

X-Ray Diffraction and DC Electrical Conductivity for $\text{Te}_{61.8}\text{Se}_{38.2}$ Thin Films

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Abstract: Vacuum alloying system was designed and built up to alloy ($\text{Te}_{61.8}\text{Se}_{38.2}$). $\text{Te}_{61.8}\text{Se}_{38.2}$ thin films were prepared by thermal evaporating system under pressure about (5×10^{-5}) mbar. Three thin films samples were prepared in one evaporation run, one in the center of the evaporation source and the other two beside it (using flat substrate holder) to study the effect of evaporation angle on properties of $\text{Te}_{61.8}\text{Se}_{38.2}$ thin film. X-ray fluorescence (XRF) has been used to investigate the purity of the primary materials and $\text{Te}_{61.8}\text{Se}_{38.2}$ alloy. Also (XRD) system has been used to investigate the structure of the $\text{Te}_{61.8}\text{Se}_{38.2}$ alloy and $\text{Te}_{61.8}\text{Se}_{38.2}$ thin films. The electrical measurements of thin film samples, showed that the center sample has an activation energy (0.575 eV and its thickness 415 nm) and for sided sample has an activation energy (0.61 eV and its thickness 340 nm).

Keywords: Amorphous materials; Thin films, X-Ray diffraction, X-Ray fluorescence and Electrical properties

1. Introduction

A homogeneous mixture of two or more metals or a metal and a nonmetal, when fused together at a certain temperature forms a new metal after solidification termed as an alloy. In the solid state an alloy may be present in one or more of the following forms [1];

- (i) As a solid solution.
- (ii) As an intermediate phase or intermediate chemical compound.
- (iii) As a finely divided mechanical mixture of solid solution.
- (iv) As a finely divided mechanical mixture of metals.
- (v) As a finely divided mechanical mixture of chemical compound of metals, the individual metals and solid solutions.

Hume-Rothery rules predict which metals will form solid-solutions based on the relative sizes and electronic properties of the metal atoms [2]. The liquid Te-Se alloy system spans a wide range of electronic behavior between metals and insulators [3]. In recent years, thin film science has grown world-wide into a major research area. Thin film of particular interest for fabrication the large area of arrays solar selective coating, solar cell, photoconductors sensor, antireflections coating, interference items, polarizer's, narrow band filters, IR detectors, wave guide coatings, temperature controller of satellites, photo thermal solar coating etc. [4]. Te and Se form alloys over the complete composition range, where the Te and Se atoms are randomized in a copolymer chain in the trigonal phase [5,6]. Electrical properties of liquid Te-Se mixtures at high temperatures and high pressures, was studied by [7]. The electrical conductivity σ and the thermoelectric power S have been measured for liquid Te-Se mixtures in a wide temperature and pressure range. Substantial changes in σ and S are induced by a slight application of pressure. The region

where such changes occur is determined the concentration-temperature plane. It is suggested that the observed semiconductor to metal transition is originated from the structural change. The main aim of this study is to investigate structural and electrical properties of thermally evaporated $\text{Te}_{61.8}\text{Se}_{38.2}$ thin films.

2. Experimental Methods

2.1 Preparation of $\text{Te}_{61.8}\text{Se}_{38.2}$ Alloy

The $\text{Te}_{61.8}\text{Se}_{38.2}$ alloy was prepared by taking the required proportions (by weight) of 99.999% pure Se and Te in Pyrex ampoule. The Pyrex glass tube was cleaned using degreasing chemicals like Alcohol and Acetone. By preheating the glass tube under vacuum to complete degreasing all the contaminations inside the tube. Materials with specific weight percent of Te-Se has been inserted in the glass tube (20 cm from the tube bottom was narrowed like neck). The encapsulations process was done in a vacuum of (2×10^{-2} mbar), then tightly sealing the capsule and inserted into the electric oven, and heated up to 973K for six hours with furnace shaken for several times during the course of synthesis to increasing the homogeneity of the alloy, and to ensure complete mixing of the constituting.

2.2 Preparation of $\text{Te}_{61.8}\text{Se}_{38.2}$ thin film

Thin films were prepared by thermal evaporation. The vacuum system which was used to prepare the thin films is from Edwards company type E306. At first time, thin aluminum poles for electrical properties study were deposited using tungsten coil as heating element and then the powder of $\text{Te}_{61.8}\text{Se}_{38.2}$ (after milling $\text{Te}_{61.8}\text{Se}_{38.2}$ ingot, which was already prepared by our alloying system) evaporated by using molybdenum boat (Mo). Three microscopic glasses were used as substrates were arranged as one in the center of the evaporation source, and the other

two glass substrates surround the centered substrate from its two sides. The evaporation process has been done in a vacuum of less than (5×10^{-5}) mbar.

3. Results and Discussion

After broken the capsule, (before depositing the films) the ingot ($\text{Te}_{61.8}\text{Se}_{38.2}$ alloy) were tested by using optical microscope. The test reveals that the outside and inside layer of the alloy was very bright, which indicates that there is no chance of oxidation associated with alloying process [8]. The qualitative chart of X-Ray fluorescence of $\text{Te}_{61.8}\text{Se}_{38.2}$ alloy points out that the alloy is very pure and the elements which have been used to prepare it are also characterized by high purity. As shown in the Fig.(1) the intensity of Se peaks

is higher than the intensity of Te peaks in the ingot of $\text{Te}_{61.8}\text{Se}_{38.2}$ alloy.

Four samples of $\text{Te}_{61.8}\text{Se}_{38.2}$ thin films were tested by X-Ray fluorescence as qualitative analysis. The four samples are:

1. AS prepared Centered sample (CS) (the position of the substrate in the center of the evaporation source)
2. AS prepared Sided sample (SS) (the position of the substrate in the side of the CS)
3. Centered sample (CS) heated at 463 K for one hour.
4. Sided sample (SS) heated at 463 K for one hour.

The X-Ray fluorescence charts of the above samples (1, 2, 3, 4) are shown in Figs. (2), (3), (4), (5).

