

Supporting Information for

3D Hierarchical Ni(PO₃)₂ Nanosheet Arrays with Superior Electrochemical Capacitance Behaviors

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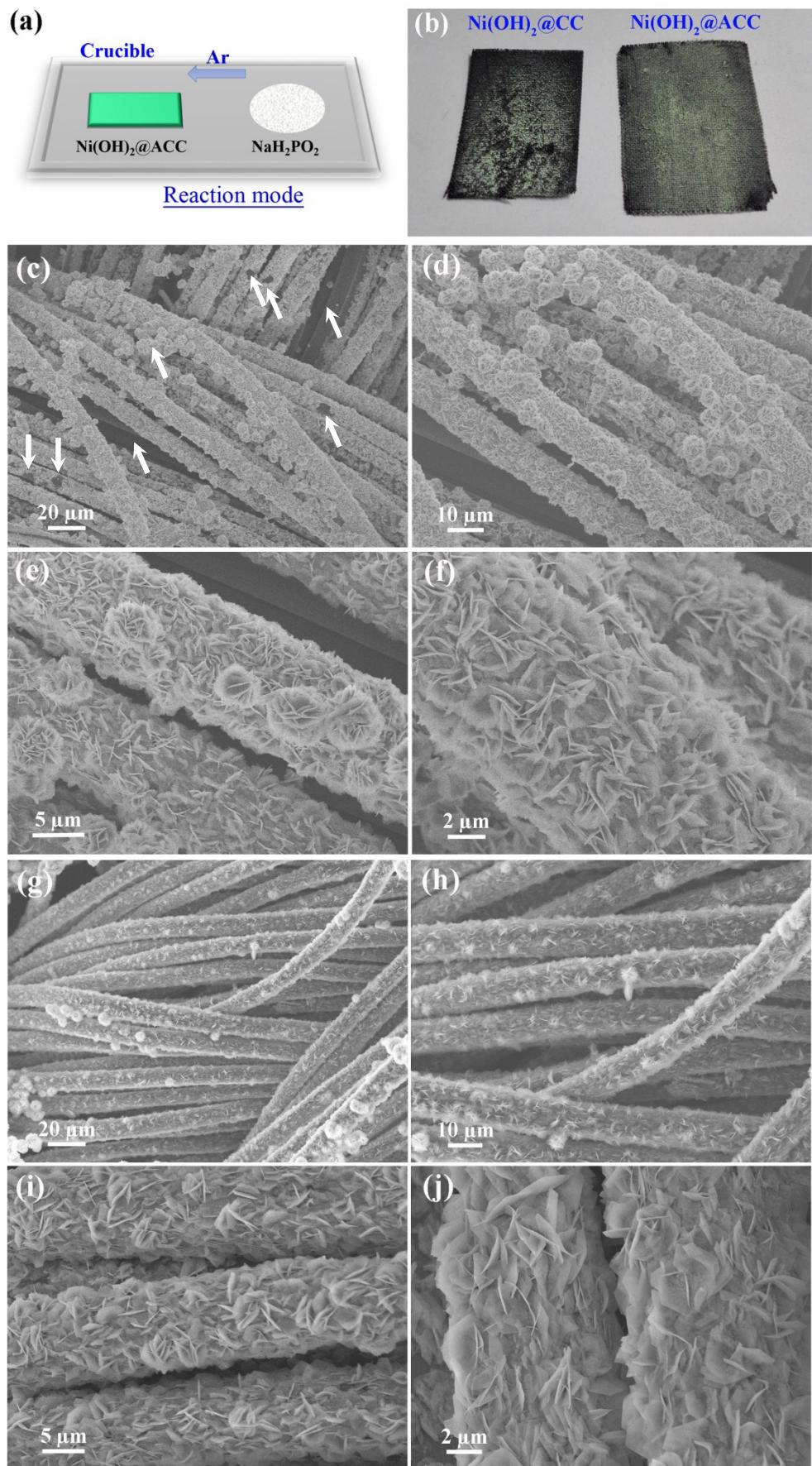


Fig. S1 (a) Schematic illumination for the synthesis of 3D hierarchical $\text{Ni}(\text{PO}_3)_2$

nanosheet arrays by the reaction of $\text{Ni(OH)}_2@\text{ACC}$ and $\text{Na}_2\text{H}_2\text{PO}_2$. (b) Digital photo of $\text{Ni(OH)}_2@\text{CC}$ and $\text{Ni(OH)}_2@\text{ACC}$. (c-f) and (g-j) Typical SEM images of $\text{Ni(OH)}_2@\text{CC}$ and $\text{Ni(OH)}_2@\text{ACC}$ under different magnifications, respectively.

