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## Electronic Supplementary Information

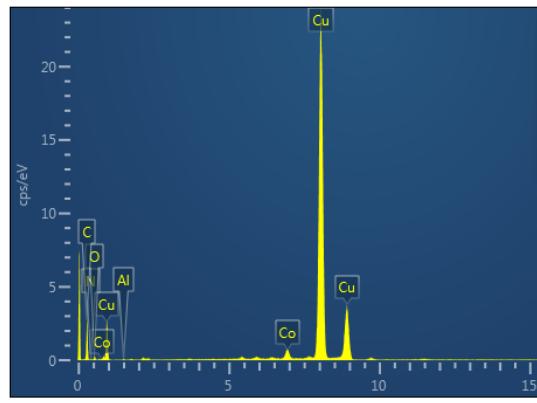
### **Boosting the hydrogen evolution activity of Co-N-C electrocatalyst by codoping with Al**

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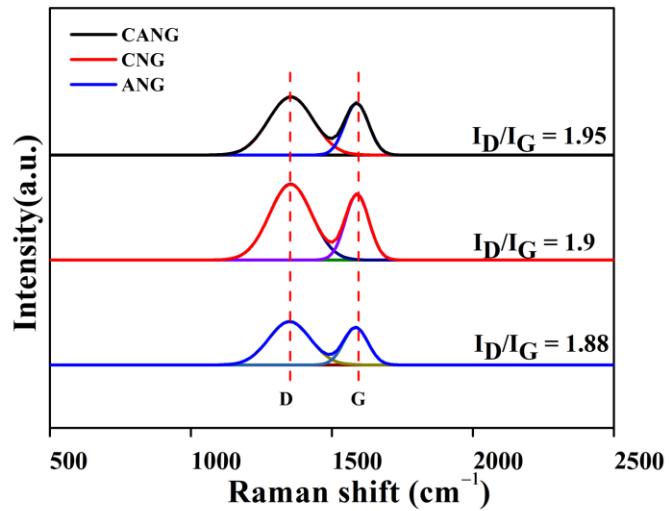
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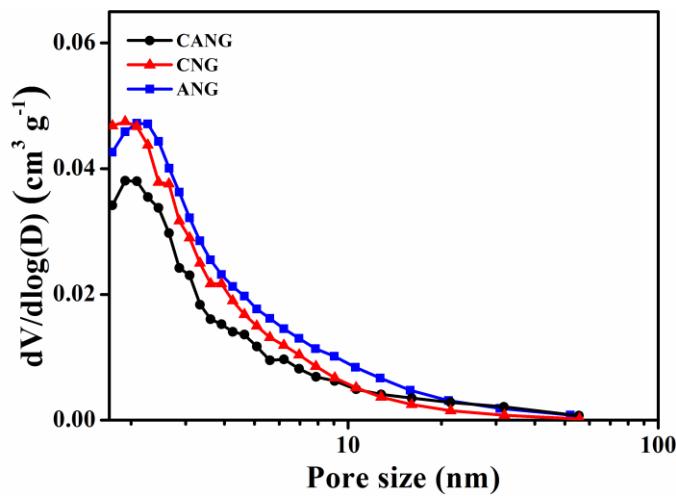
E-mail: qinyong@cczu.edu.cn



