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Development of Novel Electrolyte for Silicon based High energy density Li-ion Batteries

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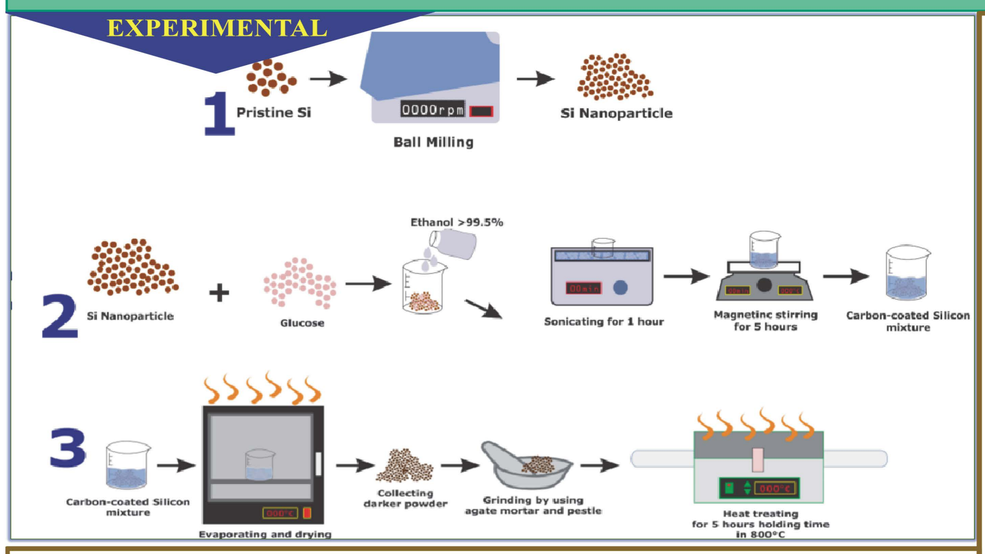
Abstract : Si is one of the promising anodes for LIBs because it has a ten-time higher theoretical capacity than graphite does. However, Si struggles with large volumetric change during lithiation/delithiation, which results in repeated breakdown-regeneration of solid-electrolyte interphase (SEI). The thickening SEI not hinders the Li^+ transport but also decreases the charge-discharge performance upon cycling. These detrimental effects are overcome in this study by modulating the lithium bis(fluorosulfonyl) imide (LiFSI) concentration in a carbonate-based (Ethylene carbonate (EC)/Diethyl carbonate (DEC)) solvent in a silicon@carbon composite anode. The effects of salt concentration on the specific capacity, rate capability, and cycling stability of Si anodes are systematically investigated. The improved properties found for the high-concentration electrolyte can be attributed to the highly conductive and robust SEI formed on the electrode surface.

