Supporting Information

Bioinspired Interfacial Engineering of $CoSe_2$ Decorated Carbon Framework Cathode towards Temperature-Tolerant and Flexible Zn-Air Batteries

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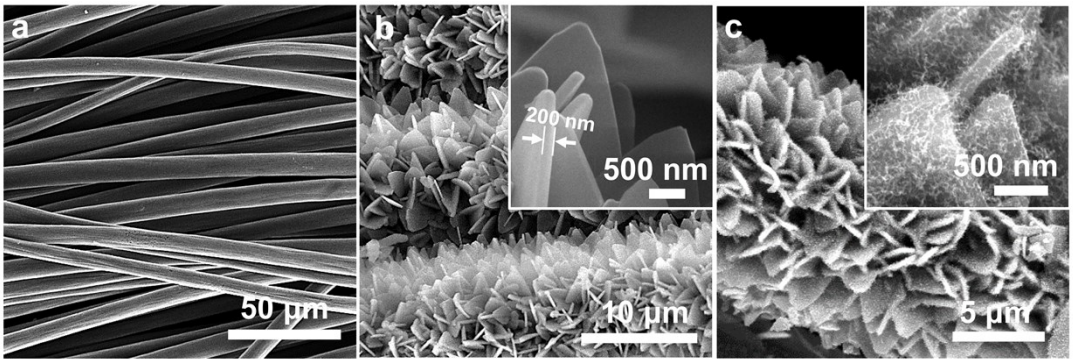

Fig. S1. SEM images of (a) carbon cloth, (b) Co-MOF NSA, and (c) Co-NCNT NSA grown on carbon cloths.

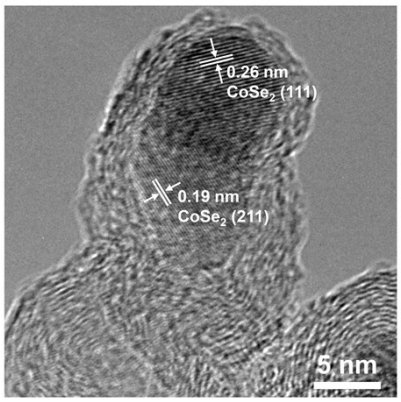

Fig. S2. HRTEM image of $CoSe_2$-NCNT NSA.

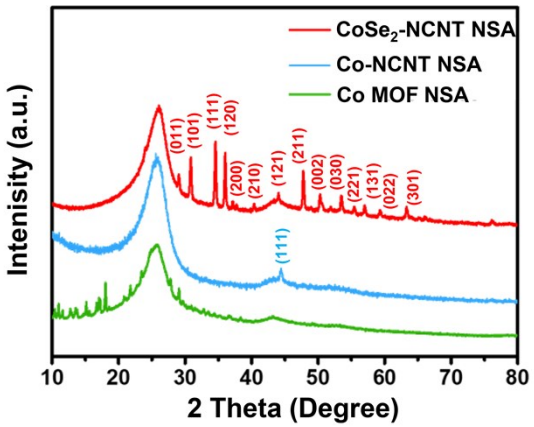

Fig. S3. XRD patterns of Co-MOF NSA, Co-NCNT NSA, and $CoSe_2$-NCNT NSA.

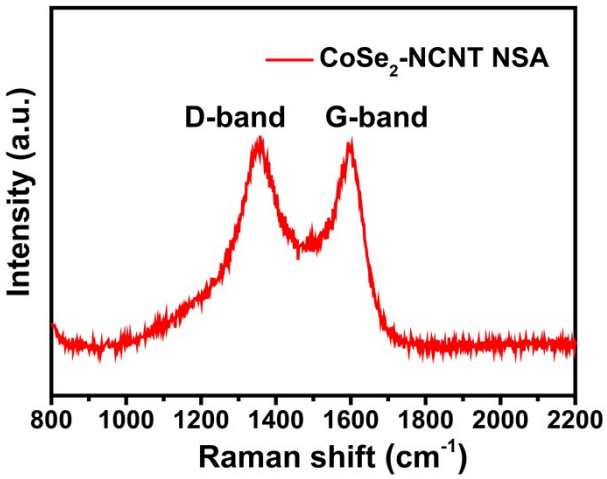

Fig. S4. Raman spectrum of the $CoSe_2$-NCNT NSA.

