ROENTGENOGRAPHIC INVESTIGATION OF Bi_2Te_3-Bi_2Se_3 <Tb> AND Bi_2Te_3-Bi_2Se_3 <Cl> FILMS

N.M. ABDULLAYEV
H.M. Abdullayev Institute of Physics of NAS of Azerbaijan
H. Javid ave., 33, Baku, AZ-1143, Azerbaijan

The real accretion of microparticle sizes, any structural reconstructions, formation of new phases don't take place at annealing of Bi_2Te_3-Bi_2Se_3 <Tb> and Bi_2Te_3-Bi_2Se_3 up to ~600K, the polycrystalline structure is observed on X-ray pattern. The calculations of interplanar spacings show that structure becomes more strength at film annealing at temperature ~600K.

Introduction

Nowadays the alternative power engineering shows the interest in film thermo-electric generators, and also the demand to infra-red radiation detectors has increased. The thermoelements or thermobatteries, covered on the substrates by the method of vacuum condensation with the use of masks and photolithography [1].

It is known, that monocrystalline films with complex technology of their preparation loose their quality during exploitation in time. The change of scattering parameter clearly reveals in small-grained films $Bi_2Te_{2,1}Se_{0,9}$ evaporated on amorphous substrate [1] in comparison with scattering which is character for these compositions in volume crystals r =0 (scattering on acoustic photons). The amorphous films have some understated characteristics than polycrystalline ones.

The substrate temperature should be in optimal limits for obtaining of qualitative film. Roentgenographic investigation of thin films had been carrying out by Frankombe and Semiletov. The temperature of (glass) substrate at which the film is formed corresponding to the composition Bi_2Te_3, has been defined at film investigation. The strong fluctuation from Bi_2Te_3 composition begins at increase t_{sub} higher than 200^0C [2]. Too low substrate temperature opposes to uniform distribution of adsorbed atoms; they group into "islands" of different thicknesses. Vice versa, too high substrate temperature leads to the separation of already settled atoms, their re-evaporation [3].

The task of X-ray investigation of system of polycrystalline films Bi_2Te_3-Bi_2Se_3 <Tb> and Bi_2Te_3-Bi_2Se_3 <Cl> obtained by thermal spraying in vacuum by the method of hot wall, has been proposed by us with the aim of improvement of physical thermoelement characteristics because of decrease of their geometric sizes, with transition on polycrystalline films, evaporated on amorphous substrate.

The given work is dedicated to X-ray investigation method of obtained polycrystalline films of bismuth telluride with terbium and chlorine up to and after annealing.

Investigation method

The investigated films Bi_2Te_3 Bi_2Te_3-Bi_2Se_3 <Tb> and Bi_2Te_3-Bi_2Se_3 <Cl> with optimal thickness 0,30 mcm prepared by evaporation of synthesized substances in installation ВУП-4 in vacuum ~10^{-4}Pa on preliminarily pre-heated NaCl crystals and glass, had been grown by the method of hot wall [1,4]. The more success conditions for steam condensation are formed on substrate, the partial steam condensation on cap walls is led to the minimum by additionally pre-heated wall, where wall temperature at spraying is 800K, substrate temperature is~ 600K at precipitation rate of thin layers is ~2nm/sec [5].

The obtained polycrystalline films Bi_2Te_3-Bi_2Se_3 <Tb> and Bi_2Te_3-Bi_2Se_3 <Cl> given below have been investigated by rontgenography method.

