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Design of high-strength Aluminum alloy castings – A case study on the importance of multicomponent diffusion to materials design

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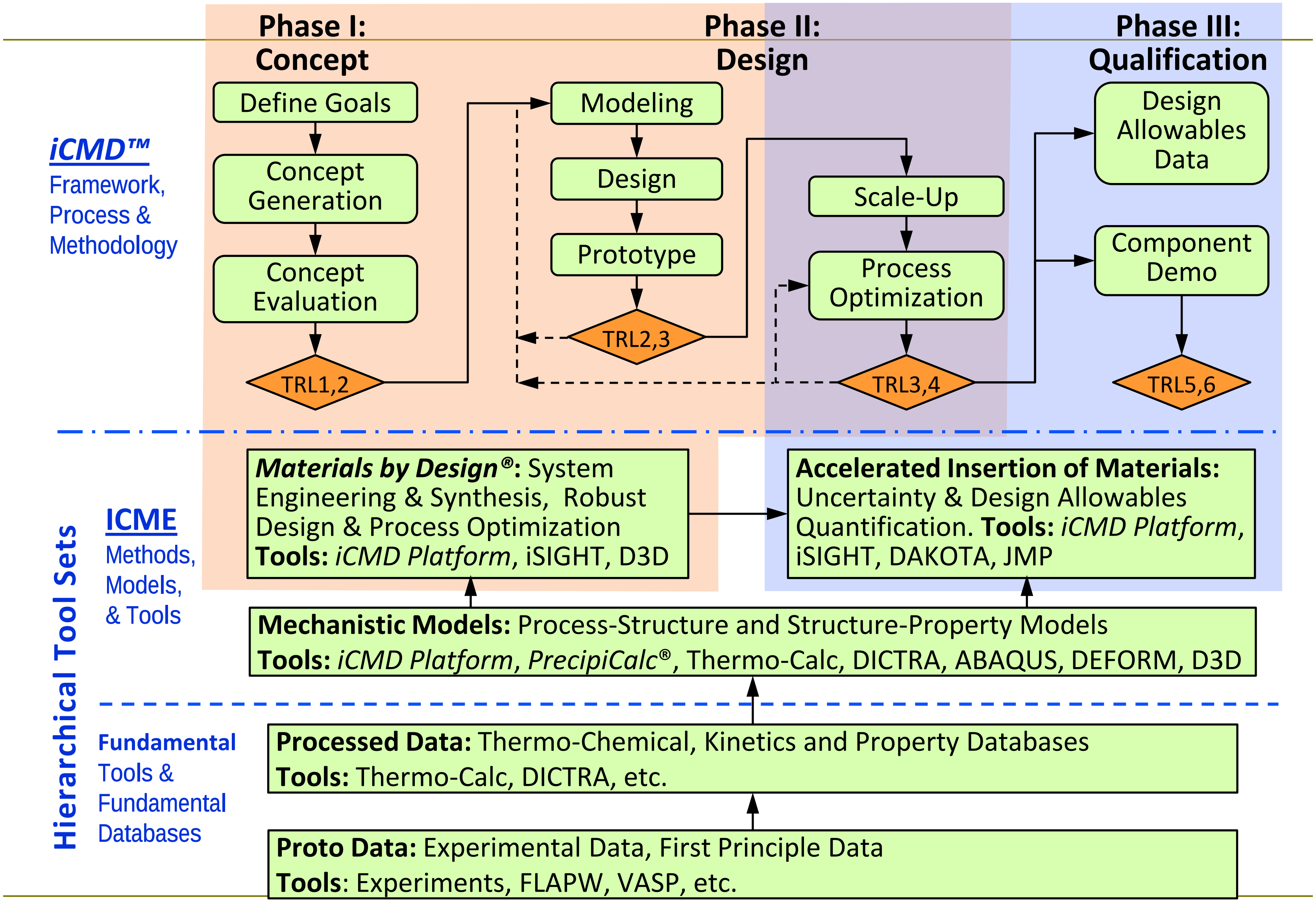
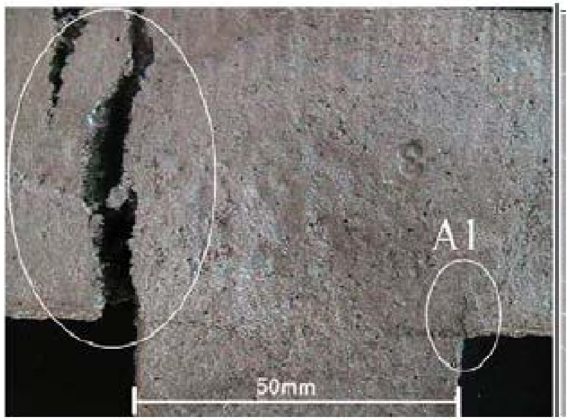
Schematic of *iCMD*™ Framework



What Problem Were We Trying to Solve?

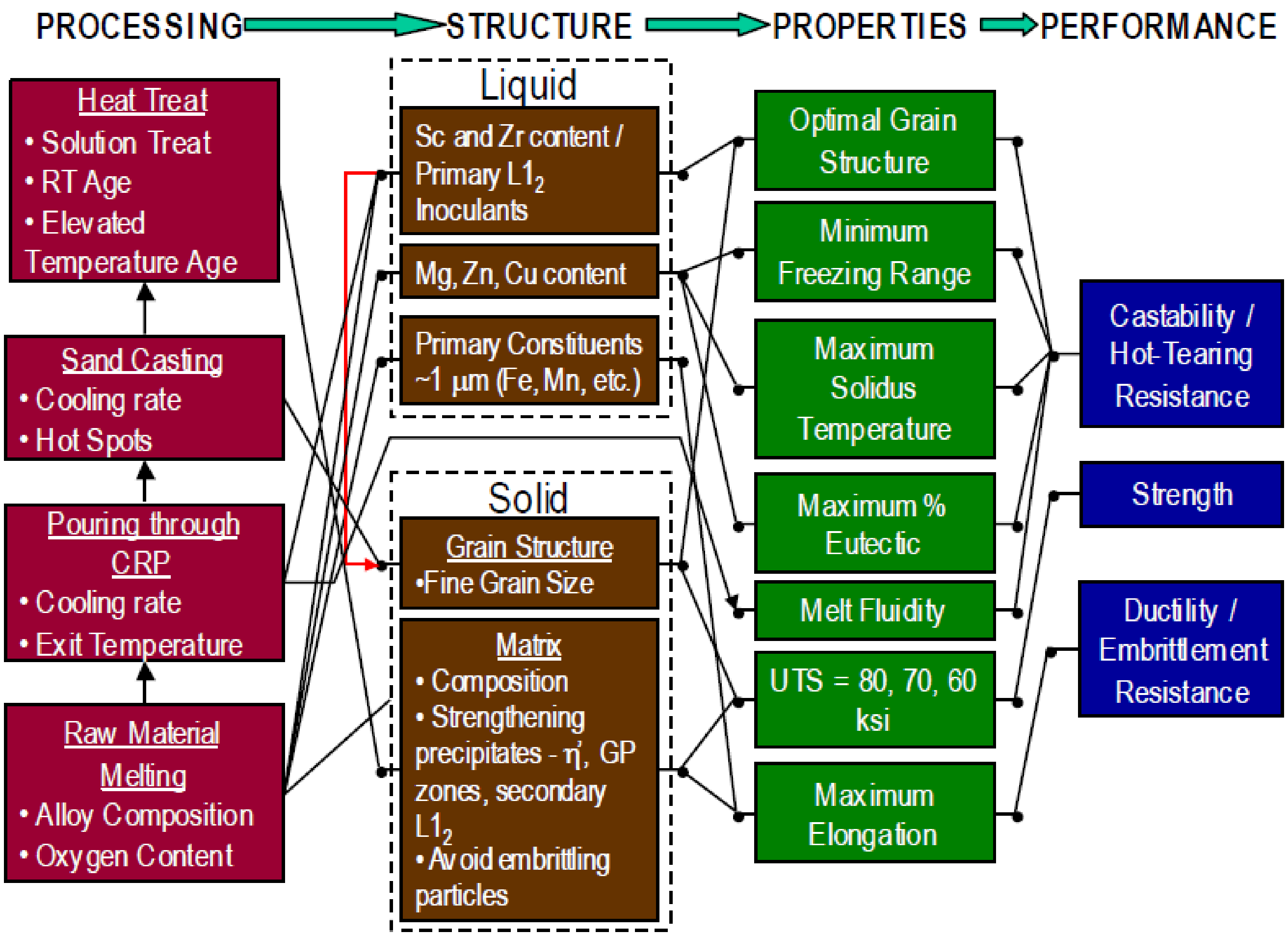
- Lightweight turbopump housings have traditionally been constructed as a welded fabrication of mostly wrought components.
- This approach introduces immense complexity and often produces multiple layers of inspection and rework that adversely impact the cost and schedule of part production.
- Transitioning to net shape cast structures is a successful strategy for controlling these costs
- However, high strength Al alloys are prone to hot-tearing during casting limiting their castability



Hot tearing of a high strength A-206 Al alloy
From M.R. Nasr Esfahani and B. Niroumand, *Materials Characterization, vol. 61, 2010, pp. 318-324*

