

# **A symmetric direct ammonia fuel cell using ternary NiCuFe alloy embedded in carbon network as electrodes**

Mengfei Zhang<sup>a</sup>, Jie Zhang<sup>b</sup>, Georgina Jeerh<sup>a</sup>, Peimiao Zou<sup>a</sup>, Boyao Sun<sup>a</sup>, Marc Walker<sup>c</sup>, Kui  
Xie<sup>b</sup>, Shanwen Tao<sup>a, d\*</sup>

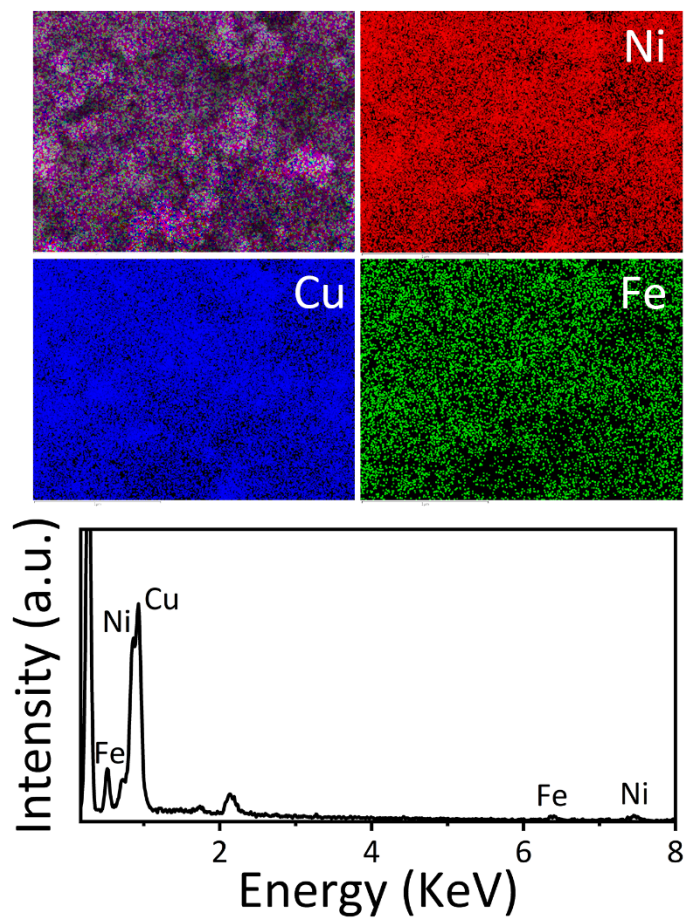
<sup>a</sup> School of Engineering, University of Warwick, Coventry, CV4 7AL, UK

E-mail: [s.tao.1@warwick.ac.uk](mailto:s.tao.1@warwick.ac.uk)

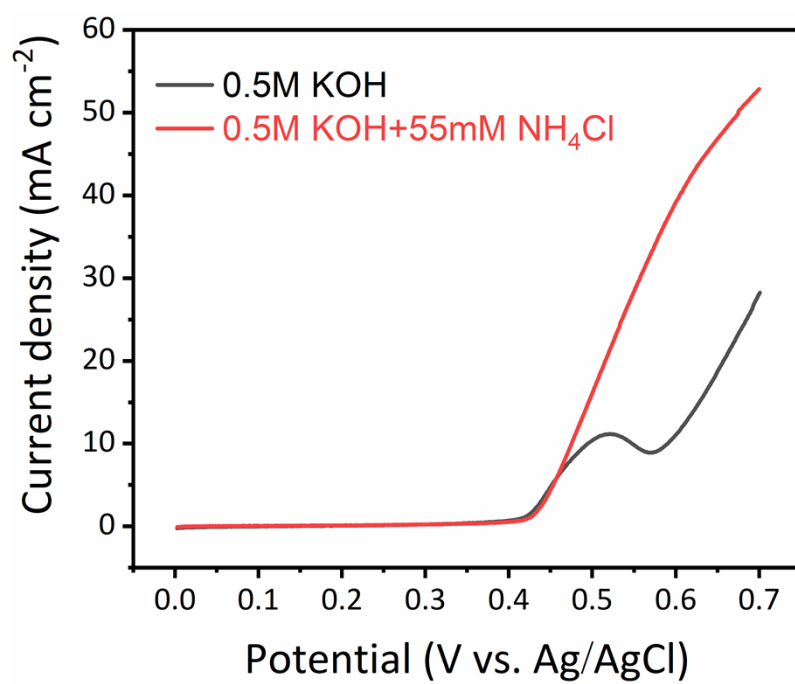
<sup>b</sup> Key Laboratory of Design and Assembly of Functional Nanostructures, Fujian Institute of Research on the Structure of Matter, Chinese Academy of Sciences, Fuzhou, Fujian 350002, China