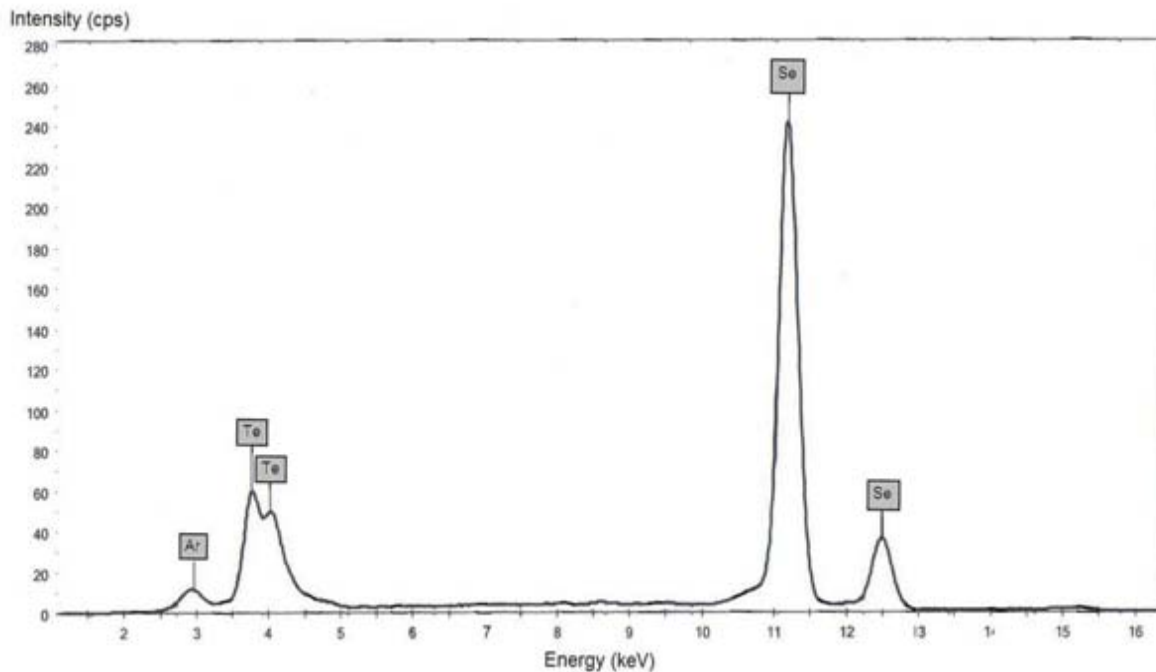


Figure 1: The X-Ray fluorescence qualitative chart of $\text{Te}_{61.8}\text{Se}_{38.2}$ alloy

