

## Supporting information

### Regulating the Layered Structure of 2D FePS<sub>3</sub> to Achieve High-performance Electrocatalytic Urea Degradation

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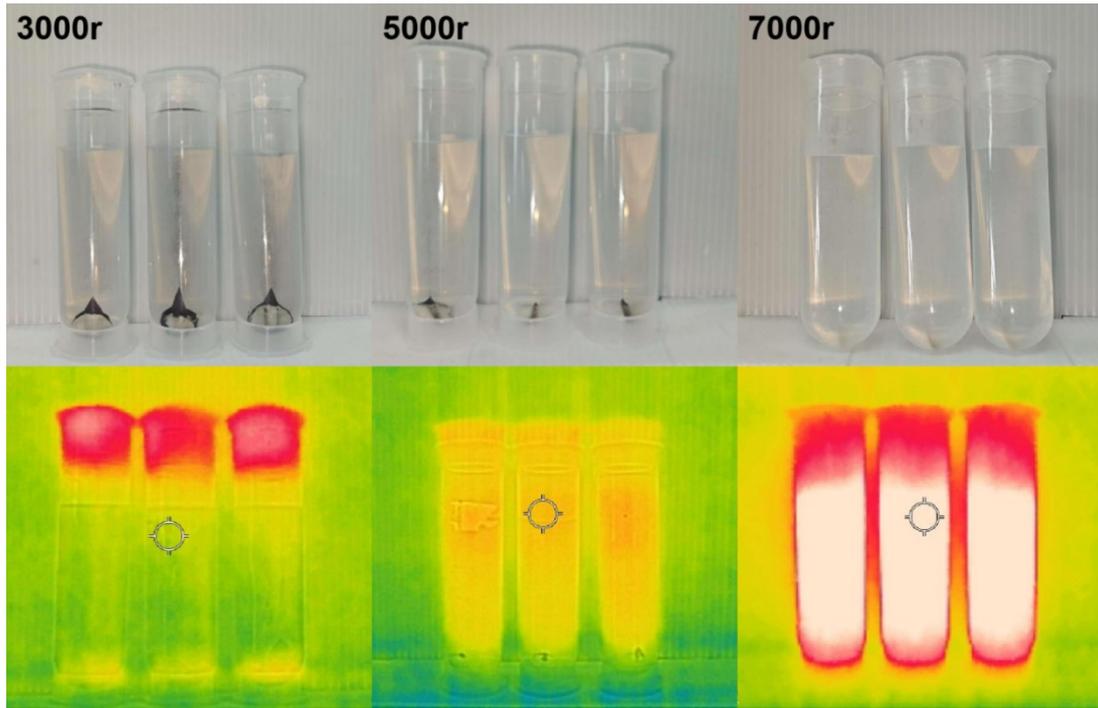
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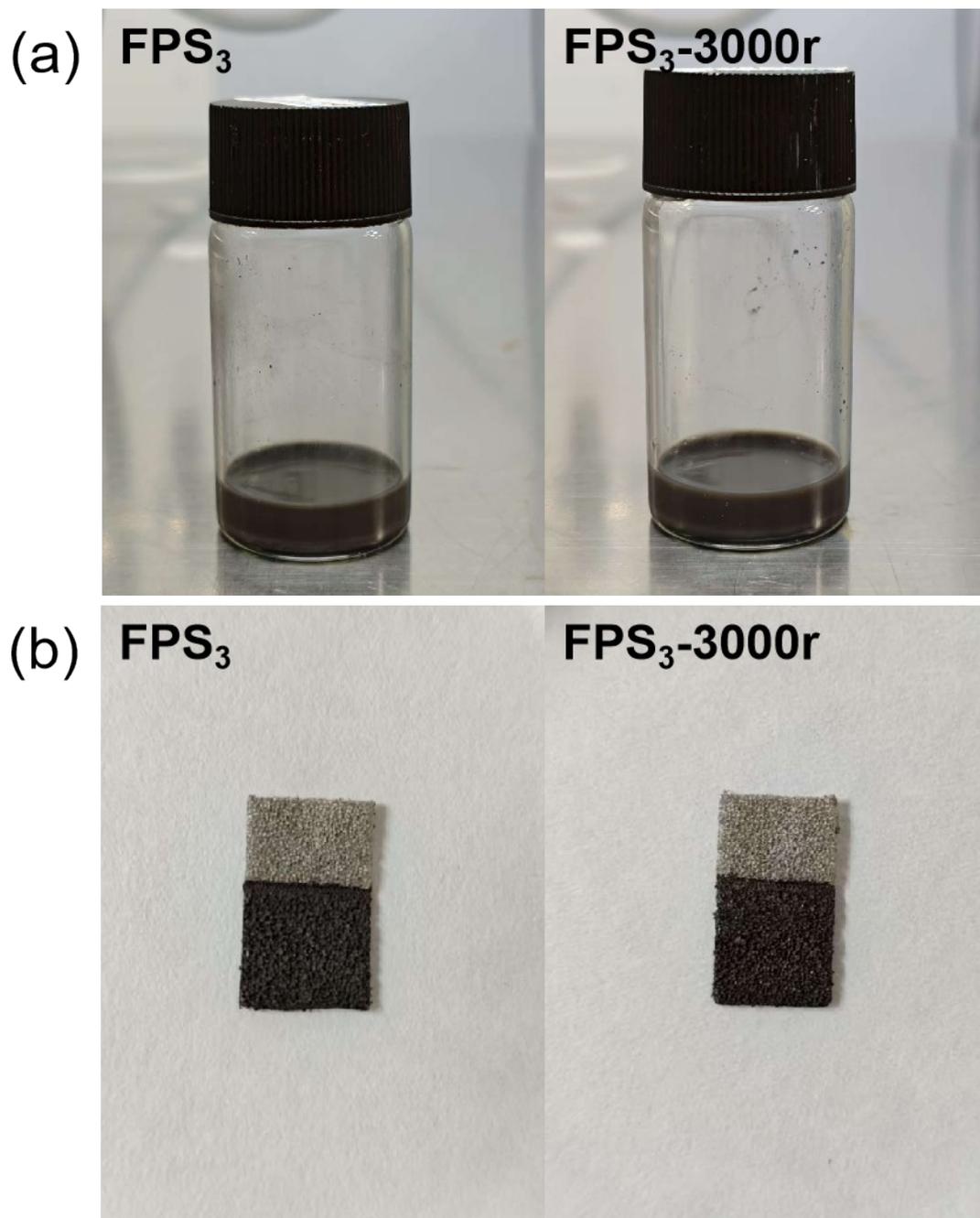
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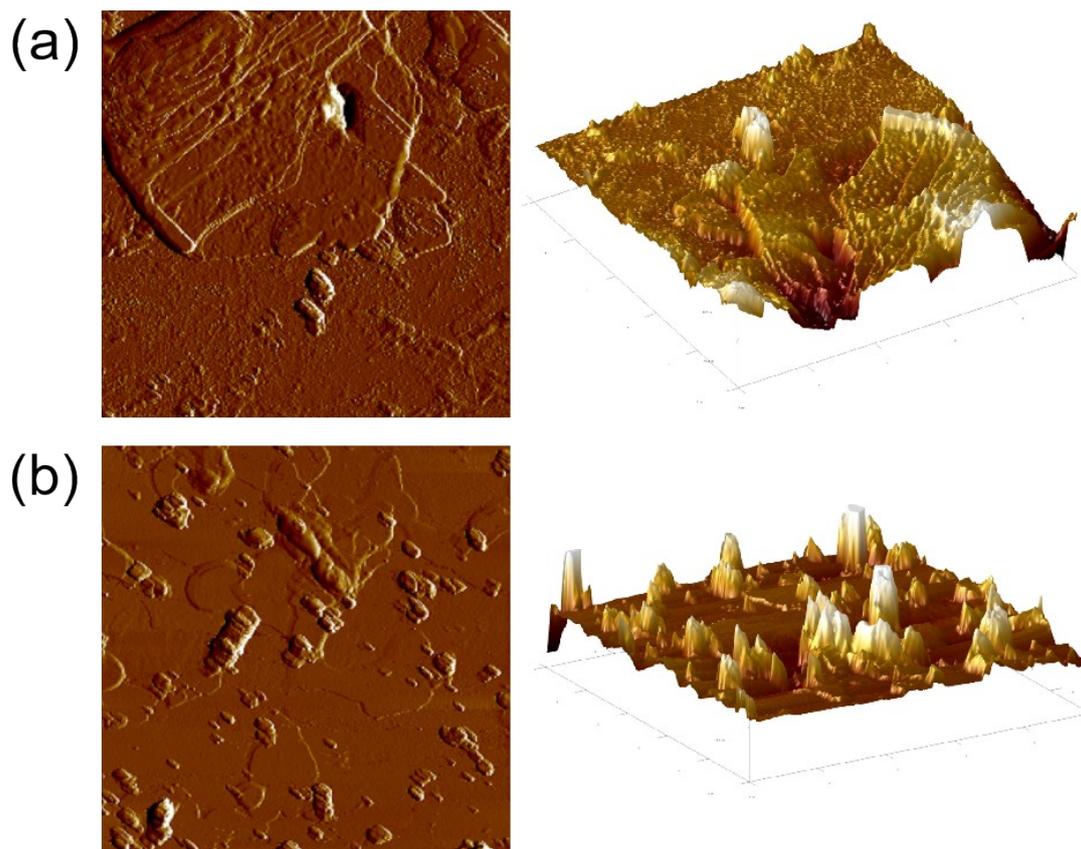
**Figure. S1** Digital and thermal infrared images of the fractions obtained by differential centrifugation.



