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C. R. Mecanique 333 (2005) 219–226



<http://france.elsevier.com/direct/CRAS2B/>

On the effect of small fluctuations in the volume fraction of constituents on the effective properties of composites

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Received 7 June 2004, accepted after revision 14 December 2004

Available online 18 January 2005

Presented by Pierre Suquet

Abstract

This study deals with three-scale composite materials comprised of nonlinear constituents. At the meso scale the composite can be considered as locally homogeneous with a macroscopic spatial variation of the constituents volume fraction. When these variations about a mean value are small, a Taylor expansion to second-order of the effective properties of the composite with respect to the fluctuations is given. This expansion can be used to discuss the beneficial or deleterious effects of clusters of inhomogeneities. It can also be used to derive new upper and lower bounds for the effective properties of nonlinear composites from dilute results. *To cite this article: P. Suquet, C. R. Mecanique 333 (2005).*

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Résumé

Effet de petites fluctuations de concentration sur les propriétés effectives des composites. Cette étude traite de composites à trois échelles nettement séparées. Ces matériaux peuvent être considérés comme localement homogènes à l'échelle mésoscopique, mais avec une fraction volumique des constituants qui varie d'un point à l'autre. Quand ces fluctuations de concentration sont faibles, un développement de Taylor des propriétés effectives macroscopiques du composite est établi. Ce développement est utilisé pour discuter l'effet des amas d'hétérogénéités. Il permet également d'obtenir des bornes supérieures et inférieures nouvelles sur les propriétés effectives macroscopiques de composites non linéaires. *Pour citer cet article : P. Suquet, C. R. Mecanique 333 (2005).*

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Keywords: Computational solid mechanics; Homogenization; Composite materials; Nonlinear behavior

Mots-clés: Mécanique des solides numérique ; Homogénéisation ; Matériaux composites ; Comportement non linéaire

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doi:10.1016/j.crme.2004.12.004

Version française abrégée

L'objet de cette Note est d'étudier l'effet de fluctuations locales de concentration autour d'une valeur moyenne sur les propriétés effectives de composites linéaires ou non linéaires. Les composites étudiés présentent trois échelles, qualifiées respectivement de microscopique, mésoscopique et macroscopique (Fig. 1). Pour simplifier on se limite à des composites biphasés et la fraction volumique de phase 1 est notée f . A l'échelle mésoscopique ces composites peuvent être considérés comme localement homogènes, mais avec une fraction volumique des constituants qui varie d'un point à l'autre. Les propriétés mésoscopiques du composite sont régies par une énergie effective mésoscopique $\tilde{w}(f, \boldsymbol{\varepsilon})$ où la fraction volumique f dépend de la variable spatiale mésoscopique \boldsymbol{x} .

Nous nous plaçons dans une situation où les variations de f autour d'une valeur moyenne f_0 sont faibles et mesurées par un petit paramètre t , $f_t(\boldsymbol{x}) = f_0 + t\delta f(\boldsymbol{x})$, avec $\langle \delta f \rangle = 0$. L'énergie effective macroscopique $\tilde{\tilde{w}}$ du composite est définie par :

$$\tilde{\tilde{w}}(t, \bar{\boldsymbol{\varepsilon}}) = \inf_{\boldsymbol{u} \in \mathcal{K}(\bar{\boldsymbol{\varepsilon}})} \left(\tilde{w}(f_t, \boldsymbol{\varepsilon}(\boldsymbol{u})) \right)$$

Un développement de Taylor de $\tilde{\tilde{w}}$ par rapport à t peut être établi. En se limitant à l'ordre 2, ce développement s'écrit :

$$\tilde{\tilde{w}}(t, \bar{\boldsymbol{\varepsilon}}) = \tilde{w}(f_0, \bar{\boldsymbol{\varepsilon}}) + \frac{t^2}{2} \left(\langle \delta f^2 \rangle \frac{\partial^2 \tilde{w}}{\partial f^2}(f_0, \bar{\boldsymbol{\varepsilon}}) - \frac{\partial^2 \tilde{w}}{\partial f \partial \boldsymbol{\varepsilon}}(\bar{\boldsymbol{\varepsilon}}) : \boldsymbol{H} : \frac{\partial^2 \tilde{w}}{\partial f \partial \boldsymbol{\varepsilon}}(\bar{\boldsymbol{\varepsilon}}) \right) + O(t^3)$$

où \boldsymbol{H} est défini par (12).

