Dynamic simulation of a deposition process

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Abstract

A new solution for the computation of a current distribution with a moving boundary is presented. The numerical scheme is based on the capabilities of two commercial codes FEMLAB 2.3 and MATLAB 6.1. First a computation is carried out with FEMLAB which provides the electrical potential distribution at the initial time. The FEM problem and its initial solution are then saved as a MATLAB program. The MATLAB code containing the whole – draw, mesh, solve, plot – sequence is then modified to automate the iterative process. A movie is finally made from the successive solutions stored as JPEG pictures. A metal deposition process is presented as a simple test but difficult benchmark. This method is validated and can be easily extended to much more complex coupled electrochemical processes.
Keywords: FEM, primary current distribution, electrodeposition, moving boundary.

1 Introduction

The problems associated to evolutionary shape change are numerous and difficult to solve. They appear in various fields of electrochemical engineering such as electrodeposition [1,4,5,7,10–16], electro-machining [17], electropolishing [8], and corrosion processes [6]. Most popular commercial codes are now able to compute primary, secondary or tertiary current distributions for any geometry. If it is easy to compute the current distribution for a given geometry, it is very difficult and time consuming to deduce the time evolution of the moving interface related to the current distribution.

One of the first attempts to model the moving deposit profiles is probably due to Deconinck [1] in 1985. Since then, many authors tried to compute moving profiles by use of different numerical methods such as finite difference method (FDM) [6,10,12,14], boundary element method (BEM) [1,5,9], finite element

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method (FEM) [7,11,12], or specific methods [16]. All these authors used the same manual iterative process but Chauvy and Landolt [17] who developed a specific FORTRAN 77 code to compute the shape evolution of cavities produced by multistep electrochemical micromachining (the CREVICER code [18] developed for corrosion purposes does not have yet this capability).

The iterative process is simple. A first current distribution gives the thickness of the first layer deposited during the initial time step by solution of the Faraday's law. A new profile is then calculated and the geometry is modified consequently. Reiterating this manual process makes possible the determination of the shape evolution with time. However, this process is time consuming even for a very simple geometry since each time step requires a new – draw, mesh, solve, plot, compute the new profile – sequence. So there is a real need of automating the iterative process for industrial purposes or simply to check different initial configurations or to test corrosion scenarios for example.

This work intends to test a new solution based on the capabilities of the two popular commercial codes FEMLAB [2] and MATLAB [3]. The tests are here voluntarily restricted to electrochemical deposits corresponding to primary current density distributions. Calculations are simpler and such tests are more severe since the primary current distribution is known to give steep profiles and be very sensitive to the mesh quality [12].

2 The current distribution and the moving boundary

Figure 1 shows the displacement of the metal-electrolyte boundary due to an electrochemical deposit carried out during a time step.

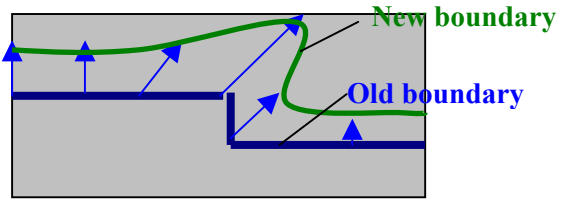

Figure 1: The moving boundary problem.

The local growth rate at any point i is governed by the Faraday's law:

$$\frac{\mathrm{d}\vec{h}}{\mathrm{d}t} = \sum_l \frac{M_l}{z_l F \rho_l} \vec{J}_l \tag{1}$$

where J_l is the local mean current density (A/m^2), M_l the molar mass (Kg/mol), ρ_l the density (Kg/m^3) and z_l the charge of an ion l present in solution. In an industrial electrolyte each individual λ_l coefficient defined as:

$$\lambda_l = \frac{M_l}{z_l F \rho_l} \tag{2}$$

is not necessarily known, so we assume that the global coefficient λ associated with the global current density J is known. Eqn. (1) is then simplified:

$$\frac{d\vec{h}}{dt} = \lambda \vec{J} \tag{3}$$

Eqn. (3) can be discretized:

$$\Delta \vec{h}_i = \lambda \vec{J} \Delta t \tag{4}$$

Let x_i and y_i the coordinates of a point i placed on the moving boundary. At the (n+1)th iteration corresponding to time $t + \Delta t$, the coordinates of point i are obtained from the coordinates computed at iteration n:

$$x_i^{n+1} = x_i^{n} + \Delta t \lambda J_x \tag{5}$$

$$y_i^{n+1} = y_i^{n+1} + \Delta t \lambda J_y \tag{6}$$

The time evolution of any point i is thus obtained from the local current density vector (J_x, J_y) deduced from the electrical potential V:

$$\vec{J} = -\sigma \vec{\nabla}.V \tag{7}$$

Then the displacement of the points of the moving interface requires the preliminary solution of the Laplace's equation:

$$\vec{\nabla}(-\sigma \vec{\nabla}.V) = 0 \tag{8}$$

When using a numerical code based on any numerical method, the first step consists in the solution of Laplace's equation at initial time. Then the current density J can be post processed from V and the new co-ordinates of each point of the border are computed using eqn. (5,6). A simple iteration scheme using eqn. (5,6) is necessary to compute the interface evolution.

