Journal of Non-Crystalline Solids 494 (2018) 31–39

Contents lists available at ScienceDirect

Journal of Non-Crystalline Solids

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

Optical and infrared spectral investigations of cadmium zinc phosphate glasses doped with WO_3 or MoO_3 before and after subjecting to gamma irradiation

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A R T I C L E I N F O

Keywords:
Cadmium zinc phosphate glass
WO_3-MoO_3
UV–vis, FTIR
Gamma irradiation

A B S T R A C T

Optical and FTIR spectral investigations of prepared undoped and WO_3-or MoO_3-doped glasses from ZnO-CdO-P_2O_5 glassy system were carried out before and after irradiation with a dose of 8 Mrad (= 8×10^4 Gy). The optical spectrum of the undoped glass reveals strong UV absorption band due to trace ferric ions present as trace impurity within the chemicals. The WO_3 or MoO_3-doped glasses show additional UV–visible bands due to the dominance presence of the low valence states of the two transition metal (TM) ions (M^{3+}, M^{4+}, M^{5+}). Gamma irradiation extends and increases the UV absorption due to suggested photochemical reactions of ferrous iron present as impurity to react with positive holes during the irradiation process to transfer to additional ferric ions possessing their absorption in the UV region. The TM-doped glasses show some shielding behavior in almost the persistence of the visible spectra due to the TM ions. FTIR spectral bands are related to phosphate groups (mainly Q^2 and Q^3 units) in accordance with the high content of P_2O_5 (70 mol%). The effects of dopant ions on the IR spectra are limited due to the low percent (0.25 to 0.75 g/100 g) and their presence in modifying positions. Gamma irradiation decreases the intensity of some IR bands due to OH, water vibrations present mainly in terminal sites.

1. Introduction

It has been established that continuous recent research activities have been identified after the review articles by Martin [1] and Brow [2] and other related publications concerning the wide properties and applications of phosphate glasses [3–5]. Phosphate glasses possess extended chemical compositions, easy melting at lower temperatures, high transmission in the UV region. The modification or the introduction of fluorides or multivalent oxides (such as Al_2O_3, Fe_2O_3, PbO) increases the applications including glass-sealing, low melting glass solders, encapsulation of some radioactive wastes, biomedical candidates and optical glasses candidates [6–10].

WO_3 and MoO_3 are very similar transition metal oxides, which exhibit close optical and electrical properties. They can exist in glasses in four valences, the trivalent, tetravalent, pentavalent and hexavalent states [11–14]. The percent of each state depends on the type and composition of the glass, condition of melting beside the concentration of the transition metal oxide.

Extended studies of WO_3 and MoO_3 doped glasses have indicated [11–14] that the hexavalent states M^{6+} of the two transition metals is the prevailing form in alkali borate and alkali silicate glasses while the lower valences (M^{5+}, M^{4+}, M^{3+}) are predominant in the phosphate glasses. Optical absorption and ESR measurements in various glasses were efficiently used to identify the various valences of molybdenum and tungsten ions [11–18]. It has been recognized that the hexavalent state of the two transition metals belongs to d^0 configuration and exhibits only a charge transfer UV band. On the other hand, the pentavalent state belongs to d^1 and possesses a single d-d band at ~750 nm, the tetravalent ion belongs to d^2 configuration with two bands at 450 and 550 nm while the trivalent ion belongs to d^3 configuration with d-d transitions at 350 and 440 nm [15–19].

WO_3 and MoO_3 can be incorporated with high percent within phosphate glasses to produce vitreous materials in which suggested MO_4 and MO_6 groups are assumed to share in the formation of the glass network [20,21].

It is accepted that the formation of radiation induced defects in pure materials originate at imperfections or dislocations present before irradiation [22]. Glasses normally possess short-range order or non-periodic structure and contain numerous intrinsic defect sites, such as non-bridging oxygens, trace impurities, flaws, etc.

When glasses are subjected to ionizing gamma irradiation, electrons are ionized and travel through the network and leaving formed positive

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https://doi.org/10.1016/j.jnoncrysol.2018.04.053
Received 11 February 2018; Received in revised form 21 March 2018; Accepted 26 April 2018
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holes. The net results of continuous irradiation include mainly ionization, charge trapping and radiolytic or photochemical effects [3,4,22,23].

Some scientists [13,14,24,25] have assumed that certain transition metal ions (e.g. V^{5+}, Cu^{2+}, Mo^{6+},W^{6+}) show obvious shielding towards successive gamma irradiation. The optical structural curves remain almost unchanged with progressive irradiation and the FTIR vibrational bands are nearly unaffected except identified minor changes in the intensities of few IR bands specifically that are related to water, OH vibrations.

The present work aims to prepare some glasses containing dopants (0.25, 0.50, 0.75 g/100 g) of WO_3 or MoO_3within the basic host glass (P_2O_5 70- ZnO 15- CdO 15 mol%) and then characterize their collective optical and FT infrared absorption spectra before and after gamma irradiation with a dose of 8 Mrad ($= 8 \times 10^4$ Gy). This specified dose (8 Mrad) was applied in this work because extended studies from our department by the sharing of some present authors; for example [26–31] have reached to the conclusion that the increase of dose of gamma rays irradiation produced continuous changes in the intensity or extension of induced bands up to 6 M rad. After this gamma dose, the changes in the optical spectra reached to saturation or show some minor changes or bleaching behavior. It is expected to characterize the detailed effects of gamma irradiation on both of the studied spectral properties and to justifying the generated induced defects in both the undoped base glass and WO_3, MoO_3-doped glasses. A final objective of the work is to find out the suitability of WO_3- or MoO_3-doped glasses to shield gamma irradiation.