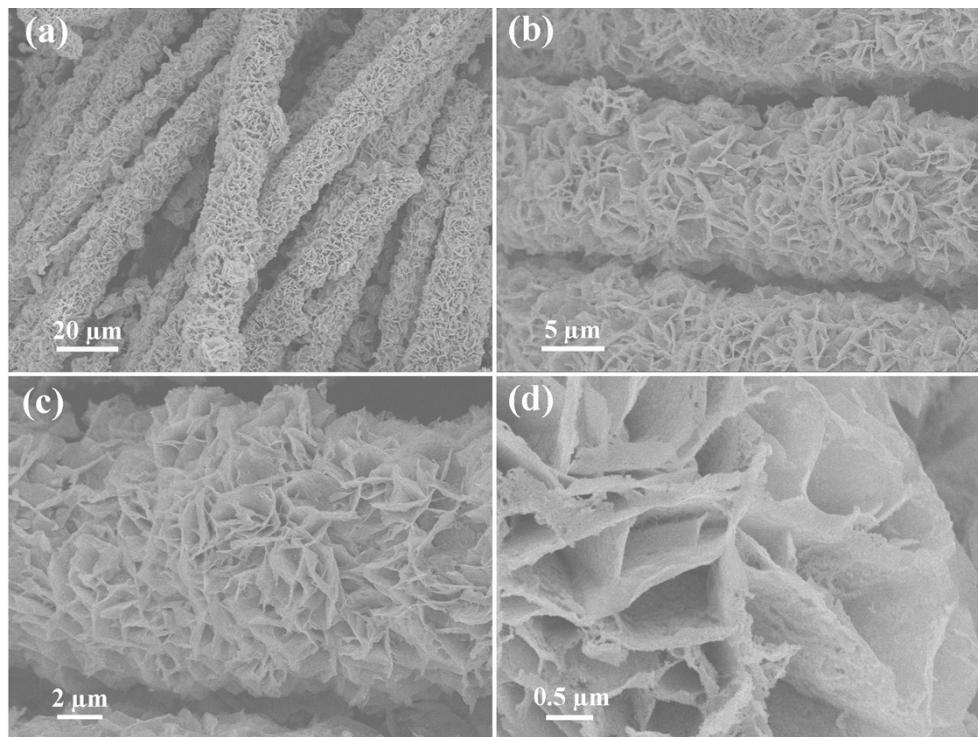


Fig. S2 Typical SEM images under different magnifications of the resultant $\text{Ni}(\text{PO}_3)_2@\text{ACC}$.

