

# **Graphene-Directed Two-Dimensional Porous Carbon Frameworks for High-Performance Lithium-Sulfur Battery Cathodes**

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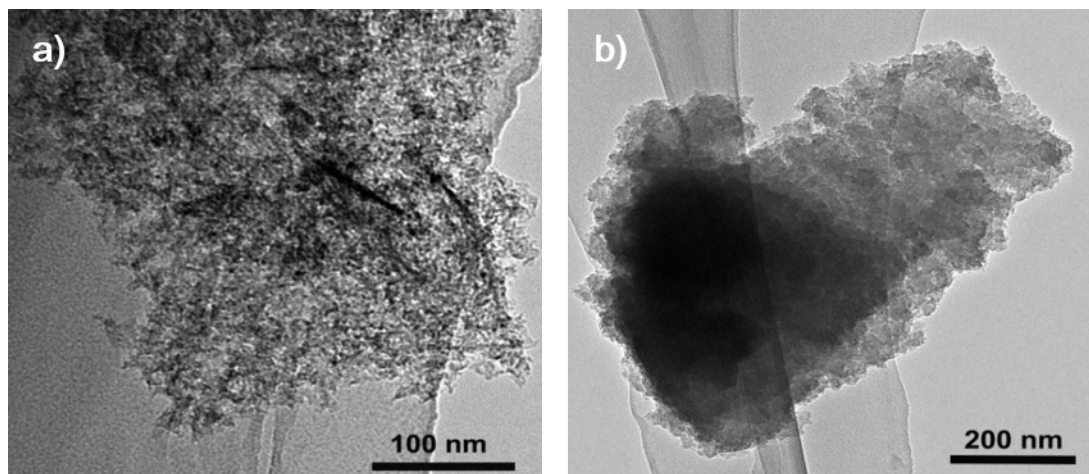
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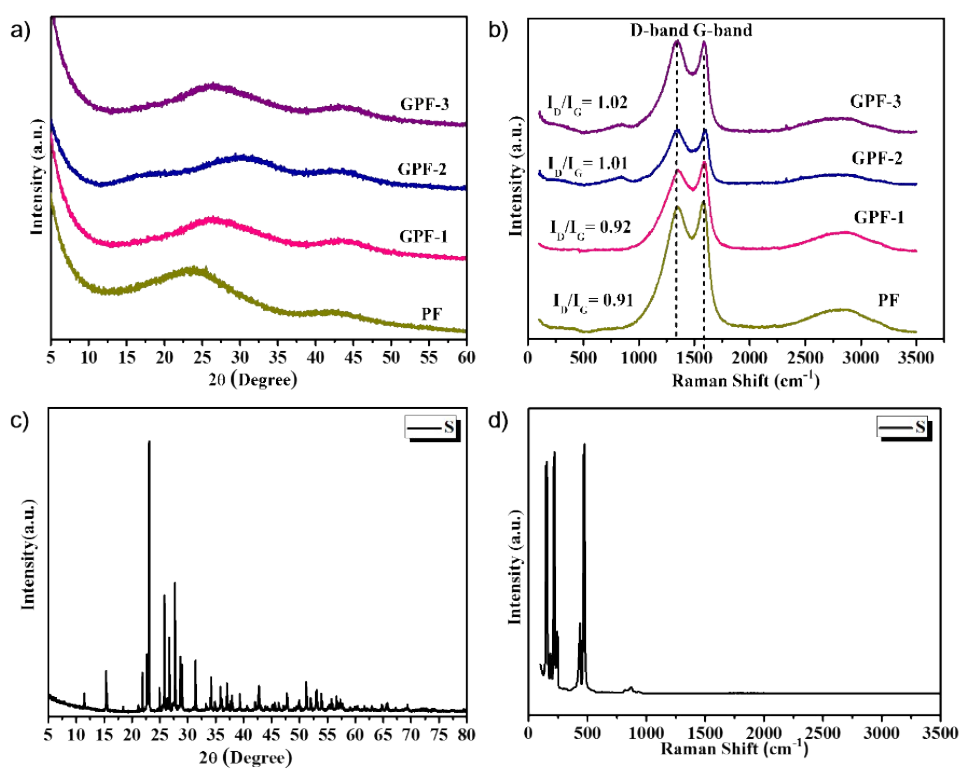
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**Fig. S1** TEM images of a) GPF-3 and b) PF.

