

## Role of Ce in enhanced performance of water oxidation reaction and urea oxidation reaction for NiFe Layered Double Hydroxide

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Turnover frequency (TOF) value of catalysts was calculated based on the equation:  $TOF = (J \times A) / (4 \times F \times n)$ , where  $J$  (mA/cm<sup>2</sup>) is the current density;  $A$  is the surface area of electrode (1 cm<sup>2</sup>);  $F$  is the Faraday constant (96485 C/mol);  $n$  is molar number of active sites on the electrode. Both Ni, Fe and Ce in NiFe LDH, Ce-NiFe LDH and Ce(OH)<sub>3</sub>@NiFe LDH are regarded as active sites. The mass activity (MA) is calculated according to the equation:  $MA = J/m$ , where  $J$  (mA/cm<sup>2</sup>) is the current density at overpotential of 300 mV;  $m$  is the mass of active sites (Ni, Fe and Ce) deposited onto nickel foam.

**Table S1.** XPS fitting parameters for the binding energies of the resulting material.

Samples	Binding energy [eV]									
	Ni <sup>2+</sup>		Ni <sup>3+</sup>		Fe <sup>3+</sup>		Ce <sup>3+</sup>		Ce <sup>4+</sup>	
	2p <sub>3/2</sub>	2p <sub>1/2</sub>	2p <sub>3/2</sub>	2p <sub>1/2</sub>	2p <sub>3/2</sub>	2p <sub>1/2</sub>	3d <sub>5/2</sub>	3d <sub>3/2</sub>	3d <sub>5/2</sub>	3d <sub>3/2</sub>
Ce(OH) <sub>3</sub> @NiFe LDH	856.45	873.0	/	/	713.3	724.1	873.6	899.1	875.5	902.5
							879.1	905.0	881.3	907.8
									884.1	917.2
Ce-NiFe LDH	856.4	872.9	/	/	713.6	724.3	873.7	899.2	875.6	902.6
							879.2	905.1	881.4	907.9
									884.2	917.3
NiFe-LDH	856.3	872.8	/	/	714.0	724.5	/	/	/	/

**Table S2.** XPS fitting parameters for the Ce peak area of the resulting material.

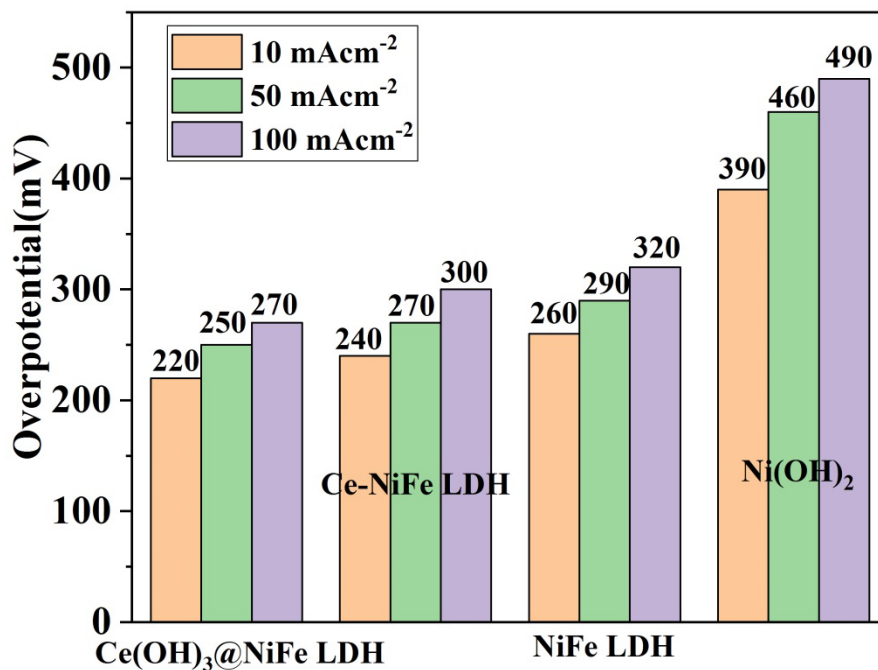
Samples (Ce <sup>3+</sup> /Ce <sup>4+</sup> ratio)	Peak area			
	Ce <sup>3+</sup>		Ce <sup>4+</sup>	
	3d <sub>5/2</sub>	3d <sub>3/2</sub>	3d <sub>5/2</sub>	3d <sub>3/2</sub>
Ce(OH) <sub>3</sub> @NiFe LDH	5648	4958	14539	5654
	6622	3790	3843	3248
Ce-NiFe LDH			2479	4256
	8137	2179	13954	15886
	21892	10237	5321	5347
		2742	2395	

