

# **Large volume variation of anisotropic graphene nanosheet electrochemical-mechanical actuator under low voltage stimulation**

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Experimental description:

GO was synthesized from natural graphite flake (325 mesh, 99.8%, ABCR GmbH & Co. KG) by a modified Hummers method and has been described in our recent work.<sup>1</sup> The reduction of GO into RGO followed the procedure described by Kovtyukhova and colleagues.<sup>2</sup> The obtained RGO solution was subjected to dialysis to completely remove residual hydrazine. The obtained RGO was filtrated through cellulose membrane obtaining a water containing gel. The weight percent of RGO in the gel is 3wt%. 600mg gel containing 18mg RGO was dispersed in 10 mL N-Methyl pyrrolidone with horn sonication in ice water bath under 0.5s on and 2s off at a 200W and 20 kHz frequency output for accumulated 15 minute sonication time using a Fisher Scientific model 500 digital sonic dismembrator to obtain a RGO solution. The as dispersed solution of RGO was casted on glass substrate. Glass substrates were placed on a horizontal platform in the oven to make sure the membrane of uniform thickness. The amount of casted solution was 2mL each time and the casting process was repeated five times to make sure total 10mL solution was casted. The casting evaporation process was achieved in the oven in 60°C and kept overnight to obtain uniform membranes and further vacuum dried in 150°C overnight obtained a fully dehydrated membrane on glass substrate. The sample was then scraped into two paralleled strips of 2mm width

and 20mm long, then silver paste was casted on the outer edge of both two paralleled strips to connect electrical conductive wires. For composite in-plane actuator fabrication, RGO gel and BmimBF<sub>4</sub> was added into N-Methyl pyrrolidone solution and horn sonicated in ice water bath followed by the above mentioned RGO membrane preparing procedure. The obtained solid membranes were preserved in vacuum dryer for characterization. FESEM images were recorded by Hitach S-4800. High resolution TEM images were recorded by FEI Tecnai G2 F20 S-Twin 200KV. Electrochemical performance was recorded by CHI660C electrochemical work station. Thickness variation was recorded by Keyence LK-G800 laser positioning system.

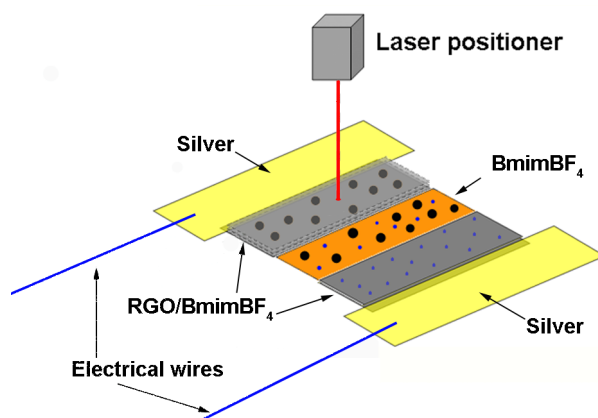


Figure S1. Illustration of in-plane graphene electrodes actuation characterization

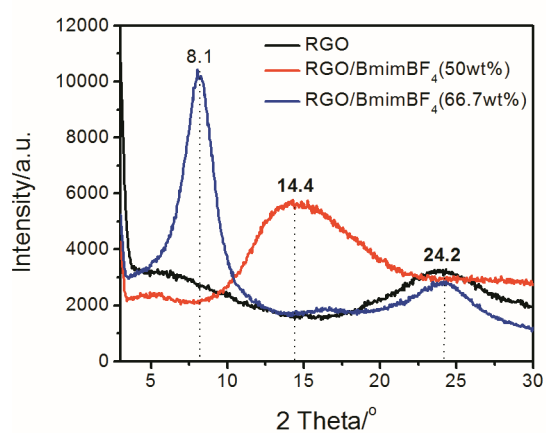


Figure S2. XRD patterns for RGO/IL membranes containing 0 wt%, 50 wt%, 66.7 wt% IL respectively

