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Vitrification of liquid waste from nuclear power plants

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Abstract

Glass is an acceptable waste form to solidify the low-level waste from nuclear power plants (NPPs) because of the simplicity of processing and its unique ability to accept a wide variety of waste streams. Vitrification is being considered to solidify the high-boron-containing liquid waste generated from Korean NPPs. This study dealt with the development of a glass formulation to solidify the liquid waste. Studies were conducted in a borosilicate glass system. Crucible studies have been performed with surrogate waste. Several developed glass frits were evaluated to determine their suitability for vitrifying the liquid waste. The results indicated that the 20 wt% waste oxides loading required could not be obtained using these glass frits. Flyash produced from coal-burning electric power stations, whose major components are SiO_2 and Al_2O_3, is a desirable glass network former. Detailed product evaluations including waste loading, homogeneity, chemical durability and viscosity, etc., were carried out on selected formulations using flyash. Up to 30 wt% of the waste oxides was successfully solidified into the flyash after the addition of 5–10 wt% Na_2O at 1200°C. © 2001 Published by Elsevier Science B.V.

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1. Introduction

The treatment and disposal of the low-level radioactive wastes (LLW) have become a major operating expense to nuclear power plants (NPPs). Volume reduction of radioactive waste is one of the most important issues for the waste management with regard to the rapidly increasing cost of the waste storage and disposal. Vitrification is much attractive among the technologies in the volume reduction for LLW. Its advantages include the large volume reduction, good durability of waste form and the complete destruction of organic compounds [1]. Vitrification technology was initially recognized and developed to immobilize high-level radioactive wastes (HLW) in the mid-1950s. Recently, many researches have focused on the development of formulations for LLW and mixed waste [2–4]. The optimization of the glass formation has involved in the examination of the following process and product constraints: product control (chemical durability, homogeneity and thermal stability) and process control (viscosity, melting temperature and melt corrosivity). Very high waste loading and large overall volume reduction can be obtained for vitrified LLW. The large amount of reductions in the final waste form minimize the long-term storage costs, making vitrification cost-effective on a life-cycle basis. Another important objective of waste immobilization is to limit the release of radionuclides to the biosphere. In this respect, chemical durability of the waste glass form is of the greatest importance. Viscosity is also a very important property of glass from the viewpoint of processing.

Liquid waste generated from the operation of NPPs is commonly converted into solid state by cementation in many plants around world and it is solidified by paraffin in Korea. However, cement and paraffin solidification are not satisfactory from the viewpoints of

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economy (volume increasing) and ecology (poor durability). Korea, stemming from an economical and technical feasibility, is showing interest in using vitrification for treating LLW introduced from NPPs to achieve the large volume reduction and good durability of the final product. The characteristics and generation trends for each waste produced in NPPs were investigated. A vitrification pilot plant started the operation in summer 1999. The pilot plant will be the base for the development of the final objective, the establishing of an industrial scale vitrification installation in Korea [5–7]. Part of the project is the developing of the glass formulations for different waste streams. The objective of this work is to develop a glass formulation to vitrify the liquid waste with a processable and durable final product.

Application of vitrification technology to liquid waste should include the glass formulation optimization, melter system selection, process control parameters determination and off-gas system operation, etc. This study focused on the glass formulation development. Although many wastes are readily vitrified, the liquid waste from NPPs that contains high content of boron can offer several challenges. Several developed glass frits were fabricated for the pilot plant testing operation and it was uncertain if they could achieve high waste loadings for vitrifying the liquid waste. Therefore, preliminary tests were performed to examine the suitability of these frits for the vitrification of liquid waste. Flyash is a by-product of burning coal in electricity generating power stations. Physically, it is a very fine powdery material, predominantly silica, with particles almost totally spherical in shape. Flyash would pollute the environment and present a headache in electric power stations. Currently, only a small part of the flyash is utilized, mainly in cement industry. The remaining amounts of flyash are used for landfilling, which is an unsatisfactory solution from both the ecological and economic points of view. Due to the large amount of glass network formers in flyash, it is cheap and a promising additive for vitrification of the waste that contains lots of glass fluxes such as liquid waste. Therefore, using flyash to vitrify liquid waste is of high desire and environmental sound [8–10]. This paper provides the results of using flyash to solidify the liquid waste.

