Supporting Information

Silicon/Graphite Composite Host for Tuned Lithiophilicity in Li Metal Anodes

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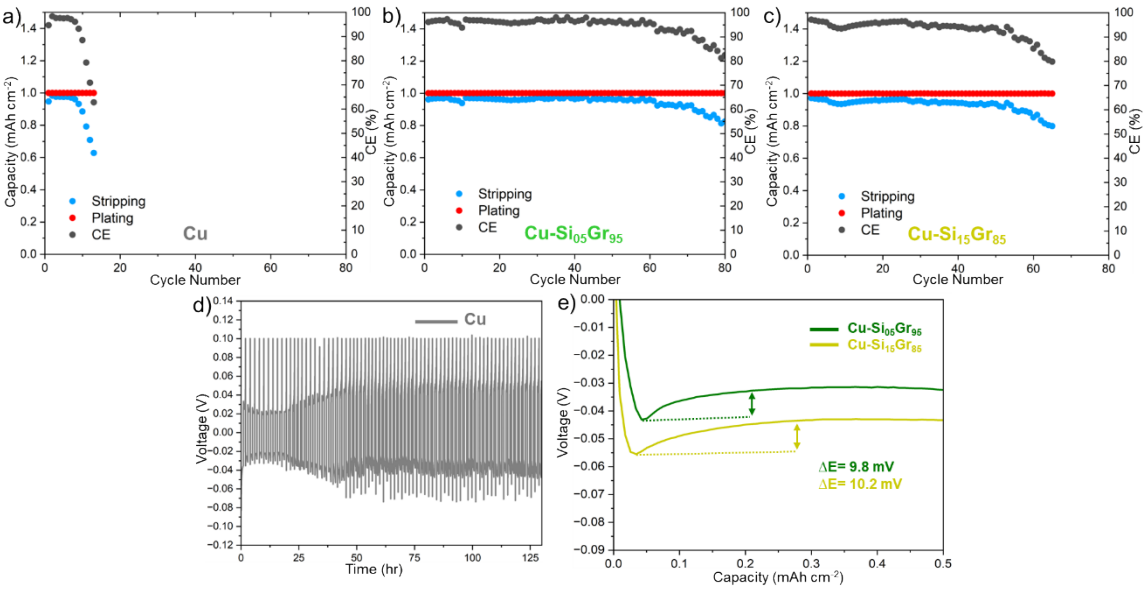


Figure S1. CE tests of a) Cu, b) Cu-$Si_{05}Gr_{95}$ and c) Cu-$Si_{15}Gr_{85}$ tested at a current density of 1 mA cm^{-2} for 1 mAh cm^{-2} Li plating/stripping. And voltage-time profile of d) Cu, as well as e) nucleation overpotentials of Cu-$Si_{05}Gr_{95}$ and Cu-$Si_{15}Gr_{85}$ tested at 1 mA cm^{-2}.

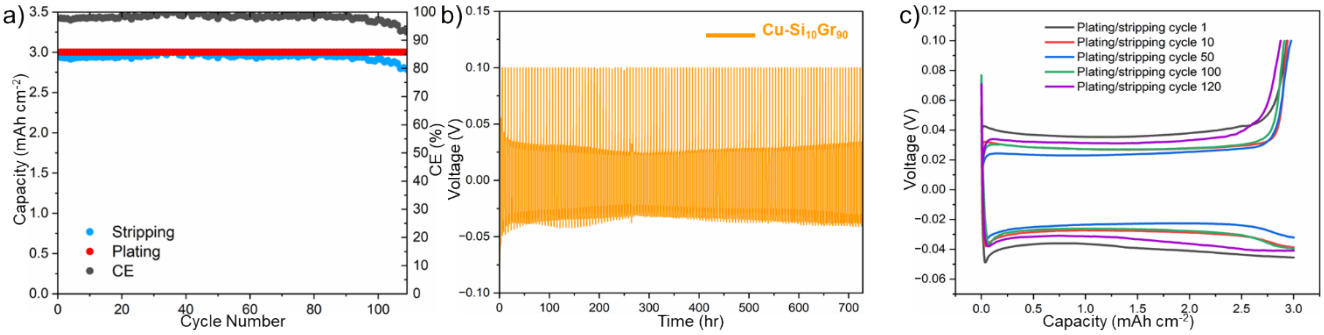


Figure S2. CE test of a) Cu-$Si_{10}Gr_{90}$@Li and b) corresponding voltage-time profile as well as c) Voltage-Areal capacity plot evaluated at a current density of 1 mA cm^{-2} and 3 mAh cm^{-2} Li plating/stripping.

