

## Supporting Information

### High-valence chromium accelerated interface electron transfer for water oxidation

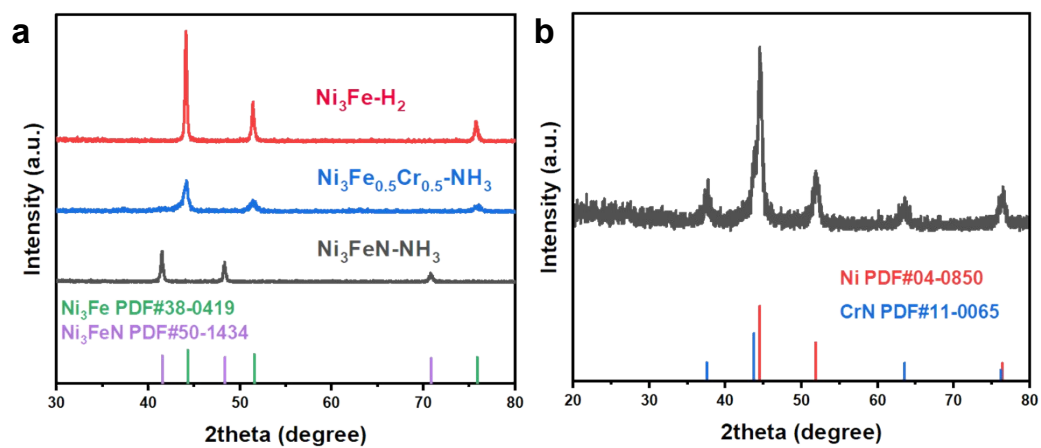
Shaoxi Kong,<sup>a†</sup> Mengfei Lu,<sup>a,b†</sup> Shicheng Yan,<sup>a\*</sup> and Zhigang Zou<sup>a,b</sup>

<sup>a</sup>Collaborative Innovation Center of Advanced Microstructures, Eco-materials and Renewable Energy Research Center (ERERC), College of Engineering and Applied Sciences, Nanjing University, No. 22, Hankou Road, Nanjing, Jiangsu 210093, P. R. China.

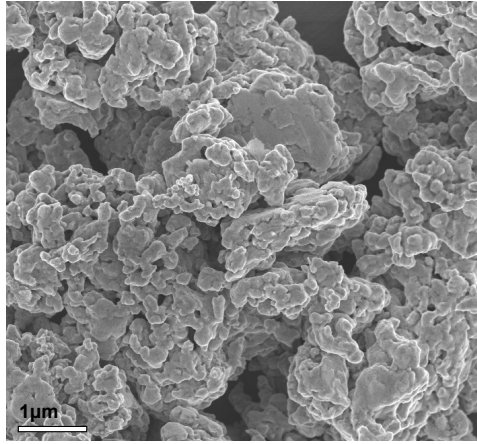
<sup>b</sup>National Laboratory of Solid State Microstructures, Jiangsu Key Laboratory for Nano Technology, School of Physics, Nanjing University, No. 22 Hankou Road, Nanjing, Jiangsu 210093, P. R. China.

\* Corresponding Author E-mail: yscfei@nju.edu.cn (S. C. Yan)

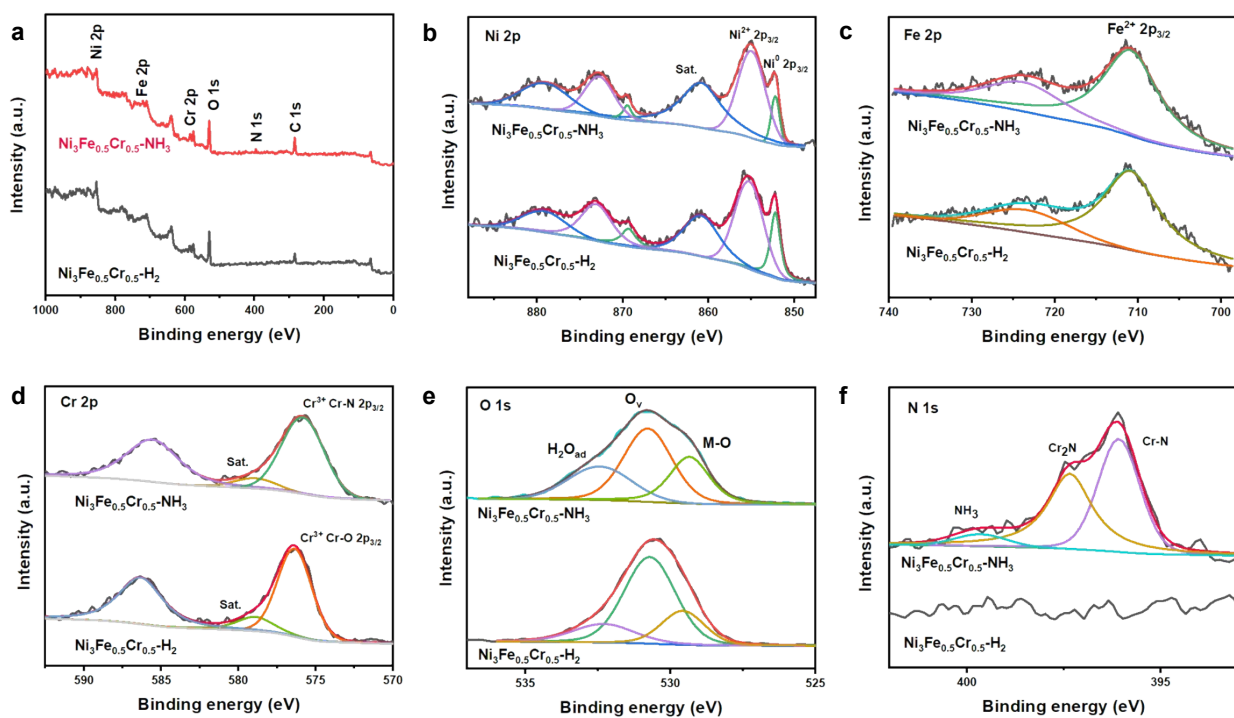
† These authors contributed equally to this work.



