

## SUPPORTING INFORMATION

# Superior Catalytic Activity of $\alpha$ -Ni(OH)<sub>2</sub> for Urea Electrolysis

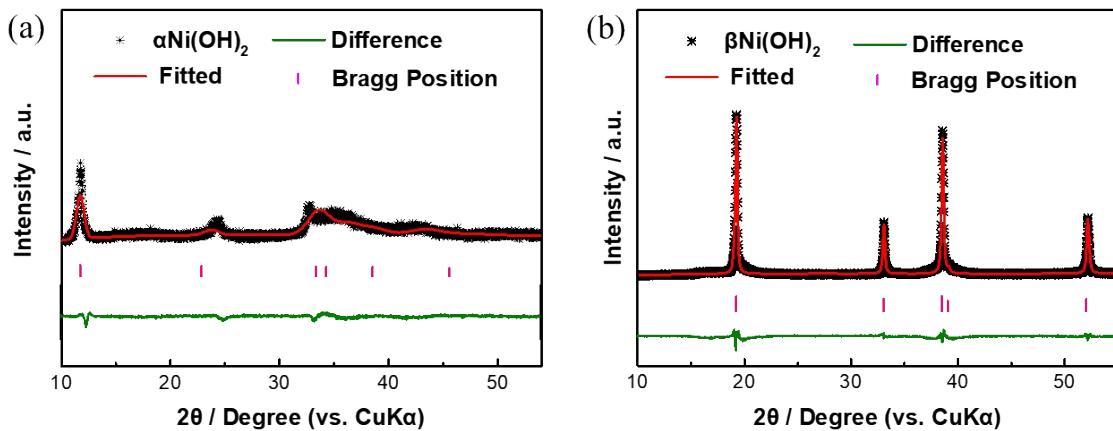
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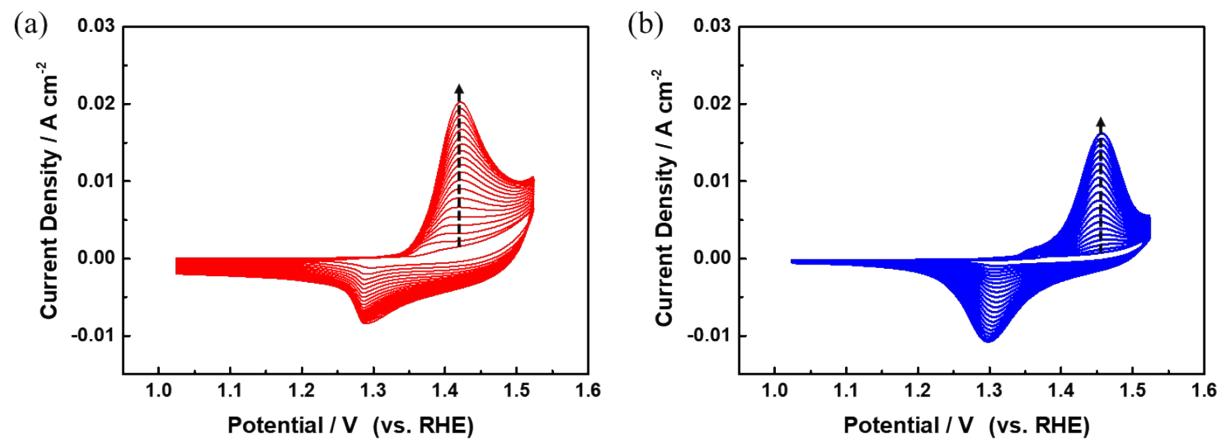
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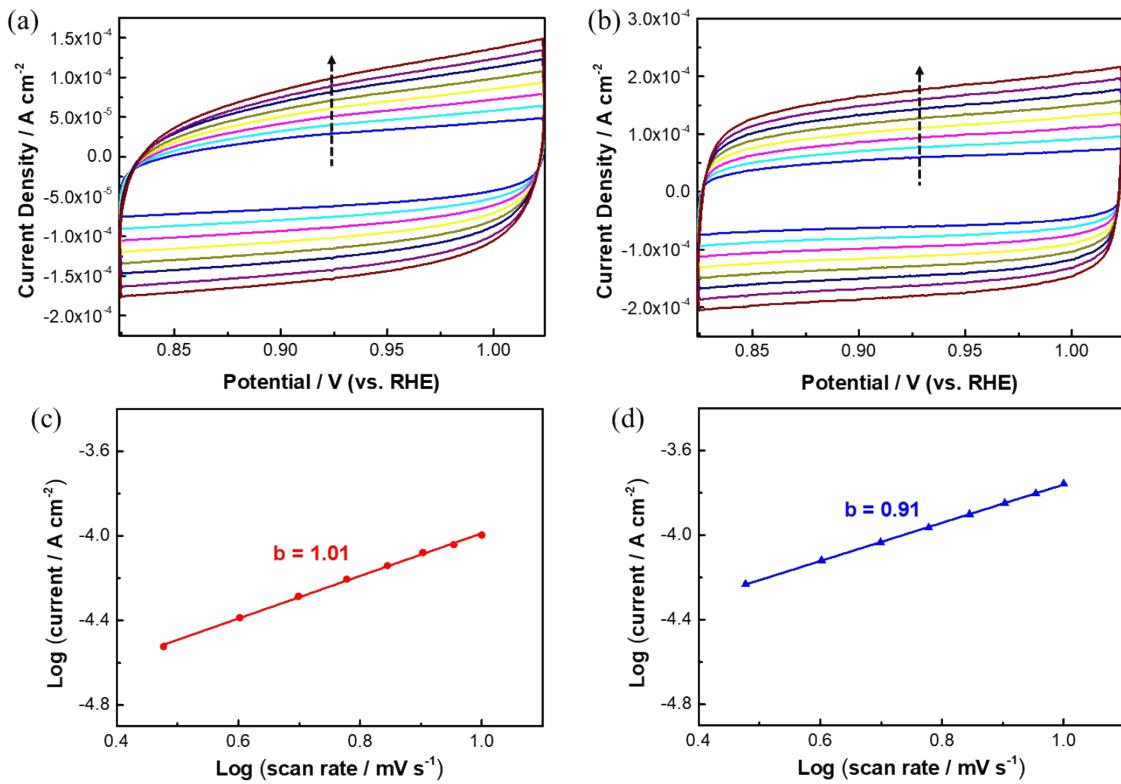
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**Figure S1** XRD refinement of (a)  $\alpha\text{-Ni(OH)}_2$  and (b)  $\beta\text{-Ni(OH)}_2$ .



