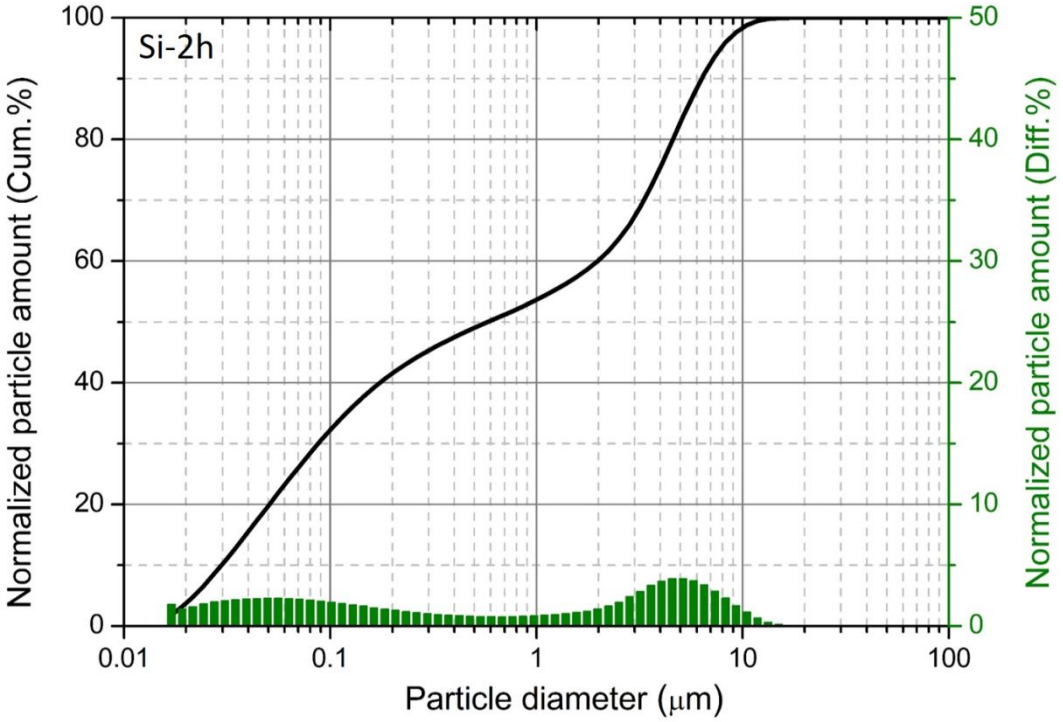
Supporting information:

Title: Electrochemical performance of ultra-thin surface oxide modulated nano-Si anode confined in graphite matrix for highly reversible lithium-ion batteries

*Maddipatla Reddyprakash, Chadrasekhar Loka, and Kee-Sun Lee**

Department of Advanced Materials Engineering, Kongju National University, Cheonan 31080, South Korea.

Corresponding author e-mail address: kslee@kongju.ac.kr (K.S. Lee)


