

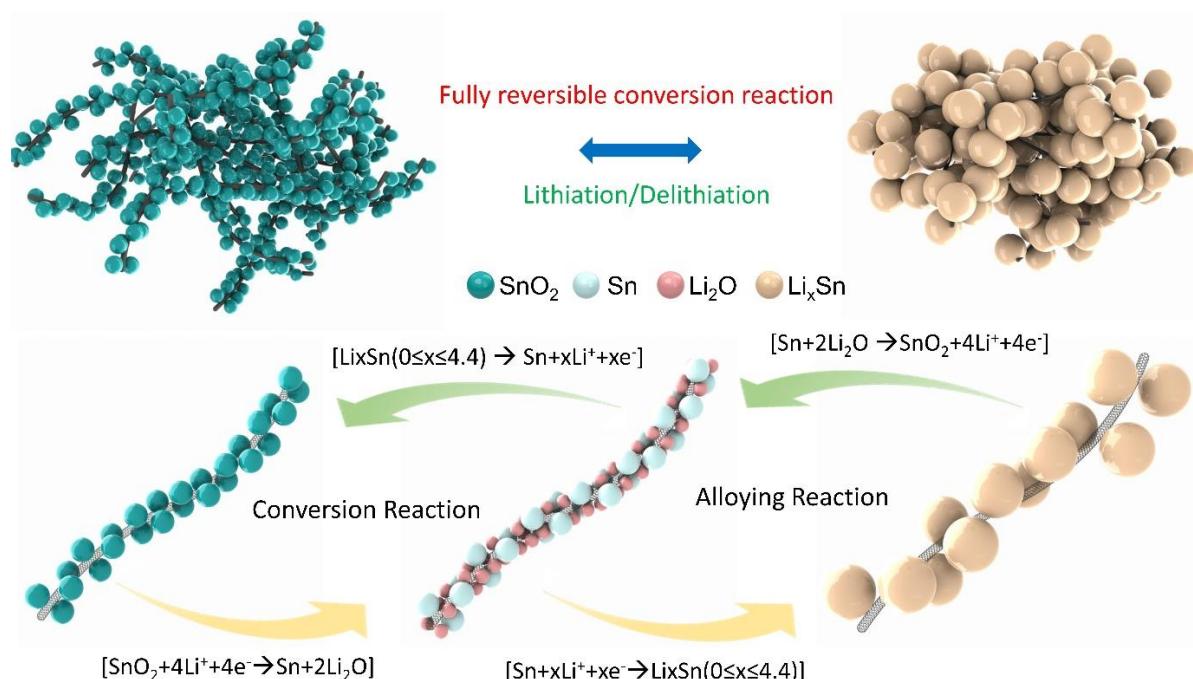
## Supplementary Information

### Unconventional Capacity Increase Kinetics of Chemically Engineered SnO<sub>2</sub> Aerogel Anode toward Long-Term Stable Lithium-Ion Batteries

