

TRANSIENT PHOTOCONDUCTIVITY IN $\text{Se}_{85-x}\text{Te}_{15}\text{In}_x$ THIN FILMS

V. Sharma, A. Thakur, N. Goyal, G. S. S. Saini, S. K. Tripathi*

Centre of Advanced Study in Physics, Panjab University, Chandigarh- 160 014, India

The present paper reports the transient photoconductivity measurements on a- $\text{Se}_{85-x}\text{Te}_{15}\text{In}_x$ ($x = 0, 4, 10$ at. %) glassy alloys. Dark conductivity (σ_d) increases and activation energy (ΔE_d) decreases as the In concentration increases. The value of pre-exponential factor (σ_0) decreases with the increase of In concentration. Rise and decay of photocurrent (I_{ph}) measurements have been carried out as a function of temperature and light intensity in all thin film samples. A peak in the rise curve is observed in all films before attaining the steady state value. This behavior decreases as the In concentration increases. From the data of the rise curves of these plots, the values of dispersion parameter (α) have been calculated. Photosensitivity (σ_{ph}/σ_d) decreases on the incorporation of In additive. The values of decay rate constant (τ_d) have been calculated. The value of τ_d increases as the concentration of In increases. The value of charge carrier concentration (n_c) increases after the In incorporation into $\text{Se}_{85}\text{Te}_{15}$ thin films. The results have been explained in terms of recombination mechanism in these materials.

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1. Introduction

Transient photoconductivity is a very useful technique to determine the energy distribution of various species of gap states, which influence the carrier mobilities and life times in chalcogenide glasses assuming the response to be controlled by multi trapping processes [1]. This method is very valuable to judge the material quality for various photoconductive applications. Transient photoconductivity employs the method of illuminating the sample with light irradiation and measuring the rise and decay of photocurrent with respect to time as a function of temperature and intensity. The decay of I_{ph} follows many different patterns. In some cases, the decay of I_{ph} has been reported to be non-exponential [2], whereas a power law decay has been observed for many other semiconducting glasses [3].

Se-Te alloys have gained much importance because of their higher σ_{ph}/σ_d , greater hardness, higher crystallization temperature (T_C), and smaller ageing effects as compared to the pure Se glass [4]. The addition of third element into Se-Te alloys expands the glass forming region and also creates compositional and configurational disorder in the system [5]. The lattice perfection and the optical gap of the material play a major role in the preparation of the device which can be modified by the addition of dopant (In). Therefore, the photoconducting properties of a- $\text{Se}_{85-x}\text{Te}_{15}\text{In}_x$ alloys are interesting to study in detail not only from the basic but also from the application point of view.

Section 2 describes the experimental details of sample preparation and photoconductivity measurements. The results are presented and discussed in section 3. The last section deals with the conclusions drawn from the present work.

* Corresponding author: surya@pu.ac.in; surya_tr@yahoo.com

2. Experimental procedure

Glassy alloys of $\text{Se}_{85-x}\text{Te}_{15}\text{In}_x$ ($x = 0, 4$ and 10) are prepared by melt quenching technique. Materials (5N pure) weighed according to their atomic percentage are sealed in quartz ampoules in a vacuum $\sim 2 \times 10^{-5}$ mbar. The sealed ampoules are kept inside a furnace where the temperature is increased upto 1025 K a heating rate of 2-3 K/min. The ampoules are frequently rocked for 24 hours at the highest temperature to make the melt homogeneous. The quenching is done in L N_2 . Thin films of the alloys are prepared by vacuum evaporation technique keeping substrates at room temperature and base pressure of $\sim 2 \times 10^{-5}$ mbar using a molybdenum boat. Pre-deposited thick indium electrodes on well degassed corning 7059 glass substrates are used for the electrical contacts. A planar geometry of the film (length ~ 1.78 cm; electrode gap $\sim 8 \times 10^{-2}$ cm) is used for electrical measurements. The thickness of the film is about 5000 Å. The film is kept in the deposition chamber in dark for 24 hrs before mounting in the metallic sample holder to attain thermodynamic equilibrium as stressed by Tripathi et al. [6].

