Electronic Supplementary Material (ESI) for RSC Advances.

Supporting information for "Binder-free lithium ion battery electrodes made of silicon and pyrolized lignin"

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Experimental

Fabrication of Si-pLig binder-free electrode

Hardwood Kraft lignin (Lig) was obtained from Mead Westvaco. Kraft lignin was first mixed with 0.5wt% polyethylene oxide (PEO, $1x10^6$ MW, Sigma Aldrich) by spatula, then the powder mixture was dissolved in dimethylformamide (DMF, Sigma Aldrich) with a weight ratio of 15%. The solution was heated to 60 °C under constant magnetic stirring for 2 hours. Afterwards, silicon nanoparticles were added to the solution with a weight ratio of 15% to DMF. The solution was magnetically stirred for 6 hours at 60 °C while going through sonication for 5 mins every 2 hours. A Mazerustar KK-250 planetary mixer was also used every 2 hours to

ensure adequate mixing. The solution was stirred and heated until reaching a suitable viscosity for slurry coating onto copper foil by a doctor blade with a thickness of 127 µm. After drying in air, the coated copper foil was dried under vacuum at 120 °C overnight (silicon nanoparticle-lignin composite prior to pyrolization is denoted as Si-Lig) to obtain a uniform Si-Lig film.

The dried Si-Lig on copper foil was pyrolized in a tube furnace with argon atmosphere at a step rate of 2 °C /min up to 800 °C, then held for 2 hours (pyrolized Si-Lig is denoted as Si-pLig) and then naturally cooled down. For making electrodes, 10 mm disks were cut and stored to be assembled into coin cells, with an average silicon mass loading of 1.4 mg cm^{-2} (post-pyrolization mass).

For comparison, silicon negative electrode using polyvinylidene fluoride (PVDF, Alfa Aesar) as the binder was prepared using conventional slurry coating method. The mass ratio of silicon, carbon additive (Super C65, Timcal), and respective binder was 3:1:1. PVDF was first dissolved in N-Methyl-2-pyrrolidone (99.5%, Alfa Aesar) solvent, then silicon nanoparticles and carbon black were mixed in the PVDF solution until a uniform slurry with reasonable viscosity was obtained. Finally, the slurry was coated onto copper foil by a doctor blade with a thickness of 127 µm.

Characterizations

Scanning electron microscopy (SEM) was performed using a Hitachi S-4300 microscope with a 15 kV voltage in the imaging mode. The structure and morphology of the Si-pLig composite electrode were examined with a JEOL 2010F transmission electron microscope (TEM). The amount of Si in the Si-pLig composite was determined by thermal gravimetric analysis (TGA) (TA Q500 in high-resolution dynamic mode) in the air. The samples were heated to 800 ˚C at a constant rate of 10 ˚C/min.

Electrochemical measurement

For electrochemical performance of Si-pLig/Si-PVDF-Super C65 on copper current collectors, coin cells (CR2025 type) were assembled in an argon filled glove box, using Li metal foil as the counter/reference electrode, Celgard 3501 membrane as the separator, and 1M $LiPF_6$ in ethylene carbonate and diethyl carbonate (EC: DEC=1:1 vol%, BASF) with 10wt% fluoroethylene carbonate (FEC) additive as the electrolyte. Electrochemical tests were performed using a Bio-Logic potentiostat (VMP-3) at room temperature. All specific capacities presented were calculated based on the weight of Si in the composite. TGA analysis was used to determine the weight percent of Si in the Si/C composite.

