HF-free synthesis of high-entropy MXene-PVA composite film and its flexible nanogenerator

Fumei Yang[1], Kam Lin Chan[1], Zehan Wu[1], Fangqing Zhao[1], Man Chung Wong[1], Sin-Yi Pang[1] and Jianhua Hao[1*]

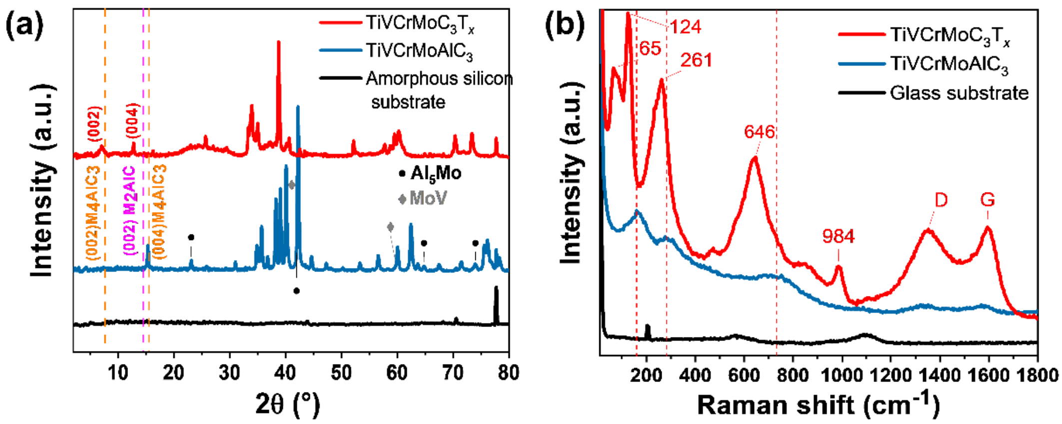

Figure S1 XRD patterns (a) and Raman spectra (b) of $TiVCrMoAlC_3$ MAX and etched high-entropy $TiVCrMoC_3T_x$ MXene nanosheets.

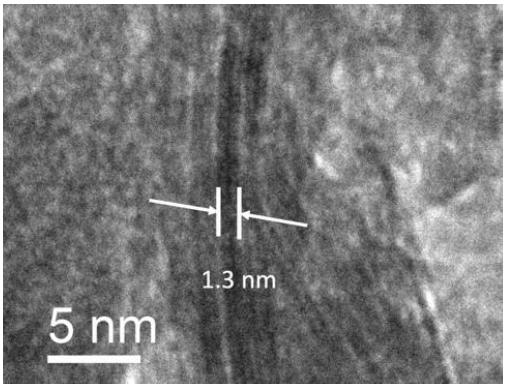

Figure S2 The TEM image of high-entropy MXene nanosheet. The TEM image confirms the predominantly multilayered nature of our MXene nanosheets, with 3 layers and a single layer thickness of ~1.3 nm.

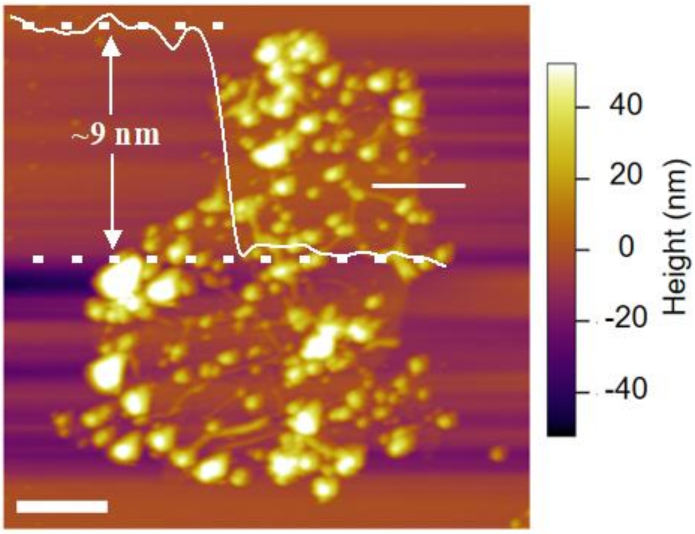

Figure S3 The AFM image and height profile of high-entropy MXene nanosheet. Scale bar: 1 μm.

