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Title: Solidification Science at Sigma Overview

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Luitjohan, Kara Eileen

Intended for: On-site presentation with non-LANL University of California employees present

Issued: 2023-07-28

Solidification Science at Sigma Overview

July 28th, 2023

Seth D. Imhoff,
Kara E. Luitjohan

Los Alamos National Laboratory
Sigma-1, Sigma Division
Foundry and Solidification Science Team

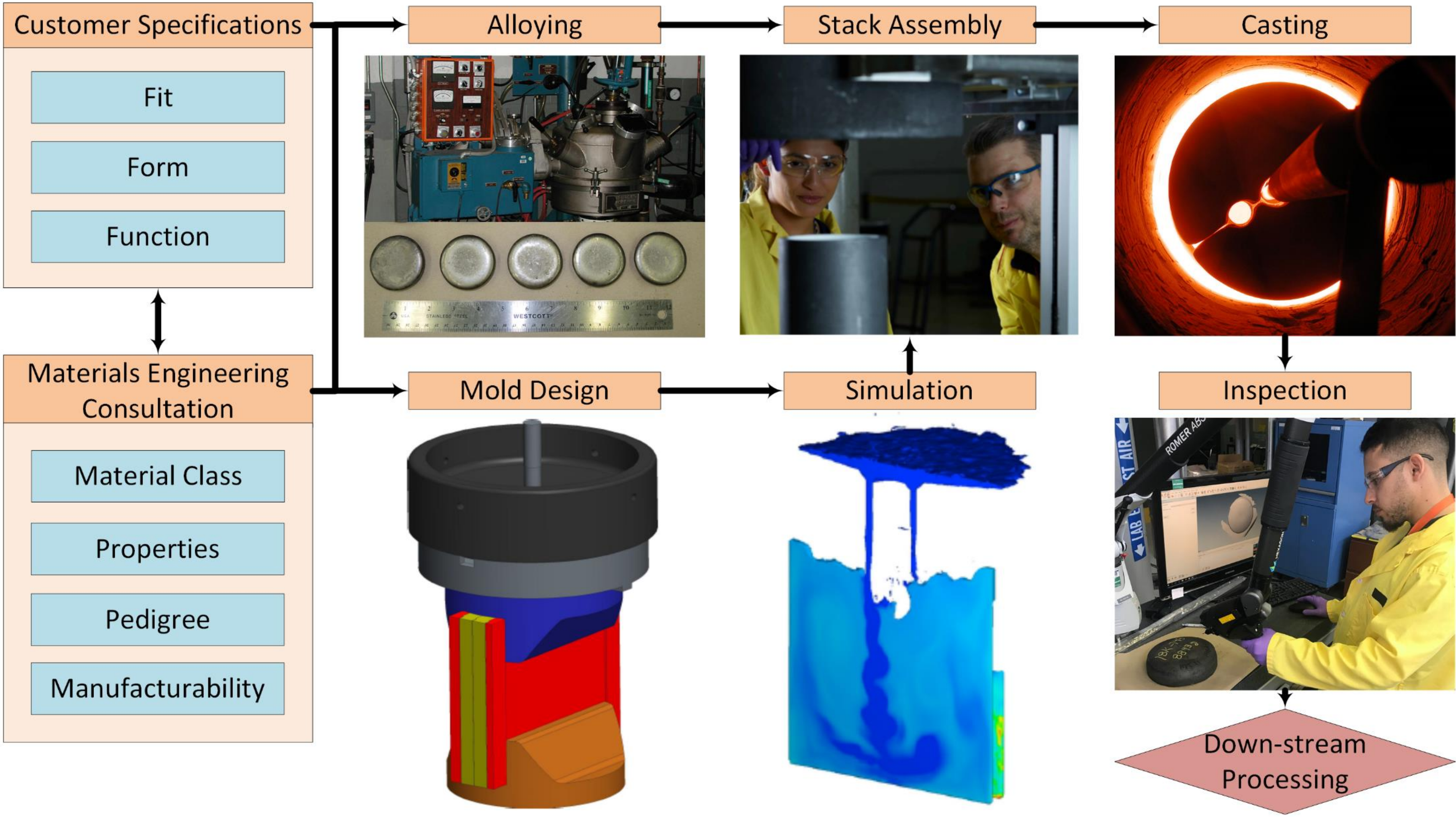
Summary of
LA-UR-20-26480
LA-UR-21-24795
LA-UR-22-28059
LA-UR-22-25533
LA-UR-23-22744
LA-UR-23-20097
LA-UR-23-21021

UNCLASSIFIED

Our laboratory and team

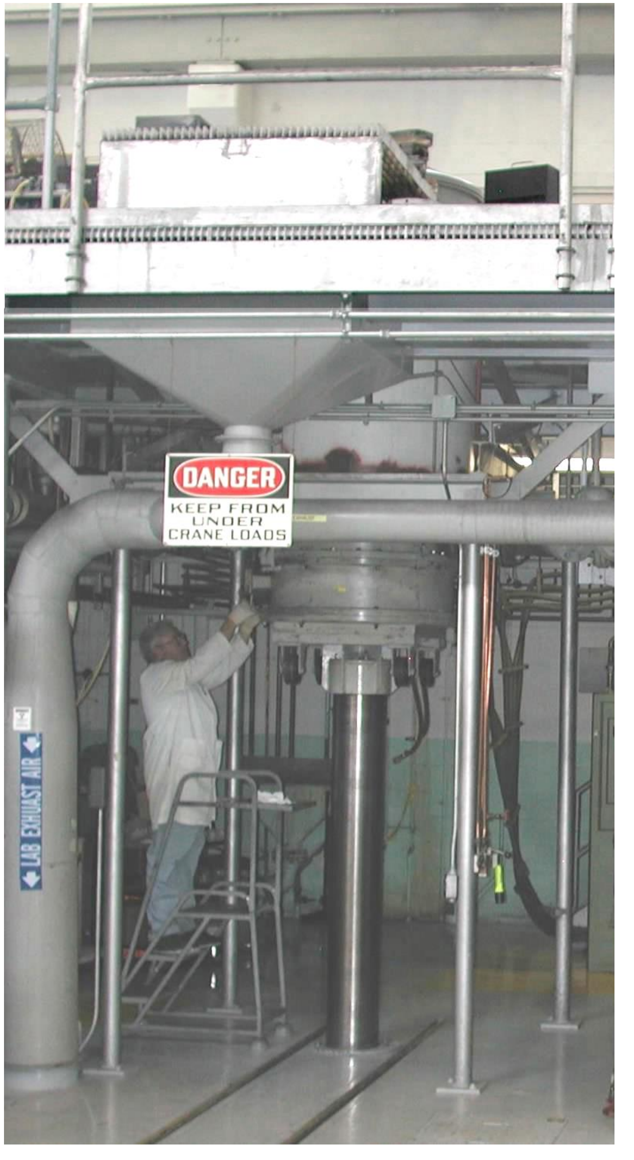
- Specialize in uranium^{238}
- Vacuum Induction Melting (VIM)
- Vacuum Arc Remelting (VAR)
- Non-consumable electrode arc melting (NCEAM)
- Plasma Arc Melting (PAM)
- Sizing operations
- Sampling and analysis operations
- …and all that goes with these processes!

End-to-end foundry service is the driver



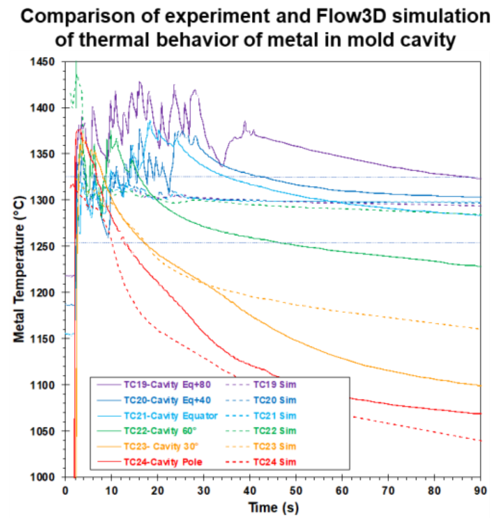
The scale of our manufacturing

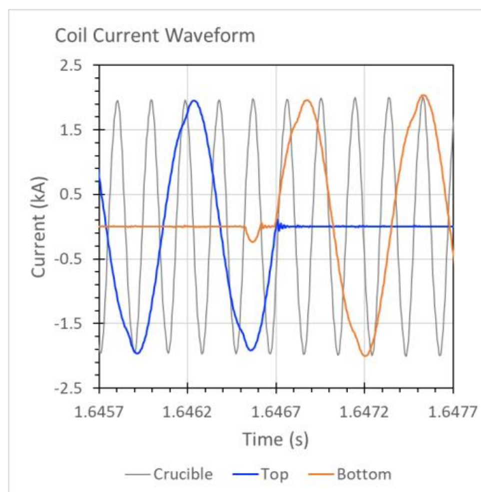
- Large research / small industrial
  - High equipment costs so upgrades are few and far between
  - Balance of natural effects is different than at the micron level
    - Think of any transport ratio that involves length scale
- Prototype manufacture
  - Deadlines
  - Low volume, high value
  - Few "mulligans" allowed
  - Control systems must cover an enormous process space so tend to be manual
  - Component manufacturing statistics are broad, but not deep