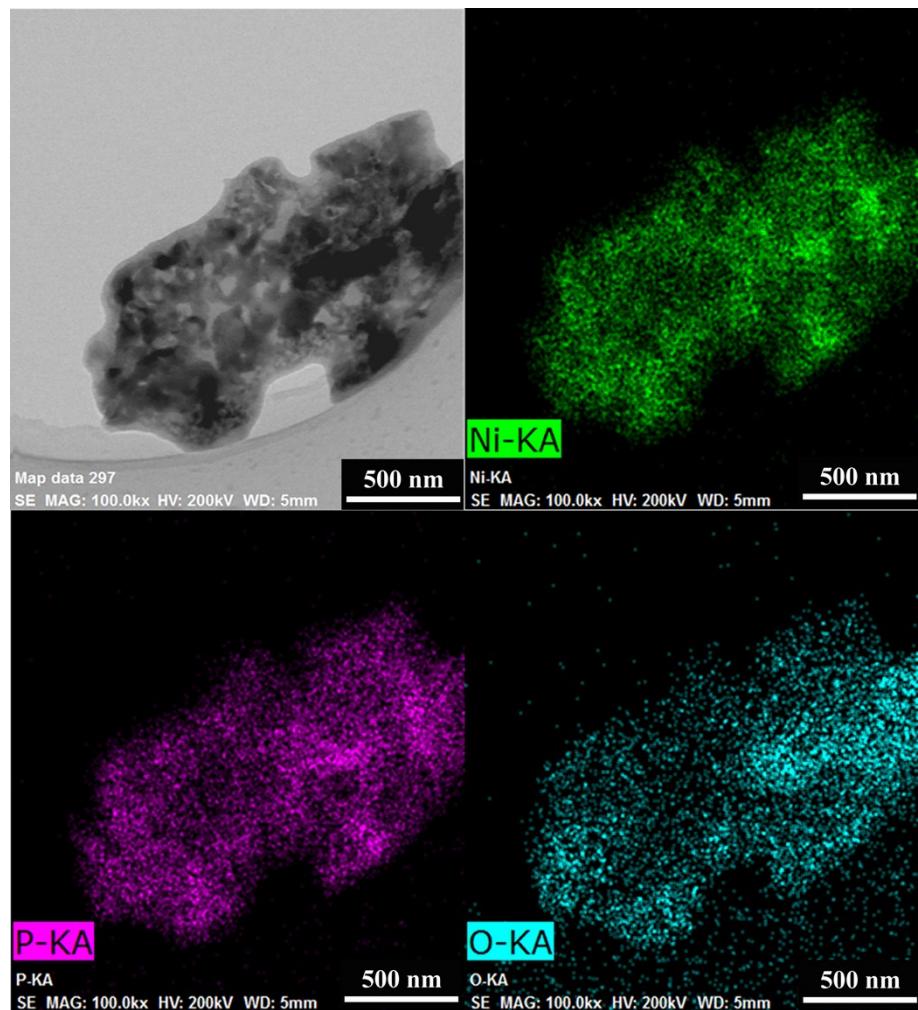


Fig. S3 Elemental mappings of Ni, P and O within the $\text{Ni}(\text{PO}_3)_2$ nanosheet.

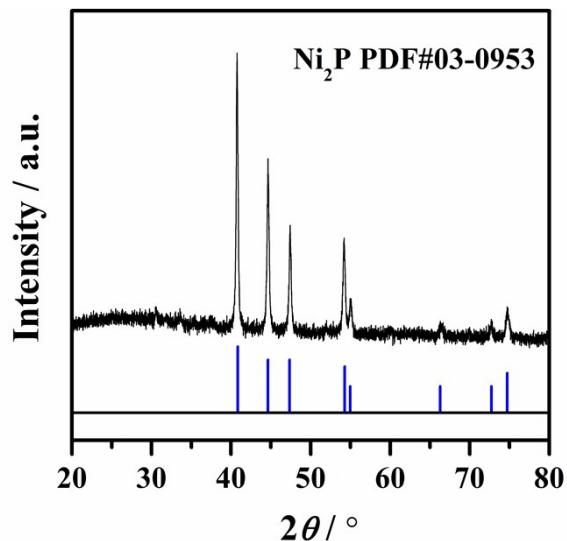


Fig. S4 Typical XRD pattern of the obtained sample by annealing $\text{Ni}(\text{OH})_2$ precursor powders and NaH_2PO_2 without the use of ACC at 300 °C in Ar atmosphere.

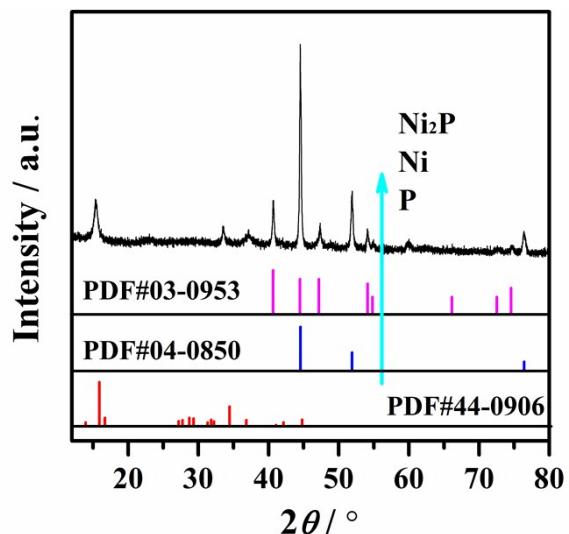


Fig. S5 Typical XRD pattern of the resultant sample by the annealing of Ni(OH)_2 precursor powders and NaH_2PO_2 at 250 °C in Ar atmosphere.