Si-2h
Normalized particle amount (Cum.%)
Normalized particle amount (Diff.%)
Particle diameter (μm)
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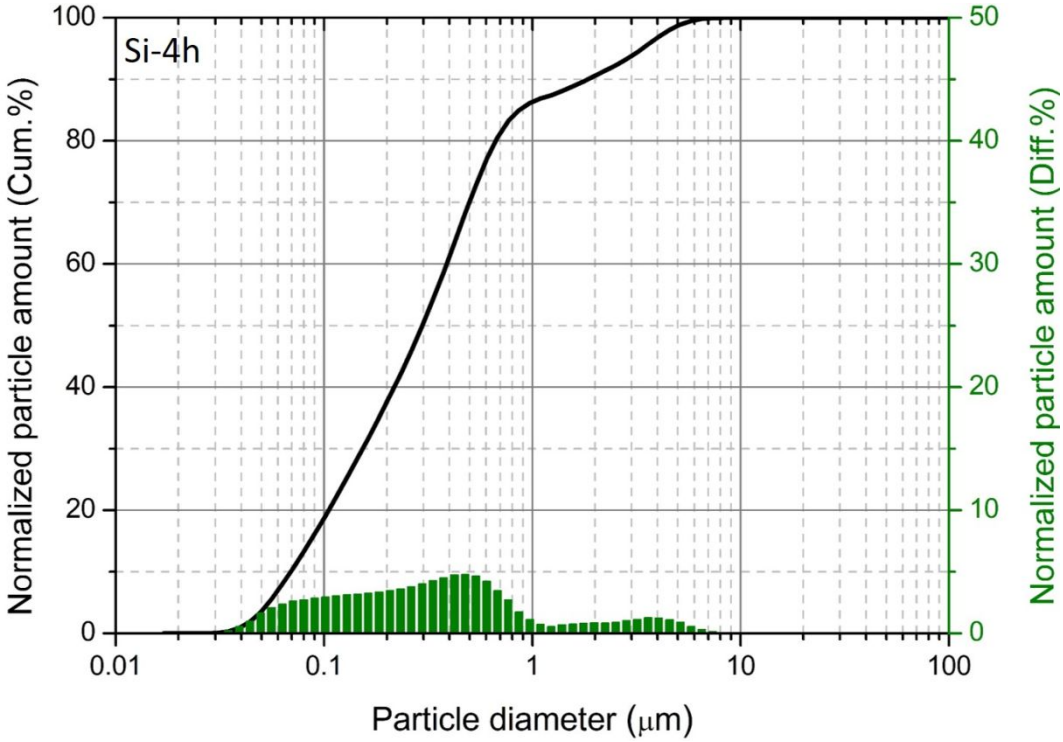
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Normalized particle amount (Diff.%)
Particle diameter (μm)
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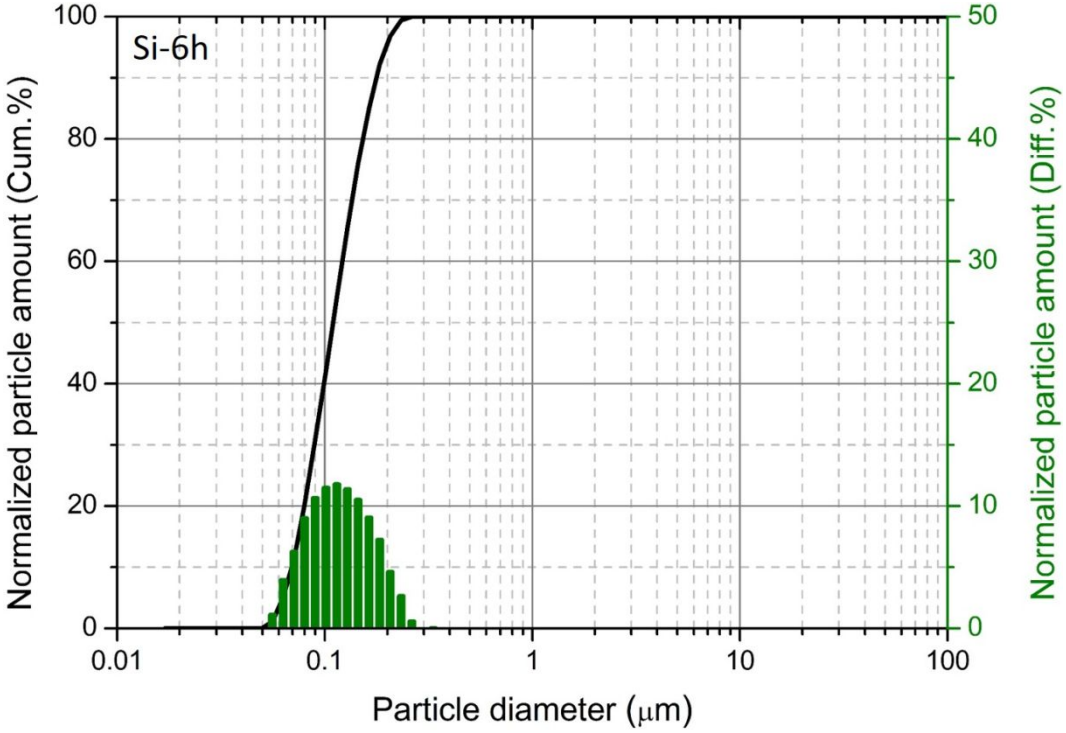

