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Characterization of Thermoelectric Power Generation Modules Made from New Materials

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ABSTRACT

Lead-Antimony-Silver-Tellurium (L-A-S-T) materials, synthesized at Michigan State University, show promising thermoelectric properties at high temperatures for use in power generation applications. Recent scaled-up quantities of L-A-S-T show a ZT=1.4 at 700 K approaching the figure of merit for samples made in small quantities [1]. These materials are of great interest for power generation applications with hot side temperatures in the range of 600-800 K. Developing these materials into working devices requires minimization of the thermal and electrical parasitic contact resistances, so various fabrication methods are under investigation. To examine each method, a new measurement system has been developed to characterize these devices under various load and temperature gradients. An introduction to the system will be presented, as well as results for devices made of the L-A-S-T materials.

INTRODUCTION

Thermoelectric generators convert energy from heat into electricity by existence of the Seebeck effect. An applied temperature gradient across the generator will force heat to flow from the hot to cold side by thermal conduction while some of this heat is converted to electricity. The generator efficiency, η, is determined by comparing the amount of electricity produced, P_{out}, to the total amount of heat induced, Q_{in}.

$$\eta = \frac{P_{out}}{Q_{in}}$$

To accurately determine the efficiency of a generator, the heat induced and electrical power produced must be carefully measured. Generator efficiencies are often calculated from the measured material properties [9]. This method indirectly yields generator efficiency through the figure of merit, requiring separate measurements of absolute Seebeck, electrical and thermal conductivities of which each are susceptible to a tolerance of errors. More direct measurements of the figure of merit have been demonstrated and are useful to obtain generator efficiency, but the measurement accuracy is limited to testing under small temperature gradients [2,3,4]. Thermoelectric generator efficiency can also be determined through comparative heat flow, and this method offers a more direct and realistic measurement of a generators efficiency [5].

EXPERIMENTAL SETUP

Figure **1** shows the method of measuring generator efficiency by comparative heat flow by which a generator is sandwiched between two standard reference materials (SRM) of known thermal conductivity. A heater source and sink induce a uniform heat flow through the reference materials and generator. For the system presented here, heat flow is monitored into and out of the generator, as well as the electrical power out of the generator when a load is attached.

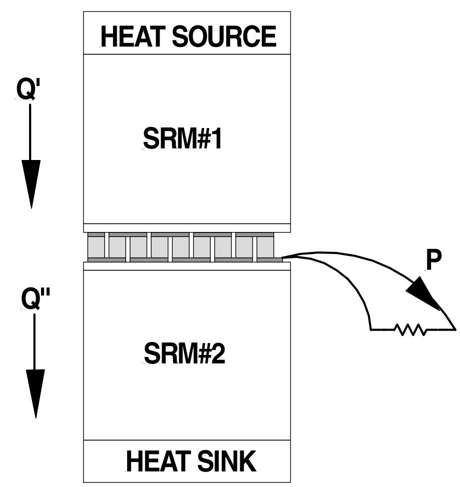

Figure 1. Generator measured efficiency by absolute heat flow.

$$\eta = \frac{P}{Q'} = \frac{P}{P + Q''}$$

SRM#1 is 304 stainless steel and SRM#2 is OFHC copper. Each SRM acts as a heat flow meter measuring heat into and out of the generator under test. A molybdenum shielded heater was used to provide the temperature gradient, and 1 mil type E thermocouples monitor the temperature gradients across the SRM's with thermocouples referenced by a platinum resistor. The heat flow is calculated from the measured temperature gradients and reference thermal conductivity data on these materials [6,7]. This method of measuring heat flow assumes conductive heat flow so the heater, heat flow meters and generator were all chosen to be of same cross sectional area to ensure uniform heat flow. Serious errors in heat flow measurements can be introduced due to convection and radiation effects, so generators are tested under a vacuum of 5µTorr using mirror finished heat flow meters. All were bonded together using silver paint.

VERIFICATION OF THE SYSTEM

Generator efficiency requires measurements of heat flow through it and electrical power generated. The electrical power is calculated from using an electronic load where the current through the load and module is measured using a Keithley instruments model 2400 source meter, and load voltage with a model 2002 multimeter. To determine the accuracy of heat flow measurements, the temperature gradient was measured across a fused quartz sample and heat flow calculated from its thermal conductivity [8]. The quartz sample was measured under varied temperature gradients providing 0-10W of heat flow, similar to the testing conditions for the thermoelectric generators in future testing.

