OPTIMIZING THE COATING METHOD FOR IMPROVED PROCESS CONTROL OF SILICON/CARBON COMPOSITE ANODES

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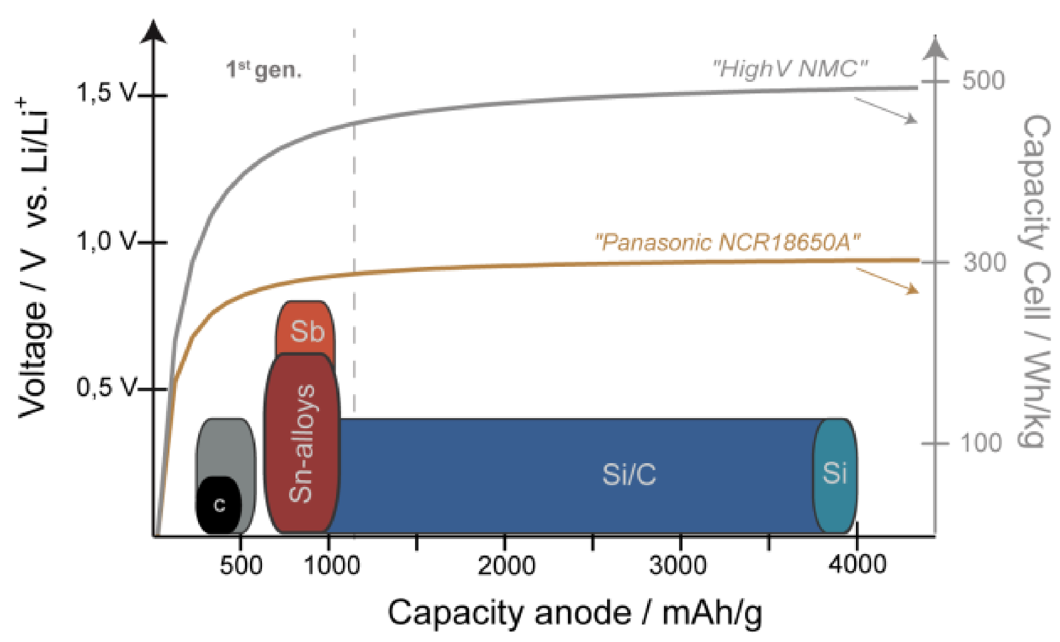
SILICON AS ANODE MATERIAL

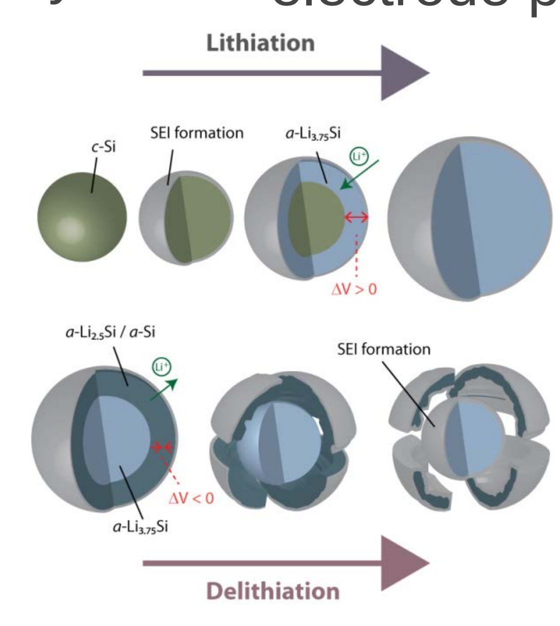
Li-ion batteries are a commercial battery technology with fast market growth. The introduction of silicon is expected to be the core of the next generation of Li-ion anodes, allowing higher battery capacity. Silicon expands during lithiation, leading to fast degradation, and limiting industrial use of silicon anodes. Biopolymer-based binder modifications are a very promising path towards longer life time of silicon-based Li-ion battery anodes, as well as the use of additives to control the SEI-formation and the following cycle stability.

PROCESS CONTROL

All components used in the Li-ion electrode and the interaction between them influence the resulting battery cell. To fully understand the result, we need to understand all the separate processes and control them. The “noise” of uncertainties in the preliminary stages of the process will be amplified as the process continues, and the noise reduction in the early phases is of great importance. Some of the parameters that are examined are the characteristics of Si, binder interactions, electrode properties and cycling parameters.



