Scaling Up Silicon-Dominant Anodes: Rheological Control and Bio-Based Binders for Pilot-Scale Battery Electrode Manufacturing

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The **GIGAGREEN project** targets the transition of novel lithium-ion cell manufacturing technologies from laboratory-scale validation (**TRL 3-4**) to scalable pilot-line manufacturing (**TRL 5-6**).

- *Target Formulations*: Scaling sustainable, water-processable anode slurries incorporating **10 wt.% silicon**.
- *Target Formulations*: Scaling sustainable, water-processable cathode slurries incorporating **LNMO**.
- *Process Transferability*: Scaling aqueous slurry formulations from laboratory scale to continuous pilot-line processing.
- *Manufacturing Reliability*: Overcoming slurry processing bottlenecks to achieve defect-free, uniform coatings at commercial mass loadings.
- *Design-to-Manufacture (DtM)*: Aligning sustainable material choices with high-throughput, energy-efficient Gigafactory requirements.

Going beyond the Gigagreen project…

Silicon-Dominant Anodes Material Limitations & Processing Bottlenecks

Silicon-Dominant Anodes (>50 wt.% Si) surpass standard graphite capacity (372 mAh g^{-1}) to achieve stable electrode capacities (~ 1000 mAh g^{-1}) but face major structural and processing bottlenecks.

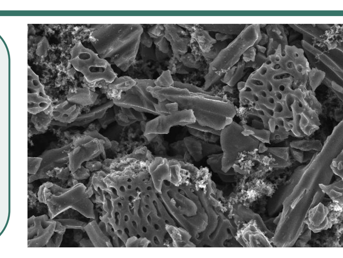
Mechanical & SEI Failure
Extreme volumetric changes (~300%) lead to particle pulverization, delamination, and continuous SEI degradation.

Particle Agglomeration
High silicon loading drastically alters slurry surface dynamics, causing severe particle clustering compared to graphite.

Slurry Instability
High tendency toward phase separation, viscosity decay, and rapid sedimentation in pilot-scale mixing vessels.

