

Supporting Information

Interface engineering of Co-CoMoS_x heterostructure nanosheets for efficient alkaline oxygen evolution reaction

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1. Chemicals and Reagents

Cobalt nitrate hexahydrate (Co(NO₃)₂·6H₂O), Sodium molybdate (vi) dihydrate (Na₂MoO₄·2H₂O), Ammonium molybdate tetrahydrate ((NH₄)₆Mo₇O₂₄·4H₂O), Cobalt(II) chloride hexahydrate (CoCl₂·6H₂O), Urea (CH₄N₂O), Thiourea (CH₄N₂S) and Sodium sulfide nonahydrate (Na₂S·9H₂O) were purchased from Macklin Reagent. All the chemicals and reagents are of analytical grade and used without further

purification. Double-distilled water was used throughout the experiment.

2. Experimental sections

2.1. Synthesis of Co-CoMoS_x nanosheets

In the first step, one piece of nickel foam (20.0 mm×20.0 mm) was washed with 3 M HCl, ethanol and deionized water for 20 minutes respectively. Then 0.25 mmol Co(NO₃)₂·6H₂O, 0.25 mmol Na₂MoO₄·2H₂O were dissolved in 30 ml deionized water and stirred for 20 minutes to form a uniform solution. The solution and treated nickel foam were then transferred to 50 ml Teflon lined stainless steel autoclave. The autoclave was maintained at 150 °C for 6 hours. After the reaction was finished, the nickel foam loaded with the precursor was washed with deionized water and then dried at 50 °C for 2 hours.

The second step, 1 mL of 0.5 M Na₂S solution was added into 30 mL deionized water under stirring. Then the sodium sulfide solution and the precursor loaded nickel foam were transferred into a autoclave and heated at 160 °C for 4 hours. After the reaction was finished, the nickel foam was washed with deionized water and then dried in a vacuum at 50 °C for 3 hours.

2.2. Synthesis of CoMoO_x nanosheets

The nickel foam with the Co-Mo precursor was annealed in the air at 400 °C for 2 hours at a heating rate of 6 °C·min⁻¹ to obtain CoMoO_x nanosheets.

2.3. Synthesis of phase-separated Mo-Ni alloy (PS-MoNi) electrode

The method for preparation of PS-MoNi electrode was reported in Advanced Energy Materials (Adv. Energy Mater. 2021, 2003511)¹. One piece of Ni foam (20.0 mm×20.0 mm) was first washed several times by HCl and deionized water. Then, 10 mmol L⁻¹ (NH₄)₆Mo₇O₂₄·4H₂O were dispersed in deionized water to form a uniform solution. Subsequently, the solution and nickel foam were transferred into a 50 mL autoclave. The autoclave was maintain at 150 °C for 20 hours. The Ni foams with precursor was rinsed with deionized water and then dried naturally. Then, the Ni foam with precursor was held at 450 °C for one hour in H₂ atmosphere.

2.4. Synthesis of Co₉S₈ electrode

In a typical procedure², 0.24 g CoCl₂·6H₂O and 0.3 g urea were dissolved in deionized water (30 mL) and stirred for 20 minutes to form a uniform solution, then the solution was transferred into an autoclave with a capacity of 50 mL. A piece of nickel foam (20.0 mm × 30.0 mm) which has been washed with 3M HCL and deionized water was soaked in the above solution. The autoclave was maintained at 95 °C for 8 hours. After the reaction is finish, the nickel foam was washed with deionized water ,then dried in a vacuum at 50 °C for 2 hours.Then 1 mL of 0.5 M Na₂S solution was added into 30 mL deionized water under stirring.Then the sodium sulfide solution and the precursor loaded nickel foam were transferred into a autoclave and maintained at 120 °C for 2 hours. After the reaction was

finished, the foam nickel was washed with deionized water and then dried in a vacuum at 50 °C for 4 hours.

