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Some Physical Properties of γ Irradiated Ge_x(As₂Te₃)_{100-x} Chalcogenide System

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Abstract

The effect of γ -radiation on the electrical conductivity, switching, and optical properties of the chalcogenide amorphous system $\text{Ge}_x(\text{As}_2\text{Te}_3)_{100-x}$ (where x = 0,1,5,10 atm %) have been studied. The results show that the radiation causes a shift of the optical gap, as well as a change in the electrical activation energy and the threshold voltage. As the γ -doses increase, the values of the allowed indirect optical energy gap E_{opt} for the different compositions decreases and the tail energy width increases.

Key Words: Chalcogenide-glasses; gamma-ray effects; visible-spectra, radiative-electrical-optical-properties; gamma-irradiation; transmission-edge-shift; defect-formation.

1. Introduction

Chalcogenide glasses have attracted attention for potential application in various solid state devices as acoustic optic devices and non linear optical devices [1-4]. They are acoustically hard materials with high impedance and may be classified as low attenuating solids [5,6]. The switching process observed in amorphous semiconductors is characterized not only by the breakdown of the high resistance state of the material but also very importantly by the presence of a positive feedback mechanism that provide the high conductance on-state. The various models proposed to explain the switching process may be categorized into homogeneous and heterogeneous models.

Many studies have been made on the switching phenomenon in chalcogenide glasses and its cause has been classified into those initiated thermally [7,8] and those initiated electronically [9-13].

The absorption of γ -radiation in chalcogenide alloys bulk and thin films depends strongly upon their electronic structure which in turn changes by the interaction with photons. The additional absorption of Te-containing glass fibers is due to the increase in the number of thermally excited free carriers with Te content, analogous to Ge and GaAs semiconductors [14].

Chalcogenide glasses are important for infrared transmission as active electronic device components for photocopying, ultramicrolithogrphy, and electronic switching [15]. It has also been shown that gamma irradiation of samples of chalcogenide glasses of As_2S_3 and Ge_2S_3 by an absorbed dose of 4.4×10^6 Gy leads to a longwave shift of their optical-transmission edge in the spectrum. The effect observed depends on the structural type of the glasses investigated. It has been suggested that the microstructural mechanism of

these changes is attributed to processes of coordination defect formation in the structural skeleton of the samples [16].

Ion-irradiation induced defect states near the Fermi level play a dominant role in the variable range hopping conduction. Bipolaron hopping conduction appears to be affected less by ion-irradiation and is quite effective to modify the electrical transport behaviour of the glass [17]. The only reported EXAFS work on amorphous selenium (a-Se) showed that the disorder in the material increases upon light irradiation [18].

The aim of the present work is to study the effect of γ -irradiation on the electrical, switching and optical properties of the $\text{Ge}_x(\text{As}_2\text{Te}_3)_{100-x}$ chalcogenide system.

2. Experimental Techniques

Bulk glasses of composition $\text{Ge}_x(\text{AsTe})_{100-x}$, (where $0 \leq = x \leq = 10 \text{ at\%}$) were prepared from 5N pure elements, Ge, As and Te in an evacuated silica ampoul at 1000^{0} C for 12 hours under vacuum of 10^{-6} torr in a rocking furnace and quenched in ice water. The prepared bulk amorphous flakes which have been obtained by the melt quench method, were used to prepare thin films of $\text{Ge}_x(\text{As}_2\text{Te}_3)_{100-x}$ by thermal evaporating techniques on glass substrate maintained at 300K under vacuum of 10^{-6} torr. The rate of deposition was kept constant at 30 nm /s for all thin films using Edwared Coating unit (306 E). The films used were characterized by a homogenous deposition. The film thickness was controlled using quartz crystal thickness monitor and was confirmed optically. The composition of the bulk was investigated using Energy Dispersive X-Ray (EDEX) Spectroscopy for $\text{Ge}_{10}\text{As}_{36}\text{Te}_{54}$ as an example (Table 1).

