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Thermoluminescence properties of barium titanate prepared by solid-state reaction

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Abstract

Barium titanate samples prepared through solid-state reaction have been analysed by X-ray diffractometry and thermoluminescence (TL) methods to ascertain their capability for usage as high gamma dose dosimeter for industrial and scientific applications. Exposure to gamma radiation doses from ^{60}Co was varied between 1 and 9 kGy. The glow curves obtained showed emission peak temperatures lying between 480 and 530 K and were found to be dose dependent. The emission peak temperature for each dose was found to shift towards high temperature as the dose increased indicating a non-first order kinetics, which could be attributed to the creation of deep level defects, caused by the irradiation. The reproducibility tests carried out on the samples showed that the sample preparation as well as the TL response of the barium titanate under gamma irradiation is reproducible and under acceptable limit. The dose response curve showed a good linearity over the dose range examined. The fading characteristics indicated that the TL spectra alter with storage time after irradiation and this has been attributed to the creation of traps of different energy level by irradiation and clustering effect leading to tunneling.

Keywords: Gamma radiation; Thermoluminescence; Dosimetry; Barium titanate; Glow curves; Radiation detector

1. Introduction

Accurate monitoring of highly energetic radiations is desirable in industrial, medical and food radiation processing where an accurate measurement of the absorbed dose can be critical for an effective outcome. In such cases, solid-state thermoluminescence (TL) dosimetry method is indispensable. It can even be extended to environmental monitoring in such places as irradiation facilities and nuclear power stations [1]. This has probably informed the growing interest in synthesising affordable dosimeter through simple, non-tedious production route. Over the years quite a number of materials have been prepared and extensively studied for dosimetry applications notable among which are, sodalime glass [2], coloured glasses [3], thin film of Li_x–$Co_{(1-x)}$–O deposited on sodalime glass [4] and zircon [5,6].

Barium titanate ($BaTiO_3$), on the other hand has not been extensively studied for dosimetry applications despite the fact that the crystal possesses excellent ferroelectric and piezoelectric features that have found usefulness in the manufacture of microelectronic devices [7] apart from being employed as optical sensors due to its large electro-optic coefficients and high photorefractive sensitivity. In addition, it is showing a good promise as a material for holographic storage and cheap diode lasers [8].

In terms of other methods of characterisation, extensive study has been carried out on defect structure in barium titanate. In general, defects in perovskite-type crystals are of two different types: one that maintains stoichiometry (Schottky) and the other that changes stoichiometry. Since this study concerns undoped barium titanate, the defect structures are those that maintains stoichiometry and of particular importance to this study are the works of Serrano et al. [9] and Pinto et al. [10] who have employed quantum-chemical method based on molecular orbital

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theory and density functional theory in the local density approximation to study F-centres as well as oxygen vacancy defects in tetragonal and cubic $BaTiO_3$ in terms of atomic movement, relaxation energy and local energy level within the band gap for the F-centres on Ba–O and Ti–O_2 layers, respectively.

The aim of the present work is therefore to use high dose gamma irradiation to study the thermoluminescence glow curve characteristics of $BaTiO_3$ prepared through solid-state reaction.

2. Experimental procedure

Barium titanate pellet was prepared from a mixture of barium oxide (BaO) and 99.9% pure titanium dioxide (TiO_2), obtained from L. Merck AG, Darmstadt, Germany, and May & Baker, England, respectively. The oxides were mixed together in 1:1 weight ratio and ground with a mortar and pestle to obtain a homogeneous powder mixture. This is followed by pre-sintering in a Carbolite furnace for 4 h at a standing temperature of 600 °C and then reground before pelletising with a pressure of 1.13×10^8 N m^{-2} to produce a disk shape of 13 mm diameter and 1 mm thickness. Since it is well known that BaO can easily absorb moisture and CO_2, the time between weighing and grinding before presintering has been kept to not more than 5 min in order to have a reduced hygroscopic effect. Neither solvent nor binder was added. The pellets were sintered at a constant temperature of 1100 °C for 8 h and furnace cooled to room temperature. The samples prepared under the conditions described above have been characterised by X-ray diffraction analyses for structural as well as phase compositional determination.

