

## Supporting information

### **Rapid Electrochemical Synthesis of Co-Ni-Mo Multifunctional Electrocatalyst on Stainless Steel Felt for Efficient Overall Water Splitting Coupled with Phenol Degradation**

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**Table.S1 The core experimental parameters for electrochemical testing**

<b>Electrodeposition synthesis</b>	
Electrode system (three electrodes)	working electrode: stainless steel felt (1 cm × 0.5 cm) counter electrode: platinum wire (∅ 0.5 mm × 37 mm) reference electrode: Ag/AgCl
Main electrochemical parameters	constant potential: -2 V vs Ag/AgCl deposition time: 600 s
Electrolyte	0.152 g of NiCl <sub>2</sub> ·6H <sub>2</sub> O, 0.228 g of CoCl <sub>2</sub> ·6H <sub>2</sub> O, 0.0117 g of Na <sub>2</sub> MoO <sub>4</sub> ·2H <sub>2</sub> O, 0.2 g of H <sub>3</sub> BO <sub>3</sub> , and 0.1 g of NaCl in 10 mL of deionized water
<b>Electrochemical active area test</b>	
Electrode system (three electrodes)	working electrode: CoxNi10-xMo@SSF composite counter electrode: carbon rod (∅ 5 mm × 90 mm) reference electrode: Hg/HgO
Main electrochemical parameters	potential range: -0.06 to -0.02 V (vs. Hg/HgO) scan rates: 10 to 100 mV·s <sup>-1</sup>
Electrolyte	1 mol·L <sup>-1</sup> KOH
<b>Ordinary HER-LSV system</b>	
Electrode system (three electrodes)	working electrode: CoxNi10-xMo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO
Main electrochemical parameters	potential range: -1.92 to -0.92 V (vs. Hg/HgO) scan rate: 2 mV·s <sup>-1</sup>
Electrolyte	1 mol·L <sup>-1</sup> KOH
<b>Ordinary OER-LSV system</b>	
Electrode system (three electrodes)	working electrode: CoxNi10-xMo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO
Main electrochemical parameters	potential range: 0 to 2 V (vs. Hg/HgO) scan rate: 5 mV·s <sup>-1</sup>
Electrolyte	1 mol·L <sup>-1</sup> KOH
<b>Ordinary HER-EIS system</b>	
Electrode system (three electrodes)	working electrode: CoxNi10-xMo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO
Main electrochemical parameters	frequency range: 0.1-10000 Hz amplitude: 5 mV potential: corresponded to a current density of 10 mA·cm <sup>-2</sup> from HER-LSV curve
Electrolyte	1 mol·L <sup>-1</sup> KOH
<b>Ordinary OER-EIS system</b>	
Electrode system (three electrodes)	working electrode: CoxNi10-xMo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO
Main electrochemical parameters	frequency range: 0.1-10000 Hz amplitude: 5 mV potential: corresponded to a current density of 80 mA·cm <sup>-2</sup> from OER-LSV curve
Electrolyte	1 mol·L <sup>-1</sup> KOH

**Continued Table.S1 The core experimental parameters for electrochemical testing**

<b>Ordinary HER i-t system</b>	
Electrode system (three electrodes)	working electrode: Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO
Main electrochemical parameters	potential: corresponded to a current density of 100 mA·cm <sup>-2</sup> from HER-LSV curve testing time: 30 hours
Electrolyte	1 mol·L <sup>-1</sup> KOH
<b>Ordinary OER i-t system</b>	
Electrode system (three electrodes)	working electrode: Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO
Main electrochemical parameters	potential: corresponded to a current density of 100 mA·cm <sup>-2</sup> from OER-LSV curve testing time: 30 hours
Electrolyte	1 mol·L <sup>-1</sup> KOH
<b>Ordinary HER stability test 3000 CV cycles</b>	
Electrode system (three electrodes)	working electrode: Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO
Main electrochemical parameters	potential range: -1.92 to -0.92 V (vs. Hg/HgO) scan rates: 50 mV·s <sup>-1</sup>
Electrolyte	1 mol·L <sup>-1</sup> KOH
<b>Ordinary OER stability test 3000 CV cycles</b>	
Electrode system (three electrodes)	working electrode: Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO
Main electrochemical parameters	potential range: 0 to 2 V (vs. Hg/HgO) scan rates: 50 mV·s <sup>-1</sup>
Electrolyte	1 mol·L <sup>-1</sup> KOH
<b>HER (KOH+ phenol) LSV system</b>	
Electrode system (three electrodes)	working electrode: Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO
Main electrochemical parameters	potential range: -1.92 to -0.92 V (vs. Hg/HgO) scan rate: 2 mV·s <sup>-1</sup>
Electrolyte	1 mol·L <sup>-1</sup> KOH + 20 mg·L <sup>-1</sup> phenol
<b>OER (KOH+ phenol) LSV system</b>	
Electrode system (three electrodes)	working electrode: Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO
Main electrochemical parameters	potential range: 0 to 2 V (vs. Hg/HgO) scan rate: 5 mV·s <sup>-1</sup>
Electrolyte	1 mol·L <sup>-1</sup> KOH + 20 mg·L <sup>-1</sup> phenol

