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## **Electronic Supplementary Information (ESI)**

## Supporting Information for

## In Situ Preparation of Interconnected Networks Constructed by Flexible Graphene/Sn Sandwich Nanosheets for High-Performance Lithium-Ion Battery Anode

Jian Qin<sup>a</sup>, Xiang Zhang<sup>a</sup>, Naiqin Zhao<sup>a, b</sup>, Chunsheng Shi<sup>a</sup>, En-Zuo Liu<sup>a</sup>, Jiajun Li<sup>a</sup>, and

Chunnian He \*<sup>, a, b</sup>

<sup>a</sup> School of Materials Science and Engineering and Tianjin Key Laboratory of Composites and

Functional Materials, Tianjin University, Tianjin +86 300072, China

<sup>b</sup> Collaborative Innovation Center of Chemical Science and Engineering, Tianjin 300072, China

\* Corresponding author.

E-mail addresses: cnhe08@tju.edu.cn (C. He)



Figure S1. (a) and (b) SEM images of the complex of  $SnCl_2-C_6H_8O_7/NaCl$  after freeze-drying process, suggesting that the NaCl particles uniformly coated with a thin layer of  $SnCl_2-C_6H_8O_7$  complex were self-assembled into 3D structure during the freeze-drying process. (c) and (d) SEM images of the C-SnO<sub>2</sub> products before eliminating the NaCl, showing that the 3D self-assembly was well preserved after low-temperature calcination of the 3D  $SnCl_2-C_6H_8O_7/NaCl$  and the 3D C-SnO<sub>2</sub> were actually formed on the surface of 3D NaCl self-assembly.



Figure S2. XRD patterns of (a) 3D SnO<sub>2</sub>/C/SnO<sub>2</sub>, (b) 3D SnO<sub>2</sub>/C/SnO<sub>2</sub>@C,

and (c) 3D G/Sn/G networks.



Figure S3. TGA of 3D SnO<sub>2</sub>/C/SnO<sub>2</sub>, 3D SnO<sub>2</sub>/C/SnO<sub>2</sub>@C, and 3D G/Sn/G networks. The Sn contents in the 3D G/Sn/G networks estimated from the thermal analysis are ca. 46.9 wt%. (The sample is annealed under air to oxidize Sn to SnO<sub>2</sub> and carbon to CO<sub>2</sub>. On the basis of the final weight of SnO<sub>2</sub>, the original content of Sn is calculated to be 46.9 wt %. ) Comparing the weight loss of 3D SnO<sub>2</sub>/C/SnO<sub>2</sub> with that of 3D SnO<sub>2</sub>/C/SnO<sub>2</sub>@C, it can be calculated that approximately 10.8 wt% carbon is added during the hydrothermal process.



Figure S4. HRTEM images of 3D G/Sn/G networks.



Figure S5. (a) STEM image of 3D G/Sn/G networks. (b) Carbon and (c) Sn element mapping images from area 1 in (a). d) SAED pattern of a typical Sn nanoparticle. e) EDX pattern from area 2 in (a).



Figure S6. (a) XRD pattern and (b) TGA of G/Sn sandwich nanosheets, showing that the Sn content is about 52 wt.%. (c) SEM and (d) TEM images of G/Sn sandwich nanosheets, indicating a sandwich-like structure with graphene layers closely binding to both sides of metallic Sn nanoparticles or nanosheets.



Figure S7. Raman spectra of (a) 3D G/Sn/G networks and (b) G/Sn sandwich nanosheets.



Figure S8. (a) Nitrogen adsorption–desorption isotherms of 3D G/Sn/G networks and G/Sn sandwich nanosheets, and (b) pore size distribution curve of 3D G/Sn/G networks.



Figure S9. Galvanostatic discharge-charge profiles of G/Sn sandwich nanosheets at a current density of 0.2 A  $g^{-1}$ .



Figure S10. Randles equivalent circuit for 3D G/Sn/G and G/Sn electrode/electrolyte interface.  $R_s$  is the electrolyte resistance and the resistance of the surface film formed on the electrodes.  $R_{ct}$  is charge-transfer resistance,  $Z_w$  is the Warburg impedance related to the diffusion of lithium ions into the bulk electrodes and CPE represents the constant phase element.

