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Macro-Segregation in Uranium-6wt%Niobium

Robert M. Aikin Jr.

Presented by John W. Gibbs

**Los Alamos National Laboratory
Materials Science & Technology: Metallurgy (MST-6)**

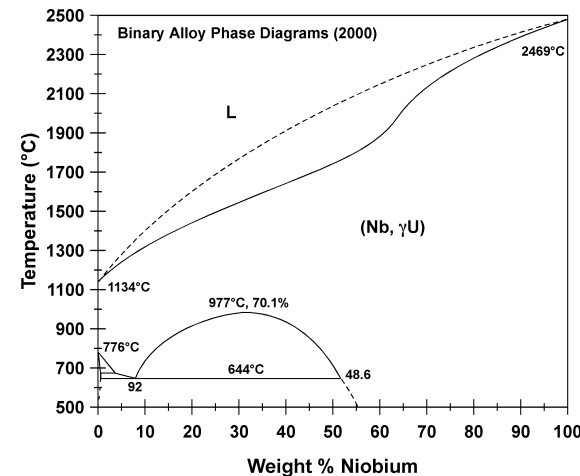
2015 TMS Annual Meeting

March 17, 2015



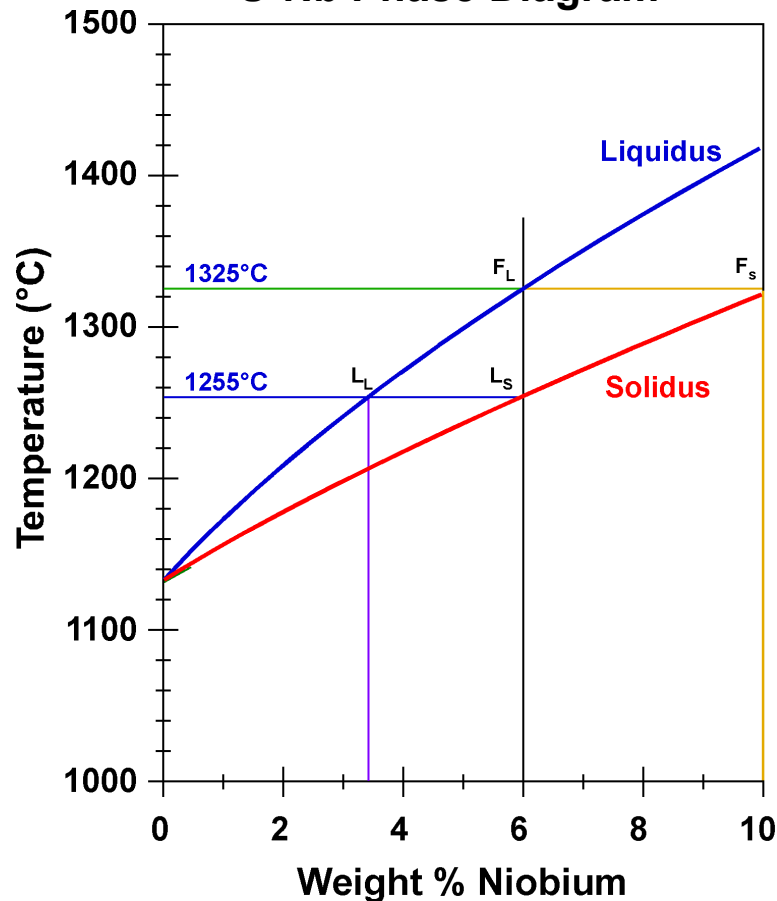
Motivation

- Historically U-Nb alloys are produced by vacuum arc-remelting (VAR) and subsequent wrought processing, but it would be convenient to produce shape castings by vacuum induction melting (VIM)
- Chemical macro-segregation of the Nb and micro-porosity remains a issue for castings produced by VIM
- This study aims to better understand macro-segregation in cast U-6wt%Nb
 - Influence of solidification time
 - Influence of casting thickness
 - Influence of solidification against and with gravity
- Understanding of solidification behavior is complicated by the fact that allotropic transformations wipe out solidification microstructure



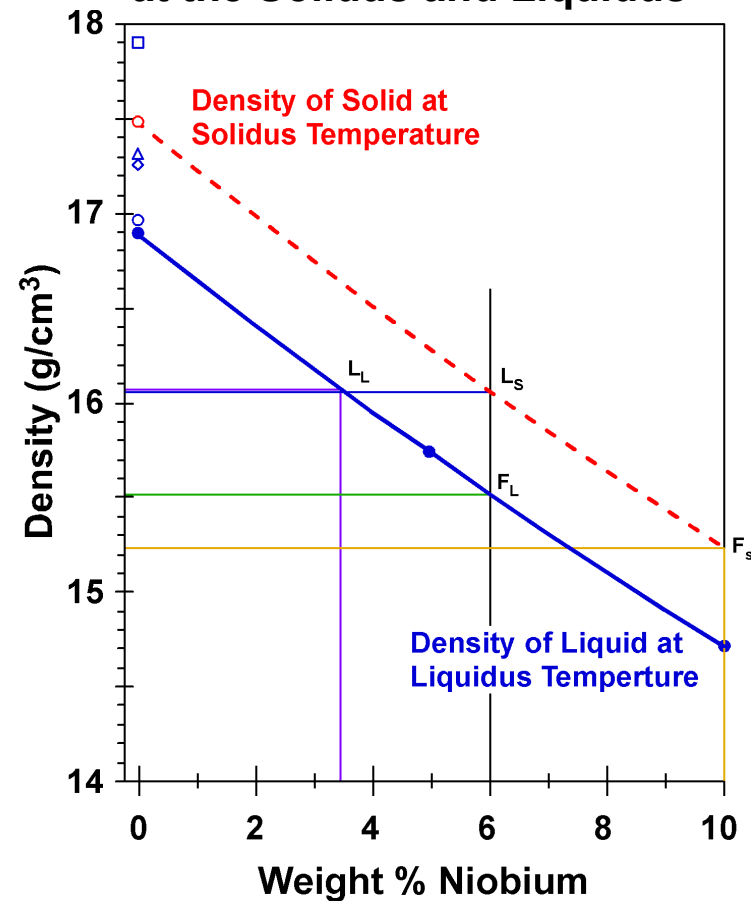
Density Changes During Solidification

Uranium Rich Portion of
U-Nb Phase Diagram



Phase diagram after:
B.A. Rogers, et al, Trans. Metall. Soc. AIME (1958)
D. J. Thoma, unpublished data (2004)

