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Physicochemical Property of Borosilicate Glass for Rare Earth Waste From the PyroGreen Process

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A study was conducted on the vitrification of the rare earth oxide waste generated from the PyroGreen process. The target rare earth waste consisted of eight elements: Nd, Ce, La, Pr, Sm, Y, Gd, and Eu. The waste loading of the rare earth waste in the developed borosilicate glass system was 20wt%. The fabricated glass, processed at 1,200°C, exhibited uniform and homogeneous surface without any crystallization and precipitation. The viscosity and electrical conductivity of the melted glass at 1,200°C were 7.2 poise and 1.1 $S{\cdot}cm^{-1}$, respectively, that were suitable for the operation of the vitrification facility. The calculated leaching index of Cs, Co, and Sr were 10.4, 10.6, and 9.8, respectively. The evaluated Product Consistency Test (PCT) normalized release of the glass indicated that the glass satisfied the requirements for the disposal acceptance criteria. Furthermore, the pristine, 90 days water immersed, 30 thermal cycled, and 10 MGy gamma ray irradiated glasses exhibited good compressive strength. The results indicated that the fabricated glass containing rare earth waste from the PyroGreen process was acceptable for the disposal in the repository, in terms of chemical durability and mechanical strength.

Keywords: Vitrification, Rare earth oxide, Characteristics, PCT, Borosilicate

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1. Introduction

The PyroGreen process, which transforms high level waste into intermediate and low level waste by improving the decontamination factor over 20 times of the Pyro Process, deservedly attracts attention for the safe recycling of spent nuclear fuel. The LiCl-KCl based eutectic waste molten salt, which contains rare earth waste, is generated from the PyroGreen process. In the PyroGreen process for spent fuel of light water reactor, Y, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, etc. are generated. Among the various rare earth elements, the generation ratio of 8 elements, Nd, Ce, La, Pr, Sm, Y, Gd, and Eu, are more than 99% [1, 2]. The composition of rare earth waste from the PyroGreen process is listed in Table 1. The majority of the rare earth nuclides, waste generated from the PyroGreen process, are separated and removed from the LiCl-KCl based eutectic waste salt as stable oxychloride and/or oxide form by repeated reaction with oxygen.

Table 1. Ratio of rare earth nuclides from the PyroRedSox waste in the PWR spent fuel

Nuclides	Composition (wt%)
Ce	22.73
Dy	0.01
Er	0.00
Eu	1.16
Gd	1.51
Ho	0.00
La	11.68
Nd	38.75
Pm	0.00
Pr	10.69
Sm	8.04
Tb	0.02
Tm	0.00
Y	5.40

Vitrification, converting radioactive waste into a glass or glass-like material, is being considered for treatment of various wastes. The vitrification offers various advantages: superior durability of resulting products, wide process window, flexibility of target waste, etc. [3]. These characteristics make vitrification the promising treatment system for radioactive waste from high-level to low-level radioactive wastes [4]. Vitrification of radioactive waste is a solidification process that combines solid and/or liquid waste with glass, resulting in a durable glass form. The radioactive waste is mixed with glass materials and heated to high temperature to produce molten glass. The stable glass that is produced incorporates radioactive elements and immobilizes them to prevent the hazardous materials reaching into the environment.

Borosilicate glass has been employed for the immobilization of radioactive waste in many studies [5, 6, 7]. Borosilicate glass was one of the first candidate glass composition for the immobilization of radioactive waste due to its physicochemical properties and well understood nature. In addition to the chemical durability, mechanical strength, and thermal stability, borosilicate glass offers sufficient flexibility with waste loadings and capability of incorporating most of the elements.

In this study, the vitrification of rare earth waste generated from the PyroGreen process was investigated using a borosilicate glass system. Also, the physicochemical properties of the developed glass were studied, in terms of the vitrification facility operation and satisfaction for waste acceptance criteria: chemical durability and mechanical strength.

