Supporting Information

Ultralow platinum-doped self-supported CoPt alloy nanosheets for efficient overall water splitting

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Experimental sections

Materials

Analytical grade $Co(NO_3)_2 \cdot 6H_2O$ and Potassium chloride (KCl, ≥99.5%) were purchased from Shanghai Chemical Corp. Titanium mesh (wire diameter: 0.32 mm) was obtained from Changsha Keliyuan. Potassium hydroxide (KOH, ≥85%) and Potassium hexachloroplatinate solution (K_2PtCl_6, 1 wt %) were purchased from Sinopharm Chemical Reagent Co. Pt/C (20 wt % Pt on Vulcan XC-72R), Iridium oxide (IrO_2), and Nafion (5 wt %) were purchased from Sigma-Aldrich Chemical Reagent Co. All chemicals were used as received without further purification. Ultrapure water was used throughout the experiments.

Preparation of Co-OH and CoPt-OH on Ti mesh

First, the titanium mesh was immersed in 6 M HCl and sonicated for 20 minutes. Then, 295.0 mg of $Co(NO_3)_2 \cdot 6H_2O$ and 300 mg of KCl were dissolved in deionized water to prepare 40 mL of electrolyte. Using a standard three-electrode system, the titanium mesh served as the working electrode, and Co-OH/Ti electrode material was obtained after electrodeposition at a constant potential of −0.7 V for 2 hours. To the same electrolyte, 200 μL of 1 wt% K_2PtCl_6 was added, and under the same conditions, CoPt-OH/Ti was obtained after another 2 hours of electrodeposition.

Preparation of CoPt-R10 on Ti mesh

The prepared CoPt-OH/Ti electrode material was immersed in a 0.5 M $NaBH_4$ solution and reduced for 10 minutes, resulting in the electrode material labeled as CoPt-R10. By controlling the reduction time to 5, 15, and 25 minutes, CoPt-R5, CoPt-R15, and CoPt-

R25 were obtained, respectively.

Preparation of Pt/C and IrO_2 on Ti mesh

Prepare the Pt/C catalyst by dissolving 5 mg of Pt/C (20 wt % Pt) into 1 mL of 50 μL of 5% Nafion solution, 500 μL of DI water, and 450 μL of ethanol. A homogeneous catalyst solution was obtained by sonicating the mixture for 30 min. Subsequently, 25 μL of the solution was added dropwise to the Ti mesh several times using a micro sampler. Finally, it was weighed after drying in the oven for subsequent testing and the loading of 20 wt % Pt/C/Ti is about 1.87 mg cm^{-2}. The preparation of IrO_2/Ti (with a loading of approximately 2.13 mg cm^{-2}) catalyst is the same as the above method.

Characterizations

Scanning electron microscope (SEM, Zeiss Supra55) set at 5.0 kV, transmission electron microscopy (TEM, Tecnai 12 F30 S-TWIN) of 200 kV were conducted for the morphology characterization of the prepared materials. The crystal structure was acquired by X-ray diffraction (XRD) on the D8 advance superspeed powder diffractometer (Bruker) with Cu Kα radiation (λ = 0.1541 nm). X-ray photoelectron spectroscopy (XPS, ESCALAB250Xi) was used to identify surface chemical species.

Electrochemistry measurements

In this work, an electrochemical workstation (CHI 660E) was utilised to complete various electrochemical tests of the catalysts with a standard three-electrode system. The self-supported catalytic electrodes were used as the working electrode, and counter and reference electrodes were graphite rods and Hg/HgO electrode, respectively. The test potentials were calculated by $E(RHE) = E(Hg/HgO) + (0.098 + 0.059\ pH)$ V with respect to the reverse hydrogen electrode (RHE). *IR*-correction was performed during the tests of the polarisation curves. The electrochemical impedance spectroscopy (EIS) reflects the conductivity of the catalyst at 10 mA cm^{-2} for HER and OER processes. Cyclic voltammetry tests were used to determine the bilayer capacitance and electrochemical active area of the self-supported catalysts. Cyclic voltammetry (CV) curves for the non-Faradaic region were recorded at different scan rates (20, 40, 60, 80, and 100 mV s^{-1}) in the potential range of 0.2 ~ 0.3 V vs RHE, and each $\Delta j/2$ was calculated at 1.15 V vs RHE to evaluate the double-layer capacitance (C_{dl}) values. The electrochemical surface area (ECSA) values were calculated by $C_{\mathrm{dl}}/C_{\mathrm{s}}$, where C_{s} is the specific capacitance using an average value of 0.040 mF cm^{-2} referred to literature.[1-4] A two-electrode system was employed to evaluate the overall water splitting performances of assembled full cells. LSV curves were obtained with a scan rate of 2 mV s^{-1}. A water displacement method was employed to collect and measure oxygen gas producing at 200 mA cm^{-2} for calculating the Faraday efficiency (FE). Chronoamperometry was applied to evaluate the stability of electrode materials for overall water splitting.

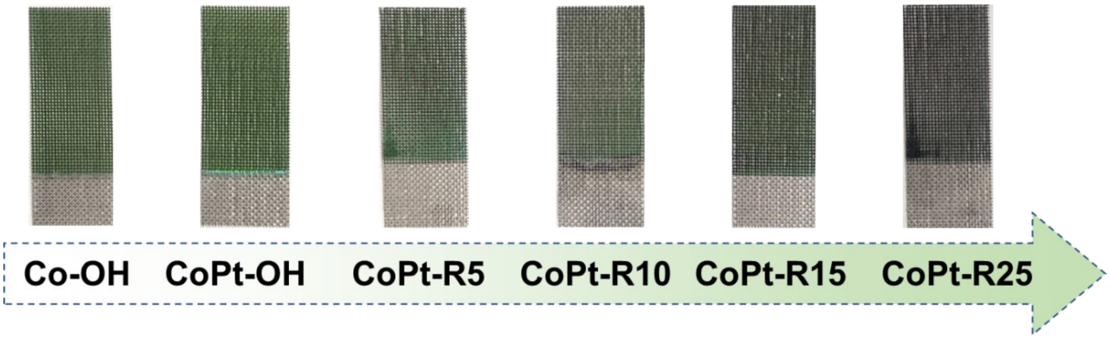

Fig. S1 Digital photographs of electrode materials.