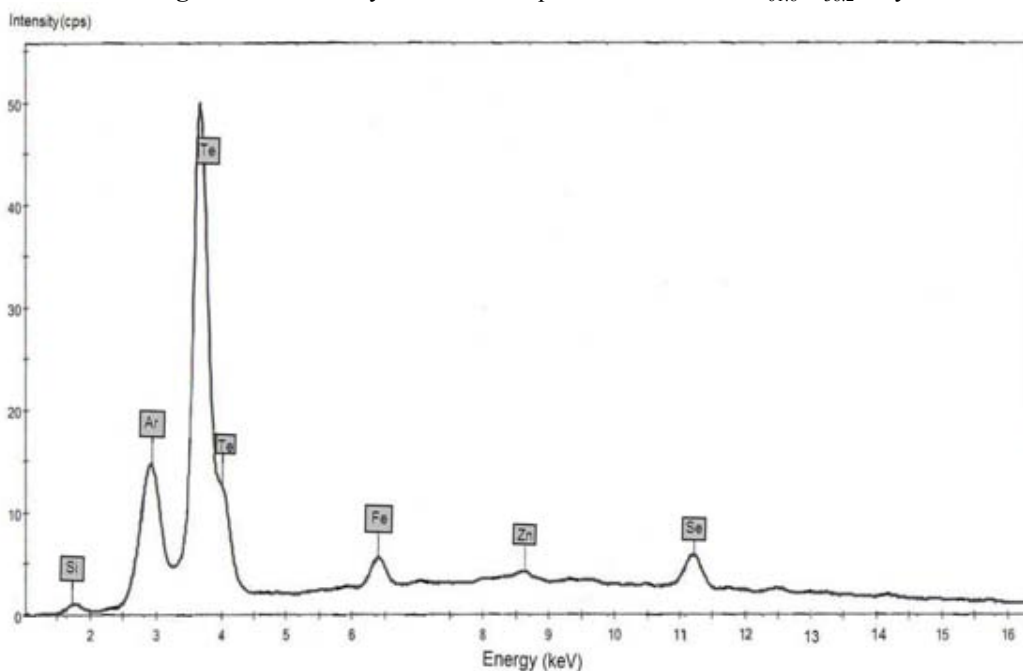


Figure 2: X-Ray fluorescence chart of (CS) film annealed at 398 K for one hour

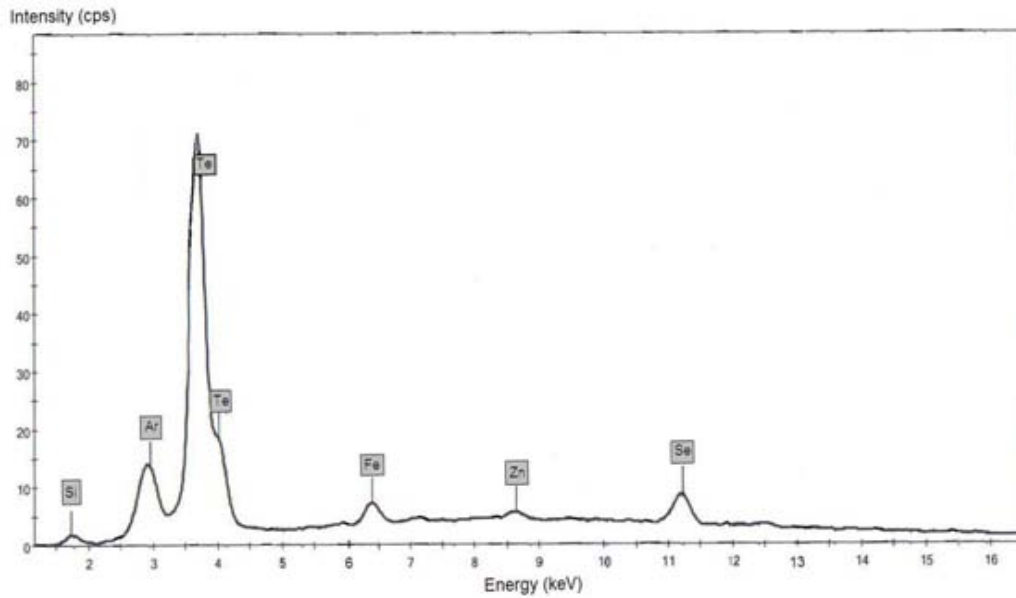


Figure 3: X-Ray fluorescence chart of (SS) film annealed at 398 K for one hour

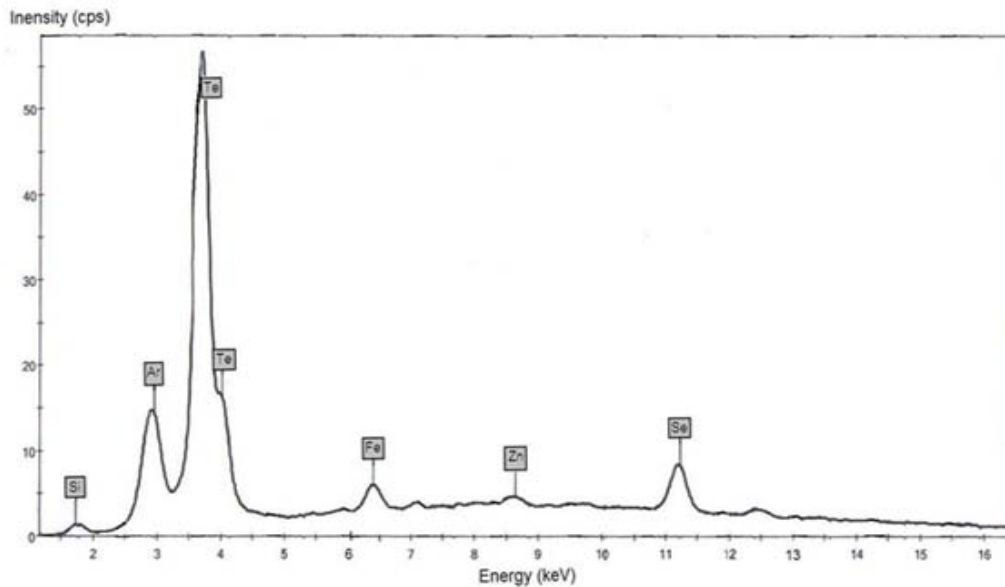


Figure 4: X-Ray fluorescence chart of (CS) film annealed at 463 K for one hour

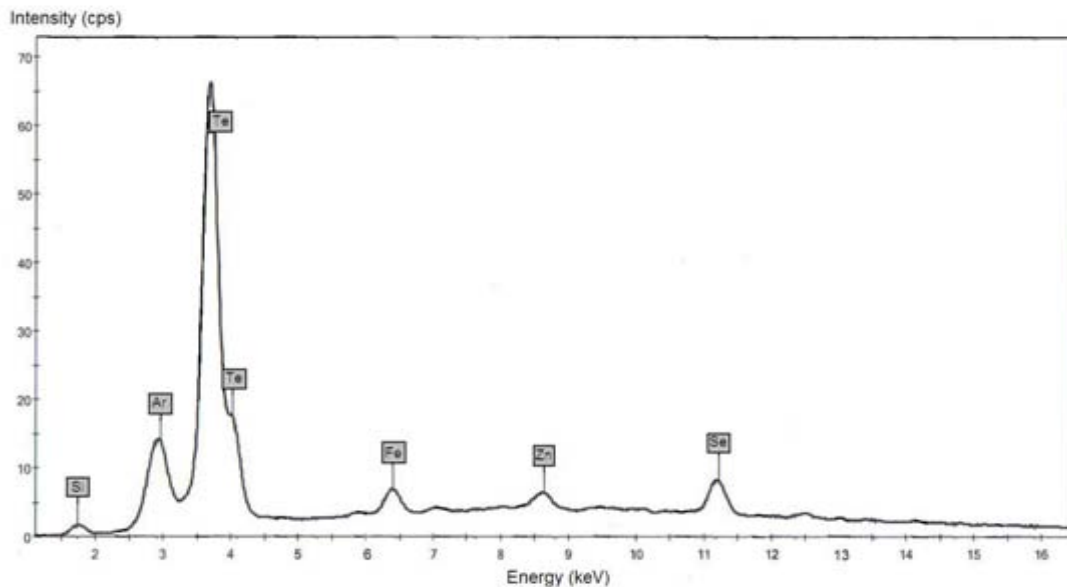


Figure 5: X-Ray fluorescence chart of (SS) film annealed at 463 K for one hour

One from figure (1) noticed that the peaks intensity of Te and Se are completely differ than that from all other thin films as seen latter. This difference in intensity is believed to be due to inhomogeneous distribution of Te and Se during alloying process, and wherever most of the Se atoms will be undeposited on the substrate due to its light weight, which consent to deflect and diffracted Se atoms from the substrate.

From the Figs. (2,3,4, and 5), the first three foreign elements (Si, Fe and Zn) appears in the four charts of the $\text{Te}_{61.8}\text{Se}_{38.2}$

thin film which are expected from the substrate slide chemical compositions which is made of glass. However, the last foreign element Ar (Argon gas) is expected to appear because they have been used Ar gas instead of He (Helium gas) in operating process of XRF device[9-12].

Same samples of thin films which were investigated by XRF unit were also tested by XRD unit. Fig. (6), (7) and (8) represent CS as deposit, and annealed at 398K and annealed at 463K for one hour, respectively.

