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Analysis of an Electroless Plating Problem

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Abstract

Electroless plating in microfluidic channels is a novel technology at the micrometer scale. As the microchannel depth varies with the flow of the chemicals, care must be taken for the channel not to run dry. Owing to the deposited chemical species the physical domain of the flow changes with time, leading to a free boundary problem. As the motion of the free boundary is small it is modeled by a transpiration approximation. With this simplification, the mathematical model, consists of a Navier-Stokes flow and an equation for the concentration of the plating chemical coupled by non standard and nonlinear boundary conditions. Existence and uniqueness are proven for the concentration equation. Numerical analysis is carried out and justifies the proposed numerical schemes and nonlinear algorithms. A numerical study is performed, in the two dimensional case, with the finite element method and an implicit Euler time-scheme.

Keywords: Electroless process, Free boundary, Finite Element Method

1 Introduction

Electroless plating is an autocatalytic process aimed at reducing complex metal cations in a liquid solution for a film or layer deposition on a base substrate [1]. This technique has been widely used in the printed circuit industry [2]. Recently, electroless process in microfluidic channels has been regarded as a promising micro or nano meter technology. Applications range from chemical etching process for electronic devices, to electrical packaging for food [3, 4]. Compared to the large-scale electroless process, the change of geometry to the micro- or nano-meter scale raises a critical issue for the deposition as the thickness becomes comparable to the dimension of flow channel. For instance, in the copper interconnecting process [5] by electroless plating, the thickness of the deposition layer of copper is large enough to risk a connection of the pillars.

From an experimental point of view, microfluidic electroless plating is very time consuming [6]. For example, even a sample preparation for investigating an electroless plating process is highly time-consuming. Hence a numerical study to predict the evolution of the domain of the fluid becomes indispensable.

Earlier simulation studies were carried out in a fixed one-dimensional domain and addressed either ion migration (e.g., [7, 8]) or emphasized the effect of flow convection [7]. Such one-dimensional

model can be useful when the fluid velocity is uniformly directed toward the rotating disk in the diffusion layer, but that is not the general case.

In this study, the mathematical analysis of the two of three-dimensional electroless plating problem is investigated. For numerical simulation, multi-dimensional electroless processes in geometrically varying micro- or nano-fluidic channels remain computationally expensive so we have investigated on bidimensional cases.

We consider a single chemical species in the electrolyte. The exchange current I_0 is given by the Butler-Volmer equation (see for example [9, 10]); it is a linear function of the electrolyte concentration c

$$I_0 = i_0 c := A \left[\exp \left(\frac{\alpha_0 z F \xi}{R \theta} \right) - \exp \left(\frac{-\beta_0 z F \xi}{R \theta} \right) \right] c, \quad (1.1)$$

where A, α_0, β_0 are physical constants, R is the perfect gas constant, F the Faraday constant and z the atomic number of the electrolyte; θ is the temperature, and ξ is the excess potential related to the interaction with other chemical species which, for our purpose is constant [7, 8]. The temperature is also assumed uniform and constant.

The plating occurs on a boundary $S(t)$ of the electrolyte, causing this interface to move inward the fluid domain but this motion is small because it is only due to plating. The plating being proportional to the concentration c , the velocity of $S(t)$ is normal to itself and given by a linear law

$$\mathbf{u} = -\alpha i_0 c \mathbf{n}$$

and α is small. On the other hand the flux of metal ion through $S(t)$ is proportional to c

$$D \frac{\partial c}{\partial \mathbf{n}} = -i_0 c$$

where D is a diffusion constant.

The concentration of the chemical species c satisfies a convection diffusion equation while the electrolyte flow is modelled by the Navier-Stokes equations.

In order to analyse this coupled problem, we approximate the small displacement of the reaction surface $S(t)$ by a transpiration approximation [11, 12] on a fixed mean surface S . It leads to an integro-differential condition on S :

$$-D \frac{\partial c}{\partial n} + i_0 c \left(1 + \frac{\alpha i_0^2}{D} \int_0^t c(s) ds \right) = 0. \quad (1.2)$$

The mathematical analysis of the Navier-Stokes equations coupled with a convection-diffusion equation for c is somewhat problematic because of the non-homogenous condition on S for the velocity. So we restrict the study to the existence of the weak solution to the convection-diffusion equation with a given fluid velocity \mathbf{u} and even this study is not straightforward. First a time-discretized approximation is shown to have a unique solution using a version of Minty-Browder's fixed point theorem and the maximum principle to prove that $0 \leq c \leq 1$. The solution of the time continuous problem is obtained as the weak limit of the of the solution of the time-discretized solutions. At the end of the paper some numerical tests are given to justify the transpiration approximation and the convergence of the backward Euler nonlinear scheme.

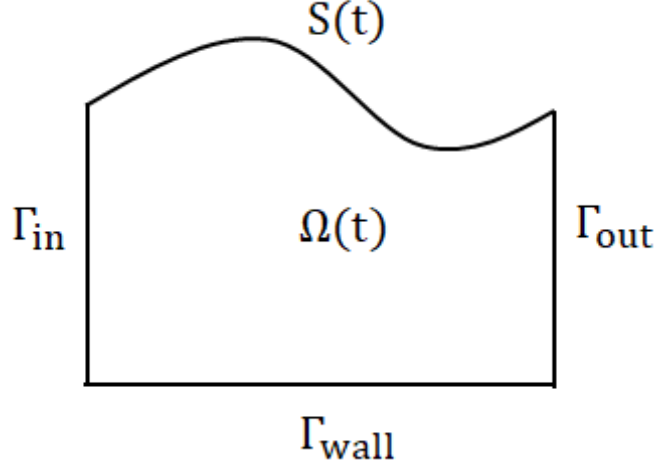


Figure 1: The physical domain is the domain occupied by the flow $\Omega(t)$; the chemical deposit is above the free boundary $S(t)$. The chemicals flow from the left boundary, Γ_{in} , to the right Γ_{out} . The bottom Γ_{wall} is a solid wall.

2 Modeling of the Physical System

The plating chemicals flow in a thin channel between a top and a bottom plate. Due to an electro-potential applied between the two plates the chemicals will deposit on the top plate. Hence the depth of the channel varies with time. A vertical cross section of the 3D system is depicted in Figure 1.

2.1 The Fluid Flow

The geometry of the fluid part is a two or three-dimensional domain $\Omega(t)$ bounded on the left by an inflow boundary Γ_{in} , on the right by an outflow boundary Γ_{out} , on the bottom by a flat wall Γ_{wall} and on the top by a time dependent boundary $S(t)$. In the three-dimensional case, the remaining boundaries are assumed to be walls. The fluid is viscous, Newtonian and incompressible, so the flow is governed by the Navier-Stokes equations for the velocity $\mathbf{u}(\mathbf{x}, t)$ and pressure $p(\mathbf{x}, t)$:

$$\partial_t \mathbf{u} + \mathbf{u} \cdot \nabla \mathbf{u} - \nu \Delta \mathbf{u} + \nabla p = 0, \quad \nabla \cdot \mathbf{u} = 0, \quad \forall \mathbf{x} \in \Omega(t), \quad \forall t \in [0, T], \quad (2.1)$$

where ν is the (constant) kinematic viscosity of the fluid. The initial velocity is given and denoted by \mathbf{u}_0 ; the inflow velocity \mathbf{u}_{in} is also given on Γ_{in} ; a no slip condition holds on $\Gamma_{wall} \cup S(t)$, and we impose a free outflow condition at Γ_{out} . So at all $t \in [0, T]$ we have:

$$\mathbf{u} = \mathbf{u}_{in} \quad \text{on } \Gamma_{in}, \quad \mathbf{u} = 0 \quad \text{on } \Gamma_{wall}, \quad -\nu \frac{\partial \mathbf{u}}{\partial n} + p \mathbf{n} = 0 \quad \text{on } \Gamma_{out}. \quad (2.2)$$

We assume that there is no back-flow on Γ_{out} : $\mathbf{u} \cdot \mathbf{n} \geq 0$ on Γ_{out} .

2.2 The Metal Ion Concentration

The metal ion concentration $c(\mathbf{x}, t)$ solves a convection-diffusion equation

$$\partial_t c + \mathbf{u} \cdot \nabla c - D \Delta c = 0, \quad \forall \mathbf{x} \in \Omega(t), \quad \forall t \in [0, T] \quad (2.3)$$

with given initial concentration c_0 ; D is the diffusion constant. The concentration is given on Γ_{in} and a no-flux condition holds on Γ_{wall} and Γ_{out} :

$$c = c_{in} \text{ on } \Gamma_{in}, \quad \frac{\partial c}{\partial n} = 0 \text{ on } \Gamma_{wall} \cup \Gamma_{out}. \quad (2.4)$$

On $S(t)$ a reaction condition is written as suggested in [7, 8],

$$-D \frac{\partial c}{\partial n} = i_0 c, \quad \mathbf{u} = -\alpha i_0 c \mathbf{n}, \quad \forall x \in S(t), \quad (2.5)$$

where i_0 and α are constants. Most important for our study: α is small.

It is also important to remember that c , being a concentration it must be non-negative and less or equal to one. In particular c_0 and c_{in} must be chosen in $[0, 1]$.

2.3 The case $\alpha = 0$

When $\alpha = 0$, there is no free boundary; consider the case $\Omega = (0, L) \times (0, 1)$. With appropriate initial and inflow conditions, the fluid velocity is

$$\mathbf{u} = (u_1, u_2)^T, \quad u_1 = y(1 - y), \quad u_2 = 0$$

Similarly, with appropriate initial and inflow conditions, the concentration depends only on time t and $y := x_2$ and solves

$$\partial_t c - D \partial_{yy} c = 0, \quad -D \partial_y c = i_0 c \text{ at } y = 1, \quad \partial_y c = 0 \text{ at } y = 0.$$

It has a closed solution $c = e^{-D\lambda^2 t} \cos(\lambda y)$ provided λ satisfies: $\lambda \tan \lambda = \frac{i_0}{D}$.

When $0 < \alpha \ll 1$, $\mathbf{u}_{in} = (y(1 - y), 0)^T$, $c_{in} = e^{-D\lambda^2 t} \cos(\lambda y)$, $\mathbf{u}_0 = \mathbf{u}_{in}$, $c_0 = c_{in}|_{t=0}$, the solution will be a linear perturbation of the above.

2.4 Transpiration Approximation

Experimental observation show that the evolution of $S(t)$ is small. Following [11, 12], we approximate (2.5) with a transpiration approximation as follows.

Let S be the initial position of $S(t)$ and let η be the distance between $S(t)$ and S normally to S , i.e.

$$S(t) = \{\mathbf{x} + \eta(\mathbf{x}, t) \mathbf{n}(\mathbf{x}) : \mathbf{x} \in S\}, \quad \eta(0) = 0$$

where $\eta(0) = 0$ is short for $\eta(\mathbf{x}, 0) = 0$ for all $\mathbf{x} \in S$. If the radius of curvature of $S(t)$ and the derivative of η along S are not large it can be shown that the difference between the normals of S and $S(t)$ is second order in η (see [11]). By definition of \mathbf{u} and by the second equation in (2.5),

$$\frac{d\eta}{dt} = \mathbf{u} \cdot \mathbf{n} = -\alpha i_0 c, \quad \eta(0) = 0, \quad \Rightarrow \quad \eta(t) = -\alpha i_0 \int_0^t c(s) ds. \quad (2.6)$$

By a Taylor expansion, the first equation in (2.5) can be written on S rather than $S(t)$:

$$-D \frac{\partial c}{\partial n}(\mathbf{x} + \eta(\mathbf{x}, t) \mathbf{n}(\mathbf{x}), t) = -D \left(\frac{\partial c}{\partial n}(\mathbf{x}, t) + \eta(\mathbf{x}, t) \frac{\partial^2 c}{\partial n^2}(\mathbf{x}, t) \right) + o(\eta)$$

$$= i_0 c(\mathbf{x} + \eta(\mathbf{x}, t) \mathbf{n}(\mathbf{x}), t) = i_0 \left(c(\mathbf{x}, t) + \eta(\mathbf{x}, t) \frac{\partial c}{\partial n}(\mathbf{x}, t) \right) + o(\eta). \quad (2.7)$$

By (2.6), it is rewritten as

$$-D \left(1 - \alpha \frac{i_0^2}{D} \int_0^t c(s) ds \right) \frac{\partial c}{\partial n}(\mathbf{x}, t) - i_0 c(\mathbf{x}, t) = \eta(\mathbf{x}, t) D \frac{\partial^2 c}{\partial n^2}(\mathbf{x}, t) + o(\eta) = O(\eta). \quad (2.8)$$

A first order in α approximation of this condition is

$$-D \frac{\partial c}{\partial n}(\mathbf{x}, t) - i_0 c(\mathbf{x}, t) = 0 \quad (2.9)$$

Neglecting $o(\eta)$ and using $\frac{1}{1-y} \approx 1+y$ and neglecting $D \frac{\partial^2 c}{\partial n^2}$ leads to

$$D \frac{\partial c}{\partial n} + i_0 c \left(1 + \frac{\alpha i_0^2}{D} \int_0^t c(s) ds \right) = 0 \quad \text{on } S. \quad (2.10)$$

In the discussion below we argue in favor of this approximation where $\eta D \frac{\partial^2 c}{\partial n^2}$ is neglected.

The second equation of (2.5) is simply written on S instead of $S(t)$. Indeed a similar Taylor expansion shows that

$$\mathbf{u} + \eta \frac{\partial \mathbf{u}}{\partial n} = -\alpha i_0 \left(c + \eta \frac{\partial c}{\partial n} \right) \mathbf{n} + o(\eta \alpha), \quad (2.11)$$

The second equation in (2.5) implies that \mathbf{u} is $O(\alpha)$; so when all normal derivatives are bounded

$$\mathbf{u} = -\alpha i_0 c \mathbf{n} + O(\eta \alpha), \quad \text{on } S \quad (2.12)$$

2.5 The Final Problem (P)

The domain and the top boundary are now fixed and denoted by Ω and S ; the boundary of Ω is

$$\Gamma := \partial\Omega = \Gamma_{in} \cup \Gamma_{wall} \cup \Gamma_{out} \cup S.$$

We propose to solve (2.1) and (2.3) in $\Omega \times (0, T)$ subject to initial conditions and boundary conditions (2.2) and (2.4) and

$$D \frac{\partial c}{\partial n} + \left(1 + \frac{\alpha i_0^2}{D} \int_0^t c(s) ds \right) i_0 c = 0 \quad \text{on } S, \quad (2.13)$$

$$\mathbf{u} = -\alpha i_0 c \mathbf{n} \quad \text{on } S. \quad (2.14)$$

2.6 Discussion:

If we had kept the term $\eta(\mathbf{x}, t) D \frac{\partial^2 c}{\partial n^2}(\mathbf{x}, t)$ in (2.10), this condition would have been second order. But even without it we expect it to be near second order when c varies slowly and Ω is elongated

because then the PDE for c , (2.3), reduces to $-D \frac{\partial^2 c}{\partial \mathbf{n}^2} \approx 0$. Whether the additional nonlinear term in (2.13) is important or not will be seen in the numerical section. In any case the first order condition is a special case of the pseudo second order condition, obtained by setting $\alpha = 0$ in (2.13). A third condition can be obtained as follows. If s denotes arc length on S , the PDE which governs c tells us that near S

$$\partial_t c + \mathbf{u} \cdot \mathbf{n} \frac{\partial c}{\partial n} = D \frac{\partial^2 c}{\partial n^2} + D \frac{\partial^2 c}{\partial s^2}$$

Assuming that the variations in s are much smaller than those in n , we obtain

$$D \frac{\partial^2 c}{\partial n^2} = \partial_t c - \alpha i_0 c \frac{\partial c}{\partial n} = \partial_t c + O(\alpha)$$

leading to

$$D \frac{\partial c}{\partial n} + i_0 c + \alpha i_0 \left(\frac{i_0^2}{D} c + \partial_t c \right) \int_0^t c(s) ds = 0 \quad \text{on } S \quad (2.15)$$

by a first order approximation and omitting the term of order $O(\alpha^2)$.