Irradiation of the barium titanate ($BaTiO_3$) samples after sintering were performed at room temperature using a ^{60}Co gamma cell irradiator (Gammacell 220 produced by the Atomic Energy of Canada) available at the Centre for Energy Research & Development (CERD), Obafemi Awolowo University, Ile-Ife, Nigeria. The dose rate during irradiation was 1135 Gy/h. Some of the samples were annealed before irradiation using a well-calibrated annealing machine at the Federal Radiation Protection Service, University of Ibadan, Ibadan, Nigeria. The annealing temperature used was fixed at 500 °C for 5 h and then furnace cooled to eliminate thermal stress. The TL glow curves of the irradiated samples were recorded in a nitrogen atmosphere using the Victoreen TL reader (model 2800M) available at the CERD. To be able to observe the influence of preheating on the nature of the glow curves, the preheating was effected at 50 and 100 °C while the heating rates employed were 8 and 10 °C/s, respectively. All the TL response in this study has been normalized by weight of the individual sample.

3. Results and discussion

3.1. X-ray diffraction

Typical X-ray diffraction pattern of the samples prepared under the conditions described above is shown in Fig. 1, which confirms the crystallinity of the specimens. The figure also indicates the appearance of the first six diffraction peaks, which corresponds to the tetragonal phase of $BaTiO_3$ and matches well

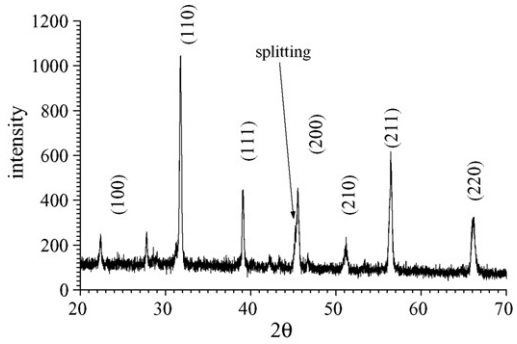

Fig. 1. X-ray diffraction pattern of $BaTiO_3$ prepared by sintering the mixture of BaO and TiO_2 at 1100 °C for 8 h.

with JCPDS data and the studies of other workers [11–13]. The lattice constants "*a*" and "*c*" have been calculated to be 3.95 and 4.48Å respectively with *c*/*a* ratio of 1.13. Comparing our result with the *c*/*a* ratio prepared through other set of chemicals [14], which is in the range of 1.04, our calculated *c*/*a* ratio of 1.13 shows that the material is strongly tetragonal.

3.2. Glow curve

The TL glow curves of $BaTiO_3$ samples annealed at a constant temperature of 500 °C and irradiated to different test doses are shown in Fig. 2. It is evident from the figure that the structure of the glow curves is different for each test dose. Below 9 kGy, all the curves show broad peaks and as the dose increases, the peaks become better defined. In addition, the peak temperature lies between 480 and 530 K. This change in peak temperature as the dose increases is probably an indication that the activation energies of the traps form a continuous distribution rather than a spectrum of discreet values. This kind of broad-peak behaviour has been observed in amorphous materials [12], but the XRD analyses confirmed the crystallinity of the specimen in this study. Therefore, either the preheating or the irradiation of the specimen must be responsible for this peak temperature shift.

Now let us consider the influence of preheating. The effect of preheating on irradiated specimen is to deplete the defect states at shallow levels with the consequence that the glow curve shifts towards high temperature as the preheating temperature

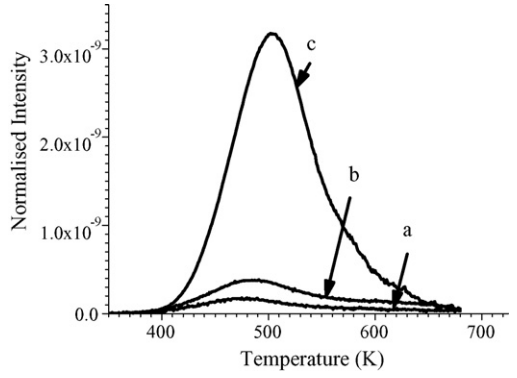

Fig. 2. Glow curve of $BaTiO_3$ irradiated with different test doses: (a) 2.27 kGy, (b) 4.54 kGy, and (c) 9.08 kGy, respectively.