

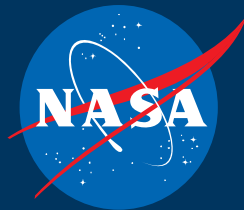
# Microstructure quantification and random forest regression models for $\text{Li}_4\text{Ti}_5\text{O}_{12}$ -Ni property prediction

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EMA 2021 – S1: Characterization of Structure–Property Relationships in Functional Ceramics  
1/22/21



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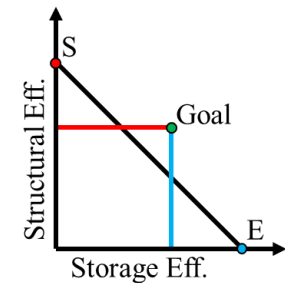
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# Motivations

- Next-generation electric propulsion systems for aerospace applications
  - Increased efficiency, reduced emissions, and noise abatement
- Two primary limitations in energy storage:
  - Insufficient energy density
  - Safety concerns
    - Flammable organic liquid electrolytes
- Inherently safe all-solid-state architectures
  - prevent Li-dendrite induced failure
  - Improve energy density (remove parasitic components)
- Multifunctional Energy Storage
  - Active battery materials in load bearing paths
    - Electrochemically and mechanically active
  - Systems level weight and volume savings
  - Distributed storage and power delivery
- Ideal microstructure for multifunctionality is unknown
  - all-solid-state battery vs structural ceramic



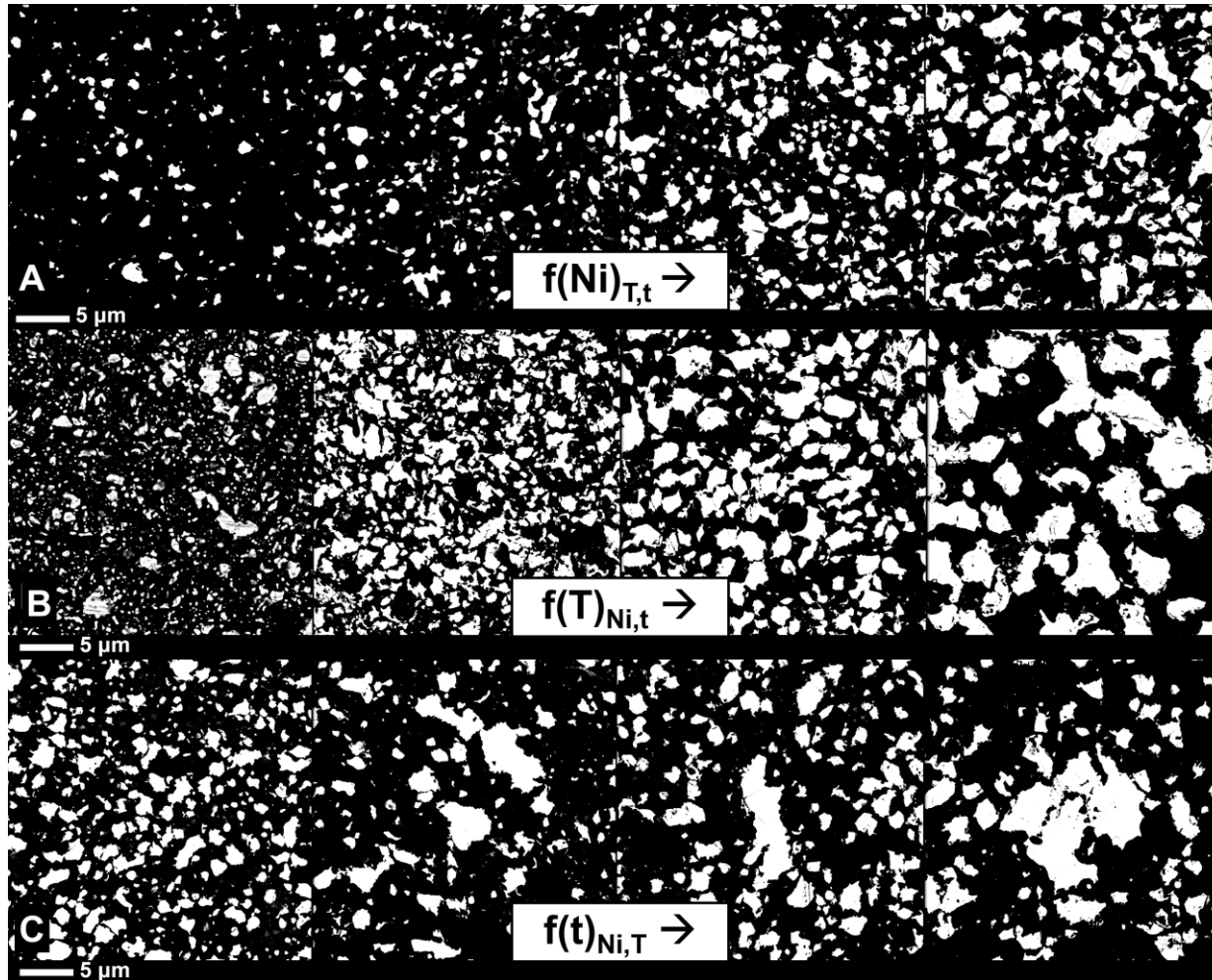
[1] <https://www.nasa.gov/centers/armstrong/Features/leaptech.html>



# Sample Matrix

High contrast SEM: black:  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  matrix, white: nickel metal current collector

38 unique Temp/Hrs/Ni conditions  
101 samples  
369 SEM images



Composition [Ni. Vol.]

