Research Article

Effect of gamma ray on some properties of bismuth borate glasses containing different transition metals

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Abstract
This work aims to study the influence of gamma irradiation on some bismuth borate glasses doped with different transition metal oxides (CuO, CoO, ZnO and CdO). The structure of the glass matrix was analyzed by Fourier transform infrared and ultraviolet–visible spectroscopy before and after gamma irradiation at doses of 1 kGy, 20 kGy and 50 kGy. Additionally, the attention is made in determining the attenuation parameters against gamma rays. Mass attenuation coefficient determined experimentally and calculated theoretically by WinXCom program, which is a significant program in the field of radiation shielding. It is found that the attenuation parameters of gamma ray have higher values of the sample without transition metal oxides and the other doped with CdO. From hardness measurements before and after irradiation, it was found that hardness increases with the addition of transition metal oxides to glass. The results of different techniques show that these glasses possess a slight effect on the network structure of the samples against gamma irradiation. So, it can be used in different applications of nuclear fields.

Keywords Bismuth borate glasses · Gamma ray · Optical and IR spectroscopy

1 Introduction

Recently, glass materials gained a growing interest in the field of radiation shielding because of their transparency to visible light and the possibility of modifying their properties by composition. Many researchers [1–4] developed the use of borate glass as radiation shielding materials. B_2O_3 is considered as one of the best glass formers, where it is has high transparency, low melting point and high thermal stability and is often used as a dielectric and insulating material.

Bismuth oxide gained a great attention as it can improve the chemical durability of glass, has a high refractive index and non-toxic nature, very good infrared transmissions and high values of nonlinear optical susceptibility and polarizability [5–7].

It was found that the addition of bismuth oxide to borate glasses develops the electrical, optical, and thermal properties of the glasses. The high third-order nonlinear optical susceptibility and long infrared cutoff of bismuth borate glasses make them very suitable for developing ultrafast optical switches, photonic devices and infrared transmission components [8, 9]. Additionally, these glasses can be used in several applications such as glass ceramics like substrates of optical and electronic devices, sensors, and reflecting windows [10, 11]. In recent decades, attention has been focused on bismuth borate-based compounds for their applications as piezoelectric, ferroelectric, pyroelectric, and nonlinear optical device materials [12, 13].

Nowadays, heavy metal oxide glasses have been used in several fields due to their favorite properties [14–16]. Moreover, transition metals (T_M) can be added during

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the preparation of the glass system to improve the optical, electrical and shielding properties of glasses [17–19], where the functions of the outer electron orbital in T_M are very sensitive to the surrounding cations. Recently, induced optical absorption in glasses due to irradiation has encouraged the utilizing of glass in several optical technology fields. The bismuth–borate glasses doped with Cu show a clear retardation effect against γ-irradiation [20]. The glasses including cobalt are preferred for glass filters or radiation-sensitive indicators and for many practical applications [21–23]. While doping glasses with heavy metal oxide ZnO gives a good result in the field of radiation shielding application [24], the addition of CdO to the glass matrix leads to stabilizing the glass structure and optimize the electrical and optical properties of glass [25].

In this work, the effect of different transition metal oxides *TMO* (CuO, CoO, ZnO and CdO) on the expense of Bi_2O_3, at concentration of 0.21 mol% in the present glasses was illustrated. The structure of the glasses was analyzed before and after gamma irradiation by Fourier transform infrared (FTIR) and ultraviolet (UV)–visible spectroscopy. Also, the influence of irradiation on the hardness was shown. Additionally, the focus is made on determining the attenuation parameters of gamma ray which are important in the field of radiation shielding materials.

2 Materials and methods

In this work, the experimental values of mass attenuation coefficient μ/ρ (g/cm^2) were compared with the theoretical values which were calculated by WinXCom program [26]. The experimental values of linear attenuation coefficient (μ) were measured using the narrow beam transmission geometry, by the following equation,

$$\mu = \ln\left(\frac{I_o}{I}\right)/x \tag{1}$$

where I_o and I are the intensities of the incident and transmitted gamma ray energies, respectively, and x is the thickness of the glass sample under measurement. The values of mass attenuation coefficient (μ_m) were then obtained by

$$\mu_m = \frac{\mu}{\rho} \tag{2}$$

where ρ is the experimental density of the sample.

By using the linear attenuation coefficient μ, the half value layer (HVL), which is the thickness required to reduce the transmitted intensity to one half the initial intensity, is determined by

$$\mathrm{HVL} = \frac{\ln 2}{\mu} \tag{3}$$

The effective atomic number Z_{eff} was calculated by

$$Z_{eff} = \frac{M\left(\frac{\mu}{\rho}\right)}{\sum_i n_i \sum_i \frac{f_i A_i}{Z_i}\left(\frac{\mu}{\rho}\right)_i} \tag{4}$$

where M, ρ, n_i, A_i, Z_i, $f_i \frac{n_i}{\sum_i n_i}$ are the molecular weight, the density, the number of formula units of the ith element, the atomic weight of the ith element, atomic number and the fractional abundance of the ith element, respectively. The Z_{eff} is an important parameter that gives an indication about how photon interacts with a substance and it changes with the photon energy.

