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Silicon anodes for lithium ion batteries

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INTRODUCTION & AIM

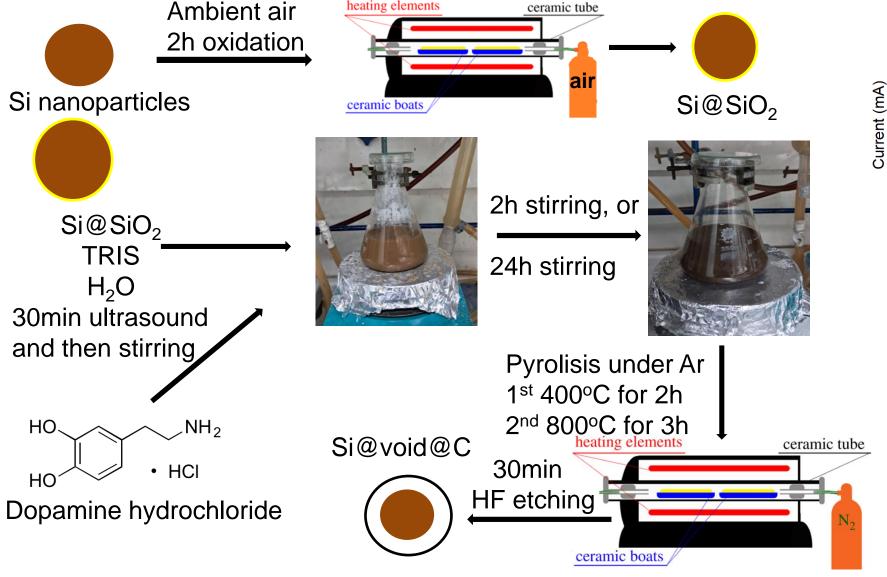
Lithium-ion batteries are among the most promising energy storage technologies. Graphite is the most commonly used anode material, with a theoretical capacity of 372 mAh/g, ongoing research is exploring alternative materials to enhance performance. One of the leading candidates is silicon, which offers an exceptionally high theoretical capacity of approximately 4200 mAh/g.^[1]

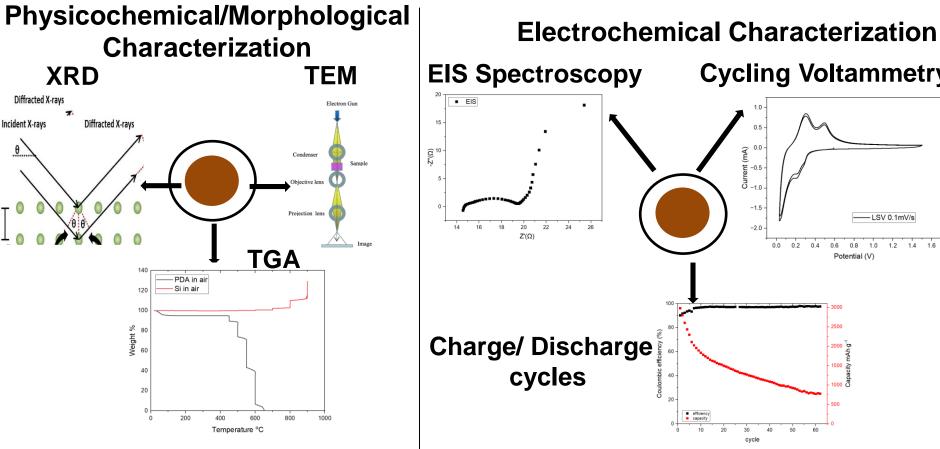
However, silicon anodes face significant challenges, particularly due to severe volume expansion (up to 300%) during lithiation. This expansion leads to mechanical stress, resulting in cracking, pulverization, and eventual degradation of the electrode structure.[2] To address these issues, current research focuses on the development of silicon-carbon yolk-shell anodes. These structures are engineered to accommodate volume changes and improve cycling stability. In yolk-shell architectures, a carbon shell encapsulates the silicon core, providing mechanical support and maintaining electrical conductivity throughout the charge-discharge cycles.