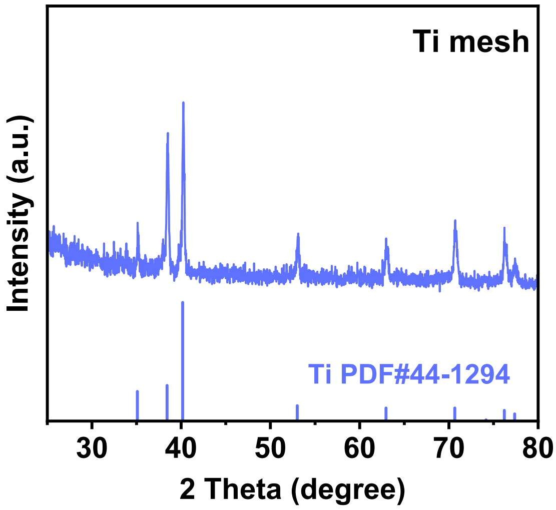

Fig. S2 XRD spectra of Ti mesh.

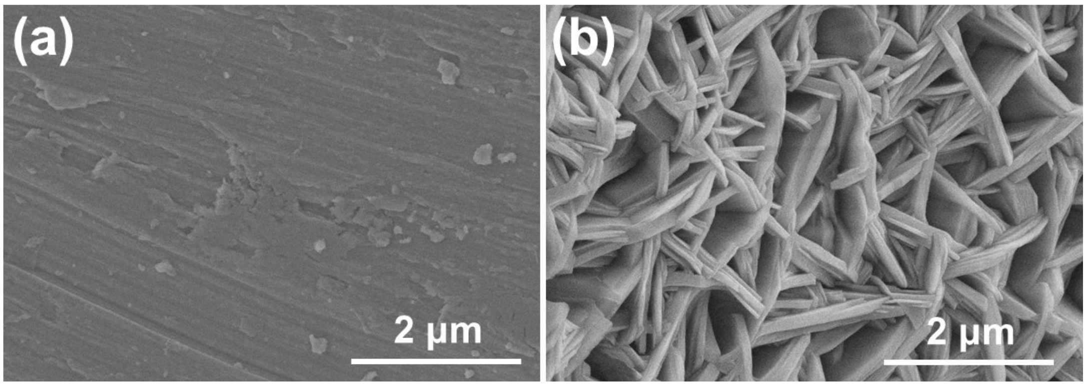

Fig. S3 SEM images of (a) pure Ti mesh and (b) Co-OH nanosheets.

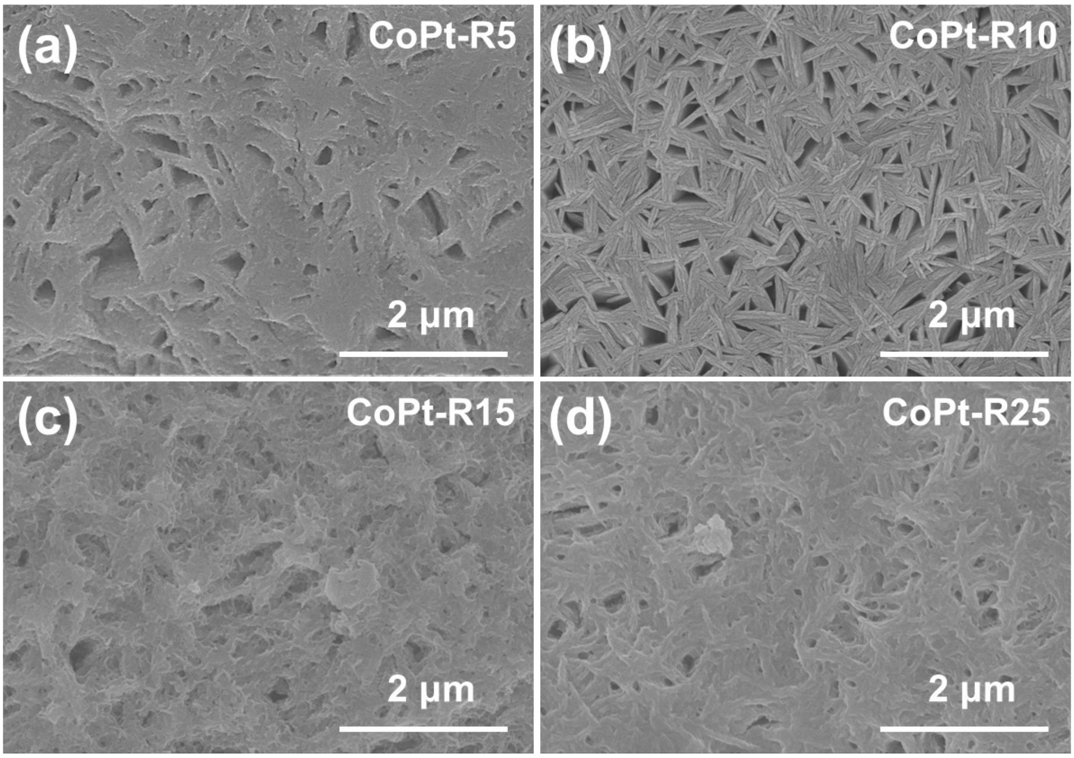

Fig. S4 (a-d) SEM images of CoPt-R5, CoPt-R10, CoPt-R15 and CoPt-R25 nanosheets.

