Regulated Surface Synergistic Layers of Layered Cathodes Through Low-Temperature Pyrolysis

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Introduction

A promising technique to improve LIBs is to construct a robust surface of carbon on Ni-rich cathodes to eliminate surface degradation and enhance battery performance [1],[2].

- Ni-rich NMC ($LiNi_xMn_yCo_{1-x-y}O_2$ where $x \geq 0.6$) suffers from capacity fading and safety concerns. Low-temperature Pyrolysis can be done with carbon precursors cellulose and urea to synthesize a carbon layer on the surface of Ni-rich NMC cathodes.

Core Hypothesis

A carbon coating layer can transform the surface structure of Ni-rich NMC consisting of carbon coating and reconstruction layer. Its thickness can be fine-tuned by post-annealing protocols, i.e., employing low-temperature carbon precursors. The synergistic layer can not only suppress the surface degradation during cycling but also increase the surface stability against a moisture environment.

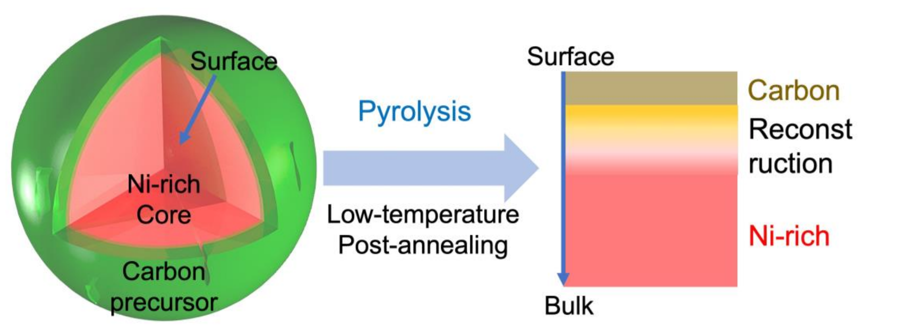
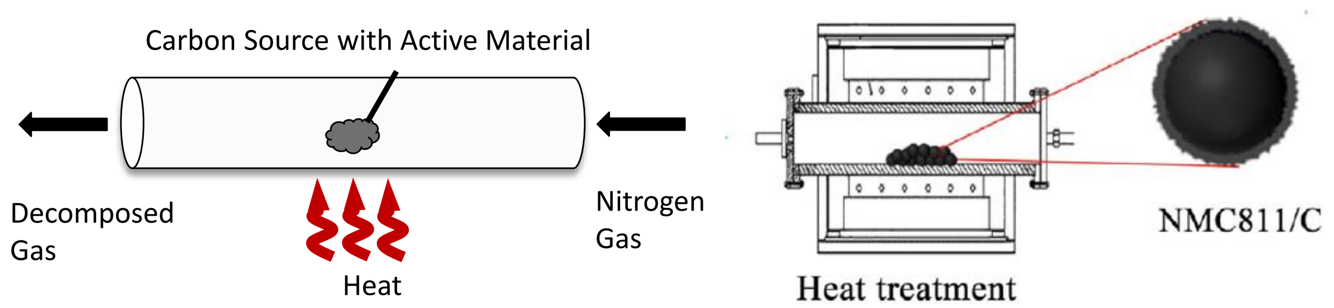


