Electronic supplementary information (ESI)

Single-Side Functionalized Graphene as Promising Cathode Catalysts in Nonaqueous Lithium-Oxygen Batteries

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Additional computational details and analysis on stability

The phonon calculations were carried out by using the finite displacement method as implemented in the PHONOPY code.[1] As shown in **Fig. S1**, there is no imaginary frequency found in all the calculated phonon dispersion spectra of the C_nX. Therefore, we confirmed that the C_nX are dynamically stable structures. First-principles molecular dynamics (FPMD) simulations were carried out by DMol[3]. Each MD simulation in the constant-volume and constant-temperature (NVT) ensemble lasted for 5 ps with a time step of 1 fs, and the temperature oscillations were controlled using the algorithm of massive generalized Gaussian moments. The temperature was set as 300 K to to assess the thermal stability of the studied C_nX under room temperature. As we can see in **Fig. S2**, the structures and bonding of C_nX are merely changed after 5 ps MD under 300 K. Thus the C_nX could keep stable under room temperature, which is enough for their applications in nonaqueous Li-O_2 battery.

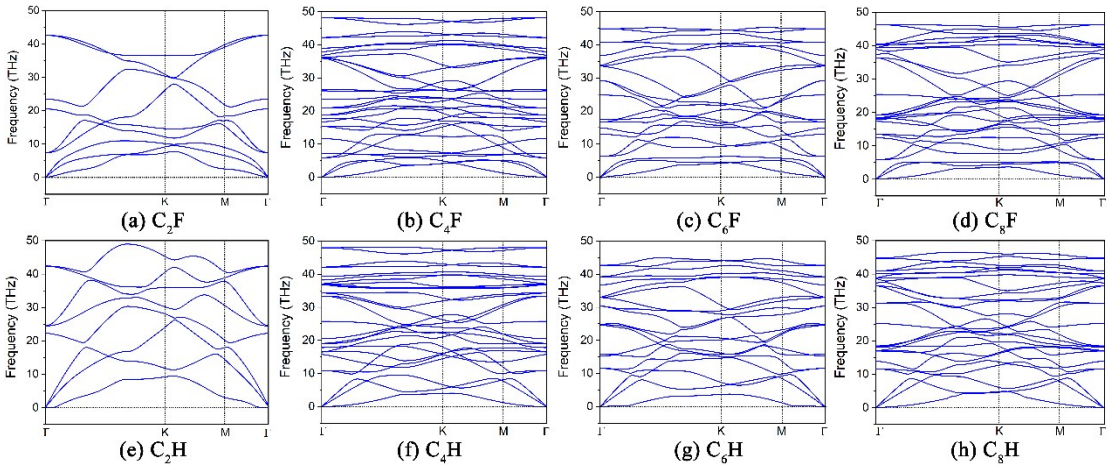


Fig. S1 Calculated phonon dispersion spectra of the C_nX.

