

# Supporting Information

## **Sono-assisted surface energy driven assembly of 2D materials on flexible polymer substrates: A green assembly method using water**

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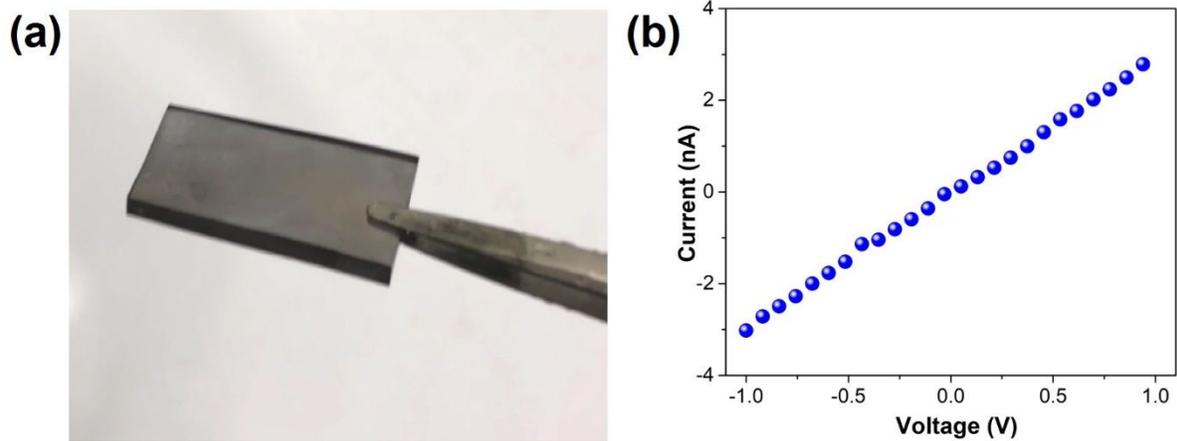
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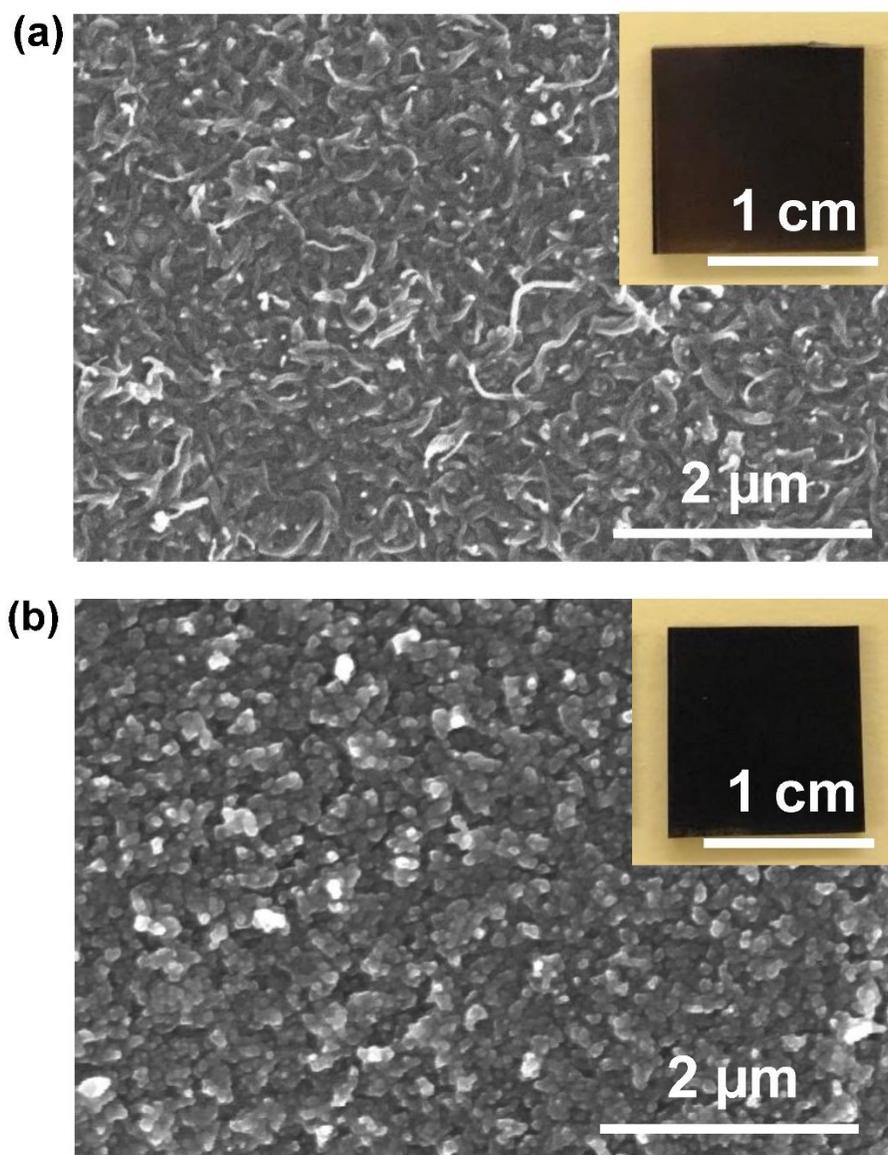
