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Role of Additives (Cd and Ge) on the Ac Conductivity and Dielectric Relaxation of $Se_{70}Te_{30}$ Glassy Alloy

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Strictly as per the compliance and regulations of :

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I. INTRODUCTION

In chalcogenide glasses, the studies of structural and chemical disorders which are due to the absence of long-range order as well as various inherent defects [1-3] in the mobility gap are very important for better understanding of transport mechanisms. The hopping conduction can be easily distinguished from that of band conduction by measuring the frequency dependence of conductivity [4], which as expected, is due to conduction in localized states. Therefore, the measurements of ac conductivity of amorphous chalcogenides can be used to obtain information about these states. This led to adapt and elaborate models allowing the electronic properties of these materials to be described [5-9]. Generally, these glasses have poor thermo mechanical properties. In order to enlarge their domain of application, it is necessary to increase their softening temperatures and mechanical strength from application and practical point of view. This leads to the necessity of changing the composition by incorporating the additives to affect the properties of binary systems as Se-Te glassy alloy system [10].

The semi-conducting compounds belonging to the cadmium chalcogenide family such as $CdSe_xTe_{1-x}$ can be advantageously used for various technical applications in particular for the conversion in photovoltaic or photo-electrochemical devices. This material with its near-ideal band gap and a high optical absorbance is a promising material for low-cost terrestrial applications.

Addition of Cd to the Se-Te system is useful in elucidating the transport mechanism in chalcogenide glasses. As well the results obtained by the incorporation of Cd can be discussed in terms of structural transitions in chalcogenide glasses [11]. In Se-Te-Ge, Ge is expected to modify the structure of Se-Te system due to its tetrahedral structure, which is quite stable as compared to chain like structure of Se-Te. Incorporation of Ge in binary Se-Te glassy system results more thermal stability in wide temperature range due to its high melting point [12]. Investigations of the temperature and frequency dependence of conductivity, the effect of impurities on the activation energy, and the effect of high electric field on the conduction mechanism is a subject of great interest, because the result of such studies provide methods to control effectively the conductivity of amorphous semiconductors.

Study of a.c conduction in Se-Te system with addition of different elements has been done by various workers [13-15]. The general frequency behavior in this type of materials is of $A\omega^s$ type over a given frequency range. Temperature dependent exponent s is determined to have a value ≤ 1. Various models have been formulated to explain this behavior and one of the starting points in these models has been formulated by Elliot in which spin pairing is energetically favoured. Hence all defect centers are either positively or negatively charged and bipolarons hop between doubly occupied D^+ states to nearby D^- states over the barrier separating them. The barrier height is correlated with the intersite separation through the Coulombic interaction between the centers. This correlated barrier hopping (CBH) model gives a temperature - dependent exponent s. This behavior is different from that predicted by the quantum mechanical tunneling (QMT) model which assumes that the a.c conduction is due mainly to the tunneling of an electron through a barrier separating

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localized states, and comes up with an exponent s that is not temperature dependent.

The present communication reports some experimental results on ac electrical conductivity and dielectric properties over the range of temperature 300 K–350 K and frequency 1 KHz–5 MHz for aforesaid glassy alloys and discuss these results in terms of the above mentioned transport models.The concept of the CBH model, incorporating the suggestions of Shimakawa, is applied to the present data. The other parameters related to dielectric measurements are also calculated and studied.

II. Experimental

Glassy alloys of $Se_{70}Te_{30}$, $Se_{70}Te_{20}Cd_{10}$ and $Se_{70}Te_{20}Ge_{10}$ systems were prepared by quenching technique. High purity (99.999 %) materials was weighed according to their atomic percentages and were sealed in quartz ampoules (length ~ 5 cm and internal dia ~ 8 mm) with a vacuum ~ 10^{-5} Torr. The ampoules containing the materials were heated to 1000°C and held at that temperature for 10 - 12 hours. The temperature of the furnace was raised slowly at a rate ~ 3 - 4 °C/min. During heating, all the ampoules were constantly rocked, by rotating a ceramic rod to which the ampoules are tucked away in the furnace for the homogeneity of the synthesized compositions. After rocking for about 10 hours, the obtained melts were cooled rapidly by removing the ampoules from the furnace and dropping to ice-cooled water. The quenched samples were taken out by breaking the quartz ampoules. Figure 1 shows the XRD plots of these glassy alloys. Compositional analysis was performed using electron probe micro- analysis (EPMA) technique.

Pellets of diameter ~10 mm and thickness ~ (1-2) mm were prepared by compressing the finely grounded powder in a die in a hydraulic press under a load of ~ 3-4 tons. Measurements were performed after coating the pellets with indium film deposited by vacuum evaporation technique. A specially designed metallic sample holder was used for the measurements of dielectric parameters in a vacuum ~ 10^{-3} Torr. The pellets were mounted in between two steel electrodes of the sample holder. The temperature was measured with the help of a calibrated copper-constantan thermocouple mounted very near to the sample, which could give measurements of temperature with an accuracy of 1^0C. For measuring real and imaginary part of dielectric constant and a. c. conductivity, conductance and capacitance were measured using a "Hioki 3532-50 LCR Hi TESTER" and by applying known relations, all the parameters are calculated. Three terminal measurements were performed to avoid the stray capacitances.

Microsoft Excel programming has been used as sources of all lines drawn on a figure. Method of least square fitting is shown having correlation coefficient (R) of the fit between 0.9 and 1 for almost all the lines obtained in the figures. The values of R^2 of all the lines plotted in the figures have been shown below the lines itself. The error bars on the data in the figures have been shown which has been estimated by the permissible error of the measuring instruments.