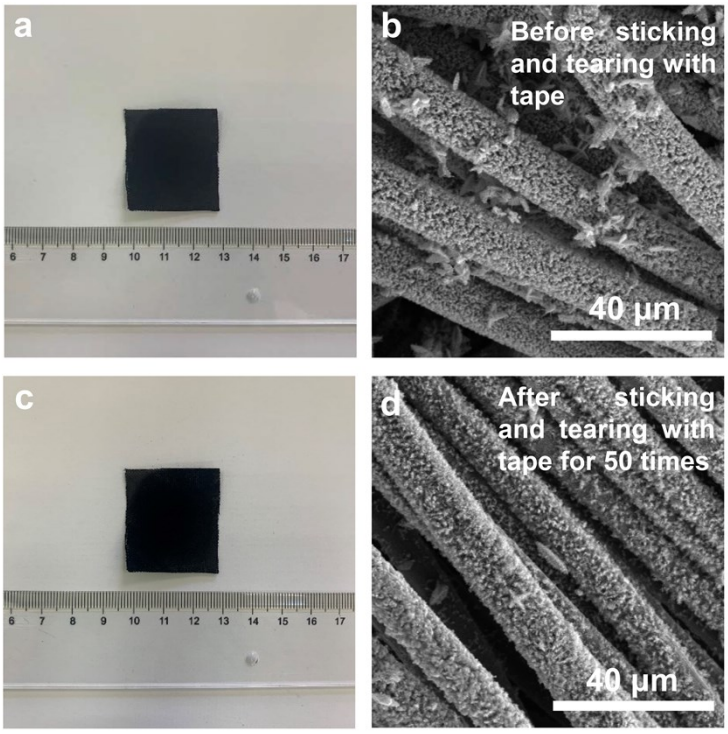

Fig. S5. Photographs and SEM images of the $CoSe_2$-NCNT NSA before (a,b), and after sticking and peeling off repeatedly with a tape for 50 times (c,d).

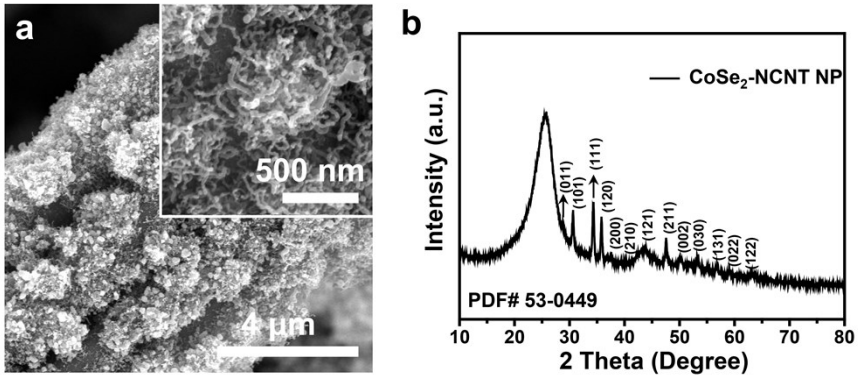

Fig. S6. (a) SEM images, and (b) XRD pattern of $CoSe_2$-NCNT NP.

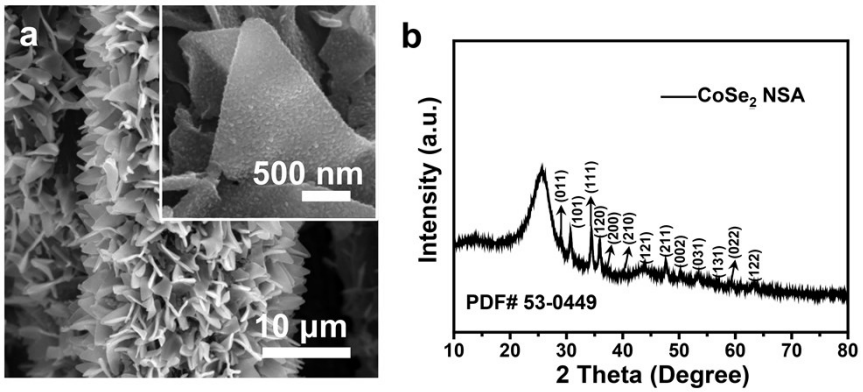

Fig. S7. (a) SEM images, and (b) XRD pattern of $CoSe_2$ NSA.