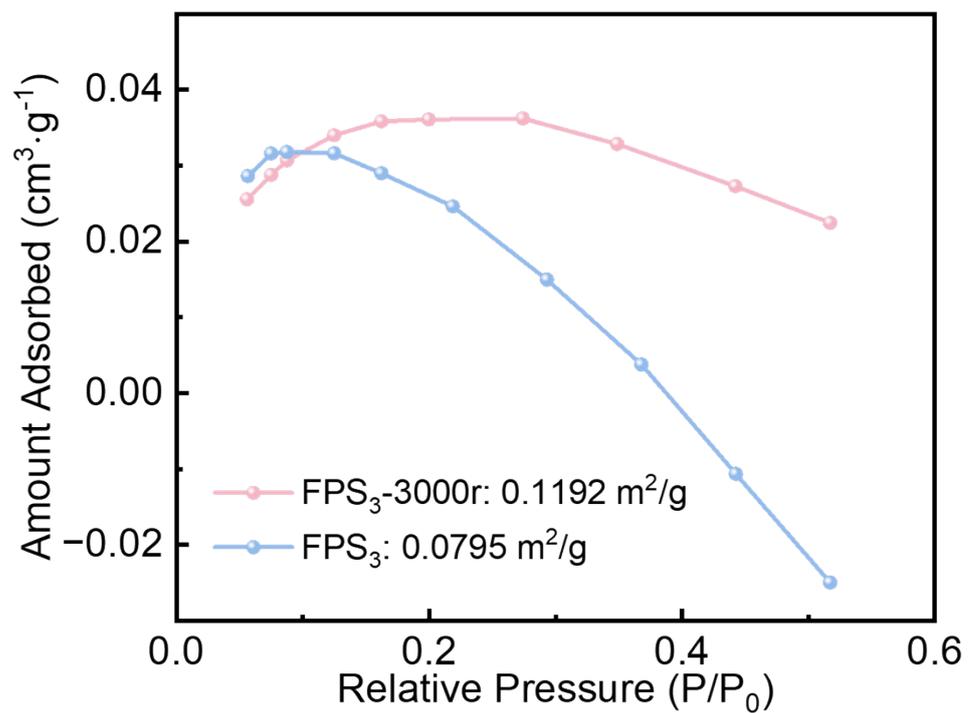
**Figure. S2** Digital photograph of the  $\text{FPS}_3$ ,  $\text{FPS}_3\text{-3000r}$ ,  $\text{FPS}_3\text{-5000r}$ , and  $\text{FPS}_3\text{-7000r}$  catalyst powders.