**Table S3.** Comparison of OER performances for Ce(OH)<sub>3</sub>@NiFe LDH/NF with other reported electrocatalysts.

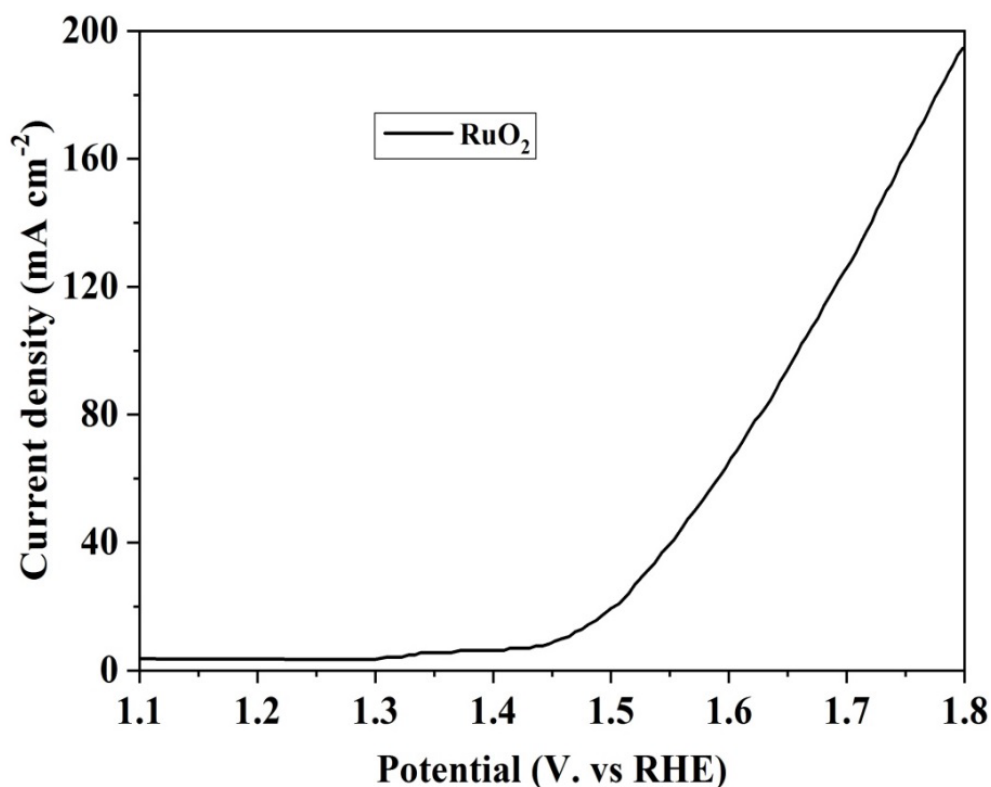
Electrocatalysts	Overpotential	References
<b>Ce(OH)<sub>3</sub>@NiFe LDH</b>	<b>220 mV at 10mA cm<sup>-2</sup></b>	<b>This work</b>
ZnCo LDH	325 mV at 10mA cm <sup>-2</sup>	J. Mater. Chem. A, 2020,8, 8692-8699
NiCo <sub>2</sub> S <sub>4</sub> @NiFe LDH	201 mV at 60 mA cm <sup>-2</sup>	ACS Appl. Mater. Interfaces 2017, 9, 15364–15372
CoO <sub>x</sub> /FeO <sub>x</sub> /CNT	308 mV at 10 mA cm <sup>-2</sup>	J. Mater. Chem. A, 2020, 8, 15140–15147
NiS <sub>2</sub> /NiSe <sub>2</sub> nanocage	290 mV at 20 mA cm <sup>-2</sup>	Small 2020, 16, 1905083
Co-Fe-V metal oxides	249 mV at 10 mA cm <sup>-2</sup>	J. Mater. Chem. A, 2020, 8, 15951–15961
P-doped NiCo <sub>2</sub> O <sub>4</sub>	300 mV at 10 mA cm <sup>-2</sup>	ACS Appl. Mater. Interfaces 2020, 12, 2763–2772
NiS/Fe <sub>3</sub> O <sub>4</sub> HNPs@CNT	243 mV at 10 mA cm <sup>-2</sup>	ACS Appl. Mater. Interfaces 2020, 12, 31552–31563
Fe <sub>3</sub> O <sub>4</sub> /FeS <sub>2</sub>	253 mV at 10 mA cm <sup>-2</sup>	J. Mater. Chem. A, 2020, 8, 14145–14151
CoMoOS nanoboxes	281 mV at 10 mA cm <sup>-2</sup>	Appl. Catal. B: Environ. 2020, 265, 118605
Ni <sub>59</sub> Cu <sub>19</sub> P <sub>9</sub>	307 mV at 10 mA cm <sup>-2</sup>	Appl. Catal. B: Environ. 2018, 237, 409–415
Fe-Ni-P-B-O	236 mV at 10 mA cm <sup>-2</sup>	ACS Nano 2019, 13, 12969–12979
hollow Fe-Co <sub>x</sub> P	300 mV at 10 mA cm <sup>-2</sup>	Chem. Eng. J. 2021, 409, 128227

