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Screen-printed radial structure micro radioisotope thermoelectric generator

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HIGHLIGHTS

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GRAPHICAL ABSTRACT

- The prototype of the micro radioisotope thermoelectric generator was fabricated by screen printing.
- The matching scheme of the thermoelectric leg material is compared.
- Three types of radioactive isotopes were considered for experiments.
- 5 pairs of thermoelectric legs has an $V_{\rm oc}$ of 68.41 mV, $I_{\rm sc}$ of 328.96 μA and an P_{max} of 5.81 μ W.

A new solution for micro-isotope power supply is proposed. The prototype of the radial structure micro RTG was fabricated by the screen printing. Curing temperature was designed for optimal material properties. The prototype's electrical performance was tested and evaluated. This type of energy harvester may provide a new idea for energy development in the future space exploration missions. It can be used not only in micro RTG but also in lightweight applications.

ABSTRACT

The micro radioisotope thermoelectric generator can be invoked as a long-life power supply in low-power device applications. Improving the current, voltage and power of power sensors by enhancing the properties of thermoelectric material composites matters in low-power device applications. The micro radioisotope thermoelectric generator driven by the temperature difference between radial thermoelectric legs printed on polyimide substrate and the loaded central heat source is reported in this study. The electrical conductivity of n-type $Bi_2Te_2.5Se_{0.3}$, p-type $Bi_0.5Sb_1.5Te_3$, and p-type Sb_2Te_3 radial thermoelectric legs are 24.57–165.8 S·cm⁻¹, with Seebeck coefficients of -176.6 , 223.3 and 139.7 µV·K⁻¹ respectively. Thermoelectric legs are prepared by screen printing with a paste consisting of epoxy resin and BiTe-based powders. The generator has five couples of radial thermoelectric legs, and their material properties are optimized through selecting the preliminary curing temperature. The electrical conductivity of n-type $Bi_2Te_2.7Se_{0.3}$, p-type $Bi_{0.5}Sb_{1.5}Te_3$, and p-type Sb_2Te_3 thermoelectric legs are 24.57–165.8 S·cm⁻¹, with Seebeck coefficients of -176.6, 223.3 and 139.7 µV·K⁻¹ respectively. When loaded with 1.5 W isotope heat sources, the prototype generator would generate an opencircuit voltage of 68.41 mV, a short-circuit current of 329.0 µA, and an output power of 5.81 µW at 39.20 mV. Stacking and series-parallel can harvest considerable energy.

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1. Introduction

The radioisotope thermoelectric generator (RTG) converting radioisotope heat into electric power by thermoelectric (TE) material without moving parts has numerous advantages, such as high reliability, long lifetime, and minimal environmental impact [\[1](#page--1-0)–4]. The increasing number of small spacecraft and studies on potential scientific applications indicates the need for RTG application at low power levels. Micro RTGs (MRTGs) mainly lend themselves to small, long-life power devices. Moreover, MRTG can provide stable long-term power output for low-power sensing devices when carrying out deep space missions [\[5\].](#page--1-1) The application range of miniature scientific instruments, for example, the small long-life meteorological/seismological stations distributed across planetary surfaces, subsurface probes, deep space microspacecrafts and sub-satellites, is very likely to extend due to such power supply [\[4\].](#page--1-2) However, the minor temperature difference, low efficiency and insufficient reliability of the miniaturized RTG currently limit its application in space missions. As regard to the output performance problem of RTG, a novel micro RTG is designed to improve the above shortcomings in this paper.