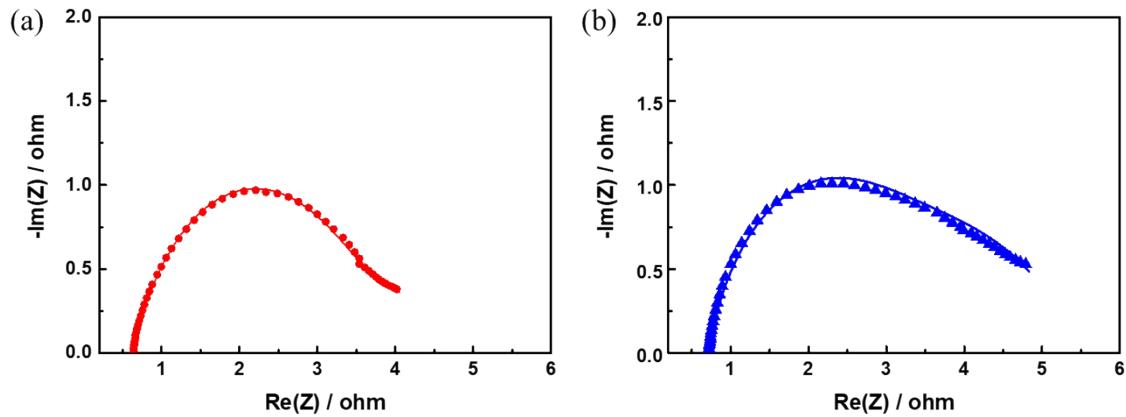
**Figure S2** The initial 20 CV curves of (a)  $\alpha\text{-Ni(OH)}_2$  and (b)  $\beta\text{-Ni(OH)}_2$  measured at the scan rate of 5 mV s $^{-1}$  in the potential window of 1.02–1.52 V in 1 M KOH electrolyte.



