

EFFECT OF γ -RADIATION DOSE ON THE OPTICAL PROPERTIES OF $(AsTe)_x(GeSe_2)_{100-x}$ ($x = 5$) THIN FILMS

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The present study deals with the effect of γ -irradiation doses in the range of (100-500 kGy) on the optical properties of quaternary $(AsTe)_5(GeSe_2)_{95}$ film. The complex index of refraction (real (n) and real (k)) and the film thickness for the as-prepared and exposed films were determined precisely based only the measured transmittance spectra. A clear red shift of the absorption edge was noted by increasing γ -irradiating doses. Such a shift leading to the observed decrease in the optical gap E_{og} . Such a behavior of E_{og} can be due to the induced defects in the films due to γ -irradiation. The index of refraction (n) has been discussed in terms of the dispersion model. The dispersion parameters were determined and well discussed in detail. The observed results show that, the $(AsTe)_5(GeSe_2)_{95}$ thin films can be employed as radiation dosimeter for radiation measurements.

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1. Introduction

In the last few decades, chalcogenide glasses which include one or more of chalcogen elements (Se, S Te) have been extensively studied due to their excellent properties and their uses in many of the technology devices [1-5]. Adding some impurities to chalcogenide glasses will be affect their physical properties. In Recent, the impurities effect on the electrical and optical properties of chalcogenide compounds as well as their thermal analysis has attracted the attention of many researchers. The electrical properties of As_2Se_3 glasses has been strongly affected when small amounts of Ga, In, Ag and/or Cu used as impurities [6-9].

Nowadays, A. Othman et al. [10] have been study the compositional effect on optical, photoelectrical and thermal properties of some As-Sb-Se chalcogenide glasses doped with Te. Chalcogenide glasses have been extensively studied because of their using in solar cells, thermal imaging detectors, optoelectronic devices, optical recording materials, optical waveguides, bio and chemical sensors [11-14]. Extensive studies were carried out to show the impurity effects on the physical properties viz. thermal, optical and electrical of their glasses [15-18]. Radiation induced effects in the optical and electrical properties of glasses and thin films [19-22]. These effects may be reversible or irreversible, allowing the use of chalcogenides in many technological applications such as high dens informatics storage, devices with large resolution display and in the fabricating the diffractive elements [23, 24]. Thermally- and photo-induced phenomena in a- $Ge_{30}S_{60}Sb_{10}$ films were investigated by Vlček et al. [25-27]. They explain the variations in the optical constants of these films in terms of the change in the densities of homo-polar bonds. Chalcogenides based IV, V and VI groups that allow the tuning of structure flexibility with composition, displays variety of both metastable and transient [28] photo induced phenomena including photo-darkening [29, 30], photo-bleaching [31] and photo-structural change [32].

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The present study comes as a continuation to our previous studies on many amorphous ternary and quaternary systems and their thin films. this work aimed to synthesis amorphous thin films have high homogeneity nature from the quaternary bulk $(As_1Te_1)(Ge_3Se_7)$ glass, where such these glasses were used over a wide range in several electronic industries such as photovoltaic, digital thermometers, sensors and many electronic devices. This work aims also to study the γ -irradiation effects of complex index of refraction $n^* = n + i k$.

2. Experimental

Conventional melt quenching technique and thermal evaporation method were used to investigates of $(AsTe)_5(GeSe_2)_{95}$ glass in the forms of bulk and thin films, respectively. Further details the perpetration methods have been detailed here [33, 34].

$(As_1Te_1)(Ge_3Se_7)$ films were exposed to γ -rays produced from a ^{60}Co -source in 4000/A Indian g-chamber with average rate of 1 Gy/s. The different doses have been controlled within the range of 100–500 kGy. JASCO-670 spectrophotometer with double beam was used for measuring the film (as-prepared and irradiated) transmittivity (T) over spectral range $400 \leq \lambda \leq 2500$ nm.

