Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Rapid Cationic Defect and Anion Dual-Regulated Layered Double Hydroxides for Efficient Water Oxidation

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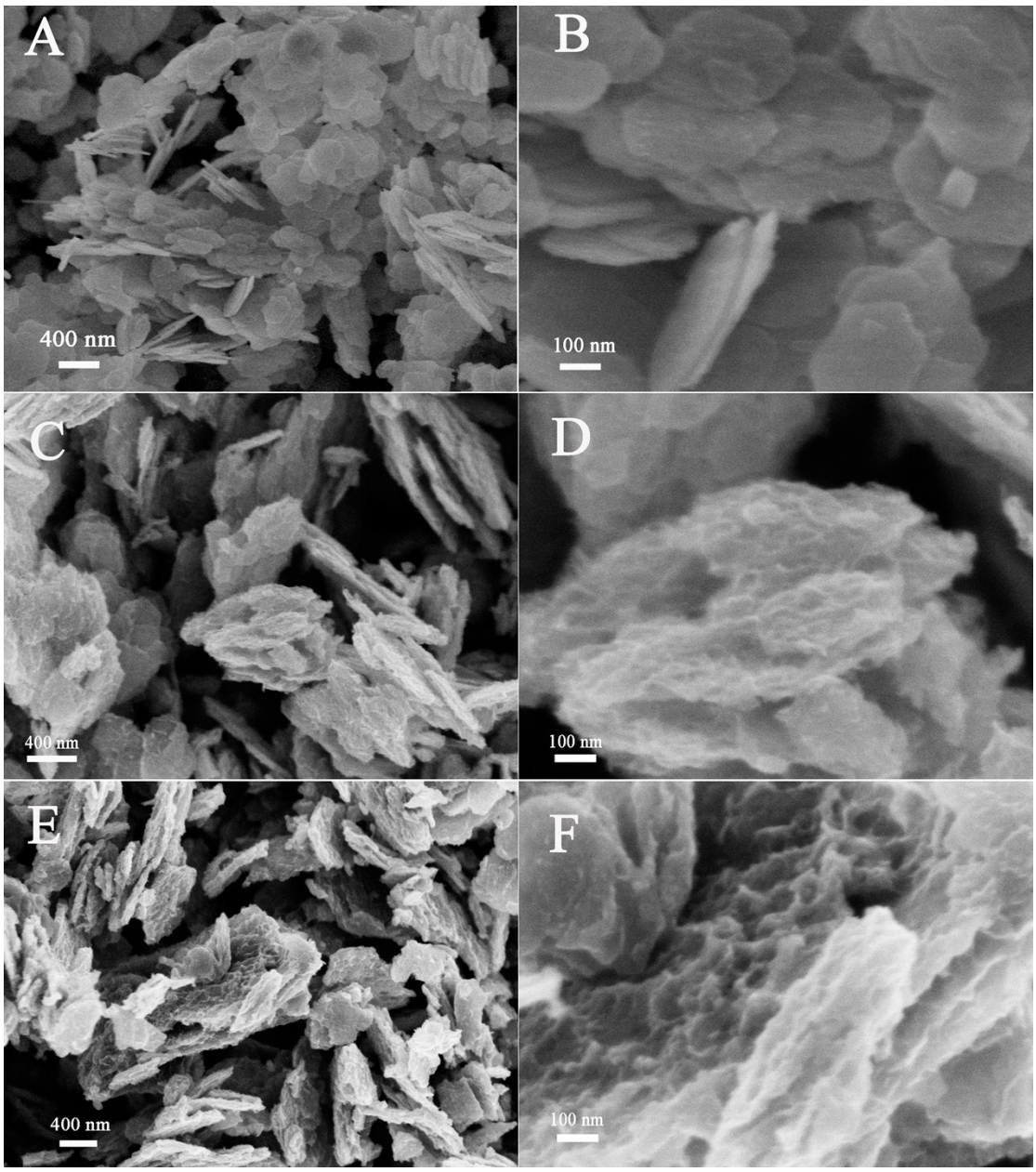

Figure S1. SEM images of (A, B) modified water DBD plasma-1 min; (C, D) modified water DBD plasma-3 min; and (E, F) modified water DBD plasma -30 min.

