

## Electronic Supplementary Information

# Interfacial Confinement of Ultra-Low Pt Single Atoms *via* Co-Pt-C/N Electronic Bridging for Efficient Hydrogen Evolution

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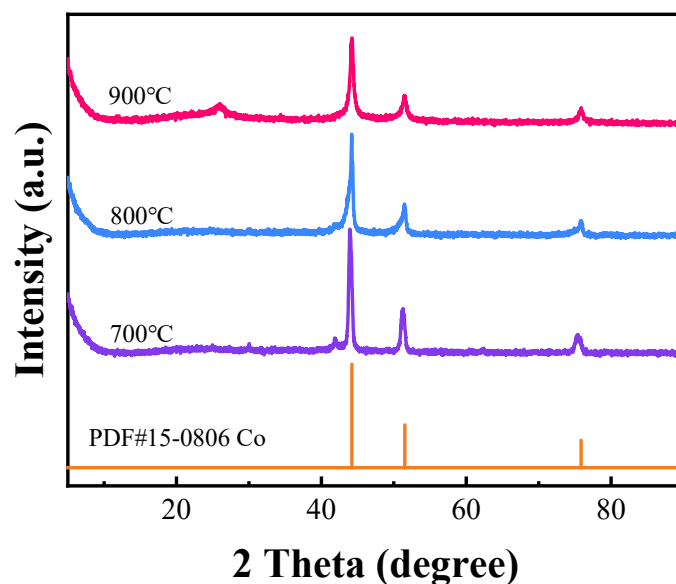
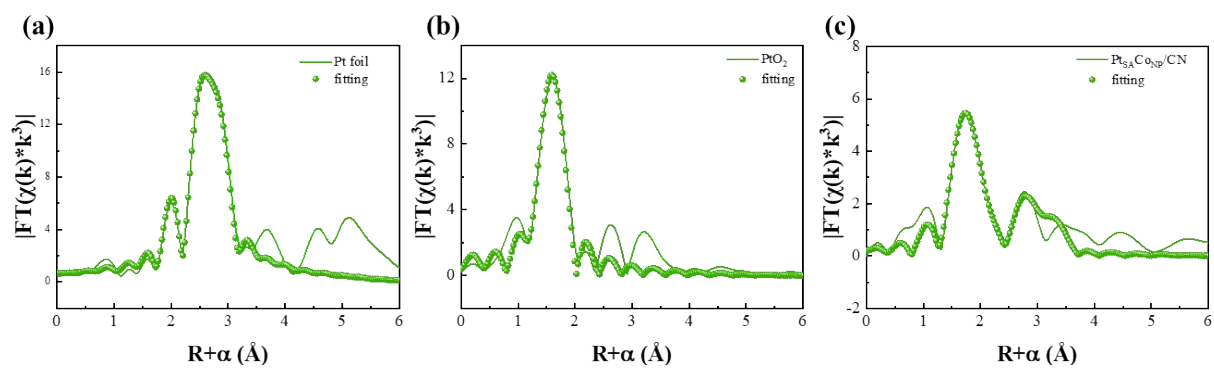
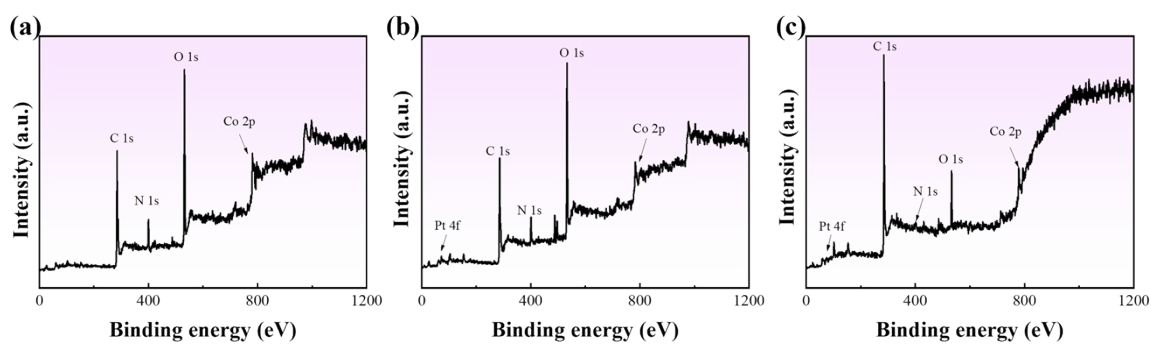


