UV-IR Spectra of the New Tellurite Glasses of The System TeO₂ -La₂O₃ and TeO₂ -V₂O₅ R.EI-Mallawany, A.Abdel-Kader, M.EI-Hawary and N.EI-Khoshkhany Physics Dept., Faculty of Science, Menofia Univ., EGYPT.

Abstract:

Binary tellurite glass systems of the forms $(TeO_2)_{(100-x)^-}(A_nO_m)_{(x)}$ where $A_nO_m = La_2O_3$ or V_2O_5 and x = 5 to 20 for La_2O_3 and 10 to 50 for V_2O_5 were prepared. UV- spectra of these glasses were recorded in the range of 200-600 nm at room temperature. The optical energy gaps $E_{optical}$ and E_{tail} have been calculated from the optical absorption edge. UV cut-off and IR cut-off have been measured. Calculations of the oxide ion average Polarizability $(\alpha_0^{2^-})$ and optical basicity (Λ) have been done.

1- Introduction:

Tellurite glasses are of scientific and technological interest because of their high refractive indices, low melting temperatures, high dielectric constants as well as their good UV and IR transmissions and considered as promising materials for non-linear optical devices as explained. Some tellurite glasses are also reported to be suitable for setting up optical fiber amplifiers [1]. The absorption edge of each glass depends on the type of network modifier. The absorption coefficient, α (ω), can be determined near the absorption edge using the formula:

$$\alpha(\omega) = \left(\frac{1}{d}\right) \ln\left(\frac{I_0}{I}\right) \tag{1}$$

where d is the thickness of the sample, I₀, I are the intensities of the incident and transmitted beams respectively. For many oxide glasses and other amorphous materials, Urbach rule [2] has been applied.

$$\alpha(\omega) = \alpha_0 \exp(\hbar \omega / E_t) \qquad (2)$$

angular frequency and E_t is the Urbach energy which is interpreted as the width of the tails of the localized states in the band gap associated with the amorphous nature of the material. When the density of states is proportional to some power of

energy $(\alpha\hbar\omega)^{1/2}$ and the relation between absorption coefficient and photon energy gap is considered [3,4], this relation can be re-written as follows:

$$(\alpha\hbar\omega)^{1/2} = A(\hbar\omega - E_{out})$$
 (3)

Where A is a constant and E_{opt} is the optical band gap determined by extra polating the linear parts of the curves to $(\alpha \hbar \omega)^{1/2} = 0$.

2- Experimental procedure:

Binary tellurite glass systems of the forms $(TeO_2)_{(100-x)}$ - $(A_nO_m)_{(x)}$ where $A_nO_m = La_2O_3$ or V_2O_5 and x = 5 to 20 for La_2O_3 and 10 to 50 for V_2O_5 were prepared as described in part 1 of this work Ref.(5).

2.1. Ultraviolet absorption spectra measurements (UV):

The UV absorption measurements for the bulk glass samples with the two parallel polished faces were measured using a UV-spectrophotmeter (Perkin-Elemer Lambada 4B) in the wavelength range 200-600nm at room temperature.

2.2. Infrared absorption spectra measurements (IR):

The IR absorption spectra of the prepared glasses in KBr matrix were recorded on a Perklin-Elemer double beam spectrophotometer model 598 at room temperature over the spectral range of 4000-200 cm⁻¹. The produced glasses were thoroughly mixed with KBr in the ratio 1:40, after that the mixtures were pressed at 10 tons for 5 min. The pellets were clear and uniform. While recording the spectra, the gain of the spectrometer was kept the same for all the samples.

3- Results and Discussion:

3-1 U.V of Tellurite glasses:

Optical absorption measurements in the spectral range 200-600 nm were made at room temperature for the glass samples of the composition of $(TeO_2)_{(100-x)^-}(La_2O_3)_{(x)}$ where [x = 5,7.5,10,12.5,15,17.5 and 20 mol%] and $(TeO_2)_{(100-x)^-}(V_2O_5)_{(x)}$ where[x =10,20,25,30,35,40,45 and 50 mol%]. The ultraviolet light absorbance of the prepared tellurite glasses containing La_2O_3 and V_2O_5 as a function wavelength are shown in Fig.(1-a&b) respectively. The absorption edge of each glass can be identified. There are no sharp absorption edges and this is

characteristic of most glassy oxide materials. It is shown in Fig.(1-a&b) that the absorption edge depends on the kind of the modifier. The position of the fundamental absorption edge shifts to lower wavelength with increasing of La_2O_3 content in binary TeO_2 - La_2O_3 glass or increasing of V_2O_5 content in binary TeO_2 - V_2O_5 glass. The shifts of the absorption edge are most likely related to the structural rearrangement of the glass and the relative concentrations of the various fundamental units.