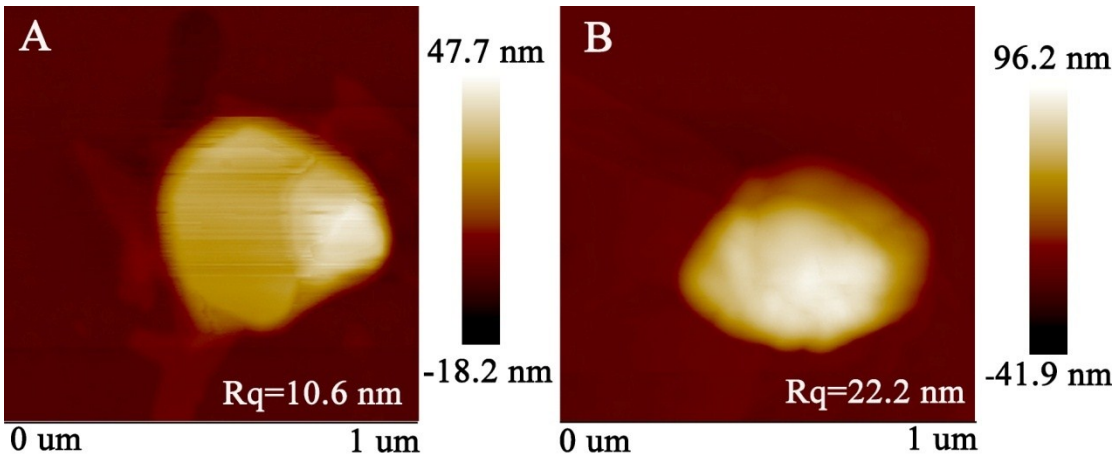

Figure S2. AFM images of A) CoAl LDHs and B) PS-CoAl LDHs. Rq= the average surface roughness (RMS).

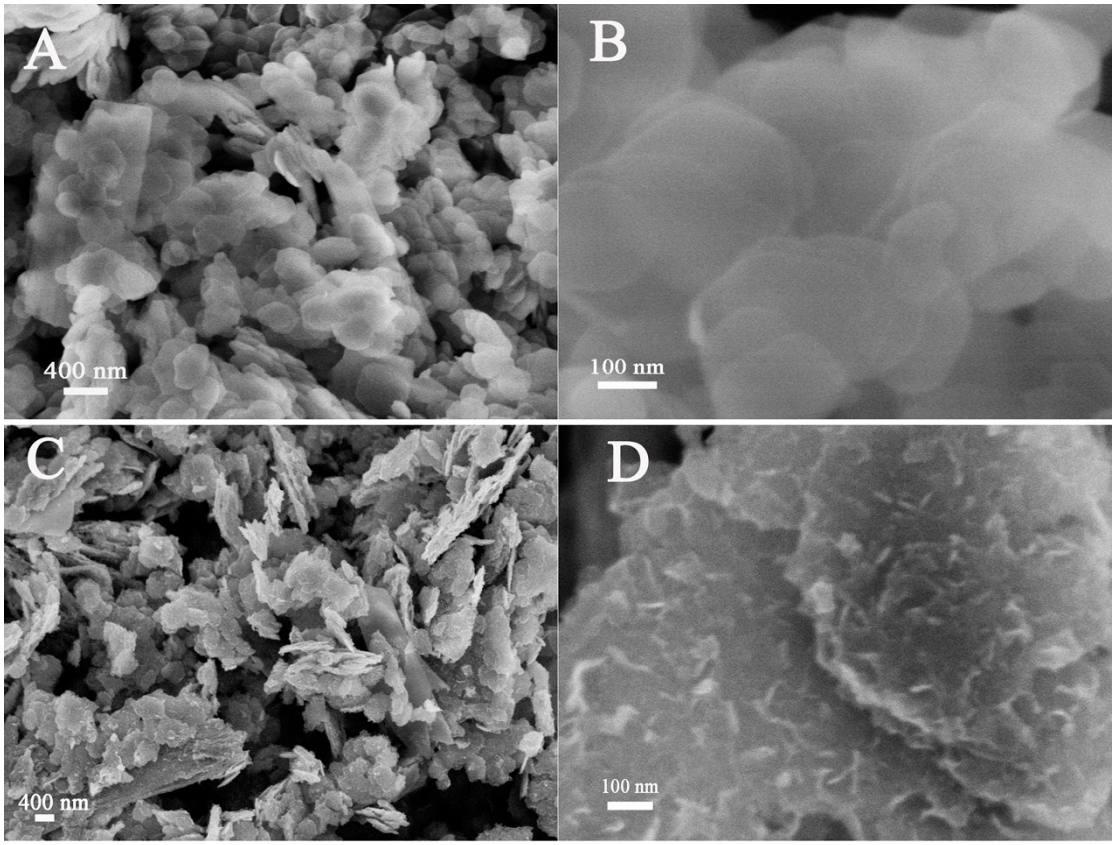

Figure S3. SEM images of (A, B) P-CoAl LDHs and (C, D) S-CoAl LDHs.

