

## DC conductivity of $Pb_xSe_{1-x}$ thin films prepared by thermal evaporation technique

Izzat M. Al-Essa\* and Ahmed Jumma Noori\*\*

\*Dept. of Physics, College of Science, University of Baghdad, Baghdad, Iraq

\*\*Directorate of Education Salahuddin, Education Department of Tuz, Karkuk, Iraq

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### ABSTRACT

In the present work, including study the effect of different concentration ( $X=0.1, 0.2, 0.3, \text{ and } 0.4$ ), substrate temperature (303 and 348) K, and annealed to  $348^{\circ}\text{K}$  on the electrical properties for  $Pb_xSe_{1-x}$  thin films, which prepared by thermal evaporation. D.C. conductivity showed increase with increasing concentration, substrate temperature and annealing temperature and all films have two activation energy. Hall measurements showed that the films at  $x=0.1$  and  $x=0.2$  was n-type, but at  $x=0.3$  and  $x=0.4$  was p-type with carrier's concentration and mobility increase with increasing of substrate temperature and annealing temperature.

**Keywords;** Lead chalcogenides, Optical absorption, Band gap, DC conductivity, lead salts, Barrier height

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### INTRODUCTION

Metal chalcogenide compounds, which are semiconductor in nature, are of considerable technical interest in the field of during the past decades [1]. The lead chalcogenides  $PbX$  ( $X= S, Se, Te$ ) have been a subject of a great amount of theoretical and experimental studies, motivated by their importance in infrared technology, and more recently, because of their utility in laser technology and as thermoelectric materials [2,3].

The lead salts exhibit properties which are unusual, relative to other semiconductors. Compared for example with the usual III-V compounds, these IV-VI chalcogens present non typical electronic and transport properties, such as higher carrier motilities, higher dielectric constants, narrow band gaps and a positive temperature coefficients, electronics and electro-optical devices [1,2,4].

Lead selenide is a polar semiconductor which has mixed ionic and overcomes the covalent bond where lead atoms connected to selenium atoms in dual ions form ( $Pb^{+2}$  and  $Se^{-2}$ ) [3,5]. It is possible to prepare n-type and p-type of  $PbSe$  thin film. The  $PbX$  compounds are narrow direct gap semiconductors, group IV-VI, which crystalline at ambient conditions in the cubic NaCl structure. The space lattice is face center cubic (f.c.c.) with lattice parameter ( $a=6.122\text{\AA}$ ), energy gap equals 0.27eV at room temperature, dielectric constant is 20, the melting point of  $PbSe$  is  $1338^{\circ}\text{K}$  and the refractive index at ( $3\mu\text{m}$ ) is 4.54 [6,7].

In this study the effect of concentration of selenium, substrate temperature and annealed temperature on the electrical properties of  $Pb_xSe_{1-x}$  thin films were investigated.

### MATERIALS AND METHODS

The  $Pb_xSe_{1-x}$  with different  $X(0.1, 0.2, 0.3, 0.4)$  compound were prepared as alloys by using high purity (99,999%) (lead and selenium) metal obtained from Balzeres company. Each element weighted according to its atomic weight and then mixing in quartz tube (length=25 cm, diameter=0.9 cm) evacuated at pressure of ( $10^{-3}$  mbar). The tube was

sealed and heated in electrical program controller furnace of type (Qallenhamp) at temperature 950 K and maintained at this temperature for about 5 hours and then allowed to cool slowly to room temperature. After that the ampoule was broken and the prepared compound of  $Pb_xSe_{1-x}$  was taken out and powdered to grain powder. This powder was used to prepare the films by thermal evaporation using Edward coating unit (model E306A) at a pressure of  $(10^{-5})$  Torr. A molybdenum boat is used as the evaporation source and the substrates are placed directly above the source at a distance of nearly 15cm. The glass substrates is freshly cleaned with a pure alcohol and distilled water followed by ultrasonic agitation.

