Optimization of Waterborne Cathode Production for Lithium-Ion Batteries: Investigating Binders, Current Collectors, and Calendering Effects

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Introduction

Lithium-ion batteries (LIBs) are essential for sustainable energy, powering electric vehicles (EVs) and broader energy storage systems. To meet increasing demand, this research explores **waterborne cathode production** as a scalable and eco-friendly alternative to traditional solvent-based methods.

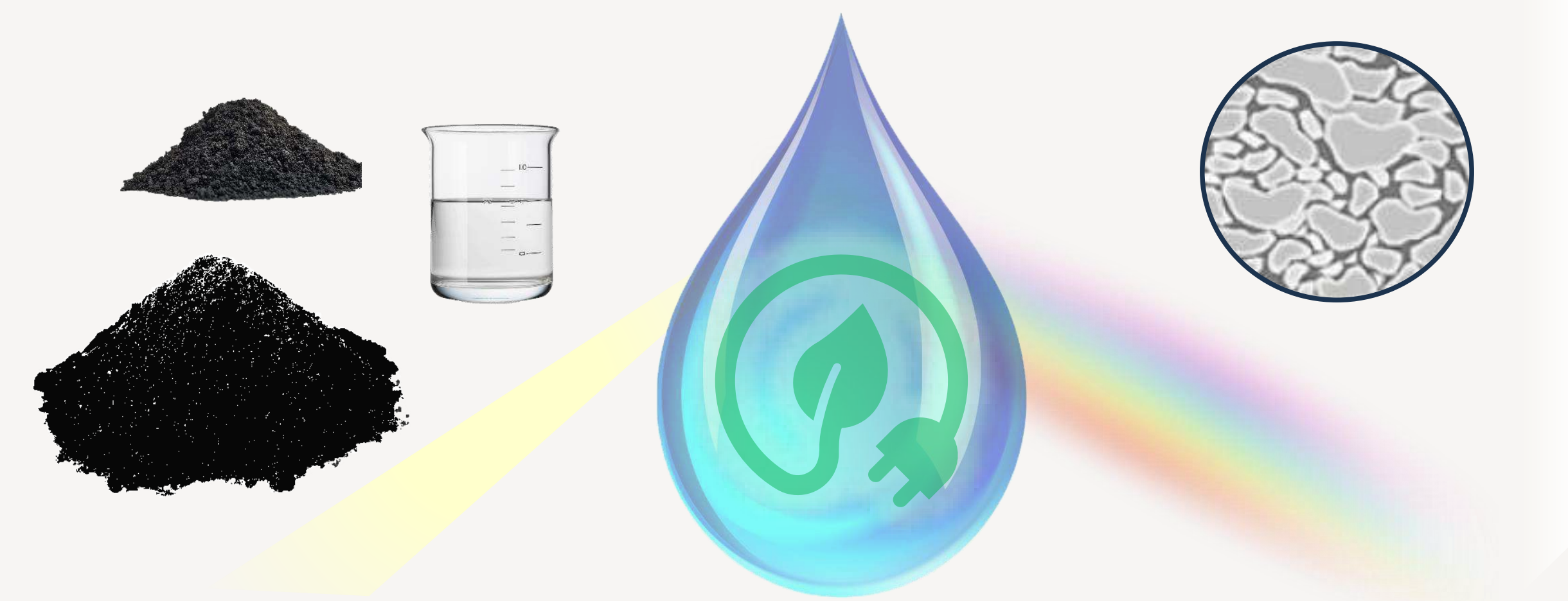
This study aims to help the transition and scale up of **LNMO-based waterborne cathodes** from lab-scale to a pilot line, investigating:
1. Binder Selection: Testing 2 different kinds of water-based emulsions (SX8684(A)-64 and TRD202A) and Carboxymethyl Cellulose (CMC) 2-component binder systems;
2. Current Collectors: Comparing pristine vs. carbon-coated aluminum foil;
3. Calendering: Evaluating post-production calendering's impact on electrode performance.
Through techniques such as **galvanostatic cycling**, **ICI** (Intermittent Current Interruption) and **PEIS** (Potentiostatic electrochemical impedance spectroscopy), this work aims to enhance LIB scalability and performance, supporting green technology adoption in EVs and energy storage.

