Binder-free growth of nickel-doped iron sulfide on nickel foam via electrochemical deposition for electrocatalytic water splitting

Subhasis Shit, †,‡ Saikat Bolar, †,‡ Naresh Chandra Murmu, †,‡ and Tapas Kuila†,‡,*

† Surface Engineering & Tribology Division, Council of Scientific and Industrial Research-Central Mechanical Engineering Research Institute, Durgapur -713209, India

‡ Academy of Scientific and Innovative Research (AcSIR), Ghaziabad- 201002, India

*Correspondence to Tapas Kuila. Contact No.: +91-9647205077; E-mail: tkuila@gmail.com; t_kuila@cmeri.res.in.

Contents:

Number of pages: 31
Number of figures: 23
Number of tables: 8
Figure S1. Chronoamperometric curves for electrodeposition at (a) pH ≈ 3.5, (b-d) pH ≈ 4 and (e) pH ≈ 4.5.
Figure S2. (a) FE-SEM image and (b) HR-TEM image of FS-0.9-30.

Figure S3. HR-TEM images of (a, b) FS-0.9-30 and (c, d) FS-0.9-45.
Figure S4. XPS survey spectra of (a) FS-0.9-15, (b) FS-0.9-30 and (c) FS-0.9-45; (d) XPS Pt 4f spectra of FS-0.9-15, FS-0.9-30 and FS-0.9-45.
Figure S5. Deconvoluted Fe 2p3/2 peaks of (a) FS-0.9-15, (b) FS-0.9-30 and (c) FS-0.9-45 after considering hyperfine slitting.

- The Fe$^{2+}$ pre-peak is designated as D, E and D in Figure S4a, S4b and S4c, respectively.
- The peaks related to hyperfine splitting of Fe$^{2+}$ are designated as:
  - A, E and C in Figure S4a
  - G and C in Figure S4b
  - E and C in Figure S4c.
- The peaks related to hyperfine splitting of Fe$^{2+}$ are designated as:
  - B, F and G in Figure S4a
  - F, A, B and D in Figure S4b
  - B, F, A and G in Figure S4c.
Figure S6. (a) XPS Ni 2p spectra of FS-0.9-15, FS-0.9-30 and FS-0.9-45; (b) VBXPS profile of FS-0.9-30 and FS-0.9-45.

