COMPTES RENDUS de l'Académie des sciences

1873-7234 (electronic)





Volume 348, Special Issue 10-11, octobre-novembre 2020 **Special issue / Numéro thématique** Contributions in mechanics of materials / Contributions en mécanique des matériaux

Guest editors / *Rédacteurs en chef invités* Julie Diani, Olivier Castelnau, Francisco Chinesta

Académie des sciences — Paris



INSTITUT DE FRANCE Académie des sciences



Comptes Rendus

Mécanique

Objective of the journal

Comptes Rendus Mécanique is an internationally peer-reviewed electronic journal covering all areas of the discipline. It publishes original research articles, review articles, historical perspectives, pedagogical texts, and conference proceedings of unlimited length, in English or French. *Comptes Rendus Mécanique* is published according to a virtuous policy of diamond open access, free of charge for authors (no publication fees) as well as for readers (immediate and permanent open access).

Editorial director: Étienne Ghys

Editor-in-Chief: Jean-Baptiste Leblond

Editorial board: Olga Budenkova, Francisco Chinesta, Jean-Michel Coron, Luc Dormieux, Florian Gosselin, Nicolas Moës, Léo Morin, Guillaume Ribert, Géry de Saxcé, Emmanuel Villermaux **Editorial secretary :** Adenise Lopes

About the journal

All journal's information, including the text of published articles, which is fully open access, is available from the journal website at https://comptes-rendus.academie-sciences.fr/mecanique/.

Author enquiries

For enquiries relating to the submission of articles, please visit this journal's homepage at https://comptes-rendus.academie-sciences.fr/mecanique/.

Contact

Académie des sciences 23, quai de Conti, 75006 Paris, France Tel: (+33) (0)1 44 41 43 72 CR-Mecanique@academie-sciences.fr



The articles in this journal are published under the license Creative Commons Attribution 4.0 International (CC-BY 4.0) https://creativecommons.org/licenses/by/4.0/deed.en



Contents / Sommaire

J. Diani, O. Castelnau, F. Chinesta	701 702
	/01-/03
Emmanuel Richaud Physico-chemical approach of polymer chemical ageing: a short review	785-795
Julie Diani Free vibrations of linear viscoelastic polymer cantilever beams	797-806
Yann Charles, Chunping Zhang, Monique Gaspérini, Brigitte Bacroix Identification methodology of a rate-sensitive constitutive law with mean field and full field modeling approaches for polycrystalline materials	807-826
O. Castelnau, K. Derrien, S. Ritterbex, P. Carrez, P. Cordier, H. Moulinec Multiscale modeling of the effective viscoplastic behavior of Mg ₂ SiO ₄ wadsleyite: bridg- ing atomic and polycrystal scales	827-846
Lu Tuan Le, Kais Ammar, Samuel Forest Efficient simulation of single and poly-crystal plasticity based on the pencil glide mech- anism	847-876
Carlos N. Tomé, Ricardo A. Lebensohn Polycrystal thermo-elasticity revisited: theory and applications	877-891
Joshua Furer, Martin Idiart, Pedro Ponte Castañeda Exact results for weakly nonlinear composites and implications for homogenization methods	893-909
David Ryckelynck, Thibault Goessel, Franck Nguyen Mechanical dissimilarity of defects in welded joints via Grassmann manifold and ma- chine learning	911-935
Ruben Ibanez, Pierre Gilormini, Elias Cueto, Francisco Chinesta Numerical experiments on unsupervised manifold learning applied to mechanical modeling of materials and structures	937-958



Contributions in mechanics of materials

Foreword

J. Diani^{*, a}, O. Castelnau^b et F. Chinesta^b

^{*a*} Laboratoire de Mécanique des Solides, UMR CNRS, Ecole Polytechnique, Institut Polytechnique de Paris, Route de Saclay, 91128 Palaiseau, France

^b Laboratoire des Procédés et Ingénierie en Mécanique et Matériaux, Arts et Métiers Sciences et Technologies, CNRS, CNAM, HESAM, 151 bd de l'hôpital, 75013 Paris, France

Courriels: julie.diani@polytechnique.edu (J. Diani), olivier.castelnau@ensam.eu (O. Castelnau), francisco.chinesta@ensam.eu (F. Chinesta)

Manuscrit reçu et accepté le 24 novembre 2020.

La mécanique des matériaux, qu'il s'agisse d'alliages métalliques, de polymères, de composites, ou encore de minéraux, est un domaine de recherche vaste s'appuyant aussi bien sur la physique, la chimie, les mathématiques, les techniques numériques, que les sciences expérimentales. Elle a la particularité de traverser les échelles de l'atome au milieu continu macroscopique. En particulier, la prise en compte des mécanismes élémentaires de déformation aux échelles pertinentes permet la construction de modèles de comportement robustes, i.e. qui soient capables non seulement de reproduire fidèlement les observations mais aussi de prédire le comportement mécanique dans des conditions inexplorées ou inexplorables expérimentalement. Ainsi, grâce à la mise en œuvre de techniques de transition d'échelle, les effets de la microstructure du matériau et de son évolution au cours du chargement sont pris en compte naturellement dans la construction du modèle. On définit ainsi des lois de comportement tridimensionnelles, physiquement fondées, prenant éventuellement en compte les effets de couplages thermique, magnétique, électrique ..., utiles à la mise en œuvre ou au dimensionnement de pièces, d'ouvrages ou de structures ...

Ce numéro spécial des comptes rendus de mécanique est l'occasion d'honorer le travail et la carrière de notre collègue et ami Pierre Gilormini qui a contribué de façon tout à fait essentielle à ce domaine de recherche. Durant sa carrière, Pierre a su naviguer entre les échelles et les matériaux. Doté d'une très grande culture scientifique associée à une expertise pointue, qui a largement profité à nombreux étudiants et collègues, il a su développer des collaborations fructueuses tout en gardant l'indépendance d'esprit qui lui est chère.

Pierre Gilormini est né en 1953, à Bastia, en Haute-Corse. En 1973, il intègre l'Ecole Centrale des Arts et Manufactures, et après un Diplôme d'Etudes Approfondies sur les propriétés mécaniques des matériaux, obtenu à l'Ecole des Mines de Paris, il devient ingénieur de recherche au

^{*} Auteur correspondant.

CEMEF (Sophia Antipolis). Il y réalise une thèse de docteur ingénieur sur la formation du copeau, à l'époque où l'optimisation des méthodes cinématiques nécessite une bonne intuition et un sens physique certain pour proposer des champs de vitesses astucieusement choisis. La simulation de procédés de formage initiée pendant cette thèse l'amènera à s'intéresser à la fois aux comportements en grandes transformations (avec P. Rougée), qui y jouent un rôle central, à l'approche du comportement d'agrégats polycristallins par homogénéisation (avec F. Montheillet, en particulier) qui en constitue un débouché naturel et aux méthodes numériques par éléments finis.

En 1986, après l'obtention d'une thèse d'état, P. Gilormini passe une année de chercheur invité à l'université de Mc Gill à Montréal, où il travaille sur la prédiction du développement de textures cristallines sous chargement avec J. Jonas. C'est la rencontre avec un chercheur d'une immense culture à la fois théorique et expérimentale qui marque le jeune chercheur. Ses travaux de simulations des procédés continueront au LMT (ENS de Cachan) qu'il intègre en 1987, avec la simulation du profilage (G. Nefussi). Ils culmineront par un travail avec B. Bacroix, mettant en jeu l'ensemble de ses compétences grâce à une approche multi-échelle utilisant un critère de plasticité, ajusté sur des calculs de plasticité cristalline, et implémenté numériquement. La simulation par éléments finis de l'emboutissage d'un cylindre prédit des ondulations dans la pièce finale imputées à l'anisotropie plastique du matériau.

Intéressé par les grandes transformations, il rejoint le G.R.E.C.O. Grandes Déformations et Endommagement créé par J. Lemaitre. Il y rencontre notamment A. Zaoui, T. Bretheau, M. Berveiller, P. Suquet. Prévoir l'endommagement lors des déformations plastiques notamment du fait des inclusions, l'amène à considérer une inclusion, puis plusieurs, en route vers l'homogénéisation.

L'étude du comportement mécanique des matériaux hétérogènes par homogénéisation est l'activité qui lui vaudra le plus de notoriété. Intéressé par la plasticité et la mise en forme des métaux, c'est naturellement que Pierre se forme à l'homogénéisation en lisant l'œuvre complète de celui qui sera l'un de ses modèles, R. Hill. La pléthore des modèles justifiés uniquement par des comparaisons avec l'expérience ne le satisfait pas. Il est alors séduit par la borne supérieure obtenue par une formulation variationnelle et proposée par un jeune chercheur (P. Ponte Castañeda). Il utilise cette borne pour montrer à l'ensemble de la communauté internationale le faible nombre de modèles qui ne la violent pas et, conscient de la grande expertise de l'école d'homogénéisation française, il organise avec M. Bornert et T. Bretheau la première école thématique du CNRS sur l'homogénéisation (2 semaines) à La Londe-les-Maures. Deux ouvrages intitulés *Homogénéisation en mécanique des matériaux*, faisant référence dans le domaine, en sont le fruit.

En 2004, il suit T. Bretheau dans une nouvelle aventure : la création d'un unique laboratoire de mécanique à l'Ecole Nationale Supérieure des Arts et métiers (Paris). C'est là qu'il découvre les réseaux polymères qui lui offriront des possibilités de recherche extrêmement variées, de la modélisation du comportement hyperélastique, viscoélastique en grandes transformations, par des lois phénoménologiques, à l'homogénéisation lorsque des charges sont ajoutées à la matrice polymère (G. Régnier, J. Diani), à la nano-impression de films polymère par lithographie (H. Teyssedre) et jusqu'à la dégradation des réseaux polymères par hydrolyse (E. Richaud, J. Verdu). A cette occasion, Pierre découvre un autre maître dans les écrits de PJ. Flory. On peut retenir deux contributions tout à fait originales : l'utilisation des théories probabilistes généralement appliquées à la gélification et étendues avec succès à la dégradation des polymères par coupures de chaînes, un dégel en quelque sorte (E. Richaud, J. Verdu) et la démonstration théorique et expérimentale que la propriété de mémoire de forme des réseaux polymères amorphes n'est que l'expression de deux de leurs propriétés intrinsèques, viscoélasticité et équivalence temps-température (J. Diani).

Pendant cette longue carrière, Pierre Gilormini a toujours préféré « savoir faire » plutôt que « faire faire ». Il a ainsi guidé de jeunes chercheurs par l'exemple : exemple de son travail, de sa rigueur, de sa curiosité, de son expertise et de son indépendance. Cette indépendance lui a permis de travailler avec un grand nombre de collaborateurs qu'il a toujours choisis pour la richesse des échanges qu'il pouvait y trouver. Il semble complètement naturel que ce soit par l'intermédiaire d'un numéro spécial des Comptes Rendus de Mécanique de l'Académie des Sciences que nous puissions rendre hommage à ce chercheur à qui la diffusion de la connaissance tient tant à cœur et dont cette revue détient l'une de ses très importantes contributions [P. Gilormini, 1995. Insuffisance de l'extension classique du modèle auto-cohérent au comportement non-linéaire. Comptes-Rendus de l'académie des sciences, série II, 320, 115-122].

> J. Diani Palaiseau julie.diani@polytechnique.edu

O. Castelnau Paris olivier.castelnau@ensam.eu

F. Chinesta Paris francisco.chinesta@ensam.eu



Contributions in mechanics of materials

Physico-chemical approach of polymer chemical ageing: a short review

Emmanuel Richaud^a

^{*a*} Laboratoire PIMM, Arts et Metiers Institute of Technology, CNRS, Cnam, HESAM Universite, 151 boulevard de l'Hopital, 75013 Paris, France *E-mail:* emmanuel.richaud@ensam.eu

Abstract. Polymers are known to degrade in their use conditions, with the risk that their properties reach unacceptable level. This paper reviews the modeling of modifications occurring at molecular and architectural changes occurring during their chemical ageing, with the aim of predicting the physical (mechanical) properties, all values being linked by structure properties relationships.

Keywords. Polymers, Ageing, Modeling, Oxidation, Hydrolysis, Kinetics.

Manuscript received 13th October 2020, accepted 19th November 2020.

1. Introduction

Polymers and composites become increasingly used because weight and costs saving but also improved specific mechanical properties such as ratio of stiffness or failure stress over density or fatigue resistance. For many industrial applications, long lasting properties are needed which arises the issue of interaction between polymer and its environment. One can usually distinguish

- physical ageing where the polymer skeleton remains unchanged but the distance between polymer chains is modified for example in case of water diffusion in an epoxydiamine thermoset networks [1, 2], or physical ageing by structural relaxation for glassy polymers aged under their glass transition temperature [3]. Diffusion of penetrants generally induces a decrease in glass transition and stiffness [1, 4, 5], contrarily to structural relaxation inducing an increase in yield stress for example [6, 7]. Physical ageing is in principle reversible [8]. In the case of water ageing, the amount of desorbed water after complete drying must correspond to amount of absorbed one during immersion (apart if penetrant reacts which is a case of chemical ageing described here below [9]).
- chemical aging where polymer slowly reacts with chemicals (water, oxygen...) under the action of UV light, temperature, or gamma rays [10–12]. Chemical changes are usually irreversible.

$$\begin{array}{c} 0 \\ 0 \\ R_1 \end{array} \xrightarrow{C} 0 \end{array} \xrightarrow{R_2} \begin{array}{c} H_2 0 \\ R_1 \end{array} \xrightarrow{O} 0 \\ R_1 \end{array} \xrightarrow{O} 0 \\ R_1 \end{array} + R_2 - OH$$

Scheme 1. Hydrolysis of an ester group.

In many cases, designers try to address the lifetime prediction issues by using very simple laws (one the best examples is the use of Arrhenius law for lifetime value [13]) but those later however fail because of the complexity of physico-chemical phenomena [14]. That is the reason why non empirical approach is needed and involves a multiscale modeling of degradation phenomena [15], the aim of which is:

- to predict the rate of degradation at molecular scale giving the rate of structural changes
- to use structure properties relationships for predicting the consequences of any given change at molecular scale.

The combination of both approaches allows predicting the time at which engineering properties are out of the range of required properties, in other words the lifetime of polymer.

2. Ageing study at molecular scale

Water permeation can induce a decrease in thermomechanical properties (glass transition, elastic modulus...) due to plasticization. However, the main damage induced by water is observed in case where water slowly reacts with specific groups in polymer (esters, amides...) generating chain scissions (Scheme 1) as observed for example in polyesters (PLA, PET) [16] or polyamides [17, 18].

For this last mechanism, a simple equation can be implemented for describing the changes of ester groups (in concentration [*e*]) belonging to polymer chain and the appearance of chain scissions:

$$\frac{\mathbf{d}[e]}{\mathbf{d}t} = -k_u[w][e]. \tag{1}$$

This means that there case where water is poorly soluble in polymer but reacts by hydrolysis (as for example PBT where the maximal water uptake is about 1%) but the damage is higher than in some case where water is more soluble but does not react with polymer (as for example the DGEBA-DDS epoxy thermoset).

The general mechanism for hydrolysis is the following but several subcases can be distinguished

- reversible hydrolysis, for example in PA11 [17]
- water diffusion controlled hydrolysis [19]
- auto-accelerated hydrolysis, either because hydrolysis induced groups induce an increase in hydrophily [18,20]

$$[w] = [w]_0 + a.s.$$
(2)

Or because carboxylic acids usually catalyze further hydrolysis reactions. In this case, a secondary equation must be added [21]:

$$e + w + ac \rightarrow s + ac$$
.

The rate expression becomes:

$$\frac{\mathrm{d}[e]}{\mathrm{d}t} = -k_u[w][e] - k_c[w][e][ac] \tag{3}$$



Scheme 2. Radical oxidation of polyethylene.

i.e., since each ester is transformed into an acid by hydrolysis:

$$\frac{d[e]}{dt} = -k_u[w][e] - k_c[w][e]([e]_0 - [e])$$
(4)

and the following expression of the ester concentration is obtained:

$$[e] = [e]_0 \frac{(1+A)\exp{-Kt}}{1+A\exp{-Kt}} \quad \text{with } K = k_c [e]_f [w] \quad \text{and:} \quad A = \frac{k_c [e]_0}{k_u}.$$
 (5)

In the case of thermal oxidation, there are other mechanisms responsible for chemical changes, as illustrated in Scheme 2 for the radical oxidation of polyethylene [22].

Basing on the Autooxidation Scheme established by Bolland and Gee [23], the study of the oxidation of aliphatic substrates lead to the following mechanistic scheme [24, 25]:

$$\begin{split} \delta POOH &\to \alpha P^\circ + \beta POO^\circ + \gamma_1 P = O + \gamma_2 s \quad k_1 \\ P^\circ + O_2 &\to POO^\circ \qquad k_2 \\ POO^\circ + PH &\to POOH + P^\circ \qquad k_3 \\ POO^\circ + POO^\circ &\to \text{inactive products} \qquad k_6. \end{split}$$

In the frame of radical kinetics, this mechanistic scheme leads to a differential system. This latter can be analytically solved in some simple case (using some simplifying hypothesis) or numerically solved. The outputs of the model are the concentration in reactive species ([P°], [POO°], [POO4]) and of stable products (carbonyls, chain scissions...):

$$\frac{d[P=O]}{dt} = \gamma_1 k_1 [POOH]^{\delta}$$
(6)

$$\frac{\mathrm{d}s}{\mathrm{d}t} = \gamma_2 k_1 [\text{POOH}]^\delta. \tag{7}$$

In most cases, the derived kinetic models are more complex (in order to take into account the occurrence of other termination processes [26], reaction of double bonds [27, 28], presence of stabilizers [29], reaction-diffusion coupling of oxygen [30]...). The finality remains the same: predict the concentration in trackers of chemical ageing (and chemical changes) versus time. Rate constants are usually determined in accelerated conditions (i.e. at enhanced temperatures) and later extrapolated to temperatures corresponding to use conditions using the well-known Arrhenius equation:

$$k_i(T) = k_{i0} \cdot \exp\left(-\frac{E_i}{RT}\right).$$
(8)

The validation of such kinetic models by comparing outputs with for example experimental observations by FTIR remains quite tricky for example:

• in a case of hydrolysis, esters are consumed to give carboxylic acids but the signal of both group overlap.

• in the case of radical oxidation of hydrocarbon polymers, a broad band appears with several maximal or shoulders (for example 1713 cm⁻¹ for carboxylic acids, 1720 cm⁻¹ for ketones, 1735 cm⁻¹ for esters, 1780 cm⁻¹ for lactones in polyethylene [31]). At first, it is not trivial to find out the mechanism of formation for some of those species (for example lactones, or esters). Moreover, some species are accompanied by a chain scission (carboxylic acid) and some other not (ketones) so that the assessment of the macromolecular damage from FTIR spectra requires to perform a reliable deconvolution and the knowledge of the molar absorptivity of each species.

In other words, approach can be flawed due to the complexity of the polymer. It is thus needed to study the effect of degradation on polymer architecture so as to predict its effect on mechanical properties.

3. Study of degradation at macromolecular scale

In a second step, the damage of polymer must be studied at macromolecular scale in order to implement structure properties involved in polymer ageing. For that purpose, experimental approaches are needed to assess the concentration in structural changes, i.e. chain scissions and/or crosslinks.

In thermoplastic polymers, the predominance of chain scissions or crosslink can be quite easily estimated from the changes in molar mass. Those latter can be measured:

• from rheometric measurements at molten state, where the Newtonian viscosity is linked to molar mass by the formula [32]:

$$\eta = k \cdot M_w^{3.4}.\tag{9}$$

There are several papers illustrating the predominance of chain scissions for example in linear polymers such as polyolefins [33], and the predominance of crosslinking in unsaturated (uncured) elastomers such as Ethylene Propylene Diene Monomer (in link with the presence of carbon carbon double bonds) [34].

• from liquid chromatography, this latter allowing to estimate both number and weight average molar mass. Using some statistical analysis proposed by Saito, the concentration in chain scission and in crosslink can be simultaneously estimated by [35]:

$$\frac{1}{M_N} - \frac{1}{M_{N0}} = s - x \tag{10}$$

$$\frac{1}{M_W} - \frac{1}{M_{W0}} = \frac{s}{2} - 2x \tag{11}$$

where *s* and *x* are respectively the concentration in chain scissions and in crosslinking.

Elastomers and thermoset networks are in essence insoluble and cannot be molten, which prevents the use of the here above experimental techniques. The "key" parameter is the concentration elastically active chains (linked to 2 crosslink nodes), which must increase in case of predominant crosslinking. Experimentally, the concentration in chain scission can be assessed

• from the residual soluble fraction (sol) using the Charlesby approach [36, 37]:

$$\frac{\nu}{\nu_0} = \left(\frac{1 - \mathrm{sol}^{1/2}}{1 - \mathrm{sol}_0^{1/2}}\right)^2 \tag{12}$$

• or the glass transition value (T_g) [38]:

$$T_g = \frac{T_{gl}}{1 - K_{DM} \cdot F \cdot x}.$$
(13)



Figure 1. Schematization of an undegraded ideal networks (all chains are elastically active and denoted by A) and of an degraded networks. NB: arrows indicate that chain are linked to other chains belong to network whereas open circle correspond to the end of a dangling chain.

The concentration in chain scissions can thus be estimated from the decrease in elastically active chains. In a simplified approach, on can assume that for a tridimensional network [39]:

 $n = n_0 - 3s + 2x$ (if chain scission and crosslinking coexist) (14)

or
$$n = n_0 - 3s$$
 (if they are only chain scissions). (15)

In this last case, some simplified relationships can be derived [36]

$$s = \frac{v_0}{3} \left(1 - \left(\frac{1 - \operatorname{sol}^{1/2}}{1 - \operatorname{sol}_0^{1/2}} \right)^2 \right)$$
(16)

$$s = \frac{T_{gl}}{2K_{DM}F} \cdot \left(\frac{1}{T_g} - \frac{1}{T_{g0}}\right).$$
 (17)

However, Equations (14)–(15) are a simple approximation which is valid only at low conversion degree. For summarizing, it is clear that one chain scission induces the loss of three elastically active chains (denoted by A in Figure 1) in an unaged network. Reversely, at higher conversion degree of the chain scission process, networks looks like depicted in Figure 1b. One can thus imagine that a chain scission can occur on a chain denoted by B, C or E dangling chain with a limited consequence on elastic properties, or on a dangling chain (denoted by D) which is linked by one single node to the rest of network, and becomes soluble.

A most refined approach was proposed by Gilormini *et al.* [21]. Network is here composed of a (elastically active chains connected to two nodes connected themselves with elastically active chains), b (elastically active chains connected to one node connected with elastically active chains and another node connected with dangling chains).

In this approach, the concentration of reactive groups hold by "a chains" and "b chains" changes with the concentration of chain scissions occurring on each kind of chain (s_A , s_B) as follows:

$$\frac{\mathrm{d}[a]}{\mathrm{d}t} = -5L\frac{\mathrm{d}s_A}{\mathrm{d}t} - 4L\frac{\mathrm{d}s_B}{\mathrm{d}t} \tag{18}$$

$$\frac{\mathrm{d}[b]}{\mathrm{d}t} = 4L\frac{\mathrm{d}s_A}{\mathrm{d}t} + 2L\frac{\mathrm{d}s_B}{\mathrm{d}t}.$$
(19)

The concentration in chain scissions can be predicted as follows

$$s = s_A + s_B \tag{20}$$

$$\frac{\mathrm{d}s_A}{\mathrm{d}t} = \frac{|a|}{[e]}\frac{\mathrm{d}s}{\mathrm{d}t} \quad \text{and} \quad \frac{\mathrm{d}s_B}{\mathrm{d}t} = \frac{|b|}{[e]}\frac{\mathrm{d}s}{\mathrm{d}t} \tag{21}$$

and

$$\frac{1}{[e]}\frac{\mathrm{d}s}{\mathrm{d}t} = \frac{K}{1 + A\exp(-Kt)}.$$
(22)

The solution of the differential system gives:

$$\frac{[a]}{[e]_0} = \left[\cos\left(\frac{\sqrt{15} \cdot KLu}{2}\right) - \frac{7}{\sqrt{15}}\sin\left(\frac{\sqrt{15} \cdot KLu}{2}\right)\right] \cdot \exp\left(-\frac{3}{2} \cdot KLu\right)$$
(23)

$$\frac{[b]}{[e]_0} = \frac{8}{\sqrt{15}} \sin\left(\frac{\sqrt{15} \cdot KLu}{2}\right) \cdot \exp\left(-\frac{3}{2} \cdot KLu\right)$$
(24)

with

$$u = -\frac{1}{K}\ln(1-x).$$
 (25)

Those equation lead to a fair prediction of decrease in elastically active chain concentrations which is sufficient for several industrial applications (Figure 2).

It remains interesting to focus on the prediction of degel, i.e. the critical stage at which chain scissions have cleared out all the elastically active chains and the network reverts back to a soluble material (with the possibility to regenerate chemicals and repolymerize them later). An original approach for predicting the degelation was developed by Gilormini *et al.* [40]: in the simplest version of this approach, a chain contains *L* reactive groups likely to undergo a chain scission process.

The probability of any chain to be uncut is given by:

$$u(x) = (1 - x)^{L}.$$
(26)

Let us focus now on the possibility for a node to belong to the gel or to belong to the soluble fraction of network (i.e. to be a free chain). The Figure 3 summarizes three possibilities:

• either the node is attached to 2 soluble chains, or at least 1 or 2 chains are attached to the network (with a probability g(x)). It gives the probability of the node to belong to the soluble fraction:

$$P(x) = [1 - g(x)]^2.$$
(27)

So that the probability for node#2 to belong to the gel:

$$P(x) = 1 - [1 - g(x)]^2 \quad \text{if } f = 3 \tag{28}$$

or:

$$P(x) = 1 - [1 - g(x)]^{f-1} \quad \text{for any } f.$$
⁽²⁹⁾

It gives the probability for a chain to be uncut and belong to the gel:

$$g(x) = u(x) \cdot [1 - (1 - g(x))^{f-1}]$$
(30)

so that, at degel:

$$x_d = 1 - \frac{1}{(f-1)^{1/L}}.$$
(31)

This approach was for example successfully validated in the case of methyl methacrylate– ethylene glycol di methacrylate radiolysis [41]. It is clear that it will be helpful in a close future given the increasing need to recycle by mechanical or chemical processes thermosets and rubbery networks.



Figure 2. Decrease in elastically active chains concentration of a polyester urethane rubbery network for several ageing conditions.

4. Structure-properties involved in the prediction of ageing induced failure

According to previous parts, it seems possible to predict the appearance of chain scissions or crosslink nodes in either thermoplastics or networks, and subsequently the changes in average molar mass (for thermoplastics) or crosslink density (for networks). It remains to establish the effect of such changes on the mechanical properties of polymers.

In the field of thermoplastic polymers undergoing mainly chain scissions, the main effect is the plasticity loss. A first end of life criteria was established basing on the observation that plastic



Figure 3. Schematization of network degradation.



Figure 4. Schematization of embrittlement criterion for thermoplastic polymers.

deformation was possible only when residual average molar mass was higher than a "critical value" (denoted by M'_c) as depicted in Figure 4a. This latter was carefully discussed [42] in the past and it was observed that:

- in amorphous thermoplastics (PMMA, PC, PS...), its value was slightly higher than the molar mass between entanglements, indicating that failure occurs when the polymer is severely damaged.
- in semi-crystalline polymers (in particular with amorphous phase in rubbery state), the ratio of "critical" molar mass to the molar mass between entanglement clearly exceeds 1, meaning that only a few chain scissions strongly reduce the plasticity. This pointed out the "morphological" origin of embrittlement. The following scenario was evidenced: chain scissions liberate "small" segments in amorphous phase and those latter join the crystalline phase which is, in essence, less deformable. This led to a "mixed" end of life criterion expressed in an "average molar mass–crystalline ratio" window (Figure 4b): polymers made with longer chains and poorly crystalline are ductile whereas polymers made of short chains with a high crystalline ratio are brittle.

In rubbery networks, one of the key properties is the changes in elastic modulus. Based on the fact that elasticity has an entropic origin in rubbery networks, it is established for years that elastic modulus depends on the concentration in elastically active chains i.e. [43]:

$$G = n_A \cdot RT \tag{32}$$

with

$$E = 3 \cdot G. \tag{33}$$

The failure of networks can be described by the critical strain energy release rate in mode I (G_{IC}). This later is shown to vary as follows in network:

- at first, *G*_{IC} can increase with the crosslinking concentration because crosslinking positively influences the elastic modulus,
- in the same time, crosslinking decreases the maximal chain extensibility.

It results in curves displaying a maximum as depicted in [44].

In thermosets, there are also data highlighting the relationship between crosslink density and toughness [45]. However, it was recently proposed that for networks with a sub-glassy relaxation such as epoxies, the end of life is governed by the depletion of groups responsible for this low temperature mobility [46].

5. Conclusions

This short paper summarizes the mathematical description of multiscale changes induced by the "chemical" ageing, i.e. the effect of the reactions of chemicals (oxygen, water) with polymer. Kinetic approaches can allow a fine prediction of the rate of chemical changes at "molecular scale". The use of statistical theories allows thus to address the changes at macromolecular scale, even ut to advanced degradation levels (such as the degelation). Last, the use of well-known structure properties relationships established for virgin polymers, together with the prediction of changes in macromolecular architecture with chains scissions or crosslinking allow predicting the changes in some physical properties and later offer to engineers a complete physico-chemical toolbox for addressing polymer lifetime issues by a non-empirical approach.

Acknowledgements

The author wants to express his sincere gratitude to Dr. Pierre Gilormini for his collaboration on degradation in polymers networks (Refs [21, 40, 41]). Besides his innovative approaches, Pierre is always a very accessible colleague, and is an example of rigor and curiosity who has inspired many young researchers in PIMM.

References

- I. Merdas, F. Thominette, A. Tcharkhtchi, J. Verdu, "Factors governing water absorption by composite matrices", *Comput. Sci. Tech.* 62 (2002), p. 487-492.
- [2] P. Gilormini, J. Verdu, "On the role of hydrogen bonding on water absorption in polymers", *Polymer* **42** (2018), p. 164-169.
- [3] G. M. Odegard, A. Bandyopadhyay, "Physical aging of epoxy polymers and their composites", J. Polym. Sci. B 49 (2011), p. 1695-1716.
- [4] B. De'Nève, M. E. R. Shanahan, "Water absorption by an epoxy resin and its effect on the mechanical properties and infra-red spectra", *Polymer* 34 (1993), p. 5099-5105.
- [5] B. de Nève, M. E. R. Shanahan, "Effects of humidity on an epoxy adhesive", Int. J. Adhes. Adhes. 12 (1992), p. 191-196.
- [6] T. D. Chang, J. O. Brittain, "Studies of epoxy resin systems. Part C: Effect of sub-Tg aging on the physical properties of a fully cured epoxy resin", *Polym. Eng. Sci.* 22 (1982), p. 1221-1227.
- [7] T. D. Chang, J. O. Brittain, "Studies of epoxy resin systems. Part D: Fracture toughness of an epoxy resin: a study of the effect of crosslinking and sub-Tg aging", *Polym. Eng. Sci.* 22 (1982), p. 1228-1236.
- [8] J. M. Hutchinson, "Physical aging of polymers", Prog. Polym. Sci. 20 (1995), p. 703-760.
- [9] J. El Yagoubi, G. Lubineau, F. Roger, J. Verdu, "A fully coupled diffusion-reaction scheme for moisture sorptiondesorption in an anhydride-cured epoxy resin", *Polymer* 53 (2012), p. 5582-5595.
- [10] J. E. Pickett, "Introduction to polymer weathering, stabilization, and testing", in Service Life Prediction of Polymers and Coatings (C. C. White, M. E. Nichols, J. E. Pickett, eds.), Elsevier, Oxford, 2020, p. 1-18.
- [11] D. J. Carlsson, "Degradation and stabilization of polymers subjected to high energy radiation", in *Atmospheric Oxidation and Antioxidants* (G. Scott, ed.), Elsevier, Amsterdam, 1993, p. 495-530.

- [12] J. E. Pickett, D. J. Coyle, "Hydrolysis kinetics of condensation polymers under humidity aging conditions", *Polym. Degrad. Stab.* 98 (2013), p. 1311-1320.
- [13] F. Gugumus, "Effect of temperature on the lifetime of stabilized and unstabilized PP films", *Polym. Degrad. Stab.* **63** (1999), p. 41-52.
- [14] M. Celina, K. T. Gillen, R. A. Assink, "Accelerated aging and lifetime prediction: review of non-Arrhenius behaviour due to two competing processes", *Polym. Degrad. Stab.* 90 (2005), p. 395-404.
- [15] M. C. Celina, "Review of polymer oxidation and its relationship with materials performance and lifetime prediction", *Polym. Degrad. Stab.* 98 (2013), p. 2419-2429.
- [16] J. E. Pickett, D. J. Coyle, "Hydrolysis kinetics of condensation polymers under humidity aging conditions", *Polym. Degrad. Stab.* 98 (2013), p. 1311-1320.
- [17] B. Jacques, M. Werth, I. Merdas, F. Thominette, J. Verdu, "Hydrolytic ageing of polyamide 11. 1. Hydrolysis kinetics in water", *Polymer* 43 (2002), p. 6439-6447.
- [18] C. El-Mazry, O. Correc, X. Colin, "A new kinetic model for predicting polyamide 6-6 hydrolysis and its mechanical embrittlement", *Polym. Degrad. Stab.* 97 (2012), p. 1049-1059.
- [19] V. Bellenger, M. Ganem, B. Mortaigne, J. Verdu, "Lifetime prediction in the hydrolytic ageing of polyesters", *Polym. Degrad. Stab.* 49 (1995), p. 91-97.
- [20] A. Ballara, J. Verdu, "Physical aspects of the hydrolysis of polyethylene terephthalate", Polym. Degrad. Stab. 26 (1989), p. 361-374.
- [21] E. Richaud, P. Gilormini, M. Coquillat, J. Verdu, "Crosslink Density Changes during the Hydrolysis of Tridimensional Polyesters, Macromol", *Theor. Simul.* 23 (2014), p. 320-330.
- [22] J. Lemaire, R. Arnaud, J.-L. Gardette, "Low temperature thermo-oxidation of thermoplastics in the solid state", *Polym. Degrad. Stab.* 33 (1991), p. 277-294.
- [23] J. L. Bolland, G. Gee, "Kinetic studies in the chemistry of rubber and related materials. II. The kinetics of oxidation of unconjugated olefins", *Trans. Faraday Soc.* 42 (1946), p. 236-243.
- [24] S. Verdu, J. Verdu, "A new kinetic model for polypropylene thermal oxidation at moderate temperatures", *Macro-molecules* **30** (1997), p. 2262-2267.
- [25] L. Audouin, V. Gueguen, A. Tcharkhtchi, J. Verdu, ""Close loop" mechanistic schemes for hydrocarbon polymer oxidation", J. Polym. Sci. A 33 (1995), p. 921-927.
- [26] E. Richaud, F. Farcas, P. Bartoloméo, B. Fayolle, L. Audouin, J. Verdu, "Effect of oxygen pressure on the oxidation kinetics of unstabilised polypropylene", *Polym. Degrad. Stab.* 91 (2006), p. 398-405.
- [27] P. Y. Le Gac, G. Roux, J. Verdu, P. Davies, B. Fayolle, "Oxidation of unvulcanized, unstabilized polychloroprene: a kinetic study", *Polym. Degrad. Stab.* 109 (2014), p. 175-183.
- [28] X. Colin, L. Audouin, J. Verdu, "Kinetic modelling of the thermal oxidation of polyisoprene elastomers. Part 1: Unvulcanized unstabilized polyisoprene", *Polym. Degrad. Stab.* 92 (2007), p. 886-897.
- [29] E. Richaud, B. Fayolle, J. Verdu, "Polypropylene stabilization by hindered phenols Kinetic aspects", Polym. Degrad. Stab. 96 (2011), p. 1-11.
- [30] L. M. Rincon-Rubio, B. Fayolle, L. Audouin, J. Verdu, "A general solution of the closed-loop kinetic scheme for the thermal oxidation of polypropylene", *Polym. Degrad. Stab.* 74 (2001), p. 177-188.
- [31] M. Gardette, A. Perthue, J.-L. Gardette, T. Janecska, E. Földes, B. Pukánszky, S. Therias, "Photo- and thermal-oxidation of polyethylene: Comparison of mechanisms and influence of unsaturation content", *Polym. Degrad. Stab.* 98 (2013), p. 2383-2390.
- [32] W. W. Graessley, "Viscoelasticity and flow in polymer mets and concentrated solutions", in *Physical Properties of Polymers* (E. Mark, A. Eisenberg, W. Graessley, L. Mandelkern, J. Koenig, eds.), ACS, Washington, DC, 1984, p. 97-153.
- [33] B. Fayolle, L. Audouin, J. Verdu, "A critical molar mass separating the ductile and brittle regimes as revealed by thermal oxidation in polypropylene", *Polymer* 45 (2004), p. 4323-4330.
- [34] A. Kumar, S. Commereuc, V. Verney, "Ageing of elastomers: a molecular approach based on rheological characterization", *Polym. Degrad. Stab.* **85** (2004), p. 751-757.
- [35] O. Saito, "On the effect of high energy radiation to polymers I. Cross-linking and degradation", J. Phys. Soc. Jpn 13 (1958), no. 2, p. 198-206.
- [36] C. Galant, B. Fayolle, M. Kuntz, J. Verdu, "Thermal and radio-oxidation of epoxy coatings", *Progr. Org. Coat.* **69** (2010), p. 322-329.
- [37] A. Charlesby, S. H. Pinner, "Analysis of the solubility behaviour of irradiated polyethylene and other polymers", *Proc. R. Soc. Lond. A* **249** (1959), p. 367-386.
- [38] E. A. DiMarzio, "On the second-order transition of a rubber", J. Res. Nat. Bur. Stand. A Phys. Chem. 68 (1964), p. 611-617.
- [39] E. Ernault, E. Richaud, B. Fayolle, "Thermal-oxidation of epoxy/amine followed by glass transition temperature changes", *Polym. Degrad. Stab.* 138 (2017), p. 82-90.
- [40] P. Gilormini, E. Richaud, J. Verdu, "A statistical theory of polymer network degradation", *Polymer* 55 (2014), p. 3811-3817.

- [41] P. Gilormini, E. Richaud, J. Verdu, "Radiochemical "degelation" of polymethyl methacrylate networks", *Polymer* **111** (2017), p. 130-136.
- [42] B. Fayolle, E. Richaud, X. Colin, J. Verdu, "Review: Degradation-induced embrittlement in semi-crystalline polymers having their amorphous phase in rubbery state", *J. Mater. Sci.* **43** (2008), p. 6999-7012.
- [43] J. E. Mark, "The rubber elastic state", in *Physical Properties of Polymers* (E. Mark, A. Eisenberg, W. W. Graessley, L. Mandelkern, J. L. Koenig, eds.), ACS, Washington, DC, 1984, p. 1-54.
- [44] P. Y. Le Gac, M. Broudin, G. Roux, J. Verdu, P. Davies, B. Fayolle, "Role of strain induced crystallization and oxidative crosslinking in fracture properties of rubbers", *Polymer* **55** (2014), p. 2535-2542.
- [45] J.-P. Pascault, H. Sautereau, J. Verdu, R. J. J. Williams, *Thermosetting Polymers (Plastics Engineering Handbook)*, 1st ed., CRC Press, 2002, Ch. 12.
- [46] E. Ernault, E. Richaud, B. Fayolle, "Origin of epoxies embrittlement during oxidative ageing", *Polym. Test.* **63** (2017), p. 448-454.



Contributions in mechanics of materials

Free vibrations of linear viscoelastic polymer cantilever beams

Julie Diani^{*a*}

^{*a*} LMS, CNRS UMR 7649, Ecole Polytechnique, Route de Saclay, 91128 Palaiseau, France *E-mail:* julie.diani@polytechnique.edu

Abstract. The free vibrations of cantilever slender beams of polymers, which are viscoelastic materials, are theoretically described using the simple Euler–Bernoulli assumption. The comparison between the theory and the experimental data collected for a thermoplastic elastomer, polyether block amide, shows very satisfactory results. Consequently, the theory is used for a thoughtful analysis of the impact of the material parameters and the beam geometry on its free vibration. Finally, the comparison of the dynamic behaviors of two polymers, using the free vibration test and a simple uniaxial tension/relaxation test, is discussed.

Résumé. Les vibrations libres d'une poutre encastrée de polymère, mesurées expérimentalement sont reproduites théoriquement à l'aide de l'hypothèse des poutres d'Euler–Bernoulli, une fois le comportement viscoélastique du matériau identifié classiquement. La théorie permet alors de simplement faire varier les paramètres matériaux et géométriques de la poutre afin de tester leurs impacts sur le test de vibration libre. En utilisant l'analyse théorique et en observant la réponse de deux matériaux lors d'un essai simple de traction/relaxation, il est possible de prédire leur comportement relatif en vibration libre.

Keywords. Polymer, Viscoelasticity, Vibration, Damping, Cantilever beam.

2020 Mathematics Subject Classification. 00X99.

Funding. This work benefited from the support of the chair "Modelling advanced polymers for innovative material solutions" led by the Ecole polytechnique (I'X) and the Fondation de l'Ecole polytechnique and sponsored by Arkema.

Manuscript received 17th April 2020, revised 12th June 2020, accepted 15th June 2020.

1. Introduction

In shoe sole applications, polymers are preferred for various reasons such as comfort, lightness, resistance to wear, cushioning effect, and so on. Good shock absorption is often desired, but in some sport applications, like running, elastic energy recovery is also crucial to providing good bouncing. For this reason, sport brands together with polymer companies are looking for materials offering a perfect compromise. To compare materials, the free vibration of a slender cantilever beam was introduced by Arkema as a characterization test. To obtain a quantitative analysis of this test, the vibration of a linear viscoelastic cantilever beam was calculated using the Euler–Bernoulli beam theory. The theory was first validated on actual experimental data and



Figure 1. Free vibration test: the cantilever beam is subjected to an initial vertical displacement at x = L and then let go.

then used to understand better the impact of the material viscoelastic parameters and of the beam geometry on its free vibration. Note that most contributions focusing on the vibrations of viscoelastic cantilever beams deal with theory (see [1–4]) without any experimental validation or application perspective. However, [5] provides a comparison between experiments and the Euler–Bernoulli theory for an excited laminated beam whose behavior is defined by a simple Kelvin–Voigt model.

In Section 2, the basic equations are briefly recalled for elastic and linear viscoelastic materials. In Section 3, the theory is validated on experimental data recorded on metal and polymer beams. Then, the impacts of the material parameters and of the beam geometry are analyzed. Finally, a simple uniaxial tension/relaxation characterization is used to predict the performance of different materials during the free vibration test.

2. Theory

2.1. Elastic problem

Let us consider a homogeneous elastic beam of length *L*, uniform cross-section *A*, mass density ρ , and Young modulus *E* (Figure 1). Using the Euler–Bernoulli beam theory [6], the equation of motion of the beam is given by

$$EI\frac{\partial^4 w}{\partial x^4} + \rho A\frac{\partial^2 w}{\partial t^2} = 0, \quad 0 \le x \le L,$$
(1)

where *I* is the second moment of area of the beam cross-section. Seeking for a solution of the form $w(x, t) = \varphi(x)\psi(t)$, Equation (1) transforms into

$$-\frac{EI}{\rho A\varphi(x)}\frac{\partial^4\varphi(x)}{\partial x^4} = \frac{1}{\psi(t)}\frac{\partial^2\psi(t)}{\partial t^2} = -\omega^2$$
(2)

with ω being constant. Therefore, the problem consists in solving a system of two differential equations

$$\begin{cases} \frac{\partial^4 \varphi(x)}{\partial x^4} - \delta^4 \varphi(x) = 0, \ \delta^4 = \frac{\rho A \omega^2}{EI} \\ \frac{\partial^2 \psi(t)}{\partial t^2} + \omega^2 \psi(t) = 0 \end{cases}$$
(3)

of general solution

$$\begin{cases} \varphi(x) = \alpha_1 \sinh \delta x + \alpha_2 \cosh \delta x + \alpha_3 \sin \delta x + \alpha_4 \cos \delta x \\ \psi(t) = \alpha_5 \sin \omega t + \alpha_6 \cos \omega t \end{cases}$$
(4)

with parameters α_1 , α_2 , α_3 , and α_4 being determined by the boundary conditions and parameters α_5 and α_6 being defined by the initial conditions. For a cantilever beam clamped at one end (*x* = 0) and free at the other end (*x* = *L*), the boundary conditions can be written as

$$w(0,t) = 0, \quad \left. \frac{\partial w}{\partial x} \right|_{x=0} = 0, \quad \left. \frac{\partial^2 w}{\partial x^2} \right|_{x=L} = 0, \quad \left. \frac{\partial^3 w}{\partial x^3} \right|_{x=L} = 0.$$
(5)

Introducing these boundary conditions into (4), parameters α_i satisfy

$$\alpha_1 + \alpha_3 = 0, \quad \alpha_2 + \alpha_4 = 0 \tag{6}$$

$$\begin{pmatrix} \sinh \delta L + \sin \delta L & \cosh \delta L + \cos \delta L \\ \cosh \delta L + \cos \delta L & \sinh \delta L - \sin \delta L \end{pmatrix} \begin{pmatrix} \alpha_1 \\ \alpha_2 \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix},$$
(7)

and the non-trivial solution is written as

$$\cos\delta_n L \cosh\delta_n L = -1. \tag{8}$$

The first few roots have already been calculated: $\delta_1 L = 1.87510$, $\delta_2 L = 4.69409$, and $\delta_3 L = 7.85340$ (see for instance [4] for a report of solutions for different boundary conditions).

Let us focus on the first mode of vibration. For the initial conditions $w(L,0) = \Delta_i$ and $\partial w/\partial t|_{t=0} = 0$, the solution is written as

$$w(x,t) = \frac{\Delta_i}{\cosh\delta_1 L - \cos\delta_1 L + (\sin\delta_1 L - \sinh\delta_1 L) \frac{\cos\delta_1 L + \cosh\delta_1 L}{\sinh\delta_1 L + \sin\delta_1 L}} \times \cos\left(\sqrt{\frac{EI}{\rho A}}\delta_1^2 t\right) \left(\cosh\delta_1 x - \cos\delta_1 x + (\sin\delta_1 x - \sinh\delta_1 x) \frac{\cos\delta_1 x + \cosh\delta_1 x}{\sinh\delta_1 x + \sin\delta_1 x}\right).$$
(9)

In order to compare the solution to an actual experimental result obtained on an elastic beam, one needs to calculate the solution, taking into account air friction and grip system friction. In such a case, Equation (1) transforms into

$$EI\frac{\partial^4 w}{\partial x^4} + c_f \frac{\partial w}{\partial t} + \rho A \frac{\partial^2 w}{\partial t^2} = 0, \quad 0 \le x \le L.$$
(10)

Variable separation still applies and by writing the solution as $w(x, t) = e^{j\omega t}\varphi(x)$ (with $j^2 = -1$), one obtains

$$EI\frac{\partial^4\varphi(x)}{\partial x^4} + (c_f j\omega - \rho A\omega^2)\varphi(x) = 0$$
(11)

$$\Leftrightarrow \frac{\partial^4 \varphi(x)}{\partial x^4} - \delta^4 \varphi(x) = 0 \quad \text{with } \delta^4 = \frac{\rho A \omega^2 - c_f j \omega}{EI}.$$
 (12)

The solution of the latter equation has been defined in (3), with values of δ_n still being the roots of (8). Therefore, for each vibration mode δ_n , the value of ω_n is obtained by solving

$$\rho A\omega_n^2 - c_f j\omega_n - EI\delta_n^4 = 0. \tag{13}$$

Note that the values of ω_n are now complex. Focusing on the first mode δ_1 only and considering the same boundary and initial conditions, the free vibration of the cantilever beam is written as

$$w(x,t) = e^{-\operatorname{Im}(\omega_{1})t} \cos(\operatorname{Re}(\omega_{1})t) \\ \times \frac{\Delta_{i}}{\cosh \delta_{1}L - \cos \delta_{1}L + (\sin \delta_{1}L - \sinh \delta_{1}L) \frac{\cos \delta_{1}L + \cosh \delta_{1}L}{\sinh \delta_{1}L + \sin \delta_{1}L}} \\ \times \left(\cosh \delta_{1}x - \cos \delta_{1}x + (\sin \delta_{1}x - \sinh \delta_{1}x) \frac{\cos \delta_{1}x + \cosh \delta_{1}x}{\sinh \delta_{1}x + \sin \delta_{1}x}\right).$$
(14)

2.2. Linear viscoelastic materials

The viscoelastic behavior of a polymer at infinitesimal strain is well described by a generalized Maxwell model. The latter consists of a parallel scheme of an elastic branch characterized by Young modulus E_{∞} and viscoelastic branches defined by relaxation times and associated Young moduli (τ_i, E_i). The vibration problem (10) is now written as [7,8]

$$I\int_{0}^{t} \left(E_{\infty} + \sum_{i} E_{i} e^{-\frac{t-\tau}{\tau_{i}}} \right) \frac{\partial}{\partial \tau} \left[\frac{\partial^{4} w(x,\tau)}{\partial x^{4}} \right] d\tau + c_{f} \frac{\partial w}{\partial t} + \rho A \frac{\partial^{2} w}{\partial t^{2}} = 0, \quad 0 \le x \le L.$$
(15)

Then, introducing the solution of the form $w(x, t) = e^{j\omega t} \varphi(x)$ leads to the differential equation

$$I\left(E_{\infty} + \sum_{i} E_{i} \frac{j\omega\tau_{i}}{1 + j\omega\tau_{i}}\right) \frac{\partial^{4}\varphi(x)}{\partial x^{4}} + (c_{f}j\omega - \rho A\omega^{2})\varphi(x) = 0$$
(16)

$$\Leftrightarrow \frac{\partial^4 \varphi(x)}{\partial x^4} - \delta^4 \varphi(x) = 0, \quad \text{with } \delta^4 = \frac{\rho A \omega^2 - c_f j \omega}{I \left(E_\infty + \sum_i E_i \frac{j \omega \tau_i}{1 + i \omega \tau_i} \right)}.$$
 (17)

Therefore, the problem to solve is similar to the previous case, and solution (14) still holds with ω_1 satisfying

$$\rho A\omega_1^2 - c_f j\omega_1 - I \left(E_\infty + \sum_i E_i \frac{j\omega_1 \tau_i}{1 + j\omega_1 \tau_i} \right) \delta_1^4 = 0.$$
⁽¹⁸⁾

The model is now tested against experimental data recorded on metal and polymer beams.

3. Model validation

3.1. Free vibration of a metal elastic beam

A metal reglet of thickness T = 0.5 mm, width W = 13 mm, and length L = 65 mm was subjected to a free vibration test. After applying an initial displacement $\Delta_i = -1.5$ mm at its free end (x = L), the reglet was set free from vibrations. The vertical motion of the tip of the reglet was measured with respect to time by a laser beam. Although the Young modulus of the reglet was unknown, it was easily estimated thanks to the vibration frequency; a realistic value of E = 121 GPa was obtained. The air friction and the clamp system damping were taken into account with the parameter c_f , which is fitted on the experimental data. A very satisfactory comparison between the experiment and the theory equation (14) is displayed in Figure 2, showing the relevance of the theory including the Euler–Bernoulli assumption.

3.2. Free vibration of a polymer beam

The thermoplastic elastomer commercialized by Arkema under the reference PEBAX®4033 was used for experimental testing. The linear viscoelasticity of the polymer was characterized using dynamic mechanical analysis in torsion. Rectangular specimens were subjected to torsion frequency sweeps, from 10^{-2} to 10 Hz, at given temperatures, from -80 °C to 80 °C with 5 °C temperature increments, using an Anton Paar MCR 502 rheometer. The time–temperature superposition assumption [9] was successfully applied. The experimental master curves for the material built at 20 °C reference temperature, as well as the linear viscoelastic fit by a generalized Maxwell model using 40 viscoelastic branches, are displayed in Figure 3. The values of the relaxation spectrum are given in Appendix A. Moreover, the elastic shear modulus at high temperature, G_{∞} , was recorded at 21 MPa. Note that the values of the shear moduli (G_{∞} , G_i) were then multiplied by a correction parameter x = 0.95 depending on the specimen geometry, rationally defined in [10] to account for the grip clamping when applying the Saint-Venant assumption [11] for rectangular



Figure 2. Comparison between the Euler–Bernoulli beam theory and the experimental data for the free vibration of a metal reglet (elastic material), taking into account air friction and clamp damping through the fit coefficient c_f .



Figure 3. Linear viscoelasticity master curves for PEBAX®4033 at 20 °C and generalized Maxwell model fit.

specimens in torsion. Finally, the Young moduli E_{∞} and E_i are simply assumed as thrice the values of G_{∞} and G_i , respectively. Although this assumption is exact for E_{∞} and some values of E_i , it probably overestimates some other E_i values. Nonetheless, the comparison between the free vibration experimental results and the theory will prove that this assumption is reasonable.

A rectangular beam of thickness T = 2.1 mm, width W = 10 mm, and length L = 65 mm of PEBAX®4033 was subjected to the free vibration test. Theoretical solutions were calculated using (14) with the initial displacement and the characterized linear viscoelastic behavior of the polymer as inputs. A very satisfactory comparison between the model and the experimental data is shown in Figure 4. Note that the theoretical solution was calculated with damping parameter c_f set to zero, assessing the first order of the material viscoelasticity in the vibration damping of the polymer beam.

The simple Euler–Bernoulli theory is relevant to reproducing the experimental data obtained with the free vibration experiment for the cantilever beam. Therefore, the theory will be used to discuss the impact of the material behavior and of the beam geometry on its free vibration.



Figure 4. Comparison between the Euler–Bernoulli beam theory and the experimental data for the free vibration of a PEBAX®4033 polymer beam.

4. Analysis and discussion

4.1. Impact of material parameters

For the purpose of simplicity, the polymer beam geometry is considered as reference. To better understand the impact of material parameters on the free vibration test, a simple Zener model is considered for the viscoelastic behavior, consisting of an elastic branch characterized by stiffness E_{∞} in parallel with a viscous branch characterized by stiffness E_v and relaxation time τ_v . For practical purposes, the reference parameters are set to $E_{\infty} = 80$ MPa, $E_v = 20$ MPa, and $\tau_v = 0.002$ s. This relaxation time is chosen to obtain a realistic duration for total damping in comparison to the experimental data displayed in Figure 4. Finally, since the air friction and clamp damping are observed to be of second order for the PEBAX® elastomer, the friction parameter is set to zero ($c_f = 0$). The beam is theoretically subjected to an initial vertical displacement approximately -3 mm as in the experiment. The free vibration is plotted with respect to time according to the material parameters.

First, several values of E_{∞} and E_v are considered while keeping the relaxation time τ_v constant. Figure 5 shows a comparison of the theoretical free vibrations when keeping E_{∞} constant and increasing E_v and when decreasing E_{∞} while keeping E_v constant. As one can expect, the stiffer the viscous branch, the faster the damping. Moreover, the damping seems to be controlled by the absolute value of E_v . For instance, the relative viscosity, defined by $E_v/(E_{\infty} + E_v)$, has an insignificant impact. Finally, the stiffness of the elastic branch E_{∞} affects the vibration frequency, which decreases when E_{∞} decreases and all other parameters are kept constant.

Second, the stiffnesses E_{∞} and E_{ν} are kept constant and τ_{ν} is varied. Figure 6 shows the comparisons of the free vibrations of the cantilever beam when multiplying or dividing the relaxation time τ_{ν} by a factor of five for reference relaxation times $\tau_{ref} = 0.001$ s and $\tau_{ref} = 0.05$ s. One notes that the damping depends on the absolute value of the relaxation time and not on a relative increase or decrease in a given reference value. In fact, considering the material and beam dimensions, the best damping was obtained for $\tau_{\nu} \approx 0.006$ s. This can be observed from Figure 7 displaying the values of the imaginary part of ω as a function of τ_{ν} .

Note that materials of different relaxation times τ_v and the same viscosity $\eta_v = E_v \times \tau_v$ may show similar free vibration but not necessarily. Two limit cases are worth mentioning. When τ becomes very large, the beam behaves like an elastic beam of Young modulus $E_{\infty} + E_v$. When τ is very small, the beam behaves like an elastic beam of Young modulus E_{∞} . Finally, note that Julie Diani



Figure 5. Impact of the Zener material stiffnesses E_{∞} and E_{ν} on the free vibration of the cantilever beam.



Figure 6. Impact of the relaxation time of the Zener material on the beam vibration.



Figure 7. Estimates of Im(ω), characterizing the beam vibration damping, with respect to the relaxation time τ_v .

it could be difficult to extend the later analysis to real polymers that present a spectrum of relaxation times.

803



Figure 8. Impact of the cantilever beam geometry on its free vibration.

4.2. Impact of beam geometry

Since the behavior of different materials may be compared using this test, let us focus on the impact of the beam geometry on its free vibration. First, let us note that in (17) and (18), the solution is independent of W. Moreover, two beams presenting the same ratios L/T show the same vibration behavior. When increasing L/T, the frequency decreases and the damping is delayed. It is therefore important when comparing two materials experimentally to consider similar geometry in terms of dimension ratio L/T.

4.3. Material comparison

In Section 3.2, it was shown that it is possible to predict the free vibration of a cantilever polymer beam when the linear viscoelasticity of the material is known. Nonetheless, the characterization of the linear viscoelasticity of a polymer, as presented in Figure 3, is rather time-consuming. Therefore, this section aims at proposing a test, simpler and faster to run, to predict how different materials will perform in terms of the free vibration test. Since most labs studying materials are equipped with standard uniaxial tensile machines and since relaxation tests are relevant to characterizing the viscoelasticity of materials, only a uniaxial tension/relaxation test is performed. For this purpose, PEBAX®4033 is compared with another PEBAX®, which is labeled 70R53. Both materials are subjected to uniaxial tension up to a small deformation ϵ of approximately 1% at a constant crosshead speed of 100 mm/min. Then the stress relaxation is recorded for 30 s. The comparison of the material stiffness with respect to time, defined as $(F/S_0)/\epsilon = E(t)$ and recorded during the relaxation step, is shown in Figure 9. PEBAX®70R53 appears significantly stiffer and undergoes more stress relaxation at a faster rate. This result is in agreement with the ratios of polyamide hard segments to polyether soft segments in both materials. PEBAX®4033 contains significantly fewer polyamide hard segments than PEBAX®70R53 [12]. From this behavior characterization combined with the previous vibration analyses, one expects the vibration frequency of PEBAX®70R53 to be significantly higher than that of PEBAX®4033 due to its higher stiffness and its vibration damping to be faster due to its higher viscosity $(E_{\nu} = E(0) - E(\infty))$.

PEBAX®70R53 slender beams of the same dimension as that of PEBAX®4033 (T = 2.1 mm, W = 10 mm, and L = 65 mm) were subjected to the same vibration test protocol. The comparison of the vibrating behaviors of both PEBAX® elastomers is displayed in Figure 10. As expected, PEBAX®70R53 vibrates at a higher frequency but for a significantly shorter duration.



Figure 9. Relaxation moduli of PEBAX® elastomers 4033 and 70R53 obtained during relaxation tests at approximately 1% strain, attained with a uniaxial tensile test at a constant crosshead speed of 100 mm/min.



Figure 10. Comparison of the free vibration of PEBAX® elastomers 4033 and 70R53 for beams having the same geometries.

As a consequence, sport equipment suppliers looking to increase the damping will favor materials with high absolute viscosity.

5. Conclusion

This study aimed at providing quantitative insight into the test of free vibration of a viscoelastic cantilever beam to compare polymers for dynamic applications. Applying the simple Euler– Bernoulli beam theory for linear viscoelastic materials, a very satisfactory quantitative comparison between the theory and the experimental data was obtained for a homogeneous slender beam of PEBAX® showing a rather extended spectrum of relaxation times.

The validated theory was then used to analyze the impact of the material parameters and the beam geometry on the free vibration behavior of a homogeneous rectangular beam described by Zener viscoelastic behavior. On the one hand, the analysis showed that the vibration frequency is related to the instantaneous stiffness of the material, and the frequency increases with the material stiffness. On the other hand, the vibration damping increases with increase in the difference between the instantaneous and long-term material stiffnesses. Although significant,

the impact of the relaxation time is more difficult to analyze; no general trend has been discerned. This is therefore especially true when considering an actual polymer beam presenting a spectrum of relaxation times. However, it was shown that the free vibration of a polymer cantilever beam is directly connected to quantities that may be measured by just a tension/relaxation test. It was actually possible to predict how two materials would compare in terms of their free vibration responses by simply comparing their mechanical responses to a tension/relaxation test.

τ_i (s)	G_i (MPa)	τ_i (s)	G_i (MPa)
8.083E-25	3.522E+01	3.776E-08	1.096E+01
5.507E - 24	3.812E+01	2.573E-07	9.180E+00
3.754E-23	4.119E+01	1.754E-06	7.412E+00
2.559E-22	4.413E+01	1.195E-05	6.096E+00
1.744E-21	4.678E+01	8.144E-05	5.239E+00
1.188E-20	4.918E+01	5.553E-04	4.576E+00
8.098E-20	5.132E+01	3.783E-03	3.952E+00
5.517E-19	5.196E+01	2.578E-02	3.312E+00
3.761E-18	4.898E+01	1.757E-01	2.698E+00
2.563E-17	4.331E+01	1.198E+00	2.208E+00
1.747E - 16	3.795E+01	8.160E+00	1.848E+00
1.190E-15	3.375E+01	5.563E+01	1.596E+00
8.114E-15	3.010E+01	3.791E+02	1.434E+00
5.532E - 14	2.670E+01	2.583E+03	1.335E+00
3.769E-13	2.340E+01	1.761E+04	1.270E+00
2.568E - 12	2.075E+01	1.200E+05	1.215E+00
1.750E-11	1.885E+01	8.175E+05	1.126E+00
1.193E-10	1.700E+01	5.573E+06	1.011E+00
8.129E-10	1.475E+01	3.798E+07	9.898E-01
5.542E-09	1.271E+01	2.588E+08	1.380E+00

Appendix A.	Viscoelastic spectrum of PEBAX®4033	3 for reference temperature 20 $^\circ$	C
-------------	-------------------------------------	---	---

References

- [1] L. Struik, "Free damped vibrations of linear viscoelastic materials", Rheol. Acta 6 (1967), p. 119-129.
- [2] S. Mahmoodi, S. Khadem, M. Kokabi, "Non-linear free vibrations of Kelvin–Voigt visco-elastic beams", Int. J. Mech. Sci. 49 (2007), p. 722-732.
- [3] M. Ilyasov, "Vibrations of linear viscoelastic materials for any herditary property", Mech. Time-Depend Mater. 11 (2007), p. 249-263.
- [4] M. Avcar, "Free vibration analysis of beams considering different geometric characteristics and boundary conditions", Int. J. Mech. Appl. 4 (2014), p. 94-100.
- [5] S. Mahmoodi, N. Jalili, S. Khadem, "An experimental investigation of nonlinear vibration and frequency response analysis of cantilever viscoelastic beams", J. Sound Vib. 311 (2008), p. 1409-1419.
- [6] S. Timoshenko, History of Strength of Materals, McGraw-Hill, New York, 1953.
- [7] R. M. Christiansen, Theory of Viscoelasticity, 2 ed., Dover, New York, 2003.
- [8] Y. Lei, S. Adhikari, M. Friswell, "Dynamic characteristics of damped viscoelastic nonlocal Euler-Bernouilli beams", *Eur. J. Mech. A/Solids* 42 (2013), p. 125-136.
- [9] J. D. Ferry, Viscoelastic Properties of Polymers, 3 ed., John Wiley & Sons, New York, 1980.
- [10] J. Diani, P. Gilormini, "On necessary precautions when measuring solid polymer linear viscoelasticity with dynamic analysis in torsion", *Polym. Test.* 63 (2017), p. 275-280.
- [11] A. B. de Saint-Venant, De la torsion des prismes avec des considérations sur leur flexion ainsi que sur l'équilibre des solides élastiques en général et des formules pratiques, Mémoires des Savants Etrangers, Paris, 1855.
- [12] J. P. Sheth, J. Xu, G. L. Wilkes, "Solid state structure-property behavior of semicrystalline poly(ether-*block*-amide) PEBAX® thermoplastic elastomers", *Polymer* 44 (2003), p. 743-756.