0, 5, 10, 20, 24, 28, 35, 40, 50 vol% Ni

Temp. [°C]

700°C, 800°C, 900°C, 1000°C, 1100°C

Time [Hrs]

5, 10, 50, 150 hrs

Huddleston W, Dynys F, Sehirlioglu A. *J Am Ceram Soc.* 2020; 103:4178-4188.  
<https://doi.org/10.1111/jace.17159>

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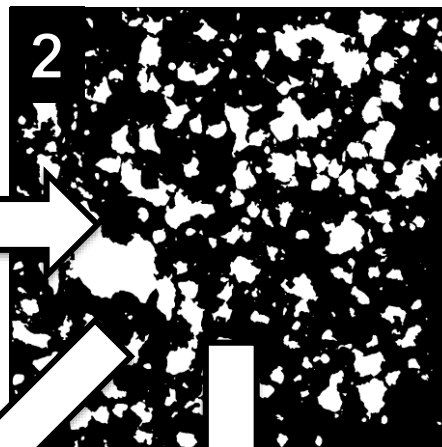
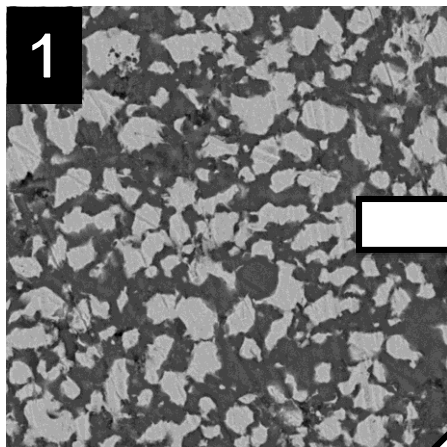
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# Methods

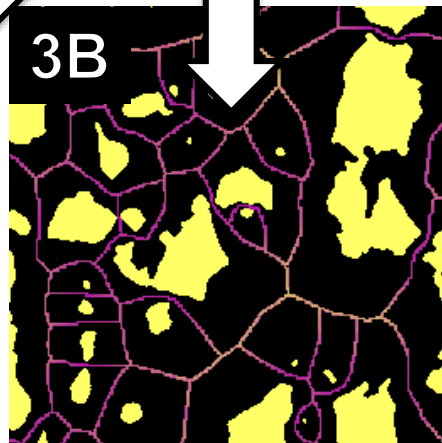


Raw SEM Image  
Bright: Ni  
Dark:  $\text{Li}_4\text{Ti}_5\text{O}_{12}$



High Contrast Image  
Particle Analyzer  
Particle Size  
Particle Location  
Count per area

Nearest Neighbor Distance  
center-to-center  
discrete



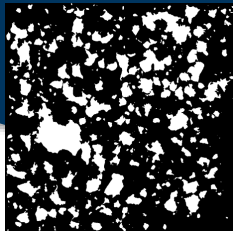
Voronoi partition map  
edge-to-edge spacing  
continuous

See also: Matt Hoffman's Poster: "Microstructural feature engineering for machine learning enabled discovery of microstructure-property relationships"

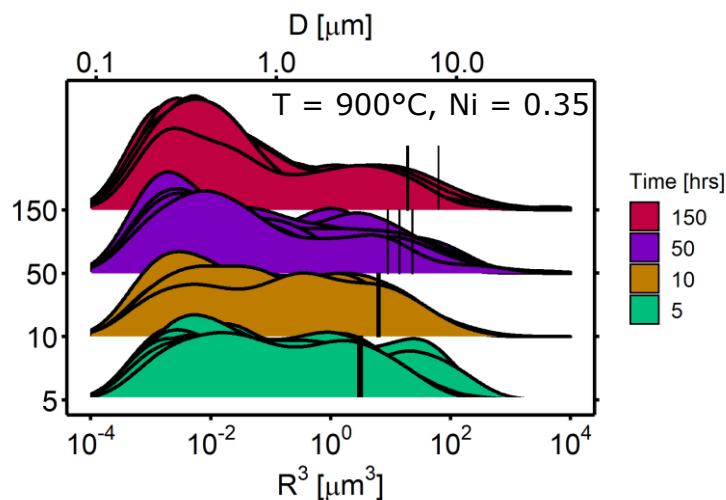
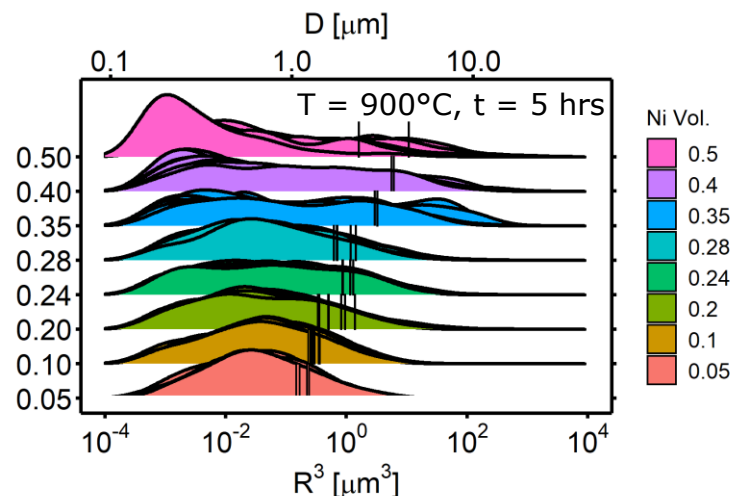
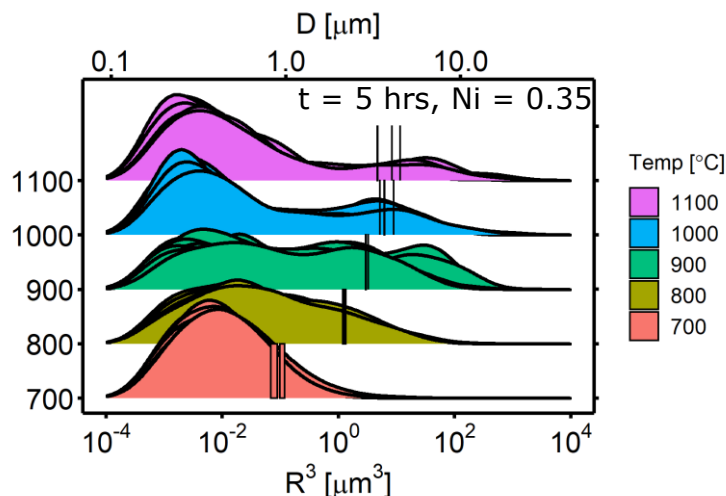
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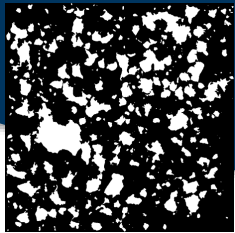


