

The Impact of Volume Expansion on Thermodynamic and Kinetic Properties of Graphite/Si Alloy Composite Anodes

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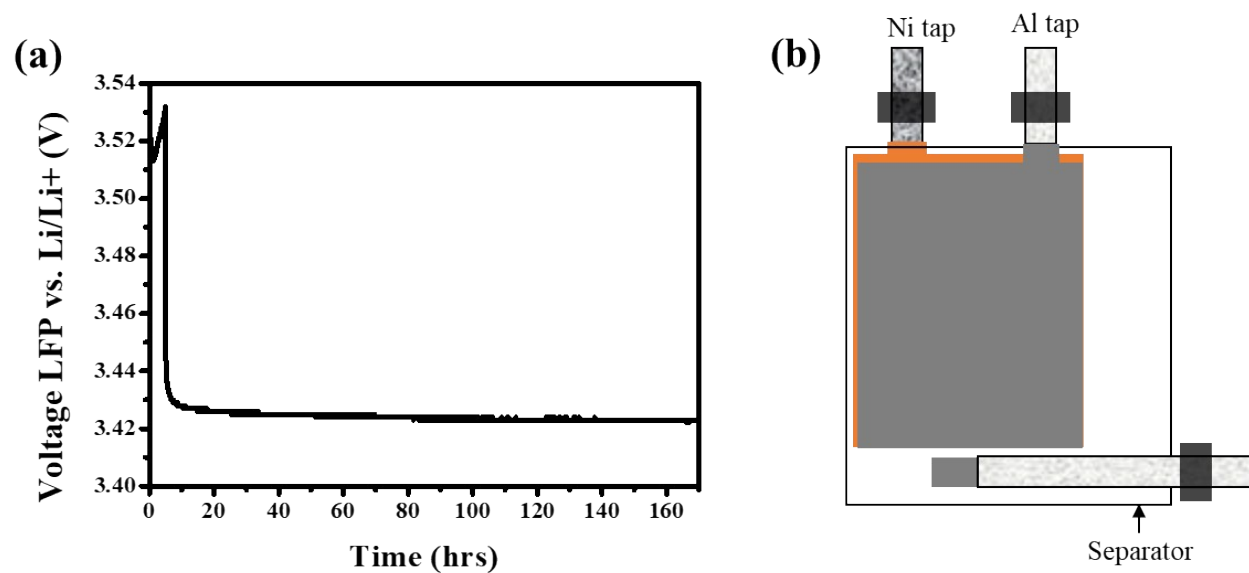


Fig. S1 (a) Voltage profile of the LFP electrode after charging at 0.1C for 5 h, demonstrating stable performance as a reference electrode. (b) Schematic illustration of the three-electrode pouch cell.

