

**Hollow Bimetallic Selenide Derived from Hierarchical MOF-based Prussian
Blue Analogue for Urea Electrolysis**

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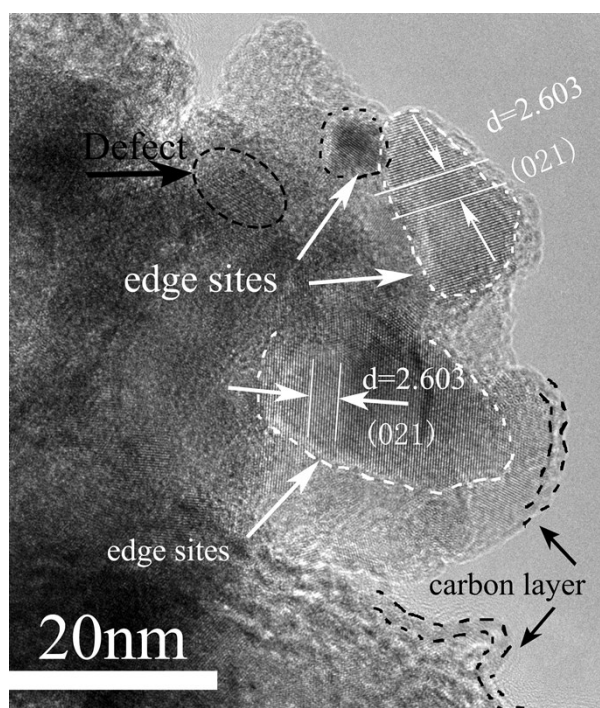


Figure S1. HRTEM image of PBA@MOF-Ni/Se.

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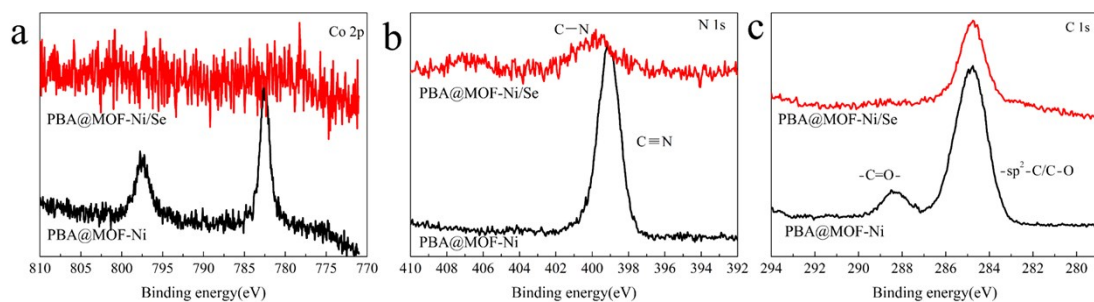


Figure S2. XPS spectra of (a) Co 2p; (b) N 1s; (c) C 1s of PBA@MOF-Ni and PBA@MOF-Ni/Se.

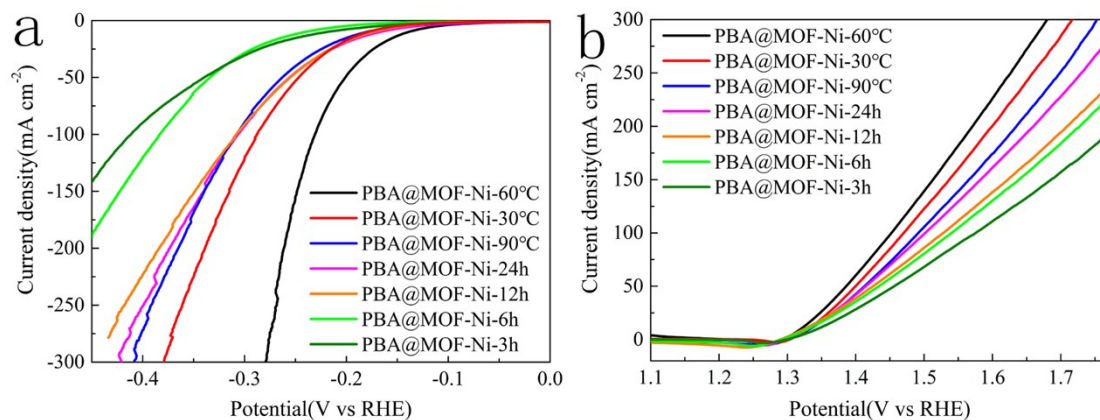


Figure S3. (a) Polarization curves of PBA@MOF-Ni/Se under different reaction conditions towards HER in 1.0 M KOH with 0.5 M urea; (b) Polarization curves of PBA@MOF-Ni/Se under different reaction conditions towards UOR in 1.0 M KOH with 0.5 M urea.

