A.C. Conductivity and Dielectric Behaviour of Chalcogenide $Ge_x Fe_x Se_{100-2x}$ Thin Films.

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Thin chalcogenide films of $Ge_xFe_x Se_{100-2x}$ (x= 2.5,5,10 and 15 at.%) with thickness about ≈ 300 nm, have been prepared by thermal evaporation technique. A.C. conductivity $\sigma_{a.c.}$ (ω) of the prepared thin films has been measured in the frequency range (0.1-20 K Hz), over the temperature range(300-345 K).Obtained data reveal that $\sigma_{a.c.}$ (ω) obey the relation, $\sigma_{a.c.}(\omega)= A\omega^s$,and the exponent, s, was found to decrease by increasing temperature. The values of, s, of the investigated thin films lie between 0.4 to 0.9. The data were analyzed in terms of different models of a.c. conduction. It was found that, the correlated barrier hopping (C.B.H.) is the dominant conduction mechanism. The dependence of dielectric constant έ and loss factor (tan δ) on both frequency and temperature has been also treated.

Introduction:

Chalcogenide glasses exhibit unique infrared transmission and electrical properties, which makes them potentially useful for applications such as threshold and memory switching [1]. Studies of the electronic nature of amorphous material give information about its electrical behaviour and this may be related to structural properties. The disorder in atomic configuration is responsible for the existence of localized electronic states within the material. Because the charge carriers are localized, a.c. technique is often employed to probe their behaviour [2,3]. The a.c. conductivity, $\sigma_{a.c.}$, in many amorphous solids has been found experimentally to obey an equation of the type $\sigma_{a.c.}(\omega)=A\omega^s$, where ω is the angular frequency of the applied field, A is a constant and s ($\leq$ 1.0) is the frequency exponent. Different conduction mechanisms can lead to ω^s type of behaviour for a.c. conductivity, but it is not easy to decide which of those mechanisms is responsible for the observed conduction mechanism. However, the behaviour of the exponent s with temperature can help in determining the possible conduction mechanism [4]. In the present study, the a.c. conductivity, $\sigma_{a.c.}$, dielectric constant έ and loss factor (tan δ) of $Ge_xFe_x Se_{100-2x}$ (x= 2.5,5,10 and 15 at.%) thin films were measured as a function of frequency and temperature with a view to determine the possible conduction mechanism. The effect of Ge and Fe contents on both conductivity and dielectric constant has been also discussed.

Experimental Procedure:

Four compositions of the $Ge_xFe_x Se_{100-2x}$ (x= 2.5,5,10 and 15 at.%) glass system were prepared by melt quenching technique [5,6]. Thin films of the compositions were prepared by thermal evaporation technique at 10^{-6} Torr using an Edwards coating system (E-306). The measurement was carried out on sandwich structure form. Metal (gold) thick film was evaporated onto glass substrate as abase electrode followed by the sample using the co- evaporation techniques, and finally the second gold electrode was evaporated. Effective area was about 0.3 cm^2.

A PM 6304 programmable automatic RCL (Philps) meter was used for measurements. The ohmic behaviour has been checked before the measurements. A.C. conductivity $\sigma_{a.c.}$, dielectric constant έ and loss factor (tan δ) of the investigated thin films were obtained in the frequency range (0.1-20 K Hz), over the temperature range (300-345 K). Sample temperature was measured using a pre-calibrated chromel-alumel thermocouple type K placed near the sample.

Results and Discussion:

The a.c. conductivity $\sigma_{a.c.}$ of Ge_xFe_x Se100-2_x (x = 2.5, 5, 10 and 15 at.%) thin films with thickness about 300 nm has been measured in the frequency range (0.1-20 KHz), over the temperature range (300-345 K). The dependence of Ln ($\sigma_{a.c.}$) on Ln (ω) is shown in Fig. (1).

