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A two-scale reaction–diffusion system with micro-cell reaction concentrated on a free boundary

Sebastian A. Meier [a], Adrian Muntean [b,*]

[a] *Centre for Industrial Mathematics (ZeTeM), FB3, University of Bremen, Postfach 330 440, 28334 Bremen, Germany*
[b] *CASA-Centre for Analysis, Scientific Computing and Applications, Department of Mathematics and Computer Science, Technical University of Eindhoven, PO Box 513, 5600 MB Eindhoven, The Netherlands*

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Abstract

We discuss the fast-reaction limit of a two-scale reaction–diffusion model. We point out that if the reaction constant a explodes to infinity, then a two-scale PDE system with free boundary at the micro cell is obtained. The aim of this note is to answer the question: Can the same two-scale free-boundary problem be obtained if we first pass to the fast-reaction limit $a \to \infty$ and then take the homogenisation limit $\varepsilon \to 0$ that is behind the derivation of the two-scale model? Here ε is the width of a thin two-dimensional strip. Using the method of asymptotic expansions, we show that it does not matter whether we first take $\varepsilon \to 0$ and then $a \to \infty$, or vice-versa. Finally, we illustrate numerically the solution behaviour of the two-scale model in case of a fast reaction. ***To cite this article: S.A. Meier, A. Muntean, C. R. Mecanique 336 (2008).***

Résumé

Sur un système de réaction–diffusion avec une frontière libre de réaction pénétrant la micro-structure. On considère un modèle de réaction-diffusion à deux échelles, dont la micro-structure contient une réaction rapide. Lorsque la constante de réaction a explose vers l'infini, le modèle à deux échelles converge vers un modèle à frontière libre concentrée dans la micro-structure. Le but de cette Note est de montrer qu'en échangeant la limite d'homogénéisation $\varepsilon \to 0$ avec celle de la réaction rapide $a \to \infty$, on ne change pas le modèle limite. Des résultats numériques sont également presentés. ***Pour citer cet article : S.A. Meier, A. Muntean, C. R. Mecanique 336 (2008).***
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Keywords: Porous media; Two-scale model; Homogenisation; Fast reaction; Free-boundary problem

Mots-clés : Milieux poreux ; Modèle à deux échelles ; Homogénéisation ; Réaction rapide ; Frontière libre

* Corresponding author.
E-mail addresses: sebam@math.uni-bremen.de (S.A. Meier), a.muntean@tue.nl (A. Muntean).

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1. Introduction

We consider the following prototypical reaction–diffusion scenario: A gaseous species A penetrates a non-saturated porous medium via the air phase of its pore space and instantaneously dissolves in the pore water where A reacts very fast with a species B. The species B becomes available by a dissolution mechanism. See, for instance, [1–3] for fast-reaction–slow-diffusion settings playing an important role in pattern formation and corrosion of porous materials, and [4,5] for further conceptually related scenarios arising in the modelling of catalytic reactors and deformation in hydrophilic swelling porous media.

In this Note, we present a coupled two-scale reaction–diffusion system whose distributed micro-structure hosts the fast reaction of A and B. Making use of singular-limit analysis we derive a non-standard free-boundary problem as the fast-reaction limit $a \to \infty$ of the two-scale model. a stands for the corresponding reaction constant. The question that we answer here is: Is the *same* two-scale free-boundary problem obtained if we first pass to the fast-reaction limit $a \to \infty$ and then take the homogenisation limit $\varepsilon \to 0$ (that is behind the derivation of the two-scale model)? Using the method of formal homogenisation, we show that interchanging the limits $\varepsilon \to 0$ and $a \to \infty$ gives the same result.

2. The microscopic system (P_a^ε) on a thin strip

Let $L > 0$ and $R \in (0, 1)$ be given lengths and let $\varepsilon > 0$ be a small number. We consider a two-dimensional micro geometry as depicted in Fig. 1(a). We define the sets $\Omega_1^\varepsilon := (0, L) \times (\varepsilon R, \varepsilon)$, $\Omega_2^\varepsilon := (0, L) \times (0, \varepsilon R)$ and $\Gamma^\varepsilon := \partial\Omega_1^\varepsilon \cap \partial\Omega_2^\varepsilon$. Then the unit normal at Γ^ε pointing towards Ω_1^ε is $\nu^\varepsilon := (0, 1)^T$. Moreover, let $\Gamma^{\varepsilon,\text{ext}} := \{0\} \times (\varepsilon R, \varepsilon)$ and $\Gamma_1^{N\varepsilon} := \partial\Omega_1^\varepsilon \setminus (\Gamma^\varepsilon \cup \Gamma^{\varepsilon,\text{ext}})$ and $\Gamma_2^{N,\varepsilon} := \partial\Omega_2^\varepsilon \setminus \Gamma^\varepsilon$.

