Electronic Supplementary Material (ESI) for Inorganic Chemistry Frontiers.

Supplementary Materials:

Fabrication dual-functional electrodes of oxygen vacancy abundant $NiCo_2O_4$ nanosheets for advanced hybrid supercapacitors and Zn-ion batteries

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Preparation of Co_3O_4 nanosheets

The electrodes of Co_3O_4 and $ZnCo_2O_4$ are synthesized using a similar method. For the preparation the Co_3O_4 on Ni foam, the used 2.5 mmol $Co(NO_3)_2 \cdot 6H_2O$, 5 mmol of NH_4F and 12.5 mmol $C_6H_{12}N_4$ were dissolved in a mixed solution of 30 mL of H_2O stirring for 30 min. After that, the mixed solution was sealed and kept at 120 °C for 5 h. The Co_3O_4 nanowires were obtained by calcination of the Co precursor at 350 °C for 2 h at a rate of 2 °C·min^{-1}.

Preparation of $ZnCo_2O_4$ nanosheets

For the synthesis of $ZnCo_2O_4$ on Ni foam, the used 1 mmol $Zn(NO_3)_2 \cdot 6H_2O$, 5 mmol $Co(NO_3)_2 \cdot 6H_2O$, 2 mmol of NH_4F and 5 mmol $C_6H_{12}N_4$ were dissolved in a mixed solution of 40 mL of H_2O stirring for 30 min. After that, the mixed solution was sealed and kept at 120 °C for 5 h. The $ZnCo_2O_4$ nanowires were obtained by calcination of the Zn-Co precursor at 400 °C for 2 hours at a rate of 2 °C·min^{-1}.

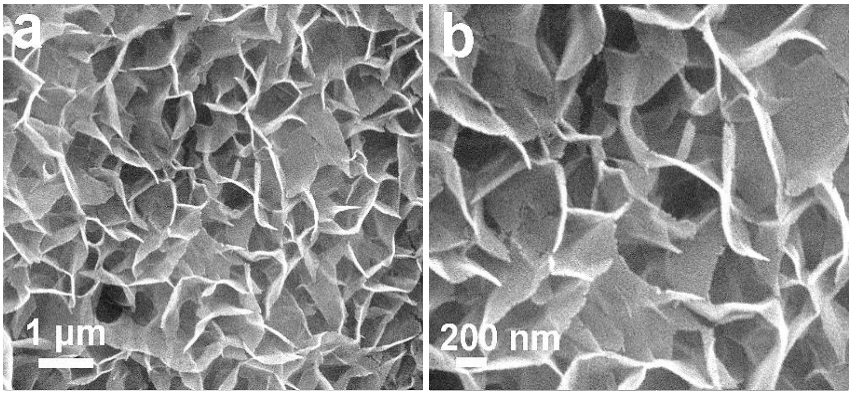


Figure S1 (a, b) SEM image of the $NiCo_2O_4$ samples.

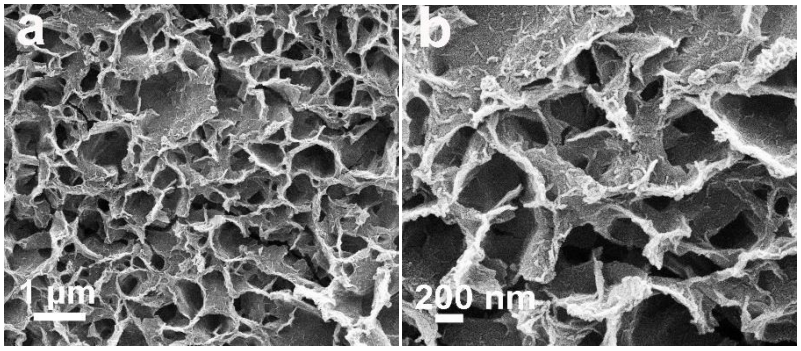


Figure S2 (a, b) SEM image of the V-$NiCo_2O_4$-3 samples.

