Characterizing the Structure of Lithium Metal Batteries using Local Ultrasonic Resonance Spectroscopy (LURS)

Matt Webster[1], Daniel Perey[1], Yi Lin[2], and Erik Frankforter[1]

[1] Nondestructive Evaluation Sciences Branch
[2] Advanced Materials and Processing Branch
NASA Langley Research Center, Hampton, VA

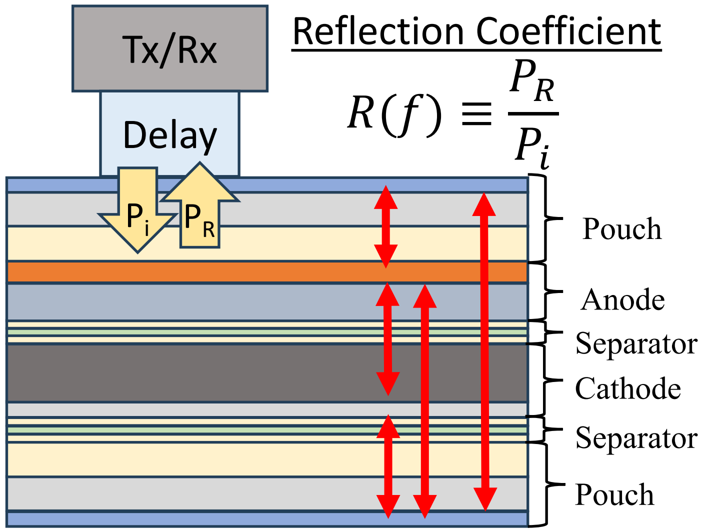
Motivation

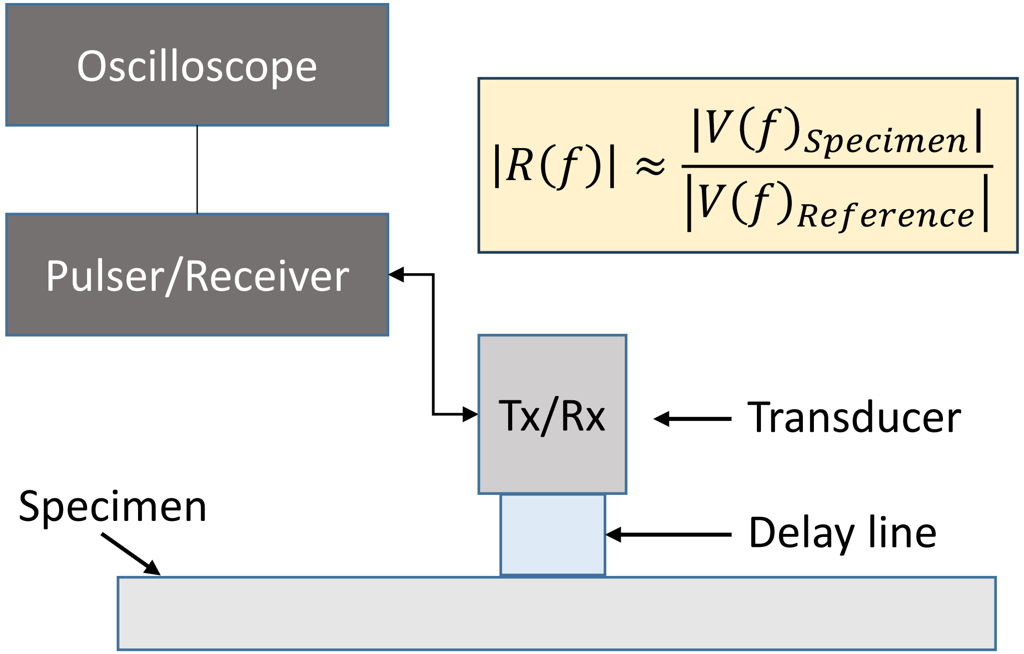
- Emergent energy demands require higher energy density batteries
- Batteries need to be safe and reliable
- Safety and performance are both influenced by the electrode structure
- Research needs:
  - Studying electrode aging mechanisms
  - Monitoring for off-nominal conditions
  - Manufacturing quality control

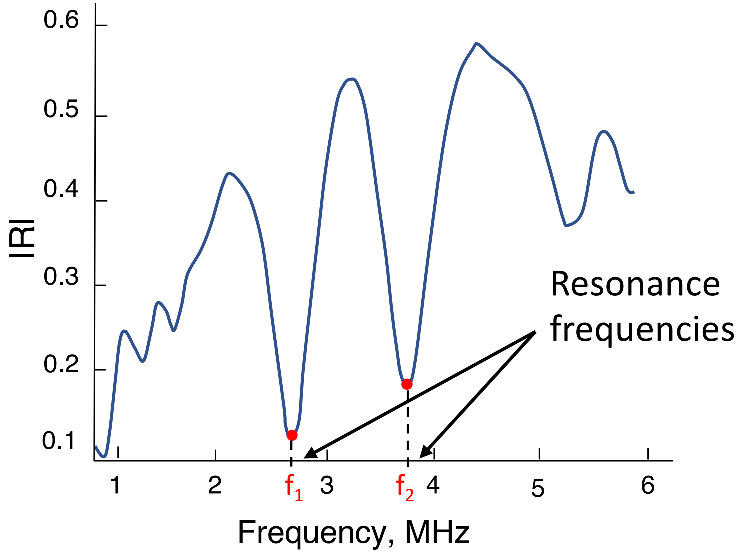
Local Ultrasonic Resonance Spectroscopy (LURS)

- Acoustic resonances occur at each frequency that produce a wavelength that is 2x the spacing between two reflectors

- LURS demonstrated by Rus and Grosse[1] on plate specimens and modified for battery inspection by Nelson[2]