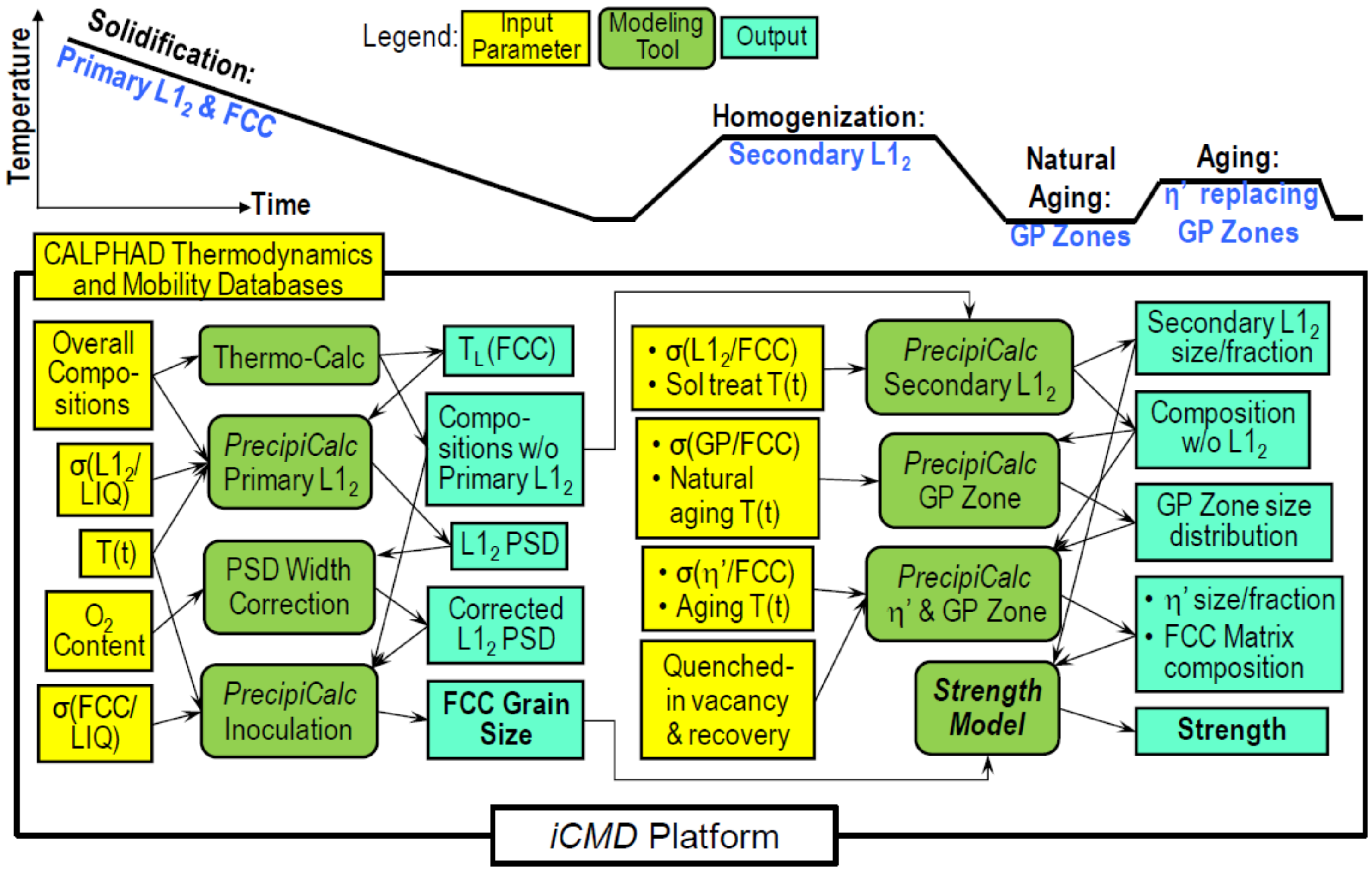
Technical Objective:
Develop a hot tearing resistant castable 7xxx-series Al alloy capable of achieving RT yield strength > 70ksi

System Design Chart for Castable 7xxx-series Al Alloys



•What structure gives the desired properties?
•What process results in the desired structure?

Integration framework for the design of castable 7xxx aluminum alloys



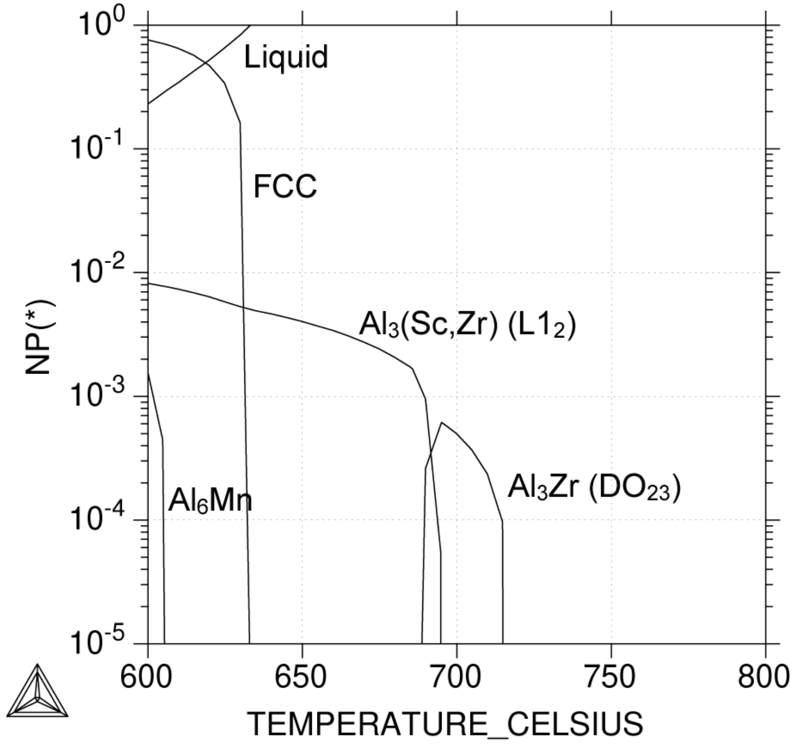
•*We have to design for the entire process – not just a single step*

Databases – Thermodynamic and Mobility – They form the backbone of CALPHAD-based design

Development of Thermodynamic Database

Multicomponent thermodynamic database for stable and metastable phases successfully developed for Sc-modified 7xxxx Al alloys

- Al-Zn-Mg-Cu description from COST507 database evaluated and revised
- Sc added to revised Al-Zn-Mg-Cu database, including all relevant binary and ternary compounds
- $L1_2$ $Al_3(Sc,Zr)$ thermodynamics developed, incorporating solubilities of other elements
- **Thermodynamic descriptions developed for metastable GP zones and η'**

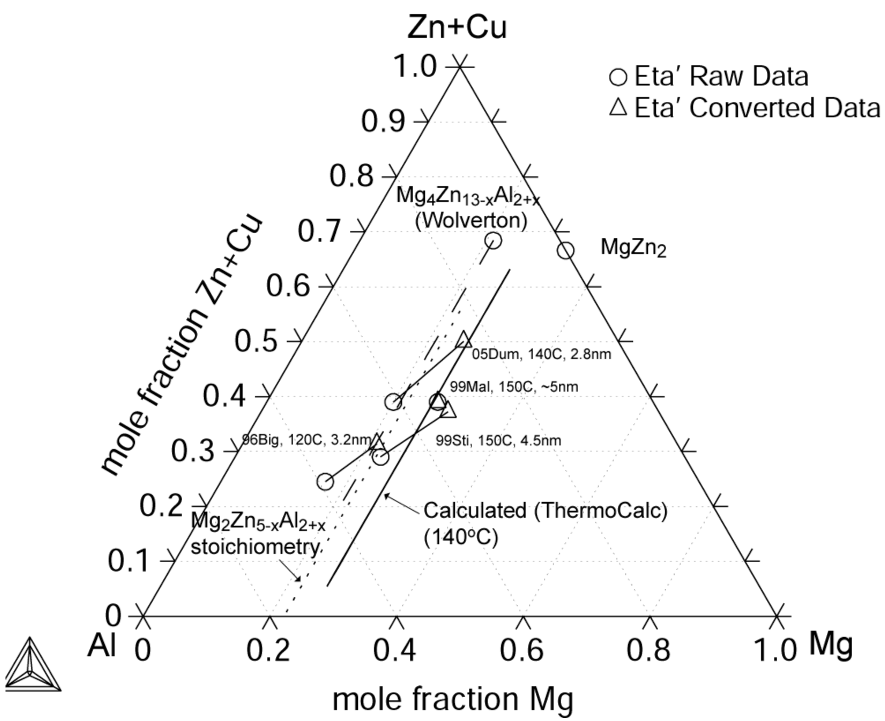


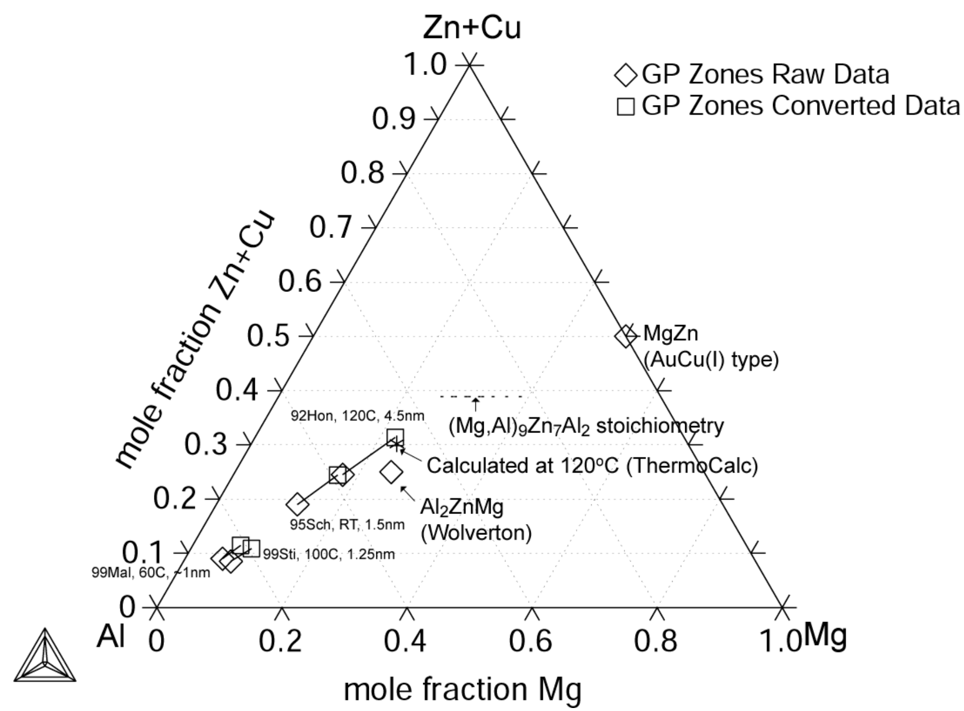
Calculated phase fractions in a 7xxx-series alloy using developed database

Metastable Phase Thermodynamics Description

- Low-temperature aging process of Al alloys – metastable phase variants are the primary strengthening phases
- For 7xxx-series alloys, typical precipitation sequence is:

  FCC (supersaturated) → GP zones → η′ (T6) → η (T7)
- Metastable phase descriptions were assessed using measured atom-probe data and DFT calculations





Mobility Database Development for Al based alloys

Liquid Phase

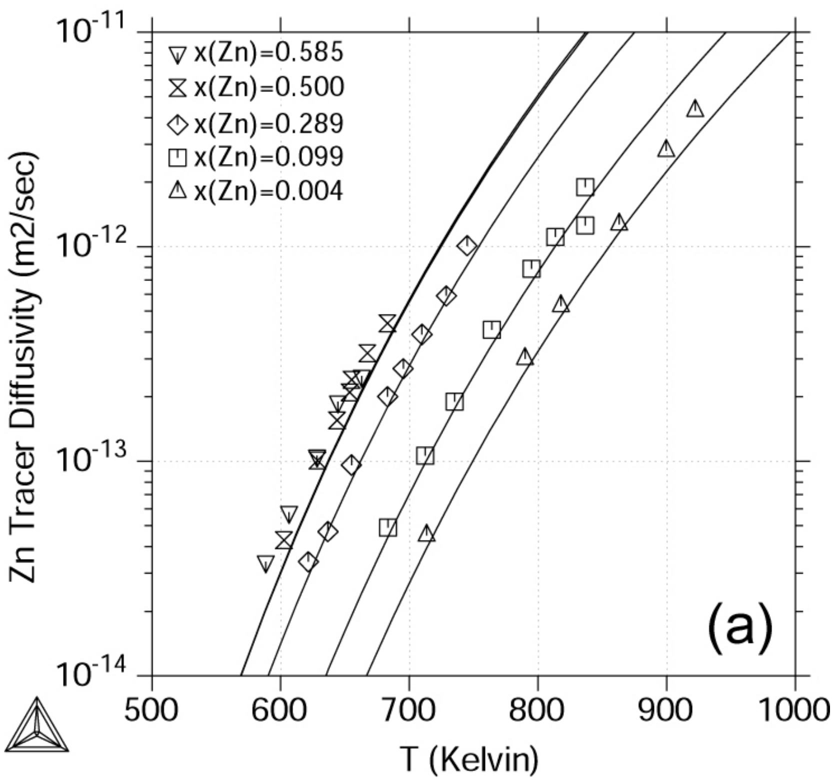
- ***Precipitation of primary $L1_2$ - $Al_3(Sc,Zr)$ from liquid is the most important phenomena***
- ***Cannot rely on D=1e-9m^2/sec for all components in liquid***
- Parameters for impurity diffusion of elements in liquid Al, liquid Cu, liquid Zn collected from literature.
- Estimates made when experimental data not available
- No interaction parameters included because of lack of experimental data.
- *PrecipiCalc* simulations carried out in the Al-Sc binary system to verify the liquid diffusivities.