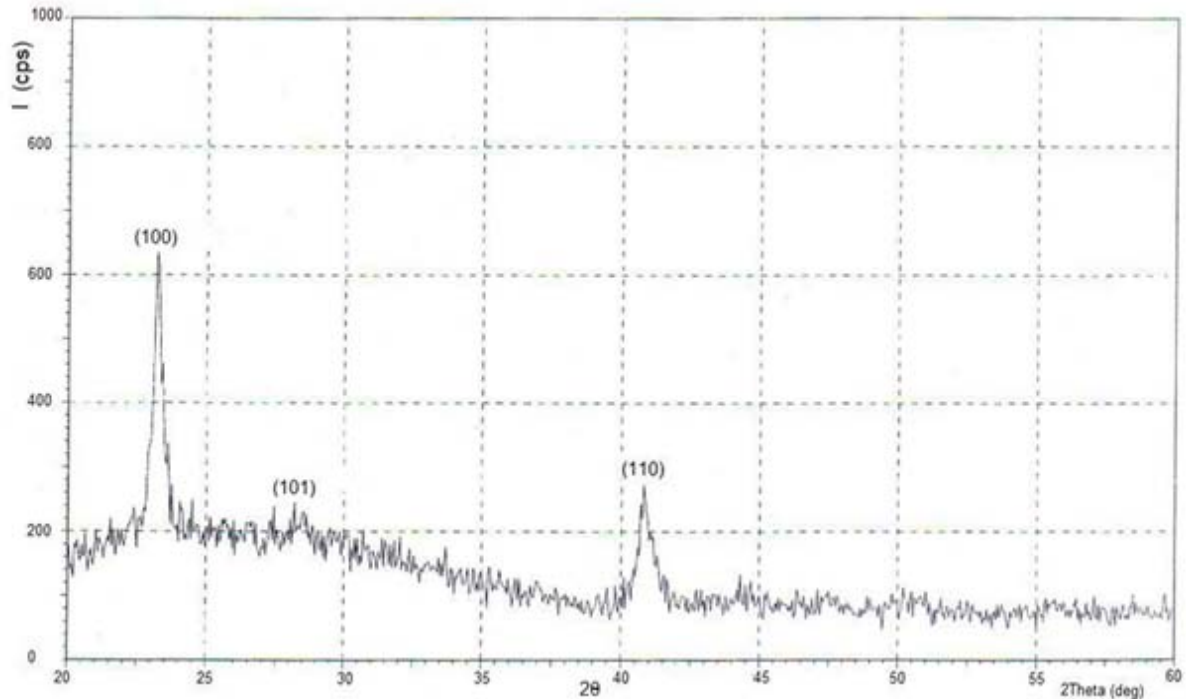


Figure 6: X-ray diffraction pattern for $\text{Te}_{61.8}\text{Se}_{38.2}$ thin film (CS as deposit)

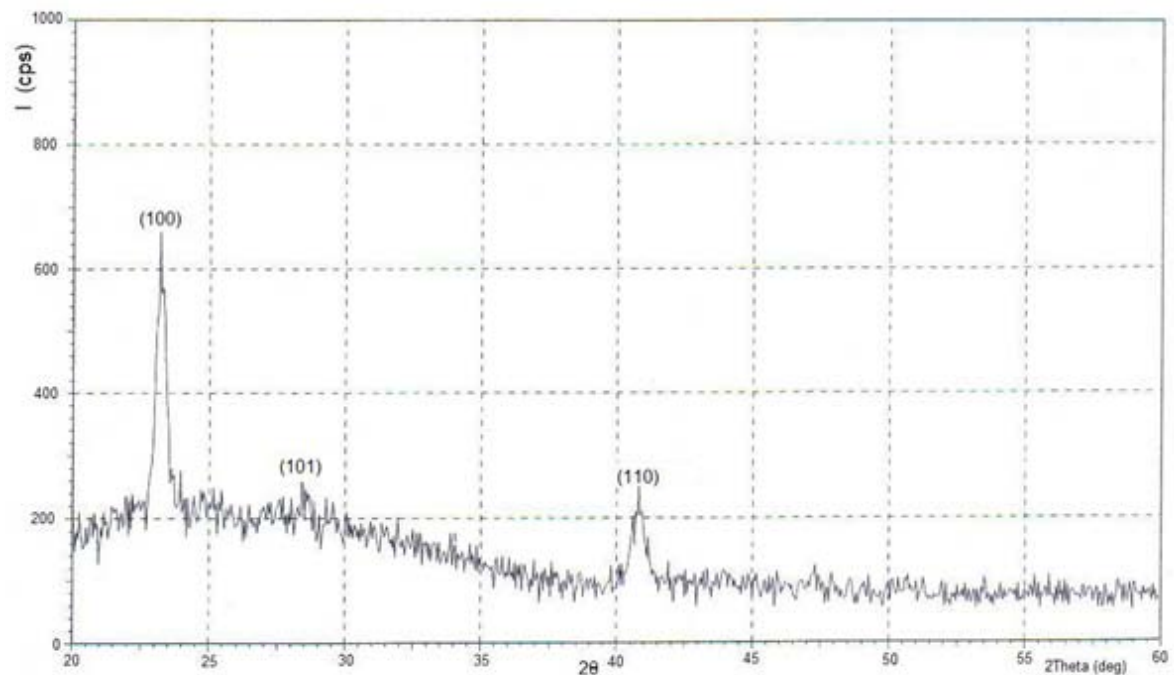


Figure 7: X-ray diffraction pattern for $\text{Te}_{61.8}\text{Se}_{38.2}$ thin film (CS annealed at 398K)

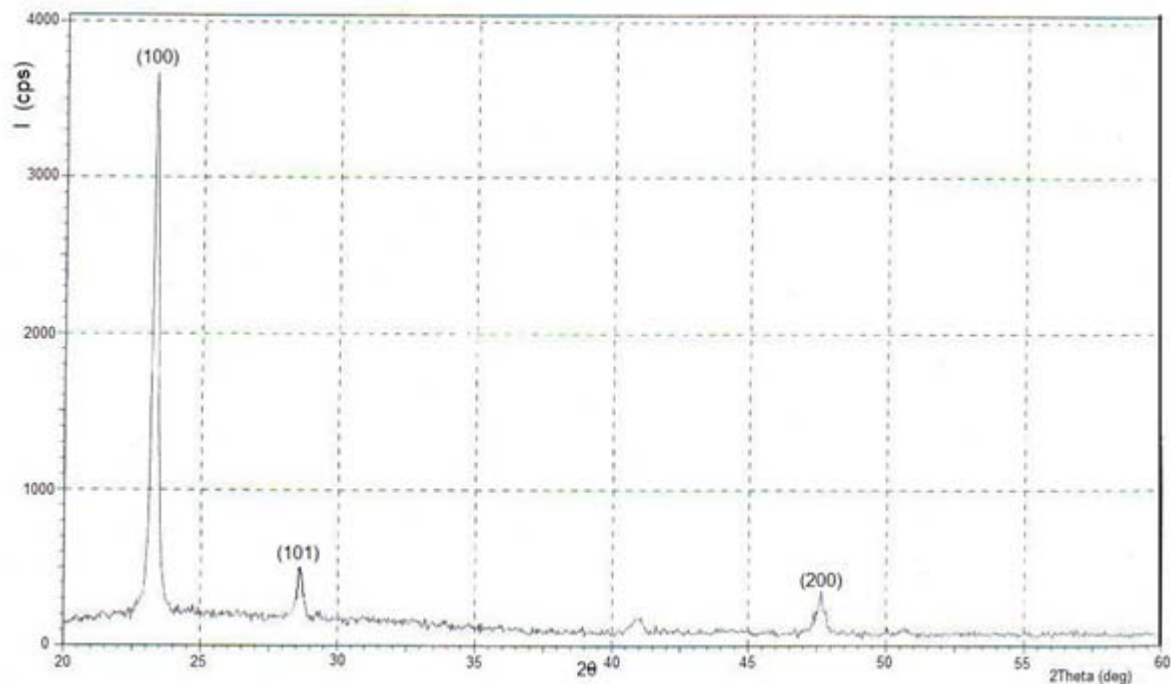


Figure 8: X-ray diffraction pattern for $\text{Te}_{61.8}\text{Se}_{38.2}$ thin film (CS annealed at 463 K)