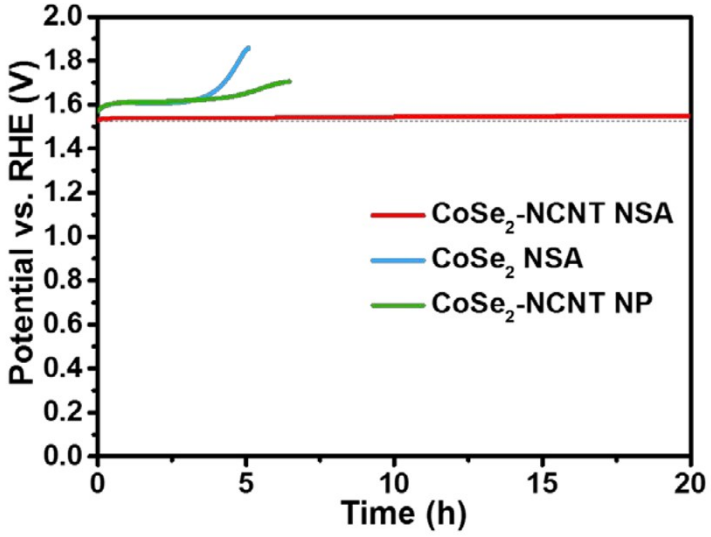

Fig. S8. OER stabilities of $CoSe_2$-NCNT NSA, $CoSe_2$-NCNT NP, and $CoSe_2$ NSA at 10 mA cm^{-2}.

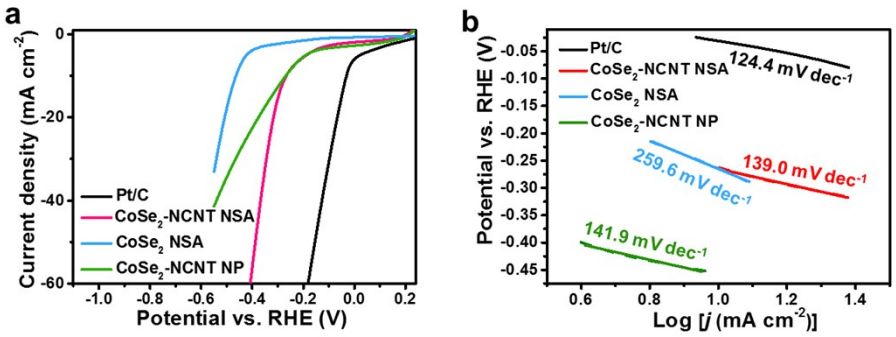

Fig. S9. (a) HER polarization curves, and (b) corresponding Tafel plots of $CoSe_2$-NCNT NSA, $CoSe_2$-NCNT NP, $CoSe_2$ NSA, and commercial Pt/C.

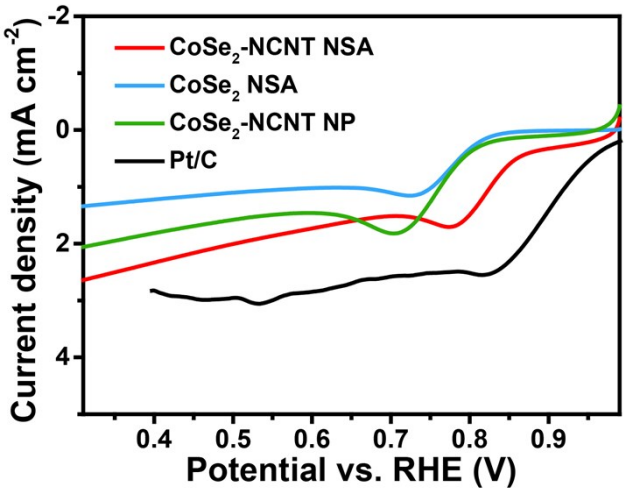

Fig. S10. ORR polarization curves of $CoSe_2$-NCNT NSA, $CoSe_2$-NCNT NP, $CoSe_2$ NSA, and commercial Pt/C.