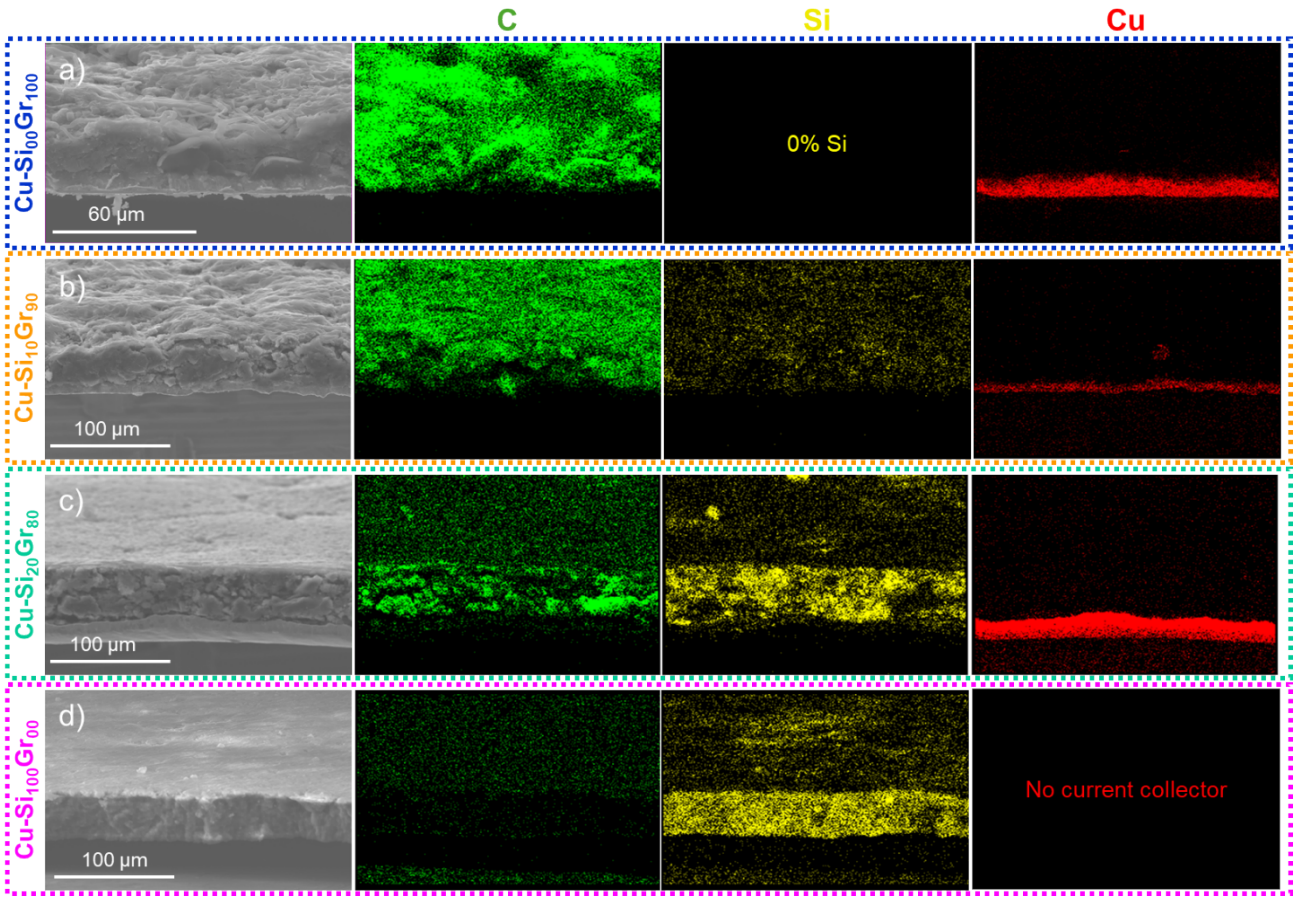


Figure S3. SEM cross section along with EDX analysis of a) of Cu-$Si_{00}Gr_{100}$@Li, b) Cu-$Si_{10}Gr_{90}$@Li, c) Cu-$Si_{20}Gr_{80}$@Li and d) Cu-$Si_{100}Gr_{00}$ after 10 plating/stripping cycles (on the plated state) using a current density of 1 mA cm^{-2} and a plating capacity of 1 mAh cm^{-2}.

