

Supporting Information

Electrodeposition of High-Capacitance 3D CoS/Graphene Nanosheets on Nickel

Foam for High-Performance Aqueous Asymmetric Supercapacitor

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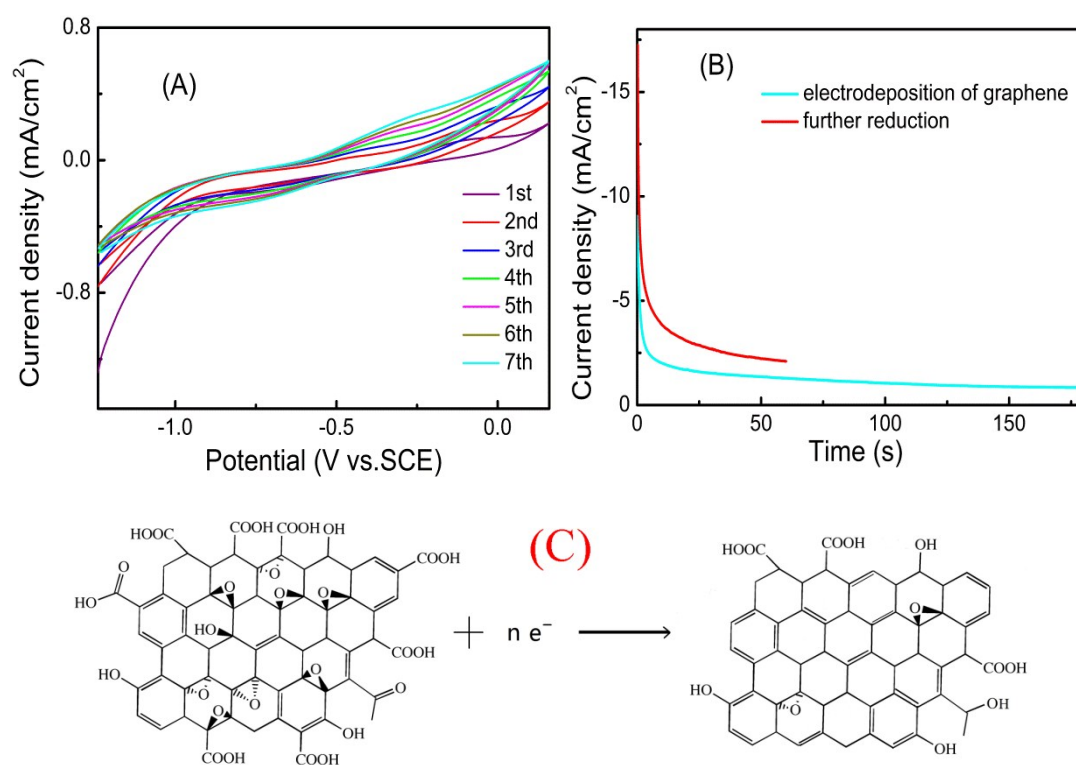


Figure S1 (A) The nanostructured cobalt sulfide film was deposited by cyclic voltammetric method in a potential interval from -1.24 to 0.16 V vs. SCE with a scan rate of 5 mV s⁻¹ for seven cycles; (B) deposition of electrochemically reduced graphene (eRG) was carried out by a potentiostatic method under a constant potential of -1.2 V vs. SCE for 180 s. After deposition, the electrode was rinsed with deionized water for several times and then immersed in deionized water for 1 h. Subsequently, the electrode was further reduced at -1.2 V vs. SCE for another 60 s in 1M LiClO₄ solution; (C) schematically illustration of electrochemically reduced graphene oxide