**Continued Table.S1 The core experimental parameters for electrochemical testing**

<b>HER (KOH+ phenol) EIS system</b>	
Electrode system (three electrodes)	working electrode: Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO frequency range: 0.1-10000 Hz
Main electrochemical parameters	amplitude: 5 mV potential: corresponded to a current density of 10 mA·cm <sup>-2</sup> from HER-LSV curve
Electrolyte	1 mol·L <sup>-1</sup> KOH + 20 mg·L <sup>-1</sup> phenol
<b>OER (KOH+ phenol) EIS system</b>	
Electrode system (three electrodes)	working electrode: Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO frequency range: 0.1-10000 Hz
Main electrochemical parameters	amplitude: 5 mV potential: corresponded to a current density of 80 mA·cm <sup>-2</sup> from HER-LSV curve
Electrolyte	1 mol·L <sup>-1</sup> KOH + 20 mg·L <sup>-1</sup> phenol
<b>HER (KOH+ phenol) i-t system</b>	
Electrode system (three electrodes)	working electrode: Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO
Main electrochemical parameters	potential: corresponded to a current density of 100 mA·cm <sup>-2</sup> from HER-LSV curve testing time: 30 hours
Electrolyte	1 mol·L <sup>-1</sup> KOH + 20 mg·L <sup>-1</sup> phenol
<b>OER (KOH+ phenol) i-t system</b>	
Electrode system (three electrodes)	working electrode: Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO
Main electrochemical parameters	potential: corresponded to a current density of 100 mA·cm <sup>-2</sup> from OER-LSV curve testing time: 30 hours
Electrolyte	1 mol·L <sup>-1</sup> KOH + 20 mg·L <sup>-1</sup> phenol
<b>HER (KOH+ phenol) stability test 3000 CV cycles</b>	
Electrode system (three electrodes)	working electrode: Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO
Main electrochemical parameters	potential range: -1.92 to -0.92 V (vs. Hg/HgO) scan rates: 50 mV·s <sup>-1</sup>
Electrolyte	1 mol·L <sup>-1</sup> KOH + 20 mg·L <sup>-1</sup> phenol

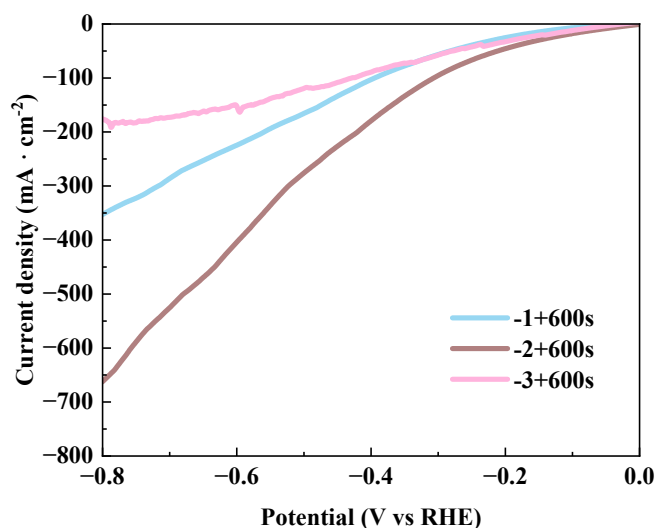
**Continued Table.S1 The core experimental parameters for electrochemical testing**

<b>OER (KOH+ phenol) stability test 3000 CV cycles</b>	
Electrode system (three electrodes)	working electrode: Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO
Main electrochemical parameters	potential range: 0 to 2 V (vs. Hg/HgO) scan rates: 50 mV·s <sup>-1</sup>
Electrolyte	1 mol·L <sup>-1</sup> KOH + 20 mg·L <sup>-1</sup> phenol
<b>Overall water splitting (KOH + phenol) LSV system</b>	
Electrode system (dual electrodes)	working electrode: Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite counter electrode and reference electrode: Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite
Main electrochemical parameters	Cell voltage: 0-3 V scan rate: 2 mV·s <sup>-1</sup>
Electrolyte	1 mol·L <sup>-1</sup> KOH + 20 mg·L <sup>-1</sup> phenol
<b>Overall water splitting (KOH + phenol) i-t system</b>	
Electrode system (dual electrodes)	working electrode: SSF, CoMo@SSF, NiMo@SSF and Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite counter electrode and reference electrode: SSF, CoMo@SSF, NiMo@SSF and Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite
Main electrochemical parameters	Cell voltage: corresponded to a current density of 100 mA·cm <sup>-2</sup> from overall water splitting LSV curve testing time: 30 hours
Electrolyte	1 mol·L <sup>-1</sup> KOH + 20 mg·L <sup>-1</sup> phenol

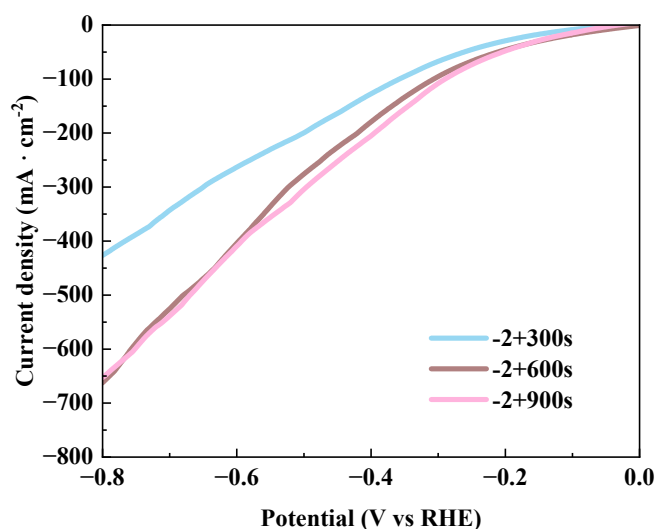
The phenol degradation test was conducted simultaneously using the constant potential chronoamperometry method (i-t) at the electrochemical workstation. The phenol degradation test lasted for 3 hours with a time interval of 30 minutes. The specific electrochemical experimental parameters were shown in Table S2. The reason for choosing different current densities in HER and OER systems is that early preliminary experiments showed that phenol began to degrade in the HER system at a current density of  $20 \text{ mA}\cdot\text{cm}^{-2}$ , while phenol did not degrade in the OER system until the current density increased to  $100 \text{ mA}\cdot\text{cm}^{-2}$ . The dual electrode system selected the same high current density as the OER system.