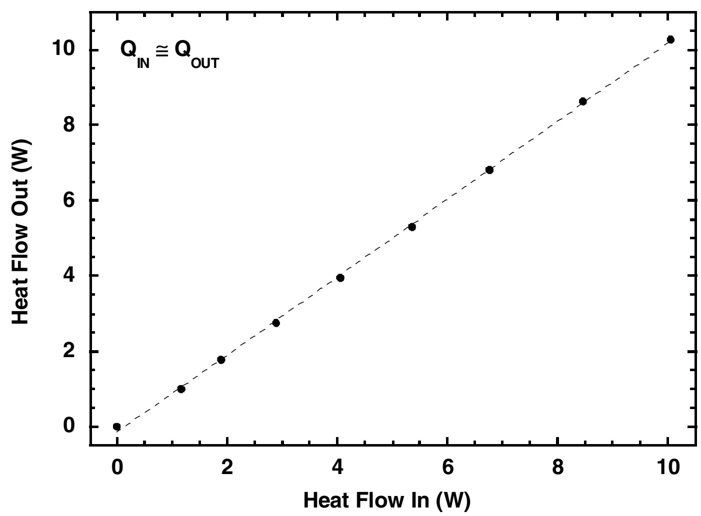

Figure 2. Heat flow verification.

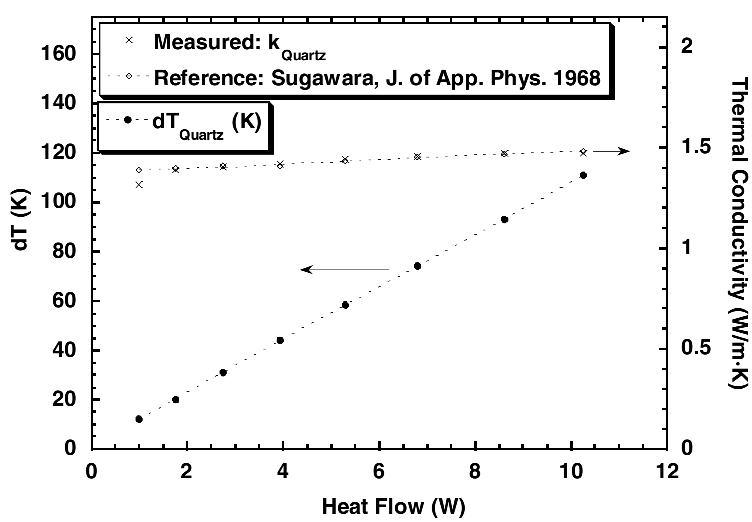

Figure 3. Heat flow measurement with quartz.

Measurements were taken on a 16mm^2 Bi_2Te_3 cooling module, and agreed well with reference data. Silver paint was used to adhere the heat flow meters to the module to provide a good thermal contact with negligible temperature gradients compared to module and heat flow meters. Thermal grease was initially used to adhere the module, but it was found that serious errors in temperature gradient measurements arise due to thermal gradients across the grease if great care is not taken to ensure a very thin layer. The measurements were taken under vacuum of 1µTorr. Each curve of Figure 4 is taken with a fixed heater power supply. The heater was designed to provide heat effectively, but not as efficiently as other designs that use guard heaters and calculate heat flow from the delivered supply power. Therefore, the power supplied to the heater is not of interest, rather the heat flow determined by the two heat flow meters.

