Supporting Information

Hoop-Strong Nanotubes for Battery Electrodes

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I. Supporting Figures

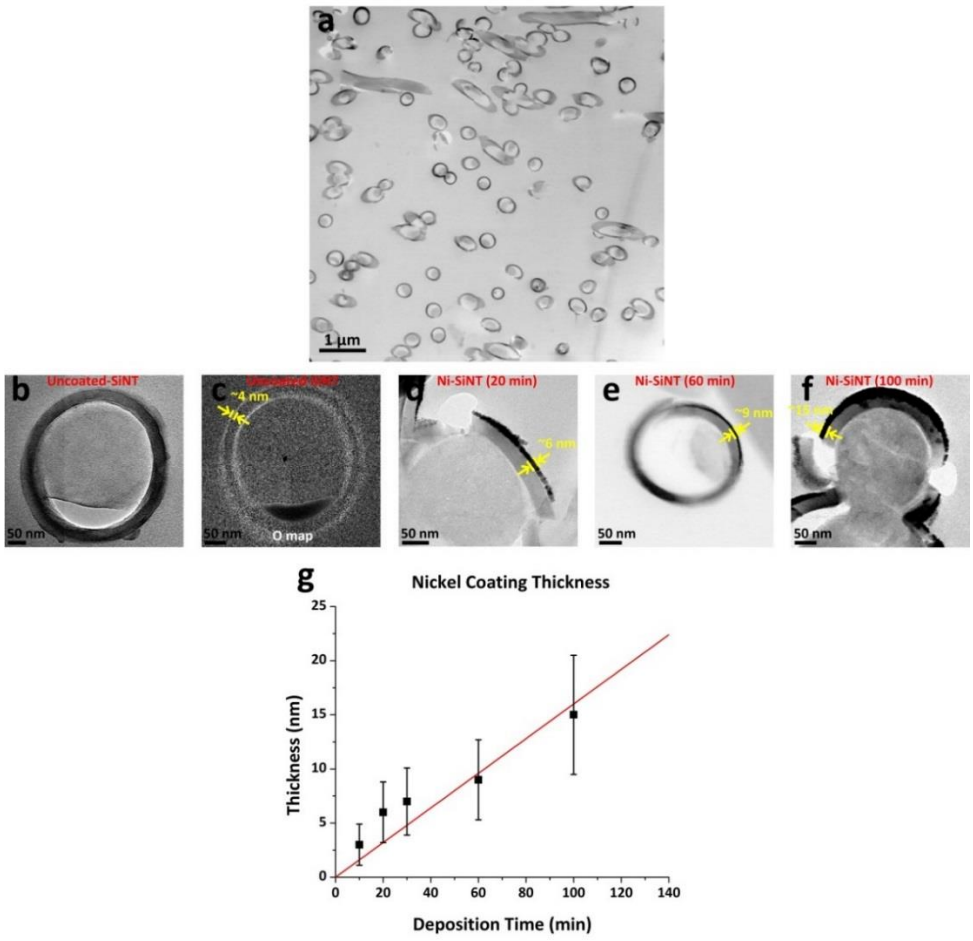


Figure S1: Surface coating thickness measurements for different SiNTs. (a) Microtomed cross-sectional Ni-coated SiNTs on a TEM Cu grid. The coating was employed using DC-magnetron sputtering at argon gas pressure of 1Pa and target power density of 4.5W/cm^2. (b) Cross-sectioned SiNT without Ni coating. (c) An EFTEM elemental mapping showing oxygen distribution in (b). (d-f) Magnified images of individual SiNTs with different Ni deposition time of 20, 60, and 100 min respectively. (g) Measured nickel coating thickness with different deposition time. To determine the thickness of Ni coatings on the surface of the SiNTs, a radial cross-section is produced by embedding the nanotubes in a cross-linked polymer and using a microtome cutting tool as shown in Figure S1a. An oxygen distribution mapping using energy-filtered transmission electron microscopy (EFTEM) is performed in a cross-sectioned SiNT without Ni coatings (Figure S1b). We judiciously determine the thickness of SiO_x surface layer to be ~4 nm (Figure S1c). The same thickness of SiO_x is also formed at the inner wall of the nanotube. For Ni coating thickness measurements, several SiNT samples with Ni deposition time of 20 min, 60 min and 100 min are selected and high-magnification TEM images are taken (Figure S1d-S1f). A thickness dependence of SiNTs is plotted with respect to different deposition time (Figure S1g). Using linear fit regression of measured Ni thickness within ±30 % error margins, we assign the thickness of Ni coatings with deposition time of 10 min, 30 min and 100 min to be ~2 nm, ~5 nm, and ~16 nm respectively. Our observations show a clean Ni-Si interface, with no indications of nickel silicide formation, which may be expected when heated above 250° C.

