Optical and transport properties of Se$_{0.7}$Te$_{0.3}$ and Se$_{0.3}$Te$_{0.7}$ alloy thin films

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Preparation and the study of optical and transport properties on Se$_{0.7}$Te$_{0.3}$ and Se$_{0.3}$Te$_{0.7}$ in the form of vacuum deposited thin films are undertaken. Material preparation consists of two processes, i.e., (i) preparation of bulk selenium telluride alloys and (ii) fabrication of thin films for characterising the above mentioned properties. The thicknesses of various films have been measured by multiple beam interference technique. Rutherford Backscattering Spectrometry (RBS) analysis clearly indicate the presence of the parental elements in two different compositions. The XRD studies revealed the polycrystalline nature of the Se$_{0.7}$Te$_{0.3}$ and Se$_{0.3}$Te$_{0.7}$ thin films. Optical constants in the 500-2500 nm region, bandgap and the transition type have been determined from the transmission data. Hall and thermopower measurements are made on these two different compositions and the Hall parameters are calculated.

Experimental Procedure

The Se$_{0.7}$Te$_{0.3}$ and Se$_{0.3}$Te$_{0.7}$ alloys were prepared by melting together high purity (99.999%) selenium and tellurium in a vacuum ($\approx 10^{-6}$ Torr) sealed quartz tube. The formation of the homogeneous alloy and its crystallinity were verified by X-ray diffraction technique. Thin films of glassy alloy of the two compositions were prepared onto well cleaned glass substrates kept at room temperature by thermal evaporation using 12A4 Hind Hivac Co., coating unit at a vacuum of $2.7 \times 10^{-3}$ Pa. Thicknesses of the films were measured using multiple-beam interferometry technique. Rutherford Backscattering spectrometry is an ideal tool to investigate the composition of thin films. The present work involves studies by the RBS technique using a 3 MeV tandem pelletron accelerator.

The transmission measurements have been made using a double beam UV-VIS-NIR spectrophotometer (Hitachi, Japan). From the measurements the corresponding refractive indices of the films ($n_t$) have been calculated using the expression computed by Ahmed and Khawaja.

$$T_t = \frac{n_2}{n_0} \frac{(1 + g_1)^2(1 + g_2)^2}{1 + g_1g_2^2 + 2g_1g_2\cos 2(\tau_1)}$$

where

$$g_1 = \frac{n_2^2 - n_1^2}{(n_0^2 + n_1^2)^2}$$

$$g_2 = \frac{n_1^2 - n_2^2}{(n_1^2 + n_2^2)^2}$$

$$\tau_1 = \frac{2\pi n_1 d}{\lambda}$$

For Correspondence
Where \( n_0 = 1, n_2 = 1.5, n_1 \) is the refractive index of the film, \( \lambda \) is the wavelength of the light and \( d \) is the thickness of the film.

The four-probe method of Vander Pauw has been used to measure the electrical conduction mechanism of the films of two different compositions. The measurements are made under a vacuum of 1.3 Pa. Temperature dependence of carrier concentration and mobility are studied in the temperature range 300-450 K. The thermal emf was measured from the temperature gradient between the two ends of the sample by the integral method in the temperature range 300-400 K. The experimental set-up for the thermo emf measurement consisted of a massive copper heat sink and a copper holder to which a heater was attached. With this set-up one end of the film can be kept at a constant temperature while the temperature of the other end of the film can be varied. The temperature of the hot end could be varied from 300-400 K while maintaining the cold end temperature at about 300 K, room temperature. The temperatures of the hot and cold ends of the films were measured with copper constantan thermocouple arrangement. The thermal emf developed across the films were measured with respect to copper as a function of hot end temperature with a Philips d.c. microvoltmeter. The thermo emf of the thermocouple recording the hot end temperature was measured using a digital millivoltmeter. All samples were annealed in vacuum at about 100°C for 8 h and then the electrical parameters were measured.