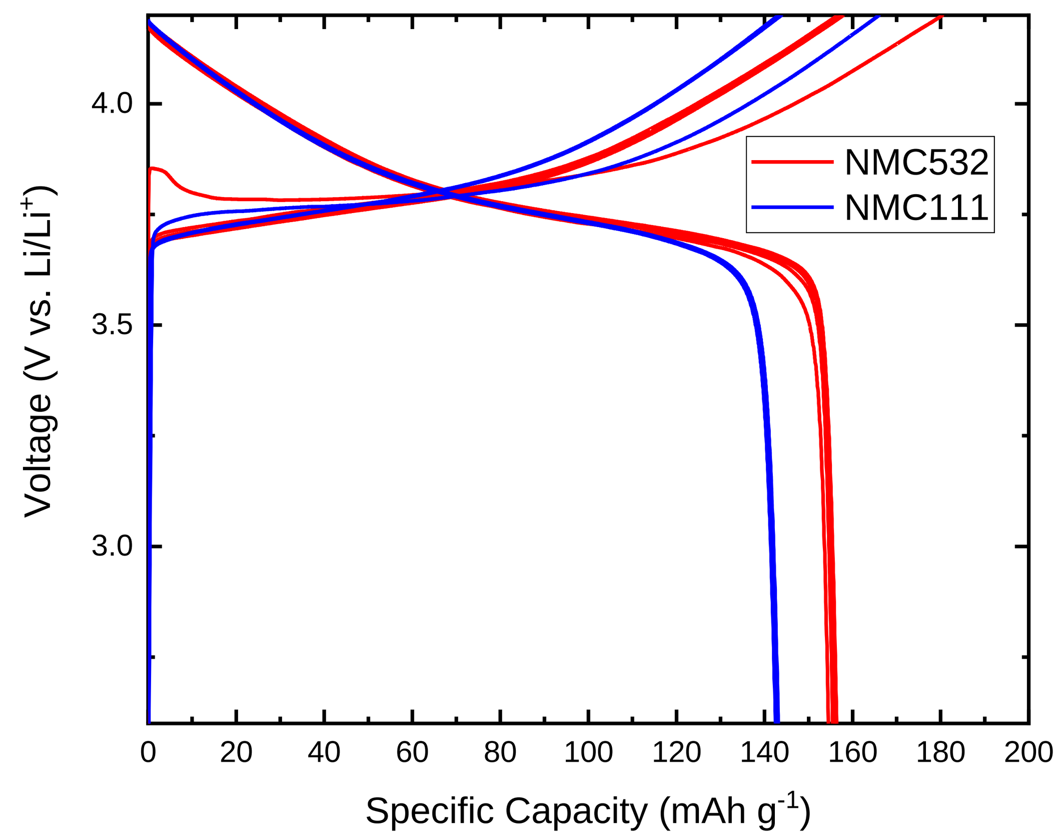
Figure 1 The low-temperature pyrolysis creates a surface carbon coating and a reconstruction layer in a Ni-rich cathode.



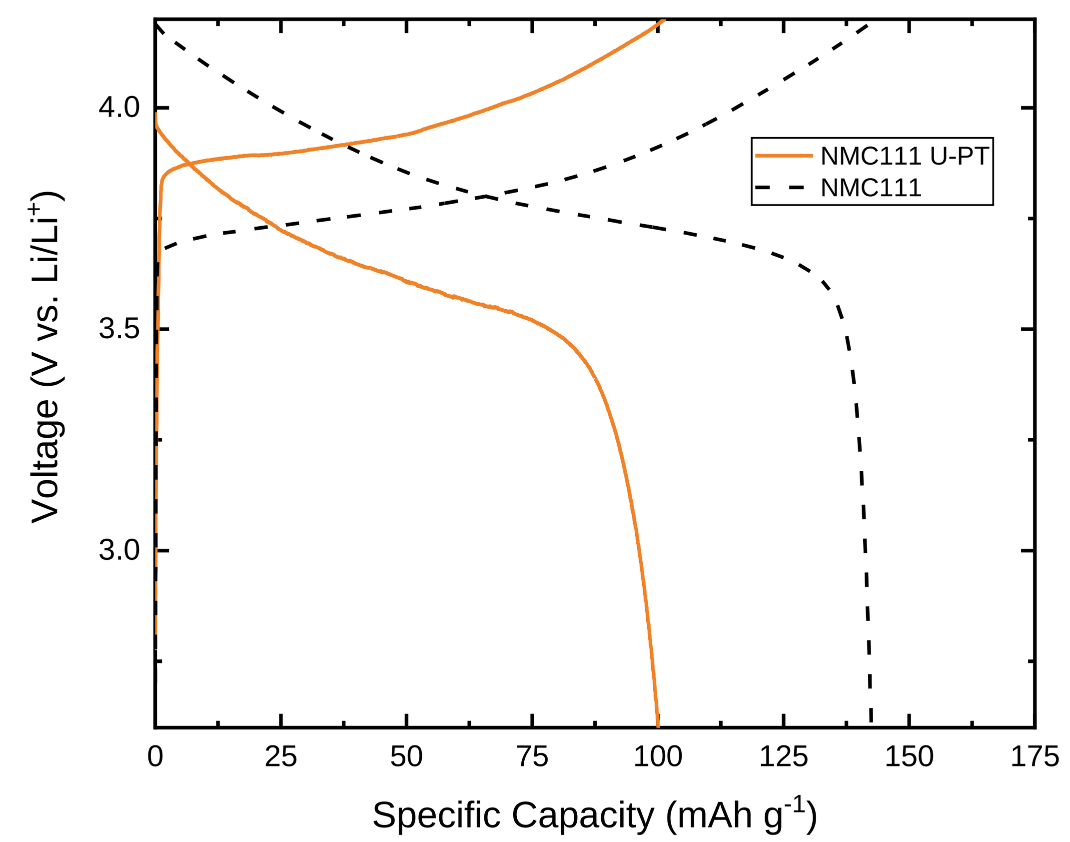
Schematic drawing of the pre-annealing process [1].