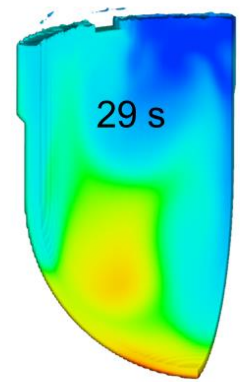


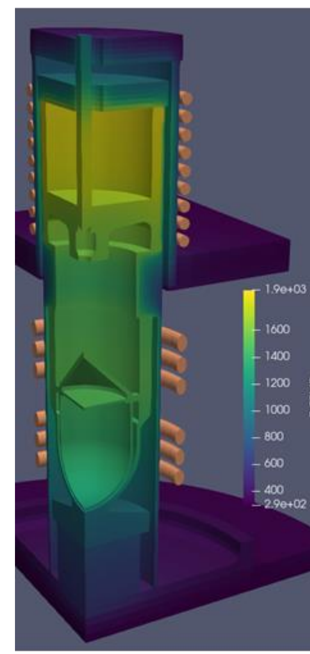
Data Capture Capabilities for V&V

Abundance of Instrumentation









250 mm dia. Hemispherical Heatup Solution (Truchas)

~50 Gb per trial for a full suite of information

We can’t cover it all today…

- Topics of interest (example)
  - Solidification Structures (U-Nb)
  - Phase relationships (U-C)
  - Solidification Pathways (Multinary)
  - Modes of solute partitioning (U-Th)
  - Control systems (VAR)

- A Deeper Dive – U-10Mo
  - A singular project
    (Alloy system behavior and property refinement)
  - How does a longer program evolve?
    (modeling focused retrospective)

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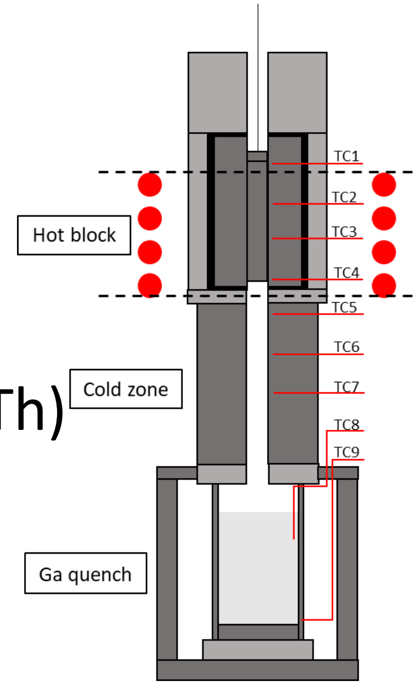

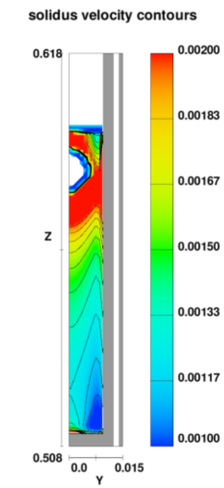

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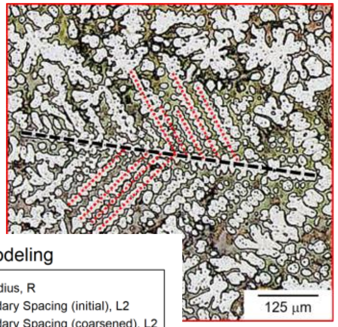

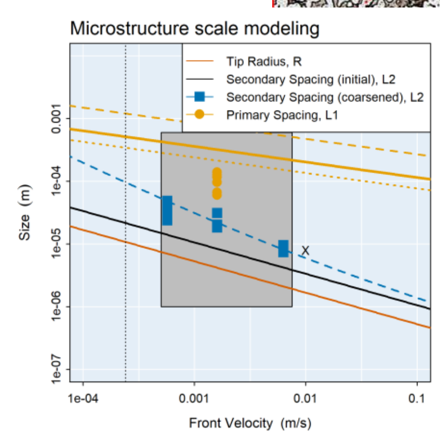

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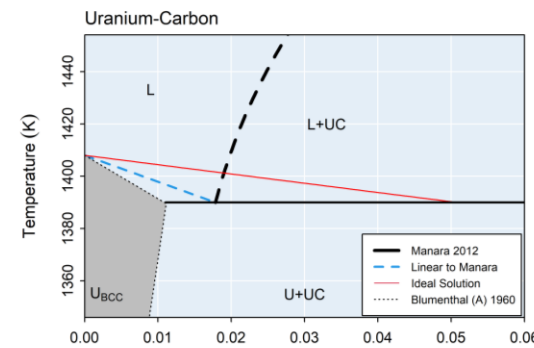

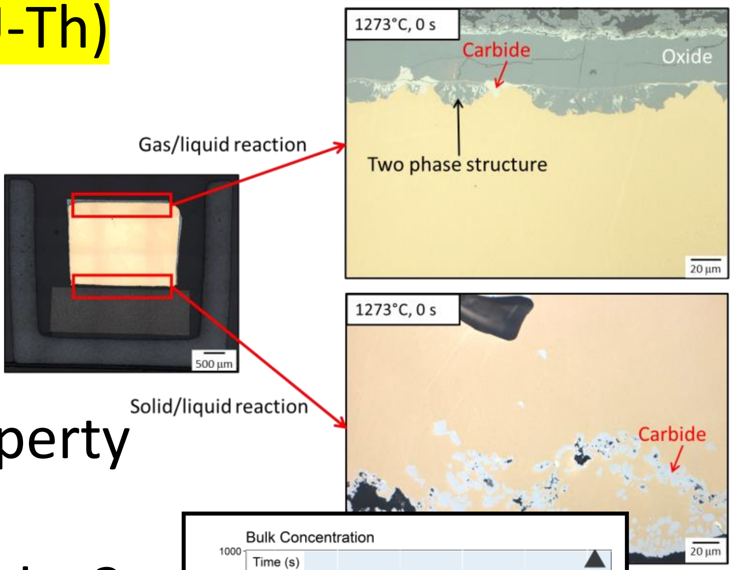

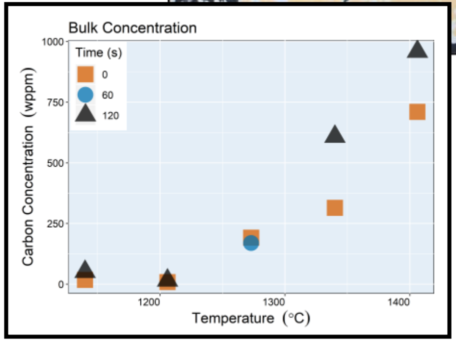

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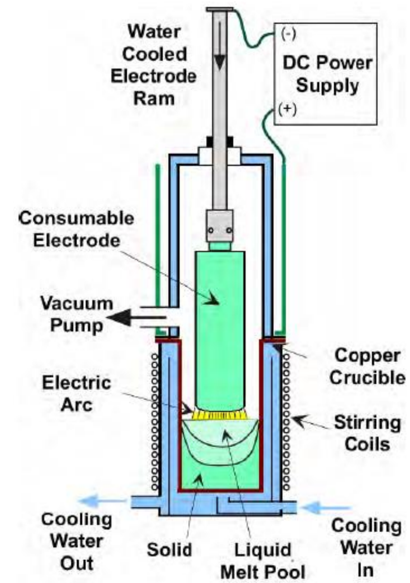

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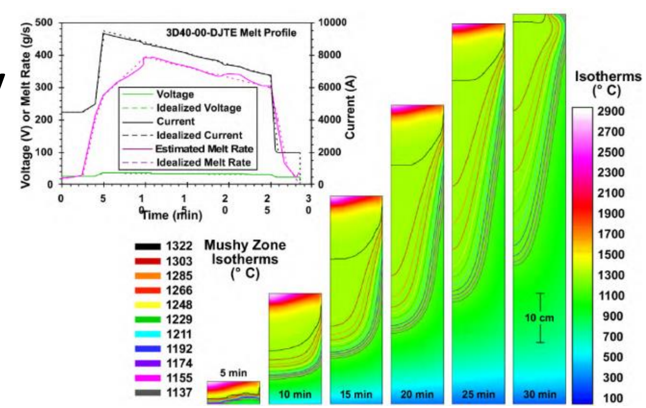