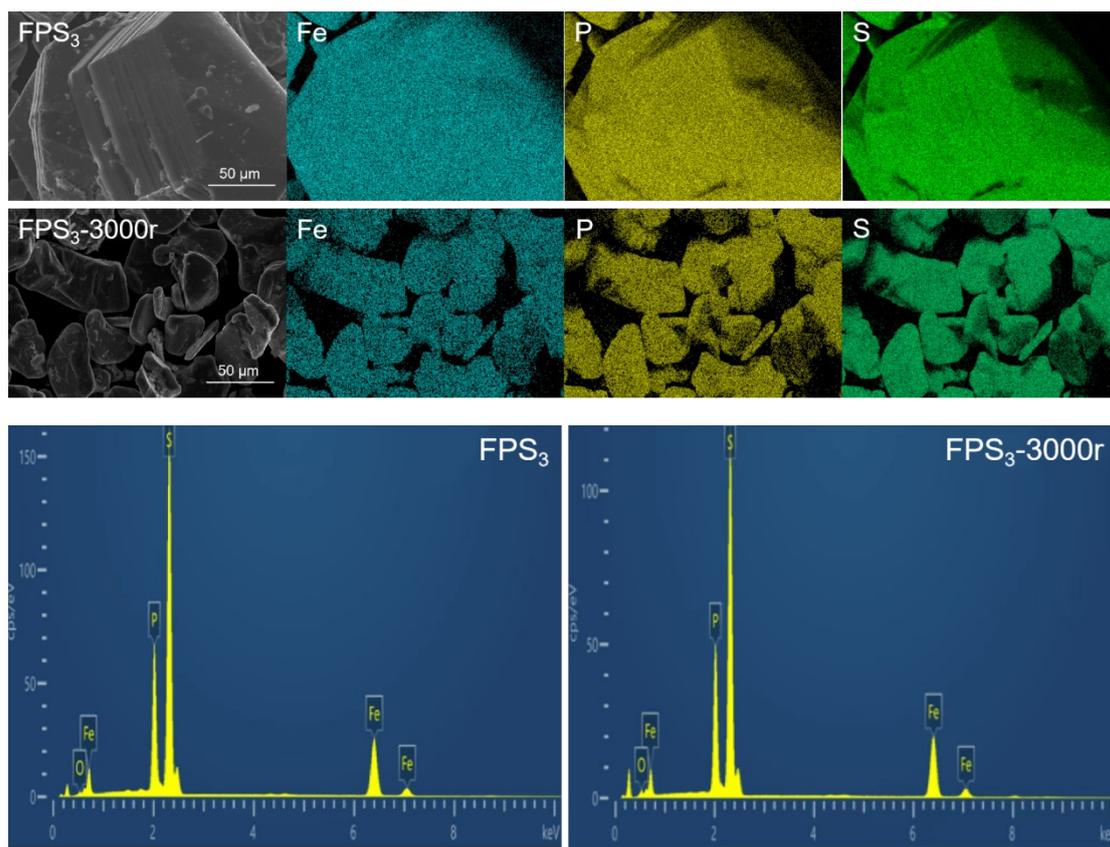
**Figure. S3** Digital photographs of (a)  $\text{FPS}_3$  and  $\text{FPS}_3\text{-3000r}$  catalyst ink, (b)  $\text{FPS}_3$  and  $\text{FPS}_3\text{-3000r}$  catalytic electrodes.



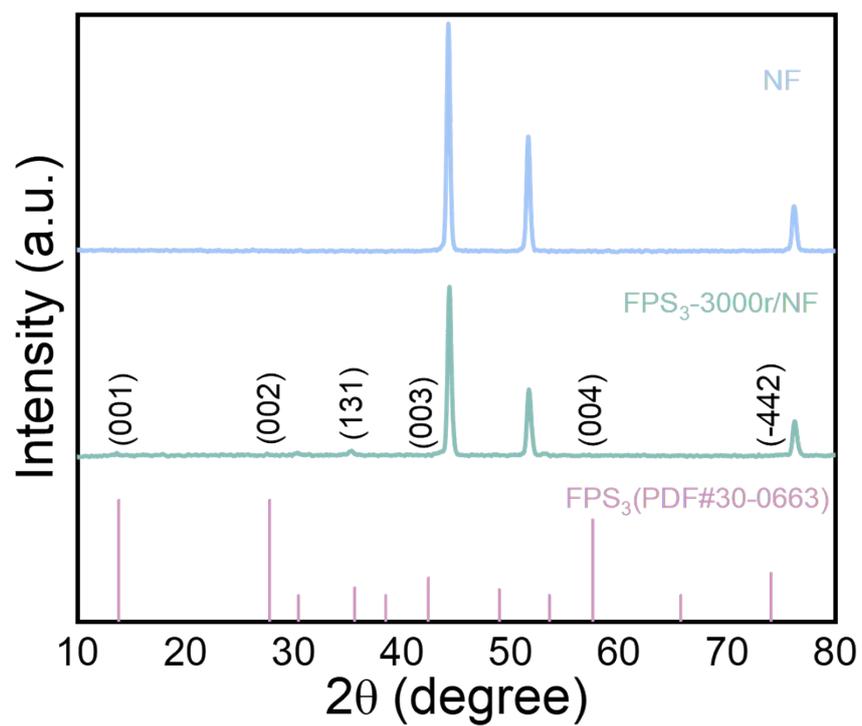
**Figure. S4** AFM images: (a) 2D planar view and 3D topography of FPS<sub>3</sub>, (b) 2D planar view and 3D topography of FPS<sub>3</sub>-3000r.



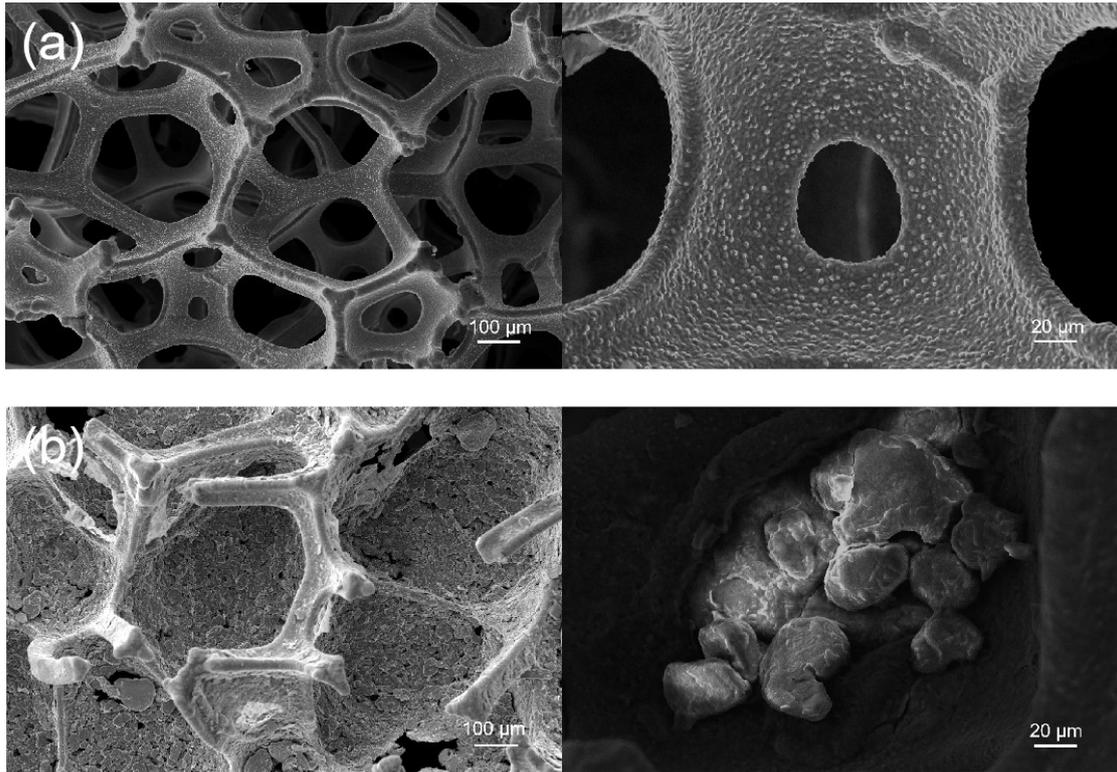
**Figure. S5** Adsorption isotherms and specific surface areas of FPS<sub>3</sub> and FPS<sub>3</sub>-3000r catalysts.



