A New Method for Defect-Rich Graphene Nanoribbons/Onion-Like Carbon@Co Nanoparticles Hybrids as an Excellent Oxygen Catalyst

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Figures

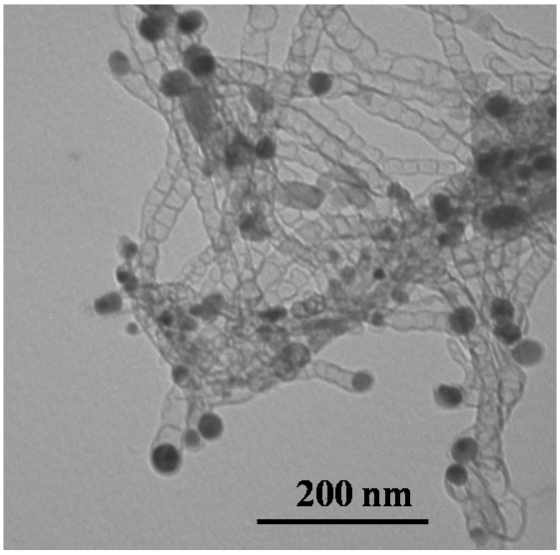

Fig. S1 TEM image of C@Co-NCNT.

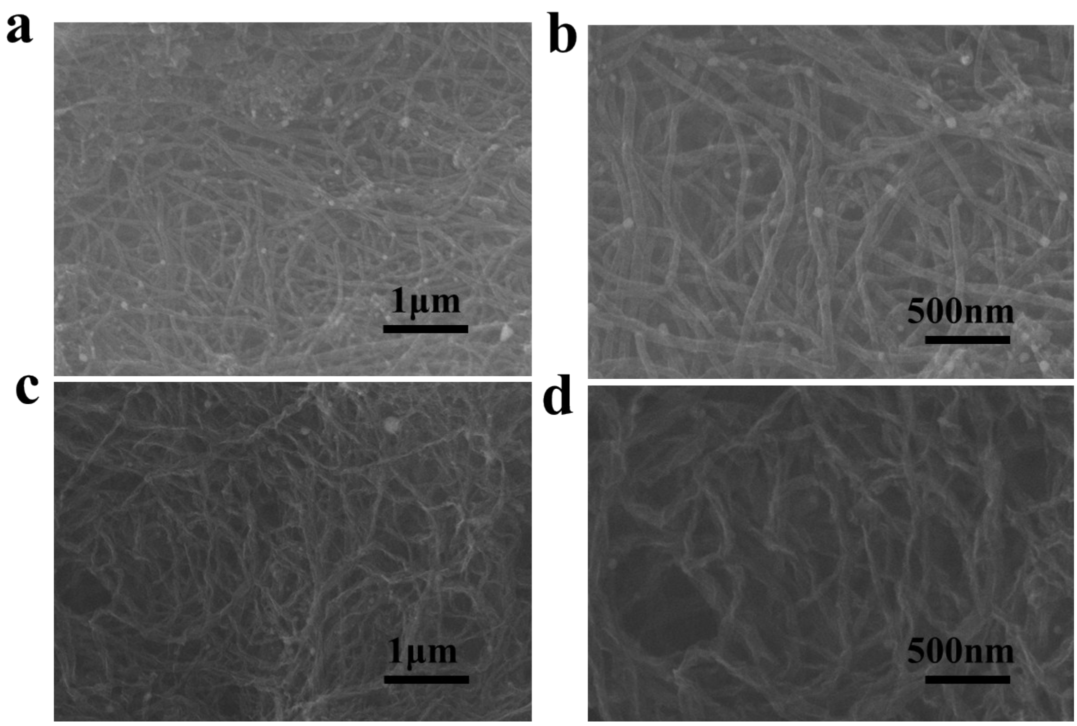

Fig. S2 SEM images of (a, b) C@Co-NCNT and (c, d) C@Co-NGR at different magnifications.

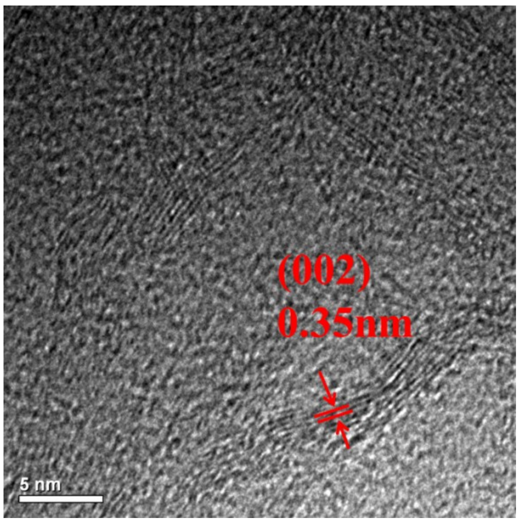

Fig. S3 HRTEM image of C@Co-NGR.