Table S1. Comparison of recently reported Fe-S based HER electrocatalysts.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Electrolyte</th>
<th>Current density (J, mA cm⁻²)</th>
<th>Overpotential @ Corresponding J (mV)</th>
<th>Tafel slope (mV dec⁻¹)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mesoporous FeS₂</td>
<td>0.1 M KOH</td>
<td>10</td>
<td>96</td>
<td>78</td>
<td>[1]</td>
</tr>
<tr>
<td>FeS₂/C/NF</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>202</td>
<td>98</td>
<td>[2]</td>
</tr>
<tr>
<td>Co-doped FeS₂</td>
<td>0.5 M H₂SO₄</td>
<td>10</td>
<td>166</td>
<td>51</td>
<td>[3]</td>
</tr>
<tr>
<td>Fe@FeOₓSᵧ</td>
<td>1.0 M KOH</td>
<td>100</td>
<td>243</td>
<td>77</td>
<td>[4]</td>
</tr>
<tr>
<td>FeS₂</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>nanoparticle@rGO</td>
<td>0.5 M H₂SO₄</td>
<td>10</td>
<td>139</td>
<td>66</td>
<td>[5]</td>
</tr>
<tr>
<td>FeS₂–RGO</td>
<td>0.5 M H₂SO₄</td>
<td>10</td>
<td>226</td>
<td>61</td>
<td>[6]</td>
</tr>
<tr>
<td>FeNi₀.₂₀S₂–rGO</td>
<td>0.5 M H₂SO₄</td>
<td>10</td>
<td>183</td>
<td>78.63</td>
<td>[7]</td>
</tr>
<tr>
<td>FeS₂-doped MoS₂</td>
<td>0.5 M H₂SO₄</td>
<td>10</td>
<td>136</td>
<td>82</td>
<td>[8]</td>
</tr>
<tr>
<td>Fe₃S₄</td>
<td>1.0 M KOH</td>
<td>50</td>
<td>279</td>
<td>87.40</td>
<td>[9]</td>
</tr>
<tr>
<td>Fe₀.₉Co₀.₁S₂/CNT</td>
<td>0.5 M H₂SO₄</td>
<td>20</td>
<td>120</td>
<td>46</td>
<td>[10]</td>
</tr>
<tr>
<td>FS-0.9-30</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>157</td>
<td>112.51</td>
<td>This</td>
</tr>
<tr>
<td>FS-0.9-30</td>
<td>10</td>
<td></td>
<td>187</td>
<td>160.75</td>
<td>Work</td>
</tr>
</tbody>
</table>
**Table S2.** Comparison of recently reported HER electrocatalysts based on sulfide of metals other than Fe.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Electrolyte</th>
<th>Current density (J, mA cm(^{-2}))</th>
<th>Overpotential @ Corresponding J (mV)</th>
<th>Tafel slope (mV dec(^{-1}))</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>CoNi$_2$S$_4$@CoS$_2$/NF</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>173</td>
<td>45</td>
<td>[11]</td>
</tr>
<tr>
<td>NiCo$_2$S$_4$</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>282</td>
<td>-</td>
<td>[12]</td>
</tr>
<tr>
<td>Co$_9$S$_8$</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>217</td>
<td>110</td>
<td>[13]</td>
</tr>
<tr>
<td>N-Ni$_3$S$_2$ NWs/NF</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>105</td>
<td>108</td>
<td>[14]</td>
</tr>
<tr>
<td>CoS$_2$ HNSs</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>193</td>
<td>100</td>
<td>[15]</td>
</tr>
<tr>
<td>Co$_3$S$_4$-L</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>270</td>
<td>124.5</td>
<td>[16]</td>
</tr>
<tr>
<td>Co(OH)$_2$/Ni-Co-S-8h</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>148</td>
<td>88</td>
<td>[17]</td>
</tr>
<tr>
<td>Ni–S–B</td>
<td>30 wt% KOH</td>
<td>10</td>
<td>240</td>
<td>121.2</td>
<td>[18]</td>
</tr>
<tr>
<td>N-Ni$_3$S$_2$@C/NF</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>113</td>
<td>90</td>
<td>[19]</td>
</tr>
<tr>
<td>NiS$_2$ nanospheres</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>147</td>
<td>105</td>
<td>[20]</td>
</tr>
</tbody>
</table>
Figure S7. Tafel plots for (a) FS-0.8-15, (b) FS-0.9-15, (c) FS-1.0-15, (d) FS-0.8-30, (e) FS-0.9-30, (f) FS-1.0-30 and (g) FS-0.9-45.
**Figure S8.** Tafel plots for (a) Pt/C (10 wt%) and NF; (b) Tafel plots near 0 V vs. RHE for FS-0.8-15, FS-0.9-15, FS-1.0-15, FS-0.8-30, FS-0.9-30, FS-1.0-30 and FS-0.9-45.
Figure S9. Experimental and Kramers-Kronig test fitted Nyquist plots (HER) for (a) FS-0.8-15, (b) FS-0.9-15, (c) FS-1.0-15, (d) FS-0.8-30, (e) FS-0.9-30, (f) FS-1.0-30 and (g) FS-0.9-45.
Figure S10. Bode phase angle plots (HER) at different overpotential for (a) FS-0.8-15, (b) FS-0.9-15, (c) FS-1.0-15, (d) FS-0.8-30, (e) FS-0.9-30, (f) FS-1.0-30 and (g) FS-0.9-45.
Figure S11. Equivalent circuits used to fit Nyquist plots for HER.

Table S3. Data obtained after fitting Nyquist plots for HER.

