Electrical Conductivity and Dielectric Properties of Se_{100-x}Te_x Alloy

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Abstract

In this study we have investigate the temperature dependent dc conductivity and dielectric properties and of Se_{100-x}Te_x alloy (x=4, 8, 12, 16). The frequency and temperature dependence of dielectric constant ε' and dielectric loss ε'' is studied in the frequency range of 1 KHz-1 MHz, and in the temperature range 290-360K. The dielectric constant ε' and dielectric loss ε'' show a decrease with increasing frequency and an increase with increasing temperature. All sample show the dielectric dispersion, due to the distribution of the relaxation time induced by polarized species. The activation energy is also evaluated from Arrhenius plot of Dc conductivity, which is due to the tunneling effect of trapped electron "hopping" from one trap site to another.

Keywords dielectric constant, dielectric loss, Dc conduction loss, Dc conductivity, Activation energy

Introduction

Chalcogenide glasses are novel materials have attracted researcher because of their potential use and extensive application in solid state devices, such as ultra high density phase change storage and memory, photo voltaic, thermal imaging, switching memory, photo detectors, photo receivers etc [1-7]. Low phonon energy makes these glasses transparent to I-R region.

Se based alloy show high transparency in mid I-R region, non-linear properties and also have a variety of photo-induced effect, which are useful for all- optical switching [8-9].

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Eminent interest has been shown in a Se-Te alloy due to higher photo sensitivity, greater hardness, higher crystallization temperature and small aging affects as compared to pure a- Se.

The Dc conductivity of amorphous semiconductor has attracted great deal of attention to understand the conduction process in these materials. Thus the study of electrical behavior of these materials is extremely important. Dielectric materials have been the subject of renewed attention. These materials application have been widely employed in various industrial devices such as dynamic access memory, microwave filters, and voltage controlled oscillator and tele-communication technologies [10-13]. The dielectric parameters are the crucial quantities in the design of devices. The study of dielectric properties of chalcogenide glasses may be useful in the determination of the structure and defect in solid, which infect can be useful for the determination of conduction mechanism in these materials. The dielectric study of chalcogenide glasses have revealed that dielectric dispersion does exist at low temperature. The dielectric dispersion is characterized by the distribution of the relaxation time induced by polarization species. In addition, the study of temperature and frequency dependence of dielectric parameter in the range of low frequency where the dielectric dispersion occurs can be important for the understanding losses happening in these materials.

In the present work, we have studied the temperature and frequency dependence of dielectric constant and dielectric losses. The Dc conductivity has also been measure to confirm the Dc conduction losses.

Experiment

Glassy alloys of $Se_{100-x}Te_x$ (x = 4, 8, 12, 16) have been prepared by meltquenching technique. High purities (99.99%) materials have been weighed according to their atomic percentage and sealed in quartz ampoules (length 5 cm and internal diameter 8 mm) under a vacuum 10⁻⁵ Torr. The sealed ampoules containing the constituent materials have been heated up to 900 K at a rate of 4 K/min and held up to 10 hours in the furnace. All the ampoules were frequently rocked while heating to obtain homogeneous glassy alloys. The ampoules have been quenched in ice-cool water to obtained glassy nature.

The quenched samples have been removed by breaking the quartz ampoules. The amorphous nature of the film has been ascertained by X-ray diffraction. The obtained melt are grinded into powder form, to obtain homogeneous alloy, which is then compressed into pellet (diameter ~ 10 mm) under a pressure of 5 tons. To obtain a good electrical contact pallet are coated on both side with silver paste. The bulk sample is then mounted between two steel electrodes inside a metallic sample holder for dielectric measurement. A vacuum of 10^{-3} Torr is maintained to avoid any moisture effect. To measure the Dc conductivity of bulk sample, a dc voltage of 1.5 V are applied across the sample and the resulting current measure by Keithley electrometer (6514). To calculate the dielectric constant ε' and dielectric loss ε'' the parallel capacitance and dissipation factor are measured by using LCZ meter (WAYNE KERR 4300).