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Los Alamos
NATIONAL LABORATORY

Summary of:

Experimental Investigation of the U-Mo Solidus & Liquidus

TMS 2023

March 19th - 23rd, 2023

Kara E. Luitjohan & Seth D. Imhoff

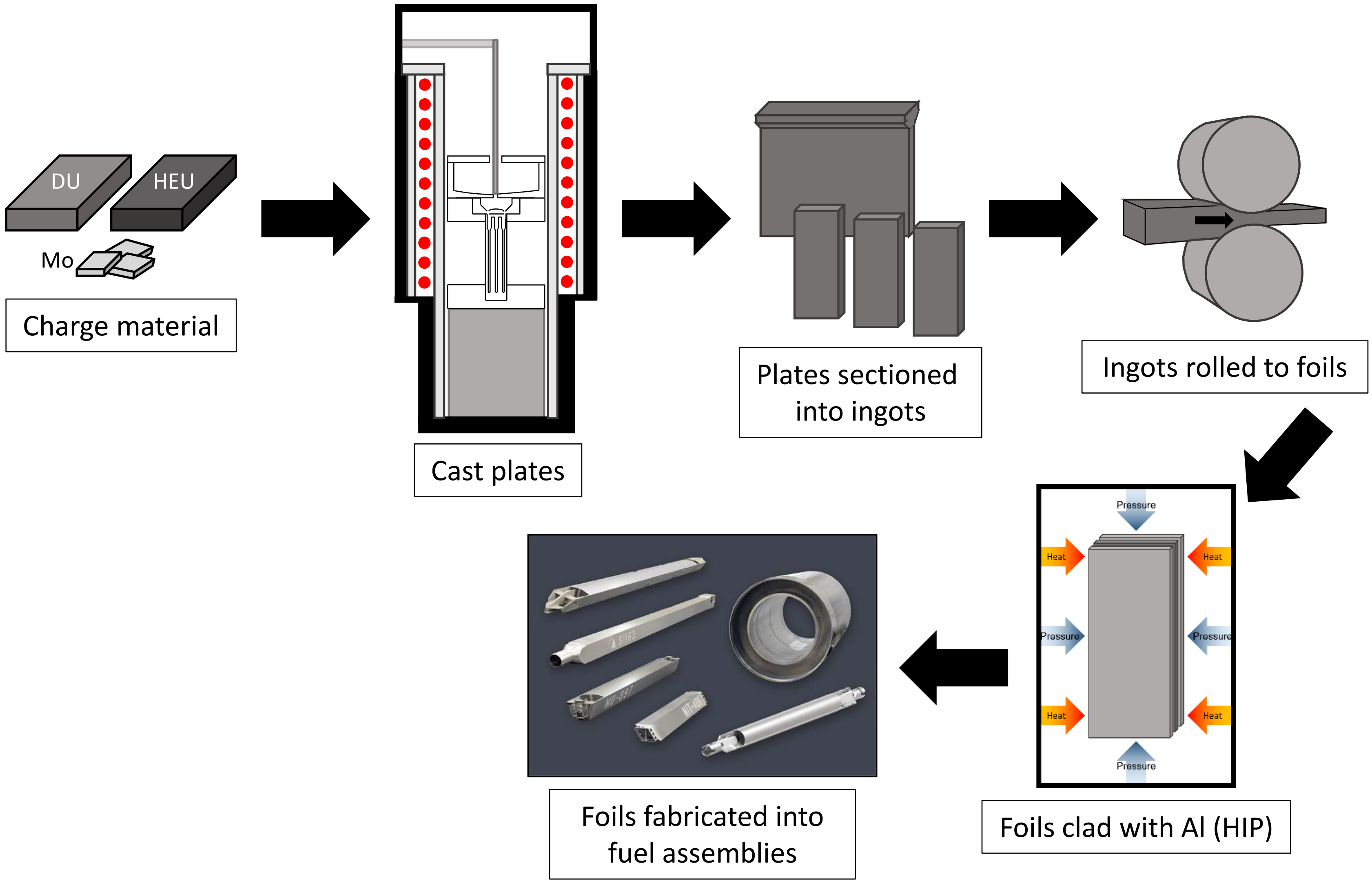
Sigma Division

United States High Performance Research Reactor (USHPRR) Project

- Convert various USHPRRs from high enriched uranium (HEU) → low enriched uranium (LEU) fuel
- Monolithic LEU-based metal fuel foil : U – 10 wt% Mo

Advanced Test Reactor (ATR) at Idaho National Laboratory (INL)

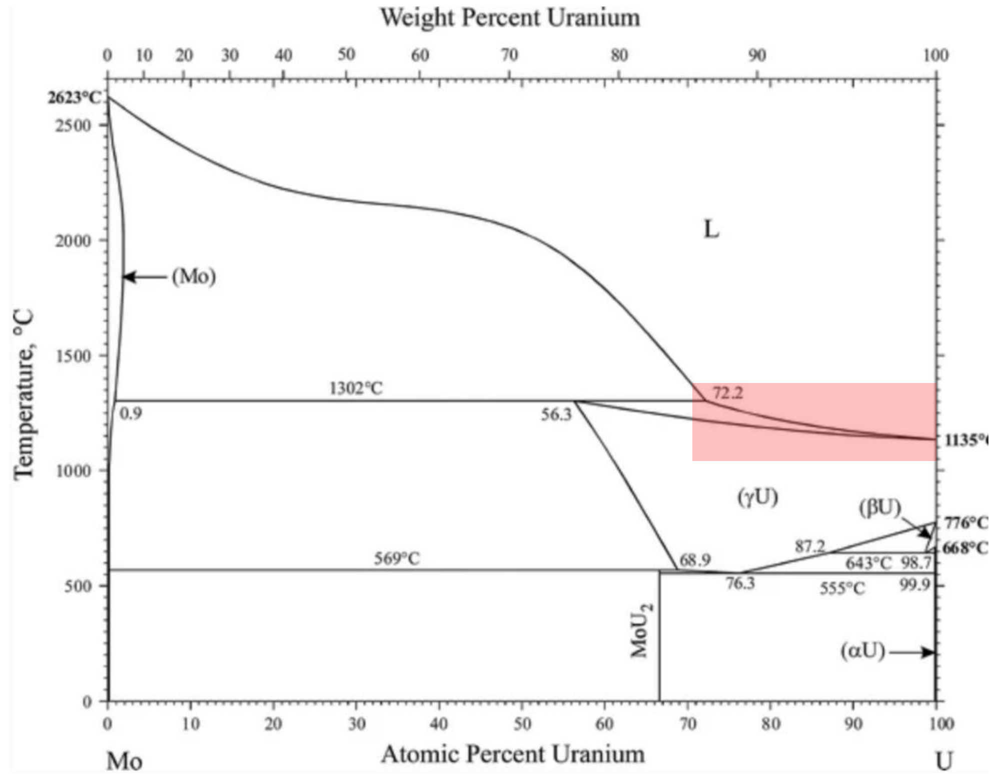
USHPRR fuel fabrication process



LR Hubbard, CL Arendt, NJ Lombardo, CK Clayton, C Lavender. PNNL-29971, 2020.

Phase relationships : 1st information referenced when making processing decisions

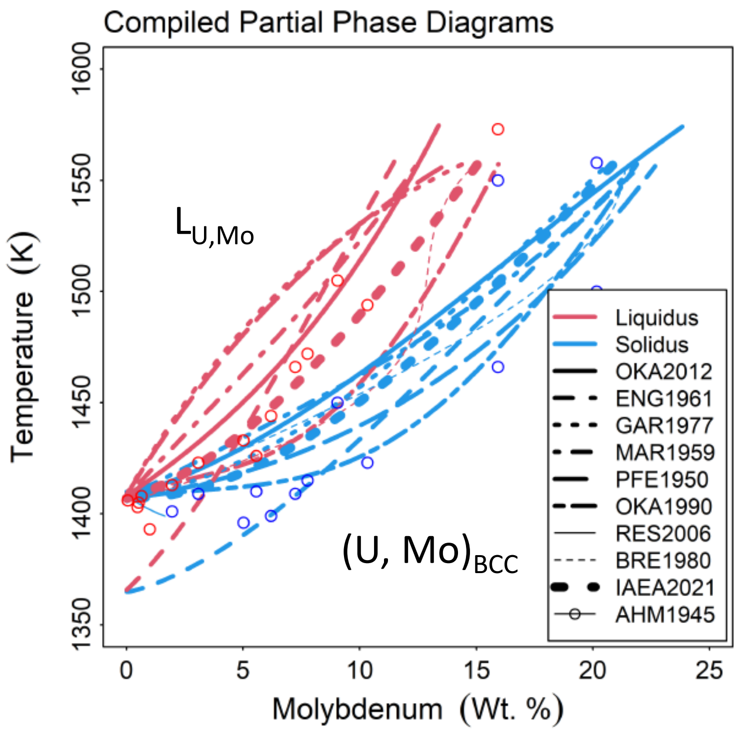
- Mo is a γ-phase (BCC) stabilizer
- Solid-state thermodynamics, kinetics, physical properties, etc. studied in depth
  - Important to fuel lifetime and performance
- Properties most relevant to casting…



