

## Supporting Information

### Compact Si/C Anodes Fabricated by Simultaneously Regulating the Size and Oxidation Degree of Si for Li-Ion Batteries

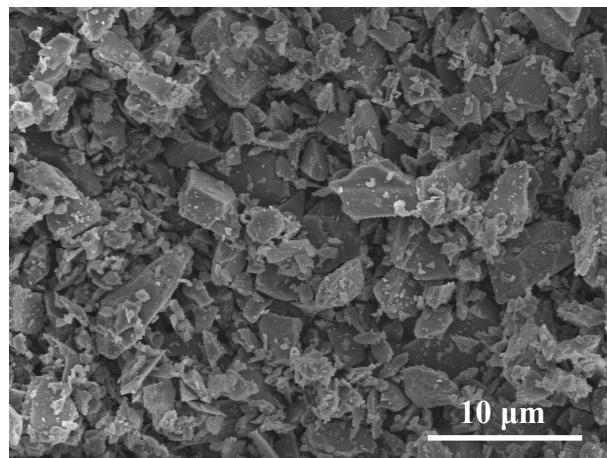
Liang Zhao <sup>a, b</sup>, Yan-Bing He <sup>a,\*</sup>, Chengfei Li <sup>a</sup>, Kelin Jiang <sup>a</sup>, Peng Wang <sup>a</sup>, Jiabin Ma <sup>a</sup>, Heyi Xia <sup>a, b</sup>, Fangyuan Chen <sup>a</sup>, Yuanbiao He <sup>a</sup>, Zhen Chen <sup>a</sup>, Conghui You <sup>a</sup>, and Feiyu Kang <sup>a, b,\*</sup>

<sup>a</sup>Shenzhen Geim Graphene Center, Tsinghua Shenzhen International Graduate School, Tsinghua University, Shenzhen 518055, PR China.

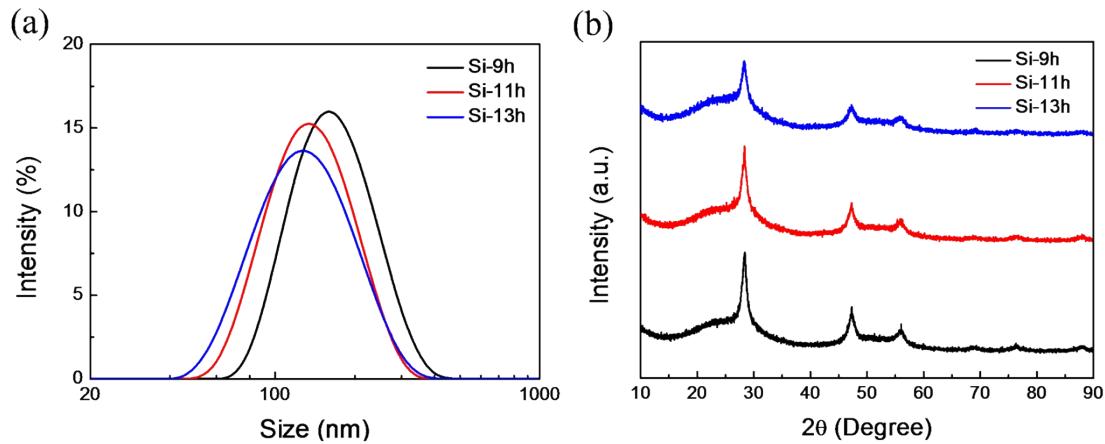
<sup>b</sup>Laboratory of Advanced Materials, Department of Materials Science and Engineering, Tsinghua University, Beijing 100084, PR China.

\* Corresponding author. E-mail address: [he.yanbing@sz.tsinghua.edu.cn](mailto:he.yanbing@sz.tsinghua.edu.cn) (Y.-B. He)