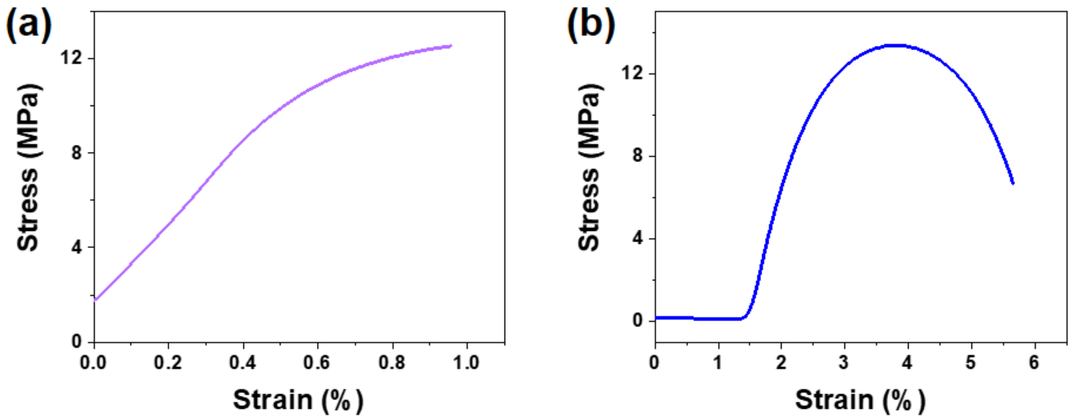

Figure S4 The stress-strain curve of MXene-PVA composite film (a) and pure PVA film (b).

The mechanical property was investigated via dynamic mechanical analyzer (Mettler Toledo DMA1). The films were clamped carefully onto the holder. The MXene-PVA composite film has a rectangular shape with dimensions of 19.28 mm in length, 10 mm in width and a thickness of 0.04 mm. As for pure PVA film, its length, width and thickness are 18 mm, 9 mm, and 0.02 mm, respectively. After the tensile strength measurement, the stress-strain curves were obtained, as shown in supplementary material, Fig. S4. From the stress-strain curves, the Young's modulus was determined by analyzing the linear region of the curve. The extracted Young's modulus of MXene-PVA composite film and pure PVA film is 2 GPa and 1 GPa, respectively.

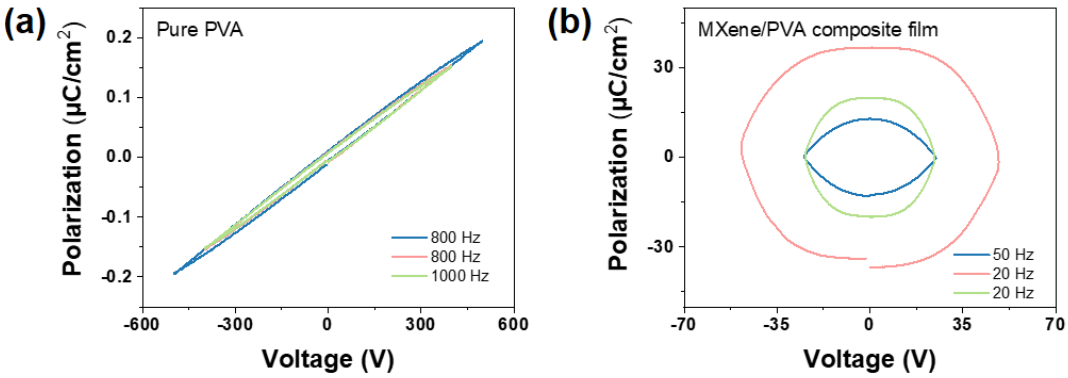

Figure S5 P-E loops of pure PVA (a) and MXene-PVA composite film (b).