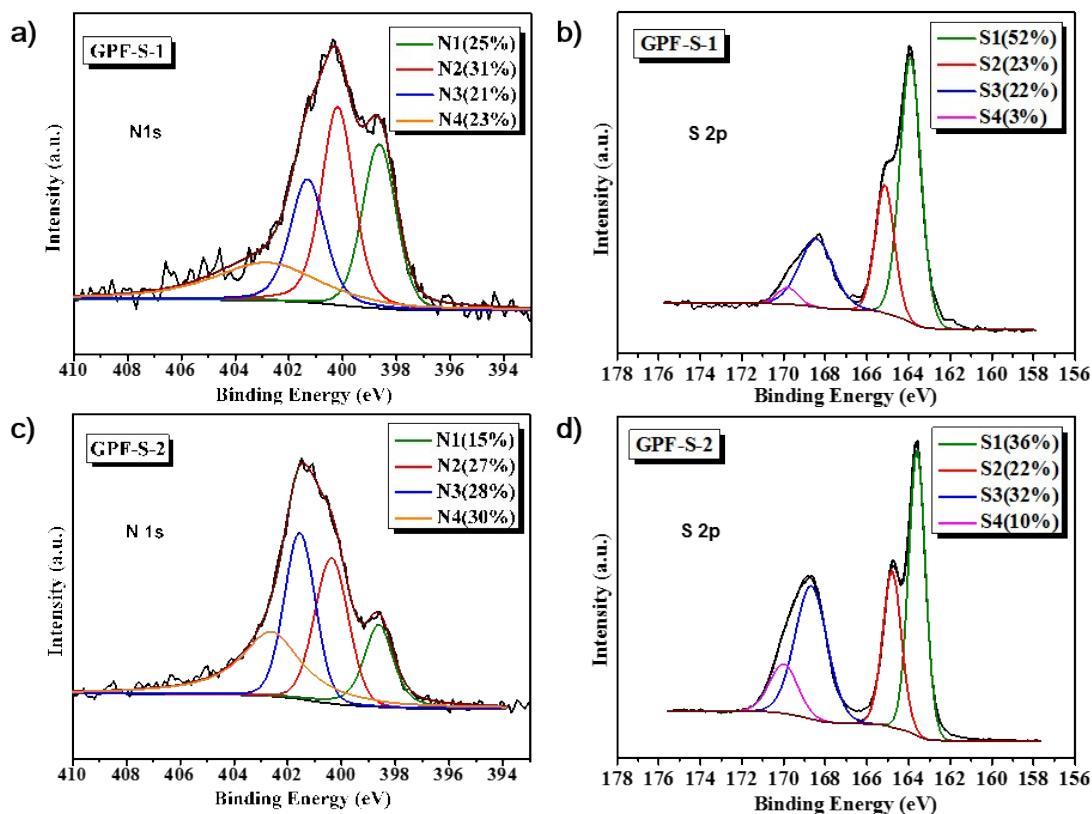


**Fig. S2** XRD patterns of a) GPF-*n*, PF (*n*=1, 2, 3) and c) sublimed sulfur; Raman spectrum of b) GPF-*n*, PF (*n*=1, 2, 3) and d) sublimed sulfur.