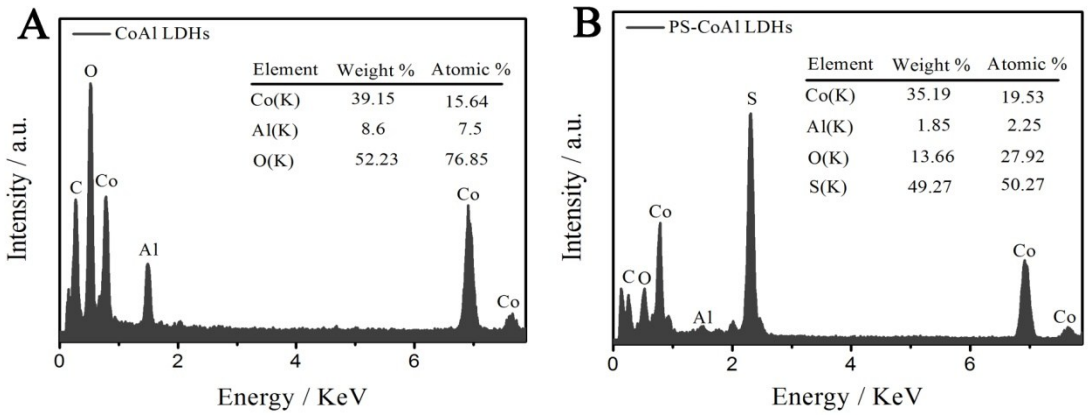

Figure S4. The energy-dispersive X-ray (EDX) images of A) CoAl LDHs and B) PS-CoAl LDHs.

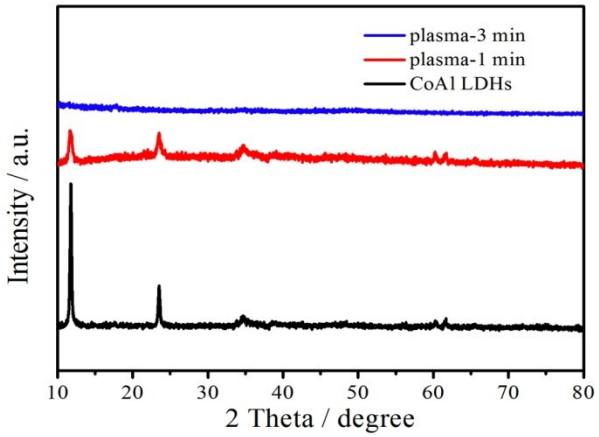

Figure S5. XRD patterns of the plasma irradiation for 1 min, 3 min, and pristine CoAl LDHs.

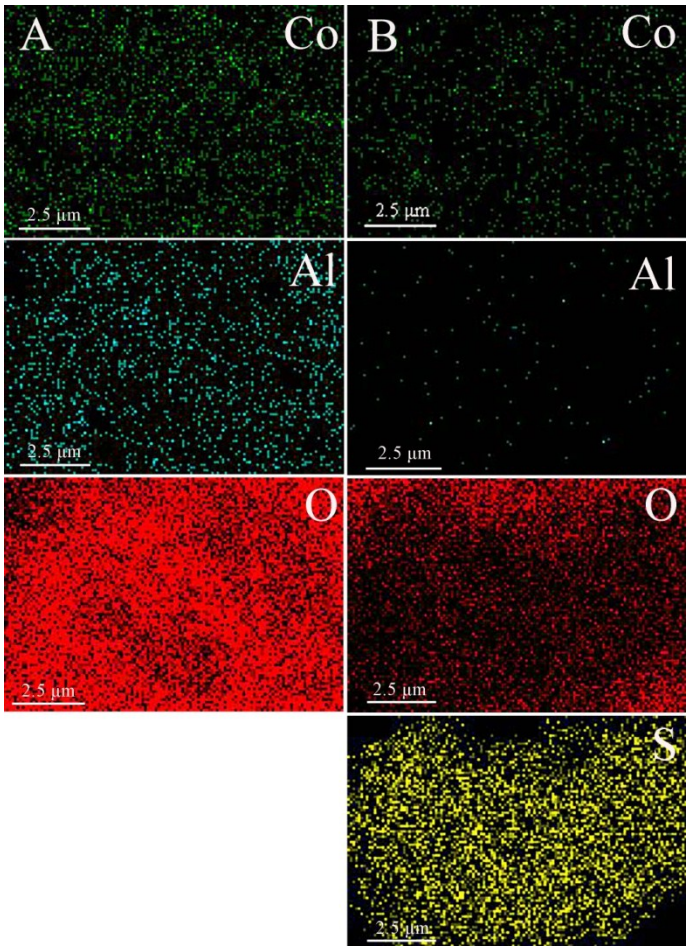

Figure S6. SEM-EDS mapping images of A) CoAl LDHs and B) PS-CoAl LDHs.

