The Effect of the (Se) percentage on Compositional, Morphological and structural properties of $Bi_2(Te_{1-x}Se_x)_3$ thin films

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Abstract

Alloys of $Bi_2[Te_{1-x} Se_x]_3$ were prepared by melting technique with different values of Se percentage (x=0,0.1,0.3,0.5,0.7,0.9 and 1). Thin films of these alloys were prepared by using thermal evaporation technique under vacuum of 10^{-5} Torr on glass substrates, deposited at room temperature with a deposition rate (12nm/min) and a constant thickness (450±30 nm).
The concentrations of the initial elements Bi, Te and Se in the $Bi_2 [Te_{1-x} Se_x]_3$ alloys with different values of Se percentage (x), were determined by XRF,The morphological and structural properties were determined by AFM and XRD techniques.
AFM images of $Bi_2[Te_{1-x} Se_x]_3$ thin films show that the average diameter and the average surface roughness increase with the increase of the percentage of Se. The X–ray diffraction measurements for bulk and thin films of $Bi_2[Te_{1-x}Se_x]_3$ have polycrystalline structure with rohmbohedral structure, with space group R^3m, and a strong (015) preferred orientation, the crystallite increase with the increase of Se percentage .

Keywords : Bismuth Selenide, Cadmium Selenide, thermoelectric, thin films, thermal evaporation, structural properties.

Introduction

Scientists and researchers recently focused on the study and expand both clean and renewable energies, and tended to diminish the credence on energy that is produced from fossil fuels (e.g. oil ,gas, petroleum and coal etc). Also because of the negatively effect on the surrounding environment for this type of fuel and in turn on human ,therefore the most frequent type of energy that scientists and researchers had concerned since 1950, was the energy that produced by thermoelectric devices, in which the materials have a thermoelectric properties, this means, materials which are able to make thermal energy to be direct conversion into electrical energy and vice versa[1].

In 1954Goldsmid demonstrated the excellent thermoelectric properties of Bismuth Telluride, attributed mainly to the large mean molecular mass, low melting temperature and partial degeneracy of the conduction and valence bands of this V-VI chalcogenide .Since that, bismuth telluride has been widely studied as a thermoelectric material with a narrow band gap, particularly in the temperature range around 300 K[2,3]. Bi_2Te_3 or Bi_2Se_3 has a rombohedral structure with space group R^3m, and the lattice is stacked in a repeated sequence of five atom layers: Te^1-Bi-Te^2-Bi-Te^1along the c-axis, Te or and (Se) and Bi layers are held together by strong ionic-covalent bonds (Te^1 or\ and Se^1 -Bi and Bi_Te^2 or \and Se^2), while The Te^1 or\and Se^1 bonds between cells are of the Van der Walls type and are extremely weak [4, 5]. Bismuth telluride compounds can be doped as either n- or p-type material by creating either a tellurium-rich composition or a bismuth-rich composition respectively [6].

The aim of study

The aim of this study is to produce thermoelectric $Bi_2(Te_{1-x}Se_x)_3$ thin films using thermal evaporation technique ,and look into the effectiveness of (Se) percentage on the structural properties , in order to use it in the future to make thermoelectric devices, which were very effective to provide a clean renewable energies to ensure a clean surrounding environment .

Experimental Procedure

Alloys of $Bi_2[Te_{1-x} Se_x]_3$ were fabricated by using exact amount of high purity (99.99%) powders of source materials (Bi, Te and Se elements), accordance with their atomic percentages of Se (x=0,0.1,0.3,0.5,0.7,0.9 and 1), Each alloy was put in an evacuated quartz ampoule to vacuum ~10^{-4} Torr , then put them in thermal oven to temperature of 923 K (650 °C) for 6 hours until ensuring homogeneous components and fused with each other, then leave them cooled gradually until they reached room temperature. The alloys were grinded well until they became powder to be manufacture a $Bi_2[Te_{1-x} Se_x]_3$ thin films with thicknesses (450±30 nm) which were deposited at room temperature on corning 7059 microscopic glass substrate by thermal evaporation method under suitable vacuum (10^{-5} Torr). The compositional, morphological and structural properties had been tested by XRF, AFM and XRD techniques.