<sup>c</sup> Department of Physics, University of Warwick, Coventry CV4 7AL, UK

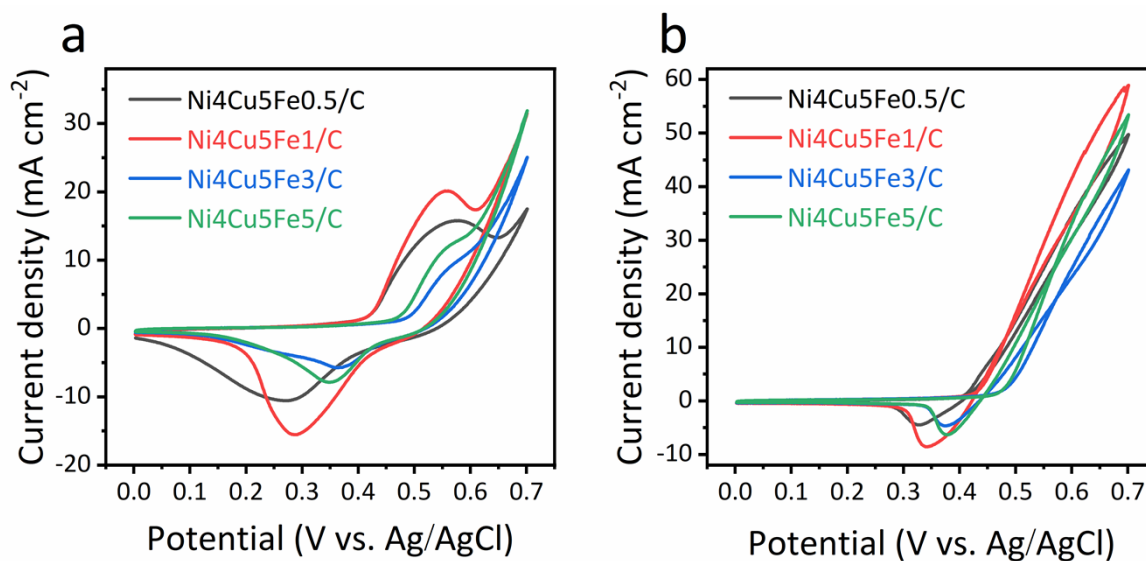
<sup>d</sup> Department of Chemical Engineering, Monash University, Clayton, Victoria 3800, Australia



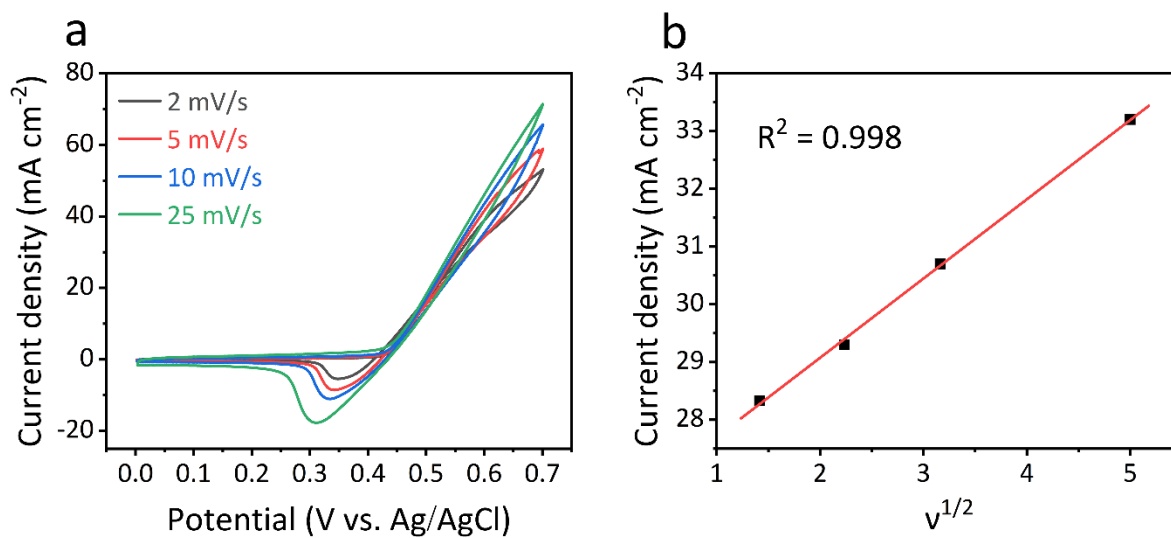
**Figure S1.** EDS mapping images and EDS spectrum of NiCuFe1/C.



