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Synthesis and characterization of nano-sized lead-telluride powder synthesized by mechanical alloying

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Abstract

Lead telluride-based alloys have the best thermoelectric properties for intermediate temperature applications (500–900 K). In this work, nanocrystalline lead telluride powder was synthesized from high-purity elements by mechanical alloying by means of a planetary ball-milling procedure. The milling medium was stainless steel, and the time of the milling was varied in order to investigate the effect on the formation time of the desired phases. Phase transformations and crystallite evolution during ball-milling were followed by powder x-ray diffraction (PXRD). The broadened PXRD peaks were analyzed with the Williamson-Hall method revealing the Nano-sized crystalline size and the introduced stress during the mechanical alloying process.

Keywords: Lead Telluride, Thermoelectric, XRD, Mechanical Alloying, nanocrystalline.

Preferred symposium: Electrical Ceramics

1. Introduction

The need to produce green energy has gained a considerable amount of attention [1]. Thermoelectric technology has great potential in increasing the efficiency of conventional energy conversion systems by directly converting waste heat into electricity [2]. The dimensionless figure of merit is a tool to determine the efficiency of thermoelectric conversion, which is defined by the following relationship:

$$ZT = (S^2\sigma T)/\kappa \qquad \text{(Eq. 1)}$$

where, S is the Seebeck coefficient, σ is the electrical conductivity, T is the absolute temperature, and κ is the total thermal conductivity [3]. Efficient thermoelectric materials should have a high power factor ($S^2\sigma$) and a low thermal conductivity at the same time [4]. The working temperature of thermoelectric materials are categorized as ambient, mid- and high- temperature [5].

PbTe system has proved to have one of the highest ZT values ever reported in the midrange temperature [6]. The conventional method to produce lead chalcogenides is melting of the

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constituent elements in vacuum furnace [7]. The conventional hydrothermal synthesis process for synthesis of the compound is very expensive and time-consuming. Therefore, researchers have tried to come up with new ways to produce PbTe powder more efficiently [8].
Nano-sized powder could be produce from the pure elements using mechanical alloying [4]. MA allows the preparation of homogeneous materials from a powder mixture of the constituent elements. In this process, energy is mechanically introduced by striking the powders in contact with each other. As the process continues, excessive plastic deformation leads to the failure of particles, which by repeated cold welding and failure, the layered structure becomes homogeneous [9]. However, some microscopic defects are produced during the process. The presence of microscopic defects and secondary phases at grain boundaries would cause a low lattice thermal conductivity, which increases the phonon scattering [10][11].

2. Materials and Methods

Elemental powders of Pb and Te were weighed and mixed with a stoichiometric ratio of Pb50Te50. The powders were loaded in a stainless steel container under argon atmosphere in a glovebox. The Ball to powder ratio was 10:1 and a rotational speed of 400 rpm was applied in a PM 2400 planetary ball mill. During the MA process, some samples were taken out in certain time intervals for XRD analysis. XRD Empream device was used for XRD analysis. The crystallite size was measured with the Williamson-Hall method. The particle size measurement was carried out using PSA Cordouan Vasco3 Device.

3. Results and Discussion

3.1. X-ray diffraction pattern and particle size analysis of as-milled powders

X-ray diffraction pattern of the mixed powder before milling is shown in Fig. 1. There is a substantial amount of lead oxide and tellurium oxide in the mixture before milling. The XRD patterns for the as milled powder after 1, 2 and 4 hours of milling are shown in Fig. 2. As reported, the crystalline phases are indexed to PbTe (ICDD: 01-078-1904) with Cubic Fm3m crystal structure [6]. After milling for 1 h most of the Pb and Te peaks are eliminated and some PbTe is produced. After 2 h of milling all elemental powders and tellurium oxide has vanished but there is still a substantial amount of lead oxide. As the milling continues to 4 h, lead oxide is still present but the crystallinity of the produced PbTe has increased due to the higher peak intensity. As shown in Fig. 3 the produced powder has a dark grey color [7].

