EFFECTS ANNEALING TREATMENT ON THERMOELECTRICITY OF LEAD TELLURIDE THIN FILMS ON Si-WAFER PREPARED BY DC MAGNETRON SPUTTERING SYSTEM

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ABSTRACT

Lead Telluride (PbTe) thin films on Si-wafer were deposited by DC magnetron sputtering system. The target was prepared from a 99.99 % impurity of Pb : Te; 1 : 1 mixed powders and then pressured into copper supporting cap at 250 MPa. The deposition process was under of Ar atmosphere in the vacuum chamber and deposition time of 30 min. As-deposited film samples were annealed at temperatures of 423 and 523 K for 60 min under a vacuum state. Phase identification, atomic composition, morphology and film thickness (d), electrical resistivity (ρ) and Seebeck coefficient (S) of as-deposited and annealed PbTe thin films were investigated and measured. This results demonstrated the as-deposited and annealed thin films samples were uniformed of crystalline phase and exhibited single phase of face-center cubic (FCC) with the annealing temperature at 523 K. The film thickness of as-deposited was explained from 460 nm to 610 nm of annealed 423 K and then reduced to 560 nm of annealed 523 K with the affected of annealing treatment. Also, thermoelectricity of PbTe thin films on Si-wafer can been changed from p–type to n–type with the S values of 128, –280 and –320 $\mu V\ K^{-1}$ for as-deposited, annealed 423 K and 523 K, respectively.

KEYWORDS: *PbTe thin film, annealing treatment, thermoelectricity, Si-wafer substrate, DC magnetron sputtering*

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INTRODUCTION

Today, thin film technology is important in manufacturing, productions and research applications such as modern microelectronic, photonic devices and sustainable energy [1,2]. Focused on sustainable energy, more than 80 % of its primary energy needs has been supplied by petroleum oil, natural gas, and coal. But, these energy resources are non-renewable and toxic to the environment with carbon dioxide (CO_2), carbon monoxide, (CO), and other environmentally harmful emissions. Hence, there has been a driving force to look for renewable energy resources that are clean, safe, and reliable long-term.

Thermoelectricity is one of these renewable energy resources. It has been widely investigated and is expected to be feasible in the near future. The performance of thermoelectric materials is considered by the dimensionless figure of merit ZT giving by;

$$ZT = \frac{S^2 T}{\rho\kappa} \quad (1)$$

where S, ρ and κ are the Seebeck coefficient, electrical resistivity, and thermal conductivity of the thermoelectric materials used to fabricate the device, respectively. T is the absolute temperature in K at which the device operates. Higher ZT values lead to thermoelectric devices of improved performance. In other words, good thermoelectric materials, which are defined as materials of high S and low ρ and κ are crucial for the fabrication of high efficiency thermoelectric devices. The highest of ZT about of 1.0 has been achieved from the state of the art thermoelectric material of Bi_2Te_3 and Sb_2Te_3 binary compounds at room temperature [3]. Moreover, other binary compounds are potential candidates for thermoelectric materials because of desirable thermoelectric properties such as: long-period crystal structure [4], low thermal conductivity [5] and electronic structure similar to semi-metallic behavior [6]. For example, chalcogenide materials suggest lead telluride (PbTe) binary compound, as is has resonant bonding and low thermal conductivity suitable for thermoelectric and phase change materials development [7,8]. Also, it can be a both p-type and n-type PbTe-based bulk TE materials, having been reported to reach a ZT between 1.4 and 1.8 [9], which is suitable for thermoelectric application.

In this work, we reported the thermoelectricity of PbTe thin film on Si-wafer substrate prepared by dc magnetron sputtering system included the structural and morphology of film within the affect of annealing treatment.