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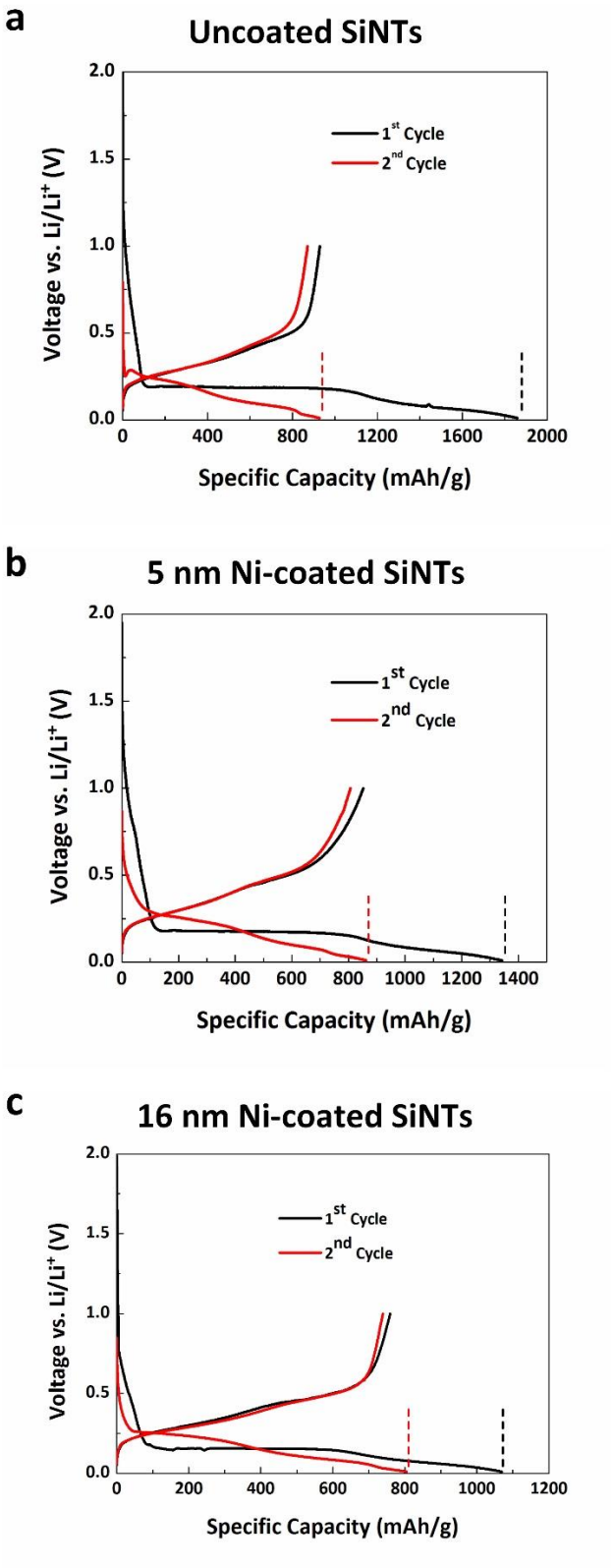


Figure S2: Galvanostatic charge/discharge profile of (a) uncoated SiNTs (b) SiNTs with ~5 nm Ni coatings (c) SiNTs with ~16 nm of Ni coatings.

