Supporting Information

 $Ni_2P_2O_7$ Nanoarrays with Decorated C_3N_4 Nanosheets as Efficient Electrode for Supercapacitors

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Experimental Section

Materials Characterization

X-ray diffraction profiles were recorded by a RIGAKU Rint-2000 X-ray diffractometer equipped with graphite monochromatized Cu-K α radiation (λ =1.54184 Å). Scanning electron microscopy and energy dispersive spectrometer mapping images was collected on a FEI Helios Nanolab 600i field emission scanning electron microscopy. Transmission electron microscopy images was produced from a FEI Tecnai G2 F20 field emission transmission electron microscopy operated at 200 kV. The X-ray photoelectron spectroscopy spectra were recorded from a Thermo Fisher ESCALAB 250Xi spectrophotometer. Fourier transform infrared spectrometer Thermo Fisher IS10 infrared spectrometer.

Three–electrodes electrochemical measurement: Electrochemical properties was measured on a Gamry Interface 1000 electrochemistry workstation in a three electrode cell by using the fabricated materials as working electrode without further treatment. Hg/HgO electrode was used as reference electrode, platinum plat electrode was acted as counter electrode, and 3 M KOH was applied as electrolyte. The cyclic voltammetry (CV), galvanostatic charge/discharge (GCD), and electrochemical impedance spectroscopy (EIS) were recorded. According to the galvanostatic discharge curves, the corresponding specific capacitance was evaluated by such an equation: $C_{sc} = (I \ \Delta t) / (S \ \Delta V)$, where the C_{sc} was the specific capacitance, I (A) was the discharge current, Δt (s) was the discharge time, S (cm²) was the areas of active materials on Ni foam, and ΔV (V) was the volt window.¹

Fabrication asymmetric supercapacitor cells

The asymmetric supercapacitor was assembled by utilizing Ni foam supported materials as positive electrode, active carbon as negative electrode, 3 M KOH as the electrolyte, and a diaphragm to separate the positive and negative electrodes. To obtain a best specific capacitance of ASCs, the charge balance the relationship between the two electrodes should

conform to $q^+ = q^-$. The mass of active materials between the two electrodes are adjusted as the following equations: $q = C \times \Delta V \times m$, $m^+/m^- = (C^- \times \Delta V^-) / (C^+ \times \Delta V^+)$, where m (g) was the active material mass on the electrode, C (F g⁻¹) was specific capacitance of electrode, and the ΔV (V) was the potential window.²



Figure S1 XRD profiles of (a) Ni foam supported NH₄NiPO₄·H₂O; (b) Ni foam/Ni₂P₂O₇, and (c) C₃N₄ coated Ni foam/Ni₂P₂O₇.



Figure S2 (a-b)SEM image of Ni foam supported $NH_4NiPO_4 \cdot H_2O$.



Figure S3 (a) EIS plots of the products; (b) enlarged curves at high frequency region.

The impedance electrochemical impedance spectroscopy was compared, as shown in Figure S3. The EIS data shows a solution resistance (Rs) and a charge–transfer resisitance (Rct). A vertical line leaning to imaginary axis represents the value of Rs. As can be seen from the inset, Rs value are 0.96, 0.85, 0.81, and 0.76 Ω for bare Ni₂P₂O₇, C₃N₄-1/Ni₂P₂O₇, C₃N₄-3/Ni₂P₂O₇, and C₃N₄-6/Ni₂P₂O₇, respectively. At high frequency region, it displays a negligible semicircle, revealing the low charge transfer resistence for Ni foam with pasted Co_{0.2}Ni_{0.8} pyrophosphates. The Rct value are 0.87, 0.94, 1.03, and 1.08 Ω for bare Ni₂P₂O₇, C₃N₄-1/Ni₂P₂O₇, C₃N₄-1/Ni₂P₂O₇, and C₃N₄-6/Ni₂P₂O₇, and C₃N₄-6/Ni₂P₂O₇, respectively.



Figure S4 The panoramic SEM images of (a) $Ni_2P_2O_7$, (b) $C_3N_4-1/Ni_2P_2O_7$, (c) $C_3N_4-3/Ni_2P_2O_7$, and (d) $C_3N_4-6/Ni_2P_2O_7$ after charge and discharge approximate 1,000 times at 2 mA cm⁻².



Figure S5 (a) Specific capacitance and (b) Ragone plot of as assembled ASC (The weight of C_3N_4 -3/Ni₂P₂O₇ is about 2.6 mg, carbon black is about 7.1 mg).

No.	Materials	Capacitance	Electrolyte	Ref.
1	Co ₃ O ₄ /CC	$400 \text{ mF/cm}^2 \text{ at } 4 \text{ mA/cm}^2$	3 М КОН	3
2	MnO@C/CC	720 mF/cm ² at 4 mA//cm ²	3 М КОН	3
3	Li ₂ Co ₂ (MoO ₄) ₃	1.03 F/cm ² at ~ 1 mA/cm ²	2 M LiOH	4
4	NiCo ₂ O ₄ nanoneedle arrays	0.99 F/cm ² at 5.56 mA/cm ²	2 М КОН	5
5	Co _{0.33} Ni _{0.67} DHs/ NiCo ₂ O ₄ /CFP	$\sim 2.3 \text{ F/cm}^2 \text{ at } 2 \text{ mA/cm}^2$	1 М КОН	6
6	Co ₃ O ₄ @MnO ₂	0.56 F/cm ² at 11.25 mA/cm ²	1 M LiOH	7
7	Na-doped $Ni_2P_2O_7//AC^{a}$	$32.6 \text{ mF/cm}^2 \text{ at } 0.5 \text{ mA/cm}^2$	PVA/KOH gel	8
8	Ni ₂ P ₂ O ₇ powders // AC ^{a)}	$\frac{30 \text{ mF/cm}^2 \text{ at } 0.1}{\text{mA/cm}^2}$	PVA/KOH gel	9

Table S1. A comparison with previously reported transition metal based materials.

^{a)} as ASCs device

Referneces

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