Methods

1

Cathodes preparation

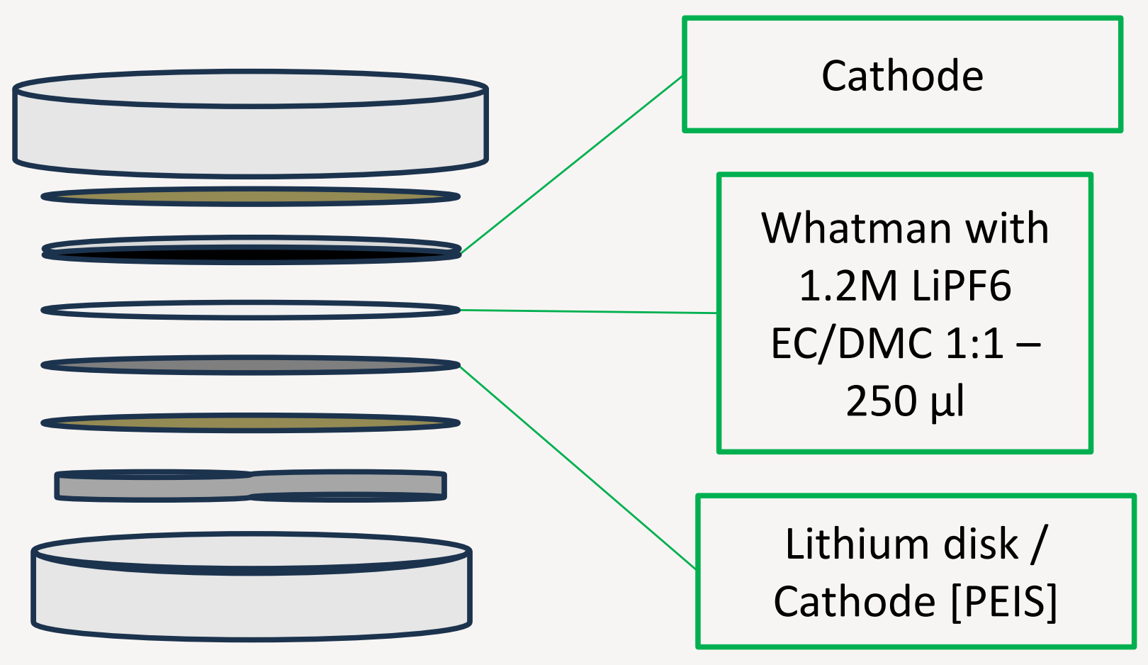
Cathode composition: 92:3:1:4
(LNMO:C45:CMC: [*SX8684(A)-64* or *TRD202A*])
Mixing method: Over-head stirring



2

Coin-cell assembly

Cell architecture: 2032 half-cell
CG/ICI: *WE:* Cathode **-** *CE:* Lithium chip
PEIS: *Symmetrical:* Double cathode
Target ML: 10-12.5 mg/cm^2



Cathode

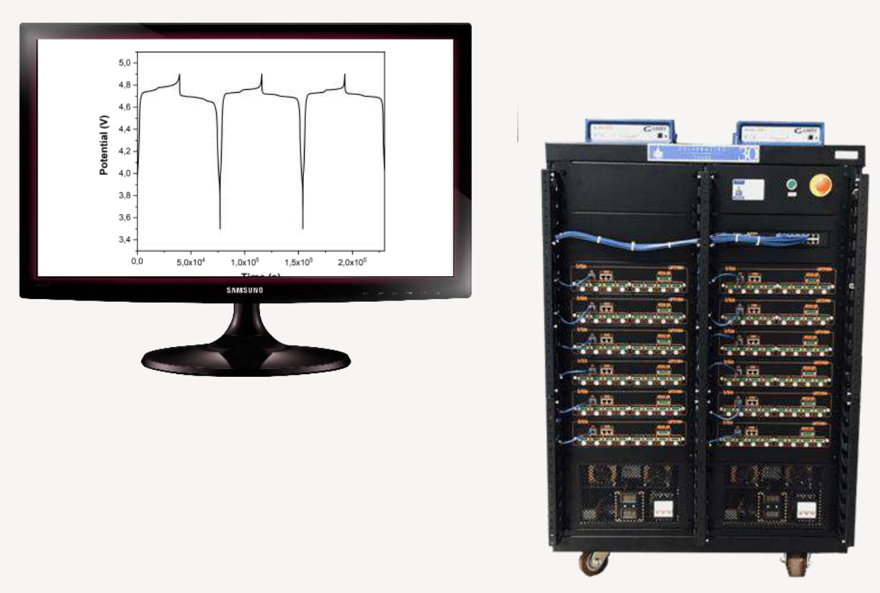
Whatman with 1.2M LiPF6 EC/DMC 1:1 – 250 µl

Lithium disk / Cathode [PEIS]