2.5. Synthesis of MoS₂ electrode

In a typical procedure³, 0.15mmol (NH₄)₆Mo₇O₂₄·4H₂O, and 6mmol thiourea are dissolved in 35 mL of deionized water and transferred into an autoclave with a capacity of 50 mL. The autoclave was heated at 180 °C for 12 hours. After the reaction was finished, The black product was centrifuged and washed three times each with water and ethanol. Finally, the precipitation was dried overnight at 50 °C in a vacuum oven.

3. Electrochemical measurements

The OER electrochemical performances were tested using a three-electrode setup in 1 M KOH solution. The nickel foams loaded with catalyst were directly used as the working electrodes. The reversible hydrogen electrode was used as reference electrode and counter electrode. The graphite rod electrode was used as counter electrode. The overall water splitting is performed in a two-electrode system. The polarization curves at a scan rate of 5 mV s⁻¹ were measured in 1 M KOH solution. Electrochemical impedance spectroscopy (EIS) measurements of the samples were tested in a three-electrode system with a frequency range from 10⁵ Hz to 0.1 Hz.

4. Materials characterization

The micromorphology of the material was observed by field emission

scanning electron microscope (Hitachi FESEM SU8220) and environmental spherical aberration correction scanning transmission electron microscope (Titan ETEM G² 80-300) of USA FEI company. X-ray diffractometer (SMARTLAB3KW, Rigaku Co., Japan) is used to determine the physical properties and structure of materials with a range of 10° to 90°. The element composition and valence state of the material were analyzed by X-ray photoelectron (ESCALAB 250XI+, Thermo Fisher Scientific company, USA) spectroscopy with the Al K α as the radiation source.

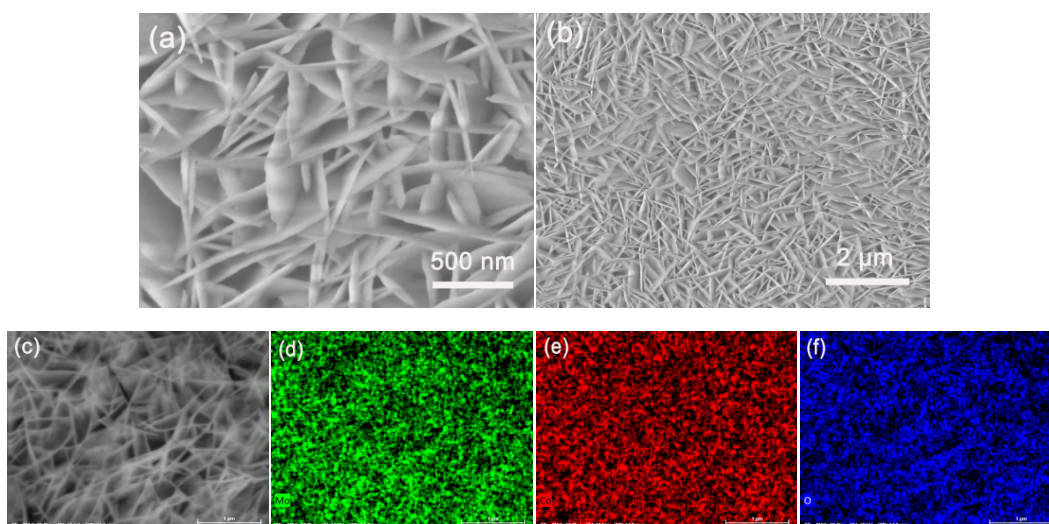


Figure S1. (a,b) SEM images and (c-f) corresponding mapping images of CoMoO_x.

