

## Supporting Information

### $\alpha$ and $\beta$ -motifs Interstratified $\beta_{bc}$ -Nickel Hydroxide for Enhanced OER and UOR Electrocatalysis

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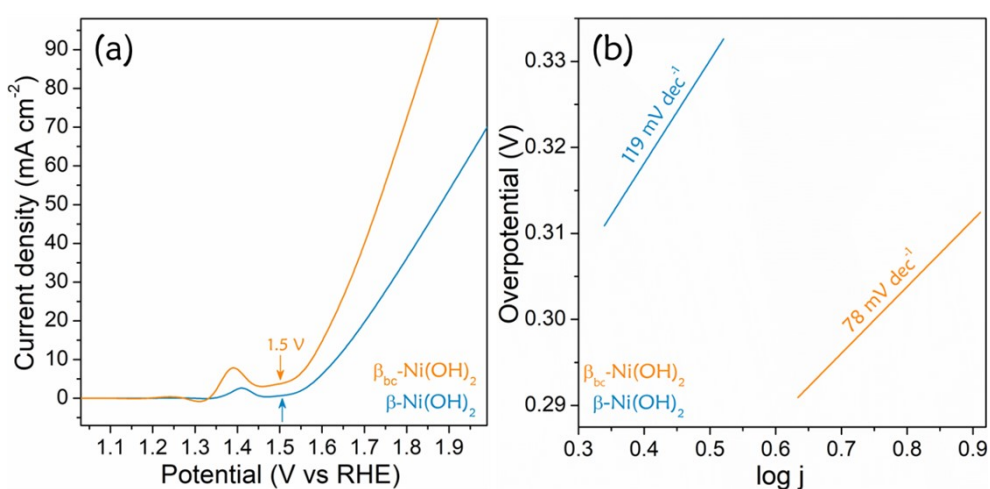
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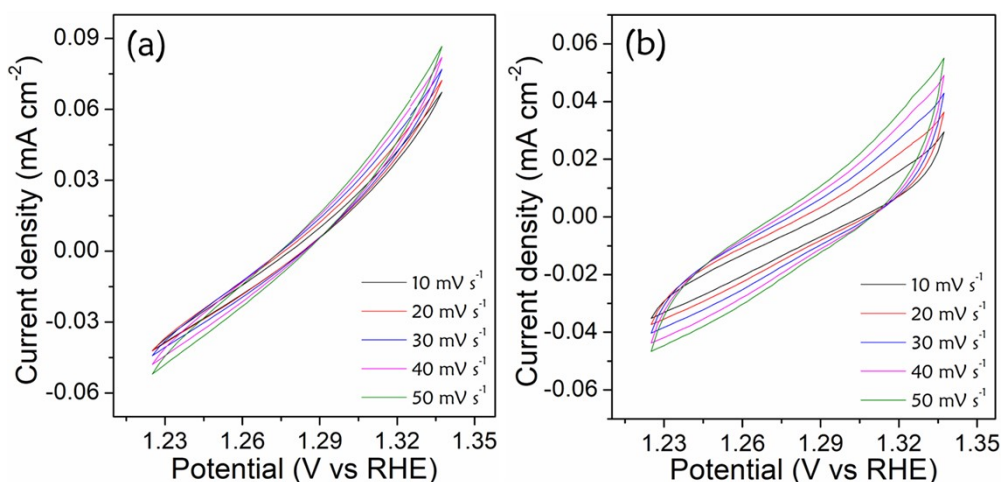
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#### DIFFaX simulation

