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SUPPLEMENTARY INFORMATION

An ionic liquid template approach to graphene-carbon xerogel composites for supercapacitors with enhanced performance

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Experimental section

The graphene was made by *in situ* catalytic graphitization of dodecylamine by Fe species, and kindly supplied by Prof. Weiping Ding at Nanjing University of China, of which the typical TEM image is shown in Fig. S1. The graphene-CXs composites were prepared by sol-gel polymerization of resorcinol-formaldehyde with Na_2CO_2 as catalyst and ionic liquids as templates following a modified method reported by Shi et al. [1] Typically, 2.20 g of resorcinol, 3.25 g of formaldehyde and 9 g of distilled water were carefully mixed, then 22.1 mg of Na_2CO_3 and 0.22 g of ionic liquid $BMImBF_4$ synthesized in our laboratory were added into the mixture. After magnetically stirring for 1 h at room temperature (RT), graphene was added, yielding a mixture that was put into an autoclave. The autoclave was heated at 80 ^oC for 24 h before being cooled back to RT, yielding a wet gel that was dried at 130 ^oC under vacuum for 3 h to get a dry gel. The dry gel was carbonized at 800 ^oC for 5 h in flowing nitrogen of 20 ml min^{-1}, yielding the graphene-CXs composites that are denoted as *x*G/CX, where *x* stands for the mass percentage of graphene to resorcinol. For comparison, the pristine CX was also synthesized in the presence of ionic liquids under the same conditions.

The materials were examined by field emission scanning electron microscopy (FE-SEM, NOVA NanoSEM 450, FEI, USA). Raman spectra of the as-made samples were recorded using a DXR Raman Microscope (Thermal Scientific) with a 532 nm laser excitation at a power of 1 mW. The XRD patterns were recorded using a Rigaku D/MAX 2400 diffractometer with a Cu Kα X-ray source operated at 40 kV and 50 mA. The Brunauer-Emmett-Teller (BET) surface area and pore-size distribution (PSD) were evaluated by the nitrogen adsorption (ASAP2020, Micromeritics, USA) at -196^oC. Before the measurement, the samples were degassed at 250 ^oC for 5 h under vacuum. The total pore volume was calculated from the amount of N_2 adsorbed at $P/P_0 \approx 1$. The micropore volumes were calculated using the t-plot method. The pore size distribution (PSD) was calculated by the BJH method using the adsorption isotherm data.

The electrode was fabricated by mixing the as-made *x*G/CX, carbon black and poly(tetrafluoroethylene) in a mass ratio of 85:5:10. The mixture was rolled into a uniform sheet, and then the sheet was punched into discs of 1.0 cm^2 with a weight of *ca.* 4.0 to 5.0 mg. The nickel foam was used as the current collector. The electrochemical test was conducted in a three-electrode system with Hg/HgO reference electrode, Pt foil (30×20 mm^2) as the counter electrode, and the as-made electrode as the working electrode. Cyclic voltammetry (CV), galvanostatic charge/discharge (GC) and electrochemical impedance spectroscopy (EIS) measurements were carried out using a CHI660D workstation (Chenhua Instruments Inc., Shanghai, China) with 6 M KOH as electrolyte. The cycle stability of the as-made composite was evaluated by GC test at a current density of 3 A g^{-1} for 5000 cycles using a Land Battery testing system.

Supplementary data

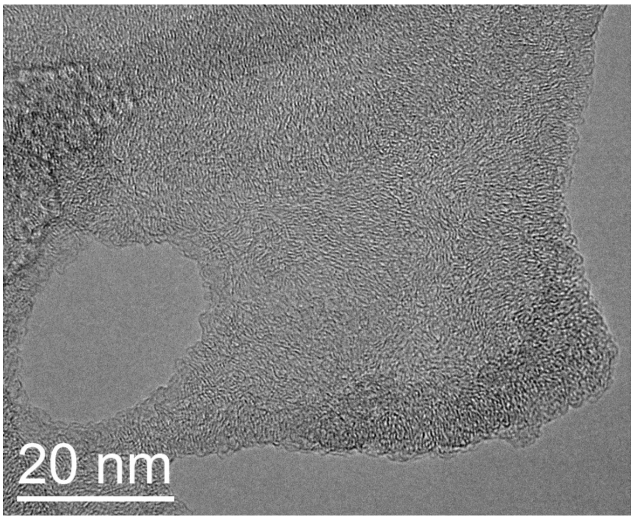

Fig. S1 The TEM image of the graphene used.

