Performance assessment of nanocrystalline thermoelectrics using Comsol Multiphysics – a comparison study

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Introduction

Despite their low efficiency values of 5-8%, thermoelectric converters working in the Seebeck or Peltier mode are implemented in applications where it is not feasible or not practical to bring in electrical mains. From water coolers to space missions, it has been shown that solid state cooling and power generation using thermoelectric converters have important applications particularly in waste heat recovery. The goal of the work presented here is two-fold: a) to summarize current methods for fabricating high efficiency thermoelectric materials and, b) to compare the performance of common and new materials by using a Comsol model for thermoelectric generator with water cooling.

Theory / Experimental Set-up

The thermoelectric (TE) effect was discovered and studied by the German scientist Thomas Johann Seebeck beginning with 1821. Seebeck discovered that two different materials with junctions held at different temperatures deflect a compass needle. Due to the temperature difference, a current is produced. According to Ampere's law, the current produces a magnetic field, which in turn will deflect a magnet. The voltage produced is proportional to the temperature difference between the "hot" and "cold" contacts and independent of the distribution of temperature along the materials. The proportionality constant is the Seebeck constant S, a primary characteristic of TE materials. This phenomenon is the basis of thermocouples. The reverse is also true. In 1834, the French scientist Jean Charles Athanase Peltier found that a voltage difference applied to a junction of two different TE materials leads to a temperature difference (the Peltier effect), currently used in Peltier coolers. The proportionality constant P, is called the Peltier constant. Neither scientist gave a theoretical explanation of the phenomena. In 1854, Lord Kelvin (formerly William Thomson) explained the phenomena and the relationship between the coefficients using principles of the thermodynamics and in the process predicted the third thermoelectric effect, the Thomson effect. Briefly, when a temperature gradient is applied to a TE material, a current will flow through the material leading to heat absorption or emission. The amount of heat is proportional to the current as well as the temperature difference through a constant called the Thomson constant. Based on the work of Edmund Altenkirch and Abraham Fedorovich Ioffe, a parameter called "figure of merit", zT, was introduced in in 1956 in order to characterize the performance of a thermoelectric device.

$$zT = T(\frac{\sigma S^2}{k})$$

where σ is the electrical conductivity, k is the thermal conductivity, S is the Seebeck coefficient and T is the absolute temperature. While heavily studied and developed in the former USSR in the midlle of the 20th century, thermoelectric have undergone cycles of high and low interest. The advancement of nanomaterials fabrication in the early 1990s renewed the interest in TE materials as it was shown that the zT can be increased well above 1.

A thermoelectric generators is made of three basic components: an array of thermoelectric material with a simple rectangular box leg, electrical contacts to connect the legs in series or parallel and two parallel isolating plates that sandwich the other two components.

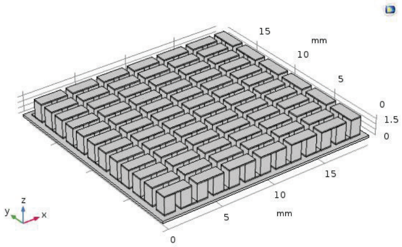


Fig. 1 A simplified schematic for a commercial TEG in series connection, drawn here without the top face

The most studies method to increase the thermoelectic efficiency is to increase the value of the Seebeck coefficient by changing the way they are fabricated. W. Li et al developed thin films of n-type and p-type thermoelectric materials and have shown increased Seebeck coefficients and power factors (2). Other groups developed complex thermoelectric materials with high zT at very high temperatures such as skutterudites and Zintl phases by using novel technologies in nanostructural engineering (1). High zT materials have also been developed by incorporating a matrix of nano-particles (3-12). Increased electrical conductivity materials have also been developed in the form of single or multi-wall carbon nano-tubes or graphene (13-15). Finally, a most exciting development in photonic topology showed that new thermal metamaterials can control the emission of a hot object by enhancing a certain portion of the spectrum and reducing the rest (16). With such material interfaces the spectrum emitted by a hot surface can be tailored to the match the energy gap of the thermoelectric material therefore promoting more charge carriers to the conduction band and reducing heat waste.

The work presented here is based on a model of a water-cooled thermoelectric generator (17, 18, 19) developed for the purpose of comparing the electric potential generated by common thermoelectric materials such as Bi_2Te_3 and PbTe with novel nanocrystalline alloys of Bismuth Antimony Telluride alloys fabricated via different methods. Various configurations for the TEG faces and electric contact materials are also investigated.