The absorption coefficient $\alpha(\omega)$ can be determined near the absorption edge from the formula (1). The most satisfactory results were obtained by plotting the quantity $(\alpha\hbar\omega)^{1/2}$ as a function of $(\hbar\omega)$ as suggested by Davis and Mott [3] as shown by Eq.(3). Absorption by indirect transitions applies to many oxide glasses. particularly at the higher values of absorption coefficient. ħ is the reduced Planck's constant and ω is the angular frequency of the incident radiation. Fig.(2-a&b) show the plots of $(\alpha\hbar\omega)^{1/2}$ against $(\hbar\omega)$ and the values of E_{opt} determined by extrapolating the linear parts of the curves to $(\alpha\hbar\omega)^{1/2}=0$. The value of E_{opt} for each glass system are given in Table (1) and represented in Fig. (3-a&b) as a function of La₂Q₃ mol% and V₂O₅ mol% respectively. From Fig.(3-a) it is clear that the value of E_{oot} increases from (3.51 to 3.61eV) due to the modification of tellurite glasses by (5 to 20) mol% La₂O₃. The values of the constant A in Eq.(3) can be determined from the slope of the linear part of the relation $(\alpha\hbar\omega)^{1/2}$ and $\hbar\omega$ which is change from (5.38 to 4.8 cm⁻¹eV⁻¹) due to the modification of tellurite glasses by (5 to 20) mol% From Fig.(3-b) it is clear that the value of Eopt increase from (2.67 to La_2O_3 3.03eV) due to the modification of tellurite glasses by (10 to 50) mol% V₂O₅. The values of the constant A in Eq.(3) can be determined from the slope of the linear part of the relation $(\alpha\hbar\omega)^{1/2}$ and $\hbar\omega$ which is change from (3.04 to 3.38 cm⁻¹eV⁻¹) due to the modification of tellurite glasses by (10 to 50) mol%V2O5 Those values are of the same order as reported by Davis & Mott [3]. The interpretation of the present optical energy gap will be based on the assumption that tellurite glasses having a high concentration of TeO₂ the triangular bipyramid TeO₄ is considered

as the basic co-ordination polyhedron in which tellurium atoms are surrounded by four oxygen and each oxygen is connected with two tellurium atoms, accomplishing an axial-equatorial bonding at which the bonds are easily deformed because of the change in the Teax-Oeq-Te angle taken place along the c-axis due to the structural incorporation of the modifier which creates defects, oxygen vacancies and microholes, and also increases the concentration of non-bridging oxygen [6]. The doping with transition metal oxides decrease the Eopt of (TeO2) which was 3.79 eV [7] to the values mentioned before and can be attributed to the increase of the non-bridging oxygen atoms. While doping with La₂O₃ gave a lower concentration of non-bridging oxygen atoms than V2O5. The estimated value of Eopt for La2O3 tellurite glasses are close to the reported values in [8]. For many amorphous materials. Urbach [2] assumed that the absorption coefficient α (ω) was an exponential function of photon energy ħω as shown by Eq.(2). Fig.(4-a&b) show the variation of $ln(\alpha)$ against $\hbar\omega$ for the present binary glassy systems. The values of Eta in Eq.(2) are calculated from the slopes of the linear regions of those curves. The values of Eta for each glasses system are given in Table (1). It is clear that the values of Eta decrease from (0.1006 to 0.0826 eV) due to the modification of tellurite glasses by (5 to 20) mol% La₂O₃ and from (0.2085 to 0.1619 eV) due to the modification of tellurite glasses by (10 to 50) mol%V2O5. These values are in the range of amorphous semiconductors between 0.046 and 0.66 eV were reported by Davis& Mott [3]. It is clear from these values that the widths of the band tails of the localized states are found to depend upon the kind of the modifier.

