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Using Physical Properties of Molten Glass to Estimate Glass Composition

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Abstract

A vitrification process is under development in KEPRI for the treatment of low- and medium-level radioactive waste. Although the project is for developing and building Vitrification Pilot Plant in Korea, one of KEPRI's concerns is the quality control of the vitrified glass. This paper discusses a methodology for the estimation of glass composition by on-line measurement of molten glass properties, which could be applied to the plant for real-time quality control of the glass product. By remotely measuring viscosity and density of the molten glass, the glass characteristics such as composition can be estimated and eventually controlled. For this purpose, using the database of glass composition vs. physical properties in isothermal three-component system of SiO_2 - Na_2O - B_2O_3, *a software TERNARY has been developed which determines the glass composition by using two known physical properties (e.g. density and viscosity).*

I. Introduction

Vitrification, the process of converting materials into a glass or glass-like substance, is increasingly being considered the accepted solution for the treatment of highly radioactive materials because of the glass properties such as potential durability of the product and the flexibility of the vitrification process. Vitrification of low-and intermediate level liquid radioactive waste has recently been considered in vigorous projects such as KEPRI project of Korea.[1,2] A most critical unit in the vitrification process so far is the glass melter. The melter operates at high temperatures of around 1100 deg-C and it releases volatile radioactive gases. Under the severe working conditions, the conformation of glass composition specifications in a glass melter can only be verified by the withrawal of molten glass sample which is then analyzed in a separate laboratory. The sampling, sample transport, and chemical and physical analysis are cumbersome because of the complexity of the remotely operated equipment. Furthermore, it takes several days to complete the necessary tasks. The application of continuous monitoring systems practiced in the chemical process industry would be very difficult in this instance, because of large number of chemical elements contained in the glass and the extremely hostile environment of high radiation and of elevated temperature. However, the glass composition can be estimated with sufficient accuracy by remotely determining certain physical properties of the molten glass in the melter.

The goal of this study is to develop a real time glass quality control system and apply it to a Vitrification Pilot Plant. To accomplish this goal, there will be a few tasks including: (a) setup of the databases of physical properties vs. chemical composition of molten waste glass at a given temperature, (b) development of the methodology of waste glass composition estimation using the database, and (c) the development of bubble generation and detection device.

The bubble rising method, a methodology of measuring viscosity and density of liquid mixture under severe conditions, has been demonstrated.[3] The method will be improved for the bubble detection accuracy and then the physical properties will be simutaneously measured in real time and analyzed for chemical composition of the molten waste glass using the database of composition vs. physical properties.

II. Estimation of Glass Composition

Triangular coordinate is a helpful tool in representing physical properties of ternary compounds as well as more complex systems which can be simplified as pseudo-ternary compounds. The properties of borosilicate glasses are conveniently represented on a ternary phase diagram as a function of SiO_2, Na_2O, and B_2O_3 concentrations. Waste glass can also be represented on a triangular phase diagram on which, for example, silicon oxides, ash and other glass frit are the primary constituents.

Variation of specific physical properties with composition can be shown as constant value lines for a given temperature as illustrated in Figure 1 and available in the literature. By determining two physical properties at the same temperature, one can estimate the corresponding composition for the point where the two isopleths intersect. The composition point will be determined more precisely if the isopleths intersect at larger angles. It illustrates in Figure 1 that viscosity, electrical resistivity, and density are composition dependent, but to a decreasing extent, respectively. It is also apparent that viscosity/density and viscosity/electrical resistivity are suitable pairs, while electrical resistivity/density pair would produce a larger margin of error in the corresponding composition.

The computer program TERNARY was developed in order to estimate the borosilicate glass composition from two physical properties at a given temperature. Since it is easier to solve the equations corresponding to the points on a two-dimensional plot, all ternary diagrams are transferred into two-dimensional plots as shown in Figure 2. In this program, the isopleths of physical properties (e.g. viscosity and density) vs. composition at 1100 deg-C were used as a database. Polynomial equations of the isopleths are fitted by using the least square method. Polynomial equations which represent the pairs of the physical property values are derived and the roots of the equations which are obtained by the Newton-Raphson method correspond to specific glass compositions.(4)

The program was tested using data from the literature for sodium borosilicate glasses. The results shown in Table 1 indicate that the composition selected from the physical properties is faborably compared with the composition calculated with TERNARY.

III. On-line Measurement of Physical Properties

Many methods are used for liquid viscometry, but the elevated temperature and the corrosiveness of the molten glass have generally limited the choices to a falling ball, flowrate through an orifice and rotating spindle methods. None of the above methods would be suitable for continuous monitoring of molten glass, especially for remote operation in an intense radioactive field. A viscometry method was developed (3) which provides near-continuous monitoring on the viscosity of molten glass by the rise velocity of gas bubble. This method also allows measurement of the density. By ascending the bubbles in molten glass, both viscosity and density are determined in real-time and the data could be then read into the computer so that TERNARY locates the composition value on a ternary diagram of borosilicate glass.