**Table S4.** Comparison of UOR performances for Ce(OH)<sub>3</sub>@NiFe LDH/NF with other reported electrocatalysts.

Catalyst	Electrolyte	potential	Ref
<b>Ce(OH)<sub>3</sub>@NiFe LDH</b>	<b>1 M KOH+0.5 M urea</b>	<b>1.40@10</b>	<b>This work</b>
Fe <sub>2</sub> P@Ni <sub>x</sub> P/NF	1 M KOH+0.5 M urea	1.34@100	Chem. Eng. J., 2021,417, 128067
Fe <sub>11.1%</sub> -Ni <sub>3</sub> S <sub>2</sub> /NF	1 M KOH+0.33 M urea	1.36@100	J. Mater. Chem. A, 2018, 6, 4346-4353
Mo-Ni <sub>3</sub> S <sub>2</sub>	1 M KOH+0.3 M urea	1.37@100	J. Mater. Chem. A, 2021, 9, 3418-3426
V-Ni <sub>3</sub> N	1 M KOH+0.5 M urea	1.39@100	J. Mater.Chem. A, 2021, 9, 4159-4166
Ni/FeOOH	1 M KOH+0.5 M urea	1.4@100	Chem. Commun., 2020, 56 ,14713-14716
Ni-S-Se/NF	1 M KOH+0.5 M urea	1.41@100	Nano Energy, 2021,81, 105605
CoS <sub>x</sub> /Co-MOF	1 M KOH+0.5 M urea	1.42@100	Inorg. Chem. Front., 2020,7, 2602-2610
MNPBA-P	1 M KOH+0.5 M urea	1.43@100	ACS Sus. Chem. Eng., 2020,8, 16037-16045
Co-Z/Se-2	1 M KOH+0.5 M urea	1.44@100	J. Power Sources 2021,491, 229592



**Fig. S1** The overpotential of Ni(OH)<sub>2</sub>/NF, NiFe LDH/NF, Ce-NiFe LDH/NF and Ce(OH)<sub>3</sub>@NiFe LDH/NF at 100 mA cm<sup>-2</sup> for OER.