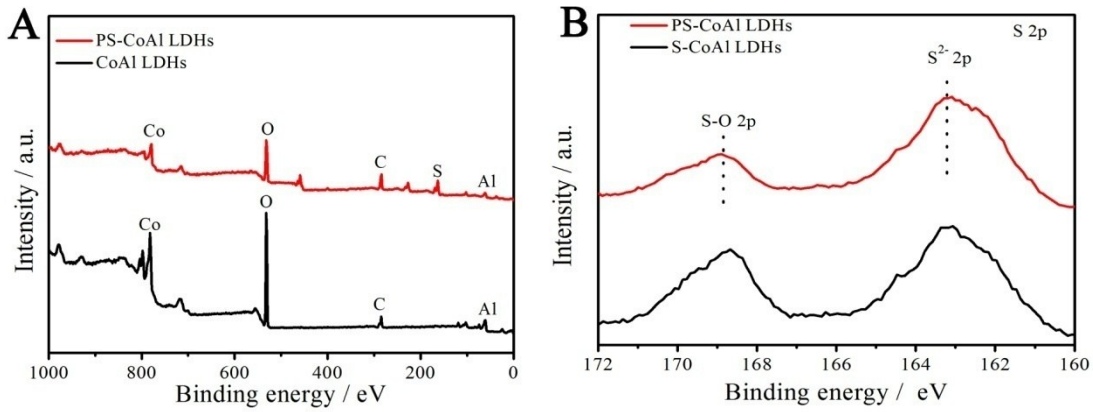

Figure S7. A) XPS survey spectra of CoAl LDHs and PS-CoAl LDHs, B) S 2p XPS peaks of S-CoAl LDHS and PS-CoAl LDHs.

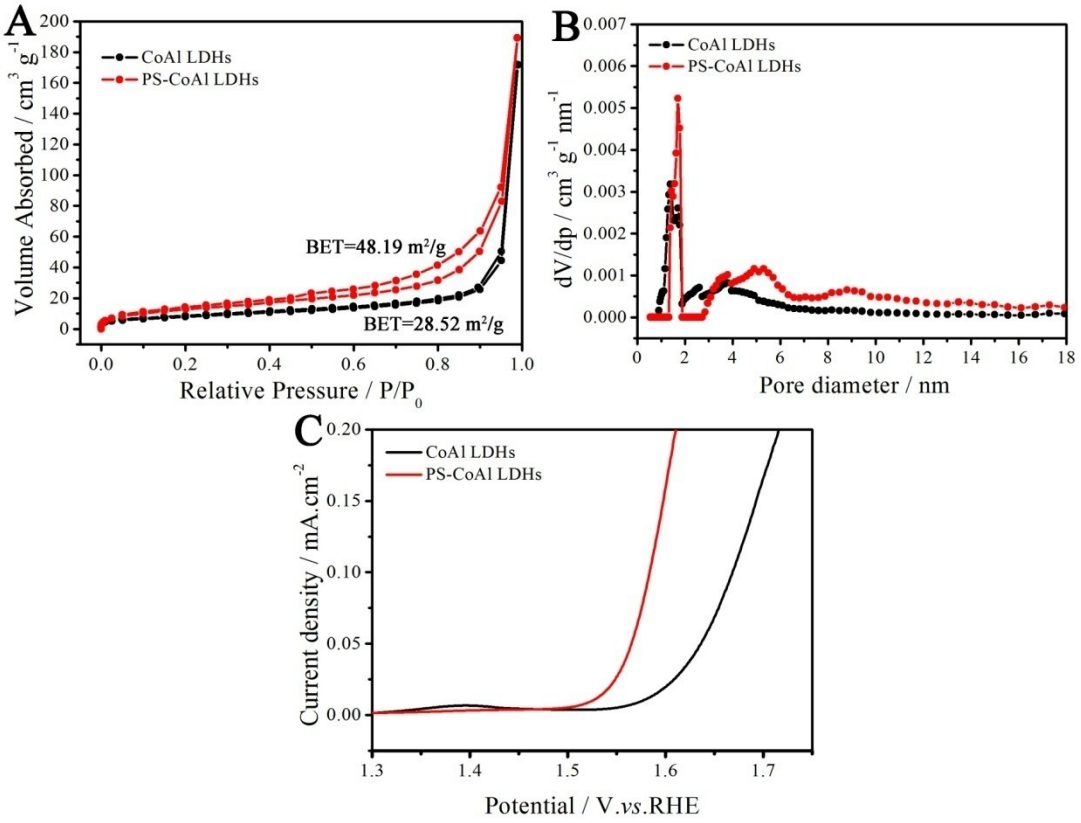

Figure S8. A) Nitrogen adsorption-desorption isotherms of CoAl LDHs and PS-CoAl LDHs; B) Corresponding pore size distribution; C) The OER performance of CoAl LDHs and PS-CoAl LDHs after BET normalization.