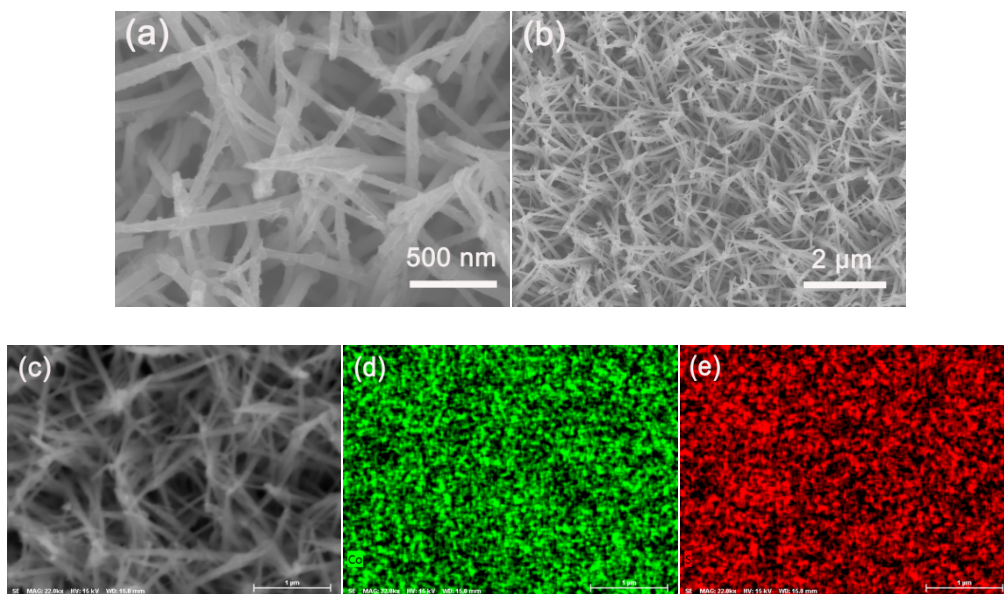


Figure S2. (a,b) SEM images and (c-e) corresponding mapping images of Co₉S₈.

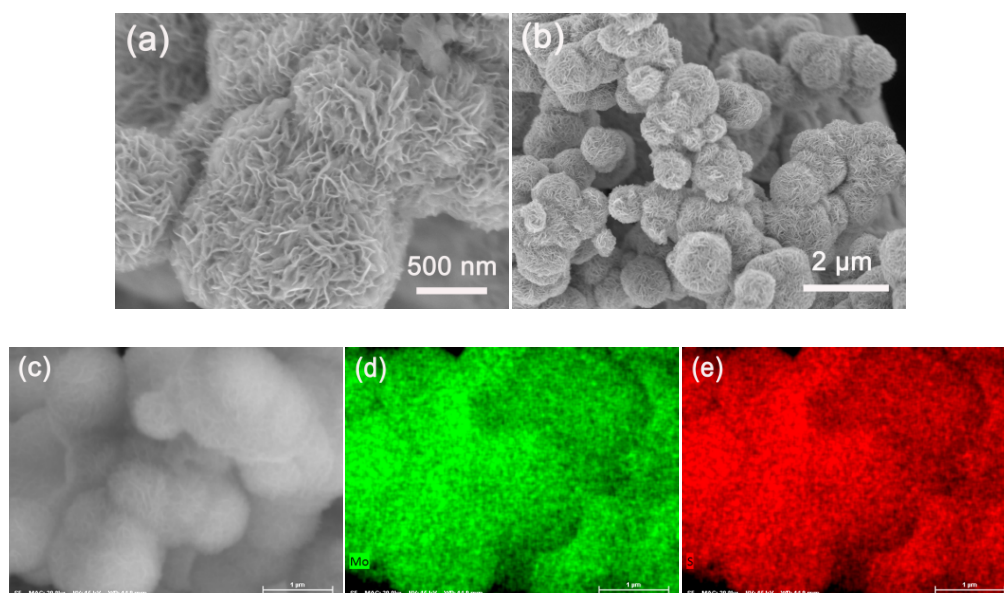


Figure S3. (a,b) SEM images and (c-e) corresponding mapping images of MoS₂.

