Holey etching strategy of siloxene nanosheets to improve the rate performance of photo-assisted Li-O_2 batteries

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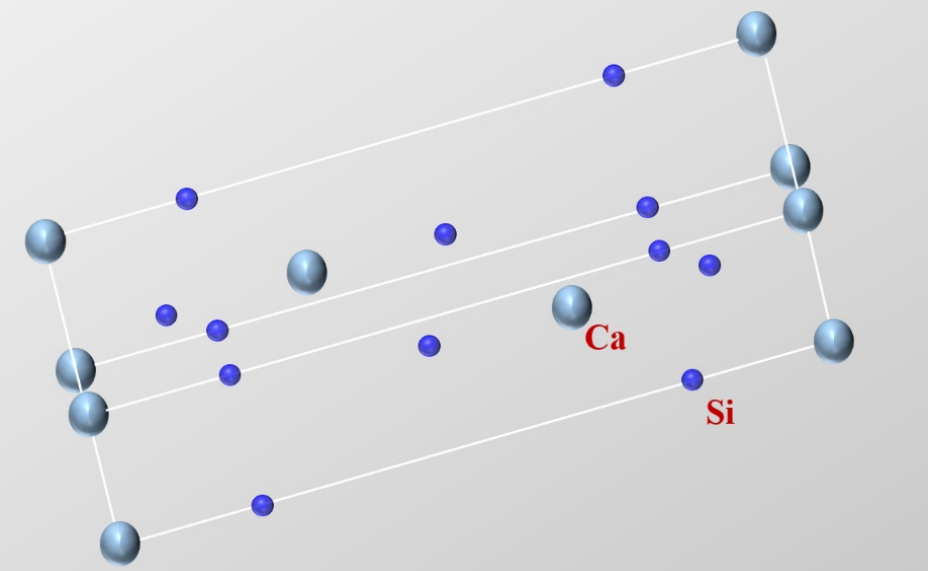

Figure S1. Crystal structure of $CaSi_2$ precursor.

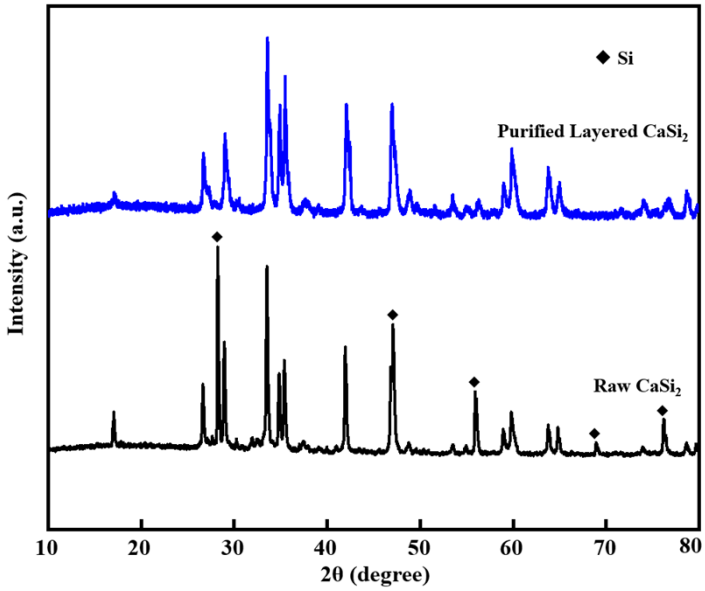

Figure S2. XRD patterns of raw $CaSi_2$ and purified layered $CaSi_2$.

