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# Multiscale Analysis of Ionic Transport in Periodic Charged Media

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***Abstract*—A macroscopic model for describing the ion transport in periodic charged porous media is rigorously derived. Our results can serve as a tool for biophysicists to analyze the ion transport through protein channels. Also, such a model is useful for describing the flow of electrons and holes in a semiconductor device.**

***Keywords*-homogenization; ion transport; the periodic unfolding method.**

## I. Introduction and setting of the problem

The goal of this paper is to obtain the effective behavior of a system of coupled partial differential equations appearing in the modeling of transport phenomena in periodic charged porous media or in the modeling of the flow of holes and electrons in semiconductors. Such a system is known in the literature as the *Poisson-Nernst-Planck system*, for the case of ion flow through membrane channels, or as *the van Roosbroeck model*, for the case of the transport of holes and electrons in semiconductors. For the physical aspects behind these models and for a review of the recent literature, we refer to [8], [13], [14], [15], [16] and [19].

Let us briefly describe the geometry of the problem. We assume, as it is customary in the literature, that the porous medium has a periodic microstructure. More precisely, for $n \geq 2$, we consider a smooth bounded connected open set $\Omega$ in $\mathbb{R}^n$, with $\mid \partial\Omega \mid = 0$. In what follows, we shall only consider the natural cases $n = 2$ or $n = 3$. We assume that the unit cell $Y = (0,1)^n$ consists of two smooth parts, the fluid part $Y_f$ and, respectively, the solid part $Y_s$, which are supposed to be open, nonempty and disjoint sets such that

$$Y = Y_f \cup \overline{Y_s}$$

and

$$\overline{Y_f} \cap \overline{Y_s} = \Gamma.$$

We suppose that the solid part has a Lipschitz boundary and does not intersect the boundary of the basic cell $Y$. Therefore, the fluid zone is connected.

Let $\varepsilon < 1$ be a real parameter taking values in a sequence of positive numbers converging to zero. For each $\mathbf{k} \in \mathbb{Z}^n$, let

$$Y_s^k = \mathbf{k} + Y_s$$

and

$$K_\varepsilon = \{\mathbf{k} \in \mathbb{Z}^n \mid \varepsilon\overline{Y_s^k} \subseteq \Omega\}.$$

We denote by

$$\Omega_\varepsilon^s = \bigcup_{\mathbf{k}\in K_\varepsilon} \varepsilon Y_s^k$$

the solid part, by

$$\Omega_\varepsilon = \Omega \setminus \overline{\Omega_\varepsilon^s}$$

the fluid part and we set

$$\theta = \left|Y \setminus \overline{Y_s}\right|.$$

It is easy to see that if the solid part is not allowed to reach the fixed exterior boundary of the domain $\Omega$, the intersection between the outer boundary $\partial\Omega$ and the

interior boundary $\Gamma_\varepsilon$ of the porous medium is empty, i.e.:

$$\Gamma_\varepsilon \cap \partial\Omega = \emptyset.$$

In such a periodic porous medium, we shall consider, at the microscale, the Poisson-Nernst-Planck system, with suitable boundary and initial conditions. The Nernst-Planck equations, describing the diffusion in the fluid phase, are coupled with the Poisson equation characterizing the electrical field which can influence the ionic transfer. More precisely, if we denote by $[0,T]$, with $T > 0$, the time interval of interest, we shall be interested in analyzing the asymptotic behavior, as $\varepsilon \to 0$, of the solution of the following system:

$$\begin{cases} -\Delta\,\Phi_\varepsilon = c_\varepsilon^+ - c_\varepsilon^- + D \quad \text{in } (0,T)\times\Omega_\varepsilon, \\ -\nabla\,\Phi_\varepsilon\cdot\nu = \varepsilon\,\sigma^\varepsilon G(\Phi_\varepsilon) \quad \text{on } (0,T)\times\Gamma_\varepsilon, \\ \nabla\,\Phi_\varepsilon\cdot\nu = 0 \quad \text{on } (0,T)\times\partial\Omega, \\ \partial_t c_\varepsilon^\pm - \nabla\cdot(\nabla c_\varepsilon^\pm \pm c_\varepsilon^\pm\,\nabla\,\Phi_\varepsilon) = F^\pm(c_\varepsilon^+, c_\varepsilon^-) \\ \qquad\qquad \text{in } (0,T)\times\Omega_\varepsilon, \\ (\nabla c_\varepsilon^\pm \pm c_\varepsilon^\pm\,\nabla\,\Phi_\varepsilon)\cdot\nu = 0 \quad \text{on } (0,T)\times\Gamma_\varepsilon, \\ (\nabla c_\varepsilon^\pm \pm c_\varepsilon^\pm\,\nabla\,\Phi_\varepsilon)\cdot\nu = 0 \quad \text{on } (0,T)\times\partial\Omega, \\ c_\varepsilon^\pm(0,x) = c_0^\pm(x) \quad \text{in } \Omega_\varepsilon. \end{cases} \tag{1}$$

Here, $\nu$ is the unit outward normal to $\Omega_\varepsilon$, $\Phi_\varepsilon$ represents the electrostatic potential, $c_\varepsilon^\pm$ are the concentrations of the ions (or the density of electrons and holes in the particular case of van Roosbroeck model), $D \in L^\infty(\Omega)$ is the given doping profile, $F^\pm$ is a reaction term and $G$ is a nonlinear function which takes into account the effect of the electrical double layer phenomenon arising at the interface $\Gamma^\varepsilon$.

We assume that

$$\sigma^\varepsilon = \sigma\left(\frac{x}{\varepsilon}\right),$$

with $\sigma(y)$ being a $Y$-periodic, bounded, smooth real function such that $\sigma(y) \geq \delta > 0$, and $G$ is a continuously differentiable function, monotonously increasing and such that $G(0) = 0$. Also, we shall suppose that there exist $C \geq 0$ and $r$, with $0 \leq r \leq n/(n-2)$ for $n = 3$ and $0 \leq r < \infty$ for $n = 2$, such that

$$\mid G'(s) \mid \leq C(1+\mid s \mid^{r-1}),\ \forall s \in \mathbb{R}.$$

Let us notice that this hypothesis concerning the smoothness of the nonlinearity $G$ can be relaxed by using a regularization technique, such as Yosida approximation (see [18]). Also, the results of this paper can be obtained, under our assumptions, without imposing any growth condition (see [17]).

In practical applications, based on the Gouy-Chapman theory, one can use the Grahame equation (see [2], [8] and [9]) in which

$$G(s) = K_1 \sinh(K_2 s), \quad K_1, K_2 > 0.$$

For the case of lower potentials, $\sinh(x)$ can be expanded in a power series of the form

$$\sinh(x) = x + \frac{x^3}{3!} + ...$$

and one can use the approximations $\sinh x \approx x$ or $\sinh x \approx x + x^3/3!$.

Concerning the reaction terms, we deal, as in [13], with the linear case in which

$$F^\pm(c_\varepsilon^+, c_\varepsilon^-) = \mp(c_\varepsilon^+ - c_\varepsilon^-),$$

but we can also address by our techniques the more general case in which

$$F^\pm(c_\varepsilon^+, c_\varepsilon^-) = \mp(a^\varepsilon c_\varepsilon^+ - b^\varepsilon c_\varepsilon^-),$$

with

$$a^\varepsilon(x) = a\left(\frac{x}{\varepsilon}\right), \quad b^\varepsilon(x) = b\left(\frac{x}{\varepsilon}\right),$$

where $a(y)$ and $b(y)$ are $Y$-periodic, bounded, smooth real functions such that $a(y) \geq a_0 > 0$, $b(y) \geq b_0 > 0$.