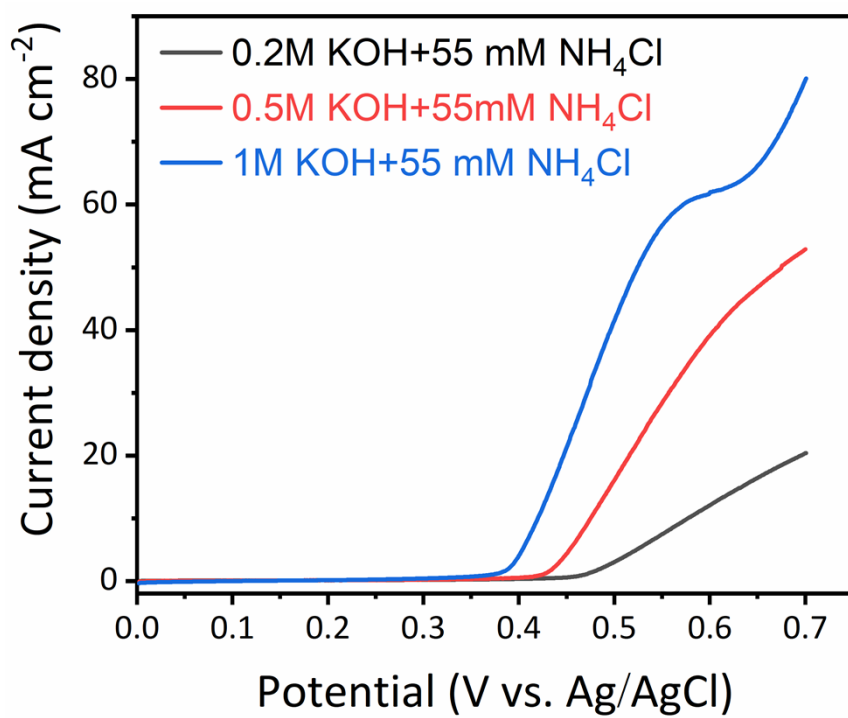
**Figure S2.** LSV curves of NiCuFe1/C in 0.5 M KOH, 0.5 M KOH+55 mM NH<sub>4</sub>Cl at a scan rate of 2 mV s<sup>-1</sup>.



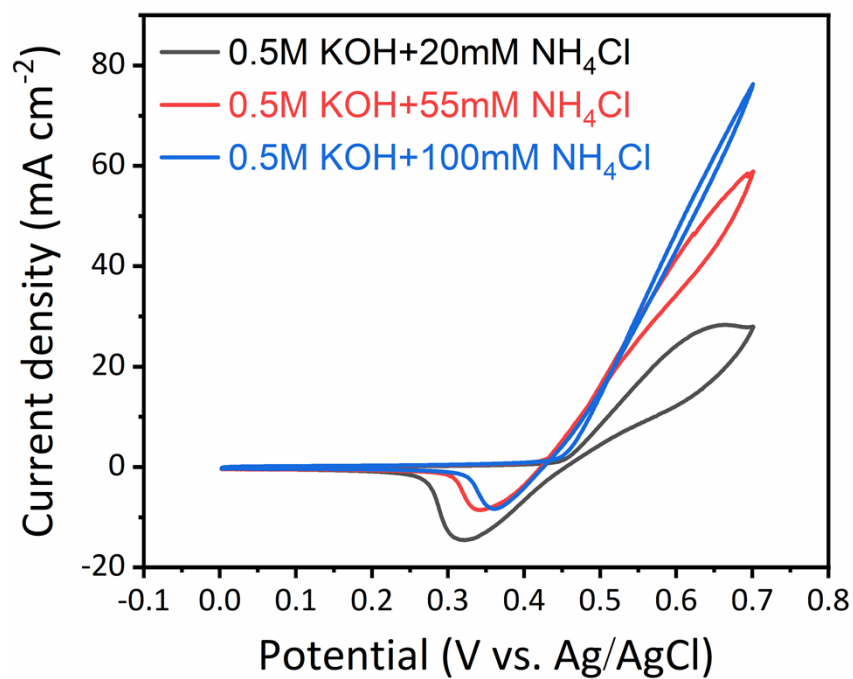
**Figure S3.** CV curves of NiCuFe<sub>0.5</sub>/C, NiCuFe<sub>1</sub>/C, NiCuFe<sub>3</sub>/C, NiCuFe<sub>5</sub>/C in (a) 0.5 M KOH, (b) 0.5 M KOH+55 mM NH<sub>4</sub>Cl at a scan rate of 5 mV s<sup>-1</sup>.