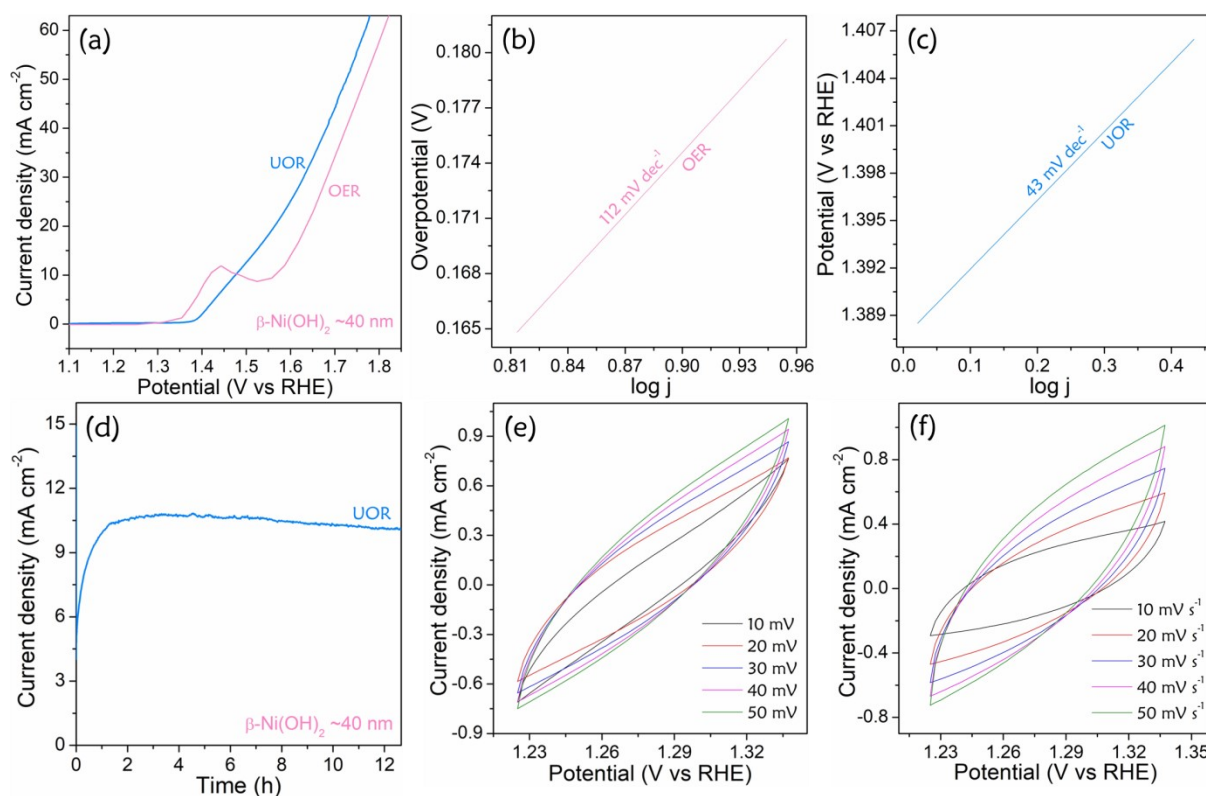
The structure parameters of  $\beta$ -nickel hydroxide were obtained from JCPDS-PDF 003-0177. Hydrogen atoms and the intercalated anions were not included in the calculation. A Lorentzian peak broadening of  $0.5^\circ$  was employed to account for the instrumental broadening and the other unaccounted causes. A platelet thickness of  $\sim 10$  nm (20 layers in registry) was assumed and the percentage of  $\alpha$ -motifs was varied from 0 to 20. The best fit was found for 12%  $\alpha$ -motifs.