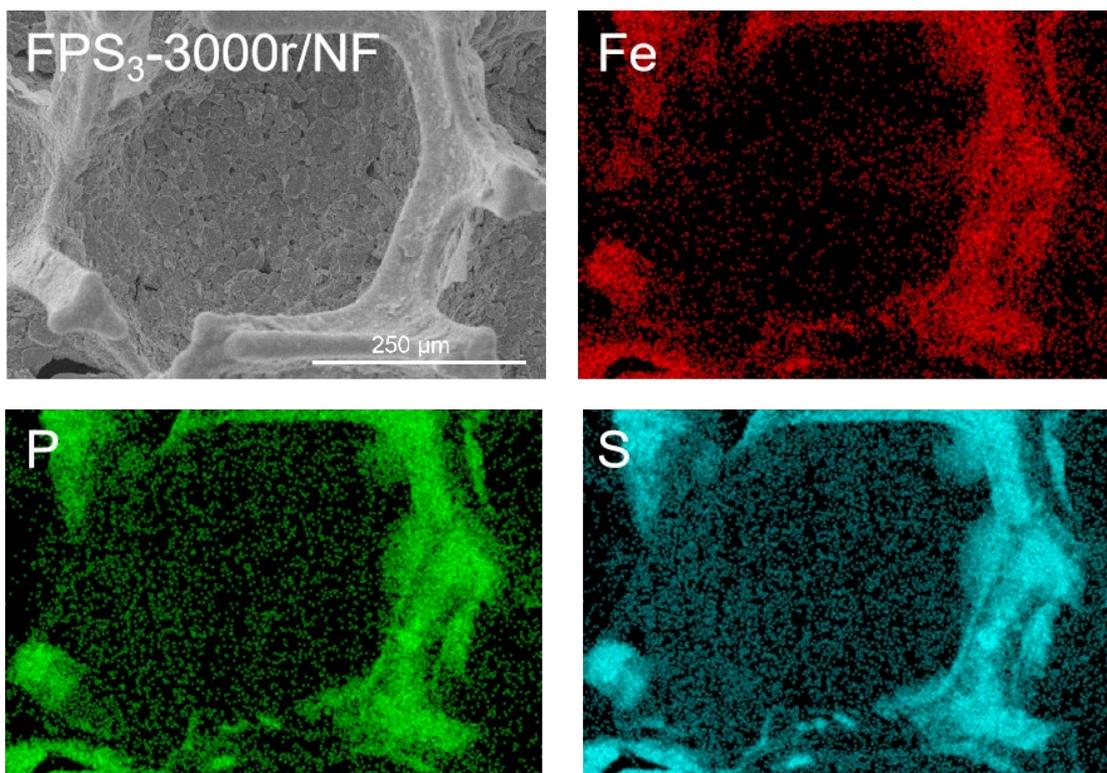
**Figure. S6** EDS element mapping and element distribution maps of FPS<sub>3</sub> and FPS<sub>3</sub>-3000r.



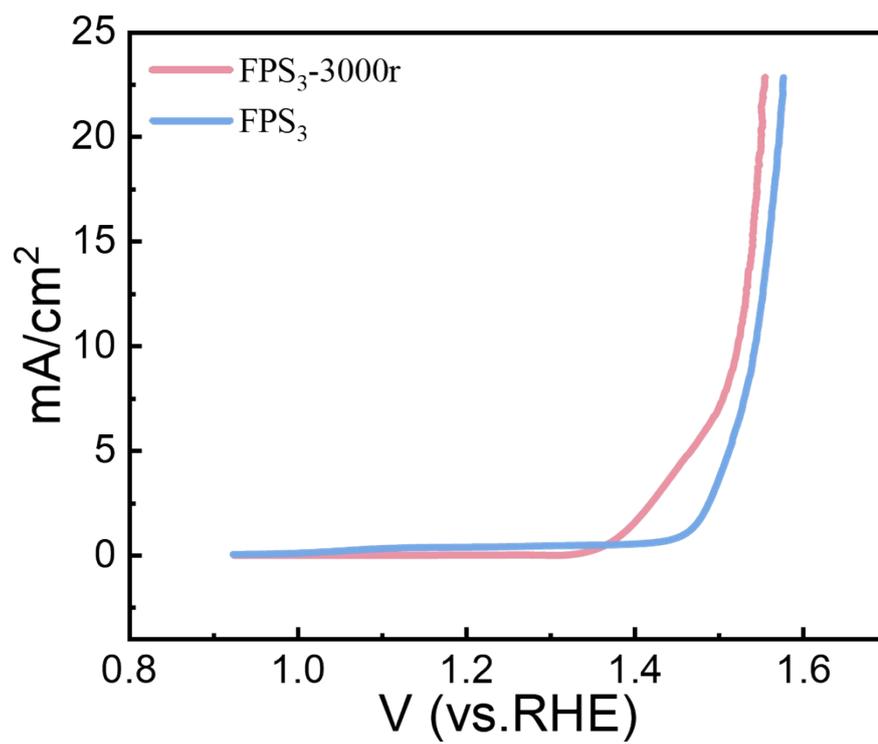
**Figure. S7** XRD patterns of the NF and FPS<sub>3</sub>-3000r/NF catalyst electrodes.



