

# Supporting Information

## Silicon-Core Carbon-Shell Nanoparticles for Lithium-ion Batteries: Rational Comparison Between Amorphous and Graphitic Carbon Coatings

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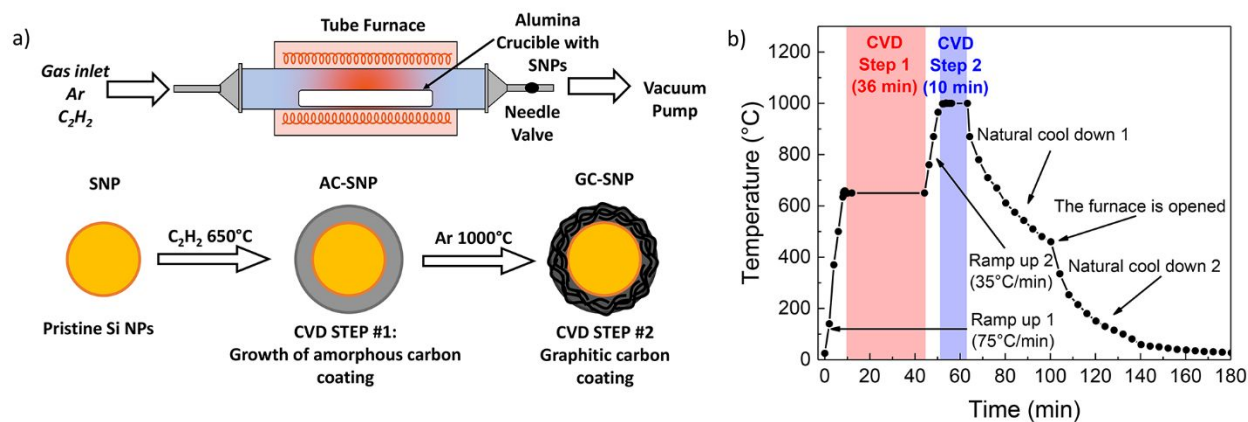
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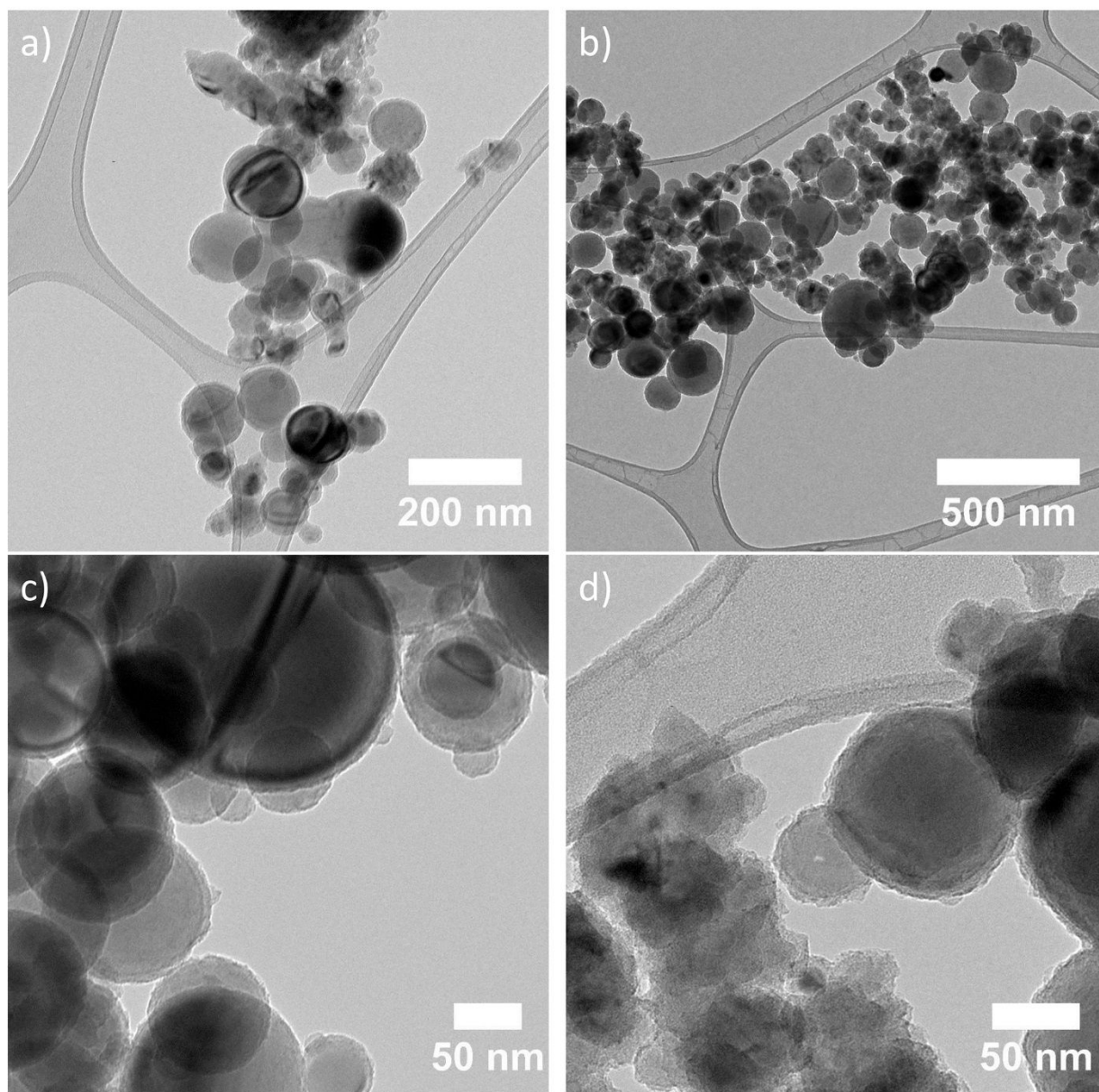
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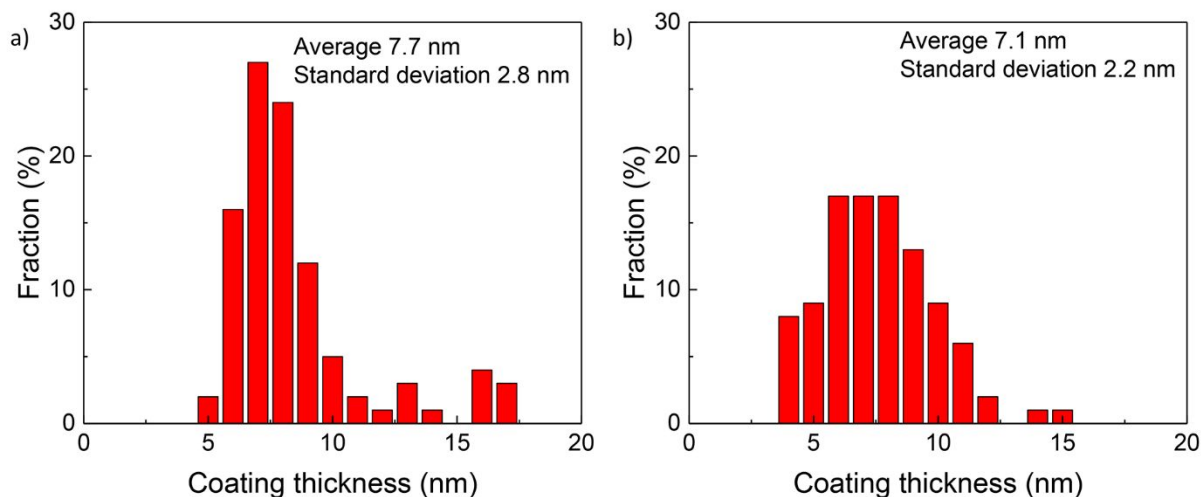
Keywords: Silicon, Amorphous Carbon, Graphitic Carbon, Chemical Vapor Deposition, Lithium-ion batteries, Additive.