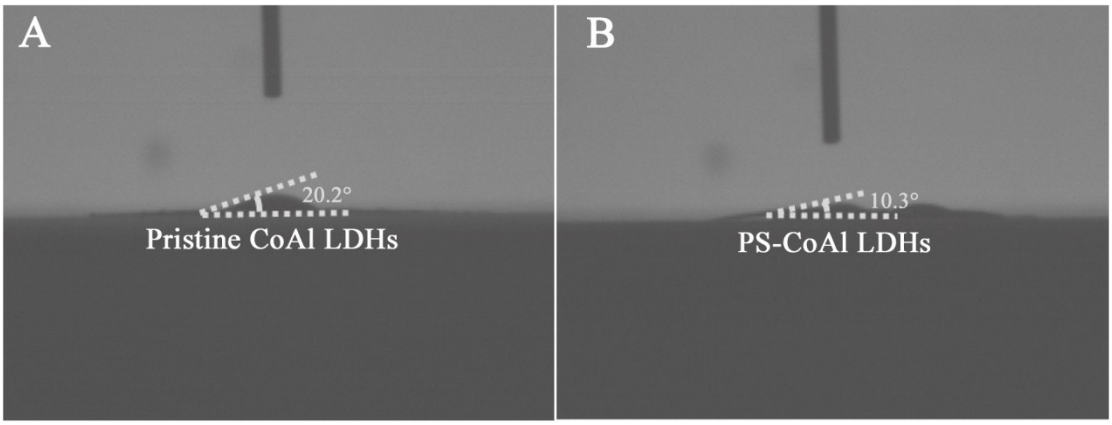

Figure S9. Contact angle measurements of A) pristine CoAl LDHs and B) PS-CoAl LDHs.

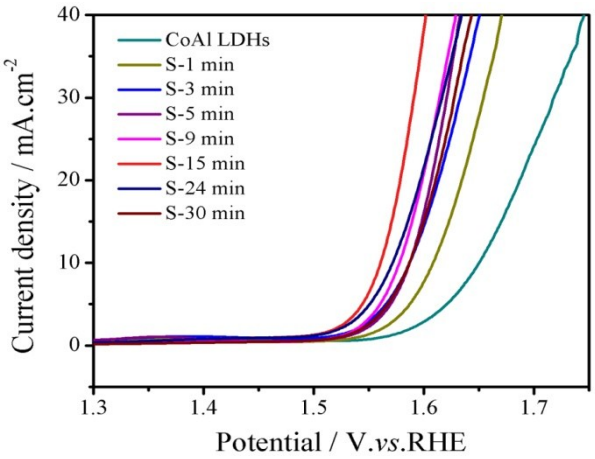

Figure S10. LSV polarization curves of CoAl LDHs by modified water plasma treatments for different time.

The optimal DBD plasma treatment time is 15 min. With the increase of time, the content of sulfur gradually increased, and the appropriate sulfur/oxygen ratio is achieved at 15 minutes to boost OER activity.

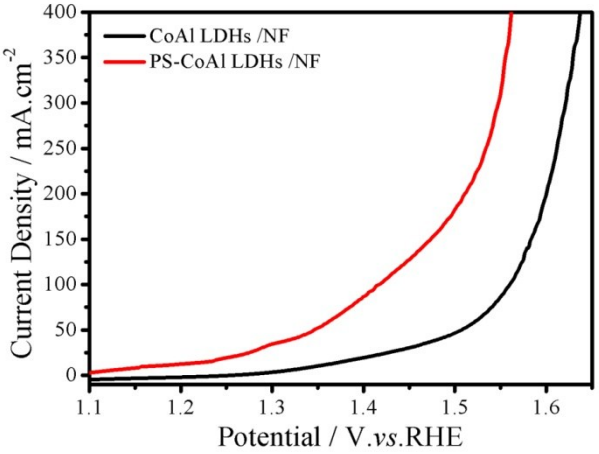

Figure S11. LSV polarization curves of CoAl LDHs and PS-CoAl LDH on NF.

