Mathematical analysis of a reaction-diffusion system modeling the phenomena of crevice corrosion in one dimension space with measure initial data

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ABSTRACT. The aim of this paper is to provide a mathematical study of nonlinear partial differential equations modeling the corrosion phenomena. We present the modelisation of our problem and the mathematical analysis of the obtained system. The originality of this work can be seen in the measure initial data and the techniques developed here to complete the mathematical study.

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1. Introduction

Crevice corrosion is a localized form of the corrosion, this attack if generally associated to the presence of small volumes of stagnant solution in occluded interstics, beneath deposits and seals, or in crevices.

Crevice corrosion is encountered particularly in metals that their structure is resistance to the stability of a passive film, since these films are unstable in the presence of high concentrations of Cl^- and H^+ ions.

The basic mechanism of crevice corrosion in passivatable alloys exposed to aerated chloride-rich media is gradual acidification of the solution inside the crevice, leading to the appearance of highly aggressive local conditions that destroy the passivity.

As dissolution of the metal M continues, an excess of M^{n+} ions is created in the crevice, which can only be compensated by electromigration of the Cl^- ions [6]. Most metallic chlorides hydrolyse, and this is particularly true for the elements in stainless steels and aluminium alloys. The acidity in the crevice increases (pH 1-3) as well as the Cl^- ion concentration (up to several times the mean value in the solution). The dissolution reaction in the crevice is then promoted and the oxygen reduction reaction becomes localized on the external surfaces close to the crevice. This "autocatalytic" process accelerates rapidly, even if several days or weeks were necessary to get it under way.

This models for crevice corrosion have been studied in electrochemical and physical literature (see G.R. Engelhard [6], S.M. Sharland [9]), and the mathematical solution was given by S.M. Sharland [10] in the steady state case.

This paper is organized as follows, we start by giving the mathematical model of the studied phenomena, we pursue it by the main result which is the existence for any measure data then we give the proof of the main result.

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2. Modeling

In the mathematical simulation of the corrosion of steels in neutral solutions, at least six species in the solution must be taken into account [10]. These species are metal ions Fe^{2+} from the dissolution process, sodium (Na^+) and chloride (Cl^-) ions to facilitate current flow within the crevice, hydrogen (H^+) and hydroxyde (OH^-) ions from the dissociation of water, and a metal hydrolysis product $(Fe(OH)^+)$. The term H_2O have no kinetic effect in the solution, since its concentration remains very close to the concentration of pure water.

The concentrations of the species are denoted as follows:

$$c_1 = [Fe^{2+}], c_2 = [Fe(OH)^+], c_3 = [Na^+], c_4 = [Cl^-], c_5 = [H^+], c_6 = [OH^-].$$

The cathodic reduction of oxygen, hydrogen ion, and water,

 $O_2 + 2H_2O + 4e \longrightarrow 4OH^-,$ $2H^+ + 2e \longrightarrow H_2,$ and, $2H_20 + 2e \longrightarrow 2OH^- + H_2$. Additionally, the two homogeneous reactions that are considered • $Fe^{2+} + H_2O \rightleftharpoons Fe(OH)^+ + H^+,$

- $H_2 O \rightleftharpoons H^+ + O H^-$.

With equilibrium constants K_1 and K_2 respectively. The forward and backward reaction rates for each reaction are denoted k_1, k_{-1} and k_2, k_{-2} , where

$$K_1 = \frac{k_1}{k_{-1}}, \quad K_2 = \frac{k_2}{k_{-2}}.$$

The evolution of the concentrations of the ions are given by the Nernst-Planck equation.

For i = 1, ..., 6

$$\frac{\partial c_i}{\partial t} = -\operatorname{div.}(N_i) + f_i.$$

We denote by c_i the concentrations of the species, f_i the rate of creation of ionic species i given as follows,

$$f_1(c) = -k_1c_1 + k_{-1}c_2c_5,$$

$$f_2(c) = k_1c_1 - k_{-1}c_2c_5,$$

$$f_3(c) = 0,$$

$$f_4(c) = 0,$$

$$f_5(c) = k_1c_1 - k_{-1}c_2c_5 + k_2 - k_{-2}c_5c_6,$$

$$f_6(c) = k_2 - k_{-2}c_5c_6.$$

and N_i the density of ion flux given by:

$$N_i = -d_i (\nabla c_i + \frac{z_i F}{RT} c_i \nabla \phi), \ i = 1, ..., 6,$$

where, d_i is the diffusion coefficient, z_i is the charge, T the temperature, R is the gas constant, F is the Faraday constant, and ϕ is the electric potential in the solution.

We consider that the species satisfy the electro-neutrality condition,

$$-\varepsilon\Delta\phi = \sum_{k=1}^{6} z_k c_k.$$

We consider the one dimension space case of the problem, and we study the problem in more general case, where the diffusion coefficients depend on time and space $d_i = d_i(t, x)$.

The electric potential satisfies the system: For $t \in [0,T]$

$$\begin{cases} -\varepsilon \frac{\partial^2 \phi}{\partial x^2} = \sum_{k=1}^6 z_k c_k & \text{ in } \Omega\\ \phi(t,s) = 0 & \text{ for } s = 0, L. \end{cases}$$

Then the concentrations satisfy the system: For i = 1, ..., 6

$$\begin{cases} \partial_t c_i - \partial_x (d_i(t, x) \partial_x c_i + m_i(t, x) c_i \partial_x \phi) = f_i \\ -\varepsilon \frac{\partial^2 \phi}{\partial x^2} = \sum_{k=1}^6 z_k c_k \end{cases}$$
(1)

where $m_i(t,x) = d_i(t,x) \frac{z_i F}{RT}$ is the mobility. At the boundary we suppose that,

$$d_i(t,s)\partial_x c_i(t,s) + m_i(t,s)c_i(t,s)\partial_x \phi(t,s) = \eta_i(t,s,c,\phi), \quad \text{for } s = 0, L, \quad t \in [0,T].$$

Where,

$$\eta_1 = \frac{i_p}{2F}, \quad \eta_2 = \eta_3 = \eta_4 = 0, \quad \eta_5 = \frac{i_{H^+}}{F}, \quad \eta_6 = \frac{i_{OH^-}}{F}$$

 i_p : the passive corrosion current density (independent of the potential),

$$i_{H^+} = A_1 c_5 exp[-\frac{\alpha_1 F E}{RT}],$$

$$i_{OH^-} = -A_2 exp[-\frac{\alpha_2 F E}{RT}],$$

$$\alpha_1 = \alpha_2 exp[-\frac{\alpha_2 F E}{RT}],$$

 α_1, α_2 are transfer coefficients, $E = E_{corr} - \phi$ is the local electrode potential, A_1, A_2 are constants that do not depend on potential.