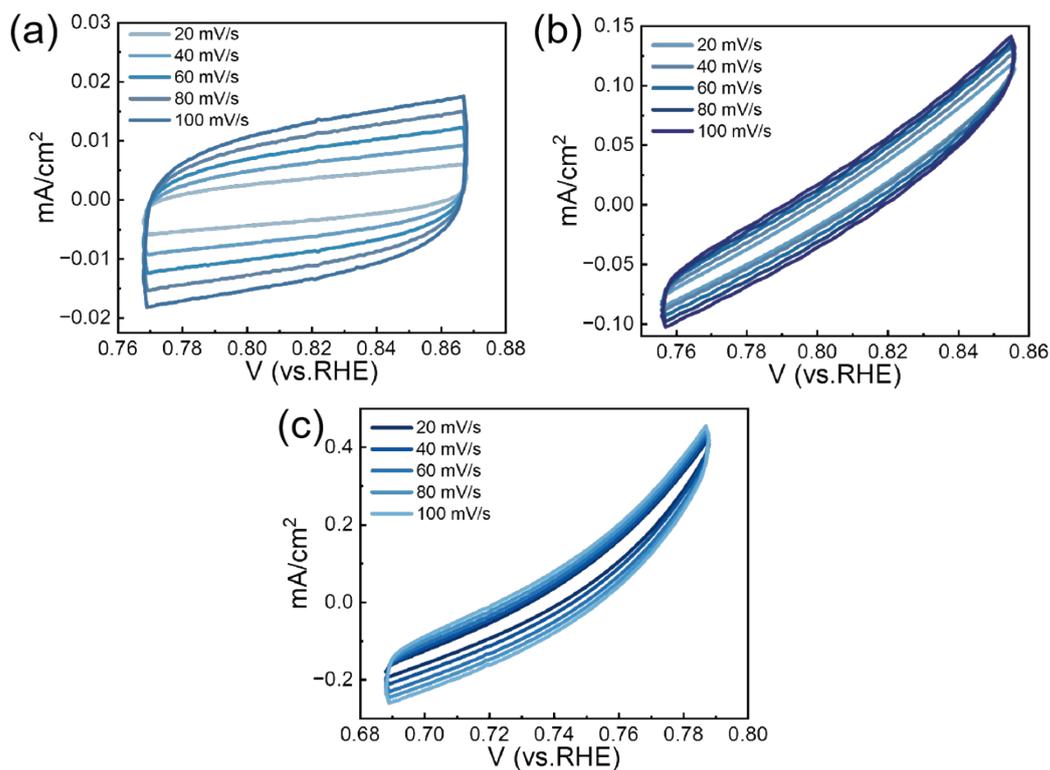
**Figure. S8** SEM image of (a) NF, (b) FPS<sub>3</sub>-3000r/NF.



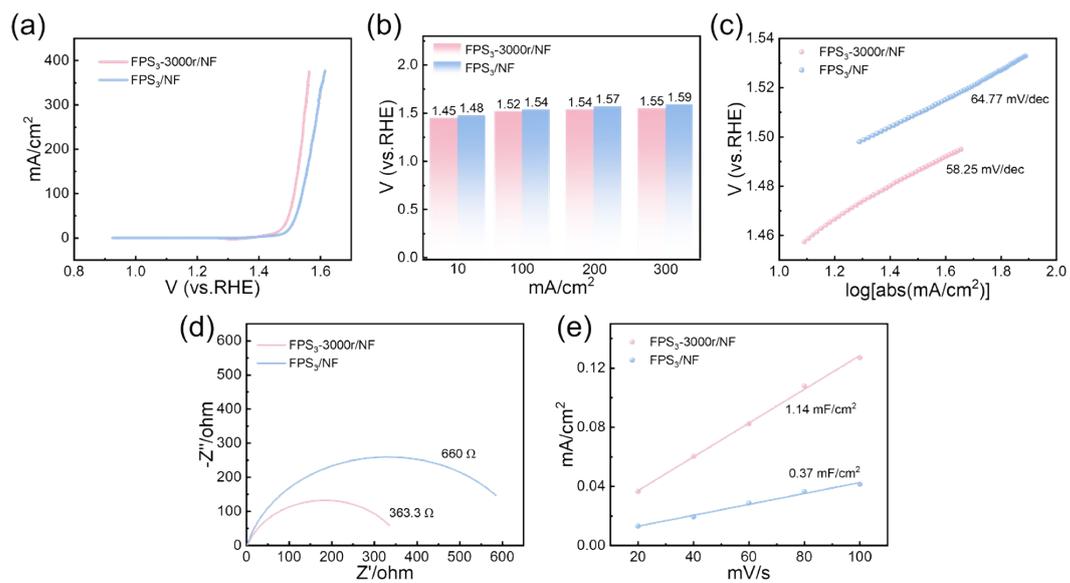
**Figure. S9** EDS elemental mapping of FPS<sub>3</sub>-3000r/NF.



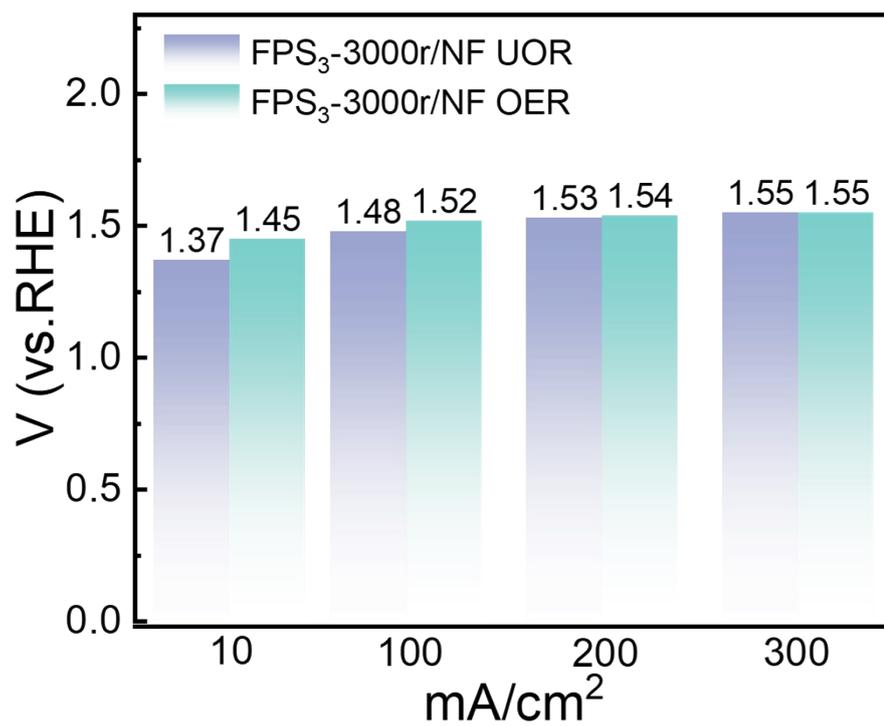
**Figure. S10** LSV curves normalized to the electrochemically active surface area (ECSA).



