

## Supporting Information

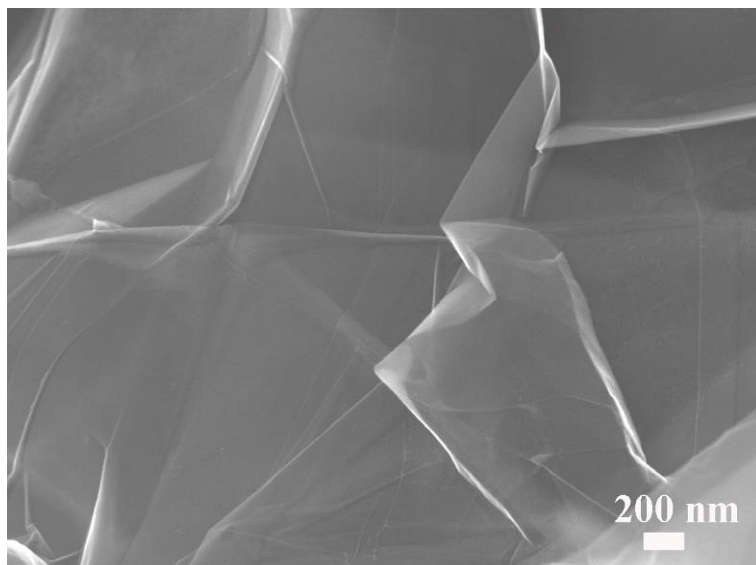
# Caterpillar-like Graphene Confining Sulfur by Restacking Effect for High Performance Lithium Sulfur Batteries

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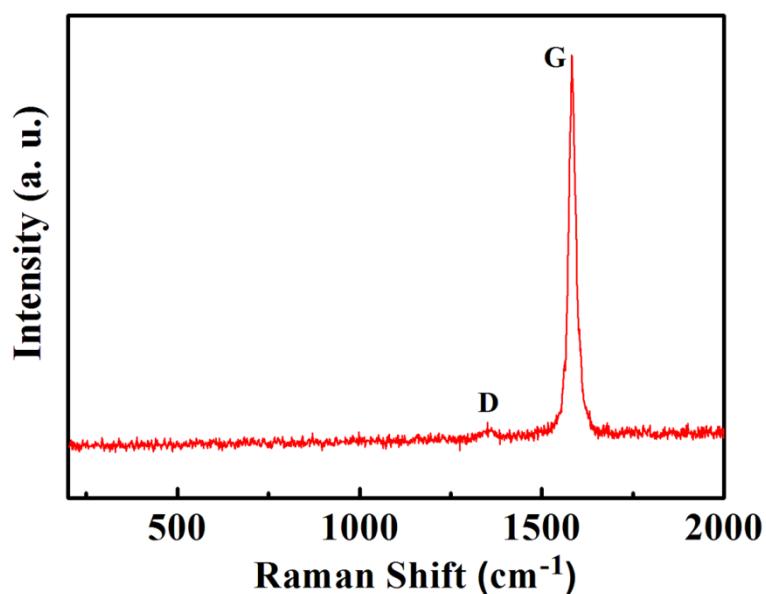
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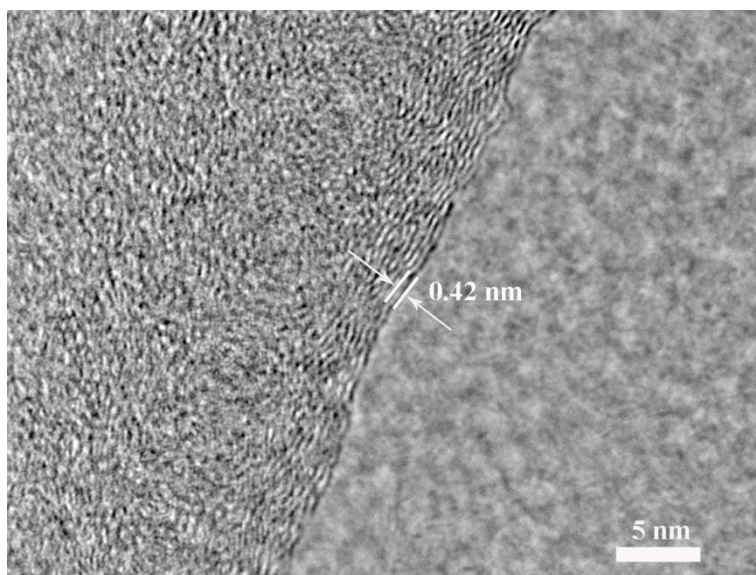
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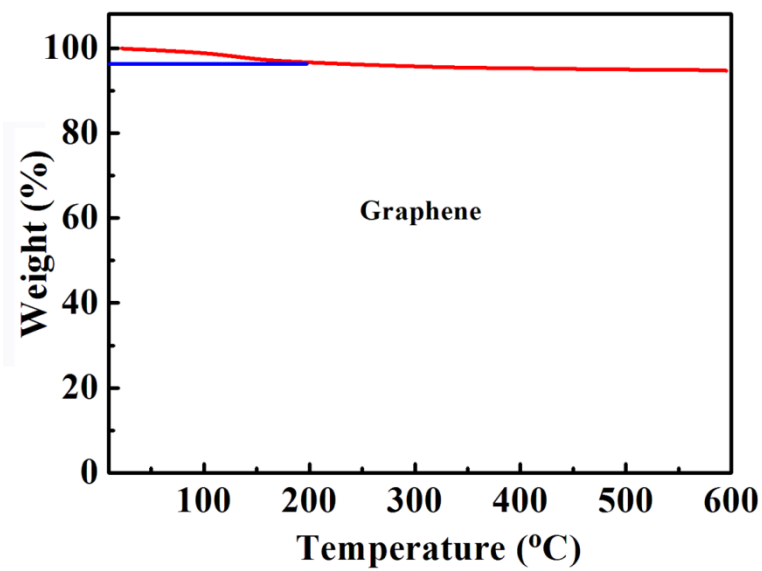
**Figure S1.** High-resolution scanning electron microscopy (HRSEM) image of graphene. The HRSEM image shows that the caterpillar-like graphene has a wrinkled structure.



