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Gamma rays interactions with CdO-doped lead silicate glasses

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Abstract
Lead silicate glass systems with different concentrations of cadmium oxide had been prepared and characterized. The XRD analysis indicated the all samples in glassy state. The FTIRpectroscopy indicated a glass modifier role of CdO that was deduced from the formation of [CdO_6] structural units. The compositional dependence of the physical parameters such as the density, the molar volume, the optical band gap, the ultrasonic velocities and the elastic moduli on CdO content were determined. The decrease of the ultrasonic velocities, the elastic moduli (experimentally determined and theoretically computed according to Makishima–Mackenzie model) with the addition of CdO was attributed to the decreased compactness and rigidity. These physical parameters revealed the glass modifier role of cadmium oxide.

Keywords CdO · Silicate glass · Gamma irradiation · Elastic moduli

1 Introduction

Nowadays, the properties of transition metallic ions (TMI) doped lead silicate glasses are interseted. A glasses which having semiconducting properties are alienated into companies, as transition metal oxide (TMO), these glasses containing (TMI) as cadmium, copper, etc. Glasses containing an excessive content(TMI)are digital electrodes (Sharma et al. 2012). The silicate glasses are fairly beneficial as they are extremely informal to prepare, this is due to structural units, in silicate glasses SiO_2 a primary make as former due to better bond strength, lesser cation extent. Transition metal oxide are incorporated into silicate glasses to improve optical behaviors. Lead oxide combined with SiO_2 with high value to form stable glass (El-Batal et al. 2012). Worrel and Henshall (1978) studied PbO doped with glasses, acts as a former and modifier. PbO is a heavy metal oxide (HMO) it used for shielding properties (Ben Kacem et al. 2017). Lead silicate glasses are very interesting

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Table 1 Chemical composition of the prepared glasses, (mol%)

Sample name	Chemical composition (mol%)		
	PbO	SiO_2	CdO
G1	30	60	10
G2	30	50	20
G3	30	40	30
G4	30	30	40
G5	30	10	60

because PbO is one of the elements which form a glass with extensive excellent of SiO_2 content. These glasses have now been examined by numerous research groups by spectroscopic properties (Ben Kacem et al. 2017). The structural study of cadmium silicate glasses has been created; these days because of the improvement of positive glass structures most of the cadmium silicate glasses that show off ohmic photoconductivity. The investigation on cadmium lead silicate glasses is interesting due to the presence of two networks forming oxides. Cadmium oxide when mixed with SiO_2 the formed cadmium silicate glasses contains Cd^{2+} ions acting as a modifier or as former. It had two different oxidation states, so it formed two types of structural units when co-existed with a network forming oxides, namely, CdO_6 that had a glass modifier role and CdO_4 that had a glass former role. The cadmium lead silicate glass gadget containing adifferent mol% of CdO is uses transducers for numerous functions. However, a nevertheless higher attention of CdO inside the cadmium silicate glasses is required to make it beneficial as piezoelectric transducers (Bahammam et al. 2017; Semin et al. 1989; Lipovskii et al. 1997; Niu et al. 2011; Hivrekar et al. 2017; Saddeek et al. 2018). A few authors studied the impact of a single (Sundar Rao et al. 2014) and more than one (Pavani et al. 2011; Kaur et al. 2013; Fayad et al. 2017) TM (transition metallic) ions as a dopant in alkali and alkaline earth oxide glasses (Abd-Allah and Nabhan 2015). Lately, Rao et al. (2006) stated the research to CdO substituted glass systems with similar compositional variations. Thus, the goal of this study is to report on the synthesis of a wide range glasses of 30 PbO–x CdO–$(70-x)$ SiO_2; where $(10 \leq x \geq 60)$, mol%. Moreover, a combined study with spectra of FTIR and UV before and after γ radiation besides and mechanical parameters of these glasses will be performed.

2 Experimental procedures

The 30 PbO–x CdO–$(70-x)$ SiO_2; where $(10 \leq x \geq 60)$, mol%. Glasses were prepared via conventional melting–quenching technique. The nominal chemical compositions with the respective codes of glasses are summarized in Table 1. The used raw materials are Analar SiO_2, PbO and CdO with 99.9% purity and grounded with each other to get a homogeneous mixture of each sample. The specified quantities of the mentioned chemicals were mixed completely and then contained in a ceramic crucible. The powder samples were melted in an electrically heated furnace at about 1100 °C for 1 h and the melt was then cast onto preheated stainless-steel mold, which was transferred immediately to the annealing furnace adjusted at 400 °C for one hour. The obtained glasses were lapped, and two opposite sides were polished to be suitable for the use in the ultrasonic velocity and UV measurements.

X-ray diffraction patterns were obtained by using Philips X-ray diffractometer (PW/1710 with Ni-filtered Cu-Kα and radiation λ = 1.542 Å).

