## Oxygen-Defect Enhanced Anion Adsorption Energy toward Super-Rate and Durable Cathode for Ni-Zn Batteries

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## Table 1 Comparison of the electrochemical performance of alkaline Zn-based batteries.

Cathode//anode materials	Electrolyte	E <sub>mid,d</sub> /V	Specific capacity /mAh g <sup>-1</sup>	<i>W<sub>M,max</sub></i> /Wh kg <sup>-1</sup>	<i>P<sub>M,max</sub></i> /kW kg <sup>-1</sup>	Cycling life /%	Ref.
O <sub>d</sub> -CNO@Ni NTs//Zn foil	$\begin{array}{c} 4 \text{ M KOH} + 2 \text{ M KF} \\ + 1 \text{ M K}_2\text{CO}_3 + \text{sat.} \\ \text{ZnO} \end{array}$	1.62	338(3.0 A g <sup>-1</sup> )	547.48	92.9(248)	93.05000 cycles	This work
Ni–NiO/CC//Zn	6 M KOH + 0.5 M ZnAc <sub>2</sub>	1.75	$256(0.625 \text{ A g}^{-1})$	441.7	41.6(287)	87.5 2000 cycles	[1]
P-NiCo <sub>2</sub> O <sub>4-x</sub> //Zn foil	1 M KOH + 50 mM ZnAc <sub>2</sub>	1.71	366(3.0 A g <sup>-1</sup> )	616.5	30.2 (388)	71.4 5000 cycles	[2]
Co <sub>3</sub> S <sub>4</sub> //Zn foil	3 M KOH + 0.1 M ZnAc <sub>2</sub>	1.70	317(1.0 A g <sup>-1</sup> )	366.1	11.8(141)	82 5000 cycles	[3]
NiS-coated $Ni_{0.95}Zn_{0.05}(OH)_2//Zn$ mesh	4 M KOH + 2 M KF +1 M K <sub>2</sub> CO <sub>3</sub> + sat. ZnO	1.71	360(1mA cm <sup>-2</sup> )	299.6	5.9(236.8)	79.03 500 cycles	[4]
Ni <sub>3</sub> S <sub>2</sub> @PANI//Zn plate	6 M KOH + 0.2 M ZnAc <sub>2</sub>	1.71	242(5.1 A g <sup>-1</sup> )	386.7	38.8 (194)	100 5000 cycles	[5]
NiCo <sub>2</sub> O <sub>4</sub> //Zn plate	6 M KOH + 0.1 M ZnAc <sub>2</sub>	1.70	$212(2.0 \text{ A g}^{-1})$	301.5	13.2 (226)	63.2 1000 cycles	[6]
Ni <sub>2</sub> P//Zn@CF	1  M KOH + 20  mM ZnAc <sub>2</sub>	1.78	$231(1.0 \text{ A g}^{-1})$	318	-	80.0 1500 cycles	[7]
M-Co <sub>3</sub> O <sub>4-x</sub> //Zn foil	1  M KOH + 50  mM ZnAc <sub>2</sub>	1.72	$288(4.0 \text{ A g}^{-1})$	772.4	17.8 (265)	100 60000 cycles	[8]
NiCo <sub>2</sub> O <sub>4</sub> @CC//Zn@C C	1 M KOH + 20 mM ZnAc <sub>2</sub>	1.71	183 (1.6 A g <sup>-1</sup> )	303.8	49.0 (159)	82.7 3500 cycles	[9]
NiCo-90//Zn foil	2.5M KOH + sat. ZnO	1.66	299(2.0 A g <sup>-1</sup> )	391.7	11.1 (235)	73.0 850 cycles	[10]