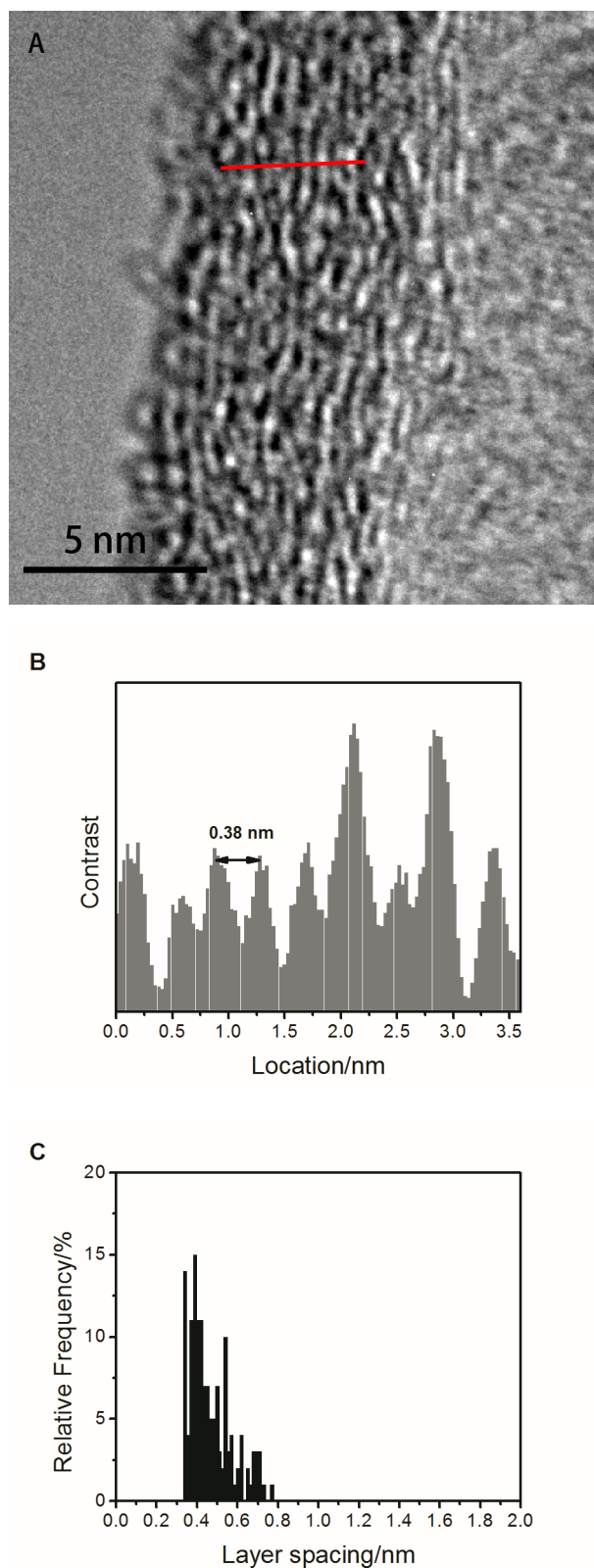


Figure S3. (A) High-resolution TEM image of RGO, (B) cross section taken along the lines in (A), (C) statistical treatment of layer spacing distribution of RGO.