3

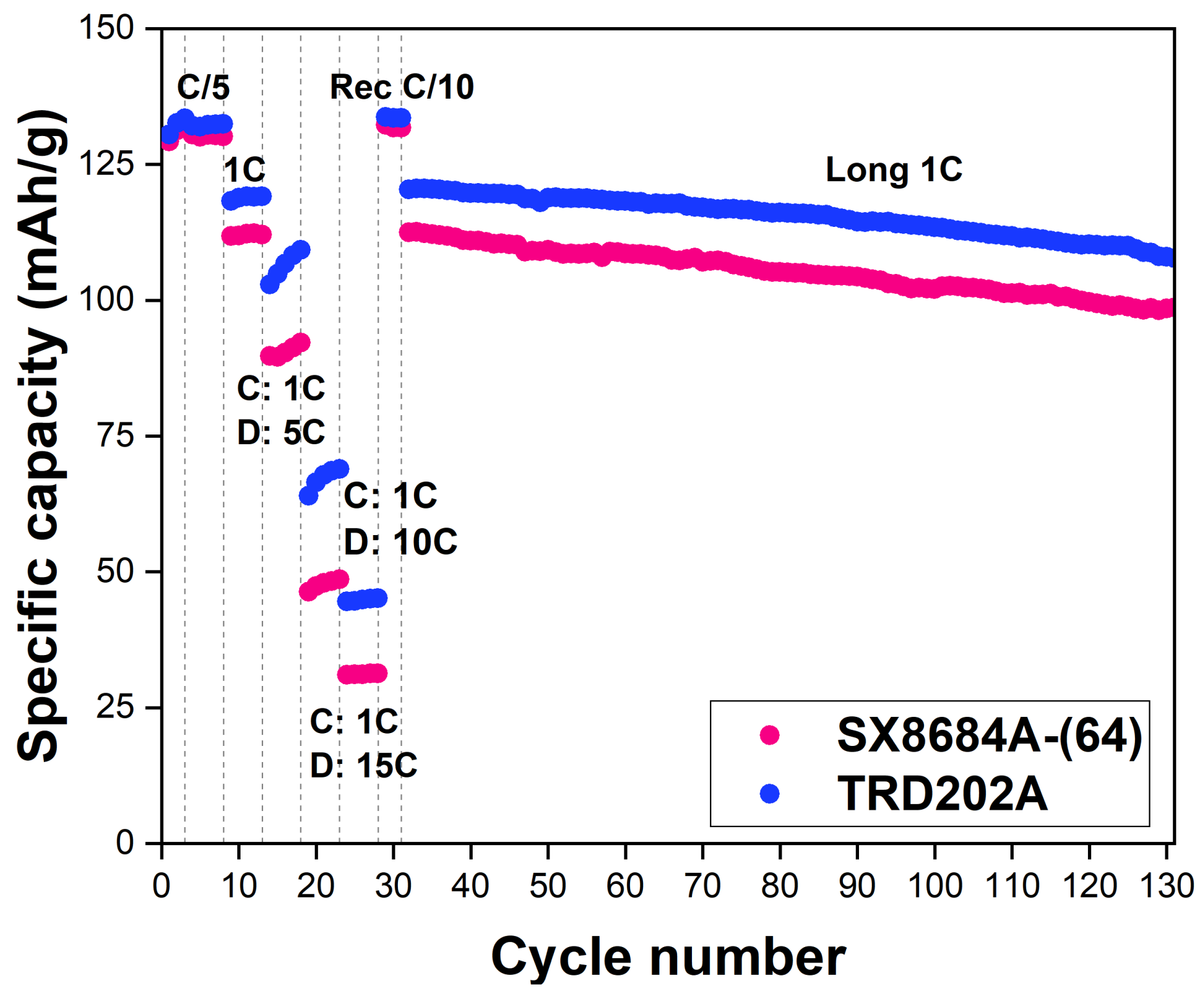
Testing

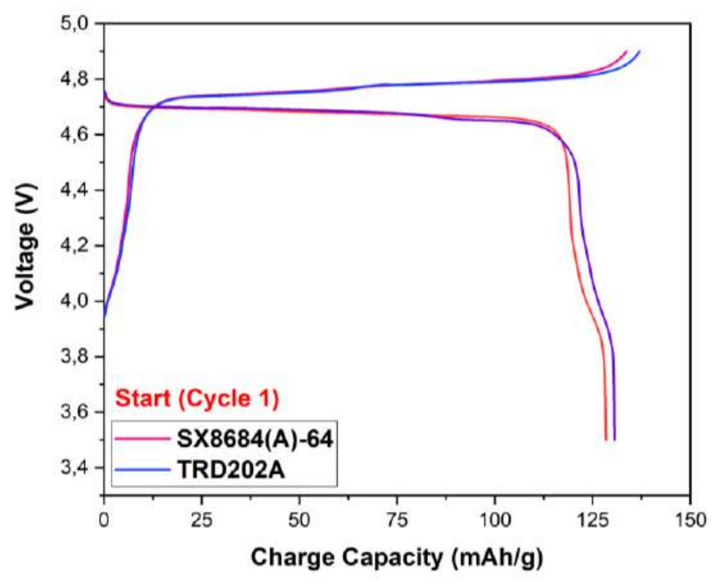
Galvanostatic cycling
ICI (Intermittent Current Interruption)
PEIS (Potentiostatic electrochemical impedance spectroscopy)

