Electronic supplementary information

Load-Tolerant, Highly Strain-Responsive Graphene Sheets

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Experimental Section

Graphene oxide was first prepared from natural graphite powder via acid-oxidation according to a modified Hummers method reported in our previous paper.[1] Reduction of the graphene oxide was then carried out with aqueous N_2H_4 in the presence of ammonia. Briefly, 2 ml 0.2 mg/ml graphene oxide was treated with 200 μl concentrated ammonium hydroxide (NH_4OH) and ~15 μl hydrazine monohydrate ($N_2H_4 \cdot H_2O$) for 1 hour at 90^{o}C. The freestanding graphene film was made by direct filtration of the aqueous reduced graphene oxide colloidal suspensions through a filter membrane with pore size of 220 nm.[2,3] The dried graphene film with a thickness of ca. 6 μm was prepared by mechanically peeled from the filter. The measured conductivity is ca. 1×10^3 S/m. These freestanding graphene film composed of layer-by-layer graphene sheets, without further optimization of its mechanical strengths or electrical properties, was used for subsequent fabrication of the graphene actuator.

Plasma treatment was carried out on a custom-built plasma apparatus powered at 13.56 MHz, 200 W (Cesar 133 RF power generator) and a pressure of 100 Pa with O_2 flow. The plasma was applied for less than 10 seconds in order to avoid possible surface damage of the graphene film.

Strain-responsive behavior was investigated in a three electrode one-cell system with a Pt wire being used as counter electrode. A certain weight was placed on the center of the mirror for load-tolerant test (Figure 1). Electrolyte is the aqueous solution 1 M $NaClO_4$. All of the electrochemical measurements in this study were carried out by using Ag/AgCl as reference electrode. Nyquist plots were obtained with amplitude of 5mV, frequency ranging 0.5Hz to 106 Hz and DC cell potential of 1 V. The electrochemical analysis and measurements of the graphene film were performed by use of a CHI660D electrochemical workstation. The morphology of the samples was examined by scanning electron microscopy (SEM, JSM-7500F).

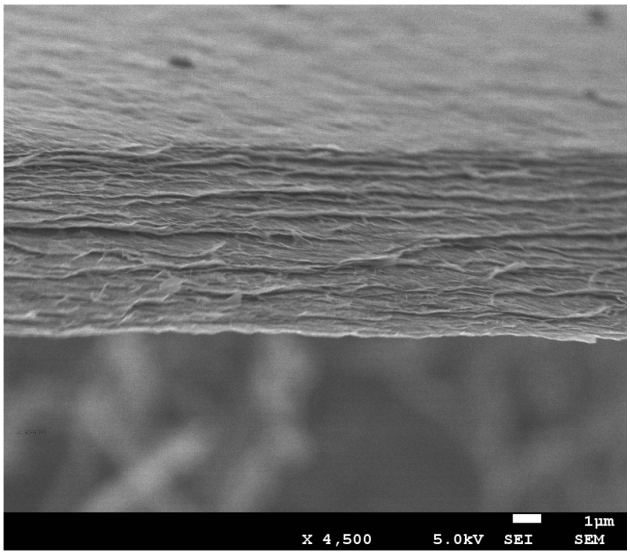

Fig. S1. SEM image of graphene film prepared by filtration.