The photoconductivity of the amorphous films have been studied by mounting them in a specially designed metallic sample holder where heat filtered white light (200 W tungsten lamp) could be shone through a transparent quartz window. A vacuum of about 10^{-3} mbar is maintained throughout these measurements. The results have been found to be the same in higher vacuum $\sim 10^{-5}$ mbar also which have been verified in the laboratory. Light intensity is measured by a digital Luxmeter (Testron, model TES-1332). The I_{ph} is obtained after subtracting dark current (I_d) from the current measured in the presence of light. For measurement of transient photoconductivity, light is shone on the sample and rise & decay of I_{ph} is noted down manually from a digital picoammeter (DPM-111 Model). The accuracy in I_{ph} measurements is typically 1pA. The films are annealed at 332 K for 2 hrs in a vacuum of about 10^{-3} mbar and then the dark and photoconductivity measurements are carried out.

3. Results and discussion

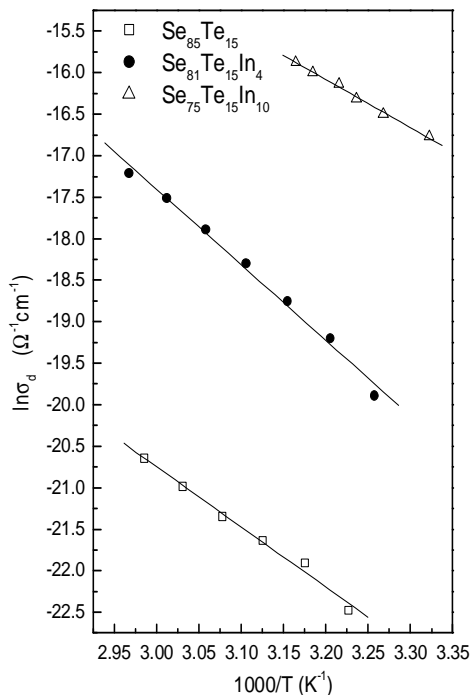


Fig. 1. Temperature dependence of σ_d for $\text{Se}_{85-x}\text{Te}_{15}\text{In}_x$ thin films.

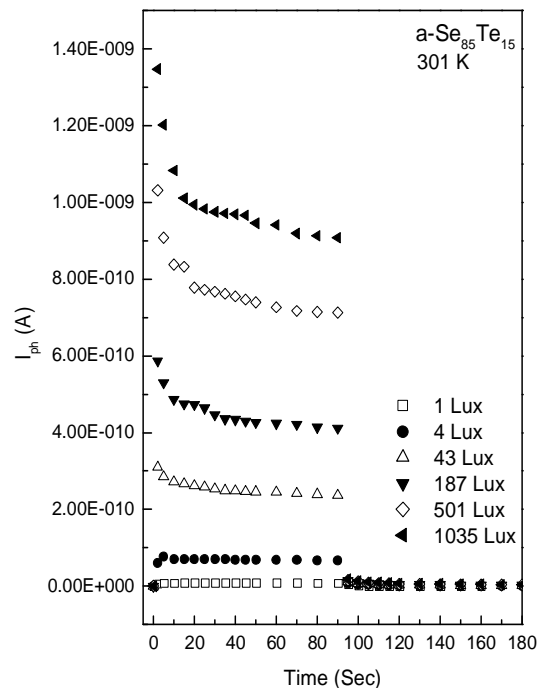


Fig. 2. Rise & decay of I_{ph} at various intensities of illumination for $\text{Se}_{85}\text{Te}_{15}$ thin film.

Fig. 1 shows the temperature dependence of σ_d in thin films of a- $\text{Se}_{85-x}\text{Te}_{15}\text{In}_x$ ($x = 0, 4$ and 10). Curves of $\ln\sigma_d$ vs $1000/T$ are straight lines for the thin films of all the three glassy alloys in the measured temperature range (301-337 K). This indicates that σ_d is a thermally activated process having single activation energy for each of the alloy. The dc conductivity obeys an Arrhenius behaviour given by the usual relation:

$$\sigma_d = \sigma_o \exp\left(\frac{-\Delta E_d}{k_B T}\right) \quad (1)$$

where σ_o is the pre-exponential factor, ΔE_d the activation energy for dc conduction and k_B is the Boltzmann constant. The values of ΔE_d are calculated for all the three alloys using the slope of the curves of Fig. 1 and are inserted in Table 1, which also contains the values of σ_d at 301 K for all thin film samples. The value of ΔE_d decreases from (0.93 ± 0.01) eV to (0.50 ± 0.01) eV and σ_d increases from $(5.22 \pm 1.00) \times 10^{-12} \Omega^{-1}\text{cm}^{-1}$ to $(5.19 \pm 0.01) \times 10^{-8} \Omega^{-1}\text{cm}^{-1}$ after adding In additive upto 10 at. % of In.

