



Effect of gamma radiation on some optical and electrical properties of lithium bismuth silicate glasses

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Abstract

Lithium bismuth silicate glasses with the composition of; 68% SiO₂, 20% Bi₂O₃, 8% Li₂O₃, 4% Na₂O doped with either 2% PbO, BaO or SrO (all in wt%) were studied to investigate some of their optical and electrical properties before and after different doses of gamma radiation. Density, oxygen packing density OPD, molar and specific volumes, optical UV absorption, optical energy gap, electrical conductivity and dielectric measurements were measured and discussed in this study regarding the effect of gamma radiation. The results showed that all glasses were more affected at the higher radiation dose (100 kGy) than the lower radiation dose (50 kGy) because of the creation of more defects and non-bridging oxygens NBOs at the higher dose. In spite of that, Pb²⁺ ions containing glass revealed better properties to resist gamma radiation than glasses containing either Ba²⁺ or Sr²⁺ ions because of its highly compacted structure and the low concentration of NBOs, defects or vacancies as well as the high polarizability of Pb²⁺ ions that plays an effective role in enhancing the glass optical and electrical properties.

1 Introduction

Bismuth silicate glasses based on heavy metal and rare earth oxides have attracted considerable interests for their potential applications, superior physical, optical and electrical properties. So they have many applications in different fields of electronics. Bismuthate glasses possess also higher refractive index, large third-order non-linear optical susceptibility, high optical basicity, large polarizability, high density, absorption cross-section and dielectric permittivity [1–3]. All of these specific characteristics make them essential for numerous applications such as synthesizing of

high-temperature ceramic superconductors [4], thermal and mechanical sensors, reflecting windows, advanced optical telecommunication, processing devices, infrared transmission components, radiation shielding, novel lasers [1–3], microwave integrated circuits and electrochemical cells (as an oxygen conductive electrolyte) [4, 5]. The large polarizability and small field strength of Bi³⁺ ions in such oxide glasses make them also suitable for optical devices such as ultra-fast optical switches, optical Kerr shutters (OKS), optical isolators and environmental guidelines [1]. Also, glasses containing Bi₂O₃ have been developed for nuclear engineering applications due to their tendency to achieve the double task of allowing visibility and absorbing radiations like gamma-rays and neutrons.

Bismuth oxide can act as an intermediated oxide in the glassy network since Bi³⁺ ion can decrease its co-ordination number from VI to III and the glass network may consist of both [BiO₆] highly distorted octahedral and [BiO₃] pyramidal units [6]. Furthermore, When Bi₂O₃ is present in smaller concentration, up to 12 mol %, bismuth mainly occupies the octahedral positions and acts as a modifier, whereas, for higher concentrations it takes the network forming positions with [BiO₃] pyramidal units.

Doping of bismuthate glasses by an alkali metal ion (Li⁺ ions) modifies their structural network and consequently it is expected to enhance its ionic conductivity because lithium oxide is used widely in semiconducting materials

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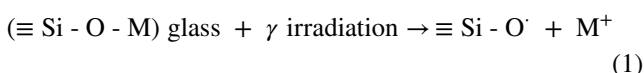
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due to its high mobility inside the glassy network and its higher tendency to carry the electric current [7–9]. Another alkali oxides like Na₂O is used as a flux in preparing glasses because it acts in the glassy network as a modifier making some disruptions in the network by creating more NBOs which in turn enhancing the melting process [10] as well as the tendency of lithium and sodium ions in enhancing the conducting properties of the glass [11, 12]. Also, it was stated by some authors [13–15] that adding BaO, SrO or PbO to Bi₂O₃ glasses gives them considerable technological importance due to enhancing their optical, mechanical, electrical and thermal properties. For instance, BaO and SrO work as modifiers and occupy the interstices within the glassy network [16]. However, lead oxide (PbO) is unique in its influence on the glass structure because it enhances the resistance against devitrification cation, improves the glass chemical durability and lowers the melting temperature. PbO can also demonstrate a dual role; it may act as a modifier or as former, giving such structural units as PbO₄ and/or PbO₃ according to its concentration in the glass matrix [16–18].

Introducing of an alkali species or modifier oxides to the glass should alter the glass density values according to the concentration of the introduced ions and the way of their interconnection with other glass components besides the composition of the host glass matrix [19, 20]. Also, occupying modifying ions the interstices in the glass network causes the creation of more non-bridging oxygens (NBOs) then a shift of the UV bands to higher wavelengths would take place.

Electrical conduction in almost alkali oxide containing glasses depends essentially on the moving of alkali ions present in the interstitial positions in the glassy network. In marketable glasses the most common conducting species are Na⁺ and Li⁺ ions that are relatively mobile and have the tendency to carry current through the glass matrix, so their presence should enhance the electrical properties of the glass [21].

The response of glasses to gamma radiation is correlated to the rate of formation and gathering of induced defects through the progressive irradiation process. The most common point defect is the oxygen-excess centers, which are found in two types; one of them is (NBO) and the other involves two NBOs on the same silicon according to the following chemical reaction [22]:



In the present study three compositions of lithium bismuth silicate glasses doped with either 2% PbO, SrO or BaO were prepared. The effect of gamma radiation on some of their physical, optical and electrical properties is also studied and discussed e.g. density, oxygen packing density, molar and specific volumes, optical UV absorption, optical energy

gap, electrical conductivity and dielectric measurements. All last parameters were investigated to examine the possibility of using glasses as semiconductors or radiation shielding materials.

