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Comparative spectral and shielding studies of binary borate glasses with the heavy metal oxides SrO, CdO, BaO, PbO or Bi_2O_3 before and after gamma irradiation

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ABSTRACT

This work aims to compare the experimental shielding behavior of some prepared binary borate glasses with SrO, CdO, BaO, PbO or Bi_2O_3 towards successive gamma irradiation by investigating their combined optical and FTIR spectral measurements before and after gamma irradiation. Optical spectra of all the samples reveal strong UV absorption which is related to the presence of unavoidable trace iron impurities (Fe^{3+} ions) contaminated within the raw materials which were used for the preparation of the studied glasses. Additional near visible bands are observed in the two lead borate and bismuth borate glasses due to characteristic absorption of Pb^{2+} and Bi^{3+} ions. Gamma irradiation causes varying responses depending on the type of glass. Bismuth borate glass shows highly marked shielding towards successive gamma irradiation while lead borate glass reveals resistance in low dose (4 Mrad) and produces induced defects at high dose (8 Mrad). The other glasses show obvious generation of induced defects upon gamma irradiation. FTIR spectra of the glasses reveal vibrational modes characteristic to combined presence of triangular and tetrahedral borate units in their specific different wavenumbers besides the presence of interfering structural Pb–O and Bi–O linkages in the lead borate and bismuth borate glasses. Gamma irradiation produces limited changes in the FTIR spectra specifically the intensity of some vibrational modes while the bismuth borate glass has proved to be the best radiation-shielding candidate when comparing the combined spectral results after successive gamma irradiation.

1. Introduction

During the last two decades, there has been an increasing interest in the preparation, characterization and applications of heavy metal oxide (HMO) glasses due to their valuable physical properties, such as high refractive index, high transmission, high infrared transparency, high density and increased third-order nonlinear optical susceptibility [1–3]. Glasses containing heavy metal oxides possess broad range of applications including the fields of optical fiber amplifiers, thermal and mechanical sensors, laser materials, reflecting windows, shielding window glass for hot camber, encapsulation of radioactive wastes.

The resistance of glasses to radiation is desirable, since fluorescence decreases significantly with radiation damage [4]. Also, it is evident that gamma irradiation affects the optical properties of various glasses with varying degrees depending on the type and composition of glass including the presence of transition metal ions even if present as impurities [5–9].

The nature of radiation damages in glasses depends on the type (i.e., ionizing vs. particle) and energy of radiation impinging on the material [10]. The resultant effects may be divided into three categories: (1) atomic displacement by momentum and energy transfer, (2) ionization and charge trapping, and (3) radiolytic or photochemical effects. The relative contributions to the net radiation damage depend on the energy of the radiation, as well as the total dose besides the presence of heavy metal oxides within the glass constituents.

When ionizing radiation (such as gamma rays, X-rays, electrons, ultraviolet radiation, and so on…) impinges on the glass, electrons are initially excited from the valence band if the incident energy is greater than the band gap. The excess energy is converted to kinetic energy and these electrons travel through the glass, they will either recombine with positively charged holes, become trapped to form color centers. Finally, when the electron energy becomes too low to ionize other electrons, they will either be trapped or recombine with holes.

Recent spectral optical and FTIR studies by various glass scientists [6–9] have arrived to the conclusion that heavy metal oxides in glasses have potential effects on gamma-radiation causing some shielding behavior because of their heavy masses and high absorption cross-section for radiation. The spectral absorption curves are observed to remain unchanged or slightly affected by successive gamma irradiation.

The present work aims to investigate combined spectral properties of some prepared selected binary borate glasses of the composition B_2O_5 55% and 45% of one of the heavy oxides: PbO, Bi_2O_3, BaO, SrO or CdO. The study includes optical (UV–Visible) and FT infrared absorption

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spectral measurements of the glasses before and after gamma irradiation with a final dose of (8 M rad = 8×10^4 Gy). This specific gamma dose is chosen on the basis of previous extended studies which indicated that the irradiation process reached saturation at this dose level for spectral changes upon gamma irradiation [5–9,11].

This work is expected to evaluate the effects of gamma irradiation on the two measured spectral properties and justify the induced defects generated from these collective heavy metal oxides with their partner borate network. Also, the study aims to compare the extent of shielding behavior or retardation effect of the different studied binary glasses towards gamma irradiation.

2. Experimental procedure

2.1. Preparation of glasses

The starting pure chemicals used in the preparation of the glasses include orthoboric acid (H_3BO_3), red lead oxide (Pb_3O_4), bismuth trioxide Bi_2O_3, heavy carbonates of $SrCO_3$, $BaCO_3$ and $CdCO_3$. The glasses were prepared by melting accurately weighed powdered batches (Table 1) in platinum crucibles at 1100–1200 °C for 90 min. The melts were rotated at intervals of 30 min to reach acceptable chemical and physical homogeneity. The homogeneous melts were poured into warmed stainless steel molds of the required dimensions. The prepared samples were immediately transferred to a muffle regulated at 400 °C for annealing (350 °C for lead borate and bismuth borate glasses). The muffle after 1 h was switched off to cool to room temperature at a rate of 30 °C/h.

