Properties of thermoelectric device with $(Bi_xSb_{1-x})_2(Te_ySe_{1-y})_3$ materials

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Thermoelectric materials are of interest for applications as heat pumps and power generators. The COP of thermoelectric devices is quantified by the figure of merit, ZT, where Z is a measure of thermoelectric devices and T is the absolute temperature. The $(Bi_xSb_{1-x})_2(Te_ySe_{1-y})_3$ alloys are state-of-the-art room temperature materials. This paper focuses on the properties of refrigeration of these kinds of materials. The electrical properties of thermoelectric materials were measured from 80-300 K. Power factor of materials were given, respectively. The properties of thermoelectric devices are studied at different temperature, the cooling power and cooling temperature of thermoelectric devices are given.

INTRODUCTION

Traditional approaches to cooling are based on thermodynamic cycles involving compression and expansion of refrigerant gases. Thermoelectric refrigeration devices, in contrast, do not rely on dynamic cycles of gases. It relies on physical a phenomena called the Peltier effect. When an electric current passes through a thermoelectric material, the heat transported by charge carriers (electron or hole) leads to a temperature gradient [1]. Heat is absorbed on the cold side and rejected at the sink, thus providing a cooling temperature.

Thermoelectric devices have several distinct advantages, including non-moving parts, non-vibration, quiet performance and spot cooling. More importance, it is a “green” refrigeration with no freon refrigerants and a negligible global warming potential. They will be applied in the cooling of CCDs (charge-coupled devices), infrared detectors, low-noise amplifiers, and computer chips. Thermoelectric refrigeration is also being considered in the automobile industry for use in the “next-generation vehicle”. The most common application of these materials is the thermoelectric cooler and warmer, which are sold at many local stores. The essence of defining a good thermoelectric material lies in material’s figure of merit, $Z = S^2\sigma/K$, where S is the Seebeck coefficient (defined as $\Delta V/\Delta T$, V is voltage and T is absolute temperature), σ is the electrical conductivity, and K is the total thermal conductivity ($K=K_e+K_L$, the lattice and electronic contributions, respectively). Alloys based on the $(Bi_xSb_{1-x})_2(Te_ySe_{1-y})_3$ system for room temperature applications have higher performance for thermoelectric refrigeration. For all the current state-of-the-art materials, the dimensionless figure of merit, ZT, is about 1. There is no theoretical or thermodynamic reason why it cannot be larger. Researchers have done lots of work to improve thermoelectric properties of materials. Up till now, how to reduce the lattice thermal conductivity becomes more important in improving materials’ figure of merit. This concept is related to Slack’s earlier

assertion of a “phonon-glass/electron-crytal” model, which suggests that a good thermoelectric material should have the electronic properties of a crystalline material and the thermal properties of a glass. In this paper, we focused on the properties of thermoelectric refrigeration devices at different temperature. The electrical conductivity and Seebeck coefficient of n-type and p-type materials were measured, and the cooling power and cooling temperature (Δ T) of the thermoelectric device are also given.

PROPERTIES OF MATERIALS

Three kinds of materials were employed in our experiments. Alloy A (p-type) and alloy B (n-type) are commercial products from Northern Refrigeration Company (Harbin, China). Alloy C is prepared by mechanical alloying method in our laboratory with the milling time for 80hrs, and its chemical composition is $(Bi_2Te_3)_{0.25}(Sb_2Te_3)_{0.75}$+3wt%Te.

XRD patterns

The XRD patterns of the alloy A and alloy B are shown in Figure 1. There are not Bi, Te, Sb and Se traces in the diffraction patterns of alloy A and alloy B. XRD patterns of alloy A and alloy B materials are almost same because they have same crystal structure.

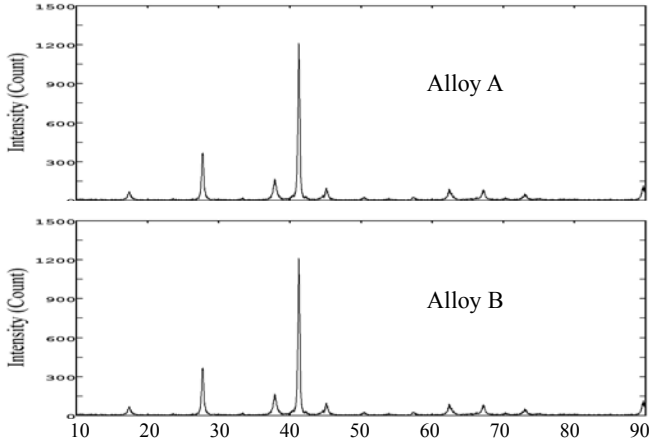

Figure 1 X-ray diffraction patterns (Cu K_α radiation) for A and B materials

Electrical properties

The electrical conductivity of the studied materials was measured using the four-probe method. The results of the measurements are shown in Figure 2. For impurity semiconductor, concentration of current carriers almost doesn’t change with the change of temperature when intrinsic excitation is not very strong. Lattice vibration has a strong impact on carrier mobility. With the increase of temperature, lattice vibration becomes stronger, so as to lead the decrease of the carrier mobility. The experimental results show that the electrical conductivity of alloy A (p-type) and alloy B (n-type) are $0.99\times10^5\ \Omega^{-1}.m^{-1}$ and $0.68\times10^5\ \Omega^{-1}.m^{-1}$ at 300K, respectively.

