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Conductive Polymer and Silicon Composite Secondary Particles for a High Area-Loading Negative Electrode

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The recent studies based on thickness of the Si electrode have shown that area-specific capacity is inversely related to the cycling performance. As the Si area loading in the electrode gets higher, the cycling performance decays significantly. This phenomenon is observed in all the Si-based and other active materials based electrodes, but more pronounced in Si-based electrodes. Both the ion-transport distance and tortuosity of the pores in the composite secondary electrode increase as the electrode's thickness increases. Therefore, lithium-ion diffusion is impeded with the increase of electrode thickness. Therefore, it is important to isolate the impact of Si volume change within micron-size domains, so as not to affect the overall electrode micro-structures. This is achieved by the following steps. First, form micron-size Si nanoparticle/conductive polymer composite secondary particles (**Fig. 1**); then build in porosity in the composite secondary particles, so the Si nanoparticle volume change during lithiation can be accommodated within the composite secondary particles (i.e., there is no volume change in the composite secondary particle level). Finally, assemble the composite secondary particles into a regular porous electrode, with pores among the composite secondary particles (**Fig. 2**).

A sonication spray-precipitation method was used to generate spherical composite secondary particles. The porosity within the composite secondary particles can provide dimensional stability in the composite secondary particle level, while the primary nanoparticles can freely expand and contract. This dimensional stability of the composite secondary particles maintains porosity among the composite secondary particles, to ensure stable ion-transport properties in the electrode level. This will allow the fabrication of higher area loading of Si electrode. Spherical composite secondary particles have an average particle size of 10 μm in diameter, as seen in the scanning electron microscope (SEM) image of the particles collected after the spray-precipitation from the methanol non-solvent.

The different Si loading electrodes based on the composite secondary particles were fabricated and tested in a coin cell with lithium metal as a counter electrode and reported in **Fig. 3**. The capacity retention during the initial cyclings of the electrodes shows the improved stability. The electrode design based on composite secondary particles, the capacity decay is minimum at moderate loading. Even at high loading of 4 mAh/cm2, the electrode made with composite secondary particles can still cycles at a small capacity fade.

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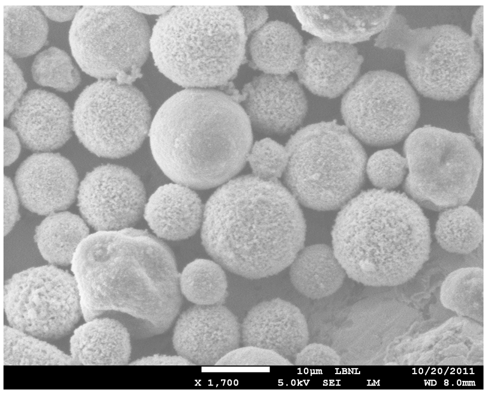

Fig. 1. Si/conductive polymer secondary composite particles made from spray precipitation process.

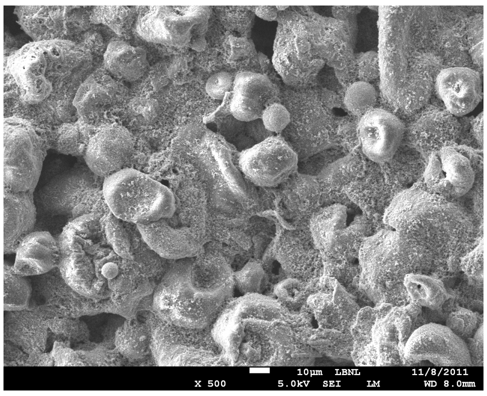

Fig. 2. Electrode made with secondary composite particles.

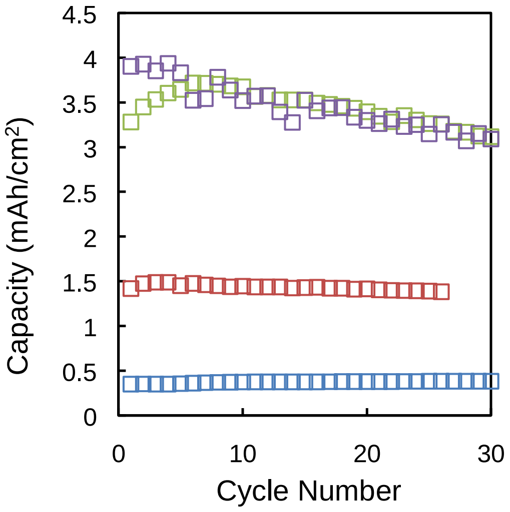

Fig. 3. The cycling performance of the Si electrode made with the secondary composite particles.