Supplementary Information

Grafting Functional Groups in Polymeric Binder Towards Enhancing Structural Integrity of Li_xSiO_2 Anode During Electrochemical Cycling

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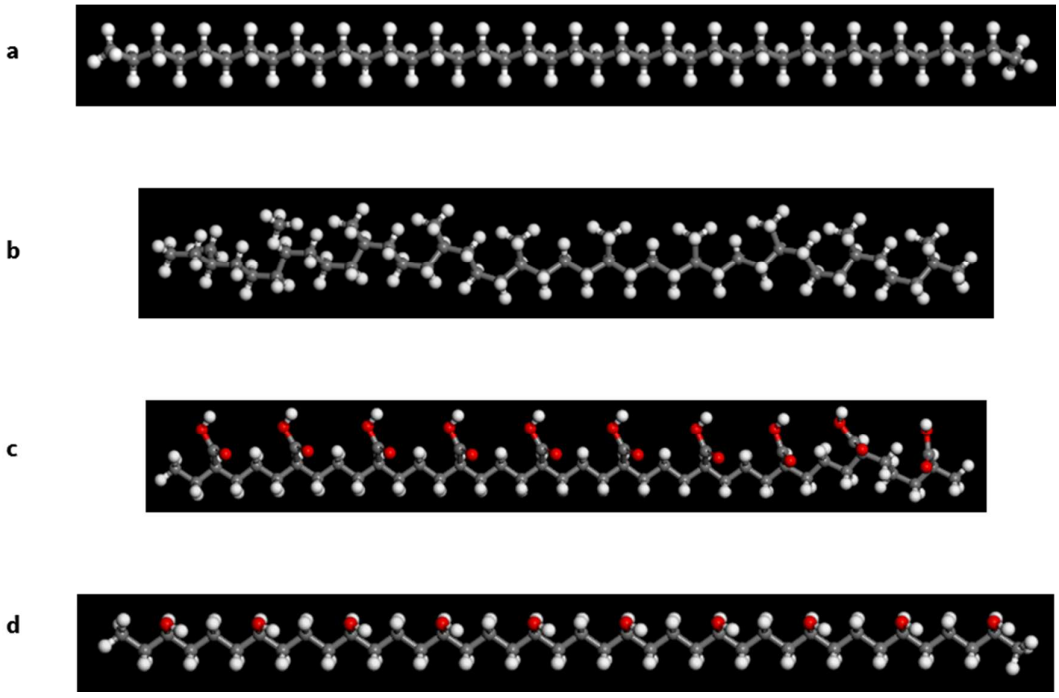


Figure S1. (a) The single chain of polyethylene (PE-H) with 20 repeating units and the grafted functional group of (b) CH_3 (PE-CH_3), (c) COOH (PE-COOH), and (d) OH (PE-OH). Each color of the ball represents H (white), C (gray), and O (red), respectively.

	Volume (Å^3)				Porosity
	Connolly	Total	Occupied	Free	
PE-CH_3	27.23	77316.90	57419.27	19897.63	25.74
PE-H	6.85	61954.84	46649.88	15304.96	24.70
PE-OH	19.30	61219.49	50950.15	10269.34	16.77
PE-COOH	37.52	75001.46	62532.21	12469.25	16.63

Table S1. Connolly volume of the functional group, total volume of the simulation box with FMP, occupied volume of FMP, free volume, and porosity for the bulk FMPs.

