Effect of Bi₂O₃ content on optical and radiation shielding properties of (Na₂O, K₂O)-SiO₂–CaO-Al₂O₃–B₂O₃ glass system

P. DARARUTANA, J. DUCHANEEPHET^a, S. PONGKRAPAN^b, N. SIRIKULRAT^a, P. WATHANAKUL^{b*}

The Royal Thai Army Chemical Department, Phaholyothin Road, Chatuchak, Bangkok 10900, Thailand; The Glass and Glass Products Research and Development Laboratory, Institute for Science and Technology Research and Development, Chiang Mai University; The Graduate School of Chiang Mai University Chiang Mai 50200, Thailand ^aDepartment of Physics, Faculty of Science, Chiang Mai University; The Glass and Glass Products Research and

Development Laboratory, Institute for Science and Technology Research and Development, Chiang Mai University, Thailand

^bDepartment of Earth Sciences, Faculty of Sciences, Kasetsart University, Bangkok, 10900 Thailand

Glasses of (Na_2O, K_2O) -SiO₂–CaO-Al₂O₃-B₂O₃ system with 5-40 wt% in concentration variations of Bi₂O₃ were fabricated in a laboratory scale using the conventional melting technique. Both refractive indices and densities of the glass samples were measured using a refractometer and the Archimedean buoyancy method, respectively, at 25°C. The gamma attenuation characteristics of the samples were studied using the photon energy of 662 keV from Cs-137. It was found that the values of refractive index and density of the samples increased and that their colors turned from colorless to yellow with increasing Bi₂O₃ contents. The relationship between refractive indices, densities, attenuation coefficients and Bi₂O₃ introducing contents were plotted and discussed. The results suggested that bismuth-bearing glass can be used as an environmentalfriendly substitute for lead- and/or barium-bearing glasses for industries besides their radiation-shielding property.

(Received July 23, 2008; accepted June 15, 2009)

Keywords: Bismuth-bearing glass, Optical property, Radiation shielding property, Local quartz sand

1. Introduction

Radiation shielding has become a subject of increasing interest among many applications in which radiation is being used, such as, the use of atomic energy and radioactive isotopes. Since glass is a solid and transparent material, there is a great effort being put into creating types that can protect users against small amounts of radiation without loss of transparency. These types of glasses have been developed to accomplish double tasks of allowing visibility while absorbing gamma radiation. An ideal shielding glass should have high absorption crosssection for radiation and, at the same time, irradiation effects on its mechanical and optical properties should be small. By comparison between other materials, lead glass is best known for gamma radiation shielding because of its high density and atomic number [1-3]. Most lead glasses exhibit high refractive indices (RI) of greater than 1.52 [4-6].

Due to toxicity of lead on human beings as well as on the environment, commercial lead-free glasses based on other heavy elements such as barium and bismuth offer comparable gamma radiation shielding.

The linear attenuation coefficient (μ) and mass attenuation coefficient (μ_m) of glasses containing oxides of B, Ba, Bi, Cd, and Pb were measured from 356 to 1332 keV. Comparison of their shielding properties with

those of standard shielding materials, such as, lead, lead glass, and concrete, has proven that they have potential applications in transparent radiation shielding [7-13]. Bismuth-bearing glasses with high refractive indices have rapidly been fabricated for desired structures and physical properties to be used in various applications, such as glass ceramics, optoelectronics, radiation-shielding, etc. [14-21].

Although the photon attenuation data are available in literatures, it is necessary to test these commercial materials experimentally for their radiation shielding efficiencies before putting into regular uses. In this study, the gamma radiation attenuation characteristics of lead-free glasses fabricated from local quartz sand and bismuth oxide have been investigated using photon from gamma radiation source with the energy range of 662 keV.

2. Experimental

Lead-free glass system of (Na_2O,K_2O) -Bi₂O₃-CaO-MgO-B₂O₃-SiO₂ was fabricated in a laboratory scale into eight 150-g batches consisting of 40 wt% dressed quartz sand from Tak – a northern province of Thailand, boric acid, sodium-, calcium-, and potassium carbonates each with different bismuth oxide (Bi₂O₃) concentrations from

5 to 40 wt%. The glass mixtures were melted in an electric furnace in normal atmosphere at 1150°C with 4-hr dwelling time. After complete melting, the molten glass was poured into a cylindrical steel mould of 3-cm diameter and of 1-cm thickness, and then cooled down to room temperature. The transparent and bubble-free cylindrical glass samples were obtained.

