Electronic Supplementary Material (ESI) for Energy & Environmental Science

[Supporting Information]

One-step Synthesis of Ni_3S_2 Nanorod@$Ni(OH)_2$ Nanosheet Core-Shell Nanostructures on Three-dimensional Graphene Network for High-Performance Supercapacitors

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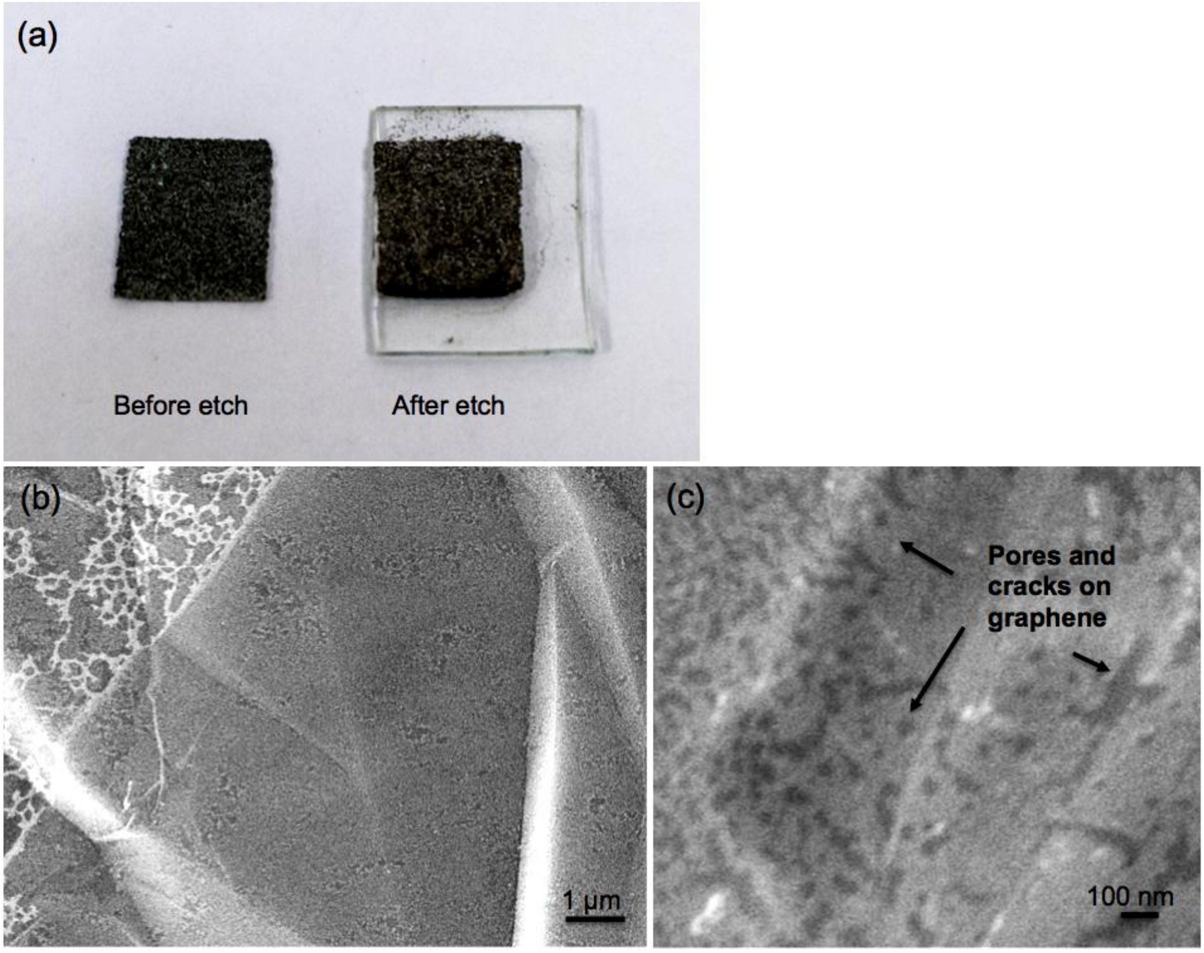

Figure S1. (a) Photos of (left) Ni_3S_2@$Ni(OH)_2$/3DGN and (right) the 3DGN placed on glass after the removal of Ni_3S_2@$Ni(OH)_2$ and Ni foam. (b, c) SEM images with different magnifications of the surface of 3DGN after complete removal of Ni_2S_3@$Ni(OH)_2$ and Ni foam in Ni_3S_2@$Ni(OH)_2$/3DGN.