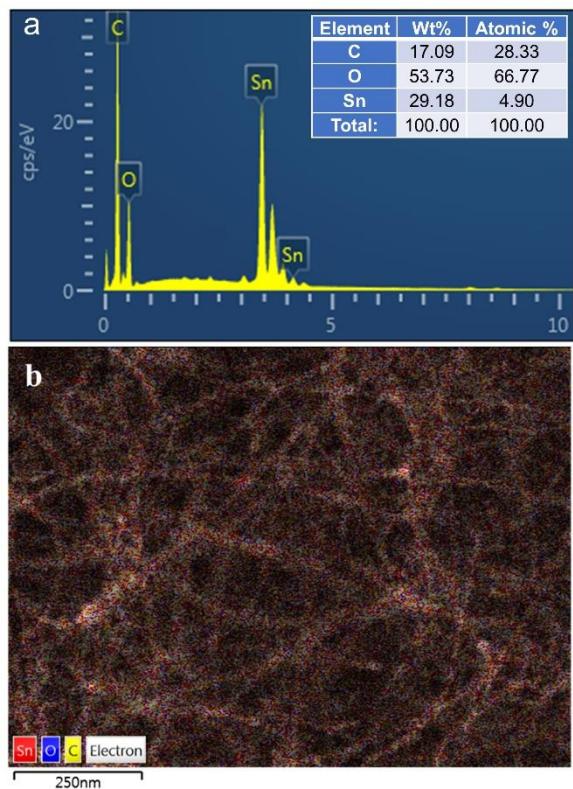
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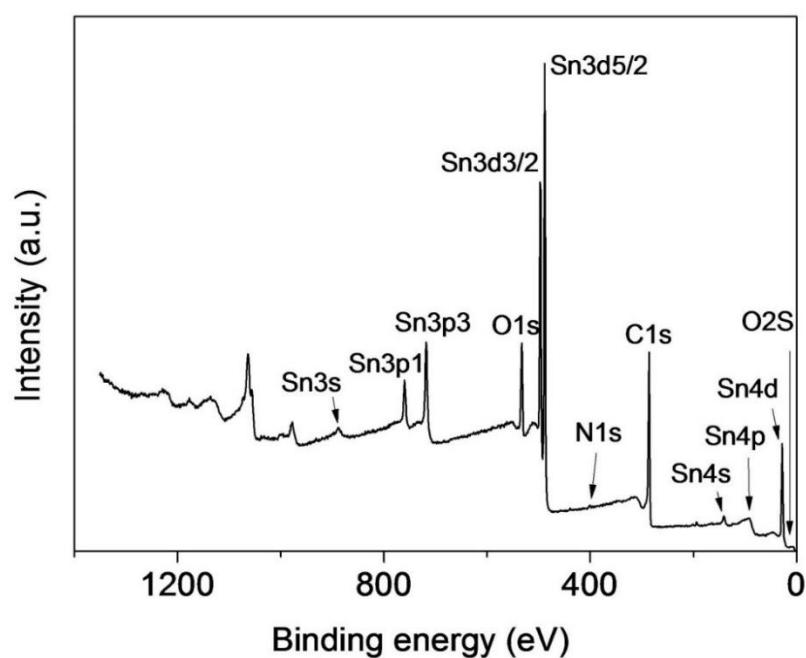
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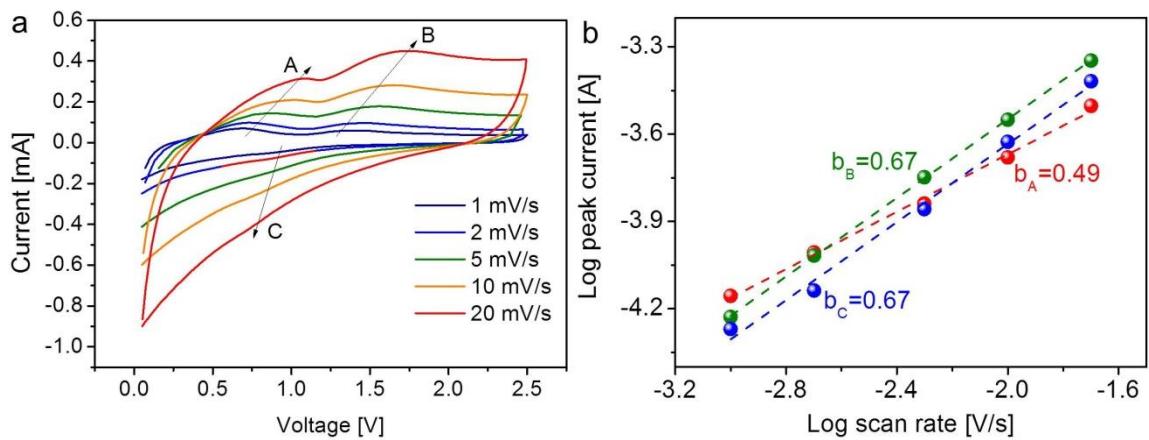
**Figure S1.** Overall electrochemical reactions between Li-ions and SnO<sub>2</sub> nanoparticles occurring during the lithiation/delithiation processes.