METHODS

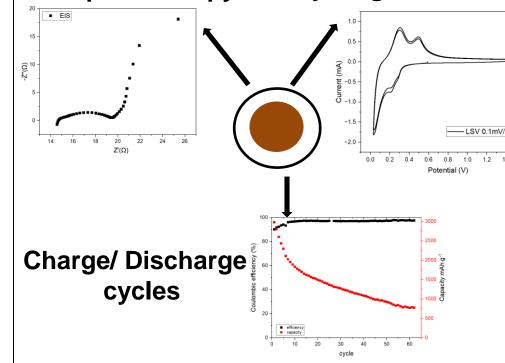
Material Synthesis

Yolk-shell structures are synthesized via dopamine polymerization conducted over varying durations (e.g., 2 hours and 24 hours). To create the void space between the carbon shell and the silicon core, bare silicon particles are first oxidized under ambient air conditions. Following polymer carbonization, the oxidized silicon layer is selectively etched, resulting in a hollow cavity that accommodates silicon expansion during lithiation.

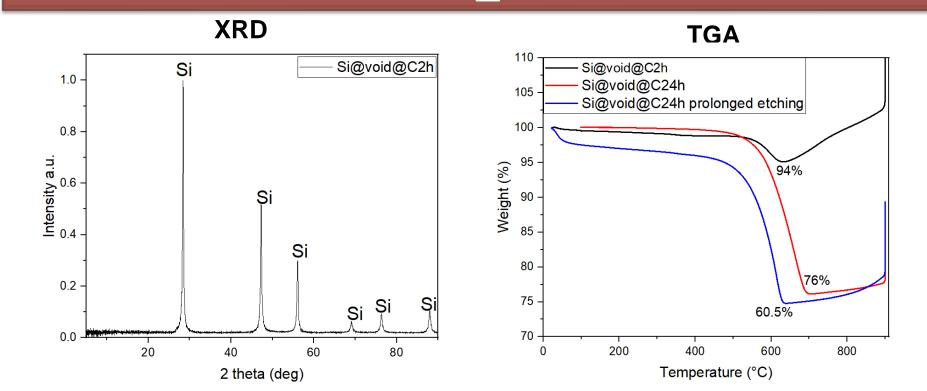




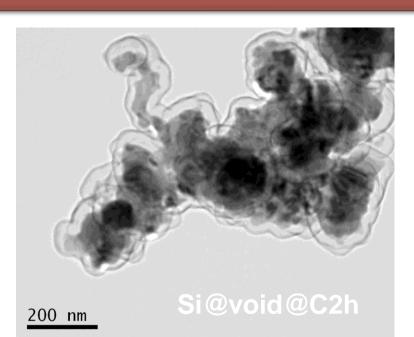
Cycling Voltammetry



RESULTS & DISCUSSION 1 Physicochemical characterization



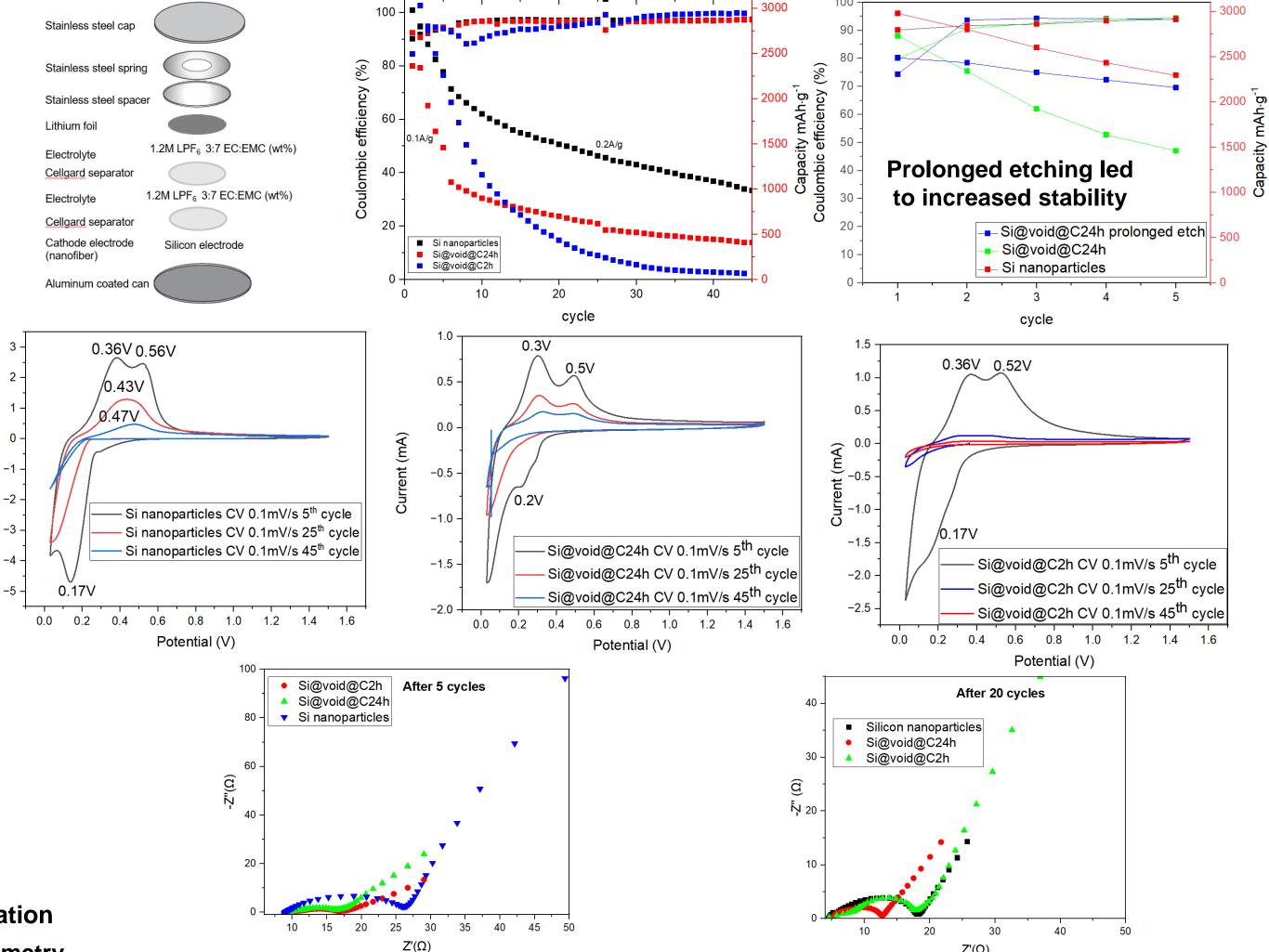
RESULTS & DISCUSSION 2 Morphological characterization







RESULTS & DISCUSSION 3 Electrochemical characterization



CONCLUSIONS

- > In this study, silicon—carbon yolk-shell materials were synthesized for application in lithium-ion batteries. TEM characterization confirmed the successful formation of the yolk-shell structure. The carbon shell thickness was found to be greater for the 24-hour dopamine polymerization (Si@void@C24h) compared to the 2-hour polymerization (Si@void@C2h). This observation is supported by thermogravimetric analysis (TGA), which showed a carbon content of 24% for Si@void@C24h and 6% for Si@void@C2h. XRD spectra revealed distinct peaks corresponding to crystalline silicon.
- > The coulombic efficiency increased during the first few cycles for all samples. This improvement is attributed to the formation and stabilization of the solid electrolyte interphase (SEI) layer.
