

Supporting Information

Carbon-coated mesoporous silicon microsphere anodes with greatly reduced volume expansion

Zheng-Long Xu, Yang Gang, Mohammad Akbari Garakani, Sara Abouali,

Jian-Qiu Huang and Jang-Kyo Kim*

Department of Mechanical and Aerospace Engineering, The Hong Kong University of
Science and Technology, Clear Water Bay, Kowloon, Hong Kong

Supplementary figures

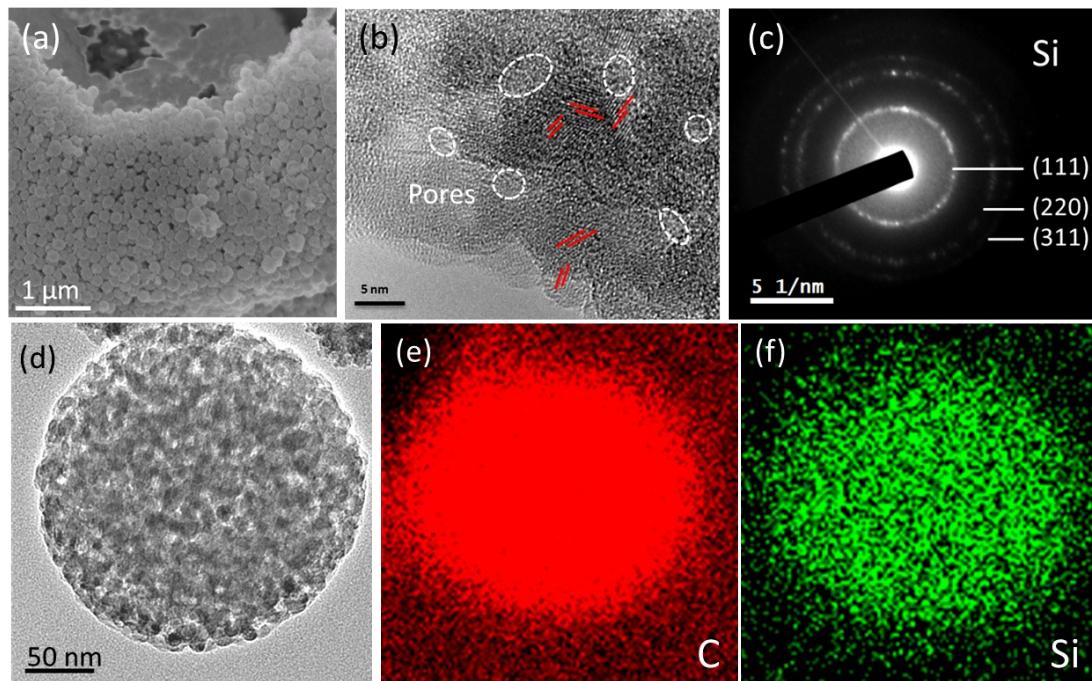


Fig. S1. (a) Low magnification SEM image of porous Si; (b) HRTEM image of porous Si showing interconnected Si nanocrystals and surrounding pores; (c) SAED pattern of porous Si; (d-f) EDS mapping of mesoporous Si/C sphere.

