

Calculation

The specific capacitance can be calculated from the CV curve using the following equations

$$C = \frac{\int ivdv}{2\mu m\Delta V} \quad (1)$$

$$C = \frac{It}{m\Delta V} \quad (2)$$

where i and v are the current and potential in the CV test (A and V), μ is the scan rate (V/s), m is the mass of active materials (g), ΔV is the potential window of discharge (0.5V here), I is the constant discharge current and t is the discharge time (s).

The specific capacitance, power density and energy density are calculated based on the galvanic charging-discharging curves using the equation as follows:

$$C = \frac{I\Delta t}{m\Delta V} \quad (3)$$

3)

$$Eg = \frac{1}{2} \times C \times \Delta V^2 \quad (4)$$

$$Pg = \frac{E}{\Delta t} \quad (5)$$

(5)

where C (Fg^{-1}) is specific capacitance, E ($Whkg^{-1}$) is energy density, P (Wkg^{-1}) is power density, ΔV is potential window (here 1.6V), I (A) is discharge current, Δt (s) is discharge time, m (g) is the sum of the masses of the positive electrode (Ni-Fe-O/r-NF, here 3mg) and negative electrode (reduced graphene oxide, here 11mg), thus here m is 14mg. When demonstrating the data, the current density is based on two electrodes, and each of them has the dimension of $2 \times 1 \times 0.05$ cm. That is to say, the $2mAcm^{-2}$ represents 8mA ($2mA \times 2$ pieces of $2cm^2$ nickel foam).

The volumetric energy density could be estimated by

$E_v = E_g \cdot m / (2 \times 2cm^2 \times 0.05cm)$ (The volume of two pieces of nickel foam as negative electrode and positive electrode)

The thickness of the rolled nickel foam is 0.05cm while the thickness of unrolled nickel foam is 0.15cm.

$P_v = P_g \cdot m / (2 \times 2cm^2 \times 0.05cm)$ (The volume of two pieces of nickel foam as negative electrode and positive electrode)

The theoretical capacitance of the asymmetric full cell is

$$\frac{1}{C_T} = \frac{1}{C_P} + \frac{1}{C_N} \quad (6)$$

where C_T is the total capacitance of the cell, C_P is the capacitance of the positive electrode (nickel oxide or nickel iron oxide, here 1600 F/g or 600 F/g as seen in Fig.7), C_N is the capacitance of the negative electrode (reduced graphene oxide, here 140F/g)

The formula is based on the book Conway, B. E. *Electrochemical supercapacitors: Scientific fundamentals and technological applications*; Kluwer Academic/Plenum Publishers:

New York, 1997. and some journals such as Liang Huang, Dongchang Chen, Yong Ding, Shi Feng, Zhong Lin Wang, and Meilin Liu *Nano Lett.* 2013, 13, 3135–3139 M. D. Stoller and R. S. Ruoff, *Energy Environ. Sci.*, 2010, 3, 1294

D. Hulicova-Jurcakova, A. M. Puziy, O. I. Poddubnaya, F. Suarez- Garcia, J. M. D. Tascon and G. Q. Lu, *J. Am. Chem. Soc.*, 2009, 131, 5026

Supplementary Figures

Table S1 BET Surface Area of Ni-Fe-O grown on rolled nickel foam with different feeding ratio of Ni/Fe

Different ratios	Ni:Fe=1:0 /r-NF	Ni:Fe=0.8:0.2 /r-NF	Ni:Fe=0.6:0.4 /r-NF	Ni:Fe=0.4:0.6 /r-NF	Ni:Fe=0.2:0.8 /r-NF	Ni:Fe=0:1 /r-NF
BET surface area(m ² g ⁻¹)	5.67	1.34	0.39	0.32	1.10	0.116