Then the concentrations satisfy the system: For i = 1, ..., 6

$$\begin{cases} \frac{\partial c_i}{\partial t}(t,x) - \frac{\partial}{\partial x}(d_i(t,x)\frac{\partial c_i}{\partial x}(t,x)) + \\ +m_i(t,x)c_i(t,x)\frac{\partial \phi}{\partial x}(t,x)) = f_i(t,x,c) & \text{in } Q_T \\ -\varepsilon \partial_{xx}\phi = \sum_{k=1}^6 z_k c_k(t,x) & \text{in } \Omega \\ c_i(0,x) = \mu_i & \text{in } \mathcal{M}_b(\Omega) \\ d_i(t,s)\partial_x c_i(t,s) + m_i(t,s)c_i(t,s)\partial_x \phi(t,s) = \\ & = \eta_i(t,s,c,\phi) & \text{for } s = 0, L \text{ and } t \in]0,T[\\ \phi(t,s) = 0 & \text{for } s = 0, L \text{ and } t \in]0,T[\end{cases}$$

where,

 $\mathcal{M}_b(\Omega) = \{\mu \text{ bounded Radon measure in } \Omega\}.$

Definition 2.1. Let $c_i \in \mathcal{C}([0, T[; L^1(\Omega)) \text{ and } \mu_i \in \mathcal{M}_b(\Omega))$. We say that $c_i(0, x) = \mu_i$ in $\mathcal{M}_b(\Omega)$ if for every $\varphi \in \mathcal{C}_b(\Omega)$

$$\lim_{t\to 0}\int_\Omega c_i(t,x)\varphi dx = <\mu_i,\varphi>$$

where, $C_b = \{\varphi : \Omega \to \mathbb{R} \text{ continuous and bounded in } \Omega\}.$

Let Ω be the open set [0, L], for T > 0 we denote by $Q_T = [0, T] \times \Omega$.

Throughout this paper we consider a general Reaction-diffusion system which involves NS species, and we assume for i = 1, ..., NS

i) $d_i \in \mathcal{C}^2(Q_T)$, for any T > 0, $x \in \Omega$ there exist $\underline{d}, \overline{d} > 0$ such that

$$0 < \underline{d} \le d_i \le d < +\infty \qquad \text{on } Q_T.$$

- ii) $\mu_i \in \mathcal{M}_b(\Omega)$.
- iii) $\eta_i(., s, ., .) \in L^{\infty}(0, T)$ for s = 0, L. iv) $f_i : Q_T \times \mathbb{R}^{NS} \longrightarrow \mathbb{R}$ measurable.
- v) $f = (f_1, ..., f_{NS}) \in \mathcal{C}^1([0, +\infty) \times \Omega \times \mathbb{R}^{NS}; \mathbb{R}^{NS})$; is quasi-positive, i.e

$$f_i(t, x, \gamma) \ge 0$$
 for any $(t, x, \gamma) \in (0, +\infty) \times \Omega \times [0, +\infty)^{NS}$

such that $\gamma_i = 0$.

vi) There exist an upper triangular invertible matrix $Q \in \mathbb{R}^{NS \times NS}$ with nonnegative diagonal entries and $b \in \mathbb{R}^{NS}_+$ a given vector such that

$$\begin{cases} QF(t, x, u) \le (1 + \sum_{1 \le j \le NS} u_j)b\\ \text{for all } u \in (\mathbb{R}^+)^{NS} \text{and a.e } (t, x) \in Q_T \end{cases}$$
(3)

where, Q is the matrix

$$\begin{pmatrix} 1 & 1 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 \end{pmatrix}$$

3. Existence for any measure data

Theorem 3.1. Assume that the assumptions (i)-(v) hold, and assume also that $\forall i =$ 1, ..., NS, $\mu_i \in \mathcal{M}_h^+(\Omega)$.

Then, there exist $c_i \in L^1(0,T;W^{1,1}(\Omega)) \cap \mathcal{C}(]0,T[;\mathcal{M}_b(\Omega)) \text{ and } \phi \in L^\infty(0,T;W_0^{1,\infty}(\Omega))$ satisfied in the following sense :

where, $c = (c_1, ..., c_{NS})$.

Proof of the Theorem 3.1.

3.1. Approximate scheme. We consider the function of truncation $\delta_n \in \mathcal{C}_0^{\infty}$ that satisfies,

$$\begin{cases} 0 \le \delta_n \le 1\\ \delta_n(r) = 1 \text{ if } |r| \le n\\ \delta_n(r) = 0 \text{ if } |r| \ge n+1 \end{cases}$$

We define for every $c \in \mathbb{R}^{NS}$

$$f_i^n(c) = \delta_n(|c|) \widetilde{f}_i(c),$$

where, $\stackrel{\sim}{f}_i = f_i(c^+)$. Let's now truncate the initial data $(\mu_i)_{1 \le i \le NS}$ as follows,

$$\begin{cases} c_{i,n}^0 \in \mathcal{C}_0^\infty(\Omega) & \text{such that } c_{i,n}^0 \ge 0, \ \|c_{i,n}^0\|_{L^1(\Omega)} \le \|\mu_i\|_{\mathcal{M}_b(\Omega)} \\ \text{and } c_{i,n}^0 \longrightarrow \mu_i \text{ in } \mathcal{M}_b(\Omega). \end{cases}$$