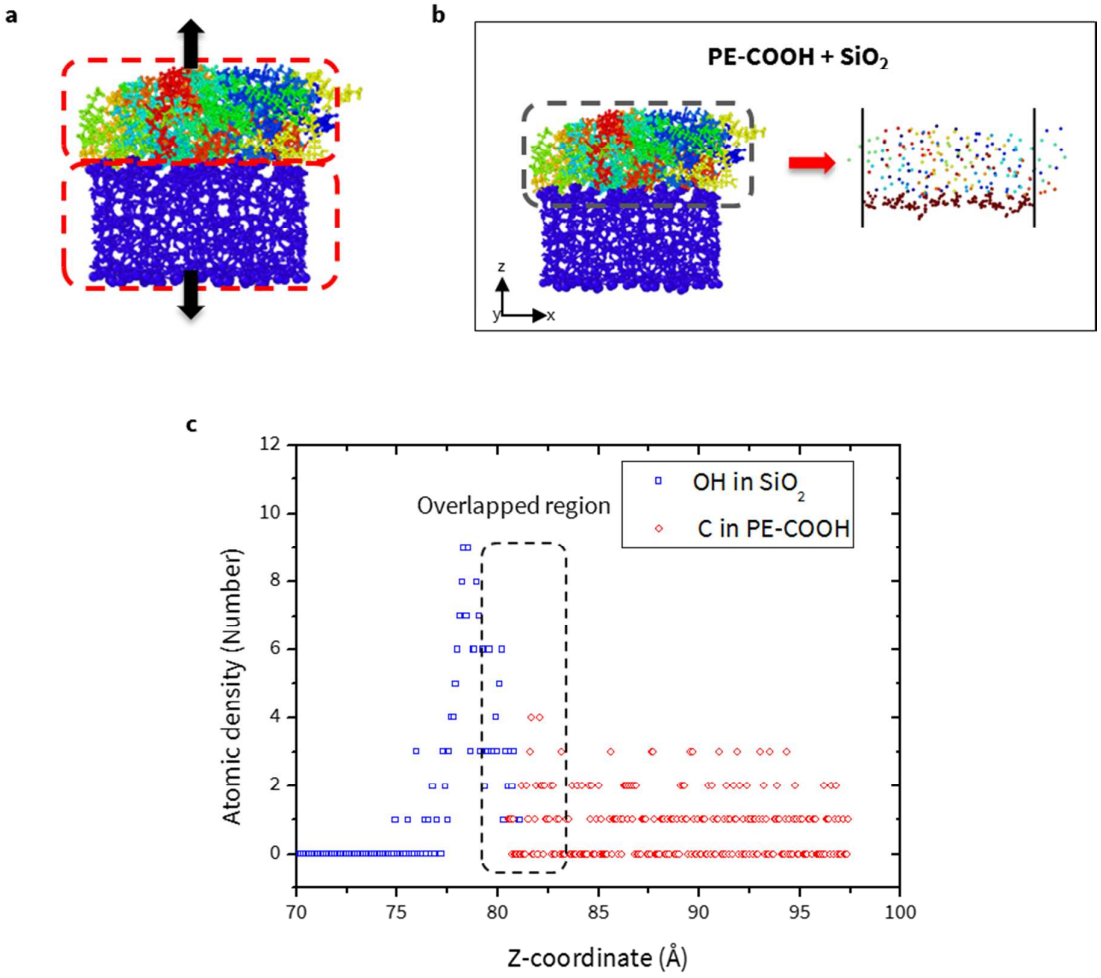
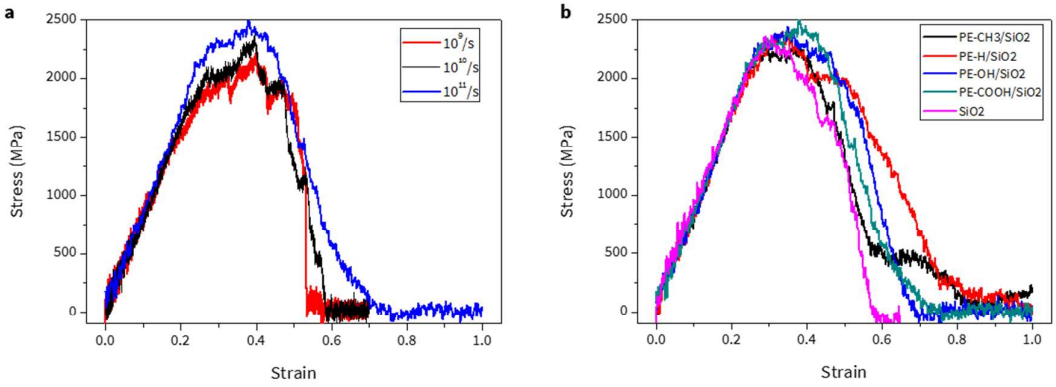


Figure S2. (a) The schematic view of how pulling test is performed. The dotted red box is where the pulling velocity is applied and the black arrow is the direction of pulling. (b) (Left) The hybrid structure of PE-COOH + SiO_2 and (Right) the distribution of atomic species of H and O in the hydroxyl group on the top of SiO_2 and C from PE and (c) their resulting atomic density in the *z*-direction.

