Conformal organic-inorganic semiconductor composites for flexible

thermoelectrics

(Supplementary Information)

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Supplementary Figure 1| Calculated band structure of Ta₄SiTe₄ with spin-orbit coupling effect. The Fermi level is set to 0 eV.



Supplementary Figure 2 Optical image of the as-prepared Ta_4SiTe_4 whiskers picked from the inner wall of quartz tube.



Supplementary Figure 3 Secondary electron image of Ta_4SiTe_4 whiskers and corresponded EDS mappings of elemental Si, Te and Ta. The image of elemental Mo is not shown here because of its very low content.



Supplementary Figure 4 Optical images of a bundle of Ta_4SiTe_4 whiskers taken before and during the bending process.



Supplementary Figure 5| Secondary electron images of PVDF/Ta₄SiTe₄ (45 *wt*%) and PVDF/Ta₄SiTe₄ (55 *wt*%) composite films.



Supplementary Figure 6| Fourier transform infrared reflection (FTIR) spectra of PVDF and PVDF/Ta₄SiTe₄ composite.



Supplementary Figure 7 a) Relative resistance as a function of various curvatures for PVDF/Ta₄SiTe₄ composite film. **b)** Relative resistance as a function of bending cycles over a curvature of 0.22 mm⁻¹ for PVDF/Ta₄SiTe₄ composite film. The resistance is practically unchanged after 5000 bending cycles.



Supplementary Figure 8| **a)** Electric conductivity and **b)** Seebeck coefficient of PVDF/Ta₄SiTe₄ composite film at 300 K during the stability test. The measurement was performed in Argon atmosphere by ZEM-5 (ULVAC, Japan).

 $\begin{array}{c|ccccc} & & & & & T_{hot}(^{\circ}\mathrm{C}) & & & T_{cold}(^{\circ}\mathrm{C}) \\ \hline & & 9.2 & & 46.8 & & 37.6 \\ & & 19.9 & & 69.2 & & 49.3 \\ & & 30.1 & & 90.2 & & 60.1 \\ & & 35.5 & & 103.2 & & 67.7 \end{array}$

Table S1 Hot side temperature (T_{hot}) and cold side temperature (T_{cold}) on the TE module under different temperature differences (Δ T).