We denote by S_T the time interval $(0, T)$, where $T > 0$ is finite and arbitrarily fixed. The active concentrations are $C_1^{A\varepsilon}(x, t)$, $C_2^{A\varepsilon}(x, t)$, and $C^{B\varepsilon}(x, t)$. By the upper indices A and B, we indicate the two reactants. The lower index concerns either phase 1 or phase 2. The species A is present in both phases while the species B only appears in phase 2. We consider the following set of balance equations acting at the micro-scale:

$$\partial_t C_1^{A\varepsilon} - D_1^A \Delta C_1^{A\varepsilon} = 0, \quad x \in \Omega_1^\varepsilon,\ t \in S_T \tag{1}$$

$$\partial_t C_2^{A\varepsilon} - \varepsilon^2 D_2^A \Delta C_2^{A\varepsilon} = -a C_2^{A\varepsilon} C^{B\varepsilon}, \quad x \in \Omega_2^\varepsilon,\ t \in S_T \tag{2}$$

$$\partial_t C^{B\varepsilon} = -a C_2^{A\varepsilon} C^{B\varepsilon}, \quad x \in \Omega_2^\varepsilon,\ t \in S_T \tag{3}$$

with boundary conditions

$$D_1^A \partial_{x_2} C_1^{A\varepsilon} = \varepsilon^2 D_2^A \partial_{x_2} C_2^{A\varepsilon}, \quad x = (x_1, x_2) \in \Gamma^\varepsilon,\ t \in S_T \tag{4}$$

$$C_1^{A\varepsilon} = C_2^{A\varepsilon}, \quad x \in \Gamma^\varepsilon,\ t \in S_T \tag{5}$$

$$C_1^{A\varepsilon} = C_1^{A,\text{ext}}(t), \quad x \in \Gamma^{\varepsilon,\text{ext}},\ t \in S_T \tag{6}$$

$$D_i^A \nabla C_i^{A\varepsilon} \cdot \nu = 0, \quad x \in \Gamma_i^{N\varepsilon},\ t \in S_T, \quad i = 1, 2 \tag{7}$$

and initial conditions

$$C_1^{A\varepsilon}(0, x) = 0, \quad x \in \Omega_1^\varepsilon, \qquad C_2^{A\varepsilon}(0, x) = 0, \quad x \in \Omega_2^\varepsilon, \qquad C^{B\varepsilon}(0, x) = C_B^0, \quad x \in \Omega_2^\varepsilon \tag{8}$$

The system (1)–(8) is referred to as (P_a^ε). For typical fast-reaction–slow-transport scenarios, the parameter a is large and defines a high Thiele modulus.

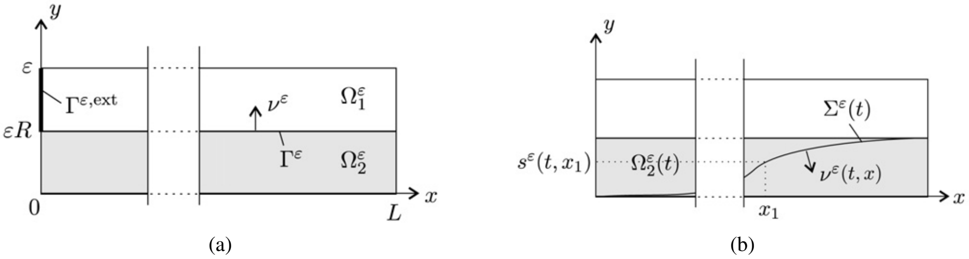
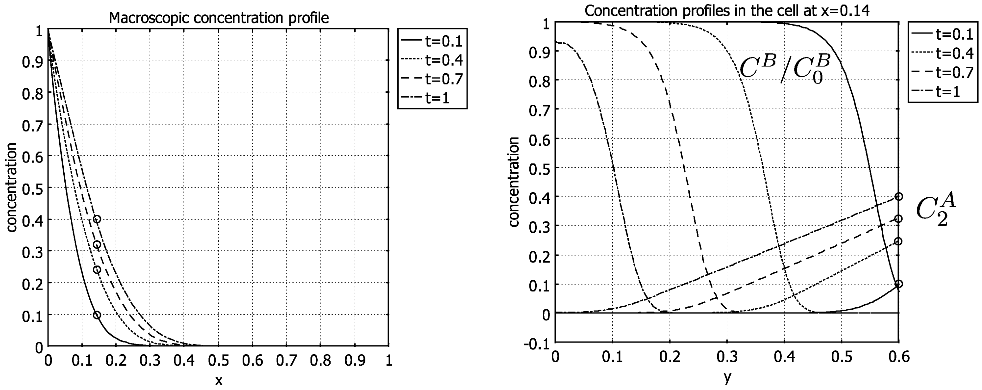


