

Electronic Absorption and Fluorescence Spectra of Trivalent Erbium Ions in LiCl-KCl Eutectic Melt at High Temperature

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1. INTRODUCTION

Extensive studies have been conducted on the pyro-chemical reprocessing technique by virtue of its intrinsic contribution for use as a recycling technique of spent nuclear fuel with a nuclear non-proliferation feature. Consequently, the reuse of spent nuclear fuel leads to a reduction of high radioactive wastes. The pyro-chemical reprocessing technique is based on the concept that electrochemical procedures associated with electro-reduction, electro-refining and electro-winning separates actinide elements from the spent nuclear fuel dissolved in a molten salt to reuse them as a nuclear fuel. Therefore, it is important to understand the chemical characteristics of fission products containing lanthanide and actinide elements in molten salts for realizing the pyrochemical reprocessing system. [1-3] In this study, we used the optical spectroscopy in conjunction with fluorescence emission, absorption spectra, and the decay curve to characterize chemical behavior of the lanthanide elements. Spectroscopic measurement was performed on trivalent Erbium (Er^{3+}) dissolved in a molten LiCl-KCl eutectic bath.

2. BODY

2.1 EXPERIMENT

An electric furnace apparatus was installed under an Ar atmosphere glove box. The inner furnace accessible via glove box is therefore filled with Ar gas that limits an exposure to oxygen and humid. A narrow rectangular quartz cell (diameter: 10 mm) was erected along the vertical path of an inner furnace. A thermocouple was linked with the quartz cell to monitor the temperature. LiCl-KCl mixture

(Sigma-Aldrich) was put into the cell and then melted by the raised temperature of 400°C. Prior to spectroscopic measurement, bubbles inside the molten salt were eliminated by waving the cell. A frequency-tripled, Q-switched, Nd:YAG laser (Quantel, Brilliant) was operated at 10 Hz. The 355-nm laser light (pulse duration: ~6 ns) was delivered to the molten salt with a pulse laser energy of 10 mJ. The fluorescence emission collected through an optical fiber ($2 \times 8 \text{ mm}^2$, 345 fibers) was fed via an entrance slit (40 μm) into a monochromator (Acton Pro, grating: 1200 lines/mm) fitted with a photomultiplier tube (PMT - Hamamatsu, R928). For the absorbance measurement, we utilized the halogen lamp as an excitation source and followed the same procedure of fluorescence measurement. Spectra-Sense software was used to acquire the spectral intensities versus wavelength. For recording the time-resolved fluorescence signals, the output of the PMT via preamplifier (SR445A, SRS) was coupled to the digital oscilloscope (Agilent, Infiniium 8000). The timing of signal acquisition in the oscilloscope is triggered and synchronized by the Q-switch signal of laser.

2.2 RESULTS AND DISCUSSION

Electronic absorption and fluorescence spectra, aspectral signature indicating the presence of excited elements, were recorded in a LiCl-KCl eutectic melt at 400°C. The absorption spectra were recorded in the wavelength region of 300-900 nm. The two absorption bands of Er^{3+} at 380nm and 524 nm corresponding to the f-f hypersensitive transitions $^4I_{15/2} \rightarrow ^4G_{11/2}$ and $^4I_{15/2} \rightarrow ^2H_{11/2}$ were observed. [4, 5] A hypersensitive transition is a transition in which the oscillator strength is sensitive to the coordination characteristics. The spectra arising from the f-f

transitions are relatively narrow. It is attributed to the fact that 4f orbitals are located below the 6s, 5p, and 5d orbitals, and therefore, the 4f electrons are well shielded from the ligand field generated by neighboring ions. The several fluorescence emissions of Er^{3+} after excitation by the 355nm radiation were observed in the wavelength region of 360-700 nm. (See Fig. 1) The five Er^{3+} ion emission peaks are seen at 380nm (${}^4\text{G}_{11/2} \rightarrow {}^4\text{I}_{15/2}$), 409nm (${}^2\text{G}_{9/2}, {}^4\text{F}_{9/2}, {}^2\text{H}_{9/2} \rightarrow {}^4\text{I}_{15/2}$), 482nm (${}^4\text{F}_{7/2} \rightarrow {}^4\text{I}_{15/2}$), 524nm (${}^2\text{H}_{11/2} \rightarrow {}^4\text{I}_{15/2}$), and 558nm (${}^4\text{S}_{3/2} \rightarrow {}^4\text{I}_{15/2}$). [6] Furthermore, we recorded the fluorescence emission for various concentrations of ErCl_3 . The ErCl_3 concentration varied from 0.0 wt.%, 0.05 wt.%, 0.1 wt.%, 0.175 wt.%, 0.5 wt.%, 1.0 wt.%. The emission intensity of Er^{3+} becomes enhanced with an increase in Er^{3+} concentration.

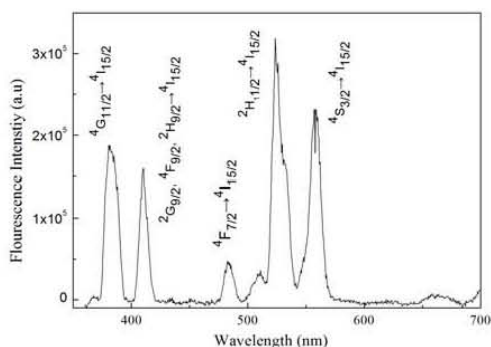


Fig. 1. Fluorescence spectrum of Er^{3+} dissolved in a LiCl-KCl eutectic containing 0.5 wt.% of ErCl_3 .

3. CONCLUSION

Spectroscopic properties of Er^{3+} dissolved in a LiCl-KCl eutectic molten salt were investigated on the basis of the absorption and emission spectra. Using excitation wavelength at 355 nm, the several prominent emission bands for Er^{3+} ions were detected.

4. ACKNOWLEDGEMENT

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5. REFERENCES

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