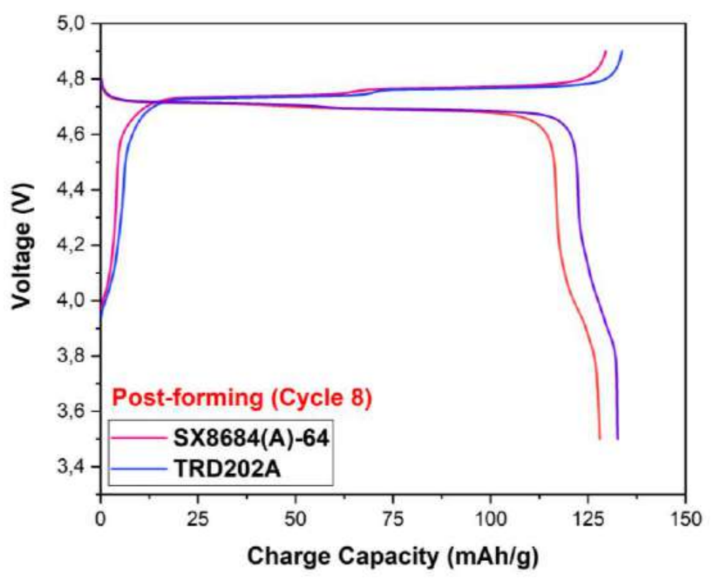


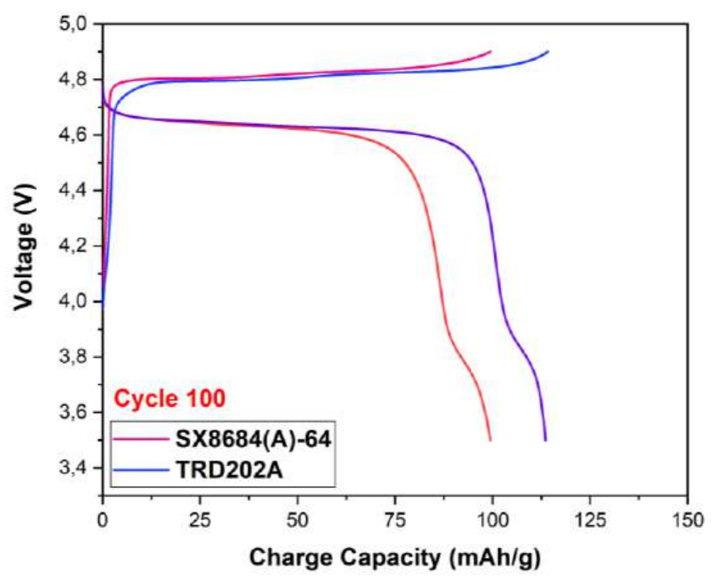
Results

Galvanostatic cycling

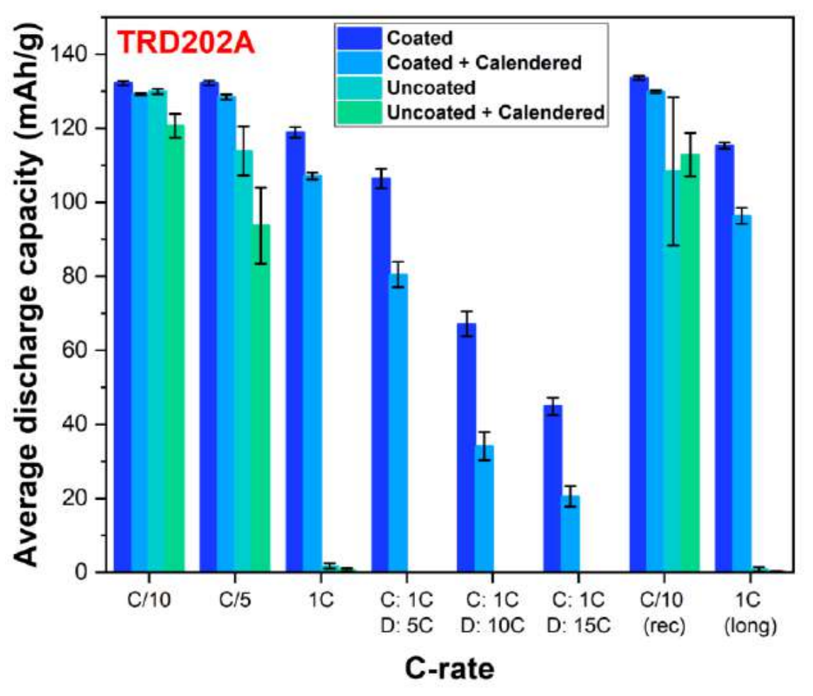


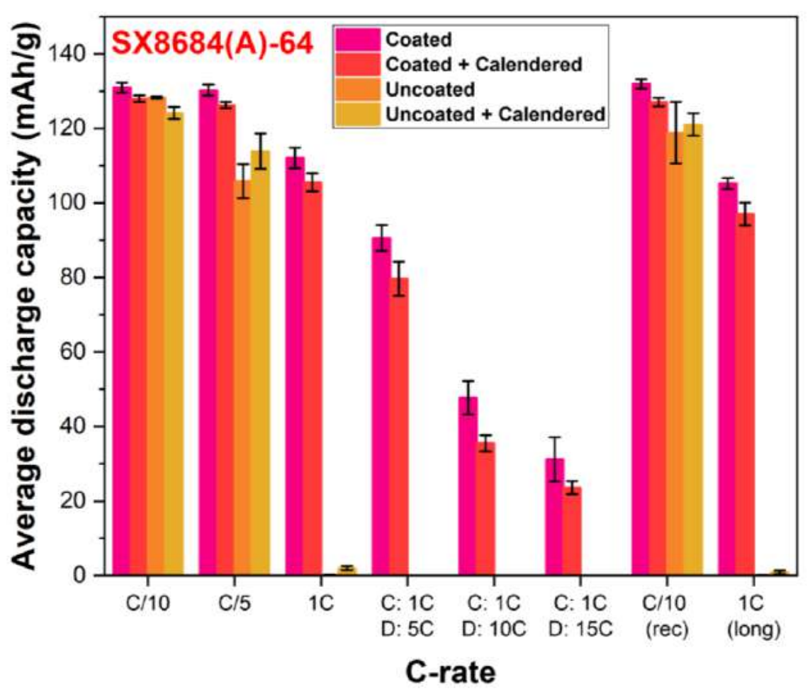






Cycling protocol [3.5-4.9V]:
Rest 12 hrs
3 cycles C/10 (CD)
5 cycles C/5 (CD)
5 cycles 1C (CD)
5 cycles 1C (C) - 5C (D)
5 cycles 1C (C) - 10C (D)
5 cycles 1C (C) - 15C (D)
3 cycles C/10 (CD)
100 cycles 1C (CD)

