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# Doping Effect of Sn in a-Se based Chalcogenide Glasses

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**Abstract:** ***In order to perform well in a variety of applications, materials must be affordable, non-toxic, environmentally friendly, simple to synthesize and well competitive. The adaptability of these materials to several functions lays the groundwork for multidisciplinary research. Materials made of chalcogenides have also demonstrated this capability and could represent the solution to this problem in the future. Tin-based compound semiconductors have attracted enormous interest in photovoltaic and optoelectronic applications due to favorable band gap (1.0–1.3 eV) and higher absorption coefficient ($10^5$ cm$^{-1}$). Doping of amorphous chalcogenide films by tin impurities assists in stabilizing the glassy matrix with respect to light exposure and thermal treatment. This review summarizes some Sn doped glass-forming systems and also the role of Sn metallic additive on structural modification, electrical and optical properties of Se rich binary, ternary and quaternary chalcogenide glasses.***

**Keywords:** Density of defect states (DOS), Optical band gap, Dark conductivity, Photosensitivity

## 1. Introduction

Chalcogenide glasses (ChGs) composed of sulfur (S), selenium (Se), tellurium (Te) and their compounds as the main components are representative of non-oxide glasses. ChGs are attracting a rigorous research essentially due to their excellent chemical stability in aggressive environments, transparency in the near and far-infrared region, low optical losses, high refraction coefficient and relatively high ion conductivity. The distinctive properties viz. lesser phonon energy, larger and wider band gap, high values of linear as well as non-linear refractive index, extraordinary transmittance range, etc. of these glasses find them a wide usage in photo-detectors, LED, IR sensors, holography and waveguides. Owing to high refractive index values and high non-linearity (~$10^2$ times of silica), chalcogenides is utilized as an ultrafast switch. Due to high chemical and thermal stability, a chalcogenide material eases fabrication of optical devices. Metallic doping in chalcogenides alters the average coordination number as well as produce structural changes viz. flexible ↔ intermediate ↔ rigid, in the glassy matrix. In chalcogenide glasses, almost all the properties are compositional dependent. Therefore the physical, optical, electrical or structural properties can be altered and controlled depending upon utilization requirement.

ChGs, having an energy gap of 3 eV, show semiconducting behavior [1]. For analyzing band structure in amorphous semiconductors, the chemical bonding environment is given more importance than the periodicity [2]. The molecular bonding in ChGs is covalent in nature and the components are kept together with attractive vanderwaals forces. In ChGs, the lone pairs enhance the bending of the bond angles and hence reduce the strain energy of the system [3]. Hence the atoms form amorphous structures readily when doped with other elements. Elemental or binary glassy systems exhibit one or two dimensional chain-like or layered structures and thus possess short-range order over a larger extent. In contrast ternary, quaternary and higher multi-component glassy systems exhibit a rigid three-dimensional structure and thus possess short-range order over a shorter extent [4].

Photoconductivity is an intrinsic property of the ChGs. It leads to a variety of interesting effects, which as a group can be labeled photo-induced effect and make structural changes. The changes in structure are caused by the absorption of above or below band gap light and the subsequent relaxation of the excited state into a new metastable structure. The ability to control the structure and properties of the ChGs with light makes them uniquely suitable for applications in optics and microfabrication. The addition of third element (Sn) expands the glass forming area and also creates compositional and configurational disorder in the system. The lattice perfection and the energy band gap of the material play a major role in the preparation of the device for a particular wavelength, which can be modified by the addition of dopants. So, the influence of additives on the electrical properties has been an important issue in the case of ChGs. As part of Group IV elements, adding tin (Sn) to the chalcogenide system complements the configuration by acting as a donor and considerably modifies the system's characteristics [5]. Such as the Sn as an impurity enhanced optical and electronic properties by permitting the absorption edge shift with the change in refractive index, optical conductivity, dielectric constant and greatly varies the optical band gap, thereby facilitating the possibilities of optoelectronic applications [6][7].