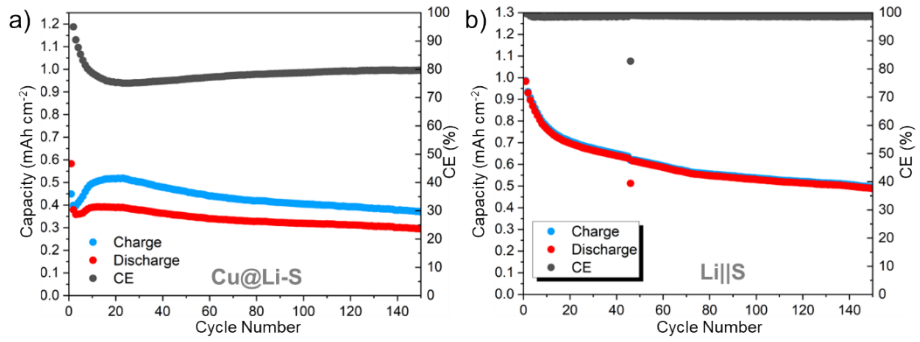


Figure S4. CE test of a) Cu@Li and b) Li cycled in a full cell against Sulfur cathode using 0.2C rate capability for 150 cycles.

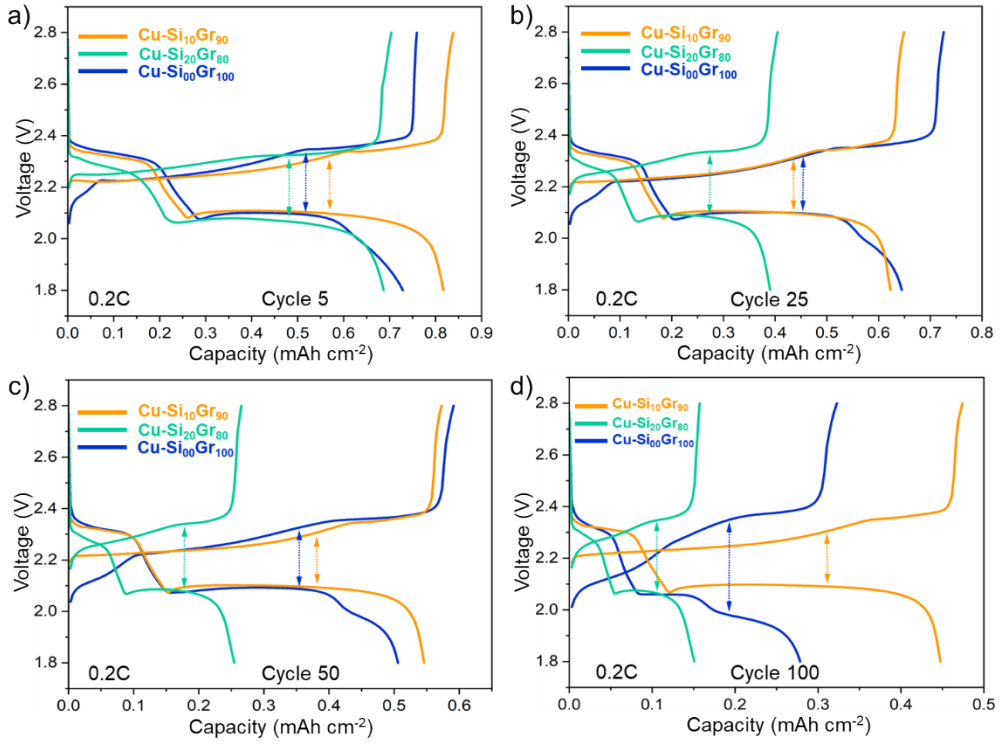


Figure S5. Voltage-Areal capacity plots at cycles a) 5, b) 25, c) 50 and d) 100 of Cu-$Si_{00}Gr_{100}$@Li-S, Cu-$Si_{20}Gr_{80}$@Li-S and Cu-$Si_{10}Gr_{90}$@Li-S cells at 0.2C.