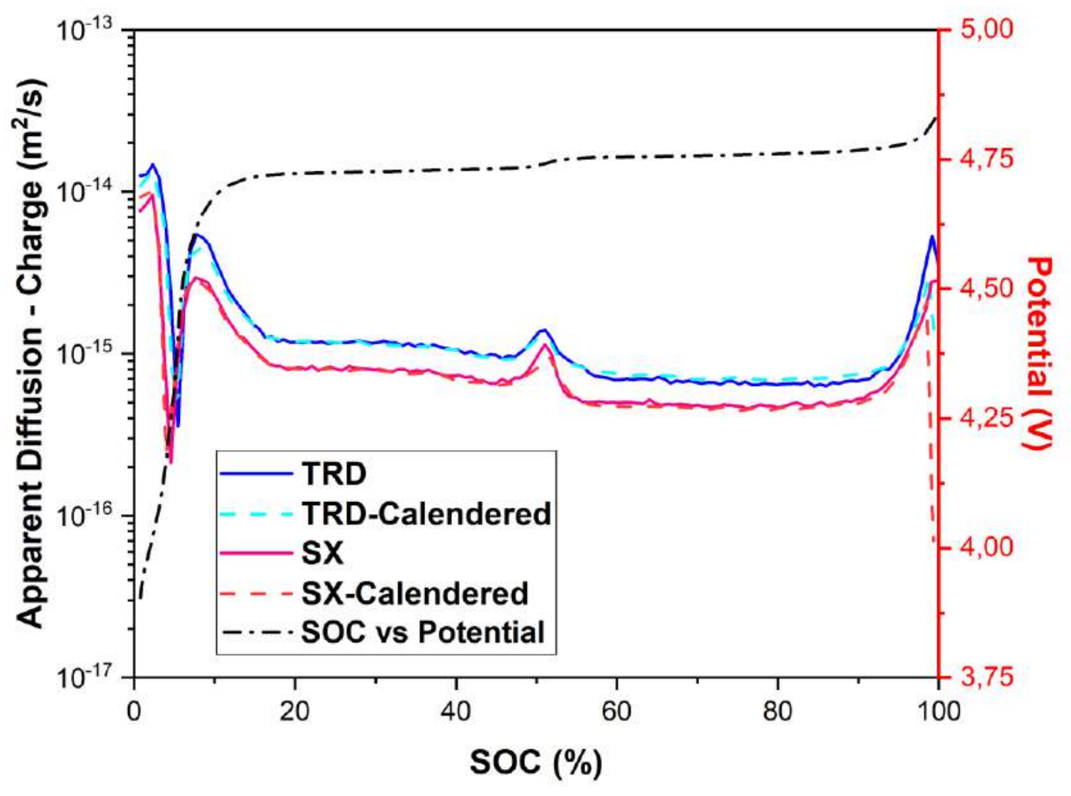


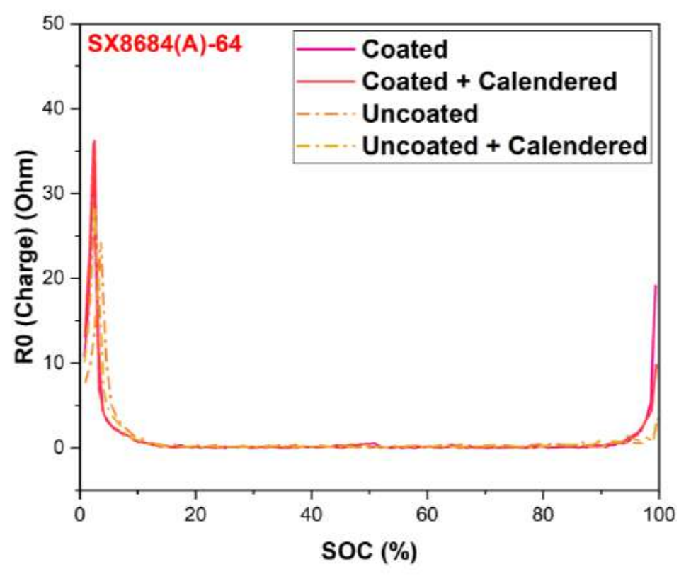


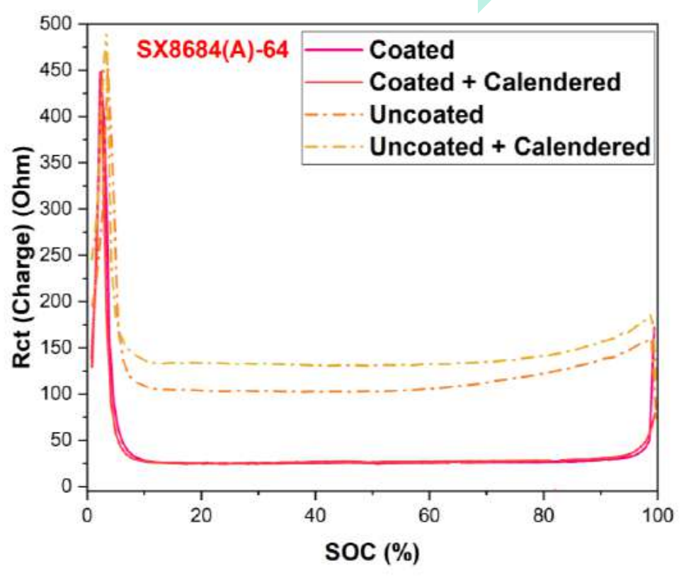
ICI (Intermittent Current Interruption)