2 Experimental details

2.1 Preparation of the glasses

Three compositions of lithium bismuth silicate glasses with the composition; 68% SiO₂, 20% Bi₂O₃, 8% Li₂O₃, 4% Na₂O doped with either 2% PbO, BaO or SrO (in wt%) were prepared from chemically pure materials of SiO₂ and Bi₂O₃ and analytical reagent grade quality of Li₂CO₃, Na₂CO₃, PbO₂, BaCO₃ and SrCO₃ from Sigma Aldrich Company.

The glasses were prepared by melting the appropriate and accurately weighed raw materials of each glass batch according to its chemical composition. Then they were mixed together and melted in platinum 2% rhodium crucible, placed in an electrically heated furnace at a temperature range from 1000 to 1100 °C for 3 h. The glass melt was stirred by rotating the crucible after each hour for attaining homogeneity and removing air bubbles. The glass sample was then cast in stainless steel molds and immediately annealed in a muffle furnace at a temperature range from 350 to 500 °C. The furnace was left over night to cool to the room temperature at a rate of 30 °C/h. Samples were then cut in dimensions of 1 × 1 × 0.2 cm³ to be suitable for each required measurement.

2.2 Density measurements

Density was measured at the room temperature based on Archimedes principle where xylene was used as the immersion liquid. All density measurements were made three times with a maximum error ± 0.001 g/cm³ and the density value was calculated according to the following formula:

$$\rho = \{a/a - b\} \times 0.86 \quad (2)$$

where ρ is the density of the glass sample, a and b are weights of the glass sample in air and in xylene respectively, and 0.86 is the density of xylene at 20 °C.

The molar volume was calculated using the formula:

$$V_m = \left(\sum X_i \cdot M_i \right) / \rho \text{ cm}^3/\text{mol} \quad (3)$$

where X_i is the molar fraction, M_i is the molecular weight of each component in the glass sample and ρ is the density of the glass.

The oxygen packing density OPD was also calculated by using the following formula:

$$\text{OPD} = \rho / M \times C \text{ g atm/l} \quad (4)$$

where M is the molecular weight of the glass sample and C is the number of oxygen atoms per unit formula.

Density, oxygen packing density, molar and specific volumes were all measured before and after successive gamma radiation doses.

2.3 UV optical absorption spectra

The optical absorption spectra measurements were carried out in the UV and visible range by using a recording double-beam spectrophotometer (Type JASCO Corp, v-570, Rel-100 Japan). The spectrophotometer was covering the range from 200 to 1000 nm. The measurement of the three investigated glasses was carried out before and after their irradiation with gamma rays.

2.4 Estimation of the optical band gap

The energy band gap (E_{opt}) values of amorphous solids were calculated through the relation given by Mott and Davis [23]:

$$\alpha h\nu = B(h\nu - E_{opt})^n \quad (5)$$

where; (E_{opt}) is the optical energy gap, (α) is the absorption coefficient, (h) is Planck's constant, (ν) is the radiation frequency, (B) is an energy independent constant and (n) is an index having values of 1/2, 3/2, 2 or 3 depending on the type of electronic transition.

2.5 Electrical conductivity measurements

The electrical conductivity measurements were performed by using a locally constructed measuring system that was connected with an electronic temperature controller. For this purpose, the samples were prepared in disc shapes and their surfaces were carefully polished and rubbed with a silver paste to give a good contact with copper cell by using

two copper wires. A programmable digital electrometer/high resistance meter (KEITHLY) was used in measuring by detecting resistivity values at constant current and voltage with a reliable fast response together with the power supply. All measurements were carried out in different temperature range from 298 to 453 K for each 20° and the electrical conductivity values were calculated from the following relation:

$$\sigma = -\log(1/\Omega) \quad (6)$$

where σ is electric conductivity ($\text{ohm}^{-1} \text{cm}^{-1}$) and Ω is the resistivity (ohm).

The measuring was taken place also before and after successive gamma irradiation of the investigated glasses.

2.6 Dielectric measurements

The dielectric properties were collected using computer assisted digital type HIOKI 2628 LCR bridge with a reliable fast response together a power supply KEITHLY 2635 A. The dielectric properties of the investigated glasses were measured in frequency range from 5 Hz to 5 MHz at a temperature range from 25 to 150 °C.

2.7 Gamma irradiation

A Co^{60} gamma cell (2000 Ci) was used as a gamma ray source with a dose rate of 1.5 Gy/s at 30 °C. The glass samples were located into gamma cell in means that every sample was exposed to the required identical dose.

3 Results and discussion

3.1 Density and molar volume

Results presented in Table 1 show that glass containing Pb^{2+} ions has density value = 2.85 g/cm³ with molar volume = 80.38 cm³/mol while Ba^{2+} and Sr^{2+} ions containing

Table 1 Characteristic parameters of the investigated glasses before and after gamma irradiation

Glass no.	Density ρ (g/cm ³)	Molar volume V_m (cm ³ /mol)	Specific volume (cm ³ /g)	Oxygen packing density OPD (g atm/l)	E_{opt} values (eV)
(2%PbO) 0 kGy	2.8555	80.38	0.3502	28.11467	3.16
(2%BaO) 0 kGy	2.7741	82.24	0.3605	27.4806	3.24
(2%SrO) 0 kGy	2.7262	83.32	0.3668	27.1242	3.36
(2%PbO) 50 kGy	2.8996	79.16	0.3449	28.5488	2.95
(2%BaO) 50 kGy	2.8885	78.98	0.3462	28.6138	2.73
(2%SrO) 50 kGy	2.8803	78.86	0.3472	28.6574	3.13
(2%PbO) 100 kGy	2.8878	79.48	0.3463	28.4326	1.93
(2%BaO) 100 kGy	2.8809	79.19	0.3471	28.5385	1.39
(2%SrO) 100 kGy	2.8779	78.93	0.3475	28.6335	1.95

glasses have density values = 2.77 and 2.72 with molar volumes values = 82.24 and 83.32 cm³/mol respectively. So it is obviously shown that lead oxide containing glass has the highest density and the lowest molar volume than glasses containing either barium or strontium oxides which reflects directly the presence of less vacancies and defects in this glass (Pb²⁺) and then its the internal compact structure among other glasses [16]. According to values listed in Table 1, glass containing lead oxide has also the lowest specific volume which gives a clue of more compaction in its structure. On the other hand it has the highest oxygen packing density which represents the fraction of space that filled with greatest possible oxygen density [24, 25]. Consequently, it can be assumed that this glass has the highest amount of bridging oxygens. While glass containing barium ions comes in the second order and that containing strontium ions comes in the third order having the lowest density and the highest specific and molar volumes which means that it has the highest amount of vacancies.