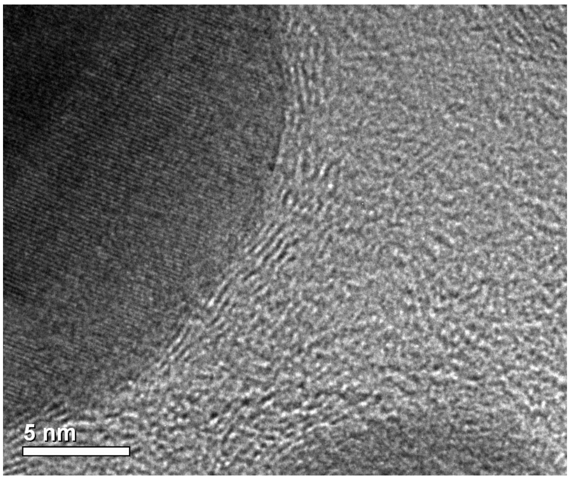

Fig. S4 HRTEM image of C@Co-NCNT.

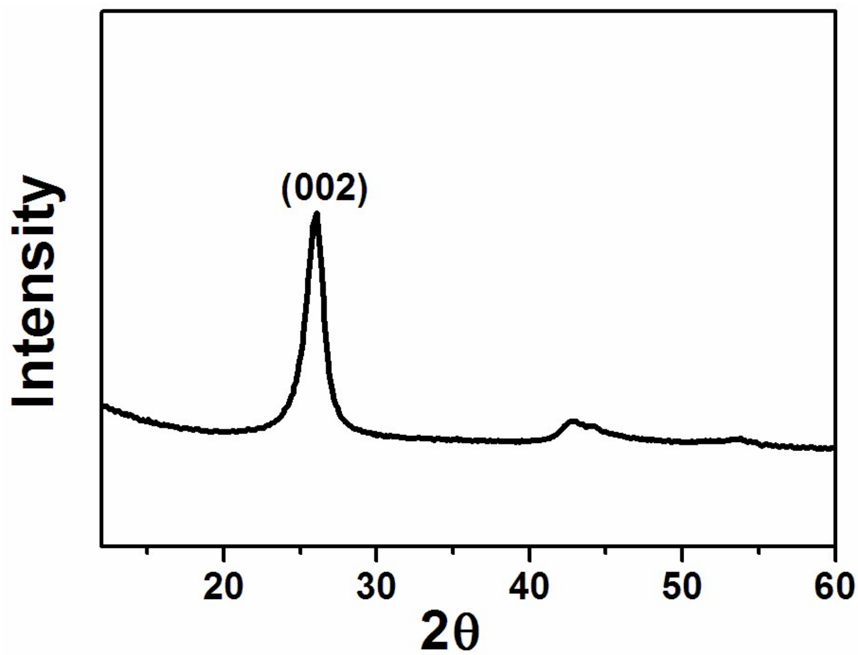

Fig. S5 XRD pattern of the commercial NCNT.

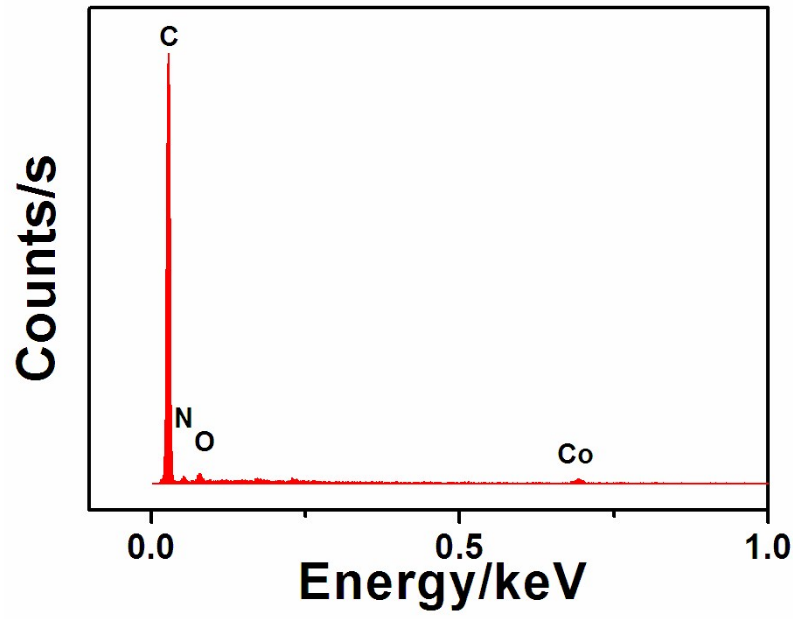

Fig. S6 EDAX spectrum of C@Co-NGR.

