Electronic Supplementary Information for

Promoting the electrocatalytic property of nickel aerogel by gold decoration for efficient electrocatalytic oxygen evolution in alkali

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Experimental section

1 Chemicals and reagents

Nickel(II) chloride hexahydrate ($NiCl_2 \cdot 6H_2O$, 99.9 %), gold(III) chloride trihydrate ($HAuCl_4 \cdot 3H_2O$, 99.9 %), sodium borohydride ($NaBH_4$) were obtained from Sigma-Aldrich. Potassium hydroxide (KOH, 85 %) and 5 wt % Nafion were obtained from Alfa Aesar. Ethanol (analytical grade) was obtained from Sinopharm Chemical Regent Co. Ltd. Carbon paper (TGP-H-060) was obtained from Toray Co. Ltd., and Ni foam was obtained from Suzhou Taili Materials Technology Co. Ltd. All the solutions were prepared using Milli-Q deionized water with resistance of 18.2 MΩ·cm.

2 Preparation of Ni and Au-Ni aerogel

To prepare Ni aerogel, 0.5 mL of 0.1 M $NiCl_2$ solution was added into 8.5 mL H_2O under stirring for about 2 min, and then 1 mL of freshly prepared 1.5 M $NaBH_4$ was quickly added into the solution. The solution was stirred for 30 seconds to get dark grey solution, and settled in the dark at room temperature for 4 hours. To get 3 % Au NPs modified Ni aerogel, 8.85 μL of 10 wt % $HAuCl_4$ was added after adding $NiCl_2$, and then the same quantity of $NaBH_4$ was added into the solution. The solution was stirred for 2 minutes to get dark grey solution, and settled in the dark at room temperature. After 4 h, fluffy black solids of both two samples settled out of solution down to the bottom of the bottle. After washing thoroughly, the samples were then supercritical-dried to obtain aerogel. To prepare Au-Ni aerogels with different Au mass, 2.95 and 14.75 μL of 10 wt % $HAuCl_4$ were used in the synthetic process with the identical steps for 1% and 5% Au-Ni, respectively. Samples with different Au amounts are labelled as *x* % Au-Ni aerogel, where *x*=1, 3, and 5, respectively. The mass density of 3% Au-Ni is about 0.031 g cm^{-3}, and the actual molar ratio of Au/Ni in 3% Au-Ni aerogel is measured to be 3.8% by an ICAP7600DUO inductively coupled plasma-optical emission spectrometer (ICP-OES).

3 Characterization

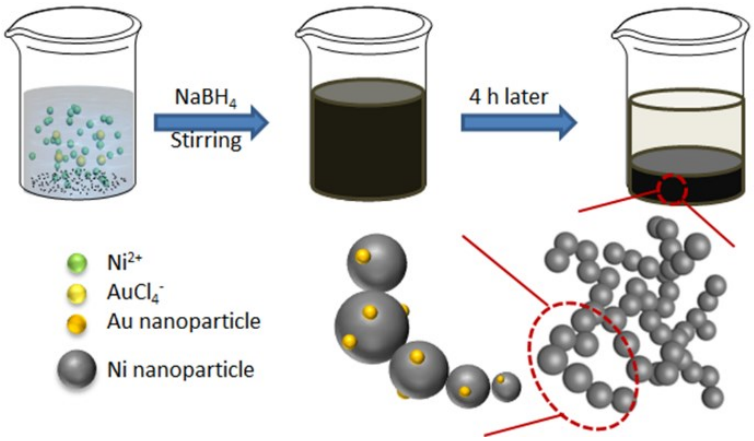
Supercritical drying was carried out on 13200J-AB obtained from SPI Supplies. Scanning electron microscopy (SEM) images were performed on a FEI Nova NanoSEM 450 scanning electron microscope with accelerating voltage of 15 kV.