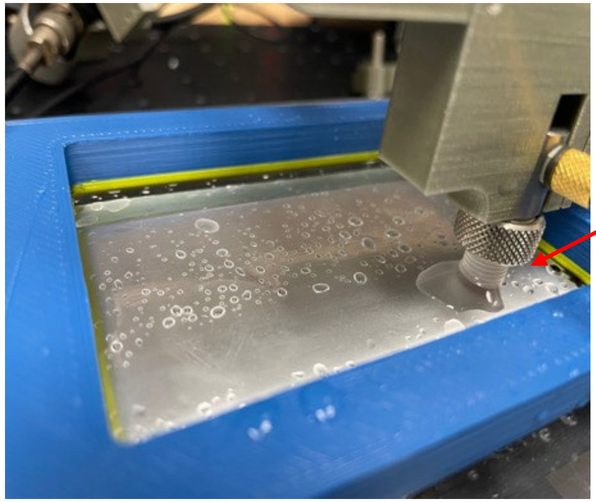
[1] J. Rus and C. Grosse, “Local Ultrasonic Resonance Spectroscopy: A Demonstration on Plate Inspection”, *J. NDE*, 2020.

[2] W. Nelson, "Local Ultrasonic Resonance Spectroscopy of Lithium Metal Batteries for Aerospace Applications." *Master’s Thesis, University of Virginia* (2021).

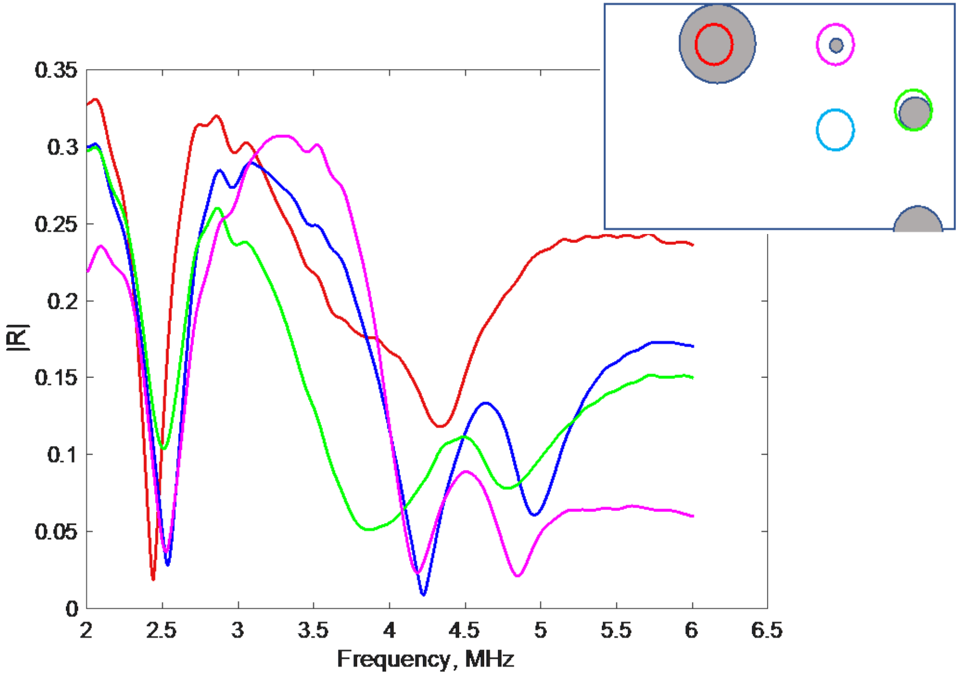
Sensitivity to Seeded Defects

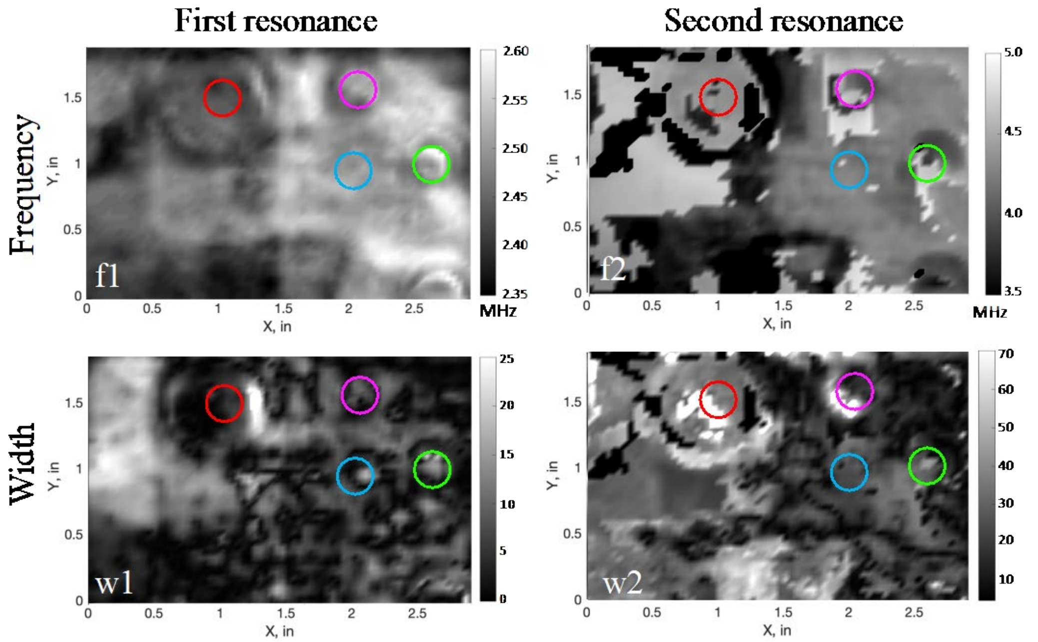
- Prior work demonstrated sensitivity to seeded lithium defects[3]
  - Contact transducer with polymer delay line