Results and Discussion

The concentrations of the initial elements Bi, Te and Se in the $Bi_2 [Te_{1-x} Se_x]_3$ alloys with different values of Se percentage (x) where (x=0,0.1,0.3,0.5,0.7,0.9 and 1), were determined by XRF, the results were tabulated in Table (1) ,it can be noticed that the compositional analysis of these alloys are in good stoichiometric percentage compared with the theoretical atomic percentages. The AFM test show increasing of the diameter and the roughness of $Bi_2[Te_{1-x}Se_x]_3$ thin films with the increase of the Se percentage as shown in the figure (1), and this result agree with the results of XRD measurements and also agree with references[7,8]. The results of AFM measurement of $Bi_2[Te_{1-x}Se_x]_3$ thin films are listed in table (2).

XRD technique has been used to calculate the miller coefficients by using the relation between the intensity and 2Ө for all samples of $Bi_2[Te_{1-x}Se_x]_3$ alloys and thin films with different values of

Se percentage (x=0,0.1,0.3,0.5,0.7,0.9 and 1). The XRD patterns of $Bi_2[Te_{1-x}Se_x]_3$ alloys showed a rombohedral structure with space group R^3m, and with a strong (015) preferred orientation ,the observed peaks have been analyzed and indexed using standard pattern Joint Committee on Powder Diffraction Standards (JCPDS) as shown in figure (2) and we noticed the peak (015) moves towards the larger 2Θ when Se percentage increases , agree with references [9,10,11],and that means there is an effect on the structure properties because of the increasing of the Se percentage . The values of the crystalline size determined by Scherrer formula that presented by (1) [12], and we found that the grain size increase with the increase of the Se percentage (table 3).

$$G.S= \frac{0.9\,\lambda}{\beta_{FWHM}\cos\theta} \ldots\ldots\ldots\ldots\ldots (1)$$

Where

β_{FWHM} :Full width at half maximum intensity

Θ: Bragg angle (degree)

λ : wavelength (nm)

The XRD of $Bi_2[Te_{1-x}Se_x]_3$ thin films have a polycrystalline structure with rohmbohedral structure, and it can be noticed that the main peak in all films is (015) as shown in table (3) and the observed peaks have been analyzed and indexed using standard pattern(JCPDS) as shown in figure(4).Also the test is shown by using Scherrer formula, the crystallite is increasing as function of Se percentage, agrees with[13], that's mean the percentage of Se is affected on the structural properties of films as shown in Table (4).

Conclusions

We can summarize the results of the present work as follows:

The compositional analyses of $Bi_2[Te_{1-x}Se_x]_3$ alloys are in good stoichiomertric percentage compared with the theoretical atomic percentage. The AFM measurement shows an increase of the diameter and the roughness of $Bi_2[Te_{1-x}Se_x]_3$ thin films were the increase of (Se) percentage. The structural properties of $Bi_2[Te_{1-x}Se_x]_3$ alloys and thin films which prepared with different x percentages (x=0 , 0.1 ,0.3 ,0.5 ,0.7,0.9 and 1) by thermal evaporation technique, were by XRD technique confirmed that all alloys and thin films have a rhombohedral structure with R^3m space group with a preferred orientation along (015), and we found that the main orientation for all samples was 015 , so the Se percentage affects the structure of the studied alloys and thin films.

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Table (1) The theoretical and experimental atomic percentage of $Bi_2(Te_{1-x}Se_x)_3$ alloys

Alloys	Elements	Atomic percentage %	
		Theoretical	Experimental
Bi_2Te_3	Bi	40	42.6
	Te	60	57.4
$Bi_2(Te_{0.9}Se_{0.1})_3$	Bi	40	41.9
	Te	50	47.2
	Se	10	11.8
$Bi_2(Te_{0.7}Se_{0.3})_3$	Bi	40	41.4
	Te	40	37.1
	Se	20	21.5
$Bi_2(Te_{0.5}Se_{0.5})_3$	Bi	40	41.08
	Te	30	31.7
	Se	30	27.62
$Bi_2(Te_{0.3}Se_{0.7})_3$	Bi	40	41.1
	Te	20	19.2
	Se	40	39.7
$Bi_2(Te_{0.1}Se_{0.9})_3$	Bi	40	40.9
	Te	10	10.2
	Se	50	48.9
Bi_2Se_3	Bi	40	40.02
	Se	60	59.88

