

Synergistic Integration of Mixed Metal Phosphate and Poly(1H-Pyrrole) for High-Performance Asymmetric Supercapacitor

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Figure S1: EDX elemental mapping analysis of optimized composite MMP-PPY2.

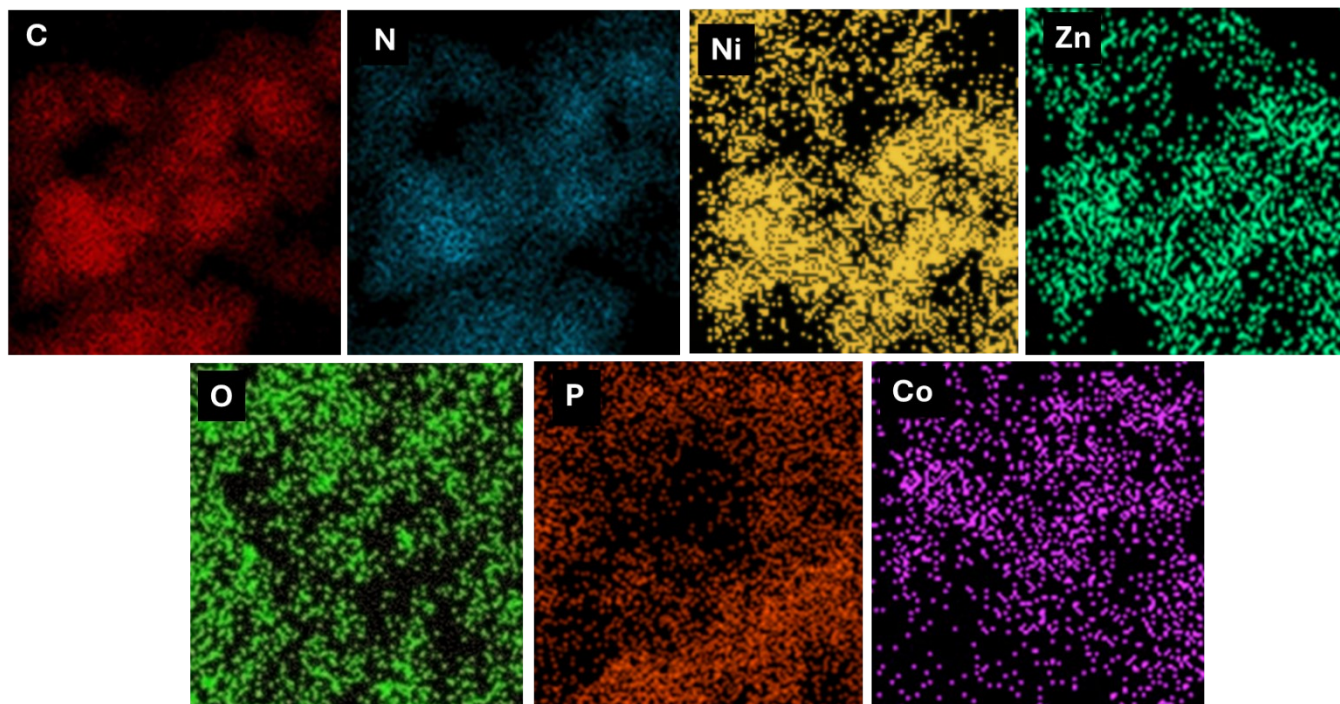
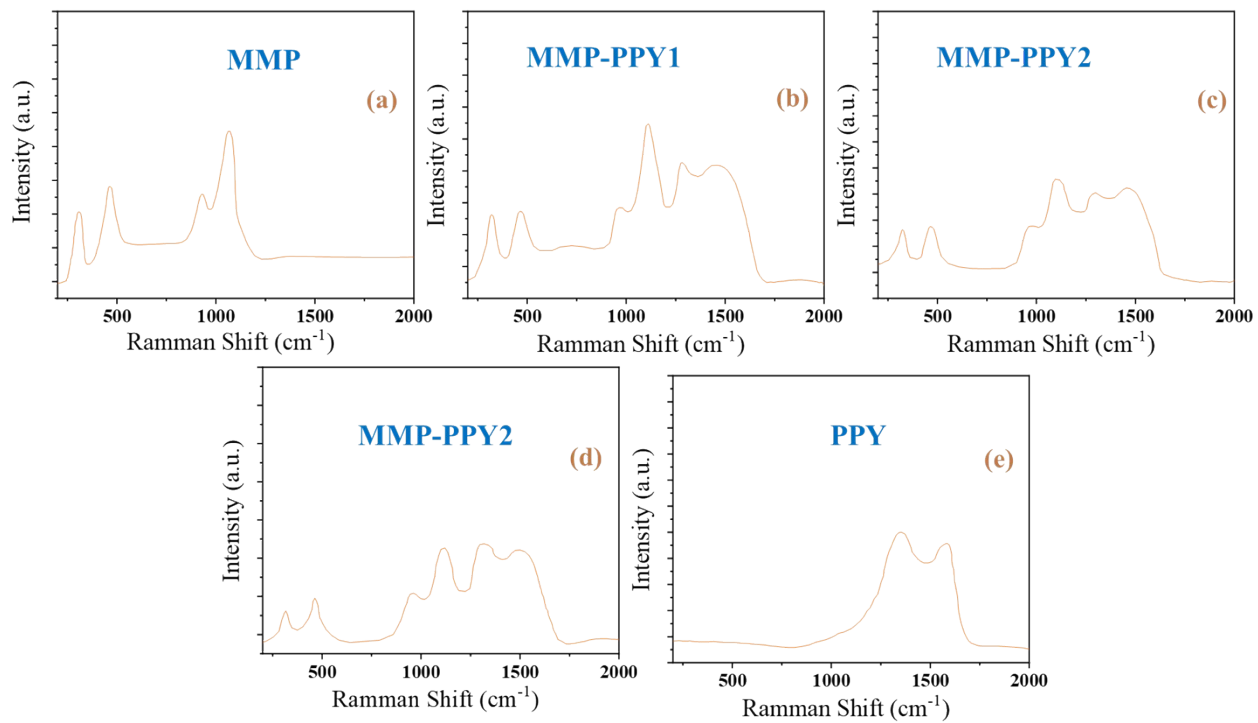