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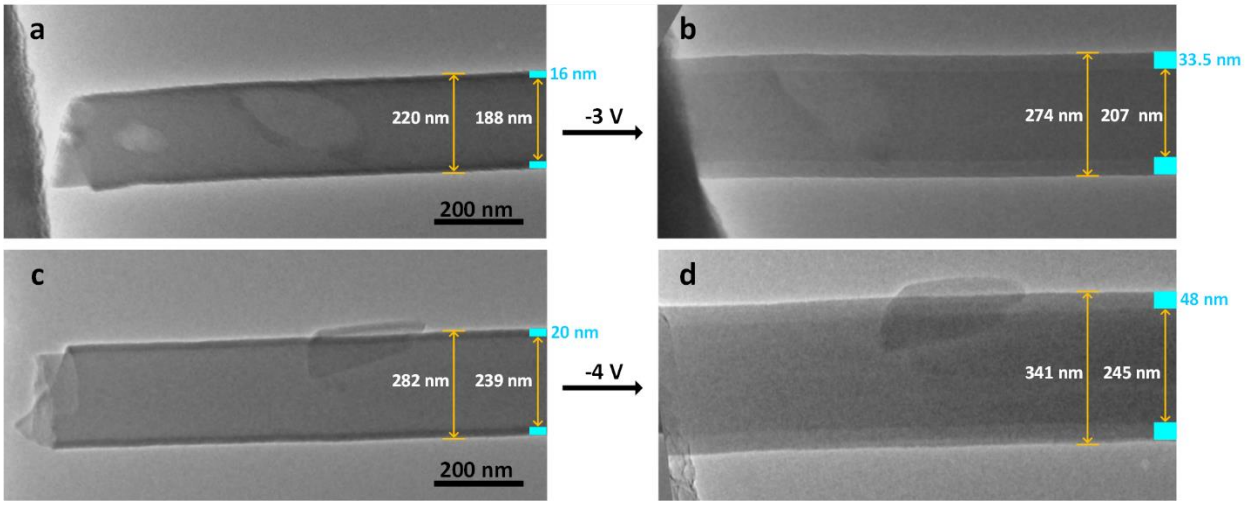


Figure S3: Lithiation of SiNT with different Si wall thickness. a) Pristine SiNT with wall thickness of 16 nm. b) Lithiation of SiNT (a) upon applying bias of -3 V with respect to Li metal. (a-b) Same scale bar. c) Pristine SiNT with wall thickness of 20 nm. d) Lithiation of SiNT (c) upon applying bias of -4 V with respect to Li metal. (c-d) Same scale bar.

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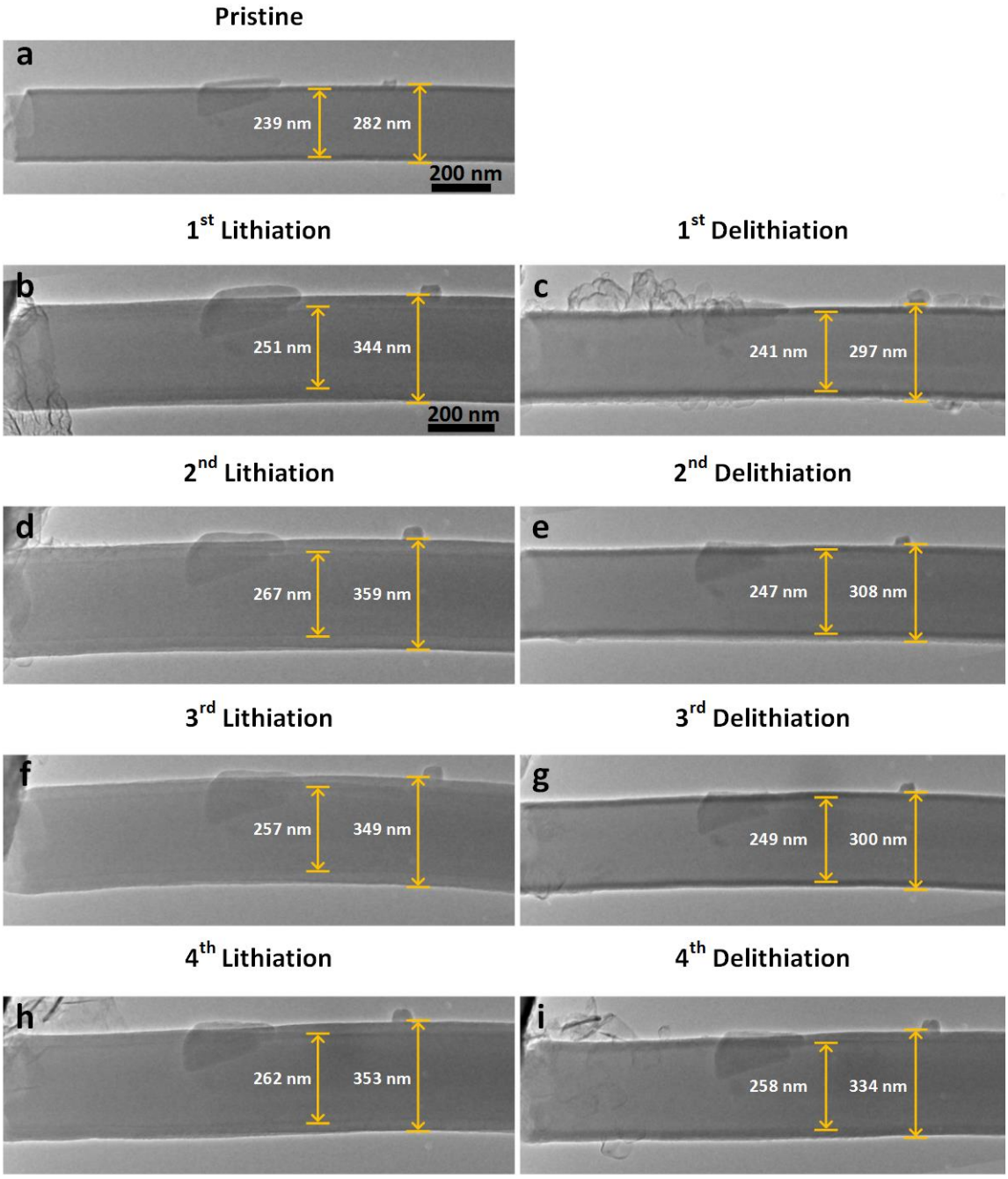