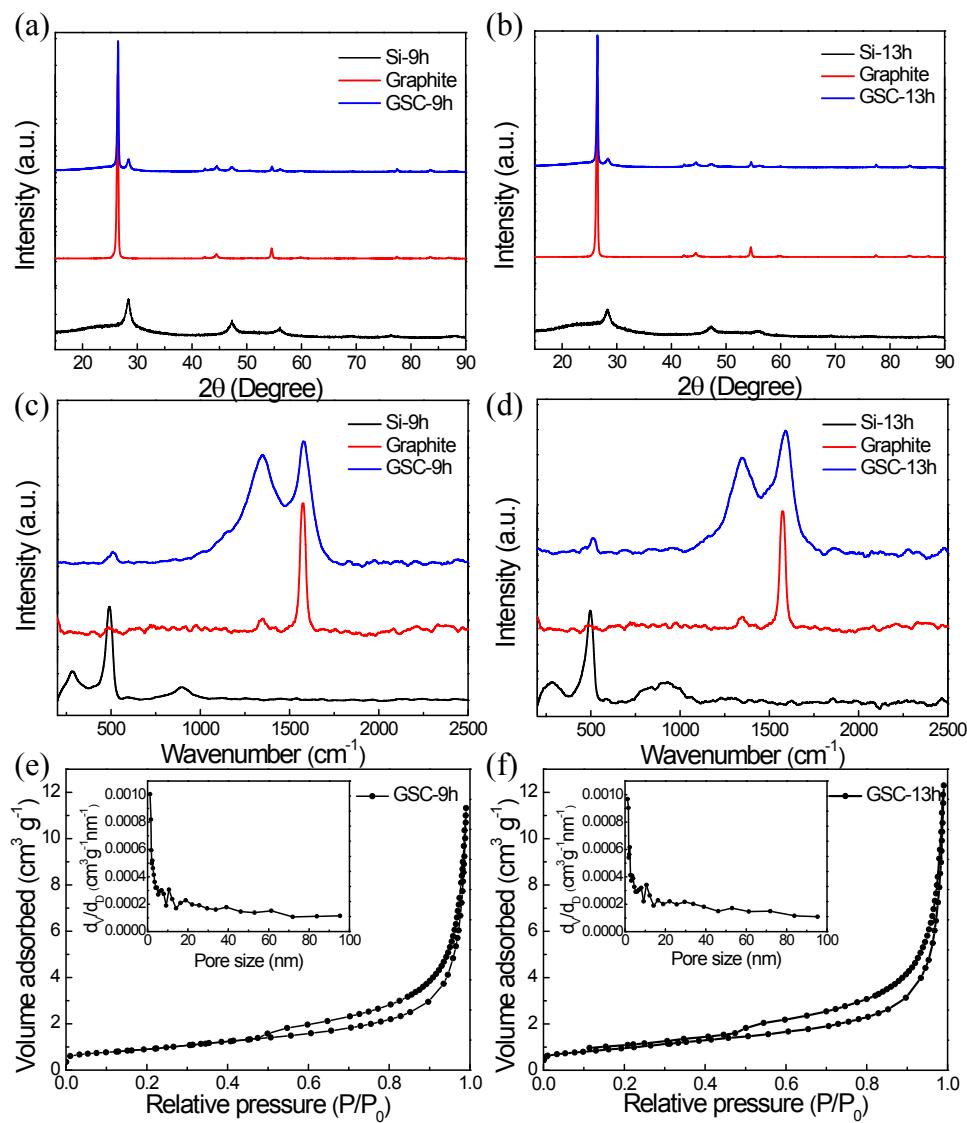
[fykang@mail.tsinghua.edu.cn](mailto:fykang@mail.tsinghua.edu.cn) (F. Y. Kang)



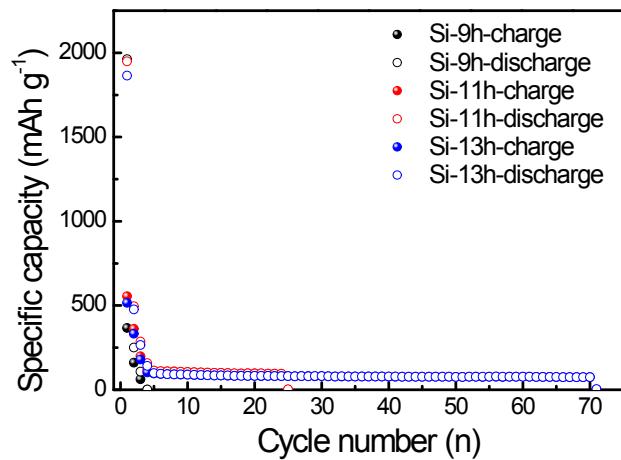
**Fig. S1** SEM image of the micro-Si particles.