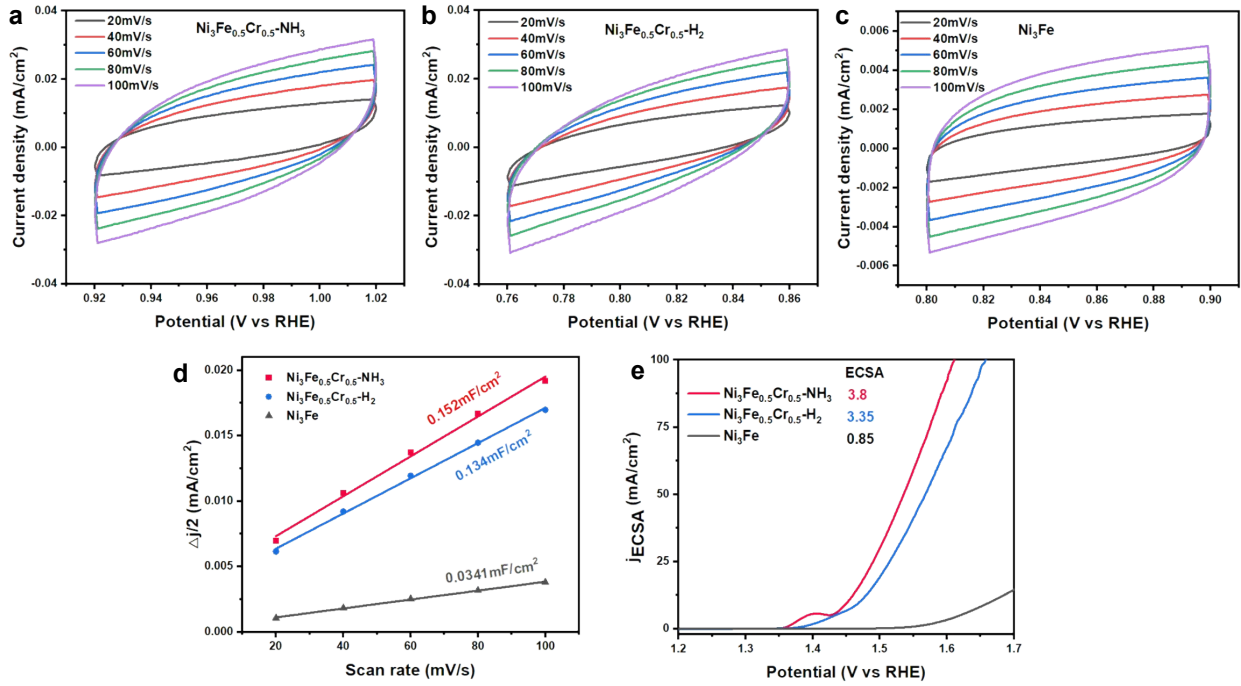
**Fig S1.** (a) XRD patterns of  $\text{Ni}_3\text{Fe}_{0.5}\text{Cr}_{0.5}\text{-NH}_3$ ,  $\text{Ni}_3\text{Fe-H}_2$  and  $\text{Ni}_3\text{FeN-NH}_3$ . (b) XRD pattern of  $\text{Ni}_3\text{Cr-NH}_3$ .