Result and Discussion

Dc Conductivity

The conduction mechanisms in dielectric materials are crucial to explain the application of dielectric. There are two types of conduction mechanism in dielectric film electrode limited conduction mechanism and bulk-limited conduction mechanism. Based on the bulk limited conduction mechanisms, some important electrical properties in the dielectric materials can be extracted, including trap spacing, trap energy level, trap density, electronic mobility, density of state (DOS) etc. The Temperature dependent dc conductivity (σ_{dc}) of Se_{100-x}Te_x in the temperature range of (290-360 K) has been measured. The plot of ln (σ_{dc}) vs. 1000/T shown in figure 1, represent a straight line which indicate that the conduction is through an activated process with single activation energy [14-15] and hence, dc conductivity can be represented by well known relation

$$(\sigma_{dc}) = \sigma_0 \exp\left[-\frac{\Delta E}{KT}\right] \tag{1}$$

Where, ΔE the activation energy k is the Boltzmann constant, σ_0 is the pre exponential factor.

The calculated values of activation energy (ΔE) and pre-exponential factor (σ_0) in the temperature range of (290-360K) are given in table 1, which suggest that the conduction is due to thermally assisted tunneling of charge carriers in the localized state present in the band tails.

Activation energy (ΔE) decreases with increases of Te concentration, this can be attributed due to the shifting of the Fermi level. The value pre-exponential factor σ_0 as suggested by Mott [16] for the conduction in localized state should be about 10³. The value of pre-exponential factor σ_0 calculated for all composition suggest that conduction mechanism is via localized state

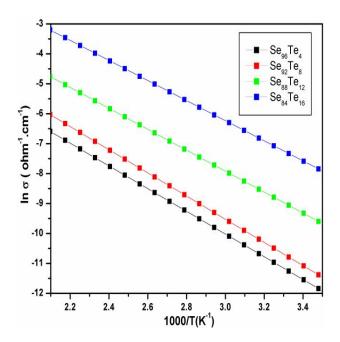


Fig 1: In (σ_{dc}) vs. 1000/T for Amorphous alloy of Se_{100-x}Te_x

Dielectric Properties

The temperature dependent of dielectric constant ε' and dielectric loss ε'' is appreciable only in certain range of temperature. The real part of dielectric constant ε' determine the maximum energy that can be store in the material.

The loss factor ε'' measure the absorption of electrical energy by the dielectric materials that is subjected to an electromagnetic field. Furthermore, the dissipation factors $\tan \delta = \varepsilon''/_{\varepsilon'}$ determine how a material cans absorb electromagnetic field [17-19]. At low temperature both ε' and ε'' are constant with temperature and increase appreciable with increase of temperature.

The temperature dependence of dielectric constant ε' and dielectric loss ε'' are studied in the frequency range (1 KHz-1 MHz) and temperature range (290-360K). Fig 2 shows that the dielectric constant ε' and dielectric loss ε'' of Se₉₆Te₄, it increases with increases of temperature and is different for different frequencies.

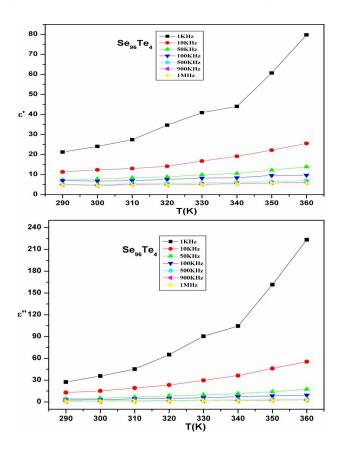


Fig 2: Dielectric Constant ε' and Dielectric loss ε'' Versus Temperature at Fixed Frequency of Se₉₆Te₄ Sample

Fig 3 shows frequency response of dielectric constant ε' and dielectric loss ε'' of Se₉₆Te₄ at fixed temperature. On increasing the frequency the dielectric constant and dielectric loss decreases.

