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Transport properties of $Ag_{0.8}Pb_{18}SbTe_{20}$ prepared by high pressure and high temperature

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ABSTRACT

In this paper, N-type thermoelectric materials $Ag_{0.8}Pb_{18}SbTe_{20}$ with enhanced power factor were prepared by high pressure and high temperature (HPHT) method. The synthetic pressure-dependent transport properties including electrical resistivity, Seebeck coefficient and power factor were studied at room temperature. The maximum power factor of 15.4 μW cm^{-1} K^{-2} was obtained at the synthetic pressure of 4.0 GPa, which is much higher than that of the same sample prepared by other methods.

1. Introduction

Thermoelectric materials have become increasingly important due to the applications in power generation and electric cooling [1–5]. The efficiency of a thermoelectric (TE) material is usually characterized by the figure of merit ZT, defined as $ZT = T\sigma S^2/\kappa$, where T is the absolute temperature, σ is the electrical conductivity, S is the Seebeck coefficient, and κ is the thermal conductivity. The electrical properties are determined by the power factor, defined as $S^2\sigma$ or S^2/ρ, where ρ is the electrical resistivity. A good thermoelectric material must have a large power factor and a low thermal conductivity.

In 2004, Hus et al. reported that the quaternary thermoelectric material $Ag_{1-x}Pb_{18}SbTe_{20}$ could achieve a ZT value of about 2.2 at 800 K [1]. The high figure of merit was originated from nanocrystals of AgSb-rich phase embedded in a PbTe-rich matrix [1]. After that, many other pursuers were inspired by the progress and started to study the $AgPb_{18}SbTe_{20}$ system by many methods [6–13]. However, the high figure of merit of Hus is difficult to replicate [8–11]. The main reason is that the power factor is too low as a result of the high electrical resistivity the sample exhibits [8–11]. Recently, Li and coworkers studied $AgPb_{18}SbTe_{20}$ system by combining mechanical alloying (MA) and spark plasma sintering (SPS). They found that large power factor (18–20 μW cm^{-1} K^{-2}) and high figure of merit (1.2–1.5) could only be obtained when the composition was rich in Pb [12,13].

Comparing to other preparation methods for thermoelectric materials, the method of HPHT has many advantages, such as restraining the disorder, phase separation and other complicating factors during the preparation for materials [14]. In our previous studies, PbTe and $AgSbTe_2$ have been successfully prepared by HPHT [14,15]. The resistivity was decreased and the power factor for these samples was improved effectively under the effect of high pressure. $AgPb_{18}SbTe_{20}$ has the same crystal structure of NaCl as PbTe and $AgSbTe_2$, which could also be regarded as the alloy of PbTe and $AgSbTe_2$. High pressure may overcome the disadvantage of low power factor for $AgPb_{18}SbTe_{20}$ synthesized at normal pressure.

In the present study, thermoelectric material $Ag_{0.8}Pb_{18}SbTe_{20}$ was synthesized by HPHT. The transport properties including Seebeck coefficient and electrical resistivity are investigated at room temperature. Our results show that HPHT is an effective method to synthesize $AgPb_{18}SbTe_{20}$ with enhanced power factor.

2. Experimental

The $Ag_{0.8}Pb_{18}SbTe_{20}$ samples were prepared with tellurium (99.999% in purity), plumbum (99.9%), antimony (99.999%) and silver (99.9%) powders as sources, which were weighed according to the stoichiometric ratio. After being uniformly mixed in an agate mortar, the mixtures were pressed into a disk with diameter of 10 mm and height of 4 mm. The pole-shaped samples were assembled for HPHT synthesis. The $Ag_{0.8}Pb_{18}SbTe_{20}$ compounds were prepared in a cubic anvil high-pressure apparatus (SPD 6× 1200) with a sample chamber of 23 mm on an edge at 1200 K and 2.0–5.0 GPa. The temperature and pressure were kept for 20 min. After that, the