L'effet des fluctuations δf peut être bénéfique ou pénalisant selon le signe du facteur de t^2 dans ce développement asymptotique : l'énergie macroscopique $\tilde{\tilde{w}}$ est alors supérieure ou inférieure à l'énergie mésoscopique \tilde{w} .

Ce développement asymptotique peut également être utilisé pour obtenir des bornes sur les propriétés effectives des composites. Considérons pour simplifier la classe des matériaux biphasés isotropes en dimension 2 et soit $\tilde{w}^-(f, \bar{\boldsymbol{\varepsilon}})$ une borne inférieure pour les propriétés effectives de cette classe réalisable par certaines microstructures. En mélangeant des microstructures optimales de fractions volumiques f_1 et f_2 différentes mais proches de leur moyenne f_0 , on génère grâce au développement asymptotique précédent une inégalité (14) qui doit être satisfaite par \tilde{w}^- . Dans le cas particulier des matériaux en loi puissance, cette inégalité prend la forme simple (20) qui dépend d'une constante β_0^- . Cette constante exprime le comportement de la borne inférieure aux faibles fractions volumiques (concentrations diluées).

1. Introduction

The composites under consideration are three-scale composites characterized by the presence of heterogeneities at different scales hereafter called the micro, meso and macro scales. These scales are characterized by different lengths d , a and L (see Fig. 1): d is the typical size of single inhomogeneities, a is the typical size of regions which contain a large number of inhomogeneities but are small with respect to the entire representative volume element and L is the size of a typical representative volume element. At the microscopic scale the composite appears to be highly heterogeneous, as shown in Fig. 1 left. At the mesoscopic scale the sharp micro-inhomogeneities are 'smeared out' as shown in Fig. 1 right and the composite appears now as an intermediate composite, or meso-composite, comprised of different constituents with a mesostructure representing the different regions, with different volume fraction of constituents, in the actual composite. Finally, at the largest scale all inhomogeneities are smeared out and the composite is seen as an homogeneous medium, the properties of which being, by definition, the effective properties of the composite.

When the three scales are 'well separated', which means that the three typical lengths satisfy the string of inequalities $d \ll a \ll L$, the effective properties of the composite can be obtained by splitting the homogenization

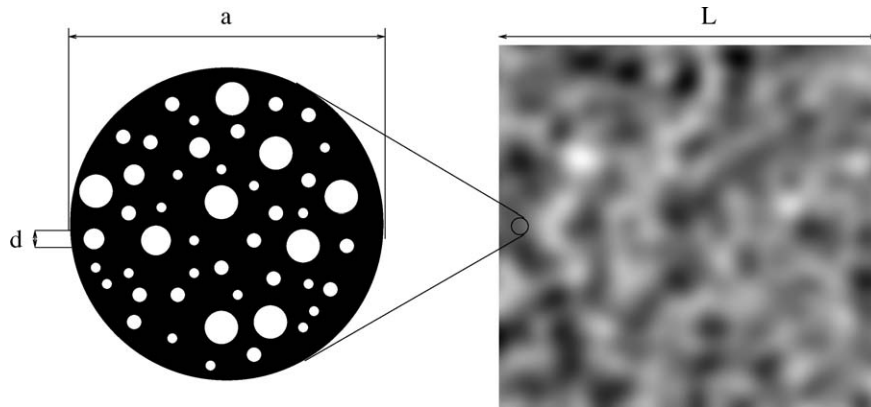


Fig. 1. Three-scale composite. Left: micro-scale. f is volume fraction of the white phase. Right: meso-scale showing fluctuations in the local volume fraction $f(\mathbf{x})$.

procedure into two successive steps. In a first step the effective properties of the mesoscopic regions can be derived by an homogenization procedure appropriate for the microstructure of each region. Then, in a second stage, the meso-composite formed by the mesoscopic regions is homogenized using an homogenization scheme appropriate to its mesostructure.

For simplicity, the composites considered in this study are, at the microscopic scale, two-phase materials, the volume fraction of phase 1 being denoted by f and the volume fraction of phase 2 being $1 - f$. When the stress–strain relations for the individual constituents derive from a strain-energy, the stress–strain relations at the mesoscopic scale also derive from a strain energy with depends on the mesoscopic strain and on other microstructural parameters including the volume fraction of phase 1, f . This mesoscopic strain-energy will be denoted as $\tilde{w}(f, \boldsymbol{\varepsilon})$ where the other microstructural variables are assumed to be fixed (a similar theory can be developed for general variations of the microstructural parameters).