2.7 Plan

The mathematical analysis of (2.1),(2.3),(2.9),(2.14) is somewhat classical, so we shall focus on the problem with the nonlinear boundary condition (2.1),(2.3),(2.13),(2.14). Then, at the end we shall argue that there is no essential new difficulty if the term $\partial_t c$ is added, namely problem (2.1),(2.3),(2.15),(2.14).

3 Variational formulation

3.1 Notations

For convenience, C , C' and C_i , $i = 1, 2, 3, \dots$, denote generic constants independent of \mathbf{u} and \mathbf{c} . We denote $d = 2, 3$ the dimension.

We use the standard notations: $f^+ = \max\{f, 0\}$ and $f^- = -\min\{f, 0\}$.

We denote by $\|\cdot\|_s$ the norm of $H^s(\Omega)$ and by $\|\cdot\|_{s,\Gamma}$ the norm of $H^s(\Gamma)$ for $\Gamma \subset \partial\Omega$.

If B is a Banach space, B' denotes its dual space. The $L^2(\Omega)$ inner product is (\cdot, \cdot) and the duality product between B and B' is $\langle \cdot, \cdot \rangle_{B, B'}$.

We define

$$W = \{w \in H^1(\Omega) : w|_{\Gamma_{in}} = 0\};$$

since W is closed in $H^1(\Omega)$ and $H^1(\Omega)$ is a Hilbert space, then so is W .

We assume that c is such that $\mathbf{u} \in L^2(0, T; H^1(\Omega)^d) \cap L^\infty(0, T; L^2(\Omega)^d)$, In variational form (2.3), (2.4), (2.13) is:

Problem (P)

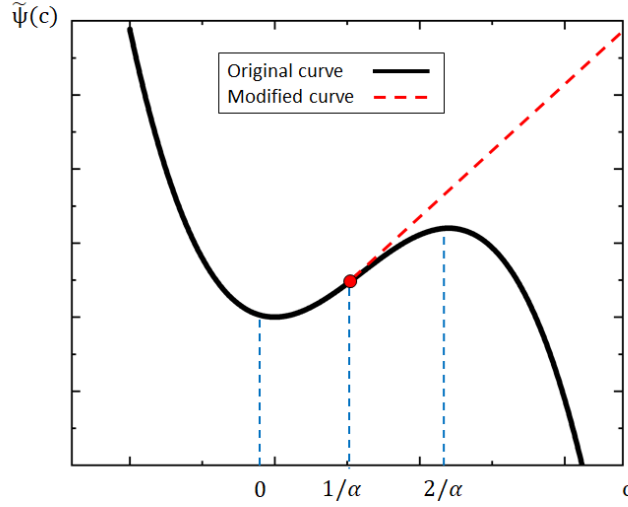


Figure 2: Graphic showing the modification of the nonconvex function $c \mapsto \frac{c^2}{2} - \frac{\alpha c^3}{6}$ into a convex one.

Find $c \in L^2(0, T; H^1(\Omega))$ with $c(0) = c_0$, satisfying $c|_{\Gamma_{in}} = c_{in}$, $\partial_t c \in W'$, such that, for all $w \in W$,

$$\begin{aligned} & \langle \partial_t c, w \rangle_{W', W} + \int_{\Omega} D \nabla c \cdot \nabla w + \frac{1}{2} \int_{\Omega} [(\mathbf{u} \cdot \nabla c)w - (\mathbf{u} \cdot \nabla w)c] \\ & + \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u} \cdot \mathbf{n})cw + \int_S \left(1 - \frac{\alpha c}{2} + \frac{\alpha i_0^2}{D} \int_0^t c(s)ds\right) i_0 cw = 0. \end{aligned} \quad (3.1)$$

Clearly, the previous expression for the convection term is consistent; it is used here to facilitate the derivation of energy estimates.

$$\int_{\Omega} (\mathbf{u} \cdot \nabla c)w = \frac{1}{2} \int_{\Omega} [(\mathbf{u} \cdot \nabla c)w - (\mathbf{u} \cdot \nabla w)c] + \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u} \cdot \mathbf{n})cw - \int_S \frac{\alpha c^2}{2}w. \quad (3.2)$$

3.2 Convexification

The term $c - \frac{\alpha c^2}{2}$ in the integral on S is problematic because it is not monotone so it makes the problem non-coercive. Indeed its primitive $\psi(c) := \frac{c^2}{2} - \frac{\alpha c^3}{6}$ is nonconvex beyond $c > \frac{1}{\alpha}$. But the physics require that $c \in [0, 1]$ and the maximum principle will insure it. So any modification of ψ outside $(0, 1)$ will not affect the solution; hence to work with a convex potential let us replace ψ by (see Figure 2).

$$\bar{\psi}(c) = \begin{cases} \frac{c^2}{2} - \frac{\alpha c^3}{6} & \text{if } c < \frac{1}{\alpha}, \\ \frac{2}{\alpha}c - \frac{1}{6\alpha^2} & \text{otherwise.} \end{cases} \quad (3.3)$$

and define

$$\phi(c) = i_0 \bar{\psi}_{,c}(c) + \frac{\alpha i_0^3}{D} c \int_0^t |c(s)| ds, \quad (3.4)$$

where $\bar{\psi}_{,c}(c)$ is the derivative of $\bar{\psi}$ with respect to c :

$$\bar{\psi}_{,c}(c) = \begin{cases} c - \frac{\alpha c^2}{2} & \text{if } c < \frac{1}{\alpha}, \\ \frac{1}{2\alpha} & \text{otherwise.} \end{cases} \quad (3.5)$$

Naturally $\bar{\psi}_{,c}$ is monotone increasing. Note that $\bar{\psi}$ is strictly convex and $\bar{\psi}'_{,c}$ is strictly increasing in $[0, \frac{1}{\alpha}]$.

The convexified variational formulation replacing (3.1) is

Problem (P_c)

Find $c \in H^1(\Omega)$ satisfying $c|_{\Gamma_{in}} = c_{in}$, $\partial_t c \in W'$, and $\phi(c) \in L^2(S)$

$$\begin{aligned} \langle \partial_t c, w \rangle_{W', W} + \int_{\Omega} D \nabla c \cdot \nabla w + \frac{1}{2} \int_{\Omega} [(\mathbf{u} \cdot \nabla c)w - (\mathbf{u} \cdot \nabla w)c] \\ + \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u} \cdot \mathbf{n})cw + \int_S \phi(c)w = 0, \quad \forall w \in W. \end{aligned} \quad (3.6)$$

Note that when $0 \leq c \leq 1$, then both c and $\phi(c) \in L^\infty(\Omega \times (0, T))$.

The proof of existence goes by steps. We assume that $\mathbf{u} \in L^2(0, T; H^1(\Omega)^d) \cap L^\infty(0, T; L^2(\Omega)^d)$, so as to focus on the equation for c with \mathbf{u} given. We will first discretize in time, show existence for the time discretized problem and then let the time step tend to zero.

4 Existence for the Time-discretized Problem

Problem (P_c^m)

Let $N \in \mathbb{N}^+$ and let $\delta t = \frac{T}{N}$ be the time step. For each integer $m \in (0, \frac{T}{\delta t})$ find $c^{m+1} \in H^1(\Omega)$ such that for all $w \in W$,

$$\begin{aligned} \int_{\Omega} \frac{c^{m+1} - c^m}{\delta t} w + D \int_{\Omega} \nabla c^{m+1} \cdot \nabla w \\ + \frac{1}{2} \int_{\Omega} [(\mathbf{u}^{m+1} \cdot \nabla c^{m+1})w - (\mathbf{u}^{m+1} \cdot \nabla w)c^{m+1}] \\ + \int_S \phi_m(c^{m+1})w + \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n})c^{m+1}w = 0 \end{aligned} \quad (4.1)$$

where $\phi_m(c^{m+1})$ is the following time approximation of $\phi(c)$,

$$\phi_m(c^{m+1}) = i_0 \bar{\psi}_{,c}(c^{m+1}) + \frac{\alpha i_0^3}{D} \left(\sum_{j=0}^m c^j \delta t \right) c^{m+1}.$$

The initial value is $c^0 = c_0$ with $c_0 \in H^1(\Omega)$, $c_0|_{\Gamma_{in}} = c_{in}$.

4.1 Existence of the Solution to the Time-discretized Problem (P_c^m)

To prove the existence, the Minty-Browder Theorem will be used.

Theorem 1 (Minty-Browder) Let B be a reflexive Banach space and $\|\cdot\|$ its norm. Let $A : B \rightarrow B'$ a continuous mapping such that

$$(i) \quad \langle Au - Av, u - v \rangle > 0 \quad \forall u, v \in B, \quad u \neq v$$

$$(ii) \quad \lim_{\|u\| \rightarrow \infty} \|u\|^{-1} \langle Au, u \rangle = +\infty.$$

Then, for any $b \in B'$, there is a unique u such that $Au = b$.

The theorem will be applied to $c^m - \tilde{c}_{in}$ where $\tilde{c}_{in} \in H^1(\Omega)$ is a lift of the boundary conditions defined as the unique solution of

$$\int_{\Omega} D \nabla \tilde{c}_{in} \cdot \nabla w = 0, \quad \forall w \in W \quad (4.2)$$

such that $\tilde{c}_{in} = c_{in}$ on Γ_{in} .

Lemma 1 If $0 \leq c_{in} \leq 1$, then \tilde{c}_{in} satisfies $0 \leq \tilde{c}_{in} \leq 1$ a.e.

Proof. The argument is classical; let us recall it for the reader's convenience. Note that $(\tilde{c}_{in} - 1)^+$ and $(\tilde{c}_{in})^-$ belong to W . Choosing $w = (\tilde{c}_{in})^-$ in (4.2), gives $\|\nabla(\tilde{c}_{in})^-\|_0^2 = 0$. Hence $(\tilde{c}_{in})^- = 0$ i.e. $\tilde{c}_{in} \geq 0$.

Now choosing $w = (\tilde{c}_{in} - 1)^+$ in (4.2) implies $\|\nabla(\tilde{c}_{in} - 1)^+\|_0^2 = 0$. Hence $(\tilde{c}_{in} - 1)^+ = 0$, i.e. $\tilde{c}_{in} \leq 1$ a.e. in Ω . Q.E.D.

Proposition 1 Let $m \geq 0$. We suppose that $0 \leq c^j \leq 1$ a.e. in Ω for all $j \leq m$, then $0 \leq c^{m+1} \leq 1$ a.e. in Ω .

Proof. Letting $w = (c^{m+1})^-$ in (4.1), gives

$$\begin{aligned} & -\frac{1}{\delta t} \|(c^{m+1})^-\|_0^2 - D \|\nabla(c^{m+1})^-\|_0^2 - \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n})(c^{m+1})^{-2} \\ & + \int_S i_0 \bar{\psi}_{,c}(c^{m+1})(c^{m+1})^- - \frac{\alpha i_0^3}{D} \sum_{j=0}^m c^j \delta t (c^{m+1})^{-2} = \frac{1}{\delta t} \int_{\Omega} c^m (c^{m+1})^- \geq 0 \end{aligned} \quad (4.3)$$

All terms on the left are obviously negative except $\bar{\psi}_{,c}(c^{m+1})(c^{m+1})^-$. Two cases: if $c \geq \frac{1}{\alpha}$ then $c^- = 0$ and $\bar{\psi}_{,c}(c)c^- = 0$; if $c < \frac{1}{\alpha}$ then $\bar{\psi}_{,c}(c)c^- = (c - \frac{\alpha}{2}c^2)c^- = -(c^-)^2 - \frac{\alpha}{2}c^2c^- \leq 0$. Hence $\bar{\psi}_{,c}(c^{m+1})(c^{m+1})^- \leq 0$ always; thus (4.3) leads to $\|(c^{m+1})^-\|_0^2 = 0$.

Define $\bar{c}^{m+1} = c^{m+1} - 1$. It satisfies

$$\begin{aligned} & \int_{\Omega} \frac{\bar{c}^{m+1} - \bar{c}^m}{\delta t} w + \int_{\Omega} D \nabla \bar{c}^{m+1} \cdot \nabla w \\ & + \frac{1}{2} \int_{\Omega} [(\mathbf{u}^{m+1} \cdot \nabla \bar{c}^{m+1})w - (\mathbf{u}^{m+1} \cdot \nabla w)\bar{c}^{m+1}] \\ & - \frac{1}{2} \int_{S \cup \Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n})w + \int_S i_0 \bar{\psi}_{,c}(c^{m+1})w + \int_S \frac{\alpha i_0^3}{D} \sum_{j=0}^m c^j \delta t c^{m+1} w \\ & + \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n})(\bar{c}^{m+1} + 1)w = 0 \end{aligned} \quad (4.4)$$

Testing with $w = (\bar{c}^{m+1})^+$, gives

$$\begin{aligned} & \frac{1}{\delta t} \|(\bar{c}^{m+1})^+\|_0^2 - \frac{1}{\delta t} \int_{\Omega} \bar{c}^m (\bar{c}^{m+1})^+ + D \|\nabla (\bar{c}^{m+1})^+\|_0^2 - \frac{1}{2} \int_S (\mathbf{u}^{m+1} \cdot \mathbf{n}) (\bar{c}^{m+1})^+ \\ & \int_S i_0 \bar{\psi}_{,c}(c^{m+1}) (\bar{c}^{m+1})^+ + \frac{\alpha i_0^3}{D} \int_S \sum_{j=0}^m c^j \delta t c^{m+1} (\bar{c}^{m+1})^+ \\ & + \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n}) ((\bar{c}^{m+1})^+ + 1) (\bar{c}^{m+1})^+ = 0. \end{aligned} \quad (4.5)$$

By the induction hypothesis $\bar{c}^m \leq 0$; observe that $\mathbf{u}^{m+1} \cdot \mathbf{n} \leq 0$ on S , $\mathbf{u}^{m+1} \cdot \mathbf{n} \geq 0$ on Γ_{out} and $\bar{\psi}_{,c}(c^{m+1}) \geq 0$ because $c^{m+1} \geq 0$. So we have that $\|(\bar{c}^{m+1})^+\|_0^2 = 0$. Q.E.D.