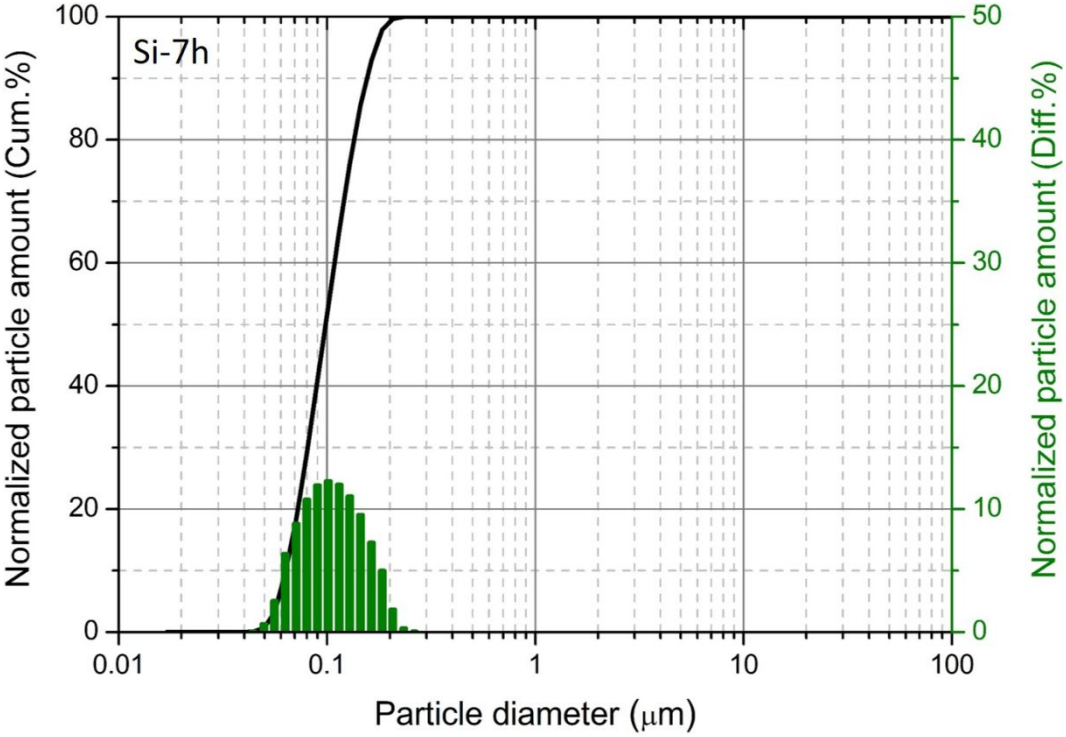

Figure S1. The particle size distribution of Si milled for 2 h, 4 h, 6 h, and 7h.