Table (2) The AFM results showing the grain size and the roughness of $Bi_2(Te_{1-x}Se_x)_3$ thin films

X	D (nm)	Roughness
0	74	0.375
0.1	75.65	0.4
0.3	79.08	0.434
0.5	80	1.96
0.7	82	2.02
0.9	92.96	2.19
1	102.63	5.48

Table (3) The parameter of 2Θ ,hkl, the interplaner of the crystals d and the grain size G.S (nm) of $Bi_2(Te_{1-x}Se_x)_3$ alloys

X	hkl	2Θ(exp)(degree)	d value (stand.)(A°)	d value (exp) (A°)	I/I∘(a.u)
0	0015	44.57	2.07	2.031	45%
	015	28.22	3.205	3.220	100%
	006	17.51	4.777	5.060	45%
0.1	015	28.32	3.12	3.230	100%
	0015	44.6	3.12	3.132	85%
	006	17.52	2.31	2.326	42%
0.3	015	28.62	2.562	2.829	100%
	006	38.56	1.283	1.268	32%
	0015	50.40	1.808	1.808	45%
0.5	006	18.23	4.85	4.783	13%
	015	28.78	3.074	3.045	85%
	0015	45.26	2.133	2.242	60%
0.7	006	17.4	5.1	4.728	40%
	0015	47.72	2	1.903	45%
	015	28.9	3.22	3.024	100%
0.9	006	18.53	3.74	4.783	62%
	015	29.03	3.237	3.044	87%
	0015	46.17	2.19	2.242	50%
1	006	18.75	3.205	4.728	60%
	0015	47.72	3.599	1.903	58%
	015	29.51	3.183	3.024	100%

Table (4) the parameter of 2Ө, hkl, the interplaner of the crystals d and the grain size G.S (nm) of $Bi_2(Te_{1-x}Se_x)_3$ thin films

X	hkl	2Ө	d_{XRD}	D_{ASTM}	G.S (nm)	I/I◦ (a.u)
0	015	26.5833	3.350	**3.222**	**9.8**	**100%**
	1010	34.3722	3.649			**20%**
	101	21.5087	4.128			**10%**
0.1	015	27. 2	3.249	**3.074**	**10.7**	**100%**
	1010	38.1402	2.357			**40%**
	101	22.12	4.128			**42%**
0.3	015	28.02	3.182	**3.074**	**12.8**	**100%**
	1010	38.56	4.575			**45%**
	101	23.13	3.842			**40%**
0.5	015	28.1059	3.172	**3.074**	**10.2**	**80%**
	101	23.2672	3.267			**30%**
	1010	39.4093	2.284			**25%**
0.7	101	21.46	3.235	3.047	11	70%
	015	28.6751	3.110			100%
	1010	40.0139	2.251			60%
0.9	015	29.16	3.06	**3.074**	**11.7**	**100%**
	101	22.86	3.21			**40%**
	1010	40.3536	2.23			**30%**
1	1010	47.311	3.26	**3.205**	**20.4**	**20%**
	101	22.5585	3.94			**80%**
	015	29.2462	3.16			**100%**

