Electronic Supplementary Material (ESI) for Dalton Transactions.

Supportting Information

Local environment-mediated efficient electrocatalytic CO_2 to CO on Zn nanosheets

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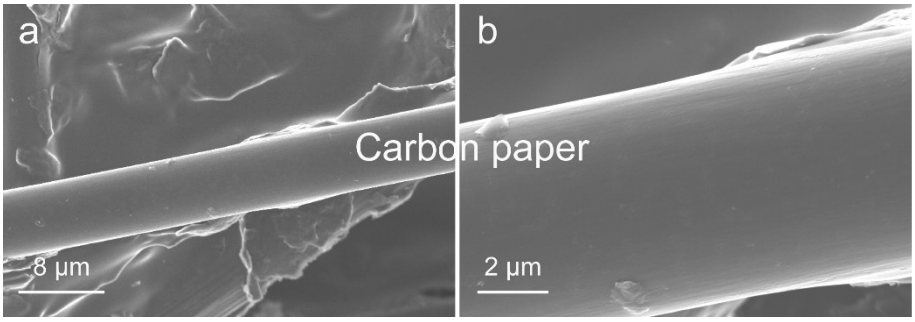

Fig. S1 (a, b) SEM images of carbon paper.

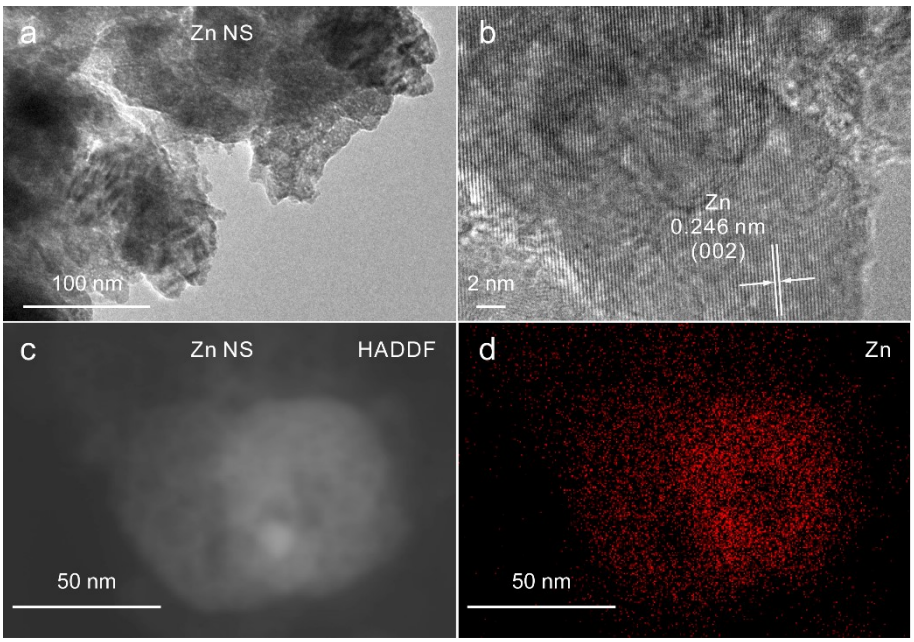

Fig. S2 (a, b) TEM images of Zn NS. (c, d) HADDF-STEM and EDS mapping images of Zn NS.

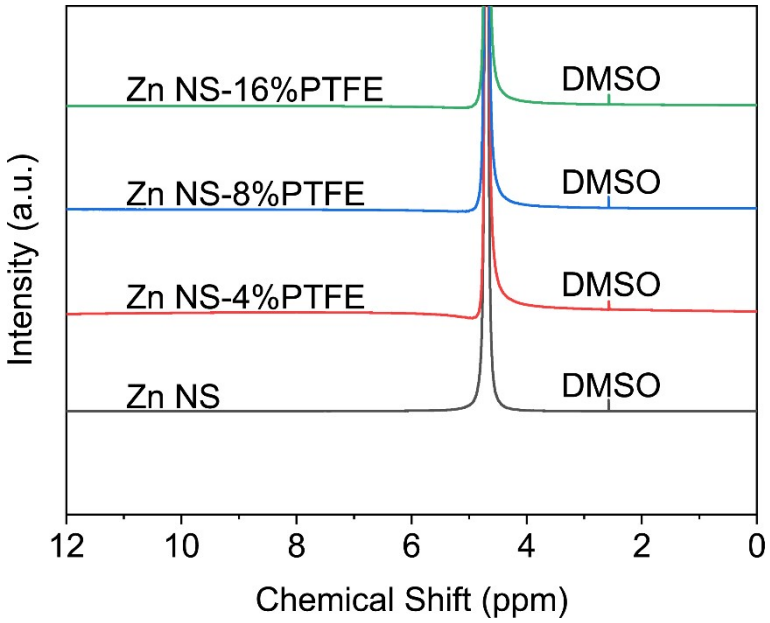

Fig. S3 The NMR spectra of electrolyte extracted from H-type cell after CO_2RR. It can be seen that no liquid products were formed from the spectra.