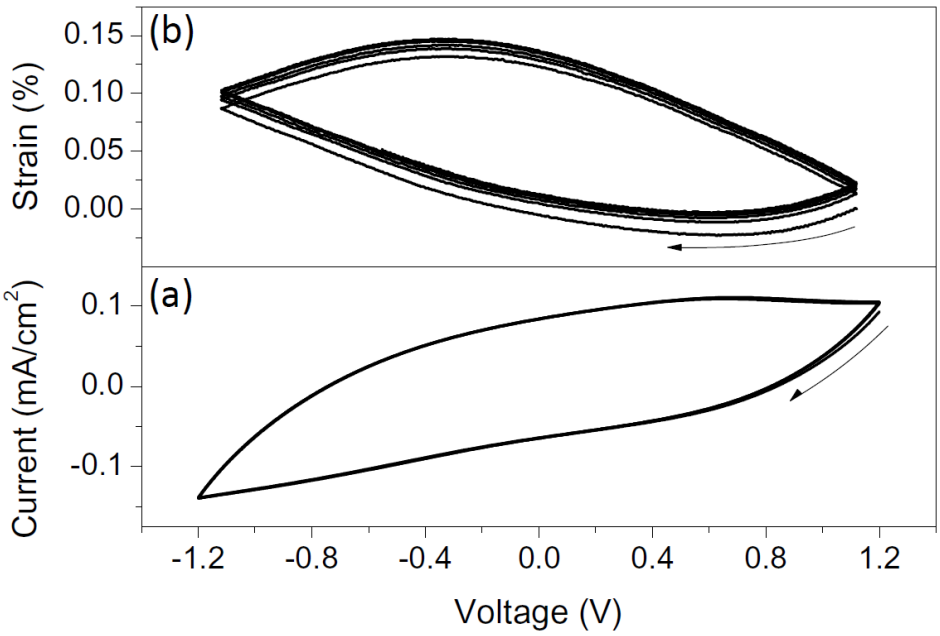

Fig. S2. Cyclic voltammograms (a) and corresponding strain response (b) of a strip-type graphene film. Scan rate: 100mV/s.

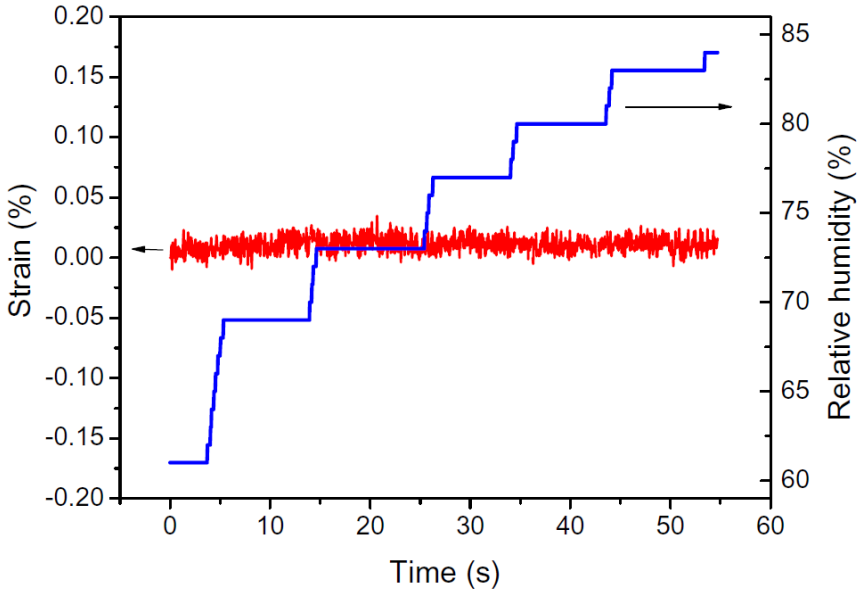

Fig. S3. Strain response of graphene strip under the changes of humidity.

