Project 30: Mechanisms of Grain Refinement in Laser Powder Bed Fusion of In-Situ Metal Matrix Composite 6061 Aluminum Alloys

Spring Meeting

April 7th – 9th 2020

- Student: Chloe Johnson (Mines)
- Faculty: Amy Clarke (Mines)
- Industrial Mentors: Paul Wilson (Boeing), Clarissa Yablinsky (LANL), John Carpenter (LANL)
- Other Participants: Joe McKeown (LLNL), Jonah Klemm-Toole (Mines)

Project 30: Mechanisms of Grain Refinement in Laser Powder Bed Fusion of In-Situ Metal Matrix Composite 6061 Aluminum Alloys

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- Student: Chloe Johnson (Mines)
- Advisor(s): Amy Clarke (Mines)

Project Duration
PhD: August 2017 to May 2021

- Problem: While inoculation presents a method to eliminate hot tearing and columnar growth in additive manufacturing (AM) of aluminum alloys, the mechanisms of grain refinement under rapid solidification conditions are not well understood.
- Objective: Understand how solidification conditions, solute content, and nucleant site density affect mechanisms controlling grain refinement in inoculated alloys in AM.
- Benefit: Inform alloy design and grain size prediction for inoculated alloys used in AM solidification conditions.

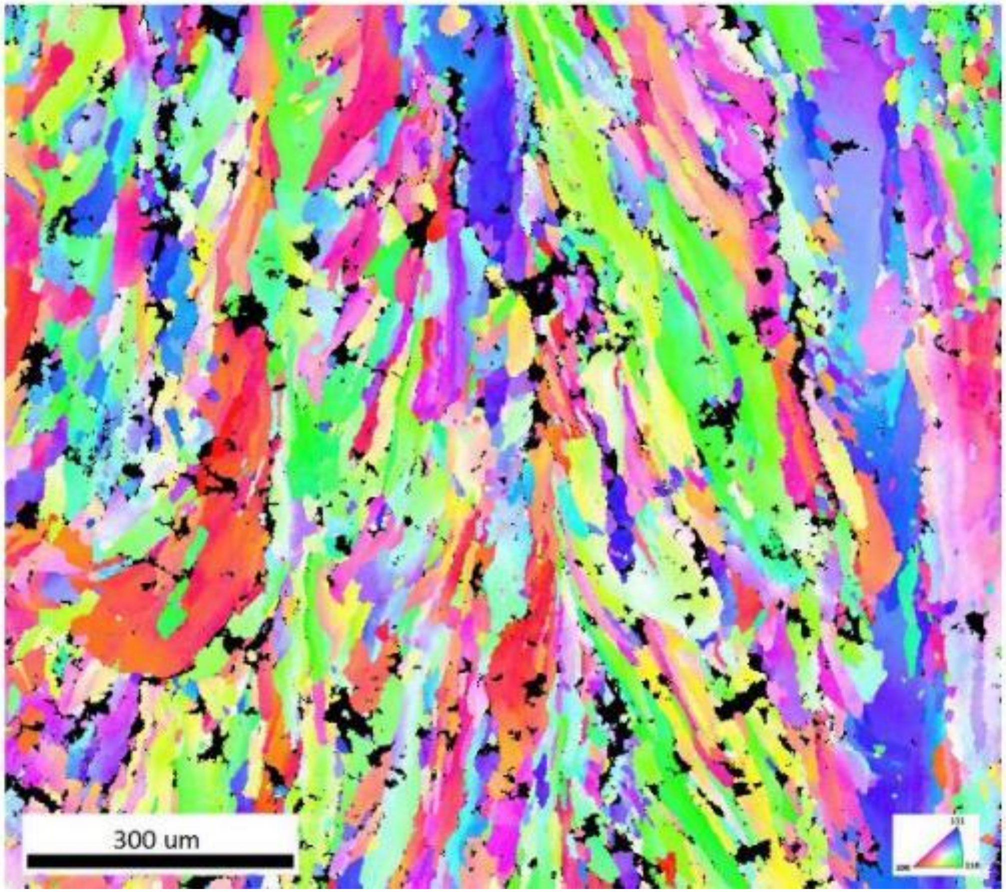
Recent Progress

- In-situ experiments performed at the Advanced Photon Source (APS) at Argonne National Laboratory (ANL) using the AM simulator in February 2020
- Microstructural characterization (namely grain and inoculant size measurements) of melt pools obtained from APS experiments
- Writing proposal, to be completed before Summer 2020
- NSF sponsored internship at Elementum 3D

Metrics

Description	% Complete	Status
1. Literature review	70%	●
2. Characterization of microstructure and measurement of modeling parameters	10%	●
3. Analysis of nucleant potency and extent of in-situ MMC reaction in AM conditions	5%	●
4. Evaluation of the effect of AM solidification conditions on grain refinement and use of the Interdependence Model	5%	●
5. Effect of inoculants and unreacted particles on post-processing heat treatment	5%	●

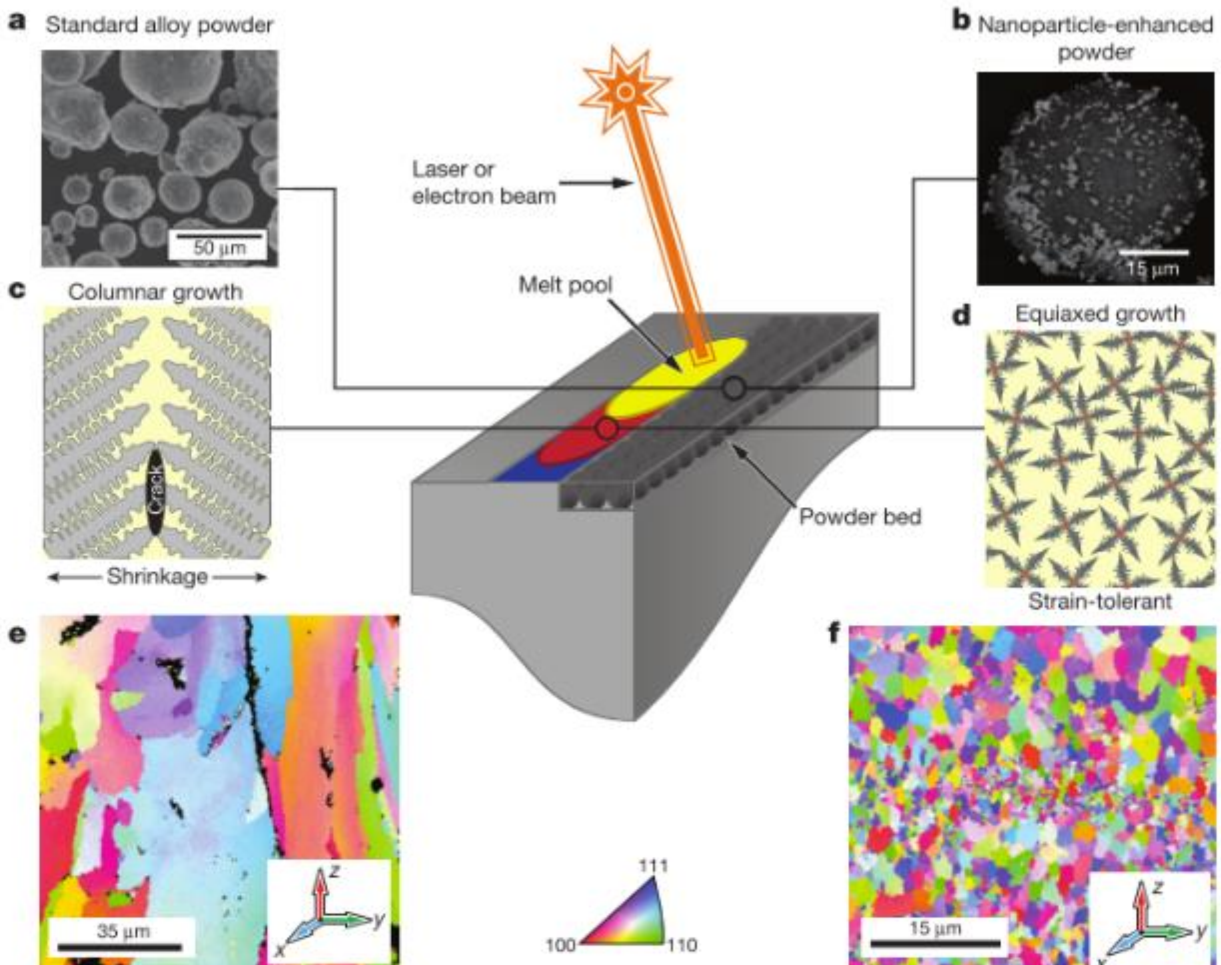
Industrial Relevance



Inverse pole figure of 3D-printed stock 7075, build direction is vertical to the page. Taken from J. H. Martin et al. *Nature,* 549 (2017) 365-369.

