SCIENTIFIC COMPUTING FOR ALUMINUM PRODUCTION

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Abstract. Numerical simulation in aluminum production envolves many different topics, this paper is dedicated to some aspects of two of them. In the first part, a numerical approach for the computation of the motion of the fluids in an electrolitic cell is presented, with an emphasis on a mesh deformation technique to track the free interface during the time. In a second part, a model for the simulation of alumina dissolution and repartition is exposed.

Key Words. Aluminum production, Electrolysis, Magnetohydrodynamics, Alumina injection and dissolution.

1. Introduction

The industrial production of aluminum is based, since the end of the 19th century, on the electrolysis process. In most countries, aluminum is produced from alumina (Al_2O_3) which itself comes from bauxite. Aluminum electrolysis is performed in big Hall-Heroult cells, of approximate size 10 meters long, 3 meters wide and 1 meter high. Figure (1) gives a schematic representation of a vertical transverse cut of such a cell. Alumina particles are injected periodically in a corrosive electrolytic bath lying over the liquid aluminum. Both fluids are kept at temperature 965°C and are immiscible.

A strong electric current (current density about $10'000 \ A/m^2$) runs through the fluids from the carbone anodes to the cathode allowing electrolysis to take place in the electrolyte. The chemical reaction:

$$2Al_2O_3$$
 + energy $\rightarrow 4Al + 3O_2$,

produces liquid aluminum and oxygen bubbles which are gradually burning the anodes and creating CO_2 . Anodes must therefore be changed regularly. Alumina is added periodically to the electrolytic bath and the aluminum is produced and recolted at the bottom of the cell every day.

All these running operations can produce perturbations of the cell rendering. Moreover, industry wants to optimize the production, this is to say increase current density as much as possible, by acting on the geometrical and electrical configurations of such electrolytic cells. Since physical observations are difficult to perform due to high temperature, current and magnetic induction in the cells, numerical simulations are very useful.

In this paper, some of the physical phenomena involved in aluminum production are presented, with emphasis on numerical simulation.

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FIGURE 1. Simplified electrolytic cell : Notations

The most important point, is that both fluids are immiscible and moving, since they are subject to electromagnetic and gravity forces. These electromagnetic forces (Lorentz forces) arise from the combination of the current density running through the fluids and the magnetic induction due to the whole electric configuration of a hall containing many cells. It is of great importance to be able to control the motion of the fluids in a cell because if the aluminum, which is a much better conductor than the electrolyte, touches the anodes then the cell would be strongly damaged.

This leads to the following magnetohydrodynamic (MHD) modelling: multifluid incompressible Navier-Stokes equations for the computation of the velocity and pressure, with an unknown interface between liquid aluminum and electrolyte determined by a level-set function, are coupled with the quasi-static Maxwell model for the computation of the magnetic induction in the whole space. The electric current density is given by Ohm's law in moving media, determined only in the cell.

Many other phenomenons may be considered. Let us mention two of them :

- (1) Temperature effects. Temperature in the electrolytic bath is maintained at a value close to 965°C. This is the result of an equilibrium between heat production mainly in the bath due to Joule's effect and heat transfer through the walls of the cell. Since the fusion temperature of the electrolytic bath is close to the mean production temperature, it can solidify on the internal walls of the cell, creating the so-called ledges. These ledges are very useful to protect the walls of the cell against the extreme chemical agressivity of the bath but they are modifying the electric currents in the cell and, of course, the shape of the fluid domain. In this document we do not deal with thermal effects but refer to [6] for a discussion on this particular question.
- (2) *Ferromagnetic effects.* The whole cell is placed in a steel container two centimers thick. Since the container is ferromagnetic, it reduces the intensity of magnetic induction into the fluids by screening and therefore the Lorentz

forces that induce movements in the fluids. Eventhough ferromagnetic effects are crucial when studying the stability of an electrolytic cell but we will not discuss this question here and refer to [4].

The contributions of this paper are the following :

- We recall the *MHD* model for the computation of the fluid motion presented in [5] and focus on the mesh deformation algorithm for the interface tracking,
- (ii) We present a numerical model to simulate the injection of alumina particles, their dissolution into the electrolyte and the repartition of the dissolved alumina concentration. This topic is linked to the previous one since we require here the knowledge of the velocity field in the fluid domain. It is of great importance when dealing with the optimization of aluminum production since a non-uniform concentration of dissolved alumina can lead to a great waste of energy.

Finally let us mention that all those phenomenons do not share the same time scale. Some of these are on a small time scale like the motion of the fluids, some are of intermediate time scale like the convection-diffusion of alumina in the bath and others are of longer scale like the formation of ledges on the cell walls. This is why we have developed two types of simulations: a simulation of transient in the cell, mainly for MHD movements and a simulation of the steady state of the cell, for solidification ledges and to compute the linear stability (see [2, 3]).

