Supporting Information

An Asymmetrically Surface-Modified Graphene Film Electrochemical Actuator

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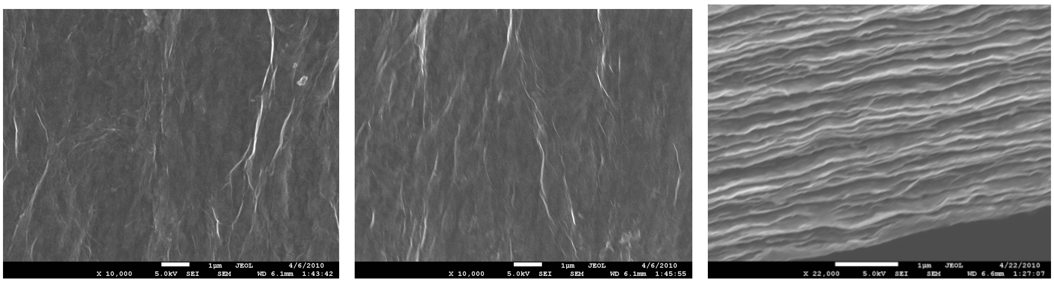


(a) (b) (c)

Figure S1. SEM images of two sides (a, b) and cross-section (c) view of the as-prepared graphene film. Scale bars: 1μm.

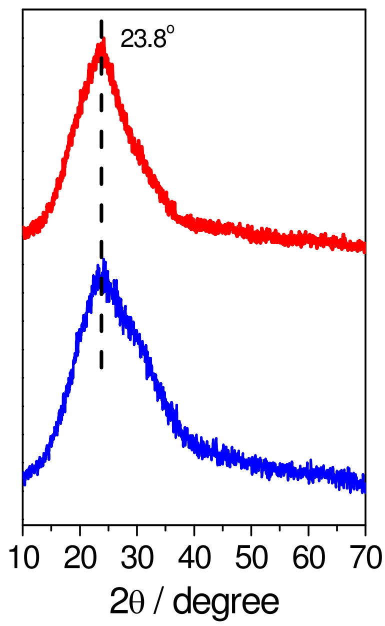


Figure S2. XRD patterns of the opposite surfaces of the as-prepared graphene film.

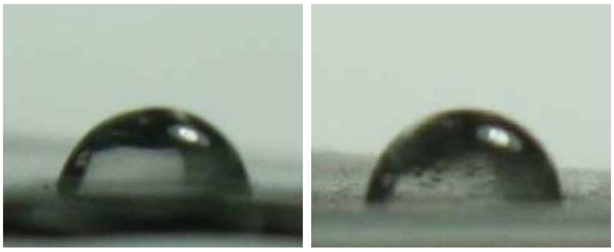


Figure S3. Photos of a 5 μL water droplet on the opposite surfaces of the as-prepared graphene film, showing the similar water contact angle of ~ 75°.

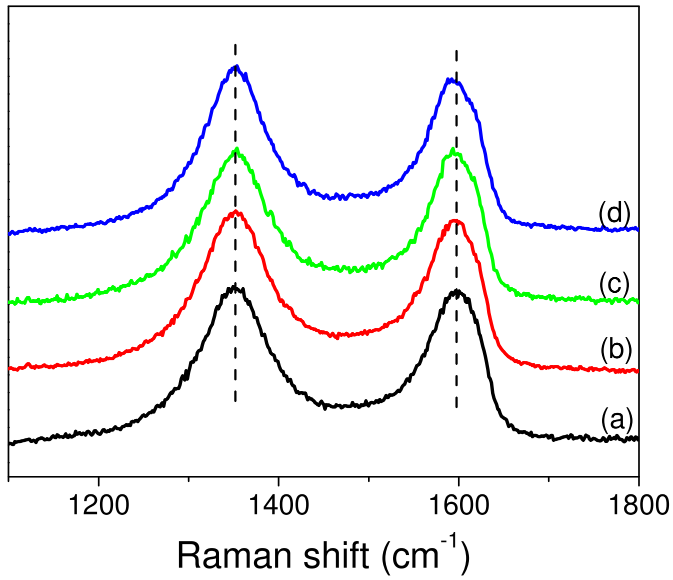


Figure S4. Raman spectra of the surfaces of the graphene film (a), Hexane-plasma-treated graphene film (b) under positive charge (c) or negative charge (d) (514 nm laser).

