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Structural Analysis of Waste Generated by Pyroprocess in Iron Phosphate Glass

Yuri Tajimi [1], Hirohide Kofuji [2], Sou Watanabe [2], Tetsuji Yano [3], Rikiya Kado [3], Takuhiro Miyawaki [3], Motoshi Iwanaga [3], and Haruaki Matsuura [1]

[1] Cooperative Major in Nuclear Engineering, Tokyo City University, 1-28-1, Tamazutsumi, Setagaya-ku, Tokyo, 1588557, Japan
[2] Japan Atomic Energy Agency, 2-4, Shirakata, Nakagun,Toukai-mura, Ibaraki, 3191195, Japan
[3] Tokyo Institute of Technology, 2-12-1, Ookayama, Meguro-ku, Tokyo, 1528552, Japan

1 Introduction

For the vitrification of high-level radioactive waste from pyroprocess, fission products are separated from spent salt as phosphates and the rest of them is vitrified as iron phosphate glass. The glass has high affinity to contain waste salts as main constituents of glass. This fact attracts attention to this system as suitable vitrification media, because this glass system is also known to have high loading capacity of nuclear wastes.

Among fission products, zirconium has been considered to be difficult to dissolve into iron phosphate glass. According to the previous study, Zr ions in glass are stabilized by the coexistence of alkali ions, Cs_2O, which is also one of radwastes generated from the nuclear power plant. Moreover, the chemical stability of the glasses depends in the concentration of these waste elements. [1]

In this study, the coordination structure around the Zr and Cs atoms in iron-phosphate glass have been evaluated by EXAFS structural analysis of the glass samples with different Cs_2O concentration in order to clarify the role of these elements in the glass properties.

2 Experiment

2 mass% of ZrO_2 with 2, 4, 8 and 16 mass% of Cs_2O, as well as 4 mass% of ZrO_2 with 0, 2, 4, 8, 16, 25 and 40 mass% of Cs_2O were added to the iron phosphate glass matrix i.e., $1Cr_2O_3$-$3(CoO)_2$-$4.5Al_2O_3$-$28Fe_2O_3$-$65P_2O_5$ (in mol%). EXAFS measurement of these samples have been carried out by transmission or fluorescence mode at KEK.

The obtained data have been analysed by EXAFS analysis program 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

In the EXAFS oscillation of Zr, there is no remarkable d ifference depending on ZrO_2 concentration. Zr-O distance shows certain dependency on Cs_2Oconcentration below 16 mass%. When Cs_2O concentration is higher than 16 mass%, oscillations become identical.

It is considered that the influence of Cs addition on the local structure of Zr almost disappears at high Cs_2O concentration region more than 16 mass%.

EXAFS analysis of Cs shows that the distance of the nearest neighbor (Cs-O) gradually becomes shorter with increasing Cs_2O concentration. Particularly in the case of the glasses with 2 mass% of ZrO_2 concentration, Cs-O peak shifted greatly between 8 and 16 mass% of Cs_2O concentration. In the case of 4 mass% of ZrO_2 glasses, the great shift is found between 16 and 25 mass% of Cs_2O concentration. It can be said that local structural correlation between Zr and Cs exists in iron-phosphate glass, which is related to the solubility and stability of Zr and Cs in this glass system.

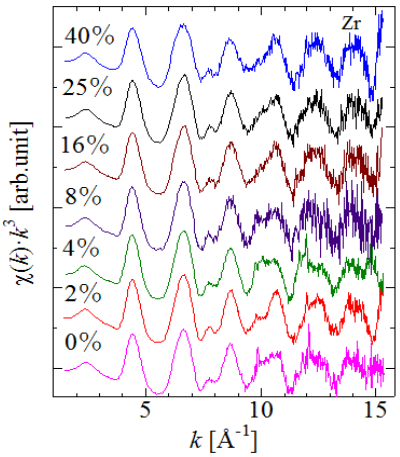

Fig. 1: EXAFS oscillations of Zr in iron phosphate glasses with various Cs_2O concentration (0, 2, 4, 8, 16, 25, 40 mass%).

4 Summary

Correlation of local structure between Zr and Cs exists in iron-phosphate glass at structural variation of Zr-O coordination below 16 mass% of Cs_2O concentration.

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References

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* g1781010@tcu.ac.jp