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Thermoelectric properties of PbTe prepared at high pressure and high temperature

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Abstract

Lead telluride (PbTe) with rock-salt structure was successfully obtained by a high-pressure and high-temperature (HPHT) method. The orientation of the PbTe samples varies with pressure increase. The results—a decrease in the Seebeck coefficient, resistivity and thermal conductivity of PbTe with pressure but an increase in the thermoelectric power figure σS^2 —indicate that the figure of merit Z of PbTe samples can be improved several times over by using HPHT.

1. Introduction

In recent years, studies of lead telluride (PbTe) have been pursued due to the device applications [1–3] in many fields such as IR detection and photoconductivity. There has been extensive work on the thermoelectric properties in the bulk state [4] and the film state [5] with the aim of improving the thermoelectric energy conversion efficiency. The performance of thermoelectric materials can be evaluated by the figure of merit defined as $Z = \sigma S^2 / \kappa$ where S is the Seebeck coefficient, σ is the electrical conductivity, and κ is the total thermal conductivity.

It is well known that improvement in the figure of merit is possible by increasing the Seebeck coefficient and electrical conductivity and by decreasing thermal conductivity. It is known that scattering phonons caused by point defects and crystal boundaries can reduce the lattice thermal conductivity. Therefore, recently some efforts have looked at the possibility of reducing the lattice thermal conductivity to improve Z [6] by means of the scattering of phonons at grain boundaries. In fact, it is reported that Z for highly disordered alloys of PbTe with a mean grain size of 1 μm is calculated to be about 10% higher than the equivalent single-crystal values [7].

The crystal and electrical structures of semiconductors, which determine the properties of the materials, can be changed by high pressure [8]. Therefore, the thermoelectric properties, both electrical conductivity and Seebeck coefficient, may be altered by high pressure. Recent

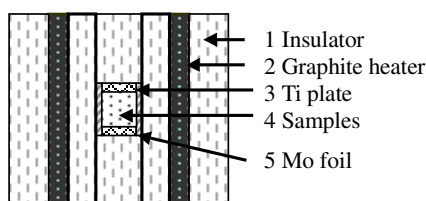


Figure 1. The sample assembly for the growth of PbTe.

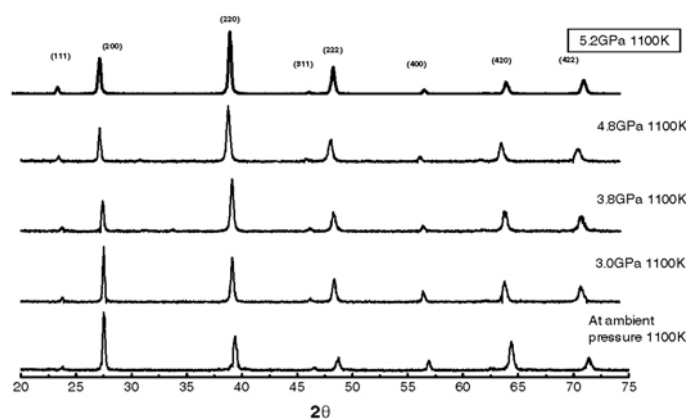


Figure 2. XRD patterns of PbTe prepared at different pressures.

experiments by Badding's group on alloys of $(\text{Ce}_{3-x}\text{Nd}_x\text{PtSb}_4)$ have suggested that values of Z somewhat above 1 can be attained under pressure [9].

In this work, we report how the thermoelectric properties of the IV–VI semiconductor PbTe, both electric resistivity and Seebeck coefficient, respond to HPHT.

2. Experimental details

The basic lead and telluride powders (high purity powder, 99.999%) were mixed completely in an agate mortar under Ar protection, and then shaped. Preparation of the PbTe compound was carried out using a cubic anvil high-pressure apparatus with a sample chamber of 23 mm. The sample assembly is shown in figure 1. The pressure was given by a calibration curve that was established by determining the applied loads corresponding to the phase transformation pressure of bismuth, thallium, and barium.

The samples obtained were characterized by x-ray diffraction (XRD). Electrical conductivity and Seebeck coefficient were measured under Ar protection by a dc four-probe apparatus with copper–constantan thermocouples and Pt wires as current leads.

