Effect of Gamma-Irradiation on Optical Properties of Glass Composition x mol%(Li_2O) (30-x) mol%(Na_2O) 65 mol%(B_2O_3):5 mol%(Sm_2O_3)

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Abstract

Nominal glass compositions x mol%(Li_2O) (30-x) mol%(Na_2O) 65mol%(B_2O_3): 5mol%(Sm_2O_3) where x=(0, 10, 20 and 30) mol% have been prepared by the conventional quenching melt annealing technique. The optical constants (α, n, k), optical energy gap (E_g), Urbach energy tail (ΔE), linear refractive index (n_0), nonlinear refractive index (n_2), linear optical susceptibility $\chi^{(1)}$ and the third order of non- linear optical susceptibility $\chi^{(3)}$ have been calculated before and after 8MRad γ- irradiation. The analysis of the absorption coefficient data revealed the existence of an allowed indirect transition both before and after γ-irradiation. It was observed that the value of optical energy gap depends on the value of lithium content (x). E_g increases from (3.15eV- 3.3eV) and from(2.93 eV -3.09eV) before and after γ- irradiation respectively when lithium content value (x) varies from 0 to 20 mol% then decreases to 3.24eV and 2.85 eV at 30 mol% of lithium content before and after γ-irradiation respectively. ΔE decreases from 0.1193 eV to 0.095 eV and from 0.565 eV to 0.401 eV with increasing lithium content from 0 to 20 mol%, then increases to 0.114 eV, 0.512 eV at 30 mol% of lithium oxide respectively before and after gamma irradiation. The linear and nonlinear optical parameters have been estimated from the obtained values of n and E_g. It was found that n_0, $\chi^{(1)}$, n_2 and $\chi^{(3)}$ vary from (2.32-2.35), (0.35- 0.363)esu., (24.1-27.71)x10^{-12}&(1.48-1.73)x10^{-12}esu.when lithium content value varies from 0 to 30 mol% before γ-irradiation respectively. The parameters n_0, $\chi^{(1)}$, n_2 and $\chi^{(3)}$ are found to vary from (2.37-2.63), (0.368-0.473), (29.332-71.626)x10^{-12}& (1.846-5.001)x10^{-12}esu.afterγ-irradiation respectively. The variation in optical band gap and Urbach energy are due to the variation in the glass structure.

Keywords: Borate Glasses, Nonlinear Optics, Optical Energy Band Gap, γ-irradiation

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Introduction

Glasses and crystals doped with various rare earth ions are important materials for diverse applications in diverse fields such as waveguides, solid state laser, optical fibers, display devices, etc [**Biji et al. 2004;Kaewnuam et al. 2016; Shaweta Mohan, 2017; Yang et al. 2015**]. The study of optical properties is one of the most useful tools to provide the information of the electronic structure of amorphous semiconductors [**Pierece, 1972**]. The investigations of glass, as host materials for rare- earth, optical and fluorescence with potential for laser and other optical applications are confirmed to borate, phosphate, silicate and fluoride glasses [**Reisfeld and Jorgensen, 1977; Gan, 1992**]. Among rare-earth ions, Sm^{+3} ion is one of the most interesting ions to analyse the optical and fluorescence properties because of its use in high- density optical storage, under sea communication and color displays and its emitting $^4G_{5/2}$level which exhibits relatively high quantum efficiency and also shows different quenching emission channels [**Manoj Kumar**, **2006;Mazurak**, **2010**]. The physical and optical properties of Sm^{+3} doped BNaLi glasses indicate that a negative value for the bonding parameter (δ) and the small value of JO parameter (Ω_2) have substantial the ionic nature of Sm^{+3} ligand bond in the Sm^{+3}- doped BNaLi glass [**Tuyen, 2014**]. The physical and optical properties of $(Li_2O)_x(B_2O_3)_{1-x}$glass were dependent on the glass composition, the increase in density with Li_2O content was due to the number of non- bridging oxygen [**Halima et al., 2014**]. The molar volume decreased as a result of Li_2O occupied interstitial position in the network. The formation of non-bridging oxygen increased with Li_2O resulted in the decrease of optical energy band gap for both direct & indirect band gap. The decrease in Urbach energy is owing to the decrease of degree of disorder in glass structure with Li_2O [**Halima et al., 2014**]. Spectroscopic studies of Sm^{+3} ions have been reported in different hosts such as water [**Camall, 1968**], crystals [**Adam et al., 1985; Do et al., 2012**] and glasses [**Ratankaram et al., 2005; Nachinmuthu et al., 1977; Jayasankar et al., 2000; Lin et al., 2005**]. The aim of the present study is to show the effect of γ-irradiation on linear optical parameters, nonlinear optical parameters and optical constants over a wide range of wavelengths and as a function of Li_2O content for x mol% (Li_2O) (30-x) mol% (Na_2O) 65 mol% B_2O_3 glass doped with 5 mol% Sm_2O_3.