The optical constants were determined from both transmittance (T) and reflectance (R), which were carried out at normal incidence using a double beam Spectrophotometer (Shimadzu model 3101) over the wavelength range 200nm - 2500nm.

For electrical measurements, samples in the form of small thin flakes 1 mm thick and thin films of thickness 0.115 μ m were used for the present work. The bulk sample was placed on a thin copper plate. A platinum probe attached to a very light pressure spring was used in contact with the top surface to maintain constant contact pressure. The current-voltage characteristics and the electrical resistance as a function of temperature for unradiated and irradiated bulk samples were carried out.. The whole sample holder was placed in a thermostatically controlled oven for measurement at higher temperatures. The current was measured using a series standard resistance (100 Ω) and a Keithley electrometer 610C. The voltage was monitored using a null detector Keithley electrometer 155.

Irradiation for bulk samples and thin films with doses (1,2,3,and 4 kGy) was performed using a Co^{60} gamma ray source model GB 150 Type B manufactured by the atomic Energy of Canada.

3. Results and Discussion

3.1. Electrical resistance

The electrical resistance R has been plotted as a function of temperature for the unradiated and the irradiated bulk samples at different doses (Figure 1a-d). The relation of log R versus $10^3/T$ is exactly linear with two distinct regions showing Arrhenius plots which obey the relation:



Figure 1a-d. The logarithm of the resistance R vs 10^3 /T of the unradiated and irradiated Ge_x(As₂Te₃)₁₀bulk samples at different doses.

$$R = R_0 \exp(W_H / kT) \tag{1}$$

where R_0 is the pre-exponential factor, and W_H is the activation energy. For the binary composition As₂Te₃ (Ge₀), it is noticed that R decreases with $10^3/T$ for 1kGy dose and then begins to increase for 2kGy and 3kGy doses, approaching the initial value. However, its change with 4kGy dose is oscillating. This may be attributed to the increase of the disorder of the samples by irradiation. The values of the hopping energy W_H are listed in Table 2 and are in accordance with the commonly accepted values published before [19,20]. For 1kGy dose, the decrease of the value of W_H is believed to result from the phase-separated morphology of these glasses and from the less rigid nature of the network of one of these phases [21]. These glasses seemed to have a chain or laminal structure and their electrical properties depended mainly on the covalent

bonds in the chain. The considerable decrease in the activation energy might be due to the breaking of these bonds. With the exception of W_H for composition $Ge_5(As_2Te_3)_{95}$ the hopping energy increases at 2kGy and 3kGy. The anomalous behaviour of the electrical resistance may be due to the small creation energy for defect centers as $C_3^+ - C_1^-$ or $C_3^+ - P_2^-$ and $C_3^+ - T_1^-$ pairs [22,23].

	Item	Min	Max	Mean	Stddev
Atomic%	Ge	9.800	11.840	10.650	1.062
Atomic%	AS	24.300	28.350	26.190	2.038
Atomic%	Te	59.810	65.390	63.160	2.954

Table 1. Energy Dispersive X-Ray (EDEX) Spectroscopy for bulk Ge₁₀As₃₆Te₅₄.

Table 2. 1-(W,W_H)(eV)(Total activation energy and hopping energy) and W_D(eV)(Disordered energy)-2,3-Optical parameters(E_{opt}(eV),A (cmeV),E_e(eV), α_o (cm)⁻¹ and finally V_{th}(V) (thrld voltage) respectively.

