Electronic Supplementary Material (ESI) for Energy & Environmental Science.

Supporting Information

High-performance n-type Ta_4SiTe_4/polyvinylidene fluoride (PVDF)/graphdiyne organic-inorganic flexible thermoelectric composites

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Details about the X_d calculation

The thickness of the fully depleted layer (X_d) in the Ta_4SiTe_4 and GDY interface can be calculated by the depletion approximation[1]

$$X_d = \left(\frac{2\varepsilon\Delta V}{qN_d 3}\right)^{1/2},$$

where ε, ΔV, q, and N_d are the dielectric constant of Ta_4SiTe_4, the built-in potential in the interfacial area, the elementary charge, and the carrier concentration of Ta_4SiTe_4. ε is assumed to be $10\varepsilon_0$ for Ta_4SiTe_4, where ε_0 is permittivity of vacuum. ΔV equals to the difference in work functions between Ta_4SiTe_4 and GDY.

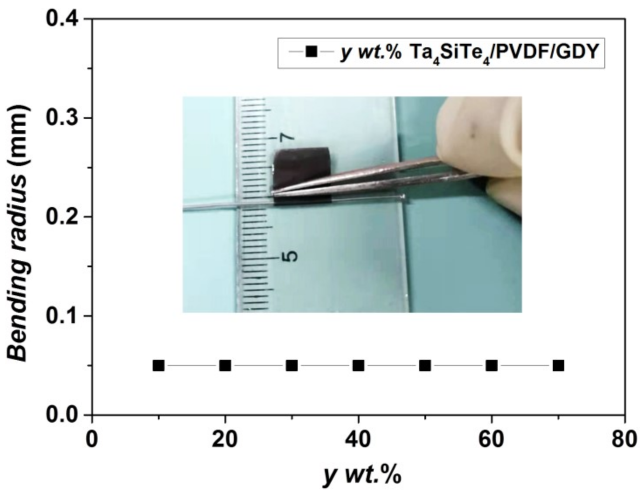

Fig. S1 Bending radius for the *y wt.*%PVDF/Ta_4SiTe_4/5.8 *wt.*%GDY composite films with different Ta_4SiTe_4 mass ratios. The inset shows the optical image of rolling the composite film on a cylindrical object with $r = 0.05$ mm. This method was proposed by Liang et al. [2]

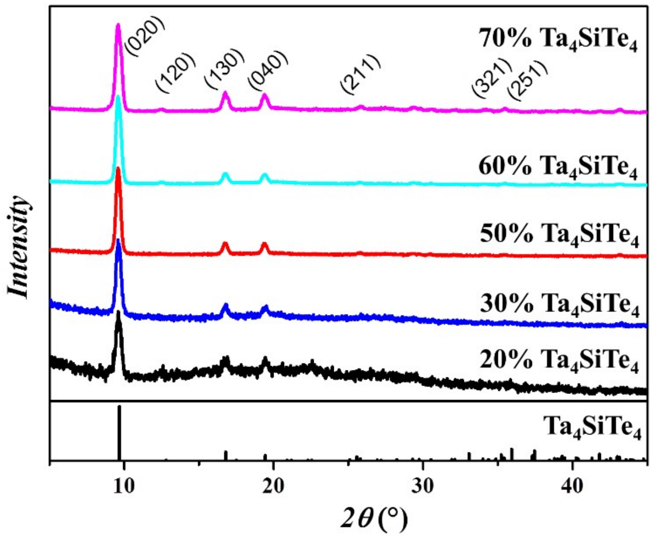

Fig. S2 X-ray diffraction patterns of the *y wt.*%PVDF/Ta_4SiTe_4/5.8 *wt.*%GDY composite films with different Ta_4SiTe_4 mass ratio.