In the present work, five glass samples having the composition ($0.45B_2O_3$–$0.1Na_2O$–0.21 *TMO*–0.24 Bi_2O_3) were prepared by melt quenching technique utilizing chemicals H_3BO_3, Na_2CO_3, Bi_2O_3, CuO, CoO, ZnO and CdO. The chemicals were mixed completely and melted in a porcelain crucible at 980 °C for 2 h. The glass samples were properly annealed at 350 °C. The density (ρ) of the samples was determined experimentally by Archimedes' method using immersion liquid tetrachloride with density 1.59 g/cm^3.

A Vicker's diamond indenture was used in a standard microhardness tester (Leco AMH 100, USA) for specimen indentation. A load of (100) gm applied for 20 s was used to make indentations in specimens of glasses. The microhardness was measured using polished samples of the glasses under investigation in the form of plates (3.5–5.6 mm) thick. Each sample was subjected to five indentations at randomly selected areas; hence, errors in the measured values corresponding to the standard deviation were found to be about 4%.

The μ values of the glass samples were measured using 3″ × 3″ NaI(Tl) scintillation detector and radioactive sources ^{137}Cs and ^{60}Co have activities 9.5 and 4.9 μCi, respectively.

The optical UV–visible transmission spectra of the prepared glass samples in the range 200 to 1000 nm were measured using Jenway 6405-UV–VIS spectrophotometer.

The infrared absorption spectra of the glasses were measured using FTIR Perkin Elmer-Spectrum in the range 400–4000 cm^{-1} at room temperature.

A ^{60}Co gamma cell has activity 1918.19 Ci was used to irradiate the glass samples at a dose rate of 1.323 kGy/h.

3 Results and discussion

Figure 1 shows a pure amorphous phase of the glass samples using the X-Ray Diffraction (XRD) spectra. Figure 1 illustrates the lack of periodicity of the studied samples due to the absence of any significant peaks of X-ray chart, and so all the studied samples are in amorphous states.

3.1 Density and hardness measurements

Density, molar volume and hardness are powerful tools capable of exploring the structure and the changes occurred in it with the change in the compositions. They are sensitive to the structural softening/compactness of the glass system, change in the geometrical configuration, coordination number, cross-link density and with the dimension of the interstitial spaces of the glasses. The composition and the experimental values of density, molar volume and hardness of the present glasses before and after irradiation are shown in Table 1. It is obvious from the table that sample 1 (parent without *TMO*) has higher values of density and molar volume. Table 1 shows that as Bi_2O_3 is replaced by *TMO,* the density and molar volume decrease. The decrease in the density is due to the smaller values of molecular weight of all *TMO* than Bi_2O_3, while the molar volume decreases due the difference of the radius of *Bi* ion and both used *TMO*. The hardness results showed raising the hardness values of the samples containing *TMO*. This may be because the shortening of the bonds leads to more bond strength which increases the hardness as Bi_2O_3 is replaced by the *TMO*. After irradiation the hardness values are slightly decreased as the irradiation dose increases, this may be due to that gamma irradiation assumed to create displacements which decrease the hardness.

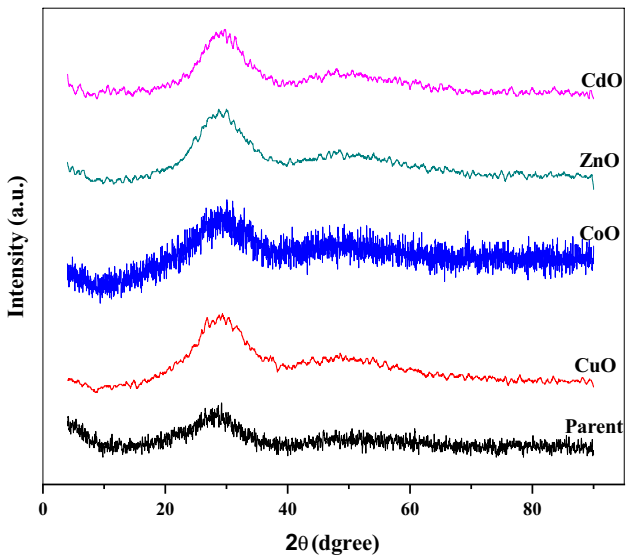

Fig. 1 The XRD of the parent sample and the samples doped with CuO, CoO, ZnO and CdO at concentration 0.21% mol

3.2 Attenuation coefficients of gamma ray

The experimental results compared with theoretical values of μ/ρ coefficient are shown in Fig. 2. It is observed that μ/ρ has higher values of sample 1 followed by sample 5 (doped with CdO); this is due to the higher density and molecular weight of these two samples than other samples.

The *HVL* results of the prepared glasses show a lower value than the results of ordinary concrete [27] and some shielding glasses RS 360 and RS 253 [28] as illustrated in Fig. 3. This means that a smaller thickness and fewer materials are needed to produce the same shielding effect against gamma ray compared with other shielding glasses.

The Z_{eff} value of a medium is considered a very useful parameter for several applications. Table 2 shows the calculated values of Z_{eff}. It is obvious from the data that the Z_{eff} values of samples 1 and 5 are higher than the other, because of the higher molecular weight of these two samples than other samples. This means that there are more electrons available for absorption; the incident photons improve the shielding characteristics.