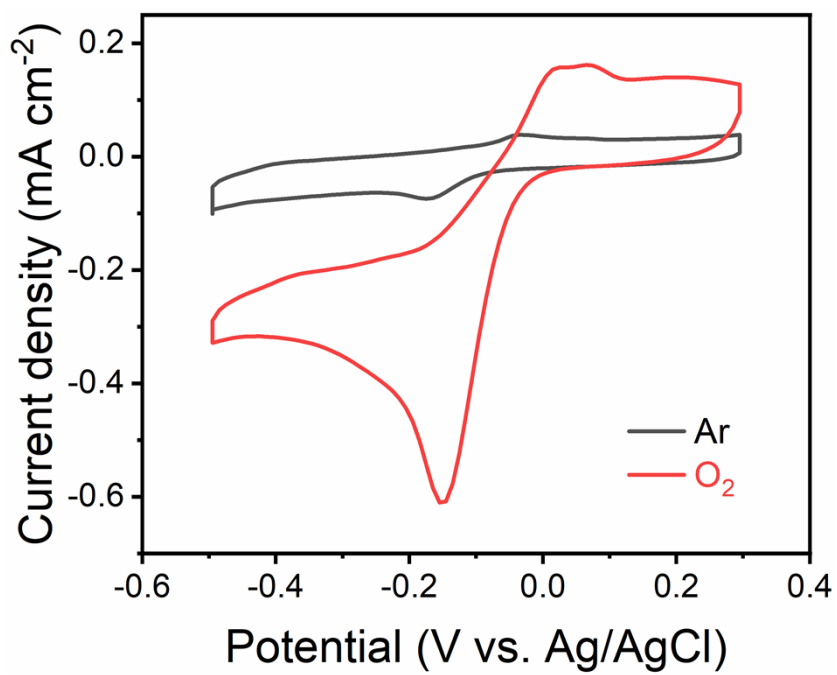
**Figure S4.** (a) CV curves of NiCuFe1/C in 0.5 M KOH+55 mM NH<sub>4</sub>Cl at different scan rates. (b) plots of the square root of the current density vary with scan rate for NiCuFe1/C.



**Figure S5.** LSV curves of NiCuFe1/C in 55 mM NH<sub>4</sub>Cl with different KOH concentration.

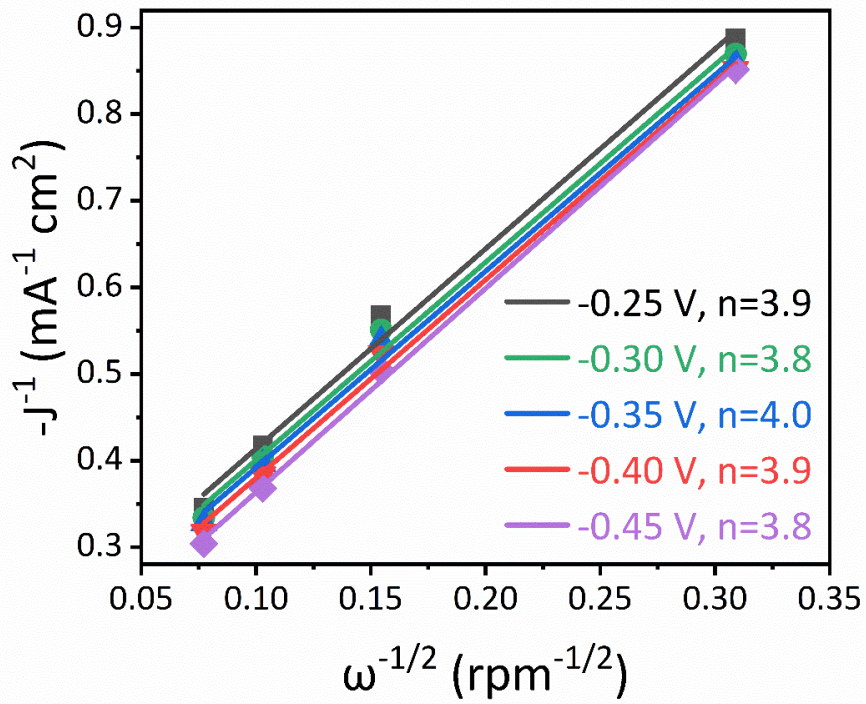


**Figure S6.** CV curves of NiCuFe1/C in 0.5 M KOH with different ammonia concentration at a scan rate of 5 mV s<sup>-1</sup>.

