Thermal Conductivities and Conduction Mechanisms of Sb-Te Alloys

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ABSTRACT

The thermal conductivities of Sb-*x* at%Te alloys (*x* = 14, 25, 44, 60, 70, and 80) have been measured by the hot strip method from room temperature up to temperature just below the respective melting points. For the intermetallic compound Sb_2Te_3 ($x = 60$), the thermal conductivity decreases from room temperature up to about 500 K and then increases up to temperature just below the melting temperature. For other Sb-*x* at%Te alloys, where $x > 60$, the temperature dependence of the thermal conductivities is negative. The thermal conductivities of the alloys are close to that of tellurium at room temperature. On the other hand, where $x < 60$, the temperature dependence is positive. The thermal conductivities of the alloys increase remarkably with decreasing Te concentration at room temperature.

Keywords Sb-*x* at%Te alloys, Thermal conductivity, Hot strip method

1. INTRODUCTION

Phase change random access memory (PCRAM) is a promising nonvolatile data storage technology for next generation memory owing to higher performance characteristics such as fast operation and higher scalability[1)]. Sb-Te binary alloys have been suggested to be suitable candidates for PCM devices because of a dramatic change in electric resistivities associated with transformation between amorphous and crystalline phases, which forms the basis for data storage[2, 3)]. The phase transformation is controlled by Joule-heating and cooling processes and, thus, accurate data for thermal conductivity of Sb-Te binary alloys are indispensable to optimal designing for PCM devices. Furthermore, the physical properties of Sb-Te alloys have drawn much attention. Onderka and Fitzner[4)] have measured the electrical resistivity of Sb_2Te_3, the intermetallic compound in Sb-Te alloys, and found that, with increasing temperature, the resistivity increases in the solid state but decreases in the liquid state. On the basis of this temperature dependence of the resistivity, Onderka and Fitzner [4)] have proposed that Sb_2Te_3 exhibits metallic conduction in the solid state and semiconductor characteristics in the liquid state. For better understanding of the physical properties of Sb-Te alloys, more accurate thermal conductivity data are desperately required. Against this background, the present work aims to determine thermal conductivities of Sb-Te binary alloys as functions of temperature and composition.

2. EXPERIMENTAL

Samples used were Sb-*x* at% Te (*x* = 14, 25, 44, 60, 70, and 80). Thermal conductivity was measured by the hot strip method as shown in Figure 1[5)]. Cylindrical samples of Sb-Te (20mm diameter and 40-50mm length) were prepared from Sb and Te powders with 99.9 mass% purity. A powder mixture (100 g) was melted in a quartz crucible (20mm inner diameter) at 973K for 4 h in vacuum, followed by furnace cooling. Subsequently, the sample was taken from the tube and cut into two along the vertical axis, and the cross sections were mechanically polished using emery papers up to #2000. Thermal conductivity measurements were conducted in argon atmosphere from 298 K up to temperatures just below the respective melting points. Prior to measurements of Sb-Te alloys, the thermal conductivity of fused silica was measured to confirm the reliability of the hot strip method because the thermal conductivity of fused silica is close to that of Sb-Te binary alloys. Two pieces of fused SiO_2 block (40×20×10 mm) were used as the samples.

3. RESULTS AND DISCUSSION

Figure 2 shows the thermal conductivity for silica glass as a function of temperature, together with the reference data. The present results show a good agreement with the reference data$^{6)}$, which validates the reliability of hot strip method.

Figure 3 shows the temperature change with time recorded during thermal conductivity measurements on Sb_2Te_3 at 298 K and 789 K. There are good linearity between temperature change and time in the time period 0.7-2 s. From the slope of the linear portion, the thermal conductivity of Sb_2Te_3 has been determined.