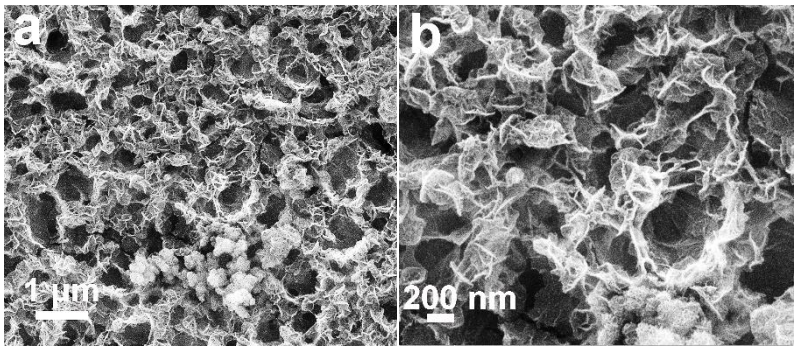


Figure S3 (a, b) SEM image of the V-$NiCo_2O_4$-5 samples.

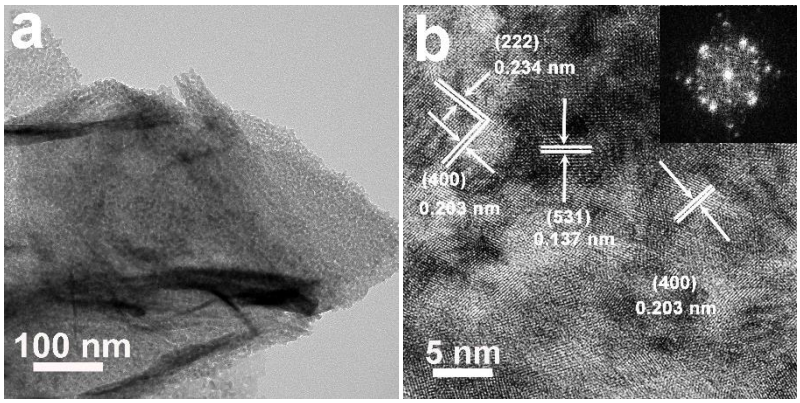


Figure S4 TEM images and (d) high-resolution TEM image of $NiCo_2O_4$.

Table S1 XPS peak area ratios of $NiCo_2O_4$ and V-$NiCo_2O_4$-4.

Peak identity / Materials	$2P_{1/2}$ Co^{2+}	$2P_{1/2}$ Co^{3+}	$2P_{3/2}$ Co^{2+}	$2P_{3/2}$ Co^{3+}	$2P_{1/2}$ Ni^{2+}	$2P_{1/2}$ Ni^{3+}	$2P_{3/2}$ Ni^{2+}	$2P_{3/2}$ Ni^{3+}	O I	O II	O III
$NiCo_2O_4$	0.48	0.52	0.53	0.47	0.43	0.57	0.60	0.40	0.26	0.37	0.42
V-$NiCo_2O_4$-4	0.63	0.37	0.68	0.32	0.52	0.48	0.63	0.37	0.32	0.55	0.13

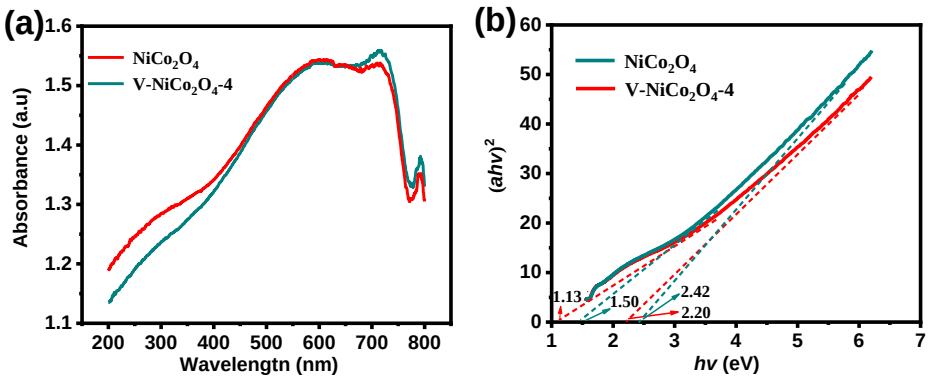


Figure S5 (a) UV-vis absorbance spectra of $NiCo_2O_4$ and V-$NiCo_2O_4$-4 samples; (c) The experimental bandgaps of $NiCo_2O_4$ and V-$NiCo_2O_4$-4 samples.