**Table.S2 The core experimental parameters for electrochemical phenol degradation testing**

<b>phenol degradation in HER three electrodes system</b>	
Electrode system (three electrodes)	working electrode: Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO
Main electrochemical parameters	potential: corresponded to a current density of $20 \text{ mA}\cdot\text{cm}^{-2}$ from HER-LSV curve testing time: 3 hours
Electrolyte	$1 \text{ mol}\cdot\text{L}^{-1}$ KOH + $20 \text{ mg}\cdot\text{L}^{-1}$ phenol
<b>phenol degradation in OER three electrodes system</b>	
Electrode system (three electrodes)	working electrode: Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite counter electrode: carbon rod reference electrode: Hg/HgO
Main electrochemical parameters	potential: corresponded to a current density of $100 \text{ mA}\cdot\text{cm}^{-2}$ from OER-LSV curve testing time: 3 hours
Electrolyte	$1 \text{ mol}\cdot\text{L}^{-1}$ KOH + $20 \text{ mg}\cdot\text{L}^{-1}$ phenol
<b>phenol degradation in overall water splitting dual electrodes system</b>	
Electrode system (dual electrodes)	working electrode: SSF, CoMo@SSF, NiMo@SSF and Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite counter electrode and reference electrode: SSF, CoMo@SSF, NiMo@SSF and Co <sub>6</sub> Ni <sub>4</sub> Mo@SSF composite
Main electrochemical parameters	Cell voltage: corresponded to a current density of $100 \text{ mA}\cdot\text{cm}^{-2}$ from overall water splitting LSV curve testing time: 3 hours
Electrolyte	$1 \text{ mol}\cdot\text{L}^{-1}$ KOH + $20 \text{ mg}\cdot\text{L}^{-1}$ phenol

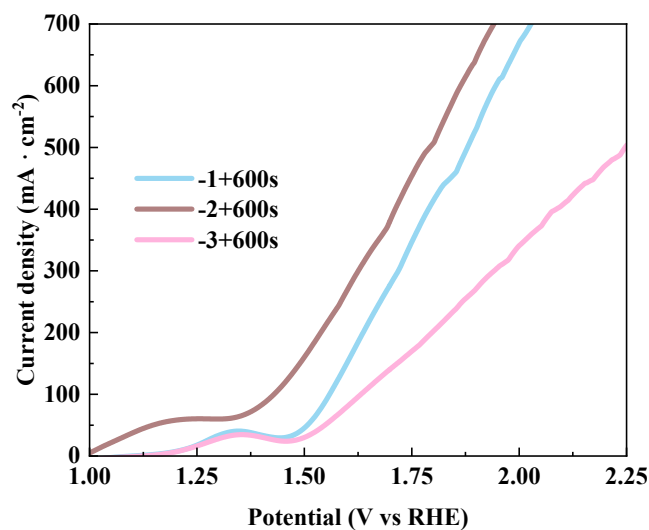


**Fig.S1 The influence of electrodeposition potential on the HER curve of synthetic materials**

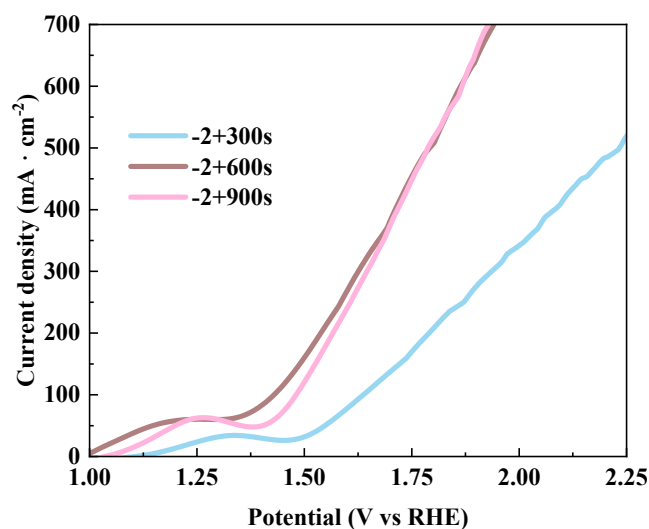


**Fig.S2 The influence of electrodeposition time on the HER curve of synthetic materials**

Fig.S1 and S2 demonstrate the effects of electrodeposition potential and time on the HER efficiency of synthesized samples. From Fig.S1, it could be seen that, under the same electrodeposition time of 600 seconds, the HER efficiency of the sample synthesized under -2 V potential was significantly better than that of -1 V and -3 V, especially the HER efficiency of sample synthesized under -3V potential was the worst. On the other hand, at a electrodeposition voltage of -2 V, the HER efficiency of sample deposited for 600 s was significantly better than that of sample deposited for 300 s. Meanwhile, the HER efficiency of sample deposited for 600 s was not significantly different from that of sample deposited for 900 s. Take both factors into consideration, the electrodeposition potential and time of -2 V and 600 s were ultimately selected as the basic experimental parameters in this study.

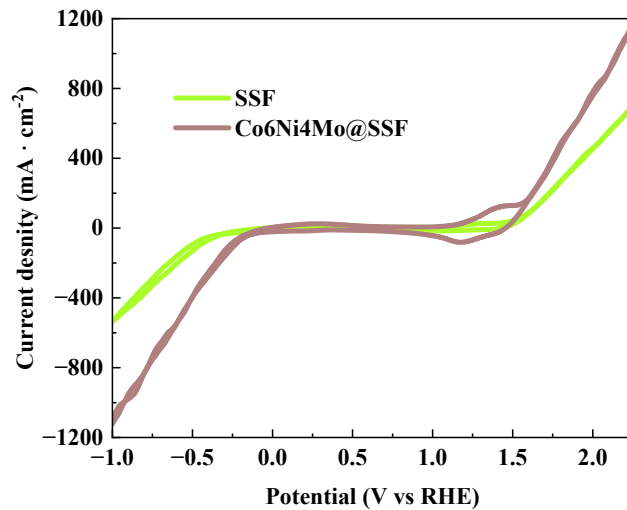


**Fig.S3** The influence of electrodeposition potential on the OER curve of synthetic materials

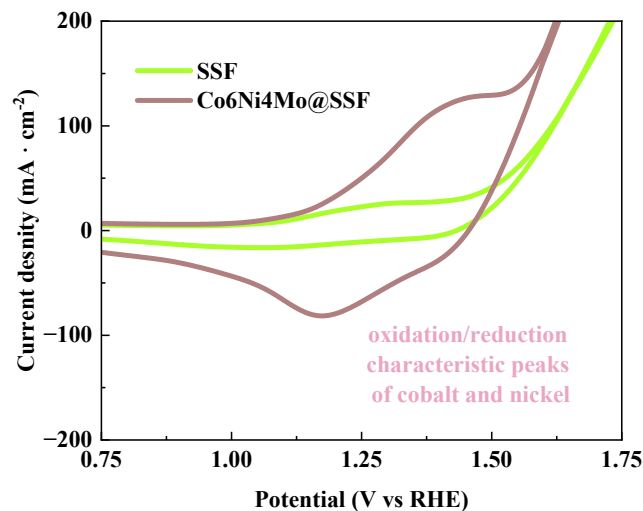


**Fig.S4** The influence of electrodeposition time on the OER curve of synthetic materials

Fig.S3 and S4 demonstrated the effects of electrodeposition potential and time on the OER efficiency of synthesized samples. From Fig.S3 and S4, it could be clearly known that the influence regularity of electrodeposition potential and time on the OER efficiency of the synthetic samples was consistent with that of HER process.