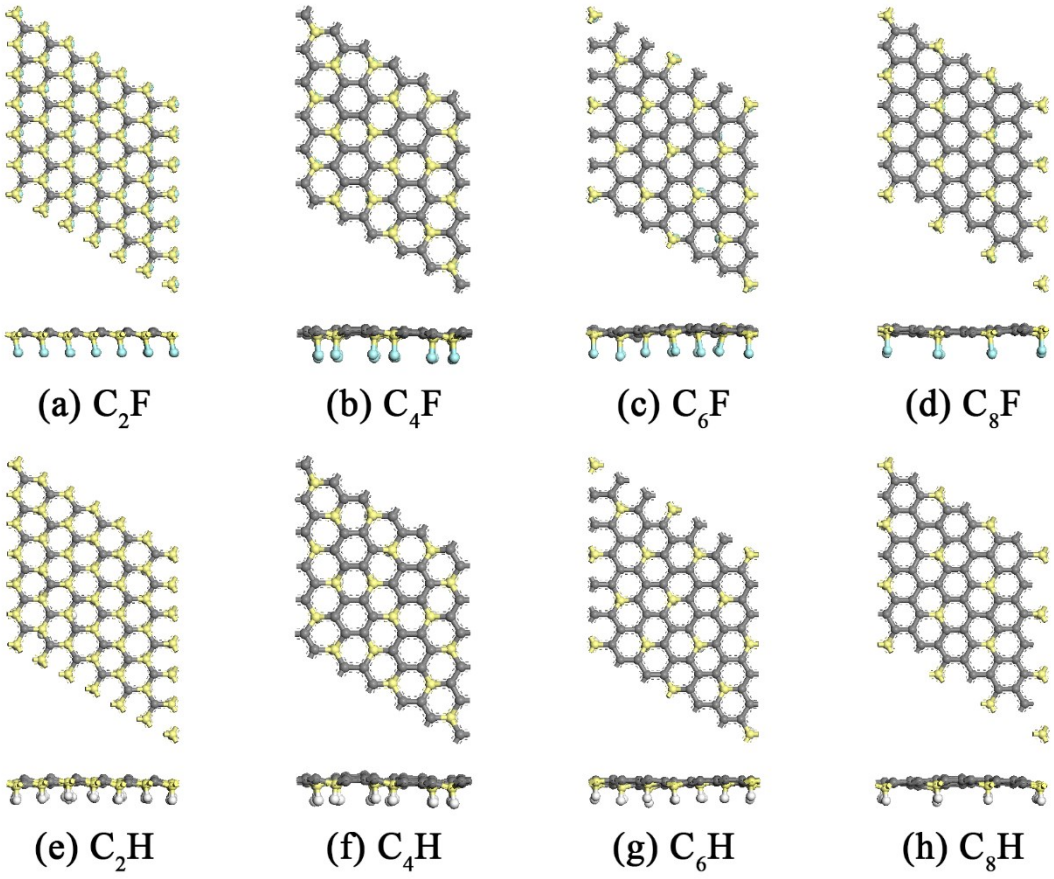


Fig. S2 Top views and side views of geometric structures of C_nX after 5 ps FPMD simulation at 300K. The grey, yellow, blue and white spheres denote sp^2-C, sp^3-C, F and H atoms, respectively.