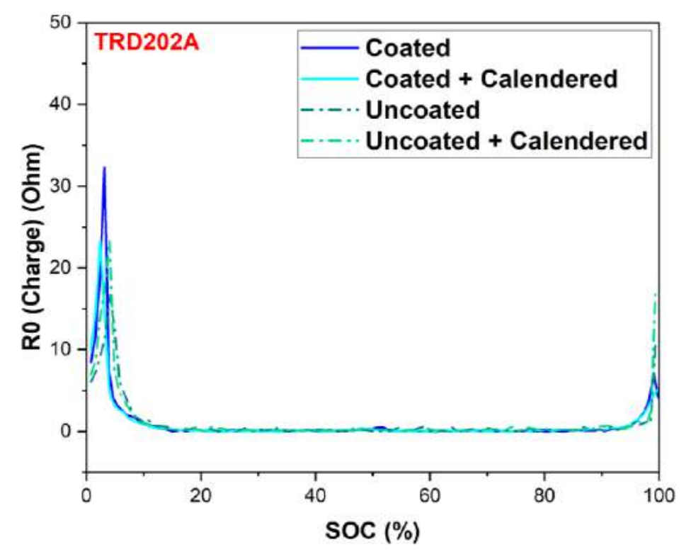
R_0 R_{ct}

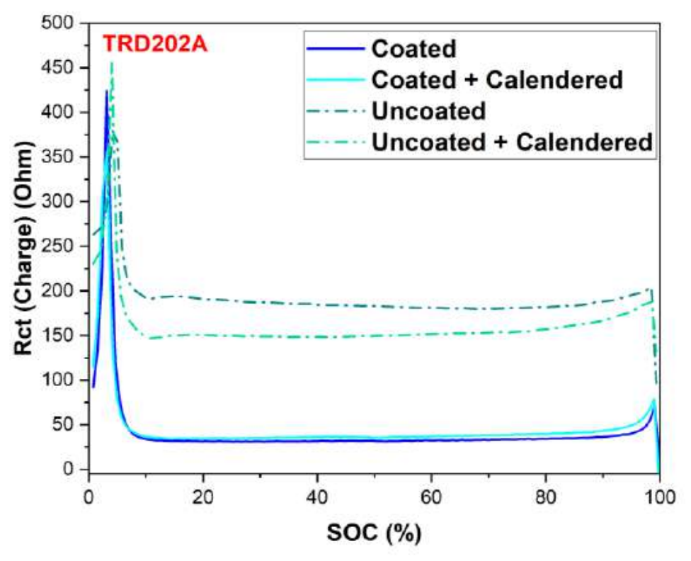
Cycling protocol[1]:
Rest 12 hrs
5 cycles C/10 (CD)
ICI (C/10, 5min pulse, 5s rest, 0.1s sampling time)