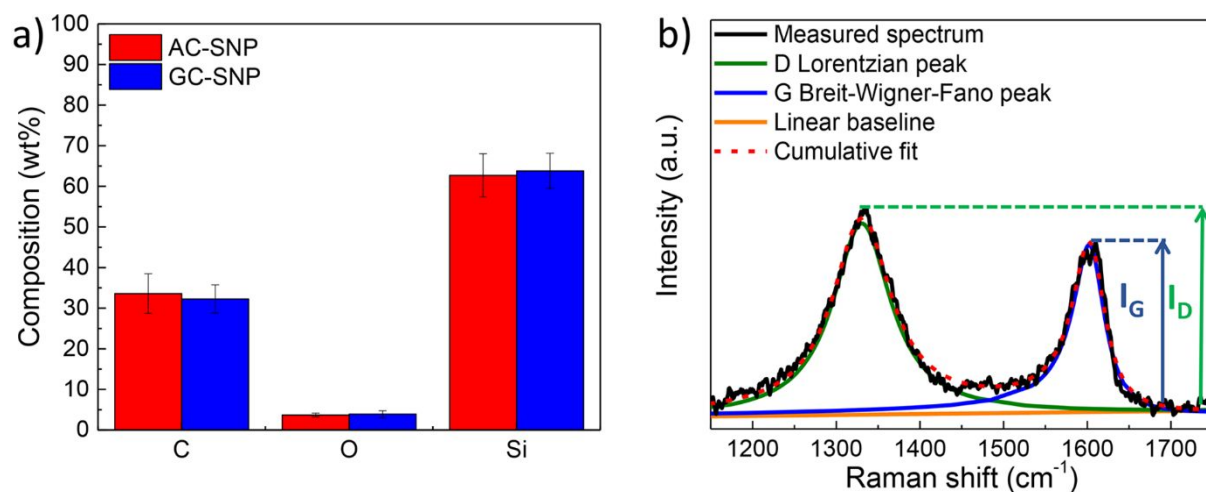
**Figure s1.** Schematic of the CVD setup used for the synthesis of the silicon-carbon composites and depiction of the effect of the two CVD steps on the carbon coating wrapping the Si particles (a). In the first low temperature step,  $C_2H_2$  is dissociated at moderate temperature (650°C), producing silicon particles wrapped with a conformal coating of amorphous carbon. This first composite is labelled as AC-SNP. In the second step the composite is annealed in Ar at high temperature (1000°C), inducing the complete graphitization of the carbon coating. This second type of composite is labeled GC-SNP. Temperature profile of the CVD process (b).