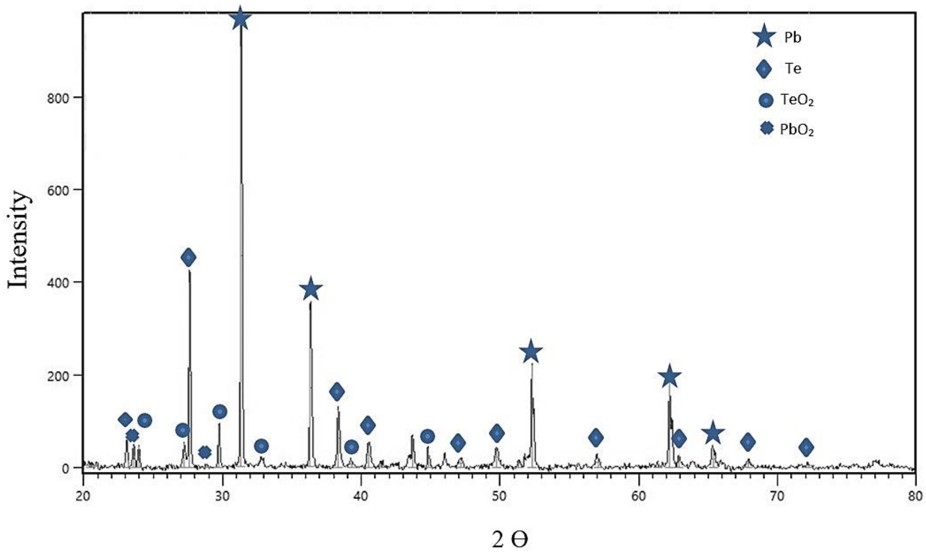

Fig. 1: XRD pattern of Lead and Tellurium powder mixture before milling.

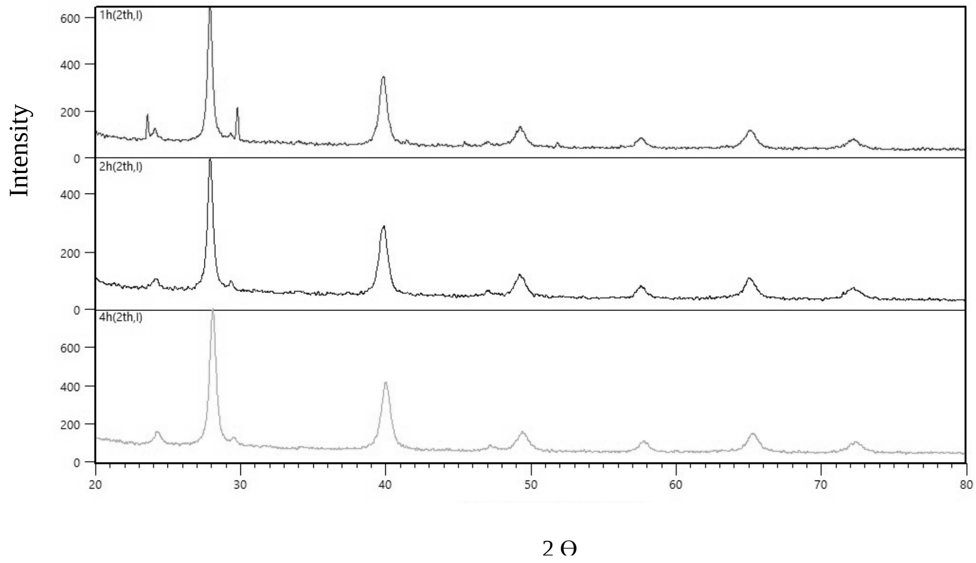

Fig. 2: XRD patterns of the PbTe compound synthesized at different milling times.