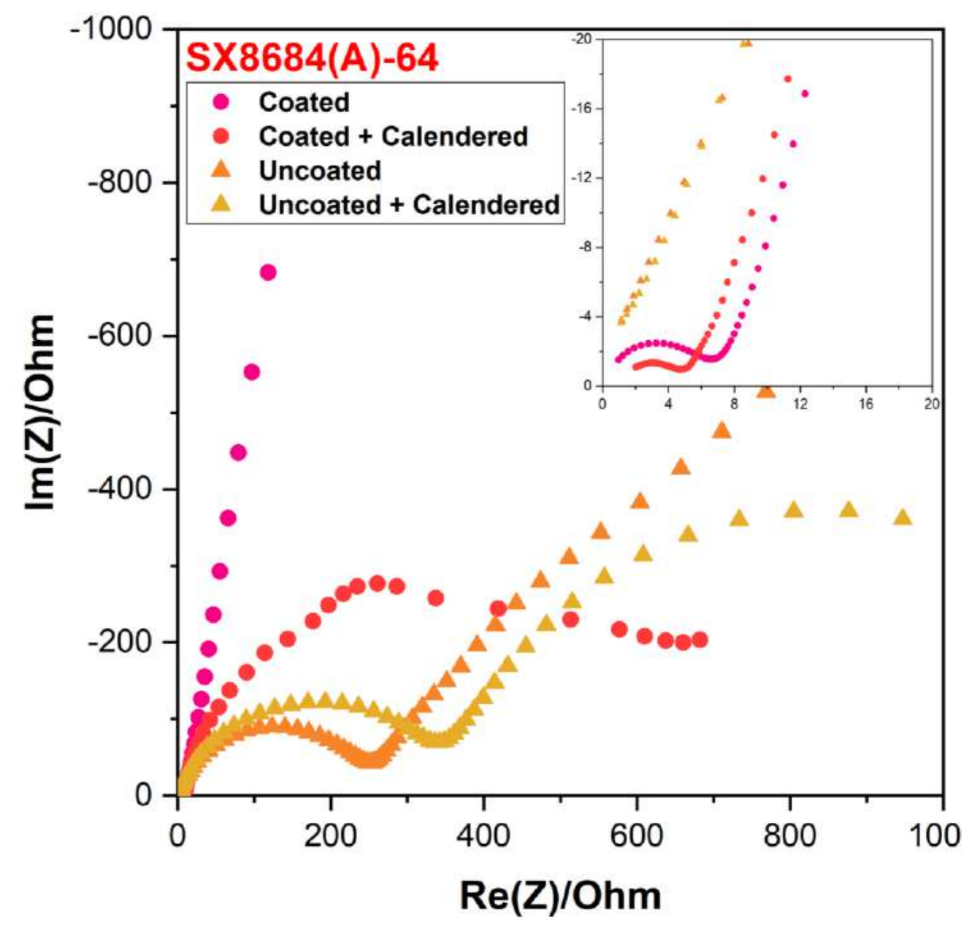


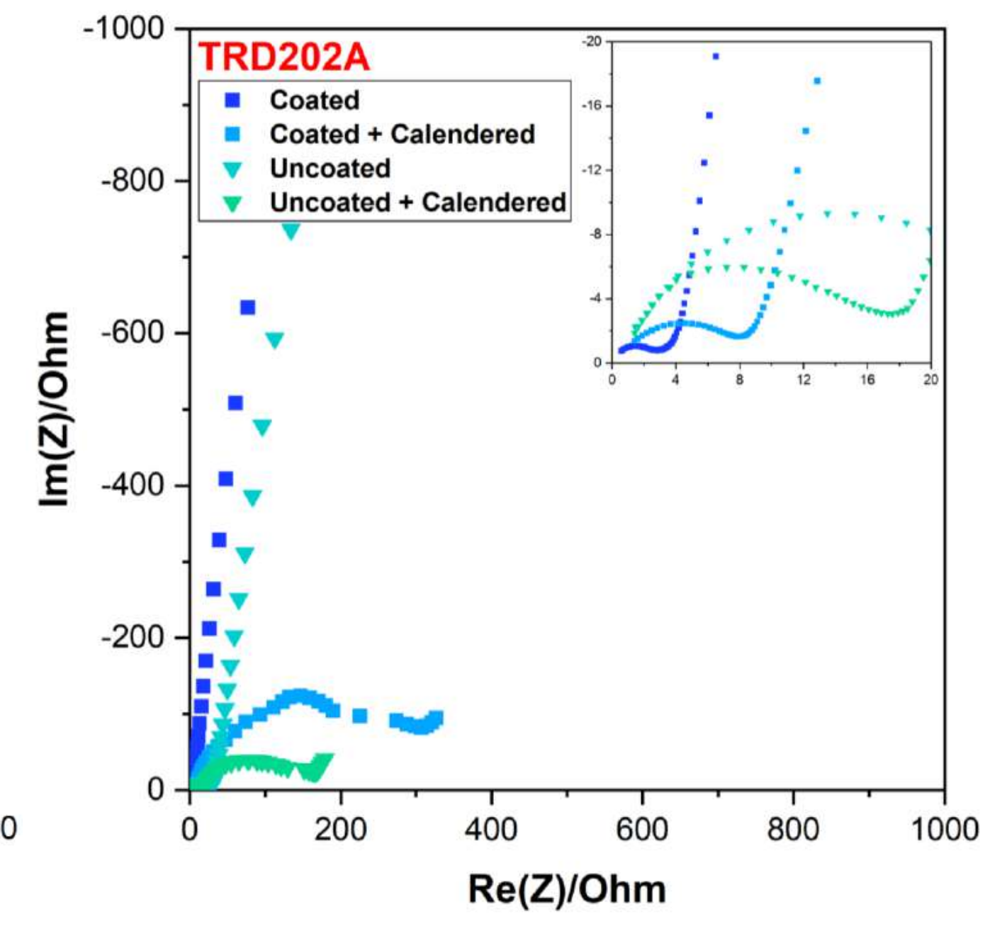






PEIS





R_{ct} estimation (TLM-Q model)[2,3] :
Frequency range: 100 kHz – 20 mHz
ΔV: 15 mV
Temperature: 25°C