Fig. 1. (a) Geometry of the model (P_a^ε). (b) Possible position of the free boundary $\Sigma^\varepsilon(t)$ in the model (P_∞^ε).



(a) (b)

Fig. 2. Solutions of the two-scale model (P_a). (a) Profiles of C_1^A. (b) Profiles of C_2^A and C^B/C_0^B for frozen $x = 0.14$.

3. First $\varepsilon \to 0$, then $a \to \infty$

3.1. Homogenisation limit $\varepsilon \to 0$ of the micro problem (P_a^ε)

It is well-known that in the limit $\varepsilon \to 0$ of (P_a^ε), we obtain the two-scale model (9)–(16), which we call (P_a). We denote $\Omega := (0, L)$ and $Y := (0, R)$. The equations are

$$\partial_t C_1^A - D_1^A \partial_{xx} C_1^A = -(1-R)^{-1} D_2^A \partial_y C_2^A(t, x, R), \quad x \in \Omega,\ t \in S_T \tag{9}$$

$$\partial_t C_2^A - D_2^A \partial_{yy} C_2^A = -a C_2^A C^B, \quad x \in \Omega,\ y \in Y,\ t \in S_T \tag{10}$$

$$\partial_t C^B = -a C_2^A C^B, \quad x \in \Omega,\ y \in Y,\ t \in S_T \tag{11}$$

with initial conditions

$$C_1^A(0, x) = 0, \quad C_2^A(0, x, y) = 0, \quad C^B(0, x, y) = C_B^0, \quad x \in \Omega,\ y \in Y \tag{12}$$

a boundary condition connecting the *micro* scale with the *macro* one

$$C_2^A(t, x, R) = C_1^A(t, x), \quad x \in \Omega,\ t \in S_T \tag{13}$$

one boundary condition at the micro scale

$$\partial_y C_2^A(t, x, 0) = 0, \quad x \in \Omega,\ t \in S_T \tag{14}$$

as well as of boundary conditions for the species living at the *macro* scale

$$C_1^A(t, 0) = C_1^{A,\mathrm{ext}}(t) \tag{15}$$

$$\partial_x C_1^A(t, L) = 0, \quad t \in S_T \tag{16}$$

The species A is active on both scales, while the species B only appears on the micro-scale. Rigorous convergence proofs are given in [6] for a linear model and in [7] for a non-linear version. This set of mass-balance equations was motivated in [2] for a double-porosity modelling approach to a fast-reaction process taking place in concrete-based materials.

As an illustration, typical solution profiles of (P_a) are plotted in Fig. 2 for parameters $L = 1$, $R = 0.6$, $D_1^A = 0.5$, $D_2^A = 5$, $C_0^B = 10$ and $a = 500$. Diffusion of C_1^A takes place at the macro scale (Fig. 2(a)). In Fig. 2(b), the local profiles of C_2^A and C^B/C_0^B for frozen $x = 0.14$ are shown. It can be seen that the reaction between A and B concentrates on a narrow zone. We expect that the faster the reaction is, the narrower this reaction zone will be. The coupling condition (13) can be read off the two plots as follows: The boundary values of C_2^A at $y = 0.6$ in the right plot coincide with the values of C_1^A at $x = 0.14$ in the left plot.

