



Influence of gamma radiation on the structural and optical properties of thulium-doped glass

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ABSTRACT

A few selected numbers of researches has been done to study the effect of gamma radiation in structural and optical properties of Tm-doped glass. Hence, the present study is done in order to overcome this problem. Tm-doped glass sample with composition of $\{[(\text{TeO}_2)_{0.7}(\text{B}_2\text{O}_3)_{0.3}]_{0.7}[\text{ZnO}]_{0.3}\}_{0.99}\{\text{Tm}_2\text{O}_3\}_{0.01}$ was prepared using melt-quenching technique. Density, FTIR, XRD and UV-VIS analyses were done before and after radiation with doses ranging from 10 until 35 kGy. With the increment of the radiation dose, the density and molar volume vary; indicating the variation in the compactness of the glass samples. FTIR analysis showed the changes in the absorption bands. The glasses still maintain its amorphous nature as confirmed by the XRD spectra. The direct and indirect optical band gaps showed a decreasing trend due to the breaking of bonds and existence of free electrons. Meanwhile, the other optical data have an increasing trend which was due to the presence of free electrons inside the glass system.

1. Introduction

There have numerous studies investigated tellurite glass due to its advantages such as low melting points, high refractive index, high dielectric constant, and good infrared transmission [1–5]. These benefits make it suitable to be used for non-linear optical devices, optical fiber amplifiers, electronic switching effects and many more. However, pure tellurite glass cannot be vitrified alone by traditional methods and needs additional substance in order to form glass [6]. Hence, many researchers start to combine another chemical into the glass system such as alkali, alkaline earth, transition metal oxides or other glass formers to improve its glass forming ability as well as other properties that are required for its appropriate applications [7]. In present study, thulium oxide is added into the glass system to enhance the optical properties of a material. According to Cho et al. [8], Tm-doped glasses have the ability to produce emission at 1.47 μm region [8]. In addition, Tian et al. [9] stated that Tm-doped glasses can provide mid-infrared emission which are very useful in military, remote sensing, eye-safe laser radar, atmosphere pollution monitoring and medical surgery [9].

The study on the structural and optical properties of a glass is very important in order to understand the nature of the material, internal structure as well as its electronic structure. This information can also be

a supporting material in the investigation of other characteristics of the glass such as elastic, thermal and electrical properties. According to Maheshvaran et al. [10], there are four types of structural units that exist in borotellurite glass that are BO_3 , BO_4 , TeO_3 and TeO_4 . They are found to be very sensitive as the other chemical oxides are added; these structural units will change from BO_3 into BO_4 and TeO_3 into TeO_4 or vice versa depending on the type of chemicals added and also the whole composition of the prepared glass. Besides, the introduction of gamma radiation into glass samples is also believed to cause changes in the structural network of the glass as well as enhancing its properties.

Ouis et al. [11] reported that there are three processes that occur during irradiation process; (i) Atomic displacement by momentum and energy transfer, (ii) Ionization and charge trapping, (iii) Radiolytic or photochemical effects. These processes are expected to operate partly or totally in the glass samples that led to the formation of induced defects and as a result causing the changes in its network structure. The response of the glass can be related to the radiation dosage rate and the sensitivity of the glass composition to gamma radiation. Shelby [12] stated that the irradiation to borate network caused the glass to be compact due to the breaking of bonds between BO_3 structural units, allowing tetrahedral BO_4 to be formed. This is also in agreement to a research that was done by Alailly et al. [13] which in turn contributed to

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the increment of density after the radiation process.

In addition, it is expected for some changes to occur in the FTIR and UV-VIS spectra after the radiation process. According to Iskandar et al. [14], network compaction and increment of the degree of disorder of the amorphous phase both occur inside the network structure. This is shown by the decrement of TeO_4 bi-pyramidal units and TeO_{3+1} (or distorted TeO_4) peaks due to the augmentation of gamma radiation dose. Besides, Xinjie et al. [15] who investigated the optical properties of rare-earth doped glass after radiation found that the optical band gap of the glass tend to decrease after being exposed to the gamma rays. This reduction shows that radiation causes the glass structure to change and it is most probably due to the generation of defects inside the glass system.

There are several researches have been done on rare-earth doped ZnO- B_2O_3 - TeO_2 glass [16–19]. However, none of them has studied the effect of gamma radiation on the structural and optical properties of this glass. Hence, the objective of this study is to provide an insight of structural and optical properties with the exposure of the glass samples to γ -radiation with dose ranging from 10 to 35 kGy. It is expected that the exposure would change the structure by breaking the bond and then rearranging it, depending on the dosage rate applied and producing a more compact network with higher density.