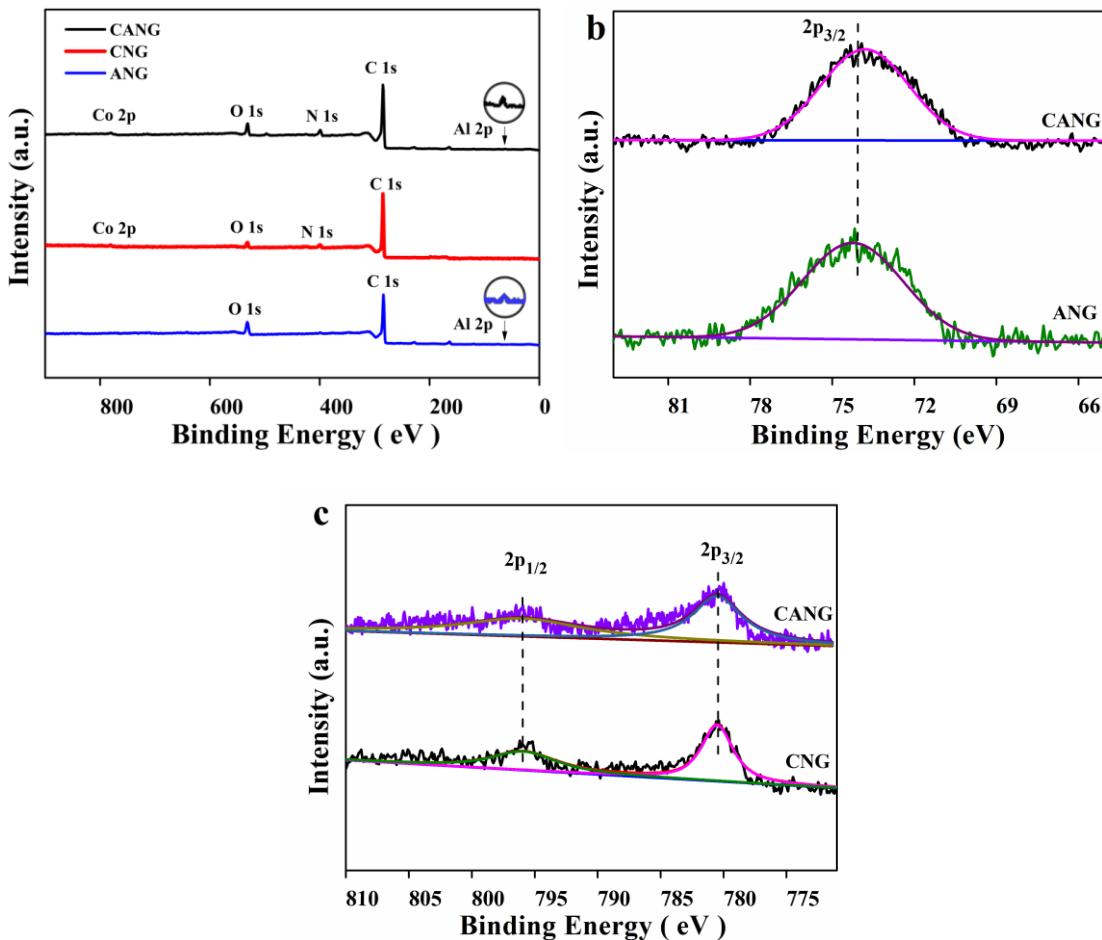
**Fig. S1** EDX of CANG by TEM.



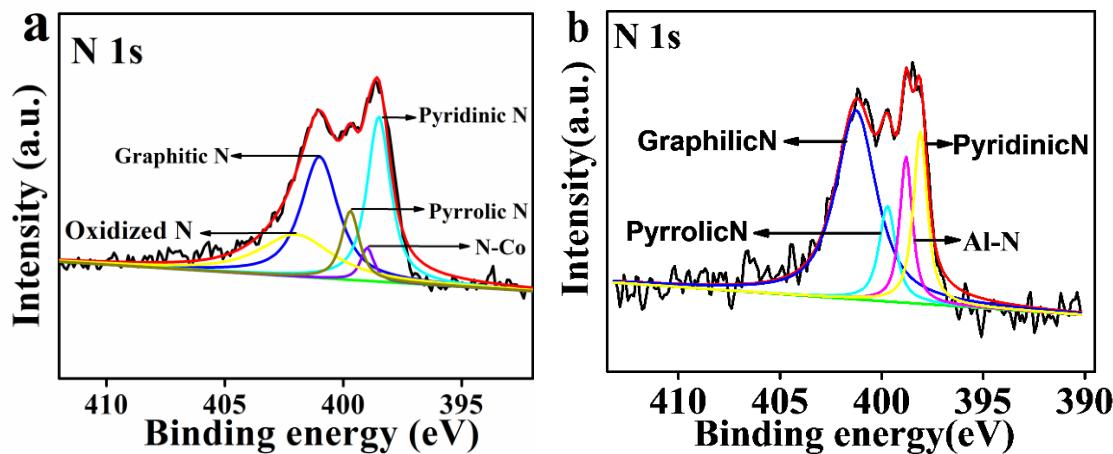
**Fig. S2** The fitted Raman spectra.