We denote by $\eta_i^n(t, x) = \eta_i(t, x, c_n, \phi_n).$ Now, let's consider the truncated system

For all
$$0 < t < T$$
, $c_{i,n} \in \mathcal{C}(0, T; L^{1}(\Omega)) \cap L^{1}(0, T; W^{1,1}(\Omega))$,
 $\phi_{n} \in L^{\infty}(0, T; W_{0}^{1,\infty}(\Omega))$
Let $v \in \mathcal{C}^{\infty}(Q_{T})$ such that $v(T, .) = 0$,
 $-\int_{Q_{T}} c_{i,n}\partial_{t}v + \int_{Q_{T}} d_{i}(t, x)\partial_{x}c_{i,n}\partial_{x}v + \int_{Q_{T}} m_{i}(t, x)c_{i,n}\partial_{x}\phi_{n}\partial_{x}v =$
 $\int_{Q_{T}} f_{i}^{n}(c_{i,n})v + \int_{\Omega} c_{i,n}^{0}v(0, .) - \int_{0}^{T} \eta_{i}^{n}(t, 0)v(., 0) + \int_{0}^{T} \eta_{i}^{n}(t, L)v(., L)$ (4)
Let $\psi \in \mathcal{C}_{0}^{\infty}(\Omega)$
 $\varepsilon \int_{\Omega} \partial_{x}\phi_{n}\partial_{x}\psi = \int_{\Omega} \sum_{i=1}^{NS} z_{i}c_{i,n}\psi$

Lemma 3.2. The problem (4) has a solution in $\mathcal{C}(]0, T[; L^1(\Omega)) \cap L^1(Q_T)$

Proof. Let $c_n = (c_{1,n}, ..., c_{NS,n})$ satisfy the problem

$$\begin{cases} \int_{Q_T} \partial_t c_{i,n} \psi + \int_{Q_T} (d_i(t, x) \partial_x c_{i,n} + m_i(t, x) c_{i,n} \partial_x \phi_n) \partial_x \psi = \\ = \int_{Q_T} f_i^n \psi - \int_0^T \eta_i^n(t, 0) \psi(., 0) + \int_0^T \eta_i^n(t, L) \psi(., L) \\ \forall \psi \in \mathcal{C}^{\infty}(Q_T), \\ \varepsilon \int_{\Omega} \partial_x \phi_n \partial_x \xi = \int_{\Omega} \sum_{j=1}^{NS} z_j c_{j,n} \xi \quad \text{a.e } t > 0, \ \forall \xi \in \mathcal{C}_0^{\infty}(\overline{\Omega}) \\ c_{i,n}(0, x) = c_{i,n}^0 \end{cases}$$
(5)

Let us introduce the following application:

$$H_{\phi_n}: L^1(Q_T) \longrightarrow L^1(Q_T)$$
$$v_i \longrightarrow c_{i,n}$$

where $\forall t \in]0, T[, \phi_n \text{ is the unique solution of the elliptic problem}$

$$\begin{cases} -\varepsilon \partial_{xx} \phi_n = \sum_{i=1}^{NS} z_i c_{i,n} & \text{in } \Omega\\ \phi_n(s) = 0 & \text{for } s = 0, L \end{cases}$$
(6)

and $H_{\phi_n}(v) = c_{i,n}$ satisfies the following system :

$$\begin{cases} \int_{Q_T} \partial_t c_{i,n} \psi + \int_{Q_T} d_i(t,x) \partial_x c_{i,n} \partial_x \psi + \int_{Q_T} m_i(t,x) c_{i,n} \partial_x \phi_n \partial_x \psi = \\ \int_{Q_T} f_i^n \psi + \int_0^T \eta_i^n(t,L) \psi(.,L) - \int_0^T \eta_i^n(t,0) \psi(.,0) \quad \forall \psi \in \mathcal{C}^{\infty}(Q_T), \\ c_{i,n}(0,x) = c_{i,n}^0. \end{cases}$$
(7)

According to Baras-Pierre [8] the problem (7) have a solution in $L^1(Q_T)$. Then H_{ϕ_n} is well defined.

Using the Schauder's fixed point theorem we prove that H_{ϕ_n} admits a fixed point:

• Let's prove that $\forall A$ bounded subset of $L^1(Q_T)$ its image by H_{ϕ_n} is relatively compact in $L^1(Q_T)$.

Let $(v_i^n) \in L^1(Q_T)$ bounded sequence, then

 $f_i^n = f_i^n(t, x, v_1^n, ..., v_{NS}^n)$ is uniformly bounded in $L^1(Q_T)$.

Let $c_i^n = H_{\phi_n}(v_i^n)$, the application $(c_{i,n}^0, f_i^n) \longrightarrow (c_{i,n})$ is compact from $L^1(\Omega) \times L^1(Q_T)$ to $L^1(Q_T)$. Then, $(c_{i,n})$ is relatively compact in $L^1(Q_T)$.

• Let us prove that H_{ϕ_n} is continuous.

Let (v_i^n) a sequence that converges to (v_i) in $L^1(Q_T)$, we extract a subsequence denoted also (v_i^n) that converges almost every where in Q_T , and $\partial_x v_i^n \longrightarrow \partial_x v_i$ almost everywhere.

Since f_i^n is continuous then $f_i^n \longrightarrow f_i$ almost everywhere in Q_T , and f_i^n is bounded, then via the Lebesgue dominated convergence theorem we deduce

$$f_i^n \longrightarrow f_i \in L^1(Q_T)$$

Then $c_{i,n}$ converges to c_i solution of (7) in $L^1(Q_T)$.