- Peak tracking approach shown for producing maps of specific resonance features



Transducer with delay line

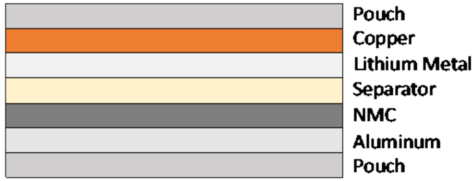


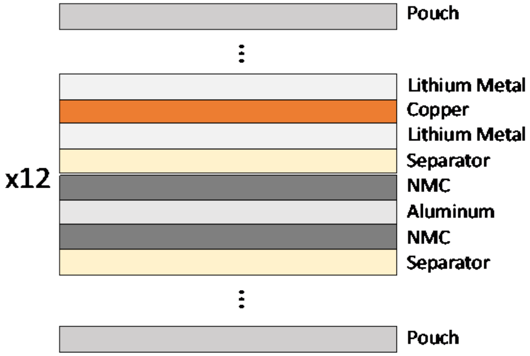


[3] Webster, M., Frankforter, E., and Juarez, P., "Evaluation of Ultrasonic Battery Inspection Techniques." *Proc. of SPIE Smart Structures and NDE, 2023.*

Cell Fabrication and Cycling

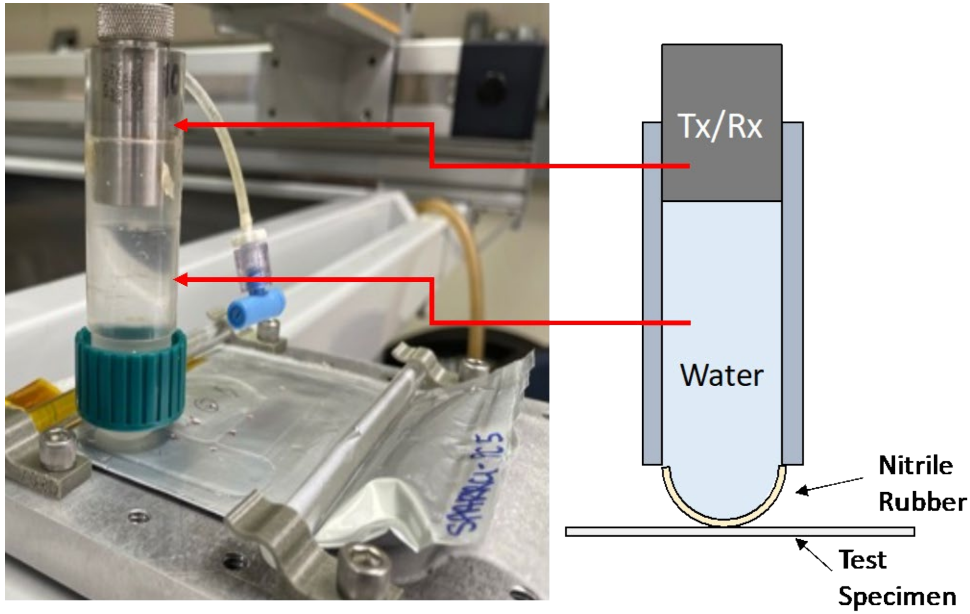
- Custom lithium metal pouch cells manufactured for testing
  - Lithium metal anode
  - Lithium nickel manganese cobalt oxide (NMC) cathode
- Single electrochemical cell specimens constructed by hand
- Multi-layer cells manufactured on a production line
- Cells cycled under constant-current conditions at charge rates (C-rates) from 0.1C – 2C

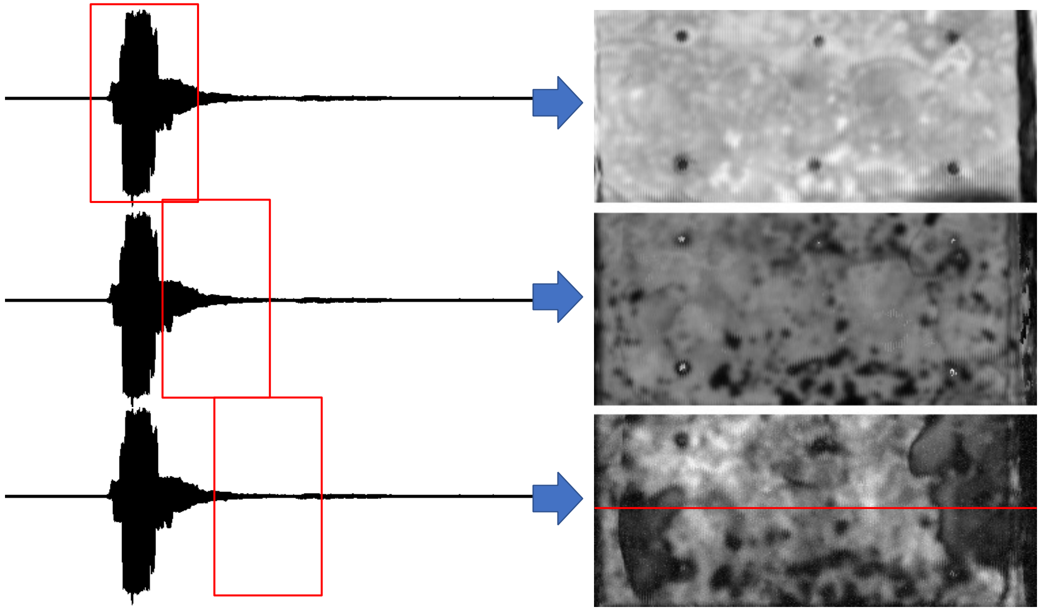


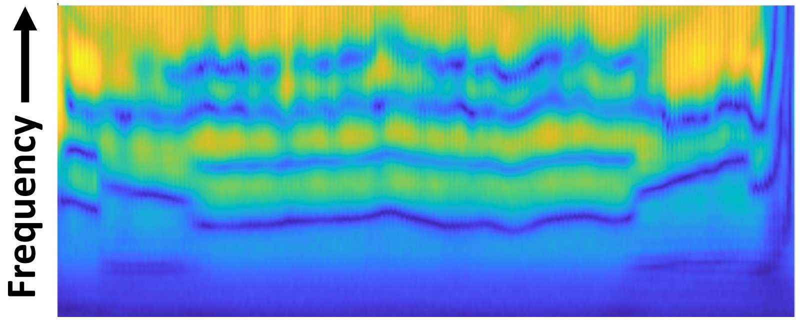


Cell Ultrasonic Inspections

- LURS performed using an ultrasonic transducer in a captured water column
  - Scans made over a 3”x2” region with 0.1” spatial resolution
- Reference measurement made using transducer in contact with glass reflector
- Full waveform data from each point processed to extract amplitude and resonance data



