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One-Step, Room Temperature Generation of Porous and Amorphous Cobalt Hydroxysulfides from Layered Double Hydroxides for Superior Oxygen Evolution Reactions

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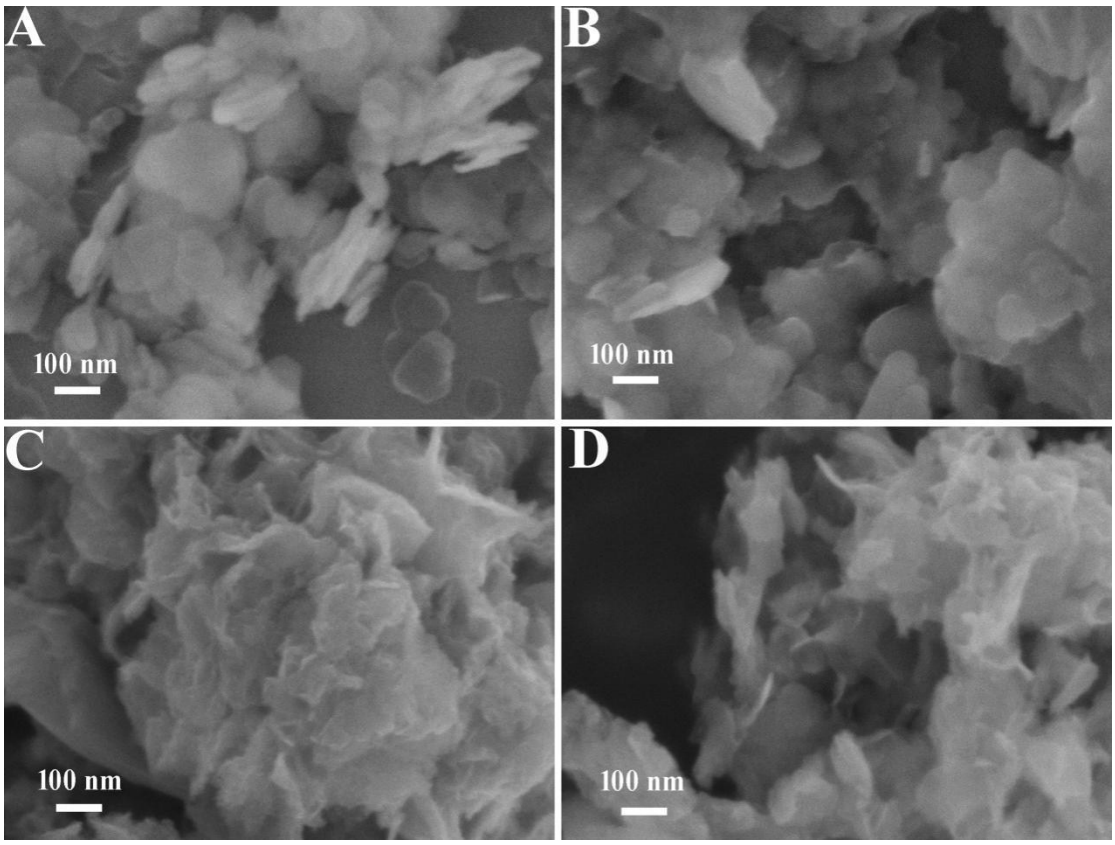

Figure S1. The SEM images of (A) CoAl-LDH, (B) OE-CoAl-LDH, (C) OS-CoAl-LDH (D) PA-$CoS_x(OH)_y$