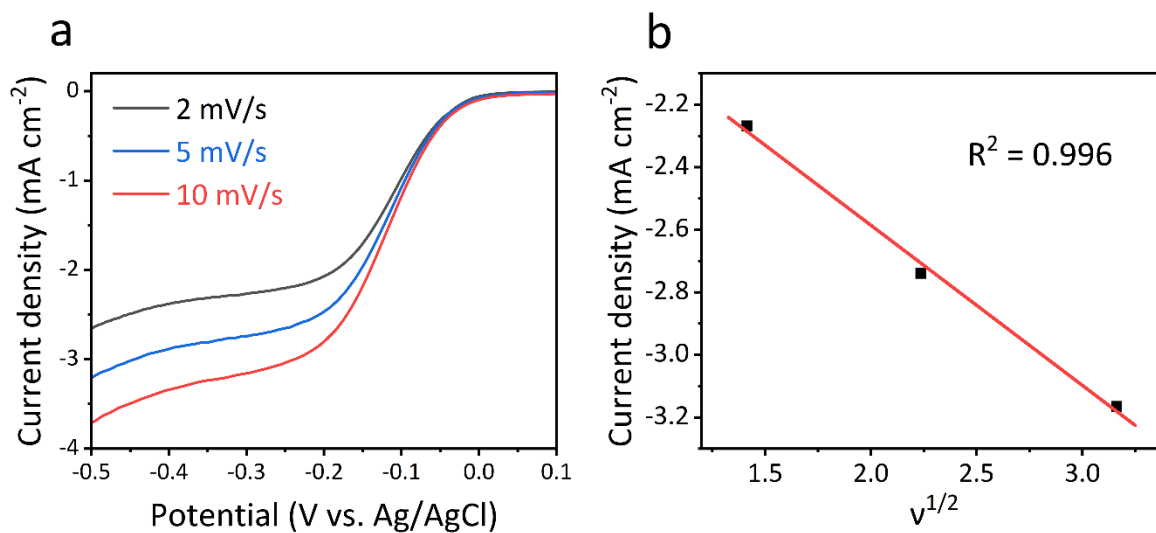


**Figure S7.** CV curves of NiCu/C in Ar or O<sub>2</sub>-saturated 0.1 M KOH.

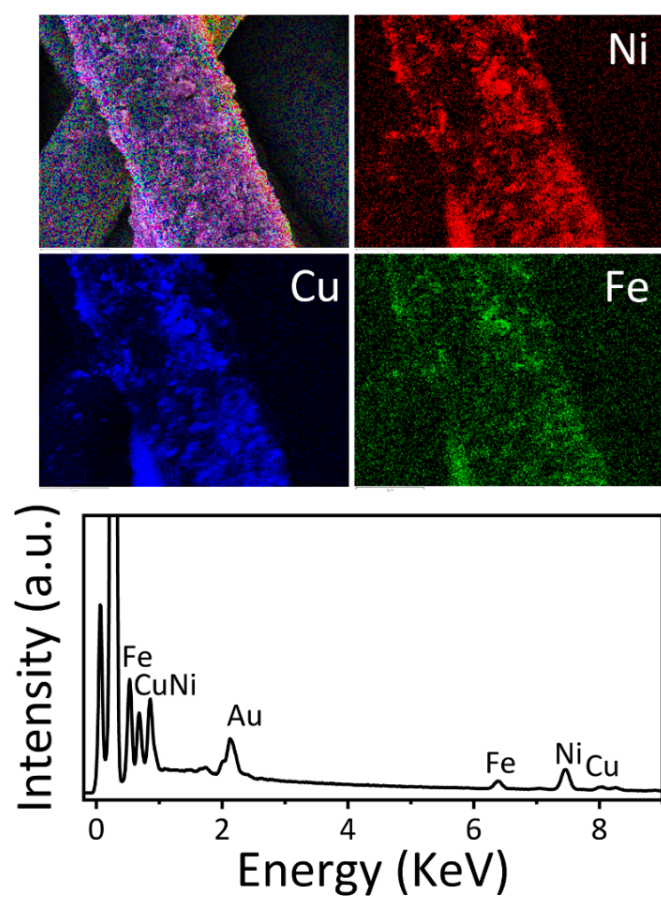




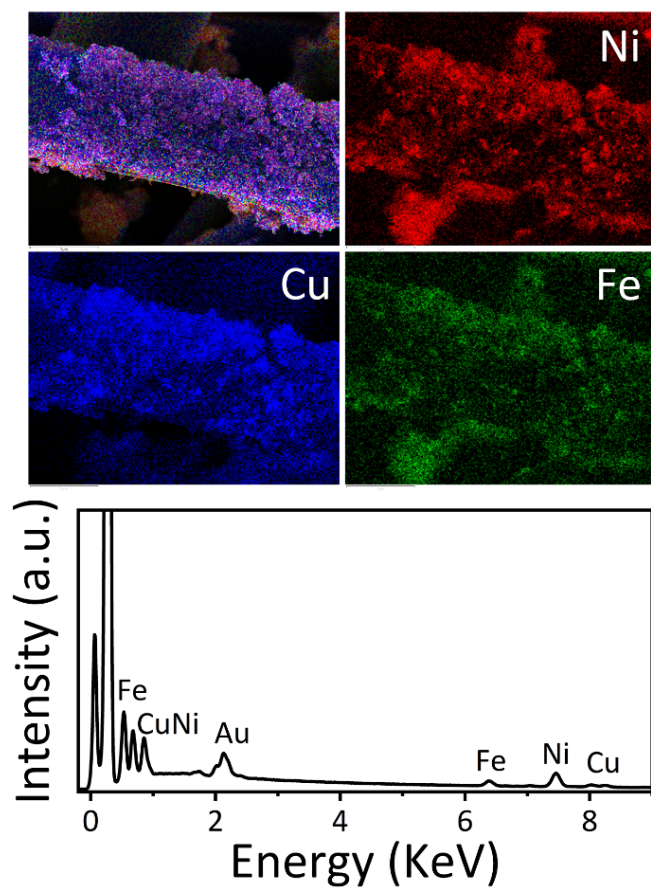
**Figure S8.** The fitted K-L plots of NiCuFe1/C at different potentials.