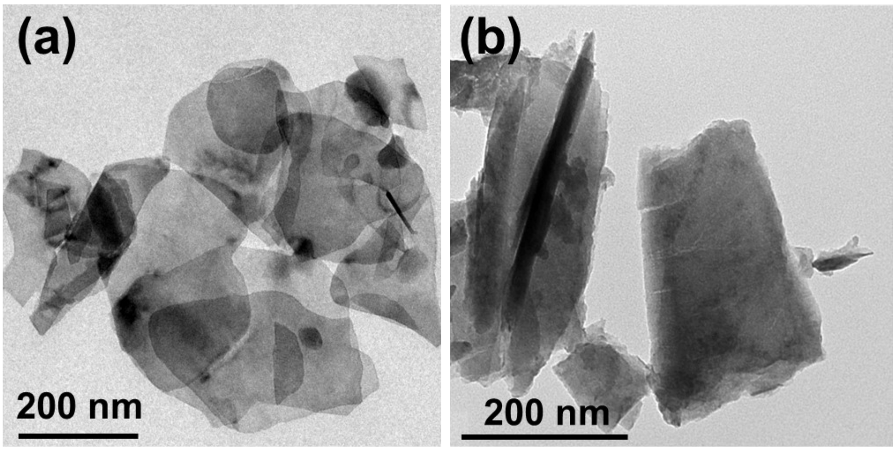

Fig. S5 (a, b) TEM images of Co-OH nanosheets.

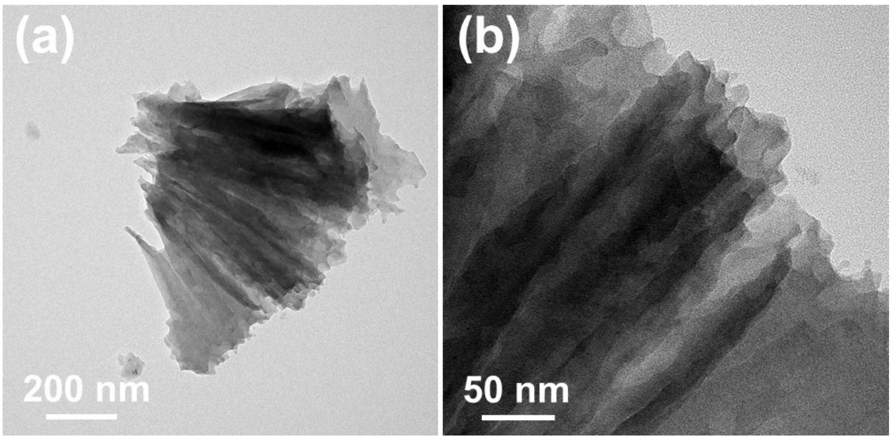

Fig. S6 (a, b) TEM images of CoPt-OH nanosheets.

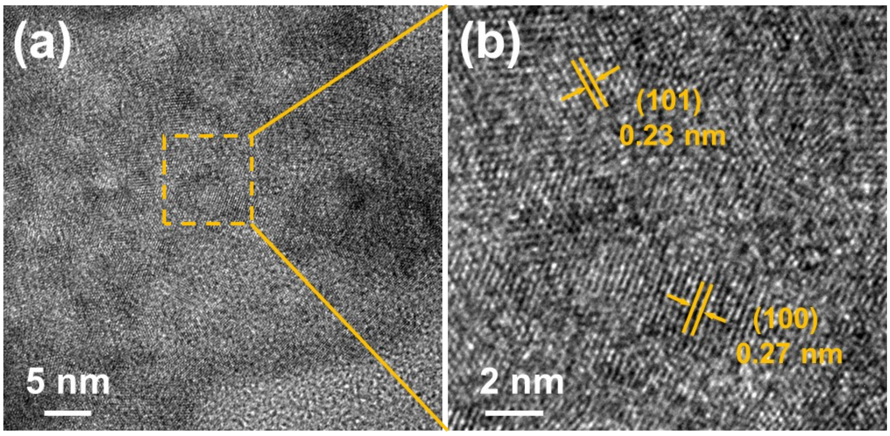

Fig. S7 (a, b) HR-TEM images of CoPt-OH nanosheets.

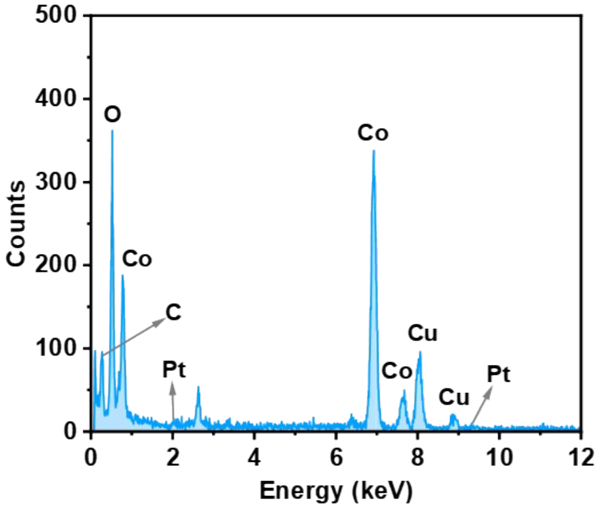

Fig. S8 EDS spectra of CoPt-R10.

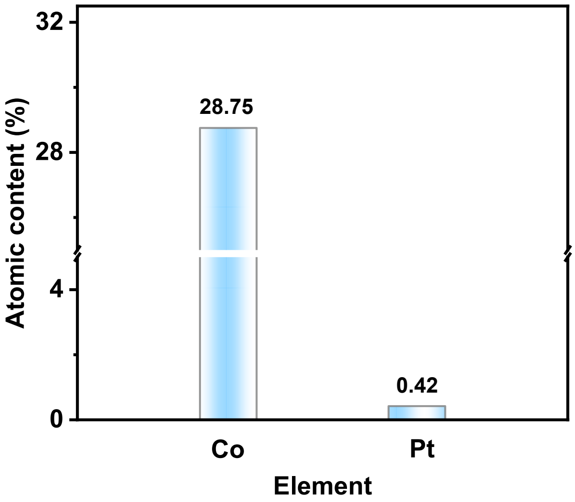

Fig. S9 Atomic percentage of the elements Co and Pt for CoPt-R10.