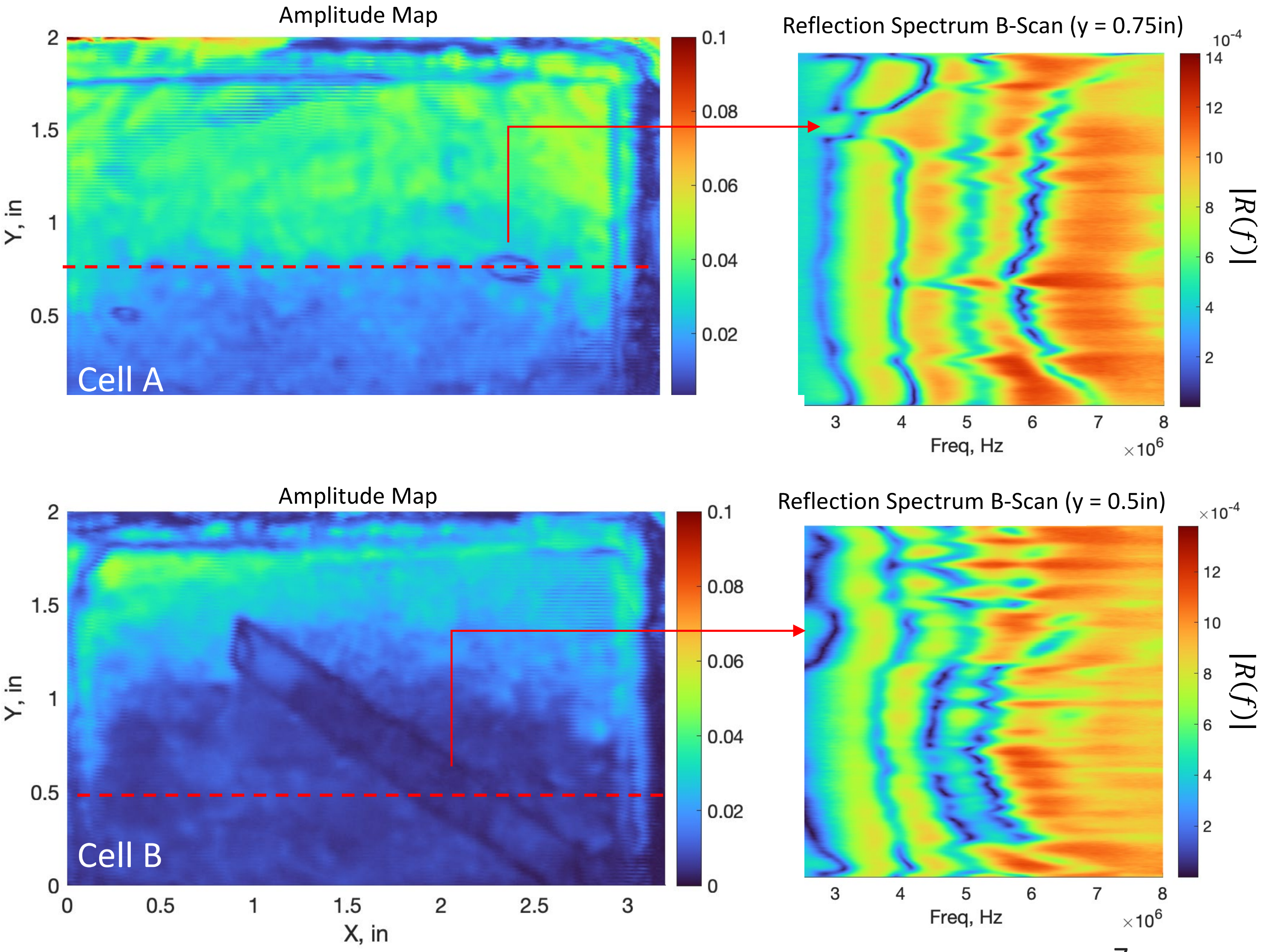




Frequency domain along red line in the amplitude C-Scan

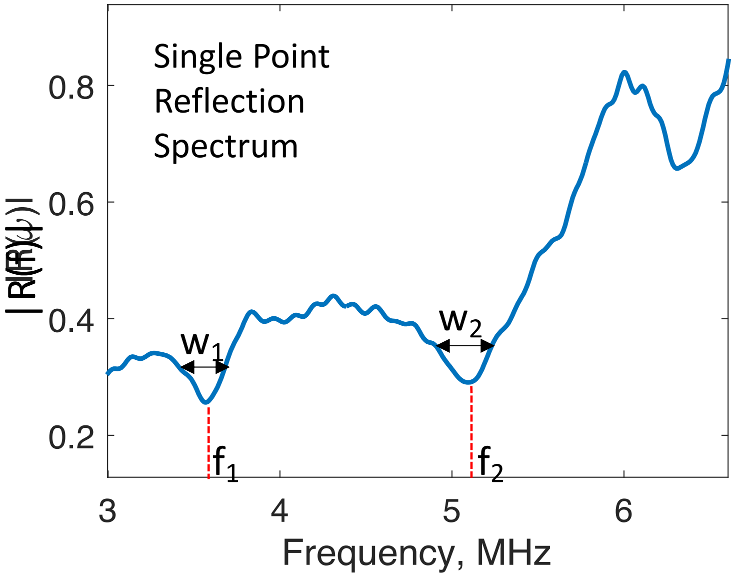
Manufacturing Defects

- Many cells contained manufacturing defects
- These often could be seen in amplitude-based C-scan data
- Resonance data provides insight into the type of defect, size, and location



