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Laser Pyrolysis Derived Silicon-Carbon Core-Shell Nanomaterials for Lithium Ion Batteries

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. Highly rational silicon-carbon architectures have been developed to accomplish this, such as the carbon clamped hollow silicon nanosphere, “yolk in shell”, and pomegranate geometries [4] which achieve high capacity and robust performance over hundreds of cycles. Guided by these achievements, we seek to develop a scalable process to synthesize silicon particle in carbon shell structures which may enable robust battery anodes with enhanced energy storage capacity.

Specifically we are developing a laser mediated pyrolysis, which has the capability to produce kg/h of high purity nanoparticles within a narrow size distribution. This technique has already been used to produce various ceramic, oxide, and metallic particles [5], and utilized for industrial scale silicon nanoparticle production. In this work we present a novel two stage pyrolysis reactor which synthesizes silicon nanoparticles in the first stage and adds a nanometric carbon shell in the second stage. The technique avoids any manipulation of the pure silicon powders prior to carbon coating, mitigating oxidation and particle degradation as well as worker exposure to nano-powders. Furthermore we discuss techniques to control particle size and coating thickness in order to optimize material performance. The capacity of current crystalline silicon core-carbon shell materials reaches ~2500 mAh/g at C/10 and retains over 70% capacity at a 2C rate over 500 cycles [6]. As amorphous silicon has been demonstrated to have improved fracture behavior, lithiation kinetics and first cycle performance [7,8], methods to produce amorphous silicon particles and their improved performance will also be discussed. Physico-chemical characterization of these materials and battery performance along with steps to mitigate first cycle irreversible behavior are to be presented.

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