Let us define

$$\tilde{c}^m := c^m - \tilde{c}_{in} \in W, \quad \forall m \in \mathbb{N} \cap [0, \frac{T}{\delta t}].$$

Remark 1 As $\tilde{c}^m \in [-1, 1]$ and $\bar{\psi}_{,c}(c^m) \in [0, 1]$, therefore $|\bar{\psi}_{,c}(c^m) \tilde{c}^m| \leq 1$.

By construction, (4.1), which defines Problem P_c^m , can be rewritten as

Problem (\tilde{P}_c^m)

Find $\tilde{c}^{m+1} \in W$ such that, for all $w \in W$,

$$\begin{aligned} & \frac{1}{\delta t} \int_{\Omega} \tilde{c}^{m+1} w + \frac{1}{2} \int_{\Omega} [(\mathbf{u}^{m+1} \cdot \nabla \tilde{c}^{m+1}) w - (\mathbf{u}^{m+1} \cdot \nabla w) \tilde{c}^{m+1}] \\ & + D \int_{\Omega} \nabla \tilde{c}^{m+1} \cdot \nabla w + \int_S \phi_m(c^{m+1}) w + \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n}) \tilde{c}^{m+1} w \\ & = -\frac{1}{2} \int_{\Omega} [(\mathbf{u}^{m+1} \cdot \nabla \tilde{c}_{in}) w - (\mathbf{u}^{m+1} \cdot \nabla w) \tilde{c}_{in}] + \frac{1}{\delta t} \int_{\Omega} \tilde{c}^m w - \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n}) \tilde{c}_{in} w \end{aligned} \quad (4.6)$$

Now define the mapping $A : W \rightarrow W'$ by

$$\begin{aligned} \langle A\rho, w \rangle &= \frac{1}{\delta t} \int_{\Omega} \rho w + \frac{1}{2} \int_{\Omega} [(\mathbf{u}^{m+1} \cdot \nabla \rho) w - (\mathbf{u}^{m+1} \cdot \nabla w) \rho] \\ & + D \int_{\Omega} \nabla \rho \cdot \nabla w + \int_S \phi_m(\rho + \tilde{c}_{in}) w + \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n}) \rho w \end{aligned} \quad (4.7)$$

Since W is closed in $H^1(\Omega)$ and $H^1(\Omega)$ is a Hilbert space, then so is W .

Lemma 2 $A : W \rightarrow W'$ defined by (4.7) is Lipschitz continuous.

The proof is fairly straightforward but long, so it is postponed to Appendix A so as not to break the thread of the proof of existence of (\tilde{P}_c^m) . From the definition of A by (4.7), there is no essential difficulty to arrive, via a sequence of inequalities, at

$$\begin{aligned} & |\langle A\rho_1 - A\rho_2, w \rangle| \\ & \leq \left(C_1 + C_2 \|\mathbf{u}^{m+1}\|_1 + C_3 (\|\rho_1\|_1 + \|\rho_2\|_1) + C_4 \delta t \sum_{j=0}^m \|c^j\|_1 \right) \|\rho_1 - \rho_2\|_1 \|w\|_1, \end{aligned} \quad (4.8)$$

Lemma 3 Let $\rho_1, \rho_2, \rho \in W$. $A : W \rightarrow W'$ defined by (4.7) satisfies

(i) $\langle A\rho_1 - A\rho_2, \rho_1 - \rho_2 \rangle > 0$ if $\rho_1 \neq \rho_2$

(ii) $\lim_{\|\rho\|_1 \rightarrow +\infty} \|\rho\|_1^{-1} \langle A\rho, \rho \rangle = +\infty$.

Proof. To show (i), we use (4.7) with $\rho_1, \rho_2 \in W$,

$$\begin{aligned} \langle A\rho_1 - A\rho_2, w \rangle &= \frac{1}{\delta t} \int_{\Omega} (\rho_1 - \rho_2) w \\ &+ \frac{1}{2} \int_{\Omega} [(\mathbf{u}^{m+1} \cdot \nabla(\rho_1 - \rho_2))w - (\mathbf{u}^{m+1} \cdot \nabla w)(\rho_1 - \rho_2)] + D \int_{\Omega} \nabla(\rho_1 - \rho_2) \cdot \nabla w \\ &+ \int_S (\phi_m(\rho_1 + \tilde{c}_{in}) - \phi_m(\rho_2 + \tilde{c}_{in}))w + \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n})(\rho_1 - \rho_2)w \end{aligned} \quad (4.9)$$

and let $w = \rho_1 - \rho_2$ to obtain

$$\begin{aligned} \langle A\rho_1 - A\rho_2, \rho_1 - \rho_2 \rangle &= \\ &\int_{\Omega} \frac{1}{\delta t} (\rho_1 - \rho_2)^2 + D \int_{\Omega} |\nabla(\rho_1 - \rho_2)|^2 + \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n})(\rho_1 - \rho_2)^2 \\ &\int_S \frac{\alpha_0^3}{D} \sum_{j=0}^m c^j \delta t (\rho_1 - \rho_2)^2 + \int_S (\bar{\psi}_{,c}(\rho_1 + \tilde{c}_{in}) - \bar{\psi}_{,c}(\rho_2 + \tilde{c}_{in}))(\rho_1 - \rho_2). \end{aligned} \quad (4.10)$$

Recall that $\mathbf{u}^{m+1} \cdot \mathbf{n} \geq 0$ on Γ_{out} . All the terms on the right are obviously positive, except the last one. Without loss of generality, we assume that $\rho_2 > \rho_1$; we know that $\bar{\psi}_{,c}$ is strictly increasing. That is, $\bar{\psi}_{,c}(\rho_2 + \tilde{c}_{in}) > \bar{\psi}_{,c}(\rho_1 + \tilde{c}_{in})$. Hence $(\bar{\psi}_{,c}(\rho_2 + \tilde{c}_{in}) - \bar{\psi}_{,c}(\rho_1 + \tilde{c}_{in}))(\rho_2 - \rho_1) > 0$. Hence

$$\langle A\rho_1 - A\rho_2, \rho_1 - \rho_2 \rangle > \frac{1}{\delta t} \|\rho_1 - \rho_2\|_0^2 + D \|\nabla(\rho_1 - \rho_2)\|_0^2.$$

Finally (ii) can be proved by taking $\rho_1 = 0$ in (i). Q.E.D.

By Theorem 1 and Lemmas 2, 3, we have

Corollary 1 There exists a unique solution to Problem (\tilde{P}_c^m) and hence also to (P_c^m) defined by (4.1).

5 Stability of the Time-discretized Problem P_c^m

Proposition 2 The following estimates hold for Problem (\tilde{P}_c^m) ,

$$\begin{aligned} \|\tilde{c}^{m+1}\|_0^2 + \|\tilde{c}^{m+1} - \tilde{c}^m\|_0^2 + D\delta t \|\nabla \tilde{c}^{m+1}\|_0^2 + \delta t \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n})(\tilde{c}^{m+1})^2 \\ \leq \|\tilde{c}^m\|_0^2 + C_1 \delta t + C_2 \delta t \|\mathbf{u}^{m+1}\|_0^2 + C_3 \delta t \|\nabla \tilde{c}_{in}\|_0^2 \end{aligned} \quad (5.1)$$

Proof. By (4.6) with $w = \tilde{c}^{m+1}$ we have

$$\begin{aligned}
& \frac{1}{2\delta t} \|\tilde{c}^{m+1}\|_0^2 + \frac{1}{2\delta t} \|\tilde{c}^{m+1} - \tilde{c}^m\|_0^2 + D \|\nabla \tilde{c}^{m+1}\|_0^2 + \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n})(\tilde{c}^{m+1})^2 \\
&= - \int_S i_0 \bar{\psi}_{,c}(c^{m+1}) \tilde{c}^{m+1} - \int_S \frac{\alpha i_0^3}{D} \sum_{j=0}^m c^j \delta t c^{m+1} \tilde{c}^{m+1} - \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n}) \tilde{c}_{in} \tilde{c}^{m+1} \\
& - \frac{1}{2} \int_{\Omega} (\mathbf{u}^{m+1} \cdot \nabla \tilde{c}^{m+1}) \tilde{c}_{in} + \frac{1}{2} \int_{\Omega} (\mathbf{u}^{m+1} \cdot \nabla \tilde{c}_{in}) \tilde{c}^{m+1} + \frac{1}{2\delta t} \|\tilde{c}^m\|_0^2
\end{aligned} \tag{5.2}$$

We estimate each term of the right hand side

$$\begin{aligned}
& - \int_S i_0 \bar{\psi}_{,c}(c^{m+1}) \tilde{c}^{m+1} \leq i_0 \int_S |\bar{\psi}_{,c}(c^{m+1}) \tilde{c}^{m+1}| \leq i_0 |S|. \\
& - \int_S \frac{\alpha i_0^3}{D} \left(\sum_{j=0}^m c^j \delta t \right) c^{m+1} \tilde{c}^{m+1} \leq \frac{\alpha i_0^3}{D} \int_S \left(\sum_{j=0}^m c^j \delta t \right) c^{m+1} |\tilde{c}^{m+1}| \leq \frac{\alpha i_0^3}{D} \int_S m \delta t \leq \frac{\alpha i_0^3 T}{D} |S|. \\
& - \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n}) \tilde{c}_{in} \tilde{c}^{m+1} \leq |\Gamma_{out}|^{\frac{1}{2}} \left(\int_{\Gamma_{out}} |\mathbf{u}^{m+1}|^2 \right)^{\frac{1}{2}} \leq C \|\mathbf{u}^{m+1}\|_1^2. \\
& - \int_{\Omega} (\mathbf{u}^{m+1} \cdot \nabla \tilde{c}^{m+1}) \tilde{c}_{in} \leq \int_{\Omega} |\mathbf{u}^{m+1} \cdot \nabla \tilde{c}^{m+1}| \leq \frac{1}{2D} \|\mathbf{u}^{m+1}\|_0^2 + \frac{D}{2} \|\nabla \tilde{c}^{m+1}\|_0^2. \\
& \int_{\Omega} (\mathbf{u}^{m+1} \cdot \nabla \tilde{c}_{in}) \tilde{c}^{m+1} \leq \int_{\Omega} |\mathbf{u}^{m+1} \cdot \nabla \tilde{c}_{in}| \leq \frac{1}{2D} \|\mathbf{u}^{m+1}\|_0^2 + \frac{D}{2} \|\nabla \tilde{c}_{in}\|_0^2.
\end{aligned}$$

Collecting all terms leads to

$$\begin{aligned}
& \frac{1}{2\delta t} \|\tilde{c}^{m+1}\|_0^2 + \frac{1}{2\delta t} \|\tilde{c}^{m+1} - \tilde{c}^m\|_0^2 + \frac{D}{2} \|\nabla \tilde{c}^{m+1}\|_0^2 + \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n})(\tilde{c}^{m+1})^2 \\
& \leq \frac{1}{2\delta t} \|\tilde{c}^m\|_0^2 + C_1 + C_2 \|\mathbf{u}^{m+1}\|_1^2 + C_3 \|\nabla \tilde{c}_{in}\|_0^2
\end{aligned} \tag{5.3}$$

Multiplying both sides by $2\delta t$ completes the proof. Q.E.D.

Summing (5.1) from 0 to m , leads to the following:

Corollary 2

$$\begin{aligned}
& \|\tilde{c}^{m+1}\|_0^2 + \sum_{j=0}^m \|\tilde{c}^{j+1} - \tilde{c}^j\|_0^2 + D \delta t \sum_{j=0}^m \|\nabla \tilde{c}^{j+1}\|_0^2 + \sum_{j=0}^m \delta t \int_{\Gamma_{out}} (\mathbf{u}^{j+1} \cdot \mathbf{n})(\tilde{c}^{j+1})^2 \\
& \leq \|\tilde{c}^0\|_0^2 + C_1 T + C_2 \delta t \sum_{j=0}^m \|\mathbf{u}^{j+1}\|_0^2 + C_3 T \|\nabla \tilde{c}_{in}\|_0^2.
\end{aligned} \tag{5.4}$$

Proposition 3

$$\delta t \sum_{j=1}^m \left\| \frac{\tilde{c}^{j+1} - \tilde{c}^j}{\delta t} \right\|_{W'}^2 \text{ is uniformly bounded.} \tag{5.5}$$

Proof. By definition

$$\left\| \frac{\tilde{c}^{m+1} - \tilde{c}^m}{\delta t} \right\|_{W'} = \sup_{w_0 \in W} \frac{1}{\|w_0\|_1} \left\langle \frac{\tilde{c}^{m+1} - \tilde{c}^m}{\delta t}, w_0 \right\rangle. \quad (5.6)$$

By (4.6), with $w = \frac{w_0}{\|w_0\|_1} \in W$,

$$\begin{aligned} \left\| \frac{\tilde{c}^{m+1} - \tilde{c}^m}{\delta t} \right\|_{W'} &= \sup_{w \in W, \|w\|=1} \left\{ - \int_{\Omega} D \nabla \tilde{c}^{m+1} \cdot \nabla w \right. \\ &- \frac{1}{2} \int_{\Omega} [(\mathbf{u}^{m+1} \cdot \nabla \tilde{c}^{m+1})w - (\mathbf{u}^{m+1} \cdot \nabla w)\tilde{c}^{m+1}] - i_0 \int_S \bar{\psi}_{,c}(c^{m+1})w \\ &- \int_S \frac{\alpha j_0^3}{D} \left(\sum_{j=0}^m c^j \delta t \right) c^{m+1}w - \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n})c^{m+1}w \\ &\left. - \frac{1}{2} \int_{\Omega} [(\mathbf{u}^{m+1} \cdot \nabla \tilde{c}_{in})w - (\mathbf{u}^{m+1} \cdot \nabla w)\tilde{c}_{in}] \right\}. \end{aligned} \quad (5.7)$$