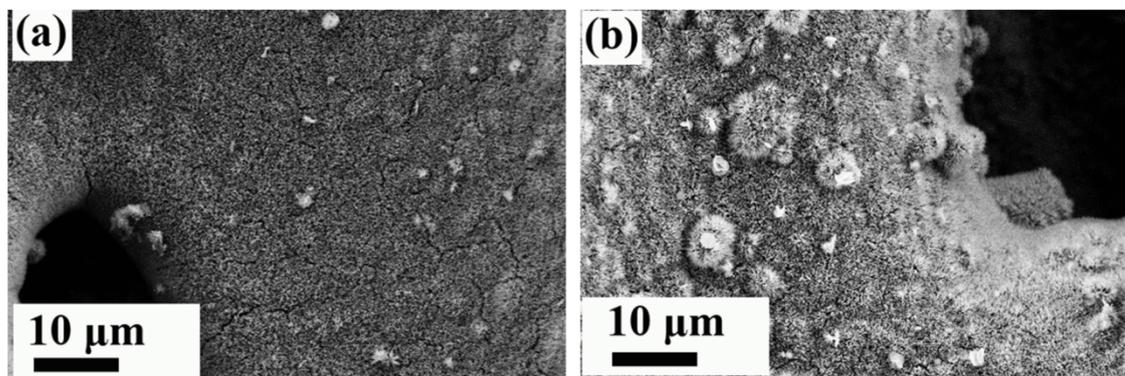


Figure S1 Typical SEM images of Ni-Fe-O grown on (a) unrolled nickel foam and (b) rolled nickel foam

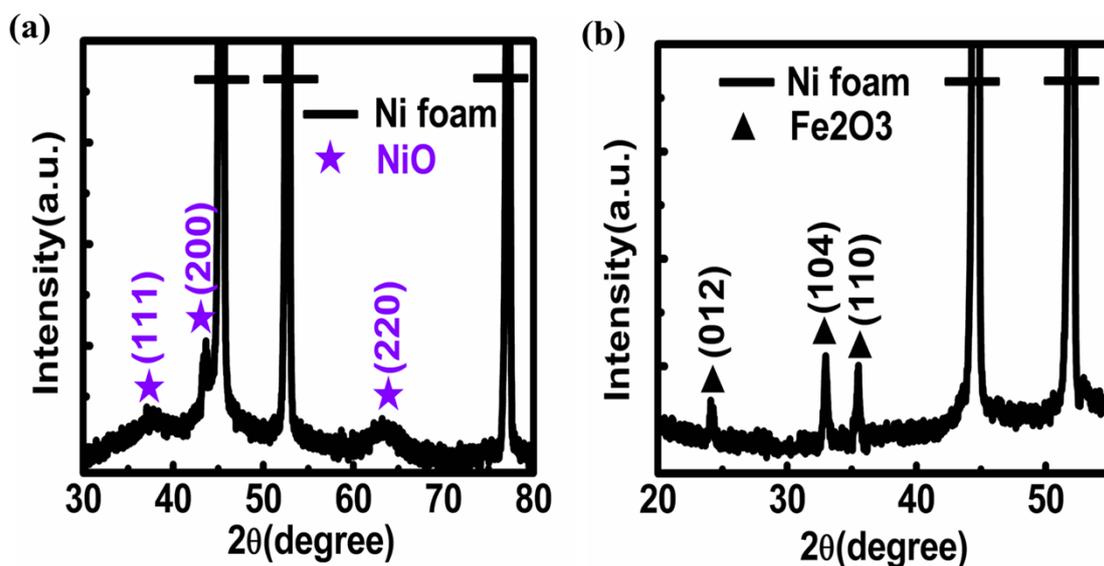


Figure S2 XRD profiles of (a) pure nickel oxide (Ni: Fe=1:0) (b) pure iron oxide (Ni:Fe=0:1)

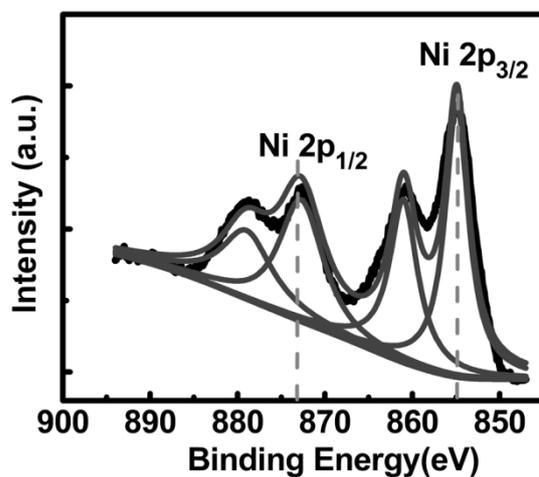


Figure S3 Ni 2p XPS and (d) Fe 2p XPS spectra of hybrid nanosheets supported on r-NF (the feeding mole ratio of Ni:Fe=0.8:0.2)