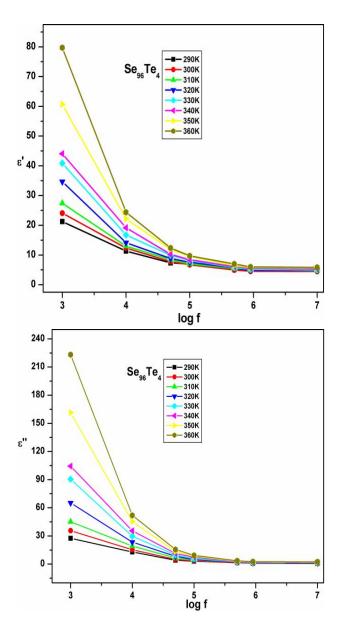


Fig 3 Dielectric Constant ε' and Dielectric Loss ε'' Versus Frequency at Fixed Temperature of Se₉₆Te₄ Sample

The temperature and frequency dependence of dielectric constant ε' for amorphous alloy Se_{100-x}Te_x (x=4, 8, 12, 16) at a fixed frequency (10 KHz) and fixed temperature 340K respectively is shown in fig 4(a) and (b). These figs indicate that dielectric constant ε' increases with increase of temperature, the increase being different at different frequencies. When the temperature increases the orientational polarization is fascinated and this increase the dielectric constant ε' . At low frequency dipole aligned themselves along the direction of the field and fully contributes to the total polarization (electronic, ionic, orientation and interfacial polarizibility) [20]. When the frequency is increased the variation of the field becomes too rapid for the molecular dipole to follow, so that their contribution to the polarization becomes less and hence the dielectric constant ε' decreases.

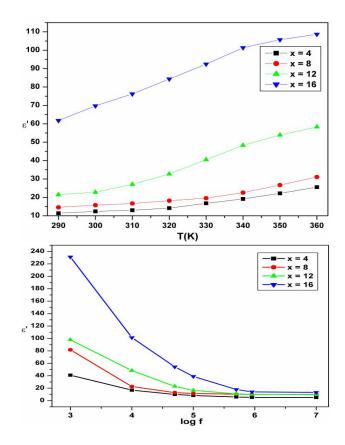


Fig 4: Temperature and Frequency Dependence of Dielectric Constant ε' for Amorphous Alloy Se_{100-x}Te_x at fix Frequency10 KHz and Temperature 340K

Similarly the temperature and frequency dependence of dielectric loss ε'' for all composition at (10 KHz) and temperature 340K respectively is shown in fig 5(a) and (b). It is clear from the fig that dielectric loss ε'' increases with temperature. The dielectric loss consists of two contribution one from the dielectric polarization processes, and other from Dc conduction [21]. Loss factor due to conduction increases with increasing temperature.

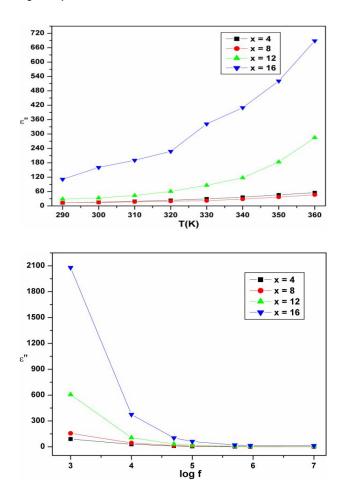


Fig 5: Temperature and Frequency Dependence of Dielectric loss ε'' for Amorphous Alloy Se_{100-x}Te_x at Fix Frequency 10 KHz and Temperature 340K.

Composition dependence of dielectric constant ε' and dielectric loss ε'' at a fixed frequency (10 KHz) and at a temperature (340 K) is shown in fig 6, from fig it is clear that dielectric constant ε' and dielectric loss ε'' both increases with Te concentration. This type of behavior has been shown by various researchers in chalcogenide glasses [22-27].

