

Copper Electrochemical Polishing Optimisation

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Abstract

A new electrochemical polishing (EP) facility will be built at CERN to process copper radiofrequency structures and this in the framework of the Future Circular Collider study. This polishing installation will be designed and assembled so that the necessary working conditions to achieve the ultimate limits of this technique are ensured. The main working variables like potential, current density, electrolyte fluid dynamics were thoroughly assessed to guarantee, not only, that polishing conditions were achieved inside the copper structure, but also that they were as even as possible. In this paper, it is described how copper electrochemical polishing optimisation was addressed starting with laboratorial data and their use to model the process with the help of COMSOL®. A set of simulations, performed with COMSOL®, to identify the best polishing parameters are presented as well and where the final result is an optimised cathode geometry.

1. Introduction

The Future Circular Collider (FCC) study develops options for potential high-energy frontier circular colliders at CERN for the post-LHC era. The main emphasis of the conceptual design study is a hadron collider with a centre-of-mass energy of the order of 100 TeV in a new 80-100 km circumference tunnel for the study of particle physics at the highest energies; the conceptual design report will include cost and energy optimisation, industrialisation aspects and provide implementation scenarios, including schedule and spending profiles.

In order to help fulfilling these objectives on the foreseen superconducting radio frequency (SRF) accelerating cavities, a new electrochemical polishing installation will be assembled at CERN capable of processing a wide range of accelerating RF structures, i.e.: 1.3 GHz, 800 MHz mono and 2-cell, 400 MHz elliptical cavities and Wide Open Waveguide Crab Cavity (WOWCC), see figure 1.

The 1.3 GHz structure will be used as R&D support before transferring acquired knowledge to the bigger structures, mainly the 400 MHz and WOWCC cavities, which are the foreseen RF structures for the FCC.

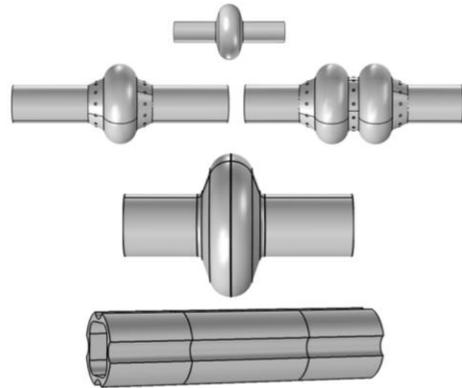


Figure 1. SRF cavities to be processed, from top to bottom: 1.3 GHz, 800 MHz mono and 2-cell, 400 MHz and WOWCC.

The new EP facility will be designed to process RF structures both in vertical and horizontal position; the latter configuration will not be covered in the present article. In the electrochemical polishing, the cavity surface is polished due to an anodic reaction that dissolves the metal burrs and spikes and takes place under appropriate electrolyte and working parameters such as voltage, current density, temperature and flow dynamics.

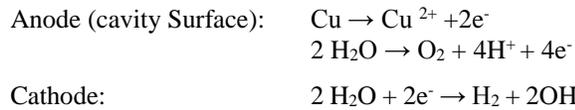
The present work used COMSOL Multiphysics® to optimise the range of working parameters and services (DC voltage and current, cooling power, coolant flow rate) and additionally define the cathode shape as already pinpointed, to ensure to reach the ultimate limits of this technique. The work presented here is focussed only on the 400 MHz structure as it is represents the largest structure to be processed and therefore it will impose by itself the most demanding requirements; all the remaining components would fit within the resulting specifications for the necessary services and facility as a whole.

The modules used in COMSOL were the secondary current distribution and laminar flow in single phase flow. With the first one, the objective was to identify the optimum electrochemical working parameters and cathode geometry that would ensure a minimum power input for the electrochemical process and an as even as possible current density across the RF structure. The interest of using this module is that the current density is proportional to the rate at which the material is removed during the EP process. The laminar flow module was applied to evaluate and optimise the impact of the flow dynamics on the EP reaction rate uniformity.