2. Experimental

2.1 Fabrication of Glass

The glass was produced by melting raw chemicals in a Pt crucible at 1,200°C without a lid. The chemical composition

Table 2. Composition of waste, base glass, and candidate glass (unit: wt%)

Oxides	Raw chemical	Waste	Base glass	Candidate glass
Al_2O_3	Al_2O_3	-	3.75	3.00
B_2O_3	B_2O_3	-	14.29	11.43
Li_2O	Li_2CO_3	-	4.28	3.43
Na_2O	Na_2CO_3	-	31.43	25.14
SiO_2	SiO_2	-	46.25	37.00
CeO_2	CeO_2	13.35	-	2.67
Eu_2O_3	Eu_2O_3	1.30	-	0.26
Gd_2O_3	Gd_2O_3	1.65	-	0.33
La_2O_3	La_2O_3	13.10	-	2.62
Nd_2O_3	Nd_2O_3	43.20	-	8.64
Pr_2O_3	Pr_6O_{11}	11.95	-	2.39
Sm_2O_3	Sm_2O_3	8.90	-	1.78
Y_2O_3	Y_2O_3	6.55	-	1.31
Sum	-	100	100	100

for the glass is shown in Table 2. The raw chemicals were mixed sufficiently in the crucible for homogeneous composition and reaction. The crucible, containing chemicals, was placed in a $MoSi_2$ furnace for 1 hr. After 45 mins reaching 1,200°C, the melted glass was stirred with a quartz rod. It was then placed in the $MoSi_2$ furnace again. After thermal treatment for glass synthesis, the melted glass was poured into the graphite mold. In the case of the glasses for analysis of mechanical strength, the melted glass was poured into the 500°C pre-heated graphite mold and cooled slowly.

2.2 High Temperature Viscosity

The viscosity of the melted glass was calculated from measured torque at various rotation speeds using the spindle attached to a viscometer within the temperature range of 900°C to 1,400°C. The measured viscosity was interpolated with respect to temperature using the Vogel-Fulcher Tammann (VFT) equation (1):

$$\ln\eta = A + B/(T - T_0) \tag{1}$$

where, A, B, and T_0 are fitting parameters.

2.3 High Temperature Electrical Conductivity

The electrical conductivity of the melted glass was determined by measuring the resistance of the melted glass as a function of the frequency using a Pt/Rh electrode probe attached to an impedance analyzer. The electrical conductivity of the melted glass was measured in the temperature range of 900°C to 1,400°C. The measured electrical conductivity was interpolated with respect to temperature using the Arrhenius equation (2):

$$\ln\sigma = A + B/T \tag{2}$$

where, A and B are fitting parameters.

2.4 Chemical Durability

The chemical stability of the glass was studied by two different methods: (1) leaching index and (2) product consistency test (PCT).

2.4.1 Leaching Index (LI)

The ANS 16.1 leach test method was used to understand the LI of the fabricated glass [8, 9, 10]. The fabricated monolithic cylindrical glass was placed in the center of 5 L glass container. The glass was placed on the Teflon holder with a hole, which was designed to maximize the contact area of the glass with the leaching solution. The container was filled with the leaching solution, i.e. DI water. The volume of the leaching solution was 10 times the surface area of the glass. At each of the ten leaching intervals, the leaching solution was exchanged with the fresh leaching solution. The cumulative leaching times for these experiments were 2, 7, and 24 hrs and 2, 3, 4, 5, 19, 47, and 90 days.

2.4.2 Product Consistency Test (PCT)

The PCT was conducted following the method denoted in ASTM C 1285-97 [11]. The glass powder (75 um – 149 um) was washed twice with flowing DI water and ultrasonically washed twice with DI water to remove any undesirable particles, which adheres to the surface of the grinded glass. The washed glass was ultrasonically washed twice again with ethanol to dissolve any organicsand dried for 12 hrs at 90°C. The prepared samples were mixed with ASTM Type I DI water in a stainless steel vessel. The ratio of the sample surface and volume of the DI water was 2,000 m^{-1}. The vessel, which was filled with the sample and DI water, was stored at 40°C for 7, 38, 69, 100 days. The leachate was filtered after completion of the PCT. The 1 ml of filtered leachate was mixed with 20 ml of 1% HNO_3. The ion composition of the resulting leachate was analyzed using inductively coupled plasma atomic emission spectroscopy (ICP-AES) for B, Li, Na, and Si and inductively coupled plasma-mass spectrometry (ICP-MS) for Ce, Eu, Gd, La, Nd, Pr, Sm, and Y.