Experimental Set-up

The Comsol model is a replica of a 5cmx5cm commercial TEG module (tegpro.com) with 1mmx1mmx1.5mm (WxDxH) thermoelectric blocks connected in series. The electric contacts are 1.5mmx1mmx0.1mm. The blocks and contacts are sandwiched between two face sheets, 50mmx50mmx1mm. A water cooler with an inlet and outlet (tegpro.com) is sandwiched between the TEG and an aluminum heatsink. The system is enclosed in a rectangular box to allow for convection cooling by air. The computer model was compared to the standard Bi_2Te_3 commercial module for a variety of hot temperature values. The water cooler was fed by a sink with the temperature of the inlet T_{inlet} = 19degC. The experimental data was used as input parameters for the Comsol model and reported previously (17).

Governing Equations / Numerical Model / Simulation / Methods / Use of Simulation Apps

The program uses the Heat Transfer Module for a stationary study. We coupled the Laminar Flow and Heat Transfer interfaces by adding a "Nonisothermal Flow" Multiphysics node and included the Thermoelectric effect. The equations describing the heat transfer by conduction and convective cooling, the electric currents and the Joule coupling, as well as the thermoelectric effect are fully described in reference (27) which was used as an example to develop the thermoelectric calculations. The program solves for heat transfer by conduction at all interfaces and water convection. The Solver was configured for the Automatic(Newton) method which significantly reduced the computation time. The materials physical constants were taken from Comsol Materials database for Bi2Te and PbTe. New materials with physical properties found in literature and/or theoretically estimated were added.

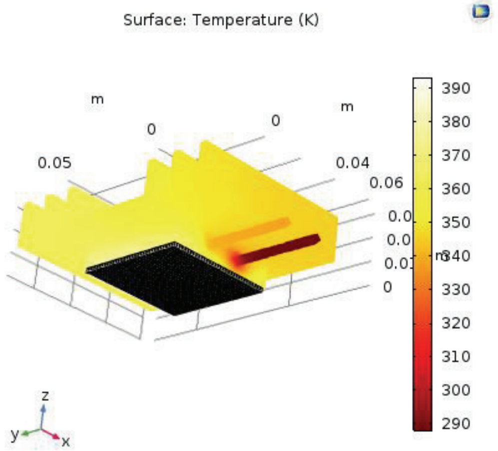
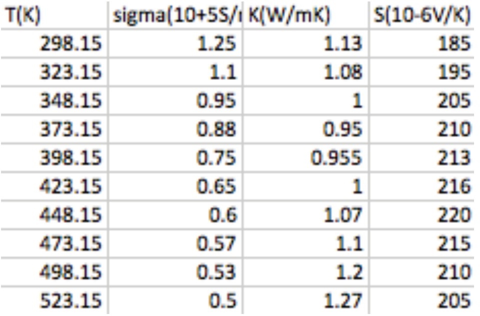


Fig. 2 Temperature graph for the complete 3-component model with TEg module, water cooler and heat sink.

Experimental Results / Simulation Results / Discussion

The bismuth antimony telluride alloys selected for the purpose of comparison are alloys that have a high zT calculated using experimental values of the physical constants k, σ and S (3, 11). They are the following:

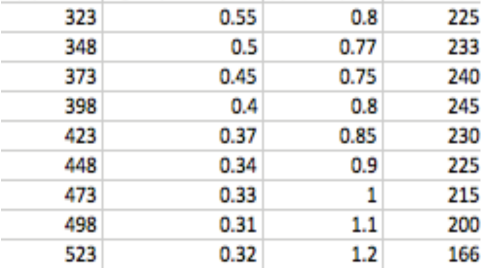
1. Nanostructures Bismuth Antimony Telluride bulk alloys $Bi_{0.5}Sb_{1.5}Te_3$ (11) fabricated from ball-milled and hot-pressed crystalline ingots under inert conditions.