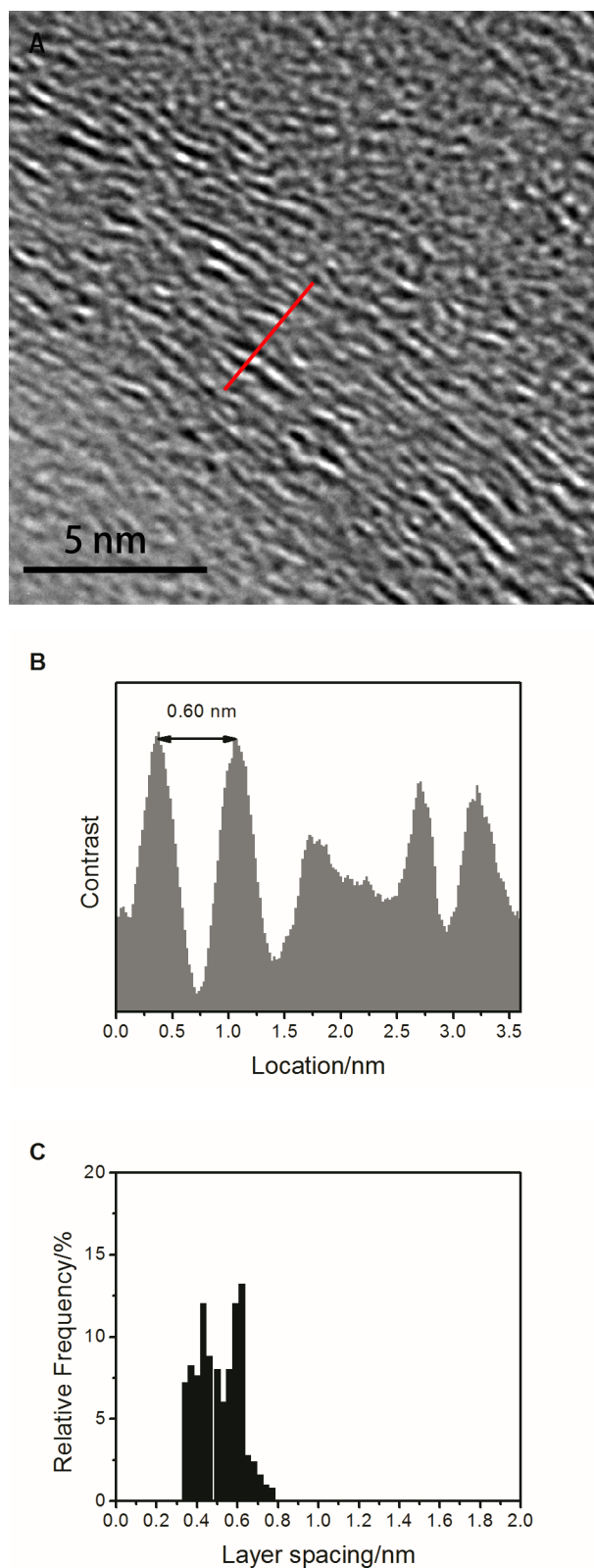


Figure S4. (A) High-resolution TEM image of RGO/BmimBF<sub>4</sub> containing 50% BmimBF<sub>4</sub>, (B) cross section taken along the lines in (A), (C) statistical treatment of layer spacing distribution of RGO/BmimBF<sub>4</sub> containing 50% BmimBF<sub>4</sub>.

