Supporting Information

NiMn Compound Nanosheets for Electrocatalytic Water Oxidation:

Effects of Atomic Structures and Oxidation States

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Figure S1. SEM images of Ni-birnessite showing the large nanosheet microstructure.



Figure S2. TEM images of (A) Ni-Birnessite, (B) NiMnO_x-B, (C) NiMn LDH and (D) NiMnO_x-L, insets are the SAED images of the corresponding samples.



Figure S3. SEM images of NiMn LDH showing the thin nanosheet microstructure.



Figure S4. AFM images and the corresponding height profiles of the four NiMn compounds. A) NiMn LDH, B) NiMnO_x-L, C) Ni-birnessite, D) NiMnO_x-B. The thicknesses of NiMn LDH and NiMnO_x-L were less than 10 nm, and those of Ni-birnessite and NiMnO_x-B were ~100 nm.



Figure S5. SEM images of NiMnOx-B showing the retained nanosheet microstructure.



Figure S6. SEM images of NiMnO_x-L showing the porous nanosheet microstructure.



Figure S7. XRD spectra of NiMn LDH before (black curve) and after (red curve) the annealing treatment.



Figure S8. XRD spectra of Ni-Birnessite before (black curve) and after (red curve) the annealing treatment.



Figure S9. Raman spectra of NiMnO_x-B and its precursor of Ni-birnessite.



Figure S10. The full XPS spectra of the catalysts. The atomic ratio of Ni/Mn for Nibirnessite, NiMn LDH, NiMnOx-B and NiMnOx-L is 0.44, 1.42, 0.24 and 1.32, respectively.



Figure S11. High resolution XPS spectra of the catalysts in (A) Ni, and (B) Mn 2p regions.

	NiMnO _x -B	NiMnO _x -L	NiMn LDH	Ni-birnessite	
Atomic	Layerd	Cubic	Layerd	Layerd	
structure					
Morphology	Microsheets	Porous	Nanosheets	Microsheets	
nanosheets					
Conductivity	2	3	1	4	
ECSA	NiMnO _x -B > NiMn LDH > NiMnO _x -L > Ni-birnessite				
Ni/Mn	0.24	1.32	1.42	0.44	
Ni (III)/Ni (II)	1.03	3.73	1.53	1.30	
Mn (III)/Total	0.87	0.15	0.15		
h (10 mA/cm ²)	362	383	386	412	
Tafel slope	69	71	83	98	

Table S1. Summary of the structure and performance of the as-prepared four samples.



Figure S12. CV scanning curves of the as-made samples after 50 cycles.



Figure S13. (A) LSV polarization curves, (B) Tafel plots of LDH samples synthesizd with different Ni/Mn ratio precursor. By comparing to the LDH sample with a precursor Ni/Mn ratio of 1, the precursor Ni/Mn ratio of LDH 2 and LDH 3 was 0.8 and 1.6 respectively.



Figure S14. Electrochemical performance of NiMnOx-B tested by using the graphitic rod as the counter electrode. (A) LSV curve, (B) Tafel plots.



Figure S15. The stability testing result of NiMnOx-B at the constant current density of 50 (left panel) and 100 mA/cm² (right panel) for more than 10000 s in even harsh conditions of 6 M KOH electrolyte and 60 °C temperature.

Samples	Concentration of metal (mg/L)				
	Pt	Ni	Mn		
NiMnO _x -B	N.D.	N.D.	N.D.		
NiMnO _x -L	N.D.	N.D.	N.D.		

Table S2. ICP test of the 1M KOH solution after the electrochemical tests.



Figure S16. Calculated LSV using ECSA as values, and the ECSA can be calculated according to the formula ECSA = C_{DL}/C_S , where a specific capacitance of $C_S = 0.040$ mF cm⁻² was used in this work.