Table 1 The composition and experimental values of density, molar volume and hardness of the prepared glasses

Sample no.	Composition (mol%)				Density (g/cm^3)	Molar volume (cm^3/mol)	Hardness kg/mm^2 average error ± 13.64			
	B2O3	Na2O	Bi2O3	Oxide (0.21% mol)			0 kGy	1 kGy	20 kGy	50 kGy
1	0.45	0.10	0.45	–	5.47	45.17	464.32	454.42	448.12	444.42
2	0.45	0.10	0.24	CuO	4.37	38.01	479.39	470.09	464.81	461.59
3	0.45	0.10	0.24	CoO	4.66	35.42	490.33	482.53	476.63	471.22
4	0.45	0.10	0.24	ZnO	4.73	35.18	519.36	510.32	504.69	500.96
5	0.45	0.10	0.24	CdO	5.02	35.16	475.34	464.61	458.97	454.64

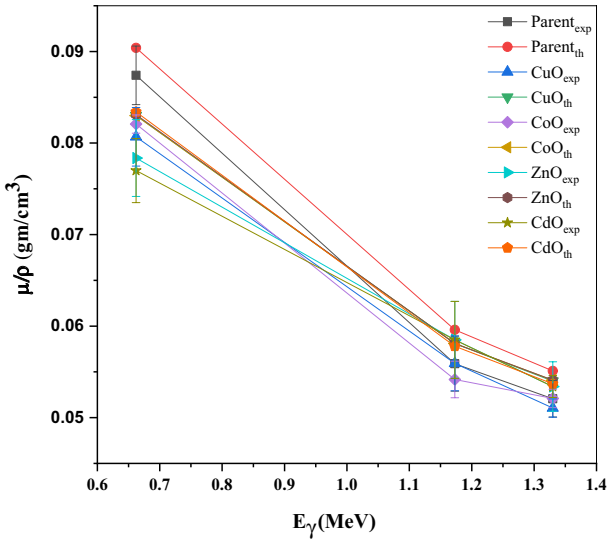

Fig. 2 The experimental and theoretical mass attenuation coefficient (μ/ρ) of the parent sample and the samples doped with *TMO* at concentration 0.21% mole

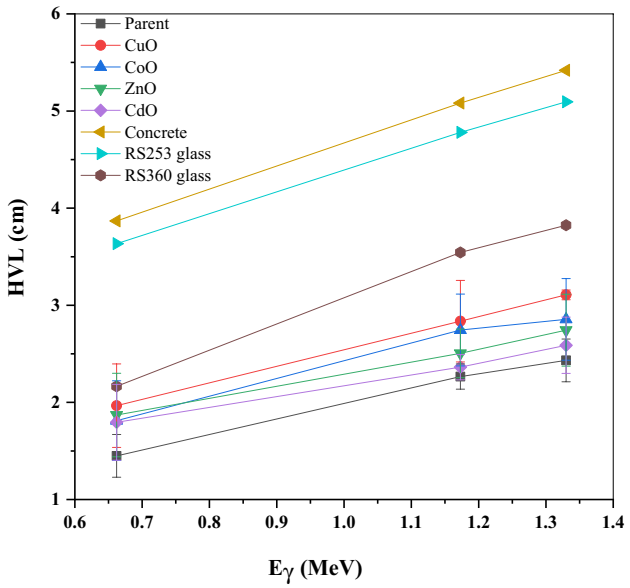

Fig. 3 The experimental *HVL* of the glass samples at different energies compared with ordinary concrete and other glasses

3.3 IR absorption spectra before irradiation

Figure 4 shows the IR spectra of the studied glasses before irradiation. The band state of the un-irradiated glasses is almost comparable; on the other hand, there is a customary change in the relative areas and intensities of a few groups.

The infrared spectra of borate glasses can be parted into three groups. The primary group found at 1200–1500 cm^{-1} is due to the asymmetric stretching vibration of B–O bonds in the triangle units BO_3. The second group at 800–1200 cm^{-1} is due to the symmetric vibration of the B–∅ bonds in $B∅_4$ tetrahedral units and from the three types of borate forms, triborate ($B_3O_5^-$), tetraborate ($B_8O_{13}^-$) and pentaborate ($B_5O_8^-$) groups, where ∅ is a bridging oxygen atom connecting two boron atoms. The third group at 800–400 cm^{-1} is due to the bending vibrations of borate groups. Bi_2O_3 has six absorption bands at 380, 425, 465, 510, 540 and 595 cm^{-1} related to the vibration of Bi–O in BiO_6; it has also five absorption bands nearly at 350, 470, 540,620 and 840 cm^{-1} related to the vibration of Bi–O in BiO_3 units, but BiO_3 doesn't include in this work because it appears at very high concentration of Bi_2O_3 [29, 30].

The band profile of the IR spectra (Fig. 4) clearly shows that sample 1 has the characteristic three regions of B_2O_3, and the distinguishing bands of Bi–O in the formulae of BiO_6 are overlapping with the borate group bands. The bands of Bi_2O_3 and B_2O_3 have been discussed in previous work [31], in which it is found that the increase in Bi_2O_3 concentration leads to the increment of the BO_4 groups and a decline of BO_3 groups. The increase in BO_4 causes an increment in the density. The decline of BO_3 decreases the strength of the bonds, which causes a decline of the hardness values.

