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Title: More On Unalloyed Plutonium Rod Castings, Characterization and Quasistatic Triaxial Tensile State Testing

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MORE ON UNALLOYED PLUTONIUM ROD CASTINGS, CHARACTERIZATION AND QUASISTATIC TRIAXIAL TENSILE STATE TESTING

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Pure plutonium possesses six allotropes between room temperature and melting at 640°C. Deformation modes vary from grain-boundary sliding in the low-symmetry monoclinic alpha and beta phases to dislocation slip in the higher symmetry cubic delta and epsilon phases [1]. Since no alloying additions are currently known that stabilize the intermediate temperature beta and gamma phases to room temperature, mechanical testing at elevated temperatures must be performed on unalloyed plutonium. Recent attempts to characterize the mechanical behavior of the higher temperature phase have shown some significant issues related to material processing, in particular casting.

Castings of high-density, unalloyed plutonium were created as part of a research and development effort at Los Alamos National Laboratory (LANL). Casting unalloyed plutonium is challenging due to large volumetric phase transformations incurred during cooling, particularly the beta to alpha transformation at 117°C which subjects the material to a 9% volume decrease inducing large internal stresses that can result in cracks and voids. A combined experimental/computational approach was used to design casting molds and determine the best process parameters needed to produce quality castings. A rod mold was machined to cast 6 rods, 20mm diameter by 110mm long. The mold and processing parameters were designed to solidify and cool the plutonium quickly to the beta phase. At this point the mold and metal were nearly isothermal and the rods slowly cooled to room temperature. By removing the heat from the system in a slow, controlled fashion, internal stresses that develop during the beta to alpha phase transformation would be allowed to dissipate. The resulting rod castings had as cast densities of 19.61 gm/cc to 19.65 gm/cc. This information was presented as the 2010 Workshop.

The cast material was characterized using a variety of techniques, including optical metallography, resonant ultrasound spectroscopy (RUS), density, dilatometry, and quasi-static mechanical testing. Three types of tensile samples were machined from the rod castings to characterize the material in a triaxial state. These specimens were nominally roundbars of 5 mm diameter by 25mm gage length with smooth, "A" notch and "D" notch configurations. Tensile testing was performed on a MTS Alliance 100 loadframe located in an inert-atmosphere glovebox located at LANL. In order to allow for accurate high-temperature testing, the testframe is equipped with an environmental chamber. In addition, a Messphysik ME-46 Videoextensometer system is employed to allow for noncontact strain measurements through both the glovebox and environmental chamber windows. Samples were tested in the alpha, beta and gamma phases at a strain rate of 10^{-1}/min.

A photo of a strained plutonium sample tested in the beta phase at 190°C, 0.1/s is shown in figure 1 along with an image of the resulting fracture surface. The resulting strains to failure were significantly lower than those reported by previous authors [2], despite higher-density starting material. The fracture surface shows a large off center void that is strongly suggestive of a preexisting defect, presumably from the casting process. It is likely that the presence of defects lead to premature failure.

Metallography of the as cast rods revealed cracks and voids, particularly within the center of the rod. Based on these results, the decision was made test sub-sized tensile specimens from smaller rod castings to better control the phase transformation path and reduce defects. The new rod mold will cast 12 rods, 10.8mm diameter by 95mm long.

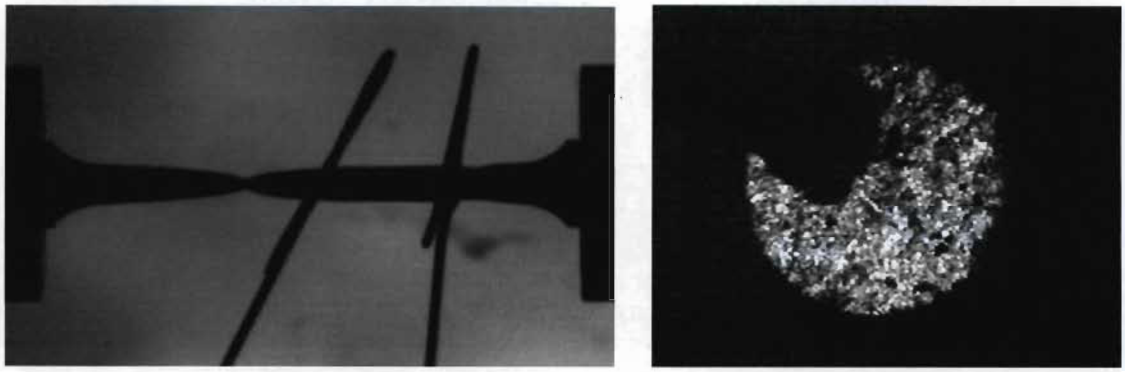

Figure 1. Left: Photo of plutonium tensile sample tested at 190C, 0.1/s as seen through the videoextensometer. This sample necked outside of the flags. Right: Fracture surface of the same specimen showing a large off center void that is strongly suggestive of a preexisting defect

The goal in casting the smaller rods will be the same as the large rods; create a thermal gradient that solidifies and directionally cools the rod from the mold bottom to top through the gamma to beta phase transformation. At this point in the casting process the mold and metal will be nearly isothermal. Removing the heat from the system on further cooling will be very slow and controlled allowing the internal stresses developed during the phase transformations to dissipate.

Other features have been added to the new mold design to correct problems that occurred with the previous design. Computer simulations have been run on the new rod design to determine the optimum process parameters such as metal pouring temperature and initial mold temperature.

The presentation will include results from the both the computer simulations and experiments as well as metallography of the as cast material.

References

[1] Nelson RD, Dahlgren SD,. Microstructures of Deformed Alpha Plutnoium. Report BNWL-981 UC-25, Metals Ceramics and Materials, 1969

[2] Wick, OJ. *Plutonium Handbook.* La Grange Park, IL: The American Nuclear Society;1980.

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Overview

Review of work presented in 2010

Metallographic characterization of 2010 rods

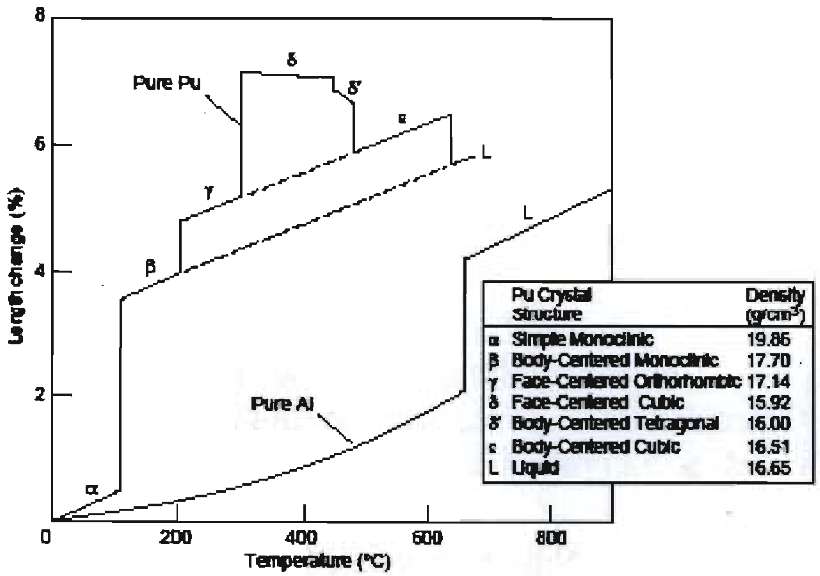
Triaxial tensile (3T) test results

Mold design for smaller 3T rods

Summary and future work

LANL history of casting unalloyed Pu

Unalloyed plutonium is difficult to cast due to the 9% volume contraction during the beta to alpha transformation. Casting defects include porosity, microcracking, and macrocracking to the point of catastrophic splitting.



The goal of this research is to produce high quality unalloyed, alpha phase plutonium for quasi-static and high strain rate mechanical testing.

1960s Del Harbur chill cast high purity (99.99wt%) Pu rods

9 to 16mm diameter, 254 mm long

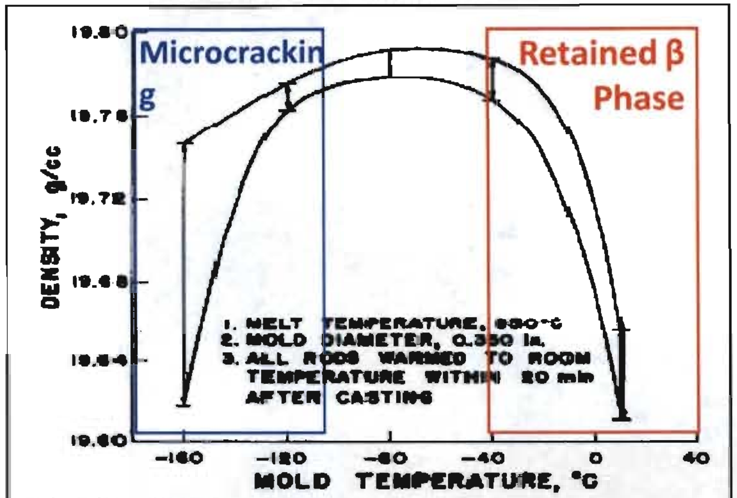


Fig. 3. Effect of mold temperature on ultimate densities of Pu rods.