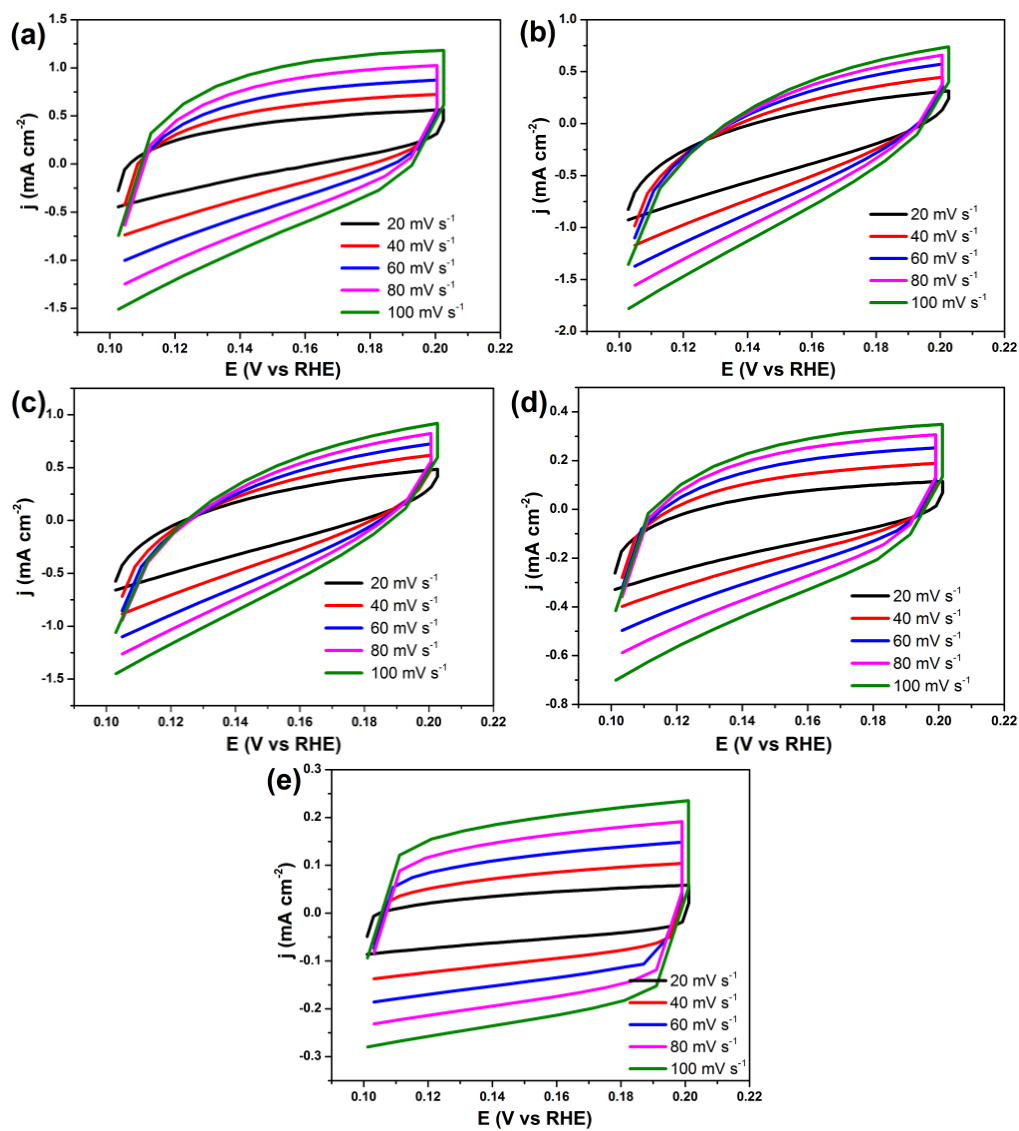


Figure S4. Electrochemical activity specific surface area of different catalysts. Cyclic voltammograms of (a) Co-CoMoS_x, (b) CoMoO_x, (c) Co-Mo precursor, (d)Co₉S₈ and (e) MoS₂ with different rates from 20 mV s⁻¹ to 100 mV s⁻¹.

References

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3. P. Kuang, T. Tong, K. Fan and J. Yu, *ACS Catalysis*, 2017, **7**, 6179-6187.