**Fig. S2** Polarization curve of the RuO<sub>2</sub> for OER with a scan rate of 5 mV s<sup>-1</sup> in 1 M KOH.

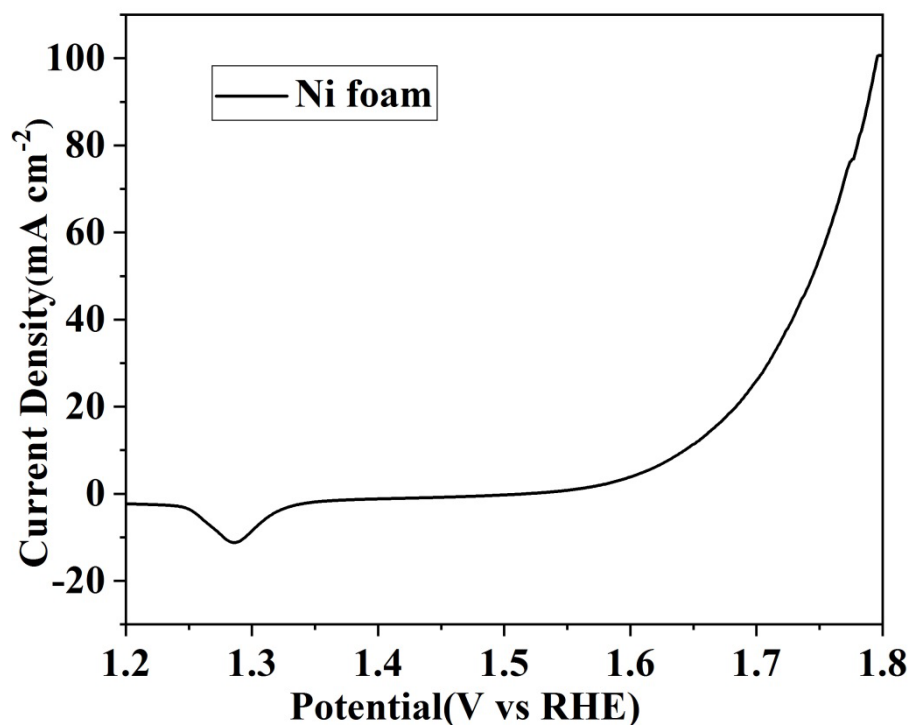


Fig. S3 Polarization curve of the Ni foam for OER with a scan rate of  $5 \text{ mV s}^{-1}$  in 1 M KOH.