Transmission electron microscopy (TEM) images, high-resolution TEM (HR-TEM), high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) images and selected area electron diffraction (SAED) pattern of aerogels were obtained on a Tecnai G2 F30 transmission electron microscope with the accelerating voltage of 200 kV. The crystalline structure of samples was analyzed by powder X-ray diffraction (XRD, Cu Kα radiation, Shimadzu XRD-6000) in the range of 10–90° (2θ). The X-ray photoelectron spectra (XPS) were measured on a ThermoFisher ESCALAB 250XI. Nitrogen physisorption isotherms were tested on a Micromeritics ASAP 2460, before measurement about 100 mg of aerogels were degassed for 12 h at 70 °C. The pore size distribution of the aerogel was assessed using the quenched solid density functional theory (DFT) equilibrium model. Molar ratio of Au/Ni is tested on an ICAP7600DUO inductively coupled plasma-optical emission spectrometer (ICP-OES).

4 Electrocatalytic measurements

The dried aerogels were dispersed in a mixture solution (500 μL water, 125 μL ethanol and 12.5 μL of 5 wt % Nafion) to obtain suspensions with weight concentration of 7.84 mg mL^{-1}. 20 μL of the suspensions was dropped onto as-prepared carbon paper (0.4 cm × 0.2 cm) with loading mass density of 1.96 mg cm^{-2}, which were dried at room temperature in the air. All electrochemical experiments were tested using CHI 660E electrochemical workstation (CHI, China) in 1.0 M KOH aqueous solution, under a three-electrode system containing working electrode loading with as-synthesized samples, reference electrode of Hg/HgO (1 M KOH) electrode and counter electrode of a platinum wire electrode. The potential was calibrated to reversible hydrogen electrode with the equation: $E_{RHE} = E_{Hg/HgO} + 0.098\ V + 0.059\ pH$. Cyclic voltammetry (CV) curves was recorded for activation at a scan rate of 10 mV s^{-1}, linear sweeping voltammetry (LSV) curves was record at a scan rate of 10 mV s^{-1}, electrochemical impedance spectra (EIS) measurements were recorded at 1.6 V (*vs.* RHE) in the frequency range from 10^5 Hz to 0.1 Hz, chronoamperometry was carried out at 1.6 V (*vs.* RHE) for 10 h. In addition, the electrochemical active surface area (ECSA) of synthesized samples was measured by CV curves under various scan rates (10, 20, 30, 40, 50 and 60 mV s^{-1}).

Supporting figures



Scheme S1. Schematic diagram for the preparation of Au-decorated Ni aerogel.