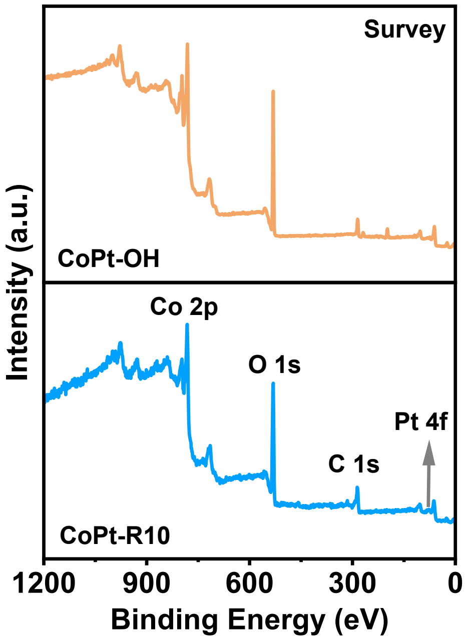

Fig. S10 XPS survey spectrum of CoPt-OH and CoPt-R10.

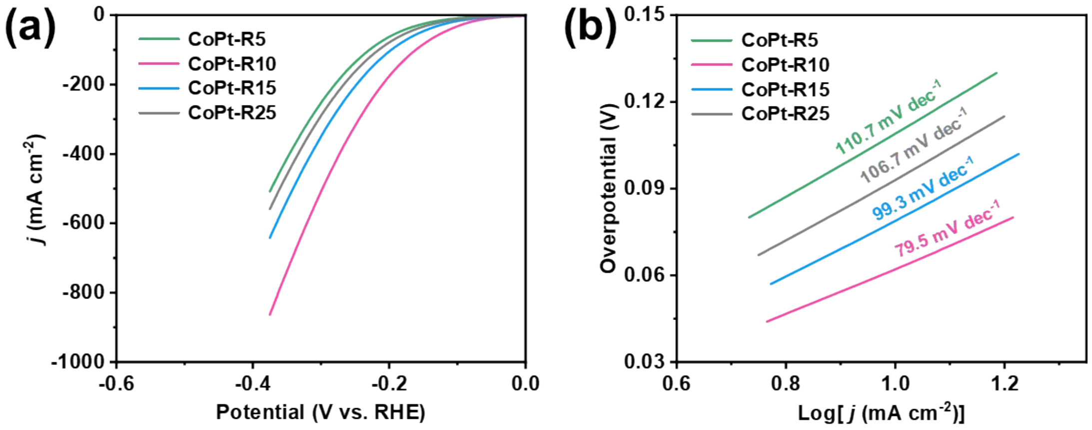

Fig. S11 HER performance: (a) LSV curves and (b) Tafel plots of CoPt-R5, 10, 15, 25.

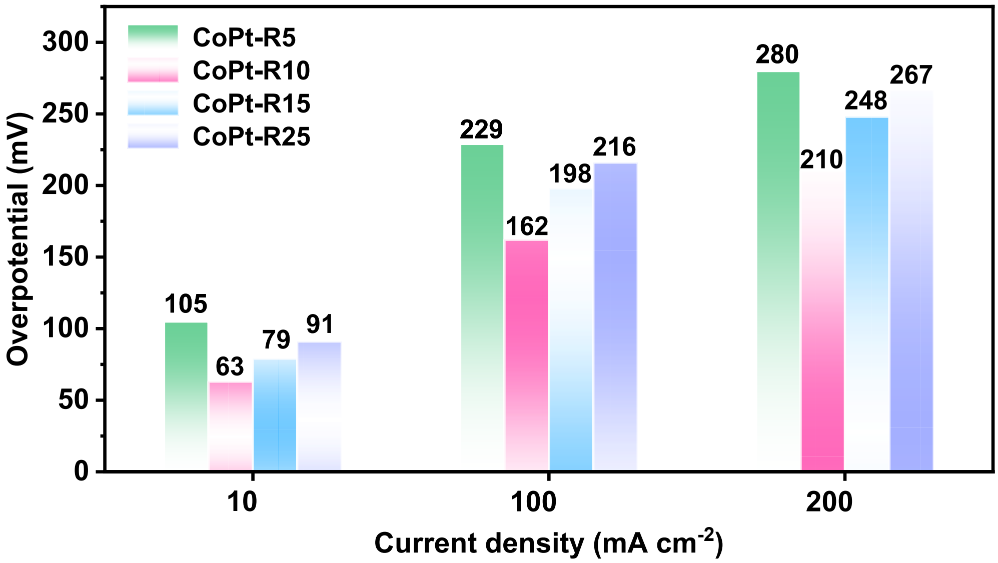

Fig. S12 Overpotentials at 10, 100 and 200 mA cm^{-2} of CoPt-R5, 10, 15, 25 for HER.

