Electrochimica Acta 164 (2015) 218–226

Contents lists available at ScienceDirect

Electrochimica Acta

journal homepage: www.elsevier.com/locate/electacta

Comparison between Numerical Simulations and Experimental Results on Copper Deposition in Rotating Cylinder Hull Cell

Jaeyeong Park [a], Sungyeol Choi [b,*], Robert Hoover [c], Kwang-Rag Kim [d], Sungjune Sohn [a], Yong-Hoon Shin [a], Supathorn Phongikaroon [e], Michael Simpson [f], Il Soon Hwang [a]

[a] *Department of Nuclear Engineering, Seoul National University, 1 Gwanak-ro, Gwanak-gu, Seoul, 151-742, Republic of Korea*
[b] *School of Mechanical and Nuclear Engineering, Ulsan National Institute of Science and Technology, UNIST-gil 50, UNIST, Ulsan, 689-798, Republic of Korea*
[c] *Department of Chemical and Materials Engineering, University of Idaho-Idaho Falls, Center for Advanced Energy Studies, 995 University Blvd, Idaho Falls, ID 83401, United States*
[d] *Korea Atomic Energy Research Institute, 1045 Daedeok-daero, Yuseong-gu, Daejeon, 305-353, Republic of Korea*
[e] *Department of Mechanical and Nuclear Engineering, Virginia Commonwealth University, 401 West Main Street, Richmond, VA, 23284, United States*
[f] *Department of Metallurgical Engineering, University of Utah, 201 Presidents Cir, Salt Lake City, Utah, 84112, United States*

ARTICLE INFO

Article history:
Received 25 September 2014
Received in revised form 16 February 2015
Accepted 18 February 2015
Available online 20 February 2015

Keywords:
rotating cylindrical Hull cell
electrorefining
overpotential distribution
current density distribution
copper electrodeposition

ABSTRACT

2D and 3D numerical models for electrorefining used in pyroprocessing have been developed by Seoul National University with the Korea Atomic Energy Research Institute and University of Idaho with the Idaho National Laboratory, respectively. To validate these models, numerical simulations are conducted on a rotating cylindrical Hull cell for copper deposition in a sulfuric acid solution. The primary current density distribution along the cathode is compared to an empirical equation of Madore. The 2D and 3D modeling results of the tertiary current density distribution along the cathode were compared. The numerical modeling results of the 2D and 3D models match each other well. In addition, the modeling results of the 3D model on the tertiary current density distributions according to the applied current densities are compared to the experimentally measured distributions. There are some discrepancies between the modeling results and experimental data. The discrepancies could be mainly explained by the hydrodynamic effect of Luggin probes used for measuring the overpotential distribution. At low Reynolds number, Luggin probes could act as a static mixer improving mass transfer near working electrode. In contrast, at high Reynolds number, Luggin probes could act as a flow obstacle dissipating flow kinetic energy.

1. Introduction

Pyroprocessing has received attention for its potential application to used nuclear fuel (UNF) management through the recycling of actinides and the generation of stable waste forms. Its high operation temperature in a strong radiation environment makes it attractive with regards to proliferation resistance [1]. In this technology, electrorefining is a key process for determining the total process time and volume of the generated waste. This unit process recovers uranium (U), which constitutes approximately 96 wt% of UNF from a Pressurized Water Reactor (PWR). Thus, the optimization of electrorefining is a significant issue for the overall process optimization.

Numerical modeling, as an alternative to experimental investigation, can dramatically reduce the cost of electrorefiner (ER) design optimization and the study of the operating parameters. For this reason, a number of numerical models of the uranium ER have been developed, including REFIN [2], PYRO [3], TRAIL [4], GPEC [5], and PRAGAMAN [6]. Each of these numerical models can be beneficial for a prediction of the electrochemical reactions; however, there are still significant discrepancies with the experimental results. These models used the diffusion boundary layer approximation without considering complicated turbulent mass transfer effects. Because of this limitation, these models cannot predict tertiary current distributions in the ER with poor mixing, relatively low concentration, or high applied current density. The local mass transfer effect in turbulent condition is significant to predict current density distributions, but it is difficult to understand because turbulent flow has typical irregularities and a thin diffusion boundary layer [7]. In addition, these existing

---

* Corresponding author. Tel.: +82 52 217 2352; fax: +82 52 217 3009.
E-mail address: chois@unist.ac.kr (S. Choi).

http://dx.doi.org/10.1016/j.electacta.2015.02.160

Table 1
Comparison of numerical simulation studies in the Hull cell.