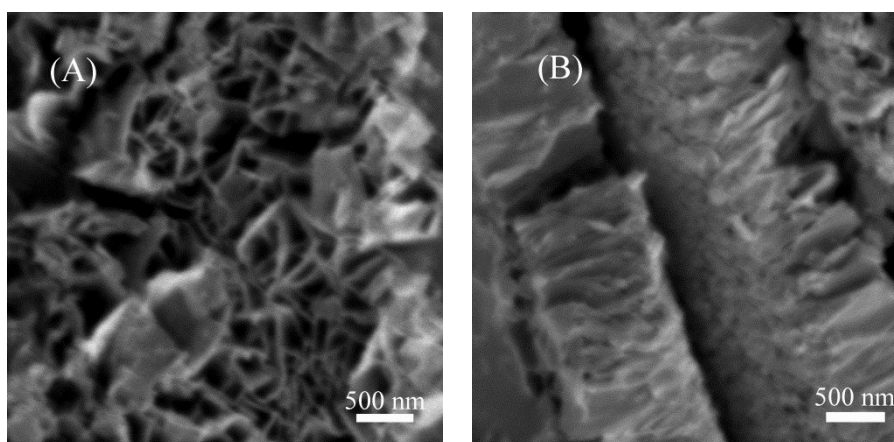


Figure S2 FE-SEM images of CoS@eRG film scratched from Ni foil through a typical synthetic method, surface (A) and cross section (B).

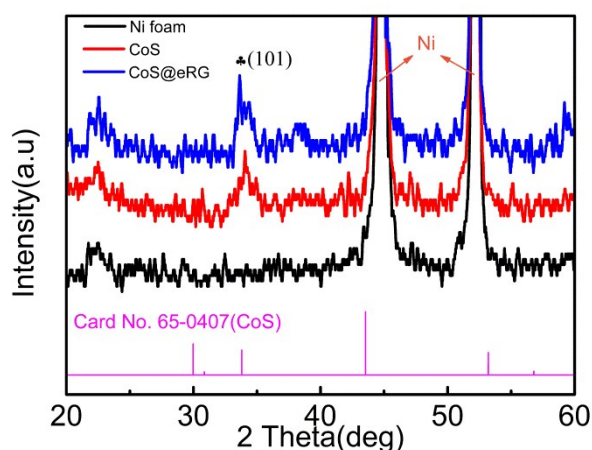


Figure S3 XRD patterns of Ni foam, CoS deposited on Ni foam and CoS@eRG composites deposited on Ni foam

The XRD patterns of bare Ni foam (black), CoS deposited on Ni foam (red) and CoS@eRG composites deposited on Ni foam (blue) are shown in Figure S3. Compared with the Ni foam, a clear diffraction peak appears at around 34° in both CoS and CoS@eRG XRD patterns, corresponding to the (101) diffraction peaks of hexagonal CoS (PDF 65–0407). The other peaks of CoS are not obvious, possibly because the highly preferred orientation along (101) plane.

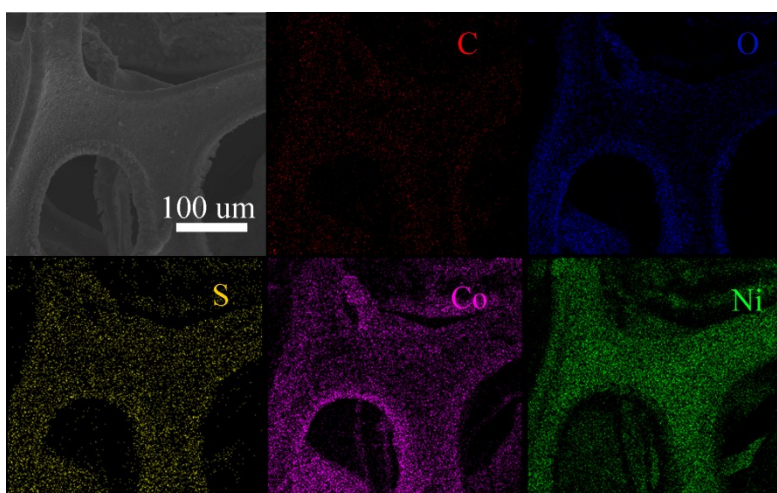


Figure S4 FE-SEM image of CoS@eRG film electrodeposited on Ni foam combined with EDS mapping of in the same area.