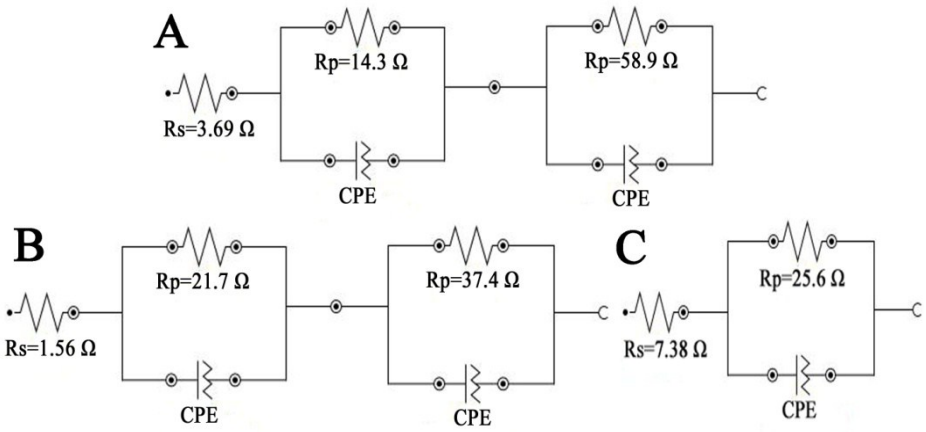

Figure S12. Electrochemical impedance spectroscopy (EIS) fitting results for A) CoAl LDHs; B) S-CoAl LDHs and C) PS-CoAl LDHs. Rs: electrolyte resistance, Rp: charge-transfer resistance, CPE: constant-phase element.

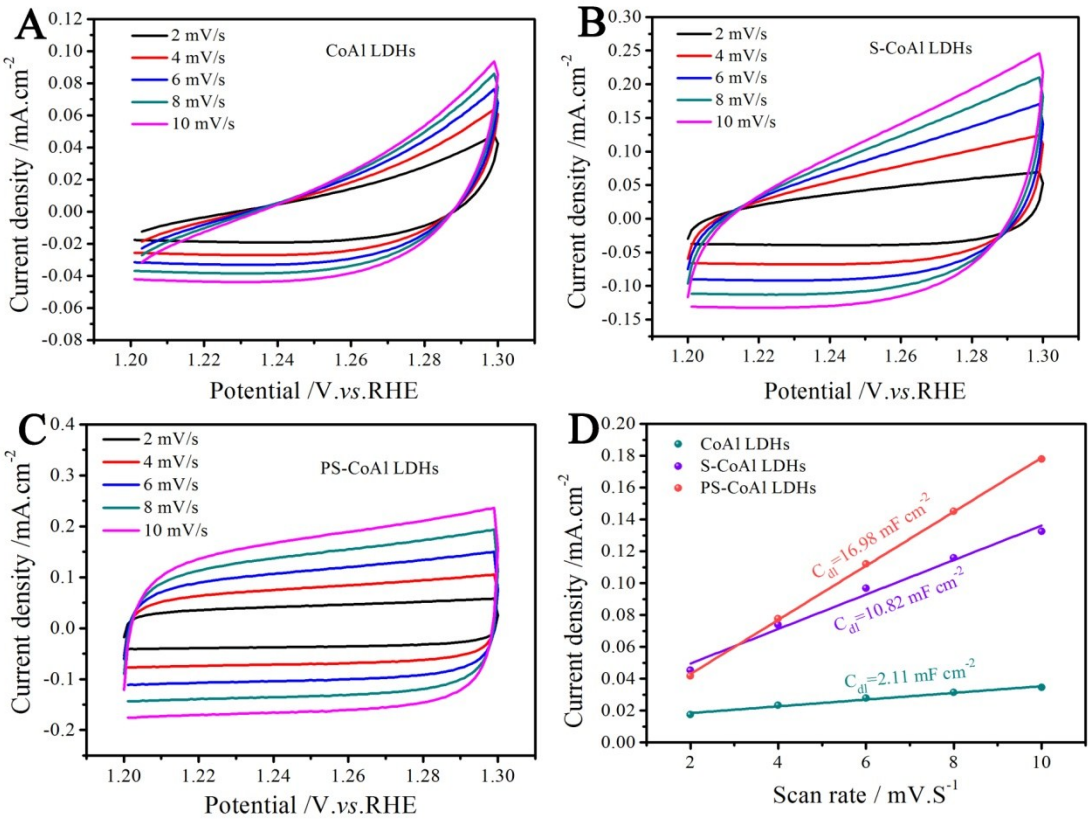

Figure S13. Electrochemical surface area (ESCA) tests of CoAl LDHs, S-CoAl LDHs and PS-CoAl LDHs towards OER in 1 M KOH. A) Cyclic voltammetry curves of A) CoAl LDHs, B) S-CoAl LDHs and C) PS-CoAl LDHs with different scanning rates. D) The capacitive current measured at 1.27 V vs RHE was plotted as a function of scan rate CoAl LDHs, S-CoAl LDHs and PS-CoAl LDHs.

