

## Supplementary Materials

### **Designing Si-based nanowall arrays by dynamic shadowing growth to tailor the performance of Li-ion battery anodes**

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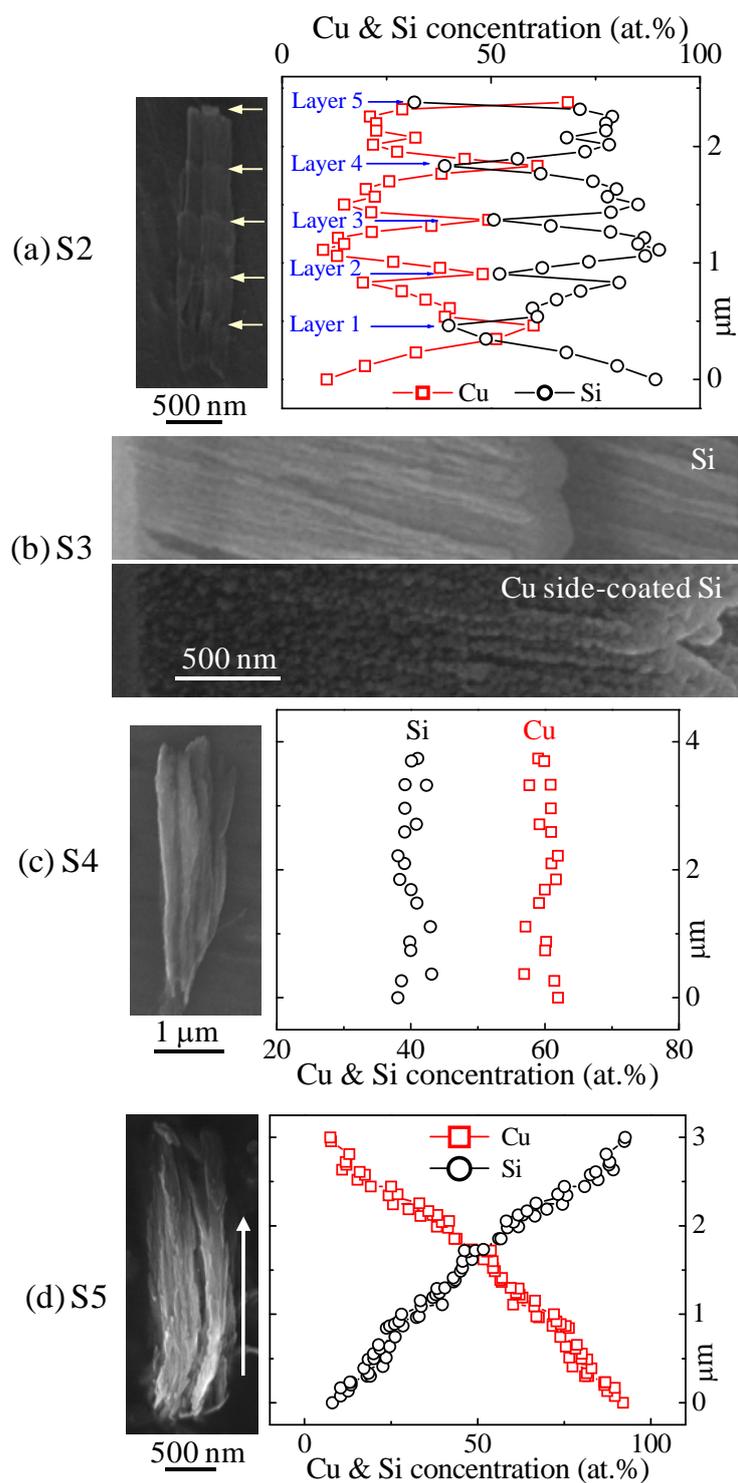
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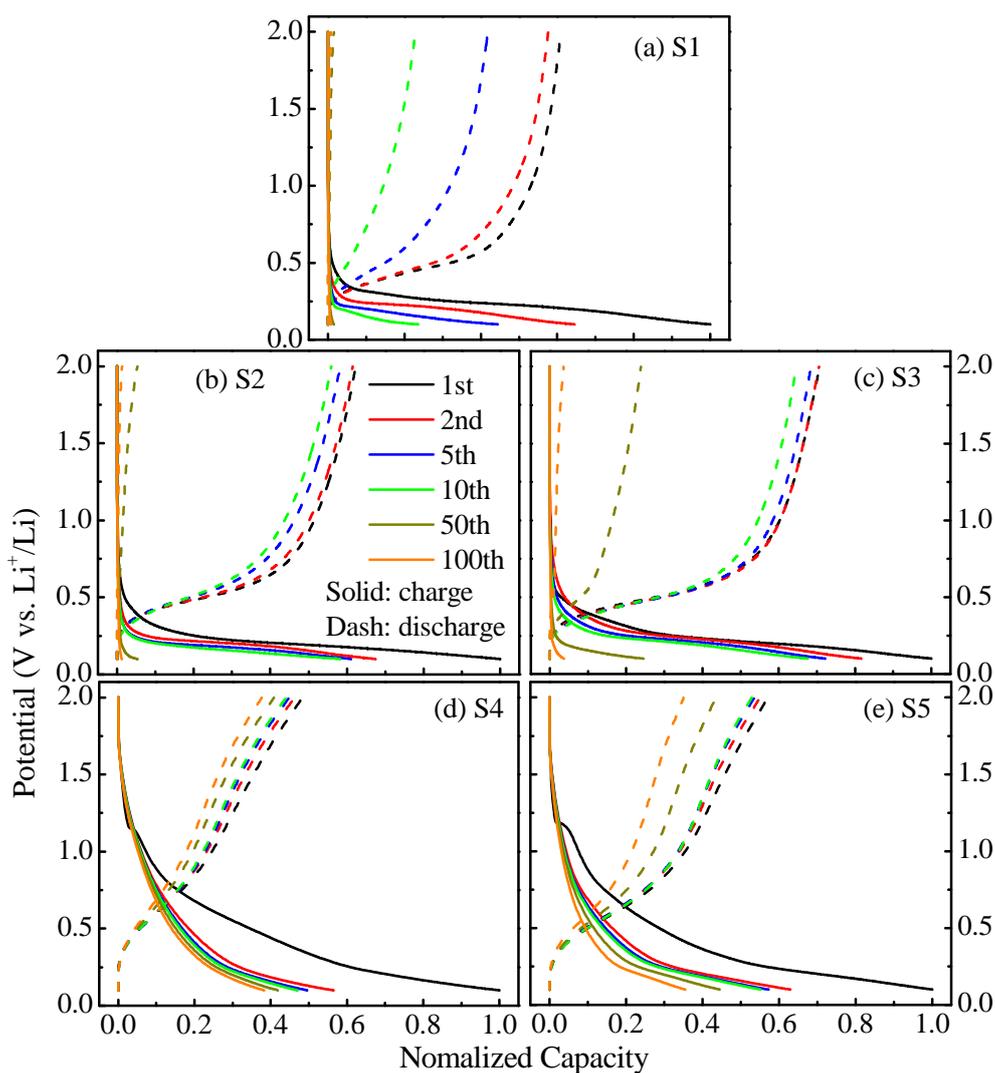
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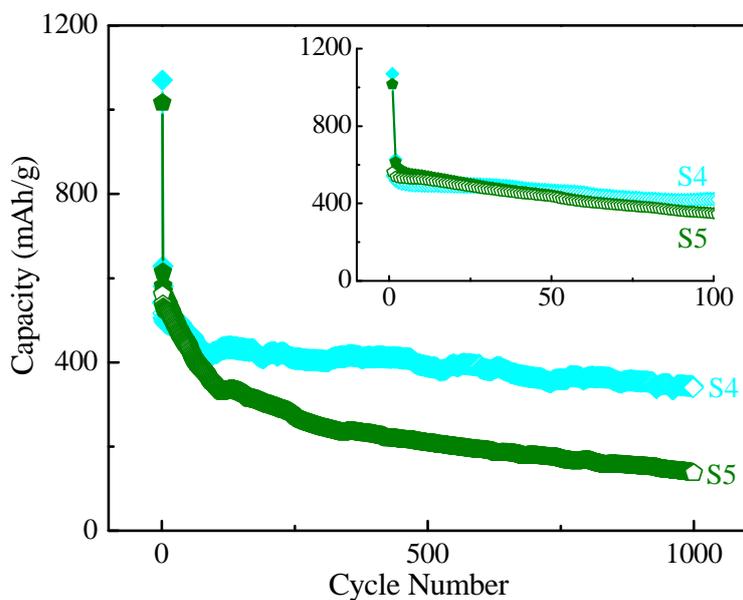