3.1.1 Effect of gamma radiation on density and molar volume of glasses

Table 1 shows that the values of density were all increased after gamma irradiation where the density of Pb-glass was increased from 2.85 g/cm³ before radiation to 2.89 at 50 kGy and 2.88 g/cm³ at 100 kGy. However, densities of Ba and Sr-glasses were increased from 2.77 and 2.73 g/cm³ before irradiation to 2.89 and 2.88 g/cm³ at 50 kGy and to 2.88 and 2.87 g/cm³ at 100 kGy respectively. The other parameters e.g. molar volume, specific volume and OPD were also changed after irradiation as shown in Table 1. As it was obvious from the table, the density values of all the investigated glasses were increased with irradiation with either 50 or 100 kGy comparing with their values before irradiation. However the rate of increase in density was slightly inhibited at the higher dose (100 kGy). This behavior may be due to the free radicals resulted from the irradiation process at the lower dose (50 kGy) that can cause only helation to the relatively small amount of defects found in the glass, while by increasing the irradiation dose to 100 kGy, the glass density is relatively decreased due to the presence of more free radicals, creating more defects and NBOs or the possible atomic displacements produced by γ -collisions within the glass structure that may significantly alter stresses in the glass network and then changes the density values [26]. In addition, the oxygen packing density was increased at 50 kGy for all glasses and return to a slight decrease at the higher dose (100 kGy) which is agreed with the density results and directly related to the formation of more non-bridging oxygen in the glass matrix at such high dose [27].

The overall view obtained by comparing results shown in Table 1, is that Pb- glass is less affected by gamma radiation

than Ba- and Sr- glasses which give a clue of more compactness in its structure and then reflects the higher stable behavior of glass containing Pb²⁺ ions than those containing either Ba²⁺ or Sr²⁺ ions.

3.2 UV absorption spectra

The optical absorption spectra of glasses at the room temperature before irradiation is shown in Fig. 1 where there are small observed peaks in the UV region 230–390 nm that can be attributed to the iron impurities of the raw materials used in glasses preparation, especially triplet ionic states of iron (Fe³⁺). From the figure it can be also noticed that all examined glasses have a cutoff at about 380 nm which is slightly shifted to a higher wavelength for glass containing lead oxide. It was concluded by Abdel-khalek and Bahgat [28] that the wavelength of the cutoff of the glass is related to the amount of non-bridging oxygens NBO present in its structure. It can be also observed from Fig. 1 that all of the examined glasses do not have sharp absorption edge which indicates the amorphous natures of the investigated glasses [29].

3.2.1 Effect of gamma radiation on optical UV absorption spectra and optical E gap

Figure 2 shows the optical UV absorption spectra of the three investigated glasses before and after three gamma radiation doses 50, 80 and 100 kGy. Figure 2 shows obviously that after irradiating glasses there are some neglected changes in the peak positions accompanied with a small

