Electronic Supplementary Information (ESI) for Dalton Transactions
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Supporting Information

Silver decorated nickel-cobalt (oxy)hydroxides fabricated via surface reconstruction engineering for boosted electrocatalytic oxygen evolution and urea oxidation

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S1 Figures in Supporting Information

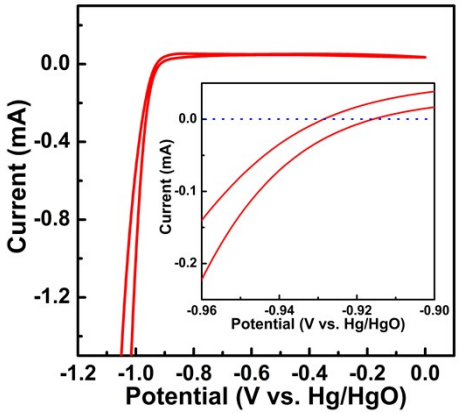

Fig. S1 CV curves of Hg/HgO electrode calibration in 1.0 M KOH. Inset: the enlarged view.

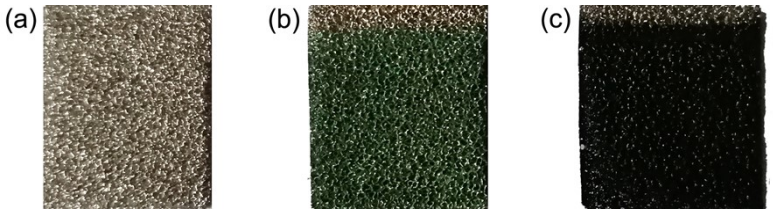

Fig. S2 Digital images of (a) NF, (b) NiCo LDH/NF, and (c) Ag@NiCo(OH)$_x$/NF.

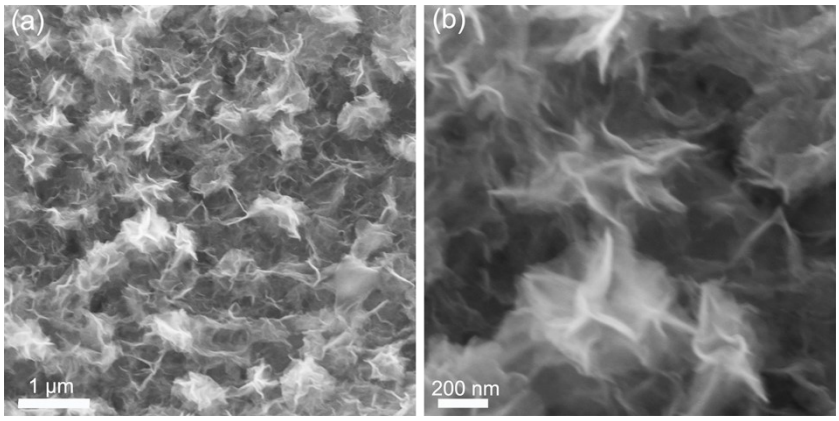

Fig. S3 (a, b) SEM images of NiCo LDH/NF.

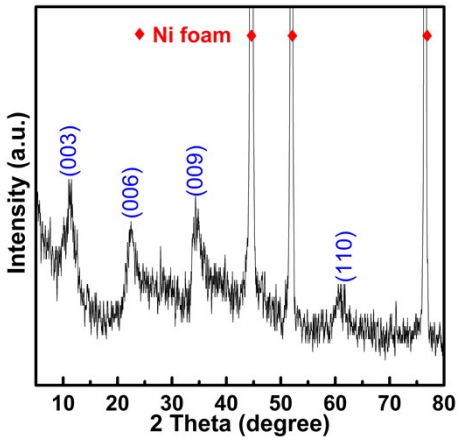

Fig. S4 PXRD pattern of NiCo LDH/NF.

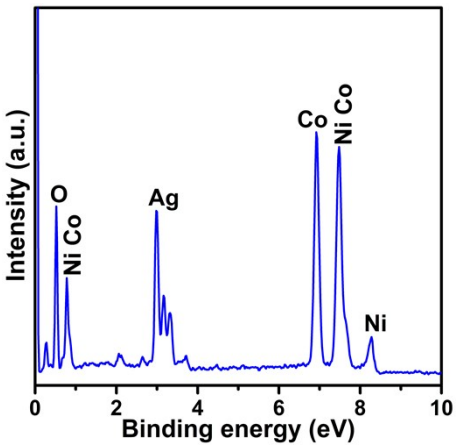

Fig. S5 EDX spectrum of the Ag@NiCo(OH)$_x$/NF.