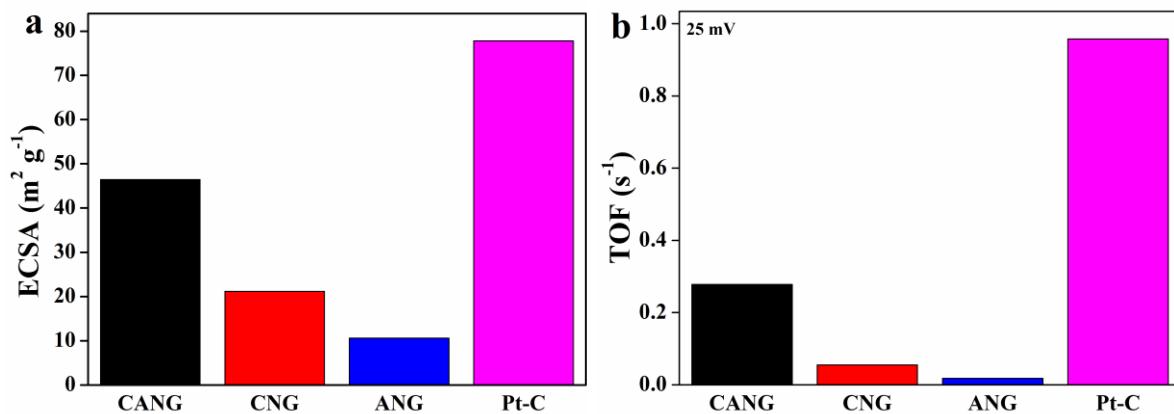
**Fig. S3** The pore-size distribution curves of CANG, CNG, and ANG



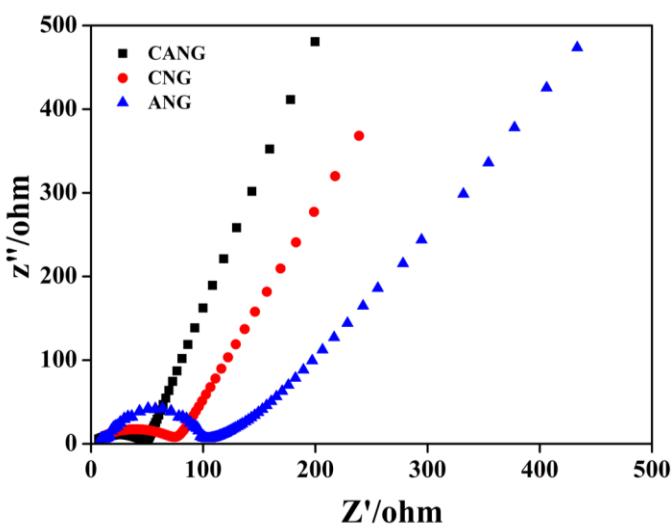
**Fig. S4** The XPS survey (a), Al2p XPS (b), and Co2p (c) XPS of CANG.



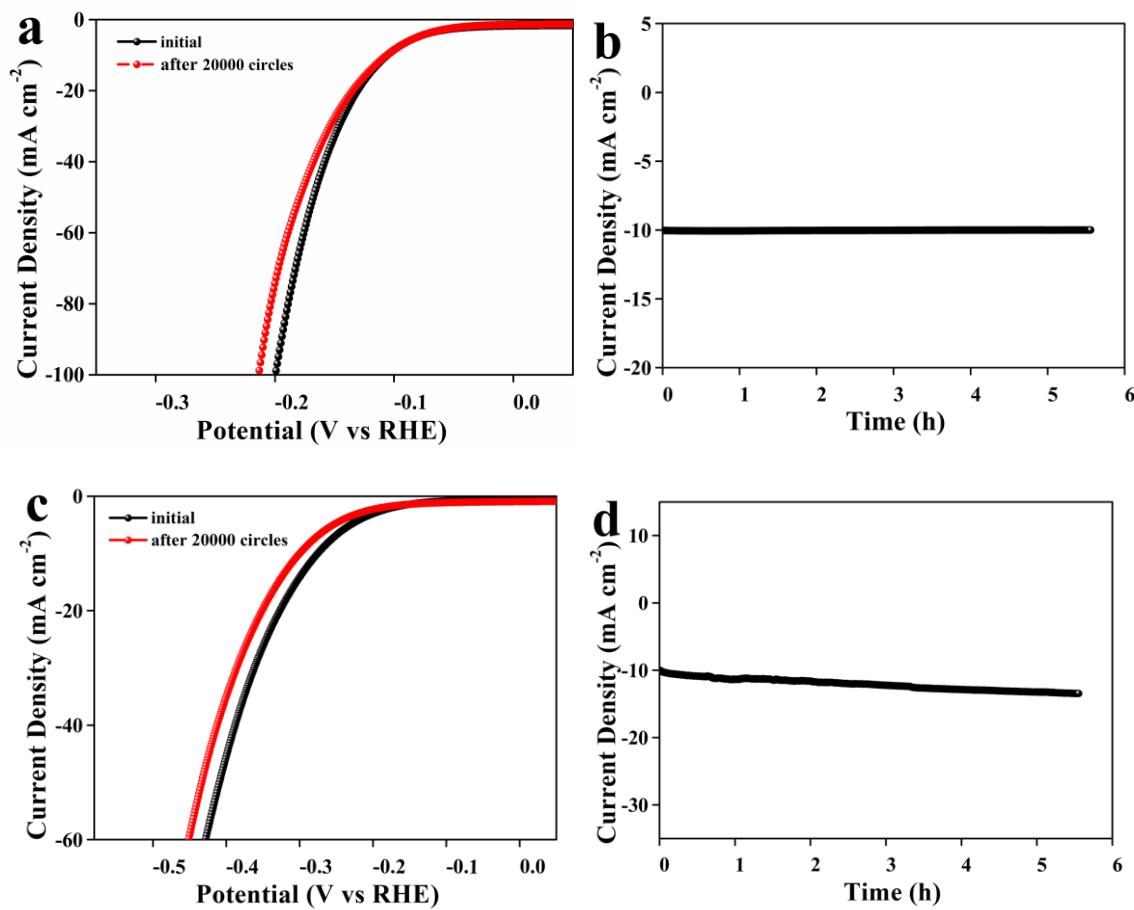
**Fig. S5** The N1s XPS of CNG (a) and ANG (b).