**Fig.S5 The CV scan curves of SSF substrate and Co6Ni4Mo@SSF composite after electrodeposition (-1 to 2.25 V vs RHE)**



**Fig.S6 The detail of CV scan curves of SSF substrate and Co6Ni4Mo@SSF composite after electrodeposition (-0.75 to 1.75 V vs RHE)**

From Fig.S5, it can be known that, after the electrodeposition process was completed, Co6Ni4Mo@SSF sample performed significantly better than SSF substrate at both HER region (bottom left) and OER region (top right). At the same time, the CV curve of Co6Ni4Mo@SSF sample showed distinct oxidation-reduction characteristic peaks of cobalt and nickel elements in the range of 0.75 to 1.75 V (Fig.S6). It was evident that both the results of Fig.S5 and S6 demonstrated the successful electrodeposition of active materials with catalytic activity onto the SSF substrate.

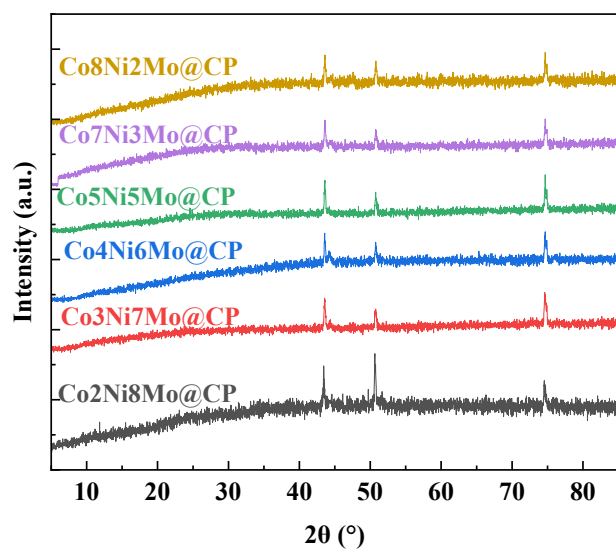


Fig.S7 The XRD patterns of composite electrocatalysts with different Co and Ni ratio

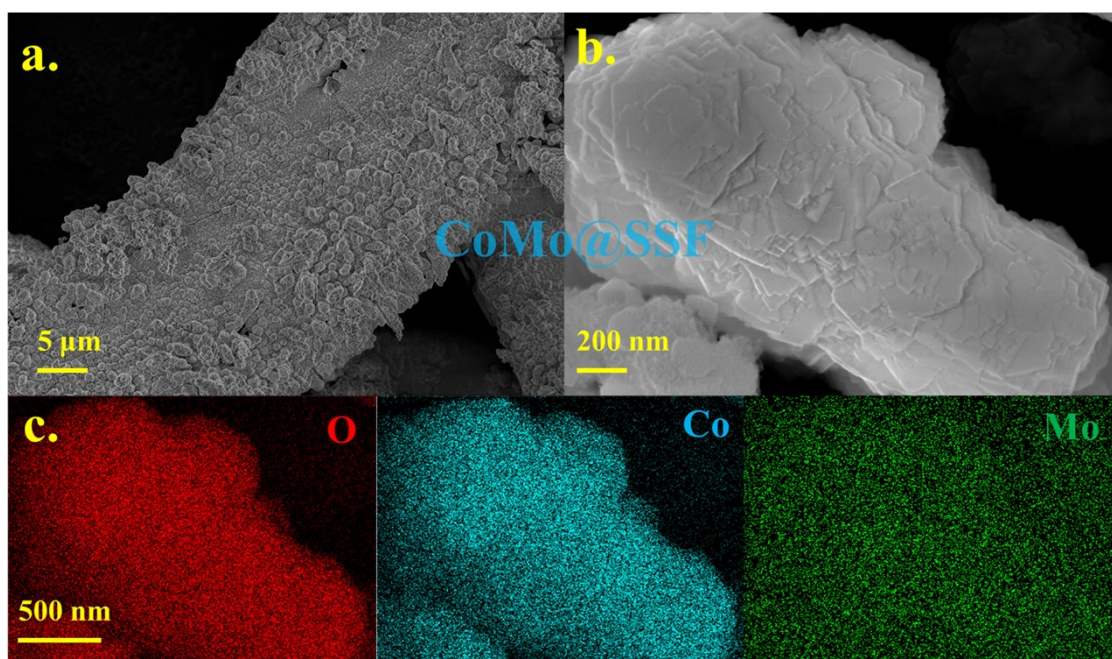


Fig.S8 The SEM morphology (a and b) and EDX-mapping (b and c) analysis results and  $\text{CoMo@SSF}$  electrocatalyst

