

Supporting Information

Seaweed Biomass Derived (Ni,Co)/CNT Nanoaerogels: Efficient Bifunctional Electrocatalysts for Oxygen Evolution and Reduction Reactions

Na Ma, Yi (Alec) Jia, Xianfeng Yang, Xilin She, Longzhou Zhang,
Zhi Peng, Xiangdong Yao and Dongjiang Yang

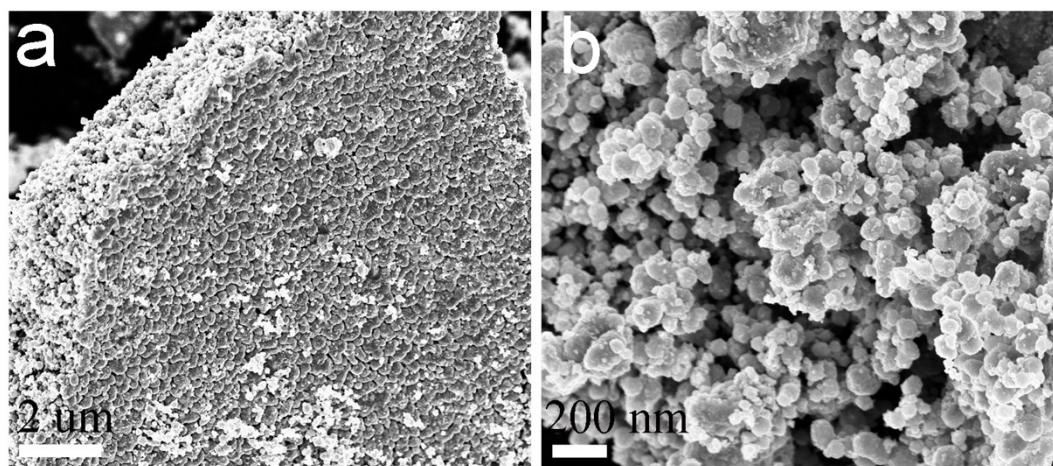


Figure S1. FESEM images of Ni/NiO/NiCo₂O₄/N-As.

Table S1. The Brunauer-Emmett-Teller (BET) surface area, pore volume and average pore size of (Ni,Co)/N-CNT-As samples.

Sample	Ni/NiO/NiCo ₂ O ₄ / N-CNT-As	Ni/NiCo ₂ O ₄ /N- CNT-As	NiCo ₂ O ₄ /N- CNT-As	NiO/NiCo ₂ O ₄ /N -CNT-As
Surface area (m ² g ⁻¹)	193	208	222	189
Pore volume(c m ³ g ⁻¹)	1.055	1.650	1.411	0.805
Average pore size (nm)	16.1	15.3	15.6	11.9