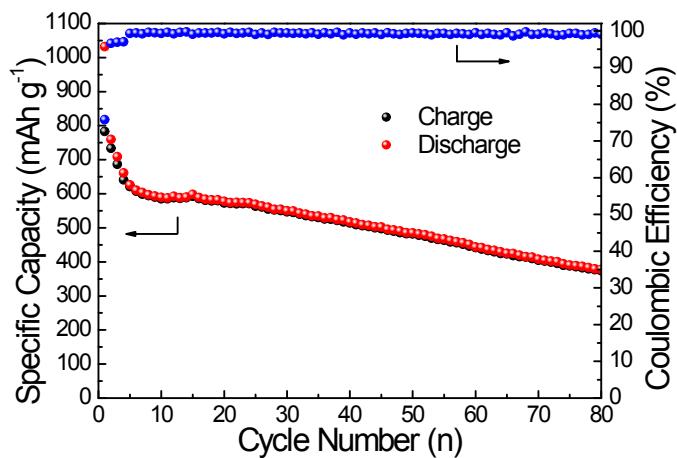
**Fig. S2** (a) The PSD curves and (b) XRD patterns of Si-9h, Si-11h, and Si-13h.



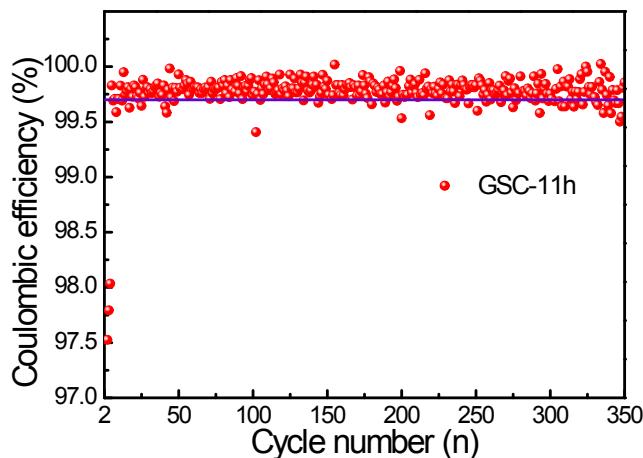
**Fig. S3** (a, b) XRD patterns, (c, d) Raman spectra, and (e, f) BET plots of GSC-9h (a, c, e) and GSC-13h (b, d, f).



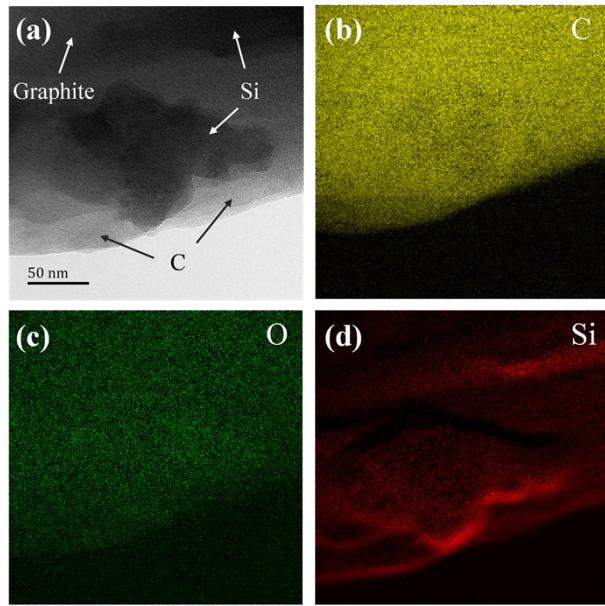
**Fig. S4** Cycling performance of Si-9h, Si-11h, and Si-13h.