<table>
<thead>
<tr>
<th>Sample</th>
<th>( R_u ) (Ω)</th>
<th>CPE(_1) (F cm(^{-2}))</th>
<th>( R_f ) (Ω)</th>
<th>CPE(_2) (F cm(^{-2}))</th>
<th>( R_p ) (Ω)</th>
<th>CPE(_3) (F cm(^{-2}))</th>
<th>( R_{CT} ) (Ω)</th>
</tr>
</thead>
<tbody>
<tr>
<td>FS-0.8-15</td>
<td>7.44</td>
<td>0.006596</td>
<td>0.40</td>
<td>-</td>
<td>-</td>
<td>0.010646</td>
<td>265.0</td>
</tr>
<tr>
<td>FS-0.9-15</td>
<td>7.40</td>
<td>0.004855</td>
<td>0.15</td>
<td>0.010731</td>
<td>210</td>
<td>0.006197</td>
<td>95.0</td>
</tr>
<tr>
<td>FS-1.0-15</td>
<td>6.41</td>
<td>0.005183</td>
<td>0.21</td>
<td>0.063838</td>
<td>90</td>
<td>0.009386</td>
<td>180.0</td>
</tr>
<tr>
<td>FS-0.8-30</td>
<td>9.04</td>
<td>0.001928</td>
<td>0.60</td>
<td>-</td>
<td>-</td>
<td>0.006919</td>
<td>283.5</td>
</tr>
<tr>
<td>FS-0.9-30</td>
<td>7.27</td>
<td>0.015822</td>
<td>0.10</td>
<td>-</td>
<td>-</td>
<td>0.030146</td>
<td>220.0</td>
</tr>
<tr>
<td>FS-1.0-30</td>
<td>5.39</td>
<td>0.009897</td>
<td>0.30</td>
<td>-</td>
<td>-</td>
<td>0.025600</td>
<td>402.0</td>
</tr>
<tr>
<td>FS-0.9-45</td>
<td>5.29</td>
<td>0.083888</td>
<td>0.20</td>
<td>-</td>
<td>-</td>
<td>1.651758</td>
<td>380.0</td>
</tr>
</tbody>
</table>
Figure S12. Fitted Nyquist plots (HER) for (a) FS-0.8-15, (b) FS-0.9-15, (c) FS-1.0-15, (d) FS-0.8-30, (e) FS-0.9-30, (f) FS-1.0-30 and (g) FS-0.9-45.
Table S4. Comparison of recently reported Fe-S based OER electrocatalysts.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Electrolyte</th>
<th>Current density (J, mA cm(^{-2}))</th>
<th>Overpotential @ Corresponding J (mV)</th>
<th>Tafel slope (mV dec(^{-1}))</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>FeS(_2)/C/NF</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>240</td>
<td>92</td>
<td>[2]</td>
</tr>
<tr>
<td>Fe@FeO(_x)S(_y)</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>238</td>
<td>82.70</td>
<td>[4]</td>
</tr>
<tr>
<td>Fe(_3)S(_4)</td>
<td>1.0 M KOH</td>
<td>50</td>
<td>255</td>
<td>46</td>
<td>[9]</td>
</tr>
<tr>
<td>FeNiS(_2) ultrathin</td>
<td>0.1 M KOH</td>
<td>10</td>
<td>310</td>
<td>46</td>
<td>[21]</td>
</tr>
<tr>
<td>FS-0.9-30</td>
<td>1.0 M KOH</td>
<td>100</td>
<td>255</td>
<td>58.29</td>
<td>This Work</td>
</tr>
</tbody>
</table>

