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Supporting Online Material

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Materials and Methods

SOM Text

Figs. S1 to S5

Sounds S1 to S6

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High-Thermoelectric Performance of Nanostructured Bismuth Antimony Telluride Bulk Alloys

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The dimensionless thermoelectric figure of merit (ZT) in bismuth antimony telluride (BiSbTe) bulk alloys has remained around 1 for more than 50 years. We show that a peak ZT of 1.4 at 100°C can be achieved in a p-type nanocrystalline BiSbTe bulk alloy. These nanocrystalline bulk materials were made by hot pressing nanopowders that were ball-milled from crystalline ingots under inert conditions. Electrical transport measurements, coupled with microstructure studies and modeling, show that the ZT improvement is the result of low thermal conductivity caused by the increased phonon scattering by grain boundaries and defects. More importantly, ZT is about 1.2 at room temperature and 0.8 at 250°C, which makes these materials useful for cooling and power generation. Cooling devices that use these materials have produced high-temperature differences of 86°, 106°, and 119°C with hot-side temperatures set at 50°, 100°, and 150°C, respectively. This discovery sets the stage for use of a new nanocomposite approach in developing high-performance low-cost bulk thermoelectric materials.

Solid-state cooling and power generation based on thermoelectric effects have potential applications in waste-heat recovery, air conditioning, and refrigeration. The efficiency of thermoelectric devices is determined by the materials' dimensionless figure of merit, defined as $ZT = (S^2\sigma/k)T$, where S , σ , k , and T are the Seebeck coefficient, electrical conductivity, thermal conductivity, and absolute temperature, respectively (1–3). An average ZT in the

application temperature range must be higher than 1 to make a thermoelectric device competitive (1–3).

There have been persistent efforts to improve ZT values since the 1950s, but the peak ZT of dominant commercial materials based on Bi₂Te₃ and its alloys, such as Bi_{1.8}Sb_{0.2}Te₃ (p-type), has remained at 1. During the past decade, several groups have reported enhanced ZT in (i) superlattices such as Bi₂Te₃/Sb₂Te₃ (4) and PbSe_{0.98}Te_{0.02}/PbTe (5), because of reductions in the lattice thermal conductivity, and (ii) new bulk materials, such as lead antimony silver telluride (LAST) and its alloys (6), including skutterudites (7). Although high ZT values were reported in superlattice structures, it has proven difficult to use them in large-scale energy-conversion applications because of limitations in both heat transfer and cost. Bulk materials with improved ZT, such as LAST and skutterudites, are ideal for high-temperature operations. However, at rela-

tively near room temperature (0° to 250°C), Bi₂Te₃-based materials still dominate.

We have pursued an approach in which the primary cause of ZT enhancement in superlattices—reduced thermal conductivity—also exists in random nanostructures (8, 9). We report a substantial ZT increase in bulk materials made from nanocrystalline (NC) powders of p-type Bi_{1.8}Sb_{0.2}Te₃, reaching a peak ZT of 1.4 at 100°C. The enhanced ZT is the result of a significant reduction in thermal conductivity caused by strong phonon scattering by interfaces in the nanostructures. There have also been reports of ZT improvements at room temperature in Bi₂Te₃-based materials caused by the addition of Bi₂Te₃ nanotubes (10) and by melt spinning (11).

Our method, on the other hand, is based on the ball milling and hot pressing of nanoparticles into bulk ingots. This approach is simple, is cost effective, and can be used on other materials. Our materials have a ZT of about 1.2 at room temperature and 0.8 at 250°C with a peak of 1.4 at 100°C. In comparison, conventional Bi₂Te₃-based materials have a peak ZT of about 1 at room temperature and about 0.25 at 250°C. The high ZT in the 25° to 250°C temperature range makes the NC bulk materials attractive for cooling and low-grade waste-heat recovery applications. The materials can also be integrated into segmented thermoelectric devices for thermoelectric power generation that operate at high temperatures. In addition to the high ZT values, the NC bulk materials are also isotropic. They do not suffer from the cleavage problem that is common in traditional zone melting-made ingots, which leads to easier device fabrication and system integration and to a potentially longer device lifetime.

Sample preparation. Nanopowders were made by ball milling bulk p-type BiSbTe alloy ingots (12). Bulk disk samples (1.25 to 2.5 cm in diameter and 2 to 15 mm in thickness) were made by hot pressing the nanopowders loaded in 1.25- to 2.5-cm (inner diameter) graphite dies (12). Disks (1.25 cm in diameter and 2 mm in thickness) and bars (about 2 mm by 2 mm by

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