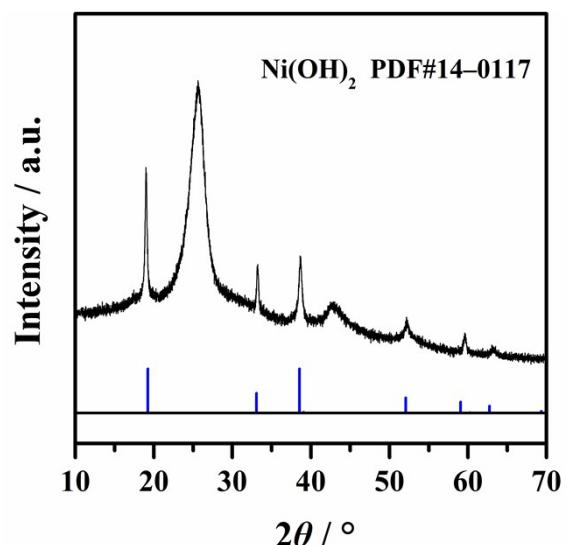


Fig. S6 XRD pattern of the annealed Ni(OH)_2 @ACC that was obtained by the heat treatment of hydrothermally-obtained hybrid at 300 °C in Ar atmosphere.

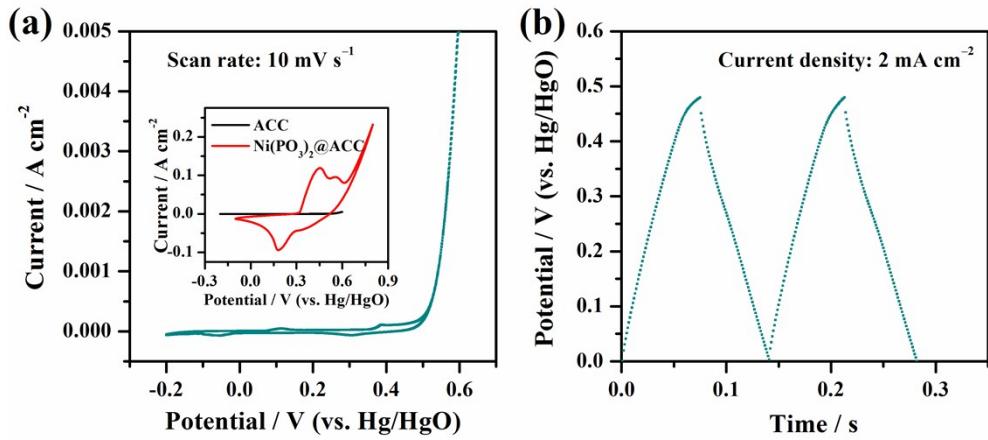


Fig. S7 Typical CV (a) and GCD (b) curves of the pure ACC electrode. The inset in Fig. S7a displays the CV comparison between ACC and Ni(PO₃)₂@ACC electrodes at the same scan rate of 10 mV s⁻¹. The *ca.* areal specific capacitance of ACC is 0.28 F cm⁻² at the discharge current of 2 mA cm⁻².