- Aluminum alloys currently used in AM are mostly traditional stock alloys (e.g. 7075, 6061, 2024)
- Under AM conditions these alloys tend to form columnar grains, and are subject to solidification cracking
- These results imply a need for alloys designed specifically for AM

Grain Size Control via Innoculants in AM Alloy Powders

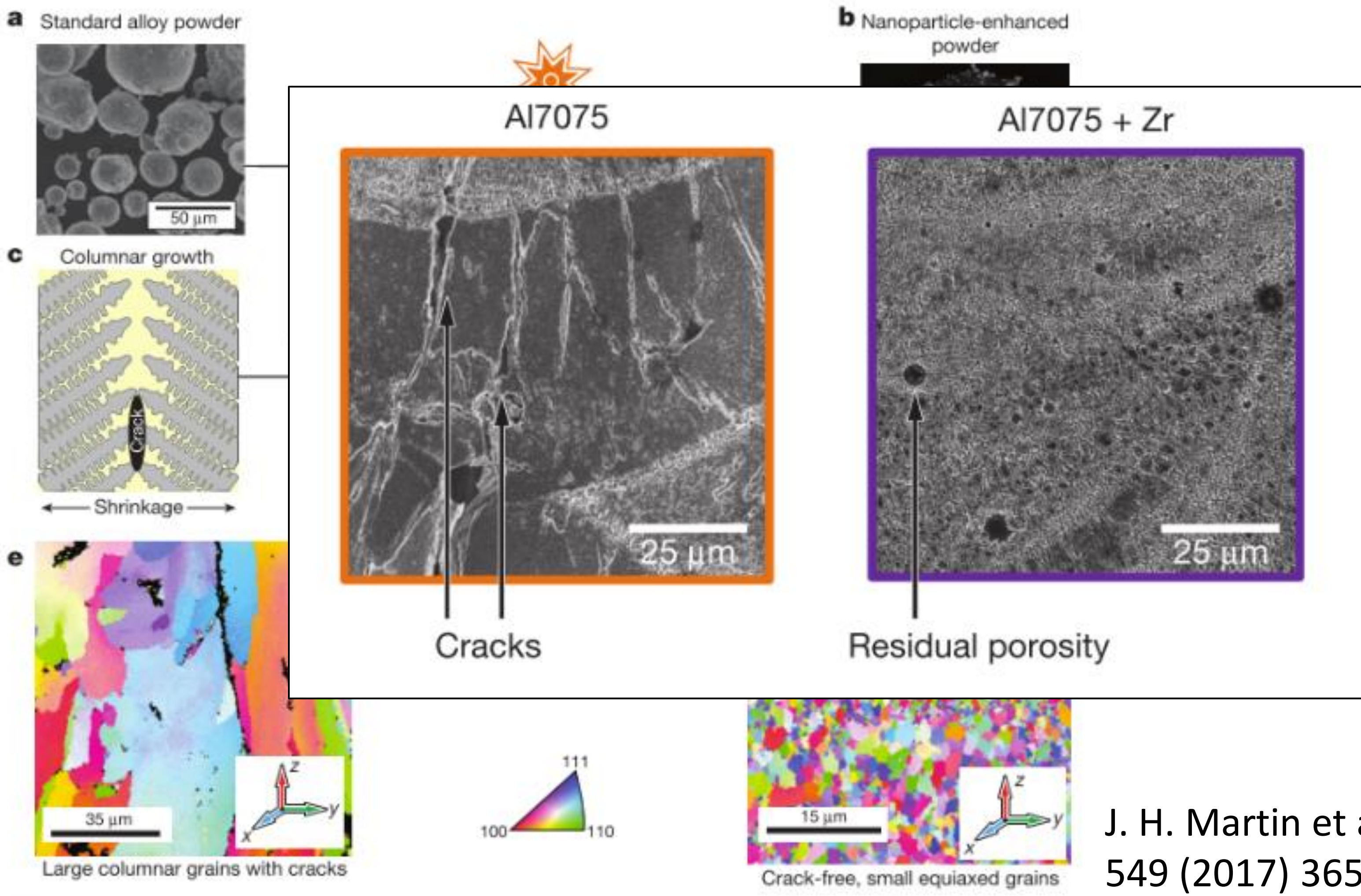


Large columnar grains with cracks

Crack-free, small equiaxed grains

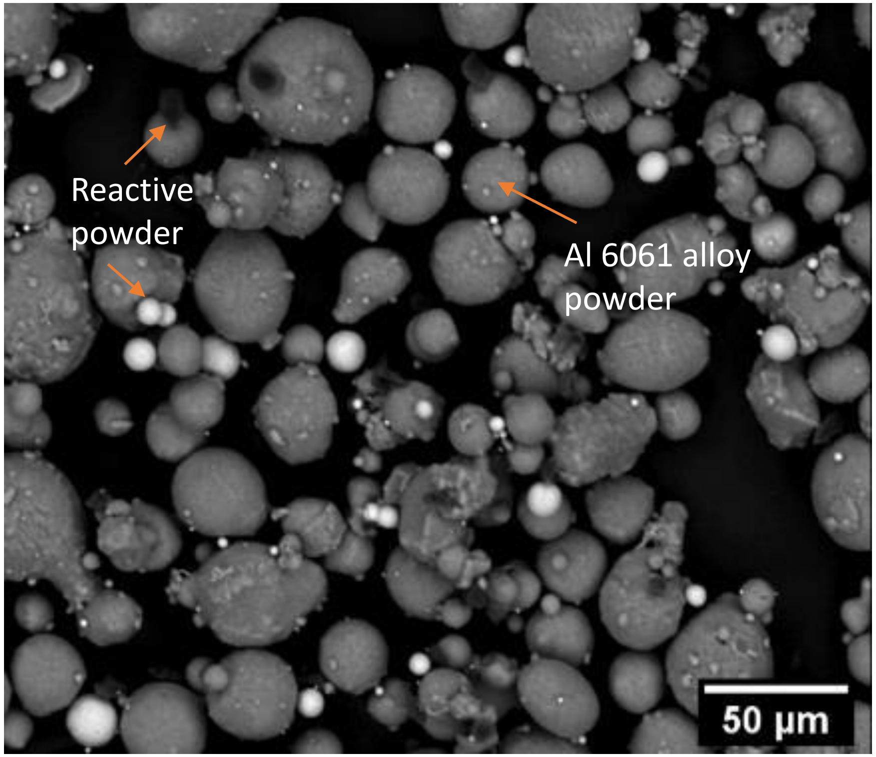
J. H. Martin et al. *Nature*, 549 (2017) 365-369.

Grain Size Control via Innoculants in AM Alloy Powders

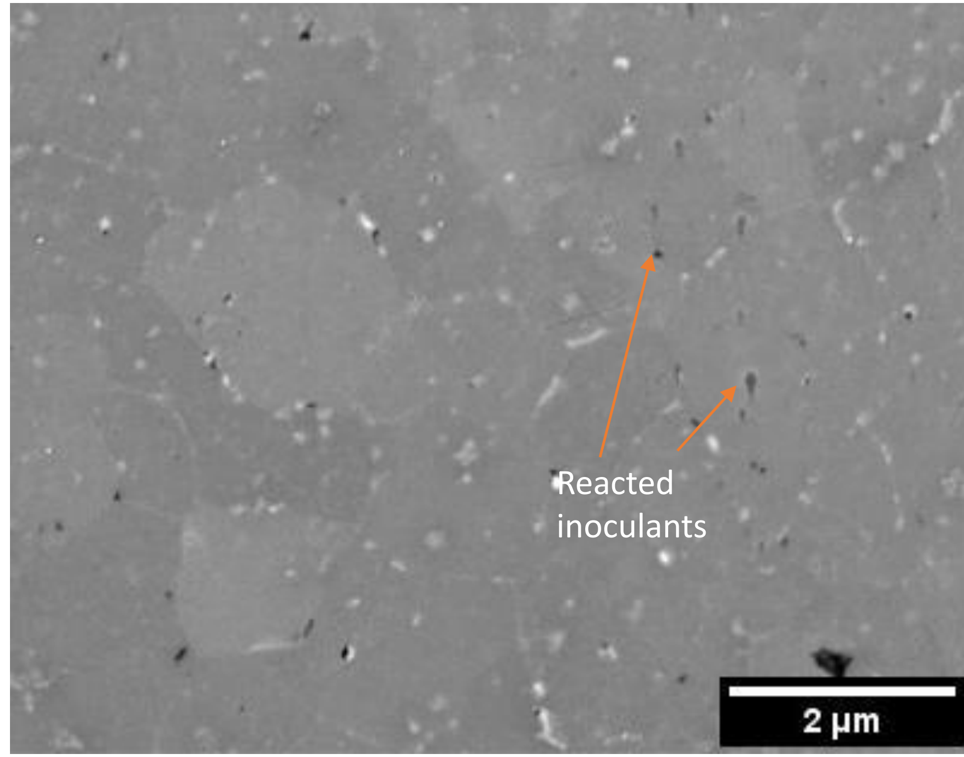


J. H. Martin et al. *Nature,* 549 (2017) 365-369.

Al 6061 Reactive Additive Manufacturing (RAM) Alloy Designed for AM: Initial Characterization



BSE SEM image of Al 6061 RAM 2% alloy powder



SEM image of as built Al 6061 RAM 2%

J. S. Neuchterlein & J. J. Iten, Reactive additive manufacturing, US Patent 20160271878 A1, priority 2015-03-17, published 2016-10-22.

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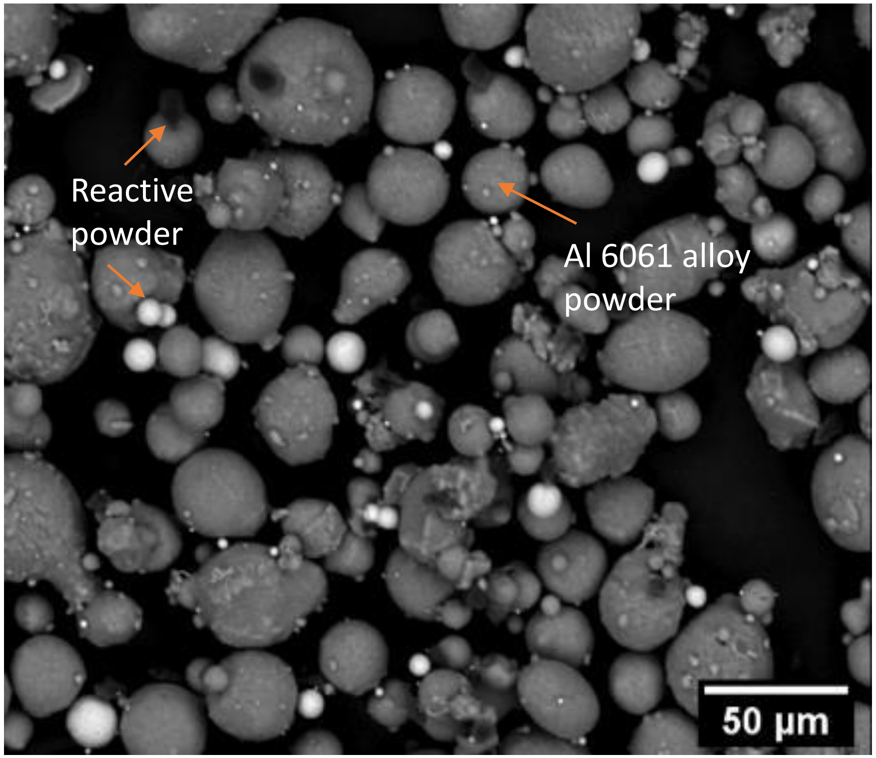