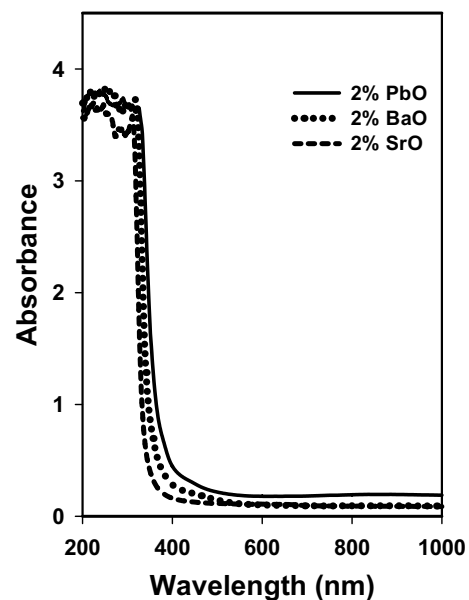


Fig. 1 UV–Vis absorption spectra of the prepared glasses

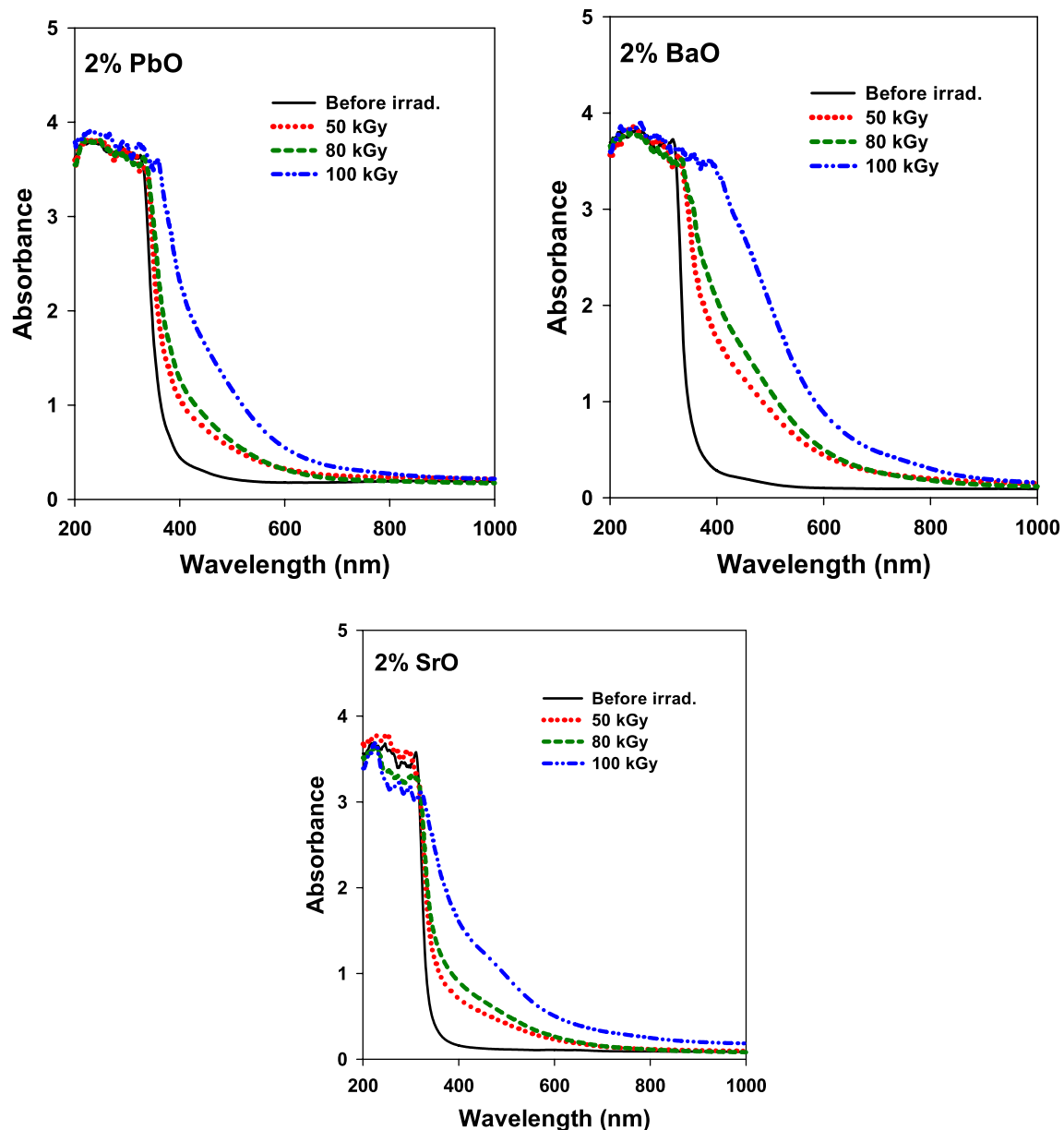


Fig. 2 UV–Vis absorption spectra of the glasses before and after gamma irradiation doses

increase in the absorbance intensity and a shifting of the cutoff to longer wavelengths with the increase of radiation dose which is related to the formation of more non-bridging oxygens because of the effect of radiation [30, 31]. Subjecting glasses to gamma radiation affects directly the host glass matrix by changing the number of NBO or the formed extrinsic defects. Therefore the observed induced absorption may be recognized also to some photochemical reactions of the present transition metal TM ions in the glass (Fe^{3+} ions) even if they were found in p.p.m concentrations according to Mo'ncke and Ehrh [32].

The optical band gap (E_{opt}) values of the studied amorphous glasses were calculated by drawing the tauc plot from

the observed absorption edge between $(\alpha h\nu)^{1/2}$ as a function with $h\nu$ as given in Eq. (5) and Fig. 3. The optical band gap E_{opt} values are ranged from 1.39 to 3.36 eV as listed in Table 1. E_{opt} represents the energy gap between the valence and conduction bands [33], so it depends mainly on how the glass structure is compacted because in the compacted network the excited electrons are more tightly bonded leading to high optical band gap E_{opt} [28]. Although the conduction band depends mainly on the glass anions however, cations have a significant indirect effect due to the change in the number of NBO.

It is obvious from Fig. 3 and Table 1 that Pb- glass have relatively lower E_{opt} values than Ba or Sr- glasses this is