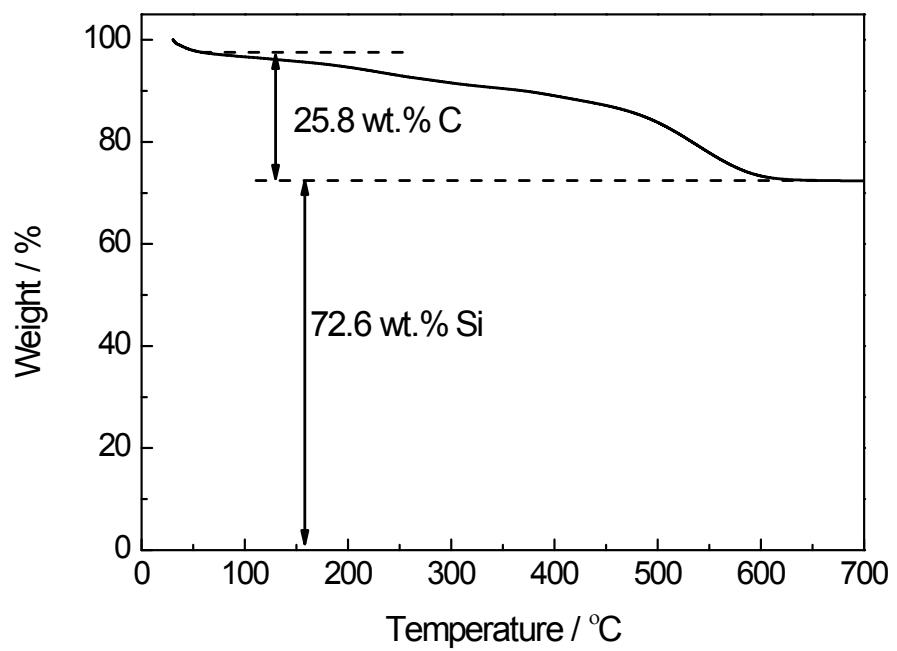


Fig. S2. TGA curve of mesoporous Si/C microspheres.

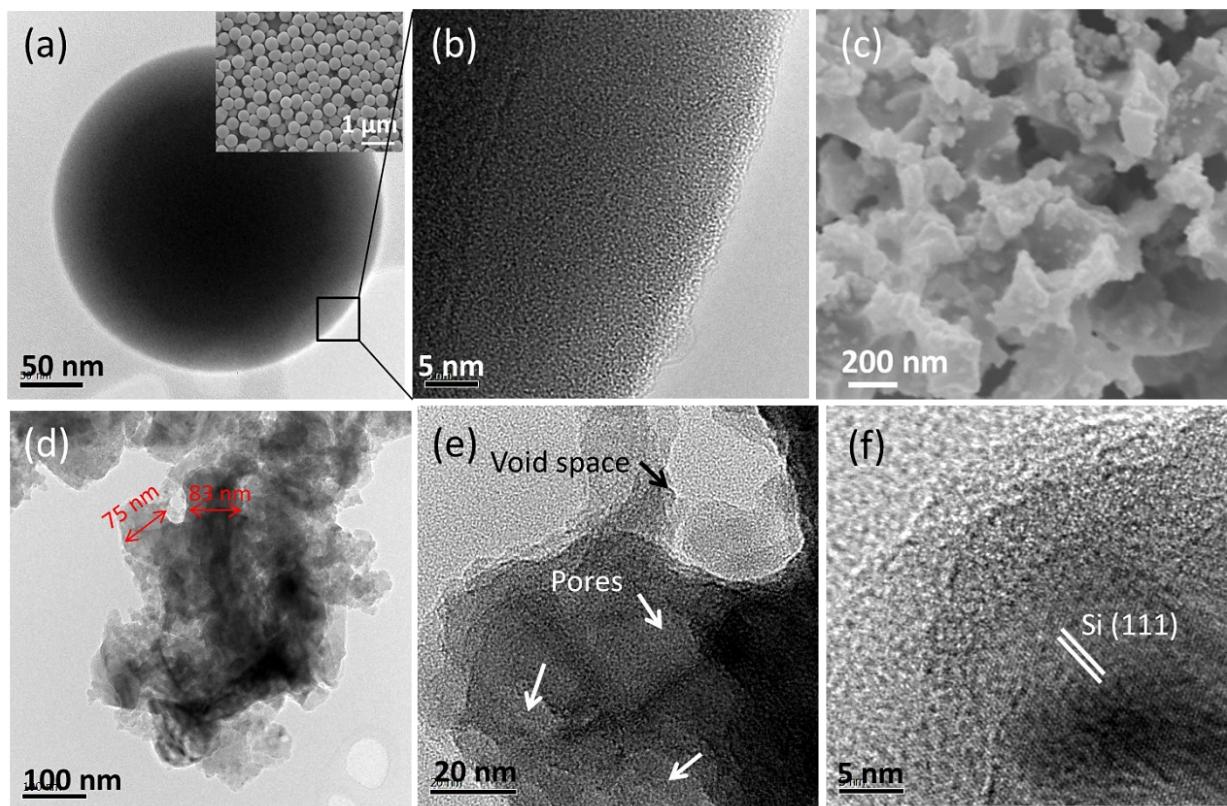


Fig. S3. (a-b) HRTEM images of solid silica particles and their SEM image in inset (a); (c) SEM image and (d-f) TEM images of macroporous Si prepared from solid silica.

