

# Electronic Supplementary Information

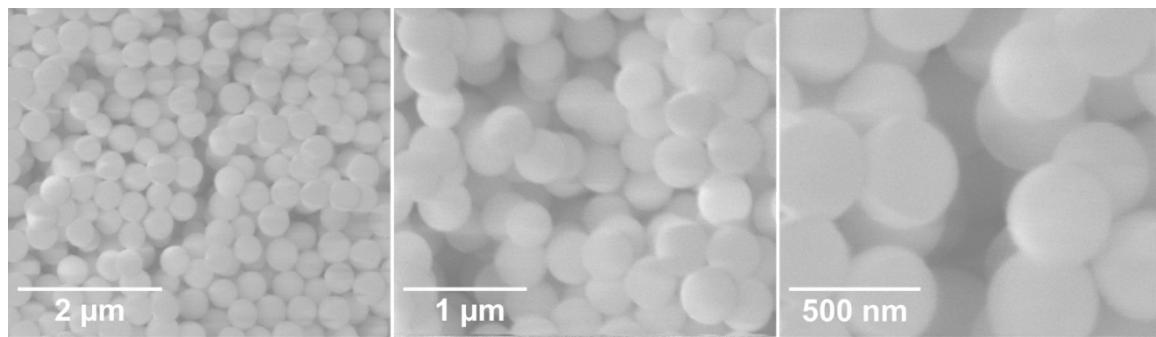
## CVD-Assisted Fabrication of Hierarchical Microparticulate Li<sub>2</sub>TiSiO<sub>5</sub>-Carbon Nanospheres for Ultrafast Lithium Storage

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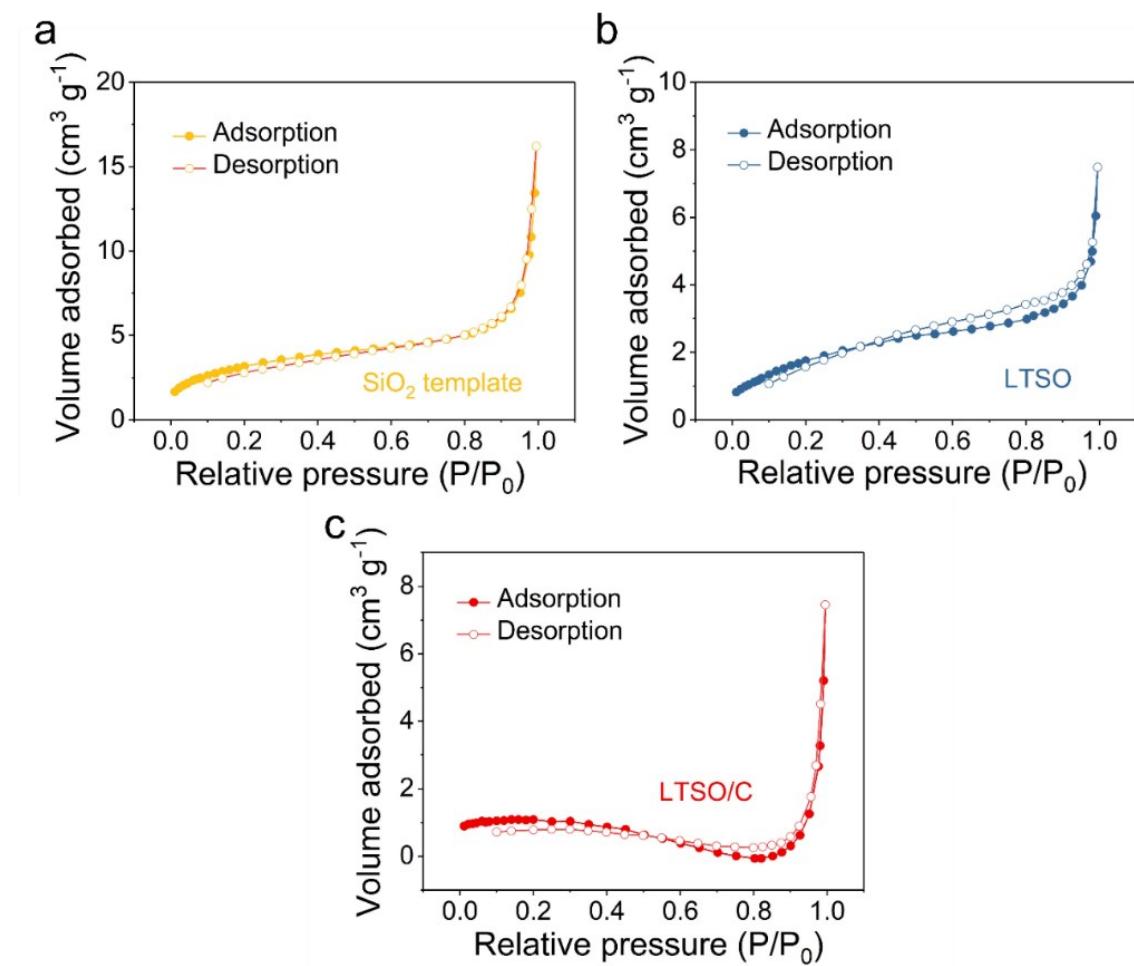
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## Supporting information