3. Results and discussions

3.1 Dispersion of the refractive index

The measured film transmittivity of the as-prepared and γ -irradiated films were represented in Figure 1. The inset of this figure shows the absorption edge changes due to γ -irradiation. A clear red shift of the absorption edge which increases by increasing the γ -irradiation doses from 100 to 500 kGy. The observed interference fringes with sharply falling at the edge of the fundamental absorption confirmed the uniformity and smoothness of the as-prepared and irradiated films. The thickness (d), refractive index of (n) and extinction coefficient (k) for the films were exactly determined using the maxima $T_{MA}(\lambda)$ and minima $T_{min}(\lambda)$ of the interference fringes of the measured transmittance $T(\lambda)$. Such analysis was suggested by Manifacier [35] and expanded by Swanepoel [36]. Fig.2 represents the transmittance $T(\lambda)$ of the as-deposited film (as an example), the generated two envelopes $T_{MA}(\lambda)$ and minima $T_{min}(\lambda)$, the geometrical mean $T_{gm}(\lambda) = (T_{Ma}(\lambda) \cdot T_{min}(\lambda))^{0.5}$ and measured substrates transmittance $T_{sub}(\lambda)$. The details of procedures used to calculate t, n and k for the films were written in our published papers [37-40]. The t value for the as-prepared film is 880 ± 7 nm.

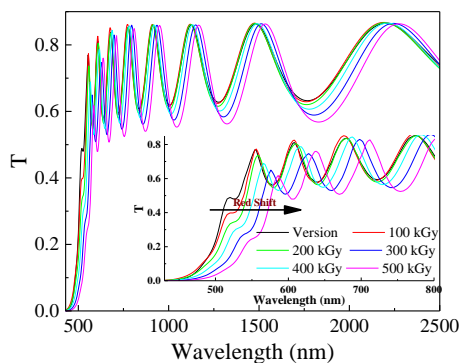


Fig. 1. The measured transmittance spectra for the as-prepared (Version) and exposed film to different doses of γ -radiation.

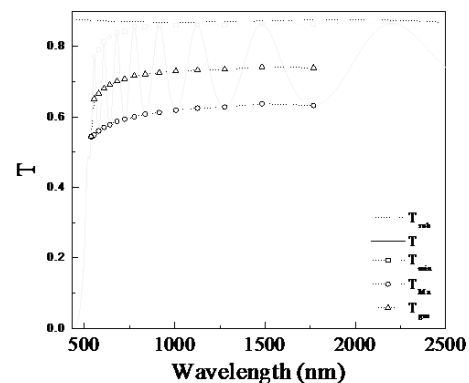


Fig. 2. The measured transmittance of the substrate T_{sub} and the as-prepared film (T), the two envelopes T_{Ma} and T_{min} and tier geometrical mean T_{gm} .

Fig. 3 represents the index of refraction dispersion for the as-prepared film and the exposed to γ -irradiation with different doses for $(AsTe)_5(GeSe_2)_{95}$ chalcogenide glassy films. Increasing the γ -radiation doses results in an increase of the index of refraction.

This may be explained by increased the film density due to irradiation because of atomic displacements or ionization results due to the collisions of γ -rays with the glass atoms that could be materially altered or change internal structure of the film. Moreover, the index of refraction for the as-prepared and exposed films with different doses of $(AsTe)_5(GeSe_2)_{95}$ thin films was fitted according to the second order Cauchy's dispersion relation:

$$n(\lambda) = a + b/\lambda^2 \quad (1)$$

where a and b are fitting parameters depending on material (solid lines in Fig. 3). The oscillating parameters viz. oscillator energy, E_0 and the dispersion energy E_d were investigated based on Wemple–DiDomenico model. According to this model the index of refraction parameter $(n^2 - 1)^{-1}$ were plotted as a function of the incident photon energy $(h\nu)^2$ as well as represented in Fig.2 . Based on the slope and intersect of this figure, it's easy to deduce both the E_0 and E_d . Furthermore, the $n(h\nu = 0)$ can be deduced from the intersect of these plots with the Y-axis. Recently, Aly et al. suggest that, the infinite wavelength (λ_0) is correlated to the E_0 and E_d values through the following expression:

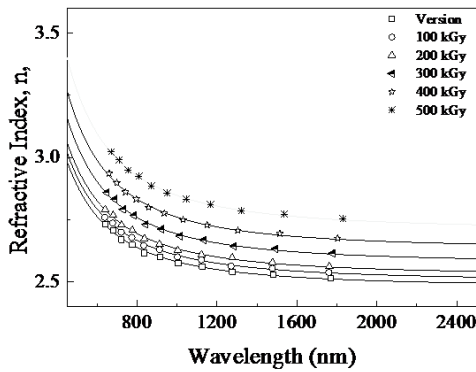


Fig. 3. The index of refraction $n(\lambda)$ for the as-prepared and exposed films.

