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CFD modelling and simulation of industrial-scale copper electrorefining process

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ABSTRACT

Both electrolyte flow and cupric ion concentration fields have been numerically calculated with the computational fluid dynamics (CFD) model in an industrial-scale copper electrorefining cell. In this CFD model, both of the natural convection induced by the electrochemical reactions and the forced convection caused by the electrolyte circulation are taken into account. In order to examine how the difference in the ways of the electrolyte circulation affects the electrolyte flow pattern associated with the concentration field, three different ways of the electrolyte circulation, "bottom inlet to top outlet", "top inlet to bottom outlet", and "side inlet to top outlet", have been modeled.

Calculation results have revealed that both "bottom inlet to top outlet" and "top inlet to bottom outlet" circulations have characteristic flow patterns that are locally formed near the inlet. On the other hand, "side inlet to top outlet" circulation forms the inherent two vortex tubes in the space below the electrodes. It is also found that the electrolyte flow patterns in the inter-electrode spaces are formed mainly by the natural convection, and they are insensitive to kinds of the electrolyte circulation way.

1. Introduction

The electrorefining process has been widely adopted to produce high purity copper. The current industrial copper electrorefining has been operated in several hundreds of cells where dozens of the anode and the cathode are aligned alternately in an electrolyte solution. It is considered that the nodular growth on the electrolytic copper cathode surfaces is caused by the adhesion of the anode slime particles and the inadequate supply of additives such as glue, thiourea, and chloride ion, to the cathodes. It is also well-known that both glue and thiourea are not only consumed by the electrochemical deposition of copper on the cathode surface, but also degraded due to high electrolyte temperature and strong sulfuric acid. In each tankhouse, the electrolyte is circulated in each unique way to supply optimally additives to the cathodes. The way of the electrolyte circulation may affect the electrolyte flow pattern in a cell, which can contribute to both the anode slime particle behavior and the additive supply to the cathodes. It is thus required to understand how the difference in the circulation ways affects the electrolyte flow pattern.

During copper electrorefining process, the natural convection develops along the vertical electrode surfaces due to the electrochemical deposition and dissolution of copper. Since pioneering analytical work by Wagner (1949) and optical work by Ibl and Muller (1958), a lot of experimental and theoretical studies have been accumulated on the mass transfer associated with the natural convection during copper electrolysis in an unstirred $CuSO_4$–H_2SO_4 aqueous electrolyte solution. With the recent improvement of computational capability, some numerical studies have been carried out. However, most of studies have focused on the small-scale laboratory systems and few studies have investigated into copper electrolysis with the electrolyte circulation system.

The present study numerically calculates both the electrolyte flow and cupric ion concentration fields in an industrial-scale copper electrorefining cell with the computational fluid dynamics (CFD) model. Both of the natural convection induced by the electrochemical reactions and the forced convection caused by the electrolyte circulation are considered in this model. In the present study, three different ways of the electrolyte circulation are modeled in order to examine how the difference in the circulation ways affects the electrolyte flow pattern as well as the concentration field of cupric ion in a whole cell. Calculation models are developed according to the tankhouse data at Naoshima Smelter and Refinery, Mitsubishi Materials Corporation. The calculated concentration profiles of cupric ion are compared with the measured values at a Naoshima electrorefining cell.

2. CFD model

Fig. 1 shows a schematic description of the electrolytic cell employed in the present study. The dimensions of this cell are

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5.4 m long and 1.2 m wide. The bottom of this cell is slanted from 1.2 to 1.35 m in depth. In this cell, there exist 54 anodes with 0.98 m in height and 0.96 m in width, and 53 cathodes with 1.0 m in height and width. Anode spacing between centers is fixed to 97 mm. It is assumed that both the anode and the cathode are geometrically flat plates hanging vertically and they do not have any distortion. As seen in Fig. 1, there exist two spaces with 100 mm in width between the side wall of the cell and the side edge of the cathode, which are called as "side regions" in this paper.

During the electrorefining process, the anode becomes thinner with time due to the electrochemical dissolution of copper, whereas the cathode becomes thicker with time due to the electrochemical deposition of copper. In the present study, four computational models are developed in order to examine the effect of the thickness changes of both the anode and the cathode. Fig. 2 shows a schematic description of four models about the electrode thicknesses and the inter-electrode spacing L in this study. It is assumed that copper cathodes are produced twice during one anode life. The process to produce the first electrolytic copper cathode is called as "1st crop operation", and the latter process as "2nd crop operation". The thicknesses of both the anode and the cathode in calculation models are set to correspond to "1st crop initial state", "1st crop final state", "2nd crop initial state", and "2nd crop final state".

