

## Supporting Information

# Mitigating Interfacial Reactions in $\text{Li}_4\text{Ti}_5\text{O}_{12}$ Anodes through Carbon Shells Synthesized by Spray Granulation

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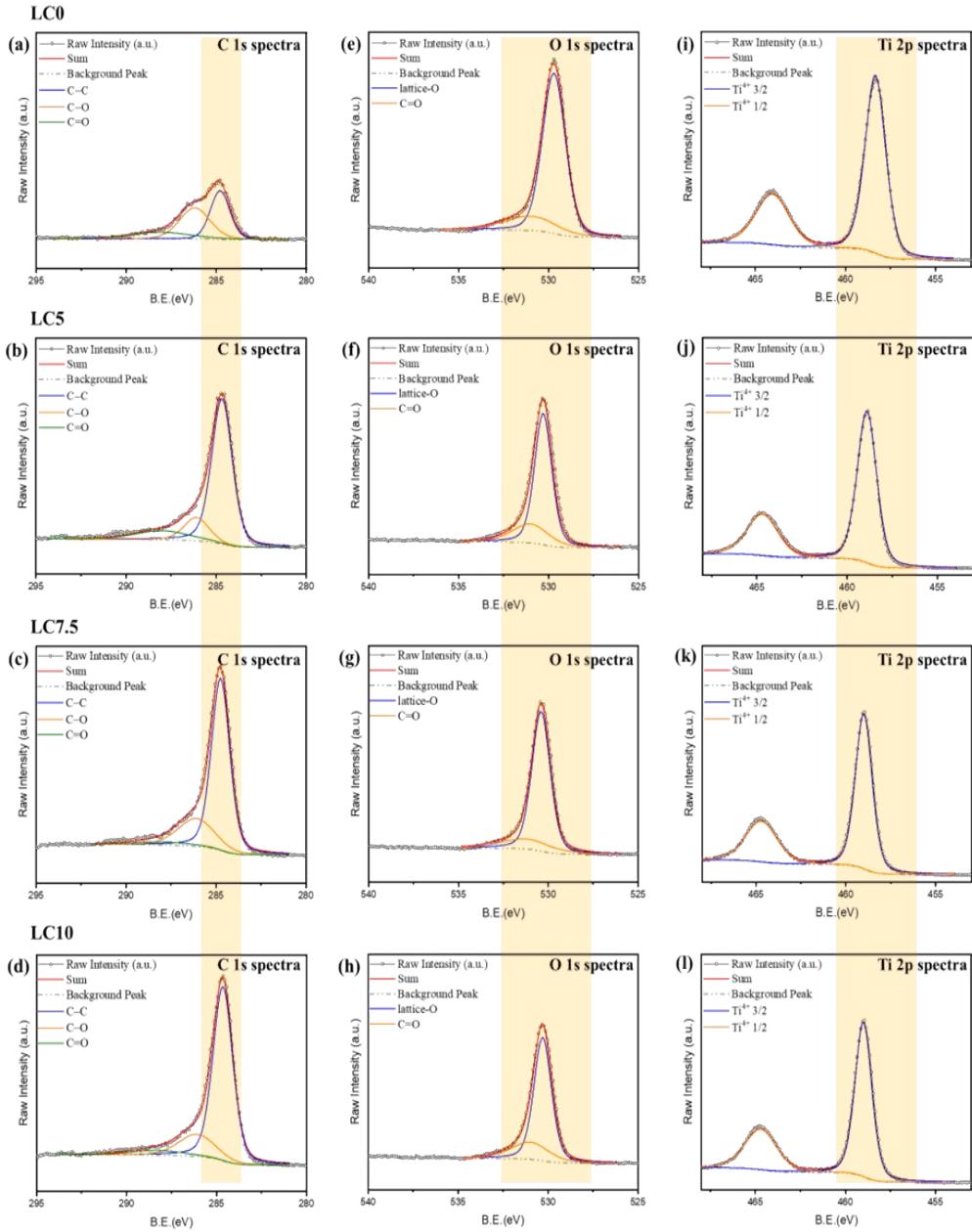
**Table S1.** Lattice parameters of LC0, LC5, LC7.5, and LC10.

