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AC Conduction and Dielectric Characterization of Vacuum Evaporated PbSe thin Films

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Abstract: Thin films of PbSe are prepared by vacuum deposition technique on to well cleaned glass substrates. Thin film capacitors of the type (Al-PbSe-Al) have been fabricated. AC Conduction and Dielectric studies performed on samples of thickness 300,500 and 700nm at various frequencies (10kHz-10MHz) and temperatures (293K-493K).From the AC conduction studies, it is confirmed that the mechanism responsible for the conduction process is hopping. Thermal activation energy is found to decrease with increase in film thickness. The variations of the dielectric constant and loss as function of frequency at different thicknesses and temperatures are observed and the results are discussed. The polarizability α is found to decrease with the increase of thickness while it increases with temperature.

Keywords: PbSe, Vacuum deposition, Conduction studies, Thin films ; Dielectric relaxation; Complex capacitance

1. INTRODUCTION

The need for microminiaturized electronic components in the microelectronics industry has provided the greatest stimulus for the investigation of electrical conduction properties of thin films of various materials. Among them, chalcogenide films for new kind of materials with immense qualities of use in many practical applications. PbSe thin film is one such candidate having a range of applications. The applications of these materials are extended to fabricate ideal infrared radiation detectors, infrared emitters and solar control coatings [1-6].For the understanding of basic features of PbSe chalcogenide films and their potentiality in application, the knowledge of the electron states and their behavior with different frequencies and at high fields becomes important. The present paper reports the AC Conduction and Dielectric studies made on PbSe thin films.

Lead chalcogenides (PbSe, PbTe, PbS) are important materials for applications such as IR detectors, photographic plates, and photo resisters [1-3]. Among the group IV-VI compounds, Lead selenide (PbSe) thin film is used as a target material in infrared sensor, grating, lenses and various opto electronic devices [4]. It has a cubic crystal structure (higher thickness) and a direct band gap of 0.27 eV in room temperature. Dielectric relaxation phenomenon shows a significant effect on the electrical properties of the capacitors, since the leakage current behavior under electric fields used in actual device operation is attributed to the dielectric relaxation. The complex plane analysis has proven to be a useful means of characterizing the electrical nature of a number of materials [11- 14]. In this technique, measured ac data are examined in complex plane of modulus, impedance and admittance. A semicircular fit of the data in any of these planes suggests an appropriate equivalent circuit representation of the observed dispersion. This approach can reveal the presence of relaxation processes and the relative contribution of defect states to the total ac response under a given set of experimental conditions. In the present paper, a study on the vacuum evaporated PbSe thin films relating to the a response have been made to describe the type of conduction mechanism and the origin of relaxation in these types of films. XRD diffraction declares the presence of large number of peaks indicates that the films with different thickness are polycrystalline in nature with prominent peaks at $2\theta=29.1^0$ which correspond to (200) plane giving rise to the rock salt cubic structure of PbSe as indicated in the ASTM [1,7, 8]. The intensity of the preferred plane for crystal growth increases with the increase of thickness .It is found that the grain size of the film increases with film thickness and the dislocation density is found to decrease.

2. Experiment

High purity (99.999% pure) selenium and lead elements obtained from Balzars (Switzerland)were used in the present investigation. The stoichiometric mixture of the selenium and lead corresponding to the composition of PbSe,weighed to an accuracy of 10^{-4} g, was sealed in a quartz ampoule under a vacuum of 2x 10^{-5} mbar. The quartz ampoule was then heated to 1273 K and then held for 5 h, kept at the same temperature with periodic shaking and finally quenched in cold water. The resultant mass was crushed into fine powder and then evaporated from a molybdenum source at a base pressure of 2x 10^{-5} mbar.Al/PbSe/Al sandwich structures of different thicknesses were fabricated and high temperature silver paste was used for the contact. The capacitance, impedance and tan δ were measured as a function of frequency (10k Hz to10 MHz), in the temperature range of 293-493 K.