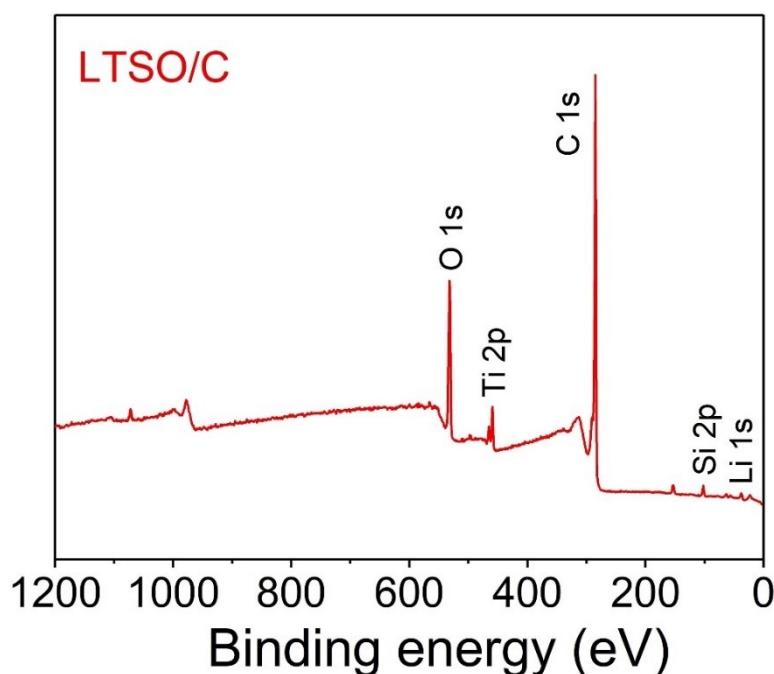
**Figure S1.** SEM images of SiO<sub>2</sub> template particles.



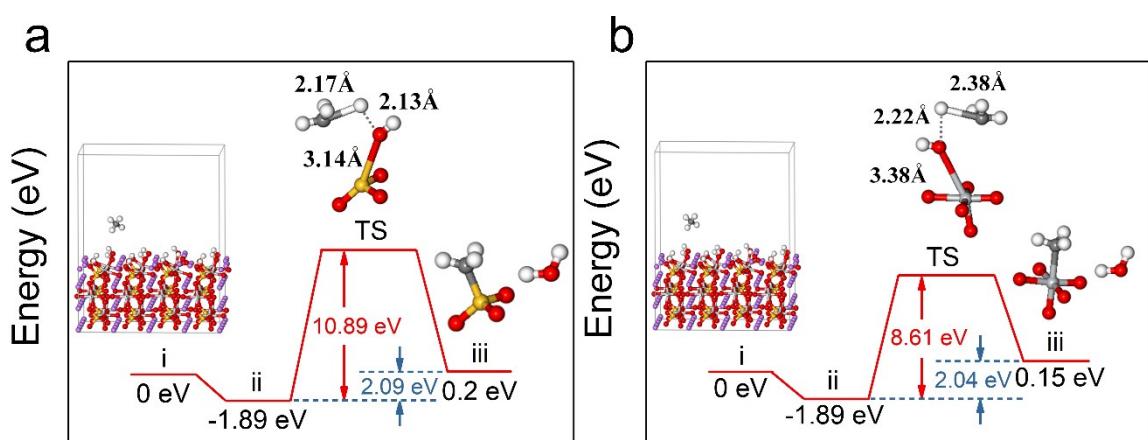
**Figure S2.** N<sub>2</sub> adsorption/desorption isotherms: a) SiO<sub>2</sub> template; b) LTSO and c) LTSO/C.



