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Characterization of Glass Melts Containing Simulated Low and Intermediate Level Radioactive Waste

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ABSTRACT

In order to examine the process parameters for the vitrification of Low and Intermediate Level radioactive Waste (LILW) generated from nuclear power plants, measurements of several melt properties was performed for four selected glasses containing simulated waste. Electrical conductivity and viscosity were determined at temperatures ranging from 1123 to 1673°C. The temperature dependences of both properties in the molten state showed a similar behavior in which their values decrease as the temperature increases. The values of the electrical conductivity and viscosity at a temperature of 1423 K adopted in an induction cold crucible melter process were 0.27~0.42 S/cm and 9.8~42 dPas, respectively.

Key words* : *Vitrification, Low and intermediate level radioactive waste, Viscosity, Electrical conductivity, Induction cold crucible melter

1. Introduction

The vitrification of Low and Intermediate Level radioactive Waste (LILW) generated from nuclear power plants has been considered as the most promising disposal method of this waste from the viewpoint of having both a large reduction effect on waste volume and long-term stability. Recently, Nuclear Environment Technology Institute (NETEC), a division of Korea Hydro & Nuclear Power (KHNP) Co., has investigated and evaluated a vitrification process using an induction Cold Crucible Melter (CCM) in which electrical currents is directly induced into a glass melt from external high frequency inductors.[1)] Although the throughput of the CCM process is lower than that of the other processes, it is attractive as the melter has a long life and no replacement of parts is required during its lifetime.

Considering the CCM process, the viscosity and electrical conductivity of melts at a process temperature of nearly 1423 K are very important properties. If the melt viscosity is higher than 100 dPa·s, not only can homogenous melts not be obtained but the ability to drain the melts to a container is reduced. In contrast, if the melt viscosity is lower than 10 dPa·s, corrosion of contacted part with melts in the crucible takes place; thus its lifespan is reduced. Unsuitable electrical conductivity of melts can lead to trouble in an external high frequency generator. Therefore, two properties of melts containing LILW should be accurately controlled in the CCM process, as shown in Table 1.

In the present work, the viscosity and electrical conductivity of glass melts containing simulated LILW were determined in order to estimate the suitability of the developed glass formulations. The results were compared with constraints for the CCM process.

2. Experimental Procedure

The promising glass formulations for the vitrification of combustible dry active waste as high and low active ion exchange resin and zeolite have been developed by NETEC in Korea using linear glass property models.[2-4)] Table 2 shows the glass compositions containing the simulated waste. Their melt properties, such as viscosity and electrical conductivity in a molten state, were determined in a laboratory.

The viscometer (RotoVisco "RV2" Hakke Co., Germany) operated by the rotating cylinder technique as shown in

Table 1. Constraint of Several Properties of Melts for CCM Process

Glass property	Constraint
Processing temperature	1423 K
Electrical conductivity	0.1~1.0 S/cm
Viscosity	10~100 dPa·s

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Table 2. Candidate Glass Compositions for Vitrification of LILW

	AG8W1	AG8W2	IG1W2	DG-2
SiO_2	43.14	41.14	41.12	41.25
Al_2O_3	12.30	12.76	12.52	7.07
Alkali oxides	20.44	24.56	23.19	19.78
Alkaline earth oxides	6.94	2.33	5.66	14.4
Transition metal oxides	6.81	8.37	7.69	4.86
B_2O_3	9.97	10.71	7.46	11.29
Others	0.40	0.13	2.36	1.35

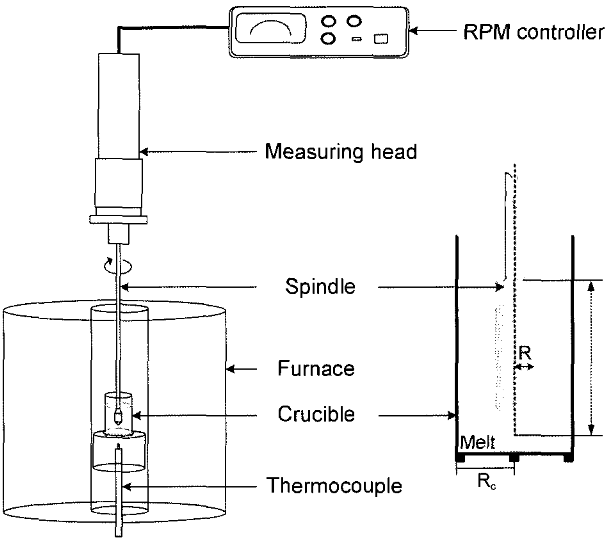

Fig. 1. Schematic description of the rotating cylinder viscometer.