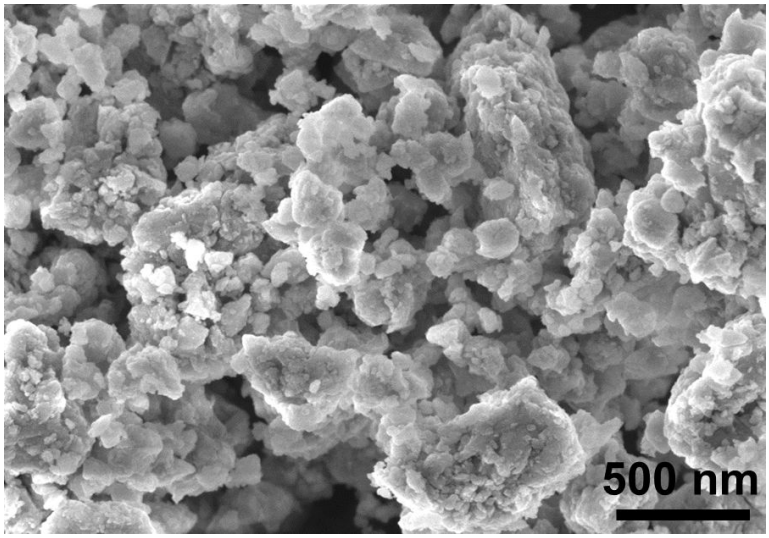

Figure S2. FE-SEM image of Si/SiO_x-2 composite.

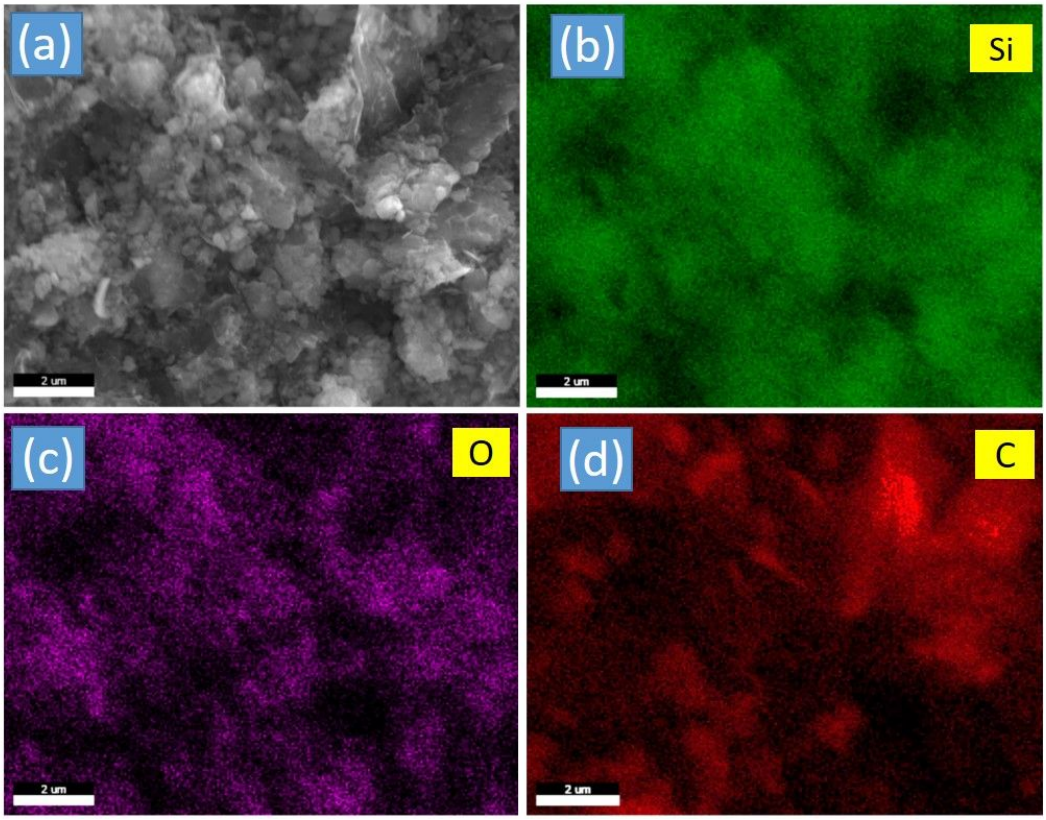

Figure S3. (a) SEM image of Si/SiO_x@C nanocomposite and (b-d) corresponding EDAX elemental mapping of silicon, oxygen, and carbon.

Table S1. The summary of XPS data of fitted Si 2p high-resolution spectra parameters for the Si/SiO_x-1, Si/SiO_x-2, and Si/SiO_x-3.