Fig.S1 XRD Patterns of Pt<sub>SA</sub>Co<sub>NP</sub>/CN pyrolysis at 700°C, 800°C, and 900°C, respectively.



**Fig. S2** Fitting of the Fourier transform  $k^3$ -weighted EXAFS spectra for Pt foil,  $\text{PtO}_2$ , and  $\text{Pt}_{\text{SA}}\text{CoNP}/\text{CN}$ .



**Fig. S3** XPS survey spectra of  $\text{Co-NH}_2\text{BDC}$ ,  $\text{PtCo-NH}_2\text{BDC}$ , and  $\text{Pt}_{\text{SA}}\text{CoNP}/\text{CN}$ .

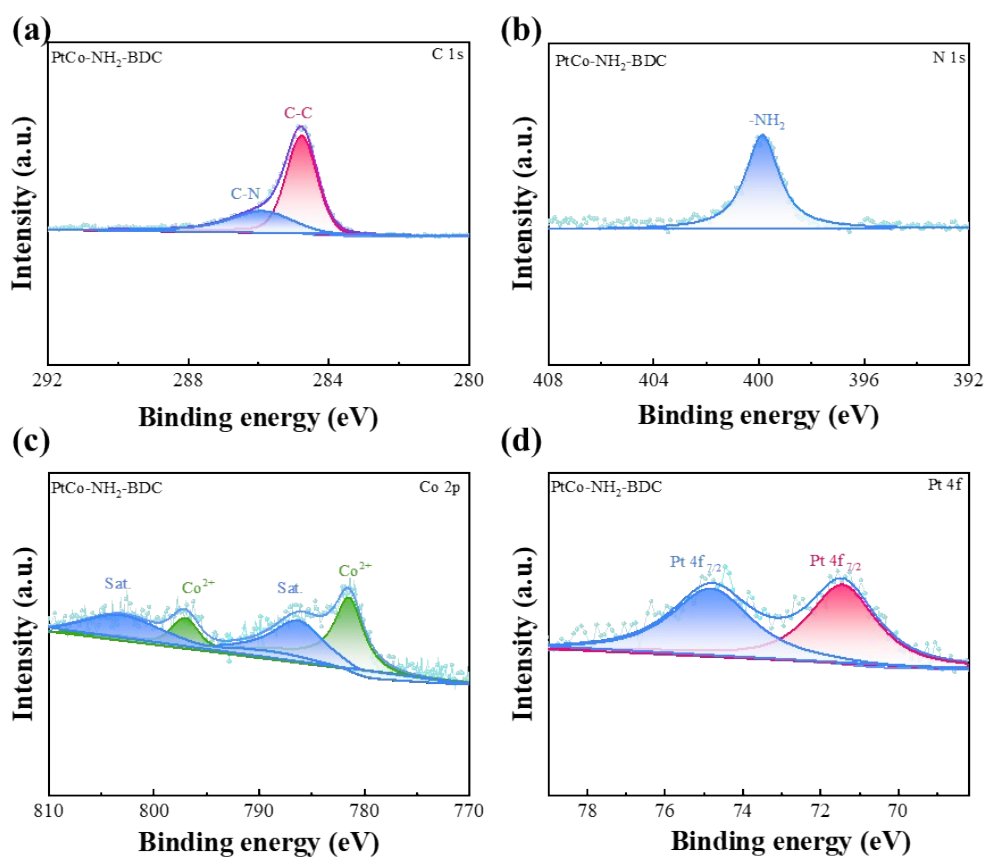
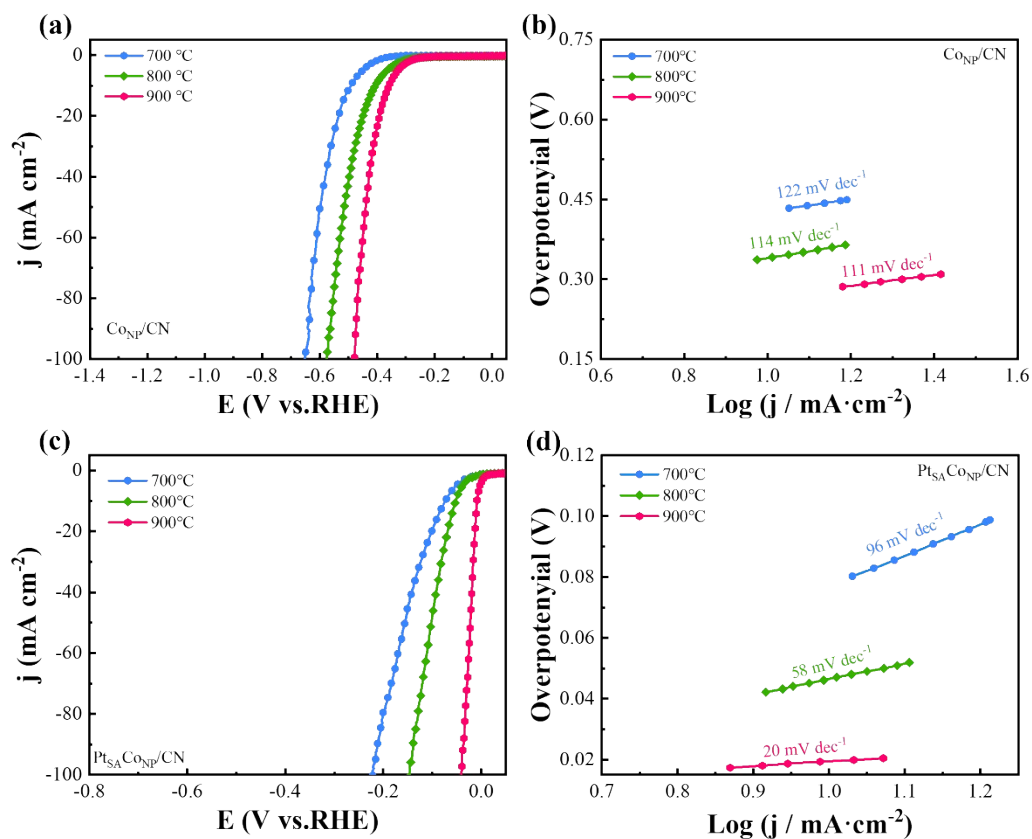
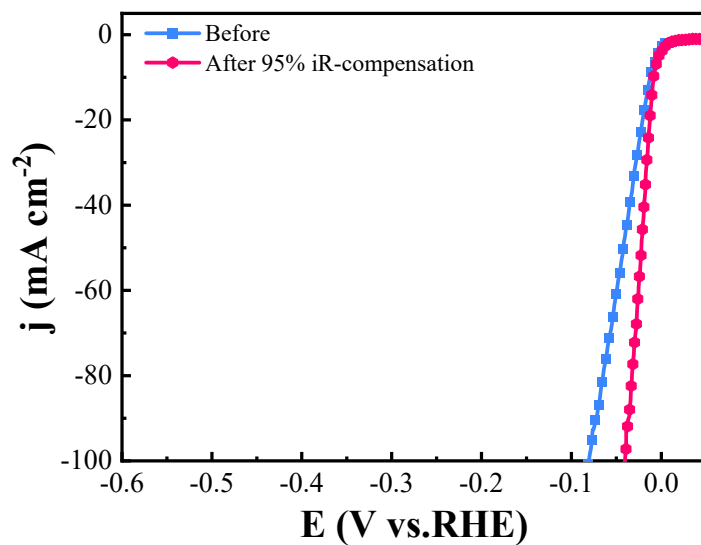