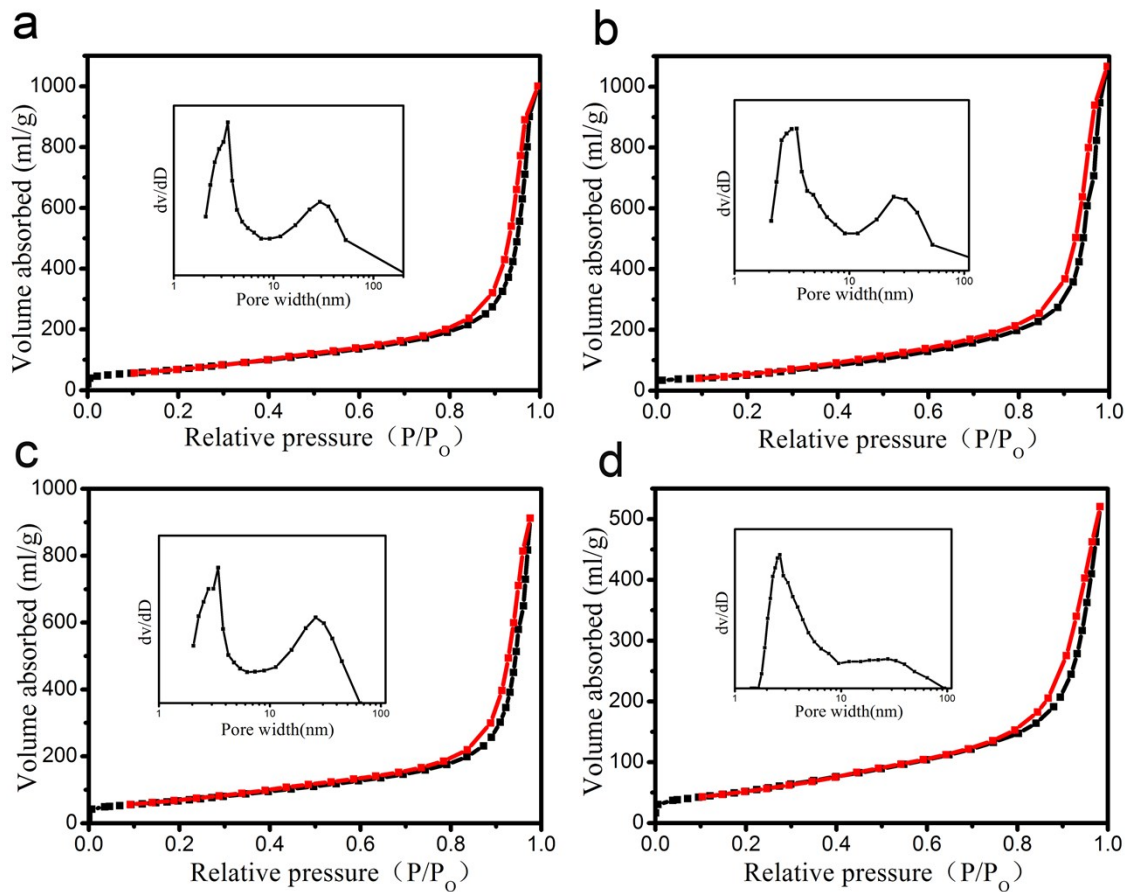


Figure S2. Nitrogen adsorption-desorption isotherms and inset pore size distributions of (a) Ni/NiO/NiCo₂O₄/N-CNT-As. (b) Ni/NiCo₂O₄/N-CNT-As. (c) NiCo₂O₄/N-CNT-As. (d) NiO/NiCo₂O₄/N-CNT-As.

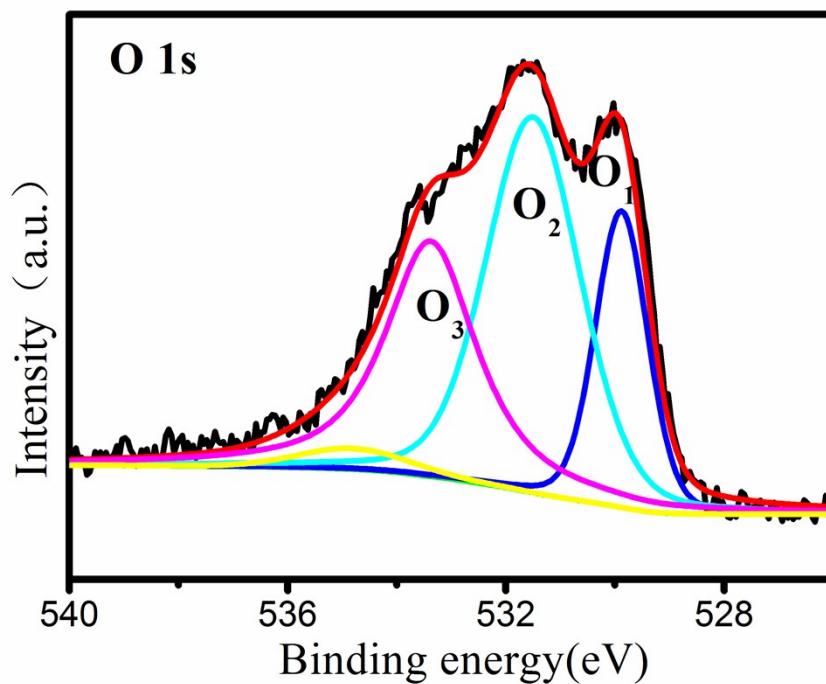


Figure S3. High resolution XPS for O 1s peak of Ni/NiO/NiCo₂O₄/N-CNT-As.