The result of EDS mapping in Figure S4 shows the presence of C, O, S, Co and Ni, in which Ni arises from Ni foam substrate and O arises from oxygen containing groups of graphene. All the element mappings in Figure S4 show continuous and uniform element distribution throughout the Ni foam surface, illustrating that the as-prepared Ni foam is covered with uniform-distributed CoS@eRG film.

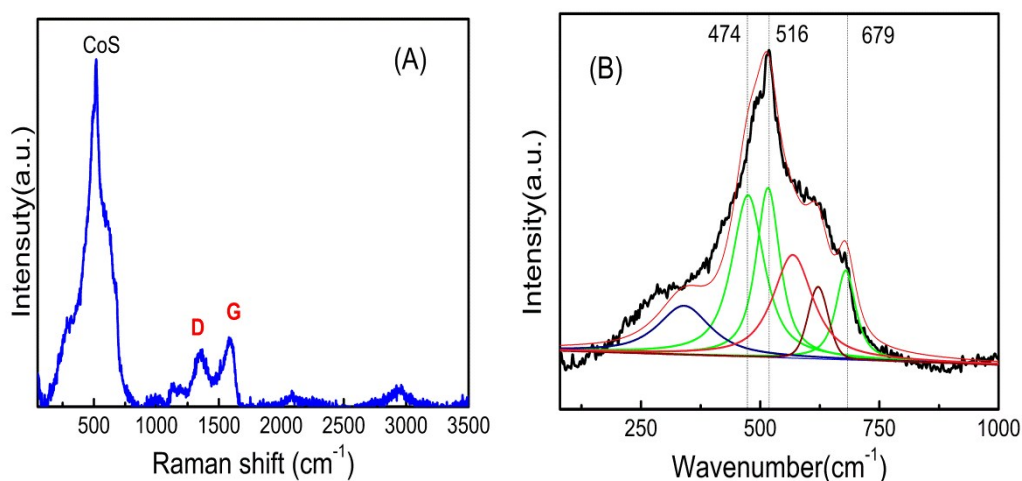


Figure S5 (A) Raman spectrum for CoS@eRG; (B) curve fitting for the CoS peak.

Generally, the Raman peak area is quantitatively proportional to the mass ratio of the particular species.^[S1] So the proportion of CoS/graphene in the nanocomposite can be revealed by the peak area ratio of CoS/(D+G). The value of CoS to graphene is

roughly calculated to be 4.5 to 1.0 from Figure S5 A. Since cobalt sulfide is a non-stoichiometric compound, the main peak of CoS could be curve fitted by several small peaks which are ascribed to the inconsequential amount of Co_xS_y ($x=1, 2\ldots 4$; $y=1, 2\ldots 9$). As shown in Figure S5 B, the main peak was curve fitted by six small peaks, in which three green peaks appeared at 474, 516 and 679 cm^{-1} representing the formation of CoS phase.

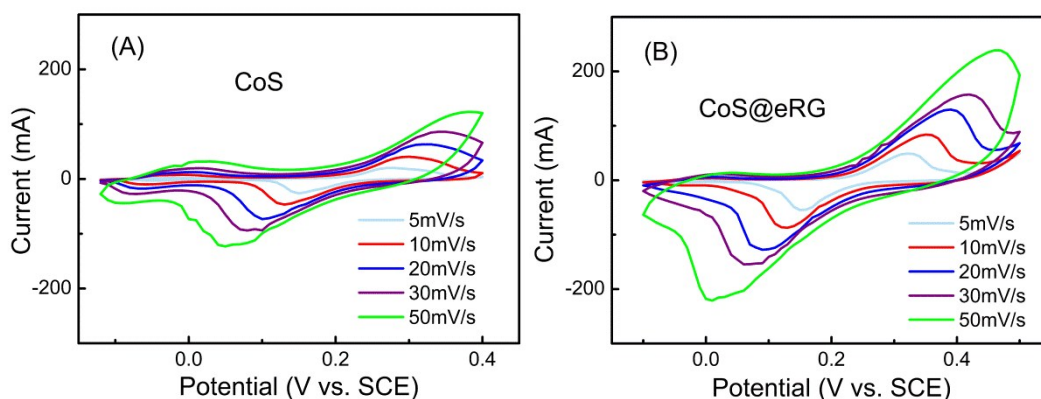


Figure S6 CV curves at various scan rates used for Randles-Sevcik equation. (A) CoS electrode, (B) CoS@eRG electrode

