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Laser pyrolysis : a method of interest for the controlled synthesis of amorphous or crystalline Si@C nanoparticles - application as anode materials in Li-Ion batteries

John Alper^{1,2}, Florent Boismain¹, Julien Sourice^{1,2}, Adrien Boulineau^{2,3}, Aurélie Habert¹, Eric De Vito^{2,3}, Willy Porcher^{2,3}, Cécile Reynaud¹, Cédric Haon^{2,3}, Nathalie Herlin-Boime^{1}*

¹NIMBE, CEA, CNRS, Université Paris-Saclay, CEA Saclay 91191 Gif sur Yvette France

²CEA, LITEN, 17 rue des Martyrs 38054 Grenoble France

³Univ. Grenoble Alpes, 38000 Grenoble France

In Lithium Ion Batteries, the replacement of graphite (372 mAh.g⁻¹) as anode active materials by higher specific capacity materials is a strategy to answer the continuous demand for increased energy storage. Silicon appears as an attractive material thanks to its high theoretical specific capacity (3579 mAh.g⁻¹). Use of silicon based anodes has not yet been realized because performances degrade rapidly. Silicon nanostructuration together with association of carbon enhances performances. In particular, core-shell silicon-carbon Si@C nanoparticles are attractive candidates to increase the capacity of Li-ion batteries while mitigating the detrimental effects of volume expansion upon lithiation processes.

Such nanoparticles were synthesized in a single step by a continuous gas phase method, the laser pyrolysis, interesting for industrial production. 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. This reactor could work in stable conditions for several hours leading to the 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 is emphasized 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 show an electrode expansion of less than 20% in volume with preserved nanostructuration.