Non-Uniform Stack Pressure

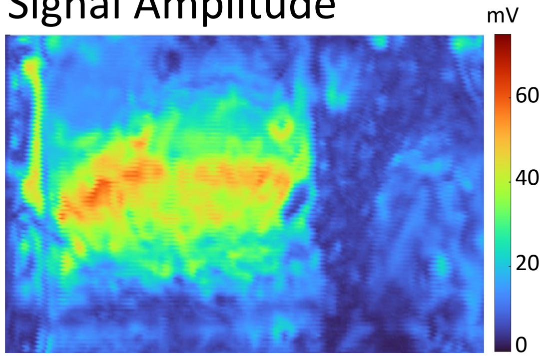
- Local changes in stack pressure altered electrode activity
- At end of life, significant heterogeneity observed in the anode
- LURS supplements amplitude data to provide details on finer features



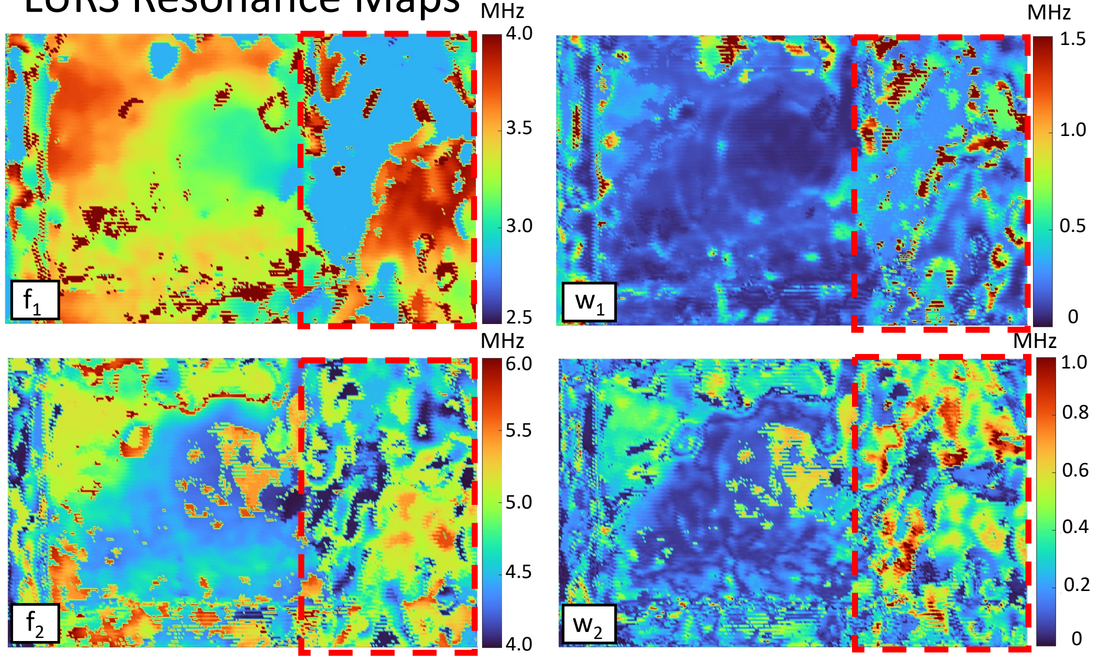
End of life Anode Structure

