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The existence of solutions to a corrosion model

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ABSTRACT

In this work, we consider a corrosion model of iron based alloy in a nuclear waste repository. It consists of a PDE system, similar to the steady-state drift–diffusion system arising in semiconductor modelling. The main difference lies in the boundary conditions, since they are Robin boundary conditions and imply an additional coupling between the equations. Using a priori estimates for the solution and Schauder's fixed point theorem, we show the existence of solutions to the corrosion model.

1. Introduction

1.1. Presentation of the model

In this work, we consider a system of partial differential equations arising in corrosion modelling. This model, presented in [1] and called the Diffusion Poisson Coupled Model, is a model of iron based alloy in a nuclear waste repository. It assumes that the metal (for instance that of a nuclear waste canister in a geological repository) is covered by an oxide layer which is in contact with a solution. In most cases, the thickness of the oxide layer ranges from nanometers to micrometers. This thickness is always much smaller than the sizes of the metal and of the solution. Therefore, a 1D modelling is sufficient to describe the system.

The oxide layer is thought of as a semiconductor: charge carriers are convected by the electric field and the electric potential is coupled to the charge densities through a Poisson equation. Moreover, the oxide layer is in contact on one side with the metal and on the other side with a solution. Charge carriers are created and consumed at both interfaces. The kinetics of the electrochemical reactions at both interfaces provides boundary conditions.

We consider here the case where only two charge carriers are taken into account: electrons and cations, Fe^{3+}. We assume that there is no evolution of the layer thickness and we consider a steady-state model. The unknowns of the problem are the density of electrons N, the density of cations P and the electric potential Ψ (there are dimensionless variables). The system is written as:

– Equation and boundary conditions for Ψ:

$$-\lambda^2 \partial^2_{xx}\Psi = -N + 3P + \rho_{hl} \quad \text{on } (0, 1), \tag{1}$$

$$\Psi - \alpha_0 \partial_x \Psi = \Delta\Psi_0^{pzc} \quad \text{at } x = 0, \tag{2}$$

$$\Psi + \alpha_1 \partial_x \Psi = V - \Delta\Psi_1^{pzc} \quad \text{at } x = 1. \tag{3}$$

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– Equation and boundary conditions for P:

$$\partial_x J_P = 0, \quad J_P = -\partial_x P - 3P\partial_x \Psi \quad \text{on } (0,1), \tag{4}$$

$$J_P = m_1^0(P_m - P)\exp(-3b_1^0\Psi) - k_1^0 P\exp(3a_1^0\Psi) \quad \text{at } x = 0, \tag{5}$$

$$J_P = m_1^L P\exp(-3b_1^L(V-\Psi)) - k_1^L(P_m - P)\exp(3a_1^L(V-\Psi)) \quad \text{at } x = 1. \tag{6}$$

– Equation and boundary conditions for N:

$$\partial_x J_N = 0, \quad J_N = -\partial_x N + N\partial_x \Psi \quad \text{on } (0,1), \tag{7}$$

$$J_N = m_2^0(N_m - N)\exp(b_2^0\Psi) - k_2^0 N\exp(-a_2^0\Psi) \quad \text{at } x = 0, \tag{8}$$

$$J_N = m_2^L N\exp(b_2^L(V-\Psi)) - k_2^L(N_m - N)\exp(-a_2^L(V-\Psi)) \quad \text{at } x = 1. \tag{9}$$

The system of partial differential equations (1), (4), (7) is the steady-state drift–diffusion system, well-known in semiconductor modelling (see [2,3]). But, while the boundary conditions in semiconductor modelling are in general mixed Dirichlet–Neumann boundary conditions (with ohmic contacts or insulating boundary segments), we have here Robin boundary conditions. They come from the kinetics of the electrochemical reactions at each interface, which are assumed to follow Butler–Volmer laws (see [4]).