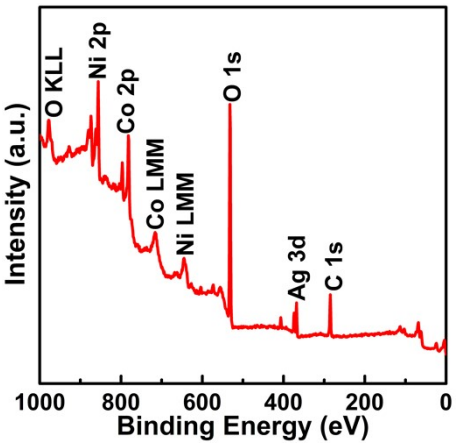

Fig. S6 XPS survey spectrum of the Ag@NiCo(OH)$_x$/NF.

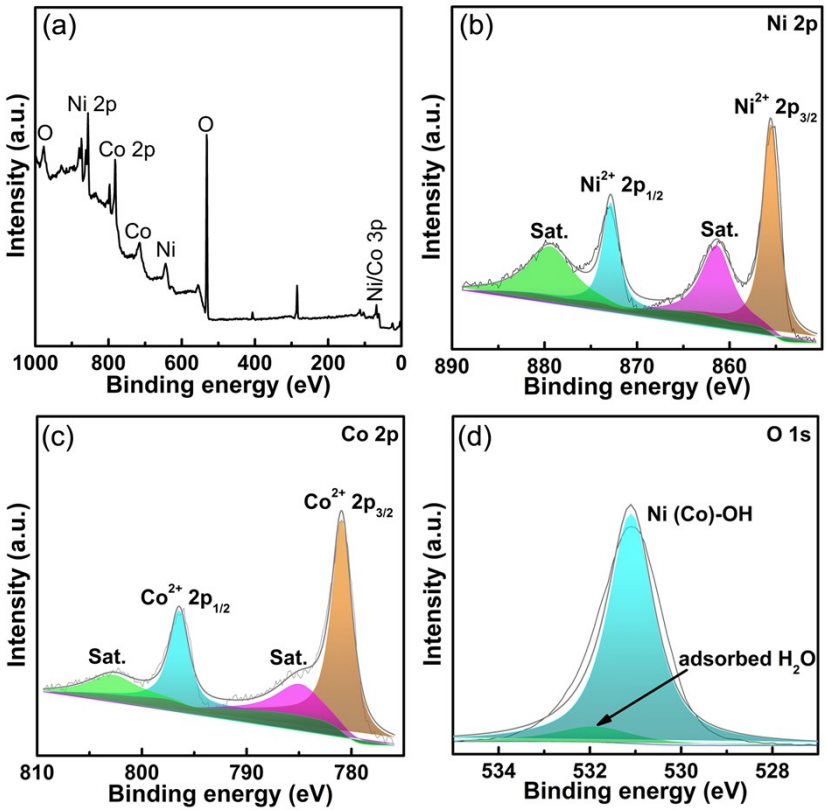

Fig. S7 (a) XPS survey spectrum, (b-d) high resolution XPS spectra of Ni 2p, Co 2p, and O 1s of NiCo LDH/NF, respectively.

As shown in Fig.S7a, the XPS survey spectrum reveals the existence of Co, Ni, and O elements in NiCo LDH/NF. In the high-resolution Ni 2p spectrum (Fig. S7b), two main peaks for Ni $2p_{3/2}$ and Ni $2p_{1/2}$ located at 855.5 and 872.8 eV are accompanied by two shake-up satellite peaks (861.4 and 879.3 eV), respectively, which are characteristic spin-orbit peaks of Ni^{2+} (Fig. S7b). For the Co XPS spectrum in Fig. S7c, the peaks appeared at 780.9 and 796.4 eV are attributed to the Co^{2+}. The other peaks (784.9 and 802.8 eV) belong to shakeup satellite peaks. As seen from O 1s XPS spectrum (Fig. S7d), the peaks at 531.1 and 531.9 eV correspond to Ni (Co)-OH and adsorbed H_2O, respectively.