The electrical conductivity has been measured as a function of temperature for  $Pb_xSe_{1-x}$  films. The measurements have been done using sensitive digital electrometer type Keithley (616) and electrical oven. The activation energies was calculated from the slope of the plot of  $\ln \sigma$  versus  $1000/T$  according to equation [8]:

$$\sigma = \sigma_0 \exp(-E_a/K_B T) \quad (1)$$

Where  $\sigma_0$  is the minimum electrical conductivity at  $0^0K$ ,  $E_a$  is the activation energy,  $T$  is the temperature and  $K_B$  is the Boltzmann's constant.

Hall Effect measurement was determined by using HMS3000 Hall measurement setting. Applying a magnetic field  $B$  [called "Hall field"] perpendicular to the electric field, yields a current ( $I$ ) then the transverse electric voltage is given by [9].

$$R_H = \frac{V_H}{I} \cdot \frac{t}{B} \quad (2)$$

where  $I$  is Hall current,  $V_H$  is Hall voltage and  $R_H$  is Hall coefficient. From the Hall coefficient equation we can determine the carrier's concentration of the semiconductor, and the carrier type, since  $R_H$  is negative for n-type and positive for p-type:

$$\text{For n-type } R_H = -1/n.e \quad (3)$$

$$\text{For p-type } R_H = 1/p.e \quad (4)$$

If the conduction is due to one carriers type e.g. electrons, then the mobility can measure [10]:

$$\mu_n = \frac{\sigma}{n.e} = \sigma |R_H| \quad (5)$$

## RESULTS AND DISCUSSION

The crystal structure of  $Pb_xSe_{1-x}$  films, which were evaporated on glass substrates with thickness  $(150 \pm 20)$  nm at room temperature were shown in figure (1). The structure become a mixture of cubic and hexagonal at  $(X=0.2, 0.3$  and  $0.4)$ . The preferred orientation lies along (200) direction. Our results are nearly in agreement with Al-Woely [11] and Kassim et al [12].

The d.c conductivity for  $Pb_xSe_{1-x}$  films has been studied as a function of  $10^3/T$  with the range of (300-473 K), at different value of  $X$  (0.1, 0.2, 0.3, 0.4), different substrate temperatures (303, and 348) K, and annealed to 348  $^0K$ . as shown in figure(2). It is clear from these figure that there were two transport mechanisms, giving rise to two activation energies  $E_{a1}$  and  $E_{a2}$ . At higher temperature range (331-393) K, the conduction mechanism is due to carrier excited into the extended states beyond the mobility edge. At lower temperature range (291 - 331) K; the conduction mechanism is due to carrier excited into localized states at the edge of the band. We can review the results as follows: From figure the  $(\sigma_{d.c.})$  of  $Pb_xSe_{1-x}$  films increases as the Pb concentration ( $x$ ) increases. This may be due to change in the localized states, structure, and composition of films as well as to the re-arrangement of atoms which yields fewer defects. This results are agreement with Kumar et al [13].