The volume fraction $f(\mathbf{x})$ of phase 1 can depend on the mesoscopic spatial variable \mathbf{x} as shown in Fig. 1. Now it is assumed that these fluctuations about a mean-value f_0 are small. In other words, the average volume fraction of the inclusions is f_0 at the level of the representative volume element. However, at the mesoscopic scale it can fluctuate slightly about this mean value and is described by a variable volume fraction $f(\mathbf{x})$ with $f(\mathbf{x}) - f_0 \ll f_0$:

$$f_t(\mathbf{x}) = f_0 + t\delta f(\mathbf{x}), \quad \text{with } \langle \delta f \rangle = 0, \quad t \ll 1 \tag{1}$$

The volume fraction fluctuations are assumed to be small so that t is a small parameter.

The aim of this Note is to assess the influence on the effective properties of the composite of small fluctuations in the volume fraction of the constituents. Do these fluctuations strengthen or weaken the composite? An expansion which is exact to second-order in the volume fraction fluctuations is given in Section 2.

An unexpected consequence of this expansion is that bounds for the effective properties of composites can be deduced from this expansion, provided that exact results are known for composites in the dilute limit (see Section 3).

2. A general result for small fluctuations in the local volume fraction of constituents

As recalled in the introduction the homogenization procedure for three-scale composites involves a first step by which the mesoscopic effective energy $\tilde{w}(f, \boldsymbol{\varepsilon})$ is determined. Then, the macroscopic effective energy of the composite, which depends on the small parameter t , is defined as

$$\tilde{\tilde{w}}(t, \bar{\boldsymbol{\varepsilon}}) = \text{Inf}_{\mathbf{u} \in \mathcal{K}(\bar{\boldsymbol{\varepsilon}})} \langle \tilde{w}(f_t, \boldsymbol{\varepsilon}(\mathbf{u})) \rangle = \langle \tilde{w}(f_t, \boldsymbol{\varepsilon}(\mathbf{u}_t)) \rangle \tag{2}$$

where $\mathcal{K}(\bar{\boldsymbol{\varepsilon}}) = \{\mathbf{v} = \bar{\boldsymbol{\varepsilon}} \cdot \mathbf{x} + \mathbf{v}^*, \mathbf{v}^* \text{ periodic}\}$. The fields \mathbf{u}_t and $\boldsymbol{\varepsilon}(\mathbf{u}_t)$ are the local displacement and associated strain fields at the meso-scale induced by appropriate boundary conditions generating an average strain $\bar{\boldsymbol{\varepsilon}}$ in V . It will be assumed that $\tilde{w}(t, \cdot)$ and \mathbf{u}_t are continuously differentiable functions of t . Since t is small, it is appropriate to look for a perturbation series expansion of \tilde{w} about $t = 0$.

The main result of this section is the following expansion

$$\tilde{w}(t, \bar{\boldsymbol{\varepsilon}}) = \tilde{w}(f_0, \bar{\boldsymbol{\varepsilon}}) + \frac{t^2}{2} \left(\langle \delta f^2 \rangle \frac{\partial^2 \tilde{w}}{\partial f^2}(f_0, \bar{\boldsymbol{\varepsilon}}) - \frac{\partial^2 \tilde{w}}{\partial f \partial \boldsymbol{\varepsilon}}(\bar{\boldsymbol{\varepsilon}}) : \mathbf{H} : \frac{\partial^2 \tilde{w}}{\partial f \partial \boldsymbol{\varepsilon}}(\bar{\boldsymbol{\varepsilon}}) \right) + \mathcal{O}(t^3) \quad (3)$$

where \mathbf{H} is a fourth-order tensor specified by (12).

Remark 1. In general, fluctuations in the volume fraction have no first-order effect on the effective potential but have a second-order effect. This is true as long as the Taylor expansion (3) is legitimate. In particular, when the tensor \mathbf{H} becomes singular, the expansion (3) is no more valid.

Remark 2. Fluctuations have a first-order effect on the local fields, reflected by the first-order terms $\dot{\mathbf{u}}_0$ and $\dot{\boldsymbol{\sigma}}_0$.

Remark 3. The fluctuations enter this second-order expansion through their covariance $\langle \delta f^2 \rangle$ and through the fourth-order tensor \mathbf{H} which depends on the geometrical arrangement of the domains where the perturbations around the mean value f_0 take place.

Remark 4. Fluctuations in volume fraction can have, at second-order, a beneficial or deleterious effect on the effective potential \tilde{w} , depending on the sign of the second-order term.

