THERMOELECTRIC PROPERTIES OF COMPLEX MAIN-GROUP TELLURIDES

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Thermoelectric materials offer unique opportunities for the construction of solid-state devices for refrigeration and power generation. The necessary combination of a high electrical conductivity (σ), low thermal conductivity (κ), and large Seebeck coefficient (S) required for high performance imposes considerable demands on materials design. This has led to a recent resurgence of interest in exploratory synthesis of chemically complex phases for thermoelectric applications. Recent reports indicate that mixed-metal tellurides related to rock-salt-type PbTe exhibit potentially technologically-useful thermoelectric properties, with n-type $AgSbPb_mTe_{m+2}$, possessing ZT values as high as 2.2 for m=18 at 800K

We are exploring the effects of chemical substitution on the electrical and thermal transport properties of mixed-metal tellurides containing main-group elements. This investigation encompasses materials with structures related to that of rock-salt together with layered variants. Materials of general formula $Tl_{1-x}MPb_mTe_{m+2}$ ($0 \leq x \leq 0.3$; $10 \leq m \leq 18$; M=Bi, Sb), adopt a rock-salt type structure and show promising thermoelectric behaviour. The materials are degenerate semiconductors, with the dominant charge carriers in the antimony-containing phases being holes, and those in the in the bismuth-containing materials, electrons. Figures-of-merit (ZT) of up to 0.65 are observed at 373K for an optimum doping level of x=0.1. The physical property data for these materials will be presented together with structural data that suggest the enhancement in properties over those of PbTe may be related to the occurrence of a degree of phase segregation on double substitution.

The ternary phase $PbBi_2Te_4$ shows thermoelectric properties comparable with those of members of the $Tl_{1-x}BiPb_mTe_{m+2}$ series. However, by contrast with the rock-salt phases, chemical substitution by tin at the lead site leads to a significant degradation in the thermoelectric behaviour of the end-member. The reduction in the figure-of-merit with increasing tin concentration may be traced to the fact that the low measured thermal conductivity (< 1 W m^{-1} K^{-1}) possessed by all quaternary phases does not compensate for the order-of-magnitude reduction in |S| that occurs with increasing levels of substitution.

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