We estimate all terms on the right hand side of (5.7)

$$-D \int_{\Omega} \nabla \tilde{c}^{m+1} \cdot \nabla w \leq D \|\nabla \tilde{c}^{m+1}\|_0 \|\nabla w\|_0 \leq D \|\tilde{c}^{m+1}\|_1 \|w\|_1 = D \|\tilde{c}^{m+1}\|_1, \quad (5.8)$$

$$\int_{\Omega} (\mathbf{u}^{m+1} \cdot \nabla w)\tilde{c}^{m+1} \leq \int_{\Omega} |\mathbf{u}^{m+1} \cdot \nabla w| \leq \|\mathbf{u}^{m+1}\|_0, \quad (5.9)$$

$$\begin{aligned} - \int_{\Omega} (\mathbf{u}^{m+1} \cdot \nabla \tilde{c}^{m+1})w &= \int_{\Omega} (\mathbf{u}^{m+1} \cdot \nabla w)\tilde{c}^{m+1} - \int_{\partial\Omega} (\mathbf{u}^{m+1} \cdot \mathbf{n})\tilde{c}^{m+1}w \\ &\leq \|\mathbf{u}^{m+1}\|_0 + \|\mathbf{u}^{m+1}\|_{\partial\Omega} \|w\|_{\partial\Omega} \leq C \|\mathbf{u}^{m+1}\|_1, \end{aligned} \quad (5.10)$$

$$- \int_S \bar{\psi}_{,c}(c^{m+1})w \leq C|S|^{\frac{1}{2}} \|w\|_1 = C|S|^{\frac{1}{2}}, \quad (5.11)$$

$$- \int_S \left(\sum_{j=0}^m c^j \delta t \right) c^{m+1}w \leq \int_S T|w| \leq CT|S|^{\frac{1}{2}} \|w\|_1 = CT|S|^{\frac{1}{2}}, \quad (5.12)$$

$$- \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n})c^{m+1}w \leq \|\mathbf{u}^{m+1}\|_{\Gamma_{out}} \|w\|_{\Gamma_{out}} \leq C \|\mathbf{u}^{m+1}\|_1 \|w\|_1 = C \|\mathbf{u}^{m+1}\|_1, \quad (5.13)$$

$$\int_{\Omega} (\mathbf{u}^{m+1} \cdot \nabla w)\tilde{c}_{in} \leq \int_{\Omega} |\mathbf{u}^{m+1} \cdot \nabla w| \leq \|\mathbf{u}^{m+1}\|_0 \|w\|_1 = \|\mathbf{u}^{m+1}\|_0, \quad (5.14)$$

$$- \int_{\Omega} (\mathbf{u}^{m+1} \cdot \nabla \tilde{c}_{in})w \leq C \|\mathbf{u}^{m+1}\|_1 \quad (\text{see (5.10)}). \quad (5.15)$$

Collecting (5.8)-(5.15) with (5.7), all multiplied by δt , gives

$$\delta t \left\| \frac{\tilde{c}^{m+1} - \tilde{c}^m}{\delta t} \right\|_{W'}^2 \leq C \delta t (1 + \|\tilde{c}^{m+1}\|_1^2 + \|\mathbf{u}^{m+1}\|_1^2). \quad (5.16)$$

By summing (5.16) from 0 to $\frac{T}{\delta t}$ and the boundedness given by Corollary 2, the proof is completed. Q.E.D.

6 Passage to the Limit $\delta t \rightarrow 0$

Let us define

$$c_\delta : [0, T] \rightarrow H^1(\Omega), \quad c_\delta(t) = c^j \quad \text{if } t \in ((j-1)\delta t, j\delta t], \quad (6.1)$$

$$\mathbf{u}_\delta : [0, T] \rightarrow H^1(\Omega)^d, \quad \mathbf{u}_\delta(t) = \mathbf{u}^j \quad \text{if } t \in ((j-1)\delta t, j\delta t], \quad (6.2)$$

$$c_h : [0, T] \rightarrow H^1(\Omega), \quad c_h(t) = \frac{t - (j-1)\delta t}{\delta t} c^j + \frac{j\delta t - t}{\delta t} c^{j-1} \quad \text{if } t \in ((j-1)\delta t, j\delta t], \quad (6.3)$$

$$c_{\delta-} : [0, T] \rightarrow H^1(\Omega), \quad c_{\delta-}(t) = c^{j-1} \quad \text{if } t \in [(j-1)\delta t, j\delta t), \quad (6.4)$$

$$C_{\delta-} : [0, T] \rightarrow H^1(\Omega), \quad C_{\delta-}(t) = \sum_{k=1}^j c^{k-1} \delta t \quad \text{if } t \in [(j-1)\delta t, j\delta t), \quad (6.5)$$

for $j = 1, \dots, N$. Note that c_δ , c_h , and $c_{\delta-}$ are in $L^2(0, T; H^1(\Omega))$ and $L^\infty(\Omega \times (0, T))$. With these notations, problem (P_c^m) reads

$$\begin{aligned} & (\partial_t c_h, w) + \frac{1}{2} [(\mathbf{u}_\delta \cdot \nabla c_\delta, w) - (\mathbf{u}_\delta \cdot \nabla w, c_\delta)] \\ & + D(\nabla c_\delta, \nabla w) + \int_S i_0 \bar{\psi}_{,c}(c_\delta) w + \int_S \frac{\alpha i_0^3}{D} c_\delta C_{\delta-} w + \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u}_\delta \cdot \mathbf{n}) c_\delta w = 0. \end{aligned} \quad (6.6)$$

Lemma 4 $C_{\delta-}$ is in $L^2(0, T; H^1(\Omega))$.

Proof.

$$\begin{aligned} & \int_0^T \int_\Omega |\partial_{x_i} C_{\delta-}(t)|^2 dx dt \leq \int_0^T \int_\Omega \left(\delta t \sum_{k=1}^N |\partial_{x_i} c^{k-1}| \right)^2 dx dt \\ & = \int_0^T \int_\Omega \delta t^2 \left(\sum_{k=1}^N |\partial_{x_i} c^{k-1}| \right)^2 dx dt \leq \int_0^T \delta t^2 \cdot \frac{T}{\delta t} \sum_{k=1}^N |\partial_{x_i} c^{k-1}|^2 dx \\ & = T^2 \delta t \sum_{k=1}^N \int_\Omega |\partial_{x_i} c^{k-1}|^2 dx \leq C \end{aligned}$$

The last inequality is due to Corollary 2.

Lemma 5

$$\|c_\delta - c_h\|_{L^2((0, T) \times \Omega)} \leq \sqrt{\frac{\delta t}{3}} \sum_{j=1}^N \|c^{j+1} - c^j\|_0^2 \quad (6.7)$$

Proof.

$$\begin{aligned} c_\delta(t) - c_h(t) &= \frac{t - j\delta t}{\delta t} (c^j - c^{j-1}) \quad \text{for } (j-1)\delta t < t \leq j\delta t, \\ \int_{(j-1)\delta t}^{j\delta t} \|c_\delta(t) - c_h(t)\|_0^2 dt &= \frac{\delta t}{3} \|c^j - c^{j-1}\|_0^2. \end{aligned}$$

The proof can be completed by taking summation from $j = 1$ to $T/\delta t$.

Corollary 3

$$c_\delta - c_h \rightarrow 0 \quad \text{in } L^2((0, T) \times \Omega) \quad \text{as } \delta t \rightarrow 0. \quad (6.8)$$

By the boundedness given by Proposition 1, 3, and Corollary 2, there are subsequences of c_δ and c_h (still denoted by c_δ and c_h), respectively such that

$$c_\delta \rightarrow c \quad \text{in } L^2(0, T; H^1(\Omega)) \quad \text{weakly,} \quad (6.9)$$

$$c_h \rightarrow c_* \quad \text{in } L^2(0, T; H^1(\Omega)) \quad \text{weakly,} \quad (6.10)$$

$$c_h \rightarrow c_* \quad \text{in } L^\infty(\Omega \times (0, T)) \quad \text{weakly,} \quad (6.11)$$

$$\partial_t c_h \rightarrow g \quad \text{in } L^2(0, T; W') \quad \text{weakly.} \quad (6.12)$$

By Corollary 3, we have $c = c_*$.

By a classical argument, see for instance [13], we have

$$g = \partial_t c. \quad (6.13)$$

Let

$$Y = \left\{ w \in L^2(0, T; H^1(\Omega)), \partial_t w \in L^2(0, T; W') \right\}.$$

By the Aubin-Lions Lemma, Y is compactly embedded in $L^2(0, T; L^q(\Omega))$ with $q < 6$ when $d = 3$ and $q < \infty$ when $d = 2$. Therefore, we have in particular

$$c_h \rightarrow c \quad \text{in } L^2((0, T) \times \Omega) \quad \text{strongly.} \quad (6.14)$$

Using Corollary 3 again, we get

$$c_\delta \rightarrow c \quad \text{in } L^2((0, T) \times \Omega) \quad \text{strongly.} \quad (6.15)$$

To see the convergence of the boundary term, we need the following lemma:

Lemma 6 Given c_δ defined by (6.1), there exists a subsequence (still denoted by c_δ) satisfying (6.9) and the followings:

$$\overline{\psi}_{,c}(c_\delta) \rightarrow \overline{\psi}_{,c}(c) \quad \text{in } L^2(0, T; H^{\frac{1}{2}}(S)) \quad \text{weakly,} \quad (6.16)$$

$$c_\delta \int_0^t c_\delta(s) ds \rightarrow c \int_0^t c(s) ds \quad \text{in } L^2(0, T; H^{\frac{1}{2}}(S)) \quad \text{weakly.} \quad (6.17)$$

Proof. Let us prove that $\overline{\psi}_{,c}(c_\delta)$ tends to $\overline{\psi}_{,c}(c)$ weakly in $L^2(0, T; H^1(\Omega))$. First, we know that $\psi_{,c}(c_\delta)$ is bounded in $\Omega \times [0, T]$ and we have

$$\int_0^T \int_\Omega |\partial_{x_i} \overline{\psi}_{,c}(c_\delta)|^2 dx dt \leq \int_0^T \int_\Omega |\partial_{x_i} c_\delta|^2 dx dt < \infty.$$

Therefore, $\overline{\psi}_{,c}$ converges weakly in $L^2(0, T; H^1(\Omega))$. To identify its limit, let $w \in C([0, T] \times \overline{\Omega})$. Then (6.17) implies that

$$\int_0^T \int_\Omega \overline{\psi}_{,c}(c_\delta) w dx dt \rightarrow \int_0^T \int_\Omega \overline{\psi}_{,c} w dx dt$$

and (6.17) and (6.9) imply that

$$\int_0^T \int_{\Omega} \partial_{x_i} \bar{\psi}_{,c}(c_{\delta}) w dx dt \rightarrow \int_0^T \int_{\Omega} \partial_{x_i} \bar{\psi}_{,c}(c) w dx dt.$$

This gives the desired convergence. By the continuity of the trace mapping

$$\phi \mapsto \phi|_{\partial\Omega}$$

for the weak topology, we deduce that

$$\bar{\psi}_{,c}(c_{\delta}) \rightarrow \bar{\psi}_{,c}(c) \text{ weakly in } L^2(0, T; H^{\frac{1}{2}}(S)). \quad (6.18)$$

For (6.17), we define $r(t, c_{\delta}) := c_{\delta} \int_0^t c_{\delta}(s) ds$ in $(0, T) \times \Omega$. To begin with, we observe that $\int_0^t c_{\delta}(s) ds \rightarrow \int_0^t c(s) ds$ strongly in $L^2((0, T) \times \Omega)$. This can be checked by the estimate:

$$\begin{aligned} \int_0^T \int_{\Omega} \left| \int_0^t (c_{\delta}(s) - c(s)) ds \right|^2 dx dt &\leq \int_0^T \int_{\Omega} t \int_0^t |c_{\delta}(s) - c(s)|^2 ds dx dt \\ &\leq \int_0^T t \int_0^T \int_{\Omega} |c_{\delta}(s) - c(s)|^2 dx ds dt = \frac{1}{2} T^2 \int_0^T \int_{\Omega} |c_{\delta}(s) - c(s)|^2 dx ds \\ &\rightarrow 0 \text{ as } \delta t \rightarrow 0. \end{aligned}$$

Let $w \in C([0, T] \times \bar{\Omega})$. Since c_{δ} strongly converges to c in $L^2((0, T) \times \Omega)$, we have

$$\int_0^T \int_{\Omega} r(t, c_{\delta}) w dx dt \rightarrow \int_0^T \int_{\Omega} r(t, c) w dx dt. \quad (6.19)$$

We differentiate $r(t, c_{\delta})$:

$$\partial_{x_i} r(t, c_{\delta}) = \partial_{x_i} c_{\delta} \int_0^t c_{\delta}(s) ds + c_{\delta} \int_0^t \partial_{x_i} c_{\delta}(s) ds. \quad (6.20)$$

We have the boundedness for the first term on the right hand side:

$$\begin{aligned} \int_0^T \int_{\Omega} |\partial_{x_i} c_{\delta}|^2 \left| \int_0^t c_{\delta}(s) ds \right|^2 dx dt &\leq \int_0^T \int_{\Omega} t |\partial_{x_i} c_{\delta}|^2 dx dt \\ &\leq T \int_0^T \int_{\Omega} |\partial_{x_i} c_{\delta}|^2 dx dt < \infty \end{aligned} \quad (6.21)$$

Using the fact that both $\partial_{x_i} c_{\delta} \rightarrow \partial_{x_i} c$ weakly in $L^2((0, T) \times \Omega)$ and $\int_0^t c_{\delta}(s) ds \rightarrow \int_0^t c(s) ds$ strongly in $L^2((0, T) \times \Omega)$, we have

$$\int_0^T \int_{\Omega} \partial_{x_i} c_{\delta} \left(\int_0^t c_{\delta}(s) ds \right) w dx dt \rightarrow \int_0^T \int_{\Omega} \partial_{x_i} c \left(\int_0^t c(s) ds \right) w dx dt. \quad (6.22)$$

The second term is bounded as well:

$$\begin{aligned}
& \int_0^T \int_{\Omega} \left| \int_0^t \partial_{x_i} c_{\delta}(s) ds \right|^2 dx dt \leq \int_0^T \int_{\Omega} t \int_0^t |\partial_{x_i} c_{\delta}(s)|^2 ds dx dt \\
& = \int_0^T t \int_0^t \int_{\Omega} |\partial_{x_i} c_{\delta}(s)|^2 dx ds dt \leq \int_0^T t \int_0^T \int_{\Omega} |\partial_{x_i} c_{\delta}(s)|^2 dx ds dt \\
& = \frac{1}{2} T^2 \int_0^T \int_{\Omega} |\partial_{x_i} c_{\delta}(s)|^2 dx dt < \infty
\end{aligned} \tag{6.23}$$

And we observe that

$$\begin{aligned}
& \int_0^T \int_{\Omega} \left(\int_0^t \partial_{x_i} c_{\delta}(s) ds \right) w dx dt \\
& = \int_0^T \int_0^t \left(\int_{\Omega} \partial_{x_i} c_{\delta}(s) w dx \right) ds dt \rightarrow \int_0^T \left(\int_0^t \int_{\Omega} \partial_{x_i} c(s) w dx ds \right) dt
\end{aligned}$$

because $w(t)$ for fixed t is continuous in Ω . Now, we have $c_{\delta} \rightarrow c$ in $L^2((0, T) \times \Omega)$ strongly and $\int_0^t \partial_{x_i} c_{\delta}(s) ds \rightarrow \int_0^t \partial_{x_i} c(s) ds$ weakly in $L^2((0, T) \times \Omega)$. This implies that

$$\int_0^T \int_{\Omega} c_{\delta} \left(\int_0^t \partial_{x_i} c_{\delta}(s) ds \right) w(t) dx dt \rightarrow \int_0^T \int_{\Omega} c \left(\int_0^t \partial_{x_i} c(s) ds \right) w dx dt$$

By (6.23) and Lemma 7 below, we have $c_{\delta} \int_0^t \partial_{x_i} c_{\delta}(s) ds \rightarrow c \int_0^t \partial_{x_i} c(s) ds$ weakly in $L^2((0, T) \times \Omega)$. Collecting all the weak convergence results above, we conclude that $r(t, c_{\delta}) \rightarrow r(t, c)$ in $L^2(0, T; H^1(\Omega))$ weakly. By the continuity of the trace for the weak topology, we have $r(t, c_{\delta}) \rightarrow r(t, c)$ in $L^2(0, T; H^{\frac{1}{2}}(S))$ weakly. Q.E.D.