Table S5. Comparison of recently reported OER electrocatalysts based on sulfide of metals other than Fe.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Electrolyte</th>
<th>Current density (J, mA cm(^{-2}))</th>
<th>Overpotential @ Corresponding J (mV)</th>
<th>Tafel slope (mV dec(^{-1}))</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Co(_9)S(_8)</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>299</td>
<td>67</td>
<td>[13]</td>
</tr>
<tr>
<td>CoS(_2) HNSs</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>290</td>
<td>57</td>
<td>[15]</td>
</tr>
<tr>
<td>Co(_3)S(_4)-L</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>350</td>
<td>84.7</td>
<td>[16]</td>
</tr>
<tr>
<td>Co(OH)(_2)/Ni-Co-S-8h</td>
<td>1.0 M KOH</td>
<td>30</td>
<td>300</td>
<td>64</td>
<td>[17]</td>
</tr>
<tr>
<td>N-Ni(_3)S(_2)@C/NF</td>
<td>1.0 M KOH</td>
<td>100</td>
<td>310</td>
<td>75</td>
<td>[19]</td>
</tr>
<tr>
<td>NiS(_2) nanospheres</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>271</td>
<td>65</td>
<td>[20]</td>
</tr>
<tr>
<td>Ni/NiS/NC</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>337</td>
<td>52</td>
<td>[22]</td>
</tr>
<tr>
<td>CoS film</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>300</td>
<td>57</td>
<td>[23]</td>
</tr>
<tr>
<td>CoS/CNT</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>330</td>
<td>142</td>
<td>[24]</td>
</tr>
</tbody>
</table>
Figure S13. FE-SEM images of (a) post-HER FS-0.9-30 and (b) post-HER FS-0.9-45; (c) XRD patterns of FS-0.9-30, post-HER FS-0.9-30, FS-0.9-45 and post-HER FS-0.9-45.

- **Subdividing the OER pathway proposed by Conway and Bourgault**

The OER proceeds via peroxide linkage formation pathway suggested by Conway and Bourgault. In this pathway the mechanistic steps involve both adsorption of oxygen-containing species and charge transfer at the electrode-electrolyte interface. However, Fletcher argued that this hypothesizes simultaneous movement of nucleus and electron which violates the Frank-Condon principle. Therefore, he suggested breaking down each step into chemical and electrochemical steps. In chemical steps the adsorption-desorption or chemical changes will occur and in the electrochemical steps the charge transfer will occur. Conway-Bourgault
pathway, therefore, can be subdivided into following seven steps following the suggestion proposed by Fletcher.

\[
\begin{align*}
OH^- & \leftrightarrow OH_{ads} & (S1) \\
OH_{ads} & \rightarrow OH_{ads} + e^- & (S2) \\
OH_{ads} + OH_{ads} & \leftrightarrow OH_{ads}OH_{ads} & (S3) \\
OH_{ads}OH_{ads} & \rightarrow OHOH_{ads} + e^- & (S4) \\
OHOOH_{ads} & \rightarrow O_{ads} + H_2O & (S5) \\
O_{ads} + OH_{ads} & \leftrightarrow O_2H_{ads} & (S6) \\
O_2H_{ads} + OH_{ads} & \rightarrow H_2O + O_2 & (S7)
\end{align*}
\]

**Figure S14.** Tafel plots for RuO$_2$ and NF.
Figure S15. Experimental and Kramers-Kronig test fitted Nyquist plots (OER) for (a) FS-0.8-15, (b) FS-0.9-15, (c) FS-1.0-15, (d) FS-0.8-30, (e) FS-0.9-30, (f) FS-1.0-30 and (g) FS-0.9-45.
Figure S16. Bode phase angle plots (OER) at different overpotential for (a) FS-0.8-15, (b) FS-0.9-15, (c) FS-1.0-15, (d) FS-0.8-30, (e) FS-0.9-30, (f) FS-1.0-30 and (g) FS-0.9-45.
Figure S17. Equivalent circuits used to fit Nyquist plots for OER.