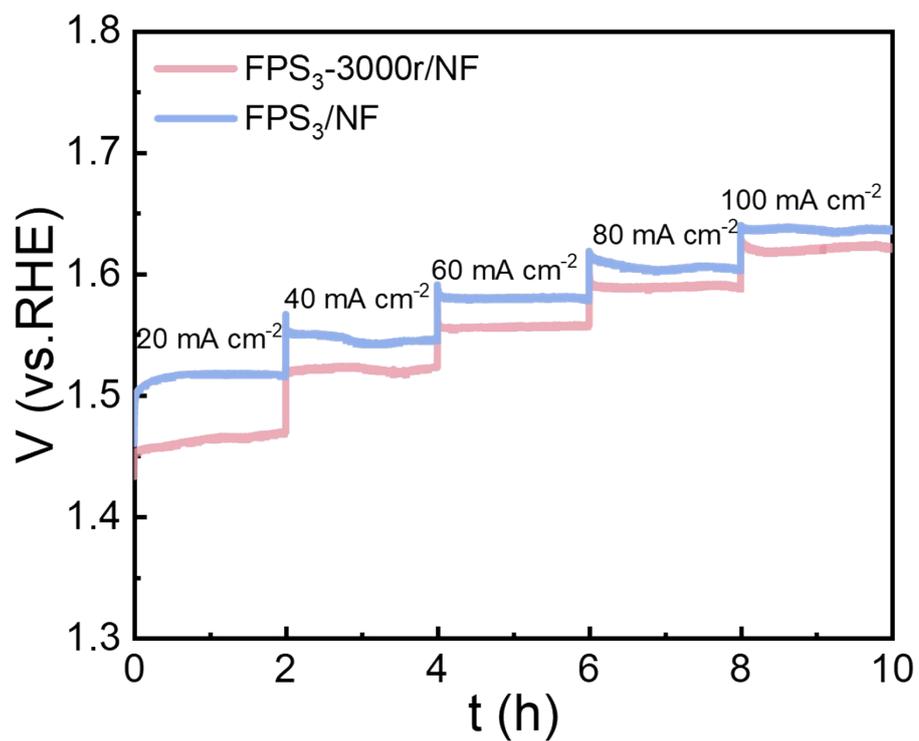
**Figure. S11** CV curves measured at different scan rates within the non-Faradaic region are presented: (a) NF, (b) FPS<sub>3</sub>/NF, and (c) FPS<sub>3</sub>-3000r/NF.



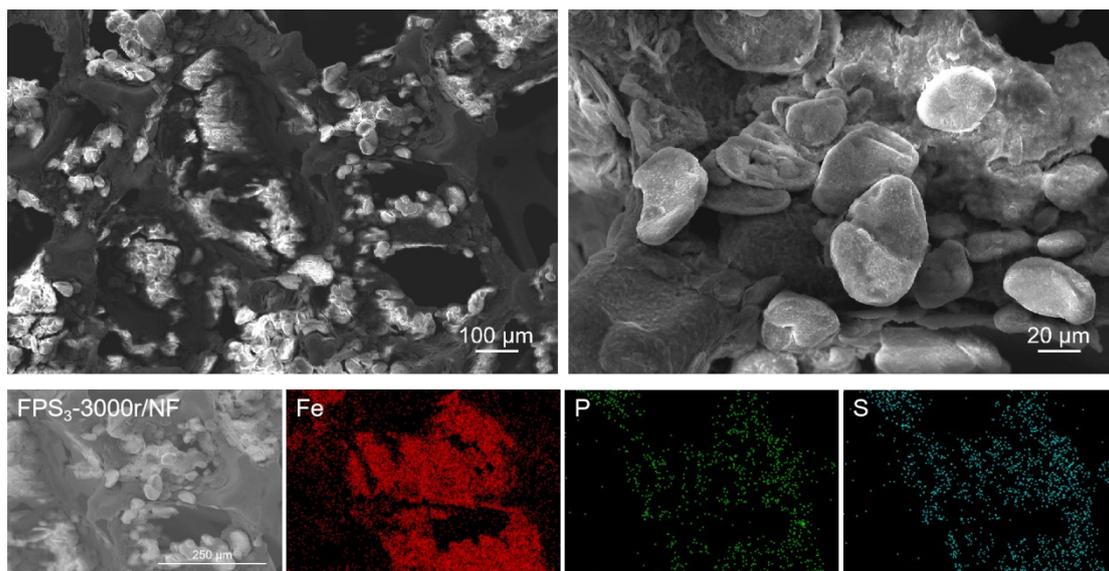
**Figure. S12** OER electrochemical performance of FPS<sub>3</sub>-3000r/NF and FPS<sub>3</sub>/NF: (a) LSV curves, (b) Current density-potential plot, (c) Tafel slopes, (d) EIS fitting curves under open circuit potential, (e) C<sub>dl</sub> determination.



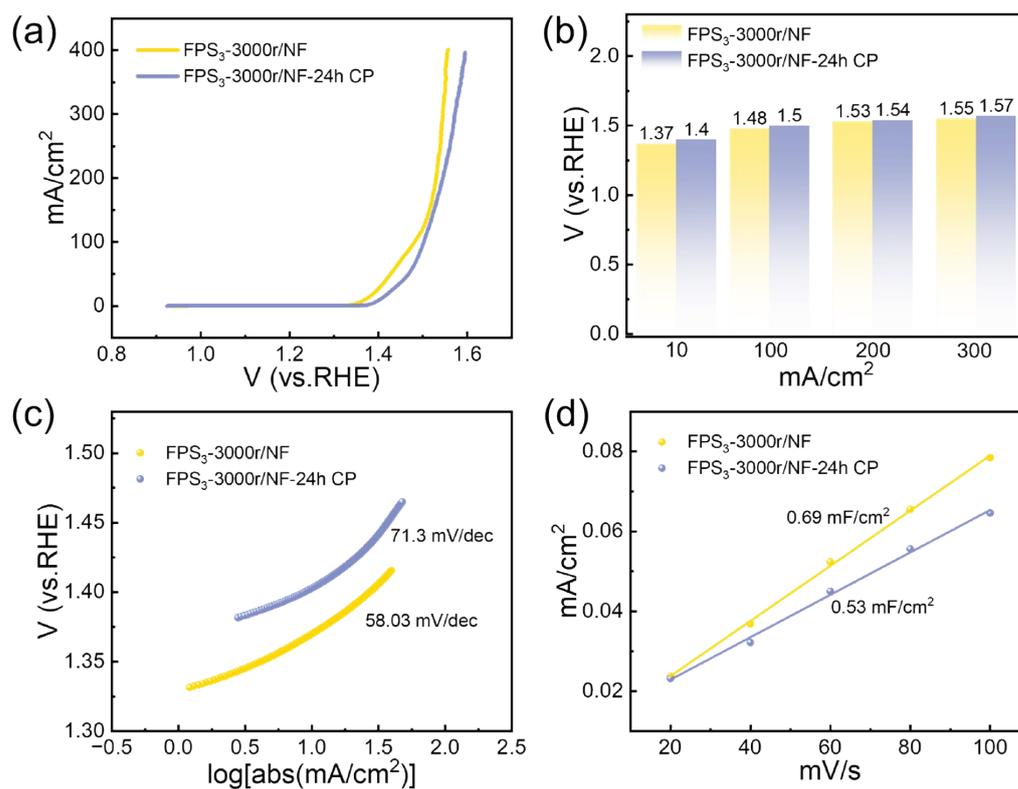
**Figure. S13** Comparison of OER and UOR performance of FPS<sub>3</sub>-3000r/NF electrode under different current densities.