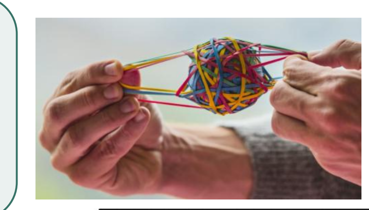
How to overcome silicon issues

Material Level
Synthesis of material with specified shapes and dimensions



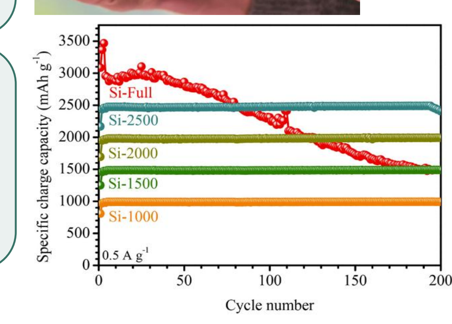
E-MAGY on with silicon

Electrode Level
Use of linear, branched, and crosslinked binders



Mechanical Properties
Robustness vs. Elasticity

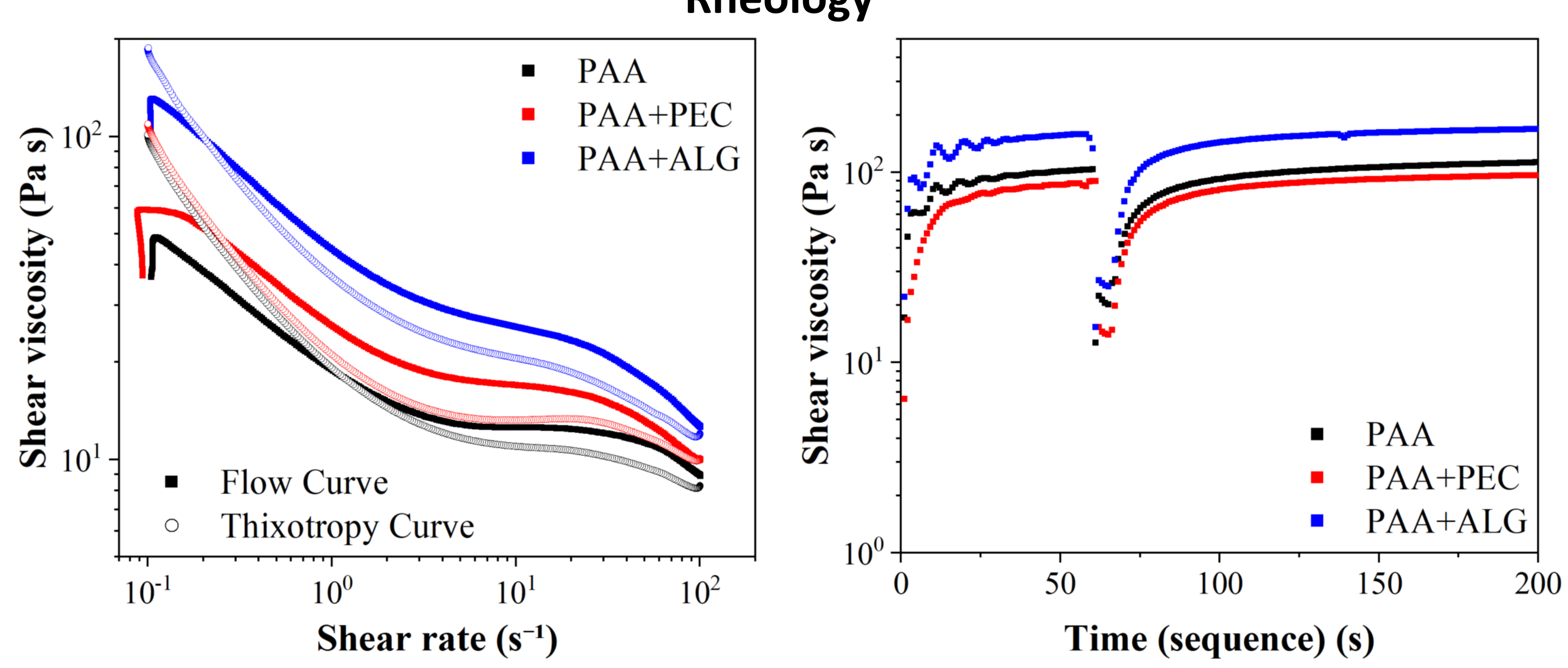
Electrochemical Application
Limiting the capacity to preserve the material structure