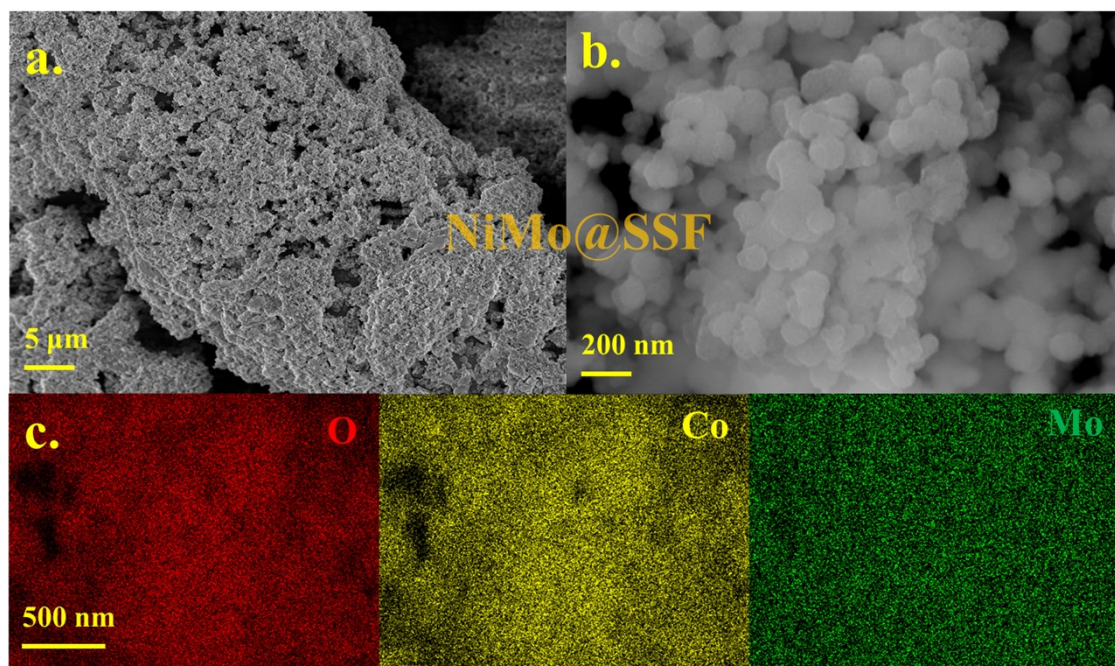


Fig.S9 The SEM morphology (a and b) and EDX-mapping (b and c) analysis results and NiMo@SSF electrocatalyst

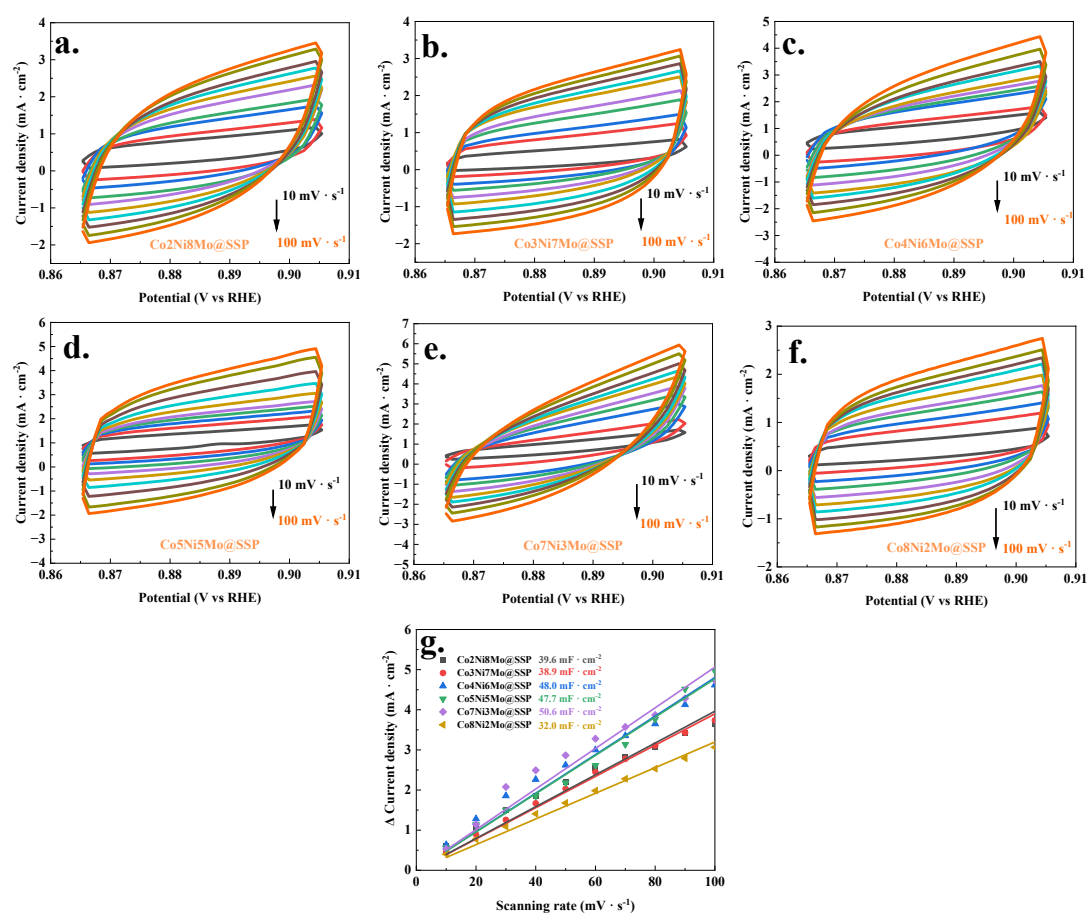


Fig.S10 The CV curves of non-Faraday region (different scanning speeds) and plots of capacitive current density vs different scan rate for  $C_{dl}$  of electrocatalysts with different Co and Ni ratios

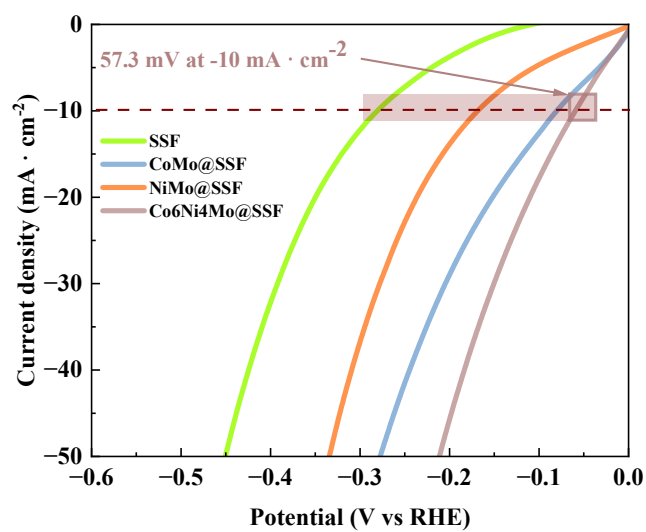


Fig.S11 The detail of HER LSV curves of Fig.6a (0 to -0.6 V vs.RHE)