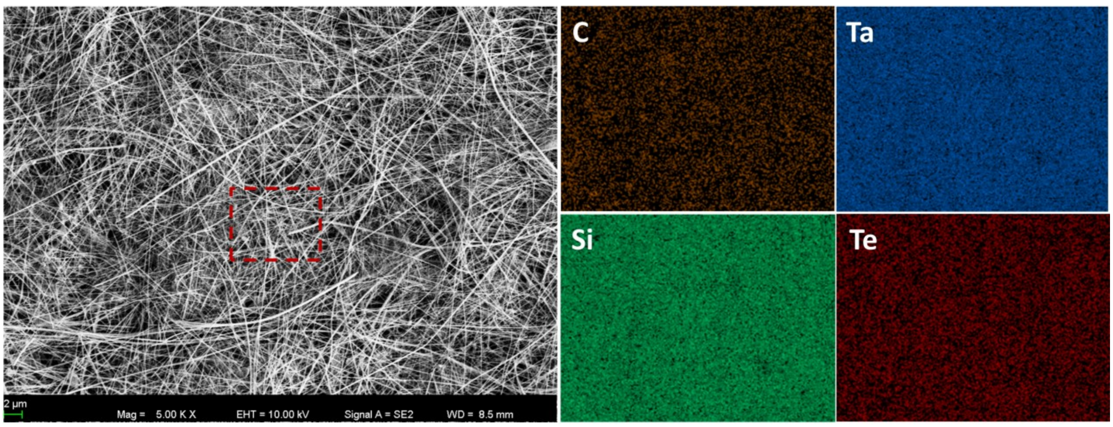

Fig. S3 SEM image of 50 *wt.*%Ta_4SiTe_4/PVDF/5.8 *wt.*%GDY composite film and the elemental mapping of Ta, Si, Te, and C.

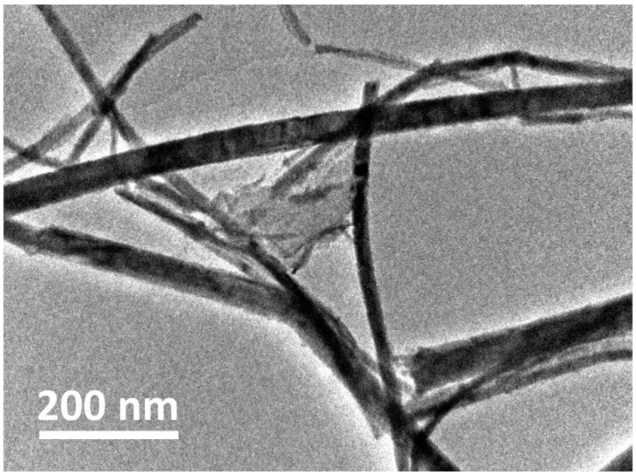

Fig. S4 Low-resolution TEM images of Ta_4SiTe_4/GDY composites.

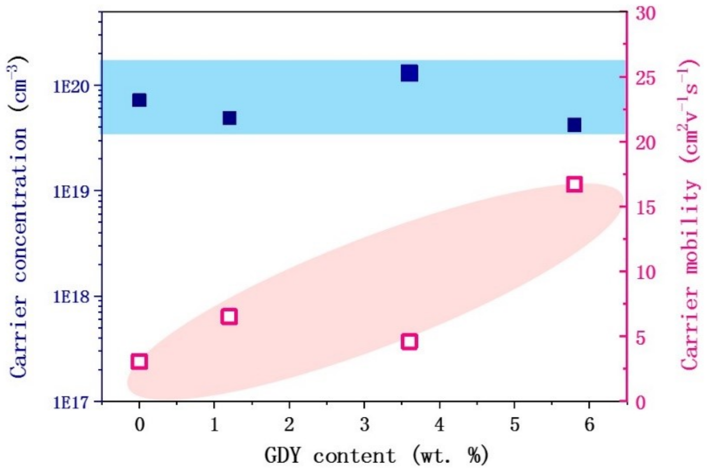

Fig. S5 Carrier concentration and carrier mobility of 50 *wt.*%PVDF/Ta_4SiTe_4/x *wt.*%GDY (x = 0, 1.2, 3.6, and 5.8) composite films.

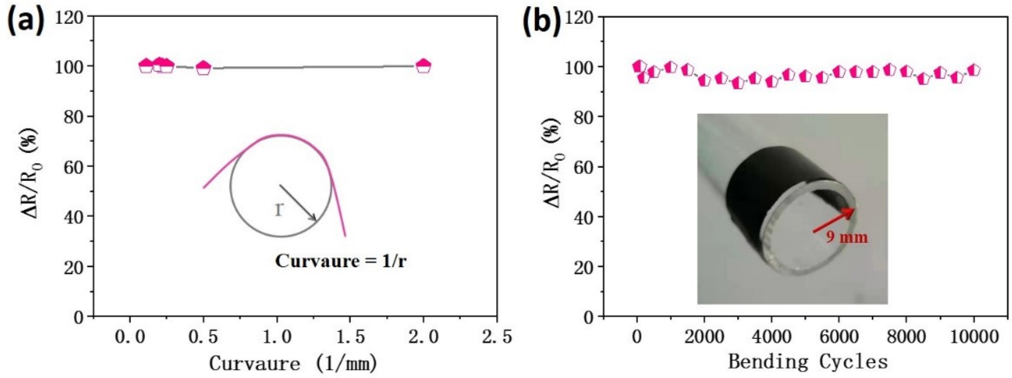

Fig. S6 (a) Relative resistance variation ($\Delta R/R_0$) as a function of various curvatures for 50 *wt.*% Ta_4SiTe_4/PVDF/5.8 *wt.*% GDY composite film. b) $\Delta R/R_0$ as a function of bending cycles over a curvature of 0.11 mm^{-1} for 50 *wt.*% Ta_4SiTe_4/PVDF/5.8 *wt.*% GDY composite film. The resistance is almost unchanged after 10000 bending cycles.