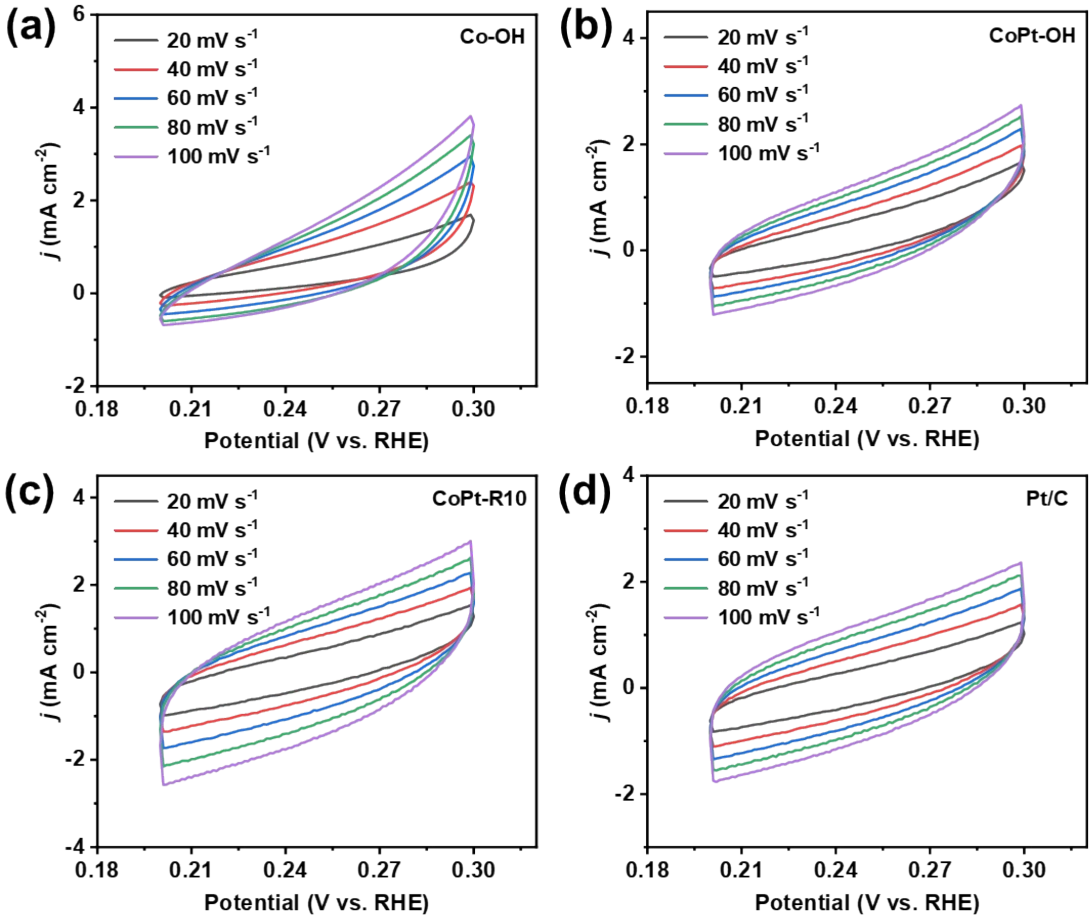

Fig. S13 Cyclic voltammetric profiles of (a) Co-OH, (b) CoPt-OH, (c) CoPt-R10 and (d) Pt/C measured at different scan rates (20 mV s^{-1} ~ 100 mV s^{-1}) with different scan rates in 1.0 M KOH.

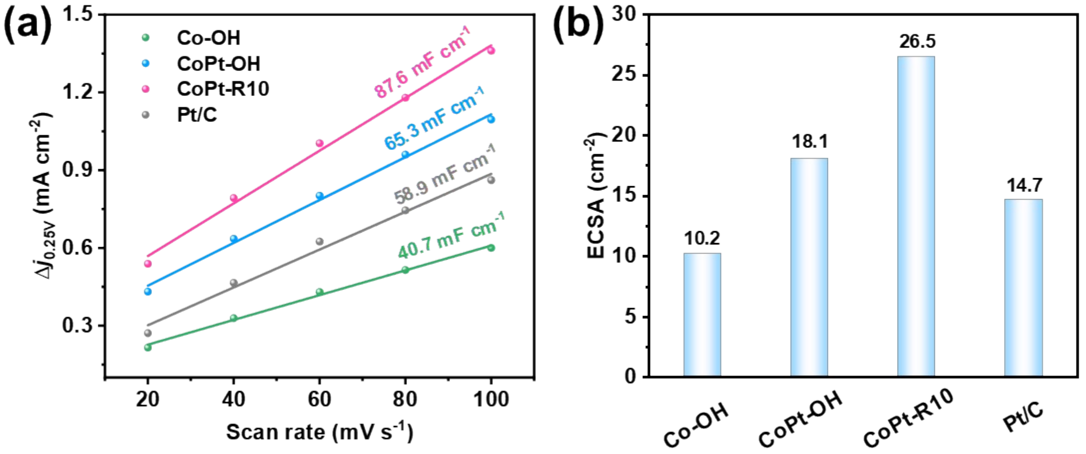

Fig. S14 (a) The double layer capacitance and (b) ECSA of Co-OH, CoPt-OH, CoPt-R10 and Pt/C.

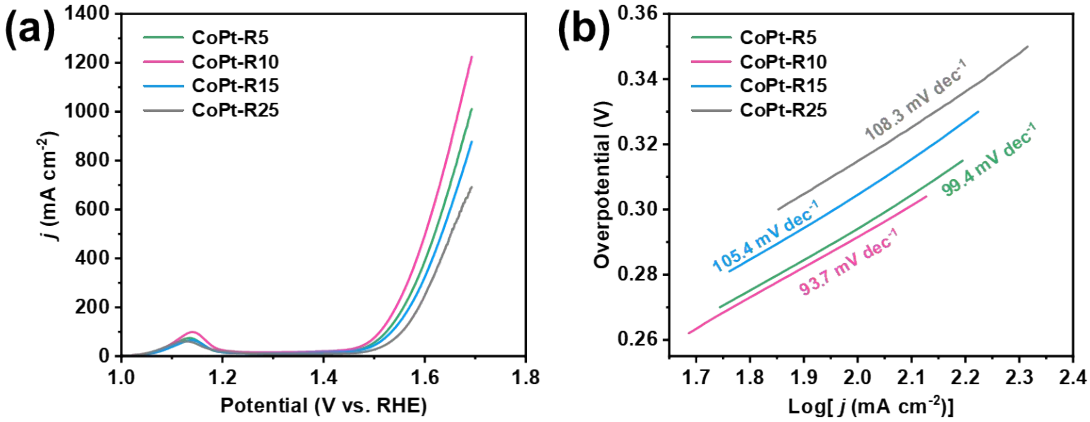

Fig. S15 OER performance: (a) LSV curves and (b) Tafel plots of CoPt-R5, 10, 15, 25.