In the present work, as a part of Bi_2O_3 is replaced with different *TMO* (CuO, CoO, ZnO and CdO) at about 0.21 mol %, this causes a change in the relative intensity of the three group bands. Figure 5 illustrates that the relative intensity of the band at ~ 700 cm^{-1} has an infinitesimal increase in the case of samples containing CoO and CdO. While there is a clear increase in the relative intensity in the

Table 2 Effective atomic number Z_{eff} of the present glass samples

Sample	Oxide (0.21%mol)	0.662 MeV		1.173 MeV		1.33 MeV	
		Experiment	Theory	Experiment	Theory	Experiment	Theory
1	–	25.62	26.96	25.19	25.45	24.80	25.30
2	CuO	20.22	20.63	20.01	20.06	19.77	20.01
3	CoO	23.54	23.81	22.69	22.74	22.17	22.68
4	ZnO	23.35	24.76	23.57	23.65	23.03	23.59
5	CdO	24.55	26.60	25.10	25.21	24.53	25.09

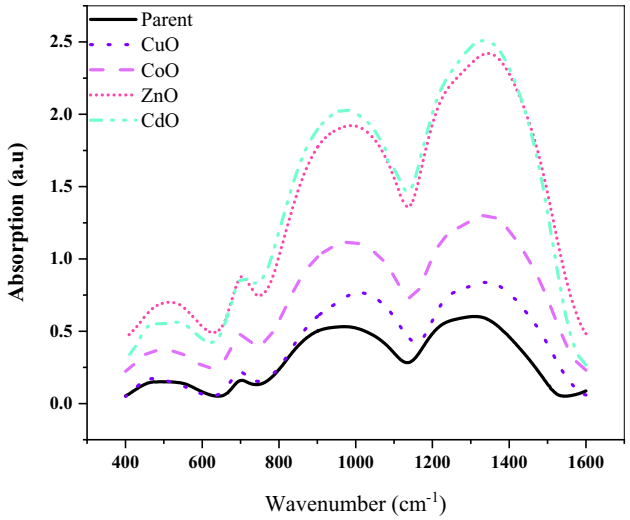

Fig. 4 The IR spectra for the present glasses with different additions of *TMO* (CuO, CoO, ZnO and CdO) at concentration 0.21% mol before irradiation

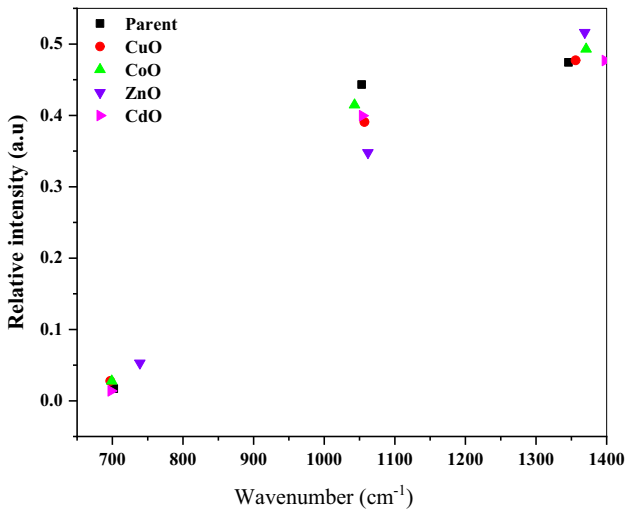

Fig. 5 The relative intensity of the three group bands at (~700, 800–1200 and 1200–1500 cm^{-1}) for the prepared glasses before irradiation

case of a sample containing ZnO, because ZnO can break the bonds between glass former ions and oxygen [32] and used in composition with B_2O_3 to break the bonds and forming BO_4 [33], while there is a decline in the relative intensity of the BO_4 band at ~800–1200 cm^{-1} and an increment in the relative intensity of the BO_3 band observed at ~1200–1500 cm^{-1}. This increment of the relative intensity of BO_3 group leads to increasing the strength of the bonds, which causes an increase in the stability of the entire glass composition and increases hardness values, while the decrease in BO_4 relative intensity leads to a decrease in the density and molar volume as shown in Table 1.

3.4 IR absorption spectra after irradiation

All the present samples were exposed to gamma radiation at doses of 1 kGy, 20 kGy and 50 kGy. Figure 6 shows that the IR spectra of the parent glass (sample 1) have no significant change in the position of the bands after irradiation at doses of 1 kGy and 20 kGy, while there is a broadness in the region of BO_4 at a dose of 50 kGy. All other samples containing *TMO* (CuO, CoO, ZnO and CdO) show a minor effect of irradiation. It is observed that there are broad bands as a result of an overlapping of some separate bands as shown in Fig. 6.

