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Screen printed flexible $\text{Bi}_2\text{Te}_3\text{-Sb}_2\text{Te}_3$ based thermoelectric generator

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Abstract. This paper reports the fabrication and testing of Bismuth Tellurium (Bi_2Te_3) – Antimony Tellurium (Sb_2Te_3) based thermocouples using screen printing technology. In this study, screen printable thermoelectric pastes were developed and the transport properties of cured material were measured. The dimension of each planer thermoleg is $39.3 \text{ mm} \times 3 \text{ mm}$ with a thickness of $67 \text{ }\mu\text{m}$ for Bi_2Te_3 leg and $62 \text{ }\mu\text{m}$ for Sb_2Te_3 leg. A single thermocouple with this dimension can generate a voltage of 6 mV and a peak output power of 48 nW at a temperature difference of 20°C . The calculated Seebeck coefficient of a single thermocouple is in the range of $262 - 282 \text{ }\mu\text{V/K}$. The Seebeck coefficient at room temperature were measured to be $-134 - -119 \text{ }\mu\text{V/K}$ and $128 - 134 \text{ }\mu\text{V/K}$ for Bi_2Te_3 and Sb_2Te_3 respectively. This work demonstrates that the low-cost screen printing technology and low-temperature materials are promising for the fabrication of flexible thermoelectric generators (TEGs).

1. Introduction

Based on Seebeck effect, the thermoelectric modules have ability to directly transfer heat into electric energy. Thermoelectric Generators (TEGs) are very attractive devices for harvesting waste thermal energy applications, for instance, powering wireless autonomous sensors [1]. The TEG devices have advantages such as silently operation, no moving parts and high reliability. It is the best approach for powering the devices in locations where there are poor levels of illumination but sufficient waste heat.

In order to evaluate the performance of a thermoelectric material, the figure of merit ZT ($Z = \alpha^2 / (\rho \cdot \lambda)$, where α is the Seebeck coefficient, ρ is the electrical resistivity, λ is the thermal conductivity and T is the temperature) is applied. In room temperature range (293 K to 303 K), Bismuth Tellurium/Antimony Tellurium ($\text{Bi}_2\text{Te}_3/\text{Sb}_2\text{Te}_3$) system has the highest ZT value among reported thermal active materials [1, 2]. Several papers have reported fabricating flexible $\text{Bi}_2\text{Te}_3/\text{Sb}_2\text{Te}_3$ based TEGs using different fabrication technologies (e.g. dispenser printing, photolithography and electrochemical deposition) [2, 3]. However, the energy transfer efficiency of conventional rigid TEGs is only around 5% [4]. Flexible TEGs are supposed to have even less transfer efficiency. Hence, in order to achieve high power output, large quality of TEG devices need to be electrically connected together. Then, an easy fabrication process is essential to low down the cost.

Screen printing is a low-cost process that is well suited for large area fabrication. It involves the deposition of synthesized thermoelectric inks that consist of thermoelectric material powders in a binder and solvent matrix. After printing only a curing process is required. The simplicity and low energy consumption is quite attractive for fabricating TEGs. The Seebeck effect and structure of a screen printed thermocouple is illustrated in figure 1. In this work, screen printable pastes filled with thermoelectric



materials were formulated. A planer TEG with 4 thermocouples was fabricated by screen printing technology and the thermal performance tested.

2. Experimental

2.1. Material synthesis

The screen printable thermoelectric pastes reported here contains thermoelectric materials Bi_2Te_3 and Sb_2Te_3 alloy powders and an epoxy binder system. A solvent was also added in to the paste to adjust the viscosity to a screen printable level.

For n-type bismuth tellurium alloy, the ratio of tellurium (Te) affects the thermoelectric performance of the device. When the atomic ratio of Te is a 64%, the bismuth tellurium alloy will have the highest negative Z value [5]. The actual formulation of this n-type semiconductor material is $\text{Bi}_{1.8}\text{Te}_{3.2}$. The powders are from Testbourne Ltd, with the size of 325 mesh and a purity of 99.99%.

An EXAKT 50 triple roll mill was used to mix the paste to achieve a uniform particles distribution. The viscosity range of these screen printable pastes is from 9000 cP to 15000 cP, measured by Brookfield High Shear CAP-1000+ viscometer.

2.2. Device fabrication

The printer used was DEK-248 screen printing machine. A polyester screen with 325 mesh was used for deposition. Kapton was used as the substrate.