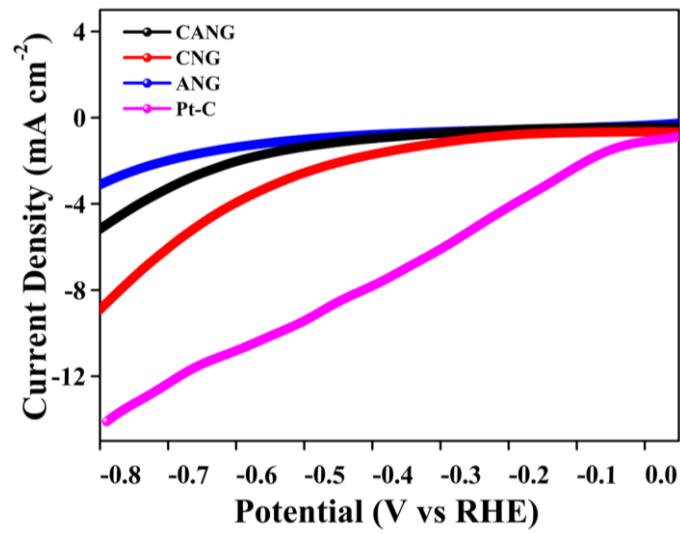
**Fig. S6** The ECSA and TOF (@25mV) of CANG, CNG, ANG and Pt/C in 0.5 M  $\text{H}_2\text{SO}_4$ .



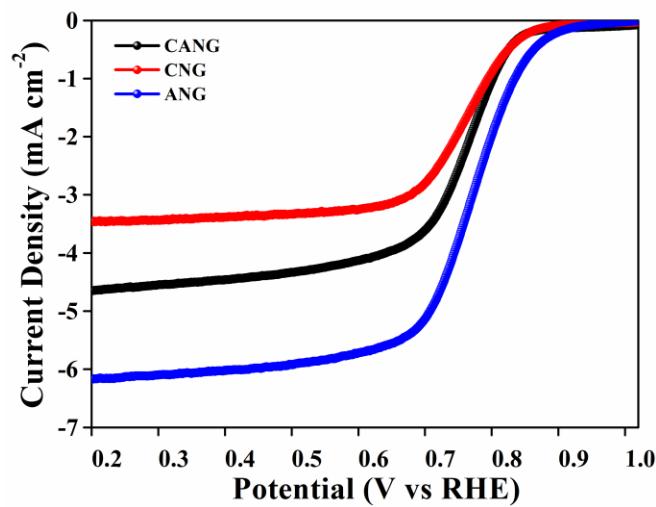
**Fig. S7** The EIS of CANG, CNG, and ANG in 0.5 M  $\text{H}_2\text{SO}_4$ .