2. Experimental procedures

2.1. Compositions of liquid waste from Korean NPPs, some developed glass frits and flyash

The liquid waste is mainly generated from the boron recovery system and waste purification system in Korean NPPs. The annual volume of low-level liquid waste from the operation of NPPs is 80 m^3 (2 × 1000 MWe PWR) [5]. Studies have found that the amount of radionuclides in liquid waste from Korean NPPs is very small and such low-level components have no effect on glass properties. Therefore, non-radioactive waste was used in the present study. The composition of the simulated liquid waste from Korean NPPs is given in Table 1. Relatively, boron concentration is high and sodium level is low. Several glass frits have been investigated and selected for the pilot plant operation. Table 2 gives their compositions [11].

Significant amount of flyash is produced permanently as a by-product of the coal combustion in Korean electric power stations. The composition of flyash varies depending on the coals burned. However, SiO_2 and Al_2O_3 are the main components. Because blending the contents of various flyash is not feasible, we selected a typical flyash from a Korean coal-burning electric power station for our preliminary study (main composition is given in Table 3). The composition of this flyash provided a basis from which we developed a glass formulation for liquid waste. Once a glass formulation was

Table 1
Average composition of the liquid waste from Korean NPPs

Components	Concentration (ppm)
B	20016
Na	6177
Ca	449
Mg	776
Cl	808

Table 2
Composition of some glass frits (oxides basis, wt%)

Oxides	Glass frit name			
	NE	FN	PF	WG
SiO_2	54.00	45.50	56.00	72.00
B_2O_3	0	18.50	15.00	0
Na_2O	28.00	20.00	21.00	13.00
Al_2O_3	18.00	10.00	5.00	1.00
CaO	0	0	0	9.00
MgO	0	0	0	4.00
Fe_2O_3	0	5.00	3.00	0
K_2O	0	0	0	1.00

Table 3
The typical composition of flyash in terms of the oxide contents

Oxides	wt%	Oxides	wt%
SiO_2	63.49	K_2O	1.46
Al_2O_3	26.70	TiO_2	1.22
Fe_2O_3	3.09	MgO	0.88
CaO	1.98	Na_2O	0.74

developed, the composition of the flyash was optimized through a statistically designed approach. In addition, the flyash contains a small amount of heavy metals or hazardous elements (such as Pb, Zn and Cr). Such low levels of heavy metals would have no significant effect on the glass properties. Previous research has demonstrated that the durable waste glass matrix would result in the less release of the radionuclides and heavy metals. Therefore, the release behavior of the heavy metals was regardless during the development of the glass formulation. The relatively high levels of SiO_2 and Al_2O_3 indicated the feasibility of the flyash to vitrify liquid waste.

2.2. Glass manufacture

The surrogate waste was prepared to the desired composition. The glass samples were melted in a 150 ml platinum crucible in a furnace at 1150°C for 2 h. After pouring, the glass was annealed at 520°C for 2 h. Glass near the crucible interface was not used for testing, in case the glass melt had reacted with the crucible. Glass from the center was cored and cracked into suitable sized particles for leach testing. Glass prepared in a small crucible might have a little different property compared with that produced in the large (actual) melter due to the process control and atmosphere variation. The study on such difference will be performed in the pilot plant operation rather in this preliminary test.

2.3. Leaching test method

In order to rapidly determine the chemical durability of the glass, a new leaching test method (named as M-PCT), which was modified from the standard PCT leaching test method, has been developed [12]. The glass sample is particles in a size of 1.0–2.0 mm. The sample was washed with acetone to remove the fine particles before testing. The surface area of the samples was not measured because the particle size was too big for the determination methods like Blaine test or BET. Therefore, the surface area was calculated using the following equation [12,13]:

$$\mathrm{SA} = \frac{6M}{\rho\phi \times 0.89}, \tag{1}$$

where SA is the surface area in cm^2, M the mass of particle glass in g, ρ the density of glass sample in g/cm^3, ϕ the average diameter of particle in cm and 0.89 the factor to convert the results from spherical particles to glass.

The ratio of glass sample surface to leachant volume (SA/V) is 50 m^{-1}. The material of the leaching container is Teflon. Leaching temperature is 70°C; leachant is deionized water, and leaching duration is 7 days. The leachates were analyzed using inductively couple plasma spectrometry (ICP) for the concentration of several main glass components, such as Si, Na, B, Al, Ca and Fe. Then the total mass loss of glass (ML) and the normalized elemental mass loss (NL_i) were obtained. The pH value of leachate was measured with a pH-meter. The ML and NL_i were used to monitor the chemical durability of glass forms during glass formulation development.

The ML was calculated as follows:

$$\mathrm{ML} = (m_0 - m_1)/(\mathrm{SA}), \tag{2}$$

where ML is the total mass loss (g/m^2), m_0 the original unleached specimen mass (g), m_1 the specimen mass after leaching (g), SA is the sample surface area (m^2).

