Supplementary information

The Mott–Schottky Co_2P/Co heterocatalyst encapsulated N,P-doped graphene/carbon nanotubes as high-efficiency trifunctional electrocatalysts for cable-type flexible Zn–air batteries and water splitting

Quynh Phuong Ngo, Thanh Tuan Nguyen, Manjinder Singh, Nam Hoon Kim, Joong Hee Lee**

E-mail: nhk@jbnu.ac.kr, jhl@jbnu.ac.kr

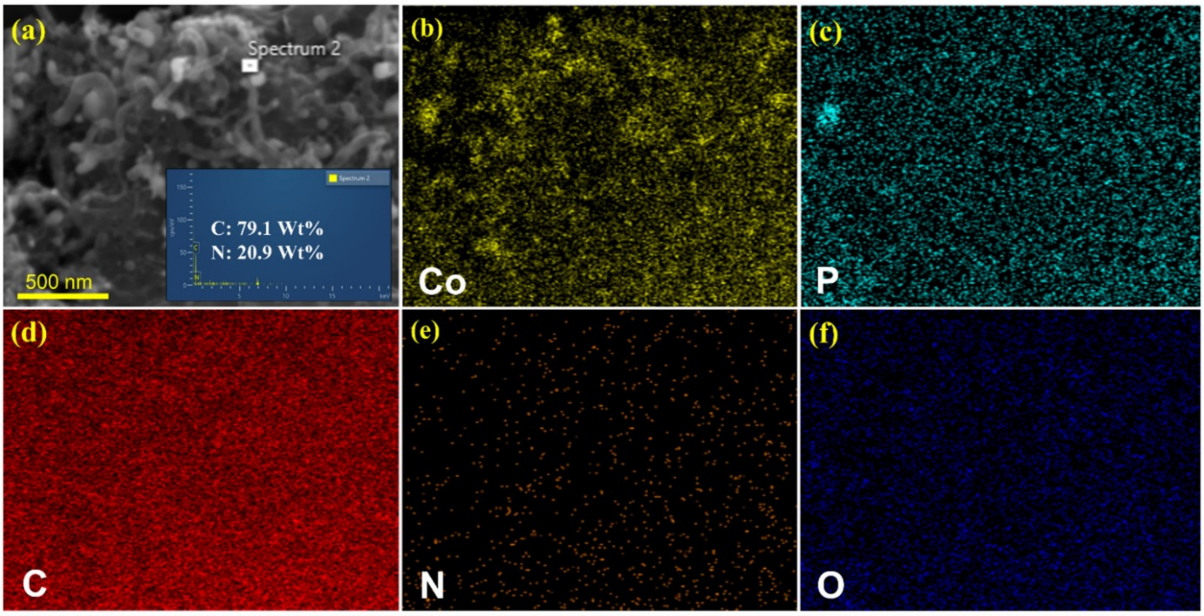

Fig. S1. (a) Overview FE-SEM and EDS color mapping of (b) Co, (c) P, (d) C, (e) N, (f) O elements and spectrum focus to N–CNT of Co_2P/Co@N–CNT/NPG nanohybrid.

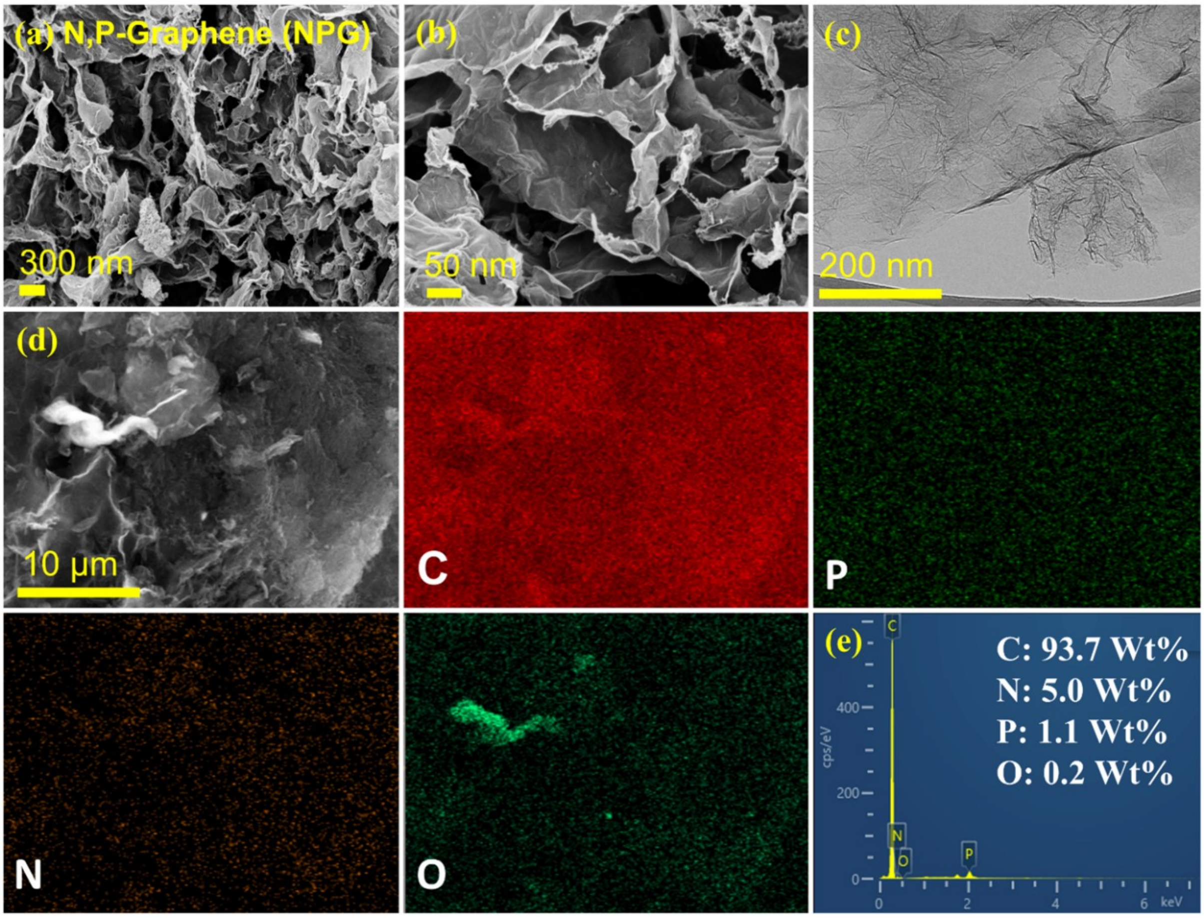

Fig. S2. (a,b) FE-SEM images, (c) TEM image, (d) Corresponding color mapping images of C, P, N, O elements, and (e) EDS of NPG.

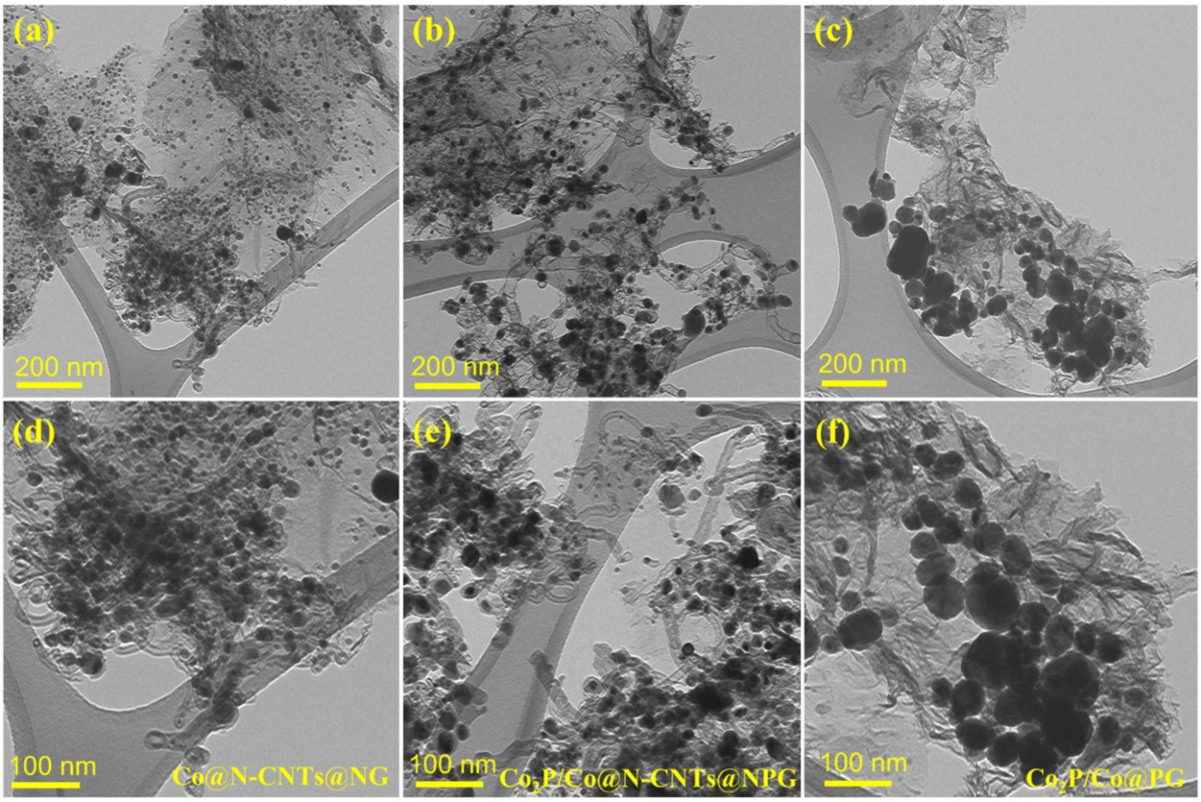

Fig. S3. TEM images of (a,d) Co@N–CNT/NG, (b,e) Co_2P/Co@N–CNT/NPG, and (c,f) Co_2P/Co@PG.

