High-throughput Electrorefining System with Graphite Cathodes and a Bucket-type Deposit Retriever

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Abstract

An electrorefining is a key element in pyroprocess because uranium, composed of more than 90% spent fuel, is recovered during this step. Therefore, it is one of the important study trends in pyroprocessing for electrorefiner to become high-throughput. To increase the throughput, KAERI applied a graphite cathode, which has a self-scraping characteristic thus the uranium deposit on the graphite electrode falls spontaneously, and a bucket-type deposit retriever, which enables to draw the uranium deposit out continuously. All of these operations were verified with a bucket-type retrieving apparatus, which showed a significant increase of electrorefining throughput.

Keywords: Electrorefining; High-throughput; Pyroprocess; Salt distillation; Ingot casting; UCl$_3$

1. Introduction

Since 1997, a Korea Atomic Energy Research Institute (KAERI) pyroprocessing comprising several processes such as head-end process, oxide reduction, electrorefining, electrowinning, and waste salt treatment has been developed. Electrorefining, a pyrochemical process to recover uranium from a metallic or electrolytically reduced spent fuel, is one of the most important processes because about 93% of spent fuel is uranium. During the electrorefining process, the uranium, transuranic (TRU), and rare earth (RE) elements in reduced metal are dissolved into LiCl-KCl eutectic salt, whereas only uranium is electro-transported and deposited as a pure dendritic form on a solid cathode.
As the size of the electrorefiner increases, some concepts to increase the throughput such as an increase of the cathodic area, a decrease in the distance between the anode and cathode electrodes, and continuous operation without interruption, have been extensively studied. Good examples are Mark-V of the US Idaho National Laboratory (INL) [1], a planar electrode electrorefiner (PEER) of Argonne National Laboratory (ANL) [2], and a high-throughput electrorefiner of Japan Central Research Institute of Electric Power Industry (CRIEPI) [3]. As a cathode is made of steel, however, the complete scraping of uranium deposits from the surface of a steel cathode is impossible [1].

To increase the throughput of an electrorefiner, KAERI has developed innovative concepts; i) separation of electrorefining into two processes: a uranium deposit process (electrorefining) with a solid cathode, and electrowinning to co-deposit TRU and RE with liquid cadmium cathode (LCC), ii) the separation of UC\(_3\) from the electrorefiner, and iii) application of graphite as a solid cathode and a bucket-type uranium deposit retriever. The first two concepts enable minimizing the interruptions owing to a change of operation mode or an addition of UC\(_3\), which results in an increase in throughput. The third process was already partially suggested several years ago when a graphite cathode and screw-type deposit transfer were applied in a high-throughput electrorefiner [4]. However, the screw-type deposit transfer was plugged up with very sticky, jelly-like uranium deposit and salt mixture, and thus was not realized at that time. Recently, we developed a new concept, a bucket-type deposit retriever, to draw a uranium deposit out of molten salt without interruption such as by scraping or reversing the cathode polarity, and successfully tested it with a uranium deposit in molten salt. Therefore, in this context, the bucket-type deposit retriever was introduced combined with a graphite cathode, which has a self-scraping characteristic.

2. Graphite cathode

When steel, which was first applied as a solid cathode, is used in an electrorefiner as a cathode electrode, the interaction between the steel and uranium deposit is too strong to scrap the deposit from the steel surface perfectly. Thus, the reverse of the cathode polarity, which results in a significant decrease of the current efficiency of the electrorefiner, might be needed to eliminate the remaining deposit and clean up the cathode surface [1]. Our previous research showed that the graphite cathode minimizes the sticking of the uranium deposit, and this kind of stripping operation is therefore not needed. During the electrorefining, uranium deposits as a uranium-graphite intercalation compound at the outermost layer, and a dendritic uranium deposit is then self-scraped, fallen down, and collected at the bottom of the electrorefiner [6].

A lab-scale cylindrical electrorefiner, in which 24 graphite cathode electrodes are located at a double layered array in the core part, and the rotating mesh type anode basket consists of 4 cartridges placed at the periphery of the electrorefiner, was developed, as shown in Fig 1(a). The photographs before and after the self-scrapping are shown in Figs. 1 (b) and (c), respectively.

![Fig. 1. (a) electrorefiner with graphite cathode; (b) before and (c) after self-scraping](image-url)
3. Bucket-type deposit retriever

The second problem to solve for an increase in throughput is how to draw out the uranium deposit, which is placed within molten salt on the bottom of the electrorefiner. Since the scrapping was performed without a stop during the electrorefining using the graphite cathode, this drawing out operation has also been attempted by an automatic and continuous method, screw-type transfer, as shown in Fig. 2(a). The screw shaft near the upper layer of molten salt was, however, plugged up with a very sticky, jelly-like uranium deposit and a salt mixture, as shown in Fig. 2(b), and thus the uranium deposit was not transferred. Our second trial using a shaftless screw, as shown in Fig. 2(c), resulted in no plugging, but the dendritic uranium deposit was grinded without a transfer.

Recently, a new concept, a bucket-type deposit retriever, was developed, as shown in Fig 3(a). A bucket with many small holes is located at the lower part of the transfer pipe, which is opened and connected to the bottom of the electrorefiner. A rotating scrapper, which is placed at the bottom of the electrorefiner, turns around and collects the dendritic uranium deposit, and then puts them in the bucket. While the bucket filled with uranium deposit moves upside, the molten salt sneaks out of the bucket. When the bucket arrives at the upper position, the lower slide of the bucket opens and the deposit pours out, as shown in Fig. 3(c). All of these operations were performed and verified without interruption in a bucket-type retrieving apparatus, as shown in Fig. 3(b).
4. Conclusion

To increase the throughput of electrorefining, KAERI has developed a graphite cathode and a bucket-type deposit retriever. The graphite cathode has a self-scraping characteristic, and thus dendritic uranium deposits on the graphite electrode fall spontaneously. Uranium deposits at the bottom of electrorefiner are collected and put into a bucket by a rotating scraper, and then retrieved out of the molten salt in the electrorefiner. All of these operations are continuously progressed without interruption, which enables throughput electrorefining.

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References