Sample	$a$ (Å)	V (Å <sup>3</sup> )	R profile
LC0	8.358	583.9	4.50
LC5	8.357	583.8	4.18
LC7.5	8.357	583.8	4.59
LC10	8.358	583.9	4.37

**Table S2.** Particle size distribution and Zeta potential of LC0, LC5, LC7.5, and LC10.

Sample	D10 (μm)	D50 (μm)	D90 (μm)	Zeta Potential (mV)
LC0	4.36	8.14	13.76	65.04±1.5
LC5	5.14	8.24	12.93	-63.03±1.5
LC7.5	5.27	8.73	13.98	-55.57±1.5
LC10	5.16	8.73	14.13	-61.03±2

The zeta potentials of all samples were measured using an electrophoretic light scattering spectrophotometer (ELS-2000, Otsuka Electronics Co., Osaka, Japan). A dispersion of 0.05 g of LTO and LTO@C in 15 ml deionized water was prepared in a quartz cell to determine the electrophoretic mobility.

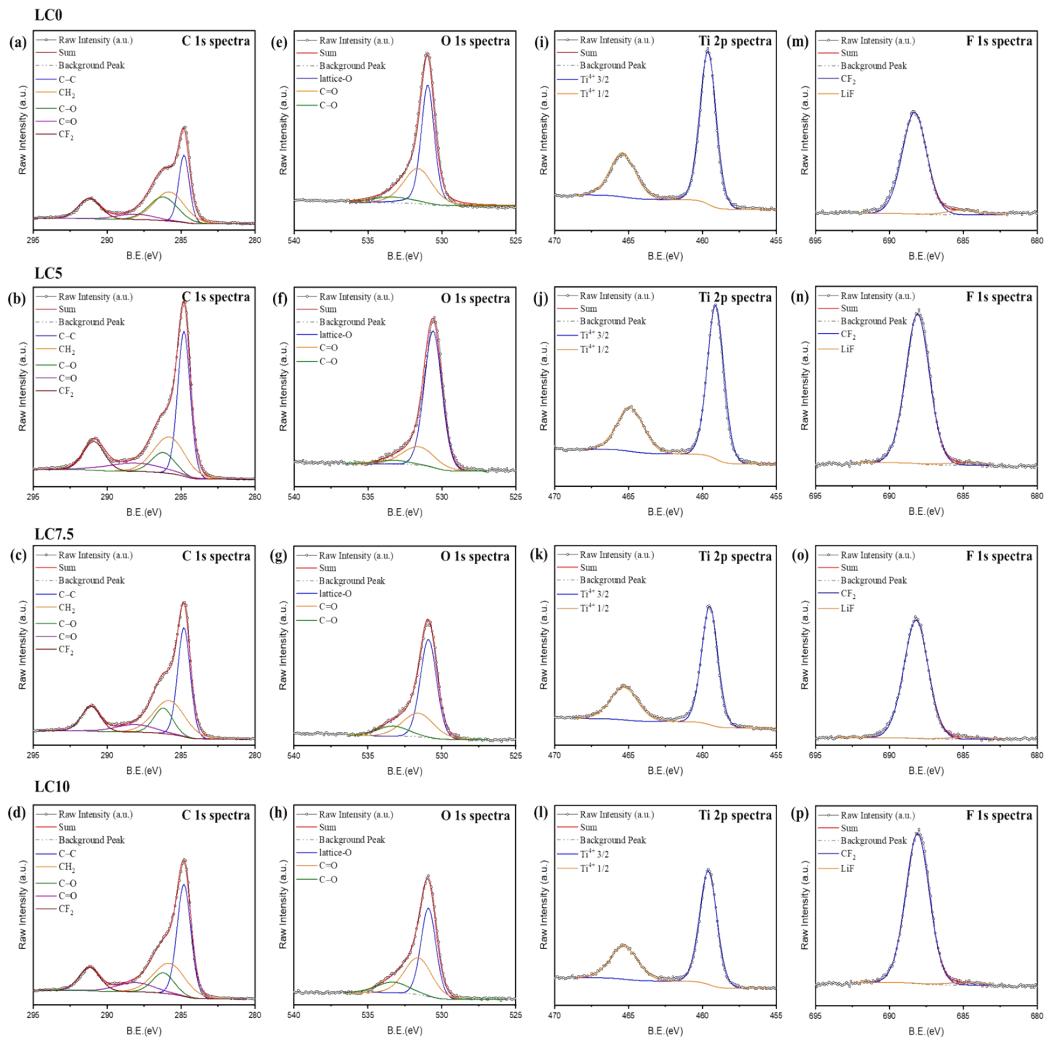


