Materials and Design 80 (2015) 20–27

Contents lists available at ScienceDirect

Materials and Design

journal homepage: www.elsevier.com/locate/matdes

Physical properties and thermoluminescence of glasses designed for radiation dosimetry measurements

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ARTICLE INFO

Article history:
Received 20 March 2015
Revised 21 April 2015
Accepted 3 May 2015
Available online 4 May 2015

Keywords:
Glasses
Thermoluminescence
Elastic properties
Physical properties

ABSTRACT

Soda lime glasses doped with CeO_2, Nd_2O_3 and MnO_2 were prepared. Thermoluminescence (TL) properties, such as glow curves and linearity of TL response on irradiation dose were investigated. Results showed that the TL properties depended on the type and concentration of the dopants. Samples were selected to calculate energy trap depth parameters. To design materials for radiation dosimetry, physical properties, ion concentration, elastic properties and effective atomic numbers are important. Theoretical bond compression models were used to determine the elastic moduli for comparison with experimental values. Results show fair agreement between theoretical and experimental measurements. The high elastic moduli of the glass samples indicated high rigidity and stability of the glass matrix structure.

1. Introduction

Glass is composed of silicon dioxide (SiO_2), commonly found in nature as quartz. It comprises more than 10% by mass of the earth's crust. Many researchers around the world have investigated the properties of glass materials such as elastic moduli, optical, radiation and electrical for design and development in many applications including laboratory, industrial, medical, nuclear waste disposal, radiation dosimetry and communication technology [1–8]. Mechanical and physical properties of glasses are determined by the composition of the material and preparation techniques. As a general rule glasses are harder than metals, with a low coefficient of thermal expansion and thermal conductivity, good electrical insulation, resistance to chemical solutions (i.e. acids, alkalines, solvents and water) and can be used at high temperatures [9]. Therefore, glasses are a possible alternative to many materials. Glass is common, readily available, rigid and easy to handle.

Nuclear power applications have become extremely important in our modern society, with increasing use of radiotherapy and industrial radiographic testing. However, nuclear power plant radiation leakages are extremely dangerous, as witnessed by the Fukushima Dai-Ichi nuclear disaster in 2011, the largest since Chernobyl in 1986. Therefore, knowledge about radiation protection, radiation shielding and radiation measurement is vital. Recently, many authors have studied the application of soda lime glass (standard window glass) as an accidental dosimeter [10–15]. Soda lime glass is widely available and this makes it very attractive for use. The thermoluminescence (TL) technique is an interesting area for personnel monitoring and research to measure high doses incurred during various radiation process applications. New series materials are now being improved with more sensitivity and linearity of TL output in a wide range of doses. Some types of glasses, doped with small concentrations of rare-earth ions (ROs) and/or transition metal ions (TMOs) as activators have been found to be very effective [8,16,17]. Thermoluminescence glass materials doped with ROs and/or TMOs now play important roles in the field of radiation detection (both basic and applied research) including nuclear power generators, radiation oncology, personal and environmental monitoring and geological dating. The ROs and/or TMOs dopant impurities in the host glasses may cause alteration to its physical properties and TL features, depending on the dose amount [18]. Therefore, the effects of the dopants on the physical properties of the glasses are interesting.

Physical properties such as ion concentration, interatomic distance and elastic moduli are very informative regarding the structures of glasses; they are directly related to the interatomic potential. The depth of scientific information derived from the study of these physical properties is important in the design of materials and applications [19–21]. The effective atomic number is also an important parameter in the field of radiological properties of dosimeters [22–24]. This study investigated the effect of ROs and TMOs on the thermoluminescence and physical properties of glass samples, with a view to design available materials for radiation dosimetric measurements.

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http://dx.doi.org/10.1016/j.matdes.2015.05.002
0261-3069/

2. Experimental details

2.1. Sample preparations

Glass samples were prepared in rectangular shapes from $(RWG)_{(90)} - (Na_2O)_{(10)} - (R_mO_n)_{(x)}$, where x is 0.001, 0.010, 0.100 and 1.000 mol%. The R_mO_n is the oxide of CeO_2, Nd_2O_3 and MnO_2. Recycled window glass (RWG) was made from common window glass (soda lime glass) purchased in Ubon Ratchathani Province, Thailand. Wavelength Dispersive X-ray Fluorescence (WDXRF) techniques were used to determine the chemical composition of RWG [25]. The preparation of RWG from window glass involved a thorough cleaning and then it was ground to a powder. The oxides of Na_2O, CeO_2, Na_2O_3 and MnO_2 used in this work were of an analytical regent grade. The raw materials were weighed using an electronic balance with an accuracy of 0.1 mg, and mixed to 50 g samples. The starting materials were mixed carefully in ceramic crucibles and then melted in an electric furnace constructed at the Glass Technology Excellent Center (GTEC), Department of Physics, Faculty of Science, Ubon Ratchathani University, until homogeneity of the glass melt was attained. The melted glass was then poured into pre-heated stainless steel moulds and immediately annealed at 500 °C for 2 h before natural cooling to room temperature. The glass samples were then cut and polished for TL and ultrasonic measurements. Details of the chemical compositions of the glass samples are shown in Table 1.