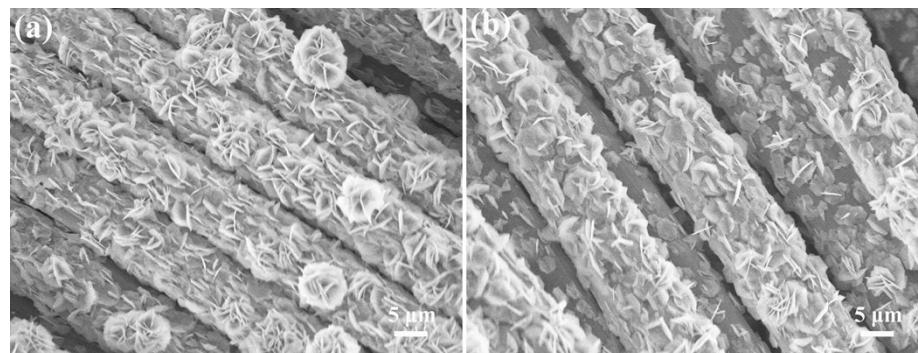


Fig. S8 SEM images of $\text{Ni}(\text{PO}_3)_2@\text{ACC}$ electrodes cycled after 800 (a) and 2000 (b) times at the discharge current of 10 mA cm^{-2} .

Table S1. Summary of comparison experiments and the crystalline phases of the corresponding products.

| Precursors | Temperature / °C | Products |
|----------------------------|------------------|--|
| Ni(OH) ₂ powder | 250 | Ni, Ni ₂ P and P |
| | 300 | Ni ₂ P |
| Ni(OH) ₂ @ACC | 250 | Ni(OH) ₂ @ACC |
| | 300 | Ni(PO ₃) ₂ @ACC |
| | 350 | Ni(PO ₃) ₂ @ACC |

Table S2. Overview of the electrochemical performance of nickel-based electrode materials for ultracapacitors.

| Materials | Electrolyte s | Specific Capacitance | Reference |
|---|---|---|-----------|
| α -Ni(OH) ₂ /graphene | 6 M KOH | 1735 F g ⁻¹ at 1 mV s ⁻¹ | 1 |
| β -Ni(OH) ₂ /graphite | 1 M KOH | 1335 F g ⁻¹ at 2.8 A g ⁻¹ | 2 |
| β -Ni(OH) ₂ /carbon cloth | 1 M NaOH | 2675 F g ⁻¹ at 5 mA cm ⁻² 7.85 F cm ⁻² at 5 mA cm ⁻² | 3 |
| amorphous Ni(OH) ₂ | 1 M KOH | 1863 F g ⁻¹ at 20 A g ⁻¹ | 4 |
| α -Ni(OH) ₂ nanosheets | 6 M KOH | 3270 F g ⁻¹ at 4 A g ⁻¹ | 5 |
| mixture of α -Ni(OH) ₂ and β - Ni(OH) ₂ /carbon fibers | 1 M KOH | 1416 F g ⁻¹ at 1 A g ⁻¹ | 6 |
| NiCo ₂ O ₄ /Ni foam | 3 M KOH | 2010 F g ⁻¹ at 2 A g ⁻¹ | 7 |
| NiCo ₂ O ₄ /carbon foam | 6 M KOH | 1231 F g ⁻¹ at 2 A g ⁻¹ | 8 |
| NiCo ₂ O ₄ /carbon cloth | 1 M KOH | 245 F g ⁻¹ at 1 A g ⁻¹ | 9 |
| NiCo ₂ O ₄ /carbon fabric | 2 M KOH | 2658 F g ⁻¹ at 2 A g ⁻¹ | 10 |
| Co _{0.5} Ni _{0.5} double hydroxide/NiCo ₂ O ₄ /carbon fiber paper | 1 M NaOH | 2.3 F cm ⁻² at 2 mA cm ⁻² | 11 |
| Ni(OH) ₂ -MnO ₂ /Ni foam | 1 M KOH | 2628 F g ⁻¹ at 3 A g ⁻¹ | 12 |
| NiCo ₂ O ₄ hollow spheres | 6 M KOH | 1141 F g ⁻¹ at 1 A g ⁻¹ | 13 |
| Co _{0.5} Mn _{0.4} Ni _{0.1} C ₂ O ₄ •nH ₂ O | 3 M KOH | 990 F g ⁻¹ at 0.6 A g ⁻¹ | 14 |
| NiFe ₂ O ₄ /carbon cloth | 1 M H ₂ SO ₄ 6 M KOH | 1135.5 F g ⁻¹ (H ₂ SO ₄) 922.6 F g ⁻¹ (KOH) at 2 mA cm ⁻² | 15 |
| α -NiS/carbon nanorods | 2 M KOH | 1092 F g ⁻¹ at 1 A g ⁻¹ | 16 |
| flower-like β -NiS | 2 M KOH | 857.76 F g ⁻¹ at 2 A g ⁻¹ | 17 |
| hollow NiS ₂ prisms | 2 M LiOH | 1725 F g ⁻¹ at 5 A g ⁻¹ | 18 |
| NiS ₂ sponge | 2 M KOH | 1685 F g ⁻¹ at 1 A g ⁻¹ | 19 |
| Ni ₃ S ₂ /Ni foam | 1 M KOH | 7.25 F cm ⁻² at 5 mA cm ⁻² | 20 |
| NiCo ₂ S ₄ aerogel | 1 M NaOH | 1400 F g ⁻¹ at 25mV s ⁻¹ | 21 |
| NiCo ₂ S ₄ /graphene | 6 M KOH | 1451 F g ⁻¹ at 3 A g ⁻¹ | 22 |
| NiCo ₂ S ₄ /carbon cloth | 1 M KOH | 1418 F g ⁻¹ at 5 A g ⁻¹ | 23 |
| Tube-like NiCo ₂ S ₄ | 6 M KOH | 1048 F g ⁻¹ at 3 A g ⁻¹ | 24 |
| Co _x Ni _{1-x} (OH) ₂ /NiCo ₂ S ₄ nanotube array on carbon fiber paper | 1 M KOH | 2.86 F cm ⁻² at 4 mA cm ⁻² | 25 |
| Ni ₂ P particles | 2 M KOH | 843.25 F g ⁻¹ at 1 A g ⁻¹ | 26 |
| Ni ₂ P/Ni foam | 2 M LiOH | 581 F g ⁻¹ at 1 A g ⁻¹ | 27 |
| Ni ₂ P/graphene | 2 M KOH | 2266 F g ⁻¹ at 5 mA cm ⁻² | 28 |
| NH ₄ NiPO ₄ •H ₂ O nanoalmonds | 3 M KOH | 1072 F g ⁻¹ at 1.5 A g ⁻¹ | 29 |

