

Supporting Information

High charge efficiency electrode by self-assembling the sulphonated reduced graphene oxide onto carbon fibre: towards an enhanced capacitive deionization

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Experiment

1. Synthesis of graphene oxide (GO)

The GO was prepared according to the modified hummers' method which has been described in our previous works [16,26]. 1 g of natural graphite flakes (SP-1, Bay carbon) was put in to an 353 K mixture of concentrated nitric acid and sulphuric acid (H₂SO₄) with volume of 100 ml separately and then mixed for 4.5 h under constant stirring. After that, the mixture was cooled down to room temperature and followed by diluting with 0.5 L of deionized water (DI) and left overnight. Then the reaction vessel with 120 ml of concentrated H₂SO₄ was immersed in an ice bath, and potassium permanganate powder was added slowly and carefully. Subsequently, the mixture was stirred for 2 h and then diluted with DI water. Further, 20 ml of 30% hydrogen peroxide was added to the mixture and the colour of mixture changed brilliant yellow along with bubbling. Then, the mixture was centrifuged and washed with hydrogen chloride acid (HCl) to remove residual metal ions and followed by rising with DI water for several times until pH almost reached 7, indicating the GO was successfully obtained.

2. Synthesis of sulphonated reduced graphene oxide (SRGO)

The synthesis of SRGO is divided into three steps. At the first step, named as partially reduction of GO, the as-prepared GO slush was added into 200 ml 0.2 M potassium hydroxide offer its pH was adjusted to 9 with sodium carbonate solution. The mixture was then maintained at 353 K for 2 hours under constant stirring. During the reaction, the dispersion turned from dark brown to dark and the aggregation was observed at the end of this step. After centrifuging at 8000 rpm and rinsing with DI water for several times, the partially reduced GO was dispensed in 200 ml DI water accompanied with vigorous stirring. To prepare the aryl diazonium salt for sulphonation, 230 mg sulfanilic acid, 90 mg sodium nitrite, 50 ml DI water and 2 g 1 M HCl were reacted in an ice bath under constant stirring for 2 hours, resulting in the formation of SRGO. Bubbles were expelled from the reaction mixture and aggregation was observed with the adding of diazonium salt solution. After centrifuging and rinsing with DI water for several times, SRGO was redispersed in 200 ml DI water and followed by adding 5 g hydrazine. Then, the reaction was kept at 373 K under constant stirring for 24 hours. After rinsing with water thoughtfully, the SRGO thus obtained

by simply filtered through a vacuum filtration system. Finally, the product was sent to the oven under the argon atmosphere at temperature of 323 K for overnight.

3. Characterization

The morphology and energy dispersive x-ray spectra of as-prepared sample was studied and collected by scanning electron microscopy (SEM, EDX, Philips XL-30). The bulk resistivity of as-prepared samples was tested using four point probes meter (SX 1944, Suzhou Baishen Technology CO., LTD) by filtering the sample on a polytetrafluoroethylene membrane with pore size of 0.25 μm . The concentration towards vacuum filtration was 10 mg/mL. The Fourier transform infrared spectra (FTIR) were recorded on a FT-IR spectrometer (PerkinElmer spectrum 100) by directly pressing the sample on sample holder. The pore size distribution and Brunauer-Emmet-Teller (BET) specific surface area were deduced from the N_2 physical adsorption measurement data which were obtained using ASAP 2010 Accelerated Surface Area and Porosimetry System (Micrometitics, Norcross, GA).

