Supplementary Information

Thermoelectric Properties of $CuInTe_2$/graphene Composites

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Experimental details

Fabrication: Graphene was prepared by ball milling method based on the expanded graphite (EG) and MgO, which has previously been reported.[1] 0.2 g EG and 1 g MgO were mixed, followed by planetary ball milling at 500 rpm for 12 h. The obtained products were dispersed in dilute hydrochloric acid to eliminate MgO. The final grapheme was obtained after filter and drying. Stoichiometric amounts of elemental copper (Cu, 99.999%, SinoReag), elemental indium (In, 99.999%, SinoReag) and elemental tellurium (Te, 99.999%, SinoReag) were mixed and sealed in evacuated quartz tubes to synthesize $CuInTe_2$. The tubes were heated to 1073 K for 12 h, and then furnace cooled to room temperature. 4 g $CuInTe_2$ and 0.1 g graphene was mixed and subsequently ball milled to obtain smaller and homogeneous composites. The obtained powders were spark plasma sintered at 500 °C for 10 min

under a pressure of 60 MPa in flowing Ar gas, yielding densified bulk samples. Sample densities were measured by the Archimedes method and the relative densities were above 97 %.

Instrumentation: The structure characters were confirmed by the powder XRD (Bruker D8 Focus) using Cu Kα radiation (λ = 1.5418 Å). Optical absorption spectra of their pulverized powders were measured at room temperature by a UV-visible-near IR spectrometer (HITACHI U-3010) equipped with an integrating sphere. Raman spectroscopy was collected in a Renishaw inVia Reflex Laser Raman with a cooled CCD detector. TEM and SEAD were conducted by a JEOL 2100F microscope, operating at 200 kV. Cross-sectional morphologies were observed with a scanning electron microscopy (SEM) JEOL-6510. For transport property measurements, bar samples about 1.5 × 2 × 10 mm^3 were used. Electrical conductivity and Seebeck coefficient were measured simultaneously by the commercial system (ZEM-3, ULVAC-RIKO). Thermal diffusivity coefficient λ was determined using a laser flash method in a flowing Ar atmosphere (Netzsch LFA 427). The thermal conductivity was calculated from $k = \rho\lambda C_p$, in which ρ is the density and C_p is the specific heat capacity. The Dulong-Petit value of C_p was used. The Hall coefficient was measured by the standard five-wire technique with silver-paint contacts at room temperature in a Physical Property Measurement System (PPMS-9T, Quantum Design Company).