| | | | |
|---|--------------------|---|-----------|
| $\text{Ni}_{11}(\text{HPO}_3)_8(\text{OH})_6$ nanotube assembly | 1 M KOH 3 M KOH | 687 F g ⁻¹ (1 M KOH) 1876 F g ⁻¹ (3 M KOH) at 0.625 A g ⁻¹ | 30 |
| amorphous NiWO_4 nanostructure | 2 M KOH | 586.2 F g ⁻¹ at 0.5 A g ⁻¹ | 31 |
| $\text{Co}_3\text{O}_4-\text{Ni}_3(\text{VO}_4)_2$ nanorods/Ni foam | 2 M KOH | 1401 F g ⁻¹ at 0.5 A g ⁻¹ | 32 |
| $\text{NiCo}_2\text{S}_4@\text{Ni}_3\text{V}_2\text{O}_8$ core/shell hybrid /Ni foam | 2 M KOH | 512 C g ⁻¹ at 1 A g ⁻¹ | 33 |
| $\text{Ni}(\text{HCO}_3)_2$ /graphene | 6 M KOH | 1200 F g ⁻¹ at 4 A g ⁻¹ | 34 |
| $\text{Ni}(\text{HCO}_3)_2$ /Ni foam | 2 M KOH | 2128.57 F g ⁻¹ at 2.5 mA cm ⁻² | 35 |
| NiMoO_4 / Ni foam | 3 M KOH | 3.4 F cm ⁻² (2138 F g ⁻¹) at 2 mA cm ⁻² | 36 |
| $\text{Ni}_{11}(\text{HPO}_3)_8(\text{OH})_6$ nanoribbons | 3 M KOH | 1876 F g ⁻¹ at 0.625 A g ⁻¹ | 37 |
| $\text{NiCo}_2(\text{PO}_4)_2$ hollow shells | 3 M KOH | 940.43 F g ⁻¹ at ~1 A g ⁻¹ | 38 |
| $\text{Ni}_2\text{P}_2\text{O}_7$ /porous carbon | 1 M NaOH | 1893 F g ⁻¹ at 2 A g ⁻¹ | 39 |
| $\text{Ni}_3\text{P}_2\text{O}_8-\text{Co}_3\text{P}_2\text{O}_8 \cdot 8\text{H}_2\text{O}$ | 6 M KOH | 1974 F g ⁻¹ at 0.5 A g ⁻¹ | 40 |
| Ni-MOF-derived porous $\text{Ni}_x\text{P}_y\text{O}_z$ | 2 M KOH | 1627 F g ⁻¹ at 1 A g ⁻¹ | 41 |
| Ni@NiO core–shell nanoparticle tube arrays | 1 M NaOH | 263 F g ⁻¹ at 2.2 A g ⁻¹ | 42 |
| NiO/Ni composites | 2 M KOH | 1204 C g ⁻¹ at 1 A g ⁻¹ | 43 |
| $\text{Ni}(\text{PO}_3)_2$ @activated carbon cloth | 3 M KOH | 2237 F g ⁻¹ at ~1 A g ⁻¹ | This work |

