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► To cite this version:

J P Alper, F Boismain, J Sourice, W Porcher, E Foy, et al.. Laser pyrolysis for the controlled synthesis of amorphous or crystalline Si@C nanoparticles - Material Synthesis and Performance Characterization in Li-Ion batteries. EUROMAT 2017, Sep 2017, Thessalonique, Greece. cea-02342073

HAL Id: cea-02342073

<https://cea.hal.science/cea-02342073>

Submitted on 31 Oct 2019

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Laser pyrolysis for the controlled synthesis of amorphous or crystalline Si@C nanoparticles -
Material Synthesis and Performance Characterization in Li-Ion batteries

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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. In Lithium Ion Batteries, the replacement of graphite (372 mAh.g⁻¹) as the anode active material by higher specific capacity materials is a strategy to meet the continuous demand for increased energy storage. Silicon appears as an attractive material because of its high theoretical specific capacity (3579 mAh.g⁻¹).

However use of silicon based anodes has not yet been commercially realized because of rapid performance degradation due to mechanical instabilities and large volume changes upon lithiation/delithiation. Silicon nanostructuring and encapsulation in carbon have been shown to increase cycle life. In this context, core-shell silicon-carbon nanoparticles (Si@C) are promising candidates for application as LiB anode materials.

Such nanoparticles were synthesized in a reactor by a continuous gas phase method, laser pyrolysis. We report here how flow simulations helped in the design of a reactor where decomposition of silane and ethylene are conducted in two successive reaction zones to produce silicon cores in the first zone and coat it with carbon in the second zone. The reactor design enables stable reactor conditions leading to the scalable single-step synthesis of amorphous or crystalline silicon nanoparticles coated with a carbon shell (a-Si@C). The advantages of the a-Si@C material are demonstrated by comparison with c-Si@C material. In particular, cyclic voltammetry demonstrates that a-Si@C composite reaches maximal lithiation during the first sweep, which is attributed to the amorphous core. After 500 charge/discharge cycles, it retains a capacity of 1250 mAh.g⁻¹ (C/5 rate) and 800 mAh.g⁻¹ (2C), with a 99.95% coulombic efficiency. Moreover, post-mortem observations prove formation of a limited solid electrolyte Interphase with an electrode expansion of less than 20% in volume and preserved nanostructuring within the electrode.