Supplementary Information

Ir-Ni bimetallic OER Catalysts prepared by controlled Ni Electrodeposition on Ir\textsubscript{poly} and Ir(111)

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Figure S1. Double-layer capacitance measurements displaying cyclic voltammograms of Irpoly measured in a potential window in which no faradic currents occur. Following scan rates were adjusted: 500 mV s\(^{-1}\), 200 mV s\(^{-1}\), 100 mV s\(^{-1}\), 50 mV s\(^{-1}\), 20 mV s\(^{-1}\) and 10 mV s\(^{-1}\).

Table S1. Calculated amount of adsorbed Ni on a polycrystalline Ir electrode in dependence of deposition potential \(E_{\text{dep}}\). The coverage of Ni (ML) was determined from ICP-MS based on the assumption of a predominant (111) face on Irpoly.

<table>
<thead>
<tr>
<th>(E_{\text{dep}} / \text{VRHE})</th>
<th>Ni / nmol</th>
<th>Ni / ML</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.05</td>
<td>3.178</td>
<td>0.36</td>
</tr>
<tr>
<td>0.10</td>
<td>3.157</td>
<td>0.36</td>
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<tr>
<td>0.20</td>
<td>1.857</td>
<td>0.21</td>
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<tr>
<td>0.30</td>
<td>1.018</td>
<td>0.12</td>
</tr>
<tr>
<td>0.40</td>
<td>0.844</td>
<td>0.09</td>
</tr>
<tr>
<td>0.50</td>
<td>0.635</td>
<td>0.07</td>
</tr>
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</table>
Figure S2. X-ray photoelectron spectroscopy measurements of (a) Ir(111) over the entire energy range. (b) High-resolution spectra of Ir(111) displaying the Ir 4f, C 1s, O 1s and Ni 2p spectra. (c) XP spectra of Ni/Irpoly OLt=20 min over the entire energy range.
Figure S3. (a) Atomic force microscopy (AFM) images of Ni/Ir(111) OL with respective height profiles. (b) Magnification of a terrace assembly of (a) and (c) higher magnification. Scale bar is 400 nm. Arrows give the direction of the calculated height profile.

Figure S4. 10th cycle of the electrocatalytic oxygen evolution reaction (OER) activities of Ni-modified Ir electrodes compared to pristine polycrystalline Ir at scan rates of 5 mV s⁻¹ in (a) 0.05 M H₂SO₄ and (b) in 0.1 M KOH of Ir(poly), Ni/Ir(poly) OL and Ni/Ir(poly) NSA. Current densities of the electrodes are normalized to the electrodes’ geometric surface area of 0.785 cm².
Figure S5. Electrocatalytic OER activities of Ni-modified electrocatalysts and pristine Ir_{poly} measured by differential electrochemical mass spectrometry (DEMS) in a dual-thin layer flow cell setup at a scan rate of 10 mV s^{-1}. (a) Faradaic current densities measured at an OER overpotential of 300 mV (1.53 V_{RHE}) normalized to the electrodes' geometric surface area of 0.785 cm^2. The electrocatalytic OER activity in H_2SO_4 was measured directly after the treatment in KOH. (b) Faraday efficiency of oxygen evolution (FEO2) determined for the catalysts.