	PE-CH_3	PE-H	PE-OH	PE-COOH
# of atoms	28	23	62	36
Volume ($Å^3$)	1863.02	2041.65	2001.02	1193.53
Density (# of atoms/volume)	0.015029	0.011265	0.030984	0.030163

Table S2. The number of H and O in the hydroxyl group on the top of SiO_2 and C from PE, overlapped volume in **Figure S2**. and their density for the slab structure of FMPs + SiO_2. For computing the volume of the occupied region, the length in z-direction (out-of-plane) is computed between the maximum and minimum value of the z-coordinate from the OH in SiO_2 and C in FMPs is obtained, respectively, while the in-plane length (x- and y-length) is given from the simulation box.



c

	Maximum strength (GPa)			Fracture strain		
	10^9/s	10^{10}/s	10^{11}/s	10^9/s	10^{10}/s	10^{11}/s
SiO_2	2217.55	2399.35	2433.54	0.472	0.572	0.587
PE-CH_3 + SiO_2	2110.84	2253.15	2359.62	0.755	0.789	0.849
PE-H + SiO_2	2049.39	2198.25	2410.31	0.696	0.765	0.832
PE-OH + SiO_2	2191.00	2352.52	2441.74	0.550	0.612	0.694
PE-COOH + SiO_2	2178.10	2390.54	2497.99	0.567	0.589	0.749

Figure S3. Stress-strain curve for the slab structure of (a) PE-COOH + SiO_2 under the strain rate of 10^9/s to 10^{11} /s and (b) FMP + SiO_2 under the strain rate of 10^{11} /s. (c) Maximum strength and the fracture strain for the slab structure of SiO_2 and FMPs + SiO_2 under the strain rate of 10^9 /s to 10^{11} /s

	Li_xSiO_2			PE-H + Li_xSiO_2			PE-COOH + Li_xSiO_2		
	Density (g/cm^3)	Volume (A^3)	Vol. Strain	Density (g/cm^3)	Volume (A^3)	Vol. Strain	Density (g/cm^3)	Volume (A^3)	Vol. Strain
Li=0	2.360	25061.35	n/a	1.735	63521.06	n/a	1.789	69784.43	n/a
Li=1	2.222	29796.21	0.188	1.794	67143.49	0.057	1.845	73238.64	0.049
Li=2	2.155	33999.83	0.357	1.730	75565.55	0.190	1.789	81248.40	0.164
Li=3	2.042	39323.92	0.569	1.674	84254.18	0.326	1.739	89522.85	0.283
Li=4	1.937	45094.81	0.799	1.653	91538.69	0.441	1.699	97672.15	0.400

Table S3. Density, volume, and volumetric strain change during lithiation for the confined structure of Li_xSiO_2, PE-H + Li_xSiO_2, and PE-COOH + Li_xSiO_2 when x=0 to 4.

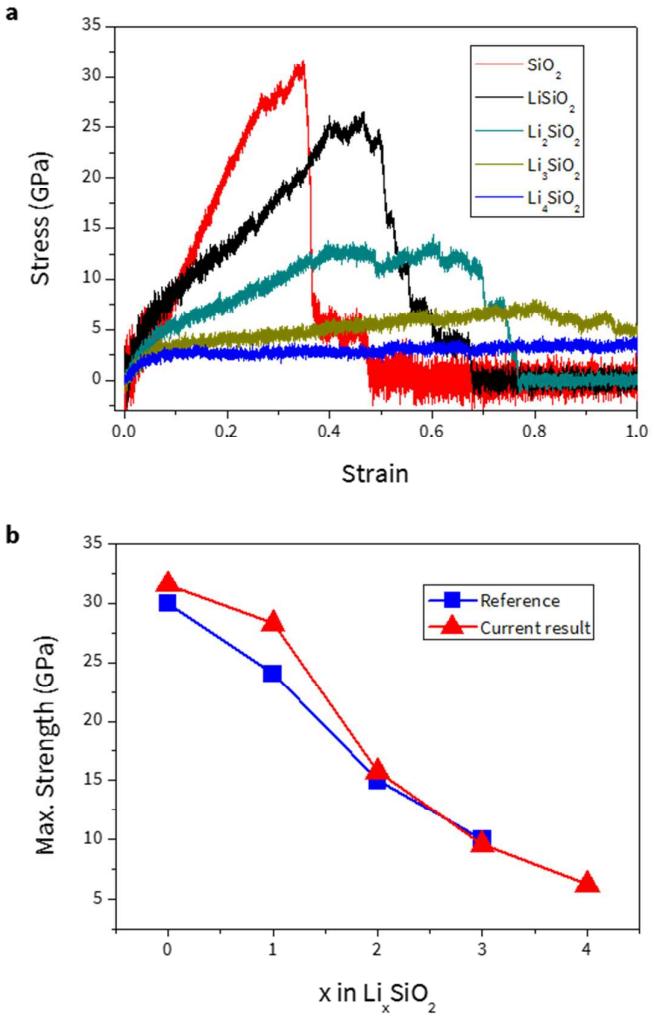


Figure S4. (a) Stress-strain curve of Li_xSiO_2 (x=0 to 4) under the strain rate of 10^{10}/s and (b) corresponding maximum strength with reference values from previous reference (J. Phys. Chem. A 2016, 120, 2114−2127).