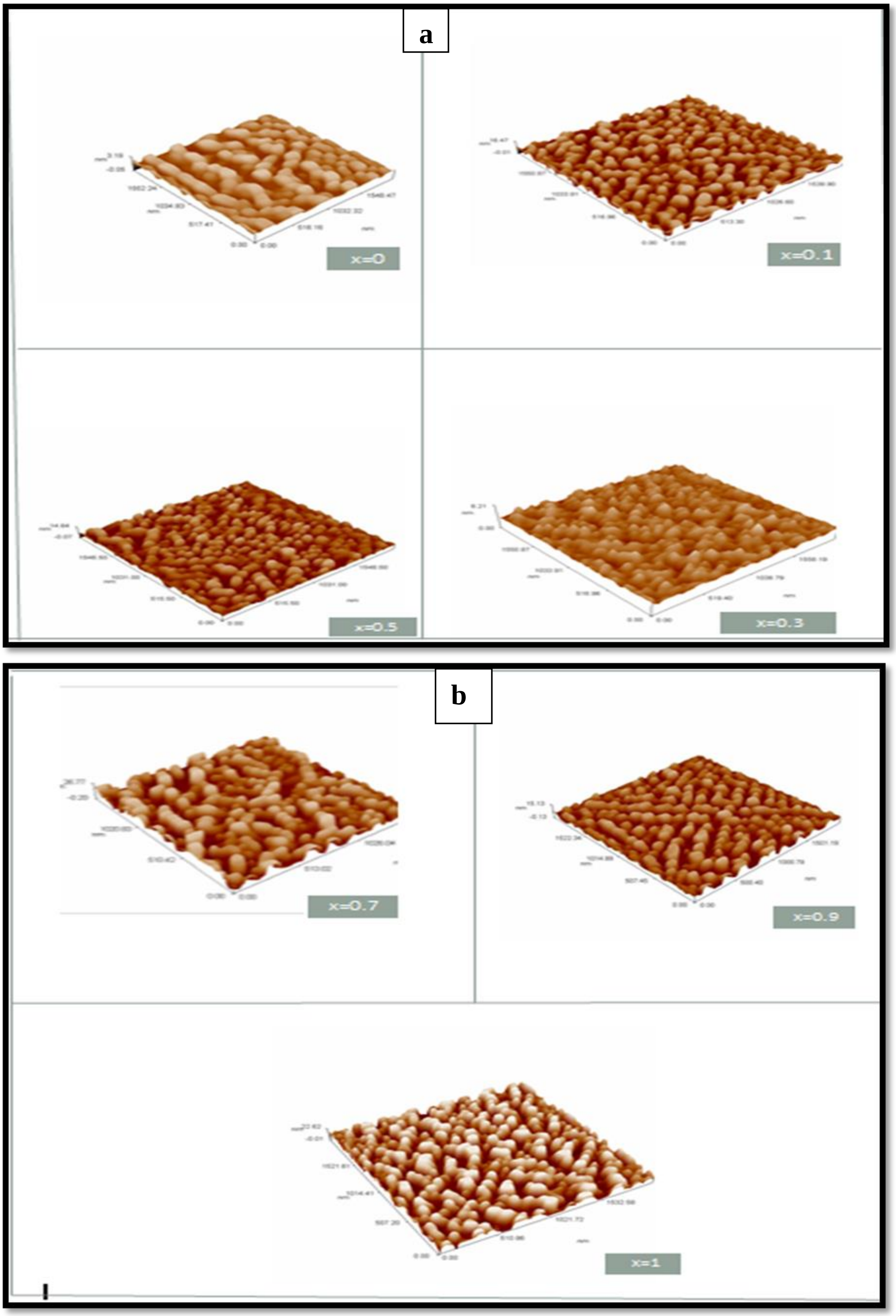

Figure (1) AFM pattern of $Bi_2(Te_{1-x}Se_x)_3$ thin films with different (Se) percentages. a) x= 0,0.1,0.3,0.5 and b) x= 0.7,0.9,1.0.