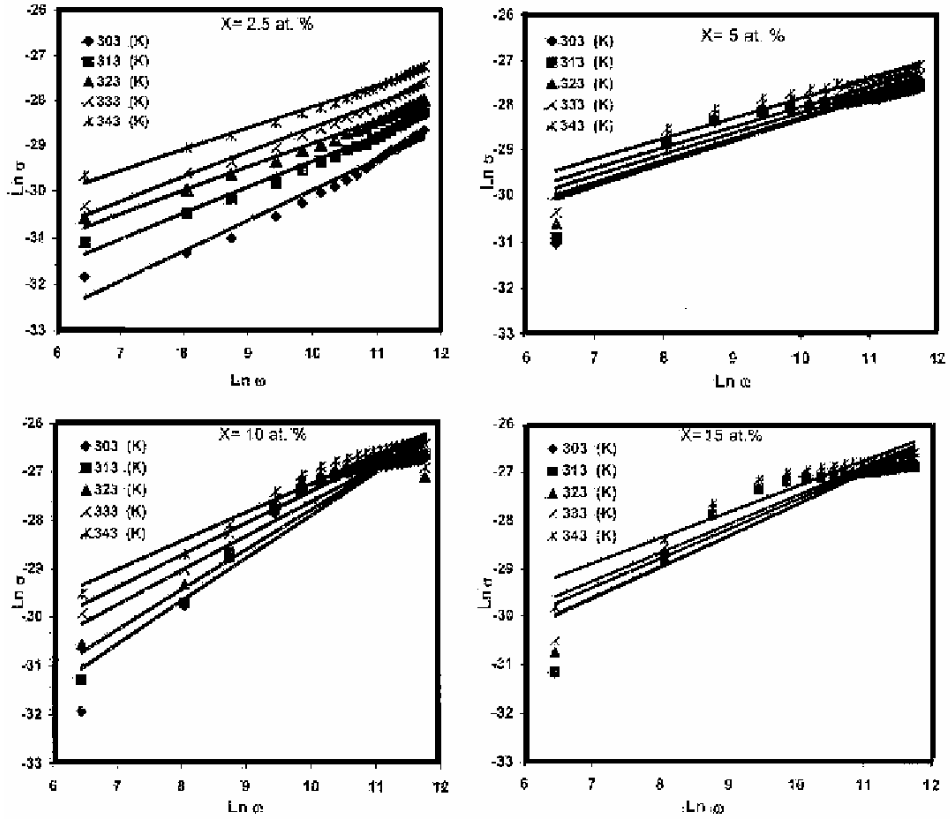

Fig. (1): Frequcency dependence of a.c. Conductivity $\sigma_{a.c.}$ for Ge_xFe_x Se_{100-2x} at various temperatures.

All samples follow a common pattern where ln ($\sigma_{a.c.}$) is linear function of ln (ω). In other words $\sigma_{a.c}$. increases with increasing frequency. Obtained data reveal that the a.c. conductivity follows the well known relation [7]:

$$\sigma_{a.c.}(\omega) = A\,\omega^s,$$

where A is constant and s ($\leq$1) is the frequency exponent. The phenomenon has been ascribed to relaxations caused by the motion of electrons or atoms. Such motion can involved hopping or tunneling between equilibrium sites [8].

Inspection of results indicates that $\sigma_{a.c.}$ increases with increasing Ge and Fe contents. The determination of a.c. Conduction mechanism implies the study of the exponent, s, as a function of temperature. The dependence of exponent, s, on temperature is shown in Fig. (2). It is observed that values of, s, lie in the range from 0.4 to 0.9. All films showed the same trend where, s, decreases by increasing temperature. Such behaviour can be observed in most amorphous semiconductors [9]. The observed behaviour of s (T) allows to conclude that, the correlated barrier hopping (C.B.H.) is the possible conduction mechanism [10].