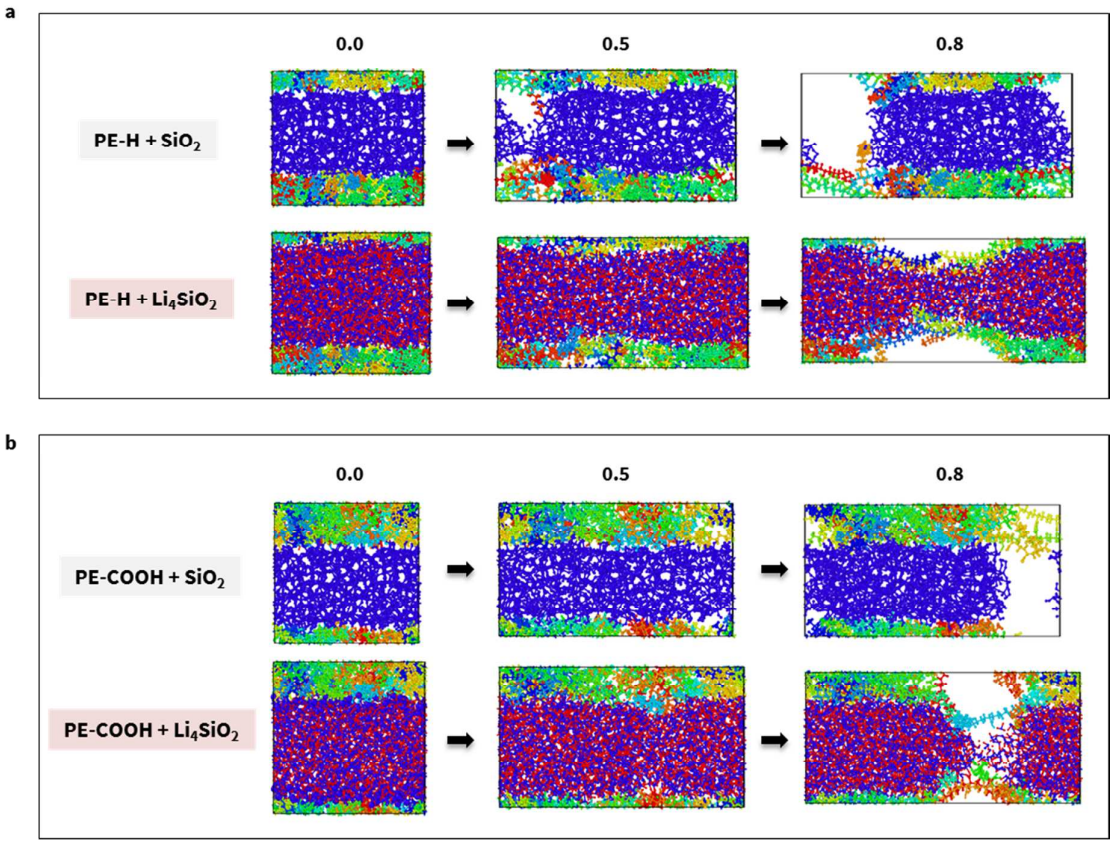


Figure S5. The atomic snapshots during tensile test for the confined structure of (a) PE-H + SiO_2 and PE-H + Li_4SiO_2 and (b) PE-COOH + SiO_2 and PE-COOH + Li_4SiO_2 at the strain rate of 10^{10}/s. Each number corresponds to the applied strain.

Interfacial surface area (Å^2)	PE-H + Li_xSiO_2	PE-COOH + Li_xSiO_2
Li=0	1737.014	1734.086
Li=1	1751.883	1736.808
Li=2	1911.877	1893.519
Li=3	2087.618	2025.066
Li=4	2198.352	2129.519

Table S4. Interfacial surface area for the confined structure PE-H + Li_xSiO_2 and PE-COOH + Li_xSiO_2 when x=0 to 4. The surface area is approximated by obtaining it from the in-plane length (x- and y-dimension).