ELECTROPOLISHING ON SINGLE-CELL: (TESLA, RE-ENTRANT AND LOW LOSS SHAPES) COMSOL MODELLING

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Abstract

In the framework of improvement of cavity electropolishing, modelling permits to evaluate some parameters not easily accessible by experiments and can also help us to guide them. Different laboratories (DESY, Fermilab) work on electro or chemical polishing modelling with different approaches and softwares. At CEA Saclay, COMSOL software is used to model horizontal electropolishing of cavity in two dimensions. The goal of this study has been motivated by improvement of our electropolishing setup by modifying the arrival of the acid. The influence of a protuberant cathode has been evaluated and compared for different shapes of single cell cavities: TESLA, ILC Low Loss (LL_{ILC}), and ILC Reentrant (RE_{ILC}).

Keywords: **single cell, electropolishing, COMSOL modelling.**

A version of this present report will be published as a CARE note for the WP 5.1 (Best EP parameters on single cell).

Table of contents

Introduction

To build the future International Linear Collider (ILC), “cold” technology has been selected. It is based on superconducting radiofrequency (SRF) cavities. Electropolishing is presently accepted like the most efficient surface treatment to reach high gradients higher than 40 MV/m for niobium RF cavities. However, this technique presents the disadvantage to be not very reproducible. Experimental researches try to improve it but modelling is also an interesting way to better understand electropolishing. It can also direct experiments. In this study, COMSOL software is used to model horizontal electropolishing on 2D single cell cavity (TESLA, RE_{ILC} and LL_{ILC}) [1, 2]. After describing general equations used for modelling, the first step will be to model our experiments. Then, new configurations are studied in order to improve acid flow in the cavity and by this way electropolishing.

1. General equations and hypothesis

1.1. Reminders about electropolishing and its kinetics

Previous studies [3, 4] have been carried out thanks to the multiphysics coupling of COMSOL. Here, chemical reactions from niobium electropolishing are introduced.

The general chemical equation for niobium oxidation is:

$$\mathbf{2\,Nb + 5\,H_2O \rightarrow Nb_2O_5 + 5\,H_2} \qquad (1)$$

Then the oxide layer Nb_2O_5 is attacked by the hydrofluoric acid following the equation [5]:

$$\mathbf{Nb_2O_5 + 12\,HF \rightarrow 2\,HNbF_6 + 10\,H_2O} \qquad (2)$$

For modelling, it is assumed that the oxide layer is already formed and so only equation (2) is considered. The kinetic v of this reaction is second order; so it is equal to

$$v = k\,[Nb_2O_5]\,[HF] \quad \text{where k is a kinetic constant in mol.l}^{-1}.\text{s}^{-1} \qquad (3)$$

As $HF \rightarrow H^+ + F^-$, it is important to note for all modelling and the following report HF concentration [HF] is not directly equal to the concentration $[H^+]$ because there is also H^+ coming from H_2SO_4 acid present in the electrolyte. So for all the modelling and the following report, $[HF] \equiv [F^-]$ only.

Considering equation (3):

$$v = \frac{1}{2} R_{[HNbF6]} \quad \text{where R is the net creation rate (mole.l}^{-1}.\text{s}^{-1}) \qquad (4)$$

By combining equation (2), (3) and (4), we obtain:

$$\mathbf{R_{HNbF6} = 2\,k\,[Nb_2O_5]\,[HF]} \qquad (5)$$
$$\mathbf{R_{Nb2O5} = -\,k\,[Nb_2O_5]\,[HF]} \qquad (6)$$
$$\mathbf{R_{HF} = -\,12\,k\,[Nb_2O_5]\,[HF]} \qquad (7)$$

If x is considered as the number of moles of $HNbF_6$ produced by reaction (2), we have:

$$v = \frac{1}{2}\frac{dx}{dt} = k\,([Nb_2O_5]_{initial} - \frac{1}{2}x)*([HF]_{initial} - 6x) \quad (8)$$

By solving equation (8), it is found that after a while, x reaches a limited value. During modelling, a limited concentration in $HNbF_6$ (and so in Nb_2O_5 and HF) was effectively found. As each concentration is directly connected to the other, only results for [HF] will be presented.

1.2. Convection diffusion and Navier-Stockes equations.

Two equations are considered [3, 4]:

- The mass balance equation:

$$\frac{\partial c_i}{\partial t} + \nabla N_i = R_i \quad (9)$$

Where c_i is the concentration of species i ($mol.m^{-3}$), N_i its flux ($mole.l^{-1}.s^{-1}$) and R_i its net creation rate ($mole.l^{-1}.s^{-1}$).