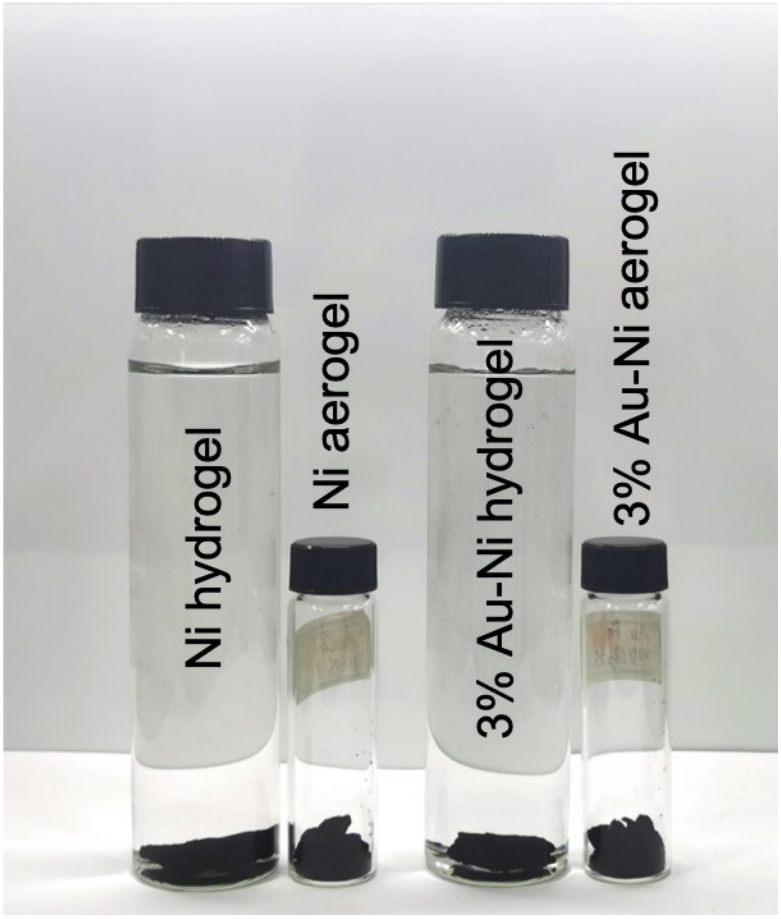


Figure s1. Photographs of the as-obtained wet gels and aerogels of Ni and 3% Au-Ni.