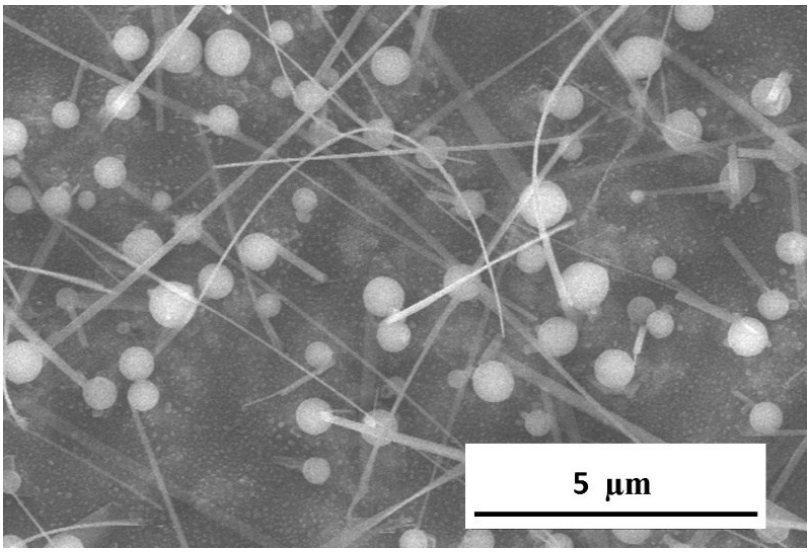
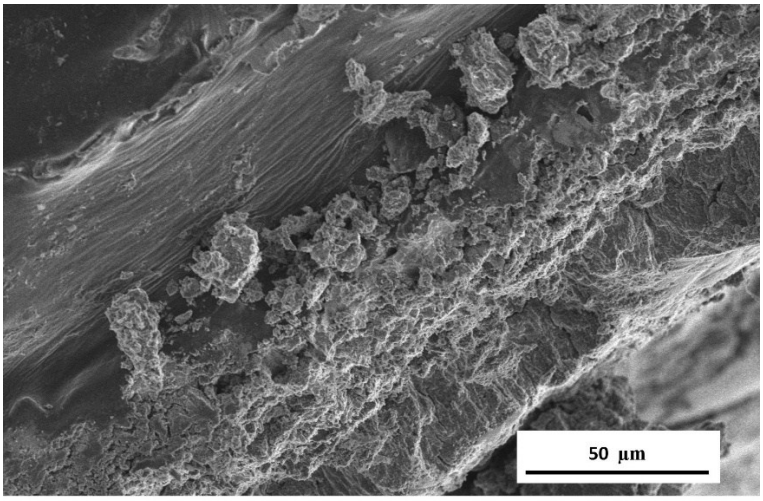
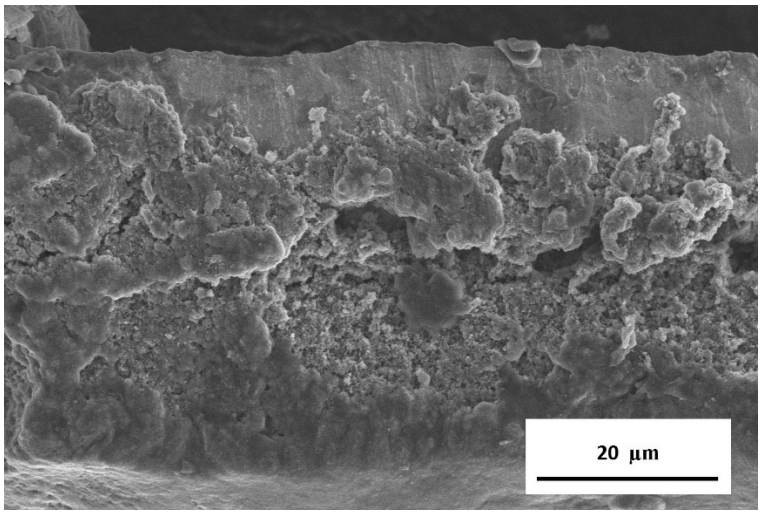


Figure S1 SEM image of Si-pLig w/o PEO.



a



b

Figure S2 a) cross-section SEM image of Si-pLig w PEO after 120 cycles. b) Cross-section SEM image of Si-PVDF after 100 cycles.

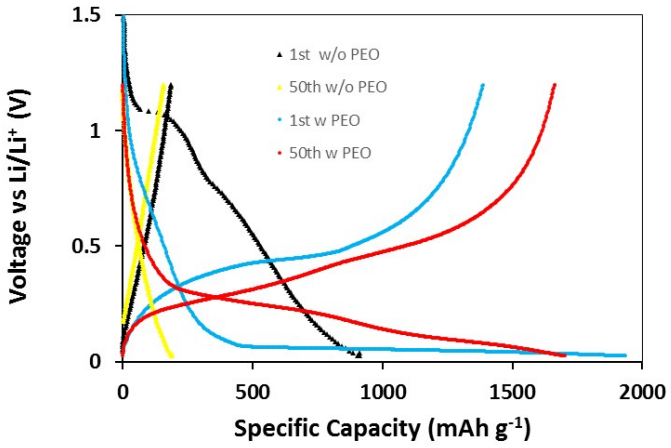


Figure S3 Voltage profile for Si-pLig w/o 0.5% PEO.

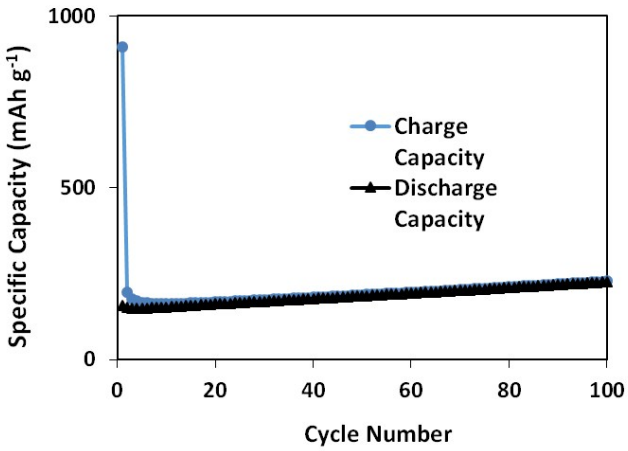


Figure S*4* Charge and discharge capacity of Si-pLig w/o 0.5% PEO. Cycling rate was 0.54 A g^{-1}