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Synthesis of Thin Film Ternary Chalcogenide Glasses and Study of its Optical Properties

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ABSTRACT

In the present work, a systematic investigation of Tellurium based ternary chalcogenide semi-conducting materials Se-Te-Ga has been undertaken. The system $Se_{70-x}Te_{30}Ga_x$ ($0<x<10$) has been studied in detail with emphasis on the effect of Gallium in Se-Te binary components. Bulk glasses of Se-Te-Ga systems and thin films are prepared by the conventional melt quenching technique and by thermal evaporation technique, respectively. The effect of compositional variation on some optical constant has been observed and discussed in the present investigation.

I. INTRODUCTION

Chalcogenide glass is a glass containing one or more chalcogens containing Se, Te and S. Such glasses are covalently bonded materials. The classical chalcogenide glasses (mainly sulfur-based ones such as As-S or Ge-S) are strong glass-formers and possess glasses within large concentration regions. Glass-forming abilities decrease with increasing molar weight of constituent elements; i.e., S > Se > Te.

Most stable binary chalcogenide glasses are compounds of a chalcogen and a group 14 or 15 element and may be formed in a wide range of atomic ratios. Ternary glasses [1] are also known. Not all chalcogenide compositions exist in glassy form, though it is possible to find materials with which these non-glass-forming compositions can be alloyed in order to form a glass. An example of this is gallium sulphide-based glasses. In the last few years owing to their interesting properties and technological applications these materials are used in optical and photonic devices. They have good electro-optic, thermo-optic, magneto-optic, acousto-optic properties, high refractive index and I R transparency. Chalcogenide glasses material heat to melt is the form of a liquid and at some instant freeze the position of every atom by quenching. Even in this freezed position they retain short range order and the position of the nearest neighbour remains nearly the same. These glasses are useful in the preparation of passive devices like lenses, windows, fibers etc. [2-4], active devices like laser fiber amplifiers [5] and non-linear components [6-8]. The present work has been carried out in order to investigate some optical properties of $Se_{70-x}Te_{30}Ga_x$ ($x=0,0.5,5,10$) system. The effect of incorporation of Gallium in Se-Te alloy can change the structural and optical properties of the system.

II. EXPERIMENTAL

Extra pure materials were weighed according to their atomic percentage and sealed in 6 cm long quartz ampoules with diameter 6 mm in the vacuum of 10^{-5} torr. The sealed ampoules were kept in a furnace at 700^0C and were held at the same temperature for about 10 hours. During heating, all the ampoules were constantly rocked for better homogenization of the alloys. This was achieved with the help of a ceramic rod to which all the ampoules were tucked in the furnace. Thereafter, the obtained melts were cooled rapidly quenched into the ice cold water. The quenched samples were taken out by breaking the ampoules. The amorphous nature of the glassy alloys was identified by X - Ray diffraction. Thin films of

glassy alloys of thickness (~300 nm) were prepared by thermal evaporation technique in which substrate was kept at room temperature at a base pressure of 10^{-4} torr using the molybdenum boat, the film was kept inside the deposition chamber for 24 hours to achieve the stable equilibrium [9]. The thickness of the film was measured by a single crystal thickness monitor.

III. RESULTS AND DISCUSSION

The value of optical absorption coefficient (α) as a function of incident photon (hν) for deposited thin films of Se-Te-Ga at room temperature is given in table 1.The absorption coefficient (α) is obtained using the relation

Absorption Coefficient (α) =Optical Density(OD)/Thickness(t)

Table 1 depicts that the value of absorption coefficient (α) increases linearly with the increase in photo energy of $Se_{70-x}Te_{30}Ga_x$ .Absorption of light [10] by amorphous solids depends on the energies of the incident photon and on the optical band gap of the material. Thus behavior may be represented by the equation given $\alpha h\nu=B (h\nu - Eg)$ as given in literature[11],where B is a constant which depends on the transition probability, hν is energy of incident photon, Eg is optical band gap of the material, r is an index which depends on the nature of transition [12] where r = 1/2 and r = 2 for allowed direct transition and indirect transition, respectively.

Table 1: Variation of Absorption coefficient (α) with photon energy in a Se-Te-Ga thin films at room temperature.

S. No.	Photon Energy E (eV)	Absorption coefficient (α) in Cm^4			
		$Se_{70}Te_{30}Ga_0$	$Se_{69.5}Te_{30}Ga_{0.5}$	$Se_{65}Te_{30}Ga_5$	$Se_{60}Te_{30}Ga_{10}$
1.	1.378	16555.656	8777.778	-	3988.889
2.	1.459	16555.656	8877.778	3988.989	3988.889
3.	1.549	15000.00	7222.222	4766.767	4766.667
4.	1.907	23555.656	7222.222	18888.989	24333.333
5.	2.066	39888.989	16555.556	29122.322	39888.889
6.	2.254	63222.232	30555.556	50877.878	57800.000
7.	2.479	80333.433	43000.000	74788.889	71000.000
8.	2.754	111111.231	57000.000	111111.111	111111.111
9.	3.541	122777.878	89666.667	173333.333	147111.111
10.	4.131	227777.878	227777.778	266666.667	266666.667

The present system obeys the rule of indirect transition and the relation between optical gap, optical coefficient (α) and the energy hν of the incident photon is given by $(\alpha h\nu)^{1/2} = A(h\nu - Eg)$ where α obeys - Urbach relation [13] $\alpha = \exp [A(h\nu - h\nu_0)]/kT$ where A is a constant of the order of unity, ν_0 is the constant corresponding to the lowest excitonic frequency.

