Local structure analysis of light elements in simulated waste glasses

Atsunobu Masuno[1], Akihiro Yamada[2], Yoshiyuki Miura[3], Toyonari Yaji[4]

1) *Graduate School of Science and Technology, Hirosaki University, 3 Bunkyo-cho, Hirosaki, Aomori 036-8561, Japan*
2) *The University of Shiga Prefecture, 2500, Hassaka-cho, Hikone, Shiga, 522-8533 Japan*
3) *Japan Nuclear Fuel Limited, 4-108 Aza Okitsuke, Oaza Obuchi, Rokkasho, Kamikita-gun, Aomori 039-3212, Japan*
4) *The SR Center, Ritsumeikan University, 1-1-1 Noji-Higashi, Kusatsu 525-8577, Japan*

Nuclear waste liquid generated in reprocessing plant of spent fuel are vitrified by mixing with base borosilicate glass matrix. The important requirements for the nuclear waste glasses are high chemical durability for corrosion due to hydrolysis and large capacity of nuclear waste incorporated in base glasses. Especially, the increase of nuclear waste in the glass is highly desired because it reduces directly the amount of vitrified glass for final disposal.

However, chemical components included in nuclear wastes are mainly heavy metal oxides such as transition metal oxides and rare earth oxides, and they are generally classified into modifier oxides. Therefore, the increase of nuclear wastes inevitably decreases glass forming ability of the base glass, and then crystallization occurs. Crystallization of Na_2MoO_4-based compositions should be avoided because they are soluble to water and reduces chemical durability of the nuclear waste glasses.

It is necessary to consider structural origin of the chemical durability and crystallization phenomena for optimization of base borosilicate glass composition to meet the requirements. However, the glass structure analysis of nuclear waste glasses is challenging work because they contain many kinds of components greater than 30[1, 2]. In this study, we focused on local structures around light elements, especially Na, in waste glasses. Local structure analyses using Na *K*-edge XAFS were carried out to investigate structure-property relationship and to develop glass compositions containing a large amount of nuclear wastes.

Simulated glasses without any radioactive elements were used for the measurements. Typical composition of simulated glasses is $10.87(Li_2O + Na_2O)$-$56.58SiO_2$-$14.81B_2O_3$-$3.84CaO$-$3.56Al_2O_3$-$2.65ZnO$ with 7.7 mol% of components derived from simulated waste liquid. The powdered glasses were pasted on a sheet of carbon tape attached to a metal substrate. Na *K*-edge XAFS spectra at room temperature were obtained at BL-10 in the SR center of Ritsumeikan University.

Figure 1 shows Na *K*-edge XANES spectra of simulated waste glasses. There were small pre-edge peak and two main peaks (b and e) in all glasses. These are characteristic features observed in aluminosilicate compounds[3]. With an increase of B_2O_3 content, the intensity of valley (v) developed, which suggests that many of Na atoms preferred to coordinate BO_3 or BO_4 structural units.

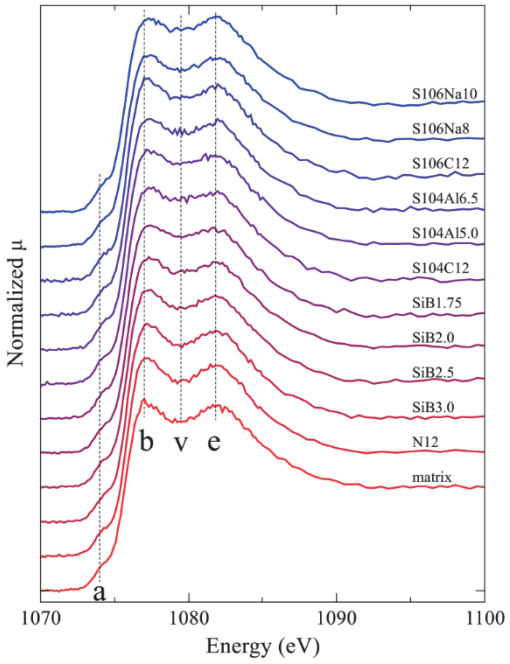

Figure 1. Na *K*-edge XANES spectra of simulated waste glasses.

Acknowledgements

This work was carried out as a part of the basic research programs of vitrification technology for waste volume reduction supported by the Ministry of Economy, Trade and Industry, Japan.

References

[1] G. Calas *et al.*, Comptes Rendus Chimie **5**, 831 (2002).
[2] G. Calas *et al.*, Procedia Material Science **7**, 23 (2014).
[3] D. R. Neuville *et al.*, European Journal of Mineralogy **16**, 809 (2004).