3. Results and discussion

3.1 A.C Conductivity

The temperature dependence of A.C conductivity was calculated using the relation $\sigma_{ac} = \omega\varepsilon_0\varepsilon_r \tan\delta$, where is the angular frequency, tan δ is the dissipation factor ,ε_r the relative dielectric constant and ε_0 is the permittivity of the free space. The frequency dependence of conductivity is shown in Fig.1 for films of different thicknesses. It is observed that the total conductivity for lower thickness (300,500nm) is frequency independent in the range 1 MHz, while it is frequency dependent for frequency >1MHz but it is frequency dependent for residual thickness, this indicted that ac conductivity become strongly frequency dependent for high thickness as power of $\sigma_{ac} \alpha f^n$ where n is a function of temperature as well as of frequency. It is observed that n varies between 0.057and 0.53 as the film thickness varied from 300 to 700 nm, while n change in non systematic manner with temperature, i.e. increases but then it showed a decreasing trend with further increase of temperature , moreover for t=300nm n increases from 0.057 to 0.66when temperature increase from 293to 443 K, while n decreases from 0.66 to 0.63 with further increase of temperature , this is in good agreement with earlier investigators [15,16] on semiconducting thin films .The power law dependence of ac conductivity is of a universal nature and corresponds to short range hopping of charge carriers through trap sites separated by energy barriers of varied heights. Each pair of potential well corresponds to a certain time constant of transition from one site to another. Naturally, the time constant for transition across a higher energy barrier is large compared to that across the smaller ones. Therefore, the traps with high activation energy can respond only at low frequencies. The dispersion in conductivity at low frequency can be explained as due to non-adiabatic hopping of charge carriers between impurity sites. If hopping takes place between a random distributions of localized charge states. The lower value of s occurs for multiple hops, while the higher value occurs for single hops. Hence, in the present investigations, Pollack’s [17] theory holds good at high frequency and at higher temperatures. In general, the values of ac conduction is found to be easily understood from the power laws:

$$\sigma_{ac} = A(T)\omega^n + \sigma_0(T)$$

where $_0$(T) and () corresponds to the D.C and A.C conductivity, respectively.The plot of ln conductivity against the inverse absolute temperature for a PbSe film of different thicknesses is shown in Fig. 2. One region are observed for all thicknesses and frequencies. The observed frequency dependence of conductivity suggests the general feature of the hopping systems. Also, we can anticipate that the numbers of traps having lower energies are more than those having higher energy values and hence, the ac conductivity at higher frequency is expected to be more. Also the polycrystalline nature of the film and the other crystallographic defects may provide additional acceptor centers [18].The activation energy is found to decrease with increasing of thickness and frequency which is given in Table2. Indeed the activation energies are found to decreases from (0.0386- 0.0297eV , 0.204- 0.0087eV and 0.0146 - 0.0066 eV for thicknesses 300, 500 and 700nm and frequencies 10kHz, 100kHz and1MHz ,which is clearly consistent with a hopping mechanism[19,20] This can be possible due to the increase of grain size and applied field frequency respectively which enhance the electronic jumps between the localized states .