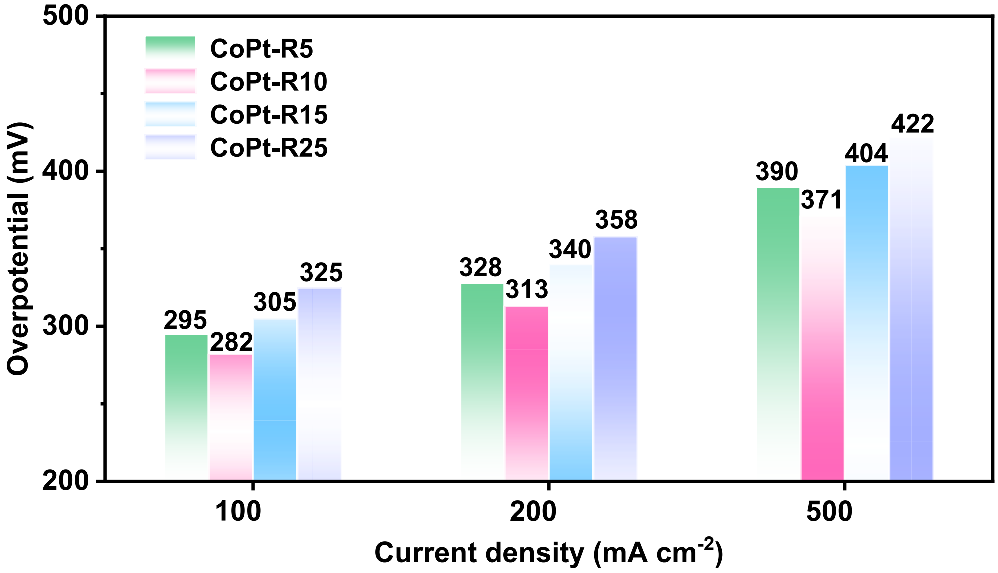

Fig. S16 Overpotentials at 100, 200 and 500 mA cm^{-2} of CoPt-R5, 10, 15, 25 for OER.

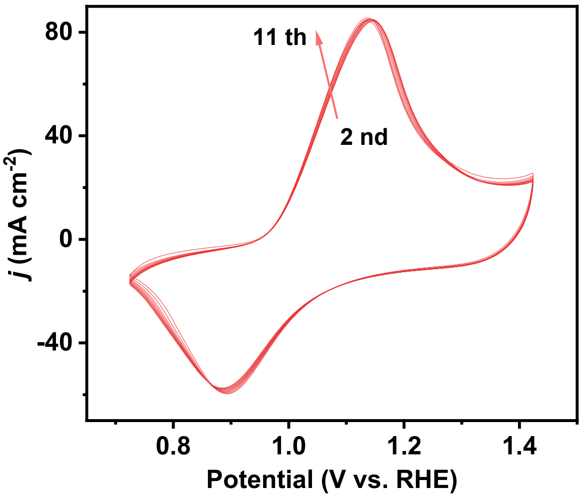

Fig. S17 CV curves of CoPt-R10 in the range of 0.72 to 1.42 V vs. RHE.

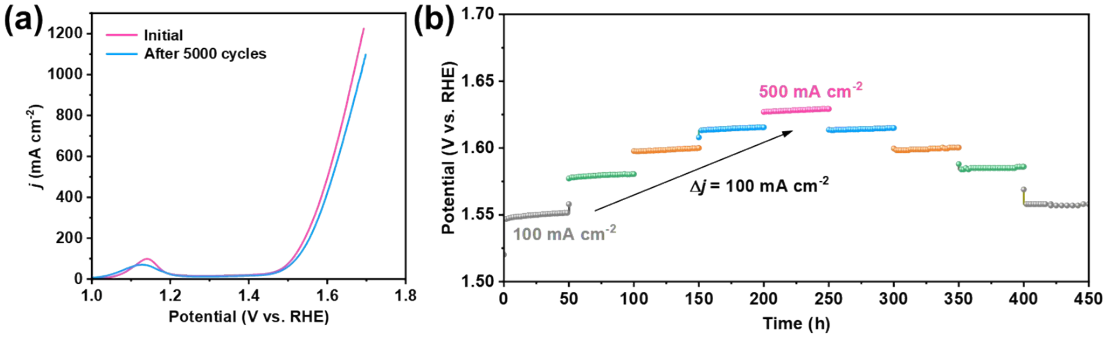

Fig. S18 (a) Polarization curves of CoPt-R10 for the OER before and after 5000 CV cycles. (b) The multicurrent step stability plots of CoPt-R10 for 450 hours.

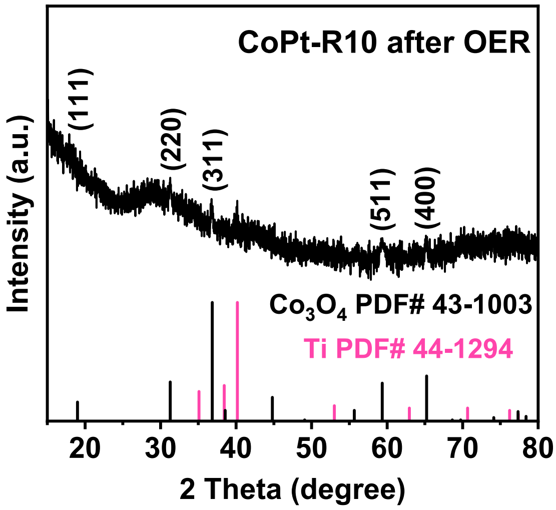

Fig. S19 XRD pattern of CoPt-R10 after stability test.