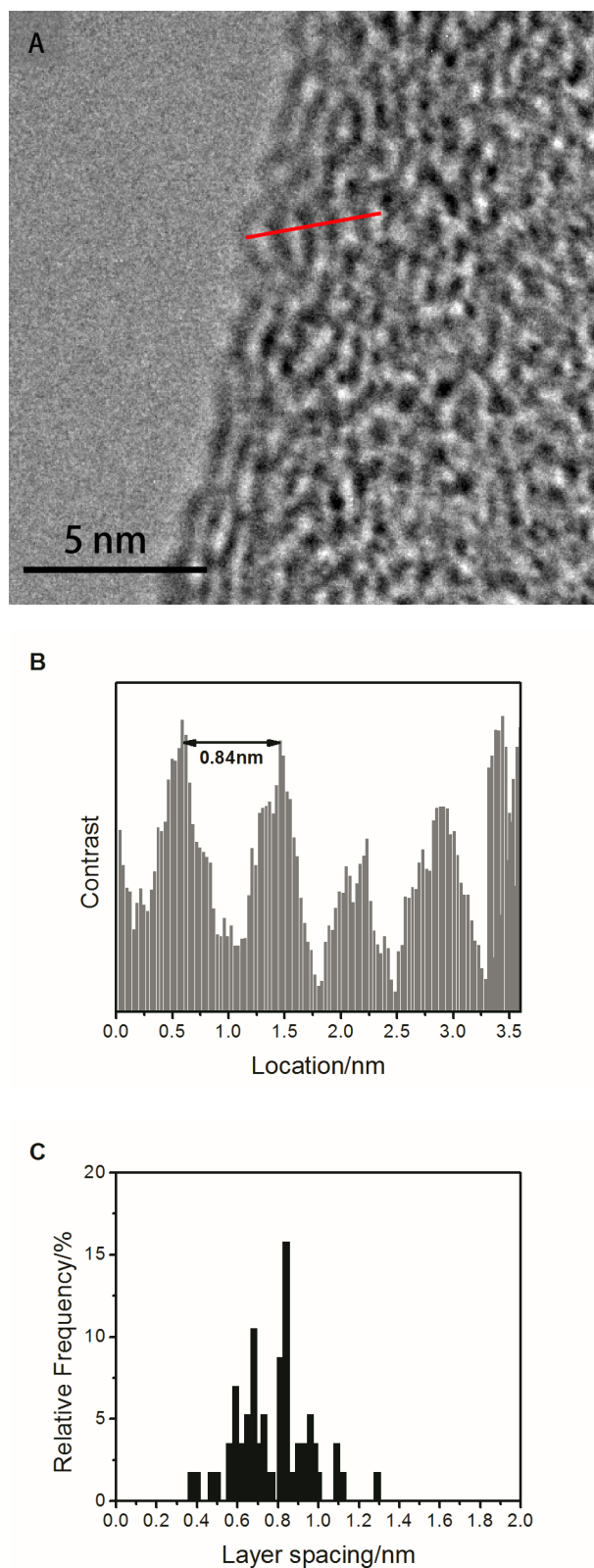


Figure S5. (A) High-resolution TEM image of RGO/BmimBF<sub>4</sub> containing 66.7% BmimBF<sub>4</sub>, (B) cross section taken along the lines in (A), (C) statistical treatment of layer spacing distribution of RGO/BmimBF<sub>4</sub> containing 66.7% BmimBF<sub>4</sub>.

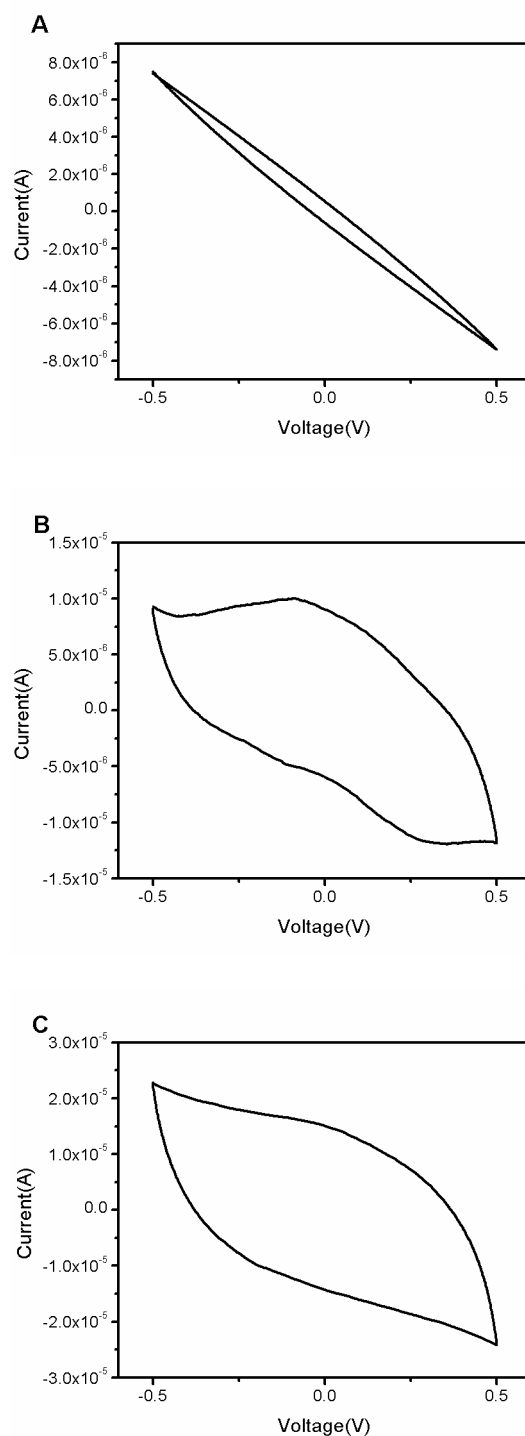


Figure S6. Cyclic voltammetry curves in scanning speed of 1mV/s for composite actuators with RGO electrodes containing (A) 0wt%, (B) 50wt%, (C) 66.7wt% ionic liquid BmimBF<sub>4</sub> separately.