Density of Solid and Liquid
at the Solidus and Liquidus



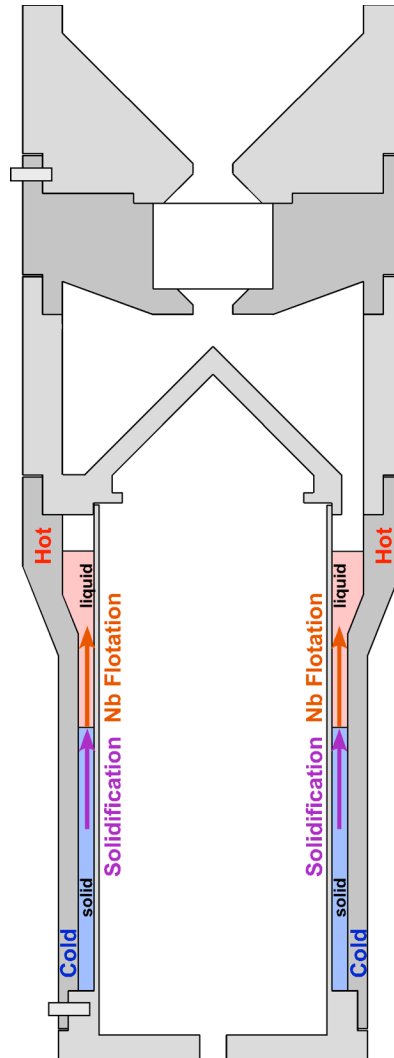
- Liquid density from W.D. Drotning, *High Temp. High Press.*, 14, 253-258 (1982)
- Solid density estimated from published pure U density and Drotning's U-Nb composition dependence of liquid

Growth Direction and Segregation

Assume Macro-Segregation by Buoyancy Driven Flow

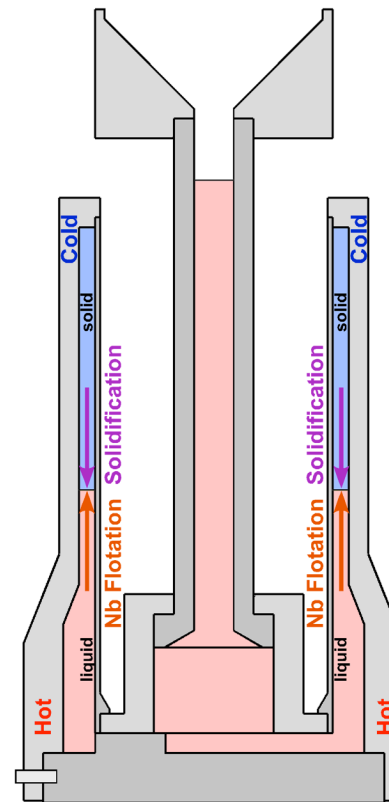
Conventional Mold Design

- Top-to-bottom fill
- Bottom-to-top solidification
- **Solidification front 'chases' Nb flotation**

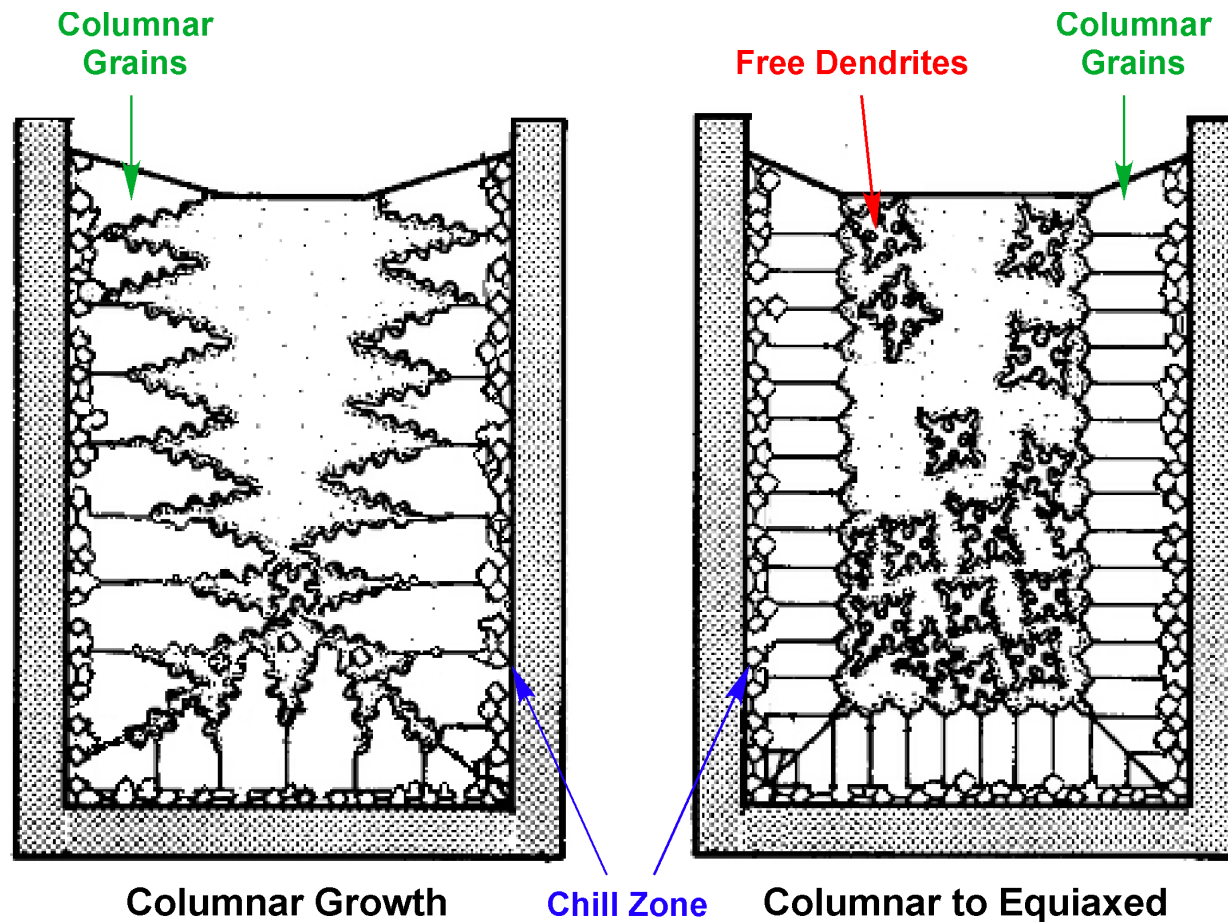


Solidification-Down Mold Design

- Bottom-to-top fill
- Top-to-bottom solidification
- **Solidification direction is opposite Nb flotation**
- Need to keep down-sprue filled and liquid to maintain pressure on casting and feeder



Columnar vs. Free-Dendritic Growth

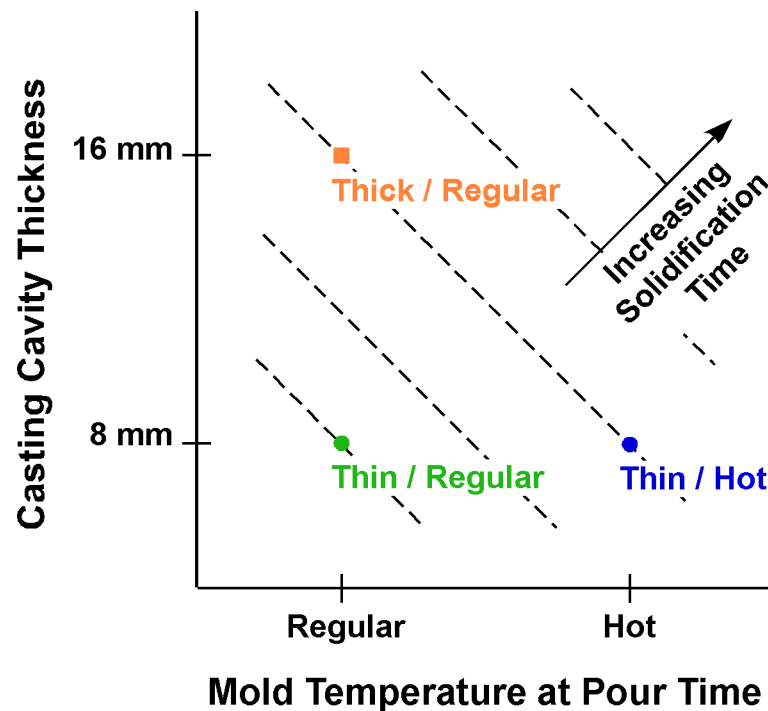


after W. Kurz and D.J. Fisher, Fundamentals of Solidification, Trans Tech Pub (1984).