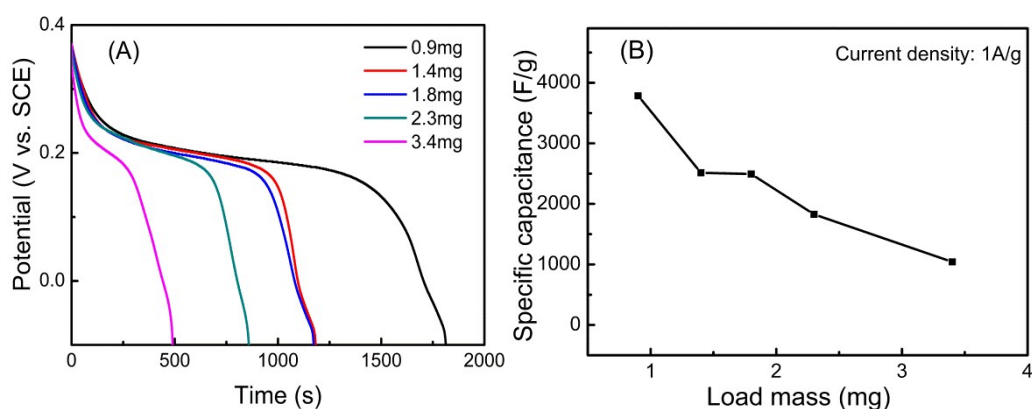


Figure S7 (A) galvanostatic discharge voltage profiles of different deposited mass of CoS@eRG at 1 A g^{-1} , (B) specific capacitance of different deposited mass of CoS@eRG at 1 A g^{-1} .

Electrodes with different mass of CoS@eRG were fabricated by controlling cyclic voltammetry scan cycles (N), and potentiostatic reduction time (t). The 0.9mg

electrode was obtained with 7 CV scans ($N=7$) and a reduction time of 120 s ($t=120$) in 1 mg ml^{-1} GO solution, 1.4mg with 10 CV scans and 180 s in 1 mg ml^{-1} GO solution, 1.8 mg with 14 CV scans and 240 s in 1 mg ml^{-1} GO solution, 2.3mg with 20 CV scans and 340 s in 1 mg ml^{-1} GO solution, 3.4mg with 30 CV scans and 510 s in 1 mg ml^{-1} GO solution.