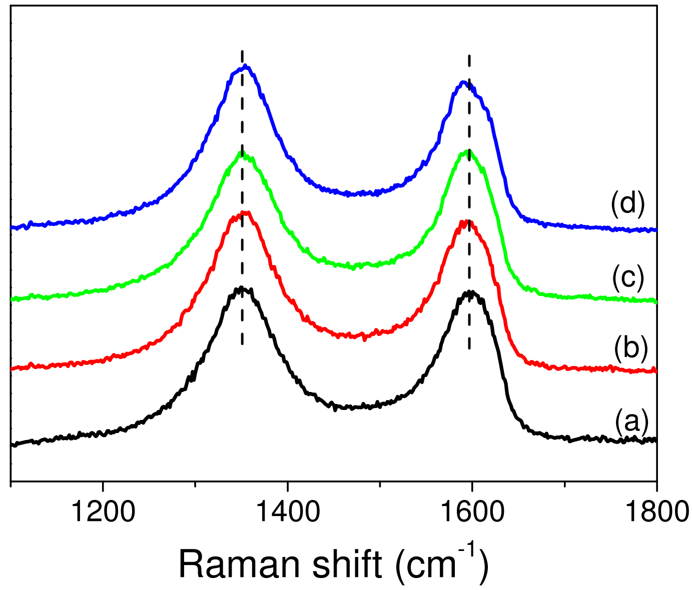
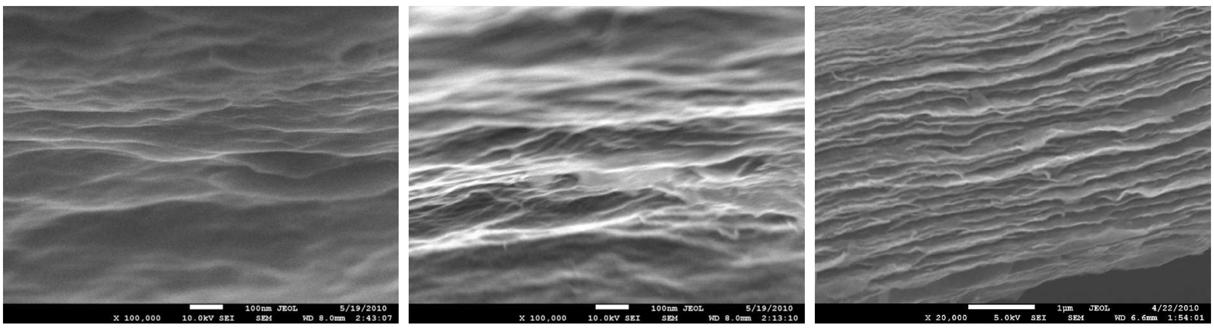


Figure S5. Raman spectra of the surfaces of the graphene film (a), the O_2-plasma-treated graphene film (b) under positive charge (c) or negative charge (d) (514 nm laser).



(a) (b) (c)

Figure S6. SEM images of the hexane (a), O_2 (b) plasma treated surfaces, and the cross section (c) of the graphene film after actuation investigation. Scale bars: 1μm.