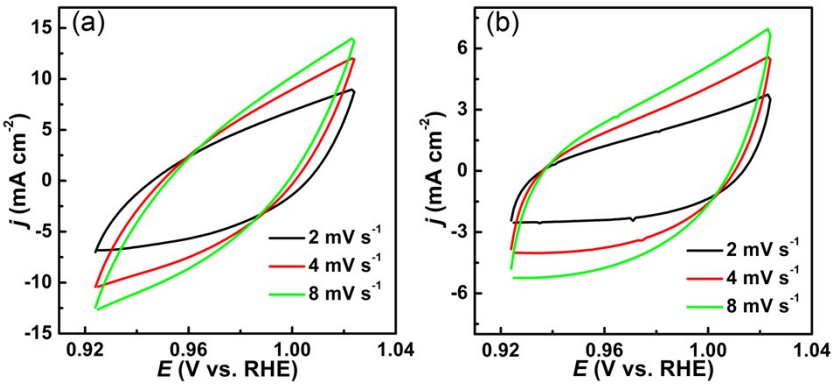

Fig. S8 Cyclic voltammograms of (a) NiCo LDH/NF and (b) Ag@NiCo(OH)$_x$/NF from 0.924 to 1.024 V vs. RHE at different scan rates in 1.0 M KOH aqueous solution.

Calculation of ECSA:

$$\mathrm{ECSA} = \frac{C_{dl}}{C_s}$$

$$\mathrm{ECSA}_{\mathrm{NiCo\ LDH/NF}} = \frac{356.5\ \mathrm{mF\,cm^{-2}}}{40\ \mu\mathrm{F\,cm^{-2}}} = 8912.5\ \mathrm{cm^{-2}}_{\mathrm{ECSA}}$$

$$\mathrm{ECSA}_{\mathrm{Ag@NiCo(OH)_x/NF}} = \frac{434.1\mathrm{mF\,cm^{-2}}}{40\ \mu\mathrm{F\,cm^{-2}}} = 10852.5\,\mathrm{cm^{-2}}_{\mathrm{ECSA}}$$

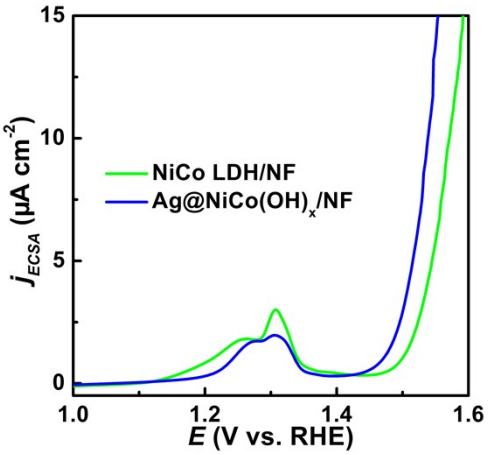

Fig. S9 LSV curves of NiCo LDH/NF and Ag@NiCo(OH)$_x$/NF normalized by C_{dl}.

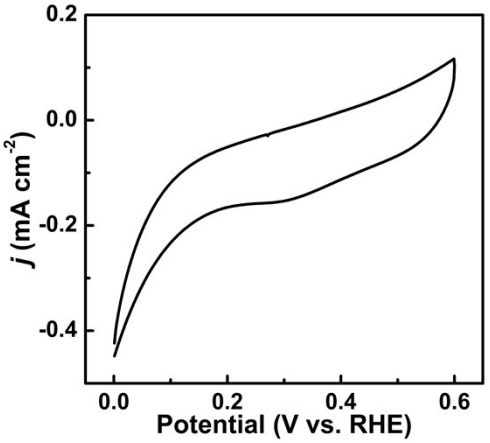

Fig. S10 CV curve for Ag@NiCo(OH)$_x$/NF measured in 1.0 M KOH.