Table1. Variation of different parameters with the change of In additive concentration in the thin films of a- $\text{Se}_{85-x}\text{Te}_{15}\text{In}_x$ glassy alloys.

x	σ_d ($\Omega^{-1}\text{cm}^{-1}$)	σ_{ph} ($\Omega^{-1}\text{cm}^{-1}$)	ΔE_d (eV)	σ_{ph}/σ_d	α	n_σ (cm^{-3})
0	$(5.22 \pm 1.00) \times 10^{-12}$	$(5.66 \pm 0.01) \times 10^{-9}$	0.93 ± 0.01	1084	0.75	8.53×10^9
4	$(1.27 \pm 0.13) \times 10^{-9}$	$(9.36 \pm 0.01) \times 10^{-8}$	0.78 ± 0.01	44	0.87	2.07×10^{12}
10	$(5.19 \pm 0.01) \times 10^{-8}$	$(7.43 \pm 0.01) \times 10^{-7}$	0.50 ± 0.01	14	0.93	8.48×10^{13}

Figs. 2, 3 and 4 show the rise and decay of I_{ph} at various intensities of illumination for the three glassy alloy thin films respectively. The figures clearly indicate that a maximum is observed at higher intensities of illumination in case of all the thin films. This maximum is not observed at lower intensities. The maximum in the rise curve is not a common feature in case of chalcogenide glasses and can be attributed to non-equilibrium recombination, where bimolecular recombination predominates over the monomolecular mode of recombination on illumination in the high intensity region. This behaviour in the rise curve has also been observed by many other workers [7, 8]. The peak in the rise curve has been explained in terms of the non-equilibrium recombination mechanism as suggested by Andriesh *et al.* [9]. According to them, such a maximum occurs in the high intensity region ($I_{ph} \gg I_d$), where the recombination is predominantly bimolecular in nature. At lower intensities ($I_{ph} \ll I_d$), the nature of recombination is mainly monomolecular and this maximum in the rise curve may be absent with the photocurrent increasing monotonically to the steady state during illumination. Mann *et al.* [10] have also found a similar maximum in a- $\text{In}_{20}\text{Se}_{80}$ and the rise of photocurrent during illumination has been attributed to the recombination between holes in valence band with electrons at the recombination centres. After adding In to the binary alloy $\text{Se}_{85}\text{Te}_{15}$, the decay of photocurrent becomes slower as compared to pure $\text{Se}_{85}\text{Te}_{15}$ thin films.

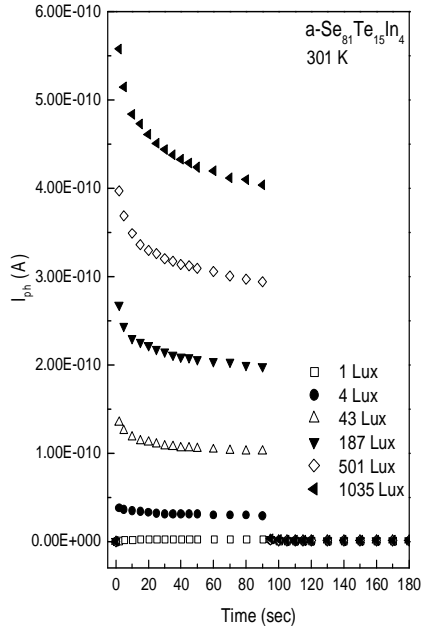


Fig. 3. Rise & decay of I_{ph} at various intensities of illumination for $Se_{81}Te_{15}In_4$ thin film.

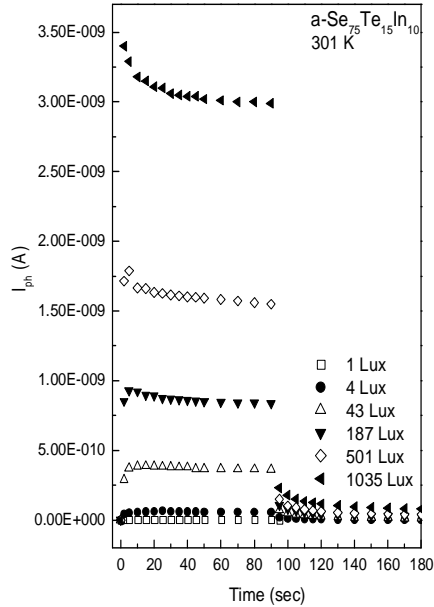


Fig. 4. Rise & decay of I_{ph} at six intensities of illumination for $Se_{75}Te_{15}In_{10}$ thin film.