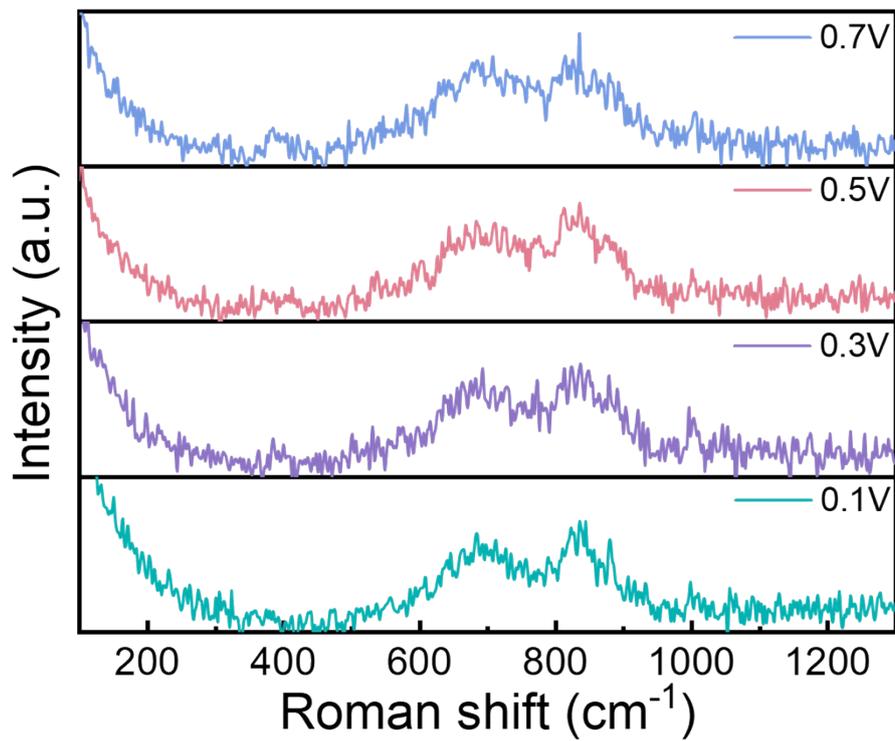
**Figure. S14** Stepped chronopotentiometry test of FPS<sub>3</sub>/NF and FPS<sub>3</sub>-3000r/NF.



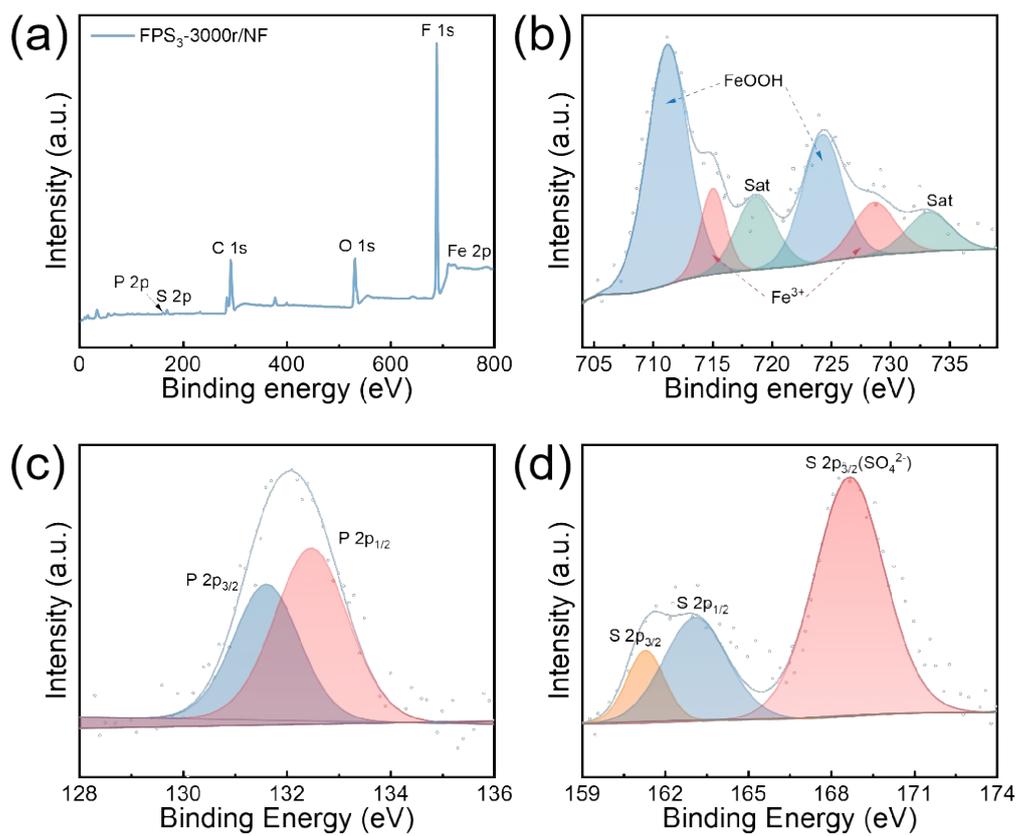
**Figure. S15** Post-reaction of FPS<sub>3</sub>-3000r/NF: SEM image and corresponding EDS elemental mapping.



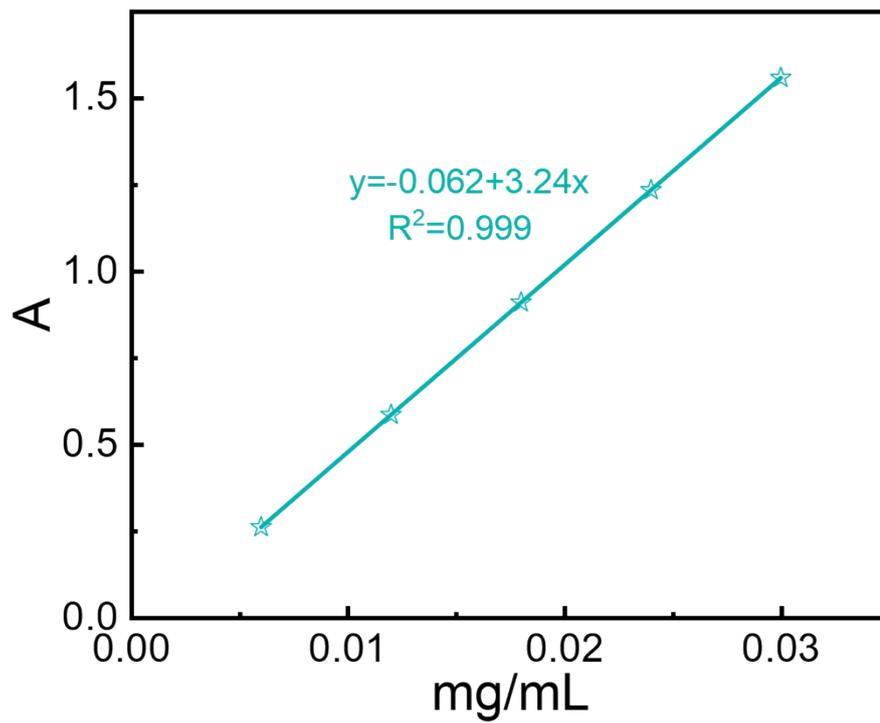
**Figure. S16** Comparison of the electrochemical performance before and after stability testing: (a) LSV curves, (b) Current density-potential plot, (c) Tafel slopes, (d) C<sub>dl</sub> determination