SX8684(A)-64	R_{ct} (Ω)
Coated	4.1
Coated + Calendered	3.5
Uncoated	130
Uncoated + Calendered	195

TRD202A	R_{ct} (Ω)
Coated	1.9
Coated + Calendered	4.6
Uncoated	16.2
Uncoated + Calendered	8.9

Discussion

Binder Performance:
The **TRD202A + CMC** binder system outperformed **SX8684(A)-64 + CMC** in all tests. **Galvanostatic cycling** showed TRD202A maintained higher capacities than SX8684(A)-64 across all C-rates, with differences especially pronounced at high rates (5C to 15C). **ICI analysis** also indicated a higher apparent diffusion coefficient for TRD202A (2×10^{-15} m^2/s vs. 9×10^{-16} m^2/s), though both systems showed similar resistance values in ICI and PEIS.

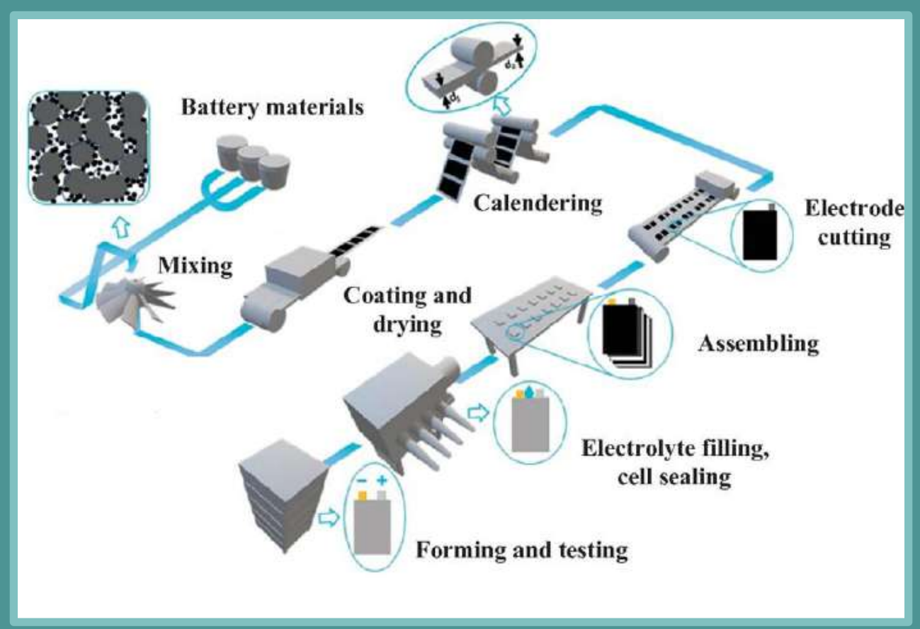
Current Collector Comparison:
Coated current collectors significantly improved performance, with coated CC cathodes retaining around **110 mAh/g** capacity at 1C, compared to near-zero for uncoated ones—valid across both binder systems and increasingly evident at higher rates. ICI showed **R_{ct} values** of 150-200 Ω for uncoated vs. below 40 Ω for coated collectors. PEIS on fresh cells confirmed generally lower R_{ct} for coated systems, especially with SX binder, highlighting the greater impact of current collector choice on performance.

Impact of Calendering:
Contrary to literature, calendered electrodes showed **reduced capacities** and higher R_{ct} values than uncalendered ones, with no significant R_0 differences. These unexpected results, likely due to the unique characteristics of the waterborne slurry (e.g., viscosity or binder distribution), suggest further optimization in slurry preparation is needed to harness the full benefits of calendering.

Conclusions

This study highlights key factors in optimizing waterborne LNMO cathodes for pilot-line scalability. The **TRD202A + CMC** binder system outperformed **SX8684(A)-64 + CMC**, demonstrating the critical role of binder selection, especially at higher C-rates. **Coated current collectors** significantly improved capacity retention and reduced resistance, reinforcing the need for surface treatments in scaled production. Unexpectedly, **calendering** slightly hindered capacity and increased resistance, suggesting adjustments in slurry viscosity and binder ratios may be needed for optimal performance.

These findings offer valuable guidance for refining waterborne cathode production on a pilot-line scale, contributing to efforts toward more scalable and efficient lithium-ion battery manufacturing.



Acknowledgements and References

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[1] Chien, Y.-C. *et al.* (2023) 'Rapid determination of solid-state diffusion coefficients in Li-based batteries via intermittent current interruption method', *Nature Communications*, 14(1). doi:10.1038/s41467-023-37989-6.
[2] Landesfeind, J. *et al.* (2016) 'Tortuosity determination of battery electrodes and separators by impedance spectroscopy', *Journal of The Electrochemical Society*, 163(7). doi:10.1149/2.1141607jes.
[3] Ogihara, N. *et al.* (2012) 'Theoretical and experimental analysis of porous electrodes for lithium-ion batteries by electrochemical impedance spectroscopy using a symmetric cell', *Journal of The Electrochemical Society*, 159(7). doi:10.1149/2.057207jes.

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