Thin films of Sn chalcogenides may find use in photovoltaic devices and nanocrystals (NCs) of such materials are attractive due to their tuneable band gaps and potential in photovoltaic, photonic and optoelectronic applications. Sn chalcogenide materials of the type SnE where E = S, Se or Te have semiconducting properties and these materials are observed to have intense absorption across the electromagnetic (EM) spectrum and narrow band gaps. They, therefore, have potential as materials for optoelectronic and photovoltaic applications. Sn is a far less toxic and, indeed, a relatively earth-abundant metal compared with many of other elements used in semiconductor materials. There has been great interest in the controlled deposition of thin films of Sn chalcogenides and the synthesis of tin chalcogenide NCs of controlled size and shape with tuneable band gaps. The doping effect of Sn on the characteristics of some a-Se based chalcogenide glassy systems were discussed in this review article.

**Sn doped Se- based chalcogenide glasses**

The doping of metals could be used to enhance and tune the host matrix's physical and chemical properties. Sn, the most likely transition metal to produce tetrahedrally coordinated sites, alters the network of the host glass by bringing about alterations in both short- and medium-range order. Tin as a dopant serves as a model probe atom to permit local structural characterization of the impurity in ChGs.

**SnSe**

Among Sn chalcogenides, tin selenide (SnSe) has demonstrated great potential in the applied material science. SnSe is a semiconducting metal chalcogenide which currently attracts major interest for photovoltaic applications. SnSe is a useful material for energy conversion devices, in particular displaying a variety of properties potentially expedient to materials in photo-electrochemical (PEC) solar cells including suppression of photocorrosion and enhancement of fill factor. Tin selenide based SnSe/$SnSe_2$ are binary compound semiconductor materials having p/n-type conductivity [8][9].

The SnSe exists in two crystallographic phases, i.e., orthorhombic (α-SnSe) and cubic (π-SnSe) and $SnSe_2$ exists in the hexagonal crystal structure. The π-SnSe material is mechanically stable and energetically comparable to α-SnSe and has already shown potential in the piezoelectric application [10]. Both the tin-selenide phases show the indirect and direct band gaps. The direct band gap of α-SnSe shows a wide tunable band gap which varies from 0.98 eV (bulk) to 1.43 eV (monolayer) [11]. $SnSe_2$ shows large variation in the band gap from 1.84 eV (bulk) to 2.04 eV (monolayer) [12]. This band gap tunability of SnSe shows its immense application possibilities in optoelectronic device applications. Tin selenide exists in two stoichiometric phases, i.e., SnSe and $SnSe_2$.Various research groups have reported another phase, $Sn_2Se_3$, but this phase is the superposition of the SnSe and $SnSe_2$ as confirmed by Nuclear Magnetic Resonance (NMR) spectroscopy. Defects present in the material also influence the properties of materials like electronic, magnetic, and optical properties. SnSe is generally a p-type semiconductor. Density functional theory (DFT) calculation was performed to study the defects during SnSe crystal growth. The Sn vacancy is present as a native defect, which causes the p-type conduction in the SnSe under Se or Sn rich conditions, as confirmed by Scanning Tunneling Microscopy (STM) studies [13][14]. $SnSe_2$ shows n-type conduction. DFT calculation shows that the vacancy of selenium and interstitial tin led to this n-type conduction in $SnSe_2$ [14]. SnSe has demonstrated versatility in thermoelectric, photodetector [15], solar cells, photocatalytic, phase change memory [16], gas sensing [17], anode material for battery, supercapacitor [18] and topological insulator (TI) [19]. These applications strongly depend upon the properties of SnSe (optical, electrical and microstructural, etc.).

Apart from material properties, material synthesis/deposition methods also play an essential role in obtaining high-quality materials [20][21][22]. John et al [23] used a 'reactive evaporation' approach to deposit SnSe thin films with elemental Sn and Se evaporated onto a common target kept at elevated temperatures (473 – 600 K). p-XRD was used to characterize the films, revealing the (400) plane as the preferred orientation. Optical measurements revealed an allowed direct band gap of 1.2 eV. Zainal and co-authors [24] used alkaline media to form SnSe films on indium tin oxide (ITO) using a chemical bath deposition approach, where sodium selenosulfate was decomposed in aqueous sodium hydroxide solution at pH 11.4 in the presence of tin(II) chloride. SnSe films were found to be polycrystalline with preferred orientation in the (201) plane. Optical absorbance measurements revealed a band gap of around 1.25 eV with a p-type direct transition.