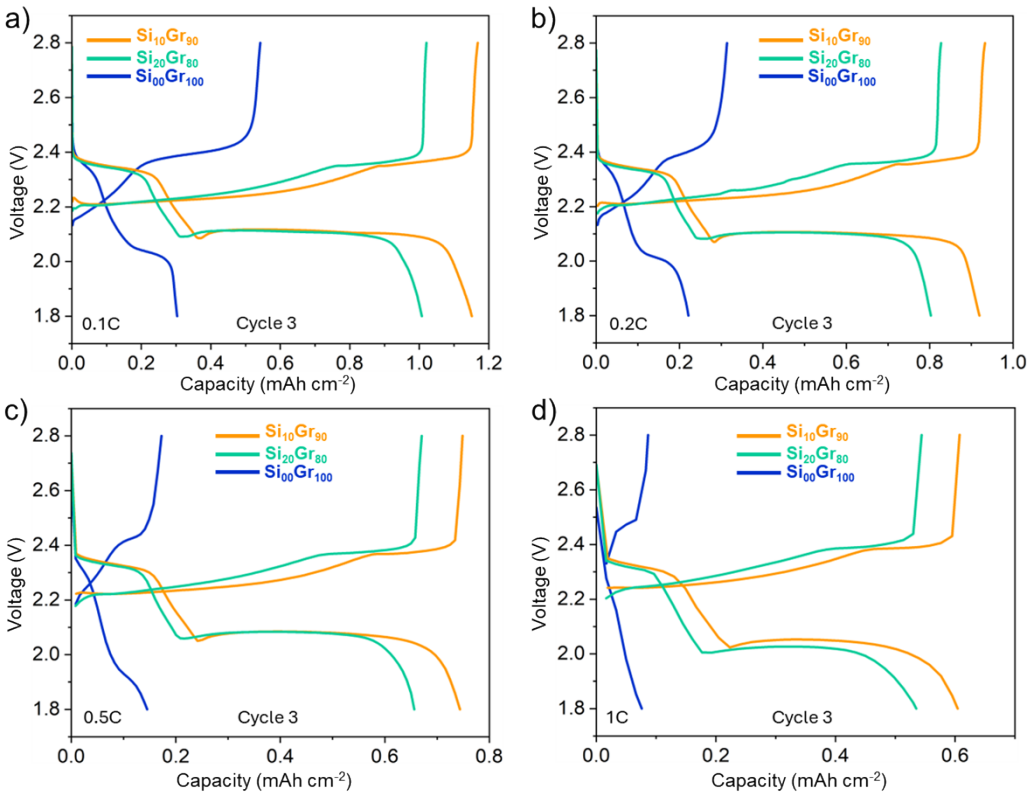


Figure S6. Voltage-Areal capacity plots at 3rd cycles of Cu-$Si_{00}Gr_{100}$@Li-S, Cu-$Si_{20}Gr_{80}$@Li-S and Cu-$Si_{10}Gr_{90}$@Li-S cells at C-rates of a) 0.1, b) 0.2, c) 0.5 and d) 1C.

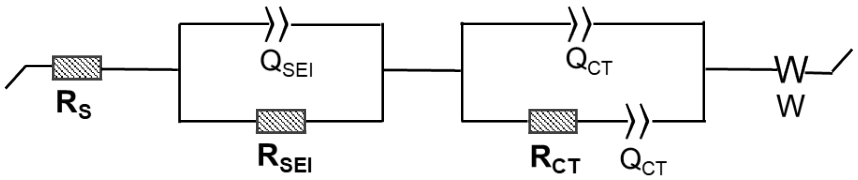


Figure S7. Applied equivalent circuit for fitting EIS data of Sulfur full cells.