Now in the present study, we calculate the electronic polarizability and optical basicity values by using average electronegativity (χ_{2av}) for binary oxide glasses TeO₂-La₂O₃ and TeO₂-V₂O₅ by using Eqs.(4,5,6) which have been stated by Reddy et al [9] as follows

$$(x_{2av}) = \frac{X_{xf} + Y_{xs}}{X + Y}$$
 (4)

$$\alpha_0^{2-} = 3.319 - 0.3422x_{2av} \tag{5}$$

$$\Lambda = 0.04375 + 0.3097 \, X_{2av} \tag{6}$$

The results are listed in Table (2). We notice that, the electronic polarizability and optical basicity values of V_2O_5 -TeO₂ equal the same values of the electronic polarizability and optical basicity of V_2O_5 -GeO₂. The optical basicity of V_2O_5 -TeO₂ show large oxide ion polarizability between 2-3 Å. These results are closed with Reddy et al [9] results.

3.3. I R of Tellurite Glass:

The room temperature IR spectra in the region 200-4000 cm⁻¹ of tellurite glasses containing (5 to 20) mol% La2O3 are shown in Fig. (5-a) and for more details Fig.(5-b) represents the IR spectra in the range 200 - 1000 cm⁻¹. The major absorption of glasses at different composition is summarized in Table (3). The region below 300cm⁻¹ for all samples did not show any sensitivity, possibly due to excessive noise and hence this region has been excluded from the discussion. No water bond or -OH stretching modes are observed in the IR spectra of the crystalline TeO2. But the binary samples exhibited a water band at 3400 cm⁻¹ and an —OH stretching at 2920 cm⁻¹. The main absorption bands are at frequencies around 610-670 cm⁻¹, a shoulder at 580-630 cm⁻¹, a small bands at 560-610 cm⁻¹ and a shoulder at 750-780 cm⁻¹ posses deformed TeO₄ groups as mentioned by Y.Dimitriev [10]. A new shoulder at 580-630 cm⁻¹ was attributed to La-O stretching vibration as measured by A.A.Bahgat [11]. A small band is observed in the spectra at 560-610 cm⁻¹, which may be attributed to Te-O bond vibrations, where the oxygen anions are considered non-bridging (NBO) as measured by E.E.Shaisha [12].

By analogy with the crystalline tellurite [13] it may be accepted that with the introduction of other oxides in tellurite glasses, part of the TeO_4 groups are transformed into TeO_3 . Tellurite containing TeO_4 groups have four band v^s_{eq} , v^{ax}_{eq} , v^{ax}_{eq} , v^{ax}_{ax} and v^s_{ax} modes and the stretching frequencies in the following manner as stated by [14]:

$$v_{eq}^{s} (TeO_2) = 780 \text{ cm}^{-1}$$
 , $v_{ax}^{ax} (TeO_2) = 675 \text{ cm}^{-1}$, $v_{eq}^{ax} (TeO_2) = 714 \text{ cm}^{-1}$, $v_{ax}^{s} (TeO_2) = 635 \text{ cm}^{-1}$.

The infrared spectra in the region 200-4000 cm⁻¹ of tellurite glasses containing (10 - 50) mol%V2O5 are shown in Fig.(6-a), and for more details Fig.(6b) represents the IR spectra in the range 200-1000 cm⁻¹. The major absorption of glasses at different composition is summarized in Table (4). The region below 300 cm⁻¹ for all samples did not show any sensitivity, possibly due to excessive noise and hence this region has been excluded from the discussion. The data exhibited a water band at 3400 cm⁻¹. The infrared spectra consist of a major band in the 650 -670 cm⁻¹, a shoulder at 770-790 cm⁻¹, a band at 980-1000 cm⁻¹, a band at 600-580 cm⁻¹ and a band at 480-500 cm⁻¹. The absorption band at 980 cm⁻¹, attributed to V=O stretching, shifted to 1000 cm⁻¹ with increase V₂O₅ content {this data are closed with Hirashima, [15]]. The band around 480-500cm⁻¹ is due to Te-O-V stretching. Absorption at 650-670 cm⁻¹ and shoulder at 770-790 cm⁻¹ both attributed to TeO3 trigonal pyramid [15] was also observed. On the other hand, absorption at 600 cm⁻¹ is due to the stretching vibration of Te-O_{ax} bond in the deformed TeO4 units [16]. From these results, we conclude that: infrared spectroscopes of vanadate tellurite glasses indicate the transformation of the basic structural unit of TeO₄ to TeO₃ with the addition of modifier.

