Recovery of RE-less U From U/RE Ingot by Electrochemical Oxidation Process

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1. Introduction

SFR fuel is majorly composed of U and Pu, where the amount of impurities such as RE (rare earth) needs to be controlled as low as possible, for example, below 1% of the fuel composition. But, it is very difficult to recover U/Pu material having so low RE amount from LCC electrowinning process [1, 2]. In order to decrease RE amount in the U/Pu/RE mixture obtained from LCC electrowinning, some treatments should be done. In this study, selective oxidation behavior of RE components in U/RE ingot was studied using an electrochemical process.

2. Experimental

Fig. 1 shows the experimental configuration for the electrochemical oxidation process, where LiCl-KCl-3.6wt% LaCl₃ was contained in an alumina crucible. U/Ce/Nd anode ingot having a weight of about 3.8 g was contained in an alumina crucible and zinc cathode buttons having a weight of about 35g in another alumina crucible as shown in Fig. 1. For the measurement of the potentials of the electrodes, Ag/AgCl electrode was used as a reference. The melting point of zinc is about 420 °C and so the zinc buttons are melted at the reaction temperature of 500 °C.

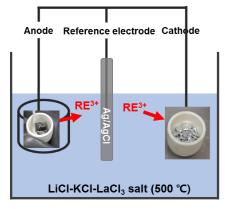
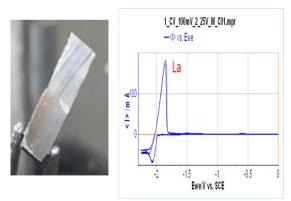


Fig. 1. Experimental configuration for electrochemical oxidation process.

Fig. 2(a) shows the U/Ce/Nd ingot, where upper and lower parts of the ingot show the different colors.

This is owing to the density difference of the U/Ce/Nd. The densities of the U and RE (Ce/Nd) was about 19 g/cm³ and 7 g/cm³, respectively. According to the EPMA analysis, some RE particles was seen in U matrix and some U particles in RE matrix. Fig. 2(b) shows the CV result of the LiCl-KCl-LaCl₃ salt at 500 °C. Any other components was not detected except La in the salt.



(a) U/Ce/Nd ingot (b) CV in LiCl-KCl-LaCl₃ salt

Fig. 2. The anode ingot and CV result of the LiCl-KCl-LaCl $_3$ salt.

3. Results and Discussion

3.1 Oxidation of the anode

After the anode and the cathode was connected to a potentiostat equipment, -1.7V was applied to the anode as a working electrode. Fig. 3(a) shows the variation of the zinc cathode potential and the current, respectively. About 250 mA was applied at the initial time and nearly 0.2 mA after 5 hours. Current value of 0 mA means no reaction between the electrodes. So, the oxidation experiment was terminated around 6 hours. Fig. 3(b) shows the CV result of the final salt to check the existence of U in the salt. U peak was not seen. During the oxidation process, it is expected that most of the Ce/Nd components was oxidized to the chloride forms and some La/Ce/Nd components was deposited to the liquid zinc cathode.

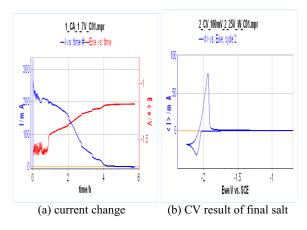


Fig. 3. Change of current value during the oxidation process and CV result of the final salt.

3.2 SEM/EPMA analysis

Fig. 4 shows the SEM/EPMA photographs of the anode ingot after the oxidation reaction. Only U part was remained and RE matrix part was all oxidized into the salt. But, RE particles still existed in the U matrix part even after the oxidation. It is predicted from this result that most of the RE particles distributed in the U matrix would not be oxidized because RE particles would be predominantly distributed in the inner part rather than the surface part of U matrix.

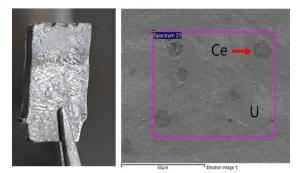


Fig. 4. SEM/EPMA analysis results of anode ingot after the oxidation reaction.

3.3 Distillation of the zinc cathode

While the anode was oxidized into the salt, metal ions of the salt would be deposited into the zinc cathode. After the oxidation was terminated, zinc cathode product was distilled in the alumina crucible around 850 °C at 1 torr to recover the deposited metal components. Fig. 5(a) shows the variation of the temperature and the weight during the distillation. Weight difference of about 35g occurred and it corresponds to the weight of the zinc cathode material. Fig. 5(b) shows the metal product remained in the alumina crucible after the distillation. The metal is being chemically analyzed to identify its composition.

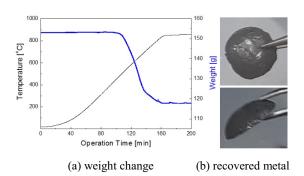


Fig. 5. Weight loss and the recovered metal after the distillation of the zinc cathode product.

4. Summary

Selective oxidation of RE elements from the U/RE metal ingot was studied in this paper using electrochemical process. Constant potential of -1.7V was applied between anode and cathode, where the potential value corresponds to standard potentials between actinide and rare earth materials. When the current values approached to nearly 0 mA, the reaction was finished. It is confirmed from the EPMA analysis that only U part of the U/RE ingot was remained. The metal recovered to the zinc cathode was obtained through the distillation process and it is being chemically analyzed in the KAERI analytical laboratory.

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REFERENCES

- [1] Li, S.X.; Herrmann, S.D.; Goff, K.M.; Simpson, M.F.; Benedict, R.W. (2009) Actinide recovery experiments with bench-scale liquid cadmium cathode in real fission product-laden molten salt. Nucl. Mater. 165:190-199.
- [2] Uozumi, K.; Iizuka, M.; Kato, T.; Inoue, T.; Shirai. O.; Iwai, T.; Arai, Y. (2004) Electrochemical behaviors of uranium and plutonium at simultaneous recoveries into liquid cadmium cathode. J. Nucl. Mater.325:34-43.