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$Co-N_{13}Se_2//Zn$ foil	1 M KOH	1.72	$152 (3.0 \text{ A g}^{-1})$	199.3	24.5 (118)	77.9 100 cycles	[11]
NCS@NCH//Zn	2 M KOH + 0.02 M ZnAc <sub>2</sub>	1.60	135(0.5 A g <sup>-1</sup> )	944.8	14.0(77.7)	105.14000 cycles	[12]
FNCP//Zn	1 M KOH	1.65	318(1.0 A g <sup>-1</sup> )	532.7	18(146)	90.6 2000 cycles	[13]
Al-CoNiDH-5%//Zn	2.5 M KOH + sat. ZnO	1.59	264(0.5mA cm <sup>-2</sup> )	-	-	64.4 2000 cycles	[14]
Ni <sub>3</sub> S <sub>2</sub> /O <sub>v</sub> -Ni(OH) <sub>2</sub> //Zn	1 M KOH + 20 mM ZnAc <sub>2</sub>	1.71	222 (1.0 A g <sup>-1</sup> )	384.6	1.73(275)	93.2 3000 cycles	[15]
CNMO-15//Zn	1 M KOH + 10 mM ZnAc <sub>2</sub>	1.77	271(2.0 A g <sup>-1</sup> )	474.1	10.3(118.8)	100 5000 cycles	[16]
NiAlCo-LDH/CNT//Zn @Cu foil	1 M KOH + 50 mM ZnAc <sub>2</sub>	1.75	278(66.7 A g <sup>-1</sup> )	324	40.0 (225)	85.0 600 cycles	[17]
β-Ni(OH) <sub>2</sub> /CNFs//Zn foil	6 M KOH +1 M LiOH + PAAS + sat. ZnO	1.80	184(5 mA cm <sup>-2</sup> )	325	11.4 (166)	96.0 1200 cycles	[18]
Ni <sub>3</sub> S <sub>2</sub> @NF//Zn foil	1 M KOH + 20 mM ZnAc <sub>2</sub>	1.78	68(5.0 A g <sup>-1</sup> )	223.3	7.3 (109)	100 100 cycles	[19]
NiSe <sub>2</sub> //Zn foil	6M KOH + sat. ZnO	1.74	244(1.45 A g <sup>-1</sup> )	328.8	91.2 (185)	91.710000 cycles	[20]
Co <sub>3</sub> O <sub>4</sub> @NiO//Zn@Cu foil	6 М КОН	1.72	183(5 mA cm <sup>-2</sup> )	316.1	5.1 (130)	89.0 500 cycles	[21]
NNA@CNH//NNA@Z n	1 М КОН	1.75	246(5.0 A g <sup>-1</sup> )	148.5	13.8 (76)	88.0 5000 cycles	[22]
NiO/CNTs//Zn plate	1 M KOH + 10 mM ZnAc <sub>2</sub>	1.75	155(1.0 A g <sup>-1</sup> )	228.3	4.4 (129)	65.0 500 cycles	[23]
CC-CCH@CMO//CC- ZnO@C-Zn	6 M KOH+1.5 M ZnO	1.65	0.7 mAh cm <sup>-2</sup> (1 mA cm <sup>-2</sup> )	235.6	12.6 (152)	71.1 5000 cycles	[24]
CC-CF@NiO//CC-CF @ZnO	2 M KOH + sat. ZnO	1.75	203(0.5mA cm <sup>-2</sup> )	355.7	17.9 (210)	72.9 2400 cycles	[25]

Co <sub>3</sub> O <sub>4</sub> @NF//Zn @CF	1 M KOH + 10 mM ZnAc <sub>2</sub>	1.78	150(3.0 A g <sup>-1</sup> )	241	14.0 (110)	80.0 2000 cycles	[26]
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*E*mid,d is the middle discharge voltage. *W*max and *P*max represent the maximum energy density and power density, respectively. The data in the parenthesis is the corresponding energy density at the maximum power density.



Fig. S1. a, b, c SEM of ZnO Nanorod Arrays (ZnO NAs) at different magnifications.d XRD patterns of ZnO NAs.



Fig. S2. XRD patterns of Ni NTs.