# Particle Size



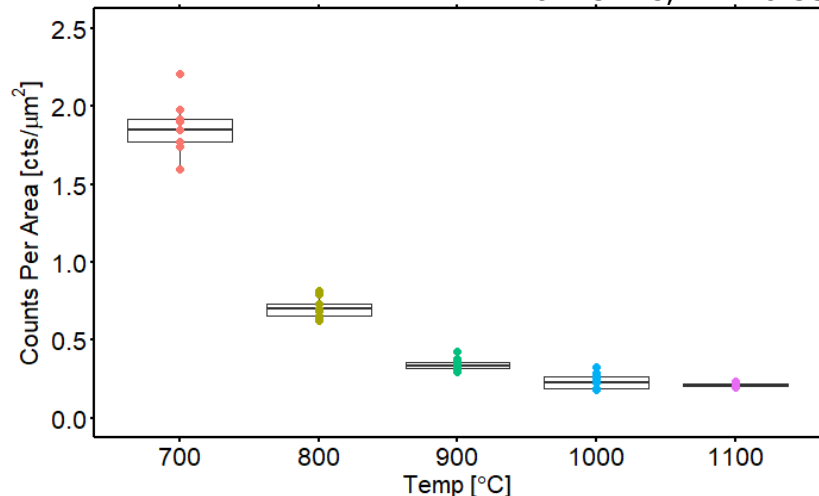
- Average Particle Size Increased
- Bimodal character Increased
  - Sintering Temperature
  - Dwell time
  - Nickel Volume Fraction
- Coarsening or Coalescence?

Plots show individual distribution curves for each image  
+ Average size values shown as vertical lines