The FT-IR spectra were measured before and after gamma irradiation at ordinary temperature in the range 4000–400 cm^{-1} by a Fourier Transform infrared spectrometer (type Bruker's VERTEX 70 FTIR Spectrometers). Absorbance A (λ) before and after gamma irradiation at ordinary occurrence for the prepared glasses have been documented at room temperature inside the variety 200–900 nm the use of a computerized double beam spectrophotometer, type JUSCO V-670. Gamma irradiation process takes place by using Co^{60} gamma cell (2000 Ci) as a gamma ray source with a dose rate of 1.5 Gy/s at 30 °C. The glass samples were placed in the gamma cell in the manner that each sample was subjected to the same gamma dose.

The densities (**ρ**) of the studied glassesis determined by the Archimedes code according to

$$\rho = \left(\frac{a}{a-b}\right)0.86 \tag{1}$$

where **a** is the weight of the glass sample in air, **b** is the weight of the glass sample when immersed in xylene of density 0.865 g/cm^3.

Molar volume $\boldsymbol{V_m}$ of prepared glasses, is determined by

$$Vm = \frac{M}{\rho} \tag{2}$$

where M the molecular weight of glass constituents.

The ultrasonic measurements were performed at room temperature by a system consisted of the Echo-graph (Krautkramer model USM3 pulsar/receiver instrument) and two transducers. The operated frequency was adjusted to 4 MHz. The was used for the determination of the longitudinal (v_L) and shear (v_T) velocities. Random errors in the measurements of the velocities were ± 10 m/s. The two velocities besides the density (Saddeek et al. 2018; Shaaban et al. 2018) were utilized in determining the

$$\text{Longitudinal modulus, } L = \rho v_l^2 \tag{3}$$

$$\text{Shear modulus, } G = \rho v_s^2 \tag{4}$$

$$\text{Poission's ratio } \sigma = \frac{1}{2} - \frac{v_T^2}{2\left(v_l^2 - v_T^2\right)} \tag{5}$$

$$\text{Young's modulus } Y = (1+\sigma)2G \tag{6}$$

$$\text{Bulk modulus, } K = L - \left(\frac{4}{3}\right)G \tag{7}$$

2.1 Makishima–Mackenzie's model

The glasses elastic moduli can be investigated using the model suggested by Makishima and Mackenzie (1973, 1975) that based on the G_i of bonds and V_i.

$$Vi = \left(\frac{3\pi}{4}\right) NA\left\{m\mathrm{R}_{\mathrm{A}}^{3} + n\mathrm{R}_{\mathrm{O}}^{3}\right\}\left(\frac{\mathrm{m}^{3}}{\mathrm{mol}}\right) \tag{8}$$

where R_A and R_O are the metallic and oxygen Pauling ionic radii.

The glass shear (G) and longitudinal (L) coefficients are:

$$L = K + \left(\frac{4}{3}\right)G \tag{9}$$

$$G = \frac{a(Vi^{2}Gi)}{(bVi)} \tag{10}$$

where a and b are constants. Then Poission's ratio according to Makishima–Mackenzie model was:

$$\sigma = \frac{1}{2} - \left(\frac{1}{7.2 * Vi}\right) \tag{11}$$

$$\text{Acoustic Impedance; } Z = v_L \rho \tag{12}$$

$$\text{Micro Hardness; } \mathrm{H} = \frac{(1-2\sigma)\mathrm{Y}}{6(1+\sigma)} \tag{13}$$

$$\text{Debye Temperature; } \theta_D = \frac{h}{k}\left(\frac{9N}{4\pi V_m}\right)^{\frac{1}{3}} M_s \tag{14}$$

where h and K are the Planck's and Boltzmann's constants and N is the Avogadro's number. The average speed of sound M_s was given by:

$$M_s = \frac{1}{3}\left(\frac{\frac{2}{v_T^3}}{\frac{1}{v_l^3}}\right)^{\frac{1}{3}} \tag{15}$$

And the coefficient of thermal Expansion (α_P) is;

$$\alpha_{P=23.2(v_L-0.57457)} \tag{16}$$

Both the oxygen molar volume (V_O) and packing density (OPD) have been calculated using the equations:

$$V_O = \left(\frac{M}{\rho}\right)\left(\frac{1}{\sum xini}\right) \tag{17}$$

$$OPD = \left(\frac{1000C}{Vm}\right)\left(\frac{Mol}{L}\right) \tag{18}$$

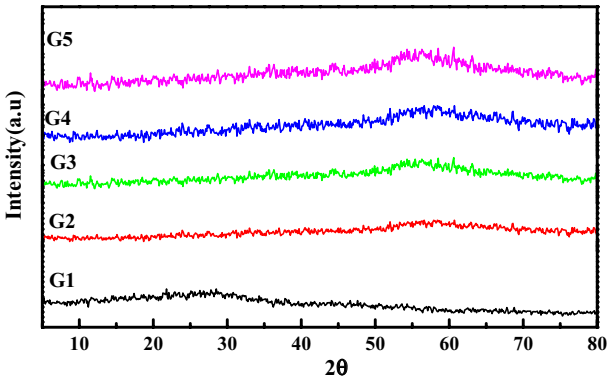

Fig. 1 XRD of the studied glasses

3 Results and discussion

Figure 1 curves confirmed XRD of the prepared glasses no distinguished hint, no sharp lines which approve the glassy nature.