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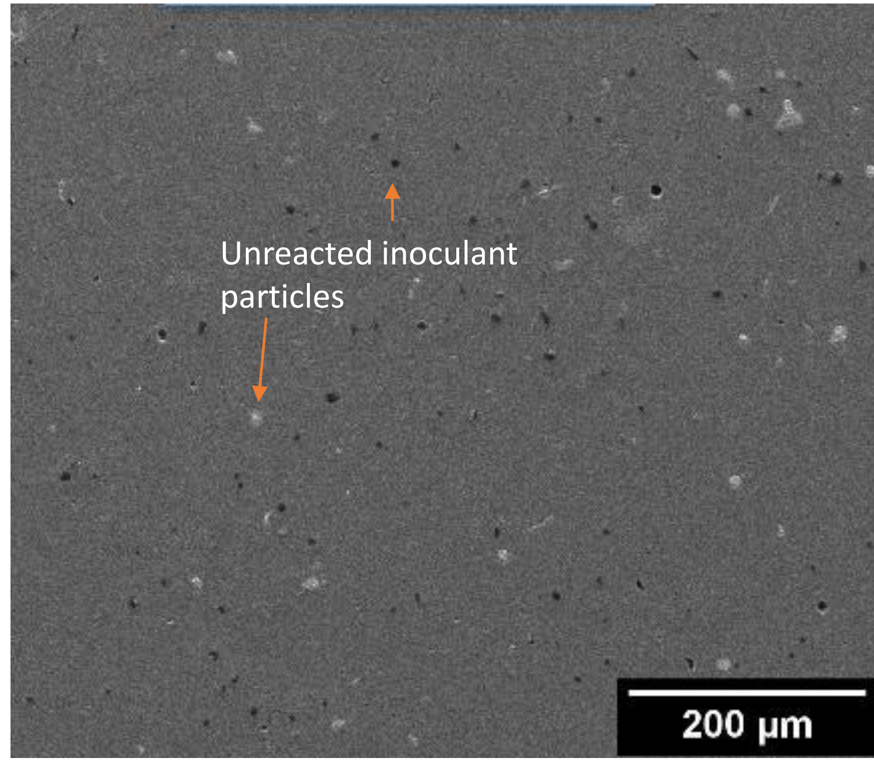

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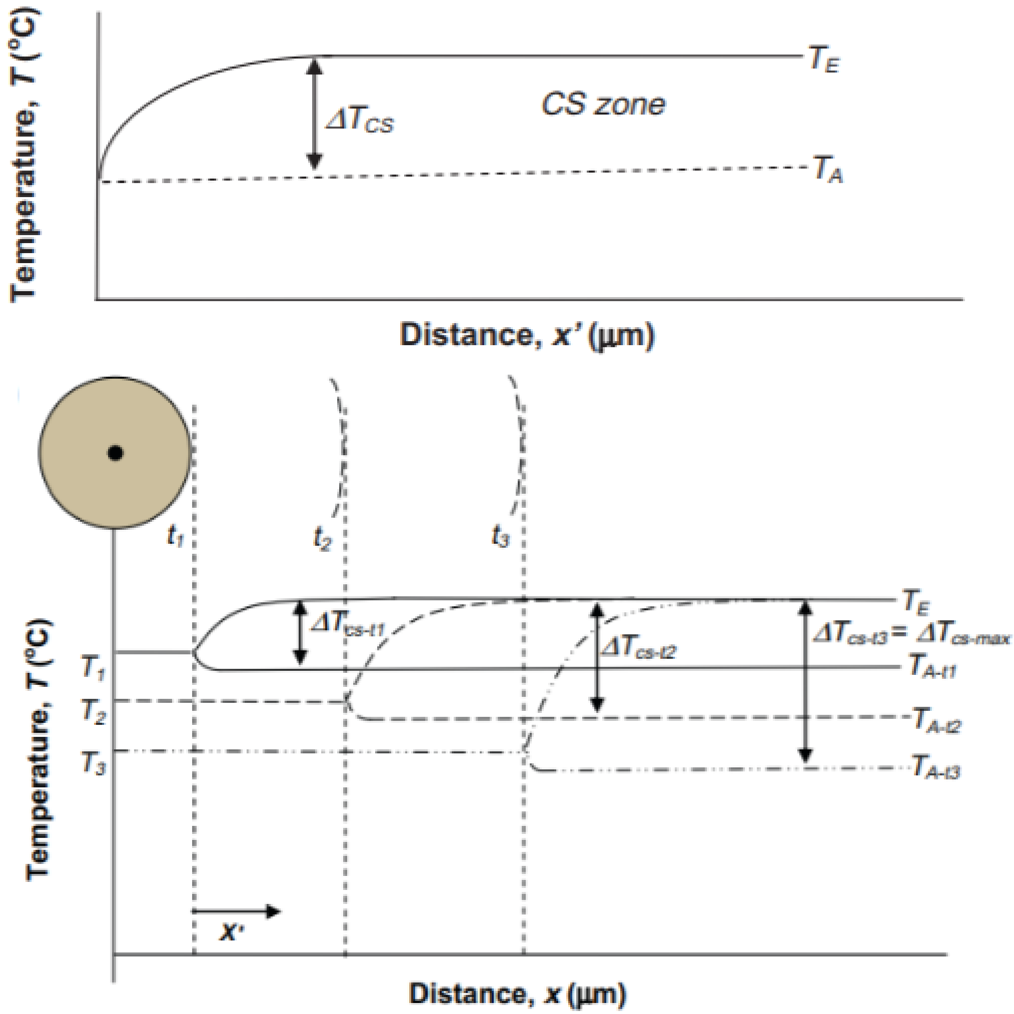
The Interdependence Model

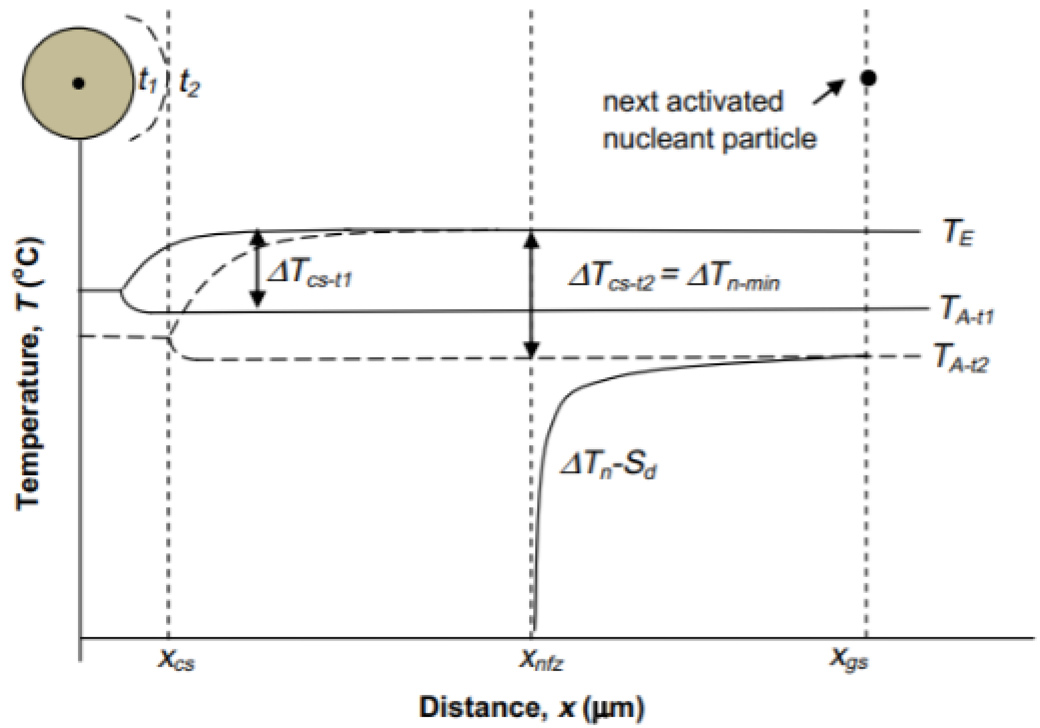
- Considers both nucleation and growth
- First nucleation events arise from thermal undercooling, then subsequent nucleation is driven by constitutional supercooling (CS) from nucleated grain



D. H. StJohn et al., Acta Mater., 59 (2011) 4907-4921

The Interdependence Model

- Once CS developed, need to consider how close the most potent nucleant particle is to this zone
- Find distance based on particle density and size distribution