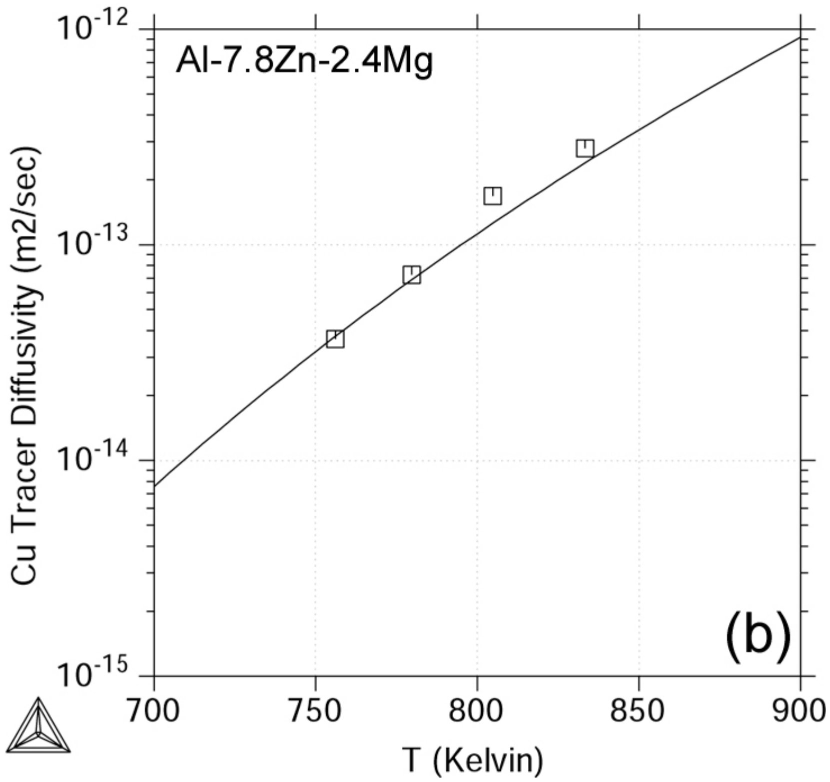
FCC Solid

- **Precipitation of strengthening phases (GP zones, η′, η-$MgZn_2$) and secondary *$L1_2$-$Al_3(Sc,Zr)$* are the important phenomena.**
- **Also important – back diffusion in FCC during solidification**
- Full mobility matrix (including binary interaction parameters) assessed for the Al-Mg-Zn-Cu system.
- Sc, Zr, Fe and Mn present in impurity amounts; Impurity mobilities should be sufficient to capture the kinetics of those components.

Calibration and Validation of Mobility Database

Mobility database successfully developed for Sc-modified 7xxxx Al alloys for the Liquid and FCC phases.



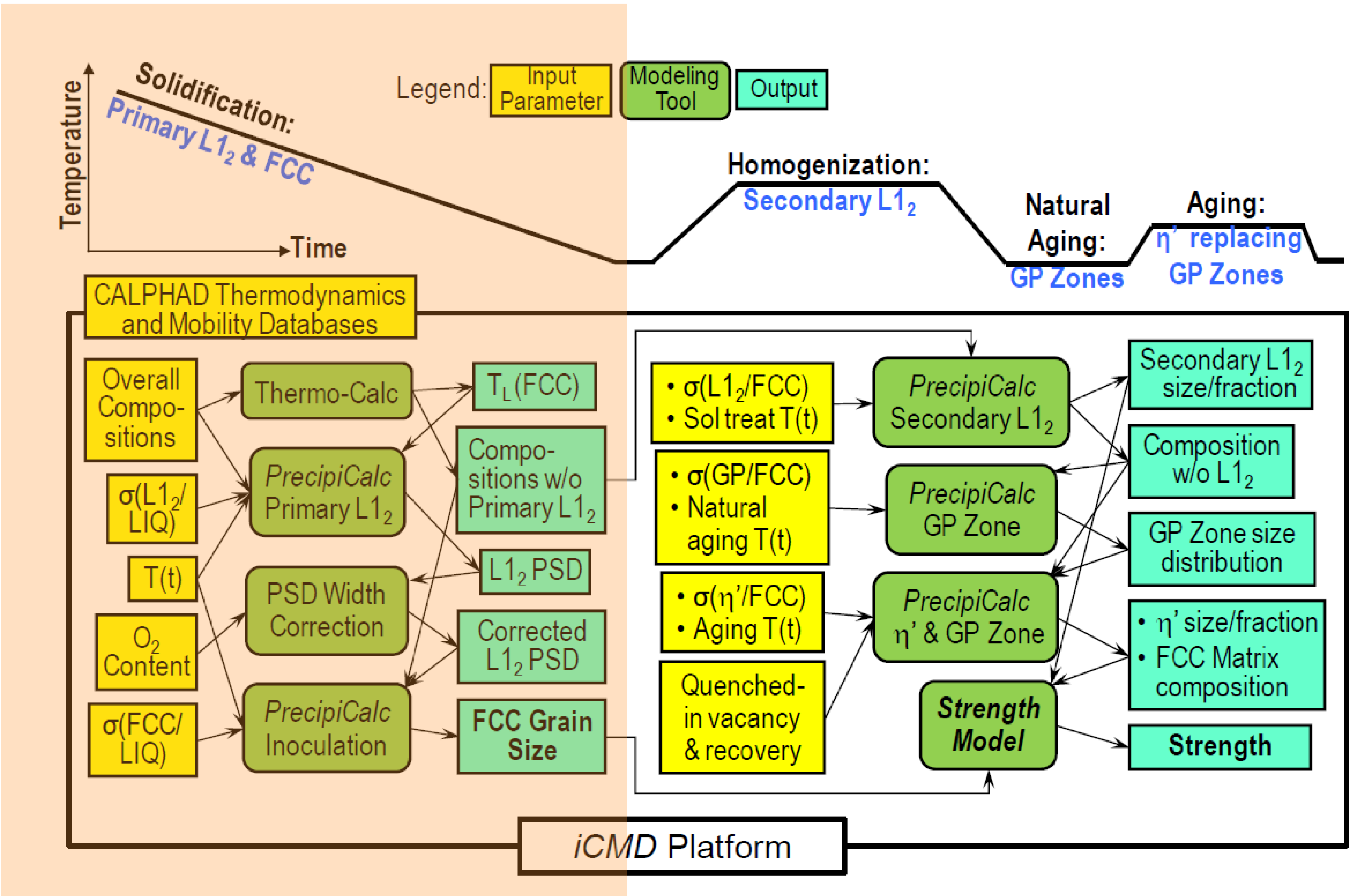


Comparison of diffusivity predictions using developed mobility database (solid lines) with experimental data (a) Zn tracer diffusion in Al (b) Cu tracer diffusion in Al-Zn-Mg alloy

In-situ Inoculation Model –

- Inoculation is a commonly practiced method to refine grain structures in cast Al alloys
- Primary $L1_2$ particles can be designed to form prior to the main solidification and act as in-situ inoculants to refine the FCC grain structure
- This results in better hot tearing resistance and castability

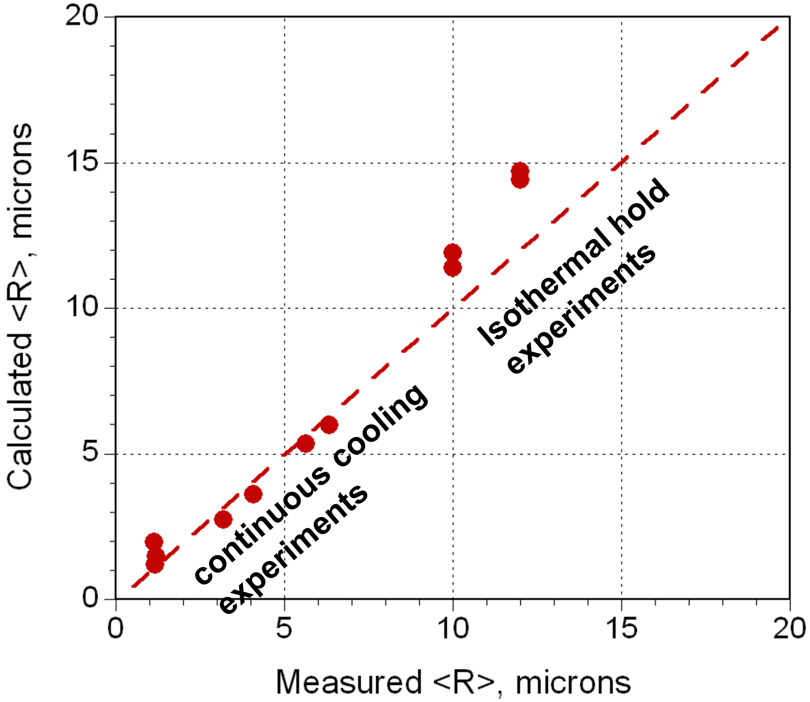
Integration of Primary $L1_2$ and Inoculation Simulation



