Supporting information

Excimer Ultraviolet-Irradiated Exfoliated Graphite Loaded with Carbon-Coated SnOₓ Small Nanoparticles as Advanced Anodes for High-Rate-Capacity Lithium-Ion Batteries

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Fig. S1 Time vs. temperature of the process of SnO$_2$ SNPs deposition onto graphene/exfoliated graphite in previous reports and this work.

Fig. S2 Raman spectra of EUV-HCl-G, EUV-G, and exfoliated graphite.
Fig. S3 XRD patterns of SnO$_2$@C-G and SnO$_2$ powders.

Fig. S4 SEM images of exfoliated graphite.

Fig. S5 (a) N$_2$ adsorption-desorption isotherms and (b) BJH pore distribution of SnO$_2$@C-G and exfoliated graphite.
Fig. S6 TG curves of SnO$_2$@C-G and SnO$_2$-G in air recorded at a heating rate of 10 °C min$^{-1}$. 
Fig. S7 (a, b) TEM images of exfoliated graphite. TEM images of SnO$_x$@C-G under (c) 10, (d) 20 and (e) 30 min EUV radiation. (f) TEM image of SnO$_x$-G under 20 min EUV radiation.
**Fig. S8** HR-TEM image of SnO$_x$@C-G.

**Fig. S8** shows discontinuous lattice fringes around the SnO$_x$ nanoparticles. Although it is difficult to distinguish the carbon coating layer and the exfoliated graphite support at this resolution, the lattice fringes of the exfoliated graphite sheets can hardly be seen in the non-edge regions without folding (**Fig. S7b**). So it can be believed that these discontinuous lattice fringes are attributed to the carbon coating.

**Fig. S9** Raman spectra of SnO$_x$@C-G and SnO$_x$-G.
**Fig. S10** Cyclic voltammogram (CV) curves of (a) SnO$_x$@C-G and (b) SnO$_x$-G electrode from 0.01 to 3 V vs. Li$^+$/Li at a scan rate of 0.1 mV s$^{-1}$.

**Fig. S11** Coulombic efficiency of (a) SnO$_x$@C-G, (b) SnO$_x$-G, and (c) exfoliated graphite electrode.
Fig. S12 (a) Galvanostatic charge/discharge profiles and (b) cycling performance of exfoliated graphite between 0.01 and 3.00 V at a current density of 0.1 A g\(^{-1}\).
Fig. S13 (a) Cycling performances of SnO$_2$@C-G-92%, SnO$_2$@C-G$'$ and SnO$_2$-G between 0.01 and 3.00 V at a current density of 0.1 A g$^{-1}$. Galvanostatic charge/discharge profiles and cycling performances of SnO$_2$@C-G$'$ (b, c) and SnO$_2$-G (d, e) at a current density of 0.1 A g$^{-1}$ for the initial 10 cycles and 1.6 A g$^{-1}$ for subsequent cycles.

The procedure to synthesize SnO$_2$@C-G-92% was the same as that of SnO$_2$@C-G. The SnO$_2$@C-G-92% anode was prepared with approx. 92% of SnO$_2$@C-G and higher loading (approx. 6 mg cm$^{-2}$). The procedure to synthesize SnO$_2$@C-G$'$ was the same as that of SnO$_2$@C-G, but reduced the exfoliated graphite quality to 20 mg, and the EUV radiation was extended to 40 min. The ratio of SnO$_2$-exfoliated graphite is 4:1 calculated based on the EDS results in Fig. S13c. The SnO$_2$-G with a mass ratio of
SnO$_2$: exfoliated graphite =1:1 was prepared by high energy ball milling with a rotating speed of 450 rpm for 4 h. Both of the two samples show the poor cycling stabilities at high current density of 1.6 A g$^{-1}$.

**Fig. S14** Morphology and element distribution of SnO$_x$@C-G and SnO$_x$-G after 250 cycles. (a) SEM mapping and (c)TEM images of SnO$_x$@C-G. (b) SEM mapping and (d) TEM images of SnO$_x$-G. (e) STEM image and EDX mapping of SnO$_x$@C-G.
Fig. S15 (a) Equivalent circuit of SnO$_x$@C-G and SnO$_x$-G ($R_S$: electrolyte resistance, $R_{SEI}$: surface film resistance, $R_{CT}$: charge transfer resistance; CPE1–3: constant phase element, $Z_w$: Warburg impedance). Nyquist plots from in-situ EIS of (b) SnO$_x$@C-G and (c) SnO$_x$-G electrode with the fitted curves calculated by the above equivalent circuit.
Fig. S16 Coulombic efficiency of (a) SnO$_x$@C-G and (b) SnO$_2$@C-G electrode corresponding to the rate performances.

Fig. S17 Comparison of rate capacity with previous reported tin oxide/graphene material anodes.
Fig. S18 The charge/discharge profiles of (a) SnO$_x$@C-G and (b) SnO$_2$@C-G electrode at varied current densities of 0.05–3.2 A g$^{-1}$.

Fig. S19 CV results of the SnO$_2$@C-G electrode recorded at increasing scan rates from 0.2 to 1.0 mV s$^{-1}$. 
Fig. S20 Schematic illustration of mechanism of the capacitive effect and the protection of carbon coating in SnO$_x$@C-G as anode of lithium-ion batteries.
Fig. S21 (a) SEM image of SnO$_x$@C-GNS, insert: EDS spectrum. (b) TEM and (c) HR-TEM images of SnO$_x$@C-GNS. (d) Galvanostatic charge/discharge curves of the initial three cycles between 0.01 and 3.0 V at a current density of 0.1 A g$^{-1}$. (e) Cycling performances of SnO$_x$@C-GNS and GNS at a current density of 0.1 A g$^{-1}$ in the initial five cycles and 1.0 A g$^{-1}$ in the subsequence cycles. (f) EIS spectra of SnO$_x$@C-GNS before and after cycling measurements. (g) CV curves of the SnO$_x$@C-GNS electrode recorded at increasing scan rates from 0.1 to 1.5 mV s$^{-1}$. (h) Charge storage contributions of SnO$_x$@C-GNS from capacitive effects and diffusion-controlled process separated with CV at a scan rate of 1.5 mV s$^{-1}$. (i) Normalized contribution ratio of capacitive capacities at different scan rates.
<table>
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<th>Sample</th>
<th>Voltage range (V)</th>
<th>Current density (A g⁻¹)</th>
<th>Cycle number</th>
<th>Capacity (mA h g⁻¹)</th>
<th>Reference</th>
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<tbody>
<tr>
<td>SnOₓ@C-G</td>
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<td>1.6</td>
<td>250</td>
<td>740</td>
<td>this work</td>
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<tr>
<td>SnO₂@P@GO</td>
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References

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