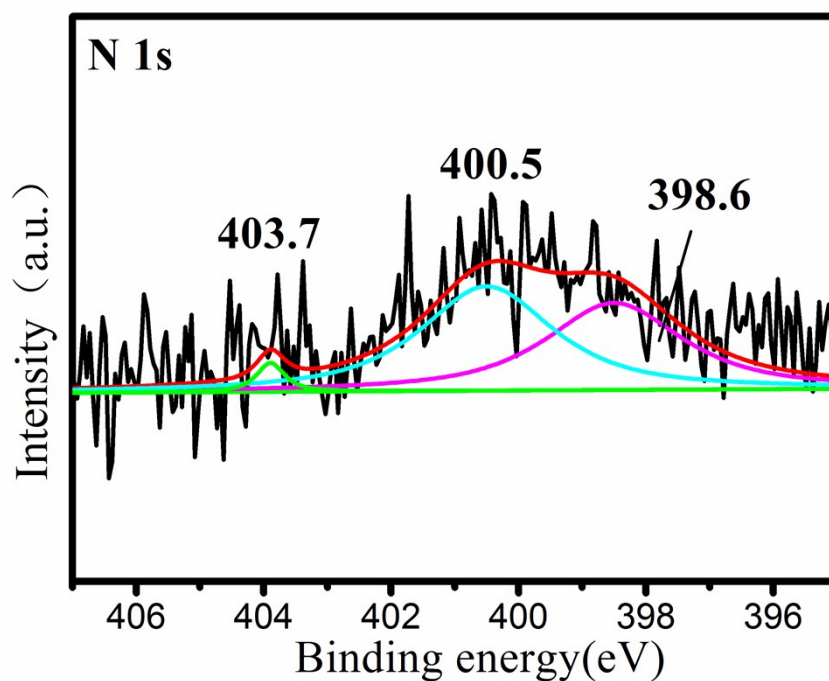


Figure S4. High resolution XPS for N 1s peak of Ni/NiO/NiCo₂O₄/N-CNT-As.

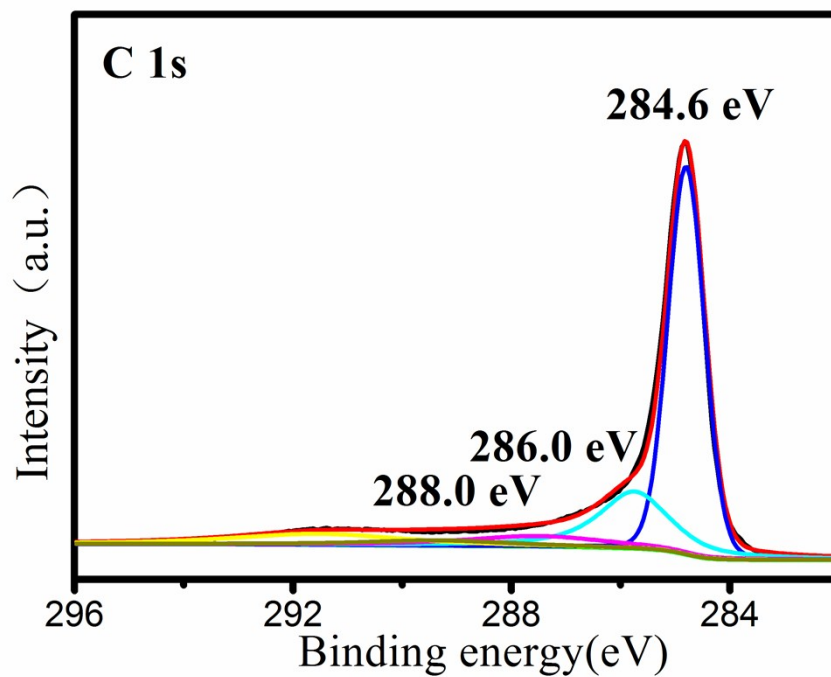


Figure S5. High resolution XPS for C 1s of Ni/NiO/NiCo₂O₄/N-CNT-As.