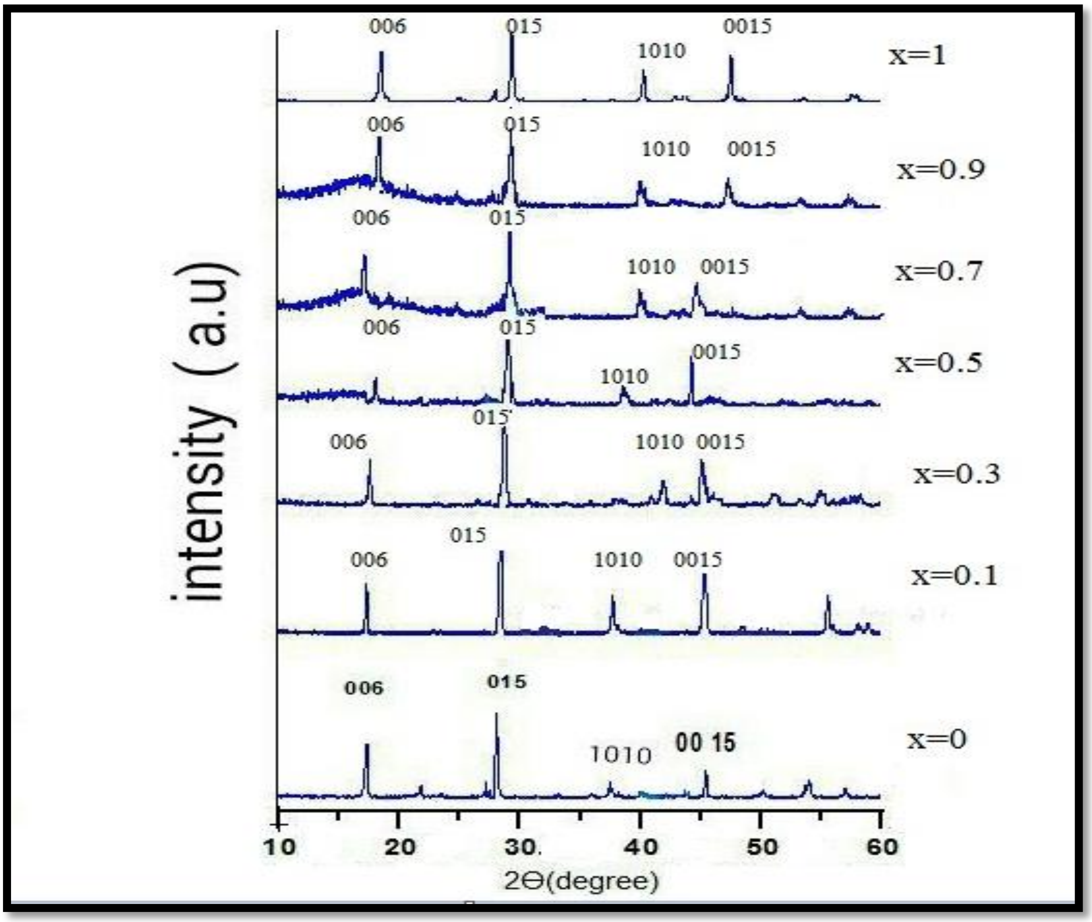

Figure (2) XRD patterns for $Bi_2(Te_{1-x}Se_x)_3$ alloys with different (Se) percentage.

