Effects of Spark Plasma Sintering Conditions on the Anisotropic Thermoelectric

Properties of Bismuth Antimony Telluride

L. Han,^a[†] S. H. Spangsdorf,^a N. V. Nong,^a[†] L. T. Hung, ^a H. N. Pham,^a Y. Z. Chen,^a A. Roch,^b

L. Stepien,^b and N. Pryds^a

^a Department of Energy Conversion and Storage, Technical University of Denmark, Risø

Campus, Roskilde, Denmark.

^b Fraunhofer Institute for Material and Beam Technology (IWS), Dresden, Germany.

†e-mail of corresponding author: ihan@dtu.dk, ngno@dtu.dk

Table of Contents:

- 1. Supplement SEM images.
- 2. SPS sample cutting schematics.
- 3. Heat capacity C_p
- 4. Metal droplets squeezed out of graphite die after SPS sintering
- 5. Calculations of Lorenz number
- 6. Sintering profiles of samples sintered with variable holding time, pressure, and ramp-rate.

1. Supplement SEM images



Fig. S1 SEM of the Commercial Bi0.4Sb1.6Te3 powder



Fig. S2 SEM-EDS element mapping of the specimen sintered by SPS at 653K.



Fig. S3 SEM-EDS element mapping of the specimen sintered by SPS at 773K.

2. SPS sample cutting schematics.



Fig. S4 SPS sample cutting schematics. The red specimens were measured for in-plane thermoelectric properties, and the grey specimens were measured for out-of-plane thermoelectric properties.

3. Heat capacity C_p



Fig S5. Temperature dependence of the specific heat at constant pressure for Bi0.4Sb1.6Te3 samples, the red line represents the measured value by DSC, and the dotted line represents the calculated values by Dulong-petit law.

4. Metal droplets squeezed out of graphite die after SPS sintering



Fig. S6 Spilled droplets squeezed out of graphite die after SPS sintering at 723 K (left) and 773 K (right).

5. Calculations of Lorenz number L_0

According to the measured n and estimated m* values, a simple parabolic band model can be applied by employing the following equations:

$$F_{\lambda}(\xi) = \int_{0}^{\infty} \frac{x^{\lambda} dx}{1 + Exp(x - \xi)}$$
(S1)

$$L_{o} = \left(\frac{k_{B}^{2}}{q^{2}}\right) \left(3F_{0}F_{2} - 4F_{1}^{2}\right) / F_{0}^{2}$$
(S2)

where $F_{\lambda}(\xi)$ is the Fermi integral and ξ is the reduced electrochemical potential. λ is a scattering parameter and 0 is assumed for acoustic phonon scattering, 1 for optical phonons scattering, and 2 for ionized impurity scattering. $k_{\rm B}$ is the Boltzmann constant, q is the unit charge of electron. ξ is calculated from the measurement Seebeck coefficient (S) using the following equation:

$$S = -\frac{k_B}{q} \left[\frac{(2+\lambda)F_{\lambda+1}}{(1+\lambda)F_{\lambda}} - \xi \right]$$
(S3)

6. Sintering profiles of samples sintered with variable holding time, pressure, and ramprate.



Fig. S7. (a) Sintering profile for SPS sintering with variable holding time. (b) Mass density of the samples as a function of holding time. (c) Degree of orientation as a function of holding time.



Fig. S8 (a) Sintering profile for SPS sintering with variable pressure. (b) Mass density of the SPS sintered samples with variable uniaxial pressure. (c) Degree of orientation as a function of uniaxial pressure.



Fig. S9 (a) Sintering profile for SPS sintering with variable ramp-rate. The inset shows the current profile during SPS sintering. (b) Mass density of the samples sintered with variable ramp-rate. (c) Degree of orientation of the samples sintered with variable ramp-rate.