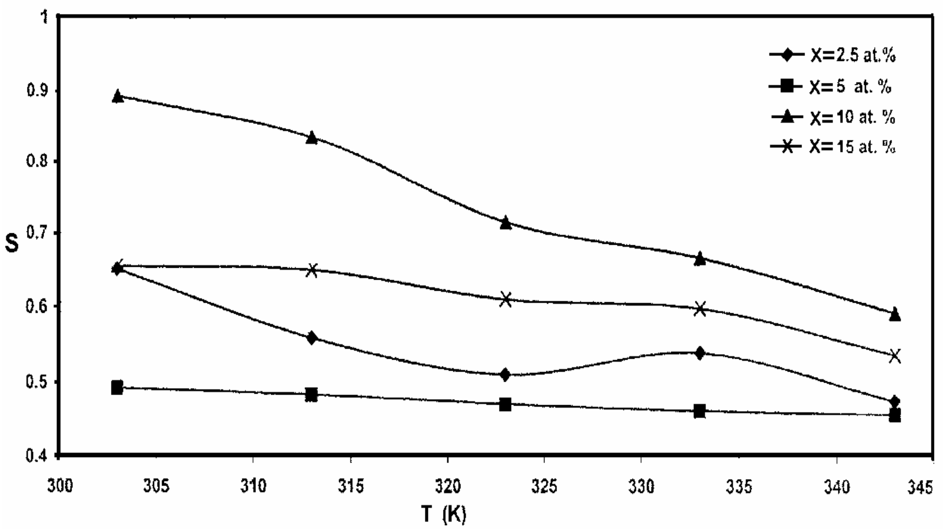

Fig. (2): Temperature dependence of frequency exponent (s) for $Ge_xFe_x Se_{100-2x}$ at various temperatures.

The dependence of a.c. conductivity $\sigma_{a.c.}$ on temperature is usually obey the well known relation [11],

$$\sigma_{a.c.} = \sigma_o \exp(- \Delta E / KT),$$

where σ_o is constant, ΔE is the activation energy, T is the absolute temperature and K is Boltzman constant. The results of the temperature dependence of the conductivity $\sigma_{a.c.}$ at a fixed frequency (10 K Hz) are shown in Fig. (3). The observed behaviour is that of semiconductors i-e $\sigma_{a.c.}$ increases with increasing temperature. The estimated values of activation energy ΔE are listed in table I. It is observed that, ΔE, decreases by increasing Ge and Fe contents. This decrease can be accounted for by the increase of disorder and the latter increases the band tail width. The observed low activation energies is in consistance with the observed dependence of a.c. conductivity $\sigma_{a.c.}$ on frequency i-e ($\sigma_{a.c.} \sim \omega^s$) [12].

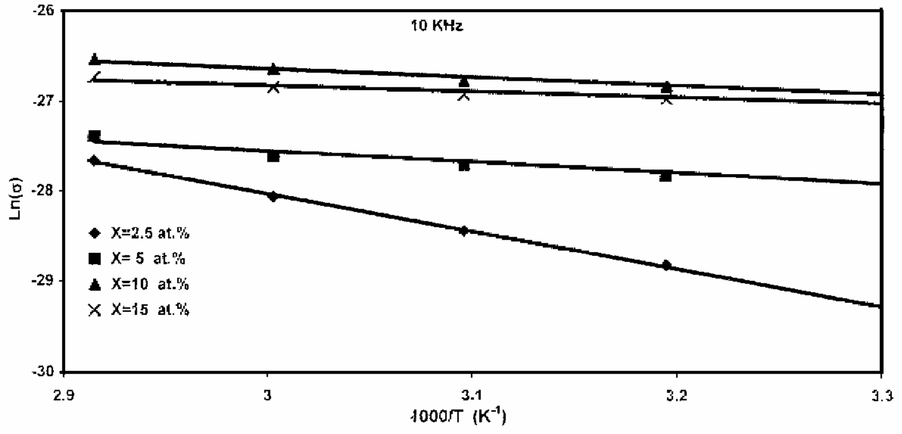

Fig. (3): Temperature dependence of a.c. conductivity, $\sigma_{a.c}$, for $Ge_xFe_x\ Se_{100-2x}$ at fixed frequency (10 K Kz).

Table (1): The values of the a.c. conductivity $\sigma_{a.c.}$, activation energy ΔE and average coordination number, calculated at room temperature and a frequency (10 K Hz) for $Ge_xFe_x\ Se_{100-2x.}$

Comp.	$\boldsymbol{\sigma_{a.c.}}$ **(Ω^{-1} cm^{-1})**	**<r>**	**ΔE (eV)**
$Ge_{2.5}Fe_{2.5}Se_{95}$	1.888×10^{-13}	2.05	0.36
$Ge_5\ Fe_5\ Se_{90}$	2.898×10^{-13}	2.10	0.11
$Ge_{10}Fe_{10}Se_{80}$	20.09×10^{-13}	2.20	0.08
$Ge_{15}Fe_{15}Se_{70}$	22.01×10^{-13}	2.30	0.06