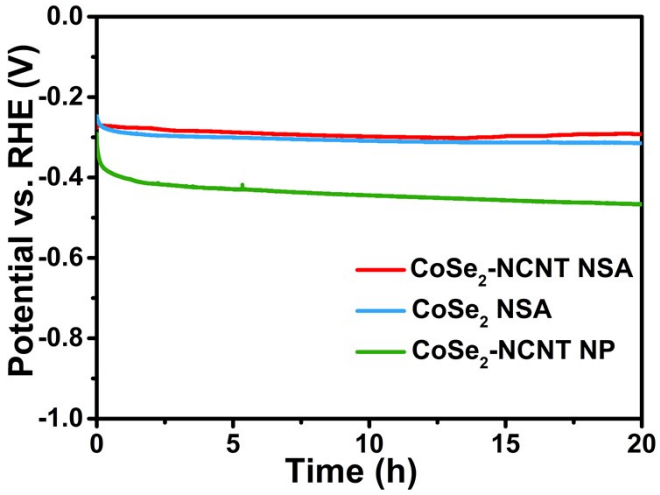

Fig. S11. HER stabilities of $CoSe_2$-NCNT NSA, $CoSe_2$-NCNT NP, and $CoSe_2$ NSA at 10 mA cm^{-2}.

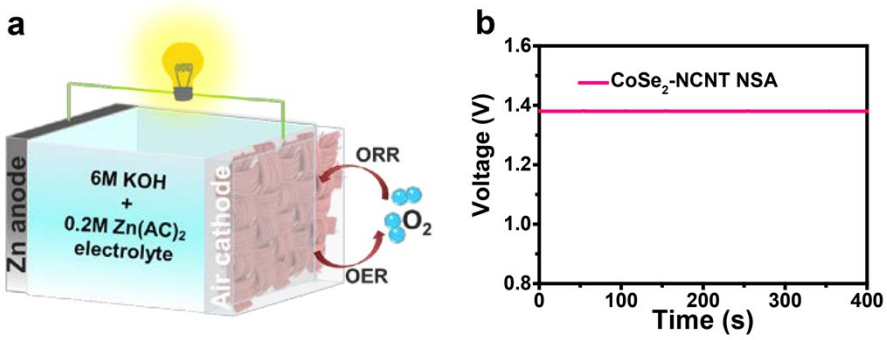

Fig. S12. (a) Schematic illustration of the configuration of Zn-air battery based on $CoSe_2$-NCNT NSA electrode with liquid electrolyte. (b) Open circuit plot of the fabricated Zn-air battery.

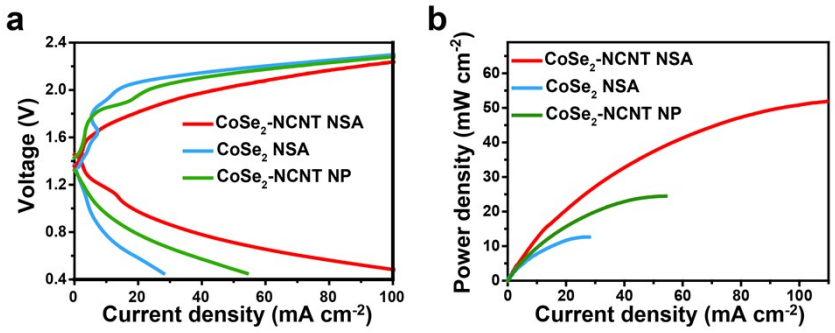

Fig. S13. (a) Charge-discharge polarization curves, and (b) power densities of $CoSe_2$-NCNT NSA, $CoSe_2$-NCNT NP, and $CoSe_2$ NSA-based Zn-air batteries.