The theoretical capacity of silicon/carbon composites



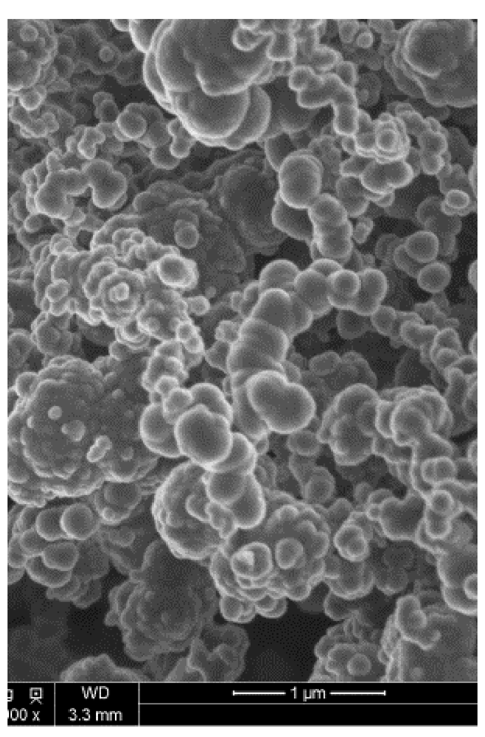
Challenges with Si: expansion upon lithiation and stability of SEI layer

Parameters that affect the process control of Si/C composite electrodes

SILICON SOURCE

The silicon particles was synthesized in a free-space reactor via a silane-based decomposition.[2]

By controlling the silane gas temperature (500-600 °C) and pressure, a suitable size distribution as well as production yield of up to 350 g/hour was obtained.

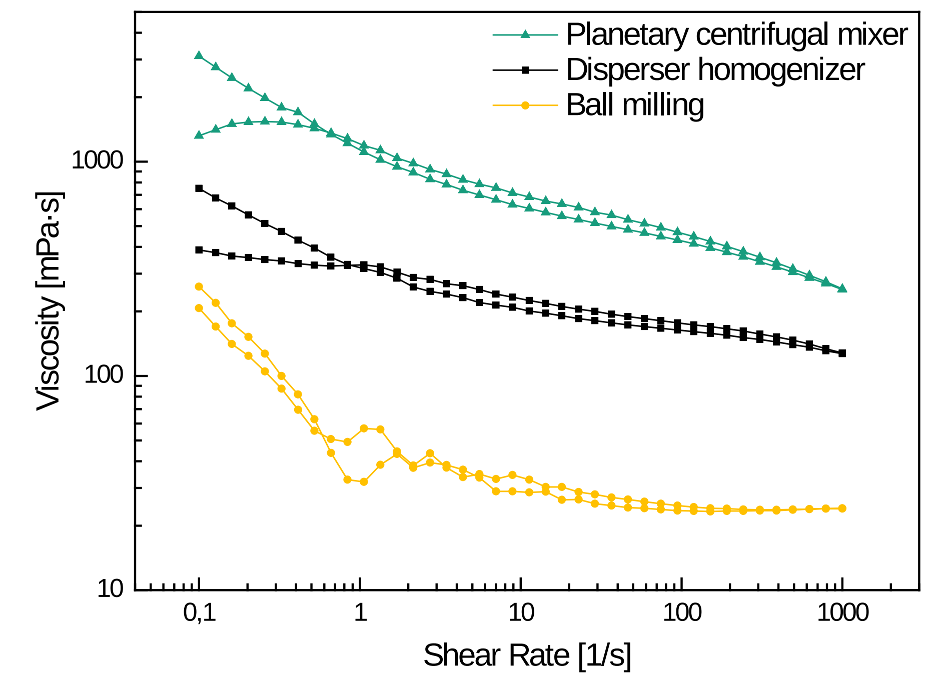


SEM image of silicon particles

MIXING METHOD

A qualitative study of three mixing methods revealed large differences in rheology properties as well as ease-of-preparation and mixing time needed to get optimal result.