The quantitative interpretation of the above absorption bands will be based on the experimental wavenumber of each oxide present in the glass as Table (5) and also by using the next relation $\begin{pmatrix} 1 & 1 & 1 \end{pmatrix}$

and also by using the next relation
$$\upsilon = \frac{1}{\lambda} = \left(\frac{1}{2\pi c}\right) \sqrt{\frac{f}{\mu}}$$
 (7)

Where v is a wave number in cm⁻¹, c is the speed of light, f is the force constant of the bond, and μ is the reduced mass of the molecule R-O and

$$\mu = \frac{(m_R m_O)}{(m_R + m_O)} \tag{8}$$

Where μ is the reduced mass of the molecule R-O and m_R , m_0 are the atomic

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weight in kg of cation
$$R$$
 and anion O respectively.
$$f = 17/r^3 (md \text{Å}^{-1}) \tag{9}$$

Where f is the bending or stretching force constant and r bond length of the cation-anion. By adopting the bond length of Te-O, La- O and V-O [17] as tabulated in Table (6). Also Table (6) summarizes the calculated reduced mass of the cation-anion stretching force constant and the theoretical wavenumber by using Eq.(7). From Table (5) and Table (6) it is clear that the theoretical values are in between the experimental values of the pure oxides that used in the present binary glass systems.

4- Conclusions:

The value of E_{opt} increased from (3.51 to 3.61 eV) with increasing La_2O_3 content from (5 to 20 mol.%) and increased from (2.67 to 3.03 eV) with increasing V_2O_5 content from (10 to 50 mol.%). The doping with V_2O_5 decreased the E_{opt} of pure TeO_2 , (E_{opt} of pure TeO_2 =3.79eV), and can attributed to the increase of the NBO atoms, while doping with La_2O_3 gave a lower concentration of NBO atoms than V_2O_5 . The value of E_{ta} decrease from (0.1006 to 0.0826 eV) with increasing La_2O_3 content from (5 to 20 mol%) and from (0.2085 to 0.1619 eV) with increasing V_2O_5 content from (10 to 50 mol%). From these values, the widths of the band tails of the localized states are found to depend upon the kind of the modifier and they are in the range of amorphous semiconductors. From the calculations of the oxide ion average Polarizability (α_0^{2-}) and optical basicity (Λ) for (100-x)TeO₂—(x)V₂O₅, where x change from (10 to 50 mol.%) by using the average electronegativity (χ_{2av}) it has been concluded that:

- (α_0^{2-}) are increased from (2.294 x 10^{-24} cm⁻³ to 2.307 x 10^{-24} cm⁻³) with increasing V₂O₅ content from (10 to 50 mol%),
- While (Λ) are decreased from (0.971 to 0.96) with increasing V₂O₅ content from (10 to 50 mol%).

From this results, the tellurite glass containing V_2O_5 show large oxide ion polarizability between 2-3 Å can be attributed to the empty d orbital of the

corresponding cation, their high coordination number towards oxide ions and relatively large cation Polarizapility.

The IR spectra the ordinary band for pure TeO_2 are 640 cm⁻¹ shift from (610 to 670 cm⁻¹) with increasing La_2O_3 content from (5 to 20 mol%) and from (650 to 670 cm⁻¹) with increasing V_2O_5 content from (10 to 50 mol%). The previous qualitative description will be analyzed quantitatively according to each stretching force constant of bond f, and reduced mass of the cation-anion. We found high correlation between the obtained experimental values of wave number and calculated wave number.

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Table (1):

Optical properties of binary TeO_2 -La₂O₃ and TeO_2 -V₂-O₅ glasses for different percentage of modifier in mol%.

