Improving the specific Capacity of Nickel Hydroxide Nanocrystals for Hybrid Supercapacitors via Yttrium Doping

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1. Experimental

1.1 Fabrication of Y-doped Ni(OH)₂ composites

In a typical synthesis, Ni(NO₃)₂·6H₂O (2 mmol) and Y(NO₃)₃·6H₂O (Ni/Y mole ratio of 10:1) was dissolved into 40 mL distilled water. 10 mL of 0.2 M ammonia were slowly dropped into the above solution with stirring for 30 min, and the green solution was sealed in 60 mL Teflon-lined autoclave. Then, the autoclave was heated to 180 °C in oven for 18 h. After cooled naturally to room temperature, the Y-Ni10 samples were collected after washed several times and dried at 80 °C overnight. For comparison, the Y-Ni0, Y-Ni5 and Y-Ni20 samples were also fabricated by the similar process (Ni/Y molar ratios of 0, 20:1 and 5:1).

1.2 Sample characterization

The as-prepared samples were characterized by scanning field-emission electron microscopy (SEM, Carl sigma 500 AMCS), transmission electron microscopy (TEM, Talos FEI), X-ray diffraction (XRD, Bruker, D8 ADVANCE) with Cu K α radiation ($\lambda = 0.15418$ nm), X-ray photoelectron spectroscopy (XPS, Thermo ESCALAB 250ZI), and the nitrogen adsorption apparatus (ASAP2460, SN:543).

1.3 Electrochemical evaluation

The working electrodes were fabricated by loading the homogeneous slurry onto nickel foam. The slurry were prepared from 80% active materials, 15% acetylene black, 5% polytetrafluoroethylene and appropriate amount of ethanol solvent. The mass loading of the active material on these electrodes is around 2.1 mg/cm². The electrochemical properties containing galvanostatic charge discharge (GCD), cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) of working electrodes were tested in three-electrode glass cell with 6 M KOH electrolytes, Hg/HgO and platinum-plate were served as the reference and counter electrodes, respectively.

The Y-Ni10//AC cell was constructed by using the Y-Ni10 as the positive and active carbon (AC) as negative electrodes. The mass ratio of was obtained from the equation of $C_{-}\times m_{-}\times V_{-}=C_{+}\times m_{+}\times V_{+}$, where C_{+} and C_{-} are the specific capacitance of Y-Ni10 and AC electrodes, V_{+} and V_{-} are the voltage range of Y-Ni10 and AC, m_{+} and m_{-} are the active mass loadings of Y-Ni10 and AC, respectively. According to the GCD studies of the individual Y-Ni10 and AC electrodes, the calculated mass ratio of 1:3.86 is used to balance the charges.

The specific capacitance (C, F g⁻¹), specific energy density (SE, W h kg⁻¹) and power density (SP, W kg⁻¹) of Y-Ni10//AC cell was obtained based on the following equation:

$$C = \frac{2I \int V dt}{MV^2 | \frac{Vf}{Vi}}$$
(1)
$$SE = \frac{I \int V dt}{M}$$
(2)

$$SP = \frac{1}{\Delta t}$$
(3)

Where I (A) is the discharge current, M (g) is the total mass of Y-Ni10 and AC materials and Δt (s) is corresponding to discharge time, V_i and V_f are the cell voltage of an initial and final value,

respectively.



Fig. S1. XRD patterns of Y-Ni0



Fig. S2. SEM images of (a) Y-Ni5, (b) Y-Ni10 and (c) Y-Ni20.



| samples | Ni norm. (wt%) | Y norm. (wt%) | Y/Ni molar ratios |
|---------|----------------|---------------|-------------------|
| Y-Ni5 | 52.66 | 3.24 | 4.42% |
| Y-Ni10 | 49.97 | 7.65 | 10.99% |
| Y-Ni20 | 54.45 | 13.98 | 18.43% |

Fig. S3. EDX of the Y-Ni0, (b)Y-Ni5, (c)Y-Ni10 and (d) Y-Ni20 samples.