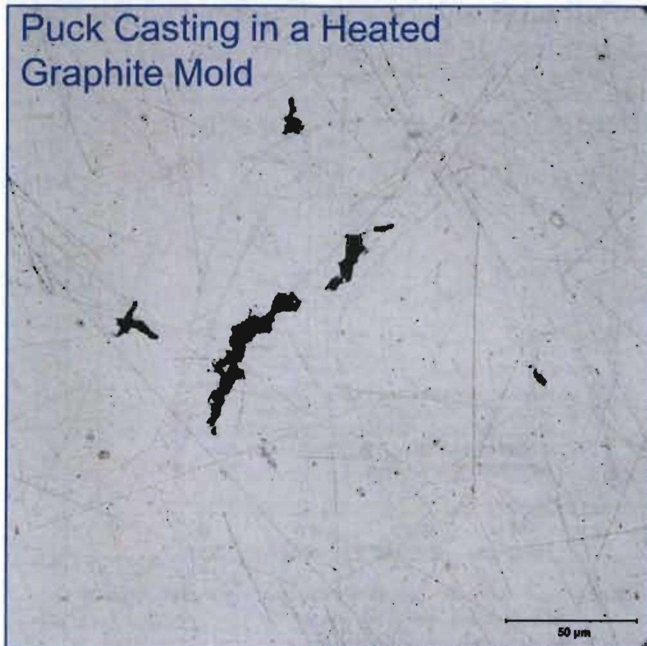
Two conclusions:

1)Rapid cooling, transform directly from ε to α, ε is quite ductile and yields, allowing significant contraction without cracking

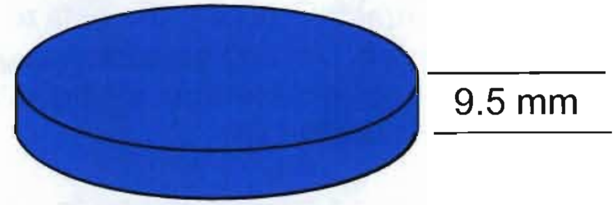
2)Transform directionally to α, (edge to center) little or no cracking occurs

LANL currently does not have a chill casting capability. We need to determine a different method of achieving the same results.

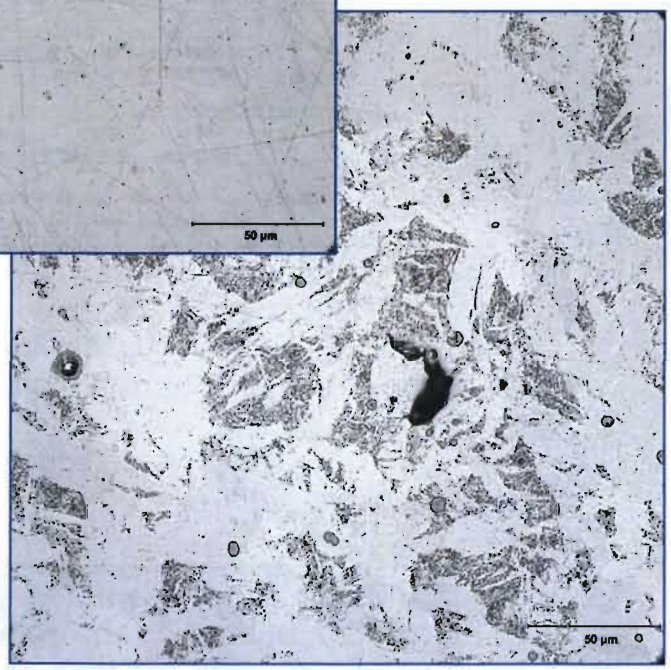
Microcracking and Retained β Phase



As-polished micrographs reveal microcracking and electro-polished micrographs reveal high temperature retained β phase (marbled microstructure).



38 mm diameter,
3.2 to 9.6 mm thick

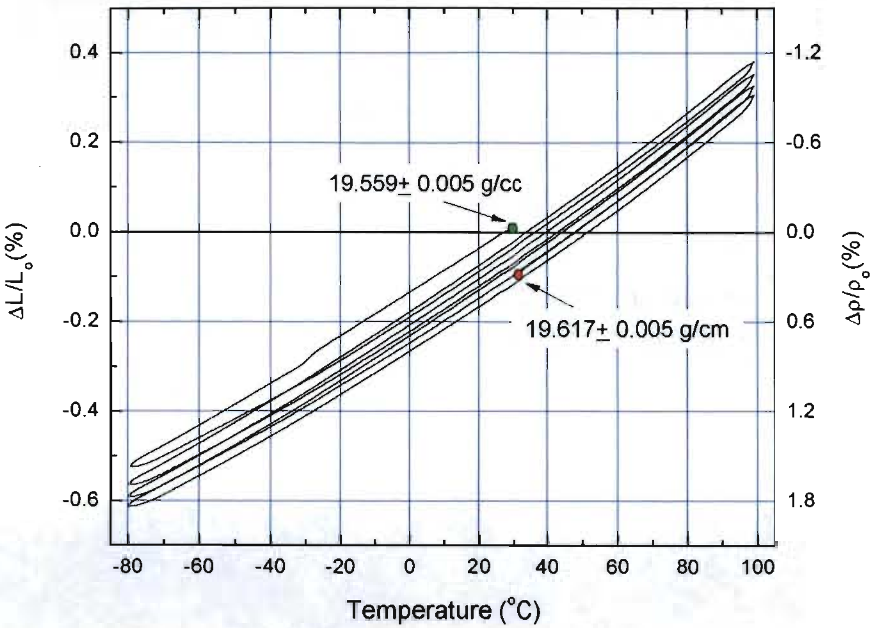


<u>As-cast α-phase unalloyed Pu pucks</u>

- Between 10-20% by volume β-phase as determined by density and thermodynamic properties. Kinetics plays a role in volume fraction of retained β-phase.
- Microcracking is revealed by metallography, but shown to be minimal (<0.2% by volume).
- Thermal expansion exhibits behavior of super-cooled phase transformations.

Thermal cycling to transform remaining beta phase

Kinetics of β→α Transformation in Cast α-Pu



Thermal cycling provides compression stress to drive the β→α transformation:

$$\Delta\sigma_T \approx \alpha_{\alpha Pu} E_{\alpha Pu} \Delta T_{Cycle} - \alpha_{\beta Pu} E_{\beta Pu} \Delta T_{Cycle}$$
$$= (\alpha_{\alpha Pu} E_{\alpha Pu} - \alpha_{\beta Pu} E_{\beta Pu}) \Delta T_{Cycle}$$
$$= 7x10^{-1} GPa$$

10 times the yield stress for β-Pu!

Time-Temperature-Transformation Diagram for β→α

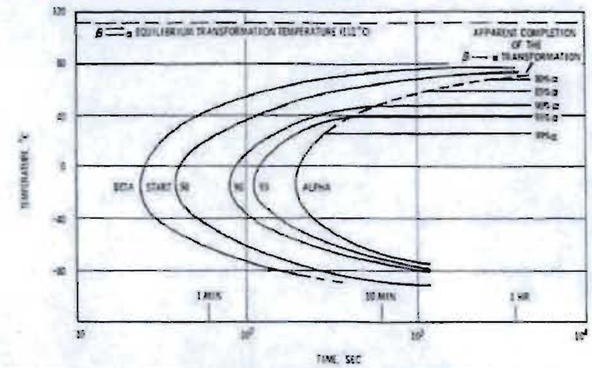
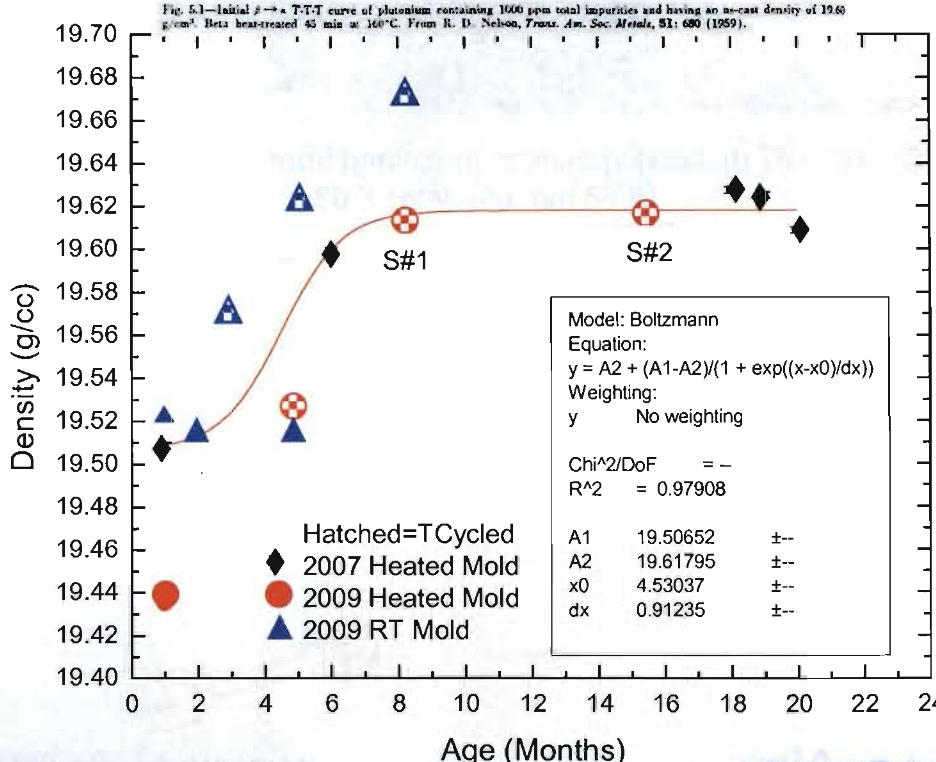
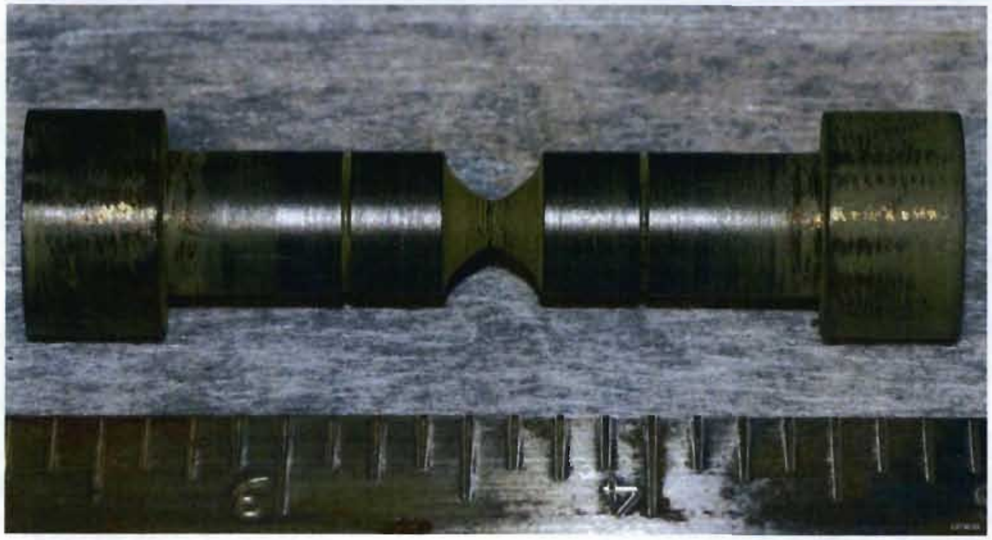