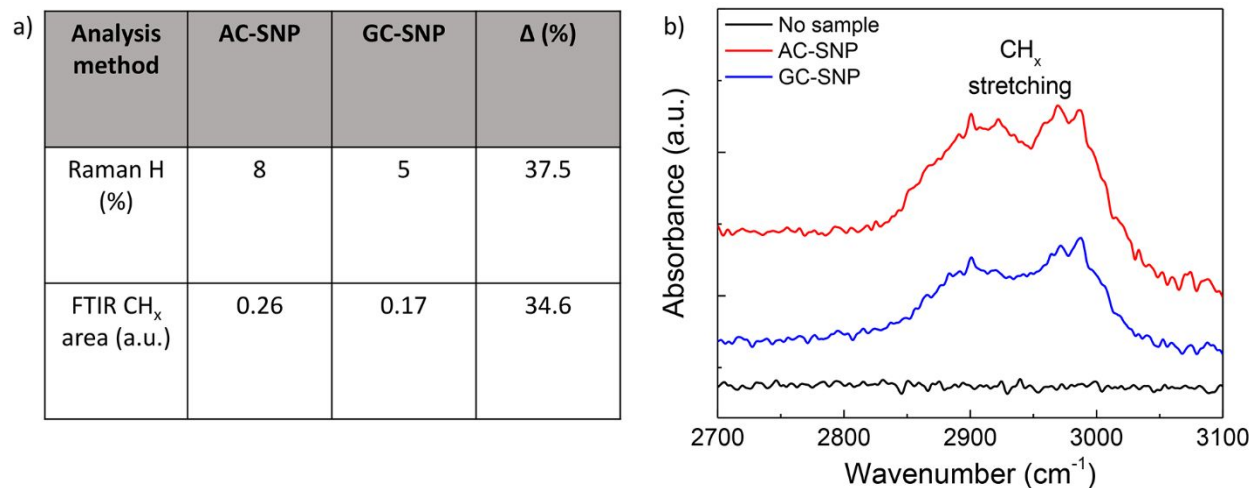
**Figure s2.** Low magnification TEM micrographs of AC-SNP (a, c) and GC-SNP (b, d).