Modeling of Primary $L1_2$ Precipitation

Continuous cooling samples

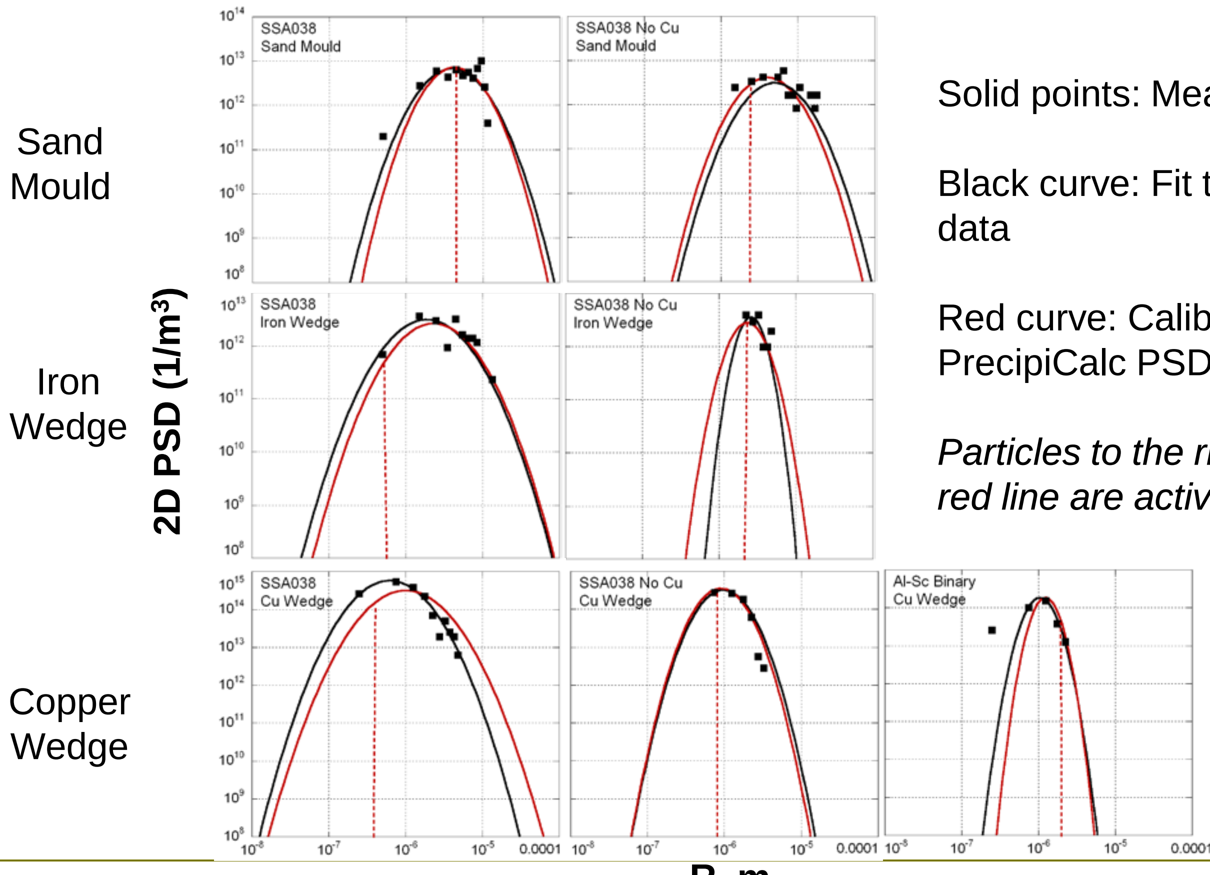
Cooling Mode	dT/dt (°C/sec)	$L1_2$ Mean Radius (μm)					
		7xxx As-Cast		7xxx-No Cu As-Cast		Al-Sc binary As-Cast	
		Exp. (2D)	Calc. (2D)	Exp. (2D)	Calc. (2D)	Exp. (2D)	Calc. (2D)
Sand Mould	~7.5	6.33	6.26	7.0	6.5	-	-
Iron Wedge	~22	4.09	4.52	3.2	2.9	-	-
Copper Wedge	~200	1.2	1.84	1.18	1.2	1.15	1.42



- $L1_2$/liquid interfacial energy adjusted within PrecipiCalc model to match measured $L1_2$ mean particle size

Heterogeneous Nucleation Treatment for Primary $L1_2$

- PrecipiCalc simulations using a homogenous nucleation model predicted a much narrower distribution of primary $L1_2$ particle sizes as opposed to measured data.
- Oxygen leads to formation of oxide particles which act as heterogeneous nucleation sites for $L1_2$ particles, and broadens PSD width.
- Experimental data was analyzed to correlate measured oxygen content with the width of measured PSD, and a calibration factor was deduced from the correlation.



Solid points: Measured data

Black curve: Fit to measured data

Red curve: Calibrated PrecipiCalc PSD

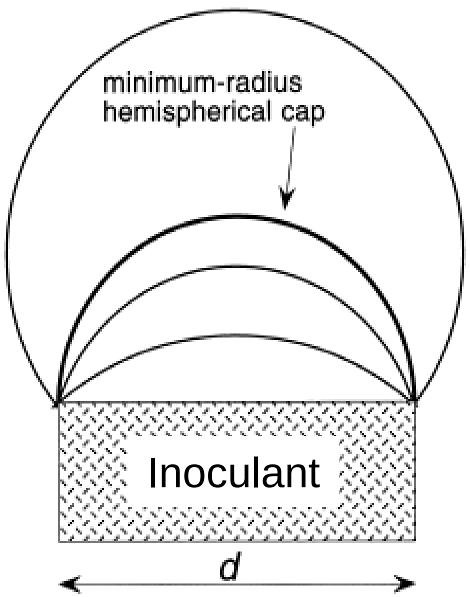
Particles to the right of dashed red line are active inoculants

Inoculation Model Mechanism

Primary $L1_2$ particles act as inoculants for the formation of fine equiaxed grains.

Mechanism of grain initiation and growth (Greer et al., Acta Mat., 2000):

- Onset of growth is controlled by **growth constraint** rather than by nucleation.
- Initial FCC nucleus grows readily across the face of nucleant particle to form thin coating.
- Further growth possible only by reducing the radius of curvature of its interface with the melt (free growth). This radius cannot go below the critical r* for nucleation unless the undercooling is increased, thus reducing r*. The Critical condition for free growth is particle size d = 2r*

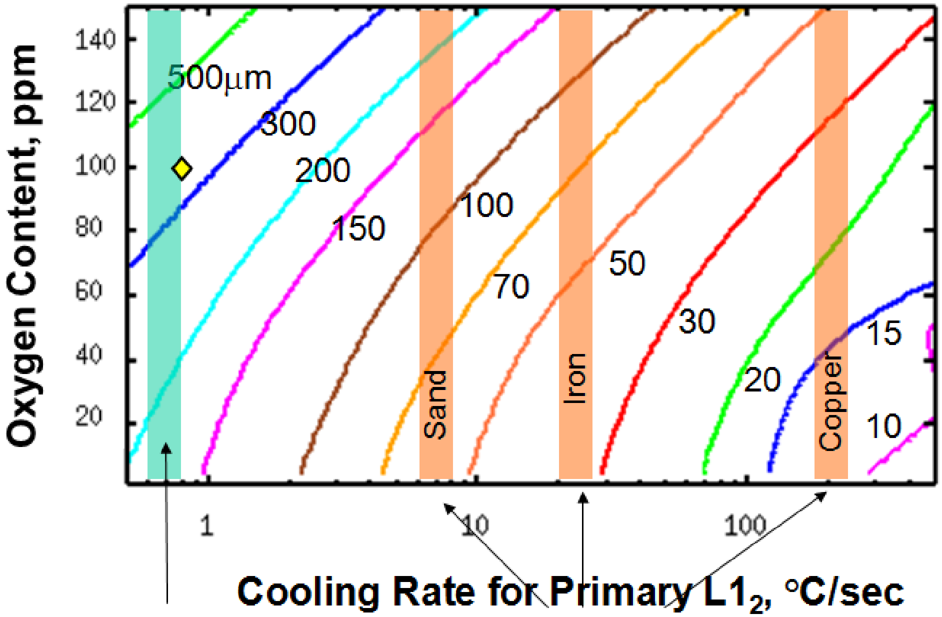


Greer's Inoculation Model implemented in PrecipiCalc:

1. Primary $L1_2$ PSD from previous step.
2. Melt cools below FCC liquidus temp.
3. **Free growth** of FCC grains allowed to occur on the largest inoculant particles which satisfy criterion d=2r* (r*=critical radius for nucleation).
4. As undercooling increases, r* decreases, and smaller $L1_2$ particles are activated as nucleants.
5. Growing FCC grains release latent heat, slowing the rate of cooling
6. Eventually the temperature rises (**recalescence**) – no more initiation of free growth, controlling the refinement of grain size.

Output of FCC Grain Inoculation Model

Predicted 7xxx Grain Size Contours with Varying Cooling Rate and Oxygen Content

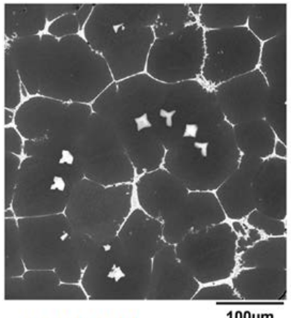


MAGMA Simulation for Component castings

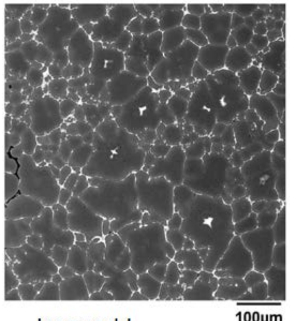
Cooling rates explored in wedge casting trials.

Inoculation model predictions.