III. Results and Discussion

a) *Frequency dependence of ac conductivity*

A common feature of all semiconductor and some other disordered systems is that ac conductivity is frequency dependent which, as expected, is due to conduction in localized states. The ac conductivity σ_{ac} (ω) was found to obey the following relation [16]

$$\sigma_{ac} = A\,\omega^{s} \qquad (1)$$

where $s \leq 1$

An alternate model by Pike [17] and Elliot [6] considers the a.c conductivity as the result of the simultaneous hopping of two carriers between oppositely charged defect centers under the influence of the external electric field. This model known as the CBH model, expresses the a.c conductivity to a first-order approximation as,

$$\sigma(\omega,T) = \Pi^2 N^2 \varepsilon' /24\ (8e^2/\varepsilon' W_m)^6\, \omega^s / \tau_0^{1-s} \qquad (2)$$

where

$$s = 1 - 6kT/W_m \qquad (3)$$

According to QMT model, the a.c. conductivity can be expressed as

$$\sigma_{ac}(\omega) = B K_B T [N(E_F)]^2 \alpha^{-5} \omega \{\ln(\nu_{ph}/\omega)\}^4 \qquad (4)$$

Where B was found to be equal to be $\Pi/3$ by Mott and Davis and $\Pi/96$ by Pollak. K is the Boltzmann constant, ε' is the dielectric constant, W_m is the maximum barrier height over which the electrons hop, τ_0 a characteristic relaxation time and N the spatial density of defect states. The frequency dependence of σ_{ac} for the investigated compositions is studied in the frequency range 1 KHz- 5 MHz at different temperatures. Figure 2 shows frequency dependence of σ_{ac} (ω) for the aforesaid compositions. It is clear from this figure that σ_{ac} (ω) increases linearly with increasing frequency. The exponent *s* is calculated from the slope of the curves plotted in Figure 2.

Temperature dependence of s is plotted in Figure 3. It is clearly seen from Figure 3 that the slope of the curves decreases with increasing temperature, indicating that s has temperature dependence. This behavior of s is in contradiction with the QMT model which is based on phonon assisted tunneling of electrons between localized states [18-21]. This model predicts a temperature-independent *s* value of 0.8. Thus the experiment result agrees with the correlated barrier hopping (CBH) [22], for a critical test of the CBH models comes from the temperature dependence of the a.c conductivity and the frequency exponent. The calculated values of σ_{ac} (ω) and s by CBH and QMT models are given in Table 1.

b) Temperature dependence of ac conductivity

The temperature dependence of ac conductivity was studied for $Se_{70}Te_{30}$, $Se_{70}Te_{20}Cd_{10}$ and $Se_{70}Te_{20}Ge_{10}$ at different frequencies. Figure 4 shows plot of ln σ_{ac} vs 1000/T for the aforesaid glassy alloys. It is clear from this figure that σ_{ac} increase linearly with decreasing the reciprocal of absolute temperature. This suggested that the ac conductivity is the thermally activated process from different localized states in the gap or its tail. The activation energy of conduction ΔE (ω) is calculated from the slopes of lines of lnσ_{ac} vs 1000/T curves by the aid of well- known relation:

$$\sigma(\omega) = \sigma_0(\omega)\ \exp\left(-\Delta E\ (\omega)/\ kT\right) \qquad (5)$$

where σ_0 is the pre-exponential factor. The values of ΔE at different frequencies for various glassy alloys are given in Table 2.

According to CBH model the charge carriers hop across the defects states D^+ and D^- . Each pair of D^+ and D^- is assumed to form a dipole with relaxation, which can be attributed to the existence of a potential barrier over which the carriers must hop [23]. It must be noticed here that the process of hopping of the charge carriers is affected also by the density of localised states $N(E_F)$, which can be calculated using the equation [24]

$$\sigma_{ac}(\omega)=1/3\Pi e^2 K_B T[N(E_F)]^2\alpha^{-5}\omega\{\ln(v_{ph}/\omega)\}^4 \qquad (6)$$

where e is the electronic charge, v_{ph} is the predominant phonon frequency and α is the exponential decay parameter of the localized states wave functions.

Taking the reported values of v_{ph} and α^{-5} [8], the density of localized states N (E_F) was calculated at frequency 1 KHz and temperature 300K. The obtained values of N (E_F) are given in Table 3 for the studied compositions. It is clear from the results obtained that density of states decreases with aforesaid impurity incorporation. Accordingly σ_{ac} (ω) also decreases with the impurity.

c) Temperature and frequency dependence of dielectric constant

Figure 5 declares that ε' increases slowly at lower temperature then increases sharply with the increase of temperature. This behavior was observed in all the three compositions studied at present. Since in chalcogenide glasses, the dielectric properties can be interpreted by considering a set of dipoles as long as the temperature is increased high. Each dipole has a relaxation time depending on its activation energy which is attributed to the existence of a potential barrier over which the carrier can hop. Because of containing dipoles, the contribution due to dipolar or orientational polarization dominates at low frequencies. As the frequency increases high, dipolar or orientational polarization normally removes due to inertia of molecules and the electronic polarization contributes. Thus the dispersion is low at high frequencies.

Since the Orientational or Dipolar polarization is associated with the thermal motion of pair of charges, the orientation of the dipoles increases as the temperature increases, leading to the increase of dielectric constant (ε'). It has also been found that the ε' decreases with increasing frequency. In dielectric spectroscopy, large frequency dependent contributions to the dielectric response, especially at low frequencies, may come from build-ups of charge. This, so-called Maxwell-Wagner polarization occurs either at inner dielectric boundary layers on a microscopic scale, or at the external electrode-sample interface on a macroscopic scale. In both cases this leads to a separation of charges. The charges are often separated over a considerable distance, and the contribution to dielectric response can therefore be orders of magnitude larger than the dielectric response due to molecular vibration which occurs at high frequencies because at high frequencies, the energy is too high to cause rotation, yet too low to effect electrons directly, and is absorbed in the form of molecular vibrations. Thus ε' decreases with increasing frequency. This type of behavior at low and high frequencies comes under interfacial polarization.

d) Temperature and frequency dependence of the dielectric loss

Figure 6 shows the temperature dependence of dielectric loss (ε'') at different frequencies for the studied composition. It is clear from this figure that ε'' increases with increasing temperature. As the temperature increases, σ_{ac}(ω) increases and so the ac conduction loss increases with increasing temperature. At low temperature values, conduction, dipole and vibration losses have the minimum value. However at higher temperatures, As the temperature increases, the hopping process increases and since the conduction is due to hopping from one localized states to the another, thus the conduction increases and so the losses due to conduction also increases. Additionally, dipole and vibration losses also increase. Thus dielectric loss increases with temperature. Figure 7 shows the frequency dependence of ε'' at different temperatures for the studied compositions.