3.1 FTIR analysis

FT-IR analysis is a useful method for confirming the structure and dynamics of glasses. The FTIR spectra of the present glassy system are represented in Fig. 2a–d. The IR assignments for the studying samples in Table 2.

3.1.1 Effect of composition on infrared spectra

The FTIR spectra of the present glassy system were represented in Fig. 2a. Shows main absorption that peaks at around ~ 465–477 cm^{-1}, ~ 716–722 cm^{-1}, ~ 890–990 cm^{-1}, ~ 11 24–1255 cm^{-1}, 1300–1560 cm^{-1}, 1650–1750 cm^{-1}. These bands listed in Table 2. The assignments of FTIR analysissummarized as the following:

(a) 465–477 cm^{-1} is ascribed to bending vibrations of Si–O–Si linkages (Khalil et al. 2010).
(b) 560–617 cm^{-1}isassociated with Pb–O stretching vibration in PbO_4 structural units (Khalil et al. 2010). Also, a small band at about 660 cm^{-1} is apportioned to Pb–O symmetrical bending Vibrations
(c) 716–722 cm^{-1} is due to symmetric stretching vibration of O–Si–O bonds (Abou Hussein and El-Alaily 2018) and a small bands at 848–883 cm^{-1} can be associated to Pb–O bond vibrations of PbO_n units with n = 3 or 4 (Saddeek et al. 2010).
(d) 890–970 cm^{-1} can be associated to the Si–O stretching mode of non-bridging oxygen's (Hwang et al. 2004).
(e) A band at about 1124–1255 cm^{-1} is detected, which is associated to antisymmetric stretching vibrations of BO's (Baccaro et al. 2007).
(f) 1300–1560 cm^{-1} is associated to Si–O–Si antisymmetric vibrations of bridging oxygen's (Rao et al. 2006).
(g) 1650–1750 cm^{-1} is associated to molecular water (Khalil et al. 2010).

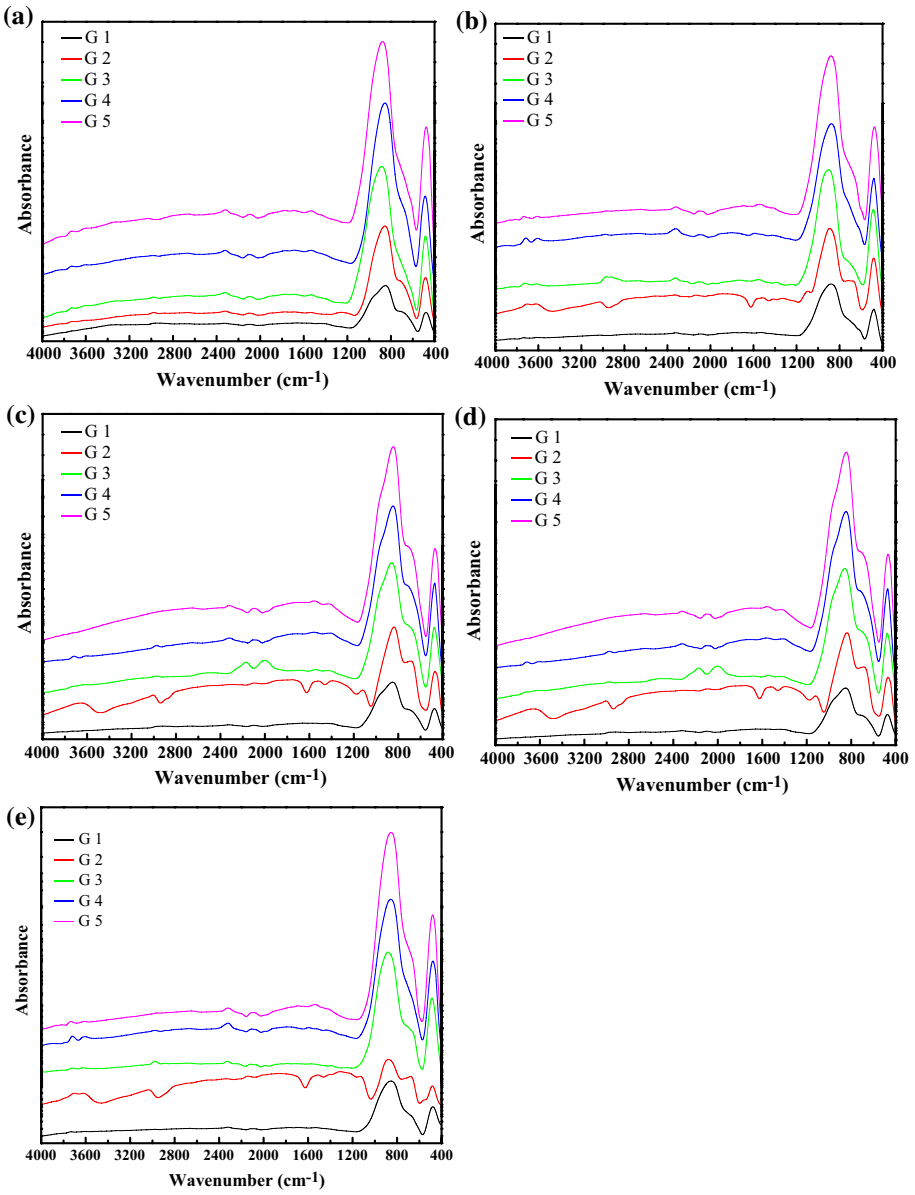

Fig. 2 **a** Infrared spectra before gamma irradiation. **b** Infrared spectra after 50 KGy gamma irradiation. **c** Infrared spectra after 100 KGy gamma irradiation. **d** Infrared spectra after 150 KGy gamma irradiation. **e** Infrared spectra after 200 KGy gamma irradiation