Glass	Cut-off	E _{opt}	Constant	E _{ta}
Composition	Wavelength	(eV)	A	(eV)
	(nm)		(cm ⁻¹ eV ⁻¹)	
TeO ₂ -La ₂ O ₃				
95 - 5	335.5	3.51	5.38	0.1006
92.5- 7.5	333	3.52	4.9	0.0987
90 - 10	331	3.54	4.825	0.0932
87.5- 12.5	329	3.55	4.815	0.927
85 - 15	327	3.578	4.71	0.0916
82.5- 17.5	325	3.59	4.785	0.0871
80 - 20	321	3.61	4.797	0.0826
TeO ₂ -V ₂ O ₅				
90 - 10	413	2.67	3.039	0.2085
80 - 20	409	2.7	3.07	0.2089
75 - 25	400	2.76	2.87	0.245
70 - 30	392.5	2.79	2.84	0.2489
65 - 35	388	2.87	3.03	0.2214
60 - 40	385	2.93	3.22	0.2192
55 - 45	381	2.97	3.21	0.1801
50 - 50	377	3.03	3.38	0.1619

$$\begin{split} & \underline{Table\ (2):} \\ & \text{Average electornegativity}\ (\chi_{2av}),\ optical\ basicity\ \Lambda_{(\chi_{2av})},\ oxide \\ & \text{ion\ polarizability}\ (\alpha_0^{2^-})_{(\chi_{2av})}\ values\ of\ binary\ tellurite\ glasses. \end{split}$$

Glass	Average	Electonic	Optical basicity $\Lambda(\chi_{2av})$	
composition	electornegativity	polarizability		
	(χ _{2av})	$(\alpha_0^{2^-})(x10^{-24}\text{cm}^{-3})$		
TeO ₂ -La ₂ O ₃				
95 - 5	2.98	2.299	0.967	
92.5- 7.5	2.968	2.303	0.963	
90 - 10	2.956	2.307	0.959	
87.5- 12.5	2.944	2.312	0.956	
85 - 15	2.933	2.315	0.952	
82.5- 17.5	2.921	2.319	0.948	
80 - 20	2.909	2.324	0.945	
TeO ₂ -V ₂ O ₅				
10 - 90	2.994	2.294	0.971	
20 - 80	2.985	2.298	0.968	
25 - 75	2.981	2.299	0.967	
30 - 70	2.976	2.301	0.966	
35 - 65	2.972	2.302	0.964	
40 - 60	2.967	2.304	0.963	
45 - 55	2.963	2.305	0.961	
50 - 50	2.959	2.307	0.960	

Table (3):

The experimental IR absorption band position for binary lanthanum tellurite glasses.

Glass	q ₁ (cm ⁻¹)	$Q_2 \text{ (cm}^{-1})$	q ₃ (cm ⁻¹)	q ₄ (cm ⁻¹)	
Composition					
TeO ₂ -La ₂ O ₃					
95 - 5	560	580	610	740	
92.5 – 7.5	560	590	630	750	
90 - 10	560	595	640	760	
87.5 - 12.5	580	600	650	770	
85 - 15	580	610	650	770	
82.5 – 17.5	590	620	655	780	
80 - 20	610	630	670	785	

Table (4):

The experimental IR absorption band position for binary vanadate tellurite glasses.

Glass	q ₁ (cm) ⁻¹	q2 (cm)-1	q ₃ (cm) ⁻¹	q ₄ (cm) ⁻¹	q 5(cm)-1
Composition					
TeO ₂ -V ₂ O ₅					
90 - 10	480	600	650	770	980
80 - 20	485	600	660	780	980
75 - 25	480	600	655	785	980
70 - 30	480	595	660	760	980
65 - 35	485	590	660	780	985
60 - 40	495	585	670	790	990
55 – 45	500	580	665	790	995
50 - 50	500	580	670	790	1000

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Table (5):

The IR absorption band position for pure tellurium and oxides.

Glass	q ₁ (cm) ⁻¹	q ₂ (cm) ⁻¹	q3(cm)-1	q ₄ (cm) ⁻¹	q ₅ (cm) ⁻¹
TeO ₂	340		640	740	
Oxide					
TeO ₂	340		635	780	
La ₂ O ₃	590		640		
V ₂ O ₅	500	550	610	820	1010

Table (6):

The theoretical IR band position of the tellurium, lanthanum and vanadium oxides.