**Figure S1.** (a-d) C 1s, (e-h) O 1s, and (i-l) Ti 2p XPS spectra of LC0, LC5, LC7.5, and LC10 powders.

We employed XPS analysis to investigate the differences in the C 1s, O 1s, and Ti 2p core-level spectra of the LTO and LTO@C powders, as shown in **Figure S1**. Obviously, the core-level spectra of LTO@C exhibited changes with increasing sucrose concentrations (5%, 7.5%, and 10%) following carbon coating. In the C 1s

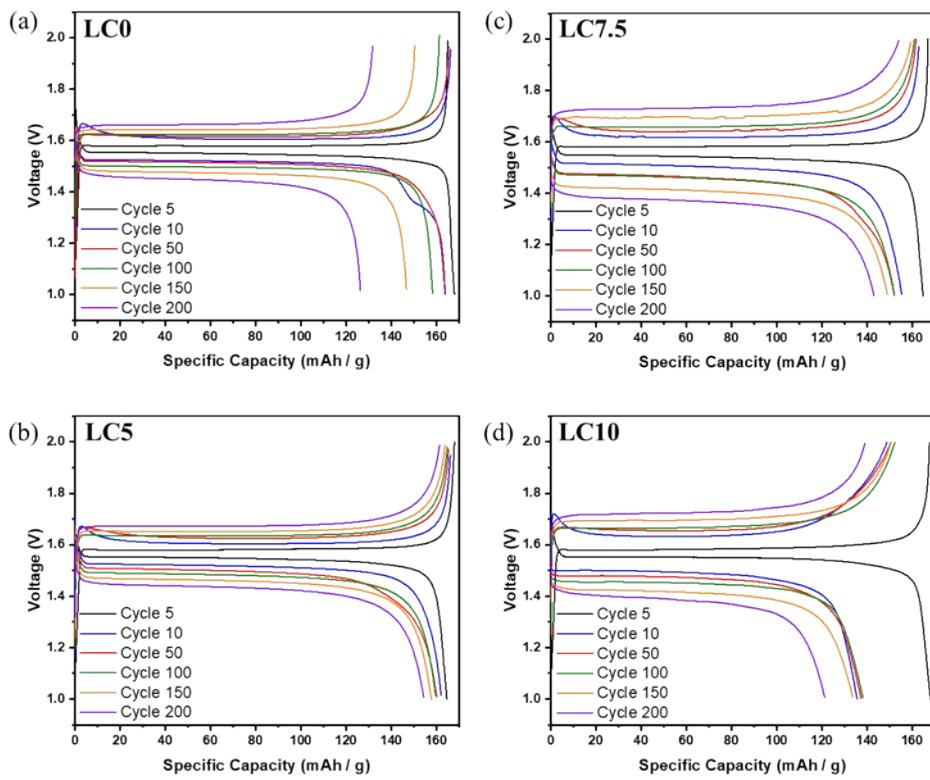
spectrum (**Figures S1 (b)-(d)**), the C-C signal intensity gradually increased due to the carbon coating, which also influenced the O 1s and Ti 2p spectra. The lattice oxygen signal intensity in the O 1s and Ti 2p spectra of LTO@C significantly decreased due to the carbon coating, with the signal peaks shifting to higher binding energies. [1]