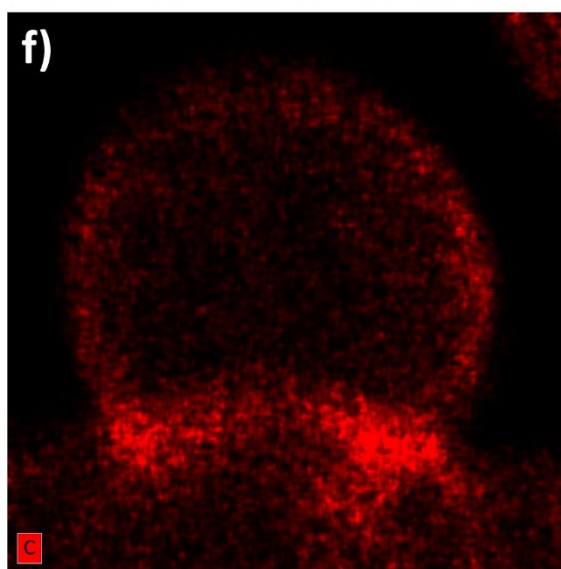
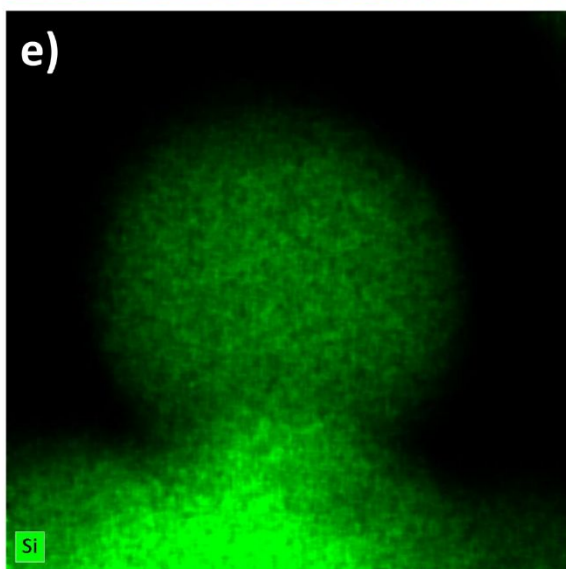
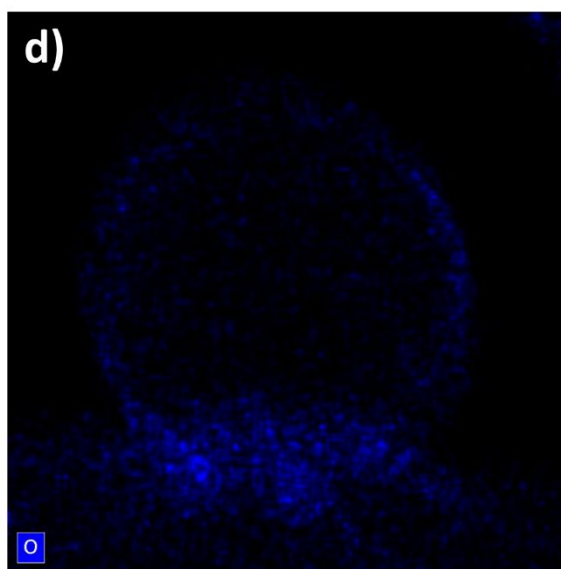
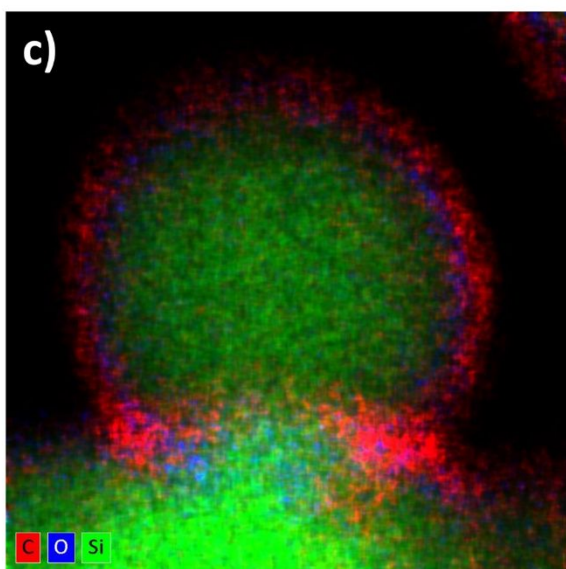
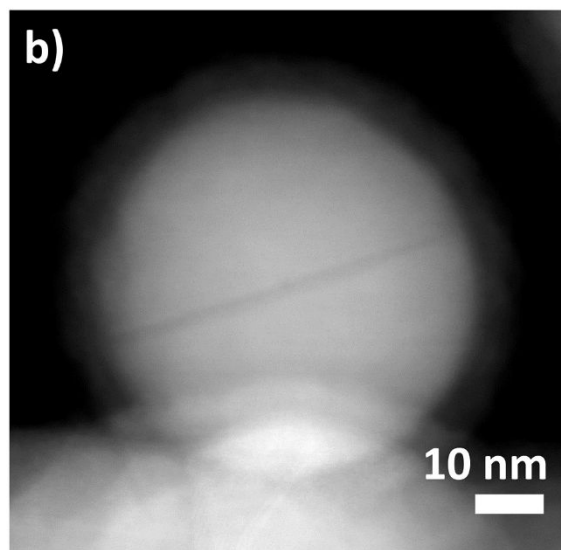
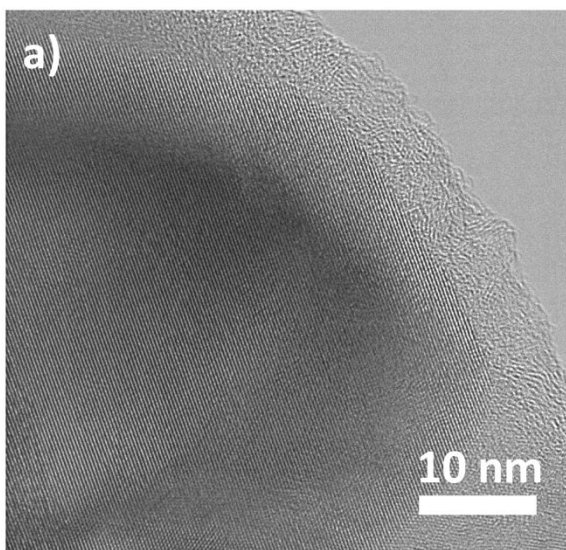
**Figure s3.** Distribution of the carbon coating thickness as measured by TEM analysis (statistical ensemble of 100 particles) for AC-SNP (a) and GC-SNP (b).