The calculation method of TOF value is according to the following equation:

$$\mathrm{TOF} = \frac{j}{4Fn}$$

where j is the measured current density (A cm^{-2}) for the OER, F is the Faraday constant (96485 C mol^{-1}) and n is the number of active sites (mol cm^{-2}). The factor 1/4 is shown because four electrons are needed to generate one oxygen molecule. The number of active sites is achieved according to cyclic voltammograms (CV) method. Specifically, the CV curve is investigated in 1.0 M KOH solution with the potential window range from 0 V to 0.6 V *vs* RHE at 50 mV s^{-1}. Then, by integrating the CV curve's charge over the whole potential range, the half value of the charge was obtained, which is the value of the surface charge density (Qs).

$$\mathrm{Qs} = Fn$$

In 1.0 M KOH solution, the TOF value of Ag@NiCo(OH)$_x$/NF is estimated to be 0.18 s^{-1} at an overpotential of 260 mV.

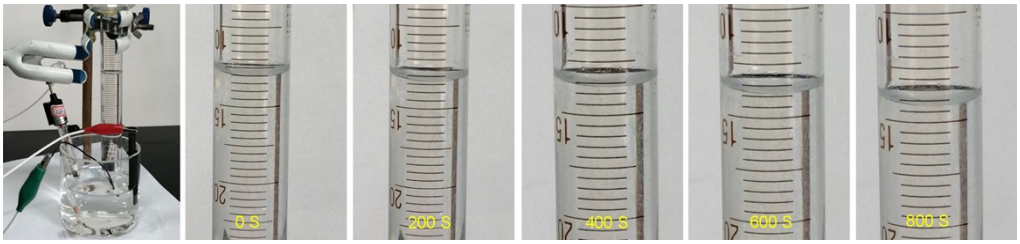

Fig. S11 Gas collection device for OER and photographs of oxygen collected at different time in 1.0 M KOH.

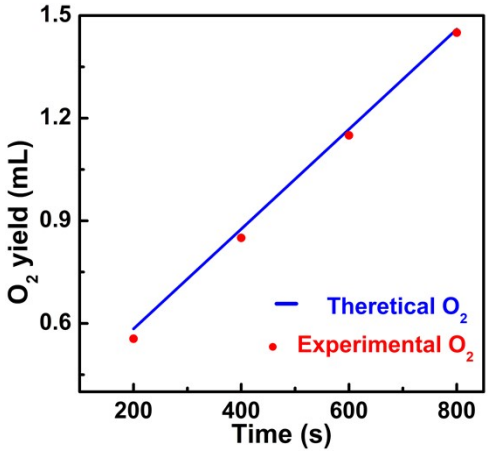

Fig. S12 Theoretically calculated and experimentally measured amount of oxygen versus time for Ag@NiCo(OH)$_x$/NF in 1.0 M KOH.

To estimate the faradaic efficiency (FE), a gas collection device was employed for the experiment (**Fig. S11**). The FE is estimated from the observed gas volume and the theoretical gas volume calculated by the charge passed through the electrode. The related formula is as follows: $\mathrm{FE} = V_{experimental} / [V_m \times Q / (NF)]$, where the N is the number of transferred electrons, F is the faraday constant (96485 C mol^{-1}), Q is the charge passed through the electrode, and V_m is the gas molar volume at 298 K and 101 kPa (24.5 L mol^{-1}). As shown in **Fig. S12**, the faradaic efficiency of Ag@NiCo(OH)$_x$/NF for the OER is calculated to be 98.1%.

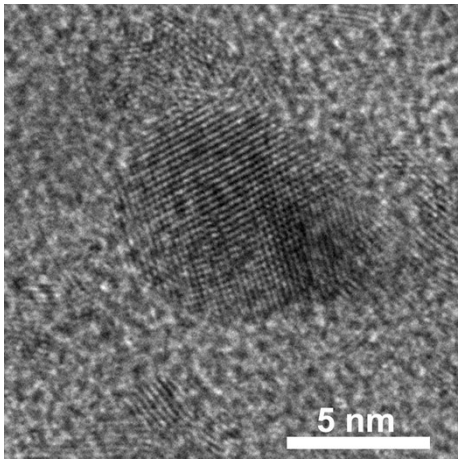

Fig. S13 HRTEM image of Ag@NiCo(OH)$_x$/NF after stability test for OER.