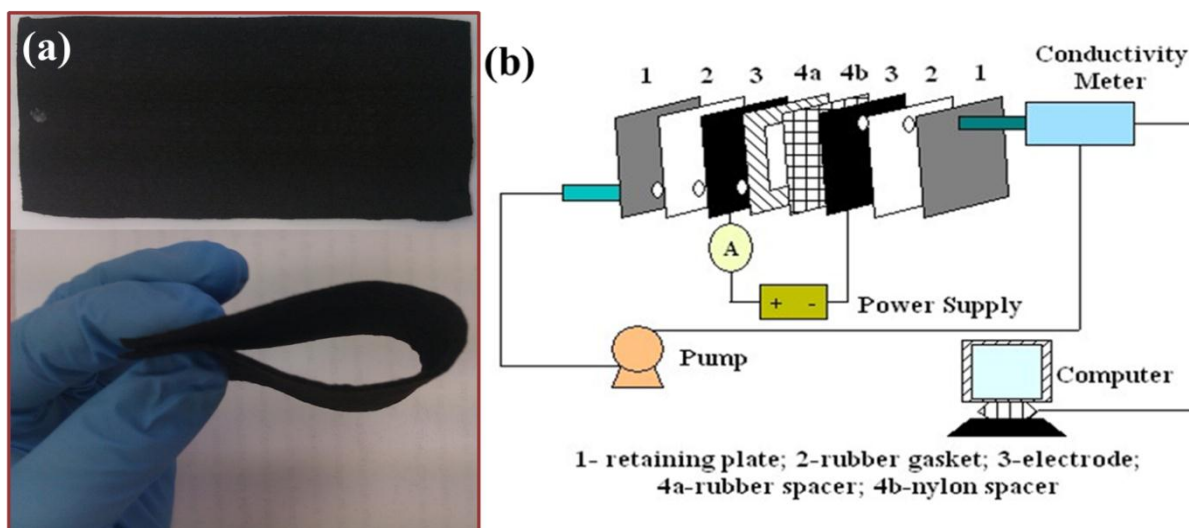


Figure S1. (a) the digital image of CFC-SRGO film, (b) CDI experimental setup.

