# Electrical Relaxation Studies of Chalcogenide xAg<sub>2</sub>S-(1-x) (0.5S-0.5Te) Glassy System

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**ABSTRACT:** Electrical relaxation of  $xAg_2S - (1-x)$  (0.5S - 0.5Te) chalcogenide glassy system has been studied in the frequency range from 42Hz to 5Mz and temperature range from 433K to 523K. Dielectric constant and dielectric loss exhibit frequency and temperature dependency. Attempts have been made to analyse dielectric relaxation data using Hunt's Model. Activation energies estimated from permittivity and complex modulus plots have been found to increase with Ag<sub>2</sub>S content. To measure DC electrical conductivity, two point probe technique has been used. Activation energies of them have also been calculated from DC conductivity measurement using Arrhenius equation. Experimental results show the dependency of DC conductivity with Ag<sub>2</sub>S content in both lower and higher temperature regions, which is found to be in close agreement with the results, obtained from dielectric relaxation.

**KEYWORDS:** Chalcogenide glasses, Transition Metal Ions, Dielectric constant, Dielectric loss, Electric Modulus, Activation energy

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# **1.INTRODUCTION**

Chalcogenide glass, the glass which contains group 16 elements of periodic table i.e., Sulphur, Selenium and/or Tellurium in addition with some more electropositive elements, acts differently from oxide glass because of its dissimilar electrical, optical, mechanical and other properties[1]. One of the major differences between oxide and chalcogenide glasses is their band gap energies [1]. Oxide glasses exhibit a wider band gap but chalcogenide glasses belong to a category of amorphous semiconductors in which band gap energies lie between 1eV and 3eV [1]. These glasses are very attractive to researchers because it is possible to tune its different properties like electrical, electronics, mechanical, chemical etc by adding impurities to this disordered system which has a conspicuous effect on changing their structures, which leads to change its conduction mechanisms [2]. This effect can be varied by varying the impurities in the system [2, 3]. Some researchers have studied the effects of different impurities in various chalcogenide glasses [4-6]. In Transition metal ions (TMI) doped glasses the small polarons forming tendency is very high [7]. The presence of valence electrons in more than one shell makes these metals intriguing to researchers [7]. These elements often exhibit several oxidation states [7]. Hopping of small polarons between different valence states of these elements can be considered as the cause of conduction in these glasses [7]. The electrical conductivity of various glasses containing TMI has been investigated by many researchers [2, 8-22].

Chalcogenide glasses have shown their potential in many applications due to their unique properties and their ability to tailor these properties according to desired requirements [6]. These glasses can be used in optical DVDs, non volatile memory devices, as waveguides and optical fibres. These glasses can also be used in the fabrication of inexpensive solar cells. electrical switches, temperature sensors, gas sensors, piezoelectric devices, LEDs, electro-photographyetc [23-35]. To know the conduction mechanism in chalcogenide semiconductors, the study of frequency-dependent electrical conductivity is very important which gives information about the transport process in localized state in the forbidden gap and the type of polarization [36]. AC conductivity and dielectric measurements of different chalcogenide semiconductors have been reported by many researchers [37-46]. But in depth investigation on them is still pending.

In this paper, the dielectric relaxation studies of  $xAg_2S-(1-x)(0.5S - 0.5Te)$ , with x = 0.35 & 0.45, chalcogenide glassy system have been reported. No research work reported by other researchers is available on this system. The motivation of this work is to study the thermal and compositional effects of dielectric relaxation and hence the electrical

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properties of Silver-Sulphide doped Sulphur-Tellurium chalcogenide glassy alloys.

# **2. EXPERIMENTAL**

The conventional melt quenching method has been used to prepare the samples of chalcogenide glassy alloys of composition, xAg<sub>2</sub>S-(1-x)(0.5S-0.5Te), with x= 0.35 & 0.45, from reagent grade chemicals Ag<sub>2</sub>S, S & Te with high purity (Adrich 99.9%). The chemicals in the appropriate proportions were mixed using a mortar and sealed in quartz ampoules under a vacuum of 10<sup>-3</sup> Torr. The mixture of chemicals in the sealed ampoules was then heated at 200°C for one hour in an electrical furnace. To obtain glassy samples, the melt was quickly quenched in ice. The glassy samples were then ground into fine powder in a mortar. To carry out dielectric relaxation analysis in the present system, electrical measurements have been done using Hioki (Model No. 3532-50) high precision LCR meter in the frequency range from 42Hz to 5MHz at different temperatures (433K to 523K).