2.5 Mechanical Strength

The compressive strength of the monolithic cylindrical glass was conducted as described in the KS F2405 (standard test method for compressive strength of concrete). According to the test method, a glass was loaded into the testing equipment so that the axis of the glass was aligned with the center of the testing equipment. The loading was applied constantly at a stress rate of 0.25 ± 0.05 $MPa \cdot s^{-1}$ (35 ± 7 $psi \cdot s^{-1}$) without any shock. The desired rate of loading should be maintained during the expected loading phase. The loading was maintained until the load indicator started to decrease steadily, and the specimen displayed a well-defined fracture pattern. The loading was applied until the sample reaches the complete fracture status. The loading rate of the experiment was 0.25 MPa per second (29.4375 kN per minute) as described by the KS F2405. The dimension of monolithic cylindrical glass specimens used for the compressive strength was roughly φ 30 mm × 60 mm.

2.5.1 Water Immersion

The glass was immersed in DI water for 90 days to evaluate degradation after 90 days of water immersion. The procedure for the water immersion test basically followed the ANS 16.1 test procedure. After 90 days, the glass was taken out of the DI water, and the compressive strength was measured.

2.5.2 Thermal Cycle

For thermal cycling, the lower and upper temperatures were set at −40°C and 60°C, respectively. The thermal cycling test was applied to the fabricated glass in a temperature controlled environment chamber. After placing the glass in the chamber, the temperature of the chamber was raised from 20°C to 60°C during 1 hr. The temperature of the chamber was maintained at 60°C for 1 hr, and decreased to 20°C during 1 hr. The temperature of the chamber was held at 20°C for 1 hr, then decreased to −40°C from 20°C during the next 1 hr and then held at −40°C for 1 hr and increased to 20°C during the following 1 hr. The temperature stayed at 20°C for 1 hr, which completed a single thermal cycle within 8 hrs. 30 thermal cycles were followed on the glass within 10 days. After completing the thermal stress cycling, the glass was subjected to a compressive strength test.

2.5.3 Gamma Irradiation

The monolithic cylindrical glass was exposed to 10 MGy of gamma ray using a Cobalt-60 irradiator (MDS Nordion INC, Canada) in the Korea Atomic Energy Research Institute (KAERI, Korea). After irradiation of the gamma ray to the glass, the compressive strength test was performed.

2.6 Microstructure

The microstructure of the glass was studied using scanning electron microscope (SEM, JSM-5600). The SEM was operated at 15–20 kV to examine the physical and chemical homogeneity. The surface of glass was investigated and images were taken in backscatter-electron mode. Elemental maps were observed to understand the major element associations. The semi-quantitative analysis of the glass was followed to examine the uniformity and composition of the material.

3. Result and Discussion

The photo, SEM micrograph, and XRD pattern of the fabricated glass containing rare earth oxides are shown in Fig. 1(a). The glass composition was optimized using the GlassForm 1.1, considering waste loading, chemical durability, viscosity, and electrical conductivity [12]. The calculated optimized waste loading of the rare earth waste was 20wt% and detailed chemical composition of the glass is summarized in Table 2.