Experiment Design

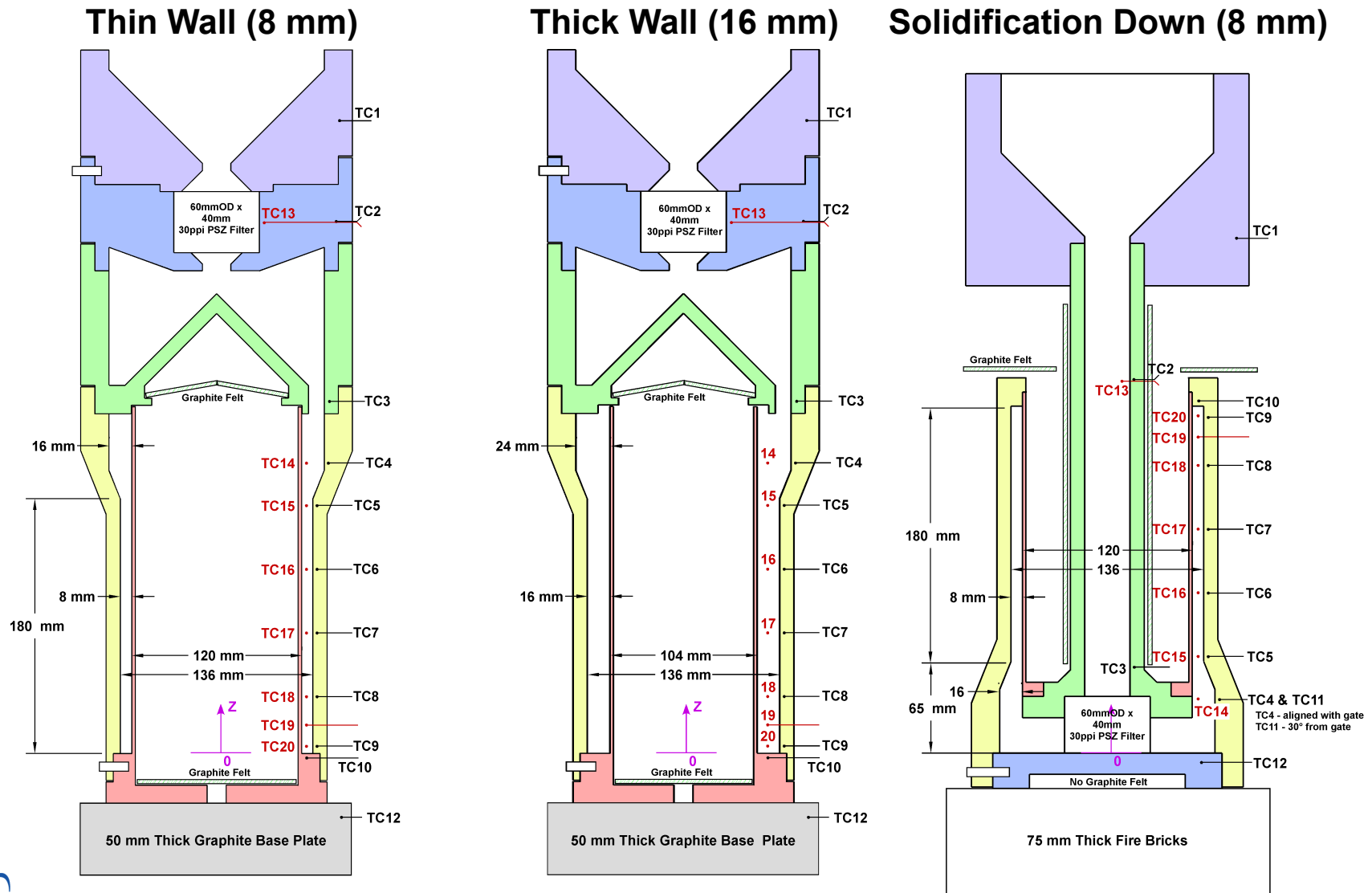
Look at three aspects of macro-segregation:

- 1) Influence of solidification time
- 2) Influence of casting thickness
- 3) Solidification-up vs. solidification-down



“Hot temperature” was determined by process modeling to try to give same solidification time as the Thin/Regular

Comparison of Molds for Cylindrical Castings



Castings Details

- Mold made from fine-grained isotropically pressed graphite (Stackpole 2020)
- Mold coated with Yttria with a cellulose binder (Type-Y from ZYP Coatings)
- Stainless-steel sheathed Type K thermocouples inserted in holes in mold
- Alumina sheathed Type C thermocouples cemented in though-holes in mold wall; bare TC junction in mold cavity
- Wrought U-6Nb plate (scrap) used as charge material
- Same metal pouring temperature used for all 4 castings (1415°C / 60°C superheat)
- Metal filtered though a 30 ppi partially-stabilized zirconia foam filter
- Mold charge and yield

Name	Charge wt (kg)	Casting wt (kg)	Casting Yield
Thick / Regular	28.07	25.28	0.90
Thin / Hot	15.02	13.47	0.90
Thin / Regular	15.13	13.36	0.88
Thin / Top Down	28.01	20.26	0.72