One proposed U-Mo phase diagram from H. Okamoto (2012)

Relatively unconstrained phase boundary problem

- Through 75 years and at least 10 relevant proposed phase diagrams, still a lack of data in the U-rich S/L region
  - Little or no direct experimentation of the solidus and liquidus formally produced since 1977

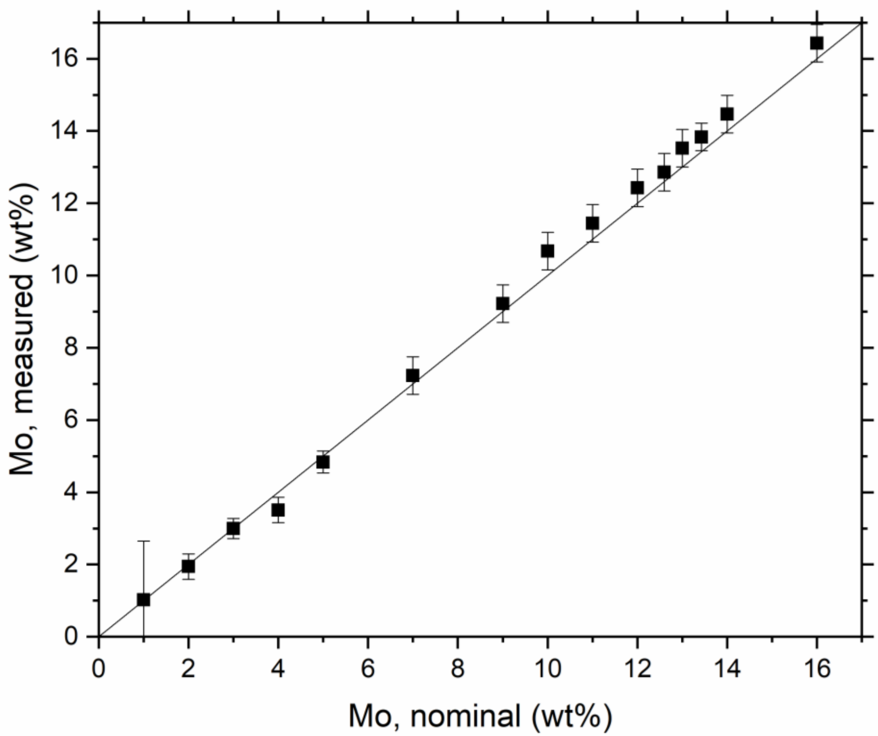


Collection of some of the liquidus and solidus boundaries as defined since 1945

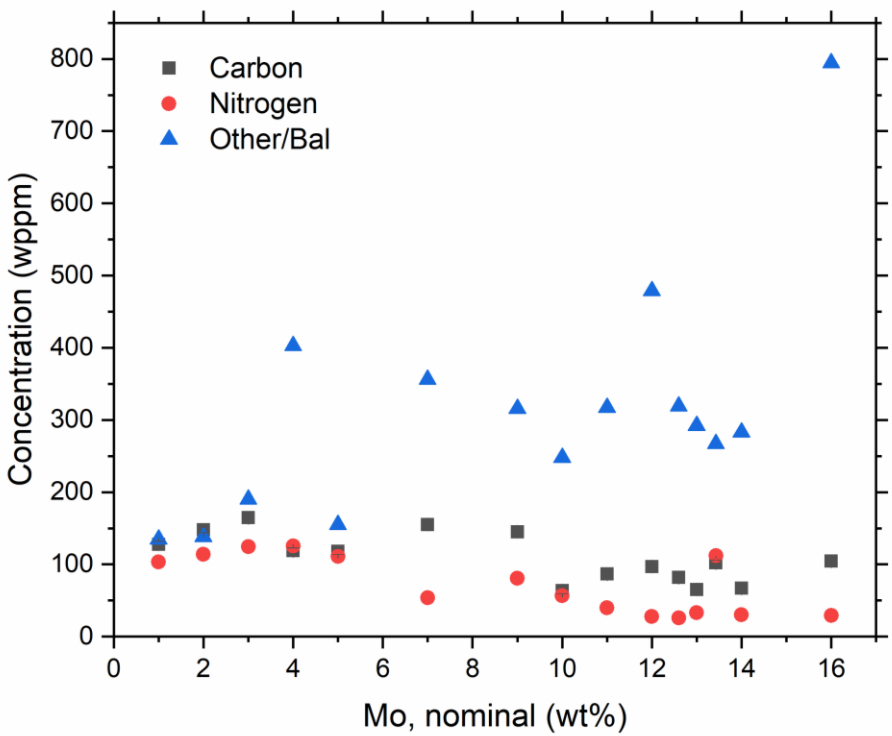
Goal : provide data with highly characterized uncertainty