Table S3. Overview of the electrochemical performances of transition metal phosphates for ultracapacitor electrodes.

| Materials | Electrolytes | Specific capacitance | Reference |
|---|--------------|---|-----------|
| $\text{NH}_4\text{NiPO}_4 \cdot \text{H}_2\text{O}$ | 3 M KOH | 1072 F g ⁻¹ at 1.5 A g ⁻¹ | 29 |
| $\text{Ni}_{11}(\text{HPO}_3)_8(\text{OH})_6$ | 3 M KOH | 1876 F g ⁻¹ at 0.625 A g ⁻¹ | 37 |
| $\text{Co}_{11}(\text{HPO}_3)_8(\text{OH})_6$ | 3 M KOH | 312 F g ⁻¹ at 1.25 A g ⁻¹ | 37 |
| $\text{NH}_4\text{CoPO}_4 \cdot \text{H}_2\text{O}$ | 3 M KOH | 340 F g ⁻¹ at 1.5 A g ⁻¹ | 44 |
| $\text{CoHPO}_4 \cdot 3\text{H}_2\text{O}$ | 3 M KOH | 413 F g ⁻¹ at 1.5 A g ⁻¹ | 45 |
| $\text{Co}_2\text{P}_2\text{O}_7$ | 3 M KOH | 367 F g ⁻¹ at 0.625 A g ⁻¹ | 45 |
| Mixed phases of $(\text{Ni},\text{Co})_3(\text{PO}_4)_2 \cdot 8\text{H}_2\text{O}$ $(\text{NH}_4)(\text{Ni},\text{Co})\text{PO}_4 \cdot 0.67\text{H}_2\text{O}$ | 6 M KOH | 1128 F g ⁻¹ at 0.5 A g ⁻¹ | 46 |
| $\text{Mn}_3(\text{PO}_4)_2 \cdot 3\text{H}_2\text{O}$ | 6 M KOH | 2086 F g ⁻¹ at 1 mV s ⁻¹ | 47 |
| $\text{Ni}_2\text{P}_2\text{O}_7/\text{C}$ | 1 M NaOH | 1893 F g ⁻¹ at 2 A g ⁻¹ | 39 |
| $\text{Co}_{0.86}\text{Ni}_{2.14}(\text{PO}_4)_2$ | 6 M KOH | 1409 F g ⁻¹ at 0.25 A g ⁻¹ | 48 |
| $\text{NiCo}_2(\text{PO}_4)_2$ | 3 M KOH | 940.4 F g ⁻¹ at ~1 A g ⁻¹ | 38 |
| $\text{Ni}(\text{PO}_3)_2 @ \text{activated carbon cloth}$ | 3 M KOH | 2237 F g ⁻¹ at ~1 A g ⁻¹ | This work |

Table S4. Overview of the electrochemical performances of supported inorganic composites for ultracapacitor electrodes.