Mold Stack – Solidification Up Mold

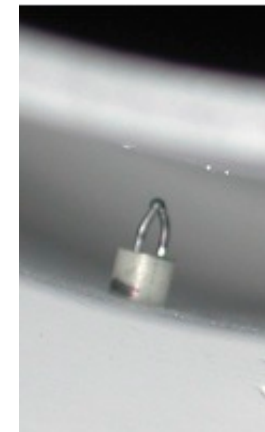
Mold stack with thermocouples



Mold stack with 12" diameter induction coils for mold heating and insulation



Bare junction type-C thermocouples in the mold cavity

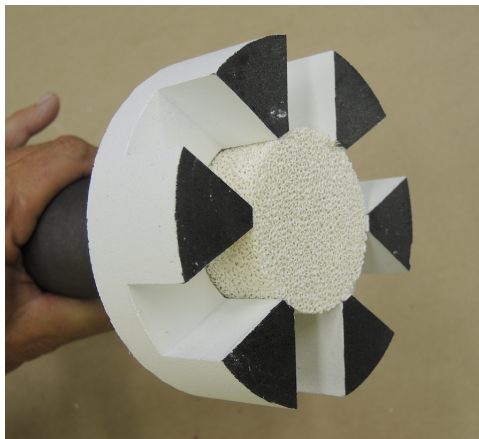


Solidification Down Casting Mold

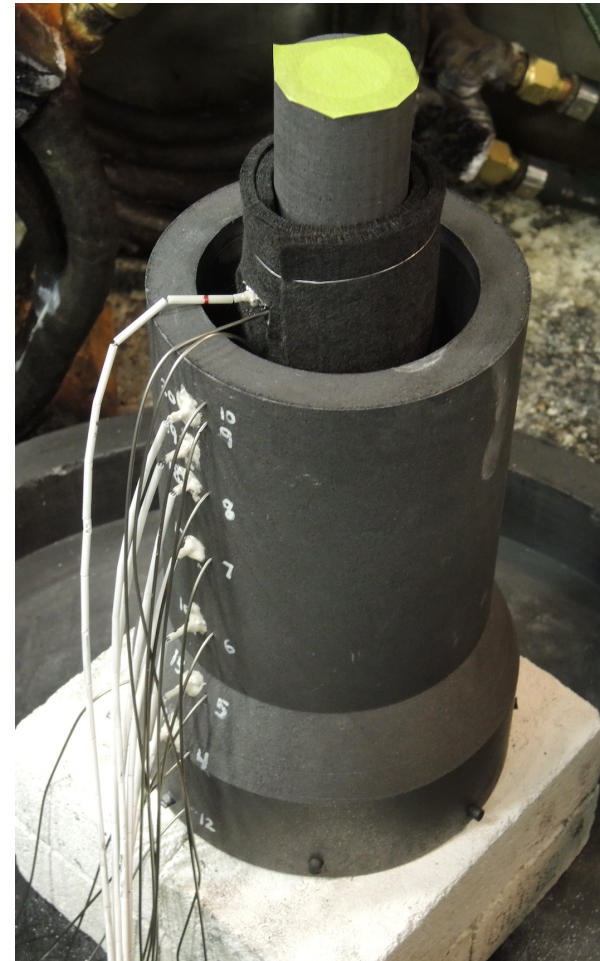
Mold Components Prior to Assembly



Detail of Sprue/Filter Holder with Porous Zirconia Filter



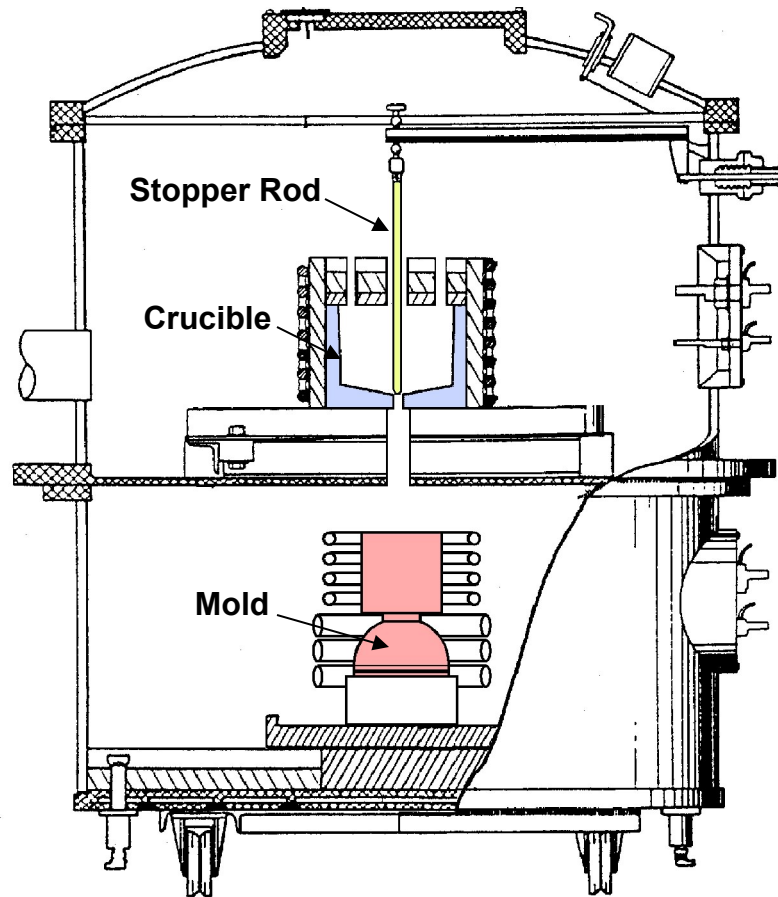
Partially Assembled Mold Showing Thermocouples in Mold and in Casting Cavity



