Electronic Supplementary Material

Interface engineering for enhancing electrocatalytic oxygen evolution reaction of CoS/CeO₂ heterostructures

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Experimental

Preparation of CoS/CC

The electrodeposition solution contained 5 mM $Co(NO_3)_2 \cdot 6H_2O$ and 0.75 M thiourea. The electrodynamic deposition was carried out in a three-electrode cell using the cleaned (2 x 2, cm x cm) CC as the working electrode, a graphite plate as the counter electrode, and Ag/AgCl as the reference electrode. The electrochemical deposition of CoS nanosheets by cyclic voltammetry (CV) was performed within the potential range between -1.2 V and 0.2 V vs. Ag/AgCl at a scan rate of 5 mV s⁻¹ for 15 scan cycles. The as-prepared CoS/CC electrode was rinsed with water and dried in vacuum at 60°C for 12 h.

Characterization

Powder x-ray diffraction (XRD) data were acquired on a RigakuD/MAX 2550 diffractometer with Cu K α radiation ($\lambda = 1.5418$ Å). For XRD, the sample of CoS/CeO₂/CC was annealed at 500°C at argon atmosphere for 1 h. The x-ray photoelectron spectroscopy (XPS) measurements were

performed on an ESCALABMK II x-ray photoelectron spectrometer using Mg as the exciting source. The scanning electron microscopy (SEM) measurements were carried out on a XL30 ESEM FEG scanning electron microscope at an accelerating voltage of 20 kV. The transmission electron microscopy (TEM) measurements were performed on a HITACHI H-8100 electron microscopy (Hitachi, Tokyo, Japan) with an accelerating voltage of 200 kV.

Electrochemical measurements

Electrochemical measurements were performed with a CHI 660E electrochemical analyzer (CH Instruments, Inc., Shanghai). All electrochemical measurements were conducted in a typical three-electrode setup with an electrolyte solution of 1.0 M KOH using CoS/CeO₂/CC as the working electrode, a graphite plate as the counter electrode, and Hg/HgO electrode as the reference electrode. Linear sweep voltammetry (LSV) measurements were conducted in 1.0 M KOH with a scan rate of 2 mV s⁻¹. The reference electrode was calibrated before the electrochemical measurements. All the potentials reported in this work were vs. RHE (reversible hydrogen electrode) in 1.0 M KOH. E (RHE) = E (Hg/HgO) + 0.924 V, $\eta = E$ (RHE) – 1.23 V. The onset potential was read from the Tafel plot for OER and determined based on the beginning of linear regime in the Tafel plot.

Density Functional Theory Calculations

Density functional theory (DFT) calculations were conducted with the Cambridge sequential total energy package (CASTEP) using the Perdew–Burke–Ernzerhof (PBE) generalized gradient approximation (GGA) functional. The energy cutoff was set as 400 eV. The CoS/CeO₂ heterostructure model was constructed by CoS (001) plane and CeO₂ (111) plane. A vacuum space with thickness more than 20 Å was set to avoid the interactions of the periodic images.



Fig. S1. XRD pattern of CoS/CeO₂/CC.



Fig. S2. Low-magnification and high-magnification SEM images of (a, b) CeO₂/CC at 0.5 mA cm⁻², (c, d) CeO₂/CC at 1 mA cm⁻², (e, f) CoS/CC.



Fig. S3. EDS spectrum of CoS/CeO₂/CC.



Fig. S4. XPS survey spectra of CeO₂/CC, CoS/CC, and CoS/CeO₂/CC.



Fig. S5. SEM image of CoS/CeO₂/CC after electrocatalytic reactions.



Fig. S6. (a) XPS Survey, (b)Co 2p, (c)Ce 3d, and (d)S 2p spectra of CoS/CeO₂/CC after

electrocatalytic reactions.



Fig. S7. Nyquist plots of CeO₂/CC, CoS/CC, and CoS/CeO₂/CC (inset: equivalent circuit model).



Fig. S8. CV curves of CoS/CeO₂/CC (a), CoS/CC (b), and CeO₂/CC (c) at various scan rates. Calculated electrochemical double-layer capacitance (c) for CoS/CeO₂/CC, CoS/CC, and CeO₂/CC.

	R _s (ohm)	R _{ct} (ohm)
CeO ₂ /CC	3.07	210
CoS/CC	3.35	118
CoS/CeO ₂ /CC	2.69	113

Table S1. The Fitting results of equivalent circuit diagram.