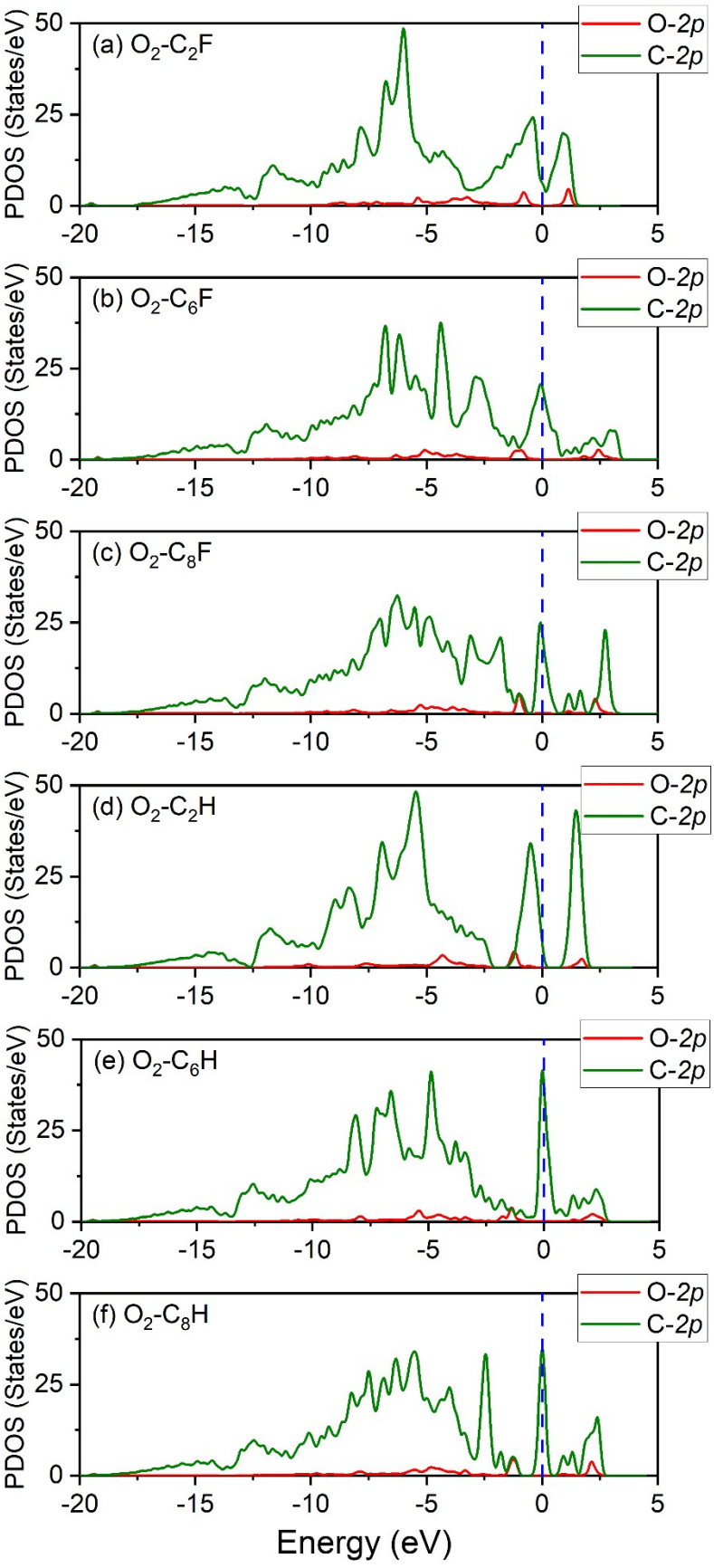


Fig. S3 Analysis on partial density of states (PDOS) of O_2 adsorbed SSX-Grs.