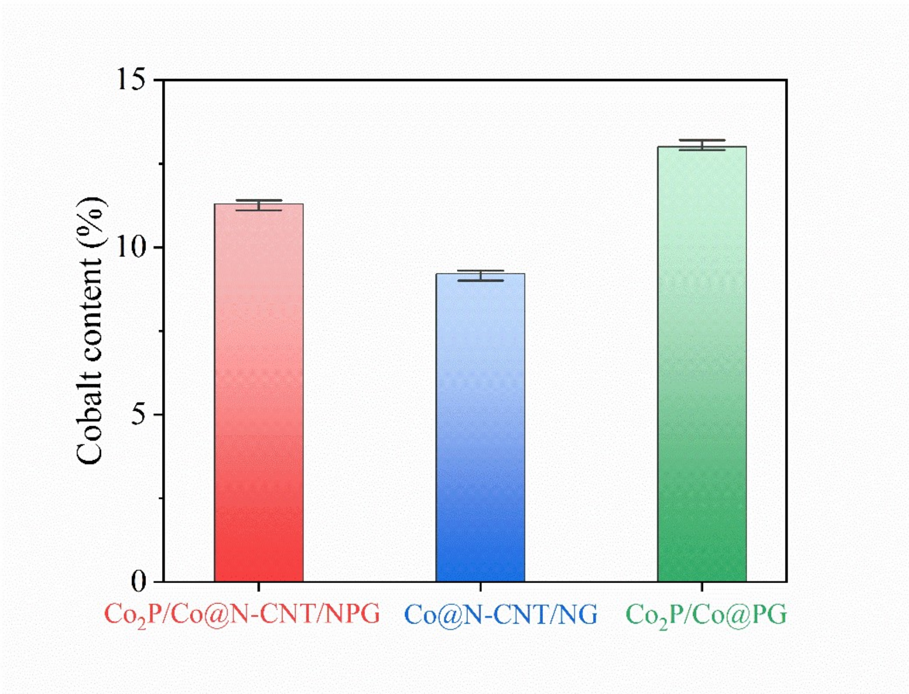

Fig. S4. The ICP−AES result of Co_2P/Co@N–CNT/NPG, Co@N–CNT/NG, and Co_2P/Co@PG.

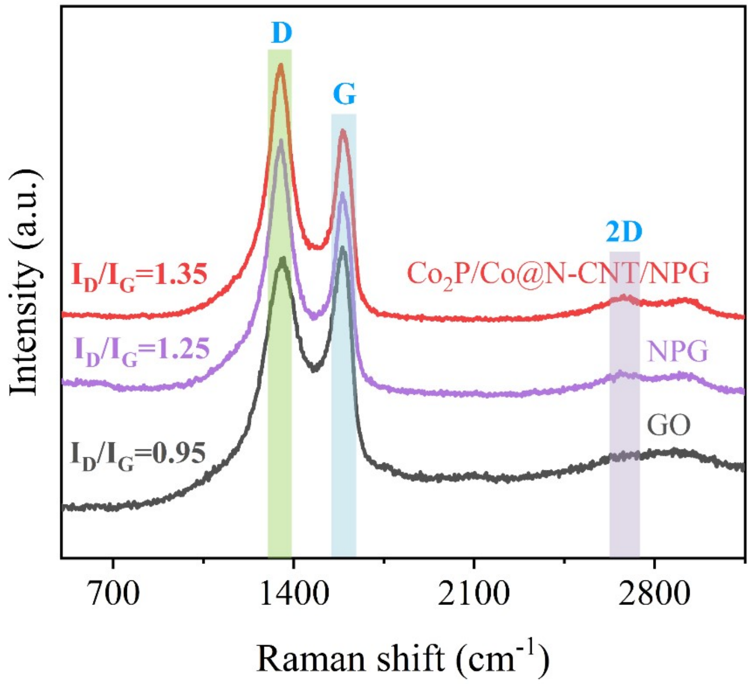

Fig. S5. Raman spectrum of Co_2P/Co@N–CNT/NPG, NPG, and GO.

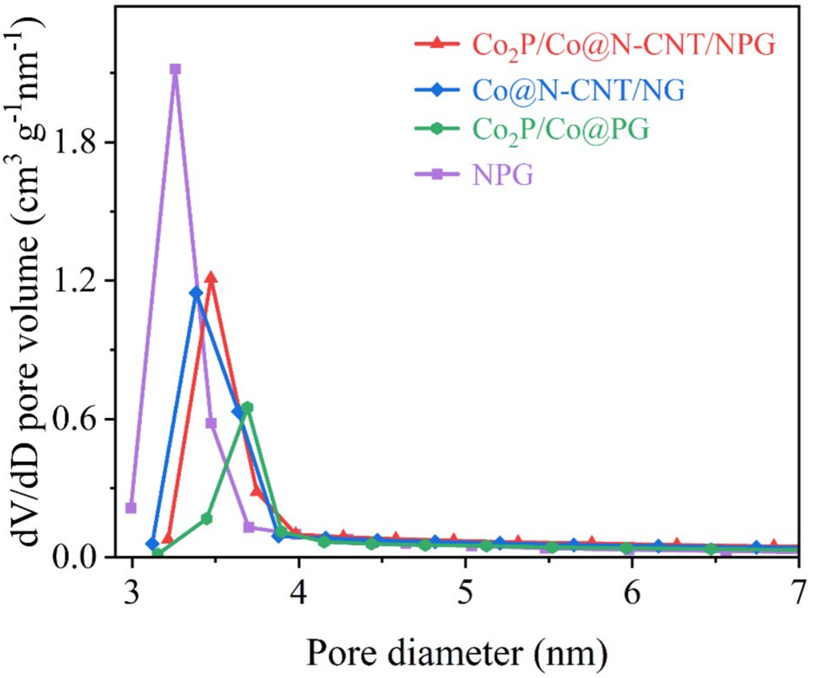

Fig. S6. Pore size distribution diagram of Co_2P/Co@N–CNT/NPG, Co@N–CNT/NG, Co_2P/Co@PG and NPG.

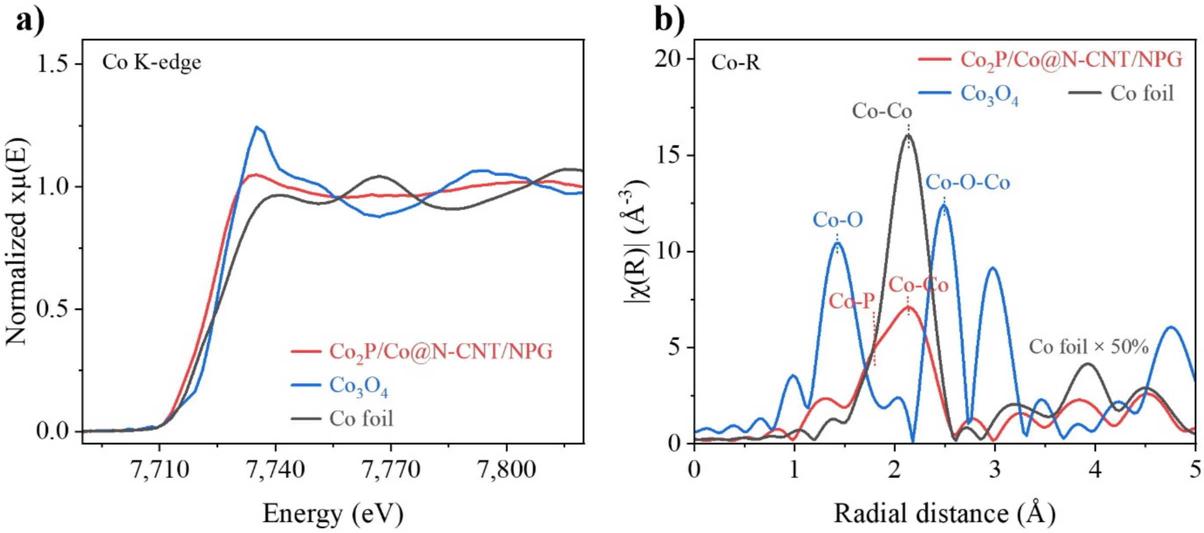

Fig. S7. XAS characterization. a) Co K-edge XANES curves and b) Fourier-transformed EXAFS spectra at the Co K-edge of Co_2P/Co@N-CNT/NPG, Co_3O_4, and Co foil reference.

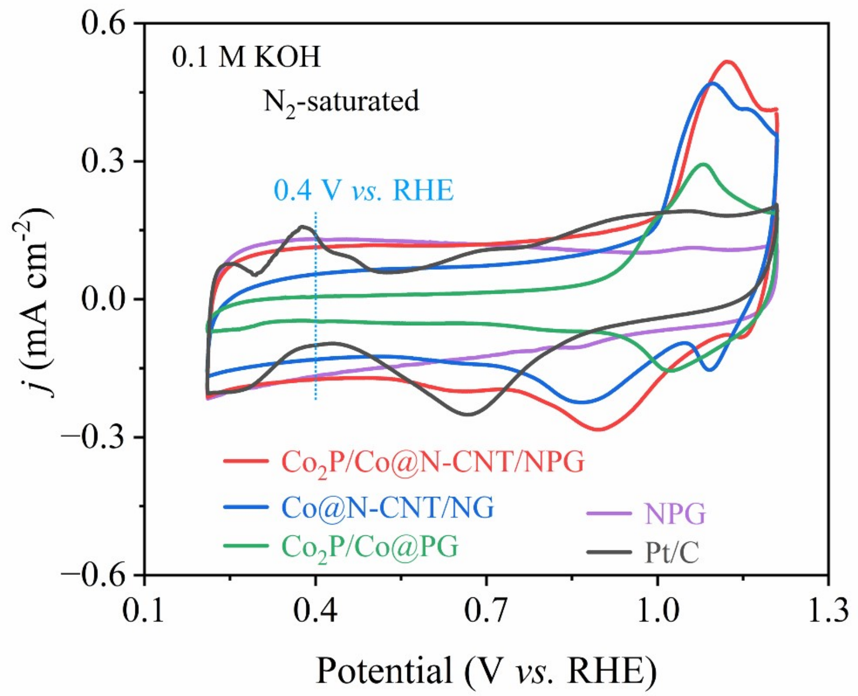

Fig. S8. The CV profiles of the synthesized materials at sweep rate of 5 mV s^{-1} in N_2-saturated 0.1 M KOH solution.