Table S6. Data obtained after fitting Nyquist plots for OER.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$R_u$ (Ω)</th>
<th>CPE$_1$ (F cm$^{-2}$)</th>
<th>$R_f$ (Ω)</th>
<th>CPE$_2$ (F cm$^{-2}$)</th>
<th>$R_{CT}$ (Ω)</th>
</tr>
</thead>
<tbody>
<tr>
<td>FS-0.8-15</td>
<td>6.25</td>
<td>0.000805</td>
<td>0.60</td>
<td>0.007159</td>
<td>29.5</td>
</tr>
<tr>
<td>FS-0.9-15</td>
<td>5.88</td>
<td>0.003412</td>
<td>0.35</td>
<td>0.009655</td>
<td>22.2</td>
</tr>
<tr>
<td>FS-1.0-15</td>
<td>7.75</td>
<td>0.000659</td>
<td>0.55</td>
<td>0.004004</td>
<td>41.5</td>
</tr>
<tr>
<td>FS-0.8-30</td>
<td>6.64</td>
<td>0.002322</td>
<td>0.70</td>
<td>0.008485</td>
<td>27.7</td>
</tr>
<tr>
<td>FS-0.9-30</td>
<td>4.81</td>
<td>0.010610</td>
<td>0.19</td>
<td>0.012065</td>
<td>17.7</td>
</tr>
<tr>
<td>FS-1.0-30</td>
<td>6.00</td>
<td>0.001842</td>
<td>8.00</td>
<td>0.002344</td>
<td>73.2</td>
</tr>
<tr>
<td>FS-0.9-45</td>
<td>5.12</td>
<td>0.016030</td>
<td>0.40</td>
<td>0.185597</td>
<td>17.8</td>
</tr>
</tbody>
</table>
Figure S18. Fitted Nyquist plots (OER) for (a) FS-0.8-15, (b) FS-0.9-15, (c) FS-1.0-15, (d) FS-0.8-30, (e) FS-0.9-30, (f) FS-1.0-30 and (g) FS-0.9-45.
Figure S19. Mott-Schottky (MS) plots for (a) FS-0.8-15, (b) FS-0.9-15, (c) FS-1.0-15, (d) FS-0.8-30, (e) FS-0.9-30, (f) FS-1.0-30 and (g) FS-0.9-45.
Figure S20. Cyclic voltammograms obtained at different scan rates for (a) FS-0.8-15, (b) FS-0.9-15, (c) FS-1.0-15, (d) FS-0.8-30, (e) FS-0.9-30, (f) FS-1.0-30 and (g) FS-0.9-45.
Table S7. The calculated C_{dl} and ECSA values for the electrodeposited materials.

<table>
<thead>
<tr>
<th>Materials</th>
<th>From ΔJ vs. ν plot</th>
<th>From ΔJν^{-1/2} vs. ν^{1/2} plot</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>C_{dl} (mF cm^{-2})</td>
<td>ECSA (cm^{2})</td>
</tr>
<tr>
<td>FS-0.8-15</td>
<td>4.54</td>
<td>113.50</td>
</tr>
<tr>
<td>FS-0.9-15</td>
<td>8.00</td>
<td>200.00</td>
</tr>
<tr>
<td>FS-1.0-15</td>
<td>5.49</td>
<td>137.25</td>
</tr>
<tr>
<td>FS-0.8-30</td>
<td>5.67</td>
<td>141.75</td>
</tr>
<tr>
<td>FS-0.9-30</td>
<td>8.65</td>
<td>216.25</td>
</tr>
<tr>
<td>FS-1.0-30</td>
<td>2.86</td>
<td>71.50</td>
</tr>
<tr>
<td>FS-0.9-45</td>
<td>2.38</td>
<td>59.50</td>
</tr>
</tbody>
</table>

