Supplementary Information

Experimental details:

Binder free cobalt oxide was synthesized using a solvothermal method. In a typical synthesis, 3 mM of cobalt nitrate was dissolved in 18 ml solution containing equal volume water and ethanol. In this solution, 300 mg of polyvinylpyrrolidone was dissolved followed by addition of 12 mM of urea (dissolved in 18 ml water/ethanol, 1:1 v/v). The resulting solution was poured in to a 45 mL Teflon lined autoclave. A pre-cleaned nickel foam was placed in the reactor. Nickel foam was cleaned using 3 M HCl, followed by washing using water and isopropanol. The reaction was carried out for 12 hours at 140 °C. Gray colored powder was uniformly deposited on the nickel foam which was washed with plenty of water and finally with ethanol. The obtained gray powder deposited nickel foam was dried at 60 °C for 8 hours and followed by calcination at 350 °C for 3 hours.

X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS) and scanning electron microscopy (SEM) was used for structural characterization of the synthesized sample. XRD and XPS were performed on the scraped out powder sample from nickel foam. The XRD powder pattern was recorded using Shimadzu X-ray diffractometer using the 2θ–θ scan with CuKα_1 (λ=1.5406 Å) radiation. The X-ray was operated at 40 kV and 30 mA. A Thermo Scientific Kα XPS system equipped with a monochromatic X-ray source at 1486.6 eV, corresponding to the Al Kα line was used for XPS measurements. The X-ray power of 75 W at 12 kV was used for the experiment with a spot size of 400 mm^2. The XPS data acquisition was performed using the "Avantage v5.932" software provided with the instrument. The morphology of the synthesized cobalt oxide was explored using a JEOL JSM-840A scanning electron microscope and an FEI Quanta 200 field emission scanning electron microscopy (FESEM) equipped with an Oxford INCA 250 silicon drift X-ray energy dispersive spectrometer (EDS).

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Electrocatalytic activities of the cobalt oxide was studied using standard three electrode system using a Versastat4-500 electrochemical workstation (Princeton Applied Research, USA). Platinum wire and saturated calomel electrode (SCE) was used as counter and reference electrode, respectively. Cobalt oxide on nickel foam was used as a working electrode. Electrochemical properties were studied using linear sweep voltammetry (LSV), cyclic voltammetry (CV), chronoamperometry and electrochemical impedance spectroscopic (EIS) techniques in 1M NaOH (pH = 13.7) solution. LSV was performed at scan rate of 1 mV/s. EIS measurements were performed in a frequency range of 0.05 Hz to 10 kHz with an applied 10 mV of AC amplitude. The potential was converted to RHE using the Nernst equation [1].
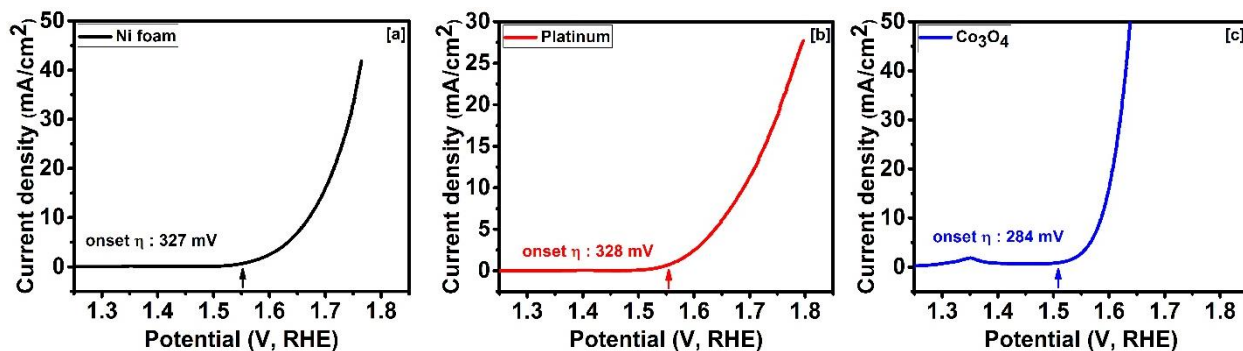

