Study of the electrical and optical properties of Ge$_{27}$Se$_{58}$Pb$_{15}$ chalcogenide glass

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**ABSTRACT**

The present article reports electrical and optical properties of Ge$_{27}$Se$_{58}$Pb$_{15}$ chalcogenide glass prepared through melt quenching technique. The dc electrical conductivity has been studied as a function of temperature and activation energy of conduction has been computed. The electrical study suggests that conduction in the glassy sample takes place via variable range hopping and thermally assisted tunneling of charge carriers. Thermally activated conduction is observed in high temperature range (348–423 K) while in lower temperature range (298–348 K), conduction follows Mott’s VRH model. Besides this, optical constants, i.e. energy band gap, refractive index, etc. were also computed for thin film of Ge$_{27}$Se$_{58}$Pb$_{15}$ glass and band gap value suggest that sample is a semiconductor.

1. Introduction

Chalcogenide glasses are interesting materials due to their transparency in visible or near infrared region up to 15 µm and their good stability for outdoor applications. At this wavelength, thermal imaging is done using exclusively single crystalline germanium, which is rare and expensive element. Chalcogenide glasses have taken the place of germanium in thermal imaging due to its low cost and possibility of obtaining glass fibers by molding due to its vitreous nature. Now days, chalcogenide glass lenses are widely used in infrared cameras and the optical performance of the system is the same as obtained using germanium [1–3]. Chalcogenide glasses based on Ge–Se are widely used for optical fibers for light transmission especially when short length and flexibility is required [4–10]. The structure of Ge–Se is basically comprised of Ge with coordination four and Se atoms with coordination two. When 33% of Ge is added to Se, chains or rings of Se are bridged by the tetrahedral bonds of Ge forming basic structural unit of Ge–Se system [9].

Ge–Se glasses are generally p-type but alloying it with suitable impurity like Pb can change the conduction from p type to n type [11–21]. The phenomenon of p to n transition makes them ideal materials for fabrication of p–n junction with several manufacturing advantages than the conventional p–n junction. Carrier type reversal (CTR) has been reported [17–21] in two series of Ge–Se–Pb glasses. In Pb$_x$Ge$_{42–x}$Se$_{58}$ ($x = 0–20$), CTR occurs at $x = 9$ while in Pb$_{50}$Ge$_{50}$Se$_{80}$ ($y = 17–24$), CTR occurs at $x = 21$. Therefore, it is expected that after CTR, Ge–Se–Pb glasses will show enhanced electrical properties. Beside this, study of optical band gap of Ge–Se–Pb glasses after CTR occurs is also expected to provide useful information in support of electrical properties of the system.

In view of this, present paper reports the temperature dependent electrical properties of Ge$_{27}$Se$_{58}$Pb$_{15}$ glass. The glass under consideration Ge$_{27}$Se$_{58}$Pb$_{15}$ is member of Pb$_x$Ge$_{42–x}$Se$_{58}$ ($x = 0–20$) system and is chosen arbitrarily to study enhanced conductivity due to p to n transition. $I$–$V$ characteristics of the sample have been recorded to determine dc electrical conductivity and conduction activation energy as a function of temperature. Besides this, optical constants, such as optical band gap, refractive index and extinction coefficient, etc. have also been computed for Ge$_{27}$Se$_{58}$Pb$_{15}$ glass.

2. Experimental details

The glassy sample of Ge$_{27}$Se$_{58}$Pb$_{15}$ has been prepared through melt quenching technique. The element powder of Ge, Se and Pb of high purity (99.999%) procured from Sigma Aldrich, were weighed in accordance with their atomic weight percentages and sealed in a ampoule at vacuum of 10$^{-5}$ Torr. The ampoule was then heated in two stages to avoid the sudden evaporation and deposition of the selenium to the inner wall of the quartz tube. The ampoule was heated slowly and maintained at 600°C for about 6 h followed by 900°C for 30 h with continuous rotation. The temperature of the furnace was raised at a rate of 3–4°C/min. The ampoule was cooled rapidly in ice-cooled water to obtain the glassy state. The so-produced ingot was then ground using a pestle and mortar to obtain the
powder form. For electrical measurements, the pellets of the sample (thickness 1 mm and diameter 5 mm) were made using pelletizer. For this, 30 mg of bulk powder of Ge$_{27}$Se$_{58}$Pb$_{15}$ glass was taken and pressure of 5 tons was applied for 5 min. The $I$–$V$ measurements were recorded on Keithley electrometer at room temperature, as well as elevated temperatures. Four probe set-up was used for dc conductivity measurements. The sample was sandwiched between copper electrodes of specially designed sample holder. The sample holder is then kept in a chamber having vacuum of $10^{-5}$ Torr in controlled argon atmosphere. A voltage of 10 V is applied to sample and temperature dependence of current is measured by placing a calibrated copper–constantan thermocouple close to the sample.