2. Methodology of the Performed Simulations

2.1. Electrochemical Simulations

The current density distribution in the 400 MHz cavity was modelled with COMSOL Multiphysics® using the secondary current distribution (SCD) physics module. The SCD interface accounts for the polishing bath resistance and for the effect of electrode kinetics, neglecting the influence of concentration variations in the electrolyte. The study was done in stationary mode, as the working conditions during the all process can be assumed to remain constant. The electron transfer reactions that take place in this process are the following:



On the electrode surfaces, the cathode and anode potentials define the boundary conditions. The cathode was defined as reference electrode, setting the parameter *external electric potential* to 0 V. Then, the cell voltage was used as boundary condition for the anode. The EP process is characterised by the polarisation curve ‘current density vs voltage’ of the electrolytic bath. The electrode kinetics were defined on the electrodes surfaces using polarisation curves previously imported. These required inputs for the electrochemical simulation plus conductivity, were obtained from a laboratory set-up using a rotating disc electrode (RDE) immersed in the EP bath [1]. A RDE is a rotating rod where the active surface is the bottom surface, as shown in figure 2.

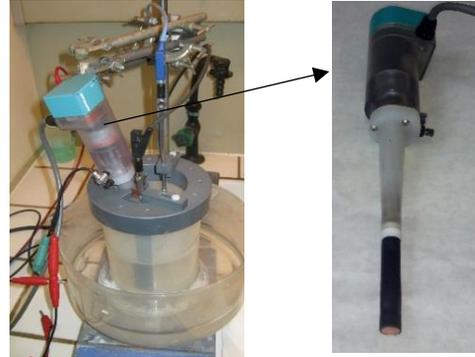


Figure 2. Laboratorial set-up.

The corresponding polarisation curve is shown in figure 3.

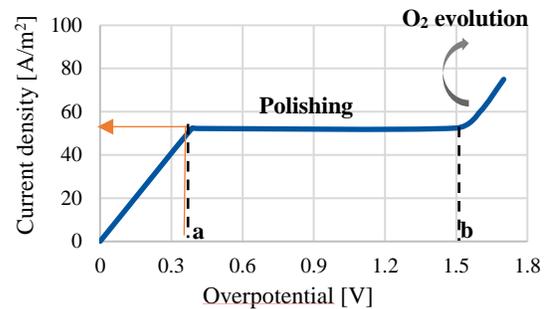


Figure 3. Anodic polarisation curve of the Cu bath measured at 15 degrees and 100 rpm in a laboratorial set-up with the help of a rotating disc electrode [1].

The optimum polishing conditions are obtained in the region between points ‘a’ and ‘b’, also known as polishing plateau. Below point ‘a’ etching is observed but no polishing; above point ‘b’ molecular oxygen starts to evolve, which disturbs the polishing [2]. Thus, it is important to define the minimum working potential that ensures being at the value of current density that corresponds to this region.

In figure 4, it is shown the 3D model used representing a 400 MHz cavity shape and the cathode geometry.

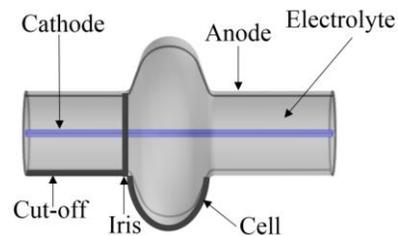


Figure 4. Drawing of the model used and terminology of the different parts of a cavity.

As a starting point for the simulation, the cathode geometry used was a simple straight rod. Then, the output of the simulation, as illustrated in figure 5, provided information on how the current was distributed in the cavity. The optimisation work consisted on defining a cathode geometry that would ensure EP across the whole structure with an even as possible current density distribution with a minimum power input. A parametric sweep was used to identify the minimum working potential.