2. Experimental

2.1. Materials, preparation of the glasses

Glasses of nominal composition x mol% (Li_2O) (30-x) mol% (Na_2O) 65mol% (B_2O_3):5 mol% (Sm_2O_3) where x= (0, 10, 20 and 30) mol% were successfully synthesized via conventional quenching melt annealing technique using analytical grade reagent. Lithium and sodium oxide are prepared using their carbonate partner, boron prepared using orthoboric acid while Sm^{+3}were used in oxide form as received.

Accurately weighted chemicals were mixed to form batches of 50 g melted in porcelain crucibles in the temperature range 1100-1250 °C depending on composition in an electrically-heated furnace for 2 h. Samples were rotated thoroughly at fixed time intervals to ensure homogeneity and to remove air bubbles. Molten glass were removed from the electrical furnace and then poured to preheated stainless steel mold of required dimensions in air and then transferred to annealing muffle adjusted at 350 ^oC. Annealing muffle was turned off and kept to cool gradually to room temperature by a rate of 10 $^oC/h$.

2.2. Technique for optical measurements

The optical transmission T% and reflection R% at normal incidence for x mol%(Li_2O) (30-x) mol%(Na_2O) 65 mol%(B_2O_3): 5 mol%(Sm_2O_3) where x=(0, 10, 20 and 30) mol%were carried out using Jasco (Type V-570) spectrophotometer in the wavelength range 200-2500 nm before and after 8MRad γ- dose irradiation for polished samples with constant thickness.

3. Results and discussion

3.1. Determination of optical energy gap

To show the effect of γ- irradiation on the optical constants, (absorption coefficient α, refractive index n and dielectric constants ε_1 and ε_2)of x mol%(Li_2O) (30-x) mol%(Na_2O) 65 mol%(B_2O_3) glasses doped with 5 mol% of $Sm_2O_3$where x= (0, 10, 20 and 30) mol% irradiated with 8MRad gamma dose were determined.

Figures (1) and (2) represent the spectral distribution curves of T% and R% for irradiated and irradiated samples respectively.

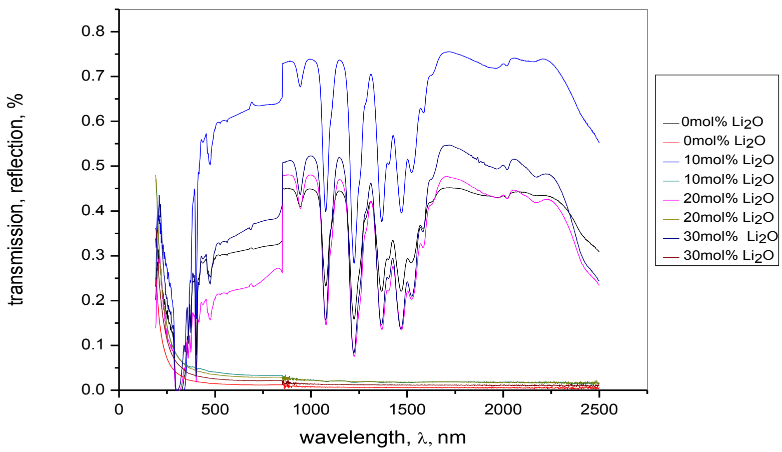


Fig. 1. The transmittance T% and the reflectance R% of unirradiated glass sample.

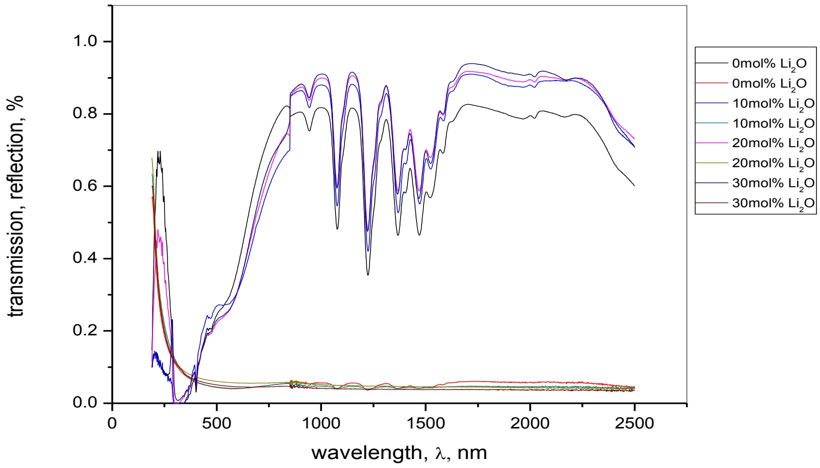


Fig. 2. The transmittance T% and the reflectance R% with 8MRadγ-irradiated glass sample.

