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Silicon/biogas-derived carbon nanofibers composites: a promising anode material for lithium-ion batteries

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Introduction

Silicon appears to be a promising anode material for increasing both the energy density and power of lithium-ion batteries due to, mainly, the high theoretical specific capacity, the relatively low working potential and the abundance in earth crust. However, the lithiation/delithiation of Si causes successive volume changes of this material leading to the fracture of the particles and the consequent poor reversibility and cycling stability of the electrode. Among different strategies which are being developed to avoid the Si-electrode degradation, this work has focused on: (i) the preparation of Si-based composites by adding a matrix (specifically carbon materials) which can help to buffer the volume changes and (ii) the limitation of the lower cut-off voltage (LCOV) which leads to a better control of these changes.

Results and discussion

Among different carbon matrices studied in this work, free-metal carbon nanofibers obtained from the catalytic decomposition of biogas (BCNFs) result the most suitable ones. Thus, the fishbone microstructure of these carbon nanofilaments, having a great number of active sites to interact with the Si particles, buffer the volume changes associated with the silicon lithiation/delithiation, preventing the electrode degradation. Specifically, an nSi:BCNFs 1:1 weight ratio in the active composite is the optimal. On the other hand, the limitation of the LCOV in the 80-100 mV range avoids the formation of the highest lithiated phase which leads to electrode degradation. Both strategies allow achieving a compromise between the specific capacity, which is greater than that of graphitic materials currently used in LIBs, and acceptable capacity retention along galvanostatic cycling. Thus, electrodes formed by 80 % of the nSi/BCNFs composite, including a 10 % of carbon black (CB), and 20 % of sodium carboxymethyl cellulose (NaCMC) as active material and binder, respectively, has been prepared in this work by a simple, fast and easily industrial-scaling process, achieving specific capacities up to \sim 520 mAh g-1 after 30 cycles with coulombic efficiency > 95 % and \sim 94 % of capacity retention along cycling at an electric current density of 100/200 mA g⁻¹ and a LCOV of 80 mV (Fig. 1).

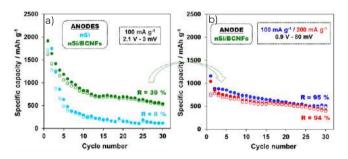


Fig. 1 Specific discharge (full circle)/charge capacity (empty circle) vs cycle number plots from the galvanostatic cycling of a) nSi and nSi/BCNFs composite at 100 mA g^{-1} in 2.1 V - 3mV potential range and b) nSi/BCNFs composite at 100-200 mA g^{-1} in the 0.9 V - 80 mV potential range