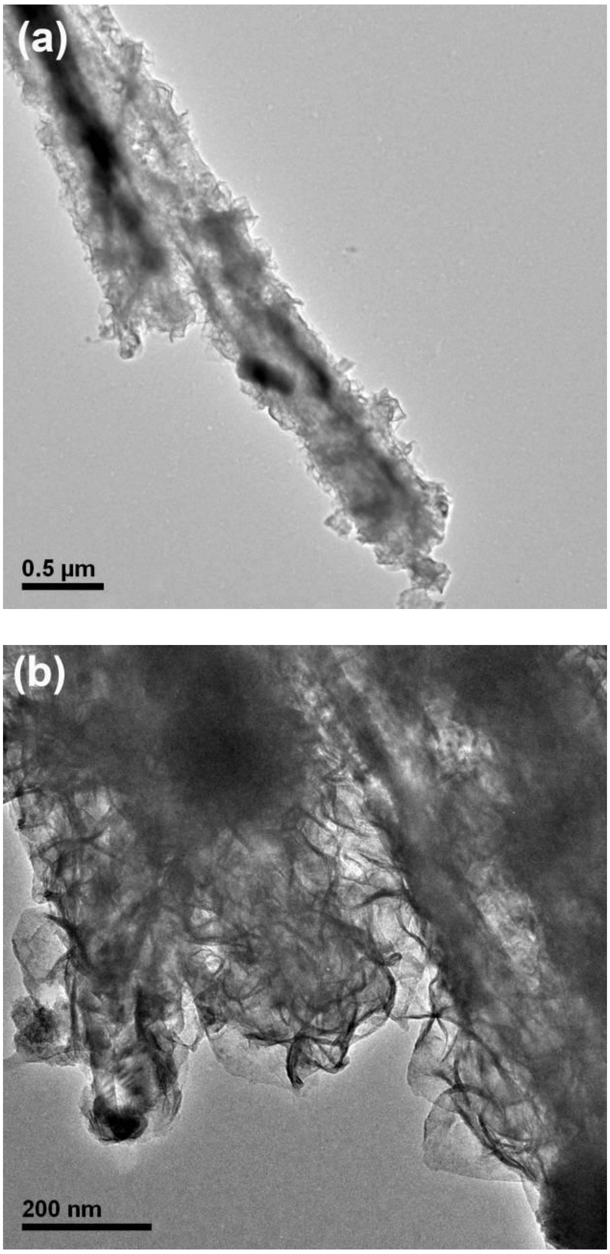

Figure S2. TEM images of the $Ni(OH)_2$ nanosheets obtained by the hydrothermal reaction at 180 ^{o}C for 24 h.