2. Magnetohydrodynamics

The liquid aluminum and the electrolytic bath are assumed to be *incompressible* viscous Newtonian fluids and their motion can be described with the incompressible Navier-Stokes equations. Note also that the two fluids are immiscible. The main physical caracteristics of these fluids are reported in Table 1.

Parameter	Unit	Symbol	Aluminum	Bath
Density	kg/m^3	ρ	2270	2130
Viscosity	$Pa \cdot s$	μ	2E-3	1E-3
Electric Conductivity	S/m	σ	3.33 E6	210

TABLE 1. Physical parameters associated with the electrolysis of aluminum.

2.1. Multifluid Navier-Stokes equations. Let Σ be the bounded domain of \mathbb{R}^3 occupied by the whole eletrolytic cell (fluids, anodes, cathodes, conductors, etc), $\Omega \subset \Sigma$ the fluid domain containing the liquid aluminum $\Omega_{al}(t)$ and the electrolytic bath $\Omega_{el}(t)$, where t is the time (see Figure 1). Clearly we have $\overline{\Omega} = \overline{\Omega}_{al}(t) \cup \overline{\Omega}_{el}(t) \subset \overline{\Sigma}$ for all time t. The interface between the two fluids will be denoted $\Gamma(t)$, i.e. $\Gamma(t) = \overline{\Omega}_{al}(t) \cap \overline{\Omega}_{el}(t)$ for all t. We assume moreover that there exists a domain $\Lambda \subset \mathbb{R}^2$ and a smooth function $g: (0, \infty) \times \Lambda \to \mathbb{R}$ such that

(1)
$$\Gamma(t) = \{ (x, y, z) \in \mathbb{R}^3 : z = g(t, x, y), (x, y) \in \Lambda, t \in (0, \infty) \}.$$

In other words, $\Gamma(t)$ can be seen as a smooth surface parametrized by x and y. In order to simplify the presentation, the density and viscosity of liquid aluminum,

denoted by ρ_{al} and μ_{al} are considered constant in space and time. The same assumption is made for the density and viscosity of electrolytic bath ρ_{el} and μ_{el} . In order to simplify the notations and if there is no ambiguity, we write in the following only μ and ρ .

If we assume that the current density \vec{j} going through the cell and the ambiant magnetic induction field \vec{B} are known, then the external force \vec{f} acting on the fluids is the sum of the gravity and of the *Lorentz forces*, i.e. $\vec{f} = \rho \vec{g} + \vec{j} \times \vec{B}$. Then the velocity \vec{u} and pressure p of the fluids satisfy the multifluid incompressible Navier-Stokes equations:

(2)
$$\begin{cases} \rho \frac{\partial \vec{u}}{\partial t} + \rho(\vec{u} \cdot \nabla)\vec{u} - \nabla \cdot (\boldsymbol{\tau}(\vec{u}, p)) = \vec{f}, \\ \nabla \cdot \vec{u} = 0, \end{cases} \quad \text{in } \Omega_{al}(t) \cup \Omega_{el}(t), \end{cases}$$

where $\boldsymbol{\tau}$ is the stress tensor defined by

(3)
$$\boldsymbol{\tau}(\vec{u},p) = 2\mu\boldsymbol{\varepsilon}(\vec{u}) - pI, \text{ with } \boldsymbol{\varepsilon}(\vec{u}) = \frac{1}{2} \left(\nabla \vec{u} + \nabla \vec{u}^T \right)$$

On the interface we assume the continuity of the velocity and write $[\vec{u}] = \vec{0}$ on $\Gamma(t)$, where $[\beta] = \beta_{el} - \beta_{al}$ is the jump of the quantity β between Ω_{al} and Ω_{el} . Considering surface tension effects we also impose that

$$(4) \qquad [\boldsymbol{\tau}\vec{n}] = \gamma H\vec{n}$$

where \vec{n} is the unit normal on $\Gamma(t)$ pointing from Ω_{al} to Ω_{el} , γ is a constant surface tension coefficient depending on the fluids and H is the Gauss curvature of the surface $\Gamma(t)$ positively counted with respect to the normal \vec{n} . The equation (4) must be completed with the value of the contact angle between $\Gamma(t)$ and the wall of the cell. In practice this contribution will be treated following the method presented in [10], by introducing in the variational formulation of (2), an integral on Γ and applying a surface divergence theorem (see below).