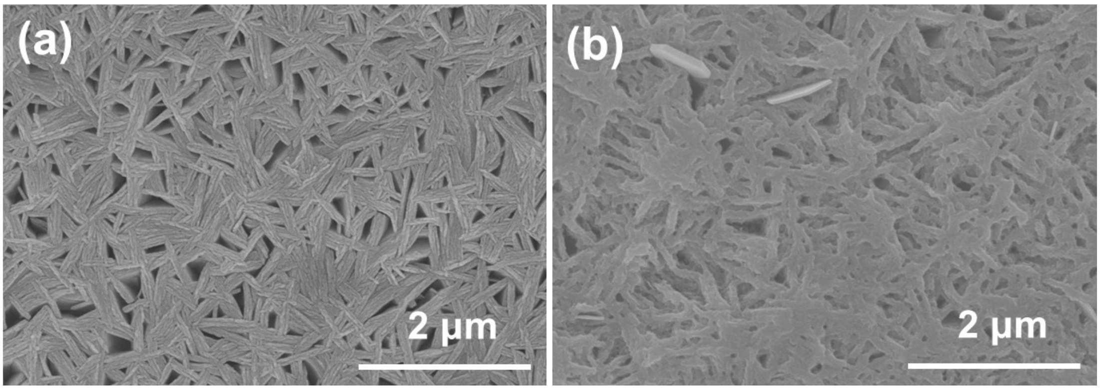

Fig. S20 SEM images of (a) CoPt-R10 and (b) CoPt-R10 after stability test for OER.

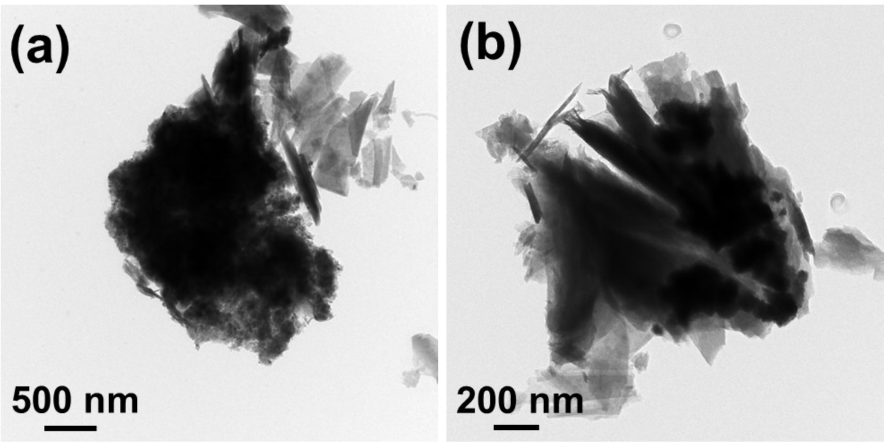

Fig. S21 TEM images of (a) CoPt-R10 and (b) CoPt-R10 after stability test for OER.

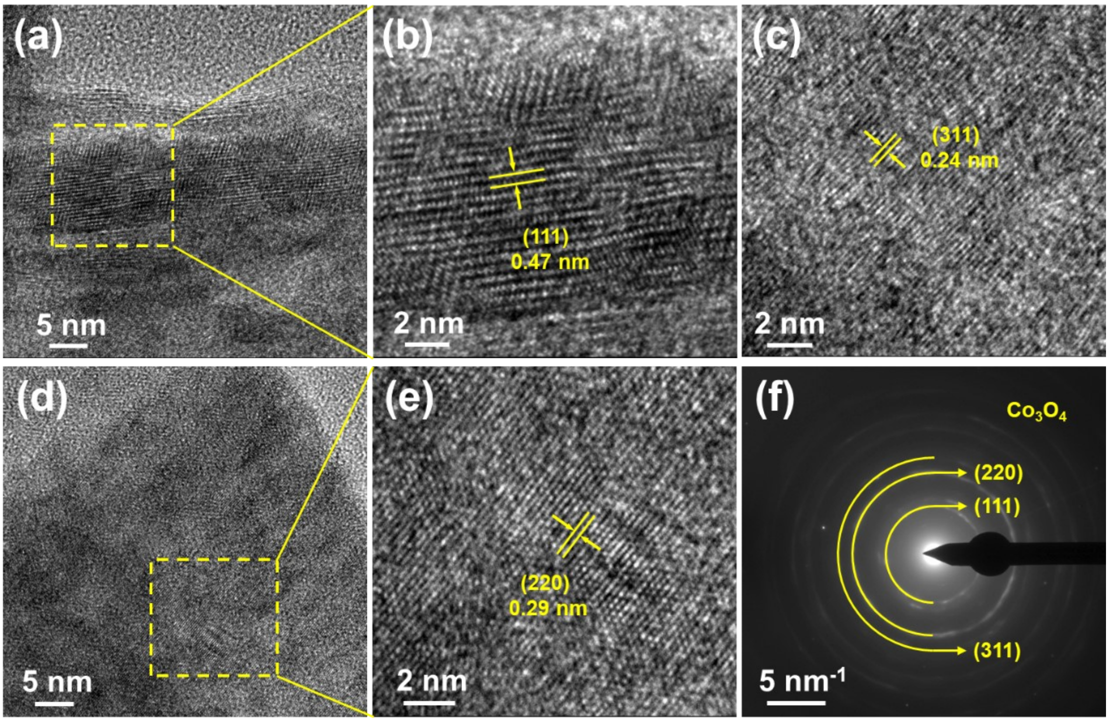

Fig. S22 (a-e) HRTEM images, and (f) SAED pattern of CoPt-R10 after stability test.