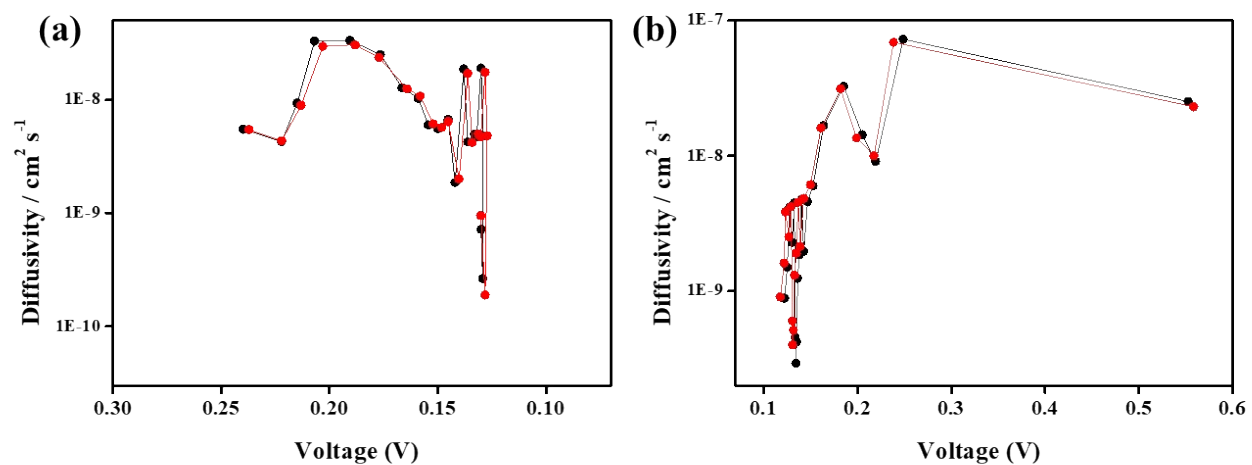


Fig. S2 Reproducibility test of graphite electrodes under identical conditions: (a) lithiation and (b) de-lithiation processes showing consistent potential profiles and diffusivity trends (deviation within 10 %).

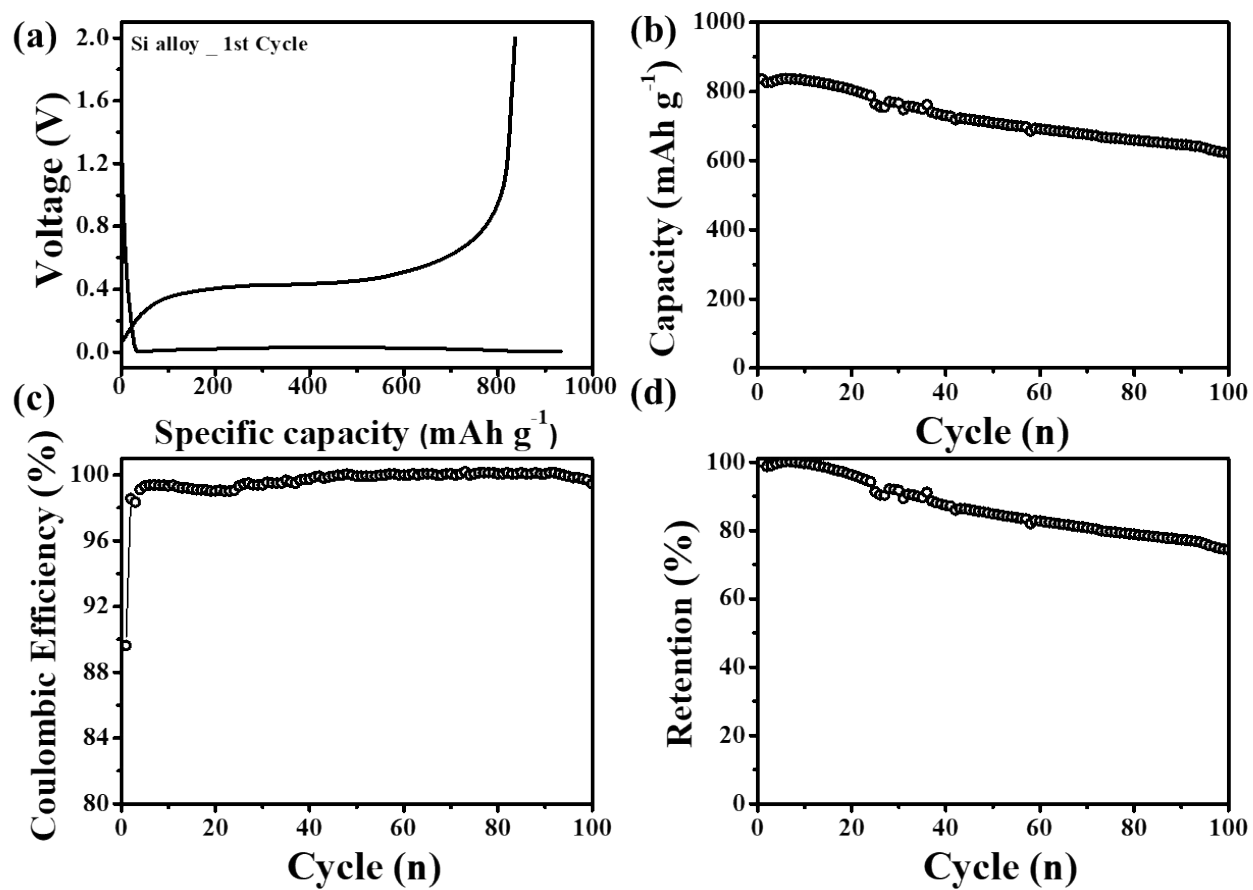


Fig. S3 Electrochemical performance of the Si alloy anode. (a) initial voltage profile, (b) cycling performance, (c) coulombic efficiency, and (d) capacity retention.