Two main different methods can be considered to model the moving boundary evolution: (i) transform the electrolyte in metal by modifying the physical properties in the deposit area (ii) deform locally the mesh near the interface to take account of the move. Both methods were tested previously and did not give satisfaction. We noticed that FEMLAB preserves the topology of the related structure named "geom" in the code when the nodes situated on the

moving boundary are manually shifted. Then it was possible to automate the moving of nodes and mesh the new geometry at each iteration. This method is described hereafter.

2.1 Electrochemical deposition in a trench

The method was checked using the very simple 2D scheme of Figure 2 which represents a simple electrochemical cell where a hole in the metal (grey) is covered by an electrolyte (white). This problem is frequently encountered in the semiconductor industry which uses electrochemical deposition processes in trenches [10,14,15]. In such conditions this very simple geometry provides a severe test for a numerical computation scheme since the primary current distribution exhibits two steep peaks at the borders of the trench.

2.2 The boundary conditions

The boundary conditions are presented in Figure 2. Dirichlet conditions are applied on anode and cathode, and symmetry conditions are applied on each side.

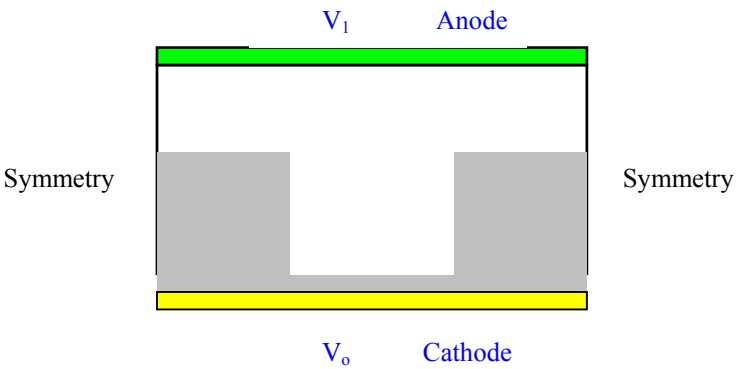

Figure 2: The boundary conditions for a trench.

3 The iterative computational process with FEMLAB-MATLAB

FEMLAB is a code based on the finite element method (FEM), developed and marketed since 1986 by COMSOL [2]. The problem is solved in the following steps thanks to a user friendly graphical interface: draw the geometry, define the boundary conditions, give the physical properties for each subdomain, mesh, refine the mesh if necessary, solve, plot the solution.

FEMLAB was first used to draw the cell and to define the physical problem. For example, in this case, a first rectangle ABCD was drawn figuring the electrolyte (zone 1). The metal part (zone 2) was obtained by tracing a polygon

starting from A and passing by the points E, F, G, H, I, J, D, A. Actually, segments were only used here to simplify the drawing, but one could just as easily use Bezier curves for a design including arcs.

In this scheme, it must be noticed that the end points of the segments are automatically selected as nodes by the mesh generator (the length of the segments constituting the moving boundary are not necessarily uniform).

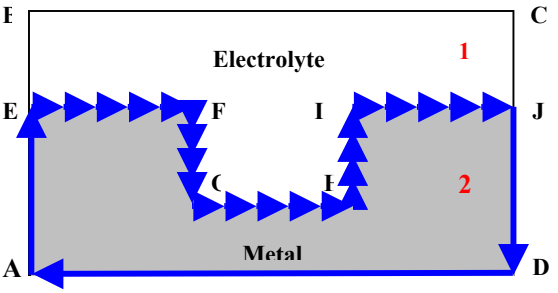

Figure 3: Drawing the trench with FEMLAB.

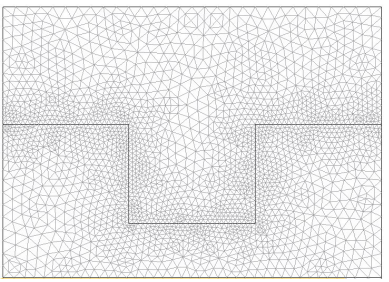

Figure 4: Initial Mesh.