With regard to the elemental concentration in leachate, the normalized element mass loss NL_i was calculated as follows

$$\mathrm{NL}_i = C_i \times V/(\mathrm{SA} \times f_i), \tag{3}$$

where NL_i is the element i mass loss (g/m^2), C_i the concentration of element i in leachate (g/m^3), V the volume of leachant (m^3); f_i is the mass fraction of element i in unleached glass.

2.4. Viscosity measurement

It is difficult to measure the high-temperature viscosity of molten glass. Although the melt viscosity in our studies was important, accurate viscosity values were not necessary because we only needed to obtain the glasses that have low viscosity (<100 Pa s). In our studies, molten glass viscosity was determined by measuring the pouring time. The pouring times of different standard viscosity oils from the melter were measured. Then a calibration curve of viscosity vs. pouring time was obtained. The relative errors for the measurements were within 20%. The melt viscosities were determined quickly by this method and the results were in good agreement with those measured by a viscometer.

3. Acceptance criteria for glass formulation development

Development of the glass formulations for a given waste stream is basically a challenging work with respect to the constrained multivariant optimization. The main constraints are the economics, processability, and chemical durability. Waste loading is a major concern in terms of the cost. Minimum addition of non-waste additives will produce the largest waste loading and also largest volume reduction. Viable glass formulations must be capable of being processed through the particular type of system considered [3,14]. Viscosity of the melt plays an important role of processability. Generally, the most important requirement for the waste form is the good chemical durability, since one of the major objectives of waste solidification is to effectively isolate the radionuclides and other contaminants and prevent their release into the environment. Chemical durability and viscosity were under first consideration on determining the acceptability of a glass formulation in this study.

Since disposal of the final stabilized waste form invariably incurs a per-unit-volume disposal cost, the volume change upon vitrification is an important economic factor. The larger waste loading results in the higher volume reduction, which will save cost. However, it is limited by the waste glass chemical durability and processability as well as by the solubility of the individual inorganic waste components. The large amount of B_2O_3 would lead to a poor durability of final product [15]. Viscosity of the melt is mainly dependent on the glass composition and it limits the waste loading as well. In this study, the waste loading should not be less than 20 wt% (oxides) according to the former economical feasibility investigation.

The glass chemical durability is mainly influenced by the glass composition. In Korea, no specific requirements have yet been imposed regarding the chemical durability of radioactive waste glasses, with the only exception that the chemical durability of the glasses should be comparable to that of other developed waste glasses. The aim of this work was to make the glass as durable as possible under the given melting conditions. During the experiment on the development of M-PCT leaching test method, we have tested several well-known glasses and compared the results with those obtained other standard leaching tests (such as MCC-1, PCT). Those glasses that have total mass losses ~ 5.0 g/m^2 (by using M-PCT, deionized water, 70°C, 7 days) could be considered as durable glasses. Thus, a glass form was acceptable in this study when its $\mathrm{ML} \leqslant 5.0$ g/m^2.

Broadly speaking, processability means that when the glass is melted at a typical melter temperature (1050–1200°C), its viscosity must be low enough to pour. Melting temperature here is defined when the viscosity of melt equals ~ 50 Pa s. In order to reduce the volatility of some waste components, the melting temperature is usually lower than 1200°C. Research of HLW vitrification showed that lower melting temperature may lead to the poorer durability of glass form [16]. The viscosity of a glass melt, as a function of temperature, is the most important variable affecting the melting rate and pourability of the glass. The viscosity determines the rate of melting of the raw feed, the rate of glass bubble release (foaming and fining) and the rate of homogenization and thus, quality of the final glass product. The viscosity of molten glass would be required to be less than 100 Pa s at 1200°C in this study.

The durability of phase-separated glass is always poorer than that of homogeneous glasses. The knowledge of the homogeneity of waste glass was obviously related to the performance of the production process. The homogeneity of waste glasses could be evaluated using electron microprobe X-ray for microhomogeneity and using chemical analysis for composition homogeneity [17]. In this study, the optical microscope was used to determine the glass homogeneity. A well-designed and controlled vitrification process should produce homogeneous blocks.

4. Experimental results and discussion

4.1. Background of glass formulation adjustment

The glass structure is usually considered as a random network [16,18]. The elements are generally classified into three types:

1. Network forming atoms: such as Si, B, P, Ge;
2. Network modifiers (or glass fluxes): such as Na, K, Li, Ca, Mg;
3. Intermediates: such as Al, Fe, Zn, Ti, Mo, etc.

The concept of using glass as a host for radioactive waste is based upon the radionuclides entering into and becoming part of the random network [19,20].