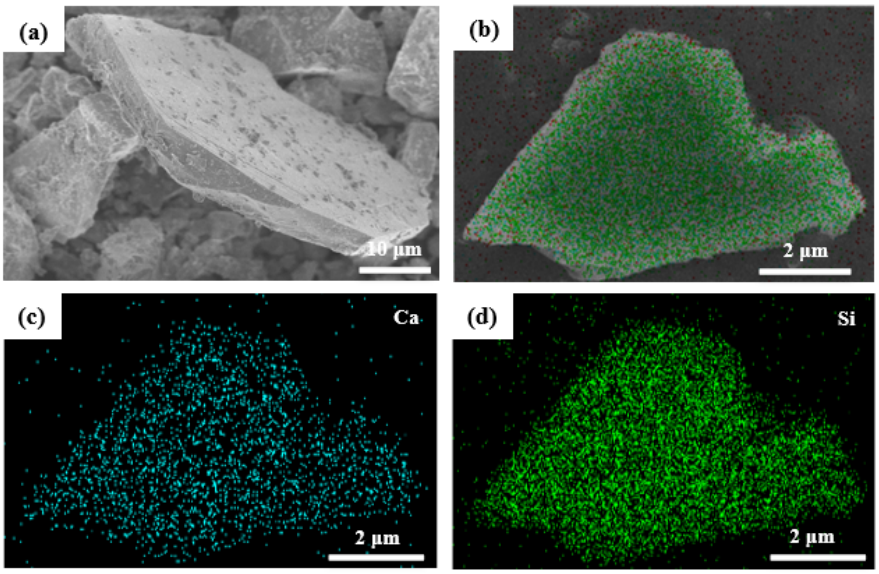

Figure S3. Morphology characteristic of purified layered $CaSi_2$: SEM image (a) and EDS-mapping images (b-d).

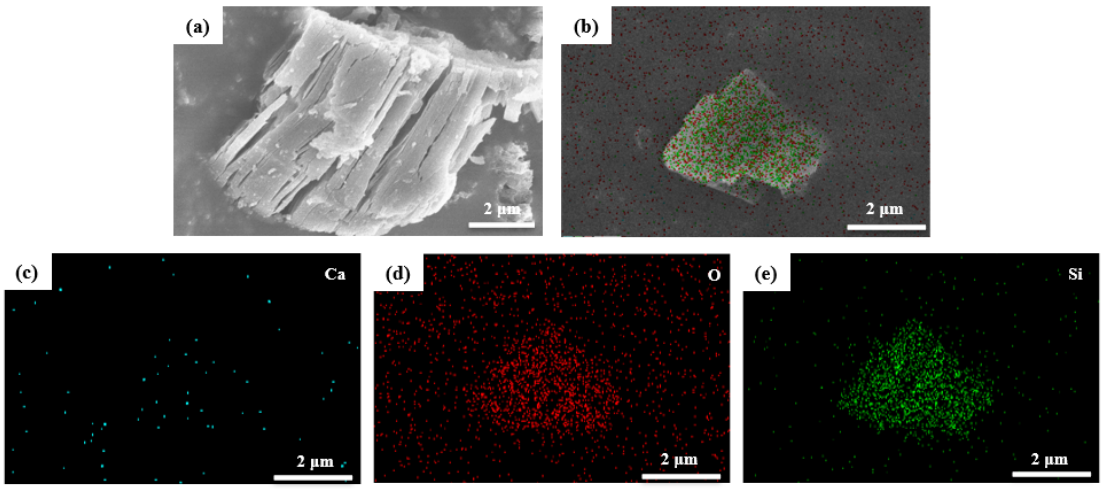

Figure S4. Morphology characterization of siloxene: SEM image (a) and the corresponding EDS-mapping images (b-e).

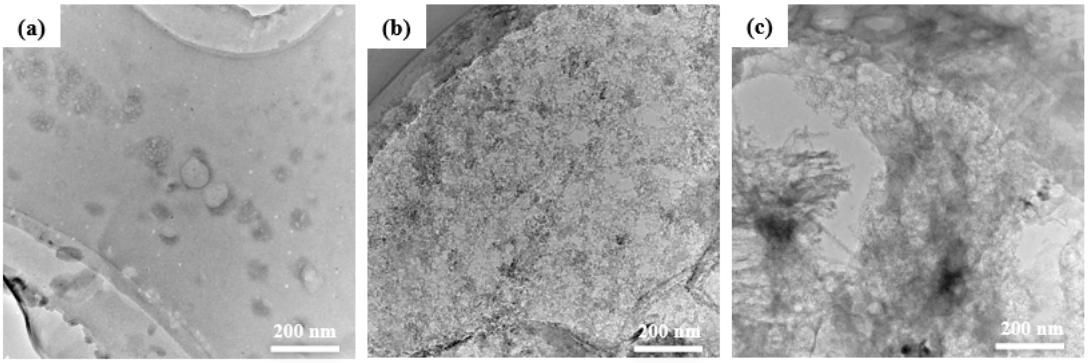

Figure S5. TEM images of the obtained products by treating siloxene NSs adsorbed Ag^+ ions with different concentrations at same $HF+H_2O_2$ solution: 0.005 mol L^{-1} (a), 0.02 mol L^{-1} (b), and 0.03 mol L^{-1} (c).

