Supplementary Information

Nickel-stabilized Si/Ni/Si/Ni multi-layer thin-film anode for long-cycling-life lithium-ion battery

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Table S1. Comparison of Si- and Sn-based material [1].

	Specific capacity (mAh/g)	Volume expansion (%)	Potential vs. Li/Li^+ (V)
Si	3579~4200	300~400	0.4
Sn	994	260	0.6

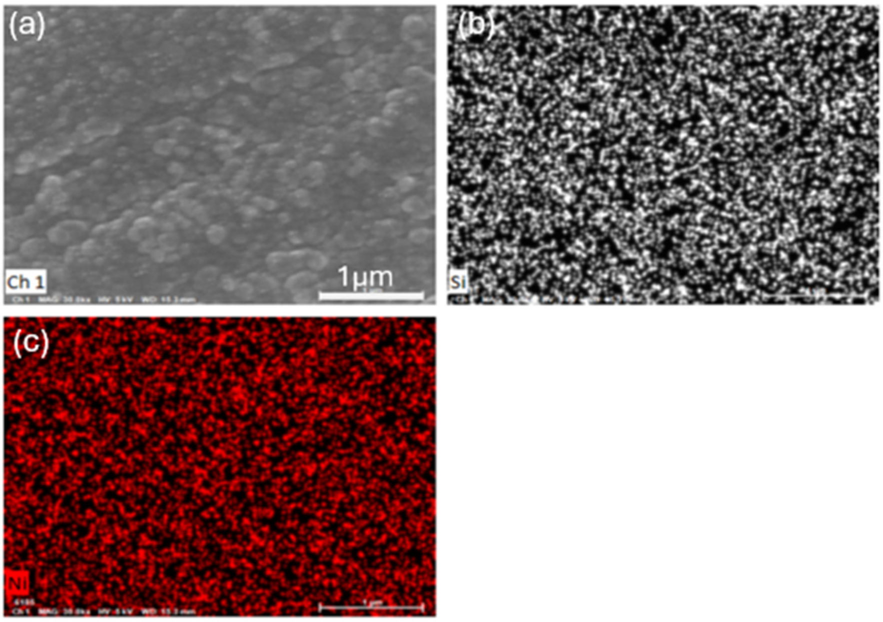


Figure S1. (a) SEM image, and EDS mapping images of (b) element Si and (c) element Ni of a Si (100nm)/Ni (20nm)/Si (100nm)/Ni (20nm) anode.

Table S2. Peak values of CV curves shown in Fig. 4.

Sample/Spot	A	B	C	D
Si thin-film anode	1^{st}:0.321 V 2^{nd}:0.329 V 3^{rd}:0.332 V	1^{st}:0.519 V 2^{nd}:0.495 V 3^{rd}:0.502 V	1^{st}:0.019 V 2^{nd}:0.028 V 3^{rd}:0.040 V	1^{st}:0.189 V 2^{nd}:0.215 V 3^{rd}:0.225 V
Si/Ni/Si/Ni thin-film anode	1^{st}:0.322 V 2^{nd}:0.340 V 3^{rd}:0.340 V	1^{st}:0.522 V 2^{nd}:0.523 V 3^{rd}:0.523 V	1^{st}:0.018 V 2^{nd}:0.027 V 3^{rd}:0.027 V	1^{st}:0.186 V 2^{nd}:0.186 V 3^{rd}:0.175 V

