Supplementary Information (SI) for Sustainable Energy & Fuels.
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Supporting Information

Layered Ammonium Metal Phosphate Based Heterostructure with Phosphate-Sulfide Interfacial Synergy for Efficient Oxygen Evolution and Urea Oxidation Reactions

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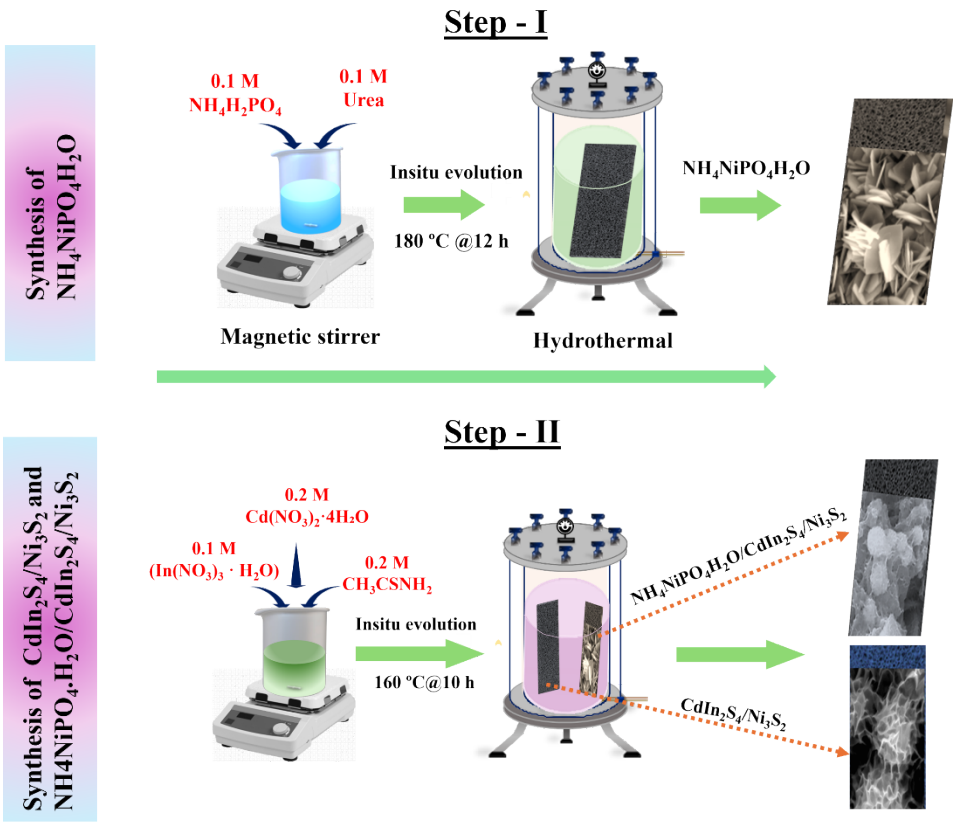
Schematic I: Stepwise synthesis of $NH_4NiPO_4 \cdot H_2O$, $CdIn_2S_4/Ni_3S_2$, and $NH_4NiPO_4 \cdot H_2O/CdIn_2S_4/Ni_3S_2$ heterostructure on Ni foam via a hydrothermal method, showcasing the sequential deposition and integration processes leading to a robust and synergistic heterostructure.