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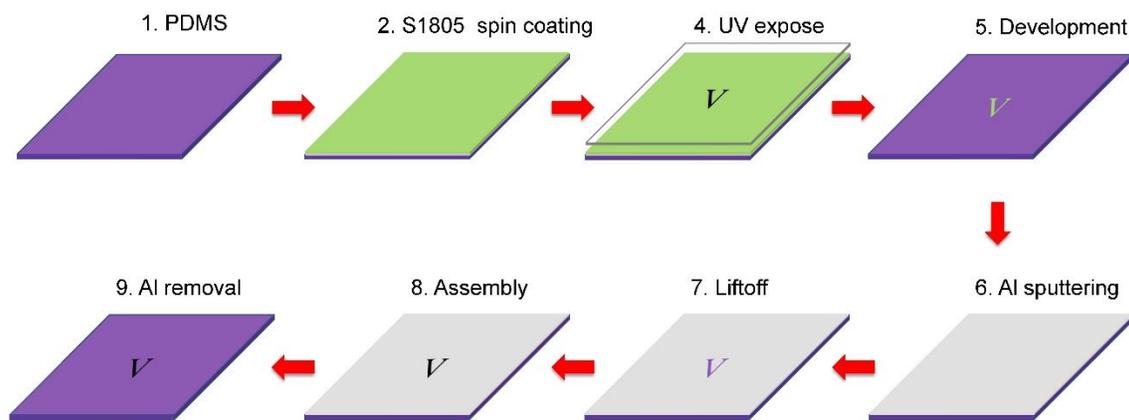
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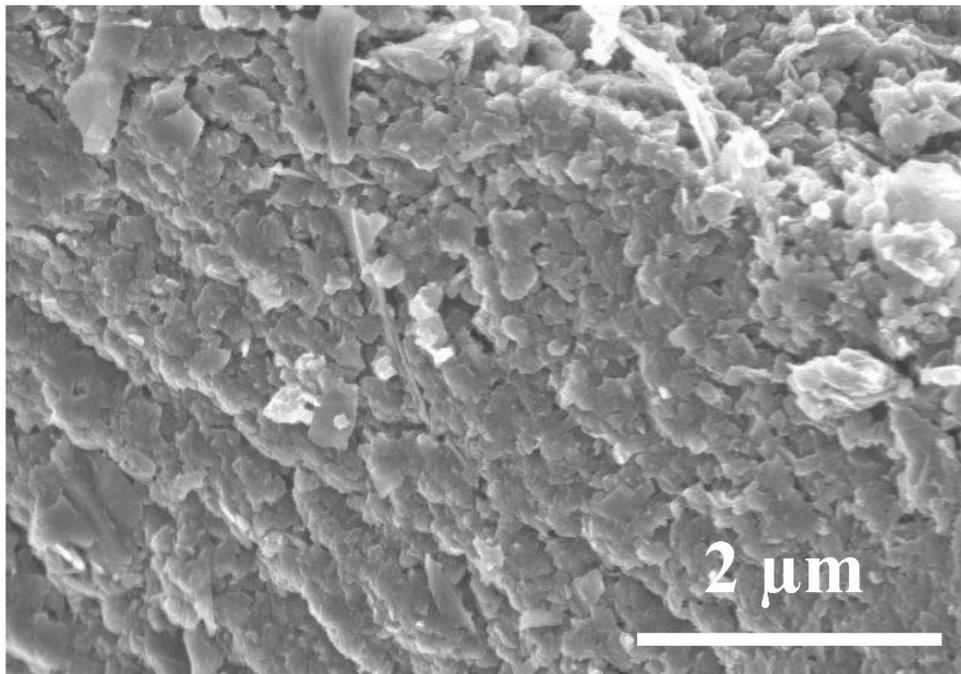
**Figure S1.** 10 s assembly of graphene on PDMS. (a), Digital camera image. (b), The corresponding I-V curve. Solution concentration 1 mg/mL.



