Supporting Information

Cobalt doping of $FePS_3$ promotes intrinsic active sites for efficient hydrogen evolution reaction

Shuang Wang[a, †], Beibei Xiao[b, †], Shijie Shen[a,*], Kai Song[a], Zhiping Lin[a], Zongpeng Wang[a], Yuchao Chen[a], Wenwu Zhong[a,*]

[a] School of Pharmaceutical and Materials Engineering, Taizhou University, Taizhou 318000, China.

[b] School of Energy and Power Engineering Jiangsu University of Science and Technology Zhenjiang, Jiangsu, 212003, China.

[†] Shuang Wang and Beibei Xiao contributed equally to this work.

E-mail: shensj@tzc.edu.cn, zhongww@tzc.edu.cn.

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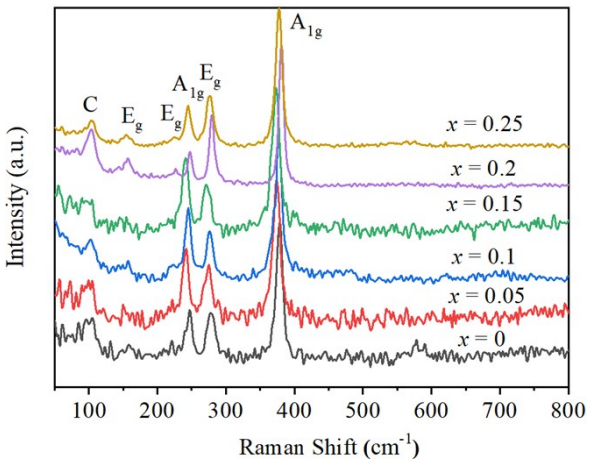

Figure S1. Raman spectra for $Fe_{1-x}Co_xPS_3$ (x = 0, 0.05, 0.1, 0.15, 0.2, 0.25).

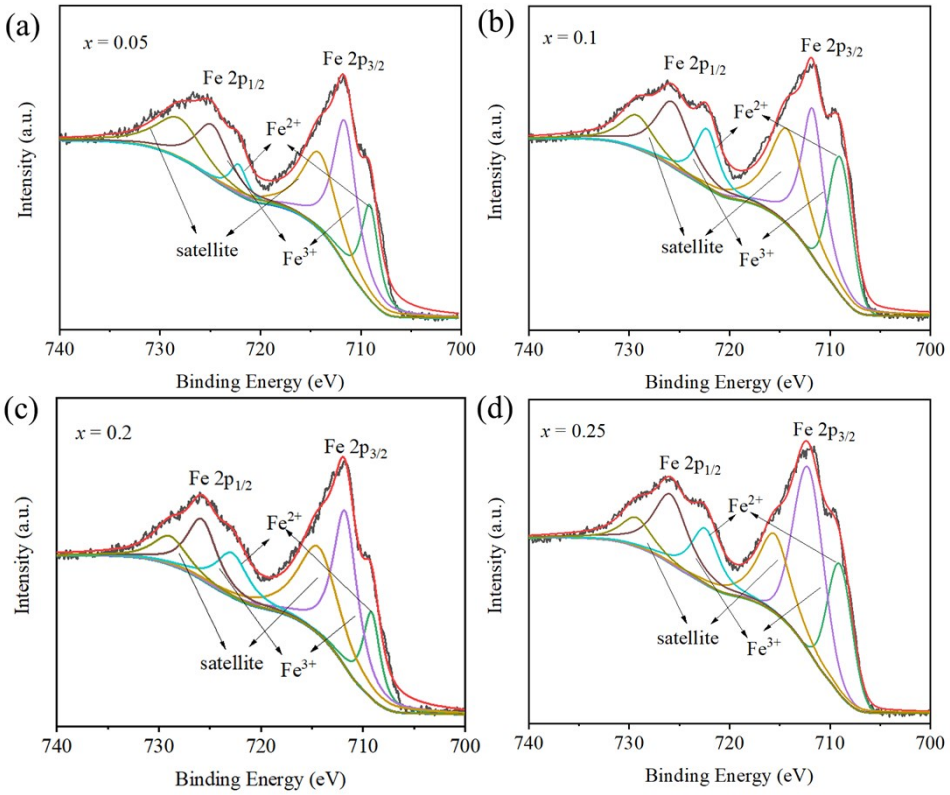

Figure S2. High-resolution XPS of Fe 2p electrons for $Fe_{1-x}Co_xPS_3$ (x = 0.05, 0.1, 0.2, 0.25).