Derivation of (3): The expansion of \tilde{w} in powers of t is formally given by its Taylor expansion at $t = 0$. The first derivatives in this expansion may be computed by successive derivation of (2); this procedure will naturally involve derivatives of \mathbf{u}_t . The problem to be solved for \mathbf{u}_t is given by

$$\boldsymbol{\sigma}_t = \frac{\partial \tilde{w}}{\partial \boldsymbol{\varepsilon}}(f_t, \boldsymbol{\varepsilon}(\mathbf{u}_t)), \quad \text{div}(\boldsymbol{\sigma}_t) = \mathbf{0}, \quad \mathbf{u}_t \in \mathcal{K}(\bar{\boldsymbol{\varepsilon}}) \quad (4)$$

By differentiation of (4), it is seen that $\dot{\mathbf{u}}_t$ (the dot denoting the derivative with respect to t) is, for all t , the solution of the systems of equations:

$$\dot{\boldsymbol{\sigma}}_t = \mathbf{L}_t : \boldsymbol{\varepsilon}(\dot{\mathbf{u}}_t) + \boldsymbol{\tau}_t, \quad \text{div}(\dot{\boldsymbol{\sigma}}_t) = \mathbf{0}, \quad \dot{\mathbf{u}}_t \in \mathcal{K}(\mathbf{0}) \quad (5)$$

where

$$\mathbf{L}_t = \frac{\partial^2 \tilde{w}}{\partial \boldsymbol{\varepsilon} \partial \boldsymbol{\varepsilon}}(f_t, \boldsymbol{\varepsilon}(\mathbf{u}_t)), \quad \boldsymbol{\tau}_t = \delta f(\mathbf{x}) \frac{\partial^2 \tilde{w}}{\partial f \partial \boldsymbol{\varepsilon}}(f_t, \boldsymbol{\varepsilon}(\mathbf{u}_t))$$

\mathbf{L}_t is the tensor of instantaneous or tangent moduli. It follows from (2) and from Hill's lemma that

$$\frac{\partial \tilde{w}}{\partial t}(t, \bar{\boldsymbol{\varepsilon}}) = \left\langle \frac{\partial \tilde{w}}{\partial \boldsymbol{\varepsilon}}(f_t, \boldsymbol{\varepsilon}(\mathbf{u}_t)) : \boldsymbol{\varepsilon}(\dot{\mathbf{u}}_t) \right\rangle + \left\langle \delta f(\mathbf{x}) \frac{\partial \tilde{w}}{\partial f}(f_t, \boldsymbol{\varepsilon}(\mathbf{u}_t)) \right\rangle = \left\langle \delta f(\mathbf{x}) \frac{\partial \tilde{w}}{\partial f}(f_t, \boldsymbol{\varepsilon}(\mathbf{u}_t)) \right\rangle \quad (6)$$

Differentiating again yields:

$$\frac{\partial^2 \tilde{w}}{\partial t^2}(t, \bar{\boldsymbol{\varepsilon}}) = \left\langle (\delta f(\mathbf{x}))^2 \frac{\partial^2 \tilde{w}}{\partial f^2}(f_t, \boldsymbol{\varepsilon}(\mathbf{u}_t)) \right\rangle + \left\langle \delta f(\mathbf{x}) \frac{\partial^2 \tilde{w}}{\partial f \partial \boldsymbol{\varepsilon}}(f_t, \boldsymbol{\varepsilon}(\mathbf{u}_t)) : \boldsymbol{\varepsilon}(\dot{\mathbf{u}}_t) \right\rangle \quad (7)$$

By means of the system of Eqs. (5) satisfied by $\dot{\mathbf{u}}_t$, one obtains finally that:

$$\frac{\partial^2 \tilde{w}}{\partial t^2}(t, \bar{\boldsymbol{\varepsilon}}) = \left\langle (\delta f(\mathbf{x}))^2 \frac{\partial^2 \tilde{w}}{\partial f^2}(f_t, \boldsymbol{\varepsilon}(\mathbf{u}_t)) \right\rangle - \left\langle \boldsymbol{\varepsilon}(\dot{\mathbf{u}}_t) : \mathbf{L}_t : \boldsymbol{\varepsilon}(\dot{\mathbf{u}}_t) \right\rangle \quad (8)$$