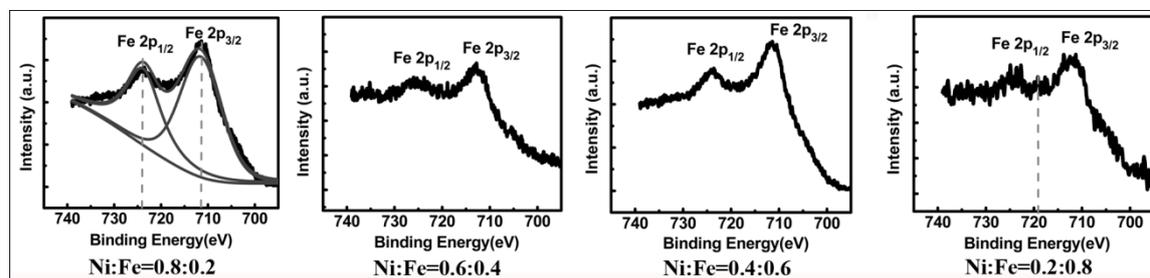


Figure S4 Fe 2p XPS spectra of hybrid nanosheets supported on r-NF (Ni:Fe different feeding mole ratio)

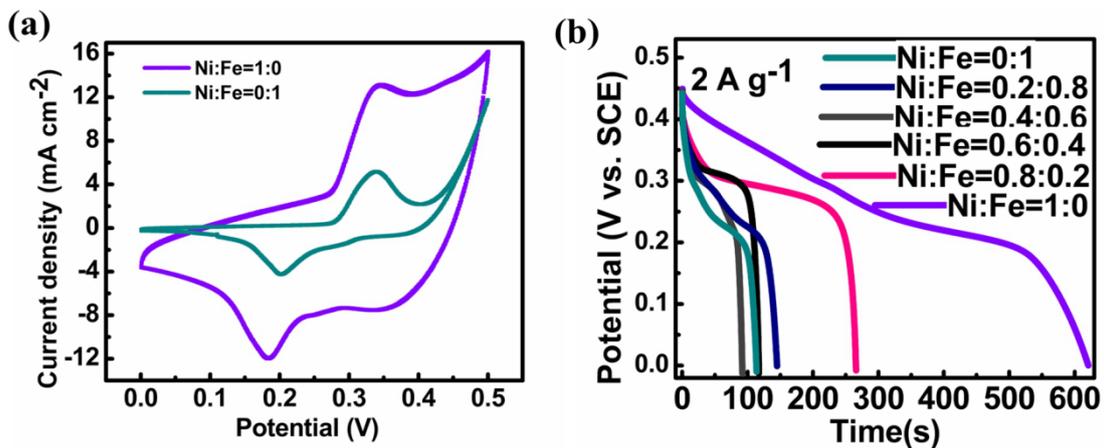


Figure S5 (a) CV curves of samples tested in 1 M KOH at a scan rate of 5 mV s⁻¹ (feeding mole ratio 1:0 and 0:1). (b) Galvanostatic charge-discharge curves of Ni-Fe hybrid electrodes with different Ni:Fe feeding mole ratios

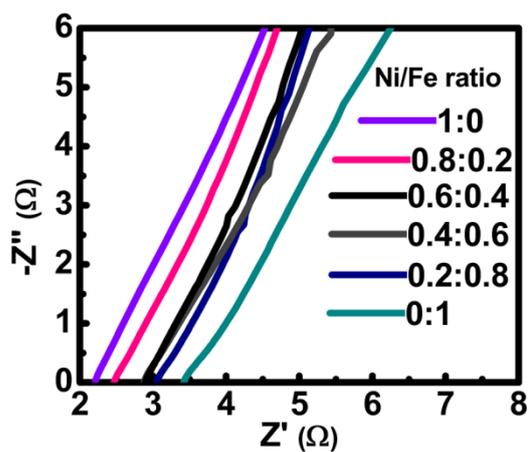


Figure S6 Comparison of Nyquist plots with different Ni:Fe feeding mole ratios