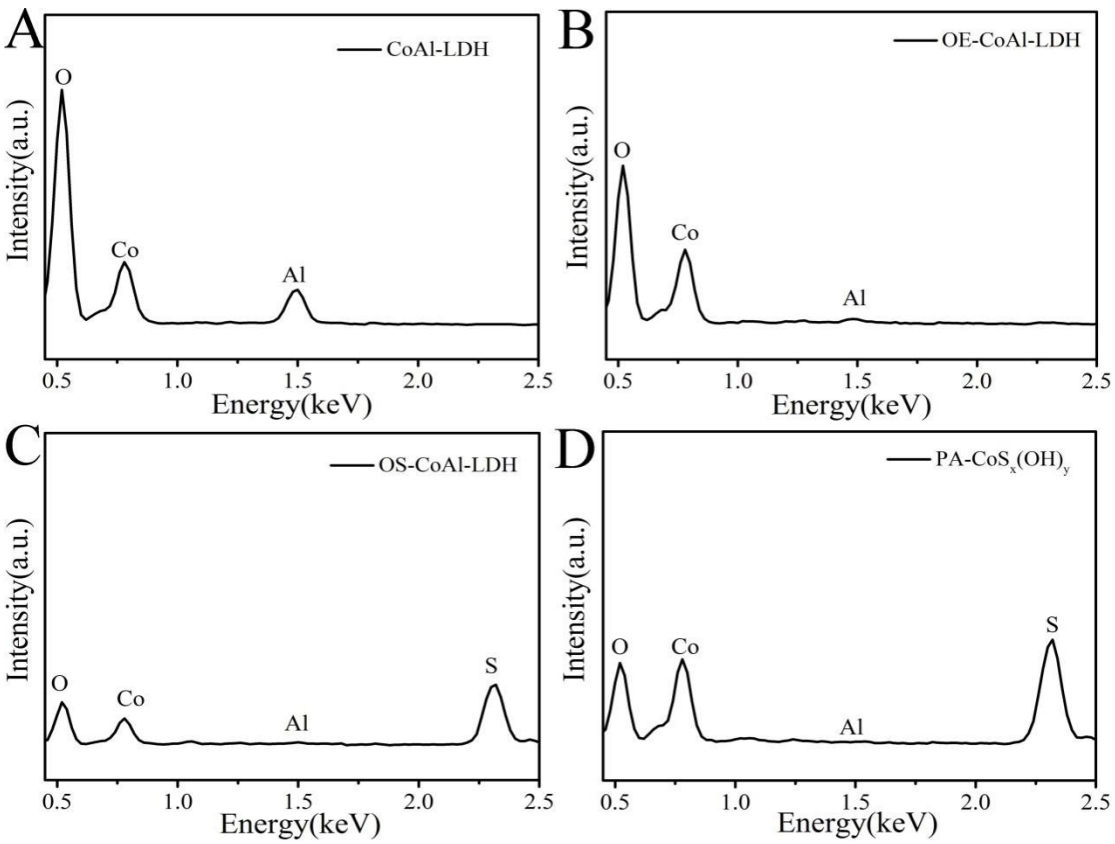

Figure S2. The EDX spectrums of (A) CoAl-LDH, (B) OE-CoAl-LDH, (C) OS-CoAl-LDH (D) PA-$CoS_x(OH)_y$

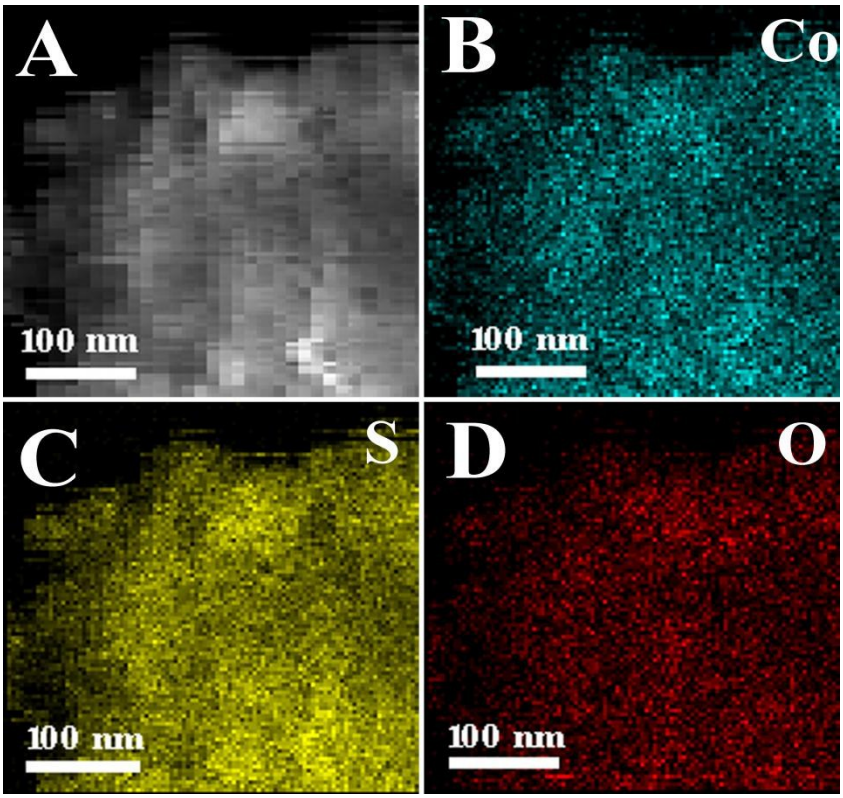

Figure S3. (A) The low-resolution TEM image of the PA-$CoS_x(OH)_y$ and corresponding elemental mapping images of (B) Co, (C) S, and (D) O in the PA-$CoS_x(OH)_y$.