New experimental setup shall include a bubble generator, two delivery tubes, a device which detects the arrival of the bubble at certain position below the air-liquid surface by measuring electrical resistivity across the passage of helium bubble, and a personal computer which controls the bubble generation, acquires data such as bubble rising velocity and transfers it into another useful form of data using the preset database. (i.e. chemical composition of waste glass in the melter).

IV. Conclusions

As a part of this study, the computer program TERNARY was developed to estimate the glass composition in a real-time basis at a given temperature, with two known values of physical properties, by integrating the database on physical properties vs. compopsition of molten borosilicate glass. The compositions of ternary and pseudo-ternary mixtures can be monitored through this TERNARY program when two physical properties are measued on-line and the data is transferred to a personal computer. In the case of molten glass, viscosity and density are the properties of choice in our study.

The on-line measurement techniques of physical properties will not be discussed here. This work will continue on the setup of bubble generation and detection device and the buildup of databases of physical properties vs. chemical composition of molten waste glass.

References

(1) Jong-Kil Park, et. al., Technical and Economical Assessment for Vitrification of Low-Level Radioactive Waste from Nuclear Power Plants in Korea, Proceedings in Waste Mangement 96, Tucson, Arizona, USA, Feb. 25-29, 1996.

(2) Ung-Kyung Chun, et. al., Waste Glass and Off-Gas Characteristics from Vitrification in a Planned LLW Treatment Facility in Korea, Proceedings in Waste Mangement 97, Tucson, Arizona, USA, Mar. 2-5, 1997.

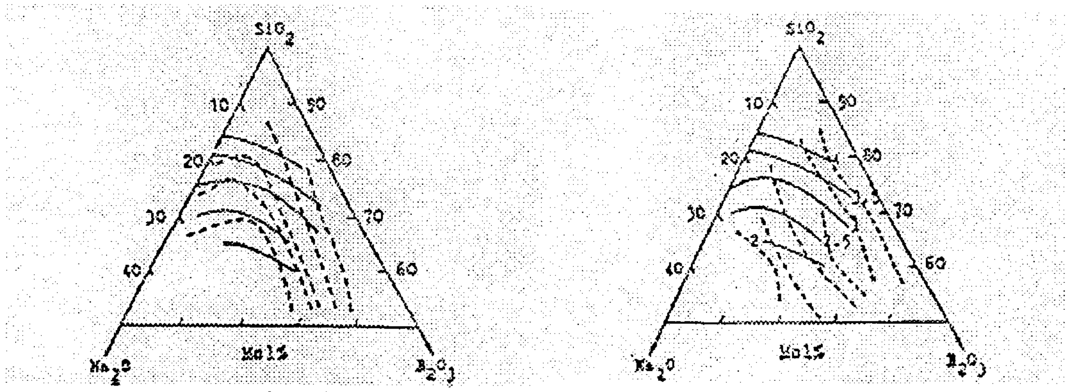
(3) A. Schneider, et. al. Final Report for Project E25-662, Georgia Institute of Technology, 1990

(4) Philip R. Bevington, Data Reduction and Error Analysis for the Physical Sciences, McGraw-Hill Co., New York, 1969.

Table 1. Test of Computer Program TERNARY

Composition Taken, mol%		Composition Calculated, mol%	
B_2O_3	SiO_2	B_2O_3	SiO_2
20.0	61.9	19.99	62.01
25.0	61.0	24.96	58.95
6.0	76.3	5.29	76.42
2.0	80.7	1.85	80.69
21.7	69.4	22.21	68.68
11.3	81.0	11.36	81.02

* the third component other than B_2O_3 and SiO_2 is Na_2O



——— log (viscosity) .... density

——— log (viscosity) .... resistivity

Figure 1. Constant Property Lines for the SiO_2 - Na_2O - B_2O_3 System at 1100 deg-C

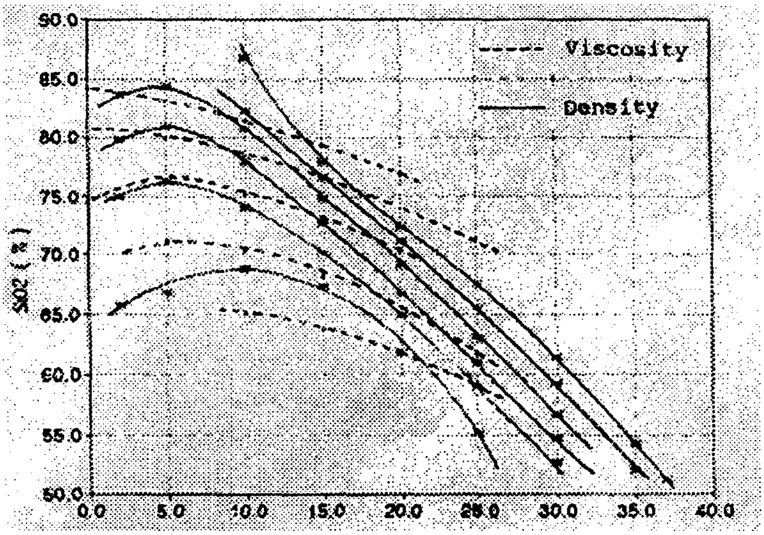


Figure 2. Two-dimensional Property Lines for Figure 1