Sample	Cycle No.	R_s (ohm)	R_{SEI} (ohm)	R_{CT} (ohm)	R_{SEI+CT} (ohm)
$Cu\text{-}Si_{00}Gr_{100}$ @Li-Sulfur	1^{st}	4.75	8.88	5.47	14.34
	100^{th}	5.46	12.6	7.92	20.52
$Cu\text{-}Si_{20}Gr_{80}$ @Li-Sulfur	1^{st}	5.73	8.41	3.78	12.19
	100^{th}	8.37	10.47	6.41	16.88
$Cu\text{-}Si_{10}Gr_{90}$ @Li-Sulfur	1^{st}	4.99	5.3	1.52	6.82
	100^{th}	5.38	7.9	2.3	10.2

Table S1. EIS fitted data table containing R_s, R_{SEI}, R_{CT} values for each $Cu\text{-}Si_{00}Gr_{100}$@Li-S, $Cu\text{-}Si_{20}Gr_{80}$@Li-S and $Cu\text{-}Si_{10}Gr_{90}$@Li-S cells.

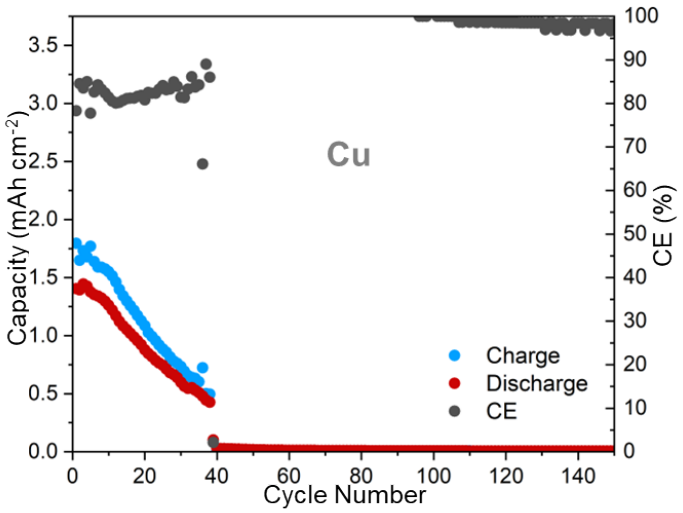


Figure S8. Cyclic performance of Cu@Li-NMC.

