**Supporting Information**

# 2D Metal-Organic Framework/$Ni(OH)_2$ Heterostructure for Enhanced Oxygen Evolution Reaction

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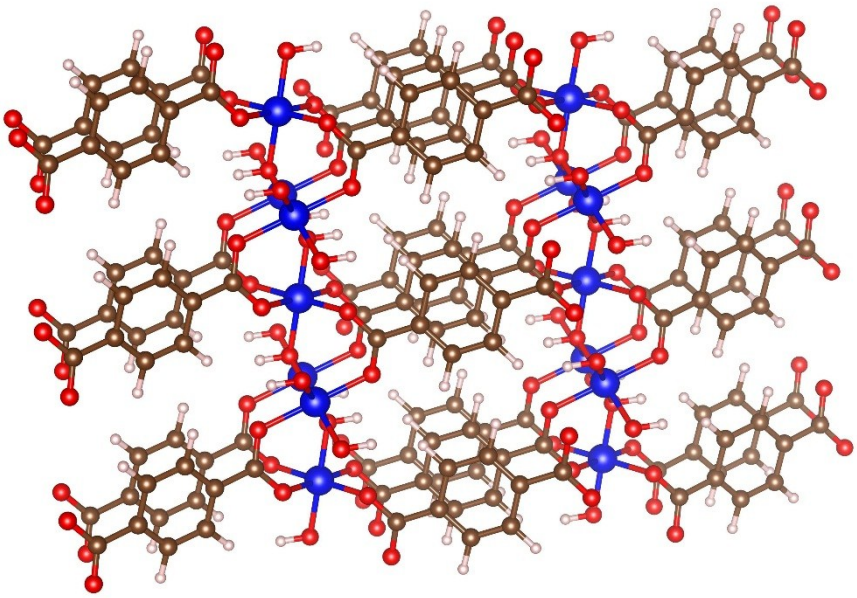

**Figure S1.** Crystal structure of Ni-BDC. Color scheme for atom representation: blue for Ni, red for O, grey for C and white for H.