Fig. 5.1—Initial β→α T-T-T curve of plutonium containing 1000 ppm total impurities and having an as-cast density of 19.60 g/cm³. Beta heat-treated 45 min at 160°C. From R. D. Nelson, *Trans. Am. Soc. Metals*, 51: 680 (1959).



Casting Pu for Tri-axial (3T) Tensile testing

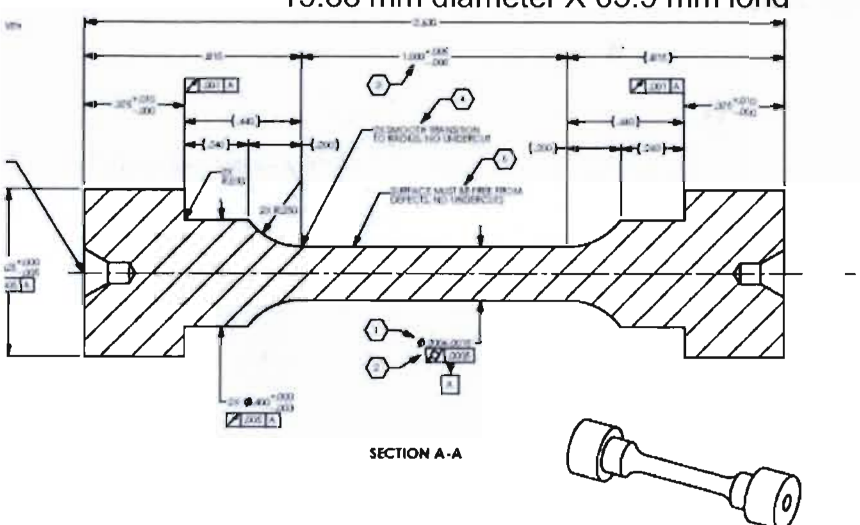


The puck dimensions are too small to machine full size tensile and 3T specimens.

Increasing the puck size to accommodate a 3T specimen would likely lead to microcracking and non-uniform properties across the puck.

A rod geometry was chosen to make larger samples of uniform cast material.

"D" notch 3T (triaxial) specimen machined from delta stabilized Pu.
15.88 mm diameter X 63.5 mm long



Perform many computer "experiments" and a few actual experiments.

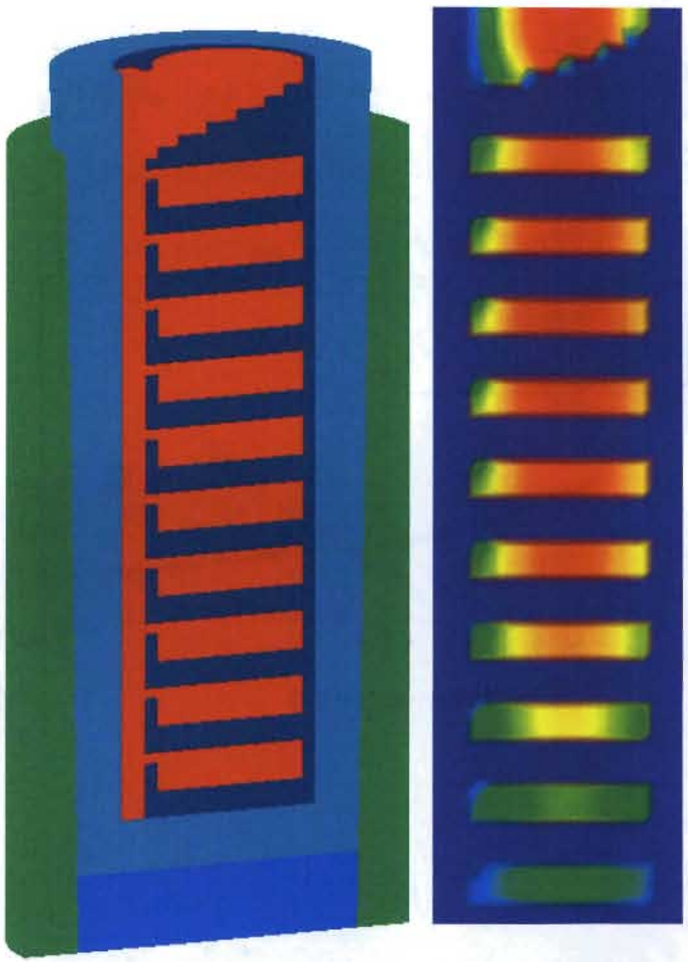
Predict and control microstructures.

Enable rapid development of optimized casting processes.

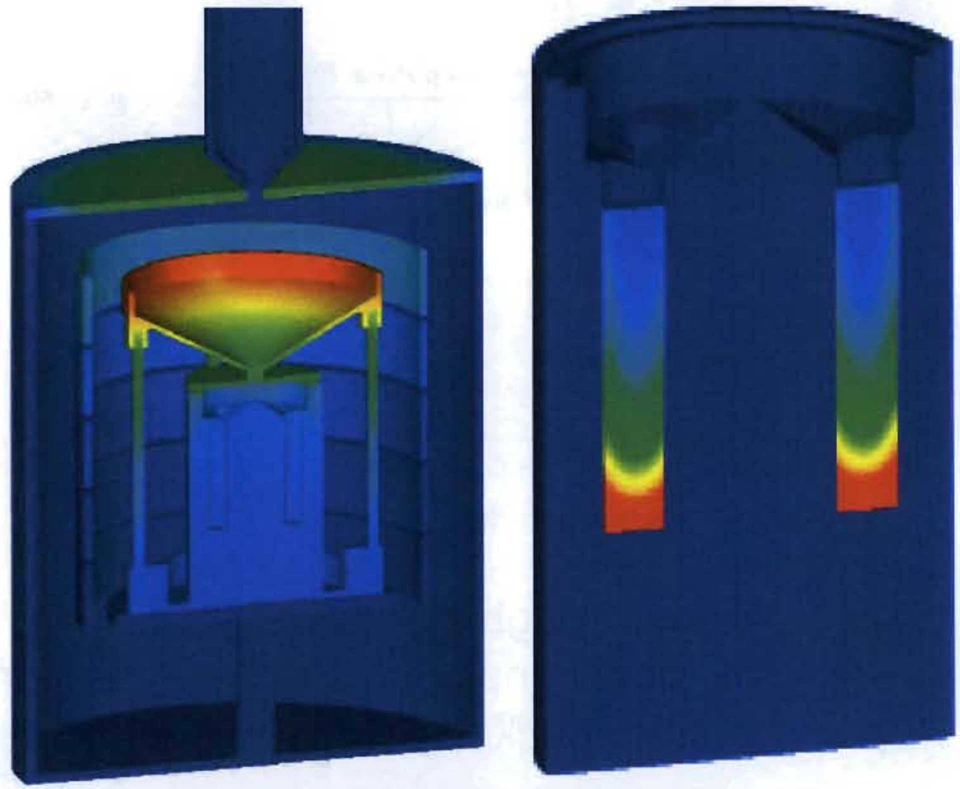
Perform highly instrumented castings to provide data for process modeling verification.

Simulations for puck and rod castings

TRUCHAS models for phase transformation studies
- **Electromagnetics**
- **Radiative heat transport with view factors**
- **Conductive heat transport**
- **Properties for pure Pu**

