On-line measurements of UV-VIS spectra in high-temperature molten salt media: Development of measuring systems

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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. High temperature electronic absorption spectroscopy challenges researchers to design and build specific apparatus/equipments and maintain certain strict physicochemical conditions. First of all, to reach to that goal, it is necessary to setup special apparatus and measuring equipments. Here, we report the details of the design of the reaction system combined with the instrumentation of the spectrometer system. Also, application result of the measuring system to U(III) involved chemical reaction in molten salt media was introduced.

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 a 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. Nd(III) spectra were obtained by dissolving $NaCl_3$ in LiCl-KCl (450 $^{\circ}$C)

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 2 presents the generation of U(III) species by the reaction of U metal with cadmium chloride in the melt (450 ^{o}C). The U(III) spectra matched well with those of Prof. Yamana's group (Kyoto University, by private communications). The information on the nature of U(III) in molten salt media is still very rare. The intensity of the absorbed peak is proportional to the concentration of U(III) produced. This implies that on-line monitoring of the process in molten salt media is possible where U(III) species are involved. This technique may be widely usable for many applications.

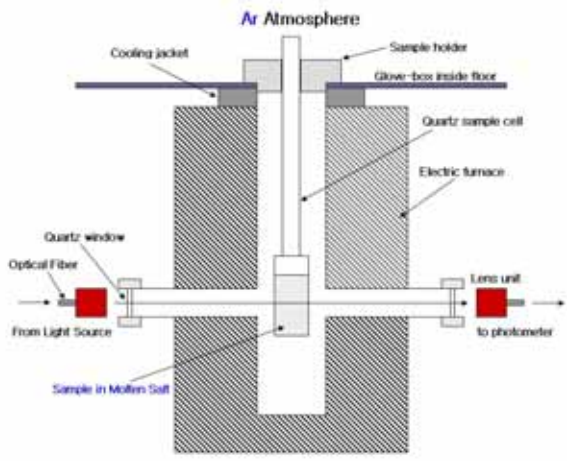

Figure 1. Block diagram of the measurement system in high-temperature molten salt media.

Also the same measurement has been applied to one lanthanide element, neodymium(III). The intensity of the spectra was much lower than that of U(III). The details of the spectra and application are introduced elsewhere.[3] The spectral feature of Nd(III) matched well with reported in recent journal. The spectral pattern was variable depending on the conditions of measurement.[4.5.6]

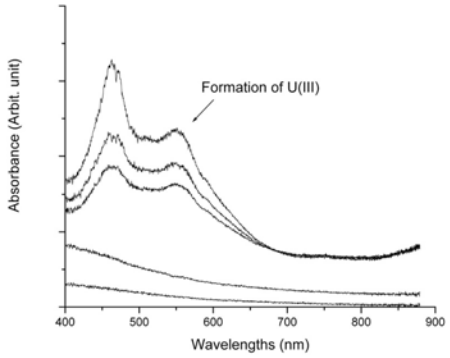

Figure 2. On-line monitoring of U(III) species generated from the oxidation of U metal by reaction with Cd(II) in molten LiCl-KCl eutectic melt..

4. Conclusions

An optical fiber based UV-VIS spectrophotometric systems combined with glove box was developed for the measurement of actinide and lanthanide elements in controlled chemical environment at high temperature molten salt media. It exhibited good performance.

By using this measuring system, on-line data acquisition of UV-VIS spectra of U(III) and Nd(III) was successful.

Acknowledgement

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