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Flow and mass transfer modelling for copper electrowinning: development of instabilities along electrodes

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ABSTRACT

A computational fluid dynamics (CFD) model has been developed to simulate the copper electrowinning (EW) process, and applied to model the flow and mass transfer in the inter-electrode gap for a single plate pair, with geometrical and operation parameters typical of industrial EW operation. The CFD model predicts a recirculation zone in all cases, driven by oxygen bubbles rising along the anode, with the electrolyte deflecting at the upper free surface and recirculating down to the base of the electrode space. The CFD model results showed that laminar natural convection driven by concentration-related density deficiency is dominant along the lower part of the cathode. Strong eddies arise along the cathode where copper depletion becomes large enough to drive buoyancy instabilities: the instability is analogous to the waves formed in natural convection on a vertical heated plate for Prandtl number much greater than 2. Limiting current density can be increased by decreasing the boundary layer thickness which can be achieved by increasing the velocity past the cathode, or by triggering flow instabilities such as the buoyancy-generated ones described above or more conventional shear-driven instabilities. Higher up the cathode, the natural convection profile becomes completely broken up by the recirculating down-flow. Similar instabilities also form close to the anode due to build-up of oxygen bubbles, the fluctuating velocities associated with the anode instabilities being much higher than those at the cathode.

1. Introduction

Copper electrowinning (EW) is the process of depositing solid copper onto a cathode from a leach solution by passing an electric current through the electrolyte. Industrial copper EW is generally carried out in large rectangular cells, in which numerous alternating cathode and anode plates are stacked vertically: current passing between the electrodes causes copper ions to deposit at the cathode, whilst oxygen bubbles are generated on the anode. It is well known that the rising oxygen bubbles cause a large recirculation zone to develop in the space between the electrodes and that this recirculation has a strong effect on mass transfer to the cathode, due to the mixing generated by the recirculation (Graydon and Kirk, 2001). A computational fluid dynamics (CFD) model of the bubble generated recirculation can allow insights into the hydrodynamic behaviour of electrolyte in the spaces between plates, and can in this way assist in understanding the details of copper mass transfer to different parts of the cathode. In particular, a CFD model can elucidate the factors affecting the transport rate of copper to the cathode surface, which is a major rate limiting factor in plating of copper on the cathode.

There are only a limited number of published studies of the hydrodynamics of the electrolyte in electrowinning cells. Ziegler (1984) and Ziegler and Evans (1986) collected limited data of the vertical velocity profile in an industrial scale system (94 cm × 12.5 cm), and compared the velocity profiles with a simple fluid dynamics model with some success. They found that a large scale recirculation is driven by oxygen bubbles rising at the anode. Turbulence intensity was predicted based on applying a two-equation turbulence model to this time-averaged flow: the region very near the anode is thus predicted (and measured) to have very high levels of turbulence, with turbulence intensity of order 50–100% being seen in both measurements and predictions. High levels of turbulence were also predicted to occur at the electrolyte surface where the upward stream is turned towards the cathode and at the top of the cathode itself. The model was also applied to an experimental arrangement for which mass transfer measurements had been made at the cathode (Ettel et al., 1974): mass transfer coefficient predicted by the CFD turbulence model agreed with the measurements at the bottom of the cathode (approximately 1.3×10^{-6} ms^{-1}) and were similar at the top of the cathode ($2.2–2.6 \times 10^{-6}$ ms^{-1}), but the detailed variation with height was not well reproduced.

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The papers of Filzwieser et al. (1999) and Filzwieser (2000) describe an extensive experimental program of detailed measurements of the velocity profiles in an EW cell, as required for reliable CFD validation. This experimental data has been used by Leahy and Schwarz (2010) to validate a detailed CFD model. Filzwieser et al. (1999), Filzwieser (2000), and Filzwieser et al. (2002) discuss some aspects of the inter-electrode hydrodynamics, including the basic recirculation zone that develops in the inter-electrode gap, and the variation in the local copper concentration close to the cathode, as a function of height along the cathode. The authors identify vertically rising flow in the lower part of the cell very close to the cathode, which they interpret as being natural convection driven by the buoyancy of electrolyte depleted of copper, the dominant flow mechanism in electrorefining (Leahy and Schwarz, 2007, 2011). (This flow was also seen by Ziegler and Evans (1986), but not mapped in detail.) A qualitative description is also given of the dynamic behaviour of the flow between the electrodes — the complexity of the fluctuating flow suggests that mass transfer will not be fully understood with a time-averaged approach. In this paper we analyse the flow dynamics in more detail, especially as it relates to mixing and transport, using geometrical and operational parameters typical of industrial cells.

The paper has the following structure: a description of the CFD model is given followed by the detailed description of the results, and conclusions.