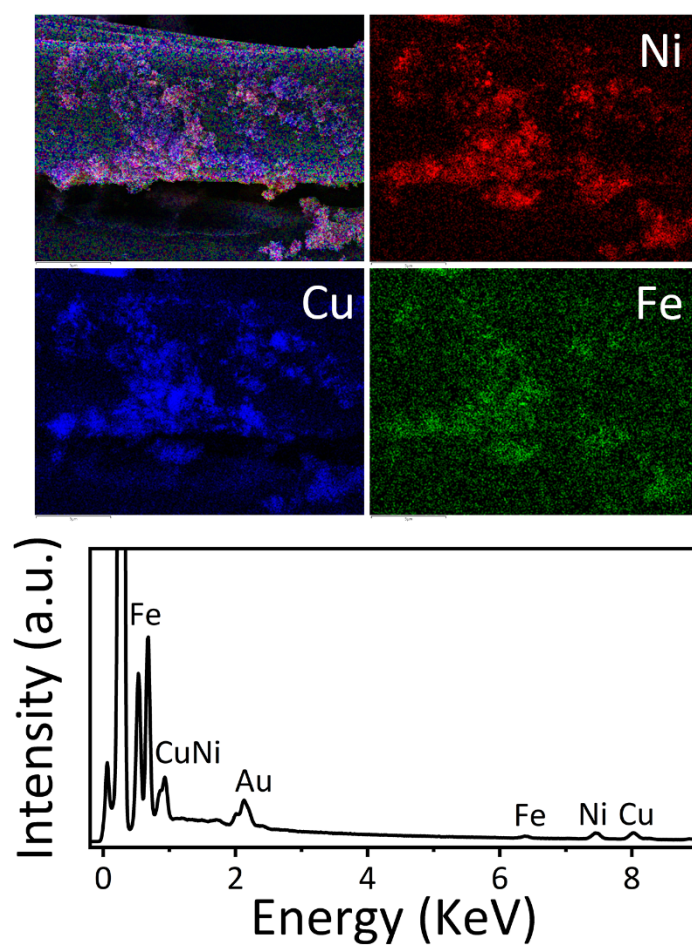
**Figure S9.** (a) LSV curves and (b) current density versus square root of the scan rate of NiCuFe1/C in O<sub>2</sub>-saturated 0.1 M KOH at different scan rates.



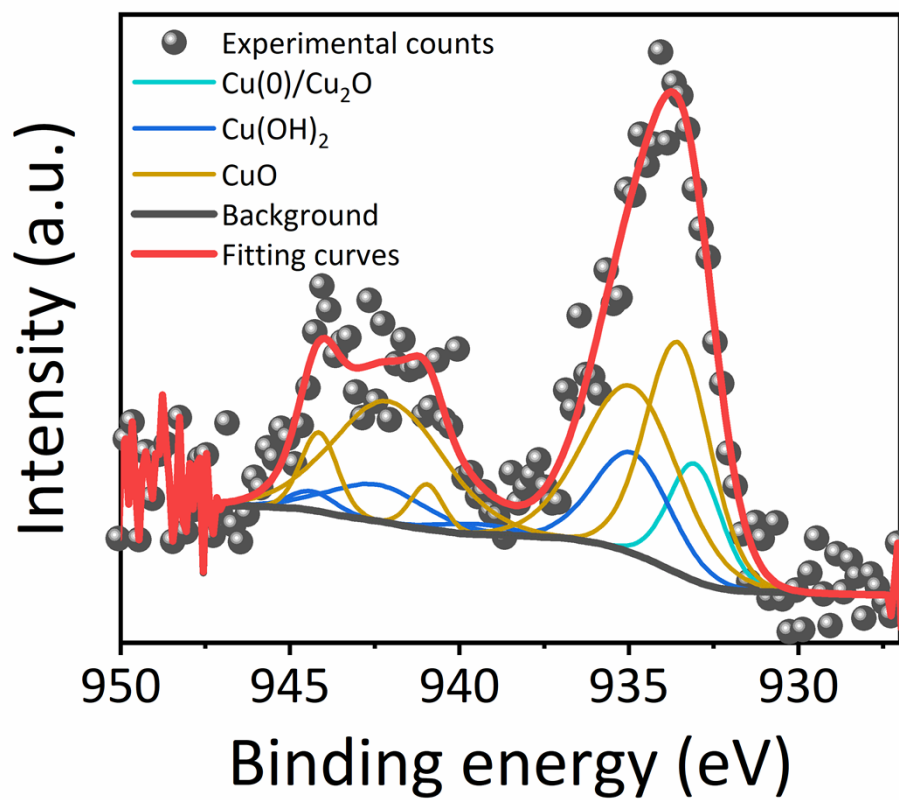
**Figure S10.** EDS Mapping images and EDS spectrum of NiCuFe1/C electrode before test.