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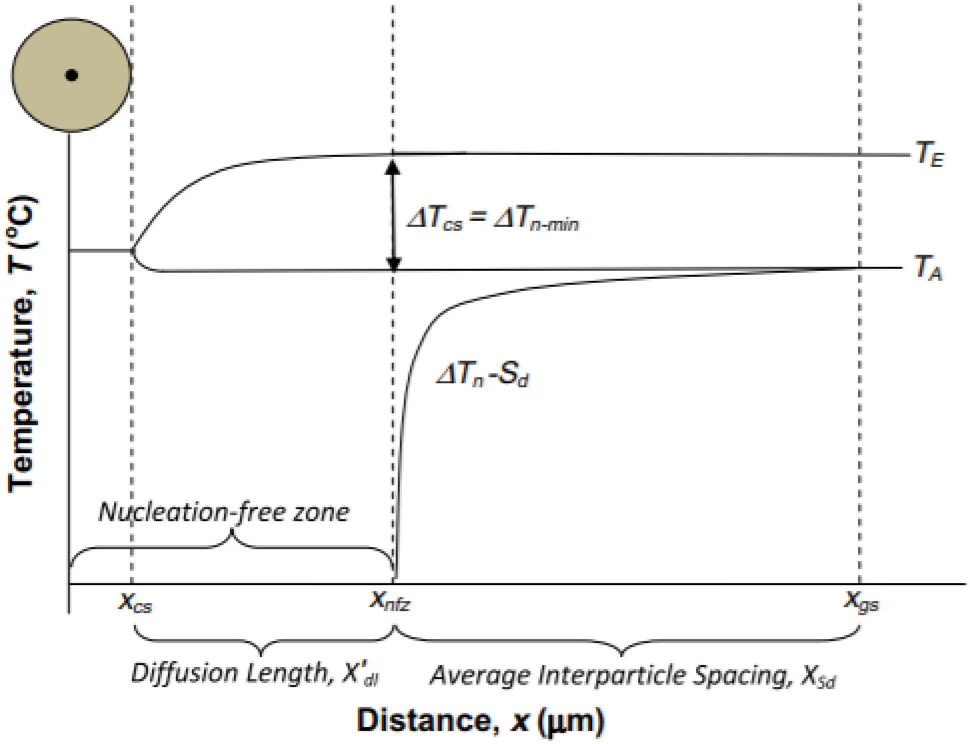
The Interdependence Model

$$d = x_{CS} + x'_{dl} + x_{Sd}$$

Grain size based off 3 distances:

- x_{CS}- distance previous grain must grow to generate sufficient undercooling for nucleation on next most potent particle
- x'_{dl}- length where undercooling from previous grain reaches a maximum
- x_{Sd}-probability of particle of highest potency being present in melt enveloped by undercooled zone



D. H. StJohn et al., Acta Mater., 59 (2011) 4907-4921

The Interdependence Model

$$d = \frac{Dz\Delta T_n}{vQ} + \frac{4.6D}{v}\left(\frac{C_l^* - C_0}{C_l^*(1-k)}\right) + x_{Sd}$$

$$\boxed{x_{CS}} \qquad \boxed{x'_{dl}}$$

$z\Delta T_n$ – incremental undercooling to cause the next most potent nucleant particle to nucleate, z is fraction of ΔT_n needed to nucleate next particle

v – solidification velocity

D – diffusion rate in liquid

C_l^* - composition of the liquid at the S-L interface

Q-growth restriction factor

The Interdependence Model

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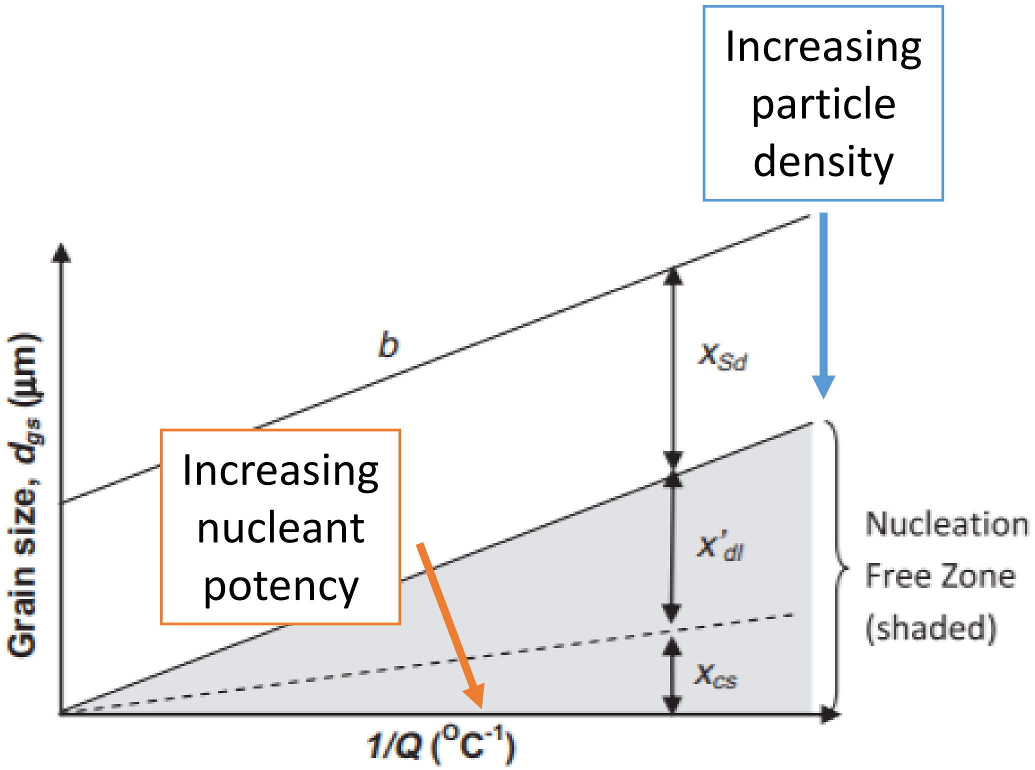

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The Interdependence Model & AM Conditions

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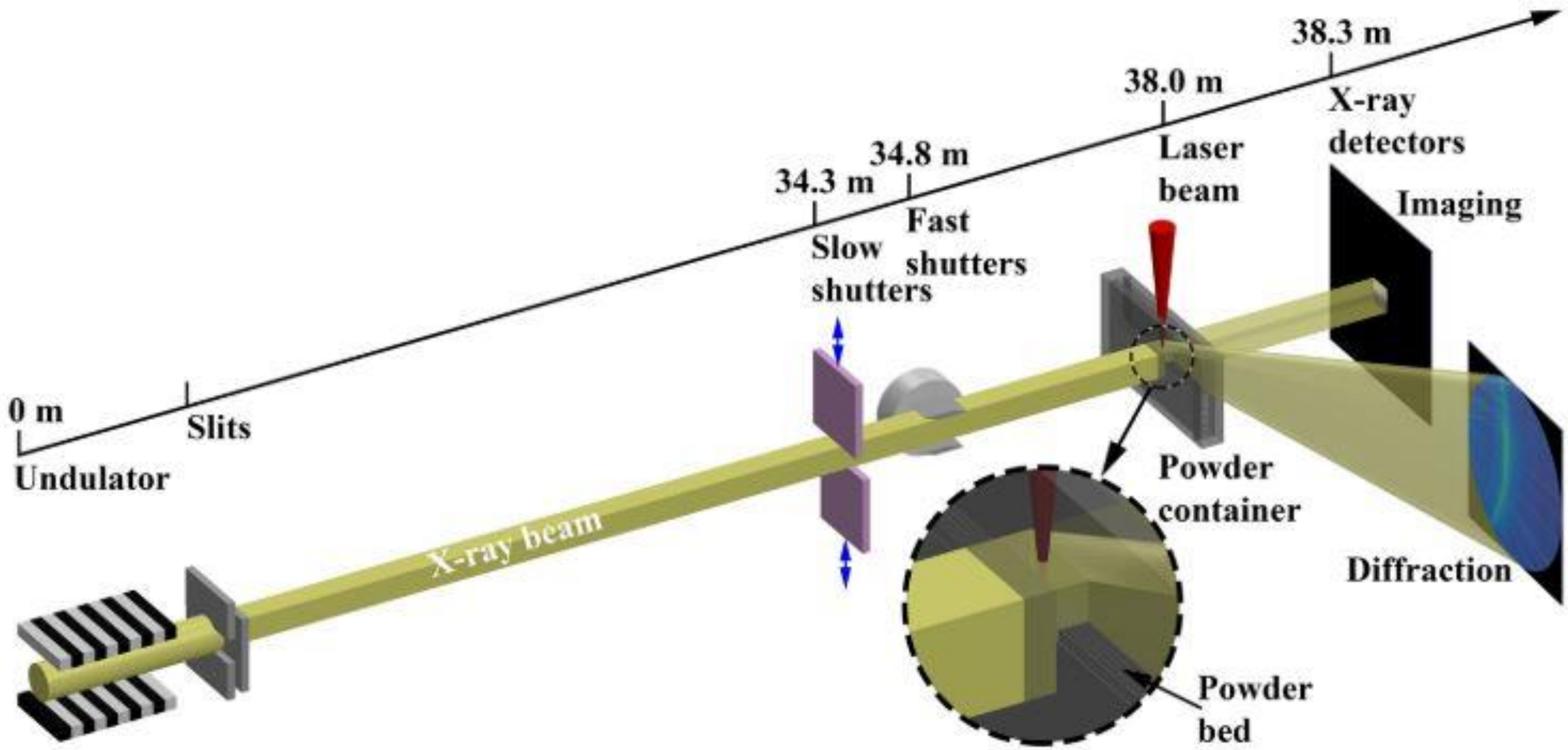
$$d = \frac{Dz\Delta T_n}{vQ} + \frac{4.6D}{v}\left(\frac{C_l^* - C_0}{C_l^*(1-k)}\right) + x_{Sd}$$