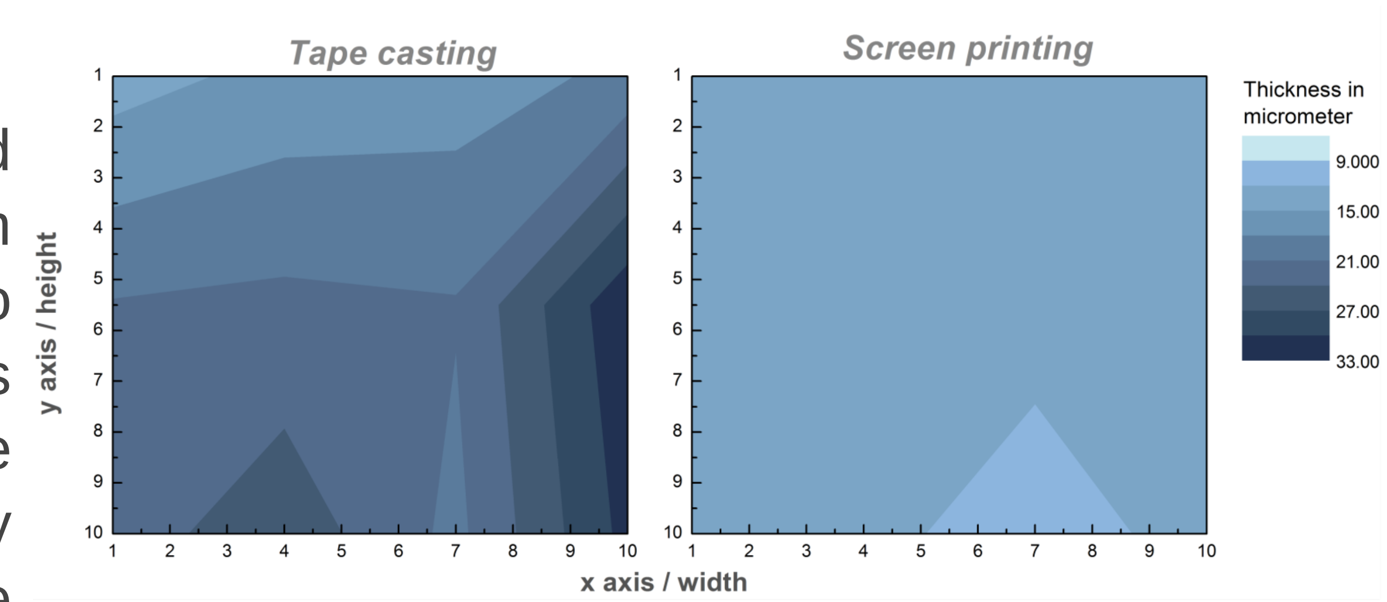
Mixing the slurry in a planetary centrifugal mixer (Thinky) gave a higher viscosity at all shear rates than with a disperser homogenizer (UltraTurrax) or with a ball miller (Fritsch).



Rheology measurements of slurries prepared by different mixing methods: large variations in viscosity will result in variances of the slurry stability and sedimentation rate. The planetary centrifugal mixer gives a higher viscosity which can be correlated to a higher slurry stability. [1]

COATING METHOD

The electrode properties depend strongly on its morphology and homogeneity. For example, the lifetime of Si/C anodes are very sensitive with regards to loading. By changing the coating method from tape casting to screen printing, a more homogeneous electrode is obtained. Both thickness and loading variations are improved with coating via screen printing. The overall loading of the electrode can be controlled by the viscosity of the slurry as well as the screen parameters. Screen parameters are mesh size, wire thickness and emulsion thickness, where the latter affects the resulting loading more strongly.