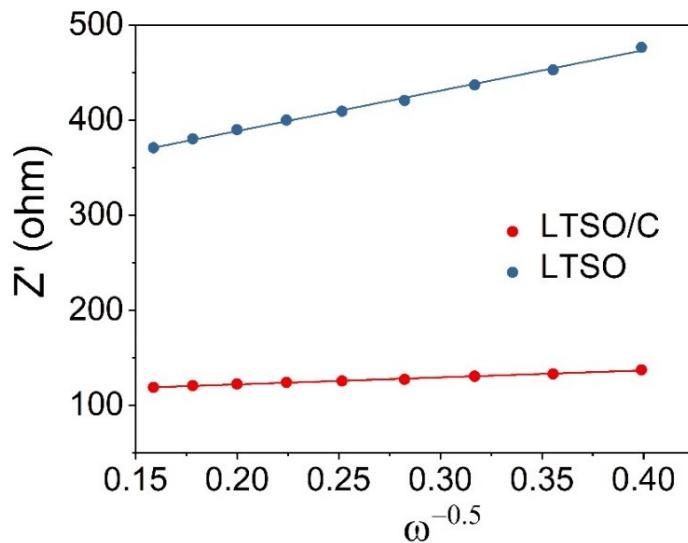
**Figure S3.** Illustration of tape density measurement of 2.5 g LTSO/C in a 4 ml glass vial.



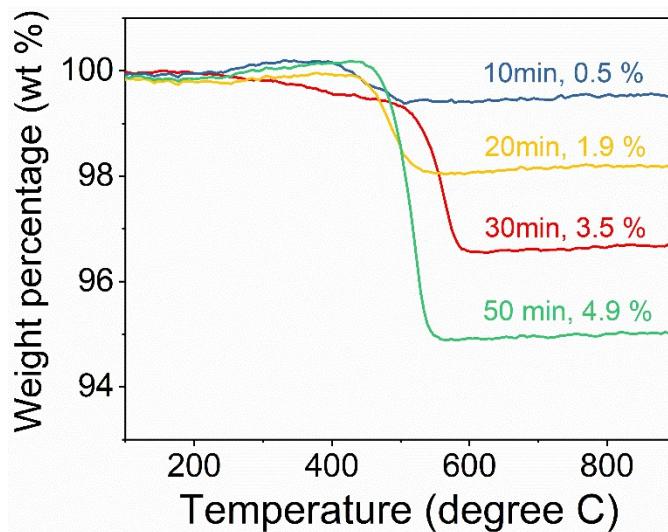
**Figure S4.** Full XPS spectra of LTSO/C.



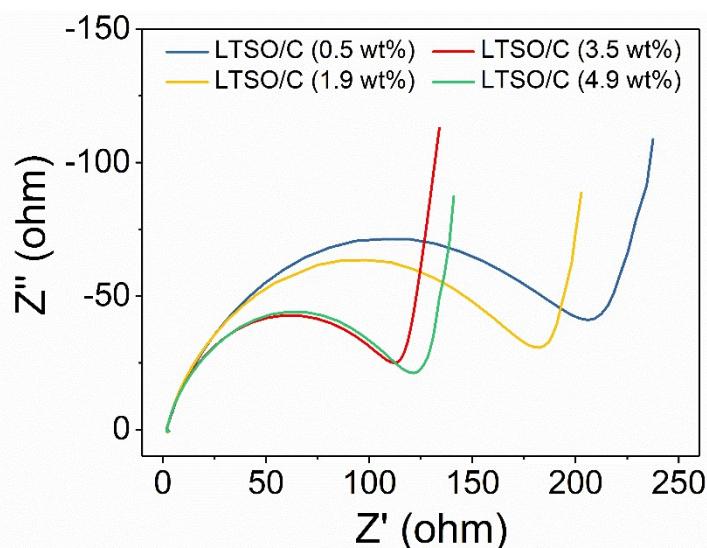
**Figure S5.** Reaction paths in CVD for LTSO with the different active sites: a) Si–OH group as active site; b) Ti–OH as active site.