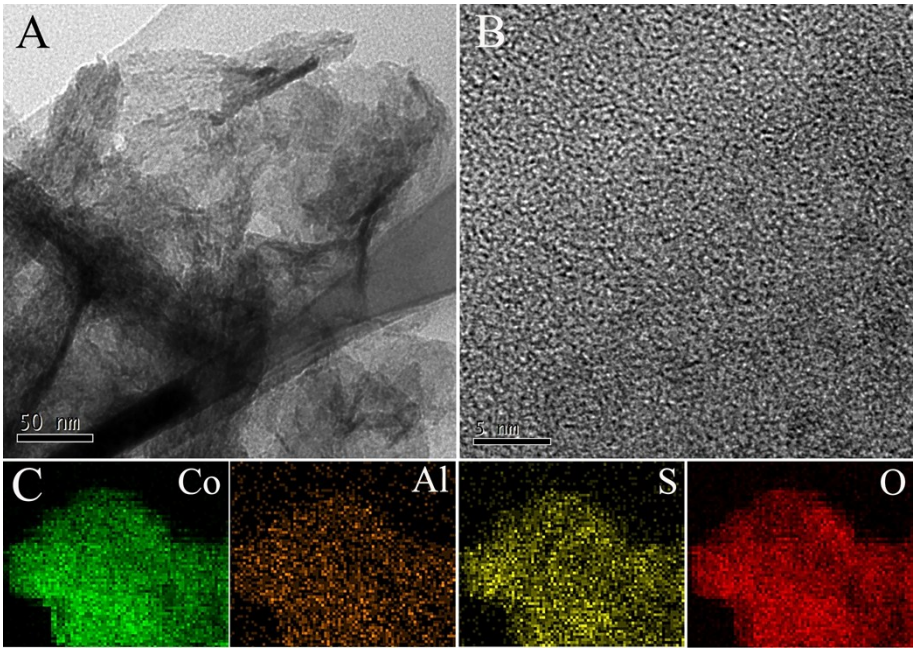

Figure S14. TEM and HRTEM images (A and B) of PS-CoAl LDHs after the electrochemical durability test; B) TEM-EDX mapping images of PS-CoAl LDHs after the electrochemical durability test.

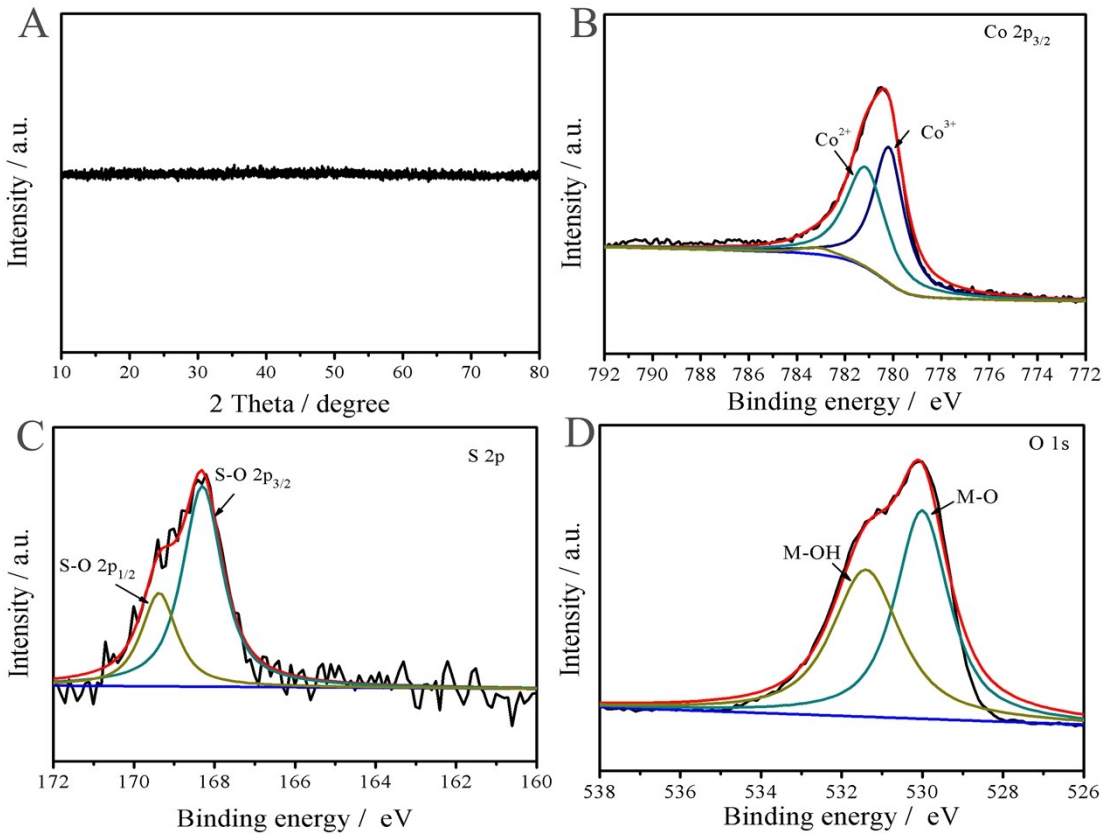

Figure S15. A) XRD pattern of PS-CoAl LDHs after the electrochemical durability test; B) Co $2p_{3/2}$, C) S 2p, and D) O 1s spectra of PS-CoAl LDHs after the electrochemical durability test.