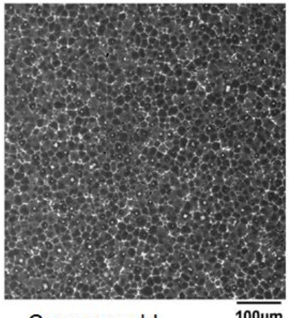
Mould	G.S (µm) (Meas.)	G.S (µm) (Pred.)
Sand	73	65
Iron	54	59.5
Copper	17	22.2



Sand mold
GS - 73 microns
Globular



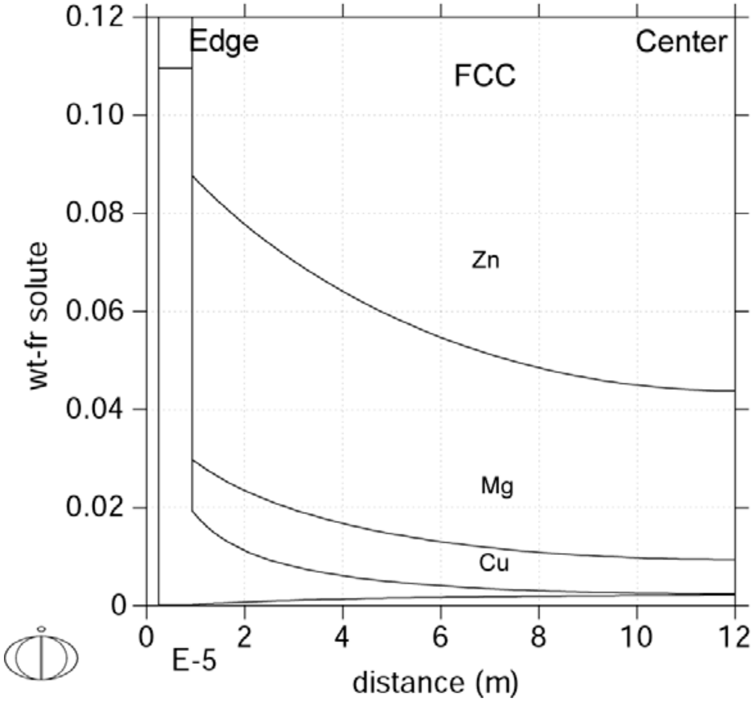
Iron mold
GS - 54 microns
Equiaxed dendritic



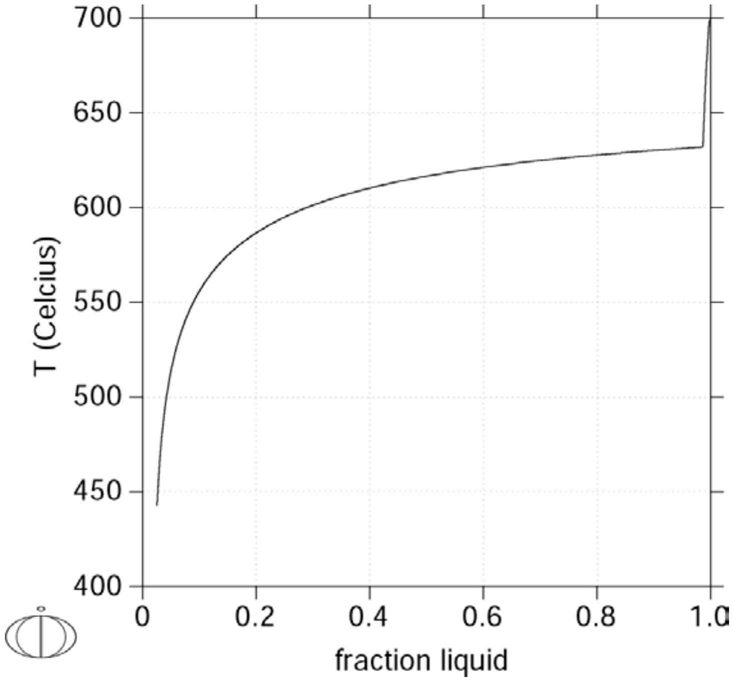
Copper mold
GS - 17 microns
Globular

Solidification: DICTRA simulations

- **Segregation within the grains can be modeled using DICTRA solidification simulations**
- **This enables us to design the subsequent homogenization/solution heat-treatment to eliminate all microsegregation**



Solute Segregation



Solidification Curve

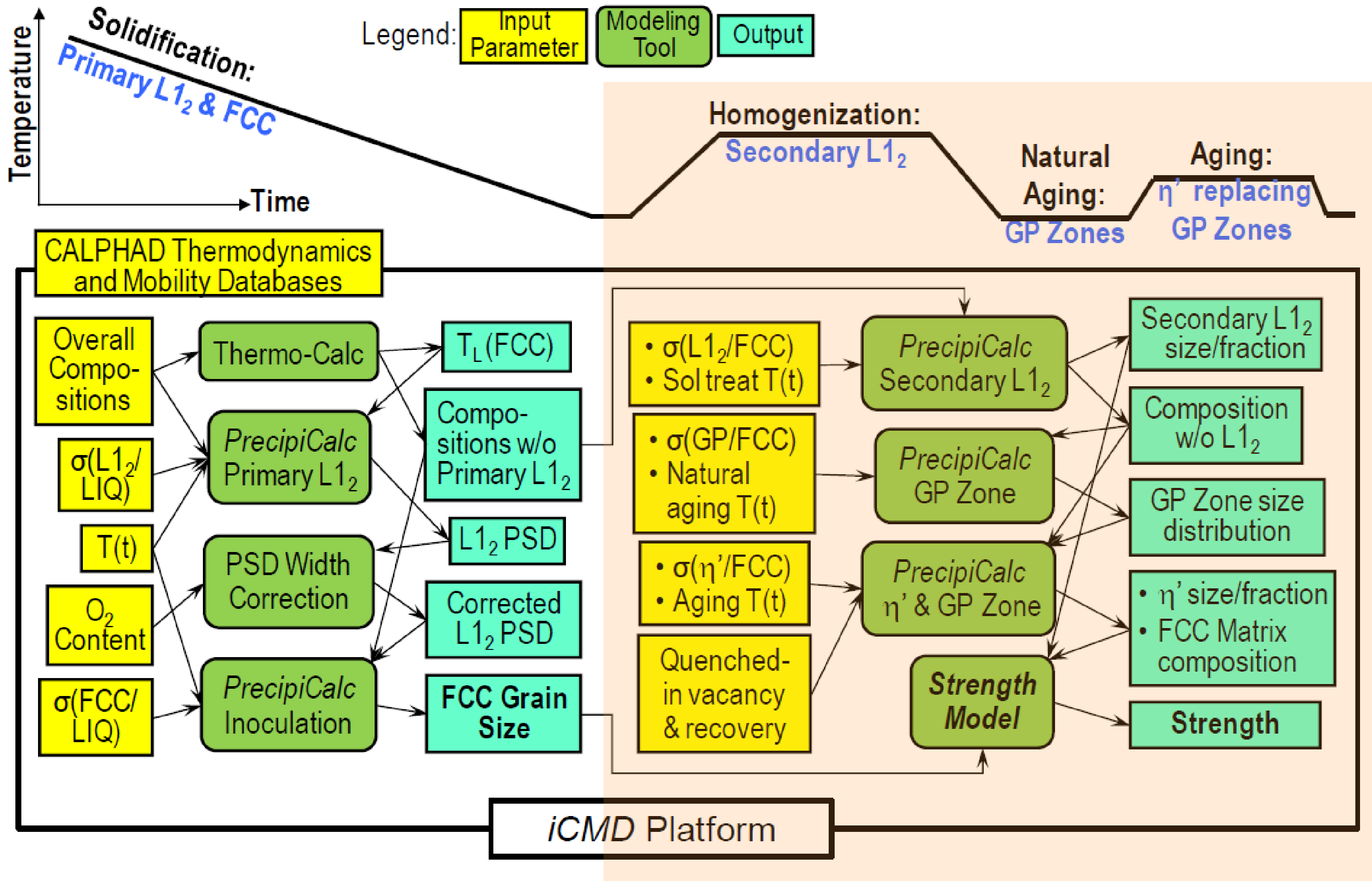
Precipitation and Strength Model -

•Low-temperature aging process of Al alloys – metastable phase variants are the primary strengthening phases

• For 7xxx-series alloys, typical precipitation sequence is:

FCC (supersaturated) → GP zones → η' (T6) → η (T7)

Strength Model Flow Chart

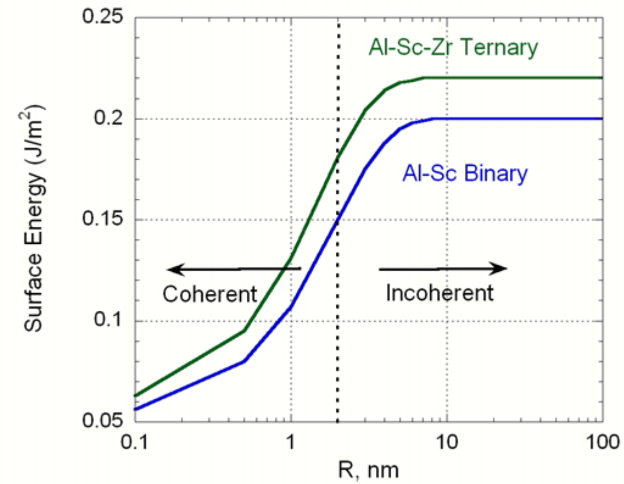


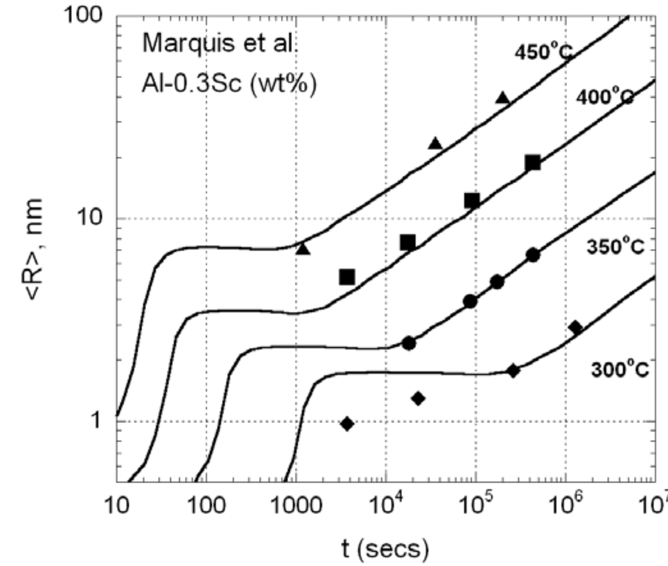
Modeling Secondary $L1_2$ Precipitation

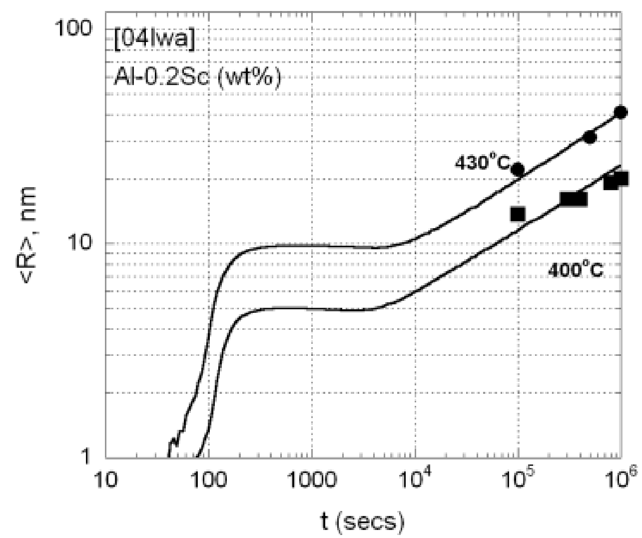
Size dependant interfacial energy function for $L1_2$ particles

For Al-Sc Binary : $\gamma(\mathrm{J/m^2}) = 0.05 \times [3 \times \tanh(4e8 \times R) + 1]$