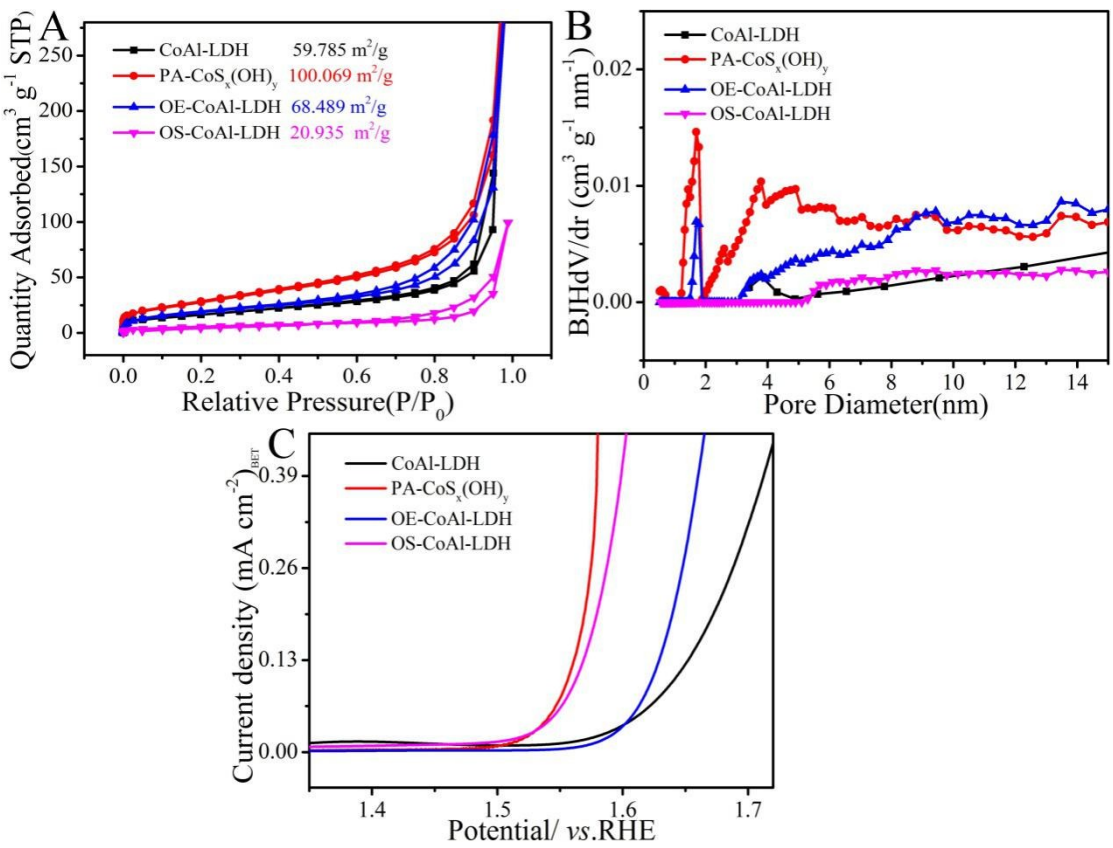

Figure S4. (A) N_2 adsorption-desorption isotherms of CoAl-LDH, PA-$CoS_x(OH)_y$, OE-CoAl-LDH and OS-CoAl-LDH. (B) Corresponding BJH pore size distribution of CoAl-LDH, PA-$CoS_x(OH)_y$, OE-CoAl-LDH and OS-CoAl-LDH. (C) The OER performance of the CoAl-LDH,PA-$CoS_x(OH)_y$, OE-CoAl-LDH and OS-CoAl-LDH after BET normalization in 1.0M KOH.