Chemistry measurements

ICP-OES

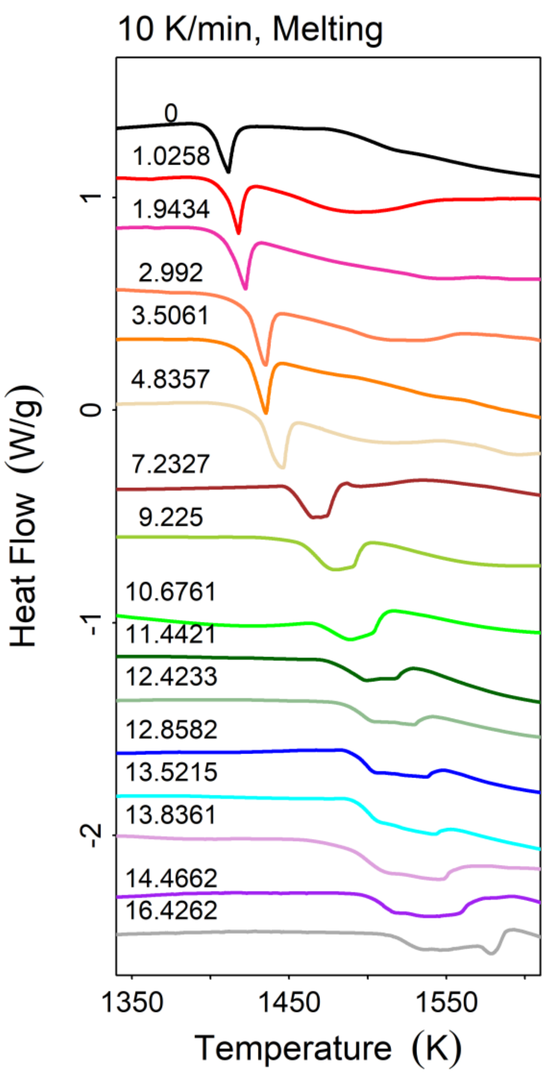


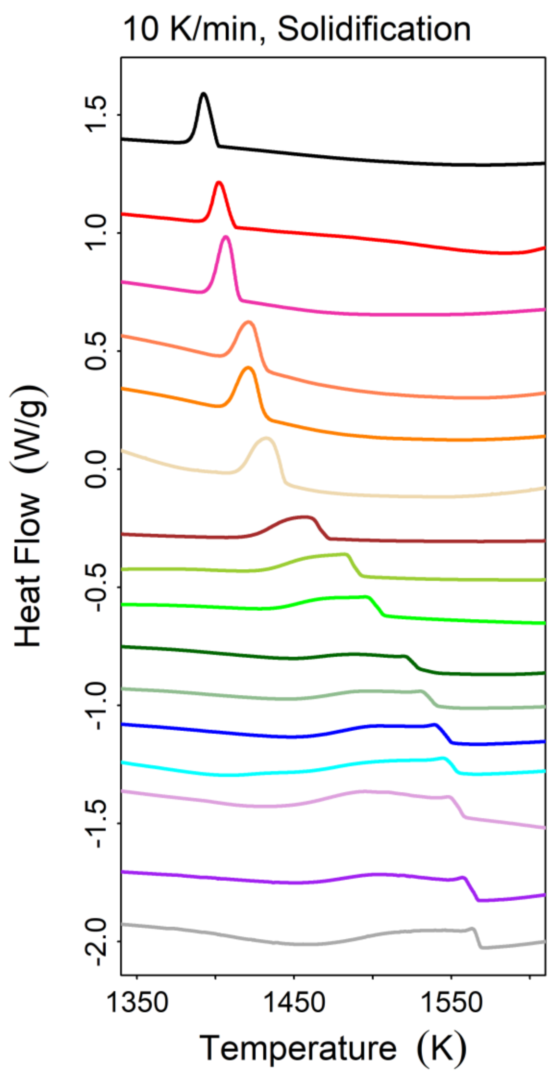
Combustion analysis (LECO) & ICP-MS

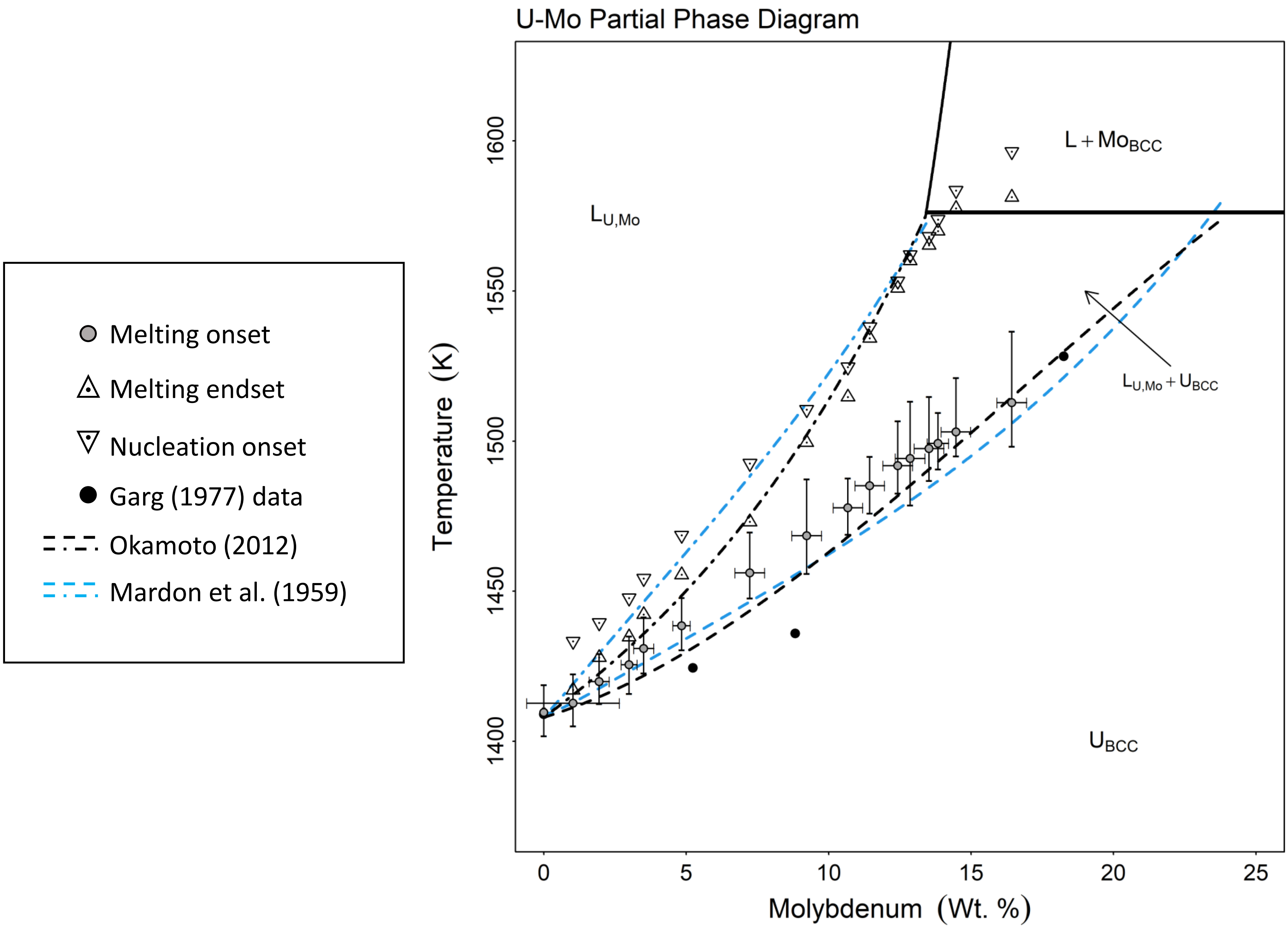


Carbon and nitrogen : standard deviation ~4 wppm

Subset of TA data for all compositions showing melting & solidification at 10 K/min







Melting onset
Melting endset
Nucleation onset
Garg (1977) data
Okamoto (2012)
Mardon et al. (1959)
U-Mo Partial Phase Diagram
L + Mo_BCC
L_U,Mo
L_U,Mo + U_BCC
U_BCC
Temperature (K)
Molybdenum (Wt. %)
1400
1450
1500
1550
1600
0
5
10
15
20
25

Our data indicates admixture model is a useful, justified assumption

- Limitations of measurement accuracy impacts ability to support any trends more detailed than this baseline behavior
- Admixture model (solid line) within the composition region of interest

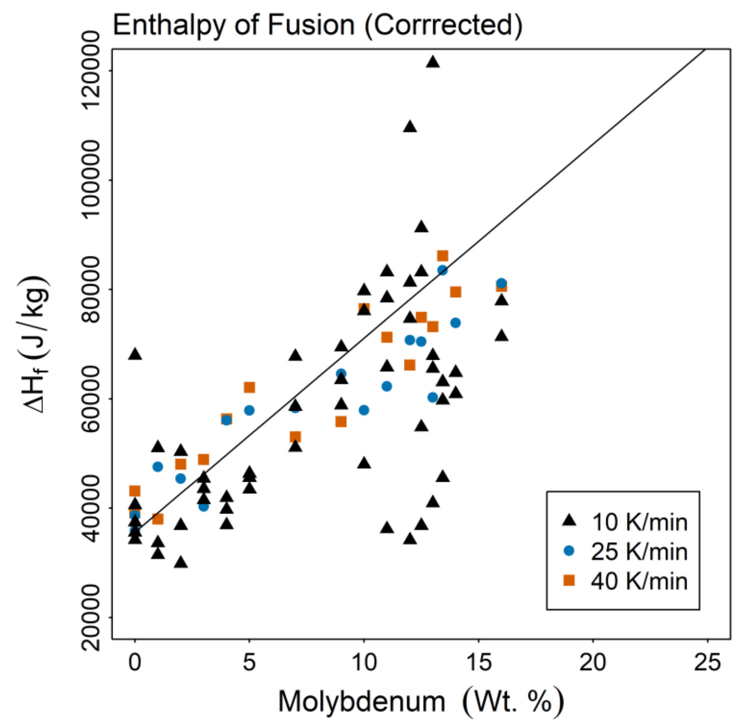
$$\Delta H_f = \frac{\left[\Delta H_{f,Mo} X_{Mo} + \Delta H_{f,U}(100 - X_{Mo})\right]}{100}$$

$$\Delta H_{f,U} = 35589.13 \frac{J}{kg}$$

Konings and Benes (2010)

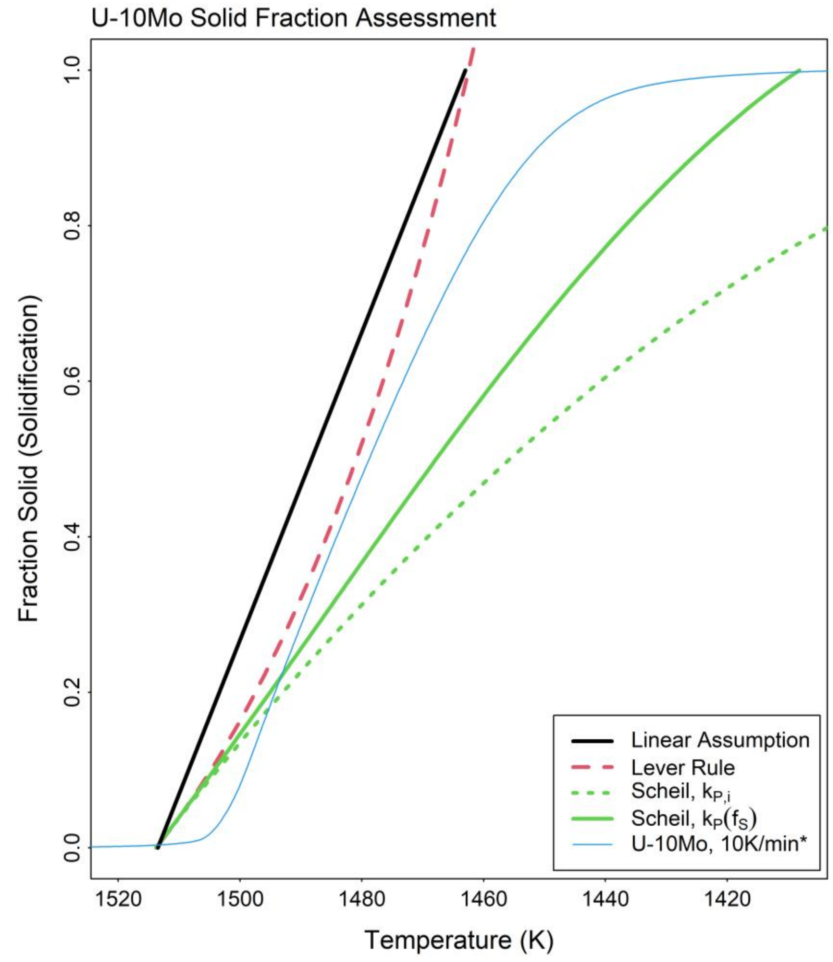
$$\Delta H_{f,Mo} = 390617.8 \frac{J}{kg}$$