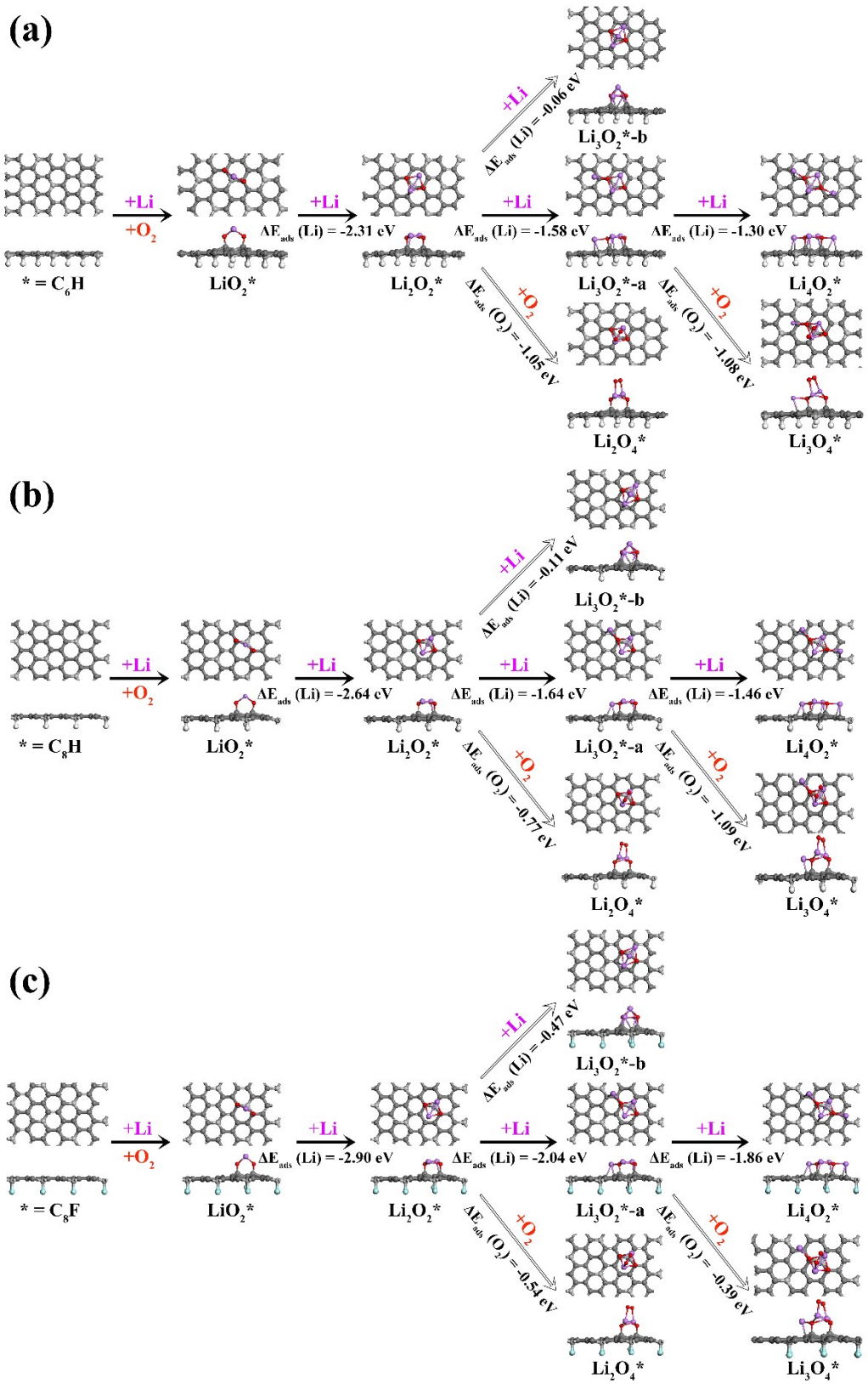


Fig. S4 Schematics of the growing pathways of Li_4O_2 on (a) C_6H, (b) C_8H and (c) C_8F. The grey, white, light blue, red and purple spheres represent C, H, F, O and Li atoms, respectively.

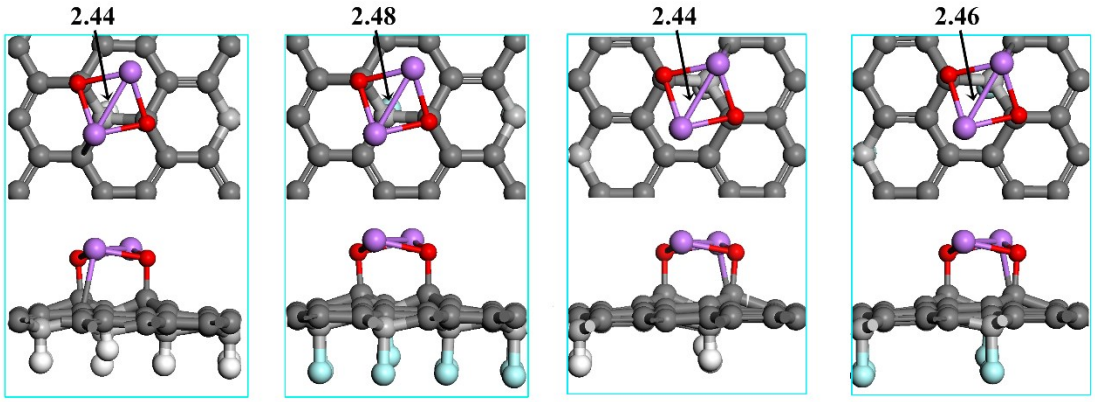


Fig. S5 The detailed structural analysis on Li_2O_2 adsorbed (a) C_6H, (b) C_6F, (c) C_8H and (d) C_8F. The distance between the two Li atoms (in Å) are denoted.