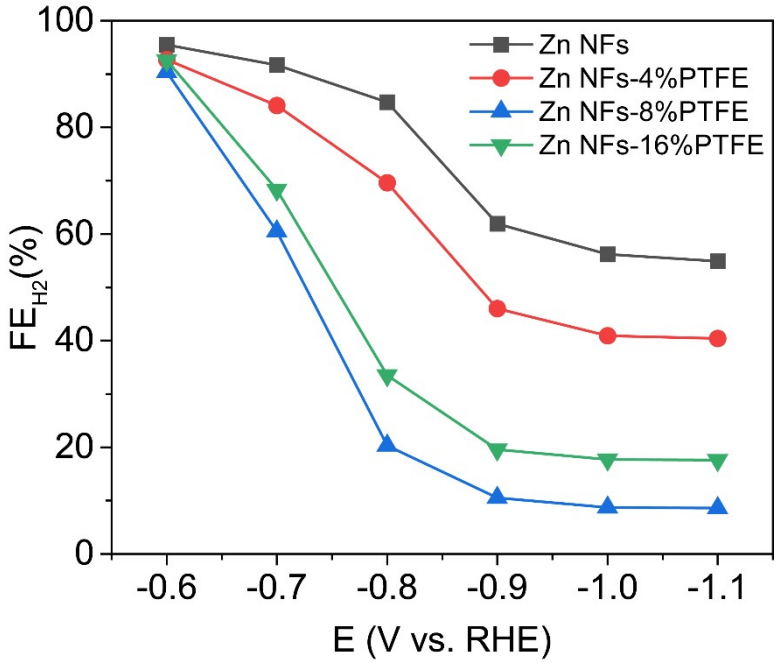

Fig. S4 H_2 Faradaic efficiency of Zn NS and Zn NS-x% PTFE (x = 4, 8, 16).

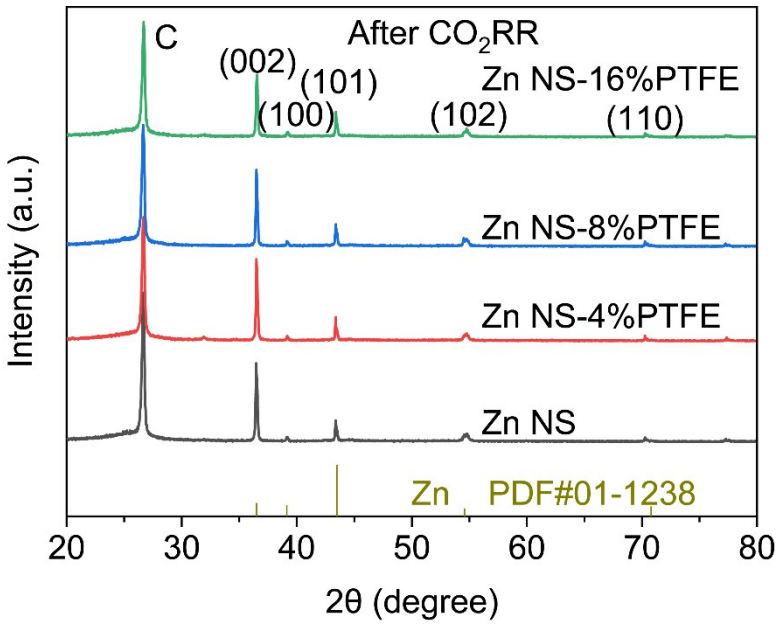

Fig. S5 XRD patterns of Zn NS and Zn NS-x% PTFE (x = 4, 8, 16) measured after CO_2RR.

