Development of 2D Kinetic Model for Multi-species Pyrochemical Electrorefining

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1. INTRODUCTION

Pyroprocessing is technology to recover uranium, transuranic elements, and long-lived fission products from spent nuclear fuel. This technology can considerably reduce the volume and the toxicity of radioactive waste [1]. The optimization of new technology requires repeated experiments that require extremely expensive and dangerous works with nuclear materials. As an alternative, we developed a 2D computational kinetic model for multi-species electrorefining.

2. MODEL DESCRIPTION

2.1 Modeling and Experimental Conditions

The laboratory scale electrorefiner in Tomczuk et al., 1992 [2] is used in this model. The geometry of the eletrorefiner is given in Fig. 1. The initial concentration in the molten salt of U is 1.61wt% and that of Pu is 1.82wt%. The run considered here lasted 56.7 h. The applied current is 1.45A, and the operation temperature was 773 K. More information are shown in Table 1.



Fig. 1. ANL Lab-scale Electrorefiner [2].

Table 1. Initial Electrorefining Conditions [2]

Parameters	Values
Temperature (K)	773
U in salt (wt%)	1.61
Pu in salt (wt%)	1.82
U in Cd pool (wt%)	1.01
Pu in Cd pool (wt%)	0.59
Current (A)	1.45
Molten-salt weight (g)	3680
Molten-salt volume (ml)	1808
Cd weight (g)	16500
Cd volume (ml)	2050

2.2 Mathematical Model

In this paper, COMSOL multiphysics code was used to develop electrorefining model. Specifically, secondary electrodeposition module, transport of diluted species module, and boundary ODEs and DAEs module were used. In the secondary electrodeposition module, electrode boundary conditions and kinetics were applied. At the cathode, -1.45A current was applied. Butler-Volmer electrode kinetics were applied at both electrodes.

$$i_{total} = \sum_{j} i_{loc,j} \quad (1)$$

$$i_{loc,j} = i_{0,j} \left(C_R^* e^{\frac{\alpha_a F \eta}{RT}} - C_o^* e^{\frac{\alpha_a F \eta}{RT}} \right) \quad (2)$$

In the transport of diluted species module, mass flux were solved. We only consider diffusion term of mass flux. Also We assumed that the concentration gradient only exist in diffusion layer domain which is 0.002cm long distance from electrode surface and assumed well mixing condition in the bulk of molten salt and Cd pool so that the concentration distribution is uniform throughout the bulk domain.

$$N_i = -D_i \nabla C_i \quad (4)$$

In the Boundary ODEs and DAEs module, the changes of bulk concentration of molten salt and Cd pool were solved.

3. RESULTS & DISCUSSION



Fig. 2. Bulk Concentration in Molten-salt (Circle: U, Square: Pu in all figures).



Fig. 3. Deposited Amount on the Solid Cathode.

Fig. 2 and Fig. 3 show us the bulk concentration change in molten salt and deposited amount on solid cathode over time. At the initial stage, by the difference of equilibrium potential at cathode, U dominantly deposit on the cathode and Pu does not. Thus U bulk concentration of molten salt keep decreasing and that of Pu keep increasing until the ratio of Pu to U reaches enough value and then gradually Pu deposition substitutes U deposition to satisfy the total current. These tendencies can be inferred from the slope changes of bulk concentration changes.



Fig. 4 show the concentration change of U and Pu in Cd pool. Although Pu has more negative standard potential than U, U dissolution rate is faster than that of Pu since the initial concentration of U is about two times that of Pu. In realistic case, after U concentration decrease, the effect of abundant concentration of U to U dissolution rate have to decrease as well and the Pu dissolution rate have to increase. However, this figure doesn't show that tendency. It must be complemented.

4. CONCLUSIONS

In this paper, the deposition and dissolution tendency of electrorefining process were studied. Based on these information, this model will contribute to find the optimum condition for high U throughput with high decontamination factor (>100) without other containments by experiments.

5. REFERENCES

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