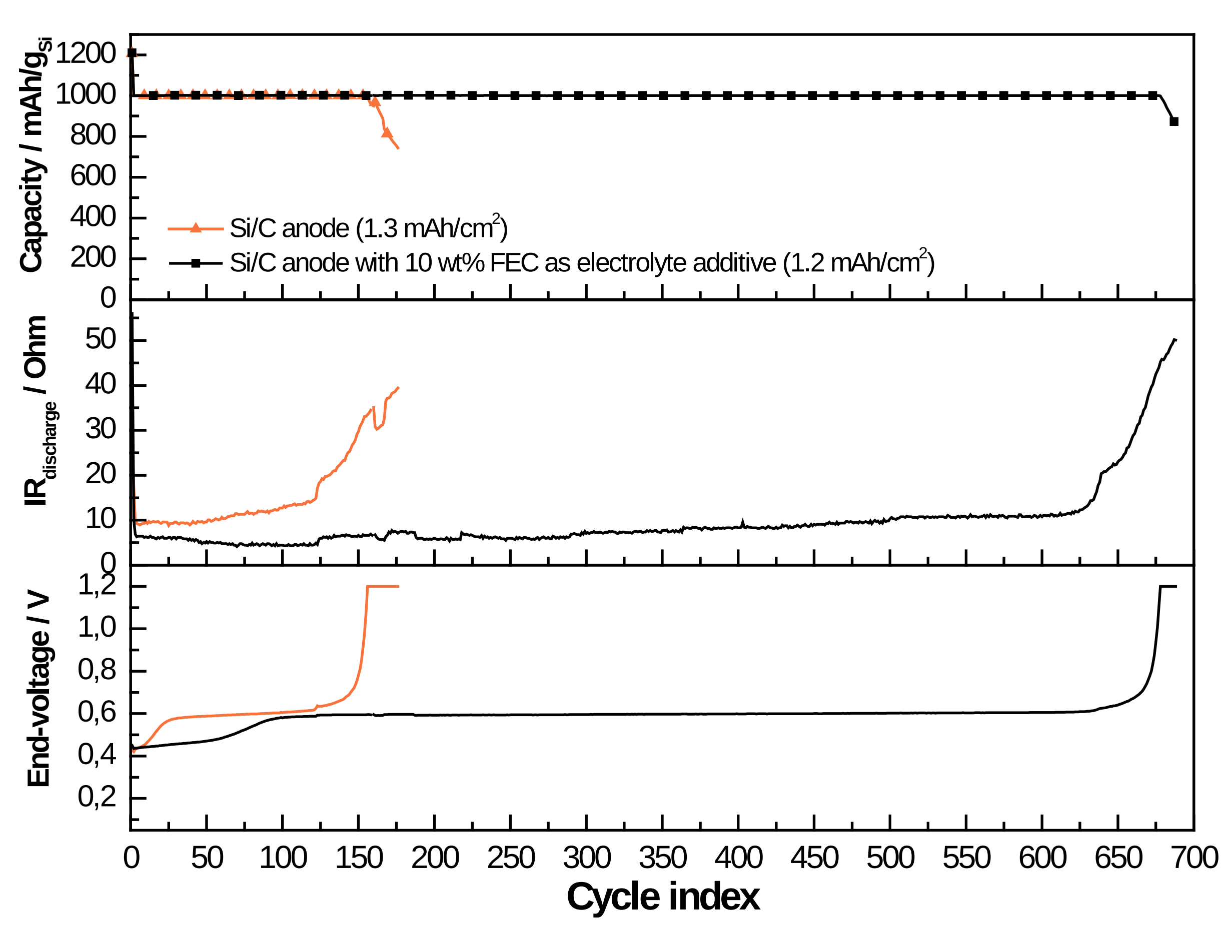


Thickness of a tape casted electrode and a screen printed electrode measured along the x- and y-axis. The homogeneity of the thickness is greatly optimized by coating via screen printing. The thickness can be increased by screen choice and slurry viscosity.

ELECTROCHEMICAL TESTING

Electrochemical performance of the Si/C-anodes was tested by galvanostatic cycling using CR2032 coin-cell-type cells. The capacity was limited to 1000 mAh/g_{Si}. Working electrodes were made by tape-casting on Cu-foils and measured against Li metal counter electrodes. The electrolyte used was 1 M $LiPF_6$ in 1:1 EC/DMC with/without electrolyte additive (10 % FEC).

The slurries were made with 60 % Si, 15 % carbon black, 10 % graphite, 15 % binder (CMC) and a buffer solvent to control the pH of the slurry.



Electrochemical cycling of Si/C electrodes prepared by screen printing. The use of FEC as an additive gives great improvement in cycle life.

[1] B. Bitsch et al. Journal of Power Sources, 265 (2014), 81-90,
[2] H. F. Andersen, H. F. et al. ECS Transactions. 62, (2014), 97–105.

CONCLUSIONS

Process control

- The homogeneity of the finished electrode can be optimized by increased process control.
- The work focuses on the use of mixing method and coating procedure for Si/C anodes.

Electrochemical testing

- Half cells with Si/C anodes were pre-pared and cycled at a limited capacity of 1000 mAh/g_{Si} for over 650 cycles.

ACKNOWLEDGEMENTS

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