Laser pyrolysis synthesis of nanoparticles for energy applications

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The laser pyrolysis method has proved efficient for the synthesis of various oxide and non-oxide nanoparticles such as SiC, Si, TiO_2... Recently we have developed the method for the one-step production of Si@C core shell nanoparticles and composites of TiO_2-carbon nanostructures.

Much attention has been paid to Si as an anode material in Li-Ion batteries in order to improve storage capacity (the theoretical limit being 3579 mAh/g in the $Li_{15}Si_4$ alloy vs 372 mAh/g for graphitic carbon). However, due large volume changes during cycling, silicon suffers several drawbacks, including rapid pulverization and SEI ripening, limiting its use. Nanostructuration and protection of silicon by a carbon coating are proven methods to improve the behavior of the silicon based anodes [1]. We have developed a two stage laser pyrolysis reactor to achieve the synthesis of silicon-carbon core-shell nanoparticles in a continuous way, without intermediate manipulations between the synthesis of the core and the shell [1]. At lab scale, the reactor is capable of stable run times up to several hours with production rates of ~8 -10 g/hour. The size of the core was tuned from 20 to 80 nm diameter with associated carbon content of up to 20 wt%. EIS spectroscopy clearly shows the influence of the carbon coating on the growth of SEI during the first cycle, with a significantly lower resistance of the SEI when Si@C nanoparticles are used by comparison with Si. The carbon coating also enables improved utilization of silicon content in the first cycles as shown by galvanostatic cycling. By adding germane in the reaction zone, we were also able to synthesize nanoparticles of Si_xGe_{1-x} with x in the range 20-80. These particles also present a core@shell organization with a silicon shell at the surface of the alloy particle. These NPs present improved columbic efficiency and stability when tested in coin cells.

In the context of transitioning to renewable energy sources, development of efficient and cost-effective solar cells is a major objective to establish an optimal energy mix. The 3rd generation of photovoltaic cells emerged to develop high efficient and low-cost cells combining the use of abundant materials and easy processes. Among them, photovoltaic cells based on perovskite materials demonstrate several significant advances with power conversion efficiencies up to 22% [3]. Nevertheless, efforts continue to improve the charge generation and collection in this kind of cell. Titanium dioxide mesoporous layers, while remaining an important component for perovskite structuration and electron transport in these high efficiency devices, is responsible of charge trapping and recombination, representing two major loss mechanisms. As carbon nanostructures are good electron transporters, the use of TiO_2/graphene nanocomposites with a clean interface between carbon and TiO_2 seems to be a promising strategy to reduce recombination phenomena and thus improve electron collection [4].

To achieve high quality of nanocomposites presenting well-controlled physical properties suitable for efficient and stable solar cells, we use laser pyrolysis, which enables the synthesis of nanoparticles in a single step with a continuous flow. This technique already proved efficient for the production of TiO_2 nanoparticles in anatase phase [5]. By adding graphene in the precursor, it was possible to obtain TiO_2/graphene nanocomposites where TiO_2 is strongly attached to the surface of graphene. Tests were conducted with a chlorine-doped methylammonium lead iodide (MAPI-Cl) reference perovskite deposited in a single-step, on TiO_2 porous electrodes composed of laser pyrolysis derived materials following a previously reported procedure [6]. Our first results show a better electron injection efficiency from the perovskite layer to the mesoporous graphene-doped TiO_2, as revealed through steady-state photoluminescence spectroscopy. Larger photocurrents and smaller series resistances are also observed for these devices, under standard illumination in the presence of graphene. More generally an increase in power conversion efficiency from 14.1 % to 15.1 % for these devices is achieved for perovskite solar cells containing graphene in the mesoporous layer, demonstrating the benefit of the laser pyrolysis process for the production of high quality electron transport layers.

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