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Thermoelectric Properties of Sintered *n*-Type and *p*-Type Tellurides

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Characterization of powder-metallurgically manufactured $(Bi_xSb_{1-x})_2(Te_ySe_{1-y})_3$ thermoelectric materials is presented. The manufacturing methods were spark plasma sintering (SPS) and hot isostatic pressing (HIP). x-Ray diffraction (XRD) and density measurements as well as transport characterization and scanning electron microscopy were performed on the materials. It is shown that both sintering techniques yield reasonable thermoelectric characteristics for *p*-type ($x = 0.2$, $y = 1$) as well as *n*-type ($x = 0.95$, $y = 0.95$) materials. Insight into the underlying reasons such as the scattering processes limiting the characteristics is gained by fitting experimental transport data using a theoretical model. The limitations and further optimization issues of our approach in thermoelectric material preparation are discussed.

Key words: Tellurides, HIP, SPS, transport properties, thermoelectricity

INTRODUCTION

Bismuth and antimony chalcogenides are the most well-established thermoelectric materials due to their high room-temperature and near-room-temperature thermoelectric figures of merit $Z = \alpha^2\sigma/\kappa$, where α is the Seebeck coefficient, σ is the electrical conductivity, and κ is the thermal conductivity. This mainly stems from their high ratio of electrical conductivity to thermal conductivity—a property widely studied but not fully understood.[1] Further improvements have been obtained by microstructuring or nanostructuring the material.[2–4] In the optimal case, the lattice thermal conductivity may be further decreased without affecting the electrical conductivity or the Seebeck coefficient. For chalcogenides, improvements of up to tens of percent have been reported[4] when compared with state-of-the-art bulk materials. In addition, other properties such as flexible manufacturability, mechanical sustainability, and increased isotropy can be achieved by methods based on micro- and nanopowders.

In this paper our aim is to study hot isostatic pressing (HIP) and spark plasma sintering (SPS) as compacting methods for powders essentially similar to those described in Ref. 3. HIP is used to manufacture fully dense, near-net-shape components with isotropic properties. SPS is a method often used to consolidate thermoelectric materials due to its short processing time that prevents excessive growth of crystallites. As the resulting microscopic and nanoscopic structure of the samples is expected to differ between the methods, we perform comparative characterization of the samples. *n*-Type thermoelectric material with SPS sintering has been previously reported in Ref. 5, and *p*-type materials with both SPS and HIP in Ref. 6. Here we provide thermoelectric transport measurements, porosity characterization, and x-ray diffraction (XRD) measurements for both *n*- and *p*-type materials compacted with both SPS and HIP. We show that, with either method, a pair of *n*- and *p*-type thermoelectric materials with reasonable properties can be produced. We fit the transport data using a theoretical model, enabling us to divide the transport quantities into their components and also gain insight into charge carrier scattering mechanisms.

EXPERIMENTAL PROCEDURES

$(Bi_xSb_{1-x})_2(Te_ySe_{1-y})_3$ thermoelectric alloys were produced by mechanical alloying in an inert

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