For the case of other nonlinear reaction rates $F^\pm$ and more general functions $G$, see [6], [10], [11] and [18].

We assume that the initial data are non-negative and bounded independently of $\varepsilon$ and

$$\int_{\Omega_\varepsilon} (c_0^+ - c_0^- + D)dx = \varepsilon \int_{\Gamma_\varepsilon} \sigma^\varepsilon G(\Phi_\varepsilon)ds. \tag{2}$$

We also assume that the potential $\Phi_\varepsilon$ has zero mean value in $\Omega_\varepsilon$.

Let us mention that, for simplifying the notation, we have suppressed in system (1) some constant physical relevant parameters.

We consider here only two oppositely charged species, i.e. positively and negatively charged particles, with concentrations $c_\varepsilon^\pm$, but all the results can be generalized for the case of $N$ species.

For the case in which we consider different scalings in (1), see [13] and [18]. Also, let us remark that we can treat the case in which the electrostatic potential is defined all over the domain $\Omega$, with suitable transmission conditions at the interface $\Gamma^\varepsilon$ (see, for instance, [8] or [16]).

From the Nernst-Planck equation, it is easy to see that the total mass

$$M = \int_{\Omega_\varepsilon} (c_\varepsilon^+ + c_\varepsilon^-) dx$$

is conserved and suitable physical equilibrium conditions hold true, at the microscale and, also, at the macroscale (see, for details, [8] and [18]).

Using similar arguments as in [8] or [13], we can prove the well posedness of problem (1) in suitable function spaces and we can obtain proper energy estimates.

The high complexity of the geometry and of the governing equations implies that an asymptotic procedure becomes necessary for describing the solution of such a problem.

Using the periodic unfolding method recently introduced by D. Cioranescu, A. Damlamian, G. Griso, P. Donato and R. Zaki (see [3], [5] and [4]), we can prove that the asymptotic behavior of the solution of our problem is governed by a new coupled system of equations (see (3)-(5)). In particular, the evolution of the macroscopic electrostatic potential is governed by a new law, similar to Grahame's law (see [8] and [9]).

An advantage of this approach is that we can avoid the use of extension operators and, therefore, we can deal in a rigorous manner with media which are less regular than those usually considered in the literature (composite materials and biological tissues are highly heterogeneous media with not very smooth interfaces, in general).

Similar problems have been considered, using different techniques, in [8], [13] or [15]. As already mentioned, our approach is based on a new method, i.e. the periodic unfolding method, which allows us to consider very general heterogeneous media. Another novelty brought by our paper consists in dealing with a general nonlinear boundary condition for the electrostatic potential and with more general reaction terms.

The rest of the paper is organized as follows: in Section 2, we formulate our main convergence result, while Section 3 is devoted to the proof of this result. The paper ends with some conclusions and a few references.

## II. The main result

Using the periodic unfolding method, we are allowed to pass to the limit in the weak formulation of problem (1) and to obtain the effective behavior of the solution of our microscopic model.

**Theorem 1.** The solution $(\Phi_\varepsilon,\, c_\varepsilon^+, c_\varepsilon^-)$ of system (1) converges, as $\varepsilon \to 0$, to the unique solution $(\Phi,\, c^+, c^-)$ of the following macroscopic problem in $(0,T)\times\Omega$:

$$\begin{cases} -\text{div}\,(D^0\,\nabla\,\Phi) + \dfrac{1}{|\,Y_f\,|}\,\sigma_0 G = c^+ - c^- + D, \\ \dfrac{\partial\, c^\pm}{\partial\, t} - \text{div}(D^0\,\nabla c^\pm \pm D^0\, c^\pm\,\nabla\,\Phi) = F_0^\pm, \end{cases} \tag{3}$$

with the boundary conditions on $(0,T)\times\partial\Omega$:

$$\begin{cases} D^0\,\nabla\Phi\cdot\nu = 0, \\ (D^0\,\nabla\, c^\pm \pm D^0\, c^\pm\,\nabla\,\Phi)\cdot\nu = 0 \end{cases} \tag{4}$$

and the initial conditions

$$c^\pm(0,x) = c_0^\pm(x),\ \forall x\in\Omega. \tag{5}$$

Here,

$$\sigma_0 = \int_\Gamma \sigma(y) ds,$$

$$F_0^\pm(c^+,c^-) = \mp(c^+ - c^-)$$

and $D^0 = (d_{ij}^0)$ is the homogenized matrix, defined as follows:

$$d_{ij}^0 = \frac{1}{|Y_f|}\int_{Y_f}\left(\delta_{ij} + \frac{\partial\chi_j}{\partial y_i}(y)\right) dy\,,$$

in terms of the functions $\chi_j$, $j=1,...,n$, solutions of the cell problems

$$\begin{cases} \chi_j \in H^1_{per}(Y_f)\,,\ \displaystyle\int_{Y_f}\chi_j = 0, \\ -\Delta\,\chi_j = 0 \ \text{ in } \ Y_f, \\ (\nabla\chi_j + \mathbf{e}_j)\cdot\nu = 0 \ \text{ on } \ \Gamma, \end{cases} \tag{6}$$

where $\mathbf{e}_i$, $1\le i\le n$, are the vectors of the canonical basis in $\mathbb{R}^n$. ∎

## III. Proof of the main result

We shall only sketch the proof of our main convergence result. For details, we refer to [18].

Let us consider now the equivalent variational formulation of problem (1):

Find $(\Phi_\varepsilon,\, c_\varepsilon^+,\, c_\varepsilon^-)$, with

$$\begin{cases} \Phi_\varepsilon \in L^\infty(0,T;H^1(\Omega_\varepsilon)), \\ c_\varepsilon^\pm \in L^\infty(0,T;L^2(\Omega_\varepsilon)) \cap L^2(0,T;H^1(\Omega_\varepsilon)), \\ \partial_t c_\varepsilon^\pm \in L^2(0,T;(H^1(\Omega_\varepsilon))') \end{cases} \tag{7}$$

such that, for any $t>0$ and for any $\varphi_1,\,\varphi_2\in H^1(\Omega_\varepsilon)$, $(\Phi_\varepsilon,\, c_\varepsilon^+,\, c_\varepsilon^-)$ satisfy:

$$\int_{\Omega_\varepsilon}\nabla\,\Phi_\varepsilon\cdot\nabla\,\varphi_1\,dx - \int_{\Gamma_\varepsilon}\nabla\,\Phi_\varepsilon\cdot\nu\varphi_1\,d\sigma =$$

$$\int_{\Omega_\varepsilon} (c_\varepsilon^+ - c_\varepsilon^- + D)\,\varphi_1\,dx, \tag{8}$$

$$\langle \partial_t c_\varepsilon^\pm,\, \varphi_2\rangle_{(H^1)',H^1} + \int_{\Omega_\varepsilon} (\nabla\, c_\varepsilon^\pm \pm c_\varepsilon^\pm\, \nabla\, \Phi_\varepsilon)\cdot \nabla\, \varphi_2 dx =$$

$$\int_{\Omega_\varepsilon} F^\pm(c_\varepsilon^+, c_\varepsilon^-)\varphi_2\,dx \tag{9}$$

and

$$c_\varepsilon^\pm(0,x) = c_0^\pm(x) \quad \text{in } \Omega_\varepsilon. \tag{10}$$

There exists a unique weak solution $(\Phi_\varepsilon,\, c_\varepsilon^+,\, c_\varepsilon^-)$ of problem (8)-(10) (see [8], [13] or [18]).

Moreover, exactly like in [13], we can prove that the concentration fields are non-negative, i.e. are bounded from below uniformly in $\varepsilon$. Also, the concentration fields are bounded from above uniformly in $\varepsilon$.