**Figure s4.** SEM/EDS of the average chemical composition of AC-SNP and GC-SNP (a). Raman fitting procedure employed for the analysis of the graphitization degree of the carbon coating in the synthesized composite materials (b). The fitting procedure encompasses a Lorentzian peak for the D mode, a Breit–Wigner–Fano (BWF) peak for the G mode and a linear baseline.

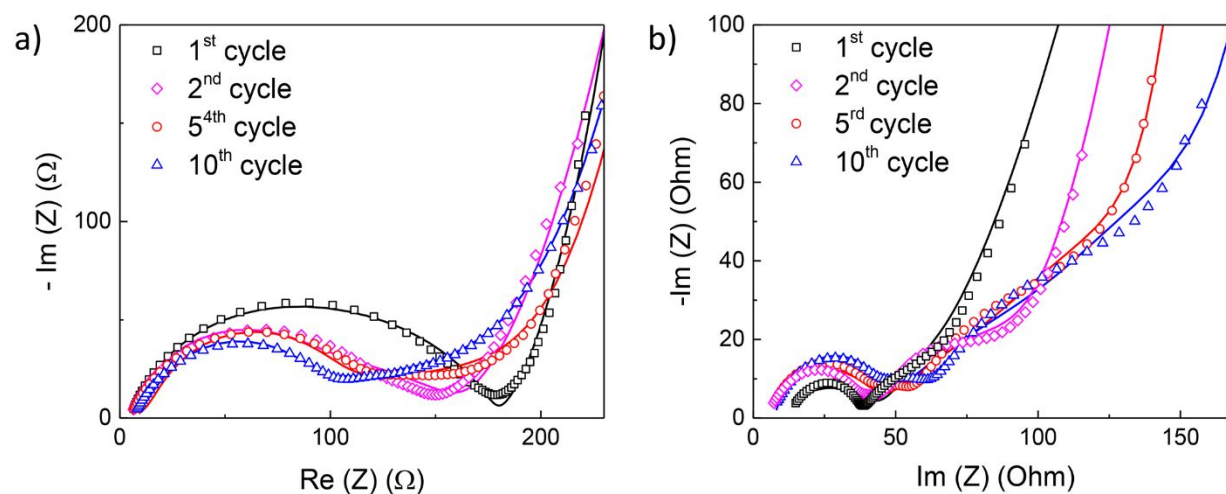


**Figure s5.** Analysis of the hydrogen content in AC-SNP and GC-SNP via Raman and FTIR analysis (a).  $\Delta$  is the difference between the hydrogen content of AC-SNP and GC-SNP expressed in percentage. FTIR spectra of AC-SNP and GC-SNP showing the CH<sub>x</sub> stretching mode (b).