- The Nernst-Planck equation:

$$N_i = -D_i \nabla c_i - z_i \omega_i F c_i \nabla \phi + u c_i \quad (10)$$

The first term relates the diffusion transport, the second the electromigration and the last one the convection. Parameters D_i, Z_i, ω_i, Φ and u represent respectively diffusion coefficient, valence, mobility, electrostatic potential and velocity of species i. F is the Faraday constant (96 485 $C.mole^{-1}$). As transport by electromigration is very weak compared to transport by convection, it can be neglected [3, 4].

The value for the diffusion coefficient is chosen equal to 10^{-8} $m^2.s^{-1}$ as determined in [3]. Mobility is defined in COMSOL by the following expression:

$$\omega_i = \frac{D_i}{RT} \quad (11)$$

- By combining equations (9) and (10), the general equation to solve is:

$$\frac{\partial c_i}{\partial t} + \nabla(-D_i \nabla c_i - z_i \omega_i F c_i \nabla \phi + u c_i) = R_i \quad (12)$$

where R_i are defined in equations (5,6,7).

- The equation (12) is coupled to Navier-Stockes equation for an incompressible fluid, in order to model the flux going through the cavity:

$$\rho \frac{\partial u}{\partial t} - \eta \nabla^2 u + \rho(u.\nabla)u + \nabla p = G \quad (13)$$

$$\nabla.u = 0 \quad (14)$$

η is the dynamic viscosity of the fluid (10^{-3} $kg.m^{-1}.s^{-1}$)
ρ the fluid density (1800 $kg.m^{-3}$)
u the fluid velocity ($m.s^{-1}$)
p the pressure ($N.m^{-2}$)
G ($N.m^{-3}$), the resultant force, only the gravity is considered here.

2. Single-cell cavity modelling

2.1. Experimental electropolishing set up at CEA Saclay

In our electropolishing test bed, niobium cavity turns horizontally around the aluminium cathode. The standard bath is composed by one volume of hydrofluoric acid for nine volumes of sulphuric acid. As H_2SO_4 is the main constituent, its density and viscosity are selected in the model and defined in equation (13).

The acid comes in by a hole located in the middle of the cathode and arises up to the extremity of the cavity as the following figure 1. Its entrance velocity is around 5.10^{-3} m/s. Half of the cavity is filled with the acid.

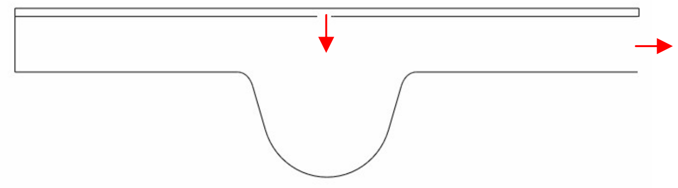


Figure 1: Acid flux during electropolishing

Intuitively this central fluid inflow contributes for an efficient acid renewal in the equator area of the cavity and for an acid stagnation (dead end) in the left beam tube (figure.1). Modelling should ratify such intuitive analysis.

2.2. Modelling Saclay electropolishing

Firstly, we will considerer the fluid velocity as constant during the electropolishing and the cavity full of acid before starting simulation. Secondly, by simplification, cavity spinning is not taking into account in this preliminary study. At last, an electropolishing can last up to 6 hours, so HF concentration will be studied up to 20 000 s (5h30). First results give us the fluid velocity for each shape (figure 2).