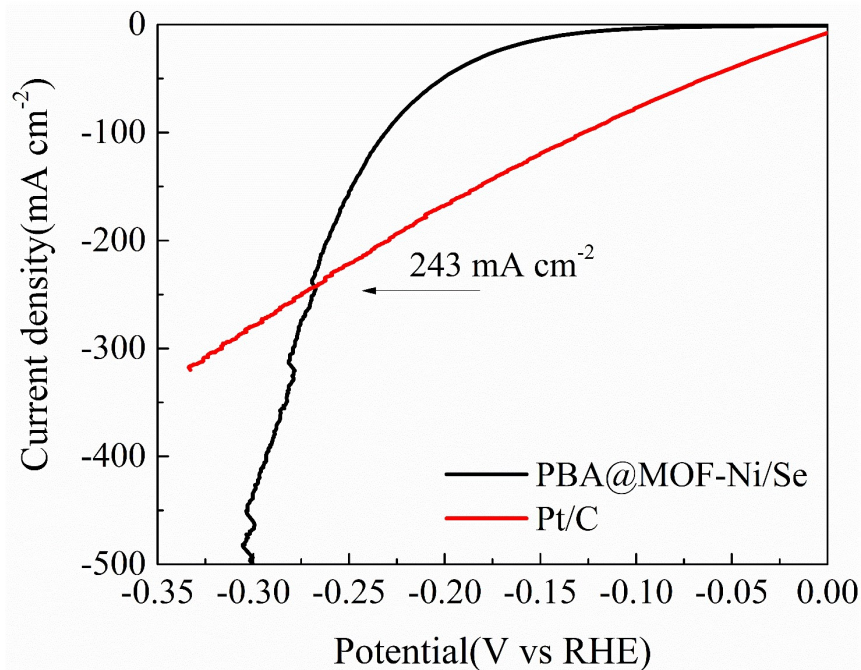


Figure S4. LSV curves towards HER of PBA@MOF-Ni/Se and commercial Pt/C.