·	Atomic	Rest mass	Reduced mass	Bond	Stretching	Theoretical
Cation	weight	Of cation	Of catio-O	Length	Force constant	Wave number
		(10 ⁻²⁷ kgU ⁻¹)	(10 ⁻²⁶ kgU ⁻¹)	(nm)	(Nm ⁻¹)	(cm ⁻¹)
Te	127.61	211.94	2.361	1.99	215.7	507.08
La	138.91	230.73	2.383	2.517	106.6	354.83
V	50.942	84.615	2.022	1.83	277.4	621.39

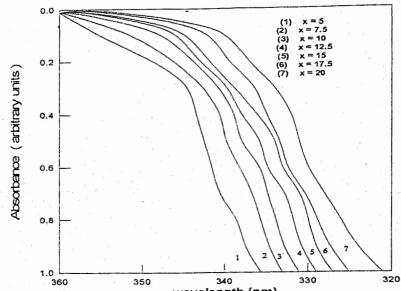


Fig.(1-a): Optical absorption spectra as a function of wavelength for

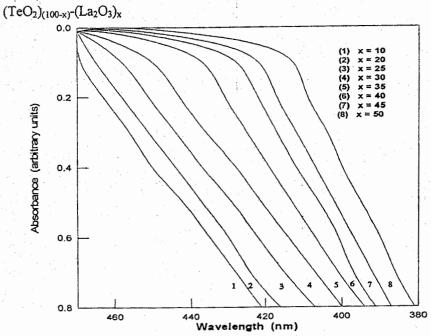


Fig.(1-b): Optical absorption spectra as a function of wavelength for $(\text{TeO}_2)_{100\text{-}x}\text{-}(V_2O_5)_x.$

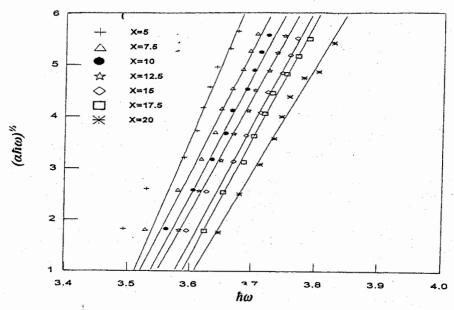


Fig.(2-a): Variation of $(\alpha\hbar\omega)^{1/2}$ with $(\hbar\omega)$ for the binary lanthanum tellurite glasses $(TeO_2)_{(100-x)}$ - $(La_2O_3)_x$

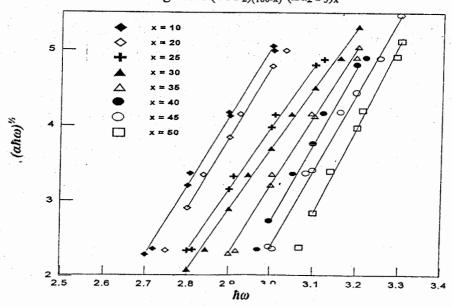


Fig.(2-b): Variation of $(\alpha \hbar \omega)^{1/2}$ with $(\hbar \omega)$ for the binary vanadate tellurite glasses $(\text{TeO}_2)_{100\text{-x}}$ - $(\text{V}_2\text{O}_5)_x$.

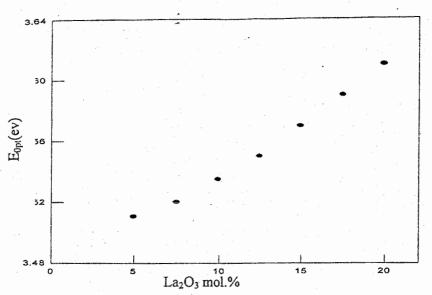


Fig. (3-a): variation of E_{opt} with $(La_2O_3 \text{ mol.}\%)$ for the binary lanthanum tellurite glasses $(TeO_2)_{(100-x)}$ - $(La_2O_3)_x$.