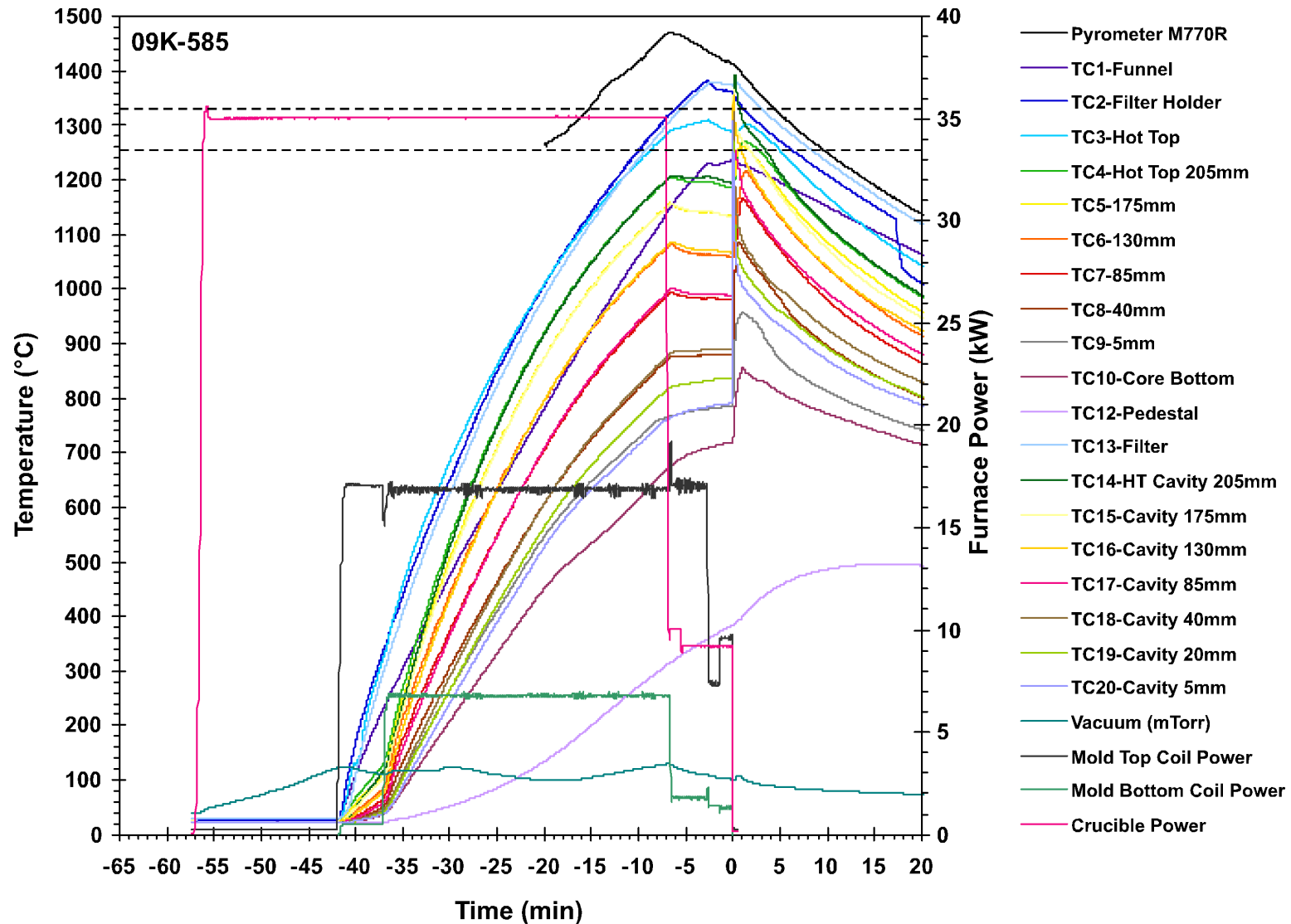
Three Zone Vacuum Induction Furnace

Three separately controlled induction coils:

- Melting crucible coil (35 kW at 9.6 kHz)
- Two mold heating coils (50 kW at 3 kHz)