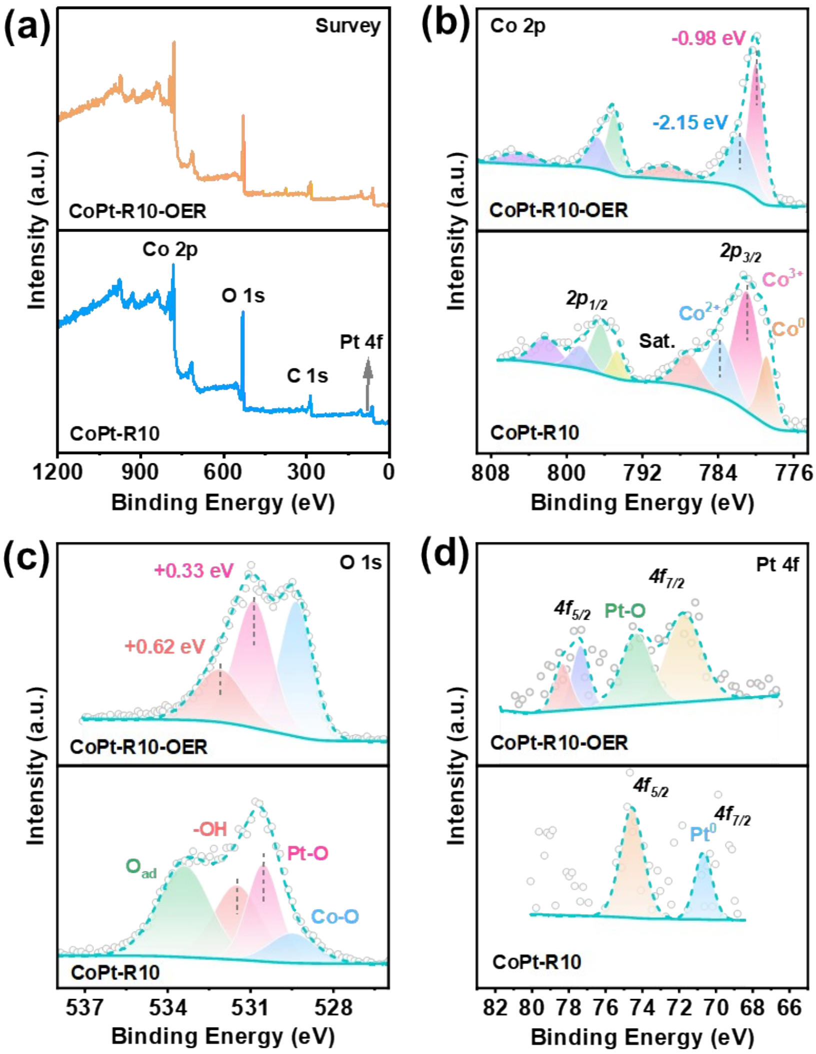

Fig. S23 (a) The XPS survey and the high-resolution spectra of (b) Co 2p, (c) O 1s and (d) Pt 4f of CoPt-R10 after OER test.

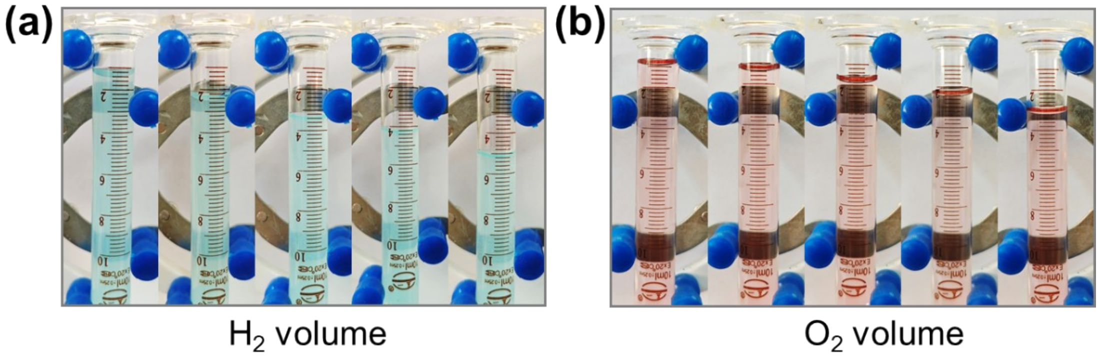

Fig. S24 Digital photographs of (a) hydrogen and (b) oxygen collected per 15 minutes.

Table S1. The amount of Co and Pt determined by ICP for CoPt-R10 initial and after long-term testing.

Element	Initial (*wt.* %)	After long-term test (*wt.* %)	Loss amount (*wt.* %)
Co	56.79	47.36	13.03
Pt	2.79	1.58	1.21

Table S2. Overall water splitting performance comparison of CoPt-R10 with recently reported catalysts in 1.0 M KOH.

Catalyst	Electrolyte	*j* (mA cm^{-2})	*E* (V)	Ref.
NiFeP/NM	1.0 M KOH	100	1.63	5
def-Ru-NiFe LDH/NCO	1.0 M KOH	100	2.05	6
CoP@CoNi LDH-Pt||CoP@CoNi LDH	1.0 M KOH	100	1.67	7
NiCoMn/NF	1.0 M KOH	100	1.75	8
NiMoFe HI	1.0 M KOH	100	1.677	9
FeNiMo NFs||Pt/C	1.0 M KOH	100	1.73	10
Ru–Co_2Ni@NCNT	1.0 M KOH	100	1.76	11
Lnp-NCM4	1.0 M KOH	100	1.75	12
Cu-NiCo LDH/NiCo@CC	1.0 M KOH	100	2.0	13
CoPt-R10	1.0 M KOH	100	1.60	This work

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