The optical absorption coefficient α could be calculated by using the measured values T% from

$$\alpha = \frac{1}{t}\ln T \tag{1}$$

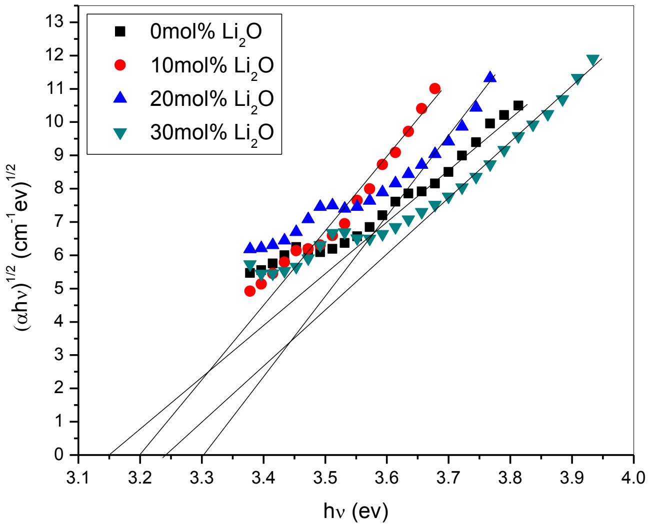
where t is the thickness of the sample

It was found that, the samples both before and after γ- irradiation obeyed the relationship for indirect transition [**Thamarai et al., 2017**].

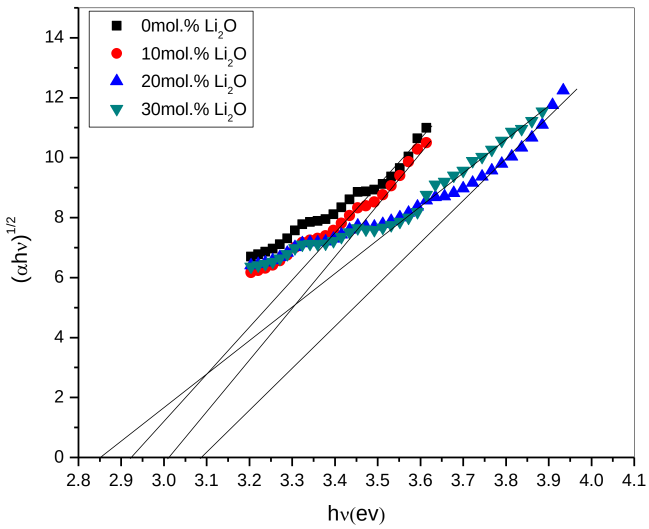
$$\alpha h\nu = A(h\nu - E_g)^2 \tag{2}$$

Where A is a constant

The functional dependence of $(\alpha h\upsilon)^{1/2}$ against the photon energy hυ for the un-irradiated and irradiated x mol%(Li_2O) (30-x) mol%(Na_2O) 65 mol%(B_2O_3) glasses doped with 5mol%Sm_2O_3 with x= (0, 10, 20 and 30) mol% are shown in fig.3 and fig.4 respectively.



(Fig.3): The dependence of $(\alpha h\upsilon)^{1/2}$ on photon energy (hυ) for lithium sodium borate glasses with sm_2O_3 before γ- irradiation.



(Fig.4):The dependence of $(\alpha h\upsilon)^{1/2}$ on photon energy (hυ) for lithium sodium borate glasses with sm_2O_3 after γ- irradiation.

Assuming the validity of Eq.(2) for indirect transition in the present system. The values of indirect optical band gap can be obtained by extrapolating the straight line on the curve to the axis at $(\alpha h\upsilon)^{1/2}=0$. The values of the indirect optical band gap for the un-irradiated and γ-irradiated indicating that, it increases from (3.15eV-3.3 eV) and from (2.93 eV-3.09 eV) with increasing Li_2O content from 0 to 20 mol%, then it decreases to 3.24 eV& 2.85 eV at 30 mol% Li_2O content respectively as shown in fig.5.

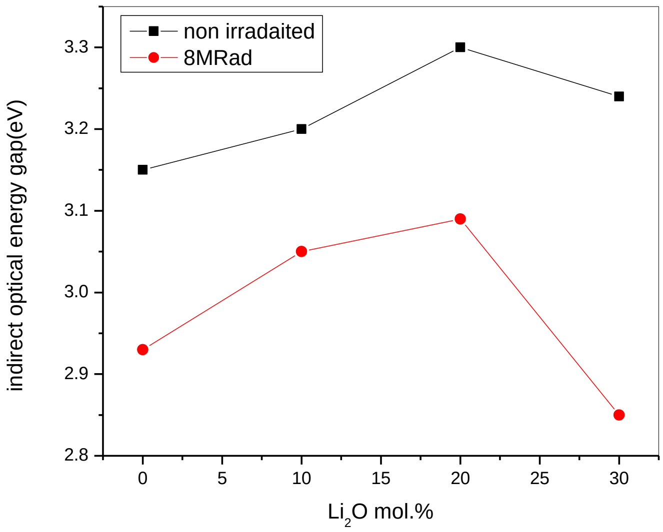


Fig. (5) Variation of indirect optical band gap vs. Li_2O content before and after γ irradiation .

It is noted that with the increase of Li_2O content the indirect optical band gap, E_g first increases and then decreases, the increase in lithium ions causes structural change in the glass network. The formation of non-bridging oxygen, which binds excited electrons less tightly than bridging oxygen increases with Li_2O content results in the decrease of an allowed indirect opical band gap [**Gayathri et al., 2011**].