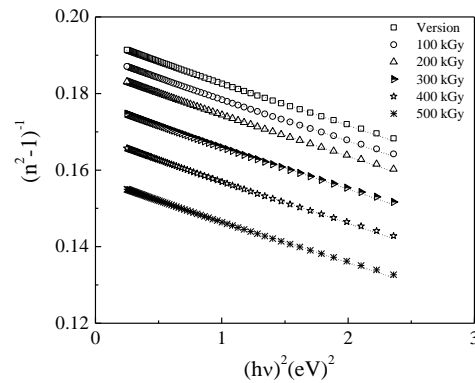


Fig. 4. $(n^2 - 1)^{-1}$ versus $(h\nu)^2$ for the as-prepared and exposed films.

$$\lambda_0 = hc \sqrt{(n_0^2 - 1)/(E_d E_0)} \quad (2)$$

With the help of λ_0 value the oscillator strength (s) can be calculated through the relationship ($s = (n_0^2 - 1)/\lambda_0^2$). The deduced values of the dispersion parameters (E_0 , E_d , $n(0)$, λ_0 , s) and the static dielectric constant ($\epsilon = (n(0))^2$) were listed in Table.1 for the as-prepared and exposed films. It clearly that, the E_0 values of decreases, while the E_d , $n(0)$ and ϵ values were increases with the increase of γ -irradiation doses from 100 to 500 kGy. The E_0 value is scales the energy gap i.e $E_0 \approx 2 E_{og}$ [37] (see the ratio of E_0/E_{og} in Table 1). Also, the changes in E_0 values due to the γ -irradiation doses can attributed to the increase of the localized state width. The E_d considered as inter-band strength value is affected by the real part of the dielectric constant (ϵ_2). In present films, the n values are increased indicating that, the area under the absorption curve is larger. Therefore, the film transmissivity shifted to the larger wavelengths side. The two parts of the dielectric constant (real (ϵ_2) and imaginary (ϵ_2)) are correlated to the index of refraction (n) and absorption coefficient (αa), respectively, any increase in the values of (αa) an indication of the absorption edge shifts towards the larger wavelengths, i.e., red shift with increase of radiation doses is benefiting an increase in the localized states density in forbidden gap. The 3ed non-linear optical susceptibility ($\chi^{(3)}$) for glassy films is a significant parameter that, determine the use of

that material in non-linear optical devices. In accordance with Miller generalized role in the limit ($v \rightarrow 0$), the value of $\chi(3)$ for the as-prepared and exposed films using the following formula [41]:

$$\chi^{(3)} = 4.02 \times 10^{15} (E_d/E_o)^4 \quad \text{esu.} \quad (3)$$

The nonlinear refractive index n_2 was correlated with $\chi^{(3)}$ through the equation ($n_2 = 12\pi \chi^{(3)}/n_o$). The investigated values of $\chi^{(3)}$ and n_2 for the as-prepared and irradiated films are listed in table 1 and they are increased with the increase of γ -irradiation doses. It is well known that, the $n_2 \propto E_{og}^4$ [42]. The tabulated values are consistent with the given relations. The obtained values of the n_2 are larger which indicate that, present films can be recognized for applications in fast optical switching devices. Furthermore, the material with large n_2 value reconnoiter third-order electronic polarization may have shortly response time and compact fiber designation, that may growing their applications in highly speed signal communication.

3.2 Optical band gap and absorption coefficient

The film absorbance (χ_a) was correlated with the absorption coefficient (αa) by this relationship ($\chi_a = e^{-\alpha a.d}$). The χ_a values as a function of λ for the as-prepared and exposed films has been investigated using Connell and Lewis equation [43]. Fig. 5 represents the absorption index for the as-prepared and exposed films. As shown represented in this figure the increase of γ -irradiation doses shifts the absorption coefficient towards the higher wavelengths i.e red shift of the absorption edge. As well discussed previously, the αa values less than 10^{-4} cm^{-1} have an exponential decadence on the wavelength according to the expression $\alpha a = \alpha a_0 \exp(Ee/hv)$ where αa_0 is constant and Ee is a band tail parameter that measuring the localized state width.