Kumar et al [25] reported the synthesis of SnSe thin films from SnSe pulverised powder using thermal evaporation onto a glass substrate at 350 K. The films were found to be polycrystalline and orthorhombic with preferred orientation in the (111) plane. Crystalline size, strain and dislocation density were calculated from the p-XRD pattern. SEM results of the studies shows that the SnSe films exhibited uniformly distributed grains over the entire surface of the substrate. The average sizes of the grains were reported to be 16 nm. Optical measurements revealed a direct band gap of 1.9 eV. A shallow donor level near conduction band, around 0.3 eV, was confirmed through calculation of activation energies from temperature dependent resistivity measurements.

SnSe thin films prepared through facile and cost-effective chemical drop-casting method was reported by Raval et al [26]. The synthesized film showed the presence of nanocrystallinity with orthorhombic crystal structure of SnSe. The as-grown thin films were found to be p-type having 1.4 eV bandgap and electrical conductivity of the order of 10 S $cm^{-1}$ which made them suitable to use it in the solar cell as an absorber layer. Sharma et al [27] reported the d.c. conductivity measurements at high electric fields in vacuum evaporated amorphous thin films of a-$Se_{100-x}Sn_x$ (x=0, 2, 4, 6) glassy alloys. I-V characteristics measured at various fixed temperatures shows ohmic behaviour at low electric fields and non-ohmic behaviour at high electric fields ($E \sim 10^4$ V/cm); analysis of the data confirms the presence of space charge limited conduction (SCLC) in the studied glassy materials. The density of defect states (DOS) near Fermi level was calculated and found to increase with increase in Sn concentration. The results were correlated with the electronegativity difference between Se and Sn used in the preparation of the glassy alloys. Sn having lower electronegativity than Se, when introduced, created positively charged defects which increases the DOS in the binary Se-Sn system beyond pure Se. Similar measurements in a-Se and a-$Se_{90}Sn_{10}$ thin films reveals that DOS increased by nearly a order of magnitude when Sn having 10 at.% is introduced in a-Se [28].

Panchal et al [29] deposited SnSe thin films from the pulverized compound material by thermal evaporation method in order to study the effect of film thickness on its structural and optical properties. SnSe thin films were grown on glass substrates held at room temperature with varying the film thicknesses from 150 nm to 500 nm. The structural analysis suggests that the thin films were polycrystalline in nature having preferred orientation in (111) direction. The surface roughness was apparently changed with increase in film thickness. The direct energy band gap calculated from the optical transmission spectra were in the range 1.74 to 1.24 eV; the energy band gap decreases with increasing the film thickness which shows its capability to be used as an absorber layer in photovoltaic application. In general, the DOS in the film increases with the film thickness which leads to decrease in the band gap. The electrical resistivity shows that the films were semi-conducting in behaviour having p-type conductivity. The narrow optical band gap (~1.3 eV) having p-type conductivity indicates its use in photovoltaic applications.

### SnCdSe

The study on the incorporation of Sn in thin layers of nanocrystalline CdSe by the chemical bath deposition technique was performed by P. K. Sahu and his co-authors [30]. X-ray diffraction pattern indicated that the formed structure is cubic with most prominent peak at (111). It has been observed that the crystallinity behaviour of the films is enhanced by doping Sn due to an increase in the grain size. Sn incorporation in CdSe decreases the optical bandgap from 2.62 eV (undoped CdSe) to 2.50 eV (5% Sn:CdSe) respectively. This increase in grain size with Sn doping significantly decreases the electrical resistivity.