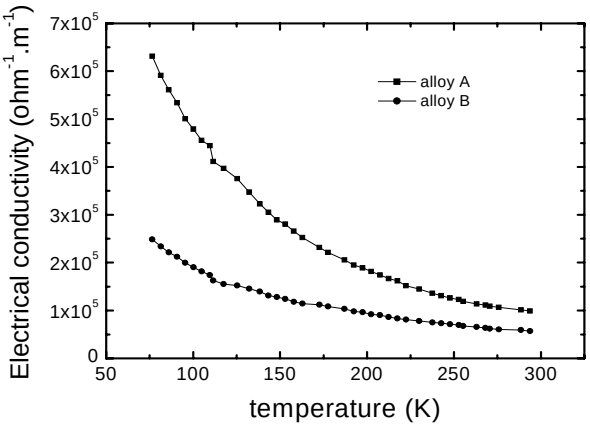

Figure 2 Materials’ Electrical conductivity changes with temperature 80-300 K

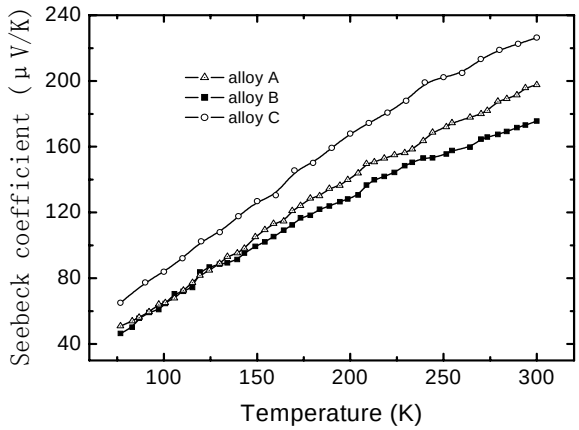

Figure 3 Seebeck coefficients of materials as a function of temperature 80-300 K

Figure 3 is the measurement results of Seebeck coefficient. It indicates that the Seebeck coefficient as a function of temperature, increases with increase of temperature. It is noted that, to aid comparison,

the absolute value of Seebeck coefficient of p-type materials was used in Figure 3. The alloy C (p-type) prepared by mechanical alloying method in our laboratory, is also studied, and the result (curve C) was also shown in Figure 3. The Seebeck coefficients of alloy B and alloy C are 197.6 and 226.3 μV/K at 300 K, respectively. Seebeck coefficient of alloy C is higher than those of the commercial materials. The thermoelectric properties of alloy C, will be studied in the future, including thermal conductivity and electrical conductivity.

The power factor of alloy A and alloy B is shown in Figure 4. It shows that power factor increases with increase of temperature below 200K and almost invariable above 200K. The data of power factor of alloy A and alloy B are 3.8×10^{-3} and 2.2×10^{-3} W/m • K^2 at 300 K, respectively.

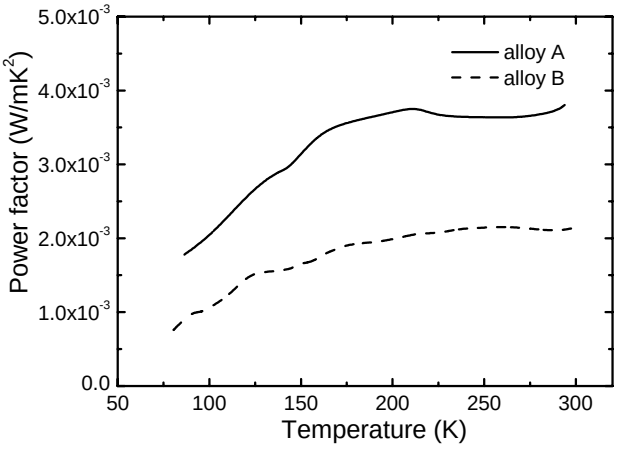

Figure 4 Power factor as a function of temperature from 80 K to 300 K

PERFORMANCE OF THERMOELECTRIC DEVICE

It is apparent that the figure of merit Z, as defined by $Z=S^2\sigma/K$, is a characteristic not of a pair of materials but, rather, of a particular couple. For a given pair of materials, we can write Z as following equation [6]:

$$Z=(S_p-S_n)^2/[(\kappa_p\rho_p)^{1/2}+(\kappa_n\rho_n)^{1/2}]^2. \quad (1)$$

Actually, above equation is rather cumbersome when attempting to find a good thermoelectric material, since it involves the properties of both thermoelements.