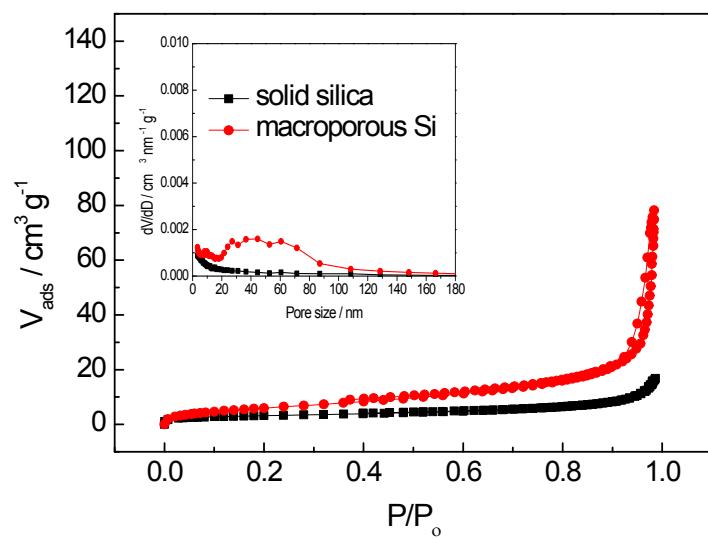


Fig. S4. N₂ adsorption/desorption isotherms and BJH pore size distributions (inset) of solid silica and macroporous Si.

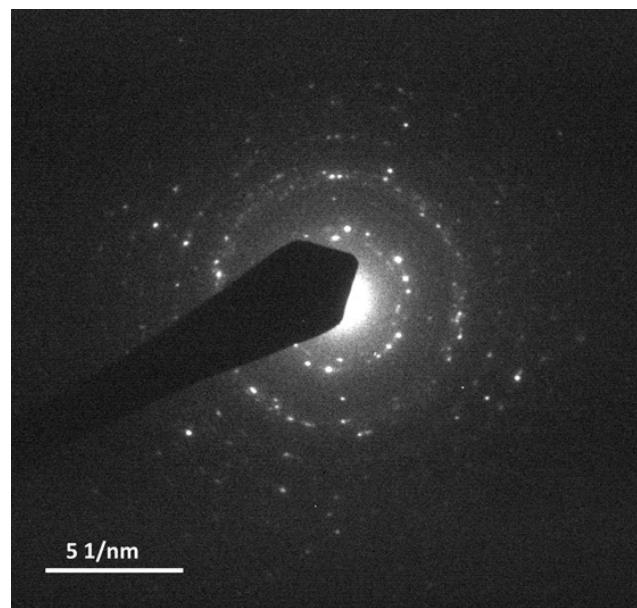


Fig. S5. SAED pattern of crystalline Li₁₅Si₄ in Fig. 4e.