Dose					
(kGy)	As_2Te_3	$\operatorname{Ge}_1(\operatorname{As}_2\operatorname{Te}_3)_{99}$	$\operatorname{Ge}_5(\operatorname{As}_2\operatorname{Te}_3)_{95}$	$Ge_{10}(As_2Te_3)_{90}$	
0	1-(0.285,0.255,	1-(0.237,0.285, 1.592,	1-(0.37,0.283,	1-(0.295,0.196,	
	1.435,0.993),0.058	1.205),0.1	1.124,1.114),0.174	1.205,0.994),0.178	
	2-0.9, 8.16×10^6	2-0.71, 5.1x10 ⁵	2-0.63, 3.78x10 ⁸	$2-0.58$, 1.51×10^5	
	3- 0.14 , 29,112	3-0.191,93.8,143	3-0.22,125.6, 225	3-0.407, 304 , 275	
1	1-(0.088,0.02,0.218,	1-(0.251,0.214),	1-(0.347,0.289,	1-(0.259, 0.196,	
	0.124) ,0.0869	,), 0.071	1.214,1.169),0.111	0.990,0.449),0.117	
	2-0.73, 2.22x10 ⁴	2-0.56, 6.93×10^4	$2-0.44$, 3.31×10^4	$2-0.38$, 2.78×10^4	
	3- 0.158,6.44,130	3-0.201, 13.3, 138	3-0.23,11.95, 222	3-0.415, 127, 118	
2	1-(0.175,0.102,	1-(0.261, 0.258,	1-(0.361, 0.30,	1-(0.486, 0.387,	
	0.237,0.227),0.126	1.293, 1.096) ,0.22	0.844 ,0.77), 0.072	0.646,0.598),0.255	
	2- 0.57, 2.97x10 ⁴	$2-0.44$, 4.34×10^4	$2-0.44$, 3.31×10^4	$2-0.38$, 2.78×10^4	
	3-0.17, 3.86, 150	3-0.21, 9.14 , 150	3-0.24 , 31.38, 265	3-0.419, 269,141	
3	1-(0.37,0.159	1-(0.259, 0.189,	1-(0.545 , 0.399,	1-(0.224, 0.099,	
	1.748,0.519),0.334	1.592,1.178),0.1292-	0.723,0.62), 0.333	,0.603), 0.047	
	$2-0.57, 2.97 \times 10^4$,0.44, 4.31x10 ⁴ 3-	$2-0.44, 3.31 \times 10^4$	$2-0.38, 2.78 \times 10^4$	
	3-0.193,12.98,200	0.215, 26.61,200	3-0.245, 21.02,275	3-0.421, 209, 204	
4	1-(,,	1-(0.329,0.181,	1-(0.548, 0.403,	1-0.224, 0.099,	
	1.785,) ,	1.729,0.846),0.252	0.703,0.598),0.268	1.389,1.014),0.047	
	2- 0.9, 8.16×10^6	2-0.56, 6.93×10^4	2-0.44, 3.31x10 ⁴	$2-0.38, 2.78 \times 10^4$	
	3-0.21, 2.4, 242	3-0.231, 70.95,242	3-0.25 ,37.85, 320	3-0.422, 155, 330	

In the beginning, the decrease of resistivity by γ -photons doses may be due to the increase of the generations of free carriers, the breaking of bonds and the creation of electron-hole pairs. For materials with large density of defects, the free carriers generated by reaction of γ -photons doses are trapped. The increase of γ -doses leads to the creation of charged recombination centers for the opposite type of carriers,

the densification of material and the formation of point defects [24]. Consequently, this leads to the increase of the electrical resistivity. Further, these results can be discussed in terms of the possible dimensional structural changes, "the size effect" and the change in the local stoichiometry. Thus, the change in the resistivity enhanced by radiation is not a structural change and it is not associated with any crystallization process.

One of the main advantages of the amorphous semiconductors is their low sensitivity to radiation. The random field in such glass separates the energies of an electron on the sites by W_D due to electron delocalization effects. Defect bonding configurations, such as dangling bonds and valence-alternation pairs are usually only present in low concentrations of less than one per 10⁴ atoms which are very important in electrical transport [25]. Then the total energy Wcan be calculated using the following relation [26]:

$$\sigma T^{1/2} \alpha \exp(W/kT) \tag{2}$$

From the plots of $\ln \sigma T^{1/2}$ vs 1/T (where σ is the conductivity) the activation energy W was calculated [26]. The activation energy W as given in the following equation [27]:

$$W = (W_D + 4W_H)^2 / 16W_H \tag{3}$$

consists of two terms: one is the hopping energy W_H and the other is the disordered energy W_D , which can be used as an indication of the degree of disorder in the system.