Figure S4: *In situ* TEM lithiation and delithiation of an uncoated SiNT performed for 4 consecutive cycles. Same scale bar (b-i).

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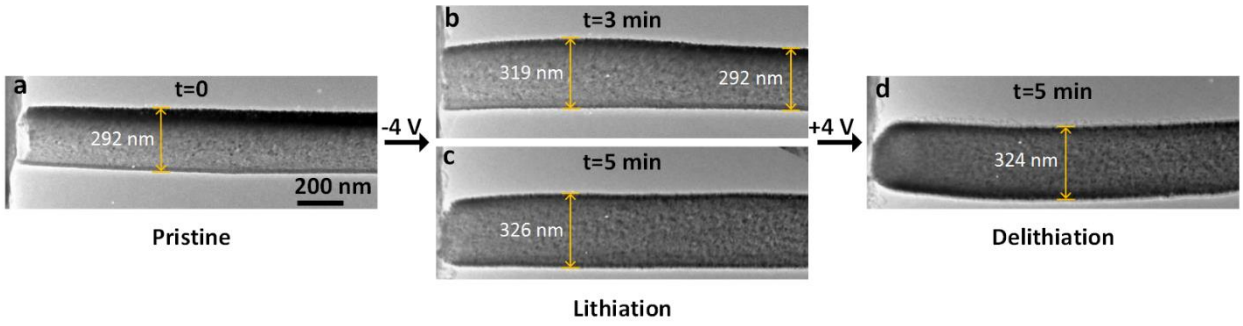


Figure S5: Lithiation and delithiation of SiNT with Ni-coating thickness of ~5 nm. Same scale bar (a-d).