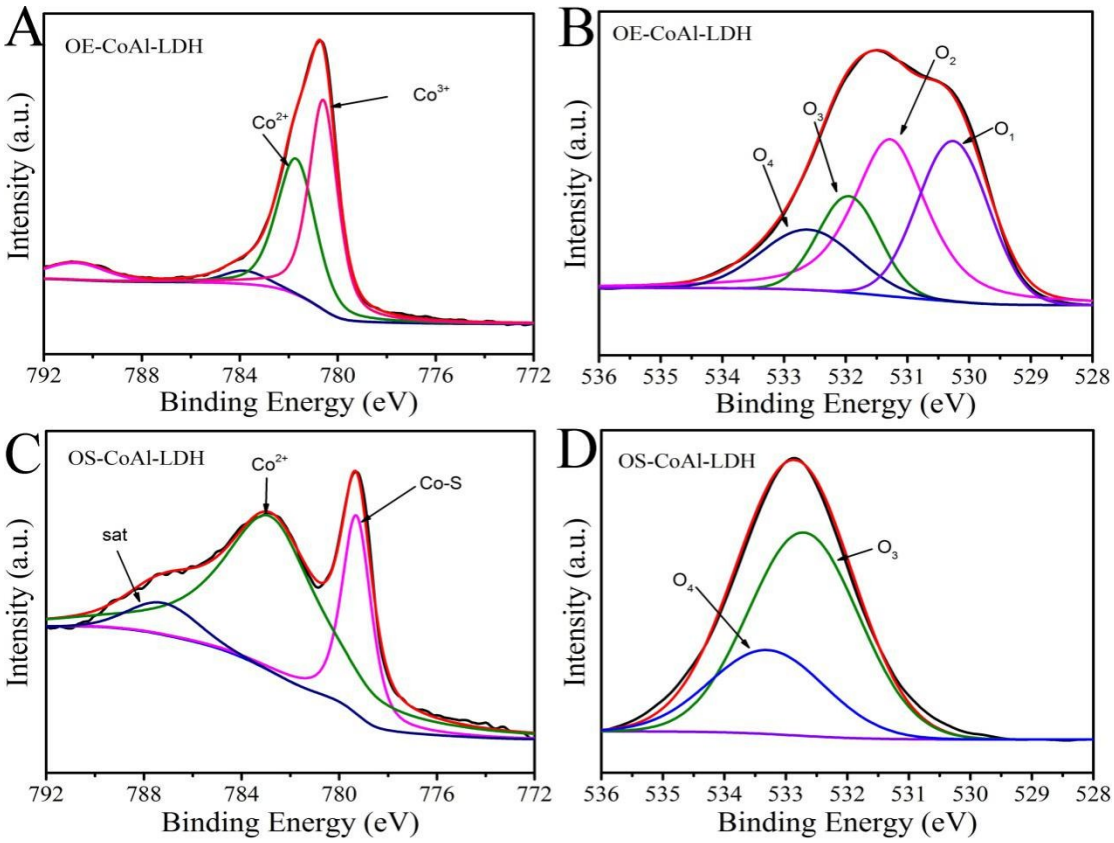

Figure S5. The XPS spectrums. (A) Co $2P_{3/2}$ of OE-CoAl-LDH, (B) O 1s of OE-CoAl-LDH (C) Co $2P_{3/2}$ of OS-CoAl-LDH (D) O 1s of OS-CoAl-LDH