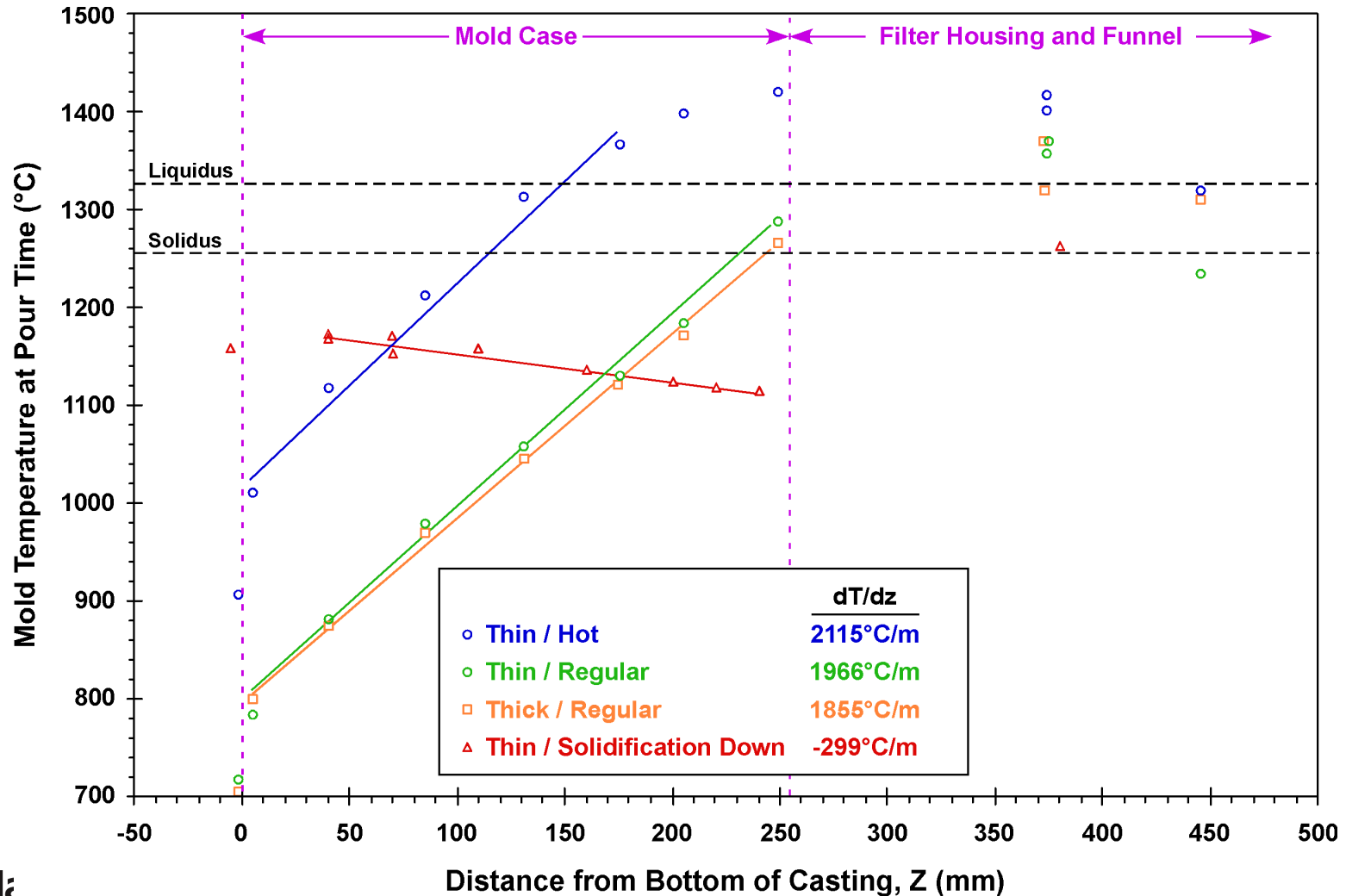


Run Chart - Thin / Regular Mold Temperature



Mold Temperature at Pouring Time

Differing Initial Mold Temperature
Same Metal Pouring Temperature = 1415°C



Cast Parts

**Thick Wall (16 mm)
Casting**



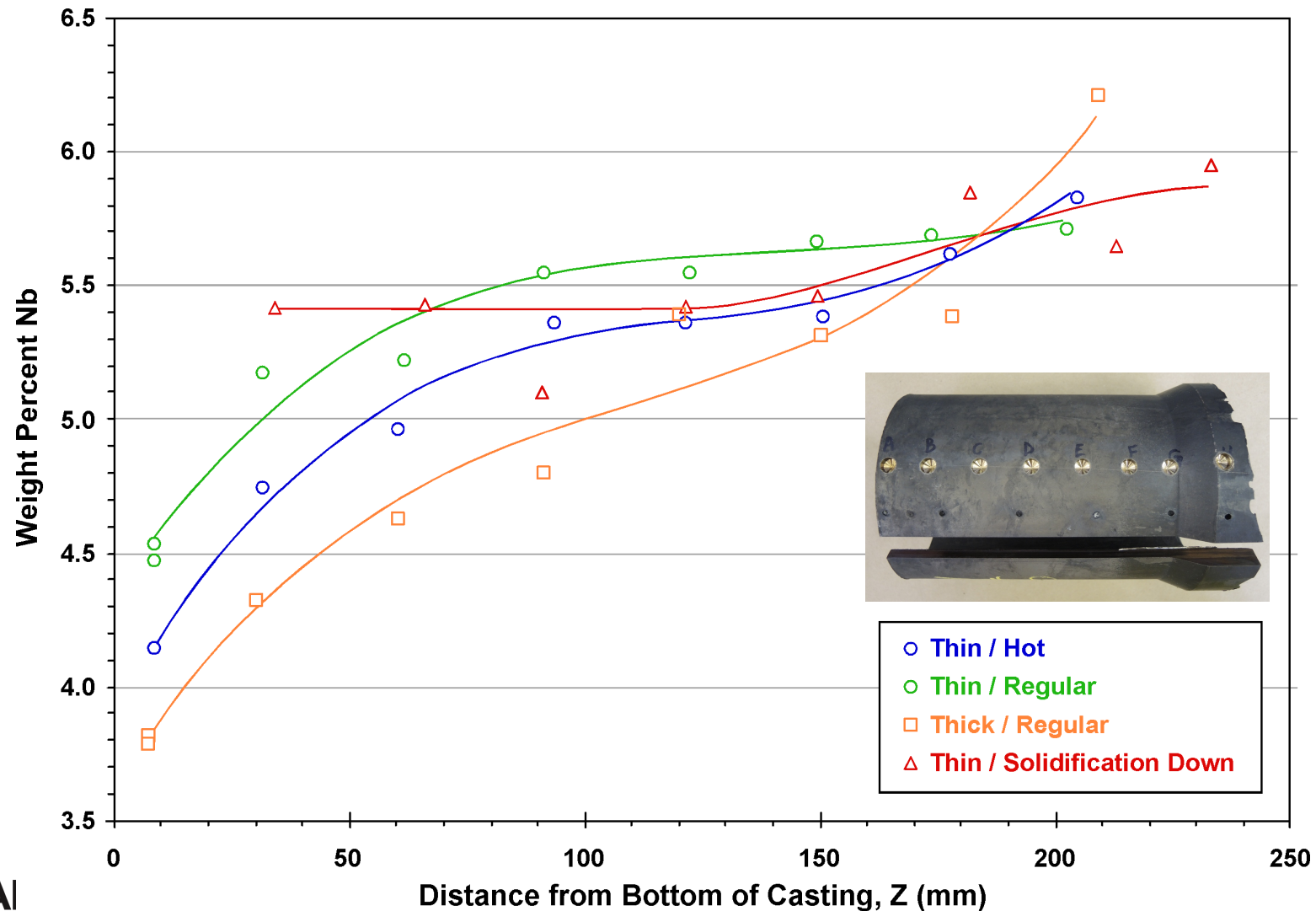
**Casting just after removal from mold.
Thermocouple remnants and residual mold
coating can be seen.**