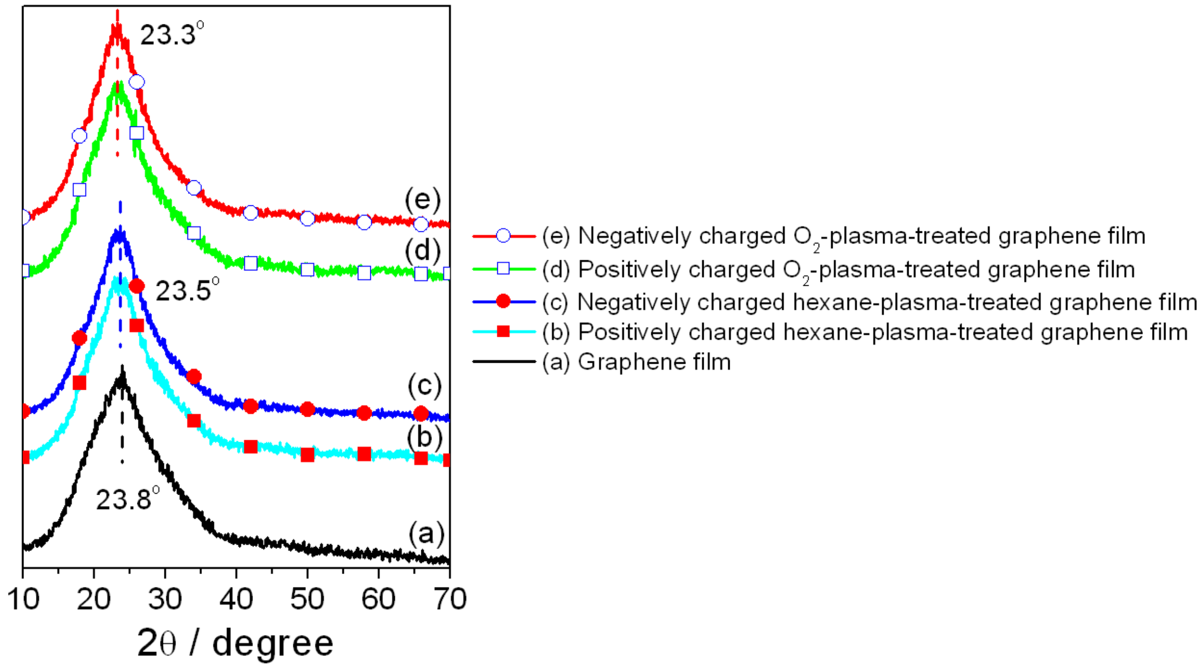


Figure S7. XRD patterns of the surfaces of the original graphene film (a) with the hexane-plasma treatment at positively (b), negatively (c) charged status, and the O_2-plasma treatment at positively (d), negatively (e) charged status, respectively.

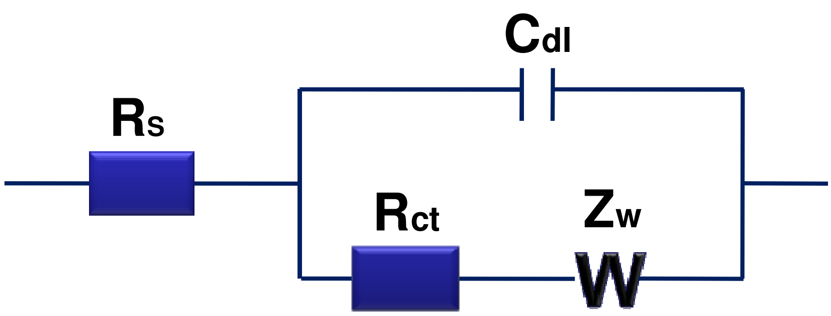


Figure S8. Randles circuit with Warburg impedances. (R_s = Resistance of the solution, C_{dl} = Double layer capacitance, R_{ct} = Charge transfer resistance and Z_w = Warburg impedance).

