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Strain-tolerant High Capacity Silicon Anodes via Directed Lithium Ion Transport for High Energy Density Lithium-ion Batteries

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Energy storage is an essential component of modern technology, with applications including public infrastructure, transportation systems, and consumer electronics. Lithium-ion batteries are the preeminent form of energy storage when high energy / moderate power densities are required. Improvements to lithium-ion battery energy / power density through the adoption of silicon anodes–with approximately an order of magnitude greater gravimetric capacity than traditional carbon-based anodes–have been limited by ~300% strains during electrochemical lithium insertion which result in short operational lifetimes. In two different systems we demonstrated improvements to silicon-based anode performance via directed lithium ion transport. The first system demonstrated a crystallographic-dependent anisotropic electrochemical lithium insertion in single-crystalline silicon anode microstructures. Exploiting this anisotropy, we highlight model silicon anode architectures that limit the maximum strain during electrochemical lithium insertion. This self-strain-limiting is a result of selecting a specific microstructure design such that during lithiation the anisotropic evolution of strain, above a given threshold, blocks further lithium intercalation. Exemplary design rules have achieved self-strain-limited charging capacities ranging from 677 $mAhg^{-1}$ to 2833 $mAhg^{-1}$. A second system with variably encapsulated silicon-based anodes demonstrated greater than 98% of their initial capacity after 130+ cycles. This anode also can operate stably at high energy/power densities. A lithium-ion battery with this anode was able to continuously (dis)charge in 10 minutes, corresponding to a power / energy density of ~1460 W/kg and ~243 Wh/kg–up to 780% greater power density and 220% higher energy density than conventional lithium-ion batteries. Anodes were also demonstrated with areal capacities of 12.7 mAh/cm^2, two orders of magnitude greater than traditional thin-film silicon anodes.

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