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## Development of Si-based Composite Anode for Superior Li-ion Battery

複合式矽基負極應用於高性能鋰離子電池之研發

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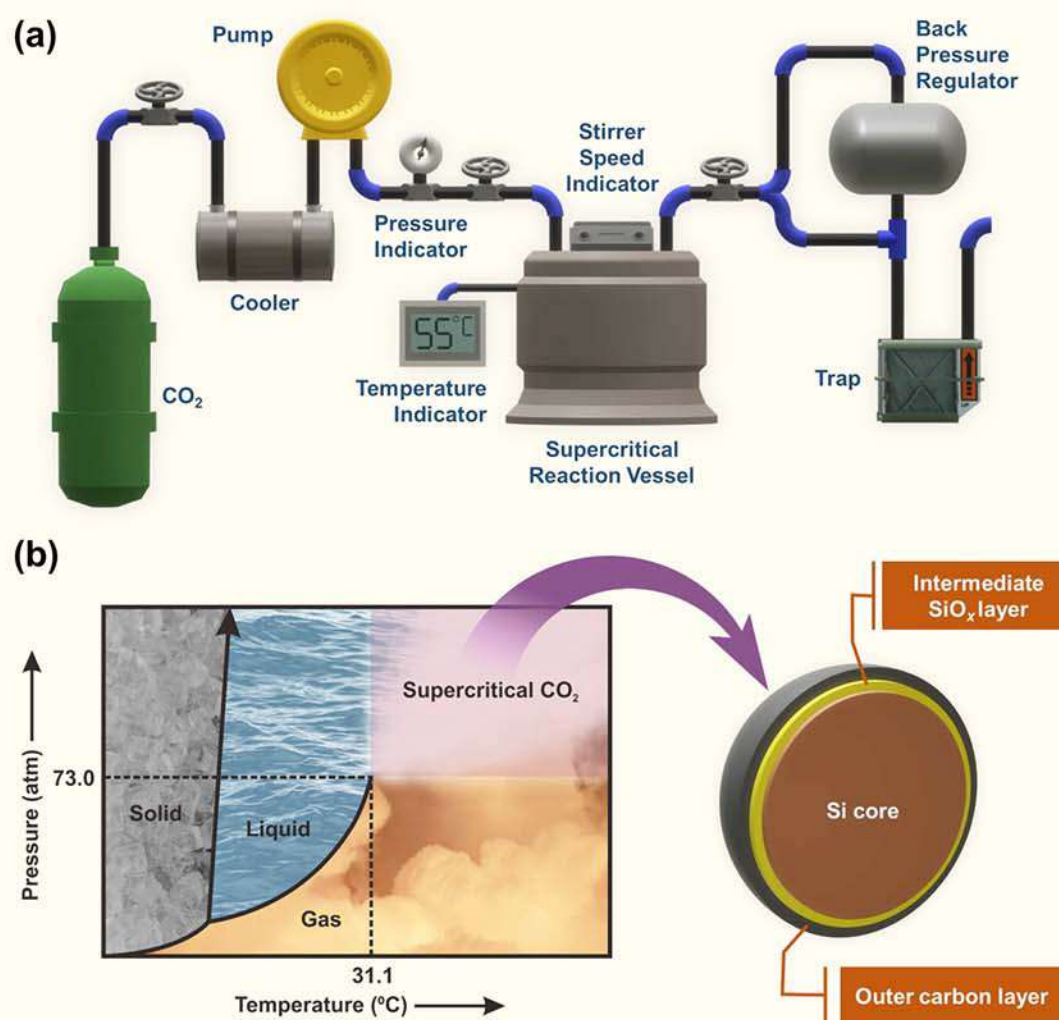
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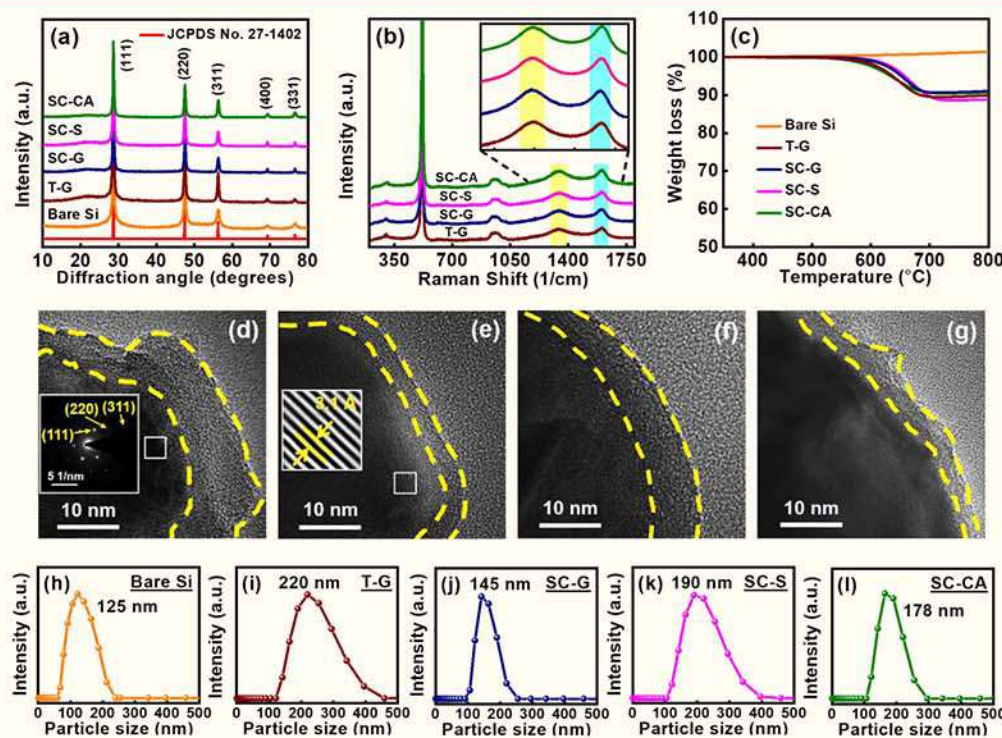
### ABSTRACT