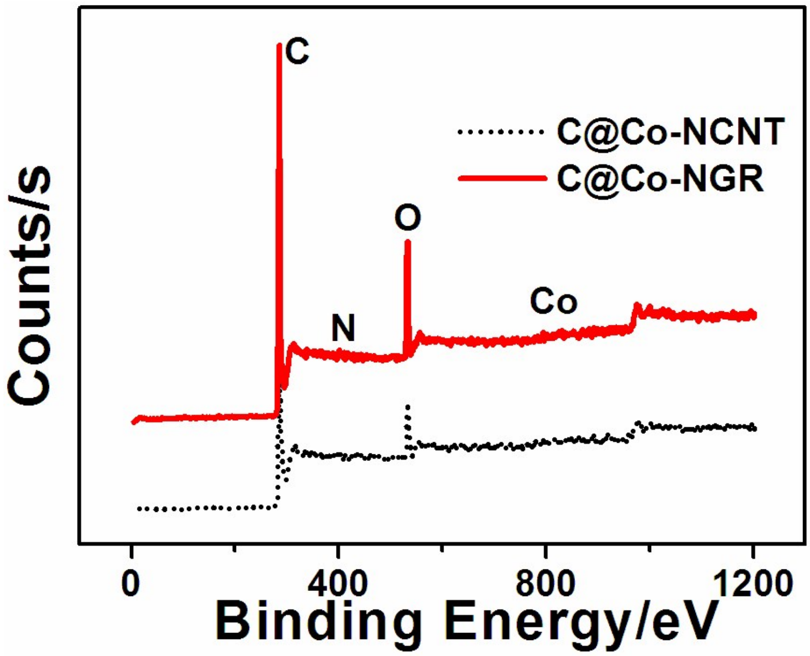

Fig. S7 XPS spectrum of C@Co-NGR and C@Co-NCNT.

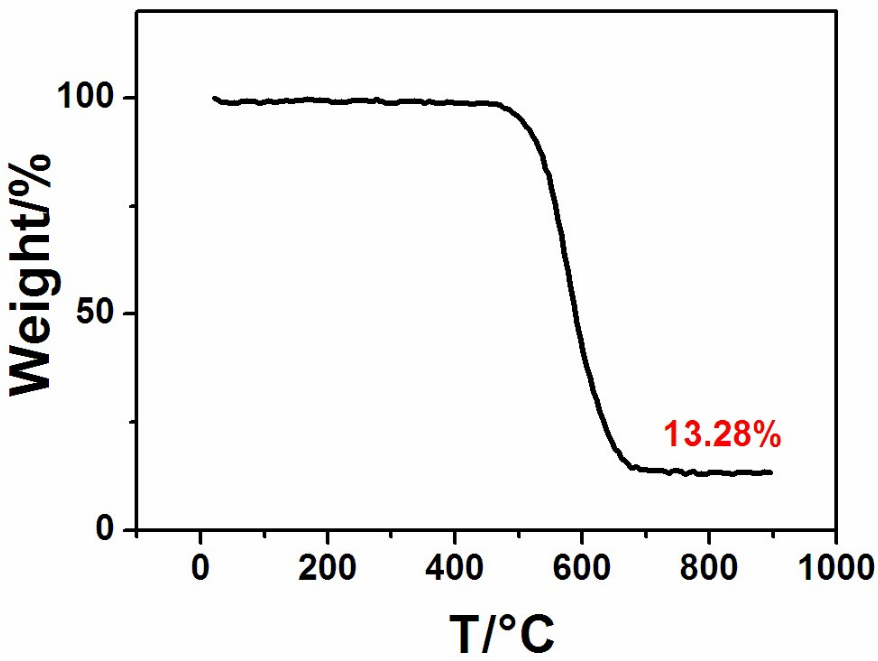

Fig. S8 TGA of C@Co-NGR was conducted at O_2 atmosphere from 25 to 900 °C with a heating rate 10 °C /min.