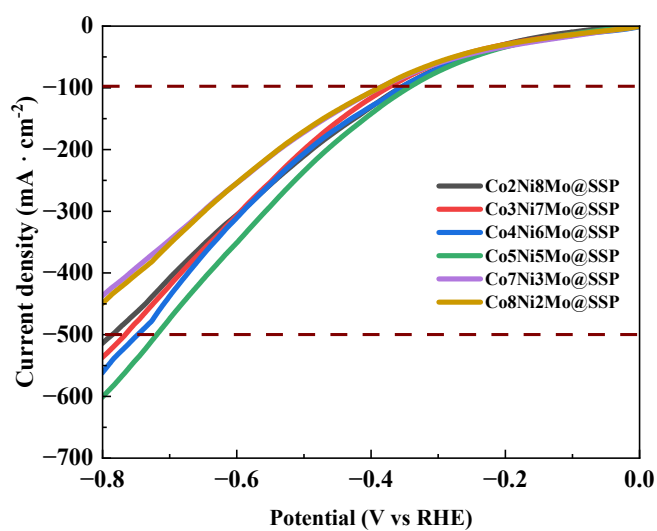


Fig.S12 The HER LSV curves of dual-element molybdate composites with various Co/Ni ratios

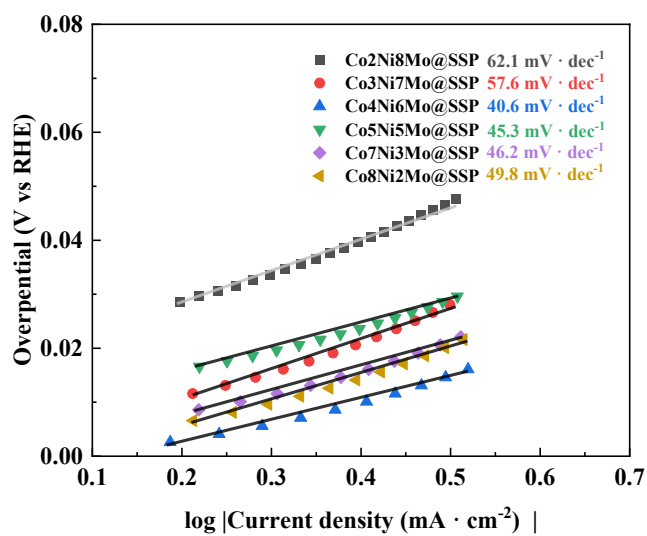


Fig.S13 The HER Tafel slope analysis results of dual-element molybdate composites with various Co/Ni ratios

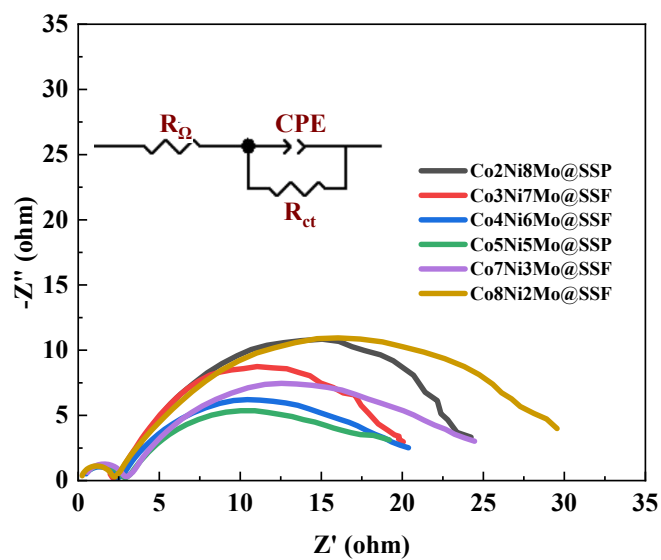
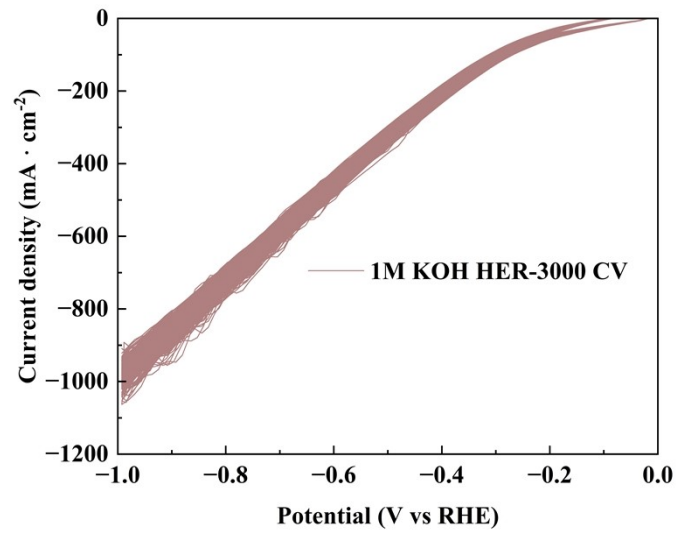


Fig.S14 The HER EIS analysis results of dual-element molybdate composites with various Co/Ni ratios



**Fig.S15** The 3000 CV cycles of HER process in 1 M KOH system

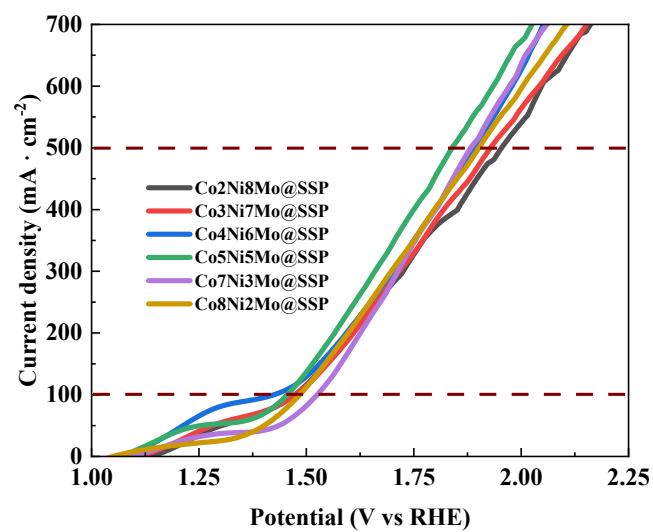


Fig.S16 The OER LSV curves of dual-element molybdate composites with various Co/Ni ratios

