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Study of the reaction between Uranium(III) and Lanthanide oxide by using the UV-VIS spectrophotometer.

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1. Introduction

Recently, ionic melts have become attractive reaction media in many fields.[1] Molten salt based electrochemical processes have been proposed as a promising method for future nuclear programs and more specifically for spent fuel processing.[2] Molten alkaline chloride based melts are considered as a promising reaction media. For this, it is interesting to understand the chemical nature of the actinides and lanthanides in high-temperature melt. Some spectroscopy provides essential information on the exact nature of f-block elements–LiCl-KCl melt system. The knowledge on the basic chemical properties of these lanthanide oxides and U(III) in molten salt media is essential for developing suitable processes. However, few studies have been reported until now on the interaction between U metal and lanthanide oxides in LiCl-KCl melt. So, we studied the interaction between U(III) and Ln(III) by using the UV-VIS spectra. UV-vis spectrometry is a strong analytical technique for characterizing chemical species and their behavior in molten salt.

2. Experimental

The spectrometer component was purchased from Ocean Optics, Inc.(Model USB 2000). Data collection was done by interfacing with PC via USB port.

The light beam passes through an optical fiber into the sample chamber. Suitable quartz lens and iris were used to collimate the beam path and adjust the intensity. Figure 1 presents the schematic diagram of the apparatus and equipments. LiCl-KCl was weighed and put into the quartz cell, and the cell was fixed in the sample holder made at the top and bottom of the reaction cell unit.

All the experiments were carried out in a glove box system. The inert atmosphere was maintained by purging with purified Ar gas. The oxygen contents and H_2O are minimized to be less than 2 ppm. The LiCl-KCl eutectic (41.5 mole percent KCl) was prepared from A.R. grade reagent. U(III) were generated by reacting uranium metal with cadmium chloride in the melt and the spectra were recorded in-situ. Eu_2O_3, Y_2O_3 and CeO_2 was obtained from Alfa Aesar Co. Ltd. (99.99% purity).

3. Results and Discussion

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By using the hardware system described in an earlier section, we were able to collect UV-VIS spectra.

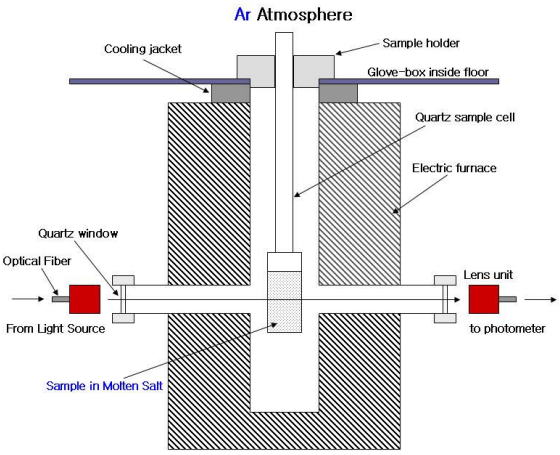

Figure 1. Block diagram of the measurement system in high-temperature molten salt media.

Figure 2 presents the generation of U(III) species by the reaction of U metal with cadmium chloride in the melt (723 K). The U(III) spectra matched well with those of Prof. Yamana's group (Kyoto University, by private communications). After Ln_2O_3 powder (Ln = Eu, Y, Ce) was added into U(III) molten salt, the changed absorption spectrum shows in Figure 2. As shown as Figure 2, first spectrum was observed typical peak of U(III). As long as pasted the time, U(III) band was decreased. In the other hand, the new band was increased after the reaction with U(III) molten salt. Then the new band was confirmed to the absorption band of $LnCl_3$. This results can be expressed by the reactions :

$$2U\ (metal) + 3CdCl_2 \rightarrow 2UCl_3 + 3Cd\ (metal)$$

$$2UCl_3 + Ln_2O_3 \rightarrow UO_2 + UO + 2LnCl_3$$

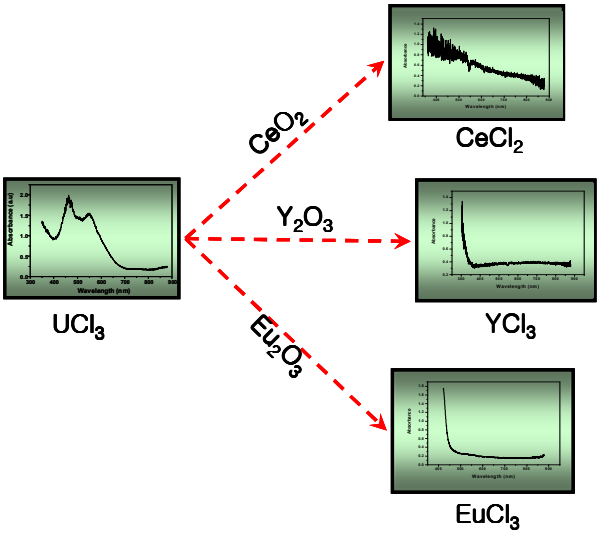

Fig 2. Uv-vis spectra of between U(III) and lanthanide oxides.

4. Conclusions

An optical fiber based UV-VIS spectrophotometric systems combined with glove box was developed for the measurement of a lanthanide elements in controlled chemical environment at high temperature molten salt media. It exhibited a good performance. By using this measuring system, we confirmed the reaction between U(III) and Ln_2O_3.

Acknowledgement

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