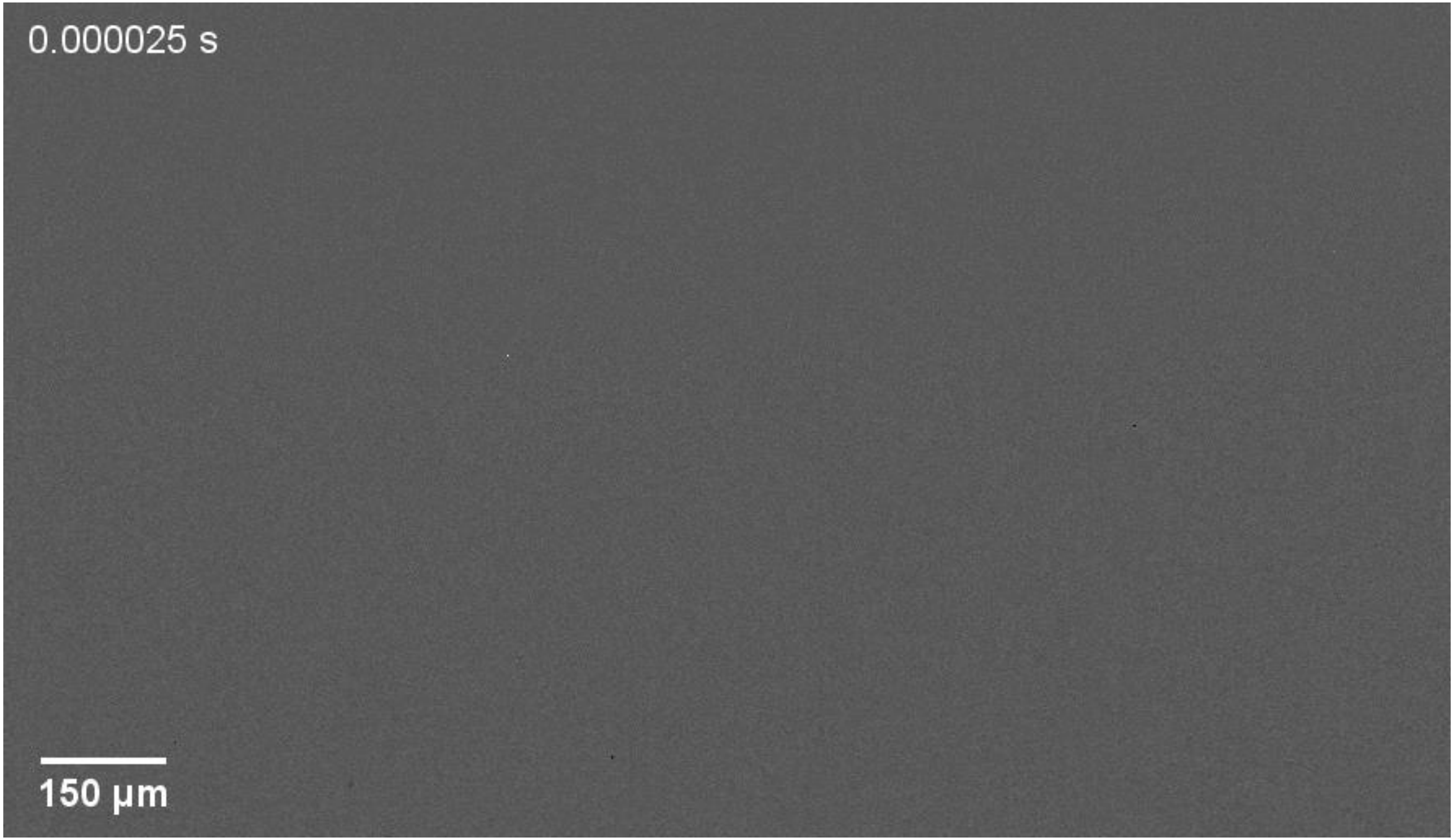
- In AM have large thermal gradients and high solidification velocity
- The larger thermal gradients are no longer negligible compared to the gradient caused by constitutional undercooling, changes how undercooling is considered ($z\Delta T_n$)
- Increasing velocity (v) decreases the distance from the S-L interface where the max constitutional undercooling is achieved

Advanced Photon Source (APS) Additive Manufacturing Simulator Set-up



Schematic of AM simulator used for in-situ experiments at ANL.
Taken from: C. Zhao et al., *Scientific Reports,* 7 (2017) 1-11.

Image Processing: Tracking of S/L Interface



Animation of laser pass on 6061 wrought + 6061 powder, 416 W, 0.5 m/s

Acknowledgement to Gus Becker for image processing

Selected Solidification & Alloy Conditions

Base Plate	Powder
Pure Al	Al RAM 5% Inoculant
Pure Al	Al RAM 10% Inoculant
Pure Al	6061 RAM 2% Inoculant
Pure Al	6061 RAM 10% Inoculant
Wrought 6061	6061 RAM 2% Inoculant
Wrought 6061	6061 RAM 10% Inoculant
6061 RAM 2% Inoculant	6061 RAM 2% Inoculant
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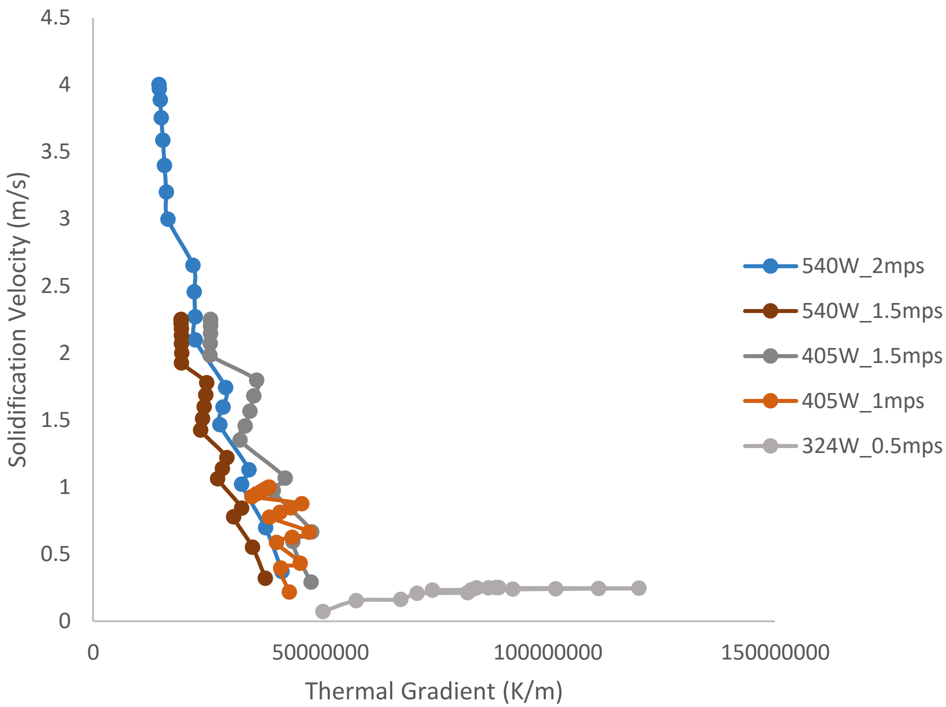

Acknowledgement to Brian Rodgers for Rosenthal modeling.

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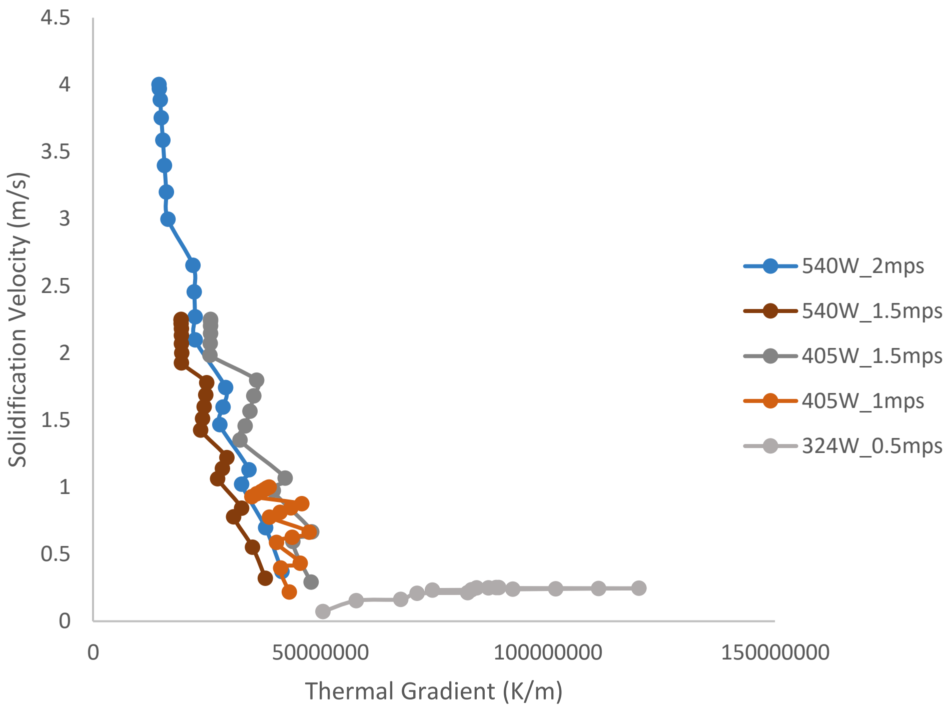

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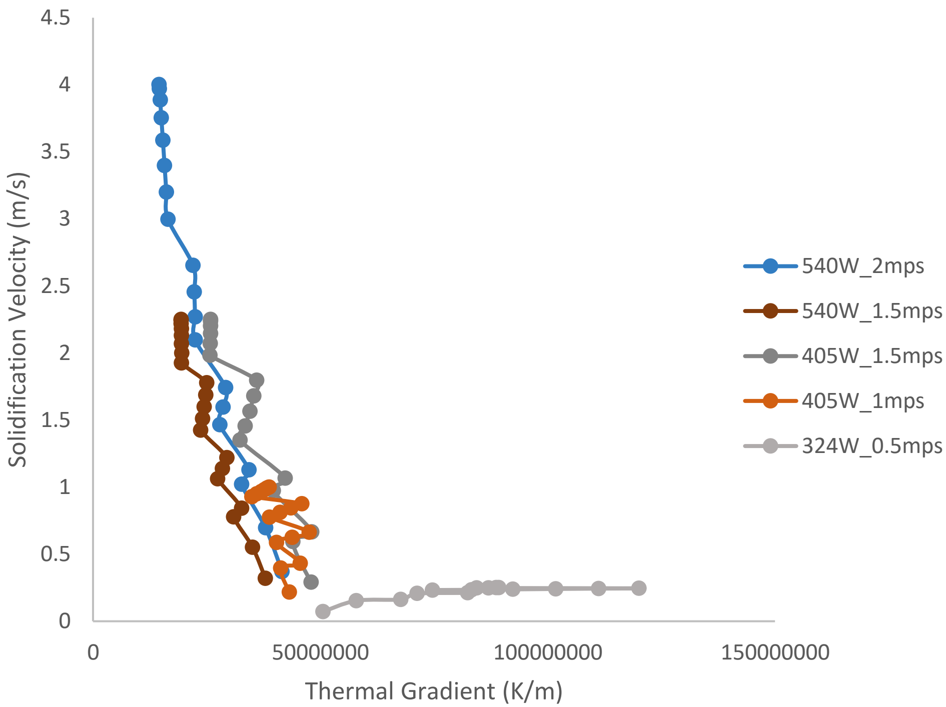