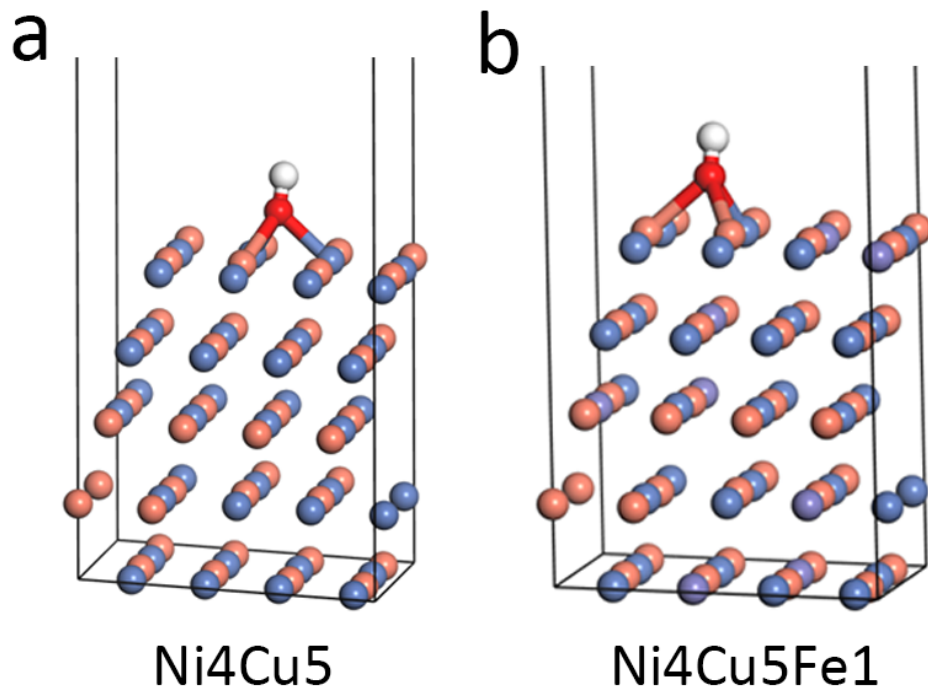
**Figure S11.** EDS Mapping images and EDS spectrum of NiCuFe1/C anode after test.



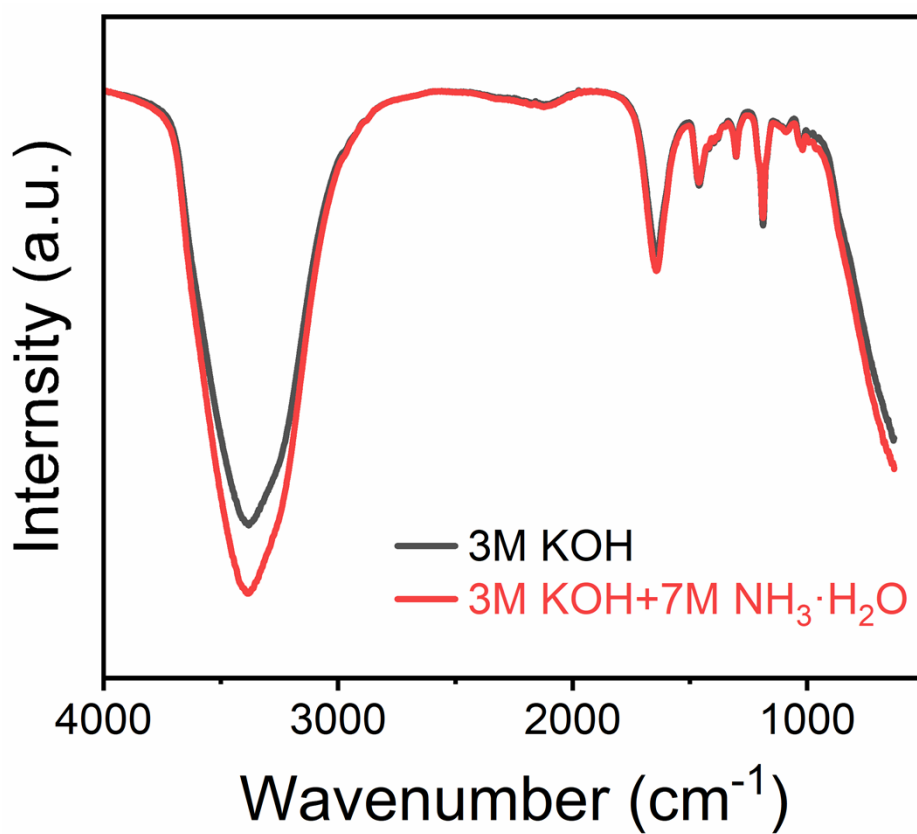
**Figure S12.** EDS Mapping images and EDS spectrum of NiCuFe1/C cathode after test.