**Figure S3** CV curves of (a)  $\alpha$ -Ni(OH)<sub>2</sub> and (b)  $\beta$ -Ni(OH)<sub>2</sub> recorded at the scan rate of 30–100 mV s<sup>-1</sup> in the potential window between 0.82 and 1.02 V for the evaluation of double-layer capacitance; b-value determination at 0.92 V of (c)  $\alpha$ -Ni(OH)<sub>2</sub> and (d)  $\beta$ -Ni(OH)<sub>2</sub>.

**Table S1** Comparison of the UOR performance of catalysts reported in the literature.

Catalysts	Electrolyte	Onset potential (V vs RHE) at 10 mA/cm <sup>2</sup>	Current density (mA cm <sup>-2</sup> ) at 1.5 (V vs RHE)
<b><math>\alpha</math>-Ni(OH)<sub>2</sub> this work</b>	<b>1 M KOH + 0.33 M urea</b>	<b>1.40</b>	<b>58</b>
Ni(OH) <sub>2</sub> nanomeshes <sup>1</sup>	1 M KOH + 0.33 M urea	1.35	~22
Ni(OH) <sub>2</sub> nanoflakes <sup>2</sup>	1 M KOH + 0.3 M urea	~1.42	~50
Ni(OH) <sub>2</sub> nanoparticles <sup>3</sup>	5 M KOH + 1 M urea	1.41	~82
Ni(OH) <sub>2</sub> nanotubes <sup>4</sup>	1 M KOH + 0.33 M urea	~1.41	~71
Ni(OH) <sub>2</sub> nanocups <sup>5</sup>	1 M KOH + 0.33 M urea	~1.47	~16
S-Ni(OH) <sub>2</sub> <sup>6</sup>	1 M KOH + 0.33 M urea	1.32	~35
NiCr/C <sup>7</sup>	1 M KOH + 0.33 M urea	1.34	~62
LaNiO <sub>3</sub> <sup>8</sup>	1 M KOH + 0.33 M urea	~1.41	~23
NiCo <sub>2</sub> O <sub>4</sub> <sup>9</sup>	1M KOH + 0.33 M urea	1.43	~22
NiMoO <sub>4</sub> <sup>10</sup>	1 M KOH + 0.50 M urea	~1.37	~125

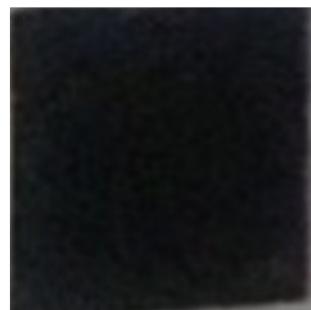


**Figure S4** EIS fitting curves of (a)  $\alpha\text{-Ni(OH)}_2$  and (b)  $\beta\text{-Ni(OH)}_2$  from Figure 5a.

**Table S2** EIS fitting results of (a)  $\alpha\text{-Ni(OH)}_2$  and (b)  $\beta\text{-Ni(OH)}_2$  from Figure 5a.

	$R_s$ ( $\Omega$ )	$R_1$ ( $\Omega$ )	$R_2$ ( $\Omega$ )
$\alpha\text{-Ni(OH)}_2$	0.63	2.91	1.57
$\beta\text{-Ni(OH)}_2$	0.71	2.58	2.18

(a)  $\alpha\text{-Ni(OH)}_2$

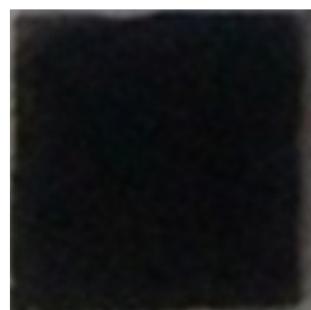


Pristine



After 20,000 s

(b)  $\beta\text{-Ni(OH)}_2$



Pristine



After 20,000 s

**Figure S5** The photograph of the (a)  $\alpha\text{-Ni(OH)}_2$  and (b)  $\beta\text{-Ni(OH)}_2$  electrode before and after stability tests.

## Notes and references

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