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Original Article

A MODEL STUDY ON MULTISTEP RECOVERY OF ACTINIDES BASED ON THE DIFFERENCE IN DIFFUSION COEFFICIENTS WITHIN LIQUID METAL

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ABSTRACT

This study presents an effective method for additional recovery of residual actinides in liquid electrodes after the electrowinning process of pyroprocessing. The major distinctive feature of this method is a reactor with multiple reaction cells separated by partition walls in order to improve the recovery yield, thereby using the interelement difference in diffusion coefficients within the liquid electrode and controlling the selectivity and purity of element recovery. Through an example of numerical simulation of the diffusion scenarios of individual elements, we verified that the proposed method could effectively separate the actinides (U and Pu) and rare-earth elements contained in liquid cadmium. We performed a five-step consecutive recovery process using a simplified conceptual reaction cell and recovered 58% of the initial amount of actinides (U + Pu) in high purity (\geq 99%).

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1. Introduction

Despite the widespread use of nuclear power to meet the growing demand for energy across the world, the treatment of spent nuclear fuel as a by-product of nuclear power generation remains a great challenge for the nuclear power industry. Pyroprocessing is one of the practical methods for solving this problem. The basic concept of pyroprocessing includes recycling of spent nuclear fuel by processing it from a pressurized water reactor into a raw material for a sodium-cooled fast reactor. This process is relatively simple and cost effective, and in particular, has an additional advantage of meeting the proliferation resistance requirement of the fuel cycle by preventing Pu from being separately recovered. Driven by the benefits of 100-fold increase in the uranium recycling rate, 100-fold decrease in the space requirement of a high-level waste disposal site, and 20-fold decrease in the volume of high-level waste compared to a direct radioactive waste treatment, recycling of spent nuclear fuel using a new method of coupling pyroprocessing to sodium-cooled fast reactors has been the object of intensive research [1].

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Pyroprocessing recovers nuclear fuel materials such as uranium and transuranic elements using the electrochemical reaction in the LiCl or LiCl-KCl molten salt medium, which is very stable at a high temperature. Pyroprocessing can be broken down into several subprocesses: (1) pretreatment, in which spent nuclear fuel assemblies are dismantled and stripped; (2) electrolytic reduction process, which converts spent oxide nuclear fuel into metal; (3) electrorefining, which selectively recovers uranium, (4) electrowinning, which recovers actinides-residual uranium and transuranic elements-remaining in the molten salt; and (5) salt waste treatment [2]. In particular, in the electrowinning stage, in which a liquid cadmium electrode is used as the anode to recover the actinides remaining in the molten salt, rare-earth fission products dissolved in the electrolyte are likely to be reduced as well because they have quasi-identical reduction potentials to those of the actinides. Consequently, rare-earth elements are found in the liquid cadmium electrode as impurities, in addition to actinides after pyroprocessing. One primary focus in the ongoing pyroprocessing research is developing methods for separating high-purity actinides from the materials gained from electrowinning, given that the separating efficiency of pyroprocessing is directly associated with spent nuclear fuel management efficiency and cost effectiveness through fuel recycling.

In this study, we designed an additional stage in pyroprocessing after electrowinning to retrieve high-purity actinides. It was intended to establish a selective separation method for separately extracting actinides (U and Pu) and rare-earth elements (Nd, Pr, Ce, and La) using the differences in solubility and diffusion coefficient of each element in a liquid cadmium electrode containing all elements reduced by electrowinning. In particular, by opening the partitioned liquid cadmium electrode cells stepwise, we first recovered high-purity actinides with high solubility and diffusion rates under a potentiostatic condition. Furthermore, by establishing a conceptual model for the aforementioned conditions, we could determine final yields and expected efficiencies, depending on the recovery time and recovery step.

2. Reactor setup and theoretical analysis

2.1. Example of a real reactor setup

Fig. 1 depicts an actual example of a reactor that implements multistep selective recovery of actinides in liquid metal electrodes. Partition walls divide the liquid cadmium electrode within the reactor into several cells (1-5) in Fig. 1). Prior to beginning the selective recovery process, only the liquid cadmium in the left-most cell contains actinides and rare-earth elements (based on a literature review, it was hypothesized in this study that only U, Pu, Nd, Pr, Ce, and La exist [3]). When the first partition wall (1) is opened and the elements contained in the liquid cadmium diffuse into the next cell, U and Pu reach the back of the cell (partition wall 2) first because of their large diffusion coefficients; they exist in high purity because other elements have smaller diffusion coefficients and thus diffuse more slowly. U and Pu are then electrochemically oxidized at the interface between the liquid cadmium and molten salt, and discharged as ions into the molten salt. These ions are then electrochemically reduced and recovered to the solid cathode. Over time, the rare-earth elements that diffuse relatively slowly-Nd, Pr, Ce, and La-reach the backside of the cell, and the purity of the recovered U and Pu decreases if the aforementioned process step continues. Limiting the first U and Pu recovery step to the time point that ensures their purity prevents this. We then discontinue the recovery process and stir the cadmium electrode to homogenize the concentration, followed by the opening of the second partition wall (2). The second recovery takes place in the same manner, and this recovery process is repeated by



Fig. 1 – Example of a reactor for multistep selective separation of actinides within a liquid metal electrode.

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