Local structure of simulated high-level radioactive wastes in borosilicate glasses containing vanadium

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[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 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.

[Experimental]

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 inbetween two alumina plates with 0.5 mm gaps. EXAFS measurements in transmission mode were carried out using Mo - K and Zr - K absorption edges on BL - 27B of PF, High Energy Acceleration Research Organization, Tsukuba. An alumina cell was installed in an electric furnace placed inbetween two ion chambers to obtain the EXAFS oscillation in the molten state, i.e.at 1200 ºC and 800 ºC. EXAFS oscillation at room temprature has been also collected.

[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 in a high temperature state has a disordered structure, the peak becomes small. In this glass system, the interatomic distance between Mo - O tends to increase at 1200 ° C, decrease at 800 ° C. and then became increase again at room temperature.

[Conclusion]

By evaluating the local structures of molten and solidified glass, it can make clarify the guidelines of searching optimum glass composition.