The parameters arising in the set of expressions Eqs. (1)–(9) satisfy the following hypotheses, denoted by $(\mathcal{H})$:

- $(k_i^0, k_i^L, m_i^0, m_i^L)_{i=1,2}$ are interface kinetic functions. In what follows, we assume that these functions are constant and verify: $k_i^0, k_i^L > 0$ and $m_i^0, m_i^L \geq 0$ for $i = 1, 2$.
- $(a_i^0, a_i^L, b_i^0, b_i^L)_{i=1,2}$ are positive transfer coefficients, which satisfy $a_i^0 + b_i^0 = 1$ and $a_i^L + b_i^L = 1$ for $i = 1, 2$.
- P_m is the maximum occupancy for octahedral cations in the host lattice, N_m is the density of states in the conduction band. They are positive constants (in the applications, $P_m = 2$ and $N_m = 1$).
- ρ_{hl} is the net charge density of the ionic species in the host lattice. In what follows, we assume that ρ_{hl} is constant throughout the whole layer, with $\rho_{hl} = -5$.
- $\Delta\Psi_0^{pzc}, \Delta\Psi_1^{pzc}$ are respectively the outer and the inner *pzc* voltages, and V is the applied voltage. Let us set $U_0 = \Delta\Psi_0^{pzc}$ and $U_1 = V - \Delta\Psi_1^{pzc}$.
- $\lambda^2, \alpha_0, \alpha_1$ are positive dimensionless parameters arising from the scaling of the model.

1.2. Changing the variables

In order to eliminate the convection terms in (4) and (7), we propose the following change of variables, which is classical in the study of the drift–diffusion system (see [2]):

$$P = e^{-3\Psi}u, \qquad N = e^{\Psi}v.$$

Variables u and v are called Slotboom variables. The system then becomes:

– Equation and boundary conditions for u:

$$\partial_x(e^{-3\Psi}\partial_x u) = 0 \quad \text{on } (0,1), \tag{10}$$

$$\partial_x u - \left(m_1^0 e^{-3b_1^0\Psi} + k_1^0 e^{3a_1^0\Psi}\right)u = -P_m m_1^0 e^{-3b_1^0\Psi}e^{3\Psi} \quad \text{at } x = 0, \tag{11}$$

$$\partial_x u + \left(m_1^L e^{-3b_1^L(V-\Psi)} + k_1^L e^{3a_1^L(V-\Psi)}\right)u = P_m k_1^L e^{3a_1^L(V-\Psi)}e^{3\Psi} \quad \text{at } x = 1. \tag{12}$$

– Equation and boundary conditions for v:

$$\partial_x(e^{\Psi}\partial_x v) = 0 \quad \text{on } (0,1), \tag{13}$$

$$\partial_x v - \left(m_2^0 e^{b_2^0\Psi} + k_2^0 e^{-a_2^0\Psi}\right)v = -N_m m_2^0 e^{b_2^0\Psi}e^{-\Psi} \quad \text{at } x = 0, \tag{14}$$

$$\partial_x v + \left(m_2^L e^{b_2^L(V-\Psi)} + k_2^L e^{-a_2^L(V-\Psi)}\right)v = N_m k_2^L e^{-a_2^L(V-\Psi)}e^{-\Psi} \quad \text{at } x = 1. \tag{15}$$

– Equation and boundary conditions for Ψ:

$$-\lambda^2\partial_{xx}^2\Psi = -e^{\Psi}v + 3e^{-3\Psi}u - 5 \quad \text{on } (0,1), \tag{16}$$

$$\Psi - \alpha_0\partial_x\Psi = U_0 \quad \text{at } x = 0, \tag{17}$$

$$\Psi + \alpha_1\partial_x\Psi = U_1 \quad \text{at } x = 1. \tag{18}$$

1.3. The main result

The goal of this work is to establish the following result:

Theorem 1. *Let the assumptions ($\mathcal{H}$) hold. Then, the problem (10)–(18) has a weak solution (u, v, Ψ), with $(u, v, \Psi) \in \mathcal{C}([0,1]) \times \mathcal{C}([0,1]) \times H^1(0,1)$, which satisfies the following L^∞-estimates:*

$$0 \le u(x) \le \max\left(\frac{k_1^L}{m_1^L} e^{3V}, \frac{m_1^0}{k_1^0}\right) P_m, \qquad 0 \le v(x) \le \max\left(\frac{k_2^L}{m_2^L} e^{-V}, \frac{m_2^0}{k_2^0}\right) N_m, \quad \forall x \in [0,1]. \tag{19}$$

The proof of this result is based on decoupling the equations for u, v and Ψ and using the Schauder fixed point theorem.