Prior to being analyzed surface of the glass samples were ground and polished to a mirror finish with a 0.3- μ m alumina paste. Refractive index (RI) of the prepared glass samples were determined using a sodium-light Reyner Duplex II refractometer with fluid $n_D \leq 1.79$ operating at room temperature. The density was measured by a Mettler Toledo AG104 analytical balance employing the Archimedean buoyancy method at 25°C.

The gamma attenuation characteristics of the prepared bismuth-bearing glasses have been studied using photons with energy range of 662 keV. The monoenergetic gamma radiation used in these measurements was from ~5 μ Ci of Cs-137 in a sealed source. The gamma radiation transmission measurements were done under a narrow beam counting geometry employing a Teledyne Brown Engineering NaI(Tl) detector with a Ludlum 2000 scaler. The lead free glass samples having various concentrations of Bi₂O₃ were interposed in the beam. The counts under the full energy absorption peak of the recorded photon spectrum were determined. From the transmitted intensity (I₀), for a density pand a thickness x of the sample, the linear attenuation coefficient (μ) and the mass attenuation coefficient (μ_m) are given by the following expressions:

$$\frac{\ln(I/I_0)}{x} = \mu \tag{1}$$

$$\frac{\mu}{\rho} = \mu_m \tag{2}$$

3. Results and discussion

As shown in Table 1, the measured densities and refractive indices of the glass samples ranged from 2.7048 to 2.5752 gcm⁻³, and from 1.520 to 1.665, respectively. The results reported were the experimental values of gamma attenuation coefficient determined in this study. The values of μ and μ_m for gamma-ray at 662 keV ranged from 0.2128 to 0.3132 cm⁻¹ and from 0.0787 to 0.0876 cm²g⁻¹, respectively.

By visual observations, colors of the glass samples were changed gradually from colorless to light yellow as Bi_2O_3 concentration was increased.

The results showed linear relationships between Bi_2O_3 concentration in the glass mixtures and refractive index, density, and attenuation coefficients of the resulted glass samples as shown in Fig. 1.

Table 2 showed values of μ and μ_m for gamma-ray at 662 keV of various glass samples. μ and μ_m of the cylindrical lead glass sample (0.25PbO.0.75B₂O₃) with density of 3.487±0.008 gcm⁻³ were 0.291 cm⁻¹ and 0.0836±0.0030 cm²g⁻¹, respectively¹⁰. Those of the lead glass (Corning 8362) with 30 wt% lead oxide and density of 3.270 gcm⁻³ were 0.28 cm⁻¹ and 0.0856 cm²g⁻¹, respectively¹. It has been previously reported that μ and μ_m for gamma-ray at 662 keV of the lead-free glass with 40 wt% BaCO₃ and density of 3.223 gcm⁻³ were 0.234 cm⁻¹ and 0.0726 cm²g⁻¹, respectively [22].

It was shown that the measured values of μ and μ_m for gamma-ray at 662 keV of the lead-free glass with 30 wt% Bi₂O₃ and density of 3.3621 gcm⁻³ were 0.2881 cm⁻¹ and 0.0857 cm²g⁻¹, respectively.

Bi ₂ O ₃ Concentration (wt%)	Properties at 25°C		Attenuation coefficient 662 keV	
	Density	RI	Linear	Mass
	(gcm^{-3})	(589nm)	(cm^{-1})	(cm^2g^{-1})
5	2.7048	1.520	0.2128	0.0787
10	2.8450	1.535	0.2287	0.0804
15	2.9784	1.555	0.2435	0.0818
20	3.1032	1.570	0.2610	0.0841
25	3.2468	1.600	0.2760	0.0850
30	3.3621	1.625	0.2881	0.0857
35	3.4077	1.650	0.2958	0.0868
40	3.5752	1.665	0.3132	0.0876

Table 1. Densities, refractive indices, and attenuation coefficients of the bismuth-bearing glass samples



concentration of bismuth oxide (wt%)

Fig. 1. Bi₂O₃ concentration vs. density, refractive index, linear attenuation coefficient and mass attenuation coefficient of the glass samples.

