Supplementary Information

Ultrahigh-rate supercapacitors based on eletrochemically reduced graphene oxide for ac line-filtering

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1. Supplementary Materials and Methods

Synthesis and purification of GO. Graphite (3.0 g) was added to concentrated sulfuric acid (70 mL) under stirring at room temperature, then sodium nitrate (1.5 g) was added, and the mixture was cooled to 0 °C. Under vigorous agitation, potassium permanganate (9.0 g) was added slowly to keep the temperature of the suspension lower than 20 °C. Successively, the reaction system was transferred to a 35 ± 5 °C water bath and stirred for about 0.5 h, forming a thick paste. Then, 150 mL of water was added, and the solution was stirred for 15 min at 90 ± 5 °C. Additional 500 mL of water was added and followed by a slow addition of 15 mL of H₂O₂ (30%), turning the color of the solution from dark brown to yellow. The mixture was filtered and washed with 1:10 HCl aqueous solution (250 mL) to remove metal ions followed by

washing with 200 mL of water to remove the acid. The resulting solid was dried in air and diluted to make a GO aqueous dispersion (0.5% w/w). Finally, it was purified by dialysis for one week using a dialysis membrane (Beijing Chemical Reagent Co., China) with a molecular weight cut off of 8,000 to 14,000 g mol⁻¹ to remove the remaining metal species.

Fabrication of ErGO electrodes. A 3 mg mL⁻¹ GO dispersion containing 0.1 M LiClO₄ (99.0%, Sinopharm Chemical Reagents Co. Ltd, Beijing, China) was used as the electrolyte. A gold foil $(0.02 \times 1 \times 3 \text{ cm})$ coated with a 3M tape on its one surface was used as the working electrode, a platinum (Pt) foil was applied as the counter electrode. All the potentials were referred to a saturated calomel electrode (SCE). The electrochemical deposition of ErGO was carried out at room temperature under a constant potential of -1.2 V for 5 -60 s using a CHI760D potentiostat-galvanostat (CH Instruments Inc.) under computer control. After electrodeposition, the graphene electrodes were washed with DI water and then immersed in DI water for 1 h to remove the residual GO absorbed on the electrodes. Successively, the ErGO electrodes were further reduced in 1 M LiClO₄ aqueous solution at -1.2 for another 30 s to increase their conductivity. Finally, the electrodes were immersed in DI water to remove LiClO₄.

Fabrication and Characterizations of ErGO-DLCs. The DLC was assembled in a symmetrical two-electrode configuration. Two identical ErGO electrodes each has a

ErGO region of 1.4 cm² were separated by a 120-µm thick microporous separator (Suzhou Beige New Materials & Technology Co. Ltd., Suzhou, china). The electrodes and the separator were wetted with the aqueous electrolyte (25% potassium hydroxide) for more than 1 h, then assembled into a layered structure and sandwiched between two pieces of Teflon slabs.

The charge/discharge tests were carried out at current densities (I_D) in the range of 40 to 800 μ A cm⁻² (the apparent surface area of the electrode is 1.4 cm²). The specific capacitance, C_s (μ F cm⁻²), was calculated by the use of following equation: $C_s = \frac{I_D}{(dV/dt)}$, where I_D is the current density (μ A cm⁻²), dV/dt is the slope of the discharge curve (V/s).

2. Structural characterizations of ErGO

The ErGO layer deposited on the surface of Au electrode was studied by Raman and x-ray photoelectron (XPS) spectra (Fig. S1). In comparison with the Raman spectrum of GO, the intensity ratio of D/G bands in the spectrum of ErGO is increased significantly, indicating the oxidized areas of GO sheets were partly restored upon reduction to forming small conjugated domains. In the C 1s XPS spectrum of ErGO, the peak associated with the oxygenated groups are much weaker than that in the spectrum of GO, revealing that most oxygen containing functional groups of GO were removed after the electrochemical reduction.



Figure S1 | **a**, 514.5 nm excited Raman spectra of GO and ErGO. **b**, C1s X-ray photoelectron spectra of GO and ErGO.



Figure S2 | Scanning electron microscope cross-sectional image of the typical ErGO

electrode with high magnification.

3. Supplementary figures related to the electrochemical performances of

ErGO-DLC



Figure S3 | The comparison of impedance phase angle versus frequency between the typical ErGO-DLC and commercial AEC.



Figure S4 | Specific capacitance versus frequency of the DLC with bare gold electrodes.



Figure S5 | Specific capacitance versus discharge current density in the range of 40 to $800 \ \mu A \ cm^{-2}$.



Figure S6 | **AC impedance characterizations of ErGO-DLC before and after the test of 10,000 cycles, in the frequency range of 1 to 100 kHz with a 5 mV ac amplitude. a,** Plots of impedance phase angle versus frequency. b, The Nyquist plots; insert: an expanded view of the high frequency region. **c,** Plots of specific capacitance versus frequency. **d,** Plots of imaginary part (**C**) of specific capacitance versus frequency.

5. Effect of electrodeposition time on the morphology of ErGO electrode and the performances of ErGO-DLCs.



Figure S7 | SEM images of the ErGO electrodes prepared by electrodeposition for different time. a,d, 5s. (d) b,e, 30 s. c, f, 60 s.



Figure S8 | Expanded view of the Nyquist plots of the DLCs based on the ErGO electrodes prepared by electrodeposition for different time.