It is clear from the figure that ε'' decreases with frequency. This decrease can be attributed to the fact that the migration of the ions in the glass is the main source of the dielectric loss at low frequencies. Accordingly, the dielectric loss at low and moderate frequencies is characterized by high values of ε'' due to the contribution of ion jump and conduction loss of ion migration, in addition to the ion polarisation loss. However at higher frequencies the ion vibrations may be the only source of dielectric loss because in this case charge carriers vibrate an appreciable distance through a dielectric but which become trapped or localized and thus decreasing vacancies or defects. So ε'' decreases at higher values of frequencies. Our results also confirm the same.

e) Impurity dependence of the dielectric parameters

The density of defect states decreases with Cd and Ge incorporation. The lone pairs and the other configuration, which the system might have undergone at the time of quenching, could be responsible for the decrease in the density of localized states. The variation can be explained in terms of charged impurity in the structure of Se-Te bonds. According to Scottmiller et. al. about 40% of Se atoms have a ring structure and 60% atoms are bonded as polymeric chains. An incorporation of Te decrease the Se ring concentration favoring Se-Te polymeric chains and mixed rings [25-26].On addition of Cd and Ge into Se-Te systems, might chains have decreased and rings get increased which decreases the defect states (D+, D-) in the Se-Te rings and chains. The effect of impurities on the dielectric parameters could also be correlated with covalent character of the studied compositions. The degree of covalency of the studied compositions can be estimated according to the following relation [27]:

The proportion of covalent character

$$= 100\% \exp [-0.25 (\chi_A - \chi_B)^2] \qquad (7)$$

where χ_A and χ_B are the electronegativities of atoms A and B respectively. The values of covalent characters are listed in Table 4. The incorporation of Cd and Ge in the pure binary $Se_{70}Te_{30}$ glassy alloy leads to decrease the value of the dielectric constant as given in Table 5. This decrease can be understood in terms of the nature of the bonding in the system.

It may be assumed that the incorporation of Cd and Ge in the binary alloy leads to increase the density of stronger bonds Cd-Cd, Ge-Ge and Cd-Te than other bonds in the network structure, i.e, decreases the weaker bond density Se-Cd and Se-Ge in the investigated compositions, which are more responsive to electric field than the stronger bonds. Thus the value of ε' decreases with Cd and Ge incorporation in the studied system. The decrease in the dielectric loss may be supposed due to the decrease in the density of defect states when the third element Cd and Ge as an impurity is incorporated in pure binary $Se_{70}Te_{30}$ glassy alloy. Our calculated data also supports the same.

IV. Conclusions

The temperature and frequency dependence of the ac conductivity and dielectric parameters in various glassy systems in the frequency range (1 KHz-5 MHz) and temperature range (300 K-350 K) have been measured. The ac conductivity seems to be both temperature and frequency dependent through the studied range of temperature and frequency. The conduction mechanism was suggested to be hopping conduction. In addition, the value of the frequency exponent s and its temperature dependence confirmed the applicability of the CBH model to the investigated compositions.

It has been also found that ε' and the ε'' both are highly dependent on frequency and temperature and found to be dependent with the nature of impurity incorporated in Se-Te glassy system. For the frequency and temperature dependence of ε' were attributed to interfacial and orientational polarisation, respectively whereas the temperature and frequency dependence of ε'' is associated with the conduction loss. The decrease in dielectric loss with the impurity is being correlated with the nature of covalent character of the studied composition and with the decrease in density of defect states.

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Table 1 : Calculated values of σ_{ac} (ω) and s for various glassy alloys at 1 KHz and 350K

Glassy Alloys	QMT σ_{ac} (ω)	QMT s	CBH σ_{ac} (ω)	CBH s
$Se_{70}Te_{30}$	1.02×10^{-10}	0.81	8.22×10^{-6}	0.53
$Se_{70}Te_{20}Cd_{10}$	1.02×10^{-10}	0.81	6.08×10^{-6}	0.22
$Se_{70}Te_{20}Ge_{10}$	1.02×10^{-10}	0.81	4.69×10^{-6}	0.23

Table 2 : Values of ΔE_{ac} (eV) for various glassy alloys at different frequencies and at 300K

Frequency KHz	$Se_{70}Te_{30}$ ΔE_{ac} (eV)	$Se_{70}Te_{20}Cd_{10}$ ΔE_{ac} (eV)	$Se_{70}Te_{20}Ge_{10}$ ΔE_{ac} (eV)
1	0.450	0.446	0.447
100	0.431	0.425	0.432
500	0.421	0.421	0.417
1000	0.403	0.411	0.413

Table 3 : Values of ac parameters at room temperature for various glassy alloys

Glassy Alloys	$\sigma_{ac}(\omega)(\Omega^{-1}m^{-1})$ (1 KHz, 300K)	N (E_F) (m^{-3})	$\sigma_{o(ac)}(\Omega^{-1}\ m^{-1})$ (1KHz, 300K)	ΔE_{ac} (ω) (1 KHz,300K)
$Se_{70}Te_{30}$	19.45×10^{-7}	1.23E24	79.51	0.448
$Se_{70}Te_{20}Cd_{10}$	12.78×10^{-7}	9.97E23	37.03	0.446
$Se_{70}Te_{20}Ge_{10}$	10.16×10^{-7}	8.89E23	31.70	0.447