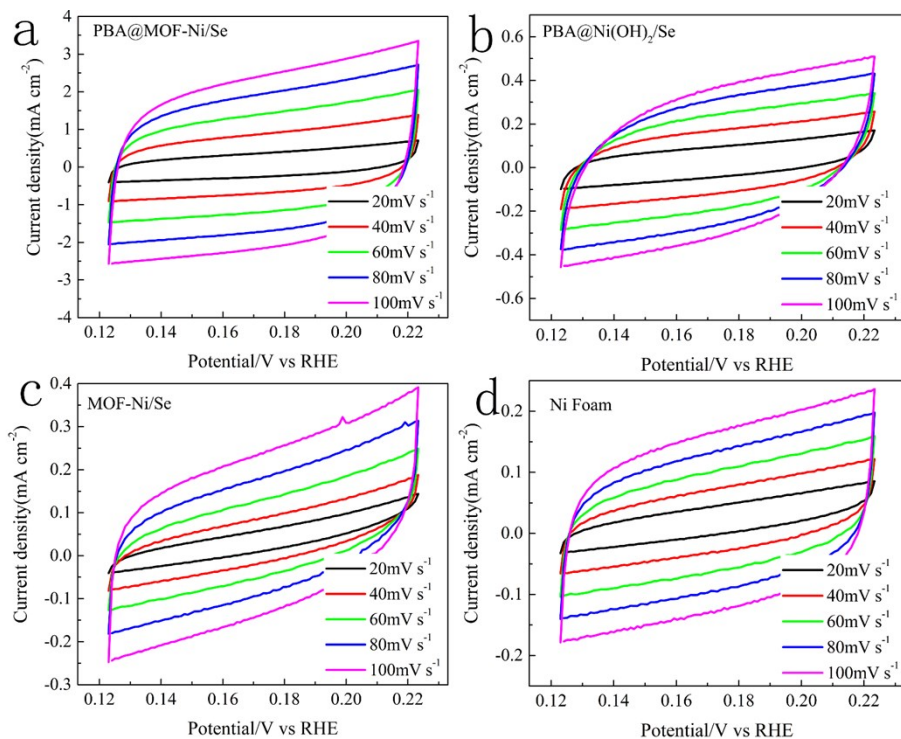


Figure S5. CV curves of (a) PBA@MOF-Ni/Se; (b) PBA@Ni(OH)₂/Se; (c) MOF-Ni/Se; (d) Ni foam; (e) ECSA evaluation towards HER in 1.0 M KOH with 0.5 M urea.

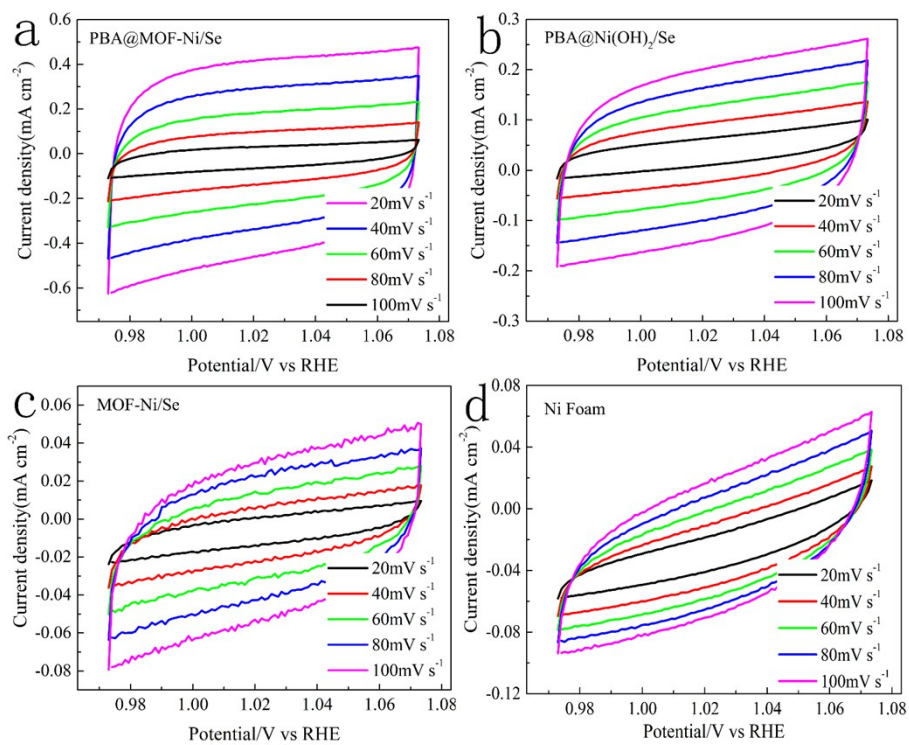


Figure S6. CV curves of (a) PBA@MOF-Ni/Se; (b) PBA@Ni(OH)₂/Se; (c) MOF-Ni/Se; (d) Ni foam; (e) ECSA evaluation towards UOR in 1.0 M KOH with 0.5 M urea.