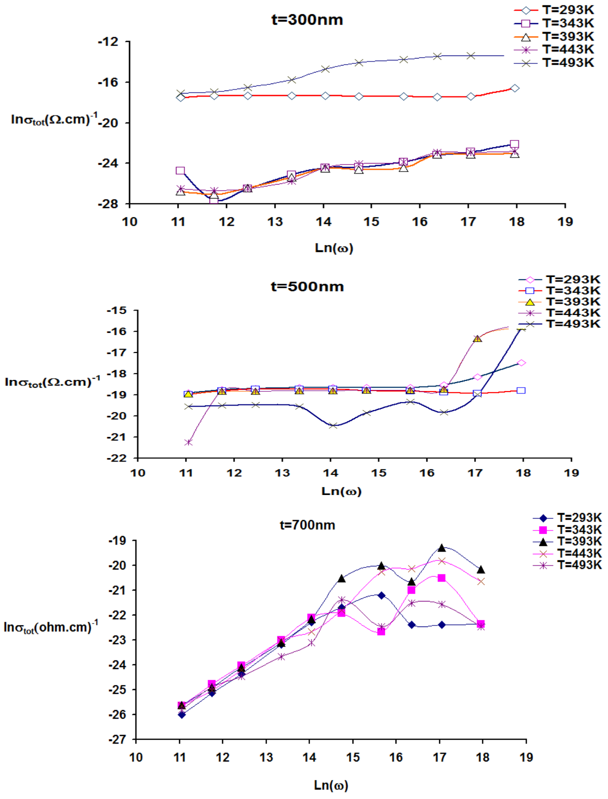
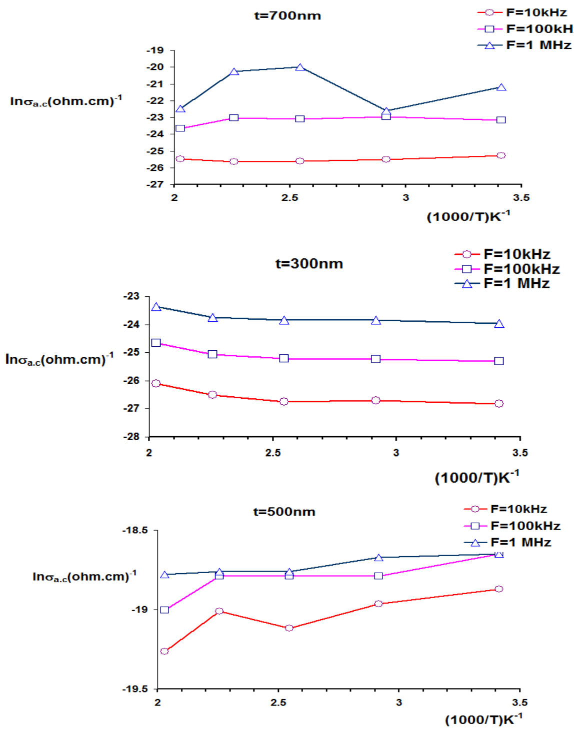


Figure 1 Frequency dependence of AC conductivity for PbSe thin film with different thickness



.Figure 2. Dependence of temperature on A.C conductivity of PbSe with different thickness

3. 2. Temperature and frequency dependence of loss tange

The temperature and frequency dependence of loss tangent tan δ were performed on Al/PbSe/Al thin films sandwich structures in order to understand the mechanism of dielectric relaxation.

Figure 3 shows the frequency dependence of loss tangent(tan δ) at different temperatures for PbSe film of different thicknesses. Tan δ has been found to decrease with increase of thickness, while Tan δ has been found to decrease with frequency at different temperatures, pass through a minimum value tan δ_{min} and thereafter increases. As the temperature is increased, the frequency at which (tan δ)min occurred shifted to higher frequencies in contrast to this behavior tan δ for PbSe thin films with higher thickness (700nm)has been found to increase with frequency at different temperatures, pass through a maximum value tan δ_{max} and thereafter decreases. As the temperature is increased, the frequency at which (tan δ)max occurred shifted to higher frequencies. This type of behavior is similar to that described by Simmons [21,22] for films having parallel RC elements of the materials in series with a parallel combination of Schottky barriers capacitance and resistance. They developed a model in which the sandwich structure is assumed to comprise a frequency independent capacitive element in parallel with a discrete temperature-dependent resistive element R, both in series with Schottky barrier capacitance and resistance. R is related to the temperature by the equation $R= R_0 \exp(\varphi/kT)$, where φ is the activation energy. From this relation, it is clear that R will decrease with an increase of temperature. Hence, tan δ_{max} shifts to the higher frequencies with increasing temperature.