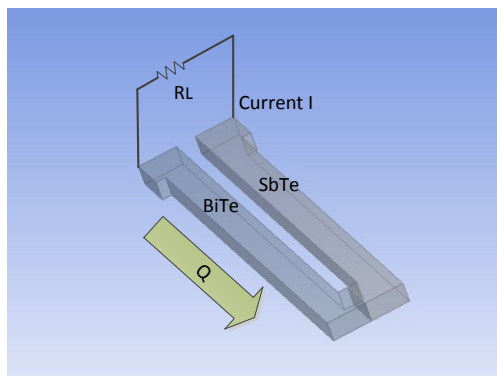


Figure 1. Operation principle of screen printed $\text{Bi}_2\text{Te}_3/\text{Sb}_2\text{Te}_3$ thermocouples

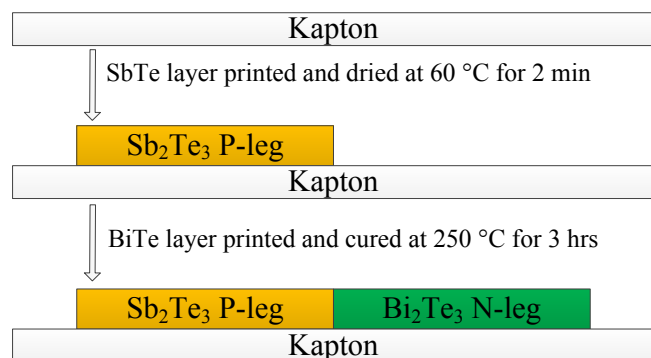


Figure 2. Fabrication process of screen printed $\text{Bi}_2\text{Te}_3/\text{Sb}_2\text{Te}_3$ thermocouple

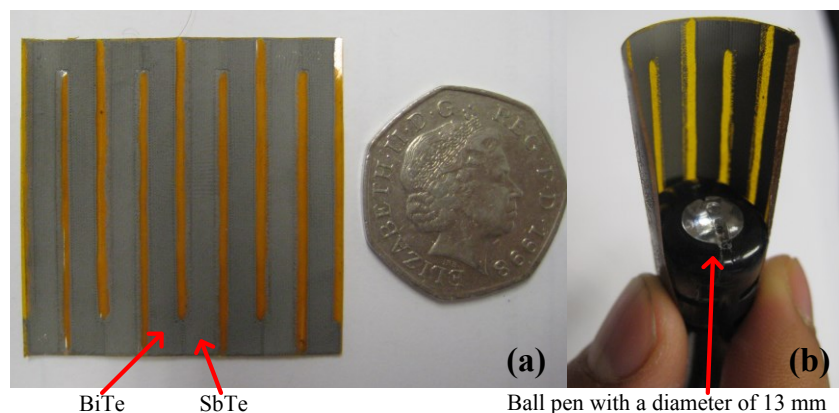


Figure 3. Printed $\text{Bi}_2\text{Te}_3/\text{Sb}_2\text{Te}_3$ thermocouples (a): sample size comparing with a 50 pence coin; (b): sample rolling on a 13 mm diameter ball pen

The screen printed $\text{Bi}_2\text{Te}_3/\text{Sb}_2\text{Te}_3$ fabrication process is shown in figure 2. It only involves deposition of two thermal material layers and a curing process. The thickness of the active thermoelectric layers can be built up by drying each layer at 60°C to evaporate the solvent and then repeating the printing step. After the two thermoelements have been deposited, they are cured at 250°C for 3 hours. The screen printed sample with 4 thermocouples is shown in figure 3.

2.3. Printed Thick-films and Device Characterization

The surface and the cross-section of the screen printed films were observed in an EVO Scanning Electron Microscope (SEM). The SEM images in figure 4 show the homogenous mixture of the powders and binder. Also, the average particle size can be identified to be around $10\ \mu\text{m}$.

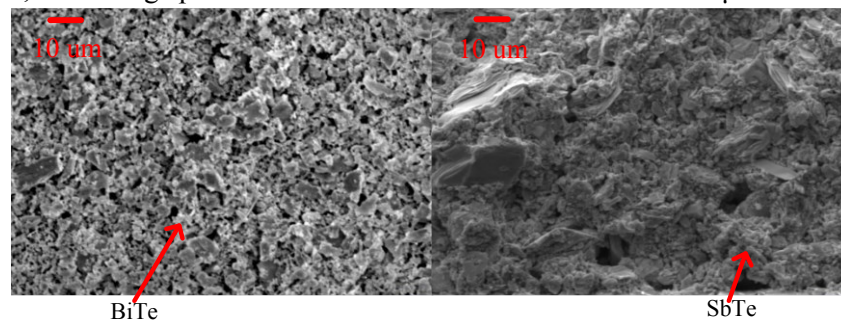


Figure 4. SEM images of printed thick-film. Left: surface of BiTe thick-film; Right: cross-section of SbTe thick-film