**Solidification Down (8 mm)
Casting**

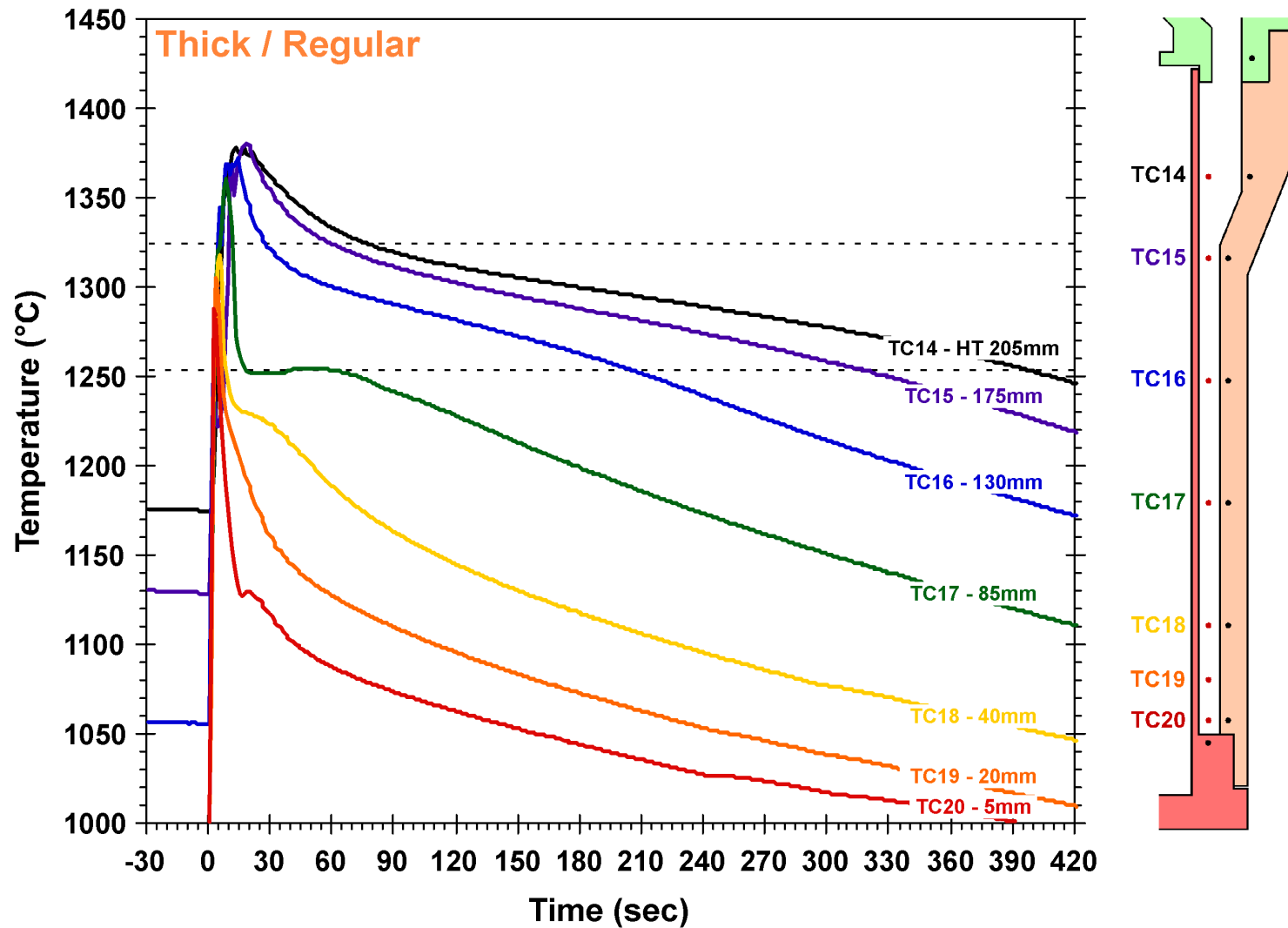


Segregation: Thickness vs. Solidification Time

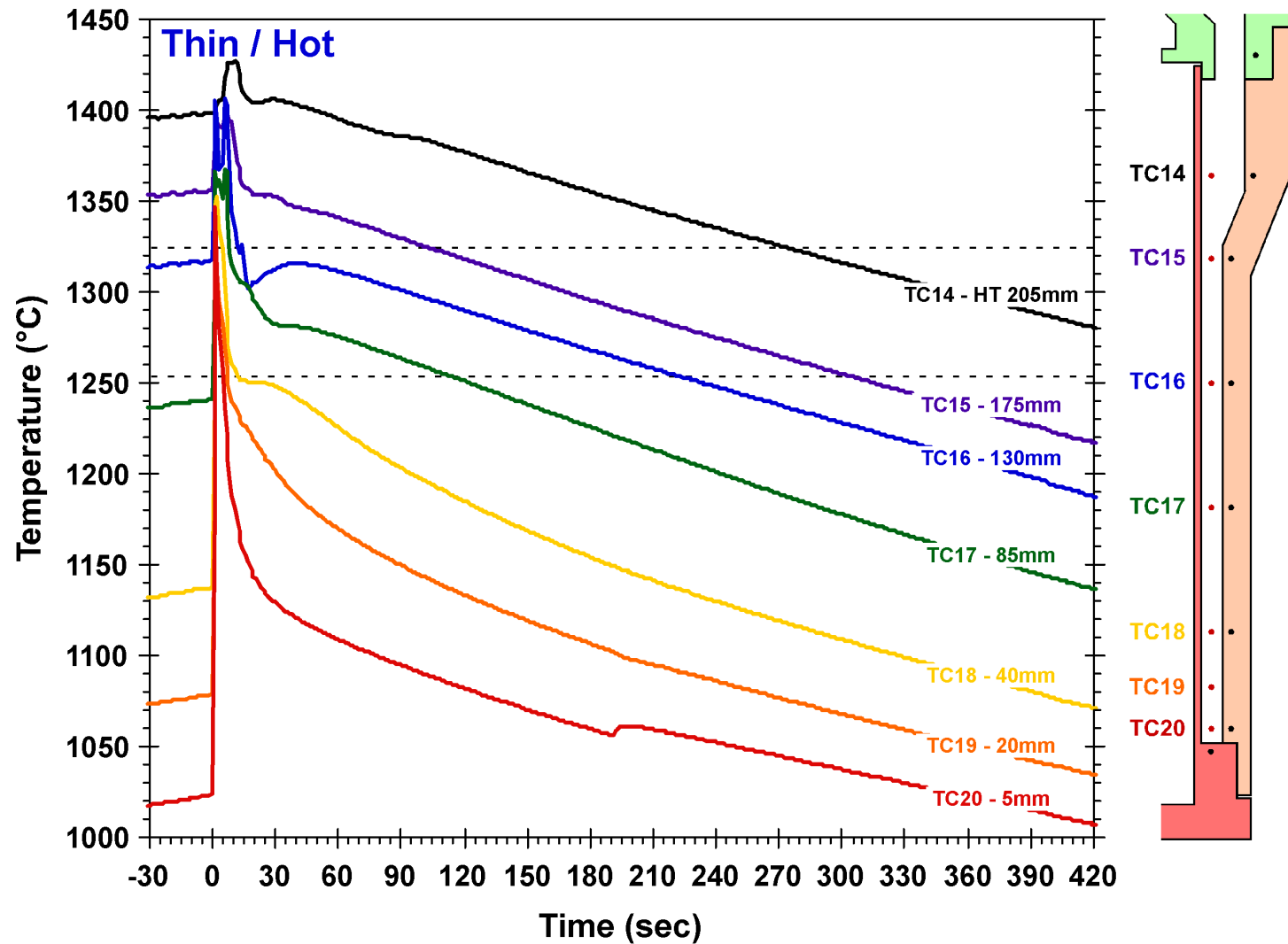
ICP Chemical Analysis Results as Function of Position
Samples from Chips Taken from 9 mm Drill Hole to Casting Center



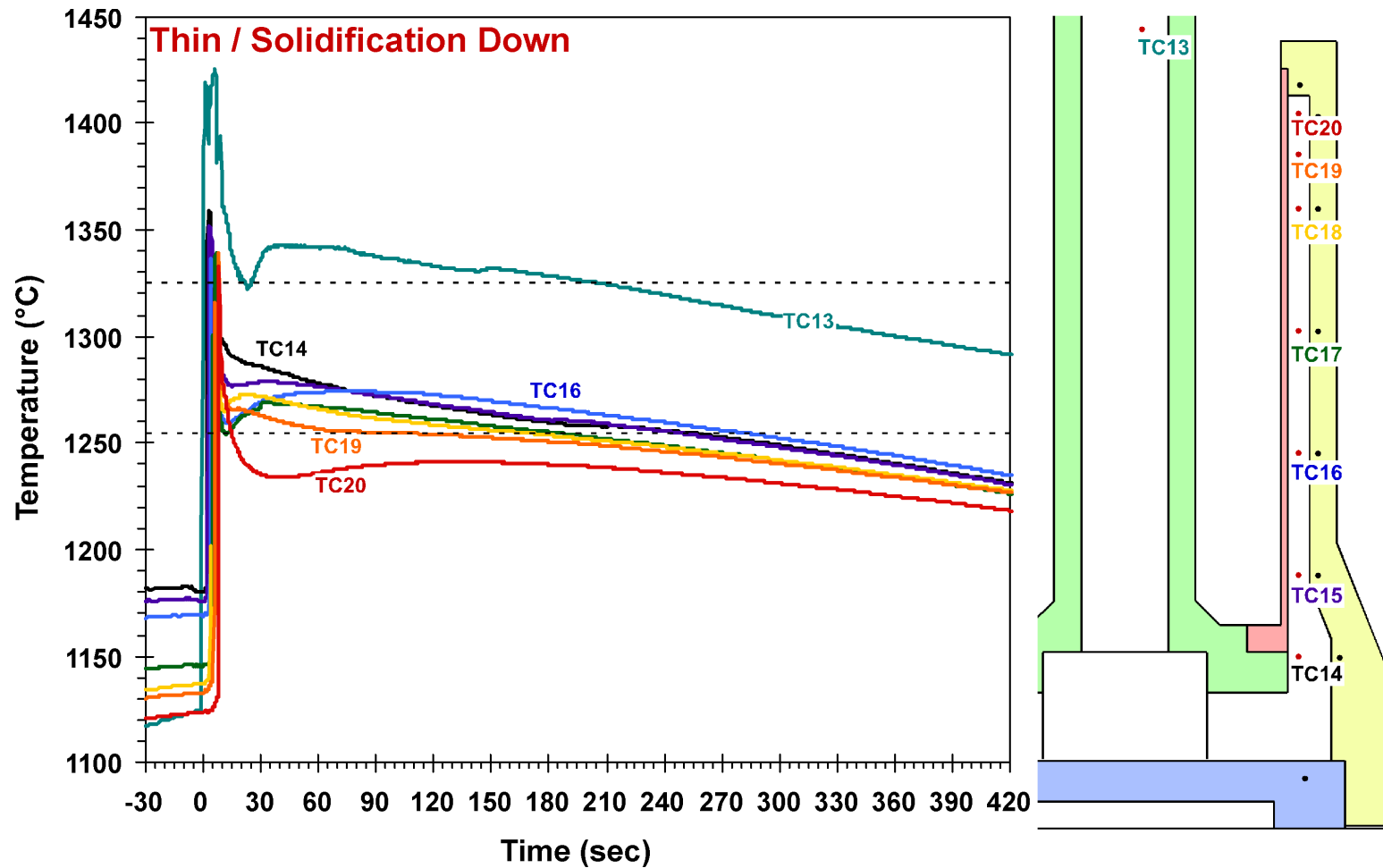
Cooling Curve - Thick / Regular Mold Temperature



