## **Supplementary Information**

## Skutterudite with Graphene-modified Grain-boundary Complexion Enhances *zT* Enabling High-efficiency Thermoelectric Device

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Fig. S1 Powder X-ray diffraction patterns of the  $Ce_{0.85}Fe_3CoSb_{12}/y$  vol% rGO (y = 0, 0.56, 1.4, 2.8)



Fig. S2 Raman Spectra of GO and 850K-SPSed rGO



Fig. S3 The EDS image of bulk  $Ce_{0.85}Fe_3CoSb_{12}/y \text{ vol}\% \text{ rGO} (y = 2.8)$ .



Fig. S4 (a-d) The surface SEM image of bulk  $Ce_{0.85}Fe_3CoSb_{12}/y$  vol% rGO ( y = 0, 0.56, 1.4, 2.8)



Fig. S5 The cross section HRTEM image of bulk  $Ce_{0.85}Fe_3CoSb_{12}/2.8\ vol\%\ rGO$ 



Fig. S6 EDS of the nano precipitate in Fig. 2d



Fig. S7 HAADF-STEM image of the p-type  $Ce_{0.85}Fe_3CoSb_{12}/0.56$  vol% rGO wrapping sample

Table S1. Comparison of the carrier concentration (*p*), mobility ( $\mu$ ), electrical conductivity ( $\sigma$ ), hall coefficient (*R*<sub>h</sub>), Seebeck coefficient (*S*) and power factor (*PF*) of Ce<sub>0.85</sub>Fe<sub>3</sub>CoSb<sub>12</sub>/y vol % rGO ( y = 0, 0.56, 1.4, 2.8) at 300 K.

р	μ	σ	R <sub>h</sub>	S	PF
$(10^{20}\mathrm{cm}^{-3})$	$(cm^2V^{-1}s^{-1})$	$(10^{5} \text{Sm}^{-1})$	$(10^{-2} \text{cm}^3 \text{C}^{-1})$	(µVK <sup>-1</sup> )	(µWcm <sup>-1</sup> K <sup>-2</sup> )
19.6	3.6	1.11	0.32	109.9	13.5
20.7	3.3	1.10	0.30	112.4	14.0
22.1	3.1	1.09	0.28	116.8	15.0
27.3	2.3	1.00	0.22	112.5	12.7
	<i>p</i> (10 <sup>20</sup> cm <sup>-3</sup> ) 19.6 20.7 22.1 27.3	$\begin{array}{c c} p & \mu \\ (10^{20}  {\rm cm}^{-3}) & ({\rm cm}^2 {\rm V}^{-1} {\rm s}^{-1}) \\ \hline 19.6 & 3.6 \\ 20.7 & 3.3 \\ 22.1 & 3.1 \\ 27.3 & 2.3 \end{array}$	p $\mu$ $\sigma$ (10 <sup>20</sup> cm <sup>-3</sup> )(cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> )(10 <sup>5</sup> Sm <sup>-1</sup> )19.63.61.1120.73.31.1022.13.11.0927.32.31.00	p $\mu$ $\sigma$ $R_h$ (10 <sup>20</sup> cm <sup>-3</sup> )(cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> )(10 <sup>5</sup> Sm <sup>-1</sup> )(10 <sup>-2</sup> cm <sup>3</sup> C <sup>-1</sup> )19.63.61.110.3220.73.31.100.3022.13.11.090.2827.32.31.000.22	$p$ $\mu$ $\sigma$ $R_h$ $S$ $(10^{20}  \mathrm{cm}^{-3})$ $(\mathrm{cm}^2 \mathrm{V}^{-1} \mathrm{s}^{-1})$ $(10^5 \mathrm{Sm}^{-1})$ $(10^{-2} \mathrm{cm}^3 \mathrm{C}^{-1})$ $(\mu \mathrm{V} \mathrm{K}^{-1})$ 19.63.61.110.32109.920.73.31.100.30112.422.13.11.090.28116.827.32.31.000.22112.5