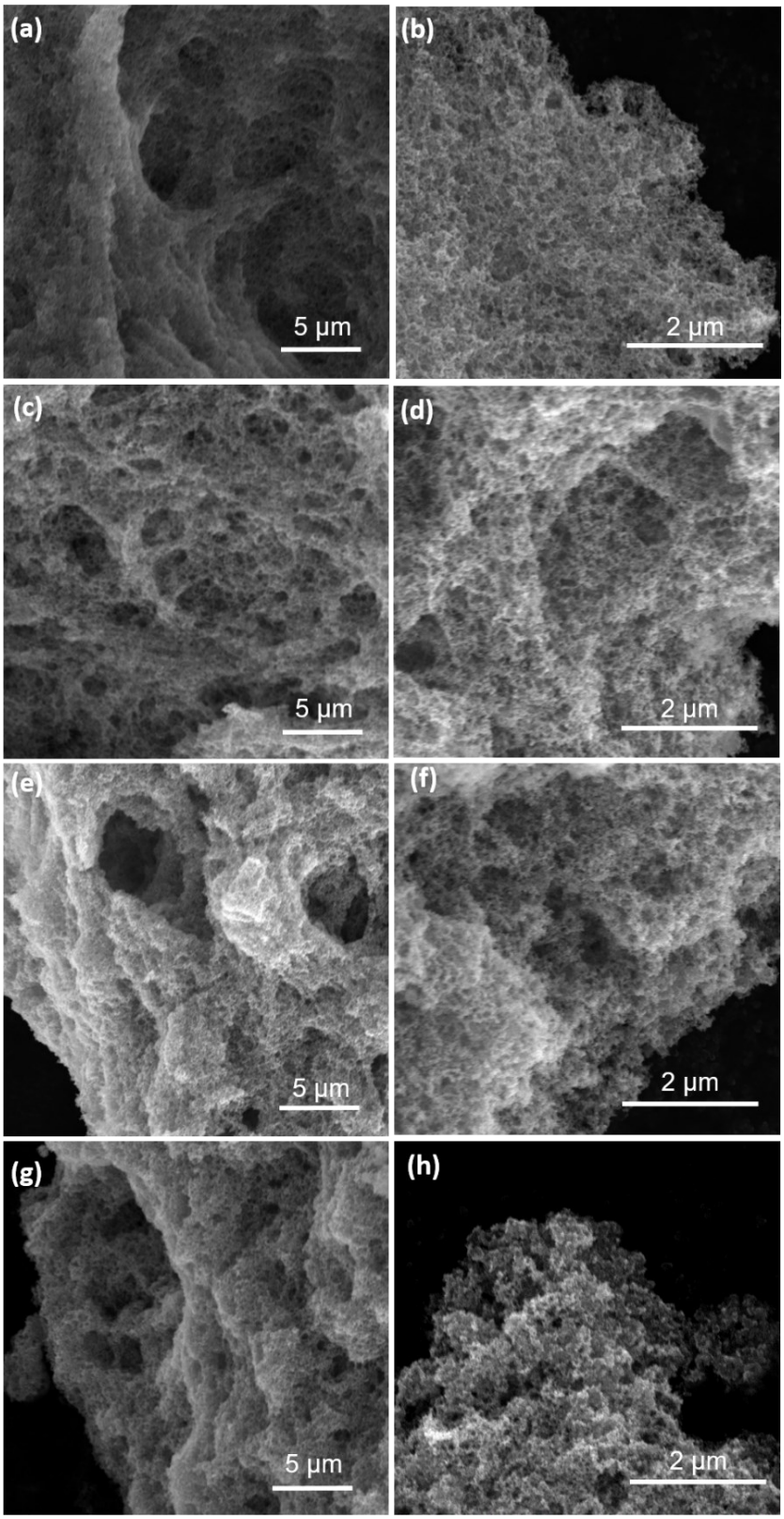


Figure s2. SEM images of Au-Ni aerogel with different Au amounts. (a, b) Ni aerogel, (c, d) 1% Au-Ni aerogel, (e, f) 3% Au-Ni aerogel, and (g, h) 5% Au-Ni aerogel.

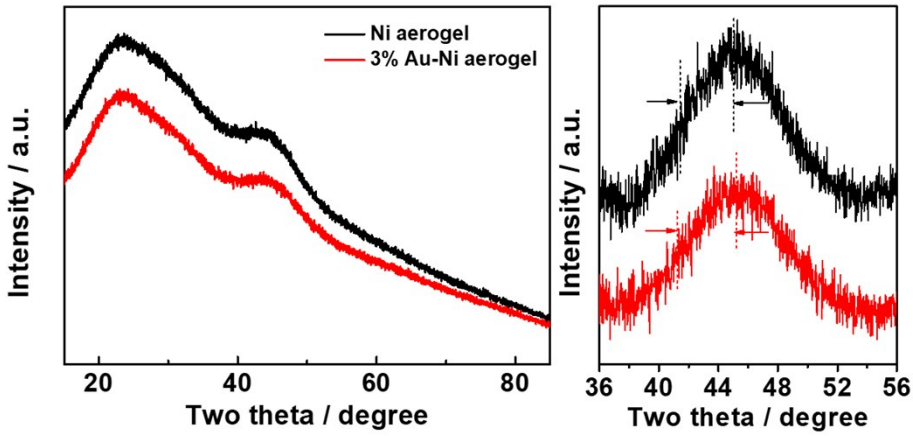