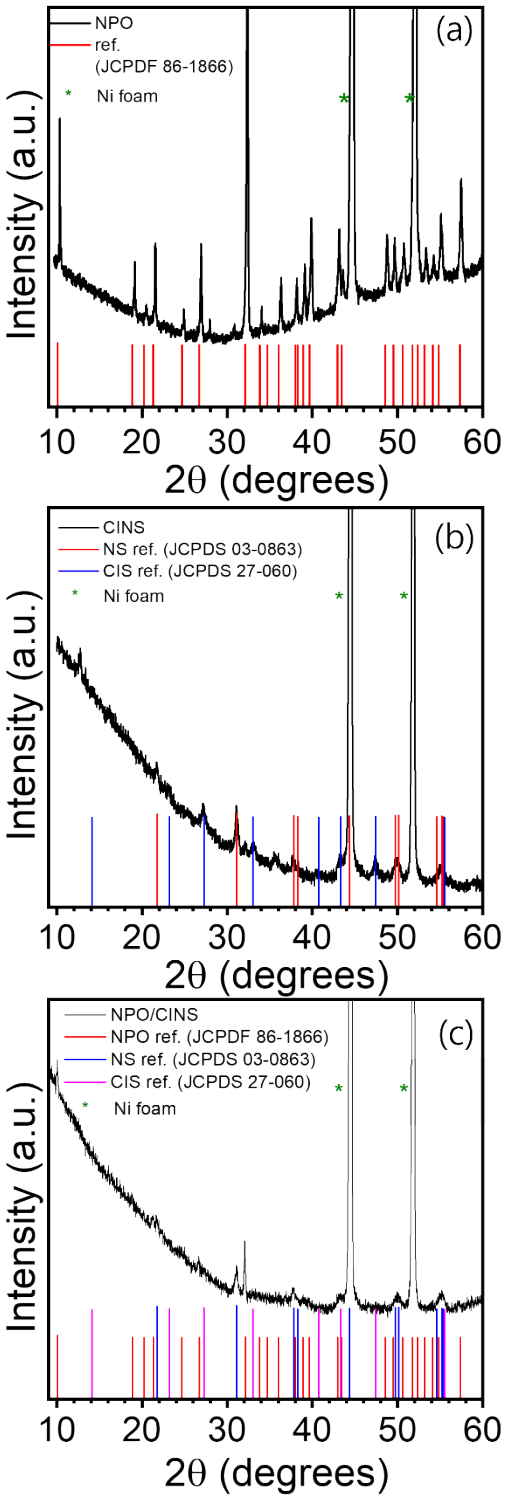
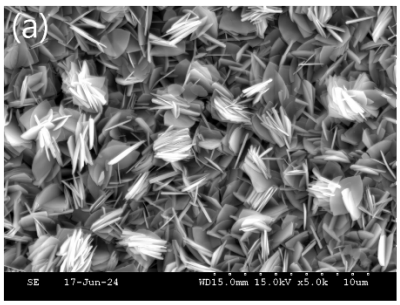
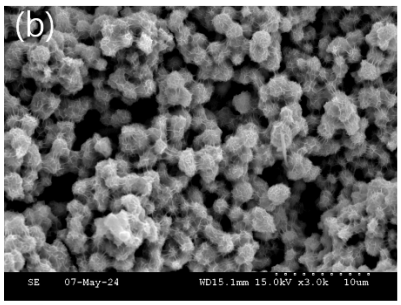
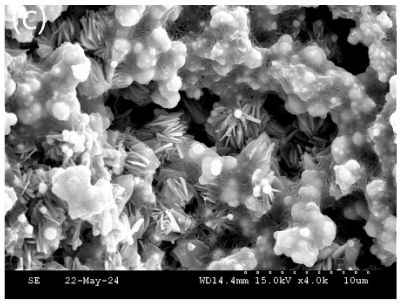


Figure S1 XRD patterns of (a) $NH_4NiPO_4 \cdot H_2O$, (b) $CdIn_2S_4/Ni_3S_4$ heterostructure and (c) $NH_4NiPO_4 \cdot H_2O/CdIn_2S_4/Ni_3S_4$ heterostructure.







(d)

Element	Wt%	Atomic %
N	0.00	0.00
Ni	33.55	16.12
O	38.65	68.15
P	11.02	10.04
Cd	4.27	1.07
In	10.06	2.47
S	2.45	2.15
Total	100	100

Figure S2 FESEM images of (a) $NH_4NiPO_4 \cdot H_2O$, (b) $CdIn_2S_4/Ni_3S_4$ and (c) $NH_4NiPO_4 \cdot H_2O/CdIn_2S_4/Ni_3S_4$ heterostructure. (d) EDS data of $NH_4NiPO_4 \cdot H_2O/CdIn_2S_4/Ni_3S_4$ heterostructure.