Dinsdale (1991)



Importance of enthalpy in casting simulations : phase fractions

- Modeling always requires assumptions
- Need to be able to justify those assumptions
  - Linear : typical default in software packages
  - Lever rule : easiest to calculate give a good diagram
  - Scheil : good limiting solution
  - Admixture model : better justification for use



Evolution of Casting Modeling at LANL for U-Mo Monolithic Fuels

Seth D. Imhoff

ADUMTT and Modeling Summit

February 7th, 2023

Los Alamos NATIONAL LABORATORY

NNSA National Nuclear Security Administration

M3

A large cast of characters over the years

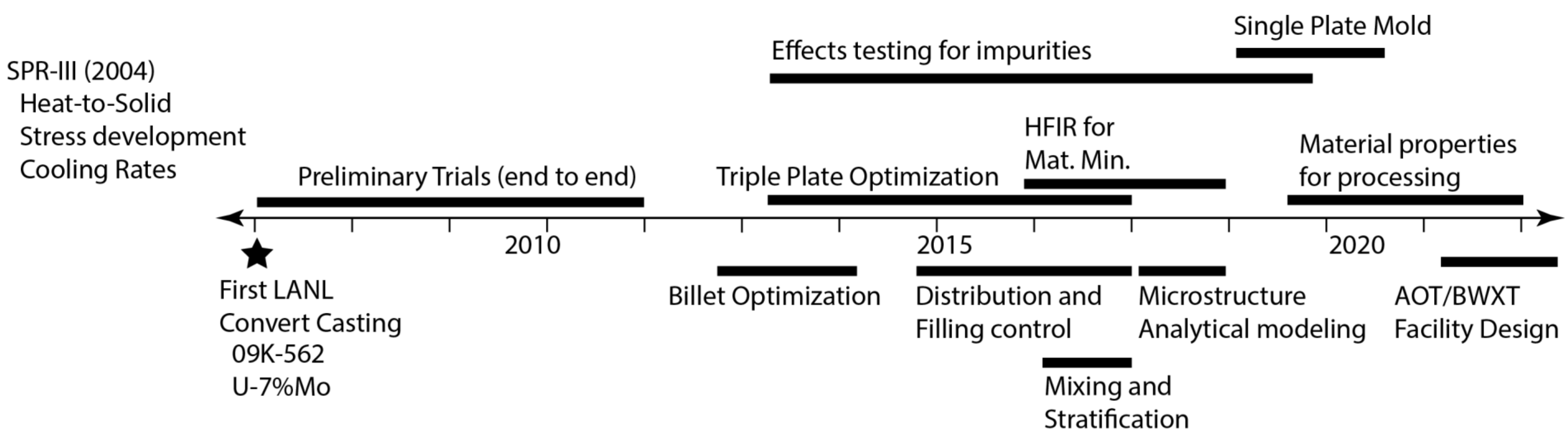
- Deniece Korzekwa
- Kin Lam
- Neil Carlson*
- Justin Crapps
- Paul Gibbs
- Gabrielle Kral
- Karen Chen
- Zach Jibben
- Rob Aikin*
- Seth Imhoff*
- Kara Luitjohan*

* Indicates work has been directly used for this talk.

This list is purely focused on modeling contributions and only a fraction of total work. My apologies to anyone that I've missed.

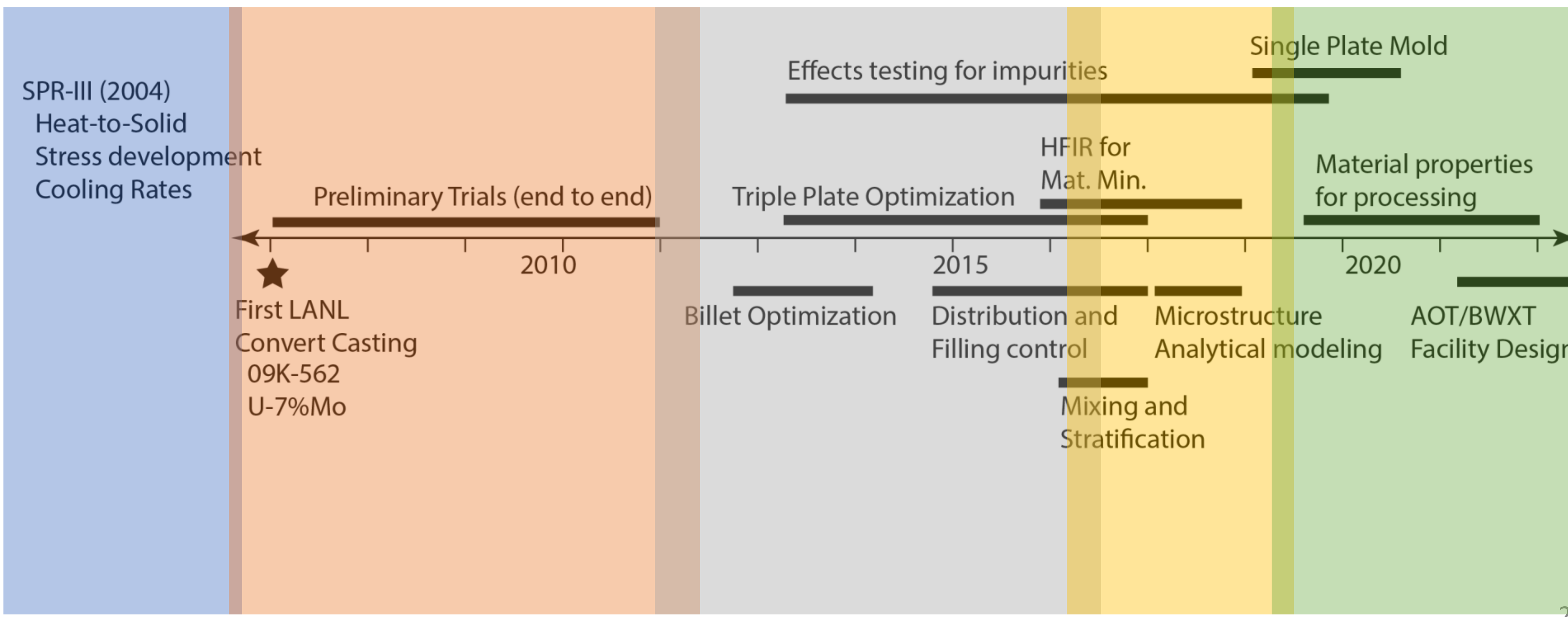
A purposeful path in the moment and hindsight

- My own introduction (the 2013 state of affairs)
  - “Model Thinking” was encouraged and rewarded
  - Mantra: Process modeling whenever possible
  - Production modeling guiding areas of attack



A purposeful path in the moment and hindsight

- Prior knowledge and familiarity
- Gaining insight, initializing baseline
- Optimization
- Solving Problems and ready for changes
- Preparing for the future



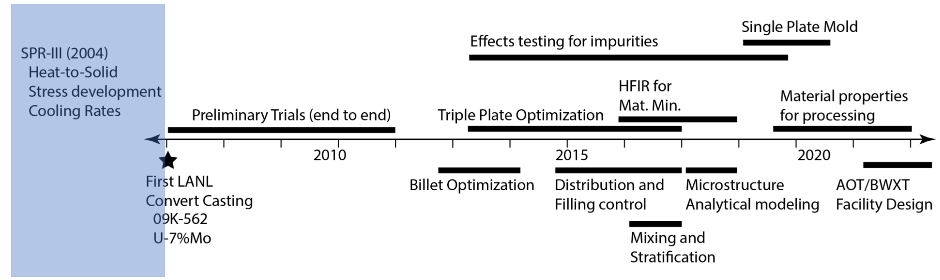
Pieces of the puzzle that need to be fit together

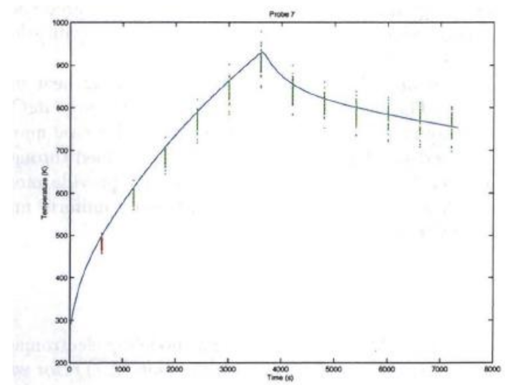
"How does this help us make fuel?"