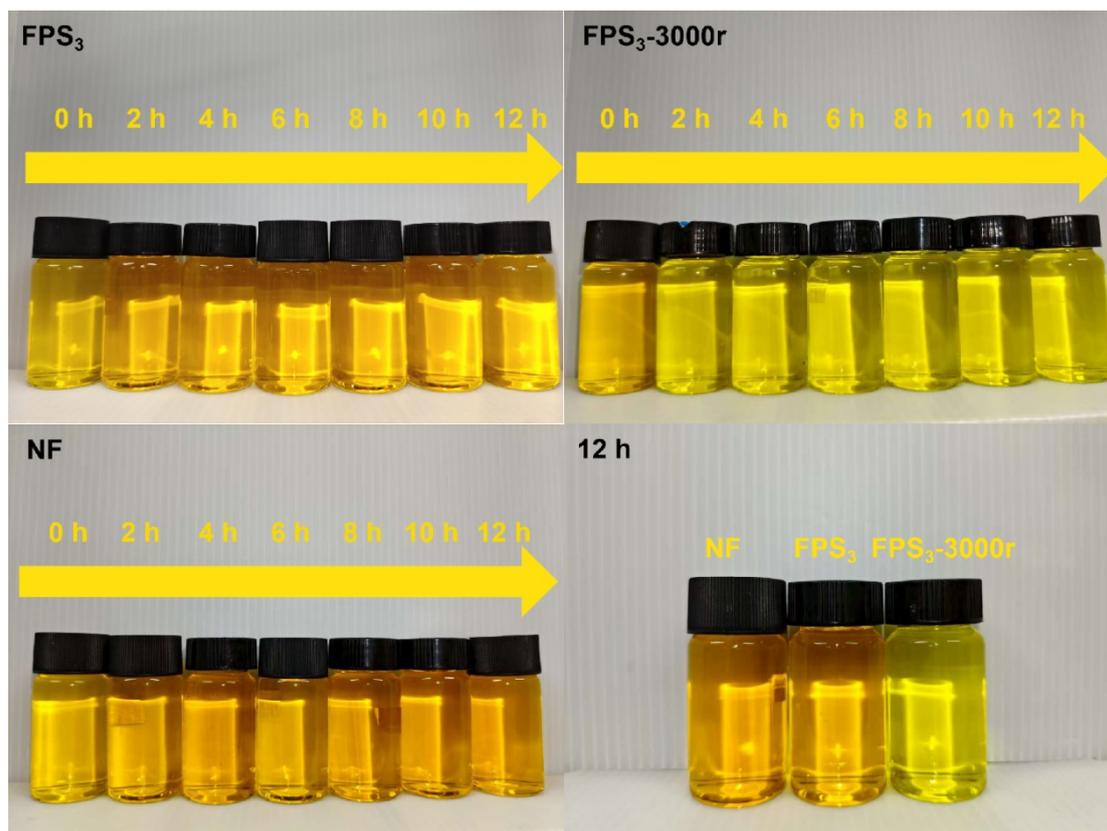
**Figure. S17** In situ Raman spectra of the FPS<sub>3</sub>-3000r/NF electrode.



**Figure. S18** post-reaction XPS spectra of the FPS<sub>3</sub>-3000r/NF: (a) Full spectra, (b) Fe, (c) P, and (d) S.



**Figure. S19** Calibration curve for urea quantification.



**Figure. S20** Digital photograph of time-tracked color evolution of the electrolyte.

**Table. S1** Elemental content of the FPS<sub>3</sub> and FPS<sub>3</sub>-3000r catalyst powders.

FPS <sub>3</sub>			FPS <sub>3</sub> -3000r		
element	Wt%	At%	element	Wt%	At%
O	1.34	3.04	O	2.81	6.31
P	16.44	19.30	P	16.07	18.64
S	49.84	56.56	S	47.89	53.67
Fe	32.38	21.09	Fe	33.23	21.38
gross	100.00	100.00	gross	100.00	100.00

**Table. S2** UOR activity parameter for various electrocatalysts.

<b>catalyst</b>	<b>electrolyte</b>	<b>Current density (mA/cm<sup>2</sup>)</b>	<b>Cell voltage V (vs. RHE)</b>	<b>Ref</b>
Ni <sub>9</sub> S <sub>8</sub> /CuS/Cu <sub>2</sub> O	1.0 M KOH + 0.33 M urea	10	1.47	1
Mo-Co-S-Se/CC	1.0 M KOH + 0.33 M urea	10	1.4	2
V-FeNi <sub>3</sub> N/Ni <sub>3</sub> N	1.0 M KOH + 0.33 M urea	10	1.46	3
CuCo <sub>2</sub> O <sub>4</sub>	1.0 M KOH + 0.33 M urea	10	1.45	4
NiCo <sub>2</sub> S <sub>4</sub> /CC	1.0 M KOH + 0.33 M urea	10	1.45	5
N-NiS/NiS <sub>2</sub>	1.0 M KOH + 0.33 M urea	10	1.62	6
NiSe <sub>2</sub> -NiO 350	1.0 M KOH + 0.33 M urea	10	1.39	7
Ni <sub>3</sub> N NA/CC	1.0 M KOH + 0.33 M urea	10	1.44	8
N <sub>2</sub> P-NF	1.0 M KOH + 0.33 M urea	10	1.47	9
O-NFF	1.0 M KOH + 0.33 M urea	10	1.38	10
NiO <sub>x</sub> /CN <sub>x</sub>	1.0 M KOH + 0.33 M urea	10	1.38	11
Ni(OH) <sub>2</sub> NS@NW/Ni	1.0 M KOH + 0.33 M urea	5	1.58	12
FePS <sub>3</sub> -3000r/NF	1.0 M KOH + 0.33 M urea	10	1.37	This work
FePS <sub>3</sub> /NF	1.0 M KOH + 0.33 M urea	10	1.477	This work

## References

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