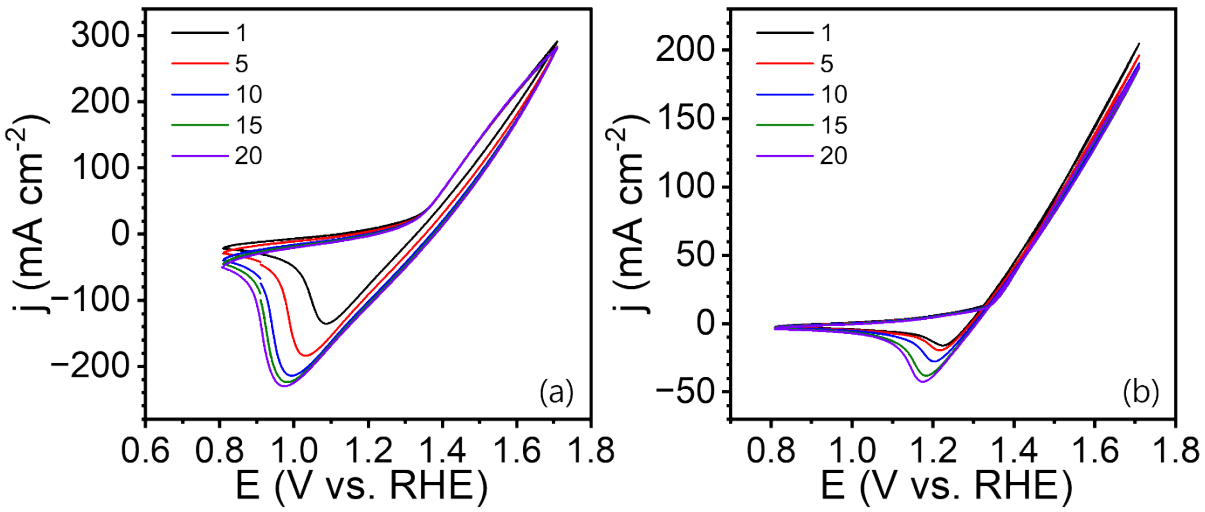


Figure S3 Cyclic voltammetry curves of $NH_4NiPO_4 \cdot H_2O/CdIn_2S_4/Ni_3S_4$ heterostructure at 100 mV/s for (a) OER and (b) UOR.

TOF calculations: The following equation is used to calculate the value of TOF:

$$TOF = (j\ A) / (4\ F\ n) \quad \text{..... (S1)}$$

Here, j (mA cm^{-2}) is the measured current density at a given potential, A is the surface area of the working electrode, 4 is the electron transfer number during O_2 production, and n is the number of moles of the active material and n is the Ni ions molar number calculated from CV curves using equation $n = Q/F$, Q is total charge calculated from the CV curve, F is the Faraday constant (F = 96485 C mol^{-1}).

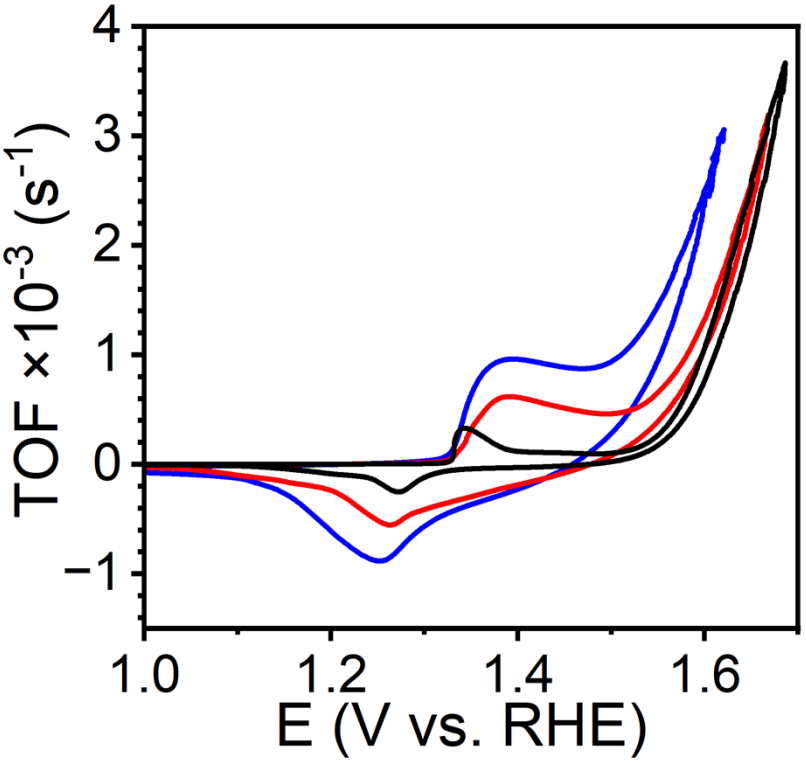
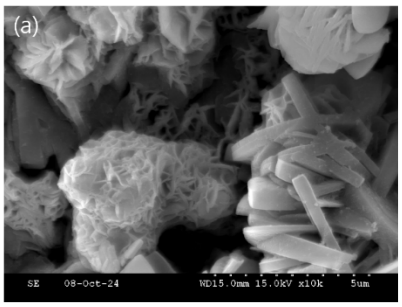
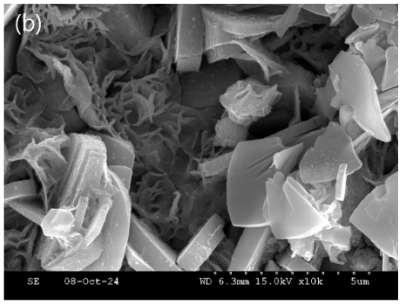