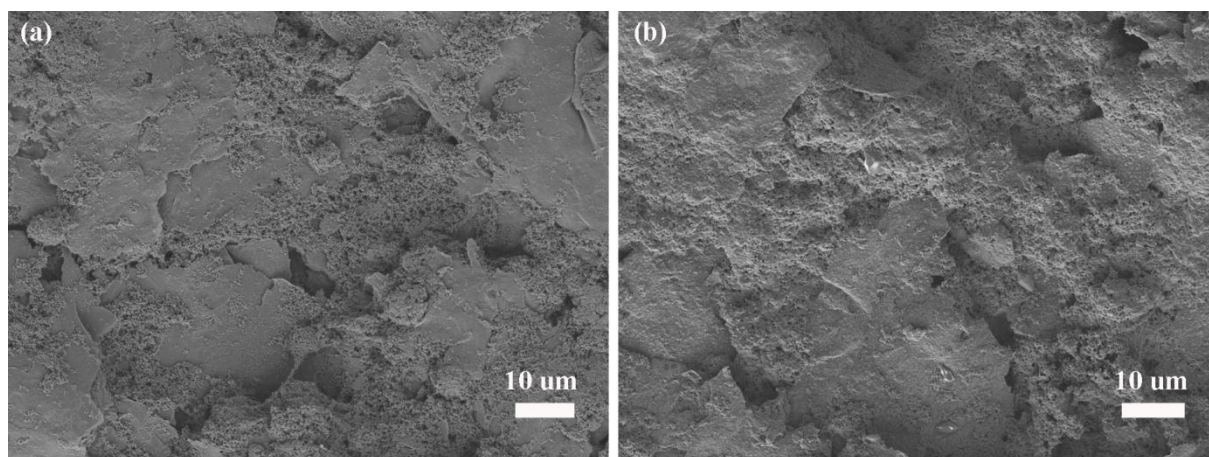
**Figure S2.** Raman spectra of the graphene. In the Raman spectra of the graphene, two bands at  $\sim 1355$  and  $\sim 1582$   $\text{cm}^{-1}$  correspond to the D-band (D) and the G-band (G), respectively. The D-band corresponds to the disorder induced the carbon and structural defects. The G-band corresponding to the  $\text{sp}^2$  carbon-bonded graphitic structure can enhance the electrical conductivity of carbon materials.



**Figure S3.** High-resolution TEM images of graphene-sulfur. After encapsulating sulfur, the layer distance of graphene-sulfur is 0.42 nm, indicating that graphene expands after sulfur intercalation.

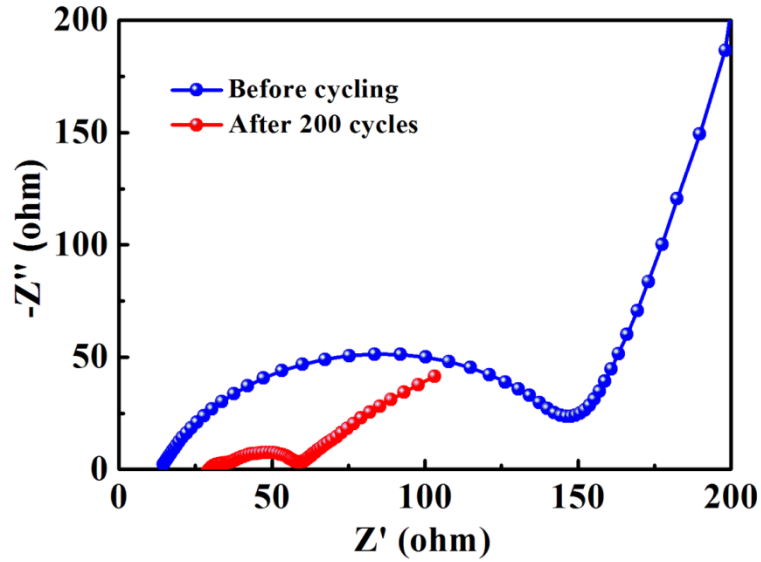


**Figure S4.** Thermal gravimetric (TG) curve of the graphene. The mass loss before 200 °C may result from the evaporation of water.



**Figure S5.** SEM images of the graphene-sulfur electrode (a) before cycling and (b) after 200 cycles at a high current density of  $1675 \text{ mA g}^{-1}$ . The small particles are Super C65. After cycling

at a high current density, the laminar structure of the graphene-sulfur is still remained, indicating the graphene-sulfur electrode has excellent mechanical stability.



**Figure S6.** Impedance plots of the graphene-sulfur electrode before cycling and after 200 cycles at a high current density of  $1675 \text{ mA g}^{-1}$ . The Nyquist plots consist of a depressed semicircle at high frequency region and an oblique line at low frequency region. The diameter of the depressed semicircle represents the charge transfer resistance ( $R_{ct}$ ). The charge transfer resistance of the cycled graphene-sulfur electrode is smaller than that of the fresh graphene-sulfur electrode, indicating that the relocation of sulfur species to the caterpillar-like graphene decreases the tendency of passivation layer formation on the electrode.