At lower levels of illumination [1-4 lux], the maximum in the rise curve of the photocurrent is not observed. At lower intensities (1-4 lux), the photocurrent rises monotonically to the steady state value. However, at higher intensities, the nature of the rise curves is quite different. The photocurrent passes through a maximum value $I_{ph}(max)$ before attaining the steady state value $I_{ph}(st)$.

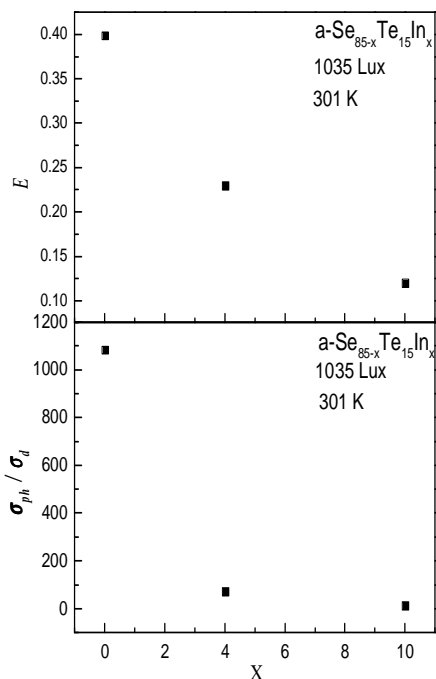


Fig. 5. Variation of E & σ_{ph}/σ_d with the concentration of In additive (x) at room temperature.

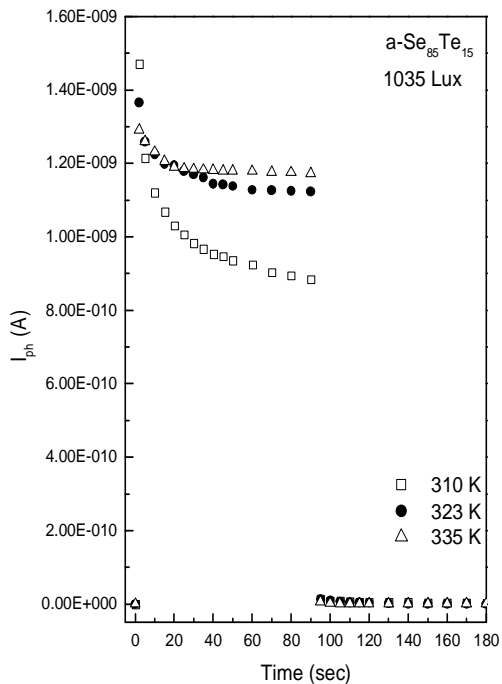


Fig. 6. Rise & decay of I_{ph} with time at different temperatures for thin film of $Se_{85}Te_{15}$.

To assess the relative variation of the anomalous effect at various intensities and temperatures, a quantity E has been used, where

$$E = \frac{[I_{ph}(\text{max}) - I_{ph}(st)]}{I_{ph}(\text{max})} \quad (2)$$

Using the data of the rise of I_{ph} for different intensities from Figs. 2, 3 and 4, the values of E have been calculated for the thin films added with different concentration of In additive and plotted in Fig. 5. The value of E decreases with the increase in In concentration in the binary alloy. The same figure also contains the values of σ_{ph}/σ_d with In additive concentration, which also decrease with increase in In content. Therefore, the maximum is observed at higher intensities and maximum decreases with increase in In content due to decrease in σ_{ph}/σ_d . The nature of variation of E and σ_{ph}/σ_d with In concentration is the same. Fig. 6 shows the rise and decay of I_{ph} at different temperatures for a- $\text{Se}_{85}\text{Te}_{15}$ thin film. The maximum in the rise curve decreases with increase in temperature. The rise and decay curves at various temperatures for Se-Te binary alloy added with different concentration of In also show a similar behaviour. The maximum in the rise curve during illumination of the thin films is more prominent at lower temperatures as compared to at higher temperatures

The value of E is observed to increase with intensity at all concentrations of In addition as indicated by Fig. 7. The same figure also contains the values of σ_{ph}/σ_d which also increases with increase in intensity justifying the observation of maximum in the rise curve only at higher intensities. Fig. 8 shows the temperature variation of σ_{ph}/σ_d and E for all concentrations of In addition, both of which decrease with increase in temperature. This indicates that the maximum in the rise curve during illumination decreases with increase in temperature due to decrease in σ_{ph}/σ_d . This shows that the nature of variation of E and σ_{ph}/σ_d with temperature is also similar. This clearly implies that the maximum in the rise curve increases as the photosensitivity increases on increase of intensity or decrease of temperature. At very low intensities, this effect is not seen at all.