2.2. Thermoluminescence measurements

All samples were annealed at 400 °C for 1 h and then 100 °C for 2 h (dual step technique). The X-ray machine (KELEX) was used (X-ray tube model MD1100). The X-ray tube has an inherent filtration by oil-insulated with vacuum system and added filtration by lead-shielded housing for reduce a low photon energy. The samples were then irradiated with X-ray photon energy (in air kerma) at 100 keV and 30 mA with a dose range of 0 to 14 mGy (with absorbed dose rate at 0.29 mGy s^{-1}) to determine the thermoluminescence responses and glow curves. The doses were measured by using pocket dosimeter (Aloka, Model: PDM-253). The irradiated glass samples were carried out TL signal immediately. TL light emitted from the glass samples were detected using a thermoluminescence detector (TLD) reader (Harshaw/Bicron Model 3500 Manual Reader). The glow curves were recorded from room temperature up to a maximum of 300 °C at a heating rate of 10 °C/s. The region of interest facility available in the TLD reader was used to evaluate the responses of different glow peaks resulting from the Computerised Glow Curve Deconvolution (CGCD) procedure. The CGCD data was carried out by take notes from the MS-DOS files to Excel program. Each datum point was obtained from an average of five detections. For investigation of trap depth parameters, general expressions for evaluating activation energy and trap depth were derived by Chen [26]. His method is useful for a broad range of energies ranging between 0.1 eV and 2.0 eV, and for values of the pre-exponential factors between $10^5\ s^{-1}$ and $10^{23}\ s^{-1}$. Chen's method does not make use of any iterative procedures and does not require knowledge of the kinetic order, which is found by using the symmetry factor ($\mu = \frac{T_2 - T_M}{T_2 - T_1}$) from the peak shape. The equations can be summarised up as Eq. (1):

$$E_\alpha = c_\alpha\left(\frac{KT_M^2}{\alpha}\right) - b_\alpha(2KT_M) \tag{1}$$

where K is the Boltzmann constant, T_M is the glow peak temperature, α is τ or δ ($\tau = T_M - T_1$ and $\delta = T_2 - T_M$), T_1 (rising end) and T_2 (falling end) are the temperatures at the half widths of the glow peak. The values of c_α and b_α are summarised in Eqs. (2) and (3).

$$c_\tau = 1.510 + 3.0(\mu - 0.42) \text{ and } b_\tau = 1.58 + 4.2(\mu - 0.42) \tag{2}$$

$$c_\delta = 0.976 + 7.3(\mu - 0.42) \text{ and } b_\delta = 0 \tag{3}$$

Frequency factor (S) is a constant characteristic of the electron trap, called the preexponential frequency factor or attempt-to-escape frequency. This parameter is proportional to the frequency of the collisions of the electron with the lattice phonons. The frequency factor can be calculated using the following Eq. (4):

$$\frac{\beta E}{KT_M^2} = (S)\exp\left(-\frac{E}{KT_M}\right)\left[1 + (b-1)\left(\frac{2KT_M}{E}\right)\right] \tag{4}$$

where β is the heating rate.

2.3. Effective atomic number calculations

The effective atomic number (Z_{eff}) for all types of materials, compounds and mixtures, can be written in terms of the fraction abundance as Eq. (5) [27]

$$Z_{eff} = \frac{\sum_i f_i A_i (\mu/\rho)_i}{\sum_j f_j \frac{A_j}{z_j}(\mu/\rho)_j} \tag{5}$$

where $f_i = \frac{n_i}{\sum_j n_j}$ is the fractional abundance of constituent element i, n_i is the total number of atoms and $\sum_j n_j$ is the total number of atoms present in the molecular formula, A_i and z_i are the atomic weight and atomic number, respectively. $(\mu/\rho)_i$ is the mass attenuation coefficient obtained from the WinXCom program [28].

2.4. Density and molar volume measurements

The density (ρ) of each sample was measured using Archimedes' principle with n-hexane as the immersion liquid. The experiments were repeated three times to obtain an accurate value of the density. The estimated error in these measurements was ±0.001 g cm^{-3}. The molar volume (V_a) was calculated for each glass sample from the expression; $V_a = \frac{M}{\rho}$, where M is the molecular weight of the glass, calculated according to the relation (6) [5].

$$M = \sum_i x_i M_i \tag{6}$$

where x_i is the mole fraction of the component oxide i and M_i is its molecular weight.

2.5. Ion concentration

The variation of ion concentration (N) can be calculated using the following equation [29]:

Table 1
Details of the chemical composition of the glass samples.

Glass sample	Compositions (mol%)				
	RWG	Na_2O	CeO_2	Nd_2O_3	MnO_2
S–Ce1	90	10	0.001	–	–
S–Ce2	90	10	0.010	–	–
S–Ce3	90	10	0.100	–	–
S–Ce4	90	10	1.000	–	–
S–Nd1	90	10	–	0.001	–
S–Nd2	90	10	–	0.010	–
S–Nd3	90	10	–	0.100	–
S–Nd4	90	10	–	1.000	–
S–Mn1	90	10	–	–	0.001
S–Mn2	90	10	–	–	0.010
S–Mn3	90	10	–	–	0.100
S–Mn4	90	10	–	–	1.000