- Part yield: Optimization directed toward defect control
  - Billet mold design
  - Triple plate distributor design
- Material yield: Optimization directed toward near net shaping
  - HFIR "shaped" mold design
  - Hot top (feeding) minimization
- Process upset response: Provide insights for where to look
  - Crucible stratification study
- Recycling: Material and defect modeling for reuse
  - Filtration and other process variables

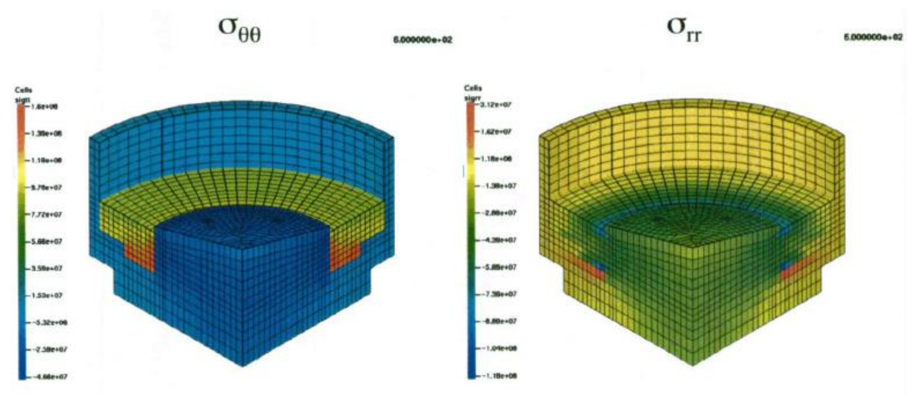
Prior knowledge and familiarity

- Leverage code validation and uncertainty
  - What has been expanded or degraded?
- Is prior data able to be accessed or applied?
- Leverage personnel
  - Which resources are still available?
  - Are we maintaining people-resources?
- Are we setting the next project/program up for success?





(a) 04K-450



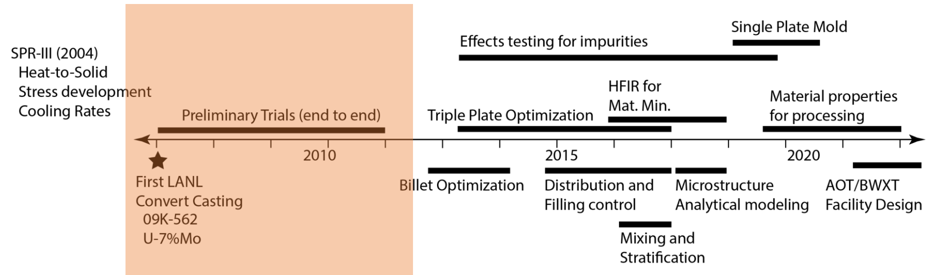
Prior Truchas verification, validation, and quantification of error work. [1,2]

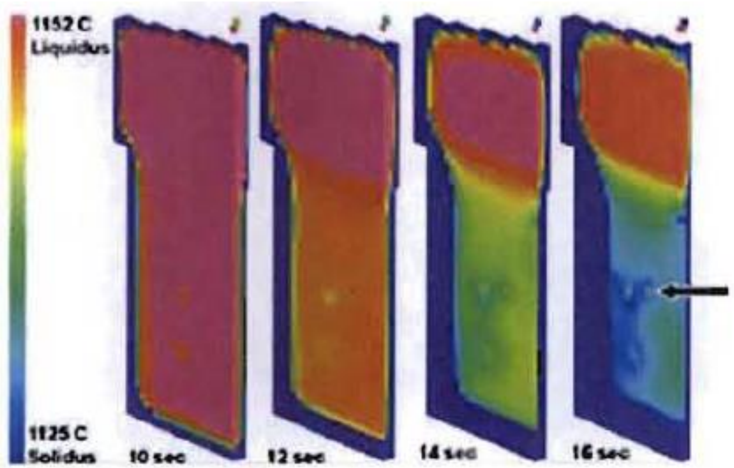
Gaining insight and initializing the baseline

- Initial models quickly become outdated however…
  - Initial trials will come with problems.
  - Models can capture information between data points to provide additional context.

- What should we expect?
  - Imperfection
  - A budding and growing base of experience.

- What could have been improved?
  - Radiographic or metallographic confirmation of suspected trouble areas. (confirm where possible)
  - Grow resources for responsiveness.

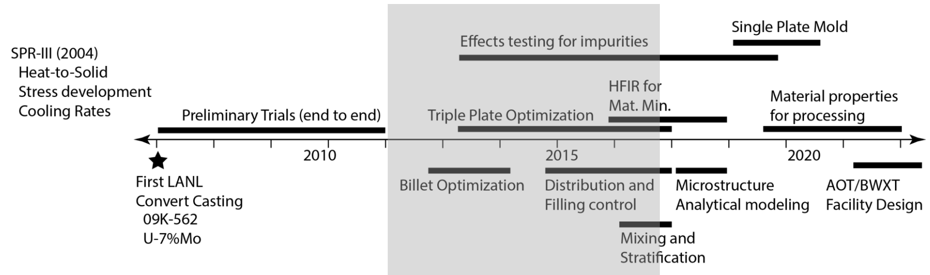


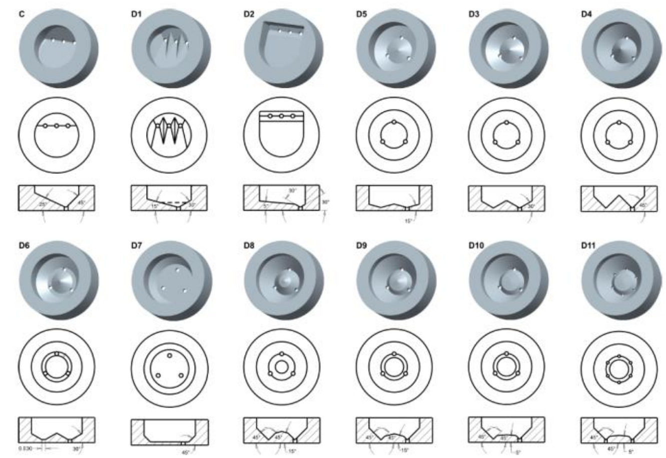


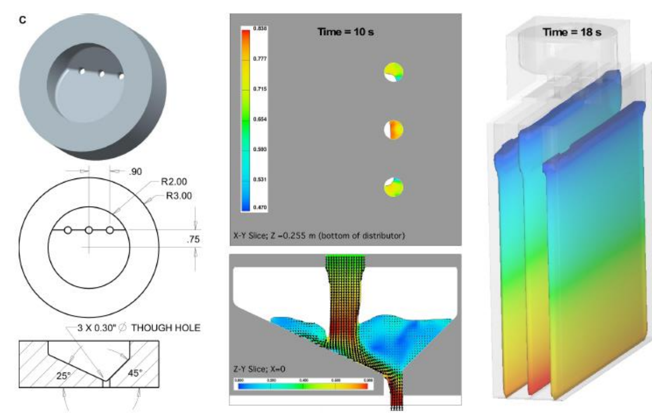
Simulated and physical first casting trial. [3]

Optimization

- Use as an engineering tool
  - Idea OR concept downselect
  - Minimization of trials
  - Experimental window assessment
- What should we expect?
  - Identified problems may be addressed directly.
  - Efficient downselect still takes time.
  - Improvement list piles up.
- What could have been improved?
  - Goals for model extension
  - Better planning to use model-experiment comparisons for V&V
  - Integration with other models or sources of information





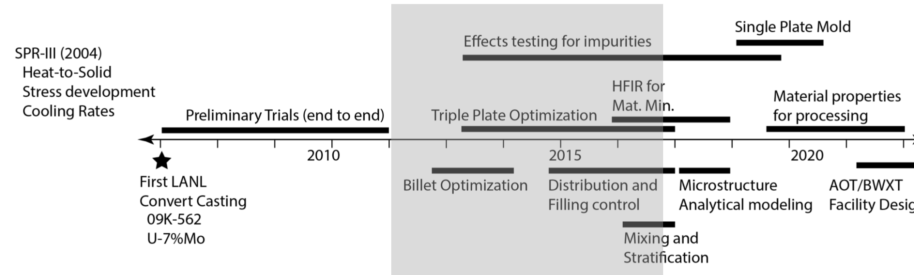


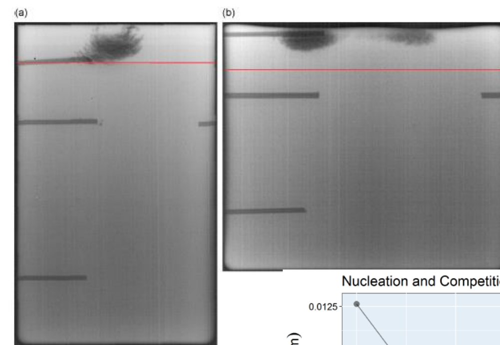
Ref [4]