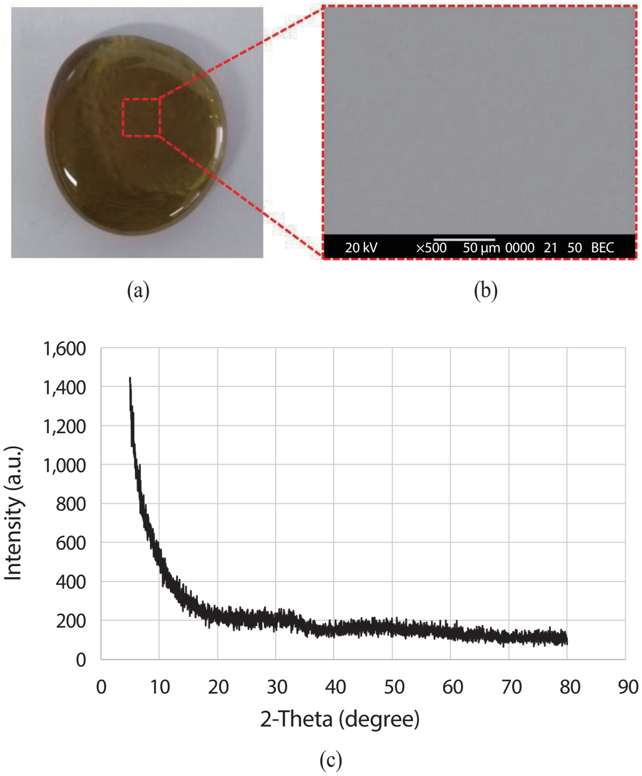


Fig. 1. (a) Photo, (b) SEM micrograph, and (c) XRD pattern of the fabricated glass containing rare earth oxides.

The SEM micrograph, shown in Fig. 1(b), of the fabricated glass indicates that the fabricated glass exhibited smooth surface, without any crystal and/or irregularities. The XRD pattern, shown in Fig. 1(c), indicates that the fabricated glass is amorphous. The element mapping of the fabricated glass was followed to understand distribution of the elements, including rare earth waste. Fig. 2 Indicates that both primary elements, which construct the glass matrix, and rare earth waste was uniformly distributed.

The physical properties of the developed glass were studied. The viscosity and electrical conductivity of the melted glass are the primary factors, when considering the appropriate operation of the vitrification facility. The reasonable range for viscosity is between 1 and 100 poise [13].

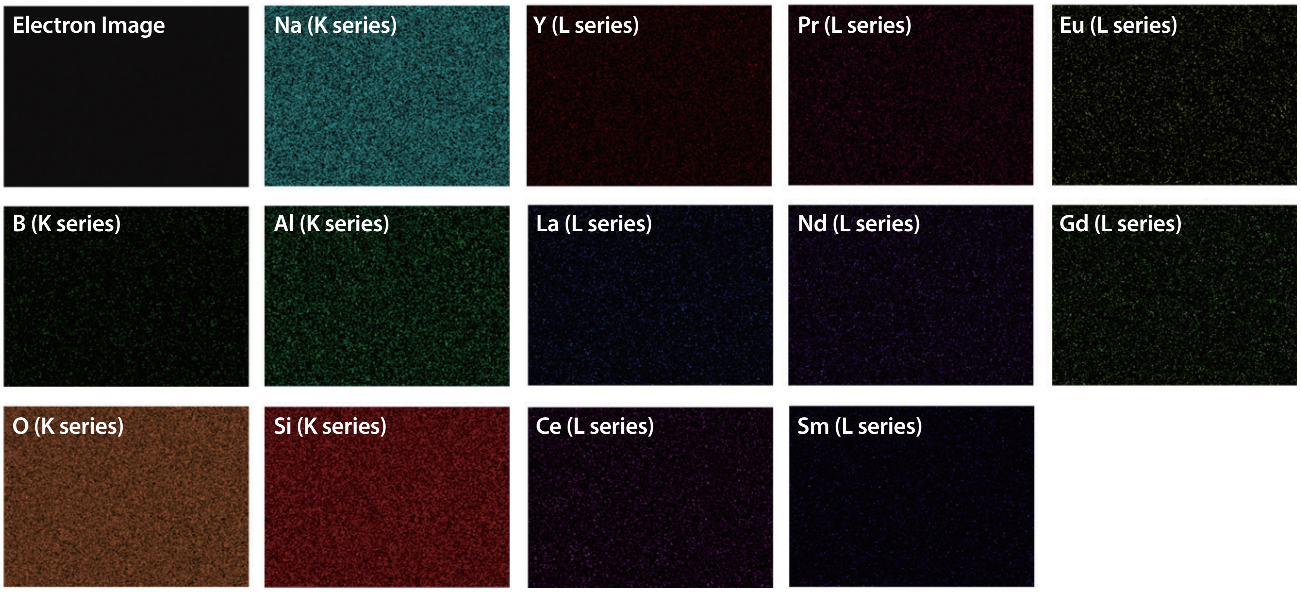
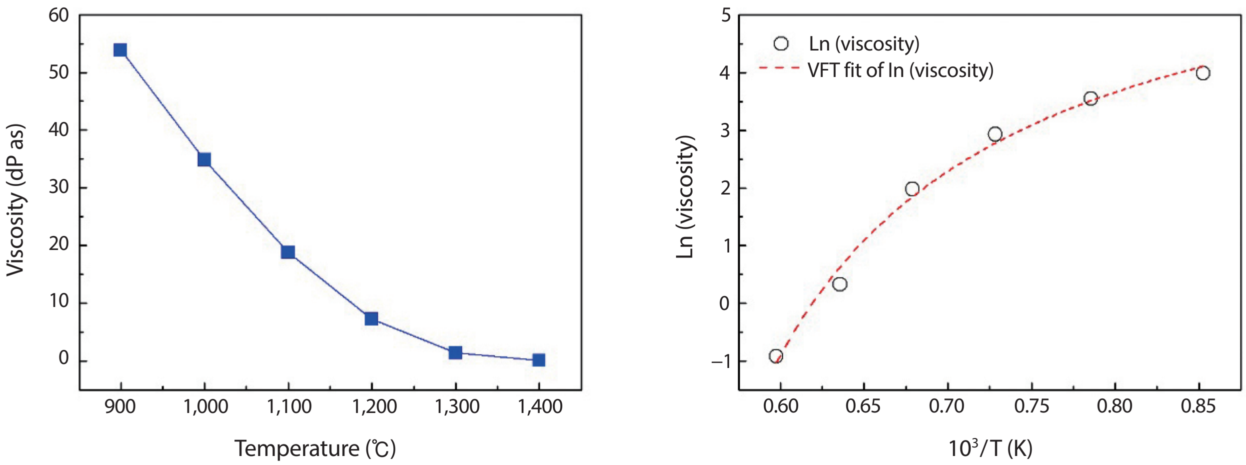