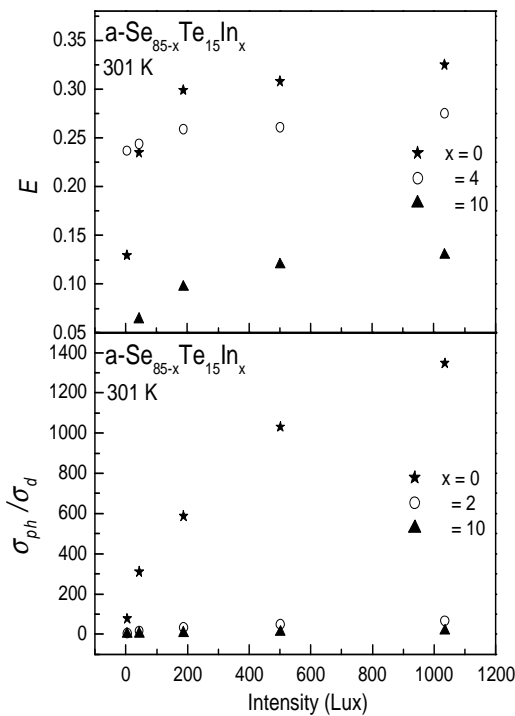


Fig. 7. Variation of E & σ_{ph}/σ_d with intensity for $\text{Se}_{85-x}\text{Te}_{15}\text{In}_x$ thin films.

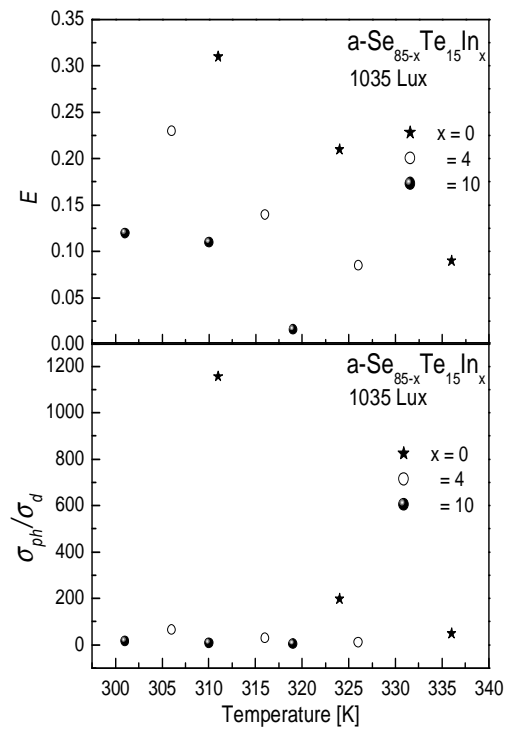


Fig. 8. Temperature variation of E & σ_{ph}/σ_d for $\text{Se}_{85-x}\text{Te}_{15}\text{In}_x$ thin films.

Andriesh *et al.* [9] observed a maximum in the rise curve of photocurrent and explained their results in terms of non-equilibrium recombination. In photoconductivity measurements, the time dependence of delocalised carrier density $p_c(t)$ is very sensitive to the level of generation rate 'g'. Under high generation rate $g > g^*$, the recombination is completely governed by bimolecular recombination mechanism.

The generation rate

$$g^* = \left(\frac{1}{2}\right) \left(\frac{N_c}{N_t}\right) \left(\frac{\tau_0}{\tau_R}\right)^{\frac{1}{\alpha}} \left(\frac{1}{R\tau_0\tau_R}\right) \quad (3)$$

In this case, for $0 \leq t < t^*$ (i.e. for the initial interval), where

$$t^* = \tau_0 \left(\frac{N_t}{N_c}\right)^{\frac{1}{1+\alpha}} \left[gR\tau_0^2\right]^{\frac{-1}{1+\alpha}} \quad (4)$$

the photocurrent rises by power law

$$p_c(t) = g\tau_0 \left(\frac{N_c}{N_t}\right)^{\alpha} \left(\frac{t}{\tau_0}\right)^{\alpha} \quad (5)$$

