

U/RE Recovery Using Large-Scale LCC Electrowinning System

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1. Introduction

The electrowinning system of KAERI is composed of three processes : (i) LCC (liquid cadmium cathode) electrowinning process for group recovery of actinides [1, 2], (ii) RAR (residual actinides recovery) process to collect most of residual actinides in a molten salt after the LCC electrowinning process, and (iii) Cd (Cadmium) distillation process of LCC deposits obtained from LCC electrowinning and RAR processes. The recovered U/TRUs are transferred to an ingot casting furnace to produce a metal fuel ingot. In this experiment, thermogravimetry (TG) was used to make a metal ingot. In this study, large-scale electrowinning system has been tested to obtain U/RE ingot using a LiCl-KCl eutectic salt containing U and rare earth elements (REs) such as Nd, Ce, and La.

2. Experimental

For the experiments of LCC process, several electrodes such as a graphite anode, a Cd cathode, a reference electrode (Ag/1mol%AgCl) and a monitoring electrode (glassy carbon anode + tungsten cathode) were used as shown in Fig. 1. The configuration of graphite anode is shown as Fig. 1(a). Namely, a graphite rod is serially surrounded by dense Al₂O₃ tube and porous SiC tube. Because electrolyte salt can be migrated and diffused to the surface of the graphite through the pores of the SiC tube, the electrode reaction can be properly happened. Cl₂ gas created on the surface of the graphite during the electrode reaction will be moved toward the Cl₂ vent line. In this stage, the dense Al₂O₃ tube plays a role of preventing Cl₂ gas from diffusing out of the anode housing. About 7.3 kg of Cd was contained in a new BeO crucible manufactured by MATERION Corporation in the USA. A potentiostat with the maximum current of 20 A (Bio Logic Company) was used for this study.

Thermogravimetry (TG) shown in Fig. 2 was used for the consolidation process of the product recovered from the Cd distiller. TG is composed of the 2-zone

furnace and glove box. Operating temperature can be increased to 1200 °C below 0.1 torr.

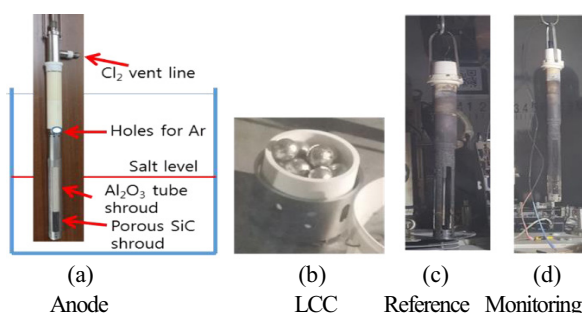


Fig. 1. Electrodes used for LCC process.

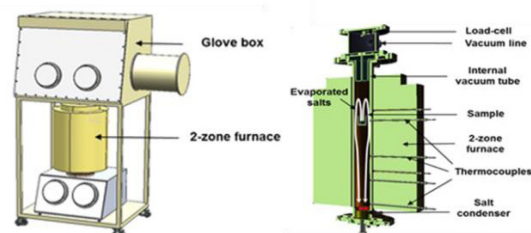
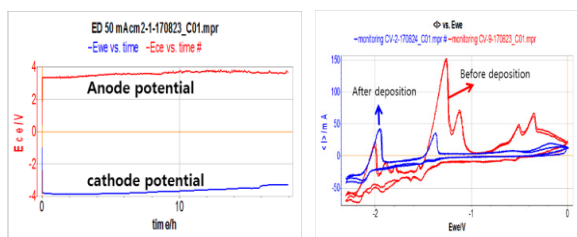


Fig. 2. Thermogravimetry equipment.

3. Results and Discussion

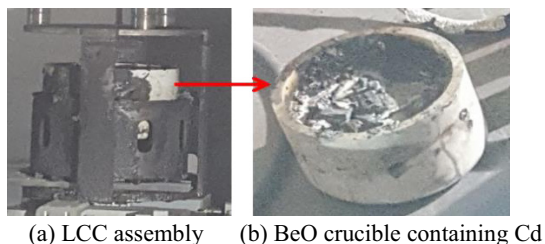
Fig. 3(a) shows the variation of the anode and cathode potential at the current density of 50 mA/cm² for 18 hours around 500 °C, where liquid Cd was used as a cathode. Cathode potential gradually moved to the positive direction as the time increases to 16 hours. Beyond that time, the cathode potential went abruptly upward. So, the deposition was terminated on that time.

Fig. 3(b) shows the comparison of the CVs before and after the deposition. Before the deposition, peak height of U was higher than that of RE, showing the concentration of U is larger than that of RE. By the way, the peak height of U is lower than that of RE in the salt after the deposition, indicating much more U ions rather than RE ions were deposited on the liquid Cd. Oxidation peaks between -0.4 and -0.6V disappeared and thereby multiple peaks were not also seen.



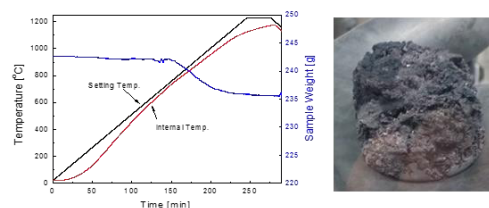
(a) Potential (b) CV result
 Fig. 3. Potential change during the deposition and comparison of CVs before and after deposition.

After the electrodeposition, LCC assembly was raised above the salt to see deposits and the status of the BeO crucible. Some deposits were observed to be attached along the outer surface of the crucible as shown in Fig. 4(a). Fig. 4(b) is the BeO crucible separated from the LCC assembly. It is observed that some deposits were attached along the inner surface of the crucible. It is predicted from Fig. 3 and Fig. 4 that some deposits began to grow out of the crucible from beyond the 16 hours deposition and so U amount recovered in Cd might be decreased compared to the theoretical U amount.



(a) LCC assembly (b) BeO crucible containing Cd
 Fig. 4. BeO crucible containing Cd deposits.

Cd ingot containing U and RE elements obtained through LCC process was transferred to Cd distiller to remove Cd. The Cd in the LCC deposit was distilled very slowly at low temperature (600 °C) to avoid a Cd splatter. And then, the distillation experiment was performed at 900 °C for the evaporation of the covered salt. U/RE product of about 107g was recovered from Cd distiller. For the consolidation process, U/RE product was contained into the Al₂O₃ crucible having a diameter of 5cm and the Al₂O₃ crucible was installed into the sample position of TG as shown in Fig. 2. After the consolidation process at 1200 °C around 0.1 torr, the weight loss of the sample was about 6g as shown in Fig. 5(a). The volume of the product in Al₂O₃ crucible decreased after the consolidation process. But, the product of Fig. 5(b) was not metal ingot and relatively easily broken to pieces by some shock. It was considered that most of the U/RE product had been considerably oxidized during the Cd distillation.



(a) Weight change (b) U/RE product
 Fig. 5. Weight change and U/RE product after consolidation.

(a) Weight change (b) U/RE product

4. Summary

U/RE was electrochemically recovered to LCC at 50 mA/cm² from LiCl-KCl salt containing 0.5wt%UCl₃, 0.22wt%NdCl₃, 0.15wt%CeCl₃ and 0.07wt%LaCl₃. The Cd in the LCC deposit was removed during the distillation using Cd distiller. U/RE product of 107g obtained from the distiller was installed to TG and then heated to 1200 °C to be consolidated. Dense U/RE metal ingot was not acquired through the consolidation process because U/RE product had been partially already oxidized during the distillation process.

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