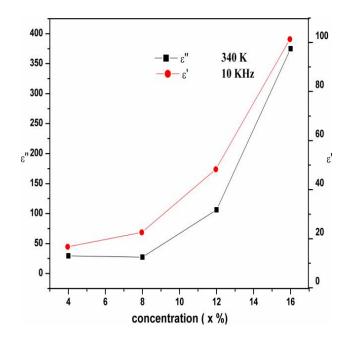


Fig 6: Dielectric Constant ε' and Dielectric loss ε'' Versus Concentration of Se_{100-x}Te_x at Fixed Frequency (10 KHz) and Temperature (340K)

So the concentration of Te play important role in increasing dielectric constant ε' and dielectric loss ε'' , this is due to the increase in defect states, these values of dielectric constant ε' and dielectric loss ε'' is shown in table 1. The $\ln \varepsilon'$ vs 1/T plot is found to be straight line in the entire sample, (result not shown) this type of temperature dependence is generally observed in molecular solids where Debye theory [28] for viscosity dependence of relaxation time hold good. This has found to be true with our sample. The above discussion emphasis that dipolar type of dispersion has been occurred in our sample. However no peak has observed in dielectric loss verses log f curve as expected in case of dipolar –type of relaxation.

Guintini et al. [29] has proposed a dipolar model for dielectric dispersion in amorphous semiconductor. This model is based on Elliot's [30] idea of hopping of charge carrier over a potential barrier between charge defect states D^+ and D^- , each pair of D^+ and D^- is assumed to form a dipole with relaxation energy between which a carrier must hop. According Guintini et al [29] at a particular frequency in the temperature range where dielectric dispersion occurs is given by

$$\varepsilon''(\omega) = (\varepsilon_0 - \varepsilon^{\infty}) 2\pi^2 N (ne^2/\varepsilon_0)^3 k T \tau_0^m W_m^{-4} \omega^m$$
(2)

Here, m is the power of angular frequency and is negative in this case which is given by

$$m = -4\pi kT / W_m \tag{3}$$

Where n is the number of electrons that hop, N is the concentration of localized sites, ε_0 and ε^{∞} are the static and optical dielectric constants, respectively, W_m is the energy required to move the electron from a site to infinity.

From (2) ε'' should obey the following relation $\varepsilon'' = B\omega^m$ Where B is constant and m is angular frequency power. The plot of $\ln \varepsilon''$ vs $\ln \omega$ should be a straight line and the slope of these lines tells us the value of m, and should be negative which is shown in Fig 7. The variation of m with temperature is shown in inset in the figure. The calculated value of m and W_m and is given in table 1 for all the sample, this indicated that the paired defect states behave as a dipole. The present result is in good agreement with the theory of hopping of charge carriers over a potential barrier as suggested by Elliott [30] in the case of chalcogenide glasses.

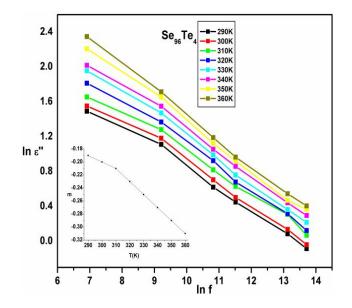


Fig 7: The Plot of $\ln \epsilon'' vs \ln \omega$ for Se₉₆Te₄Sample at different Temperature and the Inset Figure Shows the Temperature Dependence Value of m

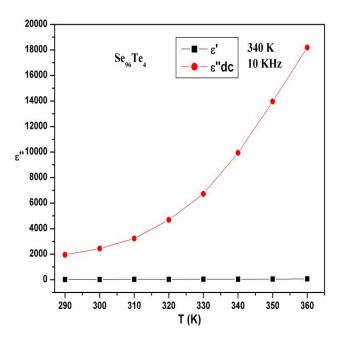
The Dc conduction loss is also an important parameter to understand the dielectric dispersion in glasses. We have calculated DC conduction loss using following relation [31-33].

$$\varepsilon''_{dc} = \frac{\sigma_{dc}}{\omega} \in_{0}$$
(4)
$$\sigma_{dc} \text{ is the DC conductivity.}$$

The calculated value of DC conduction loss is shown in table 1. It is clear from the table that Dc conduction loss is dominated over observed loss, and DC conduction loss increases with increases of Te concentration. Fig 8 show the variation of observed dielectric loss and calculated Dc conduction loss for $Se_{96}Te_4$ with temperature at fixed frequency Thus it is clear that the observed dielectric dispersion is mainly due to the DC conduction losses in the present sample.