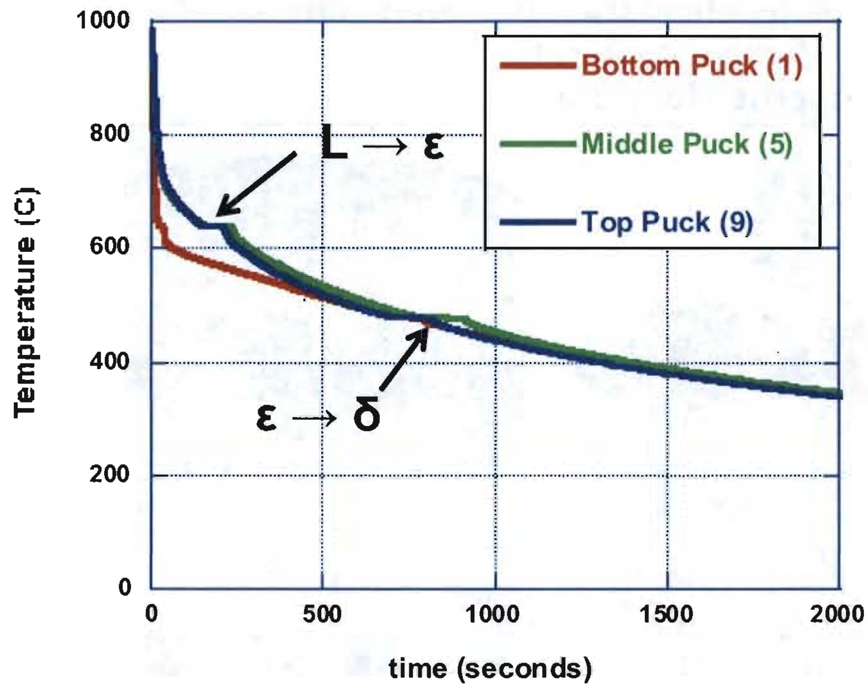


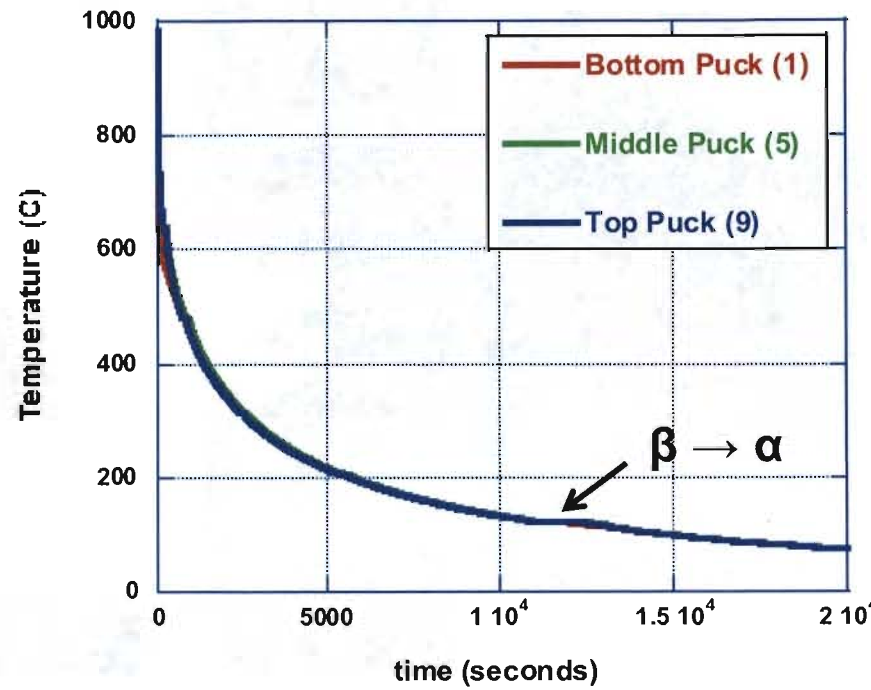
Puck casting simulations showing mold configuration and relative solidification times



Rod casting simulations showing mold preheat and beta to alpha transformation

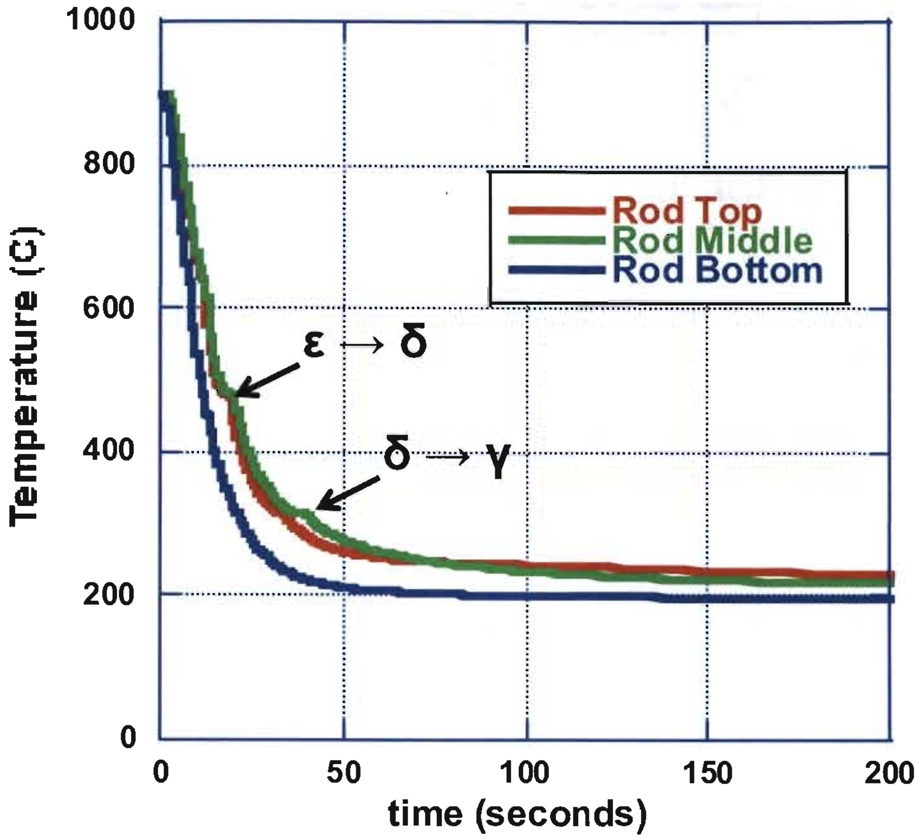
Simulation of cooling curves for puck casting

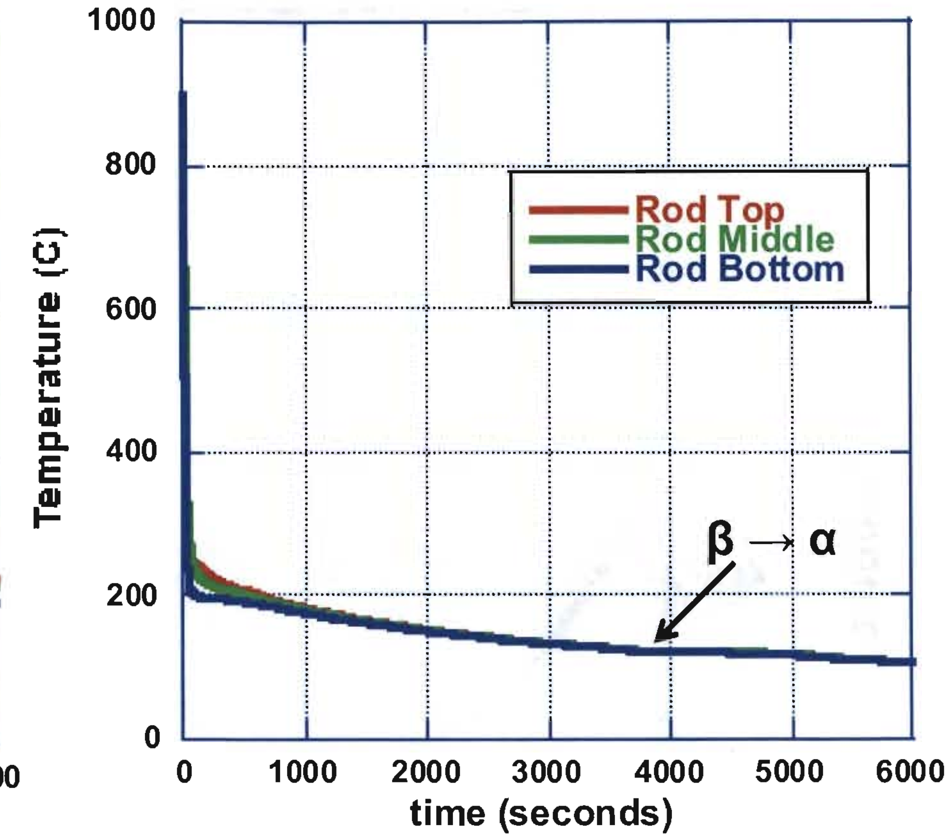




Slow cooling, through all phases
Transforms directionally from edge to center
Top and bottom pucks cool faster than center pucks