3.2. Fast-reaction limit of the two-scale problem (P_a)

Next we address the following question: What happens in the limit $a \to \infty$ with problem (P_a)? Relying on arguments from [1,8,9], we deduce that in the limit the two reactants A and B on the micro scale are completely separated by a reaction front at position $y = s(x,t) \in [0, R]$. Rigorous convergence proofs will be presented elsewhere [10]. We denote by c_1^A, c_2^B and c^B the active concentrations arising in the following limit problem

$$\partial_t c_1^A - D_1^A \partial_{xx} c_1^A = -(1-R)^{-1} D_2^A \partial_y c_2^A(t,x,R), \quad x \in \Omega,\ t \in S_T \tag{17}$$

$$\partial_t c_2^A - D_2^A \partial_{yy} c_2^A = 0, \quad x \in \Omega,\ y \in \big(s(t), R\big),\ t \in S_T \tag{18}$$

$$c^B = c_0^B, \quad x \in \Omega,\ y \in \big(0, s(t)\big),\ t \in S_T, \tag{19}$$

initial conditions

$$c_1^A(0,x) = 0, \quad c_2^A(0,x,y) = 0, \quad y \in (s_0, R), \quad s(0,x) = s_0, \quad x \in \Omega \tag{20}$$

where $0 < s_0 \leqslant R$ is the initial position of the interface s, one boundary condition matching the micro with the macro scale,

$$c_2^A(t,x,R) = c_1^A(t,x), \quad x \in \Omega,\ t \in S_T \tag{21}$$

and boundary conditions for the species living at the *macro* scale

$$c_1^A(t,0) = c_1^{A,\mathrm{ext}}(t), \quad t \in S_T \tag{22}$$

$$\partial_x c_1^A(t,L) = 0, \quad t \in S_T \tag{23}$$

The free-boundary conditions are

$$c_2^A\big(t,x,s(t,x)\big) = 0, \tag{24}$$

$$s'(t,x) c_0^B = -D_2^A \partial_y c_2^A\big(t,x,s(t,x)\big), \quad x \in \Omega,\ t \in S_T \tag{25}$$

as long as $s(t,x) > 0$. Otherwise, we have

$$s'(t,x) = 0 \quad \text{and} \quad D_2^A \partial_y c_2^A\big(t,x,s(t,x)\big) = 0 \tag{26}$$

(25) is the classical Stefan condition with c_0^B as “latent heat”. Here $y = s(t,x)$ represents the position of the free boundary at time $t \in S_T$ in the micro-cell associated to the point $x \in \Omega$. The system (17)–(26) is the free-boundary problem resulting in the *fast-reaction limit* of the two-scale model (P_a). We refer to this problem as (P_∞).

4. First $a \to \infty$, then $\varepsilon \to 0$

4.1. Fast-reaction limit $a \to \infty$ of the micro problem (P_a^ε)

We have seen that if we consider first $\varepsilon \to 0$ and then $a \to \infty$, we obtain the free-boundary problem (17)–(25). Now our second question is: Do we obtain the same PDE system as homogenised singular limit if we permute the two limits? In other words, what happens if we first take $a \to \infty$ and then $\varepsilon \to 0$?

Similar as for the system (P_∞), in the singular limit of (P_a^ε) for $a \to \infty$ the two reactants are separated by a sharp interface $\Sigma^\varepsilon(t) \subset \Omega_2^\varepsilon$, which is evolving in time (cf. Fig. 1(b)). This interface hosts the instantaneous reaction and can be represented via

$$(x_1, x_2) \in \Sigma^\varepsilon(t) \iff x_2 = s^\varepsilon(t, x_1), \quad t \in S_T,\ x_1 \in \Omega.$$

We select $s^\varepsilon(0, x_1) = s_0 \in (0, R]$ and define $\Omega_2^\varepsilon(t) := \Omega \times (s^\varepsilon(t), \varepsilon R)$. By $w^\varepsilon(t,x)$, we denote the normal velocity of $\Sigma^\varepsilon(t)$ pointing outwards from $\Omega_2^\varepsilon(t)$. By passing to the limit $a \to \infty$ in (1)–(8), we obtain the one-phase Stefan problem (27)–(34) denoted by (P_∞^ε), viz.