For optical measurements, thin film of the amorphous glassy sample was deposited by evaporating the alloy onto pre-cleaned glass substrate kept at room temperature in vacuum of $10^{-5}$ Torr in vacuum coating unit. A double beam spectrophotometer (Agilent Cary 60) was used to record the absorption and transmission spectra of thin film sample in the wavelength range 400–2500 nm at room temperature. The structural characterization of the Ge$_{27}$Se$_{58}$Pb$_{15}$ sample was done using Panalytical X pert Pro X-ray diffractometer with Cu K$_{α}$ radiation. Composition of sample was confirmed using EDXA Quanta Fe200 model.

3. Result and discussion

3.1. Structural characterization

Figure 1 shows the XRD pattern of Ge$_{27}$Se$_{58}$Pb$_{15}$ glass. No sharp peaks are observed in XRD pattern which indicate amorphous nature of sample. The composition of the sample was verified using EDXA data. Figure 2 show EDAX pattern for Ge$_{27}$Se$_{58}$Pb$_{15}$ sample.

The chemical compositions of different elements in Ge$_{27}$Se$_{58}$Pb$_{15}$ glass were found to be Ge = 26.30 at. wt%, Se = 58.55 at. wt% and Pb = 15.15 at. wt%. This is similar to the composition taken before preparation. The negligible difference in the values lies within the experimental error.

3.2. Electrical properties

The $I$–$V$ measurements were carried out at different temperatures (25, 50, 75, 100, 125, and 150°C) to observe the temperature dependence of the dc electrical conductivity of Ge$_{27}$Se$_{58}$Pb$_{15}$ glass. Figure 3 shows $I$–$V$ characteristics of Ge$_{27}$Se$_{58}$Pb$_{15}$ glass at different temperatures.

Figure 3 depicts that Ge$_{27}$Se$_{58}$Pb$_{15}$ glass follows ohm’s law in entire voltage range. Further, it was also observed that current increases with increase in voltage, as well as temperature. This indicates that sample is semiconducting in nature. The dependence of dc electrical conductivity on temperature has further been used to determine the activation energy of conduction using the following relation [22]:

$$\sigma_{dc} = \sigma_o \exp \left[ -\Delta E \right] \frac{1}{kT}$$

Where, $\sigma_o$ and $\Delta E$ represents the pre-exponential factor and activation energy, respectively, and $k$ is Boltzmann constant. The electrical conduction in amorphous semiconductors can take place by three processes viz transfer of charge carriers between delocalized states in the conduction band and valence band; transition of charge carriers in the band tails and hopping of charge carriers between localized states in the bands near the Fermi level [23–25]. In case of chalcogenide glasses, it is reported [26] that conduction is intrinsic and Fermi level is significantly shifted from the middle of the energy band gap towards the valence band. In intrinsic conduction, charge carriers hops from states close to the edge of
Figure 3. $I-V$ characteristics of Ge$_{27}$Se$_{38}$Pb$_{15$ glass at different temperatures.

The slope and intercept of the plot in higher temperature range 348–423 K was used to determine activation energy of conduction ($\Delta E$) and pre-exponential factor ($\sigma_0$) of Ge$_{27}$Se$_{38}$Pb$_{15}$ glass respectively. The values of $\Delta E$ and $\sigma_0$ are mentioned in Table 1.

Pre-exponential factor ($\sigma_0$) provides important information regarding the conduction process in chalcogenide glasses. According to Mott [23–25], for conduction in localized states, pre-exponential factor should be two to three orders smaller in magnitude than that of the conduction in extended states. Further, it was proposed that for amorphous Se and other Se rich alloys, the magnitude of $\sigma_0$ should be of the order of $10^4 \, \Omega^{-1} \, \text{cm}^{-1}$ for conduction in extended states [26,27]. In the present study, the values if $\sigma_0$ were found to be of the order of $10^3 \, \Omega^{-1} \, \text{cm}^{-1}$, therefore, localized state conduction in the tail states is likely to take place ruling out the possibility of extended state conduction.