Lemma 7 Let X be a Banach space, D a dense subset of X' , $x_n, n = 1, 2, \dots$ the bounded sequence in X . If $g(x_n) \rightarrow g(x)$ for all $g \in D$, then $x_n \rightarrow x$ weakly in X .

Proof. If x_n is a bounded sequence in X , it is an equicontinuous sequence as a sequence of functions $X' \rightarrow \mathbb{R}$. And x_n is pointwisely convergent on D by the hypothesis. Using Lemma ??, x_n is pointwisely convergent. Since x_n is bounded in norm by the hypothesis, we conclude that x_n is weakly convergent to x in X . Q.E.D.

Lemma 8 $C_{\delta-}$ defined in (6.5) satisfies

$$\left\| C_{\delta-} - \int_0^t c_{\delta}(s) ds \right\|_{L^2(0, T; H^1(\Omega))} \leq C \delta t \tag{6.24}$$

Proof. For all $t \in [(j-1)\delta t, j\delta t)$,

$$\begin{aligned}
C_{\delta-}(t) - \int_0^t c_{\delta}(s) ds & = \sum_{k=1}^j c^{k-1} \delta t - \sum_{k=1}^{j-1} c^k \delta t - c^j (t - (j-1)\delta t) \\
& = c^0 \delta t - c^j (t - (j-1)\delta t).
\end{aligned}$$

But for $t \in [(j-1)\delta t, j\delta t)$, we have $0 \leq t - (j-1)\delta t \leq \delta t$. Therefore

$$\left| C_{\delta-}(t) - \int_0^t c_\delta(s) ds \right| \leq \delta t |c^0 + c^j|.$$

On the other hand,

$$\begin{aligned} \partial_{x_i} C_{\delta-}(t) - \partial_{x_i} \int_0^t c_\delta(s) ds &= \partial_{x_i} C_{\delta-}(t) - \int_0^t \partial_{x_i} c_\delta(s) ds \\ &= \partial_{x_i} c^0 \delta t - \partial_{x_i} c^j (t - (j-1)\delta t). \end{aligned}$$

Hence, we have

$$\left| \partial_{x_i} \left(C_{\delta-}(t) - \int_0^t c_\delta(s) ds \right) \right| \leq \delta t |\partial_{x_i} (c^0 + c^j)|.$$

Therefore,

$$\begin{aligned} \left\| C_{\delta-} - \int_0^t c_\delta(s) ds \right\|_{L^2(0,T;H^1(\Omega))}^2 &= \int_0^T \left\| C_{\delta t-}(t) - \int_0^t c_\delta(s) ds \right\|_1^2 \\ &\leq \sum_{j=1}^N \int_{(j-1)\delta t}^{j\delta t} (\delta t)^2 \|c^0 + c^j\|_1^2 \\ &\leq 2 \sum_{j=1}^N \int_{(j-1)\delta t}^{j\delta t} (\delta t)^2 (\|c^0\|_1^2 + \|c^j\|_1^2) \\ &\leq 2\|c^0\|_1^2 (\delta t)^2 + 2(\delta t)^2 \sum_{j=1}^N \delta t \|c^j\|_1^2 \\ &\leq C(\delta t)^2 \end{aligned}$$

Q.E.D.

Now, we are in a position to pass to the limit in (6.6). Take any $w = v(x)\lambda(t)$, where $v \in W \cap W^{1,\infty}(\Omega)$ and $\lambda \in W_0^{1,\infty}(0, T)$. Then

$$\begin{aligned} & - \int_0^T (c_h, v) \lambda'(t) dt + \frac{1}{2} \int_0^T [(\mathbf{u}_\delta \cdot \nabla c_\delta, v) - (\mathbf{u}_\delta \cdot \nabla v, c_\delta)] \lambda dt \\ & + \int_0^T D(\nabla c_\delta, \nabla v) \lambda dt + \int_0^T \int_S i_0 \bar{\psi}_{,c}(c_\delta) v \lambda(t) dt + \int_0^T \int_S \frac{\alpha_0^3}{D} c_\delta C_{\delta-} v \lambda(t) dt \\ & + \frac{1}{2} \int_0^T \int_{\Gamma_{out}} (\mathbf{u}_\delta \cdot \mathbf{n}) c_\delta v \lambda(t) dt = 0. \end{aligned}$$

Since $\mathbf{u}_\delta \rightarrow \mathbf{u}$ strongly in $L^2(\Omega \times (0, T))$ and $\nabla c_\delta \rightarrow \nabla c$ weakly in $L^2(\Omega \times (0, T))$, the regularity of v and λ implies that

$$- \int_0^T (c_h, v) \lambda'(t) dt \rightarrow - \int_0^T (c, v) \lambda'(t) dt = \int_0^T \langle \partial_t c, v \rangle_{W', W} \lambda dt, \quad (6.25)$$

$$\frac{1}{2} \int_0^T [(\mathbf{u}_\delta \cdot \nabla c_\delta, v) - (\mathbf{u}_\delta \cdot \nabla v, c_\delta)] \lambda dt \rightarrow \frac{1}{2} \int_0^T [(\mathbf{u} \cdot \nabla c, v) - (\mathbf{u} \cdot \nabla v, c)] \lambda dt, \quad (6.26)$$

$$\int_0^T D(\nabla c_\delta, \nabla c) \lambda dt \rightarrow \int_0^T D(\nabla c, \nabla v) \lambda dt. \quad (6.27)$$

Similarly, the weak convergence of $\bar{\psi}_{,c}(c_\delta)$ to $\bar{\psi}_{,c}(c)$ in $L^2(0, T; H^{\frac{1}{2}}(S))$ implies that

$$\int_0^T \int_S i_0 \bar{\psi}_{,c}(c_\delta) v \lambda(t) dt \rightarrow \int_0^T \int_S \bar{\psi}_{,c}(c) v \lambda(t) dt, \quad (6.28)$$

and the weak convergence of $c_\delta C_{\delta-}$ to c in $L^2(0, T; H^{\frac{1}{2}}(S))$ implies that

$$\int_0^T \int_S c_\delta C_{\delta-} v \lambda(t) dt \rightarrow \int_0^T \int_S c \left(\int_0^t c(s) ds \right) v \lambda(t) dt. \quad (6.29)$$

Finally, we consider the last term $\frac{1}{2} \int_0^T \int_{\Gamma_{out}} (\mathbf{u}_\delta \cdot \mathbf{n}) c_\delta v \lambda(t)$. The assumption on \mathbf{u}_δ are: $\mathbf{u}_\delta \rightarrow \mathbf{u}$ weakly in $L^2(0, T; H^1(\Omega)^d)$ and $\mathbf{u}_\delta \rightarrow \mathbf{u}$ strongly in $L^2(0, T; L^4(\Omega)^d)$ (thanks to a sharper result than (6.15), $L^2(\Omega)$ can be replaced by any space compactly embedded into $H^1(\Omega)^d$. Here we take $L^4(\Omega)^d$, which is compatible with $d = 3$; the exponent has to be strictly less than 6). We use Green's formula:

$$\int_0^T \int_{\Gamma_{out}} (\mathbf{u}_\delta \cdot \mathbf{n}) c_\delta v \lambda = \int_0^T \int_\Omega \nabla \cdot (\mathbf{u}_\delta c_\delta) v \lambda + \int_0^T \int_\Omega (c_\delta \mathbf{u}_\delta \cdot \nabla v) \lambda \quad (6.30)$$

for all $\lambda \in L^\infty(0, T)$ and for all smooth v that vanish on $\partial\Omega \setminus \Gamma_{out}$. It suffices to prove the convergence of each term to the desired limit.

1) For all $v \in L^4(\Omega)$ and for all $\lambda \in L^\infty(0, T)$:

$$\begin{aligned} & \int_0^T \int_\Omega \nabla \cdot (\mathbf{u}_\delta c_\delta) v \lambda = \int_0^T \int_\Omega \mathbf{u}_\delta \cdot \nabla c_\delta v \lambda \\ & = \int_0^T \int_\Omega (\mathbf{u}_\delta - \mathbf{u}) \cdot \nabla c_\delta v \lambda + \int_0^T \int_\Omega \mathbf{u} \cdot \nabla c_\delta v \lambda \\ & \leq \|\mathbf{u}_\delta - \mathbf{u}\|_{L^2(0, T; L^4(\Omega))} \|\nabla c_\delta\|_{L^2(\Omega \times (0, T))} \|\lambda\|_{L^\infty(0, T)} \|v\|_{L^4(\Omega)} + \int_0^T \int_\Omega \mathbf{u} \cdot \nabla c_\delta v \lambda. \end{aligned}$$

It is noted that $\mathbf{u} v \lambda \in L^2(0, T; L^2(\Omega)^d)$ and $\nabla c_\delta \rightarrow \nabla c$ weakly in $L^2(0, T; L^2(\Omega)^d)$, we have

$$\int_0^T \int_\Omega \nabla \cdot (\mathbf{u}_\delta c_\delta) v \lambda \rightarrow \int_0^T \int_\Omega (\mathbf{u} \cdot \nabla c) v \lambda = \int_0^T \int_\Omega \nabla \cdot (\mathbf{u} c) v \lambda. \quad (6.31)$$

2) For all $v \in H^1(\Omega)$, $\lambda \in L^\infty(\Omega)$

$$\begin{aligned} & \int_0^T \int_\Omega (c_\delta \mathbf{u}_\delta \cdot \nabla v) \lambda = \int_0^T \int_\Omega c_\delta (\mathbf{u}_\delta - \mathbf{u}) \cdot \nabla v \lambda + \int_0^T \int_\Omega \mathbf{u} (c_\delta - c) \cdot \nabla v \lambda + \int_0^T \int_\Omega c (\mathbf{u} \cdot \nabla v) \lambda \\ & \leq \|\mathbf{u}_\delta - \mathbf{u}\|_{L^2(0, T; L^2(\Omega)^d)} \|c_\delta\|_{L^\infty(\Omega \times (0, T))} \|\nabla v\|_0 \|\lambda\|_{L^\infty(\Omega)} \\ & + \|\mathbf{u}\|_{L^2(0, T; L^4(\Omega)^d)} \|c_\delta - c\|_{L^2(0, T; L^4(\Omega)^d)} \|\nabla v\|_0 \|\lambda\|_{L^\infty(\Omega)} + \int_0^T \int_\Omega c (\mathbf{u} \cdot \nabla v) \lambda. \end{aligned}$$

Therefore

$$\begin{aligned}
& \int_0^T \langle \partial_t c, v \rangle_{W', W} \lambda dt + \frac{1}{2} \int_0^T ((\mathbf{u} \cdot \nabla c, v) - (\mathbf{u} \cdot \nabla v, c)) \lambda dt \\
& + \int_0^T D(\nabla c, \nabla v) \lambda dt + \int_0^T \int_S i_0 \bar{\psi}_{,c}(c) v \lambda dt + \int_0^T \int_S \frac{\alpha i_0^3}{D} c \left(\int_0^t d(s) ds \right) v \lambda dt \\
& + \frac{1}{2} \int_0^T \int_{\Gamma_{out}} (\mathbf{u} \cdot \mathbf{n}) c v \lambda dt = 0
\end{aligned} \tag{6.32}$$

for all $\lambda \in W_0^{1,\infty}(0, T)$ and for all $v \in W^{1,\infty}(\Omega)$. This gives the equations a.e. in $(0, T)$.

To recover the initial condition, we take $\lambda \in W^{1,\infty}(0, T)$, $\lambda(T) = 0$, $\lambda(0) \neq 0$, and $v \in W^{1,\infty}(\Omega)$.

We consider (6.6) such that all terms are identical except the first:

$$\int_0^T (\partial_t c_h, v) \lambda dt = \int_0^T \partial_t (c_h, v) \lambda dt = - \int_0^T (c_h, v) \lambda'(t) - (c^0, v) \lambda(0). \tag{6.33}$$

When passing to the limit, we obtain

$$\begin{aligned}
& \int_0^T \langle \partial_t c, v \rangle_{W', W} \lambda dt = - \int_0^T (c, v) \lambda'(t) - (c^0, v) \lambda(0) \\
& = \int_0^T \frac{d}{dt} (c, v) \lambda + (c(0), v) \lambda(0) - (c^0, v) \lambda(0) \\
& = \int_0^T \langle \partial_t c, v \rangle_{W', W} \lambda dt + (c(0), v) \lambda(0) - (c^0, v) \lambda(0).
\end{aligned} \tag{6.34}$$

Therefore

$$(c(0), v) = (c^0, v), \quad \forall v \in W \cap W^{1,\infty}(\Omega).$$

This implies that $c(0) = c^0$.