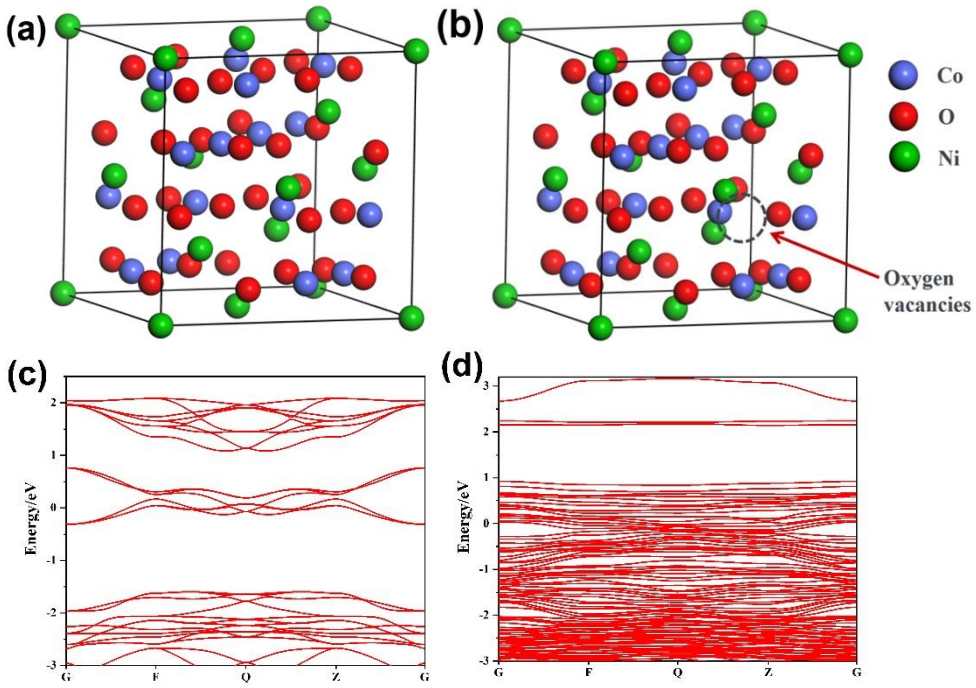


Figure S6 The atomic structure of $NiCo_2O_4$ (a) and V-$NiCo_2O_4$-4 (b); Minority spin channel in $NiCo_2O_4$ (c) and V-$NiCo_2O_4$-4 (d).

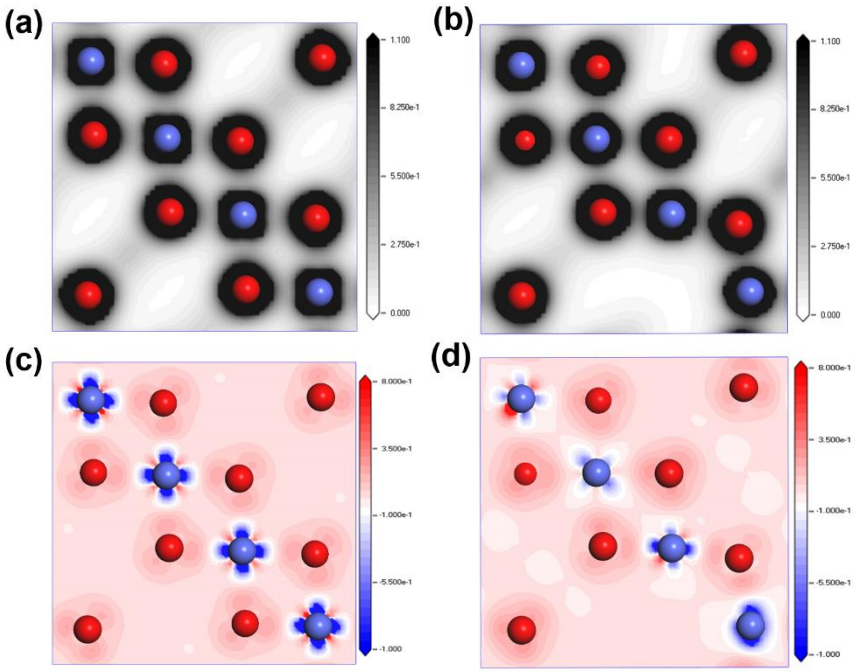


Figure S7 (a, b) electrons density image of $NiCo_2O_4$ and V-$NiCo_2O_4$-4; (c, d) electron density difference of $NiCo_2O_4$ and V-$NiCo_2O_4$-4.