Fig. 1, was used to determine viscosity of the melts. To conduct the viscosity measurements, a Pt-20%Rh spindle with 9 mm in diameter was immersed into the melt contained in a Pt-20%Rh crucible with 20 mm in diameter. The viscometer was calibrated using a standard glass of the German Society of Glass Technology (DGG) at a temperature range of 1273 to 1723 K. An apparatus constant was determined at the immersion depth of the spindle of 30 mm. The viscosity of each melt was measured within a_temperature range of 1223 to 1623 K. The measured data were interpolated to temperature using the Vogel-Fulcher-Tammann equation: $\log\eta = A + B/(T - T_0)$, where A, B, and T_0 are the fitting parameter.

For the electrical conductivity measurement of melt, a dipping electrode arrangement was applied. Fig. 2 shows the cell construction. The cell consists of an alumina crucible filled with melt, and two Pt electrodes with 1mm in diameter immersed at a 10 mm depth from the melt level. Electrical conductivity was measured between two electrodes separated by 15 mm. As a measuring bridge, a precision LCR meter (Hewlett Packard 4284A, USA) was utilized. The electrical conductivity measurements were performed using an alternating current of 1 kHz frequency in order to eliminate polarization and avoid the frequency dependence of the data.[5] The cell was calibrated directly at a temperature between 1273 and 1573 K. A binary alkali silicate glass system of which the conductivity is known was used as a melt for the calibration. After the calibration of the cell, the melt conductivity was measured in a temperature range of 1223 to 11623 K. The electrical conductivity data was interpolated to temperature using the Arrhenius equation: $\log\sigma = A/T + B$, where A and B are the fitting parameters.

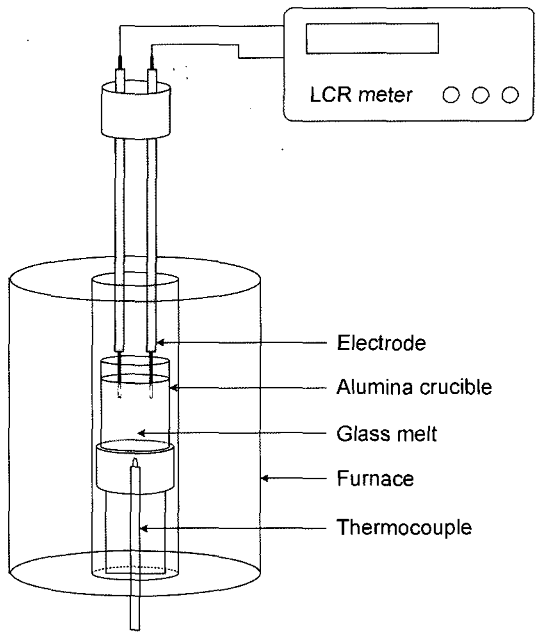

Fig. 2. Schematic description of the conductivity cell arrangement.

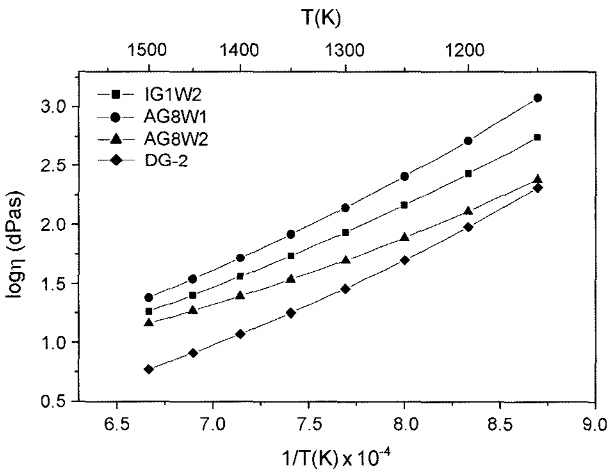

Fig. 3. Experimental plot of $\log\eta$ as a function of temperature.

