Supporting Information

# Optimizing output performance and parasitic depletion of Bi<sub>2</sub>Te<sub>3</sub>-based thermoelectric generators by using a high-density approach

Yu Tian,<sup>ab</sup> Guang-Kun Ren,<sup>a\*</sup> Zhifang Zhou,<sup>c</sup> Zhijie Wei,<sup>a</sup> Wen Fang,<sup>a</sup> Jiangfeng Song,<sup>a</sup> Yan Shi,<sup>a</sup> Xiaohong Chen,<sup>b\*</sup> Yuan-Hua Lin<sup>c</sup>

<sup>a</sup> Institute of Materials, China Academy of Engineering Physics, Jiangyou, Sichuan, 621908, P. R. China

<sup>b</sup> School of Science and Research Center for Advanced Computation, Xihua University, Chengdu, Sichuan, 610039,

P. R. China

<sup>c</sup> State Key Laboratory of New Ceramics and Fine Processing, School of Materials Science and Engineering, Tsinghua University, Beijing, 100084, P. R. China

## **Phase composition**

The X-ray diffraction patterns of p-type (Bi-Sb-Te) and n-type (Bi-Te-Se) materials are consistent with the R-3m space group with hexagonal layered structures, and no impurities are observed, as marked in Fig. S1.



Fig. S1 Phase characterization diagram of commercial  $Bi_2Te_3$ -based thermoelectric materials. The X-ray diffraction patterns of (a) p-type and (b) n-type materials.

	P-type			N-type		
Element	Bi	Sb	Te	Bi	Te	Se
at.%	8.92	32.77	58.31	40.35	55.92	3.73
wt.%	14.02	30.01	55.96	53.16	44.98	1.86

Table S1 The compositions of commercial p- and n-type Bi<sub>2</sub>Te<sub>3</sub>-based materials.

### The average Seebeck coefficient in Finite element module

The average Seebeck coefficient values derived from  $\Delta U$ - $\Delta T$  curves can be used to verify the commercial materials, and as shown in Fig. S2.



Fig. S2 Packing fraction dependence of average Seebeck coefficient values of 200-Tcs (f = 20%), 50-Tcs (f = 5%) and 50-Tcs (f = 34%) TEGs at different  $\Delta T$ .

#### Experimental contact resistivity used for V-TDE

To shed more light on the parasitic depletion, especially for contact depletion, systematic characterizations of  $r_c$  have been done based on a four-probe method, and the calculated  $\rho_c$  has been introduced into the V-TDE, Aa shown in Fig. S3(a)-(b).



Fig. S3 (a) The electrical contact resistance ( $r_c$ ) of typical reflow soldering sample, and (b) the simulation and experimental value of TEGs (*f*: 5%, 20% and 34%).

## 338-Tcs TEG with destroyed linear relationship between $P_{\text{max}}$ and f

For low temperature environment, the TEG possesses outstanding performance by remaining the maximum effective  $\Delta T$ , however, the ability would be weakened at high temperatures, resulting in higher average temperature of each leg, larger internal resistance, and lower open-circuit voltage than liner values. And the average Seebeck coefficient values are within the range made by p- and n-type materials at  $\Delta T$  of 13 K and 43K, yet they are below those at  $\Delta T$  of 73 K, as shown in Fig. S4 (a) and (b).



Fig. S4 (a) Packing fraction dependence of r,  $\Delta U$  and average Seebeck coefficient values of 50-Tcs (f = 5%), 200-Tcs (f = 20%) and 338-Tcs (f = 34%) TEGs at different  $\Delta T$ . (b) The leg height (l) dependence of  $P_{\text{max}}$ ,  $\Delta U$  and r of 338-Tcs TEG between 293K and 366K.