A silicon-based anode in lithium-ion batteries (LIBs) has a promising stage for future battery technology. However, its high fluctuation in volume expansion and compression during lithiation-delithiation, tend to demolish the structure, initiate an unstable SEI formation. A buffer matrix to restrain the volume change is important to achieve a more stable performance in Si-based anode. Supercritical carbon dioxide (SCCO<sub>2</sub>), is applied to create a SiO<sub>x</sub>/carbon multi-layer coating on Si particles. Interaction of SCCO<sub>2</sub> with Si produces a continuous SiO<sub>x</sub> layer, which can buffer Si volume change during lithiation/delithiation. Furthermore, a conformal carbon film is deposited around the Si/SiO<sub>x</sub> core. Compared to the carbon film produced via a conventional wet-chemical method, the SCCO<sub>2</sub>-deposited carbon has significantly fewer oxygen-containing functional groups and thus higher electronic conductivity. The great potential of these proposed unique anode materials with facile synthesis methods for Li-ion battery applications is well investigated in our study.

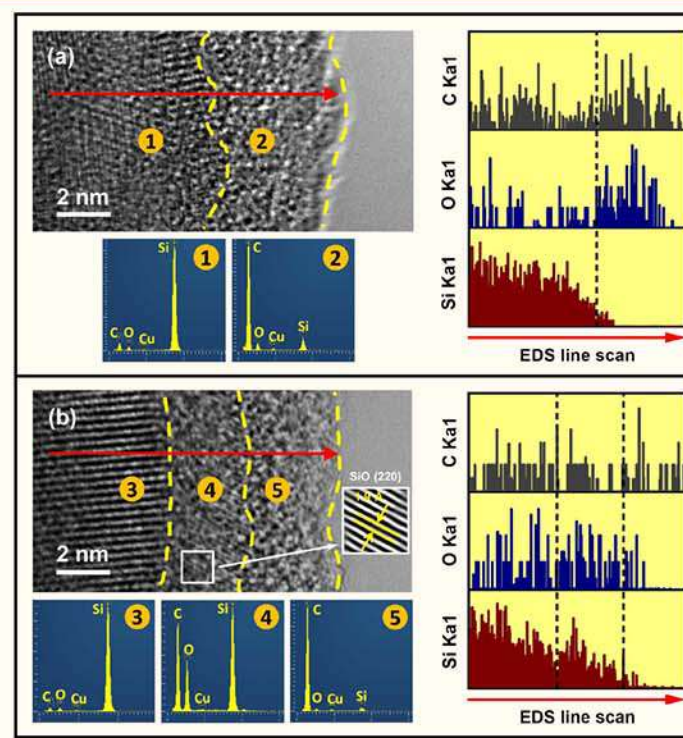
**Keywords:** carbon precursors, composite, green process, lithium-ion, silicon-based anodes.



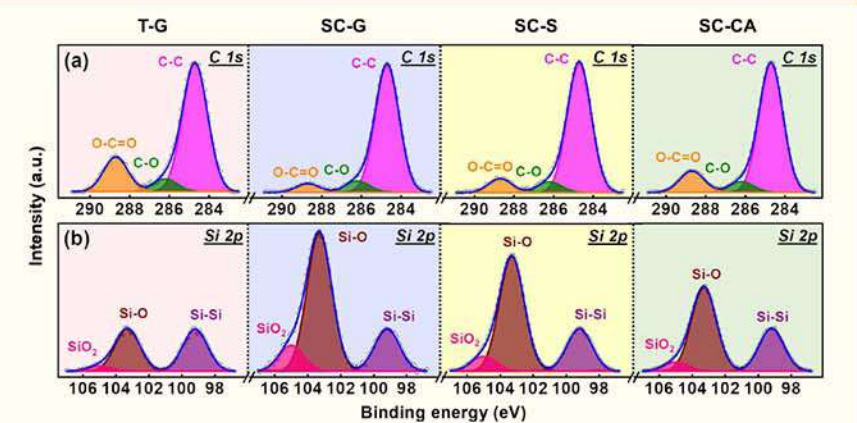
**Figure 1.** Schematic illustration of (a) SCCO<sub>2</sub> apparatus. (b) Phase diagram of CO<sub>2</sub> and scheme of SCCO<sub>2</sub>-fabricated C/SiO<sub>x</sub>/Si particle.