The relations (6) and (8), which hold for arbitrary values of t , give the first two derivatives of \tilde{w} at $t = 0$. The material is homogeneous at $t = 0$. Therefore $\mathbf{u}_0 = \bar{\boldsymbol{\varepsilon}} \cdot \mathbf{x}$ and:

$$\left. \begin{aligned} \tilde{w}(0, \bar{\boldsymbol{\varepsilon}}) &= w(f_0, \bar{\boldsymbol{\varepsilon}}), & \frac{\partial \tilde{w}}{\partial t}(0, \bar{\boldsymbol{\varepsilon}}) &= \langle \delta f \rangle \frac{\partial \tilde{w}}{\partial f}(f_0, \bar{\boldsymbol{\varepsilon}}) = 0 \quad (\text{since } \langle \delta f \rangle = 0) \\ \frac{\partial^2 \tilde{w}}{\partial t^2}(0, \bar{\boldsymbol{\varepsilon}}) &= \left(\langle (\delta f(\mathbf{x}))^2 \rangle \frac{\partial^2 \tilde{w}}{\partial f^2}(f_0, \bar{\boldsymbol{\varepsilon}}) - \langle \boldsymbol{\varepsilon}(\dot{\mathbf{u}}_0) : \mathbf{L}^{(0)} : \boldsymbol{\varepsilon}(\dot{\mathbf{u}}_0) \rangle \right) \\ \text{where } \mathbf{L}^{(0)} &= \frac{\partial^2 \tilde{w}}{\partial \boldsymbol{\varepsilon} \partial \boldsymbol{\varepsilon}}(f_0, \bar{\boldsymbol{\varepsilon}}) \end{aligned} \right\} \quad (9)$$

In these relations, $\dot{\mathbf{u}}_0$ is the solution of the linear thermoelasticity problem:

$$\dot{\boldsymbol{\sigma}}_0 = \mathbf{L}^{(0)} : \boldsymbol{\varepsilon}(\dot{\mathbf{u}}_0) + \boldsymbol{\tau}, \quad \text{div}(\dot{\boldsymbol{\sigma}}_0) = \mathbf{0}, \quad \dot{\mathbf{u}}_0 \in \mathcal{K}(\mathbf{0}), \quad \text{where } \boldsymbol{\tau}(\mathbf{x}) = \delta f(\mathbf{x}) \frac{\partial^2 \tilde{w}}{\partial f \partial \boldsymbol{\varepsilon}}(f_0, \bar{\boldsymbol{\varepsilon}}) \quad (10)$$

Given that the modulus tensor $\mathbf{L}^{(0)}$ is constant, problem (10) is a standard linear thermoelasticity problem for a homogeneous material with a distribution of body forces determined by the polarization field $\boldsymbol{\tau}$. Its solution can be expressed as $\boldsymbol{\varepsilon}(\dot{\mathbf{u}}_0) = -\boldsymbol{\Gamma}^0 * \boldsymbol{\tau}$, where $\boldsymbol{\Gamma}^0$ is the Green operator associated with $\mathbf{L}^{(0)}$.

When the fluctuations are piecewise constant, i.e. constant in N ‘regions’ V_r with characteristic function $\chi^{(r)}$, the polarization field is piecewise constant with:

$$\boldsymbol{\tau}(\mathbf{x}) = \sum_{r=1}^N \chi^{(r)}(\mathbf{x}) \boldsymbol{\tau}^{(r)}, \quad \boldsymbol{\tau}^{(r)} = \frac{\partial^2 \tilde{w}}{\partial f \partial \boldsymbol{\varepsilon}}(f_0, \bar{\boldsymbol{\varepsilon}}) \delta f^{(r)}$$

Then, taking advantage of the fact that the elastic moduli $\mathbf{L}^{(0)}$ are uniform, one obtains that:

$$\langle \boldsymbol{\varepsilon}(\dot{\mathbf{u}}_0) : \mathbf{L}^{(0)} : \boldsymbol{\varepsilon}(\dot{\mathbf{u}}_0) \rangle = \sum_{r=1}^N \sum_{s=1}^N \boldsymbol{\tau}^{(r)} : \boldsymbol{\Gamma}^{(rs)} : \boldsymbol{\tau}^{(s)}, \quad \text{where } \boldsymbol{\Gamma}^{(rs)} = \langle \chi^{(r)} \boldsymbol{\Gamma}^0 * \chi^{(s)} \rangle \quad (11)$$

In conclusion, when fluctuations have zero average around the mean value f_0 , the expansion (3) is obtained with:

$$\mathbf{H} = \sum_{r=1}^N \sum_{s=1}^N \delta f^{(r)} : \boldsymbol{\Gamma}^{(rs)} : \delta f^{(s)} \quad (12)$$