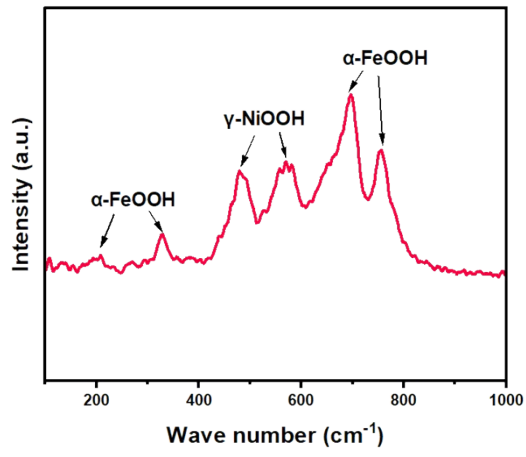
**Fig S2.** SEM image of Ni<sub>3</sub>Fe.



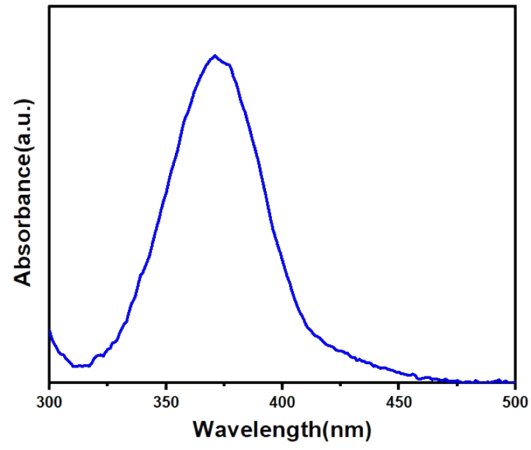
**Fig S3.** XPS spectra. (a) Survey spectra, (b) Ni 2p, (c) Fe 2p, (d) Cr 2p, (e) O 1s and (f) N 1s for  $\text{Ni}_3\text{Fe}_{0.5}\text{Cr}_{0.5}\text{-NH}_3$  and  $\text{Ni}_3\text{Fe}_{0.5}\text{Cr}_{0.5}\text{-H}_2$ .



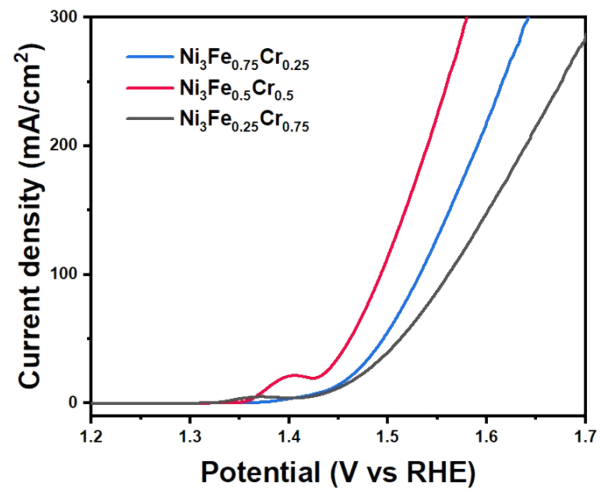
**Fig S4.** CV curves with different scan rates in non-faradic region. (a)  $\text{Ni}_3\text{Fe}_{0.5}\text{Cr}_{0.5}\text{-NH}_3$ , (b)  $\text{Ni}_3\text{Fe}_{0.5}\text{Cr}_{0.5}\text{-H}_2$ , (c)  $\text{Ni}_3\text{Fe}$  at various scan rates (20, 40, 60, 80 and 100 mV/s) in 1.0 M KOH for OER. (d) Plots of capacitive current density versus scan rate. The slopes represent  $C_{dl}$ . (e) LSV curves normalized by ECSA of the as-prepared samples. The ECSA was calculated by double-layer capacitance.



**Fig S5.** Raman spectrum of  $\text{Ni}_3\text{Fe}_{0.5}\text{Cr}_{0.5}\text{-NH}_3$  after OER.

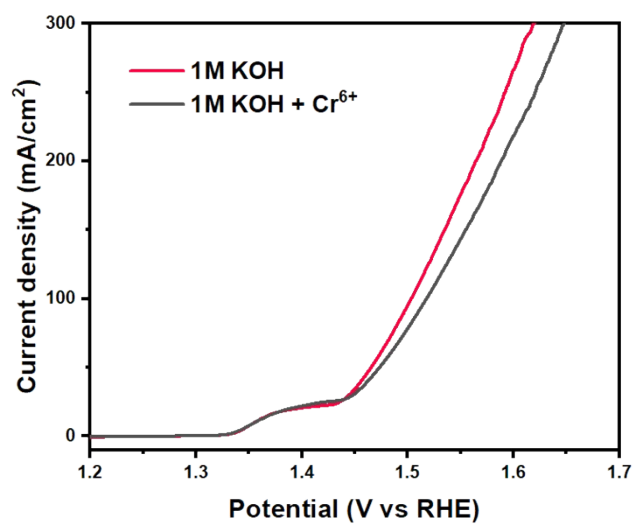


**Fig S6.** UV-vis spectroscopy of the electrolyte after the chronoamperometry measurement.



**Fig S7.**OER polarization curves of Ni<sub>3</sub>Fe<sub>1-x</sub>Cr<sub>x</sub>-NH<sub>3</sub> (x=0.25, 0.5, 0.75) with 90 % iR correction at a scan rate of 10 mV/s in 1.0 M KOH.