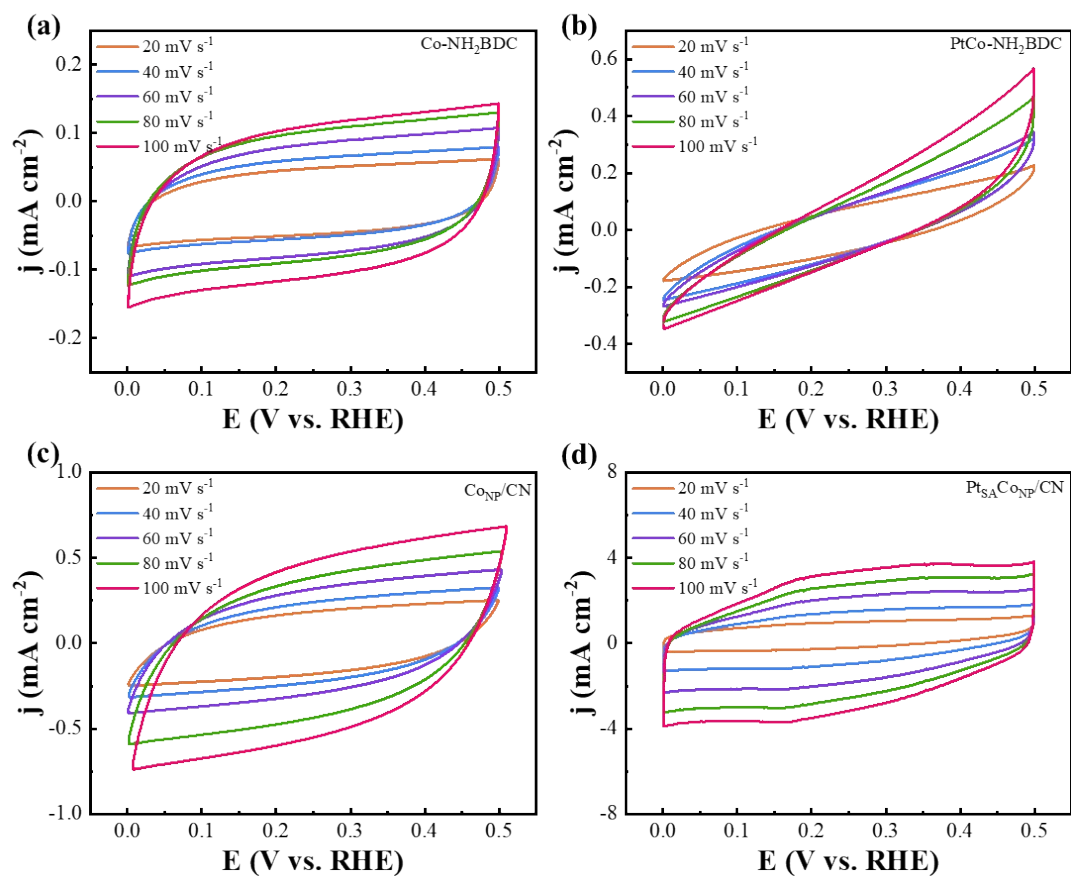
Fig. S4 C 1s, N 1s, Co 2p, and Pt 4f high-resolution XPS region of PtCo-NH<sub>2</sub>BDC.