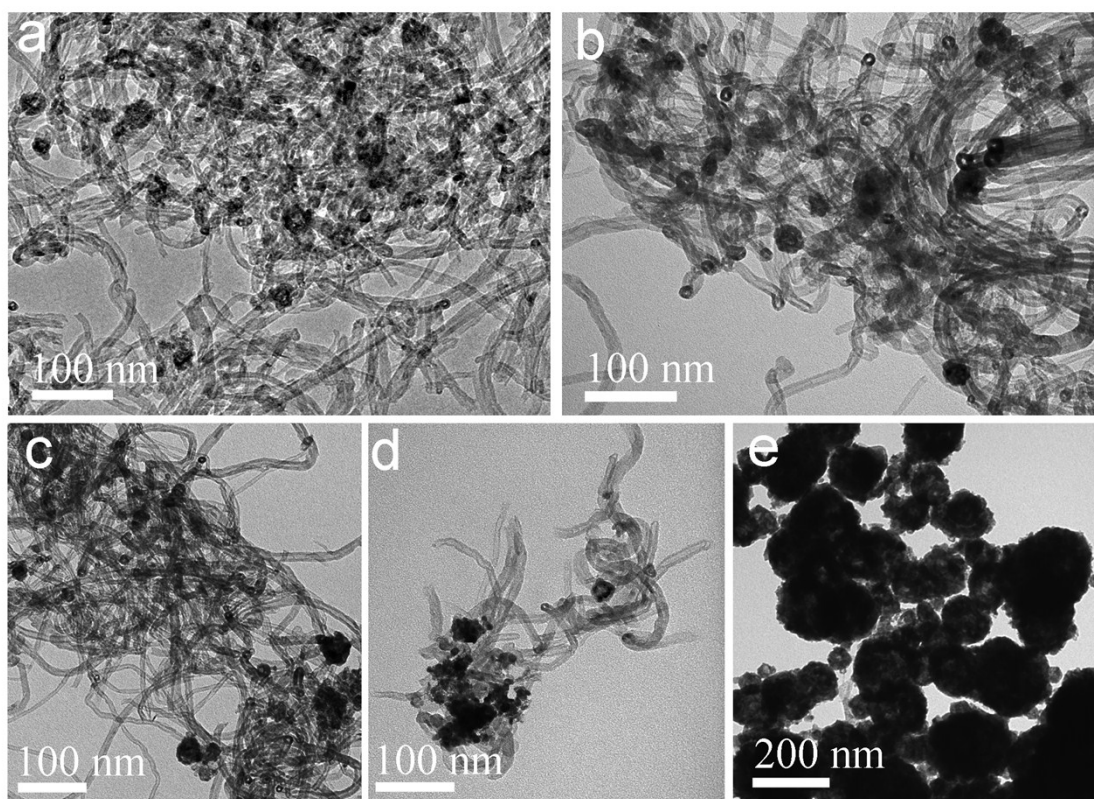


Figure S6. TEM images of (a) Ni/NiO/NiCo₂O₄/N-CNT-As. (b) Ni/NiCo₂O₄/N-CNT-As. (c) NiCo₂O₄/N-CNT-As. (d) NiO/NiCo₂O₄/N-CNT-As. (e) Ni/NiO/NiCo₂O₄/N-As.