2. Experimental details

2.1. Preparation of the glasses

The studied glasses were prepared from laboratory chemicals including: ammonium dihydrogen orthophosphate ($NH_4H_2PO_4$) for P_2O_5, cadmium carbonate ($CdCO_3$) for CdO and zinc oxide (ZnO) as such. The added dopants were introduced as (0.25, 0.50 and 0.75 wt%) of either WO_3 or MoO_3. The accurately weighed batches were melted in covered porcelain crucibles at 1150 °C ± 5 °C for 60 min in a SiC heated furnace (Vecstar, UK). The melts were rotated at intervals of 20 min to arrive for complete mixing and acceptable homogeneity.

The homogenous melts were casted into stainless steel molds. The prepared glassy samples were quickly transferred to an annealing muffle regulated at 300 °C. The muffle was switched off after 1 h and left to cool with the glasses inside to room temperature at a rate of 30 °C/h.

2.2. Optical (UV–visible) absorption measurements

Polished samples of equal thickness (2 ± 0.1 mm) were measured in a recording spectrophotometer type (Jasco, V-570 Japan) in the range of wavelength 200–2500 nm before and after gamma irradiation with a dose of (8 Mrad = 8×10^4 Gy). The optical measurements were carried out twice to confirm the accuracy of the peaks.

2.3. FT infrared absorption measurements

Fine powders of the glasses were investigated for their FTIR spectra through the KBr disc technique. 2 mg of the glass powder was mixed with 200 mg of KBr in an evocable die subjected to a load of 5 tons/cm^2 to produce clear homogenous discs. The FTIR spectra were measured immediately after the preparation of the discs using FTIR spectrometer (type Mattson, 5000, USA). The same measurements were repeated for the irradiated samples. The IR spectra were measured in the wavenumber range between 400 and 2000 cm^{-1}. At least two spectra for each sample were recorded. Infrared spectra were corrected for the dark current noises and background using the two point base line correction.

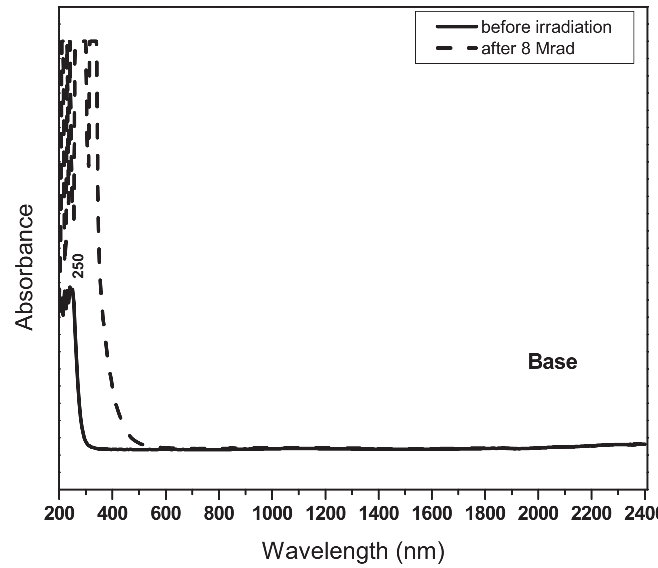

Fig. 1. Optical absorption spectra of undoped base: CdO-ZnO-P_2O_5 glass before and after gamma irradiation (dose 8 M rad).

2.4. Gamma ray irradiation facility

An Indian ^{60}Co gamma cell (2000 ci) was used as a gamma ray source with a dose of 1.5 Gy/s (150 rad/s) at a temperature of 30 °C.

3. Results

3.1. Optical absorption spectra of the undoped base glass before and after gamma irradiation

Fig. 1 illustrates the optical (UV–visible) absorption spectra of the base undoped cadmium zinc phosphate host glass. The optical spectrum before irradiation reveals a distinct strong UV absorption band with a peak at about 250 nm and with no further absorption to the rest of the measurement. On subjecting this glass to a 8 Mrad dose of gamma irradiation, the UV spectrum highly increases in intensity with the resolution of two peaks at about 250 and 320 nm and the UV absorption extends to about 400 nm and the rest of the spectrum is very close to that before irradiation.

3.2. Optical absorption spectra of WO_3-doped glasses before and after gamma irradiation

Fig. 2 reveals the optical spectra of the WO_3-doped (0.25, 0.50 and 0.75%) glasses before and after gamma irradiation. The glass doped with 0.25% WO_3 (Fig. 2a) reveals three consecutive sharp peaks extending from the UV to near visible with peaks at about 267, 363 and 450 nm with their intensities slightly decrease with the increase of wavelength. Three extra small peaks follow the previous UV-near visible peaks at about 622, 690 and 980 nm.

On gamma irradiation, the spectral curve of the 0.25% WO_3 is very close after irradiation to that before irradiation with the appearance of the three successive strong peaks and the three small visible peaks at 622, 690, and 980 nm. The glass doped with 0.50% WO_3 glass (Fig. 2) reveals three strong and sharp UV-near visible bands with peaks at about 260, 330 and 430 nm with their intensities increasing with the increase of wavelength. This is followed by the appearance of two small peaks at about 622 and 987 nm.

The spectrum of the 0.50% WO_3 glass after gamma irradiation reveals the increase of the UV absorption peaks to higher intensities while the rest visible small peaks remain unchanged in their positions and intensities.

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