3.3. *Temperature and frequency dependence of dielectric constant ε_r and ε_i*

The temperature dependence of dielectric constants ε_r *and* ε_i in the frequency range (10kHz-10MHz)and over the temperature range of 293- 493 K of *PbSe* thin films are shown in Fig.4 and 5. At all frequencies, the dielectric constant increases with temperature< 443 K, and thereafter decreases. The variation of ε_r with frequency was small for lower thicknesses. The variation of ε_r *and* ε_i with frequency was small at low temperatures. A significant variation of ε_r *and* ε_i with frequency was observed with higher thickness . In the absence of field, the charge carriers that are bound at different localized states would have different dipole orientations. The carriers can be considered localized with a strong electron- phonon interaction resulting in the formation of a small polarons [23]. An electron can hop between a pair of these centers under the action of an ac field and the hopping is equivalent to the reorientation of an electric dipole. This process would give rise to frequency dependent complex dielectric constant. Hence, the increase of dielectric constant with the decrease in frequency can be attributed to the presence of dipoles. At room temperature, the dielectric constant ε_r *and* ε_i at 10 kHz increases from 0.00141 to 0.206 and from 0.00512 to 0.80when thickness increases from 300to 500nm while ε_r *and* ε_i decrease to 0.0419 and 0.000923 respectively with further increase of thickness. The increase of $\varepsilon_r$value may be attributed to the formation of a conductor or a nonlinear capacitor with a high-energy barrier. On the other hand, in heterogeneous materials or multiphase materials, the motion of charge carriers takes place through one phase and some charge carriers may be trapped and accumulated at interfaces and defects; as a result, the electric field distorts and the dielectric constant increases. This effect depends on the conductivity of the present phases and this type of polarization is called the Maxwell–Wagner effect. As the trapping states decreases as a result of thickness increase , the electric field distortion decreases consequently the dielectric constant will be reduced.

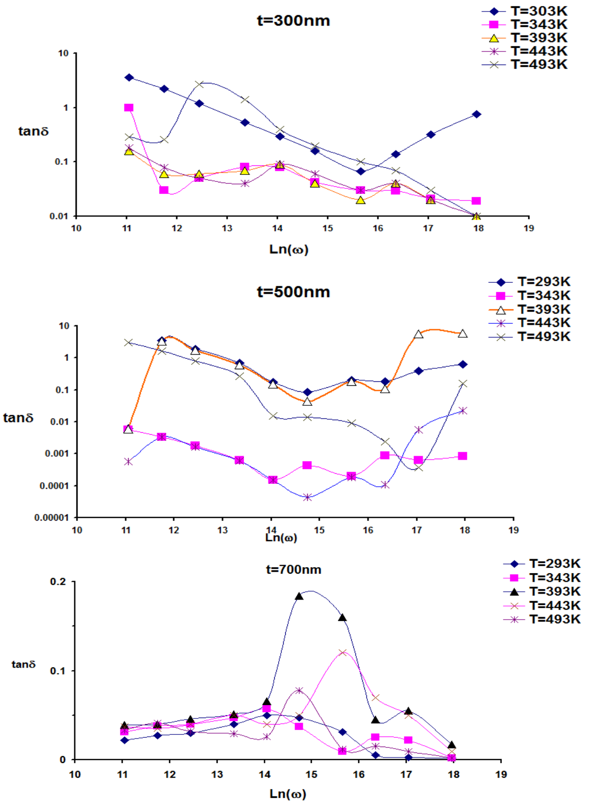


Figure 3. Frequency dependence of tanδ for PbSe thin film with different thickness