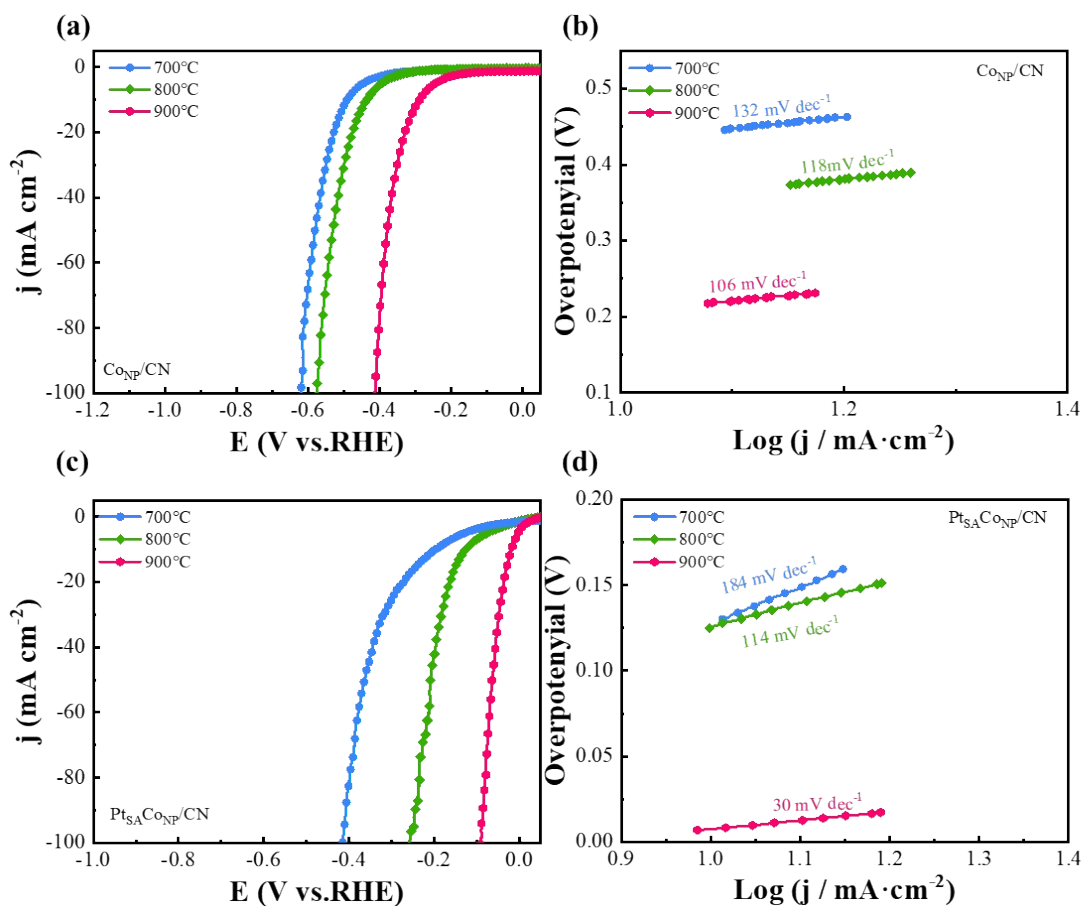
**Fig. S5** Comparative HER polarization curves and Tafel plots in 0.5 M H<sub>2</sub>SO<sub>4</sub> for (a, b) CoNP/CN and (c, d) Pt<sub>SA</sub>CoNP/CN catalysts pyrolyzed at different temperatures.



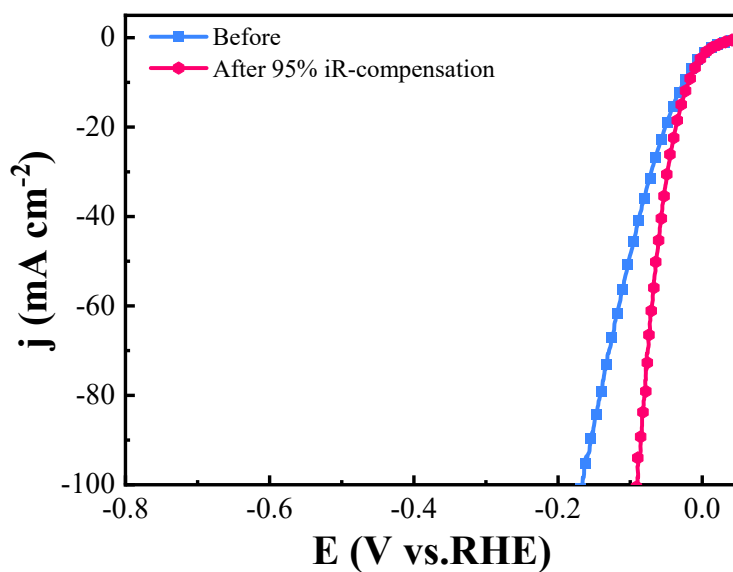
**Fig. S6** Polarization curves of Pt<sub>SA</sub>CoNP/CN (900 °C) in acidic conditions before and after iR compensation.