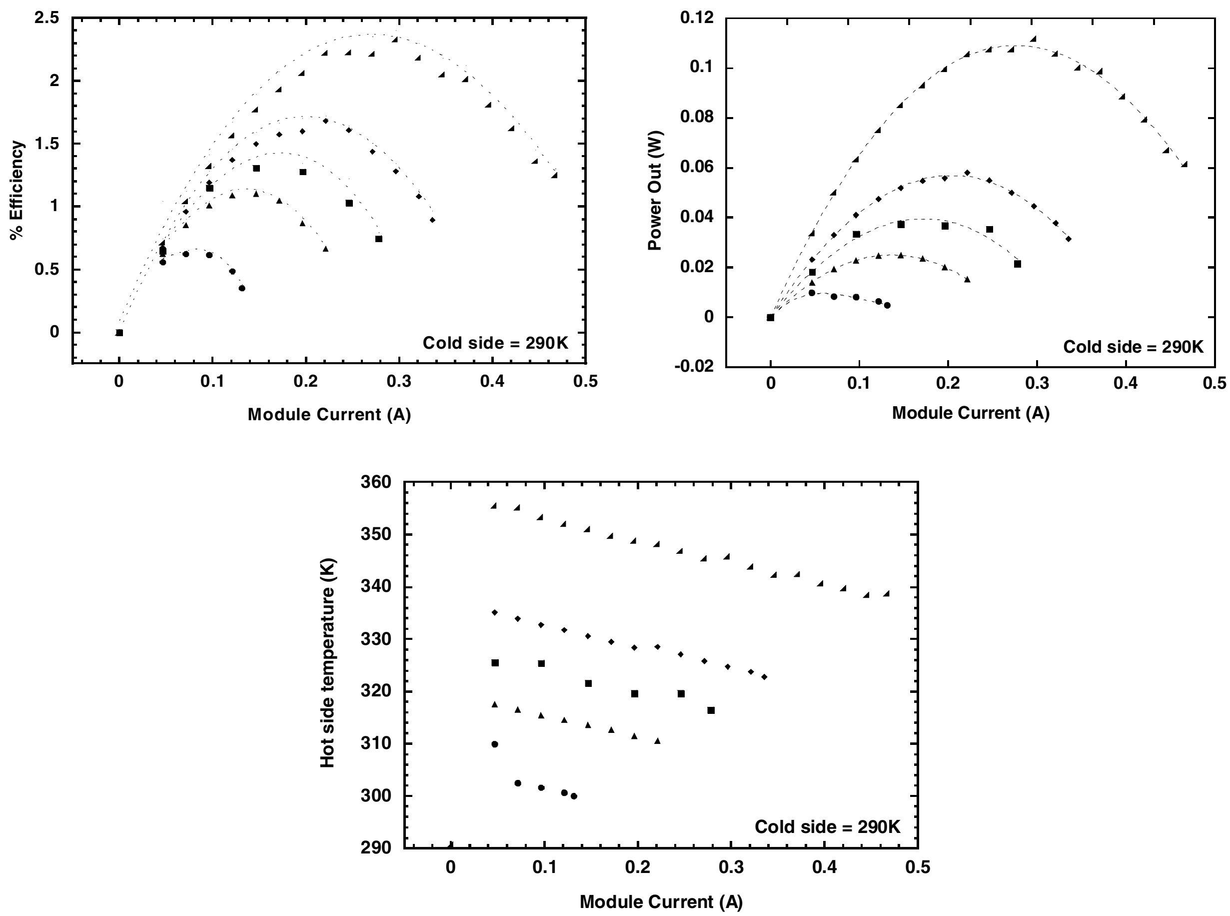

Figure 4. Measurements on a Bi_2Te_3 cooling module.

Efficiencies were calculated by two different methods. Method 1 uses the material properties of N- and P-type Bismuth Telluride and a one percent contact resistance. Method 2 uses a direct measurement of a similar modules ZT at room temperature under a small temperature gradient [4]. At peak efficiency,

$$\eta_{Calc1} = \eta|_{R_{Module}=R_{Load}} = \frac{\Delta T}{T_{hot}} \frac{\sqrt{1+ZT}-1}{1+{T_{cold}}/{T_{hot}}} \tag{1}$$

Implementing a one percent contact resistance, measured temperature conditions, and using typical N and P-type Bi_2Te_3 materials properties, the efficiencies have been calculated [9].

$$\eta_{Calc2} = \frac{\mu\eta_c}{\frac{(\mu+(1+\delta))^2}{ZT_{hot}} + (\mu+(1+\delta)) - \frac{\eta_c(1+\delta)}{2}}$$

where $\eta_c = \frac{T_{hot} - T_{cold}}{T_{hot}}$, $\mu = \frac{R_{Load}}{R_{Module}}$ and $\delta = \frac{R_{Contacts}}{R_{Module}}$

The measured results are shown in comparison with results calculated.