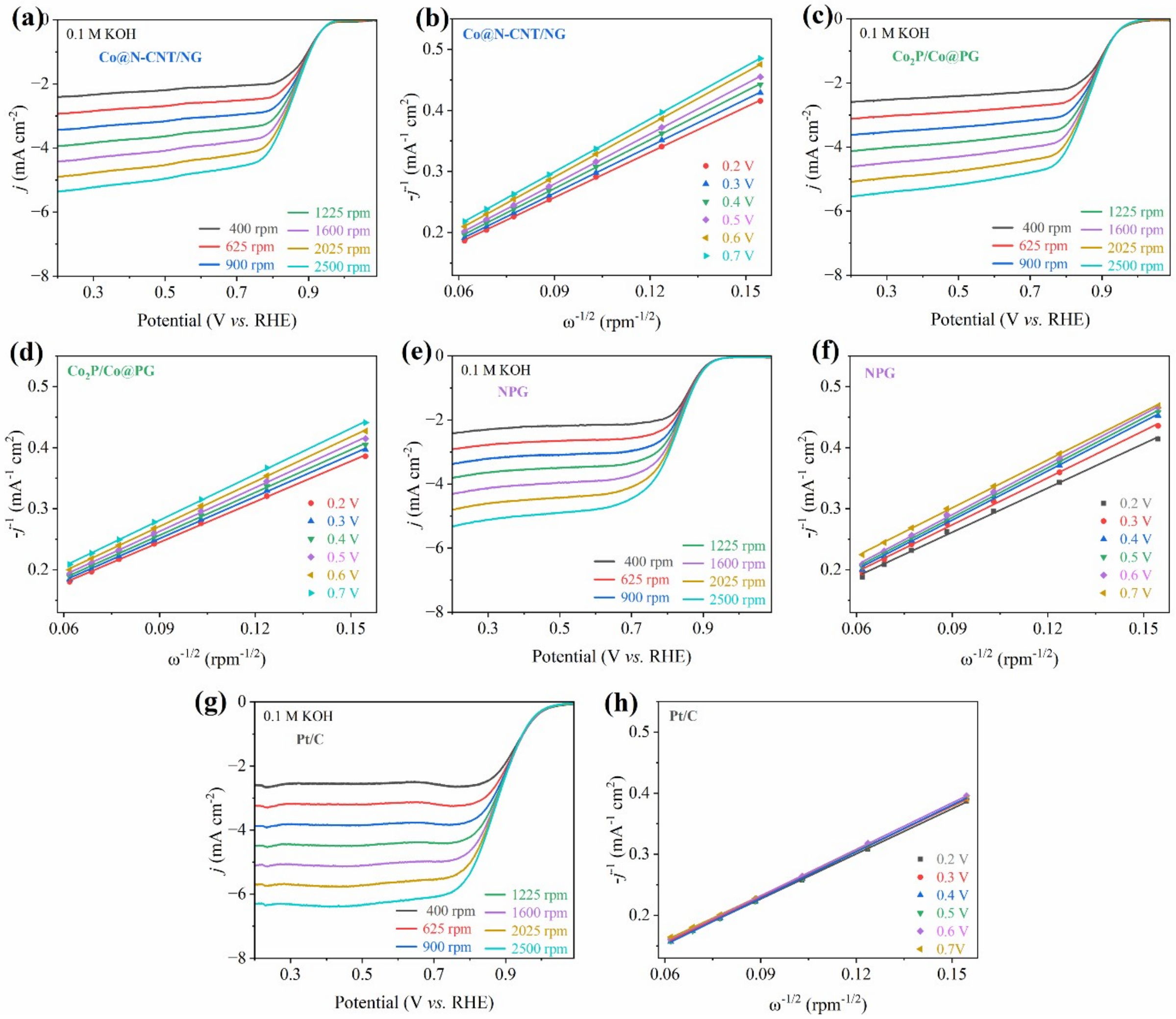

Fig. S9. LSV responses at various electrode rotation speeds and corresponding K-L plots in 0.1 M KOH aqueous solution with RDE: (a,b) Co@N–CNT/NG, (c,d) Co_2P/Co@PG, (e,f) NPG, and (g,h) Pt/C.

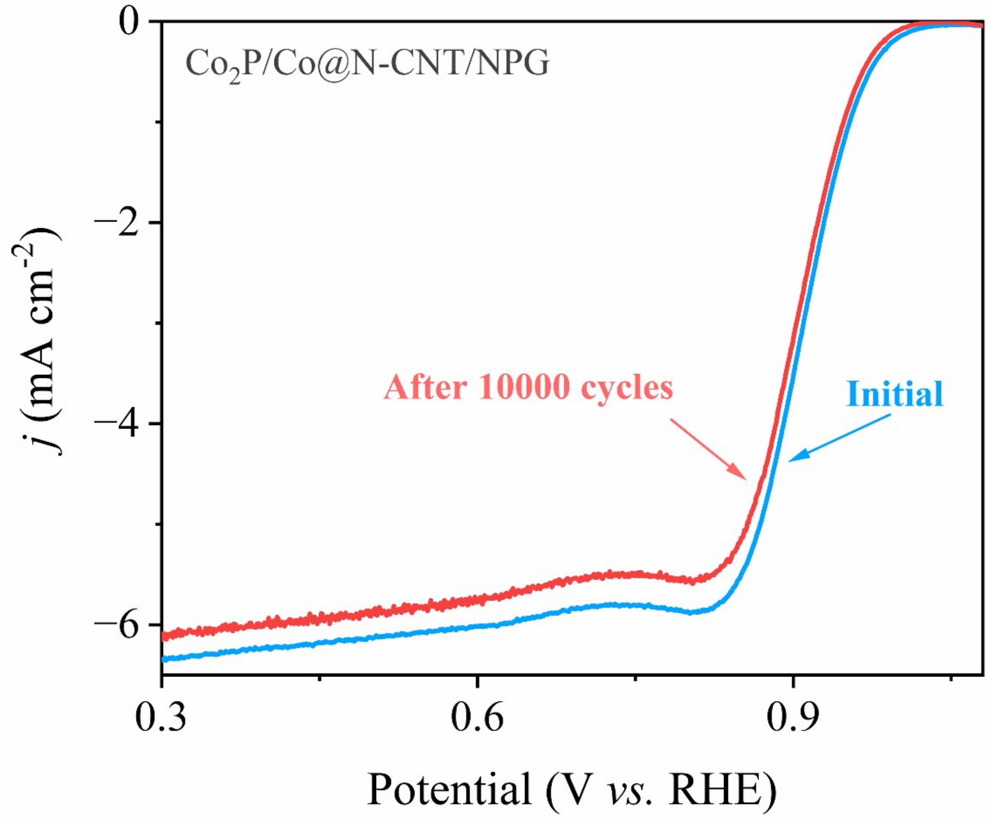

Fig. S10. ORR LSV polarization curves of Co_2P/Co@N–CNT/NPG before and after 10,000 cycles.

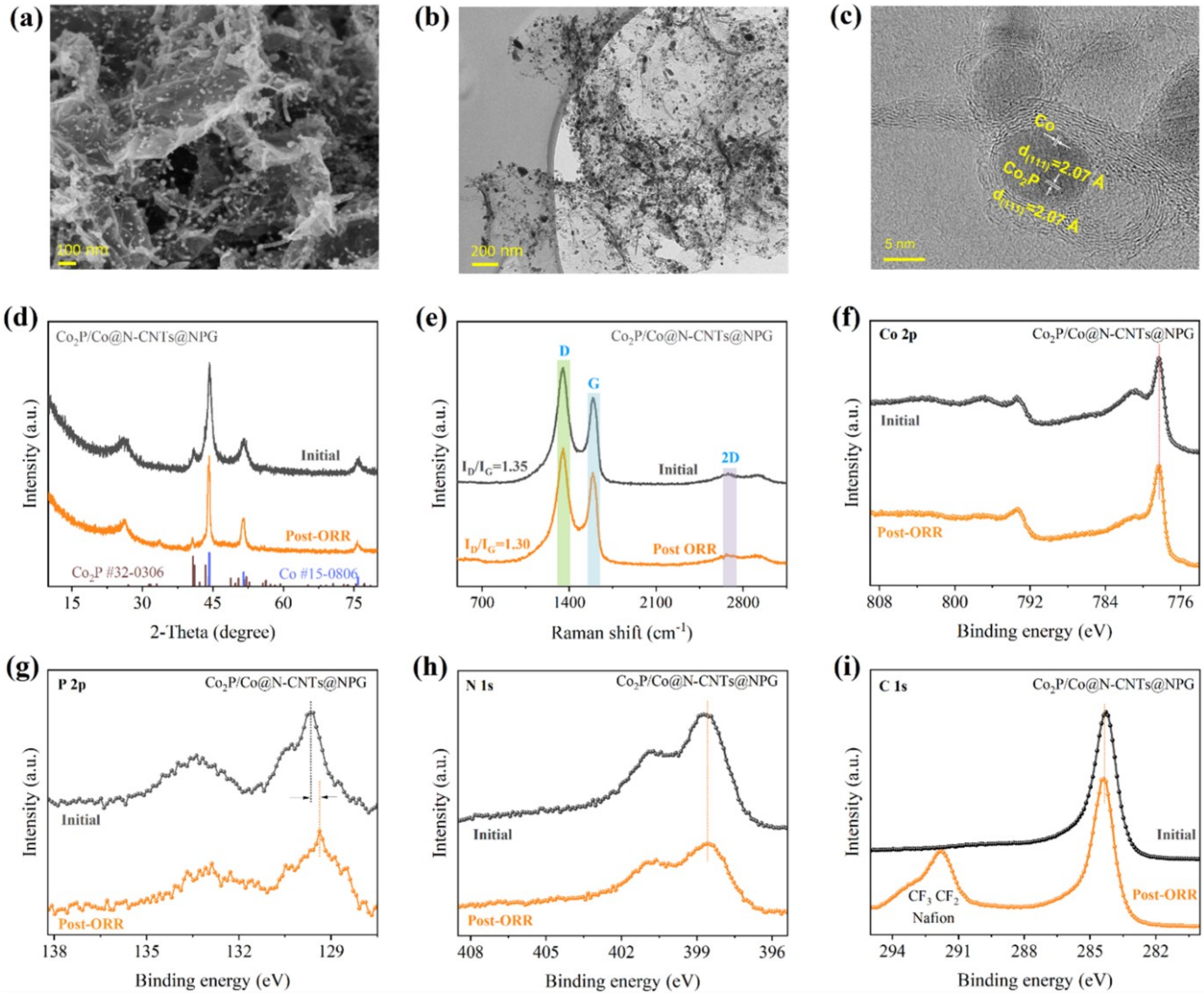

Fig. S11. (a) SEM and (b, c) TEM images at different magnification, (d) XRD patterns, (e) Raman spectra and XPS survey spectrum with high-resolution XPS spectra for (f) Co 2p, (g) P 2p, (h) N 1s, (i) C 1s of Co_2P/Co@N–CNT/NPG after ORR performance.