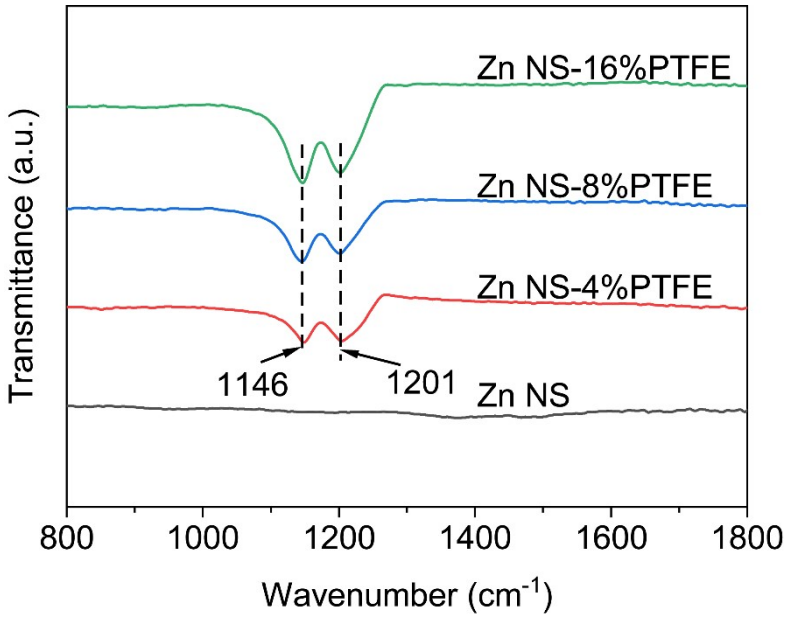

Fig. S6 FTIR spectra of the Zn NS and Zn NS-x% PTFE (x = 4, 8, 16) after CO_2RR.

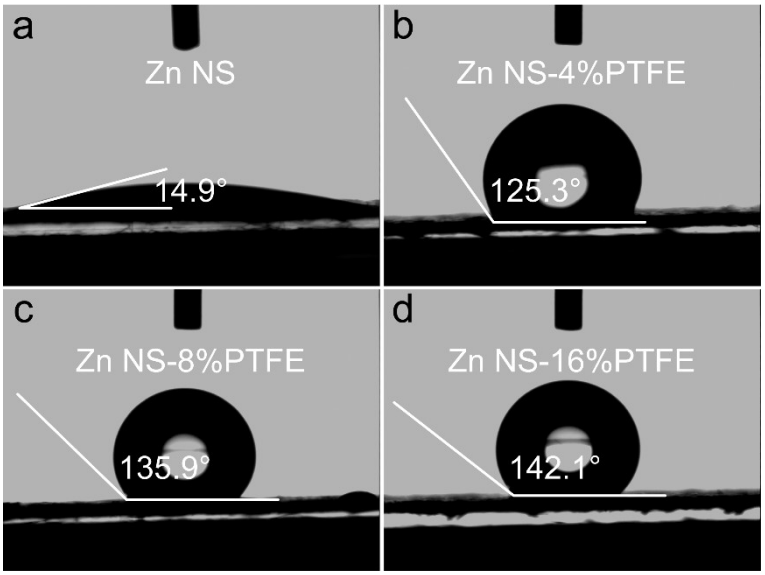

Fig. S7 Contact angle of (a) Zn NS, (b) Zn NS-4%PTFE, (c) Zn NS-8%PTFE, (d) Zn NS-16%PTFE with a drop of 0.5 M $KHCO_3$ electrolyte after CO_2RR.