**Table S1.** Porous Characteristics of GPF-*n* and PF.

| Samples | $S_{\text{BET}}^a$<br>( $\text{m}^2 \text{g}^{-1}$ ) | Total pore<br>volume $^b$<br>( $\text{cm}^3 \text{g}^{-1}$ ) | Average<br>pore size $^c$<br>(nm) | Mesopore<br>ratio<br>(%) | $S_{\text{BET}}^a$ after<br>sulfur loading<br>( $\text{m}^2 \text{g}^{-1}$ ) | Total pore volume $^b$<br>after sulfur loading<br>( $\text{cm}^3 \text{g}^{-1}$ ) |
|---------|------------------------------------------------------|--------------------------------------------------------------|-----------------------------------|--------------------------|------------------------------------------------------------------------------|-----------------------------------------------------------------------------------|
| PF      | 1157                                                 | 0.66                                                         | 2.3                               | 34                       | 20                                                                           | 0.06                                                                              |
| GPF-1   | 1285                                                 | 0.93                                                         | 2.9                               | 60                       | 15                                                                           | 0.07                                                                              |
| GPF-2   | 1414                                                 | 2.59                                                         | 7.3                               | 97                       | 20                                                                           | 0.16                                                                              |
| GPF-3   | 1683                                                 | 1.57                                                         | 3.7                               | 82                       | 14                                                                           | 0.07                                                                              |

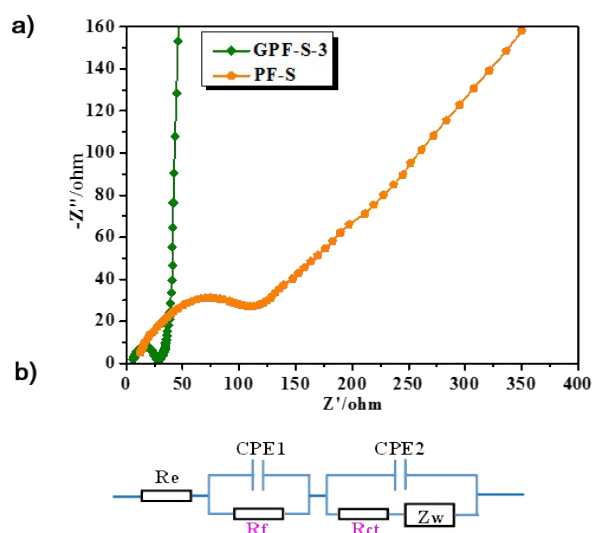
$^a$  BET specific surface area.  $^b$  At  $P/P_0 = 0.99$ .  $^c$  Determined from density functional theory (DFT).



**Fig. S3** N 1s XPS spectra of a) GPF-S-1 and c) GPF-S-2; S 2p XPS spectra of b) GPF-S-1 and d) GPF-S-2.

**Table S2.** Quantitative elemental analysis of GPF-*n*.

| Samples | Elements (wt %) |     |     |
|---------|-----------------|-----|-----|
|         | C               | N   | H   |
| GPF-1   | 86.6            | 8.7 | 4.7 |
| GPF-2   | 89.5            | 7.2 | 3.3 |
| GPF-3   | 90.0            | 6.3 | 3.7 |



**Fig. S4** a) Nyquist plots of PF-S and GPF-S-3 electrodes obtained by applying a sine wave with amplitude of 5.0 mV over the frequency range 100 kHz to 0.1 Hz; b) equivalent circuit model of the studied system ( $R_e$ : the electrolyte resistance;  $R_f$ : the resistance of the surface film formed on the electrodes, namely contact resistance;  $R_{ct}$ : charge-transfer resistance).

