COMSOL CONFERENCE BANGALORE 2013

Electrochemical Modeling of Copper Electrorefining in Lab Scale and Pilot Plant Scale

Jinomol Joy[1], D. Sujish[1], B. Muralidharan[1], G. Padmakumar[1] and K. K. Rajan[1]

[1]Fast Reactor Technology Group, Indira Gandhi Centre for Atomic Research, Kalpakkam, India
sujish@igcar.gov.in

ABSTRACT

Pyrochemical reprocessing is a non-aqueous method for reprocessing spent metallic fuels. Electrorefining is an important step in pyrochemical reprocessing. Ambient temperature electrorefiner (ATER) is a facility to demonstrate the various mechanisms and associated interlocks. Here the electrolysis of copper in acidified copper sulphate electrolyte will be carried out. Electrodeposition of copper was also numerically simulated using the secondary electrodeposition module of COMSOL and validated with the experimental results. The modelling of electrorefining of copper was done for pilot plant scale. The current density for the cathode geometry and the time required for deposition of one batch of copper was also estimated. This paper details the lab scale studies, validation of simulation and further prediction of current densities for pilot plant scale.

INTRODUCTION

Electrorefining is an important step in pyrochemical reprocessing which is a non-aqueous method for reprocessing spent metal fuel. A preliminary demonstration facility has been erected to demonstrate the various mechanisms and associated interlocks. Here the electrolysis of copper in acidified copper sulphate electrolyte will be carried out to demonstrate the electrorefining process. Laboratory scale experiments were conducted in this regard to understand the copper electrorefining process. Electrodeposition of copper was numerically simulated using a commercial multiphysics code COMSOL. The current density in each case was calculated. Current density (CD) plays an important role in electrodeposition. It depends on various factors such as composition, temperature, distance between electrodes, geometry of electrodes etc. As deposition and current density are directly proportional, for getting maximum deposition, we have to enhance CD. The experimental and theoretical current densities for various operating conditions were compared and there was very close agreement in the results. Based on the above results the modelling of electrorefining of copper was done for the as built ATER vessel. The current density for the cathode geometry was calculated and the time required for deposition of one batch of copper was also estimated.

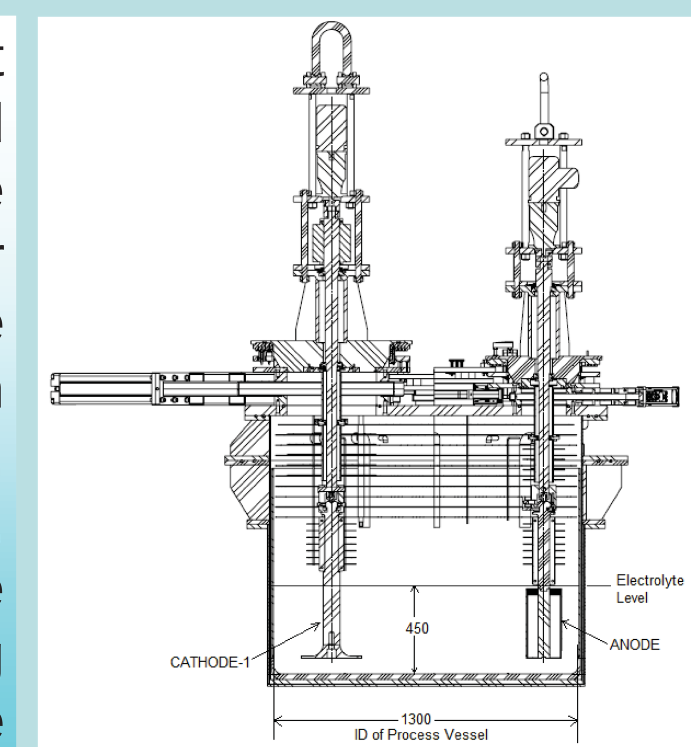

Fig.1: Electrorefiner Vessel

SALIENT FEATURES IN MODELING

MODELING IN COMSOL

Lab scale Modeling

The lab scale experiments for copper electrorefining were carried out. Numerical simulation of copper electrorefining was also carried out using COMSOL. The geometrical model and mesh details are shown in Figure 1. A typical plot of current density with time is shown in Figure 2. The experimental and numerical current densities were found to be matching.