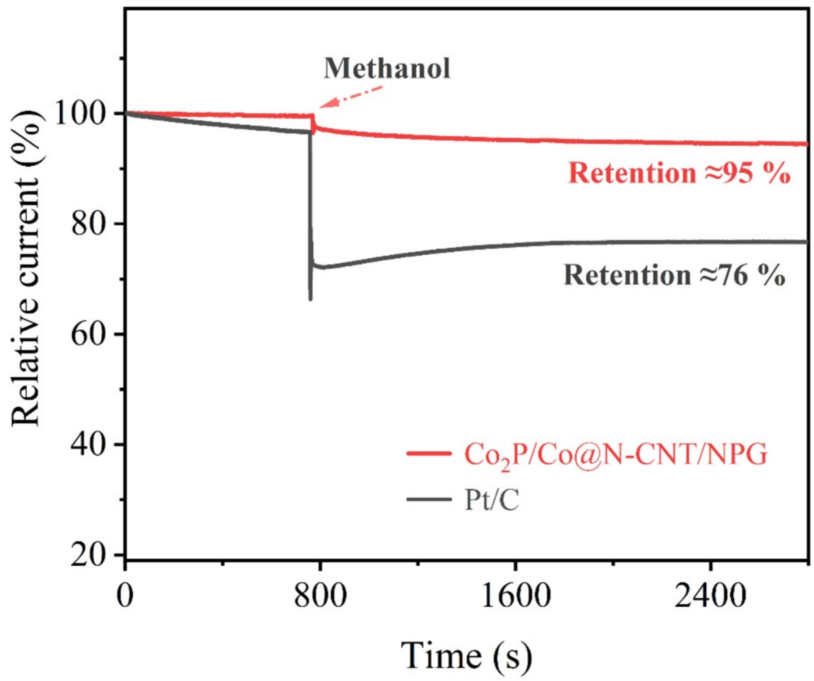

Fig. S12. The durability of Co_2P/Co@N–CNT/NPG and Pt/C under the crossover effect of 3 M methanol.

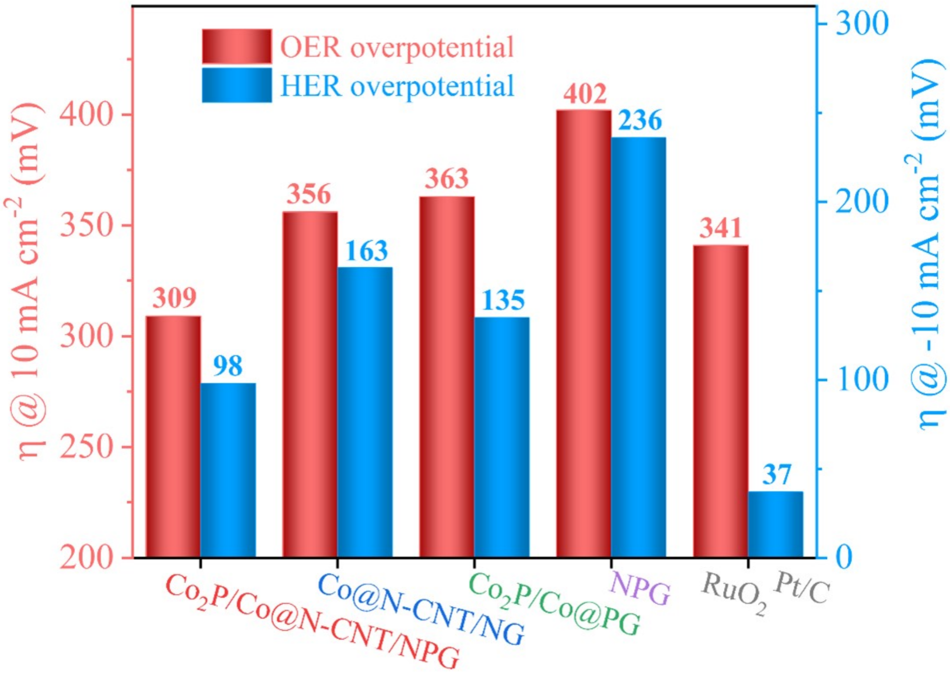

Fig. S13. The OER and HER overpotential comparison of Co_2P/Co@N–CNT/NPG, Co@N–CNT/NG, Co_2P/Co@PG, NPG, RuO_2 and Pt/C in 1 M KOH.

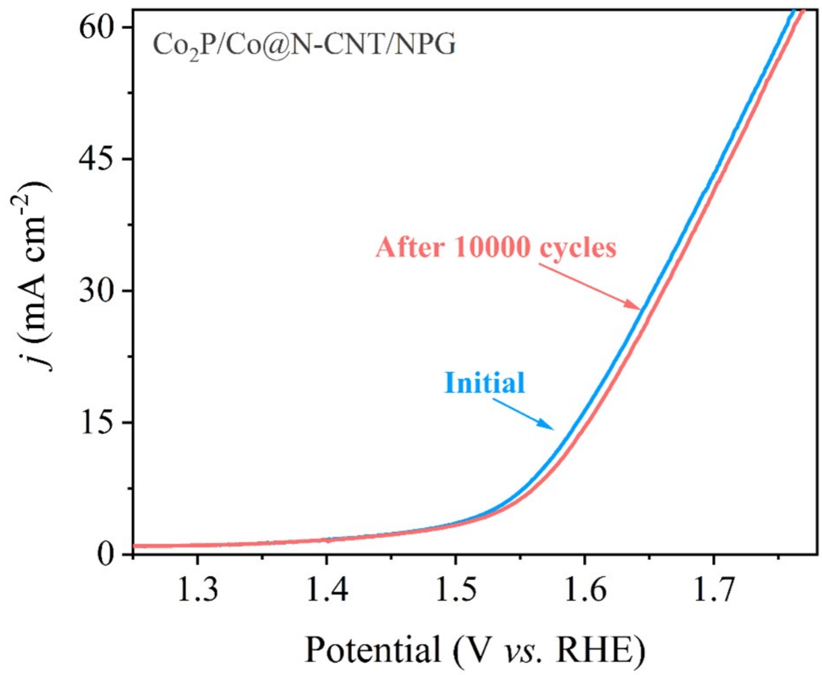

Fig. S14. OER LSV polarization curves of Co_2P/Co@N–CNT/NPG before and after 10,000 cycles.

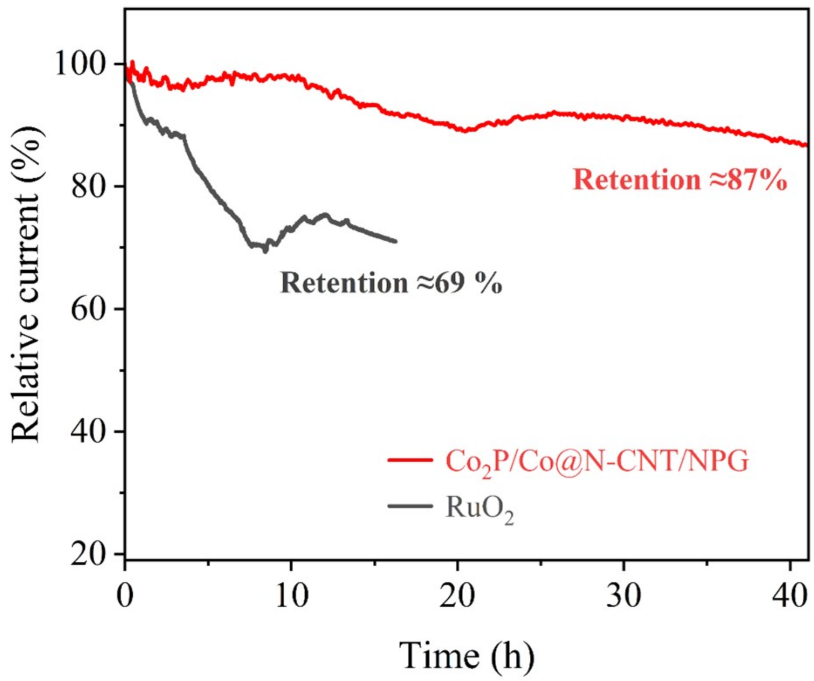

Fig. S15. Chronoamperometry stability of Co_2P/Co@N–CNT/NPG and RuO_2 for a long-term OER operation.

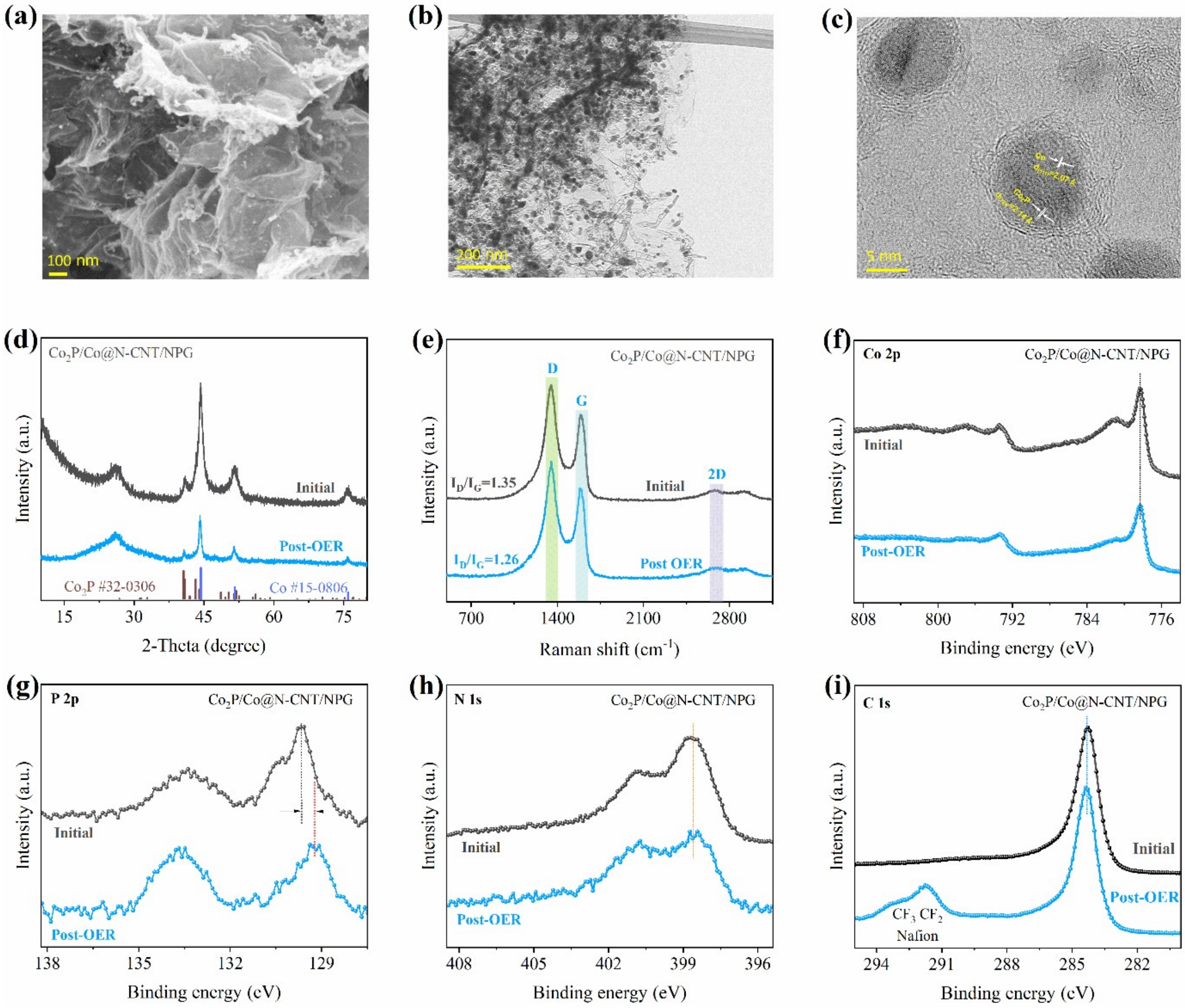

Fig. S16. (a) SEM and (b, c) TEM images at different magnification, (d) XRD patterns, (e) Raman spectra and XPS survey spectrum with high-resolution XPS spectra for (f) Co 2p, (g) P 2p, (h) N 1s, (i) C 1s of Co_2P/Co@N–CNT/NPG after OER performance.