Contributions in mechanics of materials

Identification methodology of a rate-sensitive constitutive law with mean field and full field modeling approaches for polycrystalline materials

Yann Charles^{*a*}, Chunping Zhang^{*b*}, Monique Gaspérini^{*a*} and Brigitte Bacroix^{*, c}

^{*a*} Université Sorbonne Paris Nord, Laboratoire des Sciences des Procédés et des Matériaux, LSPM, CNRS, UPR 3407, F-93430, Villetaneuse, France

^b Département de Génie Mécanique, École de Technologie Supérieure, 1100 Rue Notre-Dame Ouest, Montréal, QC H3C 1K3, Canada

^c CNRS, UPR3407, Laboratoire des Sciences des Procédés et des Matériaux, LSPM, Université Sorbonne Paris Nord, F-93430, Villetaneuse, France

E-mails: yann.charles@univ-paris13.fr (Y. Charles), chunping.zhang.1@gmail.com (C. Zhang), gasperini@univ-paris13.fr (M. Gaspérini), brigitte.bacroix@lspm.cnrs.fr (B. Bacroix)

Abstract. The present paper deals with the consideration of the rate-sensitivity mechanical behavior of metallic materials, in the framework of mean field and full field homogenization approaches. We re-examine the possibility of describing properly this rate sensitivity with a simple and widely used power law expressed at the level of the slip system, and we propose a methodology to accelerate the identification of the global material constitutive law for Finite Element (FE) simulations. For such an aim, simulations of a tensile test are conducted, using a simple homogenization model (the Taylor one, used in a relaxed constraint form) and an FE code (Abaqus), both using the same single-crystal rate-dependent constitutive law. It is shown that, provided that the identification of this law is performed with care and well adapted to the examined case (rate-sensitive or insensitive materials, static and/or dynamic ranges), the simple power law can be used to simulate the macroscopic behavior of polycrystalline aggregates in a wide range of strain rate (including both static and dynamic regimes) and strain-rate sensitivity values (up the rate-insensitive limit).

Keywords. Viscoplasticity, Polycrystalline materials, Strain-rate sensitivity, Finite Element, Constitutive law.

Manuscript received 28th July 2020, revised 8th October 2020, accepted 19th October 2020.

^{*} Corresponding author.

1. Introduction

For the modeling of the global and local mechanical behaviors of polycrystalline metals in a wide range of temperature or strain rate—with polycrystalline models, such as Taylor [1–3] or self-consistent models [4–6], with Finite Element (FE) codes [7–10], or with multiscale approaches coupling discrete dislocations dynamic approaches with FE codes [11, 12]—it is quite usual to use a rate-dependent crystalline constitutive law, which generally takes the form of the following power law expressed on the slip system s [1]¹

$$\dot{\gamma}^s = \dot{\gamma}_0^s \left(\frac{\tau^s}{\tau_0^s}\right)^n. \tag{1}$$

In this expression, n (or more often 1/n = m) characterizes the material rate sensitivity, $\dot{\gamma}^s$ is the slip rate, and τ^s the resolved shear stress. The term τ_0^s is the so-called critical (or reference) resolved shear stress (which evolves with strain when hardening is considered), while the parameter $\dot{\gamma}_0^s$ is usually called a reference shear rate [7, 8, 12]. If the exponent n is the same for all systems and for all grains of a polycrystal, it is easy to show that this exponent is also the macroscopic rate sensitivity of the polycrystal as a whole [2]. By assuming the rate dependence at the slip system level, this widely accepted phenomenological law presents the double advantage of (i) assuming that plastic strain occurs solely by crystallographic slip on well-defined slip systems, which is generally true for most metals at not too high temperatures and (ii) suppressing the long standing problem of non-uniqueness in the choice of active slip systems usually encountered in rate-independent crystalline plasticity [13–15].

Such a viscoplastic (VP) crystalline law has also been shown to be able to provide physically based large-strain-rate-sensitive constitutive models, in order to interpret experiments on metals performed up to quite large strain rates (up to 10^4 s^{-1} , see e.g. [10]), in which localization of plastic deformation is observed. Although rate sensitivity may be neglected on purpose for many metallic alloys cold-deformed at low strain rates and strains, this may no longer be valid when moderate or high strain rates are applied, or when high temperature data are considered. It is now well known that, in such a case, the resulting localization of plastic flow is indeed strongly influenced by the strain-rate sensitivity of the material [16-18], and to a lesser extent by the deformation induced anisotropy [19]. In fact, many authors have observed an increased rate sensitivity at large applied strain rates, and have postulated that this rate sensitivity increase is a characteristic of the material [1, 10, 20, 21]. However, how large the strain rate must be to observe this increased rate sensitivity is still not clear. Indeed, the strain-rate sensitivity depends in quite a complex way on strain rate, but also on strain, temperature, as well as on the underlying microstructure and possible deformation mechanisms of the tested materials [6, 22–24]. Thus, a lot of successful modeling efforts, some of them based on the very same power law as the one defined in (1), have been made to be able to simulate different strain-rate regimes, either controlled by the thermally activated interactions between dislocations at rather low strain rates or by dislocation drag at high strain rates (see, e.g., [25-28]). Conversely, some other authors claim that accounting for rate sensitivity solely by a single relationship between shear rate and shear stress is a too simplified approach even for low strain rates, and that this approach should be replaced or completed by a rate-sensitive hardening law, describing the evolution of τ_0^s with strain and strain rate (e.g., [9, 29, 30]), to account for the right influence of strain-rate sensitivity on the behavior of polycrystalline materials. Additionally, since this classical power law is still mostly used with the aim of simplifying the numerical procedures by providing a regularized

¹By using only odd values of the exponent n, we avoid in the formulation the use of absolute values. Thus, the more classically chosen value of 20 will be replaced here by 21, since the resulting difference in the macroscopic behavior is hardly detectable.

form (with a unique solution on a given slip system) of the slip criterion, the exponent *n* is usually not correctly set up (see below), resulting in an inaccurate coupling between the macroscopic rate-sensitive response and the rate-sensitive evolution of critical resolved shear stresses of slip systems. In order to solve this problem for rate-sensitive or rate-insensitive materials, some much more numerically efficient methods have been recently proposed [29,31]. The first one [29] aims at describing properly the behavior of materials in a wide range of rate sensitivity and strain rate, whereas the second one [29,31] aims at being able to treat efficiently the case of rate-insensitive materials. However, according to the authors themselves, these efficient methods are not adapted either to the treatment of rate-sensitive materials [31] or to their combined use with FE codes [32].

Thus, and especially in the framework of FE simulations, lot of works are still based on the use of the classical VP crystalline power law (1), partly also rendered popular by the user material (UMAT) subroutine developed by Huang for the commercial software Abaqus [33]. However, even in some very recent papers, the exponent of rate sensitivity is set without any justification (e.g., [12]) or is *assumed* to be "adequately chosen to neglect the rate sensitivity" (e.g., [8, 16, 34]) at the polycrystal scale. In fact, it is easy to show that it is not the case for many of the reported simulations, and that the choice made leads precisely to unexpected high strain-rate sensitivity, as soon as the strain rate varies.

The aim of this paper is thus to re-address the question, and the use, of the applied strainrate sensitivity in computations using a power-law-based crystal plasticity model, for both rateinsensitive and rate-sensitive materials. We will first recall the procedure to use adequately such a simple formulation for rate-insensitive materials. With the addition of the phenomenological saturating hardening law used in [33], we will then propose a methodology to accelerate the identification of the parameters describing the constitutive law of the material, with an FE code, for rate-sensitive materials at rather low strain rates (i.e., within the static regime). We will finally show that this identification can be extended to the case of high strain rates (dynamic regime), provided that the n value is identified differently in both regimes. This will be illustrated by some simulations of a tensile test performed using two different modeling approaches, that is, a simple homogenization model (the Taylor one, used in a relaxed constraint form) and an FE computation (using Abaqus software and dedicated user subroutines [33]), both approaches using the very same single-crystal rate-dependent constitutive law and allowing strain-rate fluctuations within the polycrystalline material.

The outline of the present paper is thus the following: the classical single-crystal VP flow rule and associated hardening law are presented in Section 2, its implementation into the Taylor model and FE Abaqus code are briefly described in Section 3, the results of the performed simulations are then analyzed in Section 4 and some conclusions are drawn in Section 5.

2. The classical single-crystal viscoplastic flow rule and associated hardening law

As already mentioned, when a grain g of a polycrystalline sample is subjected to a stress state σ , plastic strain takes place by slip on several slip systems, each labeled by index s. At the level of the slip systems, the rate-dependent slip criterion is given by (1). The resolved shear stress on system s is equal to $\tau^s = \sigma_{ij}R_{ij}^s = S_{ij}R_{ij}^s$, S being the (symmetrical) deviatoric stress tensor and R^s the orientation tensor of system s.² The components of this last tensor read

$$R_{i\,i}^{s} = \frac{1}{2} (n_{i}^{s} b_{i}^{s} + b_{i}^{s} n_{i}^{s}), \tag{2}$$

²In the following, tensors are written in bold, and the subscript "M" denotes macroscopic values.

with \vec{n}_s and \vec{b}_s characterizing respectively the slip plane normal and the slip direction for the system *s*. The strain-rate tensor $\dot{\epsilon}$ in grain *g* is then defined as a summation of individual shear-rate components on all systems *s*:

$$\dot{\varepsilon}_{ij} = \sum_{s} \dot{\gamma}_0^s \left(\frac{\tau^s}{\tau_0^s}\right)^n R_{ij}^s = \sum_{s} \dot{\gamma}_0^s \left(\frac{S_{kl}R_{kl}^s}{\tau_0^s}\right)^n R_{ij}^s.$$
(3)

As long as the parameters τ_0^s , γ_0^s , and the exponent of rate sensitivity *n* are constant, this equation represents a unique relationship between strain rate and deviatoric stress tensors at the level of grain *g*, which allows to determine both tensors from the boundary conditions imposed to the considered grain (more precisely five independent components for each tensor). The existence of a unique solution has the direct consequence that the current stress state vary with strain rate and vice versa. This constitutive law is classically accompanied by a hardening law, which can be expressed—again at the level of the slip systems—as

$$\dot{\tau}_0^s = \sum_l H_{sl} \dot{\gamma}^l,\tag{4}$$

with H_{sl} the components of the so-called hardening matrix. As in the UMAT subroutine developed for Abaqus, a saturating expression—which has proven its efficiency to reproduce experimental data concerning several metallic alloys [35, 36]—can be adopted which reads

$$\begin{cases} H_{ss} = h_0 \operatorname{sech}^2 \left| \frac{h_0 \overline{\gamma}}{\tau_{\operatorname{sat}} - \tau_{\operatorname{ini}}} \right| \\ H_{sl,s \neq l} = q h_0 \operatorname{sech}^2 \left| \frac{h_0 \overline{\gamma}}{\tau_{\operatorname{sat}} - \tau_{\operatorname{ini}}} \right| \end{cases},$$
(5)

where $\overline{\gamma}$ is the cumulated shear strain on system *s* so that

$$\overline{\gamma} = \int_0^t \sum_s |\dot{\gamma}^s| \,\mathrm{d}t. \tag{6}$$

In order to characterize completely these laws (Equations (5) and (3)), four material parameters are thus needed, namely τ_{ini} , τ_{sat} , q, and h_0 , together with two other sets of material parameters, τ_{0ini}^{s} (initial values) and $\dot{\gamma}_{0}^{S}$ —which can be reduced to two parameters τ_{0ini} and $\dot{\gamma}_{0}$, if they are assumed equal for all systems in all grains—and finally the exponent of rate sensitivity n. If one single value τ_{0ini} is imposed for all systems, then it is trivial to set it equal to τ_{ini} . In that case, the τ_{ini} and τ_{sat} values are directly linked to the initial and final values of both microscopic (at the level of the grain) and macroscopic stress values (see Figure 1 below). As for the h_0 hardening coefficient, it affects the hardening rate all along the stress–strain curve before the saturation level is reached (see again Figure 1).

Now, in order to demonstrate the usefulness of the proposed crystalline law, we are going to use two different simulations approaches (i.e., mean and full field ones) to calculate the macroscopic response of a polycrystalline aggregate in uniaxial tension. For both approaches, by construction, the local strain rate is allowed to differ from one grain to another and thus may be quite different from the macroscopic imposed one.

3. Simulation framework

3.1. The Taylor model (with full or relaxed constraints)

In the framework of the Taylor model (FC—Full Constraint or RC—Relaxed Constraint), several options are possible to simulate a tensile test along, for example, the axis 3 of the macroscopic reference system:



Figure 1. Simulated tensile curves using the RC Taylor model, with $\dot{\gamma}_0^s = \dot{\gamma}_0 = \dot{E}_{VM} = 0.0005 \text{ s}^{-1}$, q = 1.1, and n = 21. The 3 hardening parameters h_0 , τ_{ini} , τ_{sat} are varied and their values (in MPa) are given in the figure legend.

(i) The Taylor model in the FC mode, which implies that all components of the strain-rate tensor within each grain are assumed to be the same than the imposed macroscopic ones, that is, $\dot{\boldsymbol{\epsilon}} = \dot{\boldsymbol{E}}_{\mathrm{M}}$. For a general anisotropic material, the combined boundary conditions in terms of stress and strain rate then read at the level of the grain

$$\dot{\boldsymbol{\varepsilon}} = \dot{E}_{33_{\rm imp}} \begin{vmatrix} -\alpha & 0 & 0 \\ 0 & 1 - \alpha & 0 \\ 0 & 0 & 1 \end{vmatrix} \quad \text{and} \quad \boldsymbol{\sigma} = \begin{vmatrix} 0 & ? & ? \\ ? & 0 & ? \\ ? & ? & ? \end{vmatrix}$$
(7)

in which $\dot{E}_{33_{imp}}$ is the imposed macroscopic strain rate along the tensile axis and α the socalled contraction ratio, which has to be calculated, for each calculation step, by minimizing the macroscopic plastic work rate with respect to α [37]. If the material is isotropic, this factor is set to be equal to 0.5 and no extra minimization procedure is necessary in this case. This case is known to represent an upper bound for the macroscopic behavior of the material.

(ii) The Taylor model in its most relaxed version, often called the Sachs–Köchendorfer model [38, 39]. In this case, a uniaxial stress state is applied to each grain, and only the macroscopic strain rate along the tensile axis \dot{E}_{33} is imposed in each grain. The boundary conditions are then

$$\dot{\boldsymbol{\varepsilon}} = \dot{E}_{33_{\rm imp}} \begin{vmatrix} ? ? ? \\ ? ? ? \\ ? ? 1 \end{vmatrix} \quad \text{and} \quad \boldsymbol{\sigma} = \begin{vmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & ? \end{vmatrix}.$$
(8)

This option is usually applied for small strains, but it is considered to be far from the reality for moderate or large ones. It is worth mentioning that, as one of the five independent components of the strain-rate tensor is still imposed to each grain, this model does not corresponds to the so-called static model which constitutes the lower bound for the macroscopic behavior and for which the total stress tensor is the same for all grains.

(iii) an "intermediate" approach (which will be called RC Taylor model in the following), which is thought to be better for anisotropic materials although quite simplified, which consists in allowing the contraction ratio α to be different within each grain, which implies the following boundary conditions:

$$\dot{\boldsymbol{\varepsilon}} = \dot{E}_{33_{\rm imp}} \begin{vmatrix} ? & 0 & 0 \\ 0 & ? & 0 \\ 0 & 0 & 1 \end{vmatrix} \quad \text{and} \quad \boldsymbol{\sigma} = \begin{vmatrix} 0 & ? & 0 \\ ? & 0 & ? \\ ? & ? & ? \end{vmatrix}.$$
(9)

It is easy to prove that, if the sample is isotropic, we will get $\dot{E}_{M22} = \dot{E}_{M11} = 0.5$ at the level of the polycrystal and the resulting macroscopic stress state will also be purely uniaxial (like in the FC mode). Apart from being simpler and more rapid than the FC one, this version is thought to be especially well adapted to the simulation of a tensile test applied on anisotropic materials (as well as in isotropic ones of course) [40], and is consequently used in the present work.

For all these cases, it is readily seen that, among the ten independent components of $\dot{\epsilon}$ and σ which need to be determined, five are imposed and the five remaining are deduced from (3). The associated macroscopic quantities \dot{E}_M and Σ_M are then obtained by averaging on all grains.

Once the boundary conditions have been selected, an iterative calculation can be performed to simulate a tensile test, and, from the boundary conditions expressed by (9) and the resolution of (3), the tensile deviatoric stress S_{33} and strain rate $\dot{\epsilon}_{33} = \dot{E}_{33_{imp}}$ can be extracted at each step of the calculation (characterized by a time increment Δt). Then, at a given time *t* of the simulation, the total tensile strain can be obtained:

$$\Delta E_{33} = \dot{E}_{33_{\rm imp}} \Delta t \Rightarrow E_{33}(t + \Delta t) = E_{33}(t) + \dot{E}_{33_{\rm imp}} \Delta t. \tag{10}$$

At each step, the reference shear stress of each system, within each grain, is also updated

$$\tau_0^s(t+\Delta t) = \tau_0^s(t) + \sum_l H_{sl} \dot{\gamma}^l(t) \Delta t, \qquad (11)$$

noting that, at time t = 0, $\tau_0^s = \tau_{ini}$ and $\sum_l H_{sl} \dot{\gamma}^l(t) = 0$.

In order to calculate the macroscopic tensile curve, the deviatoric stress is averaged on all grains at each step to get S_M and the macroscopic true tensile stress Σ_{M33} . As

$$S_{Mii} = \langle S_{ii} \rangle$$
 and $\Sigma_{Mii} = S_{Mii} + \frac{P}{3}$, (12)

where *P* is the hydrostatic pressure, and since the macroscopic boundary conditions also impose that

$$\Sigma_{M11} = \Sigma_{M22} = 0 \tag{13}$$

then, we simply get

$$\Sigma_{M33} = \frac{3}{2} S_{M33}.$$
 (14)

Equations (10) and (14) allow then to plot the macroscopic tensile stress–strain curve. Since $\dot{E}_{33_{imp}}$ is imposed and constant along the test, the final strain can be written as

$$E_{33\text{final}} = \dot{E}_{33\text{imp}} N_{\text{step}} \Delta t, \tag{15}$$

with N_{step} being the total number of calculated steps. It is thus readily seen that if $\dot{E}_{33_{\text{imp}}}$ is modified, the product $N_{\text{step}}\Delta t$ has to be modified as well to keep the first term of (15) constant. In many of the papers quoted above, which deal with the influence of strain-rate sensitivity, the macroscopic strain rate is rarely varied and only the influence of the exponent *n* is studied. For example, it is mentioned by Canova and co-workers [1] that "all the results are normalized by the von Mises (VM) equivalent strain rate and are thus independent of the applied strain rate". In the present case, the grain VM equivalent strain rate indeed varies from grain to grain, for the selected boundary conditions (it would not be the case of course for the FC assumption).
The same expression is also valid at the macroscopic level, but if the material is macroscopically isotropic, it simply becomes equal to

$$\dot{E}_{\rm VM} = \dot{E}_{33\rm imp}.\tag{16}$$

In the simulations performed below, we will thus have $\dot{\epsilon}_{VM} \neq \dot{E}_{VM}$ in most of the grains. Also, as in many publications listed above, using either an homogenization model or an FE code, a reference case will be defined as the one performed by defining the reference shear rate equal to the macroscopic one, that is $\dot{\gamma}_0 = \dot{E}_{33imp}$, which means $\dot{E}_{33imp}/\dot{\gamma}_0 = \dot{E}_{VM}/\dot{\gamma}_0 = 1$. As a consequence, if the macroscopic strain rate \dot{E}_{VM} is multiplied or divided by a factor of say 10, without modifying the value of the reference shear rate $\dot{\gamma}_0$, N_{step} will have to be divided or multiplied by 10, if $E_{33\text{final}}$ and Δt are kept constant. The exponent n will be varied in a wide range associated classically with room temperature deformation, that is, between 10 and 200. This model will be used below to reproduce the mechanical behavior of several face centered cubic (FCC) materials (stainless steel and aluminum alloy) extracted from the literature. As in the selected examples, the texture is not documented or assumed to be isotropic, it is considered in the following as only isotropic aggregates, represented by a set of 2016 orientations, uniformly distributed within the Euler space. For all treated examples, the 12 111(110) systems will be considered. It is worth to point out that the aim of the present work is not to demonstrate the relevance of the RC Taylor model, but to be able to study the influence of the simple rate-sensitivity power law in a model allowing a strain-rate gradient within the aggregate. Some tensile curves have been simulated with this approach to illustrate the influence of the various hardening parameters on the global response. These are shown in Figure 1, where it can be clearly seen that, for a given modeling approach, the initial value (resp. final value) of the macroscopic tensile stress is directly proportional to the value of $\tau_0 = \tau_{ini}$ (resp. τ_{sat}). This proportionality is even independent of the selected hardening law for the yield stress value.

3.2. The FE Abaqus code

As the objective is to deal with strain-rate sensitivity in the framework of FE codes, some simulations have also been performed with the FE code Abaqus, which is increasingly used for the simulation of the VP behavior of polycrystalline materials, with the very same description of the single-crystal constitutive law. Again isotropic materials are considered. The simulation of a tensile test with such a code, however, is usually not performed with exactly the same boundary conditions as the ones imposed in the RC Taylor model, since displacements, instead of strain rates, are generally applied in FE simulations. Furthermore, the description of the initial microstructure of the material is also somewhat different, since it accounts for both orientations and positions of the grains within polycrystalline aggregates, whereas only the orientation distribution is accounted for in the Taylor model.

In the present case, the considered isotropic material will be represented by a cubic polycrystalline aggregate of 200 grains, made of a Voronoï tessellation obtained from Neper program [41] and imported in Abaqus CAE using python scripts [42] (see Figure 2a). The grain boundaries are simply defined as the boundaries between zones of different orientations. The choice of a reduced number of orientations compared to the previous case appears here to be a good compromise to obtain both reasonable calculation times and a macroscopic uniaxial tensile stress, as with the RC Taylor model. The polycrystal is submitted to both symmetry boundary conditions and imposed displacement on the upper and lower faces, while on the lateral ones, uniform mixed-orthogonal (or block) conditions have been defined to account for periodicity [43,44] (Figure 2b).

The crystal plasticity mechanical behavior, as defined in (1) to (6), is introduced into Abaqus using a UMAT subroutine, which is extensively described in [33], and the Euler angle set



Figure 2. (a) Example of a Voronoï tessellation made of 200 grains and (b) applied boundary conditions.

 $(\varphi_1, \Phi, \varphi_2)$ of each Voronoï cell is randomly determined in an ORIENT subroutine, describing thus an isotropic angle set.

4. Numerical results

4.1. Case study 1: the case of a rate-insensitive material

The first example treated with the simple rate-sensitive (RS) constitutive law described above is the often encountered case of a material with a very low strain-rate sensitivity (represented by 1/n). This in turns means a very high value of the *n* exponent. This exponent is usually established experimentally from flow stresses obtained at different strain rates but same microstructural state (e.g., dislocation density and thus same stress level), from the slope of the ln–ln stress–strain rate curve [45]:

$$n = \frac{\partial \ln \dot{E}_M}{\partial \ln \Sigma} \Big|_{\Sigma}.$$
(17)

When doing so, values as high as 100–400 have been reported in the literature for various materials (copper, brass, many steels,) and this is why some regularized versions of the present VP formulation have been proposed since calculations performed with very high values of *n* (above 200) are practically impossible to achieve, whatever the model used [29,31]. Another way of solving this problem is to embed the norm of the applied strain rate into the reference shear rate γ_0^s [30], by imposing it to be equal to the applied macroscopic strain rate, that is, $\dot{\gamma}_0^s = \dot{E}_{33\text{imp}}$ in the present case. It is then easy to show that in this case, the macroscopic response does not depend on the selected *n* value, and that the magnitude of the grain stress is affected only by the ratio of the grain strain-rate magnitude and the macroscopic strain-rate magnitude, which is not so large for FCC materials. This procedure is frequently used, especially when simulations are performed for one single macroscopic strain-rate value.

Let us reproduce in this way, an experimental curve obtained at room temperature for a 304L stainless steel, whose texture is assumed to be isotropic [46]. In order to identify the hardening parameters for this case, the reference shear stress has been set equal to the actual strain rate, that is, 0.001 s^{-1} , and the *n* exponent to 21, that is, one of the value most often found in the literature



Figure 3. Tensile curves obtained with the RC Taylor model for the reference case $(\dot{E}_{33imp}/\dot{\gamma_0} = \dot{E}_{VM}/\dot{\gamma_0} = 1)$ and various *n* values. Comparison with the experimental curve obtained by Chavez *et al.* [46] on a 304L stainless steel at room temperature.

Table 1. Imposed and adjusted parameters for the identification of the 304L experimental curve with the RC Taylor model

Imposed test conditions	$\dot{E}_{VM} = \dot{E}_{33imp} = 0.001 \text{ s}^{-1}$
(reference case)	$E_{33 max} = 0.35$
Imposed parameters	$\dot{\gamma}_0^s = \dot{\gamma}_0 = 0.001 \text{ s}^{-1}$ n = 21
Material adjusted parameters	$ au_0^s = 100 ext{ MPa}$ $h_0 = 260 ext{ MPa}, q = 1.1$ $ au_{ ext{sat}} = 300 ext{ MPa}, au_{ ext{ini}} = 100 ext{ MPa}$

"to reproduce negligible viscous behavior" [47] or for material which "possesses a certain ratedependency at medium to high temperatures" [14,34,48–51]. The other material parameters have been manually identified on the experimental curve (see Figure 3); they are listed in Table 1.

It is seen that, for the selected parameters, the agreement, although not 100% perfect, is quite satisfactory. An automatic identification procedure or the selection of a more physically based hardening law would probably allow for a better fit over the whole strain range, but this is out of the scope of the present paper. Much more important, for it is the focus of the present study, it is seen that when $\dot{\gamma}_0 = \dot{E}_{\rm VM}$, the influence of the exponent *n* is indeed very limited and negligible for $n \ge 21$, as reported in many studies using the same law.

But, once this reference case is satisfactorily treated, we may want to see what would happen if we change now the applied strain rate without changing the value of the reference shear stress $\dot{\gamma}_0$. This is illustrated in Figure 4, where simulations have been made using the parameters listed in Table 1, except for the applied normalized strain rate $\dot{E}_{\rm VM}/\dot{\gamma}_0$ which has multiplied or divided by ten, and the exponent *n* which has been varied from 21 to 111. It is seen that, when the normalized strain rate is set to $\dot{E}_{\rm VM}/\dot{\gamma}_0 = 1/10$, there is an influence of the strain rate on the macroscopic response, and that the curves are almost superimposed only for $n \ge 91$. It is interesting to note that the best agreement with the experimental curve is now found for n = 41, which means that both reference strain rate $\dot{\gamma}_0$ and exponent *n* should be considered simultaneously for the identification of the hardening parameters, as done in other works, see



Figure 4. Tensile curves obtained with the RC Taylor model for 2 test parameter sets different from the reference case, and various *n* values. Comparison with the experimental curve obtained by Chavez *et al.* [46] on a 304L stainless steel at room temperature.



Figure 5. Tensile curves obtained with the RC Taylor model and the Abaqus code for five different applied normalized strain-rate values (from 0.0001 up to 1 s^{-1}) and n = 21 or 111.

for example [9]: for the identification of the selected 304L tensile curve, taking $\dot{\gamma}_0 = 10 \times \dot{E}_{VM}$ with n = 41 indeed provides a slightly better fit than $\dot{\gamma}_0 = \dot{E}_{VM}$ with n = 21. It is also seen in Figure 4a that the overall stress level is increasing with n, when $\dot{E}_{VM} < \dot{\gamma}_0$, as a consequence of the used power law.

If the normalized strain rate is set to $\dot{E}_{\rm VM}/\dot{\gamma_0} = 10$, there is again a clear influence of the strain rate on the macroscopic response, and the curves are again almost superimposed only for $n \ge 91$. The overall stress level is now decreasing when n increases, which better correspond to the expected influence of rate sensitivity and has already been observed in some other works. For example, these calculations are in full agreement with the ones performed by Khan *et al.* [9] with a similar power law.

Some curves obtained for two different values of n, namely 21 and 111 and various values of $\dot{E}_{33\text{imp}}/\dot{\gamma}_0$ between 1 and 10^{-4} (that is 4 orders of magnitude) are now presented in Figure 5. These curves have been obtained with the RC Taylor model as well as with Abaqus, with the same material parameters. For the FE simulation, as the elastic part of the curve is also simulated, the following elastic constants have been considered for 304L steel: $C_{11} = 197,500$ MPa, $C_{22} = 125,000$ MPa, and $C_{44} = 122,000$ MPa. As the boundary conditions imposed to simulate a simple tensile test are not exactly expressed in the same way for the two approaches, before performing



Figure 6. Evolution of the macroscopic yield stress as a function of strain rate and strainrate sensitivity exponent for all performed simulations.

comparisons of the two sets of calculations, it has been checked that the macroscopic tensile test was indeed uniaxial in both cases (this would not have been necessarily the case for an anisotropic material). For comparison purposes, the elastic part of the curves has been taken out from the Abaqus simulations. It is interesting to note that the Abaqus curves are systematically below the RC Taylor curves, even though the Taylor model is not used in the pure FC mode (which would actually correspond to an upper bound). This means that the interactions between grains are softer in Abaqus than with the selected RC Taylor model. However, it is clear that the influence of the strain rate for given values of $\dot{\gamma_0}$ and n are similar for both approaches. It is also worth noting that $n \ge 111$, the convergence is very difficult to achieve with Abaqus, depending on the material description and imposed strain rate.

The influence of the normalized strain rate $\dot{E}_{\rm VM}/\dot{\gamma}_0$ and strain-rate sensitivity exponent *n* on the overall yield stress is now presented in Figure 6 for all performed situations. This yield stress is extracted at the first calculation step for the RC Taylor simulations and evaluated at 0.02% plastic strain for the Abaqus simulations. Clearly this stress value does not vary much with the exponent for $\dot{E}_{\rm VM}/\dot{\gamma}_0 = 1$ in both cases, but when this value is changed, the stress can strongly vary. For example, for n = 21, the stress varies from about 180 to 310 MPa when $\dot{E}_{\rm VM}/\dot{\gamma}_0$ varies from 0.0001 to 10 with the RC Taylor model. Similarly, with Abaqus, the stress varies from 170 to about 263 MPa when $\dot{E}_{\rm VM}/\dot{\gamma}_0$ varies from 0.0001 to 1. The response of the material is thus clearly rate sensitive for n = 21. The absence of strict linearity observed in the Abaqus simulations can be attributed to numerical reasons and to the local fluctuations of strain rate and stress, more significant than with the RC Taylor model, associated with weaker interactions between grains.

Now, by calculating the normalized ratio of this tensile yield stress by the reference shear stress (taken here equal to 100 MPa), the RC Taylor and Abaqus responses can be compared to the upper and lower bounds rate-insensitive approaches. It is indeed well known that, for an isotropic FCC metal in the rate-insensitive limit, this normalized stress is equal to 3.06 for the upper bound (Taylor model), 2 for the lower bound (static model) and 2.2 for the Sachs–Kochendorfer model [52]. It is also clear from Figure 6, that this rate-insensitive value can be estimated for the VP RC Taylor and Abaqus models from the yield stress obtained for $\dot{E}_{\rm VM} = \dot{\gamma}_0$ and the highest possible value for the exponent *n*. The so-obtained values are 2.78 for the RC Taylor model and about 2.62 for Abaqus. This confirms that the RC Taylor approximation is closer to the upper bound than to the lower one and that the interactions between grains are slightly softer with Abaqus. This also affects the hardening capacity as seen in Figure 5.



Figure 7. Relationship between strain rate and yield stress extracted from experimental tensile curves obtained on pure aluminum at room temperature [54].

This first study has highlighted the fact that the only way to really neglect rate sensitivity with the power law is to set $\dot{\gamma}_0 = \dot{E}_{VM}$, and to keep the macroscopic strain-rate constant; this is indeed well known from experienced users of VP mean field or full field methods, but much less from new FE code users, if we look at the recent bibliography on the subject.

4.2. Case study 2: the case of a rate-sensitive material

Furthermore, if some alternatives exist to treat efficiently the case of the rate-insensitive materials, the simple power law (1) is still largely used to treat the case of rate-sensitive materials within the static regime (typically for strain rates staying below 1.0 s⁻¹). Of course, as already said, it cannot alone account for the complex rate-sensitivity influence of a real material and the rate sensitivity of the hardening rate should also be considered [9, 32], especially if the treated problems involve strong flow localization [53, 54]. But, if it is not the case, that is, for moderate strains and strain rates, Christodoulou and Jonas [54] have shown that the use of the so-called continuous strain-rate sensitivity as defined by (17), that is, determined at constant structural state, instead of a more instantaneous one, determined during strain rate change tests, compensates to some extent for the neglect of the rate sensitivity of hardening. Thus, even with a rather simple hardening law, by paying attention to the identification procedure, it can still reproduce quite precisely the observed behavior of materials tested at different strain rates and be useful for simulations for which strong localization is not expected. Let us take the example of pure aluminum (99.99%), which is more rate sensitive than the previously studied steel. Some experimental data have been extracted from [53,54] and the rate sensitivity of the material has been estimated using (17) (Figure 7). The resulting estimated n value is found to be equal to 15 for an explored strain-rate range of 0.05 to 0.0005 s^{-1} .

Then, as in the previous example, the identification of the hardening parameters have been made for a reference case, corresponding to $\dot{\gamma_0} = \dot{E}_{\rm VM}$, arbitrarily selected equal to 0.0005 s⁻¹. We have also fixed the value of q to 1.1, classically selected for aluminum. Therefore, three parameters remain to be determined, which are h_0 , $\tau_{\rm ini}$, and $\tau_{\rm sat}$. These three parameters have been manually adjusted to reproduce the curve corresponding to $\dot{E}_{\rm VM} = 0.0005 \, {\rm s}^{-1}$, and validated then by simulating the other curves corresponding to $\dot{E}_{\rm VM} = 0.005 \, {\rm s}^{-1}$ and validated then by simulating the other curves corresponding to $\dot{E}_{\rm VM} = 0.005 \, {\rm s}^{-1}$. The result of this first identification is presented in Table 2 and Figure 8a, in which the elastic parts of the experimental curves have been suppressed. In the validation process, the parameters may be slightly re-adjusted. In that case, they have to be validated again on the reference case.



Figure 8. Stress–strain curves obtained for pure aluminum deformed in tension at various strain rates. The experimental curves are taken from [54] and the simulated ones are obtained with the RC Taylor model, using two sets of parameters (see Table 2): (a) $\dot{\gamma_0} = 0.0005 \text{ s}^{-1}$ and (b) $\dot{\gamma_0} = 0.05 \text{ s}^{-1}$.

Table 2. Imposed and adjusted parameters for the identification of the aluminum experimental curves with the RC Taylor model

$\dot{E}_{\rm VM}$ (s ⁻¹) for the	Imp	posed para	meters	Adjusted parameters			
identified curve	n	$\dot{\gamma_0}$ (s ⁻¹)	q	h_0 (MPa)	$ au_{ m ini}$ (MPa)	$\tau_{\rm sat}$ (MPa)	
0.0005	15	0.0005	1.1	4	15	35	
0.05	15	0.05	1.1	5.7	20	47	

Once the parameters have been selected for the whole explored strain-rate range, it may be desirable to modify the reference case, and thus the value of the associated reference strain rate $\dot{\gamma}_0$. In that case, it is not necessary to perform the whole identification and validation process. It can simply be noticed that for a given model, the results of a 2nd identification procedure should coincide with the result of the 1st one, since the curves to be identified are precisely the same. In other words, if the material parameters are changed, the predicted stress and strain-rate tensors at the level of the grains as well as at the level of the sample should coincide at each step of the calculation and especially at the very beginning and at the saturation level (although this level may be not reached experimentally). From (3) and the definition of τ_{ini} and τ_{sat} , this implies that

$$\frac{\dot{\gamma}_0}{\tau_{\text{ini}}^n} = \text{constant} \quad \text{and} \quad \frac{\dot{\gamma}_0}{\tau_{\text{sat}}^n} = \text{constant}$$
(18)

from which the new values of τ_{ini} and τ_{sat} associated with the new selected value of $\dot{\gamma}_0$ can be extracted. There remains then one single parameter to be identified, namely the hardening coefficient h_0 . The results of this procedure applied to the new reference case $\dot{\gamma}_0 = \dot{E}_{VM} = 0.05 \text{ s}^{-1}$ are also presented in Table 2 and Figure 8b. It is seen that the identified curves are very close to the ones obtained previously for the other set of parameters. Of course, for such a simple model, simulations are quite rapid (typically less than one minute for the calculation of one single curve in the presented examples), and thus the total identification procedure, whether performed manually as in the present case, or through an optimized identification procedure is also quite fast.

Contrary, for FE simulations, the identification of one set of parameters on one reference curve and the validation on the other two would take much longer times (depending on the computer). We can however accelerate the procedure if we first base our identification procedure for FE

Simulation	Fixed parameters $n = 15$, $\dot{E}_{VM} = \dot{\gamma}_0 = 0.0005 \text{ s}^{-1}$, $q = 1.1$				
	Parameters to be identified	h_0 (MPa)	$\tau_{\rm ini}$ (MPa)	$\tau_{\rm sat}$ (MPa)	
sim1 (up to σ_0)	h_{01} , τ_{ini1} , τ_{sat1} issued from the RC Taylor identification	4	15	35	
sim2 (up to $\sigma_{\rm sat}$)	$ au_{ m ini}$ corrected by (19) h_{01} , $ au_{ m sat1}$ issued from the RC Taylor identification	4	14	35	
sim3 (up to $\sigma_{\mathrm{expfinal}}$)	$ au_{ m sat}$ corrected by (20) Identification of h_0 with Abaqus	5	14	45	

Table 3. Imposed and adjusted parameters for the identification of the aluminum experimental curves with the Abaqus FE code

simulations on a preliminary identification step performed with a simpler model like the Taylor one as explained below.

In order to perform the identification of the three presented Al curves with Abaqus, it is first set again n = 15 and q = 1.1. The following elastic constants are also considered for aluminum: $C_{11} = 108,240$ MPa, $C_{12} = 62,160$ MPa, and $C_{44} = 28,410$ MPa. Then one reference case is selected, for example, the same as previous, $\dot{\gamma}_0 = \dot{E}_{\rm VM} = 0.0005 \text{ s}^{-1}$ and a first calculation is performed with Abaqus (sim1) with the three hardening parameters issued from the RC Taylor identification, called in the present case h_{01} , $\tau_{\rm ini1}$, and $\tau_{\rm sat1}$. As the two approaches do not lead to the same response (see Figure 5), it is expected that the predicted curve will not perfectly fit the experimental one. One first correction can be made on $\tau_{\rm ini}$ by using the proportionality relationship between initial stress σ_0 and $\tau_{\rm ini}$, illustrated in Figure 1, that is,

$$\frac{\sigma_{0\text{sim1}}}{\tau_{\text{ini1}}} = \frac{\sigma_{0\text{exp}}}{\tau_{\text{ini}}} \cong \frac{\sigma_{0\text{Tay}}}{\tau_{\text{ini}}}.$$
(19)

In this expression, $\sigma_{0\text{Tay}}$ represents the value of the yield stress estimated by the RC Taylor model. This equation allows us to immediately correct the value of τ_{ini} without any identification procedure. Once the beginning of the curve has been correctly adjusted, we can repeat the procedure for the saturation stress, by considering that, if this level is not reached on the experimental curves, it can be again correctly identified by the RC Taylor model, which means $(\sigma_{\text{sat}})_{\text{Tay}} \cong (\sigma_{\text{sat}})_{\text{exp}}$. The result of a 2nd Abaqus simulation (sim2) performed with h_{01} , τ_{ini} , and τ_{sat1} allows us to write

$$\frac{(\sigma_{\text{sat}})_{\text{Tay}}}{\tau_{\text{sat}}} = \frac{(\sigma_{\text{sat}})_{\text{sim2}}}{\tau_{\text{sat1}}},$$
(20)

and to obtain the correct value of τ_{sat} again without any identification procedure. It is worth noting, that this step implies to be able to perform one simulation with RC Taylor and one simulation with Abaqus up to the saturation level. This may involve convergence problems with Abaqus (especially if the saturation level has reached at very high strains). In this case, a classical identification procedure will have to be applied. In any case, there remains then one single parameter to be identified, which is again the hardening coefficient h_0 . The procedure is summarized in Table 3 and illustrated in Figure 9. It is seen that the agreement between experimental and predicted curves is again quite satisfactory. Again, like for the RC Taylor model, some other materials parameters can be recalculated if we change the value of $\dot{\gamma}_0$ without being obliged to perform the simultaneous identification of all parameters.



Figure 9. Stress–strain curves obtained for pure aluminum deformed in tension at various strain rates. The experimental curves are taken from [53] and the simulated ones are obtained with the RC Taylor model (parameters in Table 2) and with the Abaqus FE code, with the hardening parameters listed in Table 3, evaluated with the proposed identification method.

4.3. Case study 3: the transition between static and dynamic regimes

The previous cases have illustrated clearly the fact that the macroscopic response of a polycrystalline aggregate strongly depends on both normalized strain rate (ratio of imposed over reference strain rates) and strain-rate sensitivity exponent; especially, the only configuration where a strain-rate-independent limit can be reached is for $\dot{E}_{VM} = \dot{\gamma}_0$, with *n* greater than 20. This is true when using simple mean field models such as the RC Taylor one (used here) as well as more complex full field tools such as FE codes. However, when FE simulations are performed, the strain rate, and thus the strain-rate sensitivity, can significantly vary from one point to another, as recently underlined by Shahba and Gosh [10] who studied the behavior of strongly anisotropic Ti alloys. In that case, the choice of the VP parameters $\dot{\gamma}_0$ and *n* is not so trivial if we want, for example, to neglect the influence of strain rate in the whole sample. If one single macroscopic strain rate is investigated (and if the fluctuations within the simulated structure are expected to be moderate enough), then taking $\dot{E}_{VM} = \dot{\gamma}_0$ allows to neglect the strain-rate sensitivity as much as possible and to select an *n* exponent as small as possible to reduce the computation time, without going to extensive multiple slip due to overestimated viscosity. Typically, a value around 20 satisfies these constraints and the hardening parameters can then be simply identified on one experimental curve (as done in the present case for the 304L material). But if several values of macroscopic or microscopic strain rate need to be considered, then the selection of the two parameters can be made according to several procedures:

(i) As suggested by Khan *et al.* [9], it is possible to arbitrarily select for $\dot{\gamma}_0$ one of the investigated values of strain rate and to identify the hardening parameters on the corresponding curve and then, to identify the *n* exponent on the curves obtained for the other strain rates. According to these authors however, this option is not completely satisfactory if the associated hardening law is too simplified, as the one used in the present work, since they argue that it is not possible in this case to reproduce all experimental curves obtained, for example, on aluminum single crystals over a wide range of strain rate with one single *n* parameter. They then propose to select a more complex hardening law, which comprises an additional strain-rate sensitivity parameter. It is worth mentioning though that the behavior of the tested Al single crystals observed at varying strain rates is somewhat in contradiction with the conclusions drawn recently by Shahba and Gosh [10] who claimed that in general (and not only for the Ti alloys), the classical phenomenological power law is valid up to larger strain rates (up to 10^4 s^{-1}) than the ones investigated by Khan *et al.* [8], which do not exceed 10^3 s^{-1} .

- (ii) Alternatively, it can be recognized, as suggested by Canova *et al.* already in 1988 [1] and more recently by Uenishi and Teodosiu [55] or by Shahba and Gosh [10], that one single *n* parameter cannot reproduce all observed behaviors and that there are indeed three different regimes:
 - 1. the quasi-static regime associated with strain rates below 1 s^{-1} ,
 - 2. the dynamic regime associated with strain rates typically above 100 or even 1000 s⁻¹—depending on the authors—these two regimes being associated either with significantly different values of *n* or with different VP flow rules and even different hardening mechanisms [10, 55],
 - 3. the transition regime between the two, within which the *n* value may vary more or less smoothly.

To describe this evolution from quasi-static to dynamic regime, Canova *et al.* [1] proposed a bilinear expression for the evolution of the true stress as a function of the strain rate, whereas Uenishi and Teodosiu [55] proposed a parabolic expression, taking into account the effect of temperature within the dynamic regime. As for Shahba and Gosh [10], they propose a so-called unified flow rule, applicable to a very wide range of applied strain rates and temperatures, "uniquely capable of seamlessly accounting for the effects of location-dependent thermally activated (at low strain rates) and drag-dominated (at high strain rates) mechanisms of dislocation glide without any user intervention". Their law is thus especially well adapted to FE simulations performed for very high strain rates, for which strain rates at different locations in a heterogeneous polycrystalline microstructure are expected to vary significantly even for a uniformly applied macroscopic strain rate.

If the existence of two different regimes (and possibly of a smooth transition between the two) is thus well recognized, it is then easy to conclude that the use of the classical VP flow rule with one single set of parameters (describing both flow and hardening rules) should be allowed within one of these two regimes only. As long as the influence of temperature is not explicitly included, as, for example, in the phenomenological hardening law selected in the present work, it is also easy to recognize that the present formulation is thus more adapted to the quasi-static regime than to the dynamic one (for which additional microscopic deformation mechanisms are expected to occur). In this case, the value of $\dot{\gamma_0}$ should also be selected within the quasi-static regime, which means typically below 1 s^{-1} . If the parameters are identified as previously explained, it is then possible to reproduce the behavior of a material within the whole quasi-static regime with one single set of parameters, as already illustrated for polycrystalline aluminum in Section 4.2 but also for Al single crystals, as illustrated in Figure 10. The experimental curves obtained on Al single crystals deformed in compression by Khan et al. [9] have been reproduced with the RC Taylor model (see parameters in Table 4). It is seen that the three curves corresponding to the quasi-static regime are well reproduced (even better than in the original paper with a different, although also phenomenological hardening law), whereas the one associated with the dynamic regime (strain rate equal to 1000 s^{-1}) is not with the quasi-static parameters. If now the *n* exponent is allowed to be modified for the dynamic regime, then the curve can also be reproduced with the simple RC Taylor model and saturating hardening law. It is worth mentioning though, that the strain-rate sensitivity (1/n) is observed here to decrease at high strain rate, in contradiction with most of the other observations performed in metals [1, 10, 20, 33, 55, 56]. The reason for this quite unusual dynamic behavior of the tested Al single crystals is not explained in Ref. [8].



Figure 10. Experimental (dotted lines from [9]) and simulated (full lines, RC Taylor model) curves for Al single crystals deformed in compression.

Table 4. Imposed and adjusted parameters for the identification of the Al single crystals

 compression curves of Khan *et al.* [9] with the RC Taylor model

Imposed test conditions	$\dot{E}_{VM} = \dot{E}_{33imp} = 1 \text{ to } 0.001 \text{ s}^{-1} \text{ (quasi-static)}$ $\dot{E}_{VM} = \dot{E}_{33imp} = 1000 \text{ s}^{-1} \text{ (dynamic)}$			
Reference case for identification of the material parameters	$\dot{E}_{33imp} = 1$			
Imposed parameters	$\dot{\gamma_0^s} = \dot{\gamma_0} = 1 \text{ s}^{-1}$ n = 21 (quasi-static), n = 41 (dynamic)			
Adjusted parameters	$ au_0^s = 5 \text{ MPa}$ $h_0 = 40 \text{ MPa}, q = 1.1$ $ au_{\text{sat}} = 35 \text{ MPa}, au_{\text{ini}} = 5 \text{ MPa}$			

5. Conclusions

The aim of the present study was to rationalize the use of the simple VP power law, still widely used for its simplicity with mean field and full field approaches, to model the behavior of polycrystalline samples in a wide range of strain rates or temperatures. For this purpose, one mean field (RC Taylor model) and one full field (Abaqus FE code) approaches have been selected, with the very same description of the single-crystal behavior (FCC in the present case). These two approaches have been selected since they can predict heterogeneities of strain, stress, and strain rate within a polycrystalline sample, as experimentally observed. As a consequence, the conclusions drawn below are thought to be also applicable to other modeling choices. Through the simulation of simple tensile tests and the comparison of some simulated and experimental curves, we have shown that:

(i) with such a rate-sensitive single-crystal constitutive law, it is possible to simulate the behavior of rate-sensitive or rate-insensitive materials, in the framework of both mean field or full field approaches, provided that it is remembered that the macroscopic response of a polycrystalline aggregate will depend on both values selected for the reference strain rate $\dot{\gamma}_0$ with respect to the imposed one and the strain-rate sensitivity exponent *n*;

- (ii) for rate-insensitive materials, the strain-rate exponent value which allows to approximate the rate-insensitive limit with sufficient precision depends on the macroscopic strain rate, as long as the $\dot{\gamma_0}$ parameter has been selected. If n = 21 is high enough for a strain rate equal to $\dot{\gamma_0}$, much higher values must be selected, as soon as the strain rate is modified. As a consequence, even for n = 21 and $\dot{E}_{\rm VM} = \dot{\gamma_0}$, some strain-rate sensitivity can be observed within individual grains, as long as the deformation is not prescribed to be uniform within the whole polycrystalline sample and that strain-rate variations are thus observed. Today, this is well known only by a limited part of the FE code users;
- (iii) the VP formulation with one single exponent and associated with a simple hardening law neglecting the temperature effects, is quite adequate to describe the quasi-static regime, that is, typically for strain rates below 1/s (or below $\dot{\gamma_0}$), for rate-sensitive materials. Once all parameters have been identified for one reference case, it is then easy to modify them to treat another reference case. It has been shown that the identification procedure can be considerably simplified in this case, since two of the three hardening parameters can simply be corrected without any identification procedure;
- (iv) the identification of all parameters of the constitutive law with an FE code can be greatly accelerated by relying on the identification previously carried out with a simpler model, such as the RC Taylor model.
- (v) The simple VP formulation studied here can also be used to describe the behavior of rate-sensitive material in an extended range of strain rate (i.e., including both static and dynamic regimes), provided that the exponent *n* is identified separately in both regimes.

Finally, it is worth recalling that this widely used VP formulation is phenomenological. It is thus not adequate to describe completely all the microscopic mechanisms. If this is desired, this simple formulation should be enriched to account for a more precise description and more detailed understanding of strain rate and temperature effects.

Acknowledgments

The authors acknowledge the help of H.T. Nguyen, B. Sadriji, and A.M. Karadaniz (former Univ. Sorbonne Paris Nord students) for their contribution in the realization of a UMAT subroutine. This work has benefited from financial support from the LABEX SEAM through the National Research Agency under the "Investissements d'Avenir program" with the reference numbers ANR-11-LABX-086 and ANR-11-IDEX- 0005-02.

Two of the authors (YC and BB) have learned a lot about the use of Abaqus from P. Gilormini, with whom they have both already had the opportunity to publish research based on the advanced use of the code. They were able to appreciate and share his constant questioning on how the code is implemented, its limits and the way to overcome them, his expertise on writing and decrypting UMATs, and his deep competence also on crystal viscoplasticity. The present paper is partly a legacy of these previous collaborations with P. Gilormini.

References

- G. R. Canova, C. Fressengeas, A. Molinari, U. F. Kocks, "Effect of rate sensitivity on slip system activity and lattice rotation", *Acta Metall.* 36 (1988), no. 8, p. 1961-1970.
- [2] L. S. Toth, P. Gilormini, J. J. Jonas, "Effect of rate sensitivity on the stability of torsion textures", *Acta Metall.* **36** (1988), no. 12, p. 3077-3091.
- [3] G. I. Taylor, "The mechanism of plastic deformation of crystals. Part I. Theoretical", *Proc. R. Soc. Lond. A* 145 (1934), no. 855, p. 362-387.
- [4] R. A. Lebensohn, C. N. Tome, P. P. Castaneda, "Self-consistent modelling of the mechanical behaviour of viscoplastic polycrystals incorporating intragranular field fluctuations", *Phil. Mag.* 87 (2007), no. 28, p. 4287-4322.

- [5] J. W. Hutchinson, "Bounds and self-consistent estimates for creep of polycrystalline materials", Proc. R. Soc. Lond. A 348 (1976), p. 101-127.
- [6] I. J. Beyerlein, C. N. Tomé, "Modeling transients in the mechanical response of copper due to strain path changes", Int. J. Plast. 23 (2007), no. 4, p. 640-664.
- [7] V.-T. Phan, T.-D. Nguyen, Q.-H. Bui, G. Dirras, "Modelling of microstructural effects on the mechanical behavior of ultrafine-grained Nickel using crystal plasticity finite element model", Int. J. Eng. Sci. 94 (2015), p. 212-225.
- [8] B. Klusemann, B. Svendsen, H. Vehoff, "Investigation of the deformation behavior of Fe–3%Si sheet metal with large grains via crystal plasticity and finite-element modeling", *Comput. Mater. Sci.* 52 (2012), no. 1, p. 25-32.
- [9] A. S. Khan, J. Liu, J. W. Yoon, R. Nambori, "Strain rate effect of high purity aluminum single crystals: experiments and simulations", Int. J. Plast. 67 (2015), p. 39-52.
- [10] A. Shahba, S. Ghosh, "Crystal plasticity FE modeling of Ti alloys for a range of strain-rates. Part I: a unified constitutive model and flow rule", Int. J. Plast. 87 (2016), p. 48-68.
- [11] G. Monnet, L. Vincent, B. Devincre, "Dislocation-dynamics based crystal plasticity law for the low- and high-temperature deformation regimes of bcc crystal", *Acta Mater*. **61** (2013), no. 16, p. 6178-6190.
- [12] P. Shanthraj, M. A. Zikry, "Dislocation density evolution and interactions in crystalline materials", Acta Mater. 59 (2011), no. 20, p. 7695-7702.
- [13] R. J. Asaro, "Crystal Plast", J. Appl. Mech. 50 (1983), p. 921-934.
- [14] R. Hill, J. R. Rice, "Constitutive analysis of elastic-plastic crystals at arbitrary strain", J. Mech. Phys. Solids 20 (1972), p. 401-413.
- [15] C. Miehe, J. Schröder, "A comparative study of stress update algorithms for rate-independent and rate-dependent crystal plasticity", Int. J. Numer. Methods Eng. 50 (2001), p. 273-298.
- [16] J. W. Hutchinson, K. W. Neale, "Influence of strain-rate sensitivity on necking under uniaxial tension", Acta Metall. 25 (1977), no. 8, p. 839-846.
- [17] G. Vadillo, J. A. Rodríguez-Martínez, J. Fernández-Sáez, "On the interplay between strain rate and strain rate sensitivity on flow localization in the dynamic expansion of ductile rings", *Int. J. Solids Struct.* 49 (2012), no. 3–4, p. 481-491.
- [18] F. Zhou, J. F. Molinari, K. T. Ramesh, "Effects of material properties and strain rate on the fragmentation of brittle materials", Int. J. Fract. 139 (2006), p. 169-196.
- [19] R. J. Asaro, A. Needleman, "Overview no. 42 Texture development and strain hardening in rate dependent polycrystals", Acta Metall. 33 (1985), no. 6, p. 923-953.
- [20] R. W. Klopp, R. J. Clifton, T. G. Shawki, "Pressure-shear impact and the dynamic viscoplastic response of metals", *Mech. Mater.* 4 (1985), no. 3, p. 375-385.
- [21] Z. Leng, H. Pan, Z. Niu, C. Guo, Q. Zhang, Y. Chang, M. Zhang, F. Jiang, "Mechanical behavior, deformation and damage mechanisms of Mg–RY–Zn alloy under high strain rate", *Mater. Sci. Eng. A* **651** (2016), p. 336-340.
- [22] A. Bintu, G. Vincze, R. C. Picu, A. B. Lopes, "Effect of symmetric and asymmetric rolling on the mechanical properties of AA5182", *Mater. Design* 100 (2016), p. 151-156.
- [23] J. Luo, M. Li, W. Yu, H. Li, "The variation of strain rate sensitivity exponent and strain hardening exponent in isothermal compression of Ti–6Al–4V alloy", *Mater. Design* **31** (2010), no. 2, p. 741-748.
- [24] J. Luo, J. Gao, L. Li, M. Q. Li, "The flow behavior and the deformation mechanisms of Ti–6Al–2Zr–2Sn–2Mo–1.5Cr– 2Nb alloy during isothermal compression", J. Alloys Compd. 667 (2016), p. 44-52.
- [25] Z. N. Mao, X. H. An, X. Z. Liao, J. T. Wang, "Opposite grain size dependence of strain rate sensitivity of copper at low vs high strain rates", *Mater. Sci. Eng. A* 738 (2018), p. 430-438.
- [26] L. Peroni, M. Scapin, "Experimental analysis and modelling of the strain-rate sensitivity of sheet niobium", EPJ Web Conf. 183 (2018), article no. 01014.
- [27] A. Rusinek, J. A. Rodríguez-Martínez, A. Arias, "A thermo-viscoplastic constitutive model for FCC metals with application to OFHC copper", *Int. J. Mech. Sci.* **52** (2010), no. 2, p. 120-135.
- [28] S. Fréchard, A. Redjaïmia, E. Lach, A. Lichtenberger, "Dynamical behaviour and microstructural evolution of a nitrogen-alloyed austenitic stainless steel", *Mater. Sci. Eng. A* 480 (2008), no. 1, p. 89-95.
- [29] M. Knezevic, M. Zecevic, I. J. Beyerlein, R. A. Lebensohn, "A numerical procedure enabling accurate descriptions of strain rate-sensitive flow of polycrystals within crystal visco-plasticity theory", *Comput. Meth. Appl. Mech. Eng.* 308 (2016), p. 468-482.
- [30] S. Kok, A. J. Beaudoin, D. A. Tortorelli, "A polycrystal plasticity model based on the mechanical threshold", Int. J. Plast. 18 (2002), no. 5, p. 715-741.
- [31] S. Forest, M. B. Rubin, "A rate-independent crystal plasticity model with a smooth elastic–plastic transition and no slip indeterminacy", *Eur. J. Mech. A Solids* **55** (2016), p. 278-288.
- [32] M. Zecevic, M. Knezevic, "A new visco-plastic self-consistent formulation implicit in dislocation-based hardening within implicit finite elements: application to high strain rate and impact deformation of tantalum", *Comput. Meth. Appl. Mech. Eng.* 341 (2018), p. 888-916.

- [33] Y. Huang, A User-Material Subroutine Incorporating Single Crystal Plasticity in the ABAQUS Finite Element Program, 1991, Unknown.
- [34] L. Hu, S. Jiang, Y. Zhang, D. Sun, "Crystal plasticity finite element simulation of NiTi shape memory alloy based on representative volume element", *Met. Mater. Int.* 23 (2017), p. 1075.
- [35] D. Peirce, R. J. Asaro, A. Needleman, "An analysis of nonuniform and localized deformation in ductile single crystals", *Acta Metall.* 30 (1982), no. 6, p. 1087-1119.
- [36] R. J. Asaro, "Micromechanics of crystals and polycrystals", in Adv. Appl. Mech. (W. H. John, Y. W. Theodore, eds.), Elsevier, Cambridge, MA, USA, 1983, p. 1-115.
- [37] M. Arminjon, B. Bacroix, "On plastic potentials for anisotropic metals and their derivation from the texture function", *Acta Mech.* **88** (1991), no. 3–4, p. 219-243.
- [38] G. Sachs, "Zur Ableitung einer Filebedingung", Z. Vereines Deutscher Ingenieure 72 (1928), p. 734-736.
- [39] A. Kochendorfer, Plastiche Eigenschaften von Kristallen und Metallischen Werkstoffen, Springer, Berlin, 1941.
- [40] B. Bacroix, S. Queyreau, D. Chaubet, E. Siv, T. Chauveau, "The influence of the cube component on the mechanical behaviour of copper polycrystalline samples in tension", *Acta Mater.* 160 (2018), p. 121-136.
- [41] R. Quey, P. R. Dawson, F. Barbe, "Large-scale 3D random polycrystals for the finite element method: Generation, meshing and remeshing", *Comput. Meth. Appl. Mech. Eng.* 200 (2011), no. 17, p. 1729-1745.
- [42] Simulia, Abaqus Scripting User's Manual, Dassault Système, Providence, Rhode Island, USA, 2011.
- [43] A. Salahouelhadj, H. Haddadi, "Estimation of the size of the RVE for isotropic copper polycrystals by using elasticplastic finite element homogenisation", *Comput. Mater. Sci.* 48 (2010), no. 3, p. 447-455.
- [44] Y. Charles, R. Estevez, E. Maire, Y. Brechet, "Modelling the competition between interface debonding and particle fracture using a plastic strain dependent cohesive zone", *Eng. Fract. Mech.* **77** (2010), no. 4, p. 705-718.
- [45] W. F. Hosford, Mechanical Behaviour of Materials, Cambridge University Press, New York, NY, USA, 2010.
- [46] S. A. Chavez, G. E. Korth, D. M. Harper, T. J. Walker, "High-temperature tensile and creep data for Inconel 600, 304 stainless steel and SA106B carbon steel", *Nucl. Eng. Design* 148 (1994), no. 2, p. 351-363.
- [47] S. El Shawish, L. Cizelj, "Combining single-and poly-crystalline measurements for identification of crystal plasticity parameters: application to austenitic stainless steel", *Crystals* **7** (2017), p. 181.
- [48] Y. Zhang, S. Jiang, L. Hu, Y. Zhao, D. Sun, "Investigation of primary static recrystallization in a NiTiFe shape memory alloy subjected to cold canning compression using the coupling crystal plasticity finite element method with cellular automaton", *Model. Simul. Mater. Sci. Eng.* 25 (2017), article no. 075008.
- [49] K. Teferra, L. Graham-Brady, "A random field-based method to estimate convergence of apparent properties in computational homogenization", *Comput. Meth. Appl. Mech. Eng.* 330 (2018), p. 253-270.
- [50] L. Hu, S. Jiang, T. Zhou, J. Tu, L. Shi, Q. Chen, M. Yang, "Multiscale modeling of polycrystalline NiTi shape memory alloy under various plastic deformation conditions by coupling microstructure evolution and macroscopic mechanical response", *Mater. Design* 10 (2017), p. 1172.
- [51] L. Zhao, P. Chakraborty, M. R. Tonks, I. Szlufarska, "On the plastic driving force of grain boundary migration: a fully coupled phase field and crystal plasticity model", *Comput. Mater. Sci.* **128** (2017), p. 320-330.
- [52] A. Belkhabbaz, B. Bacroix, R. Brenner, "Investigation of the elastoplastic behavior of FCC polycrytals using a FFT numerical scheme", *Roumanian J. Techn. Sci. – Appl. Mech.* 60 (2015), p. 5-23.
- [53] N. Christodoulou, J. J. Jonas, "Flow localization in OFHC Cu and 99.99% Al", Acta Metall. 33 (1985), no. 4, p. 719-730.
- [54] N. Christodoulou, J. J. Jonas, "Work hardening and rate sensitivity material coefficients for OFHC Cu and 99.99% Al", Acta Metall. 32 (1984), p. 1655-1668.
- [55] A. Uenishi, C. Teodosiu, "Constitutive modelling of the high strain rate behaviour of interstitial-free steel", Int. J. Plast. 20 (2004), p. 915-936.
- [56] J. D. Campbell, in Dynamic Plasticity of Metals (U. CISM, ed.), Springer, Berlin, 1970.