Table 1 Electrical and Dielectric Parameter in $Se_{100-x}Te_x$ at T = 340 K and f = 10 KHz

•	$\sigma_{dc}(340K)$	ε	ε″	$\varepsilon^{''}_{\ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ \ $	ΔE	σ_0	m
<i>W_m(eV)</i> Se ₉₆ Te ₄	7.75×10 ⁻⁵	16.76	29.67	13.94×10 ³	0.33	4.29	
-0.27 1.36	1.10×10	10.70	27.07	13.74×10	0.00	7.27	
72 0	1.12×10 ⁻⁴	22.56	27.53	20.15×10^{3}	0.32	4.57	-
0.56 0.65 Se ₈₈ Te ₁₂	5.19×10 ⁻⁴	48.29	106.2	93.38×10 ³	0.29	8.24	
-0.59 0.62	5.17×10	40.27	100.2	73.30×10	0.29	0.24	
$\mathrm{Se}_{\mathrm{84}}\mathrm{Te}_{\mathrm{16}}$	2.71×10 ⁻³	101.3	375	487.6×10 ³	0.27	3.003	-
<u>0.73 0.50</u>							





Conclusion

Temperature and frequency dependence of dielectric constant and dielectric loss are studied in $Se_{100-x}Te_x$ glassy system.

Dielectric dispersion is found to occur in these glasses, which is due to Dc conduction losses, which dominate over other losses. The dielectric constant and dielectric loss increases with increase of Te concentration, which may be due to the increase in the defect state. The increase in the capacitance and hence the dielectric constant is maximum toward the low frequency region. The influence of temperature on Dc conductivity may be explained by increasing the mobility of charge carrier that increases the conductivity. On increasing the concentration of Te, Dc conductivity increases and activation energy decreases are due to increases in the density of defect states or due to decrease of the disorder in the mobility edge.