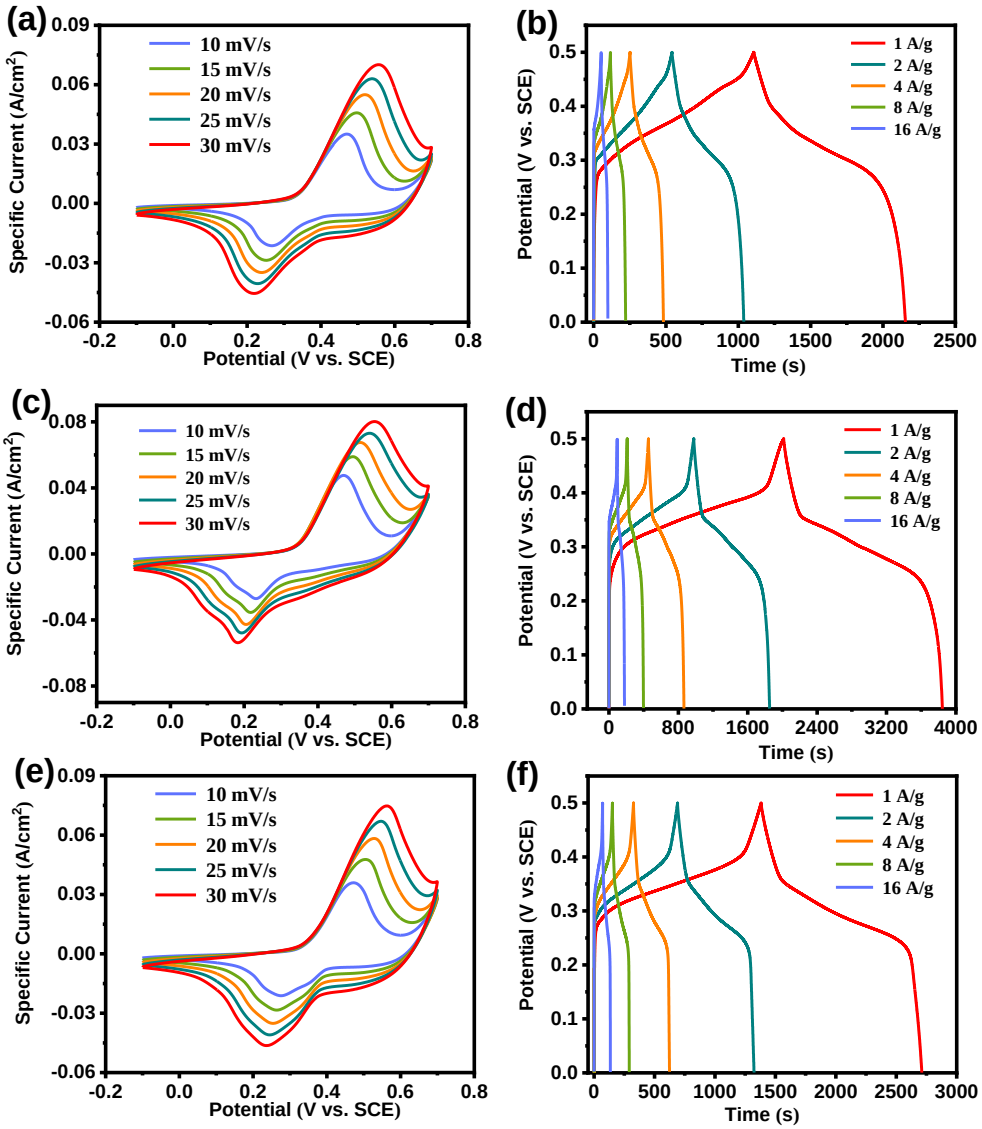


Figure S8 (a, c, e) CV curves of $NiCo_2O_4$, V-$NiCo_2O_4$-3, V-$NiCo_2O_4$-5 electrodes at different scan rates; (b, d, f) GCD curves of the $NiCo_2O_4$, V-$NiCo_2O_4$-3, V-$NiCo_2O_4$-5 electrodes at different specific currents.