Since (v_i^n) is bounded in $L^1(Q_T)$ then $(c_{i,n})$ is relatively compact in $L^1(Q_T)$, then by the uniqueness of the limit we conclude that

$$H_{\phi_n}(v_i^n) \longrightarrow c_i = H_{\phi_n}(v_i).$$

• Let us prove $H_{\phi_n}(L^1(Q_T)) \subset B(0, R)$. Let $v_i^n \in L^1(Q_T)$ and $c_{i,n} = H_{\phi_n}(v_i)$ solution of (7). Let $t \in]0, T[$, and we take 1 as a test function we get,

$$\int_{Q_t} \partial_t c_{i,n} = \int_0^t \eta_i^n(.,L) - \int_0^t \eta_i^n(.,0) + \int_{Q_t} f_i^n,$$
$$\int_{\Omega} c_{i,n}(t) = \int_0^t \eta_i^n(.,L) - \int_0^t \eta_i^n(.,0) + \int_{Q_t} f_i^n + \int_{\Omega} c_{i,n}(0).$$

We integrate over [0, T] we obtain,

$$\|c_{i,n}\|_{L^1(Q_T)} \le C_{n,T} + T(\|\mu_i\|_{\mathcal{M}_b(\Omega)} + T\sum_{s=0,L} \|\eta_i^n(.,s)\|_{L^\infty([0,T[))}),$$

= R.

Finally, H_{ϕ_n} has a fixed point $c_{i,n}$ solution of (4)

3.2. A priori estimates.

Lemma 3.3. There exists a constant M depending on $\sum_{1 \le j \le NS} \|\mu_i\|_{\mathcal{M}_b(\Omega)}$ such that

i)
$$\begin{split} &\int_{\Omega}\sum_{1\leq j\leq NS}c_{j,n}\leq M.\\ &\text{ii)} \ \left\|\phi_n\right\|_{L^{\infty}(0,T;W_0^{1,\infty}(\Omega))}\leq M. \end{split}$$

Proof. i) We have

$$\partial_t \int_{\Omega} \left(\sum_{i=1}^{NS} q_{1,i} c_{i,n} \right)(t) - \int_{\Omega} \frac{\partial}{\partial x} \left(\sum_{i=1}^{NS} q_{1,i} d_i(t,x) \frac{\partial c_{i,n}}{\partial x} + \sum_{i=1}^{NS} q_{1,i} m_i(t,x) c_{i,n} \frac{\partial \phi_n}{\partial x} \right) \\ = \int_{\Omega} \sum_{i=1}^{NS} q_{1,i} f_{i,n}.$$

$$(8)$$

Since
$$\sum_{i=1}^{NS} q_{1,i} f_{i,n} \leq b_1 (1 + \sum_{i=1}^{NS} c_{i,n})$$
 we get
 $\partial_t \int_{\Omega} (\sum_{i=1}^{NS} q_{1,i} c_{i,n})(t) - \int_{\Omega} \frac{\partial}{\partial x} (\sum_{i=1}^{NS} q_{1,i} d_i(t,x) \frac{\partial c_{i,n}}{\partial x} + \sum_{i=1}^{NS} q_{1,i} m_i(t,x) c_{i,n} \frac{\partial \phi_n}{\partial x})$
 $\leq \int_{\Omega} b_1 (1 + \sum_{i=1}^{NS} c_{i,n}),$
(9)

thus

$$\partial_t \int_{\Omega} (\sum_{i=1}^{NS} q_{1,i} c_{i,n})(t) \le \sum_{i=1}^{NS} q_{1,i} \eta_i^n(t,L) - \sum_{i=1}^{NS} q_{1,i} \eta_i^n(t,0) + \int_{\Omega} b_1 (1 + \sum_{i=1}^{NS} c_{i,n}).$$
(10)

Thanks to the nonnegativity of $(q_{1,i})_{1 \le i \le NS}$ and the boundedness of η_i^n and by using boundary conditions on ϕ_n we have the following Gronwall's inequality

$$\partial_t \int_{\Omega} (\sum_{i=1}^{NS} c_{i,n})(t) \le C + \int_{\Omega} \frac{b_1}{q_0} (1 + \sum_{i=1}^{NS} c_{i,n}), \tag{11}$$

where $q_0 = \min_{1 \le i \le NS} q_{1,i}$. let us set $W_n(t) = \sum_{i=1}^{NS} c_{i,n}(t)$. By integrating on (0,T), we obtain

$$\int_{\Omega} W_n(t) \le e^{\frac{b_1}{q_0}t} \int_{\Omega} W_n(0) + k(e^{\frac{b_1}{q_0}t} - 1),$$
(12)

where, $k = q_0(C + \int_{\Omega} \frac{b_1}{q_0})/b_1$, which implies that, for each t in the interval of existence

$$\int_{\Omega} W_n(t) \le e^{\frac{b_1}{q_0}t} \sum_{i=1}^{NS} ||\mu_i||_{\mathcal{M}_b(\Omega)} + k(e^{\frac{b_1}{q_0}t} - 1).$$
(13)

ii) We have the system satisfied by ϕ_n

$$\begin{cases} -\varepsilon \partial_{xx} \phi_n = \sum_{i=1}^{NS} z_i c_{i,n} & \text{in } \Omega, \\ \phi_n(t,s) = 0 & \text{for } s = 0, L, \ \forall \ 0 < t < T. \end{cases}$$

Then,

$$\phi_n(t,s) = \frac{1}{\varepsilon} \int_{\Omega} G(x,s) (\sum_{i=1}^{NS} z_i c_{i,n}) ds,$$

where, G is green function, given by

$$G(x,s) = \begin{cases} x(L-s) & \text{if } x \leq s \\ s(L-x) & \text{if } s \leq x \end{cases}$$

Since,

$$\sum_{i=1}^{NS} z_i c_{i,n} \|_{L^{\infty}(0,T;L^1(\Omega))} \le C.$$

Thus, we can see that

$$\left\|\phi_n\right\|_{L^{\infty}(0,T;W_0^{1,\infty}(\Omega))} \le C.$$

Lemma 3.4. There exist constants R_1 , R_2 depending on $\sum_{1 \le j \le NS} \|\mu_i\|_{\mathcal{M}_b(\Omega)}$ such that

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1)
$$\sum_{1 \le i \le NS} \int_{Q_T} |f_i^n(t, x, c_n)| \le R_1.$$

2) $\sum_{1 \le i \le NS} \int_0^T |\eta_i^n(t, s)| \le R_2 \quad \text{for } s = 0, L.$