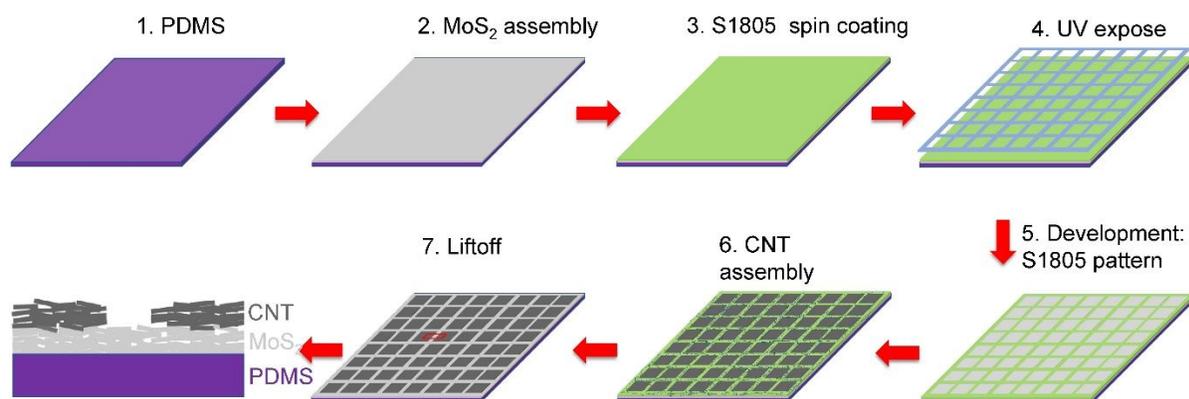
**Figure S2.** SEM images of assembled carbon nanotube **a** and carbon black **b** on PDMS substrates.



