# Tracking Internal Resistance of Single-crystalline $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$–Graphite Pouch Cells through Intermittent Current Interruption

**Art L Cleary, University of Sheffield**

Supervisors: Dr. Ashok S. Menon and Prof. Louis Piper, WMG, University of Warwick

## Abstract

The increased use of electric vehicles has led to a greater demand for high-energy-density Li-ion batteries, for which single-crystalline (SC) $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ (NMC811)–Graphite is a promising candidate.[1] However, prolonged cycling leads to their capacity fade, which is in part related to the growth of internal resistance. Therefore, by tracking the internal resistance evolution during cycling, the cell performance can be monitored. This can be achieved using the intermittent current interruption technique, or ICI.

## Methods

- ICI involves periodically stopping the current during galvanostatic cycling of a cell. During this stoppage, the voltage drop becomes proportional to the $\sqrt{time}$ for diffusion-controlled systems, i.e.,

$$\Delta E(t) = \Delta E(0) - I\sqrt{4R'/\pi C}\,\sqrt{t}, \text{ [1,2]}$$

As $R = -\Delta E(0)/I$ and $k = \sqrt{4R'/\pi C}$

$$\Delta E(t) = -IR - Ik\sqrt{t},$$

I → Current before the ICI step, E → Voltage, R' → Solution resistance, t → time & C → Double-layer capacity per unit length

- By extracting the slope and intercept with a linear regression fit using SciPy, time-independent/internal (R) and time-dependent/diffusion resistance coefficient (k) resistances can be obtained.
- A SC NMC811–Graphite pouch cells from the WMG pilot line was galvanostatically cycled using a Biologic BCS 9 potentiostat for 100 cycles between 2.5 and 4.4 V and at C/2 rate (1C = 200 mAh/g), with voltage-hold steps at 4.4 V. ICI steps spanning 10 s was performed every 10 min during cycling. Data was collected every 0.1 s. Linear regression fit was performed using the first 12 points excluding the first two points.[2, 3]