Step number	Number of cycle	Charge/discharge rate (C, $1C = 1 \text{ A}$ $g^{-1}$ )	Average reversible capacity of 3D G/Sn/G networks electrode (mAh g <sup>-1</sup> )	Average reversible capacity of G/Sn sandwich nanosheets electrode (mAh g <sup>-1</sup> )
1	20	0.2	1011	535
2	40	0.5	871	452
3	60	1	757	385
4	80	2	695	285
5	100	5	438	123
6	120	10	241	64
7	140	0.2	1098	541

Table S1. Comparison of rate performance of 3D G/Sn/G networks electrode with G/Sn sandwich nanosheets electrode on the basis of Figure 4d.

Table S2. Comparison of specific capacity and capacity retention at different rates for 3D G/Sn/G networks electrode with those of graphene/Sn sandwich nanosheets, surface-decorated graphene/Sn composites, and various Sn/C composites anodes reported.

Materials	Voltage range (V)	Current density (A g <sup>-1</sup> )	Cycle number	Specific capacity (mAh g <sup>-1</sup> )	Capacity retention (%)
3D G/Sn/G networks	0.005.2.0	2	500	665	95.5
[this work]	0.005-3.0	0.2	100	1010	97
Graphene/Sn sandwich nanosheets [S1]	0.005–2.0	0.05	60	590	69
Graphene/Sn sandwich nanosheets [S2]	0.002-3.0	1	30	501	86
	0.01.2.0	0.5	100	684.5	76
Graphene/Sn		1	100	639.7	71
	0.01-3.0	2	100	552.3	65
		5	100	359.7	51
graphene/Sn composite <sup>[S16]</sup>	0.005-2.5	0.2	50	255	20
graphene/Sn composite <sup>[S17]</sup>	0.01-3.0	0.05	30	490	55
graphene/Sn composite <sup>[S19]</sup>	0.01–3.0	0.5	200	552	87

graphene/Sn composite <sup>[S20]</sup>	0.02-1.2	0.1	100	500	80
Sn@CNT/graphene	0.005-3.0	2	100	560	82
composite <sup>[S3]</sup>		1	100	650	78.5
Sn@C/graphene composite <sup>[S4]</sup>	0.005–2.5	0.05	50	630	70
Sn@C/graphene composite <sup>[S12]</sup>	0.01–3.0	0.75	100	566	53
Sn/C composite [S6]	0.02-3.0	0.2	130	680	94
Sn/C composite [S7]	0.02-3.0	0.025	300	638	93
Sn/C composite [S15]	0.01-1.5	0.1	100	450	93
Sn/C composite [S18]	0.01-3.0	0.2	200	722	92
Sn@C@carbon fiber	0.01–3.0	0.25	200	737	88
Sn@mesoporous carbon nanowires [S9]	0.005–2.0	0.1	50	710	57
Sn@mesoporous carbon <sup>[S10]</sup>	0.005-2.0	0.05	100	560	86
Sn@carbon nanotubes <sup>[S11]</sup>	0.01–2.5	0.1	100	437	36
Sn@carbon nanotubes <sup>[S13]</sup>	0.01–2.5	1	100	335	62
	0.005–2.0	0.1	100	525	55
Sn@C nanowires <sup>[S14]</sup>		0.5	100	490	44
		1	100	486	39
		3	100	286	32
Tin-carbon/silica composite <sup>[85]</sup>	0.0–2.5	0.3	100	440	95
Sn@CNT composite [S22]	0.005-3	0.1	80	473.9	90.1
SnQDs@N-CNF	0.02-3	0.2	500	685	74
composite <sup>[S23]</sup>		0.4	200	508	67
Sn-Co-CNT	0.005-3	0.099	200	811	91.1
composite <sup>[S24]</sup>		0.99	200	612	85.7
$TiO_2 - Sn/C$	0.01-3	0.335	160	459	99.9
nanowire <sup>[825]</sup>		3.35	100	150	84.8
SnNPs @CNTs composite <sup>[S26]</sup>	0.005-2	0.1	140	648	83.7
SnNPs@h-CS composite <sup>[S27]</sup>	0.005-3	0.166	100	550	68.75
Sn NP/CNF hybrid network	0.01-3	0.2	200	460	57.93

composite <sup>[S28]</sup>					
Sn-PCNF composite <sup>[S29]</sup>	0.01-3	0.8	200	774	93
RGO/Sn composite <sup>[S30]</sup>	0.01-2	0.099	50	858	97
Sn/graphene nanocomposite <sup>[S31]</sup>	0.01-3	0.055	100	508	63.9

Table S3. Kinetic parameters of the electrodes of 3D G/Sn/G networks and G/Sn sandwich nanosheets after the rate capability test.

Samples	$R_{ m f}(\Omega)$	$R_{ m ct}(\Omega)$
3D G/Sn/G	27.79	66.03
G/Sn	34.91	141.7

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