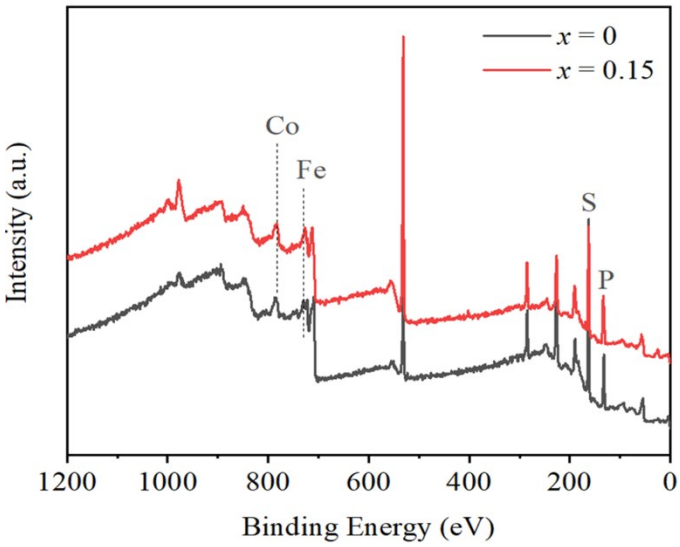

Figure S3. XPS survey for $Fe_{1-x}Co_xPS_3$ (x = 0, 0.15).

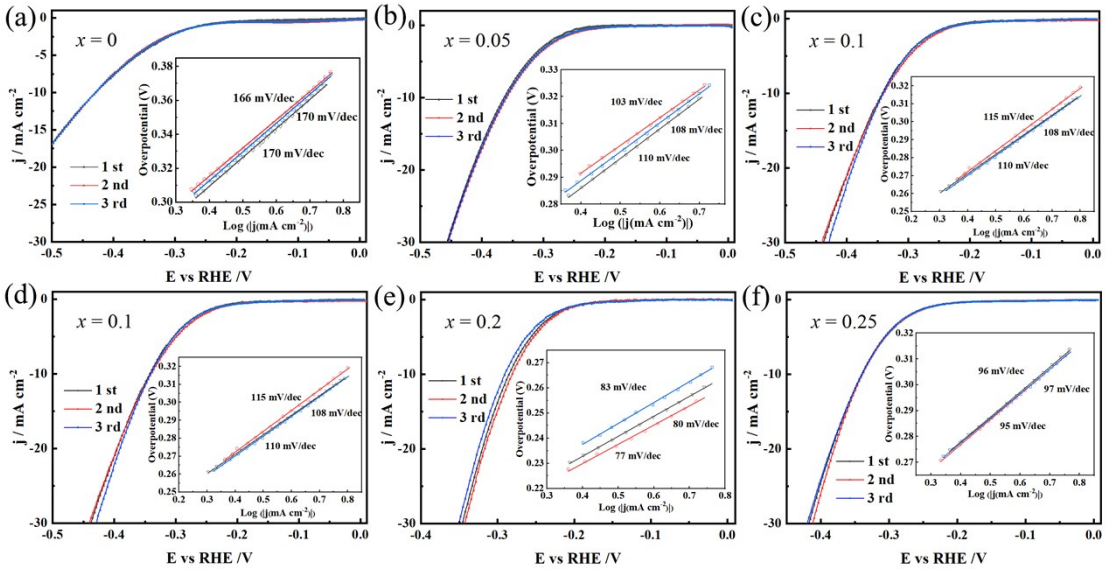

Figure S4. Linear sweep voltammetry curves and Tafel curves (inset) after i-R correction for $Fe_{1-x}Co_xPS_3$ (x = 0, 0.05, 0.1, 0.15, 0.2, 0.25) samples from three independent tests.