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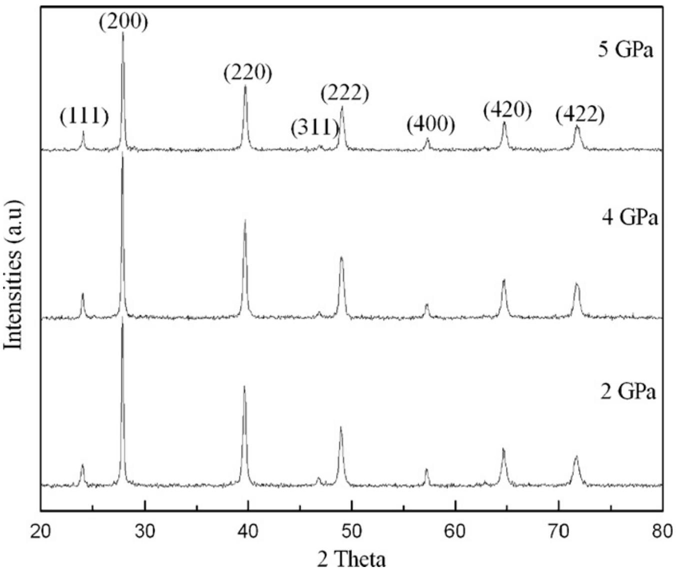

Fig. 1. XRD patterns of $Ag_{0.8}Pb_{18}SbTe_{20}$ prepared by HPHT.

electrical power for heating was cut off and the pressure was released. The pressure was estimated by the oil press load, which was calibrated by the pressure induced phase transitions of bismuth, thallium and barium metals. The temperature was estimated by the relationship of input heater power and temperature, which was measured by the platinum–rhodium thermocouples.

The collected samples were cut and polished on the surface for thermoelectric measurements which were carried out at room temperature. The density was measured by the Archimedes method. X-ray diffraction (XRD) measurements with Cu Kα radiation were performed on an X-ray diffractometer (D/MAX-RA). The fracture surface morphology was characterized by a JXA-8200 scanning electron microscope SEM. The electrical conductivity was measured by typical DC four-probe configuration. The Seebeck coefficient S was determined from thermoelectromotive force E_0 given by the temperature difference within 3–5 K between the two ends of the sample with a home-made instrument. The electromotive force was monitored by computer using the VI-Logger program provided by NI-company.

3. Results and discussion

The XRD patterns of $Ag_{0.8}Pb_{18}SbTe_{20}$ at different synthetic pressures are shown in Fig. 1. The patterns indicate that all compounds crystallize in the NaCl-type structure without noticeable secondary phase. The lattice constant of $Ag_{0.8}Pb_{18}SbTe_{20}$ evaluated from the X-ray diffraction pattern is shown in Table 1. The lattice constants are all about 0.646 nm, which is similar to the result of Kosuga et al. [9]. These results confirm that single-phase $Ag_{0.8}Pb_{18}SbTe_{20}$ compounds with PbTe-based structure have been synthesized by HPHT in a relatively short period of time.

As seen from Table 1, the density increases with an increase of synthesized pressure which may be helpful to decrease the resistivity for $Ag_{0.8}Pb_{18}SbTe_{20}$. Fig. 2 shows the SEM photograph of the fractured surface for the $Ag_{0.8}Pb_{18}SbTe_{20}$ sample synthesized at 4.0 GPa. (The SEM patterns of $Ag_{0.8}Pb_{18}SbTe_{20}$ prepared under different synthetic pressures are nearly the same.) From the photograph, we can see that the sample is considerably dense with micrometer scale grains.

The dependence of Seebeck coefficient of $Ag_{0.8}Pb_{18}SbTe_{20}$ samples prepared by HPHT on synthetic pressure is presented in Fig. 3.

Table 1
Lattice constant and density of $Ag_{0.8}Pb_{18}SbTe_{20}$ prepared by HPHT.