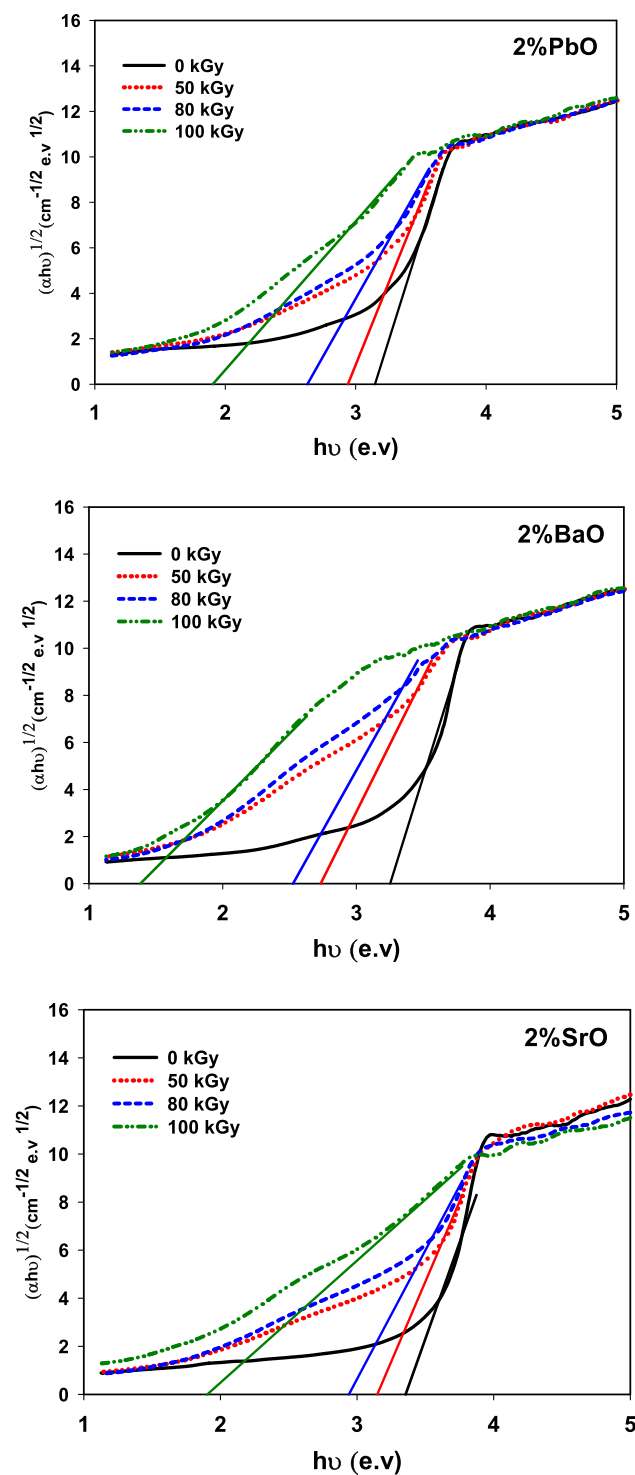


Fig. 3 Optical band gap E_{opt} of the prepared glasses before and after gamma irradiation doses

because Pb^{2+} ions have higher atomic weight than Ba^{2+} and Sr^{2+} ions and then higher polarizability. The electronic oxide polarizability depends on the amount of repulsion between PbO , BaO or SrO with Li_2O and Na_2O presuming that the

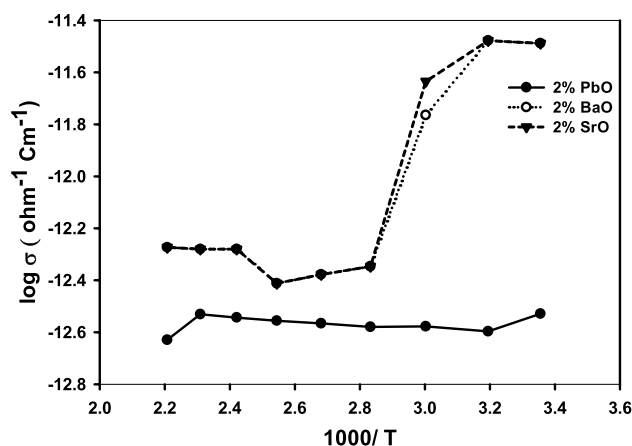


Fig. 4 Dependence of electrical conductivity on temperature of the prepared glasses

lone pair repulsion of ions with large atomic configuration [31] is smaller than that of the repulsion of ions with smaller atomic configuration. In other words the lone pair present in the valence shell of Bi^{3+} ions can be easily compensated in the presence of Li_2O or Na_2O , but the excess of Bi^{3+} ions and Pb^{2+} ions which are the highly polarizable ions can cause more polarization in the glass structure more than Ba^{2+} and Sr^{2+} ions that are less polarizable. Consequently, although Pb^{2+} ions may act in the glass network as intermediates causing more connectivity of the structure by enhancing the glass matrix, glass containing Pb^{2+} ions has relatively lower E_{opt} because of the relatively high polarizability of Pb^{2+} ions that makes the transition of free electrons easier from the valence to the conduction band causing the slight decrease in E_{opt} .

It is observed from Fig. 3 and Table 1 that E_{opt} values are decreased with increasing the dose of gamma radiation in the three investigated glasses. Since the performance of the optical band gap is directly related to the structural changes occurred in the glass network [25, 34, 35] so any changes carried out in the glass structure because of radiation would significantly affect its E_{opt} values. Previously it was assumed by Mott and Davis [23] that when the connected bridging oxygens in the glass network are converted into non-bridging oxygens, there should be a decrease in the optical band gap. Consequently defects caused by irradiation in the glass network causes an increase in the localized states concentration in the band structure by increasing NBO then more electronic transitions would take place leading to a lowering in E_{opt} values [36].

3.3 Electrical conductivity

Figure 4 shows the dependence of electrical conductivity on temperature for all the investigated glasses. From the

figure it can be observed that all glasses have low electrical conductivity and glass containing Pb^{2+} ions has the lowest conductivity which is almost stable even with increasing temperature. This result is supported by the density, UV and optical band gap results, where the results resolved that glass containing lead ions has the lowest NBO and the most tightly bonded excited electrons where the electrical conduction depends on the hopping motion of polarons separated from these tightly bonded electrons. It can be also taken into consideration that these small charge carriers (polarons) can be trapped by self-induced lattice distribution depending on the structural relaxation. In other words it can be said that these polarons are ejected from the same electrons that can be separated and producing conduction by hopping through the glass matrix, but they were captured by Li^+ or Na^+ ions, so the resulted low conductivity is predicted [37].