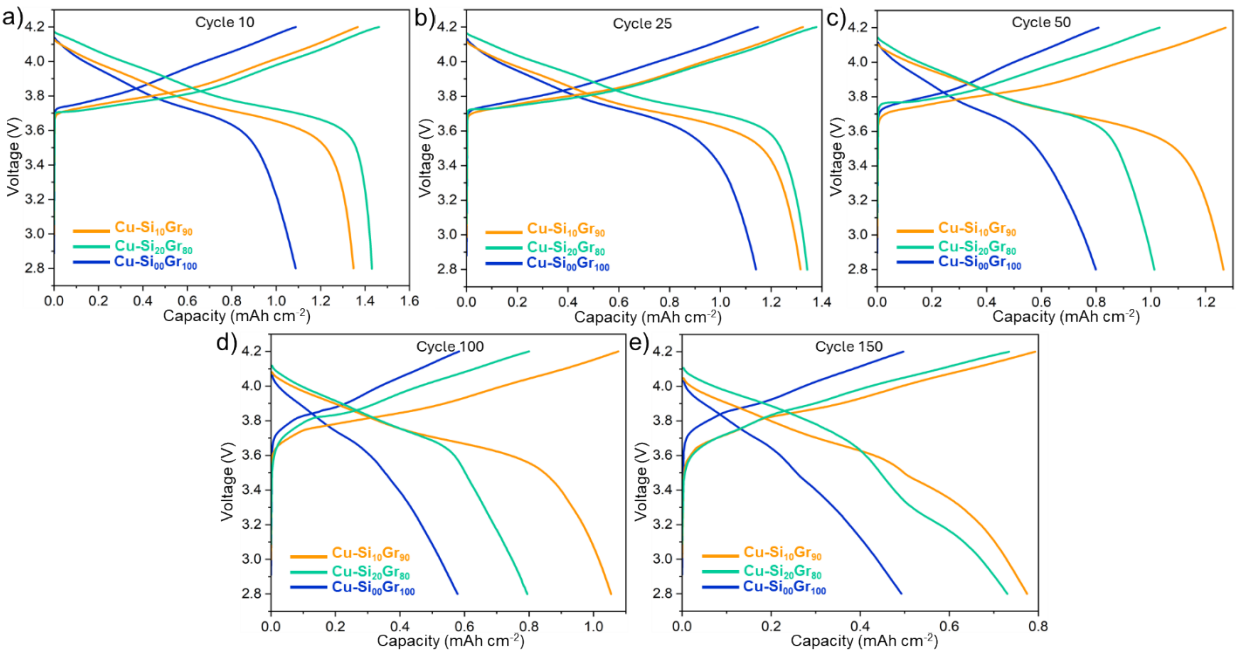


Figure S9. Voltage-Areal capacity graphs of Cu-$Si_{00}Gr_{100}$@Li-NMC, Cu-$Si_{10}Gr_{90}$@Li-NMC and Cu-$Si_{20}Gr_{80}$@Li-NMC full cells at various cycles including a) 10, b) 25, c)50, d)100 and e)150 at 0.2C.

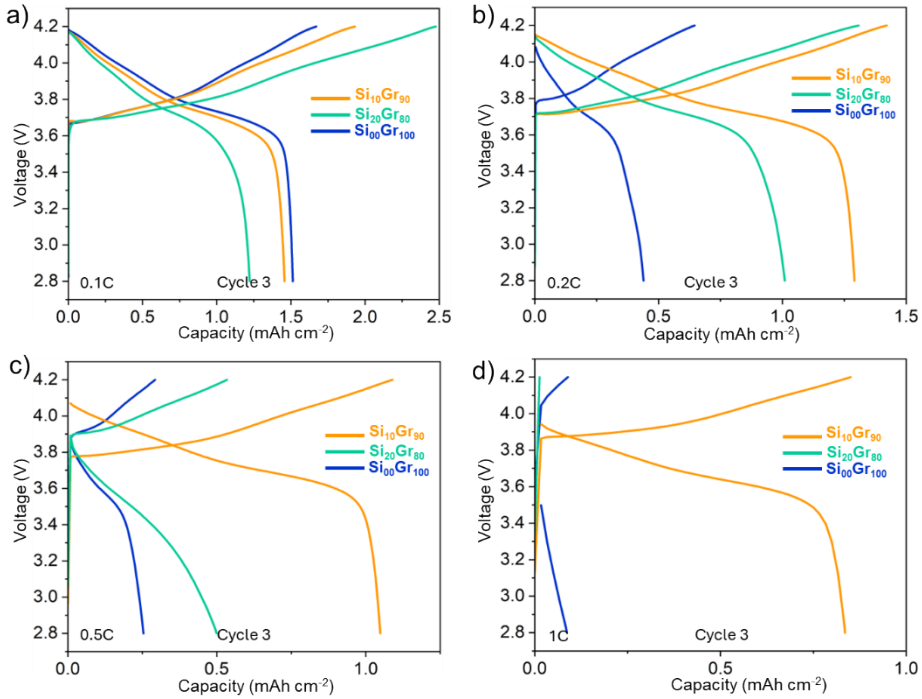


Figure S10. Voltage-Areal capacity plots at 3rd cycles of Cu-$Si_{00}Gr_{100}$@Li-NMC, Cu-$Si_{20}Gr_{80}$@Li-NMC and Cu-$Si_{10}Gr_{90}$@Li-NMC cells at C-rates of a) 0.1, b) 0.2, c) 0.5 and d) 1C.