In the lower temperature range 298–348 K, conductivity increases slowly indicating VRH conduction as suggested Mott. Mott [28,29] proposed three dimensional VRH (3D VRH) model for temperature dependence of dc electrical conductivity. The formulation is given by:

$$\sigma \sqrt{T} = \sigma' \exp \left( \frac{-B}{T^{1/4}} \right)$$  \hspace{1cm} (2)

Where, $\sigma'$ is pre-exponential factor and $B$ is given by [30]:

$$B^4 = T_o = \frac{\lambda \pi^3}{kN(E_f)}$$  \hspace{1cm} (3)

Here, $\alpha^{-1}$ describes the spatial extent of the wave function $\exp(-aR)$ associated with the localized states, $N(E_f)$ is the density of localized states at the Fermi level, $\lambda$ is a dimensionless constant (about 18), $T_o$ is degree of disorder and $k$ is Boltzmann constant.

The value of pre-exponential factor is reported as [26,27,30]

$$\sigma' = 3e^2 \gamma \left[ \frac{N(E_f)}{8\pi \alpha k T} \right]^{1/2}$$  \hspace{1cm} (4)

Where $e$ is electron charge and $\gamma$ is Debye frequency (about $10^{13}$ Hz).

Solving simultaneously Eqs. (2) and (4) gives

$$\alpha = 22.52 \sigma_o B^4 \text{cm}^{-1}$$  \hspace{1cm} (5)

and

$$N(E_f) = 2.12 \times 10^9 \times (\sigma_o')^3 B^3 \text{cm}^{-3} \text{eV}^{-1}$$  \hspace{1cm} (6)

Figure 5 shows the plot of dc electrical conductivity ($\ln \sigma(\sqrt{T})$) with $T^{-1/4}$ in the temperature range 298–348 K. From Figure 5, it is observed that conductivity increases slowly with increasing temperature.
in the temperature range 298–348 K. This suggests that conduction is due to VRH.

The values of other parameters, i.e. \(T_o, \alpha\) and \(N(E_f)\) for Ge\(_{27}\)Se\(_{58}\)Pb\(_{15}\) glass in the temperature range 298–348 K are mentioned in Table 1. The values of \(T_o\) and \(N(E_f)\) are within the range mentioned for Se-based glasses [31–33].

Mott formula for hopping distance \(R\) is given by

\[
R = \left[ \frac{9}{8\pi\alpha N(E_f)kT} \right]^{1/4} \tag{7}
\]

and hopping energy is

\[
W = \frac{3}{4\pi R^3 N(E_f)} \tag{8}
\]

Mott and Davis [28] also suggested that conduction mechanism can be identified on the basis of the value of \(W\) and \(aR\). They proposed that values of \(W\) and \(aR\) should be greater than \(kT\) (25.7 meV at 298K) and unity respectively, for VRH conduction. The values of \(W, R,\) and \(aR\) obtained for Ge\(_{27}\)Se\(_{58}\)Pb\(_{15}\) glass at different temperatures are listed in Table 2. The values of \(W\) and \(aR\) are found to be greater than \(kT\) and unity thereby satisfying the Mott’s 3D VRH model. The decrease in value of \(R\) and increase in value of \(W\) with increase in temperature is also in accordance with the concept of Mott’s VRH model.

Above study reveal that conduction mechanism changes at \(T = 348\) K. The electrical parameters obtained through both mechanisms, i.e. Arrhenius Eq. (1) and VRH Eq. (2) are electrical conductivity, pre-exponential factor and the activation energy required for conduction. The values of these parameters obtained at 348 K using both conduction mechanisms explain the observed change in conduction mechanism. The electrical conductivity at 348 K was calculated using equation \((\sigma = \frac{1}{RA})\) and was found to be \(4.45 \times 10^{-10}\) ohm\(^{-1}\) cm\(^{-1}\). The pre-exponential factor \((\sigma_o)\) obtained using thermally activated process is found to be 4.30 while using VRH, \(\sigma_o\) is found to be 12.31. Based on both Arrhenius and VRH equation, \(\sigma_o\) values suggest that for same temperature values, electrical conductivity is found to be lower in lower temperature range (303–348 K). Further, activation energy for conduction (\(\Delta E\)) for thermally activated process is found to be 5.18 eV while for conduction in lower temperature range \(\Delta E\) is found to be 0.71 eV. This suggests that in lower temperature range, low conductivity is observed due to small mobility of carriers. Low conductivity is due to absence of periodic lattice and substantial incoherent scattering. As the temperature increases, some of the localized charge carriers occasionally acquire sufficient thermal energy for conduction in band tails [28]. This conduction requires large energy for conduction, hence \(\Delta E\) is large for thermally activated process.