Results and Discussion

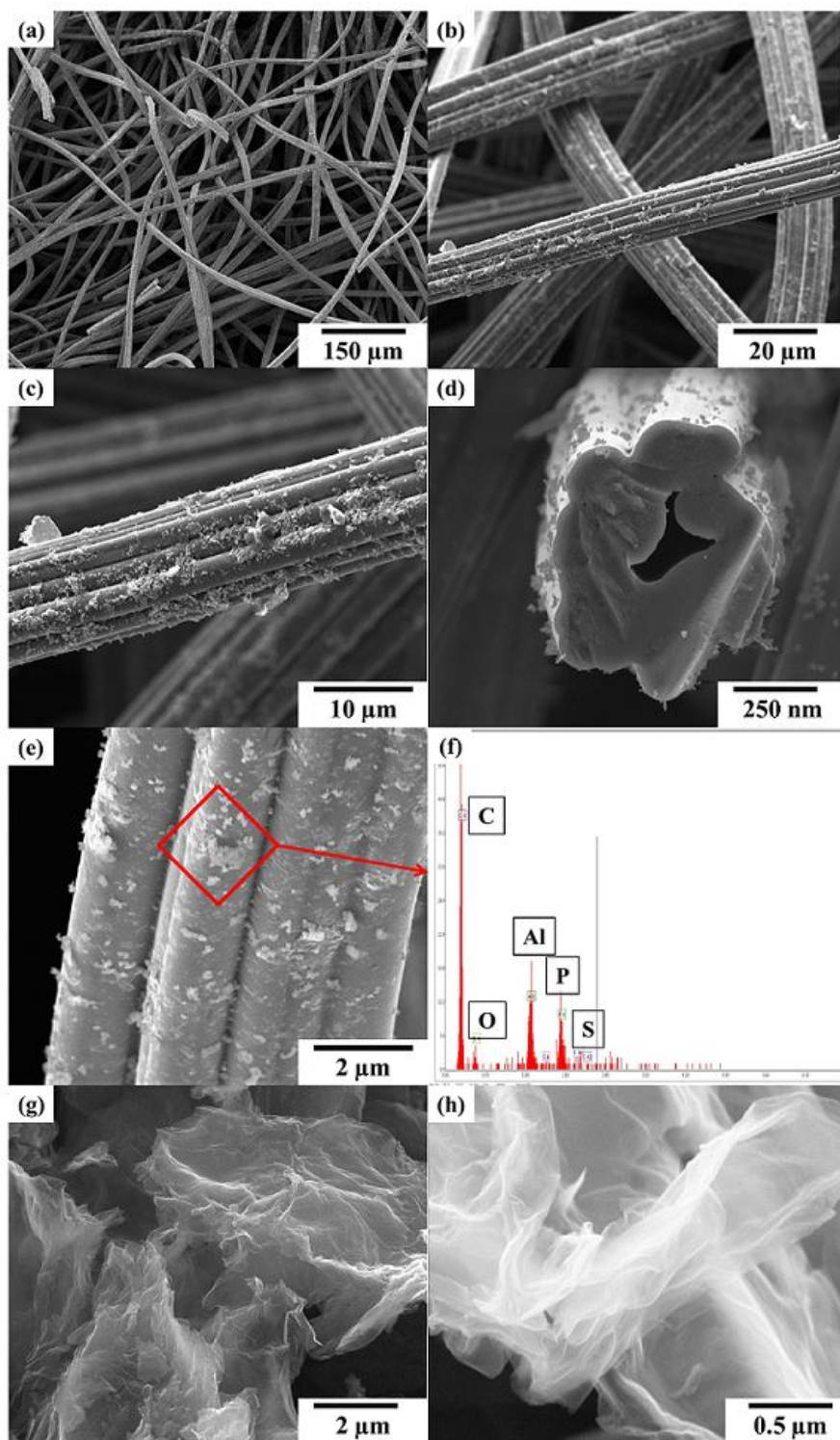


Figure S2. (a), (b), (c), (d) and (e) SEM image of CFC-SRGO composite with different magnifications, (f) EDX spectra of selected area indicated in (d), (g) and (h) SEM image of SRGO in low and high magnifications

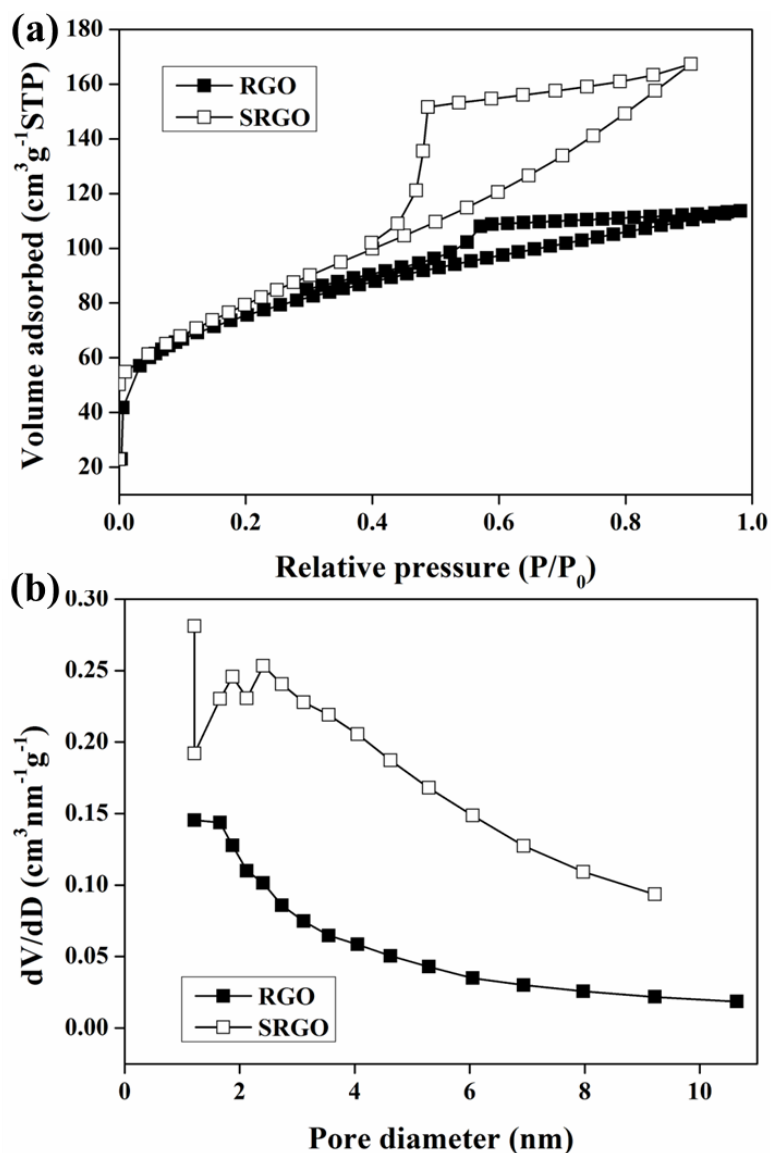


Figure S3. Nitrogen adsorption-desorption isotherms and (b) pore size distribution of RGO and SRGO.