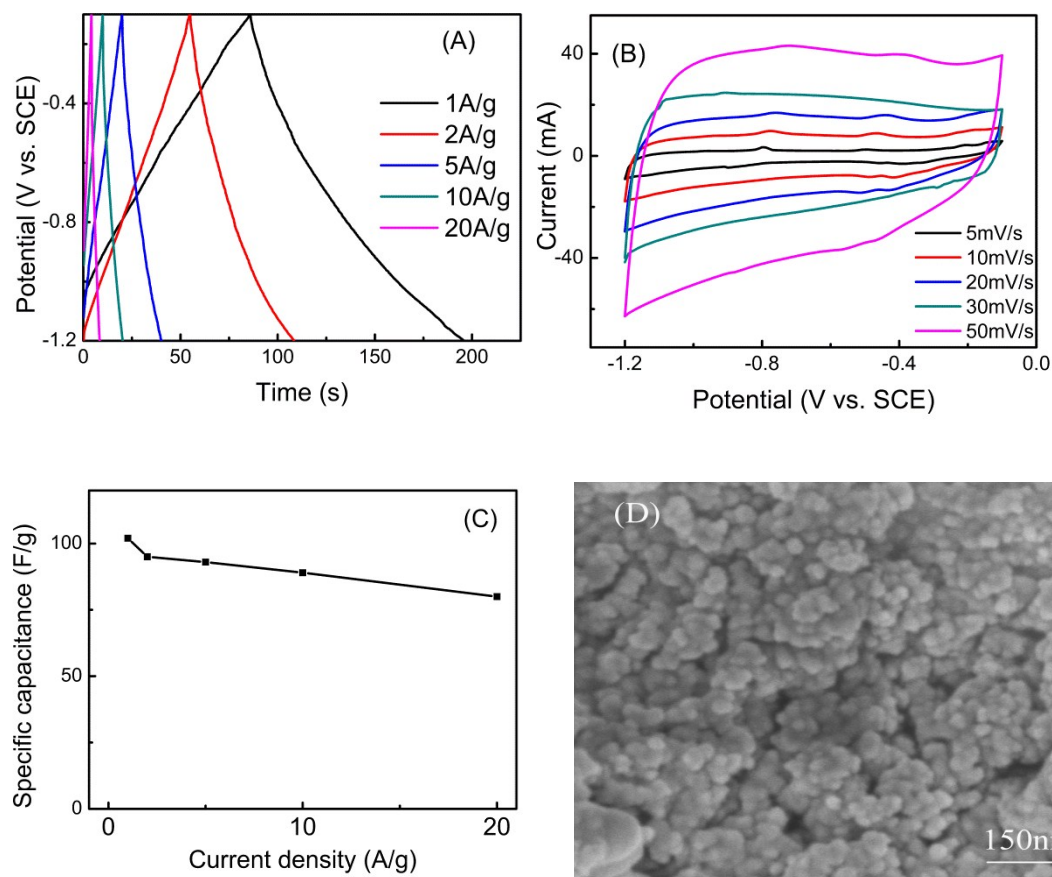


Figure S8 (A) galvanostatic charge-discharge voltage profiles of activated carbon (AC) electrode, (B) CV curves of the activated carbon electrode, (C) specific capacitance of the activated carbon electrode, (D) FE-SEM image of AC

A series of AC electrodes are made with loading mass varied from 5-15mg. When the total mass of negative electrode is 11.3 mg, the total mass of positive electrode is 0.9 mg, the asymmetric capacitor was applied with a current density of 1 A g^{-1} , the negative electrode's current density was $\sim 1 \text{ A g}^{-1}$ and the positive electrode's current density was $\sim 13.5 \text{ A g}^{-1}$. From Figure 5 B shown in the article, the CoS@eRG electrode exhibits a capacitance of $\sim 2800 \text{ F g}^{-1}$ at 13.5 A g^{-1} . And the activate carbon

electrode exhibits a capacitance of $\sim 102 \text{ F g}^{-1}$ at 1 A g^{-1} .

To ensure the charge balance between the positive and negative electrodes the following equation should be satisfied.

$$q^+ = q^-$$

$$C^+ \Delta E^+ m^+ = C^- \Delta E^- m^-$$

$$2800 \times 0.5 \times m^+ = 102 \times 1.1 \times m^-$$

$$m^+ / m^- = 0.08$$

$$\text{And } \frac{0.9}{11.3} = 0.08, \text{ so the mass of negative electrode should be } 11.3 \text{ mg.}$$

Table 1 power density and energy density for the different asymmetric supercapacitors

	Positive	Negative	Energy density (Wh kg ⁻¹)	Power density	electrolyte	voltage
Ref. S2	Co9S8	AC	31.4	200 W kg ⁻¹	30wt% KOH	1.6 V
Ref. S3	Ni(OH)2/UGF	a-MEGO	13.4	85.0kWkg ⁻¹	6 M KOH	1.8 V
Ref. S4	RGO–RuO2	RGO–PANi	26.3	250 W Kg ⁻¹	Not given	1.4 V
Ref. S5	RuO2–ILCMG	IL-CMG	19.7	6.8 kW g ⁻¹	PVA–H2SO4	1.8 V
Ref. S6	NiCo2O4	AC	17.72	25kW kg ⁻¹	1 M NaOH	1.7 V

Reference

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