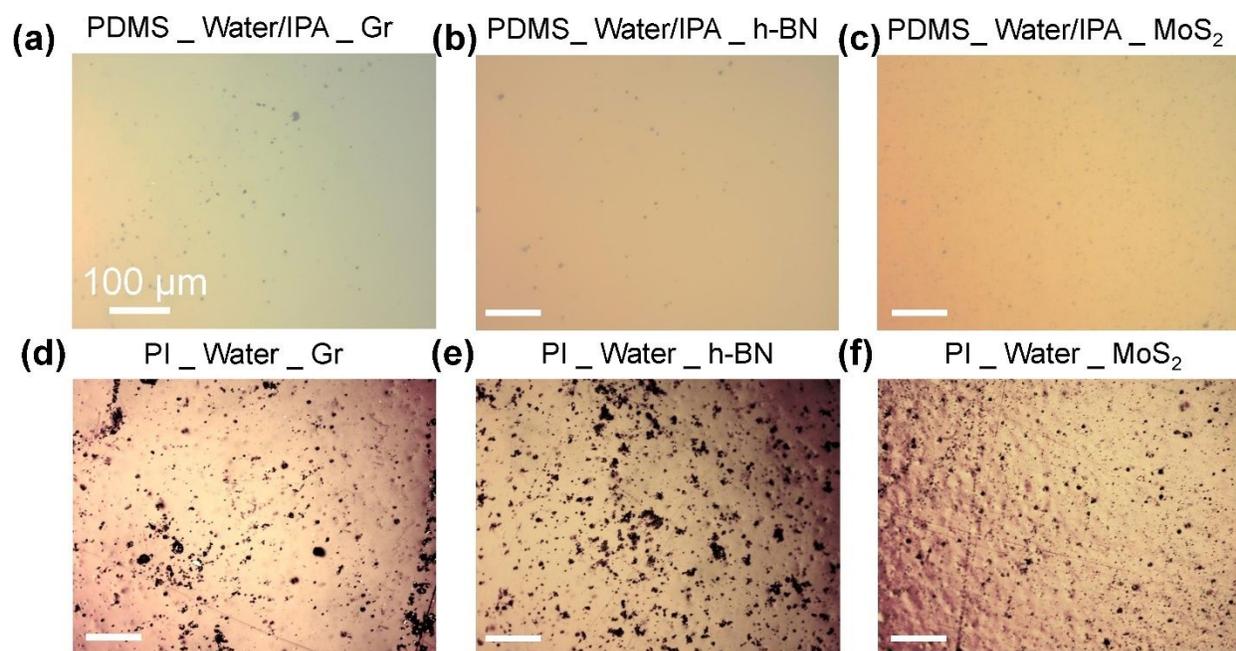
**Figure S3.** Sample preparation procedure for graphene assembly into a micro “V” letter.



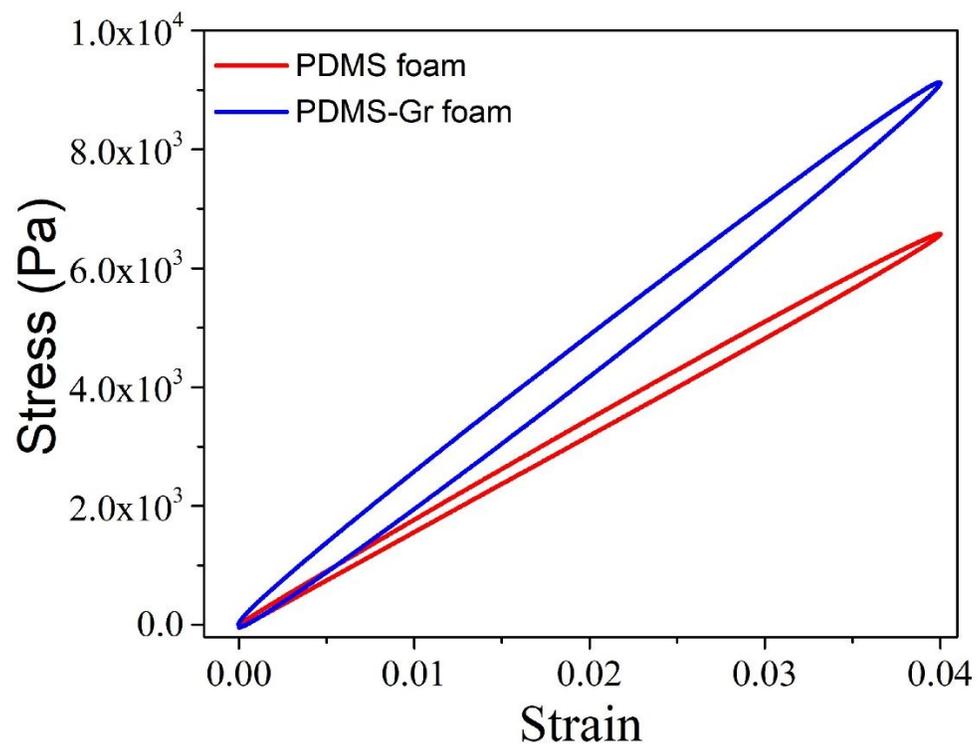
**Figure S4.** High magnification SEM image of assembled PDMS/graphene foam wall.