Table S1 The texture properties of RGO and SRGO

Sample	BET surface area (m ² /g)	Pore volume (cm ³ /g)	Micropore volume (cm ³ (STP)/g)	Average pore diameter (nm)
RGO	254	0.17	58.32	2.76
SRGO	439	0.28	99.61	2.51

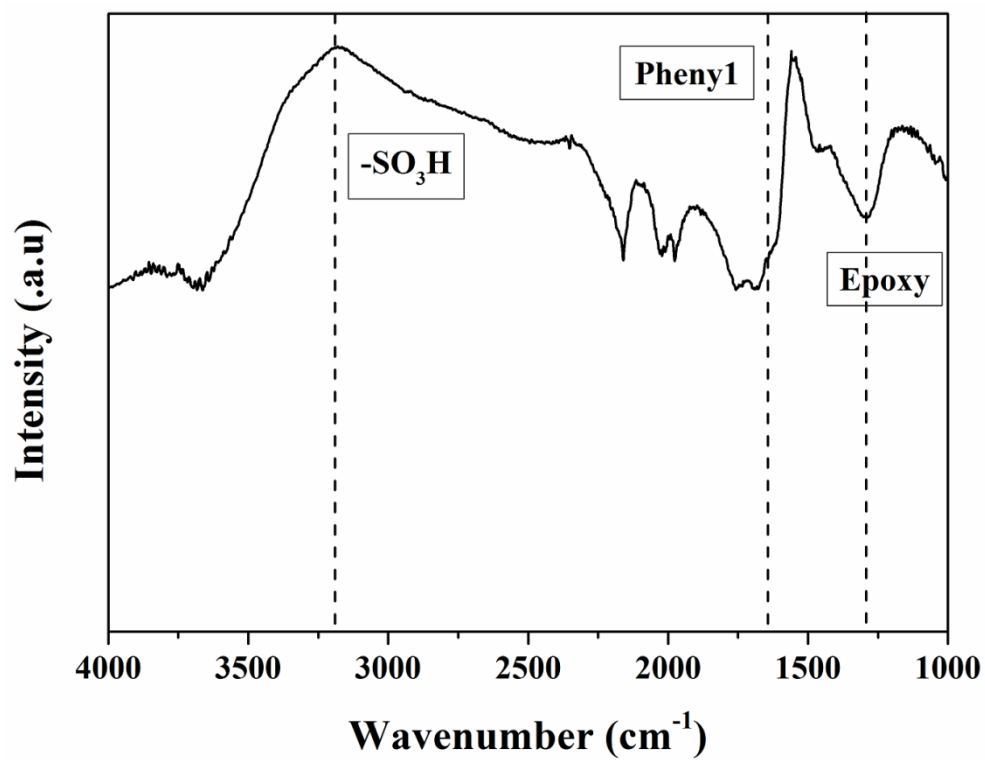


Figure S4. FTIR spectra of SRGO.