No Pressure

Signal Amplitude

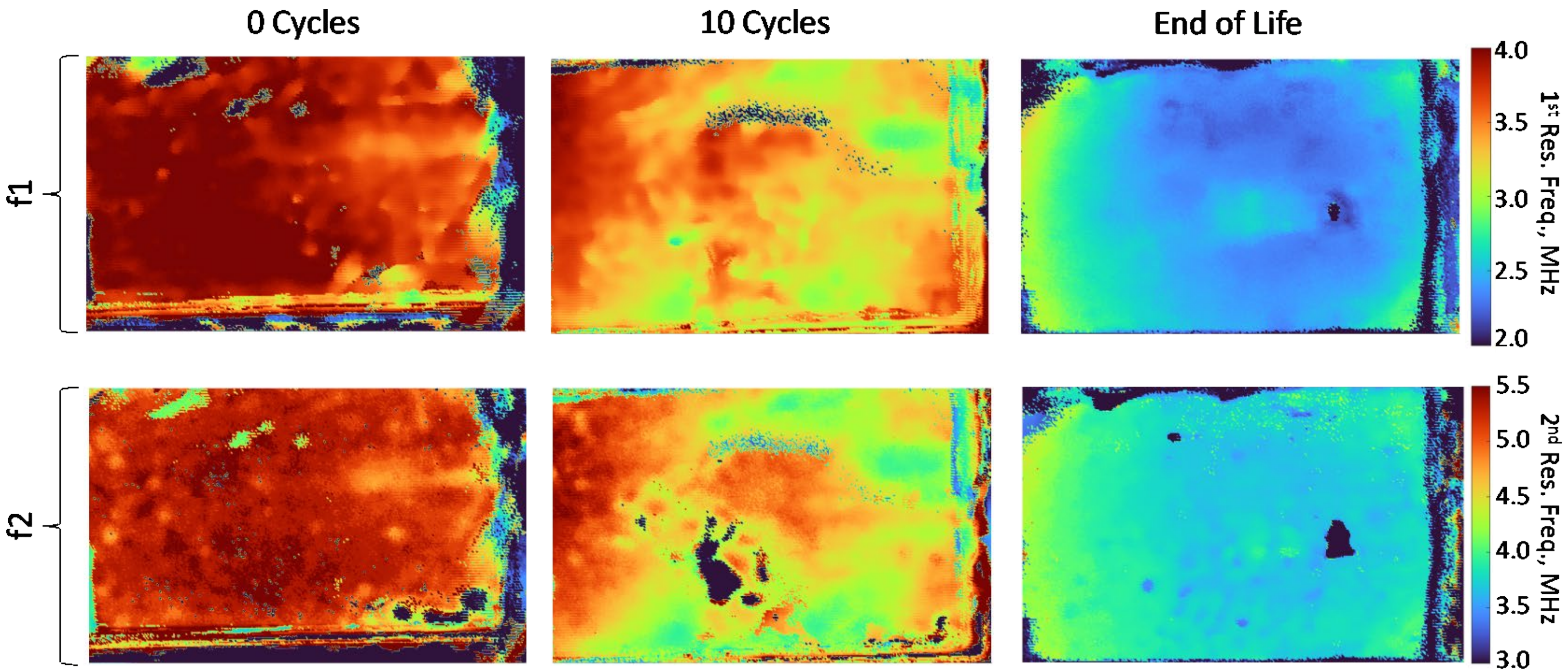


LURS Resonance Maps



Assessment of Cell Aging

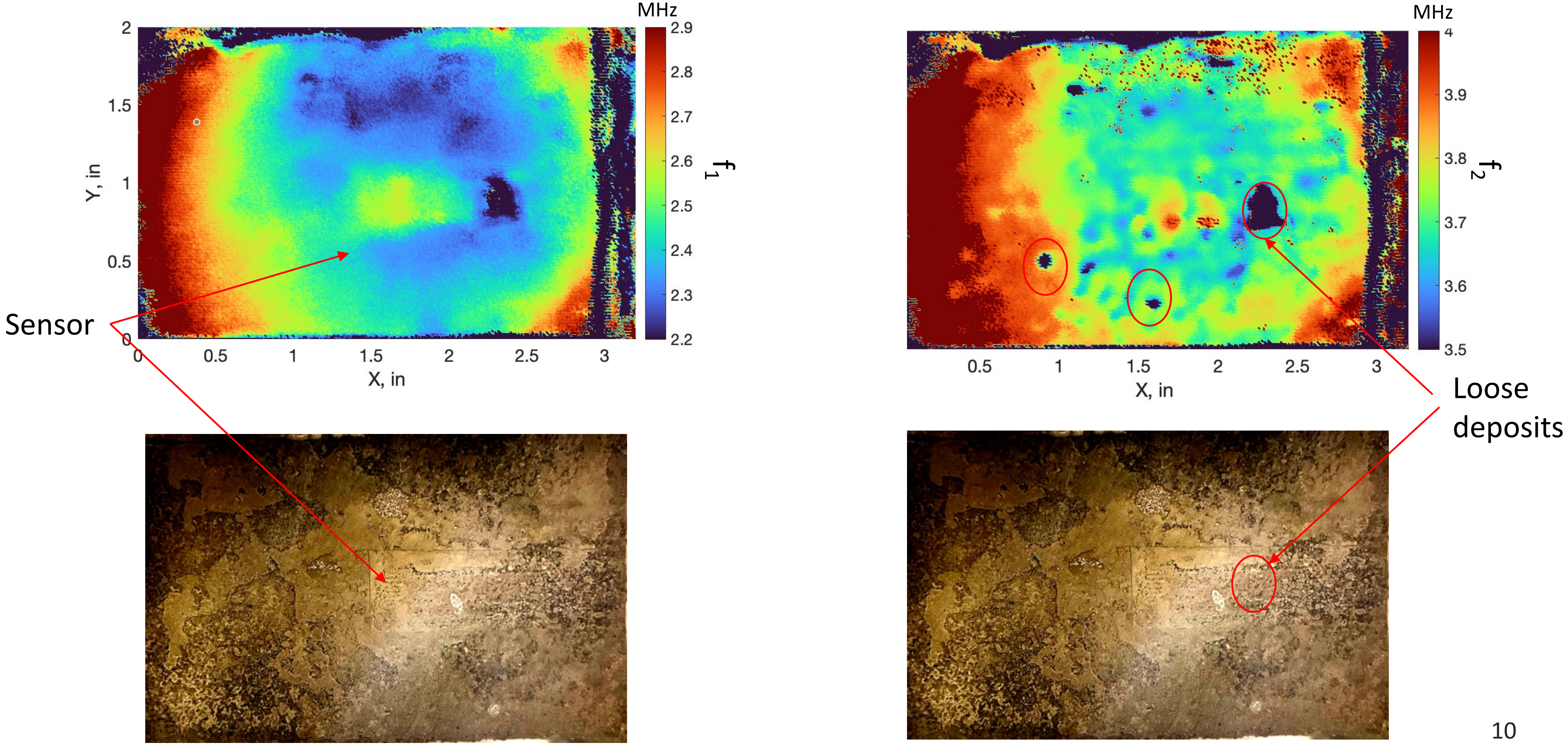
- Cell aged at room temperature at a charge rate of 2C
- Shift to lower resonance frequencies noted as degradation layer builds up on the anode surface