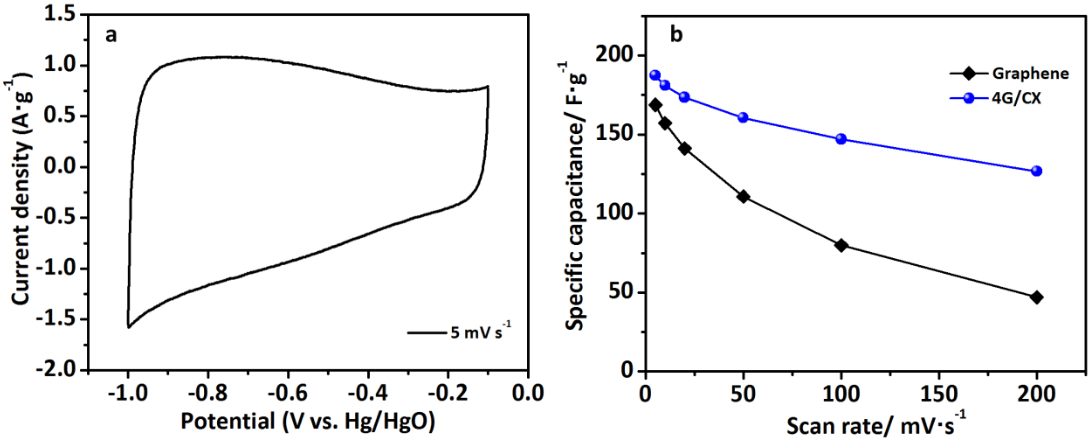

Fig. S2 Electrochemical performance of the graphene and 4G/CX: (a) CV curve of graphene at 5 mV s^{-1}, (b) the specific capacitances of graphene and *4*G/CX vs. the scan rate.