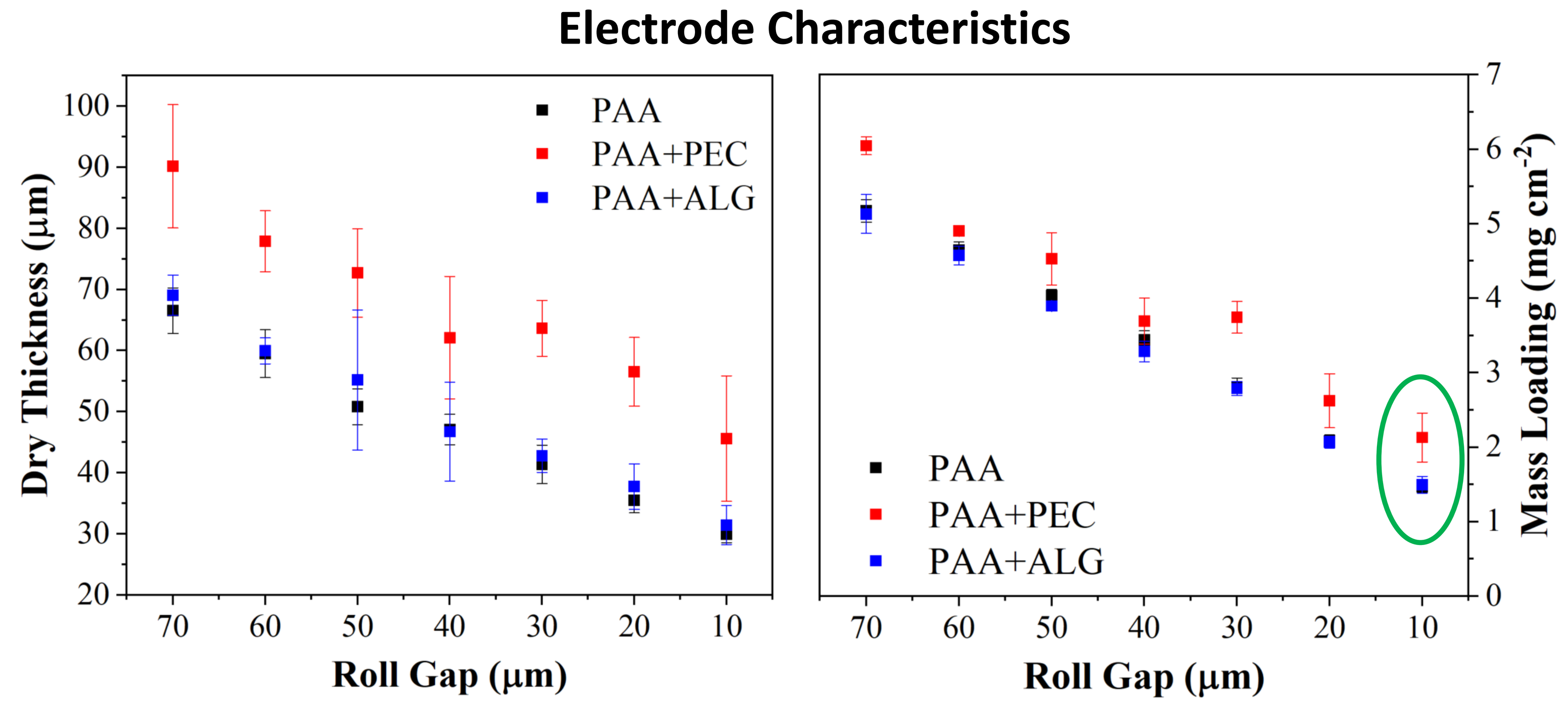
1000 mAh g^{-1} is enough to balance the cathodes [1]