Contributions in mechanics of materials

Multiscale modeling of the effective viscoplastic behavior of Mg₂SiO₄ wadsleyite: bridging atomic and polycrystal scales

O. Castelnau^{*, a}, K. Derrien^a, S. Ritterbex^{b, c}, P. Carrez^b, P. Cordier^{b, d} and H. Moulinec^e

^{*a*} Laboratoire PIMM, Arts et Metiers Institute of Technology, CNRS, Cnam, HESAM University, 151 boulevard de l'Hopital, 75013 Paris, France

 b Univ. Lille, CNRS, INRAE, Centrale Lille, UMR 8207 - UMET - Unité Matériaux et Transformations, F-59000 Lille, France

 c Geodynamics Research Center, Ehime University, 2-5 Bunkyo-cho, Matsuyama 790-8577, Japan

^{*d*} Institut Universitaire de France, 1 rue Descartes, 75005 Paris, France

^e Aix Marseille Univ, CNRS, Centrale Marseille, LMA UMR 7031, Marseille, France

E-mails: olivier.castelnau@ensam.eu (O. Castelnau), katell.derrien@ensam.eu (K. Derrien), ritterbex.sebastian_arthur_willem.us@ehime-u.ac.jp (S. Ritterbex), philippe.carrez@univ-lille.fr (P. Carrez), patrick.cordier@univ-lille.fr (P. Cordier), moulinec@lma.cnrs-mrs.fr (H. Moulinec)

Abstract. The viscoplastic behavior of polycrystalline Mg_2SiO_4 wadsleyite aggregates, a major high pressure phase of the mantle transition zone of the Earth (depth range: 410–520 km), is obtained by properly bridging several scale transition models. At the very fine nanometric scale corresponding to the dislocation core structure, the behavior of thermally activated plastic slip is modeled for strain-rates relevant for laboratory experimental conditions, at high pressure and for a wide range of temperatures, based on the Peierls–Nabarro–Galerkin model. Corresponding single slip reference resolved shear stresses and associated constitutive equations are deduced from Orowan's equation in order to describe the average viscoplastic behavior at the grain scale, for the easiest slip systems. These data have been implemented in two grain-polycrystal scale transition models, a mean-field one (the recent Fully-Optimized Second-Order Viscoplastic Self-Consistent scheme of [1]) allowing rapid evaluation of the effective viscosity of polycrystalline aggregates, and a full-field (FFT based [2, 3]) method allowing investigating stress and strain-rate localization in typical microstructures and heterogeneous activation of slip systems within grains. Calculations have been performed at pressure and temperatures relevant for in-situ conditions. Results are in very good agreement with available mechanical tests conducted at strain-rates typical for laboratory experiments.

Keywords. Earth mantle, Multiscale modelling, Dislocations, Polycrystal, Viscoplasticity.

Manuscript received 21st July 2020, revised 15th November 2020, accepted 17th November 2020.

^{*} Corresponding author.

1. Introduction

The flow of rocks in the Earth's mantle controls many large-scale geodynamic processes. Among them, mantle convection is of primary importance since it constitutes the main mechanism for the Earth to evacuate internal heat, and it drives continental drift and associated seismic events. Quantitative modeling of mantle convection, which also allows investigation of past dynamics and prediction of future events, depends on our understanding of the rheology of rocks under thermo-mechanical conditions that prevail in the Earth's interior.

The Earth's mantle extends to ca. 2900 km depth where pressure reaches 130 GPa and temperature is exceeding 3000 K. In response to increasing pressure and temperature with depth, minerals observed in rocks from the upper mantle transform at depth to denser assemblages. These phase transitions are responsible to global discontinuities of the velocities of seismic waves across the corresponding depths. The 410 km discontinuity is generally thought to be caused by the phase transition of olivine (the low pressure phase of (Mg, Fe)₂SiO₄) to wadsleyite and the 520 km discontinuity by the phase transition of wadsleyite to ringwoodite. These phase transitions give rise to the so-called transition zone which ends at the 670 km discontinuity where the lower mantle begins.

The transition zone is a major layer between the upper and lower mantles and is expected to influence the whole mantle convection depending on its rheological properties. Information on the mechanical properties of rocks come primarily from laboratory mechanical tests. This experimental approach is however rendered very challenging since the *P*, *T* conditions are in the range of 15 GPa and 1600 K for wadsleyite in the upper transition zone. Furthermore, rheological laws deduced from laboratory experiments at strain-rates of typically 10^{-5} s⁻¹ need to be extrapolated by ~10 orders of magnitudes to the extremely low strain-rate conditions (ca. 10^{-15} s⁻¹) prevailing during Earth's mantle convection.

In that context, the computational approach is an alternative to infer the viscoplastic behavior of mantle rocks and offers the potentiality to tackle the extremely low strain-rate conditions issue, provided all relevant and physical-based deformation mechanisms at play in the mantle are properly taken into account. However, this requires bridging several characteristic length scales, from sub-nanometer to sub-meter. To be able to glide, dislocations must overcome their intrinsic lattice friction, which strongly depends on their structure at the atomic scale (subnm). Core structures of dislocations belonging to given slip systems can be calculated using the Peierls–Nabarro–Galerkin method [4], relying on first principle simulations of generalized stacking fault (GSF) surfaces. This allows addressing accurately the effect of pressure on atomic bonding. Intrinsic lattice friction is then calculated and quantified by the Peierls potential. At finite temperatures, dislocation glide mobility results from thermally-activated motion of dislocations over their Peierls potentials. The obtained energy barriers for dislocation glide can then be combined with Boltzmann statistics to provide a constitutive relation at the grain level (mm scale), for each available slip systems [5, 6].

To address the rheology of polycrystalline aggregates (sub-m scale), a second scale transition, from the grain to the polycrystal, must be carried out. In minerals, this is another difficult task as few independent slip systems are generally available. For example, olivine exhibits less than 4 independent dislocation slip systems leading to an extreme viscoplastic anisotropy at the grain scale and, as a consequence, a quite challenging application of mean-field homogenization methods. Indeed, as shown by Pierre Gilormini [7, 8], many earlier homogenization methods provides an unrealistically stiff (i.e. violating a rigourous upper bound) estimation of the effective rheology. Using the more advanced Partially Optimized Second-Order (POSO) Self-Consitent (SC) estimate proposed by [9], it has been shown that the overall viscoplastic behavior (flow stress *and* stress sensitivity) in olivine is controlled by the behavior of accommodation mechanisms (dislocation climb, diffusion, grain boundary sliding, ...) which are not clearly identified yet [10–13]. Moreover, rocks are subjected to very large strain during mantle convection, inducing the development of pronounced crystallographic textures that can be partly characterized by the anisotropic velocities of seismic waves [14]. The associated effective viscoplatic anisotropy may strongly influence the mantle flow pattern in-situ [15, 16]. However, the prediction of in-situ texture distribution is tedious as, besides temperature-related mechanisms such as recrystallization and grain growth, the prediction of deformation texture is also sensitive to the used homogenization scheme [17]. Regarding the transition zone, texture development associated with large shear strain have been investigated in [18] for wadsleyite using an earlier extension of the SC scheme (the tangent approach of [19]) whose inconsistencies have been described in [7,8] and partly corrected in [20]. In Tommasi *et al.* paper [18], the effective viscosity of wadsleyite and its relation with the active slip systems has not been investigated.

The POSO version of the Self-Consistent scheme was up to recently the most accurate meanfield method for predicting the effective viscoplastic behavior of highly anisotropic materials such as mantle rocks. As shown in e.g. [21, 22], this method provides results in very good agreement with computational homogenization (providing reference solutions) e.g. based on the Fourier Transform (FFT, sometimes also denoted spectral method) introduced by [3]. Among its advantages, the POSO-SC method complies with the variational upper bound [23] that has been applied to highly anisotropic polycrystals in [24–26]. However, it lacks duality (stress and strain formulations lead to different results) and the link between the behavior of the thermoelastic polycrystal used as a reference (called Linear Comparison Composite, LCC) and the non-linear viscoplastic behavior of the real polycrystal of interest requires complex computation of some corrective terms [27] that is rarely carried out in practice. The above mentioned limitations of POSO-SC have been ruled out recently with the Fully Optimized Second-Order (FOSO) method [1, 28], which has, to the best of our knowledge, only been applied yet to porous sea ice [29].

This paper focusses on the rheological behavior of Mg_2SiO_4 wadsleyite. In Section 2, we provide the strength of the various slip systems based on computational mineral physics and the corresponding P-T dependent constitutive relation at the grain scale. We present in Section 3 how the mean-field (FOSO-SC) and full-field (FFT) homogenization schemes have been applied. Results are then presented and discussed in Section 4 and compared to the available experimental results.

2. Viscoplastic behavior of slip systems

2.1. At the dislocation scale from generalized stacking fault energies

The modelling of gliding properties of dislocations in wadsleyite has been initiated by Ritterbex *et al.* [6] for the Mg end-member of $(Mg, Fe)_2SiO_4$, i.e. the pure magnesian Mg_2SiO_4 composition. The first step of the model is to identify the most important slip systems and to determine the specific atomic arrangements which build the dislocation core. Such calculations are usually performed at the atomic scale. However, the long range displacement fields of dislocations impose the use of classical molecular dynamic simulation instead of highly accurate density functional theory based *ab-initio* calculations.

Another option to compute dislocation core properties is to use the Peierls–Nabarro model which relies on generalized stacking fault (GSF) energies. GSF's probes the ability of a perfect crystal structure to undergo a rigid body shear localized in a specific plane, and, combined with the Peierls–Nabarro approach, allows to search for potential easy slip systems of any crystalline materials. More importantly, such calculations do not require supercells containing large numbers



Figure 1. Dislocation modeling in Mg₂SiO₄ wadsleyite at 15 GPa (a) Generalized Stacking fault calculated by [30] for a {101} plane. The easy shear path is along $\langle 111 \rangle$. (b) Dislocation core profile for a $1/2\langle 111 \rangle$ {101} dislocation calculated from the Peierls–Nabarro Galerkin method. *S* is the disregistry profile.

of atoms so they can be performed by using *ab initio* methods, ensuring that the effect of high pressure on bond energy or ion interactions is accurately taken into account. In orthorhombic wadsleyite (lattice parameters a = 5.70 Å, b = 11.44 Å, c = 8.26 Å at 18.5 GPa), the easiest slip systems are [100](010) and 1/2(111){101}. An example is provided in Figure 1a which shows a GSF calculated in a {101} plane at 15 GPa. On the vertical axis is represented the excess energy associated with a rigid body shift in the considered plane. One can deduce that in {101} planes, the easiest shear path is along (111) directions.

As recalled above, the GSF serves as input for the Peierls–Nabarro model used to compute dislocation core structures by minimization of the total energy of the system composed of the elastic strain energy and the inelastic stacking fault energy across the potential glide planes. Ritterbex *et al.* [6] used a generalization of the Peierls–Nabarro model in the framework of the element-free Galerkin method which allows for the introduction of multiple glide planes in order to describe more general core structures involving spreading in several planes. In wadsleyite, for all slip systems considered, dislocations exhibit a planar core involving two well-separated partial dislocations enclosing a stacking fault (see an illustration in Figure 1b).

Besides the core structure, i.e. the atomic arrangement within the vicinity of the dislocation core, Ritterbex *et al.* computed the lattice friction experienced by dislocations on each slip system. The amount of lattice friction is often described as the height of the Peierls potential or quantified through the maximum of the derivative of the potential, the Peierls stress. Nevertheless, whereas Peierls stress or Peierls potential are strictly related to the core of dislocations, the glide of dislocations at finite temperature is thermally activated. Thermal activation means that glide at finite temperature is controlled by the nucleation and propagation of unstable kink-pairs over the Peierls barrier, assisted by the resolved shear stress. Ritterbex *et al.* provided a full description of these kink processes according to an elastic interaction model [31]. For wadsleyite, since dislocations are dissociated, the kink-pair mechanism may involve different types of kink nucleation events depending on the stress regime. Indeed, kink nucleation must occur on both partials with sequences of events that can either be correlated or uncorrelated leading to distinct nucleation enthalpy depending on the stress regime [32]. Finally, from the kink-pair nucleation enthalpy, the dislocation velocity law can be formulated as a function of temperature and resolved shear stress. The corresponding constitutive relation has to account for these two stress

Table 1. Rheological parameters of $1/2(110){110}$ and [100](010) screw dislocations in wadsleyite at 15 GPa for $\tau \ge \tau_c$. Units are m·s⁻¹ for *A*, J for *B*, and MPa for τ

	τ_c	τ_p	A_c	B_c	α_c	β_c	A_u	B_u	α_u	β_u
$1/2\langle 111\rangle \{101\}$	455	3500	2190	1.97×10^{-18}	0.5	1.6	4380	8.49×10^{-19}	1.0	5.0
[100](010)		4800	1208	2×10^{-18}	0.5	1.03				

regimes, leading to two different expressions of the shear-rate on the slip system

$$\dot{\gamma}_c(\tau) = A_c \sqrt{\rho_m} \exp\left\{-\frac{B_c}{k_b T} \left[1 - \left(\frac{\tau}{\tau_p}\right)^{\alpha_c}\right]^{\beta_c}\right\}$$
(1)

$$\dot{\gamma}_{u}(\tau) = A_{u}\sqrt{\rho_{m}}\frac{\tau-\tau_{c}}{\tau}\exp\left\{-\frac{B_{u}}{k_{b}T}\left[1-\left(\frac{\tau-\tau_{c}}{\tau_{p}}\right)^{\alpha_{u}}\right]^{\rho_{u}}\right\}$$
(2)

where indexes *c* and *u* stand for correlated and uncorrelated kink-pair nucleation regimes, respectively. Here, ρ_m is the density of mobile dislocations, *T* is the temperature, τ_p is the Peierls stress, *A* and *B* are two constants, and $k_b \simeq 1.3806 \times 10^{-23} \text{ m}^2 \cdot \text{kg} \cdot \text{s}^{-2} \cdot \text{K}^{-1}$ is the Boltzmann constant. *A* is related to the shear-rate at $\tau = \tau_c$, marking the transition stress between correlated and uncorrelated regimes, whereas *B* is an activation energy. The resolved shear stress τ acting on the slip system is given by

$$\tau = \mathbf{S} : \boldsymbol{\sigma} \tag{3}$$

where **S** is the Schmid tensor (see Appendix A), σ the local deviatoric stress tensor, and ":" the twice contracted product. The resulting shear-rate on a given slip system is given by

$$\dot{\gamma}(\tau) = \dot{\gamma}_c(\tau) \tag{4}$$

when $\tau \leq \tau_c$ and

$$\dot{\gamma}(\tau) = \frac{1}{2} [\dot{\gamma}_c(\tau) + \dot{\gamma}_u(\tau)] \tag{5}$$

otherwise. For waldsleyite, the modelling that has been performed for dislocations with screw character which are the rate-limiting ones. The easiest slips include the family $1/2\langle 111\rangle\{101\}$ with four individual slip systems $(1/2[11\bar{1}](101), 1/2[1\bar{1}\bar{1}](101), 1/2[111](10\bar{1}), 1/2[1\bar{1}1](10\bar{1}))$, and the additional system [100](010). At stresses higher than $\tau_c = 455$ MPa as considered here, the rheology of $1/2\langle 111\rangle\{101\}$ screw dislocations at 15 GPa is governed by (5) whereas that for [100](010) is given by (4). The corresponding rheological coefficients are indicated in Table 1. Finally, the local strain-rate $\dot{\epsilon}$ at position **x** inside a grain, resulting from the glide of dislocations on all slip systems, is given by

$$\dot{\boldsymbol{\varepsilon}}(\mathbf{x}) = \sum_{s=1}^{S} \mathbf{S}_{(s)} \dot{\boldsymbol{\gamma}}_{(s)}(\mathbf{x})$$
(6)

with $S_{(s)}$ the Schmid tensor for system (*s*) and *S* (= 5) the number of slip systems. Note that the four systems $1/2\langle 111\rangle \{101\}$ are independent and do not allow axial strain of the crystal lattice along the lattice direction **b** (see Appendix A). The fifth system [100](010) does not add any additional degree of freedom, and therefore the wadsleyite crystal is left with only four independent slip systems. As discussed in [13, 33], four systems are sufficient to accommodate locally any viscoplastic deformation of the polycrystalline aggregate.

2.2. Power law approximation of slip system behavior

Although homogenization models could in principle be applied with constitutive relations having an exponential form as (4) and (5), it is more common to use instead a power-law behavior

$$\dot{\gamma}_{pl} = \dot{\gamma}_0 \left| \frac{\tau}{\tau_0} \right|^{n-1} \frac{\tau}{\tau_0} \tag{7}$$

where the index *pl* stands for power-law. This is the form commonly implemented in homogenization codes. Here, $\dot{\gamma}_0$ and τ_0 are two constants to be determined, a reference shear-rate and a reference shear stress, respectively. We consider here the value $\dot{\gamma}_0 = 10^{-5} \text{ s}^{-1}$ as a typical strain-rate achieved during laboratory mechanical tests. Form (7) can be seen as a power-law approximation of (4) or (5) valid in a range of τ values close to a given reference value denoted τ_r . To find τ_0 , one expresses that the power-law approximation is tangent to the exponential form at $\tau = \tau_r$:

$$\dot{\gamma}_{pl}(\tau_r) = \dot{\gamma}(\tau_r), \quad \left. \frac{\partial \dot{\gamma}_{pl}}{\partial \tau} \right|_{\tau = \tau_r} = \left. \frac{\partial \dot{\gamma}}{\partial \tau} \right|_{\tau = \tau_r}.$$
(8)

This leads to the following expression for the stress exponent n

$$n = \frac{\partial \log \dot{\gamma}}{\partial \log \tau} \Big|_{\tau = \tau_r} = \frac{\tau_r}{\dot{\gamma}(\tau_r)} \left. \frac{\partial \dot{\gamma}}{\partial \tau} \right|_{\tau = \tau_r}$$
(9)

while the reference stress τ_0 reads

$$\tau_0 = \tau_r \left(\frac{\dot{\gamma}(\tau_r)}{\dot{\gamma}_0}\right)^{-1/n}.$$
(10)

These can be calculated using the following expressions for the derivatives in (9)

$$\frac{\partial \dot{\gamma}_c}{\partial \tau} = -a \exp\left\{ b \left[1 - \left(\frac{\tau}{\tau_p}\right)^{\alpha} \right]^{\beta} \right\} \frac{\alpha \beta b}{\tau} \left(\frac{\tau}{\tau_p}\right)^{\alpha} \left(1 - \left(\frac{\tau}{\tau_p}\right)^{\alpha} \right)^{\beta-1}$$
(11)

$$\frac{\partial \dot{\gamma}_u}{\partial \tau} = a \exp\left\{ b \left[1 - \left(\frac{\tau - \tau_c}{\tau_p} \right)^{\alpha} \right]^{\beta} \right\} \left\{ \frac{\tau_c}{\tau^2} - \frac{\alpha \beta b}{\tau} \left(\frac{\tau - \tau_c}{\tau_p} \right)^{\alpha} \left(1 - \left(\frac{\tau - \tau_c}{\tau_p} \right)^{\alpha} \right)^{\beta - 1} \right\}$$
(12)

with $a = A_{\sqrt{\rho_m}}$, $b = -B/(k_b T)$, and with indexes *c* and *u* left for the sake of clarity.

Power-law approximations of the exponential constitutive relations have been calculated under typical conditions corresponding to high *P*, *T* laboratory mechanical tests, i.e. considering a density of mobile dislocations $\rho_m = 10^{12} \text{ m}^{-2}$ and a strain-rate $\dot{\gamma} = \dot{\gamma}_0$ (leading to $\tau_0 = \tau_r$). It is observed that the power-law provides a very good approximation of the original exponential behavior, as illustrated in Figure 2a at 1700 K. The resulting power-law parameters are provided in Figure 2b for a large temperature range. It turns out that the rheology of the various slip systems is strongly non-linear. Values of *n* are between ~17 and 100 for the considered temperature range, with a different behavior for both slip systems. Moreover, the reference shear stress for the system [100](010) is significantly stiffer (~3 times) than for $1/2\langle 111\rangle$ {101}.

3. Polycrystal modeling

We now proceed to the next scale transition, i.e. estimation of the effective (average) viscoplastic behavior of a representative polycrystalline aggregate composed of a large number of grains. We consider the case for which grains are randomly oriented (random crystallographic texture) and exhibit equiaxed shapes so that the effective behavior can be considered as isotropic. In the sequel, effective (or homogenized) quantities are denoted with a tilde (.) and volume average ones with a bar (.). The effective viscoplastic behavior thus reads

$$\dot{\bar{\varepsilon}}_{eq} = \dot{\gamma}_0 \left(\frac{\bar{\sigma}_{eq}}{\bar{\sigma}_0}\right)^n \tag{13}$$



Figure 2. (left) Power-law approximations of the slip system behaviors for wadsleyite at $\dot{\gamma} = 10^{-5} \text{ s}^{-1}$: comparison of the original exponential constitutive relation with its power-law approximation (fit) at 1700 K. (right) Evolution of τ_0 and *n* of the power-law approximations of the slip system behavior at $\dot{\gamma} = 10^{-5} \text{ s}^{-1}$, for various temperatures.

where $\bar{\sigma}_{eq}$ is the effective von Mises equivalent stress ($\bar{\sigma}_{eq} = \sqrt{(3/2)\bar{\sigma}_{ij}\bar{\sigma}_{ij}}$), $\dot{\bar{\varepsilon}}_{eq}$ is the effective von Mises equivalent strain-rate ($\dot{\bar{\varepsilon}}_{eq} = \sqrt{(2/3)\dot{\bar{\varepsilon}}_{ij}\dot{\bar{\varepsilon}}_{ij}}$), where $\bar{\sigma} = \langle \sigma(\mathbf{x}) \rangle$ and $\dot{\bar{\varepsilon}} = \langle \dot{\varepsilon}(\mathbf{x}) \rangle$ are the mean deviatoric stress and strain-rate tensors respectively.

To estimate the effective reference stress $\tilde{\sigma}_0$ and effective stress sensitivity \tilde{n} , and to investigate how the stress/strain-rate are distributed within grains, we make use of two scale transition methods, the mean-field FOSO-SC scheme recently proposed by [1] and the full-field computational homogenization based on the FFT method [3]. All results below have been obtained for uniaxial deformation under a prescribed macroscopic strain-rate $\dot{\bar{\epsilon}}_{eq} = \dot{\gamma}_0$, so that $\bar{\sigma}_{eq} = \tilde{\sigma}_0$.

3.1. Mean-field approach—FOSO-SC scheme

Mean-Field homogenization of heterogeneous materials is relatively straightforward when the mechanical behavior of each mechanical phase (i.e. grains with given orientation) is linear and homogeneous. This is the case for linear thermo-elastic polycrystalline aggregates where the local constitutive relation for a phase *p* reads

$$\boldsymbol{\varepsilon}(\mathbf{x}) = \mathbf{M}^{(p)} : \boldsymbol{\sigma}(\mathbf{x}) + \boldsymbol{\varepsilon}_0^{(p)} \tag{14}$$

where $\mathbf{M}^{(p)}$ and $\boldsymbol{\varepsilon}_{0}^{(p)}$ only depend on the crystal orientation of the considered grain. This leads to an effective behavior of the same form

$$\bar{\boldsymbol{\varepsilon}} = \tilde{\mathbf{M}} : \bar{\boldsymbol{\sigma}} + \tilde{\boldsymbol{\varepsilon}}_0. \tag{15}$$

In that case, to estimate the effective compliance given by

$$\tilde{\mathbf{M}} = \langle \mathbf{M}(\mathbf{x}) : \mathbf{B}(\mathbf{x}) \rangle, \tag{16}$$

with **B** the stress concentration tensor of the purely elastic problem (i.e. when $\varepsilon_0^{(p)} = \mathbf{0}$) defined as $\boldsymbol{\sigma}(\mathbf{x}) = \mathbf{B}(\mathbf{x}) : \bar{\boldsymbol{\sigma}}$, it is sufficient to estimate the phase-average of **B**, denoted $\langle \mathbf{B} \rangle^{(p)}$. Indeed, the volume integral in (16) can be transformed into a discrete sum over all mechanical phases

$$\tilde{\mathbf{M}} = \sum_{p} f_{p} \mathbf{M}^{(p)} : \langle \mathbf{B}(\mathbf{x}) \rangle^{(p)},$$
(17)

with f_p indicating the volume fraction. This is why mean-field homogenization is very efficient numerically.

The situation for non-linear polycrystals as considered here is more complex as the local compliance $\mathbf{M}(\mathbf{x})$ defined after (6) depends on the local stress state which is itself heterogeneously distributed within deforming grains [34]. The solution (17) therefore cannot be used directly. The standard approach to address this issue relies on a linearization of the non-linear polycrystal of interest in order to end up with a thermo-elastic-like material (LCC), exhibiting the same microstructure as the original non-linear one, but for which the standard thermo-elastic homogenized solution applies. Finding the proper linearization procedure is a difficult task, this is why several propositions can be found in the literature (some of them are listed below). The exact link between the effective behavior of the non-linear polycrystal and of the linearized thermo-elastic one is another difficulty that has often been left aside [27]. The impact of the used linearization procedure on the effective behavior or high mechanical contrast between the phases. As seen in the previous section, waldseyite exhibits both very high non-linearity and anisotropy, which constitutes a challenge for mean-field homogenization.

The most advanced linearization procedure nowadays, that we will use here, is the Fully Optimized Second Order (FOSO) scheme proposed recently by [1]. In short, starting from a variational formulation of the problem, the optimal linearization, leading to the definition of the linear thermo-elastic comparison material, is defined as an optimization problem. Compared to the Partially Optimized (POSO) formulation [9], full optimization could be carried out in FOSO, hence the name. This formulation has several significant advantages compared to the previous POSO one: (i) there is no duality gap, i.e. stress and strain-rate formulations yield similar results, (ii) stress and strain-rate field statistics in the linearized and non-linear polycrystals are identical, there is no need to compute the corrective terms as in [27]. As POSO, the FOSO approach complies by construction with the known upper bounds for the effective behavior. First application of FOSO-SC to viscoplastic porous hexagonal polycrystals (similar to sea ice) yields a rheology in very good agreement with the reference results obtained by the FFT full-field homogenization [29]. In the FOSO-SC approach, the local behavior of the linear thermo-elastic comparison polycrystal reads, at the slip system level,

$$\dot{\gamma}_{(s)}(\mathbf{x}) = m_{(s)}^{(p)} \tau_{(s)}(\mathbf{x}) + \dot{e}_{(s)}^{(p)}$$
(18)

where the compliance $m_{(s)}^{(p)}$ and stress-free strain-rate $\dot{e}_{(s)}^{(p)}$ depend on both the first moment $\bar{\tau}_{(s)}^{(p)} = \langle \tau_{(s)}(\mathbf{x}) \rangle^{(p)}$ and second moment $\bar{\bar{\tau}}_{(s)}^{(p)} = \langle \tau_{(s)}^2(\mathbf{x}) \rangle^{(p)}$ of resolved shear stress in the mechanical phase *p*:

$$m_{(s)}^{(p)} = \frac{\dot{\gamma}(\hat{\tau}) - \dot{\gamma}(\hat{\tau})}{\hat{\tau} - \check{\tau}}, \quad \dot{e}_{(s)}^{(p)} = \dot{\gamma}(\check{\tau}) - m_{(s)}^{(p)}\check{\tau}$$
(19)

with $\check{\tau}$ and $\hat{\tau}$ defined as

$$\check{\tau} = \bar{\tau} - \sqrt{\frac{1-\alpha}{\alpha}} \sqrt{\bar{\bar{\tau}} - \bar{\tau}^2} \operatorname{sign}(\bar{\tau}), \quad \hat{\tau} = \bar{\tau} + \sqrt{\frac{1-\alpha}{\alpha}} \sqrt{\bar{\bar{\tau}} - \bar{\tau}^2} \operatorname{sign}(\bar{\tau}).$$
(20)

In (20), $\sqrt{\overline{t}} - \overline{t}^2$ is the standard deviation of the shear stress distribution acting on the slip system. Using the value $\alpha = 0.5$ recommended in [1], \overline{t} and \hat{t} are therefore one standard deviation below and above the mean value \overline{t} .

In comparison, the POSO formulation can be obtained using $\check{\tau} = \bar{\tau}$ and the same definition for $\check{\tau}$ as in (20) while the variational upper bound of [23] corresponds to $\check{\tau} = 0$ and $\hat{\tau} = (\bar{\tau})^{1/2}$. On the other hand, the earlier affine scheme [20], which is not based on a variational formulation, would be obtained by taking the limit of (19) for $\alpha \to 1$, therefore not making use of the shear stress fluctuation at the slip system level. The even earlier tangent formulation of [19] applied to wadsleyite in [18] is based on a similar formulation as the affine one but with a compliance divided by *n* and $\dot{e}_{(e)}^{(p)} = 0$.



Figure 3. Typical periodic microstructure considered for FFT computations.

Application to wadsleyite has been performed considering equiaxe grain shapes and using a set of 2000 crystal orientations generated by a Sobol quasi-random sequence, that provides better overall isotropy than the random orientation usually chosen. As already mentioned, wadsleyite exhibits highly anisotropic and non-linear behavior at the grain scale, and this makes the numerical convergence of the FOSO-SC model rather delicate. The used numerical procedure is detailed in Appendix B. It is worth noting that the model converges without having to introduce any additional unphysical slip systems often used in the literature to reach a total of 5 independent slip systems.

3.2. Full-Field approach—FFT numerical homogenization

The FFT method [3] relies on the 3D microstructure of the considered polycrystal, which constitutes the unit cell, submitted to periodic boundary conditions. This unit cell is discretized into $N_1 \times N_2 \times N_3$ voxels. This discretization determines a regular grid in the cartesian space x_d and a corresponding grid in the Fourier space ξ_d . The heterogeneous problem of a polycrystal exhibiting a different compliance $\mathbf{M}(\mathbf{x})$ at each position (\mathbf{x}) is rewritten equivalently as a homogeneous problem with an arbitrary homogeneous compliance \mathbf{M}^0 and an additional unknown stress-free strain-rate (or polarization) field. The solution is given by a convolution of the Green tensor associated to \mathbf{M}^0 with the polarization field of interest. In the Fourier space, this convolution turns into a direct product, hence the very high numerical efficiency of the method. An iterative scheme must be implemented to obtain, upon convergence, the compatible strain-rate field associated to the balanced stress field for nonlinear rheology, as detailed in [2]. This FFT method provides the "exact" solution (apart purely numerical errors) for the considered microstructure, but requires significantly more computing ressources than mean-field estimations. Another advantage of such full-field homogenization is that details of stress and strain-rate distributions within the microstructure can be obtained.

For the application to wadsleyite, the considered microstructure is a periodical threedimensional unit cells randomly generated by Voronoi tesselation, and containing 1000 grains (Figure 3). Crystal orientations are chosen according to the Sobol sequence introduced in



Figure 4. Macroscopic response $\tilde{\sigma}_0$ of wadsleyite for various temperatures. Predictions of FOSO-SC and FFT polycrystal models including dislocation mobility based on the GSF model are compared to experimental data. The reponse of individual slip on $1/2\langle 111\rangle(101)$ (τ value for $\dot{\gamma} = 10^{-5} \text{ s}^{-1}$ as in Figure 2) as well as the static Lower Bound (LB) are shown for comparison.

Section 3.1, leading to an effective behavior close to isotropic. The unit cell was discretized into $256 \times 256 \times 256$ voxels. The effective rheology has been obtained by averaging the model output for 10 random realizations of such synthetic polycrystalline agregates. The relative statistical uncertainty of results given below has been estimated according to the method proposed in [35]. For example, we found that using 10 random realizations leads to an error of only 0.1% on the effective stress $\tilde{\sigma}_0$.

4. Results and discussion

Results provided by the 3-scales homogenization approach described above, from the nanometer P-T dependent dislocation core structure up to the sub-meter polycrystal scale, is now given for a typical strain-rate corresponding to laboratory mechanical tests. The reference FFT solution and the FOSO-SC estimate integrate the rheology of individual slip systems obtained by the Peierls–Nabarro model described in Section 2.

Figure 4 shows the flow stress $\tilde{\sigma}_0$ for temperatures ranging between 1100 K and 2100 K. First of all, it can be observed that the mean-field homogeization scheme provides an estimation of the effective behavior that lies very close to the reference FFT numerical solution. Both estimations differ by less than 5% for the four temperatures computed by FFT (see numerical values in Table 2 for 1700 K). The FOSO-SC model therefore does an excellent job considering the very strong viscoplastic anisotropy of wadsleyite crystals (recall that axial strain along lattice direction **b** is impossible) and non-linearity ($n \approx 30$ at 1700 K). The effective stress $\tilde{\sigma}_0$ is found to be ~4.7 times larger than the flow stress of $1/2\langle 111\rangle$ (101) for the whole temperature range considered.

Concerning the effective stress sensitivity \tilde{n} , it is normally related with those of individual slip systems in a complex way due to the mechanical interaction between the grains during



Figure 5. Distribution of (left) normalized equivalent stress $\sigma_{eq}/\bar{\sigma}_{eq}$ and (right) normalized equivalent strain-rate $\dot{\epsilon}_{eq}/\dot{\bar{\epsilon}}_{eq}$ in a 2D section of a 3D FFT microstructure for wadsleyite at 1700 K.

Table 2. Effective flow stress $\bar{\sigma}_{eq}$ and standard deviations (SD) of the equivalent stress and strain-rate as predicted by various extensions of the SC scheme at 1700 K

	FFT	STAT	TGT	POSO	FOSO	AFF	VAR	TAYLOR
$ar{\sigma}_{ m eq}$	2497	1232	1699	2198	2611	3128	3232	$+\infty$
$SD(\sigma_{eq})/\bar{\sigma}_{eq}$	1.59	0	1.082	0.856	1.749	2.503	1.678	
$\mathrm{SD}(\dot{\varepsilon}_{\mathrm{eq}})/\dot{\bar{\varepsilon}}_{\mathrm{eq}}$	1.495	2.898	1.238	2.307	1.239	2.869	1.168	0

deformation. The situation for wadsleyite is somehow simplified as both FFT and FOSO-SC approaches predict no activation at all of system [100](010). In [13], it has however been shown for olivine that even when the activity of accommodation mechanisms could be considered as negligeable, their influence on the effective stress sensitivity can be significant. For example, \tilde{n} during the creep of olivine can hardly exceed a value of 2 when dislocation glide with n = 3.5 is accommodated by a linear mechanisms such as Nabarro–Herring or Coble diffusion. Olivine however exhibits only 3 independent slip systems, compared to 4 systems for wadsleyite. Here, slip on [100](010) does not relax any kinematic constraint for grains deforming with the four systems of the $1/2\langle 111\rangle\{101\}$ family. Consequently, it is observed that system [100](010) has no effect on the effective stress sensitivity, and we find numerically that \tilde{n} for wadsleyite is exactly equal to the *n* value of $1/2\langle 111\rangle\{101\}$ slip, already indicated in Figure 2: it lies between 18 and 50 in the considered temperature range.

The fields of normalized equivalent stress $\sigma_{eq}/\bar{\sigma}_{eq}$ and strain-rate $\dot{\epsilon}_{eq}/\dot{\epsilon}_{eq}$ within a section of a 3D microstructure computed by FFT at 1700 K is shown is Figure 5. Large fluctuations are observed between grains (intergranular heterogeneity) but also inside individual grains (intragranular heterogeneity). Hot spots corresponding to high values are clearly visible, they seem to be essentially located at triple junctions and grain boundaries, but note that locations corresponding to large equivalent stresses σ_{eq} do often not exhibit at the same time a large equivalent strain-rate $\dot{\epsilon}_{eq}$. In an attempt to quantify the concentration of stress at grain boundaries, we have computed for each voxel of the FFT microstructures the distance to the nearest grain boundary. We have carefully checked the behavior of many grains, and we show in Figure 6 representative results, for the two grains (#98 and #918) indicated in Figure 5. One can observe that there is a slightly smaller heterogeneity of the equivalent stress at grain interior for grain #918 compared to voxels close to



Figure 6. Effect of the distance (normalized by the grain size) to the closest grain boundary on the normalized equivalent stress for (left) grain #98 and (right) grain #918 indicated in Figure 5. Each dot correspond to a voxel of the grain.



Figure 7. Distribution of normalized equivalent stress $\sigma_{eq}/\bar{\sigma}_{eq}$ in grains #98 and #918 for (left) one microstructure and (right) average over 20 random microstructures. Mean values and standard deviation are also indicated.

grain boundaries, but such trend is not observed in grain #98. The same analysis was performed at the polycrystal level, accounting for all grains and voxels. The results are not shown here for the sake of conciseness, but a similar trend is observed. What can be said is that the largest values of equivalent stresses are observed only within voxels close to grain boundaries, but this concerns only a very small volume fraction of the material. The global picture is similar to that shown in Figure 6 with no significantly larger stress heterogeneity close to grain boundary. A similar analysis was carried out in [36] in the case of cubic polycrystals, and a larger effect of the distance was found, probably due to the fact that the plastic anisotropy at the grain scale is much smaller in cubic materials than for wadsleyite. The authors also mentioned a different behavior for hexagonal polycrystals, but without providing further details. A more detailed analysis of grain boundary effects and their dependence with the anisotropy and non-linearity of the local constitutive relation would be necessary and is left for future work.

To be more quantitative, the distribution of equivalent stress for grains #98 and #918 is shown in Figure 7. Figure 7a shows the stress distribution for these grains within a given random microstructure. One can observe that they are very different, with an intense narrow peak for grain #918 and a broad flat distribution for grain #98. None of these can be described by simple functions such as a log-normal. These are typical features encountered in many grains of the FFT computations. Stress heterogeneities in grains are due to the viscoplastic anisotropy of the grain of interest, related to its crystal orientation, and the mechanical interaction of the grain with its surrounding depending on the behavior of neighboring grains, the grain shape, and the overall polycrystal behavior. To estimate the relative importance of both sources, we have carried



Figure 8. (left) Average values of the local (voxel scale) equivalent stress $\langle \sigma_{\rm eq} \rangle^{(p)} / \bar{\sigma}_{\rm eq}$ in individual grains, and (right) standard deviation of the equivalent stress $\sqrt{\langle \sigma_{\rm eq}^2 \rangle^{(p)} - (\langle \sigma_{\rm eq} \rangle^{(p)})^2} / \bar{\sigma}_{\rm eq}$. Only 50 grains are shown out the 1000 grains of the microstructure.

out FFT computations on 20 microstructures, for which the grains size, shape, and location was random (based on Voronoi tesselation) but keeping the same set of 1000 crystal orientations. Doing so, it was possible to compute the stress distribution in grains #98 and #918 for those 20 realizations, in which those grains keep the same orientation but change size, shape, and are embedded in various environments. Results are shown in Figure 7b. Stress distributions become more similar in shape but still exhibit different mean and standard deviations. This analysis was generalized for all grains of the microstructure, and the corresponding mean and standard deviation of σ_{eq} are shown in Figure 8. In this figure, the red dots correspond to the value obtained for a given random microstructure and the blue dots are the average over 20 microstructures. It is observed that the stress distribution in all the grains is very sensitive to the specific microstructure considered. The stress fluctuation in a given grain due to microstructural effects, indicated by the spread of the red dots, is of similar amplitude than the overall stress fluctuation within the whole polycrystal. In the highly anisotropic and non-linear wadsleyite, the local mechanical state of a grain is thus affected by its crystal orientation but also significantly by the behavior of neighboring grains. A consequence of this is that special care should be taken when interpreting experimental observation of grain deformation. The static bound (STAT in Table 2), assuming a uniform stress within the whole polycrystal and in which grains deformation can be estimated by the sole knowledge of grains orientation (or associated Schmid factor), is therefore not adapted. An experimental illustration of this can be found in [37] in which, for 2D polycrystalline ice (exhibiting only two easy slip systems), no correlation was found between the grain Schmid factor and the local deformation.

Coming back to the efficiency of FOSO-SC, a challenge for mean-field homogenization models is the accurate estimation not only of the effective behavior but also of the spread of the mechanical states at the various scales, so that good results at the polycrystal scale are obtained for good reasons. The distribution of normalized stress and strain-rate over the whole polycrystal, computed with the FFT homogenization using the 20 random realizations, is given in Figure 9. Long tails are observed up to values as high as ~5. We have computed the associated standard deviations as these quantities can be also computed by mean-field models. They are defined as

$$SD(\sigma_{eq}) = \sqrt{\langle \sigma_{eq}^2(\mathbf{x}) - \bar{\sigma}_{eq}^2 \rangle}, \quad SD(\dot{\varepsilon}_{eq}) = \sqrt{\langle \dot{\varepsilon}_{eq}^2(\mathbf{x}) - \dot{\bar{\varepsilon}}_{eq}^2 \rangle}.$$
 (21)

Numerical values are indicated in Table 2. It is found that results obtained with FOSO-SC are in very good agreement with the FFT reference ones, the difference being $\sim 10\%$ for the stress heterogeneity and $\sim 15\%$ for strain-rates. This is an important result as the partially optimized version (POSO) of the SC scheme, which was the best available method since 2002, yields



Figure 9. Probability densities of (left) normalized equivalent stress $\sigma_{eq}/\bar{\sigma}_{eq}$ and (right) normalized equivalent strain-rate $\dot{\epsilon}_{eq}/\dot{\bar{\epsilon}}_{eq}$ predicted by for the whole 3D microstructure (20 random realizations) at 1700 K.

results that are clearly not as good. The variational bound (VAR) of [23] predicts stress and strain-rate heterogeneities that are here relatively close to the FFT reference but, as expected, overestimates the effective stress. Other methods not using the intraphase stress heterogeneities for the definition of the linear comparison polycrystals, i.e. the tangent (TGT) approach of [19] and the affine (AFF) one [20] significantly depart from the FFT results, both for the effective behavior and the field heterogeneities. Finally, the static uniform stress bound (STAT) completely discards stress fluctuations and underestimate the flow stress by a factor of ~2, while the Taylor uniform strain-rate bound (TAYLOR) cannot apply here due to the lack of 5 independent slip systems.

The effective response and associated stress and strain-rate heterogeneities in polycrystalline aggregates lacking 5 independent slip systems have been studied mostly in HCP materials. Hutchinson [33] has shown that in the framework of the *linear* SC scheme, used in this study to compute the linear comparison polycrystal, overall polycrystal deformation is possible with only 4 independent systems. This result has been shown to apply also for non-linear polycrystals by Nebozhyn *et al.* [26]. The same authors also showed that in ionic polycrystals with only 3 independent systems, the variational estimate of [23] predicts a flow stress that is proportional to the square-root of the mechanical contrast between the slip systems, whereas it unrealistically reaches a plateau at high contrasts for the earlier tangent approach of [19]. A very good match between the FOSO-SC approach and FFT reference results has been obtained in [1] in the case of HCP polycrystals at modest non-linearity (n = 3). The present study shows that excellent results are also obtained at a significantly larger non-linearity.

In Figure 4, our numerical results are compared to the available experimental data in the literature [38–41]. These experiments take advantage of the latest developments of experimental deformation at high *P*, *T* conditions. Pressure is created by compressing the sample assembly between two opposed anvils. This assembly contains an internal resistive heater to achieve high temperature conditions. Deformation in torsion is produced by rotating one anvil. Stress measurements are performed by X-ray diffraction using synchrotron radiation by measuring the orientation dependence and changes in lattice spacing for several diffracting planes. To ensure acquisition of a proper diffraction pattern, the grain size must be maintained small. In all these experiments, the grain size was in the range 1–5 μ m. Although the dispersion of this experimental data set is significant, it concludes consistently that wadsleyite is very strong since, despite the high temperature involved (ca. 1500–2000 K), the stress level remains very high: several GPa. Our modeling results are in excellent agreement with these observations. It must be recalled that our multiscale model relies on the strong assumption that strain is produced by dislocation glide only. The Peierls Nabarro model has shown that lattice friction is strongly affected by pressure

in this range, partly due to the strong increase of the elastic constants under pressure. Despite the small grain size, TEM observations [39–41] agree that dislocation activity is pervasive with densities of the order of 10^{13} m⁻² and dislocation configurations which emphasize the key role played by glide under high lattice friction (straight dislocations well-confined in glide planes). In our dislocation based model, glide overcoming high opposed lattice friction leads to a strongly non-linear viscoplastic behavior. Experiments provide limited constraints on stress sensitivity so far, but preliminary estimates from [40, 41] suggest stress exponents in the order of 5–6, i.e. smaller than predicted here. Although this evidence needs to be consolidated, we can tentatively invoke the role played by grain boundaries. As underlined above, the grains sizes in these highpressure experiments need to be maintained in the micrometer range. At the high temperature investigated, it is likely that some accommodation processes operate at grain boundaries. Indeed, evidence for grain boundary sliding and migration was suggested by [39]. In our polycrystalline models, the grain size is not taken into account explicitly, but grain boundaries are present and act only as barriers to dislocations glide, leading to strain localizations which cannot be accommodated by specific grain boundary relaxation mechanisms. It has been demonstrated in [13] that these accommodation mechanisms can very efficiently decrease the stress exponent. This might reconcile our numerical values to the few experimental data available.

To finish with, we would like to come back to possible geophysical implications for in situ mantle deformation, in which strain-rates are many orders of magnitude smaller than the one considered here. Ritterbex *et al.* [6] have provided evidence that dislocation glide controls the mechanical behavior of wadsleyite at lab conditions, and this allows us, in the present paper, to make the link between atomic and grain scales without the need to handle atomic diffusion. In contrast, a recent study of Ritterbex *et al.* [42] handles deformation of wadsleyite at appropriate mantle strain-rates and show that climb-controlled deformation is expected to operate rather than glide-controlled deformation as treated in this work. Therefore, the current results cannot be simply extrapolated to mantle conditions. The present work rather aims to provide a theoretical framework which enables to explain the high stress data of deformation experiments of polycrystalline wadsleyite at lab conditions. The same procedure could however be applied to these new rheological data, but this is left for future work.

5. Conclusion

This work provides the very first estimation of the rheology of a constituent of the Earth's mantle at relevant pressure and temperature in which scales from the atomistic up to polycrystalline aggregate are bridged together using state-of-the-art scale transition models. Here, the dislocation resistance to shear has been computed relying on generalized stacking faults energies incorporation the strong influence of pressure on atomic bonding, combined with Peierls-Nabarro approach. The constitutive equation of a slip system involving these dislocations is then obtained from the Orowan equation. It shows that, at high temperature and strain-rates representative of laboratory experiments (10^{-5} s^{-1}) , the rheology at the slip system level is highly non-linear due to the high lattice friction opposed to dislocation glide. The obtained constitutive relation at the slip system level has been introduced in the recent Fully Optimized Second Order Self-Consistent scheme (FOSO-SC) of Ponte Castañeda [1, 28] which allows computing the effective viscoplastic behavior at the polycrystal scale. Predictions of the latter approach have been compared with the FFT computational homogenization method [3] that provides reference results, in which the same slip system behavior has been introduced. We found that the obtained flow stress of wadsleyite polycrystals at 15 GPa matches very well with the available experimental results from the literature and lies within experimental uncertainties at least for temperatures ranging between 1500 K and 2100 K. This could be obtained thanks to the fast computation provided by the FOSO-SC method. Micromechanical modeling indicates that the stiff slip system [100](010) is not activated at all and has no effect on polycrystal deformation. Wadleyite therefore deforms with only the 4 independent systems of the $1/2\langle 111\rangle$ {101} family. The FFT results show that the deformation of individual grains (mean and standard deviation) in the polycrystalline agregate is significantly influenced by the behavior of neighboring grains. Finally, comparison with earlier mean-field approaches demonstrates the superior estimations provided by the FOSO-SC scheme, not only for the effective behavior but also for the overall stress and strain-rate heterogeneities.

Acknowledgments

This work was supported by the European Research Council under the Seventh Framework Programme (FP 7), ERC (grant number 290424 RheoMan) and under the Horizon 2020 research and innovation programme (grant number 787198 TimeMan).

Appendix A. Determination of the number of independent slip systems

Cotton and Kaufman [43] proposed a method for determining the number of independent slip systems in cubic crystals. Here we extend this method to any lattice symmetry. In that case, the number of independent slip systems not only depends on the Bravais lattice and slip system indexes, but also on the lattice parameters. For example, out of the six individual slip systems of the family $1/2(110){110}$, only two are independent in the case of a cubic crystal lattice, four are independent in the case of an orthorhombic crystal, but five for a tetragonal crystal.

Consider a crystal lattice with lattice vectors **a**, **b** and **c**. The reciprocal lattice vectors \mathbf{a}^* , \mathbf{b}^* and \mathbf{c}^* are given by

$$\mathbf{a}^* = \frac{\mathbf{b} \times \mathbf{c}}{V}, \quad \mathbf{b}^* = \frac{\mathbf{c} \times \mathbf{a}}{V}, \quad \mathbf{c}^* = \frac{\mathbf{a} \times \mathbf{b}}{V},$$
 (22)

where $V = (\mathbf{a}, \mathbf{b}, \mathbf{c})$ is the lattice volume. The vector **n** normal to a plane of indexes (*hkl*) and the vector **b** parallel to the Burgers vector of indexes [*uvw*] are given by

$$\mathbf{n} = h\mathbf{a}^* + k\mathbf{b}^* + l\mathbf{c}^*, \quad \mathbf{b} = u\mathbf{a} + v\mathbf{b} + w\mathbf{c}.$$
(23)

The strain-rate tensor resulting from dislocations glide on (hkl)[uvw] is proportional to the Schmid tensor **S** associated to that system

$$\mathbf{S} = \frac{1}{2} \left(\hat{\mathbf{n}} \otimes \hat{\mathbf{b}} + \hat{\mathbf{b}} \otimes \hat{\mathbf{n}} \right)$$
(24)

where $\hat{\mathbf{n}}$ and $\hat{\mathbf{b}}$ denote the unit vectors parallel to \mathbf{n} and \mathbf{b} , respectively. Tensor \mathbf{S} is symmetric and traceless ($S_{11} + S_{22} + S_{33} = 0$) since plastic deformation due to dislocation glide is isochoric. Therefore, it has only five independent components, S_{11} , S_{22} , S_{23} , S_{13} and S_{12} .

The number of independent slip systems in the crystal is the rank of the matrix [M] containing as many rows as individual slip systems available in the crystal and five columns filled with the five independent components of **S** for each slip system. When decomposing [M] in a row echelon form $[M]_r$, for example using the method rref of the python package sympy, the number of independent slip systems corresponds to the number of nonzero rows (rows with at least one nonzero element) of $[M]_r$.

Appendix B. Numerical method for FOSO-SC

In this section, we present the numerical method used in this work to solve for the FOSO-SC model. It relies on a linearization step to define the LCC, and an inner loop to solve for the LCC.

For the sake of comparison with [1] (their Figure 6), we have computed texture development in an ice polycrystal made of 500 grain orientations deformed under uniaxial compression up to an overall strain of 150% (in 300 steps), accounting for the evolution of grain shape, and a rheology at the grain level with n = 3 and reference shear stresses τ_0 for prismatic and pyramidal slips taken 60 times larger than for basal slip. Computation with the algorithm below (Fortran90 code) lasts 20 min using a standard laptop, compared to 20 h as indicated in [1]. This was obtained using a convergence rate parameter $\kappa = 0.5$ (step 6 below) and an accuracy $err = 10^{-4}$.

For Wadsleyite, the convergence is a little more delicate because of the high non-linearity and grain anisotropy. We had to use values of κ ranging between 0.1 and 0.005 while increasing the stress sensitivity incrementally (steps of $\Delta n \sim 5$).

B.1. Iterative linearization of the non-linear polycrystal

The outer loop of the numerical method works as follows.

- (1) Initial guess: start computing the uniform stress (static) bound, and use this solution as initial guess for the affine SC model, with the same method as described below but using the appropriated linearization (see Section 3.1). Then compute the intraphase first $\langle \boldsymbol{\sigma} \rangle^{(p)}$ and second $\langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(p)}$ moments of the stress field associated to the affine model (see Section B.2).
- (2) With the guess values of ⟨σ⟩^(p) and ⟨σ ⊗ σ⟩^(p), compute the local compliance M^(p) and stress-free strain-rate ε₀^(p) of the LCC, Equation (14), according to (3), (6) and the FOSO linearization (18)–(20).
- (3) Solve for the effective behavior (M̃, ε̃₀) of the LCC, Equation (15), using the method described in Section B.2, and compute the associated new moments (σ)^(p)_{LCC} and (σ ⊗ σ)^(p)_{LCC}.
- (4) Invert (15) to compute the effective stress $\bar{\sigma}$ associated to the prescribed strain-rate $\dot{\bar{\epsilon}}$.
- (5) Computed the following four errors:

$$\begin{aligned} \max_{i,j} |\langle \sigma_{ij} \rangle^{(p)} - \bar{\sigma}_{ij}| / \max_{k,l}(\bar{\sigma}_{kl}), \max_{i,j} |\langle \dot{\varepsilon}_{ij} \rangle^{(p)} - \dot{\varepsilon}_{ij}| / \max_{k,l}(\dot{\varepsilon}_{kl}), \\ \max_{i,j} |\langle \sigma_{ij} \rangle^{(p)}_{\text{LCC}} - \langle \sigma_{ij} \rangle^{(p)}| / \max_{k,l}(\bar{\sigma}_{kl}), \end{aligned}$$

and

$$\max_{i,j,k,l} |\langle \sigma_{ij} \sigma_{kl} \rangle_{\text{LCC}}^{(p)} - \langle \sigma_{ij} \sigma_{kl} \rangle^{(p)} | / \max_{m,n}(\bar{\sigma}_{mn}^2).$$

(6) If the largest of the above errors is larger than *err*, then compute new guesses as $\kappa \langle \boldsymbol{\sigma} \rangle_{\text{LCC}}^{(p)} + (1-\kappa) \langle \boldsymbol{\sigma} \rangle^{(p)}$ and $\kappa \langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle_{\text{LCC}}^{(p)} + (1-\kappa) \langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(p)}$ by mixing the initial guess and the estimation for the LCC, and start again with step 2 above.

B.2. Solving for the thermo-elastic self-consistent LCC

Following e.g. [44] and references therein, for given $\mathbf{M}^{(p)}$ and $\boldsymbol{\varepsilon}_{0}^{(p)}$, the effective compliance of the LCC is given by the implicit equation

$$\tilde{\mathbf{M}} + \mathbf{M}^* = \langle (\mathbf{M}^{(p)} + \mathbf{M}^*)^{-1} \rangle^{-1}$$
(25)

which is solved by a standard fixed-point iterative method. When all grains exhibit the same average shape, the Hill influence tensor \mathbf{M}^* is defined as

$$\mathbf{M}^* = \mathbf{E} : \tilde{\mathbf{M}}, \quad \mathbf{E} = (\mathbf{S}_E^{-1} - \mathbf{I})^{-1}$$
(26)

with I the identity tensor and S_E the Eshelby tensor

$$\mathbf{S}_{E} = \mathbf{P} : \tilde{\mathbf{M}}^{-1}, \quad \mathbf{P} = \frac{1}{2\pi |\mathbf{Z}|} \int_{0}^{\pi} \int_{0}^{\pi} \mathbf{H}(\boldsymbol{\xi}) \|\mathbf{Z}^{-1} \cdot \boldsymbol{\xi}\|^{-3} \sin\theta \, \mathrm{d}\theta \, \mathrm{d}\phi$$
(27)

with $H_{ijkl} = (1/2)(K_{ik}^{-1}\xi_j\xi_l + K_{jk}^{-1}\xi_i\xi_l)$ related to the acoustic tensor $\mathbf{K} = \boldsymbol{\xi} \cdot \tilde{\mathbf{M}}^{-1} \cdot \boldsymbol{\xi}$ and $\boldsymbol{\xi} = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$. **Z** is a symmetrical second order tensor describing the shape of the ellipsoidal inclusion. Here, integration of **P** is performed at each iteration of (25) using a gaussian quadrature method with an increasing number of Gauss points (as in [44]) until an accuracy of *err*/10 is reached. The implicit equation (25) is solved iteratively until a precision of *err*/10. Once $\tilde{\mathbf{M}}$ has been solved, one computes the stress concentration tensor of the purely elastic problem

$$\langle \mathbf{B} \rangle^{(p)} = (\mathbf{M}^{(p)} + \mathbf{M}^*)^{-1} : (\tilde{\mathbf{M}} + \mathbf{M}^*),$$
(28)

the effective stress-free strain-rate

$$\dot{\tilde{\boldsymbol{\varepsilon}}}_0 = \langle \dot{\boldsymbol{\varepsilon}}_0^{(p)} : \mathbf{B} \rangle \tag{29}$$

and the phase-average residual stress

$$\langle \boldsymbol{\sigma}_{\text{res}} \rangle^{(p)} = (\mathbf{M}^{(p)} + \mathbf{M}^*)^{-1} : (\dot{\boldsymbol{\varepsilon}}_0 - \dot{\boldsymbol{\varepsilon}}_0^{(p)}).$$
(30)

The intraphase first moment of the stress field is

$$\langle \boldsymbol{\sigma} \rangle^{(p)} = \langle \mathbf{B} \rangle^{(p)} : \bar{\boldsymbol{\sigma}} + \langle \boldsymbol{\sigma}_{\text{res}} \rangle^{(p)}.$$
(31)

The second moment is given by

$$\langle \sigma_{ij}\sigma_{kl} \rangle^{(p)} = \frac{1}{f_p} \left[(\bar{\boldsymbol{\sigma}} \otimes \bar{\boldsymbol{\sigma}}) :: \frac{\partial \tilde{\mathbf{M}}}{\partial M_{ijkl}^{(p)}} + 2 \frac{\partial \dot{\bar{\boldsymbol{\epsilon}}}_0}{\partial M_{ijkl}^{(p)}} : \bar{\boldsymbol{\sigma}} + \left\langle \dot{\boldsymbol{\epsilon}}_0^{(p)} : \frac{\partial \langle \boldsymbol{\sigma}_{\mathrm{res}} \rangle^{(p)}}{\partial M_{ijkl}^{(p)}} \right\rangle \right]$$
(32)

where expressions for the derivatives can be found e.g. in [44–46]. Integration of the derivative of the Eshelby tensor entering in (32) is done using a gaussian quadrature method.

References

- D. Song, P. Ponte Castañeda, "Fully optimized second-order homogenization estimates for the macroscopic response and texture evolution of low-symmetry viscoplastic polycrystals", *Int. J. Plast.* 110 (2018), p. 272-293.
- [2] P. Suquet, H. Moulinec, O. Castelnau, M. Montagnat, N. Lahellec, F. Grennerat, P. Duval, R. Brenner, "Multiscale modeling of the mechanical behavior of polycrystalline ice under transient creep", *Procedia IUTAM* 3 (2012), p. 64-78.
- [3] H. Moulinec, P. Suquet, "A numerical method for computing the overall response of nonlinear composites with complex microstructure", *Comput. Methods Appl. Mech. Eng.* 157 (1998), p. 69-94.
- [4] C. Denoual, "Modeling dislocation by coupling peierls-nabarro and element free galerkin methods", *Comput. Methods Appl. Mech. Eng.* 96 (2007), p. 1915-1923.
- [5] S. Ritterbex, P. Carrez, K. Gouriet, P. Cordier, "Modeling dislocation glide in MG₂SiO₄ ringwoodite: towards rheology under transition zone conditions", *Phys. Earth Planet. Int.* 248 (2015), p. 20-28.
- [6] S. Ritterbex, P. Carrez, P. Cordier, "Modeling dislocation glide and lattice friction in MG₂SiO₄ waldseyite in conditions of the earth's transition zone", *Am. Mineralogist* **101** (2016), p. 2085-2094.
- [7] P. Gilormini, "A critical evaluation for various nonlinear extensions of the self-consistent model", in *Proc. IUTAM Symp. on Micromechanics of Plasticity and Damage of Multiphase Materials (Sèvres, France)* (A. Pineau, A. Zaoui, eds.), Kluwer Academic Publishers, 1995, p. 67-74.
- [8] P. Gilormini, "Insuffisance de l'extension classique du modèle autocohérent au comportement non linéaire", C. R. Acad. Sci. Paris 320 (1995), no. Ser. IIb, p. 115-122.
- [9] P. Ponte Castañeda, "Second-order homogenization estimates for nonlinear composites incorporating field fluctuations. Part 1: Theory", J. Mech. Phys. Solids 50 (2002), p. 737-757.
- [10] O. Castelnau, D. K. Blackman, R. A. Lebensohn, P. Ponte Castañeda, "Micromechanical modelling of the viscoplastic behavior of olivine", J. Geophys. Res. 113 (2008), article no. B09202.
- [11] O. Castelnau, R. A. Lebensohn, P. Ponte Castañeda, D. K. Blackman, "Earth mantle rheology inferred from homogenization theories", in *Multi-Scale Modeling of Heterogeneous Materials* (O. Cazacu, ed.), John Wiley and Sons, 2008, p. 55-70.

- [12] O. Castelnau, P. Cordier, R. A. Lebensohn, S. Merkel, P. Raterron, "Microstructures and rheology of the earth's upper mantle inferred from a multiscale approach", C. R. Phys. 11 (2010), p. 304-315.
- [13] F. Detrez, O. Castelnau, P. Cordier, S. Merkel, P. Raterron, "Effective viscoplastic behavior of polycrystalline aggregates lacking four independent slip systems inferred from homogenization methods; application to olivine", J. Mech. Phys. Solids 83 (2015), p. 199-220.
- [14] J. P. Montagner, "Deep earth structure upper mantle structure: Global isotropic and anisotropic elastic tomography", in *Treatise on Geophysics* (G. Schubert, ed.), vol. 1, Elsevier, Oxford, 2nd ed., 2015, p. 613-639.
- [15] D. K. Blackman, D. E. Boyce, O. Castelnau, P. R. Dawson, G. Laske, "Effects of crystal preferred orientation on uppermantle flow near plate boundaries: rheologic feedbacks and seismic anisotropy", *Geophys. J. Int.* 210 (2017), no. 3, p. 1481-1493.
- [16] N. M. Ribe, R. Hielsche, O. Castelnau, "An analytical finite-strain parametrization for texture evolution in deforming olivine polycrystals", *Geophys. J. Int.* 216 (2019), p. 486-514.
- [17] O. Castelnau, D. K. Blackman, T. W. Becker, "Numerical simulations of texture development and associated rheological anisotropy in regions of complex mantle flow", *Geophys. Res. Lett.* 36 (2009), article no. L12304.
- [18] A. Tommasi, D. Mainprice, P. Cordier, C. Thoraval, H. Couvy, "Strain-induced seismic anisotropy of wadsleyite polycrystals and flow patterns in the mantle transition zone", J. Geophys. Res. 109 (2004), no. B12, article no. B12405.
- [19] R. A. Lebensohn, C. N. Tomé, "A self-consistent anisotropic approach for the simulation of plastic deformation and texture development of polycrystals: application to zirconium alloys", *Acta Metall. Mater.* 41 (1993), no. 9, p. 2611-2624.
- [20] R. Masson, M. Bornert, P. Suquet, A. Zaoui, "An affine formulation for the prediction of the effective properties of nonlinear composites and polycrystals", J. Mech. Phys. Solids 48 (2000), p. 1203-1226.
- [21] M. I. Idiart, H. Moulinec, P. Ponte Castañeda, P. Suquet, "Macroscopic behavior and field fluctuations in viscoplastic composites: Second-order estimates versus full-field simulations", J. Mech. Phys. Solids 54 (2006), p. 1029-1063.
- [22] R. A. Lebensohn, P. Ponte Castañeda, R. Brenner, O. Castelnau, "Full-field versus homogenization methods to predict microstructure-property relations for polycrystalline materials", in *Chapter 11 of Computational Methods* for Microstructure-Property Relationships (S. Ghosh, D. Dimiduk, eds.), Springer, 2011, p. 393-441.
- [23] P. Ponte Castañeda, "The effective mechanical properties of nonlinear isotropic composites", J. Mech. Phys. Solids 39 (1991), p. 45-71.
- [24] Y. Liu, P. Gilormini, P. Ponte Castañeda, "Variational self-consistent estimates for texture evolution in viscoplastic polycrystals", Acta Mater. 51 (2003), p. 5425-5437.
- [25] Y. Liu, P. Gilormini, P. Ponte Castaneda, "Homogenization estimates for texture evolution in halite", *Tectonophysics* 406 (2003), p. 179-195.
- [26] M. V. Nebozhyn, P. Gilormini, P. Ponte Castañeda, "Variational self-consistent estimates for viscoplastic polycrystals with highly anisotropic grains", C. R. Méc. 328 (2000), no. Ser. IIb, p. 11-17.
- [27] M. Idiart, P. Ponte Castañeda, "Field statistics in nonlinear composites. I. Theory", Proc. R. Soc. Lond. A 463 (2007), p. 183-202.
- [28] P. Ponte Castañeda, "Fully optimized second-order variational estimates for the macroscopic response and field statistics in viscoplastic crystalline composites", Proc. R. Soc. Lond. A 471 (2015), no. 2184, article no. 20150665.
- [29] S. Das, P. Ponte Castañeda, "A multiphase homogenization model for the viscoplastic response of intact sea ice: the effect of porosity and crystallographic texture", *J. Multiscale Comput. Eng.* **17** (2019), p. 121-150.
- [30] A. Metsue, P. Carrez, C. Denoual, D. Mainprice, P. Cordier, "Plastic deformation of wadsleyite: Iv dislocation core modelling based on the peierls-nabarro-galerkin model", *Acta Mater.* 58 (2010), no. 5, p. 1467-1478.
- [31] H. Koizumi, H. O. K. Kirchner, T. Suzuki, "Kink pair nucleation and critical shear stress", Acta Metall. Mater. 41 (1993), p. 3483-3493.
- [32] S. Ritterbex, P. Hirel, P. Carrez, "On low temperature glide of dissociated < 110 > dislocations in strontium titanate", *Philos. Mag.* 98 (2018), no. 15, p. 1397-1411.
- [33] J. W. Hutchinson, "Creep and plasticity of hexagonal polycrystals as related to single crystal slip", *Met. Trans.* 8A (1977), no. 9, p. 1465-1469.
- [34] P. Ponte Castañeda, P. Suquet, "Nonlinear composites", Adv. Appl. Mech. 34 (1998), p. 171-302.
- [35] T. Kanit, S. Forest, I. Galliet, V. Mounoury, D. Jeulin, "Determination of the size of the representative volume element for random composites: statistical and numerical approach", *Int. J. Solids Struct.* 40 (2003), p. 3647-3679.
- [36] G. Cailletaud, S. Forest, D. Jeulin, F. Feyel, I. Galliet, V. Mounoury, S. Quilici, "Some elements of microstructural mechanics", *Comput. Mater. Sci.* 27 (2003), p. 351-374.
- [37] F. Grennerat, M. Montagnat, P. Duval, O. Castelnau nd, P. Vacher, "Intragranular strain field in columnar ice during transient creep", Acta Mater. 60 (2012), no. 8, p. 3655-3666.
- [38] Y. Nishihara, D. Tinker, T. Kawazoe, Y. Xu, Z. Jing, K. N. Matsukage, S.-I. Karato, "Plastic deformation of wadsleyite and olivine at high-pressure and high-temperature using a rotational drickamer apparatus (rda)", *Phys. Earth Planet. Int.* **170** (2008), no. 3, p. 156-169, Frontiers and Grand Challenges in Mineral Physics of the Deep Mantle.