| Materials | Electrolytes | Areal capacitance | Reference |
|---|---|---|-----------|
| $\alpha\text{-Ni(OH)}_2/\text{Ni}$ foam | 3% KOH | 1.58 at 2 mA cm ⁻² Loading: 0.5 mg cm ⁻² | 49 |
| $\beta\text{-Ni(OH)}_2/\text{carbon cloth}$ | 1 M NaOH | 7.85 at 5 mA cm ⁻² Loading: 2.93 mg cm ⁻² | 3 |
| $\text{NiCo}_2\text{O}_4/\text{Ni}$ foam | 2 M KOH | 1.83 at 18 mA cm ⁻² Loading: 3 mg cm ⁻² | 50 |
| $\text{Co}_{0.5}\text{Ni}_{0.5}$ double hydroxide/ NiCo_2O_4 / carbon fiber paper | 1 M NaOH | 2.3 at 2 mA cm ⁻² Loading: 0.5 mg cm ⁻² | 11 |
| $\text{NiFe}_2\text{O}_4/\text{carbon cloth}$ | 1 M H ₂ SO ₄ 6 M KOH | 1.76 (H ₂ SO ₄), 1.43 (KOH) at 2 mA cm ⁻² Loading: 1.55 mg cm ⁻² | 15 |
| $\text{Co}_x\text{Ni}_{1-x}(\text{OH})_2/\text{NiCo}_2\text{S}_4$ nanotube array/carbon fiber paper | 1 M KOH | 2.86 at 4 mA cm ⁻² Loading: 7.8 mg cm ⁻² | 25 |
| $\text{NiCo}_2\text{S}_4/\text{carbon cloth}$ | 1 M KOH | 0.567 at 2 mA cm ⁻² Loading: 0.4 mg cm ⁻² | 23 |
| activated carbon cloth | 1 M H ₂ SO ₄ | ~0.076 at 5 mA cm ⁻² | 51 |
| $\text{NiCo}_2\text{O}_4@\text{MnO}_2/\text{Ni}$ foam | 1.0 M LiOH | 3.31 at 2 mV s ⁻¹ Loading: 1.4 mg cm ⁻² | 52 |
| $\text{Co}_9\text{S}_8/\text{carbon cloth}$ | 3 M KOH | 2.35 at 5 mA cm ⁻² | 53 |
| $\text{Co}_3\text{O}_4@\text{RuO}_2/\text{carbon cloth}$ | 3 M KOH | 1.18 at 1 mV s ⁻¹ Loading: 2 mg cm ⁻² | 53 |
| $\text{NiO-TiO}_2/\text{Ti}$ foil | 1 M KOH | 3 at 0.4 mA cm ⁻² | 54 |
| $\text{Co}_3\text{O}_4@\text{MnO}_2/\text{stainless steel}$ | 1.0 M LiOH | 0.56 at 11.25 mA cm ⁻² Loading: 1.5 mg cm ⁻² | 55 |
| $\text{Co}_3\text{O}_4@\text{NiO/Ni}$ foam | 2 M KOH | 1.35 at 6 mA cm ⁻² Loading: 3 mg cm ⁻² | 56 |
| $\text{WO}_{3-x}@\text{Au}@\text{MnO}_2/\text{carbon fabric}$ | 0.1 M Na ₂ SO ₄ | ~0.37 at 0.23 mA cm ⁻² MnO ₂ loading: 0.31 mg cm ⁻² | 57 |
| $\text{Fe}_2\text{O}_3/\text{carbon cloth}$ | 2.0 M Li ₂ SO ₄ | 1.784 at 2.0 mA cm ⁻² Loading: 1.8 mg cm ⁻² | 58 |
| $\text{NiCo}_2\text{O}_4@\text{Ni}$ foam | 6 M KOH | 2.1 F cm ⁻² at 1 mA cm ⁻² Loading: 2.62 mg cm ⁻² | 59 |
| $\text{Ni(PO}_3)_2@\text{activated carbon cloth}$ | 3 M KOH | 4.43 at 2 mA cm ⁻² Loading: 1.98 mg cm ⁻² | This work |

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