

XAFS analyses on yttrium fluorides added oxides at high temperatures

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Introduction

We have proposed molten alkali metal fluorides as melt baths for the pyrochemical reprocessing of nuclear fuels, and investigated the structural characteristics of some of the molten fluoride systems [1,2]. In order to develop the pyrochemical process using molten fluoride baths, structural information of these molten salts should be elucidated systematically. In general, molten fluorides can dissolve oxide species, thus it is very essential to control the oxide contents in all the process designed. In this study, we used LiF-YF₃ ($x_{\text{YF}_3}=0.2$) mixture salts, and investigated the local structure variations as increasing oxide contents at high temperatures by XAFS.

Experimental

The known amounts of LiF, YF₃, and Li₂O were homogeneously dispersed in dried boron nitride powder, pressed into pellets, and installed in specific boron nitride holders. For each measurement, a sample was installed in an electric furnace located between the ionization chambers, and heated from 300 to 1073 K. During 48 hours of beamtime, transmitted XAFS spectra have been collected, using Si (111) double crystal monochromator at Y-K X-ray absorption edge.

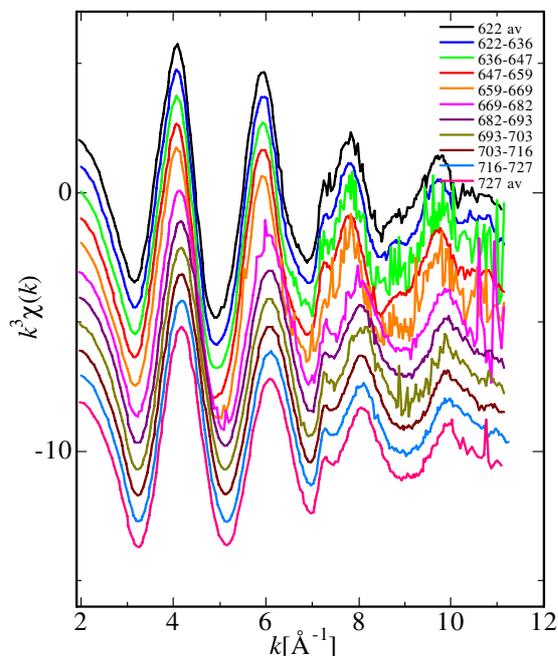


Fig. 1 EXAFS oscillations of LiF-YF₃-Li₂O ($x_{\text{YF}_3}=0.2$, $x_{\text{Li}_2\text{O}}=0.1$) at various temperatures

Results and discussion

The extracted XAFS oscillations $\chi(k) \cdot k^3$ during raising temperature 622°C to 727°C slowly are shown in Fig. 1. Between the spectra of 659°C-669°C and 669°C-682°C, one can identify slight phase shift, which is the typical tendency observed when the sample is going to be molten. In all series of measurement on LiF-YF₃-Li₂O ($x_{\text{YF}_3}=0.2$, $x_{\text{Li}_2\text{O}}=0, 0.02, 0.05, 0.1$), there are no significant variations between the temperatures when the phase shifts occur. Thus we conclude that such oxide amounts don't affect melting behaviour of the mixture LiF-YF₃ ($x_{\text{YF}_3}=0.2$). However, we carefully investigate the spectra after cooling down, and notice specific features in Fourier transformed radial structure factors, as shown in Fig. 2. In the case of LiF-YF₃-Li₂O ($x_{\text{YF}_3}=0.2$, $x_{\text{Li}_2\text{O}}=0.1$), the 2nd and even 3rd coordination contributions are evolved, which may be corresponding to generate oxide species coordinated to yttrium atoms. Thus in the following beamtime, we are planning to measure on the samples of either reduced yttrium composition or increased oxide amounts in order to investigate the oxide additional effect much precisely.

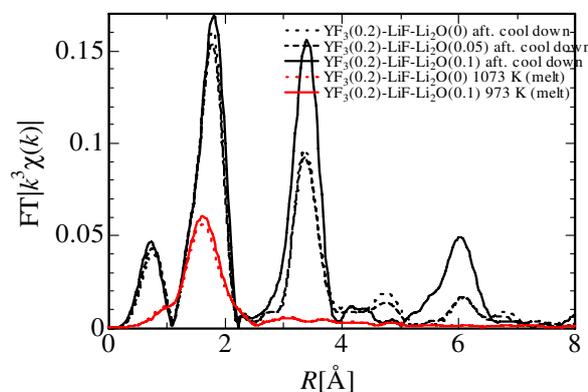


Fig. 2 Radial structure factors of LiF-YF₃-Li₂O ($x_{\text{YF}_3}=0.2$, $x_{\text{Li}_2\text{O}}=0, 0.05$ and 0.1) both in molten phases and after solidified at room temperatures

References

- [1] S. Watanabe et al., J. Alloys Compd, 408-412, 71 (2006).
- [2] S. Watanabe et al, J. Indian Chem. Soc. 82, 1059 (2005).

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