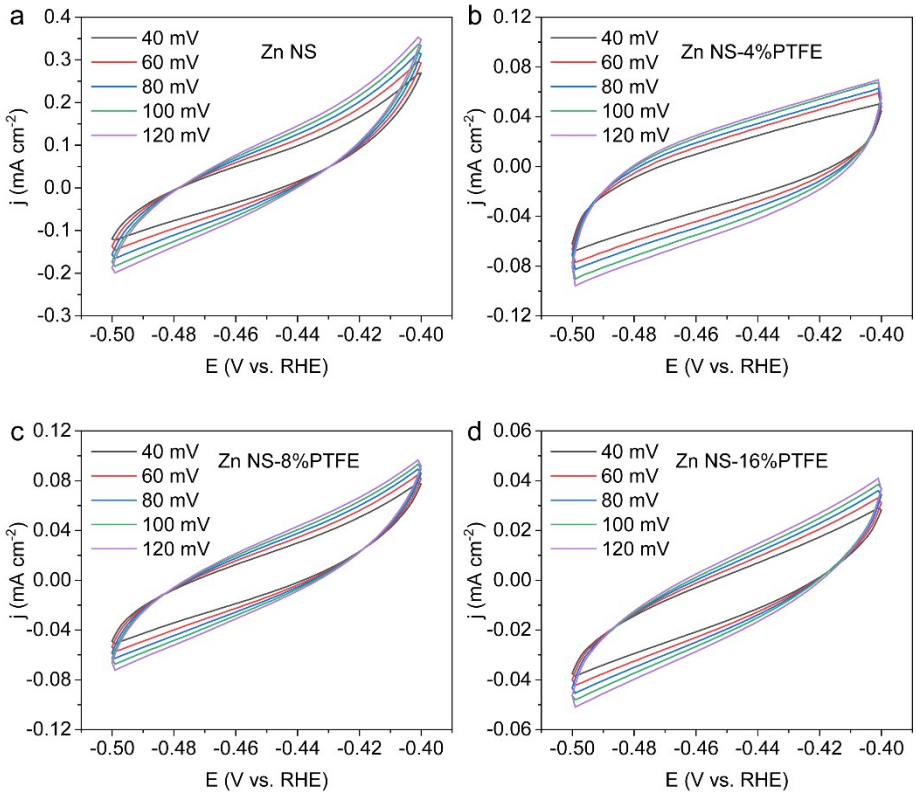

Fig. S8 CV curves of Zn NS and Zn NS-x% PTFE (x = 4, 8, 16) measured in 0.5 M $KHCO_3$ electrolyte at various scan rate (40, 60, 80, 100, and 120 mV s^{-1}).