The variation of $(\alpha h\nu)^{1/2}$ with photon energy hν for Se-Te-Ga films is given in table 3. The calculated values of Eg are given in table 2. It is evident from the table that the value of optical band (Eg) increases from 1.25 to 1.33 with increasing Ga content. The increase in the Eg with increasing Ga may be due to increase in grain size, the reduction in the disorder and decrease in density of defect states.

Table 2: Optical parameter Se-Te-Ga thin film at 700 nm at RT

X	E_g (eV)	α (Cm^{-1}) (10^4)	N	k (10^{-3})	$\varepsilon_r^{'}$	$\varepsilon_r^{''}$	R%	T%
0	1.51	0.70	2.01	0.39	4.07	0.002	15.63	65.62
0.5	1.25	0.38	2.57	0.21	6.63	0.001	29.16	70.62
5	1.33	1.16	5.27	0.65	27.86	0.006	75.62	58.33
10	1.27	1.16	2.66	0.65	7.08	0.003	31.25	64.16

Table-3: Variation of $(\alpha h\nu)^{1/2}$ with photon energy in a Se-Te-Ga thin films at Room Temperature.

S. No.	Photon Energy E (eV)	Absorption coefficient (α) in Cm^4			
		$Se_{75}In_{25}$ Pb_0	$Se_{75}In_{21}$ Pb_4	$Se_{75}In_{19}$ Pb_6	$Se_{75}In_{15}$ Pb_{10}
1.	1.377	146.338	103.477	N/A	73.169
2.	1.441	150.581	106.477	75.290	75.290
3.	1.511	147.250	98.167	85.015	85.015
4.	1.588	124.172	87.803	101.386	113.353
5.	1.674	111.310	82.966	143.701	143.701
6.	1.770	207.351	108.906	184.659	210.896
7.	1.877	283.382	179.227	237.095	283.382
8.	1.998	374.392	258.032	334.867	355.180
9.	2.136	443.382	322.608	427.898	416.485
10.	2.294	527.629	392.666	527.629	527.629
11.	2.478	580.761	478.404	649.311	620.860
12.	2.693	631.849	560.250	760.395	694.142
13.	2.950	948.379	948.379	1029.579	1029.579
14.	3.261	-	-	-	-

The value of Eg decreases from 1.33 to 1.27 with increasing Ga concentration in $Se_{70-x}Te_{30}Ga_x$ sample, since the optical absorption depends on the short range order in the amorphous state and defects associated with it. The decrease in the optical band gap in the present system may be due to reduction of the amount of disorder in the system and increase in the density of defect state which is attributed to the shift in Fermi level whose position is determined by the distribution of electrons over the localized states.The values of the variation of $(\alpha h\nu)^{1/2}$ with photon energy (hv) for $Se_{70-x}Te_{30}Ga_x$ films are given in the table 3.

The variation of reflectance (R) in percentage for the present system of Se-Te-Ga is shown in table 4. The table shows that the reflectance (R) increases with the increase in wavelength. Similarly, the transmittance (T) has also been shown for Se-Te-Ga in table 3. It is evident from the table that the percentage in the transmission increases with respect to wavelength. It is clear from the table that the value of the transmission starts decreasing after about 600 nm.

Table 4: Variation of reflection (R) in % with wave length in a Se_{70-x} Te_{30} Ga_x thin films at room temperature.

S. No.	Wave length in nm	Reflection (R) in %			
		Se-Te-Ga	Se-Te-Ga	Se-Te-Ga	Se-Te-Ga
1.	900	88.500	62.500	31.250	28.125
2.	800	71.833	54.583	60.417	13.750
3.	700	15.625	29.167	75.625	31.250
4.	600	46.875	14.583	30.208	41.667
5.	500	31.667	32.500	41.667	28.125
6.	400	30.208	31.250	36.458	31.667
7.	300	28.333	22.917	29.167	22.917

Table-5: Variation of Transmission (T) in % with wave length in a Se-Te-Ga thin films at Room Temperature.

S. No.	Wave length in nm	Transmission (T) in %			
		Se-Te-Ga	Se-Te-Ga	Se-Te-Ga	Se-Te-Ga
1.	900	40.000	56.250	75.000	82.500
2.	800	44.792	61.458	78.125	85.938
3.	700	65.625	70.625	58.333	64.167
4.	600	14.583	48.958	19.792	21.771
5.	500	2.083	15.625	3.125	3.438
6.	400	-	3.125	0.208	0.229
7.	300	-	0.208	-	-

The refractive index (n) and extinction coefficient (k) have been calculated by using the theory of reflectivity of light for the above sample discussed. According to this theory, the reflectance of light from a this film can be expressed in terms of fresnel's coefficient. The reflectivity [14-15] on a interface can be given by-$\mathbf{R=[(n-1)^2 +k^2]/[(n+1)^2 + k^2}$**and** $\alpha=4\pi K/\lambda$

The optical investigations shows that the optical band gap increases upto 4% of Ga concentration in Se-Te-Ga, with further increase of Ga content the optical band gap decreases in the present system.

CONCLUSION

From the reflectance and transmittance studies of the film of Se-Te-Ga, it may be concluded that the refractive index (n) decreases, while the value of the extinction coefficient k increases with photon energy. The increase in the optical band gap with increasing Ga content may be due to the increase in the grain size, the reduction in the disorder and decrease in the density of defect states. The value of optical band gap (Eg) decreases at higher Ga concentration. The decrease in the optical band gap in the present system may be due to reduction in the amount of disorder in the system and increase in the density of defect states.

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