

Supporting information for:

**Tunable Nano-Interfaces between MnO_x and Layered Double
Hydroxides Boost Oxygen Evolving Electrocatalysis**

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Supplementary Figures (Figures S1 – S5, Tables S1 – S4)

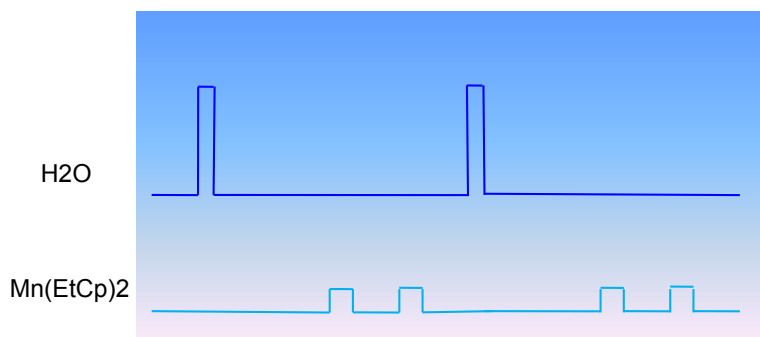


Fig. S1 Schematics for the ALD precursor sequence of growing MnO_x during each cycle.

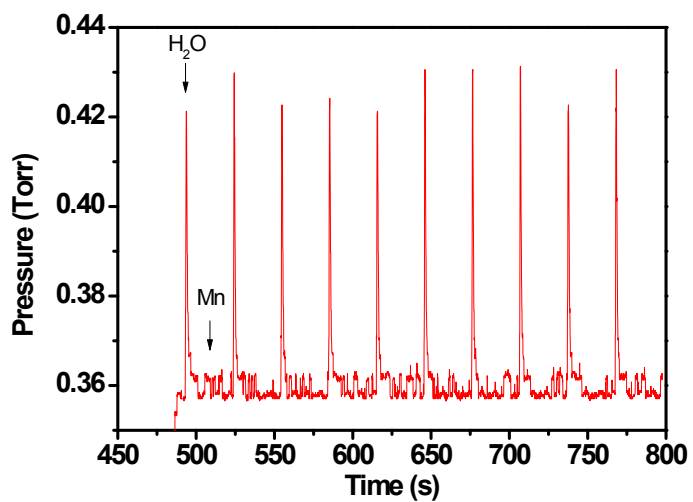
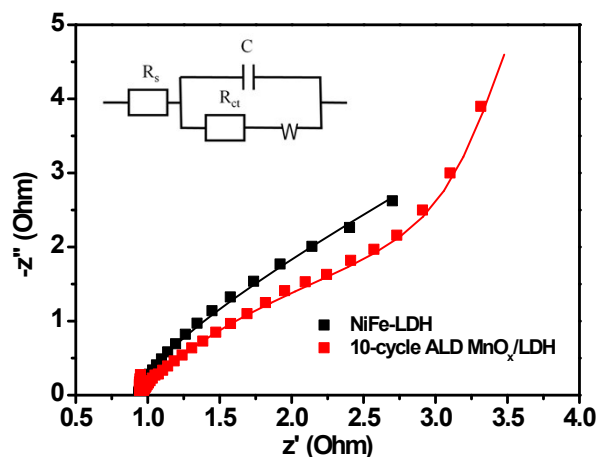


Fig. S2 Recorded ALD pulse sequences by monitoring the reactor pressure as a function of time, when using Mn(EtCp)₂ and H₂O as the precursor and the co-reactants, respectively, for ALD MnO_x growth.



| | Solution resistance (R _s), Ω | Charge transfer resistance (R _{ct}), Ω | Capacitance (C), nF cm ⁻² |
|-----------------------------------|---|---|---|
| NiFe-LDH | 0.939 | 8.125 | 18.597 |
| 10-cycle MnO _x /LDH | 0.938 | 5.890 | 18.913 |

Fig. S3 Nyquist plots of ALD MnO_x/LDH electrodes held at overpotential of 300 mV and the corresponding equivalent circuitry. Fitted values are listed in the table.