References

- Onozuka A, Oda O (1988) Electrophotographic properties of SeTeSb Halogen alloy. J Non-Cryst Solids 103: 289.
- Elliot S R (1991) Physics of Amorphous Materials, Longman Publication, London
- Fritzsche H, (2007) Why are chalcogenide glasses the materials of choice for Ovonic switching devices, J Phy Chem Solids 68: 878.
- Pattanayak P, Asokan S (2006) Anomalous electrical switching behaviour in phase-separated bulk Ge–Se–Ag chalcogenide glasses, Europhys Lett 75: 778.
- Hendrik F. Hamann, Martin O Boyle, Yves C, Martin, Michael Rooks, H Kumar Wickramasinghe, (2006) Ultra-high-density phase-change storage and memory, Nat Mater 5: 383.
- Balitska V, Shpotyuk O, Altenburg H, (2006) Bimolecular relaxation kinetics observed in radiation-optical properties of Ge–As(Sb)–S glasses, J Non-Cryst Solids 352: 4809.
- Ganjoo A, Jain H, Yu C, Song R, Ryan J V, Irudayaraj J, Ding Y J, Pantano C G, (2006), Planar chalcogenide glass waveguides for IR evanescent wave sensors, J Non-Cryst. Solids 352: 584.
- Harbold J M & Hilday F O, Wise F W, Itkain B G, (2002) Highly nonlinear Ge-As-Se and Ge-As-S-Se glasses for all-optical switching, IEEE Photon Technol Lett, 14:822.
- Seddon A B (1995) Chalcogenide glasses: A review of their preparation, properties and applications, J Non-Cryst Solids 184: 44–50.
- Ioachim A, Toacsan M I, Banciu, et al (2007) Barium strontium titanate-based perovskite materials for microwave applications, Prog Solid State Chem 35:513.
- Ota Y, Kakimoto K I, Ohsato H, Okawa T, (2004) Low-temperature sintering of Ba_{6-3x}Sm_{8+2x}Ti₁₈O₅₄ microwave dielectric ceramics by B₂O₃ and GeO₂ addition, J. Eur Ceram Soc 24:1755.
- Chen Y C, Cheng P S, Yang C F, et al (2001) Substitution of CaO by BaO to improve the microwave dielectric properties of CaO-Li₂O-Sm₂O₃-TiO₂ ceramics, Ceramics Int 27:809
- Zhou H, Wang H, Chen Y, et al (2009) Low temperature sintering and microwave dielectric properties of Ba₃Ti₅Nb₆O₂₈ ceramics with BaCu(B₂O₅) additions, Mater Chem. Phys 113:1.
- Kitao M, Yoshii K, Yamada S (1985) Thermoelectric Power of Glassy As₄₀Se_{60-x}Te_x, Physica Status Solidi 91(1) 271-277.
- Gadkari A B, Zope J K, (1988) Electrical properties of the amorphous semiconducting SeTeIn system, Journal of Non-Crystalline Solids 103 (2-3) 295-299.
- Mott NF, Davis EA (1979) Electron processes in non-crystalline materials. Clarendon Press, Oxford, p 660
- Barsoum M (1997). Fundamentals of Ceramics, McGraw Hill Series in, Materials science and Engineering, New York.
- Atyia H E, (2007) Deposition temperature effect on the electric and dielectric properties of InSbSe₃ thin films, Vacuum 81 590-598.
- Hench L L West J K (1990) Principles of Electronic Ceramics, Wiley, New York 351-360
- Tareev B, (1975) Physics of Dielectric Materials, Moscow Mir Publishers p. 67.
- Stevels J M, (1957) Handbuch der Phycik (Ed. S. Flugge) Springer: Berlin, p-350.

- Ilyas M, Zulfequar M, Khan Z H, Husain M (1998) Dielectric properties of a-Ga_xSe_{100-x} alloys (x=0, 2.5, 5.0, 7.5 and 10.0), Physica B 254 :57.
- Srivasatava KK, Kumar A, Panwar O S, (1979) Dielectric relaxation study of chalcogenide glasses, J. Non-Cryst. Solids 33:205.
- Ibrahim M M, El-Haawny S S, Hafiz M M, Abdel-Rehim M M, (1990) Indian J Pure and Appl. Phys 28: 22.
- Arora R, Kumar A, (1989) Dielectric relaxation in glassy Se and Se_{100-x}Te_x alloys Phy. Stat Sol. (A) 115:307
- Sharma A K, Bhatia K L, (1989) Frequency-dependent electrical conductivity of bismuthmodified amorphous semiconductors (GeSe_{3.5})_{100-x}Bi_x J. Non-Cryst. Solids 109:95.
- R S Kundu, Bhatia K L, Kishore N, Singh P, (1995) Effect of addition of Zn impurities on the electronic conduction in semiconducting Se_{80-x}Te₂₀Zn_x glasses Philos. Mag. B 72 :513.
- Debye P, (1929) The Chemical Cataloge Company, Polar Molecule, New York (Chapter 5).
- Guintini J C, Zanchetta J V, Jullien D, Eholie R, Houenou P, (1981) Temperature dependence of dielectric losses in chalcogenide glasses J. Non-Cryst Solids 45: 57.
- Elliott S R, (1977) A theory of a.c. conduction in chalcogenide glasses, Philos. Mag. 36 :1291.
- Zulfequar M, Kumar A (1989) Dielectric Behavior of Hot-Pressed AIN Ceramic: Effect of CaO Additive, J Electro chem Soc 136(4): 1099.
- Zulfequar M, Kumar A (1988) Adv Ceramics Matter 3:332.
- Arora R, Kumar A (1990) Electrical conductivity and dielectric relaxation in bulk glassy Se_{80-x} Te₂₀In_x, J Mater Sci Lett 9:348.