Dielectric relaxation:

The complex dielectric constant of a material medium is represent by two parts: $\varepsilon = \acute{\varepsilon} + \varepsilon''$, where $\acute{\varepsilon}$ is the real part (dielectric constant) and ε'' the imaginary part (dielectric loss). The ratio between $\acute{\varepsilon}$ and ε'' define a loss tangent $\tan\delta = \varepsilon'' / \acute{\varepsilon}$. The corresponding real and imaginary parts of a.c. conductivity obey the following relations,

$$\sigma_1(\omega) = \sigma_{a.c.} = \varepsilon_o\,\omega\,\varepsilon''(\omega),$$

$$\sigma_2(\omega) = \varepsilon_o\,\omega\,\acute{\varepsilon}(\omega),$$

where ε_o is permittivity of free space(= 8.85 $\times10^{-12}$ F/m). The temperature dependence of $\acute{\varepsilon}$ at various frequencies, for the investigated thin films is presented in Fig. (4). It can be seen that, at high frequencies (10,20 K Hz), $\acute{\varepsilon}$

remains almost constant with increasing temperature, but at low frequencies (0.1-1 K Hz), it increases with increasing temperature. The rate of increase is different at different frequencies. On the other hand, values of έ increase by increasing Ge and Fe contents. This indicates that, the increases of Ge and Fe contents play a considerable role in dielectric constant.

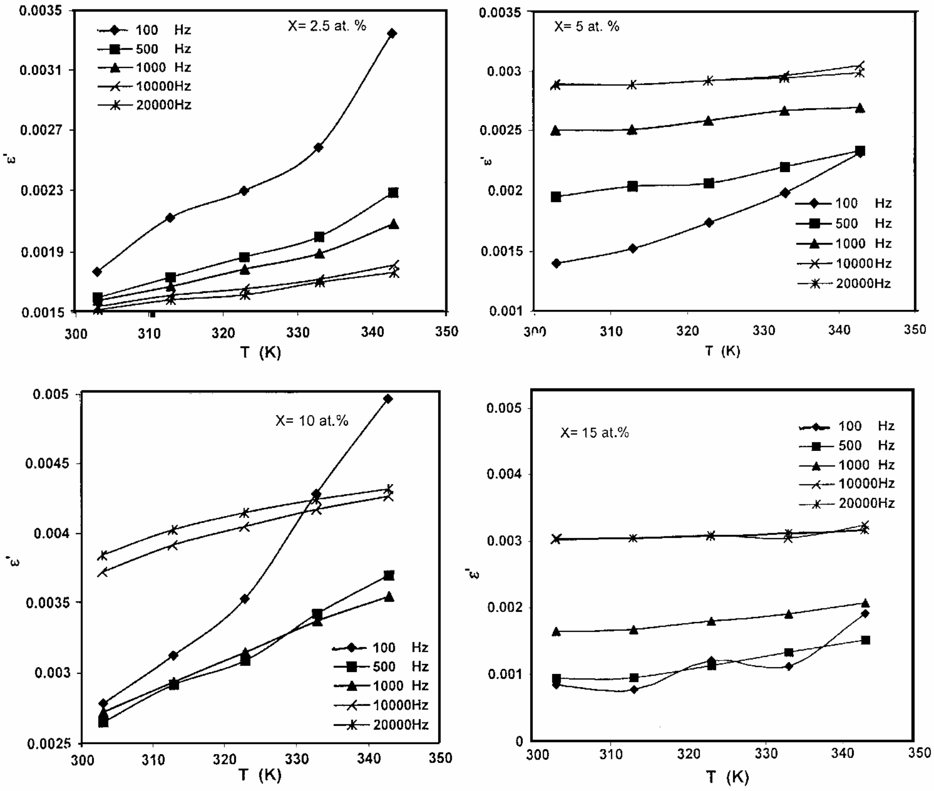

Fig. (4): Temperature dependence of dielectric constant, ε' , for $Ge_xFe_x Se_{100-2x}$ at various frequencies.

