Vitrification of the spent nuclear fuel using borosilicate and iron phosphate glasses

Cheong Won Lee *, Jae-Young Pyo, Jong Heo
Division of Advanced Nuclear Engineering and Department of Materials Science & Engineering,
Pohang University of Science and Technology (POSTECH), Pohang, Gyeongbuk, 37673, Republic of Korea
Corresponding author: schwarz@postech.ac.kr

1. Introduction

The fresh nuclear fuel made of enriched uranium oxide burned up for 3 years in the power plant. After burning, spent nuclear fuel is composed of 95 % of UO_2 and 5 % of fission products and transuranic elements. Their half-life is very long and it releases a large amount of radiation. It has to be isolated from environment. There are two methods being considered to dispose the spent nuclear fuel, direct disposal and reprocessing. However, no clear policies have been made in Korea. Approximately 760 tons of spent nuclear fuel are generated annually and stored in temporary repositories in power plants. These storages are expected to saturate in 2024 [1]. Therefore, a new technology need to be investigated for safe disposal of the spent fuel.

In this study, we propose the vitrification method as an alternative for the spent nuclear fuel disposal. Two different types of glass compositions, borosilicate and iron phosphate glasses, were investigated as potential host for the spent nuclear fuel. We assumed that spent fuel consisted of 100% UO_2 for simplicity.

2. Experimental procedure

2.1. Preparation of the glasses

The nominal compositions of the borosilicate glasses are given in Table I. Starting powders for these nominal compositions (SiO_2, H_3BO_3, Al_2O_3, Na_2CO_3, Li_2CO_3) were mixed and CeO_2 was used as a surrogate for UO_2. Batch mixtures were melted in alumina crucibles at 1500 °C for 1 hour and quenched by pouring melt between two brass molds in the air. BSA0 and BSA3 glasses were melted to form uniform black glasses. BSA5 glass contains white crystals that assumed to be CeO_2.

Table I: The nominal compositions of the borosilicate glasses (mol %)

	BSA0	BSA3	BSA5
SiO_2	55	55	55
B_2O_3	15	15	13
Na_2O	10	8	8
Li_2O	5	4	4
Al_2O_3	0	3	5
CeO_2	15	15	15
Total	100	100	100

The nominal compositions of the iron phosphate glasses are given in Table II. Starting powders for these nominal compositions ($NH_4H_2PO_4$, Fe_2O_3, H_3BO_3) were mixed and CeO_2 were used as a surrogate for UO_2. Batch mixtures were melted in alumina crucibles at 1300 °C for 1 hour and quenched by pouring melt between two brass molds in the air. IP10 glass was melted to form a uniform black glass. IP15 glass has white crystals of CeO_2.

Table II: The nominal compositions of the iron phosphate glasses (mol %) prepared

	IP10	IP15
P_2O_5	54	51
Fe_2O_3	27	25.5
B_2O_3	9	8.5
CeO_2	10	15
Total	100	100

2.1. Crystallization

Formation of unexpected and non-uniformly distributed crystals in the glass can decreases chemical durability because it absorbs glass network former, such as SiO_2, Al_2O_3 [2]. Crystallization of the glasses were analyzed by X-ray diffractometer (XRD, Rigaku D/MAX-2500/PC).

2.3. Chemical durability test

Chemical durability of the glasses (BSA0, BSA3 and IP 15) were evaluated by product consistency test (PCT) [3]. The glasses were crushed and sieved 75 to 150 μm). The glass powders were ultrasonically washed with deionized water and ethanol to remove fines and impurities. Samples (1.5 g) of powders were soaked in 15 mL of deionized water in a Teflon vessel and kept at 90 ± 2 °C for 7 days. The leachate was filtered using syringe with 0.45 μm filter. Concentration of elements in the leachate were analyzed using inductively coupled plasma mass spectroscopy (ICP-MS, NexION 350D, Perkin-Elmer SCIEX).

3. Results

3.1. Glass formation

Fig. 1 shows the XRD patterns of BSA0 and BSA3 glasses and Fig. 2 is the XRD pattern of IP10 glass. There is no evidence of crystal formation in the glasses.

