Supporting Information

Prefabrication of "Trinity" Functional Binary Layers on Silicon Surface to Develop High-Performance Lithium-Ion Batteries

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EXPERIMENTAL SECTION

Electrochemical testing

Cyclic voltammetry (CV) was carried out in a voltage range from 0.01 V to 2 V at 0.05 mV s^{-1} on an Autolab potentiostat (Autolab Instruments, Switzerland). Cycling performance and rate capability of the cells were evaluated on a Neware battery cycler (CT-4008, Shenzhen, China). Electrochemical impedance spectroscopy (EIS) measurements were conducted on Zahner Elektrik IM6 electrochemical working station over the frequency range of 10 mHz to 10^2 kHz with an alternating voltage of 5 mV. The cells were controlled at 60% depth of discharge. Galvanostatic intermittent titration (GITT) technique was measured on Neware CT-4008 with 0.1 C lasting for 15 min; followed by a relaxation time of 30 min. For full cell fabrication, the Si electrode after electrochemical pre-lithiation was assembled into a full cell with NCM523 cathode.

Material characterizations

FTIR (TENSOR 27, BRUKER OPTICS, Germany) was conducted to observe the functional groups. XRD (Rint 2000, Rigaku with Cu Kα) was applied to characterize the crystal structure. SEM was observed on Hitachi SV-8010 operated at 10 kV. TEM was conducted via FEI Tecnai G2 F20 S-TWIN under 200 KV. The chemical composition of Si electrodes was identified by XPS (Thermo Fisher, USA). The electrodes after cycles were rinsed in DMC solvent and thoroughly dried in vacuum chamber before SEM and XPS tests.

Calculation and simulation

DFT calculations were performed were optimized by using the DMol3 code.[1, 2] The all-electron PBE functional, along with a DND basis set, was used for all the geometry optimizations. The geometric structures of all molecules have been optimized. Based on the optimized structure, the lowest unoccupied molecular orbital and highest occupied molecular orbital energy of the corresponding electrolyte molecules, AZ and NSF molecules were calculated.

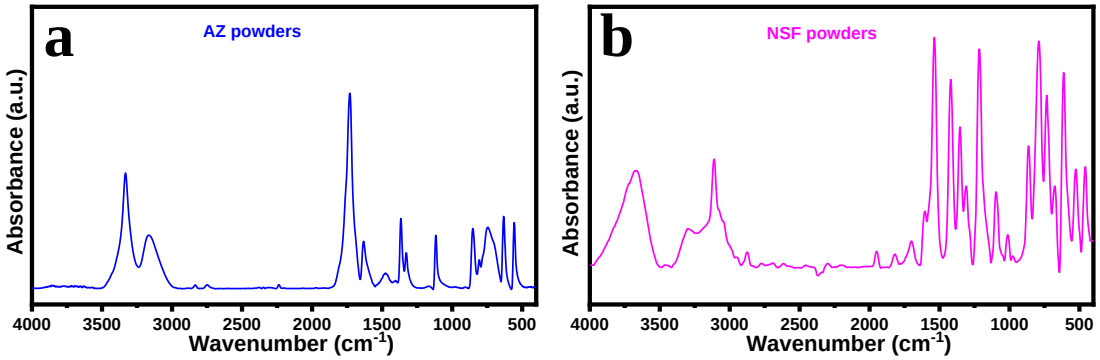

Figure S1. FTIR spectra of the pure (a) AZ and (b) NSF powders.