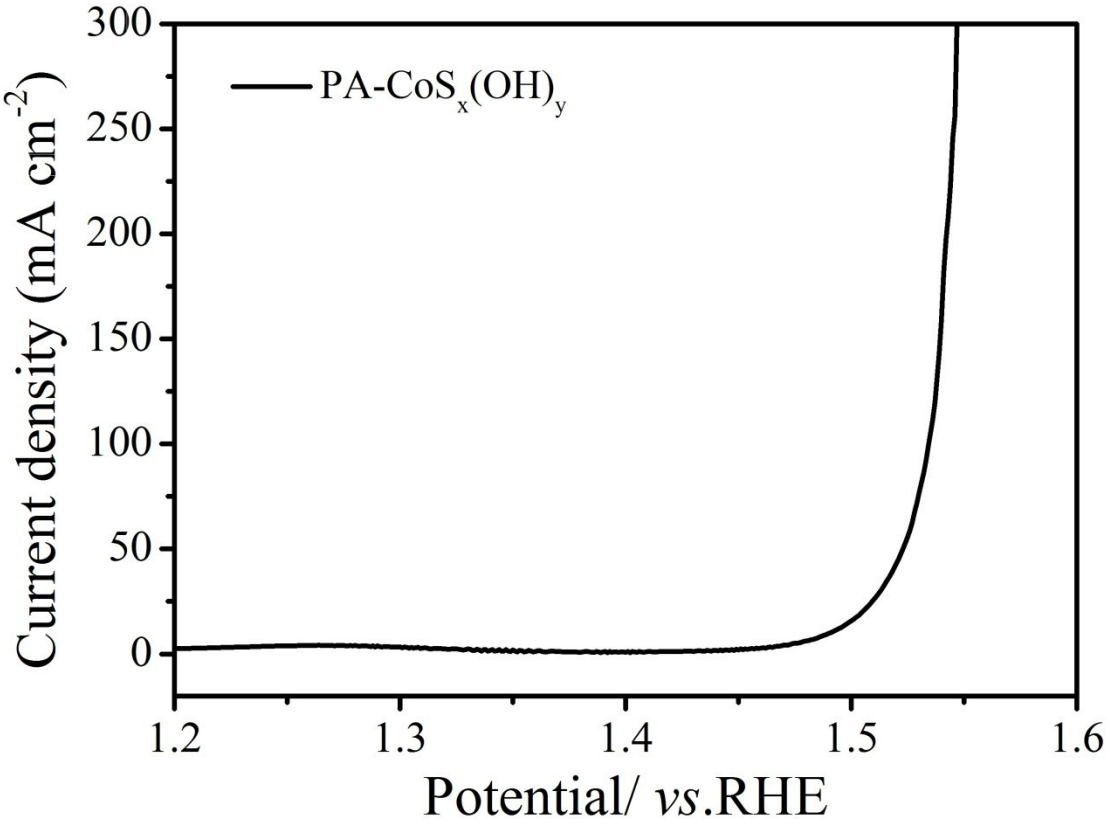

Figure S6.LSV polarization curves of PA-$CoS_x(OH)_y$/NF

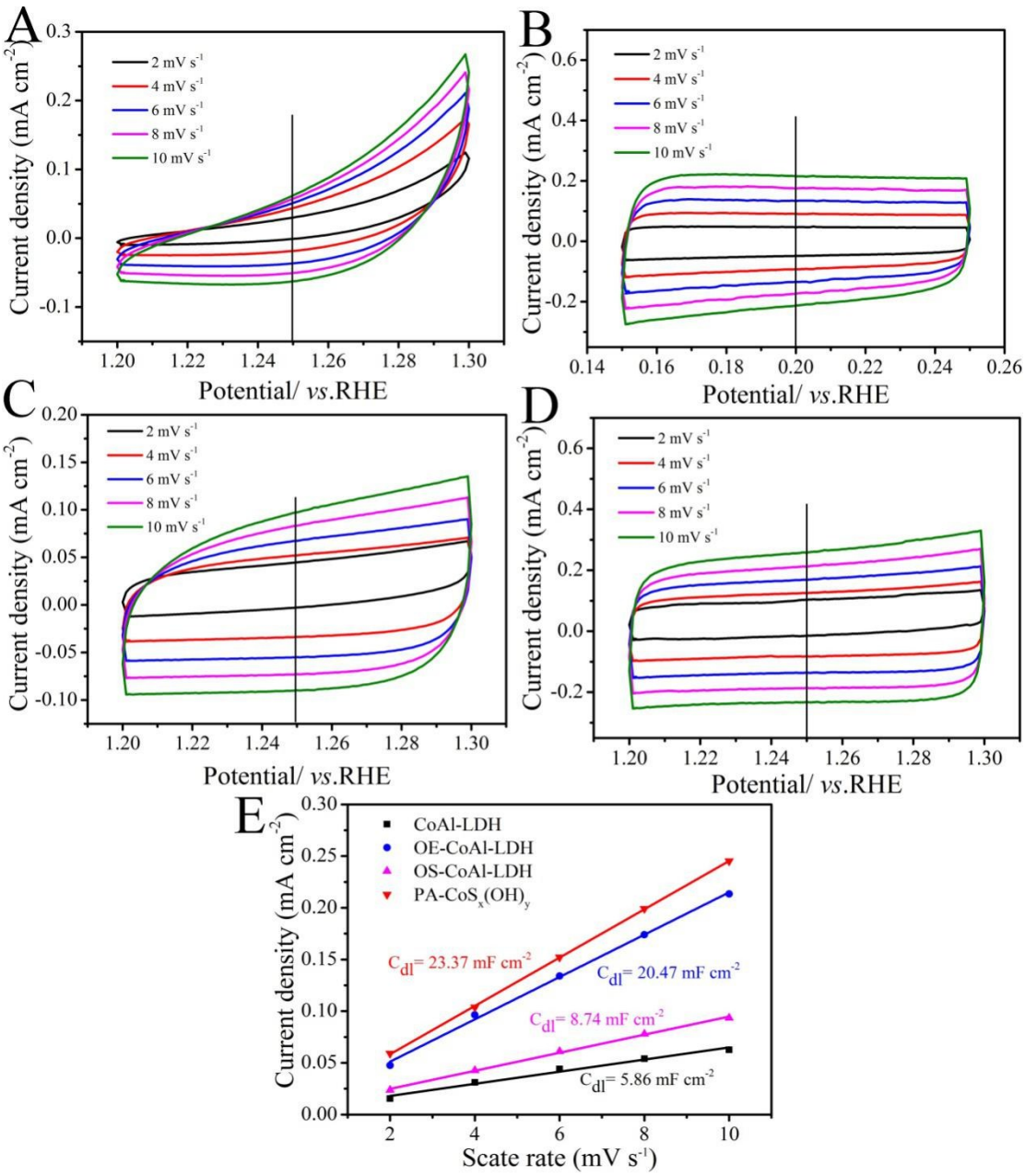

Figure S7. Tested the electrochemical surface area (ECSA) of CoAl-LDH, OE-CoAl-LDH，OS-CoAl-LDH and PA-$CoS_x(OH)_y$ in 1.0 M KOH solution. (A) Cyclic voltammetry curves of (A) CoAl-LDH, (B) OE-CoAl-LDH，(C) OS-CoAl-LDH and (D) PA-$CoS_x(OH)_y$ with different scanning rates. The capacitive current measured at 1.25 V vs RHE was plotting as a function of scan rate (E) CoAl-LDH, OE-CoAl-LDH, OS-CoAl-LDH and PA-$CoS_x(OH)_y$.

