Supporting Information

Nanospherical Solid Electrolyte Interface Layer
Formation in Binder-Free Carbon Nanotube
Aerogel / Si Nanohybrids to Provide Lithium-Ion
Battery Anodes with Long-Cycle Life and High
Capacity

Hyung Cheoul Shim,*a Ilhwan Kim, a,b Chang-Su Woo, a Hoo-Jeong Leeb and Seungmin Hyun*a

^a Department of Nano-Mechanics, Korea Institute of Machinery & Materials (KIMM), 156, Gajeongbuk-ro, Yuseong-gu, Daejeon, 34103, Republic of Korea

Department of Advanced Materials Science and Engineering, Sungkyunkwan University,
 2066, Seobu-ro, Jangan-gu, Suwon-si, Gyeonggi-do, 16419, Republic of Korea

E-mail: scafos@kimm.re.kr, hyun@kimm.re.kr

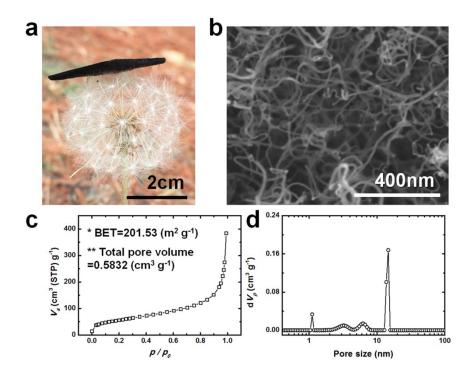


Fig. S1 (a) The photo of CNT aerogel plate. (b) SEM image of CNT aerogel. (c) N_2 absorption isotherms of CNT aerogel. (d) Meso-pore size distribution of the CNT aerogel.

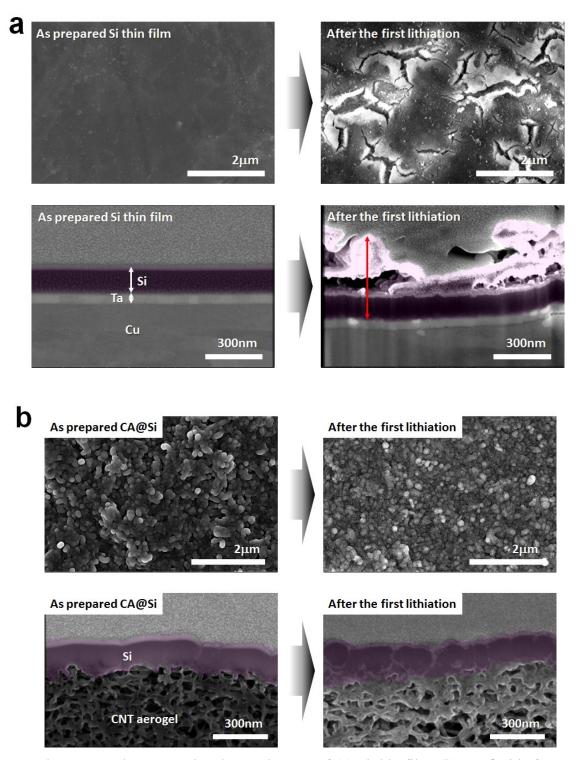


Fig. S2 The SEM and cross sectional SEM images of (a) Si thin film, (b) CA@Si before and after the first lithiation process

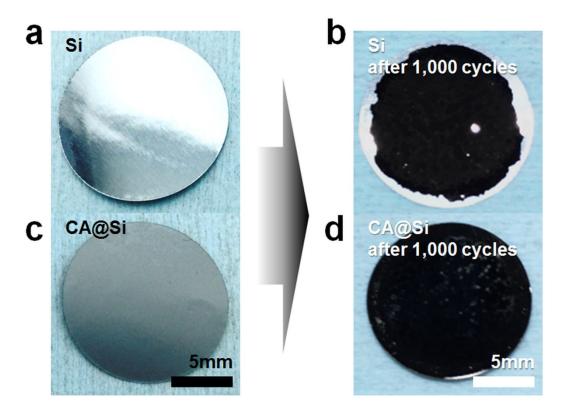


Fig. S3 Optical images of the Si and CA@Si electrode before and after cycling.

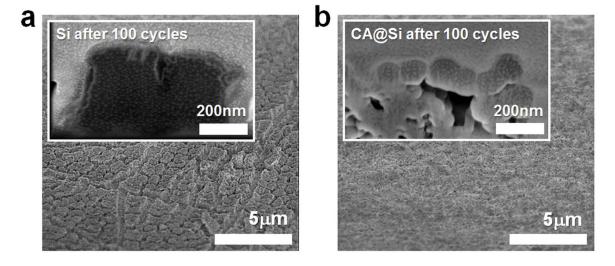


Fig. S4 Tilted FESEM images of the (a) Si and (b) CA@Si after cycling. The inset shows the corresponding cross-sectional FESEM images obtained using FIB techniques.