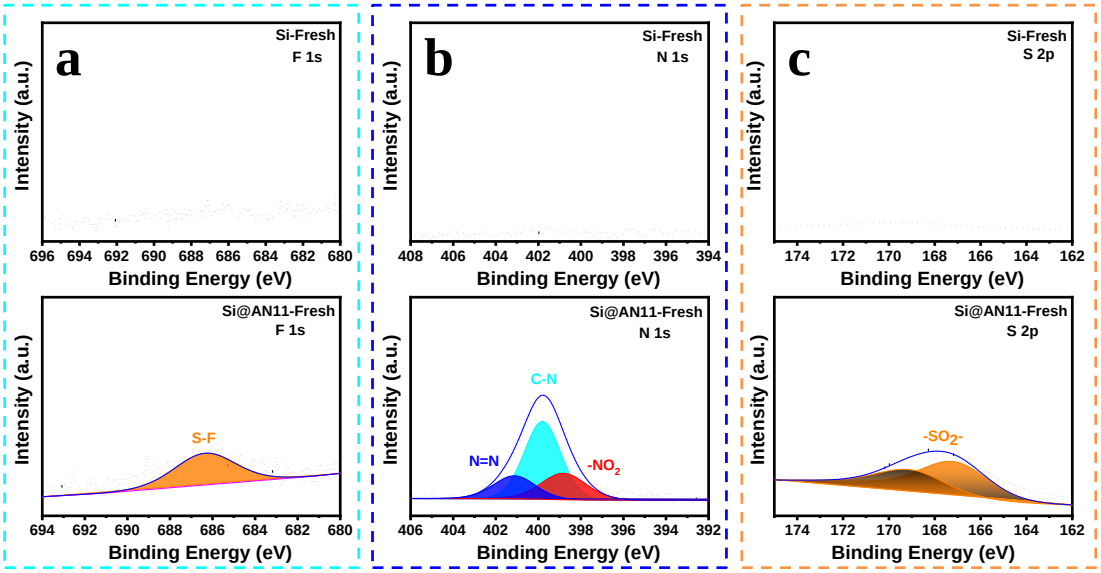

Figure S2. XPS spectra of the fresh Si and Si@AN11 electrodes: (a) F 1s spectra, (d) N 1s spectra, (c) S 2p spectra.