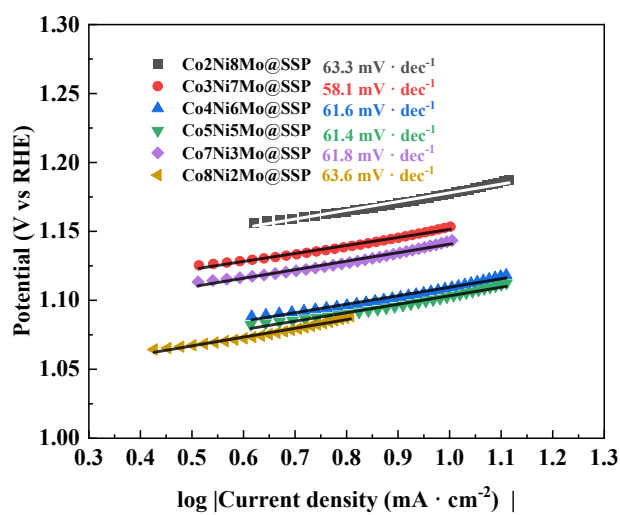


Fig.S17 The OER Tafel slope analysis results of dual-element molybdate composites with various Co/Ni ratios

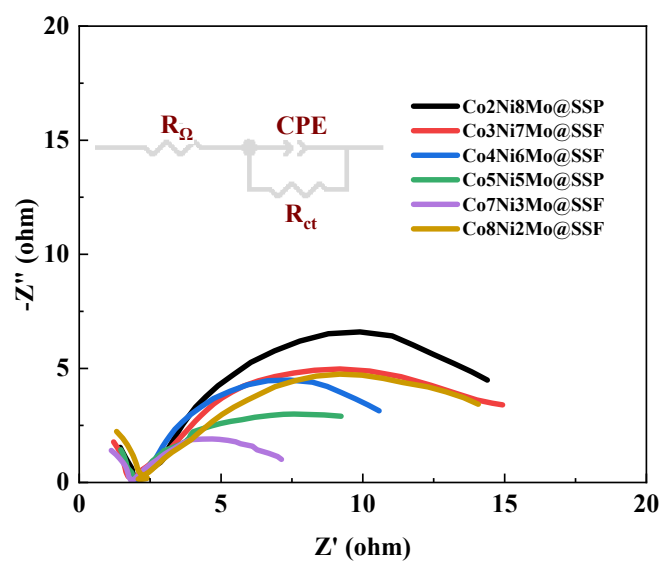


Fig.S18 The OER EIS analysis results of dual-element molybdate composites with various Co/Ni ratios

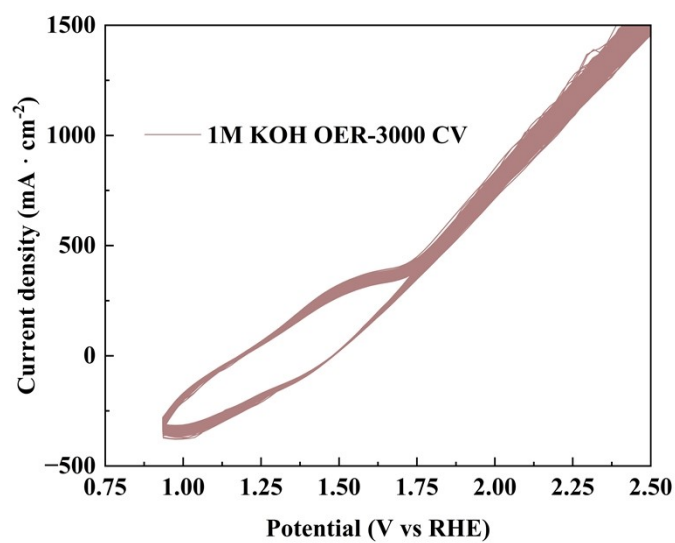


Fig.S19 The 3000 CV cycles of OER process in 1 M KOH system

**Table.S3 Summary of HER overpotentials and Tafel slopes for all reaction systems**

Catalyst sample	electrolyte	overpotential (mV) at 10 mA·cm <sup>-2</sup>	overpotential (mV) at 100 mA·cm <sup>-2</sup>	overpotential (mV) at 500 mA·cm <sup>-2</sup>	Tafel slope mV·dec <sup>-1</sup>
Co6Ni4Mo@SSF		57.3	301.8	678.3	36.9
Co6Ni4Mo@SSF after 3000 CV cycles		69.6	315.8	696.3	37.2
CoMo@SSF		77.9	389.6	801.8	59.7
NiMo@SSF		166.1	416.2	-	85.9
SSF	1 mol·L <sup>-1</sup> KOH	281.5	538.6	-	107.7
Co2Ni8Mo@SSF		103.1	354.0	786.5	62.1
Co3Ni7Mo@SSF		71.3	373.4	768.0	57.6
Co4Ni6Mo@SSF		66.2	351.8	744.7	40.6
Co5Ni5Mo@SSF		81.5	340.7	718.1	45.3
Co7Ni3Mo@SSF		61.1	385.6	-	46.2
Co8Ni2Mo@SSF		71.2	384.5	-	49.8
Co6Ni4Mo@SSF	1 mol·L <sup>-1</sup> KOH + 20 mg·L <sup>-1</sup> phenol	64.6	330.5	691.7	46.1