2.2. Optical absorption measurements

Optical (UV–visible) absorption spectral measurements were performed by using an UV–VIS spectrophotometer (type T80, PG Instruments, England). The range of measurement was from 200 to 1100 nm. Perfectly polished samples of the dimensions $1 \times 1 \times 0.2$ cm^3 were measured twice to confirm the accuracy of the absorption peaks. The thickness of the samples is constant at (2 ± 0.1 mm).

2.3. FT infrared absorption measurements

FT infrared absorption spectra were recorded at room temperature in the 400–4000 cm^{-1} range using a spectrometer of type (Mattson 5000 FTIR spectrometer, USA).

2 mg of each sample in the powder form was mixed with KBr (200 mg) and was well mixed and subjected to a load of 5 tons/cm^2 to produce clear disks. Each IR spectrum represents an average of 20 scans, normalized by using a blank KBr disk. FTIR spectral measurements were performed immediately before and after gamma irradiation.

2.4. Gamma irradiation facility

A ^{60}Co gamma cell (2000 Ci) was used as a gamma-ray source with a dose rate of 1.5 Gy/s (150 rad/s) at room temperature (30 °C). All the investigated glasses were subjected to the total gamma dose 8×10^4 Gy(8 Mrad). Using a Fricke dosimeter, the absorbed dose in water was utilized in terms of dose in glass. No cavity theory correction was made.

Table 1
Chemical composition of the prepared glasses.

Mol.%					
B_2O_3	SrO	CdO	BaO	PbO	Bi_2O_3
55	45	–	–	–	–
55	–	45	–	–	–
55	–	–	45	–	–
55	–	–	–	45	–
55	–	–	–	–	45

3. Results

3.1. Optical absorption spectra before and after gamma irradiation

Figs. (1–5) illustrate the optical absorption spectra of the prepared glasses before and after gamma irradiation with two specific gamma doses (4 Mrad, 8 Mrad). The optical spectra reveal the following spectral features:

(a) The binary strontium borate glass (Fig. 1) shows that a strong highly distinguished absorption with a distinct peak at 235 nm followed a medium absorption peak at about 375 nm and no further visible absorption can be observed. On subjecting this glass to a gamma dose of 4 Mrad, the UV absorption is observed at 235 nm and two further bands are identified at about 360 nm and a small one at about 610 nm. On increasing the gamma dose to 8 Mrad, all the absorption bands retain in their positions but the two bands at 360 and 610 nm are more identified.

(b) The binary cadmium borate glass (Fig. 2) reveals a distinct strong UV band at about 240 nm and with no further bands. On subjecting this glass to 4 Mrad gamma dose, the UV absorption shows two peaks at 230 nm and 250 nm and with no further bands. On further irradiation of the glass to 8 Mrad, the UV peaks are identified at 230 and 258 nm and the visible spectrum reveals a small curvature at 550 nm.

(c) The binary barium borate glass (Fig. 3) reveals a strong UV absorption band at 220 nm and no further visible bands are identified. On subjecting this glass to a gamma dose of 4 M rad, two induced bands are observed at 370 and 600 nm. On further irradiation to a dose of 8 Mrad, the optical spectrum reveals distinct three bands at 270, 370 and 603 nm.

(d) The binary lead borate glass (Fig. 4) shows before irradiation strong charge transfer ultraviolet absorption revealing two peaks at 231 and 285 nm and no visible peaks could be identified. On subjecting this glass to a gamma dose of 4 Mrad, all the spectral features remain similar to that before irradiation but the UV spectrum slightly decreases in intensity but the rest spectrum is almost parallel. On increasing the gamma dose to 8 M rad, the UV absorption spectrum shifts to longer wavelength and two peaks are identified at about 280 and 300 nm and the visible spectrum reveals two small broad bands centered at about 500 and 701 nm.

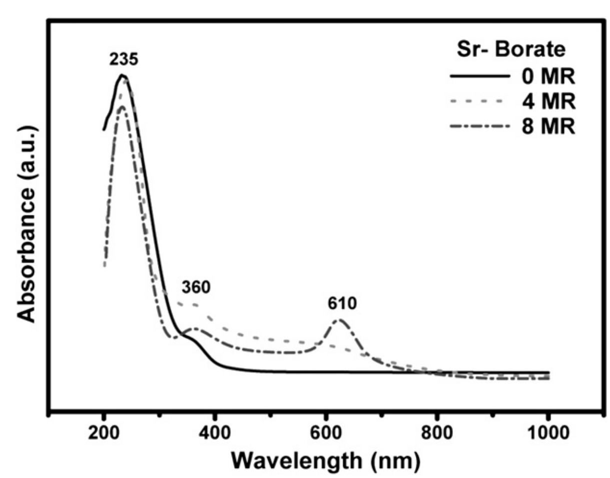

Fig. 1. UV–Visible absorption spectra of binary strontium borate before and after 4&8 Mrad gamma irradiation.