MATERIALS AND METHODS

PbTe Thin films were prepared by an in-house-built dc magnetron sputtering system. The sputtering target, 60 mm diameter and 3.0 mm thick from Pb powder (99.99 %, QRëC, New Zealand) and Te (99.99 %, QRëC, New Zealand) were mixed in 1:1 ratio and compacted in a copper supporting cap at pressure of 250 MPa. Thin films deposition process used base pressure of 3.2 mT and then applied the argon gas (Ar impurity 99.999%) in vacuum chamber to obtained operating pressure of 50 mT. The sputtering power was applied of 25 W for 30 min. As-deposited film samples were annealed at temperature of 423 K and 523 K for 1 hour in vacuum state. The thin films deposition condition is shown in Table 1.

Table 1. The thin films deposition conditions

Base pressure (mT)	3.2
Operating pressure (mT)	50
DC power (W)	25
Substrate	Si-wafer
Substrate temperature (K)	300
Deposition time (min)	30

The effect of annealing treatment on identifying phases was performed by X-ray diffractometer (XRD-6100, Shimadzu). The thickness, surface morphology and composition of thin films were analyzed by a scanning electron microscope and energy dispersive X-ray (SEM-EDX, JSM-5410, JEOL). The Seebeck coefficient and electrical conductivity of the thin films were measured by the standard steady state and four probe Van der Pauw methods, respectively [10].

RESULTS AND DISCUSSION

The XRD pattern revealed that the target consisted of a majority FCC NaCl-type structure (PDF#008-0028) with relatively good crystallinety as shown in Fig. 1.

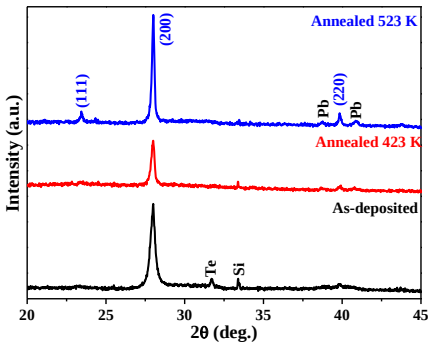

Fig. 1. XRD patterns of PbTe thin films of as-deposited and annealed 423 K and 523 K

From Fig. 1, the effect of annealing treatment has affected to increasing the crystal growth from as deposited with (200) and (220) peaks to (111), (200) and (220) for annealed at temperature 523 K. The crystallite sizes (D) and lattice strains (ε) were evaluated from full widths at half maximum (FWHM) and 2θ angle at high intensity (200) peaks from the Debye–Scherrer's formula as following by;

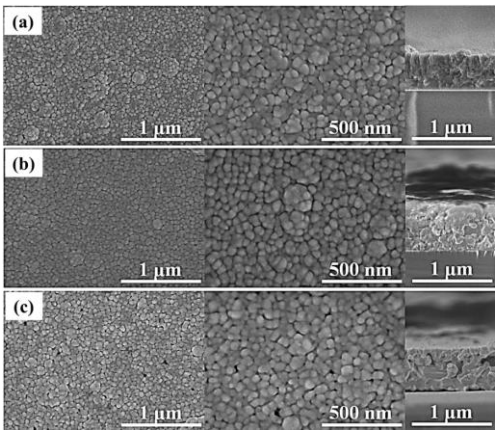

Fig. 2. SEM images of PbTe thin films of (a) as-deposited and annealed (b) 423 K and (c) 523 K