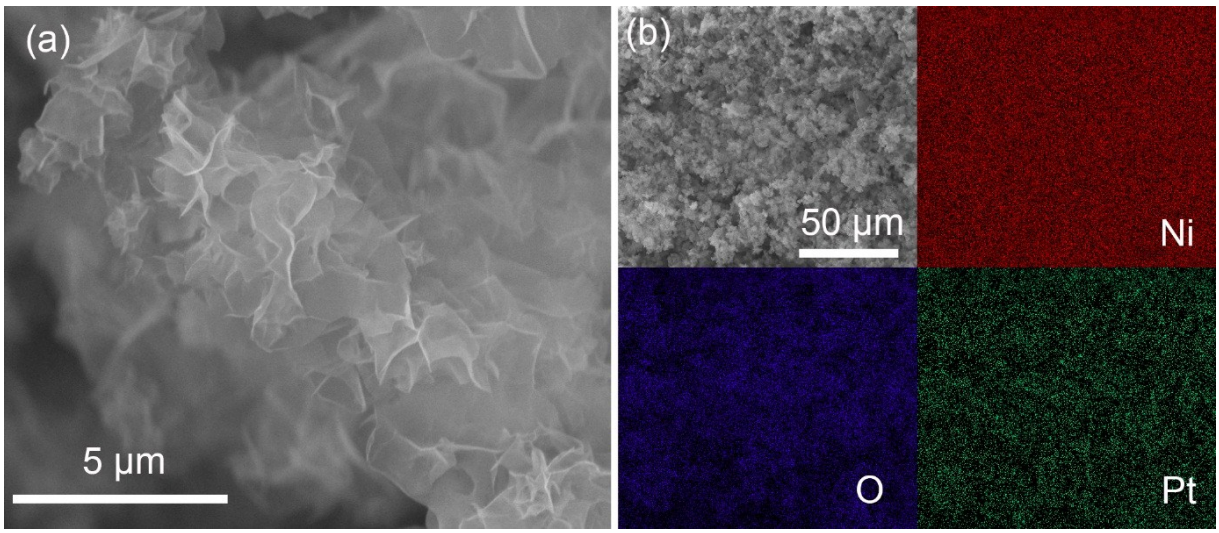


**Figure S2.** a) SEM image of $Ni(OH)_2$. b) Element mapping of $Ni(OH)_2$ for the selected area.

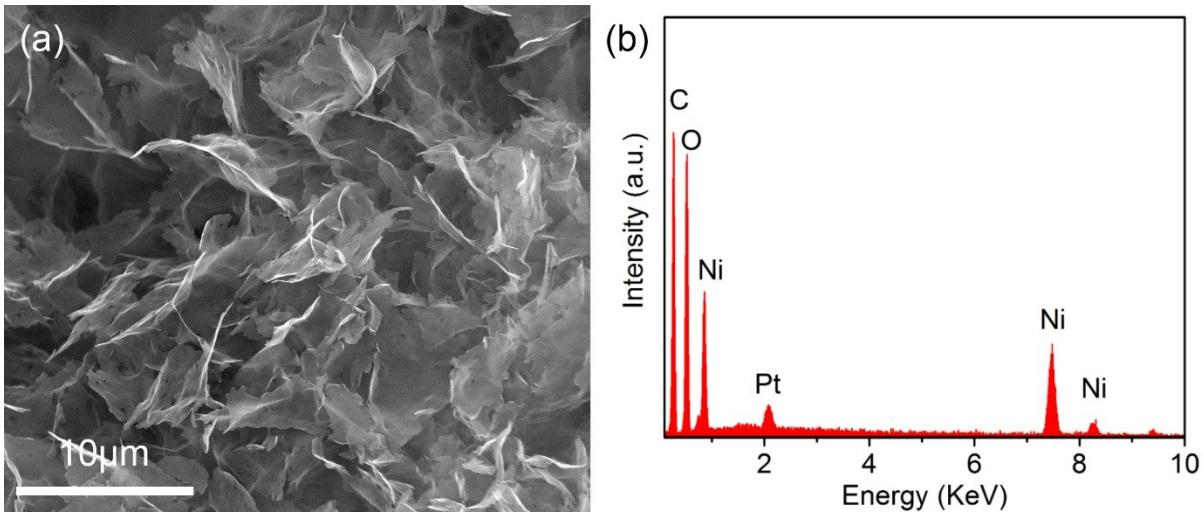


**Figure S3.** a) SEM image of Ni-BDC. b) EDS spectrum of Ni-BDC.

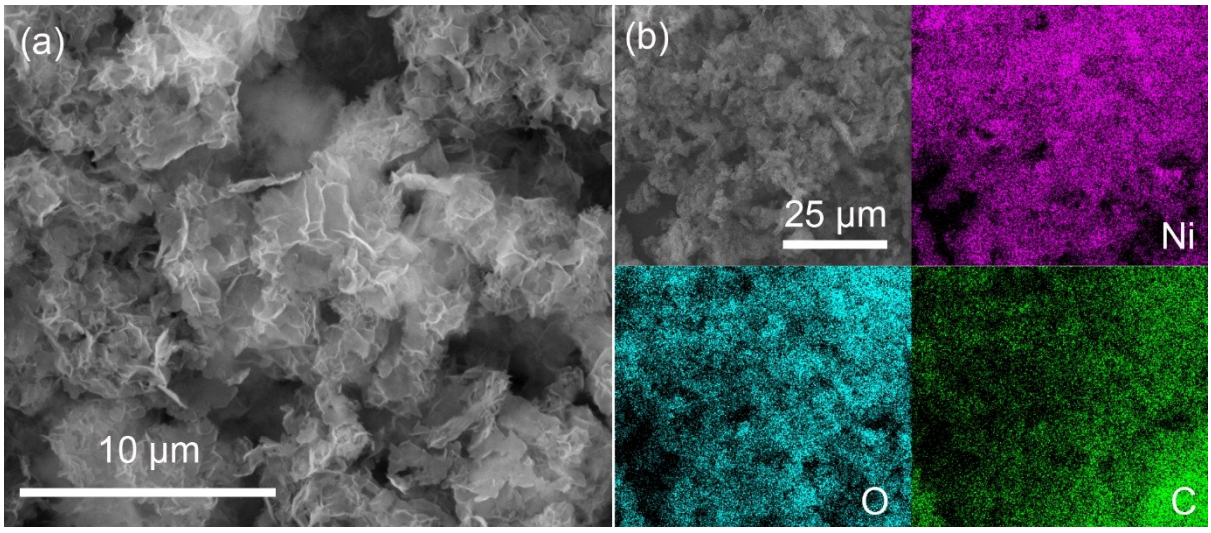


**Figure S4.** a) SEM image of Ni-BDC/$Ni(OH)_2$. b) Element mapping of Ni-BDC/$Ni(OH)_2$ for the selected area.