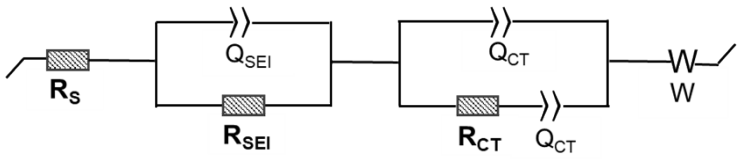
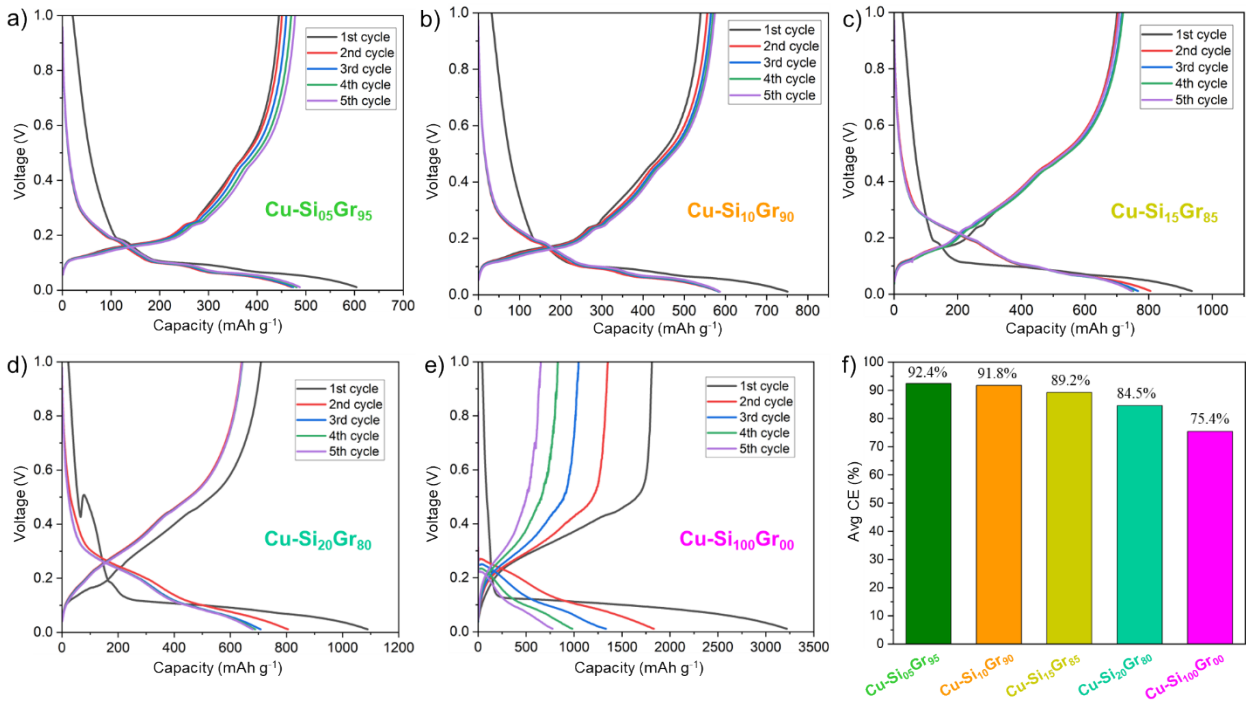


Figure S11. Applied equivalent circuit for fitting EIS data of NMC full cells.

Supporting Information

Sample	Cycle No.	R_s (ohm)	R_{SEI} (ohm)	R_{CT} (ohm)	R_{SEI+CT} (ohm)
$Si_{00}Gr_{100}$ @Li-NMC	1st	3.9	8.5	59.5	68
	100th	7.93	42.58	204.4	246.98
$Si_{20}Gr_{80}$ @Li-NMC	1st	3.4	5.36	45.01	50.37
	100th	8.3	27.65	130.9	158.55
$Si_{10}Gr_{90}$ @Li-NMC	1st	2.9	3.8	20.28	24.08
	100th	8.9	11.5	48.99	60.49

Table S2. EIS fitted data table containing R_s, R_{SEI}, R_{CT} values for each Cu-$Si_{00}Gr_{100}$@Li-NMC, Cu-$Si_{20}Gr_{80}$@Li-NMC and Cu-$Si_{10}Gr_{90}$@Li-NMC cells.