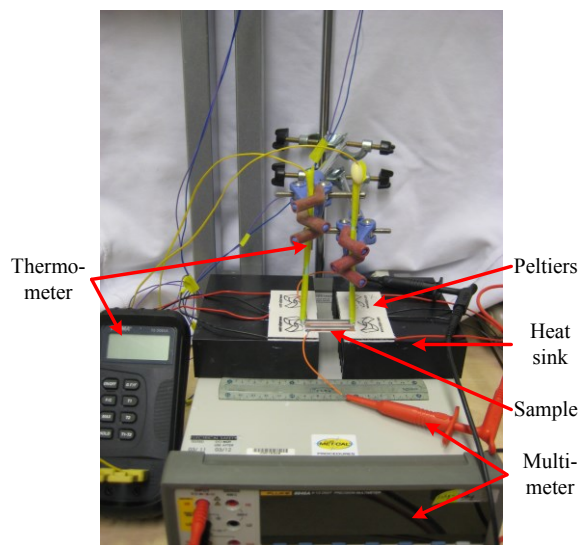


Figure 5. Seebeck Voltage measurement setup

Table 1. Seebeck coefficient of screen printed Bi_2Te_3 and Sb_2Te_3 thick-films

α ($\mu\text{V/K}$)	Bi_2Te_3	Sb_2Te_3
Day 1	-134 – -119	128 – 134
Day 100	-132 – -129	132 – 137
Bulk value [6]	-227	110

From the introduction it can be seen that the figure of merit consists of three parameters: the Seebeck coefficient α , the electrical resistivity ρ and the thermal conductivity λ . In this experiment, Hall Effect measurements were performed to test the transport properties of the printed thick films, including resistivity, using a commercial Hall Effect measurement system (HMS 3000 from Ecopia). The Seebeck coefficients of all the material samples were measured using custom-made equipment. For these measurements $1\text{ cm} \times 1\text{ cm}$ square samples were printed and tested.

The Seebeck voltage of the printed sample devices were tested by a bespoke set-up shown in figure 5. Once the device reached steady state with a constant temperature gradient, the Seebeck voltage of the device was measured using a digital multimeter. Peltier modules from European Thermodynamics provided the thermal gradient across the device. The temperature gradient across the sample was using

a Tenma digital thermometer with dual type K input. The gap between the two peltiers was fixed at 2 cm. All mechanical interfaces were sealed with thermal compound to reduce the heat loss. The electrical power output from the thermocouples was calculated from the voltage across a variable load resistance in series. All test were processed in open air at room temperature.

3. Results

3.1. Thermoelectric properties of printed thick-films

The Seebeck coefficient measurement results in comparison with the bulk value are shown in table 1. The negative sign of Seebeck coefficient indicates that the $\text{Bi}_{1.8}\text{Te}_{3.2}$ alloy powders acting as n-type material in a thermocouple, while the positive α value means that the Sb_2Te_3 alloy is p-type thermoleg. The absolute α value of $\text{Bi}_{1.8}\text{Te}_{3.2}$ thick-film is smaller than that of the bulk material, while the Seebeck coefficient of Sb_2Te_3 thick-film is higher than the bulk alloy. This is compatible with the results reported by Chen et al. [3]. The variation of the average α value is with 5% within 100 days.

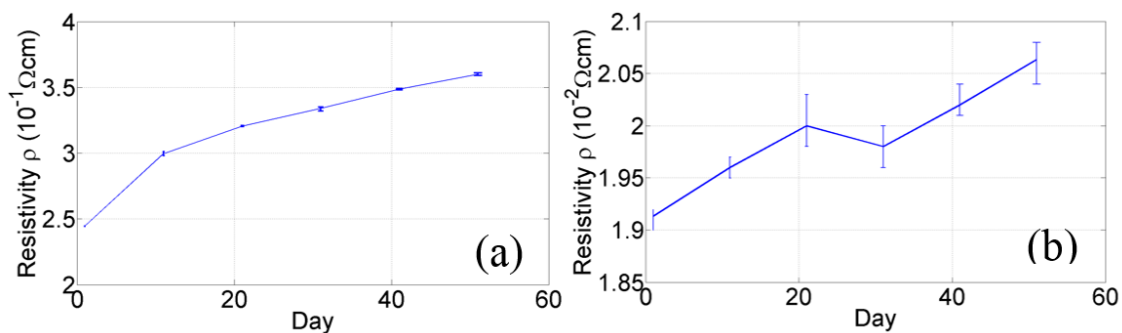


Figure 6. Resistivity of printed thick-films varying with time. (a): $\text{Bi}_{1.8}\text{Te}_{3.2}$; (b): Sb_2Te_3 .