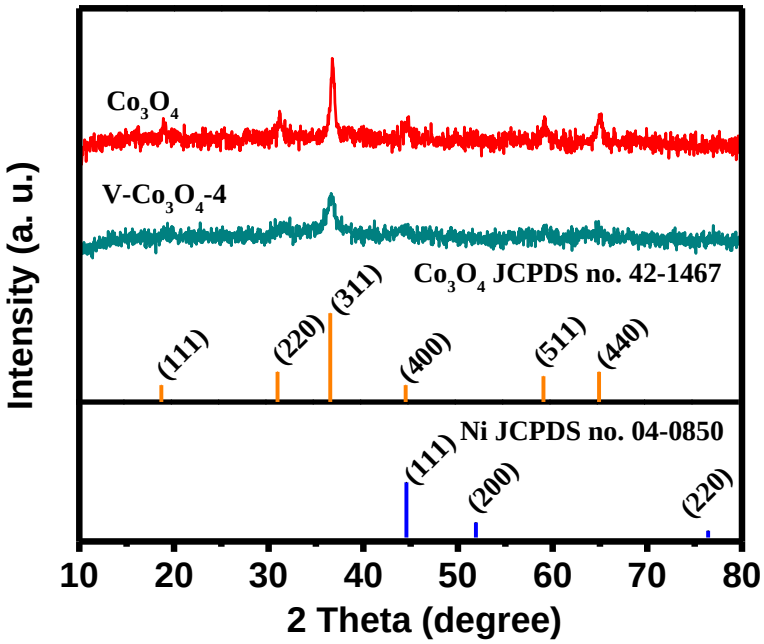


Figure S9 XRD patterns of Co_3O_4 and Co_3O_4-4.

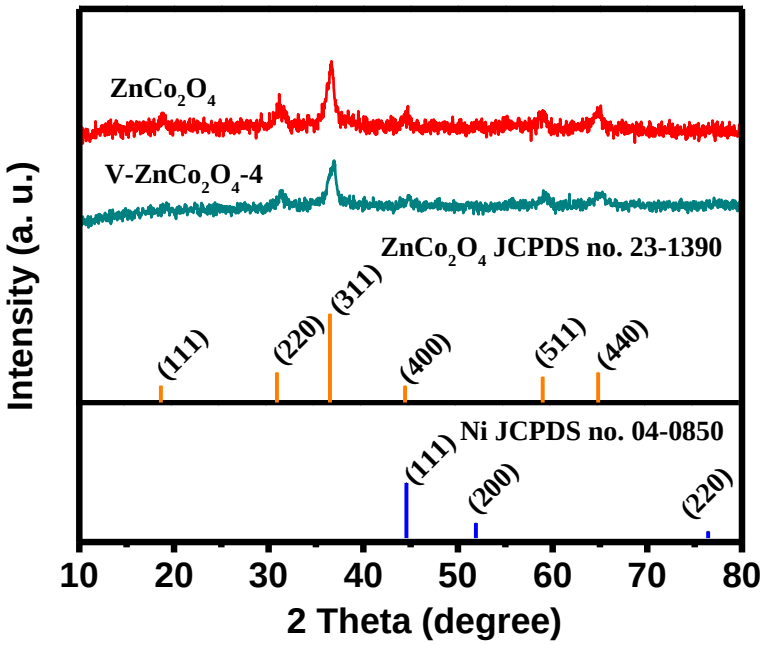


Figure S10 XRD patterns of $ZnCo_2O_4$ and V-$ZnCo_2O_4$-4.

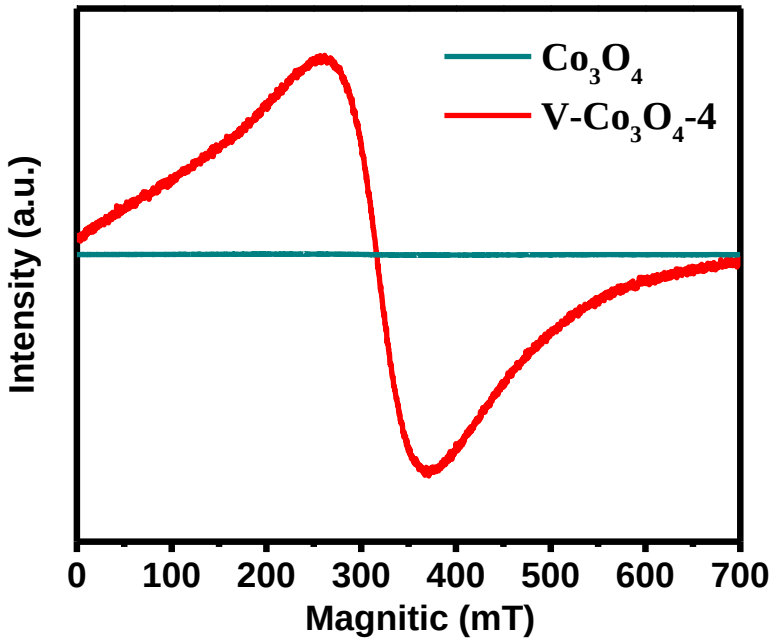


Figure S11 EPR spectra of Co_3O_4 and Co_3O_4-4.