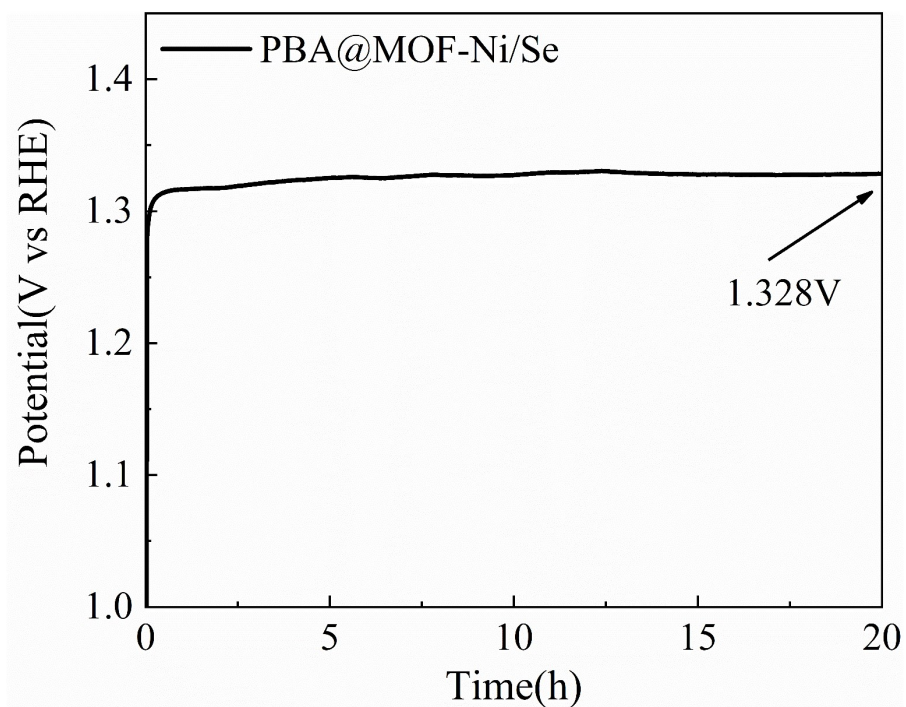


Figure S7. Chronopotentiometry curve of PBA@MOF-Ni/Se towards UOR at 10 mA cm^{-2} for 20h.

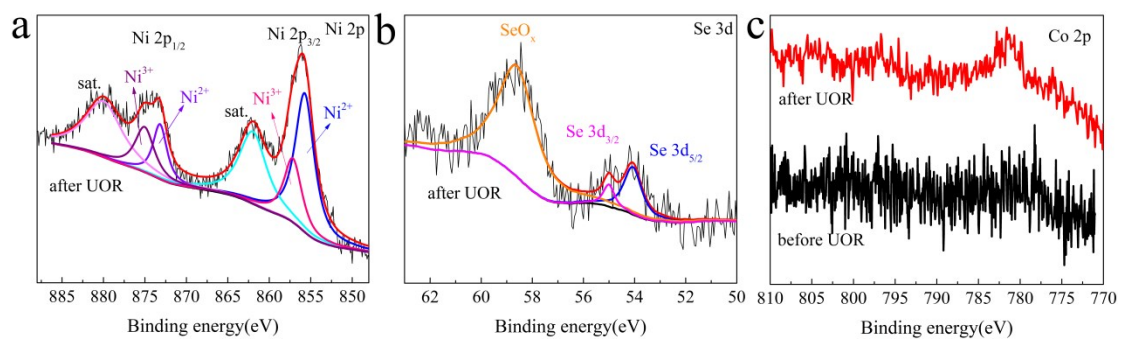


Figure S8. XPS spectra of (a) Ni 2p (b) Se 3d and (c) Co 2p of PBA@MOF-Ni/Se before and after UOR.

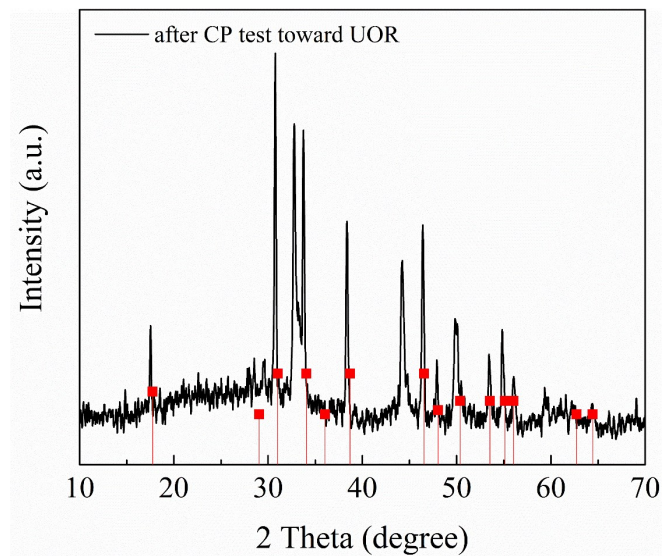


Figure S9. XRD pattern of PBA@MOF-Ni/Se after CP test towards UOR.