2. A priori estimates

This section is devoted to the proof of a priori estimates which will allow the use of a fixed point theorem. We first show that, for a given Ψ, the solutions u and v of (10)–(15) can be computed explicitly, and we deduce L^∞-estimates for u and v independent of Ψ. Then assuming that u and v are known, we prove that there exists a unique solution Ψ to the nonlinear elliptic problem (16)–(18) and we obtain estimates for Ψ.

2.1. Calculus and estimates for u and v

Proposition 2. *Let the assumptions ($\mathcal{H}$) hold and assume that $\Psi \in H^1(0,1)$ is given. Then, there exists a unique weak solution $u \in H^1(0,1)$ to (10)–(12) and a unique weak solution $v \in H^1(0,1)$ to (13)–(15). Furthermore, u and v belong to $\mathcal{C}^1([0,1])$ and verify the L^∞-estimate (19).*

Proof. The differential problems for u and v (10)–(15) are both of the form

$$\partial_x(e^{\lambda\Psi}\partial_x w) = 0 \quad \text{on } (0,1), \tag{20}$$

$$\partial_x w - A_0 w = -B_0 \quad \text{at } x = 0, \tag{21}$$

$$\partial_x w + A_1 w = B_1 \quad \text{at } x = 1, \tag{22}$$

with $A_0, A_1 > 0, B_0, B_1 \ge 0$ (thanks to ($\mathcal{H}$)) and $\Psi \in H^1(0,1) \subset \mathcal{C}([0,1])$. Using (20), there exists a constant J such that $e^{\lambda\Psi}\partial_x w = J$ and then, the solution to this problem is unique and defined by

$$w(x) = w(0) + J\int_0^x e^{-\lambda\Psi(s)}\,ds, \quad \forall x \in [0,1]$$

with

$$w(0) = \frac{B_0}{A_0} + \frac{J}{A_0}e^{-\lambda\Psi(0)} \quad \text{and} \quad J = \frac{B_1/A_1 - B_0/A_0}{\int_0^1 e^{-\lambda\Psi(x)}dx + e^{-\lambda\Psi(1)}/A_1 + e^{-\lambda\Psi(0)}/A_0}.$$

It is clear that $w \in \mathcal{C}^1([0,1])$. Furthermore, if $B_1/A_1 - B_0/A_0 \ge 0$, J is positive and the function w is increasing. Hence

$$0 \le \frac{B_0}{A_0} \le w(0) \le w(x) \le w(1) \le \frac{B_1}{A_1} \quad \forall x \in [0,1].$$

But, if $B_1/A_1 - B_0/A_0 \le 0$, J is negative and the function w is decreasing. Hence

$$0 \le \frac{B_1}{A_1} \le w(1) \le w(x) \le w(0) \le \frac{B_0}{A_0} \quad \forall x \in [0,1].$$

Therefore, in any case we have $0 \le w(x) \le \max\left(\frac{B_0}{A_0}, \frac{B_1}{A_1}\right)$ for all $x \in [0,1]$.

Let us come back to u. The system (10)–(12) for u has the form (20)–(22) with

$$\lambda = -3, \qquad A_0 = m_1^0 e^{-3b_1^0\Psi(0)} + k_1^0 e^{3a_1^0\Psi(0)}, \qquad A_1 = m_1^L e^{-3b_1^L(V-\Psi(1))} + k_1^L e^{3a_1^L(V-\Psi(1))},$$

$$B_0 = P_m m_1^0 e^{-3b_1^0\Psi(0)} e^{3\Psi(0)} \qquad B_1 = P_m k_1^L e^{3a_1^L(V-\Psi(1))} e^{3\Psi(1)}.$$