Under the above hypotheses, by standard techniques, we can show that there exists a constant $C \in \mathbb{R}_+$, independent of $\varepsilon$, such that the following *a priori* estimates hold true:

$$\|\Phi_\varepsilon\|_{L^2((0,T)\times\Omega_\varepsilon)} + \|\nabla\Phi_\varepsilon\|_{L^2((0,T)\times\Omega_\varepsilon)} \le C$$

$$\max_{0\le t\le T} \|c_\varepsilon^-\|_{L^2(\Omega_\varepsilon)} + \max_{0\le t\le T} \|c_\varepsilon^+\|_{L^2(\Omega_\varepsilon)} +$$

$$\|\nabla c_\varepsilon^-\|_{L^2((0,T)\times\Omega_\varepsilon)} + \|\nabla c_\varepsilon^+\|_{L^2((0,T)\times\Omega_\varepsilon)} +$$

$$\|\partial_t c_\varepsilon^-\|_{L^2(0,T;(H^1(\Omega_\varepsilon))')} + \|\partial_t c_\varepsilon^+\|_{L^2(0,T;(H^1(\Omega_\varepsilon))')} \le C.$$

As already mentioned, we are interested in obtaining the limit behavior, as $\varepsilon \to 0$, of the solution $(\Phi_\varepsilon,\, c_\varepsilon^+,\, c_\varepsilon^-)$ of problem (8)-(10). Our approach is based on the periodic unfolding method introduced by D. Cioranescu, A. Damlamian, G. Griso, P. Donato and R. Zaki (see [3] and [5]). This approach has the advantage that we do not need to use extension operators like in [8] or [13].

Using the properties of the unfolding operator $\mathcal{T}_\varepsilon$ introduced in [3] and [5] and the above *a priori* estimates, we can easily prove that there exist $\Phi \in L^2(0,T;H^1(\Omega))$, $\widehat{\Phi} \in L^2((0,T)\times\Omega; H^1_{per}(Y_f))$, $c^\pm \in L^2(0,T;H^1(\Omega))$, $\widehat{c}^\pm \in L^2((0,T)\times\Omega; H^1_{per}(Y_f))$, such that, up to a subsequence,

$$\mathcal{T}_\varepsilon(\Phi_\varepsilon) \rightharpoonup \Phi \text{ weakly in } L^2((0,T)\times\Omega; H^1(Y_f)),$$

$$\mathcal{T}_\varepsilon(\nabla\Phi_\varepsilon) \rightharpoonup \nabla\Phi + \nabla_y\widehat{\Phi} \text{ weakly in } L^2((0,T)\times\Omega\times Y_f),$$

$$\mathcal{T}_\varepsilon(c_\varepsilon^\pm) \to c^\pm \text{ strongly in } L^2((0,T)\times\Omega; H^1(Y_f)),$$

$$\mathcal{T}_\varepsilon(\nabla c_\varepsilon^\pm) \rightharpoonup \nabla c^\pm + \nabla_y\widehat{c}^\pm \text{ weakly in } L^2((0,T)\times\Omega\times Y_f).$$

For proving Theorem 1, let us take, first, in the Poisson equation (8), the test function

$$\varphi_1(t,x) = \psi_0(t,x) + \varepsilon\psi_1(t,x,\frac{x}{\varepsilon}),$$

with

$$\psi_0 \in \mathcal{D}((0,T); C^\infty(\overline{\Omega}))$$

and

$$\psi_1 \in \mathcal{D}((0,T)\times\Omega; H^1_{per}(Y_f)).$$

Unfolding each term by using the operator $\mathcal{T}_\varepsilon$ and passing to the limit with $\varepsilon \to 0$, we obtain (see, for details, [18]):