The figures show that there are no peaks belong to Te or Se and this means that the Te and Se elements produce substitutional solid solution without rich of Te or Se [3, 13]. It is also shown that the crystallite are nearly the same in 398K if compared with the as deposit thin film. It is thought that the energy which gained by the atoms at 398K is not enough to rearrange the atoms to produce more ordering crystallite. On the other hand, when it has been annealed

at 463K, the crystallization becomes more pronounced (more large grains).

When the sample temperature increases, the intensities of peak will also increases and indicate the increase in the degree of the crystalline [14]. Fig.(9), (10) and (11) represent SS as deposit, annealed at 398K and annealed at 463K for one hour, respectively.

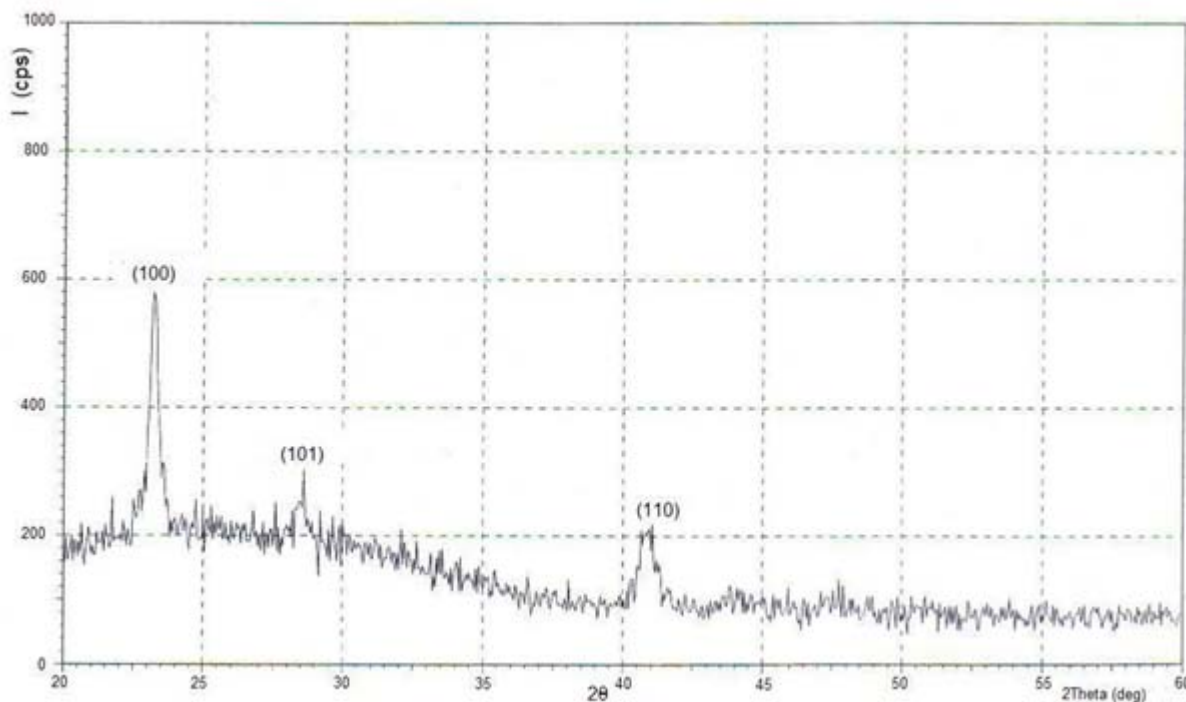


Figure 9: X-ray diffraction pattern for $\text{Te}_{61.8}\text{Se}_{38.2}$ thin film (SS as deposit)

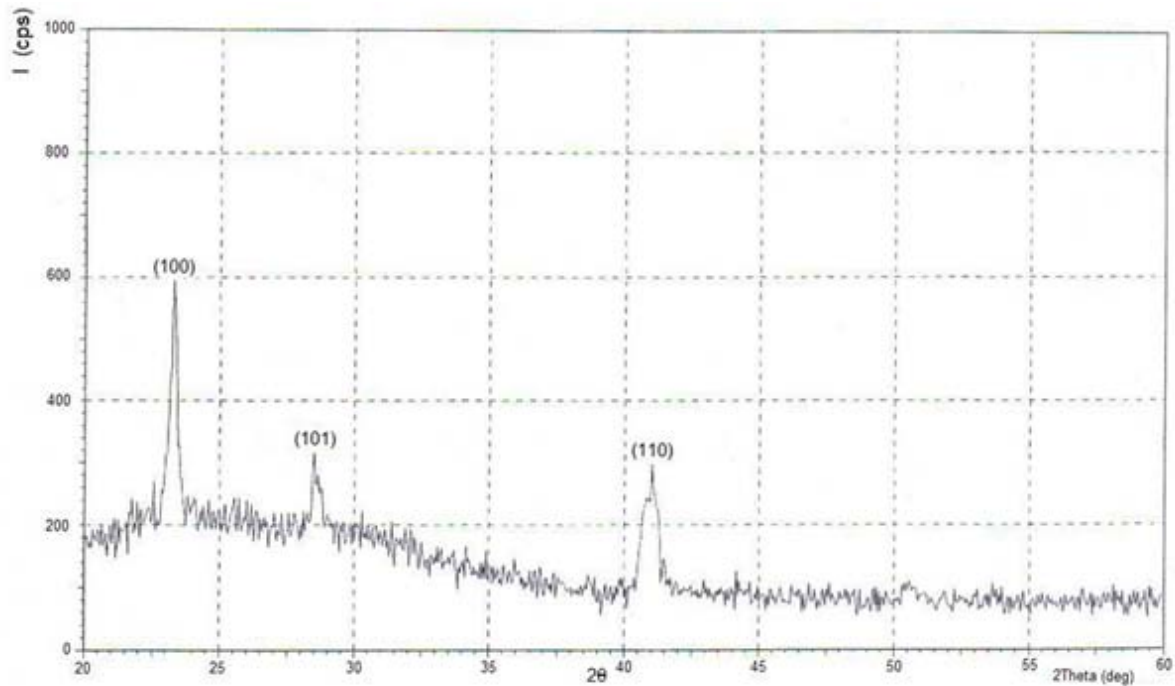


Figure 10: X-ray diffraction pattern for $Te_{61.8}Se_{32.2}$ thin film (SS annealed at 398 K)