Results

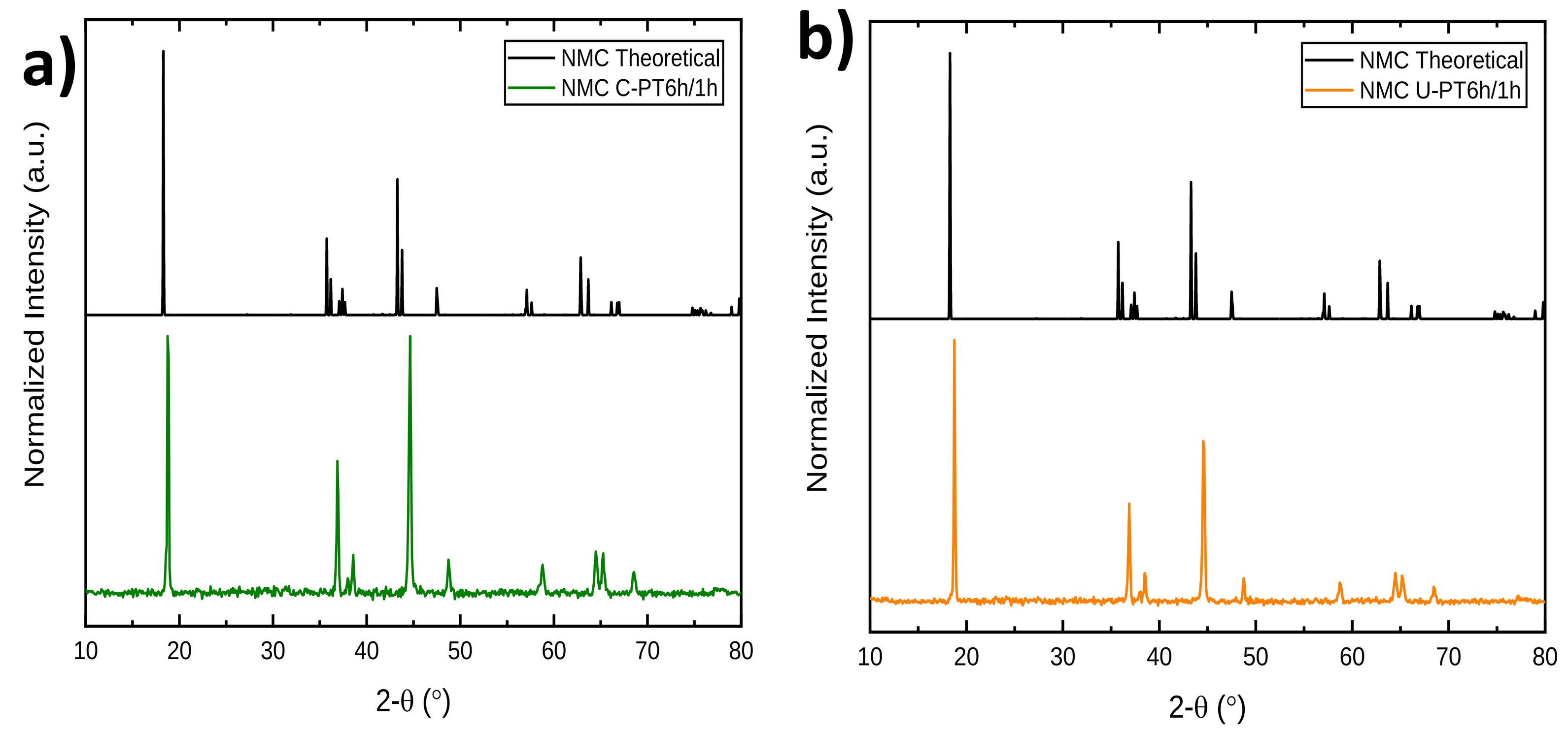
- Carbon coated NMC111 experiences a decrease in specific capacity. Pyrolysis parameters must be changed to maintain the measured specific capacity of untreated NMC111.
- Pre-treatment and longer pyrolysis time of urea increases the capacity of NMC111 coated with urea.
- NMC111 coated with cellulose must be pre-treated for cells to operate.
- Capacity fading of NMC111 coated with cellulose is very high.
- The crystal structure of NMC111 is preserved during pyrolysis.



