Supporting Information Defect Engineering of Nickel Hydroxide Nanosheets by Ostwald Ripening for Enhanced Selective Electrocatalytic Alcohol Oxidation

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Figure S1. SEM images of CF.



Figure S2. XRD patterns of 0.5, 0.75, 1.0, 1.25 and 1.5 h-Ni(OH)₂.



Figure S3. SEM images of 1.0 NiZn(OH)₂.



Figure S4. TEM images of (a) 1.0 NiZn(OH)₂, (b) 1.0 h-Ni (OH)₂.



Figure S5. SEM images of (a, b) 0.5 NiZn(OH)₂, (c, d) 0.5 h-Ni (OH)₂.



Figure S6. SEM images of (a, b) 0.75 NiZn(OH)₂, (c, d) 0.75 h-Ni (OH)₂.



Figure S7. SEM images of (a, b) 1.25 NiZn(OH)₂, (c, d) 1.25 h-Ni (OH)₂.



Figure S8. SEM images of (a, b) 1.5 NiZn(OH)₂, (c, d) 1. 5 h-Ni(OH)₂.



Figure S9. TEM images of (a, b) 1.0 NiZn(OH)₂.



Figure S10. TEM images of (a, b) 0.5 h-Ni(OH)₂.



Figure S11. TEM images of (a, b) 0.75 h-Ni(OH)₂.



Figure S12. TEM images of (a, b) 1.25 h-Ni(OH)₂.



Figure S13. EDX spectrum of 1.0 h-Ni(OH)₂.

Samples	surface O- C=O /%	Adsorbed OH /%	Lattice OH /%	Lattice O /%
0.5 h-Ni(OH) ₂	8.26	22.03	61.10	6.61
0.75 h-Ni(OH) ₂	6.48	31.76	52.35	9.41
1.0 h-Ni(OH) ₂	9.88	34.03	47.20	8.89
1.25 h-Ni(OH) ₂	10.08	31.53	50.82	7.57
1.5 h-Ni(OH) ₂	8.57	15.91	68.79	6.73

Table S1. The relative ratio of different kinds of oxygen in as-synthesized h-Ni(OH)₂.

Material	BET Surface Area /m ² g ⁻¹
0.5 h-Ni(OH) ₂	31.01
0.75 h-Ni(OH) ₂	37.94
1.0 h-Ni(OH) ₂	63.89
1.25 h-Ni(OH) ₂	88.92
1.5 h-Ni(OH) ₂	108.19

Table S2. BET specific surface area of various materials.



Figure S14. The CV curves of 0.5, 0.75, 1.0, 1.25 and 1.5 h-Ni(OH)₂.



Figure S15. Tafel plots derived from LSV curves of 0.5, 0.75, 1.0, 1.25 and 1.5 h-Ni(OH)₂.



Figure S16. Electrochemical impedance spectra of 0.5, 0.75, 1.0, 1.25 and 1.5 h-Ni(OH)₂.



Figure S17. TOF of 0.5, 0.75, 1.0, 1.25 and 1.5 h-Ni(OH)₂.



Figure S18. Durability test of $1.0 \text{ h-Ni}(\text{OH})_2$ at 1.42 V versus RHE for electrolytic water oxidation .



Figure S19. TEM images of 1.0 h-Ni(OH)₂ after durability test.



Figure S20. (a) The cyclic voltammograms of benzyl alcohol at various scan rates on 1.0 h-Ni(OH)₂. (b) I-V relationship of 1.0 h-Ni(OH)₂ from (a) in alkaline solution.



Figure S21. Conversion (%) of benzyl alcohol and selectivity (%) of oxidation products during the electrooxidation at a constant current of 20 mA in 1.0 m KOH with 40×10^{-3} M benzyl alcohol on (a) 0.5 h-Ni(OH)₂, (b) 0.75 h-Ni(OH)₂, (c) 1.25 h-Ni(OH)₂, (d) 1.5 h-Ni(OH)₂.



Figure S22. The photograph of constant current meter with concentration of 40×10^{-2} M benzoyl alcohol in ECO reaction.



Figure S23. (a) Conversion of benzyl alcohol and selectivity to benzoic acid and benzaldehyde and faradaic efficiency in 1.0 h-Ni (OH)₂ and 1.0 NiZn(OH)₂. (b) Conversion (%) of benzyl alcohol and selectivity (%) of oxidation products on 1.0 NiZn(OH)₂.



Figure S24. (a, b) TEM images and (c, d) SEM images of $1.0 \text{ h-Ni}(\text{OH})_2$ after recycling measurement.

Catalyst	Main product	Conv (%	Sel (%	FE (%)	Reference
1.0 h- Ni(OH) ₂	Benzoic acid	99.9	99.3	98.6	This work
Ni foam	Benzoic acid	93.69	75.85	81.52	This work
Co ₃ O ₄ NWs/Ti	Benzoic acid	99	92	56.82	ACS Nano, 2017, 11, 12365-12377
Co _{0.83} Ni _{0.17} /AC	Benzoic acid	100	99.4	96	New J. Chem., 2018, 42, 6381

Table S3. Comparison of 1.0 h-Ni(OH)₂ and other electrocatalysts on ECO of benzyl alcohol to benzoic acid.



Figure S25. Conversion of benzyl alcohol and selectivity to benzoic acid and benzaldehyde and Faradaic efficiency in $1.0 \text{ h-Ni}(\text{OH})_2$ after introducing TEMPO. Pentagonal stars represent Faraday efficiency.



Figure S26. TEM images of 1.0 PtO₂/h-Ni(OH)₂.



Figure S27. (a) HER polarization curves of the 1.0 $PtO_2/h-Ni(OH)_2$, 1.0 $h-Ni(OH)_2$ and CF. (b) Overpotential required at 10mA cm⁻². (c) Tafel plots derived from LSV curves of CF, 1.0 $h-Ni(OH)_2$ and 1.0 $PtO_2/h-Ni$ (OH)₂. (d) The HER Faradaic efficiency of the 1.0 $PtO_2/h-Ni(OH)_2$ catalyst in 1.0 M KOH.



Figure S28. Chronopotentiometry curve of 1.0 $PtO_2/h-Ni(OH)_2$ at -0.076 V versus RHE.



Figure S29. (a, b) The photograph of paired electrolysis system.

Models	oxygen atom in benzyl	benzyl alcohol
	alcohol	
vacancy-free Ni(OH) ₂	-1.151	-0.047
V ₀ -Ni(OH) ₂	-1.260	-0.114

Table S4. The charge numbers of benzyl alcohol and oxygen atom in benzyl alcoholon vacancy-free $Ni(OH)_2$ and V_O - $Ni(OH)_2$.