At $t = t^*$ i.e. it passes through maximum

$$p_c^* = p_c(t^*) = g\tau_0 \left(\frac{N_c}{N_t}\right)^{\frac{\alpha}{1+\alpha}} \left(gR\tau_0^2\right)^{\frac{-\alpha}{1+\alpha}} \quad (6)$$

For the interval $t^* < t < t_{st}$, where

$$t_{st} = \tau_0 \left(\frac{N_t}{N_c}\right)^{\frac{\alpha}{1+\alpha}} \left(\frac{RN_t^2}{g}\right)^{\frac{1}{1+\alpha}} \quad (7)$$

the photocurrent decays by power law

$$p_c(t) = \left(\frac{g}{R}\right)^{\frac{1}{2}} \left(\frac{N_c}{N_t}\right)^{\frac{\alpha}{2}} \left(\frac{t}{\tau_0}\right)^{\frac{-(1-\alpha)}{2}} \quad (8)$$

down to steady state value

$$p_c(st) = N_c \left(\frac{g}{RN_c N_t}\right)^{\frac{1}{1+\alpha}} \quad (9)$$

At very low generation rates $g < g_0$, where

$$g_0 = 2 \frac{-1}{\alpha} \left(\frac{N_c}{\tau_R}\right) \left(RN_t\tau_R\right)^{\frac{-1}{\alpha}} \quad (10)$$

the recombination is governed by monomolecular recombination mechanism.

For $0 \leq t < t_m$ (i.e. for initial interval), the photocurrent increases by power law eqn. (5), where

$$t_m = \tau_0 \left(\frac{N_t}{N_c}\right) \left(\frac{\tau_R}{\tau_0}\right)^{\frac{1}{\alpha}} \quad (11)$$

At $t = t_m$, the current reaches the steady state value i.e.

$$p_c(m) = g\tau_R \quad (12)$$

In this case the maximum in the photocurrent is absent.

For intermediate generation rate $g_0 < g < g^*$, in the interval $0 < t < t_b$, the monomolecular recombination dominates, and for $t > t_b$, the recombination is governed by bimolecular mechanism where

$$t_b = 2^{\frac{-1}{(1-\alpha)}} \tau_0 \left(\frac{N_c}{N_t} \right)^{\frac{\alpha}{(1+\alpha)}} (gR\tau_R^2)^{\frac{-1}{(1-\alpha)}} \quad (13)$$

Symbols:

$p \rightarrow$ total carrier density; $p_c \rightarrow$ delocalised carrier density; $g \rightarrow$ generation rate; $\tau_R \rightarrow$ characteristic time of monomolecular carrier recombination; $R \rightarrow$ bimolecular recombination constant; $\tau_0 \rightarrow$ life time of a delocalised carrier respective to trapping; $N_t \rightarrow$ total density of localised states; $N_c \rightarrow$ density of conductive states; $T \rightarrow$ Temperature (K); $k \rightarrow$ Boltzmann constant.

There is also a possibility of light induced defects responsible for such behaviour. Iovu *et al.* [11] have also found a similar maximum in Sn doped a- As_2Se_3 and AsSe films. According to them, at short times $t < t_R$ (time at which transition from recombination free to monomolecular recombination controlled I_{ph} occurs), recombination is not yet significant and the rise of I_{ph} is fully controlled by carrier trapping, yielding

$$I_{ph} \approx gt^\alpha \quad (14)$$

where g is the generation rate and α is the dispersion parameter. At longer times, I_{ph} rise depends on the excitation intensity.

At low intensities, recombination is always dominated by monomolecular recombination, the I_{ph} increases monotonically and saturates at a steady state value. At higher intensities, a quasi-stationary portion of the photocurrent is obtained after saturation, followed by a decreasing portion of the transient curve, which is governed by the equation

$$I_{ph} \approx g^2 t^{\frac{-1-\alpha}{2}} \quad (15)$$

Here the bimolecular recombination is dominant over monomolecular recombination. At even higher excitation levels, the rise is entirely controlled by the bimolecular recombination and the quasi-stationary portion of the photocurrent exhibits an overshoot.

According to Kastner *et al.* [12] and Arkhipov *et al.* [13], this overshoot in I_{ph} transient, in terms of the multiple trapping model, is due to the non-equilibrium bimolecular recombination whose intensity increases with the total carrier density. If this recombination mode is dominant, the rate of recombination sooner or later gets high enough and together with carrier trapping by states below the demarcation level, provides conditions for decreasing the density of free carriers. The maximum in the rise curve of photocurrent changes to a steady increase of I_{ph} in the crystallized states.