Further, it is clearly noticed that to increase in concentration of CdO from 10 to 60 mol%, there is slow lessening in the strength of the band that related to decreases the glass arrangement settled to give less stable structuresin cooperation of CdO in the network structural (Niu et al. 2011).

Table 2 Peak assignments for the prepared glasses

Wave number (cm^{-1})	Vibrational modes
465–477	Bending vibrations of Si–O–Si linkages
560–617	Pb–O stretching vibration in PbO_4 structural units
660	Pb–O symmetrical bending Vibrations
716–722	Can be related to Pb–O bond vibrations of PbO_n units with n = 3 and or 4
848–883	Is identified which is due to symmetric stretching vibration of O–Si–O bonds
890–970	Can be related to Si–O stretching mode of non-bridging oxygen's
1124–1255	Is related to antisymmetric stretching vibrations of bridging oxygen's
1300–1560	Is identified and related to Si–O–Si antisymmetric vibrations of bridging oxygen's
1650–1750	Is related to molecular water
3200–3700	Is related to stretching of (OH) group and molecular water

3.1.2 Effect of gamma irradiation on FTIR analysis

Figure 2b–d shows IR absorbance of glasses after gamma irradiation, When glasses sample is exposed to different doses of gamma ray, its spectra and its peak seem to remain in the same position. However, the only limited observed change is the increase or the prominence of the main bands reflects the changes taking place in the glass structure after interaction with gamma irradiation (Kaur et al. 2012). The constancy of glasses to gamma irradiation due to the manifestation of HMO and also to the capability of both Cd^{2+} and Pb^{2+} ions to participate in the network structure by forming additional PbO_4 and CdO_4 or CdO_6) units (Kaur et al. 2012; El-Batal et al. 2010; Wong and Angell 1976). Experimental data indicate that the IR spectra of all glasses after gamma irradiation reveal the same main vibrational bands with nearly no changes in their numbers or positions. This indicates general stability of such glasses towards gamma irradiation due to the presence of heavy metal cations.

3.2 Density and molar volume

Figure 2e represented densites (ρ) and Fig. 3 represented molar volume (V_m) of the glasses before and after gamma irradiation with CdO concentration. The glass density values are shown in Table 3. The obtained data showed that reason of density slightly decrease, may be accorded that that Cd^{2+} ions will occupy the glassy vacancies, along with Pb^{+2} ions act as a glass modifier. There are many factors affect directly the density and molar volume values, such as the difference in the density of both SiO_2 and CdO, the differences in their molecular weights and the differences in the atomic radii of the replaced cations [Si (1.11Å) and Cd (1.61Å)]. In other arguments, it can be comprehended that the large ionic radius leads to an increase the number bridging oxygen NBO to give more open structure and the decrease in density would take place. As shown in Table 3. In the studied glasses, it was observed that the density of irradiation samples after successive gamma irradiation doses leads to obvious decrease for most samples in the density values with comparing before gamma irradiation which may be that electric defects and breaks in the bonds, the above data is agreement with the data obtained by agreement with the data obtained by El-Batal et al. (2010) and Manfredo and Pye (1978).

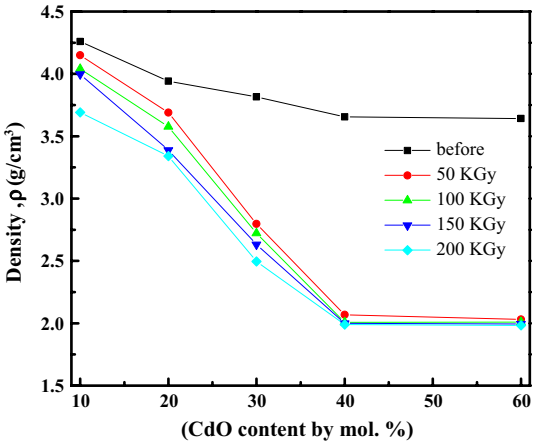

Fig. 3 Densities of the investigated glasses before and after gamma irradiation with CdO content by mol%

Table 3 The densities and molar volumes (before and after irradiation), of the studied glasses