Fluctuations constant on two subregions: Consider the simple case where the volume fraction f takes only two different values f_1 and f_2 , close to each other, on two subregions V_1 and V_2 with volume fraction $c^{(1)}$ and $c^{(2)} = 1 - c^{(1)}$. Let $f_0 = c^{(1)} f_1 + c^{(2)} f_2$ be the average volume fraction in the composite and $\Delta f = (f_1 - f_2)$. Then

$$t^2 \langle \delta f^2 \rangle = c^{(1)} c^{(2)} (\Delta f)^2, \quad t^2 \mathbf{H} = c^{(1)} c^{(2)} (\Delta f)^2 \mathbf{P}, \quad \mathbf{P} = \frac{1}{c^{(1)} c^{(2)}} \langle \chi^{(1)} \boldsymbol{\Gamma}^0 * \chi^{(1)} \rangle$$

Finally the expansion (3) reduces in this case to

$$\tilde{w}(0, \bar{\boldsymbol{\varepsilon}}) = \tilde{w}(f_0, \bar{\boldsymbol{\varepsilon}}) + \frac{c^{(1)} c^{(2)} (\Delta f)^2}{2} \left(\frac{\partial^2 \tilde{w}}{\partial f^2}(f_0, \bar{\boldsymbol{\varepsilon}}) - \frac{\partial^2 \tilde{w}}{\partial f \partial \boldsymbol{\varepsilon}}(f_0, \bar{\boldsymbol{\varepsilon}}) : \mathbf{P} : \frac{\partial^2 \tilde{w}}{\partial f \partial \boldsymbol{\varepsilon}}(f_0, \bar{\boldsymbol{\varepsilon}}) \right) + O(t^3) \quad (13)$$

3. Connection with upper and lower bounds for nonlinear composites

3.1. Deriving bounds for the effective potentials of nonlinear composites from dilute results

The above expansion can be used to derive upper and lower bounds for the effective potential of composites from dilute results.

Let us consider for simplicity a two-phase isotropic composite. Let $\tilde{w}^-(f, \bar{\boldsymbol{\varepsilon}})$ be an attainable lower bound for the effective energy of the composite where f is the volume fraction of one phase. Since \tilde{w}^- is attainable for every volume fraction f in $[0, 1]$, there exists a microstructure which achieves $\tilde{w}^-(f, \cdot)$. In particular there exist two isotropic microstructures corresponding to f_1 and f_2 where f_1 and f_2 are two volume fractions close to f_0 , such that $f_1 \leq f_0 \leq f_2$ and $f_0 = c^{(1)}f_1 + (1 - c^{(1)})f_2$ with $c^{(1)} \in [0, 1]$. A new composite can be formed by assembling (isotropically) different regions made of these two microstructures in proportions $c^{(1)}$ and $c^{(2)} = 1 - c^{(1)}$. The resulting composite is an isotropic composite in which the volume fraction of phase 1 is f_0 . Its effective energy $\tilde{\tilde{w}}$ is certainly larger than $\tilde{w}^-(f_0, \cdot)$ since \tilde{w}^- is a lower bound for all isotropic composites made of the two constituents under consideration in proportion f_0 . Since the expansion to second-order of $\tilde{\tilde{w}}$ is given by (13), the second-order term in this expansion has to be positive and this leads to the inequality:

$$\frac{\partial^2 \tilde{w}^-}{\partial f^2}(f_0, \bar{\boldsymbol{\varepsilon}}) - \frac{\partial^2 \tilde{w}^-}{\partial f \partial \boldsymbol{\varepsilon}}(f_0, \bar{\boldsymbol{\varepsilon}}) : \mathbf{P} : \frac{\partial^2 \tilde{w}^-}{\partial f \partial \boldsymbol{\varepsilon}}(f_0, \bar{\boldsymbol{\varepsilon}}) \geq 0 \quad (14)$$

This differential inequality can be used to derive a lower bound for \tilde{w}^- .

For simplicity let us consider the case of two-phase, two-dimensional, incompressible, power-law materials deriving from the energy $w^{(r)} = (\sigma_0^{(r)} \varepsilon_0 / (m + 1)) (\varepsilon_{\text{eq}} / \varepsilon_0)^{m+1}$. In dimension 2, when all constituents have the same nonlinearity exponent m and when the composite has overall isotropy, the effective potential for the composite has the form:

$$\tilde{w}(f, \boldsymbol{\varepsilon}) = \frac{\tilde{\sigma}_0(m, f) \varepsilon_0}{m + 1} \left(\frac{\varepsilon_{\text{eq}}}{\varepsilon_0} \right)^{m+1} \quad (15)$$