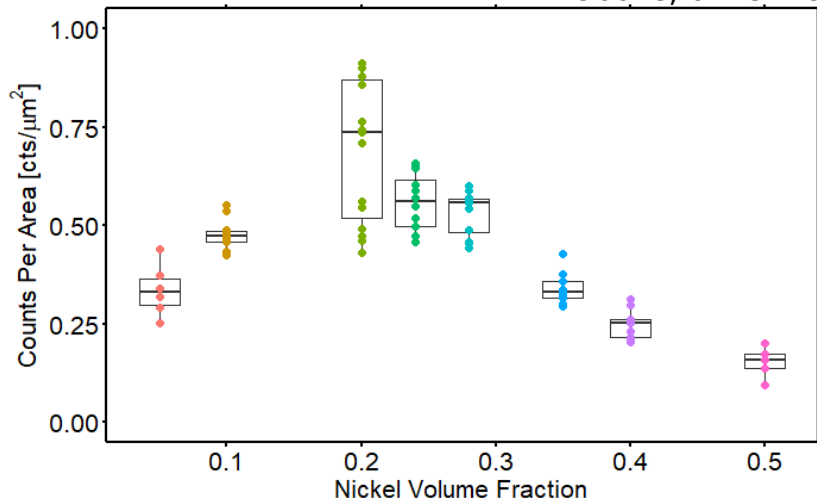


# Particle Counts

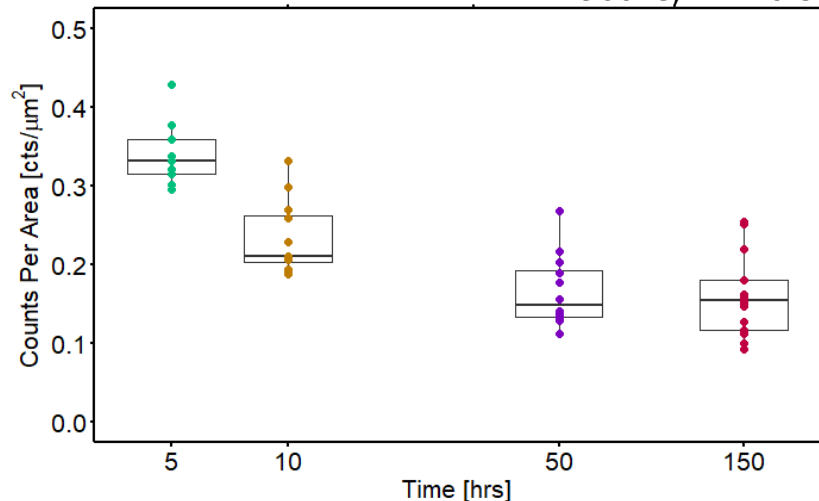
$t = 5 \text{ hrs}$ ,  $\text{Ni} = 0.35$



$T = 900^{\circ}\text{C}$ ,  $t = 5 \text{ hrs}$

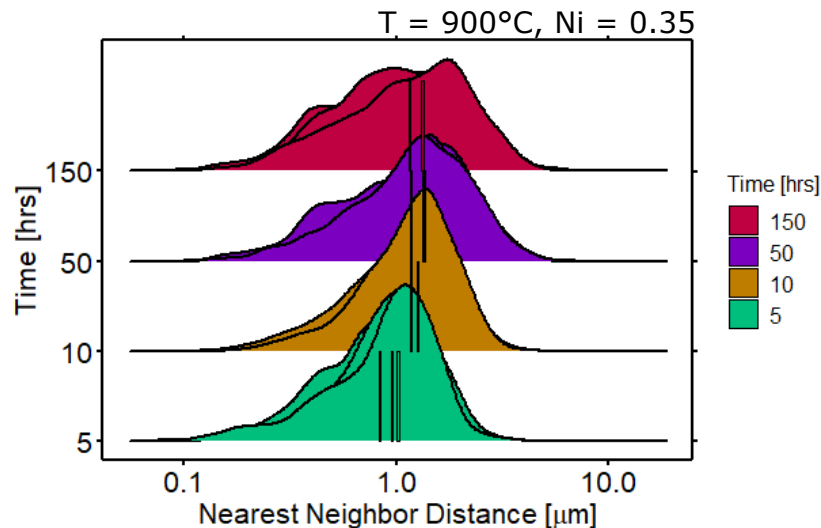
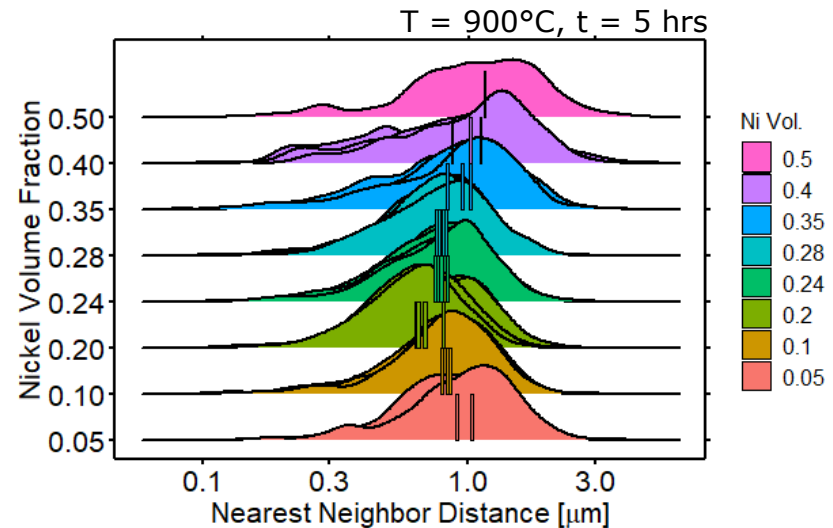
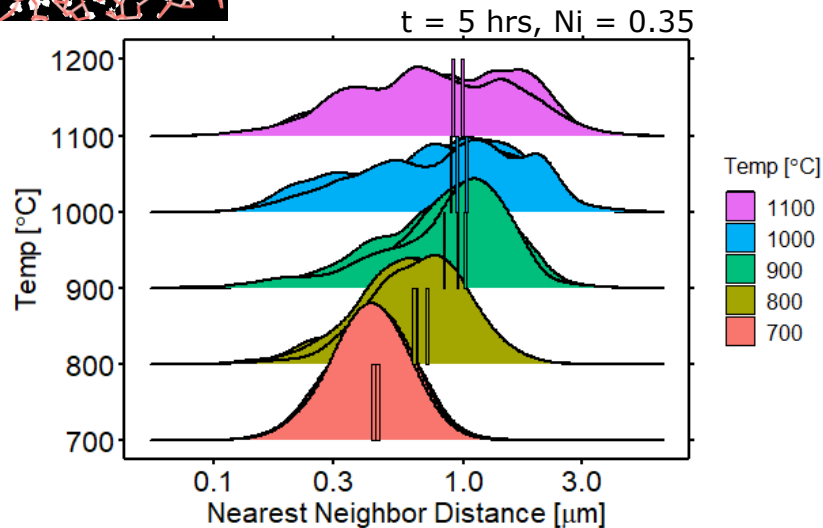
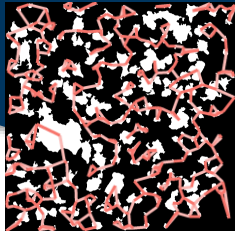