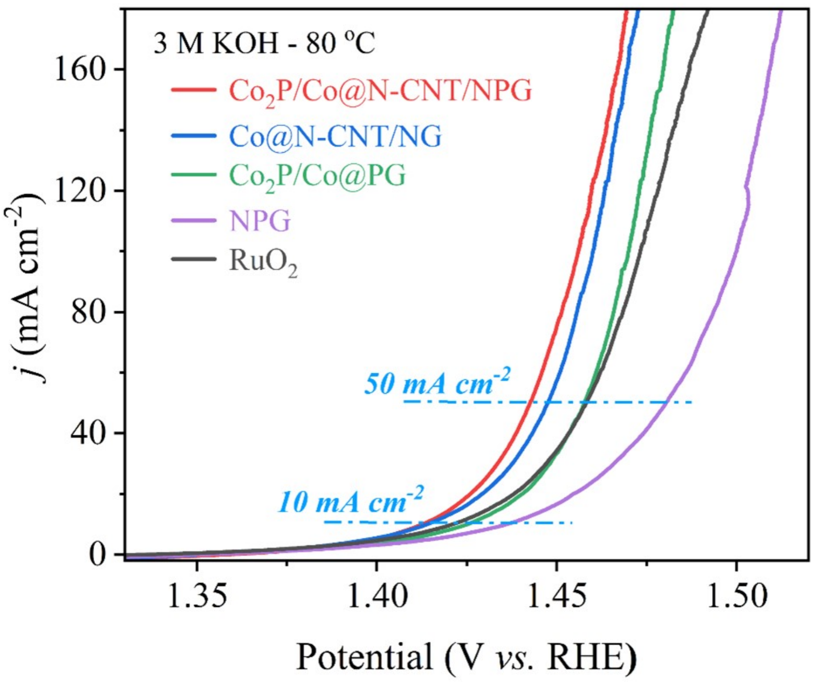

Fig. S17. OER LSV polarization curves of Co_2P/Co@N–CNT/NPG, Co@N–CNT/NG, Co_2P/Co@PG, NPG and RuO_2 in 3 M KOH at 80 °C.

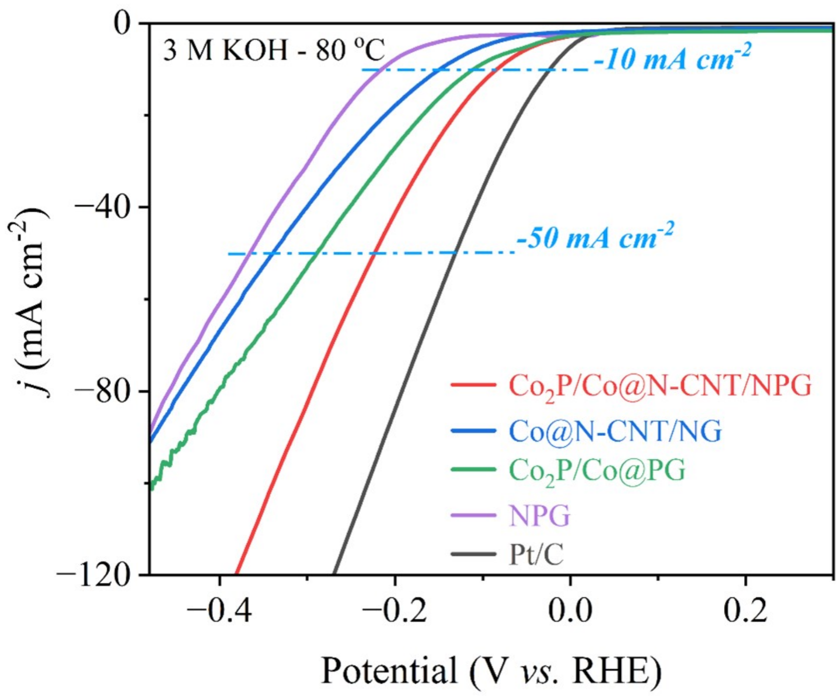

Fig. S18. HER LSV polarization curves of Co_2P/Co@N–CNT/NPG, Co@N–CNT/NG, Co_2P/Co@PG, NPG and RuO_2 in 3 M KOH at 80 °C.

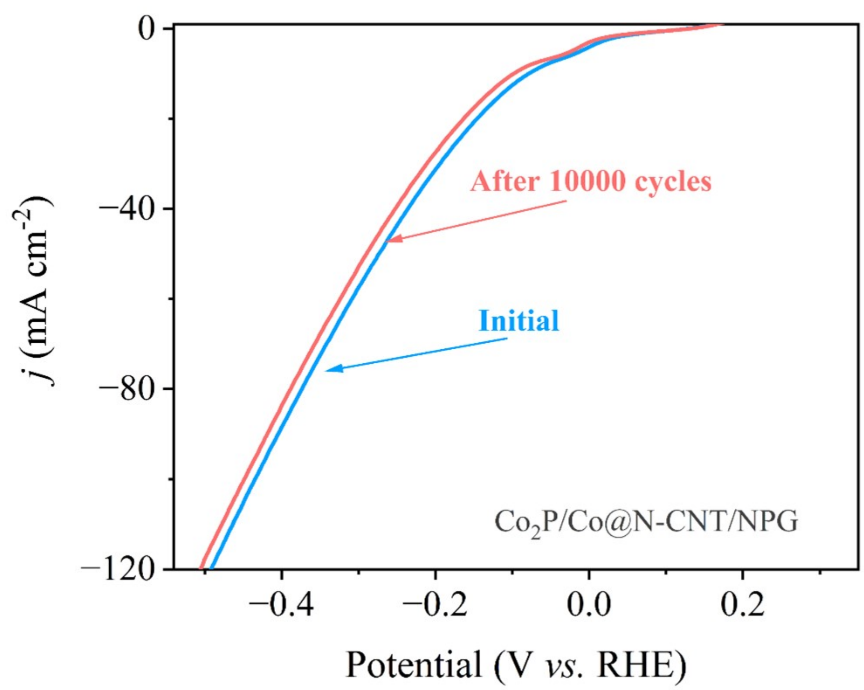

Fig. S19. HER LSV polarization curves of Co_2P/Co@N–CNT/NPG before and after 10,000 cycles.

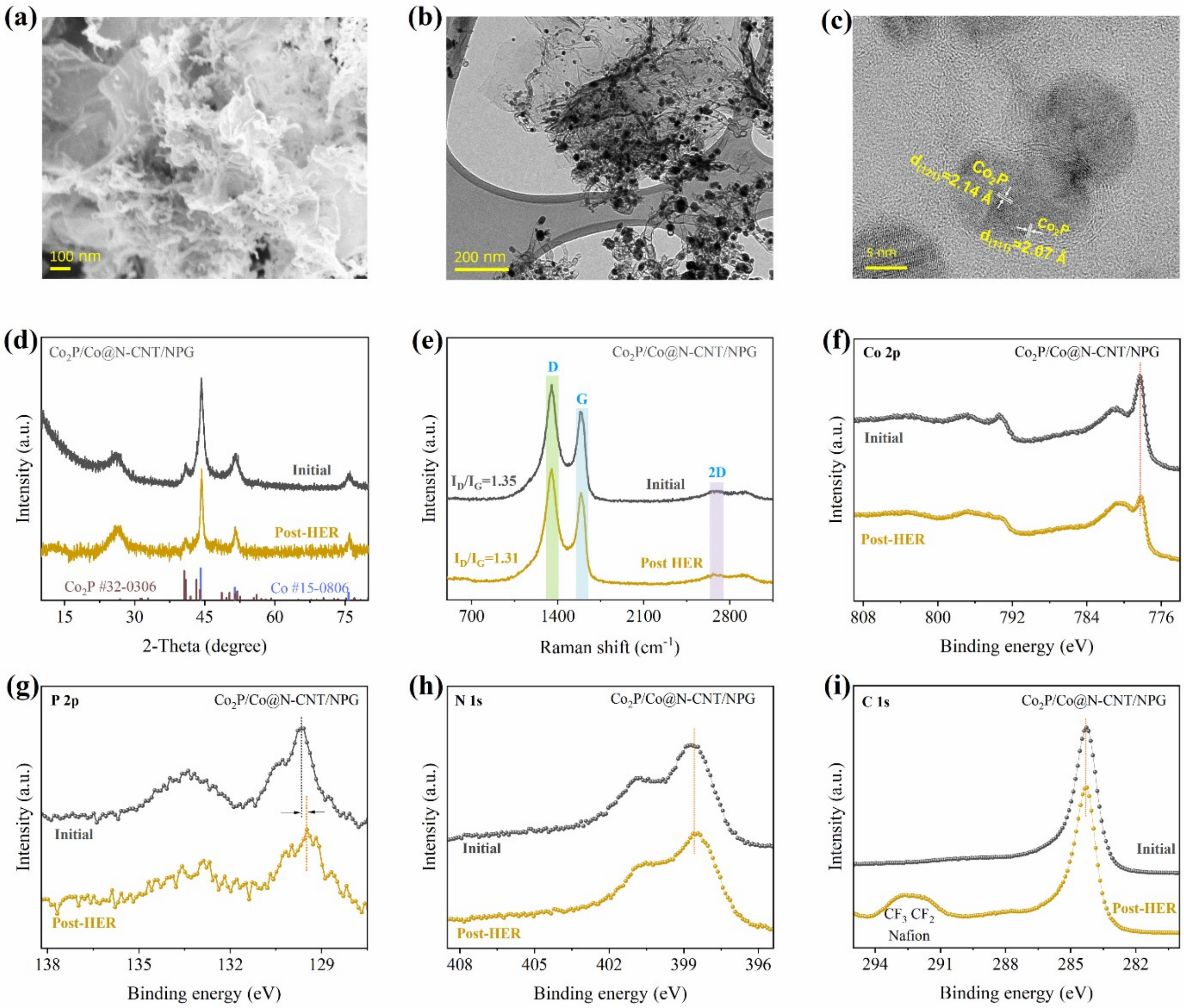

Fig. S20. (a) SEM and (b, c) TEM images at different magnification, (d) XRD patterns, (e) Raman spectra and XPS survey spectrum with high-resolution XPS spectra for (f) Co 2p, (g) P 2p, (h) N 1s, (i) C 1s of Co_2P/Co@N–CNT/NPG after HER performance.