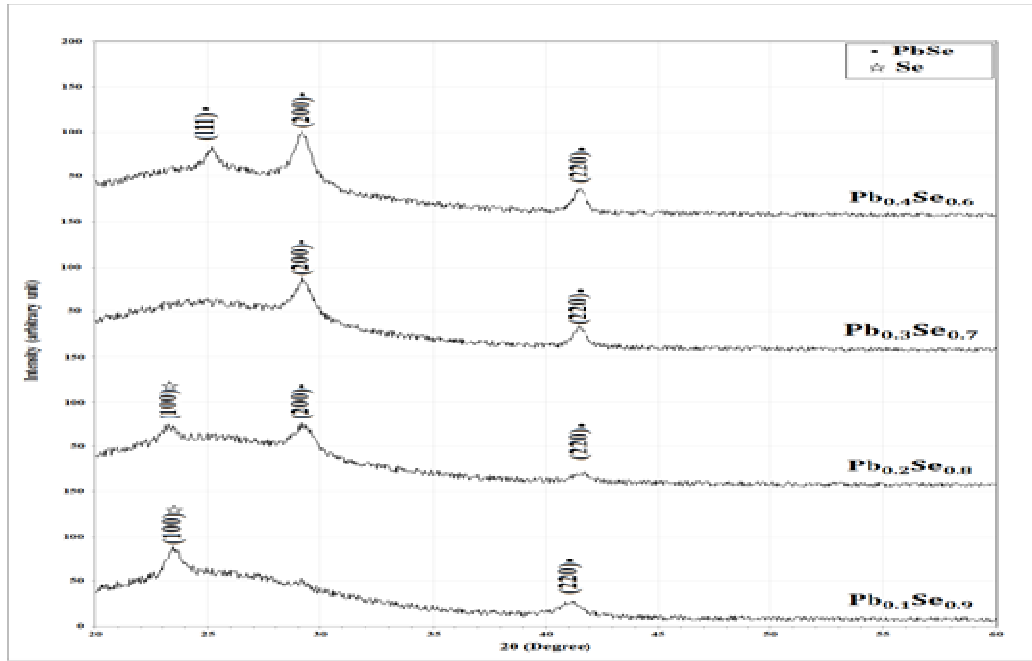


Fig. (1) X-ray diffraction patterns of  $Pb_xSe_{1-x}$  prepared at room temperature with different  $x$  (0.1, 0.2, 0.3 and 0.4)