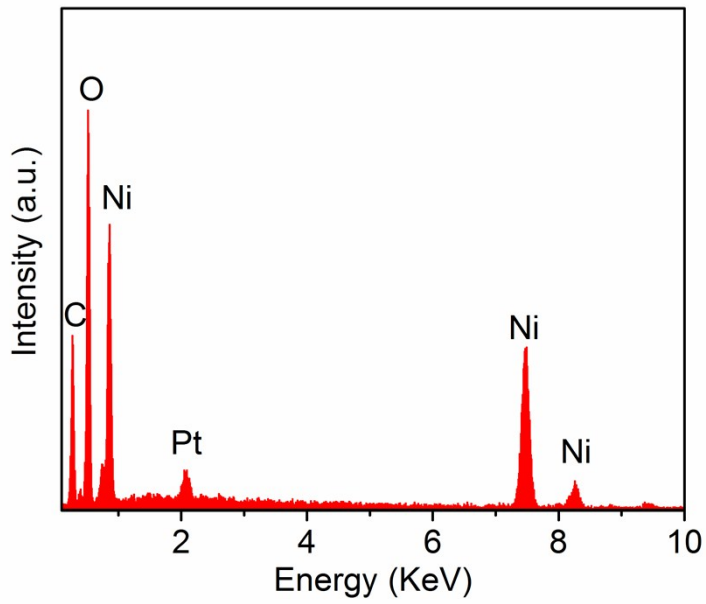


**Figure S5.** EDS spectrum for Ni-BDC/$Ni(OH)_2$ containing Ni, O and C.

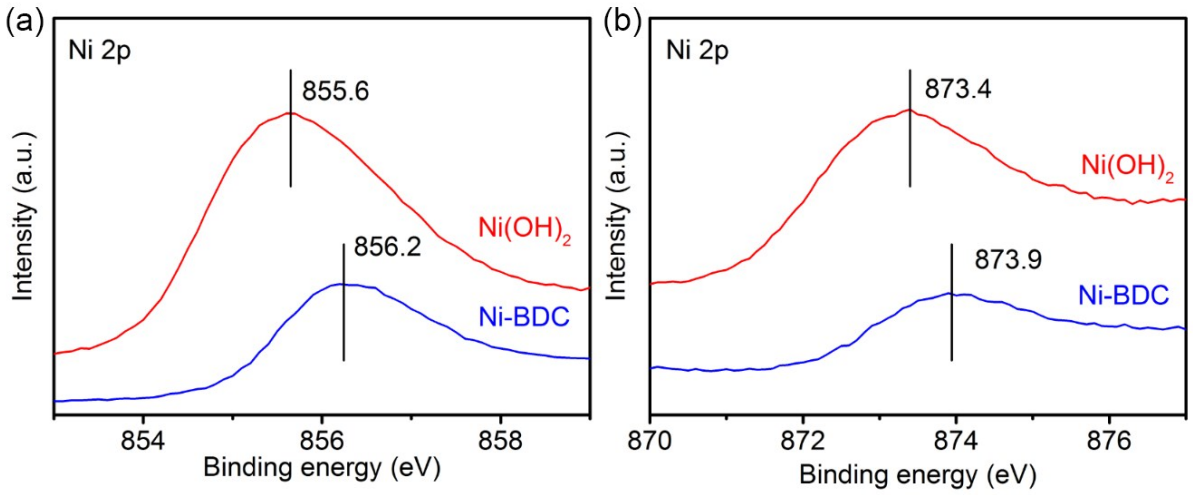


**Figure S6.** a) Magnified XPS spectra of Ni $2p_{3/2}$ for $Ni(OH)_2$ and Ni-BDC. b) Magnified XPS spectra of Ni $2p_{1/2}$ for $Ni(OH)_2$ and Ni-BDC.

