Supporting Information

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

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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 ~1355 and ~1582 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 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 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 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.