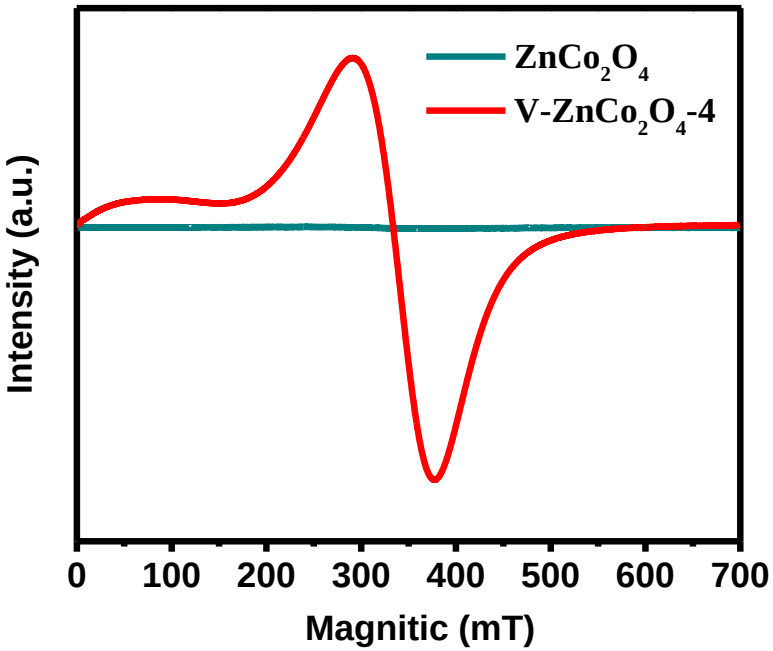
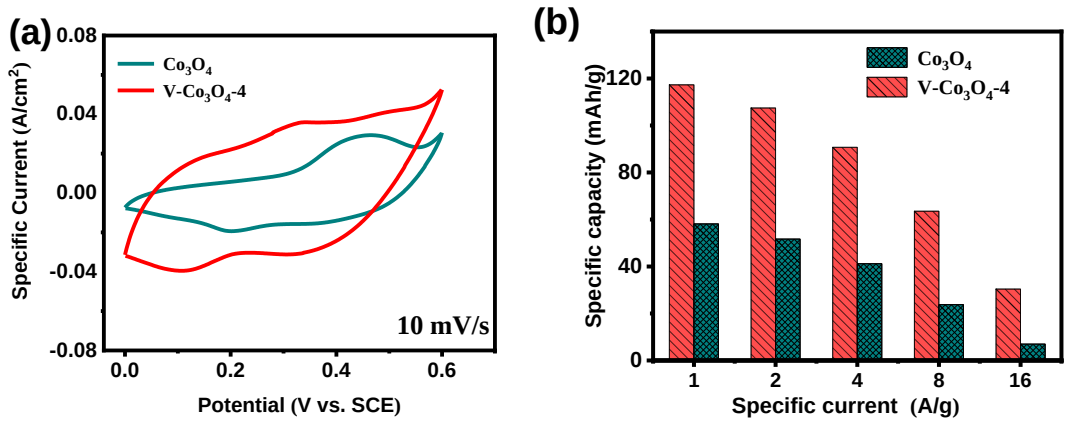


Figure S12 EPR spectra of $ZnCo_2O_4$ and V-$ZnCo_2O_4$-4.



ure S13 (a) CV curves of Co_3O_4 and Co_3O_4-4 at the scan rate of 10 $mV \cdot s^{-1}$; (b) Specific discharge capacitance at different current densities of Co_3O_4 and Co_3O_4-4 h.