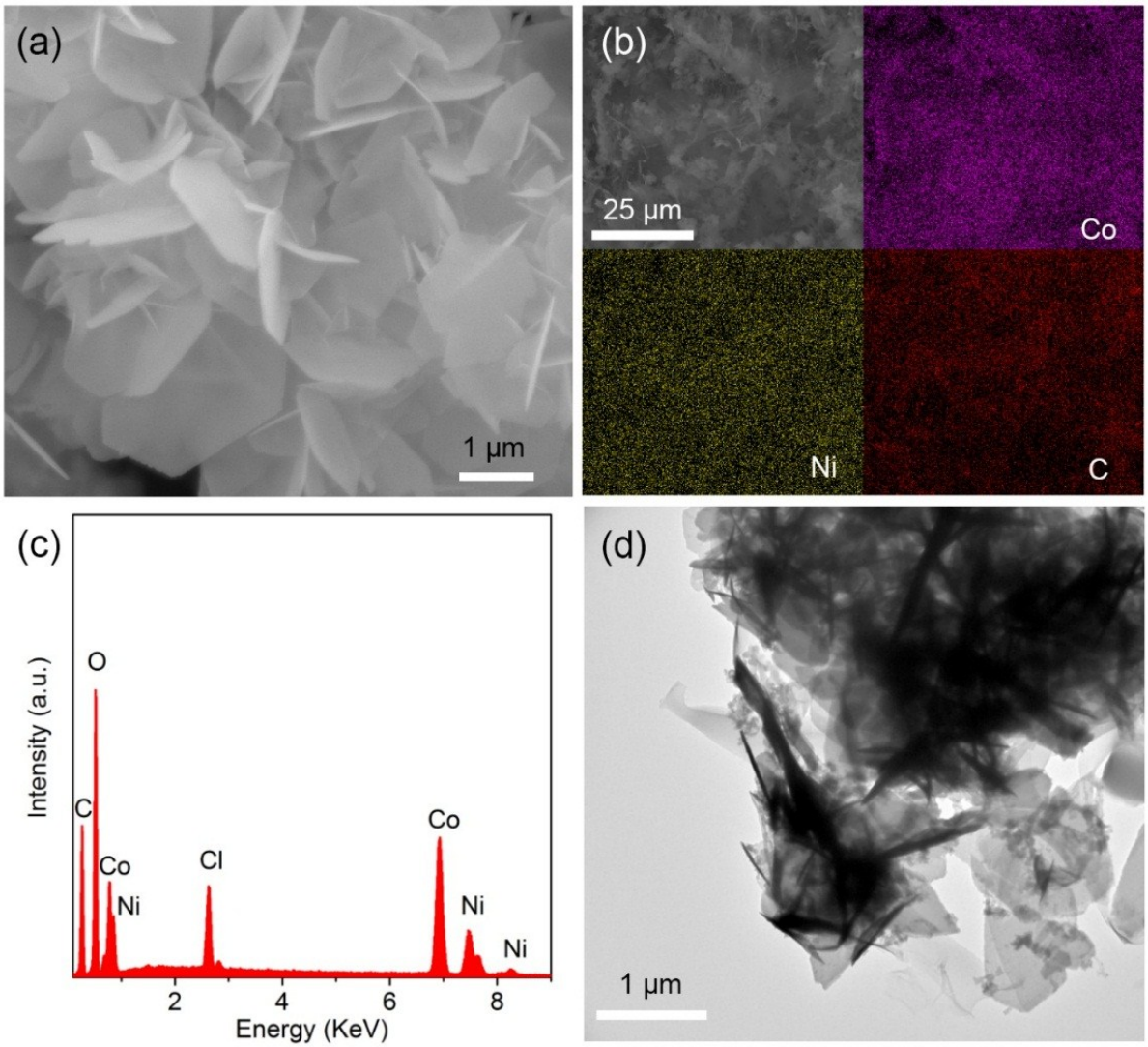


**Figure S7.** a) SEM image of $Co(OH)_2$ nanosheets. b) Element mapping of Ni-BDC/$Co(OH)_2$ for the selected area. c) EDS spectrum of Ni-BDC/$Co(OH)_2$. d) TEM image of Ni-BDC/$Co(OH)_2$. The $Co(OH)_2$ nanosheets were fabricated by a homogeneous precipitation method (Adv. Mater. Interfaces 2018, 5, 1700709). The as-fabricated $Co(OH)_2$ nanosheets were added to the solution containing BDC and $NiCl_2 \cdot 6H_2O$, and Ni-BDC/$Co(OH)_2$ hybrid nanosheets were obtained by the same sonication-assisted solution method as for Ni-BDC/$Ni(OH)_2$.