Research focus



Results and discussion

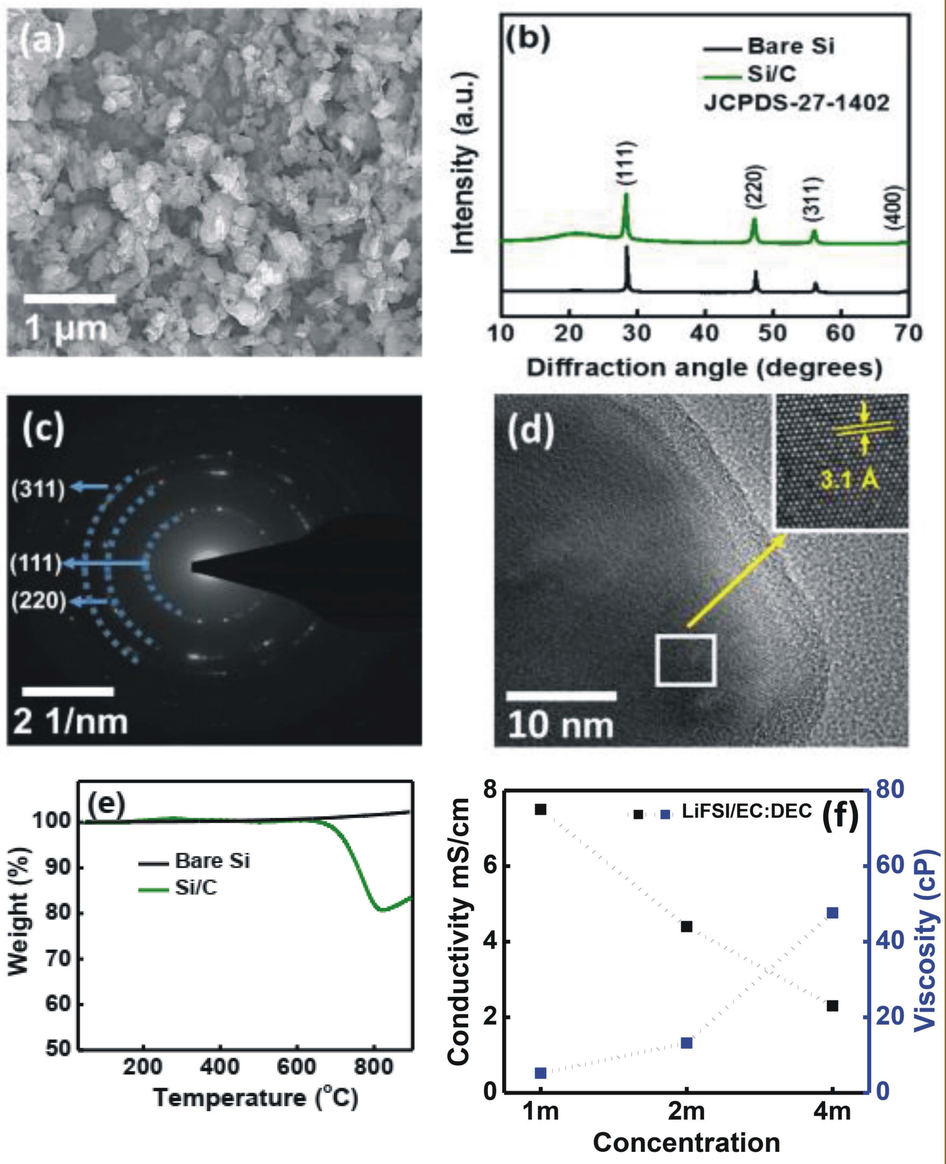


Figure 1: (a) SEM image, (b) XRD pattern, (c) electron diffraction pattern, (d) high-resolution TEM image of synthesized Si/C sample. (e) TGA data of bare Si and Si/C samples, and (f) conductivity and viscosity measurements of various electrolytes with different LiFSI concentrations.

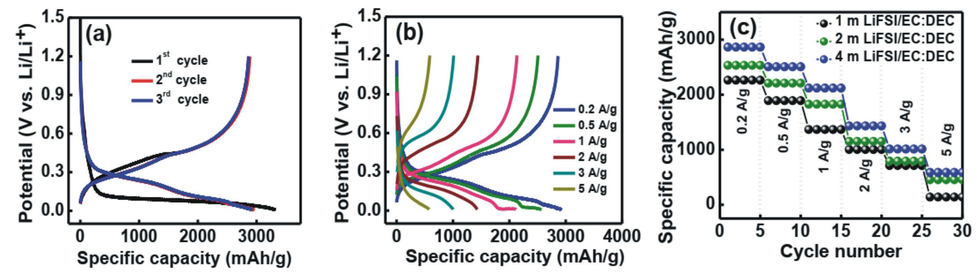


Figure 2. (a) Initial charge-discharge curves and (b) charge-discharge curves at various rates of Si/C electrode recorded in 4 m LiFSI/EC:DEC electrolyte. (c) Comparative rate performance for various electrolytes.