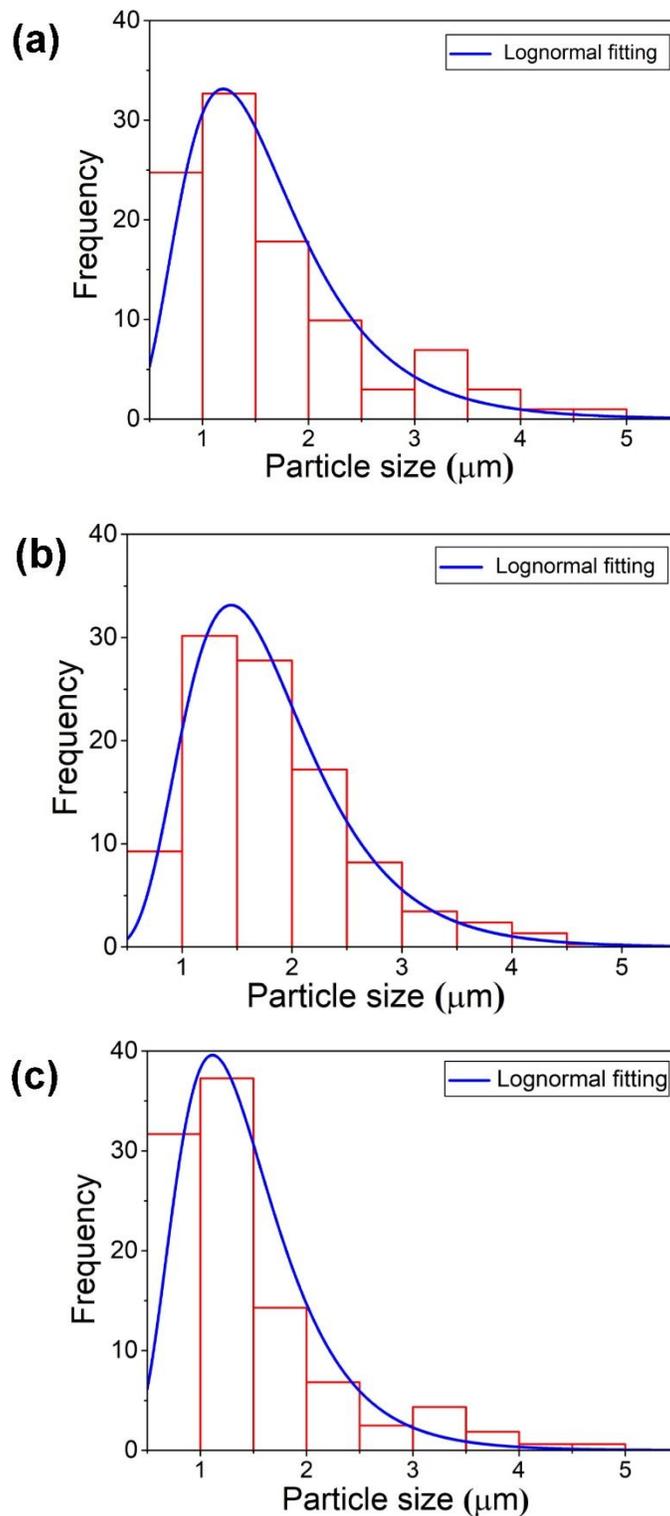
**Figure S5.** Combined micropatterning and multiple SASEDA processes Multiple SASEDA processes for MoS<sub>2</sub>/CNT microscale device fabrication.