O. Castelnau et al.

- [39] T. Kawazoe, S.-I. Karato, J. Ando, Z. Jing, K. Otsukaand, J. W. Hustoft, "Shear deformation of polycrystalline wadsleyite up to 2100 k at 14–17 gpa using a rotational drickamer apparatus (rda)", *J. Geophys. Res.* **115** (2010), p. 1-11.
- [40] J. Hustoft, G. Amulele, J.-I. Ando, K. Otsuka, Z. Du, Z. Jin, S.-I. Karato, "Plastic deformation experiments to high strain on mantle transition zoneminerals wadsleyite and ringwoodite in the rotational drickamer apparatus", *Earth Planet. Sci. Lett.* 361 (2013), p. 7-15.
- [41] R. Farla, G. Amulele, J. Girard, N. Miyajima, S.-I. Karato, "High-pressure and high-temperature deformation experiments on polycrystalline wadsleyite using the rotational drickamer apparatus", *Phys. Chem. Miner.* 42 (2015), p. 541-558.
- [42] S. Ritterbex, P. Carrez, P. Cordier, "Deformation across the mantle transition zone: A theoretical mineral physics view", *Earth Planet. Sci. Lett.* 547 (2020), article no. 116438.
- [43] J. D. Cotton, M. J. Kaufman, "A simplified method for determining the number of independent slip systems in crystals", *Scripta Metal. Mater.* 25 (1991), p. 2395-2398.
- [44] R. Brenner, O. Castelnau, L. Badea, "Mechanical field fluctuations in polycrystals estimated by homogenization techniques", *Proc. R. Soc. Lond. A* **460** (2004), no. 2052, p. 3589-3612.
- [45] R. A. Lebensohn, C. N. Tomé, P. Ponte Castañeda, "Self-consistent modeling of the mechanical behavior of viscoplastic polycrystals incorporating field fluctuations", *Philos. Mag.* 87 (2007), no. 28, p. 4287-4322.
- [46] R. Brenner, R. L. Lebensohn, O. Castelnau, "Elastic anisotropy and yield surface estimates", Int. J. Solids Struct. 46 (2009), p. 3018-3026.



Contributions in mechanics of materials

Efficient simulation of single and poly-crystal plasticity based on the pencil glide mechanism

Lu Tuan Le^{*a*}, Kais Ammar^{*a*} and Samuel Forest^{*, *a*}

 a MINES Paris Tech, PSL University, Centre des matériaux (CMAT), CNRS UMR 7633, BP 87 91003 Evry, France

E-mails: lutuan.le@gmail.com (L. T. Le), kais.ammar@mines-paristech.fr (K. Ammar), samuel.forest@mines-paristech.fr (S. Forest)

Abstract. The present work demonstrates that the pencil glide mechanism is a physically reliable and a computationally efficient model to simulate the nonlinear behaviour of b.c.c. single and polycrystals. For that purpose, the pencil glide extension of Schmid's criterion used by Gilormini [1] is incorporated in a single crystal model and in a homogenized polycrystal model accounting for large elastoviscoplastic deformations. The response of the pencil glide model in terms of stress-strain curves and lattice rotation is compared to the prediction based on the consideration of all ($\{110\}\langle111\rangle + \{112\}\langle111\rangle$) slip systems. In the case of α -iron single crystals both approaches are shown to accurately reproduce recent experimental results [2, 3]. The comparison is extended to α -iron polycrystals behaviour under tension, compression, rolling and simple shear loading conditions. The evolution of crystallographic textures obtained either based on pencil glide or using the 24 slip systems is analyzed and compared to classical experimental results from the literature. Limitations of the approach, especially in the case of simple shear textures, are also pointed out. The pencil glide approach can be viewed as a reduced order model enhancing computational efficiency of crystal plasticity simulations involving many slip mechanisms.

Keywords. Crystal plasticity, Pencil glide, Single crystal, Polycrystal, b.c.c. crystal, Homogenization, Texture.

Manuscript received 10th June 2020, accepted 30th July 2020.

1. Introduction

Many crystals with b.c.c. (body centered cubic) structure exhibit a specific plastic behaviour at low temperature characterized by the difficulty of identifying the slip planes along which dislocations are gliding, whereas the slip direction (given by the Burgers vector) is clearly defined. The reason is that screw dislocations can easily cross-slip on several planes containing each slip

^{*} Corresponding author.

direction which is a zone axis of the crystal. Slip lines are observed to be sinuous and this phenomenon was called *pencil glide* and *non-crystallographic slip* by Taylor and Elam [4]. Modifications of the well-known Schmid law for crystal plasticity were proposed to accommodate pencil glide and used to predict yield surfaces of polycrystals [5]. Gilormini [1] and Becker [6] applied such a theory to predict rolling textures based on a homogenization polycrystal approach. According to the pencil glide model used in [1], the Schmid law is applied to slip systems with fixed crystallographic slip directions and optimized slip plane to maximize the resolved shear stress at each strain increment. Rolling textures were predicted in [1] using the Taylor model [7] and the relaxed constraint model [8]. It was shown that the results of the latter model were in better agreement with experimental findings [1]. However, these two homogenization techniques are known to provide bounds of the actual behaviour. From a computational perspective, an advantage of the pencil glide model is to reduce the number of slip systems and associated internal variables and corresponding material parameters. This is the main incentive of the present contribution.

Hill [9], Mecking [10, 11], Kocks [12], van Houtte [13] and Arminjon [14, 15] have proposed several classes of polycrystal models allowing for non homogeneous stress and strain values between the various orientation classes of grains. These models essentially lead to deformation textures in good agreement with measured ones. Depending on specific weighting parameters present in these models, they can account for the whole span of the solution domain between the lower and upper bounds. Another class of non-homogeneous models which make use of the Eshelby theory to estimate the local tress and strain is the viscoplastic self-consistent scheme (VPSC) [16, 17]. All grains having the same orientation within a given precision are gathered in a single monocrystalline inclusion embedded in the effective medium. This reflects explicitly some nonlinear interaction of each grain with its homogenized surroundings [18]. For the quasi-rate-independent case, there also exists a self-consistent polycrystal plasticity model relying on the multiplicative decomposition of the deformation gradient, see [19].

In the present work, a fast computational homogenization polycrystal model is introduced, the so-called β -model which is a tunable extension of the Standard Self-Consistent (SSC) scheme to investigate the elasto-plasticity of polycrystals [20–25]. The grain/aggregate interaction is still taken into account via the use of the Eshelby tensor. The β -model incorporates interphase accommodation variables $\beta^{(g)}$ to replace the local plastic strain $\varepsilon_p^{(g)}$ in usual SSC localization formula. The evolution law for the β -variables contains tunable transition parameters to be calibrated from full-field simulations of polycrystalline aggregates [26]. The β -model formulation from [23] is extended in the present work to allow for finite deformations using the concept of local objective frames [21]. The evolution texture in b.c.c. metals is simulated using the β -model. In the simulations presented in this work, we consider either the restricted glide ({110}{111} + {112}{111}) or the pencil glide (111) as the plastic slip modes. The responses of these mechanisms will be compared for single and poly-crystals for various loading conditions in order to assess the ability of the pencil glide model to mimick the response of the full slip model.

The article is organized as follows. First, the finite deformation framework and the pencil glide model are depicted. The polycrystal homogenization β -model is then presented in the case of large deformations. The third section is dedicated to the comparison of the responses of the pencil glide model and the consideration of all ({110}{111} + {112}{111}) slip systems in the case of single crystals in tension and compression. In the case of polycrystals treated in Section 4, it is first necessary to calibrate the free parameters of the β -model from a full field simulation of polycrystalline aggregates. Polycrystal simulations are then performed in the case of tension, compression, torsion and simple shear loading conditions, considering either pencil glide or the 24 slip systems. Results are compared in terms of overall stress-strain curves, lattice rotation and evolution of texture components. Computation time of simulations for 1000 grains is also an important information provided in this work.
In the following paper, an orientation is given by the three Euler angles $\varphi_1, \Phi, \varphi_2$ where the Bunge notation is used. The distribution of orientations, the pole figures and the ODF (Orientation Distribution Functions) maps are obtained by means of the ATEX software (http://www.atex-software.eu/).

2. Model description

2.1. Single crystal plasticity at large deformations

The formulation of the present single crystal plasticity model makes use of local objective frames to develop constitutive models at finite strains [27]. It was proposed in [21] for single and polycrystals and used recently in [28]. It departs from Mandel's classical formulation relying on the multiplicative decomposition of the deformation gradient **F** [29]. The velocity gradient **L** is decomposed into its symmetric part {**L**} defining the strain rate tensor, and its skew-symmetric part }**L**{, called spin tensor. The time-dependent rotation ${}^{c}\mathbf{Q}$ linking the corotational space frame to the current one is defined as

$$\mathbf{c}\dot{\mathbf{Q}}^{\mathbf{c}}\mathbf{Q}^{T} = \mathbf{L}\{\text{ and } \mathbf{c}\mathbf{Q}(\mathbf{t}=\mathbf{0})=\mathbf{1}.$$
 (1)

The strain rate tensor is pulled back to the corotational frame and split into elastic and viscoplastic parts:

$$^{\mathbf{c}}\mathbf{D} = ^{\mathbf{c}}\mathbf{Q}^{T}\{\mathbf{L}\}^{\mathbf{c}}\mathbf{Q} = \dot{\boldsymbol{e}}^{\boldsymbol{e}} + \dot{\boldsymbol{e}}^{\boldsymbol{p}}.$$
(2)

The Cauchy stress tensor σ and the corotational stress **S** are computed from the following elastic law:

$$\mathbf{S} = \mathbb{C} : \boldsymbol{e}^{\boldsymbol{e}}, \quad \text{with } \mathbf{S} = \det(\mathbf{F})^{\mathbf{c}} \mathbf{Q}^{T} \boldsymbol{\sigma}^{\mathbf{c}} \mathbf{Q}$$
(3)

where $\mathbb C$ denotes the fourth order tensor of elastic moduli.

Plastic deformation is the result of the contribution of all gliding slip systems of the crystal:

$$\dot{\boldsymbol{e}}^{\boldsymbol{p}} = \sum_{\boldsymbol{s}\in S} \dot{\boldsymbol{\gamma}}^{\boldsymbol{s}} \{^{\mathbf{c}} \mathbf{m}^{\boldsymbol{s}} \otimes {}^{\mathbf{c}} \mathbf{n}^{\boldsymbol{s}}\}$$
(4)

where ${}^{c}\mathbf{m}^{s}$ and ${}^{c}\mathbf{n}^{s}$ respectively are the slip direction and the normal to the slip plane for slip system *s* in the corotational space frame, γ^{s} being the associated slip amount.

The link between the lattice space frame and the corotational space frame is the rotation ${}^{\#}\mathbf{Q}$ defined as

$${}^{\#}\dot{\mathbf{Q}}^{\#}\mathbf{Q}^{T} = \sum_{s \in S} \dot{\gamma}^{s} {}^{s} {}^{\mathbf{c}}\mathbf{m}^{s} \otimes {}^{\mathbf{c}}\mathbf{n}^{s} {}^{s} {}^{a} {}^{a}\mathbf{n} {}^{d} {}^{\#}\mathbf{Q}(\mathbf{t}=\mathbf{0}) = \mathbf{Q}_{\mathbf{0}} {}^{(5)}$$

whose initial value depends on the initial crystal orientation. Crystallographic directions are known in the lattice frame: ${}^{c}\mathbf{m}^{s} = {}^{\#}\mathbf{Q}\mathbf{m}^{s}$ and ${}^{c}\mathbf{n}^{s} = {}^{\#}\mathbf{Q}\mathbf{n}^{s}$. The rotation is calculed by using the exponential map described in Appendix A.

2.2. Pencil glide

The directions along which slip can occur in b.c.c crystals are $\langle 111 \rangle$ with associated unit vectors \mathbf{m}^k , k = 1, 2, 3, 4. Slip under pencil glide conditions can occur on any plane with normal \mathbf{n}^k which is parallel to one given \mathbf{m}^k vector. The driving force for plastic slip along the slip system is the resolved shear stress τ^k computed as

$$\tau^{k} = (\mathbf{Sn}^{k}) \cdot \mathbf{m}^{k} = (\mathbf{Sm}^{k}) \cdot \mathbf{n}^{k}.$$
(6)

According to the pencil glide theory [5, 30, 31], the most probable active slip plane is the one(s) maximizing the value of the resolved shear stress. This optimization procedure provides the following formula, valid for non-vanishing shear stress:

$$\mathbf{n}^{k} = \frac{\mathbf{m}^{k} \times (\mathbf{S}\mathbf{m}^{k}) \times \mathbf{m}^{k}}{\tau^{k}}$$
(7)

which implies that

$$\tau^{k} = \|\mathbf{m}^{k} \times (\mathbf{S}\mathbf{m}^{k})\|.$$
(8)

2.3. Hardening mechanisms

The plastic slip rate $\dot{\gamma}$ for a pencil glide slip system *s* is expressed by a rate-dependent phenomenological flow rule. The following classical form, taken for example from [32], is adopted:

$$\dot{\gamma}^{s} = \left\langle \frac{|\tau^{s}| - r^{s}}{K} \right\rangle^{n} \operatorname{sign}(\tau^{s}).$$
(9)

The material parameters *K* and *n* characterize the viscosity, and *r*^s is the critical resolved shear stress including the following form of isotropic hardening, according to [33]:

$$r^{s} = r_{0} + Q \sum_{l} h^{sl} (1 - \exp(-bv^{l})), \quad \text{with } \dot{v}^{l} = |\dot{\gamma}^{l}|.$$
(10)

Here, r_0 denotes the initial critical resolved shear stress, Q, b are material hardening parameters, h^{sl} is the interaction matrix which represents self-hardening and latent hardening. It must be noted that, due to the fact that slip plane is likely to change at each load increment, the slip variable integrated from (9) has no real crystallographic meaning and rather is a nonholonomic cumulative slip variable depending on the path.

2.4. Polycrystal homogenization: the β -model

Gilormini [1] has used the pencil glide mechanism to predict the rolling textures in b.c.c. polycrystals with the Taylor model. The Taylor model provides qualitative agreement with some experimental results. However other polycrystal homogenization schemes are eligible for the use of the pencil glide model. In the present work, a generic homogenization model for polycrystals is applied, namely the β -model which can be regarded as a class of reduced order models [20, 23–25, 32, 34, 35]. The approach is based on the introduction of an interphase accommodation variable $\boldsymbol{\beta}^{(g)}$ related to the mean plastic strain $\boldsymbol{e}_p^{(g)}$ in each grain orientation number g. Still using (1) to (3), the stress tensor in the corotational frame is computed by means of the effective (homogenized) tensor of elastic moduli:

$$\mathbf{S} = \mathbb{C}_{\text{Homo}} : \boldsymbol{e}^{\boldsymbol{e}}.$$
 (11)

The self-consistent homogenization model is used to compute the effective moduli \mathbb{C}_{Homo} as the limit of the series:

$$\mathbb{C}_{\text{Homo}}(n) = \sum_{g} f_{(g)} \mathbb{C}^{(g)} : [\mathbb{I} - \mathbb{S}_{\text{Esh}}(n-1) : (\mathbb{I} - \mathbb{C}_{\text{Homo}}^{-1}(n-1) : \mathbb{C}^{(g)})]$$
(12)

with $\mathbb{C}_{\text{Homo}}(0) = \sum_{g} f_{(g)} \mathbb{C}^{(g)}$, $f_{(g)}$ being the volume fraction of grain orientation *g*. The equations to compute \mathbb{S}_{Esh} are detailed in Appendix B.

The following scale transition rule is adopted to compute the mean stress for each grain orientation *g*:

$$\mathbf{S}^{(g)} = \mathbb{B}^{(g)} : (\mathbf{S} + \mathbb{L}_C^* : (\boldsymbol{\beta} - \boldsymbol{\beta}^{(g)}))$$
(13)

where $\mathbb{B}^{(g)}$ is the localization tensor defined as

$$\mathbb{B}^{(g)} = \mathbb{C}^{(g)} : [\mathbb{I} + \mathbb{S}_{\text{Esh}} : \mathbb{C}_{\text{Homo}}^{-1} : (\mathbb{C}^{(g)} - \mathbb{C}_{\text{Homo}})]^{-1} : \mathbb{C}_{\text{Homo}}^{-1}.$$
(14)

In the latter expression, \mathbb{I} is the fourth order identity tensor w.r.t. to symmetric second rank tensors, \mathbb{S}_{Esh} is the Eshelby tensor, $\mathbb{C}^{(g)}$ is the elastic stiffness tensor for grain number *g* and

$$\mathbb{L}_{C}^{*} = \mathbb{C}_{\text{Homo}} : (\mathbb{I} - \mathbb{S}_{\text{Esh}}).$$
(15)

The $\boldsymbol{\beta}^{(g)}$ variables are obtained by integrating time-independent nonlinear evolution equations reminiscent of the nonlinear kinematic hardening rule:

$$\dot{\boldsymbol{\beta}}^{(g)} = \dot{\boldsymbol{e}}_p^{(g)} - \mathbb{D} : \boldsymbol{\beta}^{(g)} \| \dot{\boldsymbol{e}}_p^{(g)} \|.$$
(16)

These variables are initialized at 0 in the present work. The plastic strain rate for each grain orientation *g* is computed as

$$\dot{\boldsymbol{e}}_{\boldsymbol{p}}^{(g)} = \sum_{s \in S} \dot{\boldsymbol{\gamma}}_{s}^{(g)} \{^{\mathbf{c}} \mathbf{m}_{s}^{(g)} \otimes {^{\mathbf{c}}} \mathbf{n}_{s}^{(g)} \}.$$
(17)

The Equation (16) involves a constant phenomenological tensor \mathbb{D} whose components must be calibrated. They are determined by comparing the model responses between β -model and a reference full-field FEM simulation of a polycrystalline aggregate following the strategy depicted in [36]. The effective accommodation variable β is given by [25, 37]:

$$\boldsymbol{\beta} = \sum_{g} f_{(g)} \mathbb{L}_{C}^{*-1} : \mathbb{B}^{(g)} : \mathbb{L}_{C}^{*} : \boldsymbol{\beta}^{(g)}.$$
(18)

The macroscopic plastic strain e^p generally deviates from the average plastic strain over all grain orientations by the following contribution:

$$\mathbf{e}^{p} = \sum_{g} f_{(g)} \mathbf{e}_{\mathbf{p}}^{(\mathbf{g})} + \sum_{g} f_{(g)} [\mathbb{C}^{(g)-1} : \mathbb{B}^{(g)} : \mathbb{L}_{C}^{*} : (\boldsymbol{\beta} - \boldsymbol{\beta}^{(g)})].$$
(19)

Depending on the choice of \mathbb{D} , the β -model can reproduce the response of the Kröner model or other self-consistent schemes better accounting for elasto-plastic accommodation of the grains than the usual Taylor model [18].

3. Assessment of the pencil glide model for single crystals

3.1. Validation: comparison with Mandel's original scheme

The formulation of crystal plasticity using two local objective frames will be compared with Mandel's model [29] which is based on the multiplicative elastic-plastic decomposition of the deformation gradient. The comparison is made for simple shear $\mathbf{F} = \mathbf{1} + \gamma \mathbf{e}_1 \otimes \mathbf{e}_2$, with prescribed overall shear γ . According to Mandel's theory, the Mandel stress tensor is used to compute the resolved shear stress on each slip system whereas the present theory makes use of the corotational stress. Due to the fact that elastic strains in metals generally remain small, no significant discrepancy is expected. The initial orientation of the crystal is $\mathbf{e}_1 = [100]$ and $\mathbf{e}_2 = [010]$. These simulations are conducted in the absence of strain hardening with a constant critical resoved shear stress r_0 , and considering 12 slip systems $\{110\}\langle 111\rangle$. It can be checked from Figure 1 that the Cauchy stress components predicted by the Mandel and corotational models are identical.

The "fish-like" oscillations of the stresses observed in Figure 1 for this orientation of a f.c.c. single crystal under simple shear were discussed in [21, 38]. It is of interest to consider the response of b.c.c. crystals to the same loading conditions. Figure 2 provides three Cauchy stress components for three slip system families, namely 12 slip systems $\{110\} < \{111\}, 24$ slip systems $\{110\} + \{112\} + \{123\}$ or 4 pencil glide mechanisms. We have found that the response of 12 b.c.c. slip systems, see Figure 1, is the same as in the f.c.c. case. The consideration of 24 slip systems leads to a similar periodic response with slightly lower stress values. The lower envelope of these curves is obtained using the pencil glide model. In all cases, the rotation rate of the lattice is $-\dot{\gamma}/2$, i.e. the



Figure 1. Simple glide test for a single crystal endowed with 12 slip systems $\{110\}\langle 111\rangle$: stress responses of the Mandel and corotational single crystal models.



Figure 2. Simple glide test for a b.c.c. single crystal endowed with 12 slip systems $\{110\}\langle 111\rangle$, 24 slip systems $\{110\} + \{112\} + \{123\}$ or pencil glide: stress components (MPa) computed using the single crystal model based on local objective frames, with $r_0 = 50$ MPa, in the absence of hardening.

spin of the corotational frame. This is a remarkable feature of simple shear for ideally oriented single crystals.

3.2. Tension of α -iron single crystals

The relevance of the Schmid law is well-known for many f.c.c. crystals deforming by slip on $\{111\}$ planes at low and intermediate temperatures. In the case of b.c.c. metals, in particular α -iron at room temperature, the situation is more complicated since multiple slip planes are available, namely $\{110\}$ and $\{112\}$, see Table 1. Several experiments [39, 40] have highlighted the fact that thermal effects are still at play. Slip planes $\{123\}$ are also possible for the accommodation of plastic strain [41]. Screw dislocations gliding in $\{110\}$ slip planes and all dislocations gliding

Notation	Plane	b	Notation	Plane	b
2A	(011)	[Ī11]	6'A	(112)T	[Ī11]
3A	(101)	[111]	2″A	(211)AT	[111]
6A	(110)	[111]	3'A	(121)T	[111]
2B	(011)	[111]	4''B	(121)AT	[111]
5B	(110)	[111]	2′B	(211)T	[111]
4B	(101)	[111]	5″B	(112)AT	[111]
1C	(011)	[11]	3″C	(Ī21)AT	[11]
5C	(110)	[11]	5'C	(112)T	[11]
3C	(101)	[111]	1″C	(211)AT	[111]
1D	(011)	[111]	6′D	(Ī12)T	[111]
4D	(101)	[111]	1″D	(211)AT	[111]
6D	(110)	[111]	4′D	(121)T	[111]

Table 1. The slip planes in b.c.c. crystals, with notations from [45, 46]

Table 2. Chemical elements in ARMCO® commercial iron (% weight), according to [3]

С	Мо	Si	Р	S	Ti	Cr	Mn	Cu	Ni	Al	Ν	Va	Sn	Pb
0.0026	0.001	0.004	0.002	0.0047		0.016	0.080	0.009	0.011	0.002	0.006		0.002	

in {112} planes experience an energy barrier to overcome lattice friction [42, 43]. Besides, crossslip in iron is easier owing to the dislocation compact core in b.c.c. crystals [44]. The existence of multiple cross-slip planes leads to the formation of wavy slip lines characterizing the pencil glide.

The objective of this section is to compare the description of the tensile behaviour of α -iron single crystals by crystal plasticity models involving either all slip systems of Table 1 or only 4 pencil glide systems. The results of experiments with ARMCO® iron that contains impurities reported in Table 2 are taken from [3]. The dimensions of the matrix h^{sl} for b.c.c. materials is usually 12 × 12 considering only one family of slip systems, but in the case of α -iron, the dimensions is 24 × 24. The difficulty is that the interaction matrix required at least 17 independent coefficients, as summarized in Table 3. Some of them could be predicted by Dislocation Dynamics simulations [47–49] but many are essentially unknown. Values taken from the works [46, 50, 51] are used in the sequel.

In contrast, the pencil glide model requires a 4 × 4 interaction matrix and only two parameters must be given to define this matrix, see Table 4. Calibration of these parameters is performed in order to obtain closest agreement with experimental curves.

The material parameters of both models are reported in Table 5. The power law value n = 20 and low *K* value ensure rate–independence of the results for the considered range of strain rates.

Initially, the crystallographic axes (X_1 , X_2 , X_3) of the samples coincide with the axes of the sample frame, labelled N, S and L to specify the directions normal to the sample observed surface, the sample side face and the sample loaded face, respectively. Tensile loading is applied along axis X_3 (parallel with L). During the test, the axial material fibers remain parallel to the loading axis, which means that $F_{13} = F_{23} = 0$. In compression along axis X_3 , material planes normal to X_3 are assumed to remain parallel, leading to the conditions $F_{31} = F_{32} = 0$. A third condition is that the rotation with respect to axis X_3 vanishes, which leads to $F_{12} = F_{21}$ for both tension and compression.

Table 3. The interaction matrix in b.c.c. single crystals using $\{110\} + \{112\}$ slip planes and the associated parameter values used for α -iron. The notations and parameters values are taken from [46, 50]

	2A	3A	6A	2B	4B	5B	1C	3C	5C	1D	4D	6D	2'A	3'A	6'A	2"B	4'B	5'B	1'C	3"C	5"C	1"D	4"D	6"D
2A	DP	Ω_1	Ω_1	CP	B1	B1	Α	B1	С	Α	С	B1	Ω_2	Ω_3	Ω_3	B2	B3	B3	D	Е	F	D	F	Е
3A		DP	Ω_1	B 1	Α	С	B1	CP	B1	С	Α	B1		Ω_2	Ω_3	E	D	F	B3	B2	B3	F	D	Е
6A			DP	B1	C	Α	С	B1	Α	B1	B1	CP			Ω_2	Е	F	D	F	Е	D	B3	B3	B2
2B				DP	Ω_1	Ω_1	Α	С	B1	Α	B1	С				Ω_2	Ω_3	Ω_3	D	F	Е	D	Е	F
4B					DP	Ω_1	С	Α	B1	B1	CP	B1					Ω_2	Ω_3	F	D	Е	B3	B2	B3
5B						DP	B1	B1	CP	С	B1	А						Ω_2	B3	B3	B2	F	Е	D
1C							DP	Ω_1	Ω_1	CP	B1	B1							Ω_2	Ω_3	Ω_3	B2	B3	B3
3C								DP	Ω_1	B1	Α	С								Ω_2	Ω_3	Е	D	F
5C									DP	B1	C	Α									Ω_2	Е	F	D
1D										DP	Ω_1	Ω_1										Ω_2	Ω_3	Ω_3
4D											DP	Ω_1											Ω_2	Ω_3
6D												DP												Ω_2
2'A													DP	Ω_1	Ω_1	G	I	I	Н	Ι	J	Н	J	I
3'A														DP	Ω_1	I	Н	J	Ι	G	Ι	J	Н	Ι
6'A															DP	I	J	Н	J	Ι	Н	Ι	Ι	G
2"B																DP	Ω_1	Ω_1	Н	J	Ι	Н	Ι	J
4'B																	DP	Ω_1	J	Н	I	Ι	G	I
5'B																		DP	I	I	G	J	I	Н
1°C																			DP	Ω_1	Ω_1	G	Ι	Ι
3"C																				DP	Ω_1	Ι	Н	J
5"C																					DP	I	J	Н
1"D																						DP	Ω_1	Ω_1
4"D																							DP	Ω_1
6"D																								DP
																	_						_	
DP =	= 0.07	CP	P = 0.07	7 Ω	= 0.7	B1	= 0.0	$8 \mid A$	= 0.1	$4 \mid C$	= 0.0'	$7 \mid B$	2 = B3	B = 0.1	E	= 0.08	D =	= F = (0.11	G = I	H = J	= 0.09	I =	0.08

Table 4. The interaction matrix in b.c.c. crystals using pencil glide, where H1 = 0.07 and H2 = 0.09 as found in the present work

	[111]	[Ī11]	[111]	[111]
[111]	H1	H2	H2	H2
[111]		H1	H2	H2
[111]			H1	H2
[111]				H1

Table 5. Material parameters used for both pencil glide and $\{110\}\langle 111\rangle + \{112\}\langle 111\rangle$ single crystal models

$C_{11} = 284 \text{ GPa}$	$C_{12} = 149 \text{ GPa}$	$C_{44} = 105 \text{ GPa}$
$r_0 = 27 \text{ MPa}$	Q = 1285 MPa	b = 1.5
<i>n</i> = 20	$K = 5 \text{ MPa}^{1/n}$	

Simulations are presented for 5 distinct single crystal orientations:

- *Orientation 1 (Figure 3a):* For this orientation in the centre of the reference triangle, the primary active slip system leads to a rotation of the L axis towards the [111] slip direction without activation of the secondary (101)[111] slip system.
- *Orientation 2 (Figure 3b):* Although the initial orientation for this case is close to a boundary [001] [011], and although the secondary traces do not differ much from those of the primary system, the measured rotation of the L axis towards the [111] slip direction provides unambiguous evidence of single slip even in the overshoot regime.



Figure 3. Rotations of the three axes of the sample measured by EBSD (left), and simulated rotations of the three sample axes: (middle) $\{110\} + \{112\}$ slip systems, (right) pencil glide. The initial orientation is marked by a square symbol.

This rotation of the L axis indicates that the primary system is $(\overline{1}01)[111]$, which has initially a slightly higher Schmid factor. The reported measured rotations of the N and S axes also correspond with the simulated ones.

- Orientation 3 (Figure 3c): This orientation lies on the boundary $[001] [\bar{1}11]$: the two equally loaded $\{110\}$ systems were found to activate in spatially separated parts of the sample with dense and straight slip lines. Then one of the two systems rapidly takes over and single slip mainly takes place for the rest of the strain path in most of the sample volume, as assessed by the lattice rotation. The lattice rotation predicted by the pencil glide model displays some discrepancy with experimental results. The reason is that it allows here for only one slip system. The interaction between 2 $\{110\}$ systems is a junction, and the corresponding coefficient in the interaction matrix has the same value as the self-hardening, however these coefficients are different when using the pencil glide model.
- Orientation 4 (Figure 3d): This orientation is situated close to the corner of the domain where the (112)[111] system has the highest Schmid factor. At the corner, two other slip systems of the {110} type have equal Schmid factors. The rotation of the L axis was found to be towards the [111] slip direction, which confirmed the activation of the (112)[111] slip system, with good agreement between both models.
- *Orientation 5 (Figure 3e):* The highest Schmid factor for this orientation is for the (211)[111] system, in competition with the collinear (101)[111] slip system. However, the Schmid factor ratio is initially 5% higher in favour of the {112} system type, with good agreement between both models.

Figure 4a shows the experimental stress-train curves for α -iron at ambient temperature. It can be seen that the hardening modulus for single slip systems {110} and {112} are different. The tensile tests performed have also reported the dissymmetric occurrence of {112} slip for orientations near the boundaries of the standard triangle (near direction [001] for twinning direction, and near direction [011] for anti-twinning direction). The value of the hardening modulus for the systems {112}AT (activated in antitwinning direction) is greater than for the systems {110}, and the hardening modulus for the systems {110}. When multiple slip systems are active, the hardening moduli are larger than for single slip even if the activation of multiple slip systems is not stable.

Corresponding tensile curves for the full single crystal and the reduced pencil glide models are provided in Figure 4b. It is found that the pencil glide model predicts the tensile response of α iron as accurately as classical crystal plasticity with 24 slip systems. There is little difference even if the same parameters are used for both models (see Table 5) except the interaction matrix which differs for both models (see Tables 3 and 4 respectively). The simulated curves are essentially in good agreement with experimental results. Lattice rotation is also well-predicted in general. Some discrepancies can be observed in the stress-train curves that can be attributed to the fact that we did not account for the dissymmetric activation of slip systems {112}. This can be done but it is out of the scope of the present paper.

4. Assessment of the pencil glide model for α -iron polycrystals

4.1. Identification of scale bridging parameters

In the β -model, \mathbb{D} denotes a fourth order phenomenological constant tensor to be calibrated. Its components are identified by comparing the model responses between the β -model and



Figure 4. True stress–true strain curves for 6 single crystal orientations: (a) experimental results, (b) simulation results: $\{110\} + \{112\}$ slip systems (continuous line), pencil glide model (stars). The corresponding initial orientations are indicated in the standard triangle by a square of the same color.

reference full-field simulations. In this work, the following form is considered for \mathbb{D} , as proposed by [22, 23] for the anisotropic elasto-plastic properties of multi-axial aggregates:

$$[\mathbb{D}\beta^{(g)}] = \begin{bmatrix} D_{11} & D_{12} & D_{23} & & \\ D_{12} & D_{33} & D_{23} & & \\ D_{23} & D_{23} & D_{33} & & \\ & & D_{44} & & \\ & & & D_{55} & & \\ & & & & D_{55} \end{bmatrix} \begin{bmatrix} \beta_{11}^{(g)} \\ \beta_{22}^{(g)} \\ \beta_{33}^{(g)} \\ \sqrt{2}\beta_{12}^{(g)} \\ \sqrt{2}\beta_{23}^{(g)} \\ \sqrt{2}\beta_{23}^{(g)} \\ \sqrt{2}\beta_{31}^{(g)} \end{bmatrix}$$
(20)

(~)

where the components are given in the material frame (X_1, X_2, X_3) of the metal sample, assuming transverse isotropy for the textures considered in the following simulations. The β variables



Figure 5. Meshes for full-field finite element model (FEM), and initially isotropic 1000 grain orientation distribution. Each grain contains eight finite elements with quadratic interpolation and reduced integration.

Table 6. Identified parameters for the β -model

$$D_{11} = 10$$
 $D_{12} = 1$ $D_{23} = 1$ $D_{44} = 5$ $D_{55} = 5$

are assumed to be deviatoric, i.e. $trace(\boldsymbol{\beta}^{(g)}) = 0$, so that the following additional condition is enforced [22]:

$$D_{11} = D_{33} + D_{23} - D_{12}.$$
 (21)

We keep five independent coeficients D_{ij} to be determined by an identification procedure. With this form of \mathbb{D} , it will be found that β -model predictions fit satisfactorily the results obtained by the full-field FEM reference for the anisotropic behaviour of polycrystalline aggregates. For that purpose, an elementary volume V made of a parallelepipedic tessellation of $(10 \times 10 \times 10)$ 1000 grains is considered, see Figure 5. Periodic boundary conditions are prescribed. The parameters D_{11} , D_{12} , D_{23} can be calibrated from a tensile test on this volume, whereas D_{44} and D_{55} are obtained from shear tests. An isotropic distribution of 1000 orientations is considered for the simulation and distributed randomly among the square grains of Figure 5. Almost isotropic \mathbb{C}_{Homo} and \mathbb{D} tensors are obtained. The found parameters for \mathbb{D} are given in Table 6. The same set of parameters is found to provide a correct fit for both $\{110\} + \{112\}$ and pencil glide models, as illustrated in Figure 6.

The FEM simulations were performed twice for each loading: one simulation taking the 24 slip systems into account in each grain, and one with the 4 pencil glide systems. The predicted yield stresses are very close to both models but the pencil glide model predicts significantly lower hardening. This is due to the fact that the applied resolved shear stresses on the pencil glide systems are greater than on the $\{110\} + \{112\}$ slip systems. Accordingly, the pencil glide model provides the lowest stress-strain relation possible due to an increase in degrees of freedom for the selection of the slip plane. Furthermore, the larger number of available slip systems in the reference model leads to more interactions via the interaction matrix and associated additional hardening. The mean field β -model is found to overestimate hardening compared with the reference FEM. It has been checked that different D_{ij} combinations will not reduce this deviation. It is noted that the same material parameters are used for the single crystal model in the FEM and mean field simulations. Better agreement can be reached using more



Figure 6. Tensile (called S22) and shear (called S12) curves for a 1000-grain polycrystal according to the full field (continuous line) and mean field (dashed lines) models: blue and green curves for the {110} + {112} crystal plasticity model; red and orange curves for the pencil glide model. Stress (resp. strain) values correspond to the Cauchy stress components σ_{22} and σ_{12} (resp. F_{22} – 1 and F_{12} prescribed components).



Figure 7. Activation zone for the slip direction [11] according to the pencil glide model (left) and activation zones for slip planes families according to Schmid's law (right).

sophisticated evolution equations for the β -variables [24] but this is not the focus of the present work. As such, these results demonstrate that the fast computational β -model is an efficient homogenization tool correctly reflecting the responses of full-field and mean field models at least for the considered loading conditions.

4.2. Tensile test

Tensiles tests have been performed for the mean field models with 352 random orientations. The result is a combination of all the examples of the previous section. In simple tension of single crystals, single slip leads to rotation of the tensile axis towards a $\langle 111 \rangle$ slip direction. In the case of polycrystals, because of the deviation of stress and strain from uniaxial tension, the phenomenon is more subtle. But the average rotation of grains can be similar. Figure 7 shows the activated zone associated with slip direction [$\bar{1}11$] for pencil glide (left) and the activated zone in standard



(0)

Figure 8. (a) Initial isotropic (left) and final grain orientations after 50% tensile straining using $\{110\} + \{112\}$ slip systems (middle) and pencil glide systems (right). (b) Trajectories of individual grains in the standard triangle in tension using $\{110\} + \{112\}$ slip systems (left) and pencil glide systems (right).

triangle by Schmid's law. The lattice rotation will be different if the initial orientation lies in the green or orange zones. More complex orientation paths are observed for initial orientations close to boundaries of the standard triangle.

Figure 8a shows the initial and final grain orientations as predicted by the crystal plasticity model using $\{110\} + \{112\}$ slips systems or pencil glide systems. The tensile axis at the end of the two simulations after 50% strain is close to the boundary [001] - [011], and the zone $[\bar{1}12] - [111]$. The pencil glide predicts similar results as the $\{110\} + \{112\}$ model, however with a more pronounced texture development.

Figure 8b shows the trajectories of individual grains according to both model predictions in tension. In both cases, if the tensile axis is close to $[011] - [\bar{1}11]$, the tensile axis will start rotating towards the slip direction [111] (with activation of $(\bar{2}11)[111]$ slip system), as long as single slip conditions dominate. At larger deformations, a secondary slip system (211)[$\bar{1}11$] is activated, associated with [$\bar{1}11$] slip direction. The tensile axis will then turn towards [011] direction and stay there. In contrast, if the tensile axis is close to $[001] - [\bar{1}11]$ boundary, at the beginning, double slip starts right away and the tensile axis turns down to [001] direction. When the grains reach the activation zone for {110} slip systems, the tensile axis turns down toward [111] direction because the ($\bar{1}01$)[111] slip system starts gliding.

If the initial orientation of the tensile axis is close to direction [001], one or multiple slip systems {112} are activated. But this direction is not stable in tension, so that at the end the tensile axis will turn up towards slip directions $\langle 111 \rangle$ (if the slip plane is $(1\bar{1}2)$, the slip direction is $[\bar{1}11]$). At large deformations, the tensile axis will rotate towards the activation zone of {110} slip systems. Their actual activation depends on the interaction matrix and the corresponding stress value that would allow for the activation of one or 2 slip systems, $(\bar{1}01)$ or (011). In the case of single slip, the



Figure 9. Activation zone of {110} + {112} slip systems (color) and pencil glide systems (dots).

tensile axis will turn down to direction [111]. In the case of double slip, the tensile axis could remain stable at $[\bar{1}12]$ direction. It is observed that both models provide the same results in this respect.

If the tensile axis lies in the central region of the standard triangle, the activated slip system family is {110}, and the tensile axis will rotate towards the slip direction [111]. The trajectory could pass the boundary [001] – [011], in this case latent hardening will postpone activation of new systems. At large deformations, the secondary slip system (101)[$\bar{1}11$] is activated so that the tensile axis will turn up again to the slip direction [$\bar{1}11$]. At the end the tensile axis may stabilize at the boundary [001]–[011]. For such initial orientations, larger differences are observed between the two model predictions (Figure 8b). With pencil glide, the tensile axis will turn down to boundary [001] – [011] whereas the consideration of {110} + {112} slip systems stabilizes the tensile axis at the upper corner of the green–orange intersection due to three activated slip systems: (1 $\bar{1}2$)[$\bar{1}11$], ($\bar{1}01$)[111].

Figure 9 shows the comparison of the activation zones for $\{110\} + \{112\}$ slip systems associated with [111] slip direction (orange, blue) and for pencil glide associate with the same slip directions (dotted region). Only slight differences can be observed. The circled region coincides with the 3 corner zone of 3 activated slip systems: $(1\bar{1}2)[\bar{1}11], (\bar{1}01)[111], (101)[\bar{1}11]$. More precise comparison is possible looking at the ODF maps of Figure 10. These maps are rather similar for both models, but in the case of pencil glide, the intensity of hot spots is a little higher: 7.66 compare to 6.44.

4.3. Compression test

In axial compression along axis X3, assuming that material planes remain parallel to the compression plane leads to the conditions $F_{31} = F_{32} = 0$. The third condition is taken as the rotation $F_{12} - F_{21} = 0$. Figure 11 shows the initial and final orientations after 0.5 compression straining $(F_{33} = 0.5)$ using $\{110\} + \{112\}$ slip systems or pencil glide systems. The compression axis of most grains at the end of both simulations is close to the boundary $[001] - [\bar{1}11]$ and particularly in the [001] and $[\bar{1}11]$ directions. Both models predict similar trends. It should be noted that, in tension, the slip directions of $\{110\} + \{112\}$ slip systems and the pencil glide are the same $\langle 111 \rangle$ but in compression, the difference between the $\{110\} + \{112\}$ model and pencil glide can reach 15°.

The trajectories of individual grains are given in Figure 11b. If the compression axis is close to $[011] - [\overline{1}11]$, the activated slip system is $(\overline{2}11)[111]$, so that the compression axis will rotate



(b) Pencil glide

Figure 10. ODF maps for 352 grains after 0.5 tensile straining according to two crystal plasticity models.

toward the slip plane normal $[\bar{2}11]$ close to $[\bar{1}11]$ direction. At very large deformations, the secondary slip systems $\{112\}$ can be activated and then the compression axis remains stable in the direction $[\bar{1}11]$.

If the compression axis lies in the middle of the standard triangle, the active slip planes are {110}, and the compression axis will turn up to the slip plane [$\bar{1}01$]. Then when the compression axis goes close to the boundary [001] – [$\bar{1}11$], the secondary slip system (011)[$\bar{1}\bar{1}1$] could be activated, leading to a slow rotation of the compression axis towards direction [$\bar{1}11$]. If the loading direction is at the boundary [001] – [011], two slip systems {110} could be activated, then the compression axis will turn towards the [001] direction.

If the compression axis is close to [001], the activated slip system is $(1\bar{1}2)[\bar{1}11]$. The compression axis turns down towards $[1\bar{1}2]$. When it passes close to the [001] direction, multi-slip can occur and [001] will be a stable orientation.

At the end the compression axis will stay close to $[001] - [\overline{1}11]$ zone and most probably close to the corner directions [001] or $[\overline{1}11]$. According to the model using $\{110\} + \{112\}$, the



Figure 11. (a) Initial isotropic texture (left) and final texture after compression straining using $\{110\} + \{112\}$ slip systems (middle) and pencil glide systems (right). (b) Trajectories of individual grains in the standard triangle during compression straining using $\{110\} + \{112\}$ slip systems (left) and pencil glide systems (right).

compression axis could stabilize at the corner where three systems are activated: $(1\bar{1}2)[\bar{1}11]$, $(\bar{1}01)[111]$, $(101)[\bar{1}11]$. This cannot occur when pencil glide is considered. The ODF maps of Figure 12 show that the pencil glide and $\{110\} + \{112\}$ essentially predict the same texture in simple compression. The maximum intensity in the case of pencil glide is almost identical: 12.71 compared to 12.62.

4.4. Rolling

Rolling textures are crucial to evaluate the capabilities of crystal plasticity models. Rolling is simulated here by plane strain compression along axis X_3 , with the plane strain direction along axis X_2 (ND): $F_{ij} = 0$ for $i \neq j$, $F_{22} = 1$, $F_{33} = 1 + \epsilon$ is prescribed. The rolling direction RD is X_1 .

The rolling textures of b.c.c. metals are generally composed of certain orientation fibers and their main features can thus be represented in a condensed manner by plotting the orientation density along these fibers. The most important fibers and the ideal rolling texture components are described in Table 7 and illustrated in Figure 13. Figure 14 (top figure) shows the textures obtained from experiments in [52] that can be compared with the results of several crystal plasticity models.

The bottom Figure 14 presents the pole figures predicted by the Taylor model at 20% deformation. The experimental and predicted textures are similar. The main fibers like α or γ can be observed. At small deformations the Taylor model is known to provide good predictions of rolling textures at limited strain levels. Limitations of the Taylor model are expected at large deformations.

Figure 15 displays the rolling textures as predicted by the β -model at 100% logatithmic strain ($F_{33} = 0.36$) using the {110} + {112} slip systems and the pencil glide. The comparison shows



(b) Pencil glide

Figure 12. ODF maps for 352 grains after 0.5 compression straining according to two crystal plasticity models.

Table 7. The important fibers and texture components for crystallographic textures of b.c.c., after [53, 54]

Fiber name	Fiber axis	Important texture components
α -fiber	{110} parallel to RD	$\{001\}\langle 110\rangle, \{112\}\langle 110\rangle, \{111\}\langle 110\rangle$
γ-fiber	{110} parallel to ND	$\{111\}\langle 112\rangle, \{111\}\langle 110\rangle$
η -fiber	{001} parallel to RD	$\{001\}\langle100 angle,\{011\}\langle100 angle$

that pencil glide leads to an increase in the intensity of texture. This result is known from other works [32]. The final textures are rather similar. However there are some differences in the development of the two fibers α and γ between the models. Figure 16 shows the ODF sections for $\varphi_2 = 45^\circ$ as predicted by the β -model using the $\{110\} + \{112\}$ slip systems and the pencil



Figure 13. Description of α -fiber and γ -fiber, after [55].



Figure 14. Top: {110} pole figures for ARMCO® iron (a) as-received; (b) after cold rolling to 64% rolling reduction [52]. Bottom: texture predicted by the Taylor model with {110} + {112} slip systems: (a) {100} pole figure, (b) {110} pole figure, after [56].



(b) Pencil glide.

Figure 15. Rolling pole figures predicted by the β -model based on two different slip mechanisms.



(b) Pencil glide.

Figure 16. ODF section for $\phi_2 = 45$ after 100% logarithmic rolling strain.

glide models. One can observe the distinct development of the two fibers α and γ according to both models, see also the discussion in the ideal case in [57].

For a more accurate comparison, the evolution of the intensity of the α fiber is plotted in Figure 17a. All the models predict that the α fiber continuously grows with strain. The shape, the number of peaks and their location are similar for the two models considered in this work, at least after 70%. Clearly the final intensity resulting from the pencil glide mechanism is larger, 18, than considering the {110} + {112} slip systems, 12. Comparison with other models shows that the number of available slips systems controls the intensity of the fiber [59]. For instance



(a) $\{110\} + \{112\}$ slip systems (left) and pencil glide (right)



(b) $\{110\}$ slip systems (left) and $\{110\} + \{112\} + \{123\}$ (right) Taylor model



(c) {110} slip systems (left) and {110} + {112} + {123} (right) VPSC model

Figure 17. Comparison of rolling texture fibers as predicted by (a) the β -model, (b) the Taylor model [58] and (c) the VPSC model, for various combinations of slip system families [58], against pencil glide.



Figure 18. Experimental pole figures for ARMCO® iron sheared, after [61].

Table 8. The important fibers and texture components for crystallographic textures of b.c.c.metals, after [60]

Components	{hkl}	$\langle uvw \rangle$	ϕ_1	Φ	ϕ_2
D_1	{111}	$\langle 011 \rangle$	125.26	45.0	0.0
D_2	$\{\bar{1}\bar{1}1\}$	$\langle 011 \rangle$	54.74	45.0	0.0
E_1	$\{1\bar{1}1\}$	$\langle 121 \rangle$	270.0	35.26	45.0
E_2	$\{\bar{1}\bar{1}1\}$	$\langle 112 \rangle$	90.0	35.26	45.0
F	{ 1 00}	$\langle 011 \rangle$	180.0	45.0	0.0
J_1	$\{1\bar{2}1\}$	$\langle 111 \rangle$	270.0	54.74	45.0
J_2	$\{\bar{1}\bar{1}2\}$	$\langle 111 \rangle$	90.0	54.74	45.0
Т	{91 0 10}	$\langle 15536 \rangle$	46.5	55.3	17.1

in Figure 17b, a different development of the peaks and the intensity values is observed for the Taylor model [58]. Including the {123} family in addition to {110} + {112} slip systems makes the Taylor prediction closer to the predictions of Figure 17a (right), with a maximal intensity of about 11 and larger peak width. After 50% strain, the β -model predicts a single peak whereas two peaks are observed in the Taylor simulations. The results of the β -model are closer to the VPSC (Viscoplastic Self-Consistent) estimates, reproduced in Figure 17c from [58], but the intensities are found to be higher. The maximum intensities according to the VPSC model for {110} + {112} + {123} slip systems respectively are 11 and 14, instead of 12 and 18 (pencil glide). It follows that not only the number of slips systems controls the intensity but also the chosen homogenization scheme. The other parameters influencing the peak sharpness are the interaction matrix and the hardening modulus.

4.5. Simple shear case

The simple shear test conditions correspond to a prescribed value of the component $F_{12} = \gamma$, whereras all other components of the deformation gradient $\mathbf{F} - \mathbf{1}$ vanish. Experiments are often performed using torsion tests, which may not be equivalent to the simple shear considered here for simplicity. In b.c.c. materials, the crystallographic texture is composed of several main components listed in Table 8, after [60].

There are fewer texture evolution results under simple shear in the literature for b.c.c. metals than for f.c.c. Williams [61] tested the ARMCO® iron at room temperature, up to a shear strain of 2.1 (Figure 18). The final texture is dominated by the $\{112\}\langle 111\rangle$ (J1,J2), $\{110\}\langle 001\rangle$ (F) components.

The β -model provides a good estimation of the experimental results as shown in Figure 19. At $\gamma = 1$. there is a slight difference between the pencil glide and the {110} + {112} slip systems.



(a) Shear amount $F_{12} = \gamma = 1$. using $\{110\} + \{112\}$ slip systems (top), and pencil glide (bottom)



(b) Shear amount $F_{12} = \gamma = 1.5$ using $\{110\} + \{112\}$ slip systems (top), and pencil glide (bottom)



(c) Shear amount $F_{12} = \gamma = 2.1$ using $\{110\} + \{112\}$ slip systems (top), and pencil glide (bottom)

Figure 19. Pole figures showing the crystallographic textures induced by simple shear using the β -model.



(a) Shear amount $F_{12} = \gamma = 1$ using $\{110\} + \{112\}$ slip systems (left), and pencil glide (right)



(b) Shear amount $F_{12} = \gamma = 1.5$ using $\{110\} + \{112\}$ slip systems (left), and pencil glide (right)



(c) Shear amount $F_{12} = \gamma = 2.1$ using {110} + {112} slip systems (left), and pencil glide (right)

Figure 20. Volume fractions of main components after simple shear according to the β -model.

Looking at the volume fraction of these components plotted in Figure 20a shows that the texture is dominated by the $\{112\}\langle 111\rangle$ (J1,J2) and $\{110\}\langle 001\rangle$ (F) components. But the component F is more important in the pencil glide case (volume fraction = 0.4) than in the $\{110\} + \{112\}$ case (volume fraction = 0.26), see Figure 20a.

Increasing the shear strain γ leads to significant texture changes, and the pencil glide does not describle accurately the experimental results. At $\gamma = 1.5$ the texture induced by pencil glide

871

is dominated by the {112}(111) (J1,J2) components (volume fraction = 0.35) even though the F component is still present (volume fraction = 0.18). In contrast, {110} + {112} slip leads to a dominating {110}(001) (F) component (volume fraction = 0.41) (Figure 20b).

At $\gamma = 2.1$ the texture resulting from pencil glide is dominated by the {112} (111) (J1,J2) components (volume fraction = 0.4) and the F component is fading away. The texture resulting from {110} + {112} slip systems is dominated only by the {110}(001) (F) components (volume fraction = 0.5) (Figure 20c).

5. Discussion

Recent detailed studies on iron single crystals show that the pencil glide must be regarded as a simplified view of actual slip processes occurring on many slip planes [2, 3, 62]. In the present work we insist on the merits of the pencil glide model from the point of view of computational efficiency. In metals and alloys deforming by means of many simultaneous plasticity mechanisms like multiple slip families, twinning and martensitic transformation (like in TRIP-TWIP steels), accounting for all mechanisms leads to a dramatic increase in the number of internal variables to be intregrated. This can be accompanied by numerical instabilities or excessive computation time and storage. The use of pencil glide systems can therefore become advantageous. The analyses of the previous sections show that the pencil glide model can mimick the results obtained considering $\{110\} + \{112\}$ slip systems in many situations, even though significant differences have been evidenced in some loading cases.

Some comparisons between both approaches were performed in the past using the Taylor model for some limited loading conditions. In the present work the more general β -model was used and new situations were considered for comparison, namely the single crystal case, the simple shear of polycrystals and detailed analyses of tension, compression and rolling.

It must be emphasized that the final texture also depends on the interaction matrix and hardening parameters. On the one hand, 24 slip systems are used with the complete interaction matrix. On the other hand, the pencil glide approach makes use of a reduced interaction matrix. The full interaction matrix requires as much as 17 parameters: one for self-hardening, one for the dipolar interaction, twelve for junction formation and three for collinear annihilation. Some of them controls the activation of secondary slip system, that can be evidenced by the tension and compression tests. In the work [63], the authors used "full constraint" and "relaxed constraint" Taylor models. When glide is limited to {112} slip systems, the simulated texture is very sharp and the tube of orientations {111}(uuw) is not well described by any of these models. On the other hand, when glide is restricted to {110} systems, the simulations predict all the observed components. The simulated textures in this case are less sharp. A combination of the simulated textures however provides most of the features observed experimentally.

The collinear annihilations are known as the strongest interactions between dislocations because they decrease the length of dislocation sources [64]. It follows that collinear slip systems are not activated in many cases. This is in contrast to experimental observations showing that the cross slip mechanism is ubiquitous in b.c.c. crystals, so that dislocations can change plane to overcome obstacles. This is at the origin of the pencil glide mechanism. In the present work, we have still adopted the collinear annihilations as the strongest interaction in the matrix. Changing this value could result in significantly different final textures.

The β -model at large deformations used in the present work is an extended version of the one proposed in [21] where isotropic elasticity was assumed and the tensor \mathbb{D} was reduced to a scalar. Improvements are still needed like the account of grain shape changes during deformation. The presented β -model assumes that, in contrast to the strain rate tensor, the material spin is the same for all grains. Special localization rules should also be developed for the spin tensor.

6. Conclusion

The homogenization β -model at large deformations has been used to predict the crystallographic textures of b.c.c. metals when accounting for 24 slip systems or when introducing the pencil glide mechanism instead.

The effect was first analyzed in the case of single crystal behaviour including a detailed comparison with recent experimental results for α -iron. For simple tension and compression of various single crystal orientations, no significant difference was found in the overall stress–strain curves and in lattice rotation whether pencil glide or the 24 {110} + {112} slip systems were used.

In the rolling and simple shear tests where the number of slips systems and the interaction matrix control the texture, some discrepancies were found in the predictions based on pencil glide or $\{110\} + \{112\}$ slip systems, especially at larger deformation values. These discrepancies are not significant for strain values below 100%.

Finally, it must be underlined that the computational time required for the pencil glide simulations is three times smaller than for the 24 slips systems. This lower computing time is a major advantage of the pencil glide approach. This makes the pencil glide model particularly well-suited for finite element simulations of alloy forming processes using polycrystal models which are still computationally demanding problems at the present time [28, 65].

Dedication

Dedicated to Pierre Gilormini in remembrance of his early contributions on pencil glide modelling.

Acknowledgements

This study was funded by Agence Nationale de la Recherche through the ANR TITWIP project (grant no. ANR-15-CE08-0013).

Appendix A.

In this section, the parameterization of space rotations by means of exponential mapping used in the code is reviewed. Any rotation **R** can be represented by the element Φ of the associated Lie group such that:

$$\mathbf{R} = \exp\left(\mathbf{1} \times \mathbf{\Phi}\right) \quad \text{and} \quad \mathbf{\Phi} = \theta \mathbf{n} = [\Phi_1, \Phi_2, \Phi_3] \tag{22}$$

where θ is the rotation angle (in radians) around a specified unit axis **n**. One can rewrite the expression using the standard matrix exponential series, noting that: exp (**A**) = $\sum_{k=0}^{\infty} 1/k! \mathbf{A}^k$,

$$\mathbf{R} = \exp\left(\mathbf{1} \times \boldsymbol{\Phi}\right) = \cos\theta \mathbf{1} + \frac{1 - \cos\theta}{\theta^2} \boldsymbol{\Phi} \otimes \boldsymbol{\Phi} + \frac{\sin\theta}{\theta} \mathbf{1} \times \boldsymbol{\Phi}.$$
 (23)

In the matrix form:

$$\mathbf{R} = \begin{bmatrix} \cos\theta + \frac{1 - \cos\theta}{\theta^2} \Phi_1^2 & \frac{1 - \cos\theta}{\theta^2} \Phi_1 \Phi_2 - \frac{\sin\theta}{\theta} \Phi_3 & \frac{1 - \cos\theta}{\theta^2} \Phi_1 \Phi_3 + \frac{\sin\theta}{\theta} \Phi_2 \\ \frac{1 - \cos\theta}{\theta^2} \Phi_1 \Phi_2 + \frac{\sin\theta}{\theta} \Phi_3 & \cos\theta + \frac{1 - \cos\theta}{\theta^2} \Phi_2^2 & \frac{1 - \cos\theta}{\theta^2} \Phi_2 \Phi_3 - \frac{\sin\theta}{\theta} \Phi_1 \\ \frac{1 - \cos\theta}{\theta^2} \Phi_1 \Phi_3 - \frac{\sin\theta}{\theta} \Phi_2 & \frac{1 - \cos\theta}{\theta^2} \Phi_2 \Phi_3 + \frac{\sin\theta}{\theta} \Phi_1 & \cos\theta + \frac{1 - \cos\theta}{\theta^2} \Phi_3^2 \end{bmatrix}$$
(24)

Defining the spin tensor $\dot{\mathbf{R}}\mathbf{R}^T = \mathbf{L} \{ = \mathbf{\Omega}, \text{ the following relation can be worked out:}$

$$\dot{\mathbf{\Phi}} = \mathbf{T}^{-1} \hat{\mathbf{\Omega}}.$$
(25)

Note that any skew-symmetric matrix Ω can be represented by the pseudo-vector $\hat{\Omega}$ such that: $\Omega \mathbf{b} = \hat{\Omega} \times \mathbf{b}$. In matrix form:

×

$$\Omega = \begin{bmatrix} 0 & -\hat{\mathbf{\Omega}}_3 & \hat{\mathbf{\Omega}}_2 \\ \stackrel{\times}{\mathbf{\Omega}}_3 & 0 & -\hat{\mathbf{\Omega}}_1 \\ \stackrel{\times}{-\mathbf{\Omega}}_2 & \hat{\mathbf{\Omega}}_1 & 0 \end{bmatrix} \text{ so that } \hat{\mathbf{\Omega}} = [\hat{\mathbf{\Omega}}_1, \hat{\mathbf{\Omega}}_2, \hat{\mathbf{\Omega}}_3]^T.$$
(26)

The matrix form of the operator **T** is

$$\mathbf{T} = \begin{bmatrix} \frac{\sin\theta}{\theta} + \frac{1 - \frac{\sin\theta}{\theta}}{\theta^2} \Phi_1^2 & \frac{1 - \frac{\sin\theta}{\theta}}{\theta^2} \Phi_1 \Phi_2 - \frac{1 - \cos\theta}{\theta^2} \Phi_3 & \frac{1 - \frac{\sin\theta}{\theta}}{\theta^2} \Phi_1 \Phi_3 + \frac{1 - \cos\theta}{\theta^2} \Phi_2 \\ \frac{1 - \frac{\sin\theta}{\theta}}{\theta^2} \Phi_1 \Phi_2 + \frac{1 - \cos\theta}{\theta^2} \Phi_3 & \frac{\sin\theta}{\theta} + \frac{1 - \frac{\sin\theta}{\theta}}{\theta^2} \Phi_2^2 & \frac{1 - \frac{\sin\theta}{\theta}}{\theta^2} \Phi_2 \Phi_3 - \frac{1 - \cos\theta}{\theta^2} \Phi_1 \\ \frac{1 - \frac{\sin\theta}{\theta}}{\theta^2} \Phi_1 \Phi_3 - \frac{1 - \cos\theta}{\theta^2} \Phi_2 & \frac{1 - \frac{\sin\theta}{\theta}}{\theta^2} \Phi_2 \Phi_3 + \frac{1 - \cos\theta}{\theta^2} \Phi_1 & \frac{\sin\theta}{\theta} + \frac{1 - \frac{\sin\theta}{\theta}}{\theta^2} \Phi_3^2 \end{bmatrix}$$
(27)

The rotation angle can be obtained from the rotation matrix **R** by the following operation:

$$\theta = \arccos\left(\frac{\operatorname{trace}(\mathbf{R}) - 1}{2}\right) \tag{28}$$

and, if $\theta \neq 0$ and \mathbf{R}_{ij} are the components of **R**, the rotation axis is

$$\mathbf{n} = \frac{\mathbf{\Phi}}{\theta} = \frac{1}{2\sin\theta} [R_{32} - R_{23}, R_{13} - R_{31}, R_{21} - R_{12}].$$
(29)

Appendix B.

Each grain is represented by an ellipsoidal inclusion (a_1, a_2, a_3) embedded in a homogeneous effective medium (HEM) resulting from the average over all the grains. The Eshelby tensor S^{Esh} is calculated by using the Green function that depends on the inclusion shape and the elasticity tensor of the HEM \mathbb{C}^{Homo} :

$$\mathbb{S}_{ijkl}^{\text{Esh}} = \frac{\mathbb{C}_{mnkl}^{\text{Homo}}}{8\pi} \int_{-1}^{1} \mathrm{d}\zeta_3 \int_{0}^{2\pi} \left[\mathbb{G}_{imjn}(\xi) + \mathbb{G}_{jmin}(\xi)\right] \mathrm{d}\omega$$
(30)

with

$$\mathbb{G}_{ijkl}(\xi) = \xi_k \xi_l \frac{N_{ij}(\xi)}{D(\xi)}$$
(31)

$$\xi_1 = \frac{\sqrt{1 - \zeta_3^2}}{a_1} \cos \omega, \quad \xi_2 = \frac{\sqrt{1 - \zeta_3^2}}{a_2} \sin \omega, \quad \xi_3 = \frac{\zeta_3}{a_3}$$
(32)

$$K_{ik} = \mathbb{C}_{ijkl}^{\text{Homo}} \xi_j \xi_l \tag{33}$$

$$N_{ij}(\xi) = \frac{1}{2} \boldsymbol{\varepsilon}_{ijk} \boldsymbol{\varepsilon}_{jmn} K_{km} K_{ln} \tag{34}$$

$$D(\xi) = \boldsymbol{\varepsilon}_{mnl} K_{m1} K_{n2} K_{l3} \tag{35}$$

 ε_{ijk} is the Levi-Civita symbol. The Gaussian quadrature method is used to compute the integration in (22):

$$\mathbb{S}_{ijkl}^{E} = \frac{\mathbb{C}_{mnkl}^{\text{Homo}}}{8\pi} \sum_{p=1}^{M} \sum_{q=1}^{N} [\mathbb{G}_{imjn}(\omega_q, \zeta_{3p}) + \mathbb{G}_{jmin}(\omega_q, \zeta_{3p})] W_{pq}.$$
(36)

Here *M*, *N* are the number of nodes, and W_{pq} are the weights at the Gauss points. In the present article, the values M = 32 and N = 32 are applied.