This simple form permits to derive explicit expressions for $\mathbf{L}^{(0)}$, \mathbf{P} and $\boldsymbol{\tau}$. Then, following similar developments in Ponte Castañeda and Suquet [1], one obtains:

$$\boldsymbol{\tau} : \mathbf{P} : \boldsymbol{\tau} = C(m) \frac{\varepsilon_0}{\tilde{\sigma}_0(m, f)} \left(\frac{\partial \tilde{\sigma}_0}{\partial f}(m, f) \right)^2 \left(\frac{\varepsilon_{\text{eq}}}{\varepsilon_0} \right)^{m+1}, \quad C(m) = \frac{1}{m + \sqrt{m}} \quad (16)$$

Finally the second-order correction in the volume fraction fluctuations reads as

$$\frac{\partial^2 \tilde{w}}{\partial f^2}(f_0, \bar{\boldsymbol{\varepsilon}}) - \boldsymbol{\tau} : \mathbf{P} : \boldsymbol{\tau} = \frac{\varepsilon_0}{m + 1} \left(\frac{\varepsilon_{\text{eq}}}{\varepsilon_0} \right)^{m+1} \left[\frac{\partial^2 \tilde{\sigma}_0}{\partial f^2}(m, f) - \frac{(m + 1)C(m)}{\tilde{\sigma}_0(m, f)} \left(\frac{\partial \tilde{\sigma}_0}{\partial f}(m, f) \right)^2 \right] \quad (17)$$

\tilde{w}^- is a lower bound for all isotropic microstructures when the associated flow stress $\tilde{\sigma}_0^-$ satisfies the inequality:

$$\frac{\partial^2 \tilde{\sigma}_0^-}{\partial f^2}(m, f) - \frac{(m + 1)C(m)}{\tilde{\sigma}_0^-(m, f)} \left(\frac{\partial \tilde{\sigma}_0^-}{\partial f}(m, f) \right)^2 \geq 0 \quad (18)$$

This inequation can be re-written as:

$$\frac{\partial}{\partial f} \left[\frac{1}{\tilde{\sigma}_0^-(m, f)^\alpha} \frac{\partial \tilde{\sigma}_0^-}{\partial f}(m, f) \right] \geq 0, \quad \alpha(m) = (m + 1)C(m) = \frac{m + 1}{m + \sqrt{m}} \quad (19)$$

(19) can be integrated twice from 0 to f . The result of this integration reads when $m < 1$ (so that $\alpha > 1$):

$$\tilde{\sigma}_0^-(m, f) \geq \sigma_0^{(2)} \left\{ 1 + [1 - \alpha(m)] \beta_0^-(m) f \right\}^{1/(1-\alpha(m))} \quad (20)$$

where $\sigma_0^{(2)} = \tilde{\sigma}_0^-(0)$ is the flow stress of phase 2 and $\beta_0^-(m) = \frac{1}{\sigma_0^{(2)}} \frac{\partial \tilde{\sigma}_0^-}{\partial f}(m, 0)$. The coefficient β_0^- , which depends on m and on the contrast between the microscopic phases, characterizes the dependence of the lower bound $\tilde{\sigma}_0^-(f, m)$ on f as f approaches 0 (dilute limit).

When $m = 1$ (then $\alpha = 1$) the double integration of (19) yields

$$\tilde{\sigma}_0^-(f, m) \geq \sigma_0 \exp(\beta(1) f) \quad (21)$$

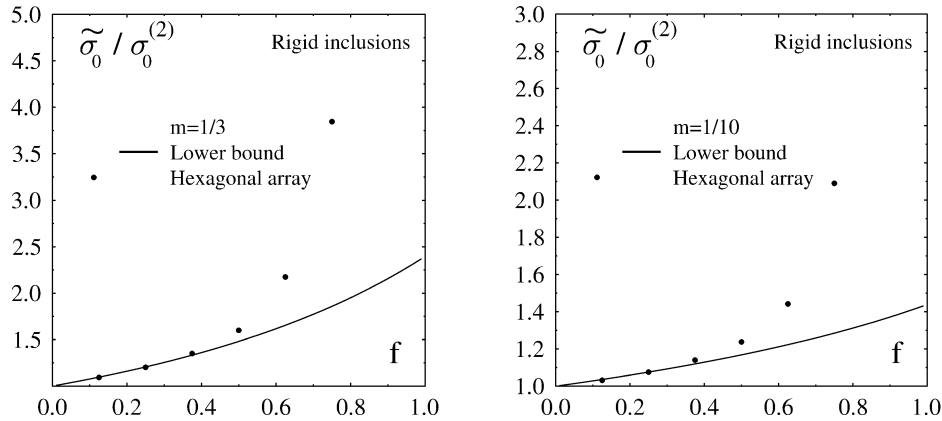


Fig. 2. Rigidly reinforced two-dimensional composites. Effective flow-stress (15) as a function of the volume fraction $c^{(1)}$ of the rigid phase. Left: $m = 1/3$. Right $m = 1/5$.