**Table.S4 Summary of OER overpotentials and Tafel slopes for all reaction systems**

Catalyst sample	electrolyte	overpotential (mV) at 100 mA·cm <sup>-2</sup>	overpotential (mV) at 500 mA·cm <sup>-2</sup>	Tafel slope mV·dec <sup>-1</sup>
Co6Ni4Mo@SSF		207.1	558.1	53.8
Co6Ni4Mo@SSF after 3000 CV cycles		237.1	570.1	55.3
CoMo@SSF		307.5	757.0	60.9
NiMo@SSF		308.3	758.2	60.1
SSF	1 mol·L <sup>-1</sup> KOH	394.0	823.9	79.6
Co2Ni8Mo@SSF		246.8	728.4	63.3
Co3Ni7Mo@SSF		246.5	701.3	58.1
Co4Ni6Mo@SSF		210.3	662.8	61.6
Co5Ni5Mo@SSF		223.1	605.6	61.4
Co7Ni3Mo@SSF		301.2	656.6	61.8
Co8Ni2Mo@SSF		258.2	675.7	63.6
Co6Ni4Mo@SSF	1 mol·L <sup>-1</sup> KOH + 20 mg·L <sup>-1</sup> phenol	229.0	559.2	44.9

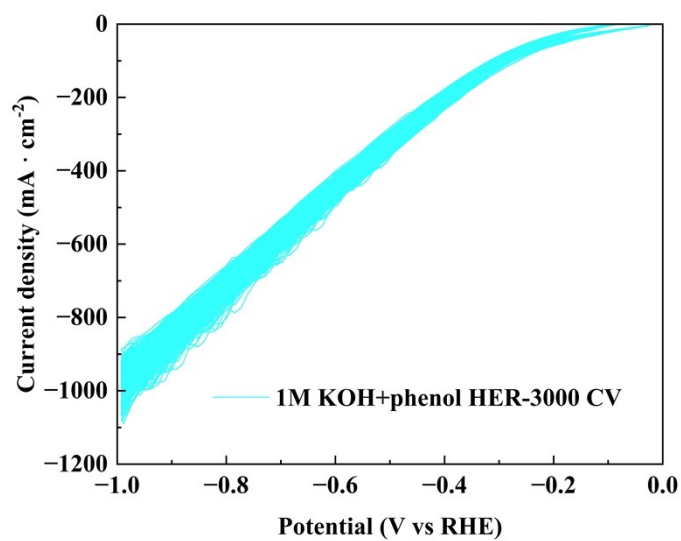


Fig.S20 The 3000 CV cycles of HER process in 1 M KOH + phenol system

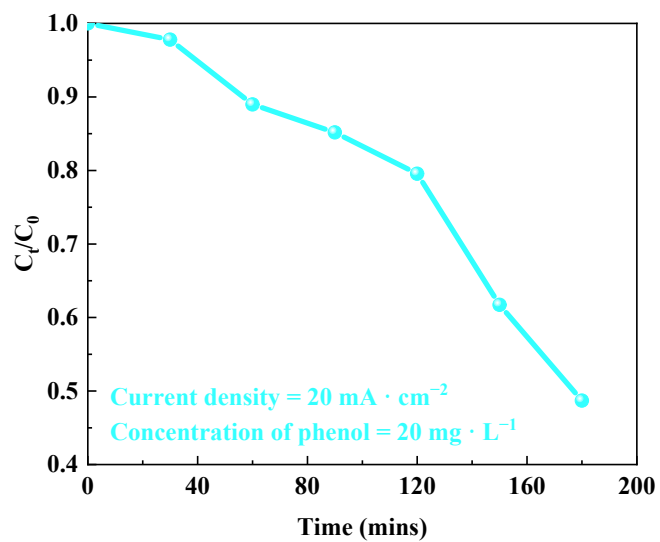


Fig.S21 The phenol degradation curve in the i-t test of HER

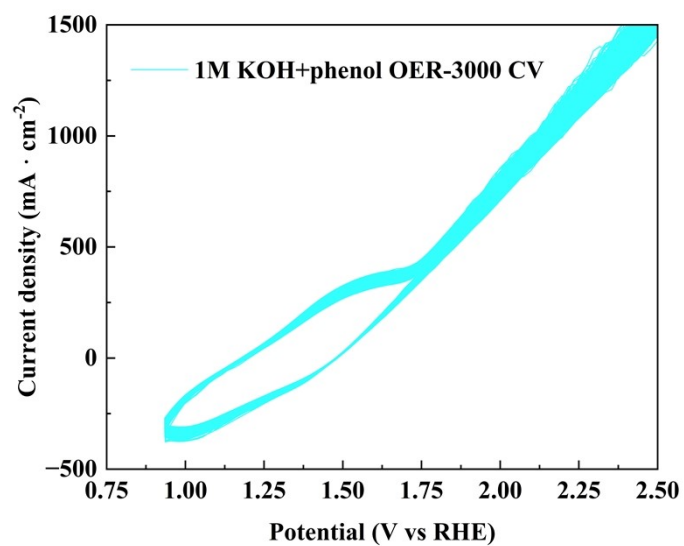


Fig.S22 The 3000 CV cycles of OER process in 1 M KOH + phenol system

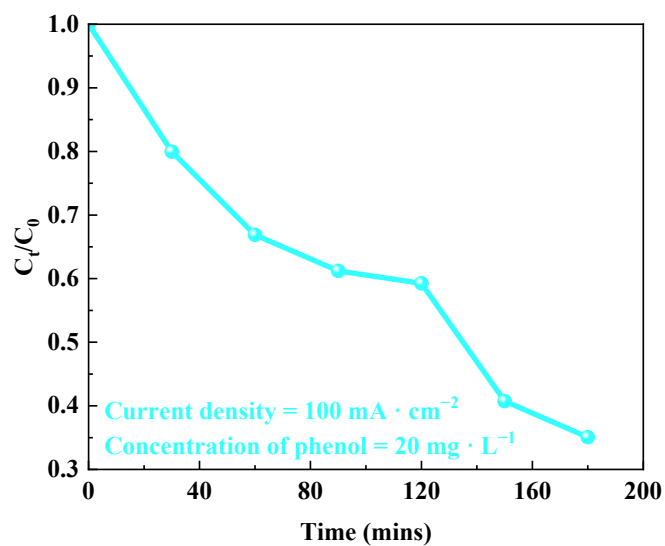


Fig.S23 The phenol degradation curve in the i-t test of OER