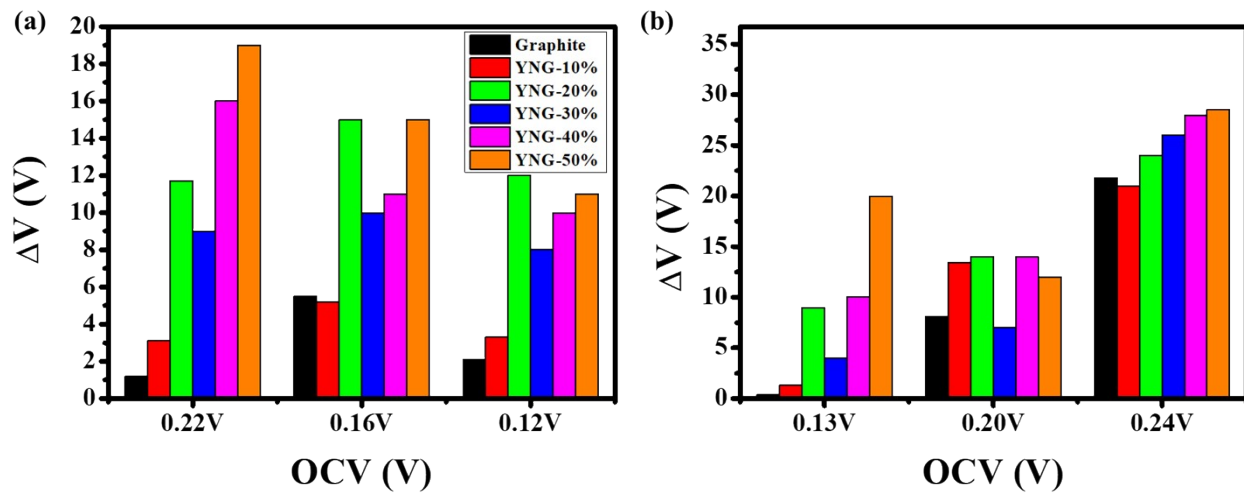


Fig. S4 ΔV as a function of OCV during (a) lithiation and (b) de-lithiation.