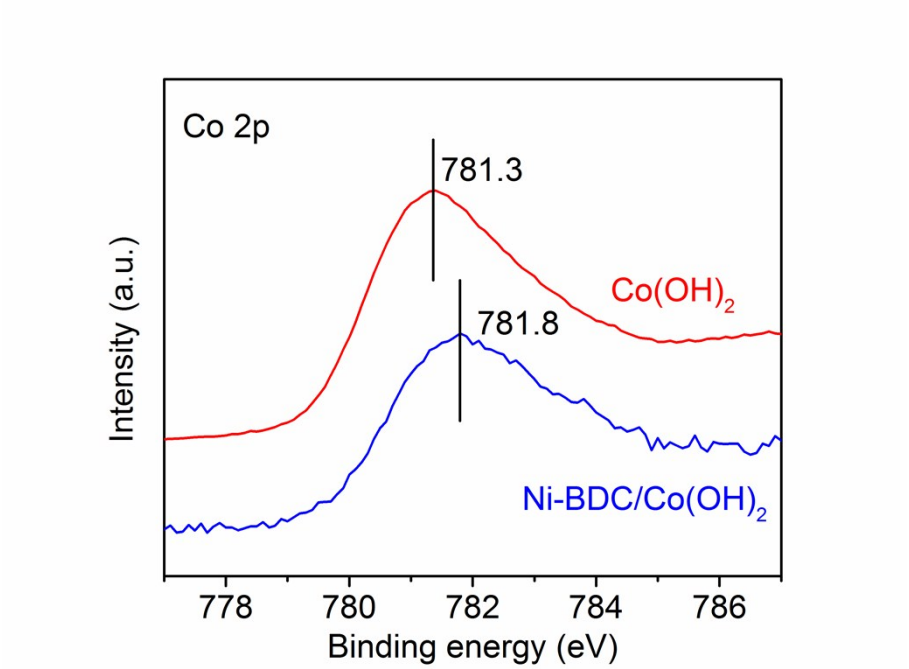


**Figure S8.** Magnified XPS spectra of Co $2p_{3/2}$ for $Co(OH)_2$ and Ni-BDC/$Co(OH)_2$.

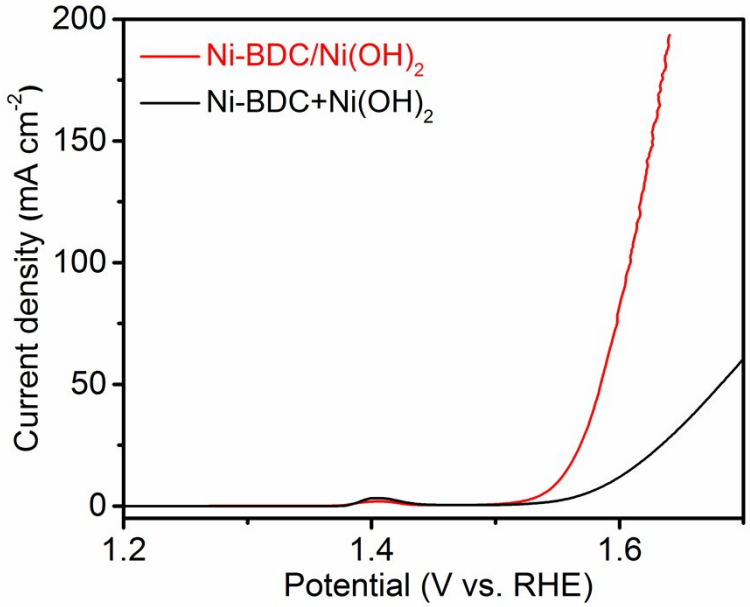


**Figure S9.** LSV curves for Ni-BDC/$Ni(OH)_2$ and physically mixed Ni-BDC+$Ni(OH)_2$ sample in 1.0 M KOH at a scan rate of 1 mV $s^{-1}$.