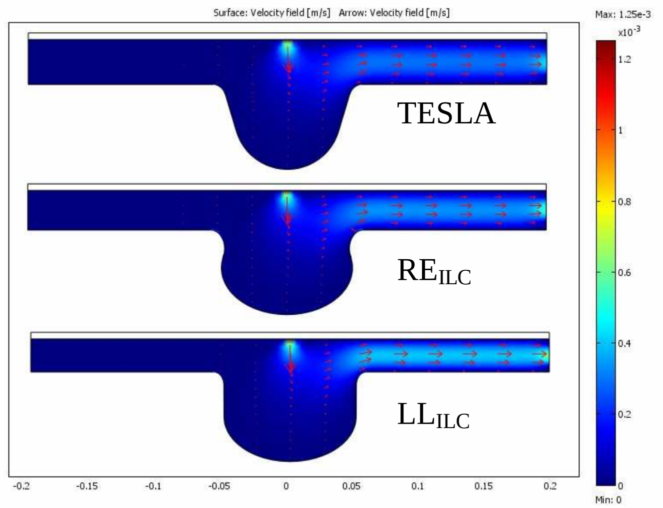
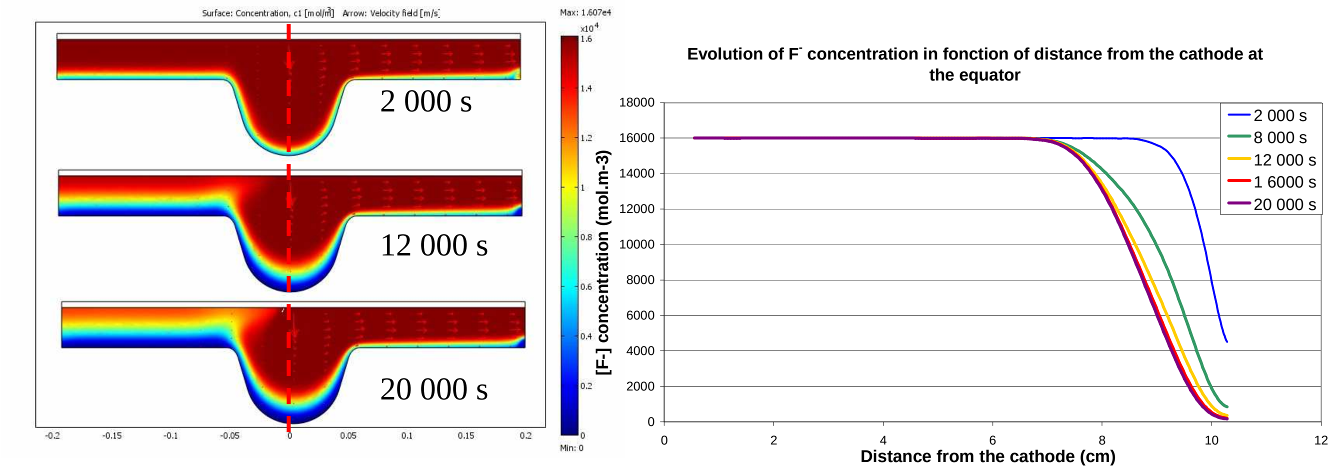


Figure 2: Fluid velocity for TESLA, RE_{ILC} and LL_{ILC} shapes

This study confirms that a dead end exists on the left beam tube, however without a very well acid renewal in the cavity body as expected in 2.1 section. So, once the cavity has been filled and once a stationary state has been reached with the acid circulation, an “old” acid might stay in the bottom of the cell. By studying the HF concentration versus time, it is possible to confirm this assumption. HF concentration is shown at the beginning (2 000 s), in the middle (12 000 s) and at the end (20 000 s) of the electropolishing.

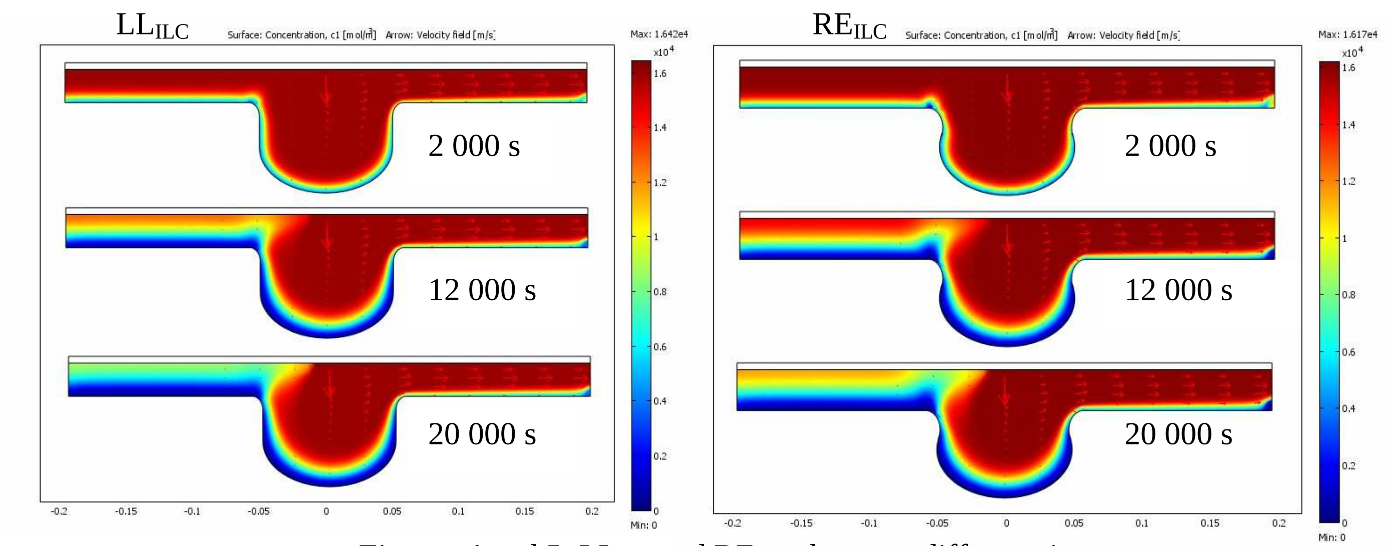
For TESLA shape, results are the following (figures 3a and b):