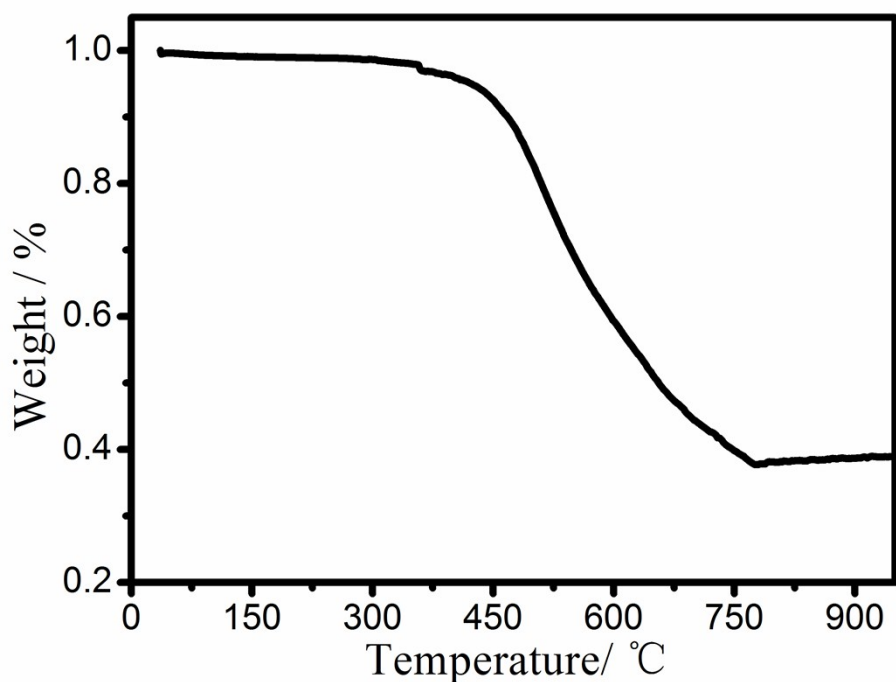


Figure S7. TGA weight change curves of Ni/NiO/NiCo₂O₄/N-CNT-As tested in air with a ramp rate of 10 °C/min.

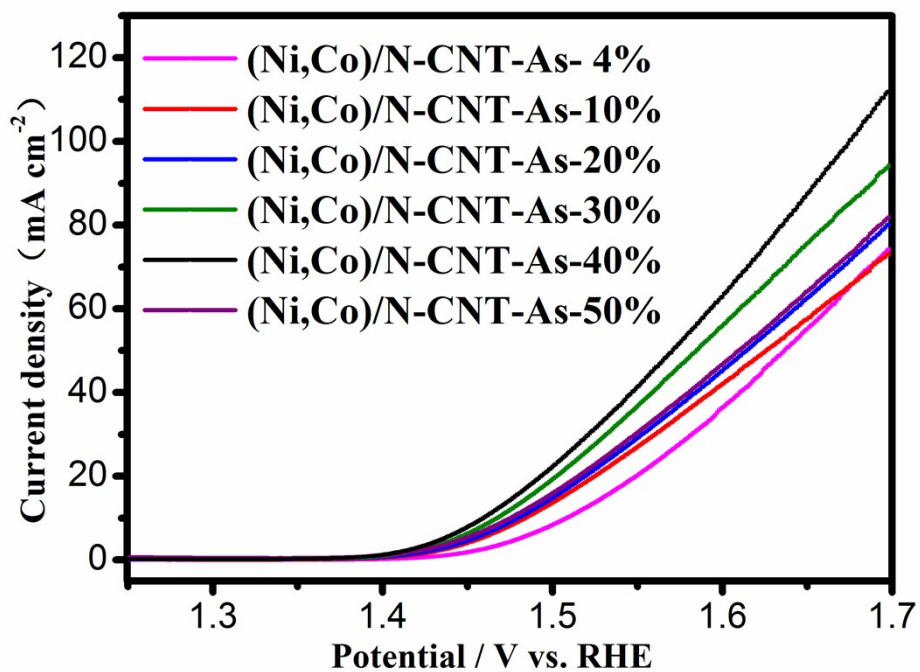


Figure S8. LSV plots of Ni/NiO/NiCo₂O₄/N-CNT-As samples with different amounts

of CNTs in 1 M KOH.