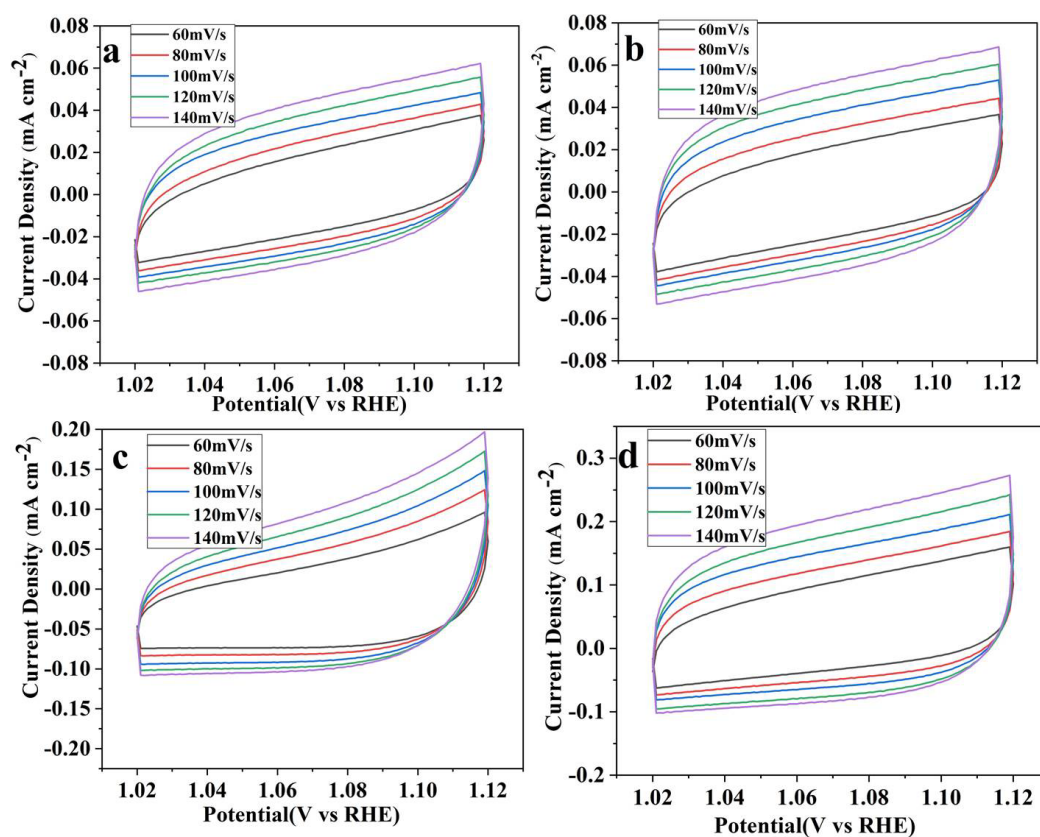


Fig. S4 CV curves with different scan rates for OER, Ni(OH)<sub>2</sub>/NF (a), NiFe LDH/NF (b), Ce-NiFe LDH/NF (c) and Ce(OH)<sub>3</sub>@NiFe LDH/NF (d).

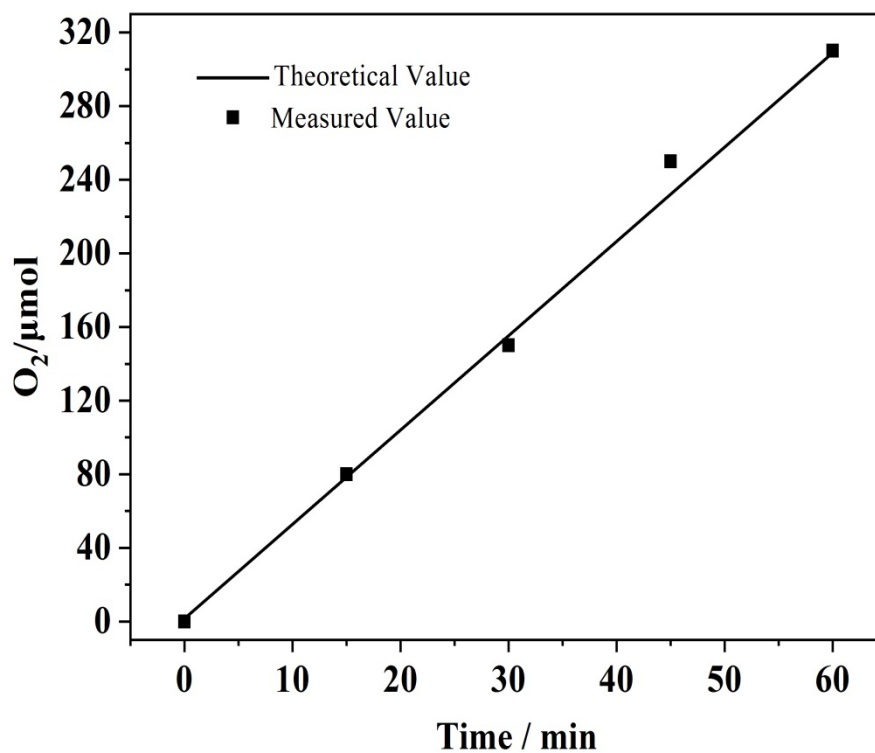


Fig. S5 Electrocatalytic efficiency of O<sub>2</sub> production over Ce(OH)<sub>3</sub>@NiFe LDH/NF.