Anode surface at end of life

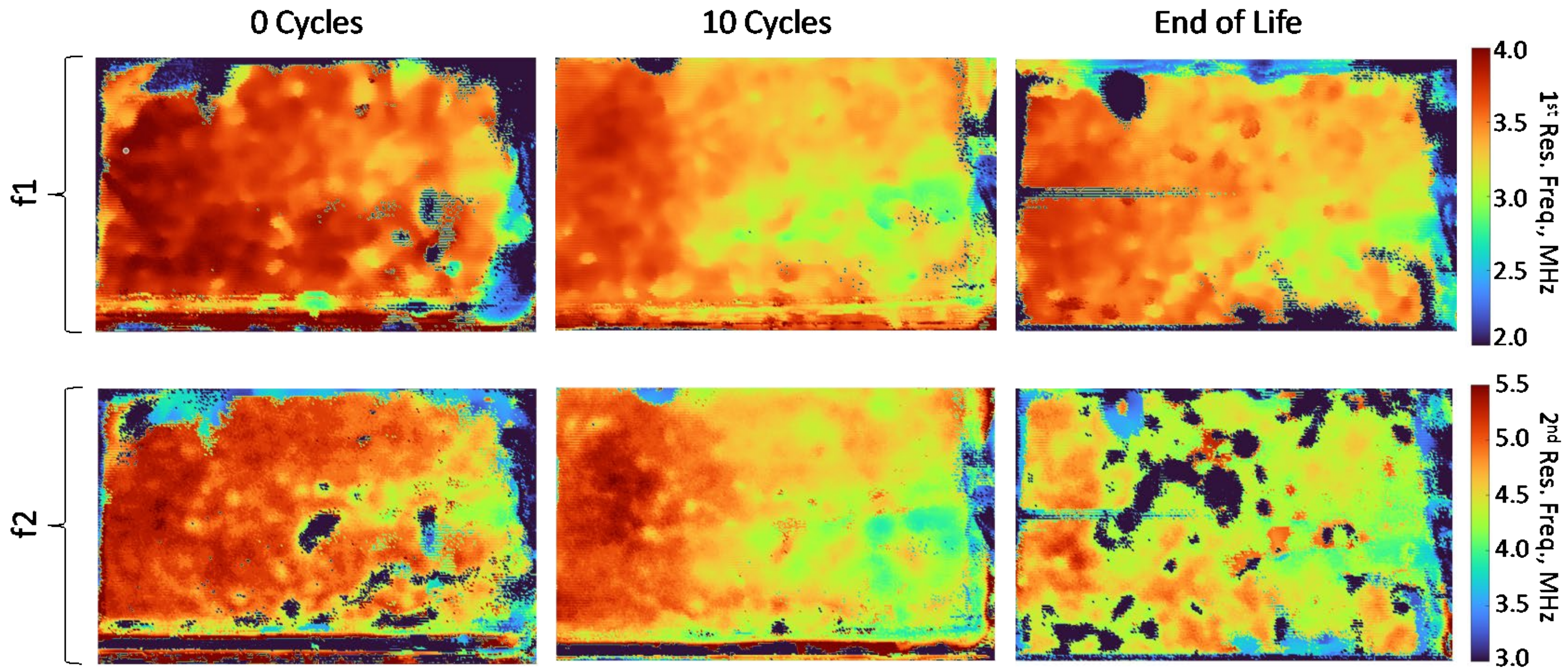
Assessment of Cell Aging

- Re-scaled resonance maps from end of life



Assessment of Cell Aging

- Cell cycled under low temperature (-20°C) condition at 0.5C
- Much less build up of degradation on the anode surface resulting in less change in average resonance



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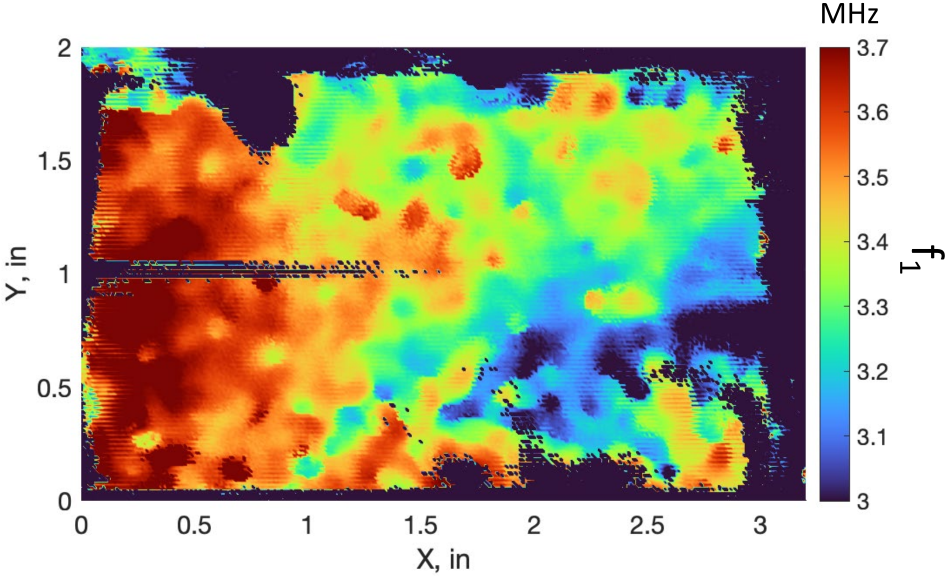
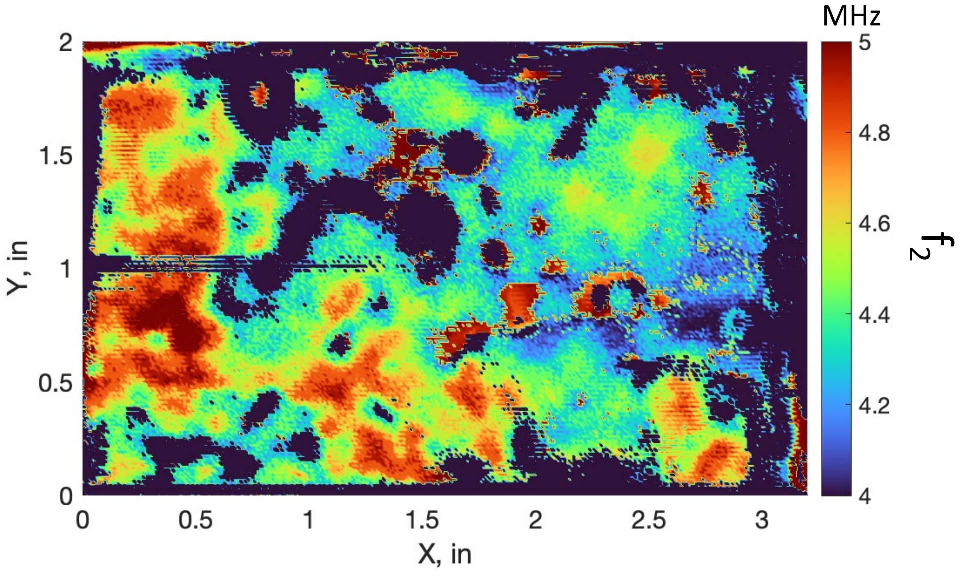
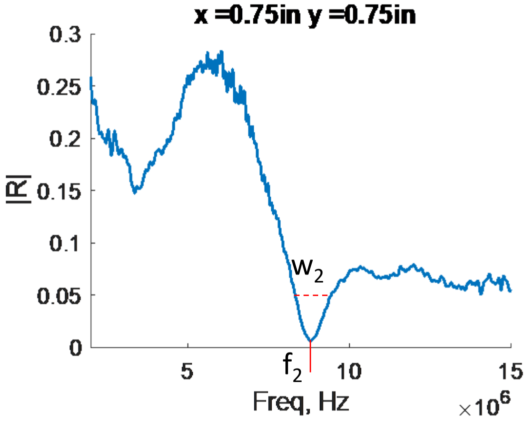


