## **Supporting Information**

## Silicon-Nanoparticles Isolated by *In-Situ* Grown Polycrystalline Graphene Hollow Spheres for Enhanced Lithium-Ion Storage

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**Figure S1.** Schematic illustration of the influence of the amount of the catalyst precursor on the structure and degree of graphitization of the carbon shell: (a) At an appropriate FeSO<sub>4</sub> concentration, the discrete graphene nanoislands produced around each Fe nanoisland will interconnect to form a compact and uniform polycrystalline graphene shell throughout the Ag surface, (b) At a low FeSO<sub>4</sub> concentration, an amorphous carbon-rich shell is obtained due to the excessive carbon source, (c) At a high FeSO<sub>4</sub> concentration, the size of the *in-situ* formed Fe nanoparticles is too large to catalyze the growth of an integrated polycrystalline graphene shell. The obtained coating layer is in fact composed of a series of closely packed graphene encapsulated Fe nanoparticles. The blue spheres represent the Fe nanoislands (or nanoparticles).



Figure S2. (a) TGA thermogram of the Si@void@amorphous carbon nanocomposites, (b) Raman spectra of the SiNPs and Si@void@amorphous carbon nanocomposites and (c)  $N_2$  sorption isotherms of the pure SiNPs and Si@void@amorphous carbon nanocomposites.







**Figure S3.** TEM images of (a) Si@void@graphene and (b) Si@void@amorphous carbon electrodes after the initial ten cycles in their fully discharged (lithiated) state.



**Figure S4.** Reversible charge (delithiation) capacity and Coulombic efficiency versus cycle number profiles of the pure SiNPs electrodes.



Figure S5. SEM images of the Si@void@amorphous carbon electrode after 600 cycles.