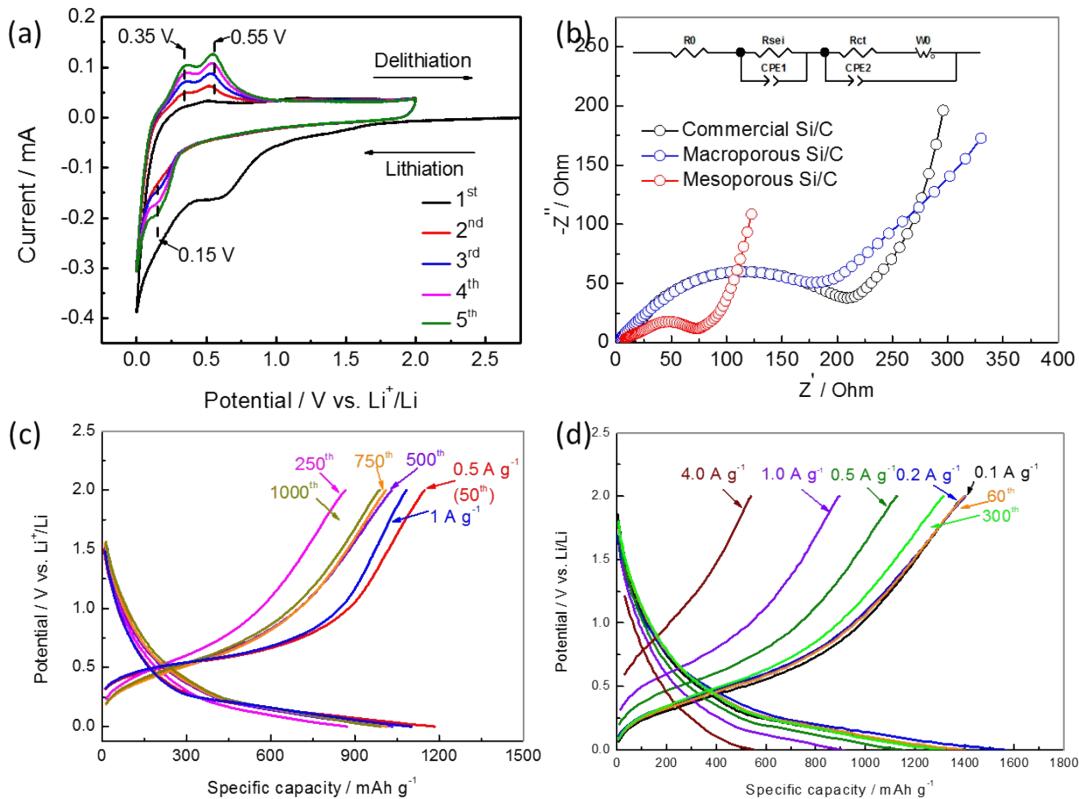


Fig. S6. (a) CV curves of mesoporous Si/C; (b) Nyquist plots of three different electrodes after cycles (see Fig. 6b) and equivalent circuit model (inset); (c-d) potential/capacity profiles of the mesoporous Si/C electrodes measured at different cycles (see Fig. 6c) and different current densities (see Fig. 6d).

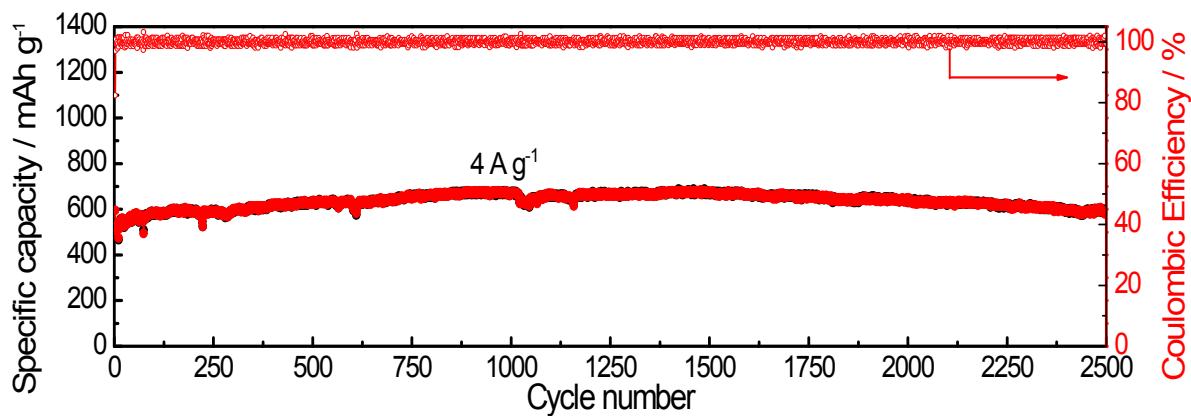


Fig. S7. Exceptional cyclic stability of mesoporous Si/C electrodes measured at 4 A g⁻¹ for 2500 cycles.