3. Results and discussion

Typical XRD patterns of PbTe obtained using $\text{Cu K}\alpha$ radiation are given in figure 2. The XRD results show that the samples synthesized at ambient pressure and high pressure are polycrystalline with NaCl-type structure. It can be seen that the orientation of the samples changes as the synthesis pressure increases. At ambient or a low pressure, the crystal growth is mainly along the (200) direction. When the pressure increases, the crystal face (200) becomes

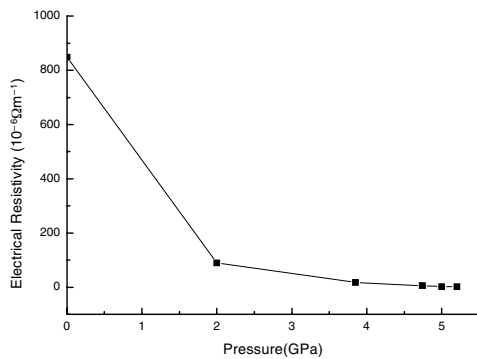


Figure 3. The dependence of resistivity on pressure for PbTe.

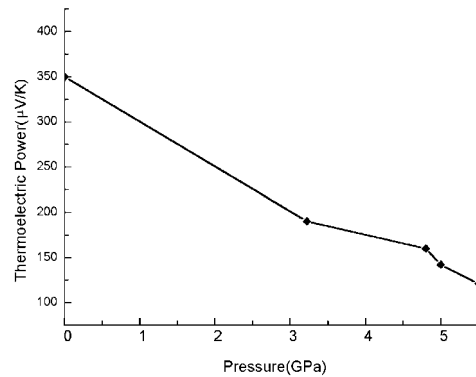


Figure 4. A plot of Seebeck coefficient versus synthesis pressure for PbTe. The curve was measured at ambient pressure.

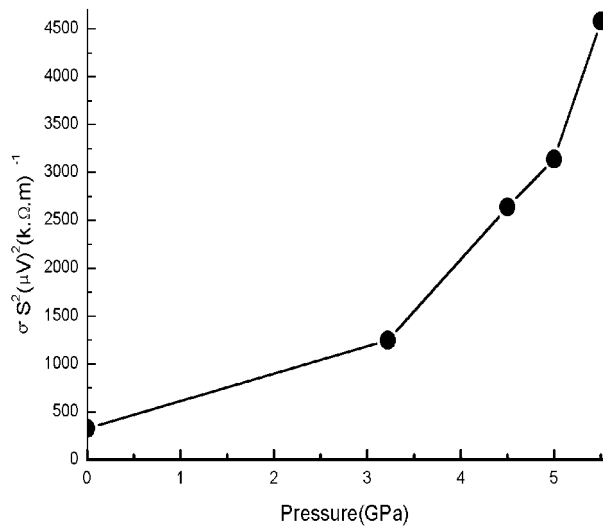


Figure 5. The relationship between the thermoelectric power factor and pressure for PbTe.

prominent. Although the structural parameters change with pressure and temperature, the structure is still a NaCl-type structure.

The resistivity of PbTe as a function of the preparation pressure is shown in figure 3. We find that the resistivity of PbTe varies clearly with the synthesis pressure. The resistivity of the samples synthesized at high pressure and high temperature is two orders of magnitude less than that for samples synthesized at ambient pressure. This may be caused by the change of the electrical structure and the doping concentration increasing under high pressure.

The dependence of the Seebeck coefficient of PbTe on the preparation pressure is shown in figure 4. We find that the Seebeck coefficient decreases with synthesis pressure increase. Moreover, the increase in the carrier concentration reduced the mobility of the carriers, which causes the Seebeck effect to decrease with pressure increase. This result is in good agreement with other experiments [11, 12].

σS^2 as a function of synthesis pressure is plotted in figure 5. It can be seen that σS^2 increases as the pressure gets larger. Although the Seebeck coefficient decreases with

Table 1. The thermal conductivity of PbTe synthesized at different pressures.

Sample	Theoretical value	3.8 GPa	4.2 GPa	4.5 GPa	4.8 GPa
κ (W K ⁻¹ mol ⁻¹)	2.3	1.05	1.19	1.28	1.58

increasing pressure, the increase of σS^2 indicates that the effect of pressure on the electrical resistivity is larger than that on the Seebeck coefficient in PbTe synthesized at high pressure.

The thermal conductivities of PbTe prepared at different pressures are shown in table 1. One observes that the thermal conductivity of PbTe synthesized at high pressure is lower than that for samples prepared at ambient pressure. This may be caused by the change of orientation in PbTe.

From the above analysis, it can be concluded that the figure of merit Z is improved by preparation pressure.

4. Conclusions

PbTe was prepared successfully at high pressure and high temperature. The XRD results show that the samples are polycrystalline with rock-salt crystal structure, and the orientation of the samples changes with the pressure. The Seebeck coefficient, resistivity and thermal conductivity decrease with synthesis pressure increase. However, the thermoelectric power figure σS^2 increases simultaneously with pressure. The figure of merit Z is enhanced nearly several times over by pressure. Pressure is a useful tool in synthesizing thermoelectric materials and improving the thermoelectric properties.

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