We may conclude the above result by the theorem:

Theorem 2 There exists $c \in L^2(0, T; H^1(\Omega))$ with $\partial_t c \in L^2(0, T; W')$ such that

$$\begin{aligned}
& \int_0^T \langle \partial_t c, w \rangle_{W', W} dt + \frac{1}{2} \left(\int_0^T \int_{\Omega} [(\mathbf{u} \cdot \nabla c) w - (\mathbf{u} \cdot \nabla w) c] dx dt \right) \\
& + D \int_0^T \int_{\Omega} \nabla c \cdot \nabla w dx dt + \int_0^T \int_S i_0 \bar{\psi}_{,c}(c) w dt \\
& + \int_0^T \int_S \frac{\alpha i_0^3}{D} c \left(\int_0^t c(s) ds \right) w dt + \frac{1}{2} \int_0^T \int_{\Gamma_{out}} (\mathbf{u} \cdot \mathbf{n}) c w dt \\
& = 0.
\end{aligned} \tag{6.35}$$

for all $w \in L^2(0, T; W)$. Moreover, c satisfies the initial condition $c(0) = c_0$ and the boundary condition $c = c_{in}$ on Γ_{in} .

Corollary 4 Problem (P_c) has a unique solution and because it satisfies $0 \leq c \leq 1$, it is also the solution of (P) .

6.1 On the boundary condition (2.15) which contains $\partial_t c$

To prove existence a similar strategy is taken: $\partial_t c$ is replaced by $(c^{m+1} - c^m)/\delta t$, existence is shown and then convergence when $\delta t \rightarrow 0$.

The proof of existence of the time-discretized problem is exactly the same but with ϕ redefined as

$$\phi_m(c^{m+1}) = i_0 \bar{\psi}_{,c}(c^{m+1}) + \alpha i_0 \left(\frac{i_0^2}{D} + \frac{1}{\delta t} \right) \left(\sum_{j=0}^m c^j \delta t \right) c^{m+1}.$$

Convergence with $\delta t \rightarrow 0$ requires more regularity, which can be obtained from the PDE differentiated in time.

7 Numerical Simulations

The rectangular domain of size $0.025 \text{ mm} \times 0.005 \text{ mm}$ is the initial physical domain. The electro-process is simulated up to time $T = 5000$.

7.1 Scalings

The simulation will be done with dimensionless variables. Let L, C and U be representative length, concentration and velocity of the physical system. Then it is easy to see that the dimensionless equation for c is the same as the original equation but with $\tilde{D}/(LU)$ instead of D , where \tilde{D} is the physical molecular diffusion. Similarly because (2.5) becomes

$$-D \frac{\partial c}{\partial n} = \frac{i_0}{U} c, \quad \mathbf{u} = -\alpha C \frac{i_0}{U} \mathbf{c} \mathbf{n},$$

the original form holds but with i_0/U redefined as i_0 and αC redefined as α .

It is well known that the dimensionalized Navier-Stokes equation has the inverse Reynolds number $\tilde{\nu}/(UL)$ redefined as ν , where $\tilde{\nu}$ being the kinematic viscosity.

The parameters of nickel ion given in [7] are $i_0 = \tilde{i}_0/(zF)$ with $\tilde{i}_0 = 0.001 \text{ A} \cdot \text{mm}^{-2}$, the number of electrons involves in the reaction $z = 2$ and the Faraday constant $F = 96487 \text{ s} \cdot \text{A} \cdot \text{mol}^{-1}$, $C = 3 \times 10^{-7} \text{ mol} \cdot \text{mm}^{-3}$.

For electrodeless plating we may take $U = 1 \text{ mm} \cdot \text{s}^{-1}$, $L = 0.005 \text{ mm}$, $\tilde{D} = 1 \times 10^{-4} \text{ mm}^2 \cdot \text{s}^{-1}$, $\alpha = 6590 \text{ mm}^3 \cdot \text{mol}^{-1}$, and $\tilde{\nu} = 1.2 \text{ mm}^2 \cdot \text{s}^{-1}$.

So in the end the numerical tests are done on a rescaled domain $\Omega = (0, 5) \times (0, 1)$ with

$$\alpha = 0.002085, \quad i_0 = 0.017273, \quad D = 0.02, \quad \nu = 240,$$

7.2 Numerical algorithm

The finite element method is used for spacial discretization. We use the P^1 element for Problem (P_c^m) which defines c^m . So let W_h denotes the space of piecewise affine continuous functions over a triangulation of Ω which are *zero* on Γ_{in} . Then one must solve the finite dimensional nonlinear

Problem (P_h^m) defined to be (P_c^m) with W_h instead of W in (4.1). Denote by $\{c_h^m\}_{m \geq 1}$ the solution. Find $c_h^{m+1} \in C_h$ satisfying

$$\begin{aligned} & \int_{\Omega} \frac{c_h^{m+1} - c_h^m}{\delta t} w_h dx + \frac{1}{2} \int_{\Omega} [(\mathbf{u}_h^{m+1} \cdot \nabla c_h^{m+1}) w_h - (\mathbf{u}_h^{m+1} \cdot \nabla w_h) c_h^{m+1}] dx \\ & + D \int_{\Omega} \nabla c_h^{m+1} \cdot \nabla w_h dx + \int_S \phi_m(c_h^{m+1}) w_h + \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u}_h^{m+1} \cdot \mathbf{n}) c_h^{m+1} w_h = 0, \end{aligned} \quad (7.1)$$

for all $w_h \in C_h$.

For the Navier-Stokes equation we use the P^2/P^1 Taylor-Hood element [14] and we denote by \mathbf{u}_h^m , p_h^m the finite element solution and by V_h , Q_h the corresponding finite element space. The variational formulation is: Find $\mathbf{u}_h^{m+1} \in V_h$ and $p_h^{m+1} \in P_h$ satisfying

$$\begin{aligned} & \int_{\Omega} \frac{\mathbf{u}_h^{m+1} - \mathbf{u}_h^m}{\delta t} \cdot \mathbf{v}_h + \int_{\Omega} [(\mathbf{u}_h^m \cdot \nabla) \mathbf{u}_h^{m+1}] \cdot \mathbf{v}_h + \nu \int_{\Omega} \nabla \mathbf{u}_h : \nabla \mathbf{v}_h \\ & - \int_{\Omega} (p_h \nabla \cdot \mathbf{v}_h + q_h \nabla \cdot \mathbf{u}_h + \epsilon p_h q_h) = 0, \quad \mathbf{u}_h^{m+1} = u_{in} \text{ on } \Gamma_{in}, \end{aligned} \quad (7.2)$$

for all $\mathbf{v}_h \in V_h$ and $q_h \in P_h$; ϵ is a small regularization parameter.

The coupled system (7.1)-(7.2) is solved iteratively. We replace c_h^{m+1} by c^* in (7.1) and solve (7.2). We denote the solution by \mathbf{u}^* . Then we replace \mathbf{u}_h^{m+1} by \mathbf{u}^* in order to get the new c^* , until $\|\mathbf{u}_{new}^* - \mathbf{u}_{old}^*\|_0 + \|c_{new}^* - c_{old}^*\|_0$ is sufficiently small.

To validate the method we need to compare with the original free boundary problem. It is solved with a similar iterative fixed point like process but the mesh needs to be rebuilt when the free boundary is updated. It is done by a scaling on y -coordinate at each time step t^j : $y \mapsto (1 - \alpha i_0 c_h^j \delta t) y$.

```

Data:  $\mathbf{u}_h^m, p_h^m, c_h^m$ , and  $y$ 
1 Set initial data  $\mathbf{u}_0, c_0$ ;
2 for  $m$  do
3    $c^* = c_h^m$ ;
4   while  $\|\mathbf{u}_{new}^* - \mathbf{u}_{old}^*\|_0 + \|c_{new}^* - c_{old}^*\|_0 \geq tolerance$  do
5     Solve (7.1) to get  $c_{new}^*$ ;
6     Solve (7.2) to get  $\mathbf{u}_{new}^*$  and  $p_{new}^*$ ;
7   end
8    $c_h^{m+1} = c_{new}^*$ ;
9    $\mathbf{u}_h^{m+1} = \mathbf{u}_{new}^*$ ;
10  For the free boundary case change the mesh by  $y \leftarrow (1 - \alpha i_0 c_h^{m+1} \delta t) y$ ;
11 end

```

7.3 Numerical results at low Reynolds number

The initial and inflow values are

$$c_0 = \cos(\lambda y), \quad \mathbf{u}_0 = y(1 - y); \quad c_{in} = c_0|_{\Gamma_{in}} \exp(-D\lambda^2 t), \quad \mathbf{u}_{in} = \mathbf{u}_0|_{\Gamma_{in}}.$$

A uniform triangular mesh 150×30 for the initial domain.

δt	L^2 error	δt	H^1 error	δt	L^2 error	δt	H^1 error
0.16	1.01723×10^{-5}	0.16	1.03365×10^{-5}	0.16	1.01722×10^{-5}	0.16	1.03365×10^{-5}
0.08	4.74704×10^{-6}	0.08	4.82368×10^{-6}	0.08	4.74701×10^{-6}	0.08	4.82367×10^{-6}
0.04	2.03444×10^{-6}	0.04	2.06729×10^{-6}	0.04	2.03443×10^{-6}	0.04	2.06729×10^{-6}
0.02	6.78147×10^{-7}	0.02	6.89107×10^{-7}	0.02	6.78142×10^{-7}	0.02	6.89103×10^{-7}

Table 1: Convergence when $\delta t \rightarrow 0$: L^2 and H^1 relative error at $T = 100$ for the scheme with the nonlinear transpiration approximation and $\nu = 240$ (left columns) and $\nu = 0.01$ (right columns). A uniform triangular mesh 150×30 is used.; $c_{\delta t=0.01}$ is used as reference solution.

We compare the results obtained using a time dependent domain (Figure 3a) with the results using a fixed domain and the linear condition (2.5) (see Figure 3b) and finally with the nonlinear condition (2.10) (see Figure 3c).

On Figure 4 the free boundary and the reconstructed free boundaries are displayed using η given by (2.6).

The convergence with respect to time step size is shown on Table 1, computed at an intermediate time $T = 100$. Since no exact solution is available, the numerical solution with time step $\delta t = 0.01$ is taken as the reference solution. The numerical results in Table 1 show a first order convergence in L^2 error conformed with the estimates given in Appendix B (see Figure 5). The weak first order convergence of H^1 error is also proved in Appendix B. However the numerical results show strong first order H^1 convergence for this test problem (see Figure 6).

7.4 Numerical results at larger Reynolds number

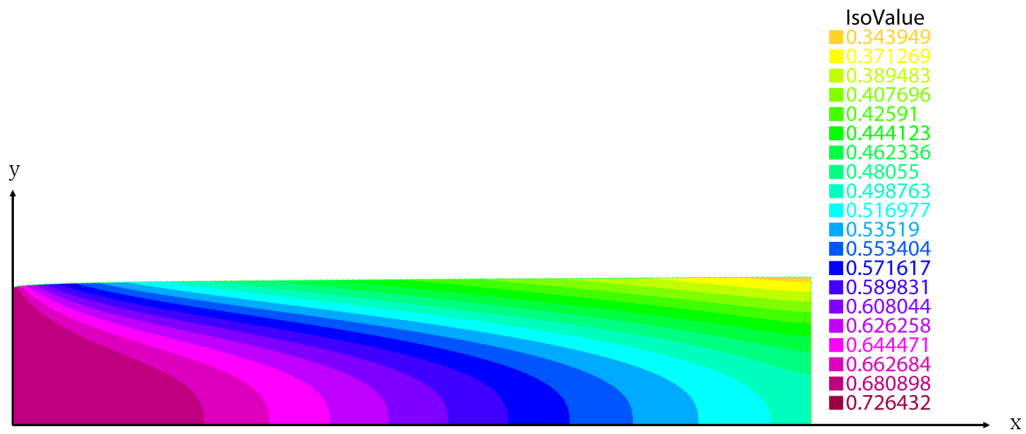
In the previous example, where the values of the parameters correspond to the physical design of [6], we could have neglected the inertial terms and work with the Stokes approximation. In order to validate the algorithm at higher Reynolds number, which may be the case for other plating problems, we keep all parameters given in the end of Section 7.1 but change the Reynolds number to the inverse of $\nu = 0.01$. The same experiments are conducted as in Section 7.3. The numerical results obtained for the low Reynolds number and the larger Reynolds number are very similar; no visible changes can be seen (see the right side of Figure 4) so we do not display the plots of Figure 3 for the high Reynolds number case.

7.5 Influence of the term $\partial_t c$ in (2.15)

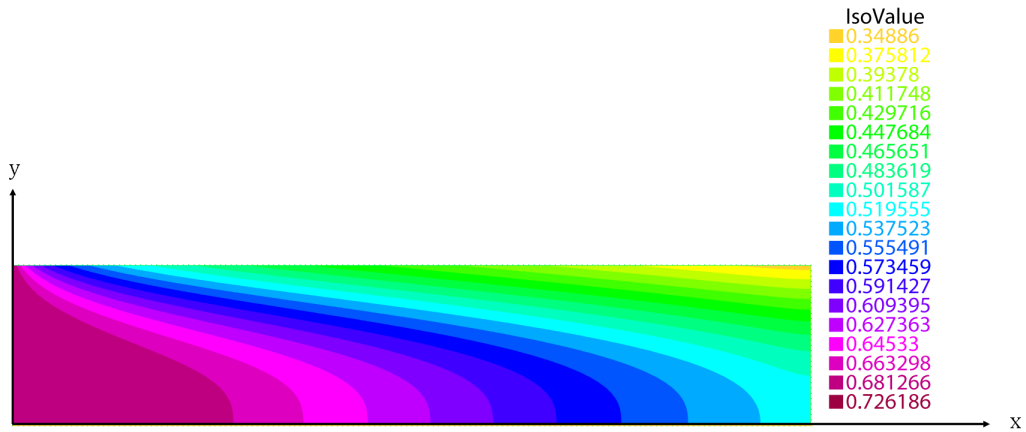
For the geometry considered in these numerical test no visible difference could be observed between (2.10) and (2.15) .

8 Conclusion

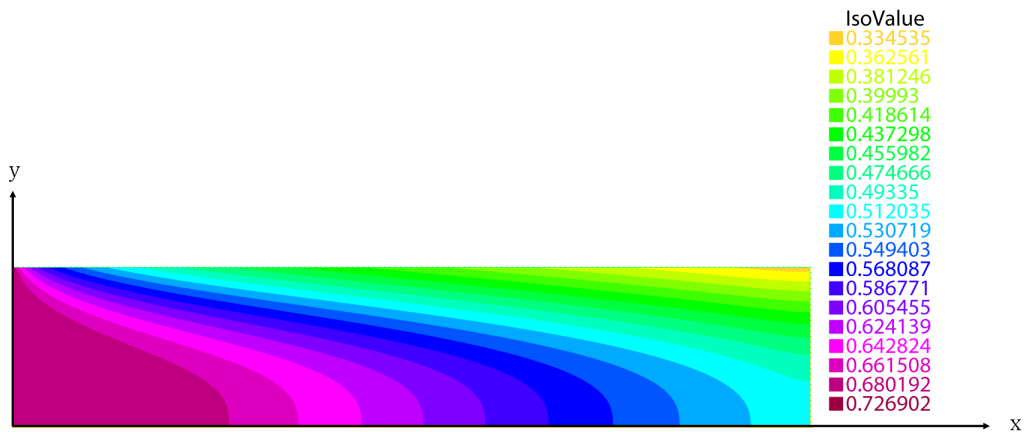
We have proposed a simplified model which approximates the Electroless process of [6] by replacing the time dependent domain occupied by the reacting chemical by a fixed domain using a transpiration approximation. We have validated the approximation numerically with a finite element method in space and a fully implicit in time approximation. We have constructed an existence



(a) Intensity map of c computed with a free boundary on a moving mesh.