2. Electrowinning CFD model

2.1. Model structure and equations

The CFD electrowinning model is two dimensional (2D) in the Y-Z plane, and is applied to a cross section of the cell, as shown in Fig. 1 with the assumption that the flow is uniform in the third (horizontal) dimension (X direction), parallel to the electrodes. The rectangular geometry of the gap region allows the use of an orthogonal mesh, as illustrated in Fig. 1. The typical number of cells in the Y and Z directions is 100 and 200 respectively, with a geometric progression of cell widths in the Y direction ensuring that the cells near the electrodes are of order 20 μm wide. The height of cells is also refined in the region near the liquid surface to approximately 1 mm. The high aspect ratio of the gap geometry causes high cell aspect ratio, but the orthogonality of the mesh maintains good discretisation accuracy. Compute times were typically less than a day running in serial mode. Three dimensional simulations required several weeks.

A two-phase Eulerian–Eulerian ("two-fluid") gas liquid CFD model is employed in ANSYS CFX (2007), with the liquid phase (electrolyte) treated as the continuous phase, and the oxygen bubbles as the dispersed phase. The technique models the dispersed phase as a quasi-continuum, and has been successfully applied to bubble plumes and the recirculation driven by the plumes (Schwarz, 1996). We assume mono-sized bubble distribution of diameter 150 micron. This estimate is based on reports that the bubbles generated are fine with a distribution from 50 micron to 200 micron (Filzwieser, 2000; Ziegler and Evans, 1986) — the predicted flow in the rising bubble plume is insensitive to the exact bubble size provided the terminal (slip) velocity is small relative to the plume velocity.

A mass balance equation is solved, which ensures conservation of mass of each phase. This is known as the equation of continuity, and for each phase is given by

$$\frac{\partial(\alpha_i\rho_i)}{\partial t} + \nabla\cdot(\alpha_i\rho_i\mathbf{v}_i) = S_i \tag{1}$$

where for phase i ($i=1$ is liquid and $i=2$ is gas), S_i is the mass source/sink term (for example at the anode and free surface where gas enters and leaves), ρ_i is the phase density, and $\mathbf{v}_i$ is the velocity. A momentum equation known as the Navier-Stokes equations is solved for each phase, which balances the forces present in the two-phase flow. The Navier-Stokes equation in vector form is given by

$$\frac{\partial(\alpha_i\rho_i\mathbf{v}_i)}{\partial t} + \nabla\cdot(\alpha_i\rho_i\mathbf{v}_i\otimes\mathbf{v}_i) = -\alpha_i\nabla p' + \nabla\cdot\left[\alpha_i\left(\mu_i+\mu_{\mathrm{T},i}\right)\left(\nabla\mathbf{v}_i+(\nabla\mathbf{v}_i)^T\right)\right] + \mathbf{M}_i + S_i\mathbf{v}_i \tag{2}$$

where p' is the (modified) pressure, and $\mathbf{M}_i$ is the sum of the body forces, described below. The laminar viscosity is denoted μ_i ($\mathrm{kg\cdot m^{-1}\,s^{-1}}$), and $\mu_{\mathrm{T},i}$ ($\mathrm{kg\cdot m^{-1}\,s^{-1}}$) is the turbulent viscosity, described below. The transient terms in Eqs. (1) and (2) are not included when the model is run as a steady state.

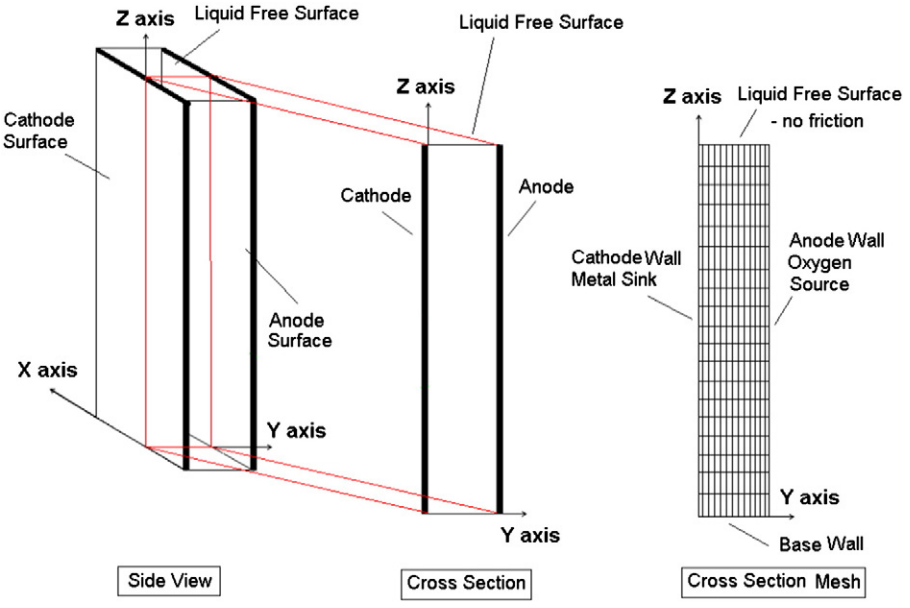

Fig. 1. Schematic diagram of the CFD geometry — side, cross section and cross section mesh views. A much larger number of cells than shown is actually used in the CFD model.