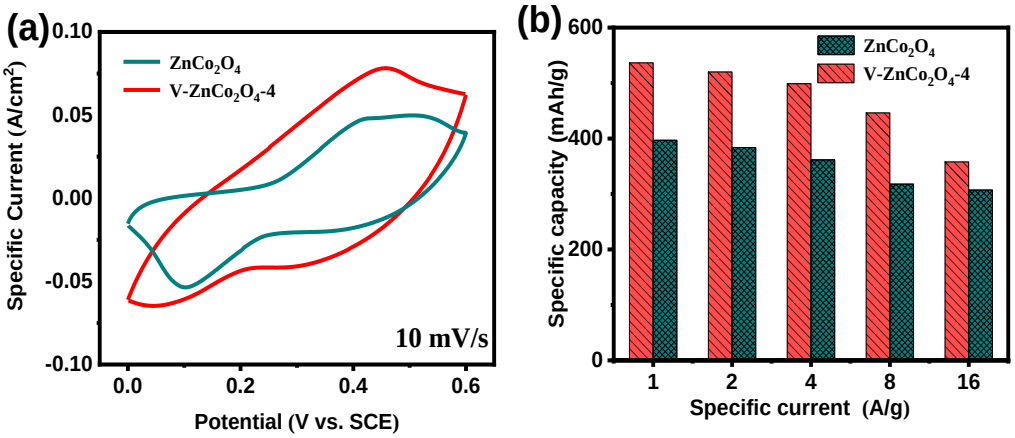


Figure S14 (a) CV curves of $ZnCo_2O_4$ and V-$ZnCo_2O_4$-4 at the scan rate of 10 $mV \cdot s^{-1}$; (b) Specific discharge capacitance at different current densities of $ZnCo_2O_4$ and V-$ZnCo_2O_4$-4.

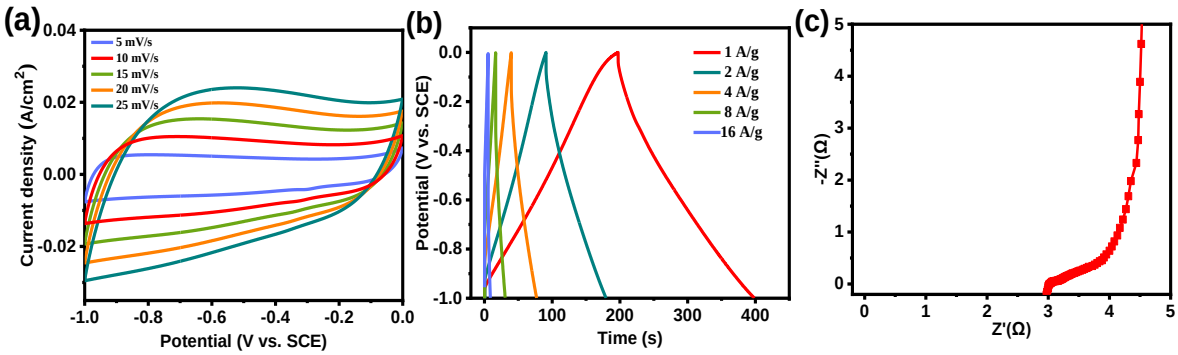


Figure S15 (a) CV curves of AC; (b) GCD curves of AC; (c) EIS curve of AC.

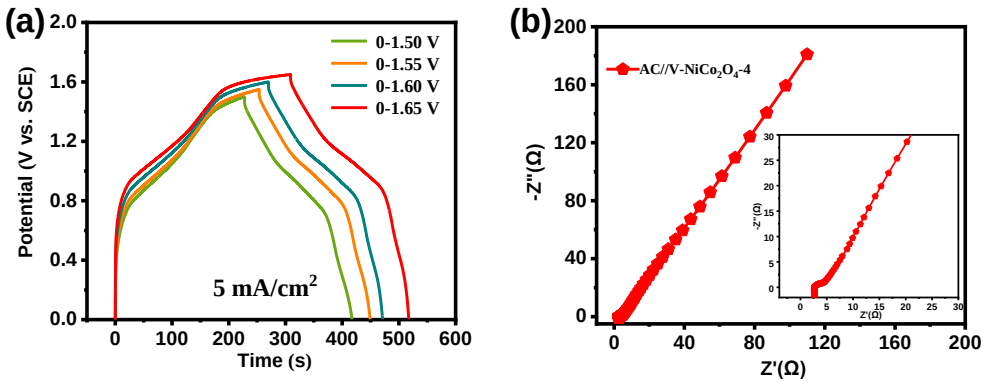


Figure S16 (a) GCD curves of the V-$NiCo_2O_4$ -4//AC HSC device at different voltages (from 1.5 to 1.8 V) at a specific current of 30 $mA \cdot cm^{-2}$; (b) EIS curves of the as-assembled V-$NiCo_2O_4$-4//AC HSC device.