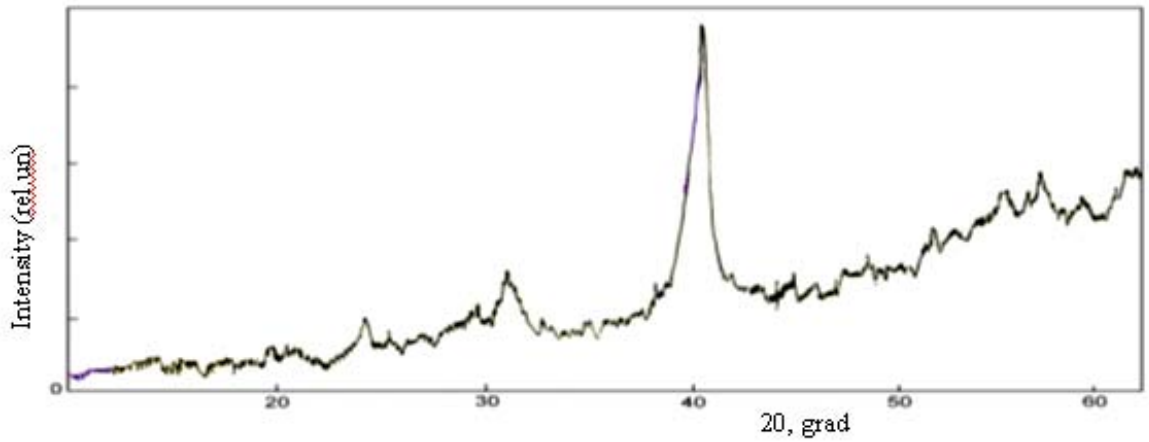

Fig.1. Roentgenogram of unannealed Bi_2Te_3-Bi_2Se_3<Tb> film.

The film samples suitable for roentgenographic investigations by thickness 30 nm are prepared by sublimation of synthesized compound of Bi_2Te_3 and Bi_2Te_3-Bi_2Se_3 <Tb> composition, i.e. $(Bi_2Te_3$-$Bi_2Se_3)_{1-x}Tb_x$ (x=0,15) and Bi_2Te_3-Bi_2Se_3 <Cl> on freshly cleaved bounds of haloid crystal NaCl and glass substrates.

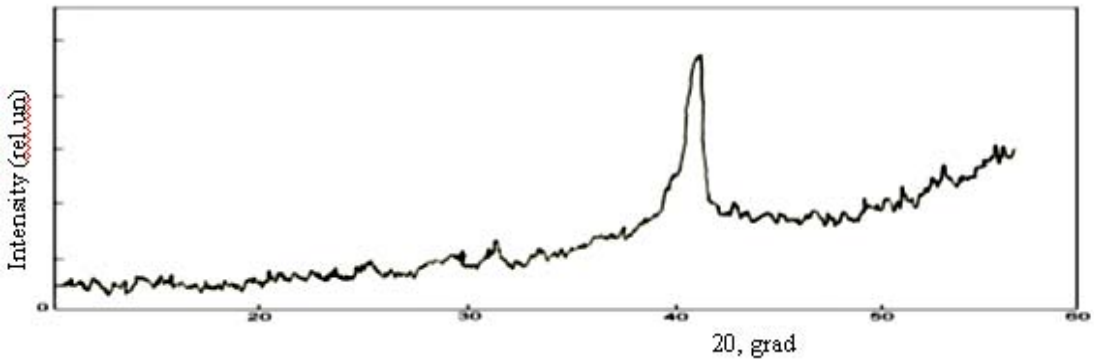

Fig. 2. Roentgenogram of unannealed Bi_2Te_3-Bi_2Se_3<Cl> film.

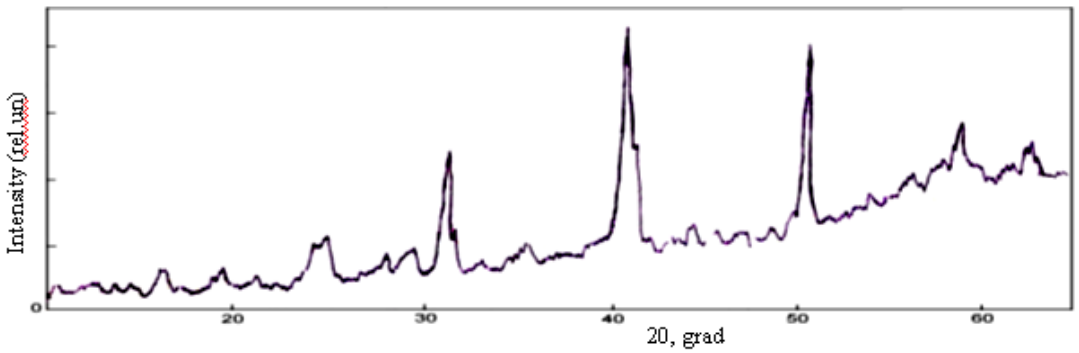

Fig. 3. Roentgenogram of annealed Bi_2Te_3-Bi_2Se_3<Tb> film.