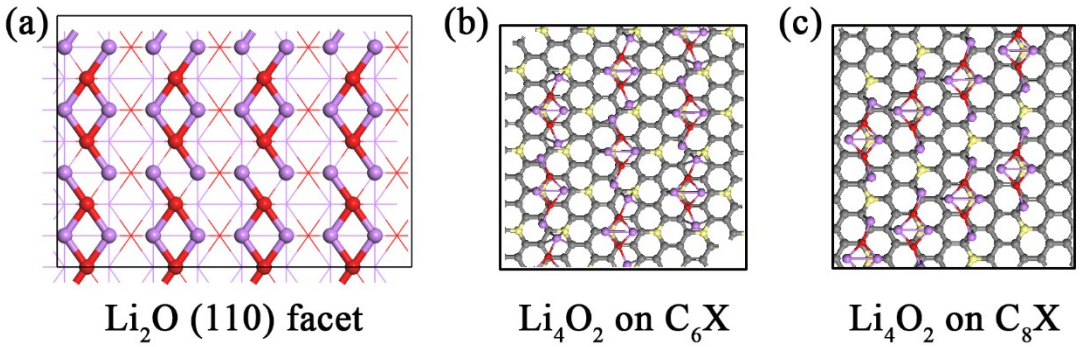


Fig. S6 (a) The (110) facet cutting from bulk Li_2O, the Li_4O_2 units in the surface are displayed as ball and stick model for clarity. (b) The simulated C_6X that is fully covered by Li_4O_2 units. (c) The simulated C_8X that is fully covered by Li_4O_2 units.

Table S1. The adsorption configurations and corresponding E_{ads} (eV), and charge transferred (Q_T, |e|) of O_2 on different SSX-Grs.

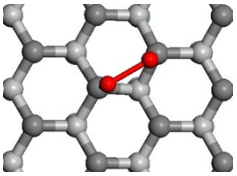
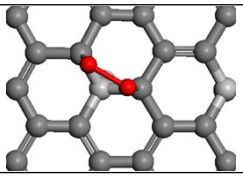
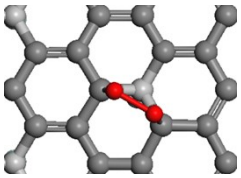
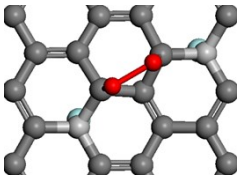
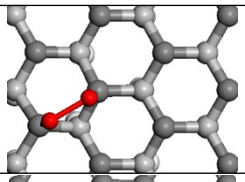
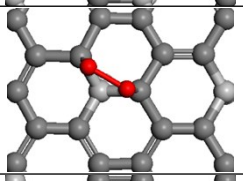
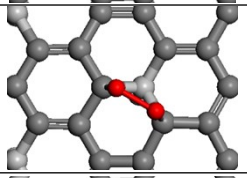
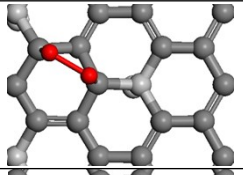
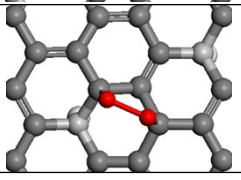
	Adsorption configuration	E_{ads}	Q_T
C_2F		-3.47	-0.55
C_6F		-1.02	-0.45
C_8F		-0.69	-0.46
		0.43	-0.47
C_2H		-3.52	-0.52
C_6H		-1.39	-0.47
C_8H		-1.21	-0.48
		-0.34	-0.51
		-0.15	-0.49

Table S2. The adsorption energy (E_{ads}) of the LiO_2 and O_2Li on different SSX-Gr.

Substrate	Adsorbate	E_{ads} (eV)
C_6F	LiO_2	-3.48
	O_2Li	-1.78
C_8F	LiO_2	-3.09
	O_2Li	-1.66
C_6H	LiO_2	-4.63
	O_2Li	-2.00
C_8H	LiO_2	-3.98
	O_2Li	-1.88

Reference

1. A. Togo, F. Oba and I. Tanaka, *Phys. Rev. B*, 2008, **78**, 134106.