The equation (2) is completed with a suitable initial condition for the velocity

$$\vec{u}(0) = \vec{u}_0$$
 such that $\nabla \cdot \vec{u}_0 = 0$,

by homogenous Dirichlet boundary conditions for the velocity $\vec{u} = 0$ on a part Γ_D of the border $\partial\Omega$ and by slip boundary conditions, i.e. $\vec{u} \cdot \vec{n} = 0$ and $\tau \vec{n} \cdot \vec{t_i} = 0, i = 1, 2$, on $\Gamma_S = \partial\Omega \setminus \Gamma_D$ (see section 2.2), where $\vec{t_1}, \vec{t_2}$ are two orthogonals vectors in the normal plane to \vec{n} . Remark that the temperature drop between the interior and the exterior of the domain Ω usually leads to the creation of solidified ledges near the border of the cell. However we will not include those thermic effects in our model and refer to [6] for a discussion on this particular question.

Considering the spaces $L^2_0(\Omega) = \left\{ q \in L^2(\Omega) : \int_\Omega q \, dx = 0 \right\}$ and

(5)
$$\widetilde{H}^1(\Omega) = \left\{ \vec{v} \in H^1(\Omega)^3 \text{ satisfying } \vec{v} = 0 \text{ on } \Gamma_D \text{ and } \vec{v} \cdot \vec{n} = 0 \text{ on } \Gamma_S \right\},\$$

an appropriate weak formulation of the problem (2) with interface conditions (3) and (4) is the following : find two mappings $\vec{u}: (0,\infty) \to \tilde{H}^1(\Omega)$ and $p: (0,\infty) \to$

 $L_0^2(\Omega)$ satisfying

(6)
$$\int_{\Omega} \left(\rho \frac{\partial \vec{u}}{\partial t} \cdot \vec{v} + \rho(\vec{u} \cdot \nabla) \vec{u} \cdot \vec{v} + \boldsymbol{\tau}(\vec{u}, p) : \boldsymbol{\varepsilon}(\vec{v}) \right) = \int_{\Omega} \vec{f} \cdot \vec{v} + \int_{\Gamma(t)} \gamma H \vec{v} \cdot \vec{n},$$
(7)
$$\int_{\Omega} q \operatorname{div}(\vec{u}) = 0,$$

for all $\vec{v} \in \tilde{H}^1(\Omega)$ and all $q \in L^2_0(\Omega)$. Here $L^2(\Omega)$ is the standard space of square integrable functions and $H^1(\Omega)$ the standard Sobolev space. We refer to [13] and [11] for the discussion on the existence of the solution and to [5] for the description of a numerical scheme in time to deal with these equations.

Introducing $\vec{n}_{\partial\Omega}$ the outward unit normal to $\partial\Omega$ and $\vec{n}_{\Gamma} = \vec{n}$ the normal to the interface $\Gamma(t)$, we define on the boundary $\partial\Gamma(t) = \Gamma(t) \cap \partial\Omega$ the vectors

$$\vec{t}_{\partial\Gamma} = \vec{n}_{\Gamma} \times \vec{n}_{\partial\Omega}, \quad \vec{t}_{\partial\Omega} = \vec{n}_{\partial\Omega} \times \vec{t}_{\partial\Gamma} \quad \text{and} \quad \vec{m} = \vec{t}_{\partial\Gamma} \times \vec{n}_{\Gamma}.$$

Denoting by $\nabla_S \vec{v}$ the gradient of \vec{v} along the surface $\Gamma(t)$ (see [10] for details) and by θ the physical contact angle defined as the angle between \vec{m} and $\vec{t}_{\partial\Omega}$ we can rewrite the surface tension term in (6) as

(8)
$$\int_{\Gamma(t)} \gamma H \vec{v} \cdot \vec{n} = \int_{\partial \Gamma(t)} \gamma \cos(\theta) \ \vec{t}_{\partial \Omega} \cdot \vec{v} - \int_{\Gamma(t)} \gamma \operatorname{tr} (\nabla_S \vec{v})$$

Since the dimensions of the cell are large, the surface tension could be neglected as it influences the shape of the interface only close to the boundary. However, in particular cases where the dimensions are smaller, this term can have a dominant contribution.

2.2. Free Surface. The resolution of (2) requires the knowledge of the shape of the domains $\Omega_{al}(t)$ and $\Omega_{el}(t)$ at time t, i.e. the time evolution of the interface $\Gamma(t), \forall t \geq 0$. For this purpose, we follow here a *Level-Set method* (see [15]) and define a C^1 function $\varphi : (0, \infty) \times \Omega$ satisfying for all time t:

(9)
$$\varphi(t,x) \begin{cases} > 0 & \text{in the bath,} \\ < 0 & \text{in the liquid aluminum} \\ = 0 & \text{on the interface.} \end{cases}$$

The zero isosurface of φ defines the interface $\Gamma(t)$ and φ is the solution of the following linear convection equation:

(10)
$$\frac{\partial \varphi}{\partial t} + \vec{u} \cdot \nabla \varphi = 0 \quad \text{in } (0, \infty) \times \Omega,$$

where \vec{u} is the fluid velocity. In fact the Level-Set method deals with the distance between x and the interface $\Gamma(t)$ but since we numerically solve equation (10) on small time steps, we re-normalize φ after each time step (see [15]).