In the present study, following three different ways of the electrolyte circulation are modeled, as shown in Fig. 3, to examine how the difference in the circulation ways affects the electrolyte flow pattern as well as the concentration field.

(a) "Bottom inlet to top outlet".
(b) "Top inlet to bottom outlet".
(c) "Side inlet to top outlet".

"Bottom inlet to top outlet" circulation has been most conventionally adopted for copper electrorefining cells worldwide, although there may be some differences in detail. The electrolyte enters the cell through the bottom inlet box located at one end, and it then overflows through the top outlet box located at the opposite end. On the other hand, in the case of "top inlet to bottom outlet" circulation, the electrolyte enters the cell through the top inlet box located at one end, and it is then drained through the bottom outlet box located at the opposite end. The electrolyte circulation way of "side inlet to top outlet", which is shown in Fig. 4, has been adopted at Mitsubishi Materials Corporation's Naoshima copper tankhouse. Contrary to the conventional electrolyte circulation way, the electrolyte is fed to the cell through the inlet holes on two pipes placed at the cell bottom, and it then overflows through the top outlet box. 53 Inlet holes with 10 mm in diameter are opened alternately on two pipes, which are designed to feed the electrolyte toward every cathode.

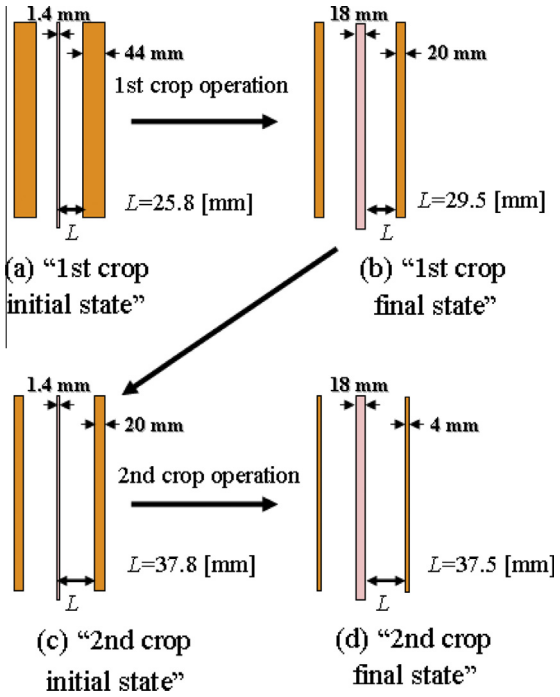

Fig. 2. Schematic description of four models about the electrode thicknesses and the inter-electrode spacing.

In the present study, the CFD model is set up within the ANSYS FLUENT framework. The following continuity equation and steady state Navier–Stokes equations for the incompressible Newtonian fluid are numerically solved.

$$\nabla \cdot (\rho \mathbf{u}) = 0 \tag{1}$$

$$\nabla \cdot (\rho \mathbf{u}\mathbf{u}) = -\nabla p + \eta \nabla^2 \mathbf{u} + \rho \mathbf{g} \tag{2}$$

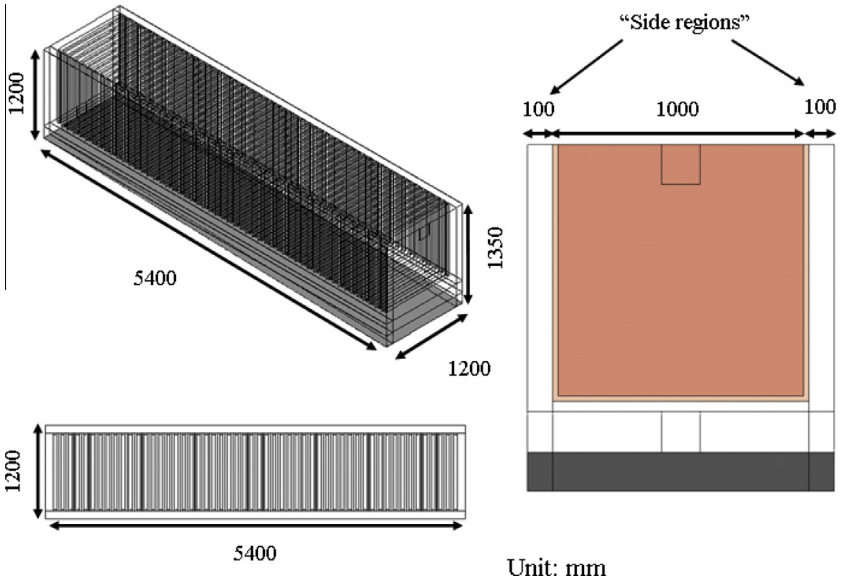

Fig. 1. Schematic diagram of the electrolytic cell.