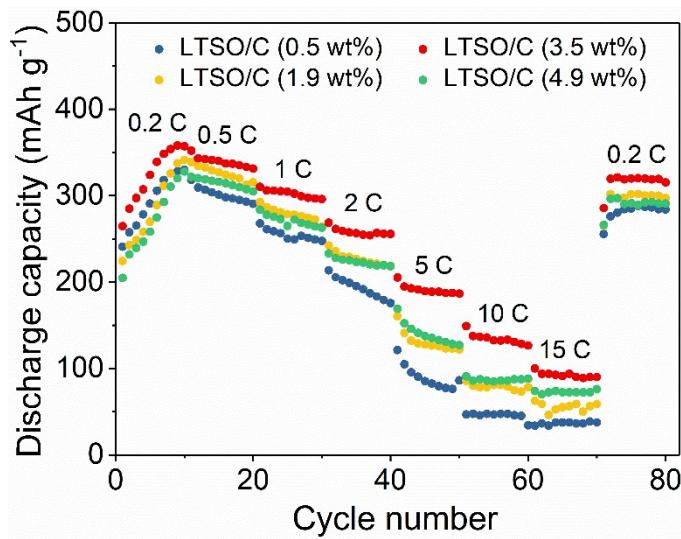
**Figure S6.** Fitting lines for calculation of  $\text{Li}^+$  diffusion coefficient in the Warburg diffusion region.



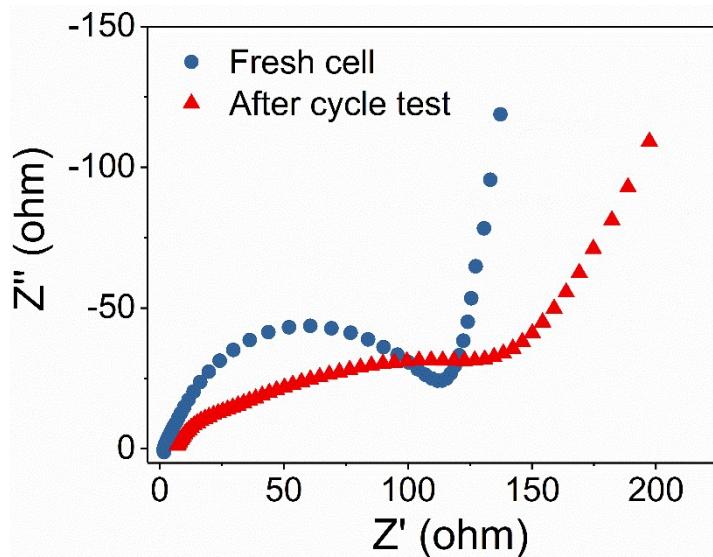
**Figure S7.** TG results of LTSO/C composites with different CVD treatment durations.



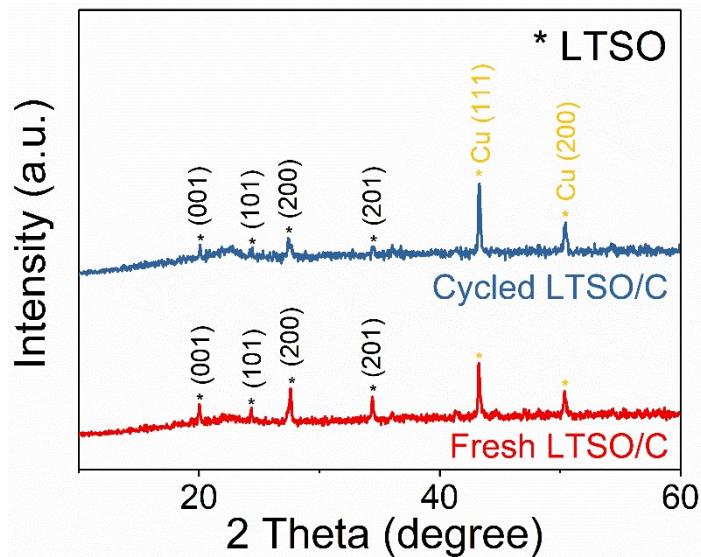
**Figure S8.** Nyquist plots of LTSO/C composites with different carbon contents.



