Multi-Dimensional Modeling of Electrorefining Process of Used Nuclear Fuel With Liquid Cadmium and Rotating Cylinder Electrodes

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1. Introduction

Electrorefining process is the actual process separating the recyclable U species from the products of electroreduction process [1]. Thus the separation efficiency of electrorefining process determines the efficiency of the whole pyroprocessing.

This study aimed to construct computational electrorefining model. Electrorefining process accompanies electrochemical reaction and mass transport. To reflect these multi-physics in the electrorefining reaction and optimize the computational load, the 3D hydrodynamic model and 2D multi-species model were developed.

The results were validated by the experimental data of lab-scale electrorefining process conducted by Argonne National Laboratory (ANL) at 1992 [2].

2. Method

2.1 Cell configuration

This model brought initial conditions and geometry from the lab-scale ANL experiment. The configuration of electrorefining cell is shown in Fig. 1.

The molten salt region in contact with the cathode and anode, where the region highlighted by the red line in the Fig. 1, was only considered in this model to focus on the reactions over the surface of the electrodes.

2.2 Numerical approach

In this paper, 3D and 2D computational electrorefining models were developed with a CFD software COMSOL 5.1.

The 3D model only considered U single species and calculated velocity distribution and the thickness of diffusion dominant region caused by the rotating cylinder cathode.

The calculated thickness was applied to the 2D model which ignores fluid dynamics and assumes well mixed state of bulk region and only consider mass transport of diffusion in the diffusion dominant region. 2D model involved three species U, Pu, and Nd and focused on the competitive reactions of them.

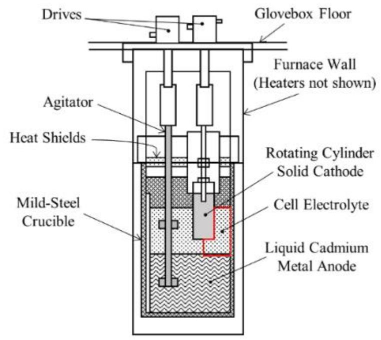

Fig. 1. ANL lab-scale electro-refining cell [2].

2.3 Mathematical equations

The Nernst-Planck equation was calculated to reflect mass transport caused by the convection and diffusion. The electromigration was neglected because of the sufficient supporting electrolyte.

$$N_i = -D_i \nabla C_i + C_i v - \frac{n_i F}{RT} D_i C_i \nabla \phi \qquad (1)$$

The Navier-Stokes equation was applied to the 3D hydrodynamic model to reflect the convection effect of rotating cylinder cathode.

$$\rho\left(\frac{\delta v}{\delta t} + v \cdot \nabla v\right) = -\nabla p + \nabla \cdot \mu(\nabla v + (\nabla v)^T) + f \qquad (2)$$

The modified Butler-Volmer equation was applied to the 2D multi-species model to calculate the competitive reactions of reactants.

$$i_{loc,i} = i_{ex,i} \frac{exp(\frac{\pm\alpha_i n_i F\eta_i}{RT})}{1+\frac{i_{ex,i}}{i_{L,i}}exp(\frac{\pm\alpha_i n_i F\eta_i}{RT})} \quad (3)$$

The limiting current was defined as follow:

$$i_{L,i} = \frac{n_i F D_i C_{bulk,i}}{\delta} \quad (4)$$

3. Results and Discussions

3.1 3D hydrodynamic model

The velocity distribution of U in the molten salt was investigated with the 3D model (Fig. 2). The highest velocity was formed in the vicinity of the rotating cathode. On the other hand, the lowest velocity was formed right below the cathode bottom and the region far from the cathode. Because of the active mass transport by the convection, very thin diffusion dominant layer was formed near the cathode.

The calculated thickness of the diffusion dominant layer was 0.0045 cm and it was applied to the 2D model. However, we expect the actual thickness should be thinner because we only consider the convection effect of the cathode.

3.2 2D multi-species model

The amount of metal deposition on the cathode was investigated with 2D multi-species model (Fig. 3). The early deposition reaction was dominated by the U. However, the time after 50 hours, the Pu started to participate in the reduction reaction and the reduction rate of U started to decrease. Although the Nd also exist in the molten salt, Nd was not involved in the metal deposition on the cathode until the end of the process. It is because Nd is chemically very stable in chloride form [3].

4. Conclusion

In this paper, the hydrodynamic effect of the rotating cylinder electrode was investigated with 3D model and the calculated thickness of diffusion dominant region was applied to the 2D multi-species model. The simulated result of metal deposition on the cathode was well matched to the reference experimental data of ANL.

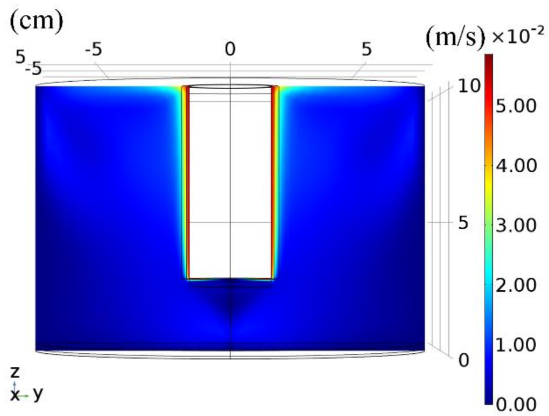

Fig. 2. Velocity (m/s) of U in molten salt at the *yz*-cross section of 3D hydrodynamic model analyzing a rotating cylinder solid electrode.

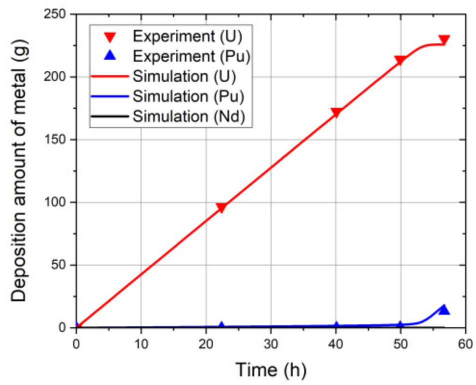

Fig. 3. Amount of metal deposition on the cathode.

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