Simulation of cooling curves for rod casting



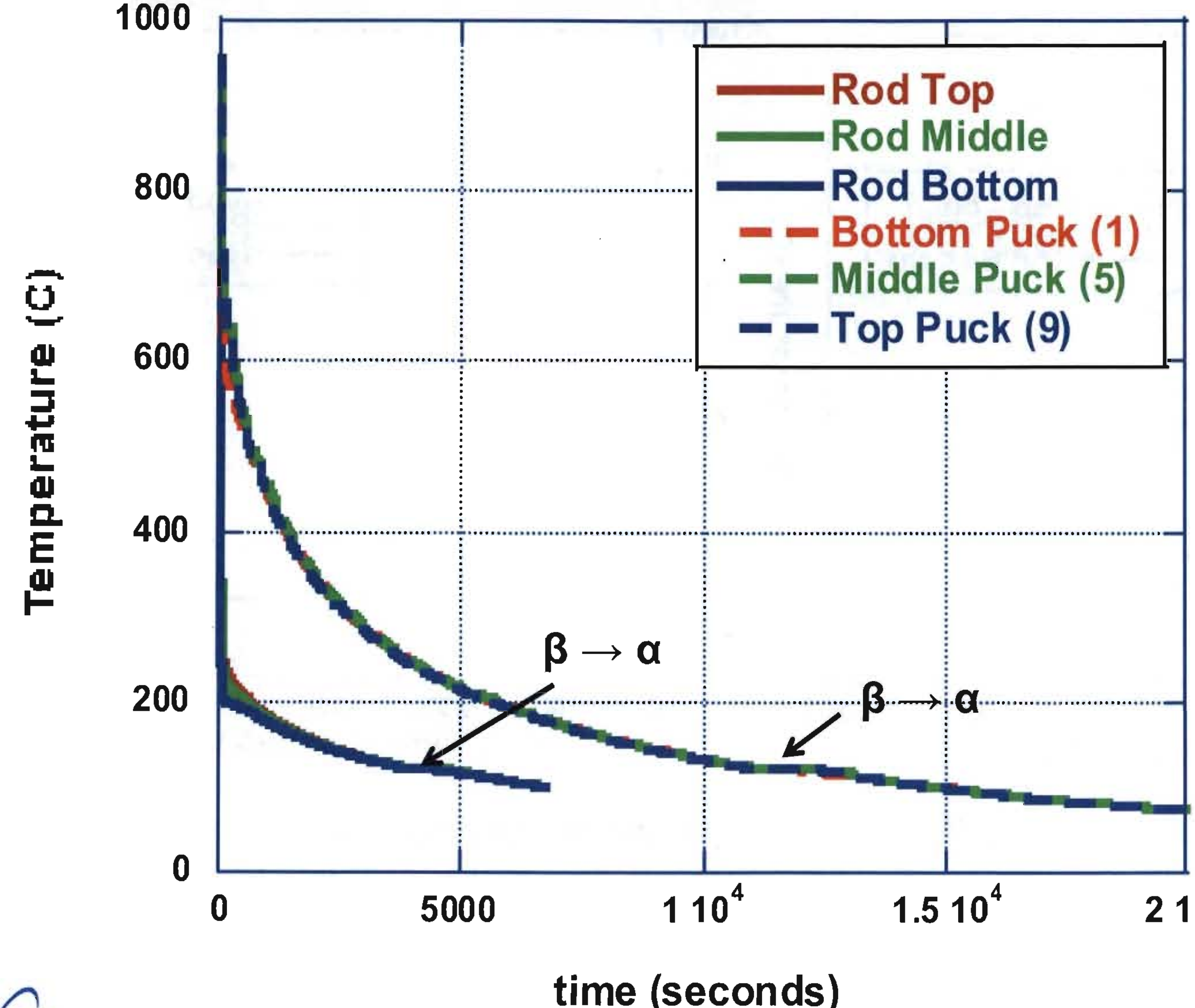


Initial cooling is rapid to ~ 225°C

γ → β transition is at ~215°C

Subsequent cooling is slow through the beta phase and the beta to alpha transformation.

Comparison of simulated of cooling curves



The pucks cool much slower through all phase transformations.

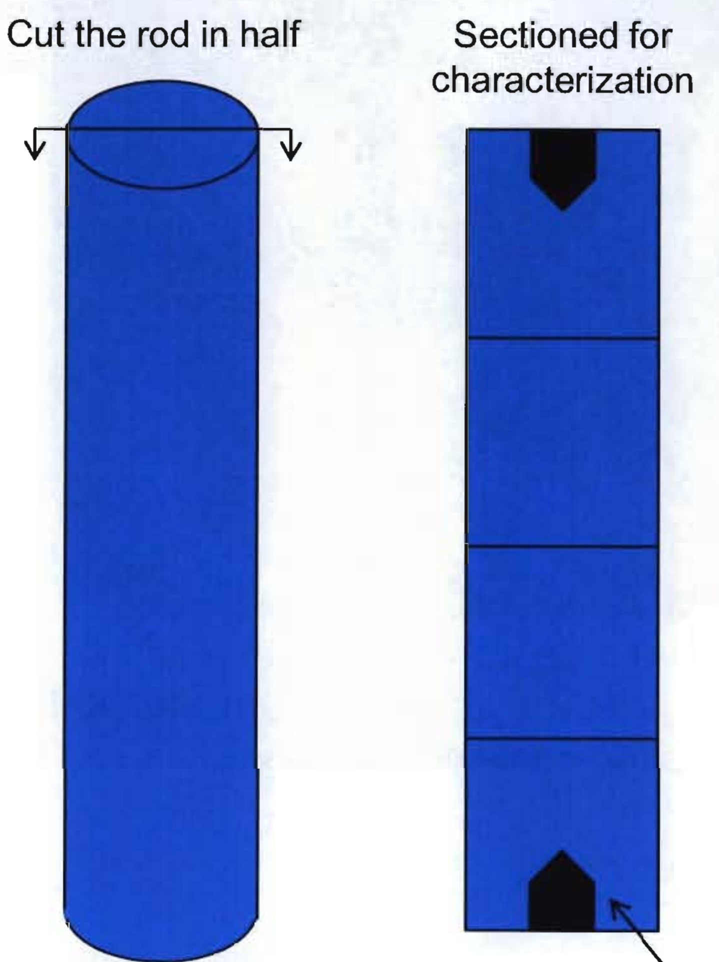
Casting of 500 ppm Ga alloyed Pu in rod mold

The heat up of the rod mold was exactly as predicted. The rods were poured at a funnel temperature of 575°C and a mold temperature of 150°C. The metal temperature was 900°C. The first casting contained 500 ppm Ga. The second casting contained 300ppm.

Both castings had non-symmetric fills. The cause is still unknown.

The average initial as-cast density from the rods was 19.66 gm/cm^3 ranging from 19.63 to 19.69. The rods were machined into tensile bars. One rod was sectioned for characterization.

Characterization of 300 ppm Ga alloyed Pu rod



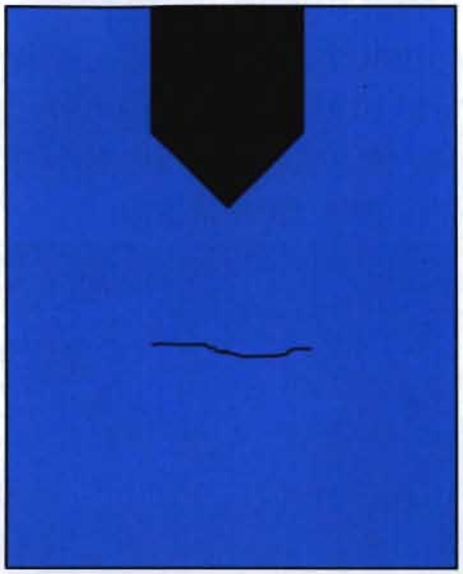
One half :
Metallographic analysis of all sections

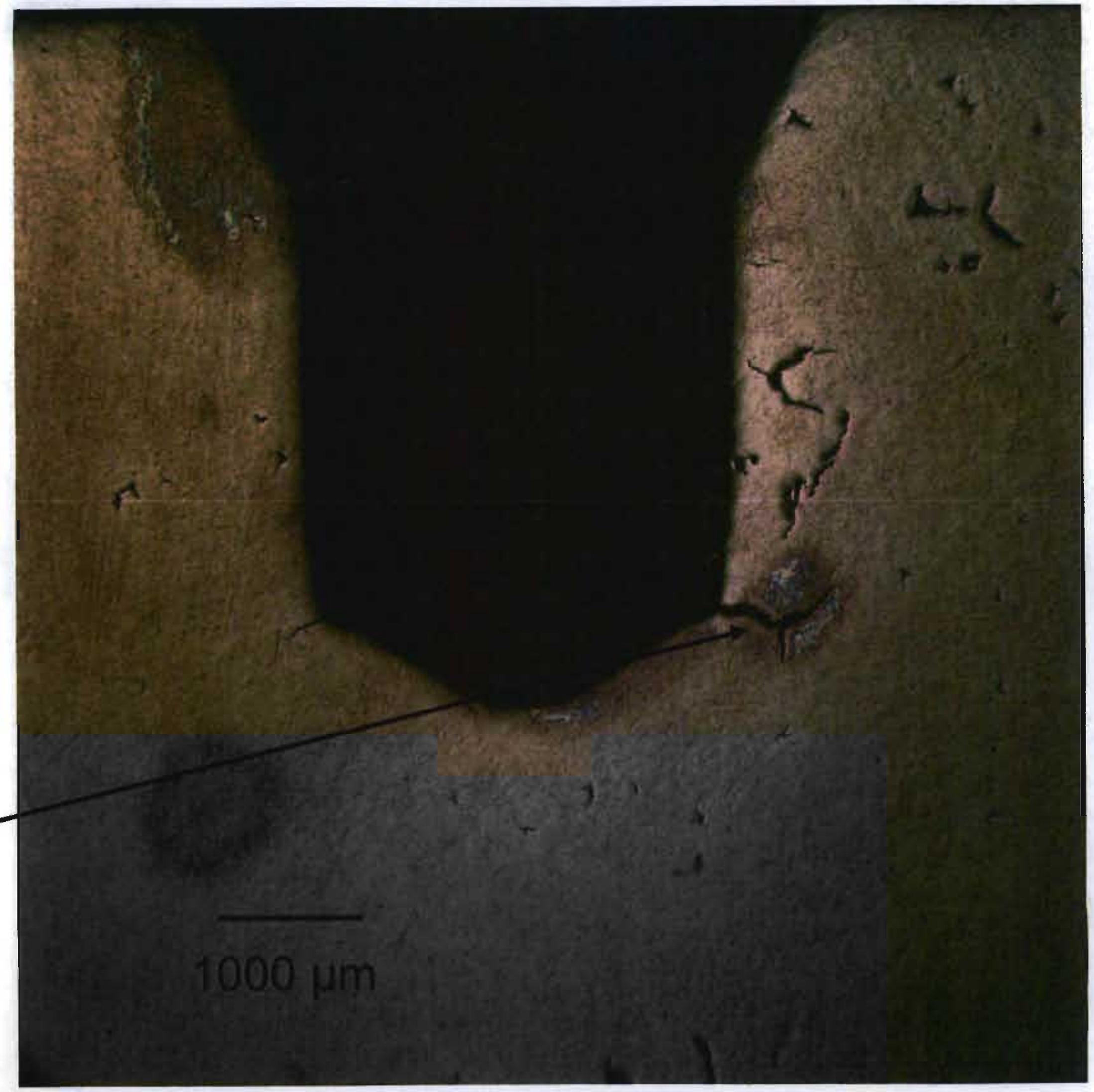
Other half:
Thermo dynamic properties on samples from middle section:
- Dilatometry (3 specimens) and
- Differential Scanning Calorimetry (6 specimens)
- 3 quasi static specimens (not yet tested)