Remark that this approach influences the choice of the boundary conditions of the Navier-Stokes problem. Liquid aluminum and electrolytic bath being viscous fluids, we should impose homogeneous Dirichlet boundary conditions everywhere on $\partial\Omega$. The problem is that this would freeze the interface on the boundary $\partial\Omega$ and though it is preferable to impose slip conditions on the lateral wall and therefore permit the interface to move vertically. For this purpose, on the lateral wall of the fluid domain Γ_S , slip boundary conditions while on $\Gamma_D = \partial\Omega \setminus \Gamma_S$, homogeneous Dirichlet boundary conditions are imposed.

494

2.3. Mesh Deformation. In general the Level-Set method is associated with the use of a fixed mesh. Thus, in order to have a good approximation of the free surface, fine meshes are needed, at least in a neighbourhood of the interface. This can not be done easily here since the computational domain is very complex from the geometrical point of view. The fluid mesh typically represents less than 50 % of the elements of the whole mesh. In this context it seems reasonable to introduce a deformation technique that forces the mesh to follow the interface motion.

2.3.1. Interface mesh deformation. Let us introduce the reference fluid domain $\hat{\Omega}$ corresponding to the case when the interface Γ between the two fluids is horizontal at the altitude $z_0 \in \mathbb{R}$. For some h > 0, we associate to this domain an isotropic conforming tetrahedral mesh $\hat{\mathcal{T}}_h$ with N_h nodes and M_h elements of typical size h. $\hat{\mathcal{T}}_h$ is compatible with the physical domains in the sense that an element is either entirely in the aluminum domain either in the bath domain. In this configuration, the interface Γ_h is the union of some triangular faces of elements belonging to $\hat{\mathcal{T}}_h$.

In the following we introduce an algorithm that allows us to build, for any time t_n , a mesh \mathcal{T}_h^n of the fluid domain at time t_n by deforming the reference mesh $\hat{\mathcal{T}}_h$.

Suppose that we know at time t_n the mesh of the fluid domain \mathcal{T}_h^n and the velocity field \vec{u}_h^{n+1} computed from (6-7) on \mathcal{T}_h^n during the time interval (t_n, t_{n+1}) . We now explain how to construct the new triangulation \mathcal{T}_h^{n+1} . For each node x_i^n of the mesh \mathcal{T}_h^n , $i = 1, \ldots, N_h$, we can define φ_h^n as

(11)
$$\varphi_h^n(x_i^n) = \begin{cases} \operatorname{dist}(x_i^n, \Gamma_h^n), & \text{if } x_i^n \in \Omega_{el}(t_n), \\ -\operatorname{dist}(x_i^n, \Gamma_h^n), & \text{if } x_i^n \in \Omega_{al}(t_n), \end{cases}$$

where $\operatorname{dist}(x, \Gamma_h^n)$ is the vertical distance function between $x \in \Omega$ and the surface Γ_h^n .

Solving with a finite element method, the transport equation (10) on (t_n, t_{n+1}) with φ_h^n as initial condition and \vec{u}_h^{n+1} as velocity, we compute the convected levelset function φ_h^{n+1} on \mathcal{T}_h^n . The zero isosurface of φ_h^{n+1} yields the new interface Γ_h^{n+1} .

The first step of the deformation algorithm is to move the N_h^{Γ} nodes x_i^n of Γ_h^n to their new position x_i^{n+1} in order to assure that

(12)
$$\varphi_h^{n+1}(x_i^{n+1}) = 0, \quad \forall i = 1, \dots, N_h^{\Gamma}.$$

To do this, we first associate to each node $x_i^n \in \Gamma_h^n$ a trajectory line, say a curve in \mathbb{R}^3 parametrized by $\gamma_i : [0,1] \to \mathbb{R}^3$ on which the node x_i^n is allowed to move. In most situation we can choose straight lines as trajectories. The goal here is to keep a conforming mesh at each time step and insure that the nodes do not cross the cell boundary $\partial \Omega$ (see Figure 2). Clearly, if the lateral wall is vertical, all trajectories are parallel. This is often not the case in cells where lateral faces of $\partial \Omega$ are usually inclined.



FIGURE 2. Example of possible trajectory lines depending on cell geometry. Left : vertical boundary, right : inclined boundary. The nodes of the interface are only allowed to move along these trajectroy lines.