In the present case also, the peak in the rise of photocurrent has been observed at higher intensities only. At much lower intensities, this peak has not been observed. The results are in accordance with the theory proposed by Andriesh *et al.* [9]. Based upon the above theory, the values of dispersion parameter ' α ' have been calculated from double logarithmic plots using the data of Figs. 2, 3 and 4. Fitting these data, the dispersion parameter α , is calculated and values for all the three thin films at room temperature and an intensity of 1035 lux are given in Table 1. The value of α increases with the increase in In concentration in a- $\text{Se}_{85}\text{Te}_{15}$ binary alloy, indicating a lower value of maximum in the rise of I_{ph} . The value of α is less than one in case of all the three glassy alloys and is in good agreement with those reported by others [7,11]. The value of α has been observed to change by additives which describes the exponential density of localised states as observed by Pfister and Morgan [14]. They observed that the time dependent current is then result only of a time dependent average mobility and found the transit time to decrease with the addition of In additive.

The study of peak in the rise curve during illumination at different temperatures shows that the peak in rise is maximum for maximum value of σ_{ph}/σ_d at a particular temperature (Figs. 2, 3 and

4). As the temperature range of the present measurements has been close to the glass transition temperature ($T_g = 336$ K) of the a- $\text{Se}_{85}\text{Te}_{15}$ alloy [12], another explanation of the rise of I_{ph} can be given in terms of the structural relaxation processes which may affect the physical parameters near the T_g . The structural relaxation rate depends on the annealing temperature and also on the light exposure [13]. It can be argued that, near the T_g , the relaxation rate in the presence of light becomes so large that the defects start equilibrating on the time scale of present measurements. Thus, the observed maximum in the rise of I_{ph} may be a consequence of light and temperature induced structural relaxation effects.

The above discussion shows that the peak in the rise of I_{ph} in present case can be examined equally well in terms of non-equilibrium recombination at higher intensities as well as structural relaxation in the presence of light near the glass transition temperature. Measurements at temperature much lower than T_g can help in ruling out the possibilities of structural relaxation effects. It has not been possible to go below 301 K in the present experiment because of the high resistance of the films.

In the present case, the value of σ_0 decreases (from $\sim 1.54 \times 10^4 \Omega^{-1}\text{cm}^{-1}$ to $11.55 \Omega^{-1}\text{cm}^{-1}$ for $\text{Se}_{85}\text{Te}_{15}$ to $\text{Se}_{75}\text{Te}_{15}\text{In}_{10}$ respectively) with the increase in In concentration. A decrease in the value of σ_0 indicates that the density of localised states increase on the addition of In to $\text{Se}_{85}\text{Te}_{15}$ binary alloy [15]. A decrease in the value of σ_{ph}/σ_d (from 1084 to 14) after In incorporation, also indicates that there is an increase in the density of localised states. The charge carrier concentration (n_σ) for different samples has been calculated using the equation [4,16]

$$n_\sigma = 2 \left(\frac{2\pi m k_B T}{h^2} \right)^{\frac{3}{2}} \exp \left(\frac{-\Delta E_\sigma}{k_B T} \right) \quad (16)$$

where m is the mass of the charge carrier, k_B is the Boltzmann constant and ΔE_σ the Fermi energy level position of the glassy semiconductor.

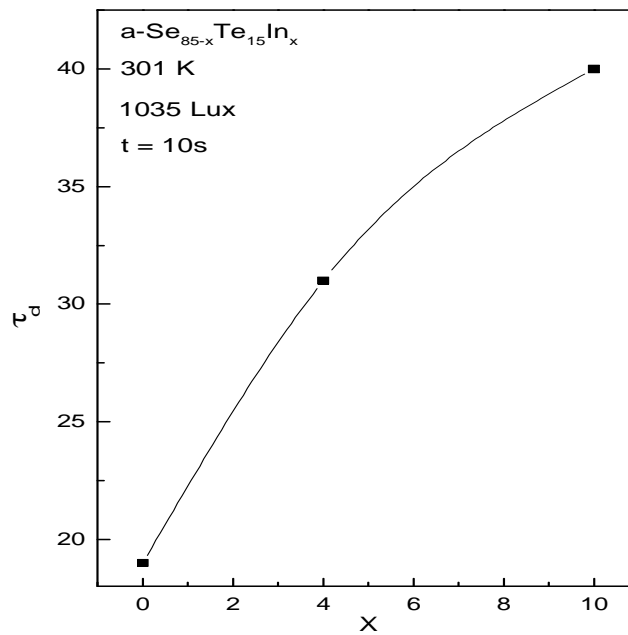


Fig. 9. Variation of τ_d with In concentration (x) for $\text{Se}_{85-x}\text{Te}_{15}\text{In}_x$ thin films.