As shown in **Figures S1 (a), (e), and (i)**, the C 1s spectrum of LTO powder showed signals at 284.8 eV (C-C), 286.1 eV (C-O), and 289 eV (C=O). The C-C signal mainly originates from adventitious carbon, while C=O is contributed by the residual  $\text{Li}_2\text{CO}_3$  on the surface. In the O 1s spectrum, the signal at 529.8 eV mainly corresponds to the lattice oxygen of LTO, while the signal at 531 eV (C=O) is attributed to  $\text{Li}_2\text{CO}_3$ . The Ti 2p spectrum consists of a doublet of peaks located at 458.3 eV and 464 eV, corresponding to the  $\text{Ti}^{4+}$  ion of LTO. [2, 3]

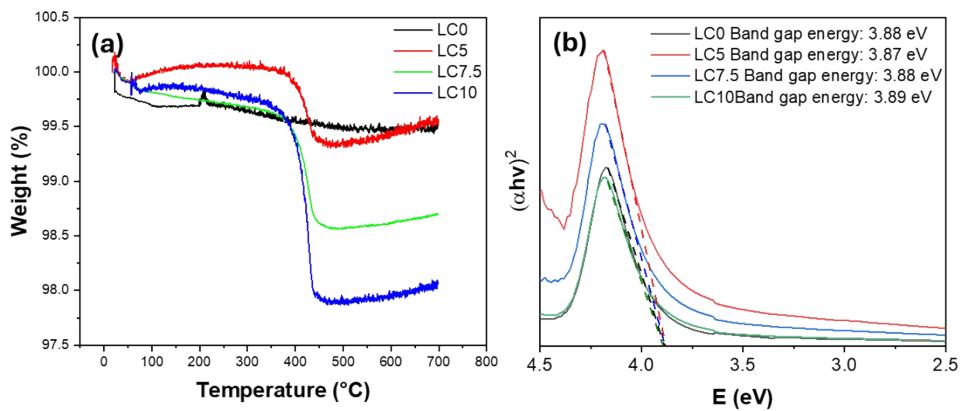


**Figure S2.** The XPS spectra of the LC0, LC5, LC7.5, and LC10 electrodes before 200 cycles. (a-d) C 1s, (e-h) O 1s, (i-l) Ti 2p, and (m-p) F 1s regions.

**Figure S2** presents the XPS spectra of the LC0, LC5, LC7.5, and LC10 electrodes. The C 1s spectra reveal additional signals at 285.7 eV ( $\text{CH}_2$ ) and 291 eV ( $\text{CF}_2$ ) beyond those observed from the powder, attributed to the PVDF binder. The (C-C) signal also includes contributions from carbon black (Super P). The O 1s and Ti 2p spectra remained largely unchanged compared to those of the powder. In the F 1s spectra, signals at 685 eV (LiF) and 688 eV ( $\text{CF}_2$ ) were detected, with the  $\text{CF}_2$  signal originating from PVDF and the small LiF peak likely resulting from surface contamination during sample preparation. [4-7]



**Figure S3.** Charge-discharge curves (a) LC0, (b) LC5, (c) LC7.5, and (d) LC10 under 1C with potential range examined from 1 V to 2 V.



**Figure S4.** (a) TGA curves of LC0, LC5, LC7.5, and LC10 powders. (b) Tauc plots of LC0, LC5, LC7.5, and LC10 samples derived from UV-Vis reflectance spectra.

## References

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