Here, in order to prevent the influence of foam nickel substrate on the test, we use Ti sheet as the electrodeposition substrate to prove that the electrodeposition product is nickel elemental.



**Fig. S3. a, b** SEM images of CNO at different magnifications. **c, d** SEM images of O<sub>d</sub>-CNO at different magnifications.



**Fig. S4.** Nitrogen adsorption–desorption isotherms and pore size distributions of O<sub>d</sub>-CNO@Ni NTs, O<sub>d</sub>-CNO and CNO

Here, since our sample is an array structure grown in situ on the nickel foam, the Brunauer-Emmett-Teller (BET) test results include the quality of the Ni foam.



Fig. S5. XRD patterns of CNO and O<sub>d</sub>-CNO.

Here, in order to prevent the influence of nickel foam substrate on the test, we pump and filter the powder sample for testing.



Fig. S6. The SAED pattern for the CNO sample.



Fig. S7. Energy dispersive spectrum (EDS) of the O<sub>d</sub>-CNO@Ni NTs sample.

**Table 2.** EDX results in elements content from the area identified in TEM micrograph(Fig 2 e).

Element	Spectral peak area	Area Sigma	K factor	wt%	wt% Sigm a	Atomic perce nt
СК	406	68	2.208	1.70	0.28	5.94
N K	141	63	2.965	0.79	0.35	2.37
ОК	3371	113	1.810	11.55	0.36	30.31
Co K	11533	205	1.239	27.06	0.43	19.27
Ni K	24982	298	1.245	58.90	0.54	42.11
total				100.00		



Fig. S8. Optical photograph of the material preparation process.



Fig. S9. XPS survey spectra of the O<sub>d</sub>-CNO@Ni NTs.



Fig. S10. capacity contribution ratio of a Od-CNO@Ni NTs. b Od-CNO. c CNO.



Fig. S11. EIS equivalent circuit diagram.

	Rct	W
CNO	0.72 Ω	0.64 Ω
O <sub>d</sub> -CNO	0.40 Ω	0.33 Ω
Od-CNO@Ni NTs	0.35 Ω	0.07 Ω

Table 3. The EIS results of CNO, Od-CNO, Od-CNO@Ni NTs.



**Fig. S12.** The CNO cathode **a** The CV curve at different voltage Windows at 10 mV s<sup>-1</sup>. **b** The CV curve at different voltage Windows at 20 mV s<sup>-1</sup>. **c** CV curves. **d** CV curves of O<sub>d</sub>-CNO cathodes.



Fig. S13. a Galvanostatic charge-discharge curves of the CNO cathode. b Galvanostatic charge-discharge curves of the  $O_d$ -CNO cathode.



**Fig. S14.** Element analysis of dissolved Co ions in electrolyte during cycling by ICP-OES.



Fig. S15. The SEM images of O<sub>d</sub>-CNO@Ni NTs, O<sub>d</sub>-CNO and CNO electrodes with different number of cycles



Fig. S16. The TEM images of CNO electrodes before and after cycles



Fig. S17. The total density of states of CNO bulk.



Fig. S18. Ex-situ SEM images at difffferent states.



Fig. S19. Ex-situ XRD patterns at difffferent states.



Fig. S20. Ex-situ XPS data at difffferent states.



Fig. S21. a, b, c Ex-situ HRTEM images at difffferent states. d, e, f EDX element mappings at difffferent states.



Fig. S22. The time vs. voltage curve of the O<sub>d</sub>-CNO@Ni NTs//Zn aqueous battery at  $60 \text{ A g}^{-1}$ .



**Fig. S23.** Comparison of the CV curves of **a** 2 mV s<sup>-1</sup> **b** 3 mV s<sup>-1</sup> **c** 5mV s<sup>-1</sup> **d**10 mV s<sup>-1</sup>.



Fig. S24. Comparison of the CV curves before and after a fire and b knock.

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