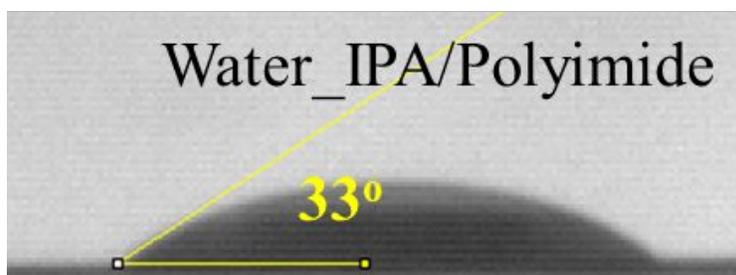
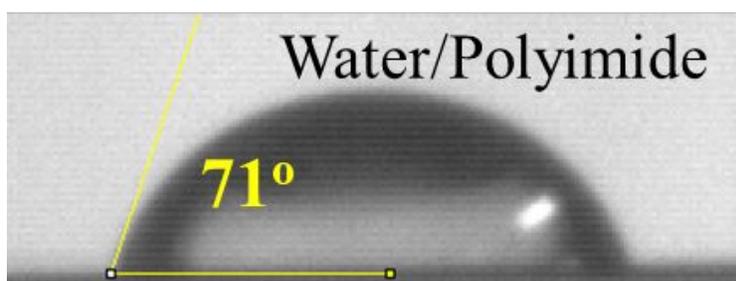
**Figure S6.** Optical images of static assembled samples. (a) (b) (c) are graphene, h-BN, and MoS<sub>2</sub> assembly on PDMS in water/IPA, respectively. (d) (e) (f) are graphene, h-BN and MoS<sub>2</sub> assembly on PI in water, respectively. The scale bars are 100 μm.



**Figure S7.** Mechanism properties of pure PDMS foam and assembled graphene/PDMS foam



**Figure S8.** Size distribution of graphene C-300 (a), h-BN (b) and MoS<sub>2</sub> (c). The projected area diameters were calculated and summarized. At least 100 particles were measured for each sample.



**Figure S9.** Contact angle data on polyimide substrate for water and water/IPA (1:1).

**Table S1.** Calculated interfacial energy and separation energy.

	$\gamma_{pn}$	$\gamma_{ps}$	$\gamma_{ns}$	$W_{psn}$	$W_{pns}$
PDMS/Water/Gr	11.62	38.84	13.89	41.11	-13.33
PDMS/Water/h-BN	22.1	38.84	2.95	19.69	-13.79
PDMS/Water/MoS <sub>2</sub>	10.12	38.84	11.96	40.68	-16.76
PDMS/Water_IPA/Gr	11.62	3.91	5.31	-2.4	13.02
PDMS/Water_IPA /h-BN	22.1	3.91	8.37	-9.82	26.56
PDMS/Water_IPA /MoS <sub>2</sub>	10.12	3.91	3.27	-2.94	9.48
Polyimide/Water/Gr	24.47	11.2	13.89	0.62	27.16
Polyimide/Water/h-BN	12.96	11.2	2.95	1.19	4.71
Polyimide/Water/MoS <sub>2</sub>	19.2	11.2	11.96	3.96	19.96

**Table S2.** The summary of surface tension and surface tension components

	Surface tension	Dispersive components	Polar components	Reference
Water	72.75	22.10	50.65	1
Water/IPA (1:1)	25.13	16.96	8.17	1
Graphene	53.0	39.1	13.9	2
MoS <sub>2</sub>	44.5	32.09	14.41	3
h-BN	58.27	27.99	30.28	4
PDMS	19.8	19.0	0.8	5
Polyimide <sup>a)</sup>	37.2	29.1	8.1	

<sup>a)</sup>The surface tension and components of polyimide were calculated using the contact angles from Figure. S9 and the Fowkes surface energy theory: <sup>2</sup>

$$\gamma_l(\cos\theta + 1) = 2(\gamma_l^{\text{di}}\gamma_s^{\text{di}})^{1/2} + 2(\gamma_l^{\text{po}}\gamma_s^{\text{po}})^{1/2}$$

where  $\gamma_l^{\text{di}}$  and  $\gamma_l^{\text{po}}$  are the liquid dispersive and polar components, respectively, and  $\gamma_s^{\text{di}}$  and  $\gamma_s^{\text{po}}$  are the solid dispersive and polar components, respectively.  $\theta$  is the contact angle.

#### Supplementary References

1. Shen, J.; He, Y.; Wu, J.; Gao, C.; Keyshar, K.; Zhang, X.; Yang, Y.; Ye, M.; Vaita, R.; Lou, J. *et al.* Liquid Phase Exfoliation of Two-Dimensional Materials by Directly Probing and Matching Surface Tension Components. *Nano Lett.* **2015**, *15*, 5449-5454.

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