### SnAsSe

Doping of a high concentration of Sn in the As–Se system causes an increase in the growth of the tetrahedral unit of structure $Sn(Se_{1/2})_4$ [31].Tin as an impurity significantly influences the absorption edge, causing the exponential slope to decrease, widening the spectral range of photoconductivity and delaying the recombination of the photogenerated holes. The AsSe amorphous films usually became darkened under action of light from the region of fundamental optical absorption $h\nu \geq Eg$ and so called photodarkening effect takes place. The increasing of the absorption is caused by the red shift of the absorption edge, due to broadening of the valence band, is accompanied of the respective increasing of the refractive index. Metal impurities effectively reduce the photodarkening and the degree of reduction becomes stronger as the impurity concentration is increased. The optical study under light exposure in amorphous films $As_2Se_3$ doped with Sn (0.5 to 5.0 at% Sn) was studied by M. S. Iovu and his co- authors [32]. It was shown that it effectively reduces photodarkening and this degree of reduction becomes stronger up to 2.0% concentration, then photo darkening increases again. The doping of amorphous chalcogenide films by Sn serves as a stabilizer for stabilizing the glass matrix in terms of light exposure and heat treatment.

### GeSeSn

The Sn doping also affects linear optical properties effectively. The linear optical properties of amorphous $GeSe_{2-x}Sn_x$ (where $0 \leq x \leq 0.8$) films were studied using spectrophotometric measurements of transmittance ($T$) and reflectance ($R$) at typical frequencies of light in the wavelength range of 200–2500 nm [33]. Linear optical properties showed that the optical band gap decreases and the refractive index increases with an increase in Sn content in the Ge–Se system. It was found that the third-order nonlinear optical susceptibility is a function of Sn in the Ge–Se system. The values of the third-order nonlinear optical susceptibility increased with an increase in Sn content. Figure 1(a) shows the transmission spectra of $GeSe_{2-x}Sn_x$ ($0 \leq x \leq 0.8$). At $\lambda > 850$ nm, all the films became transparent, indicating a lower diffusion and absorption; therefore, the transmission intensity is 80% higher than the intensity on the visible region. The absorption edge increased showed red-shift with Sn dopant concentration increase. The band gap value estimation for Sn doped GeSe using Tauc relation has been shown in Figure 1(b). The addition of Sn atoms at the place of Se atoms induces the formation of Ge–Sn and Sn–Sn bonds at the place of some Ge–Se bonds. This indicates an increase in the density of weaker bonds at the expanse of stronger bonds because of the higher atomic weight of a Sn atom than that of a Se atom. This consequently decreases the optical band gap energy of $GeSe_{2-x}Sn_x$ by increasing the Sn content. The effect of film composition on the spectral behaviour of the dispersion curves has been shown in Figure 1(c). The refractive index ($n$) for all films increases with an increase in λ up to 800 nm. After that, it decreases with the increase of wavelength. This increase in $n$ value with an increase in doping concentration could be ascribed towards an increase in the valence of the current carriers. This refers to the increase in polarizability associated with larger Sn atoms.

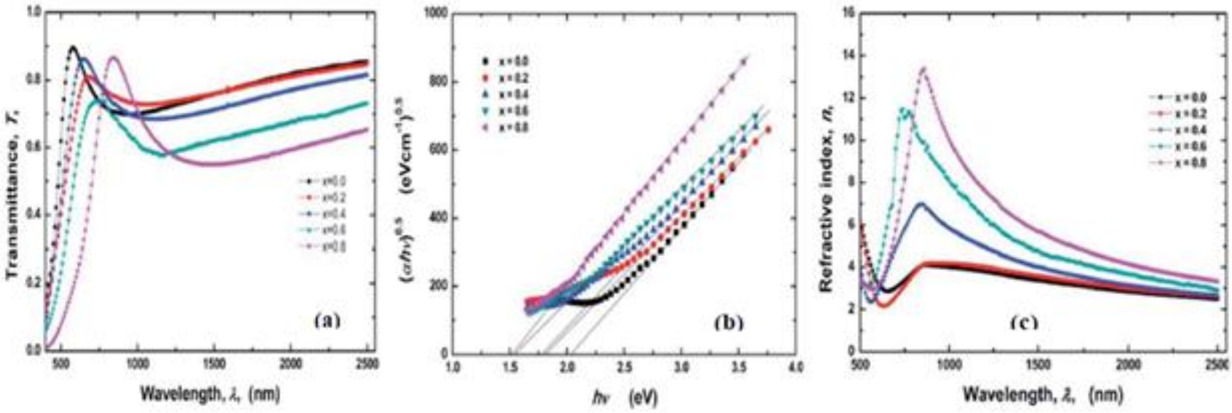

**Figure 1:** (a) Optical transmission (T) of $GeSe_{2-x}Sn_x$ films ($0 \leq x \leq 0.8$) as a function of wavelength of light λ, (b) $(\alpha h\nu)^{1/2}$ versus $h\nu$ for $GeSe_{2-x}Sn_x$ films, (c) variation of refractive index (n) against wavelength. [33]