When Lithium ions were added in a small amount, there was an increase in the number of BO_4 units against membered borate rings with only one BO_4 tetrahedron appeared and it can be in triborate, tetraborate or penta borate forms. Thus the numbers of non-bridging oxygen increase with further addition of Li_2O and break up the regular structure of the borate and boroxyl rings and as a result the band gap decreases [**Hager,2009**].

3.2.Determination of Urbach energy

The absorptionedge in many disordered materials follows the Urbach rule given by [**Dhineshbabu et al., 2017**]

$$\alpha(\omega) = B \exp\left(\frac{\hbar\omega}{\Delta E}\right) \tag{3}$$

where α(ω) is the absorption coefficient at an angular frequency of ω=2πf, ΔE is the width of tail of localized states in the band gap (Urbach energy) and was determined from the reciprocal of the slope of the plot lnα(ω) versus ħω. The Urbach energy is well known that the shape of the fundamental absorption edge in the exponential (Urbach) region can be yield information on the disorder effects [**Kesavulu et al., 2010**]. Materials with larger Urbach energy would have greater tendency to convert weak bonds into defects [**Rani et al., 2009**]. It was found that Urbach energy (ΔE) for xmol%(Li_2O) (30-x) mol%(Na_2O) 65 mol%(B_2O_3)

glasses doped with 5 mol% Sm_2O_3wherex= (0, 10, 20 and 30) mol%decreases from 0.1193 to 0.095 eV and from 0.565 to 0.401 eV with increasing lithium oxide content from 0 to 20 mol%, then it increases to 0.114 eV, 0,512 eV at 30 mol% before and after γ- irradiation respectively.

The decrease in Urbach energy is due to the decrease in the degree of disorder in glass structure with the increasing of Li_2O mol% content [**Upender et al., 2010**].

Fig.6. Shows the dependence of Urbach energy ΔE on the Li_2O mol% content for x mol% (Li_2O) (30-x) mol% (Na_2O) 65 mol% (B_2O_3) glasses doped with 5mol% Sm_2O_3 with x= (0, 10, 20and 30) mol%

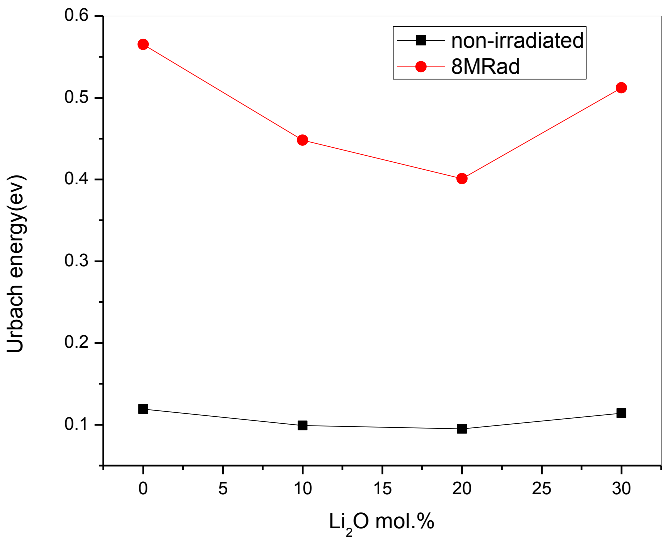


Fig. (6) Variation of Urbach energy vs. Li_2O mol.% content before and after γ irradiation .

The data of an allowed indirect optical energy gap and Urbach energy for x mol% (Li_2O) (30-x) mol% (Na_2O) 65 mol% (B_2O_3) glasses doped with 5mol% of Sm_2O_3 with x=(0, 10, 20and 30) mol% before & after γ irradiation are listed in table 1.

Table1. The data of indirect energy gap optical transition data for alkali borate glass containing Samarium oxide and the effect of irradiation

Lithium content mol%	Un-irradiated		8MRad	
	E_g (ev)	ΔE (ev)	E_g (ev)	ΔE (ev)
0	3.15	0.119	2.93	0.565
10	3.2	0.099	3.05	0.448
20	3.3	0.095	3.09	0.401
30	3.24	0.114	2.85	0.512

Figures [7], [8], [9] and [10] a&b show the dispersion curves of refractive index n, real part of complex dielectric constants ($\varepsilon_1=n^2-k^2$), extinction coefficient k and imaginary part of dielectric constant ε_2 = 2nk for x mol%(Li_2O) (30-x) mol%(Na_2O) 65 mol%(B_2O_3) glasses doped with 5 mol% Sm_2O_3 with x= (0, 10, 20 and 30) mol% before and after γ- irradiation respectively.

