Methodologies for Three-Dimensional Computational Modeling on Electrorefiner

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1. Introduction

Pyroprocessing have been actively researched in some major countries as a partitioning technology for treating spent nuclear fuel which has high proliferation resistance based on group separation of actinides and its high radiation and temperature conditions that make it hard to approach to nuclear material [1]. In this technology, electrorefining play the leading part as a step in which most of uranium in spent nuclear fuel can be recovered [2]. In order to recycle recovered uranium for nuclear fuel, it is important to acquire high purity uranium without other elements in this process. Furthermore, high throughput electrorefiner should be designed in order for pyroprocessing to become a feasible solution for spent nuclear fuel problem. These two goals could be more economically and effectively achieved based on three-dimensional design with computational analysis by reducing trial and error in experiment.

However, since three-dimensional numerical analysis on multi-element electrochemical reaction requires expensive computational resources, it is impractical and inefficient to obtain all information on electrorefiner at once. In this paper, the several methods for three dimensional modeling on electrorefining process according to needed information are presented.

2. Methods and Results

2.1 Modeling on Applied potential and overpotential

2.1.1 Single element assumption

Applied potential refers to the sum of anode and cathode overpotential, ohmic drop and cell potential which can be obtained by adding anode and cathode potential represented in Eq. (1). [3]

$$E_{cell} = E_{anode} + E_{cathode} + \eta_a + \eta_c + IR_{Ohmic} \quad (1)$$

Electrorefining for uranium recovery from spent nuclear fuel is a multispecies system including uranium, actinides, fission product and so on. Since rapid changes of species concentration and potential could occur in multi-element system, three-dimensional multispecies computational analysis might be a time consuming and inefficient way. However, electrorefiner simulation could be conducted more efficiently based on single element assumption. Because most of spent nuclear fuel consists of uranium and electrorefiner aims to recover of pure uranium without other elements, a great portion of anodic and cathodic currents is determined by uranium in the case of normal operation of electrorefiner except the initial stage when some elements are dissolved from anode as described in Fig. 1 obtained by one-dimensional electrochemical simulation.

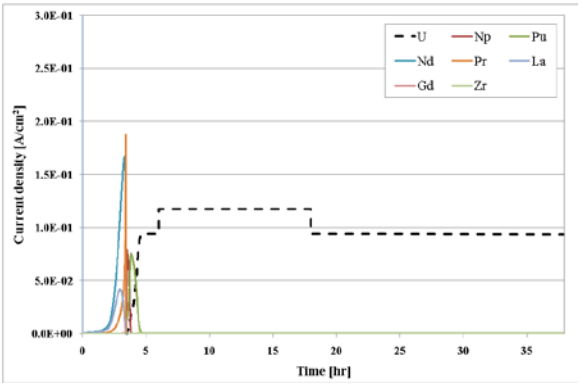

Fig. 1. Anode current density on 38 hours electrorefining which was calculated by one-dimensional electrochemical computation

Therefore, single element assumption considering only uranium could be regarded as a reasonable approach for uranium recovery process modeling. The equation can be simplified as Eq. (2) because anode and cathode potential cancel each other in single element assumption.

$$E_{cell} = \eta_a + \eta_c + IR_{Ohmic} \quad (2)$$

2.1.2 Determination of effective anode area

Since overpotential is related to current density by Butler-Volmer equation and Ohmic drop might be changed over time based on diameter increase of cathode which reduce the physical distance between anode and cathode, determination of detailed geometry configuration such as effective electrode area is really significant to calculate more exact overpotential and ohmic drop in electrorefining modeling. However, it is tough decision because of complicated mechanisms of electrode geometry change just as dendrite growth of cathode and porosity formation of anode. In addition, if fuel claddings are loaded in anode basket with spent nuclear fuel, the complexity and uncertainty of anode geometry may increases because Zr metal layer can be developed on perforated anode basket surface by repetition of dissolution and deposition of Zr at anode [4]. In this case, although mass of loaded spent nuclear fuel converge to zero, Calculated applied potential using the anode profile which comes close to the area of

anode basket area shows better agreement with experiment which is conducted by Idaho National Laboratory than that using the anode profile which moves toward zero as represented in Fig. 2.

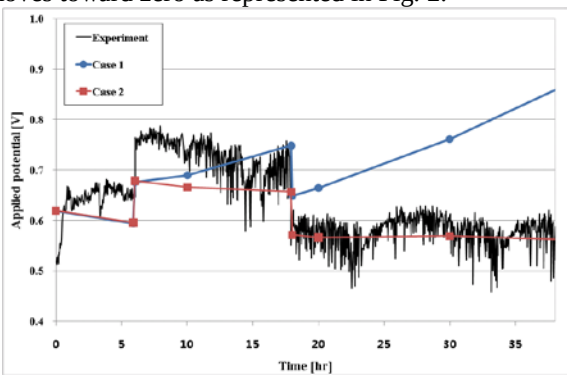

Fig. 2. Comparison between experimental data and calculated applied potentials based on effective anode area profiles which converge to zero (case1) and anode basket area (case2)

2.2 Modeling on TRU contamination on recovered uranium

Designing electrorefiner which has high uranium throughput without TRU decontamination is one of the most important issues in pyroprocessing but it is hard to achieve because of irregularity of mass transfer which result in increase of local overpotential. Therefore, in order to design more efficient and profitable electrorefiner, it is necessary to conduct three-dimensional computational modeling. As mentioned above, because three-dimensional multispecies analysis requires expensive resources and long computation time, analysis on TRU contamination could be indirectly conducted by combining one-dimensional electrochemical calculation and results on three-dimensional single element mass transfer simulation as represented in Fig.3.[5]

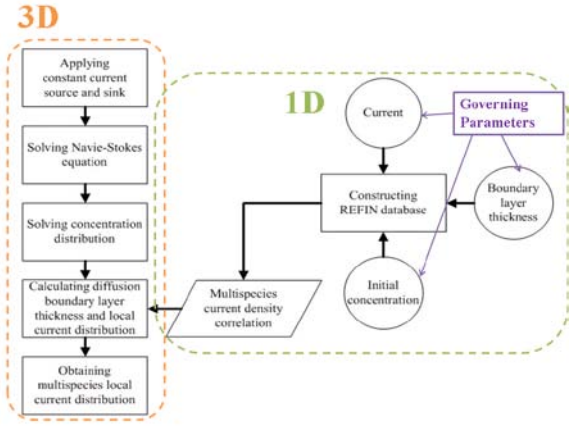

Fig. 3. Overall approaches for calculating TRU decontamination by combining one dimensional electrochemical calculation and three dimensional fluid dynamics simulation

In one-dimensional electrochemical calculation, current density correlation as function of boundary layer thickness, initial concentration actinides and so on can be obtained. Therefore, applying this correlation to three-dimensional fluid dynamics results, degree and location of TRU decontamination could be confirmed [5].

3. Conclusions

Because of the limitation of computational resources, it might be hard to obtain all information at once by three-dimensional multispecies calculation. Therefore, it is advisable to select appropriate method according to need information. Single element assumption could be applied for calculating overpotential and applied potential in normal operation of electrorefiner. Determination of effective anode area is also significant because it directly affects current density on electrode surface. Modeling on TRU contamination could be conducted indirectly but more efficiently and effectively by combining one-dimensional electrochemical calculation and three-dimensional mass transfer simulation.

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