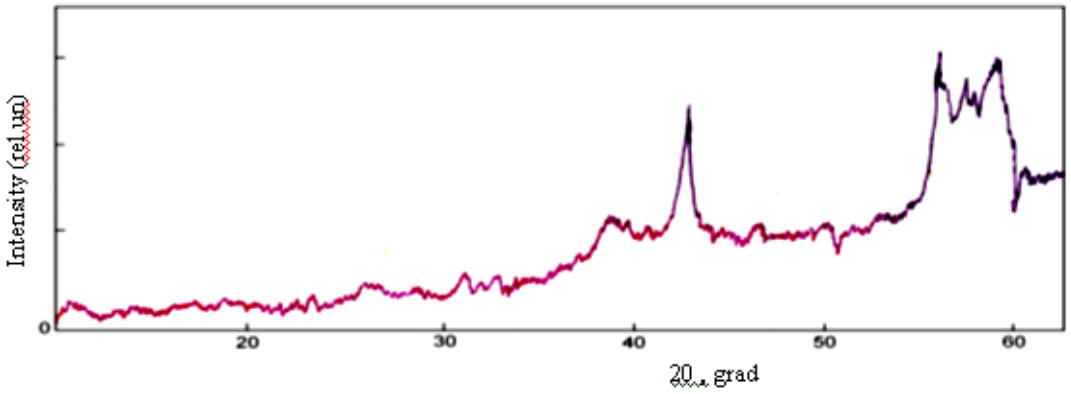

Fig. 4. Roentgenogram of annealed Bi_2Te_3-Bi_2Se_3<Cl> film.

The obtained samples are treated by roentgenographic analysis on DRON-2,0 (CuK_α is radiation, Ni is filter) at the mode 35kV, 10mA. 10, 13, 15, 16 clear diffraction reflections for Bi_2Te_3 analogues have been fixed in the limits $5^o \leq 2\theta \leq 70^o$ correspondingly. The obtained diffraction patterns are almost identical ones with small difference of reflex intensity and angles of reflection. The film roentgenogram of Bi_2Te_3-Bi_2Se_3 <Tb> and Bi_2Te_3-Bi_2Se_3 <Cl> polycrystals before and after annealing are well indicated on the base of Bi_2Te_3 hexagonal lattice [6].

Results

The thermo-annealing influence at ~600 on the film structure and properties, precipitated on the glass has been investigated. The films, precipitated on the glass, are treated by annealing in the vacuum for the elimination of elastic stress fields. The annealing with duration 24 hours with temperature decrease in 25 grad/min is carried out in vacuum ~10^{-4}Pa.

Table 1.

Roentgenographic investigation of Bi_2Te_3-Bi_2Se_3<Tb> and Bi_2Te_3-Bi_2Se_3<Cl> films.

№	d_{sc}, p-tip	d_{exp}, p-tip	I/I_0	Hkl	d_{exp}, n-tip	I/I_0
1	5,050	5,415	2	222	5,523	1
2	3,770	3,849	10	110	3,889	5
3	3,210	3,292	1	221	3,366	1
4	-	2,716	10	332	3,029	10
5	-	2,593	10	444	2,869	7
6	2,370	2,450	20	433	2,629	10
7	2,230	2,344	2	443	2,469	7
8	2,190	2,215	6	011	2,230	5
9	2,030	2,110	9	555; 231; 544	-	-
10	1,996	2,007	5	554; 11 1;200	-	-
11	1,809	1,868	2	220; 342; 311	1,926	10
12	-	1,802	17	331	1,819	10
13	1,696	1,755	15	665; 442	-	-
14	1,608	1,611	20	453	1,637	10
15	1,486	1,526	20	665 442	1,551	10
16	1,450	1,458	10	533	1,492	5

The roentgenogram obtained at the radiation of polycrystalline films Bi_2Te_3-Bi_2Se_3 <Tb> and Bi_2Te_3-Bi_2Se_3 <Cl> are well indicated on the base of hexagonal lattice of Bi_2Te_3 polycrystal (a=0,43835, c=3,0487nm; sp.gr. D_{3d}^5, R_{3m}, $Z=3$) and satisfies data [7]. The all reflexes having strong and average intensities which are character for the given structure are observed on roentgenogram. The results of calculated hkl, I/I_0 and experimental interplanar spacings d_{exp} in the films Bi_2Te_3-Bi_2Se_3 <Tb> of p-type and Bi_2Te_3-Bi_2Se_3 <Cl> of n-type in comparison with science data d_{sc} are given in the table 1 [7-8].