- > Cyclic voltammetry (CV) analysis showed a cathodic peak at ~0.2 V, corresponding to lithium alloying with silicon, and anodic peaks between 0.3-0.5 V, associated with dealloying during delithiation. Among the samples, Si@void@C exhibited the best electrochemical reversibility and cycling stability.
- > EIS revealed lower resistance values for both Si@void@C materials compared to uncoated silicon, confirming that the carbon shell enhances electronic conductivity while mitigating resistive SEI and charge-transfer barriers.
- > A gradual decrease in peak current from the 5th to the 45th cycle was observed, likely due to silicon pulverization, loss of active material, and structural degradation. This decline in electrochemical activity is further supported by the increase in impedance over cycling.
- > Electrochemical testing showed that bare silicon nanoparticles exhibited higher initial capacity than both yolkshell electrodes. This may be attributed to the relatively small void space between the silicon core and carbon shell, as observed in TEM images, which limits the accommodation of volume expansion during lithiation. Consequently, silicon pulverization leads to particle detachment, carbon shell fracture, and capacity fading.
- > This study led to the prolonged HF etching of Si@void@C24h. Initial results show promising stability.

References

[1] Y. Ma, P. Guo, M. Liu, P. Cheng, T. Zhang, J. Liu, D. Liu, D. He, Journal of Alloys and Compounds, 905 (2022) 164189. [2] F. Zhang, Z. Jia, C. Wang, A. Feng, K. Wang, T. Hou, J.Liu, Y. Zhang, G. Wu, Energy 195 (2020) 117047.