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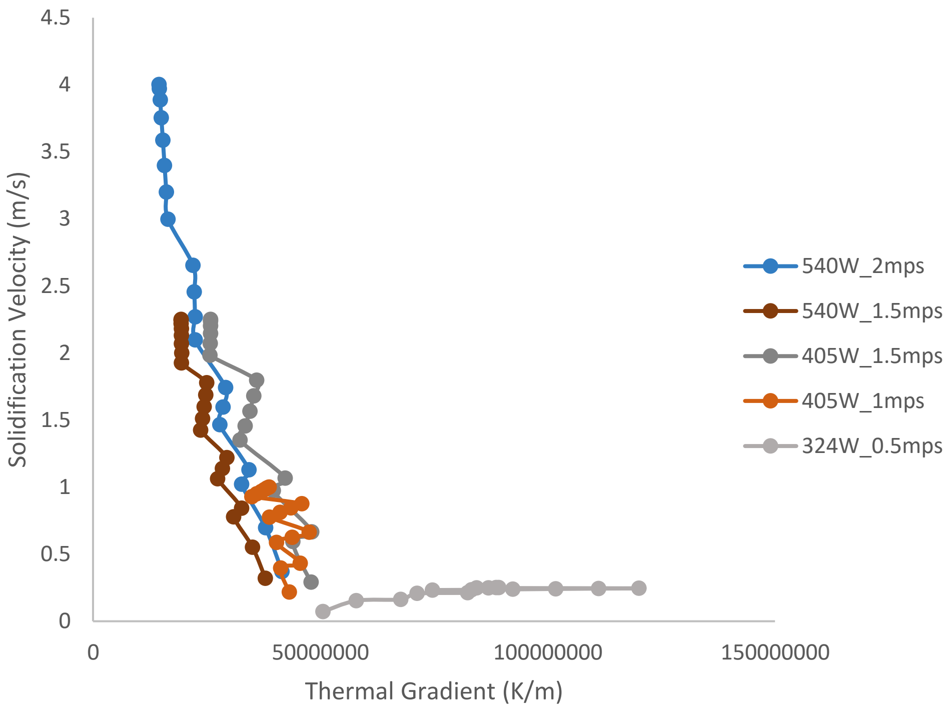

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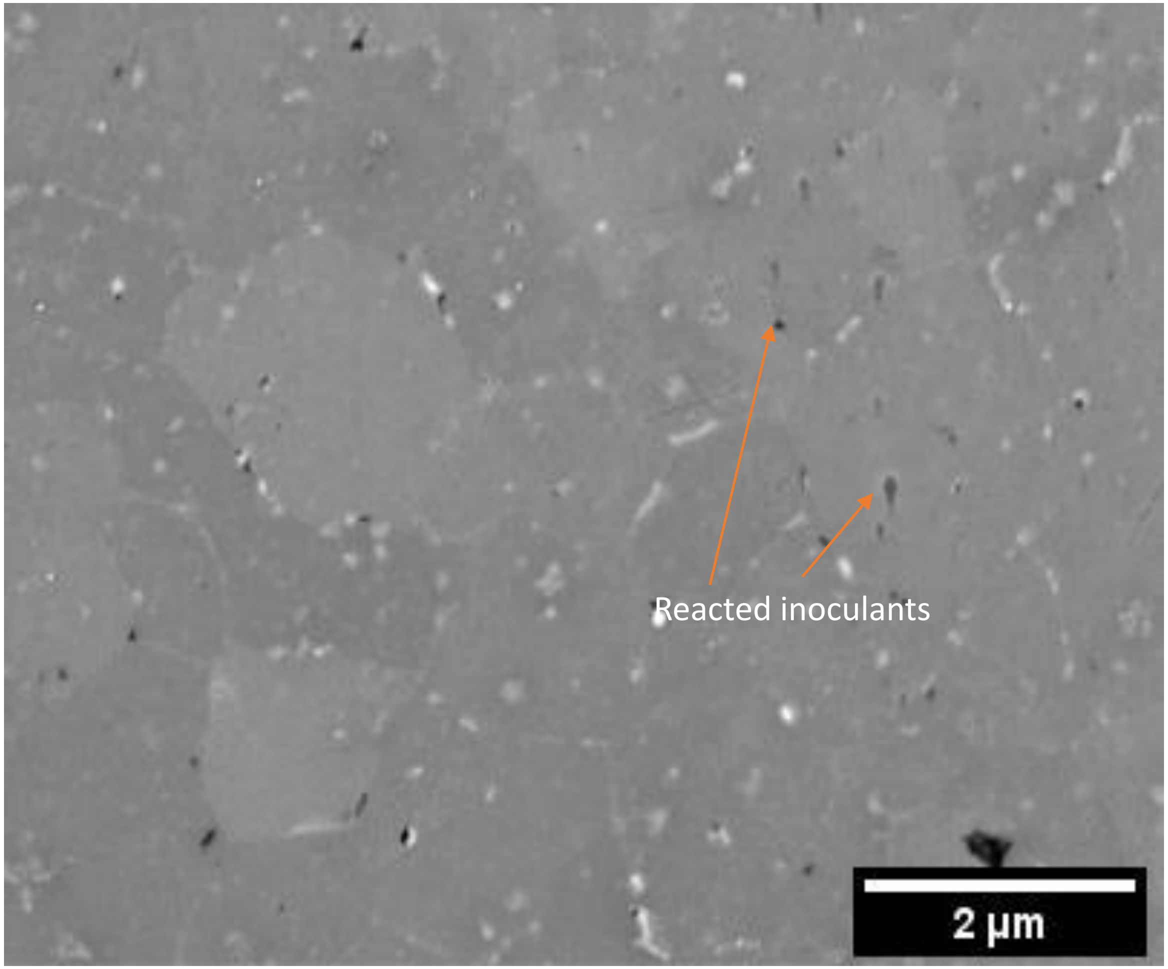
Postmortem Measurements: Inoculant Diameter