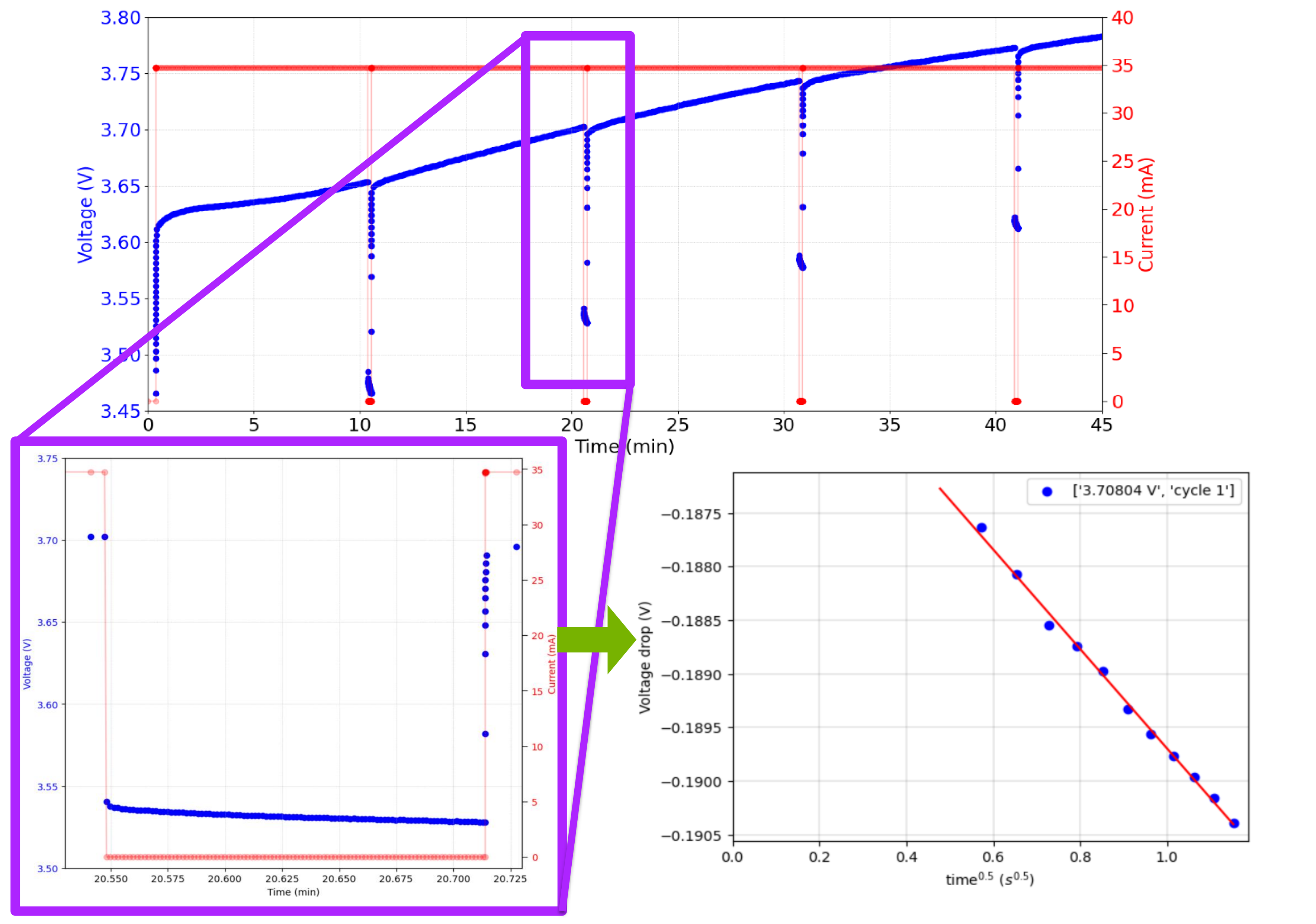



Figure 1a: Voltage (blue) evolution during ICI steps. Current is shown in red. A zoomed-in section of one ICI step is shown below. 1b: Linear regression fit of voltage vs. $\sqrt{time}$ during the ICI step.

## Observations

- ICI cycling is comparable to conventional galvanostatic cycling [3]
- With prolonged cycling overpotential and voltage hysteresis increase, as can be seen from figure 2. This is reflected in the internal resistance increase, which can be tracked using ICI.
- Figures 3A and 3B show increasing internal resistance (R) with cycle number.
- Figure 3C shows the evolution of current flow during the constant-voltage hold.
- The rate of R and the voltage-hold increase appears to reduce towards the end of cycling.

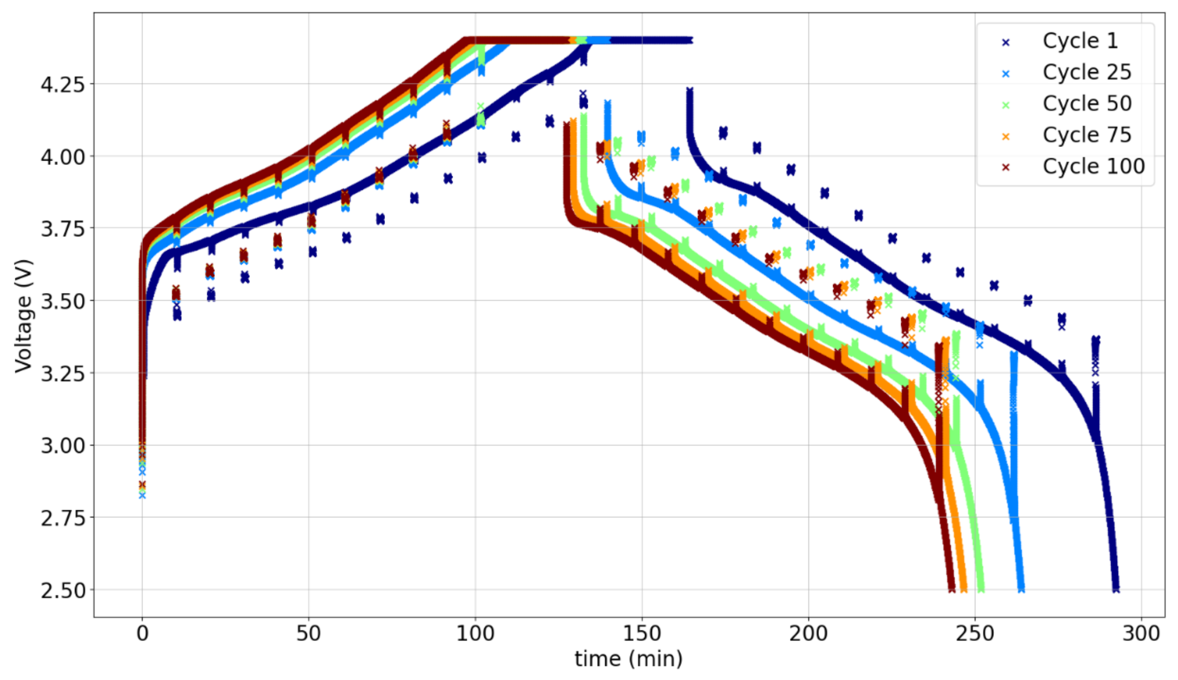



Figure 2: Pouch cell cycling data for selected cycles. The 'spikes' correspond to the ICI steps.