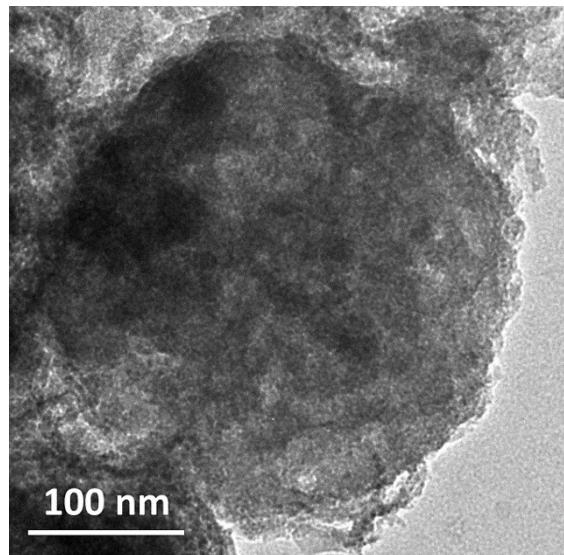


Fig. S8. TEM image of mesoporous Si/C electrode taken after 500 cycles (see Fig. 6b).

Supplementary tables

Table S1 Surface area, pore volume and pore size of silica, Si and Si/C composites.

Sample	Surface area / m ² g ⁻¹	Pore volume / cm ³ g ⁻¹	Pore size / nm
Porous silica	738	0.74	~3.6
Porous Si	544	0.56	~7, ~1.9
Mesoporous Si/C	257	0.47	~5
Solid silica	11	0.024	/
Macroporous Si	92	0.16	~60

Table S2 Comparison of volume expansion between the current mesoporous Si/C electrodes

and the representative Si/C composites measured by *in-situ* TEM.

Structure	Initial diameter /nm	Volume expansion /%	Ref.
Neat Si	~145	~330	S1
Si/C	~145	~310	S1
Si/graphene	~90	~248	S2
Si/CNT	~20	~180	S3
Mesoporous Si/C	~242	~85	This work

Table S3 Resistance parameters, R_0 , R_{sei} and R_{ct} , determined from the simulation data in Fig. S6b.

Electrodes	R_0 / Ω	R_{sei} / Ω	R_{ct} / Ω
Mesoporous Si/C	5.0	16.6	26.7
Macroporous Si/C	4.9	25.2	143.4
Commercial Si/C	5.1	86.1	107.1

Table S4 Comparison of electrochemical performance between the current mesoporous Si/C electrodes and the representative Si-based electrodes prepared by magnesiothermic reduction in terms of current density ($A g^{-1}$), cycle number, residual capacity ($mAh g^{-1}$) and capacity retention (%).

Structure	Current density $/A g^{-1}$	Cycle number	Residual capacity	Capacity retention / %	Ref.
			$/ mAh g^{-1}$		
Bubble-sheet Si/C	1	200	1018	93	17
Rice husk based Si/C	2	300	1500	86	20
Rice husk Si/rGO	1	30	1000	~64	21
Sand based Si/C	2	1000	1024	~58	22
Sand based Si/C	1	100	~1500	~60	23
Porous Si	0.05	50	1004	~30	24
Electrospun Si fiber	2	300	1363	70	25
Electrospun Si/C	1	100	547	~57	26
Porous Si	1	100	1440	~80	27
Void Si/TiO ₂	0.4	100	804	48	28
3D porous Si/C	0.8	100	1409	94	32
3D Si/graphene	0.2	200	2050	~51	33
Si/SiO ₂	1	50	900	75	34
Mesoporous Si/C	0.5	500	1199	~86	This work
	1	1000	990	~90	

Supplementary Movie:

Movie S1: The lithiation process of mesoporous Si/C microsphere. The movie is played at 10 x of the real speed.

Reference

S1 Z.L. Xu, K. Cao, A. Abouali, M. Akbari Garakani, J. Huang, J.Q. Huang, E. Kamali Heidari, H. Wang, J.K. Kim, *Energy Storage Mater.* 2016, **3**, 45.

S2 L. Luo, J. Wu, J. Luo, J. Huang, V.P. Dravid, *Sci. Rep.* 2014, **4**, 3863.

S3 W. J. Yu, C. Liu, P. X. Hou, L. Zhang, X. Y. Shan, F. Li and H. M. Cheng, *ACS Nano*, 2015, **9**, 5063.