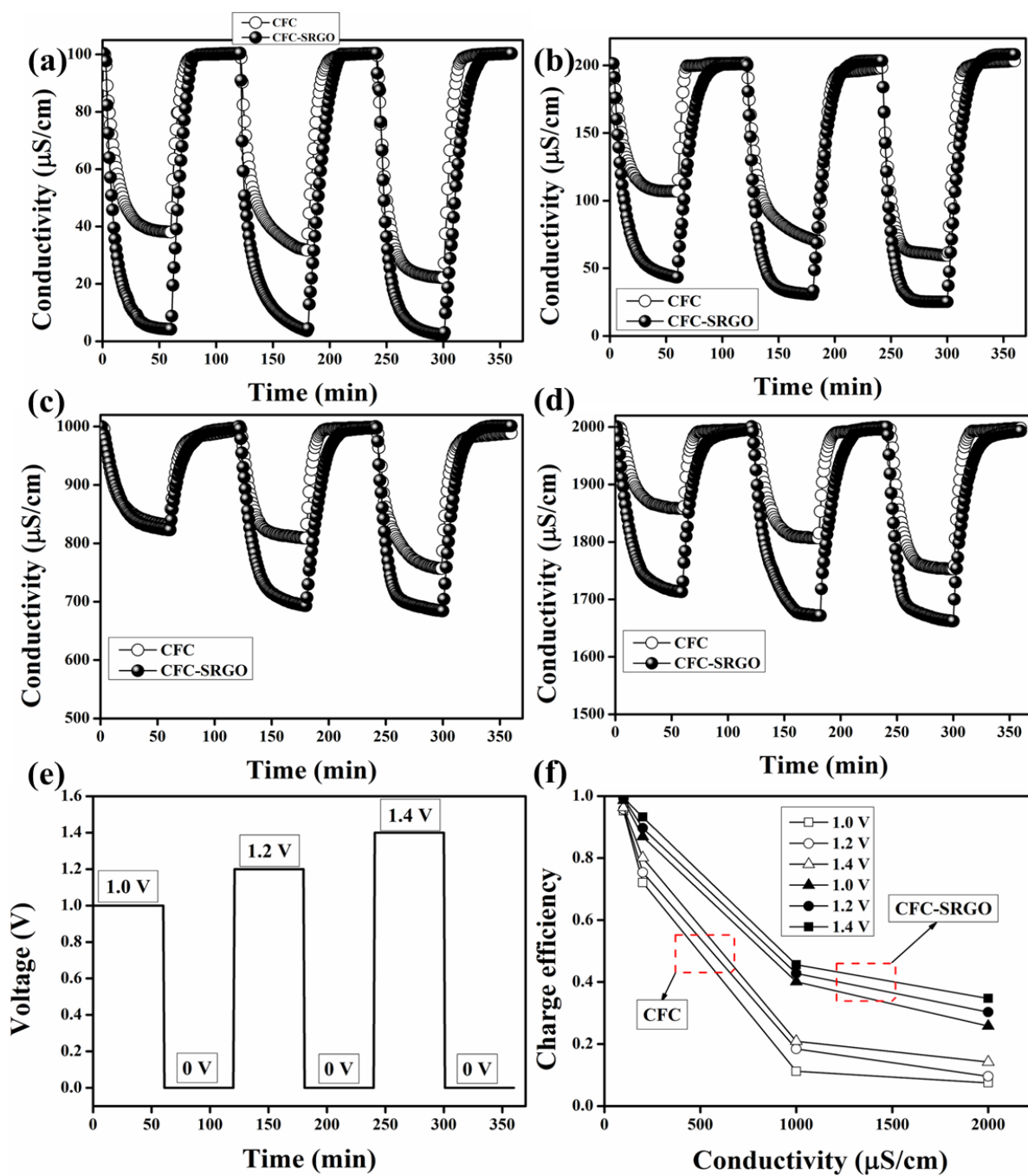


Figure S5. The electrodesorption performance of CFC and CFC-SRGO electrode at the cell voltage of 1.0, 1.2 and 1.4 V in NaCl solution with an initial conductivity of (a) 100, (b) 200, (c) 1000 and (d) 2000 $\mu\text{S/cm}$, respectively; (e) the cell voltage corresponds to electrodesorption-desorption cycles; (f) the charge efficiency dependence of CFC and CFC-SRGO electrodes on cell voltage and solution conductivity.