	West et al., 1992 [22]	Madore et al., 1992a [19]	Madore et al., 1992b [14]	Low et al., 2007 [13]	This paper
Cell systems	Traditional Hull cell	Rotating cylinder Hull (RCH) cell	RCH cell	RCH cell	RCH cell
Mass transport control	Difficult to control	Well controllable	Well controllable	Well controllable	Well controllable
Current density distributions	Primary	Primary	Primary, secondary	Primary, secondary, tertiary	Primary, tertiary
Analytical solutions for current density distribution	A curve fit to an analytical solution	A curve fit to an analytical solution	No	No	No
Potential distributions	No	No	No	Yes	Yes
Experimental comparisons	No (compared with reported empirical formula)	No	Deposit thickness with current density distributions	No	Measured potential distributions with calculated ones
Numerical methods	Analytical method	Boundary element method	Boundary element method	Finite element method	Finite element method
Simulation dimension	2D	2D	2D	2D	2D, 3D

models have a limitation in that it is hard to simulate electrochemical reactions with a specific geometry. To predict the local contamination on the cathode and improve the ER more efficiently, a multi-dimensional numerical analysis reflecting the specific geometries is required.

For this reason, Seoul National University (SNU) with the Korea Atomic Energy Research Institute (KAERI) and University of Idaho (UI) with the Idaho National Laboratory (INL) have developed three dimensional (3D) [7–10] and two dimensional (2D) [11,12] computational electrorefining models, respectively. To validate these two models, benchmark studies using a Rotating Cylinder Hull (RCH) cell with a copper sulfate electrolyte solution were performed. The RCH cell with copper sulfate system was selected, because it has been widely utilized as a standard technique for quantitative electrodeposition studies and its high availability of reliable materials property data [13]. This paper includes comparisons of the 2D and 3D electrorefining models with the RCH cell experimental results conducted at SNU using a copper sulfate solution.

Herein, we provide a description of the geometry and conditions of the RCH cell and the suggested mathematical equations for both the 2D and 3D computational models. The results of the RCH cell experiments conducted at SNU are presented, and the computational results of the two models are compared and validated against the experimental results. Furthermore, discussions on the discrepancies among the two models and the experiments are being delivered in detail.

2. Rotating Cylindrical Hull cell

The RCH cell for electroplating research has been used as an experimental tool for the investigations of single metal, alloy and composite deposition. This cell proposed by Madore et al. [14] provides an analytical solution for primary current distribution

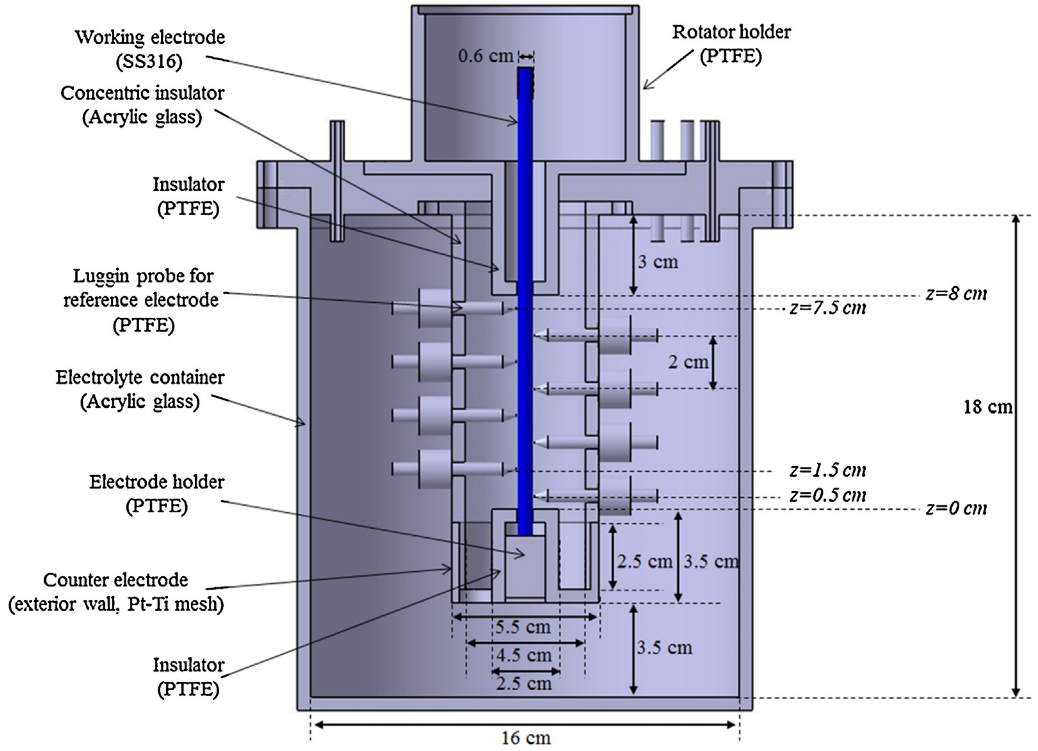

Fig. 1. Sectional view of the rotating cylinder Hull cell design.