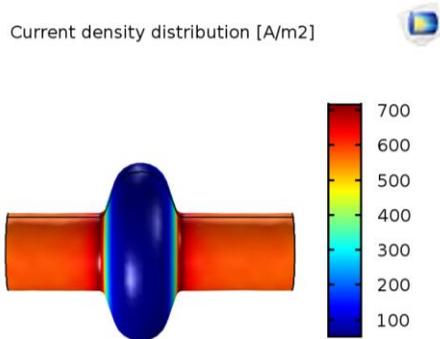


Figure 5. Current density distribution across the cavity surface [A/m^2], output of the electrochemical simulation.

2.2. Fluid Dynamics Simulations

The simulation of the electrolyte velocity distribution in the 400 MHz cavity was done using the Laminar Flow physics combined with the Wall Distance interface. The approach made was to model the electrolyte flow with the cavity in vertical position following previous set-up used at CERN. From the facility point of view, the vertical EP is simpler than the horizontal set-up, as no rotating interfaces are required; the same for the modelling and simulation, as it would imply the use of two phase flow interface and adding the rotation of the cavity. In figure 6, it is presented the 3D model used.

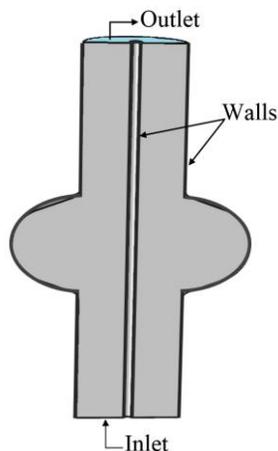


Figure 6. Model geometry

The laminar flow was solved using the Navier-Stokes equations. The model consists of a single domain which represents the electrolyte. The flow inlet was defined on the bottom area of the cavity with the boundary condition of laminar inflow. The outlet, on the top area of the cavity, was set with the pressure boundary condition. The effect of gravity was included with the volume force interface. The cavity wall was defined by a no slip boundary condition representing the fact that in the vertical position, the cavity wall does not move.

In the wall distance interface, the wall boundary condition was added to select the boundaries of the cavity wall, from which we wanted to calculate the distance, and then extract the fluid dynamic parameters. In figure 7, it is presented the information obtained from the simulation.

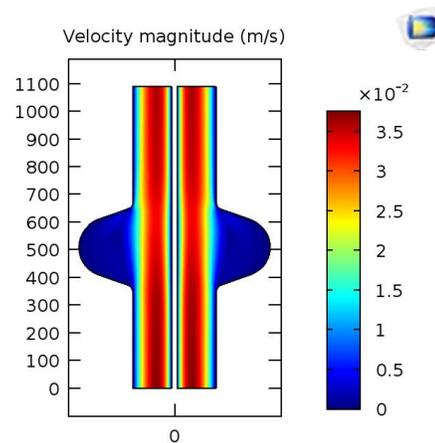


Figure 7. Electrolyte velocity distribution inside cavity.

This model was used to evaluate the electrolyte velocity distribution in the 400 MHz cavity with different cathode geometries. Starting with the geometry of the cathode, which was optimised on the basis of the electrochemical criteria, a new version was made in order to try to improve the homogeneity of the electrolyte velocity.

2.3. Fluid dynamics impact on the etching rate

A 2D model of the laboratory set-up using the RDE was created in COMSOL to get a parameter linked to the fluid dynamics in the active area of the RDE under the different conditions of rotation. In figure 8, it is shown a scheme of the model created.

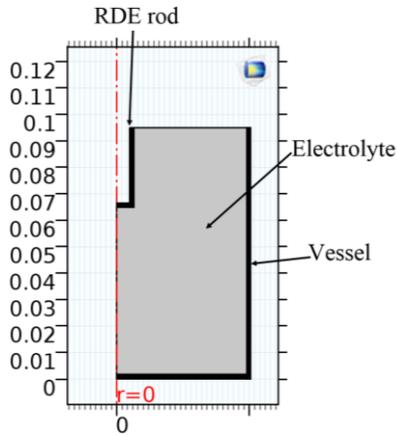


Figure 8. Scheme of the 2D model used in COMSOL.