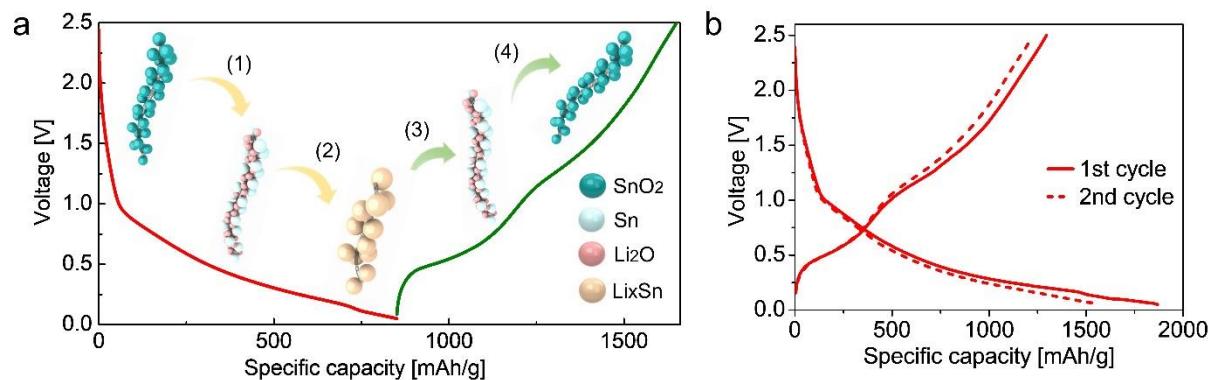
**Figure S2.** (a) EDS spectrum and (b) element mapping image of the  $\text{SnO}_2$  aerogel active material.



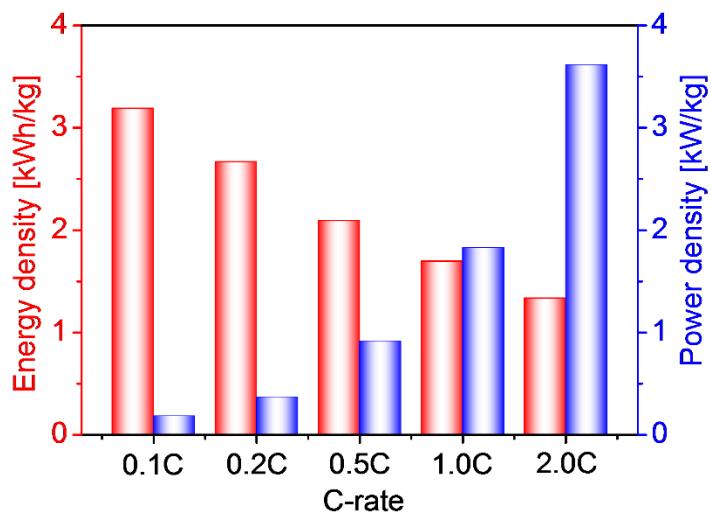
**Figure S3.** The full XPS spectrum of the  $\text{SnO}_2$  aerogel active material.



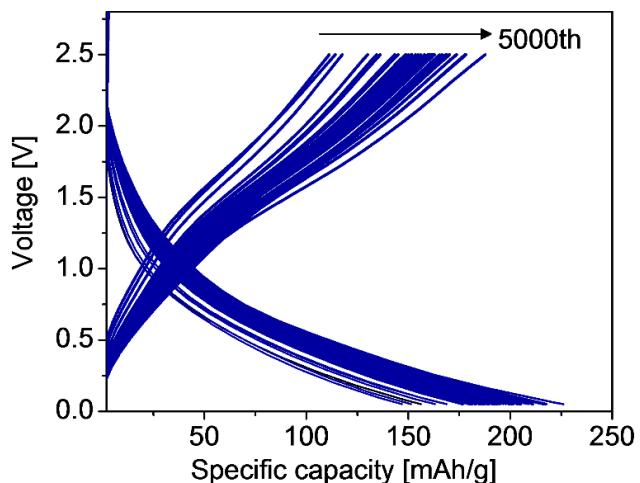
**Figure S4.** (a) Sequential CV curves with various scan rates of 1, 2, 5, 10, and 10 mV/s. (b)  $b$ -value at the anodic (A, B) and cathodic (C) peaks in the CV curves.