References

- [1] P. Gilormini, "The theory of rate sensitive pencil glide application to rolling textures", *Acta Metall.* **37** (1989), p. 2093-2101.
- [2] L. Le, P. Franciosi, "Expectable specific features of BCC crystal plastic flow and consistency with the Schmid law", *Phil. Mag.* **93** (2013), p. 3589-3611.
- [3] P. Franciosi, L. Le, G. Monnet, C. Kahloun, M.-H. Chavanne, "Investigation of slip system activity in iron at room temperature by SEM and AFM in-situ tensile and compression tests of iron single crystals", *Int. J. Plast.* 65 (2015), p. 226-249.
- [4] G. Taylor, C. Elam, "The distortion of iron crystals", Proc. R. Soc. Lond. A 112 (1926), p. 337-361.
- [5] P. Gilormini, B. Bacroix, J. Jonas, "Theoretical analyses of (111) pencil glide in b.c.c. crystals", Acta Metall. 36 (1988), p. 231-256.
- [6] R. Becker, "Pencil glide formulation for polycrystal modelling", Scr. Metall. Mater. 32 (1995), p. 2051-2054.
- [7] G. Taylor, "Plastic strain in metals", J. Inst. Metals 62 (1938), p. 307-324.
- [8] G. Sachs, "Zur Ableitung einer Fliessbedingung", Z. Verein. Dtsch. Ing. 72 (1928), p. 734-736.
- [9] J. Bishop, R. Hill, "A theory of the plastic distortion of a polycrystalline aggregate under combined stresses", *Lond. Edinburgh Dublin Phil. Mag. J. Sci.* 42 (1951), no. 327, p. 414-427.
- [10] H. Honnef, H. Mecking, "A method for the determination of the active slip systems and orientation changes during single crystal deformation", in *Proceedings of the 5th International Conference on Texture of Materials*, Springer Verlag, 1978, p. 265-275.
- [11] H. Mecking, "Low temperature deformation of polycrystals", in *Proceedings of the 2th International Symposium on Metallurgy and Materials Science: Deformation of Polycrystals*, Danmarks Tekniske Universitet, 1981, p. 73-86.
- [12] U. Kocks, G. Canova, "How many slip systems and which?", in *Deformation of polycrystals: Mechanisms and Microstructures, 2nd International Symposium on Metallurgy and Materials Science* (N. Hansen, A. Horsewell, T. Leffers, H. E. Liholt, eds.), Risø National Laboratory, Roskilde, Denmark, 1981, p. 35-44.
- [13] P. Van Houtte, "On the equivalence of the relaxed taylor theory and the bishop-hill theory for partially constrained plastic deformation of crystals", *Mater. Sci. Eng.* **55** (1982), p. 69-77.
- [14] M. Arminjon, "Théorie d'une classe de modèles de Taylor Hétérogènes: application aux textures de déformation des aciers", Acta. Metall. 35 (1987), p. 615.
- [15] M. Arminjon, C. Donadille, "Présentaion d'un modèle polycrystallin extrémal: application aux aciers d'un modèle approché", Mém. Et. Sci. Revue Métall. 87 (1990), p. 359.
- [16] A. Molinari, "Extensions of the self-consistent tangent model", Model. Simul. Mater. Sci. Eng. 7 (1999), p. 683-697.
- [17] R. Lebensohn, C. Tome, "A self-consistent visco-plastic model: calculation of rolling textures of anisotropic materials", *Mater. Sci. Eng. A* 175 (1994), p. 71-82.
- [18] M. Berveiller, A. Zaoui, "An Extension of the self-consistent scheme to plastically flowing polycrystals", J. Mech. Phys. Solids 26 (1979), p. 325-344.
- [19] P. Lipinski, J. Krier, M. Berveiller, "Elastoplasticité des métaux en grandes déformations: comportement global et évolution de la structure interne", *Rev. Phys. Appl.* 25 (1990), p. 361-388.
- [20] G. Cailletaud, "A micromechanical approach to inelastic behaviour of metals", Int. J. Plast. 8 (1992), p. 55-73.
- [21] S. Forest, P. Pilvin, "Modelling finite deformation of polycrystals using local objective frames", Z. Angew. Math. Mech. 79 (1999), p. 199-202.
- [22] K. Sai, G. Cailletaud, S. Forest, "Micro-mechanical modeling of the inelastic behavior of directionally solidiffed materials", Mech. Mat. 38 (2006), no. 3, p. 203-217.
- [23] G. Martin, N. Ochoa, K. Sai, E. Hervé-Luanco, G. Cailletaud, "A multiscale model for the elastoviscoplastic behavior of directionally solidified alloys: application to FE structural computations", *Int. J. Solids Struct.* 51 (2014), p. 1175-1187.
- [24] G. Cailletaud, F. Coudon, "Scale transition rules applied to crystal plasticity", in *Materials with Internal Structure: Multiscale and Multifield Modeling and Simulation* (P. Trovalusci, ed.), Springer International Publishing, 2016, p. 1-15.
- [25] F. Coudon, G. Cailletaud, J. Cormier, L. Marcin, "A multiscale model for nickel-based directionally solidified materials", *Int. J. Plast.* 115 (2019), p. 1-17.
- [26] P. Pilvin, "Une approche inverse pour l'identification d'un modèle polycristallin élastoviscoplastique", in 3ème Colloque National en Calcul des Structures, 20–23 mai, Giens, Presses Académiques de l'Ouest, 1997, p. 207-212.
- [27] J. Besson, G. Cailletaud, J.-L. Chaboche, S. Forest, M. Blétry, in *Non–Linear Mechanics of Materials*, Solid Mechanics and Its Applications, vol. 167, Springer-Verlag, Berlin, Heidelberg, 2009.

- [28] N. Hfaiedh, A. Roos, H. Badreddine, K. Saanouni, "Interaction between ductile damage and texture evolution in finite polycrystalline elastoplasticity", *Int. J. Damage Mech.* 28 (2019), p. 481-501.
- [29] J. Mandel, "Equations constitutives et directeurs dans les milieux plastiques et viscoplastiques", Int. J. Solids Struct. 9 (1973), p. 725-740.
- [30] A. Krawietz, "Efficient integration in the plasticity of crystals with pencil glide and deck glide", *Tech. Mech.* **21** (2001), p. 243-250.
- [31] R. Glüge, "Glide mechanisms for bundle- and plate-like structures", Tech. Mech. 35 (2015), p. 62-69.
- [32] C. Guillemer, M. Clavel, G. Cailletaud, "Cyclic behavior of extruded magnesium: experimental, microstructural and numerical approach", Int. J. Plast. 27 (2011), p. 2068-2084.
- [33] M. Lindroos, G. Cailletaud, A. Laukkanen, V.-T. Kuokkala, "Crystal plasticity modeling and characterization of the deformation twinning and strain hardening in Hadfield steels", *Mat. Sci. Eng.* 720 (2018), p. 145-159.
- [34] P. Pilvin, "The Contribution of Micromechanical Approaches to the Modelling of Inelastic Behaviour of Polycrystals", in *Fourth Int. Conf. on Biaxial/multiaxial Fatigue and Design (London)* (A. Pineau, G. Cailletaud, T. Lindley, eds.), ESIS, vol. 21, Mechanical Engineering Publications, 1996, p. 3-19.
- [35] G. Martin, L. Nazé, G. Cailletaud, "Numerical multi-scale simulations of the mechanical behavior of β -metastable titanium alloys Ti5553 and Ti17", *Procedia Eng.* **10** (2011), p. 1803-808.
- [36] P. Pilvin, F. Onimus, R. Brenner, S. Pascal, X. Feaugas, K. Saï, "Finite element assessment of an affine self-consistent model for hexagonal polycrystals", *Eur. J. Mech. - A/Solids* 61 (2017), p. 345-356.
- [37] T. Gu, J.-R. Medy, V. Klosek, O. Castelnau, S. Forest, E. Hervé-Luanco, F. Lecouturier-Dupouy, H. Proudhon, P.-O. Renault, L. Thilly, P. Villechaise, "Multiscale modeling of the elasto-plastic behavior of architectured and nanostructured Cu-Nb composite wires and comparison with neutron diffraction experiments", *Int. J. Plast.* **122** (2019), p. 1-30.
- [38] A. Bertram, M. Kraska, "Determination of finite plastic deformations in single crystals", Arch. Mech. 47 (1995), p. 203-222.
- [39] D. Caillard, "Kinetics of dislocations in pure Fe. Part I. In situ straining experiments at room temperature", Acta Mater. 58 (2010), p. 3493-3503.
- [40] J. Kumagai, S. Takaki, S. Suzuki, H. Kimura, "Orientation dependence of quasi-three-stage work hardening in high purity iron single crystals", *Mater. Sci. Eng.* A 129 (1990), no. 2, p. 207-215.
- [41] B. Jaoul, Etude de la plasticité et application aux métaux, 1st published, Presses des Mines de Paris, 1965.
- [42] G. Monnet, D. Terentyev, "Structure and mobility of the edge dislocation in BCC iron studied by molecular dynamics", Acta Mater. 57 (2009), no. 5, p. 1416-1426.
- [43] S. Queyreau, J. Marian, M. Gilbert, B. Wirth, "Edge dislocation mobilities in bcc Fe obtained by molecular dynamics", *Phys. Rev. B* 84 (2011), no. 6, article no. 064106.
- [44] C. Domain, G. Monnet, "Simulation of screw dislocation motion in iron by molecular dynamics simulations", *Phys. Rev. Lett.* 95 (2005), no. 21, article no. 215506.
- [45] P. Franciosi, "Etude théorique et expérimentale du comportement élastoplastique des monocristaux métalliques se déformant par glissement : modélisation pour un chargement complexe quasi-statique", PhD Thesis, Université Paris 13, 1984.
- [46] R. Madec, L. Kubin, *Dislocation Interactions and Symmetries in BCC Crystals*, Springer, Netherlands, 2004, 69-78 pages.
- [47] R. Madec, B. Devincre, L. Kubin, "From dislocation junctions to forest hardening", *Phys. Rev. Lett.* **89** (2003), article no. 255508.
- [48] L. P. Kubin, R. Madec, B. Devincre, "Dislocation intersections and reactions in FCC and BCC crystals", MRS Proc. 779 (2003), article no. W1.6.
- [49] R. Madec, B. Devincre, L. Kubin, T. Hoc, D. Rodney, "The role of collinear interaction in dislocation-induced hardening", *Science* **301** (2003), p. 1879-1882.
- [50] S. Queyreau, G. Monnet, B. Devincre, "Slip systems interactions in a-iron determined by dislocation dynamics simulations", *Int. J. Plast.* 25 (2009), no. 2, p. 361-377.
- [51] L. T. Le, "Etude par essais in situ MEB et AFM sur monocristaux des mécanismes de glissement á température ambiante de la ferrite de pureté commerciale", PhD Thesis, Université Paris Nord, 2013.
- [52] E. Bruder, "Mechanical properties of ARMCO $^{\delta}$ iron after large and severe plastic deformation-application potential for precursors to ultrafine grained microstructures", *Metals* **8** (2018), p. 191-203.
- [53] P. Coulomb, Les textures dans les métaux de réseau cubique, Dunod, Paris, France, 1972.
- [54] U. Kocks, C. Tomé, H. R. Wenk, Texture and Anisotropy, Cambridge University Press, 1998.
- [55] A. Das, "Calculation of crystallographic texture of BCC steels during cold rolling", J. Mat. Eng. Perform. 26 (2017), no. 6, p. 2708-2720.
- [56] D. Ceccaldi, F. Yala, T. Baudin, R. Penelle, F. Royer, M. Arminjon, "Deformation textures and plastic anisotropy of steels using the Taylor and nonhomogeneous models", *Int. J. Plast.* 10 (1994), p. 643-661.

Lu Tuan Le et al.

- [57] M. Hoelscher, D. Raabe, K. Luecke, "Rolling and recrystallization textures of BCC steels", Steel Res. Int. 62 (1991), p. 567-575.
- [58] S. M'Guil, W. Wen, S. Ahzi, J. J. Gracio, "Modeling of large plastic deformation behavior and anisotropy evolution in cold rolled bcc steels using the viscoplastic ϕ -model-based grain-interaction", *Mat. Sci. Eng.: A* **528** (2011), p. 5840-5853.
- [59] D. Raabe, "Investigation of contribution of {123} slip planes to development of rolling textures in bcc metals by use of Taylor models", *Mater. Sci. Technol.* **11** (1995), p. 455-460.
- [60] J. Baczynski, J. J. Jonas, "Texture development during the torsion testing of α -iron and two IF steels", *Acta Mater.* **44** (1996), p. 4273-4288.
- [61] O. W. Williams, "Shear texture in copper, brass, aluminum, iron and zirconium", *Trans. Metall. Soc. AIME* **224** (1962), p. 129-139.
- [62] C. Kahloun, G. Monnet, S. Queyreau, L. Le, P. Franciosi, "A comparison of collective dislocation motion from single slip quantitative topographic analysis during in-situ AFM room temperature tensile tests on Cu and Feα crystals", *Int. J. Plast.* 84 (2016), p. 277-298.
- [63] J. L. Raphanel, P. Van Houtte, "Simulation of the rolling textures of b.c.c. metals by means of the relaxed taylor theory", Acta Metall. 33 (1985), p. 1481-1488.
- [64] L. P. Kubin, R. Madec, B. Devincre, "Dislocation intersections and reactions in FCC and BCC crystals", MRS Proc. 779 (2003), article no. W1.6.
- [65] G. Cailletaud, P. Pilvin, "Utilisation de modèles polycristallins pour le calcul par éléments finis", Rev. Européenne des Éléments Finis 3 (1994), p. 515-541.



Contributions in mechanics of materials

Polycrystal thermo-elasticity revisited: theory and applications

Carlos N. Tomé^a and Ricardo A. Lebensohn^{*, a}

^{*a*} Los Alamos National Laboratory, Los Alamos, NM 87544, USA *E-mails*: tome@lanl.gov (C. N. Tomé), lebenso@lanl.gov (R. A. Lebensohn)

Abstract. The self-consistent (SC) theory is the most commonly used mean-field homogenization method to estimate the mechanical response behavior of polycrystals based on the knowledge of the properties and orientation distribution of constituent single-crystal grains. The original elastic SC method can be extended to thermo-elasticity by adding a stress-free strain to an elastic constitutive relation that expresses stress as a linear function of strain. With the addition of this independent term, the problem remains linear. Although the thermo-elastic self-consistent (TESC) model has important theoretical implications for the development of self-consistent homogenization of non-linear polycrystals, in this paper, we focus on TESC applications to actual thermo-elastic problems involving non-cubic (i.e. thermally anisotropic) materials. To achieve this aim, we provide a thorough description of the TESC theory, which is followed by illustrative examples involving cooling of polycrystalline non-cubic metals. The TESC model allows studying the effect of crystallographic texture and single-crystal elastic and thermal anisotropy on the effective thermo-elastic response of the aggregate and on the internal stresses that develop at the local level.

Keywords. Homogenization, Self-consistent methods, Thermo-elasticity, Polycrystals, Anisotropy, Metals.

Manuscript received 7th May 2020, revised and accepted 6th July 2020.

1. Introduction

The self-consistent (SC) theory is the most commonly used mean-field homogenization method for estimating the mechanical response behavior of polycrystals based on the knowledge of the properties and orientation distribution of constituent single-crystal grains. The SC method was proposed independently by Hershey [1] and Kröner [2] for linear elastic polycrystals, and it was subsequently expanded for this constitutive regime by Hill [3] and Zeller and Dederichs [4]. Although in the early days of the formulation, computational restrictions limited the applications of SC models to relatively simple cases, a general implementation of the elastic SC model is now part of homogenization-based polycrystal mechanics codes such as the VPSC code [5].

Consideration of thermo-elasticity adds a stress-free strain to the elastic constitutive relation that expresses stress as a linear function of strain. With the addition of this independent term,

^{*} Corresponding author.

the problem remains linear. The solution of a thermo-elastic self-consistent (TESC) problem for composites, consisting of a phase made of inclusions embedded in a matrix phase with different mechanical properties, was introduced by Walpole [6] and further reformulated and extended by, for example, Willis [7], Buryachenko [8], and Milton [9]. These works identified and, in some cases, also tackled the case of polycrystals as a special composite made of many *mechanical* phases, with distinct properties associated with different single-crystal orientations, with no matrix phase. Tomé *et al.* [10] presented the TESC formulation specialized for polycrystals to predict the thermo-mechanical response of hexagonal close packed (hcp) Zr aggregates, which, as almost every non-cubic material, has anisotropic thermal properties at the single-crystal level. The TESC formulation also has important theoretical implications for the development of SC homogenization of non-linear polycrystals. Indeed, for aggregates of grains deforming in the viscoplastic regime, the different available SC theories vary in the procedure used to linearize the non-linear mechanical behavior at the grain level, but eventually all of them make use of the TESC theory applied to the linearized behavior of an iteratively adjusted generalized thermo-elastic polycrystalline material.

Despite its relevance, a thorough exposition of the TESC theory with applications to actual thermo-elastic polycrystal problems is lacking in the literature. Only a brief description can be found in [10] as one among several applications of different SC models to different deformation regimes (TESC being one of them) for the interpretation of experimental data on Zr alloys. The present contribution intends to fill this gap by presenting the polycrystal TESC theory in a comprehensive manner, which is followed by illustrative examples involving cooling of polycrystalline non-cubic metals to study the effect of crystallographic texture and single-crystal elastic and thermal anisotropy on the effective and local thermo-elastic responses of the aggregate.

2. Model

In the context of the SC theory, a polycrystal is represented by a set of weighted crystal orientations. The orientations represent mechanical phases (i.e. set of single-crystal grains with the same orientation and morphology surrounded by different neighborhoods) or statistically representative "grains" (in what follows, "grains" and mechanical phases will be used interchangeably), and the weights represent volume fractions. The set of orientations and weights is chosen to reproduce the initial crystallographic texture of the material. Each grain is treated as an *ellipsoidal* elastic inhomogeneous inclusion embedded in an effective homogenized thermo-elastic medium. The ellipsoidal shape (or distribution of ellipsoidal shapes) represents the morphologic texture of the polycrystal. The inhomogeneity character derives from the difference in elastic properties between each individual grain and the effective homogenized medium when expressed in the same reference system. The inclusion character derives from the possible presence of eigenstrains such as thermal strains. The inhomogeneous inclusions and the medium generally have fully anisotropic properties, deriving from the intrinsic single-crystal anisotropy, crystallographic texture, and/or non-equiaxed morphology. The effective medium represents the average environment surrounding each orientation. Deformation is based on anisotropic crystal elasticity and thermal expansion, which in the case of non-cubic crystals is also anisotropic.

2.1. Local constitutive behavior and homogenization

The local thermo-elastic constitutive behavior in a material point **x** of the polycrystal, relating the local strain $\boldsymbol{\varepsilon}(\mathbf{x})$ and stress $\boldsymbol{\sigma}(\mathbf{x})$ is described by the linear relation

$$\varepsilon_{ij}(\mathbf{x}) = C_{ijkl}^{-1}(\mathbf{x})\sigma_{kl}(\mathbf{x}) + \alpha_{ij}(\mathbf{x})\delta T(\mathbf{x}) = \varepsilon_{ij}^{\text{el}}(\mathbf{x}) + \varepsilon_{ij}^{\text{th}}(\mathbf{x}),$$
(1)

$$\sigma_{ij}(\mathbf{x}) = C_{ijkl}(\mathbf{x})(\varepsilon_{kl}(\mathbf{x}) - \varepsilon_{kl}^{\text{tn}}(\mathbf{x})), \tag{2}$$

where the moduli $\mathbf{C}(\mathbf{x})$ and $\boldsymbol{\alpha}(\mathbf{x})$ are the elastic stiffness and the thermal expansion tensors of the single crystal containing the material point \mathbf{x} , which, in general, can be considered to be piecewise constant within the domain of each grain (g) and thus can be denoted by $\mathbf{C}^{(g)}$ and $\boldsymbol{\alpha}^{(g)}$, respectively; $\delta T(\mathbf{x})$ is the change in local temperature from the temperature at which the aggregate is free of thermal stresses; and $\boldsymbol{\epsilon}^{\rm el}(\mathbf{x})$ and $\boldsymbol{\epsilon}^{\rm th}(\mathbf{x})$ are the elastic and thermal strains, respectively. The thermal strain can be regarded as a stress-independent eigenstrain induced by temperature change.

Assuming uniform temperature distribution, the following relation holds within the domain of grain (*g*):

$$\boldsymbol{\sigma}(\mathbf{x}) = \mathbf{C}^{(g)} \colon \boldsymbol{\varepsilon}(\mathbf{x}) - \mathbf{C}^{(g)} \colon \boldsymbol{\alpha}^{(g)} \delta T,$$
(3)

$$\boldsymbol{\varepsilon}(\mathbf{x}) = \mathbf{C}^{(g)^{-1}} : \boldsymbol{\sigma}(\mathbf{x}) + \boldsymbol{\alpha}^{(g)} \delta T, \tag{4}$$

where δT is the uniform change in temperature from the temperature at which the aggregate is free of thermal stresses. At the aggregate level, a similar constitutive law holds, involving the effective elastic and thermal moduli:

$$\overline{\boldsymbol{\sigma}} = \overline{\mathbf{C}} : \, \overline{\boldsymbol{\varepsilon}} - \overline{\mathbf{C}} : \, \overline{\boldsymbol{\alpha}} \delta \, T, \tag{5}$$

$$\overline{\boldsymbol{\varepsilon}} = \overline{\mathbf{C}}^{-1} : \, \overline{\boldsymbol{\sigma}} + \overline{\boldsymbol{\alpha}} \delta \, T. \tag{6}$$

The bars on top indicate the polycrystal's effective tensors. The effective elastic stiffness **C** and effective thermal expansion $\overline{\alpha}$ are *a priori* unknown and need to be determined. From Hill's lemma [11], the volume averages of the local stress and strain tensors, denoted here as $\langle \rangle$, correspond to the effective magnitudes $\overline{\boldsymbol{\epsilon}} = \langle \boldsymbol{\epsilon}(\mathbf{x}) \rangle$ and $\overline{\boldsymbol{\sigma}} = \langle \boldsymbol{\sigma}(\mathbf{x}) \rangle$, respectively. Taking volumetric average of (3) and (4), and using (5) and (6), respectively, leads to the following relations:

$$\overline{\mathbf{C}}: \overline{\boldsymbol{\varepsilon}} - \overline{\mathbf{C}}: \overline{\boldsymbol{\alpha}} \delta T = \langle \mathbf{C}^{(g)}: \boldsymbol{\varepsilon}(\mathbf{x}) \rangle - \langle \mathbf{C}^{(g)}: \boldsymbol{\alpha}^{(g)} \rangle \delta T,$$
(7)

$$\overline{\mathbf{C}}^{-1}: \overline{\boldsymbol{\sigma}} + \overline{\boldsymbol{\alpha}} \delta T = \langle \mathbf{C}^{(g)^{-1}}: \boldsymbol{\sigma}(\mathbf{x}) \rangle + \langle \boldsymbol{\alpha}^{(g)} \rangle \delta T.$$
(8)

These relations couple the elastic and thermal tensors of the grain with those of the aggregate, and they are completely general. In this work, we provide the algorithm to solve for $\overline{\mathbf{C}}$ and $\overline{\boldsymbol{\alpha}}$ within the framework of the TESC formulation. However, before describing the latter, it is instructive to present the also commonly used upper-bound (UB) and lower-bound (LB) approaches. The simplest expressions for $\overline{\mathbf{C}}$ and $\overline{\boldsymbol{\alpha}}$ obtained from (7) or (8) follow from assuming uniform strain $\boldsymbol{\varepsilon}(\mathbf{x}) = \overline{\boldsymbol{\varepsilon}}$ or uniform stress $\boldsymbol{\sigma}(\mathbf{x}) = \overline{\boldsymbol{\sigma}}$ in the aggregate, respectively. In the former case (known as Voigt assumption in elasticity) and if (7) has to be valid for an arbitrary strain and an arbitrary temperature increment, the overall moduli are given by

$$\overline{\mathbf{C}}^{\mathrm{UB}} = \langle \mathbf{C}^{(g)} \rangle \quad \text{and} \quad \overline{\boldsymbol{\alpha}}^{\mathrm{UB}} = \overline{\mathbf{C}}^{UB^{-1}} \colon \langle \mathbf{C}^{(g)} \colon \boldsymbol{\alpha}^{(g)} \rangle.$$
(9)

Conversely, if the stress is assumed to be uniform in the aggregate (Reuss assumption in elasticity) and (8) holds for an arbitrary stress and an arbitrary temperature increment, the overall moduli are given by the average of the crystal compliances and thermal tensors:

$$\overline{\mathbf{C}}^{\mathrm{LB}} = \langle \mathbf{C}^{(g)^{-1}} \rangle^{-1} \quad \text{and} \quad \overline{\boldsymbol{\alpha}}^{\mathrm{LB}} = \langle \boldsymbol{\alpha}^{(g)} \rangle.$$
(10)

The uniform strain approach fulfills compatibility (but not necessarily equilibrium) within the polycrystal and represents an upper bound for the elastic energy of the aggregate. The uniform stress approach fulfills equilibrium (but not necessarily compatibility) in the aggregate and represents a lower bound for the energy. Neither approach fulfills the two conditions given by (7) and (8) simultaneously. In addition, the elastic constants and thermal expansion coefficients given by (9) and (10) are different because the average of the inverses does not coincide with the inverse of the average. Only for the trivial case of elastically and thermally isotropic crystals

are the tensors independent of grain orientation and both averaging procedures give the same results.

The estimation of the effective elastic stiffness $\overline{\mathbf{C}}$ and the effective thermal expansion $\overline{\boldsymbol{\alpha}}$ by means of the SC approach is described in Section 2.5. For this purpose, we first solve the problem of an elastic inclusion embedded in a homogeneous medium. Invoking Mura's [12] concept of equivalent inclusion, the constitutive behavior at the grain level can be rewritten in terms of the homogeneous macroscopic moduli by adding and subtracting $\overline{\mathbf{C}} : \boldsymbol{\epsilon}(\mathbf{x}) - \overline{\mathbf{C}} : \overline{\boldsymbol{\alpha}} \delta T$ to (3) to obtain

$$\boldsymbol{\sigma}(\mathbf{x}) = \overline{\mathbf{C}}: \left(\boldsymbol{\varepsilon}(\mathbf{x}) - \boldsymbol{\varepsilon}^*(\mathbf{x})\right) - \overline{\mathbf{C}}: \, \overline{\boldsymbol{\alpha}} \, \delta \, T, \tag{11}$$

where

$$\boldsymbol{\varepsilon}^*(\mathbf{x}) = \overline{\mathbf{C}}^{-1} \colon [(\mathbf{C}^{(g)} - \overline{\mathbf{C}}) \colon \boldsymbol{\varepsilon}(\mathbf{x}) - (\mathbf{C}^{(g)} \colon \boldsymbol{\alpha}^{(g)} - \overline{\mathbf{C}} \colon \overline{\boldsymbol{\alpha}})\delta T].$$
(12)

In this way, the inhomogeneous inclusion is replaced by an equivalent homogeneous inclusion with elastic stiffness \overline{C} and eigenstrain field $\varepsilon^*(\mathbf{x})$. Subtracting (5) from (11) gives

$$\tilde{\boldsymbol{\sigma}}(\mathbf{x}) = \overline{\mathbf{C}}: \, (\tilde{\boldsymbol{\varepsilon}}(\mathbf{x}) - \boldsymbol{\varepsilon}^*(\mathbf{x})), \tag{13}$$

where the symbol "~" denotes local deviations of the corresponding tensor from macroscopic values: $\tilde{\sigma}(\mathbf{x}) = \sigma(\mathbf{x}) - \overline{\sigma}$ and $\tilde{\boldsymbol{\varepsilon}}(\mathbf{x}) = \boldsymbol{\varepsilon}(\mathbf{x}) - \overline{\boldsymbol{\varepsilon}}$.

In the absence of external body forces, the equilibrium equation is

$$\sigma_{ij,j}(\mathbf{x}) = (\overline{\sigma}_{ij} + \widetilde{\sigma}_{ij}(\mathbf{x}))_{,j} = \widetilde{\sigma}_{ij,j}(\mathbf{x}) = 0.$$
(14)

Replacing (13) in (14), using the relation $\tilde{\varepsilon}_{ij}(\mathbf{x}) = (1/2)(\tilde{u}_{i,j}(\mathbf{x}) + \tilde{u}_{j,i}(\mathbf{x}))$, and accounting for the symmetry $\overline{C}_{ijkl} = \overline{C}_{ijlk}$, we get a set of partial differential equations for the local deviation of the displacement gradient field:

$$\overline{C}_{ijkl}\tilde{u}_{k,lj}(\mathbf{x}) + f_i^{\text{ext}}(\mathbf{x}) = 0,$$
(15)

where the artificial body force field associated with the heterogeneity of elastic and thermal constants is defined as

$$f_i^{\text{ext}}(\mathbf{x}) = -\overline{C}_{ijkl} \varepsilon_{kl,j}^*(\mathbf{x}) = \sigma_{ij,j}^*(\mathbf{x}).$$
(16)

The field $\sigma^*(\mathbf{x}) = -\overline{\mathbf{C}}$: $\varepsilon^*(\mathbf{x})$ is an eigenstress field, which is also known as polarization field.

2.2. Green function method

The system defined by (15) consists of three differential equations with three unknowns, namely, the components of the local deviation of the displacement vector field $\tilde{u}_i(\mathbf{x})$. A system of N linear differential equations with N unknown functions and an inhomogeneity term, such as (15), can be solved using the Green function method. Let us call $G_{km}(\mathbf{x} - \mathbf{x}')$ the Green function associated with $u_i(\mathbf{x})$ of a homogeneous infinite medium with elastic stiffness $\overline{\mathbf{C}}$ that gives, in the absence of any other sources of strain, the displacement at position \mathbf{x} along direction k induced by a unit external body force, with a single non-vanishing component m, applied at \mathbf{x} :

$$\overline{C}_{ijkl}G_{km,lj}(\mathbf{x}-\mathbf{x}') + \delta_{im}\delta(\mathbf{x}-\mathbf{x}') = 0,$$
(17)

where $\delta(\mathbf{x} - \mathbf{x}')$ is the Dirac delta function and δ_{im} is the Kronecker delta function. Once the solution of (17) is obtained, the solution of (15) is given by the convolution integral, that is, a linear superposition of elementary contributions to the displacement field given by the Green function:

$$u_k(\mathbf{x}) = \int_{R^3} G_{km}(\mathbf{x} - \mathbf{x}') f_m^{\text{ext}}(\mathbf{x}') \, \mathrm{d}\mathbf{x}'.$$
 (18)

Assuming zero macroscopic strain, the same expression holds for $\tilde{u}_i(\mathbf{x})$:

$$\tilde{u}_k(\mathbf{x}) = \int_{R^3} G_{km}(\mathbf{x} - \mathbf{x}') f_m^{\text{ext}}(\mathbf{x}') \, \mathrm{d}\mathbf{x}'.$$
(19)

Taking the Fourier transform of (17),

$$\overline{C}_{ijkl}FT[G_{km,lj}(\mathbf{x}-\mathbf{x}')] + \delta_{im}FT[\delta(\mathbf{x}-\mathbf{x}')] = 0,$$
(20)

and applying the properties of the Fourier transform of the derivative and of the Dirac delta function,

$$\overline{C}_{ijkl}i^2k_lk_j\hat{G}_{km}(\mathbf{k}) + \delta_{im} = 0$$
⁽²¹⁾

where **k** is a point/vector of the 3D Fourier space of angular frequencies and $i = \sqrt{-1}$ is the imaginary unit. Denoting **k** = $k\alpha$, where the scalar k and the vector α (this notation is only used here and in the next section, and it should not be confused with the thermal expansion tensors α_{ij} used elsewhere) are the modulus and the unit vector associated with **k**, respectively, we obtain

$$\overline{C}_{ijkl}\alpha_l\alpha_j[k^2\hat{G}_{km}(\mathbf{k})] = \delta_{im}.$$
(22)

Defining

$$A_{ik}(\boldsymbol{\alpha}) = \alpha_j \alpha_l \overline{C}_{ijkl} \tag{23}$$

allows us to obtain the algorithmic expression to calculate the Fourier transform of the Green function:

$$k^2 \hat{G}_{ij}(\boldsymbol{\alpha}) = A_{ij}^{-1}(\boldsymbol{\alpha}). \tag{24}$$

Since the matrix A_{ij} is a real function of $\boldsymbol{\alpha}$, so is $k^2 \hat{G}_{ij}$. This property leads to real integrals in the derivation that follows.

2.3. Elastic inclusion and Eshelby tensor

Replacing the eigenstress given by (16) in (19) and taking derivatives yield for the local deviation of the displacement gradient field

$$\tilde{u}_{k,l}(\mathbf{x}) = \int_{\mathbb{R}^3} G_{ki,l}(\mathbf{x} - \mathbf{x}') \sigma^*_{ij,j}(\mathbf{x}') \, \mathrm{d}\mathbf{x}'.$$
(25)

Using the relation $\partial G_{ij}(\mathbf{x} - \mathbf{x}')/\partial \mathbf{x} = -\partial G_{ij}(\mathbf{x} - \mathbf{x}')/\partial \mathbf{x}'$, we can rewrite the integrand of (25) as $G_{ki,l}(\mathbf{x} - \mathbf{x}')\sigma_{ij,j}^*(\mathbf{x}') = (G_{ki,l}(\mathbf{x} - \mathbf{x}')\sigma_{ij}^*(\mathbf{x}'))_{,j} + G_{ki,lj}(\mathbf{x} - \mathbf{x}')\sigma_{ij}^*(\mathbf{x}')$ for integration by parts. Using the Gauss theorem and the vanishing character of the Green function at infinity, we obtain [12]

$$\tilde{u}_{k,l}(\mathbf{x}) = \int_{\mathbb{R}^3} G_{ki,jl}(\mathbf{x} - \mathbf{x}') \sigma_{ij}^*(\mathbf{x}') \,\mathrm{d}\mathbf{x}'.$$
(26)

Equation (26) provides an exact implicit solution to the problem of a general eigenstrain field $\boldsymbol{\varepsilon}^*(\mathbf{x})$ being accommodated elastically and inducing an eigenstress field $\boldsymbol{\sigma}^*(\mathbf{x})$. Such a solution requires knowing the local dependence of the eigenstrain or the eigenstress tensor. Here, we are interested in the specific application to thermo-elastic aggregates, where the elastic stiffness $\mathbf{C}^{(g)}$, the thermal expansion tensor $\boldsymbol{\alpha}^{(g)}$, and the thermal dilatation $\boldsymbol{\alpha}^{(g)}\delta T$ are constant over the domain of each grain. This leads to the eigenstrain $\boldsymbol{\varepsilon}^{*(g)}$ adopting a constant value in the domain of the inclusion and being zero in the domain of the effective medium. Under these conditions, Eshelby [13] and Mura [12] demonstrated that for a single inclusion of ellipsoidal shape embedded in an infinite medium, the elastic strain field is uniform in the domain of the inclusion. Here, however, we follow the more general approach of Berveiller *et al.* [14], expressing the average of the local deviation of the displacement gradient field given by (25) over the domain Ω^g of the grain:

$$\tilde{u}_{k,l}^{(g)} = \left(-\frac{1}{\Omega^g} \int_{\Omega^g} \int_{\Omega^g} G_{kl,jl}(\mathbf{x} - \mathbf{x}') \, \mathrm{d}\mathbf{x} \, \mathrm{d}\mathbf{x}' \right) \overline{C}_{ijmn} \varepsilon_{mn}^{*(g)}.$$
(27)

Equation (27) can be interpreted in two ways. If the effective elastic modulus **C** is assumed to be the stiffness of an infinite homogeneous matrix in which an ellipsoidal inclusion with constant eigenstrain $\boldsymbol{\epsilon}^{*(g)}$ is embedded, $\tilde{u}_{k,l}^{(g)}$ is the constant value inside domain Ω^{g} obtained from the

solution of Eshelby's inclusion problem. On the other hand, in the actual case of a polycrystal, $\tilde{u}_{k,l}^{(g)}$ should be interpreted as the average value (first moment) of the corresponding field in grain (g).

Expressing the second derivative of the Green tensor in terms of its Fourier transform, we obtain

$$\widetilde{u}_{k,l}^{(g)} = \left(\frac{1}{8\pi^3 \Omega^g} \int_{\Omega^g} \int_{\Omega^g} \int_{K^3} (\alpha_l \alpha_j k^2 \widehat{G}_{ki}(\mathbf{k})) \exp[-i\mathbf{k} \cdot (\mathbf{x} - \mathbf{x}')] \, \mathrm{d}\mathbf{k} \, \mathrm{d}\mathbf{x} \, \mathrm{d}\mathbf{x}'\right) \overline{C}_{ijmn} \varepsilon_{mn}^{*(g)}
= T_{klij} \overline{C}_{ijmn} \varepsilon_{mn}^{*(g)}.$$
(28)

Transforming the integral over the reciprocal space to spherical coordinates such that $d\mathbf{k} = \sin\theta \, d\theta \, d\phi \, k^2 \, dk$ and using (22), the tensor T_{klij} can be rewritten as

$$T_{klij} = \frac{1}{8\pi^3 \Omega^g} \int_0^{2\pi} \int_0^{\pi} \alpha_l \alpha_j A_{ki}^{-1}(\boldsymbol{\alpha}) \Lambda(\boldsymbol{\alpha}) \sin\theta \, \mathrm{d}\theta \, \mathrm{d}\varphi, \tag{29}$$

where θ and ϕ are the spherical coordinates of the Fourier unit vector $\boldsymbol{\alpha}$ and

$$\Lambda(\boldsymbol{\alpha}) = \int_0^\infty \left(\int_{\Omega^g} \int_{\Omega^g} \exp[-ik(\mathbf{x} - \mathbf{x}')] \, \mathrm{d}\mathbf{x} \, \mathrm{d}\mathbf{x}' \right) k^2 \, \mathrm{d}k. \tag{30}$$

Integration of (30) inside an ellipsoidal domain Ω^g with main axes (a, b, c) gives [14]

$$\Lambda(\boldsymbol{\alpha}) = \frac{8\pi^3}{3} \frac{(abc)^2}{[\rho(\boldsymbol{\alpha})]^3},\tag{31}$$

where

$$\rho(\alpha) = \sqrt{(a\alpha_1)^2 + (b\alpha_2)^2 + (c\alpha_3)^2}.$$
(32)

Replacing (31) in (29), and recalling that $\Omega^g = (4/3)\pi(abc)$, provides an expression of T_{klij} for an ellipsoidal grain:

$$T_{klij} = \frac{(abc)}{4\pi} \int_0^{2\pi} \int_0^{\pi} \frac{\alpha_j \alpha_l A_{ki}^{-1}(\boldsymbol{\alpha})}{[\rho(\boldsymbol{\alpha})]^3} \sin\theta \, \mathrm{d}\theta \, \mathrm{d}\varphi.$$
(33)

For arbitrary elastic anisotropy, the double integral in (33) has to be solved numerically using, for instance, a Gauss–Legendre technique [5]. For the purpose of calculating the symmetric strain tensor, the following symmetric Eshelby tensor is defined:

$$S_{ijkl} = \frac{1}{4}(T_{ijmn} + T_{jimn} + T_{ijnm} + T_{jinm})\overline{C}_{mnkl}.$$
(34)

2.4. Interaction and localization equations

Taking the symmetric part of (28), and using the Eshelby tensor defined in (34), we obtain the average deviation of strain in grain (g) with respect to the macroscopic strain:

$$\tilde{\boldsymbol{\varepsilon}}^{(g)} = \mathbf{S} : \boldsymbol{\varepsilon}^{*(g)}. \tag{35}$$

An expression equivalent to (13), relating local deviations in stress, strain, and eigenstrain fields, also holds for the deviations of average stress, average strain, and average eigenstrain in the grains with respect to the effective medium. It is straightforward to demonstrate this by integrating (13) over the grain domain:

$$\tilde{\boldsymbol{\sigma}}^{(g)} = \overline{\mathbf{C}} : (\tilde{\boldsymbol{\varepsilon}}^{(g)} - {\boldsymbol{\varepsilon}}^{*(g)}).$$
(36)

Replacing the eigenstrain given by (35) in (36), the following interaction equation is obtained:

$$\tilde{\boldsymbol{\sigma}}^{(g)} = -\tilde{\mathbf{C}} : \tilde{\boldsymbol{\varepsilon}}^{(g)}, \tag{37}$$

where the interaction tensor is defined as

$$\tilde{\mathbf{C}} = \overline{\mathbf{C}} : (\mathbf{I} - \mathbf{S}) : \mathbf{S}^{-1}.$$
(38)

Using the polycrystal constitutive relation (5) and the grain-averaged constitutive relation obtained from averaging (3) in the domain of grain (g),

$$\boldsymbol{\sigma}^{(g)} = \mathbf{C}^{(g)} : \boldsymbol{\varepsilon}^{(g)} - \mathbf{C}^{(g)} : \boldsymbol{\alpha}^{(g)} \delta T,$$
(39)

the interaction equation (37) can be rewritten in terms of total strain tensors. After some algebraic manipulation, the following *strain localization equation* is obtained:

$$\boldsymbol{\varepsilon}^{(g)} = \mathbf{A}^{(g)} \colon \overline{\boldsymbol{\varepsilon}} + \mathbf{D}^{(g)} \delta T, \tag{40}$$

where the strain localization tensors are defined as

$$\mathbf{A}^{(g)} = (\mathbf{C}^{(g)} + \tilde{\mathbf{C}})^{-1} \colon (\overline{\mathbf{C}} + \tilde{\mathbf{C}}), \tag{41}$$

$$\mathbf{D}^{(g)} = (\mathbf{C}^{(g)} + \tilde{\mathbf{C}})^{-1} \colon (\mathbf{C}^{(g)} \colon \boldsymbol{\alpha}^{(g)} - \overline{\mathbf{C}} \colon \overline{\boldsymbol{\alpha}}).$$
(42)

2.5. Self-consistent equations

The derivation presented in the previous sections solves the problem of a thermo-elastic inclusion embedded in a thermo-elastic effective medium being subject to external loading conditions. In this section, we use the previous result to construct a polycrystal model, consisting in regarding each grain as an ellipsoidal inclusion embedded in a homogeneous medium with the same properties as those of the homogenized polycrystal. The properties of such a medium are not known *a priori*, and they will be obtained following an iterative SC procedure. The corresponding equations are derived in this section.

The homogenized medium assumption is central to our derivation and needs some discussion. Even if the solution for each individual ellipsoid representing a grain embedded in a homogeneous medium is exact, in a real aggregate, grains are not ellipsoidal and are not surrounded by a homogeneous continuum. Rather, the surrounding neighbors have different orientations and induce stress gradients inside the grains and across grain boundaries. In addition, grains with the same orientation in the aggregate are surrounded by different neighbors, and as a consequence, their associated average stress is not the same. As a result, the grain stress and strain given by the effective medium approximation have to be regarded as representing the average state of all grains having the same orientation in the aggregate.

We use the localization equation for the strain (40) to derive two equations. The first equation follows from replacing (40) in (39):

$$\boldsymbol{\sigma}^{(g)} = \mathbf{C}^{(g)} : \boldsymbol{\varepsilon}^{(g)} - \mathbf{C}^{(g)} : \boldsymbol{\alpha}^{(g)} \delta T = \mathbf{C}^{(g)} : \mathbf{A}^{(g)} : \overline{\boldsymbol{\varepsilon}} + (\mathbf{C}^{(g)} : \mathbf{D}^{(g)} - \mathbf{C}^{(g)} : \boldsymbol{\alpha}^{(g)}) \delta T.$$
(43)

The second equation follows from taking the average of (40) over all grains:

$$\langle \boldsymbol{\varepsilon}^{(g)} \rangle = \langle \mathbf{A}^{(g)} \rangle \colon \overline{\boldsymbol{\varepsilon}} + \langle \mathbf{D}^{(g)} \rangle \delta T.$$
(44)

We recall that $\langle \rangle$ denotes the average over the representative grains weighted by the associated grain volume fraction. Extracting the effective strain of the medium $\overline{\boldsymbol{\epsilon}} = \langle \boldsymbol{\epsilon}^{(g)} \rangle$ from (44), replacing it in (43), and averaging the grain stresses give

$$\langle \boldsymbol{\sigma}^{(g)} \rangle = \langle \mathbf{C}^{(g)} : \mathbf{A}^{(g)} \rangle : (\langle \mathbf{A}^{(g)} \rangle^{-1} : \langle \boldsymbol{\varepsilon}^{(g)} \rangle - \langle \mathbf{A}^{(g)} \rangle^{-1} : \langle \mathbf{D}^{(g)} \rangle \delta T) + \langle \mathbf{C}^{(g)} \mathbf{D}^{(g)} - \mathbf{C}^{(g)} : \boldsymbol{\alpha}^{(g)} \rangle \delta T.$$
(45)

Enforcing the condition that the effective stress is equal to the average of the grain stresses $\overline{\sigma} = \langle \sigma^{(g)} \rangle$,

$$\overline{\boldsymbol{\sigma}} = \overline{\mathbf{C}} \colon \overline{\boldsymbol{\varepsilon}} - \overline{\mathbf{C}} \colon \overline{\boldsymbol{\alpha}} \delta T = \langle \boldsymbol{\sigma}^{(g)} \rangle, \tag{46}$$

leads to

$$\overline{\mathbf{C}}: \overline{\boldsymbol{\epsilon}} - \overline{\mathbf{C}}: \overline{\boldsymbol{\alpha}} \delta T = \langle \mathbf{C}^{(g)}: \mathbf{A}^{(g)} \rangle : \langle \mathbf{A}^{(g)} \rangle^{-1}: \overline{\boldsymbol{\epsilon}} - \{ \langle \mathbf{C}^{(g)}: \mathbf{A}^{(g)} \rangle : \langle \mathbf{A}^{(g)} \rangle^{-1}: \langle \mathbf{D}^{(g)} \rangle - \mathbf{C}^{(g)}: \mathbf{D}^{(g)} + \mathbf{C}^{(g)}: \boldsymbol{\alpha}^{(g)} \} \delta T.$$
(47)

For this identity to hold, the strain and temperature factors on the right-hand side have to coincide with those on the left-hand side. This condition leads to

$$\overline{\mathbf{C}} = \langle \mathbf{C}^{(g)} \colon \mathbf{A}^{(g)} \rangle \colon \langle \mathbf{A}^{(g)} \rangle^{-1}, \tag{48}$$

which is an implicit equation for the stiffness because the localization tensor $\mathbf{A}^{(g)}$ is a function of the interaction tensor $\mathbf{\tilde{C}}^{(g)}$ (38), which in turn is a function of $\mathbf{\bar{C}}$ and the Eshelby tensor \mathbf{S} , which is also a function of $\mathbf{\bar{C}}$. As a consequence, Equation (48) is a fixed-point equation for the effective polycrystal's elastic stiffness.

The condition on the term associated with a temperature change is

$$\overline{\mathbf{C}}: \overline{\boldsymbol{\alpha}} = \langle \mathbf{A}^{\dagger(g)}: \mathbf{D}^{(g)} \rangle + \langle \mathbf{C}^{(g)}: \boldsymbol{\alpha}^{(g)} \rangle, \tag{49}$$

where

$$\mathbf{A}^{\dagger(g)} = (\overline{\mathbf{C}} + \widetilde{\mathbf{C}}^{(g)}) : (\mathbf{C}^{(g)} + \widetilde{\mathbf{C}}^{(g)})^{-1}.$$
(50)

Using (42) for $\mathbf{D}^{(g)}$ leads, after some algebraic manipulation, to a direct equation giving the effective thermal coefficients of the aggregate:

$$\overline{\mathbf{C}}: \overline{\boldsymbol{\alpha}} = \langle \mathbf{A}^{\dagger(g)} \rangle^{-1}: \langle \mathbf{A}^{\dagger(g)}: \mathbf{C}^{(g)}: \boldsymbol{\alpha}^{(g)} \rangle.$$
(51)

The iterative solution of (48) gives the polycrystal's elastic stiffness, and the subsequent use of (51) gives the polycrystal's thermal coefficients once the effective stiffness is known.

We end this section mentioning that by taking derivatives with respect to the local moduli of the effective properties given by (48) and (51), it is possible to access information on intragranular heterogeneity in the form of second moments of the stress field distribution in each mechanical phase (*g*). The algorithms to calculate these second moments from the SC theory for thermoelastic polycrystals (based on the original TESC expressions for composites [15,16]) can be found in [17–19].

3. Applications

The applications discussed in this section include the calculation of effective elastic and thermal moduli of zirconium, beryllium, and uranium and the evolution of macroscopic strain and internal stresses during cooling processes.

3.1. Cooling of zirconium

The purpose of this example is to show how texture affects the effective elastic and thermal constants and also the evolution of internal stress during cooling. We simulate cooling of Zr from 900 to 300 K for two different crystal orientation distributions: a non-textured aggregate (see discussion in the next paragraph) and an aggregate with axisymmetric texture, typical of an extruded bar. The axisymmetric texture is displayed in Figure 1, and the single-crystal elastic and thermal constants (or coefficients of thermal expansion) are given in Tables 1 [20] and 2 [21], respectively. In this simulation, the boundary condition is that the macroscopic stress components are zero, and at 900 K, the grains in the aggregate are assumed to be stress-free.

The UB, LB, and TESC elastic constants calculated using 1000 orientations from a Sobol sequence [22], obtained by maximizing the separation (in Euler space) of the next orientation in the sequence from all the previous orientations, are given in Table 1. The effective thermal moduli are given in Table 2. Observe that the effective properties may deviate slightly from the exact isotropic conditions ($\overline{C}_{11} = \overline{C}_{22} = \overline{C}_{33}$ and $\overline{\alpha}_1 = \overline{\alpha}_2 = \overline{\alpha}_3$); for example, note some small fluctuations in the least significant digits in the predicted values for non-textured aggregates. The reason is that despite the aforementioned Sobol sequence optimizing the generation of a
	C_{11}	C_{22}	C ₃₃	C_{23}	C_{13}	C_{12}	C_{44}	C_{55}	C_{66}
Crystal	143.5	143.5	164.9	65.4	65.4	72.5	32.1	32.1	35.5
		ľ	Non-tex	tured a	ıggrega	ite			
UB	144.1	144.1	144.1	71.1	71.0	71.1	36.5	36.5	36.5
TESC	143.5	143.5	143.5	71.2	71.2	71.2	36.1	36.1	36.1
LB	142.9	142.9	142.9	71.4	71.4	71.4	35.8	35.8	35.8
			Textu	red agg	regate				
UB	146.9	146.5	142.4	70.0	69.9	71.3	35.0	34.9	37.8
TESC	145.9	145.7	142.2	70.2	70.1	71.6	34.8	34.7	37.2
LB	145.1	144.8	142.0	70.4	70.3	71.9	34.6	34.6	36.7

Table 1. Zr single-crystal elastic constants [20] and polycrystal elastic constants predicted using UB, LB, and TESC models for non-textured and extrusion-texture aggregates (Voigt notation; units of [GPa])

Table 2. Zr single-crystal thermal constants [21] and polycrystal thermal constants predicted using UB, LB, and TESC models for non-textured and extrusion-texture aggregates (Voigt notation; units of $[10^{-6} \text{ K}^{-1}]$)

	α_1	α_2	α_3	α_1	α_2	α_3	
Crystal	5.7	5.7	10.5	5.7	5.7	10.5	
	No	n-texti	ured	Textured			
UB	7.28	7.29	7.28	8.09	8.02	5.71	
TESC	7.23	7.23	7.23	7.98	7.91	5.87	
LB	7.23	7.23	7.23	7.87	7.82	6.00	

discrete set of orientations to a uniform distribution, which should produce the same effective response as that for a very large set of random orientations, the set of 1000 orientations used here for representing the aggregate is not strictly uniform. In what follows, the polycrystal represented by these 1000 orientations is referred to as "non-textured aggregate."

Since Zr is mildly anisotropic elastically, the stiffness components of the textured aggregate are not very different from those of the non-textured aggregate, but they display a clear axial symmetry, that is, $C_{11} \cong C_{22} \neq C_{33}$, $C_{13} \cong C_{23} \neq C_{12}$, and $C_{44} \cong C_{55} \neq C_{66}$. On the other hand, the large thermal anisotropy of the single-crystal thermal coefficients makes the effective properties strongly dependent on texture. As we show next, it also has an important effect on the internal stress evolution during cooling.

Figure 2 displays the evolution of overall diagonal strain components as a function of decreasing temperature predicted using TESC. As is to be expected, for the non-textured aggregate, all three directions contract by the same amount; but for the textured aggregate, this is not the case. A concentration of basal poles perpendicular to the axial direction of the bar, combined with a larger thermal coefficient along the *c*-axis, leads to a larger contraction perpendicular to the bar axis. This is consistent with the effective thermal coefficients of the aggregate reported in Table 2.

Figure 2 also shows the evolution with temperature of the strain normal to basal (0002) and prismatic (10–10) planes either perpendicular to axis 1 (radial direction) or axis 3 (axial direction) of the texture. In the case of the non-textured aggregate, both directions are equivalent, and the result reflects the fact that the tendency of the crystal is to contract more than the effective medium along the *c*-axis and less than the effective medium along the *a*-axis. The interaction of the grain (inclusion) with the effective medium partially prevents such behavior, and this leaves



Figure 1. Basal pole figure of extruded Zr bar.



Figure 2. Microstrain evolution in (10–10) and (0002) planes and macroscopic strain evolution with temperature in non-textured Zr and extruded Zr bar when cooling from 900 K to 300 K, predicted using TESC.

the *c*-axis in tension and the *a*-axis in compression. The same mechanism applies to internal stress evolution in the textured polycrystal except that now the average contraction is different along the axial and radial directions and so is the interaction with grains oriented in either of these directions. The TESC model captures well such a directional interaction. Observe that the final elastic strains are large $(~1 \times 10^{-3})$ and correspond to normal stresses of order 100 MPa. The larger thermal coefficient perpendicular to the basal plane induces a larger thermal contraction, which is counteracted by the interaction with the surrounding medium. As a consequence, the strain along the *c*-axis is always positive. The opposite is true for the direction normal to the prismatic planes of the crystal. Texture changes the properties of the effective medium and affects the result quantitatively. These predictions are consistent with measurements in a Zrly-2 bar reported by MacEwen *et al.* [21].

3.2. Cooling of beryllium

To show the relevance of the single-crystal anisotropic moduli, here we perform the same calculations for another hcp material, beryllium, and compare the results against those for zirconium. For this purpose, we simulate cooling of Be from 900 to 300 K for a non-textured aggregate. The single-crystal elastic and thermal constants are given in Tables 3 [20] and 4 [23], respectively. As before, the boundary condition is zero macroscopic stress components and an internal stress-free aggregate at 900 K. The latter is in reasonable agreement with the internal stresses measured by Brown *et al.* [24] as a function of temperature in a non-textured aggregate.

88	7
----	---

	C_{11}	C_{22}	C_{33}	C_{23}	C_{13}	C_{12}	C_{44}	C_{55}	C_{66}	
Crystal	292.3	292.3	336.4	14.0	14.0	26.7	162.5	162.5	132.8	
			Non-textured aggregate							
UB	313.6	313.6	313.6	14.9	15.9	14.9	149.3	149.3	149.3	
TESC	312.5	312.6	312.5	15.3	15.3	15.3	148.6	148.6	148.6	
LB	311.5	311.3	311.5	15.6	15.6	15.6	148.0	148.0	148.0	

Table 3. Be single-crystal elastic constants [20] and polycrystal elastic constants predicted using UB, LB, and TESC models for a non-textured aggregate (Voigt notation; units of [GPa])

Table 4. Be single-crystal thermal constants at 450 K [23] and polycrystal thermal constants predicted using UB, LB, and TESC models for a non-textured aggregate (Voigt notation; units of $[10^{-6} \text{ K}^{-1}]$)

	α_1	α_2	α_3
Crystal	13.9	13.9	10.4
	Noi	n-textu	ired
UB	12.7	12.7	12.7
TESC	12.7	12.7	12.7
LB	12.7	12.7	12.7

Beryllium is a peculiar hcp material in two aspects: it exhibits an unusually low Poisson modulus ($\nu \sim 0.045$; varies depending on the crystal direction) and its thermal expansion coefficients have been reported to increase two-fold from 300 to 900 K [23]. For the purpose of this demonstration, we have made no attempt at accounting for the variation in thermal coefficients with temperature and have adopted the coefficients corresponding to 450 K. Another difference from Zr is that thermal contraction along the *c*-axis is smaller than perpendicular to it since $\alpha_{33} < \alpha_{11}$.

The UB, LB, and TESC polycrystal elastic constants are given in Table 3 and the corresponding polycrystal thermal moduli are given in Table 4.

Figure 3 displays the evolution of overall diagonal strain components as a function of decreasing temperature predicted using TESC. As is to be expected for a non-textured aggregate, all three directions contract by the same amount, about 0.8% at room temperature. Figure 3 also shows the evolution with temperature of the strain normal to basal (0002) and prismatic (10–10) planes, and the result reflects the tendency of the crystal to contract less than the effective medium along the *c*-axis and more than the effective medium along the *a*-axis. The interaction of the grain (inclusion) with the effective medium opposes such a trend, and this leaves the *c*-axis in compression and the *a*-axis in tension, which is opposite to what happens in Zr. The TESC model captures such a directional interaction and predicts the final crystallographic elastic strains of ~0.5 × 10⁻³, which correspond to large normal stresses of order ~100 MPa in prism planes and of ~-200 MPa in basal planes (Figure 3). These values are in good agreement with the measurements reported by Brown *et al.* [24]. Although the internal stresses are somewhat smaller in a textured aggregate, they are expected to play an important role in early yielding, stress differential, and elastoplastic transition when a Be aggregate is subjected to mechanical loading.

3.3. Cooling of uranium

As in the previous cases, the purpose of this example is to show how texture affects the effective elastic and thermal constants, and the evolution of internal stress, during cooling of α -U. In α -U,



Figure 3. Microstrain evolution in (10–10) and (0002) planes and macroscopic strain evolution with temperature predicted using TESC for non-textured Be when cooling from 900 to 300 K (left); evolution of stress normal to (10–10) and (0002) planes (right).



Figure 4. (100), (010), and (001) poles of rolled α -uranium.

the elastic and thermal constants are more anisotropic than those of Zr, and the single-crystal symmetry is orthotropic instead of hexagonal.

As before, to evidence texture effects, we simulate the cooling of α -U from 900 to 300 K for two different crystal orientation distributions: a non-textured aggregate and an aggregate with rolling texture, see Figure 4. The single-crystal elastic and thermal coefficients are reported in Tables 5 and 6, respectively. The evolution of internal strains on {100}, {010}, and {001} planes along directions x_1 (rolling), x_2 (transverse), and x_3 (normal) is also reported. The simulation imposes all the macroscopic stress components to be zero while cooling from 900 to 300 K.

The UB, LB, and TESC elastic constants calculated for the non-textured and textured aggregates are given in Table 5 and the effective thermal moduli are given in Table 6. The large anisotropy of the α -U single-crystal thermal coefficients makes the effective properties strongly dependent on texture. In addition, as shown in Figure 5 (and also in Figure 2 for Zr), the macroscopic strains induced by cooling under the condition of zero macroscopic stress are contractive. The grains, however, develop increasingly compressive and tensile components of stress and elastic strain that average to zero.

The larger thermal coefficients along the [100] and [001] crystal directions, compared with [010], induce a larger thermal contraction perpendicular to those planes, which is counteracted by the interaction with the surrounding medium. As a consequence, the associated internal strains are always positive. The opposite is true for the [010] crystal direction, which has a nearly zero thermal coefficient. Texture changes the properties of the effective medium and affects the results quantitatively but not qualitatively.



Figure 5. Evolution of microstrains in {100}, {010}, and {001} planes and evolution of macroscopic strain components in non-textured and rolling-texture α -U when cooling from 900 to 300 K.

Table 5. α -uranium single-crystal elastic constants [25] and polycrystal elastic constants predicted using UB, LB, and TESC models for a non-textured and a rolling-texture aggregate (Voigt notation; units of [GPa])

	C_{11}	C_{22}	C ₃₃	C_{23}	C_{13}	C_{12}	C_{44}	C_{55}	C_{66}
Crystal	214.8	198.6	267.1	107.6	21.8	46.5	124.4	73.4	74.3
			Non-te	xtured a	ıggrega	ite			
UB	232.2	232.3	232.1	55.9	56.1	55.9	88.0	88.1	87.9
SC	225.1	225.2	225.1	56.7	56.7	56.6	84.1	84.2	84.1
LB	219.0	219.1	219.0	57.6	57.7	57.5	80.6	80.7	80.6
			Textu	ured agg	regate				
UB	214.0	224.3	241.3	52.3	65.4	58.6	92.3	94.5	85.8
SC	208.5	219.0	233.7	52.8	65.7	59.2	88.7	90.2	81.9
LB	203.8	214.5	227.0	53.5	66.1	59.9	85.5	86.3	78.5

Table 6. α -uranium single-crystal thermal constants [26]. Polycrystal thermal constants predicted using UB, LB, and TESC models for a non-textured and a rolling-texture aggregate (Voigt notation; units of $[10^{-6} \text{ K}^{-1}]$)

	α_1	α_2	α_3	α_1	α_2	α_3	
Single crystal	25.41	0.65	20.65	25.41	0.65	20.65	
	No	n-textu	red	Textured			
UB	15.11	15.15	15.11	10.64	17.17	17.44	
SC	15.38	15.41	15.37	10.01	17.46	18.61	
LB	15.56	15.60	15.55	9.21	17.64	19.86	

4. Conclusions

In this paper, the TESC formulation is presented in detail and applied to the cooling of polycrystalline single-phase non-cubic metals. The TESC model captures the effect of crystallographic texture and single-crystal elastic and thermal anisotropy on effective and local thermo-elastic responses. Although not shown here, the TESC model can be applied to multiphase polycrystalline aggregates. In such cases, the heterogeneity in the local thermal response captured by the model arises from single-crystal thermal anisotropy (i.e. for non-cubic phases) and/or the difference in thermal properties between phases (including cubic phases).

The results presented here show that depending on temperature change, texture of the aggregate, and level of single-crystal elastic and thermal anisotropy, internal stresses at the grain level can be significant even in the absence of applied stress, for example, cooling under zero external stresses, as in all the cases presented. These internal stresses may play an important role in promoting plastic yield during cooling or during loading after annealing as well as in determining subsequent stress differential, for example, different yield in tension versus compression. In other words, these thermal stresses can significantly affect yielding and the elastoplastic transition compared with the case of aggregates with stress-free grains. Evidently, the TESC formulation presented here is not able to capture this effect by itself, but it needs to be combined with model extensions that include crystal plasticity mechanisms such as elastoplastic [27] and elastoviscoplastic [28, 29] formulations.