Fig. 2. Element x-ray mapping of the fabricated glass.



(a) (b)

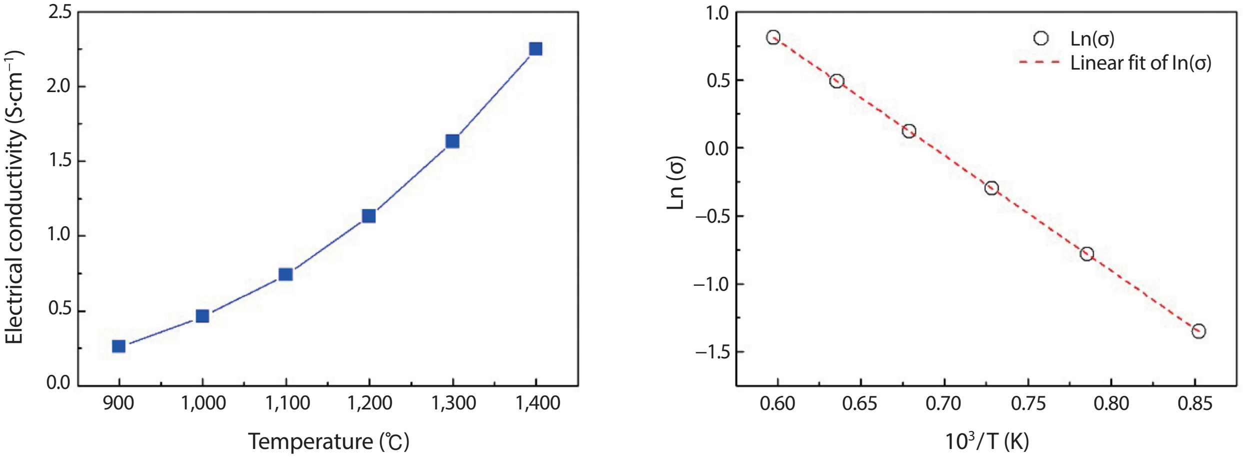
Fig. 3. Plot of (a) viscosity and (b) logarithm of viscosity of the glass as a function of temperature.