But, since $a_1^L + b_1^L = 1$ and $a_1^0 + b_1^0 = 1$,

$$\frac{B_1}{A_1} = P_m \frac{e^{3\Psi(1)}}{1+\frac{m_1^L}{k_1^L} e^{3\Psi(1)} e^{-3V}} \leq \frac{k_1^L}{m_1^L} e^{3V} P_m,$$

$$\frac{B_0}{A_0} = P_m \frac{e^{3\Psi(0)}}{1+\frac{k_1^0}{m_1^0} e^{3\Psi(0)}} \leq \frac{m_1^0}{k_1^0} P_m.$$

Similar computations for v conclude the proof of (19) and of Proposition 2. □

2.2. Estimates for Ψ

Proposition 3. *Let the assumptions $(\mathcal{H})$ hold and assume that $u,\ v \in \mathcal{C}([0,1])$ are given and satisfy (19). There exists a unique weak solution $\Psi \in H^1(0,1)$ to (16)–(18). Furthermore, there exists M depending only on the data of the problem (in $(\mathcal{H})$) such that*

$$\|\Psi\|_{H^1(0,1)} \leq M \quad \text{and} \quad \|\Psi\|_{\mathcal{C}([0,1])} \leq M. \tag{23}$$

Proof. A weak solution to (16)–(18) is a function $\Psi \in H^1(0,1)$ such that for all $\varphi \in H^1(0,1)$

$$\int_0^1 \partial_x\Psi\, \partial_x\varphi\, dx + \frac{1}{\alpha_1}\Psi(1)\varphi(1) + \frac{1}{\alpha_0}\Psi(0)\varphi(0) = \frac{U_1}{\alpha_1}\varphi(1) + \frac{U_0}{\alpha_0}\varphi(0) - \frac{1}{\lambda^2}\int_0^1 \left(e^{\Psi} v - 3e^{-3\Psi} u + 5\right)\varphi\, dx. \tag{24}$$

The uniqueness of a weak solution is obtained by contradiction. We assume that there exist two weak solutions Ψ_1 and Ψ_2 and we take the difference of (24) for Ψ_1 and Ψ_2. With $\varphi = \Psi_1 - \Psi_2$ as the test function, using the monotonicity of the application $\Psi \to e^{\Psi} v - 3e^{-3\Psi} u$ (because u and v are nonnegative), we get

$$\int_0^1 |\partial_x(\Psi_1 - \Psi_2)|^2\, dx + \frac{1}{\alpha_1}|\Psi_1(1) - \Psi_2(1)|^2 + \frac{1}{\alpha_0}|\Psi_1(0) - \Psi_2(0)|^2 \leq 0,$$

which yields $\Psi_1 = \Psi_2$.

Let us now introduce $\mathcal{J}$ defined on $H^1(0,1)$ by

$$\mathcal{J}(\Phi) = \frac{1}{2}\int_0^1 (\partial_x\Phi)^2 + \frac{1}{2\alpha_1}\Phi(1)^2 + \frac{1}{2\alpha_0}\Phi(0)^2 + \frac{1}{\lambda^2}\int_0^1 (e^{\Phi} v + e^{-3\Phi} u + 5\Phi) - \frac{U_1}{\alpha_1}\Phi(1) - \frac{U_0}{\alpha_0}\Phi(0).$$

It is a continuous and strictly convex function on $H^1(0,1)$ and (24) corresponds to the Euler–Lagrange equation for the function $\mathcal{J}$. Therefore, we prove the existence of a weak solution to (16)–(18) by proving the existence of a minimum for $\mathcal{J}$. As $H^1(0,1)$ is reflexive and $\mathcal{J}$ strictly convex, it remains to verify that $\mathcal{J}$ is coercive (see for instance [5]).

We provide $H^1(0,1)$ with its usual norm $\|\cdot\|_{H^1}$ and a new norm $|||\cdot|||_{H^1}$ defined by

$$\|\Phi\|_{H^1(0,1)}^2 = \int_0^1 (\partial_x\Phi)^2 + \int_0^1 \Phi^2 \quad \text{and} \quad |||\Phi|||_{H^1(0,1)}^2 = \int_0^1 (\partial_x\Phi)^2 + \Phi(0)^2 + \Phi(1)^2.$$

Using the compact injection from $H^1(0,1)$ to $\mathcal{C}([0,1])$ and an adaptation of Poincaré inequality, it can be shown that these two norms are equivalent.