$$\int_0^T \int_{\Omega\times Y_f} (\nabla\Phi(t,x) +$$

$$\nabla_y\widehat{\Phi}(t,x,y))\,(\nabla\psi_0(t,x) + \nabla_y\psi_1(t,x,y))\,dx\,dy\,dt +$$

$$\sigma_0 \int_0^T \int_\Omega G\psi_0(t,x)\,dx\,dt =$$

$$\int_0^T \int_{\Omega\times Y_f} (c^+(t,x) - c^-(t,x) +$$

$$D(x))\psi_0(t,x)dx\,dy\,dt. \tag{11}$$

Then, by density, it follows that (11) holds true for any $\psi_0 \in L^2(0,T;H^1(\Omega))$ and $\psi_1 \in L^2((0,T)\times\Omega; H^1_{per}(Y_f))$.

Taking $\psi_0(t,x) = 0$, we obtain

$$\begin{cases} -\Delta_y\,\widehat{\Phi}(t,x,y) = 0 \quad \text{in } (0,T)\times\Omega\times Y_f, \\ \nabla_y\,\widehat{\Phi}\cdot\nu = -\nabla_x\,\Phi(t,x)\cdot\nu \quad \text{on } (0,T)\times\Omega\times\Gamma, \\ \widehat{\Phi}(t,x,y) \quad \text{periodic in } y. \end{cases}$$

By linearity, we get

$$\widehat{\Phi}(t,\,x,\,y) = \sum_{j=1}^n \chi_j(y)\frac{\partial\Phi}{\partial x_j}(t,\,x), \tag{12}$$

where $\chi_j$, $j = \overline{1,n}$, are the solutions of the cell problems (6).

Taking $\psi_1(t,x,y) = 0$, integrating with respect to $x$ and using (12), we easily get the macroscopic problem for the electrostatic potential $\Phi$.

Now, taking in the Nernst-Planck equation (9) the test function

$$\varphi_2(t,x) = \psi_0(t,x) + \varepsilon\psi_1(t,x,\frac{x}{\varepsilon}),$$

with

$$\psi_0 \in \mathcal{D}((0,T); C^\infty(\overline{\Omega}))$$

and

$$\psi_1 \in \mathcal{D}((0,T)\times\Omega; H^1_{per}(Y_f)),$$

unfolding each term by using the operator $\mathcal{T}_\varepsilon$ and passing to the limit with $\varepsilon \to 0$, we get (see [18])

$$-\int_0^T \int_{\Omega\times Y_f} (c^\pm(t,x))\,\partial_t\psi_0(t,x)\,dx\,dy\,dt+$$

$$\int_0^T \int_{\Omega\times Y_f} (\nabla\, c^\pm(t,x) + \nabla_y \widehat{c^\pm}(t,x,y))(\nabla_x\psi_0(t,x)+$$

$$\nabla_y\psi_1(t,x,y))\,dx\,dy\,dt =$$

$$\int_0^T \int_{\Omega\times Y_f} F_0^\pm(c^+,c^-)\psi_0(t,x)\,dx\,dy\,dt. \qquad (13)$$

By standard density arguments, we see that (13) holds true for any $\psi_0 \in L^2(0,T;H^1(\Omega))$ and $\psi_1 \in L^2((0,T)\times\Omega; H^1_{per}(Y_f))$.

Taking, first, $\psi_0(t,x) = 0$, and, then, $\psi_1(t,x,y) = 0$, we obtain exactly the macroscopic problem for the concentrations $c^\pm$.

Since $\Phi$ and $c^\pm$ are uniquely determined (see [13] and [18]), the whole sequences of microscopic solutions converge to a solution of the unfolded limit problem and this completes the proof of Theorem 1. ∎

## IV. Conclusion

Using the periodic unfolding method, the macroscopic behavior of the solution of a system of equations describing the ion transport in periodic charged media is analyzed.

Our model is relevant for studying the ion transport through protein channels or the flow of electrons and holes in a semiconductor device.

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