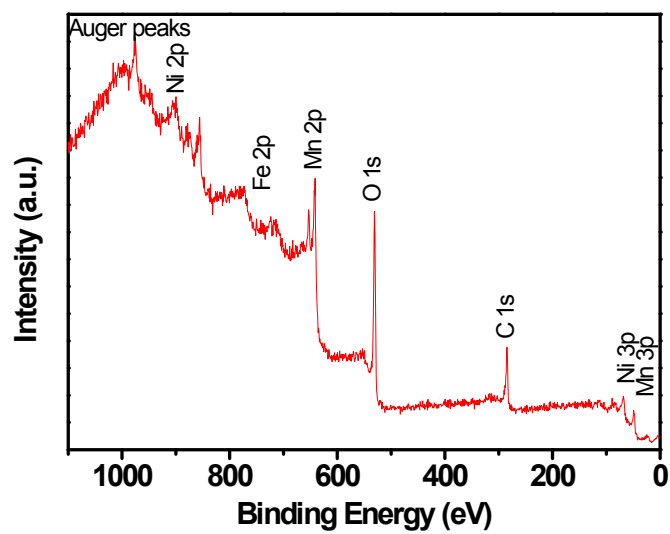


Fig. S4 XPS survey spectrum of 10-cycle ALD $\text{MnO}_x/\text{NiFe-LDH}$ samples.

Table S1 Fitting parameters used to fit the Mn 2p^{3/2} peaks (Pk).

| | Pk 1 (eV) | Pk 2 (eV) | Pk 3 (eV) | Pk 4 (eV) | Pk 5 (eV) | Pk 6 (eV) | FWHM (eV) |
|------------------------------------|--------------|--------------|--------------|--------------|--------------|--------------|--------------|
| MnO _x | 640.2 | 641.2 | 642.1 | 643.05 | 644.2 | 645.9 | 1.23 |
| MnO _x /LDH before EC | 640.2 | 641.2 | 642.1 | 643.05 | 644.2 | 645.9 | 1.23 |
| MnO _x /LDH after EC | 640.2 | 641.2 | 642.1 | 643.05 | 644.2 | 645.9 | 1.23 |

Table S2 Fitting parameters used to fit the Ni 2p^{3/2} and Fe 2p^{3/2} peaks (Pk).

| | Pk Ni2p ^{3/2} (eV) | Pk Fe2p ^{3/2} (eV) |
|-----------------------|-----------------------------|-----------------------------|
| MnO _x /LDH | 856.0 | 713.0 |
| NiFe-LDH | 856.8 | 715.0 |

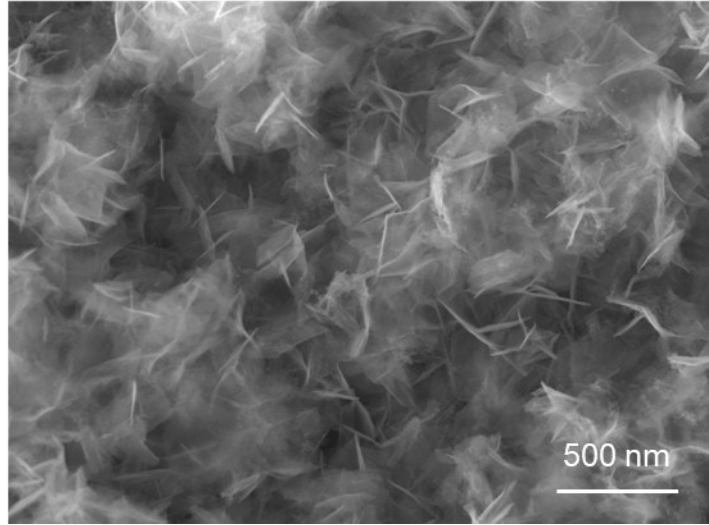


Fig. S5 SEM image of the 10-cycle ALD $\text{MnO}_x/\text{NiFe-LDH}$ electrode after 10 h stability testing.

Table S3 ICP-MS results of the NiFe-LDH samples.

| Sample | Ni | Fe | Mn |
|--|----------|----------|----------|
| NiFe-LDH | 0.441 mg | 0.570 mg | NA |
| 10 MnO _x /NiFe-LDH | 0.462 mg | 0.559 mg | 0.067 mg |
| 10 MnO _x /NiFe-LDH after stability test | 0.431 mg | 0.540 mg | 0.053 mg |

Table S4 The TOF values of the as-prepared samples calculated at 300 mV overpotentials.

| Sample | TOF (s ⁻¹) |
|-------------------------------|------------------------|
| NiFe-LDH | 0.0043 |
| 10 MnO _x /NiFe-LDH | 0.0066 |