In the case of low viscosity, the melted glass is likely to negatively affect the durability of the vitrification facility. On the other hand, smooth pouring of the melted glass is not available for the higher viscosity, over 100 poise. Fig. 3 shows the viscosity of the melted glass. The observed viscosity of the melted glass at 1,200°C was 7.2 poise. The obtained VFT equation is

$$\log\eta = 2.81 + \frac{728}{T - 1,573}$$

where, T is temperature.

The activation energy for the viscosity, an indication of the energy required to cleave inorganic bonds within the glass network to initiate flow, of the glass was 159.3



(a)

(b)

Fig. 4. Plot of (a) electrical conductivity and (b) logarithm of electrical conductivity of the glass as a function of temperature.

Table 3. Chemical analysis of leaching solution from water immersion test after 2, 7, and 24 hrs and 2, 3, 4, 5, 19, 47, and 90 days (unit: $\mu g \cdot L^{-1}$).

Element	Time									
	2 hrs	7 hrs	24 hrs	2 days	3 days	4 days	5 days	19 days	47 days	90 days
Cs	N.D.*	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	3.91	8.76	14.4
Co	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	2.65	5.76	8.50
Sr	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	7.54	16.00	19.8

*N.D.: Not Detected

$kJ \cdot mol^{-1}$, which is similar to the previously reported results [14, 15, 16].

Fig. 4 indicates that the electrical conductivity of the melted glass increased, as the temperature increased. Generally, electrical conductivity over 0.1 $S \cdot cm^{-1}$ at the operation temperature is favorable for the reasonable process of the vitrification facility. The obtained electrical conductivity of the melted glass at 1,200°C was 1.1 $S \cdot cm^{-1}$. The activation energy for the electrical conductivity of the glass was 70.5 $kJ \cdot mol^{-1}$, which is similar to the previously reported results [14, 15, 16].

The measured viscosity and electrical conductivity were within the acceptable ranges of 1–100 poise and over 0.1 $S \cdot cm^{-1}$ at a temperature range of 1,000 to 1,300°C, respectively. It is reasonable to conclude that developed glass composition is suitable for the operation of the vitrification facility.

The chemical durability of the fabricated glass was studied using the LI test, which is based on ANS 16.1, and PCT. The ANS 16.1 protocol was adopted to evaluate the surface release and diffusivity of constituents in a monolithic vitrified form. Table 3 shows the chemical composition of the dissolved elements in the water. The spiking elements, which were Cs, Co, and Sr, were not detected until 120 hrs.

The effective diffusivity is calculated using equation (3)

$$D = \pi \left[\frac{(an/A_0)}{(\Delta t)_n} \right]^2 \left(\frac{V}{SA} \right)^2 T \tag{3}$$

$$T = [1/2(t_n^{1/2} + t_{n-1}^{1/2})]^2 \tag{4}$$

where D is effective diffusivity ($cm^2 \cdot s^{-1}$) during leach

Table 4. Chemical analysis of leachate from PCT after 7, 38, 69, 100, and 323 days at 40°C (unit: $\mu g \cdot L^{-1}$)

Element	Time (day)				
	7	38	69	100	323
Al	N.D.*	0.20	0.20	0.24	0.30
B	24.60	41.70	68.10	71.30	92.90
Li	9.49	14.90	13.80	14.60	28.90
Na	108.70	178.20	121.10	145.20	343.00
Si	33.20	48.10	68.80	71.80	82.90
Ce	N.D.	N.D.	N.D.	N.D.	N.D.
Eu	N.D.	N.D.	N.D.	N.D.	N.D.
Gd	N.D.	N.D.	N.D.	N.D.	N.D.
La	N.D.	N.D.	N.D.	N.D.	N.D.
Nd	N.D.	N.D.	N.D.	N.D.	N.D.
Pr	N.D.	N.D.	N.D.	N.D.	N.D.
Sm	N.D.	N.D.	N.D.	N.D.	N.D.
Y	N.D.	N.D.	N.D.	N.D.	N.D.

*N.D. : Not Detected

interval, V is volume of glass (cm^3), SA is geometric surface area of the glass (cm^2), T is the leaching time representing the mean time of leaching interval, an is quantity of an element released from the glass during the leaching interval, A_0 is total quantity of an element in the glass at the beginning of the first leaching interval, and Δt_n is duration of the nth leaching interval (s).