For Al-Sc-Zr ternary : $\gamma(\mathrm{J/m^2}) = 0.055 \times [3 \times \tanh(5e8 \times R) + 1]$

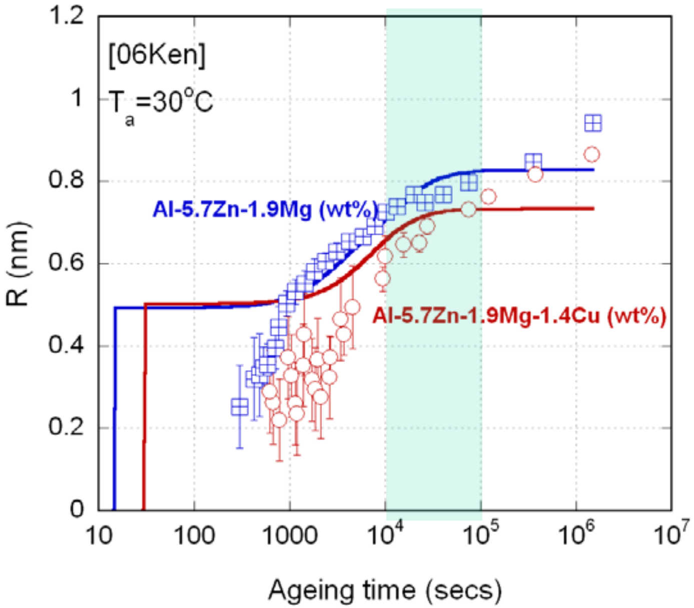


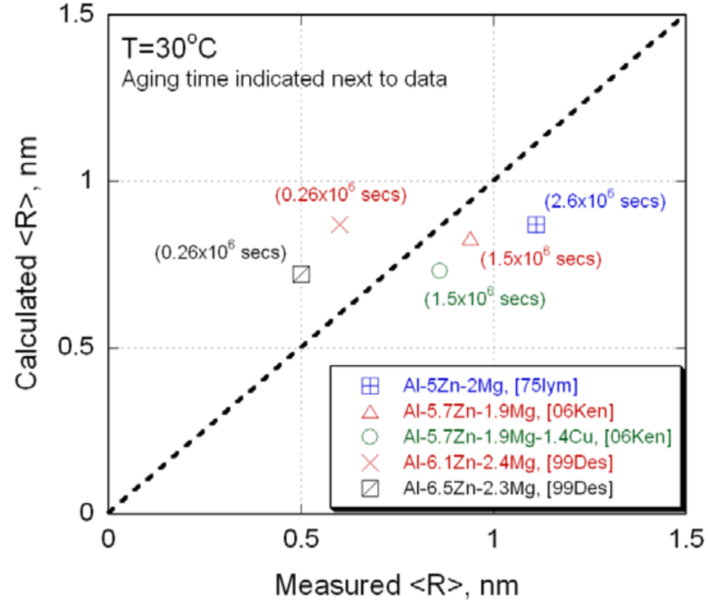




- PrecipiCalc model was calibrated to existing literature data for secondary $L1_2$ precipitation
- Solid lines: PrecipiCalc Simulations; Data points: Experimental literature data.
- Accurately reproduced **in region of practical interest**

Modeling GP zone Precipitation





- GP zones precipitate homogenously at RT – act as nucleating sites for subsequent η′ formation
- Role of quenched-in vacancies is extremely important – used as a multiplication factor to diffusivity matrix
- Accurately reproduced **<u>in region of practical interest</u>**

Quenched-in vacancies

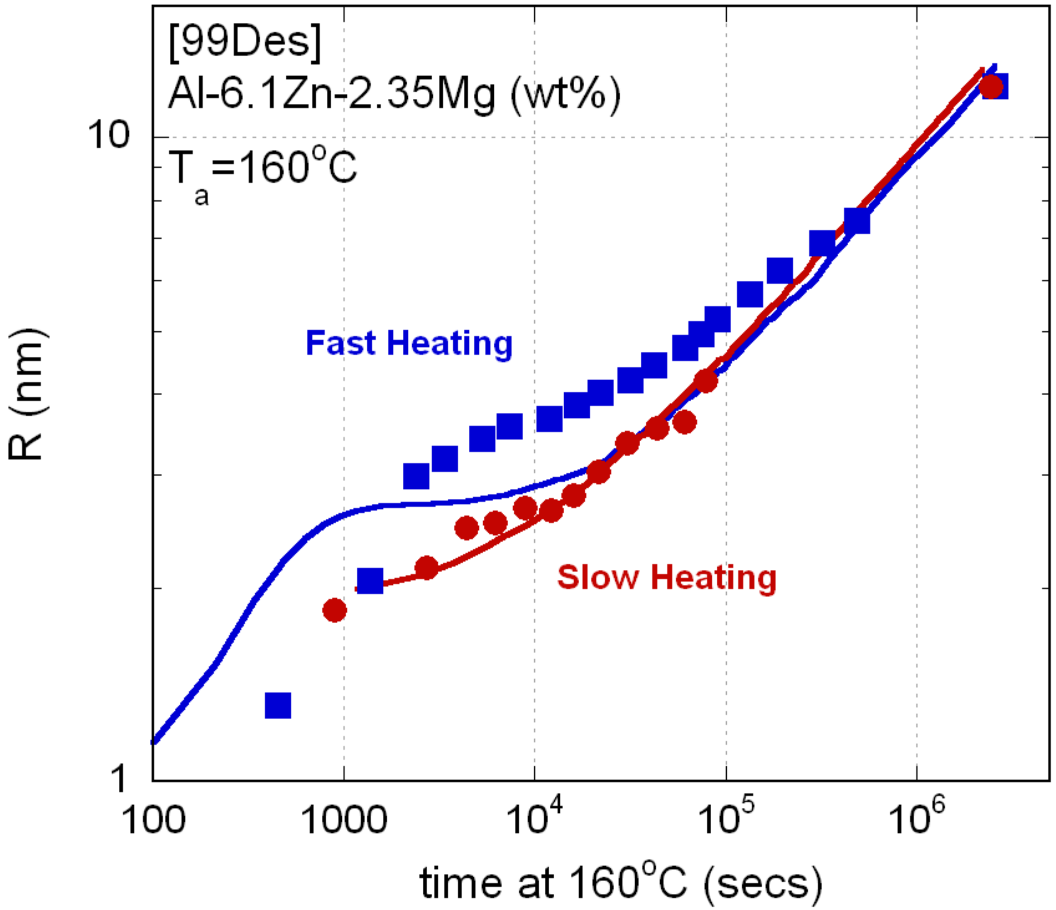
$$S = \frac{C_V^{Tq}}{C_V^{298}} = \exp\left[\frac{-E_f}{k_B}\left(\frac{1}{T_q} - \frac{1}{298}\right)\right]$$

Annealing of vacancies

$$\frac{C_V^{Excell}}{C_V^{Equilibrium}} = 1 + S\exp\left(-K_1 t\right)$$

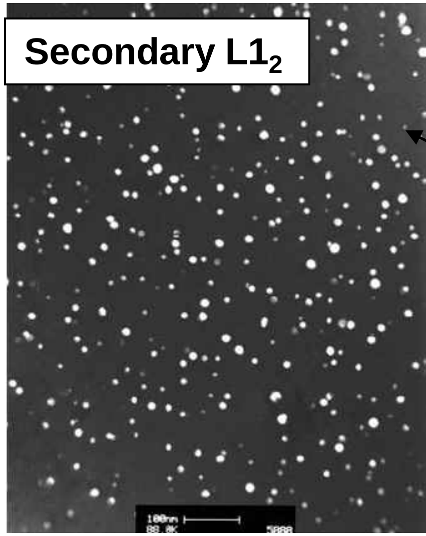
$$K_1 \approx \rho \upsilon \lambda^2 \exp\left(\frac{-E_m}{k_B T}\right)$$

Modeling Eta-prime Precipitation



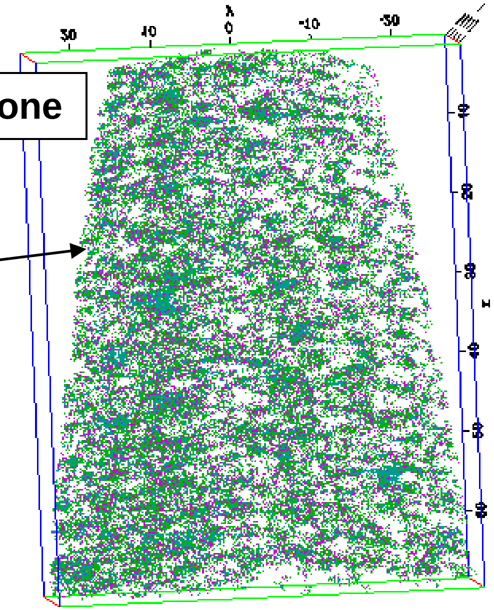
- η′ particles are the primary strengthening particles in 7xxx series Al alloys in the T6 peak-aged condition, and nucleate heterogeneously on existing GP zones upon heating to T>70°C
- Typical T6 treatment temperatures for 7xxx-type alloys are 120-150°C, at which point most GP zones dissolve and the predominant precipitates are η′ particles
- Accurately reproduced **<u>in region of practical interest</u>**

Characterization of Precipitates in a 7xxx Alloy

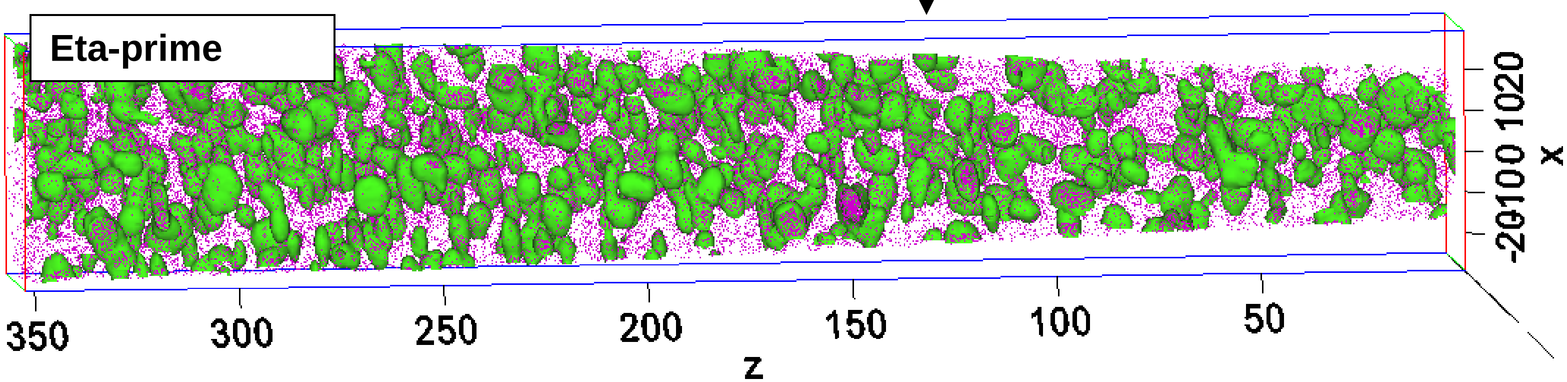


460°C/20hrs
(TEM)