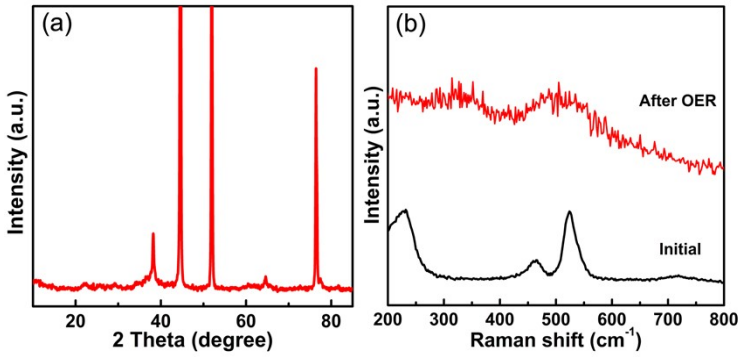

Fig. S14 (a) PXRD pattern and (b) Raman spectra of Ag@NiCo(OH)$_x$/NF after stability test for OER.

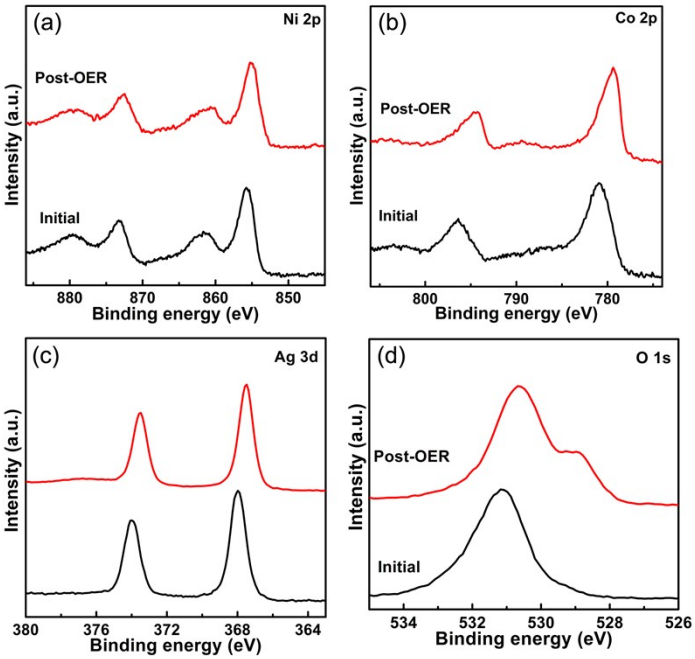

Fig. S15 (a-d) High-resolution XPS spectra of Ni 2p, Co 2p, Ag 3d, and O 1s for Ag@NiCo(OH)$_x$/NF initial and after stability test for OER.

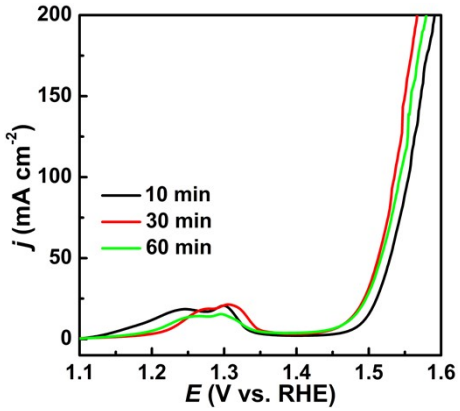

Fig. S16 (a) LSV curves for the OER of Ag@NiCo(OH)$_x$/NF with different immersion time (10, 30, and 60 min).

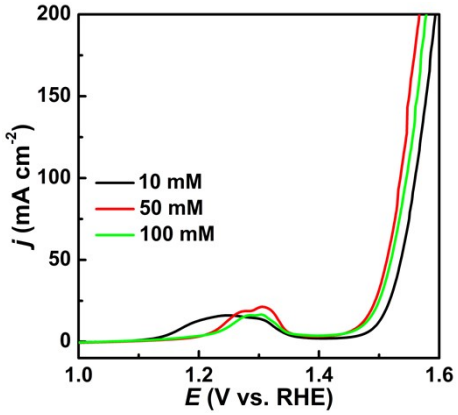

Fig. S17 (a) LSV curves for the OER of Ag@NiCo(OH)$_x$/NF with different concentration of $AgNO_3$ solution (10, 50, and 100 mM).