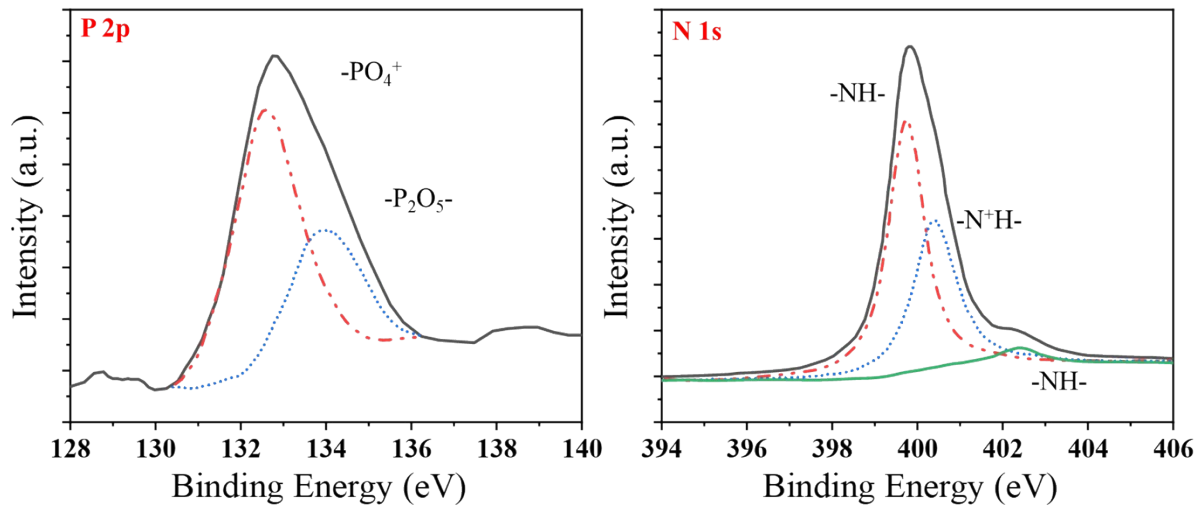
Figure S2: Raman spectra of (a) pure Zn-doped Ni–Co phosphate, (b–e) Zn-doped Ni–Co



phosphate/polypyrrole (PPy) composites with increasing PPy content, and (f) PPy.