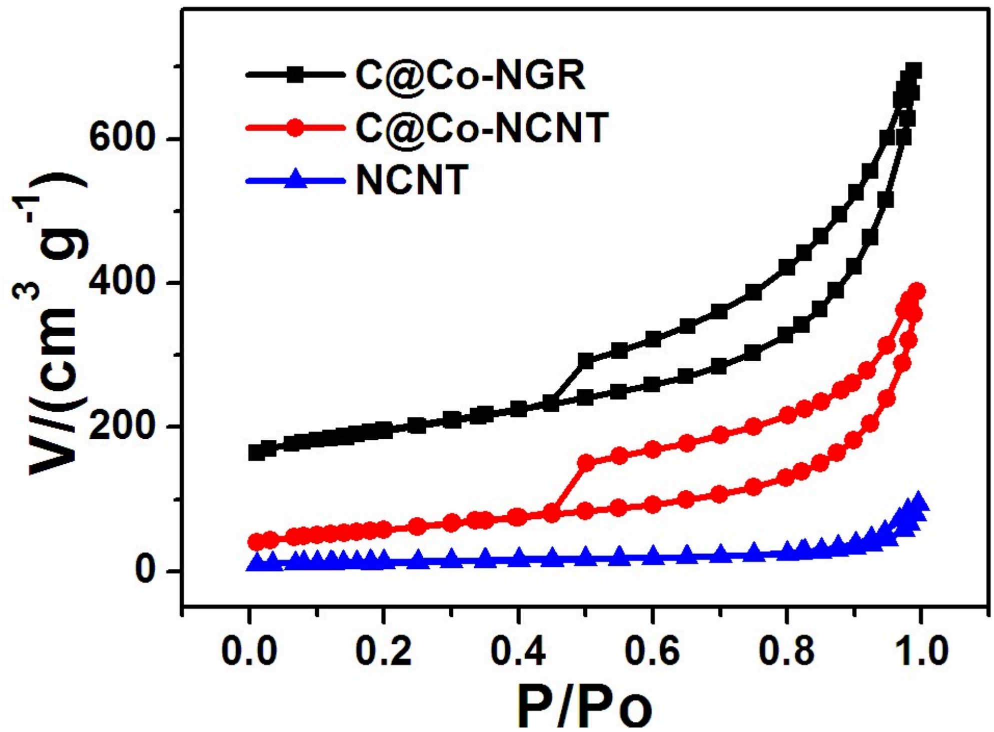

Fig. S9 N_2 adsorption/desorption isotherm curves of the resultant C@Co-NGR, C@Co-NCNT, and NCNT.

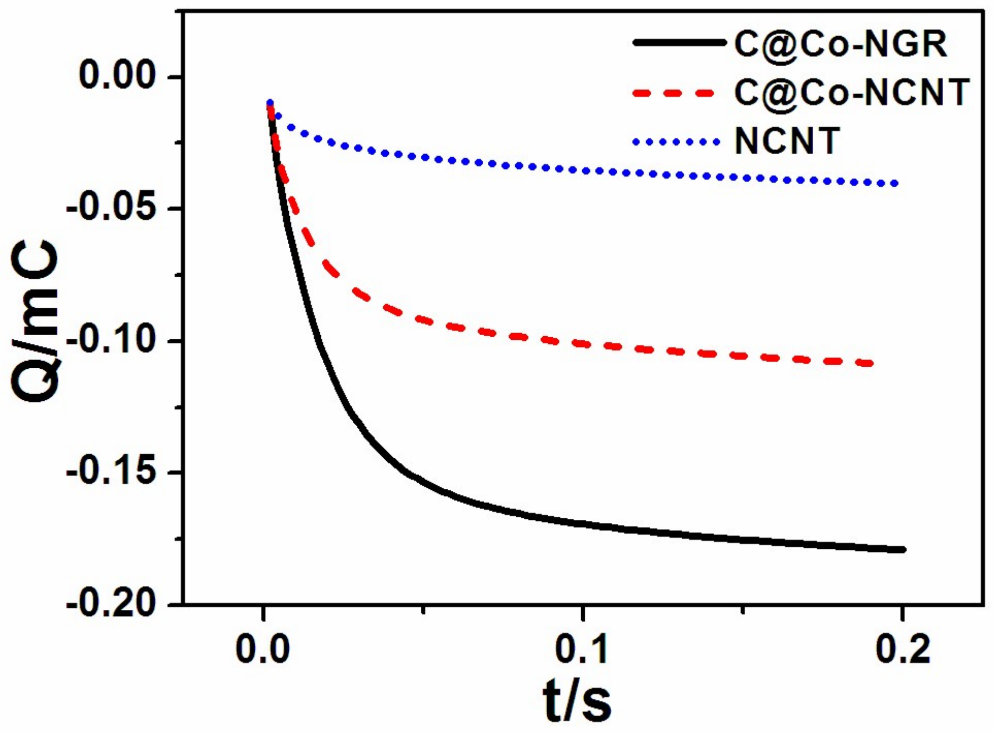

Fig. S10 Chronocoulometric curves of C@Co-NGR, C@Co-NCNT, and NCNT in 1.0 mM $K_3Fe(CN)_6$ solution containing 2.0 M KCl.