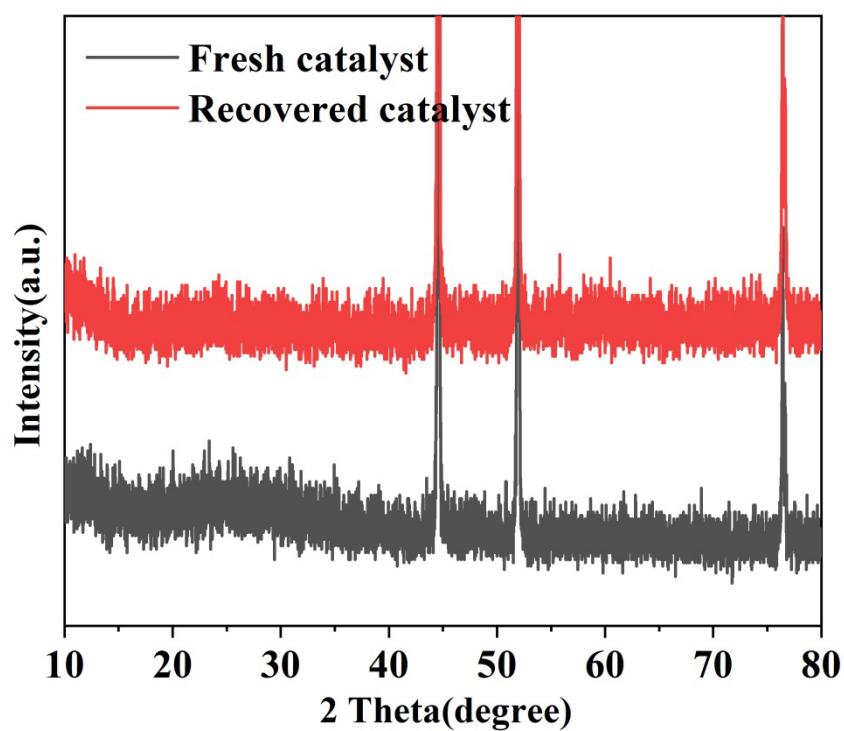
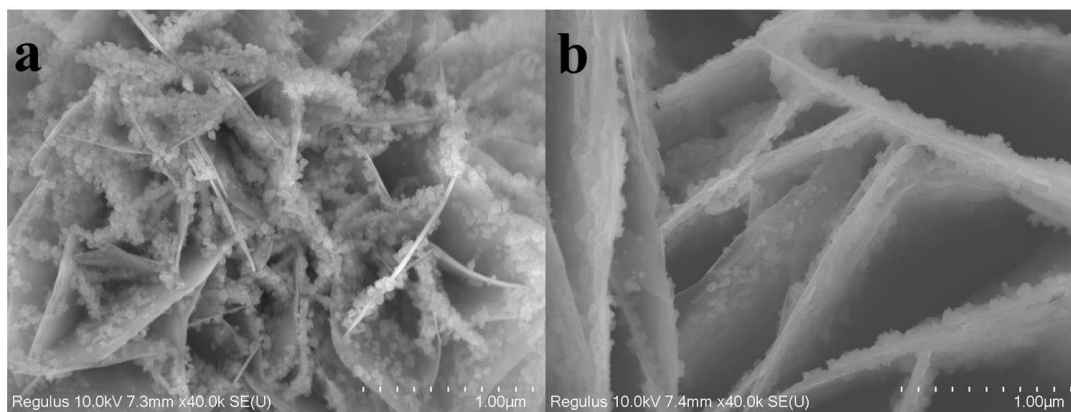
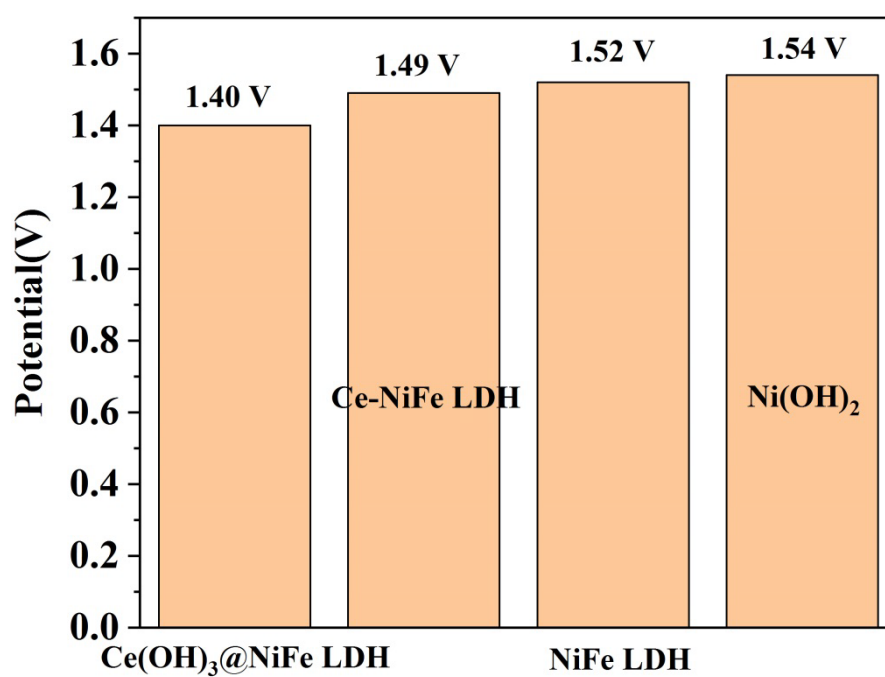


Fig. S6 XRD of fresh (a) and recovered (b) Ce(OH)<sub>3</sub>@NiFe LDH/NF for OER.