**Fig S8.** OER polarization curves of  $\text{Ni}_3\text{Fe}_{0.5}\text{Cr}_{0.5}\text{-NH}_3$  with 90 % iR correction at a scan rate of 10 mV/s in 1.0 M KOH and 1.0 M KOH with  $\text{Cr}^{6+}$ .

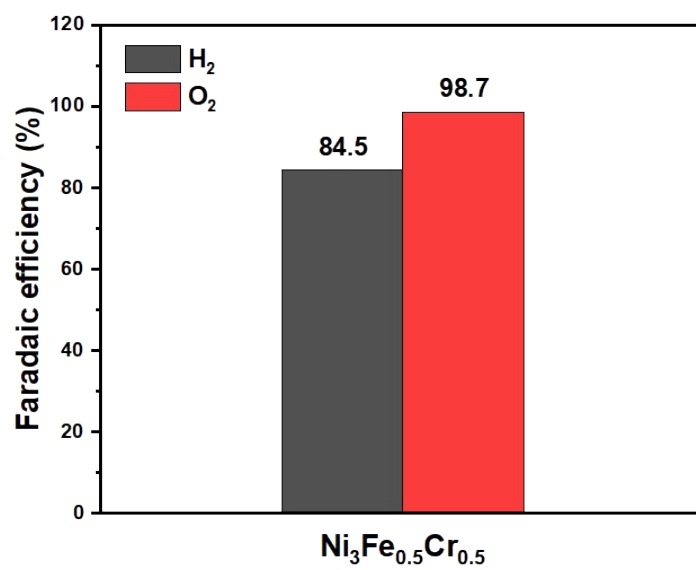


Fig S9. The faradaic efficiency of Ni<sub>3</sub>Fe<sub>0.5</sub>Cr<sub>0.5</sub>-NH<sub>3</sub>.

**Table S1.** Compositions of the samples determined by ICP-AES.

Samples	The content of metal ions (mmol/L)			Mole ratio		
	Ni	Fe	Cr	Ni	Fe	Cr
$\text{Ni}_3\text{Fe}_{0.5}\text{Cr}_{0.5}$	1.20	0.21	0.18	3.08	0.54	0.46
After 10h OER	0.2835	0.0500	0.0047	3.06	0.54	0.051
After 20h OER	0.1894	0.0340	0.0033	2.99	0.54	0.05

**Table S2.** Concentrations of Cr Fe and Ni for Ni<sub>3</sub>Fe<sub>0.5</sub>Cr<sub>0.5</sub>-NH<sub>3</sub> after operating in different electrolytes, as shown in Fig 5b, determined by the atomic absorption spectrum.

	<b>0-10h OER</b>	<b>10-20h OER</b>
<b>Concentration of Cr(<math>\mu\text{g/L}</math>)</b>	237.66	ND
<b>Concentration of Fe(<math>\mu\text{g/L}</math>)</b>	ND	ND
<b>Concentration of Ni(<math>\mu\text{g/L}</math>)</b>	ND	ND

**Table S3.** Comparison of the OER performance of Ni<sub>3</sub>Fe<sub>0.5</sub>Cr<sub>0.5</sub>-NH<sub>3</sub> to the recently reported Cr-NiFe catalysts.

Catalyst	Electrolyte	Substrates	Method	j (mA cm <sup>-2</sup> )	η (mV)	Reference
Ni <sub>3</sub> Fe <sub>0.5</sub> Cr <sub>0.5</sub> -NH <sub>3</sub>	1.0 M KOH	Carbon paper	Dropping	25	209	This work
				50	232	
NiFeCr-LDH	1.0 M KOH	Ni foam	Dropping	100	242	[1]
NiFeCr-LDH/MoS <sub>2</sub>	1.0 M KOH	glassy carbon	Dropping	10	270	[2]
NiFeCr LDH	1.0 M KOH	Carbon paper	In situ growth	25	225	[3]
h-NiFeCr	1.0 M KOH	Ni foam	Electrodeposition	10	220	[4]
NiFeCr	1.0 M KOH	Ni foam	Electrodeposition	100	260	[5]
Cr <sub>1</sub> /FeNi-LDH	1.0 M KOH	stainless steel	Electrodeposition	10	202	[6]
CS-NiFeCr	1.0 M KOH	Copper foil	Electrodeposition	10	200	[7]
				50	230	

## References

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