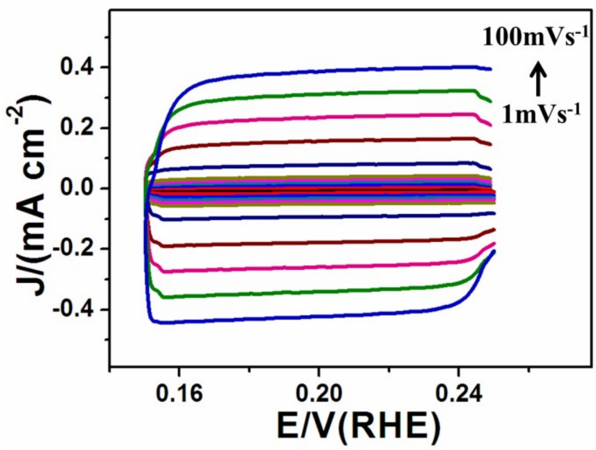

Fig. S11 CVs of C@Co-NGR in 0.50 M H_2SO_4 at different scan rates of 1, 2, 4, 6, 8, 10, 20, 60, 80, and 100 mV s^{-1}, respectively.

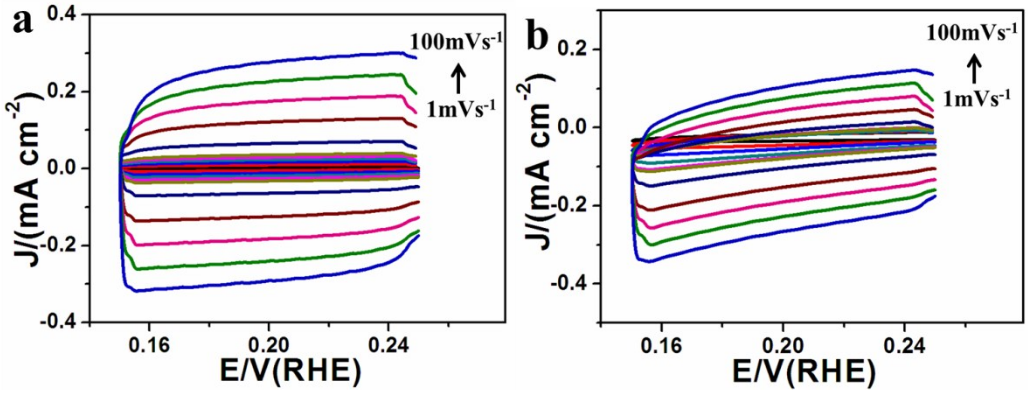

Fig. S12 CVs of (a) C@Co-NCNT and (b) commercial NCNT in 0.50 M H_2SO_4 at different scan rates of 1, 2, 4, 6, 8, 10, 20, 60, 80, and 100 mV s^{-1}, respectively.

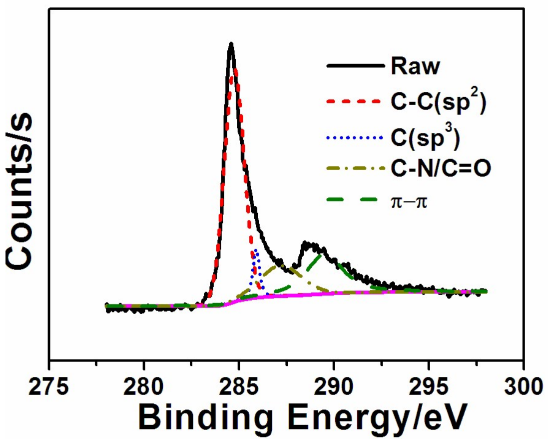

Fig. S13 High-resolution C 1s XPS spectrum for the resultant C@Co-NCNT.