The introduction of Sn in the $Ge_{1-x}Sn_xSe_3$ system results in the shifting of band-edge with an increase in x values as Sn gets substituted at the position of Ge, as reported by Fadel et al [34]. This indicates a decrease of band gap with an increase in Sn concentration. This behaviour is due to the intrinsic defect states contributed to the deposition of Sn. The shifting behaviour of the refractive index towards the lower energy values is attributed to the increase of Sn concentration in the $Ge_{1-x}Sn_xSe_3$ system.

**SnSbSe**

Structures of melt quench-synthesized samples of $Sn_xSb_5Se_{95-x}$ system, where x =0, 5, 10 and 12.5-mole% have been studied using Fourier transform infrared spectroscopy (FTIR) and Raman spectroscopy [35]. FTIR spectra (Figure 2a) show that addition of Sn-mole% to the system causes a shift in IR-peak's intensity and width from long to the short wavelength. This change implies the breaking of Se chains (Se-chain mode occurs in binary system) that appeared around 210–254 $cm^{-1}$ and the occurrence of pyramidal $SbSe_3$ around 147–210 $cm^{-1}$ (asymmetrical stretching of pyramidal $SbSe_3$ mode is dominant) and asymmetrical tetrahedral $SnSe_4$ mode (occurrence of new transmission bands) around 117–145 $cm^{-1}$ for Sn =5 mole% up to 180 $cm^{-1}$ in Sn =12.5 mole% spectra. Raman spectra (Figure 2b) show that a pyramidal $SbSe_3$ peak is cited at 190 $cm^{-1}$. Addition of Sn-mole% increases the peak intensity and causes Raman shift towards 183 $cm^{-1}$, indicating the occurrence of Sn–Se bonds. The results suggest that 4, 3 and 2 are co-ordination numbers of Sn, Sb and Se, respectively, and are the most preferable coordination numbers that enhance the glass formation in the Sn–Sb–Se system.