**Fig. S7** SEM of fresh (a) and recovered (b)  $\text{Ce(OH)}_3\text{@NiFe LDH/NF}$  for OER.



**Fig. S8** The potential of  $\text{Ni(OH)}_2\text{/NF}$ ,  $\text{NiFe LDH/NF}$ ,  $\text{Ce-NiFe LDH/NF}$  and  $\text{Ce(OH)}_3\text{@NiFe LDH/NF}$  at  $10 \text{ mA cm}^{-2}$  for UOR.

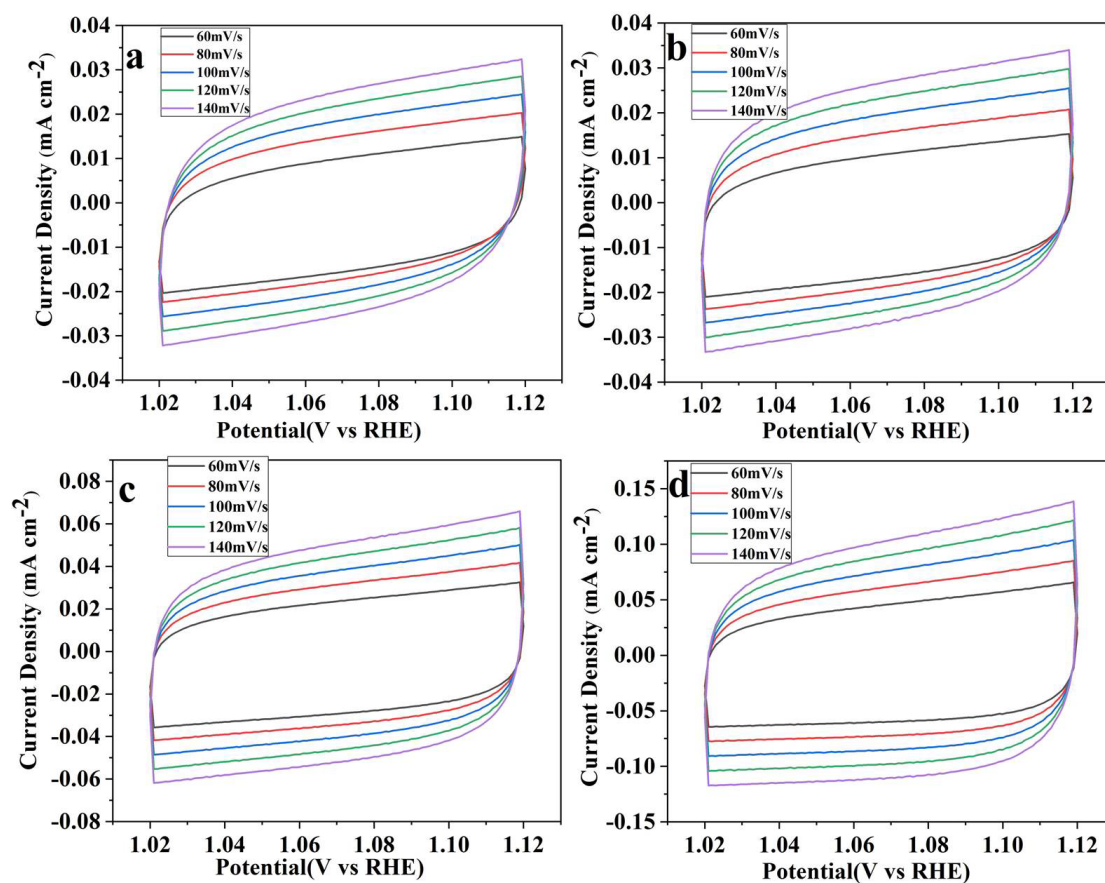


Fig. S9 CV curves with different scan rates for OER, Ni(OH)<sub>2</sub>/NF (a), NiFe LDH/NF (b), Ce-NiFe LDH/NF (c) and Ce(OH)<sub>3</sub>@NiFe LDH/NF (d).