Fig. 3: the sythesized Lead telluride powder.

Fig. 4 shows the particle size curve for the as-milled powder. The average size of the particles is less than 10 nm. The crystallite size calculated by the Williamson-hall method is presented in Table 1. The crystallite size is about 30 nm and does not change largely by increasing milling time. Therefore, 4 h milling time is sufficient for producing crystalline PbTe powder although the starting lead oxide would still be present after 4 h.

Table 1. Crystallite size determined by Williamson-Hall method.

Milling time (h)	1	2	4
Crystallite size (nm)	30	35	33

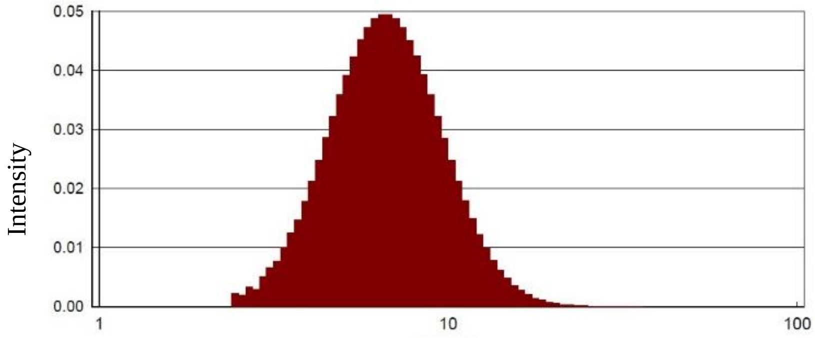

Fig. 4. Cumulative particle size distribution of the as-milled powder.

Conclusion

Lead telluride nano-sized powder was produced by mechanical alloying and the effect of milling time on phase formation and crystallinity was investigated. The synthesized powder was nanocrystalline based on Williamson-Hall method calculations and an average particle size of about 10 nm was achieved based on PSA particle size analysis. XRD result showed that after 2 h of milling the phase formation of PbTe was complete and as the milling time increases the crystallinity of the powder improves due to higher intensity of the peaks.

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References

[1]Ray, P., *Renewable energy and sustainability*. Clean Technologies and Environmental Policy, 2019. **21**(8), 1517-1533.
[2]Rowe, D.M., *Thermoelectrics handbook: macro to nano*. 2018: CRC press.
[3]Vollstedt, A.-M., *Thermoelectric Power Generation and Refrigeration Systems*. 2001.
[4]Aminzare, M., et al., *Effect of single metal doping on the thermoelectric properties of SnTe.* Sustainable energy & fuels, 2019, **3**(1), 251-263.

[5]Twaha, S., et al., *A comprehensive review of thermoelectric technology: Materials, applications, modelling and performance improvement.* Renewable and sustainable energy reviews, 2016, **65**, 698-726.

[6]N. Bouad1, R.M. Marin-Ayral, J.C. Te´denac,” Mechanical alloying and sintering of lead telluride”, Journal of Alloys and Compounds, 297, 2000, 312–318.

[7]He, J., et al., *On the origin of increased phonon scattering in nanostructured PbTe based thermoelectric materials.* Journal of the American Chemical Society, 2010, **132**(25), 8669-8675.

[8] N. Bouada, R.M. Marin-Ayrala , G. Nabiasa,b ,*, J.C. Te´denaca,” P hase transformation study of Pb–Te powders during mechanical alloying”, Journal of Alloys and Compounds 353, 2003, 184–188.

[9]Benjamin, J.S. *Fundamentals of mechanical alloying*. Trans Tech Publ.

[10]Nolas, G.S., J. Sharp, and J. Goldsmid, *Thermoelectrics: basic principles and new materials developments*. Vol. 45. 2001: Springer Science & Business Media.

[11]Shevelkov, A.V., Chemical aspects of the design of thermoelectric materials. Russian Chemical Reviews, 2008. 77(1), p. 1.