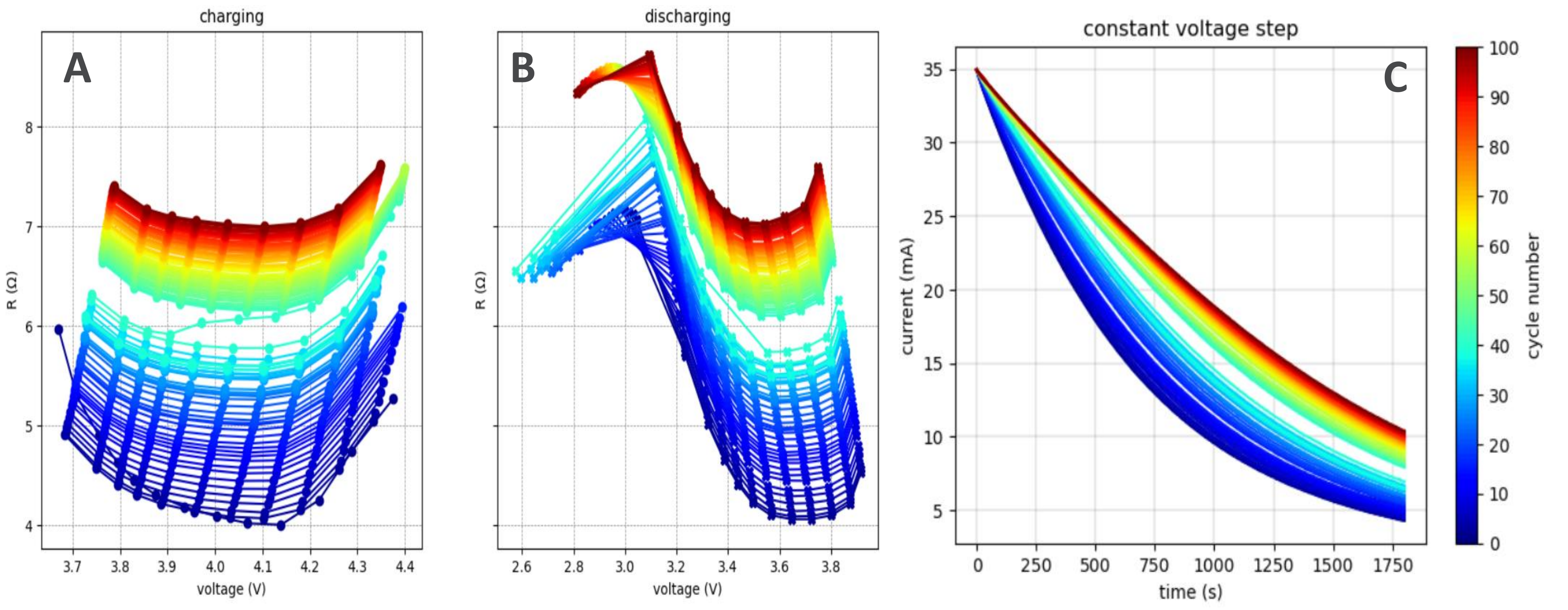



Figure 3: R obtained from ICI during charging (A) and discharging (B). C shows the current flow during the constant voltage hold.

## Analysis and conclusion

- ICI can perform real-time internal resistance tracking.
- Bulk and surface changes in the electrodes with extended cycling [4] lead to irreversible changes that reduce the cell performance, resulting in increasing R.
- This is also reflected in the voltage-hold current evolution, which shows that a higher current is required to maintain the voltage with increased cycling. This is linked to the increase in the heterogeneity in the cathode.

## References

1. DOI: 10.26434/chemrxiv-2023-zs9kp
2. Chien, Yu-Chuan. Operando Characterisation of Lithium–Sulfur Batteries. Diss. Acta Universitatis Upsaliensis, 2021
3. Electrochimica Acta 427 (2022): 140888
4. Journal of Energy Storage 57 (2023): 106211

## Future steps

- Extend this to other battery chemistries
- Implement ICI cycling in conventional battery cycling during the periodic capacity-check cycles
- Apply this method to the 3 electrode full cells to understand how the individual resistance evolution occurs on each electrode

## Intern bio

Art L Cleary is a physics student going into 3rd year at the University of Sheffield. During his studies he has gained an interest in energy storage and data analysis.

*Email = artlcleary@gmail.com, LinkedIn =* in

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