But the much better approach would be to presuppose the fixed pre-exponential factor (σ_0), to calculate the ΔE_σ and then the n_σ , the procedure usually taken for other amorphous semiconductors [17]. Using this way, the charge carrier concentrations have been calculated for

these $\text{a-Se}_{85-x}\text{Te}_{15}\text{In}_x$ thin films. For the calculation of the values of n_σ , the value of $\sigma_0 \approx 1.54 \times 10^4 \Omega^{-1}\text{cm}^{-1}$ of the $\text{a-Se}_{85}\text{Te}_{15}$ film has been taken. The calculated values of n_σ are given in Table 1. The value of n_σ increases with the increase of In concentration indicating an increase in the density of defect states on the incorporation of In additive.

This observation is also supported by the transient photoconductivity measurements where the value of differential life time constant (τ_d) has been observed to increase with the increase in In concentration. The differential life time (τ_d) has been calculated by using the expression as suggested by Fuhs and Stuke [3]

$$\tau_d = - \left[\frac{1}{I_{ph}} \cdot \frac{dI_{ph}}{dt} \right]^{-1} \quad (17)$$

From the slopes of these curves (decay of I_{ph} vs. time curves of Figs. 2, 3 and 4 for maximum intensity of illumination at $T = 301$ K), the values of τ_d have been calculated using the above equation at time $t = 10$ sec. Corrected values of I_{ph} have been used for these calculations. The results have been plotted in Fig. 9. The value of τ_d increases with the increase in In concentration. This indicates a slower rate of decay of I_{ph} , on switching off the illumination, with the incorporation of In additive in binary $\text{Se}_{85}\text{Te}_{15}$ binary alloy.

The σ_{ph}/σ_d depends on the life time of the excess charge carriers which, in turn depends on the density of defect states in a particular material. Higher the density of defect states, lower will be the life time as these defect states may act as recombination centres in the presence of light forming intimate valence alternation pairs (IVAPs) under illumination. These defects are converted by bond switching reactions to random pairs of charged defects, known as light induce metastable defects (LIMDs) [18-20]. The LIMDs act as electron/hole trapping centers and decrease the photocurrent. In other words, the more the density of defect states, the more will be IVAPs. Therefore, σ_{ph}/σ_d should decrease with the increase in the density of defect states. Such an increase in the density of defect states after incorporation of In additive may be understood in terms of the value of electron affinity of In as suggested by Onozuka *et al.* [21].

The Se and Te are elements of the same group (group VI) of the periodic table. Therefore, most tellurium atoms are probably mixed in selenium atom chains but, some of the tellurium atoms are not incorporated in chains. The Te atoms not incorporated into chains would act as ionized impurities forming positively charged localised states, since the electron affinity of Te (1.97 eV) is lower than that of Se (2.02 eV). Similarly, the addition of In will induce more and more positively charged localised states, since the electron affinity of In is much lower than that of Se and Te. Therefore, an increase in defect states after putting In concentration in binary $\text{Se}_{85}\text{Te}_{15}$ alloy may be understood in terms of very low electron affinity of In (0.30 eV) as compared to Se or Te. Khan *et al.* [22] have also studied the effect of In incorporation on the electrical conductivity and thermoelectric power measurements on $\text{a-Se}_{80-x}\text{In}_x$ and $\text{Se}_{80-x}\text{Ge}_{20}\text{In}_x$ thin films. They have shown that the number of charge carriers calculated using equation (16) increase with the increase in In concentration.

4. Conclusions

The effect of In impurity on the transient photoconductivity has been studied in amorphous $\text{Se}_{85}\text{Te}_{15}$ binary alloy. The maximum in the rise curve during illumination decreases with the incorporation of In additive. This peak in the rise curve increases with increase in intensity and decreases with increase in temperature. The decay of I_{ph} on switching off the illumination becomes slow in the thin films added with In. This indicates an increase in the density of localised states in the mobility gap of the Se-Te binary alloy on the addition of In. Both σ_d and σ_{ph} increase after the addition of In. The ΔE_d decreases as the In concentration is increased. The σ_{ph}/σ_d decreases sharply after In incorporation.

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