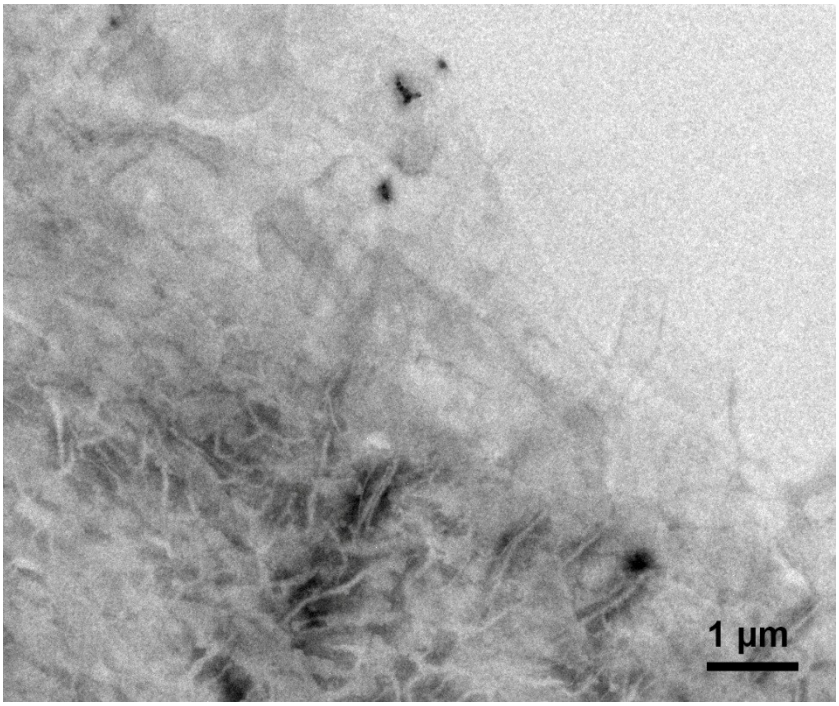


**Figure S10.** TEM image of Ni-BDC/$Ni(OH)_2$ sample after OER test.

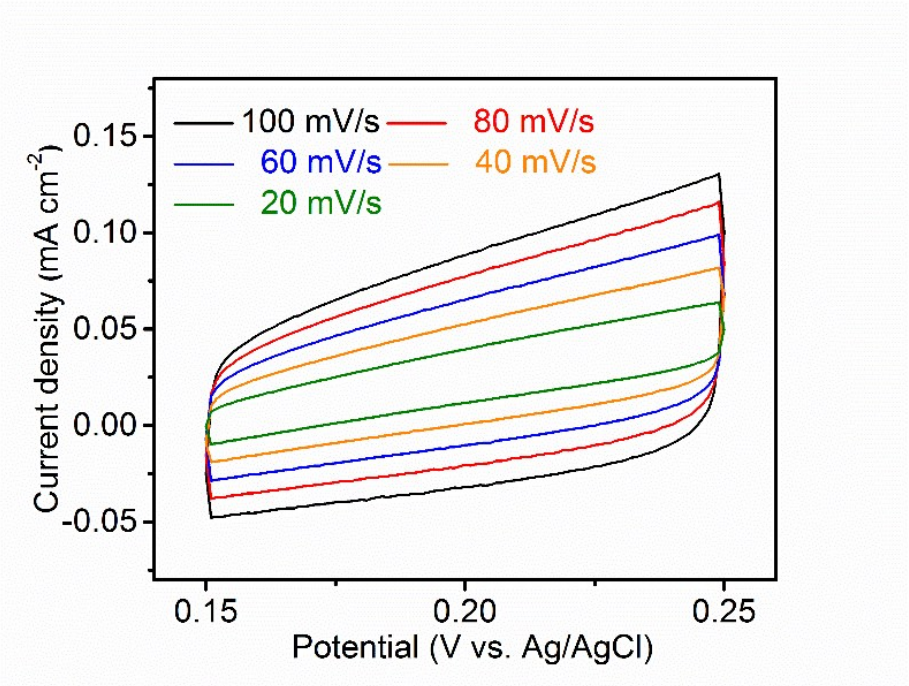


**Figure S11.** CV curves of $Ni(OH)_2$ at scan rates from 20 to 100 mV $s^{-1}$.

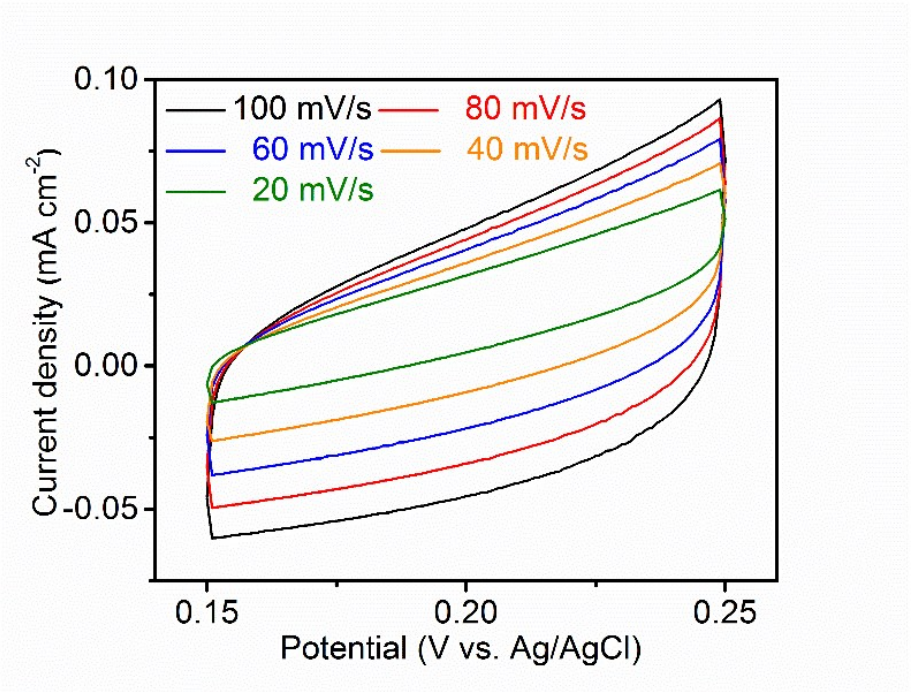


**Figure S12.** CV curves of Ni-BDC at scan rates from 20 to 100 mV $s^{-1}$.

**Table S1.** Comparison of OER activity between MOF-based or $Ni(OH)_2$-based catalysts deposited on glassy carbon or FTO in alkaline solution.

| **Catalyst** | **Overpotential @ 10 mA cm$^{-2}$ (mV)** | **Current density @ 1.6V (vs. RHE) (mA cm$^{-2}$)** | **Tafel slop (mV dec$^{-1}$)** | **Reference** |
|---|---|---|---|---|
| Ni-BDC/$Ni(OH)_2$ | 320 | 82.5 | 41 | This work |
| Ni-BDC | 358 | 15.1 | 57 | This work |
| $Ni(OH)_2$ | 395 | 4.0 | 60 | This work |
| NNU-23 ($Fe_2Ni$-MOF) | 365 | 11 | 81.8 | Angew. Chem. Int. Ed. **2018**, 57, 9660. |