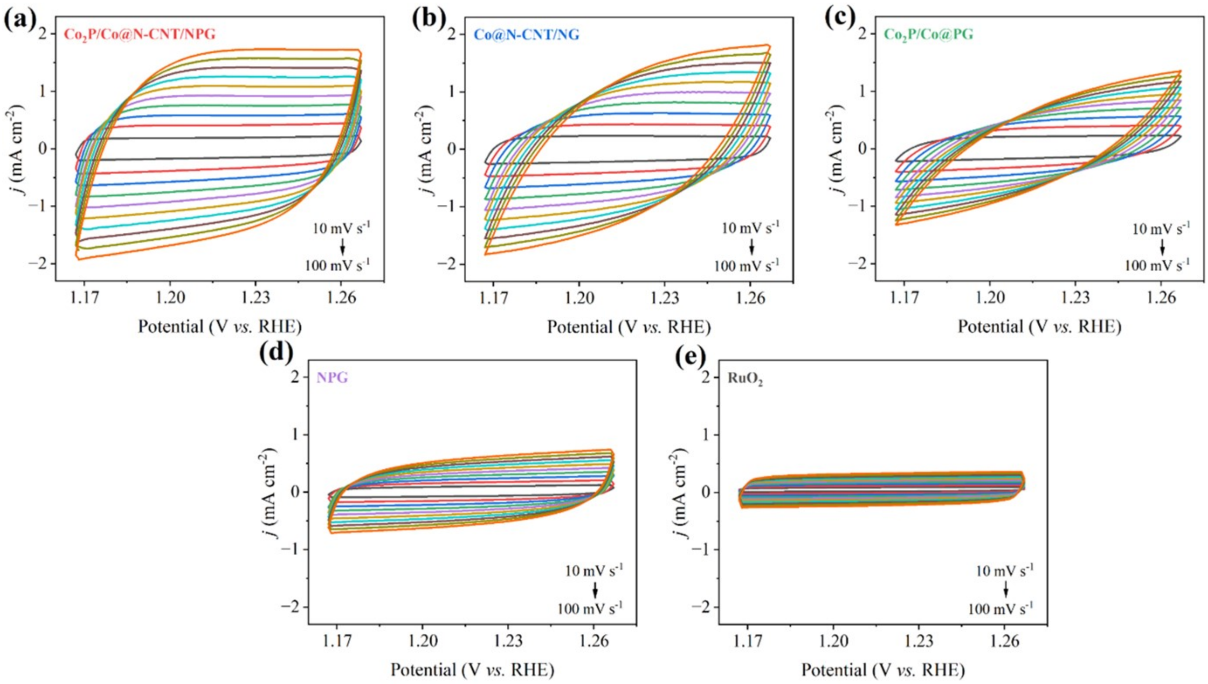

Fig. S21. CV curves of catalysts at various scan rates in 1 M KOH aqueous solution: (a) Co_2P/Co@N–CNT/NPG, (b) Co@N–CNT/NG, (c) Co_2P/Co@PG, (d) NPG, and (e) RuO_2.

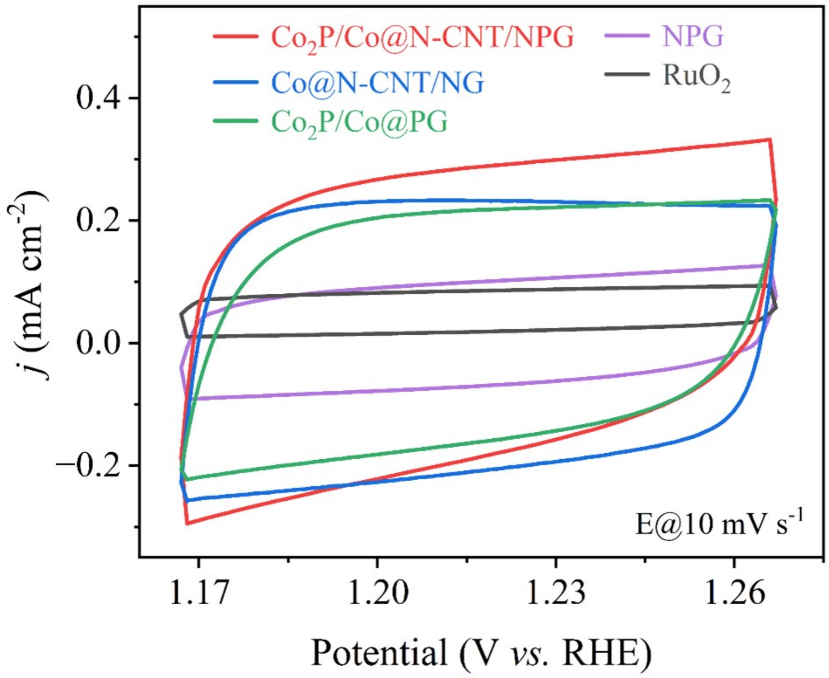

Fig. S22. CV measurement of different synthesized catalysts at a sweep rate of 10 mV s^{-1}.

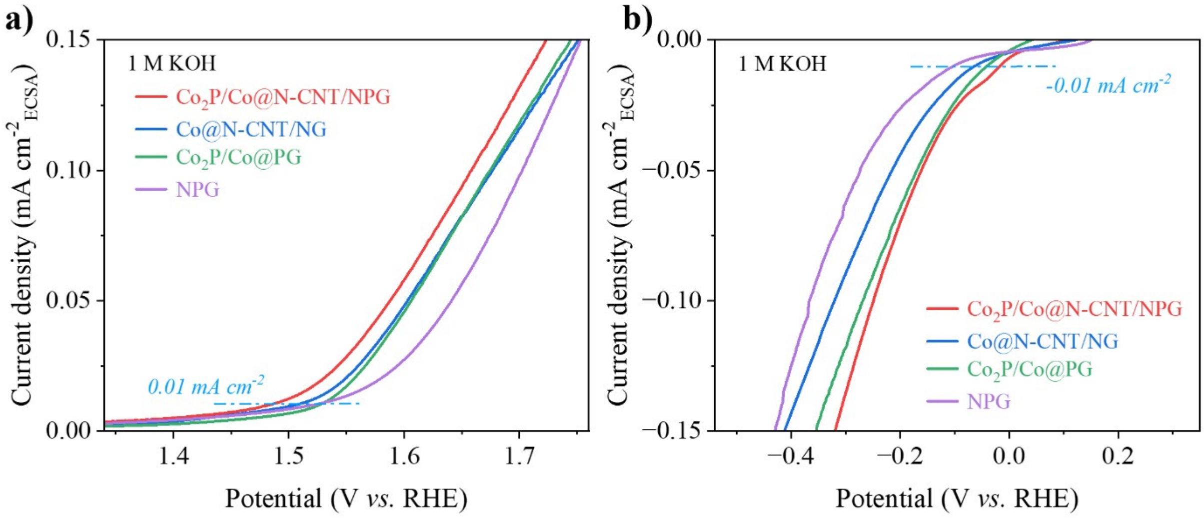

Fig. S23. ECSA-normalized LSV curves of as-prepared electrocatalysts for a) OER and b) HER.

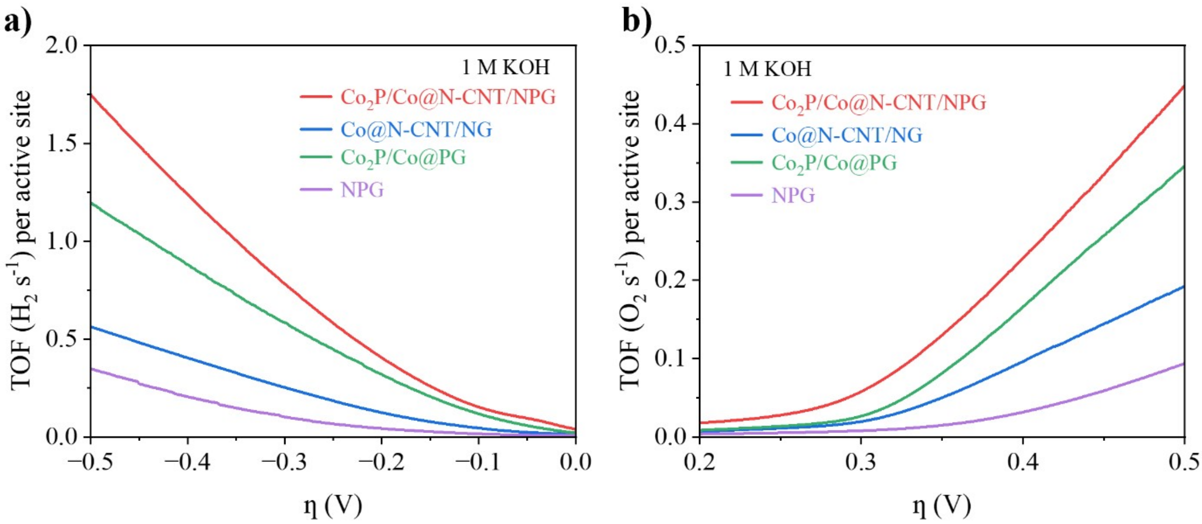

Fig. S24. TOF of as-prepared electrocatalysts for a) HER and b) OER.