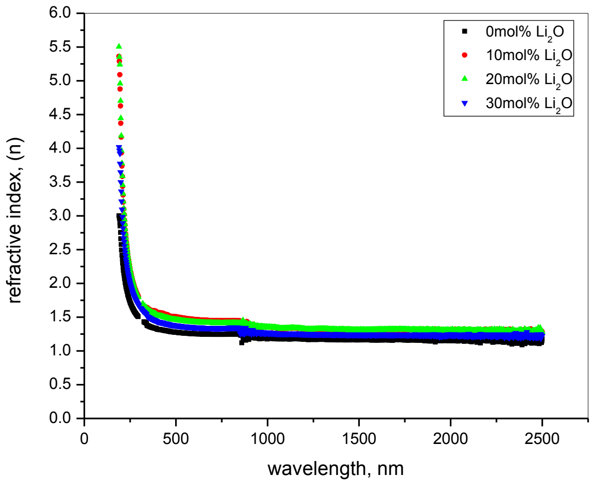


Fig.7a. Dispersion curve of refractive index n with wavelength before γ- irradiation.

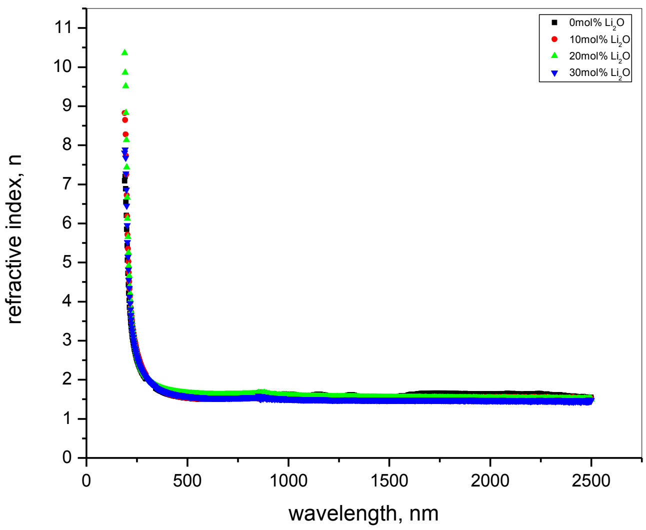


Fig.7b. Dispersion curve of refractive index n with wavelength after γ- irradiation.

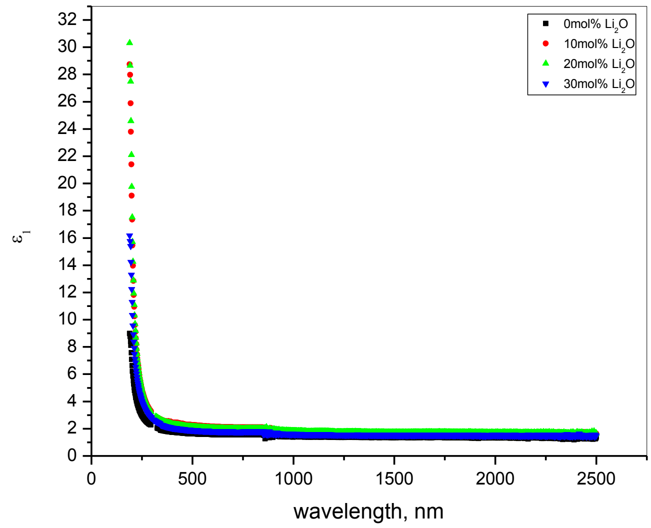


Fig.8a. Dispersion curve of real part of complex dielectric constants (ε_1) with wavelength before γ- irradiation.

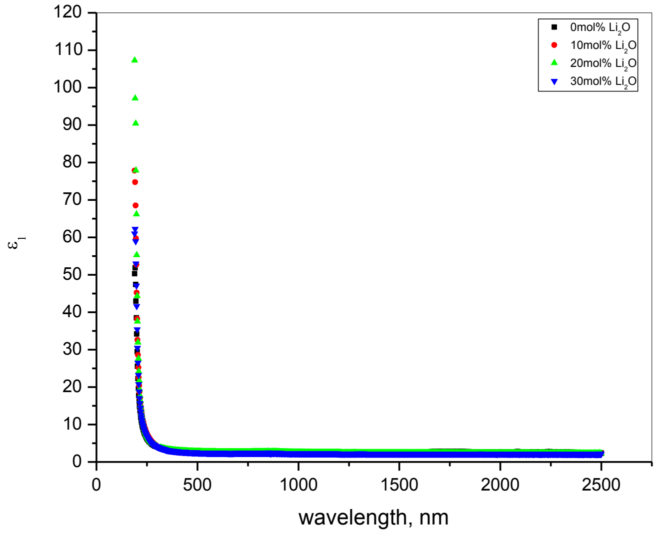


Fig.8b. Dispersion curve of real part of complex dielectric constants (ε_1) with wavelength after γ- irradiation.