Figures 3a. 3b.: [F-] at different times for TESLA cavity
a: longitudinal, b: transversal axis at the equator – dashed line

From figure 3a., it is easy to see that there is not a symmetric electropolishing in the cavity: there is a beam tube where the HF concentration is lower than in the other one. The same observation can be made for the iris. After a long time (>16 000 s), no evolution of acid concentration is reported in the bottom of the cell. The limit concentration as predicted by the differential equation (8) is reached. On the figure 3b., an acid depletion with time is easily noticed at the equator. An aging of the acid in this cavity part could explain a bad electropolishing at the equator compared to the beam tubes.

Results on TESLA cavity can be compared to other shapes like RE_{ILC} and LL_{ILC}:



Figures 4 and 5: LL_{ILC} and RE_{ILC} shapes at different times

The same analysis can be made: there is a bad renewal of acid in the bottom of the cell and in the left beam tube. Furthermore, a strong dissymmetry in acid renewal of the "left" iris compared to the other one can be noticed, especially for RE_{ILC} shape.

Regarding these modelling results highlighting a possible non symmetric electropolishing in the experimental case, a new configuration of acid inflow should be considered (cf. figure 6). It should be expected:

- A similar electropolishing in the both beam tubes.
- A better sweeping of the cell.
- A symmetric electropolishing of e both iris for RE_{ILC} shape.

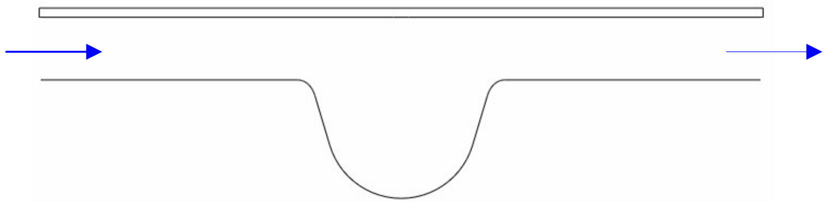


Figure 6: New acid inflow

2.3. New configuration study

The fluid velocity and the HF concentration are studied for each shape, as previously.

2.3.1. Study of the velocity field

The results for velocity field are presented on the following pictures:

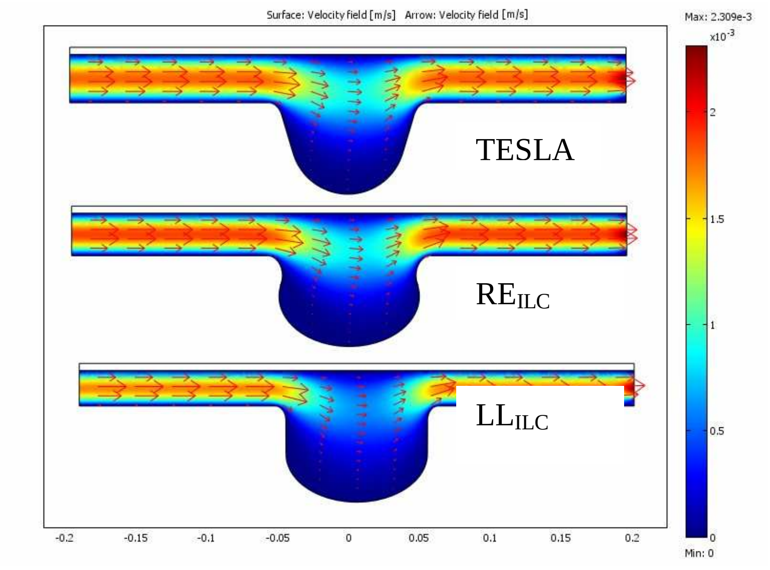


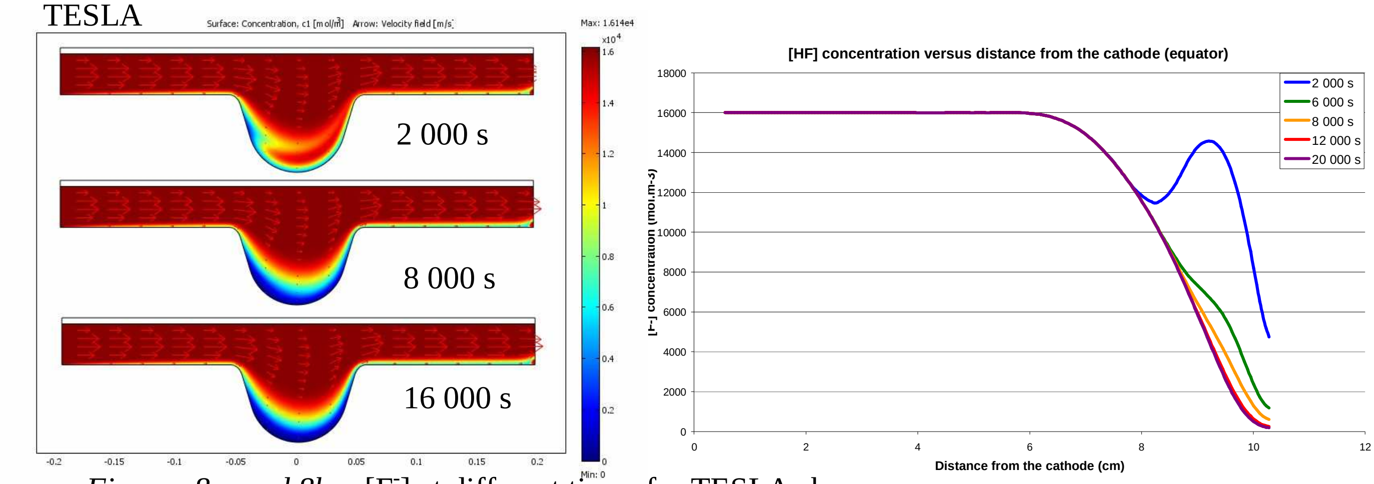
Figure 7: Fluid velocity for TESLA, RE_{ILC} and LL_{ILC} shapes. New acid inflow.

As expected, there is a very good and symmetric sweeping of the beam tubes. However the velocity of the fluid is very low in the bottom of the cell. That means a bad renewal of the acid.

2.3.2. HF Concentration versus time

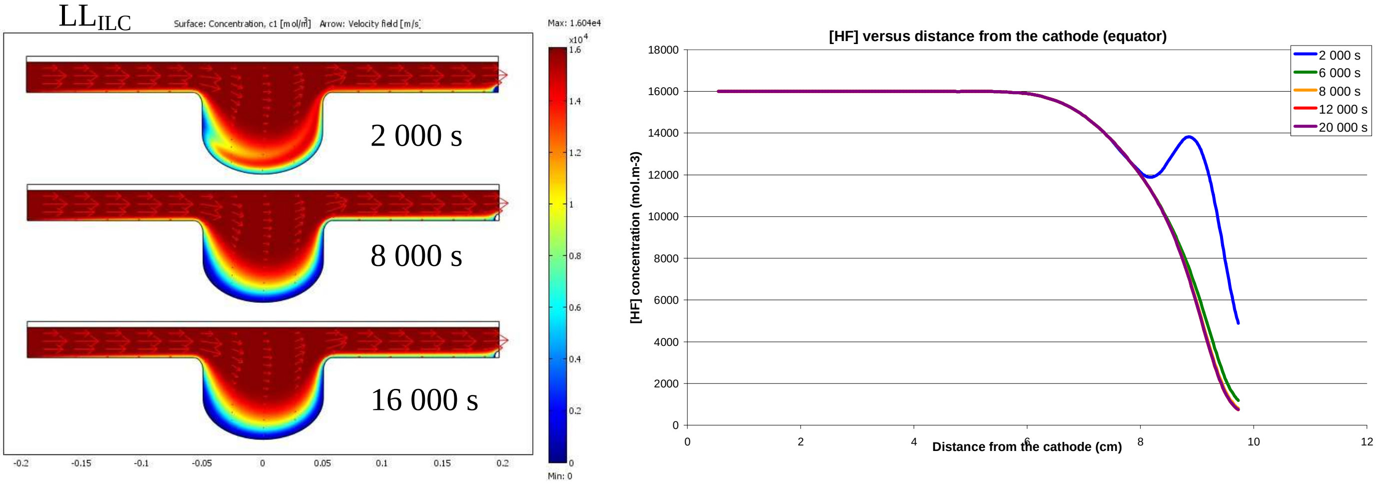
This hypothesis is confirmed by the HF concentration profile:

TESLA



Figures 8a. and 8b. : [F⁻] at different times for TESLA shape
a: longitudinal and b: transverse profile at the equator

LL_{ILC}



Figures 9a. and 9b. : [F⁻] at different times for Low Loss shape
a: longitudinal and b: transverse profile at the equator

For these two shapes, a dissymmetric electropolishing is observed at the beginning. Then there is a uniform electropolishing in the cell correlated to a depletion of HF in the bottom of the cell (figures 8b. and 9b.): after 8 000 s for LL_{ILC} and 12 000 s for TESLA shape, there is no evolution of HF concentration. From this analysis, few conclusions can be related:

- With this new acid inflow, LL_{ILC} cavity might be electropolished faster than the TESLA one.
- Electropolishing in the cell is faster (in the previous configuration, there is no evolution of HF concentration after 16 000 s).
- However, at the beginning (2 000 s), even if both beam tubes are electropolished in the same way, the cell is not electropolished symmetrically.

In the next paragraph, the evolution of HF concentration for RE_{ILC} shape will be especially analysed at the iris and in the bottom of the cell.

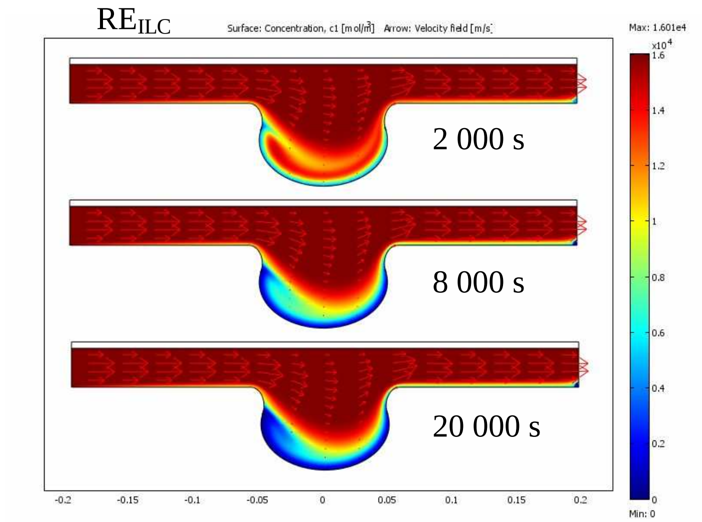


Figure 10: F^- concentration for RE_{ILC} shape for different times

For RE_{ILC} shape, this acid inflow configuration is not very favourable: even after 20 000 s, there is still a dissymmetric electropolishing in the cell.

To conclude, in spite of a good electropolishing of beam tubes, this new acid inflow is not optimized for symmetric electropolishing of the central part of the cell, especially for the RE_{ILC} shape. A solution should be found to avoid such trouble by adding a protuberance on the cathode to force the acid to sweep the bottom of the cell.