The restriction of the level-set function φ_h^{n+1} on such a line will be given by the function $\chi_i: [0,1] \to \mathbb{R}$ such that

(13)
$$\chi_i(s) = \left(\varphi_h^{n+1} \circ \gamma_i\right)(s), \quad \forall s \in [0,1] \text{ and } i = 1, \dots, N_h^{\Gamma}.$$

The new interface mesh can be identified as the isosurface $\varphi_h^{n+1} = 0$ by solving the following 1D fixed point problems: for all $i = 1, \ldots, N_h^{\Gamma}$, find $\bar{s} \in [0, 1]$ such that

(14)
$$\chi_i(\bar{s}) = 0 \text{ and set } x_i^{n+1} = \gamma_i(\bar{s}).$$

Note that any fixed point method can be used here but since φ_h^{n+1} is often close to a linear function, a simple *secant method* converges very fast. It can be written as follows: if s_0 and s_1 are given numbers in [0, 1], we compute until convergence

$$s_{k+1} = s_k - \frac{s_k - s_{k-1}}{\chi_i(s_k) - \chi_i(s_{k-1})} \chi_i(s_k) , \quad k = 1, 2, \dots$$

The main difficulty here is to efficiently compute $\chi_i(s_k)$ for each k which requires a fast 3D interpolation of the function φ_h^{n+1} at point $\gamma_i(s_k)$.

2.3.2. Fluid mesh deformation. Above we have seen how to combine the levelset technique with a mesh deformation algorithm to identify at each time t_{n+1} the interface mesh Γ_h^{n+1} . Below we now describe how to propagate this deformation to the fluid mesh \mathcal{T}_h^n .

Let us first introduce a zone around the interface in which the mesh is allowed to be deformed. Let $z_m, z_p \in \mathbb{R}$, $z_m < z_0 < z_p$ be such that the volume in Σ between the altitude z_m and z_p belongs to the fluid domain (no other electric conductors). In most cells z_m is the top altitude of the cathode and z_p is the bottom altitude of the anodes. Recall that z_0 is the altitude of the interface in \hat{T}_h .

The mesh of the fluid domain \mathcal{T}_h^{n+1} at time t_{n+1} can be constructed from $\hat{\mathcal{T}}_h$ and Γ_h^{n+1} as follows:



FIGURE 3. "Elastic" deformation of the mesh.

• Aluminum : For each node $\hat{x}_i \in \hat{\mathcal{T}}_h \cap \Omega_{al}$ with altitude $z \in \mathbb{R}$, verifying $z_m \leq z \leq z_0$, we define $\alpha \in [0, 1]$ such that

(15)
$$\alpha = \frac{z - z_m}{z_0 - z_m}.$$

If $\gamma_i : [0,1] \to \mathbb{R}^3$ is the trajectory line associated with \hat{x}_i , then there exist $a, b \in \mathbb{R}$ such that $\gamma_i(a)$ is on the interface formed by the triangles of $\hat{\Gamma}_h$ and $\gamma_i(b)$ is on the interface formed by the triangles of Γ_h^{n+1} (see Figure 3). The node \hat{x}_i can then be moved as follows:

(16)
$$x_i^{n+1} = \hat{x}_i + \alpha \cdot (\gamma_i(b) - \gamma_i(a)).$$

The coefficient α assure that the deformation is proportional to the distance between a node and the interface in the original mesh. In particular if $z \approx z_m$ the deformation tends to zero. This can be seen as a kind of *elastic* deformation of $\hat{\mathcal{T}}_h$.

• Bath: For each node $\hat{x}_i \in \hat{\mathcal{T}}_h \cap \Omega_{el}$ with altitude $z \in \mathbb{R}$, verifying $z_0 \leq z \leq z_p$, we define $\alpha \in [0, 1]$ such that

(17)
$$\alpha = \frac{z_p - z}{z_p - z_0}.$$

The new position of \hat{x}_i is then defined by the equation (16) and, here, the deformation tends to zero if $z \approx z_p$.

For understanding, the Figure 4 presents an 2D simplified illustration where we impose a given deformation of the interface.

2.4. Electromagnetic equations. We have seen that the source term \vec{f} in Navier-Stokes equations (2) is the sum of the gravity $\rho \vec{g}$ and of the Lorentz forces $\vec{j} \times \vec{B}$, with \vec{j} the current density flowing trough the cell and \vec{B} the magnetic induction produced by the electric configuration of the conductors. Since the liquid metal is much more conductive than the electrolythic bath, the current distribution is strongly dependent of the position of the interface. In order to correctly simulate the motion of the fluids in a cell, one has to couple the evolutive Navier-Stokes equations (2) with the following time-dependent Maxwell equations in which we



FIGURE 4. Mesh deformation in a two dimensional simplified example. On the left $\hat{\mathcal{T}}_h$, on the right \mathcal{T}_h^{n+1} .

neglect the displacement currents (see [14] for a justification):

(18)
$$\begin{cases} \begin{cases} \frac{\partial B}{\partial t} + \nabla \times \vec{E} = 0, \\ \nabla \times \vec{B} = \mu_0 \vec{j}, & \text{in } \mathbb{R}^3, \\ \nabla \cdot \vec{B} = 0, \\ \vec{j} = \sigma \left(\vec{E} + \vec{u} \wedge \vec{B} \right), & \text{in } \Sigma. \end{cases}$$

where σ is the electric conductivity of the materials, μ_0 the magnetic permeability of the vaccum, \vec{E} the electric field and \vec{u} the velocity of the fluids. Since μ_0 is constant, ferromagnetic effects are neglected but in real situations, the ferromagnetic screen effect due to the steel shell containing the cell must be added (see [4]).