Sample	Peak identity	Binding Energy (eV)
Si/SiO_x-1	Si^{0+}	99.7
	Si^{2+}	102.57
	Si^{3+}	104.38
Si/SiO_x-2	Si^{0+}	99.75
	Si^{2+}	102.2
	Si^{3+}	104.26
Si/SiO_x-3	Si^{0+}	99.5
	Si^{3+}	103.6
	Si^{4+}	105.5

Table S2. Comparison of electrochemical performance of the various Si/SiO_x based anode materials for lithium-ion batteries.

Materials	1st Cycle Discharge Capacity (mAh g^{-1})	1st Cycle Coulombic Efficiency	Current density (mAg^{-1})	Cycle number	Discharge Capacity (mAh g^{-1})	Discharge Capacity retention	Ref.
$Si-SiO_x$ composite	2270	85%	200	70	1600	75.4%	[1]
Nano-Si/SiO_x composite	1715	60%	100	50	1445	84.2%	[2]
$Si-SiO_x$ composite	2245	58.4%	100	50	55	2.5%	[3]
Si-core-SiO_x shell nanocomposite	2030	64.41%	---	50	1622	79.92%	[4]
Si/SiO_x composite	2905	83%	420	100	1105	38%	[5]
Porous Si/SiO_2 composite	2505	70.4%	---	100	205	8.1%	[6]
Si/SiO_x nanocomposite	2759	76%	500	260	1532	76.7%	This work

Table S3. Capacities, coulombic efficiencies and capacity retention values of the Si/SiO_x, and Si/SiO_x@C nanocomposite electrodes.

Sample name	1st Discharge capacity (mAh g^{-1})	1st Charge capacity (mAh g^{-1})	1st Coulombic efficiency	Discharge capacity after 100 cycles (mAh g^{-1})	Discharge capacity retention after 100 cycles (1C-rate)
Si/SiO_x-1	2759	2100	76%	1849	88.2%
Si/SiO_x-2	2524	1643	65%	1127	70.9%
Si/SiO_x-3	2274	1419	62%	822	67.7%
Si/SiO_x@C	1748	1531	87%	1505	97.7%

Table S4. R_{SEI} and R_{ct} values of the Si/SiO_x-1 and Si/SiO_x@C nanocomposites after the 1st, 30th and 100th cycles.

Sample name	R_{SEI} (Ω)			R_{ct} (Ω)		
	1st cycle	30th cycle	100th cycle	1st cycle	30th cycle	100th cycle
Si/SiOx-1	8.6	5.44	4	49	26.5	23.3
Si/SiOx@C	5.2	3.1	1.4	34.8	16	9

References:

(1) Yoo, H.; Lee, J. I.; Kim, H.; Lee, J. P.; Cho, J.; Park, S. Helical Silicon/Silicon Oxide Core-Shell Anodes Grown onto the Surface of Bulk Silicon. *Nano Lett.* **2011**, *11* (10), 4324-4328.

(2) Hoeltgen, C.; Lee, J. E.; Jang, B. Y. Stepwise Carbon Growth on Si/SiO_x Core-Shell Nanoparticles and Its Effects on the Microstructures and Electrochemical Properties for High-Performance Lithium-Ion Battery's Anode. *Electrochim. Acta* **2016**, *222*, 535-542.

(3) Wang, J.; Zhou, M.; Tan, G.; Chen, S.; Wu, F.; Lu, J.; Amine, K. Encapsulating Micro-Nano Si/SiO_x into Conjugated Nitrogen-Doped Carbon as Binder-Free Monolithic Anodes for Advanced Lithium Ion Batteries. *Nanoscale* **2015**, *7* (17), 8023-8034.

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(6) Wang, K.; Tan, Y.; Li, P.; Xue, B.; Sun, J. Facile Synthesis of Double-Layer-Constrained Micron-Sized Porous Si/SiO_2/C Composites for Lithium-Ion Battery Anodes. *ACS Applied Materials & Interfaces* **2019**, *11* (41), 37732-37740.