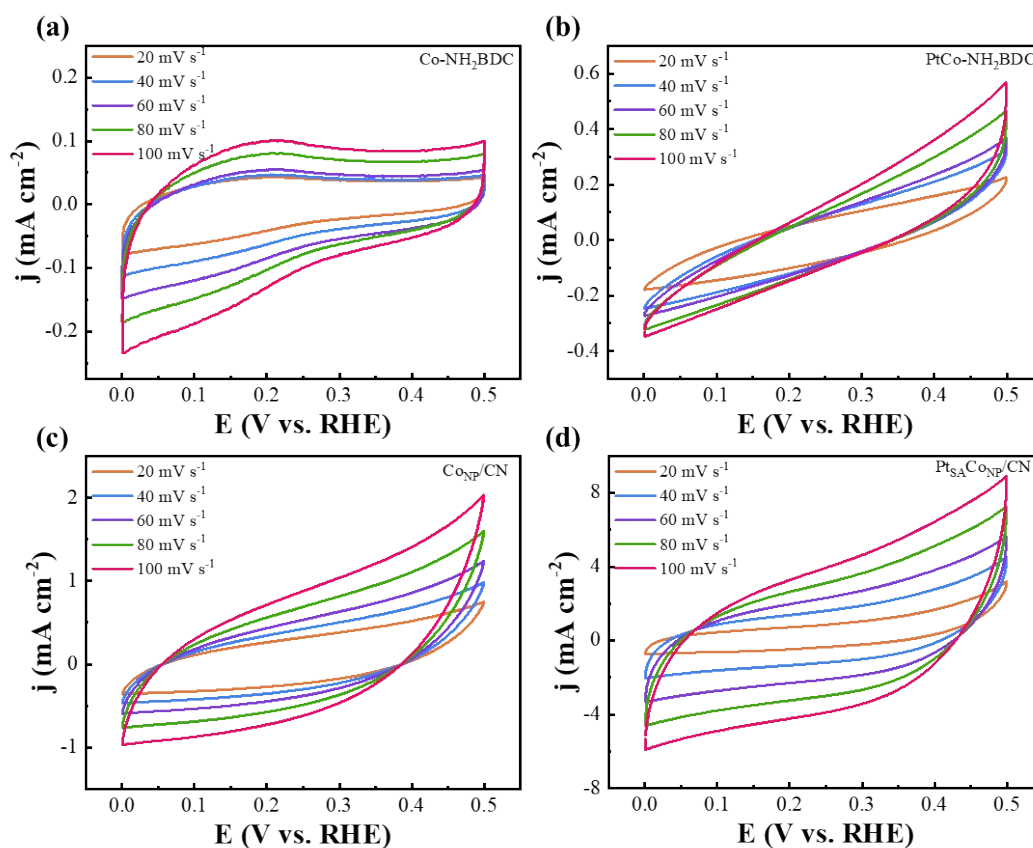
**Fig. S7** Cyclic voltammetry (CV) curves of (a)  $\text{Co-NH}_2\text{BDC}$ , (b)  $\text{PtCo-NH}_2\text{BDC}$ , (c)  $\text{Co}_{\text{NP}}/\text{CN}$  and (d)  $\text{Pt}_{\text{SA}}\text{Co}_{\text{NP}}/\text{CN}$  in  $0.5 \text{ M H}_2\text{SO}_4$ .



**Fig. S8** Comparative HER polarization curves and Tafel plots in 1.0 M KOH for (a, b)  $\text{Co}_{\text{NP}}/\text{CN}$  and (c, d)  $\text{Pt}_{\text{SA}}\text{Co}_{\text{NP}}/\text{CN}$  catalysts pyrolyzed at different temperatures.