(b) Intensity map of c computed by the linear transpiration approximation.



(c) Intensity map of c computed by nonlinear transpiration approximation.

Figure 3: The solution profiles of numerical experiments with $\nu = 240$.

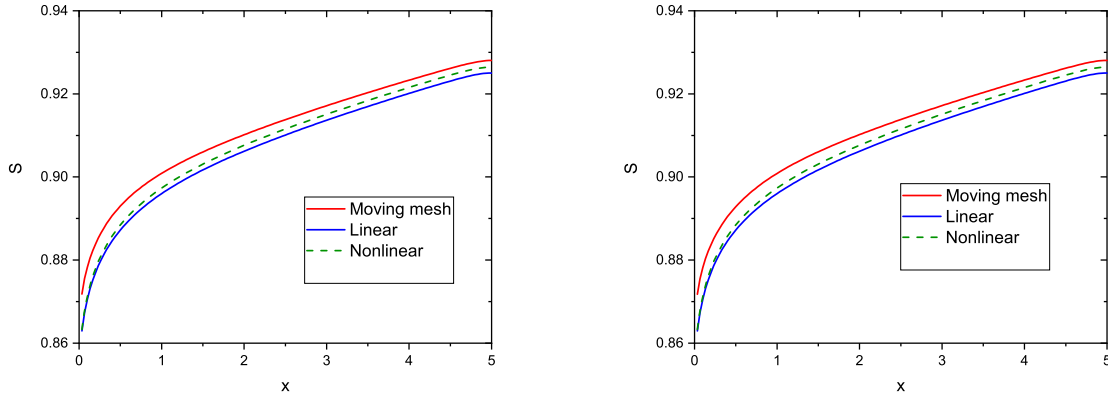


Figure 4: $S(T)$ calculated by 3 experiments at $T = 5000$. The red curve is the height of $S(T)$ computed by moving mesh. The blue curve is computed by the displacement $\eta(T)$ with linear condition. The green dash curve is computed by the displacement $\eta(T)$ with nonlinear condition. If the curve of moving mesh is regarded as the reference solution, it is easy to see that the nonlinear approximation does better than the linear approximation. Left figure corresponds to with $\nu = 240$ and Right figure to $\nu = 0.01$.

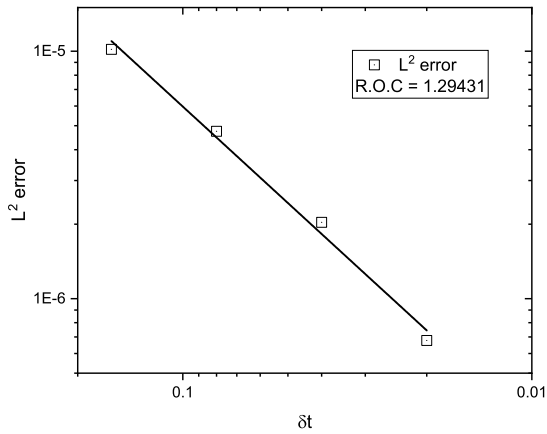


Figure 5: L^2 relative error versus δt at $T = 100$

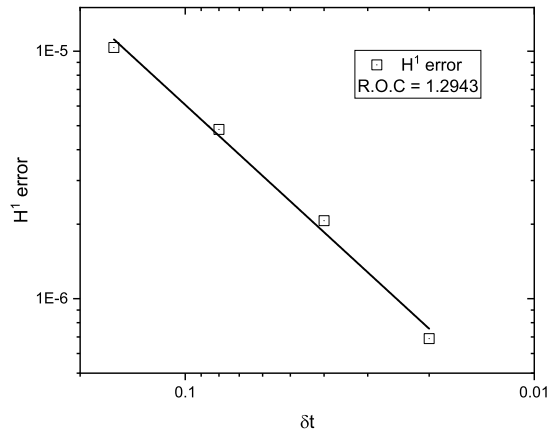


Figure 6: H^1 relative error versus δt at $T = 100$.

proof by using variational convex analysis or fixed point arguments. The proof is technical and long because the nonlinearity is on the boundary condition and because it required a convexification of the energy potential and the maximum principle. However it was worth the effort because it gives a stable ground for the numerical studies and it may be useful for other similar problems. We plan to extend this study to two phase flows to take into account the formation of bubbles.

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A Proof of Lemma 2.

Let $\rho_1, \rho_2 \in W$. Formula (4.7) gives

$$\begin{aligned} \langle A\rho_1 - A\rho_2, w \rangle &= \frac{1}{\delta t} \int_{\Omega} (\rho_1 - \rho_2) w dx \\ &+ \frac{1}{2} \int_{\Omega} [(\mathbf{u}^{m+1} \cdot \nabla(\rho_1 - \rho_2))w - (\mathbf{u}^{m+1} \cdot \nabla w)(\rho_1 - \rho_2)] dx + D \int_{\Omega} \nabla(\rho_1 - \rho_2) \cdot \nabla w dx \\ &+ \int_S (\phi_m(\rho_1 + \tilde{c}_{in}) - \phi_m(\rho_2 + \tilde{c}_{in}))w + \frac{1}{2} \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n})(\rho_1 - \rho_2)w \end{aligned} \quad (\text{A.1})$$

We estimate each term on the right hand side of (A.1):

$$\frac{1}{\delta t} \int_{\Omega} (\rho_1 - \rho_2) w dx \leq \frac{1}{\delta t} \|\rho_1 - \rho_2\|_0 \|w\|_0 \leq \frac{1}{\delta t} \|\rho_1 - \rho_2\|_1 \|w\|_1 \quad (\text{A.2})$$

$$\begin{aligned} \int_{\Omega} (\mathbf{u}^{m+1} \cdot \nabla(\rho_1 - \rho_2))w dx &\leq \|\mathbf{u}^{m+1}\|_{L^4(\Omega)} \|\nabla(\rho_1 - \rho_2)\|_0 \|w\|_{L^4(\Omega)} \\ &\leq C \|\mathbf{u}^{m+1}\|_1 \|\rho_1 - \rho_2\|_1 \|w\|_1 \end{aligned} \quad (\text{A.3})$$

$$\begin{aligned} \int_{\Omega} (\mathbf{u}^{m+1} \cdot \nabla w)(\rho_1 - \rho_2) dx &\leq \|\mathbf{u}^{m+1}\|_{L^4(\Omega)} \|\nabla w\|_0 \|\rho_1 - \rho_2\|_{L^4(\Omega)} \\ &\leq C \|\mathbf{u}^{m+1}\|_1 \|\rho_1 - \rho_2\|_1 \|w\|_1 \end{aligned} \quad (\text{A.4})$$

$$\int_{\Omega} \nabla(\rho_1 - \rho_2) \cdot \nabla w dx \leq \|\nabla(\rho_1 - \rho_2)\|_0 \|\nabla w\|_0 \leq \|\rho_1 - \rho_2\|_1 \|w\|_1 \quad (\text{A.5})$$

Let $x_1, x_2 \in \mathbb{R}$. If $x_1 + \tilde{c}_{in}$, and $x_2 + \tilde{c}_{in} > \frac{1}{2\alpha}$, then $|\bar{\psi}_{,c}(x_1 + \tilde{c}_{in}) - \bar{\psi}_{,c}(x_2 + \tilde{c}_{in})| = 0$. If $x_1 + \tilde{c}_{in} \leq \frac{1}{2\alpha}$ and $x_2 + \tilde{c}_{in} > \frac{1}{2\alpha}$, then

$$\begin{aligned} |\bar{\psi}_{,c}(x_1 + \tilde{c}_{in}) - \bar{\psi}_{,c}(x_2 + \tilde{c}_{in})| &\leq |\bar{\psi}_{,cc}(x_1 + \tilde{c}_{in})| |x_1 - x_2| \\ &\leq |1 - \alpha(x_1 + \tilde{c}_{in})| |x_1 - x_2| \leq (1 + \alpha + \alpha|x_1|) |x_1 - x_2|. \end{aligned}$$

If $x_1 + \tilde{c}_{in}$, and $x_2 + \tilde{c}_{in} \leq \frac{1}{2\alpha}$, then

$$\begin{aligned} |\bar{\psi}_{,c}(x_1 + \tilde{c}_{in}) - \bar{\psi}_{,c}(x_2 + \tilde{c}_{in})| &= |x_1 + \tilde{c}_{in} - \frac{\alpha}{2}(x_1 + \tilde{c}_{in})^2 - (x_2 + \tilde{c}_{in}) + \frac{\alpha}{2}(x_2 + \tilde{c}_{in})^2| \\ &= |x_1 - x_2 - \frac{\alpha}{2}(x_1 + \tilde{c}_{in})^2 + \frac{\alpha}{2}(x_2 + \tilde{c}_{in})^2| \\ &= |x_1 - x_2 - \frac{\alpha}{2}(x_1 + x_2)(x_1 - x_2) - \alpha\tilde{c}_{in}(x_1 - x_2)| \\ &\leq (1 + \alpha + \frac{\alpha}{2}|x_1| + \frac{\alpha}{2}|x_2|) |x_1 - x_2|. \end{aligned}$$

Now we can conclude that

$$\begin{aligned} \int_S (\bar{\psi}_{,c}(\rho_1 + \tilde{c}_{in}) - \bar{\psi}_{,c}(\rho_2 + \tilde{c}_{in}))w &\leq \int_S (1 + \alpha + \frac{\alpha}{2}|\rho_1| + \frac{\alpha}{2}|\rho_2|) |\rho_1 - \rho_2| |w| \\ &\leq (1 + \alpha) \|\rho_1 - \rho_2\|_S \|w\|_S + \frac{\alpha}{2} (\|\rho_1\|_{L^3(S)} + \|\rho_2\|_{L^3(S)}) \|\rho_1 - \rho_2\|_{L^3(S)} \|w\|_{L^3(S)} \\ &\leq C_1 \|\rho_1 - \rho_2\|_1 \|w\|_1 + C_2 (\|\rho_1\|_1 + \|\rho_2\|_1) \|\rho_1 - \rho_2\|_1 \|w\|_1. \end{aligned} \quad (\text{A.6})$$

Now,

$$\begin{aligned}
& \int_S \sum_{j=0}^m c^j \delta t ((\rho_1 + \tilde{c}_{in}) - (\rho_2 + \tilde{c}_{in})) w \\
&= \int_S \sum_{j=0}^m c^j \delta t (\rho_1 - \rho_2) w \leq \delta t \int_S \sum_{j=0}^m |c^j| |\rho_1 - \rho_2| |w| \\
&\leq \delta t \sum_{j=0}^m \|c^j\|_{L^3(S)} \|\rho_1 - \rho_2\|_{L^3(S)} \|w\|_{L^3(S)} \leq C \delta t \sum_{j=0}^m \|c^j\|_1 \|\rho_1 - \rho_2\|_1 \|w\|_1
\end{aligned} \tag{A.7}$$

And finally,

$$\begin{aligned}
& \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n}) (\rho_1 - \rho_2) w \leq \int_{\Gamma_{out}} |\mathbf{u}^{m+1} \cdot \mathbf{n}| |\rho_1 - \rho_2| |w| \\
&\leq \|\mathbf{u}^{m+1}\|_{L^3(\Gamma_{out})} \|\rho_1 - \rho_2\|_{L^3(\Gamma_{out})} \|w\|_{L^3(\Gamma_{out})} \leq C \|\mathbf{u}^{m+1}\|_1 \|\rho_1 - \rho_2\|_1 \|w\|_1
\end{aligned} \tag{A.8}$$

Combining (A.2)-(A.8), we have

$$\begin{aligned}
& |\langle A\rho_1 - A\rho_2, w \rangle| \\
&\leq \left(C_1 + C_2 \|\mathbf{u}^{m+1}\|_1 + C_3 (\|\rho_1\|_1 + \|\rho_2\|_1) + C_4 \delta t \sum_{j=0}^m \|c^j\|_1 \right) \|\rho_1 - \rho_2\|_1 \|w\|_1
\end{aligned} \tag{A.9}$$

This completes the proof. Q.E.D.