Now, using the Young inequality and positivity of u and v we have

$$\left|\frac{U_1}{\alpha_1}\Phi(1)\right| \leq \frac{1}{4\alpha_1}\Phi(1)^2 + \alpha_1 U_1^2, \qquad \left|\frac{U_0}{\alpha_0}\Phi(0)\right| \leq \frac{1}{4\alpha_0}\Phi(0)^2 + \alpha_0 U_0^2, \qquad \int_0^1 (e^{\Phi} v + e^{-3\Phi} u) \geq 0.$$

Hence

$$\mathcal{J}(\Phi) \geq \min\left(\frac{1}{2}, \frac{1}{4\alpha_1}, \frac{1}{4\alpha_0}\right)|||\Phi|||_{H^1(0,1)}^2 + \frac{5}{\lambda^2}\left(\int_0^1 \Phi\right) - \alpha_1 U_1^2 - \alpha_0 U_0^2, \quad \forall \Phi \in H^1(0,1).$$

Using Cauchy–Schwartz inequality, equivalence of norms and again the Young inequality, we obtain

$$\mathcal{J}(\Phi) \geq \mu |||\Phi|||_{H^1(0,1)}^2 - \nu \quad \forall \Phi \in H^1(0,1), \tag{25}$$

where $\mu > 0$ and $\nu \geq 0$ only depend on the data $U_0, U_1, \alpha_0, \alpha_1$ and λ^2. Therefore $\mathcal{J}$ is coercive and admits a minimum Ψ on $H^1(0,1)$. From $\mathcal{J}(\Psi) \leq \mathcal{J}(0) = \frac{1}{\lambda^2}\int_0^1 (u+v)$ and (25), we get

$$\|\Psi\|_{H^1(0,1)} \leq M(\sup u, \sup v, U_0, U_1, \alpha_0, \alpha_1, \lambda^2).$$

With the compact injection of $H^1(0,1)$ into $\mathcal{C}([0,1])$, we conclude the proof of (23). □

3. The existence of steady-state solutions

In this section, we prove Theorem 1. To this end, we introduce

$$\mathcal{S} = \left\{(u, v) \in \mathcal{C}([0, 1])^2;\ 0 \le u(x) \le \mathcal{U},\ 0 \le v(x) \le \mathcal{V},\ \forall x \in [0, 1]\right\}$$

with $\mathcal{U} = \max\left(\frac{k_1^L}{m_1^L} e^{3V}, \frac{m_1^0}{k_1^0}\right) P_m$ and $\mathcal{V} = \max\left(\frac{k_2^L}{m_2^L} e^{-V}, \frac{m_2^0}{k_2^0}\right) N_m$.

Then, we consider the mapping $H : (u_0, v_0) \in \mathcal{S} \mapsto (u, v) \in \mathcal{S}$ defined by the two following steps:

- $\beta : (u_0, v_0) \in \mathcal{S} \to \Psi \in H^1(0, 1)$, the unique solution to (16)–(18) (with u_0, v_0 instead of u, v).
- $\gamma : \Psi \in H^1(0, 1) \to (u, v) \in \mathcal{S}$, the unique solution to (10)–(15).

Thanks to Propositions 2 and 3, we know that this mapping is well defined. Furthermore, each fixed point (u, v) of H determines clearly a weak solution (Ψ, u, v) to (10)–(18).

Note that $\mathcal{S}$ is a closed and convex subset of $\mathcal{C}([0, 1])$. We shall now show that $H : \mathcal{S} \to \mathcal{S}$ is continuous and $H(\mathcal{S})$ precompact. When this result is established we conclude the existence of a fixed point of H in $\mathcal{S}$ from Schauder's theorem (see [6]).

The first step: continuity of γ.