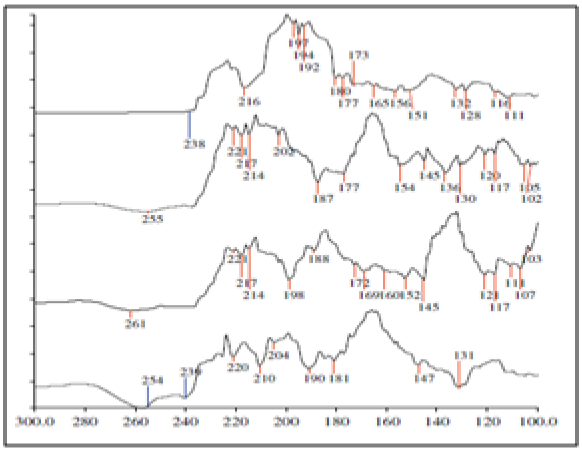

**Figure 2(a):** Infrared transmission spectra of $Sn_x$-$Sb_5$-$Se_{95-x}$ system. [35]

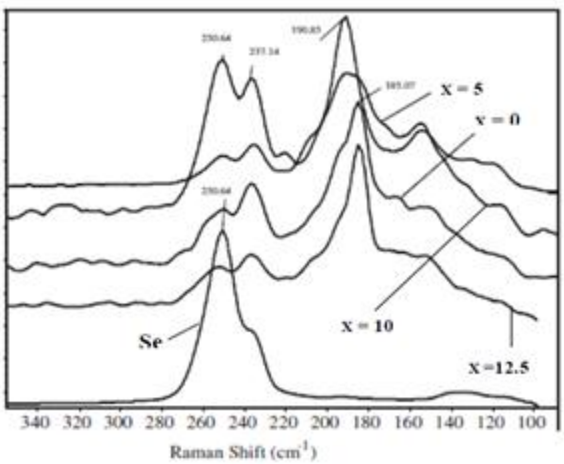

**Figure 2(b):** Raman spectra of amorphous Se and $Sn_x$-$Sb_5$-$Se_{95-x}$ system, where x= 0, 5, 10, and 12.5. [35]

**SeTeSn**

The effect of Sn additive on the Dark conductivity ($\sigma_d$) and Photoconductivity ($\sigma_{ph}$) in amorphous $Se_{85}Te_{15}$ binary alloy have been reported [36] and the results were explained on the basis of increase in the DOS present in the mobility gap and in terms of the electron affinity values of the various constituents of the thin films. The electrical measurements have been carried out on a-$Se_{85-x}Te_{15}Sn_x$ (x = 0, 2, 4, 6 and 10 at. %) thin films. Both $\sigma_d$ (Figure 3a) and $\sigma_{ph}$ were found to increase up to x = 6 at.% and then decreases. Activation energy ($\Delta E_d$) decreases up to x = 2 at. % of Sn addition, but it increases on further Sn addition. An increase in $\sigma_d$ with a corresponding decrease in the $\Delta E_d$ as the Sn concentration is increased, indicates an increase in the DOS. The charge carrier concentration ($n_\sigma$) also increases as the Sn concentration increases up to x = 6 at. % and then decreases as the concentration of Sn is increased. Photosensitivity ($\sigma_{ph}/\sigma_d$) decreases sharply and quite appreciably after the incorporation of Sn additive and having a minimum at x = 4 (Figure 3b). As the concentration of Sn is increased further, the value of $\sigma_{ph}/\sigma_d$ increases. The photosensitivity depends on the lifetime of the excess charge carriers $\tau_d$ (differential life time) which in turn depends on the density of localized states (DOS) in a particular material. The higher the density of states, the lower will be lifetime, as these defect states may act as recombination centers in presence of light. A sharp decrease in the $\sigma_{ph}/\sigma_d$ by two orders of magnitude and an increase in $\tau_d$ indicate an increase in the DOS. An increase in defect states after putting Sn concentration in binary $Se_{85}Te_{15}$ alloy may be due to the reason that the addition of Sn induce more and more positively charged localized states, since the electron affinity of Sn is much lower than that of Se and Te.

The effect of Sn incorporation in the density of defect states of two binary Se-Te glassy systems, comparing the properties of a- $Se_{0.75}Te_{0.25}$, a- $Se_{0.85}Te_{0.15}$ and a-$Se_{0.75}Te_{0.15}Sn_{0.10}$ glassy alloys were reported by Sharma et al [37]. Properties of d.c. conductivity at high electric fields in vacuum were examined; and current-voltage (I-V) characteristics have been measured at various fixed temperatures. A non-ohmic behaviour is observed at high electric fields (E ~ $10^4$ V/cm). DOS near Fermi level was calculated using the theory of SCLC for uniform distribution of traps. The addition of Sn decreases the DOS in the pure

Se-Te glassy network; the decrease is less in $Se_{0.85}Te_{0.15}$ than in $Se_{0.75}Te_{0.25}$. The difference in order of decrease in DOS in the two $Se_{0.75}Te_{0.25}$ and $Se_{0.85}Te_{0.15}$ binary glassy systems was explained on the basis of the electro-negativity difference between the constituent elements. When Sn, having lower electro-negativity than Se and Te, is introduced in $Se_{0.85}Te_{0.15}$ (density of traps near Fermi level $g_o$ $=6.93 \times 10^{14}$ $eV^{-1}cm^{-3}$), positively charged defects will be created, but the extent of their creation will be smaller since the amount of Te is same in both ($Se_{0.75}Te_{0.15}Sn_{0.10}$, $g_o = 2.34 \times 10^{14}$ $eV^{-1}cm^{-3}$. On the other hand, when the same Sn is incorporated in $Se_{0.75}Te_{0.25}$, the DOS may decrease more sharply due to decrease in percentage of Te atom in $Se_{0.75}Te_{0.15}Sn_{0.10}$ than $Se_{0.75}Te_{0.25}$ ($g_o = 1.69 \times 10^{15}$ $eV^{-1}cm^{-3}$ ) binary glassy system.