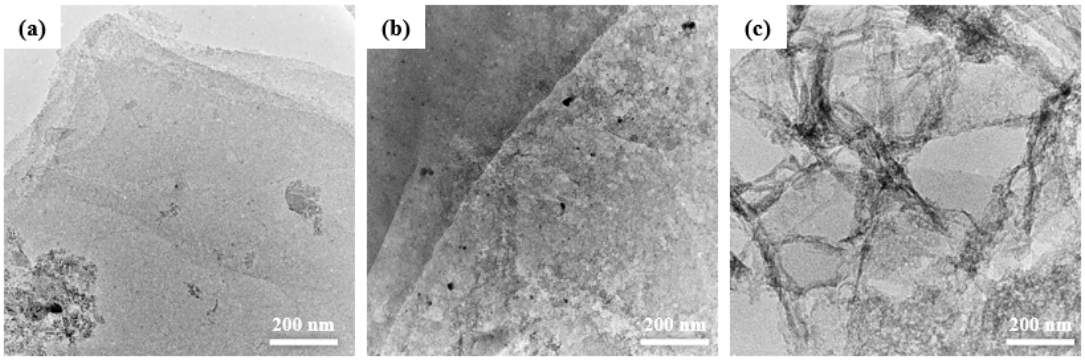

Figure S6. TEM images of the obtained products by treating siloxene NSs adsorbed 0.545 mol L^{-1} Ag^{+} ions with different HF concentrations: 0.452 mol L^{-1} (a), 0.753 mol L^{-1} (b), and 1.13 mol L^{-1} (c).

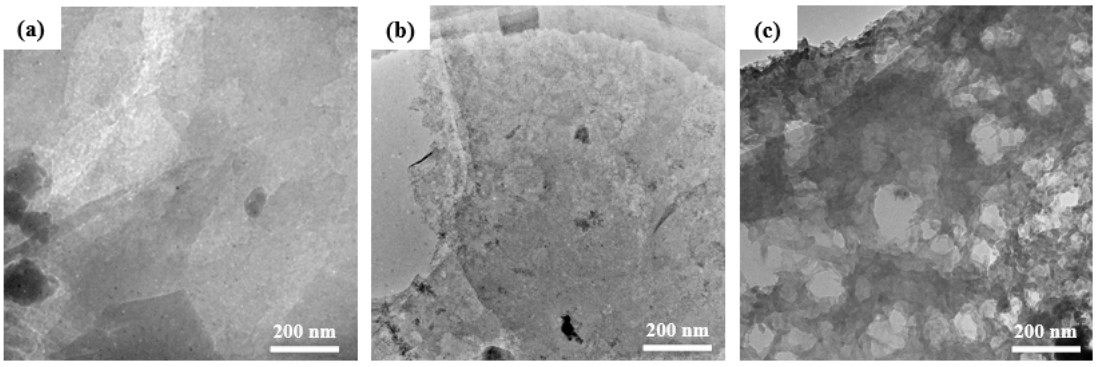

Figure S7. TEM images of the obtained products by treating siloxene NSs adsorbed 0.327 mol L^{-1} Ag^{+} ions with different H_2O_2 concentrations: 0.196 mol L^{-1} (a), 0.245 mol L^{-1} (b), and 0.49 mol L^{-1} (c).