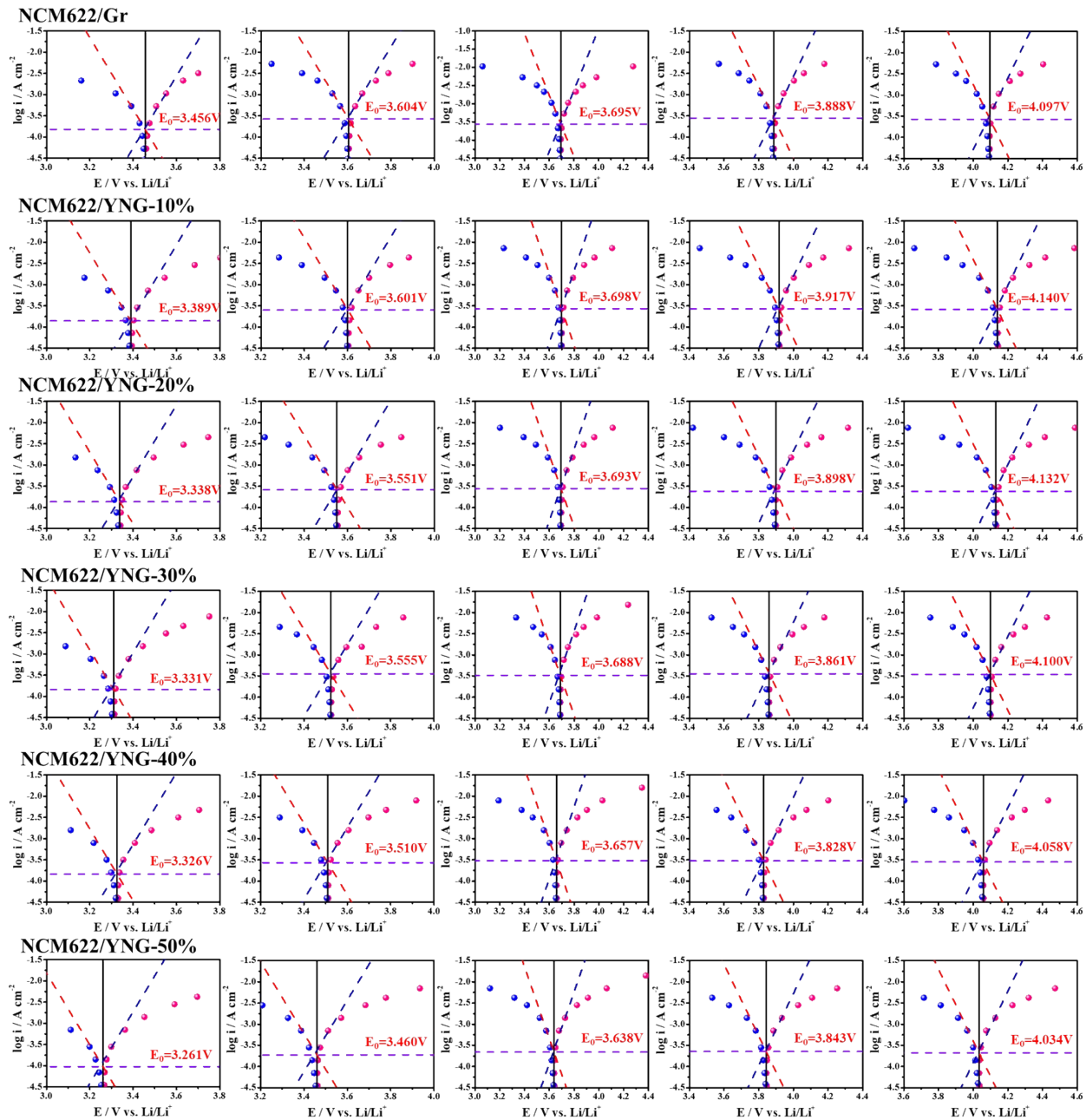


Fig. S5 Tafel plots of the graphite/Si alloy anode at different states of charge (SOC, from left to right: 10, 30, 50, 70, and 90 %).