Table S2. Comparison of the similar device properties of the oxygen-deficient metal oxide as cathode

Material	Electrolyte	Performance	Cycling stability of Device	Sr. No
L-$CuCo_2O_4$	3 M KOH	139.72mAh·g^{-1} at 1 A·g^{-1}	85.5% after 10000 cycles	1
150-N:$ZnCo_2O_4$	3 M KOH	422.73 mAh·g^{-1} at 5 A·g^{-1}	95.4% after 3000 cycles	2
OV-$MgCo_2O_4$	3 M KOH	54.11 mAh·g^{-1} at 1 A·g^{-1}	82% after 10000 cycles	3
OV-$ZnCo_2O_4$	6 M KOH	293.14 mAh·g^{-1} at 1 A·g^{-1}	No cycling	4
$ZnMoO_4$-OV	6 M KOH	209.12 mAh·g^{-1} at 1.4 A·g^{-1}	87.4% after 10000 cycles	5
P-$NiMoO_4$	1 M KOH	142.88 mAh·g^{-1} at 1.4 A·g^{-1}	98.7% after 5000 cycles	6
N-Bi_2MoO_6	6 M KOH	155.13 mAh·g^{-1} at 0.5 A·g^{-1}	79% after 10000 cycles	7
Ov-NiMn-LDH	2 M KOH	32.8.6 mAh·g^{-1} at 1 A·g^{-1}	No cycling	8
Co_3O_4@Co/NC-HN	3 M KOH	273.9 mAh·g^{-1} at 1 A·g^{-1}	92.6% after 4000 cycles	9
v-Co_3O_4/CC	2 M LiOH	51.75 mAh·g^{-1} at 1 A·g^{-1}	81.4% after 5000 cycles	10
N-GNTs@OV-Bi_2O_3	6 M KOH	196.47 mAh·g^{-1} at 1 A·g^{-1}	85% after 10000 cycles	11
Vo-NiCo LDH	6 M KOH	217.1 mAh·g^{-1} at 1 A·g^{-1}	75% after 10000 cycles	12
MoO_{3-x}	1 M H_2SO_4	273.33 mAh·g^{-1} at 5 A·g^{-1}	75% after 10000 cycles	13
Pd-Co_3O_4	6 M KOH	181.92 mAh·g^{-1} at 2 .06 A·g^{-1}	92.5% after 4000 cycles	14
L-$CoFe_2O_4$/C	2 M KOH	66.67 mAh·g^{-1} at 1 A·g^{-1}	No cycling	15
Ov-MnO_2@ MnO_2	1 M Na_2SO_4	125.67 mAh·g^{-1} at 1 A·g^{-1}	82% after10000 cycles	16
α-MnO_2	1 M KOH	204.53 mAh·g^{-1} at 1 A·g^{-1}	80.6% after10000 cycles	17
LOV-MnO_2	1 M Na_2SO_4	126.42 mAh·g^{-1} at 1 A·g^{-1}	92.2% after10000 cycles	18
V-$NiCo_2O_4$-4	**2 M KOH**	**751.67 mAh·g^{-1} at 1 A·g^{-1}**	**91.9% after 10000 cycles**	**This work**

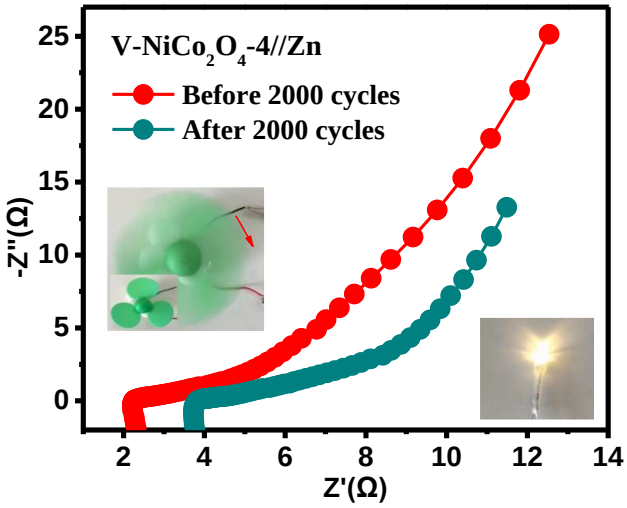


Figure S17 EIS curves of the as-assembled V-$NiCo_2O_4$-4//Zn batteries.

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