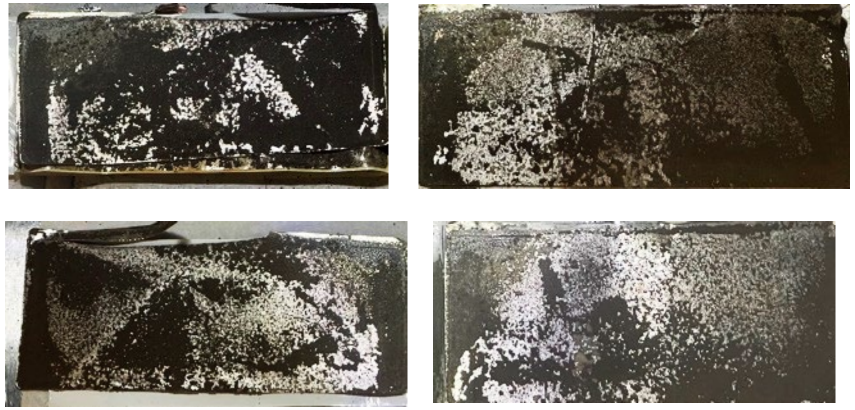


Multi-layer cells

- Multi layer cells inspected at end of life show significant irregularity in electrode structure

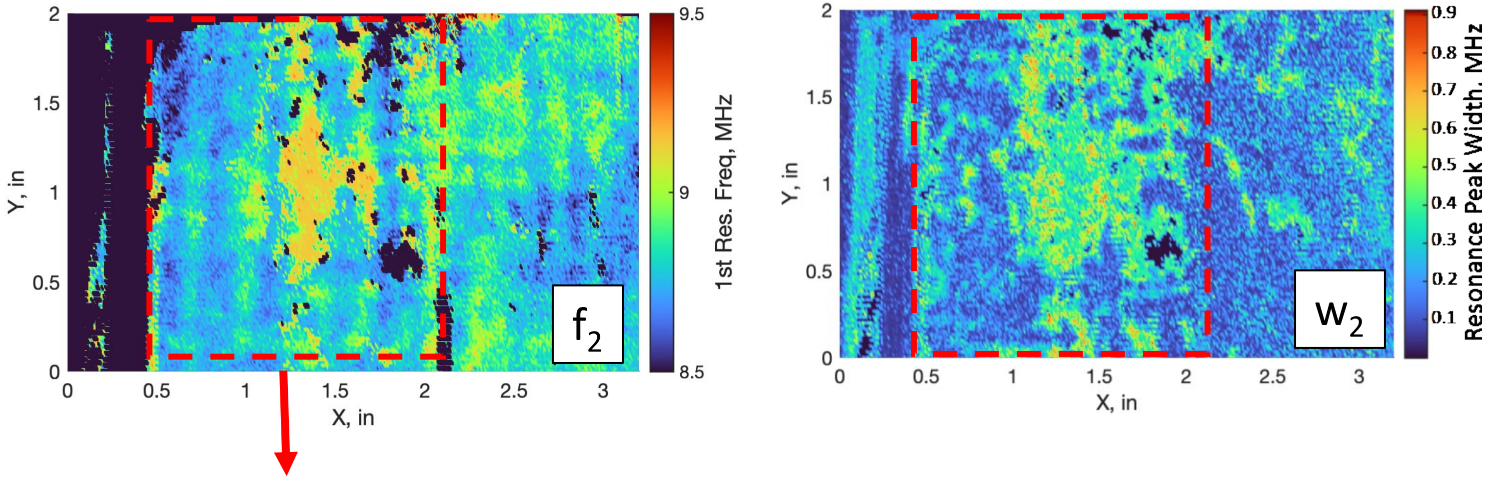
- Resonance influenced by multiple layer interactions, but show hot spots of increased scattering and irregularity





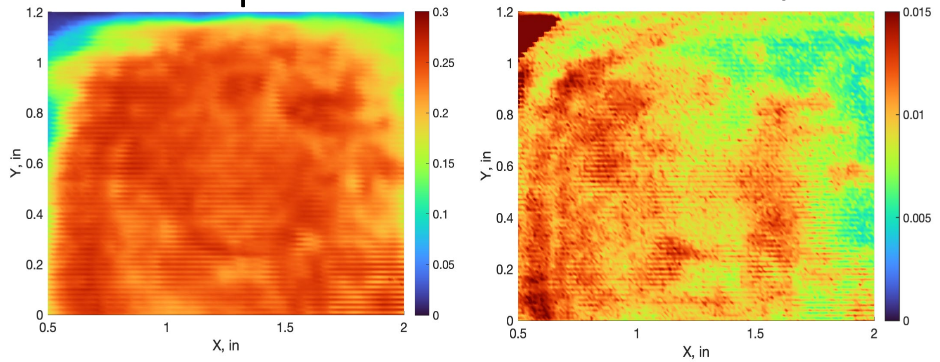
Example anode surfaces at end of life

Resonance Maps



Peak Amplitude

Post-Peak Amplitude



Conclusions

- LURS approach is sensitive to changes in battery structure both from manufacturing defects and aging
- Resonance data supplements amplitude-based analysis, providing additional information on the location, sizing, and type of defect
- Modeling is critical to fully understand the implications of resonance spectra on underlying structure in multi-layer cells
- Future work includes in-situ application for prognostics and quantitative structural analysis based on resonance data