$T = 900^{\circ}\text{C}$ ,  $\text{Ni} = 0.35$



- Discrete Ni particle counts
  - Decreased with Sintering Temp. & Dwell time
  - Maximized near the Ni percolation threshold
- Little change observed at high T/t

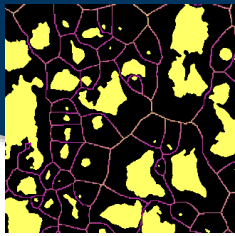
# NND



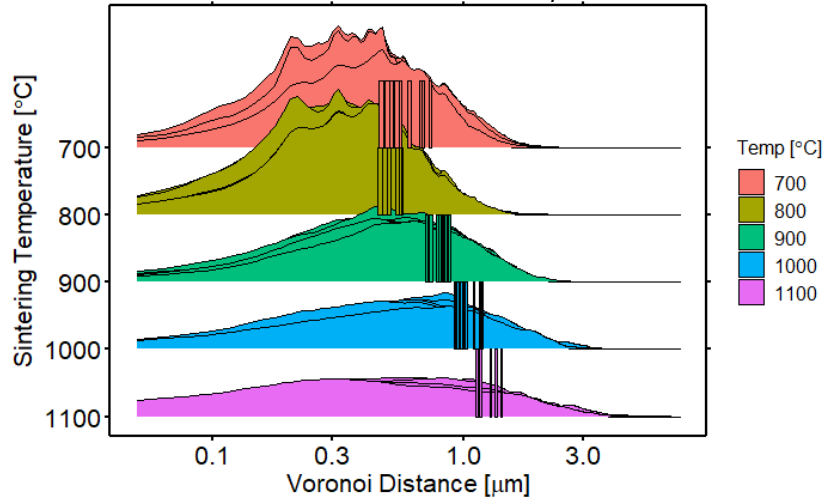
- Nearest Neighbor (center-to-center) Distances
  - Increased with Sintering Temp. & Dwell time
  - Minimized near the Ni percolation threshold
- Little change observed at high T/t

Plots show individual distribution curves for each image  
+ Average NND values shown as vertical lines

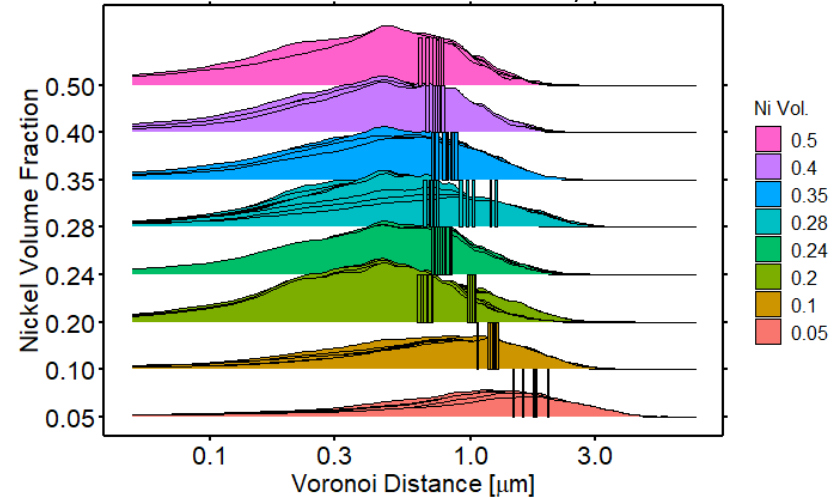
# Voronoi



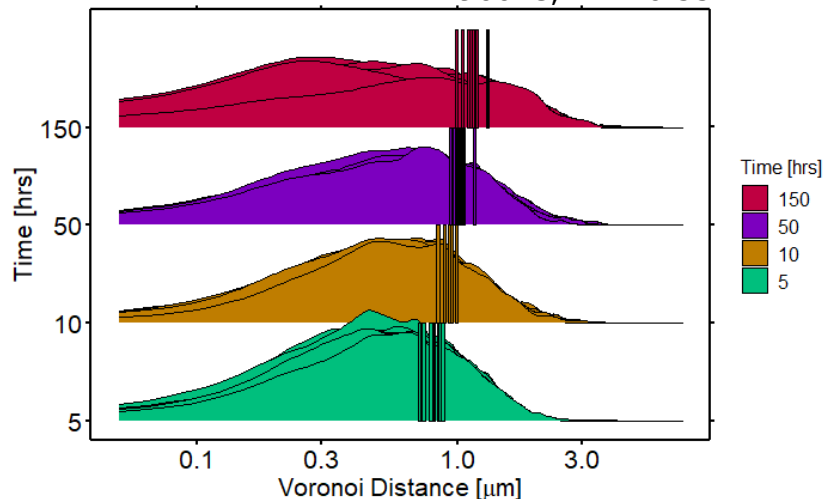
$t = 5 \text{ hrs}$ ,  $\text{Ni} = 0.35$