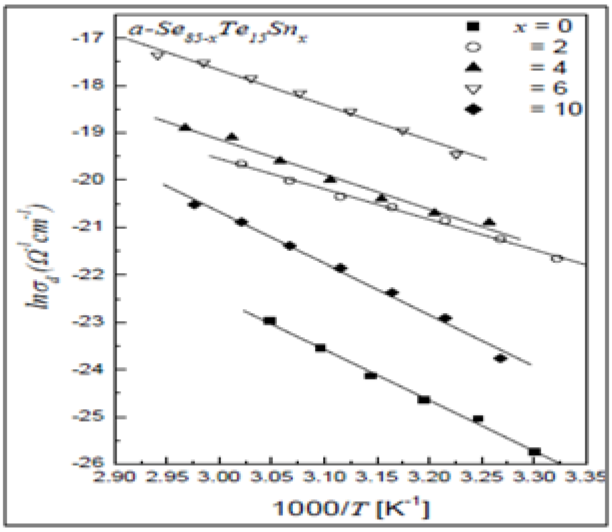

**Figure 3(a):** Temperature dependence of dark conductivity for thin films of a-$Se_{85-x}Te_{15}Sn_x$ (x = 0, 2, 4, 6 and 10 at. %) glassy alloys. [36]

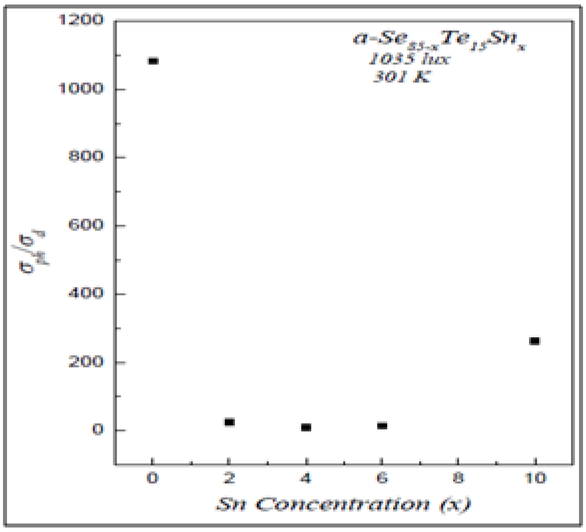

**Figure 3(b):** Composition dependence of photosensitivity ($\sigma_{ph}/\sigma_d$) at room temperature for different a-$Se_{85-x}Te_{15}Sn_x$ (x = 0, 2, 4, 6 and 10 at. %) glassy alloy thin films. [36]

**GeSeSbSn**

Kang et al [38] studied the effect of the addition of Sn on the structural, physical and electrical properties of vacuum evaporated $(Ge_{20}Se_{80})_{90-x}Sb_{10}Sn_x$(x = 2, 4, 6 and 10 at %) thin films. The temperature-dependent $\sigma_d$ measurements in the temperature range 293 K−363 K reveals that the conduction in Ge-Se-Sb-Sn thin films is through an activated process with single activation energy and $(Ge_{20}Se_{80})_{86}Sb_{10}Sn_4$ composition has the highest conductivity as compared to the other compositions in the series. The formation of weaker Sb − Sb bonds leads to a decrease in conductivity for the higher concentration of Sn element. It was found that the Meyer-Neldel rule was obeyed by $(Ge_{20}Se_{80})_{90-x}Sb_{10}Sn_x$(x = 2, 4, 6 and 10 at %) glassy systems. Chemically ordered network model, consistent with (8 − N) rule, has been utilized in estimating various physical parameters which reveals that Ge-Se-Sb-Sn glassy systems are thermally stable glasses.

## 2. Conclusion

Metal doping in chalcogenide materials attracts much interest due to their wide application in several fields of physics. In particular, Sn doping in Se based chalcogenide glasses affects the structure, electrical and optical properties of host matrix. This review helps in understanding the properties and possible applications of tin selenide-based materials for next generation technology.

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