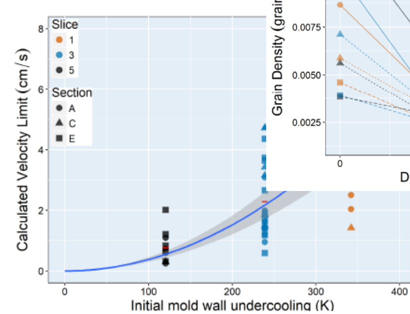
Optimization Supplement

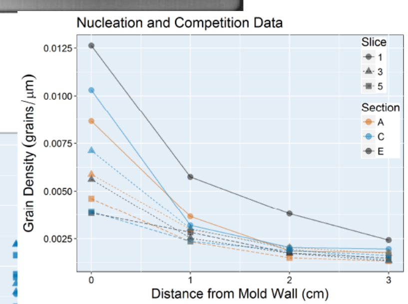
- The pressure may be on, but successes can mount
  - Billet mold optimized. (not selected) [5]
    - Anticipated part yield improvement
  - Triple plate distributor design finalized [4]
    - Anticipated part and material yield improvement
  - HFIR Complex Casting Shape initialized [6]
    - Anticipated material yield improvement
    - Paired with novel deformation processing
  - Triple plate casting improved (selected) [4]
  - As-cast microstructural modeling [6]
    - Prediction of basic structure information.
    - Able to be paired with homogenization models
  - Effects testing for impurities [7, 8]
    - Enabled cost-benefit for concepts such as filtration

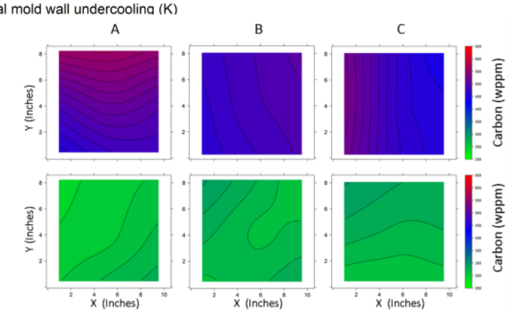
- A lot of information gets generated. This must be managed at a high level!

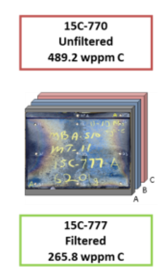






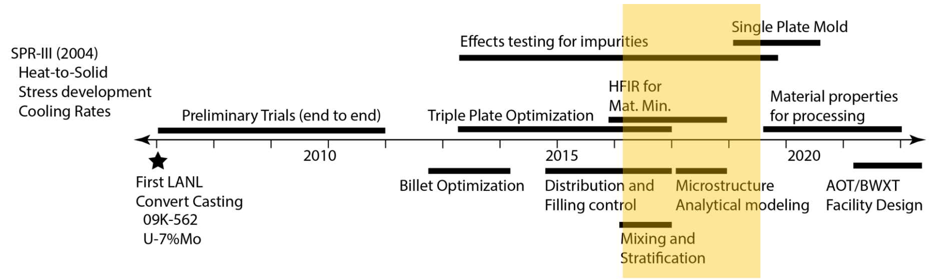


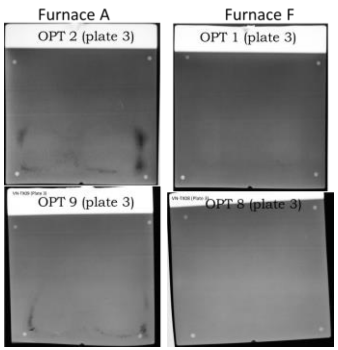


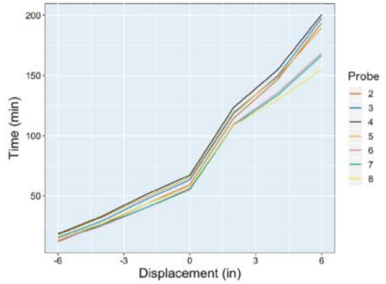


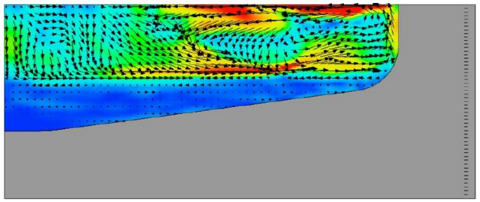
Solving Problems, ready for changes

- Responsiveness becomes key
  - Final downselect is pushing schedule
  - Identification or quantification of process upset
  - Experimental window assessment
- What should we expect?
  - “We saw this, does the model give us a hint?”
  - What information is useful to the equipment operator?
  - More physics may be required…
- What could have been improved?
  - Testing of model extension
  - Communication can always be better.





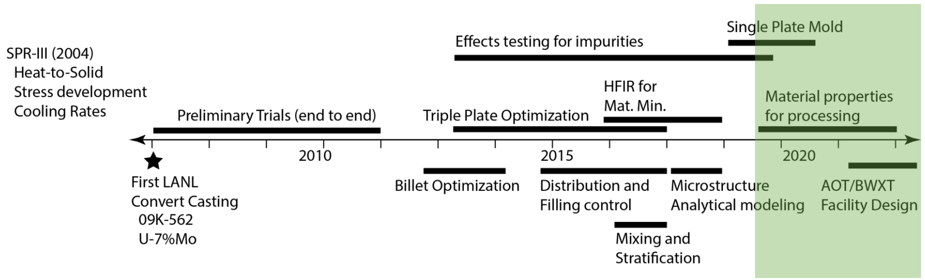




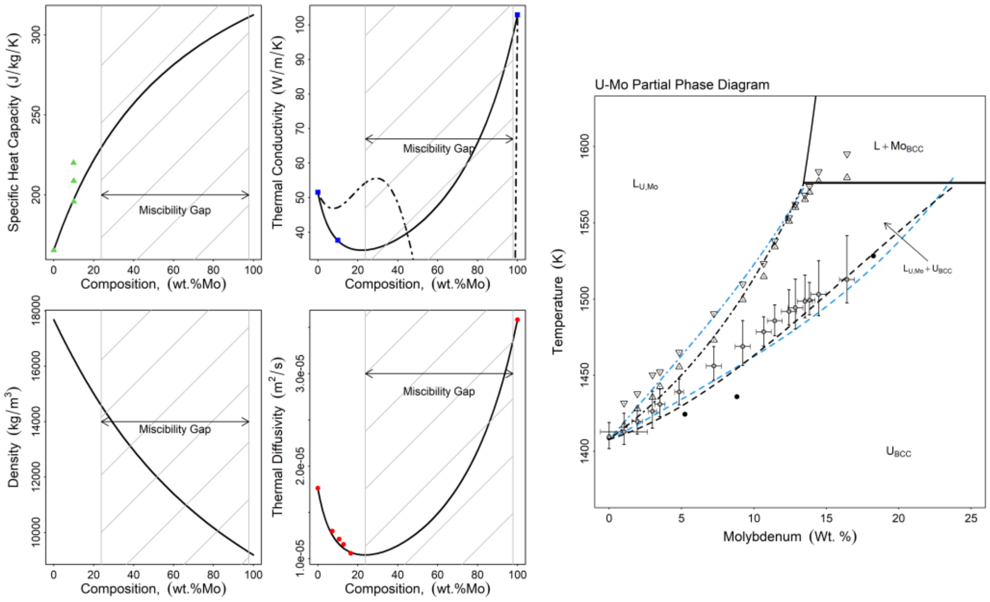
Outputs for fast-response problem solving [9, 10]

Preparing for the future

- Retrospectives to inform
  - Were we missing something?
  - What is coming next?



- What should we expect?
  - Implementing new physics models
  - Improving datasets [11, 12]
  - Improving assumptions
  - Assessing for gaps in documentation.
  - Assessment of prior work for V&V and Q of U.
  - Anticipation of the next customer



- What could have been improved?
  - We will know in the future!

What did we get and where do we go?

- Modeling made a significant impact on how stages of development were tackled, explained, and resolved.

- Modeling helps gets the most out of experiment

- Where were the opportunities in hindsight?
  - Integration with other modeling efforts
  - Early verification of simulated hints
  - Including new features/physics early.

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Thank you