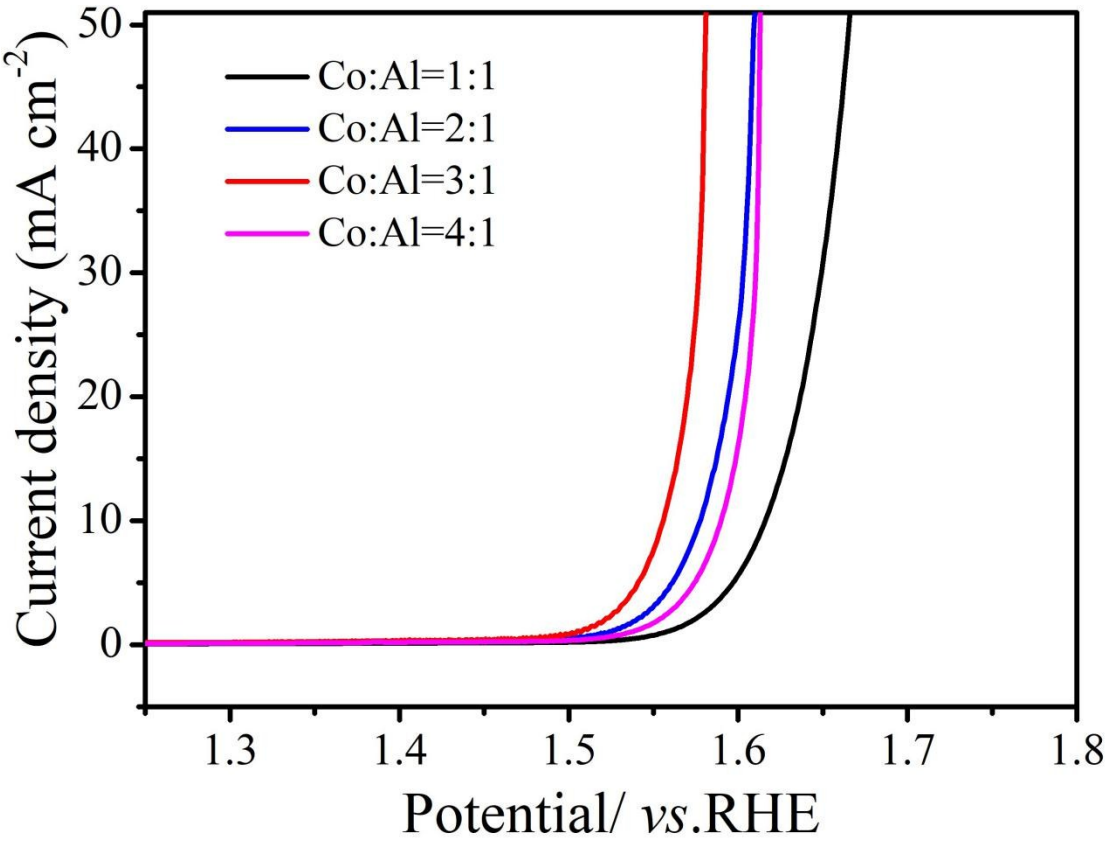

Figure S8. The OER performances of those samples with different Co:Al ratios were treated in mixed solution of 5 M NaOH and 3 M Na_2S.

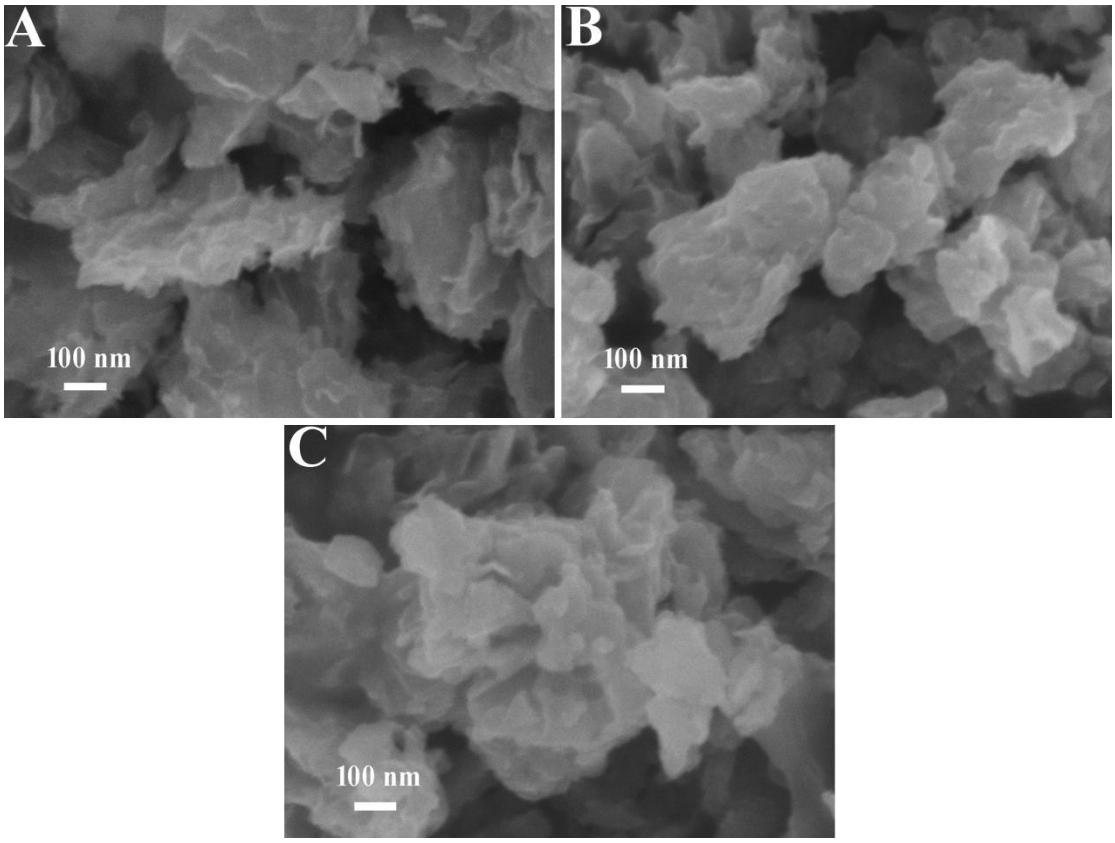

Figure S9. The SEM images of the CoAl-LDH nanosheets treatment conditions with different concentrations of NaOH (A) 2 M, (B) 8M, and (C) 10 M.