The Laminar Flow interface was used to model this set-up. The axial symmetry boundary condition was used to define the symmetry plane of the model. The boundaries corresponding to the RDE were defined with the sliding wall boundary condition that allowed the definition of the speed. At the boundaries representing the vessel, a no slip condition was defined. The top boundary was defined with the symmetry boundary condition, and the relative pressure was set to 0 in a point in the top right corner. The values of current density from the polishing plateau extracted from laboratory setup at different rotating speeds were coupled with the shear rate values from the model and the outcome is the correlation shown in figure 9. The shear rate, which represents the gradient of velocity at the vicinity of a reference layer, was considered a good variable to work with as it can be easily assessed with the software and is directly defined on a boundary (cavity wall).

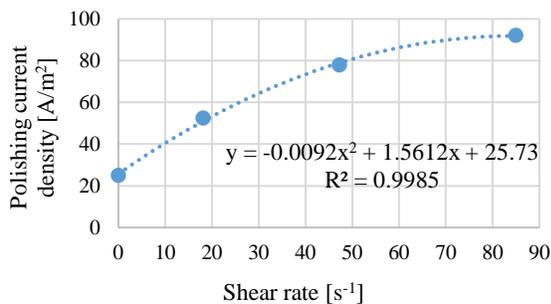


Figure 9. Correlation between polishing current density and shear rate.

Afterwards, the defined correlation was imported in the model of the 400 MHz cavity with the different cathode geometries to assess the impact of the fluid dynamics in the etching rate and this for each configuration.

3. Simulation Results

3.1. Electrochemical Simulations

Two cathode geometries that maximise an even current density distribution across the cavity surface were selected based on the output of the electrochemical simulations. In figure 10, it is presented the starting cathode geometry (1) and the resulting two optimised cathodes. The purple colour indicates the active parts of the cathodes.

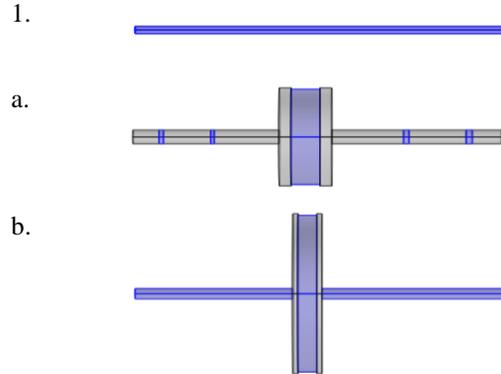


Figure 10. Cathode geometries used in the SCD study.

The geometry of cathode ‘a’ was defined taking into account the constraint defined by the cut-off diameter, which defines a upper limit for the size to be inserted in the cavity; whereas cathode ‘b’ was defined by neglecting this limitation and so as to evaluate the possible benefit, knowing that this would be a teaser for a more complex cathode assembly. In figure 11, it is presented the profile of the current density distribution through the cavity length in function of the minimum working potential, obtained for each of the three previous cathodes. The black dashed line represents the minimum current density that is necessary to ensure that the cavity is being electropolished.

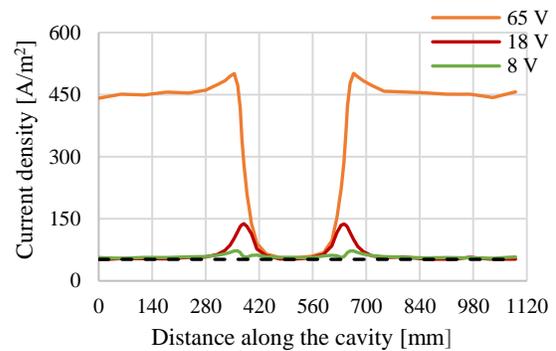


Figure 11. — Starting cathode, — cathode ‘a’, — cathode ‘b’, — min. current density to EP.