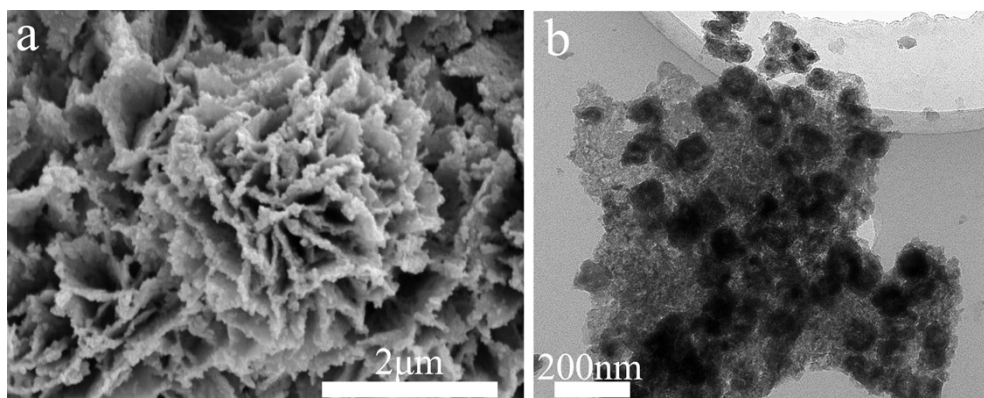


Figure S10. (a) SEM image of PBA@MOF-Ni/Se after CP test towards UOR; (b) TEM image of PBA@MOF-Ni/Se after CP test towards UOR.

Table S1. Comparison of the electrocatalytic activity of PBA@MOF-Ni/Se towards UOR in alkaline media with catalysts reported previously.

Electrocatalysts	Electrolyte	Potential (V vs. RHE) at 10 mA cm ⁻²	Ref
PBA@MOF-Ni/Se	1.0 M KOH 0.5 M Urea	1.319 V	This work
CoS ₂	1.0 M KOH 0.3 M Urea	1.4 V	1
Ni ₂ P/Fe ₂ P	1.0 M KOH 0.5 M Urea	1.36 V	2
Ni/NiO	1.0 M KOH 0.33 M Urea	1.33 V	3
MnO ₂ /MnCo ₂ O ₄	1.0 M KOH 0.5 M Urea	1.33 V	4
NiMoO ₄ ·H ₂ O	1.0 M KOH 0.5 M Urea	1.35 V	5
Ni MOF	1.0 M KOH 1.0 M Urea	1.36 V	6

HC-NiMoS/Ti	1.0 M KOH 0.5 M Urea	1.38 V	7
NF-G-Mn	1.0 M KOH 0.5 M Urea	1.33 V	8
CoN NF/NF	1.0 M KOH 0.5 M Urea	1.342 V	9
Ni _(10%) Pd _(10%) /OMC	1.0 M KOH 0.33 M Urea	1.346 V	10

Table S2. Comparison of the electrocatalytic performance of PBA@MOF-Ni/Se || PBA@MOF-Ni/Se towards overall urea electrolysis in alkaline media with catalysts reported previously.

Catalyst	Electrolyte	Cell voltage at 10 mA cm ⁻²	Ref
PBA@MOF-Ni/Se	1.0 M KOH 0.5 M Urea	1.495 V	This work
CoS ₂	1.0 M KOH 0.3 M Urea	1.59 V	1
MnO ₂ /MnCo ₂ O ₄	1.0 M KOH 0.5 M Urea	1.55 V	4
HC-NiMoS/Ti	1.0 M KOH 0.5 M Urea	1.59 V	7
NF-Pt/C	1.0 M KOH 0.5 M Urea	1.63 V	11
NF-Pt/C NF-IrO ₂	1.0 M KOH 0.5 M Urea	1.72 V	11
NiMoS	1.0 M KOH 0.5 M Urea	1.59 V	12

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