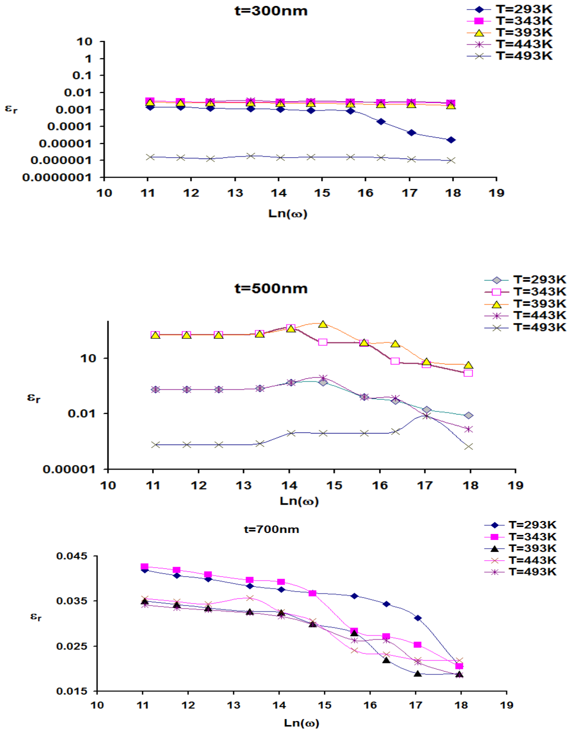
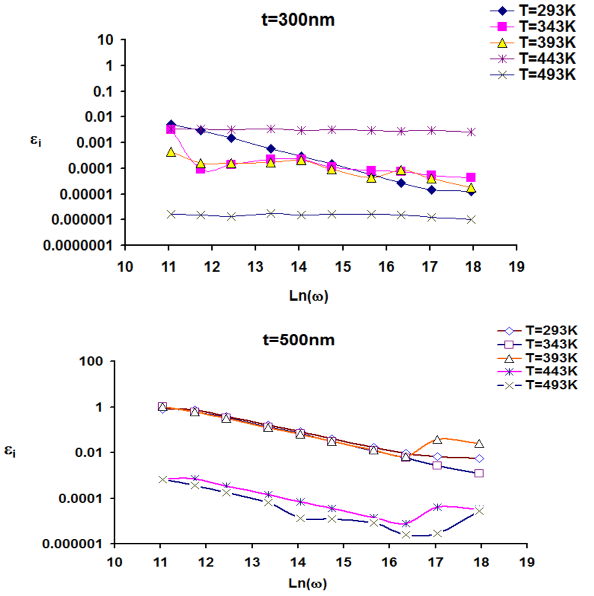


Figure 4. Frequency dependence of ε_r for PbSe thin film with different thickness ..



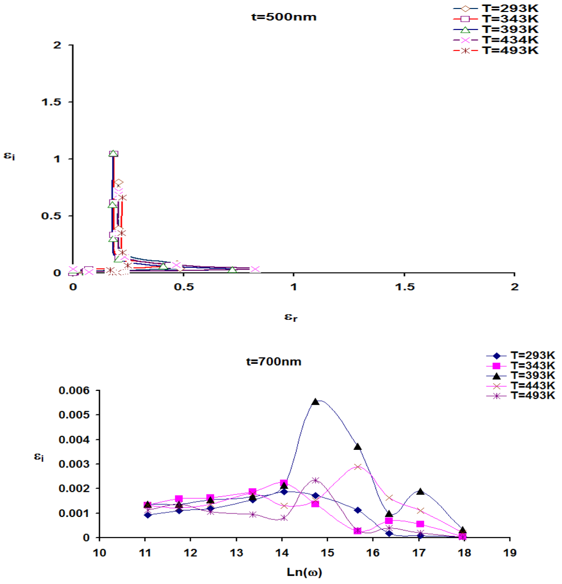


Figure 5. Frequency dependence of ε_i for PbSe thin film with different thickness .

4. COLE-COLE DIAGRAMS

There are three dispersion peaks which are (α ,β , and γ) each of which represents an individual relaxation process, (1) weak α-peak observed at temperature range 30 and 80^0C,(2) a well pronounced. a β-peak at approximately 0^0C,(3)γ-peak in the temperature range -50 and -100^0C. The γ-dispersion is probably due to torsional motion of the chain units, the origin of α-dispersion is the relaxation process in the crystalline phase, the β-dispersion produced by far the largest peak and is due to the motion of comparatively long chain segments in the amorphous phase the intensity of β-peak depends on the relative concentration of noncrystalline phase, thus the measurements of dielectric loss lead to analogous results regarding the molecular motion responsible for the relaxation processes.