$T = 900^\circ\text{C}$ ,  $t = 5 \text{ hrs}$



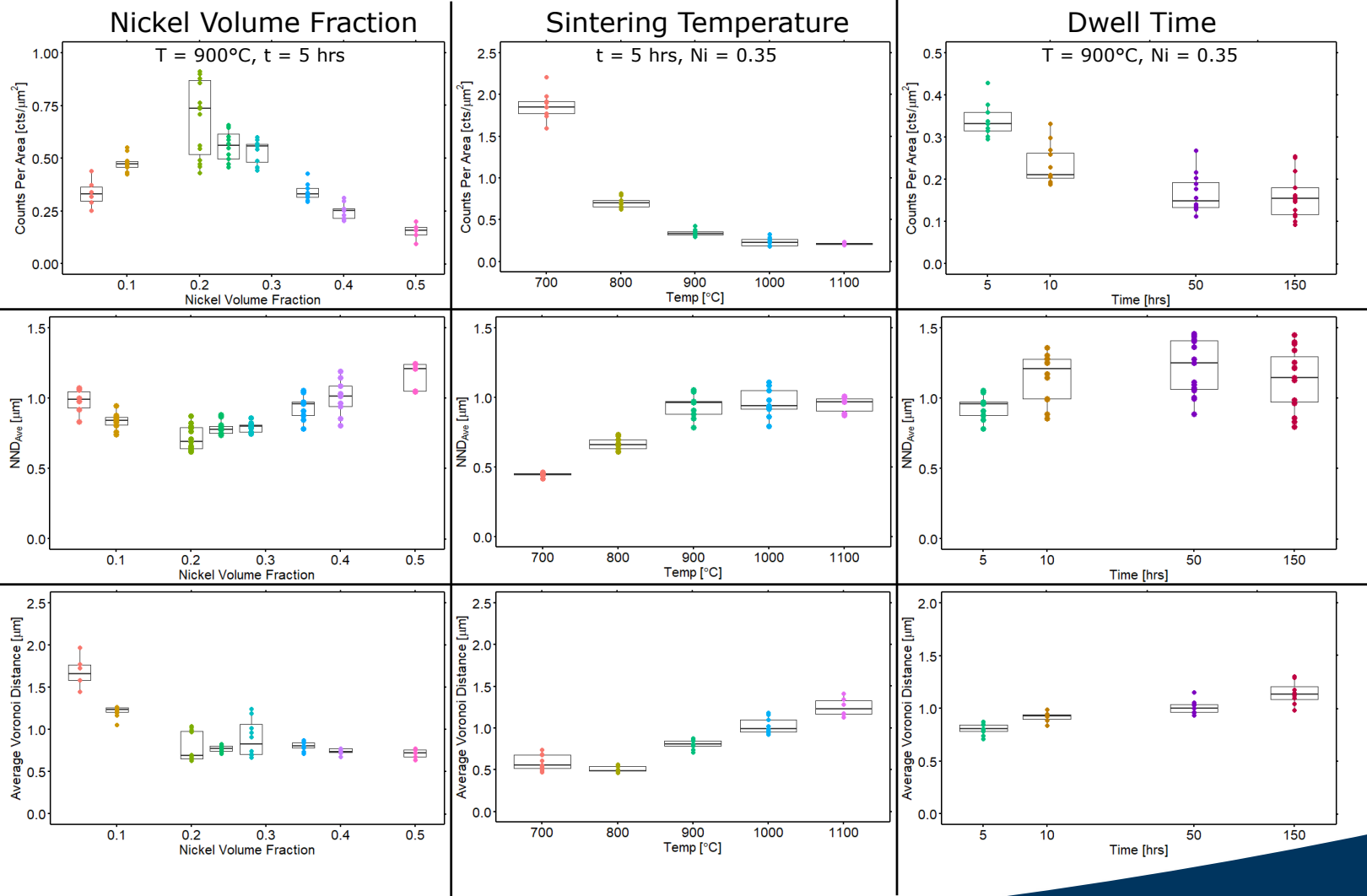
$T = 900^\circ\text{C}$ ,  $\text{Ni} = 0.35$



