Short Range Structure Of Molten Cscl - Nacl Mixtures Obtained By Xafs Analysis

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Introduction

In the course of development of pyrochemical reprocessing of nuclear oxide fuels, uranium and plutonium oxide (MOX) co-electrodeposition process is most crucial from the technical point of view. Usually, molten 2CsCl-NaCl is preferred as melt bath for MOX codeposition, however the mechanism of the process is still unknown. Furthermore, even the physico-chemical properties and microscopic structure of the melt bath themselves are still unclear. In the present study, we mainly focused on the structural elucidation of the melt bath, and the coordination behaviour around the Cs ions by using XAFS method.

Experimental

Several grams of finely grained chemicals were inserted into specially designed thin quartz containers of varying X-ray paths in an Ar atmosphere glove box. The quartz containers were sealed under vacuum. The electric furnace used differed slightly from the one originally designed by us for the laboratory XAFS equipment of Rigaku Corp[1)] in that it now had a cylindrical core. We carried out XAFS measurements of CsCl and 2CsCl-NaCl systems at room temperature and above the melting point, by using Cs-K X-ray absorption edge (35.98keV) at BL01B1 and BL19B2, SPring-8.

Results and Discussion

The Extracted □(k) EXAFS spectra and Fourier transformed structural functions of pure CsCl and 2CsCl-NaCl at room temperatures and 973 K are shown in Fig. 1 and 2, respectively. Similar to the observation in [2)], small phase shift in the current EXAFS spectra during melting is observed. Due to shortage of the core-hole lifetime at the X-ray absorption edge of Cs, XAFS oscillations detected are relatively small than those previously reported for molten $LaCl_3$ [3)]. The Cs–Cl distance, $r_{\mathrm{Cs\text{-}Cl}}$, and the coordination number, $N_{\mathrm{Cs\text{-}Cl}}$, corresponding to the Cs–Cl 1st coordination shell are obtained by the curve-fitting method at the first peak of FT|k^3□(k)| in R-space, including the 3rd and 4th cumulants. Both, $r_{\mathrm{Cs\text{-}Cl}}$ and $N_{\mathrm{Cs\text{-}Cl}}$ decrease drastically on melting, as already suggested for several other alkali metal halides. In the mixture with NaCl, the $r_{\mathrm{Cs\text{-}Cl}}$ distance is found to be slightly longer than that in pure molten CsCl. We will provide structural parameters obtained by XAFS as well as another complementary techniques in the presentation.

References

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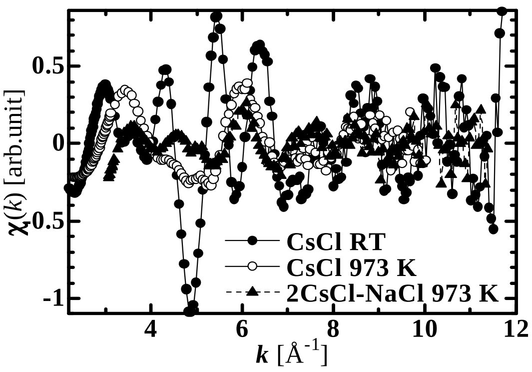

Fig. 1 **EXAFS $\chi(k)$ spectra of CsCl and 2CsCl - NaCl.**

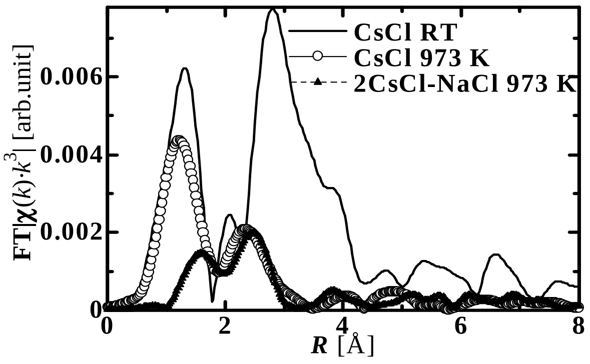

Fig. 2 **Structural functions of CsCl and 2CsCl – NaCl obtained by XAFS.**