$$\partial_t c_1^{A\varepsilon} - D_1^A \Delta c_1^{A\varepsilon} = 0, \quad x \in \Omega_1^\varepsilon,\ t \in S_T \tag{27}$$

$$\partial_t c_2^{A\varepsilon} - \varepsilon^2 D_2^A \Delta c_2^{A\varepsilon} = 0, \quad x \in \Omega_2^\varepsilon(t),\ t \in S_T \tag{28}$$

with boundary conditions

$$D_1^A \partial_{x_2} c_1^{A\varepsilon} = \varepsilon^2 D_2^A \partial_{x_2} c_2^{A\varepsilon}, \quad x = (x_1, x_2) \in \Gamma^\varepsilon, \ t \in S_T \tag{29}$$

$$c_1^{A\varepsilon} = c_2^{A\varepsilon}, \quad x \in \Gamma^\varepsilon, \ t \in S_T \tag{30}$$

$$c_1^{A\varepsilon} = c_1^{A,\mathrm{ext}}(t), \quad x \in \Gamma^{\varepsilon,\mathrm{ext}}, \ t \in S_T \tag{31}$$

$$D_1^A \nabla c_1^{A\varepsilon} \cdot \nu = 0, \quad x \in \Gamma_1^{N\varepsilon}, \ t \in S_T \tag{32}$$

and initial conditions

$$c_1^{A\varepsilon}(0,x) = 0, \quad x \in \Omega_1^\varepsilon, \qquad c_2^{A\varepsilon}(0,x) = 0, \quad x \in \Omega_2^\varepsilon(0), \qquad s^\varepsilon(0,x_1) = s_0, \quad x_1 \in \Omega \tag{33}$$

The limit problem is closed by the interface conditions

$$w^\varepsilon(t,x) c_0^B = -\varepsilon^2 D_2^A \nabla c_2^{A\varepsilon}(t,x) \cdot \nu^\varepsilon(t,x), \quad c_2^{A\varepsilon}(t,x) = 0, \quad t \in S_T, \ x \in \Sigma^\varepsilon(t) \tag{34}$$

as long as $s(t,x) > 0$, and otherwise

$$w^\varepsilon(t,x) = 0 \quad \text{and} \quad -\varepsilon^2 D_2^A \nabla c_2^{A\varepsilon}(t,x) \cdot \nu^\varepsilon(t,x) = 0, \quad t \in S_T, \ x \in \Sigma^\varepsilon(t) \tag{35}$$

4.2. Homogenisation limit $\varepsilon \to 0$ of the free-boundary problem (P_∞^ε)

We sketch now the homogenisation procedure by asymptotic expansions; see [11,12], e.g. analogous technique have also been applied in [13,14] for similar models with varying microstructure. We assume that the functions $c_1^{A\varepsilon}$ and $c_2^{A\varepsilon}$ admit the asymptotic representations

$$c_1^{A\varepsilon}(t,x) = c_1^{A0}\left(t, x_1, \frac{x_2}{\varepsilon}\right) + \varepsilon c_1^{A1}\left(t, x_1, \frac{x_2}{\varepsilon}\right) + \cdots, \quad x_1 \in \Omega, \ x_2 \in (R\varepsilon, \varepsilon)$$

$$c_2^{A\varepsilon}(t,x) = c_2^{A0}\left(t, x_1, \frac{x_2}{\varepsilon}\right) + \varepsilon c_2^{A1}\left(t, x_1, \frac{x_2}{\varepsilon}\right) + \cdots, \quad x_1 \in \Omega, \ x_2 \in (0, R\varepsilon)$$

valid for small ε, where all coefficients are Y-periodic functions w.r.t. their third coordinate. Moreover, we rescale

$$s^\varepsilon(t,x_1) =: \varepsilon s^0(t,x_1), \quad t \in S_T, \ x_1 \in \Omega \tag{36}$$

By (36), we obtain the important geometrical relations

$$\nu^\varepsilon(t,x) \approx \begin{pmatrix} 0 \\ -1 \end{pmatrix} + \mathcal{O}(\varepsilon) \tag{37}$$

$$w^\varepsilon(t,x) \approx -\varepsilon \partial_t s^0(t,x_1) + \mathcal{O}(\varepsilon^2), \quad t \in S_T, \ x \in \Sigma^\varepsilon(t) \tag{38}$$

The limit equations are calculated by plugging into (27)–(34) the asymptotic expansions introduced above. The model (P_∞) is obtained by collecting the zeroth order (in ε) terms.

Remark. Note that the asymptotic expansion are only formally correct. Convergence of the homogenisation process may be proven by transformation to a fixed geometry; see [15,16].

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