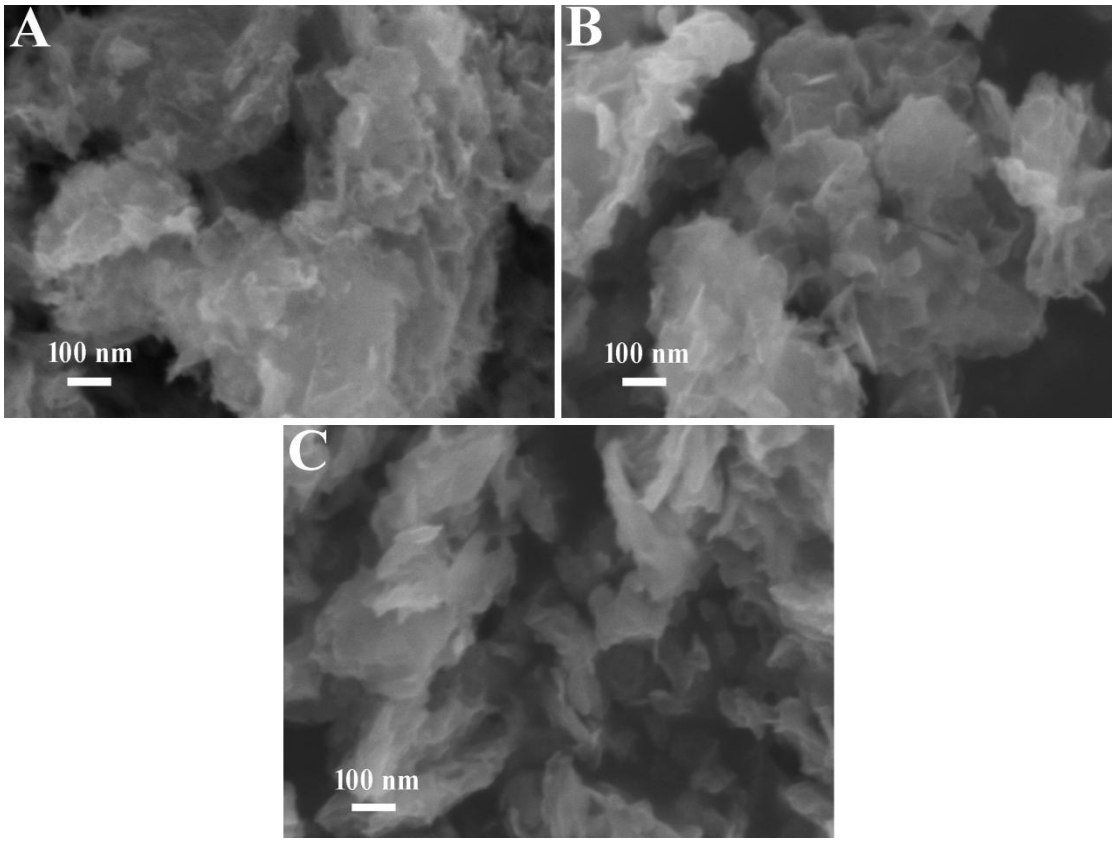

Figure S10. The SEM images of the CoAl-LDH nanosheets treatment conditions with different concentrations of Na_2S (A) 1 M, (B) 2M, and (C) 5 M.

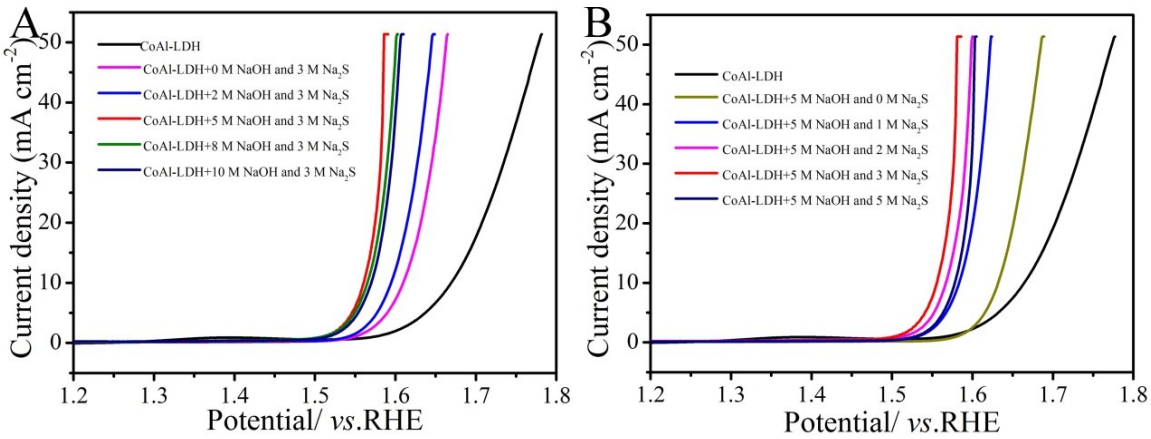

Figure S11. (A) The OER performances of the nanosheets treatment conditions with different concentrations of NaOH. (B) The OER performances of the nanosheets treatment conditions with different concentrations of Na_2S.