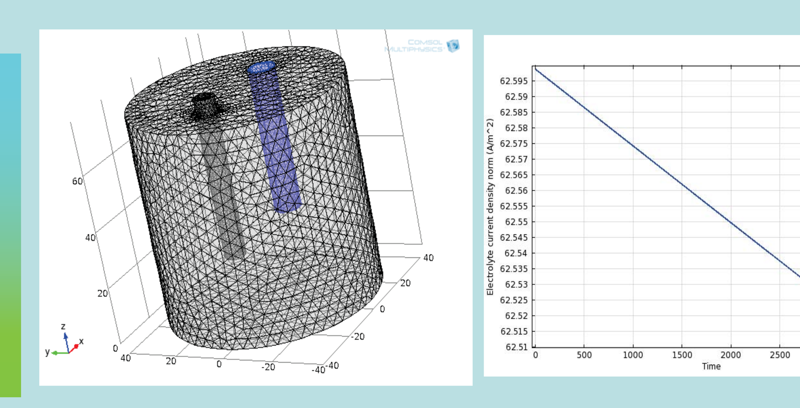

Fig.2: Mesh details of lab scale model.

MATHEMATICAL MODELLING

$i = -\sigma \nabla \phi$ Ohm's law

$\nabla^2 \phi = 0$ Laplace equation

$V = \phi_0$

$\eta = V - \phi_0$ Overpotential

Butler –Volmer equation

$$i = i_0 \left[e^{(\alpha_a F \eta_s)/RT} - e^{(-\alpha_c F \eta_s)/RT} \right]$$

Domain Equations

$\nabla . i_k = Q_k$

$i_s = -\sigma_s \nabla \phi_s$

$i_k = -\sigma_k \nabla \phi_k$ (Electrode)

$i_l = -\sigma_l \nabla \phi_l$ (Electrolyte)

$\eta_m = \phi_s - \phi_l - E_{eq.m}$

Boundary Conditions

The ATER vessel is 1300mm in diameter. The centre to centre distance between the anode and the solid cathode is 700mm. The anode basket is modeled as porous electrode with porosity 0.4. Copper rodlets are dump packed into the anode basket to a height of 103mm in all the four legs. A voltage of 1V is applied externally to the anode. The cathode is a cylindrical rod of 65 mm diameter. Acidified copper sulphate has been modeled as an electrolyte with electrical conductivity 44 S/m. The other inputs to the model include exchange current density which is 16A/m2, Anode transfer coefficient which is 0.5 and cathode transfer coefficient 1.5. Number of participating electrons is 2 and the equilibrium potential at reference temperature of 298.15K is given by the relation 0.34+ (0.0591/2)*log(0.6). Meshing of the geometry is done by extra fine physics controlled meshes.

RESULTS

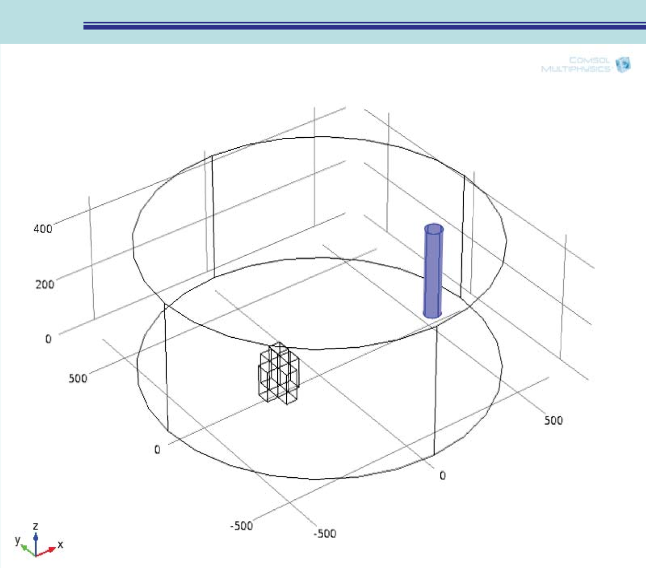

Fig.3: Model of pilot plant scale copper electrorefining.

