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Supporting Information

MXene Nanosheet/Organic Superlattice for Flexible Thermoelectrics

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S1. Process reproducibility and reliability

In this work, 3 batches of MX, MX SL, SL-300, SL-400 and SL-500 films were prepared. For each composition in each batch, 3 samples were cut out from different regions. As shown in Figure S1, for a specific composition, the fluctuation of σ and *S* was within 5% and 10%, respectively, in the same batch; while they increased to be within 10% and 15%, respectively, from batch to batch, implying good reproducibility and reliability of the process.



Figure S1. Fluctuation of the electrical conductivity, σ and Seebeck coefficient, *S* of SL-400 film (a) In different regions of the same batch, and (b) From batch to batch.

S2. Setup for Seebeck coefficient measurement

The Seebeck coefficient was measured in ambient conditions based on a homemade apparatus. As shown in Figure S2a, the ends of the sample are placed on two reversibly installed Peltier modules and the temperature difference between the two ends of the sample is achieved by applying different current to the Peltier modules. A pair of thermocouples are pointed at the sample to collect the temperature difference (Δ T) and voltage difference (Δ V), and the distance between the probes was about 1.5 cm. Finally, a linear relationship was fitted based on the recorded Δ V and Δ T, and the slope of this linearity was noted as the Seebeck coefficient (Figure S2b).



Figure S2. (a) Schematic of the experimental setup for the Seebeck coefficient measurement; (b) Fitting of $\Delta V vs. \Delta T$.

S3. Setup for electrical conductivity measurement

As shown in Figure S3a, the electrical conductivity was measured on a commercial Hall measurement system (CH-Magnetoelectricity Technology, CH-100) in ambient conditions based on the Van der Pauw method. As shown in Figure S3b, the samples were cut into squares with dimension of 1 cm \times 1 cm, the 4 corners of which were contacted with four gold probes with silver serving as the contacting material. The distance between adjacent probes was about 1 cm.



Figure S3. (a) Configuration for electrical conductivity measurement using the Van der Pauw method, (b) Photography of the testing configuration.

S4. Molecule alignment after intercalation



Figure S4. (a) Schematic microstructure of MXene/HA mixture, and (b) MXene/HA/NMF (MX SL)

film.

S5. Film morphology



Figure S5. Morphology of the peeled-off MX SL film (a) At flat state, and (b) Under bending.

S6. Output voltage of the TE module



Figure S6. Demonstration of the voltage output of the TE module when attaching one end to finger skin.



S7. Air stability of the MXene-based films

Figure S7. The decrease rate of electrical conductivity ($\sigma_0 - \sigma_s / \sigma_0$, σ_s for the exposed, σ_0 for the original) of MX, MX SL, MX SL-300, MX SL-400 and MX SL-500 films after being exposed in air (temperature = ~26 °C and relative humidity = ~55%) for 3 months.

S8. Reproducibility of the TE module

ΔT (K)	U (mV)			Power (nW)			Power density (W m ⁻²)		
	1st	2nd	3rd	1st	2nd	3rd	1st	2nd	3rd
10	0.53	0.41	0.49	2.5	2.3	2.8	0.016	0.014	0.017
30	1.33	1.19	1.25	20.3	16.1	23.5	0.127	0.100	0.146
50	2.31	2.35	2.26	58.6	63.4	55.5	0.366	0.396	0.346

Table S1. Performance of the TE modules based on SL-400 films of 3 batches

S9. Air stability of the TE module



Figure S8. The decrease rate of output voltage (U_0-U_s/U_0) and power (P_0-P_s/P_0) of TE module (where U_s , and P_s are output voltage and power of TE module for the exposed, U_0 , and P_0 for the original) after being exposed in air (temperature = ~26 °C and relative humidity = ~55%) for 3 months.