Figure (5) shows the temperature dependence of loss factor (tan δ) at various frequencies. It can be seen that (tan δ) increases with increasing temperature and the variation is more pronounced above 1.0 K Hz, but it remains almost constant at (10,20 K Hz). Moreover the increase of Ge and Fe contents leads to decrease of (tan δ). It may be noted that, $\sigma_{a.c.}$ is related to έ and (tan δ). Therefore as the temperature increases, the conductivity of thin film, and hence both έ and (tan δ) would increase.

From the above results, it can be seen that, both έ and (tan δ) exhibit strong temperature dependence at higher temperature and lower frequencies. This behaviour can be accounted for by assuming that the contribution of dipole orientation would be insignificant [13]. The main contribution to the dielectric constant in the high frequency region may be due to the electronic polarization. The έ is strongly temperature dependent in the high-temperature region. This may related to the fact that, at high temperatures the loss is dominated by thermally activated electron hopping where as in the low-temperature region such an activated process is frozen out, resulting in a decrease of έ at low temperature[14].

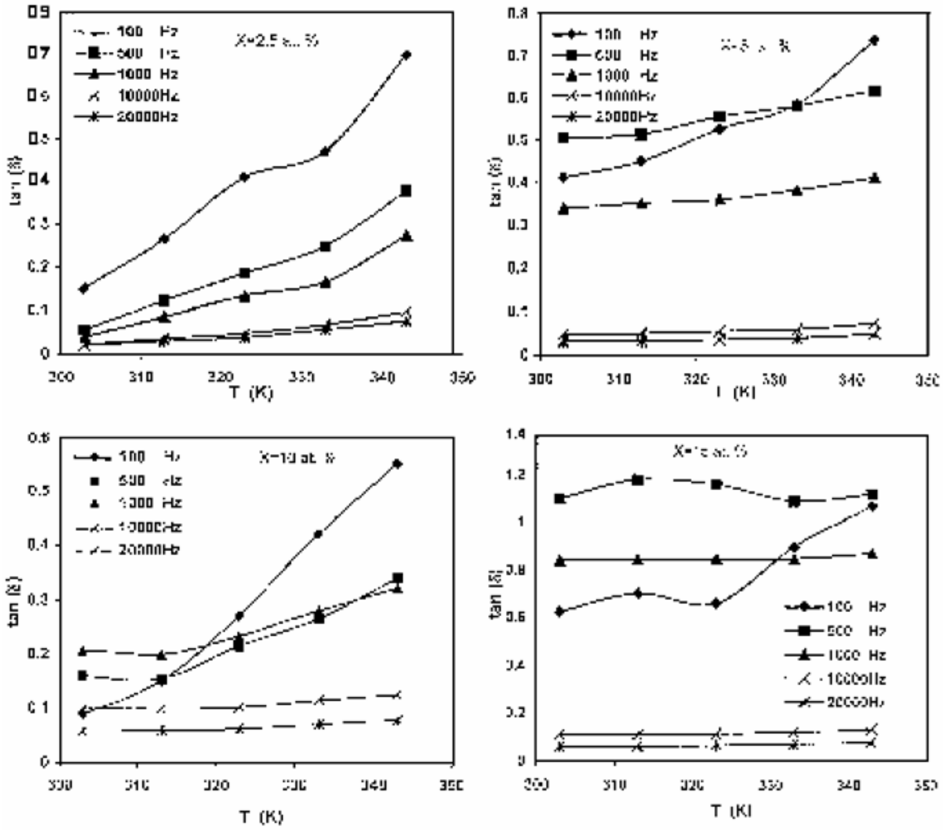

Fig. (5): Temperature dependence of tan δ for $Ge_xFe_x\ Se_{100-2x}$ at various frequencies.

Conclusions:

The a.c. conductivity $\sigma_{a.c.}$ of the investigated thin films is observed to be proportional to ω^s where the exponent, s, decreases by increasing temperature. This behaviour is found to be consistent with the Elliot model (correlated barrier hopping). The dependence of dielectric constant έ and loss factor (tan δ) on both frequency and temperature could be discussed with the frame of the effect of Ge and Fe contents on both conductivity and dielectric behaviour.

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