A deconvolution process [34] was utilized to isolate these groups to study the characteristic parameters of each discrete band. These parameters are the center "*C*" which determine the structure group and the relative zone "*A*" which is corresponding to the concentration of this group. The relation between radiation dosages of each studied glass sample and deconvolution information is demonstrated in Fig. 7, which shows that there is a minor change on the relative intensities of the bands and the main vibrational units approximately unchanged their position. This limited effect of irradiation may be due to a minor degree of disorder or some variations in the bond angles and/or bond lengths of the structural units which agrees with Mostafa et al. [35] and supports the small change in the hardness values after irradiation. The IR results after irradiation show minor changes in the position and relative intensities of the studied bands of both the parent and the *TMO*-doped glasses which indicate that there is a stability of the entire glass network. This leads to stability in the values of gamma-ray attenuation parameters of the prepared glasses when used as shielding for gamma irradiation in the studied ranges.

3.5 Optical properties before and after gamma irradiation

Figure 8 illustrates the transmittance spectra of the UV–visible region before irradiation of the parent sample and samples doped with *TMO*. The optical spectrum shows a robust ultraviolet absorption extending from 200 to 310 nm followed by visible transmission approaches saturation with a longer wavelength. The observed region at 200–310 nm may be due to the presence of ineluctable trace iron impurities (Fe^{3+} ions) within the raw materials combined with the effect of absorption due to the presence of high Bi^{3+} ions [36, 37].

Figure 9 shows the UV–visible transmittance spectra of each glass sample before and after successive

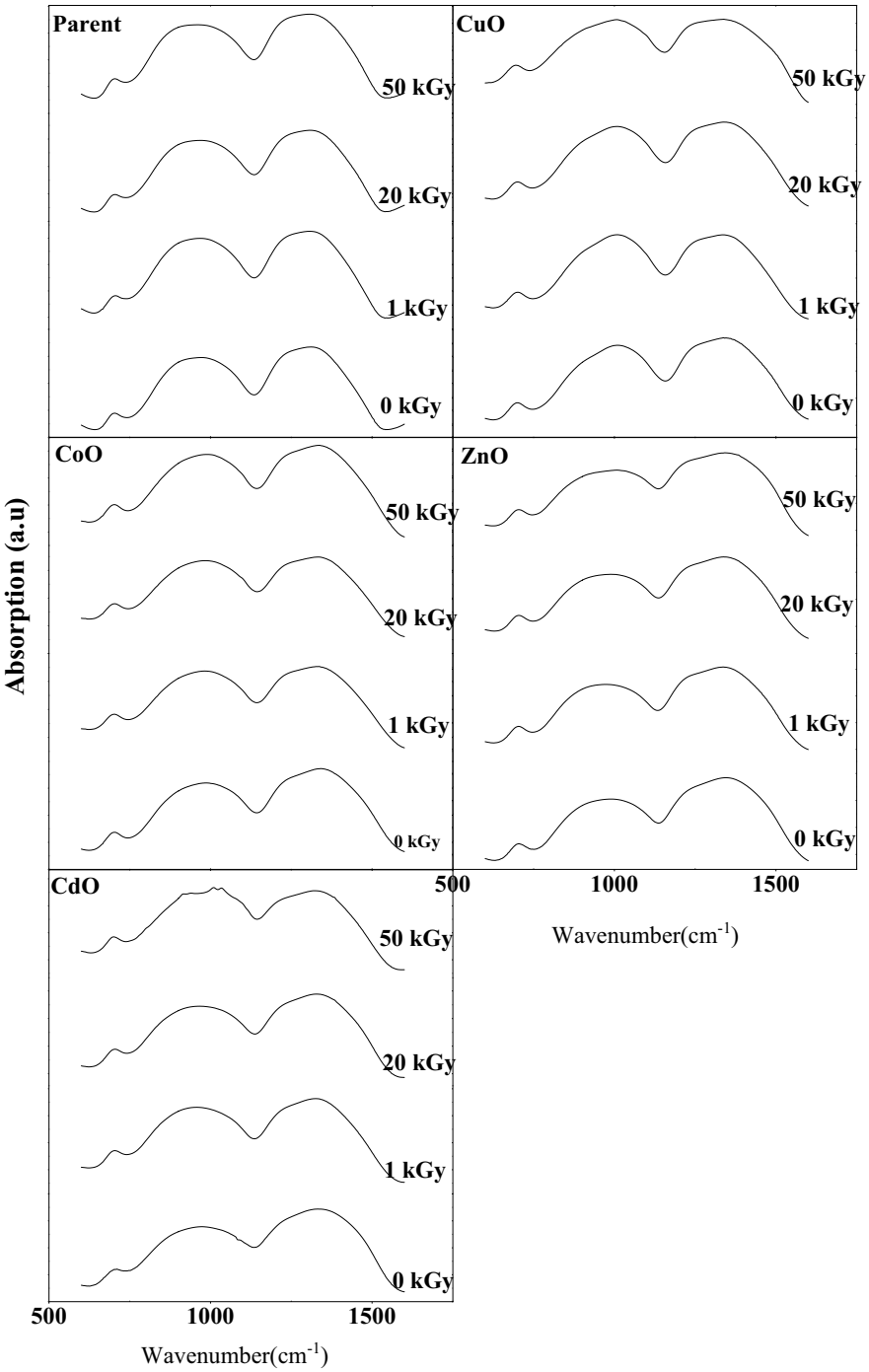

Fig. 6 The IR absorption spectra for the present glasses before and after irradiation with doses 1, 20 and 50 kGy

gamma irradiation. On progressive gamma irradiation, the UV–visible spectra show a decrease in the intensities of the percentage transmission spectra at 1 kGy and 20 kGy doses, followed by a higher intensity at 50 kGy without a considerable change in the general character of the spectra. It is obvious that the parent sample and the sample doped with CuO have a minimum effect in the percentage transmittance intensity after successive gamma irradiation. This behavior shows a perceptible resistance to irradiation which agrees with several researchers [38, 39].