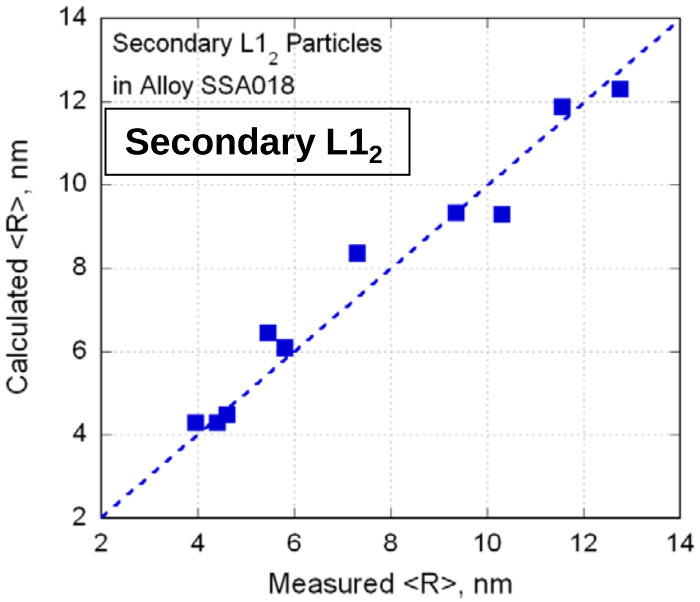
ST+ 30°C/ 2 days
(LEAP)



SHT+ 30°C/ 2 days + 140°C/10hrs
(LEAP)



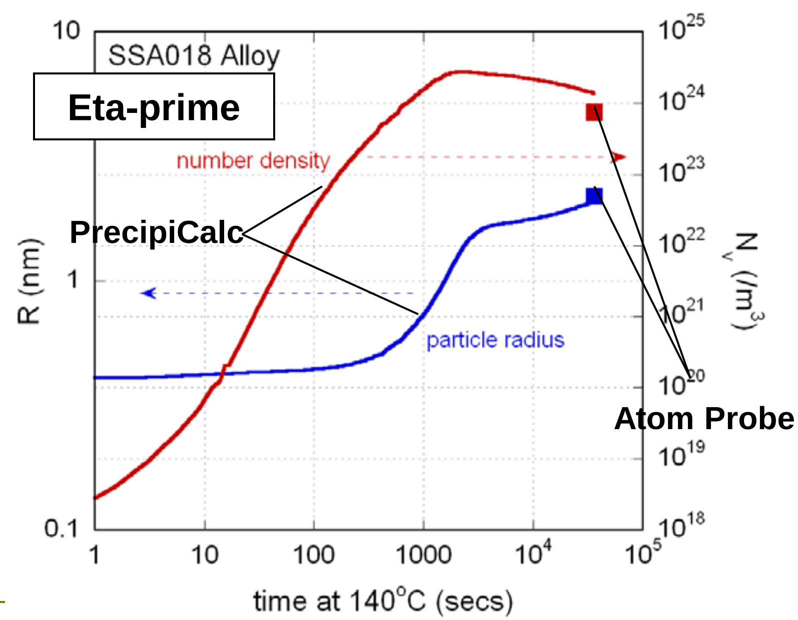
Validation of PrecipiCalc model predictions



GP zone

Analysis	Particle Radius (nm)	Number Density (#/m^3)	Volume Fraction (%)
PrecipiCalc: (Reported SSA018 composition [05Sen])	0.72	3.6e25	7%
LEAP: 1st Experiment	0.56	2.6e25	4.6%
PrecipiCalc: (Composition from 1st LEAP experiment)	0.71	2.7e25	4.8%
LEAP: 2nd Experiment	0.58	2.5e25	2.6%
PrecipiCalc: (Composition from 2nd LEAP experiment)	0.71	1.1e25	1.8%

- PrecipiCalc model predictions show good agreement with experimentally measured data for a 7xxx-series alloy



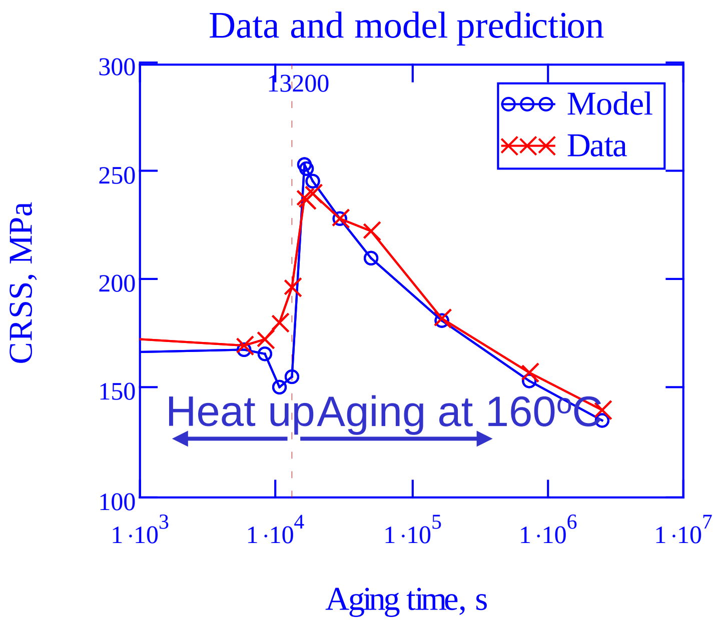
Strength Modeling: Calibration

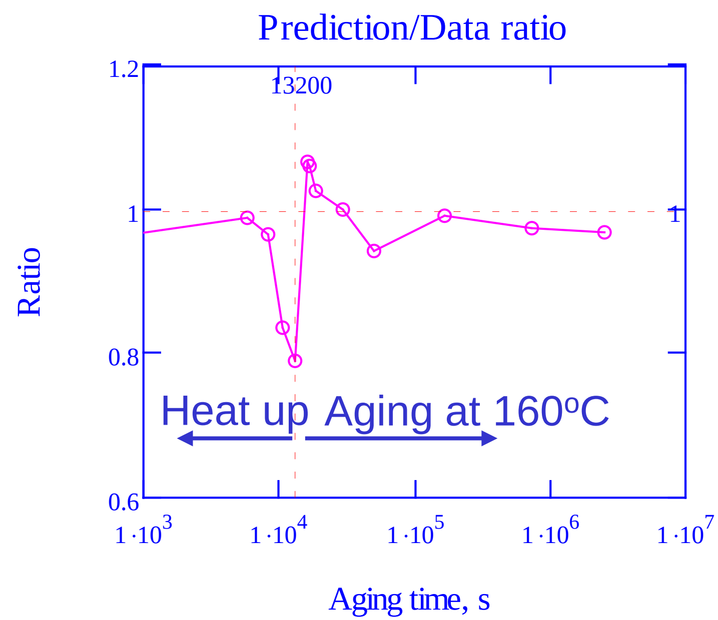
- Mechanistic precipitation strength model for Al-alloys developed at QuesTek, based on:
  - Orowan by-pass and Friedel shear-cutting models.
  - Solid solution strengthening: atomic size and elastic modulus mismatch.
  - Dislocation strengthening: based on Nes's dislocation recovery model.
  - Hall-Petch grain size effect.
- Strength model was first calibrated to literature data.
- Developed model was used to predict the strength of aged SSA018 alloy, at different thermal treatment conditions.

$$\sigma_y = M\left(\tau_0 + \tau_d + \tau_s + \tau_p\right)$$

$$\tau_p = \left(\sum_i \left(\tau_{p,i}\right)^K\right)^{\frac{1}{K}}$$

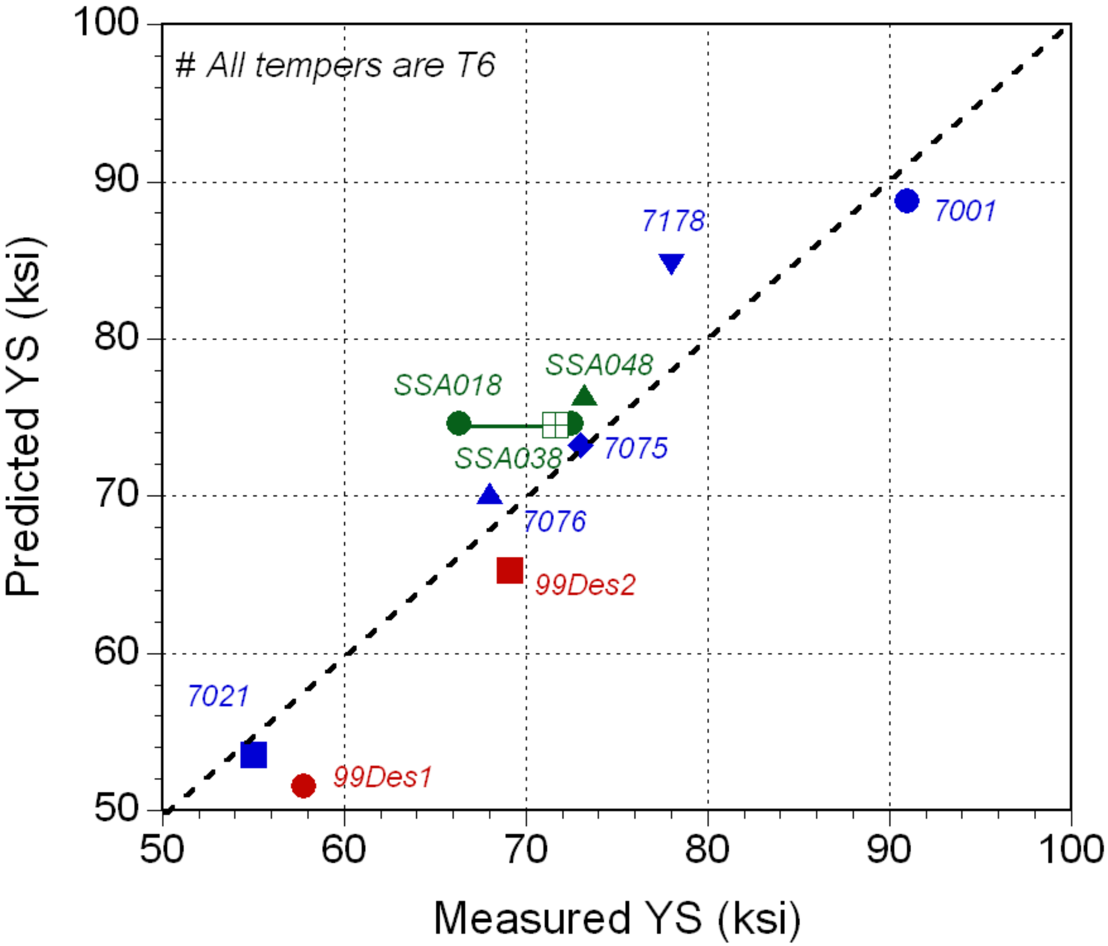
$$\tau_d(t) = \alpha\mu b\sqrt{\rho(t) + \frac{k_I\theta}{b\delta(t)}}$$





Experimental data from *A. Deschamps et al, Acta Mat., vol. 47, 1999, pg. 293-305*

Comparison of Measured Yield Strength Values with Predictions



Most predictions are within ±5 ksi.