$$\Delta T_n = \frac{4\sigma}{\Delta S_v d}$$

σ – S-L interfacial energy

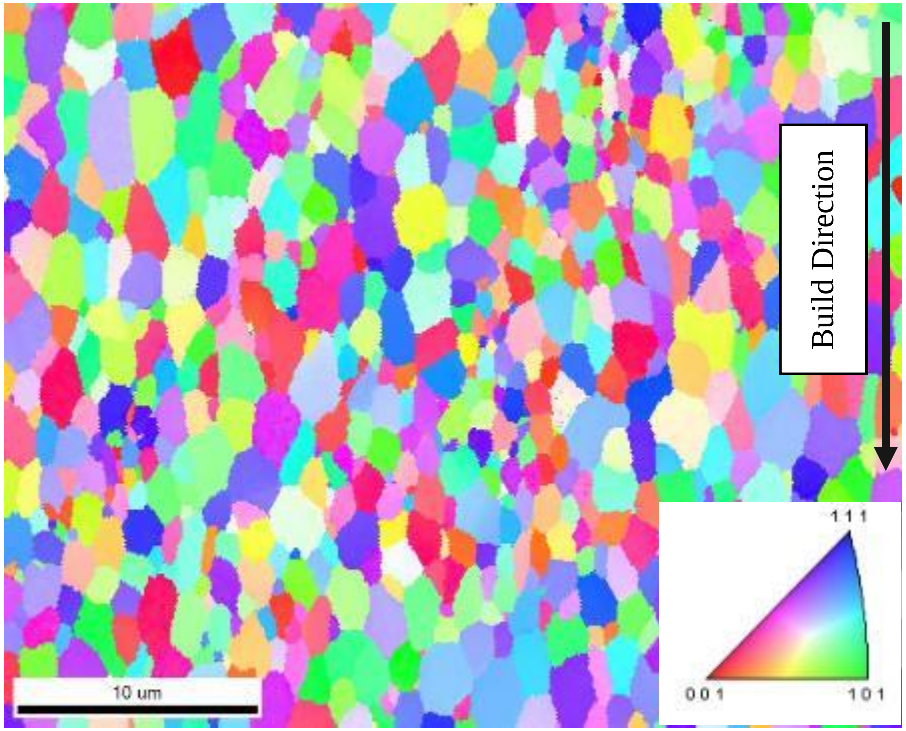
ΔS_v - entropy of fusion

d – nucleant particle diameter

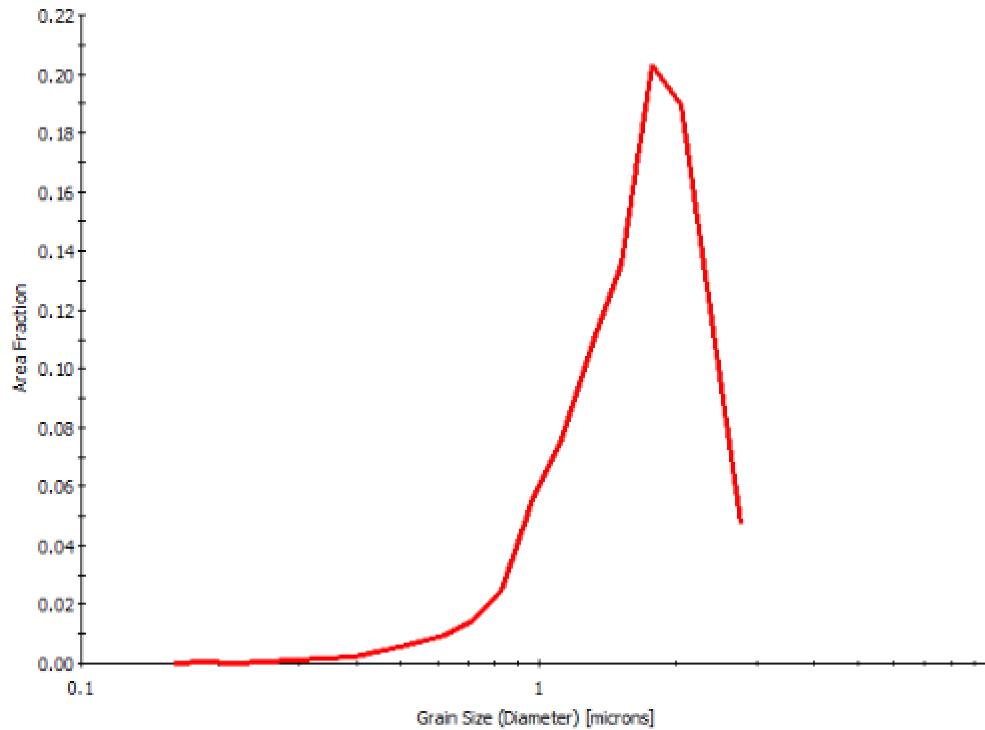


SEM image of Al 6061 RAM 2% build

Postmortem Measurements: Grain Size

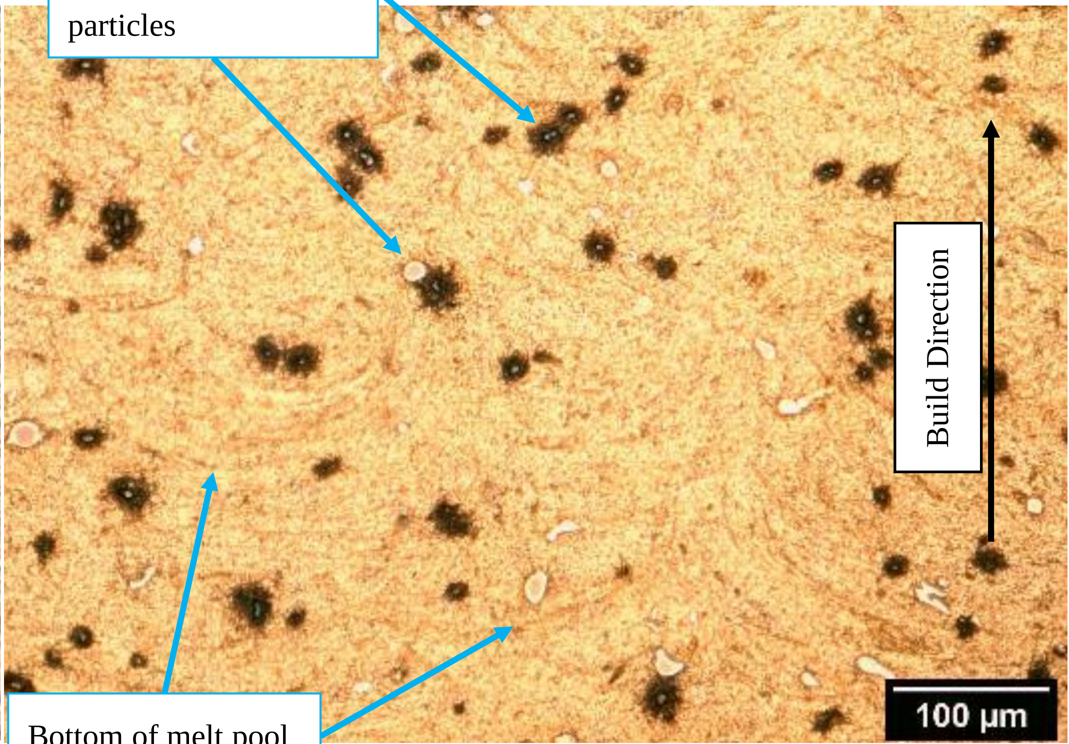


EBSD inverse pole figure map (IPF) taken from an Al 6061 RAM 2% rectangular build



Grain size (diameter) measurement taken from EBSD IPF (left) of Al RAM 6061 2%

Postmortem Measurements: Microstructural Trends in Melt Pool & % Reacted



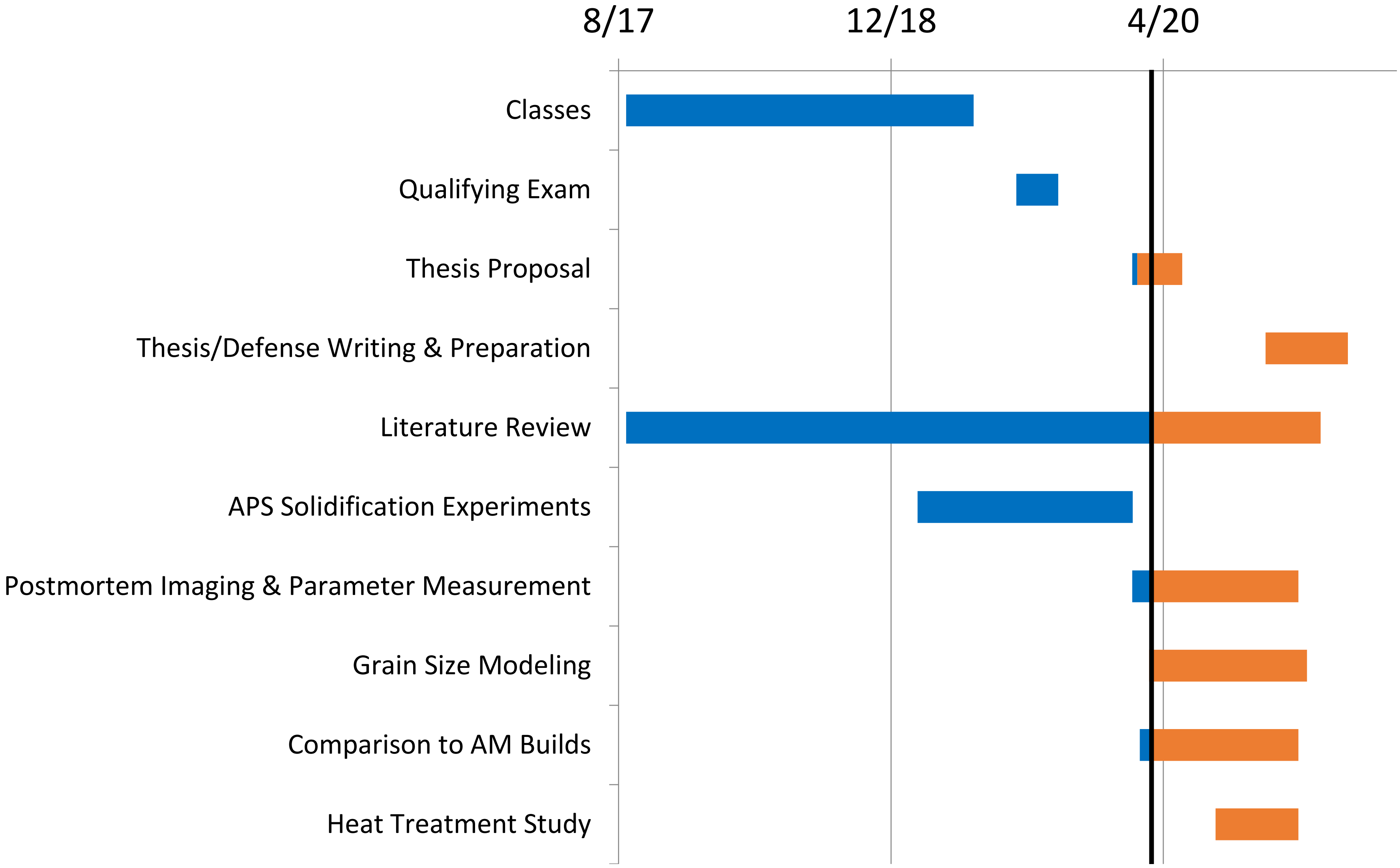
Light optical images of Al 6061 RAM 2% build etched with:

1) Phosphoric acid and NaCl at 70°C for 1 s
2) NaOH and $KMnO_4$ (Weck's reagent) for 5 min

Future Work

- Measure parameters from in-situ radiography and post-mortem characterization (i.e. solidification velocity, grain size, nucleant size, and fraction of unreacted particles)
- Comparison of interdependence model to experimental results and evaluate the utility for AM solidification conditions
- Compare microstructural characteristics from in-situ experiments to AM builds
- Perform heat treatment study of AM builds

Progress


8/17
12/18
4/20
Classes
Qualifying Exam
Thesis Proposal
Thesis/Defense Writing & Preparation
Literature Review
APS Solidification Experiments
Postmortem Imaging & Parameter Measurement
Grain Size Modeling
Comparison to AM Builds
Heat Treatment Study

Challenges & Opportunities

- Challenges
  - Getting representative data from imaging, measurements, and enough of a variety of solidification conditions
  - Modeling for AM is challenging, could run into some issues
- Opportunities
  - Validate existing models with respect to AM
  - Inform heat treatment design changes needed for inoculated alloys or MMCs for post-processing

Thank you!