TE modules in RTG can convert the energy between heat energy and electric energy directly, diversely applied in harvesting and sensing energy at the same time $[6,7]$. The optimization of structure and material is important to the properties of thermoelectric devices [\[8\]](#page--1-4). Whalen et al. have used 11 pairs of 215 μ m-thick Bi₂Te₃ in the design of a wheel spoke thermocouple, and optimized the thermopile through cutting the thermoelectric material by four equal parts. [\[9\]](#page--1-5). Menon et al. have stacked 15p–n radial TE couples in series, and further obtained the voltage and power increasing in linear along with TE [\[10,11\].](#page--1-6) The application of block and thick-film devices with this structure has been reported in previous work. The number of elements and power in a given area needs to be balanced [\[12,13\].](#page--1-7) Semiconductor alloy materials based on $Bi₂Te₃$ have the brittleness characteristics [\[14\]](#page--1-8). In particular, the devices of long-life micro radioisotope of TEGs based on printed thick film BiTe requires a high degree of mechanical stability and flexibility. Physical cutting methods can no longer meet the thin film requirements for fabricating devices with radial structures. Thus methods of molding the thick film TE material need to be changed. As for the 2D additive manufacturing method, the screen printing has the advantages of low cost, rapid prototyping and mass manufacturing. Son and Cho et al. have applied the screen printing method to produce high-performance thermoelectric generator by using all-inorganic and hybrid viscoelastic inks in recent years [15–[17\].](#page--1-9) A large number of thermocouples can be manufactured rapidly by screen printing, able to provide high electric power, which is indeed a developed and appropriate program [\[18,19\].](#page--1-10) RTG's reliability is similar to the requirements on flexibility and output performance of a wearable thermoelectric device [\[20](#page--1-11)–22]. Polymer thermoelectric composite

materials are very attractive owing to its low cost, flexibility and high power density [\[23\]](#page--1-12). Madan et al. have reported a flexible thermoelectric generator of polymer thermoelectric composite, used in flexible and high output practical wireless sensor networks [\[24\]](#page--1-13). Gima et al. have reported 50-couple screen printed $Bi₂Te₃/Bi_{0.5}Sb_{1.5}Te₃$ -epoxy annular thermoelectric generators, used in ultra-low-power sensors, with the average power of 0.068 nW at a 20 K temperature gradient, 26nA and 2.6 mV [\[25\]](#page--1-14). In this study, we have investigated the TE properties of MRTG composite materials. In addition, a new manufacturing process is optimized, and the properties of the composite semiconductor material are explored. Furthermore, a prototype with five pairs of TE legs, diameter of 1.5 cm, and an area of 1.77 cm^2 is prepared. It is tested by loading a radioisotope heat source. The output performance of the MRTG is further evaluated.

Moreover, it will improve the energy conversion efficiency by fabricating planar array and space stack thermoelectric devices. Thus, the thermoelectric device technology deserves to be promoted because of the simple heat treatment process, equipment manufacturing process, heat treatment conditions, and the whole process posing no harm to the environment. The use of new thermoelectric device manufacturing technology is not limited to micro RTG. Various lightweight devices, such as low-cost flexible wearable generators, solar thermal generators and pipe powering wireless sensors are also welcomed [\[20,26\].](#page--1-11)

2. Materials and methods

2.1. Bi₂Te₃-epoxy TE paste synthesis

TE bulk ingots are prepared as TE powder (i.e., p-type $Bi_{0.5}Sb_{1.5}Te_3$, $Sb₂Te₃$ and n-type $Bi₂Te_{2.7}Se_{0.3}$) through airflow milling. The powder needs to be sieved through a 325 mesh screen before use. The polymer binder is an epoxy system formulated by the polypropylene glycol diglycidyl ether epoxy resin (Sigma-Aldrich) and the methylhexahydrophthalic anhydride (Sigma-Aldrich). Among them, the equivalent weight ratio of epoxy and hardener is 1:0.85. Furthermore, use the 1- Cyanoethyl-2-ethyl-4-methylimidazole (0.5 wt%; Shikoku Chemicals) as the catalyst in the system.

The TE powder occupies 45–50 vol% of and paste (Fig. S1). Add the butyl acetate (Sigma-Aldrich) to the resin blend to reduce the viscosity of the ink as a nonreactive diluent. Furthermore, the low percentages of organic solvents are used to extend the shelf life of the epoxy system so as to adjust the viscosity for printing, and the epoxy system is selected in this work owing to its low viscosity and extended pot life. Use the planetary mixer to mix the powder and solvent cement uniformly. Mix the turbid liquid system at 1000 rpm for 3 min, being held for 1 min, and mix it again at 1800 rpm for 3 min.

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