B Error Estimates

In this section, we further assume that

$$\begin{aligned}
\text{(A1)} \quad & \int_0^T \|\partial_{tt} c\|_{-1}^2 dt \leq M_1 \quad \text{for some constant } M_1 \text{ and} \\
\text{(A2)} \quad & \sup_{t \in [0, T]} \|\partial_t c\|_{H^1(\Omega)'} < M_2 \quad \text{for some constant } M_2.
\end{aligned} \tag{B.1}$$

For convenience, we define

$$\begin{aligned}
B(u, v) &:= \left(1 + \frac{\alpha i_0^2}{D} u\right) i_0 v, \\
b(u, v, w) &:= \int_S \left(1 + \frac{\alpha i_0^2}{D} u\right) i_0 v w, \\
G(u, v, w) &:= \int_S \frac{\alpha i_0^3}{D} u v w.
\end{aligned} \tag{B.2}$$

The difference equation for the exact solution of c defined by (2.3) can be expressed as:

$$\frac{c(t^{m+1}) - c(t^m)}{\delta t} + \mathbf{u}^{m+1} \cdot \nabla c(t^{m+1}) - D \Delta c(t^{m+1}) = R^m, \tag{B.3}$$

where

$$R^m = \frac{1}{\delta t} \int_{t^m}^{t^{m+1}} (t - t^m) \partial_{tt} c(t) dt \quad (\text{B.4})$$

Defining $\epsilon^j = c(t^j) - c^j$, the error equation can be expressed by

$$\frac{\epsilon^{m+1} - \epsilon^m}{\delta t} + \mathbf{u}^{m+1} \cdot \nabla \epsilon^{m+1} - D \Delta \epsilon^{m+1} = R^m \quad (\text{B.5})$$

subject to the boundary condition

$$\begin{aligned} \epsilon^{m+1} &= 0 \quad \text{on } \Gamma_{in}, & \frac{\partial \epsilon^{m+1}}{\partial n} &= 0 \quad \text{on } \Gamma_{wall} \cup \Gamma_{out} \\ D \frac{\partial \epsilon^{m+1}}{\partial n} + B \left(\int_0^{t^{m+1}} c(s) ds, c(t^{m+1}) \right) - B \left(\sum_{j=0}^m c_j \delta t, c^{m+1} \right) &= 0 \quad \text{on } S. \end{aligned} \quad (\text{B.6})$$

The symmetrized weak formulation to (B.3) is

$$\begin{aligned} & \int_{\Omega} \frac{c(t^{m+1}) - c(t^m)}{\delta t} w + D \int_{\Omega} \nabla c(t^{m+1}) \cdot \nabla w \\ & + \frac{1}{2} \int_{\Omega} [(\mathbf{u}^{m+1} \cdot \nabla c(t^{m+1})) w - (\mathbf{u}^{m+1} \cdot \nabla w) c(t^{m+1})] dx + \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n}) c^{m+1} w \\ & + \int_S \left(1 - \frac{\alpha c^{m+1}}{2} + \frac{\alpha i_0^2}{D} \sum_{j=0}^m c^j \right) i_0 c^{m+1} w = \int_{\Omega} R^m w dx \end{aligned} \quad (\text{B.7})$$

Subtracting (B.7) by (4.1), we have

$$\begin{aligned} & \int_{\Omega} \frac{\epsilon^{m+1} - \epsilon^m}{\delta t} w dx + D \int_{\Omega} \nabla \epsilon^{m+1} \cdot \nabla w dx \\ & + \frac{1}{2} \int_{\Omega} [(\mathbf{u}^{m+1} \cdot \nabla \epsilon^{m+1}) w - (\mathbf{u}^{m+1} \cdot \nabla w) \epsilon^{m+1}] dx + \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n}) \epsilon^{m+1} w \\ & - \int_S \frac{\alpha i_0}{2} (c(t^{m+1}) + c^{m+1}) \epsilon^{m+1} w + b \left(\int_0^{t^{m+1}} c(s) ds, c(t^{m+1}), w \right) \\ & - b \left(\sum_{j=0}^m c^j \delta t, c^{m+1}, w \right) = \int_{\Omega} R^m w dx \end{aligned} \quad (\text{B.8})$$

Before investigating the error estimate, some auxiliary results are needed. We collect them in the Remark below:

Remark 2 We have

$$\begin{aligned} & B \left(\int_0^{t^{m+1}} c(s) ds, c(t^{m+1}) \right) - B \left(\sum_{j=0}^m c^j \delta t, c^{m+1} \right) \\ & = \left(1 + \frac{\alpha i_0^2}{D} \sum_{j=0}^m c^j \delta t \right) i_0 \epsilon^{m+1} + \frac{\alpha i_0^3}{D} \left[\int_0^{t^{m+1}} c(s) ds - \sum_{j=0}^m c^j \delta t \right] c(t^{m+1}) \end{aligned} \quad (\text{B.9})$$

Defining

$$\xi^m = \int_0^{t^{m+1}} c(s) ds - \sum_{j=0}^m c^j \delta t \quad (\text{B.10})$$

we have

$$\xi^m = \sum_{j=0}^m \epsilon^j \delta t + \phi^m, \quad (\text{B.11})$$

where $\phi^m = \sum_{j=0}^m \partial_t c(\theta^j) \delta t^2$ for some $\theta^j \in (t^j, t^{j+1})$. By (B.9), (B.10), (B.11) and letting $w = \epsilon^{m+1}$ in (B.8), we have

$$\begin{aligned} & \frac{1}{\delta t} \|\epsilon^{m+1}\|^2 - \frac{1}{\delta t} \int_{\Omega} \epsilon^{m+1} \epsilon^m dx + D \|\nabla \epsilon^{m+1}\|^2 + b \left(\sum_{j=0}^m c^j \delta t, \epsilon^{m+1}, \epsilon^{m+1} \right) \\ & + G(c(t^{m+1}), \sum_{j=0}^m \epsilon^j \delta t, \epsilon^{m+1}) + G(c(t^{m+1}), \phi^m, \epsilon^{m+1}) + \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n}) (\epsilon^{m+1})^2 \\ & - \int_S \frac{\alpha i_0}{2} (c(t^{m+1}) + c^{m+1}) (\epsilon^{m+1})^2 = \int_{\Omega} R^m w dx \end{aligned} \quad (\text{B.12})$$

Multiplying the both sides by δt , we get

$$\begin{aligned} & \|\epsilon^{m+1}\|^2 + D \delta t \|\nabla \epsilon^{m+1}\|^2 + \delta t b \left(\sum_{j=0}^m c^j \delta t, \epsilon^{m+1}, \epsilon^{m+1} \right) \\ & + \delta t G(c(t^{m+1}), \sum_{j=0}^m \epsilon^j \delta t, \epsilon^{m+1}) + \delta t G(c(t^{m+1}), \phi^m, \epsilon^{m+1}) + \delta t \int_{\Gamma_{out}} (\mathbf{u}^{m+1} \cdot \mathbf{n}) (\epsilon^{m+1})^2 \\ & - \delta t \int_S \frac{\alpha i_0}{2} (c(t^{m+1}) + c^{m+1}) (\epsilon^{m+1})^2 = \delta t \int_{\Omega} R^m w dx + \int_{\Omega} \epsilon^{m+1} \epsilon^m dx \end{aligned} \quad (\text{B.13})$$

Q.E.D.

Theorem 3 There is a generic constant C such that

$$\|\epsilon^{m+1}\| \leq C \delta t \quad \forall 0 \leq m \leq \frac{T}{\delta t} - 1 \quad (\text{B.14})$$

and

$$\|\epsilon^{m+1}\|_1 \leq C \delta t^{\frac{1}{2}} \quad \forall 0 \leq m \leq \frac{T}{\delta t} - 1. \quad (\text{B.15})$$

Proof. By a recurrence argument, we are going to show that if the statements (B.14) and (B.15) hold simultaneously for all ϵ^j and for all $j \leq m$, then they hold as well for ϵ^{m+1} . Notice that it is true when $m = 0$.

Defining $G_1 = |G(c(t^{m+1}), \sum_{j=0}^m \epsilon^j \delta t, \epsilon^{m+1})|$ and $G_2 = |G(c(t^{m+1}), \phi^m, \epsilon^{m+1})|$, we have the estimates:

$$\begin{aligned}
G_1 &\leq \frac{\alpha i_0^3}{D} \sum_{j=0}^m \int_S |c(t^{m+1}) \epsilon^j \epsilon^{m+1} \delta t| \\
&\leq \frac{\alpha i_0^3}{D} \sum_{j=0}^m \int_S |\delta t \epsilon^j \epsilon^{m+1}| \leq \frac{\alpha i_0^3}{D} \left(\sum_{j=0}^m \|\epsilon^j\|_S \delta t \right) \|\epsilon^{m+1}\|_S \\
&\leq C \left(\sum_{j=0}^m \|\epsilon^j\|_1 \delta t \right) \|\epsilon^{m+1}\|_1 \leq C(m+1) \delta t^{\frac{3}{2}} \|\epsilon^{m+1}\|_1 \leq C \|\epsilon^{m+1}\| \delta t^{\frac{1}{2}}.
\end{aligned} \tag{B.16}$$

Using (A2), we have

$$\begin{aligned}
G_2 &\leq \frac{\alpha i_0^3}{D} \int_S |c(t^{m+1}) \phi^m \epsilon^{m+1}| \leq \frac{\alpha i_0^3}{D} \int_S |\phi^m \epsilon^{m+1}| \\
&\leq \frac{\alpha i_0^3}{D} \|\phi^m\|_S \|\epsilon^{m+1}\|_S \leq C \|\phi^m\|_1 \|\epsilon^{m+1}\|_1 \leq C \delta t^2 \|\epsilon^{m+1}\|_1.
\end{aligned} \tag{B.17}$$

By (A1), we have

$$\begin{aligned}
&\delta t \left| \int_{\Omega} R^m \epsilon^{m+1} dx \right| \\
&\leq \frac{D}{4} \delta t \|\epsilon^{m+1}\|_1^2 + C \delta t^{-1} \|R^m\|_{H^1(\Omega)'}^2 \\
&= \frac{D}{4} \|\epsilon^{m+1}\|_1^2 + C \delta t^{-1} \left\| \int_{t^m}^{t^{m+1}} (t - t^m) \partial_{tt} c dt \right\|_{H^1(\Omega)'}^2 \\
&\leq \frac{D}{4} \|\epsilon^{m+1}\|_1^2 + C \delta t^{-1} \int_{t^m}^{t^{m+1}} \|\partial_{tt} c\|_{H^1(\Omega)'}^2 dt \int_{t^m}^{t^{m+1}} (t - t^m)^2 dt \\
&\leq \frac{D}{4} \|\epsilon^{m+1}\|_1^2 + C \delta t^2 \int_{t^m}^{t^{m+1}} \|\partial_{tt} c\|_{H^1(\Omega)'}^2 dt \\
&\leq \frac{D}{4} \|\epsilon^{m+1}\|_1^2 + C \delta t^2
\end{aligned} \tag{B.18}$$

$$\begin{aligned}
&b \left(\sum_{j=0}^m c^j \delta t, \epsilon^{m+1}, \epsilon^{m+1} \right) - \int_S \frac{\alpha i_0}{2} (c(t^{m+1}) + c^{m+1}) (\epsilon^{m+1})^2 \\
&\geq \int_S \left(1 + \frac{\alpha i_0^2}{D} \sum_{j=0}^m c^j \delta t \right) i_0 (\epsilon^{m+1})^2 - \int_S \alpha i_0 (\epsilon^{m+1})^2 \\
&\geq \int_S \left((1 - \alpha) + \frac{\alpha i_0^2}{D} \sum_{j=0}^m c^j \delta t \right) i_0 (\epsilon^{m+1})^2 \geq 0.
\end{aligned} \tag{B.19}$$

$$\int_{\Omega} \epsilon^{m+1} \epsilon^m dx \leq \frac{1}{2} \|\epsilon^m\|^2 + \frac{1}{2} \|\epsilon^{m+1}\|^2. \tag{B.20}$$

Combining (B.13), (B.16)-(B.20), and since the boundary term of Γ_{out} in (B.13) is nonnegative, we have

$$\left(\frac{1}{2} - \frac{1}{4}D\delta t\right)\|\epsilon^{m+1}\|^2 + \frac{3}{4}D\delta t\|\epsilon^{m+1}\|_1^2 \leq C\|\epsilon^{m+1}\|_1\delta t^{\frac{3}{2}} + C\delta t^2. \quad (\text{B.21})$$

This implies (B.15). Now using (B.21) and (B.15), we get (B.14). Q.E.D.

Theorem 4 (Improved estimate) For $0 \leq m \leq \frac{T}{\delta t} - 1$, we have

$$\delta t \sum_{j=0}^m \|\epsilon^{j+1}\|_1^2 \leq C\delta t^2 \quad (\text{B.22})$$

Proof. Putting $w = 2\epsilon^{m+1}$ in (B.13) and using the estimates in Theorem 3, we have

$$\begin{aligned} \|\epsilon^{m+1}\|^2 + 2D\delta t\|\nabla\epsilon^{m+1}\|^2 &\leq 2\delta tC_1\|\epsilon^{m+1}\|_1 \left(\sum_{j=0}^m \|\epsilon^j\|_1\delta t \right) \\ + 2C_2\delta t^3\|\epsilon^{m+1}\|_1 + \frac{D}{4}\delta t\|\epsilon^{m+1}\|_1^2 + C_3\delta t^2 \int_{t^m}^{t^{m+1}} \|\partial_{tt}c\|_{H^1(\Omega)'}^2 dt + \|\epsilon^m\|^2 \end{aligned} \quad (\text{B.23})$$

Note that

$$\|\epsilon^{m+1}\|^2 + 2D\delta t\|\epsilon^{m+1}\|_1^2 = (1 - 2D\delta t)\|\epsilon^{m+1}\|^2 + 2D\delta t\|\epsilon^{m+1}\|_1^2.$$

Taking the sum of (B.23) from 0 to m and using (A1), we have

$$\begin{aligned} \left(1 - \frac{9}{4}D\delta t\right)\|\epsilon^{m+1}\|^2 + \frac{7}{4}\sum_{j=0}^m D\delta t\|\epsilon^{j+1}\|_1^2 &\leq \sum_{j=0}^m 2\delta tC_1\|\epsilon^{j+1}\|_1 \left(\sum_{k=0}^j \|\epsilon^k\|_1\delta t \right) \\ &\quad + \sum_{j=0}^m 2C_2\delta t^3\|\epsilon^{j+1}\|_1 + C_3\delta t^2. \end{aligned} \quad (\text{B.24})$$

The first term on the right hand side of (B.24) can be estimated by

$$\begin{aligned} &\sum_{j=0}^m 2\delta tC_1\|\epsilon^{j+1}\|_1 \left(\sum_{k=0}^j \|\epsilon^k\|_1\delta t \right) \\ &\leq \sum_{j=0}^m \frac{D}{4}\delta t\|\epsilon^{j+1}\|_1^2 + C\delta t^2 \sum_{j=0}^m \sum_{k=0}^j \|\epsilon^k\|_1^2 \\ &\leq \frac{D}{4}\delta t\|\epsilon^{m+1}\|_1^2 + C\delta t \sum_{j=0}^m \|\epsilon^j\|_1^2. \end{aligned} \quad (\text{B.25})$$

Similarly, the second term can be estimated by

$$\begin{aligned}
\sum_{j=0}^m 2C_2\delta t^3 \|\epsilon^{m+1}\|_1 &\leq \sum_{j=0}^m 2C\delta t^3 \left(\frac{1}{2\nu} \|\epsilon^{m+1}\|_1^2 + \frac{\nu}{2} |\Omega| \right) \\
&\leq \nu CT |\Omega| \delta t^2 + \frac{C}{\nu} \delta t^2 \sum_{j=0}^m \|\epsilon^{m+1}\|_1^2 \\
&\leq C\delta t^2 + \frac{D}{4} \delta t \sum_{j=0}^m \|\epsilon^j\|_1^2 + \frac{D}{4} \delta t \|\epsilon^{m+1}\|_1^2
\end{aligned} \tag{B.26}$$

for every $\nu > 0$.

Finally, employing (B.24)-(B.26), we have

$$\left(1 - \frac{11}{4} D\delta t \right) \|\epsilon^{m+1}\|_1^2 + \frac{5}{4} D\delta t \sum_{j=0}^m \|\epsilon^{j+1}\|_1^2 \leq C_1 \delta t^2 + C_2 \delta t \sum_{j=0}^m \|\epsilon^j\|_1^2. \tag{B.27}$$

By induction on m , we can easily show that

$$\frac{5}{4} D\delta t \sum_{j=0}^m \|\epsilon^{j+1}\|_1^2 \leq C\delta t^2. \tag{B.28}$$

Q.E.D.