Three remaining sections reserved for thermal cycling studies (not yet performed)

Rod was rough machined to clean up outer surface
Center drilled for machining

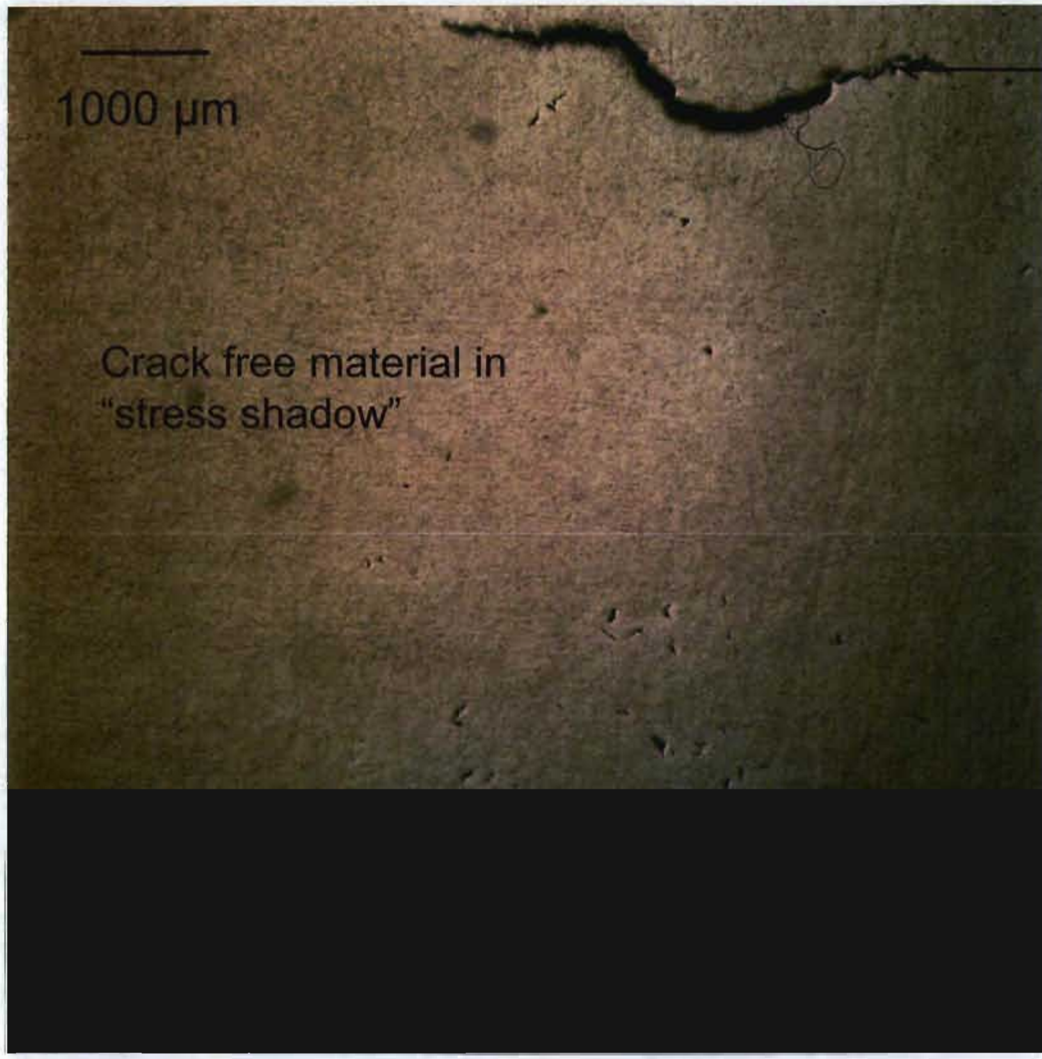
Metallography of 300ppm Ga alloyed Pu rod



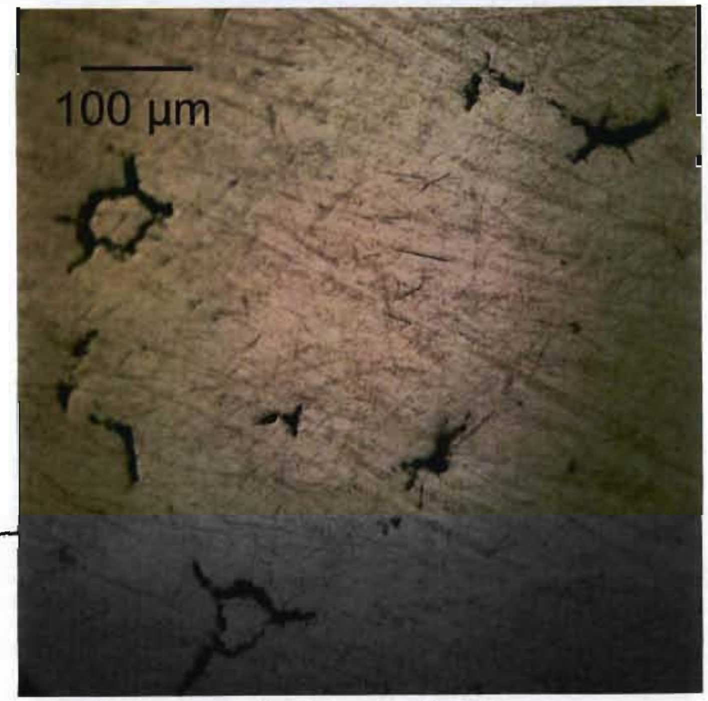


Machining induced cracks:
The center drilling operation heated the material into the beta phase. Upon subsequent cooling, cracks developed during the transformation back to the alpha

Metallography of 300 ppm Ga alloyed Pu rod



Crack initiated during delta to gamma transition. Crack growth and opening during cooling can accommodate subsequent strain producing a "stress shadow"

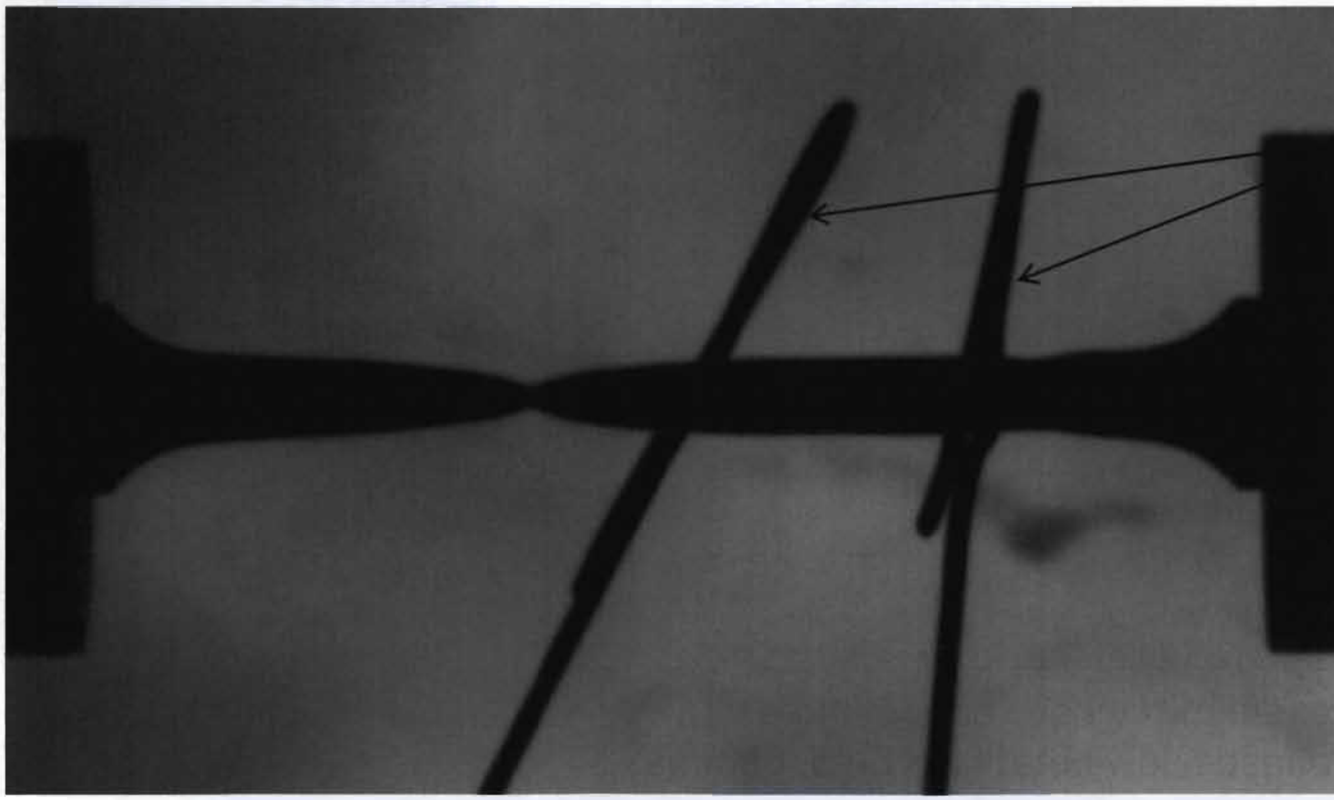


Cracks form during beta to alpha transformation most likely nucleating at triple points.

High temperature tensile testing with video extensometry

Video extensometry allows accurate non-contact strain measurements, as well as helping to normalize specimen area from room temperature measurements.

Black/white contrast is critical to quality measurements.