Sample name	ρ (g/cm^3)					Molar volume (V_m (m^3 mol^{-1})				
	Before	50	100	150	200	Before	50	100	150	200
		Dose (KGy)					Dose (KGy)			
G1	4.259	4.15	4.04	3.998	3.691	28.33	24.57	25.24	25.5	27.62
G2	3.942	3.689	3.576	3.388	3.34	32.34	29.28	30.21	31.88	32.34
G3	3.815	2.797	2.722	2.632	2.495	35.21	40.79	41.91	43.35	45.73
G4	3.655	2.069	2.008	2	1.99	38.62	58.07	59.83	60.07	60.38
G5	3.642	2.03	2.01	1.994	1.983	42.51	62.17	62.79	63.29	63.65

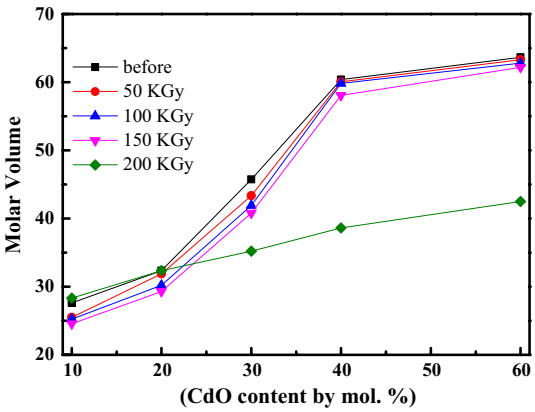

Fig. 4 Molar volumes of the investigated glasses before and after gamma irradiation with CdO content by mol%

3.3 Ultrasonic velocities and elastic constants

The ultrasonic non-damaging testing has been determined to be one of the quality strategies to look at the microstructure, characterization, mechanical properties, and phase adjustments as well as to evaluate elastic constants (El-Alaily et al. 2017, 2018). One

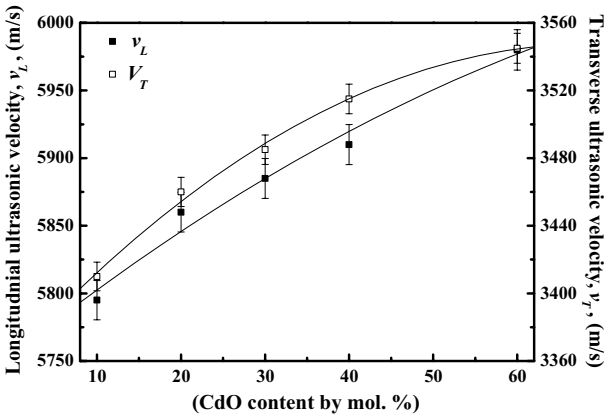

Fig. 5 The longitudinal and shear ultrasonic velocities v_L and v_T of the investigated glasses with CdO by mol%

also can signify the materials inclusive of semi engaging in glasses, high-quality undertaking glasses, glass ceramics, bio-energetic glasses and many others, by means of this nondestructive testing approach. The acoustic wave spread in bulk glasses has been of a tremendous hobby to apprehend their mechanical properties (Shaaban et al. 2017). Both ultrasonic velocites (longitudinal, v_L and shear, v_T) decreases with the cadmium content increase Table 3. The decrease in ultrasonic velocity is specifically accredited to the variability in the (ρ) and the elastic moduli (Shaaban et al. 2017). This decrease in the velocity may be associated with the decrease in the rigidity and the (ρ) of the glasses (Shaaban et al. 2017). Figure 4 represented of (v_L) and (v_T) of the glass with concentration of CdO. It found that, both velocities (v_L and v_T) decreased.

Figures 5 and 6 represented of elastic moduli with CdO content. The values of elastic moduli had been showing decrease with the increase of CdO content. The decrease in K may be associated with the changing inside the coordination number with an increasing inside the CdO awareness, the decrease in the average force constant and the decreasing in elastic moduli may be associated with the crosslink density. While CdO increases (ρ) decrease and (V_m) increase which, makes the glasses is less compact, the bond strength between Cd–O (60 K Cal.) is lower than the bond strength of Si–O (106 K Cal.) (Kannappan et al. 2009).

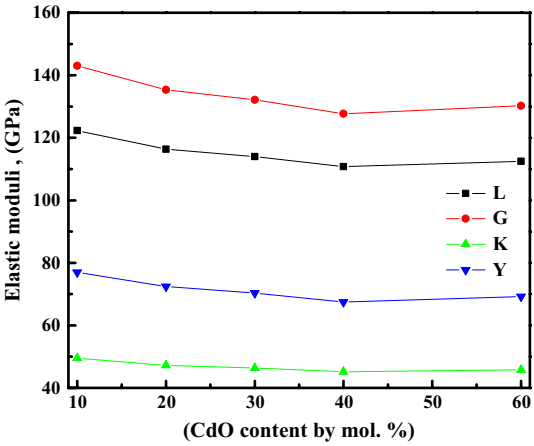

Fig. 6 The elastic moduli experimentally of the investigated glasses with CdO by mol%

Table 4 The values of sound velocities (v_L and v_T), elastic moduli (calculated) of the studied glasses, oxygen molar volume, oxygen packing density, poison's ratio and thermal expansion coefficient (α_P), of prepared glasses