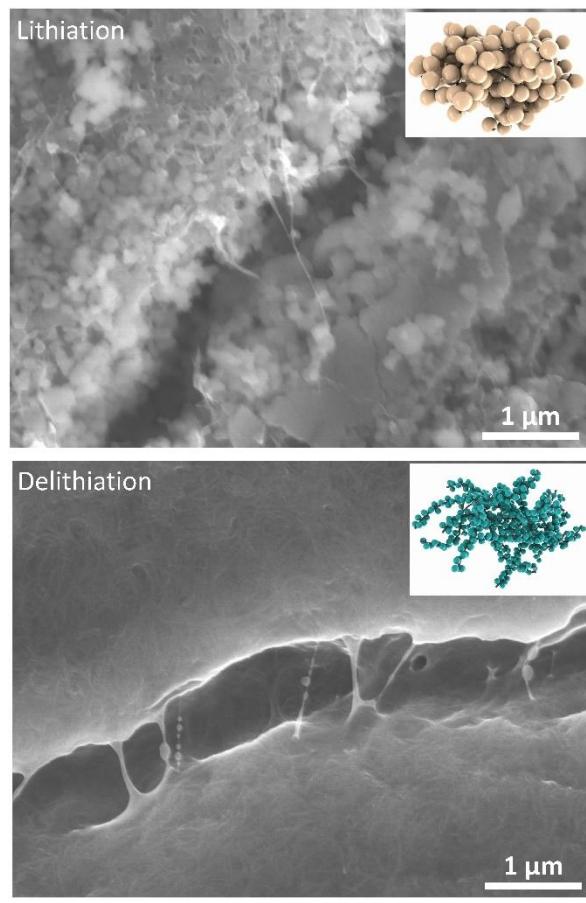
**Figure S5.** (a) Reaction mechanism according to the lithiation/delithiation cycle, which was representatively measured at a current density of 395 mA g<sup>-1</sup>. (b) Curves of first and second lithiation/delithiation processes at a current density of 79 mA g<sup>-1</sup>.



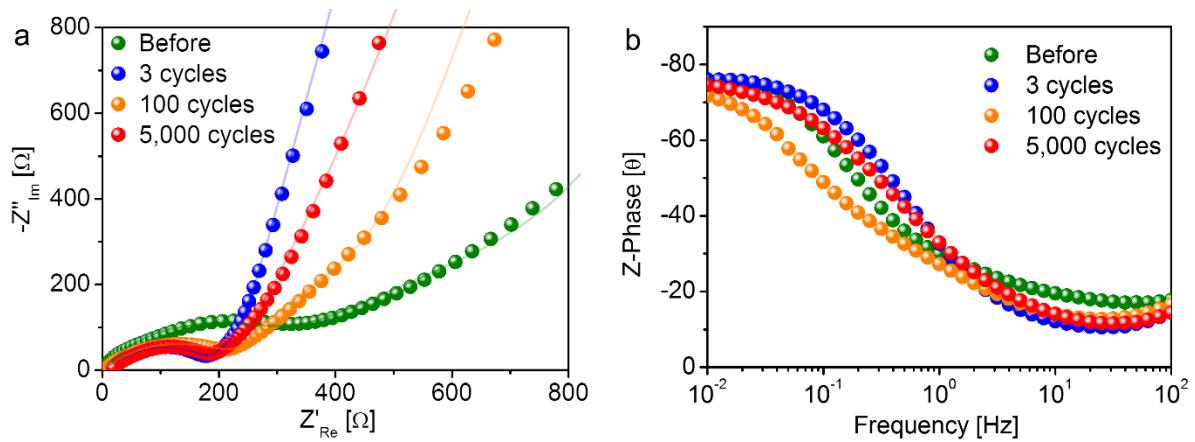
**Figure S6.** Energy and power densities of the SnO<sub>2</sub> aerogel device extracted from low C-rates (0.1 C to 2 C) of Figures 4a and 4b.



**Figure S7.** Real time-charge/discharge curves of the SnO<sub>2</sub> aerogel device at 10 C (7900 mA g<sup>-1</sup>).



**Figure S8.** FE-SEM images of the  $\text{SnO}_2$  aerogel electrode after 10 cycles at 10 C during lithiation and delithiation processes.