# 3. RESULTS AND DISCUSSIONS 3.1. Dielectric Permittivity

The electrical properties of a material as a function of frequency of applied electric field, temperature and crystal structure can be obtained from dielectric

relaxation study of the materials[47]. The complex

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$$\varepsilon^* = \varepsilon / - j\varepsilon // \tag{1}$$

where  $\varepsilon'$  is the real part of the permittivity called dielectric constant which represents the maximum amount of energy that can be stored in the dielectric material and  $\varepsilon'/$  is the imaginary part, which indicates the dielectric loss, in terms of energy loss in the sample[47].The dielectric constant  $\varepsilon'$  can be estimated by using the relation:

$$\varepsilon' = C_p t / \varepsilon_0 A$$
 (2)

where  $C_P$  is the capacitance of the sample in Farad, t is the thickness of the sample under study,  $\epsilon_0$  is the permittivity of free space (8.854x10^{-12} F/m) and A is the area of the sample under study.  $\epsilon^{\prime/}$  can be presented as:



Fig.1.  $\epsilon'$  versus log  $\omega$  plot of sample with (a) x=0.35 and (b) x=0.45



Fig.2.  $\varepsilon''$  versus log  $\omega$  plot of sample with (a) x=0.35 and (b) x=0.45

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#### $\varepsilon^{\prime\prime} = \varepsilon^{\prime} \tan \delta$

where, tan  $\delta$  is the dielectric loss tangent, which is proportional to the loss of energy dissipated as heat in the sample.

(3)

The frequency dependence of dielectric constant  $\epsilon$ / of the as prepared sample, with x = 0.35 and 0.45, is shown in Fig. 1(a) and 1(b) respectively at different temperatures. Figs. 2 (a) and (b) represents the dielectric loss  $\epsilon^{\prime\prime}$  versus frequency plots at various temperatures for x = 0.35 &x= 0.45 respectively. It is observed from Fig.1 & Fig. 2 that both  $\epsilon'$  and  $\epsilon'/are$ found to decrease with the frequency. High values of dielectric constant in the low frequency region are due to electrode polarization, occurred from the accumulation of space charge at glass-electrode interface [38].As the frequency increases, the values of both  $\varepsilon$ / and  $\varepsilon$ //are found to decrease because the variation of the electric field becomes too fast for molecular dipoles to follow, which reduces the involvement in polarization. In higher frequency region, both  $\varepsilon'$  and  $\varepsilon''$  shows frequency independent behaviour. It can also be observed from Fig.1 & Fig. 2 that both  $\epsilon'$  and  $\epsilon''$ are found to increase with temperature. This effect is usually associated with the decrease in bond energies as the rise in temperature weakens the intermolecular forces and increases the thermal agitation and hence enhances/disturbs the orientational vibrations, which may increase polarization. It can also be observed from Fig. 1 and Fig. 2that both the dielectric constant  $\varepsilon$ / and dielectric loss  $\varepsilon$ // are found to increase with the decrease of Ag<sub>2</sub>S concentration in the compositions, which may be ascribed to the

$$\omega_{\rm m} = \omega_0 \exp[-(W_{\rm f}/K_{\rm B}T)] \tag{6}$$

where,  $\omega_0$  is the pre-factor,  $W_f$  is the activation energy related with dielectric loss process,  $K_B$  is the Boltzmann constant and T is the absolute temperature. In Fig.3, angular frequency,  $\omega$ , values at different temperatures and at fixed dielectric loss,  $\epsilon'/=50000$ , is presented. The data for both x=0.35 and 0.45 have been linearly fitted and the activation energy related to the dielectric loss process,  $W_f$ , has higher conductivity of the glassy system,  $xAg_2S - (1 - x) (0.5Te - 0.5S)$  with x = 0.35, compared to that with x= 0.45.

To analysis the dielectric relaxation processes Hunt's Model [48, 49]has been used. According to Hunt's Model [48, 49], the total conductivity can be expressed in two different frequency domains,  $\omega$ <  $\omega_m \& \omega > \omega_m$ , where  $\omega_m$  represents the peak frequency in dielectric loss plots. In the high frequency region  $(\omega > \omega_m)$  the hopping of carriers in pairs between ions is considered to be responsible for relaxation process, whereas in low frequency region ( $\omega < \omega_m$ ) the relaxation process can be described as the transportation of individual particles over macroscopic distance in clusters [50]. The conductivity in these two regions can be expressed as

 $\sigma_{t}(\omega) = \sigma_{dc} (1 + A(\omega/\omega_{m})^{s}) \text{ for } \omega > \omega_{m}$ (4)

$$\sigma_{t}(\omega) = \sigma_{dc}(1 + K(d)(\omega/\omega_{m})^{r}) \text{ for } \omega < \omega_{m}$$
(5)

where r=1+d-d<sub>f</sub>, d is the dimensionality of space containing pertinent clusters, d<sub>f</sub> is the functional dimensionality of clusters, A & K(d) are constants. In Fig. 2, we can see that due to the absence of any peak, the characteristics frequency,  $\omega_m$  of the dielectric loss for the present glassy system cannot be determined. It has been reported by some researchers [48-50] that the peak frequency  $\omega_m$ follows Arrhenius temperature dependence, which can be expressed by the following expression:

been calculated from the slopes of the sample best fitted lines. Table 1 shows the values of  $W_f$  for x=0.35 & 0.45. It can be observed from Table 1 that the activation energy related to the dielectric loss increases with  $Ag_2S$  content in the compositions.