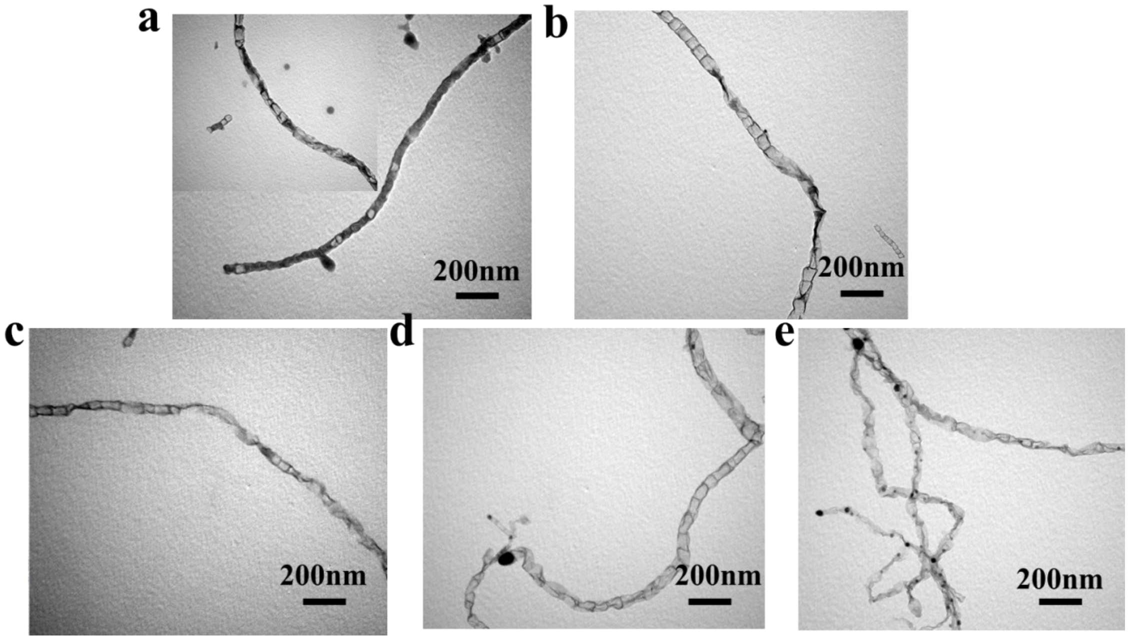

Fig. S14 TEM images of (a) C@Co-NGR-NH_4Cl, (b) C@Co-NGR-$NH_3{\bullet}H_2O$, (c) C@Co-NGR-HCl, (d) C@Co-NGR-$HClO_4$, and (e) C@Co-NGR-$Co(NO_3)_3$.

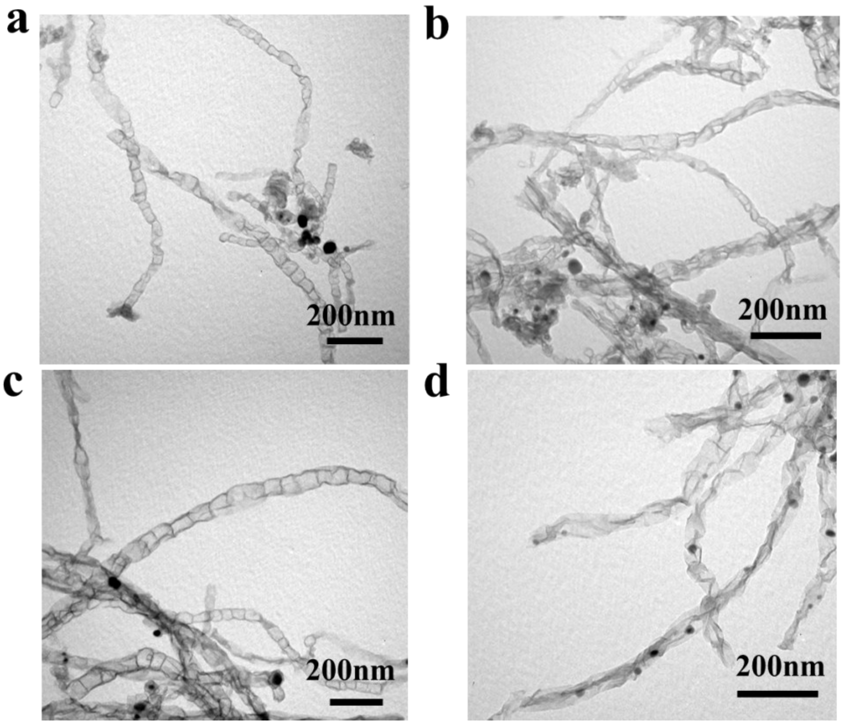

Fig. S15 TEM images of (a) C@Co-NGR-0, (b) C@Co-NGR-300, (c) C@Co-NGR-500, and (d) C@Co-NGR-800.

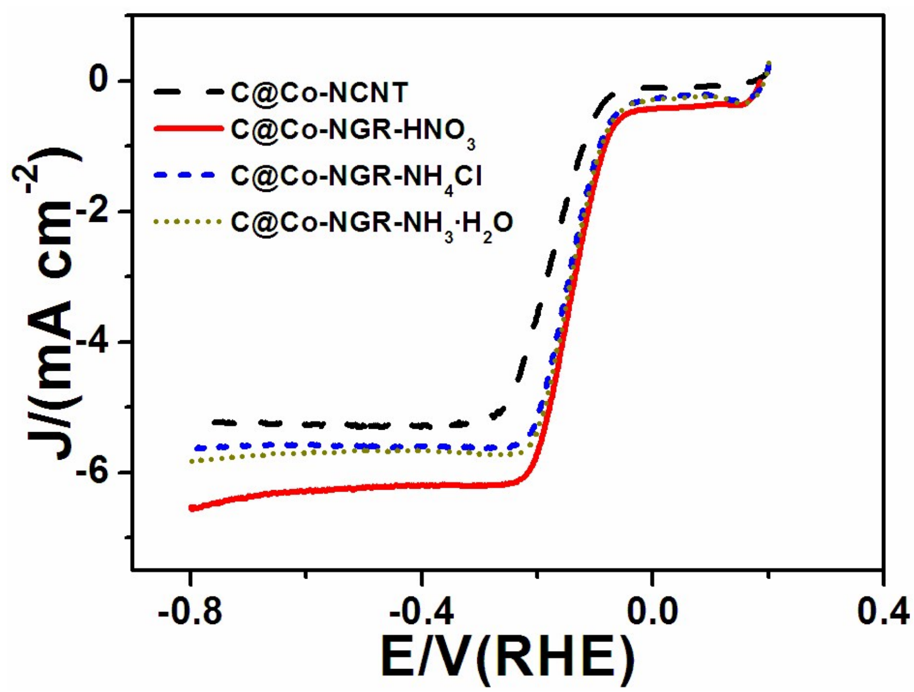

Fig. S16 LSV curves of different materials for ORR in O_2-saturated 0.10 M KOH. Scan rate is 5 mV/s and rotation rate is 1600 rpm.