**Results and Discussion**

**XRD and RBS analysis**—Figs 1a and 1b shows the X-ray diffractograms obtained for \( \text{Se}_{0.7}\text{Te}_{0.3} \) and \( \text{Se}_{0.3}\text{Te}_{0.7} \) thin films of thicknesses 187 and 76 nm respectively. The observed peaks in the diffractogram reveal that the deposited films possess polycrystalline structure. Similar observations have been reported by earlier workers. Also the RBS analysis clearly indicate the presence of the parental elements in \( \text{Se}_{0.7}\text{Te}_{0.3} \) and \( \text{Se}_{0.3}\text{Te}_{0.7} \) thin films and hence their stoichiometry.

**Optical studies**—Fig. 2 illustrate the transmission characteristics of \( \text{Se}_{0.7}\text{Te}_{0.3} \) and \( \text{Se}_{0.3}\text{Te}_{0.7} \) alloy thin films respectively. The oscillatory variation of transmittance observed in \( \text{Se}_{0.7}\text{Te}_{0.3} \) films arises due to the interference maxima and minima condition being satisfied for the particular combination of wavelengths and optical thickness of the medium. In \( \text{Se}_{0.3}\text{Te}_{0.7} \) films, the transmittance increases gradually and becomes maximum in the NIR region and decreases slowly for the different thicknesses studied. As the thickness of the film increases the transmittance maximum increases and shifts towards higher wavelength in the NIR region. The transmittances decreases with the increase of film thickness when the wavelength becomes less than 1500 nm. It is clearly observed that these films have very low transmittance of 20 to 40% in the higher wavelength region (1500-2000 nm). Since Se and Te are isoelectronic, as more Te is incorporated in a-Se, both structural and compositional disorders increase. The increased disorder enhances tailing of these localized states. Soon a situation arises when these start...
dominating over the charged defect states/energies. This may be the reason that dependence of \% T on thickness is more prominent for higher Te content and the trend appears to be different for films with lower Te content.

In Se\textsubscript{0.7}Te\textsubscript{0.3} and Se\textsubscript{0.3}Te\textsubscript{0.7} thin films \((h \alpha)^2\) versus \(h\) plots (Fig. 3) reveals the existence of direct allowed transitions with optical band gap 1.4 and 0.7 eV for films of thickness 76 and 138 nm respectively. The band gap decreases with increase of film thickness. This may be due to the increase of particle size and a decrease of strain. The corresponding refractive indices \((n_g)\) have been calculated using the expression\(^7\) for Se\textsubscript{0.7}Te\textsubscript{0.3} and Se\textsubscript{0.3}Te\textsubscript{0.7} thin films. The extinction coefficients \((k_f)\) are evaluated by making use of Lambert's equation. Fig. 4 shows the plot between refractive index and wavelength \((\lambda)\) for two compositions of different thicknesses. The refractive index decreases initially and attains a constant value around 2100 nm. Such decrease in \(n_f\) value is attributed to the strong effect of surface and volume imperfections on the microscopic scale\(^11\). The extinction coefficients vary from 2.4 to 0.6 and 2.6 to 0.9 for a typical film of thicknesses 76 and 138 nm of two different compositions Se\textsubscript{0.7}Te\textsubscript{0.3} and Se\textsubscript{0.3}Te\textsubscript{0.7} respectively.

The optical band gaps of Se\textsubscript{1-x}Te\textsubscript{x} alloys \((x=0.3, 0.5\) and 0.7\) are plotted in Fig. 5 as a function of At \% concentration of Te, i.e., \((x)\). Optical band gaps of glass alloys decreases with increase in the Te concentration, but not linearly over the whole range of \(x\) investigated. It is seen that the optical band gap energy values are higher for lower Te At \%. Also optical band gap decreases faster in beginning, the decrease becomes less beyond 50 At \%. It is expected that as more Te is introduced in glassy Se-Te alloys, the compositional disorder will increase, which will lead to the enhanced tailing of the bands in the gap region.