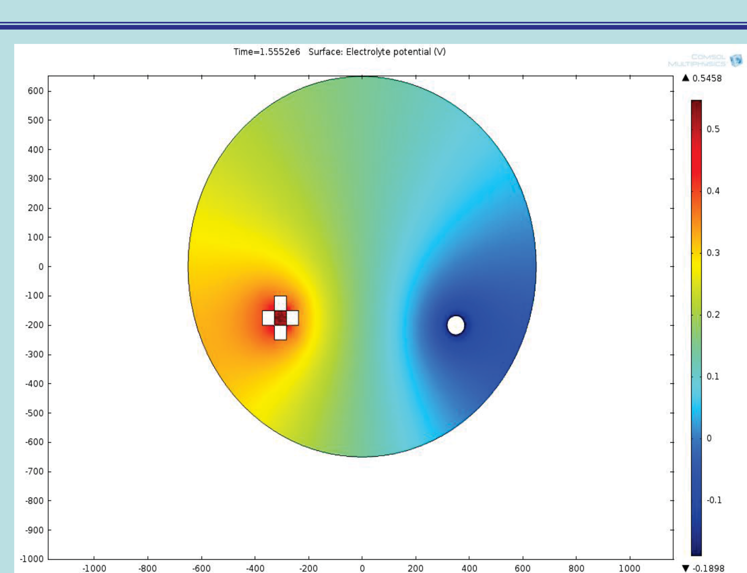

Fig. 4: Electrolyte potential distribution in ATER

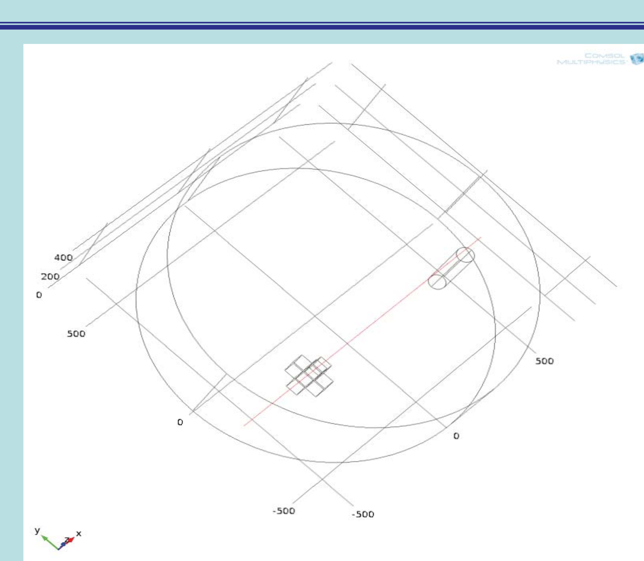

Fig. 5: Cut-line across electrodes and electrolyte

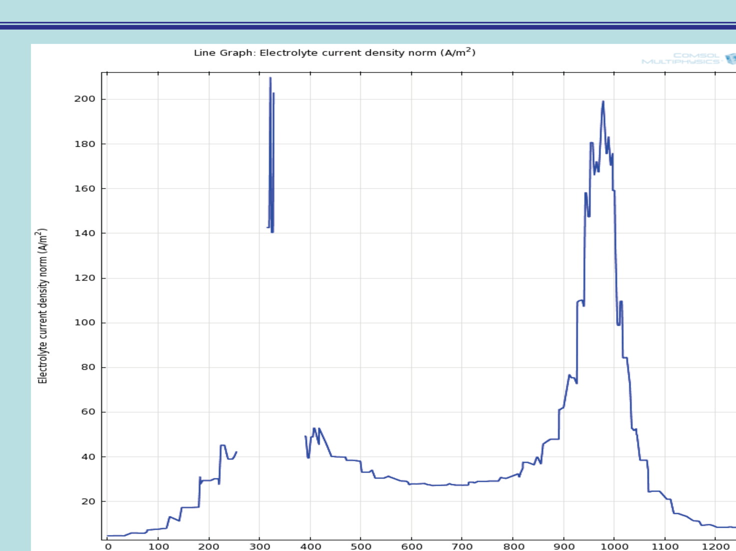

Fig. 6: Current density plot along the cut-line

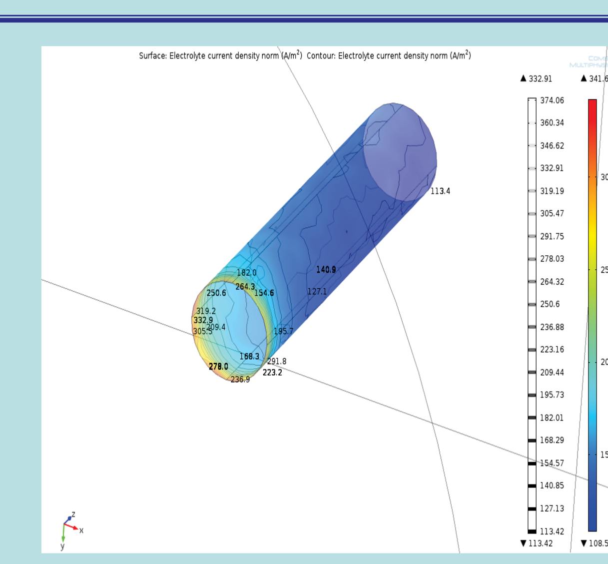

Fig. 7: Contour plot of cathode current density

CONCLUSIONS

The secondary electodeposition module of COMSOL was used to simulate the electrodeposition of copper in laboratory scale. The model was validated by evaluating the experimental and theoretical current densities at various operating conditions. The same module has been used here to evaluate the current density for as built ATER. The current density for this system is around 143.47 A/m2. Accordingly the time taken to deposit of 5 kg of copper on the solid cathode would be 20 days.