Figure S3: The XPS spectra of the Zn–NiCo(PO₄)₂/PPy composite (MMP-PPY2).

The P 2p spectrum shows a characteristic peak around ~133–134 eV corresponding to PO₄³⁻ and P₂O₅ species, while the N 1s spectrum exhibits peaks at ~398–402 eV attributed to different nitrogen states in polypyrrole, including neutral amine (=N–/–NH–) and positively charged nitrogen (N⁺). Notably, these binding energies remain nearly unchanged compared to the individual materials, indicating that the electronic environments of phosphorus and nitrogen are preserved. This absence of significant peak shifts or new components suggests that no chemical bonding or charge transfer occurs between Zn–Ni–Co(PO₄)₂ and PPy.



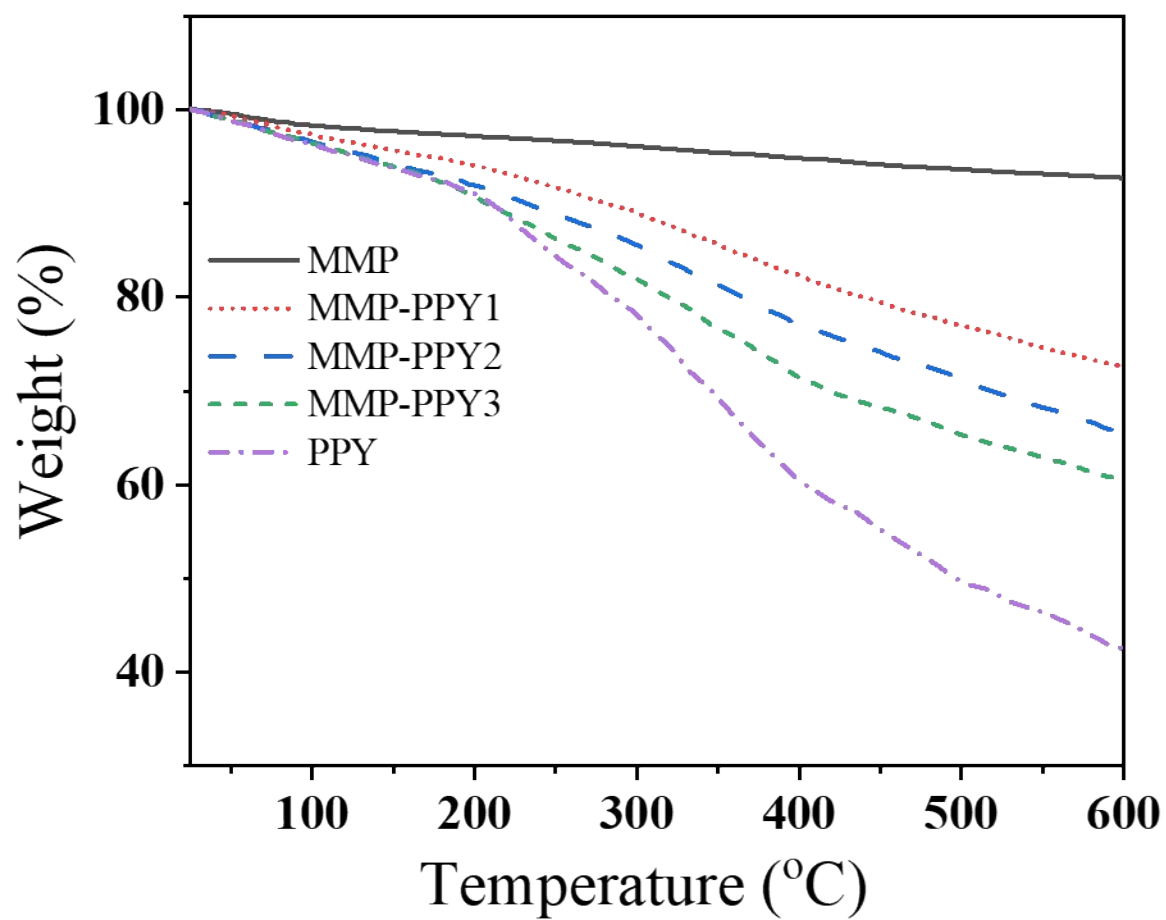


Figure S4: Thermogravimetric analysis (TGA) curves of all the samples.

Figure S5: CV curves at multiple scan rates for MMP-PPY1 and MMP-PPY2.

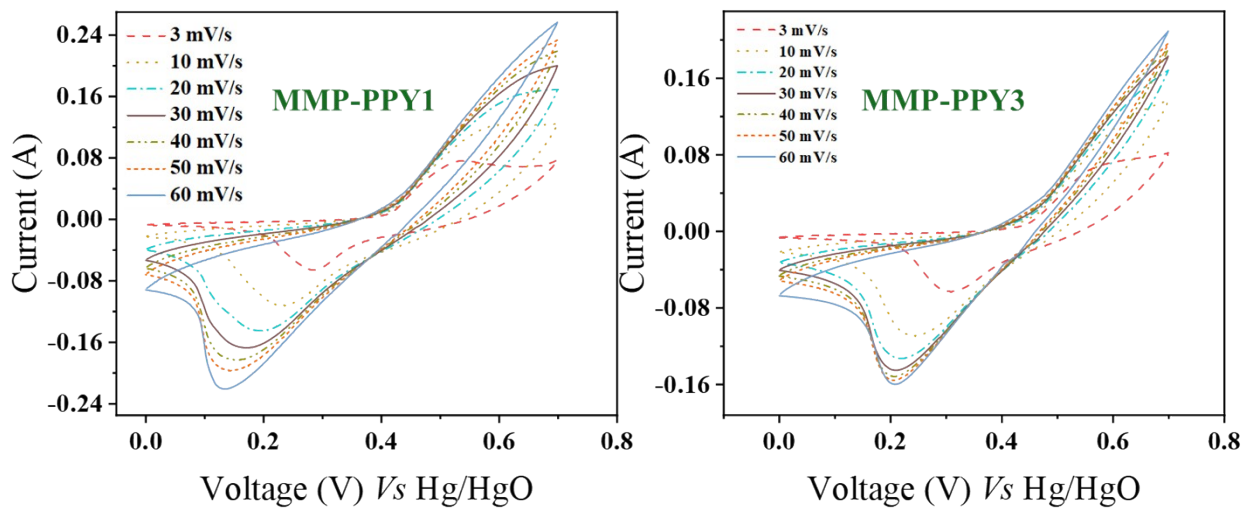
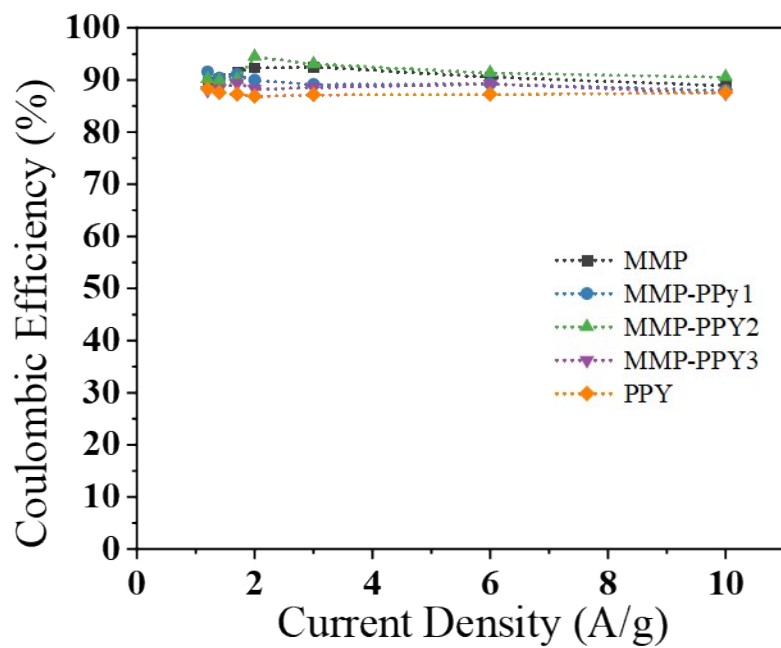


Figure S6: Columbic efficiency of all the samples at multiple scan rates.