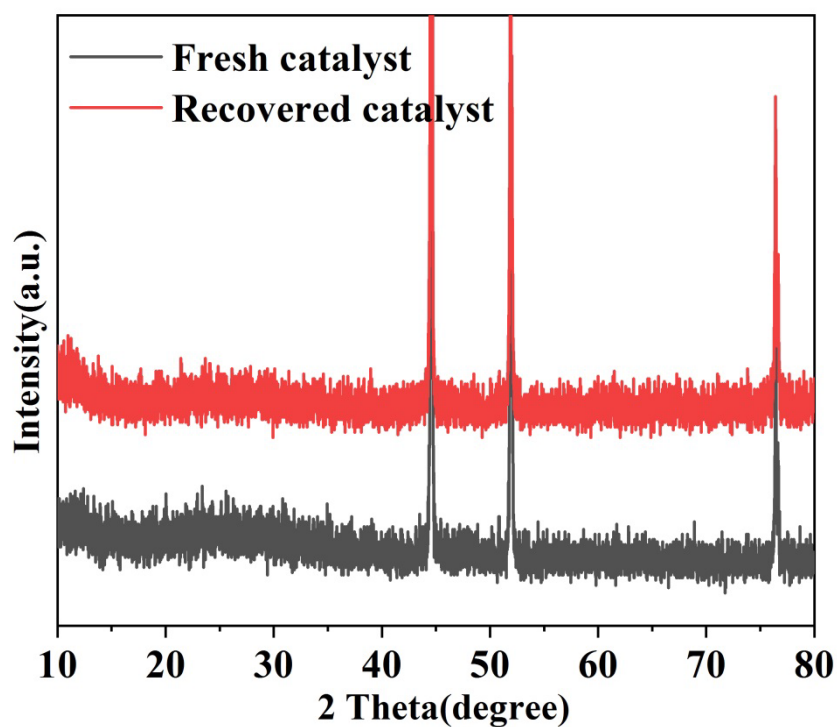
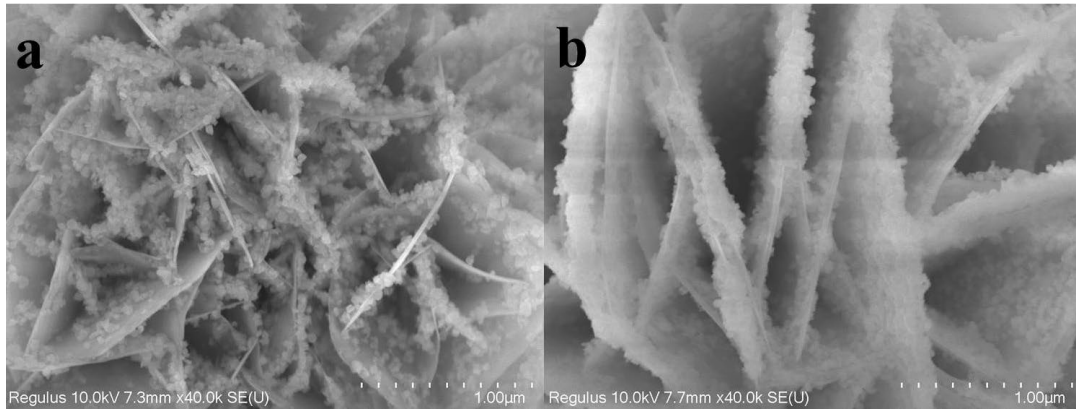


Fig. S10 XRD of fresh (a) and recovered (b) Ce(OH)<sub>3</sub>@NiFe LDH/NF for UOR.





**Fig. S11** SEM of fresh (a) and recovered (b)  $\text{Ce}(\text{OH})_3@ \text{NiFe}$  LDH/NF for UOR.