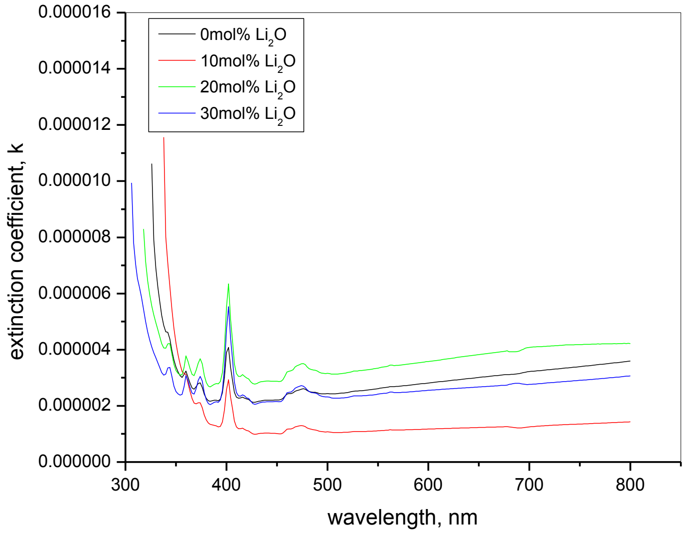


Fig.9a. Dispersion curve of extinction coefficient (k) with wavelength before γ- irradiation.

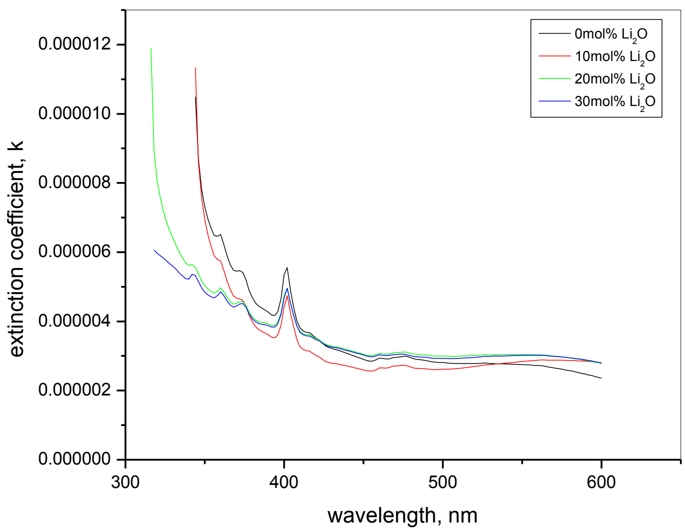


Fig.9b. Dispersion curve of extinction coefficient (k) with wavelength after γ- irradiation

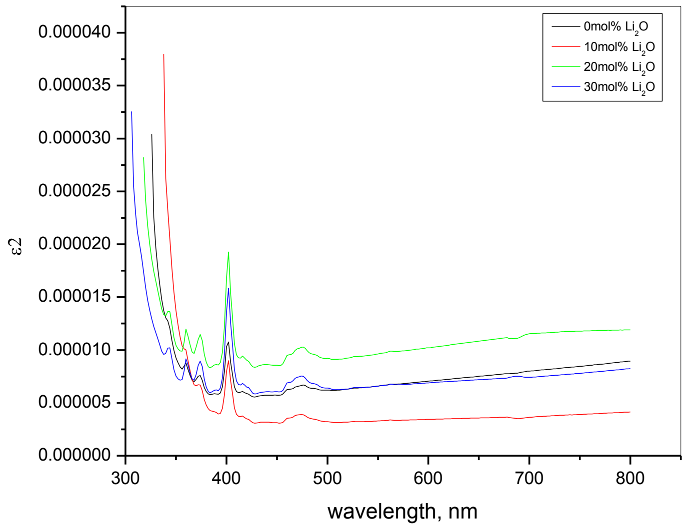


Fig.10a. Dispersion curve of imaginary part of complex dielectric constants (ε_2) with wavelength before γ- irradiation.

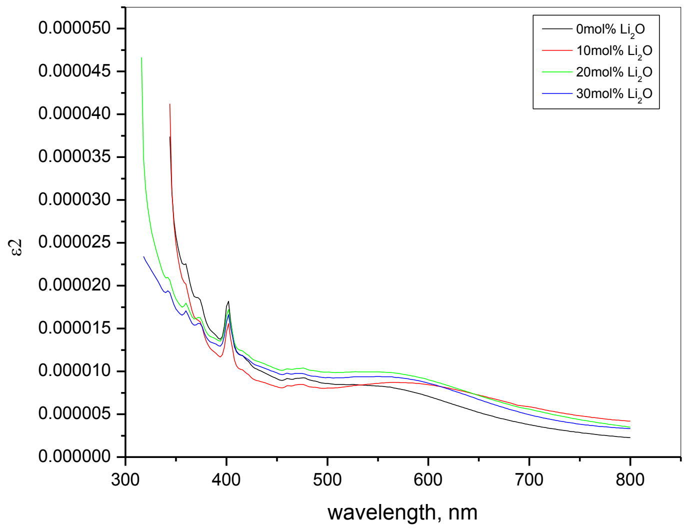


Fig.10b. Dispersion curve of imaginary part of complex dielectric constants (ε_2) with wavelength after γ- irradiation.