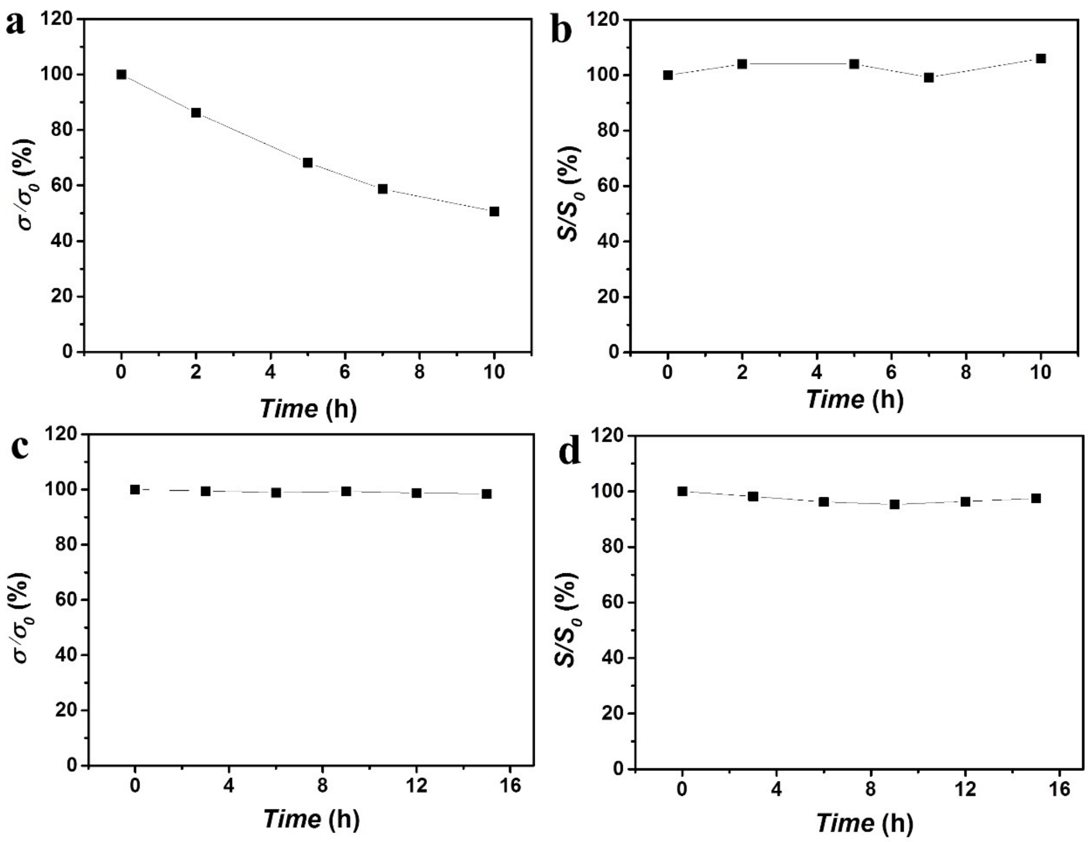

Fig. S7 Electrical conductivity and Seebeck coefficient variations of 50 *wt.*%PVDF/Ta_4SiTe_4/5.8 *wt.*%GDY composite film during the stability test (a-b) in air and (c-d) Argon atmosphere.