After gamma irradiation, the elicited ultraviolet light and visual defects result from the impact of irradiation on the glass composed of borate network and inessential trace impurities. Additionally, owing to the non-periodic nature of glass and the presence of non-bridging oxygen and trace impurities, most glasses acquire

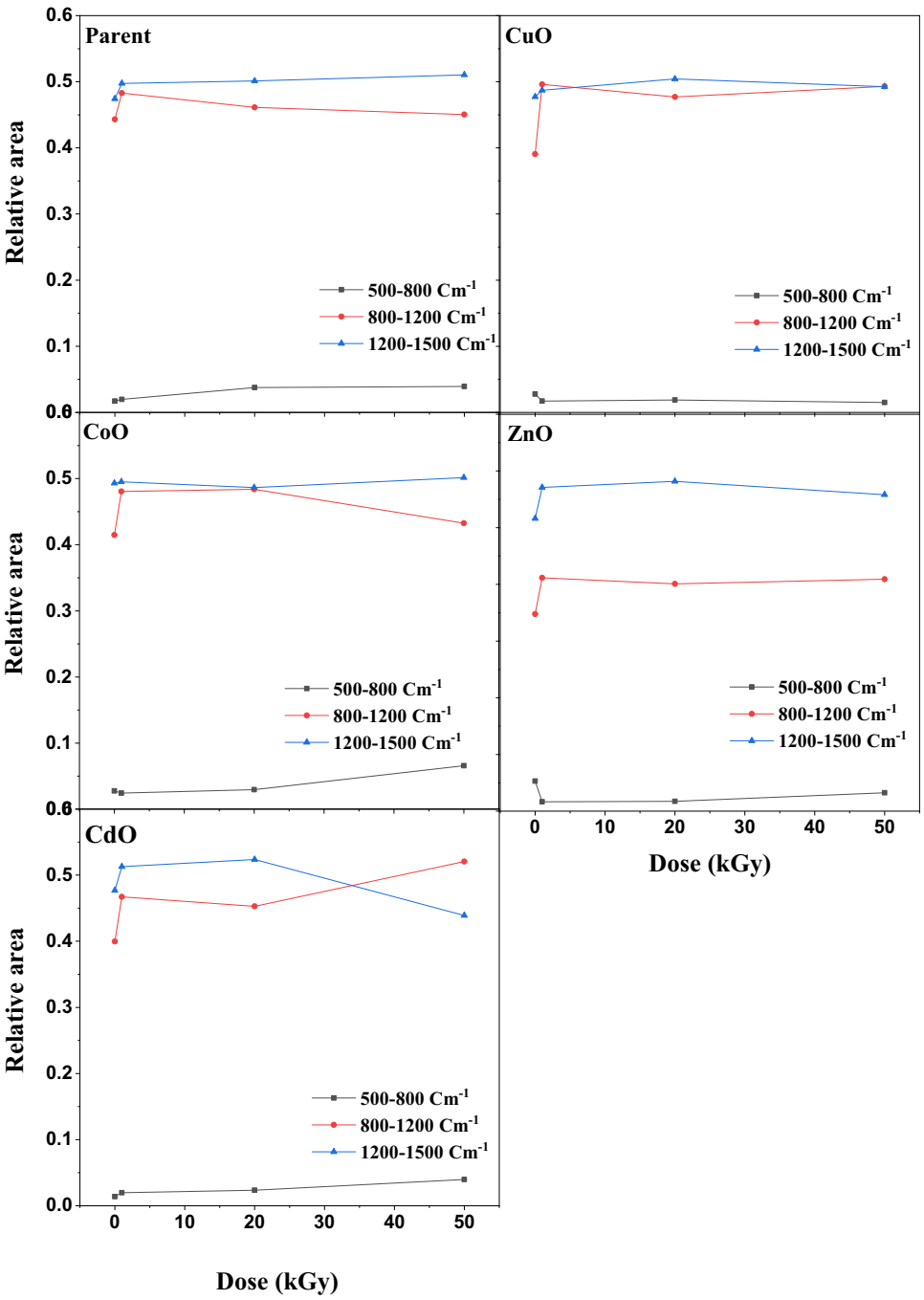

Fig. 7 The deconvolution data as a function of the radiation doses for the studied glass samples

radiation-induced defects that create characteristic optical absorption.