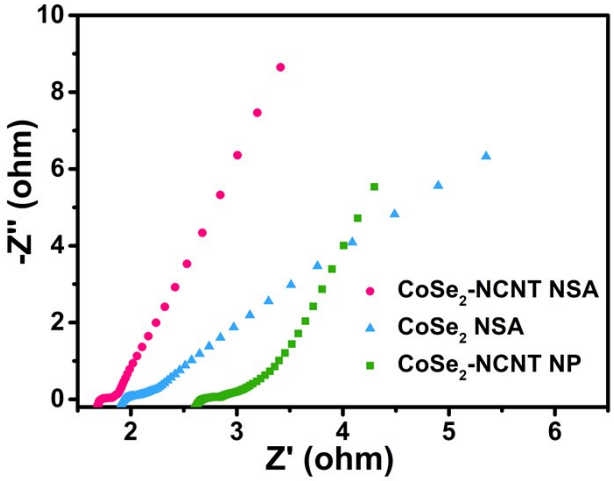

Fig. S14. Nyquist plots of $CoSe_2$-NCNT NSA, $CoSe_2$-NCNT NP, and $CoSe_2$ NSA.

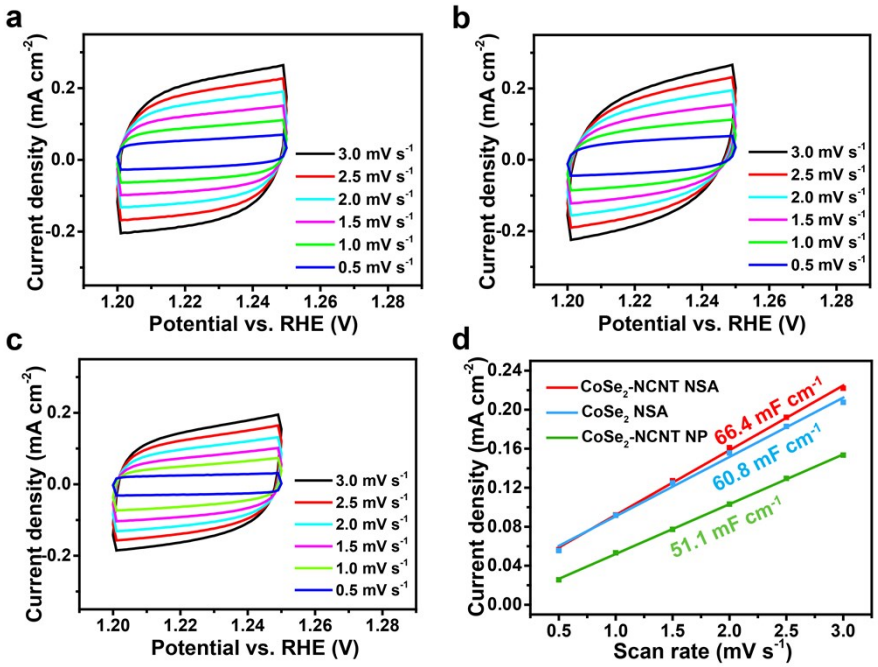

Fig. S15. CV curves of (a) $CoSe_2$-NCNT NSA, (b) $CoSe_2$-NCNT NP, and (c) $CoSe_2$ NSA in 1.0 M KOH in a non-Faradaic potential range at different scan rates. (d) Calculated C_{dl} for $CoSe_2$-NCNT NSA, $CoSe_2$-NCNT NP, and $CoSe_2$ NSA.

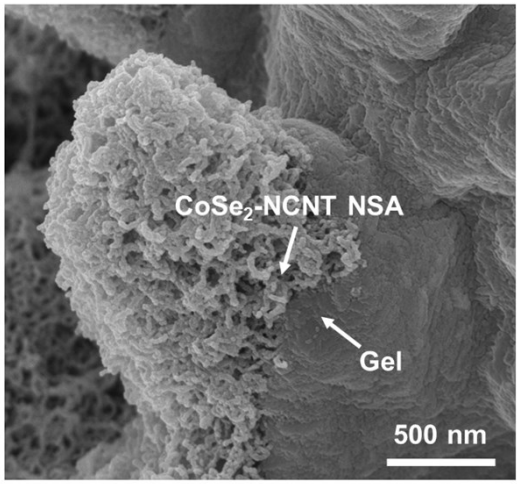

Fig. S16. High-magnification SEM image of the electrode-electrolyte interface of $CoSe_2$-NCNT NSA cathode.