**Figure S9.** Rate performances of LTSO/C composites with different carbon contents.



**Figure S10.** EIS results of fresh and cycled LTSO/C cells.



**Figure S11.** XRD results of fresh and cycled LTSO/C cells.

**Table S1.** Fitting results of electrochemical impedance spectroscopy (EIS).

Materials	$R_s$ (ohm)	$R_{ct}$ (ohm)
LTSO	2.1	350.8
LTSO/C	1.7	115.6

**Equation S1.** Lithium-ion diffusion coefficient at open circuit.

The  $\text{Li}^+$  diffusion coefficient can be figured out through the following equation:

$$D_{Li^+} = R^2 T^2 / 2 A^2 n^4 F^4 C^2 A_w^2$$

Where  $D_{Li^+}$  represents the  $\text{Li}^+$  diffusion coefficient in LTSO ( $\text{cm}^2 \text{s}^{-1}$ ),  $R$  represents the gas constant ( $8.31 \text{ J mol}^{-1} \text{ K}^{-1}$ ),  $T$  represents the absolute temperature (298 K),  $A$  represents the surface area of the electrode ( $0.785 \text{ cm}^2$ ),  $n$  represents the number of electrons transferred per molecule during the electrochemical reaction,  $F$  represents the Faraday constant (96485  $\text{C mol}^{-1}$ ),  $C$  represents the molar concentration of  $\text{Li}^+$  in LTSO, and  $A_w$  represents the Warburg coefficient associated with  $Z'$  and  $\omega^{-0.5}$ .

**Equation S2.** Randles-Sevcik equation.

$$I_p = 2.69 \times 10^5 n^{1.5} A D_{Li^+}^{0.5} C v^{0.5}$$

Where  $I_p$  represents the peak current in amps,  $n$  represents the number of electrons transferred during the redox reaction per molecule,  $A$  represents the surface area of the electrode ( $\text{cm}^2$ ),  $C$  represents the concentration of  $\text{Li}^+$  in LTSO ( $\text{mol cm}^{-3}$ ),  $D_{Li^+}$  represents the diffusion coefficient of  $\text{Li}^+$  in LTSO ( $\text{cm}^2 \text{s}^{-1}$ ), and  $v$  represents the scanning rate ( $\text{V s}^{-1}$ ).

According to this principle,  $D_{Li^+}$  is proportional to  $I_p^2$ .

	LTSO/C						Graphite						Ref 39 - Graphite <sup>1</sup>						
Tap density (g mL <sup>-1</sup> )	1.3						1.1						1.1						
Carbon content	Graphitic carbon / 3.5 wt%						Graphite						Graphite flakes						
Current density (A g <sup>-1</sup> )	0.15	0.3	0.6	1.5	3	4.5	0.2	0.5	1	2	5	10	0.2	0.5	1	2			
Volumetric capacity (mAh cm <sup>-3</sup> )	441.1	393.1	335.5	248.6	174.6	120.1	418	363	275	91.3	46.2	14.3	385	297	176	77			
	Ref 40 - LTO <sup>2</sup>					Ref 41 - LTO <sup>3</sup>						Ref 42 - LTO <sup>4</sup>				Ref 43 - LTO <sup>5</sup>			
Tap density (g mL <sup>-1</sup> )	1.23					0.82						1.4				1.78			
Carbon content	Carbide / 1.88 wt%					Carbide / 6 wt%						N/A				Carbide / 28 wt%			
Current density (A g <sup>-1</sup> )	0.175	0.35	0.875	1.75	3.5	0.16	0.32	0.64	1.28	1.6	3.2	0.175	0.35	0.525	0.875	0.175	0.35	0.875	1.75
Volumetric capacity (mAh cm <sup>-3</sup> )	199.8	195.3	180.3	170	136.5	123	118.9	113.2	109.1	105.8	103.3	233.8	214.2	200.2	169.4	329.3	284.8	208.3	178
	Ref 44 - TiO <sub>2</sub> <sup>6</sup>					Ref 45 - TiO <sub>2</sub> <sup>7</sup>					Ref 46 - TiO <sub>2</sub> <sup>8</sup>					Ref 47 - TiO <sub>2</sub> <sup>9</sup>			
Tap density (g mL <sup>-1</sup> )	N/A					1.1					1.06					0.63			
Carbon content	N/A					N/A					Graphene oxide / 1.2 wt%					Graphene oxide / 4.4 wt%			
Current density (A g <sup>-1</sup> )	0.1675	0.335	0.67	1.34	0.2	0.5	1	2	0.168	0.336	0.84	1.68	3.36	0.5	1	1.5	2	3	
Volumetric capacity (mAh cm <sup>-3</sup> )	370	260	155	75	315	245	195	145	200.4	177	144.6	107.8	79.5	130.2	120.3	115.2	112.8	106.8	

**Table S2.** Detailed data sheets of electrochemical performance comparison result for Fig. 5d in the main text.

## References

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