Electronic Supplementary Information (ESI)

In situ semi-transformation from heterometallic MOFs to Fe-Ni LDH/MOF hierarchical architectures for boosted oxygen evolution reaction

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**Fig. S1** PXRD patterns of FeNi-MOF-74 precursors with different metal ratio.

**Fig. S2** The SEM of MOF-74(Fe:Ni=2:1).
**Fig. S3** The SEM of FeNi-MOF-74 (Fe:Ni=1:1).

**Fig. S4** The SEM of MOF-74 (Fe:Ni=2:3).
**Fig. S5** EDS of MOF-74(Fe:Ni=2:3), FeNi-MOF-74 and MOF-74(Fe:Ni=2:1).
**Fig. S6** Elemental mapping images of C, O, Fe, and Ni in FeNi-MOF-74.

**Fig. S7** PXRD patterns of Fe-Ni LDH/MOF-a2, Fe-Ni LDH/MOF-b2, Fe-Ni LDH/MOF-c2 and FeNi-MOF-74 using FeNi-MOF-74 (Fe:Ni=1:1) as precursor.
**Fig. S8** PXRD patterns of Fe-Ni LDH/MOF-b2-3h, Fe-Ni LDH/MOF-b2-6h, Fe-Ni LDH/MOF-b2-12h and FeNi-MOF-74 using FeNi-MOF-74 (Fe:Ni=1:1) as precursor.

**Fig. S9** PXRD patterns of Fe-Ni LDH/MOF-b1, Fe-Ni LDH/MOF-b2, Fe-Ni LDH/MOF-b3 and MOF-74(Fe:Ni=2:1) using FeNi-MOF-74(Fe:Ni=2:1) as precursor.
**Fig. S10** PXRD patterns of Fe-Ni LDH/MOF-a2, Fe-Ni LDH/MOF-b2, Fe-Ni LDH/MOF-c2 and MOF-74(Fe:Ni=2:1) using MOF-74(Fe:Ni=2:1) as precursor.

**Fig. S11** The SEM of Fe-Ni LDH/MOF-b1 using FeNi-MOF-74 as a precursor.
Fig. S12 The TEM of Fe-Ni LDH/MOF-b1 using FeNi-MOF-74 as a precursor.

Fig. S13 TEM image of ultra-thin LDH nanoarrays near the edge Fe-Ni LDH/MOF-b2.
**Fig. S14** The SEM of Fe-Ni LDH/MOF-b3 using FeNi-MOF-74 as a precursor.

**Fig. S15** The TEM of Fe-Ni LDH/MOF-b3 using FeNi-MOF-74 as a precursor.
Fig. S16 EDS of Fe-Ni LDH/MOF-b1, Fe-Ni LDH/MOF-b2 and Fe-Ni LDH/MOF-b3.
Fig. S17 Elemental mapping of Fe, Ni, C and O of Fe-Ni LDH/MOF-b2 that by using the FETEM-EDS.

Fig. S18 The SEM of Fe-Ni LDH/MOF-a2 using FeNi-MOF-74 as a precursor.
Fig. S19 The SEM of Fe-Ni LDH/MOF-c2 using FeNi-MOF-74 as a precursor.

Fig. S20 The SEM of Fe-Ni LDH/MOF-74 (Fe : Ni = 2 : 1)-b2 using MOF-74(Fe:Ni=2:1) as a precursor.
**Fig. S21** The XPS spectra of C 1s the FeNi-MOF-74, Ni-MOF-74 and Fe-Ni LDH/MOF-b2.

**Fig. S22** The full cyclic voltammograms (CV) of FeNi-MOF-74 and Fe-Ni LDH/MOF-b2 at a scan rate of 10 mV·s⁻¹ without iR correction.
**Fig. S23** (a) The LSV curves of MOF-74(Fe:Ni=2:3), FeNi-MOF-74 and MOF-74(Fe:Ni=2:1); (b) Overpotential at 10 mA·cm⁻² for different MOF precursors.

**Fig. S24** The LSV curves of Fe-Ni LDH/MOF-b2-3h, Fe-Ni LDH/MOF-b2-6h, Fe-Ni LDH/MOF-b2-12h and FeNi-MOF-74 using FeNi-MOF-74 as a precursor.
**Fig. S25** The LSV curves of Fe-Ni LDH/MOF-a2, Fe-Ni LDH/MOF-b2, Fe-Ni LDH/MOF-c2 and FeNi-MOF-74 using FeNi-MOF-74 as a precursor.

**Fig. S26** The LSV curves of Fe-Ni LDH/MOF-b1, Fe-Ni LDH/MOF-b2, Fe-Ni LDH/MOF-b3 and MOF-74(Fe:Ni=2:1) using MOF-74(Fe:Ni=2:1) as a precursor.
Fig. S27 The LSV curves of Fe-Ni LDH/MOF-a2, Fe-Ni LDH/MOF-b2, Fe-Ni LDH/MOF-c2 and MOF-74(Fe:Ni=2:1) using MOF-74(Fe:Ni=2:1) as a precursor.

Fig. S28 The LSV curves of Fe-Ni LDH/MOF-b1, Fe-Ni LDH/MOF-b2, Fe-Ni LDH/MOF-b3 and MOF-74(Fe:Ni=2:3) using MOF-74(Fe:Ni=2:3) as a precursor.
Fig. S29 Summary of the overpotential from the concentration effect with different MOF precursors.

Fig. S30 The CV curves of the as-prepared Fe-Ni LDH/MOF-b1, Fe-Ni LDH/MOF-b2, Fe-Ni LDH/MOF-b3 and FeNi-MOF-74 at different scan rate of 20, 40, 60, 80, 100 and 120 mV s$^{-1}$. 
**Fig. S31** TOF curves of FeNi-MOF-74 precursors and the hierarchical Fe-Ni LDH/MOF-b2 for OER reaction at different potentials.

**Fig. S32** Theoretical and experimental amounts of O\textsubscript{2} evolved during the OER at the current density of 10 mA·cm\textsuperscript{-2}.
**Fig. S33** Stability test of Fe-Ni LDH/MOF-b3 for 24 h at current density of 10 mA·cm$^{-2}$.

**Fig. S34** Stability test of FeNi-MOF-74 for 24 h at current density of 10 mA·cm$^{-2}$. 
**Fig. S35** LSV curve of Fe-Ni LDH/MOF-b2 after 24h stability testing.

**Fig. S36** The LSV curves of Fe-Ni LDH/MOF-b2 after 1000 cycles.
Fig. S37 XRD and SEM images for Fe-Ni LDH/MOF-b2 after 24 h stability test.
**Fig. S38** High-resolution XPS spectra of Ni 2p, Fe 2p, C 1s and O 1s for Fe-Ni LDH/MOF-b2 after 24 h stability test.