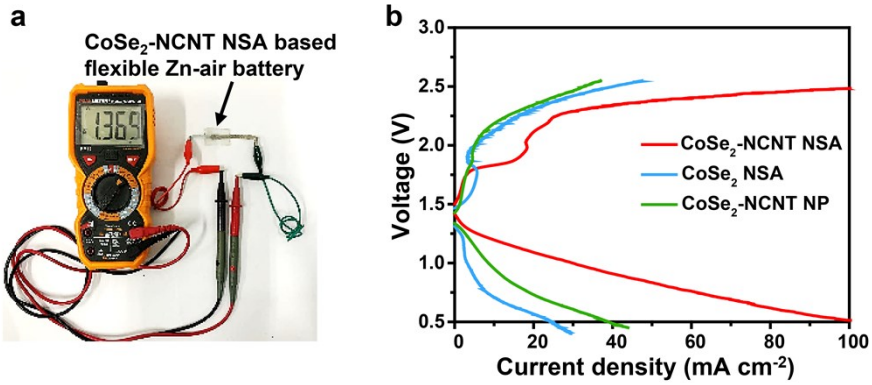

Fig. S17. (a) Photograph of the assembled flexible Zn-air battery showing an open-circuit voltage of ~1.37 V. (b) Charge-discharge polarization curves of the flexible Zn-air batteries based on $CoSe_2$-NCNT NSA, $CoSe_2$-NCNT NP, and $CoSe_2$ NSA electrodes.

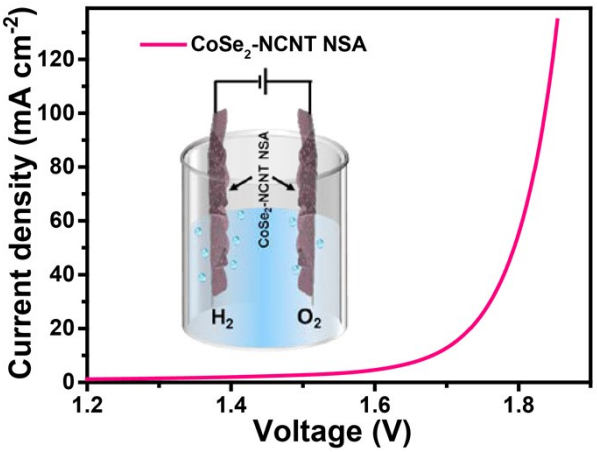

Fig. S18. Polarization curve of the overall water splitting device measured in 1.0 M KOH, which was driven by two $CoSe_2$-NCNT NSA-based flexible Zn-air batteries connected in series.

Table S1. Comparison of multifunctional electrocatalytic peformances.