Calculations Procedure:

To ensure the optimal performance and reliability of the optimized MMP-PPY2 composite material, the asymmetric supercapacitor device has been fabricated. The masses of the active materials for the positive (MMP-PPY2) and negative (activated carbon) electrodes were carefully balanced based on the principle of charge balance.

$$\frac{m_+}{m_-} = \frac{C_{s-} \times \Delta V_-}{Q_{s+}} \quad (S1)$$

In above equation, m_- and m_+ signifies the active mass of the negative and positive electrode, respectively. Whereas C_{s-} and Q_{s+} depicts the specific capacitance and specific capacity of the negative and positive electrodes in a three-electrode setup. The corresponding PW of negative electrode is displayed by ΔV_- . Through utilizing the aforementioned equation, the active mass of the negative and positive electrodes was obtained as 3.50 and 6.50 mg, respectively.

The specific capacity Q_s (C/g) is calculated using the standard formula

$$\text{(using CV results)} \quad Q_s = \frac{1}{mv} \int I(V) dV \quad (S2)$$

$$\text{(using GCD results)} \quad Q_s = \frac{I \times \Delta t}{m} \quad (S3)$$

The integral part represents the area under the CV curve; m is the total mass of active material in both electrodes (positive + negative) in grams v is the scan rate and Δt is the discharge time in seconds.

Table S1: Calculation of Q_s via CV outcomes.

Sample	Active Mass of Electrode Material (g)	Area Under CV Curve (3 mV/s)	Area Under CV Curve (5 mV/s)	Area Under CV Curve (10 mV/s)	Area Under CV Curve (20 mV/s)	Area Under CV Curve (30 mV/s)	Area Under CV Curve (40 mV/s)	Area Under CV Curve (50 mV/s)
MMP	0.004	0.01428	0.021452	0.034126	0.048557	0.056007	0.062669	0.070119
MMP- PPY1	0.004	0.018544	0.028495	0.046857	0.069351	0.081161	0.091038	0.102705
MMP- PPY2	0.004	0.021461	0.028146	0.037216	0.044641	0.038302	0.037469	0.037609
MMP- PPY3	0.004	0.024762	0.034041	0.052207	0.080041	0.094024	0.105969	0.111375
PPY	0.004	0.010695	0.016045	0.02634	0.039518	0.045219	0.045542	0.042207

Table S2: Calculation electrochemical outcomes for real device.

Current density	Discharge time (s)	Current	Specific	energy	Power
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(A g ⁻¹)		(A)	capacity (C g ⁻¹)	density (Wh Kg ⁻¹)	density (W kg ⁻¹)
1.9	233.7	0.019	444.0	104.9	1615
2.2	183.7	0.022	404.1	95.4	1870
2.5	149.6	0.025	374.0	88.3	2125
3	113	0.030	339.0	80	2550
4	76.6	0.040	306.4	72.3	3400
6	45.4	0.060	272.4	64.4	5100
8	30.8	0.080	246.4	58.2	6800
11	20	0.110	220.0	51.9	9350
14	14.6	0.140	204.4	48.4	11900

Table S3: EIS Fitting parameters.

Sample	Rs (Ω)	Rct (Ω)	CPE n	Warburg σ (Ω·s ^{-1/2})
MMP	1.35	4.00	0.87	1.8
MMP-PPy1 (15%)	1.04	2.20	0.88	1.0
MMP-PPy2 (25%)	1.08	0.88	0.92	0.40
MMP-PPy3 (35%)	1.40	6.00	0.82	2.5