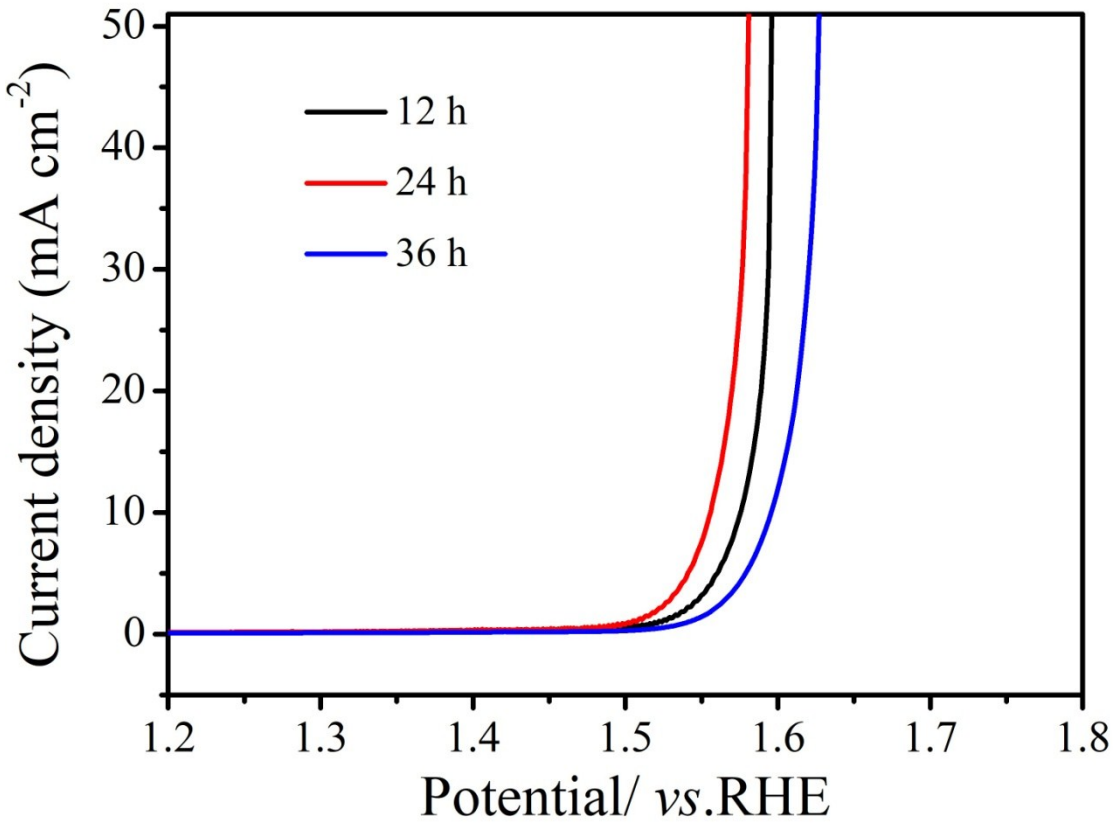

Figure S12.The OER performances of those samples treated at different times in mixed solutions of 5M NaOH and 3 M Na_2S.

Table S1. The element analysis of the CoAl-LDH, OE-CoAl-LDH，OS-CoAl-LDH and PA-$CoS_x(OH)_y$

Materials	Co (atom percentage content, %)	Al (atom percentage content, %)	S (atom percentage content, %)	O (atom percentage content, %)	The element mole ratio of Co/Al	The element mole ratio of Co/S
CoAl-LDH	18.65	5.04	-	65.35	3.70	-
5M NaOH+3M Na_2S	17.31	0.14	15.08	24.63	123.64	1.15
5M NaOH	15.64	0.46	-	39.81	34.00	-
3M Na_2S	7.51	0.3	12.28	22.33	25.03	0.61

Table S2.Comparison of OER performance for PA-$CoS_x(OH)_y$ with other Co-based materials in 1.0M KOH.

Electrocatalyst	Electrolyte solution	Current density (mA cm^{-2})	Overpotential at the corresponding Current density (mV)	Reference
PA-$CoS_x(OH)_y$	1M KOH	10	320	This work
PA-$CoS_x(OH)_y$/NF	1M KOH	10	261	
Co-S/Ti	1M KOH	10	361	[1]
Bulk CoS_2	1M KOH	10	480	[2]
CoOx film	1M KOH	10	403	[3]
CoCo LDH	1M KOH	10	393	[4]
Co-P film	1M KOH	10	345	[5]
Co-C_3N_4/CNT	1M KOH	10	380	[6]
Ni–Co Double Hydroxides Nanocage	1M KOH	10	350	[7]
$NiCo_2O_4$ nanowire/Ti	1M KOH	10	360	[8]
Co_3S_4-L	1M KOH	10	360	[9]
CoS2/N,S-GO	1M KOH	10	380	[10]

Table S3. The element analysis of the treatment of CoAl-LDH with different sulfuration time.

Materials	Co (atom percentage content, %)	Al (atom percentage content, %)	S (atom percentage content, %)	O (atom percentage content, %)	The element mole ratio of Co/S
5MNaOH+3M Na_2S - 12 h	11.26	0.33	8.93	59.04	1.26
5MNaOH+3M Na_2S - 24 h	17.31	0.14	15.08	24.63	1.15
5MNaOH+3M Na_2S - 36h	8.64	0.14	16.46	36.76	0.52

Reference

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