Proof. 1) Considering the hypothesis (v)

$$\sum_{i \le j \le NS} q_{ji} f_j^n \le b_i (1 + \sum_{1 \le k \le NS} c_{k,n}).$$

For i = NS, we have

$$q_{NS,NS} f_{NS}^n \leq b_{NS} (1 + \sum_{1 \leq k \leq NS} c_{k,n}),$$

$$\int_{Q_T} |f_{NS}^n| \leq \int_{Q_T} \frac{b_{NS}}{q_{NS,NS}} (1 + \sum_{1 \leq j \leq NS} |c_{j,n}|),$$

$$\int_{Q_T} |f_{NS}^n| \leq K (\sum_{1 \leq i \leq NS} \|\mu_j\|_{\mathcal{M}_b(\Omega)} + mes(Q_T)) \leq M_{NS}.$$

For i = NS - 1,

$$q_{NS-1,NS-1}f_{NS-1}^{n} + q_{NS-1,NS}f_{NS}^{n} \le b_{NS-1}(1 + \sum_{1 \le k \le NS} c_{k,n}),$$

and,

$$\begin{split} f_{NS-1}^{n} &= (f_{NS-1}^{n} + \frac{q_{NS-1,NS}}{q_{NS-1,NS-1}} f_{NS}^{n}) - \frac{q_{NS-1,NS}}{q_{NS-1,NS-1}} f_{NS}^{n}, \\ |f_{NS-1}^{n}| &\leq |f_{NS-1}^{n} + \frac{q_{NS-1,NS-1}}{q_{NS-1,NS-1}} f_{NS}^{n}| + |\frac{q_{NS-1,NS}}{q_{NS-1,NS-1}} f_{NS}^{n}|, \\ \int_{Q_{T}} |f_{NS-1}^{n}| &\leq \int_{Q_{t}} \frac{b_{NS-1}}{q_{NS-1,NS-1}} (1 + \sum_{1 \leq j \leq NS} |c_{j,n}|) + \frac{q_{NS-1,NS-1}}{q_{NS-1,NS-1}} M_{NS}, \\ &\leq K_{1}(mes(Q_{T}) + \sum_{1 \leq i \leq NS} \|\mu_{j}\|_{\mathcal{M}_{b}(\Omega)}) + K_{2} \leq M_{NS-1}. \end{split}$$

Doing the same as above for every $1 \le i \le NS$ we get,

$$\int_{Q_T} |f_i^n| \le M_i(\|\mu_j\|_{\mathcal{M}_b}, (q_{i,j})_{j=i,..,NS}).$$

Thus we obtain the desired result which is,

$$\sum_{1 \le i \le NS} \int_{Q_T} |f_i(t, x, c_n)| \le \sum_{1 \le i \le NS} M_i = R_1.$$

2) Set

$$\begin{cases} \eta_i^n = 0 & \text{for } i \in U_0 \\ \eta_i^n = -A_i exp(-\frac{\alpha_i F.E^n}{RT}) & \text{for } i \in U_1 \\ \eta_i^n = A_i c_i^n exp(-\frac{\alpha_i F.E^n}{RT}) & \text{for } i \in U_2 \end{cases}$$

Where, $E^{n} = E_{corr} - \phi_{n}, U_{0} \cup U_{1} \cup U_{2} = [1, ..., NS].$ Then we have,

$$\sum_{1 \le i \le NS} \int_0^T |\eta_i^n(t,s)| = \sum_{i \in U_1} \int_0^T |\eta_i^n(t,s)| + \sum_{i \in U_2} \int_0^T |\eta_i^n(t,s)|.$$

$$\begin{split} \text{If } i \in U_1 \\ \int_0^T |\eta_i^n(t,s)| &= \int_0^T |A_i \exp(-\frac{\alpha_i F.E^n(s)}{RT})| \le \|A_i \exp(-\frac{\alpha_i F.E^n(s)}{RT})\|_{L^\infty(0,T)}.T, \\ &= M_1(A_i,\alpha_i,\|\phi_n\|_{L^\infty(0,T,W^{1,\infty}(\Omega))}). \end{split}$$

And if $i \in U_2$

$$\int_{0}^{T} |\eta_{i}^{n}(t,s)| = \int_{0}^{T} |A_{i}c_{i}^{n}(s)\exp(-\frac{\alpha_{i}F.E(s)}{RT})|, \\
\leq ||A_{i}\exp(-\frac{\alpha_{i}F.E}{RT})||_{L^{\infty}(0,T)}||c_{i}^{n}(s)||_{L^{\infty}(0,T)}, \\
\leq M(A_{i},\alpha_{i},||\phi_{n}||_{L^{\infty}(0,T,W^{1,\infty}(\Omega))}).||c_{i}^{n}(s)||_{L^{\infty}(0,T)}) = M_{2}.$$

Thus,

$$\sum_{1 \le i \le NS} \int_0^T |\eta_i^n(t,s)| \le M_1 + M_2 = R_2.$$

3.3. Convergence. Our purpose is to prove that (c_n, ϕ_n) solution of the approximated problem (4) converge to (c, ϕ) solution of (2). From the work of Baras, Hassan and Veron [3] we have, the application $(c_{i,n}^0, f_i^n) \longrightarrow c_{i,n}$ is compact from $L^1(\Omega) \times L^1(Q_T)$ into $L^1(Q_T)$. Then, we deduce the existence of a subsequence also denoted (c_n, ϕ_n) , such that For i = 1, ..., NS

$$\begin{cases} c_{i,n} \longrightarrow c_i & \text{strongly in } L^1(Q_T), \\ c_{i,n} \longrightarrow c_i & \text{almost every where in } Q_T, \end{cases}$$

and,

$$c_{i,n}^0 \longrightarrow \mu_i \text{ in } \mathcal{M}_b(\Omega).$$

Since ϕ_n is uniformly bounded in $L^{\infty}(0,T;W_0^{1,\infty}(\Omega))$, we conclude the existence of $\phi \in L^{\infty}(0,T;W_0^{1,\infty}(\Omega))$ such that

$$\partial_x \phi_n \longrightarrow \partial_x \phi$$
 for the topology $\sigma(L^{\infty}(Q_T), L^1(Q_T))$.