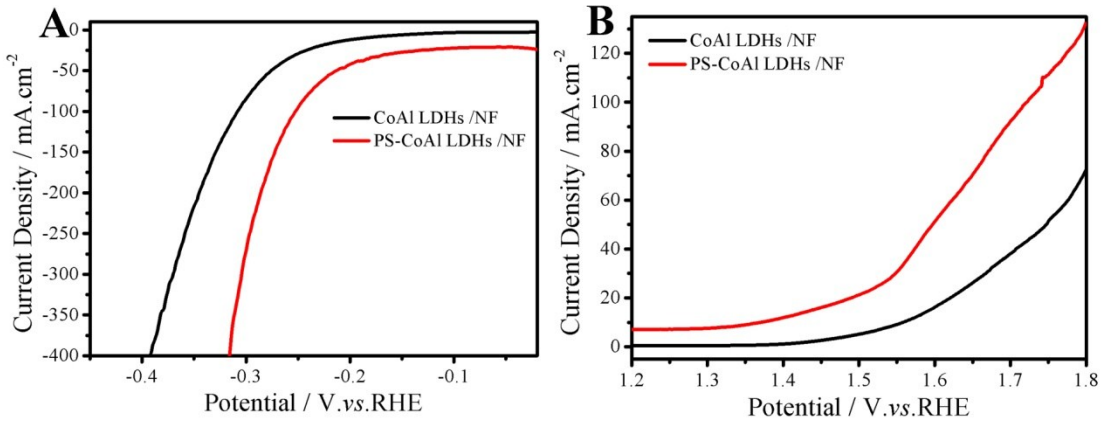

Figure S16. A) LSV polarization curves of CoAl LDHs and PS-CoAl LDH on NF; B) LSV polarization curves of CoAl LDHs and PS-CoAl LDH on NF for overall water splitting.

Table S1. Summarization the content of different elements in CoAl LDHs and PS-CoAl LDHs

Sample	Co (At%)	Al (At%)	S (At%)	O (At%)
CoAl LDHs	20.35	7.86	0	71.79
PS-CoAl LDHs	10.12	0.83	30.99	58.06

At %:atom percentage content

Table S2.Comparison results of the XPS for Co $2p_{3/2}$ spectra of CoAl LDHs, S-CoAl LDHs, and PS-CoAl LDHs.

Co species (At %)-CoAl LDHs	**Co species (At %)-S-CoAl LDHs**		**Co species (At %)-PS-CoAl LDHs**	
Co^{2+}-OH/O	Co^{2+}-OH/O	Co-S	Co^{2+}-OH/O	Co-S
54.75	52.65	47.35	30.61	69.39

Table S3.Comparison results of the XPS for O 1s spectra of CoAl LDHs, S-CoAl LDHs, and PS-CoAl LDHs.

O species (At %)-CoAl LDHs	**O species (At %)-S-CoAl LDHs**	**O species (At %)-PS-CoAl LDHs**		
M-OH	M-OH	M-O	M-OH	H_2O
100	100	25.21	56.22	18.57

Table S4. Comparison of OER activity of the PS-CoAl LDHs with recently reported catalyst.

Catalysts	Electrolyte solution	Substrate	η (mV) at 10 mA cm^{-2}	Tafel slope (mV dec^{-1})	Reference
PS-CoAl LDHs	1 M KOH	glass carbon electrode	329	61	**This work**
$Co_{0.5}Fe_{0.5}S$@N-MC	1 M KOH	glass carbon electrode	410	159	S[1]
Co_9S_8/S-C	1 M KOH	glass carbon electrode	339	64	S[2]
Co-P film	1 M KOH	copper foils	345	47	S[3]
$Co_5AlS_{1.5}(OH)_{10}$	0.1M KOH	glass carbon electrode	414		S[4]
CoS-$Co(OH)_2$@a MoS_{2+x}	1 M KOH	glass carbon electrode	380	68	S[5]
Monocrystalline CoZn hydroxides	1 M KOH	Zn foil	430	68	S[6]
Ultrathin CoMn LDHs	1 M KOH	glass carbon electrode	350	43	S[7]
$Co(OH)(C_6H_5COO)\cdot H_2O$ Nanobelts	1 M KOH	glass carbon electrode	360	76	S[8]
NiFe@g-C_3N_4/CNTs	1 M KOH	glass carbon electrode	326	67	S[9]
$Co_3O_4/NiCo_2O_4$ double-shelled nanocages	1 M KOH	Ni foam	340	88	S[10]

References:

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