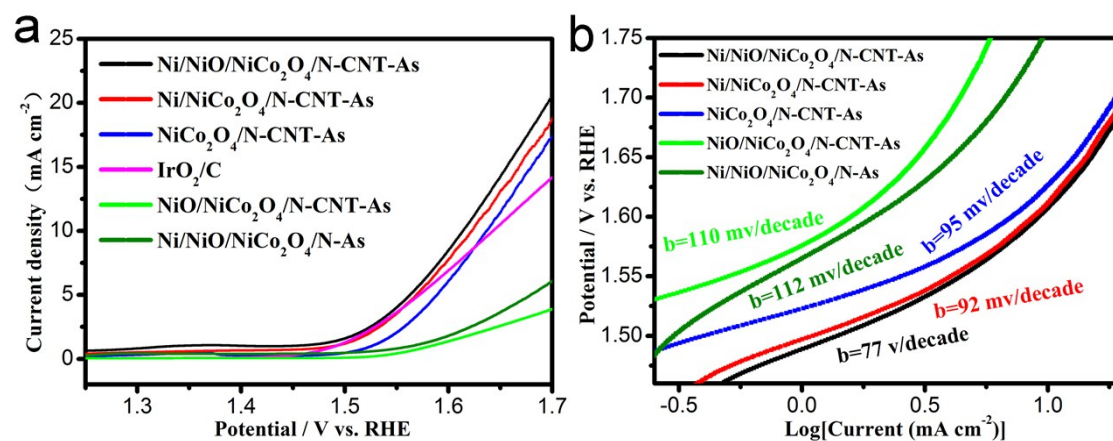


Figure S9. (a) (Ni,Co)/N-CNT-As samples and commercial IrO₂/C at 10mV s⁻¹ in 0.1 M KOH. (b) Tafel plots for (Ni,Co)/N-CNT-As samples in 0.1M KOH.

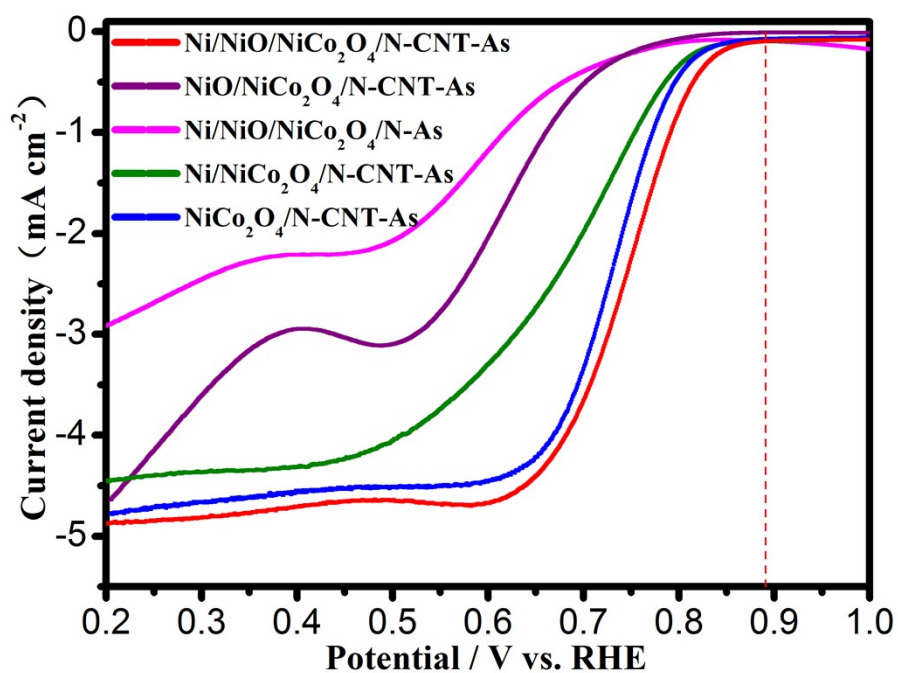


Figure S10. LSV plots of (Ni,Co)/N-CNT-As samples in 0.1 M KOH.