There has been much research focused on the composition-durability relationships of glass. A great deal of information is available in simple systems. Unfortunately, results from one study cannot be compared quantitatively with those from other studies because of the absence of a uniform approach for testing or measuring durability [14,16]. Many results are qualitative. However, some general rules may still be applied, such as the following:

1. The components that form the strongest bonds in glasses result in the greatest improvement to glass and waste durability, whereas those that form the weakest bonds generally prove the greatest detriment to glass and waste glass durability;
2. Adding SiO_2, Al_2O_3, B_2O_3 and ZrO_2 may improve durability;
3. Adding alkali metal oxides may decrease the durability;
4. The mole ratio between Na_2O and $(Al_2O_3 + B_2O_3)$ of ~ 0.9 is suitable.

The glass formers are the major constituents of all waste glasses. If the inorganic oxides from the waste have insufficient glass formers to fall within an accepted glass formulation range, additional glass formers must be added through the process. Although the network modifiers (such as alkali metals) may decrease the durability, they are critical for controlling melted glass viscosity (and electrical conductivity, etc.) The addition of fluxes to the glass melt is controlled as increased flux composition generally lowers melting temperature, glass viscosity and leaching resistance. If the waste does not contain proper ratios of the materials for the formation of a glass, additives may be necessary. Because of its good chemical durability and inexpensiveness, SiO_2 is the most widely used glass forming oxides and therefore the major ingredient of the glasses. B_2O_3 is the second most used glass forming oxides. Although B_2O_3 is usually considered as a network former, however, borosilicate glasses composed of large amount of B_2O_3 are generally not chemically durable. The most effective glass modifier is Na_2O. The waste containing high level of B_2O_3 needs to be co-fed with other additives in order to achieve acceptable properties.

4.2. Test of some developed glass frits

Table 4 presents the experiment results of using the developed glass frits for vitrifying the liquid waste stream. The compositions of the frits are given in Table 2. Initial tests were centred on waste loadings around 20 wt%. The glass products were all homogeneous. It was ascertained that the viscosity of all melts was low enough for pouring except for glass NE20. High level of Na_2O and B_2O_3 in glasses resulted in low viscosity, but led to the poor chemical durability of final products. Only the NE20 has acceptable chemical durability. Increasing the waste loading to 30 wt% for NE decreased the chemical durability. The glass products of NE cracked easily even after having been annealed at 520°C. There is no quantitative method to measure the glass crack condition in our laboratory, but it is easy to find if there is a crack by visual inspection. The crack of glass after cooling is mainly due to its high thermal expansion coefficient. The borosilicate glasses usually have relatively higher thermal expansion coefficient than silicate or phosphate glasses. The increasing amounts of B_2O_3 and Al_2O_3 will increase the glass thermal expansion coefficient. The glass frits of FN, PF and WG could not satisfactorily immobilize the liquid waste. Too high level of B_2O_3 in FN and PF sharply decreased the chemical durability of the final products. Relatively high level of CaO in WG led to a poor durable product as well. The NE can incorporate <30 wt% of waste oxides, but the final products have high thermal expansion coefficient. Then we have no further studies on these glass frits due to the poor properties of their final products.

4.3. Flyash as an additive to vitrify liquid waste

Many glass forms for different waste streams have been developed [11,21]. Based on the composition of liquid waste and the past research, it is predicted that the ideal frit to solidify this liquid waste should have high content of $SiO_2 + Al_2O_3$. The flyash from the coal-burning electric power stations contains a large amount of SiO_2 and Al_2O_3, which would be a desirable network former. The flyash was tested at various waste loadings without other additives. Table 5 provides some typical experiment results. These glass forms were totally homogeneous. The minor component from flyash into glass decreased the thermal expansion coefficient of the final product and then improved its crack condition. The increasing of waste loading decreased the melt viscosity, but decreased the chemical durability as well. Because these glasses contained very low Na_2O, the pH value of leachate was relatively low. When the glass has the waste loading greater than 30 wt%, the chemical durability is very poor. FAB-1 glass has excellent chemical durability, but its melting temperature is higher than 1350°C. These results indicated that the waste stream mixed with flyash did not contain proper ratios of the materials for the formation of a quality glass and some additives might be required. The content of flyash in the glass should be less than 70 wt% in order to obtain a relatively low-melting temperature.