The calculation of specific capacitance followed by the equation:

$$C = \frac{1}{s \cdot m \cdot \Delta V} \int_{V_0}^{V_0 + \Delta V} i dV$$

S is the scanning speed of CV, 1mV/s

m is the electrode weight, 0.000384 g

$\Delta V$  is the voltage range for CV, 1 V

The integration is the cumulated electrical quantity in the CV process

The specific capacitance for actuators have been found to be of 1.97F/g for pristine RGO, 30.68F/g for composite membrane of 50wt% and 60.00F/g for that of 66.7 wt% BmimBF<sub>4</sub> separately.

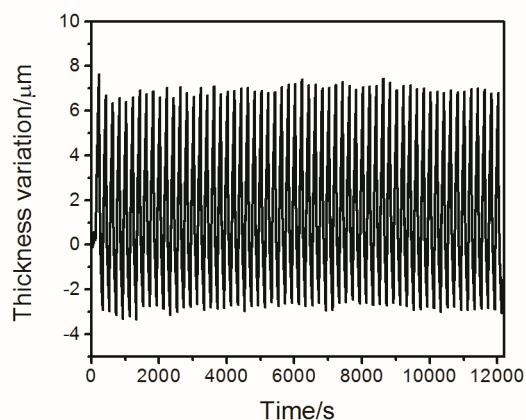


Figure S7. Thickness variation curve for in-plane actuator containing 50wt% BmimBF<sub>4</sub> under 0.005Hz, 2V square wave electrical stimulation.

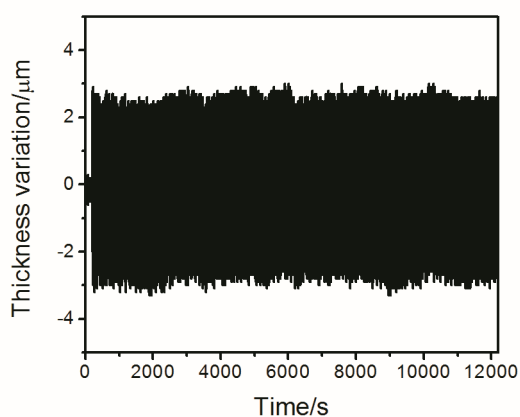


Figure S8. Thickness variation curve for in-plane actuator containing 66.7 wt% BmimBF<sub>4</sub> under 0.05Hz, 2V square wave electrical stimulation.

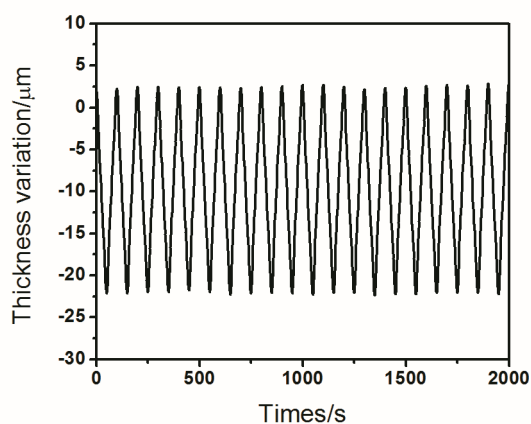


Figure S9. Thickness variation curve for in-plane actuator containing 66.7 wt% BmimBF<sub>4</sub> under 0.01Hz, 2V square wave electrical stimulation.

## References:

- 1 J. Liu, Z. Wang, L. Liu, W. Chen, *Phys Chem Chem Phys.* **2011**, 7, 13216
- 2 D. Li, M. B. Müller, S. Gilje, R. B. Kaner, G. G. Wallace, *Nature Nanotechnology* **2008**, 3, 101