**Figure S9.** (a) Nyquist plots at the high- and middle frequency region. (b) Z-phase vs. Frequency as a function of cycles.

**Table S1.** Comparison of discharge capacity and energy density using SnO<sub>2</sub>/carbon compound electrodes.

Anode (SnO <sub>2</sub> contents)	OPW[V]	Discharge capacity [mAh/g]	Es[Wh/kg]	Ref.
3D porous SnO <sub>2</sub> aerogel (37wt%)	0.05~2.5	1361, 0.079A/g (5th cycles)	3188	this work
		1919, 0.79A/g (Recovery)	4528	
CNT/Perforated SnO <sub>2</sub> (~83.1wt%)	0.001~3.0	1108, 1.5A/g	3323	S1
CNT/c-SnO <sub>2</sub> (~72 wt%)	0.01~3.0	1140, 0.05A/g	3409	S2
CNTH/SnO <sub>2</sub> (~57.6wt%)	0.01~3.0	1109.5, 0.1A/g	3317	S3
MWCNT/SnO <sub>2</sub> (~75wt%)	0.001~2.5	~682, 0.05A/g	1704	S4
MWCNT/SnO <sub>2</sub> (~75.5wt%)	0.005~2.5	963, 0.0782A/g	2403	S5
SWNT Paper/SnO <sub>2</sub> (~34wt%)	0.01~2.0	~669.52, 0.025A/g	1332	S6
porous-CNT/SnO <sub>2</sub> (~64.7wt%)	0.01~3.0	968,0.1A/g	2894	S7
Activated CNT/SnO <sub>2</sub> (~65wt%)	0.01~2.5	829.5, 0.2mA/cm2	2037	S8
Gr/CNT/SnO <sub>2</sub> (~49.5wt%)	0.01~3.0	947, 0.1A/g	2832	S9
Gr/CNT/SnO <sub>2</sub> (~55.3wt%)	0.001~3.0	864, 0.05A/g	2273	S10
Carbon/CNT/SnO <sub>2</sub> (~62.39wt%)	0.01~2.5	1572, 0.2A/g	3914	S11
Carbon coated-CNT Sponge/SnO <sub>2</sub> (~22.9wt%)	0.01~3.0	~943, 0.1A/g	2820	S12
GF/ SnO <sub>2</sub> nanorod array/ PANI (~77wt%)	0.05~3.0	740, 0.1A/g	2183	S13
SGF(Gr Foam)/SnO <sub>2</sub> (~45.56wt%)	0.01~3.0	918.1, 0.2A/g	2745	S14
C@SnO <sub>2</sub> @C HNSs (~54wt%)	0.005~3.0	1123, 0.1A/g	3363	S15
Gr/SnO <sub>2</sub> (~67wt%)	0.01~3.0	1025, 0.1A/g	3065	S16

