Supplementary Information

Three-Dimensional Porous MoNi₄ Networks Constructed by Nanosheets as Bifunctional Electrocatalysts for Overall Water Splitting

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Table S1. HER, OER and overall water splitting activities of the porous MoNi<sub>4</sub> networks and reported catalysts.

<table>
<thead>
<tr>
<th>Catalyst (mass loading)</th>
<th>Electrolyte</th>
<th>HER Potential vs. RHE (V) @ 10 mA cm&lt;sup&gt;-2&lt;/sup&gt;</th>
<th>OER Potential vs. RHE (V) @ 10 mA cm&lt;sup&gt;-2&lt;/sup&gt;</th>
<th>Overall Water Splitting Potential (V) @ 10 mA cm&lt;sup&gt;-2&lt;/sup&gt;</th>
<th>Overall Water Splitting Onset Potential (V)</th>
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<tr>
<td>porous MoNi&lt;sub&gt;4&lt;/sub&gt; networks (~1.09 mg/cm&lt;sup&gt;2&lt;/sup&gt;)</td>
<td>1 M KOH</td>
<td>-0.028</td>
<td>1.51</td>
<td>1.58</td>
<td>1.45&lt;sup&gt;this work&lt;/sup&gt;</td>
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<td>NiCo&lt;sub&gt;2&lt;/sub&gt;O&lt;sub&gt;4&lt;/sub&gt; hollow microcuboids&lt;sup&gt;[1]&lt;/sup&gt; (~1 mg/cm&lt;sup&gt;2&lt;/sup&gt;)</td>
<td>1 M KOH</td>
<td>-0.110</td>
<td>1.52</td>
<td>1.65</td>
<td></td>
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<tr>
<td>NiSe nanowire film/Ni foam&lt;sup&gt;[2]&lt;/sup&gt; (2.8 mg/cm&lt;sup&gt;2&lt;/sup&gt;)</td>
<td>1 M KOH</td>
<td>-0.096</td>
<td></td>
<td>1.63</td>
<td>~1.5</td>
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<td>Ni&lt;sub&gt;3&lt;/sub&gt;P&lt;sub&gt;4&lt;/sub&gt; Films/Ni foil&lt;sup&gt;[3]&lt;/sup&gt; (~3.5 mg/cm&lt;sup&gt;2&lt;/sup&gt;)</td>
<td>1 M KOH</td>
<td>-0.15</td>
<td>1.56</td>
<td>&lt;1.7</td>
<td>~1.53</td>
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<td>Ni&lt;sub&gt;2&lt;/sub&gt;P nanoparticles&lt;sup&gt;[4]&lt;/sup&gt; (0.14 mg/cm&lt;sup&gt;2&lt;/sup&gt;)</td>
<td>1 M KOH</td>
<td></td>
<td>1.52</td>
<td>1.63</td>
<td>~1.5</td>
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<tr>
<td>Co-P films/Cu foil&lt;sup&gt;[5]&lt;/sup&gt; (2.71 mg/cm&lt;sup&gt;2&lt;/sup&gt;)</td>
<td>1 M KOH</td>
<td>-0.094</td>
<td>1.575</td>
<td>1.64</td>
<td>1.57</td>
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<td>Porous Cobalt-Based Thin Film&lt;sup&gt;[6]&lt;/sup&gt; (~0.1 mg/cm&lt;sup&gt;2&lt;/sup&gt;)</td>
<td>1 M KOH</td>
<td>-0.38</td>
<td></td>
<td>1.53</td>
<td></td>
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<td></td>
<td>0.5 M H&lt;sub&gt;2&lt;/sub&gt;SO&lt;sub&gt;4&lt;/sub&gt;</td>
<td></td>
<td>-0.15</td>
<td></td>
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<td>Ni&lt;sub&gt;3&lt;/sub&gt;S&lt;sub&gt;2&lt;/sub&gt; Nanosheet Arrays /Ni foam&lt;sup&gt;[7]&lt;/sup&gt; (~1.6 mg/cm&lt;sup&gt;2&lt;/sup&gt;)</td>
<td>1 M KOH</td>
<td>~0.22</td>
<td>~1.49</td>
<td>~1.76(@ ~13 mA cm&lt;sup&gt;-2&lt;/sup&gt;)</td>
<td></td>
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<tr>
<td></td>
<td>neutral media</td>
<td></td>
<td>~0.17</td>
<td></td>
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<tr>
<td>NiFeOₓ/CFP after two lithium galvanostatic cycles[8] (−1.6 mg/cm²)</td>
<td>1M KOH</td>
<td>-0.088</td>
<td>1.46</td>
<td>1.55</td>
<td>~1.5</td>
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<td>-</td>
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<td>-</td>
<td>-</td>
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<td>-</td>
<td>1.51 (increasing the mass loading)</td>
<td>-</td>
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</table>
**Figure S1.** SEM images of (a) nickel foam, (b) porous MoNi$_4$ networks annealed at 450°C at low magnification.

**Figure S2.** XRD patterns of nickel foam.
Figure S3. SEM images of Mo-Ni based precursors at different magnifications.

Figure S4. TEM images of Mo-Ni based precursors at different magnifications.
**Figure S5.** XRD patterns of the porous MoNi₄ networks annealed at 300°C, 450°C and 600°C.

**Figure S6.** Higher resolution SEM image of the porous MoNi₄ networks annealed at 450°C.

**Figure S7.** The corresponding energy dispersive X-ray (EDX) spectrum of porous MoNi₄ networks annealed at 450°C.
Figure S8. SEM images of porous MoNi₄ networks annealed at 300°C at different magnifications.

Figure S9. TEM images of porous MoNi₄ networks annealed at 300°C at different magnifications.
**Figure S10.** SEM images of porous MoNi$_4$ networks annealed at 600°C at different magnifications.

**Figure S11.** TEM images of porous MoNi$_4$ networks annealed at 600°C at different magnifications.
Figure S12. XPS spectra of the MoNi₄ networks annealed at different temperature. (a) Ni 2p peaks. (b) Mo 3d peaks.
Figure S13. SEM images of porous MoNi$_4$ networks annealed at 450°C after the HER stability test.

Figure S14. (a) TEM images and (b) the corresponding selected-area electron diffraction (SAED) pattern of porous MoNi$_4$ networks annealed at 450°C after the HER stability test.
**Figure S15.** SEM images of porous MoNi$_4$ networks annealed at 450°C after the OER stability test.

**Figure S16.** (a-c) TEM images and (d) the corresponding selected-area electron diffraction (SAED) pattern of the porous MoNi$_4$ networks annealed at 450°C after the OER stability test.
**Figure S17.** Dissolved quantity - time curves of Mo and Ni during OER.

**Figure S18.** XPS spectra (Ni 2p peaks) of the MoNi₄ networks annealed at different temperature after OER.
**Figure S19.** Electrochemically surface area measurements. The corresponding $j_{\text{geo}}$ vs scan rates plots of the porous MoNi$_4$ networks annealed at 450°C.

**Figure S20.** The Nyquist plots of the porous MoNi$_4$ networks annealed at 450°C at (a) -100 mV vs. RHE and (b) 1.6 V vs. RHE. All of the potentials and voltages are without iR corrected.
References