It is clear that, refractive index (n) and dielectric constant (ε_1) decreases with increasing both wavelength and lithium oxide content. An addition of Li_2Ocauses changed in coordination number and creates more non-bridging oxygen. Thus it has a higher average coordination number of studied glasses which results in the increase of refractive index [**Saddek et al., 2008**].

3.3. Calculation of optical linear parameters and Non -linear Optical parameters

The linear optical refractive index n_0 can be calculated from the data obtained from the optical energy gap of indirect transition according to the relation:

$$\frac{n_0{}^2-1}{n_0{}^2+2} = 1 - \sqrt{E_g/20} \tag{4}$$

It was noted that, the observed data of refractive index are in a good agreement with the obtained data [**Marzouk, 2012**]. The estimation of non-linear optical parameters is mostly obtained from the third order non-linear optical susceptibility $\chi^{(3)}$ and linear refractive index n_o data. $\chi^{(3)}$ can be calculated from $\chi^{(1)}$ from miller's rule [**Miller, 1967**] according to the relation:

$$\chi^{(3)} = [\chi^{(1)}]^4 x10^{-10} = \left(\frac{n_0{}^2-1}{4\Pi}\right)^4 x10^{-10} \text{e.s.u} \tag{5}$$

Where $\chi^{(1)}$ is the linear optical susceptibility.

The non-linear refractive index n_2 are related to both the real part of third non-linear optical susceptibility and linear optical refractive index n_o by the expression [**Nasu, 1981**]:

$$n_2 = \frac{12\pi Re\ \chi^{(3)}}{n_0} \tag{6}$$

The data of optical linear & non-linear optical parameters before and after 8MRad γ dose irradiation are listed in Table 2.

Table.2: The data of linear refractive index, linear optical susceptibility $\chi^{(1)}$, third order non-linear optical susceptibility $\chi^{(3)}$ and non-linear refractive index n_2 for x mol% (Li_2O) (30-x) mol% (Na_2O) 65 mol% (B_2O_3) doped with 5 mol% Sm_2O_3 with x= (0, 10, 20 and 30)mol% before and after γ-irradiation are listed in table 2.

Lithium oxide content mol%	Un-irradiated				8MRad γ-irradiation			
	n_o	$\chi^{(1)}$ [esu]	$\chi^{(3)}$*10^{-12} [esu]	n_2*10^{-12} [esu]	n_o	$\chi^{(1)}$ [esu]	$\chi^{(3)}$*10^{-12} [esu]	n_2*10^{-12} [esu]
0	2.358	0.363	1.733	27.71	2.416	0.385	2.2	34.277
10	2.345	0.358	1.644	26.433	2.384	0.373	1.927	30.482
20	2.321	0.35	1.483	24.1	2.373	0.368	1.846	29.332
30	2.335	0.354	1.577	25.467	2.635	0.473	5.001	71.626

The obtained linear and non-linear parameters n_o, $\chi^{(1)}$, $\chi^{(3)}$ and n_2 are in agreement with the proposed one [**Wang, 1970**] which proved that optical solid materials having high linear refractive index n_o possess higher non-linear refractive index n_2. Borate glasses have large n_o as well as $\chi^{(3)}$ depending on the highly polarizable oxide ions and cations making an easy distorsion of electron density in a strong electromagnetic field [**Dimitrov and Komatsu., 1999**].

Fig.(11) and fig.(12) show the dependence of third order non-linear optical $\chi^{(3)}$ on the linear refractive index n_o for x mol%(Li_2O) (30-x) mol%(Na_2O) 65 mol%(B_2O_3) doped with 5 mol% Sm_2O_3 with x=(0, 10, 20 and 30) mol% before and after γ-irradiation respectively.

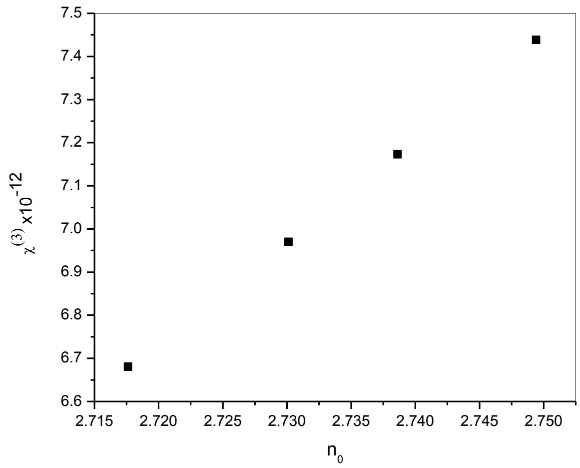


Fig.(11) The dependence of third order non-linear optical $\chi^{(3)}$ on the linear refractive index (n_o) for x mol%(Li_2O) (30-x) mol%(Na_2O)65 mol% (B_2O_3) doped with 5 mol% Sm_2O_3 with x=(0, 10, 20 and 30) mol% before γ -irradiation.

