## New design of Si negative electrodes with copper foam as current collector

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## Introduction

Lithium-ion batteries (LIBs) are widely used for various mobile devices, such as cell phones, laptop computers, and recent automobiles, including hybrid vehicles, and electric vehicles (EVs), due to their higher energy density and lower self discharge rate than other rechargeable batteries. Their energy densities are however still not sufficient for most of the demands, especially for EVs. [1,2] Silicon (Si) attracts a great attention as a new negative electrode material for high-energy density LIBs due to its extremely higher capacity (3580 mAh g<sup>-1</sup>) than conventional graphite negative electrodes (372 mAh g<sup>-1</sup>). However, the commercialization of Si-based anode has been hindered due to its poor cyclability conductivity because of the huge volume variation (above 300%) that results in particles or electrode disintegration, and because of unstable SEI.

A lot of efforts have thus been made to solve the problems mentioned here above. Extensive studies on silicon thin films, silicon nanowires, nanoparticles, nanotubes, porous structures and optimizing electrolyte additives, binders, conductive additives have shown very encouraging results when used as anode materials in lithium ion batteries and remarkably improve the cycle performance of Si-based electrodes [1-5]. However, there still remain several technological hurdles that should be overcome such as higher areal active mass loading to reach typically more than 3 mAh/cm<sup>2</sup> surface capacity. The traditional of battery design with electrodes coated onto flat metal current collectors does not allow for highr mass loadings due to the following major difficulties: (1) thick electrodes tend to delaminate from the flat current collectors after the coating process. (2) The limitations in the lithium diffusion through a thick electrode which induced strong power penalties. The concept of three dimensional (3D) battery electrodes has been used previously to enhance the energy per footprint area. Here we adapt Cu foam as a replacement for metal foil current collectors to create 3D structuration of the composite electrode.

The silicon-based composite electrodes were prepared by casting of the electrode slurry into the porosity of the Cu foam. With such design, the mass loading can reach a value as high as 10 mg/cm<sup>2</sup>. Then its SEM characterization, electrochemical property and failure mechanism were studied by means of charging– discharging tests, and electrochemical impedance spectroscopy (EIS). In comparison with common 2D design, fabricated by slurry-coating technology on flat copper foil, the 3D copper framework (Figure 1) has shown a great advantage in the cycle life vs. the active material surface loading of. This good electrochemical performance can be attributed to the good conductivity and excellent ion transport from the electrolyte to the active material surface and also from the superior charge transport between current collector (Cu foam) and electrode materials (Figure 2).

## Experimental

The silicon electrodes were composed of 80wt% micro-Si (99.9%, 1–5 micron, Alfa Aesar), 12wt% Super P carbon black (noted CB, Timcal) and 8wt% carboxymethyl cellulose (CMC, (DS = 0.7, Mw = 90 000, Aldrich) and prepared by ball milling in an aqueous pH3 buffer. The slurry was cast on Cu foam (300 $\mu$ m thick, ~ 80% porosity, Metafoam) or onto a 25  $\mu$ m thick for copper foil. The electrode was dried at room temperature and then at 100°C in vacuum to remove the water solvent.

Electrodes were cycled either with a limited capacity of 1200 mAh/g of Si in a 1:1 EC/diethyl carbonate (DEC) with 10 wt% fuoroethylene carbonate (FEC) and 2 wt% vinylidene carbonate (VC); and a Li metal disc as the counter and reference electrode.

Fig 1: Top SEM micrographs of 3D silicon-based anode Cu foam without (a) and with mass loading of active materials (b)



Fig 2: Cycle number vs. Active mass of Silicon with Cu foil (green) and Cu foam (blue) as current collector.



## References

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