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Silicon Core Carbon Shell Nanoparticles By Scalable Laser Pyrolysis for Li-Ion Alloy Anodes – Material Synthesis and Performance Characterization

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As the world moves away from distributed fossil fuel use in order to mitigate the climatic effects of carbon pollution, the need for high energy density storage devices continues to grow. Secondary lithium ion batteries (LiB) are one such attractive energy storage device. Current LiB technology relies on graphitic carbon as the anode material, with a theoretical capacity of 372 mAh/g. In order to increase the energy density of LiBs, anode materials with a greater capacity for lithium storage are under intense investigation. Materials which form alloys with lithium such as antimony, germanium, silicon, and tin, all have theoretical capacities which far surpass graphite. However silicon, as the most naturally abundant element and possessing a theoretical capacity of 3579 mAh/g in the $\text{Li}_{15}\text{Si}_4$ alloy, is the most promising for global adoption in next generation LiBs.

There are issues which require resolution before silicon can be implemented. Large volumetric changes associated with the lithiation-delithiation process (~300%) result in material pulverization and loss of electrical contact [1]. Also unstable solid-electrolyte-interphase (SEI) formation during cycling results in the consumption of lithium during operation and capacity fade [2]. Previous studies have conclusively shown that the former issue may be mitigated by utilizing nano-scale silicon materials, with particles under 150 nm in diameter remaining intact during the swelling and contraction associated with cycling [3]. It has also been demonstrated that by encapsulating the silicon materials in carbon shells shows promise in stabilizing the SEI [4].

Here we present a scalable process to achieve this core-shell morphology via laser mediated pyrolysis. The technique, which has been used to produce various ceramic, oxide, and metallic particles [5], has already been utilized on the industrial scale for silicon nanoparticle production. Previously our group demonstrated the capacity of crystalline silicon core-carbon shell materials, synthesized in a two stage pyrolysis reactor, reaches ~500 mAh/g and retains over 70% capacity at a fast 2C rate over 500 cycles [6]. Amorphous silicon, with isotropic expansion upon lithiation, holds promise in forming a more stable SEI than crystalline silicon, and hence increased capacity retention. We have tuned pyrolysis reaction parameters in order to obtain consistent production of amorphous silicon nanoparticle cores. In this talk, a comparison of the battery testing results for amorphous vs. crystalline silicon cores will be presented. Steps to overcome present challenges with the cyclability and irreversible capacity loss due to SEI formation will also be discussed.

References

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