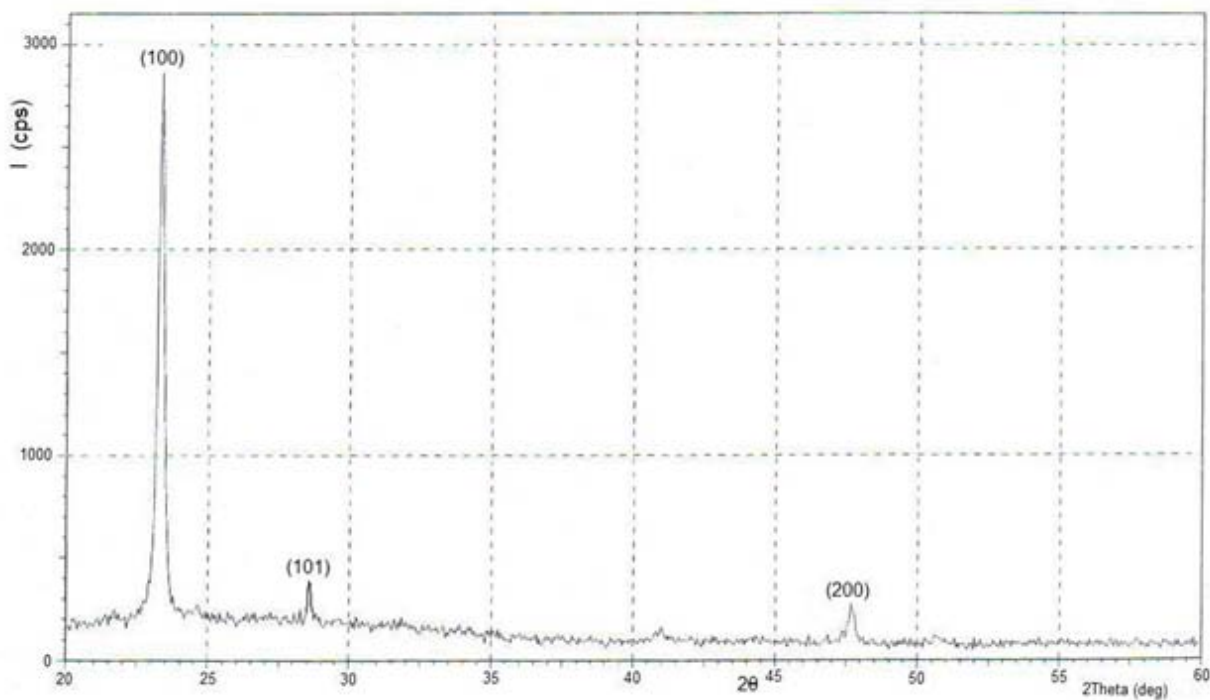


Figure 11: X-ray diffraction pattern for $Te_{61.8}Se_{38.2}$ thin film (SS annealed at 463 K)

Figs.(6) and (9), give a notification that the crystallite in Fig. (6) is more ordered due to the thickness of CS sample which is more than the thickness of SS sample. It can also be seen that the peak intensity are increases and have more sharpness as film thickness increases [9, 13]. This can be attributed to the fact of the sample position reference to the boat. The interplanar distance for each peak in the spectrum was determine dusing Bragg’s diffraction equation. The calculated values of d_{hkl} for $Te_{61.8}Se_{38.2}$ alloy, CS and SS samples are shown in Tables (1), (2) and (3), respectively.

Table 1: values of hkl and $dhkl$ for $Te_{61.8}Se_{38.2}$ alloy

hkl	$dhkl(A^\circ)$
101	3.1957
110	3.1569
102	2.0566

Table 2: values of hkl and $dhkl$ for CS film sample

As deposit		Annealed at 398 K		Annealed at 463 K	
hkl	$dhkl(A^\circ)$	hkl	$dhkl(A^\circ)$	hkl	$dhkl(A^\circ)$
100	3.8206	100	3.8221	100	3.8100
101	3.1244	101	3.1237	101	3.1244
110	2.2051	110	2.2098	200	1.9100

Table 3: values of *hkl* and *d_{hkl}* for SS film sample

As deposit		Annealed at 398 K		Annealed at 463 K	
<i>hkl</i>	<i>d_{hkl}</i> (Å°)	<i>hkl</i>	<i>d_{hkl}</i> (Å°)	<i>hkl</i>	<i>d_{hkl}</i> (Å°)
100	3.8213	100	3.8142	100	3.8083
101	3.1235	101	3.1211	101	3.1129
110	2.2047	110	2.2024	200	1.9056

In order to study the temperature dependence of the electrical conductivity of the as-deposited $Te_{61.8}Se_{38.2}$ samples, films of 415 nm thickness (CS sample) and 340 nm thickness (SS) were deposited onto glass substrates by thermal evaporation. Figs. (12 and 13) show the temperature dependence of the electrical conductivity of the CS and SS as-prepared films, respectively. Three regions of $\ln\sigma$ vs. $1/T$ plot are shown in both figures. In the first region (I), which extends from temperature 338K up to 398K, the

conductivity varies with temperature according to the Arrhenian relation $\sigma = \sigma_0 \exp(-\Delta E_a / K_B T)$, where σ_0 is the pre-exponential factor and ΔE_a the activation energy for conduction which is 1.17 eV for CS sample and 1.24 eV for SS sample. The second region (II) which is started from about 398 K to ≈ 453 K of both samples show a rapid increase in conductivity of the examined film as a result of increase in temperature and ΔE_a is 0.992 eV and 1.03 eV for CS and SS samples, respectively. It corresponds to the amorphous-crystalline transformation. The third region (III) which is started from about 453 K to ≈ 473 K explains the $\ln\sigma$ vs. $1/T$ relation in case of acrystalline state where the activation energy for conduction (ΔE_a) decreases to 0.814 eV for CS and 0.816 eV for SS samples as shown in Table (4).