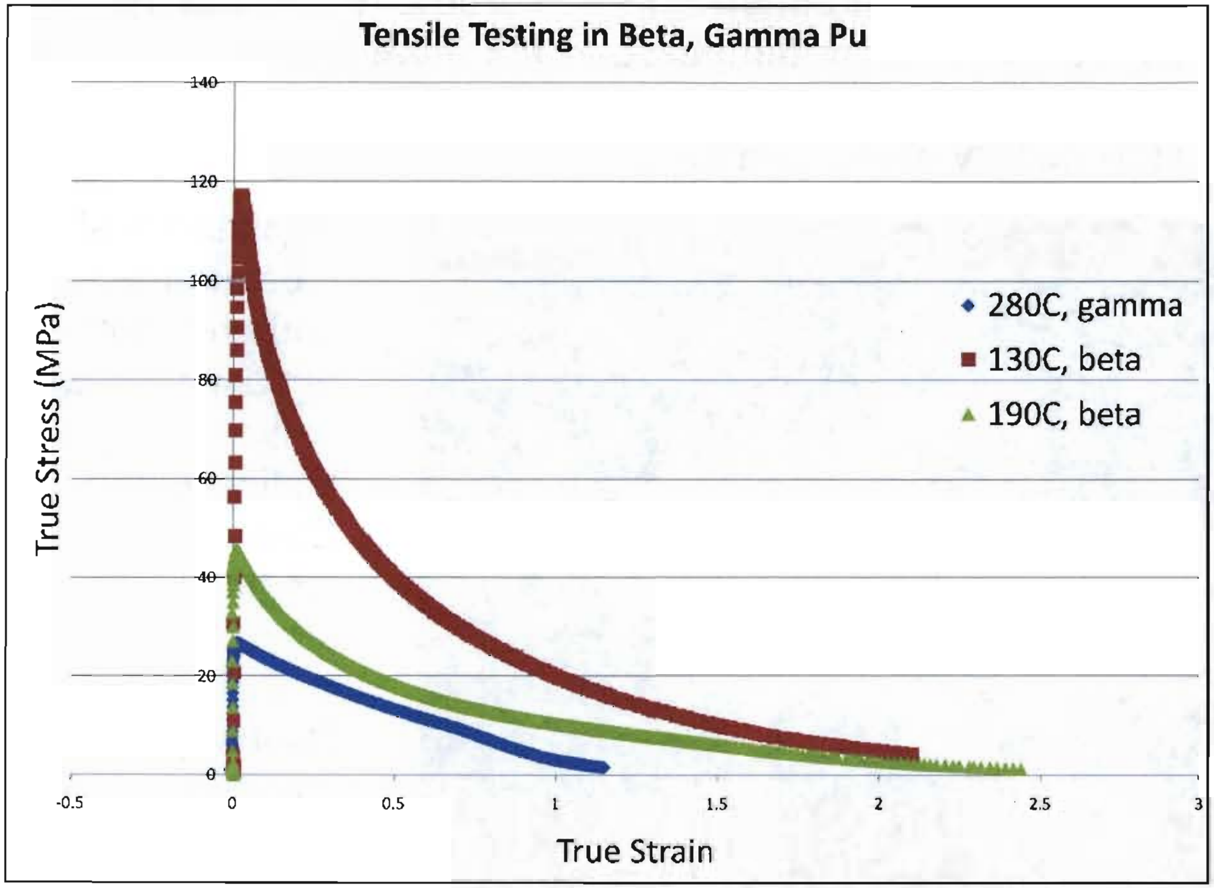
Continuous strain measurements are gathered both radially and axially.

Optical Flags- Create gauge length for axial strain

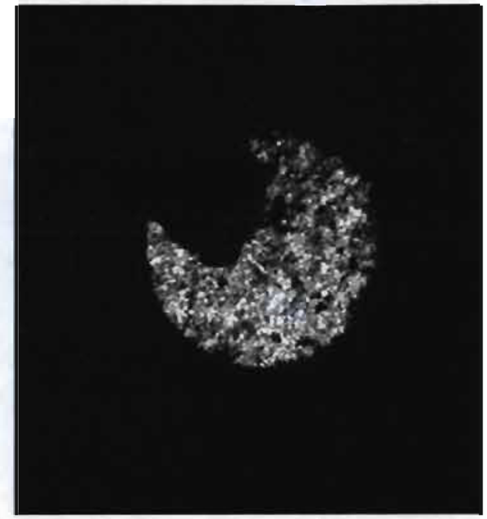
Diametral strain is calculated from the gauge section curvature in the cross section

Smooth tensile bar tested in the high beta, 190 ° C

High temperature tensile testing smooth notch bars

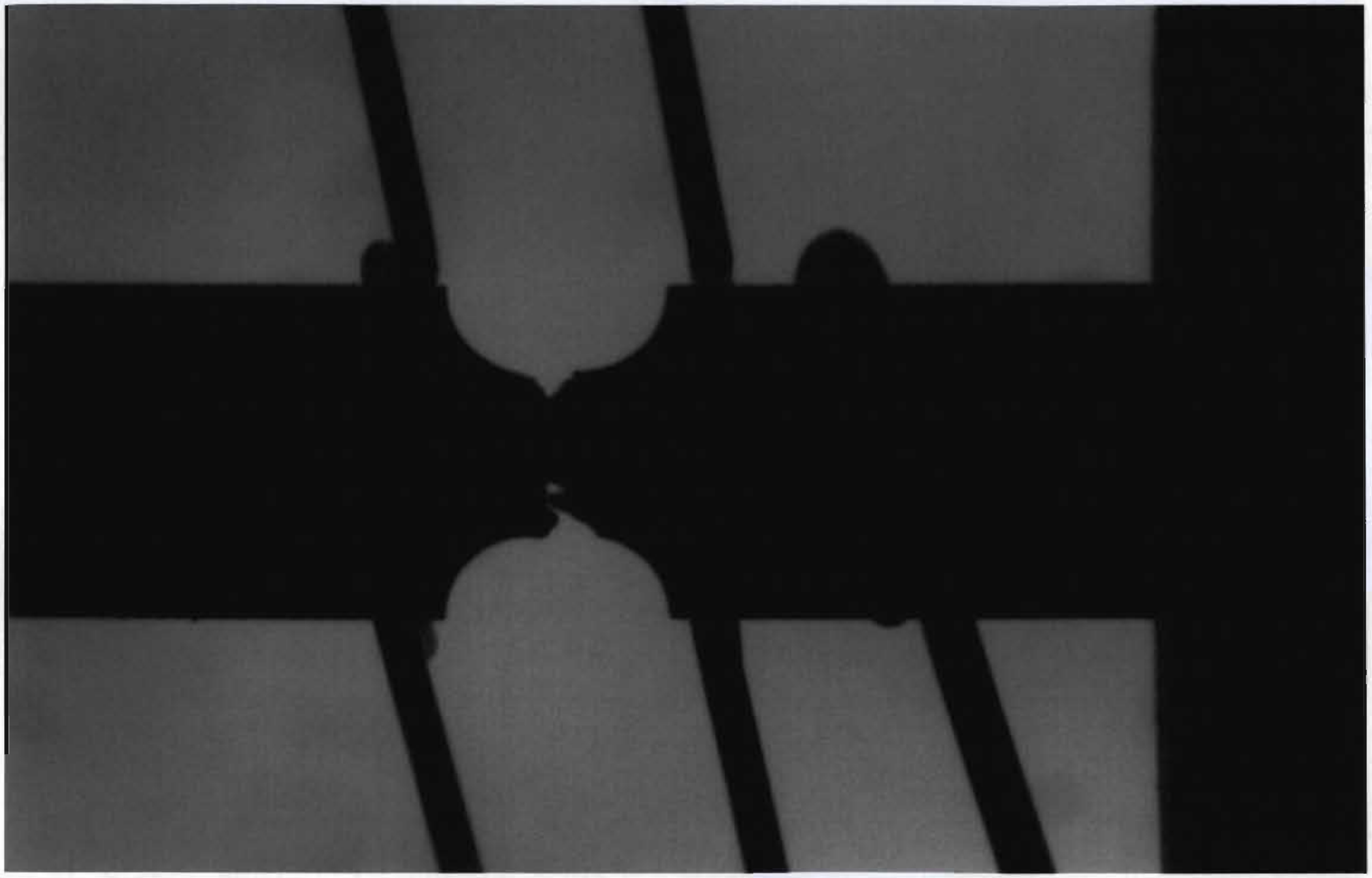


Stress strain curves of smooth tensile bars at three temperatures in two phases: low beta, high beta, gamma.