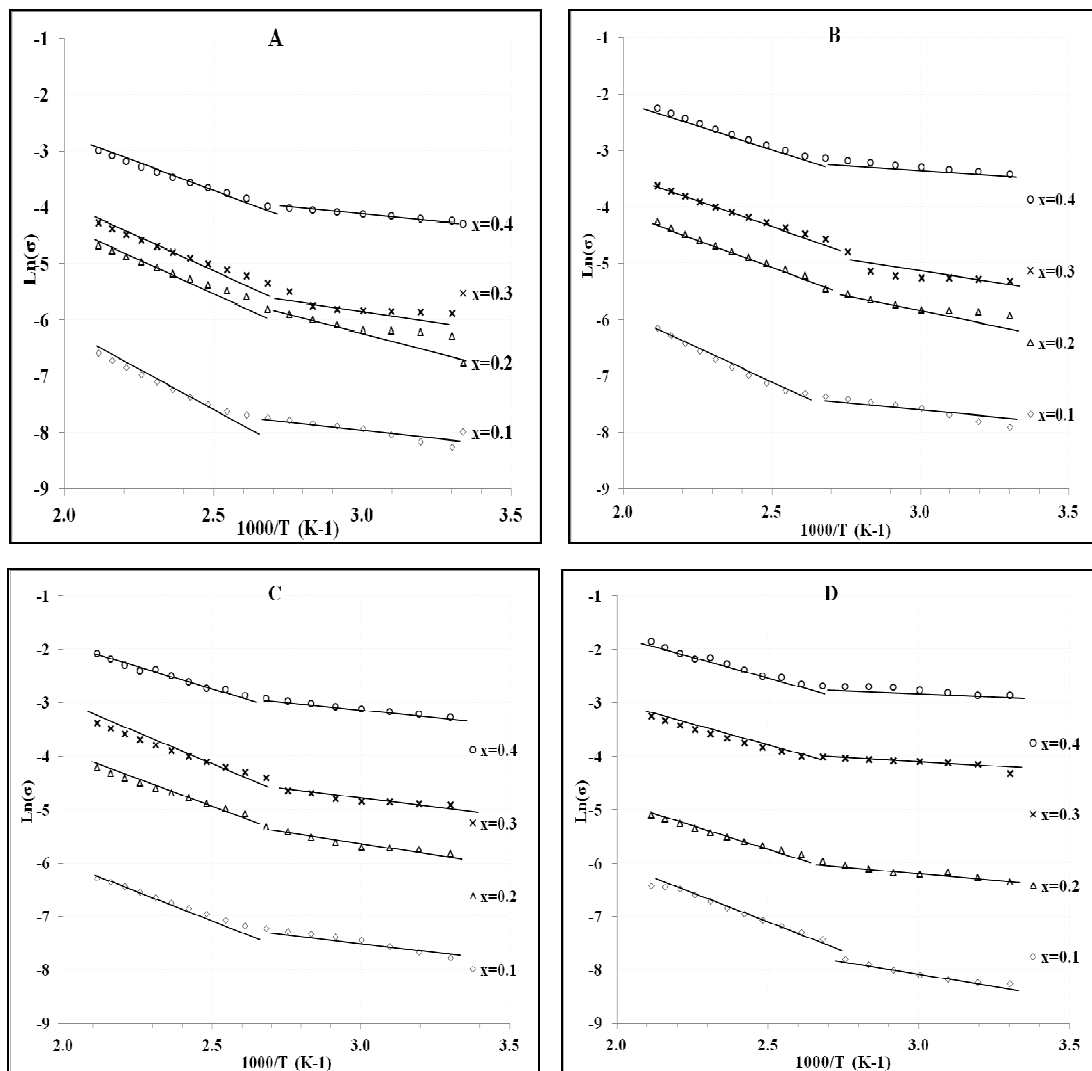


Fig.(2)  $\ln \sigma$  versus  $1000/T$  for  $Pb_xSe_{1-x}$  (A)  $T_a$  &  $T_s$  at RT, (B)  $T_a=348K$  and  $T_s=RT$  (C),  $T_a=RT$  and  $T_s=348K$ , (D)  $T_a$  &  $T_s$  at 348K

It was found that the  $\sigma_{dc}$  conductivity of  $Pb_xSe_{1-x}$  films with different x increases with increasing the substrate temperatures ( $T_s$ ) and annealing temperature ( $T_a$ ). The activation energies decrease with increasing the substrate temperature and annealing (as shown in figure 3) due to decrease the energy gap.

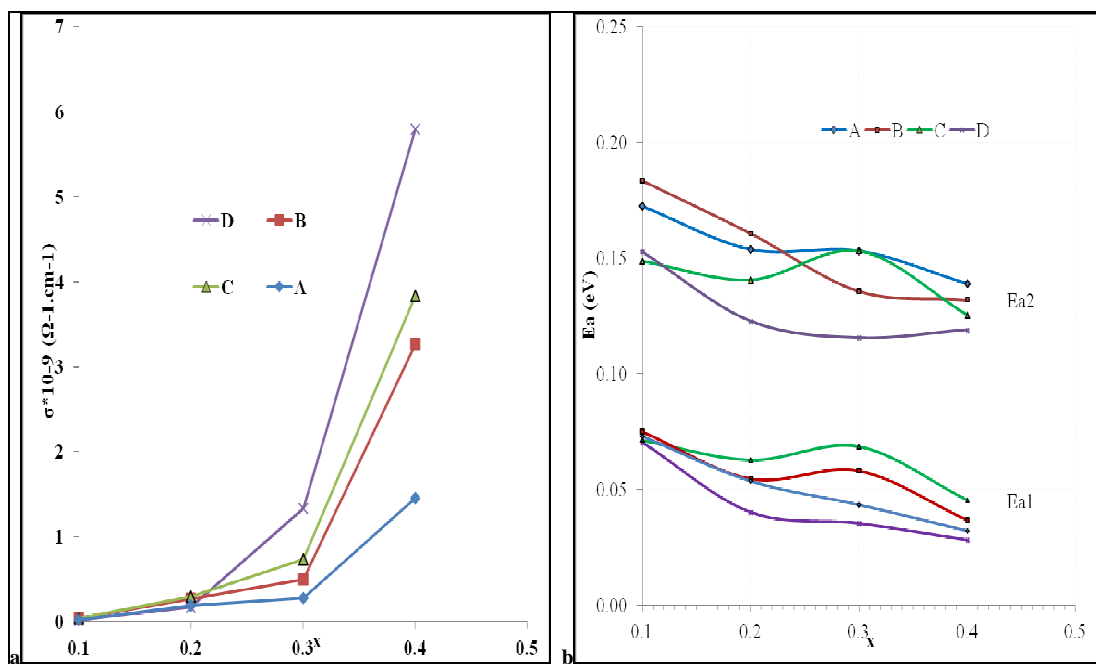


Fig.(3)(a)  $\sigma_{dc}$  vs. Pb concentration & (b)  $E_{a1}$  and  $E_{a2}$  vs. Pb concentration for  $Pb_xSe_{1-x}$  at (A)  $T_a$  &  $T_s$  at RT, (B)  $T_a=348k$  and  $T_s=RT$ , (C)  $T_a=RT$  and  $T_s=348K$ , (D)  $T_a$  &  $T_s$  at 348K