Hall and thermopower measurements—Fig. 6 shows the variation of Hall mobility \((\mu_H)\) with absolute temperature for the two different compositions \((\text{Se}_{0.3}\text{Te}_{0.7}; \text{Se}_{0.7}\text{Te}_{0.3})\) of different thicknesses. In Se\textsubscript{0.7}Te\textsubscript{0.3} films an increase in mobility is noticed with an increase of temperature, thus following the C-W relationship\(^13\). While in Se\textsubscript{0.3}Te\textsubscript{0.7} thin films the mobility slightly increases initially and then decreases with an increase of temperature. Fig. 7 show the variation of Hall coefficient \((R_H)\) with absolute temperature for two different compositions of different thicknesses. It is seen that in Se\textsubscript{0.3}Te\textsubscript{0.7} films \(R_H\) slowly decreased attaining a

![Fig. 3](image-url)  
**Fig. 3**—Plot of \((h \alpha)^2\) versus \(h\) (Se\textsubscript{0.7}Te\textsubscript{0.3}) and (Se\textsubscript{0.3}Te\textsubscript{0.7})

![Fig. 4](image-url)  
**Fig. 4**—Variation in \(n_f\) with wavelength (Se\textsubscript{0.7}Te\textsubscript{0.3}) and (Se\textsubscript{0.3}Te\textsubscript{0.7})

![Fig. 5](image-url)  
**Fig. 5**—Variation in optical band gap with Te At \%
minimum with the increase in temperature and then sharply increased as temperature is increased. The curves for Se$_{0.7}$Te$_{0.3}$ films initially show an increase and attains a constant value at higher temperatures. Fig. 8 shows the variation of carrier concentration ($n$) with temperature for two different compositions. It is interesting to see that $n$ showed a maximum and decreased with increase of temperature and also $n$ value decreased with increase of thickness for Se$_{0.3}$Te$_{0.7}$ thin films. In Se$_{0.7}$Te$_{0.3}$ films it increases only weakly with temperature and no significant dependence on thickness was found.

Transport properties of a material will be different when compared with the properties of the same material in the bulk state at a given temperature because of the fact that in the thin film state, in addition to the normal bulk scattering processes, scattering of carriers by the external and internal surfaces occurs. Since the external surface-to-volume ratio is an inverse function of the thickness of a film and the grain size is also small thus the extent of the above kind of scattering with the thickness of the film changes. Consequently, the transport properties of films will be the functions of their thickness and temperature.

Thermo emf has been measured for two different compositions of selenium telluride films (Se$_{0.7}$Te$_{0.3}$ and Se$_{0.3}$Te$_{0.7}$). The variation of thermo emf with temperature for two compositions of typical thicknesses 193 and 138 nm respectively are
shown in Fig. 9. It is observed that the plots are linear. The thermopower for these films are calculated as 100 and 75 μV/K respectively. For both compositions all the samples appear to be p-type which indicates the current transport is primarily due to holes as a result of thermal excitation of electrons and their subsequent capture by acceptor states.

Substitution of Te into Se reduces the Se₈ ring population and slightly increases the chain fraction but reduces the chain length. The atoms can occupy either intra-chain or intra-ring or chain termination positions in α-Se randomly. Since the electronegativities of Se and Te differ, this is expected to lead to preferences in the bonding positions. All of these changes in the structure of α-Se, when Te is introduced into it, lead to changes in its energy band structure and hence its electrical and optical properties. In particular, these, modify charged defect states in the gap, enhancing the tailing of the localized states and increasing the trap concentration. In SeₓTeₙ₋ₓ films the conduction takes place via hopping through the localized states.

Conclusion

Stoichiometric Se₀.₇Te₀.₃ and Se₀.₃Te₀.₇ thin films have been prepared. RBS analysis clearly indicate the presence of the parental elements in the two compositions and hence their stoichiometry. Optical studies on these films reveal the existence of interference effects. Optical band gap values are evaluated and it is found that the band gap values decrease with the increase of film thickness. The Hall mobility, Hall coefficient and carrier concentration are determined from Hall studies for the temperature range (300-450 K). The p-type nature of the films of the two different compositions has been observed from the thermoelectric power studies.

References