## **Supplementary Information**

# A solution-processed TiS<sub>2</sub>/organic hybrid superlattice film towards flexible thermoelectric devices

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## **XRD** analysis

XRD measurement has been performed on the mixed  $TiS_2$ /hexylamine (HA) powder after grinding. The experimental result is presented in Fig. s1, which shows that the resultant powder consists of intercalated  $TiS_2$ [HA] phase with larger d-spacing as well as a small amount of unintercalated  $TiS_2$  phase. Fig. s2 compares the XRD pattern of pristine  $TiS_2$  crystal and asdeposited film, which clearly shows the peak shifting towards low angel direction after organic intercalation.

#### Weight loss

We have annealed the film sample at 130 °C in vacuum and measured the weight change as a function of annealing time. The result is presented in Fig. s3, which demonstrates the remarkable weight loss during annealing.

## **Evolved** gas analysis

In order to further identify the organic substances and their stability in the film, we have performed the evolved gas analysis by gas chromatography-mass spectrometer (GC-MS). The 6h-annealed sample was scratched from substrate and loaded into a pyrolyzer system (Frontier Lab PY-2020iD) coupled with a GC-MS. The sample was heated up to 600 °C in nitrogen with a heating rate of 15°C/min. The extracted ion chromatograms of m/z 30 and m/z 59 signals are presented in Fig. s4, in which the former can be assigned to hexylamine (HA) and the latter is N-methylformamide (NMF). It clearly shows that most of NMF molecules were removed from the film at temperatures below 300°C, while HA was mainly extracted from the film at temperatures above 300°C, although NMF has a higher boiling point than HA.

## I-V curves of flexible thermoelectric module

The I-V curves of our flexible device were recorded by a programmable Keithley 4200-SCS analyzer at different temperature gradient in the range of 10-70K and the result is presented in Fig. s5.



Fig. s1 XRD pattern of mixed TiS2 and hexylamine (HA) powder after grinding



Fig. s2 XRD pattern of pristine  $TiS_2$  crystal (upper panel) and  $TiS_2$ /organics hybrid film (lower panel).



Fig. s3 The weight loss change of as-deposited film as a function of annealing time.



Fig.s4 The extracted ion chromatogram of (a) m/z 30 and (b) m/z 59 signals as a function of temperature.



Fig. s5 The measured I-V curves of our flexible thermoelectric module at different temperature gradient.