The optical energy band gap was interpreted by Davis and Mott [40] in general form $(\alpha h\nu) = B(h\nu - E_{opt})^n$, where α is the absorption coefficient, $h\nu$ is the photon energy, B is a constant, E_{opt} is the optical energy band gap, and n is an index which can have 1/2 for direct band gap transition or $n = 2$ for indirect band gap transition. The values of E_{opt} were determined by extrapolation of the linear part of each curve representing the variation of $(\alpha h\nu)^{1/2}$ with $h\nu$. The width of band tails (Urbach energy E_c) [41] can be determined from the relation $\alpha(\nu) = \alpha_0 \exp(h\nu/E_c)$, where α_0 is a constant. E_c of the present glasses were calculated by taking the reciprocal of the slope of the linear part of ln $\alpha(\nu)$ versus $h\nu$ plots. The absorption coefficient is given by the relation $\alpha(\nu) = -\ln T/t$, where T is the transmittance and t is the sample thickness [42]. Table 3 illustrates the values of the cutoff wavelengths, indirect band gap energies and the values of band tails of the prepared glass samples before and after irradiation. The

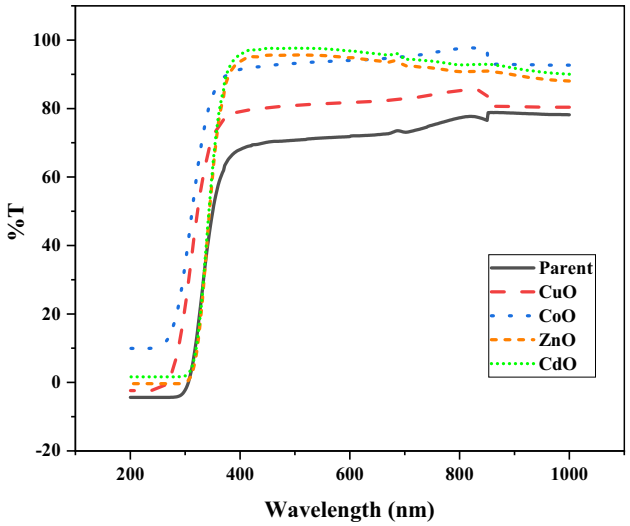

Fig. 8 The UV–visible transmittance spectra of the undoped and the doped glasses with *TMO* before irradiation

samples doped with CuO and CoO have shown a shift of the cutoff wavelength toward higher wavelength with successive irradiation. While the shift of the cutoff wavelength is toward lower wavelength of the samples doped with ZnO and CdO, it may be attributed to the formation of non-bridging oxygen. The appraised error of the optical band gap and band tail of all samples is around ± 0.005 eV and ± 0.002 eV, respectively. It seems that the optical band gap values change with the type of *TMO*. The inclusion of metal in salt structure might produce some defect states within the mid-gap, which is chargeable for band gap energy. From the FTIR analysis it was observed that the replacement of Bi_2O_3 by *TMO* leads to increase the relative intensity of BO_3 and decrease the relative intensity of BO_4 bands. The increase in BO_3 causes shortening of the bonds leads to raising of bond strength, which leads to increase the hardness and decrease the indirect band gap. While there is an increase in the indirect band gap energy of the sample containing CuO, this may be due to some overlapping of certain absorption bands given by Bi–O and B–O linkage vibrations. This overlapping makes the prediction of certain structure of Bi_2O_3–B_2O_3–CuO glass system is difficult [43]. The change in the width of the absorption edge could also be due to the presence of a localized state within the band tails. These defects cause fluctuations in bond angle leading to the increase in Urbach energy [44–46], while after irradiation at 1, 20 and 50 kGy, the optical band gap and Urbach energy values show a very slight change of each glass sample. This agrees with the limited variation in the behavior of relative intensities of BO_3 and BO_4 groups as a function of the radiation doses as shown in Fig. 7 in the *IR* study. After gamma irradiation, the elicited ultraviolet light and visual defects resulted from the impact of irradiation on the host glass composed of a borate network by increasing the concentration of non-bridging oxygen due to breaking the B–O covalent bond in glass matrix [37, 47]. The very slight change in the results of the *IR* analysis and optical properties of the samples after gamma irradiation indicates that the present glasses have a considerable resistance against irradiation in the range of 1–50 kGy.

4 Conclusion

In this work, the addition of different transition metal oxides to bismuth borate glasses modifies the stability of glass structure, which is supported by the results of Fourier transform infrared and optical spectroscopic studies. The infrared outcomes show a strong unity of the glass network due to the high intensity of BO_3 group as compared to BO_4, downsized the effect of gamma beams on the glass composition. This leads to a small outcome in the ultraviolet–visible results of all the calculated parameters after successive gamma irradiation. Additionally, the high intensity of BO_3 group leads to increase the hardness values with the addition of transition metal oxides to the glasses. The determination of gamma ray interaction parameters could indicate that the parent sample and the sample doped with CdO have lower values of half value layer and higher values of effective atomic number than those of the other samples. All the prepared glasses have lower values of half value layer than those of the other commercial shielding glasses RS 360, RS 253 and ordinary concrete, which means better attenuation effect against γ-irradiation. One can conclude that, from the analysis of the glass structure by Fourier transform infrared, which shows strong network of the glass matrix support the small change of the ultraviolet–visible and hardness measurements after successive gamma irradiation, the prepared glasses have a noticeable resistance to gamma irradiation and can be operationally beneficial as radiation shielding materials.