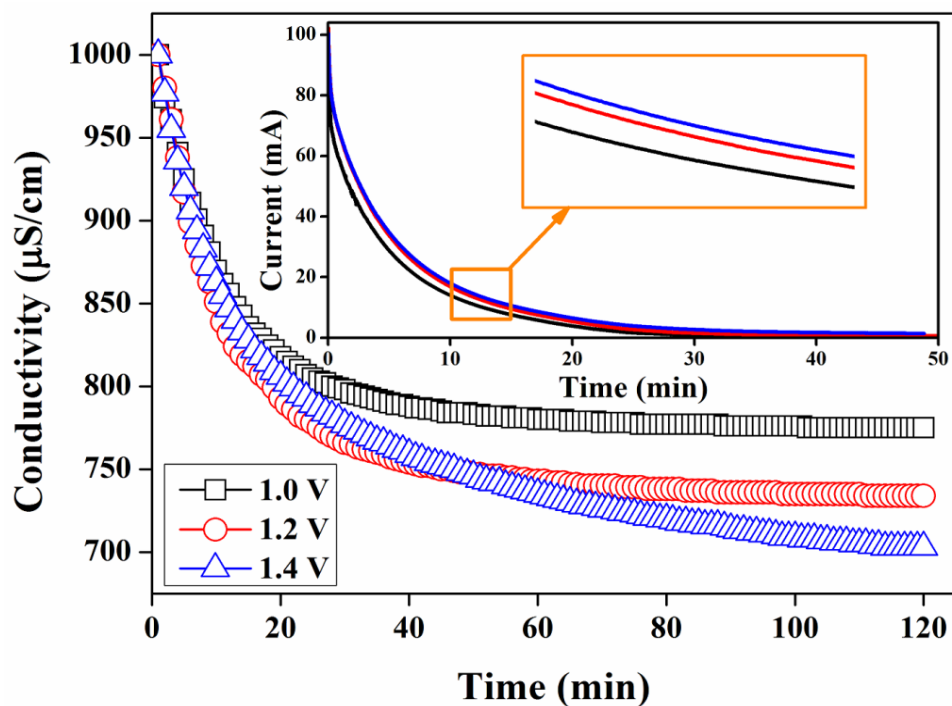


Figure S6. Electrosorption performance of CFC based MCDI at different cell voltage in NaCl solution with an initial conductivity of 1000 $\mu\text{S/cm}$, the inset shows corresponding current response.