Figure s3. XRD patterns of Ni aerogel and 3% Au-Ni aerogel.

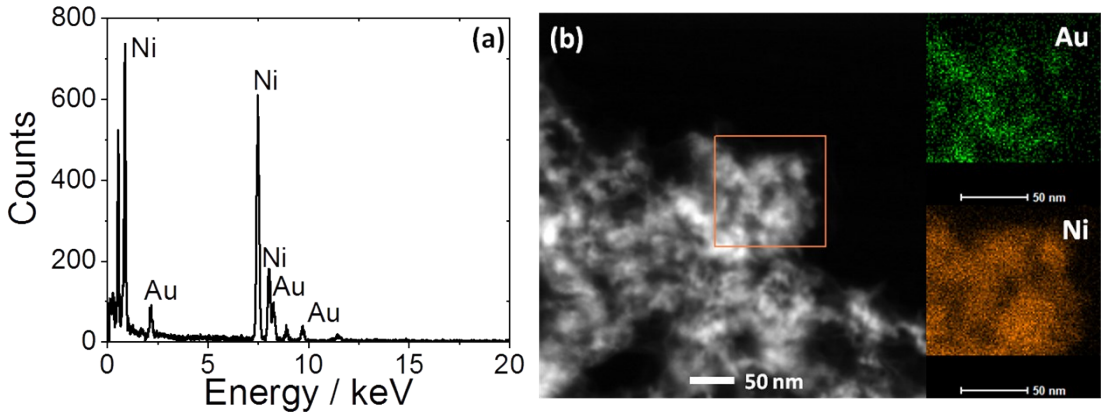


Figure s4. (a) EDS spectrum of 3% Au-Ni aerogel; and (b) mapping elements images of Au and Ni of 3% Au-Ni aerogel.