Catalysts	OER η_{10} (V)	OER Tafel slope (V dec^{-1})	HER η_{10} (V)	ORR $\eta_{\text{half-wave potential}}$ (V)	References
CoS_x@Cu_2MoS_4-MoS_2/NSG	0.351	0.062	0.118	0.890	*Adv. Energy Mater.* **2020**, 10, 1903289
FeCo/Co_2P@NPCF	0.330	0.061	0.260	0.790	*Adv. Energy Mater.* **2020**, 10, 1903854
N,P-HCNF	0.320	0.248	0.550	0.820	*Nano Energy*, **2019**, 64, 103879.
N, P, F-graphene	0.390	0.136	0.520	0.760	*Angew. Chem. Int. Ed.*, **2016**, 55, 13296.
Co/CoO_x/perovskite	0.410	0.099	0.210	0.760	*Nano Energy*, **2017**, 32, 247.
$Co_{0.85}$Se@NC	0.320	0.075	0.230	0.830	*J. Mater. Chem. A*, **2017**, 5, 7001.
Fe_2P/Fe_4N@NC	0.410	0.177	0.232	0.800	*ACS Appl. Mater. Interfaces*, **2017**, 9, 32840.
Co/CoP-NC	0.300	0.044	0.180	0.830	*Mater. Horiz.*, **2018**, 5, 108.
CoP@SNG	0.350	0.068	0.174	0.790	*Nanoscale*, **2018**, 10, 14613.
O-modified NC	0.380	0.044	0.392	0.880	*ACS Appl. Mater. Interfaces*, **2018**, 10, 11678.
CoFe-$CoFe_2O_4$/NCNT	0.310	0.063	0.204	0.740	*ACS Appl. Mater. Interfaces*, **2018**, 10, 39828.
Co−N-C	0.350	0.060	0.180	0.880	*ACS Appl. Mater. Interfaces*, **2019**, 11, 39809.
Co/NGC	0.396	0.092	0.293	0.850	*ACS Appl. Mater. Interfaces*, **2020**, 12, 5717.
CoFe/NC NS	0.321	0.061	0.230	0.849	*ACS Sustainable Chem. Eng.*, **2019**, 7, 15278.
Mo_2C@NC/Co@NG	0.424	0.051	0.142	0.867	*Nanoscale*, **2019**, 11, 12563.
Co_3C-NB	0.435	0.090	0.154	0.757	*J. Mater. Chem. A*, **2019**, 7, 14904.
$CoSe_2$-NCNT NSA	0.290	0.056	0.265	0.830	This work

Table S2. Comparison of long-term stabilities of rechargeable Zn-air batteries using liquid electrolyte.

Air cathodes	Electrolyte	Voltage decaying rate (mV h^{-1})	Cycling time (h)	Reference
$CuCo_2O_4$/N-CNTs	6 M KOH + 0.2 M $Zn(Ac)_2$	0.965	40	*Adv. Funct. Mater.*, **2017**, 27, 1701833.
Co_4N/NC fibers	6 M KOH + 0.2 M $Zn(Ac)_2$	0.313	135	*J. Am. Chem. Soc.*, **2016**, 138, 10226.
Co, N, O-graphene	6 M KOH + 0.2 M $ZnCl_2$	5.770	62	*Adv. Mater.*, **2017**, 1703185.
$Co_{0.85}Se$@NC	6 M KOH + 0.2 M $Zn(Ac)_2$	6.730	30	*J. Mater. Chem. A*, **2017**, 5, 7001.
Co_3O_4-NC	6 M KOH + 0.1 M ZnAc	0.990	210	*Adv. Mater.*, **2017**, 29, 1704117.
CoFe-$CoFe_2O_4$/NCNT	6 M KOH.	2.156	21	*ACS Appl. Mater. Interfaces*, **2018**, 10, 39828
CoP@SNG	6 M KOH + 0.2 M $Zn(Ac)_2$	3.086	30	*Nanoscale*, **2018**, 10, 14613.
Co-NC	6 M KOH + 0.1 M ZnAc	0.994	180	*ACS Catal.*, **2018**, 8, 8961.
Co@NCNT HMS	6MKOH+0.2MZn (Ac)2	0.427	150	*J. Mater. Chem. A*, **2018**, 6, 15523.
$LaNiO_3$@ FeOOH NPs	6 M KOH + 0.2 M $Zn(Ac)_2$	0.630	167	*Appl. Catal. B: Environ.*, **2020**, 262, 118291.
$Co_2P_2O_7$ NS	6 M KOH + 0.2 M $ZnCl_2$	0.540	120	*J. Colloid Interf. Sci.*, **2020**, 563, 328.
Co/N-C	6 M KOH + 0.2 M $Zn(Ac)_2$	1.140	120	*ACS Appl. Mater. Interfaces*, **2020**, 12, 5717.
N, B-Co_3C	6 M KOH + 0.2 M $Zn(Ac)_2$	4.007	180	*J. Mater. Chem. A*, **2019**, 7, 14904.
Mo_2C@NC/Co@ NG	6 M KOH + 0.2 M $Zn(Ac)_2$	4.657	80	*Nanoscale*, **2019**, 11, 12563.
$CoSe_2$-NCNT NSA	6 M KOH + 0.2 M $Zn(Ac)_2$	0.238	235	This work