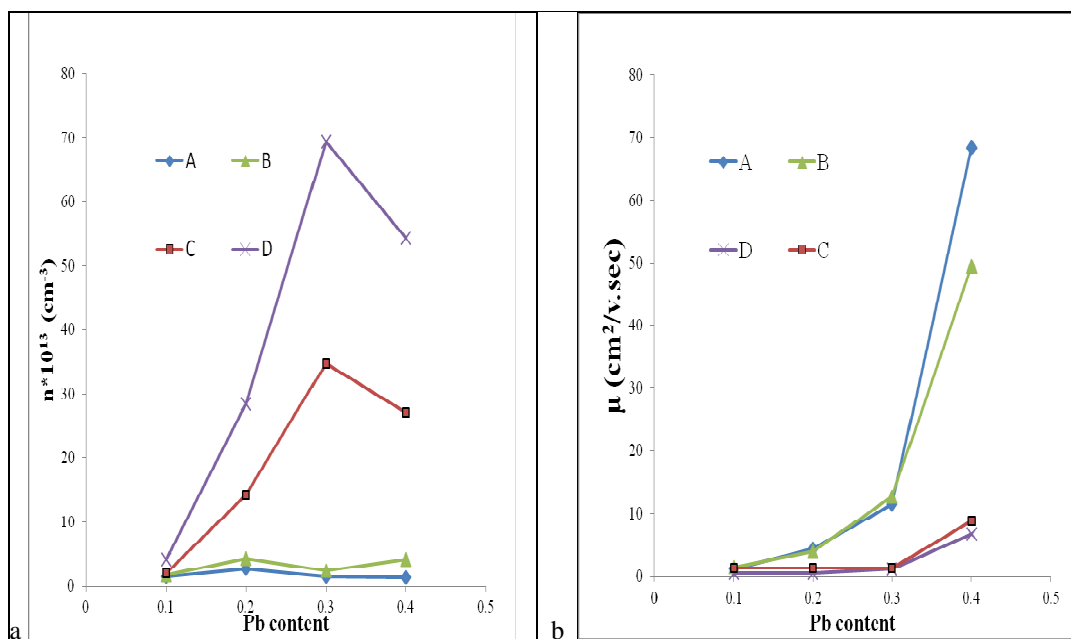
The value of ( $\sigma_{dc}$ ) and activation energies are listed in Table (1).

Table (1): D.C Electrical conductivity and activation energies at different values of (x) for  $Pb_xSe_{1-x}$  with the Effect of substrate and annealing temperature

$T_a$ (°k)	$T_s$ (°k)	x	$\sigma_{RT} \cdot 10^{-2}$	$E_{a1}$ (eV)	Range (K)	$E_{a2}$ (eV)	Range (K)
		0.1	0.026	0.0731	303-373	0.1725	373-473
RT	RT	0.2	0.187	0.0537	303-373	0.1538	373-473
		0.3	0.281	0.0434	303-373	0.1532	373-473
		0.4	1.456	0.0322	303-373	0.1389	373-473
		0.1	0.037	0.0750	303-373	0.1833	373-473
348k	RT	0.2	0.265	0.0546	303-373	0.1606	373-473
		0.3	0.498	0.0580	303-373	0.1357	373-473
		0.4	3.268	0.0366	303-373	0.1320	373-473
		0.1	0.042	0.0713	303-373	0.1487	373-473
RT	348k	0.2	0.299	0.0628	303-373	0.1406	373-473
		0.3	0.736	0.0686	303-373	0.1531	373-473
		0.4	3.831	0.0453	303-373	0.1254	373-473
		0.1	0.026	0.0705	303-373	0.1528	373-473
348k	348k	0.2	0.174	0.0401	303-373	0.1228	373-473
		0.3	1.333	0.0353	303-373	0.1156	373-473
		0.4	5.797	0.0281	303-373	0.1190	373-473

The sign of Hall coefficient ( $R_H$ ) indicates that  $Pb_{1-x}Se_x$  thin films behaves as a n-type semiconductor for ( $x=0.1$  and  $x=0.2$ ) and p-type for ( $x=0.3$  and  $x=0.4$ ).

