BL-27B/2017G623

XAFS measurement on radioactive waste simulants in various disposal glasses

Kei Shiina[1], Yuri Tajimi[1], Takafumi Uchiyama[1], Toshiaki Kakihara[2], Yasutomo Tajiri[2], and Haruaki Matsuura[1]

[1] Cooperative Major in Nuclear Engineering, Tokyo City University, 1-28-1, Tamazutsumi, Setagaya-ku, Tokyo, 1588557, Japan
[2] IHI Corp, 1, Shin-nakahara-cho, Isogo-ku, Yokohama, 2358501, Japan

1 Introduction

In Japan, high-level radioactive liquid waste generated from reprocessing of spent nuclear fuel is planning to be mixed with glass matrix, filled in a stainless steel canister, cooled down well and put into deep disposal site. At the moment, borosilicate glass is considered to be one of the best candidates of vitrified glass. Furthermore, in order to reduce the cost required for stable preserving vitrified glass for a long period, there is a demand for a technique for increasing packing ratio of wastes in the vitrified glass. In this case, molybdates tend to crystallize out from vitrified glass as a yellow phase (YP) and may affect homogeneity of vitrified glass. Therefore, in order to investigate the optimum composition ratio of vanadium-added borosilicate glass [1,2] which is supposed to suppress YP generation in the case of highly molybdenum filled, Si / B ratio, the alkali content, the alkaline earth metal content have been modified and the local structural change around molybdenum and zirconium in simulated glass has been evaluated by the EXAFS analysis.

2 Experiment

1 g of various glass samples with different Si/B ratio, alkali and alkaline earth metal contents, which also contained all simulated waste liquid components, were pulverized. These samples were installed in-between two alumina plates with 0.5 mm gaps. An alumina cell was installed in an electric furnace placed in-between two ion chambers to obtain the EXAFS oscillation in the molten state, i.e.at 1200 °C and 800 °C. EXAFS oscillation at room temperature has been also collected. The obtained data has been analyzed by the program of WinXAS version 3.02. An EXAFS oscillation was extracted using the cubic spline method and the structure function was obtained by the Fourier transformation. Structural parameters were derived from fitting based on EXAFS equation.

3 Results and Discussion

Comparing the EXAFS oscillations at the molten state and those at room temperature, it can be considered that the glass having a small deviation in the distance between Mo - O would be a stable glass from local structural point of view. Since the glass at high temperature has a disordered structure, the peak becomes small. The structural functions of glasses containing both Ca and Mg have been modified significantly depending on temperature. On the other hand, samples containing single alkaline-earth element have larger temperature dependency than those containing double elements.

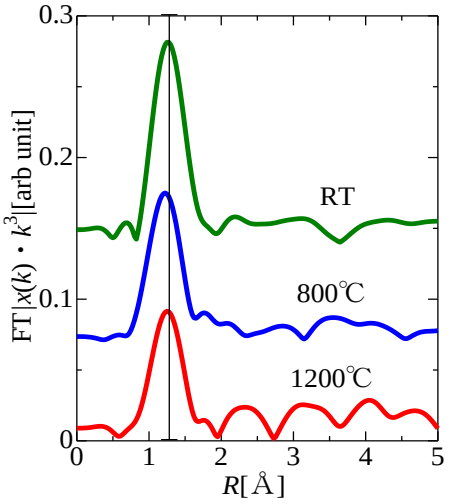

Fig.1 Structural functions of Mo-O depending on the addition of both Mg and Ca.

4 Conclusion

Since the oxygen coordination number of Mo did not change, it does not contribute to the matrix network structure of glass. Also the alkaline-earth metal plays a role of charge compensation, and it becomes more stable glass with double elements addition. By evaluating the local structures of molten and solidified glass, it can make clarify the guidelines of searching optimum glass composition.

References

[1] T. Nabemoto, T. Oniki., Molten salt and high temperature chemistry, 59, 20-24 (2016).
[2] H. Kawashima, T. Hashimoto, T. Kakihara, T. Fukui The fall meeting of the Atomic Energy Society of Japan, 1E03 (2016).

* g1781008@tcu.ac.jp