For glass containing either Ba^{2+} or Sr^{2+} ions, it can be observed from the figure that they have higher conductivity than glass containing Pb^{2+} ions, this may be due to the presence of more NBO and more holes which make the hopping of polarons easier. It can be also observed that the relation between increasing temperature and electrical conductivity

has three stages; the first stage is rapid where the decrease of the conductivity of glasses with the increase of temperature may be due to consuming of the energy gained from the raised temperature in healing these holes which in turn causes a decrease in the conduction [38]. It can be also noticed that the state of healing is high at the beginning of raising temperature, then reduced to a slow helation rate indicated by the very low decrease in the conductivity with the increases of temperature as shown in the second stage. At the third stage there is an increase in the conductivity with the increase of temperature which can be attributed to the increasing of energy to be able to eject more polarons and the presence of new holes which permits the hopping of these polarons.

3.3.1 Effect of gamma radiation on the electrical conductivity

Figure 5 shows the electrical conductivity values of the three investigated glasses after being irradiated with 50, 80 and 100 kGy. It is observed from the figure that the electrical conductivity values decreased when the glass samples were

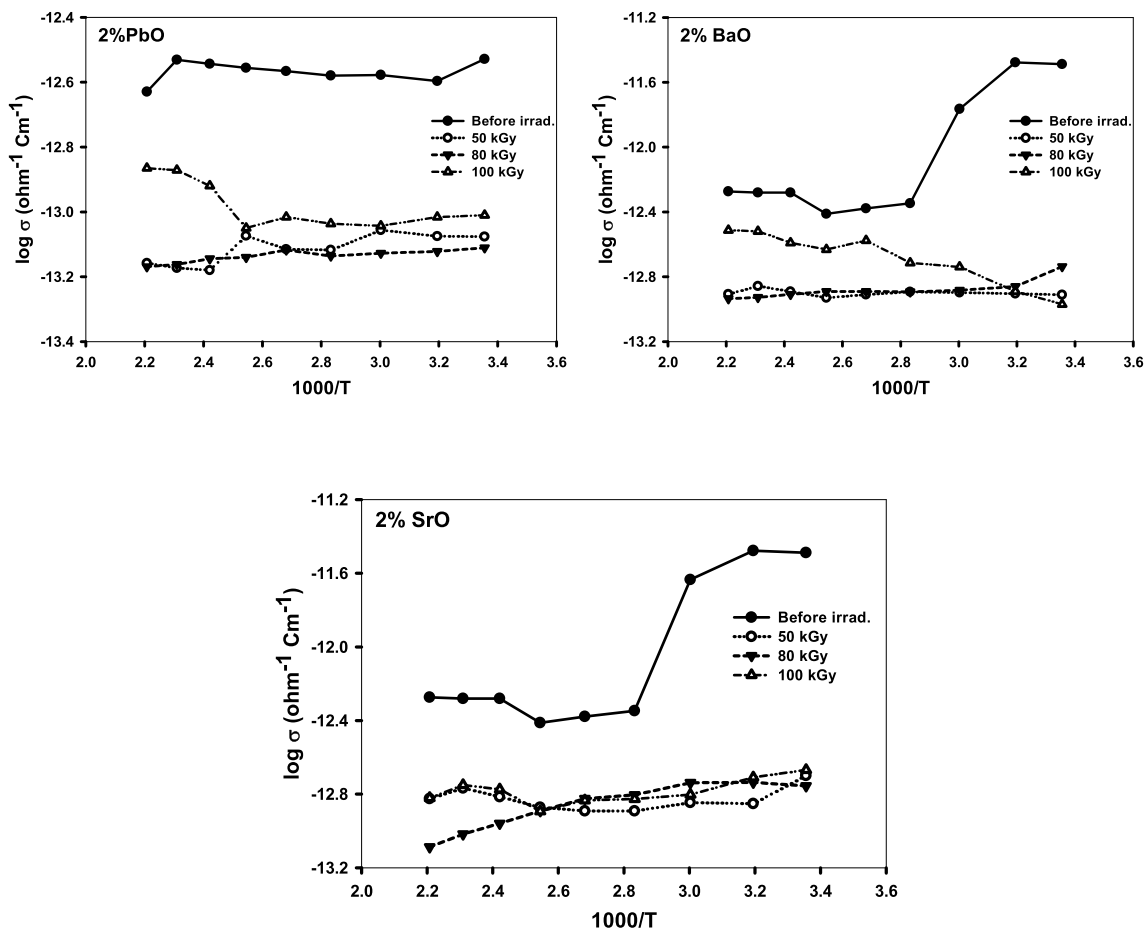


Fig. 5 Dependence of electrical conductivity on temperature of the prepared glasses before and after gamma irradiation doses

irradiated however, the dose of 50 kGy gives lower electrical conductivity values than the dose of 100 kGy. This behavior can be related to the glass density results and the other parameters tabulated in Table 1. Where density values of glasses at 50 kGy is higher than those at 100 kGy because at the last dose the complete of the helation process occurred by the ejected free radicals, while the excess of these radicals begin to cause more defects in the glass structure either holes or non- bridging oxygens. Therefore, the number of oxygen packing density were decreased however molar and specific volumes were increased comparing with their values at 50 kGy. This behavior can also explain the slight increase in electrical conductivity values at 100 kGy than those at 50 kGy where the presence of more defects, holes or NBO gives the chance for the charge carriers such as Na^+ and Li^+ ions to move more freely and carry current easier causing the observed increase in electrical conductivity values.

3.4 Dielectric properties

Figure 6 shows the variation of dielectric constant Σ' with temperature at different frequencies for the three investigated glasses, it can be seen that Σ' increases with the increase of both temperature and frequency.

From the results, it can be observed that glass containing Pb^{2+} ions has the highest dielectric constant; this indicates that this glass has the largest degree of lattice distortion which results in large proportion of space charge polarization [39]. The increase of dielectric constant with temperature can also be attributed to the ability of higher temperature to weaken the molecules/atoms binding forces which give them more freedom to increase their vibration resulting in an increase in the polarization [40]. This is because the dipolar polarization is affected by the intermolecular forces so it enhances the orientational vibration [41].