Sample name	v_L (m* s^{-1})	$_vT$ (m* s^{-1})	L (GPa)	G (GPa)	K (GPa)	Y (GPa)	V_0 cm^3/mol	O_{PD} mol/L	σ	α_P K^{-1}
G1	5980	3545	80.94	53.52	152.30	131.57	121.14	9.768	0.229	138,722.67
G2	5910	3515	72.75	48.70	137.69	119.45	102.18	11.55	0.226	137,098.67
G3	5885	3485	70.35	46.33	132.13	113.98	90.29	13.04	0.230	136,518.67
G4	5860	3460	67.17	43.76	125.51	107.85	79.1	14.85	0.232	135,938.67
G5	5795	3410	65.84	42.35	122.31	104.62	69.3	17.71	0.235	134,430.67

Table 5 Values of packing density (*Vi*), Dissociation energy (*Gi*), elastic moduli, (according to Makishima – Mackenzie Model), micro hardness (H), Debye temperature (θ_D), and acoustic impedance (Z), of prepared glasses

Sample name	*Vi* (m^3) 10^{-6}	*Gi* (kcal/kJ)	*L* (GPa)	*G* (GPa)	*K* (GPa)	*Y* (GPa)	H (H/(109 N m^{-2}))	$_D$ (K)	Z (Z/(107 kg m^{-2} s^{-1}))
G1	0.491	13.29	64.02	23.98	32.04	54.55	9.67	472.08	2.55
G2	0.415	12.46	48.04	19.92	21.48	43.25	8.89	466.21	2.33
G3	0.368	11.62	38.59	17.15	15.73	35.74	8.34	466.02	2.25
G 4	0.323	10.79	30.88	14.72	11.25	29.13	7.81	463.61	2.14
G5	0.271	9.127	21.88	11.39	6.692	20.66	7.48	468.57	2.11

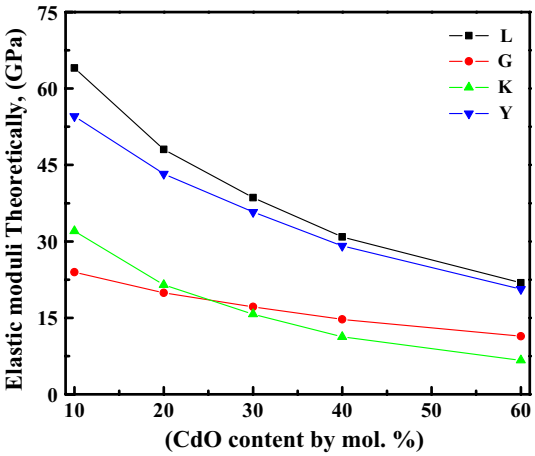

Fig. 7 The elastic moduli theoretically of the investigated glasses with CdO by mol%

Further, the decrease in micro hardness (H), oxygen molar volume, oxygen packing density, acoustic impedance as shown in Tables 4 and 5, and Debye temperature, because the network modifier (NWM) (Laopaiboon and Bootjomchai 2013) content is increased and the increasing values of thermal expansion coefficient as shown in Table 4 are due to the reduction in rigidity of the glass structure.

3.4 UV absorption spectra

Spectroscopic properties are a very beneficial technique for characterizing the optically well as electronic l properties of dissimilar materials as glass, ceramics thin films, filters and pigments (Varshneya 2013). Experimental absorption designates that the induced bands in the cadmium lead silicate glasses extend in the UV–Vis ranges are connected to essentially with the lead glasses contains of bands arising from the 6 $s^2 \rightarrow 6$ s 6p transitions, the lower mostlivelinessconforming to ν of the ^{1} s0$\rightarrow ^3$ p1 thatexpected by Saddeek et al. (2018), Sushama and Predeep (2014) and Duffy (1997). However, no different encouraged bands in the visible region and the full range lef to vers nearly unaffected which, can be associated to the occurrence of high percent 60 mol% of HMO (CdO) which takes steadiness and obstruction to the result of gamma irradiation.

Figures 7 and 8a–d, depicted the measured Absorbance (A) spectra with and without gamma irradiation of the investigated samples. It's originated that, visual absorption superiority is not suddenly definited in the current samples, which obviously designates their glassy state. The adding of CdO changes the spectrum into the lower λ adjacent (i.e. this change E_{opt}) (Duffy and Ingram 1975, 1976). The E_{opt} of the investigated samples can also be evaluated by Aly (2015) and Mott and Davis (1977):

$$\propto = \frac{A_0(\mathbf{h}\omega - E_{opt})^r}{\mathbf{h}\omega} \tag{19}$$

where A_0 is a constant, h is Planck's constant divided by 2π, $h\omega$ is the photon energy and r is the index countryside of conversion. The values of r = 1/2, 3/2, 2 or 3 correspond to direct allowed, direct forbidden, indirect allowed and indirect forbidden, correspondingly.