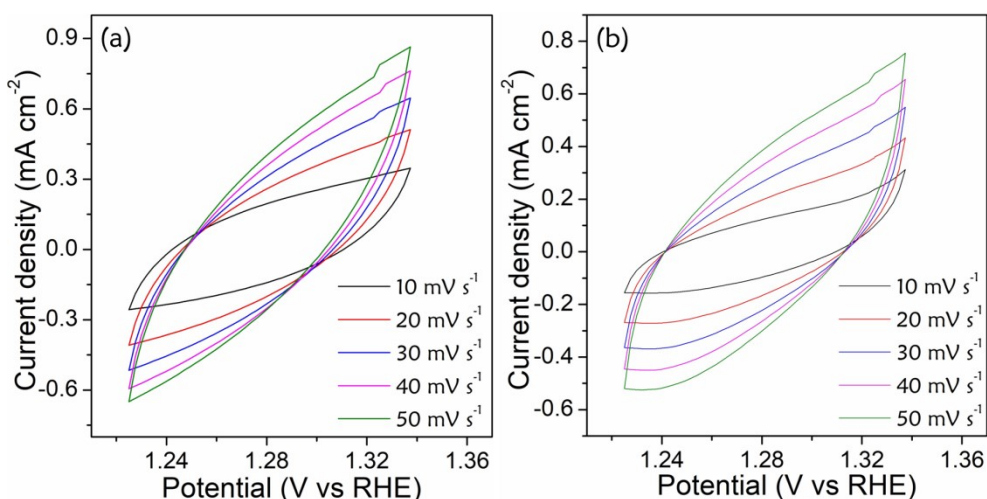
**Figure S1** Data without IR correction: (a) LSV and the corresponding (b) Tafel plot ( $\eta$  vs  $\log j$ ) of  $\beta$ -nickel hydroxide and  $\beta_{bc}$ -nickel hydroxide recorded at a scan rate of  $1 \text{ mV s}^{-1}$  respectively in 1M KOH.



**Figure S2** Cyclic voltammograms of (a)  $\beta$ -nickel hydroxide and (b)  $\beta_{bc}$ -nickel hydroxide recorded at various scan rates in 1 M KOH. This data was used to estimate the electrochemical surface area (ECSA) of the electrode materials.



**Figure S3** Electrochemical studies of  $\sim 40$  nm  $\beta$ -nickel hydroxide obtained by hydrothermal treatment of  $\beta_{bc}$ -nickel hydroxide in 0.2 M KOH for 12h. LSV @  $5 \text{ mV s}^{-1}$  in 1 M KOH without urea (OER) and with 0.33 M urea (UOR) (a); Tafel plot ( $\eta$  vs  $\log j$ ) (b and c); Chronoamperometry ( $j$  vs  $t$ ) at an applied potential of  $1.4 \text{ V}_{\text{RHE}}$  in 1 M KOH + 0.33 M urea (UOR) (d); Cyclic voltammograms at various scan rates in 1 M KOH (OER) (e) and 1 M KOH + 0.33 M urea (UOR) (f). This data was used to estimate the electrochemical surface area (ECSA) of the electrode material.



**Figure S4** Cyclic voltammograms of (a)  $\beta$ -nickel hydroxide and (b)  $\beta_{bc}$ -nickel hydroxide recorded at various scan rate in 1M KOH + 0.33 M urea. This data was used to estimate the electrochemical surface area (ECSA) of the electrode materials.

**Table S1** Comparison of the electrocatalytic OER activity of various nickel hydroxide-based materials reported in the literature.

