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. Reduction of the graphene oxide was then carried out with aqueous N₂H₄ in the presence of ammonia. Briefly, 2 ml 0.2 mg/ml graphene oxide was treated with 200 μl concentrated ammonium hydroxide (NH₄OH) and ~15 μl hydrazine monohydrate (N₂H₄·H₂O) for 1 hour at 90°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. 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 × 10³ 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 O₂ 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.

