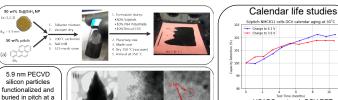


Nanoparticle Surface Passivation Strategies to Improve the Cycle and Calendar Lifetime of Silicon Anodes in Standard Carbonate Electrolytes

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Oligomer/SiO_x Passivation PEO PECVD silicon particles (5.9 nm diameter) covalently functionalized with PEC for colloidal stability to improve slurry processing and printing Si@PEO || Li

sp² Amorphous Carbon Passivation



USABC protocol: OCV-RPT Si@Pitch II NMC811 1.5 mAh/cm²

- Electrode and cycling details: 8:1:1 Si@PEO:P84:C45
- Cured at 420 °C for 4 hrs under N
- Electrolyte: 1.2M LiPF₆ in 3:7 EC:EMC with 3 wt.% FEC (GenF)
- Full cells were electrochemically prelithiated to 18% SOC
- C/3 cycling with intermittent C/10
- Voltage window 3.0-4.2 V
- All measurements at 30° C

Conclusions

- 1. High silicon content electrodes with intermediate loading show stable cycling (CE >99.9%) in standard carbonate electrolyte.
- Calendar life > 10 months

Calendar life studies

Aging Time @ 30°C (months)

USABC protocol: OCV-RPT

Si@PEO (420 C) || NMC811

Carroll et al. 2023. J. Mat. Chem A: DOI: 10.1039/D2TA08935A

Electrode and cycling details:

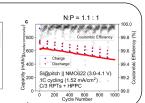
50 wt.% ratio

Si-Pitch

8:1:1 Si@Pitch:P84:C45 Cured at 350 °C for 4 hrs under N₂ Prelithiated to 8% SOC

All cycling data collected conditions as Si@PEO

0.6 0.4 0.2

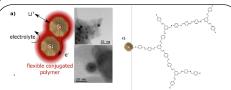


Conclusions

- 50/50 blend of pitch and silicon provides a capacity > 1000mAh/g
- Stable cycling (CE > 99.9%) at low loading
- Calendar life > 10 months

Schulze et al. manuscript in preparation

Conducting Polymer Passivation



PECVD covalently functionalized with hyperbranched polyphenylmethanimine (PolyPMI) to bury Si interface in a mixed electronic and ionic conductor

0.4 60 Cycle number Particle Diameter (nm)

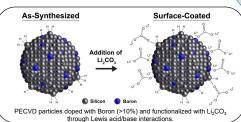
Plasma Enhanced Chemical

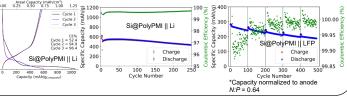
Vapor Deposition (PECVD)

Silicon Nanoparticles

- Highly controllable synthesis with surface termination (-SiH_x) for easy covalent or ionic functionalization
 - In Li-ion batteries, particle sizes dictate the cycle stability of silicon anodes

Ionic Passivation





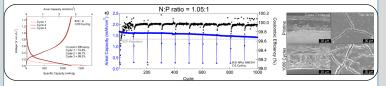
Electrode and cycling details:

- 6:2:2 Si@PolyPMI:P84:C65
- Cured at 350 °C for 4 hrs under N₂ Cycling data collected at 1C
- No electrochemical prelithiation All electrolyte is GenF

Conclusions:

- Silicon embedded and covalently attached to hyperbranched
- CE reaches values > 99.99%
- SEI remains unchanged between 1 and cycle 200
- Calendar life measurements are on going

Martin et al. 2023, Adv. Ener. Mat. : DOI:10.1002/aenm.20220392



Electrode and cycling details:

- 8:1:1 B:Si:P84:C45
- Cured at 400 °C for 4 hrs under N.
- Cycling data collected at C/3 in GenF electrolyte
- Full cells assembled from electrochemically prelithiated anodes (1%< SOC)

Conclusions:

- Boron-doped PECVD silicon particles are functionalized with Li₂CO₃ through strong acid/base interactions
- Highly compact electrode morphology Electrochemical cycling against NMC 811 displays capacity retention > 84% through 1000 cycles
- Calendar life measurements are on going

Pach et al. Manuscript in Preparation