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Preparation and Characterization of Nano porous Silica Glasses for Nuclear waste Immobilization

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Abstract

In order to nuclear waste immobilization, crystallization and phase separation of glass composition with addition of Mo_2O_3 have been investigated. At first, base glass composition prepared by melting of raw pure materials and above-mentioned additive in an alumina crucible at 1400^{O}C for 3 hrs. and casting in a metal mold. Mo_2O_3 was added in constant values of 0.5, 1.5, 2.5 and 3.5 mole%. XRD analysis carried out to ensure that no crystalline phases are formed. Then heat treatment was performed in temperature intervals of 620, 650, 680 and 710°C. SEM was used to phase separation study. The acid leaching process of phase separated glasses was performed. Porosity analyzes and measurement of specific surface area of BET were performed on acid leached samples. The results show that the highest porosity volume was related to the sample containing 1.5 Mole% Mo_2O_3 with two acid washing steps.

Keywords: Borosilicate glass, Vycor, Immobilization, Nuclear Waste.

Introduction

Recently, a variety of different materials such as ceramics, cements, metal matrices, and glasses have been considered to incorporate nuclear wastes. But the two best candidate materials are crystalline ceramics and glasses[1,3]. Fixation of radioactive wastes in glasses has been shown to be a viable technological alternative for effective management of nuclear wastes[2]. On the other hand, Borosilicate glasses have found many applications due to their low coefficient of thermal expansion and excellent chemical resistance. Phase separation is very important in this application of these glasses. Some oxides increase the tendency of phase separation in theses glasses; some oxides can also limit or inhibit phase separation. This work reports crystallization and phase separation of an alkali borosilicate glass composition in the $Na_2O–CaO–Al_2O_3–B_2O3–SiO_2$ system with addition of different amounts of Mo_2O_3 used as nuclear waste immobilization.

Experimental

Preparation of the materials

The mixture of pure raw materials such as Silica, Borax, Alumina, Magnesium Carbonate, and Calcium Carbonate, Molybdenum Oxide was melted in an alumina crucible in 1400° C for 3 hours and cast in a metal mold. Mo_2O_3 was added in values of (0.5-1.5-

2.5-3.5) mole% named 0.5M, 1.5M, 2.5M, and 3.5M and heat treatment was performed in specified intervals of 620, 650, 680, 710 ° C. XRD analysis, SEM and BET were used to characterize microstructure and porosity of the glass samples.

Results and discussion

The tendency of crystallization increased with increasing the amount of Mo_2O_3 due to decrease in viscosity of the glass melt. By increasing the amount of Mo_2O_3 in the mother glass, phase separation was achieved and crystallization occurred in samples containing molybdenum oxide. Acid leaching process of phase separated glasses was performed by HCl 10%. Porosity analyzes and measurement of specific surface area of BET were performed on acid leached samples. The results show that the highest porosity volume was related to the sample containing 2.5% Mo_2O_3 with two steps acid washing. Based on the specific surface results, the highest specific surface area was obtained for the sample containing 1.5% Mo_2O_3 with two acid washing steps (23.78 m^2 / g).

Tables

Table 1. Chemical Composition of the Specimens (Mole%)

Oxide/ Composition	**Base**	**0.5M**	**1.5M**	**2.5M**	**3.5M**
SiO_2	51.23	51.23	51.23	51.23	51.23
B_2O_3	14.81	14.81	14.81	14.81	14.81
Al_2O_3	4.94	4.94	4.94	4.94	4.94
MgO	7.41	7.41	7.41	7.41	7.41
CaO	14.2	14.2	14.2	14.2	14.2
K_2O	1.85	1.85	1.85	1.85	1.85
Na_2O	5.55	5.55	5.55	5.55	5.55
Mo_2O_3	--	0.5	1.5	2.5	3.5

Figures

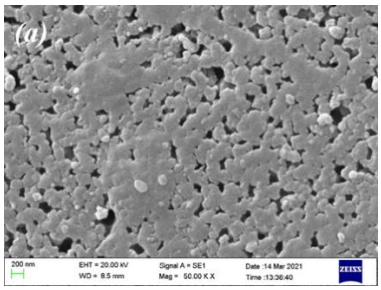

Figure 1.SEM image of phase separated of glass containing 1.5Mole% Mo_2O_3

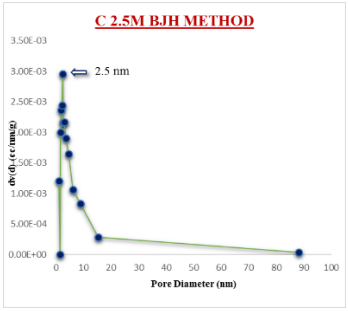

Figure 2: Porosity size distribution of C0.5M Sample. (Sample containing 0.5Mole% Mo_2O_3)

Conclusions

Borosilicate glasses are a solid in which a wide range of nuclear waste can be dissolved. Current research shows importance role of new containment additives with high chemical durability limiting release of radionuclides if the glass matrix is destroyed by aqueous tendency. Since phase separation and crystallization of the borosilicate glass composition cause inhomogeneity in these glasses, studying of these properties is most important and has high priority in this application. Studied in this work shows that with addition of different amounts of Mo_2O_3 changes crystallization and phase separation. Since getting to homogenized microstructure in glass compositions used for nuclear waste immobilization is important intensively, control and characterization of glass was prepared successfully via suitable time and temperature heat treatment of the samples.

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