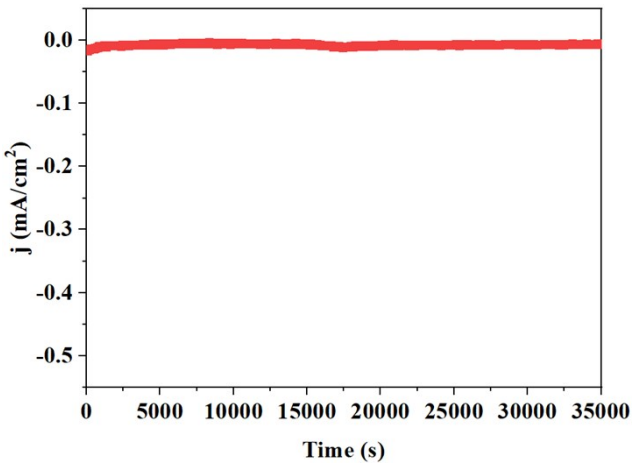

Figure S5. Time-dependent change of current density at a constant overpotential of 100 mV for $Fe_{0.85}Co_{0.15}PS_3$ as a HER catalyst.

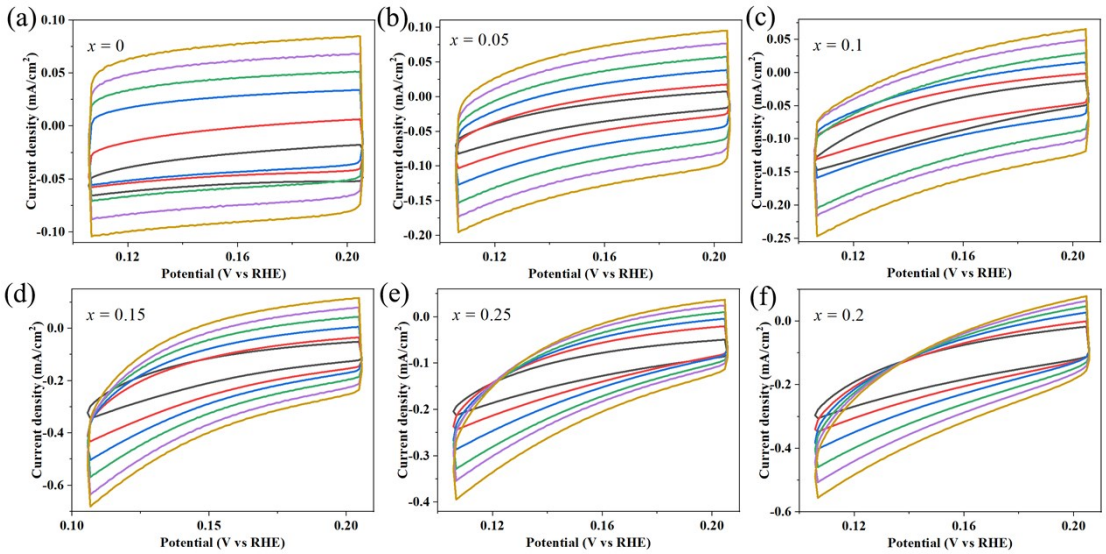

Figure S6. Cyclic voltammetry curves of $Fe_{1-x}Co_xPS_3$ (x = 0, 0.05, 0.1, 0.15, 0.2, 0.25) at various scan rates (20 mV, 40 mV, 80 mV, 120 mV, 160 mV, 200 mV,).