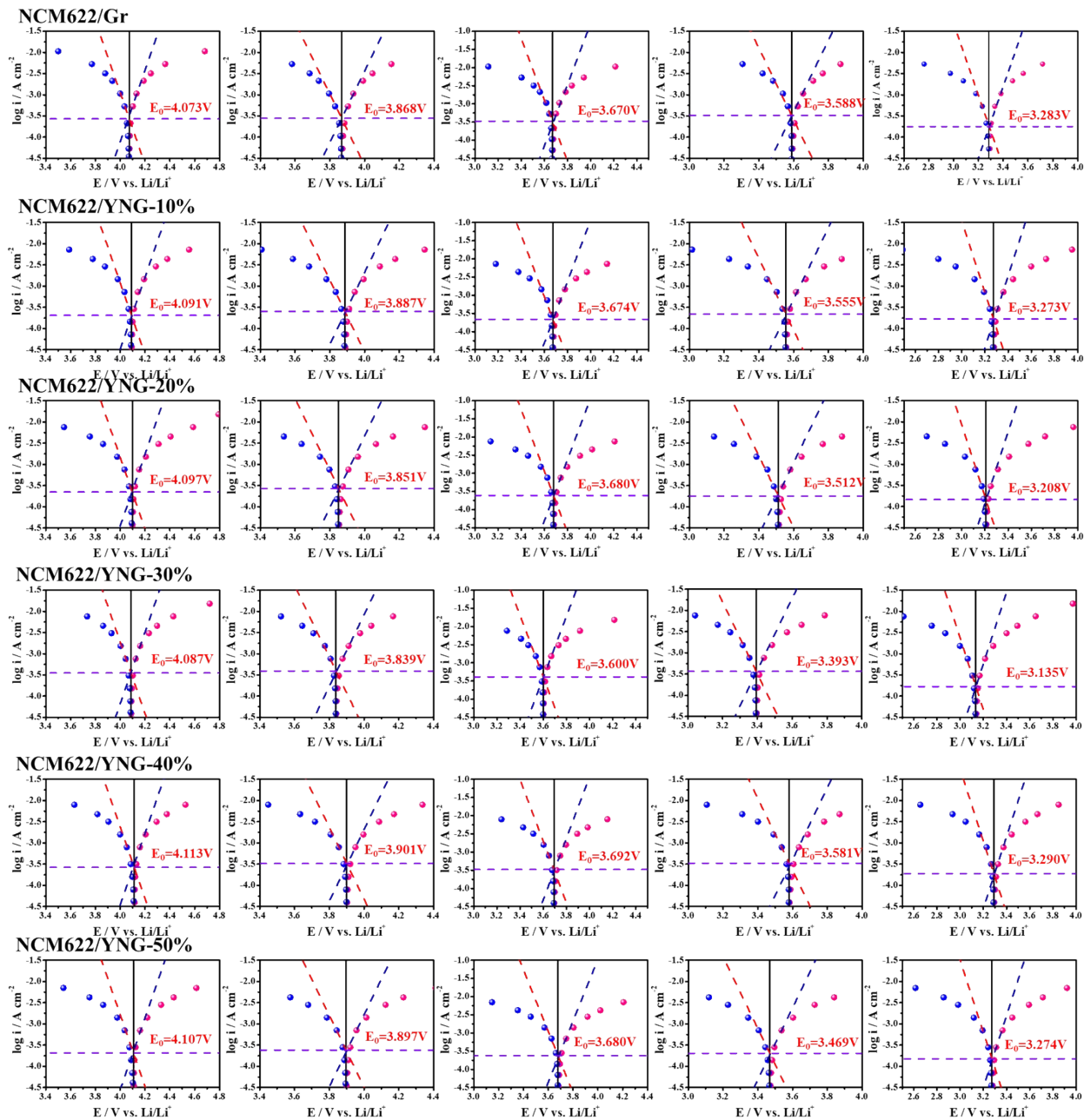


Fig. S6 Tafel plots of the graphite/Si alloy anode at different depths of discharge (DOD, from left to right: 10, 30, 50, 70, and 90 %).

Table. S1 Electrode properties of graphite, and graphite/Si composite anodes.

Anode	Graphite content (wt%)	Si alloy content (wt%)	Electrode Density (g cc ⁻¹)	Mass loading (mg cm ⁻²)	Initial Thickness (μm)
Graphite	100	0	1.60	3.84	24
YNG-10%	90	10	1.68	4.20	25
YNG-20%	80	20	1.62	4.38	27
YNG-30%	70	30	1.63	4.08	25
YNG-40%	60	40	1.60	4.43	28
YNG-50%	50	50	1.58	3.96	25

Table. S2 Equivalent circuit fitting results for EIS analysis.

Anode	OCV (V)	R_s (Ω)	R_{SEI} (Ω)	R_{ct} (Ω)
Graphite	2.71	1.63	0.90	12.4
YNG-10%	2.83	1.84	0.95	4.39
YNG-20%	2.80	1.63	1.41	1.89
YNG-30%	2.80	1.63	1.91	0.38
YNG-40%	2.81	1.52	2.23	1.85
YNG-50%	2.82	1.15	2.63	0.53

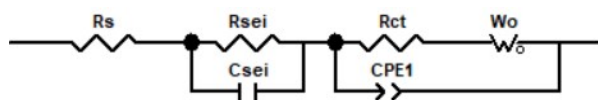


Table. S3 Parameters used for porosity calculation.

Parameter	Value
$\rho(\text{Graphite}) \text{ g cm}^{-3}$	2.22
$\rho(\text{Si-alloy}) \text{ g cm}^{-3}$	3.41
$\rho(\text{SWCNT}) \text{ g cm}^{-3}$	1.3
$\rho(\text{CMC}) \text{ g cm}^{-3}$	1.59
$\rho(\text{SBR}) \text{ g cm}^{-3}$	0.94
$\text{Porosity (\%)} = 1 - \frac{\text{electrode density}}{\text{theoretical density}} \times 100$	