Table S1. the mass ratios of the Ni and Y elements in Y-Ni5, Y-Ni10 and Y-Ni20 samples.

Fig. S4. HRTEM images of Y-Ni0 sample.



Fig. S5. N₂ absorption-desorption isotherm of (a) Y-Ni0 and (b) Y-Ni5 samples (insets of the pore distribution).

| Sample | Specific surface area | Total pore volume | Average pore diameter |
|--------|-----------------------|-------------------|-----------------------|
| | $(m^2 g^{-1})$ | $(cm^3 g^{-1})$ | (nm) |
| Y-Ni0 | 6.7931 | 0.034545 | 20.4408 |
| Y-Ni5 | 32.0665 | 0.157677 | 18.3113 |
| Y-Ni10 | 119.3531 | 0.266668 | 7.9563 |
| Y-Ni20 | 105.2588 | 0.214238 | 7.3784 |



Table S2. A comparison study of BET and pore diameter.

Fig. S6. CV curves at different scan rates of (a) Y-Ni0, (c) Y-Ni5 and (e) Y-Ni20, Galvanostatic charge-discharge curves of (b) Y-Ni0, (d) Y-Ni5 and (f) Y-Ni20.



Fig. S7. (a) Galvanostatic charge-discharge curves of Y-Ni10; (b) Specific capacitance of Y-Ni0, Y-Ni5,

Y-Ni10 and Y-Ni20 electrodes



Fig. S8. the linear relation between the oxidation/reduction peak current and the scan rates of Y-Ni0,

Y-Ni5, Y-Ni10 and Y-Ni20.



Fig. S9. CV curves at different scan rates and Galvanostatic charge-discharge curves of AC.



Fig. S10. (a) CV curves of the Y-Ni10 and AC electrodes in the three-electrode system at 10 mV⁻¹.



Fig. S11. (a) CV curves at different scan rates and (b) GCD curves of the Y-Ni10//AC device.

| Electrode materials | Specific Capacity | capacity retention | Refs | | | | |
|---|--|--------------------------------|------------|--|--|--|--|
| Ni-CO-Mn-OH/RGO | 665 C g ⁻¹ at 2 A g ⁻¹ | 64.21% at 20 A g ⁻¹ | S 1 | | | | |
| NiCoMn-OH | 757 C g ⁻¹ at 1 A g ⁻¹ | 48.7% at 50 A g ⁻¹ | S2 | | | | |
| NiCoAl-LDH nanoplates | 564.5 C g ⁻¹ at 1 A g ⁻¹ | 59% at 30 A g ⁻¹ | S3 | | | | |
| Co ₃ O ₄ @NiCoAl- LDH | 441.6 C g ⁻¹ at 1 A g ⁻¹ | 60.05% at 20 Ag ⁻¹ | S4 | | | | |
| Al -Ni(OH) ₂ nanosheets | 849 C g ⁻¹ at 1 A g ⁻¹ | 65.4 % at 6 Ag ⁻¹ | S5 | | | | |
| CoMn LDH | 531.3 C g ⁻¹ at 0.7 A g ⁻¹ | 69.1% at 28.6 Ag ⁻¹ | S6 | | | | |
| NiMn-LDH/rGO | 625 C g ⁻¹ at 1 A g ⁻¹ | 36.0% at 5 A g ⁻¹ | S7 | | | | |
| Glucose NiMn-LDH | 732 C g ⁻¹ at 1 A g ⁻¹ | 59.4% at 10 A g ⁻¹ | S8 | | | | |
| Core-Shell NiAl-LDH | 367.5 C g ⁻¹ at 2 A g ⁻¹ | 75.0% at 25 A g ⁻¹ | S9 | | | | |
| CoAl-LDH | 339.3 C g ⁻¹ at 1 A g ⁻¹ | 73.6 at 20 A g ⁻¹ | S10 | | | | |
| CoAl-LDH/FGN | 611 C g ⁻¹ at 1 A g ⁻¹ | 75.3% at 10 A g ⁻¹ | S11 | | | | |

 Table S2. The specific capacitance of various electrodes in the three-electrode system in references

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