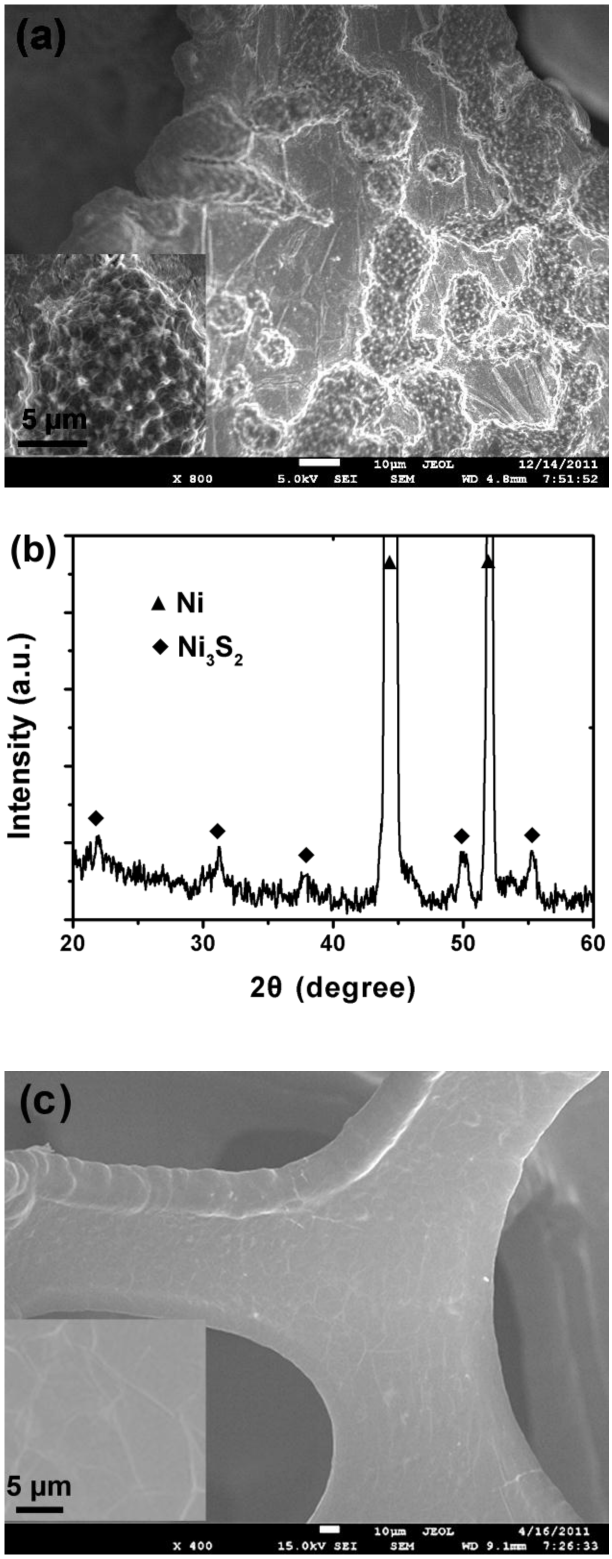

Figure S3. (a) SEM image and (b) XRD result of 3DGN with hydrothermal treatment at 180 °C for 2 h. (c) SEM images of untreated 3DGN. Inset in (a) and (c): The corresponding magnified SEM images.