Then, let's prove that

$$c_{i,n}\partial_x\phi_n \longrightarrow c_i\partial_x\phi$$
 in $\mathcal{D}'(Q_T)$.

But first we need to prove that

$$c_{i,n}\partial_x\phi_n \longrightarrow c_i\partial_x\phi$$
 for the topology $\sigma(L^1(Q_T), L^\infty(Q_T)).$

Let $\psi \in L^{\infty}(Q_T)$ we have

$$\int_{Q_T} \psi(c_{i,n}\partial_x\phi_n - c_i\partial_x\phi) = \int_{Q_T} \psi\partial_x\phi_n(c_{i,n} - c_i) + \int_{Q_T} \psi c_i(\partial_x\phi_n - \partial_x\phi).$$

For the first term we have

$$\int_{Q_T} \psi \partial_x \phi_n(c_{i,n} - c_i) \le \|\psi\|_{L^{\infty}(Q_T)} \|\partial_x \phi_n\|_{L^{\infty}(Q_T)} \|c_{i,n} - c_i\|_{L^1(Q_T)},$$

as,

$$||c_{i,n} - c_i||_{L^1(Q_T)} \xrightarrow[n \to \infty]{} 0,$$

we deduce,

$$\int_{Q_T} \psi \partial_x \phi_n(c_{i,n} - c_i) \underset{n \to \infty}{\longrightarrow} 0.$$

For the second term

$$\left|\int_{Q_T} \psi c_i(\partial_x \phi_n - \partial_x \phi)\right| \le \|\psi\|_{L^{\infty}(Q_T)} \|c_i\|_{L^1(Q_T)} \|\partial_x \phi_n - \partial_x \phi\|_{L^{\infty}(Q_T)},$$

since $|\partial_x \phi_n - \partial_x \phi| \longrightarrow 0$ for $\sigma(L^{\infty}(Q_T), L^1(Q_T))$, then

$$\int_{Q_T} \psi c_i(\partial_x \phi_n - \partial_x \phi) \longrightarrow 0.$$

Consequently,

$$\partial_t c_{i,n} - \partial_x (d_i \partial_x c_{i,n} + m_i c_{i,n} \partial_x \phi_n) \longrightarrow \partial_t c_i - \partial_x (d_i \partial_x c_i + m_i c_i \partial_x \phi) \quad \text{in } \mathcal{D}'(Q_T).$$

Otherwise, since $c_{i,n} \longrightarrow c_i$ strongly in $L^1(Q_T)$ then,

$$\sum_{i=1}^{NS} z_i c_{i,n} \longrightarrow \sum_{i=1}^{NS} z_i c_i \quad \text{strongly in } L^1(Q_T).$$

Since $f_1^n, ..., f_{NS}^n$ are continuous we have, for i = 1, ..., NS

$$f_i^n(t, x, c_n) \longrightarrow f_i(t, x, c)$$
 almost everywhere in Q_T .

To conclude we need to prove that $f_i^n \longrightarrow f_i$ in $L^1(Q_T)$ thanks to Vitalli's theorem all we have to prove is that f_i^n are equi-integrable in $L^1(Q_T)$

Lemma 3.5. For every $i = 1, ..., NS f_i^n$, is equi-integrable in $L^1(Q_T)$

Proof. Let E be a measurable set of Q_T and $\varepsilon > 0$ then,

$$\int_{E} |f_{i}^{n}| \leq \int_{E \cap [(\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji}c_{i,n} \leq \alpha]} \sup_{0 \leq c_{j,n} \leq \alpha} |f_{i}^{n}(t, x, c_{1,n}, ..., c_{NS,n})| dx dt + \int_{E \cap [\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji}c_{i,n} \geq \alpha]} |f_{i}^{n}|,$$

$$\leq I_{1} + I_{2}.$$

To investigate the terms I_1 and I_2 we are going to need the following result.

Lemma 3.6. Let σ_n be a sequence in $L^1(Q_T)$. Then the following statements are equivalent:

1)
$$\sigma_n$$
 is uniformly integrable in $L^1(Q_T)$
2)
$$\begin{cases}
There exists $J : (0, \infty) \longrightarrow (0, \infty) \text{ with } J(0^+) = 0 \text{ and} \\
(a) \quad J \text{ is convex, } J' \text{ is concave, } J' \ge 0 \\
(b) \quad \lim_{r \to +\infty} \frac{J(r)}{r} = +\infty \\
(c) \quad \sup_n \int_{Q_T} J(|\sigma_n|) \le \infty
\end{cases}$$$

Now, we choose J as given in 2) with (2-c) is replaced by

$$\sup_{n} \int_{Q_{T}} J(\sum_{1 \le i \le NS} b_{i}(1 + \sum_{1 \le k \le NS} c_{k,n})) < \infty, \qquad \sup_{n} \int_{\Omega} J(\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji}c_{i,n}^{0}) < \infty,$$
(14)

$$\sup_{n} \int_{Q_T} J(|\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji}.\eta_i^n|) < \infty.$$
(15)

This is possible by Lemma 3.6, since $\sum_{1 \le i \le NS} c_{i,n}$ converges in $L^1(Q_T)$ and

 $\sum_{\substack{1 \leq i \leq NS}} c_{i,n}^0 \text{ converges in } L^1(\Omega).$ Now we set,

$$j(r) = \int_0^r \min(J'(s), (J^*)^{-1}(s)) ds$$

where J^* is the conjugate function of J that satisfies (2-a) and (2-b) and

$$\forall r \ge 0, \ j(r) \le J(r), \qquad J^*(j'(r)) \le r.$$
(16)

For I_1 , we notice that by the assumptions on f_i and the choice of f_i^n ,

$$\sup_{0 \le c_{1,n}, \dots, c_{NS,n} \le \alpha} |f_i^n| \le \beta_i (1 + NS\alpha) \quad \text{where} \quad \beta_i = \beta_i (b_{NS}, \dots, b_i, q_{NS,NS}, \dots, q_i).$$