2. Methodology

Seven samples of glass with composition of $\{[(\text{TeO}_2)_{0.7}(\text{B}_2\text{O}_3)_{0.3}]_{0.7}[\text{ZnO}]_{0.3}\}_{0.99}\{\text{Tm}_2\text{O}_3\}_{0.01}$ was fabricated using conventional melt-quenching technique. Chemical oxides from Alfa Aesar were used with purity of 99.99%, 97.5%, 99.99% and 99.9% for TeO_2 , B_2O_3 , ZnO, and Tm_2O_3 respectively. All the chemicals were weighted using electronic balance to get the required amount. After that, the mixture was ground and stirred for 30 min to get a homogenous mixture. Then, it was put in the alumina crucible and pre-heated in the first furnace at 400 °C for 1 h to get rid of any water vapour. Next, it was melted in the second furnace at 900 °C for 1 h. The molten glass then was poured in pre-heated stainless steel mould and annealed at 400 °C for 2 h to reduce any residual thermal stresses in the sample. Lastly, the furnace was turned off and the sample was left overnight in the furnace to let it cool down to room temperature. The sample was cut using Isomet Buehler low speed saw machine according to the thickness required for the testing purposes which in this case was 2 mm thickness. After that, both sides of the sample were polished in order to obtain smooth and parallel surfaces. Then, the sample was sent for radiation process in MINTec, SINAGAMA, Malaysian Nuclear Agency (MINT), Bangi, Selangor. The dose ranges that were used varied from 10 to 35 kGy. The density of each glass sample was measured using Electronic Densimeter MD-300S with distilled water as the immersion fluid. Some of the samples were also ground into powder form for structural testing that were FTIR and XRD testing. These testing were done using Perkin Elmer FTIR Spectrometer and Philips X-ray Diffractometer X'PERT PRO PW304 with Cu $K\alpha_1$ radiation. Meanwhile, the optical testing was done by using UV-1650PC Shimadzu UV-Vis Spectrophotometer. All the parameters were investigated before and after the radiation process.

3. Results and discussion

3.1. Colour changes

After the radiation process, it can be observed that the colour of the samples changed from pale yellow into yellowish-brown as shown in Figs. 1 and 2. This modification is attributed to the formation of defects in the glass network that known as colour centres [20]. There are two types of colour centres that can be produced, which are electron and hole colour centres. Electron colour centres are created when electrons from the gamma rays replace an atom or ion in the glass network while



Fig. 1. Glass sample before radiation.



Fig. 2. Glass sample after radiation.

hole colour centres are formed when electrons are ejected from an atom and creating holes. These colour centres are able to absorb only specific energies or specific incident light wavelengths and thus make them responsible for the changes in the absorption spectrum of the glass. Some colour centres also show fluorescence, which in turn can function as laser materials [21]. The evidence for colour centres formation is shown in the transmission spectra as presented later in Fig. 7.

3.2. FTIR spectra

In FTIR testing, infrared radiation is passed through the sample and some of the radiation will be absorbed while some of it will be transmitted. The radiation will be absorbed when the energy is equal to the energy of molecular vibrations inside the sample. As a result, the resulting infrared spectrum will produce a molecular fingerprint for the sample that enable the identification of molecular bonds that exist inside the material. In this study, the FTIR testing is done in the region of 280–2000 cm^{-1} for the prepared glass before and after radiation. However, the fingerprint region for all glass samples can be seen only in the range of 560–1500 cm^{-1} as shown in Fig. 3. The spectra can be divided into three regions that are at 620 cm^{-1} , 850–1100 cm^{-1} and

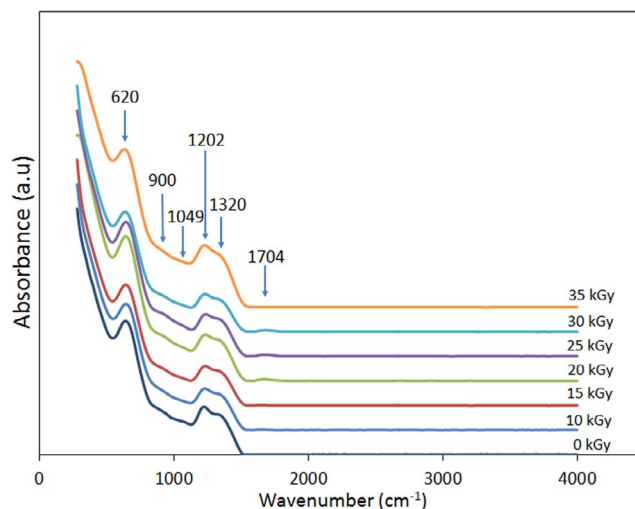


Fig. 3. FTIR spectra of thulium doped zinc borotellurite glass before and after radiation.

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