# Realization of High Performance and Flexibility Thermoelectric Module via Hybrid Carbon-Based Materials and Bi<sub>2</sub>Te<sub>3</sub> Alloy

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#### Abstract

Thermoelectric devices performance depends on device engineering minimizing heat loss as well as inherent material properties. Considering that the limited flat or angular shape of devices, a considerable amount of heat loss is inevitable. To address this issue, we demonstrate that the flexible thermoelectric module made by thermoelectric ink. The thermoelectric ink has successfully realized the in-situ growth of bismuth Tellurium alloy on the surface of carbon-based materials and then fabricated a series hybrid material by utilizing the special interaction between metal Cu, carbon-based materials, and polyimide. Due to the high ratio of homogeneously dispersed molecular carbon-based materials inside the bismuth Tellurium alloy matrix, a record-low thermal conductivity has been achieved.

## 1. Introduction

Now with the energy demand is getting increased, advancing renewable energy technology development and conducting research is indispensable. Among the several renewable energies, thermoelectric modules (TEM) [1-3] can be used not only to convert electrical energy to thermal energy but also to convert thermal energy to electrical energy. These include following advantages: clean energy, small volume, no noise, low failure rate, and without global warming gas emission. The thermoelectric material property depends on the dimensionless figure of merit ZT, which is defined as ZT =  $S^2 \sigma T \kappa^{-1}$ .  $S^2 \sigma$  is the power factor (PF), S is Seebeck coefficient (V/ $\Delta$ T),  $\sigma$  is electrical conductivity, T is absolute temperature, and  $\kappa$  is thermal conductivity. Additionally, the number of wearable devices users is increasing owing to IoT (Internet of Things) epoch approaching; there include smart watches, smart glasses even the smart clothes and sensors for medical and sports monitoring. Many wearable devices developers are focusing on how to reduce the power dissipation and how to import the renewable energy to be the power source in the device. Thus, flexible thermoelectric module (FTEM) is an attractive approach because of their ability and suitability to power miniature electrical devices for wearable applications.

## 2. Experimental

The early papers already showed lots of flexible thermoelectric modules via organic thermoelectric materials like PE-DOT:PSS. They almost had very low power output, because of low figure of merit ZT organic thermoelectric materials. Due to this reason, we think hybrid the high ZT inorganic thermoelectric materials and organic materials, that it has the possibility to keep flexibility and well power output. Carbon Nano-tube (CNT) is a very special one-dimensional material with interesting physical, chemical, and mechanical properties. It has very high carrier mobilities, large aspect ratio, super-hydrophilic properties, high thermal conduction, and high strength, and thus has been used to fabricate various composites. However, the high surface energy and flexible character of CNTs make them hard to be separated. The challenge to use CNTs for TE and other applications is developing a technique to uniformly distribute the CNTs in the matrix material. In fact, due to the presence of free p electrons, the CNTs' surface has a high chemical reactivity and it can interact with many polymers, oxides, and even pure metals. These interactions could be used to separate the agglomerated CNTs and effectively disperse them in the host matrix material. In this study, sintered polycrystalline sample of Bi<sub>2</sub>Se<sub>0.3</sub>Te<sub>2.7</sub> and Bi<sub>0.5</sub>Sb<sub>1.5</sub>Te<sub>3</sub> were prepared as follows. Stoichiometric ratio mixtures of Se (Sb), Bi and Te with purities of 99.999% were melted in a sealed vacuum quartz tube at 880°C for 48h. And then, add Cu<sub>0.03</sub> and MWCNT 0.75 wt% to do ball milling for 12h with a rotation rate 300rpm. And then, the compound was sealed vacuum quartz tube and melted at 880°C for 24h. The compound cracked into microscale by ball milling for 6h with a rotation rate 500rpm, and hybrid polyimide (PI) together.

Seebeck coefficient and electric conductivity by ZEM-3 (ULVAC-RIKO, Japan). Phase structure was analyzed by xray diffraction (XRD, Bruker D2, Germany). The morphology of particles and fractographs were observed by field emission scanning electron microscopy (FE-SEM, JSM-6700F, Japan). The thermal conductivity was measured by Laser Flash (ULVAC-TC900).

## 3. Results and Discussion

The crystal structure of n-type and p-type are identified by x-ray diffraction (XRD) to present the varying phase structures, as shown in Fig. 1 and Fig. 2. The diffraction peaks of the as-prepared binary  $Bi_2Se_{0.3}Te_{2.7}$  and  $Bi_{0.5}Sb_{1.5}Te_3$  exhibit

rhombohedral geometry (Space group  $R3\overline{m}$ ), which are the same as PDF#50-0954 and PDF#49-1713, respectively.



Fig. 1 XRD patterns of n-type  $Bi_2Se_{0.3}Te_{2.7}$  and Cu + MWCNT doping  $Bi_2Se_{0.3}Te_{2.7}$ .



Fig. 2 XRD patterns of p-type  $Bi_{0.5}Sb_{1.5}Te_3$  and Cu + MWCNT doping  $Bi_{0.5}Sb_{1.5}Te_3$ .



Fig. 3 (a) SEM image of n-type Cu + MWCNT doping Bi<sub>2</sub>Se<sub>0.3</sub>Te<sub>2.7</sub>,(b) SEM image of p-type Cu + MWCNT doping Bi<sub>2</sub>Se<sub>0.3</sub>Te<sub>2.7</sub>.

The Scanning Electron Microscope (SEM) images (Fig. 3) reveal the morphology of n-type and p-type materials, and it also shows there are some copper percolating on CNT. The

figure of merit ZT was already enhanced by copper percolation on CNT doping. (Fig.4) In the flat surface measurement, Fig. 5 shows the power output of FTEM has the best value around 210  $\mu$ W/cm<sup>2</sup>. In the curved surface measurement, Fig. 5 shows the power output of FTEM has the best value around 110  $\mu$ W/cm<sup>2</sup>.



Fig. 4 Temperature-dependent (a) S, (b)  $\sigma$ , (c)  $\kappa$ , (d) figure of merit ZT of N-type and P-type, respectively.



Fig. 5 FTEM output voltage and output power on flat surface and curve surface at different temperature difference.

#### 3. Conclusions

In conclusion, we have shown the FTEM via screen printing. The obtained FTEM has improved the faults of TTEM and is ideal for developing wearable electronic devices. And, the figure of merit ZT of Bi<sub>2</sub>Se<sub>0.3</sub>Te<sub>2.7</sub> and Bi<sub>0.5</sub>Sb<sub>1.5</sub>Te<sub>3</sub> materials are improved by copper percolation on CNT doping.

## References

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