As a consequence,

$$I_1 \leq \beta_i.meas(E)(1+NS\alpha) < \frac{\varepsilon}{2}.$$

For I_2

$$I_{2} = \int_{E \cap [\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji}c_{i,n} \ge \alpha]} |f_{i}^{n}| \le \frac{1}{j'(\alpha)} \int_{Q_{T}} j'(\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji}c_{i,n})|f_{i}^{n}|$$

Let us prove that the term $\frac{1}{j'(\alpha)} \int_{Q_T} j'(\sum_{i=1}^{NS} \sum_{j=1}^i q_{ji}c_{i,n}) |f_i^n|$ is uniformly bounded in $L^1(Q_T)$. We set

$$R_i^n = -\sum_{j=i}^{NS} q_{i,j} f_j^n + b_i (1 + \sum_{k=1}^{NS} c_{k,n}) \quad \text{for } i = 1, ..., NS .$$

we have,

$$\partial_t c_{i,n} - \partial_x (d_i(t,x)\partial_x c_{i,n} + m_i(t,x)c_{i,n}\partial_x \phi_n) = f_i^n$$

For every *i* we multiply the equation by $\sum_{j=1}^{i} q_{j,i}$ and we summate over *i* we obtain,

$$\partial_t (\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji} c_{i,n}) - \partial_x [\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji} d_i(t, x) \partial_x c_{i,n} + (\sum_{i=1}^{NS} \sum_{j=1}^{i} m_i(t, x) q_{ji} c_{i,n}) \partial_x \phi_n] + \sum_{1 \le i \le NS} R_i^n$$
$$= \sum_{i=1}^{NS} b_i (1 + \sum_{k=1}^{NS} c_{k,n}).$$

We multiply it by $j'(\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji}c_{i,n})$ and integrate over Q_T and by putting $\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji}c_{i,n} = O_n, \text{ we obtain}$ $\int_{\Omega} j(O_n)(T) - \int_{Q_T} j'(O_n)\partial_x [\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji}d_i(t,x)\partial_x c_{i,n} + (\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji}m_i(t,x)c_{i,n})\partial_x \phi_n] + \int_{Q_T} j'(O_n) \sum_{1 \le i \le NS} R_i^n$ $= \int_{Q_T} j'(O_n) \sum_{1 \le i \le NS} b_i(1 + \sum_{1 \le k \le NS} c_{k,n}) + \int_{\Omega} j(\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji}c_{i,n}^0).$

We put,

$$J_{1} = \int_{Q_{T}} j'(O_{n}) \partial_{x} \left[\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji} d_{i}(t,x) \partial_{x} c_{i,n} + \left(\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji} m_{i}(t,x) c_{i,n} \right) \partial_{x} \phi_{n} \right],$$

$$J_{2} = \int_{Q_{T}} j'(O_{n}) \sum_{1 \le i \le NS} b_{i} (1 + \sum_{1 \le k \le NS} c_{k,n}).$$

We start by investigating J_2 , we have

$$j'(r).s \le J(s) + J^*(j'(r)) \le J(s) + r.$$

Then,

$$J_2 \le \int_{Q_T} J(\sum_{1 \le i \le NS} b_i (1 + \sum_{1 \le k \le NS} c_{k,n})) + \sum_{i=1}^{NS} \sum_{j=1}^i q_{ji} c_{i,n},$$

which is bounded independently of n by Lemma 3.6.

Let us investigate J_1 , after integration by parts, we put $\theta^n = \sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji} \eta_i^n$, then we get

$$\begin{split} J_{1} &= -\int_{Q_{T}} j^{"}(O_{n})\partial_{x}(O_{n})[\sum_{i=1}^{NS}\sum_{j=1}^{i}q_{ji}d_{i}(t,x)\partial_{x}c_{i,n} + (\sum_{i=1}^{NS}\sum_{j=1}^{i}q_{ji}m_{i}(t,x)c_{i,n})\partial_{x}\phi_{n}] \\ &-\int_{0}^{T}j^{\prime}(O_{n})\theta^{n}(t,0) + \int_{0}^{T}j^{\prime}(O_{n})\theta^{n}(t,L) \\ &\leq \int_{0}^{T}j^{\prime}(O_{n})|\theta^{n}(t,L)| + \int_{0}^{T}j^{\prime}(O_{n})|\theta^{n}(t,0)| - \underline{d}\int_{Q_{T}}j^{"}(O_{n})|\partial_{x}O_{n}|^{2} - \\ &- m_{NS}\int_{Q_{T}}j^{"}(O_{n})\partial_{x}O_{n}O_{n}\partial_{x}\phi_{n} - \\ &- \int_{Q_{T}}j^{"}(O_{n})\partial_{x}(O_{n})(\sum_{i=2}^{NS}\sum_{j=1}^{i}q_{ji}(m_{i}-m_{NS})(t,x)c_{i,n})\partial_{x}\phi_{n}) \end{split}$$

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$$\leq \sum_{s=0,L} \int_{0}^{T} J(|\theta^{n}|)(t,s) + O_{n}(t,s) - \underline{d} \int_{Q_{T}} j^{"}(O_{n}) |\partial_{x}O_{n}|^{2} - m_{NS} \int_{Q_{T}} j^{"}(O_{n}) \partial_{x}O_{n} O_{n} O_{n} \partial_{x}\phi_{n} - \int_{Q_{T}} j^{"}(O_{n}) \partial_{x}(O_{n}) (\sum_{i=2}^{NS} \sum_{j=1}^{i} q_{ji}(m_{i} - m_{NS})(t,x)c_{i,n}) \partial_{x}\phi_{n}).$$

Using Young and Holder's inequalities we obtain,

$$J_1 \le \sum_{s=0,L} \int_0^T J(|\theta^n|)(t,s) + O_n(t,s) + M \int_{Q_T} j''(O_n) \left[\sum_{1\le i\le NS} |\partial_x c_{i,n}|^2 + \sum_{1\le i\le NS} |c_{i,n}|^2\right],$$

where M depends on $(\underline{d}, (m_i)_{i=1,\dots,NS}, (q)_{i,j}, \|\phi_n\|_{L^{\infty}(0,T;W_0^{1,\infty}(\Omega))})$.