References

- A. V. Hershey, "The elasticity of an isotropic aggregate of anisotropic cubic crystals", J. Appl. Mech.-Trans. ASME 21 (1954), p. 236.
- [2] E. Kröner, "Berechnung der elastischen Konstanten des Vielkristalls aus den Konstanten des Einkristalls", Z. Phys. 15 (1958), p. 504.
- [3] R. Hill, "Continuum micro-mechanics of elastoplastic polycrystals", J. Mech. Phys. Solids 13 (1965), p. 89.
- [4] R. Zeller, P. H. Dederichs, "Elastic constants of polycrystals", Phys. Status Solidi (b) 55 (1973), p. 831.
- [5] C. N. Tomé, R. A. Lebensohn, "ViscoPlastic Self-Consistent (VPSC) code", Los Alamos National Laboratory.
- [6] L. J. Walpole, "On the overall elastic moduli of composite materials", J. Mech. Phys. Solids 17 (1969), p. 235.
- [7] J. R. Willis, "Variational and related methods for the overall properties of composites", *Adv. Appl. Mech.* 21 (1981), p. 1.
- [8] V. A. Buryachenko, "Multiparticle effective field and related methods in micromechanics of random structure composites", *Math. Mech. Solids* 6 (2001), p. 577.
- [9] G. W. Milton, The Theory of Composites, Cambridge University Press, 2002.
- [10] C. N. Tomé, N. Christodoulou, P. A. Turner, M. A. Miller, C. H. Woo, J. Root, T. M. Holden, "Role of internal stresses in the transient of irradiation growth of Zircaloy-2", J. Nucl. Mater. 227 (1996), p. 237.
- [11] R. Hill, "The essential structure of constitutive laws for metal composites and polycrystals", J. Mech. Phys. Solids 15 (1967), p. 79.
- [12] T. Mura, *Micromechanics of Defects in Solids*, 2nd ed., Martinus-Nijhoff Publishers, Dordrecht, The Netherlands, 1987.
- [13] J. D. Eshelby, "The determination of the elastic field of an ellipsoidal inclusion, and related problems", Proc. R. Soc. Lond. A 241 (1957), p. 366.
- [14] M. Berveiller, O. Fassi-Fehri, A. Hihi, "The problem of two plastic and heterogeneous inclusions in an anisotropic medium", *Int. J. Eng. Sci.* 25 (1987), p. 691.
- [15] M. Bobeth, G. Diener, "Static elastic and thermoelastic fluctuations in multiphase composites", J. Mech. Phys. Solids 35 (1987), p. 37.
- [16] W. Kreher, "Residual stresses and stored elastic energy of composites and polycrystals", J. Mech. Phys. Solids 38 (1990), p. 115.
- [17] Y. Liu, "Macroscopic behavior, fluctuations and texture evolution in viscoplastic polycrystals", PhD Thesis, University of Pennsylvania, USA, 2003.
- [18] R. Brenner, O. Castelnau, L. Badea, "Mechanical field fluctuations in polycrystals estimated by homogenization techniques:", *Proc. R. Soc. Lond. A* **460** (2004), p. 3589.
- [19] R. A. Lebensohn, C. N. Tomé, P. P. Castañeda, "Self-consistent modeling of the mechanical behavior of viscoplastic polycrystals incorporating intragranular field fluctuations", *Phil. Mag.* 87 (2007), p. 4287.
- [20] G. Simmons, H. Wang, Single Crystal Elastic Constants and Calculated Aggregate Properties: A Handbook, MIT Press, 1971.
- [21] S. R. MacEwen, C. N. Tomé, J. Faber Jr., "Residual stresses in annealed Zircaloy", Acta Metall. 37 (1989), p. 979.
- [22] O. Castelnau, D. K. Blackman, R. A. Lebensohn, P. P. Castañeda, "Micromechanical modelling of the viscoplastic behavior of olivine", J. Geophys. Res. B 113 (2008), p. B09202.

- [23] P. Gordon, "A high temperature precision X-ray camera. Some measurements of the thermal coefficients of expansion of Beryllium", J. Appl. Phys. 20 (1949), p. 908.
- [24] D. W. Brown, T. A. Sisneros, B. Clausen, S. Abeln, M. A. M. Bourke, B. G. Smith, M. L. Steinzig, C. N. Tomé, S. C. Vogel, "Development of intergranular thermal residual stresses in Beryllium during cooling from processing temperatures", *Acta Mater.* 57 (2009), p. 972.
- [25] E. S. Fisher, "Temperature dependence of the elastic moduli in α -uranium single crystals", *J. Nucl. Mater.* **18** (1966), p. 39.
- [26] L. T. Lloyd, C. S. Barrett, "Thermal expansion of *α*-uranium", *J. Nucl. Mater.* **18** (1966), p. 55.
- [27] P. A. Turner, C. N. Tomé, "A study of residual stresses in Zircaloy-2 with rod texture", *Acta Metall. Mater.* 42 (1994), p. 4143.
- [28] Y. Jeong, C. N. Tomé, "Extension of the visco-plastic self-consistent model to account for elasto-visco-plastic behavior using a perturbed visco-plastic approach", *Modell. Simul. Mater. Sci. Eng.* 27 (2019), article no. 085013.
- [29] M. Zecevic, R. A. Lebensohn, "New robust self-consistent homogenization schemes of elasto-viscoplastic polycrystals", Int. J. Solids Struct. 202 (2020), p. 434.



Contributions in mechanics of materials

Exact results for weakly nonlinear composites and implications for homogenization methods

Joshua Furer^{*a*}, Martin Idiart^{*b*, *c*} and Pedro Ponte Castañeda^{*, *a*, *d*}

^{*a*} Graduate Group in Applied Mathematics and Computational Sciences, University of Pennsylvania, Philadelphia, PA 19104-6315, USA

^b Centro Tecnológico Aeroespacial / Departamento de Aeronáutica, Facultad de Ingeniería, Universidad Nacional de La Plata, Avda. 1 esq. 47 S/N, La Plata B1900TAG, Argentina

^c Consejo Nacional de Investigaciones Científicas y Técnicas (CONICET), CCT La Plata, Calle 8 N° 1467, La Plata B1904CMC, Argentina

^d Department of Mechanical Engineering and Applied Mechanics, University of Pennsylvania, Philadelphia, PA 19104-6315, USA

E-mails: jfurer@sas.upenn.edu (J. Furer), martin.idiart@ing.unlp.edu.ar (M. I. Idiart), ponte@seas.upenn.edu (P. Ponte Castañeda)

Abstract. Weakly nonlinear composite conductors are characterized by position-dependent dissipation potentials expressible as an additive composition of a quadratic potential and a nonquadratic potential weighted by a small parameter. This additive form carries over to the effective dissipation potential of the composite when expanded to first order in the small parameter. However, the first-order correction of this asymptotic expansion depends only on the zeroth-order values of the local fields, namely, the local fields within the perfectly linear composite conductor. This asymptotic expansion is exploited to derive the exact effective conductivity of a composite cylinder assemblage exhibiting weak nonlinearity of the power-law type (i.e., power law with exponent $m = 1 + \delta$, such that $|\delta| \ll 1$), and found to be identical (to first order in δ) to the corresponding asymptotic result for sequentially laminated composites of infinite rank. These exact results are used to assess the capabilities of more general nonlinear homogenization methods making use of the properties of optimally selected linear comparison composites.

Keywords. Homogenization, Nonlinear, Heterogeneous materials, Exact solution, Composite materials, Effective conductivity.

Manuscript received 12th October 2020, revised 25th November 2020, accepted 27th November 2020.

* Corresponding author.

1. Introduction

The development of multiscale approximations for the mechanical and physical properties of disordered solids greatly benefits from the identification of specific material systems whose properties at different length scales can be linked exactly, as they provide guidance and useful benchmarks for evaluating the relative merits of competing schemes. Several classes of such solvable systems have been identified when the constitutive phases exhibit *linear* responses. These include, for instance, sequential laminates (e.g., [1–3]), dilute dispersions (e.g., [4]), assemblages of neutral coated inclusions (e.g., [5–8]), symmetric materials (e.g., [9, 10]), and iterated composites (e.g., [11–13]). Comparatively fewer results are available when the constitutive phases exhibit *nonlinear* responses. Such nonlinear results are typically generated by partially extending the above linear results, and often require more involved mathematical descriptions. Examples include sequential laminates (e.g., [14, 15]), symmetric materials (e.g., [16]), and iterated composites (e.g., [17, 18]). Now, these descriptions can simplify considerably when the constitutive responses are *weakly nonlinear* (e.g., [19–22]). But, despite this observation, results for weakly nonlinear composites are seldomly exploited in the development of homogenization methods.

The purpose of this paper is to report exact results for the electrical conductivity of twophase composites exhibiting weak nonlinearity. Such composites are characterized by positiondependent dissipation potentials expressible as an additive composition of a quadratic potential and a nonquadratic potential weighted by a small parameter. This additive form carries over to the effective dissipation potential of the composite when expanded to first order in the small parameter. However, the first-order correction of this asymptotic expansion depends only on the zeroth-order values of the local fields, namely, the local fields within the perfectly linear composite conductor. This asymptotic expansion is exploited to derive the exact effective conductivity of a composite cylinder assemblage (CCA) exhibiting weak nonlinearity of the power-law type. More specifically, we consider conductors with phase constitutive relation of the type $J = \epsilon E^m$, where J is the current, E is the electric field, ϵ is the nonlinear conductivity and $m = 1 + \delta$, with $|\delta| \ll 1$. The exact asymptotic result (to first order in δ) for these weakly nonlinear composites, which has not previously been reported in the literature, is then used to assess the capabilities of various nonlinear homogenization methods.

These nonlinear homogenization methods blossomed from ideas initiated by Willis [23], who proposed a generalization of the variational bounds of [24] for linear composites, which could also handle nonlinear composites. The first application of these bounds of the Hashin-Shtrikman (HS) type to nonlinear composites was pursued by Talbot and Willis [25]. A more general variational approach consisting in the use of an optimally chosen "linear comparison composite" (LCC) was advanced by Ponte Castañeda [26, 27] (see also [28] for the special case of power-law materials). This approach is not only capable of delivering bounds of the HS type, but can also be used to generate three-point bounds and other estimates, such as self-consistent-type estimates, by means of corresponding bounds and estimates for linear composites. Because of their bounding properties, these "variational linear comparison" estimates have been shown to deliver significantly improved estimates. For example, Gilormini [29] showed that the "classical" extension [30,31] of the self-consistent method [32] for nonlinear composites violates the rigorous bounds of [26]. In addition, Suguet [33] provided a reinterpretation of the variational linear comparison method of [26] in terms of the second moments of the fields in the phases of the LCC, thus providing an alternative explanation for the improvements observed relative to the classical homogenization schemes using only the averages or first moments of the fields in the phases. However, it was found that these bounds and estimates were not able to reproduce the exact estimates of [34] for small heterogeneity contrast. For these reasons, Ponte Castañeda [35] proposed an alternative "tangent second-order" (TSO) variational approach making use of more general LCCs,

which, while not yielding bounds, was capable of reproducing exactly to second order in the contrast the asymptotic expansions of [34]. In spite of giving improved results for small contrast, the TSO estimates were not as robust as the variational estimates for strongly nonlinear composites exhibiting strong heterogeneity contrast. More recently, Ponte Castañeda [36] provided a fully optimized second-order (FOSO) homogenization method combining the advantages of the variational and TSO methods, which makes use of both the first and second moments of the fields and has the additional advantage of allowing the estimation of these moments in the nonlinear composite directly from the corresponding moments in the LCC [37]. In implementing the FOSO method, one must choose the specific value, a weight-factor α which is not dictated by the underlying variational procedure. Out of convenience, Ponte Castañeda [36] chose a value of $\alpha = 0.5$, and the choice was adopted by Furer and Ponte Castañeda [38], who suggested that the "optimal" value of α would depend, among other things, on the microstructure, nonlinearity, and properties of the phases. The exact results reported in this work are used to assess this choice. Finally, it is also shown that the exact estimate of [15] for the corresponding class of sequentially laminated nonlinear composite conductors also agrees exactly to first order in the weakly nonlinear limit with the CCA result.

2. Effective behavior of weakly nonlinear composites

We consider material systems made up of two distinct constitutive phases. The electrical conductivity of each phase is characterized by a dissipation potential $w^{(r)}$ (r = 1, 2), such that the current density **J** and the electric field intensity **E** are related by

$$\mathbf{J} = \frac{\partial w}{\partial \mathbf{E}}(\mathbf{x}, \mathbf{E}), \quad w(\mathbf{x}, \mathbf{E}) = \sum_{r=1}^{2} \chi^{(r)}(\mathbf{x}) \ w^{(r)}(\mathbf{E}), \tag{1}$$

where the characteristic functions $\chi^{(r)}$ serve to describe the microstructure, being 1 if the position vector **x** is in phase *r*, and 0 otherwise, so that $\chi^{(1)}(\mathbf{x}) + \chi^{(2)}(\mathbf{x}) = 1$. The potentials $w^{(r)}$ are assumed to be strictly convex and bounded from below. The focus is on material systems where the size of the characteristic particle size is much smaller than the size of the specimen. In that case, the functions $\chi^{(r)}$ exhibit rapid oscillations and homogenization theory states that the *overall response* of the composite is given by the relation between the average current density and the average electric field over a "representative volume element" Ω . Then, letting $\langle \cdot \rangle$ denote the volume average over Ω , and letting $\overline{\mathbf{J}} = \langle \mathbf{J} \rangle$ and $\overline{\mathbf{E}} = \langle \mathbf{E} \rangle$, the overall response can be characterized by the *effective potential* \tilde{w} , such that (e.g., [39])

$$\bar{\mathbf{J}} = \frac{\partial \tilde{w}}{\partial \bar{\mathbf{E}}} (\bar{\mathbf{E}}), \quad \tilde{w}(\bar{\mathbf{E}}) = \min_{\mathbf{E} \in \mathcal{K}(\bar{\mathbf{E}})} \langle w(\mathbf{x}, \mathbf{E}) \rangle, \tag{2}$$

where $\mathscr{K}(\overline{\mathbf{E}})$ is the set of admissible fields $\mathbf{E}(\mathbf{x})$, such that there is a continuous scalar field φ in a suitable functional space satisfying $\mathbf{E} = -\nabla \varphi$ in Ω and $\varphi = -\overline{\mathbf{E}} \cdot \mathbf{x}$ on $\partial \Omega$. In view of these differential constraint, the minimizer \mathbf{E} must be such that the associated current density field \mathbf{J} be divergence free, i.e., $\nabla \cdot \mathbf{J} = 0$ within Ω . Dual variational formulations are available involving the Legendre transform of $\widetilde{w}(\overline{\mathbf{E}})$, as given by $\widetilde{u}(\overline{\mathbf{J}}) = \widetilde{w}^*(\overline{\mathbf{E}})$.

This constitutive framework can be used to model weakly nonlinear as well as strongly nonlinear behaviors. Weakly nonlinear materials are hereby characterized by dissipation potentials of the form

$$w^{(r)}(\mathbf{E}) = w_0^{(r)}(\mathbf{E}) + \delta w^{(r)}(\mathbf{E}),$$
(3)

where $w_0^{(r)}$ is quadratic but $w^{(r)}$ is not, $w_0^{(r)}$ and $w^{(r)}$ are both bounded from below, and δ is a small parameter. The effective response of this weakly nonlinear composite can then be determined, at least formally, by expanding the effective potential (2) to first order in δ . Denoting

by \mathbf{E}_{δ} and \mathbf{E}_0 the electric field intensities attaining the minimum in (2)₂ for nonzero δ and for $\delta = 0$, respectively,

$$\widetilde{w}(\overline{\mathbf{E}}) = \langle w(\mathbf{x}, \mathbf{E}_{\delta}) \rangle = \langle w_0(\mathbf{x}, \mathbf{E}_0) \rangle + \delta \left\langle w(\mathbf{x}, \mathbf{E}_0) + \frac{\partial w_0}{\partial \mathbf{E}} (\mathbf{x}, \mathbf{E}_0) \cdot \frac{\partial \mathbf{E}_{\delta}}{\partial \delta} \bigg|_{\delta = 0} \right\rangle + O(\delta^2).$$
(4)

Then, noting that $\partial w_0 / \partial \mathbf{E}(\mathbf{x}, \mathbf{E}_0)$ is the current density field \mathbf{J}_0 within the composite for $\delta = 0$, and is therefore divergence free, and that $\partial \mathbf{E}_{\delta} / \partial \delta|_{\delta=0} = -\nabla \partial \varphi_{\delta} / \partial \delta|_{\delta=0}$, Hill's lemma implies that [40]

$$\left\langle \frac{\partial w_0}{\partial \mathbf{E}}(\mathbf{x}, \mathbf{E}_0) \cdot \frac{\partial \mathbf{E}_{\delta}}{\partial \delta} \right|_{\delta=0} \right\rangle = \left\langle \frac{\partial w_0}{\partial \mathbf{E}}(\mathbf{x}, \mathbf{E}_0) \right\rangle \cdot \left\langle \frac{\partial \mathbf{E}_{\delta}}{\partial \delta} \right|_{\delta=0} \right\rangle = \left\langle \frac{\partial w_0}{\partial \mathbf{E}}(\mathbf{x}, \mathbf{E}_0) \right\rangle \cdot \left. \frac{\partial \overline{\mathbf{E}}}{\partial \delta} \right|_{\delta=0} = 0, \quad (5)$$

since $\langle \mathbf{E}_{\delta} \rangle = \overline{\mathbf{E}}$ and the expansion is carried out for fixed $\overline{\mathbf{E}}$. The effective potential of the weakly nonlinear composite is thus given by

$$\widetilde{w}(\overline{\mathbf{E}}) = \langle w_0(\mathbf{x}, \mathbf{E}_0) \rangle + \delta \langle w(\mathbf{x}, \mathbf{E}_0) \rangle + O(\delta^2), \tag{6}$$

and therefore its evaluation to first order in δ requires knowledge of the *linear* fields only. This observation has been exploited in earlier works [19,20,22] to study composites exhibiting weakly nonlinear responses of the polynomial type. In the next section, results are derived for a different class of composites exhibiting weakly nonlinear responses of the power-law type.

3. Composite cylinder assemblages with power-law dissipation

We consider a special class of cylindrical dispersions consisting of nonoverlapping homothetic composite cylinders of infinite sizes filling up the entire material volume, each composed of an exterior cylinder coating made up of the matrix phase and an interior concentric cylinder made up of the inclusion phase. These microstructures were introduced by HS [5] and are referred to as CCAs. When the phases exhibit an isotropic linear response, the assemblage exhibits an in-plane isotropic linear effective response that can be determined exactly by solving the field equations within a single composite cylinder subject to uniform boundary conditions (e.g., [5, 41]). When the phases exhibit a nonlinear response, on the other hand, the computation of the exact response requires the solution of the field equations throughout the dispersion. In view of the expansion (6), however, the linear fields within a single composite cylinder still furnish the exact effective response when the phases exhibit a weakly nonlinear response, at least to first order. Henceforth, we compute the expansion (6) for assemblages characterized by isotropic dissipation potentials of the power-law form

$$w^{(r)}(\mathbf{E}) = \frac{\epsilon^{(r)}}{m+1} E^{m+1},$$
(7)

where $E = |\mathbf{E}|$ represents the magnitude of \mathbf{E} , $\epsilon^{(r)}$ denotes the conductivity of phase *r*, and the exponent m > 0 characterizes the degree of nonlinearity. For simplicity, the exponent *m* is taken to be the same for both phases. A weakly nonlinear response is reproduced by taking $m = 1 + \delta$ and expanding the power-law potentials to first order in δ :

$$w^{(r)}(\mathbf{E}) = \frac{\epsilon^{(r)}}{2} E^2 + \delta \, \frac{\epsilon^{(r)}}{4} E^2 (\ln E^2 - 1) + O(\delta^2). \tag{8}$$

This expression is indeed of the form (3) with

$$w_0^{(r)}(\mathbf{E}) = \frac{\epsilon^{(r)}}{2} E^2 \text{ and } w^{(r)}(\mathbf{E}) = \frac{\epsilon^{(r)}}{4} E^2 (\ln E^2 - 1).$$
 (9)

Now, it is well known that when both phases exhibit the same nonlinear exponent m, the effective potential is also of the power-law type with the same nonlinear exponent m (see, for instance, [40]). In view of the overall (in-plane) isotropy of the assemblage, we can thus write

$$\widetilde{w}(\overline{\mathbf{E}}) = \frac{\widetilde{\epsilon}}{m+1}\overline{E}^{m+1},\tag{10}$$

where $\overline{E} = |\overline{E}|$. The effective conductivity $\tilde{\epsilon}$ depends on the local conductivities $\epsilon^{(r)}$, the microstructural morphology, and, more importantly, it also depends on the exponent *m*. Thus,

$$\widetilde{w}(\overline{\mathbf{E}}) = \frac{\widetilde{\epsilon}_0}{2}\overline{E}^2 + \delta \frac{\overline{E}^2}{4} [2\widetilde{\epsilon}_1 + \widetilde{\epsilon}_0(\ln \overline{E}^2 - 1)] + O(\delta^2), \tag{11}$$

where the constants $\tilde{\epsilon}_0$ and $\tilde{\epsilon}_1$ are such that $\tilde{\epsilon} = \tilde{\epsilon}_0 + \delta \tilde{\epsilon}_1 + O(\delta^2)$, and characterize completely the effective response. Expressions (6), (9) and (11) imply that

$$\widetilde{\epsilon}_{0}\overline{E}^{2} = \langle \epsilon(\mathbf{x})E_{0}^{2} \rangle \quad \text{and} \quad 2\overline{E}^{2}[\widetilde{\epsilon}_{1} + \widetilde{\epsilon}_{0}\ln\overline{E}] = \langle \epsilon(\mathbf{x})E_{0}^{2}\ln E_{0}^{2} \rangle, \tag{12}$$

which furnish the effective constants $\tilde{\epsilon}_0$ and $\tilde{\epsilon}_1$ in terms of the linear electric field distribution. Within any composite cylinder of the assemblage occupying a domain Ω_c , the linear electric potential is solution to the field equations (see, for instance, [41])

$$-\nabla \cdot [\varepsilon(\mathbf{x})\nabla\varphi_0] = 0 \quad \text{in } \Omega_c, \quad \varphi_0 = -\overline{\mathbf{E}} \cdot \mathbf{x} \quad \text{on } \partial\Omega_c, \quad \varphi_0 \in H^1(\Omega_c), \tag{13}$$

where $\epsilon(\mathbf{x}) = \chi^{(1)}(\mathbf{x})\epsilon^{(1)} + \chi^{(2)}(\mathbf{x})\epsilon^{(2)}$. In terms of a polar coordinate system with origin at the center of the cylinder and polar axis colinear with the applied electric field $\overline{\mathbf{E}}$, the solution to these equations can be written as (see, for instance, [41])

$$\varphi_0(\mathbf{x}) = \begin{cases} \overline{E} \ a_1 r \cos\theta & 0 \le r \le r_i \text{ (inclusion phase)} \\ \overline{E} \left(a_2 r + \frac{b_2}{r} \right) \cos\theta & r_i \le r \le r_o \text{ (matrix phase),} \end{cases}$$
(14)

where r and θ are, respectively, the radial and angular coordinates, r_i and r_o are, respectively, the inner and outer radii of the matrix coating, and a_1 , a_2 , b_2 are constants given by

$$a_1 = -\frac{2\epsilon^{(2)}}{(\epsilon^{(1)} + \epsilon^{(2)}) - c(\epsilon^{(1)} - \epsilon^{(2)})},$$
(15)

$$a_{2} = -\frac{(\epsilon^{(1)} + \epsilon^{(2)})}{(\epsilon^{(1)} + \epsilon^{(2)}) - c(\epsilon^{(1)} - \epsilon^{(2)})},$$
(16)

$$b_2 = \frac{(\epsilon^{(1)} - \epsilon^{(2)})}{(\epsilon^{(1)} + \epsilon^{(2)}) - c(\epsilon^{(1)} - \epsilon^{(2)})} r_i^2.$$
(17)

In these expressions, $c = (r_i/r_o)^2$ denotes the volume fraction of inclusions. The linear electric field intensity is thus given by

$$E_0^2(\mathbf{x}) = |\nabla \varphi_0(\mathbf{x})|^2 = \overline{E}^2 \times \begin{cases} a_1^2 & 0 \le r \le r_i \\ \left(a_2 - \frac{b_2}{r^2}\right)^2 \cos^2 \theta + \left(a_2 + \frac{b_2}{r^2}\right)^2 \sin^2 \theta & r_i \le r \le r_o. \end{cases}$$
(18)

Introducing the field distribution (18) in $(12)_1$, we have that

$$\widetilde{\epsilon}_{0} = \frac{1}{\pi r_{o}^{2}} \int_{0}^{r_{i}} \int_{0}^{2\pi} \epsilon^{(1)} a_{1}^{2} r \, \mathrm{d}r \, \mathrm{d}\theta + \frac{1}{\pi r_{o}^{2}} \int_{r_{i}}^{r_{o}} \int_{0}^{2\pi} \epsilon^{(2)} \left[\left(a_{2} - \frac{b_{2}}{r^{2}} \right)^{2} \cos^{2}\theta + \left(a_{2} + \frac{b_{2}}{r^{2}} \right)^{2} \sin^{2}\theta \right] r \, \mathrm{d}r \, \mathrm{d}\theta,$$
(19)

and upon integration we obtain

$$\tilde{\epsilon}_{0} = \epsilon^{(2)} \frac{(\epsilon^{(1)} + \epsilon^{(2)}) + c(\epsilon^{(1)} - \epsilon^{(2)})}{(\epsilon^{(1)} + \epsilon^{(2)}) - c(\epsilon^{(1)} - \epsilon^{(2)})},$$
(20)

which is the effective conductivity of the linear CCA [5, 41].

In turn, introducing the field distribution (18) in $(12)_2$, we have that

$$2\tilde{\epsilon}_{1} + \tilde{\epsilon}_{0} \ln \overline{E}^{2} = \frac{1}{\pi r_{o}^{2}} \int_{0}^{r_{i}} \int_{0}^{2\pi} \epsilon^{(1)} a_{1}^{2} (\ln a_{1}^{2} + \ln \overline{E}^{2}) r \, dr \, d\theta + \frac{1}{\pi r_{o}^{2}} \int_{r_{i}}^{r_{o}} \int_{0}^{2\pi} \epsilon^{(2)} \left[\left(a_{2} - \frac{b_{2}}{r^{2}} \right)^{2} \cos^{2} \theta + \left(a_{2} + \frac{b_{2}}{r^{2}} \right)^{2} \sin^{2} \theta \right] \times \left(\ln \left[\left(a_{2} - \frac{b_{2}}{r^{2}} \right)^{2} \cos^{2} \theta + \left(a_{2} + \frac{b_{2}}{r^{2}} \right)^{2} \sin^{2} \theta \right] + \ln \overline{E}^{2} \right) r \, dr \, d\theta,$$
(21)

which in view of (19) simplifies to

$$\widetilde{\epsilon}_{1} = \frac{1}{\pi r_{o}^{2}} \int_{0}^{r_{i}} \int_{0}^{2\pi} \frac{\epsilon^{(1)}}{2} a_{1}^{2} \ln a_{1}^{2} r \, \mathrm{d}r \, \mathrm{d}\theta + \frac{1}{\pi r_{o}^{2}} \int_{r_{i}}^{r_{o}} \int_{0}^{2\pi} \frac{\epsilon^{(2)}}{2} \left[\left(a_{2} - \frac{b_{2}}{r^{2}} \right)^{2} \cos^{2}\theta + \left(a_{2} + \frac{b_{2}}{r^{2}} \right)^{2} \sin^{2}\theta \right] \\ \times \ln \left[\left(a_{2} - \frac{b_{2}}{r^{2}} \right)^{2} \cos^{2}\theta + \left(a_{2} + \frac{b_{2}}{r^{2}} \right)^{2} \sin^{2}\theta \right] r \, \mathrm{d}r \, \mathrm{d}\theta,$$
(22)

and upon integration yields (see Appendix A for details)

$$\tilde{\epsilon}_1 = c\epsilon^{(1)}a_1^2 \ln|a_1| + (1-c)\epsilon^{(2)} \left[a_2^2 \ln|a_2| + c(1+\ln|a_2|) \left(\frac{b_2}{r_i^2}\right)^2 \right].$$
(23)

~

Replacing the various constants by their expressions (15)–(17), we finally obtain

$$\widetilde{\epsilon}_{1} = \widetilde{\epsilon}_{0} \ln \left[\frac{(\epsilon^{(1)} + \epsilon^{(2)})}{(\epsilon^{(1)} + \epsilon^{(2)}) - c(\epsilon^{(1)} - \epsilon^{(2)})} \right] + \frac{c\epsilon^{(2)}}{[(\epsilon^{(1)} + \epsilon^{(2)}) - c(\epsilon^{(1)} - \epsilon^{(2)})]^{2}} \left(4\epsilon^{(1)}\epsilon^{(2)} \ln \left[\frac{2\epsilon^{(2)}}{\epsilon^{(1)} + \epsilon^{(2)}} \right] + (1 - c)(\epsilon^{(1)} - \epsilon^{(2)})^{2} \right).$$
(24)

For later reference, it is noted that in the limiting case of perfectly insulating inclusions ($\epsilon^{(1)} = 0$), the exact results (3) and (24) simplify to

$$\frac{\widetilde{\epsilon}_0}{\epsilon^{(2)}} = \frac{1-c}{1+c} \quad \text{and} \quad \frac{\widetilde{\epsilon}_1}{\epsilon^{(2)}} = \frac{1-c}{1+c} \left[\frac{c}{1+c} - \ln(1+c) \right].$$
(25)

4. Infinite-rank laminates with power-law dissipation

It is well known that material systems with disparate classes of underlying microgeometries are most likely to exhibit different effective responses when the local behavior is nonlinear, even if exhibiting coincident effective responses when the local behavior is linear. Motivated by this general rule, we consider here material systems with two-dimensional microgeometries belonging to a fairly rich class confected in [15]. These microgeometries are of "particulate" type and are constructed by resorting to a differential scheme in combination with two-phase sequential laminations of infinite rank, and are such that when the constituent phases and the inclusion distribution are isotropic, and the local constitutive response is linear, their effective conductivity agrees exactly with that of the CCAs considered in the previous section. More generally, the effective dissipation potential of these infinite-rank laminates is given by

$$\widetilde{w}(\overline{\mathbf{E}}) = \widehat{w}(\overline{\mathbf{E}}, -\ln c), \tag{26}$$

where $c = c^{(2)}$ is the volume fraction of the inclusion phase, and $\widehat{w}(\overline{\mathbf{E}}, t)$ is solution to the Hamilton–Jacobi equation

$$\frac{\partial \widehat{w}}{\partial t}(\overline{\mathbf{E}}, t) + \widehat{w} + H\left(\overline{\mathbf{E}}, \frac{\partial \widehat{w}}{\partial \overline{\mathbf{E}}}\right) = 0 \quad \text{with } \widehat{w}(\overline{\mathbf{E}}, 0) = w^{(1)}(\overline{\mathbf{E}})$$
(27)

and Hamiltonian

$$H(\overline{\mathbf{E}}, \overline{\mathbf{J}}) = \max_{a(\mathbf{n})} \langle a\mathbf{n} \cdot \overline{\mathbf{J}} - w^{(2)} (\overline{\mathbf{E}} + a\mathbf{n}) \rangle_{\nu}.$$
 (28)

In this last expression, $\langle \cdot \rangle_{v} = \int_{|\mathbf{n}|=1} (\cdot) v(\mathbf{n}) ds(\mathbf{n})$ is an orientational average weighted by the reduced H-measure $v(\mathbf{n})$ of the microgeometry. For statistically isotropic composites with power-law dissipation potentials, this formalism generates an exact result for the effective conductivity given by (see [42])

$$\widetilde{\epsilon} = \widehat{\epsilon}(-\ln c),\tag{29}$$

where $\hat{\epsilon}(t)$ is solution to the ordinary differential equation

$$\frac{\mathrm{d}\widehat{\epsilon}}{\mathrm{d}t} + h(\widehat{\epsilon}) = 0 \quad \text{with } \widehat{\epsilon}(0) = \epsilon^{(1)}$$
(30)

and

$$h(z) = z + \frac{1}{2\pi} \int_0^{2\pi} ((m+1)z \, a_*(\theta, z) \cos\theta - \epsilon^{(2)} [1 + 2a_*(\theta, z) \cos\theta + a_*(\theta, z)^2]^{(m+1)/2}) \, \mathrm{d}\theta.$$
(31)

In this last expression, the function $a_*(\theta, z)$ is solution to the optimality condition

$$z\cos\theta - \epsilon^{(2)}(a_*(\theta, z) + \cos\theta)(1 + 2a_*(\theta, z)\cos\theta + a_*(\theta, z)^2)^{(m-1)/2} = 0.$$
 (32)

This differential equation can be written in integral form as

$$\int_{\epsilon^{(1)}}^{\tilde{\epsilon}} \frac{\mathrm{d}z}{h(z)} = \ln c. \tag{33}$$

We now set $m = 1 + \delta$ and $\tilde{\epsilon} = \tilde{\epsilon}_0 + \delta \tilde{\epsilon}_1 + O(\delta^2)$, and proceed with the expansion of (33) to first order in δ :

$$\int_{\varepsilon^{(1)}}^{\widetilde{\varepsilon}_0+\delta\ \widetilde{\epsilon}_1+O(\delta^2)} \frac{\mathrm{d}z}{h_0(z)+\delta\ h_1(z)+O(\delta^2)} = \int_{\varepsilon^{(1)}}^{\widetilde{\varepsilon}_0} \frac{\mathrm{d}z}{h_0(z)} + \delta\left[\frac{\widetilde{\epsilon}_1}{h_0(\widetilde{\epsilon}_0)} - \int_{\varepsilon^{(1)}}^{\widetilde{\epsilon}_0} \frac{h_1(z)}{h_0(z)^2} \mathrm{d}z\right] + O(\delta^2) = \ln c, \tag{34}$$

where use has been made of Leibniz's rule; thus,

$$\int_{\varepsilon^{(1)}}^{\widetilde{\varepsilon}_0} \frac{\mathrm{d}z}{h_0(z)} = \ln c \quad \text{and} \quad \widetilde{\varepsilon}_1 = h_0(\widetilde{\varepsilon}_0) \int_{\varepsilon^{(1)}}^{\widetilde{\varepsilon}_0} \frac{h_1(z)}{h_0(z)^2} \,\mathrm{d}z. \tag{35}$$

The first identity furnishes the linear conductivity $\tilde{\epsilon}_0$, while the second identity furnishes the firstorder correction $\tilde{\epsilon}_1$. These identities require the expansion of the integrand h(z), which in turn requires the expansion of the function $a_*(\theta)$ as given by (32). The latter is given by

$$a_*(\theta, z) = \frac{z - \epsilon^{(2)}}{\epsilon^{(2)}} \cos\theta - \delta \frac{z}{\epsilon^{(2)}} \ln\left[\sin^2\theta + \frac{z}{\epsilon^{(2)}} \cos^2\theta\right] \cos\theta + O(\delta^2),\tag{36}$$

while the former is given by

$$h_0(z) = \frac{z^2 - \epsilon^{(2)2}}{2\epsilon^{(2)}} \quad \text{and} \quad h_1(z) = \frac{z^2 - \epsilon^{(2)2}}{4\epsilon^{(2)}} - \frac{z^2 + \epsilon^{(2)2}}{2\epsilon^{(2)}} \ln\left(\frac{z + \epsilon^{(2)}}{2\epsilon^{(2)}}\right). \tag{37}$$

In this last expression, use has been made of the integrals (49). Introducing $(37)_1$ into $(35)_1$ and evaluating the integral we obtain

$$\frac{\widetilde{\epsilon}_0 - \epsilon^{(2)}}{\widetilde{\epsilon}_0 + \epsilon^{(2)}} \frac{\epsilon^{(1)} + \epsilon^{(2)}}{\epsilon^{(1)} - \epsilon^{(2)}} = c.$$
(38)

The solution to this equation for the linear effective conductivity $\tilde{\epsilon}_0$ agrees exactly with (3). In turn, introducing (37) and (38) into (35)₂ and evaluating the integral we obtain an expression that agrees exactly with expression (24) for $\tilde{\epsilon}_1$. In conclusion, these infinite-rank laminates exhibit the same effective response as the CCAs of the previous section not only when the phases exhibit linear behavior but also when the phases exhibit weakly nonlinear behavior of the power-law type (to first order in the nonlinearity perturbation parameter δ). The fact that the same weakly nonlinear conductivity is displayed by the infinite-rank laminates as for the CCAs suggests that the effective parameter $\tilde{\epsilon}_1$ may be only modestly sensitive to microgeometrical details (as is the case for the effective parameter $\tilde{\epsilon}_0$ for linear composites).

5. Estimates for weakly nonlinear composites based on linear comparison composites

This section provides the relevant formulas for the various nonlinear homogenization estimates considered in this work, after being specialized to weakly nonlinear, power-law, composite conductors in two dimensions. In all cases, the effective properties of the LCC are determined via the estimates of [5] for two-phase composite conductors (specialized to 2D). Therefore, all the nonlinear estimates recover the exact conductivity of the CCA of the previous section when the phases exhibit a linear response.

First, it is recalled that variational (VAR) bounds of the HS type for two-phase nonlinear conductors have been given by Ponte Castañeda [27]. Expressions for the VAR bounds for weakly nonlinear composites were obtained by Ponte Castañeda [43] for general nonlinear correction $w^{(r)}$, which can be specialized for power-law conductors by means of expression (9)₂ for $w^{(r)}$. The form of the resulting correction $\tilde{\epsilon}_1$ for general two-phase composites is somewhat too cumbersome to include here and it will simply be noted that the result does not recover the exact result for the CCA given by expression (24), although of course it still provides a bound. For the special case when the inclusions are assumed to be perfectly insulating ($\epsilon^{(1)} \rightarrow 0$), the result for the effective nonlinear conductivity (for power-law behavior) simplifies considerably to the expression

$$\frac{\tilde{\epsilon}^{\text{VAR}}}{\epsilon^{(2)}} = \frac{1-c}{(1+c)^{(m+1)/2}},$$
(39)

while the corresponding result for the weakly nonlinear correction reduces to

$$\frac{\widetilde{\epsilon}_1^{\text{VAR}}}{\epsilon^{(2)}} = -\frac{1}{2} \frac{1-c}{1+c} \ln(1+c), \tag{40}$$

which is indeed seen to be different from the corresponding exact expression (25)₂ for the CCA.

Next, it is recalled that the TSO estimates for nonlinear composite conductors were given by Ponte Castañeda [44]. Two different results were obtained depending on whether one starts with the primal formulation (\tilde{w}), as given by (2), or by the dual formulation obtained by means of the Legendre transformation ($\tilde{u} = \tilde{w}^*$). The result for the primal formulation (\tilde{w}) is given by

$$\tilde{\epsilon}^{\text{TSO}} = \sum_{r=1}^{2} c^{(r)} \epsilon^{(r)} \left[E^{(r)(1+m)} + \frac{1+m}{2} E^{(r)m} (1-E^{(r)}) \right], \tag{41}$$

where $cE^{(1)} + (1 - c)E^{(2)} = 1$ and $E^{(2)}$ is solution to

$$1 - \frac{\epsilon^{(1)}}{\epsilon^{(2)}} \frac{1 - (1 - c)E^{(2)}}{(cE^{(2)})^m} = \frac{1 - E^{(2)}}{cE^{(2)}}\sqrt{m}.$$
(42)

The solution to this equation admits the following asymptotic expansion in the small parameter $\delta = m - 1$:

$$E^{(2)} = \frac{\epsilon^{(1)} + \epsilon^{(2)}}{(\epsilon^{(1)} + \epsilon^{(2)}) - c(\epsilon^{(1)} - \epsilon^{(2)})} + \delta \frac{c}{2} \frac{\epsilon^{(2)} - \epsilon^{(1)} - 4\epsilon^{(1)} \ln \left[\frac{\epsilon^{(1)} + \epsilon^{(2)}}{2}\right]}{[(\epsilon^{(1)} + \epsilon^{(2)}) - c(\epsilon^{(1)} - \epsilon^{(2)})]^2} \epsilon^{(2)} + O(\delta^2).$$
(43)

Upon introducing this expression into (41) and expanding in δ , it can be verified that the TSO estimate reproduces the exact correction $\tilde{\epsilon}_1$ for the conductivity of the weakly nonlinear CCA, as given (24). The corresponding result for the dual formulation (\hat{u}) is similar in form to the above result for the primal (\tilde{w}) formulation and will not be repeated here, for brevity. However, it should be noted that while the primal and dual TSO estimates are not identical (i.e., there is a duality gap), it can be shown that the dual formulation also leads to the exact result for the weakly nonlinear correction $\tilde{\epsilon}_1$ of the CCA.

Finally, we consider the FOSO estimates of [36]. The FOSO method—like the VAR method does not have a duality gap, but the final form of the estimate depends on the formulation (primal or dual). In fact, the initial form of the potentials for the LCC in the primal (\tilde{w}) and dual (\tilde{u}) formulations were chosen by Ponte Castañeda [36] to be slightly inconsistent, for simplicity of the calculations, and as a consequence of this "nonsymmetric" choice of the LCC, the FOSO delivers two different sets of estimates depending on whether the derivation is based on electric field potentials (w), giving rise to the FOSO(W) formulation, or on the current density potentials (u), giving rise to the so-called FOSO(U) formulation. In spite of the inconsistency in the predictions of the two formulations, which is due to the lack of symmetry in the form of the energy potential of the LCC, it should be emphasized that both formulations are fully consistent and do not exhibit a duality gap. By using a more symmetric choice for the potential of the LCC, Furer and Ponte Castañeda [38] proposed a "symmetric" version of the FOSO method, which predicts the same effective response whether one starts from the primal (\tilde{w}) or dual (\tilde{u}) formulations. As will be seen below, all three formulations deliver the same estimates in the weakly nonlinear limit.

Since the expressions for the different versions of the FOSO estimates are a bit complicated, we will consider here only the case of perfectly insulating inclusions ($\epsilon^{(1)} \rightarrow 0$), which corresponds to the case of infinite heterogeneity contrast (recall that the results are exact to second order in the heterogeneity contrast) and should be representative of more general cases. The expressions for general nonlinearity are provided in Appendix B. As already mentioned, in the weakly nonlinear limit, all three estimates are identical to first order in $\delta = m - 1$ with correction given by

$$\frac{\tilde{\epsilon}_{1}^{\text{FOSO}}}{\epsilon^{(2)}} = -\frac{1-c}{1+c}\ln(1+c) + \frac{(1-c)}{2(1+c)^{2}}[(1-\alpha)\hat{j}^{2}\ln(\hat{j}^{2}) + \alpha\check{j}^{2}\ln(\check{j}^{2})], \tag{44}$$

where

$$\hat{j}^2 = (1-\alpha)\left(1+\sqrt{2c}\sqrt{\frac{\alpha}{1-\alpha}} + \frac{\alpha c}{1-\alpha}\right), \quad \check{j}^2 = \alpha\left(1-\sqrt{2c}\sqrt{\frac{1-\alpha}{\alpha}} + \frac{(1-\alpha)c}{\alpha}\right)$$
(45)

and where α corresponds to a certain weight factor usually set equal to 1/2.

6. Results and discussion

The exact results derived in Section 3 for CCAs exhibiting weak power-law nonlinearity are now exploited to assess in more detail the capabilities of the various homogenization estimates of Section 5. (Since the corresponding results of Section 4 for the infinite-rank laminates are identical to the results for CCAs, it will not be necessary to refer to those results explicitly in this section, although the fact that they are identical should of course be kept in mind.) We begin by recalling that both versions (primal and dual) of the TSO estimates reproduce the exact result for any volume fraction and heterogeneity contrast. Thus, these estimates are found to be asymptotically exact not only for weakly heterogeneous systems—to second order in the contrast—but also for weakly nonlinear systems—to first order in the nonlinearity parameter $\delta = m - 1$. However recomforting, this observation is not necessarily expected to hold for more general weakly nonlinear systems.

We now focus on the VAR and FOSO estimates, which—unlike the TSO estimates—do not reproduce the exact nonlinear correction $\tilde{\epsilon}_1$ on the effective conductivity. However, it is recalled that the VAR estimate provides a lower bound for $\tilde{\epsilon}_1$, while the various versions of the FOSO reduce to the same identical result for $\tilde{\epsilon}_1$ and depend on a certain weight factor α . Figure 1 provides plots of these estimates for the case of perfectly insulating inclusions, along with the exact result. For the FOSO estimates, three different values of the parameter α are considered ($\alpha =$ 0.25, 0.5, 0.75). Part (a) displays plots of the nonlinear correction $\tilde{\epsilon}_1$ as a function of the inclusion volume fraction *c*, normalized by the matrix conductivity $\epsilon^{(2)}$. Among all these estimates, the VAR estimate is found to be the least accurate. This is related to the fact that the VAR estimate actually provides rigorous bounds for the effective conductivity of all composites with statistically isotropic microstructures—including for the CCAs. However, as shown recently by Furer and



Figure 1. (a) Exact result and corresponding estimates for the weakly nonlinear correction $\tilde{\epsilon}_1$ as a function of inclusion volume fraction *c*, normalized by the matrix conductivity $\epsilon^{(2)}$. (b) The value of α for which the FOSO estimate for $\tilde{\epsilon}_1$ reproduces the exact result.

Ponte Castañeda [45], the VAR bounds also hold for microstructures that, while isotropic in the linear case, are anisotropic in the nonlinear case. Thus the VAR estimate provides an upper bound on the dissipation potential, which translates into a lower bound on the correction $\tilde{\epsilon}_1$. The FOSO estimates are seen to comply with this bound for the three values of α considered. Interestingly, the FOSO predictions are seen to be asymmetric about $\alpha = 0.5$. Of the three values of α considered, $\alpha = 0.5$ is found to generate the most accurate predictions. Moreover, these predictions are quite close to the exact result for the entire range of inclusion volume fraction. A comparison of the analytical expressions for the exact result and the FOSO estimates with arbitrary α reveals that the FOSO estimate reproduces the exact result if α is chosen to satisfy the equation

$$2c = (1-\alpha)\left(1+\sqrt{2c}\sqrt{\frac{\alpha}{1-\alpha}} + \frac{\alpha c}{1-\alpha}\right)\ln\left(1+\sqrt{2c}\sqrt{\frac{\alpha}{1-\alpha}} + \frac{\alpha c}{1-\alpha}\right) + \alpha\left(1-\sqrt{2c}\sqrt{\frac{1-\alpha}{\alpha}} + \frac{(1-\alpha)c}{\alpha}\right)\ln\left(1-\sqrt{2c}\sqrt{\frac{1-\alpha}{\alpha}} + \frac{(1-\alpha)c}{\alpha}\right).$$
(46)

The solution to this equation is plotted in Figure 1(b) as a function of the volume fraction of inclusions *c*. While clearly dependent on *c*, this "optimal" value of α is seen to be very close to the value $\alpha = 0.5$ advocated by Furer and Ponte Castañeda [38] based on comparisons with other estimates. In fact, it can be verified that the value $\alpha = 0.5$ leads to the *exact* asymptotic result, $\tilde{\epsilon}_1/\epsilon^{(2)} \sim -c^2/2$, in the dilute limit ($c \ll 1$). These observations for an infinitely contrasted system, and the fact that the FOSO estimates are exact for weakly contrasted systems to second order, suggest that the use of $\alpha = 0.5$ for general material systems provides a reasonable compromise between accuracy and simplicity.

Next, the influence of α on the FOSO predictions for strongly nonlinear power-law assemblages is considered. Thus, Figure 2 shows results for the effective conductivity $\tilde{\epsilon}$, as a function of nonlinearly *m*, for a fixed value of *c* = 0.2. For comparison purposes, results are included for the three different versions of the FOSO methods—nonsymmetric FOSO(U) and FOSO(W) [36] and symmetric FOSO [38]—as well as the VAR bounds, the two versions of the TSO estimates (TSO(U) and TSO(W)) and the exact weakly nonlinear limit to first order in $\delta = m - 1$ (and therefore linear in *m*). The pairs of Figures 2(a) and (b) and Figures 2(c) and (d) show, respectively, the effective



Figure 2. Estimates for the effective conductivity with α equal to (a, b) 0.5 and (c, d) the value for which the FOSO method gives the exact correction ($\alpha \approx 0.5178$). Results are presented as a function of the nonlinear *m*, for a fixed volume fraction *c* = 0.2 of the perfectly insulating phase.

conductivity for both the entire range of nonlinearities (left column), as well as zoomed-in versions for weak nonlinearities (right column), for $\alpha = 0.5$ (top row) and the value of α ($\alpha \approx 0.5178$) for which the FOSO estimate recovers the exact result in the weakly nonlinear limit (bottom row). It is noted that the order in which the different methods appear in each legend corresponds to the order in which the values predicted by the various methods at m = 0 also appear. In particular, the topmost entry in the legend is the method predicting the largest value of the effective conductivity at m = 0, while the bottom-most entry in the legend is the method predicting the smallest value at m = 0. As was remarked by Furer and Ponte Castañeda [38], the symmetric FOSO estimate always lies somewhere between the FOSO(U) and FOSO(W) estimates, with a maximum difference of about 5% between the symmetric and nonsymmetric versions for m = 0. Moreover, the effect of changing α on the various FOSO predictions is relatively minor; for example, as can be seen in Figures 2(a) and (c), the symmetric FOSO method predicts a value of the effective conductivity for m = 0 slightly above 0.65 when $\alpha = 0.5$, while it gives a value slightly below 0.65 when $\alpha \approx 0.5178$. On the other hand, the TSO estimates, which are independent of any weight factors, are very close for values of m between 0.5 and 1, but diverge dramatically in the strongly non-

linear limit as *m* approaches zero. In fact the TSO(U) violates the upper bound provided by the VAR estimates for values of *m* less than about 0.04. In addition, it is seen in Figure 2(a) that the FOSO(U) method and the TSO(W) method are rather close for values of $0.5 \le m \le 1$, while, in Figure 2(c), this agreement continues up to $m \approx 0.2$. In fact, the FOSO(U) estimate crosses above the TSO(W) estimate for 0.3 < m < 1 in Figure 2(a) for $\alpha = 0.5$, but remains always below it in Figure 2(c) when $\alpha \approx 0.5178$.

The magnified results in Figures 2(b) and (d) show more clearly the effect of the choice of α on the comparisons of the FOSO estimates with the other estimates. As expected, the two TSO estimates, which give the exact weakly nonlinear behavior, lie tangent to the first-order approximation in the limit as $m \rightarrow 1$ in both figures. On the other hand, the various FOSO estimates only do so for $\alpha \approx 0.5178$, as can be seen in Figure 2(d). In fact, consistent with earlier observations for this value of α , it can also be seen in Figure 2(d) that the FOSO(U) and TSO(W) estimates appear to agree, as do the FOSO(W) and TSO(U) estimates, even for nonlinearities up to m = 0.8, again with the symmetric FOSO lying roughly in between the two sets of estimates. Finally, also consistent with earlier observations, all the estimates are seen to be significantly lower than the VAR bound. Again, this can be explained by the fact that the VAR bound must hold for certain nonlinearly anisotropic microstructures, including the finite-rank microstructures attaining the corresponding linear bounds [45].

7. Closing remarks

This paper provides exact asymptotic estimates for weakly nonlinear power-law composite conductors with CCA and infinite-rank laminated microstructures. It is known that both of these microstructures attain exactly the 2-D HS bounds [5], when the material behavior is linear. The objective of this work is to test more general estimates obtained by various nonlinear homogenization methods making use of corresponding estimates for LCCs against these exact results in order to assess their accuracy and capabilities. The first finding is that the TSO estimates of the HS type [44] agree to first order in the nonlinearity parameter with the exact asymptotic result. However, this surprising result has been found (details not presented in this paper) not to carry over to the corresponding 3-D conductivity results, and may therefore be little more than a coincidence. Indeed, it is known that the TSO method performs poorly for strongly nonlinear behavior, as a consequence of the fact that it neglects the effect of the field fluctuations which become very significant in the strongly nonlinear limit. In particular, the TSO estimates are known to exhibit a duality gap that increases from zero in the weakly nonlinear limit to very large values in the strong nonlinear limit.

For these reasons, improved FOSO estimates have been proposed [36], which make use of both the first and second moments of the field fluctuations in the linearization procedure, albeit at the expense of having to introduce certain weight factors α (corresponding to weights of the multiple stationary points of the error function in the LCC methods) in order to ensure that the resulting estimates do not exhibit duality gaps. Nonetheless, they have the added advantage that they allow the estimation of the macroscopic behavior, as well as of the first and second moments of the field statistics, directly from the corresponding estimates for the LCC. The second important finding of this work is that the FOSO estimates depend on the weight factors even in the weakly nonlinear limit. However, the FOSO can be made to be consistent with the weakly nonlinear limit for an appropriate choice of the weight factor α . More importantly, the resulting value for α , at least in this very special case, is found to be very close to 1/2, which is the value initially proposed by Ponte Castañeda [36], and found to provide reasonably accurate estimates also by Furer and Ponte Castañeda [38] in other situations. In fact, the choice $\alpha = 1/2$ for the FOSO delivers the exact result for the weakly nonlinear correction to the effective conductivity in the limit of dilute concentrations of perfectly conducting particles. Consequently, the choice of 1/2 for the weight factors α is recommended more generally, at least until this issue is better understood (see also [46]).

The third significant finding is that the VAR bounds of the HS type [27]—while still rigorous bounds for weakly nonlinear composites—are not exact to first order in the nonlinearity parameter. However, this result is not surprising in view of recent findings by Furer and Ponte Castañeda [45] showing that the VAR bound of the HS type, which makes use of the HS bounds for the corresponding LCC, must include microstructures that, while isotropic for linear material behavior, can be anisotropic when used for nonlinear composites. In fact, it was shown by Furer and Ponte Castañeda [45] that the same finite-rank sequential laminates that have been shown to attain the HS bounds in the linear case [2], also attain the VAR bounds of the HS type for certain special loadings.

Finally, it was also shown that the estimates of [15] for infinite-rank sequential laminates coincide with the corresponding exact results for the weakly nonlinear CCA microstructures— to first order in the nonlinearity parameter. In fact, we have recently verified that this is also true in 3D conductivity, where the effective properties of the composite sphere assemblage (CSA) and isotropic infinite-rank laminates agree to first order in the nonlinearity parameter. (Details are not shown here for brevity.) In any case, this final result seems to suggest that the effective behavior of weakly nonlinear composites should be relatively insensitive to the specific type of particulate microstructure involved, just as for linear composites where the CCA/CSA and sequential laminates (of various ranks) lead to identical macroscopic response.

Acknowledgments

JF and PPC acknowledge support by the National Science Foundation under Grant No. DMS-1613926. MII acknowledges support by the Air Force Office of Scientific Research (USA) under award number FA9550-19-1-0377.

Appendix A. Integration in the weakly nonlinear constant (22)

The integration over the angular variable θ in the second term of the right-hand side of (22) requires the evaluation of

$$\int_{0}^{2\pi} \left[\left(a_2 - \frac{b_2}{r^2} \right)^2 \cos^2\theta + \left(a_2 + \frac{b_2}{r^2} \right)^2 \sin^2\theta \right] \ln \left[\left(a_2 - \frac{b_2}{r^2} \right)^2 \cos^2\theta + \left(a_2 + \frac{b_2}{r^2} \right)^2 \sin^2\theta \right] d\theta.$$
(47)

Making use of standard identities for squared trigonometric functions in the argument of the logarithm, this expression can be written as

$$\left(a_{2} - \frac{b_{2}}{r^{2}}\right)^{2} \int_{0}^{2\pi} \cos^{2}\theta \ln\left[a_{2}^{2} - 2a_{2}\frac{b_{2}}{r^{2}}\cos(2\theta) + \left(\frac{b_{2}}{r^{2}}\right)^{2}\right] d\theta + \left(a_{2} + \frac{b_{2}}{r^{2}}\right)^{2} \int_{0}^{2\pi} \sin^{2}\theta \ln\left[a_{2}^{2} - 2a_{2}\frac{b_{2}}{r^{2}}\cos(2\theta) + \left(\frac{b_{2}}{r^{2}}\right)^{2}\right] d\theta.$$
(48)

The two integrals in this expression can be evaluated by noting that for $r_i \le r \le r_o$ the ratio $|b_2/r^2/a_2| < 1$, see expressions (16) and (17), and by invoking the integrals (see integrals 4.397-14 4.397-15 in [47])

$$\int_{0}^{\pi/2} \cos^{2}\theta \ln\left[1 + 2a\cos(2\theta) + a^{2}\right] d\theta = -\int_{0}^{\pi/2} \sin^{2}\theta \ln\left[1 + 2a\cos(2\theta) + a^{2}\right] d\theta = \frac{\pi a}{4}$$
(49)

for |a| < 1. Thus, the integral (48) is

$$2\pi \left[a_2^2 + \left(\frac{b_2}{r^2}\right)^2 \right] \ln a_2^2 + 4\pi \left(\frac{b_2}{r^2}\right)^2.$$
 (50)

In view of this last expression, the second term of the right-hand side of (22) is given by

$$\frac{\epsilon^{(2)}}{2} \frac{1}{\pi r_o^2} \int_{r_i}^{r_o} 2\pi \left\{ \left[a_2^2 + \left(\frac{b_2}{r^2} \right)^2 \right] \ln a_2^2 + 2 \left(\frac{b_2}{r^2} \right)^2 \right\} r \, \mathrm{d}r$$

$$= \frac{\epsilon^{(2)}}{2} \int_{c}^{1} \left\{ \left[a_2^2 + \left(\frac{b_2}{r_o^2} \right)^2 z^{-2} \right] \ln a_2^2 + 2 \left(\frac{b_2}{r_o^2} \right)^2 z^{-2} \right\} \, \mathrm{d}z$$

$$= \epsilon^{(2)} (1-c) \left[a_2^2 \ln |a_2| + c(1+\ln |a_2|) \left(\frac{b_2}{r_i^2} \right)^2 \right]. \tag{51}$$

Appendix B. Fully optimized second-order estimates for power-law conductors with perfectly insulating inclusions

For the case of perfectly insulating inclusions considered here, the dual (\tilde{u}) formulation is the easiest to work with and therefore we start with the FOSO(U) version. We then give the corresponding results for the symmetric FOSO version, which is similar in form. Finally, the results of the FOSO(W) version, which makes use of the more complex primal (\tilde{w}) formulation, is given.

FOSO(U) version. Converting the results of [36] for 2-D viscoplastic composites with porous inclusions to the mathematically analogous case of 2-D power-law conductors with perfectly insulating inclusions, it is found that the effective conductivity $\tilde{\epsilon}^{\text{FOSO}}$, defined by (10), is given by

$$\frac{\widetilde{\epsilon}^{\text{FOSO}}}{\epsilon^{(2)}} = (1-c) \left[\sup_{k} \{(1-\alpha) \widehat{j}^{n+1} + \alpha \widecheck{j}^{n+1}\} \right]^{-1/n}$$
$$= (1-c) \left[\sup_{k} f(k(n), n) \right]^{-1/n},$$
(52)

where n = 1/m and

$$\hat{j} = \sqrt{\hat{j}_{\parallel}^{2}} + \hat{j}_{\perp}^{2}, \quad \check{j} = \sqrt{\check{j}_{\parallel}^{2}} + \check{j}_{\perp}^{2},$$

$$\hat{j}_{\parallel} = 1 + \sqrt{\frac{\alpha}{1-\alpha}} \sqrt{\frac{c}{2}} k^{1/4}, \quad \check{j}_{\parallel} = 1 - \sqrt{\frac{1-\alpha}{\alpha}} \sqrt{\frac{c}{2}} k^{1/4},$$

$$\hat{j}_{\perp} = \sqrt{\frac{\alpha}{1-\alpha}} \sqrt{\frac{c}{2}} k^{-1/4}, \quad \check{j}_{\perp} = -\sqrt{\frac{1-\alpha}{\alpha}} \sqrt{\frac{c}{2}} k^{-1/4}.$$
(53)

In addition, k = k(n) is the anisotropy ratio of the LCC, and the optimal value k^* is determined as a solution to the equation

$$\frac{\partial f(k^*(n), n)}{\partial k} = 0.$$
(54)

Then, letting $k = k^*(n)$, as determined by the stationarity condition (54), and making use of the fact that $\partial/\partial m = -m^{-2}(\partial/\partial n)$, where n = 1/m, it is found that

$$\frac{\tilde{\epsilon_1}^{\text{FOSO}}}{\epsilon^{(2)}} = \frac{\partial}{\partial m} \frac{\tilde{\epsilon}^{\text{FOSO}}}{\epsilon^{(2)}} = (1-c) \frac{\partial}{\partial m} [f(k^*(n), n)]^{-1/n}$$

$$= -\frac{(1-c)}{m^2} \frac{\partial}{\partial n} [f(k^*, n)]^{-1/n}$$

$$= -\frac{(1-c)}{m^2} [f(k^*, n)]^{-1/n} \left[\frac{\log(f(k^*, n))}{n^2} - \frac{1}{nf(k^*, n)} \left(\frac{\partial f(k^*, n)}{\partial k} \frac{\partial k^*}{\partial n} + \frac{\partial f(k^*, n)}{\partial n} \right) \right].$$
(55)

C. R. Mécanique, **2020**, 348, nº 10-11, 893-909

Note that the derivative with respect to *n* in the last line is taken with k^* held fixed, while the term $(\partial f(k^*, n))/\partial k$ vanishes on account of (54). Using the fact that $k^*(1) = 1$ and f(1, 1) = (1 + c), it follows that

$$\frac{\tilde{\epsilon}_1^{\text{FOSO}}}{\epsilon^{(2)}} = \left. \frac{\partial}{\partial m} \frac{\tilde{\epsilon}^{\text{FOSO}}}{\epsilon^{(2)}} \right|_{m=1} = \frac{(1-c)}{(1+c)^2} \left[(1-\alpha) \hat{j}^2 \log(\hat{j}) + \alpha \check{j}^2 \log(\check{j}) \right] - \frac{1-c}{1+c} \log(1+c), \tag{56}$$

where \hat{j} and \check{j} are given as in (53), with k = 1.

Symmetric FOSO version. The symmetric FOSO version is also given (see [38] for details) by an expression of the form (52), with \hat{j}_{\parallel} and \check{j}_{\parallel} as given in (53), but with \hat{j}_{\perp} and \check{j}_{\perp} obtained as the solution of equations

$$\hat{j}_{\perp}\check{j}_{\perp}((\hat{j}_{\parallel}-\check{j}_{\parallel})+k(1-2\alpha)(\hat{j}_{\parallel}+\check{j}_{\parallel}))-2k((1-\alpha)\check{j}_{\parallel}\hat{j}_{\perp}^{2}-\alpha\hat{j}_{\parallel}\check{j}_{\perp}^{2})=0,$$
(57)

$$4\alpha(1-\alpha)\hat{j}_{\perp}\check{j}_{\perp} + (1-2\alpha)\left((1-\alpha)\hat{j}_{\perp}^2 - \alpha\check{j}_{\perp}^2\right) + \frac{c}{2\sqrt{k}} = 0,$$
(58)

and chosen in just a way that $\check{j}_{\perp} < \hat{j}_{\perp}$. Then, following the same procedure as above, and noting that in this context, when k = 1, the expressions for \hat{j}_{\perp} and \check{j}_{\perp} using the symmetric FOSO method is the same as those for the FOSO(U) method, we conclude that they will give the same first-order correction, given by (56).