**Table S2.** Comparison of cycle stability using SnO<sub>2</sub>/carbon compound electrodes.

Anode (SnO <sub>2</sub> contents)	OPW[V]	Discharge capacity [mAh/g]	Cyclability [%] and conditions	Ref.
3D porous SnO <sub>2</sub> aerogel (37wt%)	0.05~2.5	1919, 0.79A/g	600	0.79A/g this work
		224, 7.9A/g	10,000	
CNT/Perforated SnO <sub>2</sub> (~83.1wt%)	0.001~3.0	1108, 1.5A/g	1000 (>74.5%)	4.0A/g S1
CNT/c-SnO <sub>2</sub> (~72 wt%)	0.01~3.0	1140, 0.05A/g	500 (>72.0%)	1.0A/g S2
CNTH/SnO <sub>2</sub> (~57.6wt%)	0.01~3.0	1109.5, 0.1A/g	100 (>74.2%)	0.2A/g S3
MWCNT/SnO <sub>2</sub> (~75wt%)	0.001~2.5	~682, 0.05A/g	100 (>10.1%)	0.05A/g S4
MWCNT/SnO <sub>2</sub> (~75.5wt%)	0.005~2.5	963, 0.0782A/g	100 (>90%)	3.91A/g S5
SWNT Paper/SnO <sub>2</sub> (~34wt%)	0.01~2.0	~669.52, 0.025A/g	100 (>67.8%)	0.025A/g S6
porous-CNT/SnO <sub>2</sub> (~64.7wt%)	0.01~3.0	968,0.1A/g	500 (>114.1%)	1.0A/g S7
Activated CNT/SnO <sub>2</sub> (~65wt%)	0.01~2.5	829.5, 0.2mA/cm2	50 (>89.7%)	1mA/cm2 S8
Gr/CNT/SnO <sub>2</sub> (~49.5wt%)	0.01~3.0	947, 0.1A/g	300(>78.0%)	0.6A/g S9
Gr/CNT/SnO <sub>2</sub> (~55.3wt%)	0.001~3.0	864, 0.05A/g	300(>55.9%)	1.0A/g S10
Carbon/CNT/SnO <sub>2</sub> (~62.39wt%)	0.01~2.5	1572, 0.2A/g	150(>60.7%)	1.0A/g S11
Carbon coated-CNT Sponge/SnO <sub>2</sub> (~22.9wt%)	0.01~3.0	~943, 0.1A/g	100(>90.56%)	0.1A/g S12
GF/ SnO <sub>2</sub> nanorod array/ PANI (~77wt%)	0.05~3.0	740, 0.1A/g	50(>76.8%)	0.5A/g S13
SGF(Gr Foam)/SnO <sub>2</sub> (~45.56wt%)	0.01~3.0	918.1, 0.2A/g	50(>73.9%)	0.2A/g S14
C@SnO <sub>2</sub> @C HNSs (~54wt%)	0.005~3.0	1123, 0.1A/g	1000(>92%)	10.0A/g S15
Gr/SnO <sub>2</sub> (~67wt%)	0.01~3.0	1025, 0.1A/g	300(>84%)	0.1A/g S16
dual carbon shells coated SnO <sub>2</sub> hollow nanospheres(41.7%)	0.01~3.0	694, 0.2A/g	300(78.7%)	0.2A/g S17
dual carbon shells coated SnO <sub>2</sub> hollow nanospheres(41.7%)	0.01~3.0	400, 5A/g	10000	5A/g S17
Sn-SnO <sub>2</sub> @CNT composite	0.01~3.0	744, 0.5A/g	1000(86%)	0.5A/g S18

**Table S3.** EIS fitted parameters of the SnO<sub>2</sub> aerogel cells before cycling and after 3, 100, and 5000 cycles. The fitted typical equivalent circuit model was conducted using ZsimpWin.

Samples	Rs ( $\Omega$ )	CPE <sub>SEI</sub> [Y <sub>0</sub> , (Ss <sup>n1</sup> cm <sup>-2</sup> )]	n <sub>1</sub>	R <sub>SEI</sub> ( $\Omega$ )	CPE <sub>CT</sub> [Y <sub>0</sub> , (Ss <sup>n2</sup> cm <sup>-2</sup> )]	R <sub>CT</sub> ( $\Omega$ )	n <sub>2</sub>	W (Y <sub>0</sub> , Ss <sup>0.5</sup> cm <sup>-2</sup> )	D <sub>Li<sup>+</sup></sub> [cm <sup>2</sup> s <sup>-1</sup> ]
Fresh	3.3	$8.72 \times 10^{-5}$	0.68	304.7	$3.24 \times 10^{-3}$	706.4	0.57	$0.562 \times 10^{-2}$	$2.38 \times 10^{-12}$
3 Cycles	3.7	$3.63 \times 10^{-4}$	0.71	28.5	$1.43 \times 10^{-2}$	195.2	0.86	$1.66 \times 10^{-2}$	$10.22 \times 10^{-12}$
100 Cycles	3.9	$4.24 \times 10^{-4}$	0.67	34.2	$3.22 \times 10^{-2}$	208.7	0.81	$2.32 \times 10^{-2}$	$17.22 \times 10^{-12}$
5,000 Cycles	3.8	$4.03 \times 10^{-4}$	0.62	33.5	$2.93 \times 10^{-2}$	190.2	0.81	$2.03 \times 10^{-2}$	$11.94 \times 10^{-12}$

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