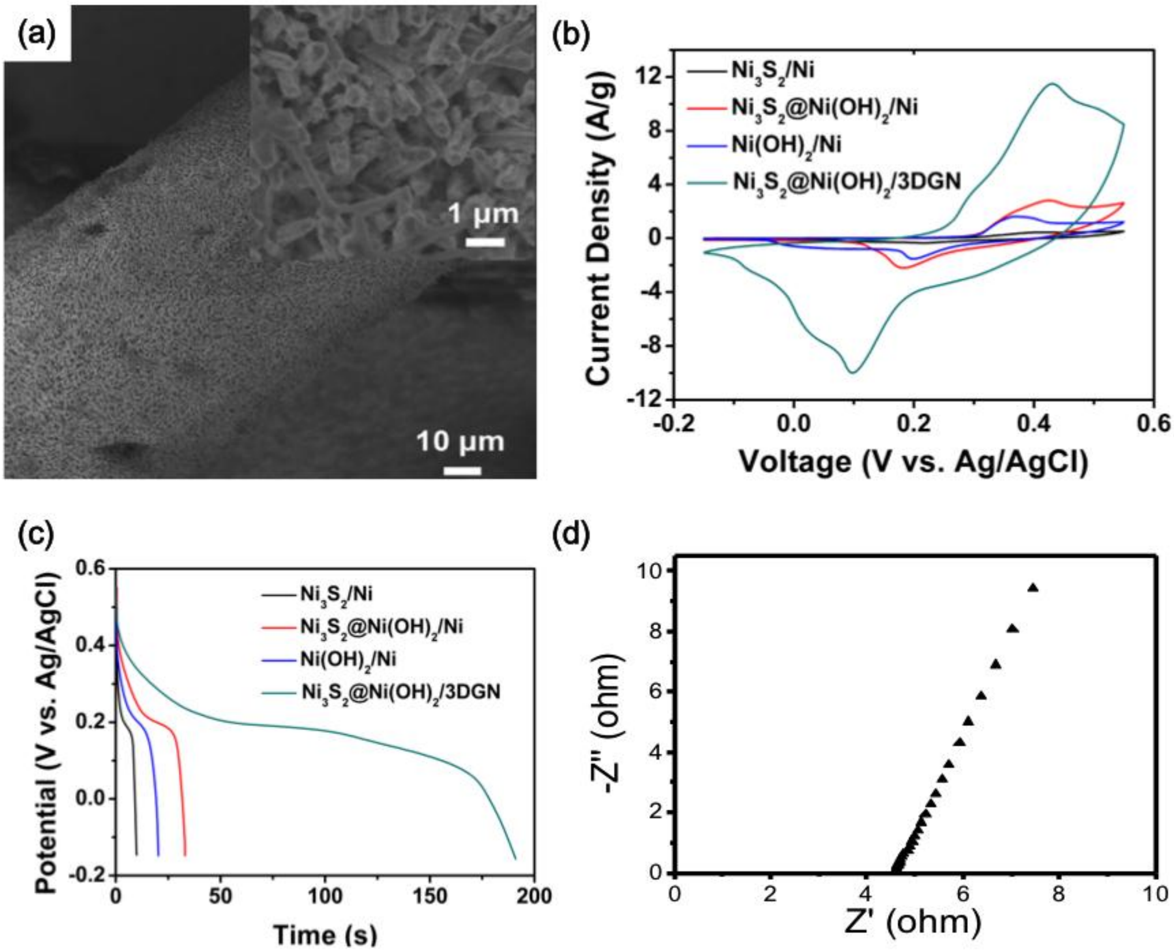

Figure S4. (a) SEM images of Ni_3S_2@$Ni(OH)_2$/Ni obtained by hydrothermal treatment at 180 ^{o}C for 12 h. Inset: High-magnification SEM image of Ni_3S_2@$Ni(OH)_2$/Ni. Comparison of Ni_3S_2/Ni, Ni_3S_2@$Ni(OH)_2$/Ni, $Ni(OH)_2$/Ni and Ni_3S_2@$Ni(OH)_2$/3DGN electrodes: (b) cyclic voltammograms at scan rate of 5 mV/s, and (c) discharge curves at the current density of 5.1 A/g. (d) Nyquist plots of Ni_3S_2@$Ni(OH)_2$/Ni.

The specific capacitance of Ni_3S_2/Ni, Ni_3S_2@$Ni(OH)_2$/Ni and $Ni(OH)_2$/Ni, calculated from CV curves at 5 mV/s is 183, 246 and 231 F/g, respectively (Figure S4b). The galvanostatic discharge curves in Figure S4c show specific capacitances of 72, 241 and 147.8 F/g at the current density of 5.1 A/g for Ni_3S_2/Ni, Ni_3S_2@$Ni(OH)_2$/Ni and $Ni(OH)_2$/Ni, respectively.

Calculations:

Specific capacitance was calculated from CV and charge-discharge curves by the equations (1) and (2), respectively, where I1 is the response current, △V is the voltage window, ν is the scan rate, I2 is the constant discharge current, △t is the discharging time and m is the total mass of the active materials. $m_{total} = m_{graphene}$ (~0.2 mg/cm^2) + $m_{composite}$. Because the Ni_3S_2@$Ni(OH)_2$/3DGN and Ni_3S_2@$Ni(OH)_2$/Ni were synthesized in one step, at moment it is difficult to define the exact mass ratio of Ni_3S_2 and $Ni(OH)_2$ in the composite electrodes. To calculate $m_{composite}$, the Ni_3S_2@$Ni(OH)_2$ was considered as pure Ni_3S_2 or $Ni(OH)_2$. The calculation is described as following: the weight increment (*x* mg) of Ni foam or 3DGN can be directly weighted after the synthesis of Ni_3S_2@$Ni(OH)_2$ on Ni foam or 3DGN. If assuming the composite is pure $Ni(OH)_2$, $m_{composite} = x$ mg × ($M_{Ni(OH)2}/2M_{(OH)}$) = x mg × (92.7/34) = 2.73x mg, where M is the molecular weight or atomic weight. If assuming the composite is pure Ni_3S_2, $m_{composite} = x$ mg × ($M_{Ni3S2}/2M_S$) = x mg × (240/64) = 3.75x mg. Based on the equations (1) and (2), the capacitance of composite electrodes calculated by Ni_3S_2 is less than that calculated by $Ni(OH)_2$, which was shown in Figure S5. In our paper, we use the lower capacitance values based on Ni_3S_2. Therefore, the actual capacitance value of Ni_3S_2@$Ni(OH)_2$/3DGN is higher than the presented value in the paper.

$$C_{m1} = \frac{\int I_1 dV}{vm\triangle V} \qquad (1)$$

$$C_{m2} = \frac{I_2 \triangle t}{m\triangle V} \qquad (2)$$

Areal capacitances were calculated from CV and charge-discharge curves by the equations (3) and (4), respectively. *S* is the geometrical area of the electrode.

$$C_{S1} = \frac{\int I_1 dV}{vS\triangle V} \qquad (3)$$

$$C_{S2} = \frac{I_2 \triangle t}{S\triangle V} \qquad (4)$$

Energy density (*E*) and Power density (*P*) were calculated by the (5) and (6) equations, respectively.