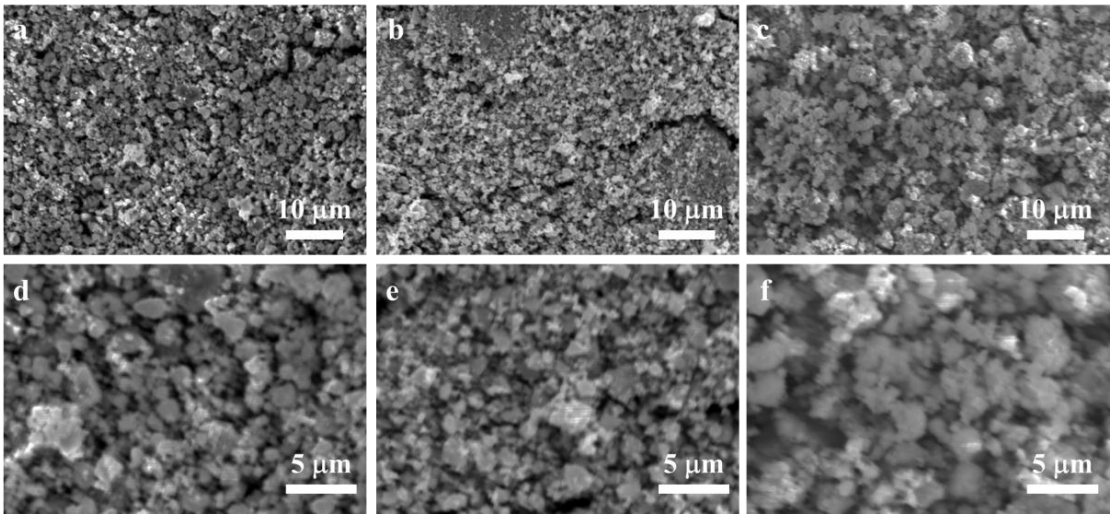

Fig. S1. Low- and high-magnified SEM images of (a,d) $CuInTe_2$, (b,e) $CuInTe_2$/graphene (80:1) and (c,f) $CuInTe_2$/graphene (40:1) powders after ball-milling before SPS process.

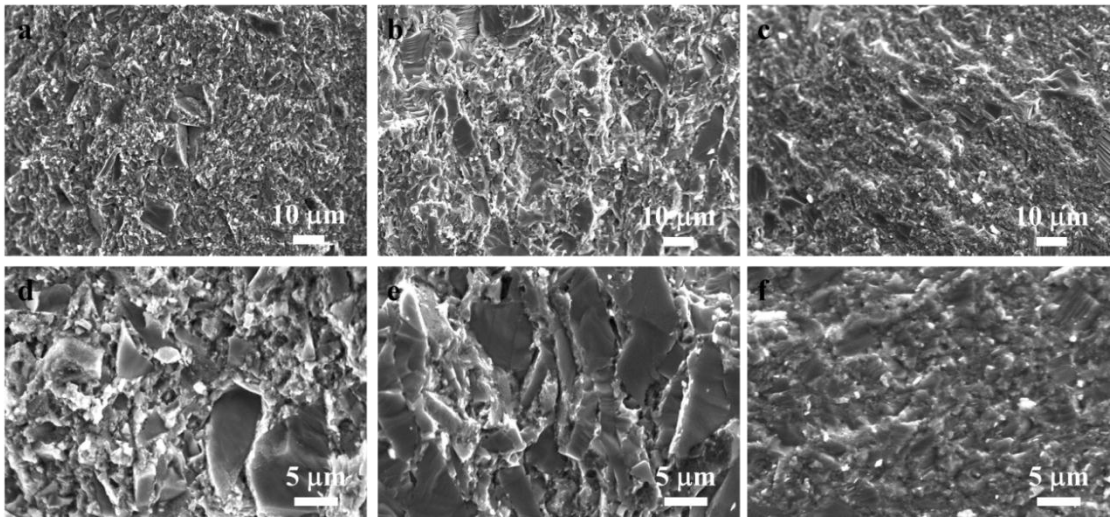

Fig. S2. Low-magnified cross-sectional SEM images of (a,d) $CuInTe_2$, (b,e) $CuInTe_2$/graphene (80:1) and (c,f) $CuInTe_2$/graphene (40:1).