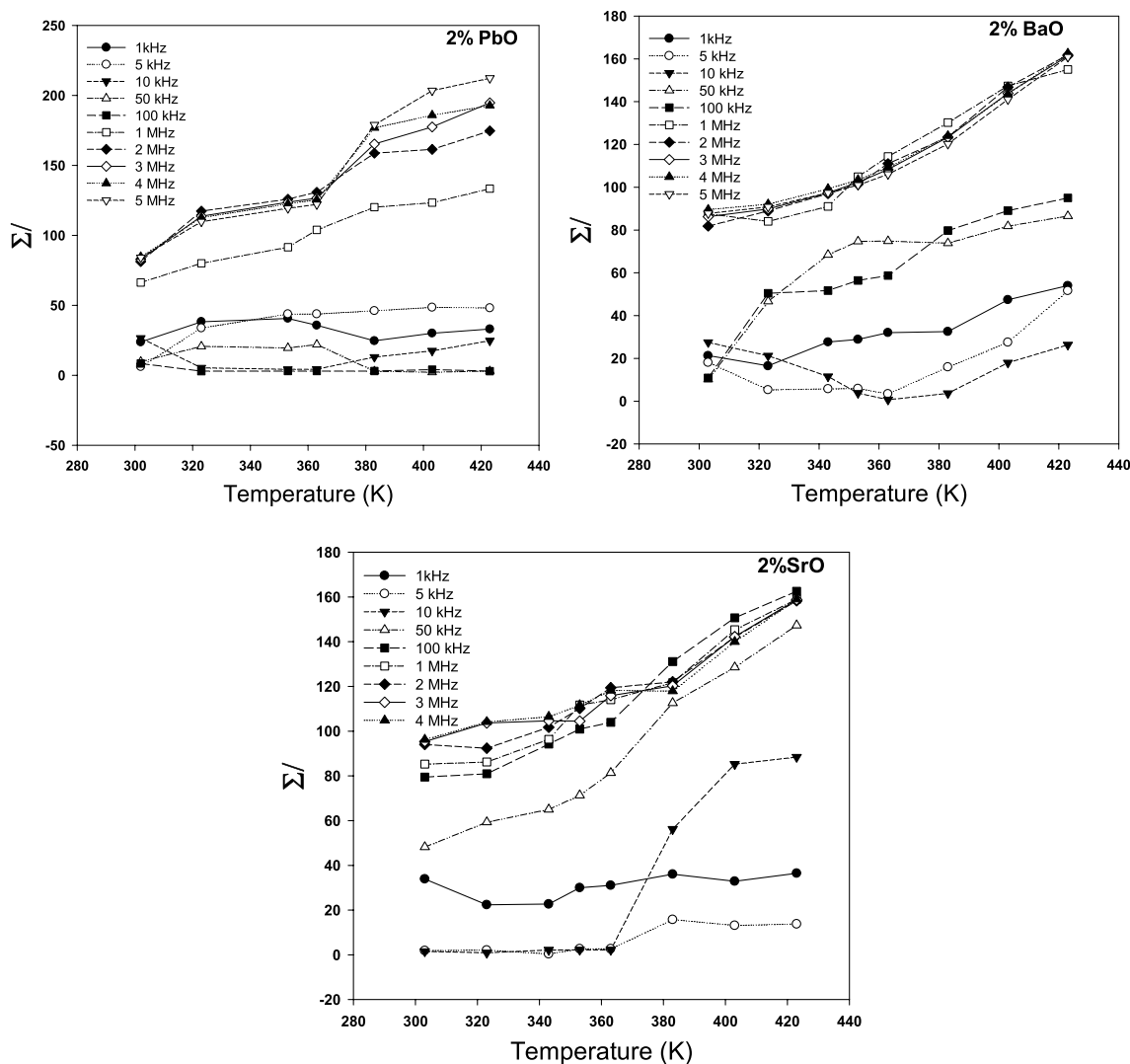


Fig. 6 Variation of dielectric constants Σ' with temperature at different frequencies for the three investigated glasses

It is common for most glasses that the dielectric constant decreases with the increase of frequency. This is due to the decrease of polarizability contribution where the ionic sources and orientation become completed due to the ions inertia. However in our case, it is observed from Fig. 6 that the dielectric constant increases with the increase of frequency for all the examined samples. This may be attributed to the increase of the amount of distortion and polarization inside the lattice to act as separated electrical circuits causing the observed increase in \sum'' with the increase of frequency.

Figure 7 shows the dielectric loss of the three investigated glasses which represents the migration of ions at low frequencies, while at high frequencies, the contribution of jumping ions, conducting ions and polarization loss is rising. From the figure, it can be observed that glass containing

Pb^{2+} ions has the highest dielectric loss than the other two glasses which is agreed with the above previous results.