Table 2. Comparison of density and the attenuation coefficients at 662 keV of lead- and lead-free glass samples.

Tumo	Density	Attenuation coefficient		
rype	(gcm^{-3})	Linear (cm ⁻¹)	Mass (cm^2g^{-1})	
Lead glass [10]	3.487	0.291	0.0836	
Lead glass [1]	3.270	0.280	0.0856	
Lead free glass				
Ba-bearing	3.223	0.234	0.0726	
[22]	3.3621	0.2281	0.0857	
Bi-bearing				

4. Conclusions

Low-density glass samples will give rise to less attenuation than high-density ones. A wide variety of glasses could be produced both in terms of transparency and radiation shielding.

By comparing the properties, one can say the prepared bismuth-bearing glass with 30 wt% Bi_2O_3 is closely equivalent to lead glasses that have been used as standard radiation shielding glasses.

It can be concluded that the lead-free glasses prepared from local quartz sand and bismuth oxide as the main compositions are environmental friendly and can be used as gamma radiation shields. The utilization of local raw materials for producing radiation shielding glasses will help to lower the country's loss of trade equilibrium.

Acknowledgements

Part of the research has been funded by the Graduate School of Chiang Mai University. The Glass and Glass Products Research and Development Laboratory, Institute for Science and Technology Research and Development of Chiang Mai University supported the experiments in the glass fabrication.. The analytical balance and the refractometer were supported by the Department of Earth Sciences, Faculty of Science, Kasetsart University. The radiation measurements were supported by the Faculty of Technical of Medical Science, Chiang Mai University.

References

- W. S. Rohwell, Bull. PE-50 Corning Works, New York, 1958.
- [2] Premier Technology Inc., 2004.
- [3] Aura Lens Products Inc., 2005.
- [4] F. J. T. Malony, Doubleday & Company Inc., New York, 1968.
- [5] G. Holloway, Wykeham Publications Ltd., London, 1973.
- [6] H. G. Pfaender, Van Nostrand Reinhold Company, New York, 1983.
- [7] C. M. Davission, R. D. Evans, Rev. Mod. Phys. 24, 79 (1952).
- [8] B. Goswami, N. Chaudhuri, Phys. Rev. A 7, 1912 (1973).
- [9] M. J. Berger, J. M. Hubbell, NBSIR 87, 1987.
- [10] A. Khanna et al., Nuclear Instruments and Methods in Physics Research B 114, 217 (1996).
- [11] M. A. Abdel-Rahman et al., Nuclear Instruments and Methods in Physics Research A 447, 432 (2000).
- [12] R. Nathuram, Proc. of the 10th Int. Cong. of the International Radiation Protection Association, Hiroshima, P-6a-327, 2000.
- [13] H. Singh et al., Nuclear Science and Engineering, 142, 342 (2002).
- [14] K. Singh et al., Nuclear Instruments and Methods in Physics Research B **194**, 1 (2002).
- [15] H. Singh et al., Nuclear Instruments and Methods in Physics Research B 207, 257 (2003).
- [16] H. Singh et al., Physics and Chemistry of Glasses 44, 5 (2003).
- [17] I. I. Oprea et al., Optical Materials 26, 235 (2004).
- [18] S. Sindu et al., Materials Chemistry and Physics 90, 83 (2005).
- [19] F. H. El Batel, Nuclear Instruments and Methods in Physics Research B 254, 243 (2007).
- [20] F. H. El Batel et al., Physica B 391, 88 (2007).
- [21] I. Ardelean, D. Rusu, J. Optoelectron. Adv. Mater. 10, 66 (2008).
- [22] P. Dararutana et al., Abs. of 11th Int. Conf. on Radiation Shielding, Georgia, PSTR-16, 2008.

^{*}Corresponding author: pwathanakul@gmail.com