Explanation of T6 temper:

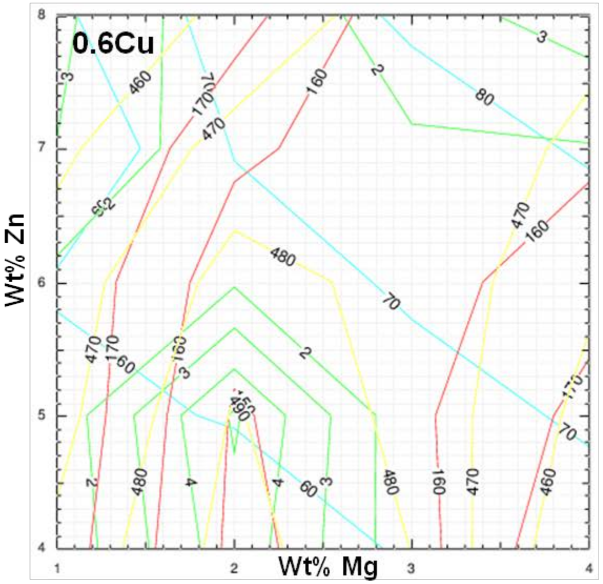
- For SSA0xx alloys: 460°C/48hrs+RT/2days+120°C/19hrs
- For 99Des alloys: 475°C/1hr+RT/3days+160°C/1-5hrs
- For wrought 7xxx alloys: 465°C/1hr+RT/1day(assumed) + 120°C/24hrs

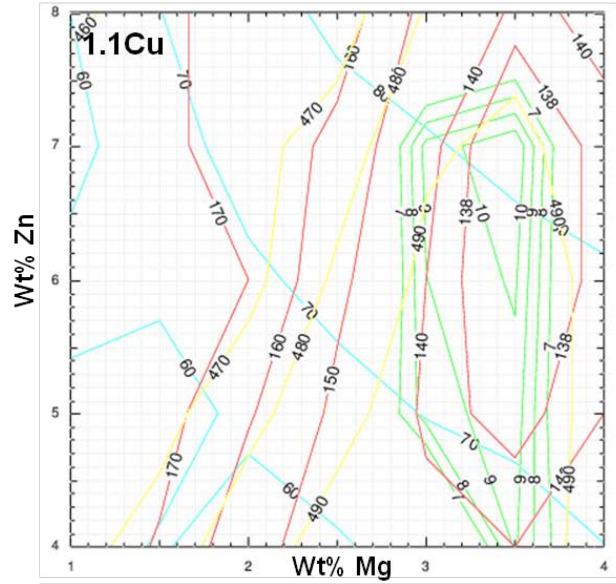
Data Source

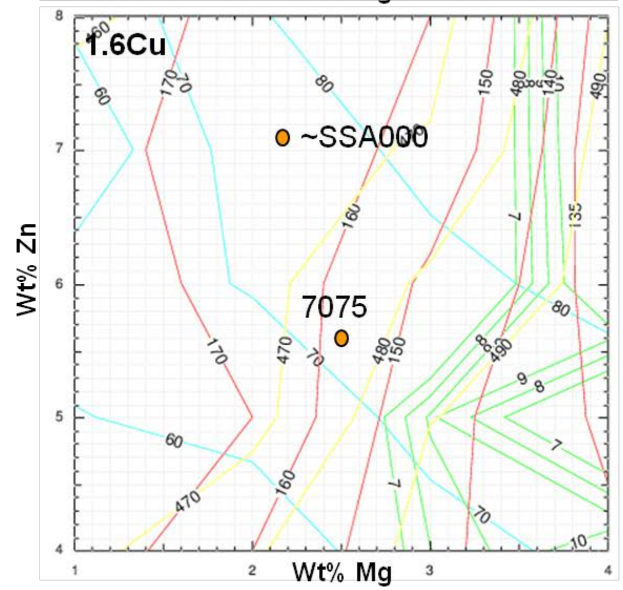
SSA0xx: O.N. Senkov et al., Met. & Mat. Trans. A, vol. 36A, Aug 2005, pp. 2115-2126.
99Des: A. Deschamps et al., Mat. Sc. & Tech., vol. 15, Sep. 1999, pp. 993-1000.
7xxx: ASM Metals Handbook, Desk Edition, 1995

Design Optimization – Design of high strength 7xxx-series castable alloys by using the developed models

Strength-Solidification Maps with Varying Zn, Mg, and Cu







Constraints:

- Strength
- Freezing range
- Scheil solidus temperature
- % Eutectic

Blue lines: Yield strength iso-contours (ksi)

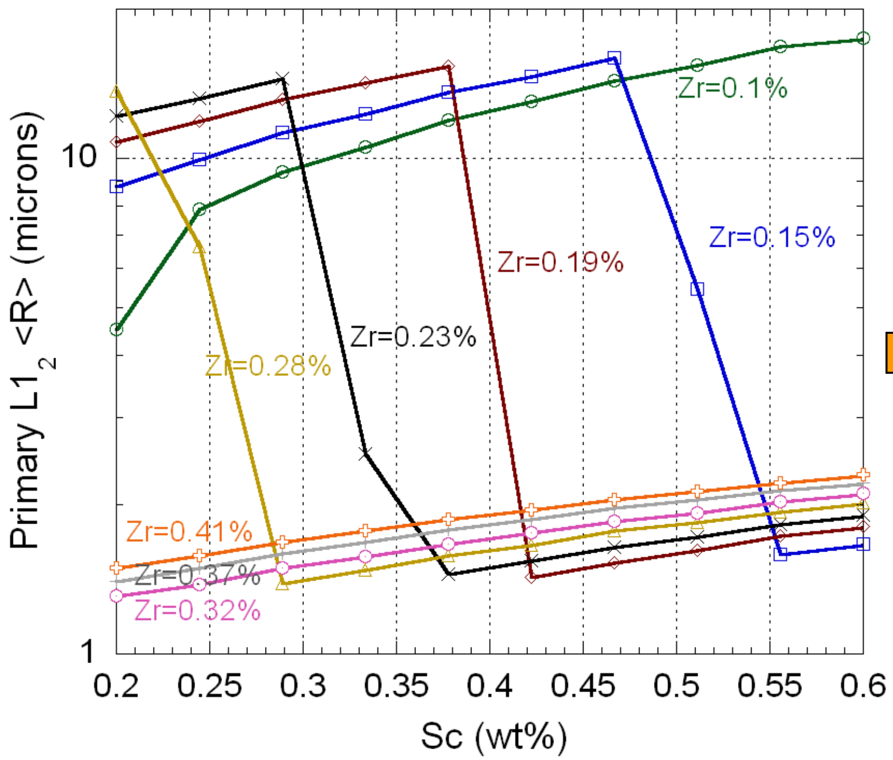
Green lines: % Eutectic (Scheil)

Red lines: Scheil Freezing range (°C)

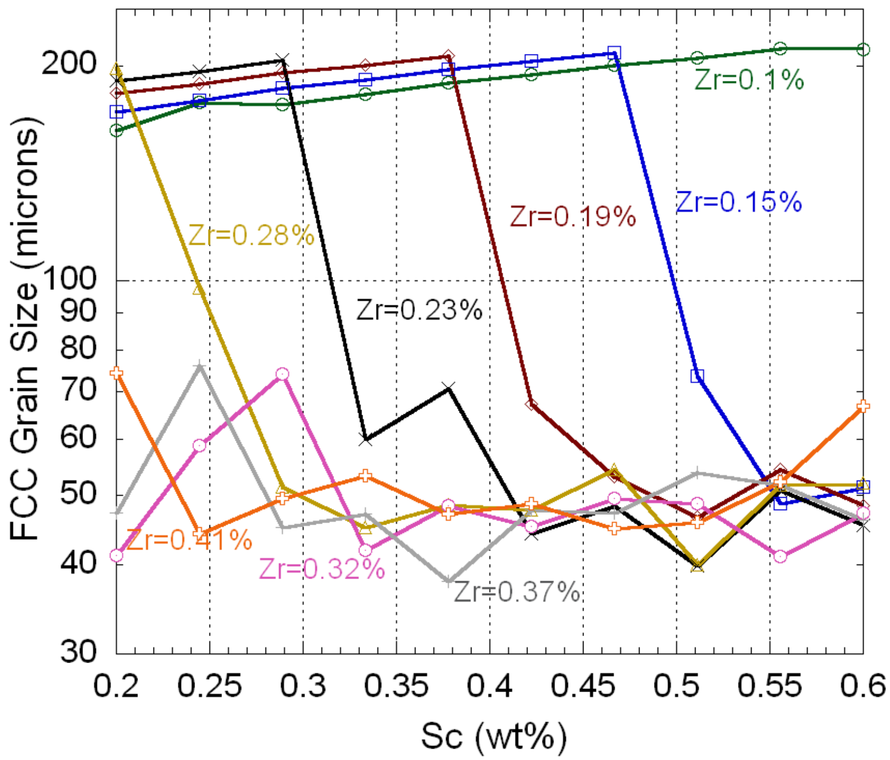
Yellow lines: Scheil solidification Temperature (°C)

Simulation of Casting Process: Primary $L1_2$ <R> and FCC Grain Size

Primary $L1_2$



FCC Grain Size



Assumed 80 ppm O

Results

Alloys were designed to achieve a cast FCC grain-size of ~50μm (for optimal hot-tearing resistance), and yield strength levels of 70ksi and 78ksi.

Alloy	Grain size (microns)		YS (ksi)	
	Predicted	Measured	Predicted	Measured
M1	54	48.5	72	70
H1	48	45	78	76

Alloys showed good resistance to hot-tearing in dog-bone testing due to in-situ inoculation and solidification optimization

Summary

- Alloy design involves number of complex structures and processes
  - We have to account for all of these different structures/processes
- Multicomponent diffusion plays a key role in almost all processing steps
  - Without accurate databases designs will be empirical and limited – impossible to design for complex industrial processes
- The required fidelity of the databases is driven largely by the design needs
  - We need to be able to accurately model the “region of interest”
- Top-down approach
  - Design framework drives model/database development