Table 4 : Calculated covalent character of bonds for considered compositions

Bonds for bond type	% covalent character
Se-Se	100.00
Se-Te	97.78
Se-Cd	88.47
Se-Ge	91.39
Te-Te	100.00
Te-Cd	96.08
Te-Ge	79.85
Cd-Cd	100.00
Cd-Ge	99.75
Ge-Ge	100.00

Table 5 : Values of dielectric parameters for various glassy alloys

Glassy alloys	ε' (At 1 KHz, 300 K)	ε'' (At 1 KHz, 300 K)
$Se_{70}Te_{30}$	33.34	34.67
$Se_{70}Te_{20}Cd_{10}$	26.20	23.06
$Se_{70}Te_{20}Ge_{10}$	20.00	18.20

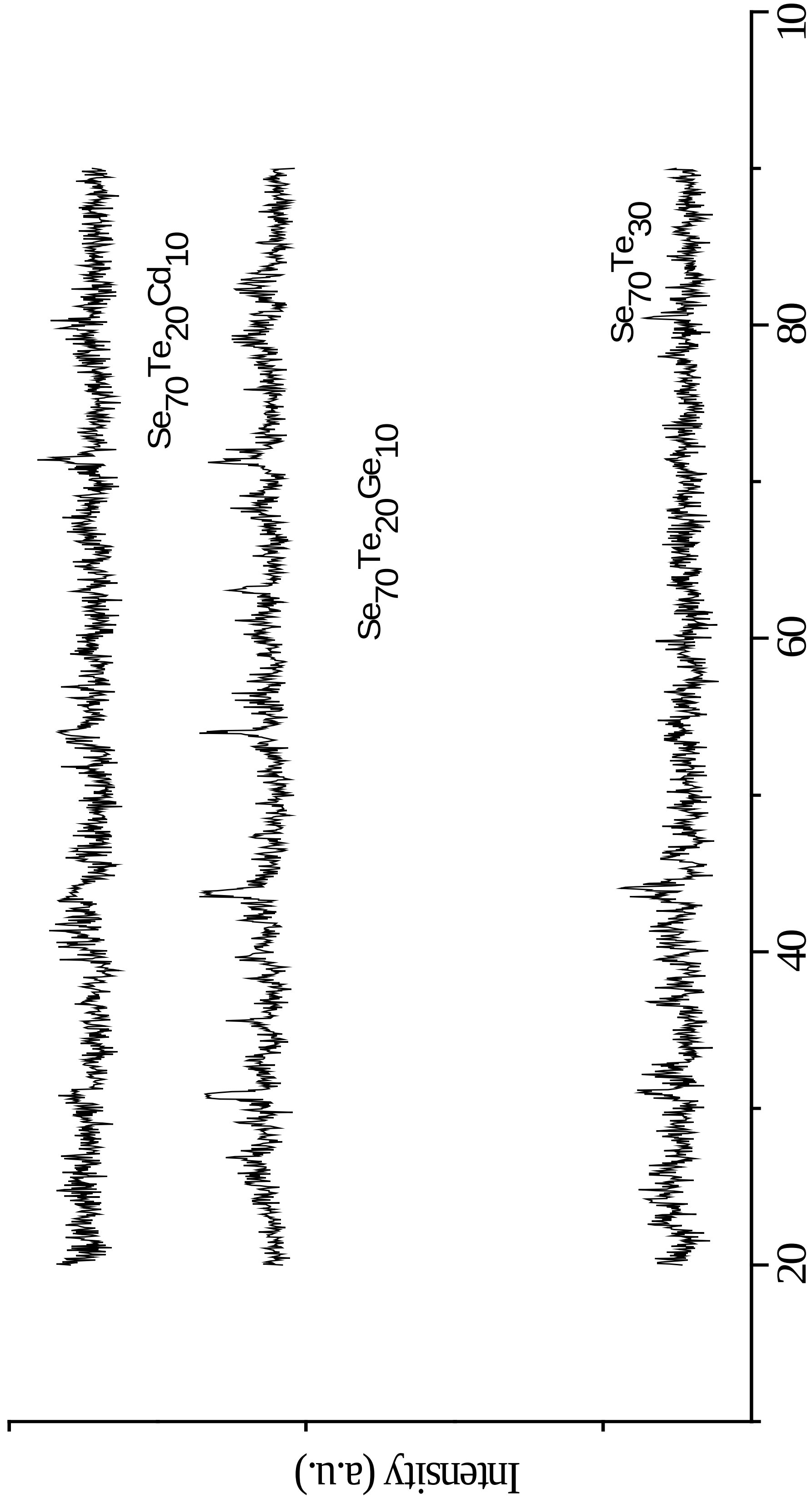

Fig. 1: XRD plots of some binary and ternary glassy alloys

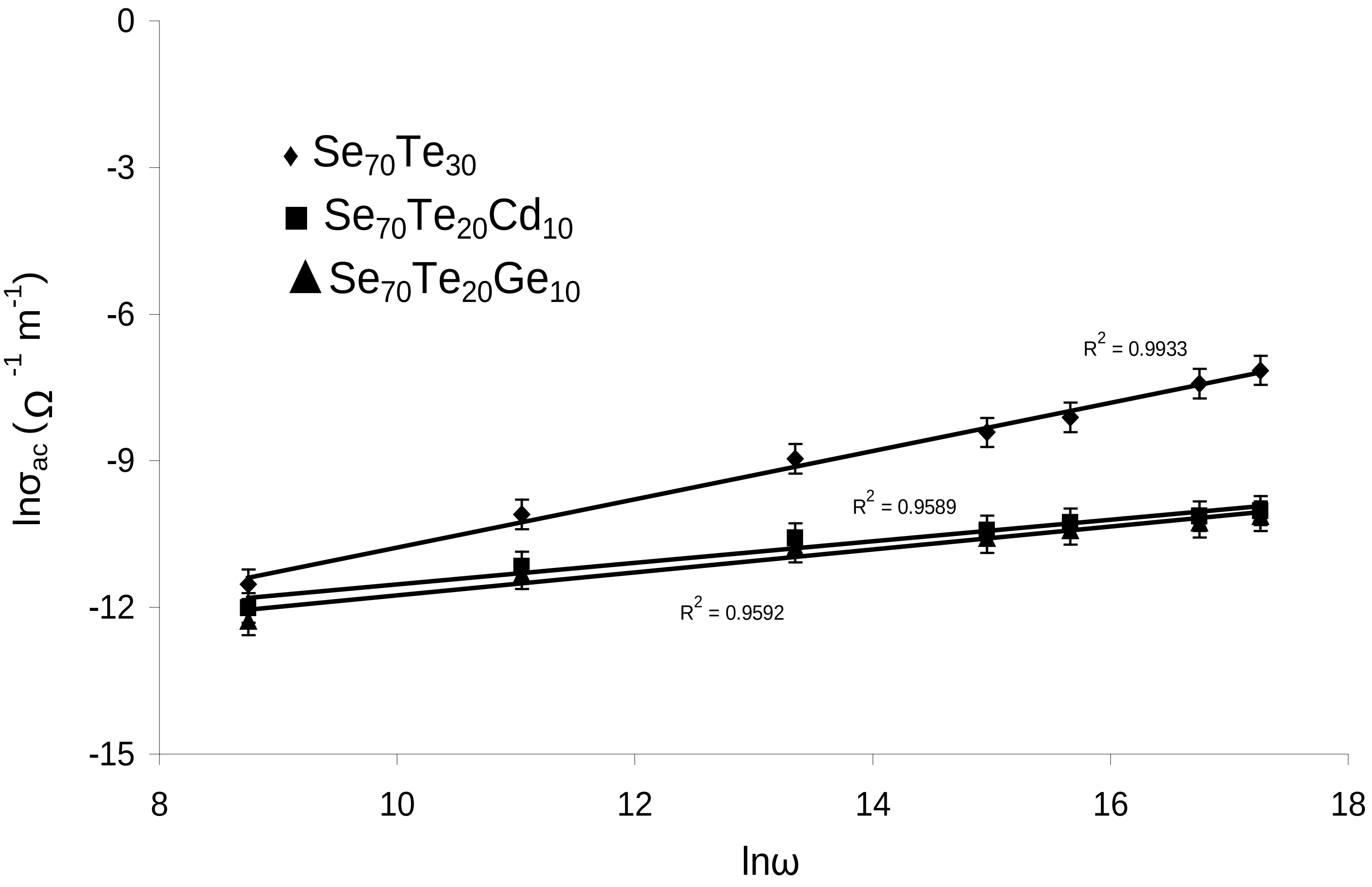

Fig. 2: Frequency dependence of ac conductivity in $Se_{70}Te_{30}$, $Se_{70}Te_{20}Cd_{10}$ and $Se_{70}Te_{20}Ge_{10}$ glassy alloys at 350 K