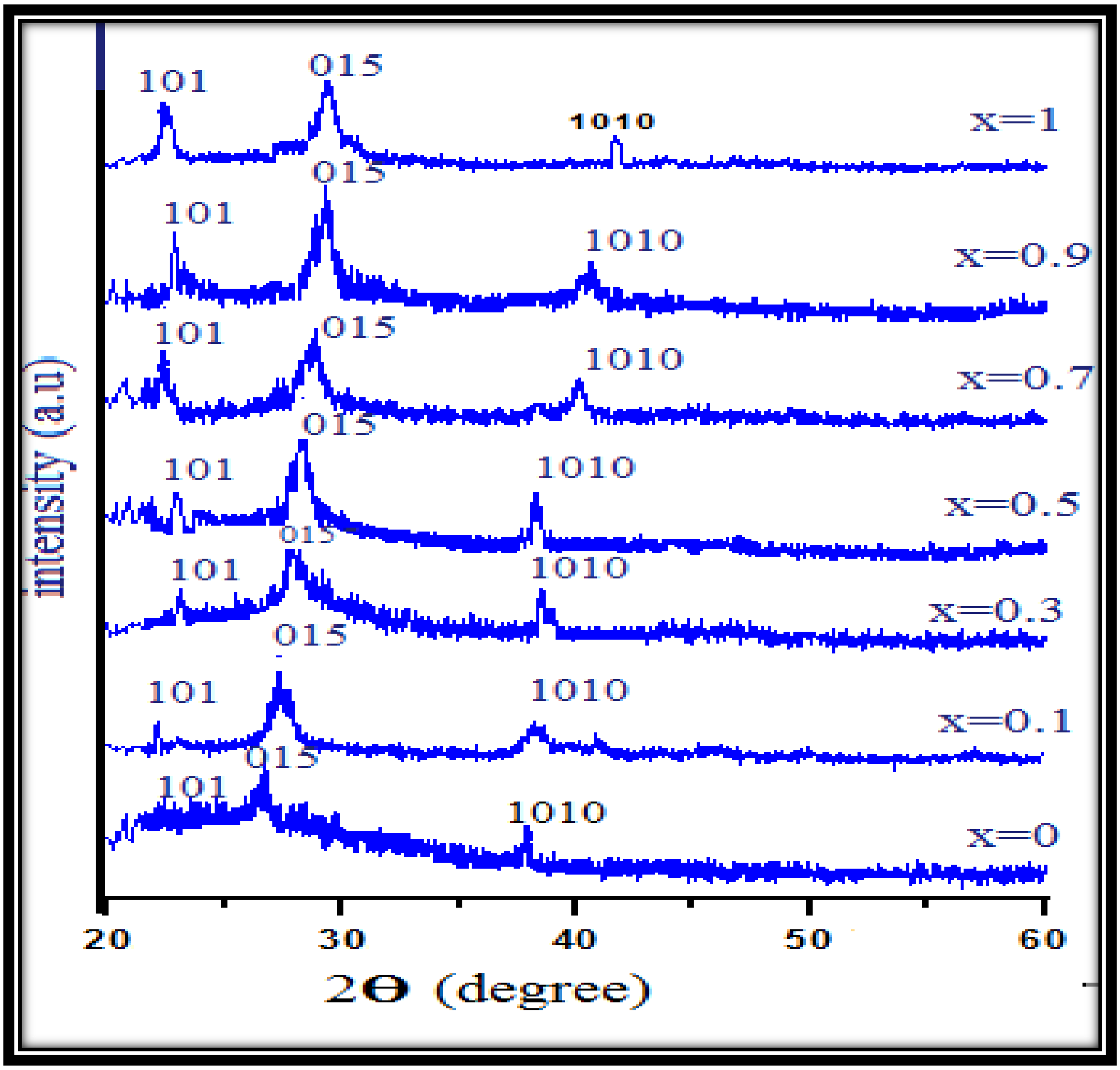

Figure (3) XRD patterns of $Bi_2(Te_{1-x}Se_x)_3$ thin films with different (Se) percentages

تأثير نسبة (Se) على الخواص السطحية والتركيبية لاغشية $Bi_2(Te_{1-x}Se_x)_3$ الرقيقة

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الخلاصة

تم تحضير سبائك المركب $Bi_2(Te_{1-x}Se_x)_3$ بعملية صهر المواد الاولية وفق نسب مختلفة لنسبة السيلينيوم (x=0 , 0.1 ,0.3 ,0.5 ,0.7,0.9 and 1).
الاغشية الرقيقة لهذا المركب تم تحضيرها باستخدام تقنية التبخير الحراري تحت ضغط فراغ يصل الى (10^{-5} torr) حيث تم ترسيب المادة على قواعد زجاجية نظيفة في درجة حرارة الغرفة بمعدل ترسيب(12nm/min) بسمك (450±30 nm.) .
قد تم التأكد من نسب المواد الأولية (Bi ,Te and Se) باستخدام جهاز فلورة الاشعة السينية XRF، اما الخواص التركيبية للسبائك والاغشية الرقيقة لمركب $Bi_2(Te_{1-x}Se_x)_3$ باختلاف نسبة السيلينوم فيها بواسطة جهاز حيود الاشعة السينية XRDاما الخواص السطحية للاغشية الرقيقة تم فحصها باستعمال جهاز AFM وقد وجد ان اقطار الحبيبات تزداد بزيادة نسبة السيلينيوم فيها وكذلك يزداد معدل الخشونة فيها ان قياس حيود الاشعة السينية وجد ان السبائك والاغشية الرقيقة لها تركيب متعدد التبلور ذو تركيب سداسي وفسحة مجموعةR^3m وكانت القمة السائدة والاقوى هي (015) وكذلك بينت ان حجم البلوريات يزداد بزيادة نسبة السيلينيوم في المركب .

الكلمات المفتاحية : بزموث تيلورايد ، بزموث سيلنايد، خواص كهرو حرارية ،اغشية رقيقة، التبخير الحراري، وخواص تركيبية .