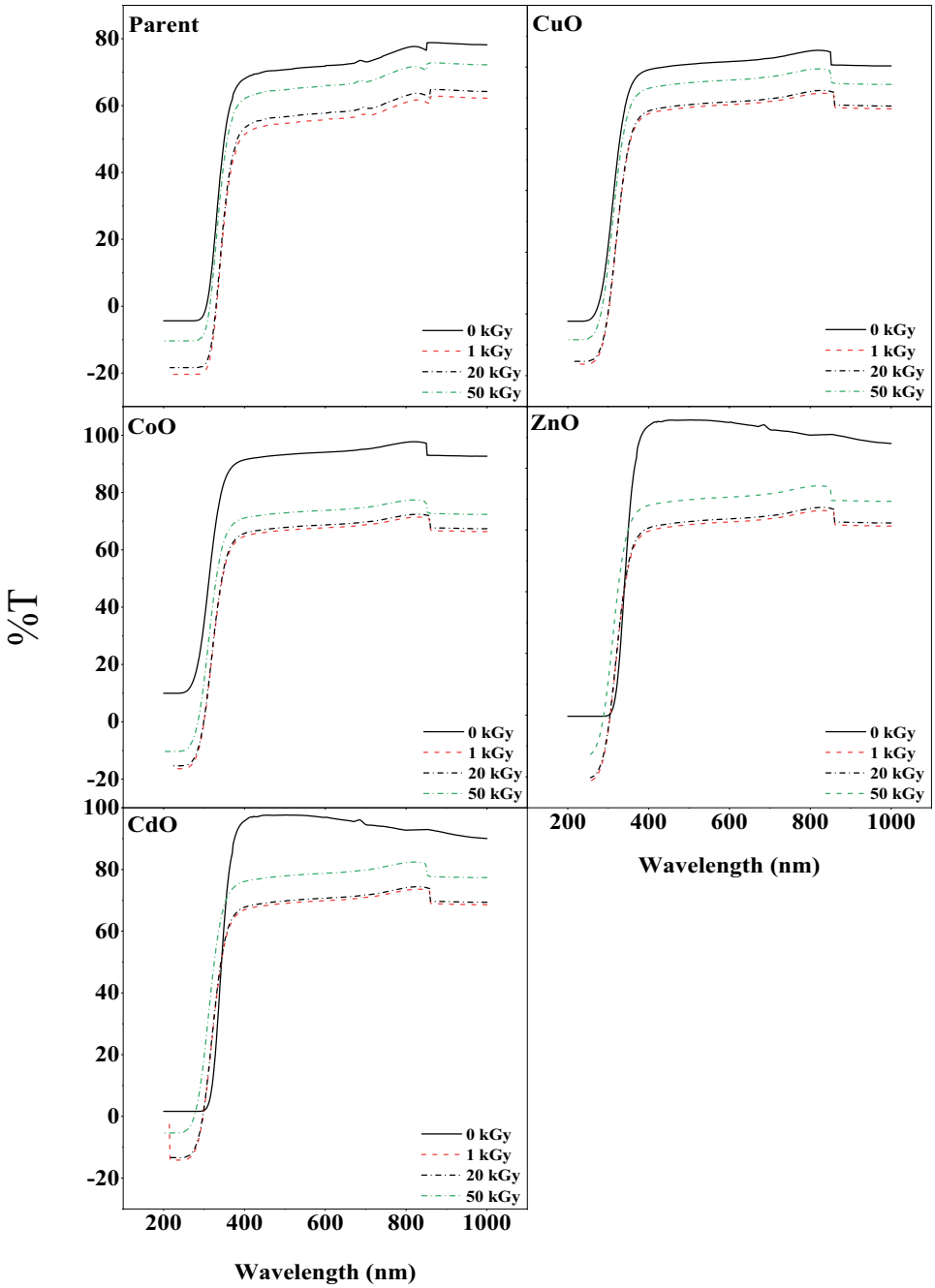

Fig. 9 The UV-transmittance spectra of each glass sample before and after irradiation

Table 3 The cutoff wavelengths, indirect band gap and breadth of tail values versus *TMO* contents at concentration 0.21% mol before and after irradiation

Samples	Cutoff wavelengths (nm) at doses				Indirect band gap (eV) at doses				Band tail (eV) at doses			
	0 kGy	1 kGy	20 kGy	50 kGy	0 kGy	1 kGy	20 kGy	50 kGy	0 kGy	1 kGy	20 kGy	50 kGy
Parent	293	313	310	300	3.91	3.71	3.72	3.91	0.64	0.62	0.61	0.63
CuO	257	280	283	268	4.52	4.02	4.11	4.31	0.71	0.67	0.66	0.73
CoO	259	288	278	270	3.72	3.7	3.69	3.68	0.67	0.67	0.66	0.66
ZnO	310	287	290	280	3.7	3.68	3.67	3.66	0.75	0.75	0.74	0.74
CdO	312	281	280	261	3.69	3.68	3.68	3.67	0.69	0.69	0.68	0.68

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Compliance with ethical standards

Conflict of interest The authors declare that they have no competing interests.

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