FOSO(W) version. The implementation of the FOSO(W) version makes use of the primal (\tilde{w}) formulation, which we spell out next and is a bit more complicated, as the average electric field in the phases are unknown. Nonetheless, it can be shown that an estimate for the effective conductivity can equivalently be estimated via

$$\frac{\tilde{\epsilon}^{\text{FOSO}}}{\epsilon^{(2)}} = (1-c) \sup_{k,\overline{E}^{(2)}} \{(1-\alpha)\hat{E}^{m+1} + \alpha\check{E}^{m+1}\}$$

$$= (1-c) \sup_{k,\overline{E}^{(2)}} g(k(m),\overline{E}^{(2)}(m),m),$$
(59)

where $\overline{E}^{(2)}$ is the average electric field in the matrix phase. The optimal values of k^* and $\overline{E}^{(2)*}$ are then determined via the equations

$$\frac{\partial g(k^*, \overline{E}^{(2)*}, m)}{\partial k} = 0,$$

$$\frac{\partial g(k^*, \overline{E}^{(2)*}, m)}{\partial \overline{E}^{(2)}} = 0.$$
 (60)

As above, the form of $g(k, \overline{E}^{(2)}, m)$ is different depending on whether one uses the FOSO(W) method or the symmetric FOSO method. In the former case, we have

$$\widehat{E} = \sqrt{\widehat{E}_{\parallel}^{2} + \widehat{E}_{\perp}^{2}}, \quad \widecheck{E} = \sqrt{\widecheck{E}_{\parallel}^{2} + \widecheck{E}_{\perp}^{2}}, \\
\widehat{E}_{\parallel} = \overline{E}^{(2)} - (\overline{E}^{(2)} - 1)\sqrt{\frac{\alpha}{1 - \alpha}} \frac{k^{-1/4}}{\sqrt{2c}}, \quad \widecheck{E}_{\parallel} = \overline{E}^{(2)} + (\overline{E}^{(2)} - 1)\sqrt{\frac{1 - \alpha}{\alpha}} \frac{k^{-1/4}}{\sqrt{2c}}, \\
\widehat{E}_{\perp} = -(\overline{E}^{(2)} - 1)\sqrt{\frac{\alpha}{1 - \alpha}} \frac{k^{1/4}}{\sqrt{2c}}, \quad \widecheck{E}_{\perp} = (\overline{E}^{(2)} - 1)\sqrt{\frac{1 - \alpha}{\alpha}} \frac{k^{1/4}}{\sqrt{2c}}, \quad (61)$$

while in the latter case, \hat{E}_{\parallel} and \check{E}_{\parallel} are the same, while \hat{E}_{\perp} and \check{E}_{\perp} are again related through

$$\widehat{E}_{\perp}\check{E}_{\perp}(k(\widehat{E}_{\parallel}-\check{E}_{\parallel})+(1-2\alpha)(\widehat{E}_{\parallel}+\check{E}_{\parallel}))-2((1-\alpha)\check{E}_{\parallel}\widehat{E}_{\perp}^{2}-\alpha\widehat{E}_{\parallel}\check{E}_{\perp}^{2})=0,$$
(62)

$$4\alpha(1-\alpha)\widehat{E}_{\perp}\check{E}_{\perp} + (1-2\alpha)((1-\alpha)\widehat{E}_{\perp}^2 - \alpha\check{E}_{\perp}^2) + (E^{(2)}-1)^2\frac{\sqrt{\kappa}}{2c} = 0,$$
(63)

and chosen so that $\check{E}_{\perp} < \widehat{E}_{\perp}$.

C. R. Mécanique, 2020, 348, nº 10-11, 893-909

Once again, when m = 1, so that k = 1, the expressions are the same whether one uses the FOSO(W) or symmetric FOSO method. Moreover, using the fact that $\overline{E}^{(2)*}(1) = (1 + c)^{-1}$, it can be shown that the FOSO(W) version leads to the same correction $\tilde{\epsilon}_1$ as the FOSO(U) version, as given by (56).

References

- [1] J. C. Maxwell, A Treatise on Electricity and Magnetism, Clarendon Press, Oxford, UK, 1873.
- [2] L. Tartar, "Estimations fines des coefficients homogénéisés", in *Ennio de Giorgi Colloquium* (P. Krée, ed.), Research Notes in Math., vol. 125, Pitman Publishing Ltd., London, 1985, p. 168-187.
- [3] G. Francfort, F. Murat, "Homogenization and optimal bounds in linear elasticity", *Arch. Ration. Mech. Anal.* 94 (1986), p. 307-334.
- [4] A. B. Movchan, S. K. Serkov, "The Pólya-Szegó matrices in asymptotic models of dilute composites", Eur. J. Appl. Math. 8 (1997), p. 595-621.
- [5] Z. Hashin, S. Shtrikman, "A variational approach to the theory of the effective magnetic permeability of multiphase materials", J. Appl. Phys. 33 (1962), p. 3125-3131.
- [6] K. Schulgasser, "Sphere assemblage model for polycrystals and symmetric materials", J. Appl. Phys. 54 (1983), p. 1380-1382.
- [7] P. Gilormini, "Realizable compressibility and conductivity in isotropic two-phase composites", C. R. Acad. Sci. Paris IIb–Mech. 329 (2001), p. 851-855.
- [8] G. W. Milton, S. K. Serkov, "Neutral coated inclusions in conductivity and anti-plane elasticity", Proc. R. Soc. Lond. A 457 (2001), p. 1973-1997.
- [9] A. M. Dykhne, "Conductivity of a two-dimensional two-phase system", Zh. Eksp. Teor. Fiz. 59 (1970), p. 110-115.
- [10] V. L. Berdichevskii, "Heat conduction of checkerboard structures", Vestnik Moskov. Univ. Ser. I Mat. Mekh. 40 (1985), p. 56-63.
- [11] D. A. G. Bruggeman, "Berechnung verschiedener physikalischer Konstanten von heterogenen Substanzen. I. Dielektrizitätskonstanten und Leitfähigkeiten der Mischkörper aus isotropen Substanzen", Ann. Phys. 416 (1935), p. 636-664.
- [12] M. Avellaneda, "Optimal bounds and microgeometries for elastic two-phase composites", SIAM J. Appl. Math. 47 (1987), p. 1216-1228.
- [13] M. I. Idiart, "Multiphase conductors realizing Aleksandrov's mean", SIAM J. Appl. Math. 76 (2016), p. 1792-1798.
- [14] I. Harriton, G. deBotton, "The nearly isotropic behaviour of high-rank nonlinear sequentially laminated composites", Proc. R. Soc. Lond. A 459 (2003), p. 157-174.
- [15] M. I. Idiart, "Modeling the macroscopic behavior of two-phase nonlinear composites by infinite-rank laminates", J. Mech. Phys. Solids 56 (2008), p. 2599-2617.
- [16] G. Francfort, P. Suquet, "Duality relations for nonlinear incompressible two-dimensional elasticity", Proc. R. Soc. Edinb. 131A (2001), p. 351-369.
- [17] J. M. Duva, "A self-consistent analysis of the stiffening effect of rigid inclusions on a power-law material", J. Eng. Mater. Technol. 106 (1984), p. 317-321.
- [18] O. Lopez-Pamies, "An exact result for the macroscopic response of particle-reinforced Neo-Hookean solids", J. Appl. Mech. 77 (2010), article no. 021016.
- [19] X. C. Zeng, D. J. Bergman, P. M. Hui, D. Stroud, "Effective medium theory for weakly nonlinear composites", *Phys. Rev. B* 38 (1988), p. 10970-10973.
- [20] O. Levy, D. J. Bergman, "Weakly nonlinear composites of random composites: a series expansion approach", J. Statist. Phys. 82 (1995), p. 1327-1344.
- [21] O. Levy, R. V. Kohn, "Duality relations for non-ohmic composites, with applications to behavior near percolation", J. Statist. Phys. 90 (1998), p. 159-189.
- [22] Y. P. Pellegrini, "Field distributions and effective-medium approximation for weakly nonlinear media", *Phys. Rev. B* 61 (2000), p. 9365-9372.
- [23] J. R. Willis, "The overall response of composite materials", ASME J. Appl. Mech. 50 (1983), p. 1202-1209.
- [24] Z. Hashin, S. Shtrikman, "A variational approach to the theory of the elastic behavior of multiphase materials", J. Mech. Phys. Solids 11 (1963), p. 127-140.
- [25] D. R. S. Talbot, J. R. Willis, "Variational principles for inhomogeneous nonlinear media", IMA J. Appl. Math. 35 (1985), p. 39-54.
- [26] P. Ponte Castañeda, "The effective mechanical properties of nonlinear isotropic composites", J. Mech. Phys. Solids 39 (1991), p. 45-71.
- [27] P. Ponte Castañeda, "Bounds and estimates for the properties of nonlinear heterogeneous systems", *Phil. Trans. R. Soc. Lond. A* 340 (1992), p. 531-567.

- [28] P. Suquet, "Overall potentials and extremal surfaces of power law or ideally plastic materials", J. Mech. Phys. Solids 41 (1993), p. 981-1002.
- [29] P. Gilormini, "A shortcoming of the classical nonlinear extension of the self-consistent model", C. R. Acad. Sci. Paris IIb–Mech. 320 (1995), p. 115-122.
- [30] R. Hill, "Continuum micro-mechanics of elastoplastic polycrystals", J. Mech. Phys. Solids 13 (1965), p. 89-101.
- [31] J. W. Hutchinson, "Bounds and self-consistent estimates for creep of polycrystalline materials", Proc. R. Soc. Lond. A 348 (1976), p. 101-127.
- [32] E. Kröner, "Berechnung der elastischen konstanten des vielkristalls aus den konstanten des einkristalls", Z. Phys. 151 (1958), p. 504-518.
- [33] P. Suquet, "Overall properties of nonlinear composites: a modified secant moduli theory and its link with Ponte Castañeda's nonlinear variational procedure", C. R. Acad. Sci. Paris II **320** (1995), p. 563-571.
- [34] P. Suquet, P. Ponte Castañeda, "Small-contrast perturbation expansions for the effective properties of nonlinear composites", C. R. Acad. Sci. Paris II 317 (1993), p. 1515-1522.
- [35] P. Ponte Castañeda, "Exact second-order estimates for the effective mechanical properties of nonlinear composites", J. Mech. Phys. Solids 44 (1996), p. 827-862.
- [36] P. Ponte Castañeda, "Stationary variational estimates for the effective response and field fluctuations in nonlinear composites", J. Mech. Phys. Solids 96 (2016), p. 660-682.
- [37] M. I. Idiart, P. Ponte Castañeda, "Field statistics in nonlinear composites. I. Theory", Proc. R. Soc. A 463 (2007), p. 183-202.
- [38] J. Furer, P. Ponte Castañeda, "A symmetric fully optimized second-order method for nonlinear homogenization", Z. Angew. Math. Mech. 98 (2018), p. 222-254.
- [39] J. R. Willis, "Variational estimates for the overall response of an inhomogeneous nonlinear dielectric", in *Homogenization and Effective Moduli of Materials and Media* (J. L. Ericksen *et al.*, eds.), Springer-Verlag, New York, 1986, p. 247-263.
- [40] P. Ponte Castañeda, P. Suquet, "Nonlinear composites", Adv. Appl. Mech. 34 (1998), p. 171-302.
- [41] G. W. Milton, The Theory of Composites, Cambridge University Press, Cambridge, UK, 2002.
- [42] M. I. Idiart, P. Ponte Castañeda, "Estimates for two-phase nonlinear conductors via iterated homogenization", Proc. R. Soc. A 469 (2013), article no. 20120626.
- [43] P. Ponte Castañeda, G. deBotton, G. Li, "Effective properties of nonlinear inhomogeneous dielectrics", Phys. Rev. B 46 (1992), p. 4387-4394.
- [44] P. Ponte Castañeda, M. Kailasam, "Nonlinear electrical conductivity in heterogeneous media", Proc. R. Soc. Lond. A 453 (1997), p. 793-816.
- [45] J. Furer, P. Ponte Castañeda, "On the optimality of the variational linear comparison bounds for porous viscoplastic materials", J. Mech. Phys. Solids 138 (2020), article no. 103898.
- [46] J. C. Michel, P. Suquet, "Effective potentials in nonlinear polycrystals and quadrature formulae", Proc. R. Soc. Lond. A 473 (2017), article no. 20170213.
- [47] I. Gradshteyn, I. M. Ryzhik, Table of Integrals, Series, and Products, Academic Press, London, UK, 1994.



Contributions in mechanics of materials

Mechanical dissimilarity of defects in welded joints via Grassmann manifold and machine learning

David Ryckelynck[®] *, ^a, Thibault Goessel^b and Franck Nguyen^a

^a Mines ParisTech PSL University, Centre des Matériaux, Evry, France

^b Mines ParisTech PSL University, France

E-mails: david.ryckelynck@mines-paristech.fr (D. Ryckelynck),

thibault.goessel@mines-paristech.fr (T. Goessel), franck.nguyen@mines-paristech.fr (F. Nguyen)

Abstract. Assessing the harmfulness of defects based on images is becoming more and more common in industry. At present, these defects can be inserted in digital twins that aim to replicate in a mechanical model what is observed on a component so that an image-based diagnosis can be further conducted. However, the variety of defects, the complexity of their shape, and the computational complexity of finite element models related to their digital twin make this kind of diagnosis too slow for any practical application. We show that a classification of observed defects enables the definition of a dictionary of digital twins. These digital twins prove to be representative of model-reduction purposes while preserving an acceptable accuracy for stress prediction. Nonsupervised machine learning is used for both the classification issue and the construction of reduced digital twins. The dictionary items are medoids found by a k-medoids clustering algorithm. Medoids are assumed to be well distributed in the training dataset according to a metric or a dissimilarity measurement. In this paper, we propose a new dissimilarity measurement between defects. It is theoretically founded according to approximation errors in hyper-reduced predictions. In doing so, defect classes are defined according to their mechanical effect and not directly according to their morphology. In practice, each defect in the training dataset is encoded as a point on a Grassmann manifold. This methodology is evaluated through a test set of observed defects totally different from the training dataset of defects used to compute the dictionary of digital twins. The most appropriate item in the dictionary for model reduction is selected according to an error indicator related to the hyper-reduced prediction of stresses. No plasticity effect is considered here (merely isotropic elastic materials), which is a strong assumption but which is not critical for the purpose of this work. In spite of the large variety of defects, we provide accurate predictions of stresses for most of defects in the test set.

Résumé. L'évaluation de la nocivité des défauts à partir d'images est de plus en plus courante dans l'industrie. Aujourd'hui, ces défauts peuvent être insérés dans des jumeaux numériques qui visent à reproduire dans un modèle mécanique ce qui est observé sur un composant. Ainsi, un diagnostic à partir d'image peut être mis en place. Mais la variété des défauts, la complexité de leur forme et la complexité de calcul des modèles d'éléments finis liés à leur jumeau numérique, rendent ce type de diagnostic trop lent pour toute application pratique. Nous montrons dans cet article qu'une classification des défauts observés permet de définir un dictionnaire des jumeaux numériques. Ces jumeaux numériques se révèlent représentatifs pour la réduction de modèle, tout en conservant une précision acceptable pour la prévision des contraintes. Un

^{*} Corresponding author.

apprentissage automatique non supervisé est utilisé à la fois pour la question de la classification et pour la construction de jumeaux numériques réduits. Les éléments du dictionnaire sont des médoïdes trouvés par l'algorithme de partitionnement k-médoïdes. Les médoïdes sont censés être bien répartis dans l'ensemble des données d'entraînement, selon une métrique ou une mesure de dissimilitude. Dans cet article, nous proposons une nouvelle mesure de dissimilitude entre les défauts. Elle est fondée théoriquement sur les erreurs d'approximation des prévisions hyperréduites. Ce faisant, les classes de défauts sont définies en fonction de leur effet mécanique et non directement en fonction de leur morphologie. En pratique, chaque défaut de l'ensemble de données d'entraînement est encodé comme un point sur une variété de Grassmann. Cette méthodologie est évaluée au moyen d'un ensemble de défauts tests totalement différents de l'ensemble de données d'apprentissage utilisé pour calculer le dictionnaire des jumeaux numériques. L'élément le plus approprié du dictionnaire, pour la réduction du modèle, est sélectionné en fonction d'un indicateur d'erreur lié à la prévision hyperréduite des contraintes. Aucun effet de plasticité n'est considéré ici (simplement des matériaux élastiques isotropes), ce qui est une hypothèse forte mais qui n'est pas critique pour l'objectif de ce travail. Malgré la grande variété de défauts, nous montrons des prévisions précises des contraintes pour la plupart des défauts de l'ensemble de test.

Keywords. Data encoding, Hyper-reduction, Reduced order model, ROM-net, Taxonomy of defects, Computer vision.

Mots-clés. Encodage de données, Hyper-réduction, Réduction d'ordre de modèles, ROM-net, Taxonomie de défauts, Vision par ordinateur.

Manuscript received 27th April 2020, revised 15th July 2020 and 23rd September 2020, accepted 23rd September 2020.

1. Introduction

Mechanical modeling based on images is becoming increasingly important in material science and in industrial applications for the assessment of harmfulness of defects. The early detection of defects in industrial processes has been studied for more than a decade. In fact, in 2011, scientists tried to improve the efficiency of early fault detection on gears, which are critical in many machinery operations [1]. Moreover, nondestructive inspection techniques are able to detect and locate voids for a wide range of materials and welding processes: resistance seam welding of aluminum, zinc, and galvanized steel [2]; resistance spot welding of ferritic/martensitic steels [3]; electron beam welding of steel to Fe–Al alloy [4]; laser welding of stainless steels [5] and aluminum alloys [6].

Here, machine learning enhances the value of data related to defects observed in the past if these data are available in a memory storage system. In the framework of image-based diagnosis, machine learning aims to consider the following assertion: if two defects are similar, they have equivalent harmfulness. Accounting for this similarity should facilitate the prediction of the harmfulness of new defects by using a training dataset of defects. The purpose of this paper is to sample a training dataset of defects so that a set of representative defects is defined. The representative defects are assumed to be well distributed in the training dataset according to an appropriate metric. This metric classifies defects according to the displacement fluctuations they cause around them. Here, the sets of both experimental data and simulation data, related to the representative defects, define a dictionary of digital twins. This dictionary contains the simulation data for the construction of local reduced order bases [7] in the nonparametric space of the observed defect. Section 2 presents a schematic view of these ideas.

Pure data-driven approaches have been detailed in the literature for defect diagnosis. Provided that a wide range of data is available, machine learning methods can eventually detect defects automatically and classify them into different classes given some prescribed criteria. For instance, supervised machine learning is used for defect classification issues related to a freezing process in [8]. Convolutional neural networks (CNNs) [9], which are very helpful in computer vision [10], have been trained to detect and diagnose defects. In [11], a CNN detects defects on an automotive damper. Another encouraging study published recently uses CNNs to diagnose defects on freight trains with a precision of approximately 80% [12]. An amazing property of CNNs is their ability to learn features, or kernels [10], solely by using labeled data and deep learning. However, in our opinion, using deep learning does not necessarily imply forgetting the available knowledge about the mechanics of materials when it comes to predicting the harmfulness of defects. The purpose of this paper is to couple machine learning and mechanical modeling so that transfer learning [13] is achieved.

In the mechanics of materials, image-based meshing methods [14,15] enable generating complex finite element meshes of digital images obtained via X-ray computed tomography [16]. The finite element models used as digital twins of mechanical components are fed by a huge knowledge on mechanical behavior for various materials, metals [17], composites [18], and concrete [19] for instance. Unfortunately, they cannot be used as a tool to assess the quality of a component in a serial production framework. The required fine meshes of defective components generally lead to prohibitive computational time as explained in [20]. However, the use of mechanical knowledge in digital twins would ensure data continuity between the component design phase and the diagnosis of defect harmfulness [20]. Besides, projection-based model order reduction methods enable reusing knowledge about materials science while using machine learning to generate approximation spaces for a fast solution of partial differential equations (PDEs) [21, 22]. Therefore, physics principles and material constitutive equations are preserved in weak formulations of partial derivative equations. In this paper, the machine learning task is restricted to the construction of an approximation space for the purpose of model order reduction. Here, this approximation space is piecewise constant over the space of all possible defects. Its numerical representation is a dictionary of digital twins based on representative defects. Usually, the manifold containing the solution of a PDE can be accurately depicted as residing in a small vector space. Therefore, linear machine learning methods, such as proper orthogonal decomposition [23], singular value decomposition (SVD), and noncentered principal component analysis, aim at learning this reduced vector space from simulation data. This subspace is then used as a single approximation space for the projection-based model order reduction of PDE. In practice, the finite element shape functions are replaced by vectors, or empirical modes, that span the reduced approximation space. In some situations, the solution of the governing PDEs lies in a manifold that cannot be covered by a single vector subspace without increasing its dimension, thus degrading the computational complexity of projection-based model reduction. In these cases, deep learning algorithms are useful for reducing the complexity of approximation spaces by using simulation data. For instance, in [24], physics-informed neural networks are proposed for solving supervised learning tasks while respecting any given laws of physics described by general nonlinear PDEs. These networks are no longer used as projection-based model order reduction schemes. However, such reduced schemes are found in [25] and [22], where a deep classifier recommends a reduced order model depending on input variables having a tensor format (e.g., images). In [25], a CNN recommends a reduced order model related to a loading environment seen on the image of an experimental setup. In [22], a deep classifier using CNN is trained to recommend hyperreduced order models for lifetime prediction depending on a three-dimensional (3D) stochastic temperature field. These two contributions follow the same neural network architecture termed ROM-net [22]. Such ROM-nets are trained by using simulation data encoded as points on a Grassmann manifold, which is a set of vector subspaces of given common dimensions in the same ambient space. This data encoding is quite general when considering simulation data in the framework of projection-based model order reduction. In this paper, it is extended for the classification of voids according to their mechanical effects on mechanical components.

In the current work, observational data are two-dimensional (2D) slices of experimental 3D images of voids. These 3D images have been obtained via X-ray computed tomography by Lacourt [26]. The reduced approximation space for displacements is spanned by two types of vectors: macroscopic modes of an ideal defect-free medium and fluctuation modes around each defect. By following the two-scale machine learning approach proposed in [20], the fluctuation modes are computed by assuming dilution conditions and scale separation for each defect separately. The volume fraction of the defect is negligible when computing the fluctuation modes. In linear elasticity, each defect admits an exact reduced basis of fluctuation modes for strains and displacements. For 2D problems, this ideal reduced basis contains three modes; for 3D problems, it contains six modes. This reduced basis is said to be ideal because its computation requires the finite element solution of an elastic problem, which is specific to the defect. In the proposed approach, such solutions are available only during the training phase of the approximation space. In the test phase, the ideal reduced basis is not available. The methodology, introduced in [22] for ROM-nets, aims to take in a dictionary the reduced approximation space related to a similar defect as a substitute for the ideal reduced basis. Here, this dictionary is called the dictionary of digital twins. This requires defining the following:

- a dissimilarity evaluation between defects,
- a dictionary of representative defects containing the related digital twins with a small number of items,
- a classifier that finds the best item in the dictionary for the construction of an approximation space dedicated to the target digital twin in a test set or for real application.

In this paper, the representative defects are medoids selected by the k-medoids algorithm [27]. The number of medoids has to be prescribed prior to data clustering via the k-medoids algorithm. Too many items in the dictionary make the classifier too complex and would eventually entail no complexity reduction for image-based modeling. In this paper, the best item in the dictionary is approximately selected via an error indicator and not via a CNN as proposed in [22]. This error indicator measures the discrepancy between the stresses predicted via hyper-reduction (HR) for the target digital twin and the equilibrated stresses [28]. A theoretical analysis of the convergence of the hyper-reduced approximation shows that a partial approximation error has an upper bound that scales linearly with the sines of the principal angles between the ideal modes and the modes involved in the approximation space. Identical reduced bases have principal angles equal to zero as well as sines. The convenient space to measure these angles is a Grassmann manifold [29, 30]. The sines of these angles are termed chordal distances [31]. Hence, the proposed dissimilarity criterion accounts for the mechanical effect of the defects via model reduction of displacement fluctuations around defects. It is not a direct evaluation of morphology dissimilarity.

For the sake of simplicity, the target problem used for defect diagnosis is similar to the micromechanical problem that defines the fluctuation modes. The reader can find in [20, 32, 33] more complex target mechanical problems that are solved by the HR method used in this paper. As the finite element approximation space is specific to each defect, we need to design a common ambient space for the computation of Grassmann distances. This is performed through an encoding mesh. We have paid particular attention to the compromise to be made between the accuracy of numerical approximations and the memory space required to save the simulation data related to the encoding mesh.

The present paper is structured as follows. Section 2 is a commented graphical abstract. Section 3 presents the projection-based model-reduction method, an upper bound for partial errors on displacement predictions, and the encoding mesh associated with the Grassmann manifold. Section 4 details the training dataset of defects, the partition of these data by using the



Figure 1. (a) Raw data in their original ambient space, (b) the proposed ambient space for dissimilarity measurement between defects according to a Grassmann distance related to the subspace spanned by fluctuation modes around each defect.

k-medoids algorithm in the Grassmann manifold, and some validation results. Section 5 presents the stress prediction results for the test set of defects and the classifier used to select an item from the dictionary of digital twins. Section 6 draws the conclusion of this paper.

2. Commented graphical abstract

Assume that a training dataset of defects is available with detailed digital twins and all related data. A schematic view of the training dataset is plotted in Figure 1. Training data are plotted as blue points in this figure.

We propose to select representative defects and their data to form a dictionary of digital twins for the purpose of model reduction. Representative defects are assumed to be well distributed in the training dataset according to a dissimilarity measurement. Two representative defects are plotted in red and green in Figure 1. The main novelty is a proper dissimilarity definition between defects so that the representative defects are medoids selected by using the k-medoids algorithm [27].

The original ambient space (Figure 1(a)) that contains the predictions of fluctuations around defects in the training dataset does not account for the linearity of elastic balance equations. We know that displacement fluctuations belong to a small vector subspace spanned by fluctuation modes, which is denoted by V^* for each defect individually. These vector subspaces are points in a manifold termed Grassmannian. As a result, prior to dissimilarity computation between defects, the training data are placed in a proper ambient space: a Grassmannian. A sketch of the Grassmannian ambient space is shown in Figure 1(b).

Displacement fluctuations or stress fluctuations around defects have magnitudes directly related to the loading magnitude defined in the mechanical target problem. They can be easily computed. This is the reason why fluctuation magnitudes do not matter in similar linear predictions. Then the Grassmannian ambient space is plotted as a unit circle. The geodesic distance on this circle is the angle θ shown in Figure 1(b). One can also consider in Figure 1 the chordal distance, which is the length of the straight line between two points on this circle for small values of θ . This chordal distance is closely related to an upper bound of partial approximation errors.



Figure 2. On the left, an experimental defect and its close-up. On the right, the horizontal component of $\Delta u^{\star(j)}$ for two traction modes related to $\mathbf{E}^{(1)}$ and $\mathbf{E}^{(2)}$ and one shear mode related to $\mathbf{E}^{(3)}$.

In what follows, the simulation data of the representative defects are the training data used for the HR of digital twins.

Data in the test set cannot be plotted in Figure 1 because their related fluctuation fields have to be predicted via the proposed dictionary-based model-reduction method.

3. Theoretical results on elastic fluctuation modes

3.1. Projection-based hyper-reduction of the target mechanical problem

This section describes the target mechanical problem used as a digital twin of a target defect. In the mechanics of materials, the harmfulness of defects is evaluated through the magnification of the Cauchy stress around each defect. We restrict our attention to the prediction of the Cauchy stress via the approximate solution of PDEs in linear elasticity of isotropic materials. The principal variable of these equations is the displacement field.

The domain occupied by the material, denoted by Ω^* , is a surrounding box around each defect. We give an example in Figure 2. Formally, Ω^* has no parameter. Its morphology totally depends on the observation of a defect through a digital image. In the following, the superscript \star refers to mathematical objects that are specific to the digital image of a defect. This notation emphasizes the variability due to the input image. All mathematical objects with the superscript \star can be seen as the output of an implicit function depending on Ω^* without introducing any parameter for Ω^* . We avoid giving a morphological definition of an input space that contains all possible values of Ω^* . Hence, the proposed setting is a nonparametric mechanical modeling for digital twins.

The outer boundary of the surrounding box is denoted by $\partial_E \Omega^*$. The boundary of the defect, which is modeled as a void, is denoted by $\partial_V \Omega^*$. The local frame follows the principal axes of the second-order moments of the volume distribution in the defect. These principal axes are denoted by \mathbf{e}_1 and \mathbf{e}_2 . All defects have a circumscribed circle of diameter equal to D. The length of Ω^* is 30D. The displacement field and the Cauchy stress are denoted by \mathbf{u}^* and $\boldsymbol{\sigma}^*$, respectively.

The fluctuation of the displacement field is denoted by $\Delta \mathbf{u}^*$. It is defined as the counterpart of the homogeneous displacement field such that

$$\mathbf{u}^{\star}(\mathbf{x}) = \mathbf{E} \cdot \mathbf{x} + \Delta \mathbf{u}^{\star}(\mathbf{x}), \quad \forall \mathbf{x} \in \Omega^{\star}, \quad \Delta \mathbf{u}^{\star} \in \mathcal{V}^{\star}, \tag{1}$$

where **E** is the macroscopic strain tensor imposed on $\partial_E \Omega^*$. It is a given symmetric second-order tensor. Here, **E** · **x** is the defect-free macroscopic mode for displacements; \mathcal{V}^* is the usual finite element approximation space that accounts for the defect geometry and Dirichlet boundary conditions (2) on $\partial_E \Omega^*$.

Here, Ω^* is sufficiently large to fulfill a dilution assumption and the following boundary condition on Δu^* :

$$\Delta \mathbf{u}^{\star}(\mathbf{x}) = 0, \quad \forall \mathbf{x} \in \partial_E \Omega^{\star}.$$
⁽²⁾

The Hooke tensor for elasticity is denoted by C. Hence, the elastic constitutive equation reads as

$$\boldsymbol{\sigma}^{\star} = \mathbf{C} : \boldsymbol{\varepsilon}(\mathbf{u}^{\star}), \tag{3}$$

where $\boldsymbol{\varepsilon}(\mathbf{u}^*)$ is the deformation tensor, that is, the symmetric part of the displacement gradient. The weak form of the elastic equilibrium equation reads as follows: find $\Delta \mathbf{u}^* \in \mathcal{V}^*$ such that

$$\Delta \mathbf{u}^{\star}(\mathbf{x}) = \sum_{i=1}^{\mathcal{N}^{\star}} \boldsymbol{\varphi}_{i}^{\star}(\mathbf{x}) \boldsymbol{q}_{i}^{\star}, \quad \forall \mathbf{x} \in \Omega^{\star}$$

$$\tag{4}$$

$$\int_{\Omega^{\star}} \boldsymbol{\varepsilon}(\boldsymbol{\varphi}_{i}^{\star}) : \mathbf{C} : (\mathbf{E} + \boldsymbol{\varepsilon}(\Delta \mathbf{u}^{\star})) \, \mathrm{d}\Omega = 0 \quad \forall i \in \{1, \dots, \mathcal{N}^{\star}\},$$
(5)

where $(\boldsymbol{\varphi}_i^{\star})_{i=1}^{\mathcal{N}^{\star}}$ are the finite element shape functions that span \mathcal{V}^{\star} such that $\boldsymbol{\varphi}_i^{\star} = 0$ on $\partial_E \Omega^{\star}$. Here, **E** is a symmetric second-order tensor; it has only three components for 2D problems. Therefore, as the elastic equations are linear, there exists an ideal reduced basis containing three fluctuation modes denoted by $(\boldsymbol{\psi}_k^{\star})_{k=1,\dots,3}$ such that

$$\Delta \mathbf{u}^{\star}(\mathbf{x}) = \sum_{k=1}^{N} \boldsymbol{\psi}_{k}^{\star}(\mathbf{x}) \overline{\boldsymbol{\gamma}}_{k}^{\star}, \quad \forall \mathbf{x} \in \Omega^{\star}, \quad \boldsymbol{\psi}_{k}^{\star} \in \mathcal{V}^{\star}, \quad k = 1, \dots, N$$
(6)

where $\overline{\gamma}^* \in \mathbb{R}^3$ is a vector of exact reduced coordinates. As the fluctuation modes are vectors of the approximation space, one can introduce the reduction matrix \mathbf{V}^* , which contains the finite element coordinates of the fluctuation modes:

$$\boldsymbol{\psi}_{k}^{\star}(\mathbf{x}) = \sum_{i=1}^{\mathcal{N}^{\star}} \boldsymbol{\varphi}_{i}^{\star}(\mathbf{x}) V_{ik}^{\star}, \quad k = 1, 2, 3, \quad \forall \mathbf{x} \in \Omega^{\star}.$$
(7)

In what follows, approximate reduced bases for fluctuation modes are denoted by **V**. In Figure 1(b), V^* is represented by the blue arrow and **V** is represented by the red arrow. The parameter **V** may not be totally specific to Ω^* . The approximate continuous modes are

$$\boldsymbol{\psi}_{k}(\mathbf{x}) = \sum_{i=1}^{\mathcal{N}^{\star}} \boldsymbol{\varphi}_{i}^{\star}(\mathbf{x}) V_{ik}, \quad k = 1, 2, 3, \quad \forall \mathbf{x} \in \Omega^{\star}.$$
(8)

For a given reduced matrix V, the approximate displacement fluctuation reads as

$$\Delta \mathbf{u}(\mathbf{x}) = \sum_{k=1}^{N} \boldsymbol{\psi}_{k}(\mathbf{x}) \boldsymbol{\gamma}_{k}, \quad \forall \mathbf{x} \in \Omega^{\star}.$$
(9)

The two reduced bases span two vector spaces of the same dimension N in the same ambient space $\mathbb{R}^{\mathcal{N}^*}$, $N < \mathcal{N}^*$. Each of these vector spaces is a point in a Grassmann manifold, which is denoted by $\operatorname{Gr}(N, \mathcal{N}^*)$. This manifold is a huge space as it contains all the vector subspaces of dimension N in $\mathbb{R}^{\mathcal{N}^*}$. For instance, it contains also the vector subspaces spanned by all sets of N vibration modes. Such vibration modes are not relevant here. However, the Grassmann manifold is equipped with geodesic metrics, which is very convenient for the definition of the dissimilarity

between defects, as explained in Section 3.2, when considering approximation errors due to the HR method.

The HR method [34] aims at computing reduced coordinates γ introduced in (9) by projecting the equilibrium equation on **V** via a restriction of the domain Ω^* to a reduced integration domain (RID) denoted by Ω_R^* . By following the empirical interpolation method [35], interpolation points are computed for column vectors in **V**. We choose the RID Ω_R^* such that it contains the interpolation points related to both the reduced basis for displacement and a reduced basis for stresses. We give more details about the construction of Ω_R^* in Appendix. We define a set of test reduced functions denoted by Ψ_{Ri} :

$$\mathscr{F} = \left\{ i \in \{1, \dots, \mathscr{N}^{\star}\}, \int_{\Omega^{\star} \setminus \Omega_R^{\star}} (\boldsymbol{\varphi}_i^{\star})^2 \,\mathrm{d}\Omega = 0 \right\}$$
(10)

$$\boldsymbol{\psi}_{Rj}(\mathbf{x}) = \sum_{i \in \mathscr{F}}^{\mathscr{N}^{\star}} \boldsymbol{\varphi}_{i}^{\star}(\mathbf{x}) V_{ij}, \quad \forall \mathbf{x} \in \Omega^{\star}, \quad j = 1, 2, 3.$$
(11)

As explained in [34], these test functions are null on the interface between Ω_R^{\star} and the counterpart of the domain as if Dirichlet boundary conditions were imposed. On this interface, the displacement follows the shape of the modes ψ_k . The HR method gives access to reduced coordinates γ that fulfill the following balance equations:

$$\Delta \mathbf{u}(\mathbf{x}) = \sum_{k=1}^{N} \boldsymbol{\psi}_{k}(\mathbf{x}) \boldsymbol{\gamma}_{k}, \quad \forall \mathbf{x} \in \Omega_{R}^{\star}$$
(12)

$$\int_{\Omega_R^*} \boldsymbol{\varepsilon}(\boldsymbol{\psi}_{Rj}) : \mathbf{C} : (\mathbf{E} + \boldsymbol{\varepsilon}(\Delta \mathbf{u})) \, \mathrm{d}\Omega = 0, \quad \forall j = 1, \dots, N.$$
(13)

The matrix form of the hyper-reduced balance equations reads as follows: find $\gamma \in \mathbb{R}^N$ such that

$$\Delta \mathbf{u}(\mathbf{x}) = \sum_{i=1}^{\mathcal{N}^{\star}} \boldsymbol{\varphi}_{i}^{\star}(\mathbf{x}) q_{i}^{\mathrm{HR}}, \quad \forall \mathbf{x} \in \Omega_{R}^{\star}$$
(14)

$$\mathbf{q}^{\mathrm{HR}} = \mathbf{V}\boldsymbol{\gamma} \tag{15}$$

$$\mathbf{K}^{\mathrm{HR}}\boldsymbol{\gamma} = \mathbf{V}[\mathscr{F},:]^{T} \mathbf{F}^{\star}[\mathscr{F}]$$
(16)

$$\mathbf{K}^{\mathrm{HR}} = \mathbf{V}[\mathscr{F},:]^{T} \mathbf{K}^{\star} [\mathscr{F},:] \mathbf{V}$$
(17)

$$K_{ij}^{\star} = \int_{\Omega^{\star}} \boldsymbol{\varepsilon}(\boldsymbol{\varphi}_{i}^{\star}) : \mathbf{C} : \boldsymbol{\varepsilon}(\boldsymbol{\varphi}_{j}^{\star}) \,\mathrm{d}\Omega, \quad i, j = 1, \dots, \mathcal{N}^{\star}$$
(18)

$$F_i^{\star} = -\int_{\Omega^{\star}} \boldsymbol{\varepsilon}(\boldsymbol{\varphi}_i^{\star}) : \mathbf{C} : \mathbf{E} \,\mathrm{d}\Omega, \quad i = 1, \dots, \mathcal{N}^{\star},$$
(19)

where $\mathbf{V}[\mathscr{F},:]$ denotes a row restriction of matrix \mathbf{V} to indices in \mathscr{F} . We assume that the matrix \mathbf{K}^{HR} is of full rank. This assumption is always checked in numerical solutions of hyper-reduced equations. Rank deficiency may appear when the RID construction does not account for the contribution of a reduced basis dedicated to stresses.

In this paper, the hyper-reduced prediction is supplemented by the following equilibrium step over the RID: find $\delta \mathbf{u}^*$ such that

$$\delta \mathbf{u}^{\star}(\mathbf{x}) = \sum_{i \in \widetilde{\mathscr{F}}} \boldsymbol{\varphi}_{i}^{\star}(\mathbf{x}) \delta q_{i}^{\star}, \quad \forall \mathbf{x} \in \Omega_{R}^{\star}$$
(20)

$$\int_{\Omega_R^*} \boldsymbol{\varepsilon}(\boldsymbol{\varphi}_i^*) : \mathbf{C} : (\mathbf{E} + \boldsymbol{\varepsilon}(\Delta \mathbf{u} + \delta \mathbf{u}^*)) \, \mathrm{d}\Omega = 0 \quad \forall i \in \widetilde{\mathscr{F}},$$
(21)

where $\widetilde{\mathscr{F}}$ is the set of all degrees of freedom in Ω_R^* except those belonging to elements connected to the interface between Ω_R^* and its counterpart. During this correction step, displacements are frozen on elements connected to the interface between Ω_R^* and its counterpart. There is no natural boundary condition on this interface. Here, they are forecast by the hyper-reduced prediction. This correction step has been proposed in [36] for the evaluation of contact forces. This is a local correction step over Ω_R^* . The solution $\mathbf{u}^{eq} = \mathbf{E}\mathbf{x} + \Delta \mathbf{u} + \delta \mathbf{u}^*$ is a hybrid solution that weakly couples HR and a finite element approximation over the RID. We refer the reader to [36–38] for more details about hybrid HR schemes. The equilibrated stress is quite accurate compared to the stress computed via HR solely. The hyper-reduced stress prediction is denoted by $\boldsymbol{\sigma}^{\text{HR}}$, while the equilibrated stress is denoted by $\boldsymbol{\sigma}^{\text{eq}}$. Here, $\delta \boldsymbol{\sigma}^*$ is the correction term for stress predictions:

$$\boldsymbol{\sigma}^{\mathrm{HR}} = \mathbf{C} : (\mathbf{E} + \boldsymbol{\varepsilon}(\Delta \mathbf{u})) \tag{22}$$

$$\delta \boldsymbol{\sigma}^{\star} = \mathbf{C} : \boldsymbol{\varepsilon}(\delta \mathbf{u}^{\star}) \tag{23}$$

$$\boldsymbol{\sigma}^{\mathrm{eq}} = \boldsymbol{\sigma}^{\mathrm{HR}} + \delta \boldsymbol{\sigma}^{\star}. \tag{24}$$

Property 1. If \mathbf{K}^{HR} is of full rank, then the hyper-reduced balance equations are equivalent to an oblique projection of the finite element prediction:

$$\mathbf{q}^{\mathrm{HR}} = \mathbf{V} (\mathbf{\Pi}^T \mathbf{V})^{-1} \mathbf{\Pi}^T \mathbf{q}^{\star}, \qquad (25)$$

where $\mathbf{\Pi} = \mathbf{K}^{\star}[:, \mathcal{F}]\mathbf{V}[\mathcal{F}, :]$. Hence, the hyper-reduced prediction of the reduced coordinate vector $\boldsymbol{\gamma}$ is a minimizer for $f(\boldsymbol{\beta})$:

$$\boldsymbol{\beta} \in \mathbb{R}^{N}, \quad f(\boldsymbol{\beta}) = \|\boldsymbol{\Pi}^{T}(\mathbf{V}\boldsymbol{\beta} - \mathbf{q}^{\star})\|_{2}^{2}.$$
(26)

Here, Π is a projector for elastic stresses in Ω_{R}^{\star} according to the reduced test functions:

$$\sum_{i=1}^{\mathcal{N}^{\star}} \Pi_{ik} (\mathbf{V} \boldsymbol{\gamma} - \mathbf{q}^{\star})_{i} = \int_{\Omega_{R}^{\star}} \boldsymbol{\varepsilon}(\boldsymbol{\psi}_{Rk}) : (\boldsymbol{\sigma}^{\mathrm{HR}} - \boldsymbol{\sigma}^{\star}) \,\mathrm{d}\Omega,$$
(27)

where σ^{\star} is the finite element stress prediction.

The proof is straightforward. Here, $\mathbf{K}^{\text{HR}} = \mathbf{\Pi}^T \mathbf{V}$. The Jacobian matrix for f reads as $\mathbf{J} = \mathbf{V}^T \mathbf{\Pi} \mathbf{\Pi}^T \mathbf{V} = (\mathbf{K}^{\text{HR}})^T \mathbf{K}^{\text{HR}}$. If \mathbf{K}^{HR} is of full rank, then \mathbf{J} is symmetric definite positive and $\mathbf{J}^{-1} = (\mathbf{K}^{\text{HR}})^{-1}$. Then, both the minimization problem and the hyper-reduced equation have a unique solution. The solution of the minimization problem is

$$\mathbf{q}^f = \mathbf{V}(\mathbf{J})^{-1} \mathbf{V}^T \mathbf{\Pi} \mathbf{\Pi}^T \mathbf{q}^{\star}$$
(28)

$$= \mathbf{V}(\mathbf{K}^{\mathrm{HR}})^{-1} \mathbf{\Pi}^T \mathbf{q}^{\star}.$$
 (29)

Since $\Pi^T \mathbf{q}^* = \mathbf{V}[\mathscr{F}, :]^T \mathbf{F}^* [\mathscr{F}], \mathbf{q}^f = \mathbf{q}^{\mathrm{HR}}.$

We can note that if $\Omega_R^* = \Omega^*$, $\widetilde{\mathscr{F}}$ contains all degrees of freedom indices and $\delta \mathbf{u}^* = \Delta \mathbf{u}^* - \Delta \mathbf{u}$. However, the correction step has the same computational complexity as the full finite element model. In this case, the size of Ω^* has been carefully chosen so that the reduction in complexity is not trivial. Here, the most complex operations are indeed the computation of \mathbf{K}^{HR} and the solution of the reduced linear system of equations. They scale linearly with $\operatorname{card}(\mathscr{F})N^2$ and N^3 , respectively. Hence, N^3 has to be sufficiently small compared to \mathscr{N}^* if we consider the computational complexity for the solution of sparse linear systems in the finite element method.

3.2. Approximation errors

Let us introduce three canonical macroscopic strains, $\mathbf{E}^{(1)}$, $\mathbf{E}^{(2)}$, and $\mathbf{E}^{(3)}$:

$$\mathbf{E}^{(1)} = \boldsymbol{\varepsilon}(x_1 \mathbf{e}_1) \tag{30}$$

$$\mathbf{E}^{(2)} = \boldsymbol{\varepsilon}(x_2 \mathbf{e}_2) \tag{31}$$

$$\mathbf{E}^{(3)} = \boldsymbol{\varepsilon} \left(\frac{x_1 \mathbf{e}_2 + x_2 \mathbf{e}_1}{2} \right). \tag{32}$$

In what follows, the three finite element solutions obtained for each of the three canonical macroscopic strains $E^{(1)}$, $E^{(2)}$, and $E^{(3)}$ are saved in a matrix Q^{\star} such that the three displacement fields read as

$$\mathbf{u}^{\star(j)}(\mathbf{x}) = \mathbf{E}^{(j)} \cdot \mathbf{x} + \Delta \mathbf{u}^{\star(j)}(\mathbf{x}) \quad \forall \mathbf{x} \in \Omega^{\star}, \quad j = 1, 2, 3$$
(33)

$$\Delta \mathbf{u}^{\star(j)}(\mathbf{x}) = \sum_{i=1}^{\mathcal{N}} \boldsymbol{\varphi}_i^{\star}(\mathbf{x}) Q_{ij}^{\star}, \quad \forall \mathbf{x} \in \Omega^{\star}, \quad j = 1, 2, 3.$$
(34)

All mechanical simulations are run with the Z-set software suite. One can find more information on the software website (http://www.zset-software.com/). The reduced basis V^* is obtained by using a truncated SVD of Q^* :

$$\mathbf{Q}^{\star} = \mathbf{V}^{\star} \mathbf{S}^{\star} \mathbf{W}^{\star T}, \quad \mathbf{V}^{\star} \in \mathbb{R}^{\mathcal{N}^{\star} \times 3}, \quad \mathbf{V}^{\star T} \mathbf{V}^{\star} = \mathbf{I},$$
(35)

where **I** is the 3×3 identity matrix. Because of the linearity of the elastic problem above, **C** is proportional to the Young modulus. Hence, **V**^{*} does not depend on the value of the Young modulus thanks to the normalization of the modes. Regarding the Poisson coefficient, for the sake of simplicity, we restrict our attention to isotropic materials having a Poisson coefficient equal to 0.3. An example of displacement fluctuations is shown in Figure 2.

Similarly to Céa's lemma, but in the finite dimension, there exists an upper bound for the approximation error observed through the projector Π . The best projection of the exact solution in the approximation space via the 2-norm is denoted by γ_p :

$$\boldsymbol{\gamma}_{P} = \operatorname{argmin}_{\mathbf{q} = \mathbf{V}\mathbf{g}} \| \mathbf{q}^{\star} - \mathbf{q} \|_{2}^{2}, \tag{36}$$

where $\mathbf{q}^{\star} = \mathbf{V}^{\star} \overline{\boldsymbol{\gamma}}^{\star}$ and $\mathbf{K}^{\star} \mathbf{q}^{\star} = \mathbf{F}^{\star}$. Hence, $\boldsymbol{\gamma}_{P} = \mathbf{V}^{T} \mathbf{V}^{\star} \overline{\boldsymbol{\gamma}}^{\star}$.

Property 2. There exists a stability coefficient c^* , which does not depend on F^* (the loading condition), such that the partial approximation error has the following upper bound:

$$\|\mathbf{\Pi}^{T}(\mathbf{q}^{\star} - \mathbf{V}\boldsymbol{\gamma})\|_{2} \le c^{\star} \|\mathbf{q}^{\star} - \mathbf{V}\boldsymbol{\gamma}_{P}\|_{2},$$
(37)

where $\|\mathbf{\Pi}^T(\mathbf{q}^* - \mathbf{V}\boldsymbol{\gamma})\|_2$ is the partial approximation error that does not account for errors in stress predictions outside Ω_R^* . Hence, the smaller the Euclidean distance between the subspace spanned by \mathbf{V} and the finite element prediction, the better the prediction of the stress in Ω_R^* according to the projector $\mathbf{\Pi}$. It is therefore relevant to train \mathbf{V} by using a training dataset of defects. When the RID covers the full domain, a certification of the reduced projection can be achieved, where all errors admit an upper bound, by following the constitutive relation error proposed in [39, 40].

The proof of the previous property is straightforward in the finite dimension. Let us denote by α^* an upper bound of the highest singular value of **II**. Then,

$$\|\mathbf{\Pi}^{T}(\mathbf{q}^{\star}-\mathbf{q})\|_{2}^{2} \leq (\alpha^{\star})^{2}\|\mathbf{q}^{\star}-\mathbf{q}\|_{2}^{2} \quad \forall \mathbf{q}$$

Moreover, as $\boldsymbol{\gamma}$ is a minimizer for $f(\cdot)$, we obtain

$$\|\mathbf{\Pi}^{T}(\mathbf{q}^{\star} - \mathbf{V}\boldsymbol{\gamma})\|_{2}^{2} \leq \|\mathbf{\Pi}^{T}(\mathbf{q}^{\star} - \mathbf{V}\boldsymbol{\gamma}_{P})\|_{2}^{2}$$
(38)

and

$$\|\mathbf{\Pi}^{T}(\mathbf{q}^{\star} - \mathbf{V}\boldsymbol{\gamma})\|_{2}^{2} \le (\alpha^{\star})^{2} \|\mathbf{q}^{\star} - \mathbf{V}\boldsymbol{\gamma}_{P}\|_{2}^{2}.$$
(39)

Property 3. If $\mathbf{V}^* \in \mathbb{R}^{\mathcal{N}^* \times N}$ and $\mathbf{V} \in \mathbb{R}^{\mathcal{N}^* \times N}$ are two orthonormal matrices of the same ambient space $\mathbb{R}^{\mathcal{N}^*}$, then they span subspaces that belong to the same Grassmann manifold. The partial approximation error has an upper bound depending on the chordal distance [31], denoted by $d^{\operatorname{Ch}}(\mathbf{V}^*, \mathbf{V})$, between the subspaces spanned by \mathbf{V}^* and \mathbf{V} , respectively:

$$\|\mathbf{\Pi}^{T}(\mathbf{q}^{\star} - \mathbf{V}\boldsymbol{\gamma})\|_{2} \le \alpha^{\star} d^{\mathrm{Ch}}(\mathbf{V}^{\star}, \mathbf{V})\sqrt{N} \|\overline{\boldsymbol{\gamma}}^{\star}\|_{2}.$$
(40)
Here, the chordal distance [31] uses the principal angles $\boldsymbol{\theta} \in \mathbb{R}^N$, $\theta_k \in [0, \pi/2]$ *for* k = 1, ... N *computed via the following SVD:*

$$\mathbf{V}^{T}\mathbf{V}^{\star} = \mathbf{U}\cos(\boldsymbol{\theta})\mathbf{U}^{\star T}, \quad \mathbf{U}^{T}\mathbf{U} = \mathbf{U}^{\star T}\mathbf{U}^{\star} = \mathbf{I}$$
(41)

$$d^{\mathrm{Ch}}(\mathbf{V}^{\star},\mathbf{V}) = \|\sin(\boldsymbol{\theta})\|_{F}$$

$$\tag{42}$$

$$\|\mathbf{U}^{\star}\|_{F}^{2} = N,\tag{43}$$

where $\|\cdot\|_F$ is the Frobenius norm. Here, $\cos(\theta)$ and $\sin(\theta)$ are the cosine and sine diagonal matrices, respectively. In addition, the following property holds when a full SVD is computed:

$$\mathbf{U}^{\star}\mathbf{U}^{\star T} = \mathbf{I}.\tag{44}$$

The proof is the following:

$$\|\mathbf{q}^{\star} - \mathbf{V}\boldsymbol{\gamma}_{P}\|_{2}^{2} = \overline{\boldsymbol{\gamma}}^{\star T} (\mathbf{V}^{\star} - \mathbf{V} \mathbf{V}^{T} \mathbf{V}^{\star})^{T} (\mathbf{V}^{\star} - \mathbf{V} \mathbf{V}^{T} \mathbf{V}^{\star}) \overline{\boldsymbol{\gamma}}^{\star} = \overline{\boldsymbol{\gamma}}^{\star T} (\mathbf{I} - \mathbf{V}^{\star T} \mathbf{V} \mathbf{V}^{T} \mathbf{V}^{\star}) \overline{\boldsymbol{\gamma}}^{\star}.$$
(45)

Hence,

$$\|\mathbf{q}^{\star} - \mathbf{V}\boldsymbol{\gamma}_{P}\|_{2}^{2} = \overline{\boldsymbol{\gamma}}^{\star T} (\mathbf{I} - \mathbf{U}^{\star} \cos(\boldsymbol{\theta})^{2} \mathbf{U}^{\star T}) \overline{\boldsymbol{\gamma}}^{\star}$$
(46)

$$\|\mathbf{q}^{\star} - \mathbf{V}\boldsymbol{\gamma}_{P}\|_{2}^{2} = \overline{\boldsymbol{\gamma}}^{\star T} \mathbf{U}^{\star} (\mathbf{I} - \cos(\boldsymbol{\theta})^{2}) \mathbf{U}^{\star T} \overline{\boldsymbol{\gamma}}^{\star}$$

$$\tag{47}$$

$$\|\mathbf{q}^{\star} - \mathbf{V}\boldsymbol{\gamma}_{P}\|_{2}^{2} = \overline{\boldsymbol{\gamma}}^{\star T} \mathbf{U}^{\star} \sin(\boldsymbol{\theta})^{2} \mathbf{U}^{\star T} \overline{\boldsymbol{\gamma}}^{\star}$$
(48)

$$= \|\sin(\boldsymbol{\theta})\mathbf{U}^{\star T}\overline{\boldsymbol{\gamma}}^{\star}\|_{2}^{2}.$$
(49)

For all matrices $\mathbf{A} \in \mathbb{R}^{n \times m}$ and $\mathbf{B} \in \mathbb{R}^{m \times n}$, the following property holds:

$$\|\mathbf{A}\mathbf{B}\|_F \leq \|\mathbf{A}\|_F \|\mathbf{B}\|_F,$$

and for $\mathbf{a} \in \mathbb{R}^n$: $\|\mathbf{a}\|_F = \|\mathbf{a}\|_2$.

Thus,

$$\|\mathbf{q}^{\star} - \mathbf{V}\boldsymbol{\gamma}_{P}\|_{2}^{2} \le \|\sin(\boldsymbol{\theta})\mathbf{U}^{\star T}\|_{F}^{2}\|\overline{\boldsymbol{\gamma}}^{\star}\|_{2}^{2} \le \|\sin(\boldsymbol{\theta})\|_{F}^{2}N\|\overline{\boldsymbol{\gamma}}^{\star}\|_{2}^{2}$$
(50)

and

$$\|\mathbf{\Pi}^{T}(\mathbf{q}^{\star} - \mathbf{V}\boldsymbol{\gamma})\|_{2} \leq \alpha^{\star} \|\sin(\boldsymbol{\theta})\|_{F} \sqrt{N} \|\overline{\boldsymbol{\gamma}}^{\star}\|_{2}.$$
(51)

Property 3 is a convergence property for hyper-reduced predictions. If $\theta = 0$, then $VU = V^*U^*$ and

$$0 = \mathbf{\Pi}^T (\mathbf{V}^* \boldsymbol{\gamma}^* - \mathbf{V} \boldsymbol{\gamma}) \tag{52}$$

$$= \mathbf{\Pi}^T \mathbf{V} \mathbf{U} (\mathbf{U}^{\star T} \boldsymbol{\gamma}^{\star} - \mathbf{U}^T \boldsymbol{\gamma})$$
(53)

$$= \mathbf{K}^{\mathrm{HR}} \mathbf{U} (\mathbf{U}^{\star T} \boldsymbol{\gamma}^{\star} - \mathbf{U}^{T} \boldsymbol{\gamma})$$
(54)

$$\Rightarrow \boldsymbol{\gamma} = \boldsymbol{\mathbf{U}} \boldsymbol{\mathbf{U}}^{\star T} \boldsymbol{\gamma}^{\star}$$
(55)

$$\Rightarrow \mathbf{q}_{HR} = \mathbf{q}^{\star} \quad \text{and} \quad \boldsymbol{\sigma}^{HR} = \boldsymbol{\sigma}^{\star}. \tag{56}$$

Hence, the hyper-reduced prediction is the same as the finite element prediction. However, it uses fewer floating-point operations.

In what follows, we assume that there is a correlation between the approximation error $\sigma^{\text{HR}} - \sigma^*$ in Ω_R^* and the upper bound in (40). This correlation is evaluated through a numerical experiment in Section 4.1.

3.3. Grassmannian ambient space

As explained in [29, 30], Grassmann manifolds are the adequate concept when considering interpolation of reduced order models. Property 3 shows that it is also relevant for data encoding in a proper ambient space and for the classification of voids according to their mechanical effects. We state that the target defect Ω^* is similar to a defect having **V** for the ideal reduced basis if



Figure 3. Encoding mesh with four elements on each half-side of the middle square having a regular mesh. From left to right: the surrounding box, a close-up, and the same close-up of a defect.

the chordal distance $d^{Ch}(\mathbf{V}^*, \mathbf{V})$ is sufficiently small. In what follows, each defect in the training dataset is encoded by using its ideal reduced basis \mathbf{V}^* as shown in Figure 1(b).

A common Grassmann manifold is defined for all defects by using a common encoding mesh. Each finite element prediction related to the training dataset of defects is transferred on this encoding mesh prior to any machine learning including SVD and dissimilarity computations. To facilitate the transfer of simulation data, the prediction $\Delta \mathbf{u}^{\star(j)}$ is extended inside the defect by introducing a very small Young modulus in the void $(10^{-3}E)$. Figure 3 shows the encoding mesh.

This data encoding from the reduced mechanical response of the input, has the huge advantage to be universal. This encoding may be applied in other fields. Each data in the dataset is supplemented by a reduced basis $\mathbf{V}^{\star(i)}$, where *i* is the defect index.

Several geodesic distances are available between points in Grassmann manifolds. The chordal distance is one of them. The Grassmann distance is another one, which is more common in the literature. The Grassmann distance between the subspaces spanned by V^* and V, respectively, is denoted by $d^{Gr}(V^*, V)$. It reads as

$$d^{\mathrm{Gr}}(\mathbf{V}^{\star},\mathbf{V}) = \frac{\|\boldsymbol{\theta}\|_2}{\pi/2}.$$
(57)

In the following numerical experiment, the Grassmann distance magnifies the distance between subspaces compared to the chordal distance similarly to the one-dimensional equation $\theta \ge \sin(\theta)$ for $\theta \in [0, \pi/2]$. As a result, we choose d^{Gr} for the partition of the training dataset of defects into classes of defects.

The encoding mesh must have the number of degrees of freedom \mathcal{N} as small as possible to lower the memory use in a storage system, but it must be sufficiently fine to

- lower the norm of transfer errors from the original mesh to the encoding mesh;
- · detect dissimilarities between defects up to a given accuracy.

To highlight the influence of the encoding mesh on transfer errors, we varied the number of elements on each half-side of the middle square that has a regular mesh. Figure 4 presents the convergence of the Grassmann distance between a perfect circle and an isotropic defect or an anisotropic defect. Both isotropic and anisotropic defects are shown in Figure 5. Grassmann distances are calculated for several encoding meshes. The ideal reduced basis for the fluctuation modes around the circular defect is denoted by $\mathbf{V}^{\star(0)}$. Here, $\mathbf{V}^{\star(1)}$ and $\mathbf{V}^{\star(2)}$ denote the reduced



Figure 4. Convergence of Grassmann distances with respect to the encoding mesh for the evaluation of dissimilarity between a circular and an anisotropic (d_1) or an isotropic defect (d_2) .



Figure 5. Examples of defects (defect in blue, steel in red). The isotropic defect (left) and the anisotropic defect (right).

bases related to the anisotropic and isotropic defects, respectively. The two Grassmann distances are

$$d_1 = d^{\rm Gr}(\mathbf{V}^{\star(1)}, \mathbf{V}^{\star(0)}) \tag{58}$$

$$d_2 = d^{\rm Gr}(\mathbf{V}^{\star(2)}, \mathbf{V}^{\star(0)}). \tag{59}$$

In Figure 4, the Grassmann distances and the encoding mesh successfully distinguish the two defects, and we observe a convergence of the distances when the mesh exceeds four elements on each half-side of the middle square. A difference of 20% in the Grassmann distance between the two defects is consistent as the pilot circular defect and the anisotropic defect are mechanically very different. The gap may be sufficient to distinguish them during the partition of training data. Therefore, we suggest keeping the encoding mesh with 609 nodes (four elements on each half-side).

4. Numerical results on the training dataset of defects

Three-dimensional images of voids have been obtained by Laurent Lacourt [20] via X-ray computed tomography of welded joints. These 3D images have been cut into slices so that we have more 2D samples. The dataset contains $n_I = 2,745$ samples of 2D images with only one defect per image. However, one defect may involve several voids that have a high mechanical interaction. Eighty percent of these data have been randomly selected as training data. The remaining data belong to the test set.

4.1. Data clustering

Clustering methods have been already used for the construction of local reduced order bases in parametric spaces. We refer the reader to [41, 42], for instance, and for more recent examples to [43]. Local reduced order bases are known to be more accurate over a global reduced basis [41]. Here, the set of possible defects is not a parametric space. For this reason, k-means is useless because no barycentric coordinates are available here. The k-medoids algorithm [27] circumvents this difficulty in the case of nonparametric modeling via the selection of representative defects.

All available images in the dataset have been converted into finite element predictions via image-based digital twins. Displacement fluctuations have been transferred to the encoding mesh prior to the computation of the related reduced basis $\mathbf{V}^{\star(i)}$ ($i = 1, ..., n_I$). Then, a dissimilarity matrix has been computed, accounting for all the Grassmann distances between defects in the training dataset:

$$D_{i\,i}^{\mathrm{Gr}} = d^{\mathrm{Gr}}(\widehat{\mathbf{V}}^{\star(i)}, \widehat{\mathbf{V}}^{\star(j)}), \quad i, j \in \{1, \dots, n_I\}.$$

$$(60)$$

A k-medoids clustering algorithm [27] has been used for the partition of data according to their Grassmann distance. The k-medoids algorithm proposed in [27] can be summarized as follows:

- Initialization step: select *K* rows in \mathbf{D}^{Gr} as indices of initial medoids (m_1, \dots, m_K) .
- · Repeat the following two steps until convergence:
 - Data assignment step: assign each point of the dataset to the cluster corresponding to its closest medoid:

$$\mathscr{L}_{i}^{\mathrm{Gr}} = \operatorname{argmin}_{k \in \{1, \dots, K\}} (\mathbf{D}^{\mathrm{Gr}}[[m_{1}, \dots, m_{k}, \dots], i]) \quad i \in \operatorname{training} \operatorname{dataset}.$$
(61)

 Medoid update step: for each cluster, update the medoid by finding the point that minimizes the sum of distances to all the points in the cluster:

$$\mathscr{C}_{k} = \{i \in \text{training dataset} | \mathscr{L}_{i}^{\text{Gr}} = k\}, \quad k = 1, \dots, K$$
(62)

$$m_{k} = \operatorname{argmin}_{j \in \mathscr{C}_{k}} \left(\sum_{i \in \mathscr{C}_{k}} \mathbf{D}^{\operatorname{Gr}}[j, i] \right).$$
(63)

We arbitrarily set the number of clusters to K = 5. The set \mathscr{C}_k contains the indices of defects in the cluster number k. Hence, the sizes of the clusters are $\operatorname{card}(\mathscr{C}_1) = 554$, $\operatorname{card}(\mathscr{C}_2) = 200$, $\operatorname{card}(\mathscr{C}_3) = 202$, $\operatorname{card}(\mathscr{C}_4) = 901$, and $\operatorname{card}(\mathscr{C}_5) = 339$.

The defects located in the medoids are shown in Figure 6. These defects have different anisotropies. The most isotropic defect is m_4 . The most anisotropic defects are m_2 and m_3 . These last defects are rather symmetric. In future work, this symmetry should be removed from the dataset.

Defects m_2 and m_3 contain two voids. They are so close to each other that fluctuation modes account for the local interactions between these voids.

4.2. Validation of hyper-reduced predictions for the training dataset

The meta-parameters of HR are set up by using the simulation data available in the training dataset.

For each medoid $m^{(k)}$ and for each target image number *i*, we define an image-specific hyperreduced model denoted by HR(k, i) without using the encoding mesh here. The hyper-reduced





model incorporates a reduced basis for displacement fluctuations and a reduced basis for stresses $(\mathbf{V}^{(k,i)}, \mathbf{V}^{\sigma(k,i)})$. These reduced bases are computed after the transfer of simulation data from the mesh of the medoid onto the mesh of the target problem, which is defect-dependent. The matrix of macroscopic modes related to a defect-free mechanical problem is denoted by $\mathbf{V}^{\text{macro}} \in \mathbb{R}^{\mathcal{N}^* \times 3}$. The column number j in $\mathbf{V}^{\text{macro}}$ is related to the macroscopic displacement $\mathbf{E}^{(j)}\mathbf{x}$. The complete reduced basis for displacement reads as

$$\overline{\mathbf{V}}^{(k,i)} = [\mathbf{V}^{\text{macro}}, \mathbf{V}^{(k,i)}] \in \mathbb{R}^{\mathcal{N}^{\star} \times 6}$$

The hyper-reduced equilibrium equation is (13) with empirical modes obtained by substituting $\mathbf{V}^{(k,i)}$ for \mathbf{V} in (8). A zone of interest is designed automatically around each defect. It contains four layers of elements from the border of the defect $\partial_V \Omega^*$. The construction of the RID follows the procedure explained in Appendix so that Ω_R^* contains the zone of interest.