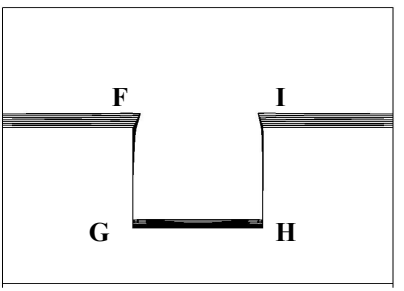

Figure 5: First deposit steps.

Thereafter the algorithm moves only the nodes located at the ends of the segments appearing between the points E and J of Figure 3. This set of segments constitutes the border between electrolyte and metal, border which will be moved during the deposit process. Figure 4 shows the mesh given by the automatic mesh generator of FEMLAB after the grid initialization and one refining process. Figure 5 shows the five first successive steps that were hand made using the global algorithm described in Figure 6. It can be seen that the deposit is not perfectly symmetrical and the points F and I tend to approach, while the points G and H go up. These observations are discussed hereafter.

3.1 Automation of the iterative scheme

When the solution is obtained with FEMLAB it can be saved as a genuine MATLAB file. This file includes the whole structure describing the topology, the

boundary conditions, the physical data and all the information necessary for the FEM solution. Since this ASCII file can be edited and modified, it is thus easy to add in the program the loop on the steps 4–7 of Figure 6 to obtain the iterative scheme and the periodic plot of the moving boundary solution stored as a JPEG file. Collecting the successive pictures it is then easy to make a movie showing deposit evolution with the time. Only stages 4 and 7 of the scheme must be programmed, all the other stages appearing without any change in the original MATLAB file.

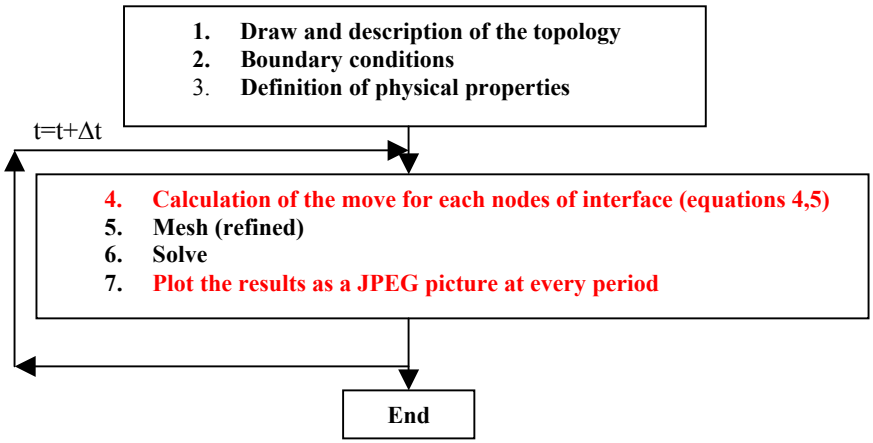

Figure 6: Iterative computation algorithm.

Due to the structure of the code, it is remarkable that any displacement of the points located at the moving interface does not change the boundary conditions, as long as the points remain in the same domain. Moreover, since the topological structure is not altered, the region where the deposit replaces the electrolyte receives automatically the physical properties of metal. This MATLAB equivalence of the FEMLAB solution has the advantage of giving direct access to the geometrical structure and to all the parameters and the physical data used in the computations in every point of each sub domain.

4 Numerical results

Figure 7 shows in black the deposit evolution for 200 iterations when the edges of the hole are tilted. It appears immediately that the side peaks grow more quickly than the deposit in the centre of the hole. It can be noticed that after 150 iterations a second small peak appears at the side of the larger one. This peak can be interpreted as a shadow effect of the principal peak. A corner effect is also seen at the edges of the central part of the hole. These effects are overvalued and would be less important in a real process, since this calculation of primary distribution of current neglects all the limiting effects, particularly the overpotentials.

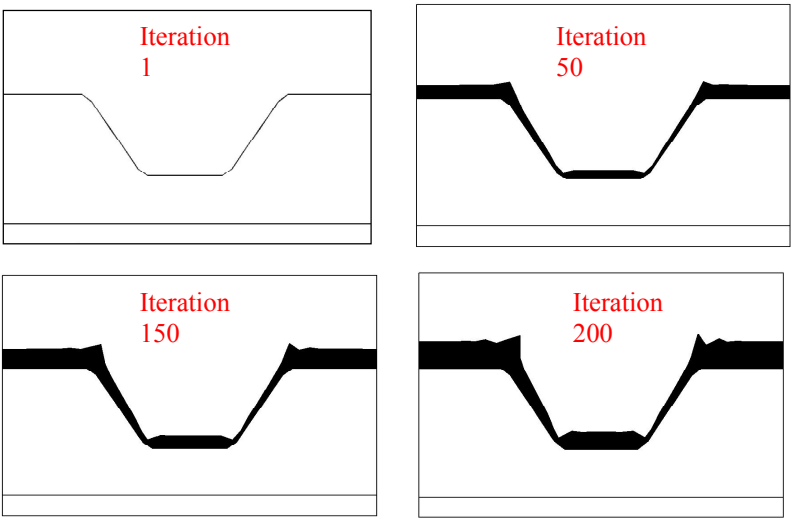

Figure 7: Evolution of deposit for 200 iterations.