| $Co_2(\mu\text{-}OH)_2(bbta)$ | 387 | 6 | N/A | J. Am. Chem. Soc. **2016**, 138, 8336. |
| $Ti_3C_2T_x$–CoBDC nanosheets | 410 | 4 | 48.2 | ACS Nano **2017**, 11, 5800. |
| Co-BDC nanosheets | 371 | 10 | 103 | Nat. Energy, **2016**, 1, 16184. |
| Ni-BDC nanosheets | 321 | 20 | 65 | Nat. Energy, **2016**, 1, 16184. |
| NiPc–MOF | 390 | 8 | 74 | J. Mater. Chem. A, **2018**, 6, 1188. |
| β-$Ni(OH)_2$ nanomeshes | NA | 100 | 132 | Adv. Mater. **2017**, 29, 1604765 |
| $Ni_{0.75}V_{0.25}$-LDH nanosheets | 318 | 27 (1.58V vs. RHE) | 50 | Nat. Commun. **2016**, 7, 11981. |
| NiFe LDH/graphene | 337 | 20 | 45 | Adv. Mater. **2015**, 27, 4516. |
| α-$Ni(OH)_2$ hollow spheres | 331 | 45 | 42 | J. Am. Chem. Soc. **2014**, 136, 7077. |
| MWCNTs/$Ni(OH)_2$ | 474 | < 5 | 87 | J. Mater. Chem. A, **2014**, 2, 11799. |
| CoMn LDH nanoplates | 324 | NA | 43 | J. Am. Chem. Soc. **2014**, 136, 16481 |