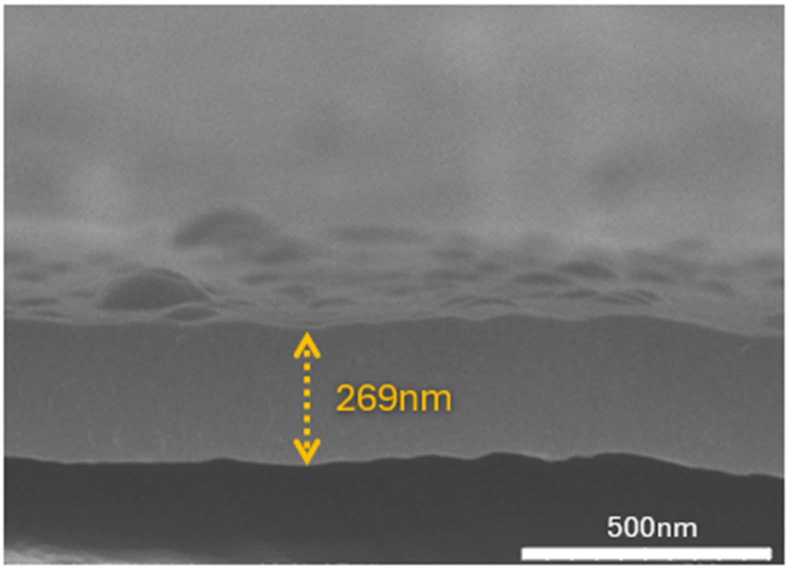
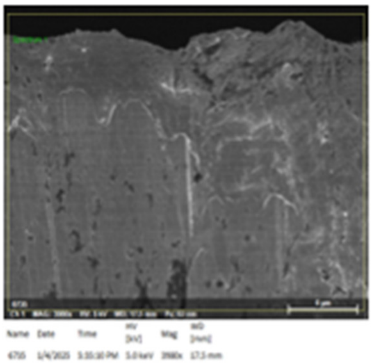
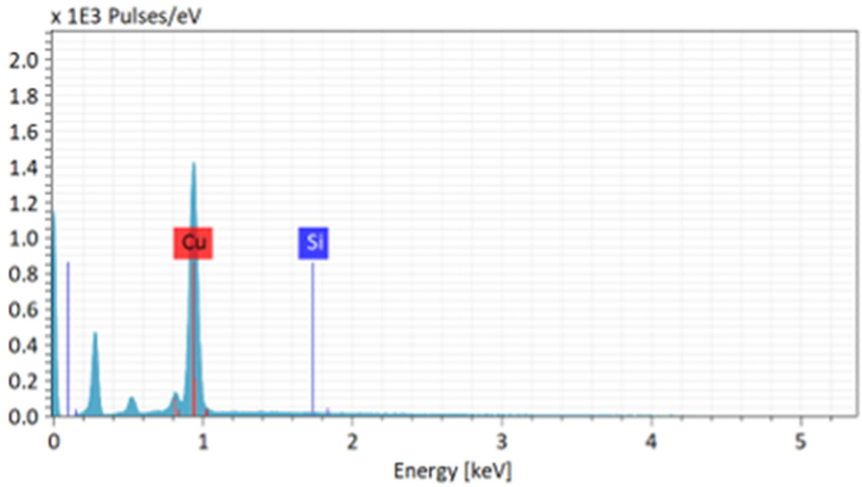


Figure S2. Cross-sectional SEM image of Si (200nm) anode before cycling.





Spectrum	Si	Cu	Sum
Spectrum 1	0.02	99.98	100.00

Figure S3. A cross-sectional SEM image (upper) and an EDS spectrum (lower) showing elemental contents of a Si (200nm) anode, of which the active layer has broken apart from the current collector after 100 cycles of charge–discharge under 1mA/cm^2.

As depicted in Figure S3, after 100 cycles of charge–discharge, the cross-sectional image of a Si anode contains only the copper collector, and no Si content was detected. The Si active layer pulverized or was detached from the copper collector.