Cooling Curve - Thin / Hot Mold Temperature



Cooling Curve - Solidification Down



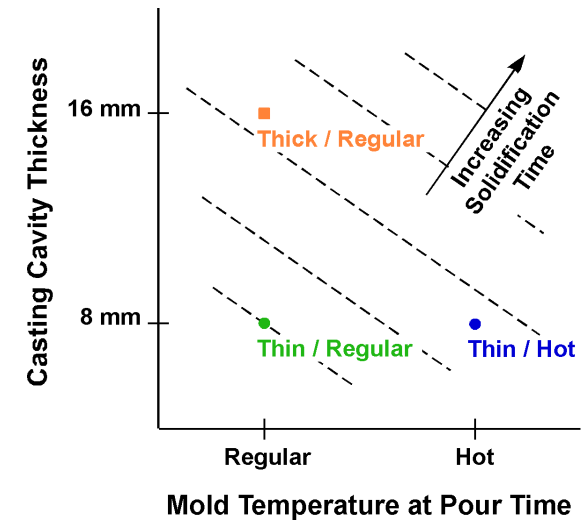
- Mold progressively filled from bottom to top
- Mold *mostly* solidified from top to bottom (TC16 is a hot spot)
- Very shallow thermal gradient; everything mushy at same time

Solidification Time

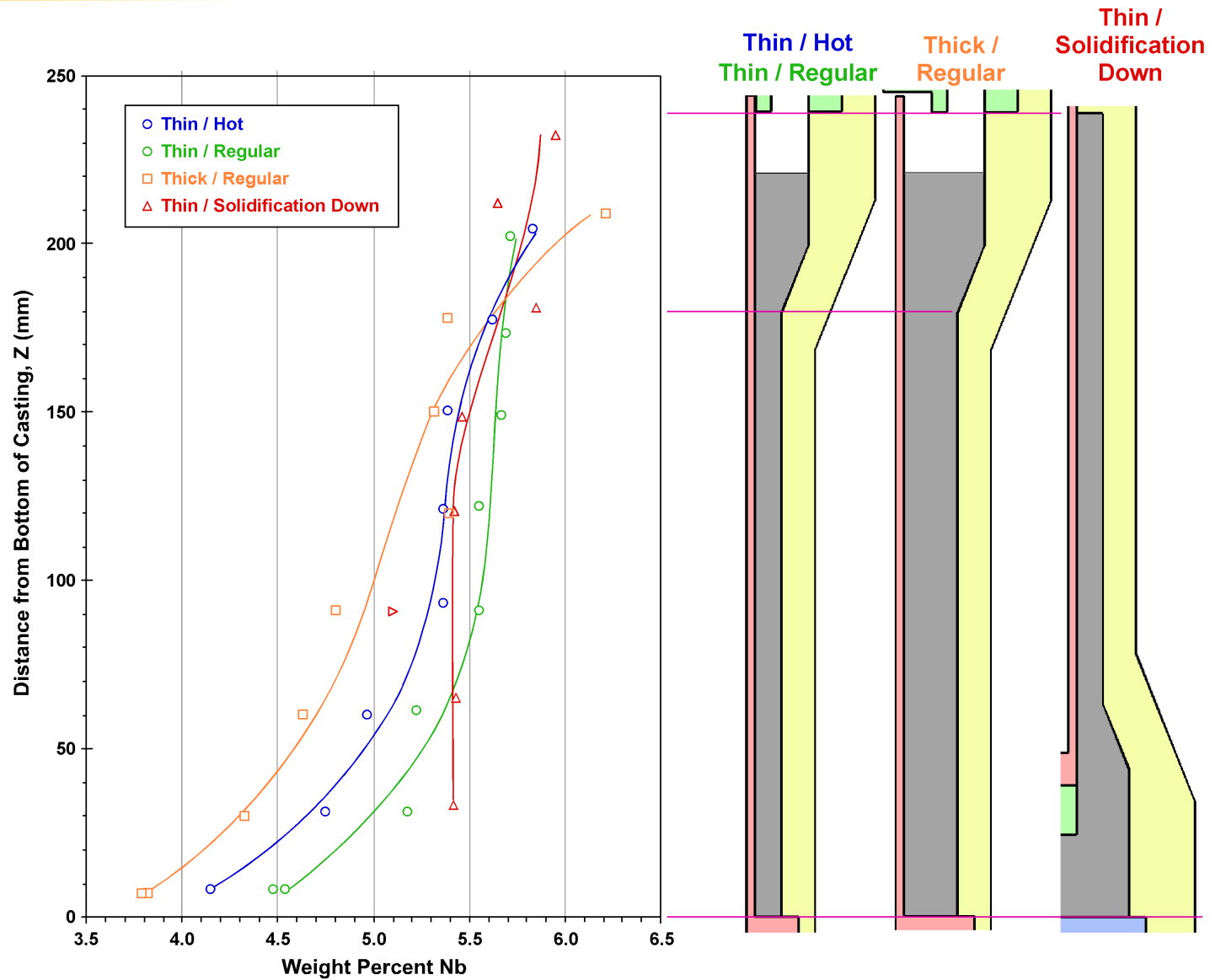
Solidification and Local Solidification Time from Cooling Curves

		Cavity							Down Sprue
Thermocouple ID		TC20	TC19	TC18	TC17	TC16	TC15	TC14	TC13
Distance from Bottom, Z (mm)		5	20	40	85	130	175	205	
Solidification Time (s)	Thin/Hot	5	7	11	109	217	298	506	
	Thin/Regular	3	4	6	14	54	107	206	
	Thick/Regular	4	6	8	59	198	312	389	
Distance from Top (mm)		5	20	40	85	130	175	205	-25
Distance from Bottom, Z (mm)		240	225	205	160	115	70	40	270
Solidification Time (s)	Thin / Top Down	17	74	158	176	275	239	244	365

		Distance from Bottom, Z (mm)	5	20	40	85	130	175	205	
Local Solidification Time (s)	Thin/Hot				7	100	206	211	252	
	Thin/Regular				3	9	49	103	165	
	Thick/Regular					53	174	264	326	
		Distance from Top (mm)	5	20	40	85	130	175	205	-25
		Distance from Bottom, Z (mm)	240	225	205	160	115	70	40	270
Local Solidification Time (s)	Thin / Top Down		8	69	158	168	269	233	239	636



Segregation: Thickness vs. Solidification Time



Conclusions

- **For U-6wt%Nb castings solidified bottom-to-top:**
 - Segregation is most strongly related to solidification time
 - Macro-segregation is most severe in the bottom of the conventional castings; this is the region which also solidifies most rapidly
 - For similar solidification times there does appear to be a thickness effect (for the 8 to 16 mm thickness examined)
- **For casting solidified down (top-to-bottom):**
 - Macro-segregation is mostly eliminated
 - A higher thermal gradient than obtained in this experiment would be beneficial to better cast structure and a better cast part
- **The observed macro-segregation is consistent with free-dendritic growth with flotation (buoyancy driven and/or filling driven advection of liquid?)**