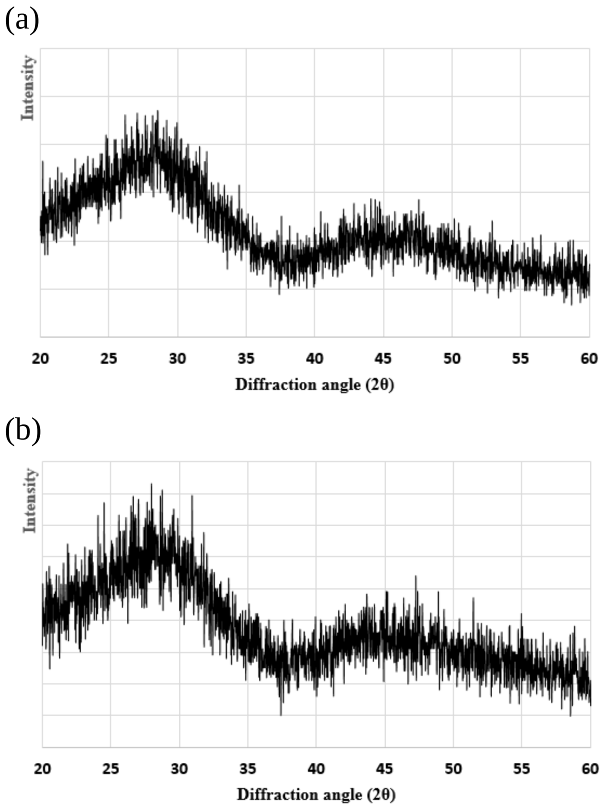

Fig. 1. XRD patterns of (a) BSA0 and (b) BSA 3 specimens

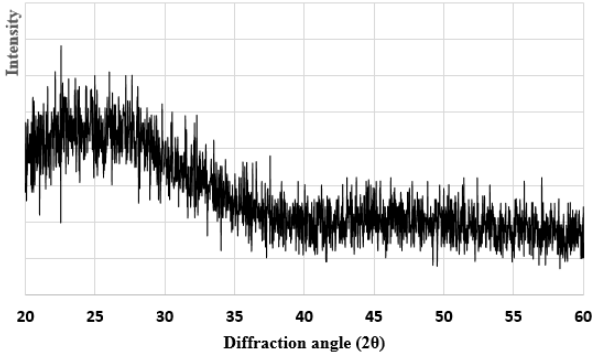

Fig. 2. XRD pattern of IP10 specimen

3.2. Chemical durability

Normalized elemental releases, r_i (g/m^2), were calculated by following formula:

$$r_i = \frac{C_i}{f_i(A/V)} \quad (1)$$

C_i is concentration of *i*th element in the leachate (ppm), f_i is the mass fraction of *i*th element in the glass (unitless), and A/V is ratio of the glass surface area to solution volume (m^{-1}). A/V value of the borosilicate glasses were 1810 m^{-1}, calculated from the glass density, 2.96 g/cm^3 and iron phosphate glass was 1700 m^{-1}, calculated from the glass density, 3.15 g/cm^3

Results of BSA0/BSA3 and IP10 are listed in Tables III and IV, respectively values of the normalized releases of Na^+ and Li^+ from BSA0 was higher than US regulation, < 2 g/m^2. It indicates that BSA0 glass is not suitable for the immobilization of spent fuel. On the other hand, the normalized releases of all elements from BSA3 and IP10 glasses were < 2 g/m^2. BSA3 and IP10 glasses appear to possess sufficient chemical durability as hosts.

Table III: Concentration C_i (ppm) and normalized elemental releases r (g/m^2) from BSA0 and BSA3 glasses

Elements	BSA0		BSA3	
	C_i (ppm)	r_i (g/m^2)	C_i (ppm)	r_i (g/m^2)
Si	151.40	0.42	20.35	0.057
B	241.97	1.36	4.25	0.050
Na	146.70	3.52	4.26	0.21
Li	57.43	3.17	2.71	0.057
Al	-	-	4.02	0.11
Ce	0.05	9.7×10^{-5}	0.16	3.2×10^{-4}

Table IV: Concentration C_i (ppm) and normalized elemental releases r (g/m^2) from IP10 glass

Elements	C_i (ppm)	r_i (g/m^2)
P	10.14	0.026
Fe	0.0023	6.4×10^{-6}
B	2.74	0.12
Ce	0.14	8.3×10^{-4}

4. Conclusion

Two different compositions of glasses were prepared as potential hosts for the spent fuel. Borosilicate glass containing 15 mol% of CeO_2 (BSA3) was melted at 1500 °C and iron phosphate glass containing 10 mol% of CeO_2 (IP10) was melted at 1300 °C. Normalized elemental releases of all elements from both glasses were < 0.21 g/m^2 that satisfy US regulations.

5. Acknowledgement

This research was supported by Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Science and ICT (NRF-2017M2B2B1072405).

REFERENCES

[1] B.H. Park, Assessment of spent nuclear fuel amounts to be managed based on disposal option in Republic of Korea, Annals of Nuclear Energy, Vol. 109, pp. 199-207, 2017

[2] P. Hrma, Crystallization during processing of nuclear waste glass, Journal of Non-crystalline Solids, Vol. 356, pp. 3019-3023, 2010

[3] Standard test methods for determining chemical durability of nuclear, hazardous, and mixed waste glasses and multiphase glass ceramics: The Product Consistency Test (PCT), C 1285-02, ASTM International, West Conshohocken, PA, 2008 <www.astm.org>.