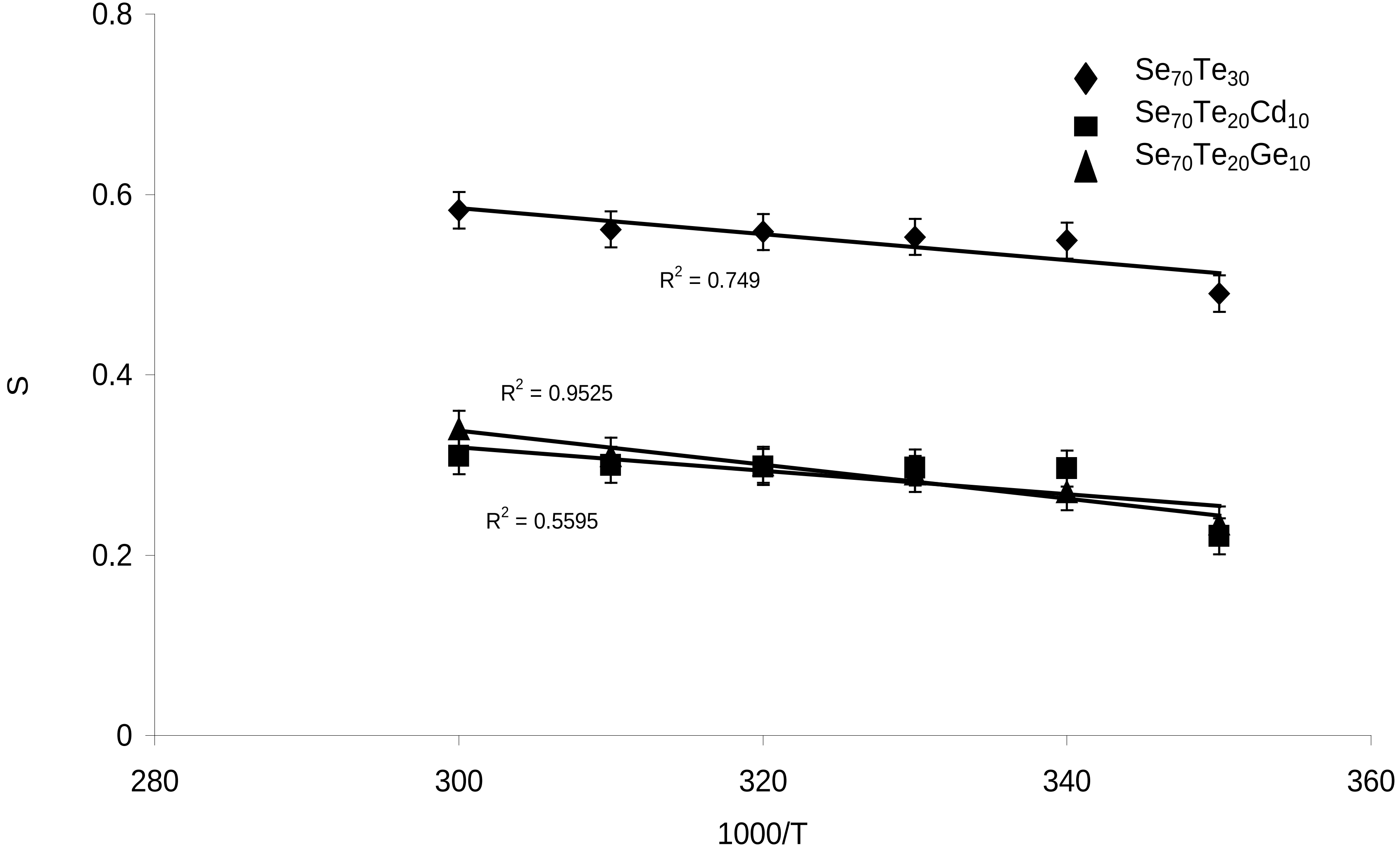

Fig. 3: S vs T curve in $Se_{70}Te_{30}$, $Se_{70}Te_{20}Cd_{10}$ and $Se_{70}Te_{20}Ge_{10}$ glassy alloys.

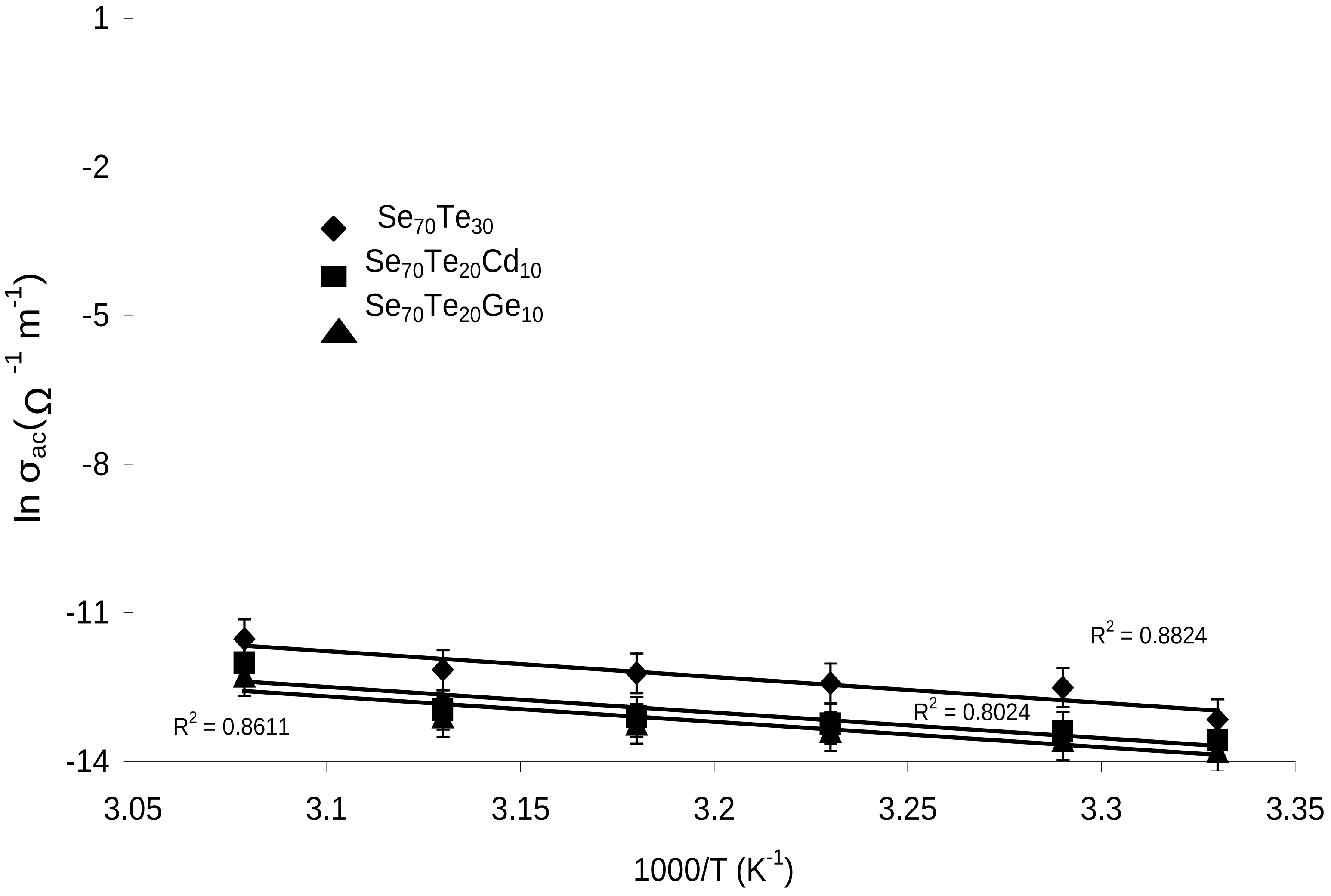

Fig. 4: Temperature dependence of ac conductivity in $Se_{70}Te_{30}$, $Se_{70}Te_{20}Cd_{10}$ and $Se_{70}Te_{20}Ge_{10}$ glassy alloys at 1 KHz.