Figure 6 shows the resistivity of both thick-films and their variation with time. Both materials also show an increase in resistivity with time, while this is larger for the BiTe material. In 50 days, the $\text{Bi}_{1.8}\text{Te}_{3.2}$ thick-film shows an increase ρ of 47% compared with the original value. For Sb_2Te_3 thick-films, the variation is only 7.8% over 50 days.

3.2. Device characterization

Figure 7 shows the tested Seebeck voltage of 4 screen printed BiTe/SbTe thermocouples. A single Bi_2Te_3 - Sb_2Te_3 thermocouple can generate 17 mV voltage at a temperature difference of 60 °C, which is better than the 5 mV of a single Bismuth-Tellurium thermocouple achieved by our group at the same temperature difference [6]. From the definition of Seebeck Effect ($\alpha = V/\Delta T$, where V is the Seebeck Voltage, ΔT is the temperature difference), for a single thermocouple of this sample, the Seebeck coefficient is in the range of 262–282 $\mu\text{V/K}$ (figure 8).

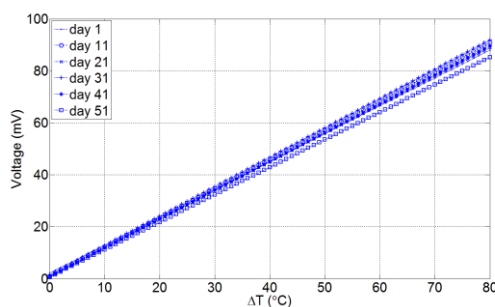


Figure 7. Generated Seebeck voltage of four printed thermocouples, varying with time

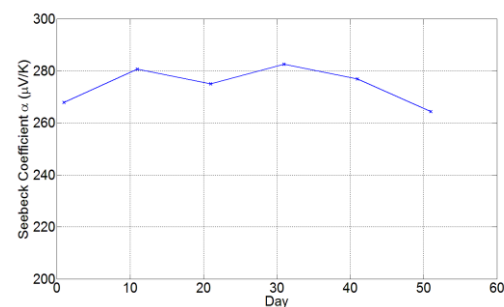


Figure 8. Calculated Seebeck coefficient for single thermocouple varying with time

The power output versus load resistance at a temperature difference of 20 °C is shown in figure 9. The voltage across the load increases with the increasing load resistance and the maximum output power occurs when the load resistance matched the TEGs resistance. The power output P is calculated by $P=V^2/R$, where V is the voltage across the load resistance and R is the load resistance. For a single thermocouple, the original power is 48 nW. This value decreased to 23 nW after 50 days.

The reason for the low power mainly comes from the high resistivity of the BiTe thick-film. From figure 4 it can be seen that there are pores existing between nearby particles, which prevent a better Ohm contact between particles.

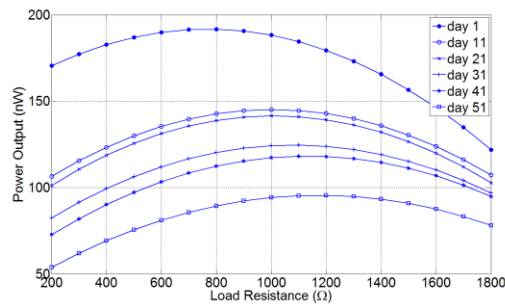


Figure 9. Power output of 4 printed BiTe/SbTe thermocouples varying with load resistance

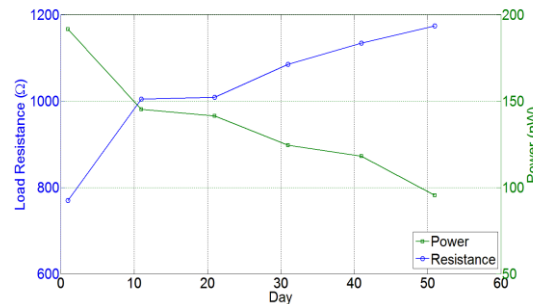


Figure 10. Maximum power output and self-resistance of 4 printed BiTe/SbTe thermocouples varying with time

4. Conclusion

This work demonstrates that the low cost screen printing technology can be used to fabricate BiTe/SbTe based TEGs for room temperature energy harvesting applications. Although the power output is low for a single thermocouple, the flexibility allows the screen printed TEGs to be connected in series and rolled in to a coil. Then, higher power output can be achieved by deploying more thermocouples in this way. Future work includes the measurement of thermal conductivity which will enable the calculation of ZT.

However, the high and increasing electrical resistivity of $\text{Bi}_{1.8}\text{Te}_{3.2}$ thick-film is an issue for reaching a higher power output for this screen printed TEG. Oxidization of bismuth tellurium powders is the likely cause. Future work will explore the optimization of design and material processing to decrease the resistivity. To reduce the resistivity variation and prevent oxidization, an encapsulation layer on top of the printed thermal materials layer will be investigated.

5. Acknowledgment

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