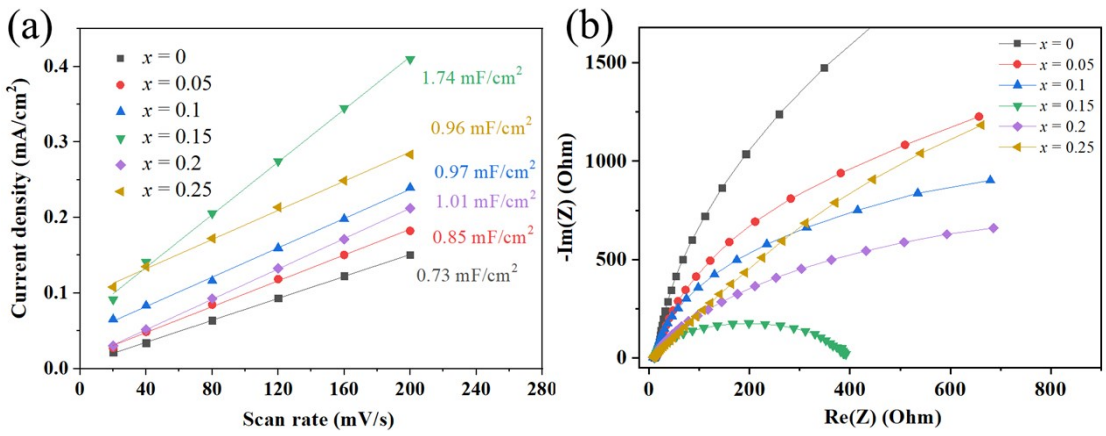

Figure S7. (a) The plots showing the derived C_{dl} and (b) EIS Nyquist plots for $Fe_{1-x}Co_xPS_3$ (x = 0, 0.05, 0.1, 0.15, 0.2, 0.25).

Table S1. Fe/Co atomic ratios of $Fe_{1-x}Co_xPS_3$ (x = 0.05, 0.1, 0.15, 0.2, 0.25) samples by ICP.

Sample (x)	0.05	0.1	0.15	0.2	0.25
Fe/Co (nominal)	19	9	5.7	4	3
Fe/Co (by ICP)	19.3	8.4	5.2	3.5	2.8

Table S2. Comparison of HER activity for related MPS_3 (M = Fe, Co, Ni, Mn, Zn, Cd, Sn, etc.).

Material	Overpotential (mV)	Tafel slope (mV/dec)	Electrolyte	Reference
$Fe_{0.85}Co_{0.15}PS_3$ crystal	263	80	0.5M H_2SO_4	This work
$FePS_3$ nanosheets (about 8 atomic layers)	241	94	0.5M H_2SO_4	*J. Mater. Chem. A*, 2019, **7**, 13928-13934.
$FePS_3$ crystal	>950	~200	0.5M H_2SO_4	*ACS Appl. Mater. Inter.*, 2017, **9**, 12563-12573.
$FePS_3$ nanosheets (few layer)	211	42	0.5M H_2SO_4	*ACS Energy Lett.*, 2016, **1**, 367-372.

$FePS_3$ nanosheets/rGO (few layer)	108	54	0.5M H_2SO_4	*ACS Energy Lett.*, 2016, **1**, 367-372.
$FePS_3$ nanosheets (about 5 atomic layers)	>350	109	1M KOH	*ACS Catal.*, 2017, **7**, 8549-8557.
$FePS_3$ nanocrystals/ carbon paper	175	137	1M KOH	*Small Methods*, 2019, 1900632
$CoPS_3$ crystal	~680	84	0.5M H_2SO_4	*ACS Appl. Mater. Inter.*, 2017, **9**, 12563-12573.
$NiPS_3$ crystal	~600	56	0.5M H_2SO_4	*ACS Appl. Mater. Inter.*, 2017, **9**, 12563-12573.
$NiPS_3$ nanosheets (few layer)	297	69	0.5M H_2SO_4	*Chemelectrochem*, 2016, **3**, 1392-1399.
$SnPS_3$ crystal	>1100	~150	0.5M H_2SO_4	*ACS Appl. Mater. Inter.*, 2017, **9**, 12563-12573.
$ZnPS_3$ crystal	>1300	75	0.5M H_2SO_4	*ACS Appl. Mater. Inter.*, 2017, **9**, 12563-12573.
$CdPS_3$ crystal	~1350	~140	0.5M H_2SO_4	*ACS Appl. Mater. Inter.*, 2017, **9**, 12563-12573.
$MnPS_3$ nanosheets (~38 nm)	~835	not given	0.5M H_2SO_4	*Adv. Funct. Mater.*, 2019, **29**, 1805975.

Table S3. Electrical resistivity under room temperature for $Fe_{1-x}Co_xPS_3$ (x = 0, 0.05, 0.1, 0.15, 0.2, 0.25).

Sample (x)	0	0.05	0.1	0.15	0.2	0.25
Resistivity (Ω m)	83	44	32	21	38	51