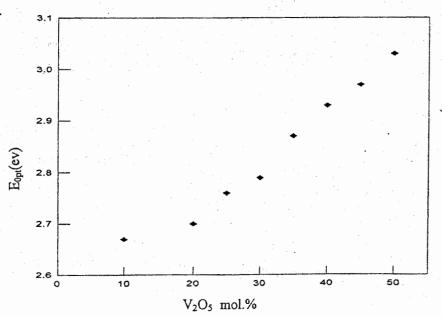


Fig. (3-b): variation of E_{opt} with $(V_2O_5 \text{ mol.}\%)$ for the binary vanadate tellurite glasses $(TeO_2)_{100-x}$ - $(V_2O_5)_x$.

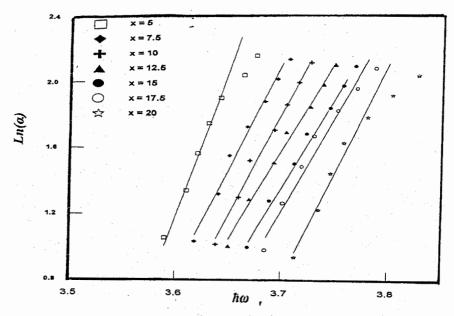


Fig.(4-a): Variation of ln (a) with ($\hbar\omega$) for lanthanum tellurite glasses $(\text{TeO}_2)_{(100\text{-x})}\text{-}(\text{La}_2\text{O}_3)_x\;.$

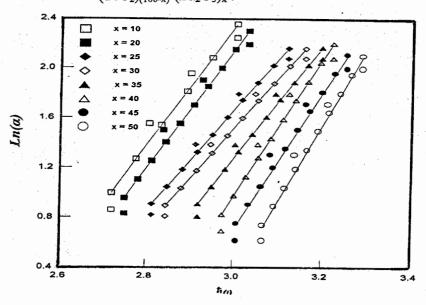


Fig.(4-b): Variation of $\ln (\alpha)$ with $(\hbar \omega)$ for vanadate tellurite glasses $(\text{TeO}_2)_{(100-x)}$ - $(\text{La}_2\text{O}_3)_x$.

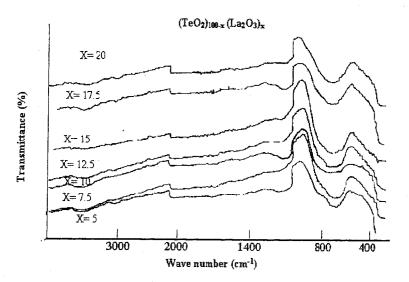


Fig. (5-a) experimental IR spectra of the binary lanthanum tellurite glasses.

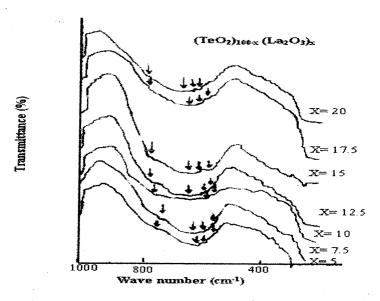


Fig. (5-b) experimental IR absorption bands of the binary lanthanum tellurite glasses in the range 200-1000 $\,\mathrm{cm}^{-1}$

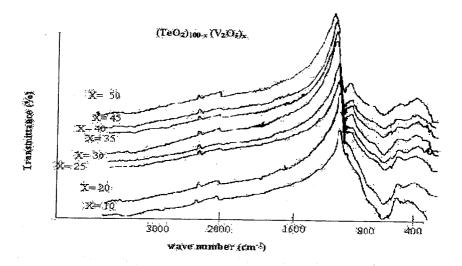


Fig. (6-a) experimental IR spectra of the binary vanadium tellurite glasses.

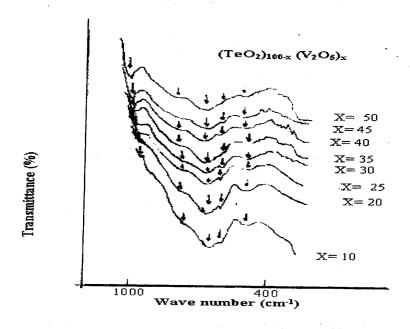


Fig. (6-b) experimental IR absorption bands of the binary vanadium tellurite glasses in the range 200-1000 cm⁻¹.