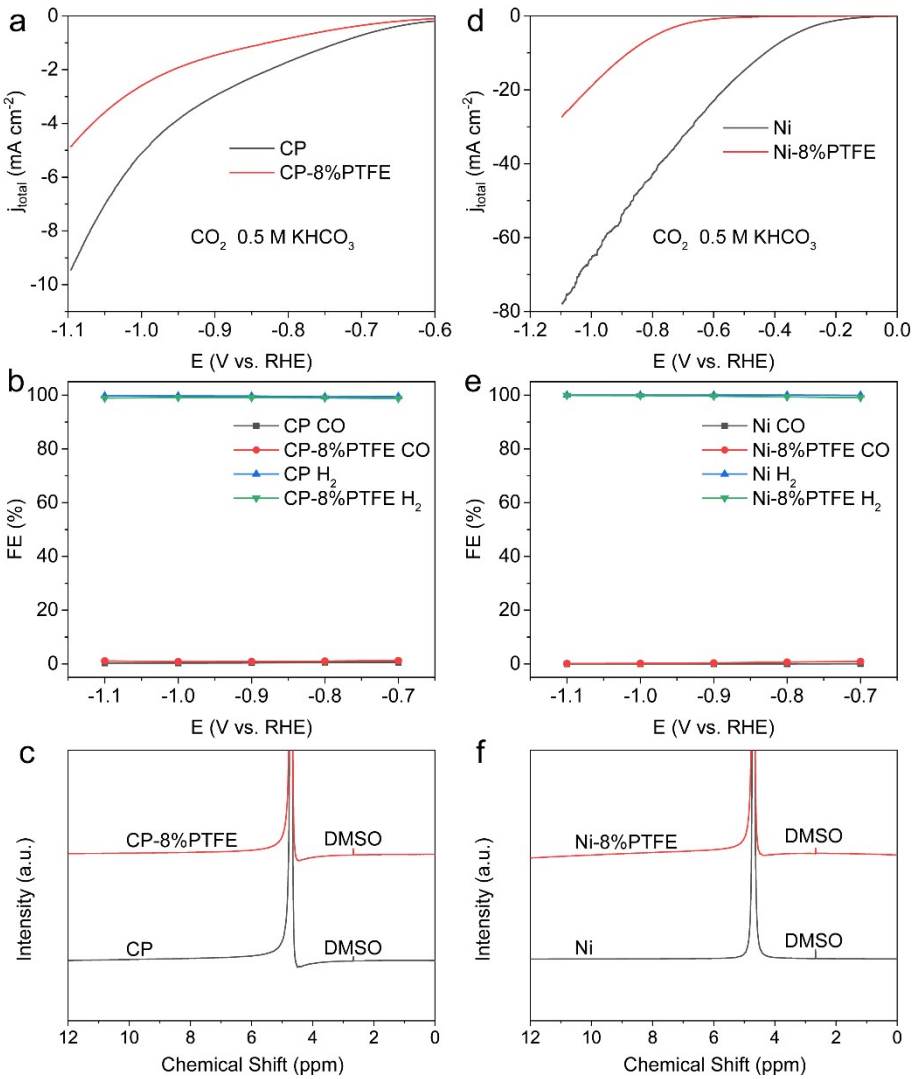

Fig. S9 (a ~ c) LSV, Faradaic efficiency (FE) and NMR spectra of CP and CP-8%PTFE. (d ~ f) LSV, Faradaic efficiency (FE) and NMR spectra of Ni and Ni-8%PTFE.

Electrode	Electrolyte	E vs. RHE (V)	FE_{CO} (%)	J_{CO} (mA cm^{-2})	Reference
Zn NS-8% PTFE	0.5 M $KHCO_3$	-1.0	90.2	-7.1	This work
Zn/Cu-Na200	0.1 M $KHCO_3$	-1.0	83.2	-9.8	1
$Sn_{0.5}Zn_{0.5}O_y$	0.1 M $KHCO_3$	-0.9	~ 24	-1.5	2
ZnLa-1	0.1 M $KHCO_3$	-1.5	25.99	-2.5	3
HP-Zn	0.1 M $KHCO_3$	-1.0	~ 85	-0.6	4
NiZn	0.1 M $KHCO_3$	-0.9	7	-1.7	5
Mo-ZnO	0.5 M $NaHCO_3$	-0.8	39.8	-1.8	6
ZnO@Cu	0.1 M $KHCO_3$	-1.1	~ 56	/	7
O-Zn-200	0.1 M $KHCO_3$	-1.0	79.7	-3.1	8
$Zn_{0.65}$-$Ni_{0.35}$	0.1 M KCl	-1.5 $V_{Ag/AgCl}$	55	-4.6	9

Table S1 Electrochemical CO_2 reduction to CO performance reported in the literature.

Electrode	Before	After
Zn NS	36.1°	14.9°
Zn NS-4% PTFE	128.3°	125.3°
Zn NS-8% PTFE	136.8°	135.9°
Zn NS-16% PTFE	143°	142.1°

Table S2 Contact angle of Zn NS and Zn NS-x% PTFE (x = 4, 8, 16) with a drop of 0.5 M $KHCO_3$ electrolyte (5 μL) before and after electrochemical performance testing.

References

1 P. Pinthong, P. Klongklaew, P. Praserthdam and J. Panpranot, *Nanomaterials*, 2021, **11**, 1671.

2 J. Jiang, B. Huang, R. Daiyan, B. Subhash, C. Tsounis, Z. Ma, C. Han, Y. Zhao, L. H. Effendi, L. C. Gallington, J. N. Hart, J. A. Scott and N. M. Bedford, *Nano Energy*, 2022, **101**, 107593.

3 Z. Liang, L. Song, M. Sun, B. Huang and Y. Du, *Science Advances*, 2021, **7**, eabl4915.

4 J. Wang, Z. Zhu, X. Wei, Z. Li, J. S. Chen, R. Wu and Z. Wei, *J. Phys. Chem. C*, 2021, **125**, 23784-23790.

5 X. Zhang, Y. Zhou, H. Zhang, H. Li, K. Liu, H. Li, H. Pan, J. Hu, J. Fu, S. Chen and M. Liu, *Journal of Energy Chemistry*, 2021, **63**, 625-632.

6 J. Wang, G. Wang, J. Zhang, Y. Wang, H. Wu, X. Zheng, J. Ding, X. Han, Y. Deng and W. Hu, *Angew. Chem. Int. Edit.*, 2021, **60**, 7602-7606.

7 S. B. Varandili, D. Stoian, J. Vavra, K. Rossi, J. R. Pankhurst, Y. T. Guntern, N. López and R. Buonsanti, *Chemical Science*, 2021, **12**, 14484-14493.

8 T.-L. Chen, H.-C. Chen, Y.-P. Huang, S.-C. Lin, C.-H. Hou, H.-Y. Tan, C.-W. Tung, T. S. Chan, J.-J. Shyue and H. M. Chen, *Nanoscale*, 2020, **12**, 18013-18021.

9 M. Beheshti, S. Kakooei, M. C. Ismail and S. Shahrestani, *Electrochim. Acta*, 2020, **341**, 135976.