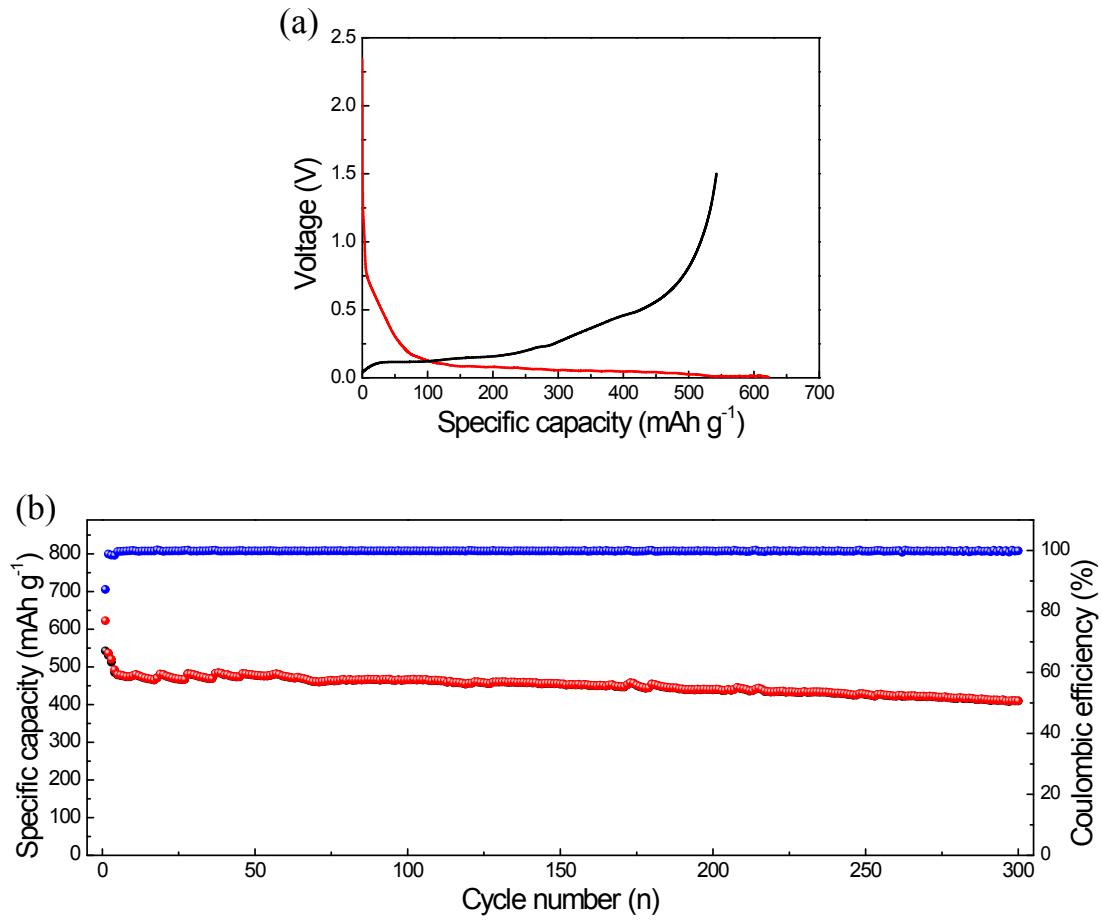
**Fig. S5** Cycling performance of SC-11h.



**Fig. S6** Coulombic efficiency of GSC-11h from 2<sup>nd</sup> to 350<sup>th</sup> cycle.



**Fig. S7** (a) TEM and (c-d) EF-TEM images of the GSC-11h after 200 cycles.



**Fig. S8** (a) The initial charge and discharge curves and (b) cycling performance of the GSC-graphite anode.



**Fig. S9** The optical photograph of  $\text{LiNi}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3}\text{O}_2/\text{GSC-graphite}$  full batteries.

**Table S1** The proportion of different valence states of Si in GSC-9h, GSC-11h, and GSC-13h according to the XPS results in Figure 3e.

	Si <sup>0</sup>	Si <sup>1+</sup>	Si <sup>2+</sup>	Si <sup>3+</sup>	Si <sup>4+</sup>
GSC-9h	17.6%	4.64%	31.03%	25.28%	21.45%
GSC-11h	14.46%	5.37%	29.02%	31.34%	19.82%
GSC-13h	11.99%	8.44%	29.12%	36.23%	14.23%

**Table S2** The EIS simulation results of Figure 4d.

	R <sub>b</sub> [Ω]	R <sub>SEI</sub> [Ω]	R <sub>ct</sub> [Ω]	R <sub>t</sub> [Ω]
GSC-9h	2.92	4.05	10.71	17.68
GSC-11h	4.11	3.75	8.08	15.94
GSC-13h	4.38	2.48	15.37	22.23

**Table S3** The EIS simulation results of Figure 6d.

	R <sub>b</sub> [mΩ]	R <sub>SEI</sub> [mΩ]	R <sub>ct</sub> [mΩ]	R <sub>t</sub> [mΩ]
1st	43.76	3.53	58.00	105.29
50th	46.20	12.01	10.83	69.04