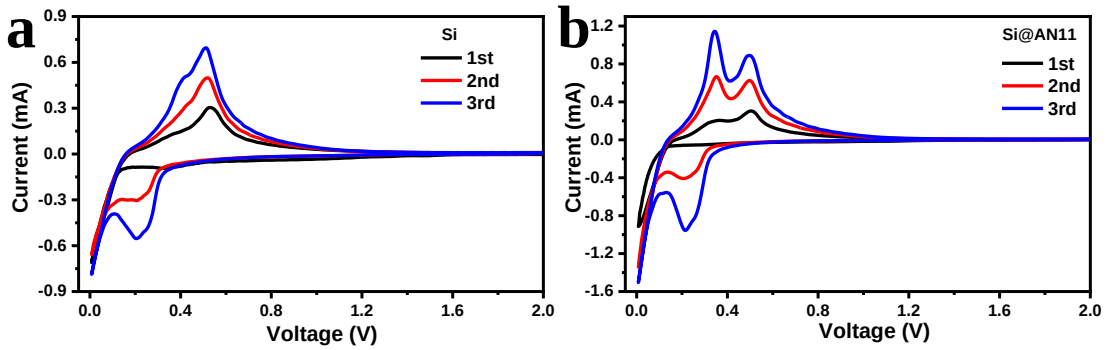

Figure S3. Initial three CV curves of Si and Si@AN11 anodes at 0.05 mV s^{-1}

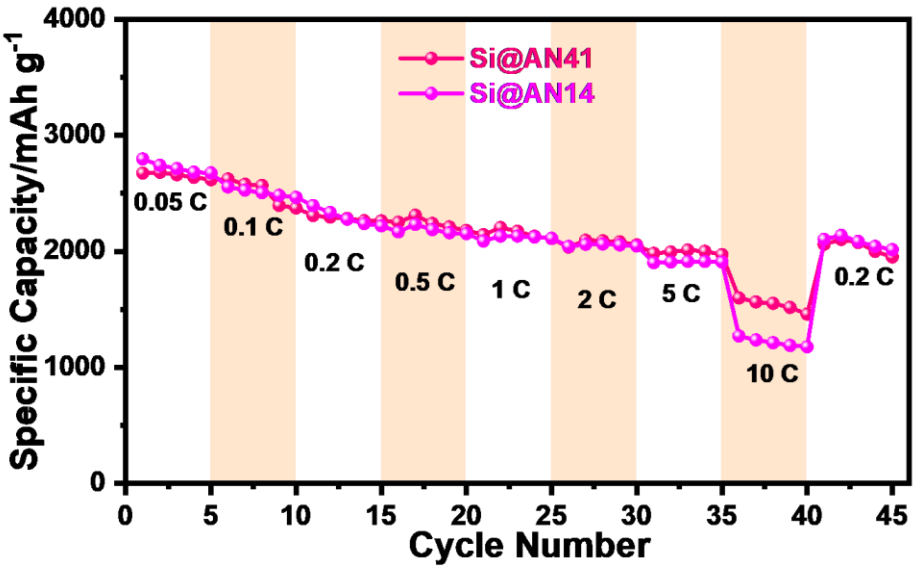

Figure S4. Rate capability for Si@AN41 and Si@AN14 anodes

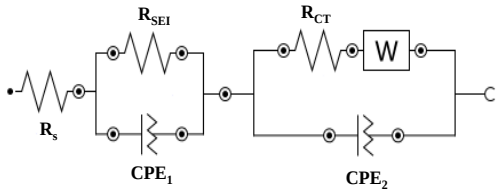

Figure S5. The equivalent circuit used for fitting the impedance spectra.

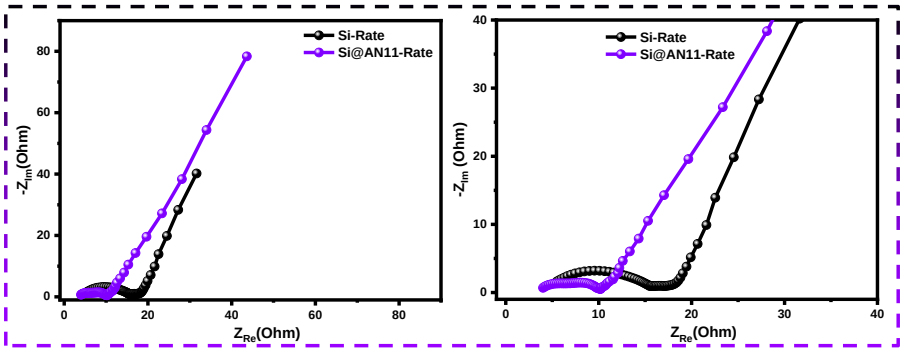

Figure S6. Nyquist plots and partially enlarged curves of Si and Si@AN11 anodes after the rate test.

Table S1. The corresponding resistance values of Si and Si@AN11 anodes after the rate test.

State	R_s/Ohm	R_{SEI}/Ohm	R_{ct}/Ohm
Si-Rate	3.84	12.2	8.47
Si@AN11-Rate	3.69	1.1	5.87

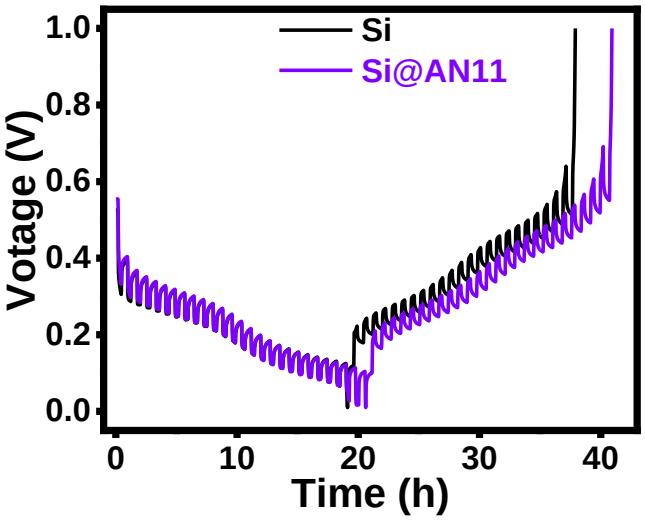

Figure S7. GITT profiles of the naked Si and Si@AN11 anodes.