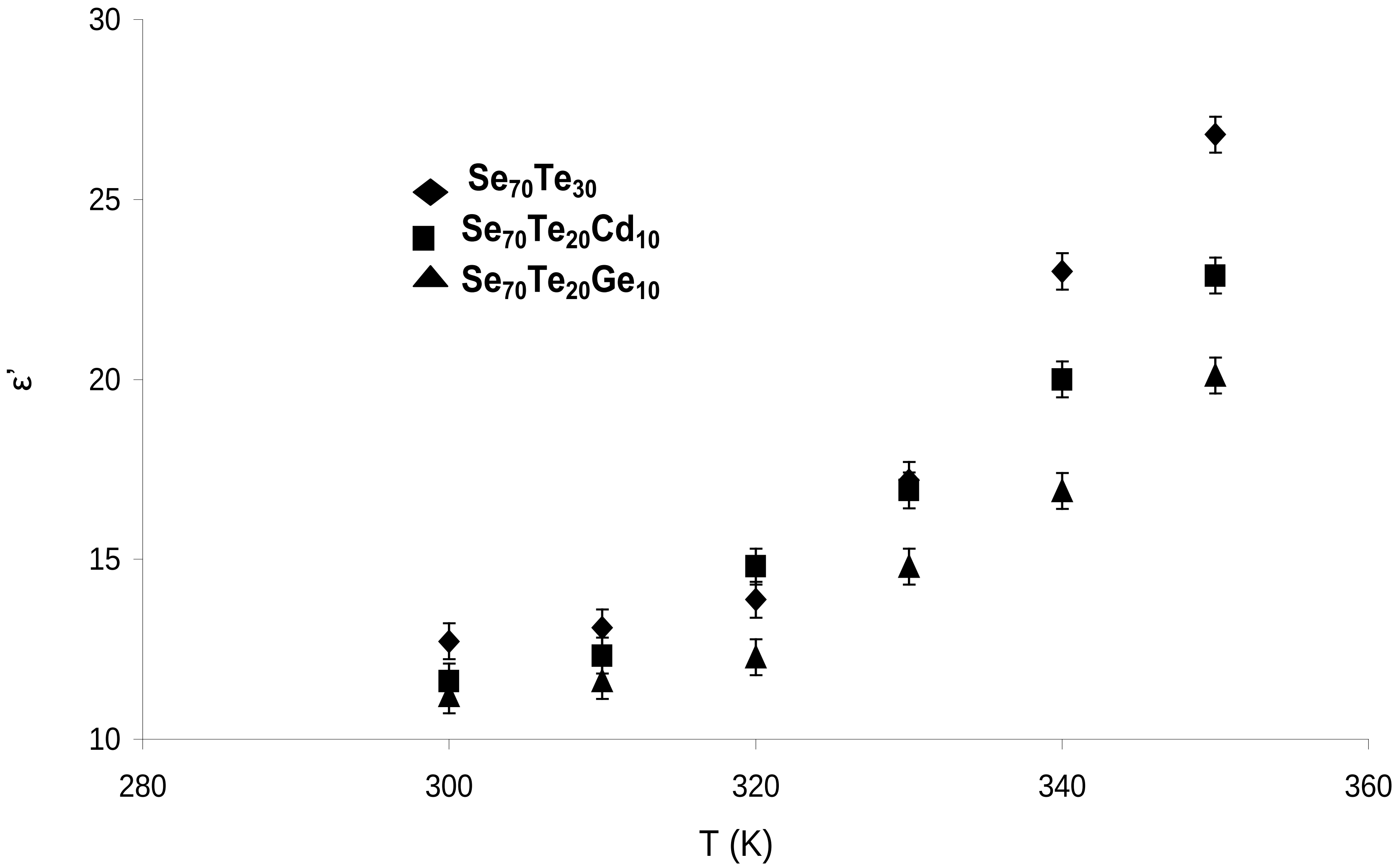

Fig. 5: Temperature dependence of dielectric constant (ε') in $Se_{70}Te_{30}$, $Se_{70}Te_{20}Cd_{10}$ and $Se_{70}Te_{20}Ge_{10}$ glassy alloys at 5 MHz.