A Peltier module is composed of thermoelectric couples（alloy A and alloy B）, heat conducting ceramic plates and some electrically conducting wire. Ceramic plates form the cold and hot surfaces of the module and provide good heat transfer and low electrical conductivity. The ceramic face size of the tested device was $3.5\times 4.0 cm^2$, the test was carried out at 1atm. The hot side plate was cooled using water

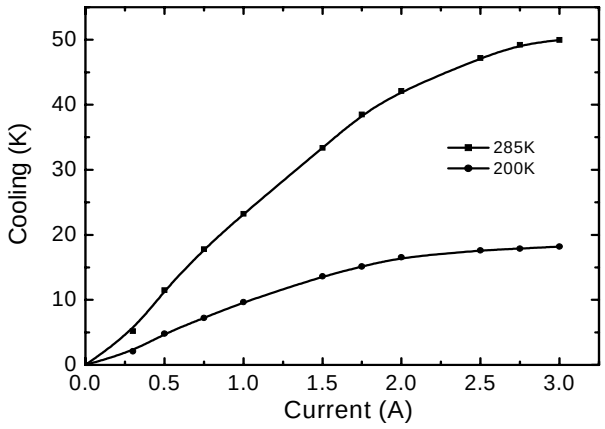

Figure 5 Observed cooling temperature as a function of current in a 3.5×4 cm^2 rectangle

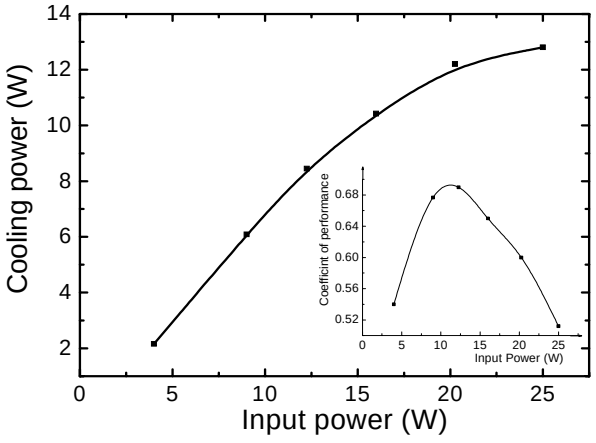

Figure 6 Cooling power as a function of input power (hot side temperature was 285 K)

at 285 K and LN_2 as a cold background at 200 K. Other researchers have done a lot of work in thermoelectric device at room and low temperature [2-5].

The observed cooling temperatures and cooling power (285 K and 200 K hot side plate temperature) as functions of current or input power are shown in Figure 5 and Figure 6 respectively. There are 49.5 K and 17.5 K cooling temperatures under hot side temperatures of 285 K and 200 K, respectively. In this bulk device, 13 W output power at 25 W input power is obtained when the hot side temperature is 285 K and cooling temperature is 20 K.

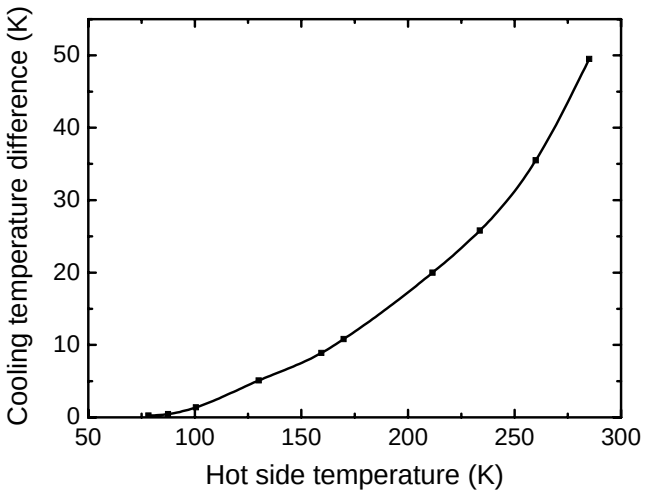

Figure 7 Cooling temperature as a function of hot side temperature

Figure 7 shows the change of cooling temperature differences at different hot side temperatures. The temperature difference of 1.4 K was obtained when the hot side temperature was 100 K. The temperature difference was only 0.45 K when the hot side temperature was 80 K.

SUMMARY

In summary, the electrical properties of materials have been measured as a function of temperature. The properties of bulk thermoelectric device have been measured. When the hot side temperatures are 285 K and 200 K, the cooling temperature differences are 49.5 K and 17.5 K, respectively. The maximum cooling power obtained is 13 W when the input power is 25 W.

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