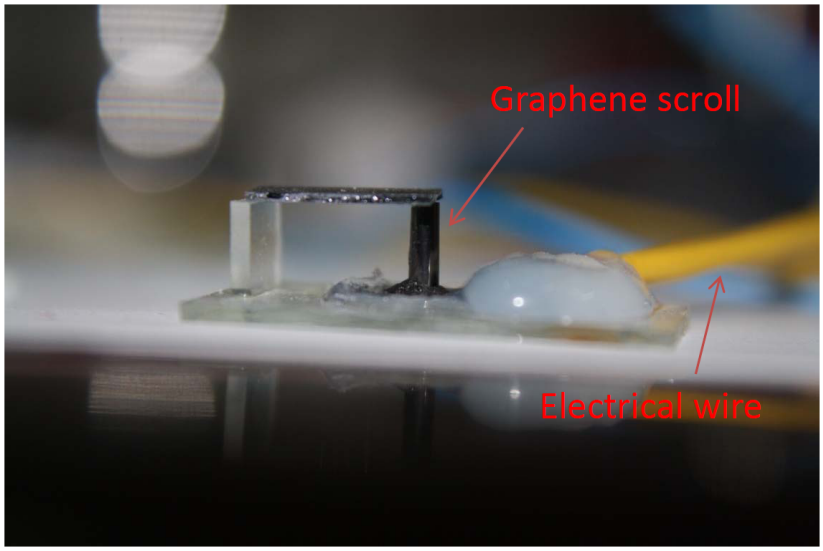

Fig. S4. A photo for a final fabricated device for characterizing the scroll-type graphene film (3mm in diameter).

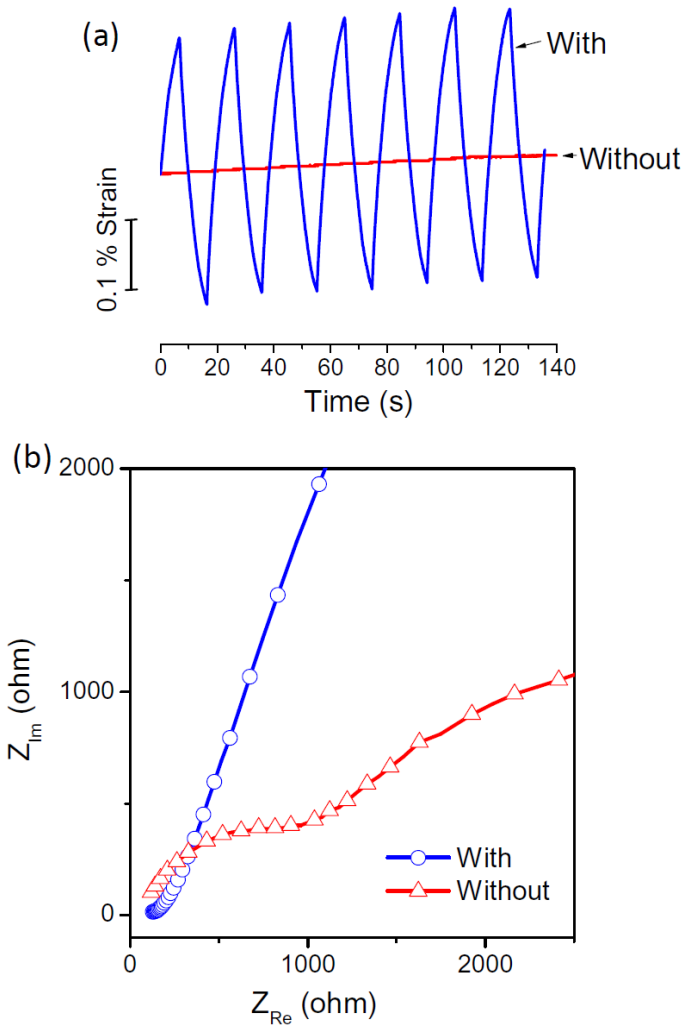

Fig. S5. (a) The comparison of strain response of graphene film with and without O_2 plasma treatment at applied square wave potential of ±1.2V and a frequency of 0.05 Hz, and (b) Nyquist plots of graphene film with and without O_2 plasma treatment.

1 Y. X. Xu, H. Bai, G. W. Lu, C. Li and G. Q. Shi, *J. Am. Chem. Soc.*, 2008, **130**, 5856.

2 H. Chen, M. B. Muller, K. J. Gilmore, G. G. Wallace and D. Li, *Adv. Mater.*, 2008, **20**, 3557.

3 D. Li, M. B. Muller, S. Gilje, R. B. Kaner and G. G. Wallace, *Nat. Nanotechnol.*, 2008, **3**, 101.