

PVDF-Supported Graphene Foam as Robust Current Collector for Lithium Metal Anode

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Figure S1

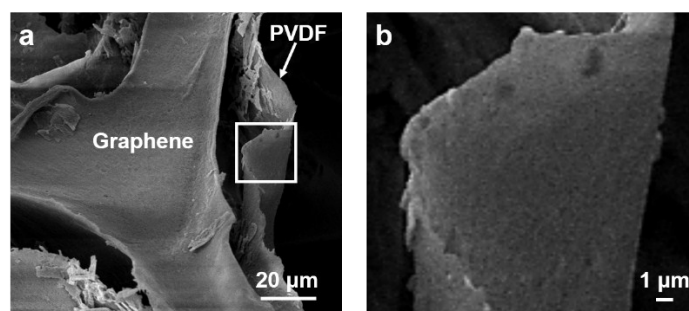


Figure S1 a. SEM image of the PSGF showing an obvious dividing part between the graphene foam and the PVDF layer. b. The magnified image of the marked area in a.

The thickness of the PVDF layer coated over the graphene was determined by the filtration speed and the amount of PVDF solution used in the vacuum-filtration coating process. Experimentally, 1 mL 5% solution of PVDF in NMP was filtrated through the Ni@G foam in 20 seconds. As a result, the thickness of PVDF layer coated over graphene was less than 100 nm.

Figure S2

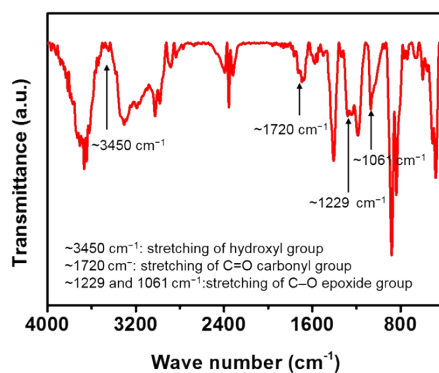


Figure S2 FTIR spectrum of PSGF.

As shown in Figure S2, the lithiophilic oxygen function groups were supposed to be –COO- groups.

Table S1

Peak	Type	Position BE (eV)	FWHM (eV)	Raw Area (cps eV)	RSF	Atomic Mass	Atomic Conc %	Mass Conc %
F 1s	Reg	684.300	1.951	34317.7	1.000	18.998	23.88	32.98
O 1s	Reg	528.250	2.162	1950.7	0.780	15.999	1.83	2.12
C 1s	Reg	282.900	1.448	24096.7	0.278	12.011	74.30	64.89

Table S1 Quantification report of XPS conducted on the surface of PSGF

XPS conducted on PSGF (Table S1) identified the existence of O atoms on the carbonous surface, which help create the lithiophilic surface for lithium deposition. Because of the limit of XPS in quantitative characterization, the atomic concertation and mass concertation are not precise and just for reference.

Figure S3

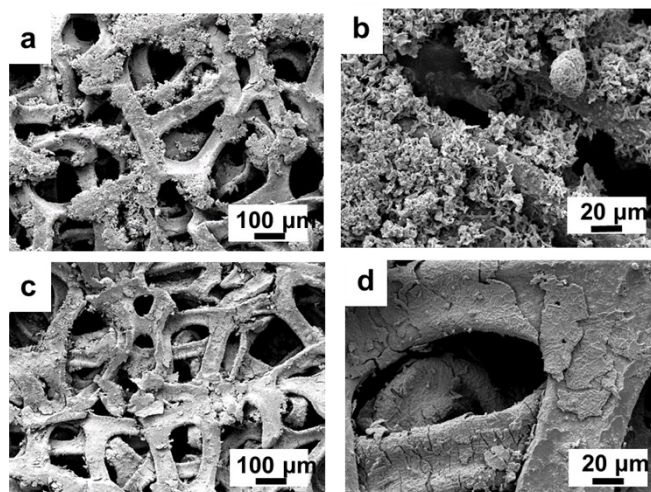


Figure S3 **a, b.** SEM images of the morphologies of lithium deposited on nickel foam (a, b) and PSGF (c, d) under different magnifications.

To demonstrate the critical function of the surface chemistry on tailoring the lithium deposition behavior, the nickel foam was taken as the reference current collector for lithium metal anode, which

has the three-dimensional morphology same with the PSGF. As shown in Figure S3a and b, the lithium deposition on nickel foam shows a dendritic and uneven morphology. However, the lithium deposition on the PSGF exhibits a much more even and flat morphology. It can be ascribed to the effectiveness of the surface chemistry on tailoring the lithium deposition behavior. The lithiophilic O atoms on the surface of the PSGF were supposed to decrease the nucleation barrier of lithium deposition and thus enabling an even plating behavior of lithium metal.

Figure S4

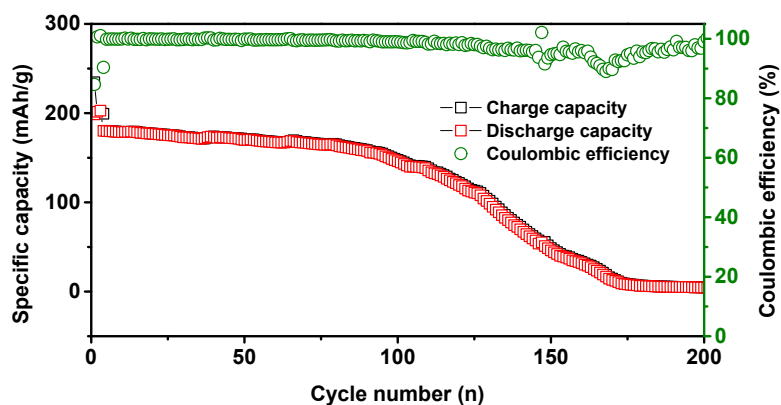


Figure S4 Cycling performance of the full cell comprising of Cu@Li anode and NCA cathode under the current density of 1C (1 C = 220 mA/g).

Figure S4 shows the cycling performance of the full cell comprising of Cu@Li anode and NCA cathode at 1C. The capacity was stable in the early 100 cycles, but an obvious drop was observed after 100 cycles. The failure of the cycling performance was mainly caused by the exhaustion of active lithium ions, as well as the low coulombic efficiency of the Cu@Li anode.

Figure S5

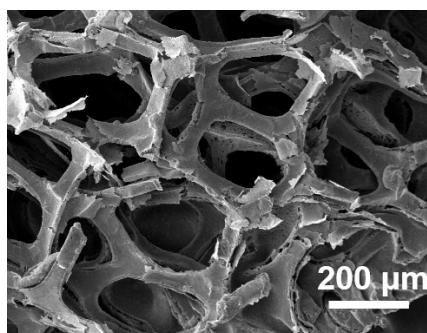


Figure S5 SEM image of the PSGF after 200 cycles in the full cell.

We disassembled the full cell comprising of PSGF@Li anode and NCA cathode after 200 cycles

and removed the surface SEI film and lithium metal by the organic solvent (DMC : Actone : Ethonal=1:1:1). The cycled PSGF current collector exhibits a well maintained three-dimensional morphology (Figure S5), which is benefits from the effective supporting function of the PVDF. The robust feature of the PSGF current collector is critical for the long lifespan of the corresponding lithium metal anode.