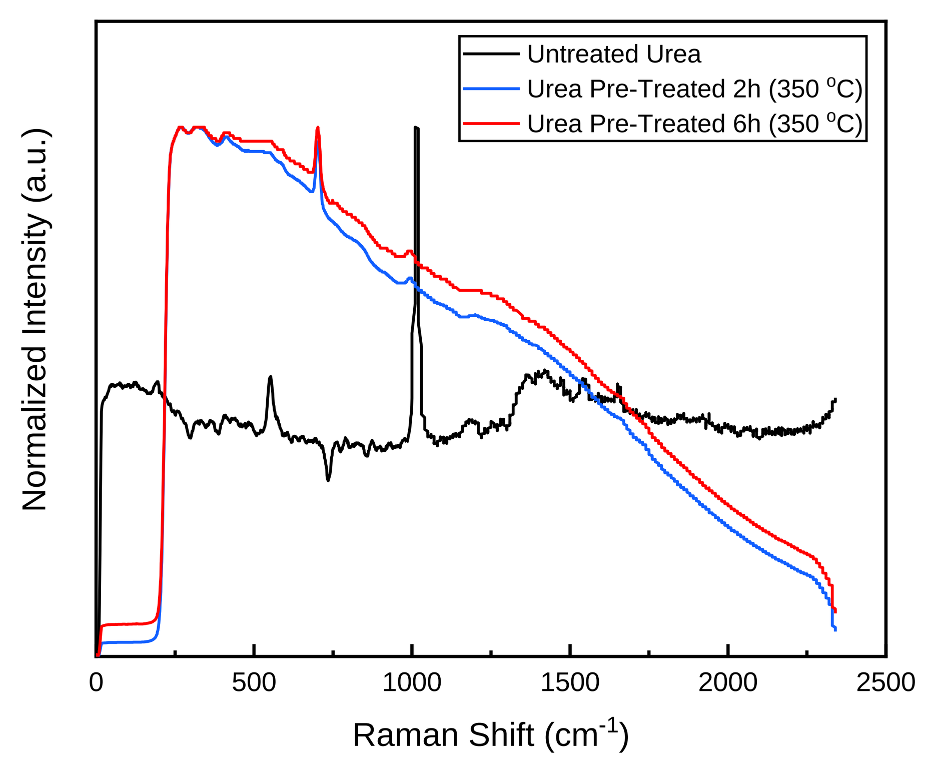
Preliminary voltage profiles of the first 10 cycles of NMC111 and NMC532.



Voltage profile of the 10th cycle of Urea Pre-Treated at 350 °C under N_2 gas for 2 hours, synthesized with NMC111 at 350 °C under N_2 gas for 2h, compared with preliminary data of pure NMC111.



X-Ray Diffraction (XRD) Spectra of **a)** cellulose and **b)** urea pre-treated at 350 °C under N_2 gas for 6 hours, synthesized with NMC111 at 350 °C under N_2 gas for 1 hour.



Raman Spectroscopy of urea with various pre-treatments times.

Results

Sample	Specific Capacity (mAh g-1)
Urea 111 (2h Pre-T 350 °C + 2h Pyro 350 °C)	100
Urea 111 (6h Pre-T 350 °C + 1 Pyro 350 °C)	30
Urea 111 (10h 350 °C)	90
Urea 111 (2h 350 °C)	55
Urea 111 (6h Pre 350 °C + 2h Pyro 350 °C)	55
Cellulose 111 (2h Pre-T 350 °C + 2 Pyro)	70 (one cycle)
Cellulose 111 (6h Pre-T 350 °C + 1 Pyro 350 °C)	90 (one cycle)
Cellulose 111 (10h 350 °C)	Failed
Cellulose 111 (2h 350 °C)	Failed

Next Steps

- X-Ray Photoelectric Spectroscopy (XPS)
- Cycling using BioLogic battery testing
- Optimize the pyrolysis parameters (time and temperature)

References

[1] Gang Chen *et al.* A robust carbon coating strategy toward Ni-rich lithium cathodes, Ceramics International, Volume 46, Issue 13, 2020
[2] Nutthaphon Phattharasupakun *et al.* Core-shell Ni-rich NMC-Nanocarbon cathode from scalable solvent-free mechanofusion for high-performance 18650 Li-ion batteries, Energy Storage Materials, Volume 36, 2021

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