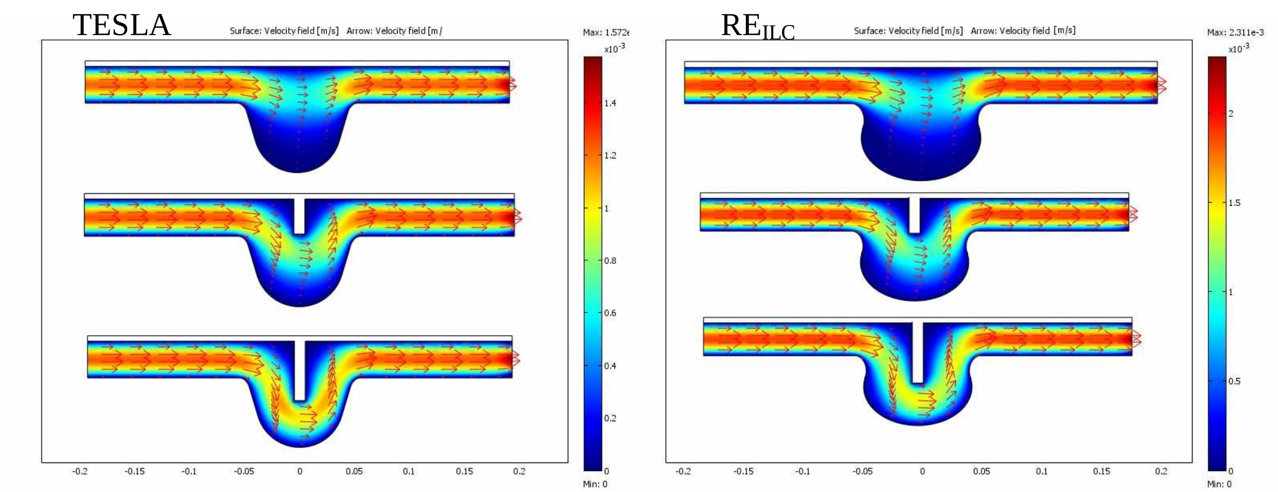
2.4. Protuberance on the cathode

The protuberance is located in the middle of the cathode. Two different sizes are studied:

- Protuberance length quite smaller than the beam tube radius. It could be introduced easily in the cavity.
- The other one is longer. It could be designed like an "umbrella" system.

2.4.1. Study of the velocity field

The velocity field of the acid is studied for both TESLA and RE_{ILC} shapes:

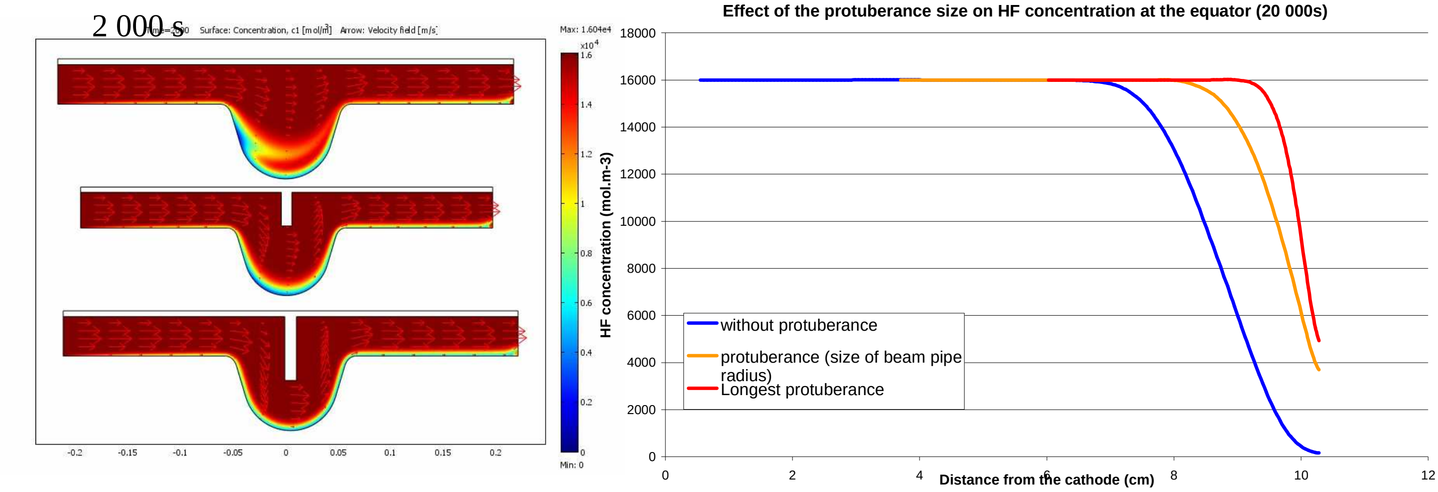


Figures 10 and 11: Study of velocity field for TESLA (10) and RE_{ILC} (11) shapes with a protuberance on the cathode

As expected, the effect of the protuberance is very efficient: it forces the fluid to sweep the bottom of the cell; a better renewal of the acid can be hoped.