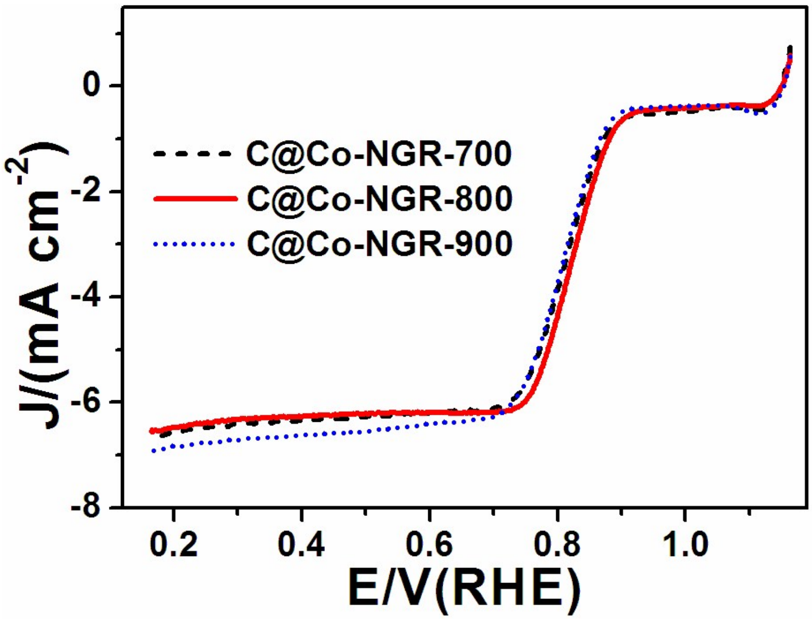

Fig. S17 LSV curves of different materials for ORR in O_2-saturated 0.10 M KOH. Scan rate is 5 mV/s and rotation rate is 1600 rpm.

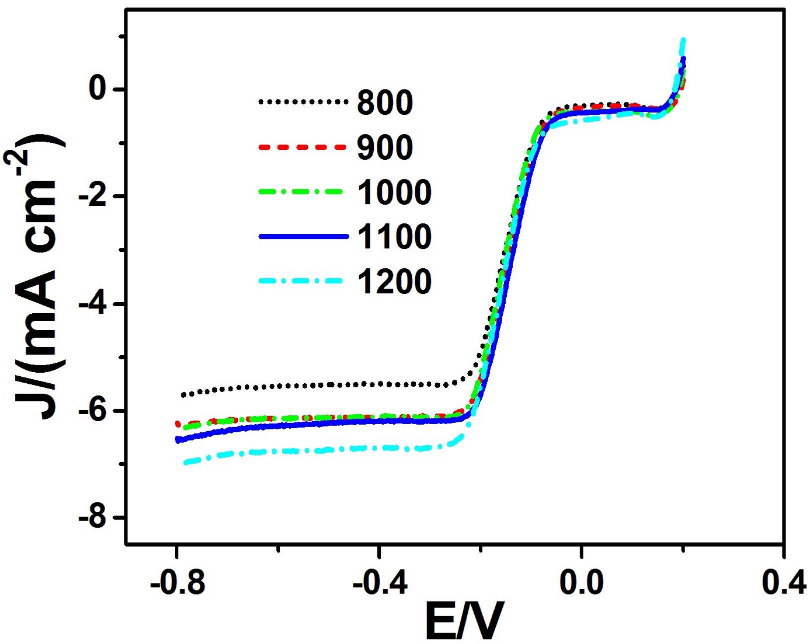

Fig. S18 LSV curves of C@Co-NGR with different loading amount (X=800, 900, 1000, 1100, 1200 μg cm^{-2}) for ORR in O_2-saturated 0.10 M KOH. Scan rate is 5 mV/s and rotation rate is 1600 rpm.