**Fig. A1** The SEM images and Cu & Si depth profiles reveal the expected structures: (a) 5-layer Cu/Si in S2; (b) Cu side-coated Si in S3, where the Cu layer in the form of connected nanoparticles covers continuously one side of each Si nanowall from bottom to top, by comparing the SEM images of Cu side-coated Si and pure Si samples; (c) uniform distribution of

Cu (~ 60 at.%) and Si (~ 40 at.%) through the whole length of co-deposited CuSi composite nanorods in S4; (d) graded distribution of Cu and Si in S5 (ref. Y. P. He, J. G. Fan and Y. P. Zhao, *Cryst. Growth Des.*, 2010, **10**, 4954-4958). The depth profiles were obtained by multiple energy dispersive X-ray spectroscopy (EDX) measurements along the samples shown in the corresponding SEM images. In the experiment, during the deposition of CuSi anodes on Cu foils, a piece of flat Si substrate was used as a control. A bundle of CuSi nanorods formed on the Si substrate was transferred onto a conductive carbon tape for composition analysis by EDX.



**Fig. A2** The charge and discharge curves in the 1<sup>st</sup>, 2<sup>nd</sup>, 5<sup>th</sup>, 10<sup>th</sup>, 50<sup>th</sup>, and 100<sup>th</sup> cycles for five different Si-based nanowall structures: (a) S1, (b) S2, (c) S3, (d) S4, and (e) S5.



**Fig. A3** The battery cycling performance for both the uniform CuSi composite (S4) and graded CuSi composite (S5) nanowall arrays cycled at a current of 0.05 mA ( $\sim 1/4C$  rate) in the potential range of 0.1 to 2 V versus  $\text{Li}^+/\text{Li}$  for 1000 cycles. Inserted are the corresponding 100-cycle curves. Solid and hollow symbols denote charge and discharge capacity data, respectively. The results show that, for S4, the capacity only decays from the first discharged 542 mAh/g to 421 mAh/g after 100 cycles (78% retention) and to 340 mAh/g after 1000 cycles (63% retention); while for S5, the capacity decays from the first discharged 561 mAh/g to 346 mAh/g (62% retention) and to 137 mAh/g (24% retention).