The analysis of obtained data and calculated values show on the fact that compositions of annealed films Bi_2Te_3-Bi_2Se_3<Tb> and Bi_2Te_3-Bi_2Se_3<Cl> form the isostructure on the base of hexagonal structure of bismuth telluride chalcogenide.

Conclusion

On the base of X-ray investigation of the thin film structures obtained by thermal spraying on the glass, it has been established that polycrystalline films of Bi_2Te_3-Bi_2Se_3<Tb> and Bi_2Te_3 - Bi_2Se_3<Cl> compositions by thickness 0,30 mcm form at substrate temperature ~600K. Therefore, the real increase of microparticle sizes, any structural reconstructions, formations of new phases aren't observed at annealing of Bi_2Te_3-Bi_2Se_3<Tb> and Bi_2Te_3 - Bi_2Se_3<Cl> up to ~600K, the polycrystalline structure are observed on roentgenogram. The calculations of interplanar spacings show that structure becomes more strength at film annealing at temperature ~600K.

The author is thankful to G.G. Huseynov for help in the carrying out of investigations.

[1] *N.S. Lidorenko.* Plyonochniye termoelementi: fizika i primeneniye. M., Nauka, 1985, 3, 7, 179, 199.(in Russian)

[2] *B.M. Golcman, V.A. Kudinov, I.A. Smirnov*. Poluprovodnikoviye termoelektricheskiye materiali na osnove Bi_2Te_3. M., Nauka, 1972, 18, 302. (in Russian)

[3] *S.I.Mehdiyeva, N.Z.Jalilov, N.M.Abdullayev, N.R.Memmedov, M.I.Veliyev, V.Z.Zeynalov.* TPE-06, 3-rd Intern.Conf. on Techn.&Phys.Probl. in Pow.Engin., Ankara, Turkey, May 29-31, 2006, 695.

[4] *S.I. Mekhtiyeva, N.Z. Dzhalilov, N.M. Abdullayev, N.R. Memmedov, V.Z. Zeynalov*. Mikrostruktura plyonok Bi_2Te_3-Bi_2Se_3 legirovannikh. AMEA, Xeberler, XXVII, № 2, Bakı, 2007, 148. (in Russian)

[5] *A.G. Abdullayev, E.I. Veliyulin, S.Sh. Kakhramanov*. Vliyaniye legirovanya i interkalirovanya na svoystva khalkogenidov vismuta. Baku, 1991. (in Russian)

[6] *D.I. Ismailov, G.M. Akhmedov, R.Sh. Shafizade*. Dokl. ANAzerb.SSR, 45, №4, 1998, 6-8. (in Russian)

[7] Spravochnik. Minerali. T. I, Izd. AN SSSR, M. 1960, 573. (in Russian)

[8] *S.S. Tolkachev*. Tablici mejploskostnikh rasstoyaniy. Izd. «Khimiya» Leningradskoye otd.. 1968, 78.

N.M. Abdullayev

Bi_2Te_3-Bi_2Se_3 <Tb> və Bi_2Te_3-Bi_2Se_3 <Cl> TƏBƏQƏLƏRİN RENTGENOGRAFİK TƏDQİQİ

Bi_2Te_3-Bi_2Se_3<Tb> və Bi_2Te_3-Bi_2Se_3<Cl> nazik təbəqələri ~600K-dək qızdırdıqda yeni fazaların əmələ gəlməsi və hər hansı bir struktur dəyişikliyi, mikrohissəciklərin ölçülərində real böyüməsi baş vermir.

Н.М. Абдуллаев

РЕНГЕНОГРАФИЧЕСКОЕ ИССЛЕДОВАНИЕ ПЛЁНОК Bi_2Te_3-Bi_2Se_3 <Tb> и Bi_2Te_3-Bi_2Se_3 <Cl>

При отжиге пленок Bi_2Te_3-Bi_2Se_3<Tb> и Bi_2Te_3-Bi_2Se_3<Cl> до ~600К не происходит реального увеличения размера микрочастиц, каких либо структурных перестроек, образование новых фаз: на рентгенограмме наблюдается поликристаллическая структура. Расчёты межплоскостных расстояний показывают, что при отжиге пленок структура упрочняется.

Received: 01.07.08