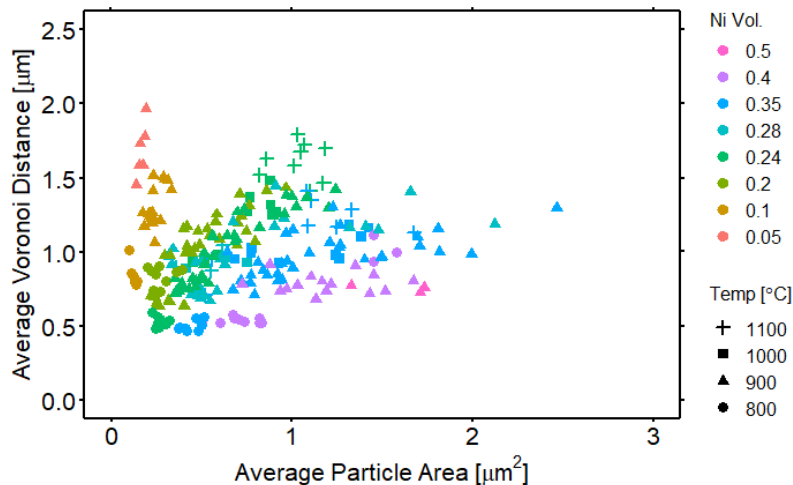
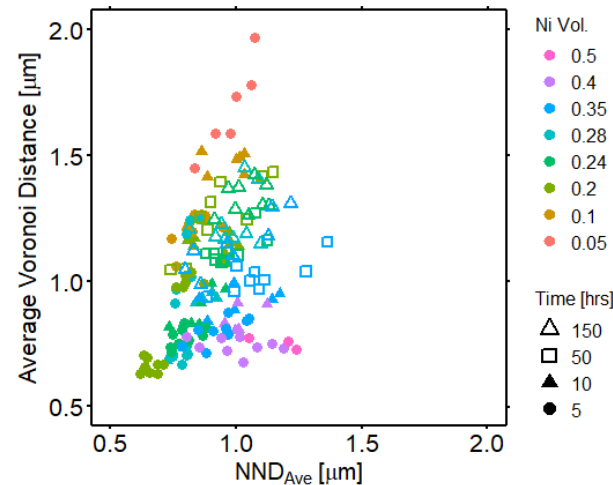
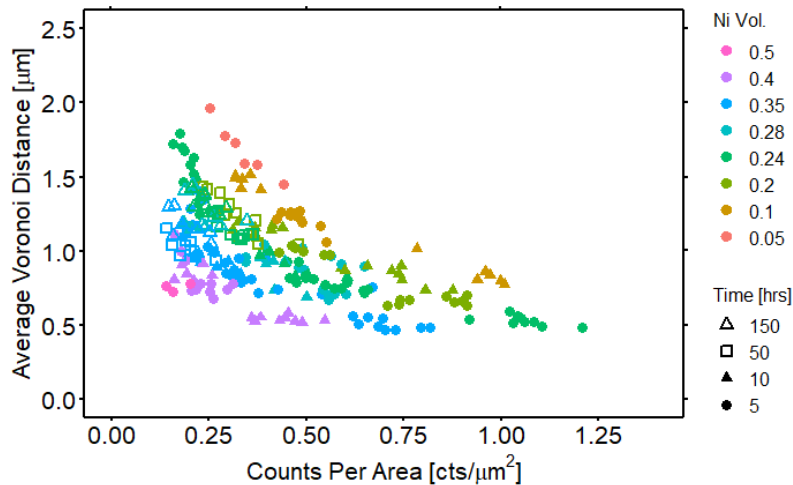
- Voronoi (edge-to-edge) Spacing
  - Increased with Sintering Temp. & Dwell time
  - Decreased with Nickel Volume Fraction
- Little change observed at low T, high Ni

Plots show individual distribution curves for each image  
+ Average VD values shown as vertical lines

# Quantifier Comparison



# Quantifier Comparison

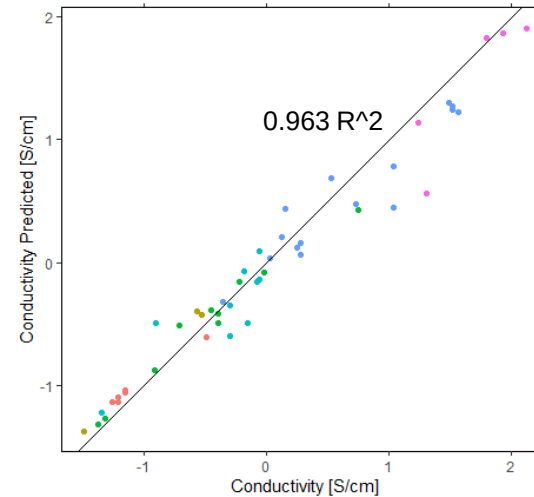
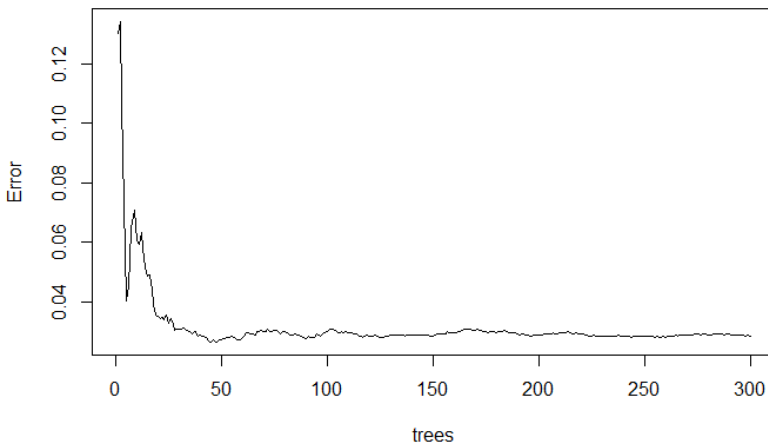


- Processing-microstructure-property landscapes
  - Compare microstructure quantifiers
  - Beyond individual slices of the matrix
  - Compare all data together
  - Observe additional trends
- Predict properties from microstructure?