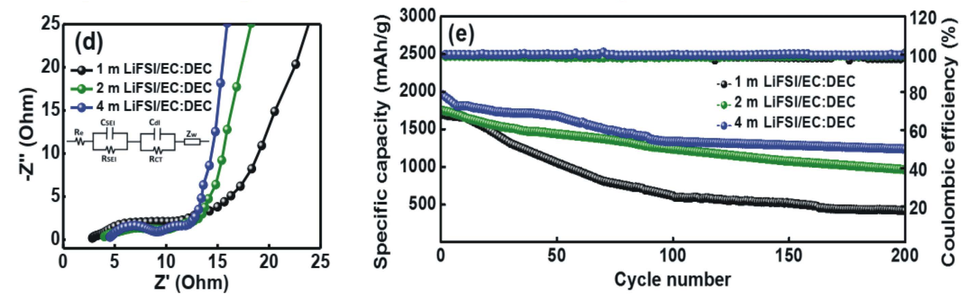


Figure 3: (d) EIS spectra, and (e) cycling stability of Si/C electrodes measured in various EC:DEC-based electrolytes.

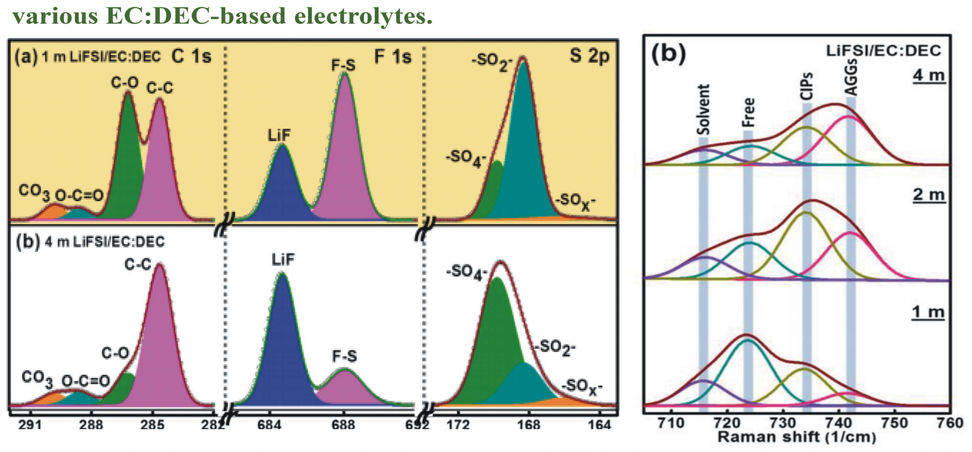


Figure 4: XPS C 1s, F 1s, and S 2p spectra of Si/C electrodes after being cycled 20 times in (a) 1 m LiFSI/EC:DEC and 4 m LiFSI/EC:DEC electrolytes, (b) Raman spectra of EC:DEC-based electrolytes with various concentrations of LiFSI.

Conclusion

In summary, we demonstrate a carbonate-based concentrated electrolyte. the superior performance of Silicon/carbon composite as anode for LIBs, 4 m LiFSI-EC:DEC can be used to efficiently improve the cycling stability of Si/C electrodes as a potential lithium-ion battery electrolyte. A high reversible capacity and only 30% capacity fading rate after cycle are delivered in HCE. From the XPS results reveled that SEI formed on Si/C anode is rich In LiF that can effectively inhibit the continuous electrolyte decomposition. The achieved results demonstrate that increasing the electrolyte concentration consequences in an improved cell performance with higher capacity, especially for the first stabilization cycles. The notable electrochemical performances of Si@C anode validate that the optimized 4 m LiFSI in EC: DEC can be a promising alternative for potential LIBs electrolyte.

Publications

1. **Bharath Umesh**, Purna Chandra Rath, Rahmandhika Firdauzha Hary Hernandha, Jeng-Yu Lin, S. B. Majumder, Quan-Feng Dong, Jeng-Kuei Chang. Moderate-Concentration Fluorinated Electrolyte for High-Energy-Density Si//LiNi $_{0.8}$Co$_{0.1}$ Mn $_{0.1}$O$_2$ Batteries. ACS Sustainable Chemistry and Engineering.

2. Anif Jamaluddin, **Bharath Umesh**, Fuming Chen, Jeng-Kuei Chang and Ching-Yuan Su. Facile synthesis of core–shell structured Si@graphene balls as a high-performance anode for lithium-ion batteries. Nanoscale.

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