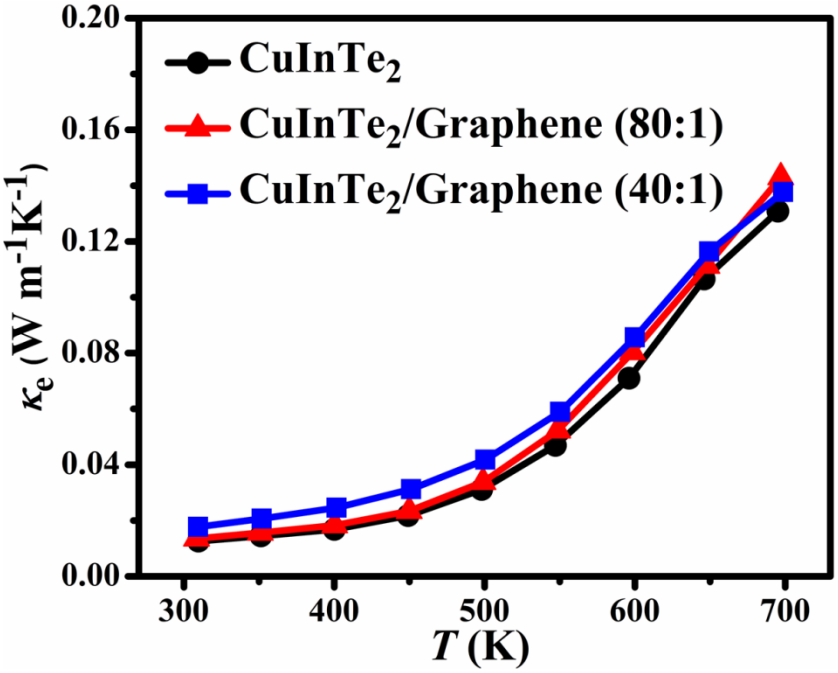

Fig. S3. Temperature dependence of electronic thermal conductivity (κ_e).

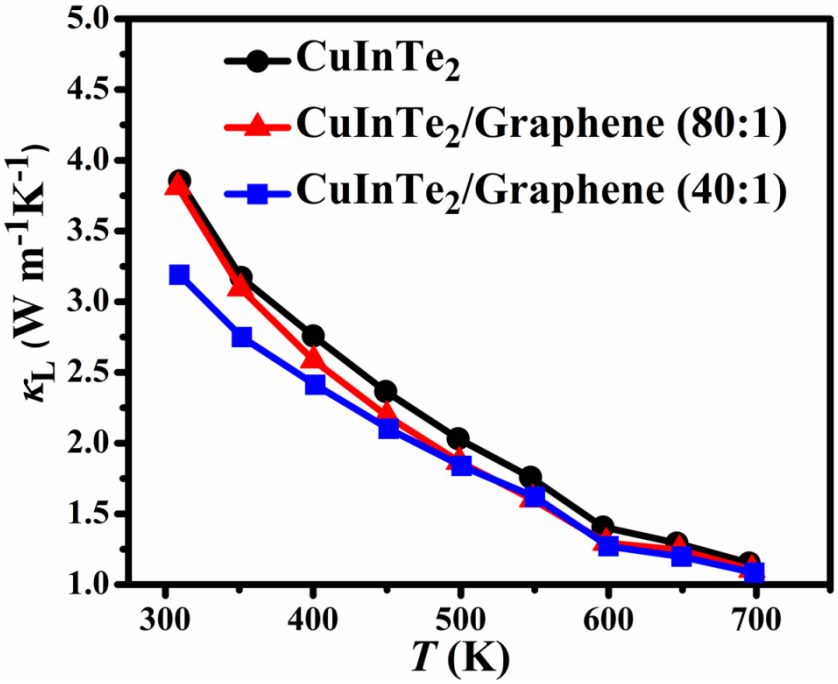

Fig. S4. Temperature dependence of phonon thermal conductivity (κ_e).

Table S1. The atomic positions and occupancies of $CuInTe_2$.

Element	***x***	***y***	***z***	**Occupancies**
Cu	0	0	0	1
In	0	0	0.5	1
Te	0.225	0.25	0.125	1

Table S2. Comparison of the electrical conductivity (σ), Seebeck coefficient (S), thermal conductivity (κ), electronic thermal conductivity (κ_e), phonon thermal conductivity (κ_L) and dimensionless figure of merit (ZT) at 700 K.

Sample	σ **(S·m^{-1})**	S **(μV·K^{-1})**	PF **(mW·m^{-1}K^{-2})**	κ **(W·m^{-1}K^{-1})**	κ_e **(W·m^{-1}K^{-1})**	κ_L **(W·m^{-1}K^{-1})**	ZT
$CuInTe_2$	12547	239	0.72	1.28	0.13	1.15	0.39
$CuInTe_2$/graphene (80:1)	13676	228	0.71	1.25	0.14	1.10	0.40
$CuInTe_2$/graphene (40:1)	13140	225	0.67	1.22	0.14	1.08	0.38

References

S1. T. Lin, J. Chen, H. Bi, D. Wan, F. Huang, X. Xie and M. Jiang, *Journal of Materials Chemistry A*, 2013.