Estimation and Measurement of Centerline Temperature of the Glass Waste Form

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1. Introduction

High level glass waste forms are to be stored in the storage building before final disposal. The waste form storage vault should be operated using mechanically induced cooling system to remove the decay heat preventing overheat of the vault and devitrification of glass waste form in the canister [1,2]. However, even with the air cooling system in the storage vault, when the waste loading of heat generative nuclides are high, the centerline temperature of a glass waste form could exceed its glass transition temperature, leading to the increase of leaching rate of radioactive nuclides due to the devitrification of glass waste form [3]. Therefore, the estimation of centerline temperature of glass waste form for each waste stream is very essential in the period of storage. Also, the centerline temperature is important when a molten glass waste form is drained from vitrification equipment to a metal canister because the glass can be devitrified if the temperature profile from melting to room temperature is slow. Here, the centerline temperature of rare earth glass waste form generated from pyrochemical process has been estimated [4] and the temperature profile after drain process was measured.

2. Experimental

2.1 Fabrication of rare earth glass waste form

The rare earth glass waste form was fabricated using a SiO_2-Al_2O_3-B_2O_3 glass frit and Nd_2O_3/Gd_2O_3 as a surrogate waste material of rare earth fission products. For a vitrification of mixed oxides, the crucible was heated to 1,450°C with a heating rate of 6°C/min and maintained at 1,450°C for 4 hours. Thermal conductivity that is required in the centerline temperature calculation was measured using NETZSCH-LFA457 under an Ar atmosphere with a sample size of 12.7 mm in diameter and 2 mm in thickness.

2.2 Centerline temperature calculation

Centerline temperatures of waste forms for each transuranic elements (TRU) recovery ratio in the electrowinning process of the pyrochemical process were calculated using steady-state conduction equation (see equation (1) and (2)) in a long and solid cylinder with uniform heat generation and constant thermal conductivity. The heat generations of the waste form for each TRU recovery ratio were calculated using ORIGEN-S code.

$$\frac{1}{r}\frac{\partial}{\partial r}\left(kr\frac{\partial T}{\partial r}\right)+\frac{1}{r^2}\frac{\partial}{\partial \phi}\left(k\frac{\partial T}{\partial \phi}\right)+\frac{\partial}{\partial z}\left(k\frac{\partial T}{\partial z}\right)+\dot{q}=\rho c_p\frac{\partial T}{\partial t} \quad (1)$$

$$T_c = T(r=0) = \frac{-A+\sqrt{A^2+2B\left(AT_S+\frac{B}{2}{T_S}^2+\frac{\dot{q}}{16}D^2\right)}}{B} \quad (2)$$

2.3 Measurement of centerline temperature

Centerline temperature profile was measured by metal canister having R-type thermocouples that can measure temperature profiles at characteristic points. Test glass (general borosilicate glass) was poured into the metal canister (cylinder type, 300 mm diameter) and the centerline temperature profile from melting temperature to room temperature was obtained.

3. Results and Discussion

During the pyrochemical process, TRUs could be involved in the rare earth waste owing to the TRU recovery ratio (hereafter TRR) in the electrowinnng process. The nuclide formulation of each waste form after electrowinning process was calculated according to the TRR, where TRR-100 indicates 100% TRU recovery ratio. TRR-99, TRR-95, TRR-90, TRR-85, and TRR-80 indicate TRU recovery ratio of 99%, 95%, 90%, 85%, and 80%, respectively.

The centerline temperatures of each waste form having 0.3 m diameter were calculated according to the scheme in Fig. 1. In case of TRR-100, when there are no TRUs in the rare earth waste, the centerline temperature was 138.34^{o}C at the initial stage of storage. The centerline temperature was increased according to the decrease of TRU recovery ratio, however, even the TRR-80 case showed the centerline temperature of 201.62^{o}C, which is far below the glass transition temperature of the rare earth glass waste form (T_g=769.46^{o}C). Therefore, it is concluded that thermal stability of waste form in case of 0.3 m diameter is not affected by the TRU recovery ratio (even with the TRR-80 case) in the electrowinning process.

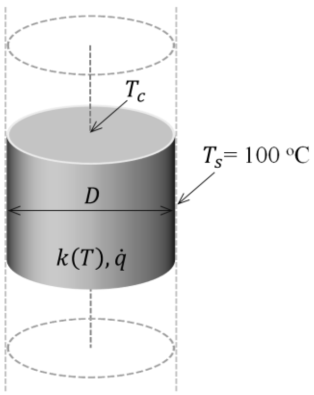

Fig. 1. Scheme of centerline temperature (Tc) calculation (Ts :surface temperature, k: thermal conductivity, q: heat generation, D: diameter).

Meanwhile, the centerline temperature after drain process was obtained using metal canister with 300 mm diameter. It was found that the molten glass poured at 1,350^{o}C was cooled to room temperature within 12 hours.

4. Conclusion

In order to determine thermal stability of the waste form immobilizing rare earth waste generated from the electrowinning process of the pyrochemical process, the centerline temperature of the rare earth glass waste form was calculated using steady-state conduction equation in a long and solid cylinder type waste form of constant thermal conductivity and uniform heat generation. To verify the effects of TRU content in the rare earth waste on the centerline temperature, the TRU recovery ratio (TRR) was varied from 80% to 100%. It was revealed that thermal stability of waste form in case of 0.3 m diameter was not affected by the TRU recovery ratio in the electrowinning process, meaning that the waste form size is thermally reasonable due to the low centerline temperature being far below the glass transition temperature of the rare earth glass waste form. Also, the devitrification tendency with current size waste form in the fabrication process can be assessed using temperature cooling profile obtained from current study.

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