Figure S4 Turnover frequency values (TOF) of (a) $NH_4NiPO_4 \cdot H_2O$, (b) $CdIn_2S_4/Ni_3S_4$ and (c) $NH_4NiPO_4 \cdot H_2O/CdIn_2S_4/Ni_3S_4$ heterostructure. before and after OER and UOR activities.



Spectrum 1		
Element	Wt%	Atomic %
N	0.00	0.00
Ni	54.15	33.22
O	22.01	49.55
P	10.30	11.98
Cd	1.03	0.33
In	11.27	3.54
S	1.23	1.39
Total	100	100



Spectrum 1		
Element	Wt%	Atomic %
N	0.00	0.00
Ni	50.43	25.91
O	36.56	68.92
P	1.23	1.20
Cd	3.81	1.02
In	6.69	1.76
S	1.28	1.20
Total	100	100

Figure S5 FESEM images and EDS data of $NH_4NiPO_4 \cdot H_2O/CdIn_2S_4/Ni_3S_4$ heterostructure after (a) OER and (b) UOR.

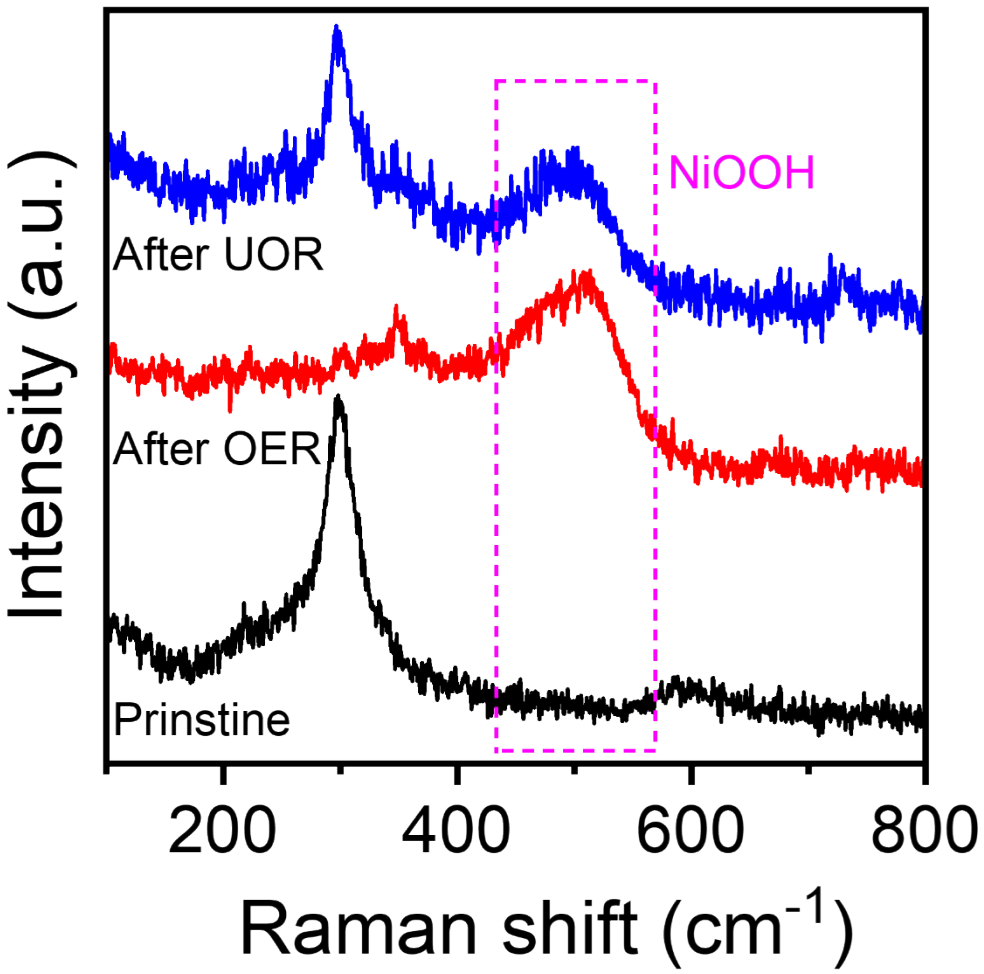


Figure S6. Raman spectra of $NH_4CoPO_4 \cdot H_2O/CdIn_2S_4/Ni_3S_4$ heterostructure before and after OER and UOR activities.

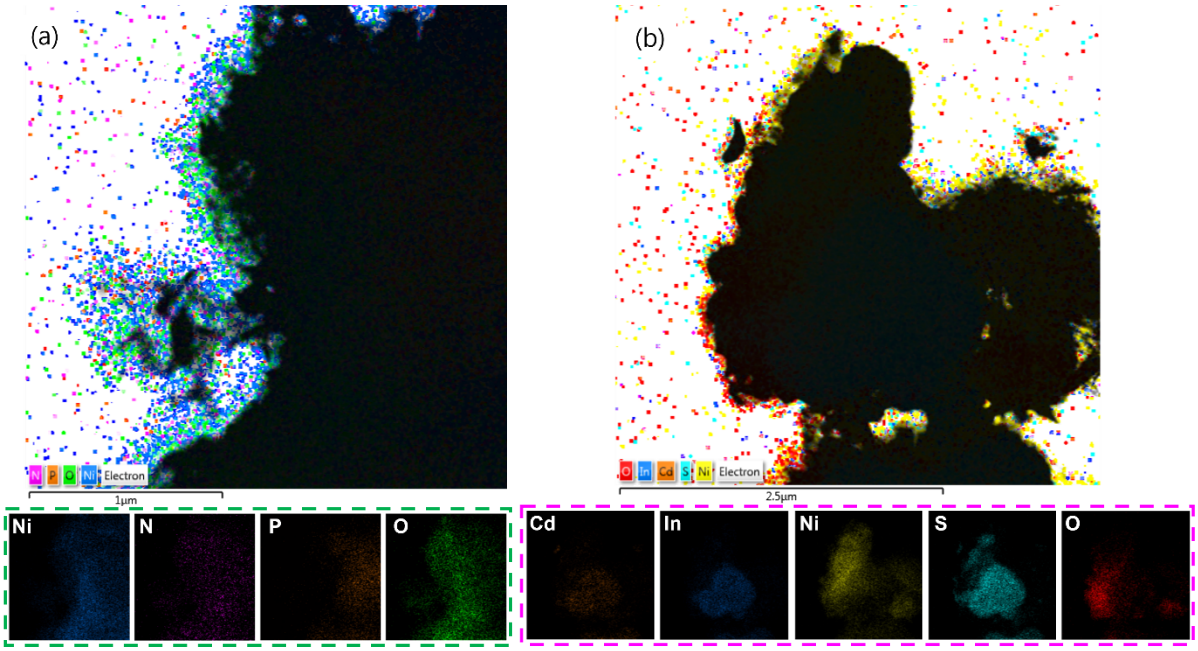


Figure S7. EDX mapping images from TEM of NPO/CINS heterostructure (a) NPO phase and (b) CINS phase.