Calculation and saving of the successive images on a PC - HP VECTRA 6800 last about only 20 minutes for the two examples presented above.

Thus when the methodology is developed, the user can easily carry out many tests very quickly. The same methodology can be used for research purposes to rapidly evaluate some assumptions.

5 Problems related to the algorithm

Actually, the algorithm presented in Figure 6 failed in some specific cases. Three problems were detected in the case of the trench with vertical edges.

5.1 Junction of two consecutive nodes

This problem appears when the length of a vector included in the moving boundary decreases during the iterative scheme. The nodes of the vector become closer and closer and can touch each other. This event is not improbable if we look at points I and F of Figure 5. When two nodes are confounded, the topology of the geometry is modified and this is not compatible with the present algorithm. Moreover, when two consecutive nodes come close, the automatic meshing generates locally smaller and smaller triangles, and then the number of degrees of freedom of the system blows up and the iterative process stops for lack of memory space. Then the user must draw a new geometry that integrates the fusion of the confounded nodes and restart the automatic process.

5.2 Unsymmetrical evolution of a symmetrical geometry

Our tests were systematically carried out on symmetrical cells in order to point out some possible numerical defects related to the algorithm. The Figure 8 exhibits a symmetry defect observed at the beginning of the computation when plotting the current density distribution along the path ab. This problem is obviously related to the discretization scheme which can produce small but significant numerical cut out errors on the large peaks appearing mainly in primary current distributions.

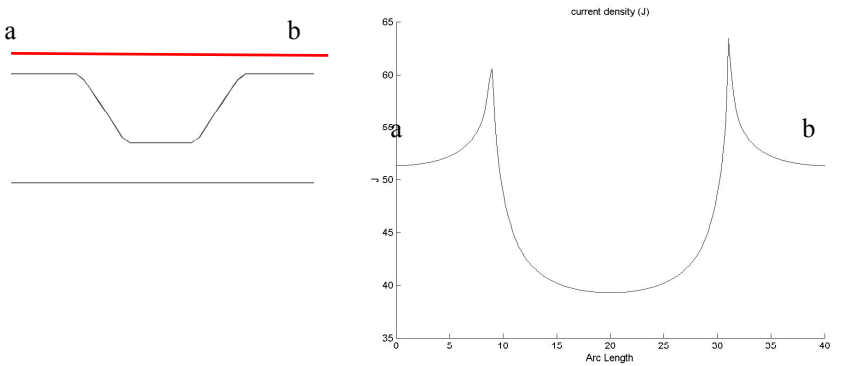

Figure 8: Dissymmetry of current distribution along ab.

Such a cut out effect is unpredictable: it can be compensated from an iteration to the other or can be accumulated. It is inherent to the numerical method and cannot be fixed without an improvement of the precision of the numerical computation. It probably would be less apparent for the smoother peaks of the secondary or tertiary current distributions.

5.3 Wrong displacement towards the inner part of metal

This defect did not appear in the electro polishing tests. It was only noticed in the most severe tests of electrodeposition. This is due to a wrong displacement when the current flow lines come successively through metal, electrolyte and metal in the vicinity of points H and G of Figure 3. These irregular node displacements (1) as well as a lack of symmetry of Figure 8 were attributed to the finite precision of the computation. The problem was fixed by checking that every deposit displacement was well done in the proper direction, i.e. from metal to electrolyte.

6 Conclusions

This work has shown that it is possible to automate the tedious manual computation of a moving interface in primary distribution of current by using the

FEMLAB and MATLAB codes successively. It also revealed the limitations inherent to the numerical discretization. The numerical instabilities encountered with this simple algorithm could be fixed and the defects would not appear in secondary current distribution. So the iterative scheme is now well validated.

This work is obviously criticisable from a strict electrochemical point of view insofar it does not take into account the physical limiting phenomena. However, as the algorithm is now validated, it is possible to automate the treatment of coupled phenomena thank to the Multiphysics capabilities of FEMLAB. Calculations would be heavier, but in fact there is no additional issue. Taking account of nonlinear boundary conditions would either be straightforward since they do not interfere with the iterative algorithm described here. The first results allow considering more complex applications taking into account of current distribution coupled with heat, mass or momentum transfers. Now the problems related to the moving of nodes are fixed, the algorithm works properly and a movie describing the time evolution of a profile can be obtained in a couple of hours with a cheap usual personal computer.

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