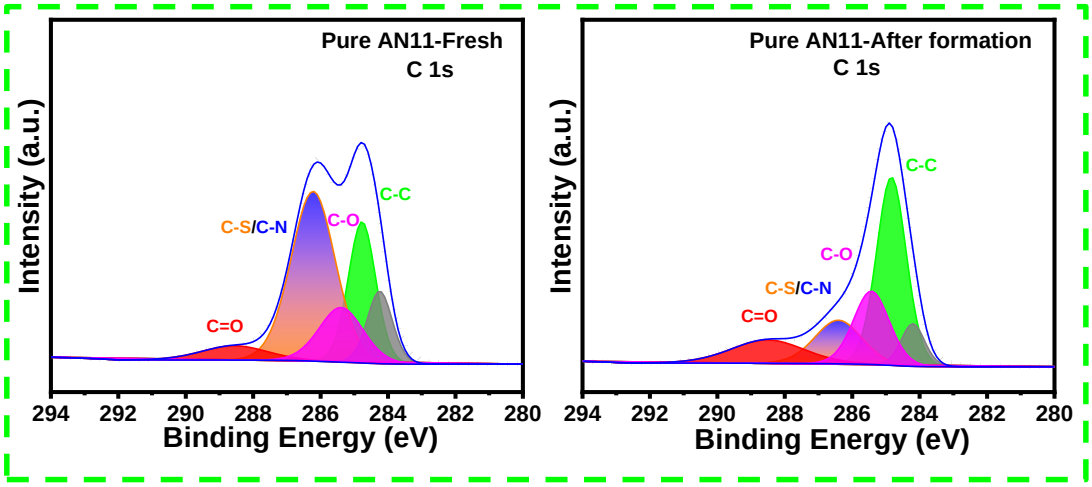

Figure S8. C 1s spectra of the fresh and as-formed AN11 electrodes.

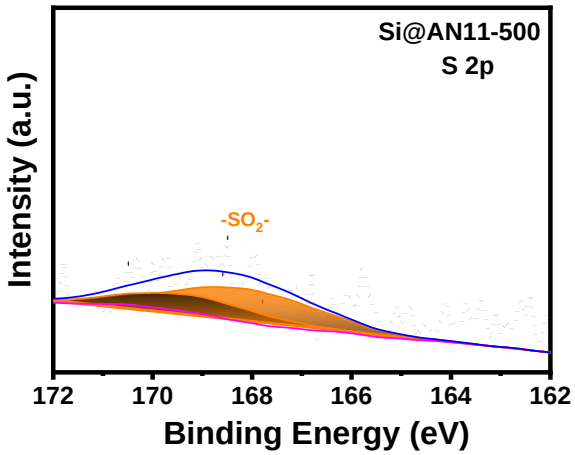

Figure S9. S2p spectrum of the Si@AN11 anode after 500cycles.

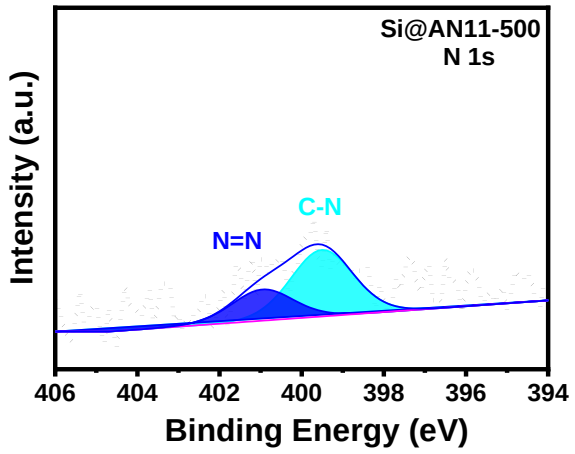

Figure S10. N 1s spectrum of the Si@AN11 anode after 500cycles.