Figure S21. FE-SEM images of (a-b) post-OER FS-0.9-30; (c) XRD patterns of FS-0.9-30 and post-OER FS-0.9-30.
Table S8. Comparison of recently reported non-noble metal based overall water splitting electrocatalysts.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Electrolyte</th>
<th>Current density (J, mA cm(^{-2}))</th>
<th>Required potential (V)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>FeS(_2)/C/NF</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>1.72</td>
<td>[2]</td>
</tr>
<tr>
<td>Fe@FeO(_x)S(_y)</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>1.65</td>
<td>[4]</td>
</tr>
<tr>
<td>Fe(_3)S(_4)</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>1.68</td>
<td>[9]</td>
</tr>
<tr>
<td>Co(_9)S(_8)</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>1.66</td>
<td>[13]</td>
</tr>
<tr>
<td>CoS(_2) HNSs</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>1.68</td>
<td>[15]</td>
</tr>
<tr>
<td>Co(_3)S(_4)-L</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>1.63</td>
<td>[16]</td>
</tr>
<tr>
<td>Co(OH)(_2)/Ni-Co-S-8h</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>1.62</td>
<td>[17]</td>
</tr>
<tr>
<td>Ni(_3)S(_2)@C/NF</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>1.57</td>
<td>[19]</td>
</tr>
<tr>
<td>NiS(_2) nanospheres</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>1.66</td>
<td>[20]</td>
</tr>
<tr>
<td>Ni/NiS/NC</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>1.61</td>
<td>[22]</td>
</tr>
<tr>
<td>Ni–Co–P hollow nanobricks</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>1.62</td>
<td>[27]</td>
</tr>
<tr>
<td>(Ni(_x)Fe(_y))(_2)(_P)</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>1.61</td>
<td>[28]</td>
</tr>
<tr>
<td>NFP@NG</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>1.69</td>
<td>[29]</td>
</tr>
<tr>
<td>CuO@Ni/NiFe hydroxides</td>
<td>1.0 M KOH</td>
<td>10 or 40</td>
<td>1.73 or 1.73</td>
<td>[30]</td>
</tr>
<tr>
<td>CoP/NF</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>1.65</td>
<td>[31]</td>
</tr>
<tr>
<td>CoFeZr oxide nanosheet</td>
<td>1.0 M KOH</td>
<td>10</td>
<td>1.63</td>
<td>[32]</td>
</tr>
<tr>
<td>FS-0.9-30</td>
<td></td>
<td></td>
<td>1.727</td>
<td></td>
</tr>
<tr>
<td>FS-0.9-30 (+) II FS-0.9-45</td>
<td>1.0 M KOH</td>
<td>10</td>
<td></td>
<td>This work</td>
</tr>
<tr>
<td>(-) Asymmetric</td>
<td></td>
<td></td>
<td>1.588</td>
<td></td>
</tr>
</tbody>
</table>

- Faradaic efficiency (FE) calculation of the overall water splitting

The current density at 1.8 V potential = 137 mA cm\(^{-2}\)

The calculated geometric area of the electrode = 2.5 cm\(^2\)

Charge passed per second = \((0.137 \times 2.5 \times 1)\) coulomb = 0.3425
0.3425 coulomb = (0.3425/96485) mole of electron

The oxygen produced per second = (0.3425/(96485 × 4)) mole, as 4 electrons are involved for one electron generation

The oxygen produced per second = ((0.3425 × 22400)/(96485 × 4)) mL = 0.0199 mL, as 1 mole gas = 22400 mL gas at STP

Figure S22. (a) Fabricated setup for calculating the Faradaic efficiency of the overall water splitting; (b) chronoamperometric curve for FS-0.9-30 (+) || FS-0.9-45 (-) biased at 1.8 V potential; (c) theoretically calculated and measured amount of the evolved O₂.

An online gas chromatography (GC) was unavailable at present thus an experimental electrolyzer prototype (as shown in Figure S22a) was constructed. The amount of the O₂ evolved was
measured from the water displacement in the collection tube. The area of the electrodes in the setup was kept high so that large amount of gas could evolve. The FE at 1.8 V potential was calculated using the following equation.

\[
FE = \frac{\text{Amount of } O_2 \text{ evolved} \times 100}{\text{Theoretically calculated amount of } O_2}
\]  

(S8)

**Figure S23.** Comparison of (a) HER catalytic activity of Pt/C (10 wt%) and (b) OER catalytic activity of RuO$_2$ after drop-casting with and without polymeric binder.

Both Pt/C and RuO$_2$ showed superior electrocatalytic activity when those were drop casted on the NF without any binder in comparison to when drop casted with binder. However, the electrocatalysts came off from the NF due to the generation of H$_2$/O$_2$ gases and thus the obtained polarization curves were not reliable. This experiment supported that the usage of polymeric binder can suppress the electrocatalytic activity of the electrocatalyst.

**REFERENCES**


[8] Zhao, X.; Ma, X.; Lu, Q.; Li, Q.; Han, C.; Xing, Z.; Yang, X.; FeS2-Doped MoS2 Nanoflower with the Dominant 1T-MoS2 Phase as an Excellent Electrocatalyst For High-