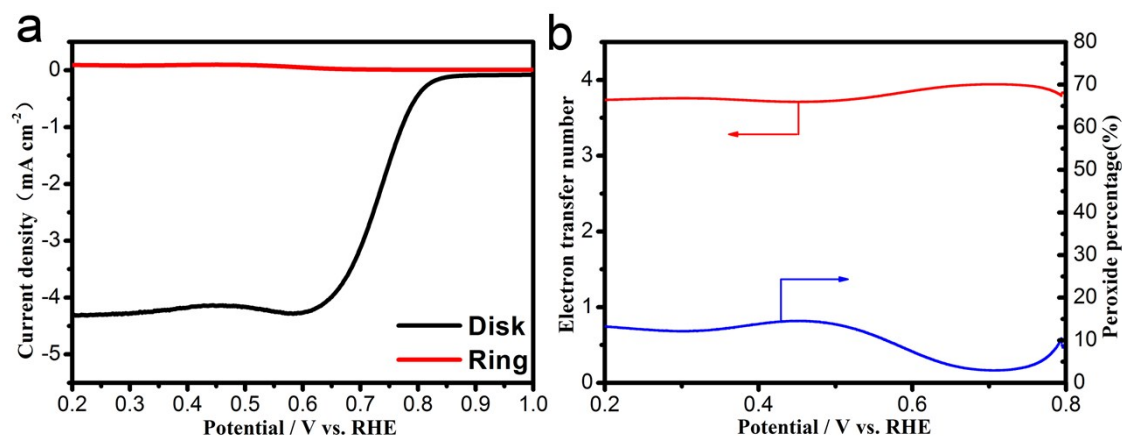


Figure S11. (a) Rotating ring-disk electrode (RRDE) voltammograms recorded with the Ni/NiO/NiCo₂O₄/N-CNT-As in O₂-saturated 0.1 M KOH at 1600 rpm. Disk current (I_D) is shown on the lower half and the ring current (I_R) is shown on the upper half of the graph. (b) The calculated percentage of peroxide and the electron transfer number (n).

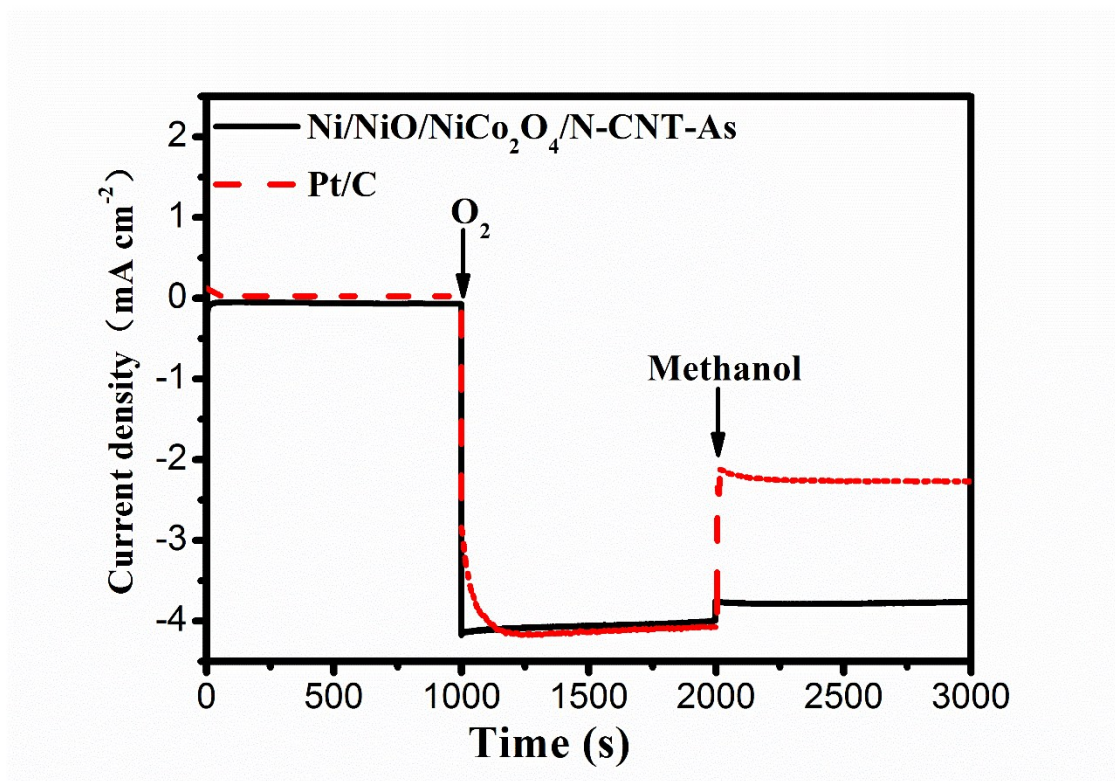


Figure S12. Methanol crossover resistance (at 0.69 V vs.RHE) test of Ni/NiO/NiCo₂O₄/N-CNT-As and 20% Pt/C in 0.1 M KOH.

Table S2. Comparison of OER performance of Ni/NiO/NiCo₂O₄/N-CNT-As with reported metal oxides catalysts in 1 M KOH solution.