A direct evidence of the existence of multi-relaxation time in PbSe films is obtaining by plotting Cole-Cole diagrams as shown in Fig.6 for different thicknesses. It has been observed that for all films reported here (ε_r) versus(ε_i) curves represent the arc of circles having their centers lying below the absicca axis. This confirms the existence of distribution of (τ) in all films. By measuring the angles (απ/2)the vales of the polarizability (α) had been determined and are listed in Table.1.α has been found to increase with increase of thickness and temperature pass through a maximum value and thereafter decreases, the decrease of (α) with the increase in thickness results from rise of the forces of the intermolecular [24,25],while the increase of (α)value comes from the weaken the forces.

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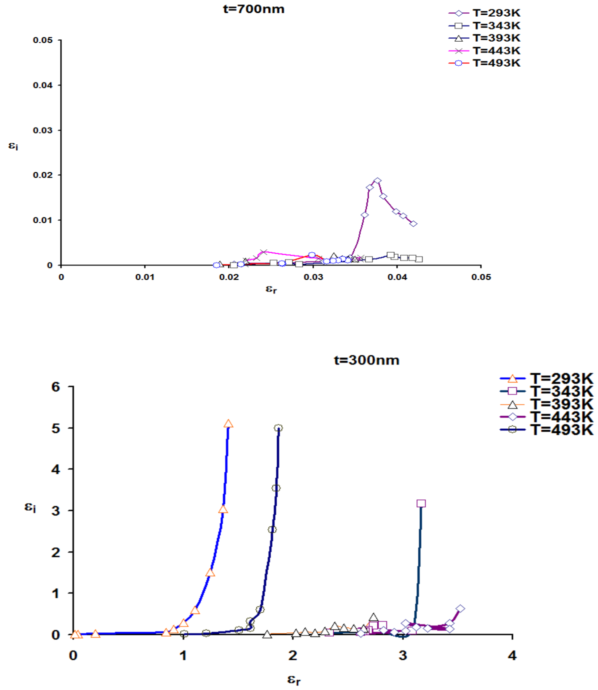

Fig.6 Cole – Cole diagrams of PbSe films for different thickness and temperatures.

5. CONCLUSION

In the present work, we can conclude the following:

1-The A.C conductivity ,ε_r andε_i are frequency independent for lower thicknesses and they are frequency dependent for higher thickness .

2- All A.C parameters like A.C conductivity, ε_r andε_i are frequency dependent in the high frequency range.

3- It is found that increases of thickness increases the force of the intermolecular, while increase of temperature reduces the force of the intermolecular i.e. formation a barrier.

4- It is more convenient to use PbSe with 500nm in thickness as a capacitor in electronic circuits since the values of polarisability in general (α≥0.5) and to use PbSe with residual thickness (300 and 700nm) as a resistor since the values of polarisability in general (α<0.5)

Table.1 Illustrates the values of s andα of PbSe films.

Thickness (nm)	Oven Temperature (K)	s	
300	293	0.057	0.1111
	343	0.618	0.2553
	393	0.632	0.3108
	443	0.660	0.3333
	493	0.631	0.2775
500	293	0.143	0.5328
	343	0.004	0.3663
	393	0.37	0.6666
	443	0.54	0.5555
	493	0.293	0.3996
700	293	0.538	0.1665
	343	0.591	0.0888
	393	0.92	0.1443
	443	0.877	0.1332
	493	0.575	0.1776

Table 2. Illustrates the $E_{a.c}$ values of PbSe films.

Thickness (nm)	$E_{a.c}$(eV)		
	F=10kHz	F=100kHz	F=1MHz
300	0.0386	0.0204	0.0146
500	0.0334	0.0171	0.0207
700	0.0297	0.0087	0.0066

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