As observed in figure 11, with the starting cathode, the whole cavity surface is electropolished applying 65 V. However, the current density is so high in the cut-offs, that the current will be mainly used to produce oxygen rather than removing copper. Based on this result, the cathode shape of cathode ‘a’ was designed to increase the resistance in the cut-offs by reducing locally its active surface. As a result, with this cathode, the current density was globally reduced and applying 18V results in an almost uniform distribution except on the iris of the cavity.

The working parameters (potential and total current) of cathode ‘a’, resulting from this simulation, were used to calculate the power dissipated in the electropolishing process and will be taken into account to dimension the main system equipment and services such as the DC power supply and chiller cooling power.

The results obtained with cathode ‘b’ show an almost uniform current density distribution along the cavity surface, which would mean a lower power input. However, the geometry of such cathode implies the design of a complex mechanical assembly to enable the insertion in the cavity through the cut-off. For time constraints this will not be the first choice for the processing of the elliptical cavities prototypes such as the 400 MHz, but could be considered in a future development.

3.2. Fluid Dynamics Simulation

In figure 12, it is presented the geometry of cathode ‘a’, optimised on the basis of the electrochemical criteria, and of a new cathode, cathode ‘c’, designed with the aim of achieving a more homogeneous electrolyte velocity, without compromising the benefits of ‘a’. Cathode ‘a’, is a massive piece of copper and cathode ‘c’, has the same envelope geometry as ‘a’, but it consists of a solid tube with a central disc that enables the flow of the electrolyte through it.

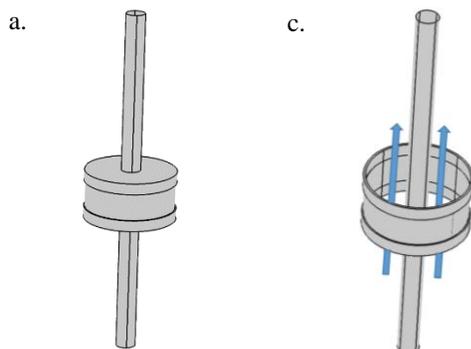


Figure 12. Cathode geometries assessed.

In figure 13, it is illustrated a 2D representation of the electrolyte velocity distribution inside the 400 MHz cavity for the two cathodes using a flow rate of 50 litres per minute. The white arrows represent the path followed by the electrolyte. The blue colour shows the lower range of velocities and the red colour the higher values.

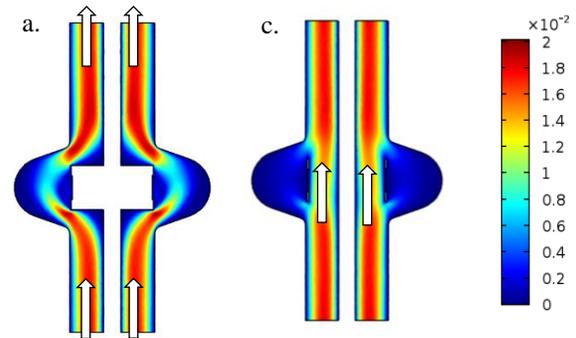


Figure 13. Electrolyte velocity distribution [m/s] with: cathodes ‘a’ and ‘c’.

From these images it is visible that with cathode ‘a’ the electrolyte velocity is higher close to the surface at the iris of the cavity. With cathode ‘c’, as the central disc of the cathode is hollow, it does not present a flow constriction, close to the iris of the cavity, between the cathode and the cavity wall. In figure 14, it is represented the electrolyte velocity profile determined at 20 mm away from the cavity wall through the cavity length.