Chloe Johnson

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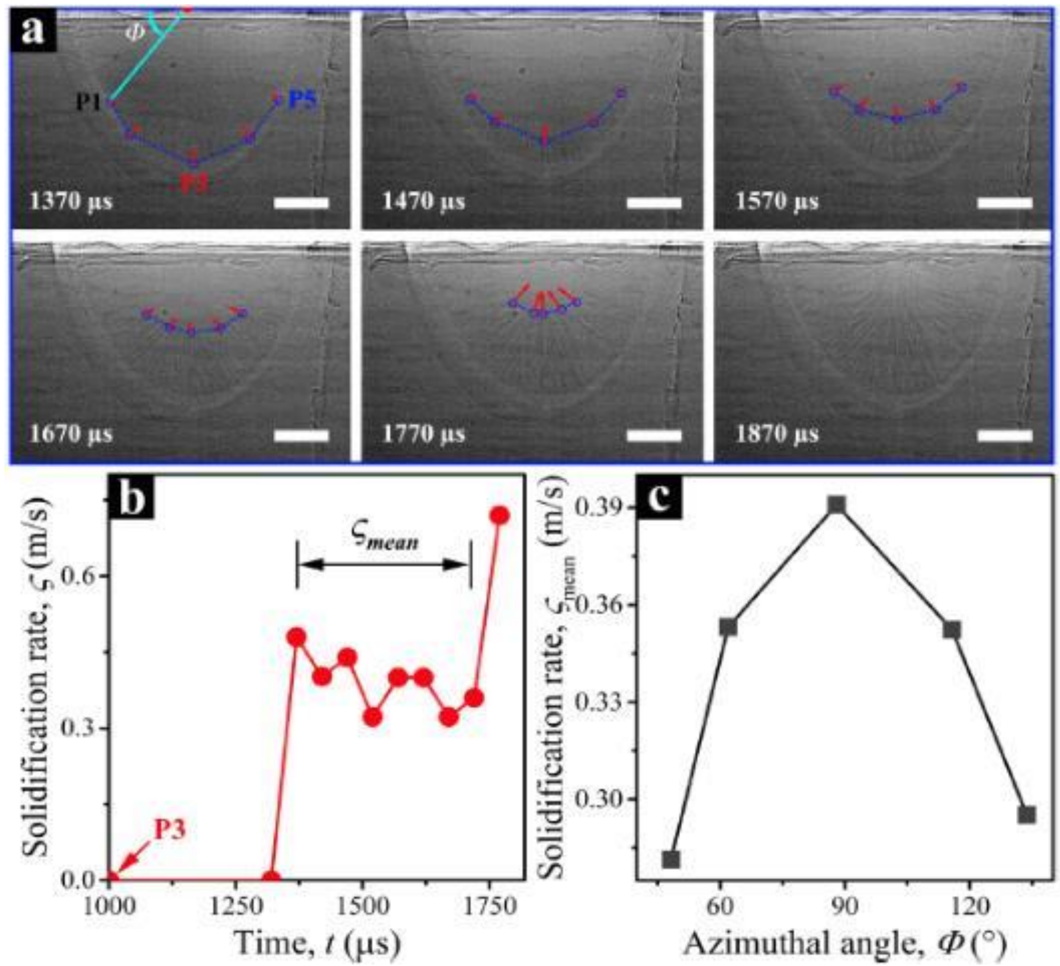
Heat Treatment Study Parameters

Sequence Number	Solution Temperature (°C)	Solution Time (hr)	Aging Temperature (°C)	Aging Time (hr)					
				#1	#2	#3	#4	#5	#6
1	530	1.5	150	3	8	12	18	24	48
2	530	1.5	165	3	8	12	18	24	48
3	530	1.5	175	1	3	5	8	12	18
4	530	1.5	190	1	3	5	8	12	18
5	500	1.5	165	3	8	12	18	24	48

- Each heat treatment will be performed on Al 6061 RAM 2% as well as traditional 6061 for comparison
- These will be evaluated with microhardness maps and microstructural analysis on a selected few
- Will also add compare samples of Al 6061 RAM 2% build samples printed with no preheat, 80°C, and 200°C build plate temperatures at one set of heat treatment parameters

APS Additive Manufacturing Simulator Set-up



Synchrotron x-ray imaging of a Ti-6Al-4V plate sample in laser melting processes and solidification rate measurements. C. Zhao et al., *Scientific Reports* (2017).

Exploring Grain Refinement in Inoculated Alloys for AM

- To build on and adjust the Interdependence Model for AM conditions, use in-situ synchrotron and ex-situ postmortem imaging to measure parameters more accurately
- Nucleant potency and the effect of limited kinetics in rapid solidification will be evaluated by comparing initial nucleation site density to the amount of unreacted particle measured after solidification
- Solute and nucleation site density effects will be considered by varying the alloy content (i.e. comparing pure Al and 6061) and the volume % of RAM, respectively
- Modeling of single raster samples from synchrotron experiments will then be compared to bulk scale builds
- Heat treatments will be performed on build samples to evaluate the effect of inoculants on post processing performance

Grain Refinement in Traditional Casting

$$d = a + \frac{b}{Q}$$

M. Easton et. al, Met. and Mater. Trans. A, 36A (2005) 1911-1920.

Grain Refinement in Traditional Casting

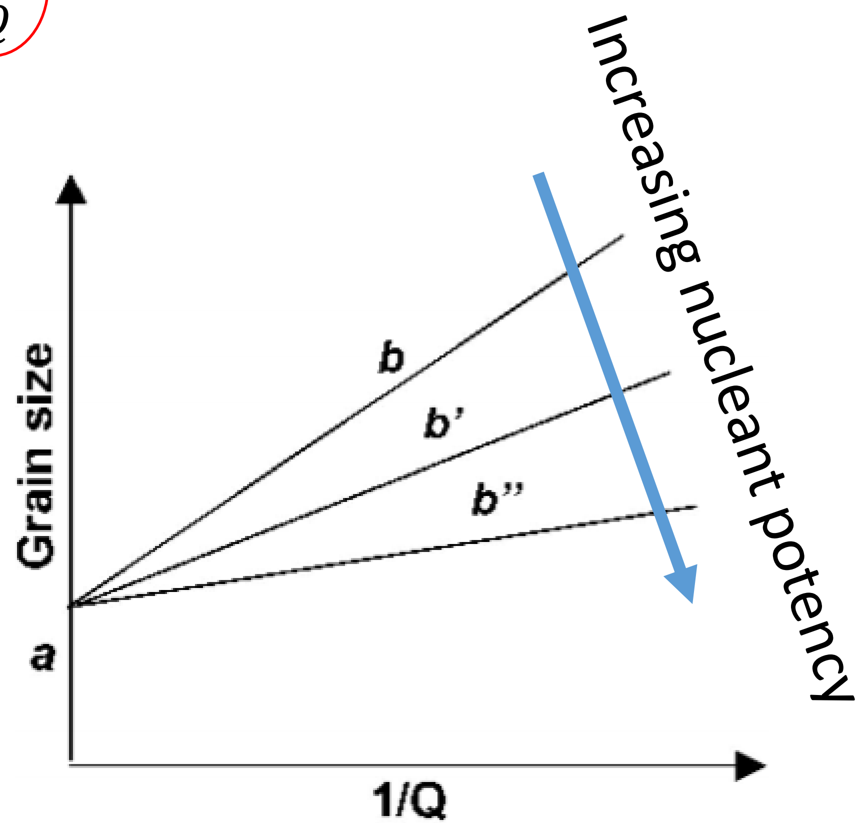
$$d = a + \frac{b}{Q}$$

- b-constant
- Q – growth restriction factor:

$$Q = m_l C_0 (k - 1)$$

m_l - liquidus slope
C_0 - alloy composition
k – partition coefficient

- Related to constitutional undercooling needed to nucleate grains
- This is inversely proportional to nucleant potency



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Grain Refinement in Traditional Casting

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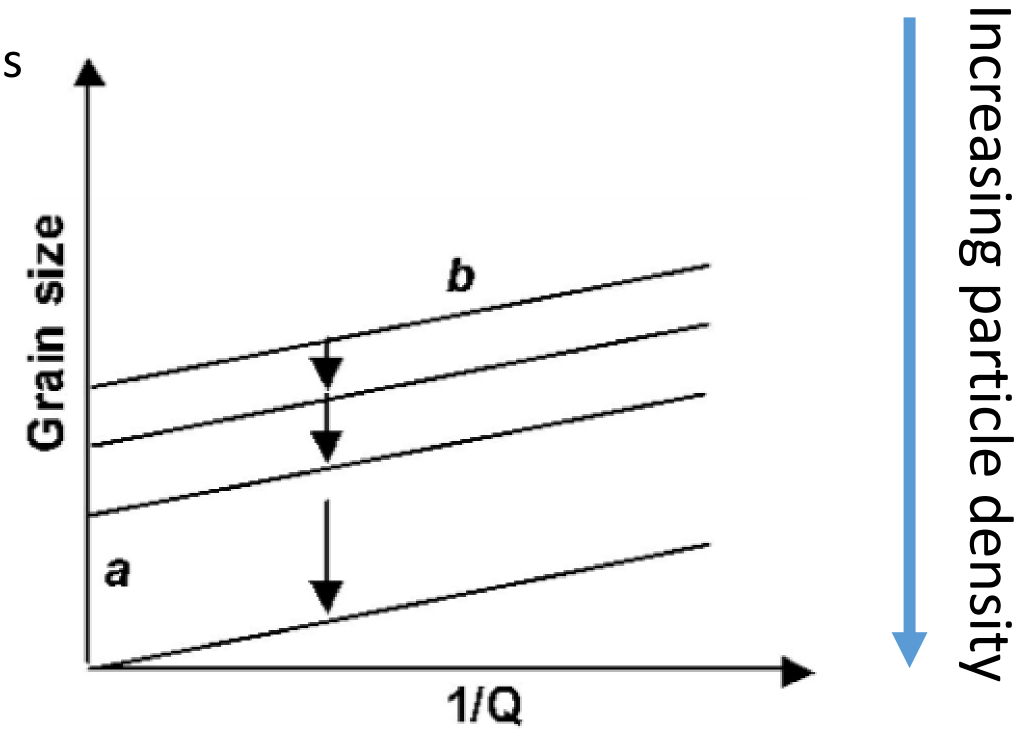
$$d = a + \frac{b}{Q}$$

- grain size intercept, characterizes minimum grain size that can be achieved in a system
- highly dependent on amount of solute, as lower solute, higher grain size limit
- Also dependent on particle concentration



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