Since j' is concave, $j''(r) \le \frac{j'(r)}{r}$ we have,

$$\int_{Q_T} j'' (\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji} c_{i,n}) \sum_{1 \le i \le NS} |c_{i,n}|^2 \le \int_{Q_T} j' (\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji} c_{i,n}) \sum_{1 \le i \le NS} c_{i,n},$$
$$\le \int_{Q_T} J (\sum_{1 \le i \le NS} c_{i,n}) + \sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji} c_{i,n}.$$

Which is uniformly bounded in $L^1(Q_T)$, for the second term we are going to use the equation satisfied by $c_{NS,n}$, we put $\gamma = \frac{b_{NS}}{q_{NS,NS}}$

$$\begin{split} \int_{\Omega} j(c_{NS,n})(T) + \underline{d} \int_{Q_{T}} j^{"}(c_{NS,n}) |\partial_{x}c_{NS,n}|^{2} \\ &\leq \overline{m} \int_{Q_{T}} j^{"}(c_{NS,n}) |c_{NS,n} \partial_{x}c_{NS,n}| |\partial_{x}\phi_{n}| + \gamma \int_{Q_{T}} j'(c_{NS,n})(1 + \sum_{k=1}^{NS} c_{k,n}) \\ &\quad + \int_{0}^{T} j'(c_{NS,n}) \eta^{n}_{NS}(t,0) - \int_{0}^{T} j'(c_{NS,n}) \eta^{n}_{NS}(t,L) + \int_{\Omega} j(c_{NS,n})(0), \\ &\leq M_{\varepsilon} \int_{Q_{T}} j''(c_{NS,n}) |c_{NS,n}|^{2} + \varepsilon \int_{Q_{T}} j''(c_{NS,n}) |\partial_{x}c_{NS,n}|^{2} + \gamma \int_{Q_{T}} j'(c_{2,n})(1 + \sum_{k=1}^{NS} c_{k,n}) \\ &\quad + \sum_{s=0,L} \int_{0}^{T} j'(c_{NS,n}) |\eta^{n}_{NS}|(t,s) + \int_{\Omega} j(c_{NS,n})(0), \end{split}$$

where, M_{ε} depends on $\underline{m}, \|\phi_n\|_{L^{\infty}(0,T;W_0^{1,\infty}(\Omega))}$, then,

$$\int_{\Omega} j(c_{NS,n})(T) + (\underline{d} - \varepsilon) \int_{Q_T} j''(c_{NS,n}) |\partial_x c_{NS,n}|^2 \\ \leq M_{\varepsilon} \int_{Q_T} j'(c_{NS,n}) |c_{NS,n}| + \gamma \int_{Q_T} j'(c_{NS,n}) \left(1 + \sum_{1 \le k \le NS} c_{k,n}\right) \\ + \sum_{s=0,L} \int_0^T j'(c_{NS,n}) |\eta_{NS}^n|(t,s) + \int_{\Omega} j(c_{NS,n})(0).$$

To conclude we choose ε small enough then we can see that $\int_{Q_T} j''(c_{NS,n}) |\partial_x c_{NS,n}|^2$ is uniformly bounded.

Adding the information $j''(O_n) \leq j''(c_{NS,n})$, now we can conclude that the term $\int_{Q_T} j''(O_n) |\partial_x c_{NS,n}|^2$ is uniformly bounded, doing the same as above we see that

 $\int_{Q_T} j^{"}(O_n) |\partial_x c_{i,n}|^2$ for i = 1, ..., NS is uniformly bounded so is J_1 .

Thus, $\int_{Q_T} j' (\sum_{i=1}^{NS} \sum_{j=1}^i q_{ji} c_{i,n}) \sum_{1 \le i \le NS} R_i^n$ is uniformly bounded. By the definition of R_i^n we have

$$\begin{split} \sum_{i=1}^{NS} R_i^n &= -(q_{1,1}f_1^n + (q_{12} + q_{22})f_2^n + \ldots + \sum_{j=1}^{NS} q_{j,NS}f_{NS}) + b_i(1 + \sum_{k=1}^{NS} c_{k,n}), \\ q_{1,1}f_1^n &+ (q_{12} + q_{22})f_2^n + \ldots + \sum_{j=1}^{NS} q_{j,NS}f_{NS} = -\sum_{i=1}^{NS} R_i^n + b_i(1 + \sum_{k=1}^{NS} c_{k,n}), \\ \sum_{i=1}^{NS} \alpha f_i^n &\leq \sum_{i=1}^{NS} R_i^n + b_i(1 + \sum_{k=1}^{NS} c_{k,n}), \\ \sum_{i=1}^{NS} f_i^n &\leq \frac{1}{\alpha} \sum_{i=1}^{NS} R_i^n + b_i(1 + \sum_{k=1}^{NS} c_{k,n}), \end{split}$$

where $\alpha = min(q_{1,1}, q_{12} + q_{22}, \dots, \sum_{j=1}^{n} q_{j,NS}).$

Finally we obtain,

$$\int_{Q_T} j' (\sum_{i=1}^{NS} \sum_{j=1}^{i} q_{ji} c_{i,n}) \sum_{1 \le i \le NS} |f_i^n| < \infty.$$

Going back to the term I_2

$$I_2 \le \frac{1}{j'(\alpha)} \int_{Q_T} j'(\sum_{i=1}^{NS} \sum_{j=1}^i q_{ji} c_{i,n}) |f_i^n|$$

By choosing $\alpha = \alpha(\varepsilon)$ large enough depending only on ε , I_2 can be made less than $\frac{\varepsilon}{2}$. Thus,

$$\int_E |f_i| < \varepsilon$$

Finally, this proves the equi-integrability of f_i^n for i = 1, ..., NS in $L^1(Q_T)$.

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