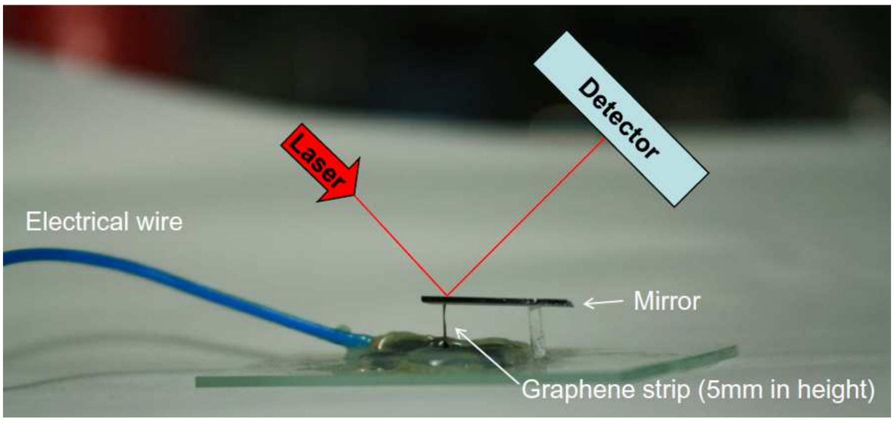
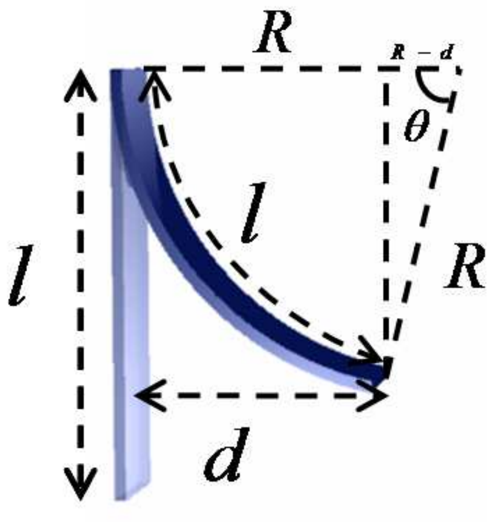


Figure S9. The setup for measuring the length changes of the graphene strips treated with Hex-P and O_2-P under applied square wave potential.

A cut strip (1.5mm in width) of graphene film was anchored perpendicularly to glass slide by conductive epoxy as working electrode, which was connected with an electrical wire and covered with nonconductive silicon rubber. A glass slide with the same height to the graphene strip was also stood on glass by superglue. A thin oxidized Si (SiO_2/Si) wafer supported by graphene strip and glass slide was used as the mirror for laser reflection. The reflected laser, associated with the length change of graphene strip, was recorded by a displacement detector. The actuation investigation was carried out by using an Ag/AgCl reference electrode and a Pt counter electrode in 1 M $NaClO_4$ aqueous solution.



Scheme S1. The relative parameters of a bending graphene strip for curvature calculation.

The curvature is defined to be the reciprocal radius (1/R). Scheme S1 exhibits the relative parameters of a bending graphene strip for curvature calculation. Each parameter is defined as follows:

l: The length of the graphene actuator.

R: The radius to the arc of the curved graphene actuator.

θ: The angle of the arc of the curved graphene actuator.

d: The horizontal displacement of the end of graphene actuator.

Obviously,

$$\cos\theta = \frac{R - d}{R} \tag{1}$$

Accordingly to the Taylor expression:

$$\cos\theta = 1 - \frac{\theta^2}{2} + \frac{\theta^4}{2\cdot3\cdot4} - \frac{\theta^6}{2\cdot3\cdot4\cdot5\cdot6} + \cdots \approx 1 - \frac{\theta^2}{2} \qquad (2)$$

The equation could be extracted as below.

$$1 - \frac{\theta^2}{2} = \frac{R-d}{R} \qquad (3)$$

In terms of the equation:

$$R\theta = l \qquad (4)$$

It is deduced that

$$Curvature = \frac{1}{R} = \frac{2d}{l^2} \qquad (5)$$