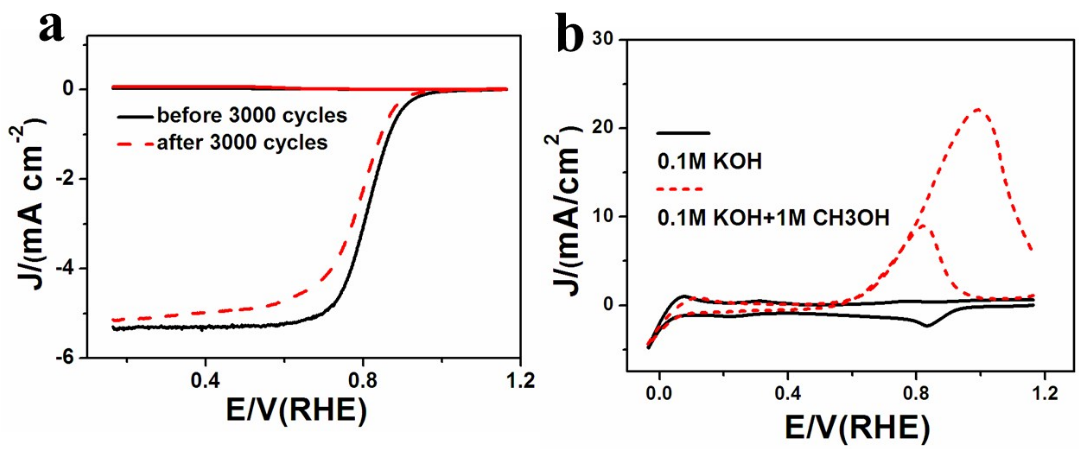

Fig. S19 LSV curves of commercial Pt/C for ORR in O_2-saturated 0.10 M KOH before and after 3000 cycles. (f) CVs of the commercial Pt/C in O_2-saturated 0.10 M KOH without and with 1.0 M CH_3OH at a scan rate of 50 mV s^{-1}.

Table S1 Summary of the properties of the resulting materials.

Samples	BET surface area (m^2 g^{-1})	I_D/I_G	ECASA (cm^2)	ΔE_p (mV)	C_{dl} (mF cm^{-2})	η(mV *vs* RHE) at 10 mA cm^{-2}	Tafel slope (mV dec^{-1})	J_0 (mA cm^{-2})
C@Co-NGR	343.2	1.01	3.30	70	8.06	38	34.5	0.83
C@Co-NCNT	204.8	0.88	2.18	90	5.68	149	46.1	0.21
Commercial NCNT	45.3	0.97	0.576	83	2.98	-	-	-

Table S2 Distribution of C species obtained from the de-convolution of the C1s peaks by XPS

	C-C(sp^2)/%	C(sp^3)/%	C-N/C=O/%	π-π*/%	sp^3/sp^2 (ratio)
C@Co-NCNT	**53.05**	**7.578**	**15.9**	**23.45**	**0.14**
C@Co-NGR	**74.35**	**15.15**	**4.67**	**3.69**	**0.21**

Table S3 Comparisons of HER, OER, and ORR performances for **C@Co-NGR** with other non-precious metal carbon electrocatalysts.

Catalysts	ORR		OER		References
	E^b_{onset}/V	$E^b_{1/2}$/V	E^a_{onset}/V	η^a at 10 mA/cm^2 (mV)	
Fe-derived NCNT	0.89	0.71			1
pPMF	0.973	0.879			2
G/CNT/Co	0.95	0.86			3
BCN-FNHs	-	0.861			4
Co@N-CNTs-m	0.929	0.849			5
Co-C@NWCs	0.939	0.83			6
Co@Co_3O_4@PPD	~0.864	~0.794			7
HDPC	0.95	0.79			8
FeNi@NC			1.44	280	9
Co_3O_4C-NA			1.47	290	10
Co_3O_4-HS				405	11
$Fe_xCo_{(1-x)}$-N/PC		0.812		405	12
Co-MOF@CNTs (5 wt%)	0.91	0.82		340	13
CoO@N/S-CNF	0.84	0.722		320	14
Co–N–C		0.8		310	15

Co/NC		0.83		460	16
Co_3O_4/NRGO	0.92	0.83		420	17
C@Co-NGR	0.910	0.830	1.49	410	This work

E^b: potential in basic solution; **E^a**: potential in acidic solution

pPMF: porous bamboo-like carbon nanotube/Fe_3C nanoparticles

BCN-FNHs: bamboo-like carbon nanotube (b-CNT)/Fe_3C nanoparticle (NP) hybrids

Co@N-CNTs-m: Co nanoparticle-encapsulated N-doped carbon nanotube

HDPC: heteroatom (N, P, Fe) ternary-doped, porous carbons

FeNi@NC: single layer graphene encapsulating FeNi

Co_3O_4C-NA: Co_3O_4-carbon porous nanowire arrays

Co_3O_4-HS: Co3O4 hollow spheres

CoO@N/S-CNF: CoO nanoparticles into nitrogen and sulfur co-doped carbon nanofiber networks

Co_3O_4/NRGO: Co_3O_4/N-doped reduced graphene oxide

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