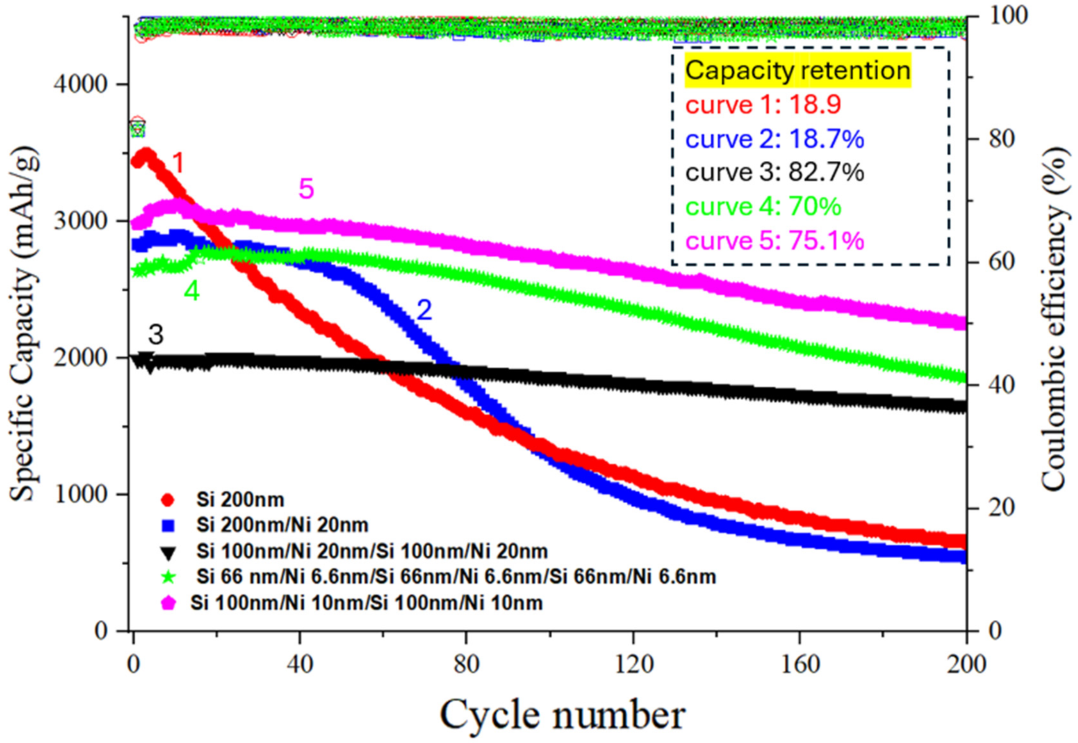


Figure S4. Cyclic performance of Si (200nm), Si (200nm)/Ni (20nm), Si (100nm)/Ni (20nm)/Si (100nm)/Ni (20nm), Si (66nm)/Ni (6.6nm)/Si (66nm)/Ni (6.6nm)/Si (66nm)/Ni (6.6nm), and Si (100nm)/Ni (10nm)/Si (100nm)/Ni (10nm) under 0.05mA/cm2 test current.

Figure S4 presents results for cells with different electrode structures or thicknesses. After 200 cycles, the multilayer Si/Ni/Si/Ni structure (curves 3 and 5) exhibited superior capacity retention to the other configurations. This is further evidenced by the fact that, under identical mass loading of the active material (curves 2, 4, and 5), the Si/Ni/Si/Ni structure consistently demonstrated the best performance.

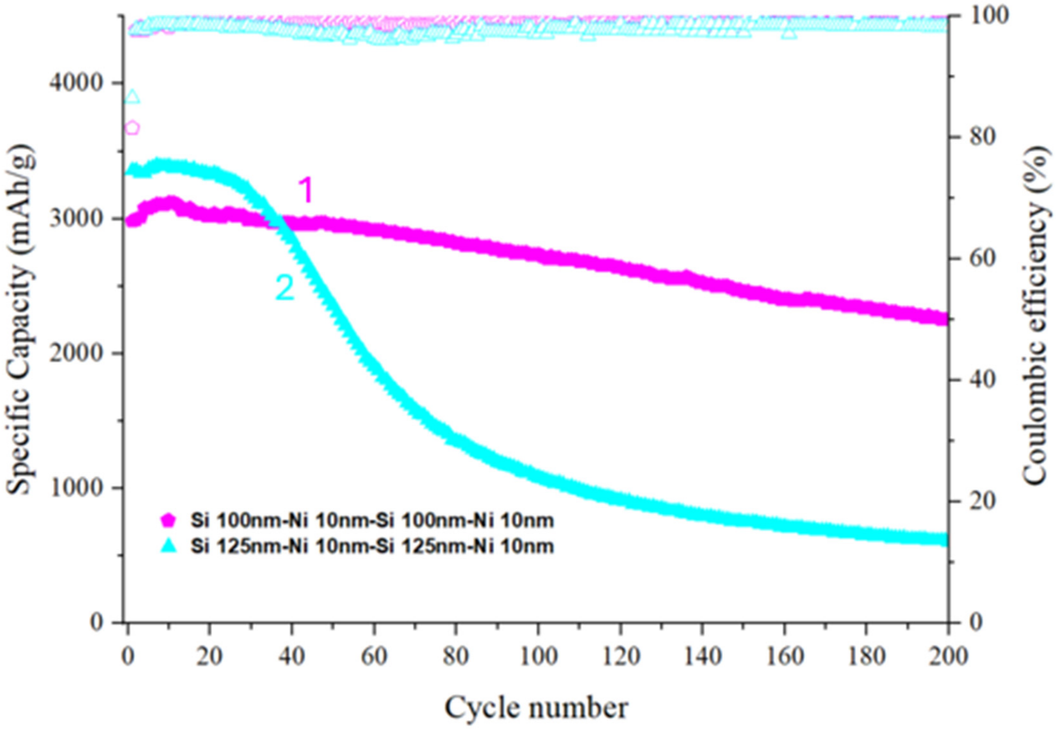


Figure S5. Cyclic performance of Si (100nm)/Ni (10nm)/Si (100nm)/Ni (10nm) anode with different Si thicknesses under 0.05mA/cm^2 test current density.

As depicted in Figure S5, a significant capacity retention degradation was observed after 200 cycles when the active material Si layer thickness was increased from 100 to 125 nm. This degradation is primarily attributed to the substantial volume expansion of Si during the alloying and de-alloying process. The 10 nm Ni protective layer proved insufficient to accommodate this volume expansion and maintain structural integrity. To further enhance the performance of the Si 100nm/Ni/Si 100nm/Ni anode, the thickness of the Ni layer was increased to 20 nm.

35. Li, W., Sun, X., & Yu, Y. (2017). Si-, Ge-, Sn-based anode materials for lithium-ion batteries: from structure design to electrochemical performance. Small Methods, 1(3), 1600037.