The LI is calculated using equation (5)

$$LI_n = -\log(D_n) \quad (5)$$

where LI_n is the leachability index and D_n is the effective diffusivity ($cm^2 \cdot s^{-1}$) for the element during the leach interval n.

The LI, derived from the ANS 16.1 test, evaluates diffusion controlled contaminant release, which were Cs, Co, and Sr in this experiment, with respect to the time. Since the LI indicates the specific chemical durability of the element of interests, the LI is generally used as an effective tool to assess the acceptability for the disposal in the repository [8, 9]. According to the waste acceptance criteria, the immobilizing waste form is considered an effective material when the LI is equal to or greater than 6. The calculated LI for Cs, Co, and Sr were 10.4, 10.6, and 9.8, respectively. The results indicate that the fabricated glass for rare earth waste is acceptable for the disposal in the repository, in terms of chemical durability.

The PCT of the fabricated glass was followed to understand the long term chemical durability of the fabricated glass. The detected chemical composition from the leachate is listed in Table 4. The rare earth elements were not found from the all leachates.

The PCT normalized release ($g \cdot m^{-2}$) is calculated from Table 4 using the equation (6).

$$NL_i = \frac{C_i \times V}{f_i \times SA} \quad (6)$$

where C_i is the concentration of element in the leachate ($um \cdot L^{-1}$) and f_i is the mass fraction of element in the glass, which was evaluated using the analyzed glass composition

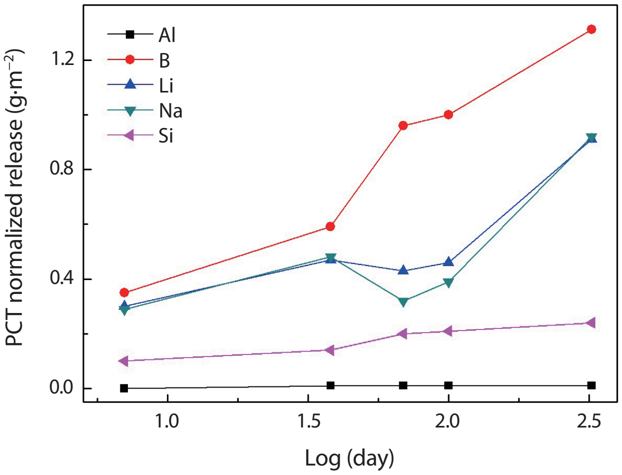


Fig. 5. PCT normalized release of the fabricated glass as a function of logarithm of time (day).

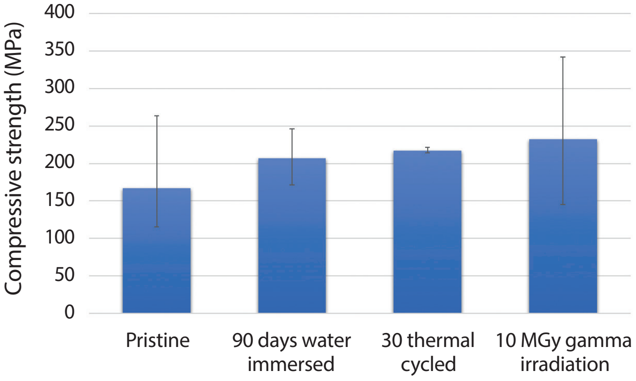
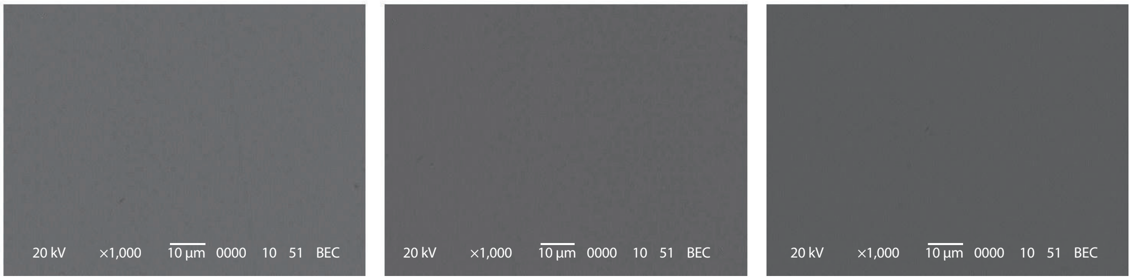


Fig. 6. Compressive strength of the monolithic cylindrical glasses: pristine, 90 days water immersion, 30 thermal cycles, and 10 MGy gamma ray irradiation (unit: MPa).