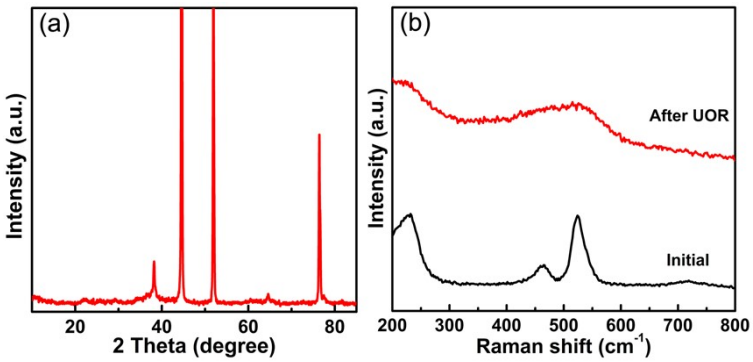

Fig. S18 (a) PXRD pattern and (b) Raman spectra of Ag@NiCo(OH)$_x$/NF after stability test for UOR.

S2 Tables in Supporting Information

Table S1. Comparison of the OER performance of Ag@NiCo(OH)$_x$/NF with other reported electrocatalysts.

Catalysts	Tafel slope (mV dec^{-1})	Overpotential (mV) at 10 mA cm^{-2}	References
Ag@NiCo(OH)$_x$/NF	**64.6**	**236**	**This work**
Ag@NiV$_{0.2}$Co$_{0.2}$	38.2	255	*Chem. Eng. J.* 2021, 425, 131662.
Ag-doped CoOOH	64.6	256	*ACS Catal.* 2020, 10, 562−569
Ag NW@NiMn-LDHs(1:2)	40.2	270	*ACS Nano* 2020, 14, 1770−1782
CoSeP	87	255	*ACS Energy Lett.* 2019, 4, 987.
Fe, Co-NiSe$_2$	63	251	*Adv. Mater.* 2018, 30, 1802121.
Ag@CoxP	76.4	310	*ACS Catal.* 2018, 7, 7038-7042.
Ni(OH)$_2$-NP	78.6	260	*ACS Nano* 2018, 12, 3875.
2D NiCoFe NSs/NF	58	240	*Nanoscale* 2018, 10, 12975.
ANF@NW	103	382	*Small 2018* , 14, 1800294.
NF@Ni/C	54	265	*Energy Environ. Sci* . 2018, 11, 2363.
Co–Ni–B@NF	120	313	*J. Mater. Chem. A* 2017, 5, 12379.
CoO-MoO$_2$	70	312	*Adv. Funct. Mater.* 2017, 27, 1702324.
NiCoP/NF	87	280	*Nano Lett.* 2016, 16, 7718-7725.

Table S2. The concentration of $AgNO_3$ aqueous solution, immersion time, and the OER overpotential at 50 mA cm^{-2} of different Ag@NiCo$(OH)_x$ /NF samples.

Concentration of $AgNO_3$ (mM)	Immersion time (min)	Overpotential (mV) @ 50 mA cm^{-2}
10	30	307
50	30	280
50	10	302
50	60	291
100	30	289

Table S3. Comparison of the UOR performance of Ag@NiCo(OH)$_x$/NF with other reported electrocatalysts.

Catalysts	Electrolyte	Potential at 10 mA cm^{-2}	References
Ag@NiCo(OH)$_x$/NF	**1 M KOH + 0.33 M urea**	**1.24**	**This work**
CoFeCr LDH/NF	1 M KOH + 0.33 M urea	1.305	*Appl. Catal. B: Environ.* , 2020, 272, 1189596.
C-350	1 M KOH + 0.33 M urea	1.337	*ACS Sustainable Chem. Eng.* 2020, 8, 7414.
$NiCo_2S_4$	1 M KOH + 0.33 M urea	1.32	*ACS Sustainable Chem. Eng* . 2018, 6, 5011.
Fe-Ni_3S_2/NF	1 M KOH + 0.33 M urea	1.44	*J. Mater. Chem. A* , 2018, 6, 4346.
Ni-MOF	1 M KOH + 0.33 M urea	1.36	*Chem. Commun.* 2017, 53, 10906.
$Ni(OH)_2$	1 M KOH + 0.33 M urea	1.38	*Angew. Chem. Int. Ed* . 2016, 55, 12465.