The values obtained for W, W_H , and W_D are given in table (2). W_D is the disordered energy which might exist between the localized states. As shown in the table, W_D decreases for the first dose and increases as the radiation doses increase for all compositions showing the increase of disorder. The calculated values of W_D are smaller than W_H as predicted for other chalcogenide glasses [28,20] (Table 2).

3.2. Switching effect

The results of static I-V characteristic for bulk samples (1 mm thick) with different doses for the four glassy compositions are studied, example in $Ge_{10}(As_2Te_3)_{90}$, (Figure 2)). All compositions showed the same behaviour where V_{th} decreases at 1kGy dose and then increases as the value of the dose increases (Table 2).



Figure 2. The static I-V characteristic for $Ge_{10}(As_2Te_3)_{90}$ samples (1 mm thick).

The switching characteristics of multicomponents chalcogenide glasses [29-31] and the threshold switching phenomenon in chalcogenide semiconductor have been studied extensively. It is believed that the switching phenomenon in the bulk devices is initiated by a current-aided thermal process or an energy-controlled process [32-34]. The results show that there is a critical field (V_{th}) below which the material remains in a low conducting state (OFF state). The non-linear behaviour before switching to the negative state is discussed according to the Poole-Frenkel field emission of electrons from deep traps and modification of the energy difference between filled and empty sites [35]. Further, the nonlinearities are determined largely by the abundance of the most-polarizable constituent [36,37]. Above this field the Joule heating term predominates and conductivity rises rapidly. It is proposed therefore that thermal effects play an important part in the electrical behaviour [38]. The defects in the sample cause a change in the thermal conductivity and structure of the material. The current in the highly resistive state is not electrode limited but the electric field is essentially uniform through the bulk of the material. Under the effect of the energy of the photons, the trap centers become less effective, and the movement of carriers may be hindered mainly by the scattering process. The interaction of photons with the glass in the filament region causes a rapid rise in temperature which is assumed to cause a localized change in the structure of the glass, by breaking up the lattice bonding, thus, leaving many dangling bonds [39] and increasing the threshold voltage for switching.

3.3. Optical properties

The substrate effects and interface reflections were eliminated from the observed transmittance (T) and reflectance (R). The transmittance (T_f) and reflactance (R_f) of the film are obtained by the aid of the following relations:

$$T_f = T_{0bs} (1 - R_{obs} R_s) / (1 + R_s) \tag{4}$$

and

$$R_f = R_{obs} - [T_{(f)}^2 R_s / (1 - R_{obs} R_s)]$$
(5)

where $R_s = (n_s - 1)^2/(n_s + 1)^2$, n_s is the refractive index of the glass (1.54), and T_{0bs} and R_{obs} are the observed transmittance and reflectance of the film deposited on glass substrate.

Using transmittrance and reflectance $(T_f \text{ and } R_f)$ of each film the absorption coefficient α can be calculated using the following relation [40]:

$$T_f = (1 - R_f)^2 \exp(-\alpha d) \tag{6}$$

Where, α is the absorption coefficient and d is the film thickness. (Figure 3) show the dependence of α on the photon energy E at different γ -doses example for Ge₁₀(As₂Te₃)₉₀ samples. It is observed that the absorption decreases with radiation doses for all samples. This may be attributed to the activation of the recombination processes induced by irradiation. For higher values [$\alpha > 10^4$ cm⁻¹], the absorption coefficient α (where the absorption is associated with interband transitions) yields the power part of the relation [41-43]:



Figure 3. Relation between the optical absorption coefficient (α) and the photon energy(E) for Ge₁₀(As₂Te₃)₉₀ thin films.

$$\alpha * h\nu = \beta (h\nu - E_q)^m \tag{7}$$

where β is a constant and m is the power which characterizes the type of optical transition process, (m = 2 for the indirect optical transitions). The energy gaps were determined by plotting $(\alpha h\nu)^{1/2}$ vs E = h ν as constructed in Figure 4 for Ge₁(As₂Te₃)₉₉ γ -irradiated films with different γ -doses. These spectra show the evolution of permanent changes with γ -irradiation and indicates that the absorption edges for the allowed indirect transition shift to low photon energies as the γ -doses increase. The reported values of E_{opt} for all samples are listed in Table 2, with those previously published for unirradiated Ge_x(As₂Te₃) [19]. Absorption of photons creates electron-hole pairs. In turn, the field of such pairs may modify the electronic structure and hence optical properties of the Ge_x(As₂Te₃)_{100-x} glass.



Figure 4. Plots of $(\alpha E)^{0.5}$ versus (E) for $Ge_1(As_2Te_3)_{99}$ thin films.

In the exponential edge region where $(1 < \alpha < 10^4 \text{ cm}^{-1})$, the absorption coefficient α is governed by the relation [44].

$$\alpha = \alpha_0 \exp(h\nu/E_c) \tag{8}$$

where α_0 is a constant and E_c is the Urbach energy, interpreted as the width of the tails of localized states in the band gap. The band tail width for irradiated samples was calculated by plotting $\ln \alpha$ vs $E = h\nu$ for all thin films Figure 5 (for Ge₁₀ as example). The obtained values of E_c are listed in table (2) with those previously published for unirradiated Ge_x(As₂Te₃)_{100-x} [19].



Figure 5. Relation between $\ln \alpha$ and (E) for $Ge_{10}(As_2Te_3)_{90}$ thin films.

The increase of γ -doses would lead to systematic changes in density of states. Some of these changes were recovered when the γ -doses were turned off. The Urbach tail was relaxed by photoirradiation and recovered, an effect which is associated with the effect of the photoenhanced far-infrared band broadening in amorphous-chalcogenide films [45]. Hopping conduction in the tail of localized states was proposed in parallelism with conduction in extended states [46]. It can be noticed that the values of E_c increase as γ doses increase. So we can conclude that γ -doses cause the breaking of bonds leading in turn to the increase of dangling bonds and of defects, as well as the trapping of the generated carriers. This may be the cause for high resistance, high threshold voltage and the increase in band tail width. The dose dependence of Gamma radiation induced densification and compaction in $Ge_x(As_2Te_3)_{100-x}$ by the total energy absorbed. Moreover, the density change was found to be consistent for all radiation types [47].

The optical properties of unirradiated $Ge_x(As_2Te_3)_{100-x}$ thin films were studied and the optical absorption was found to be due to indirect transition [19]. Furthermore, the band tail width obeyed the Urbach's empirical relation.

4. Conclusion

The effect of γ irradiation on the electrical and switching properties for $\text{Ge}_x(\text{As}_2\text{Te}_3)_{100-x}$ were studied. It is noticed that R decreases with T 1kGy dose and then begin to increase for the higher doses. This may be attributed to the increase in the amorphousity and/or the increase of the disorder of the samples by

irradiation. The disordered energy W_D decreases at 1kGy dose and increases as the radiation doses increase for all compositions showing the increase of disorder and compaction.

The optical properties are influenced by irradiation. The absorption coefficient decreases, the same behaviour for the optical energy gap E_{opt} shifted to lower energy and the values of E_c which represent the band tail width of the localized states in the forbidden gap increase as γ -doses increase. So we can conclude that γ - doses cause the breaking of bonds leading to the increase of dangling bonds and of defects, as well as the trapping of the generated carriers.

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