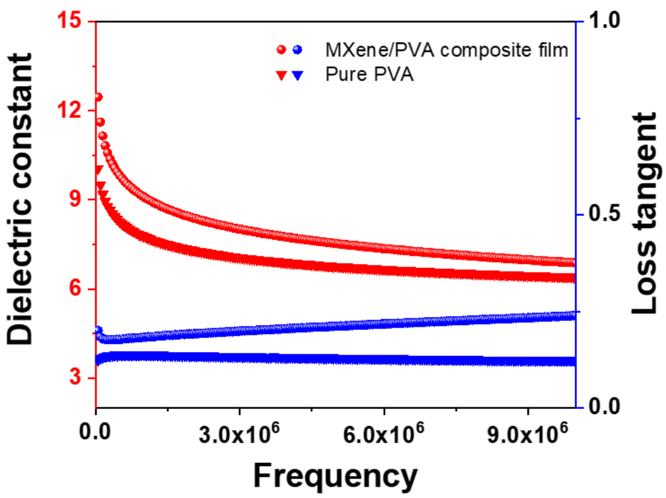

Figure S6 Dielectric constant and loss tangent of pure PVA and MXene-PVA composite film.

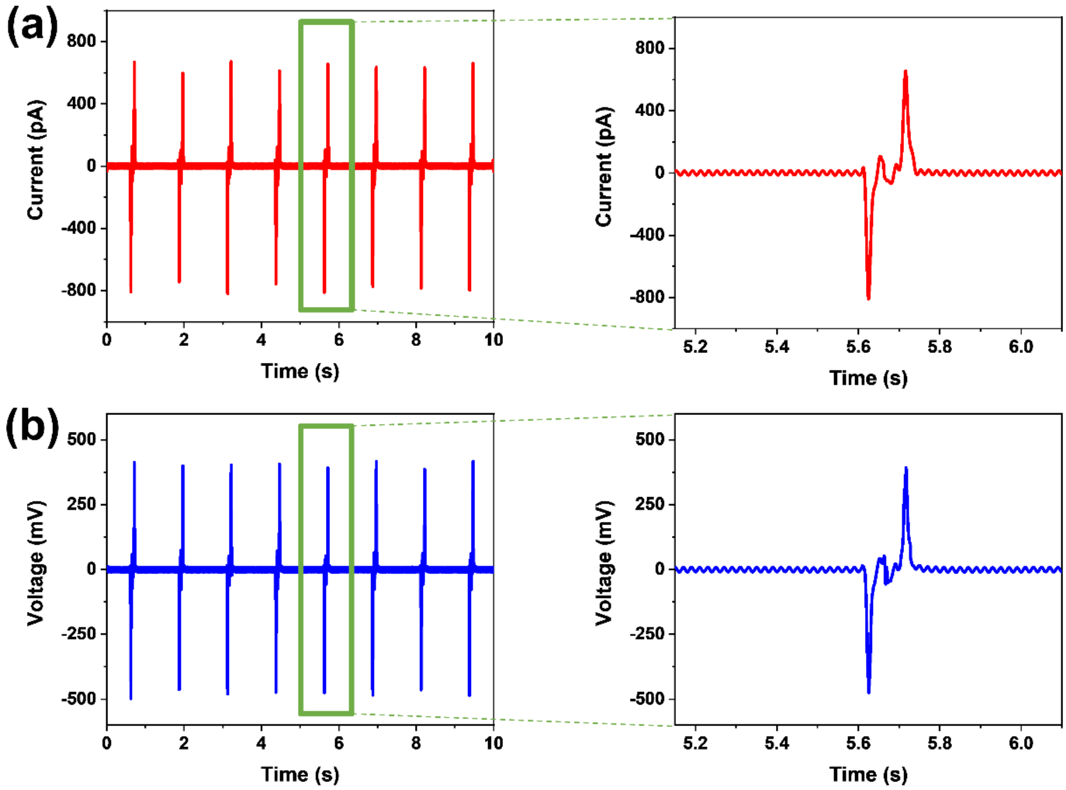

Figure S7 Switched polarity in current (a) and voltage (b) signals of MXene/PVA-PENG at 3.47 N compressive force with forward and reverse connection, respectively.