**Table S3.** Kinetic parameters of GPF-S-3 and PF-S electrodes.

| Samples | $R_f (\Omega)$ | $R_{ct} (\Omega)$ |
|---------|----------------|-------------------|
| PF-S    | 56.89          | 146.50            |
| GPF-S-3 | 18.52          | 24.33             |

**Table S4.** Comparison of the electrochemical performance of GPF-3 and other literature reported porous carbon-based cathode materials for Li-S battery.

| Samples              | $S_{\text{BET}}/\text{m}^2 \text{ g}^{-1}$<br>( $V_{\text{tot}}/\text{cm}^3 \text{ g}^{-1}$ ) | Specific capacity/ $\text{mAh g}^{-1}$ |                                                      |                                                                                        | Ref.      |
|----------------------|-----------------------------------------------------------------------------------------------|----------------------------------------|------------------------------------------------------|----------------------------------------------------------------------------------------|-----------|
|                      |                                                                                               | Initial capacity (current density)     | Stabilized capacity (current density; cycle numbers) | Capacity at large current density (current density)                                    |           |
| Hollow carbon sphere | 648                                                                                           | 1070(0.5 C)                            | 970(0.5 C;100)                                       | 450(3 C)                                                                               | 1         |
| a-PCNS               | 1800(5.4)                                                                                     | 1300(1 C)                              | 720(1 C;100)                                         |                                                                                        | 2         |
| L-GPC                | 2500(1.937)                                                                                   | 900(0.5 C)                             | 650(0.5 C;100)                                       | 600(5 C)                                                                               | 3         |
| GSH@APC              | 513(0.853)                                                                                    | 1070(1 C)                              | 877(1 C;150)                                         | 800(10 C)                                                                              | 4         |
| CA                   | 642(1.02)                                                                                     | 1441(0.1 A $\text{g}^{-1}$ )           | 820(0.1 A $\text{g}^{-1}$ ;50)                       | 521(3.2 C)                                                                             | 5         |
| MCNs-1               | 857(0.45)                                                                                     |                                        |                                                      | 450(1.8 C)                                                                             | 6         |
| DTG                  | 1628(2.0)                                                                                     | 110(1 C)                               | 750(1 C;200)                                         | 720(10 C)                                                                              | 7         |
| OMC                  | 2445(2.63)                                                                                    | 1050(1 C)                              | 965(1 C;100)                                         |                                                                                        | 8         |
| PAF                  | 2023(1.73)                                                                                    | 850(0.05 C)                            | 700(0.05 C;50)                                       |                                                                                        | 9         |
| TTCN                 | 822.8(1.77)                                                                                   | 1270(0.5 A $\text{g}^{-1}$ )           | 915(0.5 A $\text{g}^{-1}$ ;50)                       | 565(6 A $\text{g}^{-1}$ )                                                              | 10        |
| N-ACNT/G             | 217(0.72)                                                                                     | 1150(1 C)                              | 900(1 C;80)                                          | 780(5 C)                                                                               | 11        |
| PDA-NHC-S            | 1538(2.02)                                                                                    | 1180(0.2 C)                            | 720(0.2 C;200)                                       |                                                                                        | 12        |
| MMCS                 | 1013(1.26)                                                                                    | 1640(0.1 C)                            | 1014(0.1 C;100)                                      | 600(2 C)                                                                               | 13        |
| DUT-86               | 2450(4.97)                                                                                    | 1190(0.1 C)                            | 880(0.1 C;100)                                       | 200(1 C)                                                                               | 14        |
| MNCS/CNT             | 615(2.07)                                                                                     | 1438(0.84 mA $\text{cm}^{-2}$ )        | 1200(1.68 mA $\text{cm}^{-2}$ ;200)                  |                                                                                        | 15        |
| 850PCNTs             | 367.2(1.68)                                                                                   | 1080(1 C)                              | 600(1 C;250)                                         | 150(15 C)                                                                              | 16        |
| 3D rGO-HCS           | 445.07(2.28)                                                                                  | 901(0.5 C)                             | 772(0.5 C;400)                                       | 770(4 C)                                                                               | 17        |
| GPF-3                | 1683 (1.57)                                                                                   | 1461(2 A $\text{g}^{-1}$ )             | 962(2 A $\text{g}^{-1}$ ;120)                        | 890(5 A $\text{g}^{-1}$ );<br>652(15 A $\text{g}^{-1}$ )<br>591(20 A $\text{g}^{-1}$ ) | This work |

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