Synthetic pressure (GPa)	Lattice constant (rim)	Density (g/cm^3)
2	0.6462	8.055
4	0.6456	8.108
5	0.6462	8.134

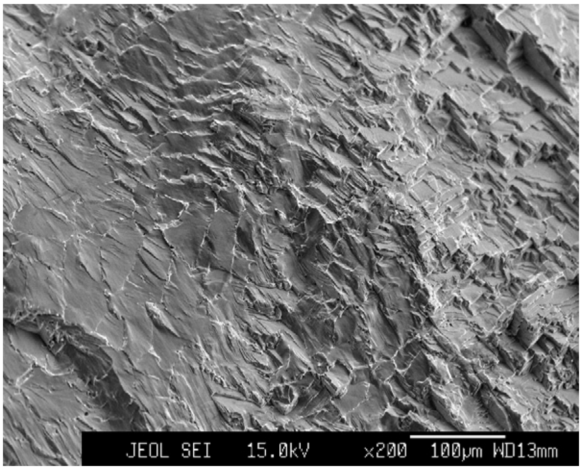

Fig. 2. SEM image of section plane inner of $Ag_{0.8}Pb_{18}SbTe_{20}$.

The Seebeck coefficients of all the samples are negative, which indicate that the majority of charge carriers are electrons (n-type). The Seebeck coefficient in absolute value for $Ag_{0.8}Pb_{18}SbTe_{20}$ decreases with an increase of synthetic pressure from $-235.9\,\mu V\,K^{-1}$ at 2.0 GPa to $-35.3\,\mu V\,K^{-1}$ at 5.0 GPa. The pressure-dependent Seebeck coefficient is similar to the results of PbTe we reported early [14].

The electrical resistivity of $Ag_{0.8}Pb_{18}SbTe_{20}$ as a function of synthetic pressure from 2.0 to 5.0 GPa is shown in Fig. 4. The electrical resistivity decreases with an increase of synthetic pressure, which is similar to the results of in situ measurement for PbTe-based samples under high pressure [14]. The electrical resistivity of the samples prepared by HPHT is lower than that of the samples prepared at normal pressure reported by other pursuers [8–10]. It may be helpful to enhance its thermoelectric properties, which has been testified by the following experimental results (see Fig. 5).

The power factor for $Ag_{0.8}Pb_{18}SbTe_{20}$ calculated by Seebeck coefficient and electrical resistivity is shown in Fig. 5. With the increasing synthetic pressure, the power factor increases when the pressure is lower than 4.0 GPa and then decreases. The maximum power factor for $Ag_{0.8}Pb_{18}SbTe_{20}$ synthesized at 4.0 GPa reaches to $15.4\,\mu W\,cm^{-1}\,K^{-2}$.

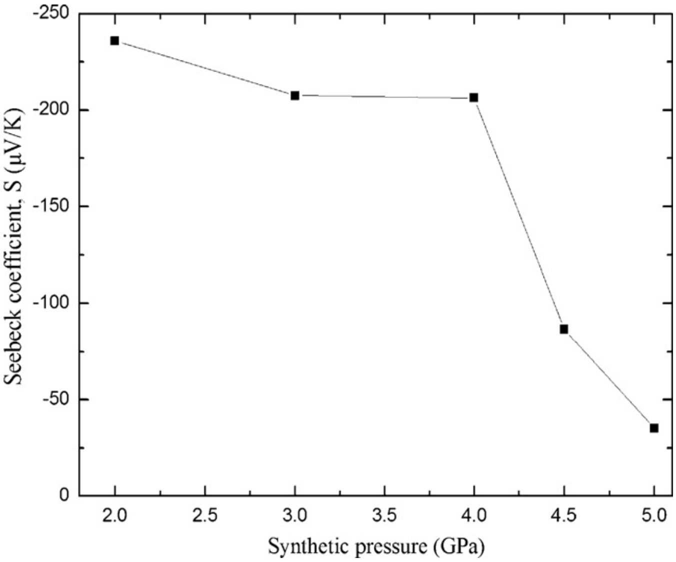

Fig. 3. Relationship between Seebeck coefficient and synthetic pressure for $Ag_{0.8}Pb_{18}SbTe_{20}$ measured at room temperature.