3. Results and Discussion

The temperature dependence of the viscosity is shown in

Fig. 3. For all melts, the viscosity decreases when the temperature is increased from 1150 to 1500 K. The viscosities of all melts at a process temperature are 9.8 to 42 dPa · s, and met the processing constraint in Table 1. Table 4 shows the activation energy for the viscous flow of each glass melt at a temperature range between 1150 and 1500 K. The activation energy of the glasses varied from 108.77 to 152.64 kJ/mol. As the glass compositions used in this study are fairly complicated, it is difficult to interpret the relation between viscosity and composition. However, the outstanding viscous behavior of AG8W2 may be related to the CaO content of the alkaline earth oxide in the melt. According to the work of Gehlhoff and Thomas,[6] an addition of CaO to sodium silicate glass lowers the viscosity at a high temperature, and raises the viscosity at a low temperature. Therefore, the long-glass behavior of AG8W2 in comparison with the other melts is attributed to lowest content of CaO in the glass melts.

The temperature dependency of the electrical conductivity in each melt is shown in Fig. 4. The values of the measured

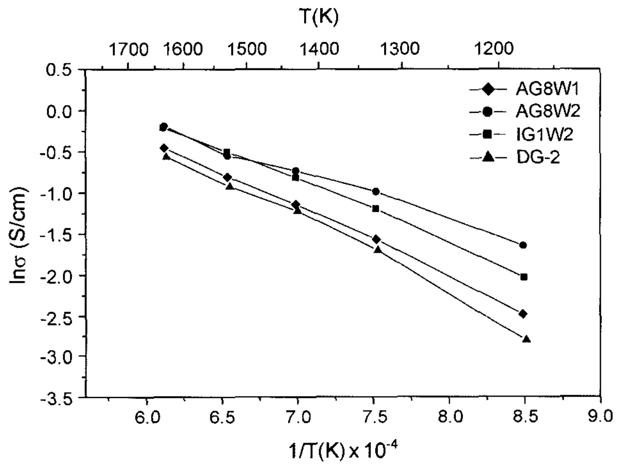

Fig. 4. Experimental plot of logσ as a function of temperature.

Table 3. Viscosity and Electrical Conductivity (or Resistivity) Values of Simulated Waste Melts at 1150°C

	AG8W1	AG8W2	IG1W2	DG-2
η (dPa · s)	42.78	21.57	30.45	9.87
σ (S/cm)	0.30	0.42	0.41	0.27
log η (in dPa · s)	1.63	1.33	1.48	0.99
log ρ (in Ωcm)	0.52	0.37	0.39	0.57
logη/logρ	3.1	3.6	3.8	1.7

Table 4. Activation Energy for Viscous Flow and Electrical Conduction at Measured Temperature Range

	AG8W1	AG8W2	IG1W2	DG-2
E_η (kJ/mol)	152.64	108.77	133.70	137.88
E_σ (kJ/mol)	70.46	49.02	63.39	77.73
E_η/E_σ	2.17	2.22	2.11	1.77

electrical conductivity varied from 0.27 to 0.42 S/cm at a processing temperature, and were acceptable for the CCM process. The activation energies for the electrical conductivity of each melt, as shown in Table 4, were calculated using the slope of the fitting line. Although there are various alkali oxides in melts, the high activation energies for the electrical conduction of AG8W1 and DG-2 can be explained in terms of the high viscosity at the process temperature and the mixed alkali effect,[7] respectively. In the case of DG-2, its mole fraction of K_2O/R_2O, 0.23 is closer to the mole fraction of the conductivity minima of nearly 0.5 compared to the others, 0.06, 0.02, and 0.04.

The work of Morey[8] demonstrated that the temperature dependence of the viscosity and electrical resistivity ($\rho = 1/\sigma$) of melts is often mutually dependent on the relationship, log$\eta \approx$ 3logρ. All melts except the DG-2 displayed similar relationship, which was determined to be 3.1 to 3.8 in logη/logρ_2 as shown in Table 3. However, judging from the viscosity and the resistivity of DG-2 it appears likely that there is no quantitative relationship. This was determined to be 1.7. In fact, since the mechanism between the viscous flow and the electrical conduction is similar in the material transport process, the relationship between the two properties based on their temperature dependence has been studied from a practical viewpoint.[9-13] According to the experimental results performed in various melt systems, plot of logç versus logñ showed a linear relationship, and it was concluded that the slope of this linearity denotes E_η/E_s,[14,15] whose value depends on the glass composition, as shown in Table 4.

4. Conclusions

The results of this study indicate that viscosity and electrical conductivity of simulated LILW melts fulfill the melt property constraints for an induction cold crucible melter system. The viscosity and electrical conductivity of all melts show a somewhat different behavior with temperature. This must be related to the glass composition. For these reasons, the direct determination of the glass properties in a molten state can contribute to either the development of a glass formulation or the modification of a developed one.

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