The numerical resolution of (18) will not be discussed here (see [5, 7] for details) but we just notice that the main difficulty is to efficiently compute the magnetic induction \vec{B} for all time t, since it is defined in the whole space \mathbb{R}^3 .

2.5. Numerical results.

In this section, we consider a simplified electrolytic cell whose geometry is a cylinder with a 3.5 cm base radius. The current is entering the cell from a partially immerged anode and is leaving it trough a cathode covering the whole base of the fluid domain (see Figure 5). Note that, in this experiment, surface tension effects are not negligibles since the capillary length is

$$\lambda_c = \sqrt{\frac{\gamma}{\Delta \rho g}} = 1.91 \text{ cm} \approx \text{ interface radius/2},$$

where $g = 9.81 \ m/s^2$, the surface tension coefficient γ for the aluminum/electrolyte interface is 0.5 N/m and $\Delta \rho = \rho_{al} - \rho_{el} = 140 \ kg/m^3$.

When the contact angle $\theta = \pi/2$, starting from an inclined interface as initial condition, Lorentz forces in the cell induce a rotational motion of the interface called the "metal pad rolling" (see [1], [8] and [9]). This is due to the big jump of



FIGURE 5. Geometrical configuration of the simplified cell.

conductivity between the bath and the aluminum ; indeed, the current path minimizes the Joule's effect and so there will be more current going trough the side of the interface close to the anode and less on the other. As the current density is uniformly distributed at the bottom of the cathode, horizontal currents are created in the aluminum and consequently we also have the creation of horizontal Lorentz forces, orthogonal to the current density and magnetic induction. Depending on the current and magnetic induction intensity, interface oscillations can either increase or decrease. In the aluminum industry a cell is said to be stable whenever the oscillations of the interface do not increase.

Suppose the contact angle on the lateral wall is greater than $\pi/2$. Then, in absence of electrical currents, the steady state of the interface will be a curved surface (see Figure 6). Moreover we can see that, in presence of electrical currents and external magnetic induction, this steady state is extremely stable in the sense that small perturbations vanish very quickly. This means that in this particular geometrical configuration, surface tension effects have a dominant contribution.

3. Dissolution and distribution of alumina in the bath

Let us assume that we are in presence of a stationary MHD configuration and consequently that Ω_{el} and \vec{u} do not depend on time.

In the industrial process, particles of alumina are added periodically at several locations between the anodes. It is assumed that the alumina particles follow the trajectories of the velocity field, dissolving at the same time and increasing thereby the concentration of liquid alumina. Liquid alumina is also transported by the



FIGURE 6. Steady state position of the interface. On the right: numerical simulation. On the left : X-ray picture of a lab experiment (source : RioTinto-Alcan, Laboratoire de recherche et fabrication, St-Jean de Maurienne (F)).

bath, diffusing slightly, and it is transformed into aluminum by electrolysis. Since aluminum is heavier than the bath it sinks to the bottom of the cell. This last phenomena is not contained in the model presented hereafter.

3.1. Model. We denote $c = c(\vec{x}, t)$ the concentration of liquid alumina and n_p the particle density of solid alumina. Alumina particles are supposed to be spherical and their radii change with time. In an Eulerian description, we thus have $n_p = n_p(\vec{x}, R, t)$, where R is the particle radius. The physical unit of the concentration is mol·m⁻³ and for the particle density it is (number of particles)·m⁻⁴. In a Lagrangian description we assume that the kinetics of the particle dissolution can be described by

(19)
$$R(t) = f(R(t))$$

along the trajectories $\vec{x}(t)$ given by the velocity \vec{u} . If, for example, dissolution kinetics are due to diffusion only, then f would be defined as (see for example [12],[16])

(20)
$$f(R) = -\frac{DM(c_{\text{sat}} - c(\vec{x}(t), t))}{\rho R},$$

where c_{sat} is the saturation concentration of liquid alumina in the bath, D is its diffusion coefficient, M is the molar mass of alumina, and ρ is its mass density.

Going back to the Eulerian reference frame, if the number of alumina particles in the bath is kept constant the particle density follows a convection equation in four dimensions,

(21)
$$\frac{\partial n_p}{\partial t} + \vec{u} \cdot \nabla n_p + \frac{\partial}{\partial R} (n_p f) = \psi,$$

where ψ stands for a source due to the injection of the particles. Observe that this equation does not change the number of particles but it does change the total mass of the particles because particle radii become smaller by dissolution.