For each image #*i* in the training dataset, a hyper-reduced model is built. It is denoted by HR(k, i) for each medoid (k = 1, ..., 5). Hyper-reduction predictions are performed for the three macroscopic strains $\mathbf{E}^{(j)}$ (j = 1, ..., 3). The predicted stresses on the RID are denoted by $\sigma^{\text{HR}(k,i,j)}$. The finite element prediction of this stress is denoted by $\sigma^{\star(i,j)}$. The exact error on the stress prediction in $\Omega_R^{\star(i)}$, $i \in \{1, ..., n_I\}$, reads as

$$e^{\mathrm{HR}(k,i)} = 100\sqrt{\frac{\sum_{j=1}^{3} \|\sigma^{\star(i,j)} - \sigma^{\mathrm{HR}(k,i,j)}\|_{\Omega_{R}}^{2}}{\sum_{j=1}^{3} \|\sigma^{\star(i,j)}\|_{\Omega_{R}}^{2}}},$$
(64)

where $\|\cdot\|_{\Omega_R} = \int_{\Omega_R} \cdot :\cdot d\Omega$ for stress tensors. Figure 7 reports the correlation between the exact error $e^{\operatorname{HR}(k,i)}$ and the chordal distance to the medoid. This plot contains $5 \times n_I$ points. The coordinates of these points are $(D_{m_k,i}^{\operatorname{Ch}}, e^{\operatorname{HR}(k,i)})$. The average chordal distance is 0.7. The average error is 29%.

The correlation between e^{HR} and d^{Ch} is not perfect, but it is sufficient here for the clustering of simulation data. In some situations, e^{HR} and d^{Ch} may not be correlated. A good correlation requires that the target mechanical problem activate all the modes in $\mathbf{V}^{\star(i)}$ and $\mathbf{V}^{(k,i)}$, which means that the related reduced coordinates have no null component. This is the case here. If one of the reduced components is null, the upper bound can be obviously simplified without using all the principal angles in $d^{\text{Ch}}(\mathbf{V}^{\star(i)}, \mathbf{V}^{(k,i)})$.



Figure 7. Correlation plot between $e^{\text{HR}(k,i)}$ and d^{Ch} for all points in the training dataset and for all hyper-reduced models. The red dot is the average point (x = 0.7, y = 29%).



Figure 8. Density distribution for $e^{\text{HR}(1,i)}$ (%) in cluster #1 for $i \in \mathcal{C}_1$.

For each cluster of data, we have reported in Figures 8–12 the histograms of the density distribution of $e^{\text{HR}(k,i)}$, for $i \in \mathcal{C}_k$, inside each cluster separately.

The clustering results are interesting because most of the errors are below 29% (the average error on the whole dataset). However, many points have an error e^{HR} bigger than 20%. The clustering may not perform well for clusters where there is a lack of similar data. Here, clusters \mathscr{C}_2 and \mathscr{C}_3 have few points with an error lower than 20%.

Some clusters could be rejected for HR in a sense that a full finite element prediction may be preferable for the defects in these clusters. In what follows, we do not reject any cluster, and we include simulation data related to all the medoids in the dictionary of digital twins.

5. Results on the test set of defects



Figure 9. Density distribution for $e^{\text{HR}(2,i)}$ (%) in cluster #2 for $i \in \mathcal{C}_2$.



Figure 10. Density distribution for $e^{\text{HR}(3,i)}$ (%) in cluster #3 for $i \in \mathcal{C}_3$.

5.1. Selection of a reduced digital twin in the dictionary

In this section, we restrict our attention to simulation data in the test set. This test set aims to evaluate the full modeling procedure for the fast prediction of stresses around defects. The average speedup for the solution of linear systems is 0.03/0.003 = 10 for hyper-reduced predictions. The average speedup for the computation of the stresses in this equation is 0.01/0.005 = 2. Speedups of approximately 1000 are obtained for similar 3D problems [20].

An error indicator has been developed for the selection in the dictionary of the medoid that is expected to give the best stress prediction via HR. This error indicator is similar to the error $e^{\text{HR}(k,i)}$, where the exact stress σ^* is replaced by the equilibrated stress σ^{eq} . The error indicator



Figure 11. Density distribution for $e^{\text{HR}(4,i)}$ (%) in cluster #4 for $i \in \mathcal{C}_4$.



Figure 12. Density distribution for $e^{\text{HR}(5,i)}$ (%) in cluster #5 for $i \in \mathcal{C}_5$.

reads as

$$\eta^{(k,i)} = 100 \sqrt{\frac{\sum_{j=1}^{3} \|\delta\sigma^{(k,i,j)}\|_{\Omega_R}^2}{\sum_{j=1}^{3} \|\sigma^{\operatorname{eq}(i,j)},\|_{\Omega_R}^2}}$$
(65)

where $\delta \sigma^{(k,i,j)}$ is the stress correction in (23), which is computed by the equilibrium step related to HR(k, i).

For each defect in the test set, we select a medoid $k^{\star(i)}$ for stress prediction such that

$$k^{\star(i)} = \operatorname{argmin}_k \eta^{(k,i)}.$$
(66)

The best medoid selection aims to lower the error in the prediction of stresses:

$$\overline{k}^{\star(i)} = \operatorname{argmin}_{k} e^{\operatorname{HR}(k,i)}.$$
(67)



Figure 13. Correlation plot between $e^{\text{HR}(k,i)}$ and $\eta^{(k,i)}$ for points in the test set. The color dots are related to the cluster index *k*.

Then, we obtain an automatic labeling of the defects in the test set. The estimated labels are denoted by \mathscr{L}_i^{η} : $\mathscr{L}_i^{\eta} = \operatorname{argmin}_k \eta^{(k,i)}$. The perfect labels are denoted by \mathscr{L}_i : $\mathscr{L}_i = \operatorname{argmin}_k e^{\operatorname{HR}(k,i)}$. Figure 13 reports the correlation between $e^{\operatorname{HR}(k,i)}$ and the error indicator $\eta^{(k,i)}$. There is a strong correlation between $e^{\operatorname{HR}(k,i)}$ and $\eta^{(k,i)}$ for low errors: $e^{\operatorname{HR}(k,i)} \approx \eta^{(k,i)}$ for

There is a strong correlation between $e^{\text{HR}(k,i)}$ and $\eta^{(k,i)}$ for low errors: $e^{\text{HR}(k,i)} \approx \eta^{(k,i)}$ for $e^{\text{HR}(k,i)} < 40\%$. This correlation exists for every cluster. As a result, this indicator helps us to find the medoid as if we have the exact finite element prediction.

The better the σ^{eq} approximates the σ^{\star} , the more accurate the error indicator. The error on equilibrated stresses reads as

$$e^{\operatorname{eq}(k,i)} = 100\sqrt{\frac{\sum_{j=1}^{3} \|\sigma^{\star(i,j)} - \sigma^{\operatorname{eq}(k,i,j)}\|_{\Omega_{R}}^{2}}{\sum_{j=1}^{3} \|\sigma^{\star(i,j)}\|_{\Omega_{R}}^{2}}}.$$
(68)

Figure 14 shows the correlation between $e^{eq(k,i)}$ and $\eta^{(k,i)}$. Such a correlation is sufficient to select the same hyper-reduced order model for the predictions with or without an equilibrium step. The range of errors on equilibrated stresses is much smaller than that on σ^{HR} . Therefore, the equilibrated stresses are the simulation outputs of interest for the reduced digital twin.

In Figures 15–19, for each cluster of data, we have reported the histograms of the density distribution of $e^{eq(k,i)}$, for $\mathcal{L}_i^{\eta} = k$, inside each cluster separately.

Most of the errors on equilibrated stresses are lower than 5%. The modeling procedure is very accurate. Errors related to medoids m_1 , m_4 , and m_5 are more concentrated below 5% than those for medoids m_2 and m_3 . Hyper-reduced predictions attached to medoids m_2 and m_3 are less accurate. Such results have been anticipated during the clustering procedure.

5.2. Detailed numerical results on hyper-reduced predictions

In this section, we report local numerical results for defects in the test set. The index of the first image is i = 1664. Its label via the error indicator is $\mathscr{L}_i^{\eta} = 3$. This label is the optimal: $\mathscr{L}_i^{\eta} = \mathscr{L}_i$. It is intentionally related to a medoid of the small cluster \mathscr{C}_3 for which there is certainly a lack of observed defects. Related numerical results are shown in Figure 20. The prediction is globally accurate ($e^{\text{eq}(3,1664)} = 5\%$).



Figure 14. Correlation plot between $e^{eq(k,i)}$ and $\eta^{(k,i)}$ for points in the test set. The color dots are related to the cluster index *k*.



Figure 15. Density distribution for $e^{eq(1,i)}$ (%) in cluster #1 for $\mathscr{L}_i^{\eta} = 1$.

Better results are obtained for the test data attached to m_4 , the medoid of the largest cluster. The index of the second image is i = 1987. The global error is $e^{eq(4,1987)} = 0.2\%$. Local predictions of the shear stress for $\mathbf{E}^{(3)}$ are reported in Figure 21.

Local predictions are very accurate for this defect certainly because cluster C_4 involves a large number of observed defects.

6. Conclusion

In this paper, local defects are compared according to a mechanical metric using model order reduction techniques and a Grassmann manifold. Each simulation data related to a defect in a training set is encoded as a point on a Grassmann manifold by using an ideal reduced basis of



Figure 16. Density distribution for $e^{eq(2,i)}$ (%) in cluster #2 for $\mathscr{L}_i^{\eta} = 2$.



Figure 17. Density distribution for $e^{eq(3,i)}$ (%) in cluster #3 for $\mathscr{L}_i^{\eta} = 3$.

displacement fluctuations around each defect. This approach can be easily extended to many types of local defects in homogeneous or locally homogeneous materials.

Five categories of defects are proposed by using a k-medoids algorithm via the Grassmann distance between the reduced bases attached to each defect. Each category of defect (also called a "cluster") has a representative defect in the training set, which is a medoid. It turns out that most defects in the proposed training set are almost spherical. However, some have a very complex shape, including several voids in interactions. The hyper-reduced predictions of the stresses around the defects in a given cluster are compared to the true simulation data. This pertains to the training data only as a validation of the modeling process. Most of the errors are below 29%. More training data would have probably improved the stress prediction by increasing the number of defect categories.



Figure 18. Density distribution for $e^{eq(4,i)}$ (%) in cluster #4 for $\mathscr{L}_i^{\eta} = 4$.



Figure 19. Density distribution for $e^{eq(5,i)}$ (%) in cluster #5 for $\mathscr{L}_i^{\eta} = 5$.



Figure 20. Test defect #1664. From left to right: finite element prediction of the shear stress related to $\mathbf{E}^{(3)}$, the related equilibrated stress σ^{eq} on the RID around the defect, and the error map for this stress component.



Figure 21. Test defect #1987. From left to right: finite element prediction of the shear stress related to $\mathbf{E}^{(3)}$, the related equilibrated stress σ^{eq} on the RID around the defect, and the error map for this stress component.

The distances to all the representative defects, for data in the test set or for newly observed defects, are evaluated by a fast error estimator on the hyper-reduced prediction of stress. The fact that the closest representative defect is a defect in the training set facilitates the interpretation of its selection.

The error estimation is restricted to an RID around the input defect. The equilibrated stresses involved in this error estimation are very accurate, with global errors on stress below 5% for data in the test set, when using the best representative defect. Such excellent results can be explained by the following facts: (i) the Grassmann metric is theoretically founded for hyper-reduced predictions as shown in this paper; (ii) both the simulation data of selected representative defects and the hyper-reduced equations enable the prediction of accurate Dirichlet boundary conditions on the boundary of the RID for the computation of equilibrated stresses. The larger the extent of this reduced domain, the more accurate the boundary conditions because they tend toward a given macroscopic value. However, the larger the RID, the higher the computational complexity of the error estimator. This complexity limits the number of defect categories when selecting the best representative defect for stress predictions.

To develop this promising approach to defect classification, more data are needed. Both data acquisition and data augmentation techniques are in development. In the future, more defect categories must be defined. In such a case, the selection of the best representative defect could be carried out by a deep classifier as in ROM-nets.

Appendix. Details about RID construction

Let us introduce two mathematical operators. The first mathematical operator collects the degrees of freedom of a subdomain Ω_{α} :

$$\mathscr{C}(\Omega_{\alpha}) = \left\{ i \in \{1, \dots, \mathcal{N}\}, \int_{\Omega_{\alpha}} \boldsymbol{\varphi}_{i}^{2} \, \mathrm{d}\Omega > 0 \right\}$$

The second mathematical operator aggregates the support of finite element shape functions having their index in a set \mathcal{G} :

$$\mathscr{L}(\mathscr{G}) = \bigcup_{i \in \mathscr{G}} \operatorname{supp}(\boldsymbol{\varphi}_i), \quad \mathscr{L}(\mathscr{G}) \subset \Omega.$$

The extension of this subdomain by adding *n* layers of connected elements reads as

$$(\mathcal{L} \circ \mathcal{C})^n \circ \mathcal{L}(\mathcal{G})$$

The operator \mathscr{L} is suitable for displacement fields because they are approximated by finite element shape functions. A similar operator is also introduced for stresses. When collecting simulation data related to stresses, in the matrix \mathbf{Q}^{σ} , we store all the stress components at all

Gauss points for all elements. Each row of \mathbf{Q}^{σ} is related to one component of the stress tensor at a Gauss point in an element. Then, the DEIM algorithm applied to \mathbf{V}^{σ} gives a set of indices of components of the stress tensor at some Gauss points in some elements. This set is denoted by \mathscr{P}^{σ} . We denote by $\mathscr{L}^{\sigma}(\mathscr{P}^{\sigma})$ the support of the elements related to set \mathscr{P}^{σ} . Here, $\mathscr{L}^{\sigma}(\mathscr{P}^{\sigma})$ is a subdomain of Ω .

In this paper, the RID construction is the following:

$$\Omega_R = (\mathscr{L} \circ \mathscr{C}) \circ (\mathscr{L}(\mathscr{P}) \cup \mathscr{L}^{\sigma}(\mathscr{P}^{\sigma})) \cup \Omega_{\text{ZOI}},$$

where Ω_{ZOI} is the zone of interest.

References

- Z. Li, X. Yan, C. Yuan, Z. Peng, L. Li., "Virtual prototype and experimental research on gear multi-fault diagnosis using wavelet-autoregressive model and principal component analysis method", *Mech. Syst. Signal Process.* 25 (2011), p. 2589-2607.
- [2] T. Mira-Aguiar, C. Leitão, D. M. Rodrigues, "Solid-state resistance seam welding of galvanized steel", Int. J. Adv. Manufact. Technol. 86 (2016), no. 5, p. 1385-1391.
- [3] T. Uwaba, Y. Yano, M. Ito, "Resistance spot weldability of 11cr-ferritic/martensitic steel sheets", J. Nucl. Mater. 421 (2012), no. 1, p. 132-139.
- [4] S. K. Dinda, J. M. Warnett, M. A. Williams, G. G. Roy, P. Srirangam, "3D imaging and quantification of porosity in electron beam welded dissimilar steel to Fe–Al alloy joints by X-ray tomography", *Mater. Des.* 96 (2016), p. 224-231.
- [5] J. D. Madison, L. K. Aagesen, "Quantitative characterization of porosity in laser welds of stainless steel", *Scr. Mater.* 67 (2012), no. 9, p. 783-786.
- [6] A. Haboudou, P. Peyre, A. B. Vannes, G. Peix, "Reduction of porosity content generated during Nd:YAG laser welding of A356 and AA5083 aluminium alloys", *Mater. Sci. Eng. A* 363 (2003), no. 1, p. 40-52.
- [7] D. Amsallem, C. Farhat, "An online method for interpolating linear parametric reduced-order models", SIAM J. Sci. Comput. 33 (2011), p. 2169-2198.
- [8] S. Moomkesh, S. A. Mireei, M. Sadeghi, M. Nazeri, "Early detection of freezing damage in sweet lemons using Vis/SWNIR spectroscopy", *Biosys. Eng.* 164 (2017), p. 157-170.
- [9] Y. Le Cun, B. E. Boser, J. S. Denker, D. Henderson, R. E. Howard, W. E. Hubbard, L. D. Jackel, "Handwritten digit recognition with a back-propagation network", in *Advances in Neural Information Processing Systems 2* (D. S. Touretzky, ed.), Morgan-Kaufmann, 1990, http://papers.nips.cc/paper/293-handwritten-digit-recognition-with-aback-propagation-network.pdf, p. 396-404.
- [10] A. Krizhevsky, I. Sutskever, G. E. Hinton, "Imagenet classification with deep convolutional neural networks", Adv. Neural Inf. Process. Syst. 2 (2012), p. 1097-1105, cited By 33336.
- [11] T. Zehelein, T. Hemmert-Pottmann, M. Lienkamp, "Diagnosing automotive damper defects using convolutional neural networks and electronic stability control sensor signals", *Sensor Actuator Networks* **9** (2020), p. 1-19.
- [12] L. Xiao, B. Wu, Y. Hu, J. Liu, "A hierarchical features-based model for freight train defect inspection", *IEEE Sens. J.* 20 (2020), p. 2671-2678.
- [13] S. J. Pan, Q. Yang, "A survey on transfer learning", IEEE Trans. Knowl. Data Eng. 22 (2010), no. 10, p. 1345-1359.
- [14] D. Ulrich, B. van Rietbergen, H. Weinans, P. Ruegsegger, "Finite element analysis of trabecular bone structure: a comparison of image-based meshing techniques", *J. Biomech.* **31** (1998), no. 12, p. 1187-1192.
- [15] F. N'Guyen, "Morphologie mathématique appliquée au développement d'outils de maillage EF automatiques dans le cas de microstructures hétérogènes bi et multiphasées", PhD Thesis, Lille 1 University, 2014.
- [16] P. Henry, G. Nicolas, F. Samuel, L. Wolfgang, "Incipient bulk polycrystal plasticity observed by synchrotron in-situ topotomography", *Materials* 11 (2018), p. 1-18.
- [17] Y. Amani, S. Dancette, P. Delroisse, A. Simar, E. Maire, "Compression behavior of lattice structures produced by selective laser melting: X-ray tomography based experimental and finite element approaches", *Acta Mater.* 159 (2018), p. 395-407.
- [18] A. Madra, P. Breitkopf, A. Rassineux, F. Trochu, "Image-based model reconstruction and meshing of woven reinforcements in composites", *Int. J. Numer. Methods Eng.* **112** (2017), no. 9, p. 1235-1252.
- [19] Y. Huang, Z. Yang, W. Ren, G. Liu, C. Zhang, "3d meso-scale fracture modelling and validation of concrete based on in-situ X-ray computed tomography images using damage plasticity model", *Int. J. Solids Struct.* 67–68 (2015), p. 340-352.
- [20] L. Lacourt, D. Ryckelynck, S. Forest, V. de Rancourt, S. Flouriot, "Hyper-reduced direct numerical simulation of voids in welded joints via image-based modeling", *Int. J. Numer. Methods Eng.* **121** (2020), no. 11, p. 2581-2599.

- [21] D. Ryckelynck, "Hyper-reduction of mechanical models involving internal variables", Int. J. Numer. Methods Eng. 77 (2009), no. 1, p. 75-89.
- [22] T. Daniel, F. Casenave, N. Akkari, D. Ryckelynck, "Model order reduction assisted by deep neural networks (ROMnet)", *Adv. Model. Simul. Eng. Sci.* **7** (2020), no. 1, p. 16.
- [23] J. Lumley, "The structure of inhomogeneous turbulence", in *Atmospheric Turbulence and Wave Propagation*, Nauka, Moscow, 1967, p. 166-178.
- [24] M. Raissi, P. Perdikaris, G. E. Karniadakis, "Physics-informed neural networks: a deep learning framework for solving forward and inverse problems involving nonlinear partial differential equations", J. Comput. Phys. 378 (2019), p. 686-707.
- [25] F. Nguyen, S. M. Barhli, D. P. Muñoz, D. Ryckelynck, "Computer vision with error estimation for reduced order modeling of macroscopic mechanical tests", *Complexity* 2018 (2018), p. 1-10.
- [26] L. Lacourt, "Lifetime assessment of welded structures containing defects", PhD Thesis, Mines ParisTech-PSL University, 2019.
- [27] H. S. Park, C. H. Jun, "A simple and fast algorithm for k-medoids clustering", *Expert Syst. Appl.* 36 (2009), p. 3336-3341.
- [28] D. Ryckelynck, L. Gallimard, S. Jules, "Estimation of the validity domain of hyper-reduction approximations in generalized standard elastoviscoplasticity", *Adv. Model. Simul. Eng. Sci.* **2** (2015), p. 6.
- [29] D. Amsallem, C. Farhat, "Interpolation method for adapting reduced-order models and application to aeroelasticity", *AIAA J.* **46** (2008), p. 1803-1813.
- [30] R. Mosquera, A. Hamdouni, A. El Hamidi, C. Allery, "Pod basis interpolation via inverse distance weighting on grassmann manifolds", *Discrete Contin. Dyn. Syst., Series S* 12 (2018), p. 1743-1759.
- [31] K. Ye, L. H. Lim, "Schubert varieties and distances between subspaces of different dimensions", SIAM J. Matrix Anal. Appl. 37 (2016), no. 3, p. 1176-1197.
- [32] B. Miled, D. Ryckelynck, S. Cantournet, "A priori hyper-reduction method for coupled viscoelastic-viscoplastic composites", *Comput. Struct.* 119 (2013), p. 95-103, cited By 8.
- [33] M. Horák, D. Ryckelynck, S. Forest, "Hyper-reduction of generalized continua", *Comput. Mech.* 59 (2017), no. 5, p. 753-778.
- [34] D. Ryckelynck, K. Lampoh, S. Quilici, "Hyper-reduced predictions for lifetime assessment of elasto-plastic structures", *Meccanica* **51** (2016), p. 309-317.
- [35] M. Barrault, Y. Maday, N. C. Nguyen, A. T. Patera, "An empirical interpolation method: application to efficient reduced-basis discretization of partial differential equations", C. R. Math. 339 (2004), no. 9, p. 667-672.
- [36] J. Fauque, I. Ramiere, D. Ryckelynck, "Hybrid hyper-reduced modeling for contact mechanics problems", Int. J. Numer. Methods Eng. 115 (2018), no. 1, p. 117-139.
- [37] J. Baiges, R. Codina, S. Idelson, "A domain decomposition strategy for reduced order models. Application to the incompressible Navier-Stokes equations", *Comput. Methods in Appl. Mech. Eng.* 267 (2013), p. 23-42.
- [38] P. Kerfriden, J. C. Passieux, S. Bordas, "Local/global model order reduction strategy for the simulation of quasi-brittle fracture", Int. J. Numer. Methods Eng. 89 (2012), no. 2, p. 154-179.
- [39] P. Kerfriden, J. J. Ródenas, S. P.-A. Bordas, "Certification of projection-based reduced order modelling in computational homogenisation by the constitutive relation error", *Int. J. for Numer. Methods Eng.* 97 (2014), no. 6, p. 395-422.
- [40] K. C. Hoang, P. Kerfriden, S. P. A. Bordas, "A fast, certified and "tuning free" two-field reduced basis method for the metamodelling of affinely-parametrised elasticity problems", *Comput. Methods Appl. Mech. Eng.* 298 (2016), p. 121-158.
- [41] S. Sahyoun, S. M. Djouadi, "Control of nonlinear pdes based on space vectors clustering reduced order systems", IFAC Proc. Volumes 47 (2014), no. 3, p. 5181-5186, 19th IFAC World Congress.
- [42] M. Ghasemi, E. Gildin, "Localized model reduction in porous media flow", *IFAC-PapersOnLine* 48 (2015), no. 6, p. 242-247, 2nd IFAC Workshop on Automatic Control in Offshore Oil and Gas Production OOGP 2015.
- [43] M. Hess, A. Alla, A. Quaini, G. Rozza, M. Gunzburger, "A localized reduced-order modeling approach for pdes with bifurcating solutions", *Comput. Methods Appl. Mech. Eng.* 351 (2019), p. 379-403.



Contributions in mechanics of materials

Numerical experiments on unsupervised manifold learning applied to mechanical modeling of materials and structures

Ruben Ibanez^{*a*}, Pierre Gilormini^{*a*}, Elias Cueto^{*b*} and Francisco Chinesta^{*, *a*}

 $^{\it a}$ PIMM lab, Arts et Metiers Institute of Technology, 151 Boulevard de Hôpital, 75013 Paris, France

^b Aragon Institute of Engineering Research, Universidad de Zaragoza, Maria de Luna s/n, 50018 Zaragoza, Spain

E-mails: ruben.ibanez-pinillo@ensam.eu (R. Ibanez), pierre.gilormini@ensam.eu (P. Gilormini), ecueto@unizar.es (E. Cueto), francisco.chinesta@ensam.eu (F. Chinesta)

Abstract. The present work aims at analyzing issues related to the data manifold dimensionality. The interest of the study is twofold: (i) first, when too many measurable variables are considered, manifold learning is expected to extract useless variables; (ii) second, and more important, the same technique, manifold learning, could be utilized for identifying the necessity of employing latent extra variables able to recover single-valued outputs. Both aspects are discussed in the modeling of materials and structural systems by using unsupervised manifold learning strategies.

Keywords. Nonsupervised manifold learning, State variables, Dimensionality reduction, *k*-PCA, Structural analysis, Material constitutive equations.

Manuscript received 21st June 2020, revised 12th July 2020, accepted 7th October 2020.

1. Introduction

Recently, data-driven description of materials has been gaining popularity. Many complex material behaviors resisting traditional modeling procedures, or that are too complex from a microstructural viewpoint, are approached by using data-based descriptions. Different approaches are being considered. Among them include those based exclusively on measured data, others that extract the manifolds related to data, and others that attempt to enforce thermodynamic and thermomechanical consistency. The interested reader can refer to [1–7] and the numerous references therein. This work focuses on techniques based on the use of manifolds and their associated manifold learning procedures for extracting them from the available data.

^{*} Corresponding author.



Figure 1. Multidimensional data on one- (left), two- (center), and three-dimensional (right) manifolds embedded in \mathbb{R}^D .

In general, data involve many dimensions. Consider first a sequence of three-dimensional (3D) fields defined in a domain $\Omega \subset \mathbb{R}^3$ partitioned into D voxels. Each of these fields contains many data, one datum at each voxel. Each field can be represented as a point in a vector space of dimension D (the number of voxels), where we can presume each of the D coordinate axes as reporting the value that the field of interest takes in the associated voxel. Thus, each field becomes a point in that high-dimensional space of dimension D, \mathbb{R}^D . If important correlations exist among the different fields, these points are expected to be distributed on a low-dimensional subspace embedded in the D-dimensional space. Techniques aiming at extracting these reduced subspaces, the so-called slow manifolds, sketched in Figure 1, are key tools for manipulating data and extracting their hidden information.

Thus, data define in general slow manifolds embedded in very large vector spaces due to the significant hidden correlations among them. The number of uncorrelated explicative dimensions usually becomes much smaller than the a priori assumed dimension of the space for accommodating the data. The extraction of these slow manifolds can be successfully accomplished by using linear and nonlinear dimensionality reduction techniques such as principal component analysis (PCA) in the linear case and its nonlinear counterparts (ℓ PCA, kernel-based PCA [k-PCA], LLE, tSNE, etc.) [8–12].

These techniques can be applied to several physical systems. Moreover, when the slow manifold is determined, the solution at any point on it can be computed very accurately from a simple interpolation of neighboring data on the manifold [13], enabling almost real-time predictions and the associated real-time decision-making.

However, extracting knowledge from data associated with an existing but hidden model requires the following:

- identifying the manifold intrinsic dimension,
- · discovering hidden parameters,
- · discarding useless parameters, and
- · discovering the models originating the data.

These questions are addressed, illustrated, and discussed in the present work in a purely methodological manner, aiming at illustrating the key concepts that could open numerous future possibilities in the field of mechanics of materials, processes, structures, and systems.

In our previous works, we addressed problems involving thousands of dimensions [13–16], proving that despite the apparent richness in many cases, the embedding regards a relatively low-dimensional space. However, in most of the problems that we have treated until now, their complexity prevented fine analyses of their solutions. The present paper, which is purely methodological, considers simple problems defined in low dimensions, with known solutions

being easily visualizable, for facilitating an analysis and discussion. Of course, and as proved in the works just referred to, all the methodologies apply to multidimensional settings.

2. Unsupervised manifold learning

Let us consider a vector $\mathbf{y} \in \mathbb{R}^D$ containing experimental or synthetic data from measurements or numerical simulation. These results are often referred to as *snapshots*. If they are obtained by numerical simulation, they consist of nodal values of the essential variable. Therefore, these variables will be somehow correlated and, notably, there will be a linear transformation **W** defining the vector $\boldsymbol{\xi} \in \mathbb{R}^d$, with d < D, which contains the still unknown *latent variables* such that

$$\mathbf{y} = \mathbf{W}\boldsymbol{\xi}.\tag{1}$$

The $D \times d$ transformation matrix **W**, which satisfies the orthogonality condition $\mathbf{W}^T \mathbf{W} = \mathbf{I}_d$, is the main ingredient of the PCA and can be computed as detailed in Appendix A from the covariance matrix associated with a number (M) of snapshots $\mathbf{y}_1, \dots, \mathbf{y}_M$, which constitute the columns of matrix **Y**.

While PCA works with the covariance matrix (i.e., $\mathbf{Y}\mathbf{Y}^T$), multidimensional scaling (MDS) works with the Gram matrix containing scalar products (i.e., $\mathbf{S} = \mathbf{Y}^T \mathbf{Y}$) as described in Appendix A.

On the other hand, the *k*-PCA is based on the fact that data not linearly separable in *D* dimensions could be linearly separated if previously projected to a space in Q > D dimensions. However, the true advantage arises from the fact that it is not necessary to write down the analytical expression of that mapping as described in Appendix A.

3. An illustrative structural mechanics case study

Consider first a hypothetical mechanical system consisting of a prismatic beam whose three dimensions, height, width, and length, are denoted, respectively, by *h*, *b*, and *L*, all of them being measurable quantities. In what follows, we consider a particular output *P* that constitutes also a measurable quantity (buckling critical load, etc.) assumed related to those parameters from an existing but actually hidden model even if in what follows we will consider hypothetical, and most of the time, unphysical models.

Thus, we consider a set of data composed of M measures $\mathbf{y}_i = \{h_i, b_i, L_i, P_i\}$, i = 1, ..., M, with h_i , b_i , and L_i being randomly chosen from a uniform probability distribution in their respective intervals of existence, \mathcal{I}_h , \mathcal{I}_b , and \mathcal{I}_L , respectively, defined from the following:

$$\begin{cases} \mathscr{I}_{h} = [h_{\min}, h_{\max}] \\ \mathscr{I}_{b} = [b_{\min}, b_{\max}] \\ \mathscr{I}_{L} = [L_{\min}, L_{\max}]. \end{cases}$$
(2)

Without any other pre-existing knowledge, one expects the output depending on the three geometrical parameters (i.e., P = P(h, b, L)). In what follows, we consider three different scenarios.

3.1. Output depending on a single parameter

In this section, we assume a quite simple model that relates the output to a single parameter,

$$P = \alpha b, \quad \alpha \in \mathbb{R}^+, \tag{3}$$

where $\alpha = 1000$ in the numerical tests carried out. We perform M = 1000 measures, where $\mathscr{I}_h \in [0.1, 0.2], \mathscr{I}_b \in [0.15, 0.2]$, and $\mathscr{I}_L \in [1, 1.5]$.



Figure 2. Eigenvalues $\lambda \in [\lambda_1 10^{-6}, \lambda_1]$.

The measures constitute a set of M = 1000 points in \mathbb{R}^4 on which the *k*-PCA is applied by using the Gaussian kernel

$$\kappa(\mathbf{y}_i, \mathbf{y}_j) = \exp^{-\frac{\|\mathbf{y}_i - \mathbf{y}_j\|^2}{2\beta^2}},\tag{4}$$

where $\beta = 10$.

Figure 2 depicts the highest eigenvalues among M resulting from the *k*-PCA, those lying between the highest value λ_1 and $10^{-6}\lambda_1$.

The slow manifold associated with $\boldsymbol{\xi}$ is represented by selecting the first three reduced coordinates (ξ_1, ξ_2, ξ_3) as shown in Figure 3, where its one-dimensional (1D) intrinsic dimension is noted. This result was expected from the considered model expressed by (3). The points on the slow manifold are colored depending on the values of *h*, *b*, *L*, and *P*, evidencing that *b* constitutes the latent variable and that the output *P* scales (visually) linearly with it.

The process of coloring the data points in the embedded manifold deserves some additional comments due to the fact that this is used in all the analyses reported in the present paper. Manifold learning techniques look for a low-dimensional manifold defined by data points. As soon as the slow manifold is extracted, the different data points can be mapped on it. This visualization is only possible when the number of dimensions allows a simple graphical representation (as is the case for the problems addressed in the present paper). Then, these points can be colored depending on the value of the different initial coordinates, and one expects that if there is a correlation (direct or inverse and linear or nonlinear) between the initial and reduced coordinates, the colors must exhibit a certain grading.

Even if this analysis seems quite dependent on the low dimensionality of the embedding, in higher dimensional embeddings, the analysis can be performed by using local statistics. Thus, by considering a data point in the slow manifold and its closest neighbors, a local statistical analysis can be easily performed with the standard deviation indicating the dispersion of the data (equivalent to the local dispersion of colors).

One could be slightly surprised by the nonlinearity that the manifold exhibits despite the linearity of model (3). This nonlinearity is an artifact of the nonlinear kernel (4) used. As the model is linear, one could expect the ability of the PCA to address the problem at hand. For this purpose, it suffices transforming the kernel into its linear counterpart, giving rise to the PCA

$$\kappa(\mathbf{y}_i, \mathbf{y}_j) = \mathbf{y}_i \cdot \mathbf{y}_j. \tag{5}$$



Figure 3. Slow manifold in the 3D space defined by the first reduced coordinates (ξ_1, ξ_2, ξ_3) . Each point is colored according to the value of the coordinate *h* (top left), *b* (top right), *L* (bottom left), or *P* (bottom right).



Figure 4. Slow manifold ξ when considering the PCA linear dimensionality reduction. Each point is colored according to the value of the variable *b*.

In this case, as expected, a single nonzero eigenvalue results, and consequently the dimension of the reduced space becomes one (i.e., $\xi = \xi$). The associated manifold is depicted in Figure 4.

Now, we consider a slightly different model, again depending on a single variable but in a nonlinear manner, according to

$$P = \alpha h^2, \tag{6}$$



Figure 5. Slow manifold in the 3D space defined by the first three reduced coordinates (ξ_1, ξ_2, ξ_3) . Each point is colored according to the value of the coordinate *h* (left) or *P* (right).



Figure 6. Slow manifold in the 3D space defined by the first three reduced coordinates (ξ_1, ξ_2, ξ_3) . Each point is colored according to the value of the coordinate *L* (left) or *P* (right).

where again $\alpha = 1000$. Figure 5 depicts the 1D slow manifold, where points are colored according to the values of the variables *h* and *P*. Here, even if the direct relation can be noted, its nonlinearity is much less evident to visualize.

Finally, we consider the model

$$P = \frac{\alpha}{L^2},\tag{7}$$

where $\alpha = 1000$. Figure 6 depicts the 1D slow manifold, where points are colored according to the values of the variables *L* and *P* to emphasize the inverse relation between them.

3.2. Output depending on two parameters

In this section, we consider a model involving two of the three variables, in particular,

$$P = \alpha \frac{h^3}{L^2},\tag{8}$$

where $\alpha = 1000$.

Figure 7 depicts the two-dimensional (2D) slow manifold, where points are colored according to the values of variables *h*, *b*, *L*, and *P*. Here, the direct and inverse effects of *h* and *L* with respect to *P* can be noted as well as the fact that parameter *b* seems, and in fact is, useless.



Figure 7. Slow manifold in the 3D space defined by the first three reduced coordinates (ξ_1, ξ_2, ξ_3) . Each point is colored according to the value of the coordinate *h* (top left), *b* (top right), *L* (bottom left), or *P* (bottom right).

3.3. Identifying hidden variables

The previous case studies revealed the ability to extract the intrinsic dimensionality of the slow manifold as well as the possibility to identify useless parameters. The present case addresses a very different situation in which the model involves the three variables in the discussed case, h, b, and L. However, only two of them were measured, namely h and L with the output P, with b remaining inaccessible.

Thus, we have

$$P = \alpha \frac{bh^3}{L^2},\tag{9}$$

where $\alpha = 1000$. Therefore, the M = 1000 collected data \mathbf{y}_i , $i = 1, \dots, M$, reads as $\mathbf{y}_i = \{h_i, L_i, P_i\} \in \mathbb{R}^3$.

Figure 8 depicts the reduced points (ξ_1, ξ_2, ξ_3) , which as can be seen are distributed in a domain $\omega \subset \mathbb{R}^3$. However, no dimensionality reduction is noted, and the embedding remains 3D. A direct consequence is that many values of the output *P* exist for the same values on the measured inputs *h* and *L*, related to the different values of *b*, which affect the output *P*. However, as *b* is not measured, its value is not considered in the data points.

Such a multivalued output does not represent any conceptual difficulty. It indicates that even if both variables participate in the output, there may be others that were not considered or those considered may be useless for explaining the output.



Figure 8. Reduced representation $\xi_i \in \mathbb{R}^3$ of the data $\mathbf{y}_i \in \mathbb{R}^3$, where the reduced points are colored according to the value of the output *P*.



Figure 9. Reduced points colored according to the values of the coordinates *h* (left) and *L* (right).

To conclude about the pertinence of these variables with respect to the considered output, we consider coloring the reduced data depending on the h and L values (refer to Figure 9). We compare them with the one where the color scales with the output P reported in Figure 8.

Thus, one could conclude that both variables h and L are relevant for explaining the output P. If we assume that the output P should be univocally explained from a small number of variables, clearly only two variables (here h and L) are not sufficient. One extra dimension suffices for recovering a single-valued output, that is, considering the reduced points in four dimensions \mathbb{R}^4 .

As visualizing things in four dimensions is quite a difficult task, for the sake of clarity in the exposition, in what follows, we propose addressing a simpler model involving lower dimensional spaces.

We consider the simpler model

$$P = \alpha b h^3, \tag{10}$$

where $\alpha = 1000$. However, the M = 1000 collected data \mathbf{y}_i , i = 1, ..., M, only deal with h and P, (i.e., $\mathbf{y}_i = \{h_i, P_i\} \in \mathbb{R}^2$).

Figure 10 depicts the dataset $\mathbf{y}_i = \{h_i, P_i\}$, where it can be noted that many values of the output *P* are found for the same value of the variable *h*.

For univocally expressing the output *P*, we consider again the *k*-PCA. We compute the reduced dataset in a 3D space by considering the first three coordinates (ξ_1 , ξ_2 , ξ_3), while coloring these



Figure 10. Dataset $y_i = \{h_i, P_i\}, i = 1, ..., M$.



Figure 11. Reduced points colored according to the values of the coordinates *h* (left) and *P* (right).

points with respect to h, to prove that h represents an explanatory variable, or with respect to P (refer to Figure 11). Figure 12 depicts the same manifold but now colored by using the hidden variable b, which proves that it contributes to explaining the output P and constitutes the hidden variable. Even if we just proved that a latent variable exists, and that it corresponds to b, as in practice we ignore this fact, we never measure the quantity b. Furthermore, it is even possible that we ignore its existence; the proposed procedure is only informative but not constructive.

Obviously, there is no unique choice. The same behavior is obtained by coloring the reduced dataset with respect to any function $b^p h^q$, where $p, q \in \mathbb{R}$. Thus, any measurable variable ensuring such a uniform color grading could be used as the latent variable for constructing the model. There are an infinite number of possibilities but with certain constraints; in the present case, it must involve *b*.

To conclude this section, we address a similar but more complex situation. We consider now the richer model (9) with $\alpha = 1000$. The M = 1000 collected data \mathbf{y}_i , i = 1, ..., M, only deal with the input variable h and the output P (i.e., $\mathbf{y}_i = \{h_i, P_i\} \in \mathbb{R}^2$), where two variables, b and L, involved in model (9) remain hidden.

Figure 13 depicts the dataset $\mathbf{y}_i = \{h_i, P_i\}$, where again it can be noted that many values of the output *P* are found for the same value of the variable *h*.

For univocally expressing the output *P*, we consider again the *k*-PCA. We compute the reduced dataset in a 3D space by considering again the first three coordinates (ξ_1 , ξ_2 , ξ_3) while coloring



Figure 12. Reduced points colored according to the value of the output *b*.



Figure 13. Dataset $\mathbf{y}_i = \{h_i, P_i\}$.

these points with respect to *h* to prove that *h* represents an explanatory variable of the output *P* as Figure 14 proves. Figure 15 clearly reveals that by coloring the reduced points with respect to the two variables *b* and *L* taken solely, they do not represent the unique hidden variable able to explain using *h* the output *P*. The hidden latent variable according to model (9) should combine *b* and *h*. Figure 16 proves that the combined parameter b/L^2 perfectly works when compared with the manifold colored with respect to the output *P*.

It is important to note that the latent variable must involve the term b/L^2 up to any power and eventually a multiple of any power of *h*.

3.4. Discussion

The previous numerical experiments allow drawing a conclusion on the ability of unsupervised manifold learning techniques for identifying useless data and the existence of latent variables.

The procedure is based on the ability of embedding high-dimensional data in a lowdimensional space, where the dimension represents an approximation of the inherent dimensionality of the data.



Figure 14. Reduced points colored according to the value of the coordinate *h*.



Figure 15. Reduced points colored according to the values of the variables *b* (left) and *L* (right).



Figure 16. Reduced points colored according to the values of the variables b/L^2 (left) and *P* (right).

When the expected dimensionality reduction does not occur and the k-PCA reveals univocally that an extra dimension is required to accommodate the data, this indicates the existence of a hidden latent variable.

However, when the dimensionality reduction applies, by analyzing locally the manifold (any



Figure 17. Elastic behavior (left), elastic–perfectly plastic behavior (center), and elastoplasticity with linear hardening (right).

embedded data point with its closest neighbors) with respect to every original coordinate attached to each embedded data point, one can conclude that correlations exist between the initial and reduced coordinates (color grading or low statistical variance). When the dispersion increases to a large extent, one can infer that the initial coordinate analyzed is not directly correlated (even if it could be correlated in a more complex or combined manner) as discussed.

To prove the generality of these conclusions, Section 4 addresses a more complex scenario. It deals with the constitutive modeling of materials, particularly those whose behavior depends on the deformation history of the material.

4. Simple mechanical behaviors

In this section, we consider some simple 1D mechanical behaviors as that depicted in Figure 17. Figure 18 shows a set of M = 1500 strain–stress pairs $\mathbf{y}_i = \{\varepsilon_i, \sigma_i\}, i = 1, ..., M$, related to an hypothetical 1D elastic–plastic behavior with linear hardening. The depicted points represent the final mechanical state (strain–stress) of M loading–unloading random trajectories. It can be noted that with only the information of possible mechanical states, given a strain value, there are many probable values of permissible stresses and vice versa.

There is nothing physically inconsistent in having multivalued fields, but if one is looking for single-valued stress, then an extra latent variable must be introduced.

If, as in Section 3, we apply the *k*-PCA on the dataset \mathbf{y}_i , i = 1,...,M, as expected, the 2D manifold appears embedded in the \mathbb{R}^3 space as depicted in Figure 19. This indicates that three mechanical variables, stress and strain completed by a latent extra variable *p*, are mutually related (e.g., $\sigma = \sigma(\varepsilon, p)$), giving rise to a 2D manifold embedded in \mathbb{R}^3 .

As previously discussed, many latent variables could be certainly considered for obtaining for example a single-valued stress value. Historically, the plastic strain ε_p , illustrated in Figure 20, was widely considered as a latent variable. Coloring the reduced points ξ_i depicted in Figure 19 according to the plastic strain results in the colored manifold shown in Figure 21. This validates the choice of the plastic strain as a latent variable.

5. Toward alternative representations of the mechanical state

An alternative representation of the mechanical state, avoiding the choice of nonevident latent variables, consists in assuming that the stress at time *t* depends on the whole strain history, that is, on $\varepsilon(\tau)$, $\tau \le t$.

When memory effects are neglected, the mechanical state can be described at time *t* from the strain and stress increments and the present state of stress–strain. That is, $\forall t_i = i\Delta t$, $\mathbf{y}_i = \{\varepsilon_i, \sigma_i, \Delta\sigma_i/\Delta\varepsilon_i\}$, where $\Delta\varepsilon_i$ has a given magnitude and two possible signs (positive in loading and negative in unloading).



Figure 18. Strain–stress mechanical states related to elastic–plastic behavior with linear hardening.



Figure 19. Elastic-plastic manifold embedded in the 3D space.

Different scenarios can be analyzed:

- Linear elastic regime without plastic strain. In this case, as expected, the slow manifold constituted by the reduced states *ξ_i* related to the mechanical states *y_i*, depicted in Figure 22, in the 3D space (*ξ*₁, *ξ*₂, *ξ*₃) becomes 1D.
- Nonlinear elastic behavior (with the tangent modulus $E_T = E\varepsilon^2$) without plastic strain. In this case, the results are similar to those just discussed. In Figure 23, the nonlinear behavior can be noted.



Figure 20. Plastic strain definition (left) and mechanical states \mathbf{y}_i colored according to the plastic strain (right).



Figure 21. Manifold colored according to the plastic strain.

- Linear elastic regime with nonzero plastic strain (perfect plasticity—no hardening). In the present case, in the mechanical state within the elastic domain, we recover from \mathbf{y}_i a reduced slow manifold of dimension two as expected, which is depicted in Figure 24.
- Linear elastic regime with nonzero plastic strain (perfect plasticity—no hardening) with activated damage. In the present case, with respect to the previously discussed scenario, we presume that the material degrades with the magnitude of the plastic strain and the tangent modulus decreases accordingly. As Figure 25 depicts, this case is quite similar to the previous scenario. However, when coloring with respect to the tangent modulus, the expected uniform degraded map is noted.
- The present case study considers, in addition to the points inside the elastic domain, another set of mechanical states on the elastic domain boundary with a different



Figure 22. Slow manifold related to linear elastic behavior and colored according to the tangent modulus (i.e., $\Delta \sigma_i / \Delta \varepsilon_i$).



Figure 23. Slow manifold related to nonlinear elastic behavior and colored according to the tangent modulus (i.e., $\Delta \sigma_i / \Delta \varepsilon_i$).

instantaneous tangent modulus when loading and unloading. As Figure 26 reveals, the mechanical manifold is now richer with points on the elastic domain boundary separated from the elastic manifold. In the solution depicted in Figure 26, damage is not activated. In the presence of damage, the reduction in elastic tangent modulus scales with the plastic strain. The slow manifold is colored according to the tangent modulus as shown in Figure 27.

• The last scenario adds an extra richness to the constitutive behavior. At the mechanical states within the elastic domain, the elastic tangent modulus is affected by a latent



Figure 24. Strain–stress points associated with states within the elastic domain with nonzero plastic strains (left) and the associated slow manifold colored according to the tangent modulus (right).



Figure 25. Slow manifold related to damageable elastic–plastic behavior operating within the elastic domain colored according to the tangent modulus.

variable that is the product of the plastic strain with another extra variable. The latter, which could represent strain-rate sensitivity, is assumed to take arbitrary values here due to the fact that we are more interested in methodological aspects than in physical considerations. The considered mechanical states are depicted in Figure 28. When applying the *k*-PCA to the set of mechanical states $\mathbf{y}_i = \{\varepsilon_i, \sigma_i, \Delta\sigma_i/\Delta\varepsilon_i\}$, the resulting manifold remains 3D. As expected, no dimensionality reduction is accomplished as Figure 29 reveals, where many values of the elastic tangent modulus can be found for the same values of the stress and strain. To investigate the nature of this behavior, we depict in Figure 30 the elastic tangent modulus versus the plastic strain, where as expected it can be noted that the former does not depend exclusively on the latter. By applying the *k*-PCA to the data shown in Figure 30, which are nonseparable in two dimensions, one expects



Figure 26. Slow manifold related to elastic–plastic behavior operating within the elastic domain and on the elastic domain boundary colored according to the tangent modulus.



Figure 27. Slow manifold related to elastic–plastic behavior operating within the elastic domain and on the elastic domain boundary, colored according to the tangent modulus, when damage is activated.

to separate them by embedding in a 3D space as previously discussed and as Figure 31 proves. Finally, Figure 32 presents the slow manifold from Figure 31 but now colored with respect to the plastic strain or the extra latent variable.



Figure 28. Mechanical states within the elastic domain, colored according to the tangent modulus, the last depending on the product of two latent variables, the plastic strain, and another arbitrarily chosen variable.



Figure 29. Slow manifold related to elastic–plastic behavior operating within the elastic domain, colored according to the tangent modulus, with the last scaling with the product of the plastic strain and an extra latent variable.



Figure 30. Elastic tangent modulus versus the plastic strain.



Figure 31. Slow manifold related to the data consisting of the elastic tangent modulus and the plastic strain.



Figure 32. Slow manifold of the elastic tangent modulus colored according to the plastic strain (left) and the extra latent variable (right).

6. Conclusion

The present work introduced, tested, and discussed issues related to manifold dimensionality with two major purposes: (i) first, when too many measurable variables are employed, manifold learning is able to discard the useless variables; (ii) second and more important, the same technique can be employed for discovering the necessity of employing and then measuring an extra latent variable that is able to recover and ensure single-valued outputs. Both issues were analyzed and discussed in two case studies, one with respect to structural mechanics and the other with respect to path-dependent material constitutive behaviors.

Indeed, the physical interpretation of the discovered latent variable could imply the introduction of other measurable variables. This topic should be deeply analyzed.

The main interest in discovering these manifolds is that for a new accessible mechanical state, the output can be inferred by a simple interpolation from its neighbors on the manifold. The main aim of the present work is the construction and analysis of these manifolds. Future works, currently in progress, will focus on their use in performing data-driven simulations.

Conflicts of interest

The authors declare no competing financial interests.

Dedication

This manuscript was written with the help of the contributions of all authors. All authors have given approval to the final version of the manuscript.

Acknowledgments

The first, third, and fourth authors are supported by their respective ESI Group research chairs; their support is gratefully acknowledged. The first author is supported by CREATE-ID ESI-ENSAM research chair. The third author is supported by the ESI Group Chair at the University of Zaragoza. The fourth author is supported by CREATE-ID ESI-ENSAM research chair.

Appendix A. From principal component analysis to its kernel-based counterpart

A.1. Principal component analysis

Let us consider a vector $\mathbf{y} \in \mathbb{R}^D$ containing experimental results or synthetic data from a numerical simulation. These results are often referred to as *snapshots*. If they are obtained by numerical simulation, they consist of nodal values of the essential variable. Therefore, these variables will be somehow correlated and, notably, there will be a linear transformation \mathbf{W} defining the vector $\boldsymbol{\xi} \in \mathbb{R}^d$, with d < D, which contains the still unknown *latent variables*, such that

$$\mathbf{y} = \mathbf{W}\boldsymbol{\xi}.\tag{11}$$

The $D \times d$ transformation matrix **W**, which satisfies the orthogonality condition $\mathbf{W}^T \mathbf{W} = \mathbf{I}_d$, is the main ingredient of the PCA [8].

Assume that there exist M different snapshots $\mathbf{y}_1, \dots, \mathbf{y}_M$, which we store in the columns of a $D \times M$ matrix **Y**. The associated $d \times M$ reduced matrix Ξ contains the associated vectors $\boldsymbol{\xi}_i$, $i = 1, \dots, M$.

.

The PCA usually works with centered variables. In other words,

$$\begin{cases} \sum_{i=1}^{m} \mathbf{y}_i = \mathbf{0}, \\ \sum_{i=1}^{M} \boldsymbol{\xi}_i = \mathbf{0}, \end{cases}$$
(12)

implying the necessity of centering data before applying the PCA.

The PCA proceeds by guaranteeing maximal preserved variance and minimal correlation in the latent variable set $\boldsymbol{\xi}$. The latent variables in $\boldsymbol{\xi}$ are therefore uncorrelated, and consequently the covariance matrix of $\boldsymbol{\xi}$,

$$\mathbf{C}_{\xi\xi} = \mathbf{E}\{\mathbf{\Xi}\mathbf{\Xi}^T\},\tag{13}$$

should be diagonal.

To extract the d uncorrelated latent variables, we proceed from

$$\mathbf{C}_{yy} = \mathrm{E}\{\mathbf{Y}\mathbf{Y}^T\} = \mathrm{E}\{\mathbf{W}\mathbf{\Xi}\mathbf{\Xi}^T\mathbf{W}^T\} = \mathbf{W}\mathrm{E}\{\mathbf{\Xi}\mathbf{\Xi}^T\}\mathbf{W}^T = \mathbf{W}\mathbf{C}_{\xi\xi}\mathbf{W}^T.$$
(14)

Pre- and post-multiplying by \mathbf{W}^T and \mathbf{W} , respectively, and making use of the fact that $\mathbf{W}^T \mathbf{W} = \mathbf{I}$, give us

$$\mathbf{C}_{\xi\xi} = \mathbf{W}^T \mathbf{C}_{\gamma\gamma} \mathbf{W}.$$
 (15)

The covariance matrix \mathbf{C}_{yy} can then be factorized by applying the singular value decomposition,

$$\mathbf{C}_{yy} = \mathbf{V} \mathbf{\Lambda} \mathbf{V}^T, \tag{16}$$
where V contains the orthonormal eigenvectors; Λ is a diagonal matrix containing the eigenvalues sorted in descending order.

Substituting (16) into (15), we arrive at

$$\mathbf{C}_{\boldsymbol{\xi}\boldsymbol{\xi}} = \mathbf{W}^T \mathbf{V} \mathbf{\Lambda} \mathbf{V}^T \mathbf{W}.$$
 (17)

This equality holds when the *d* columns of **W** are taken to be collinear with *d* columns of **V**. We then preserve the eigenvectors associated with the *d* nonzero eigenvalues,

$$\mathbf{W} = \mathbf{V}\mathbf{I}_{D \times d},\tag{18}$$

which gives

$$\mathbf{C}_{\xi\xi} = \mathbf{I}_{d \times D} \mathbf{\Lambda} \mathbf{I}_{D \times d}.$$
 (19)

We therefore conclude that the eigenvalues in Λ represent the variance of the latent variables (diagonal entries of $C_{\xi\xi}$).

A.2. Multidimensional scaling

The PCA works with the covariance matrix of the experimental results, $\mathbf{Y}\mathbf{Y}^{T}$. However, the MDS works with the Gram matrix containing scalar products (i.e., $\mathbf{S} = \mathbf{Y}^{T}\mathbf{Y}$) [8].

The MDS preserves pairwise scalar products:

$$\mathbf{S} = \mathbf{Y}^T \mathbf{Y} = \mathbf{\Xi}^T \mathbf{W}^T \mathbf{W} \mathbf{\Xi} = \mathbf{\Xi}^T \mathbf{\Xi}.$$
 (20)

Computing the eigenvalues of S, we arrive at

$$\mathbf{S} = \mathbf{U}\mathbf{\Lambda}\mathbf{U}^{T} = (\mathbf{U}\mathbf{\Lambda}^{1/2})(\mathbf{\Lambda}^{1/2}\mathbf{U}^{T}) = (\mathbf{\Lambda}^{1/2}\mathbf{U}^{T})^{T}(\mathbf{\Lambda}^{1/2}\mathbf{U}^{T}),$$
(21)

which in turn gives

$$\boldsymbol{\Xi} = \mathbf{I}_{d \times M} \boldsymbol{\Lambda}^{1/2} \mathbf{U}^T.$$
⁽²²⁾

A.3. Kernel-based principal component analysis

The *k*-PCA is based on the fact that data not linearly separable in *D* dimensions could be linearly separated if they are previously projected to a space in Q > D dimensions. However, the true advantage arises from the fact that it is not necessary to write down the analytical expression of that mapping.

The symmetric matrix $\mathbf{\Phi} = \mathbf{Z}^T \mathbf{Z}$, with \mathbf{Z} containing the snapshots $\mathbf{z}_i \in \mathbb{R}^Q$, i = 1, ..., M, associated with $\mathbf{y}_i \in \mathbb{R}^D$, has to be decomposed into eigenvalues and eigenvectors. The procedure for centering data \mathbf{z}_i is carried out in an implicit way.

The eigenvector decomposition reads as

$$\boldsymbol{\Phi} = \boldsymbol{\mathsf{U}}\boldsymbol{\Lambda}\boldsymbol{\mathsf{U}}^T,\tag{23}$$

giving rise to

$$\boldsymbol{\Xi} = \mathbf{I}_{d \times M} \boldsymbol{\Lambda}^{1/2} \mathbf{U}^T.$$
⁽²⁴⁾

The difficulties of operating in a high-dimensional space of dimension, in general, $Q \gg D$, and the mapping unavailability are circumvented by introducing the kernel functional κ (also known as the kernel trick). This allows computing scalar products in \mathbb{R}^Q while operating in \mathbb{R}^D by applying the Mercer theorem. This theorem establishes that if $\kappa(\mathbf{u}, \mathbf{v})$ (where $\mathbf{u} \in \mathbb{R}^D$ and $\mathbf{v} \in \mathbb{R}^D$) is continuous, symmetric, and positive definite, then it defines an inner product in the mapped space \mathbb{R}^Q . Many different kernels exist; some of them are reported in [8].

References

- T. Kirchdoerfer, M. Ortiz, "Data-driven computational mechanics", Comput. Methods Appl. Mech. Eng. 304 (2016), p. 81-101.
- [2] M. A. Bessa, R. Bostanabad, Z. Liu, A. Hu, D. W. Apley, C. Brinson, W. Chen, W. K. Liu, "A framework for data-driven analysis of materials under uncertainty: countering the curse of dimensionality", *Comput. Methods Appl. Mech. Eng.* 320 (2017), p. 633-667.
- [3] Z. Liu, M. Fleming, W. K. Liu, "Microstructural material database for self-consistent clustering analysis of elastoplastic strain softening materials", *Comput. Methods Appl. Mech. Eng.* **330** (2018), p. 547-577.
- [4] D. Gonzalez, F. Chinesta, E. Cueto, "Thermodynamically consistent data-driven computational mechanics", *Contin. Mech. Thermodyn.* 31 (2019), p. 239-253.
- [5] R. Ibanez, E. Abisset-Chavanne, J. Aguado, D. Gonzalez, E. Cueto, F. Chinesta, "A manifold learning approach to datadriven computational elasticity and inelasticity", Arch. Comput. Methods Eng. 25 (2018), no. 1, p. 47-57.
- [6] M. Latorre, F. Montans, "What-you-prescribe-is-what-you-get orthotropic hyperelasticity", *Comput. Mech.* 53 (2014), no. 6, p. 1279-1298.
- [7] P. Ladeveze, D. Neron, P.-W. Gerbaud, "Data-driven computation for history-dependent materials", C. R. Méc. 347 (2019), no. 11, p. 831-844.
- [8] J. A. Lee, M. Verleysen, Nonlinear Dimensionality Reduction, Springer, New York, 2007.
- [9] L. Maaten, G. Hinton, "Visualizing data using t-SNE", J. Mach. Learn Res. 9 (2008), p. 2579-2605.
- [10] S. T. Roweis, L. K. Saul, "Nonlinear dimensionality reduction by locally linear embedding", *Science* 290 (2000), no. 5500, p. 2323-2326.
- [11] N. Kambhatla, T. Leen, "Dimension reduction by local principal component analysis", *Neural Comput.* **9** (1997), no. 7, p. 1493-1516.
- [12] Z. Zhang, H. Zha, "Principal manifolds and nonlinear dimensionality reduction via tangent space alignment", SIAM J. Sci. Comput. 26 (2005), no. 1, p. 313-338.
- [13] A. Badias, S. Curtit, D. Gonzalez, I. Alfaro, F. Chinesta, E. Cueto, "An augmented reality platform for interactive aerodynamic design and analysis", *Int. J. Numer. Methods Eng.* **120** (2019), no. 1, p. 125-138.
- [14] D. Gonzalez, J. Aguado, E. Cueto, E. Abisset-Chavanne, F. Chinesta, "kPCA-based parametric solutions within the PGD framework", Arch. Comput. Methods Eng. 25 (2018), no. 1, p. 69-86.
- [15] E. Lopez, D. Gonzalez, J. V. Aguado, E. Abisset-Chavanne, E. Cueto, C. Binetruy, F. Chinesta, "A manifold learning approach for integrated computational materials engineering", *Arch. Comput. Methods Eng.* 25 (2018), no. 1, p. 59-68.
- [16] E. Lopez, A. Scheuer, E. Abisset-Chavanne, F. Chinesta, "On the effect of phase transition on the manifold dimensionality: application to the Ising model", *Math. Mech. Complex Syst.* 6 (2018), no. 3, p. 251-265.

Comptes Rendus

Mécanique

Objet de la revue

Les *Comptes Rendus Mécanique* sont une revue électronique évaluée par les pairs de niveau international, qui couvre l'ensemble des domaines de la discipline. Ils publient des articles originaux de recherche, des articles de revue, des mises en perspective historiques, des textes à visée pédagogique ou encore des actes de colloque, sans limite de longueur, en anglais ou en français. Les *Comptes Rendus Mécanique* sont diffusés selon une politique vertueuse de libre accès diamant, gratuit pour les auteurs (pas de frais de publication) comme pour les lecteurs (libre accès immédiat et pérenne).

Directeur de la publication : Étienne Ghys

Rédacteur en chef: Jean-Baptiste Leblond

Comité éditorial : Olga Budenkova, Francisco Chinesta, Jean-Michel Coron, Luc Dormieux, Florian Gosselin, Nicolas Moës, Léo Morin, Guillaume Ribert, Géry de Saxcé, Emmanuel Villermaux **Secrétaire éditoriale :** Adenise Lopes

À propos de la revue

Toutes les informations concernant la revue, y compris le texte des articles publiés qui est en accès libre intégral, figurent sur le site https://comptes-rendus.academie-sciences.fr/mecanique/.

Informations à l'attention des auteurs

Pour toute question relative à la soumission des articles, les auteurs peuvent consulter le site https://comptes-rendus.academie-sciences.fr/mecanique/.

Contact

Académie des sciences 23, quai de Conti, 75006 Paris, France Tél. : (+33) (0)1 44 41 43 72 CR-Mecanique@academie-sciences.fr





Mécanique

Volume 348, nº 10-11, octobre-novembre 2020

Special issue / Numéro thématique Contributions in mechanics of materials / *Contributions en mécanique des matériaux*

Guest editors / *Rédacteurs en chef invités* Julie Diani, Olivier Castelnau Francisco Chinesta

J. Diani, O. Castelnau, F. Chinesta

Cover illustration / *Illustration de couverture* © Katell Derrien, Hervé Moulinec, Olivier Castelnau

Foreword 781-783 **Emmanuel Richaud** Physico-chemical approach of polymer chemical ageing: a short review 785-795 Julie Diani Free vibrations of linear viscoelastic polymer cantilever beams 797-806 Yann Charles, Chunping Zhang, Monique Gaspérini, Brigitte Bacroix Identification methodology of a rate-sensitive constitutive law with mean field and full field modeling approaches for polycrystalline materials 807-826 O. Castelnau, K. Derrien, S. Ritterbex, P. Carrez, P. Cordier, H. Moulinec Multiscale modeling of the effective viscoplastic behavior of Mg₂SiO₄ wadslevite: bridging atomic and polycrystal scales 827-846 Lu Tuan Le, Kais Ammar, Samuel Forest Efficient simulation of single and poly-crystal plasticity based on the pencil glide mechanism 847-876 Carlos N. Tomé, Ricardo A. Lebensohn Joshua Furer, Martin Idiart, Pedro Ponte Castañeda Exact results for weakly nonlinear composites and implications for homogenization methods 893-909 David Ryckelynck, Thibault Goessel, Franck Nguyen Mechanical dissimilarity of defects in welded joints via Grassmann manifold and machine learning 911-935 Ruben Ibanez, Pierre Gilormini, Elias Cueto, Francisco Chinesta Numerical experiments on unsupervised manifold learning applied to mechanical modeling of materials and structures 937-958

1873-7234 (electronic)