### 3.2. Electric Modulus

The complex electric modulus spectrum describes the electrical relaxation and microscopic properties of the present glassy system.



Fig.3.  $\log_{10}\omega$  versus 1000/T at  $\epsilon$ " = 50000 of glass xAg<sub>2</sub>S-(1-x)(0.5Te-0.5S) where x=0.35 & 0.45.



Fig.4. (a) M' versus log  $\omega$ , (b) M" versus log  $\omega$  plots of the sample xAg2S-(1-x)(0.5Te-0.5S) where x=0.35



Fig.5. log  $\omega_m$  versus 1000/T plot obtained from M" peak at different temperatures of the glass  $xAg_2S$ -(1-x)(0.5S-0.5Te) where x=0.35 & 0.45

Table 1 Activation energies of the glass xAg <sub>2</sub> S – (1- x) (0.5S – 0.5Te) obtained from D	Dielectric		
loss & Electric Modulus plots			

x	Activation Energy from Dielectric loss plots, W <sub>f</sub> (eV)	Activation Energy from Electric Modulus, E <sub>m</sub> (eV)
0.35	0.36	0.4344
0.45	0.824	0.6149

The advantage for introducing complex electric modulus,  $M(\omega)$  spectra is to reflect the dynamic properties of the samples alone by excluding the polarization effects at sample-electrolyte interface[51].The complex electric modulus is defined as the reciprocal of the complex permittivity [51] and is represented as:

$$M^* = (\varepsilon^*)^{-1} \tag{7}$$

$$M^* = M' - jM''$$
(8)

$$M^{*} = \varepsilon^{//[(\varepsilon^{/})^{2} + (\varepsilon^{//})^{2}] + j \cdot \varepsilon^{///[(\varepsilon^{/})^{2} + (\varepsilon^{//})^{2}]}$$
(9)

where,  $M^*$  is the complex conjugate of electric modulus,  $\epsilon^*$  is the complex conjugate dielectric permittivity, M' is the real and M'' is the imaginary parts of the electric modulus respectively.

The real and imaginary parts of the electric modulus can be calculated using Eq. (9). Fig. 4 (a) and (b) show the frequency dependence of real and imaginary parts of the electric modulus respectively for x = 0.35. Similar nature for M/and M// plots have been obtained for the system with x=0.45. It can be concluded from Fig. 4(a) that at all the temperatures M/ approaches zero at low frequencies due to the suppression of the electrode polarization. It can be observed from Fig 4(b) that with the increase of temperature, the  $M^{//}$  peak has been shifted to higher frequencies, which indicates that the relaxation process in this glassy system is dependent on temperature. The shifting of M// peak is an indication of the system stabilization in a short time for an external force at elevated temperature. The M<sup>//</sup> peak indicates the frequency at which transition of charge carriers mobility takes place from long range to short range

distances. In Fig.5, the variation of peak frequency  $\omega_m$ , obtained from Fig 4(b), with reciprocal temperature is shown. Thermally activated  $\omega_m$  can be expressed from the Arrhenius relation:

 $\omega_{\rm m} = \omega_0 \exp(-E_{\rm m}/KT)$ (10)where, $\omega_m$  is the frequency at which M// peak occurs, E<sub>m</sub> is the activation energy corresponding to  $\omega_m$ , Kis the Boltzmann constant and T is the temperature in Kelvin. Emcan be estimated from the slopes of the least-squares straight line fitting of the log  $\omega_m$  versus 1000/T plot. In Table 1,  $E_m$ values are also included. It is observed from the Table 1 that the activation energy (from complex modulus plots) increases and hence the relaxation time ( $\tau$ ) increases, as $\omega_{m}$ . $\tau = 1$  [52, 53], with Ag<sub>2</sub>S content. This result is in close agreement with the activation energy obtained from dielectric loss of the glass. It is not essential to discuss a comparative analysis on the validity of Eq. (6) and Eq. (10). The concept of introducing permittivity does not yield  $\omega_m$ . So Hunt's model does not utilize to explain conductivity spectra. But this barrier can be overcome by considering the concept of electric modulus spectra. Distinct peaks in the M// versus frequency plots may give the validity of Hunt's model over here.

# **4. CONCLUSION**

The dielectric permittivity and electric modulus analysis have been carried out over the frequency range from 42 Hz to 5MHz and at different temperatures (433K-523K) for the chalcogenide glassy system  $xAg_2S - (1 - x) (0.5S - 0.5Te)$ , with x = 0.35 & 0.45, prepared by melt quenching method. It has been observed from permittivity relaxation studies that the dielectric constant and corresponding loss are found to decrease with frequency, which generally occurs due to electrode polarization, and are found to increase with temperature, which is associated with the decrease in bond energies. The electric modulus study reveals the temperature dependency of relaxation processes in this glassy system. Activation energies of these glasses have been calculated from dielectric loss and complex modulus analysis. Observations show the increase of activation energies and hence decrease of relaxation time with theAg<sub>2</sub>S content in the compositions.

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