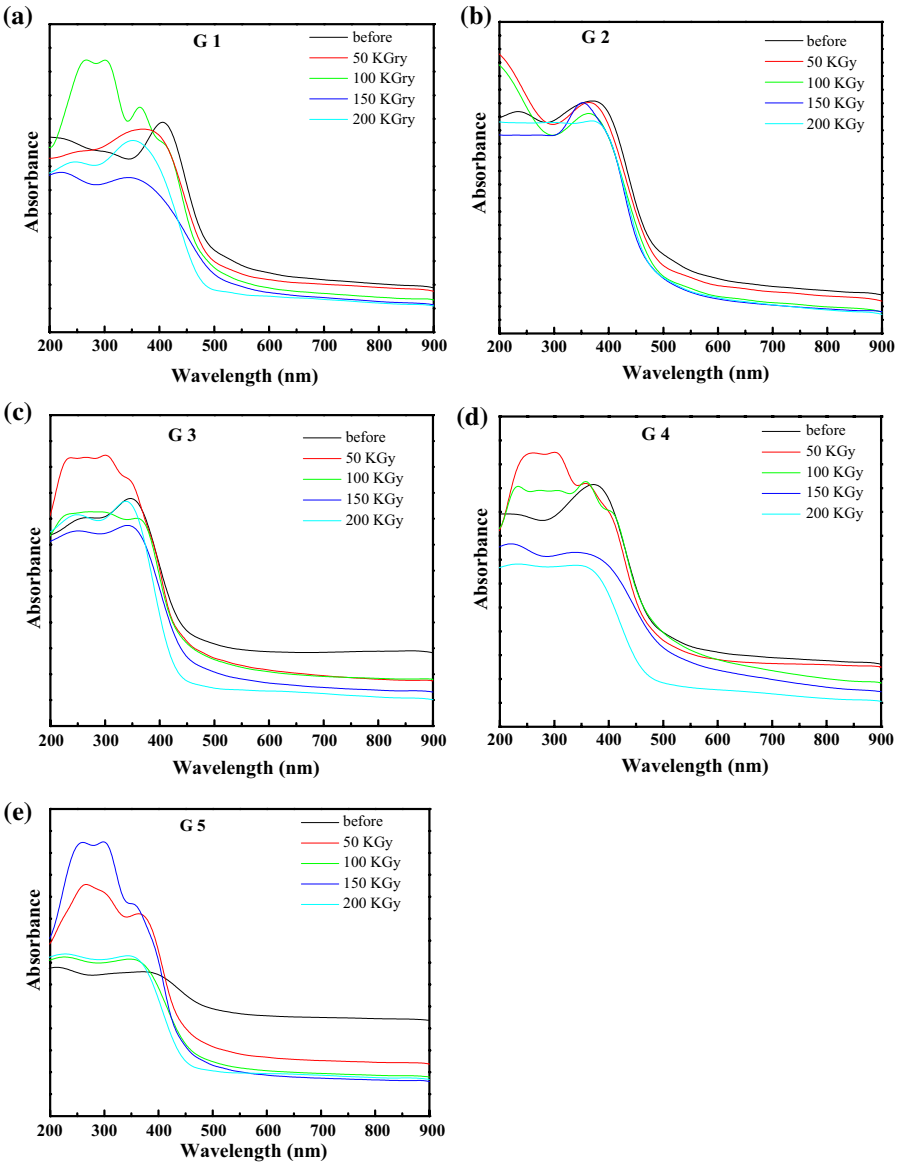

Fig. 8 **a** Absorption spectra of G1 before and after gamma irradiation. **b** Absorption spectra of G2 before and aftegamma irradiation. **c** Absorption spectra of G3 before and after gamma irradiation. **d** Absorption spectra of G4 before and after gamma irradiation. **e** Absorption spectra of G5 before and after gamma irradiation

The E_{opt} value is shown in Fig. 8e and Table 6. This decreasing of E_{opt} can be accredited to the increase of bonding deficiencies and NBO's and increase of attendance of give rmid-point, clues to the decrease of E_{opt} with and without gamma irradiation.