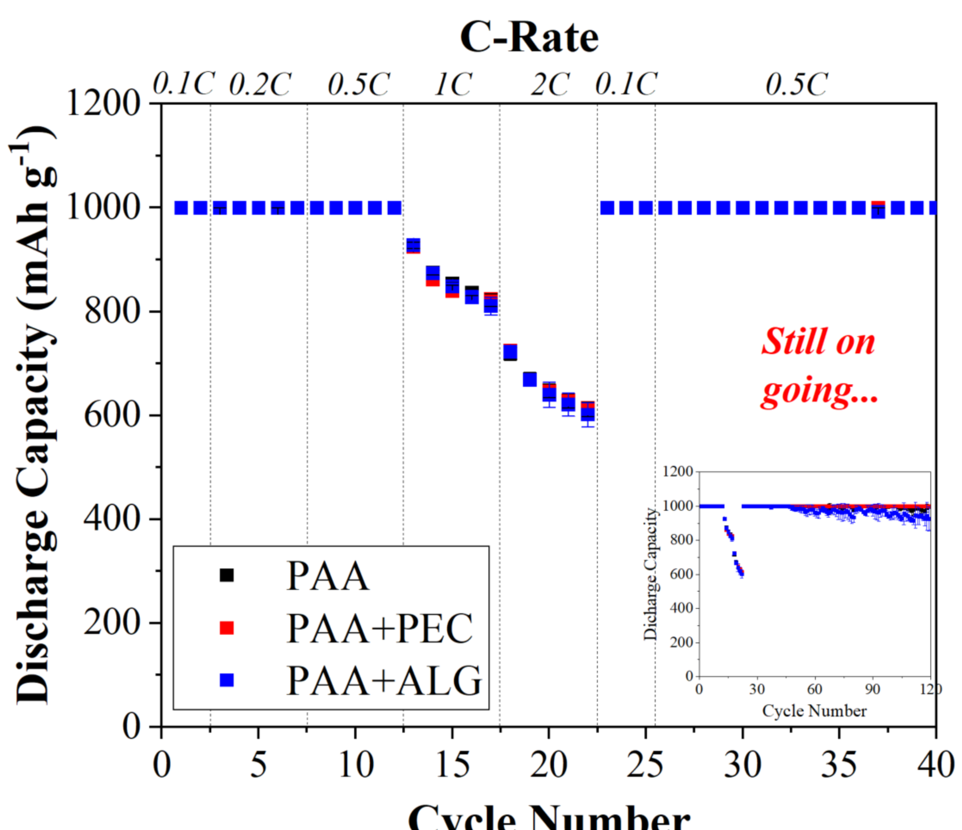
Slurries and Electrode properties

Rheology



Electrode Characteristics



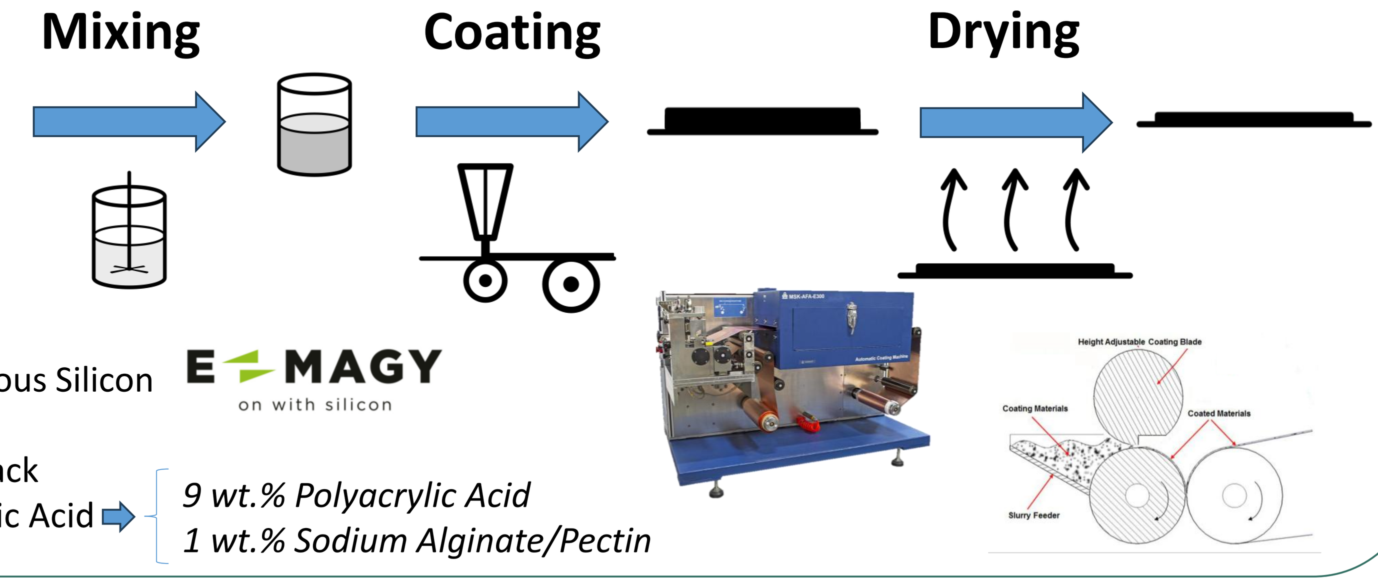


Electrochemical Characterization

- **High-Rate Capability**: All binder formulations show full capacity recovery (~1000 mAh g^{-1}) after high C-rates.
- **Comparable Long-Term Stability**: Bio-based blends perform comparably to pure PAA over 120+ cycles.