Catalyst	OER: $\eta_{10}$ (mV)	Tafel slope (mV/dec)	Ref
$\beta_{bc}$ -Ni(OH) <sub>2</sub>	320	60	this work
$\beta$ -Ni(OH) <sub>2</sub>	370	95	
$\alpha$ -Ni(OH)NO <sub>3</sub> hollow spheres	331	42	1
$\beta$ -Ni(OH) <sub>2</sub> hexagonal plates	444	111	
CNT/ultra-thin $\beta$ -Ni(OH) <sub>2</sub> nanoplate	474	87	2
Ultrathin $\beta$ -Ni(OH) <sub>2</sub>	470	60	
Ultrathin $\beta$ -Ni <sub>0.78</sub> Fe <sub>0.22</sub> (OH) <sub>2</sub>	320	35	3
$\beta$ -Ni(OH) <sub>2</sub> films	421	58	
rGO- $\beta$ -Ni(OH) <sub>2</sub>	378	56	4
Electrodeposited Ni(OH) <sub>2</sub> on CNT	358	-	
CoNi(OH) <sub>2</sub> Nanoflowers @V <sub>2</sub> O <sub>5</sub>	350	108	6
Ultrathin Ni(OH) <sub>2</sub> nanosheet	330	41	7
$\beta$ -NiS@ $\beta$ -Ni(OH) <sub>2</sub>	330	53	8
NiV-LDH	318	50	9
Ni/Ni(OH) <sub>2</sub> Hybrid	310	75	10
Electrodeposited $\alpha$ -Ni(OH) <sub>2</sub> on Ti	310	42.6	11
$\beta$ -Ni(OH) <sub>2</sub> nanoparticles	300	53	12
NiFe-dodecyl sulfate LDH	318	80	
NiFe-dodecyl sulfate LDH	300	36	14
NiFe LDH nanosheets	300	40	15
Ni(OH) <sub>2</sub>	290	123.5	16
Sn-Ni(OH) <sub>2</sub>	246	65.5	
CoFe <sub>2</sub> O <sub>4</sub> / $\beta$ -Ni(OH) <sub>2</sub>	278	67	17
$\beta$ -Fe-Ni(OH) <sub>2</sub>	260	32	18
NF/NiMoO <sub>4</sub> /NiS/Ni(OH) <sub>2</sub>	260	110	19
Yolk-Shell $\alpha/\beta$ -Ni(OH) <sub>2</sub> /Carbon	254	55	20
$\beta$ -Fe-Ni(OH) <sub>2</sub> /nickel foam	219	53	21
$\beta$ -Ni(OH) <sub>2</sub> nanoribbon	210	74	22

Ultrathin $\alpha$ -Ni(OH) <sub>2</sub> /Ni Foam	192	108	23
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**Table S2:** Comparison of the electrocatalytic UOR activity of various nickel hydroxide-based materials reported in the literature.

Catalyst	Onset (mV)	Potential (mV) @ 10mA cm <sup>-2</sup>	Tafel slope (mV/dec)	Ref
$\beta_{bc}$ -Ni(OH) <sub>2</sub>	1.36	1.39	25	this work
Ultrathin Ni(OH) <sub>2</sub> nanoflakes	1.37	1.41	36	1
Ni(OH) <sub>2</sub> /CuCo/Ni(OH) <sub>2</sub> @ Ni foam	1.23	1.33	92	2
1% Ce: $\alpha$ -Ni(OH) <sub>2</sub> /NF	1.29	1.31	25	3
1% Ce: $\beta$ -Ni(OH) <sub>2</sub> /NF	1.31	1.35	52	
Ni(OH) <sub>2</sub> nanosheets @ Carbon Cloth	1.31	1.32	81	4
Single layer $\alpha$ -Ni(OH) <sub>2</sub> @ Carbon Cloth	1.37	1.39	-	5
Oxygen vacancy rich V-Ni(OH) <sub>2</sub>	1.36	1.38	29	6
vanadium doped $\alpha$ -Ni(OH) <sub>2</sub>	1.31	1.33	32	7
Metallic Ni(OH) <sub>2</sub> nanosheets	1.37	1.39	-	8
V <sub>Ni</sub> rich $\alpha$ -Ni(OH) <sub>2</sub>	1.37	1.39	30	9
Ni(OH) <sub>2</sub> @ Nickel Foam	1.34	1.35	24	10
$\alpha$ -nickel hydroxy chloride	1.32	1.34	41	11
$\beta$ -Ni(OH) <sub>2</sub>	1.35	1.38	74	
1% Cu: $\alpha$ -Ni(OH) <sub>2</sub> /NF	1.31	1.34	38	12
Ni(OH)S	1.31	1.34	52	13
NiCo-LDHs	1.35	1.37	-	14
$\beta$ -Ni(OH) <sub>2</sub> -NF	1.33	-	122	15
$\alpha$ -Ni(OH) <sub>2</sub> /NF	1.33	-	61	
Co-Ni(OH) <sub>2</sub> /NF	1.30	1.35	24	
Porous Ni(OH) <sub>2</sub> nanosheets	1.42	1.45	43	16
Ru nanoparticles – V doped NiFe LDH	1.37	1.4	-	17

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