$$E = \frac{1}{2} C_{m2} (\triangle V)^2 \qquad (5)$$

$$P = \frac{E}{\triangle \mathrm{t}} \qquad (6)$$

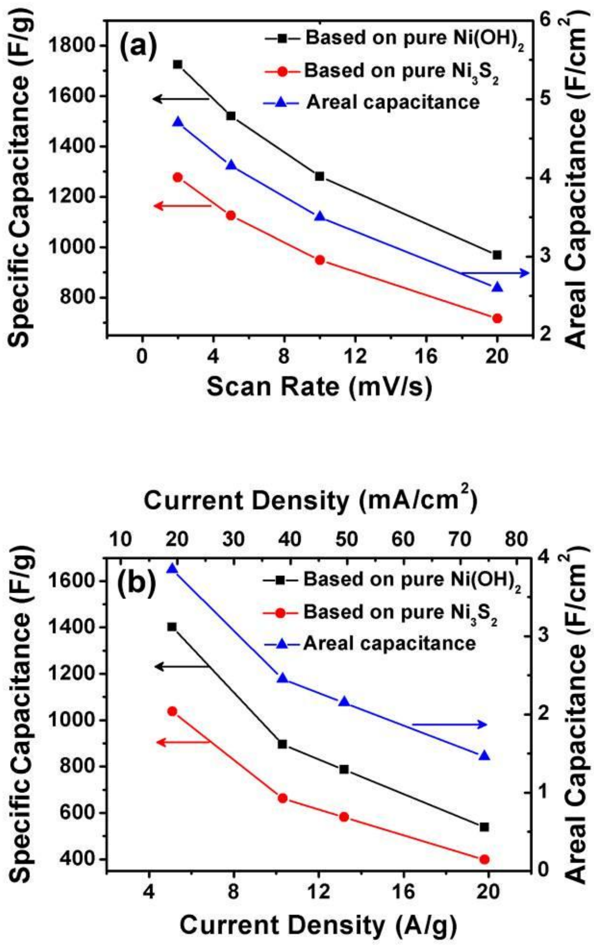

Figure S5. Calculated specific capacitance and areal capacitance as a function of (a) scan rate and (b) current density for Ni_3S_2@$Ni(OH)_2$/3DGN electrode.

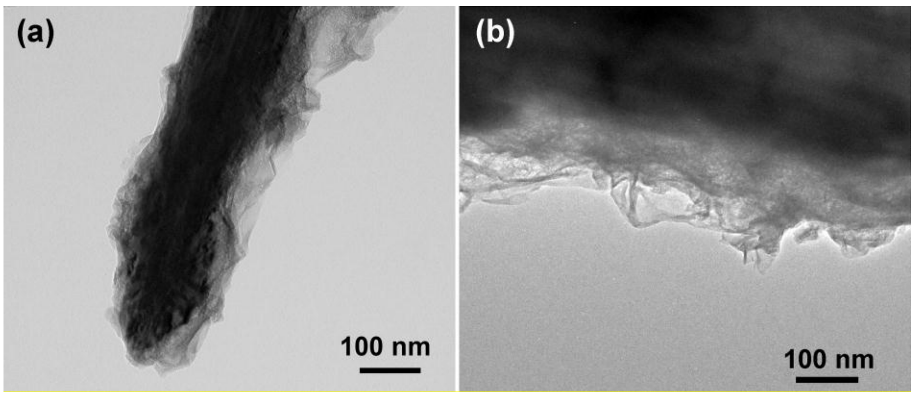

Figure S6. TEM images of the Ni_3S_2@$Ni(OH)_2$ sample after 2000 cycles.

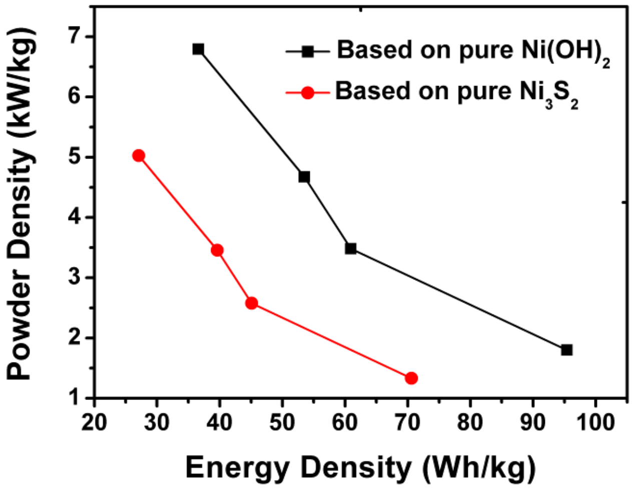

Figure S7. Ragone plot of energy density (*E*) versus power density (*P*) for Ni_3S_2@$Ni(OH)_2$/3DGN electrode.