# Conductivity Random Forest

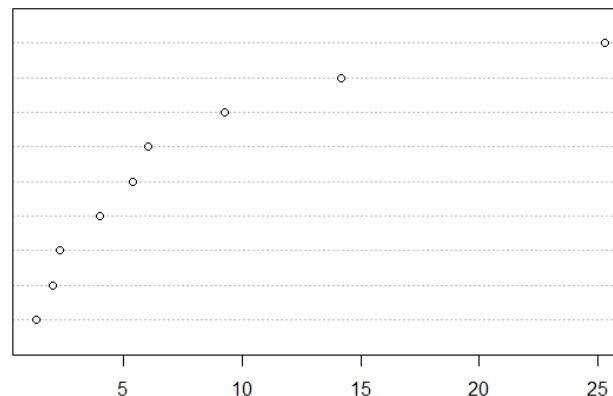
Data Split: 70% Training - 30% Testing

Conductivity Random Forest - 97.13 % Var Explained



Conductivity RF

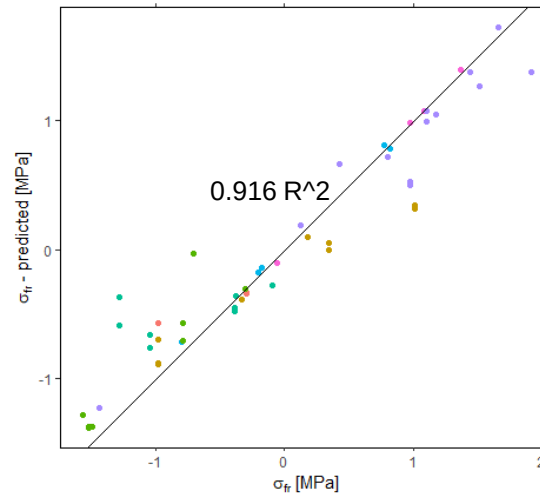
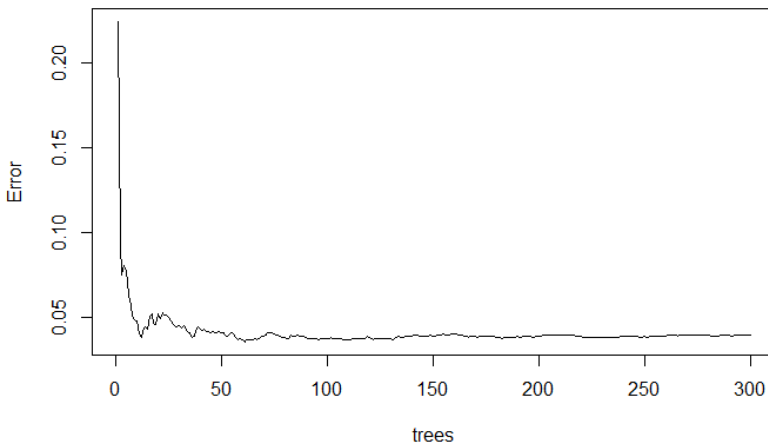
Total Nickel Area Coverage  
Average Nickel Particle Area  
Voronoi Map Coverage  
Average Voronoi Spacing  
Count per area  
Bulk Density  
Dwell Time  
NND<sub>AVE</sub>  
Sintering Temp



# Fracture Stress Random Forest

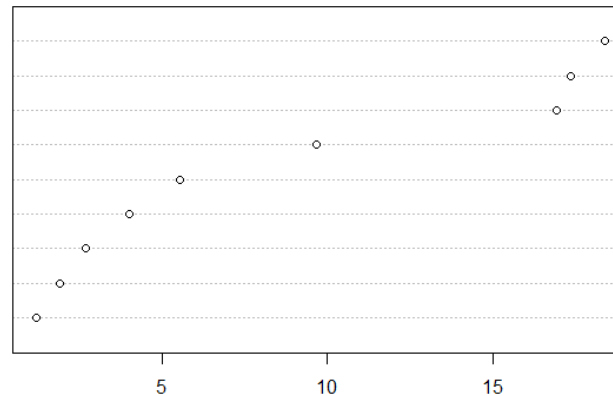
Data Split: 70% Training - 30% Testing

Stress Random Forest - 96.02 % Var Explained



Stress RF

Total Nickel Area Coverage  
Average Nickel Particle Area  
Voronoi Map Coverage  
Bulk Density  
Count per area  
Sintering Temp.  
NND<sub>AVE</sub>  
Average Voronoi Spacing  
Dwell Time

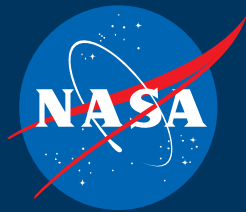


# Summary

- $\text{Li}_4\text{Ti}_5\text{O}_{12}$ -Ni composites
- Microstructures were quantified using several measures
  - Particle Size
  - Counts Per Area
  - Nearest Neighbor Distance
  - Voronoi Spacing
- Random Forest Regression Models were developed for
  - Conductivity
  - Fracture Stress
- Future work
  - Modeling from full distributions, improve over average measures

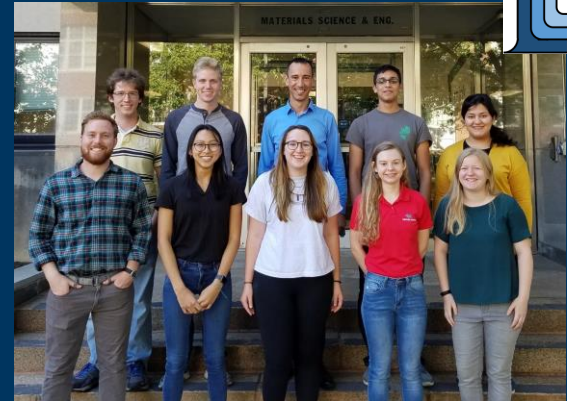
# Acknowledgements

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More Questions → [whh36@case.edu](mailto:whh36@case.edu)