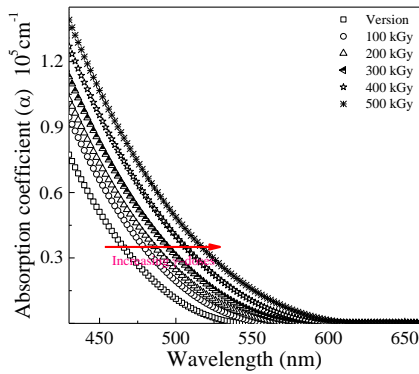


Fig. 5. The $\alpha(\lambda)$ for the as-prepared and exposed films.

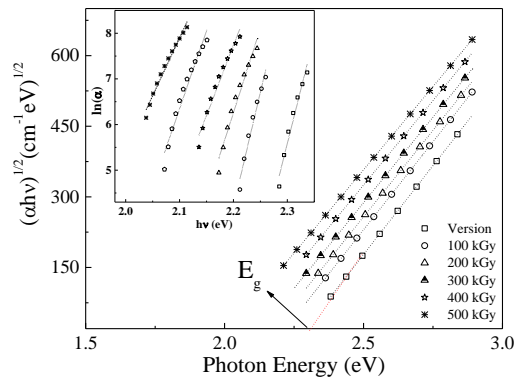


Fig. 6. The $\sqrt{\alpha a \cdot hv}$ vs. hv (high absorption region) and the inset of the figure shows the $\ln(\alpha a)$ vs hv (low absorption region) for the as-prepared and exposed films.

On the other side, for $\alpha a \geq 10^{-4} \text{ cm}^{-1}$ the absorption coefficient was found to obey the non-direct transition rule in the form of:

$$\sqrt{\alpha a \cdot hv} = \sqrt{B} (hv - E_{og}) \quad (4)$$

Whereas B is namely Tauc's edge slope and is inversely proportional to the width of localized states (Ee) [44] and E_{og} is the optical gap. Fig. 6 and it's inset shows the absorption coefficient indexes ($\ln(\alpha a)$, $\sqrt{\alpha a \cdot hv}$) versus the incident photon energy for the as-prepared and exposed

films. The observed E_e, E_{og}, \sqrt{B} values were tabulated in Table 1. As one can see from this table that, with increasing γ -irradiation doses the values of E_{og} and \sqrt{B} decrease while E_e values increases. The observed decrease in E_{og} may be due to the disorder changes and/or defects presented in non-crystalline materials. Taking in mind that, exposing the films to γ -irradiation produced defects results as disorder in the film structure. The \sqrt{B} measures the structure disorder in amorphous materials. The material with higher \sqrt{B} has lower structure disorder. In other words, Mott and Davies model decided that, the localized states width nearby the edge affected by the deficiencies and through the amorphous matrix i.e the E_{og} values are inversely proportional to E_e values. Finally, with the help of E_{og} value, the plasma frequency (ω_p) can be determined using Mikove et al. relationship [45]:

$$(n(0))^2 = 1 + (\hbar\omega_p/E_g)^2, \quad \omega_p^2 = 4\pi e^2(N/m^*) \quad (5)$$

The observed values of ω_p and the ration of the free carriers (N) to the effective mass (m^*) were listed in Table 1. Both the ω_p^2 and N/m^* behaves as well as the E_{og} which is consistent with the above relation.

4. Conclusions

The measured transmittance spectra of gamma irradiation (100–500 kGy doses) for $(AsTe)_5(GeSe_2)_{95}$ chalcogenide thin films were characterized to study the effect of γ -irradiation upon dispersion parameters and optical properties of the investigated sample. Remarkably, a clear difference in the measured film transmittivity ($T(\lambda)$) before and after exposed films of different doses of γ -irradiation. Any change of ($T(\lambda)$) affect the values of optical constants viz. absorption coefficient, refractive index and energy band gap. Such results show that, the $(AsTe)_5(GeSe_2)_{95}$ chalcogenide films can be serve as radiation dosimeter for radiation measurements.

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