The carrier's mobility was calculated from equation (5). From the figure(4 a and b) it was found that all the carriers concentration increases with substrate temperature and annealing temperature, but mobility decrease with substrate temperature and annealing temperature increases.



(4)(a) and (b) Variation of Hall mobility and carrier concentration as the versus X for Pb<sub>x</sub>Se<sub>1-x</sub> films at (A) T<sub>a</sub>=RT and T<sub>s</sub>=RT (B) T<sub>a</sub>=348K and T<sub>s</sub>=RT (C) T<sub>a</sub>=RT and T<sub>s</sub>=348K (D) T<sub>a</sub>=348K and T<sub>s</sub>=348K

This may be due to the re-crystallization process, which leads to the decrease of defects in the film during the film growth, and consequently a decrease of the carriers scattering at the defect. The increasing of re-crystallization leads to rising the potential barrier, for that reason the mobility increasing. Our results are agreement with Nayef [4] and Arivazhagan et al [15].

It can be observed from table (2) that the carrier's concentration and mobility increasing with increase of the Pb concentration. The substrate temperature and annealing temperature considered as an effective parameter on the carrier concentration and mobility.

Table: (2): Hall parameters of Pb<sub>x</sub>Se<sub>1-x</sub> at X=0.1,0.2,0.3 and 0.4 with the effect of substrate temperature and annealing temperature

T <sub>a</sub> (K)	T <sub>s</sub> (K)	x	R <sub>H</sub> (cm <sup>3</sup> /c)	n*10 <sup>15</sup> (cm <sup>-3</sup> )	σ (Ω.cm) <sup>-1</sup>	μ(cm <sup>2</sup> /v.sec)	type
RT	RT	0.1	4.40E+03	1.42	0.0026	1.1	n
		0.2	2.30E+03	2.72	0.001869	4.3	n
		0.3	4.10E+03	1.52	0.002806	11.5	p
		0.4	4.70E+03	1.33	0.014556	68.4	p
348	RT	0.1	3.70E+03	1.69	0.000372	1.4	n
		0.2	1.47E+03	4.25	0.002653	3.9	n
		0.3	2.54E+03	2.46	0.004975	12.6	p
		0.4	1.52E+03	4.13	0.03268	49.5	p
RT	348	0.1	3.00E+03	2.08	0.000421	1.3	n
		0.2	4.40E+02	14.20	0.002988	1.3	n
		0.3	1.80E+02	34.72	0.007358	1.3	p
		0.4	2.30E+02	27.17	0.038314	8.8	p
348	348	0.1	1.50E+03	4.17	0.000261	0.4	n
		0.2	2.20E+02	28.41	0.001736	0.4	n
		0.3	9.00E+01	69.44	0.013333	1.2	p
		0.4	1.15E+02	54.35	0.057971	6.7	p

### CONCLUSION

The Pb<sub>x</sub>Se<sub>1-x</sub> with different X(0.1, 0.2, 0.3, 0.4) compound were prepared successfully as alloys by using high purity (99,999%) lead and selenium metal. The D.C. conductivity for all films increases as the Pb concentration, substrate temperature and annealing temperature increase. There are two transport mechanisms of the charge carriers. In general the activation energies decrease with increasing Pb concentration, substrate temperature and annealing temperature. The Hall measurements showed that Pb<sub>x</sub>Se<sub>1-x</sub> films were n-type at x=0.1 and x=0.2, and p-type at x=0.3 and x=0.4. Carrier's concentration and mobility increase with increasing with the Pb concentration. While the carrier's concentration increases with substrate temperature and annealing temperature, but mobility decreases

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