Sample	Onset E(V)	$\eta@10\text{mAcm}^{-2}$ (V)	$j@\eta=0.3$ V (mAcm ⁻²)	Loading (mgcm ⁻²)	Tafel slopes (mV/Dec)	References
Ni/NiO/NiCo ₂ O ₄ /N-CNT-As	~1.43	0.24	33	0.24	45	This work
IrO ₂ /C	~1.47	0.35	-	0.24	-	This work
NCNT/CoO-NiO-NiCo	~1.46	0.27	19	0.21	40	<i>Angew. Chem., Int. Ed.</i> , 2015, 54, 9654-9658
Co ₃ O ₄ /N-rmGO	-	0.31	-	1	67	<i>Nat. Mater.</i> , 2011,10,780-786

Note: η is the overpotential and j is Current Density.

Table S3. Comparison of bifunctional oxygen electrode activities of Ni/NiO/NiCo₂O₄/N-CNT-As with reported metal oxides catalysts in 0.1 M KOH solution.

Sample	Onset OER E(V) (vsRHE)	E _{OER} ^a (V) @10 mA cm ⁻²	E _{ORR} ^b (V) @ -3 mA cm ⁻²	Oxygen electrode (E _{OER} -E _{ORR}) ΔE ^c (V)	Loading (mg cm ⁻²)	References
Ni/NiO/NiCo ₂ O ₄ / N-CNT-As	~1.51	1.60	0.74	0.87	0.24	Thiswork
NCO-A ₁	~1.52	1.62	0.78	0.84	0.90	<i>Nanoscale</i> , 2014, 6, 3173-3181
NiCo ₂ O ₄	~1.55	1.72	0.75	0.97	-	<i>J. Mater. Chem. A</i> , 2013, 1, 12170-12177
NiCo ₂ O ₄ /G	~1.54	1.67	0.54	1.13	0.41	<i>J. Mater. Chem. A</i> , 2013, 1, 4754-4762
Co ₃ O ₄ /2.7Co ₂ MnO ₄	~1.58	1.77	0.68	1.09	0.093	<i>Nanoscale</i> , 2013, 5, 5312-5315
macro/meso-NC-NH ₃ ⁺ Co ₃ O ₄ microtrepangs	~1.67	~1.72	~0.82	~0.90	0.255	<i>Energy Environ. Sci.</i> , 2015, 8, 3274-3282
Co ₃ O ₄ /C fiber	~1.53	-	-	-	0.30	<i>Nanoscale</i> , 2015, 7, 1830-1838

Note: ^a E_{OER}: the potential at an OER current density of 10 mA cm⁻². ^b E_{ORR}: the potential at an ORR current density of -3 mA cm⁻². ^c ΔE: the difference of the potentials between E_{ORR} and E_{OER}.

Table S4. A comparison of the Zn-air battery performance of this work with recent literatures in terms of the charge voltage.

Sample	Voltage of charge (initial) v	Voltage of charge (final) v	Voltage gap between discharge and charge (V)	References
Ni/NiO/NiCo ₂ O ₄ /N-CNT-As	1.95 (5A/g)	1.94	0.75	Thiswork
Ni/NiO/NiCo ₂ O ₄ /N-CNT-As	1.95 (10 A/g)	1.90	0.75	Thiswork
Co ₃ O ₄ /C fiber	~2.1 (2 mA/cm ²)	~2.1	~0.9	<i>Nanoscale</i> , 2015,7,1830-1838
Co ₃ O ₄ /C fiber mat	~2.04 (20 mA/cm ²)	~2.04	~0.85	<i>Nanoscale</i> , 2015,7,1830-1838
NCNT/CoO-NiO-NiCo	~2.1 (20 mA/cm ²)	-	~0.9	<i>Angew. Chem., Int. Ed.</i> , 2015, 54, 9654-9658
MnO ₂ and CNT composite	~2.6	~2.73	1.5	<i>Electrochim. Acta</i> , 2012, 69,295-300
Co ₃ O ₄ NP modified MnO ₂ Nanotubes	~2.1	~2.2	~0.85	<i>Nanoscale</i> , 2013,5,4657-4661
Co ₃ O ₄ NC/N-CNT	2.16	-	1.02	<i>ChemSusChem</i> , 2015,8,3129-3138
macro/meso-NC-NH ₃ ⁺ Co ₃ O ₄ microtrepangs	-	~2.0	0.7	<i>Energy Environ. Sci.</i> , 2015,8, 3274-3282
Co ₃ O ₄ NW	2.0	-	1.02	<i>Adv. Energy Mater.</i> , 2014, 4, 1301389