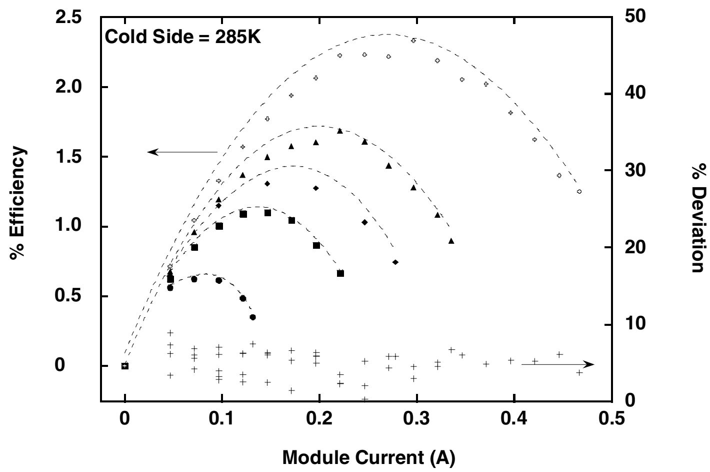

Figure 5. Five percent average deviation in efficiency measurements, dashed lines are calculated values.

The slight deviation can be attributed to testing under larger gradients where ZT shifts from it's peak value as a cooling device, to a lower value tested at higher average temperature conditions.

MEASUREMENTS ON L-A-S-T GENERATORS

High temperature thermoelectric compounds have been synthesized at Michigan State University and show excellent properties for use in power generation applications. Previous research at MSU focused on identifying new materials for power generation applications, whereas present research additionally addresses fabricating some of the more promising materials, like L-A-S-T, into working generators. Thermoelectric generators are being produced from these materials and have been tested under varied load and gradient conditions. Fabrication and investigation of producing ohmic contacts to these materials are being investigated elsewhere [10]. While the best contacts have yet to be identified, generators have been tested with present contact methods and scaled-up material quantities to track the progress of develop L-A-S-T based generators.

L-A-S-T modules have been fabricated and measured results agree well with the expected performance. For example, one module was tested with a 30 percent room temperature contact resistance. Its efficiency was calculated for the measured testing conditions and material properties, and data is provided in figure 6 below. Data collection had started for this module with a load of 160mΩ per unicouple, and the measured result of 1.25% agreed quite well with expected 1.37%. This module was thermally cycled during the test, however the electrically insulating plate of alumina bonded to the metal unicouple strips broke inhibiting further data collection. Nevertheless, L-A-S-T modules are being fabricated and tested under large temperature gradients with expected device performance given the less than ideal fabrication processes.

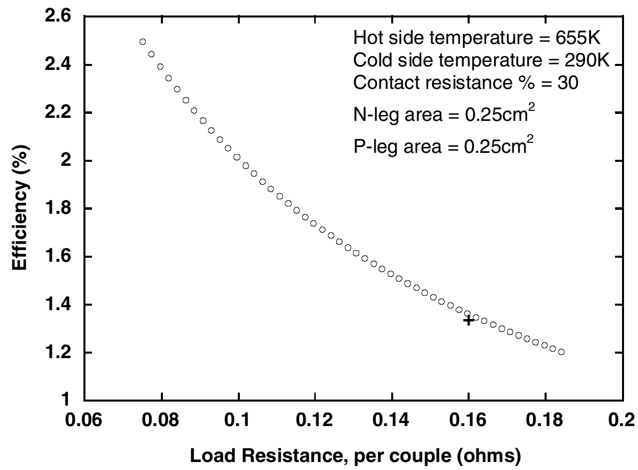

Figure 6: Calculated versus Measured Efficiency for a LAST module.

The modules fabricated to date with L-A-S-T have much lower efficiencies than initially expected. Some reasons can be attributed to the cross sectional area of legs not optimized for appropriate current densities, high contact resistances, and material properties different for the large scale up ingot materials used in these modules than data presented elsewhere [1].

CONCLUSION

A new measurement system has been developed to characterize L-A-S-T based generators under various load and temperature gradients. The system accuracy has been verified with reference materials, and is ready to continue use for investigation of fabricating devices made of the L-A-S-T materials. Recent scaled up quantities of L-A-S-T have different properties than the initial smaller ingots, and present studies are progressing to make ingots with higher yields of the smaller ingots high performance characteristics. Furthermore, consistent fabrication of low resistance contacts is being investigated to improve the measured efficiencies of LAST generators.

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