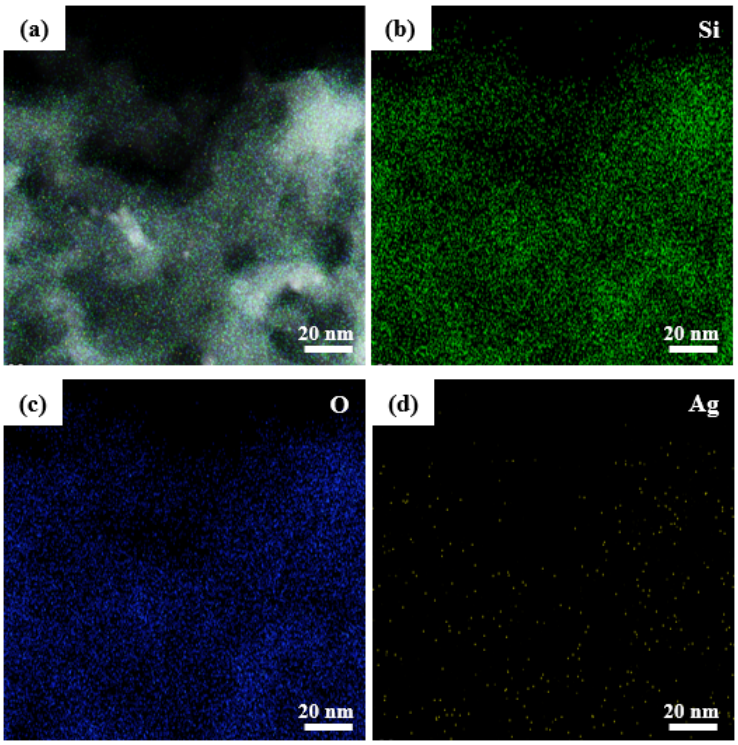

Figure S8. Energy spectrum images of P-siloxene NSs by spherical aberration electron microscope.

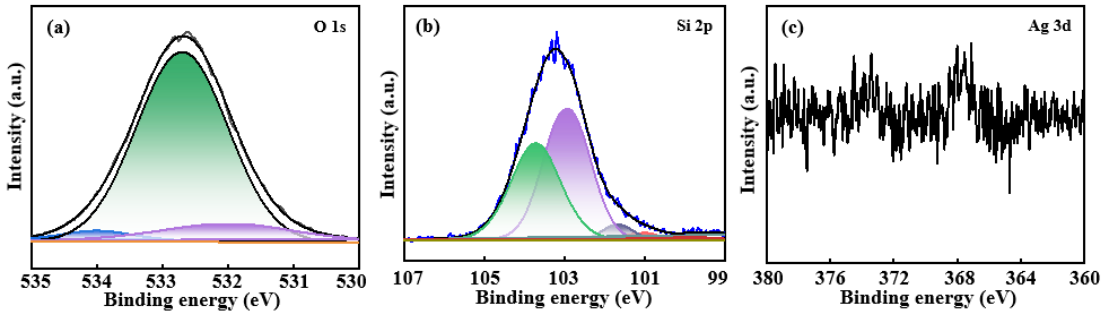

Figure S9. XPS spectra of P-siloxene NSs: O 1s spectrum (a), Si 2p spectrum (b), and Ag 3d spectrum (c).

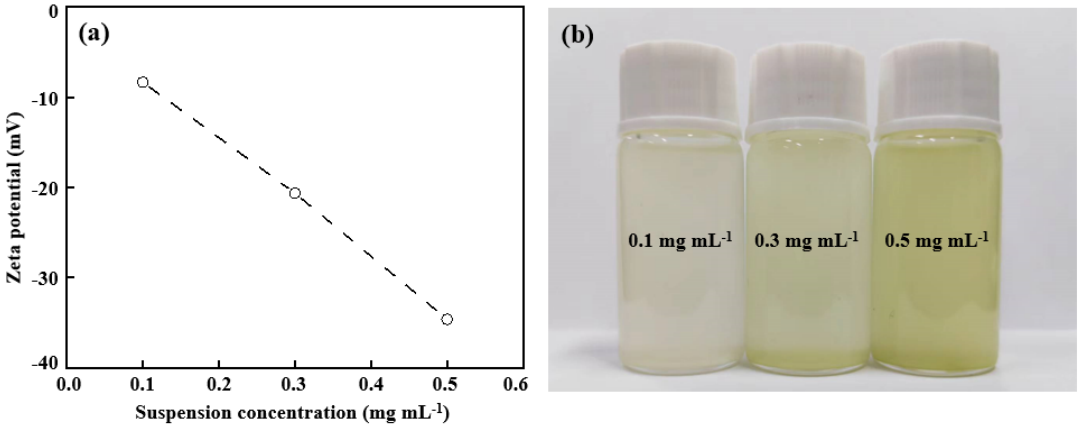

Figure S10. Zeta potential (a) and the optical images (b) of P-siloxene NSs suspensions with different concentrations.