3.2. Comments

1. The inequality (14) shows that any attainable lower bound $\tilde{w}^-(f, \boldsymbol{\epsilon})$ has to be a convex function of f , since \mathbf{P} is a positive fourth-order tensor.

2. An upper bound can be obtained by similar arguments. Indeed if \tilde{w}^+ is an attainable upper bound the associated effective flow stress $\tilde{\sigma}_0^+$ must satisfy the inequality:

$$\frac{\partial^2 \tilde{\sigma}_0^+}{\partial f^2}(m, f) - \frac{(m+1)C(m)}{\tilde{\sigma}_0^+(m, f)} \left(\frac{\partial \tilde{\sigma}_0^+}{\partial f}(m, f) \right)^2 \leq 0 \tag{22}$$

which gives by integration:

$$\tilde{\sigma}_0^+(m, f) \leq \sigma_0^{(2)} \{ 1 + [1 - \alpha(m)] \beta_0^+(m) f \}^{1/(1-\alpha(m))} \tag{23}$$

where $\beta_0^+(m)$ now characterizes the dependence of the upper bound $\tilde{\sigma}_0^+$ on f in the dilute limit.

3. As expected, the lower bound (21) is weaker than the lower Hashin–Shtrikman bound for two-phase (two-dimensional) composites made of incompressible linear phases, since the HS lower bound is indeed optimal.

4. On the positive side, only very few lower bounds improving on the Reuss bound are known for nonlinear composites (when their energy grows more slowly than a quadratic function). Noticeable exceptions are the works of Talbot and Willis [2] and Garroni et al. [3]. The inequality (20) seems to be a new lower bound (certainly not optimal). On the negative side, (20) depends on a coefficient β which is not given by the theory and which has to be determined by other means (see for instance [4] for an example of such a determination).

5. When m goes to 0, the scalar $(m+1)C(m)$ tends to $+\infty$. Therefore the inequality (18) implies that any lower bound for $\tilde{\sigma}$ (assumed to be a twice-differentiable function of f) has to be such that $\lim_{m \rightarrow 0} \partial \tilde{\sigma}_0^- / \partial f = 0$. In conclusion, under the assumption that $\partial \tilde{\sigma}_0^- / \partial f(m, f)$ is a continuous function of m , $\tilde{\sigma}_0^-(0, f)$ has to be constant (with respect to f). In particular when $f = 0$ and $f = 1$, this constant must be less than the flow stress of the individual phases. The best constant meeting this constraint is the Reuss lower bound, i.e. the smallest flow stress of the phases. In other words, under the above assumptions of differentiability with respect to f and continuity with respect to m , the Reuss lower bound is an optimal lower bound when $m = 0$. In a similar (but slightly different) problem for dielectric materials, Garroni et al. [3] reached the same conclusion by completely different means. They actually showed that there exist isotropic microstructures for which the Reuss bound is exact.

3.3. A numerical example

A comparison with numerical simulations can be performed to assess the magnitude of the underestimation provided by the lower bound (20) for specific microstructures which correspond to ‘weak’ composites. When phase 1 is rigid, the weakest microstructures are those where the fibers are dispersed in the deformable matrix. The case of an hexagonal array has been investigated by J.C. Michel (private communication). Although the nonlinear effective properties of such arrays are not exactly isotropic, the deviation from isotropy remains reasonable and the comparison with the lower bound (20) is sensible. As shown in Fig. 2, the predictions of the lower bound lie below the computed effective properties at all volume fraction. Numerical values for the coefficient β appearing in (20) are taken from Ponte Castañeda [5] (which itself makes use of numerical calculations by Lee and Mear [4]). The effective properties for the hexagonal array tend to blow-up when the volume fraction of the rigid phase approaches the compact piling-up volume fraction ($f = \pi/2\sqrt{3}$). In contrast, the lower bound (20) remains finite as f tends to 1.

Acknowledgements

The author is indebted to J.C. Michel for providing him with the numerical results for hexagonal arrays of rigid fibers shown in Fig. 2.

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