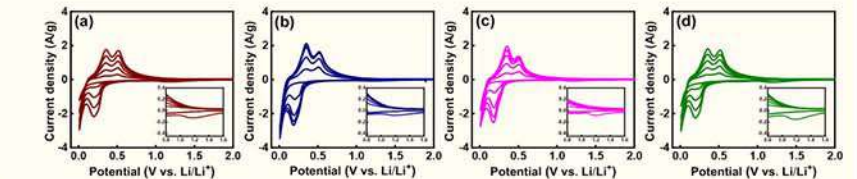
**Figure 2.** (a) XRD patterns, (b) Raman spectra, and (c) TGA curves of bare Si and various coated Si samples. High-resolution TEM images of (d) T-G, (e) SC-G, (f) SC-S, and (g) SC-CA. Particle size distribution data of (h) bare Si, (i) T-G, (j) SC-G, (k) SC-S, and (l) SC-CA measured using DLS.



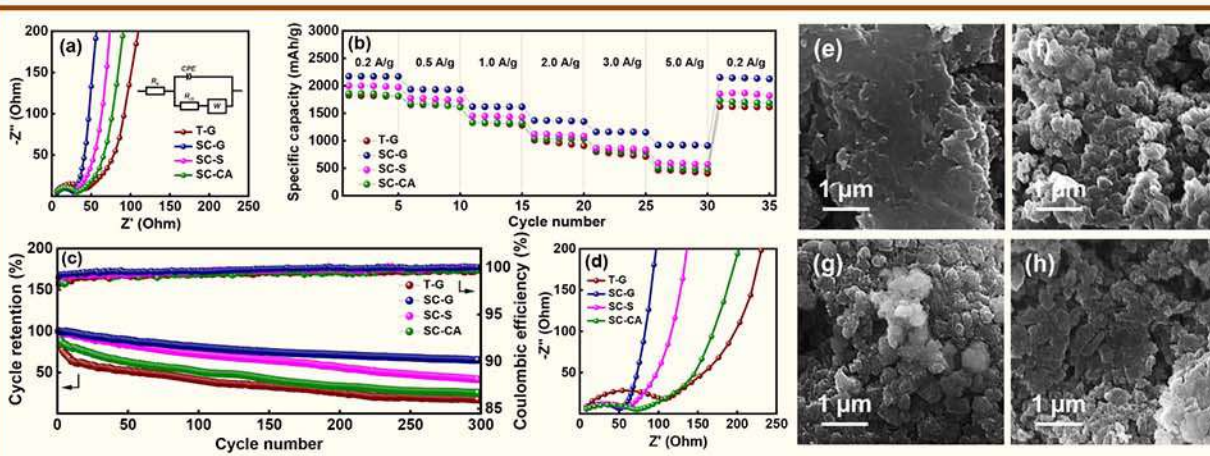
**Figure 3.** High-resolution TEM images, EDS spectra, and EDS line-scan data of (a) T-G and (b) SC-G samples. The EDS spectra are taken at the positions labeled in the TEM images.



**Figure 4.** XPS (a) C 1s and (b) Si 2p spectra of various samples.



**Figure 5.** CV curves of (a) T-G, (b) SC-G, (c) SC-S, and (d) SC-CA electrodes measured at a potential sweep rate of 0.1 mV s<sup>-1</sup>.



**Figure 6.** (a) EIS spectra of various electrodes and equivalent circuit used for data fitting. (b) Comparative rate performance of various electrodes. (c) Cycling stability of various electrodes measured at 1 A g<sup>-1</sup>. (d) EIS spectra of various electrodes after 300 charge-discharge cycles. SEM images of (e) T-G, (f) SC-G, (g) SC-S, and (h) SC-CA electrodes after cycling.

### SUMMARY

A SCCO<sub>2</sub> coating method for producing C/SiO<sub>x</sub> multilayers on Si nanoparticles was developed. The low oxygen-containing functional groups of the carbon layer, leading to higher electronic conductivity. Its first-cycle Coulombic efficiency was 84%. After 300 cycles, the electrode retains ~65% of its initial capacity. Thus, the proposed anode and material design/synthesis strategy have great potential for high-energy-density and high-power-density LIBs applications.

### REFERENCE

*Adv. Funct. Mater.* 2021, 31, 2104135.



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