Figure 4 shows the thermal conductivity for solid Sb_2Te_3 as a function of temperature, in comparison with values measured by other investigators$^{7-11)}$ and calculated from the Wiedemann−Franz (WF) law$^{12)}$ using the resistivity data reported by Onderka$^{4)}$ and the theoretical Lorentz number, 2.45×10^{-8} $W\Omega/K^2$. Thermal conductivity data obtained during the heating cycle are in very good agreement with those obtained during the cooling cycle. The data obtained in the present work show an interesting temperature dependence: the thermal conductivity decreases with increasing temperature up to approximately 500 K and then increases. The data obtained at room temperature in the present work are close to those reported from Yokota and Katayama$^{11)}$. The present data are also in fairly good agreement with values calculated by the WF law at temperatures lower than about 600 K, above which they deviate from each other.

Figure 5 shows the present thermal conductivity results of Sb_2Te_3 along with the thermal conductivity due to free electron (λ_e), phonon (λ_{ph}) and ambipolar diffusion (λ_{ab}) estimated by Yokota and Katayama$^{11)}$. These results indicate that the contribution from the free electrons is relatively small, i.e., 1 W m^{-1} K^{-1}, and thus phonon diffusion dominates thermal conduction at temperatures lower than 300 K. With increasing temperature, however, both contributions to the thermal conductivity become smaller, whereas the contribution from ambipolar diffusion increases. This contribution is unnoticeable at low temperature but increases remarkably with increasing temperature to become comparable to the contribution from phonon diffusion. The increase of thermal conductivity of Sb_2Te_3 above 600 K would be due to the ambipolar diffusion.

For other Sb-x at%Te alloys, where $x > 60$, the temperature dependence of the thermal conductivities is negative. The thermal conductivities of the alloys are close to that of tellurium at room temperature. On the other hand, where $x < 60$, the temperature dependence is positive. The thermal conductivities of the alloys increase remarkably with decreasing Te concentration at room temperature.

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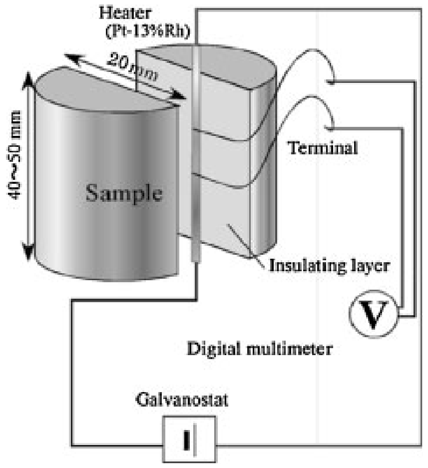

Fig. 1 Schematic diagram for hot strip method

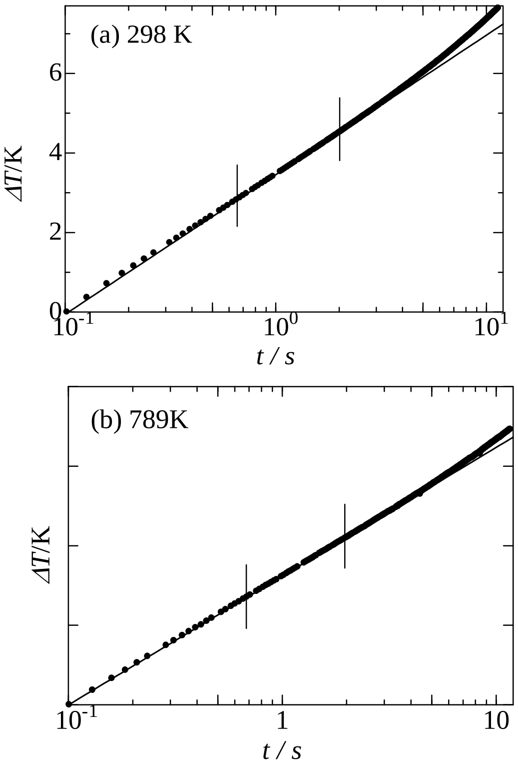

Fig.3 Temperature rise of heater during thermal conductivity measurements on Sb_2Te_3 with time at (a) 298 K and (b) 789 K