### 3.3. Optical properties

Figure 6 shows the transmission spectrum of thin film of Ge\(_{27}\)Se\(_{58}\)Pb\(_{15}\) glass. Figure depicts several maxima and minima, which suggest that thin film is homogeneous. The optical constants of the film have been evaluated using Swaneopeol method [34]. The upper and lower envelopes of the interference fringes were drawn and value of refractive index \((n)\) of the film in the spectral region of medium and weak absorption was evaluated using the expression [35]

\[
n = \left[ N + (N^2 - n^2)\frac{1}{2} \right]^{1/2} \tag{9}
\]

Where,

| Composition          | \(\sigma_o\) (Ω\(^{-1}\) cm\(^{-1}\)) | \(\Delta E\) (eV) | \(\sigma_o\) (Ω\(^{-1}\) cm\(^{-1}\)) | \(\alpha\) (cm\(^{-1}\)) | \(\Delta\sigma_o\) (Ω\(^{-1}\) cm\(^{-1}\)) | \(\sigma\) (Ω\(^{-1}\) cm\(^{-1}\)) | \(N(E_f)\) (cm\(^{-3}\) eV\(^{-1}\)) | \(T_o\) (K) |
|---------------------|-------------------------------------|-------------------|-------------------------------------|-------------------|-------------------|-------------------|-------------------|------------|
| Ge\(_{27}\)Se\(_{58}\)Pb\(_{15}\) | 0.86 \times 10^{-10}               | 5.18              | 4.30                                | 1.15 \times 10^{5} | 1.92 \times 10^{15} | 1.48 \times 10^{5} |                      |            |

### Table 1. Values of dc electrical conductivity \((\sigma)\) at 298 K, conduction activation energy \((\Delta E)\) and pre-exponential factor \((\sigma_o)\) in temperature range (348–423 K), spatial extent of wave function \((\alpha)\), density of states \((N(E_f))\) near fermi level and degree of disorder \((T_o)\) for Ge\(_{27}\)Se\(_{58}\)Pb\(_{15}\) glass.

| Temperature (K) | \(R\) (cm) | \(W\) (meV) | \(aR\) |
|-----------------|------------|-------------|--------|
| 298             | 1.58 \times 10^{-5} | 31           | 1.817  |
| 323             | 1.55 \times 10^{-5} | 33           | 1.782  |
| 348             | 1.52 \times 10^{-5} | 35           | 1.748  |

Table 2. Values of hopping distance \((R)\), hopping energy \((W)\) and \(aR\) for Ge\(_{27}\)Se\(_{58}\)Pb\(_{15}\) glass in the temperature range 298–348 K.
\[ N = \frac{2n_s(T_M - T_m)}{T_M T_m} + \frac{n_s^2 + 1}{2} \]  
(10)

Where \( T_M \) and \( T_m \) are the maximum and minimum values of the upper and lower envelopes at a certain wavelength (710 nm) and \( n_s \) is the refractive index of the glass substrate.

The refractive index of the glass substrate (\( n_s \)) has been calculated from the well-known relation [36]

\[ n_s = \frac{1}{T_s} + \left( \frac{1}{T_s^2} - 1 \right)^{1/2} \]  
(11)

Where \( T_s \) denotes the transmittance spectra of the glass substrate. The value of \( n_s \) for present study was found to be 1.5.

If \( n_1 \) and \( n_2 \) are the refractive indices at two adjacent maxima (or minima) at wavelengths \( \lambda_1 \) and \( \lambda_2 \) respectively then formula for film thickness is given by,

\[ d = \frac{\lambda_1 \lambda_2}{2(\lambda_1 n_2 - \lambda_2 n_1)} \]  
(12)

The above equation is derived from the basic equation of interference fringes,

\[ 2nd = m_o \lambda \]  
(13)

where the order number \( m_o \) is an integer for maxima and a half integer for minima.