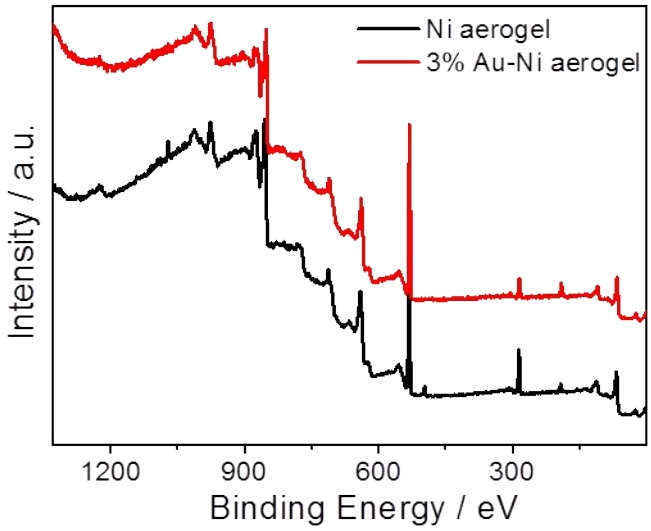


Figure s5. XPS spectra of survey for Ni aerogel and 3% Au-Ni aerogel.

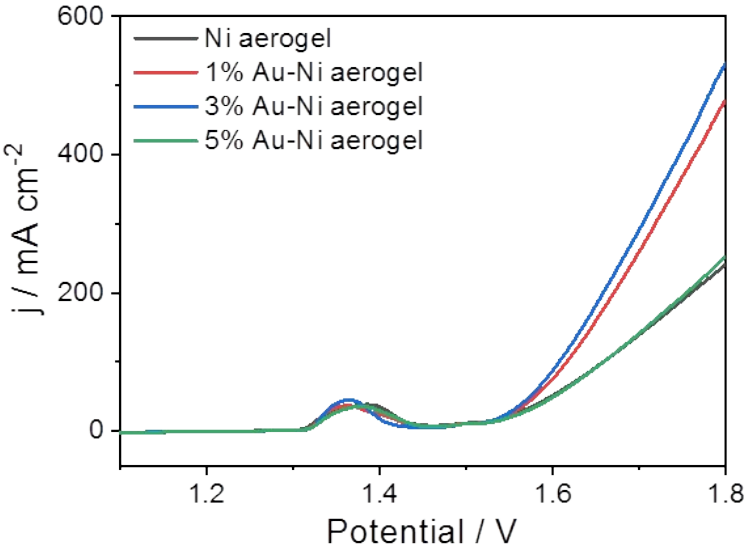


Figure s6. LSV curves of Au-Ni aerogel with different Au amount

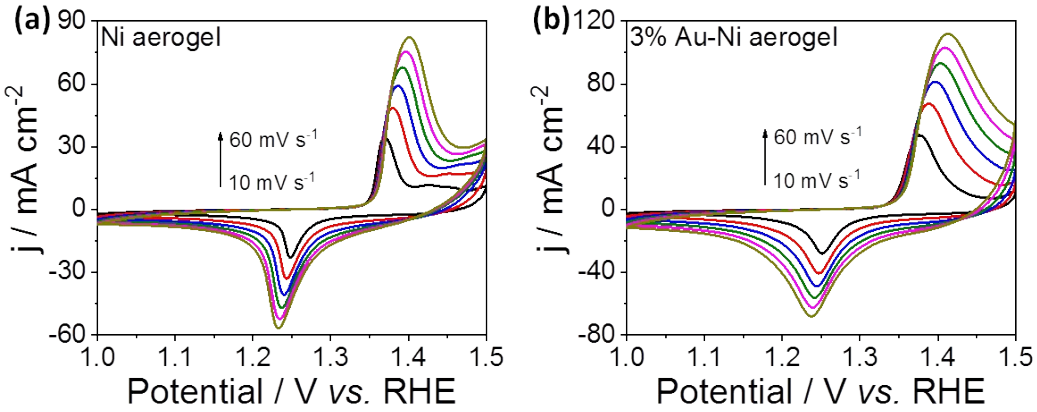


Figure s7. CV curves with various scan rates of (a) Ni aerogel and (b) 3% Au-Ni aerogel.

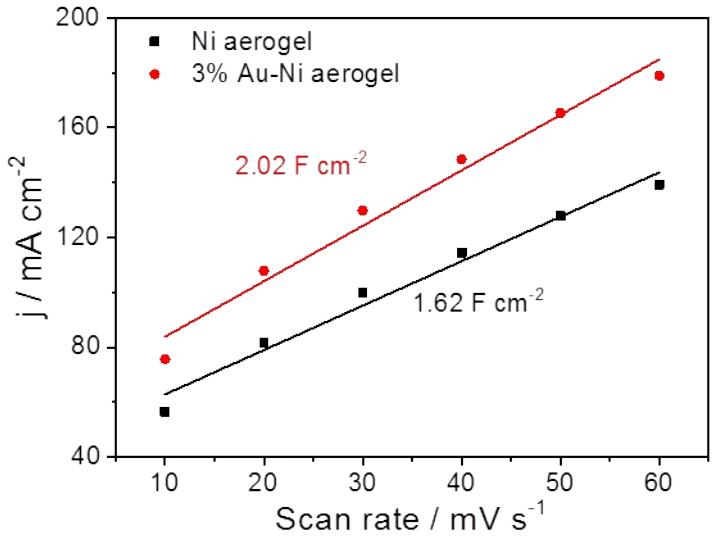


Figure s8. Oxidation peak current density versus scan rate plot for Ni aerogel and 3% Au-Ni aerogel.

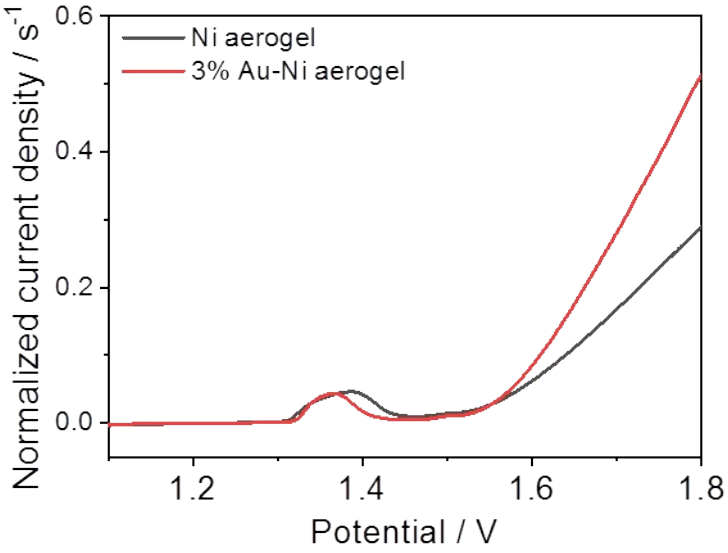


Figure s9. Normalized current density of Ni aerogel and 3% Au-Ni aerogel.

