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Thermoelectric figure of merit of Zn_4Sb_3 samples grown by mechanical alloying and subsequent sintering

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Abstract – Zn_4Sb_3 alloys were prepared by mechanical alloying followed by a sintering process of different duration (0 to 50 hours). Transport properties involving electrical resistivity ρ(T), Seebeck coefficient S(T) and thermal conductivity κ(T) were studied in the temperature range between 100 and 290K. The Seebeck coefficient and electrical resistivity increase with the sintering time reaching maximum values close to 350 μV/K and 18 mΩcm, respectively. The thermal conductivity decreases with the annealing time up to values less than 1 W/K-m. From ρ(T), S (T) and κ(T) data it was possible to determine the thermoelectric power factor (PF) and the dimensionless figure of merit (ZT), which reach maximum values close to 25 μW/K^2-cm and 0.35, respectively.

The challenge of thermoelectric materials research is to find compounds with high ZT ($ZT = S^2/\rho\kappa$) values. In order to increase the magnitude of ZT, it is necessary to reduce the thermal conductivity and/or to enhance the thermoelectric power factor [1]. Among the thermoelectric materials recently studied, the Zn_4Sb_3 compound is a promising thermoelectric material, due mainly to its high figure of merit values, which is a result of its particular low thermal conductivity [2].

Zinc and antimony powders (99.999% pure) were used to prepare Zn_4Sb_3 samples, these components were mixed and submitted to a mechanical alloying process followed by a sintering treatment at 500^{0}C, the duration of this thermal process took values from 0 to 50 hours.

The x-ray diffraction analysis shows that the samples are multi-phased with the presence of rombohedral (hex.) semiconducting Zn_4Sb_3 and small quantities of ZnSb, metallic Zn and Sb as secondary phases.

The electrical resistivity shows a weak metallic temperature behavior, its magnitude increases with the processing time from 1 mΩ-cm to 18 mΩ-cm. The absolute Seebeck coefficient (figure 1a) shows positive values, indicating a p-type conduction. Seebeck coefficient experimental data fits well to the Mott model (solid lines in figure 1a). Thermal conductivity shows a linear temperature dependence, its magnitude decreases with the processing time to values less than 1 W/K-m. With the thermal process its electronic component decreases from 10% to 1% of the total thermal conductivity.

From S(T), ρ(T) and κ(T) experimental data the thermoelectric power factor and dimensionless figure of merit were calculated. These parameters enhance with the processing time up to reach maximum values close to 25 μW/K^2-cm and 0.35 in the sample annealed during 20 hours (figures 1b and 1c).

Despite these values being smaller than those of conventional semiconductors, the exhibited values by the performance parameters suggest that mechanical alloying followed by solid-state synthesis is an alternative path to prepare these kind of compounds, however, it is necessary to control the growth of ZnSb and the losses of Zn and Sb during the thermal processes.

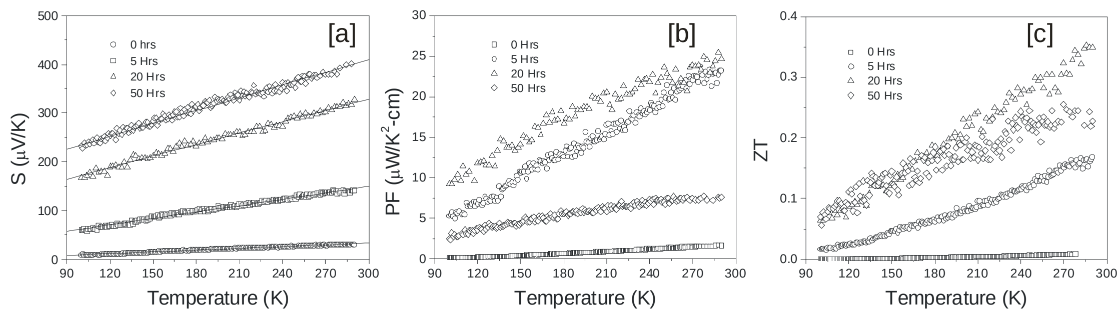

Figure 1. Seebeck coefficient, thermoelectric power factor and dimensionless figure of merit of Zn_4Sb_3 compounds, grown by a combination of mechanical alloying and solid state sintering.

References

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