Vitrification of the Spent Nuclear Fuel Using Iron Phosphate Glasses

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1. Introduction

Average amount of the spent nuclear fuel (SNF) being discharged from the nuclear power plants (NPP) in Korea reached 760 MTU per year. They have been stored in the spent fuel pool inside the NPP and the storages will be saturated by 2024 [1]. Direct disposal inside the deep geological repository has been considered for a safe disposal of SNF. We propose the direct vitrification using phosphate glass can reduce the amount of SNF that needs to be stored inside the repository.

2. Experimental procedure

2.1 Preparation of the glasses

The nominal compositions of the iron phosphate glasses are given in Table 1. Starting powders of $NH_4H_2PO_4$, Fe_2O_3 and Al_2O_3 were mixed and CeO_2 powders were used as a surrogate for UO_2. Batch mixtures were calcined in alumina crucibles at 220°C for 1 hour and melted at 1300°C for 1 hour. Melts were then quenched by pouring and pressing between two brass molds in the air to form glasses black in color.

Table 1. Nominal and analyzed composition of the iron phosphate glass prepared

Elements	Nominal composition (mol%)	Nominal composition (wt.%)	Analyzed composition (wt.%)
P_2O_5	60	57.30	55.45
Fe_2O_3	15	16.12	18.43
Al_2O_3	5	3.43	4.20
CeO_2 (UO_2)	20	23.16 (32.10)	21.92 (30.38)
Total	100.00	100.00	100.00

2.2 Properties analysis

X-ray fluorescence (XRF) spectroscopy was used to analyze the glass composition quantitatively. Density was measured at room temperature by the Archimedes method using deionized water as a medium. Glass transition temperature (T_g) and specific heat (C_p) were determined using a differential scanning calorimetry (DSC) at a heating rate of 10°C/min and thermal conductivity (k) was measured using laser flash method (LFA).

Chemical durability of the glasses were evaluated using the product consistency test (PCT) [2]. Glasses were crushed and sieved to 75 ~ 150 μm in size. Glass powders were then washed with deionized water and ethanol and 1.5 g of powders were soaked in 15 mL of deionized water inside a Teflon vessel and kept at 90 ± 2 °C for 7 days. The leachate was filtered using syringe with a 0.45 μm filter. Concentrations of elements in the leachate were analyzed using inductively coupled plasma mass spectrometer (ICP-MS).

Assessment of nuclear criticality safety and the stabilities of the glasses against the heat was performed using Monte Carlo N-particle 6 (MCNP 6) and computational fluid dynamics (CFD).

3. Results

3.1 General properties

Compositions of the glasses analyzed were similar to the nominal composition without any significant loss of individual components (Table 1). The glass contains 21.92wt.% of CeO_2, If UO_2 were used, concentration should be 30.38wt.%. Normalized elemental release of Ce after PCT was 2.3 x 10^{-4} g/m^2 and all other elements were < 0.2 g/m^2 significantly below US regulation of 2 g/m^2. The iron phosphate glasses have T_g = 540±2°C, C_p = 320.0±15 J/kg °C and k = 0.33±0.015 W/m K with a density of 3.15 g/cm^3

3.2 Evaluation of the vitrified SNF

Values of effective multiplication factor (k_{eff}) were 0.495 and 0.755 when the canisters were in the normal state and accident case, respectively. Both values were much lower than regulation of 0.95. It indicated that vitrified SNF has enough nuclear criticality safety for a disposal inside the repository.

Temperature of the buffer in repository for vitrified SNF reaches as high as 82.23°C when a spacing between canisters is 2m and distance between disposal tunnels is 10m. Area efficiency of the vitrified SNF disposal is almost 7 times higher than the case of the direct disposal [3]. In addition, the highest temperature expected at the center of the glass is 82.73°C and it is considerably below the glass transition temperature of the glass prepared (540°C).

4. Conclusion

Iron phosphate glasses containing CeO_2, as a surrogate for UO_2, were developed as a potential waste form for immobilization of SNF. The waste loading of CeO_2 (UO_2) in the glass was 21.92wt.% (30.38wt.%). Normalized elemental releases of all elements were < 0.2 g/m^2, well below the US regulations. k_{eff} (0.495 and 0.755) were below the criticality safety regulations (0.95). Glass transition temperature (540°C) of the iron phosphate glass is much higher than the peak temperature of the vitrified SNF (82.73°C) inside the repository, thus providing the thermal stability of the waste forms.

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