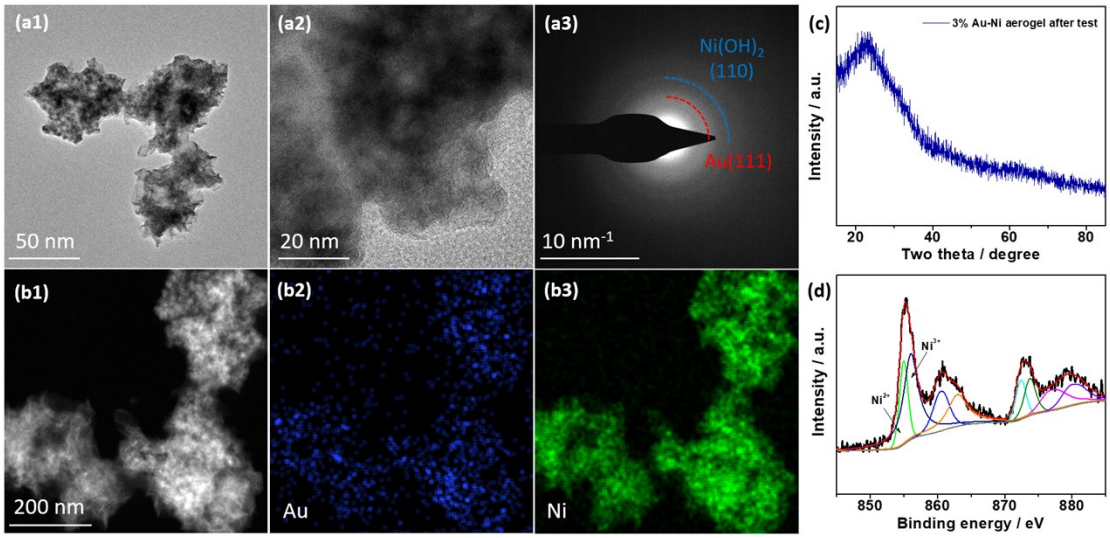


Figure s10. (a) TEM images and SAED pattern, (b) HAADF and elemental mapping images, (c) XRD, and (d) XPS of Ni 2p spectrum for 3% Au-Ni aerogel after OER stability test.

Table s1. Comparison of OER catalytic activity of various Ni-based electrocatalysts.

Catalyst	Electrolyte	Current density (mA cm^{-2}) @ overpotential	Tafel slope (mV dec^{-1})	Reference:
Ni aerogel	1.0 M KOH	100 @ 377 mV	85.3	This work
3% Au-Ni aerogel	1.0 M KOH	100 @ 428 mV	129.6	This work
3DG−Au-Ni_3S_2	1.0 M KOH	91.2 @ 370 mV	106	ACS Appl. Energy Mater., 2019, 2, 3708.
$Ni(OH)_2$-TCNQ/Cu foam	1.0 M KOH	100 @ 354 mV	110	ACS Catal., 2018, 8, 1, 651
Fe-doped Ni_2P	1.0 M KOH	20 @ 330 mV	39	Chem. Commun., 2018, 54, 863
Ni-NC700	0.1 M KOH	10 @ 430 mV	100	ACS Sustainable Chem. Eng., 2019, 7, 2187
2%Au/Ni_3N-10	1.0 M KOH	10 @ 280 mV	52	ChemElectroChem, 2019, 6, 5744
Au/$Ni_{12}P_5$	1.0 M KOH	10 @ 340 mV	49	Nano Res., 2017, 10, 3103
PO-Ni/Ni-N-CNFs-1000 + 300	1.0 M KOH	10 @ 420 mV	113.1	Nano Energy, 2018, 51, 286
Ni/$Ni(OH)_2$	1.0 M KOH	10 @ 270 mV	70	Adv. Mater., 2020, 32, 1906915
A-Ni@DG	1.0 M KOH	10 @ 279 mV	47	Chem, 2018, 4, 285
Ni/NiO@G-SH	1.0 M KOH	10 @ 270 mV	46	ACS Appl. Energy Mater., 2019, 2, 363
Ni-BDC/$Ni(OH)_2$	1.0 M KOH	82.5 @ 370 mV	41	Nanoscale, 2019, 11, 3599