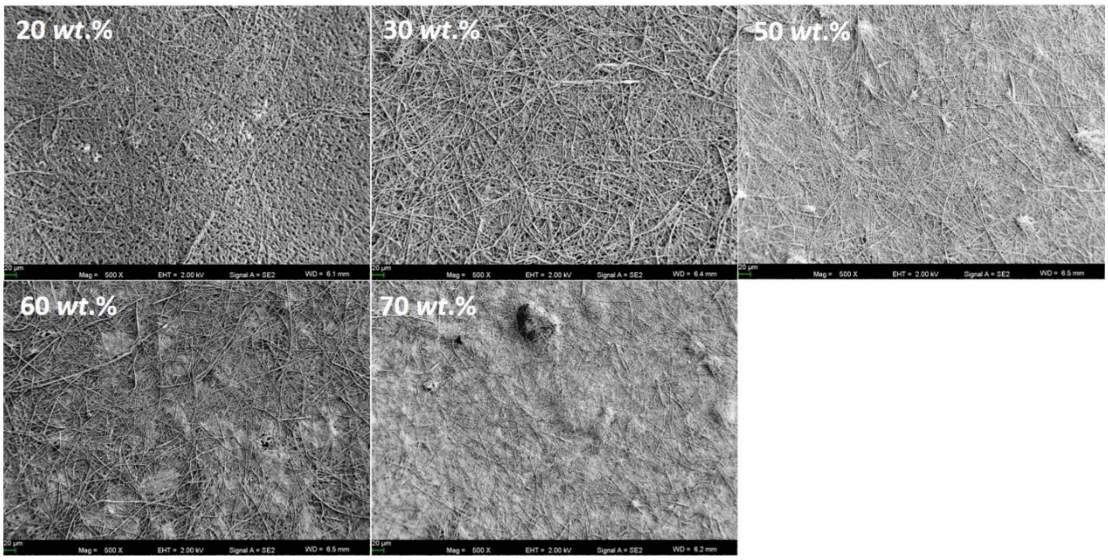

Fig. S8 SEM images of *y wt.*%Ta_4SiTe_4/PVDF/5.8 *wt.*%GDY (*y* = 20, 30, 50, 60, and 70) composite films.

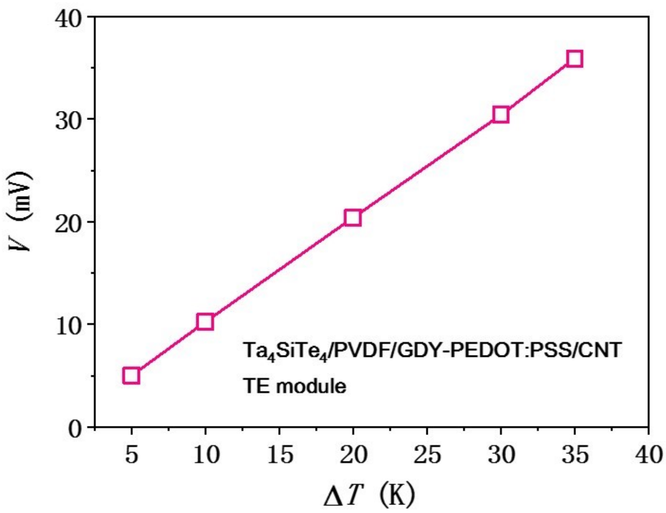

Fig. S9 Open voltage (*V*) as a function of temperature gradient (ΔT) for the prototype Ta_4SiTe_4/PVDF/GDY-PEDOT:PSS/CNT TE module.

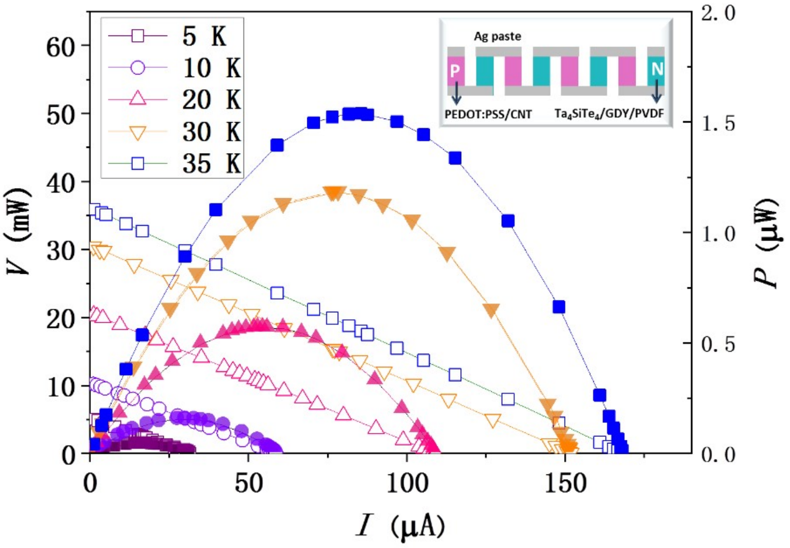

Fig. S10 Output voltage (*V*) and power (*P*) as a function of current (*I*) for prototype Ta_4SiTe_4/PVDF/GDY-PEDOT:PSS/CNT TE module under different operating temperatures. The inset shows the sketch map of the prototype module.

References

[1] D. A. Neamen, Semiconductor Physics and Devices, *Basic Principles*, New York: The McGraw-Hill Companies, Inc.

[2] L. R. Liang, C. Y. Gao, G. M. Chen, C-Y Guo, *J. Mater. Chem. C*, 2016, **4**, 526.