Other optical constants, such as energy band gap, extinction coefficient and dielectric constant has been evaluated with the help of absorption coefficient (\( \alpha \)). Absorption coefficient is given by

\[ \alpha = \frac{1}{d} \ln \left( \frac{1}{X} \right) \]  
(14)

Here, \( d \) is the thickness of the film and \( X \) is the absorbance. The thickness of Ge\(_{27}\)Se\(_{58}\)Pb\(_{15}\) glass film is found to be 731 nm.

The extinction coefficient is then obtained from the following relation:

\[ k = \frac{\alpha \lambda}{4\pi} \]  
(15)

The values of extinction coefficient (\( k \)) and refractive index (\( n \)) has further been used to determine real (\( \varepsilon' \)) and imaginary (\( \varepsilon'' \)) dielectric constant using following relations

\[ \varepsilon' = n^2 - k^2 \]  
(16)

\[ \varepsilon'' = 2nk \]  
(17)

The values of refractive index, extinction coefficient and dielectric constant (real and imaginary) at wavelength 710 nm are mentioned in Table 3.

The value of absorption coefficient in high absorption region (\( \alpha \geq 10^4 \text{ cm}^{-1} \)) has been used to determine the band gap using Tauc relation [37] given by

\[ \alpha(h\nu) = \frac{B(h\nu - E_g)^2}{h\nu} \]  
(18)

Where, \( h\nu \) and \( E_g \) are photon energy and the optical band gap respectively. \( B \) is a parameter that depends on transition probability. Figure 7 shows the plot of \((ahv)^{1/2}\) versus \(h\nu\) for Ge\(_{27}\)Se\(_{58}\)Pb\(_{15}\) glass film. The extrapolation of the straight line to \((ahv)^{1/2} = 0\) axis gives the value of the energy band gap. The energy band gap value of Ge\(_{27}\)Se\(_{58}\)Pb\(_{15}\) glass has been mentioned in Table 3.

From Table 3, it is observed that energy band gap of Ge\(_{27}\)Se\(_{58}\)Pb\(_{15}\) glass lies within the semiconducting range (0.2–2 eV), which suggest that sample is semiconducting in nature. The electrical study of Ge\(_{27}\)Se\(_{58}\)Pb\(_{15}\) glass also indicated the temperature dependent semiconducting nature of sample.

**Table 3.** Values of refractive index (\( n \)), extinction coefficient (\( k \)), real (\( \varepsilon' \)) and imaginary (\( \varepsilon'' \)) dielectric constant and energy band gap (\( E_g \)) of Ge\(_{27}\)Se\(_{58}\)Pb\(_{15}\) glass.

| Sample            | \( n \)  | \( k \)  | \( \varepsilon' \) | \( \varepsilon'' \) | \( E_g \) (eV) |
|-------------------|---------|---------|--------------------|---------------------|--------------|
| Ge\(_{27}\)Se\(_{58}\)Pb\(_{15}\) | 1.647   | 0.012   | 2.712              | 0.041               | 1.65         |

**Figure 6.** Transmission spectrum of thin film of Ge\(_{27}\)Se\(_{58}\)Pb\(_{15}\) glass.

**Figure 7.** Plot of \((ahv)^{1/2}\) versus \(h\nu\) for Ge\(_{27}\)Se\(_{58}\)Pb\(_{15}\) glass thin film.
Further, lower values of extinction coefficient and dielectric constant makes it suitable for application in anti-reflection coatings.

4. Conclusions
The electrical study of Ge$_{27}$Se$_{83}$Pb$_{15}$ glass reveal that sample shows temperature dependent semiconducting nature and conduction in sample takes place via two conduction processes. In lower temperature range (298–348 K), conduction takes place via VRH while at higher temperature range (373–423 K), conduction takes place by thermally assisted tunneling of charge carriers. The values of hopping range and hopping energy also confirm that conduction in lower temperature range follows Mott’s VRH model. Besides this, optical constants, i.e. energy band gap, refractive index etc. were also computed for thin film of Ge$_{27}$Se$_{83}$Pb$_{15}$ glass and band gap value suggest that sample is a semiconductor. Further, lower value of extinction coefficient and dielectric constant makes this glass ideal candidate for anti-reflection coatings.

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