The motion of liquid alumina concentration can be described by a convectiondiffusion equation,

(22)
$$\frac{\partial c}{\partial t} + \vec{u} \cdot \nabla c - D\Delta c = \dot{Q}(n_p),$$

where $\dot{Q}(n_p)$ is the source of liquid alumina which consists of two parts. On the one hand we have the mass which is lost by dissolution of the particles and which is the source of concentration of liquid alumina:

(23)
$$\dot{q}_1(n_p) = -4\pi \frac{\rho}{M} \int_0^\infty n_p f R^2 dR.$$

On the other hand, concentration diminishes by the electrolytic process. This is supposed to happen uniformly in the whole bath, giving

$$\dot{q}_2 = -\frac{I}{6FV},$$

where I is the total electric current, F the Faraday constant, and V the volume of Ω_{el} . The quantity $\dot{Q}(n_p)$ is the sum of $\dot{q}_1(n_p)$ and \dot{q}_2 .

We are thus looking for $c = c(\vec{x}, t)$ and $n_p = n_p(\vec{x}, R, t)$ verifying

(25)
$$\frac{\partial c}{\partial t} + \vec{u} \cdot \nabla c - D\Delta c = \dot{Q}(n_p)$$
 in $\Omega_{el} \times (0, T)$

(26)
$$\frac{\partial c}{\partial n} = 0$$
 on $\partial \Omega_{el} \times (0, T)$

(27)
$$\frac{\partial n_p}{\partial t} + \vec{u} \cdot \nabla n_p + \frac{\partial}{\partial R} (n_p f) = \psi$$
 in $\Omega_{el} \times (0, \infty) \times (0, T)$,

with f defined as above.

Without the injection of alumina particles (i.e. $\psi = 0$) and without electrolysis (i.e. $\dot{q}_2 = 0$) the total mass of alumina in the bath does not change over time, i.e.

(28)
$$\frac{d}{dt}\left(M\int_{\Omega_{el}}cd\Omega + \frac{4}{3}\pi\rho\int_{\Omega_{el}}\left(\int_0^\infty n_p R^3 dR\right)d\Omega\right) = 0.$$

3.2. Numerical scheme. Let us choose a discretization of the time t by

$$0 < t_1 < t_2 < \ldots < t_n < t_{n+1} < \ldots$$

If c^k, n_p^k are known approximations of c and n_p at time t_k , we compute c^{k+1} and n_p^{k+1} at time t_{k+1} as follows. On each time step $(t_{k+1} - t_k)$ we solve successively the equation for the concentration c and the equation for the particles n_p . For solving this last equation we use a splitting method in time in order to separate the space-time part from the radius-time part in Equation (27). With this splitting we avoid solving a problem in \mathbb{R}^4 . So we numerically solve:

• Convection-diffusion equation on (t_k, t_{k+1}) ,

(29)
$$\frac{\partial c}{\partial t} + \vec{u} \cdot \nabla c - D\Delta c = \dot{Q}(n_p^k), \qquad \text{in } \Omega_{el}$$

 $(30) c(t_k) = c^k.$

ΔR in μm	Δt in seconds	relative mass difference in $\%$
40	0.01	15.9
20	0.005	7.2
10	0.0025	3.1

TABLE 2. Relative mass difference of the total mass after 100 seconds and the total mass at time 0.

• Convection equation in space on (t_k, t_{k+1}) ,

(31)
$$\frac{\partial \hat{n}_p}{\partial t} + \vec{u} \cdot \nabla \tilde{n}_p = \psi, \qquad \text{in } \Omega_{el},$$

(32)
$$\tilde{n}_p(t_k) = n_p^{\kappa},$$

where \tilde{n}_p is a prediction of the quantity n_p .

• Particle dissolution on (t_k, t_{k+1}) ,

(33)
$$\frac{\partial n_p}{\partial t} + \frac{\partial}{\partial R}(n_p f) = 0,$$

(34)
$$n_p(t_k) = \tilde{n}_p(t_{k+1}).$$

We set $c^{k+1} = c(t_{k+1})$ and $n_p^{k+1} = n_p(t_{k+1})$. The convection-diffusion equation (29) and the convection equation (31) are solved by an Euler implicit method in time and by a continuous piecewise linear finite element method with SUPG stabilization in space. The particle dissolution (33) is solved using the method of characteristics on time and radius, where a piecewise constant discretization of the radius is used. All these problems are numerically solved on the same tetrahedral mesh of Ω_{el} . The linear systems are solved by the GMRES method.