Half Cell: Silicon Anode vs. Lithium
Anode Mass Loading: 1-2 mg cm^{-2}
Electrolyte: 150 μL of 1 M $LiPF_6$ in EC:DEC 1:1 + 10% FEC + 2% VC

Anode Production Process

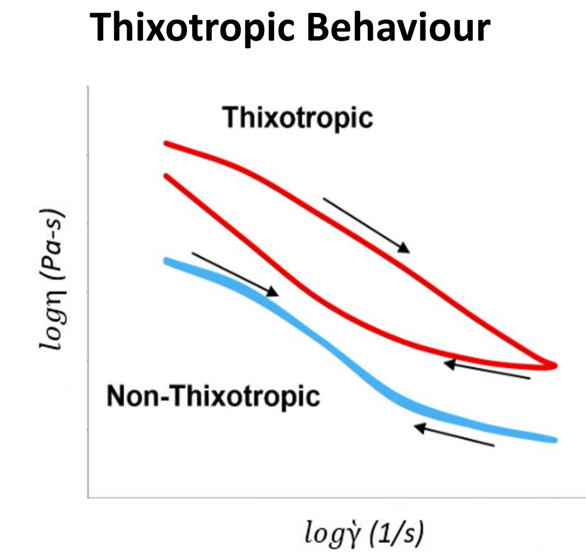


Composition
80 wt.% Nanoporous Silicon
5 wt.% Graphite
5 wt.% Carbon Black
10 wt.% Polyacrylic Acid ⇒ *9 wt.% Polyacrylic Acid*, *1 wt.% Sodium Alginate/Pectin*

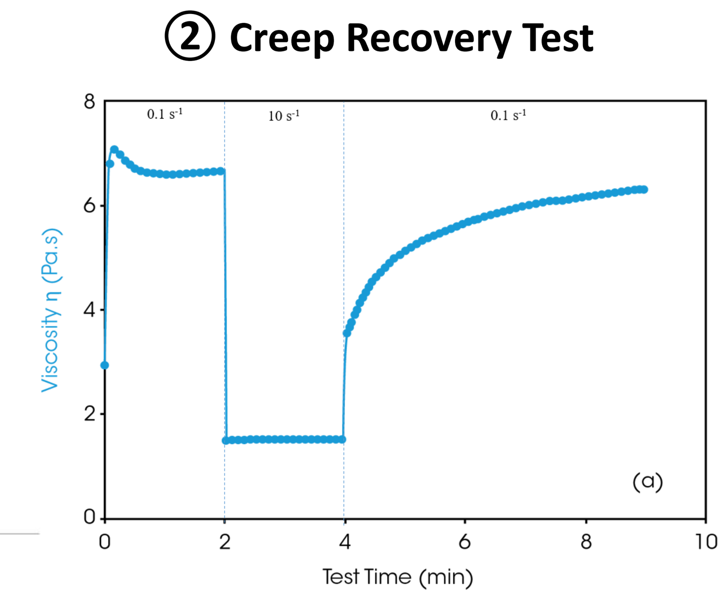
E-MAGY on with silicon

The importance of rheology

① Flow Curve & Thixotropic Behaviour



② Creep Recovery Test



Conclusions

- ✓ **Rheology as a Predictive Tool:** Fundamental for tuning slurry behavior, ensuring processability, and preventing defects in continuous coating lines.
- ✓ **Pilot-Scale Process Control:** Strong parameter-property relationships allow precise tuning of mass loading and coating thickness for uniform electrodes.
- ✓ **Ongoing Challenges for Si-Dominant Anodes:** While controlled capacity (~1000 mAh g^{-1}) ensures stability, further research is needed to address volumetric expansion, long-term degradation, and full-cell scalability.

[1] Xuming Yang, Shengrui Tang, Liewu Li, Yaqi He, Menghao Li, Xianbin Wei, Jiangtao Hu, Jionghui Wang, Xiaoping Ouyang, M. Danny Gu, Qianlin Zhang, Jianhong Liu; Lithiation Depth Regulation of Silicon Anodes toward Excellent Stability. *J. Phys. Chem. Lett.* 18 July 2024; 15 (28): 7320–7326. https://doi.org/10.1021/acs.jpclett.4c01599

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