4 Conclusion

In this work, some physical and electrical properties of lithium bismuth silicate glasses containing 2% of PbO , BaO or SrO were studied. Also, the effect of gamma radiation on these properties was also discussed. The results indicated that glass containing PbO has the highest density and oxygen packing density and the lowest molar and specific volumes. Also the density values of the glasses were increased at 50 kGy but decreased at 100 kGy because the free radicals resulted from the irradiation process at 50 kGy can cause only relation to the amount of defects found in the

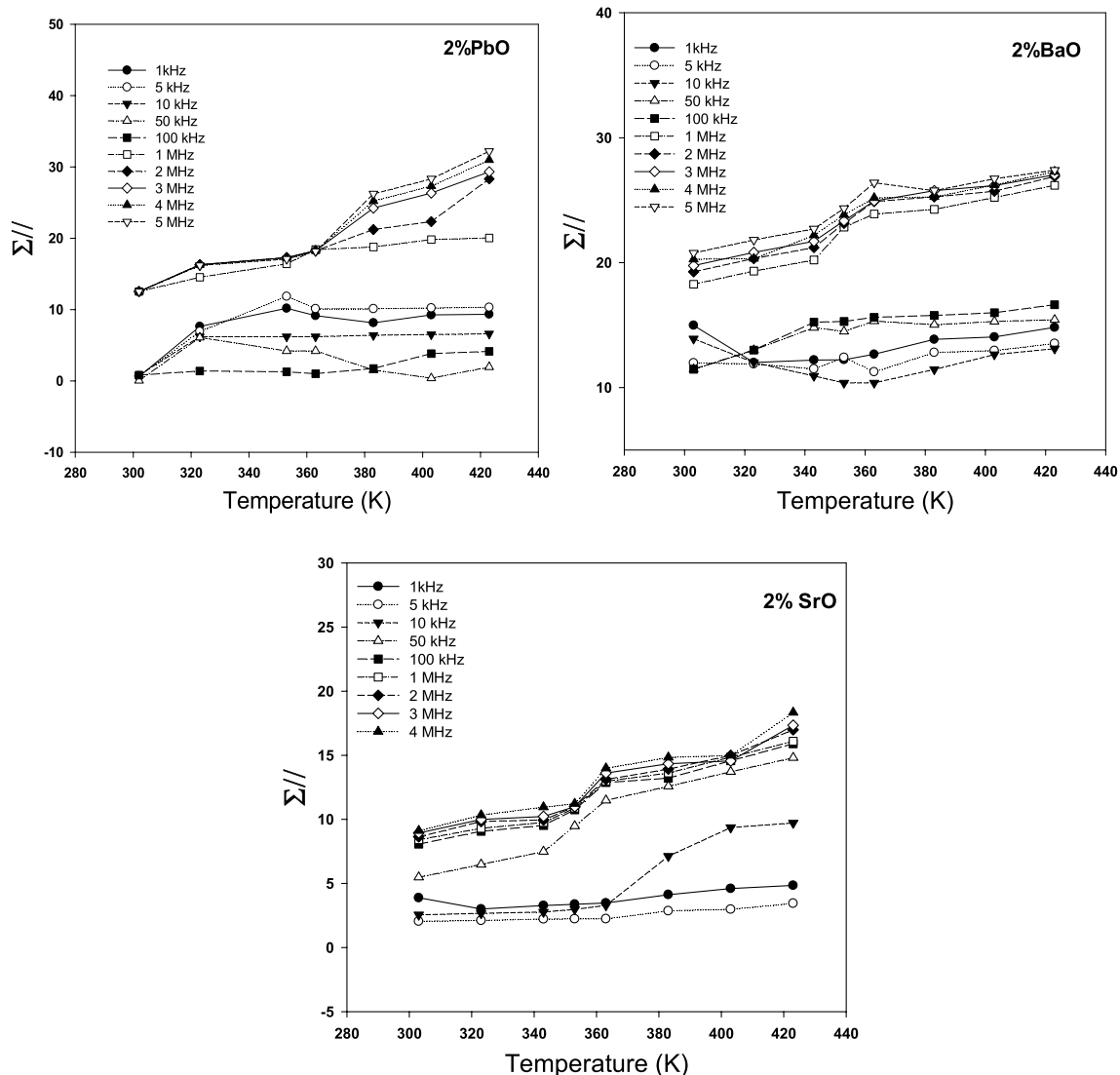


Fig. 7 Variation of dielectric loss Σ'' with temperature at different frequencies for the three investigated glasses

glass however, at 100 kGy the glass density was relatively decreased because of creating more free radicals and NBO at such high dose. The UV results showed the appearance of small peaks at 230–390 nm related to Fe^{3+} impurities of the used raw materials with no visible bands and a cutoff at about 380 nm which was shifted to a higher wavelength with irradiation indicating the formation of more non-bridging oxygens. Optical band gap values revealed a decrease after irradiation because of increasing in the localized states concentration in the band structure allowing more electronic transitions to take place. Pb- glass has lower E_{opt} than Ba^{2+} and Sr^{2+} glasses although it has the most compacted structure because of its high polarizability which makes the transition of free electrons easier from the valence to the conduction band. The electrical conductivity results showed an obvious decrease with irradiation in the three glasses; however the dose of 50 kGy gives lower electrical conductivity values than the dose of 100 kGy agreeing with the density results because at the highest dose (100 kGy) a complete of helation process takes place by the ejected free radicals. Pb-containing glass has the lowest electrical conductivity values which were almost stable even with the increase of temperature since it has the highest polarizability where the ejected polarons responsible for electrical conductivity were captured by either Li^+ or Na^+ ions however the polarization of both Bi^{3+} and Pb^{2+} ions increased with either increasing of temperature or frequency. This may indicate that Pb^{2+} ions tend to occupy the network forming positions rather than modifying ones. Also Pb- containing glass has the highest values of dielectric constant at different frequencies because of increasing the amount of distortion and polarization inside the lattice to act as separated electrical circuits causing the observed increase in \sum^j with the increase of frequency.

From all the last results; it can be suggested that the investigated glass compositions are good shielding materials for gamma radiation especially the glass containing Pb^{2+} ions which is less affected by irradiation than Ba^{2+} and Sr^{2+} glasses because of its highly compacted structure and the high polarizability of the heavy lead ions.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

Ethical approval This article does not contain any studies with human participants or animals performed by any of the authors.

Informed consent Informed consent was obtained from all individual participants included in the study.

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