(a) (a) (a)

Fig. 7. SEM micrograph (×1,000) of the glasses (a) after 90 days water immersion, (b) 30 thermal cycle, and (c) 10 MGy gamma ray irradiation.

given in Table 2. The SA/V, which was 2,000 m^{-1} for this calculation, is the ratio of the sample surface area of vitrified form to volume of leachate (m^{-1}). The plot of PCT normalized release with respect to the logarithm of time (day) is shown in Fig. 5. The result indicates that all the PCT normalized release for Al, B, Li, Na, and Si are lower than 2 $g \cdot m^{-2}$ [8, 9]. Interestingly, the tendency of the PCT normalized release for the individual elements was not congruent. It means that the mechanism for the dissolution of the elements is not identical.

The mechanical strength of the vitrified form was studied using the KS F2405. In the case of destruction of the glass, the effective surface of the glass increases, compared to the designed condition. Since the higher effective surface increases the possibility to affect the environment, the mechanical strength of the vitrified form is considered as an important factor. In the case of disposal in the repository, the vitrified form is exposed to various environments, such as exposure to water, radiation, and temperature change. The mechanical strength of pristine, 90 days water immersed, 30 thermal cycled, and 10 MGy gamma ray irradiated glass were evaluated. The compressive strengths, which were measured 3 times, are evaluated by dividing the maximum loading on the glass during the measurement by the cross sectional area of the glass. The calculated compressive strength is summarized in Fig. 6. The compressive strength of the vitrified form shall be higher than 3.44 MPa (500 psi) when tested in accordance with ASTM 2010 from the US

NRC's technical position on waste form (US NRC 1991) [8, 9]. All the glasses, pristine, 90 days water immersed, 30 thermal cycled, and 10 MGy gamma ray irradiated, exhibited acceptable compressive strength. Fig. 7 shows the SEM micrograph of the glasses after 90 days water immersion, 30 thermal cycles, and 10 MGy gamma ray irradiation. The SEM analysis indicated that phase separation and/or crystallization of the glass were not observed under listed external stress. According to the study on mechanical strength and SEM analysis, it is reasonable to conclude that the glass for rare earth waste is free from the concern of water immersion, severe temperature change, and gamma ray irradiation lower than 10 MGy, in terms of mechanical strength.

The waste acceptance criteria (WAC) is suggested by disposal facility operator, Korea Radioactive Waste Agency (KORAD). The required properties of solidified forms, specified in WAC, are compressive strength, thermal cycle test, leaching test, etc. The experiment results indicate that the fabricated glass satisfies the characteristics requirements for waste solidified forms.

4. Conclusions

The glass composition for the vitrification of the rare earth oxide waste, containing Nd, Ce, La, Pr, Sm, Y, Gd, and Eu generated from the PyroGreen process, was studied. The optimized waste loading of the rare earth waste in the developed borosilicate glass system was 20wt%. The SEM image and element mapping results indicated that the fabricated glass, processed at 1,200°C, showed an uniform and homogeneous surface. The viscosity and electrical conductivity of the melted glass at 1,200°C were 7.2 poise and 1.1 $S\cdot cm^{-1}$, respectively. The calculated leaching index of Cs, Co, and Sr were 10.4, 10.6, and 9.8, respectively. The evaluated PCT normalized release of the glass were lower than 2 $g\cdot m^{-2}$. The pristine, 90 days water immersed, 30 thermal cycled, and 10 MGy gamma ray irradiated glasses exhibited an acceptable compressive strength. The results indicated that the fabricated glass containing rare earth waste from the PyroGreen process were acceptable for the operation of the vitrification facility and satisfied the waste acceptance criteria for the disposal repository, in terms of chemical durability and mechanical strength.

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