Figure 1S: OER polarization curves for (a) Ni foam, (b) platinum and (c) Co_3O_4 showing onset overpotential (defined as potential at which current increases sharply minus 1.23V, vs RHE) in 1M NaOH.
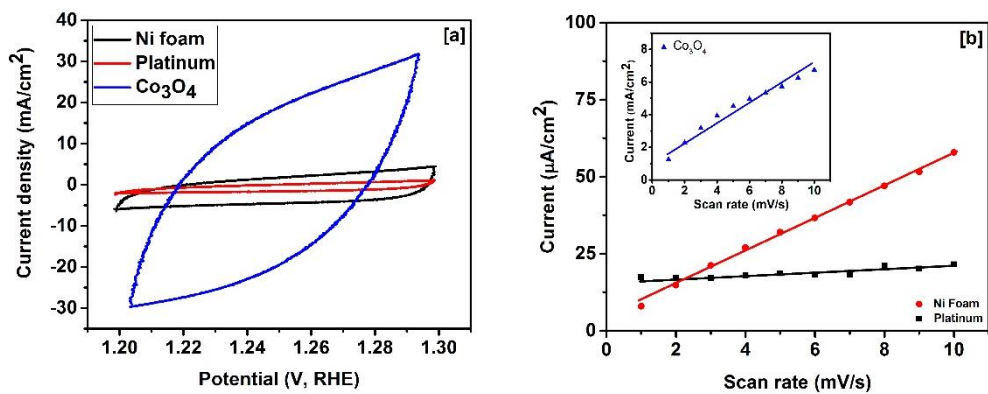
Figure 2S: (a) Comparison of CV curves and (b) variation of current densities against scan rate at 1.25 V for nickel foam, platinum and cobalt oxide in 1M NaOH.
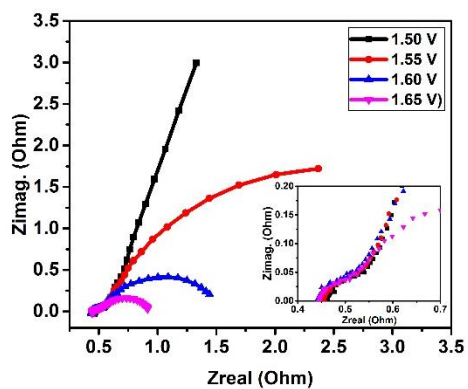

Figure 3S: Nyquist plots of Co_3O_4 electrode at different potentials (vs RHE).
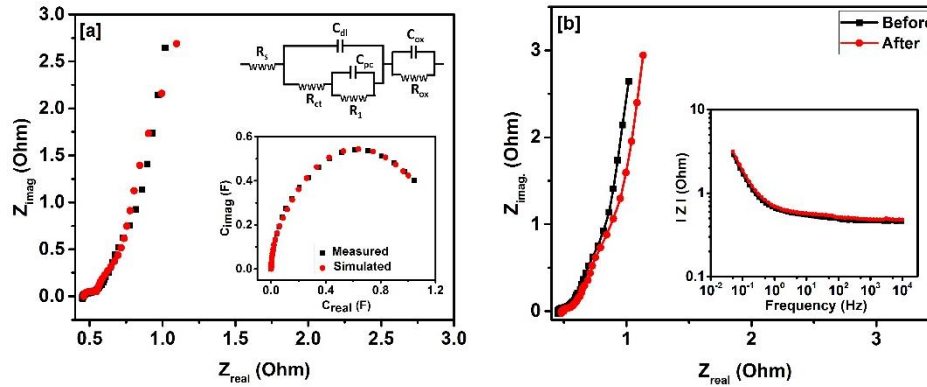
Figure 4S: (a) Measured and simulated Nyquist plots for Co_3O_4 before stability test (inset figure shows equivalent circuit used for fitting (top) and measured and simulated real and imaginary capacitance (bottom), (b) Nyquist plots for Co_3O_4 before and after stability test (inset figure shows variation of IZI as a function of frequency) in 1M NaOH.

Table 1S: NLLS-fit parameters of the elements before and after stability test of Co_3O_4 using an equivalent circuit of Figure 9a.

	R_s (Ω)	C_{dl} (F)	R_{ct} (Ω)	C_{pc} (F)	R_1 (Ω)	C_{ox} (F)	R_{ox} (Ω)
Before stability test	0.459	0.046	0.099	1.119	18.83	1.333	0.020
Before stability test	0.493	0.038	0.985	1.001	23.04	1.186	0.270

References

[1] H. Wang, H.-W. Lee, Y. Deng, Z. Lu, P.-C. Hsu, Y. Liu, D. Lin, Y. Cui, Bifunctional non-noble metal oxide nanoparticle electrocatalysts through lithium-induced conversion for overall water splitting. Nature Communications 6, 7261 (2015).