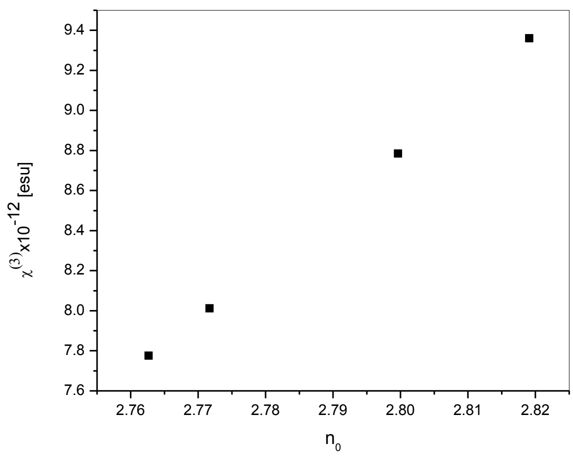


fig.(12) The dependence of third order non-linear optical $\chi^{(3)}$ on the linear refractive index (n_o) for x mol%(Li_2O) (30-x) mol%(Na_2O) 65 mol%(B_2O_3) doped with 5 mol% Sm_2O_3 with x=(0, 10, 20 and 30) mol% after γ-irradiation.

Conclusion

X mol% (Li_2O) (30-x) mol% (Na_2O) 65mol% (B_2O_3) glass doped with 5 mol% of $Sm_2O_3$where x= (0, 10, 20 and 30) mol% were synthesized using conventional quenching melt annealing technique. The optical properties of the prepared samples were dependent on the glass composition and effect of γ-irradiation. The formation of non-bridging oxygen increased with Li_2O content on the decrease of allowed indirect optical energy band gap for both un-irradiated and γ-irradiated. This is because non-bridging oxygen binds excited electrons less tightly than bridging oxygen. The decrease in Urbach energy is owing to the decrease of degree of disorder in glass structure with Li_2O content. A decrease in optical energy gap (E_g) and an increase of Urbach energy (ΔE) after gamma irradiation due to defect formation.

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عنوان البحث:

تأثير التشعيع بأشعة جاما على الخواص الضوئيه لمركبات الزجاج

[x mol% Li_2O (30-x) mol% Na_2O 65 mol% B_2O_3:5 mol% Sm_2O_3] where x= (0,10, 20 and 30) mol%

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الملخص العربى

تم تحضير زجاج الليثيوم الصوديوم بورات المطعم بالسامريوم بتركيز 5 مول% وبنسب وزنيه لاكسيد الليثيوم 0و10و 20و 30 مول% باستخدام التبريد المناسب للصلب المذاب ومن ثم دراسة الخواص الضوئيه وحساب المعاملات الضوئيه الخطيه وغير الخطيه قبل التشعيع وبعد التشعيع لاشعة جاما بجرعة 8 ميجا راد . وقد تم قياس معامل النفاذيه ومعامل الانعكاس عند سقوط عمودى فى مدى من الاطوال الموجيه (200-2500) نانومتر ثم حساب معامل الامتصاص(α) ومعامل الانكسار (n) ودالة الامتصاص k وثوابت العزل الكهربى ε_1 و ε_2 ومعرفة نوع الانتقال الالكترونى ومعامل الانكسار الخطى n_0 ومعامل القابليه الضوئيه الخطى $\chi^{(1)}$ومعامل القابليه الضوئيه الغير خطيه من الرتبه الثالثه $\chi^{(3)}$ ومعامل الانكسار الغير خطى من الرتبه الثانيه n_2 قبل وبعد التشعيع.

وقد اظهرت النتائج ان الانتقال الالكترونى هو انتقال مسموح غير مباشر قبل وبعد التشعيع وان قيم طاقة الفراغ تزداد من 3.15 -3.3 الكترون فولت ومن 2.43 الى 3.09 الكترون فولت بزيادة تركيز الليثيوم من صفر الى 20 مول% ثم تقل الى 3.24 الكترون فولت و 2.85 الكترون فولت عند تركيز 30 مول% لمحتوى الليثيوم فى الزجاج قبل وبعد التشعيع على الترتيب . وتقل كذلك طاقة عرض المستويات المحدده فى منطقة الفراغ الممنوع (طاقة اور بخ) من (0.193 -0.095) الكترون فولت ومن (0.565 -0.401) الكترون فولت بزيادة تركيز الليثيوم من صفر مول % الى20 مول% ثم تزداد الى 0.114,0.512 عند تركيز 30 مول% لاكسيد الليثيوم قبل وبعد التشعيع على الترتيب. وتتراوح قيم المعاملات الضوئيه الخطيه n_0 ، $\chi^{(1)}$ (2.32 – 2.35) ، (0.35- 0.363) وحده كهروستاتيكيه قبل التشعيع و (2.37 – 2.635) ، (0.363- 0.473) وحده كهروستاتيكيه بعد التشعيع على الترتيب وتتراوح قيم المعاملات الضوئيه غير الخطيه $\chi^{(3)}$ ، n_2 من (1.84- 5.00)$\times 10^{-12}$ وحده كهروستاتيكيه ، (29.332-71.626)$\times 10^{-12}$ بعد التشعيع على الترتيب.