Electronic Supplementary Information for

Rough Ni@MoN coral for the hydrogen evolution reaction in acidic and alkaline media

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Fig. S1. XRD patterns of (a) NiMo nanorod, (b) Ni@MoN-650/700/750, (c) Ni-700 and Mo-700.

Fig. S2. SEM images of (a) NiMoO$_4$ nanorod, (b) Ni-700, (c) Mo-700.

Fig. S3. Raman of Ni@MoN-650/700/750.
Fig. S4. XPS spectra of the Ni@MoN-650 catalysts: (a) survey, (b) C 1s, (c) N 1s, (d) O 1s, (e) Ni 2p and (f) Mo 3d.

Fig. S5. XPS spectra of the Ni@MoN-750 catalysts: (a) survey, (b) C 1s, (c) N 1s, (d) O 1s, (e) Ni 2p and (f) Mo 3d.
**Fig. S6.** XPS spectra of the Ni-700 catalysts: (a) survey, (b) C 1s, (c) N 1s, (d) O 1s and (e) Ni 2p.

**Fig. S7.** XPS spectra of the Mo-700 catalysts: (a) survey, (b) C 1s, (c) N 1s, (d) O 1s and (e) Mo 3d.
Fig. S8. XPS of Ni@MoN-700 after argon plasma etching, Ni 2p.

Fig. S9. (a) CV curves of Ni@MoN-650 electrode, (b) CV curves of Ni@MoN-700 electrode, (c) CV curves of Ni@MoN-750 electrode, (d) CV curves of Ni-700 electrode, (e) CV curves of Mo-700 electrode, in 1 M KOH.
Fig. S10. (a) The HER-TOFs of the Ni@MoN-650/700/750 in 1 M KOH solution (b) The HER-TOFs of the Ni@MoN-650/700/750 in 0.5 M H$_2$SO$_4$ solution.

Fig. S11. (a) CV curves of Ni@MoN-650 electrode, (b) CV curves of Ni@MoN-700 electrode, (c) CV curves of Ni@MoN-750 electrode, (c) CV curves of Ni-700 electrode, (c) CV curves of Mo-700 electrode, in 0.5 M H$_2$SO$_4$.

Fig. S12. Contact angles of (a) Ni@MoN-650, (b) Ni@MoN-700 and (c) Ni@MoN-750.
Fig. S13. XPS spectra of the Ni@MoN-700 catalysts after long-term electrolysis in 1M KOH: (a) survey, (b) C 1s, (c) N 1s, (d) O 1s, (e) Ni 2p and (f) Mo 3d.

Fig. S14. XPS spectra of the Ni@MoN-700 catalysts after long-term electrolysis in 0.5 M H₂SO₄: (a) survey, (b) C 1s, (c) N 1s, (d) O 1s, (e) Ni 2p and (f) Mo 3d.
**Table S1.** The concentration of oxygen vacancies for different samples as calculated form the O 1s XPS spectra.

<table>
<thead>
<tr>
<th>Catalyst</th>
<th>$S_{O-V}/S_{total	ext{ other peak}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni@MoN-650</td>
<td>0.43</td>
</tr>
<tr>
<td>Ni@MoN-700</td>
<td>0.54</td>
</tr>
<tr>
<td>Ni@MoN-750</td>
<td>0.35</td>
</tr>
<tr>
<td>Ni-700</td>
<td>0.43</td>
</tr>
<tr>
<td>Mo-700</td>
<td>0.37</td>
</tr>
<tr>
<td>KOH-after</td>
<td>0.41</td>
</tr>
<tr>
<td>H$_2$SO$_4$-after</td>
<td>0.16</td>
</tr>
</tbody>
</table>

**Table S2.** Comparison of the RCT values of the prepared materials.

<table>
<thead>
<tr>
<th>Sample</th>
<th>RCT (Ohm)-KOH</th>
<th>RCT (Ohm)-H$_2$SO$_4$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni@MoN-650</td>
<td>1.64</td>
<td>1.62</td>
</tr>
<tr>
<td>Ni@MoN-700</td>
<td>1.43</td>
<td>1.40</td>
</tr>
<tr>
<td>Ni@MoN-750</td>
<td>1.58</td>
<td>1.56</td>
</tr>
<tr>
<td>Ni-700</td>
<td>1.71</td>
<td>1.65</td>
</tr>
<tr>
<td>Mo-700</td>
<td>1.74</td>
<td>1.68</td>
</tr>
</tbody>
</table>
**Table S3.** Comparison of the catalytic activities of HER on Ni@MoN-700 with recently reported catalysts in the 1.0 mol·L$^{-1}$ KOH medium

<table>
<thead>
<tr>
<th>Catalyst</th>
<th>$\eta$/mV vs. RHE</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni@MoN-700</td>
<td>30</td>
<td>This work</td>
</tr>
<tr>
<td>Ni/NiO-cp</td>
<td>124</td>
<td>[2]</td>
</tr>
<tr>
<td>W-MoP</td>
<td>71</td>
<td>[7]</td>
</tr>
<tr>
<td>Mo-N/Mo-C</td>
<td>135</td>
<td>[8]</td>
</tr>
<tr>
<td>Mo–N/C@MoS$_2$</td>
<td>117</td>
<td>[9]</td>
</tr>
<tr>
<td>MoS$_2$/MoN</td>
<td>132</td>
<td>[10]</td>
</tr>
<tr>
<td>NiSA-MoS$_2$</td>
<td>98</td>
<td>[17]</td>
</tr>
<tr>
<td>Ni$_3$N</td>
<td>74</td>
<td>[24]</td>
</tr>
<tr>
<td>NiO/Ni-CNT</td>
<td>80</td>
<td>[25]</td>
</tr>
<tr>
<td>NiCoDPA</td>
<td>112</td>
<td>[41]</td>
</tr>
</tbody>
</table>

Note: $\eta$ is the overpotential measured at 10 mA·cm$^{-2}$

**Table S4.** Comparison of the catalytic activities of HER on Ni@MoN-700 with recently reported catalysts in the 0.5 mol·L$^{-1}$ H$_2$SO$_4$ medium

<table>
<thead>
<tr>
<th>Catalyst</th>
<th>$\eta$/mV vs. RHE</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni@MoN-700</td>
<td>76</td>
<td>This work</td>
</tr>
<tr>
<td>Co–N–V$_5$S$_4$</td>
<td>268</td>
<td>[1]</td>
</tr>
<tr>
<td>MoS$_2$/MoN</td>
<td>117</td>
<td>[10]</td>
</tr>
<tr>
<td>VN/Mo$_2$C</td>
<td>140</td>
<td>[15]</td>
</tr>
<tr>
<td>NiSA-MoS$_2$</td>
<td>110</td>
<td>[17]</td>
</tr>
<tr>
<td>PCN@MoS$_2$@C</td>
<td>130</td>
<td>[19]</td>
</tr>
<tr>
<td>Cu electrode</td>
<td>182</td>
<td>[39]</td>
</tr>
<tr>
<td>CoP$_3$/CoMoP</td>
<td>125</td>
<td>[40]</td>
</tr>
<tr>
<td>CoMoOF/GF</td>
<td>94</td>
<td>[42]</td>
</tr>
</tbody>
</table>

Note: $\eta$ is the overpotential measured at 10 mA·cm$^{-2}$. 