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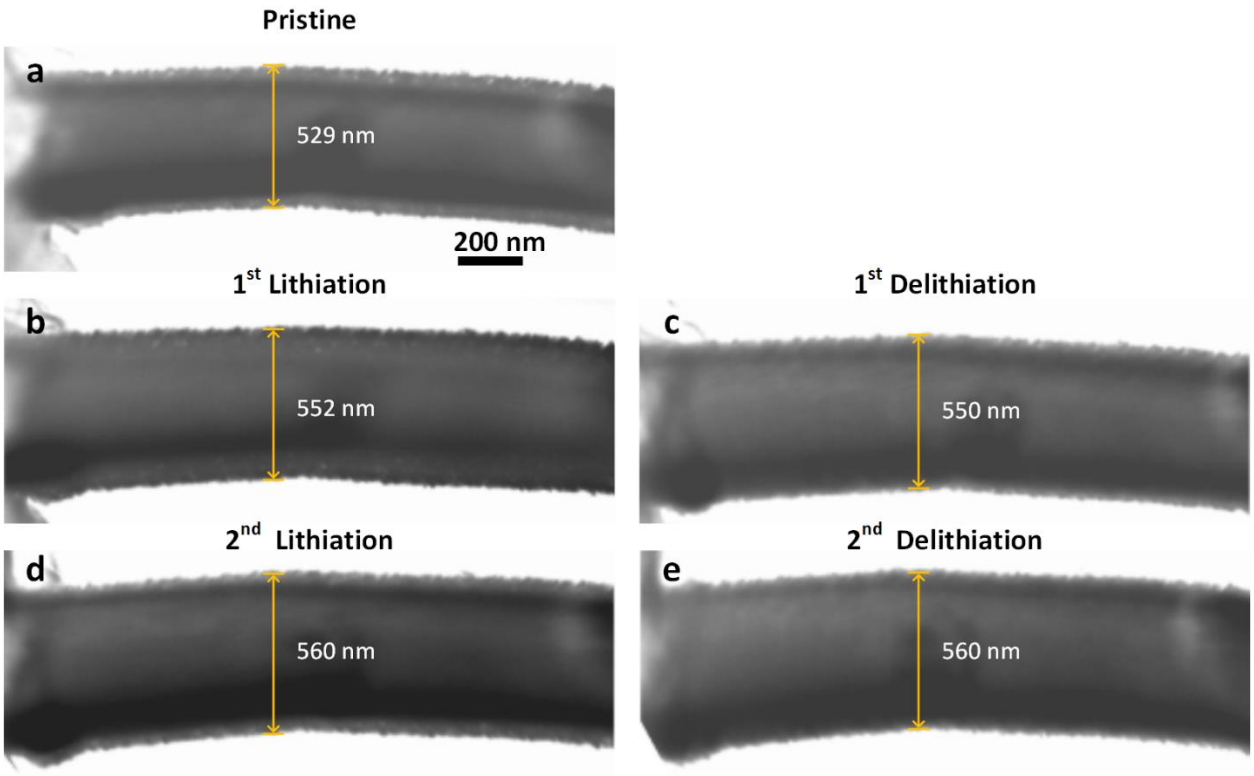


Figure S6: Lithiation and delithiation of SiNT with Ni-coating thickness of ~16 nm performed for 2 consecutive cycles. Same scale bar (a-e).

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II. Supporting Table

	Outer Diameter (nm)		Inner Diameter (nm)	
	Before Lithiation	**After Lithiation**	**Before Lithiation**	**After Lithiation**
Uncoated-SiNTs				
# 1.	269	365	233	254
# 2.	253	333	213	229
# 3.	285	360	231	242
~2 nm Ni-Coated SiNTs				
# 1.	324	380	263	245
# 2.	280	346	227	212
# 3.	271	327	217	199
~5 nm Ni-Coated SiNTs				
# 1.	263	290	218	203
# 2.	284	311	246	229
# 3.	300	324	253	242
~16 nm Ni-Coated SiNTs				
# 1.	672	678	570	521
# 2.	605	606	406	329
# 3.	526	527	400	330

Table S1: Measured outer and inner diameters of uncoated-SiNTs and Ni-coated SiNTs with different thicknesses before and after lithiation.

Note: The selection of ~16 nm Ni-coated SiNTs with larger diameters is motivated to achieve better contrast for the interior of the SiNT by TEM. SiNTs with smaller diameters presents certain unfortunate ambiguities in resolving the inner diameter owing to the relatively thicker Ni coatings, the low scattering density of lithium alloys, and the issue of electron transparency in such nanotubes in the TEM (operating at 200kV). The different sizes of the SiNTs used in our study have little or no effect on the nature of volume expansion during lithiation, which agrees with the coin-cell studies on coated SiNT shown in Figure 1.

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II. Supporting Movie

Movie M1: Lithiation behavior of an uncoated SiNT showing outer volume expansion of Si walls. The video is sped up four times from the real-time.

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