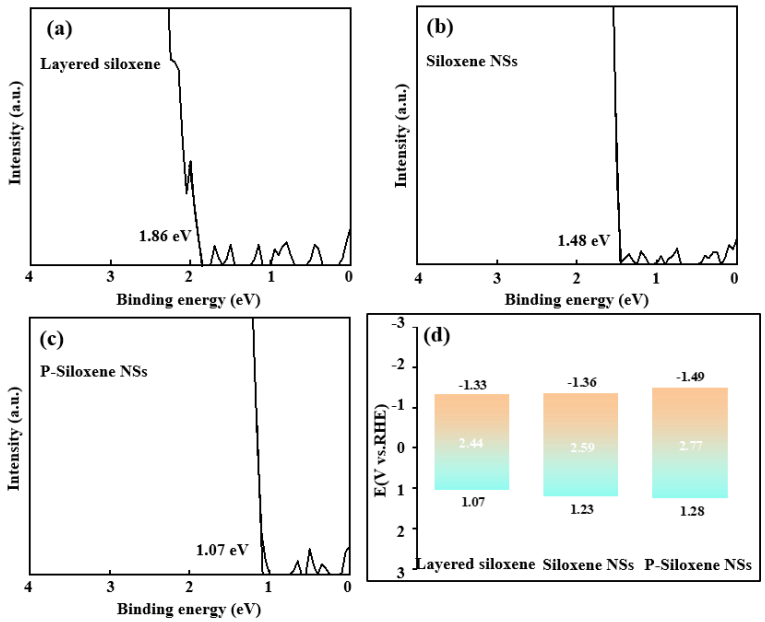

Figure S11. UPS and fitting curves of three siloxene photo-catalytic with different structures: layered siloxene (a), siloxene nanosheets (b), and P-siloxene NSs (c). Energy structure diagram of layered siloxene, siloxene NSs, and P-siloxene NSs.

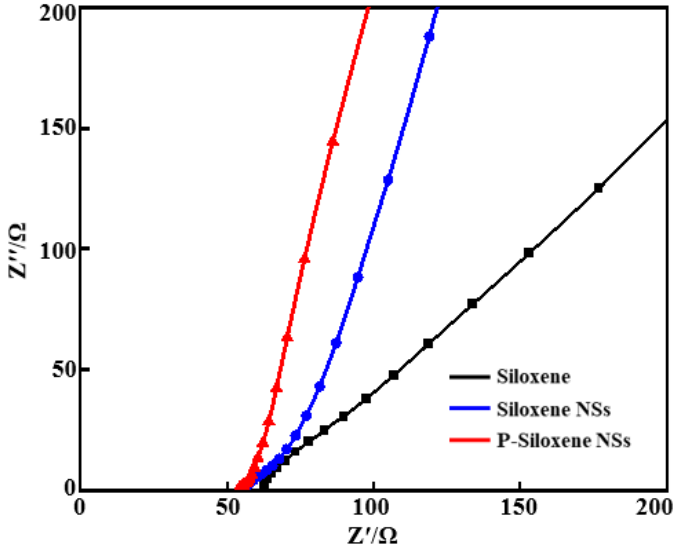

Figure S12. EIS spectra of layered siloxene, siloxene NSs, and P-siloxene NSs in three electrodes.

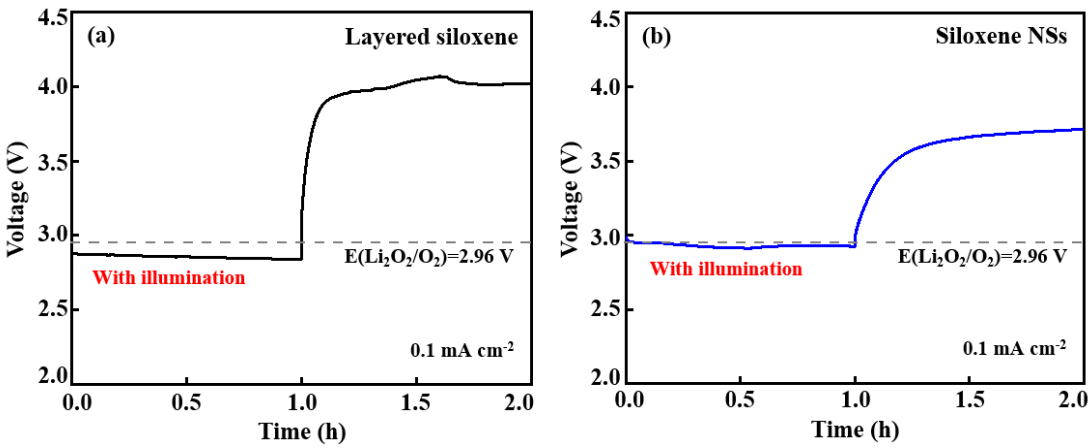

Figure S13. Charge and discharge potentials of photo-assisted Li-O_2 batteries assembled with layered siloxene (a) and siloxene NSs (b) with illumination.