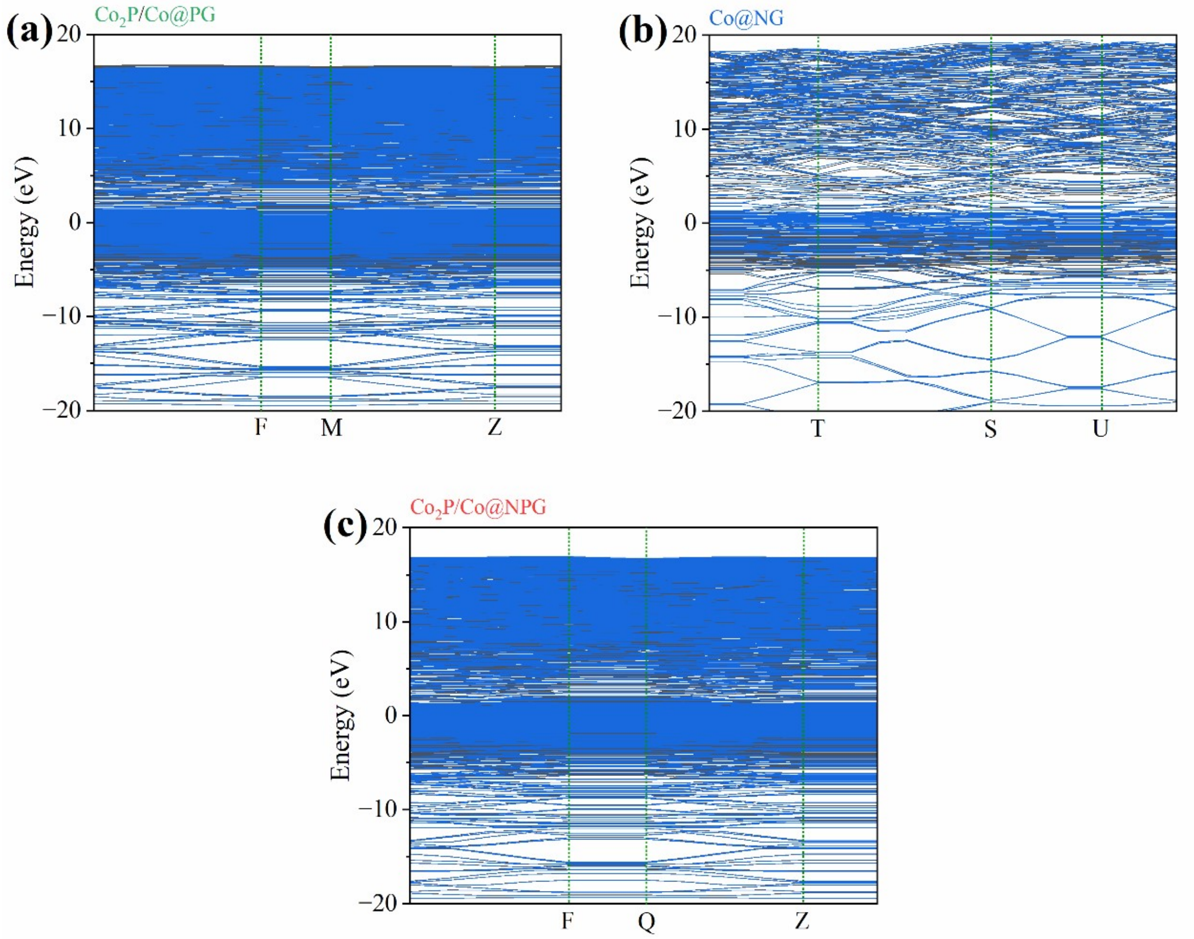

Fig. S25. The electronic band structure of Co_2P/Co@PG, Co@NG, and Co_2P/Co@NPG.

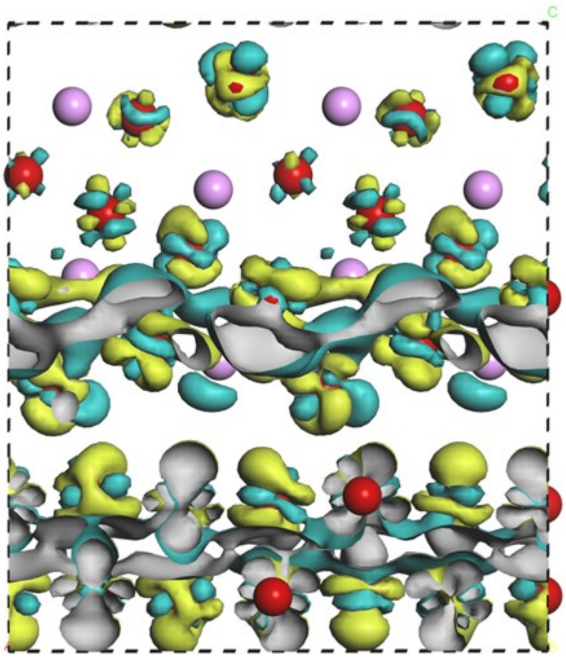

Fig. S26. Energy density difference calculation for Co_2P/Co model.

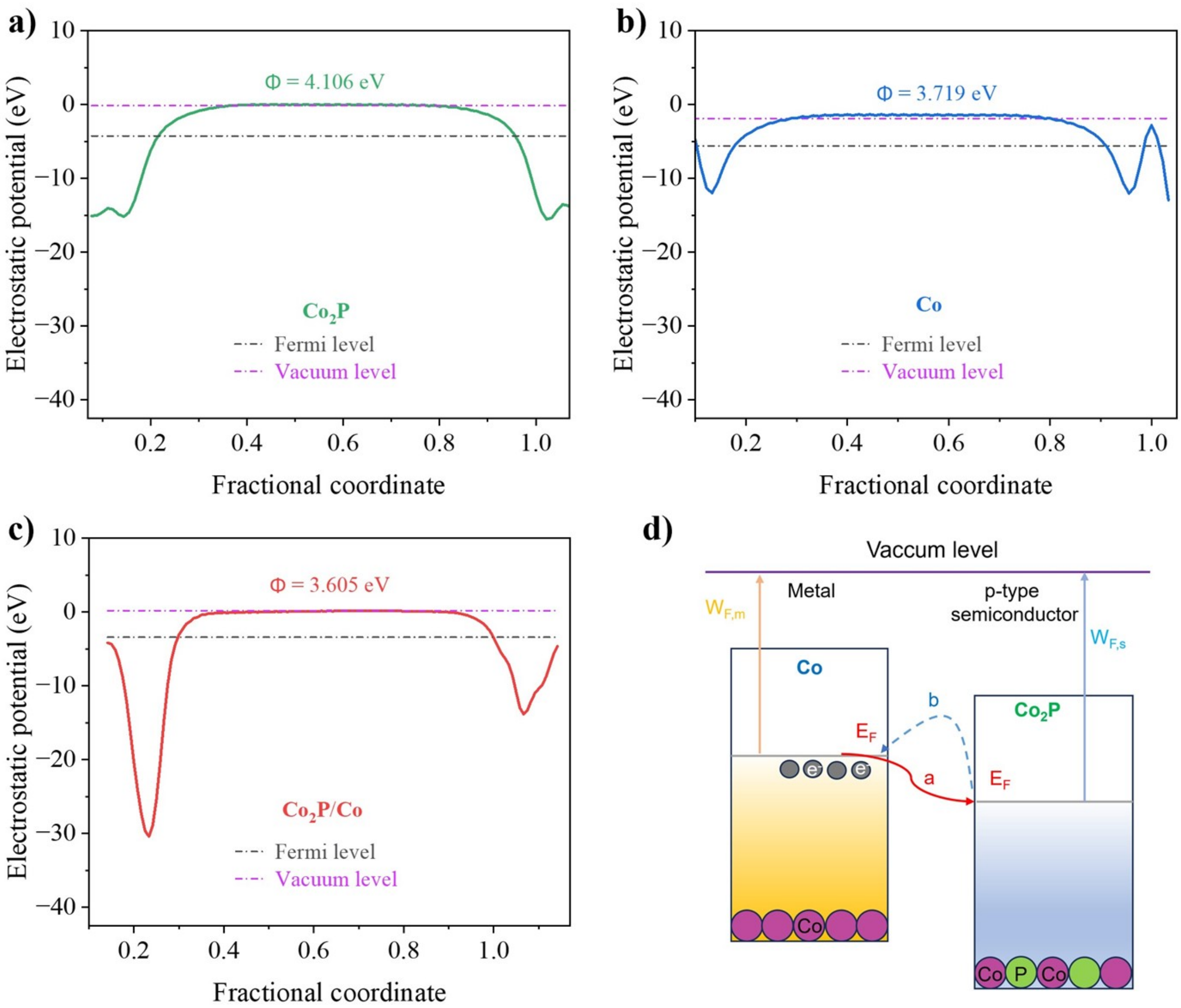

Fig. S27. (a-c) The calculation results for work function for Co_2P, Co and Co_2P /Co, (d) Mott–Schottky model for the interaction between Co sites and Co_2P sites in Co_2P/Co.

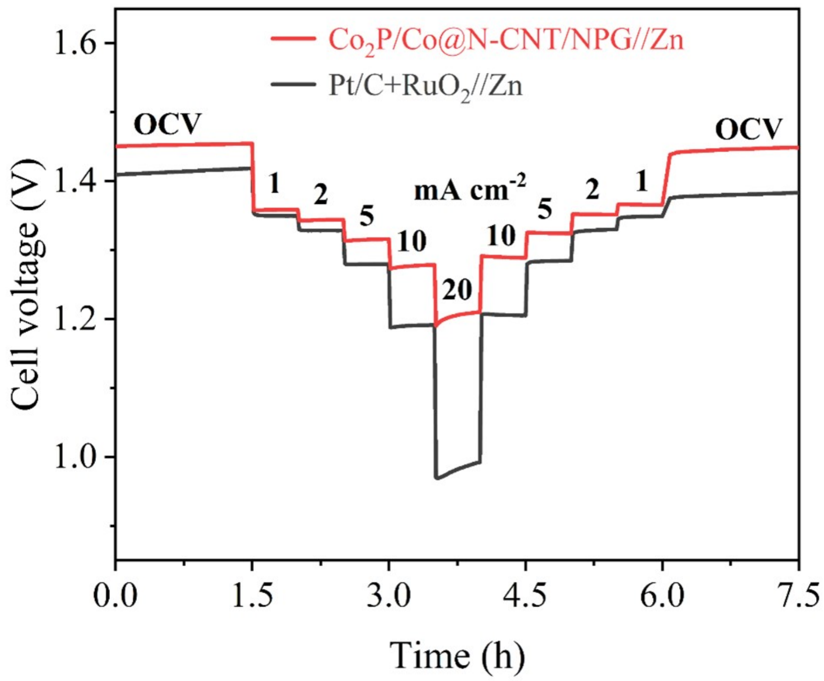

Fig. S28. Rate discharge curves the voltage with various current density of the rechargeable ZAB based on the Co_2P/Co@N–CNT/NPG and Pt/C+RuO_2 cathodes.