In order to decrease the melting temperature, Na_2O was added as the only additive. Glasses, include 60–65 wt% of flyash, 25–30 wt% of waste oxides and 5–10 wt% of Na_2O, have good chemical durability and suitable viscosity. Glass FAB-5 and FAB-6 in Table 5 have the acceptable properties. Due to the additive of Na_2O, the pH value of leachate increased. Fig. 1 provides the NL_i values of tested glasses. Na and B are much easily be released. The same tendency of ML and NL_i was observed for all glasses. Understanding of the leaching behavior

Table 4
Composition of some glasses with high waste loading

Glass sample No.	Type of glass frit				
	NE		FN	PF	WG
	NE20	NE30	FN20	PF20	WG20
Waste loading (wt%)	20	30	20	20	20
Homogeneous	Homogeneous	Homogeneous	Homogeneous	Homogeneous	Homogeneous
Viscosity[a]	~50	~50	~20	~20	~20
Cracked	Cracked	Cracked	Cracked	Cracked	No crack
ML (g/m^2)	2.61	7.36	61.27	43.26	14.86
Leachate pH	9.1	9.4	9.1	9.6	9.5

[a] Viscosity is measured at 1200°C, Pa s.

Table 5
Glass formulations and test results

Glass No.	Composition (wt%)			Viscosity[a] (Pa s)	ML (g/m^2)	pH
	Flyash	Waste oxides	Additive (Na_2O)			
FAB-1	70.00	30.00	0	100	2.86	8.6
FAB-2	60.00	40.00	0	~100	39.82	8.2
FAB-3	50.00	50.00	0	~30	214.2	7.8
FAB-4	60.00	30.00	10.00	~30	5.47	9.3
FAB-5	65.00	30.00	5.00	~50	4.07	8.9
FAB-6	65.00	25.00	10.00	~50	3.37	9.1
FAB-7	75.00	12.00	13.00	> 100	1.26	9.3

[a] Viscosity was measured at 1200°C.

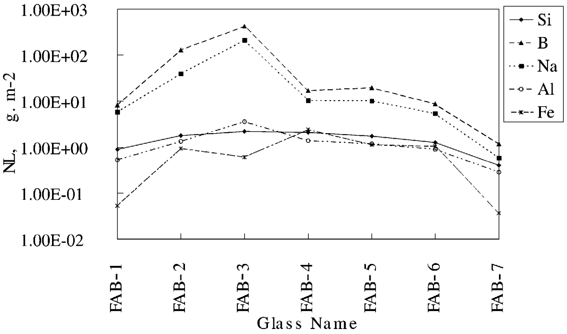

Fig. 1. Normalized elemental mass losses of glasses (M-PCT, 70°C, 7 days, deionized water).

was limited because of the insufficient leaching data in this study. To fully understand the chemical properties of a glass form, the leaching behavior and glass surface studies must be performed. Our future research will focus on the detailed characterization of glass surface and leaching behavior of glass FAB-5 and FAB-6.

5. Conclusion

Vitrification is a promising immobilization technology. It destroys organic compounds and then produces a high stable product. Vitrification is being considered for the treatment of the LLW NPPs in Korea. The glass formulation development includes the simultaneous optimization of the product control and process control. The waste loading, product chemical durability and melt viscosity are the main concerns in this study. Efforts were made to find a glass frit (or glass forming chemicals) that could be added to the liquid waste to produce a durable waste form with a high waste loading. The borosilicate glass has been worldwide used to stabilize the high-level waste. It also could be used for the liquid waste. The waste stream is utilized as a resource for the glass-making process for minimizing the need for purchased additives usually required for stabilization. The boron in the glass was totally taken from the waste stream. Preliminary glass formulation development involved in melting trials followed by visual homogeneity examination, viscosity measurement, crack condition examination and short-term leaching test. Several developed glass frits have been tested to solidify the liquid waste from Korean NPPs with the waste loading of 20 wt% (oxides). The results of these tests indicated that these glass frits could not satisfactorily vitrify the liquid waste stream. The frit NE could immobilize about 20 wt% of waste oxides to produce durable glass, but the final product cracked easily. Other frits could not produce durable glasses with 20 wt% of waste oxides. Results suggested that these frits mixing with the liquid waste did not have the appropriate proportion of compositions to make durable glasses. The flyash produced from coal-burning electric power plant contains large amount of SiO_2 and Al_2O_3, which are the best glass network former. It is desirable to mix the flyash with high-boron-containing waste to produce durable borosilicate glass. The melting experiments have shown that it is possible to vitrify liquid waste stream by using flyash as an additive. Adding Na_2O as network modifier especially as an agent to reduce viscosity is necessary. Formulations having 25–30 wt% waste oxides, 5–10 wt% Na_2O and 65–70 wt% flyash were found to be promising for incorporation of the liquid waste. The glasses showed good durability and suitable viscosity at 1200°C.

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