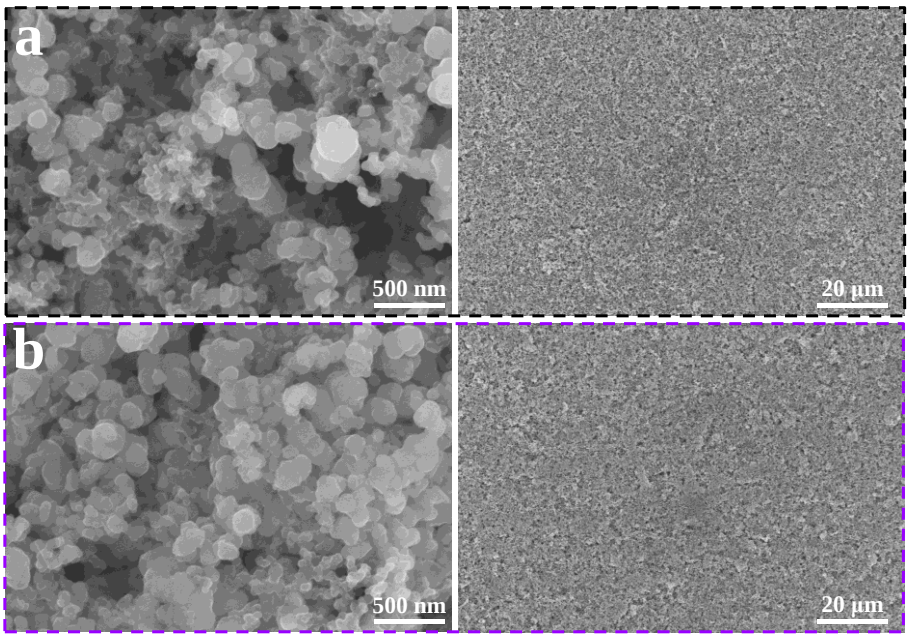

Figure S11. SEM images of the fresh (a) Si and (b) Si@AN11 electrodes.

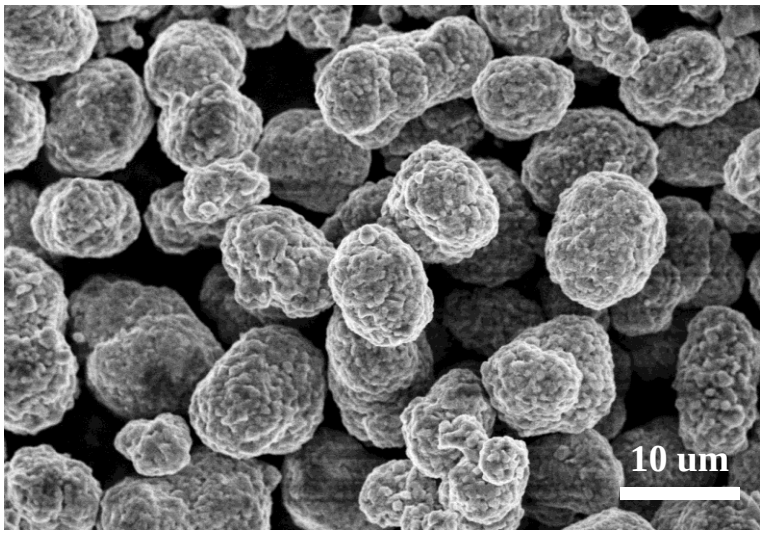

Figure S12. SEM image of NCM523 powders.

REFERENCES

1. Mulliken, R. S., Electronic Population Analysis on LCAO–MO Molecular Wave Functions. I. *J, Chem. Phys.* **1955,** *23* (10), 1833-1840.

2. Delley, B., From molecules to solids with the DMol3 approach. *J. Chem. Phys.* **2000,** *113* (18), 7756-7764.