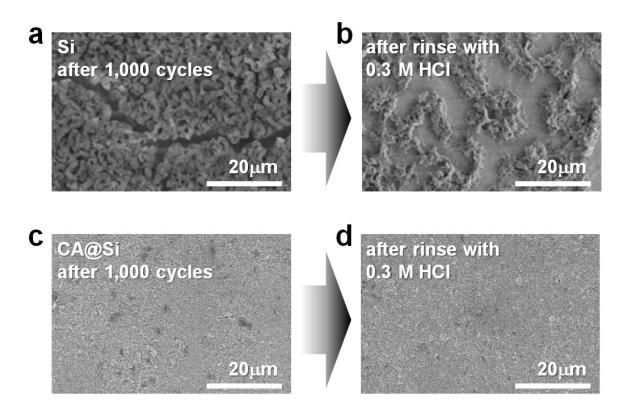


Fig. S5 FESEM image of the (a) Si and (c) CA@Si electrode after cycling. FESEM image of the (b) Si and (d) CA@Si after 1000 cycles with selective removal of SEI layers using chemical etching.

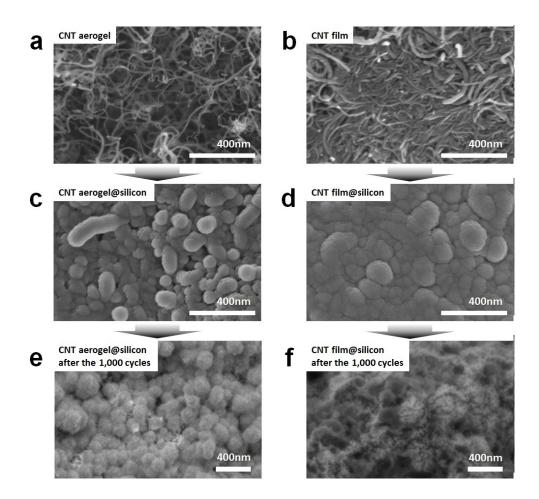


Fig. S6 FESEM images of (a) CNT aerogel (CA), (b) CNT film (CF), (c) Silicon deposited onto CA (CA@Si), (d) Silicon deposited onto CF (CF@Si), (e) CA@Si and (f) CF@Si after 1,000 electrochemical cycles.

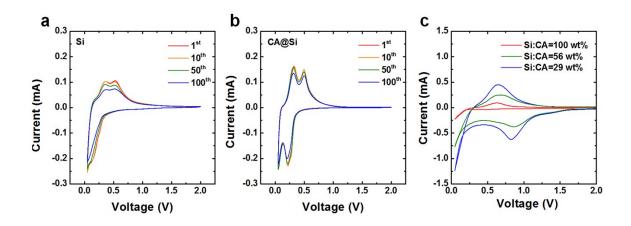


Fig. S7 CV curves (scan rate: 0.1 mV s⁻¹) of the 1, 10, 50, 100 cycles for (a) Si, (b) CA@Si electrodes. (c) CV curves of the CA@Si electrode for various Si/CA ratios.

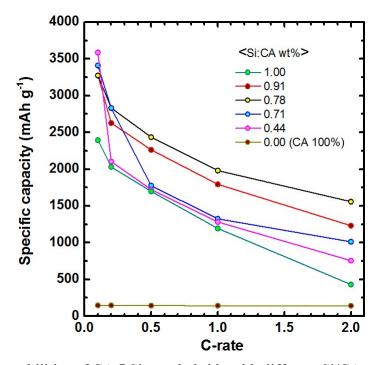


Fig. S8 The rate capabilities of CA@Si nanohybrids with different Si/CA ratio.

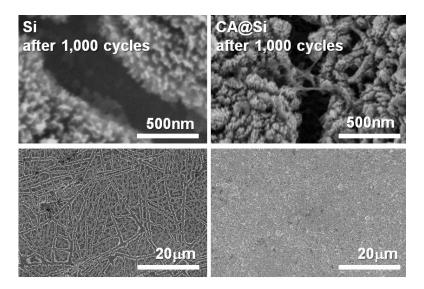


Fig. S9 FESEM image of the Si and CA@Si electrode after 1,000 cycles with selective removal of SEI layers using chemical etching. Note that the CNT aerogel maintained the conducting networks in the CA@Si electrode, while the cycled Si electrode lost the conducting path after cycling.

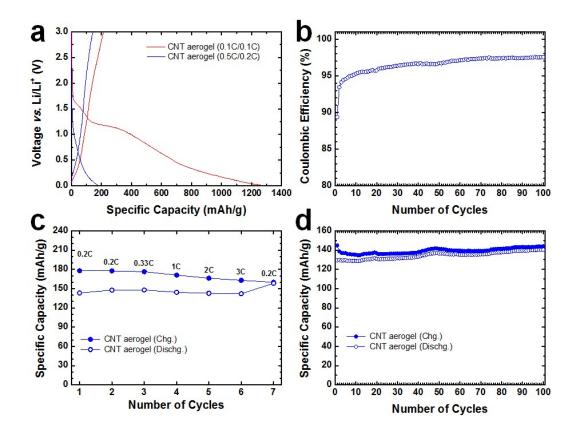


Fig. S10 Electrochemical performance of the CA. (a) charge-discharge profiles, (b) Coulombic efficiency, (c) rate capability and (d) charge-discharge curve as a function of the number for cycles.