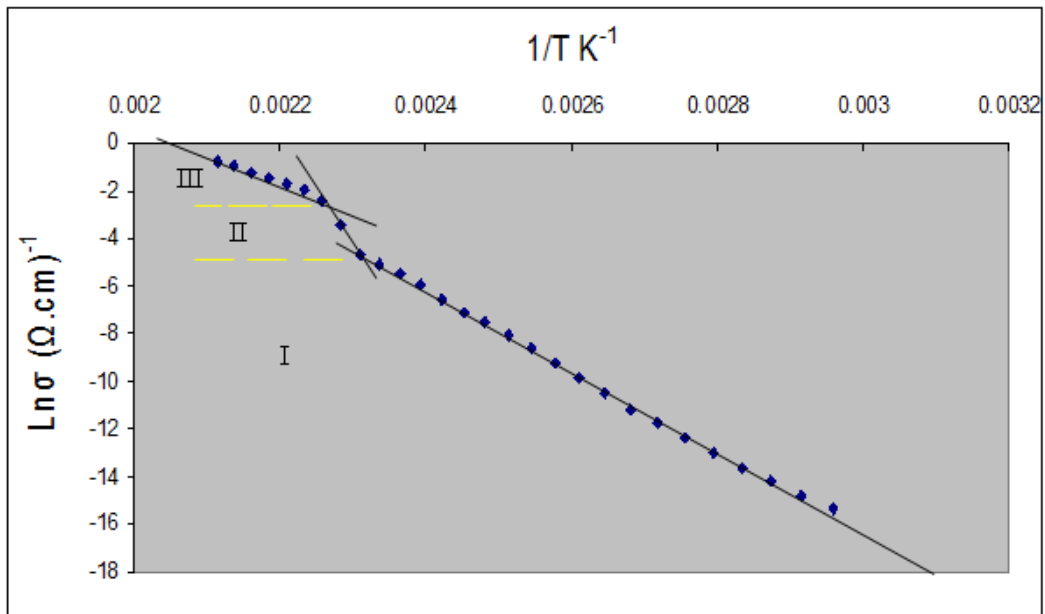


Figure 12: Temperature dependence of DC conductivity for $Te_{61.8}Se_{38.2}$ thin film of (CS sample)

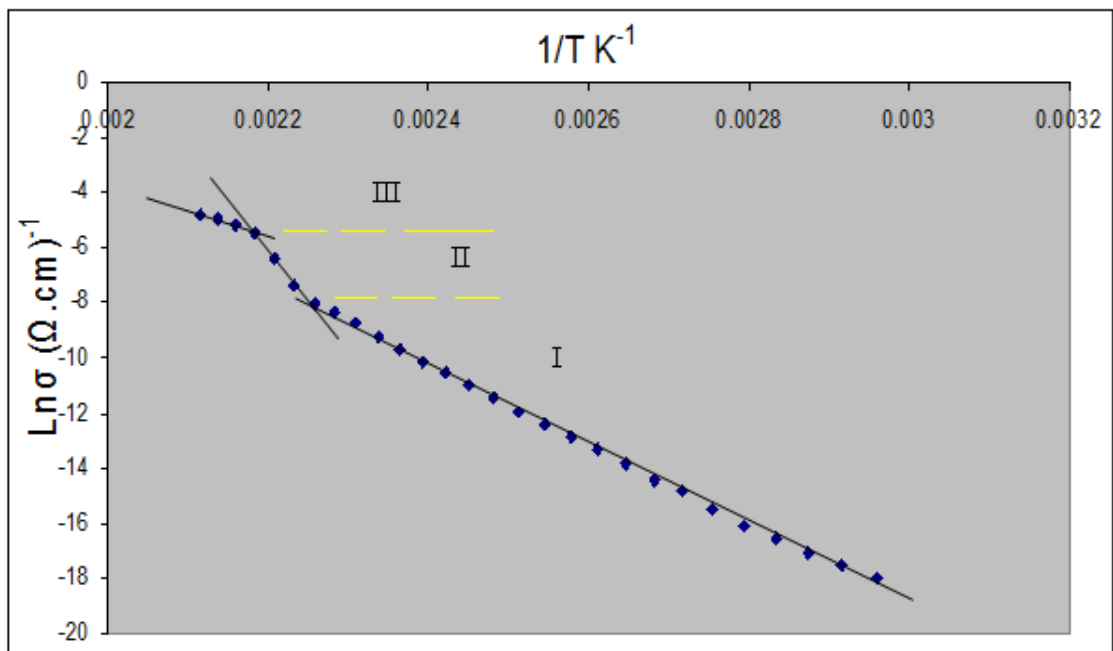


Figure 13: Temperature dependence of DC conductivity for $Te_{61.8}Se_{38.2}$ thin film of (SS sample)

By heating the as-deposited films (CS and SS) from room temperature to $T=463$ K and keeping the high temperature constant for one hour changes the three regions curve of both samples into a single straight line with activation energy equals 0.575 eV for CS and 0.61 eV for SS sample, as shown in Fig. 14 and 15, respectively. In general, the conductivity of the annealed film becomes higher than that

of the as-prepare done. As shown in Table (4), the measurements of DC- electrical conductivity show that the conductivity increases with increasing temperature but the activation energy is decreasing, activation energy ΔE_a gradually decreases with corresponding gradual increase in conductivity σ [3,13].