T(K)	sigma(10+5S/	K(W/mK)	S(10-6V/K)
298.15	1.25	1.13	185
323.15	1.1	1.08	195
348.15	0.95	1	205
373.15	0.88	0.95	210
398.15	0.75	0.955	213
423.15	0.65	1	216
448.15	0.6	1.07	220
473.15	0.57	1.1	215
498.15	0.53	1.2	210
523.15	0.5	1.27	205

Table 1. Thermoelectric constants for Nanostructures Bismuth Antimony Telluride $Bi_{0.5}Sb_{1.5}Te_3$ bulk alloys (11)

2. Bulk nanocrystalline $Bi_{0.4}Sb_{1.6}Te_3$ solid solution (3) fabricated by Spark Plasma Sintering.



T(K)	sigma(10+5 S/m)	K(W/mK)	S(10-6 V/K
323	0.55	0.8	225
348	0.5	0.77	233
373	0.45	0.75	240
398	0.4	0.8	245
423	0.37	0.85	230
448	0.34	0.9	225
473	0.33	1	215
498	0.31	1.1	200
523	0.32	1.2	166

Table 2. Thermoelectric constants for $Bi_{0.4}Sb_{1.6}Te_3$ bulk alloys (3) fabricated by Spark Plasma Sintering at 450K

3. B_2Te_3 and PbTe from the Comsol materials database (19)

The first data set obtained is the electric potential generated as a function of the hot surface temperature for four thermoelectric materials, Bi_2Te_3, PbTe, nanostructured $Bi_{0.5}Sb_{1.5}Te_3$ and $Bi_{0.4}Sb_{1.6}Te_3$. The data shown in Fig. 3 is for a TEG that has Copper electrical contacts and graphite faces. Fig. 3 below shows the electric potential for these materials for the full TEG module with both water and heat sink cooling. The cold water inlet is maintained at 291.15K. $Bi_{0.4}Sb_{1.6}Te_3$ is the best performing material, exceeding the electric potential of the common Bi_2Te_3 and PbTe by 34% at 393K. At T=393K, $Bi_{0.5}Sb_{1.5}Te_3$ generates an electric potential 11% higher than Bi_2Te_3 at 393K, making it the he second best performing material. The second set of data is represented in Fig. 4. It summarizes the performance for the same ranger of T but Aluminum contacts instead of Copper. As expected, $Bi_{0.4}Sb_{1.6}Te_3$ remains the top performer, however, the percent difference is slightly less than the configuration with Copper as the elextric conductivity of Cu is higher than Al.

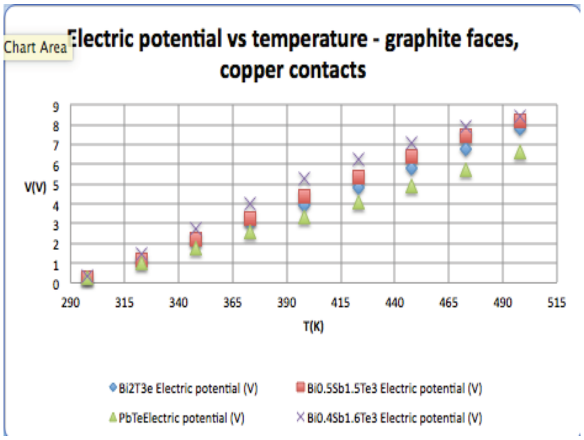


Fig. 3 Electrical potential versus temperature for TEG with Cu contacts and graphite faces. T inlet water = 292K.

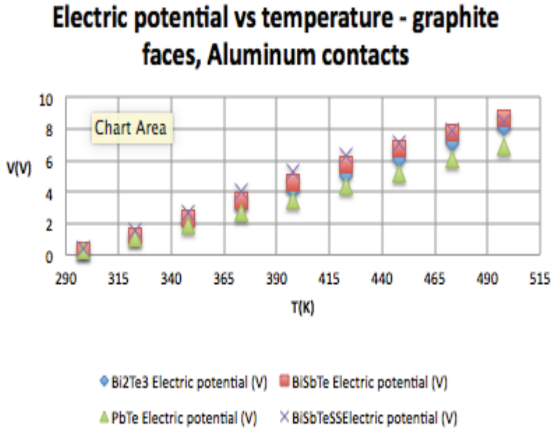


Fig. Electrical potential versus temperature 4 TEG with Al contacts and graphite faces. T inlet water = 292K

Conclusions

SPS sintered $Bi_{0.4}Sb_{1.6}Te_3$ and Nanostructured $Bi_{0.5}Sb_{1.5}Te_3$ prove to be superior novel thermoelectrics under water cooling conditions for temperatures up to 498K. The fabrication method for $Bi_{0.5}Sb_{1.5}Te_3$ is less elaborate and less expensive than the SPS method. Both materials increase have superior performance in the temperature range of many practical applications. Several aspects could be further investigates such as the specific heat capacity at constant pressure which might be slightly different between the two bismuth antimony telluride alloys considered as well as small differnces in the physical density due to the fabrication process.

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