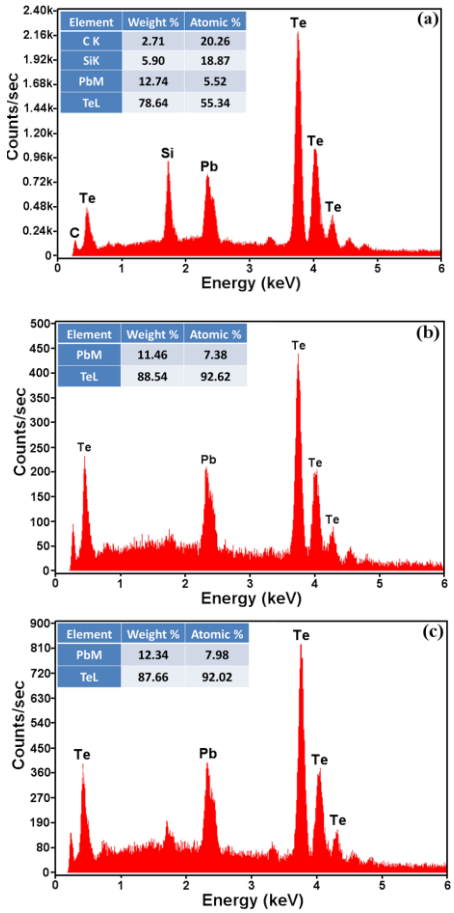

Fig. 3. EDX spectra of PbTe thin films of (a) as-deposited and annealed (b) 423 K and (c) 523 K

$$D = \frac{K\lambda}{\beta \cos\theta} \quad (2)$$

and

$$\varepsilon = \frac{\beta}{4\tan\theta} \quad (3)$$

where K, λ, and θ are the shape factor, wavelength of CuKα radiation, and diffraction angle, respectively. The various annealing temperatures affected to FWHM, grain sizes and lattice strains of the PbTe thin films, as shown in Table 2. From Table 2, the crystal size of the PbTe thin films were decreased with the annealing temperature increasing as confirmed by SEM images as shown in Fig. 2.

Table 2. Grain sizes and lattice strains of the PbTe thin films

Sample	(200) peak (deg.)	FWHM (deg.)	*D* (nm)	ε (%)
Asd.	13.9998	0.2282	35.88	0.40
423 K	13.9976	0.1736	47.17	0.30
523 K	14.0042	0.1103	74.22	0.20

Figure 2 shows the surface topologies and cross sections of the PbTe films. The as-deposited film showed a homogeneous morphology, and perfect adherence to the substrate. The annealed films showed stronger grooving features along the grain boundaries. The surface roughness was increased up to the annealing of 523 K. The crystal growth of PbTe thin films was increased with the temperature annealing function to corresponding XRD pattern preferred orientation and film thickness expansion.

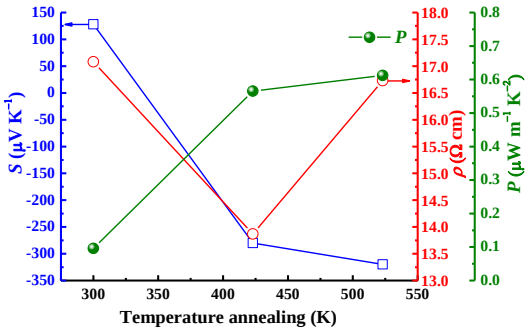

Fig. 4. Seebeck coefficient, electrical resistivity and power factor of PbTe thin films at the annealing temperature functions

Figure 3 shows the composition of PbTe thin films, which was quite different from that of the sputtering target with 1:1. A primary reason the as-deposited film yielded such composition was because of high plasma density, which enabled high sputtering rate of charged and neutral species from

the targets [11,12]. However, more Te atomic composition enhanced Seebeck coefficient into power factor, as shown in Fig. 4.

Figure 4 shows the thermoelectric properties of PbTe thin films comprises Seebeck coefficient, electrical resistivity and power factor, respectively. Seebeck coefficient measurement values demonstrated that the thermoelectricity of PbTe thin films on Si-wafer changed from p–type to n–type due to temperature annealing affected corresponding to [9] reported. Hence, this result indicated that the PbTe material has both hole and electron concentration which it depended on for atomic composition variation.

CONCLUSION

PbTe thin films on Si-wafer can be prepared by DC magnetron sputtering system. The effect of annealing treatment can change the thermoelectricity of PbTe thin films from p–type to n–type. Both electrical resistivity decrease and Seebeck coefficient increase can enhance the power factor of PbTe thin films increase with the temperature annealing increasing.

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