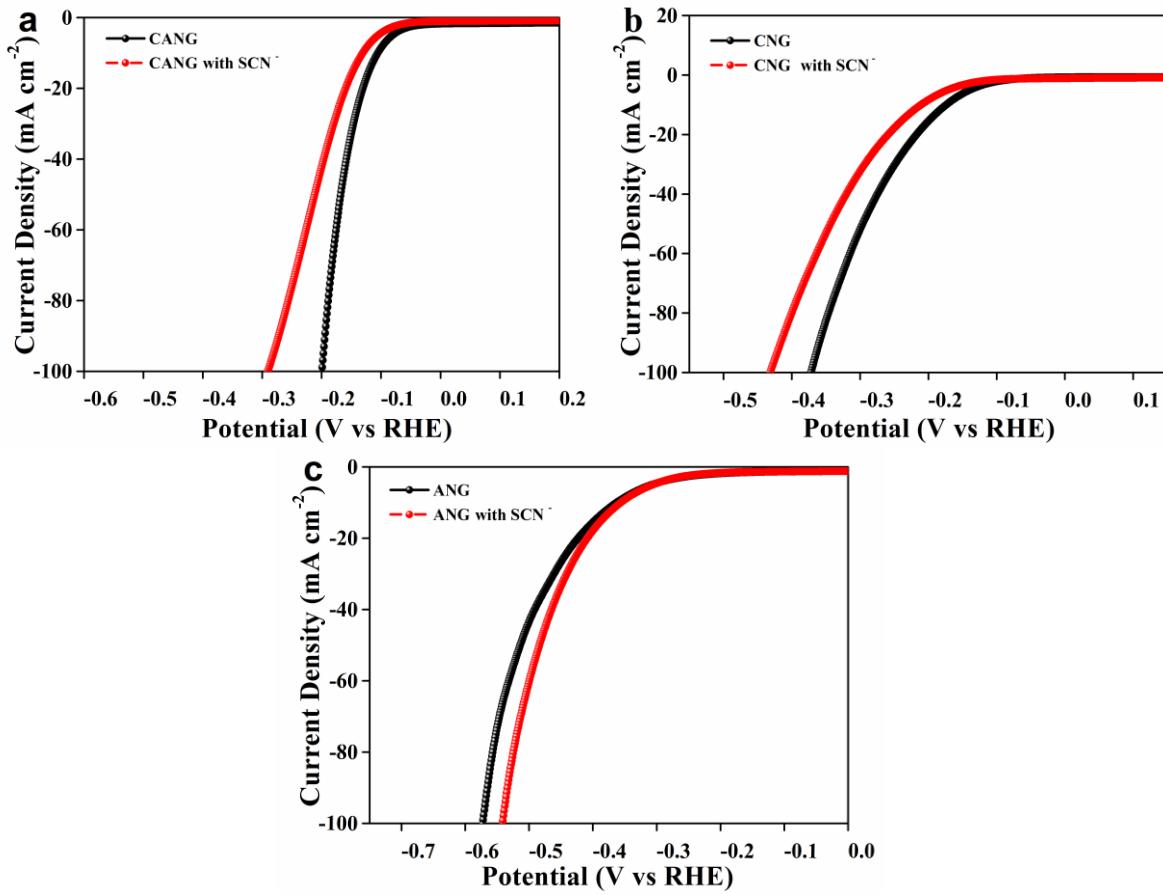
**Fig. S8** The cyclic stability of CANG in 0.5 M  $\text{H}_2\text{SO}_4$  (a, b) and 1 M KOH (c, d). (a, c) LSV curves, (b, d) i-t curves.



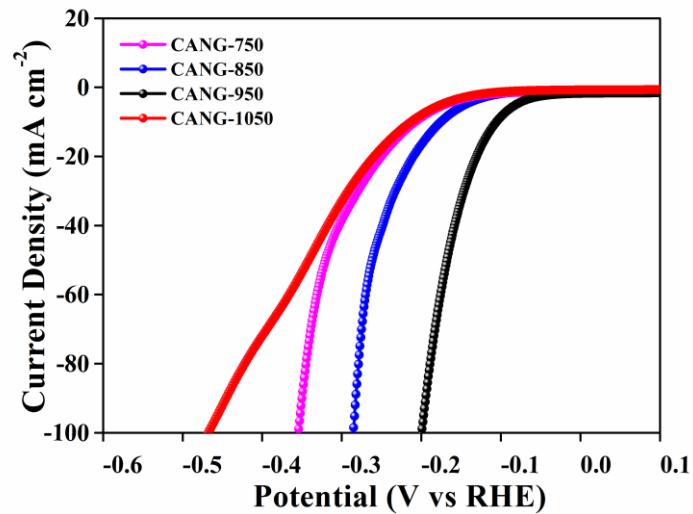
**Fig. S9** The HER activity of CANG, CNG, ANG, and Pt/C in a neutral media (1 M PBS).