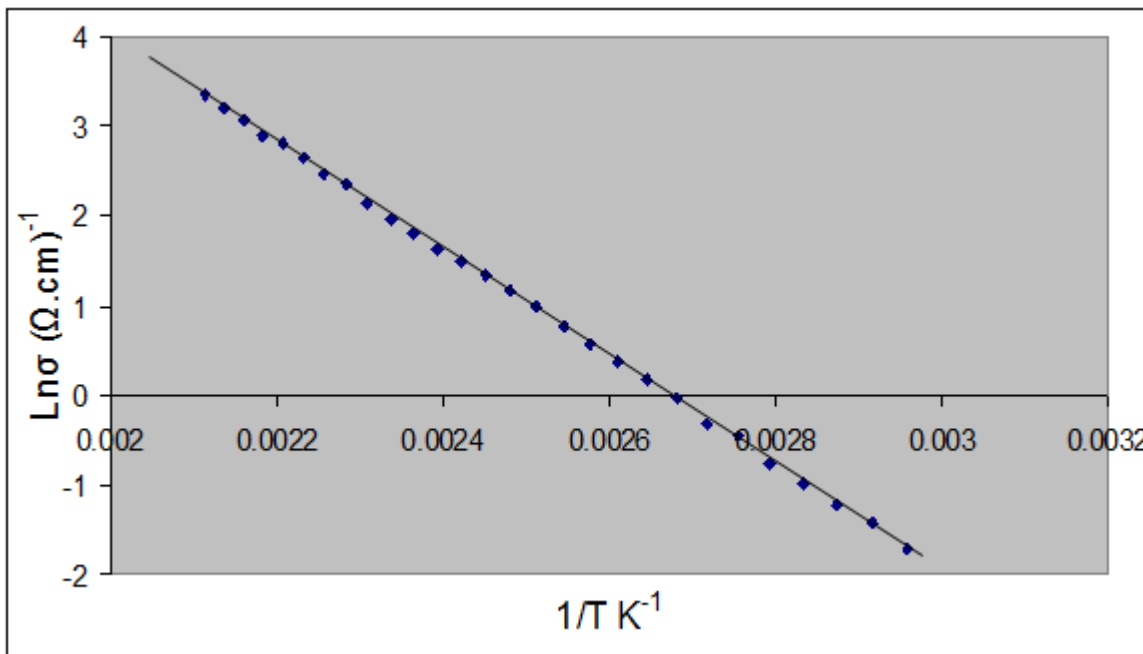


Figure 14: DC conductivity of the CS thin films after annealed at 463 K (More Crystalline)($t=415$ nm, $\Delta E_a=0.575$ ev)

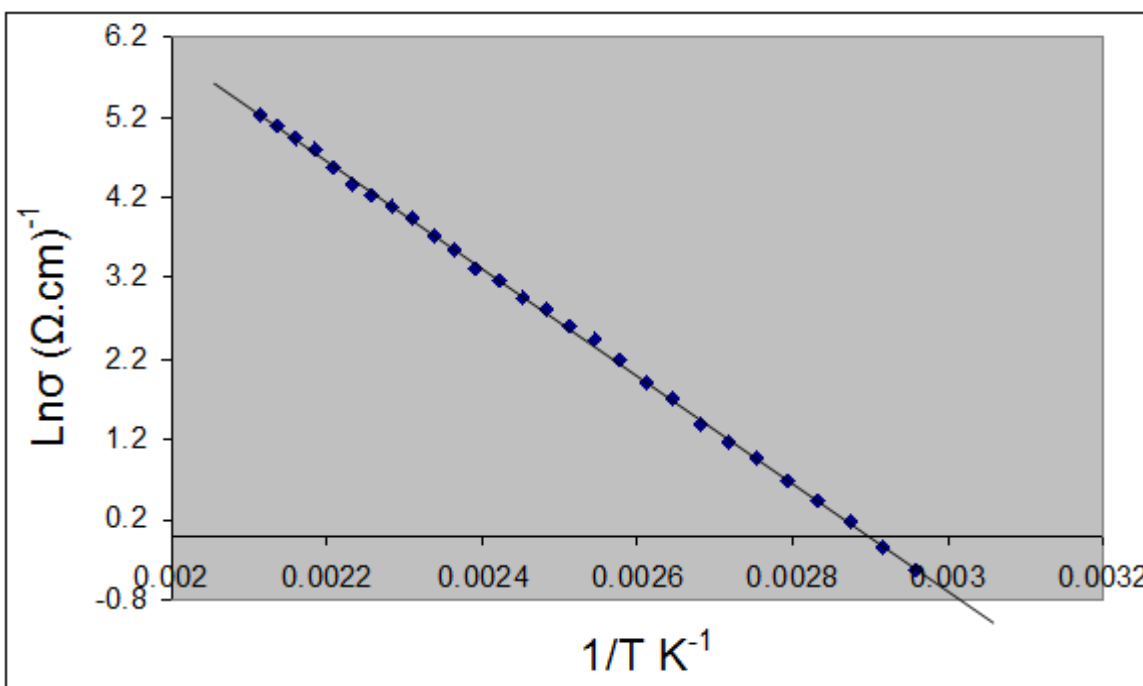


Figure 15: DC conductivity of the SS thin films after annealed at 463 K($t=340$ nm)($\Delta E_a=0.61$ ev)(More Crystalline)

The conductivity σ and activation energy ΔE_a for CS and SS samples have been correlated with that of the structural characteristics. It is clear from the behavior of conductivity and values of activation energies how the structure

approaches the more crystalline form by annealing which improved by the structural study as mentioned before [13,15].

Table 4: Activation energy of CS and SS sample at as deposited and annealed in this study and those found by previous studies

CS as deposit		SS as deposit		CS annealed at 463 K	SS annealed at 463 K	Previous ΔE_a (eV)
T (K)	ΔE_a (eV)	T (K)	ΔE_a (eV)	ΔE_a (eV)	ΔE_a (eV)	
338 to 398	1.17	338 to 398	1.24	0.575	0.61	1.4 as deposit
398 to 453	0.992	398 to 453	1.03			0.6 annealed at
453 to 473	0.814	453 to 473	0.816			453K [13]

4. Conclusions

In summary, we have demonstrated that the alloying system which was considered in this work is a very suitable system to alloy binary. The results indicate that the position of thin film substrate has a significant role in the thin film characteristics, such as thickness and electrical properties (activation energy). Furthermore, we have investigated the electrical properties of $Te_{61.8}Se_{38.2}$ thin film deposited on glass by thermal evaporation technique. The electrical properties for (CS and SS) samples are affected by annealing. Therefore, the samples CS and SS, which are annealed at 463K, were found to be more crystalline and activation energy decreased. The activation energy for CS (thickness 415 nm) and SS (thickness 340 nm) is 0.575eV and 0.61eV, respectively.

5. Future Scope

In this paper we study the preparation of $Te_{61.8}Se_{38.2}$ alloy (by weight) and thin film of $Te_{61.8}Se_{38.2}$. Additionally, we have investigated the temperature dependence of the electrical conductivity of the as-prepared $Te_{61.8}Se_{38.2}$ specimen, films of 415 nm thickness (CS sample) and 340 nm (SS sample). The future scope of annealing of these samples is to transfer thin film to crystalline state and decrease the activation energy for conduction (ΔE_a). As a result, the conductivity of the annealed film becomes higher than that of the as-deposited one. In addition, the future scope of this work is to produce thin film greater hardness, higher photosensitivity, higher crystallization temperature, and lower ageing effects as compared with pure amorphous Se.

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