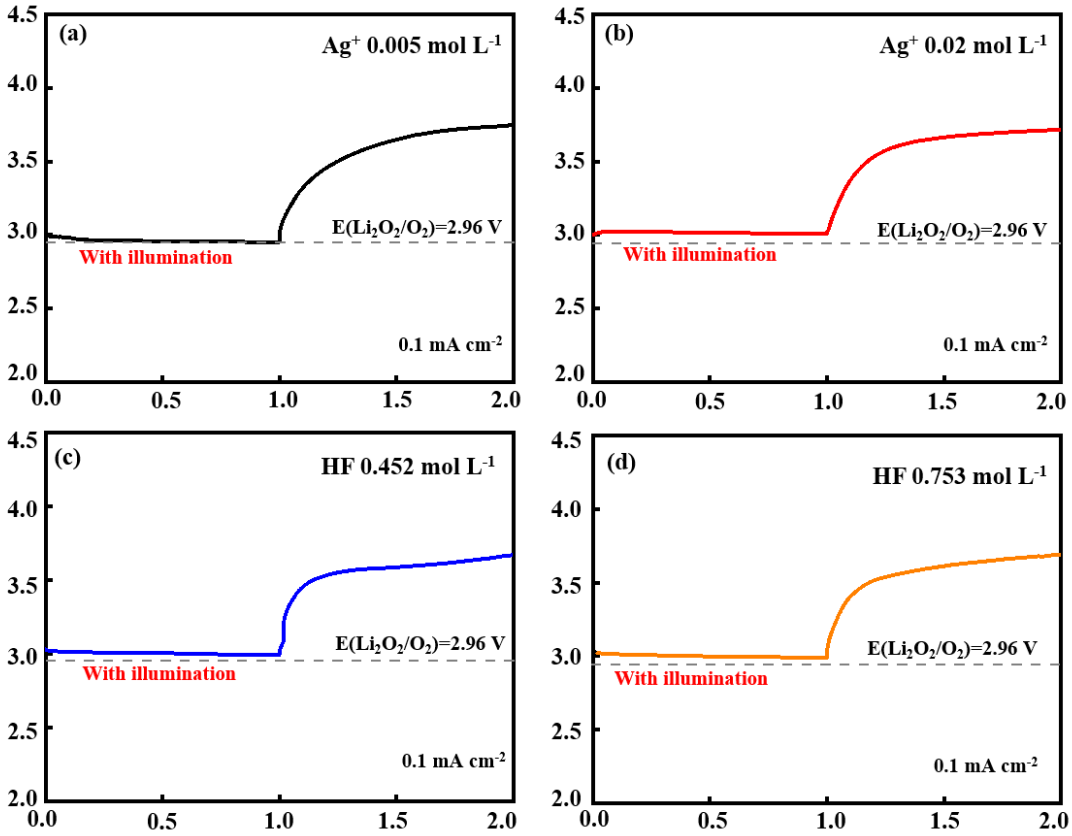

Figure S14. Charge and discharge potentials of photo-assisted Li-O_2 batteries assembled with Ag^+ concentrations of 0.005 mol L^{-1} (a), 0.02 mol L^{-1} (b) and HF concentrations of 0.452 mol L^{-1} (c), 0.753 mol L^{-1} (d) of P-siloxene NSs with illumination.

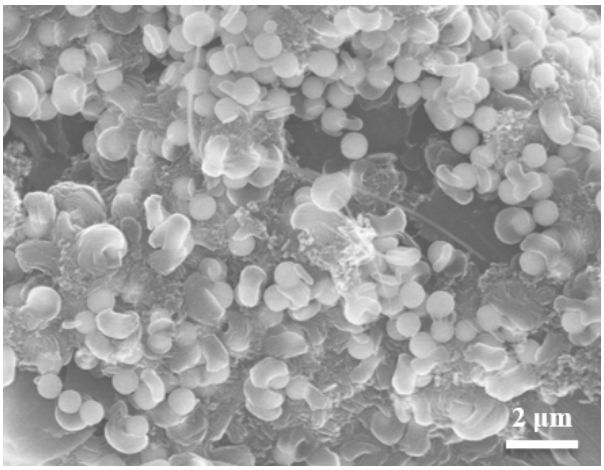

Figure S15. SEM image of Li_2O_2 intermediate product after complete discharge.