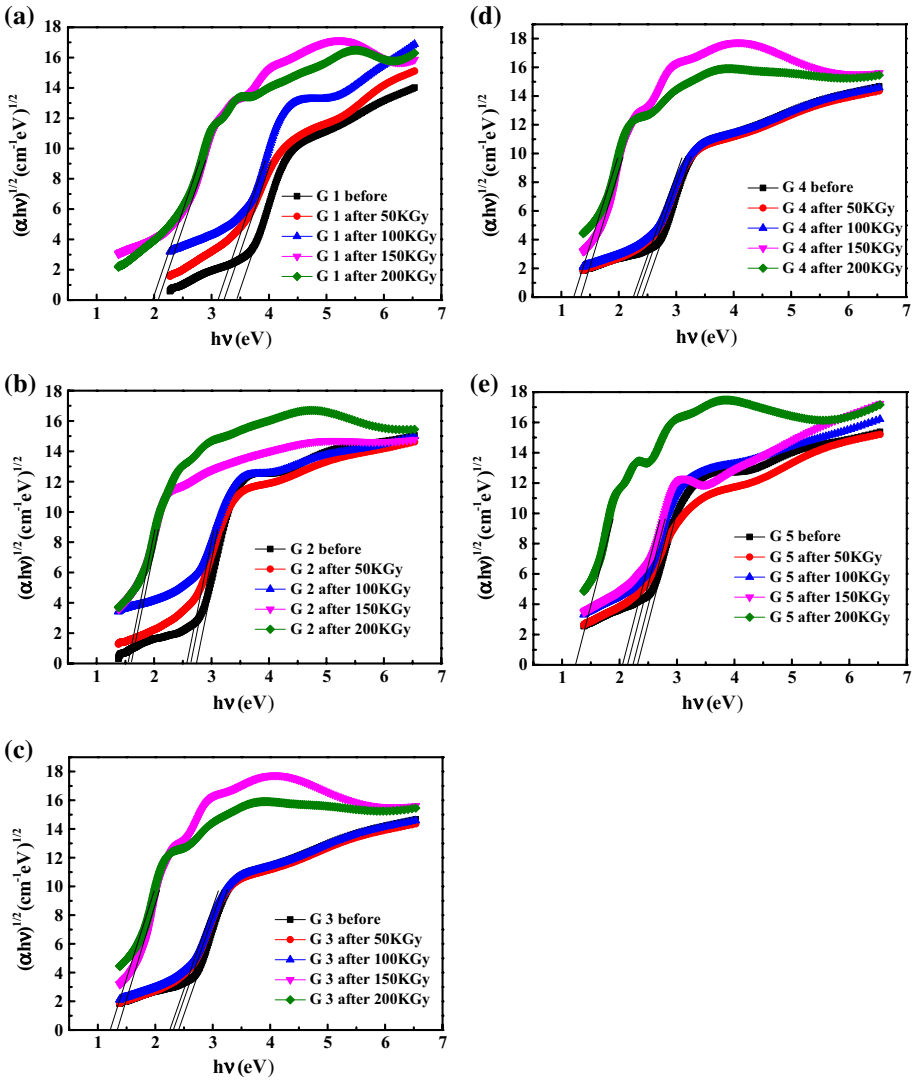

Fig. 9 **a** E_{opt} of G1 before and after gamma irradiation. **b** E_{opt} of G2 before and after gamma irradiation. **c** E_{opt} of G3 before and after gamma irradiation. **d** E_{opt} of G4 before and after gamma irradiation. **e** E_{opt} of G5 before and after gamma irradiation

The refractive index (n) are calculated as:

$$n = \frac{(1-R)^2 + k^2}{(1+R)^2 + k^2} \tag{20}$$

The k values have been determined using the relation ($k = \alpha\lambda/4\pi$). According to the Lorentz–Lorenz equation (Wahab and Shaaban 2018; Nabhan et al. 2017; Marzouk 2012; Marzouk et al. 2013, 2015; Singh et al. 2008), density of material can be affect on the (n) in a straight proportion and contrariwise proportional to (V_m). Thus, the decrease in the

Table 6 The optical band gab $E_{opt.}$ (before and after irradiation), of the studied glasses

Sample name	E_g (eV)				
	Before	50 (KGy)	100	150	200
G1	3.455	3.25	3.11	2.1	1.97
G2	2.75	2.65	2.58	1.61	1.55
G3	2.43	2.34	2.27	1. 34	1.23
G4	2.41	2.31	2.26	1.34	1.23
G5	2.3	2.22	2.08	2	1.2

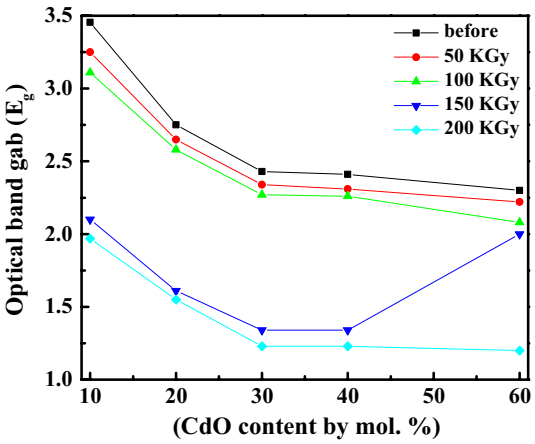

Fig. 10 E_{opt} of the glass system before and after gamma irradiation with CdO content by mol%

values of (n) is ascribed to the decrease in density of the glass. Refractive index of studing glasses decreases with CdO as shown in Figs. 9a–d and 10 refractive index is inversely proportional to the (V_m).

4 Conclusions

Different concentrations of cadmium doped with lead silicate glasses were synthesized. These glasses have been analyzed by FTIR, UV spectroscopic, and γ gamma radiation. FTIR spectroscopy confirmed that the tybe structure of these glass will be less compact, this result leads to decrease the densities and refractive index with increasing the CdO with and without gamma irradiation. The two velocities (v_L) and (v_T) of the current glasses with different mol% of CdO content are decreased. The elastic moduli decrease with increasing of CdO content. According to Makishima–Mackenzie, both of Shear modulus and longitudinal modulus decrease with increasing of CdO content. This decrease in elastic moduli may be connected to the changing in the coordination number with increases in the CdO concentration, the decrease in the average force constant and the crosslink density. The value of E_{opt} with and without gamma irradiation wasdecreased with increasing of CdO, This decrease of E_{opt} can be accredited to type structure of glass.

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