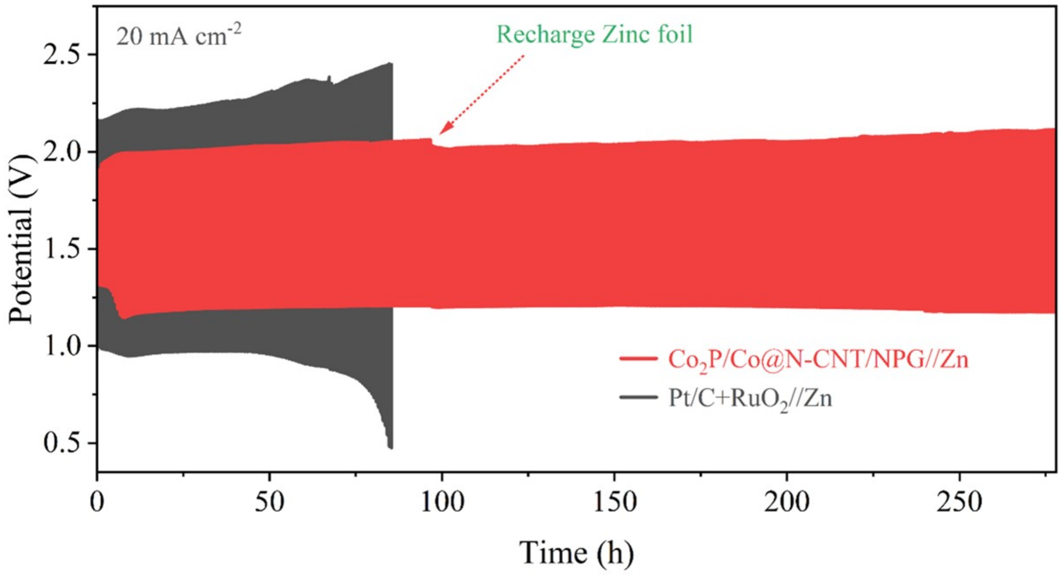

Fig. S29. Cycle stability of ZAB at a high current density of 20 mA cm^{-2}.

Table S1. Elemental composition of as-synthesized catalysts estimated.

Samples	Co (wt.%)	P (wt.%)	N (wt.%)	O (wt.%)	C (wt.%)
Pure NPG	-	1.32	6.84	5.46	86.38
Co@N–CNT/NG	9.56	-	5.58	4.61	80.08
Co_2P/Co@PG	12.46	4.01	-	5.71	77.42
Co_2P/Co@N–CNT/NPG	10.24	3.75	6.45	8.70	70.85

Co, P, N, O, and C contents were detected by XPS analysis.

Table S2 Compare ORR performance with the most recently reported non-noble metal ORR catalysts in 0.1 M KOH.

ORR catalysts	Catalyst loading (mg cm^{-2})	Onset potential (V vs. RHE)	$E_{1/2}$ (V vs. RHE)	n	References
Co_2P/Co@N–CNT/NPG	0.2	1.05	0.91	3.95	This work
20% Pt/C		1.02	0.89	3.97	
Co_xP@N,P-C	0.28	-	0.82	3.95	1
Fe_xP/NPCS	0.16	0.918	0.832	3.98	2
$MnVO_x$@N-rGO	0.3	1.14	0.80	~3.9	3
HCA-Co	-	0.92	0.87	-	4
W_2N/WC	0.6	0.93	0.827	-	5
SHG	0.5	1.01	0.87	3.81-3.96	6
$NH_4Co_xFe_{1-x}PO_4$	0.35	0.83	0.74	3.64-4.01	7
P single bond HPC	0.4	0.88	0.79	3.5–3.9	8
CoP-PBSCF	0.25	-	0.752	~4	9
Mo_2C@NC	0.28	0.9	0.78	-	10
2D-PPCN	-	0.92	0.85	~3.6	11
Fe–N/C	0.1	0.923	0.809	~3.96	12
N,S-CD/rGO	0.35	-	0.69	3.90	13
Co-NTMCs@NSC	0.2	0.945	0.833	3.91	14

Table S3 Compare OER performance with the most recently reported non-noble metal OER catalysts in 1 M KOH.

OER catalysts	Catalyst loading (mg cm^{-2})	Overpotential (mV)	Tafel (mV dec^{-1})	References
$Co_2P/Co@N–CNT/NPG$	0.2	309	60.5	This work
RuO_2		341	74.2	
$Co_xP@N,P-C$	0.28	320	87	[1]
W_2N/WC	0.6	320	94.5	[5]
SHG	0.5	330	71	[6]
$NH_4Co_xFe_{1-x}PO_4$	0.35	313	58.92	[7]
FeP nanorods	-	350	63.6	[15]
VN-Co-P	-	335	-	[16]
CP/CTs/Co-S	-	308	72	[17]
CoP/GO	0.28	340	66	[18]
FeCoNi@Gr	0.32	325	60	[19]
Ni_3S_2/NF	-	312	111	[20]
Co@NPC-H	0.2	350	57	[21]
$Na_{1.95}CoP_2O_7$	0.28	360	51	[22]
$Co_9S_8@MoS_2$	0.4	342	94	[23]
PNPC	0.28	313	83.7	[24]

Table S4 Compare catalytic activities of the reported non-noble bifunctional catalysts for both OER and ORR.

Both OER and ORR Catalysts	Electrolyte (M KOH)	$E_{1/2}$ (V vs. RHE)	$E_{j=10}$ (V vs. RHE)	ΔE (V vs. RHE)	References
$Co_2P/Co@N–CNT/NPG$	0.1	0.91	1.74	0.82	This work
20% Pt/C		0.89	1.91	1.02	
RuO_2		0.75	1.88	1.13	
$MnVO_x@N$-rGO	0.1	0.80	1.65	0.85	3
Co@NPC-H	0.1	0.72	1.58	0.86	21
$Na_{1.95}CoP_2O_7$	0.1	0.73	1.59	0.86	22
SL $Ni(OH)_2$	0.1	0.64	1.57	0.93	25
SWCNT@NPC	1	0.85	1.678	0.83	26
$CoV_{2–x}Fe_xO_4$	1	0.664	~1.53	0.83	27
Fe_2P/EWC	0.1	0.83	1.53	0.87	28
CoFe/N-GCT	0.1	0.79	1.67	0.88	29
$NiO/NiCo_2O_4$	0.1	0.73	-	0.86	30
Fe_2Ni_2N/Co	0.1	0.76	1.63	0.87	31
$Fe/Fe_3C@C$	1	0.84	1.68	0.84	32
Fe@N–C	0.1	0.83	1.71	0.88	33
CoNC-CNF	0.1	0.8	1.68	0.88	34
Ni_3Fe/N-C	0.1	0.78	1.62	0.84	35

Table S5 Compare HER performance with the most recently reported non-noble metal HER catalysts in 1 M KOH.

HER catalysts	Catalyst loading (mg cm^{-2})	Overpotential (mV)	Tafel (mV dec^{-1})	References
Co_2P/Co@N–CNT/NPG	0.2	98	104.6	This work
Pt/C	0.2	37	61.4	
W_2N/WC	0.6	148.5	47.4	[5]
SHG	0.5	230	112	[6]
VN-Co-P		137	81	[16]
CP/CTs/Co-S	-	190	131	[17]
CoP/GO	0.28	150	38	[18]
FeCoNi@Gr	0.32	211	77	[19]
Ni_3S_2/NF	-	131	96	[20]
Co_9S_8@MoS_2	0.4	143	117	[23]
Co/NBC	-	117	146	[36]
NiS/Ni_2P/CC	-	111	78.1	[37]
$NiCo_2S_4$ NA/CC	4	230	141	[38]
$CoCO_3$@NiFe LDH	-	171	168.2	[39]
NiSP/NF	-	93	107	[40]
N-NiS/NiS_2	-	185	106	[41]

Table S6 The comparative performance of the liquid ZAB with recently reported non-noble metal air cathode-based liquid ZAB devices in alkaline electrolytes.

Air cathode	OCP (V)	Power density (mW cm^{-2})	Specific capacity (mAh g_{zn}^{-1}@mA cm^{-2})	Energy density (W h g^{-1} @mA cm^{-2})	Durability (cycles/h @ mA cm^{-2})	References
Co_2P/Co@N–CNT/NPG	1.458	145	781@10	987.2@10	1200/800@10 375/275@20	This work
Pt/C+RuO_2	1.411	98	608@10	728.9@10	825/280@10 225/75@20	This work
$NH_4Co_xFe_{1-x}PO_4$	1.36	74.6	750@10	-	-/30@10	7
NiO/$NiCo_2O_4$	1.47	99.0	814.4@10	911@10	-/175@10	30
Fe/Fe_3C@C	1.37	101.3	682.6@10	764.5@10	297/99@10	32
Fe@N–C	~1.40	220	-	-	100/-@10	33
Ni_3Fe/N-C	-	-	528@10	~634@10	420/105@10	35
CoFeN-NCNTs//CCM	1.455	145	778.4@10	-	1355/445@10	42
N-GCNT/FeCo	1.48	89.3	872.2@100	653@100	240/40@10	3
Co_4N@NC	1.49	98.6	644.3@10	736.2@10	200/-@10	44
CoS_x/NCNTs/Ni	1.45	131	-	-	200/200@5	45
Co_2SiO_4/N-C	1.412	138.2	-	-	600/400@5	46
Co–VN–NC	1.44	237.8	-	-	-	47
$Fe_{0.5}Co_{0.5}O_x$/NrGO	1.43	86	709@25	806@25	120/120@25	48

Table S7 The comparative performance of the solid-state flexible ZAB with recently reported non-noble metal air cathode-based solid-state flexible ZAB devices.

Air cathode	OCV (V)	Durability (cycles/h@mA cm^{-2})	References
Co_2P/Co@N–CNT/NPG	1.415	615/205@5	This work
Pt/C+RuO_2	1.392	210/70@5	
HCA-Co	1.4	50/33.3@1	4
QAFCGO	~1.4	30/10@1	49
Fe-Co_4N@N-C	1.34	45/-@4	50
Co_4N/CNW/CC	1.4	408/136@10	51
NC-Co SA	1.41	125/42@10	52
CoFe/N-GCT	1.48	1600/270@2	53
$LaNiO_3$/NCNT	1.3	120/25@-	54
$NiCo_2O_4$@MnO_2-CNTs	1.36	540/9@3	55
DN-CP@G	1.34	170/170@1	56
P–O/FeN_4–CNS	1.41	-/20@25	57
CC@VG	1.38	108/36@2	58
Fe,N-CNS	1.38	60/10@5	59
Fe-N_x-C	1.49	-/120@1	60
CoNCNTF/CNF	1.34	68/11@0.5	61

Table S8 The comparative performance of the overall water splitting with recently reported non-noble metal electrode-based water splitting devices in 1 M KOH.

Catalysts	Overpotential at 10 mV cm^{-2} (V)	References
Co_2P/Co@N–CNT/NPG	1.66	This work
Pt/C+RuO_2	1.54	
SHG	1.68	[6]
CP/CTs/Co-S	1.74	[17]
CoP/GO	1.7	[18]
FeCoNi@Gr	1.687	[19]
Ni_3S_2/NF	1.68	[20]
Co/NBC	1.68	[36]
NiS/Ni_2P/CC	1.67	[37]
$NiCo_2S_4$ NA/CC	1.68	[38]
$CoCO_3$@NiFe LDH	1.67	[39]
NiSP/NF	1.7	[40]
N-NiS/NiS_2	1.74	[41]
NiCoP–$NiCoSe_2$	1.7	[62]
Co_9S_8/CC	1.74	[63]
$NiSe_2$	1.7	[64]

References

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