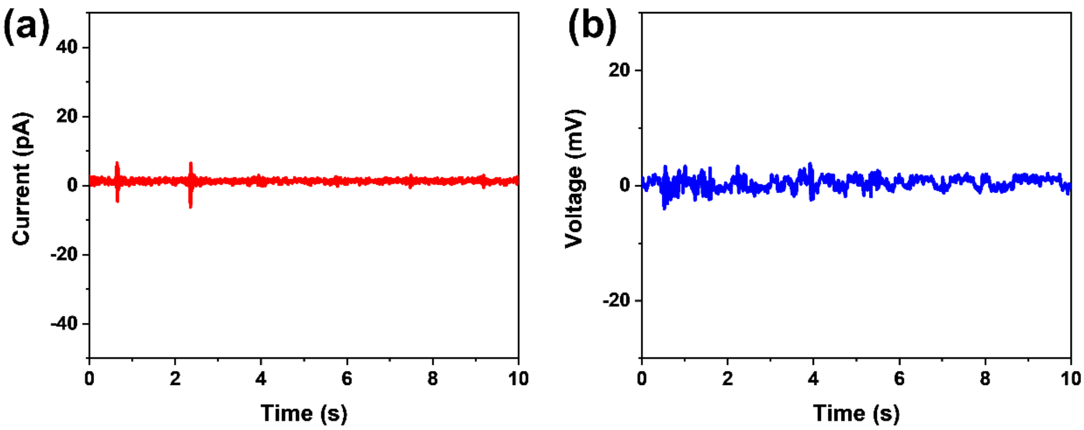

Figure S8 The output signals of pure PVA device. (a) Output current. (b) Output voltage.

Table S1 The comparison of common MXene synthesis methods

Method	**Temperature**	**Etching Agent**	**Processing Time**	**Yield**	**Safety concern**	**Scalability**
HF Etching [1, 2]	RT	HF	24-72 h in air	~40-98%	Hazardous HF handling required	Limited by safety concerns
LAMS (Lewis Acid Molten Salt) [3-5]	>550°C	Lewis acidic salts ($ZnCl_2$, $CuCl_2$, $FeCl_2$, etc.)	4-24 h in inert gas (heating and cooling time excluded)	~50-75%	High temperature, inert gas required	Good - Scalable process with few safety limitations
MS³ (salt shielded) [6]	~700°C	Molten salts	10-40 mins in air (heating and cooling time excluded)	Not reported	High temperature process	Moderate
Thermal-assisted electrochemical etching (this work and ref. [7])	25-80°C (optimally at 50°C)	Diluted HCl	3-9 h in air	~50% (up to ~75% with recycling)	Diluted HCl	Good - Simple setup, recyclable
MS-E-etching (Molten-salt-assisted electrochemical etching) [8]	450°C	LiCl-KCl melts	12 h in inert gas	~95%	High temperature, inert gas required	Good - One-pot process, reusable salts

Table S2 Comparison of output performance in MXene-based PENGs. V_{oc}: open-circuit voltage; I_{sc}: short-circuit current.

PENGs	Applied strain/force	V_{oc} (mV)	I_{sc} (pA)	Power density (mW m^{-2})
Monolayer $Ti_3C_2T_x$ MXene-PENG [9]	1.08%	~0.27	300	6.5
Monolayer oxide $Ti_3C_2T_x$ MXene-PENG [10]	1.30%	~1.25	~1150 ± 200	~31.04
MXene/PVA-PENG (this work)	3.47 N	500	790	0.14

Reference

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