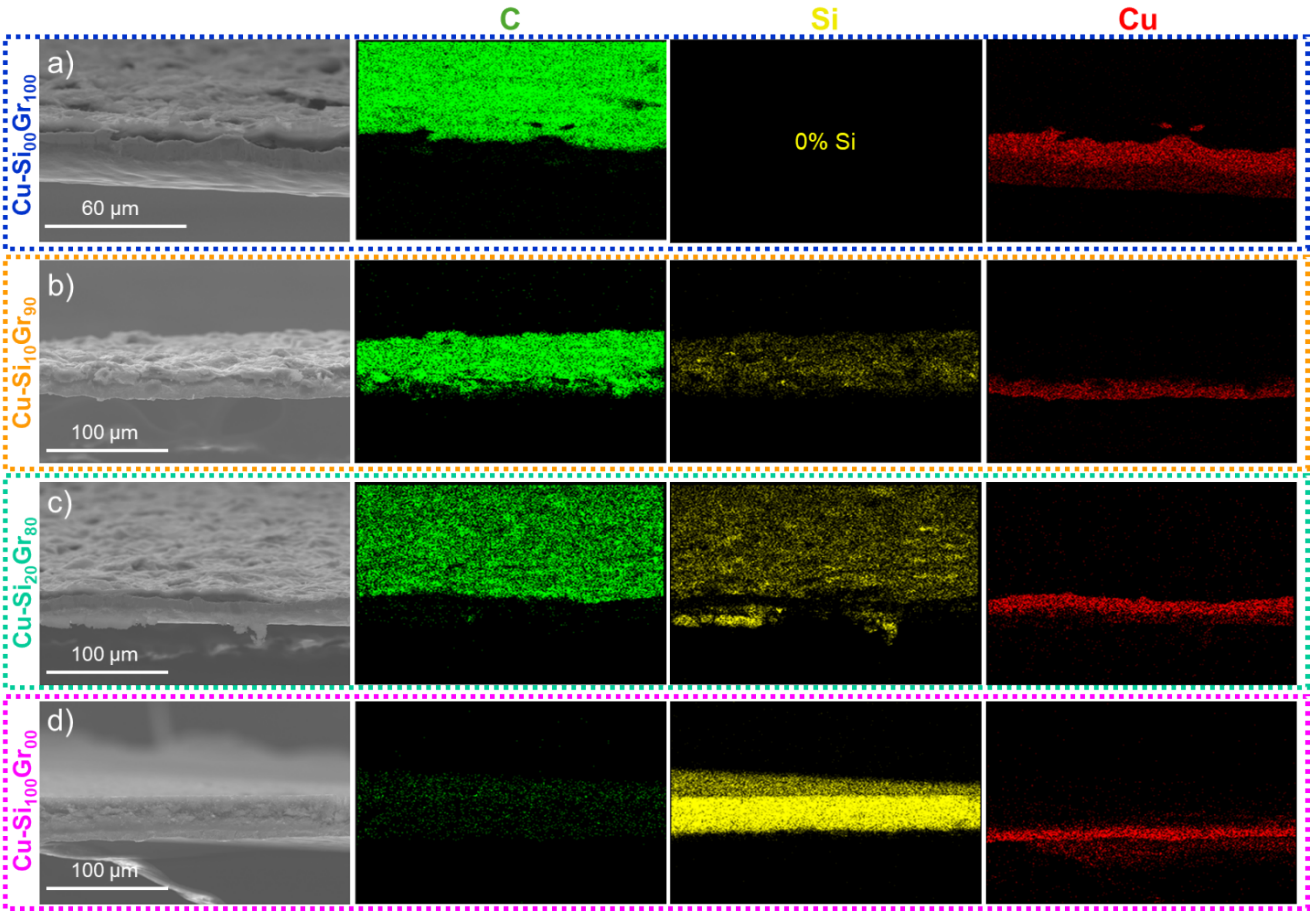


Figure S12. Voltage-capacity plots for the initial five charge/discharge cycles of a) Cu-$Si_{05}Gr_{95}$, b) Cu-$Si_{10}Gr_{90}$ c) Cu-$Si_{15}Gr_{85}$ d) Cu-$Si_{20}Gr_{80}$ and e) Cu-$Si_{100}Gr_{00}$ half cells prior to Li plating/stripping. f) the average CE for the initial charge/discharge cycles.

Figure S13. SEM cross section along with EDX analysis of pristine a) of Cu-$Si_{00}Gr_{100}$@Li, b) Cu-$Si_{10}Gr_{90}$@Li, c) Cu-$Si_{20}Gr_{80}$@Li and d) Cu-$Si_{100}Gr_{00}$.