Let $\Psi \in H^1(0, 1)$ and set $(u, v) = \gamma(\Psi)$. We have already seen in Section 2.1 that u and v can be explicitly computed. For u for instance, we have

$$u(x) = u(0) + J(\Psi) \int_0^x e^{3\Psi(s)}\, ds \quad \forall x \in [0, 1], \text{ with} \tag{26}$$

$$u(0) = \frac{B_0(\Psi)}{A_0(\Psi)} + \frac{J(\Psi)}{A_0(\Psi)} e^{3\Psi(0)} \quad \text{and} \quad J(\Psi) = \frac{B_1(\Psi)/A_1(\Psi) - B_0(\Psi)/A_0(\Psi)}{\int_0^1 e^{3\Psi(x)} dx + e^{3\Psi(1)}/A_1(\Psi) + e^{3\Psi(0)}/A_0(\Psi)}. \tag{27}$$

All the maps involved in the computation of u (and similarly of v) are continuous from $H^1(0, 1)$ to $\mathcal{C}([0, 1])$. Therefore γ is continuous.

The second step: continuity of β.

For (u_0, v_0) and $(\widetilde{u_0}, \widetilde{v_0})$ in $\mathcal{S}$, we set $\Psi = \beta(u_0, v_0)$, $\widetilde{\Psi} = \beta(\widetilde{u_0}, \widetilde{v_0})$ and $\theta = \Psi - \widetilde{\Psi}$. Subtracting (24) written for Ψ and $\widetilde{\Psi}$ and applying with θ as a test function, we get

$$\begin{aligned}\int_0^1 |\partial_x \theta|^2 + \frac{1}{\alpha_1}\theta(1)^2 + \frac{1}{\alpha_0}\theta(0)^2 &= -\frac{1}{\lambda^2}\int_0^1 \left(e^{\Psi} v_0 - e^{\widetilde{\Psi}} \widetilde{v_0}\right) \theta dx + \frac{3}{\lambda^2}\int_0^1 \left(e^{-3\Psi} u_0 - e^{-3\widetilde{\Psi}} \widetilde{u_0}\right) \theta \\ &\le \frac{1}{\lambda^2}\int_0^1 e^{\Psi} (v_0 - \widetilde{v_0})\, \theta dx + \frac{3}{\lambda^2}\int_0^1 e^{-3\Psi} (u_0 - \widetilde{u_0})\theta dx.\end{aligned}$$

Setting $\delta = \min(1, 1/\alpha_1, 1/\alpha_0)$, we have

$$\begin{aligned}\lambda^2 \delta \|\theta\|^2_{H^1(0,1)} &\le \int_0^1 e^{\Psi} |v_0 - \widetilde{v_0}|\ |\theta| dx + 3\int_0^1 e^{-3\Psi} |u_0 - \widetilde{u_0}|\ |\theta| dx, \\ &\le C\left(\|v_0 - \widetilde{v_0}\|_{L^2(0,1)} \|\theta\|_{L^2(0,1)} + \|u_0 - \widetilde{u_0}\|_{L^2(0,1)} \|\theta\|_{L^2(0,1)}\right),\end{aligned}$$

thanks to (23) and the Cauchy–Schwarz inequality. Using the Young inequality, we obtain

$$\|\theta\|^2_{H^1(0,1)} \le C'\left(\|v_0 - \widetilde{v_0}\|^2_{H^1(0,1)} + \|u_0 - \widetilde{u_0}\|^2_{H^1(0,1)}\right).$$

Thus, β is a continuous map.

The third step: compactness of H.

It remains to prove that H maps $\mathcal{S}$ into a precompact subset of $\mathcal{C}([0, 1])$. Therefore, we prove that u and v defined by $(u, v) = H(u_0, v_0)$ with $(u_0, v_0) \in \mathcal{S}$ are bounded in $H^1(0, 1)$. Indeed, u, for instance, is defined by (26)–(27) and thanks to (23), there exists C depending only on the data introduced in $(\mathcal{H})$ such that

$$\|u\|_{H^1(0,1)} \le C.$$

We have a similar result for v.

Finally, with the compact injection of $H^1(0, 1)$ into $\mathcal{C}([0, 1])$, $H(\mathcal{S})$ is precompact and $H : \mathcal{S} \to \mathcal{S}$ is continuous. Hence, by Schauder's fixed point theorem, H has a fixed point in $\mathcal{S}$ and the system (10)–(18) admits a solution. This achieves the proof of Theorem 1.

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