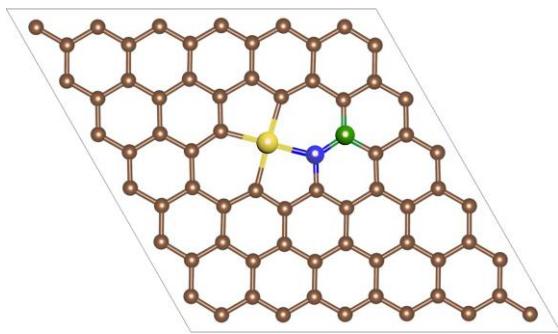
**Fig. S10** The ORR activity of CANG, CNG, and ANG in 0.1 M KOH.



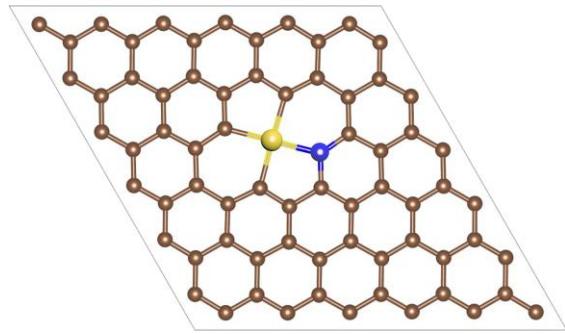
**Fig. S11** The  $\text{SCN}^-$  tests of CANG (a), CNG (b), and ANG (c) in 0.5 M  $\text{H}_2\text{SO}_4$ .



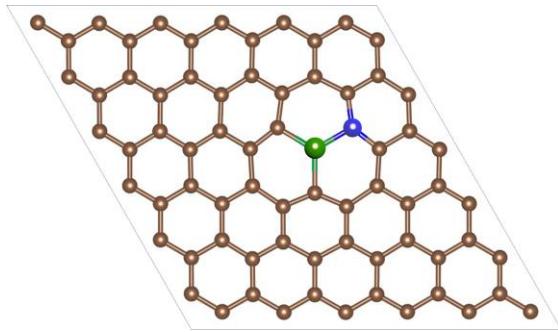
**Fig. S12** The HER activity of CANG fabricating at various temperatures ranged from 750 °C to 1050 °C in 0.5 M  $\text{H}_2\text{SO}_4$ .



(CANG)



(CNG)



(ANG)

**Fig. S13** The structural model of active centre of CANG, CNG, and ANG.

**Tab. S1** The comparison of HER activity of our CANG with those reported Co-based catalyst in literature.

Catalyst	Medium	$\eta_{10}$	Tafel slope	Reference
		(mV)	(mV dec <sup>-1</sup> )	
CANG	0.5 M H <sub>2</sub> SO <sub>4</sub>	106	81.5	This work
	1 M KOH	275	120	
PANICo750A	0.5 M H <sub>2</sub> SO <sub>4</sub>	138	55	[1]
CoN <sub>x</sub> /C	0.5 M H <sub>2</sub> SO <sub>4</sub>	133	57	[2]
Co-NG	0.5 M H <sub>2</sub> SO <sub>4</sub>	147	82	[3]
Co <sub>3</sub> C/Co-N-C/G	0.5 M H <sub>2</sub> SO <sub>4</sub>	103	99	[4]
NC-Co <sub>0.85</sub> Se	0.5 M H <sub>2</sub> SO <sub>4</sub>	127	34.1	[5]
CoP/C	0.5 M H <sub>2</sub> SO <sub>4</sub>	151	128	[6]
CeO <sub>2</sub> /Co (OH) <sub>2</sub>	1 M KOH	317	140	[7]
Co <sub>9</sub> S <sub>8</sub>	1 M KOH	293	127	[8]
Co <sub>9</sub> S <sub>8</sub> HMs-140/C	1 M KOH	250	108	[9]

**Tab. S2** The elemental component of CANG fabricated at various temperatures.

	750 °C	850 °C	950 °C	1050 °C
Co	1.57%	1.84%	2.18%	1.84%
Al	0.21%	0.50%	0.51%	0.82%
N	7.55%	5.51%	4.50%	3.33%

## Reference

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