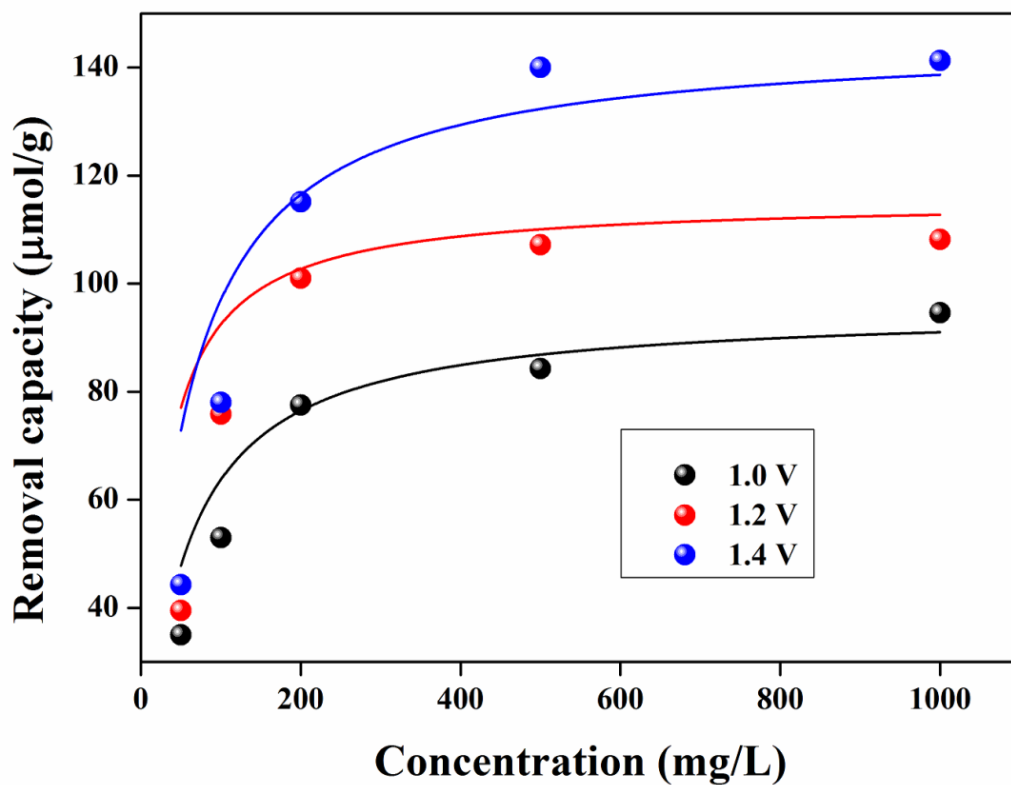


Figure S7. The electrosorption isotherm of CFC electrode at cell voltage of 1.0, 1.2 and 1.4 V, respectively.

Table S2 Parameters determined from various isotherms of CFC electrode.

Voltage (V)	Isotherm	q_m ($\mu\text{mol/g}$)	K_L	r_L^2	Isotherm	K_F	n	r_F^2
1.0		95.57	0.0198	0.95		15.82	3.76	0.84
1.2	Langmuir	155.55	0.0382	0.93	Freundlich	24.83	4.41	0.63
1.4		145.23	0.0201	0.96		20.46	3.41	0.79

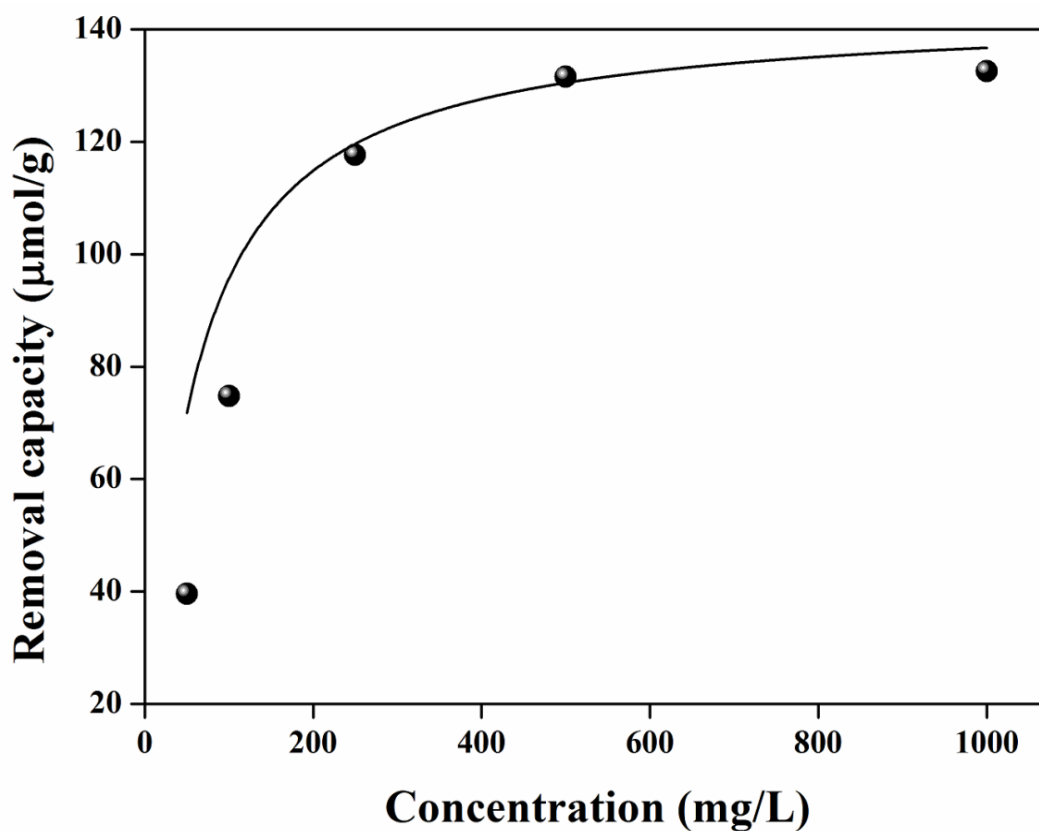


Figure S8. The electrosorption isotherm of CFC-SRGO electrode at temperature of 273 K, the cell voltage is 1.2 V.