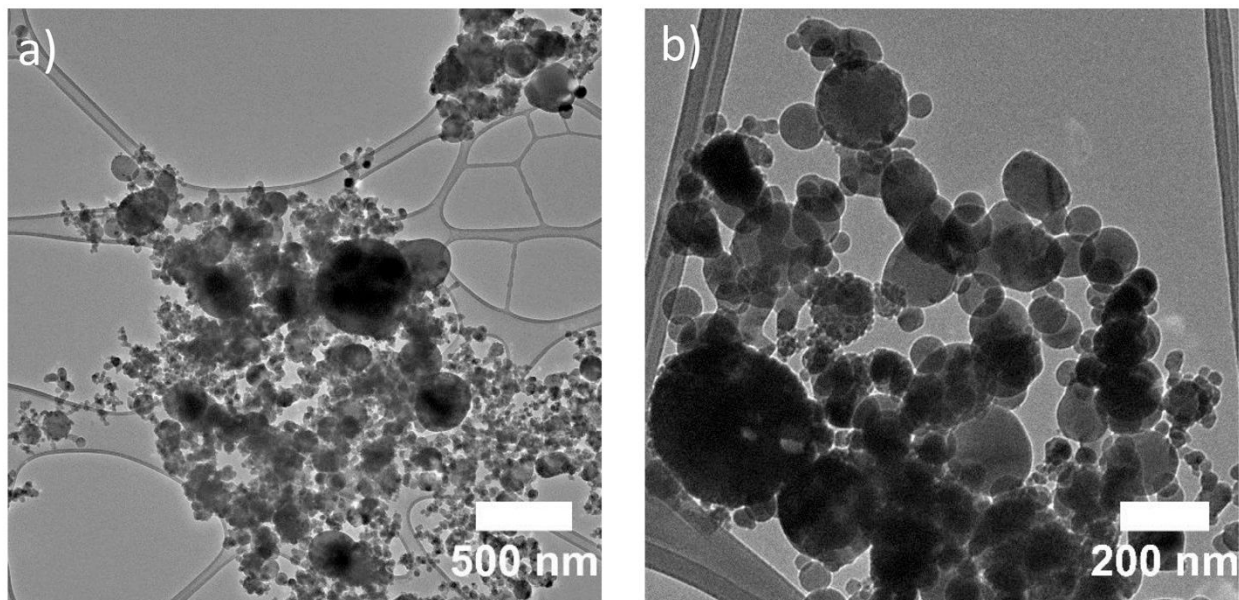




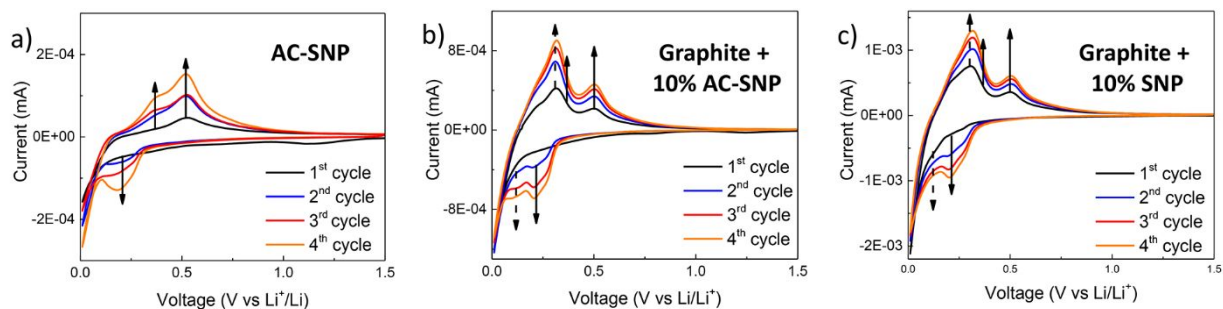
**Figure s6.** Bright-field STEM image of a single AC-SNP particle (a). The image highlights the neat interphase between silicon and carbon. Dark field STEM (b) and STEM-EDS micrographs of AC-SNP (c-f).



**Figure s7.** EIS analysis of AC-SNP (a) and GC-SNP (b) after the 1<sup>st</sup>, 2<sup>nd</sup>, 5<sup>th</sup> and 10<sup>th</sup> lithiation cycles. The fit for each dataset is represented by the corresponding solid line.



**Figure s8.** Low magnification TEM micrographs of pristine SNPs.



**Figure s9.** CV curves of electrodes fabricated with AC-SNP (a), graphite +10 wt% AC-SNP (b) and graphite +10 wt% SNP (c). The solid arrows show the lithiation and delithiation peaks of Si while the broken arrows show the lithiation and delithiation peaks of graphite.