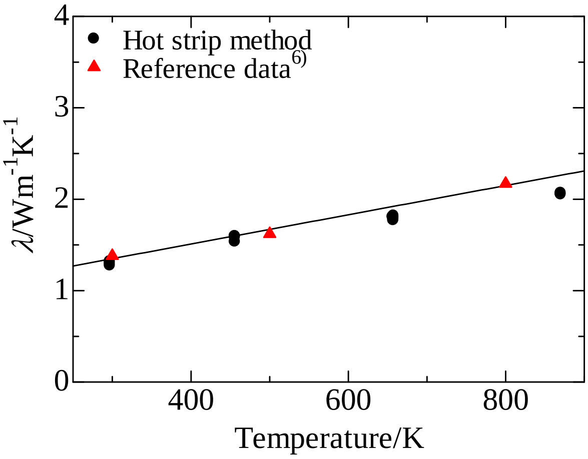

Fig. 2 Thermal conductivity of silica glass as function of temperature

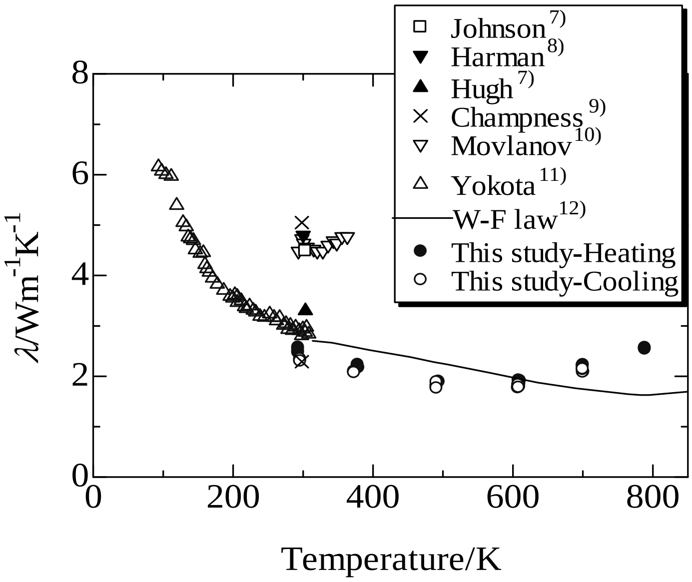

Fig.4 Temperature dependence of thermal conductivity for solid Sb$_2$Te$_3$ by hot-strip method along with data measured and calculated from W-F law

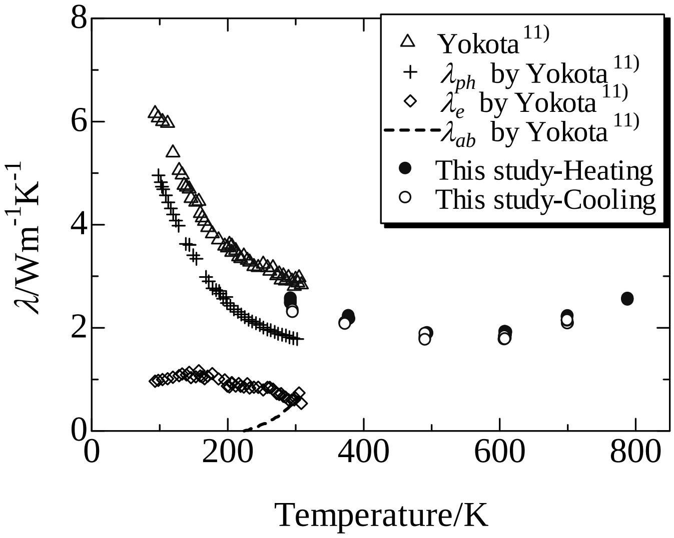

Fig.5 Present thermal conductivity results of Sb$_2$Te$_3$ along with thermal conductivity due to free electron (λ_e), phonon (λ_{ph}) and ambipolar diffusion (λ_{ab}) estimated by Yokota and Katayama