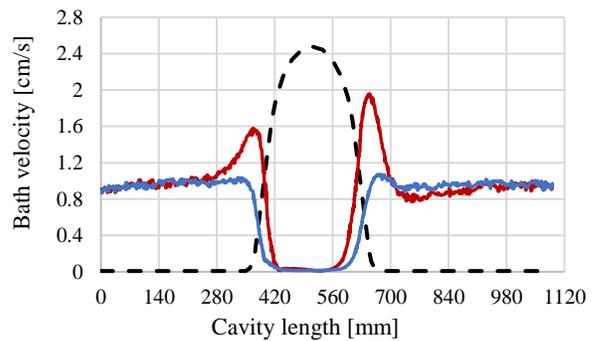


Figure 14. — Cathode ‘a’, — cathode ‘c’, - - cavity shape.

As observed with cathode ‘c’, the electrolyte velocity along the cavity is more homogeneous, without a velocity peak near the iris of the cavity.

3.3. Impact of fluid dynamics in the current density distribution

Using the correlation as defined in §2.3, polishing current density vs shear rate, the impact of the fluid dynamics on the polishing rate for the different cathode geometries was made possible, as the current density is proportional to the rate at which the material is removed. The results are presented in figure 15.

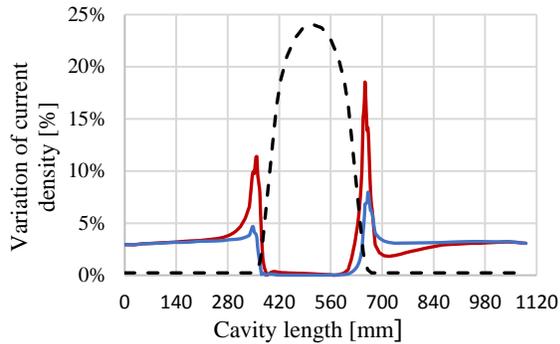


Figure 15. — Cathode 'a', — cathode 'c', - - cavity shape.

As observed in figure 15, the cell of the cavity, where the shear rate is close to 0 (as visible in figure 14 the cell has a low gradient of the velocity), presents no variation of the current density compared to the one set only with the SCD. However, in the cut-offs and iris, the shear rate is higher and thus, current density increases which means that the rate of material removal will be higher as well.

4. Conclusions

The simulations of the current density distribution across the cavity surface were used to define an optimal cathode geometry using first an electrochemical approach. The resulting cathode, cathode 'a', consists of a small tube with a big copper central disc with two isolating rings placed at both sides. This geometry allows a more homogeneous polishing rate, minimizing the regions with high current density where molecular oxygen bubbles are produced. To perform the electropolishing of the 400 MHz cavity using this cathode it is required a minimum working potential of 18 V, and the total current supplied is around 135 A. As consequence, the cooling power required just for the EP process is approximately 2.5 kW.

The fluid dynamics of the process was studied to evaluate the impact of the cathode shape in the distribution of the electrolyte velocity. Cathode 'c', which has a hollow central disc, was found to show the best results, as a more homogeneous electrolyte velocity distribution was achieved, which means a more uniform material removal all through the cavity surface.

The definition of a correlation between the current density and the shear rate allowed to quantify the impact of the fluid dynamics in the EP etching rate obtained in the 400 MHz cavity, observing an increase between 11 and 18% at the iris of the cavity, where the shear rate is higher.

The simulation of the electrolyte flow was made using a vertical set-up. For the future work, it will be important to evaluate the impact of the fluid dynamics on the horizontal electropolishing, where the cavity is half filled with the acid and kept under rotation.

References

1. T.D. Ecclestone, 'Defining the optimum tools and working conditions for copper electrochemical polishing', CERN, Geneva, Switzerland, <https://edms.cern.ch/document/1844296/1>.
2. V. Palmieri, 'Fundamentals of electrochemistry- the electrolytic polishing of metals: Application to copper and Niobium', Proceedings of the 11th Workshop on RF-Superconductivity, September 2013, DESY, Germany.

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