**Figure S13.** XPS spectrum of Cu $2p_{3/2}$  in Ni<sub>4</sub>Cu<sub>5</sub>Fe<sub>1</sub>/C anode.



**Figure S14.** Slab model and OH adsorption on the surfaces of NiCu and NiCuFe1.



**Figure S15.** FTIR spectra of AEMs after immersed in 3 M KOH and 3 M KOH + 7 M  $\text{NH}_3 \cdot \text{H}_2\text{O}$  for 24 h.



**Table S1** The composition calculated from XRF results.

Samples	XRF results					The composition calculated from XRF
	Ni	Cu	Fe	P	S	
Ni4Cu5	41.1	57.4	0	0.64	0.42	Ni4Cu5.59
Ni4Cu5Fe0.5	39.8	52.9	2.83	0.64	0.40	Ni4Cu5.32Fe0.28
Ni4Cu5Fe1	37.8	53.2	7.71	0.58	0.37	Ni4Cu5.63Fe0.82
Ni4Cu5Fe3	33.2	45	20.5	0.53	0.34	Ni4Cu5.42Fe2.47

**Table S2** Comparison of DAFC performance with literature results for DAFCs with non-precious metal catalysts in the electrodes.

Anode	Cathode	Fuel	T/°C	OCV/V	Peak current density/ mA cm <sup>-2</sup>	Peak current density / mW cm <sup>-2</sup>	Ref.
NiCu/C	SrCo <sub>0.8</sub> Cu <sub>0.1</sub> Nb <sub>0.1</sub> O <sub>3-<math>\delta</math></sub> /C	1M NaOH + 35% NH <sub>3</sub> ·H <sub>2</sub> O	25	0.45	~2.2	0.25	1
NiCu/C	SrFe <sub>0.8</sub> Cu <sub>0.1</sub> Nb <sub>0.1</sub> O <sub>3-<math>\delta</math></sub> /C	1M NaOH + 5M NH <sub>3</sub> ·H <sub>2</sub> O	25	0.46	~2.5	0.35	2
NiCu/C	$\alpha$ -MnO <sub>2</sub> /C	3M NH <sub>3</sub> ·H <sub>2</sub> O	25	0.39	2.1	0.35	3
Ni/C	MnO <sub>2</sub> /C	35% NH <sub>3</sub> ·H <sub>2</sub> O	25	0.77	~10	3.6	4
CDN/C	MnO <sub>2</sub> /C	35% NH <sub>3</sub> ·H <sub>2</sub> O	80	0.8	32	9	5
Ni <sub>4</sub> Cu <sub>5</sub> Fe <sub>1</sub> /C	Ni <sub>4</sub> Cu <sub>5</sub> Fe <sub>1</sub> /C	3M KOH+7M NH <sub>3</sub> ·H <sub>2</sub> O	80	0.62	31	8.9	<b>This work</b>

## References

1. P. Zou, S. Chen, R. Lan and S. Tao, *ChemSusChem*, 2019, **12**, 2788-2794.
2. P. Zou, S. Chen, R. Lan, J. Humphreys, G. Jeerh and S. Tao, *Int. J. Hydrog. Energy*, 2019, **44**, 26554-26564.
3. M. Zhang, P. Zou, G. Jeerh, S. Chen, J. Shields, H. Wang and S. Tao, *ACS Sustain. Chem. Eng.*, 2020, **8**, 12817-12824.
4. R. Lan and S. Tao, *ECS Electrochem. Lett.*, 2013, **2**, F37-F40.
5. R. Lan and S. Tao, *Electrochem. Solid-State Lett.*, 2010, **13**, B83-B86.