**Fig. S9** Polarization curves of  $\text{Pt}_{\text{SA}}\text{Co}_{\text{NP}}/\text{CN}$  in alkaline conditions before and after iR compensation.



**Fig. S10** Cyclic voltammetry (CV) curves of (a) Co-NH<sub>2</sub>BDC, (b) PtCo-NH<sub>2</sub>BDC, (c) Co<sub>NP</sub>/CN and (d) Pt<sub>SA</sub>Co<sub>NP</sub>/CN in 1 M KOH.

**Table S1.** HER parameter of the catalysts under acidic conditions.

Electrocatalyst	$\eta_{10}$ /mV	Tafel/mV dec <sup>-1</sup>	$C_{dl}$ /mF cm <sup>-2</sup>	$R_{ct}$ /ohm
		1		
Co-NH <sub>2</sub> BDC	662	119	0.79	906
Co <sub>NP</sub> /CN(700°C)	493	122	/	/
Co <sub>NP</sub> /CN(800°C)	409	114	/	/
Co <sub>NP</sub> /CN(900°C)	363	111	4.24	138
PtCo-NH <sub>2</sub> BDC	290	200	0.83	441
Pt <sub>SA</sub> Co <sub>NP</sub> /CN(700°C)	77	96	/	/
Pt <sub>SA</sub> Co <sub>NP</sub> /CN(800°C)	51	58	/	/

Pt <sub>SA</sub> Co <sub>NP</sub> /CN(900°C)	8	20	30.77	54
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**Table S2.** HER parameters of the catalysts under alkaline conditions.

Electrocatalyst	$\eta_{10}/\text{mV}$	Tafel/ $\text{mV dec}^{-1}$	$C_{dl}/\text{mF cm}^{-2}$	$R_{ct}/\text{ohm}$
Co-NH <sub>2</sub> BDC	579	141	0.72	585
Co <sub>NP</sub> /CN(700°C)	494	132	/	/
Co <sub>NP</sub> /CN(800°C)	439	118	/	/
Co <sub>NP</sub> /CN(900°C)	287	106	6.01	259
PtCo-NH <sub>2</sub> BDC	416	240	0.82	386
Pt <sub>SA</sub> Co <sub>NP</sub> /CN(700°C)	208	184	/	/
Pt <sub>SA</sub> Co <sub>NP</sub> /CN(800°C)	124	114	/	/
Pt <sub>SA</sub> Co <sub>NP</sub> /CN(900°C)	18	30	41.20	43

**Table S3.** Comparison of the HER performance of advanced materials reported in the literature and in this work.

Catalysts	Acidic Overpotential ( $\eta$ )@j (mV @ mA cm <sup>-2</sup> )	Acidic Tafel (mV dec <sup>-1</sup> )	Alkaline Overpotential ( $\eta$ )@j (mV @ mA cm <sup>-2</sup> )	Alkaline Tafel (mV dec <sup>-1</sup> )	Ref.
Pt <sub>SA</sub> Co <sub>NP</sub> /CN	8@10	20	18@10	30	This work
Pt <sub>SA</sub> /Co <sub>AC</sub> -O@ACTP	61@10	43	77@10	53	1
Pt <sub>1</sub> Co <sub>6</sub> /np-Co <sub>2</sub> P	/	/	21@10	31	2
PtSe <sub>2</sub> /PtCo	33@10	22	/	/	3
CoPt <sub>3</sub> /a-FCWO-NS	/	/	135@10	80.69	4
CoPt@C	/	/	15@10	29	5
CoPt-Pt <sub>SA</sub> /NDPCF	/	/	31@10	43.6	6
CNs@CoPt	4.2@10	54.3	19.1@10	88.7	7
Co <sub>n</sub> -Pt <sub>1</sub> @NPC	/	/	20@10	29.07	8

Pt <sub>sa</sub> -PtCo NCs/N-CNTs-900	/	/	47@10	41	9
CoPt-Co <sub>SA</sub> @NSC	/	/	23@10	37	10

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