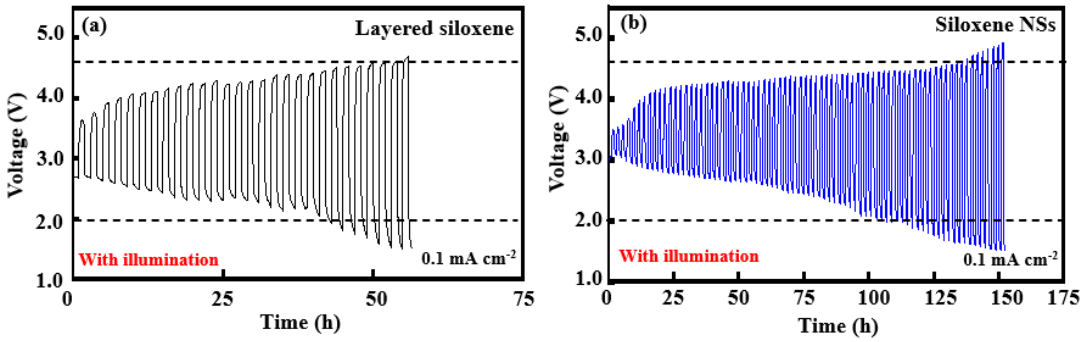

Figure S16. Cycle stability of photo-assisted Li-O_2 batteries assembled with layered siloxene (a) and siloxene NSs (b).

Table S1. Comparison of photo-electrochemistry performance for P-siloxene NSs photoelectrode with other reported photoelectrodes

Types of Photocatalysts	Current Density (mA cm^{-2})	Discharge voltage (V)	Charge voltage (V)	Round-trip efficiency (%)	Rate performance current density (mA cm^{-2})	Ref.
C_3N_4	0.04	3.22	3.38	95.3	3.10/3.50 0.15	1
g-C_3N_4	0.01	2.70	1.90	142	2.70/2.25 0.03	2
TiO_2/Fe_2O_3	0.01	3.01	3.20	94	2.80/3.75 0.05	3
Fe_2O_3/C_3N_4	0.1	3.13	3.19	98.2	2.75/3.50 0.4	4
Au/Nv-C_3N_4	0.05	3.16	3.26	97	3.00/3.27 0.25	5
Co-TABQ	0.1	3.12	3.32	94	3.60/2.80 0.5	6
Siloxene QDs	0.075	3.72	1.60	230	3.50/2.16 3.0	7
P-Siloxene NSs	0.1	3.05	3.40	90	2.87/3.82 0.8	This study

Supplementary References

1. Z. Zhu, X. Shi, G. Fan, F. Li, J. Chen, *Angew. Chem. Int. Ed.* 2019, **131**, 19197-19202.
2. Y. Liu, N. Li, K. Liao, Q. Li, M. Ishida, H. Zhou, *J. Mater. Chem. A* 2016, **4**, 12411-12415.
3. M. Li, X. Wang, F. Li, L. Zheng, J. Xu, J. Yu, *Adv. Mater.* 2020, **32**, 1907098.
4. Z. Zhu, Q. Lv, Y. Ni, S. Gao, J. Geng, J. Liang, F. Li, *Angew. Chem. Int. Ed.* 2022, **134**, 202116699.
5. Z. Zhu, Y. Ni, Q. Lv, J. Geng, W. Xie, F. Li, J. Chen, *PNAS* 2021, **118**, 2024619118.
6. Q. Lv, Z. Zhu, S. Zhao, L. Wang, Q. Zhao, F. Li, L. A. Archer, J. Chen, *J. Am. Chem. Soc.* 2021, **143**, 1941-1947.
7. C. Jia, F. Zhang, N. Zhang, Q. Li, X. He, J. Sun, R. Jiang, Z. Lei, Z.-H. Liu, *ACS Nano* 2023, **17**, 1713-1722.