

Structure and Transport Properties of Bulk Nanothermoelectrics Based on $\text{Bi}_x\text{Sb}_{2-x}\text{Te}_3$ Fabricated by SPS Method

L.P. BULAT,^{1,6} I.A. DRABKIN,² V.V. KARATAYEV,² V.B. OSVENSII,²
YU.N. PARKHOMENKO,² M.G. LAVRENTEV,² A.I. SOROKIN,²
D.A. PSHENAI-SEVERIN,³ V.D. BLANK,⁴ G.I. PIVOVAROV,⁴
V.T. BUBLIK,⁵ and N.YU. TABACHKOVA⁵

1.—National Research University ITMO, St. Petersburg, Russia. 2.—GIREDMET Ltd., Moscow, Russia. 3.—Ioffe Physical Technical Institute, St Petersburg, Russia. 4.—Technological Institute of Superhard and New Carbon Materials, Troitsk, Russia. 5.—National University of Science and Technology “MISIS”, Moscow, Russia. 6.—e-mail: LBulat@mail.ru

Ball milling with subsequent spark plasma sintering (SPS) was used to fabricate bulk nanothermoelectrics based on $\text{Bi}_x\text{Sb}_{2-x}\text{Te}_3$. The SPS technique enables reduced size of grains in comparison with the hot-pressing method. The electrical and thermal conductivities, Seebeck coefficient, and thermoelectric figure of merit as functions of temperature and alloy composition were measured for different sintering temperatures. The greatest value of the figure of merit $ZT = 1.25$ was reached at the temperature of 90°C to 100°C in $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$ for sintering temperature of 450°C to 500°C. The volume and quantitative distributions of size of coherent dispersion areas (CDA) were calculated for different sintering temperatures. The phonon thermal conductivity of nanostructured $\text{Bi}_x\text{Sb}_{2-x}\text{Te}_3$ was investigated theoretically taking into account phonon scattering on grain boundaries and nanoprecipitates.

Key words: Thermoelectrics, bulk nanostructures, thermoelectric figure of merit, bismuth telluride, coherent dispersion areas, thermal conductivity

INTRODUCTION

Important scientific results on improvement of the thermoelectric figure of merit ZT using bulk nanostructures have been achieved during the last few years; see, for example, Refs. 1–4 ($ZT = T\sigma\alpha^2/\kappa$, where T is the absolute temperature and σ , κ , and α are the electrical and thermal conductivities and the Seebeck coefficient, respectively). In particular, promising materials for use in thermoelectric coolers from the points of view of cost and efficiency are bulk nanostructures based on $\text{Bi}_x\text{Sb}_{2-x}\text{Te}_3$ solid solutions.^{1–8}

Fabrication of bulk nanostructures should include two steps: preparation of nanoparticles and consolidation of nanoparticles (nanopowder) into a bulk

sample. Ball milling is the most widely known and used method of nanoparticle preparation.^{5–8} As the second step, hot pressing or spark plasma sintering (SPS) can be utilized. It was reported⁵ that $ZT = 1.4$ at temperature of 100°C was obtained in nanothermoelectrics based on $\text{Bi}_x\text{Sb}_{2-x}\text{Te}_3$, using ball milling with subsequent hot pressing with pulsed direct current (DC)⁵ (the laser flash method being utilized for thermal conductivity measurement). The value $ZT = 1.1$ was reached at $T = 80^\circ\text{C}$ to 90°C by using ball milling with subsequent hot pressing^{7,8} (the thermal conductivity being measured by the Harman method).

A possible way to reduce the average size of nanograins through the recrystallization process during hot pressing consists in fabrication of nanocomposites, i.e., bulk structures with inclusion of nanoparticles with another chemical composition.⁸ It has been recognized that the size of coherent dispersion areas (CDA) in such nanocomposites is