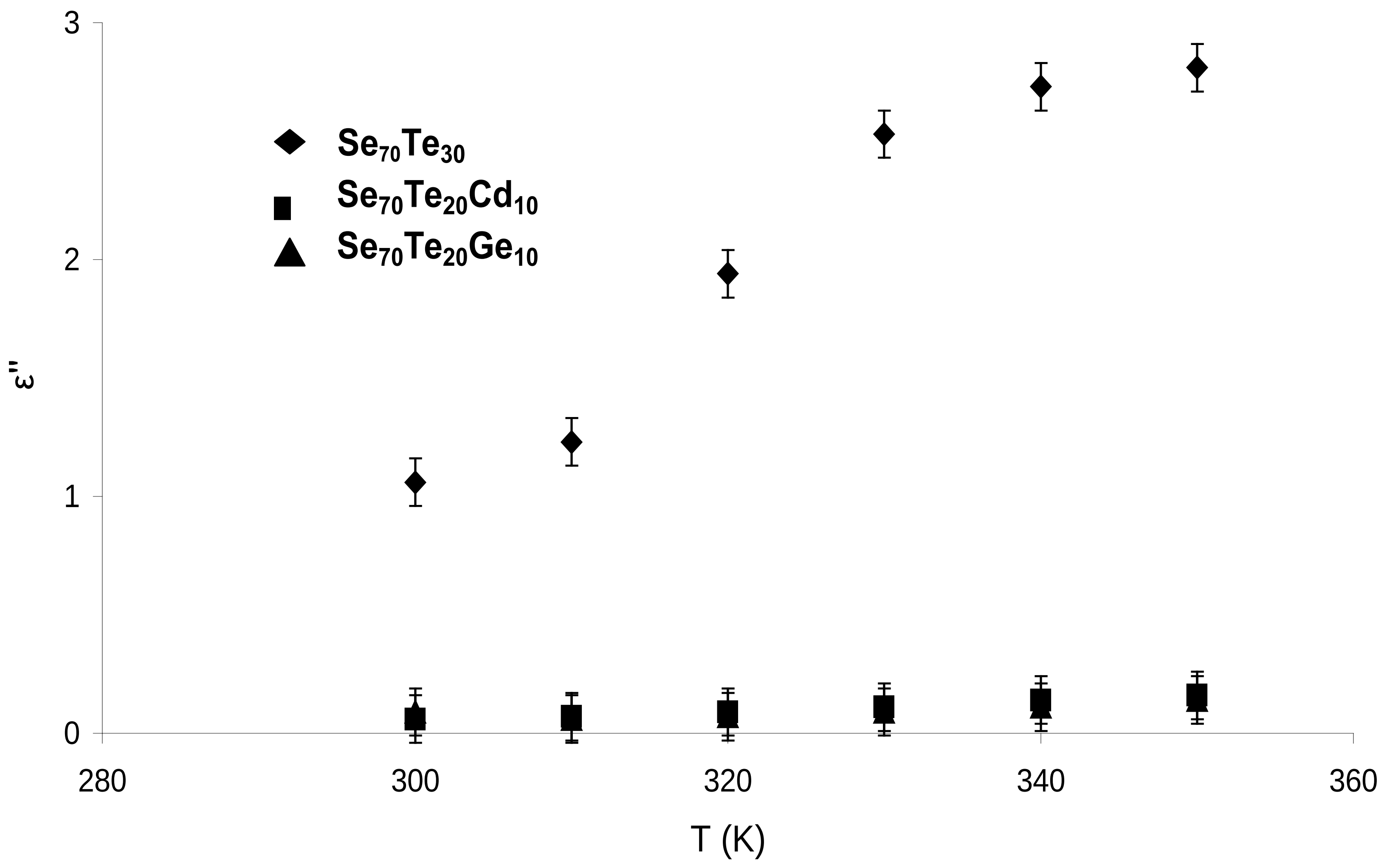

Fig. 6: Temperature dependence of dielectric loss (ε'') in $Se_{70}Te_{30}$, $Se_{70}Te_{20}Cd_{10}$ and $Se_{70}Te_{20}Ge_{10}$ glassy alloys at 5 MHz.

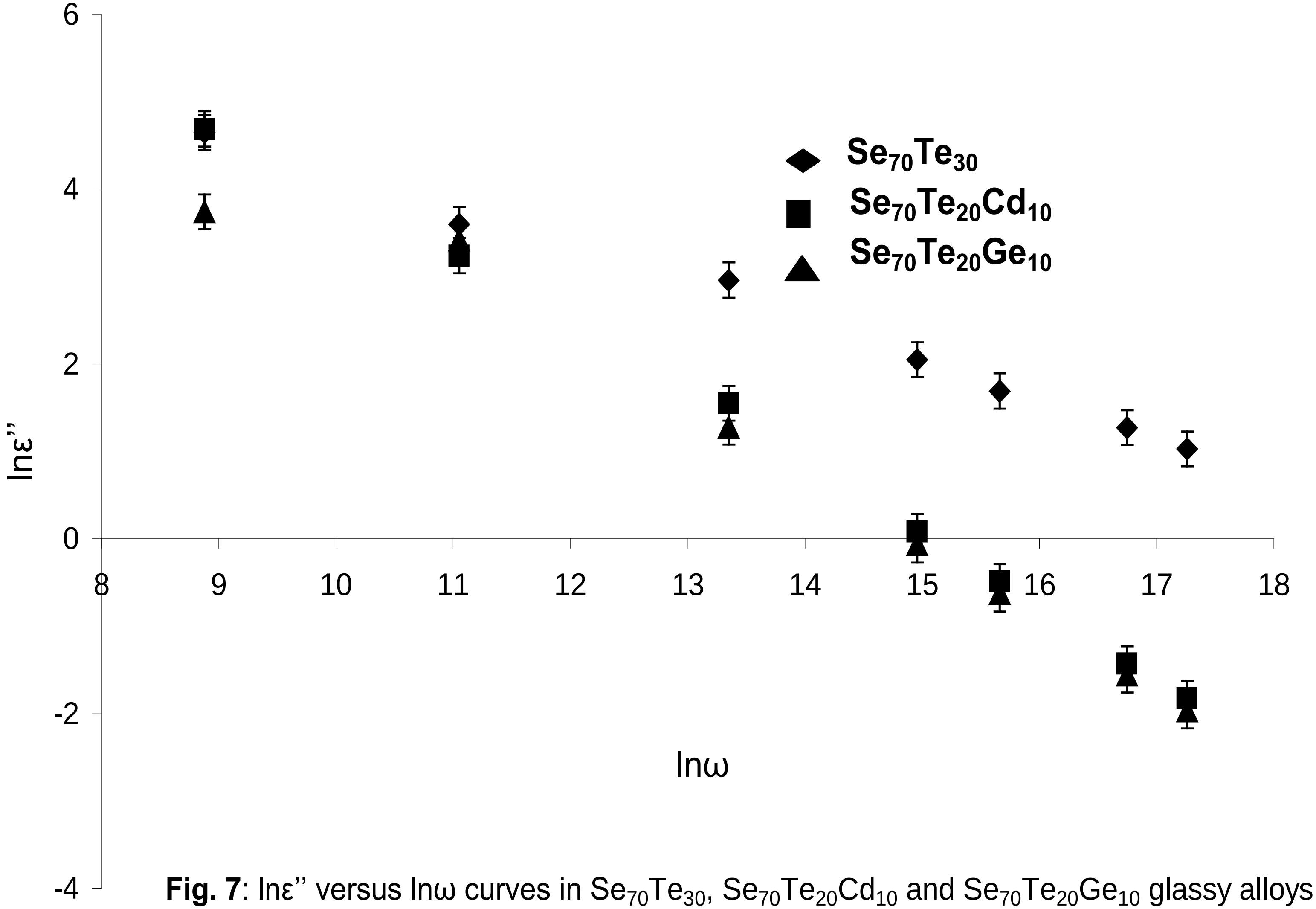

Fig. 7: lnε'' versus lnω curves in $Se_{70}Te_{30}$, $Se_{70}Te_{20}Cd_{10}$ and $Se_{70}Te_{20}Ge_{10}$ glassy alloys at 350K.