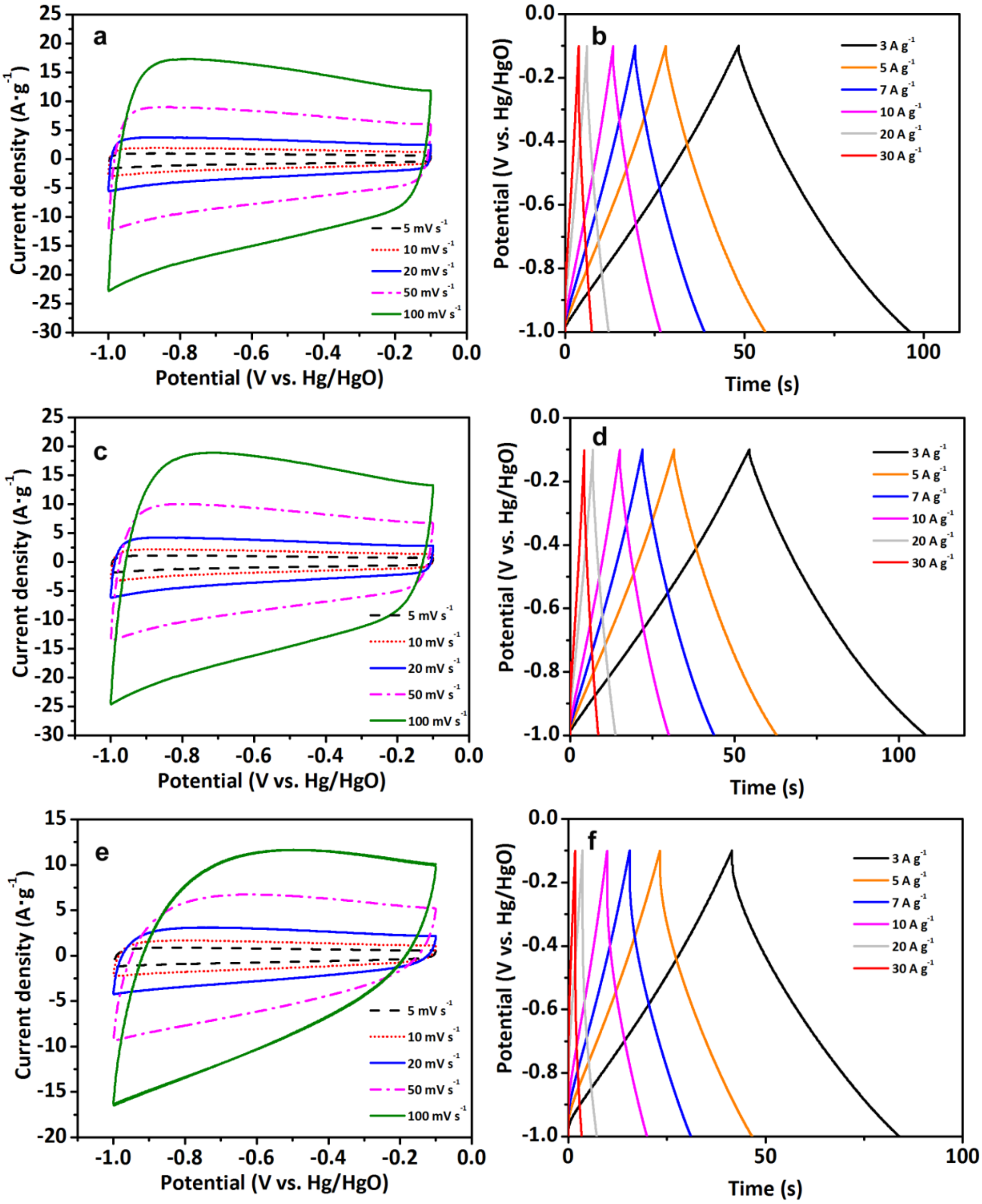

Fig. S3 Electrochemical performance of *1*G/CX (a, b), *4*G/CX (c, d), *10*G/CX (e, f), electrochemical tests were conducted in a three-electrode system with Pt and Hg/HgO as the counter and reference electrodes and 6 M KOH as the electrolyte.

Table S1

Precursor	Method	Surface area/ m^2 g^{-1}	Capacitance/ F g^{-1}	Interfacical capacitance/ □ F cm^{-2}	Testing conditions	Electrolyte	Ref.
Resorcinol-formaldehyde	Ionic liquid template and graphene composite	761	230 192 188 181	30.2	0.1 A g^{-1} (3E)[a] 1 A g^{-1} 5 mV s^{-1} 10 mV s^{-1}	6 M KOH	This work
Resorcinol-formaldehyde	Polyethyleneimine-modified graphene oxdie doping combined with chemical activation	1158	221	18.2	10 mV s^{-1} (3E)	6 M KOH	2
Resorcinol-formaldehyde	Graphene oxide doping, solvent exchange, ambient drying	733	85	11.6[b]	1 A g^{-1} (3E)	6 M KOH	3
Phenol-formaldehyde	Graphene oxide cross-link	378	116	30.7[b]	5 mA (2E)	30 % KOH	4
Resorcinol-formaldehyde	Ambient drying	706	197	27.9[b]	5 mV s^{-1} (3E)	1 M H_2SO_4	5
Resorcinol-formaldehyde	cobalt doping	-	100	-	10 mV s^{-1} (3E)	1 M H_2SO_4	6
Resorcinol-formaldehyde	Inverse emulsion polymerization, solvent exchange, microwave-drying	1670	150	9.0[b]	1 A g^{-1} (2E)	6 M KOH	7
Resorcinol-formaldehyde	Assisted by cationic polyelectrolyte	725	210 110	28.9 14.5	1 mV s^{-1} (3E)	1 M H_2SO_4 1 M KNO_3	8
Resorcinol-formaldehyde	Freeze-drying, chemical modification with ammonia borane	621	About 75	12.1[b]		TEATFB	9
Resorcinol-formaldehyde	Ambient drying	626	183.6	29.3[b]	5 mV s^{-1}	6 M KOH	10

					(3E)		
Resorcinol-formaldehyde	Solvent exchange, ambient drying	-	110.06	-	1 mV s^{-1} (3E)	6 M KOH	11
Resorcinol-formaldehyde	Solvent exchange, ambient drying Post heat-treated at 300°C in air	-	40 35	-	5 mA cm^{-2} (2E)	3 M H_2SO_4 4 M KOH	12
Resorcinol-formaldehyde	Solvent exchange CO_2 activation	1760	23.8[c]	1.4[b]	5 mV s^{-1} (3E)	1 M H_2SO_4	13
Pyrocatechol-formaldehyde	Solvent exchange and CO2 supercritical drying	519	167	32.2[b]	1 mV s^{-1} (3E)	1 M H_2SO_4	14
Reduced graphene oxide (rGO) and single-walled carbon nanotubes (SWCNT)	Mixing and vacuum filtration	-	290.6	-	0.5 A g^{-1} (2E)	1 M KCl	15
SWCNT/rGO	Spray-coating	-	194	-	0.8 A g^{-1} (3E)	1 M KOH	16
GO-SWCNT	Dip casting	-	295	-	0.5 A g^{-1} (2E)	6 M KOH	17
SWCNT/GO	Vacuum filtration	-	171.85	-	5 A g^{-1} (3E)	1 M KOH	18

[a] 3E and 2E stand for a three-electrode system and a two-electrode system;

[b] calculated based on the specific capacitance and specific surface area;

[c] calculated based on the density and volumetric capacitance in the paper.

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