3.3. Numerical results. We consider a cube of volume 1 m³. We impose a velocity field \vec{u} by assuming the cube to be a driven cavity. Thus, to compute the velocity field, we impose zero velocity at the bottom surface, zero normal velocity at the lateral surfaces $(\vec{u}.\vec{n}=0)$ and a velocity of 1 m/s from the left to the right at the top surface. The initial particle density n_p is as in Figure 7, top left, while the initial concentration of liquid alumina c is set to zero. Figure 7 shows the evolution of the particle density n_p and the concentration c. The evolution of the total mass, the mass of alumina particles and the mass of liquid alumina is shown in Figure 8. We notice that the total mass is slightly increasing. In Table 2 we show the relative mass difference between the total mass after 100 seconds and the total mass at the beginning. We do this on a fixed mesh, for different time steps Δt and for different radius meshsizes ΔR . We see that the relative mass difference is divided by two if Δt and ΔR are divided by two.

4. Conclusion

This paper presented : i) a physical model for the movements of the interface between liquid aluminum and electrolyte in an aluminum electrolytic cell, ii) a physical model for the alumina dissolution and convection in these fluids. Numerical methods to effectively approximate and compute the time-dependent solutions to these models have been introduced as well and some results obtained on simplified geometries have been presented.



FIGURE 7. Evolution of particle density n_p (left) and concentration c (right)



FIGURE 8. Evolution of the total mass, the mass of alumina particles and the mass of liquid alumina during the simulation

Our final aim is the simulation of an industrial configuration of a cell in a reasonable time and with a good precision taking into account fluid motion, solidifications of ledges, ferromagnetic effects and alumina convection-dissolution.

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References

- P.A. Davidson, R.I. Lindsay, Stability of interfacial waves in aluminium reduction cells, J. Fluid Mech., 362:273-295, 1998.
- [2] J. Descloux, M. Flück, M. V. Romerio, Spectral aspects of an industrial problem. In Spectral analysis of complex structures. Hermann éditeurs des sciences et des arts, Paris, 17-33, 1995.
- [3] J. Descloux, M. Flück, M. V. Romerio, A modelling of the stability of aluminum electrolysis cells. In Nonlinear partial differential equations and their applications. Collège de France Seminars, Vol XIII. D. Cioranescu and J.L. Lions editors, Pitman Research Notes in Mathematics Series 391. Addison Wesley Longman 1998, 117-133.
- [4] M. Flück, T. Hofer, A. Janka, J. Rappaz, Numerical methods for ferromagnetic plates, Scientific report in Analysis and Numerical Analysis, EPFL, 2008.
- [5] M. Flück, A. Janka, C. Laurent, M. Picasso, J. Rappaz, G. Steiner, Some Mathematical and Numerical Aspects in Aluminum Production, Journal of Scientific Computing, 2008.
- [6] M. Flück, J. Rappaz, Y. Safa, Numerical Simulation of thermal problems coupled with magnetohydrodynamics effects in aluminum cell, International Journal for numerical methods in engineering, Applied Mathematical Modelling, In Press, Corrected Proof, Available online 29 February 2008, doi:10.1016/j.apm.2008.02.011.
- [7] M.Flück, J. Rappaz, G. Steiner, On a domain decomposition method for numerically solving a magnetic induction problem. Scientific report in Analysis and Numerical Analysis, EPFL.

- [8] J.-F. Gerbeau, C. Le Bris, T. Lelièvre, Mathematical methods for the magnetohydrodynamics of liquid metals. Numerical Mathematics and Scientific Cumputation Series. Oxford Science Publications 2006.
- J.-F. Gerbeau, C. Le Bris, T. Lelièvre, Metal pad roll instabilities, proceeding of the 2002 TMS Annual Meeting and Exhibition, Light Metals, 483-487, 2002.
- [10] J.-F. Gerbeau, T. Lelièvre, Variational formulation of the Generalized Navier Boundary Condition, 2nd International Conference on Scientific Computing and Partial Differential Equations, Hong-Kong, 2006.
- [11] R. Glowinski: Finite element methods for incompressible viscous flow. Handbook of Numerical Analysis, Vol IX. P.G. Ciarlet and J.L. Lions editors. North-Holland, Elsevier 2003, 498-541.
- [12] R.G.Haverkamp, B.J.Welch, Modelling the dissolution of alumina powder in cryolite, Chemical Engineering and Processing, 37(1998), 177-187.
- [13] P.L. Lions, Mathematical Topics in Fluid Mechanics, Vol. 1 : Incompressible Models, Oxford University Press, Oxford, UK, 1996.
- [14] D.Munger, Simulation numérique des instabilités magnétohydro-dynamiques dans les cuves de production de l'aluminium, Master's thesis, Université de Montréal, 2004.
- [15] J.A.Sethian, Level Sets Methods. Cambridge Monographs on applied and Computational Mathematics, 1996.
- [16] J.Wang, D.R.Flanagan, General Solution for Diffusion-Controlled Dissolution of Spherical Particles. 1. Theory. Journal of Pharmaceutical Sciences, 88(1999), no.7, 731-738.

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504