2.4.2. HF concentration study (TESLA shape)

The influence of a cathode protuberance is observed at different times:



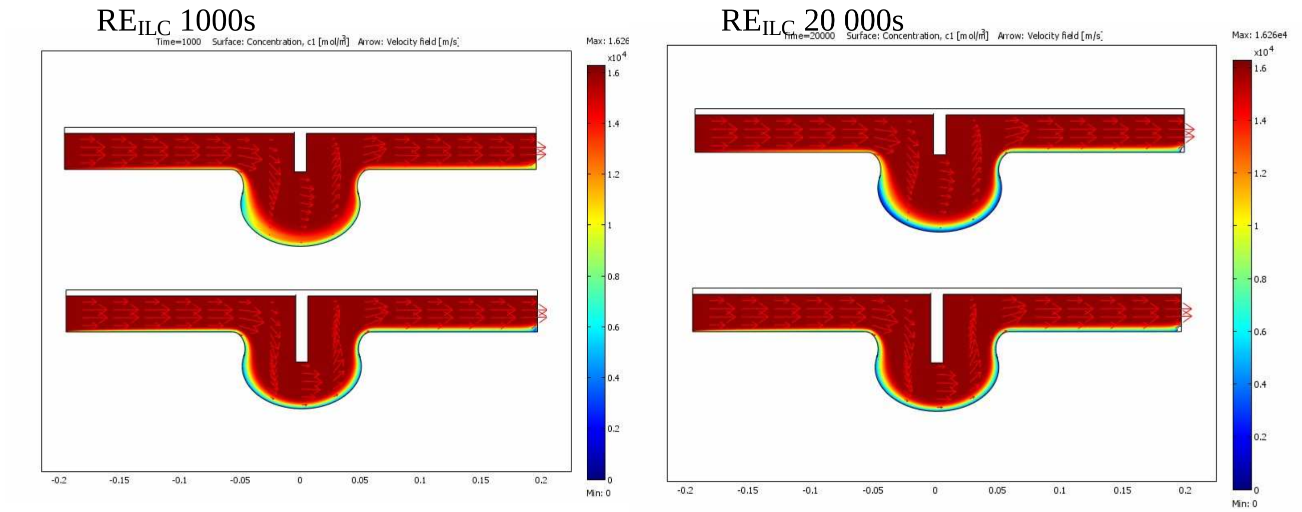
Figures 12a. 12 b.: Influence of the protuberance on the TESLA mono cell cavity
a: longitudinal profile at 2 000 s, b: transverse profile at 20 000 s

These results at 2 000 s and 20 000 s confirm those found for the velocity fluid. There is a beneficial effect on electropolishing with the protuberance:

- No more perturbation in the cell after 2 000 s (figure 12a.).
- After 20 000 s, there is smaller HF depletion comparing to the case without protuberance (figure 12b.).

2.4.3. RE_{ILC} shape

As shown in figures 13 protuberant cathode is also a benefit for RE_{ILC} shape, especially to attenuate the dissymmetric electropolishing of the cell. The HF concentration is studied at the beginning of the electropolishing (1000 s) and at the end (20 000 s).



Figures 13a. 13b.: Influence of the protuberance on the RE_{ILC} mono cell cavity
a:1 000 s, b: 20 000 s

The result is the one expected:

- With the short protuberance, there is only a small dissymmetric electropolishing which is totally softened after 1 000 s.
- With the longest one, there is a symmetric electropolishing.

A protuberance on the cathode is the best one for TESLA and RE_{ILC} shapes:

- It forces the acid to sweep the bottom of the cell, and so to renew it at this place.
- There is quasi no dissymmetric electropolishing in the cell.

Conclusion

A basis work has been done on modelling of single-cell cavity electropolishing with different shapes (RE_{ILC}, LL_{ILC} and TESLA):

- The required equations are convection diffusion ones for the species transport coupled to Navier-Stockes equation for the flow of the acid. Kinetic equations are used to model the niobium consumption by HF.
- First step was to model Saclay experimental set-up with an acid inflow by a hole in the middle of the cathode. It has been concluded that for each shape there is a dead end in one beam tube, and there is no acid renewal in the bottom of the cell.
- Acid flowing along the main revolution axis of the cavity improves electropolishing of beam tubes however it is not efficient. Furthermore, it induces a dissymmetric electropolishing of the cell, especially for RE_{ILC} shape.
- Nevertheless a protuberant cathode has a very good effect on cell electropolishing for each shape and dissymmetric electropolishing for RE_{ILC} disappears.

Same work as shown in the present report should be done for the half re-entrant shape: this optional profile combines RF parameters of the low loss and re-entrant cavities [6]. However, the next step will be modelling electropolishing for nine-cell “ICHIRO” cavities in collaboration with KEK in order to understand if poor RF performances are related to electropolishing [7]. Then, it should be interesting to confront these results to vertical nine-cell re-entrant cavities of Cornell to evaluate the gravity influence on electropolishing [8]. Future improvements should be completed by adding temperature effect, cavity spinning and 2D axi-symmetric modelling.

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