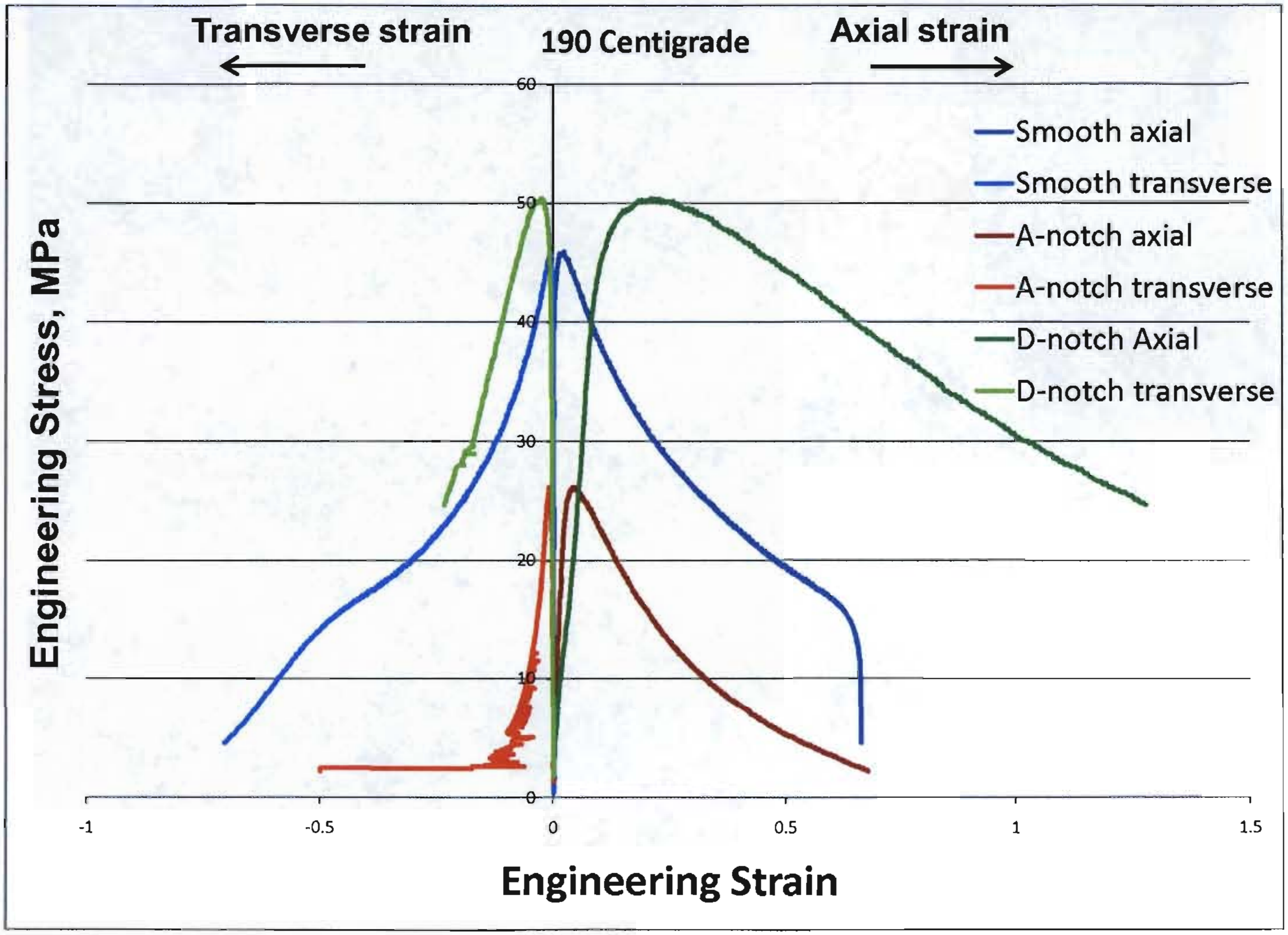
Macrograph of fracture surfaces indicates the existence of a large flaw.

High temperature tensile testing A notch bar



190C, A notch 0.100" gage length, 10^{-1}/min

Comparison of 3T tests at 190°C (high beta)



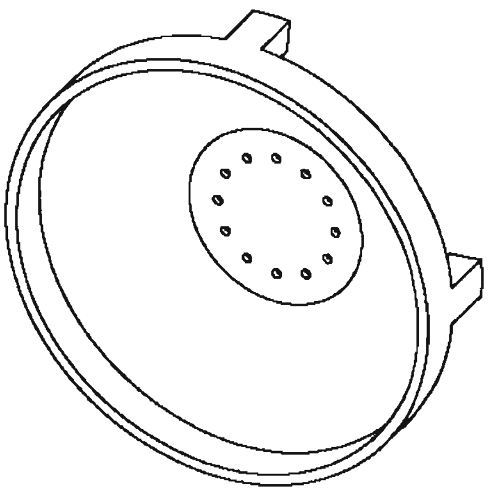
New mold design for half scale 3T rods

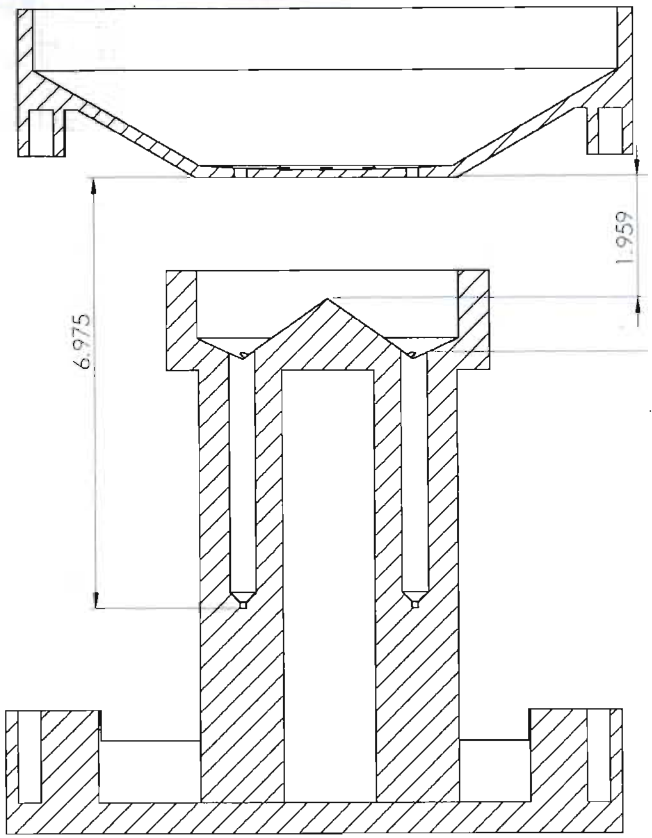
New casting

12 rods - 5 mm Ø X 100 mm long

New funnel with 12 pour holes to reduce non symmetric filling problems.

Original mold design had hollow core, this was later removed to create a better cooling profile.





Conclusions from large rod castings

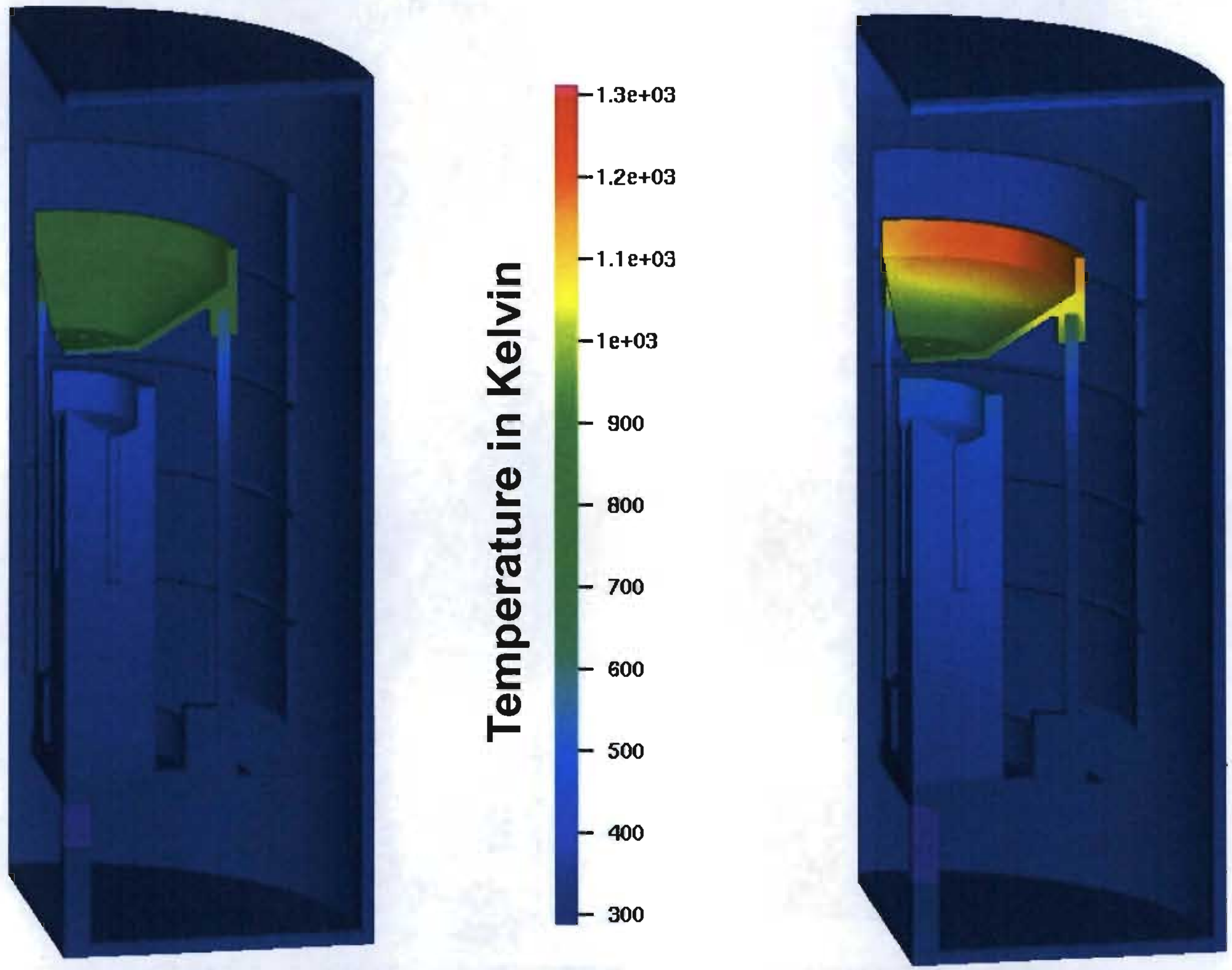
- Single phase unalloyed α-Pu is difficult to produce due to microcracking and retained β-Pu. Although computer simulations help drive mold and process parameter design, it appears that for the current application, the rod cross section and volume is too big to accommodate the transformation stresses using our available casting equipment.

- Better, more consistent data will aquired by testing smaller 3T rods, scaled by a factor of 2 with a gauge diameter of 0.100" (2.54 mm).

- Equilibrium thermodynamics is not sufficient to define phase transformations in plutonium - kinetics, microstructure, and impurity composition are important.

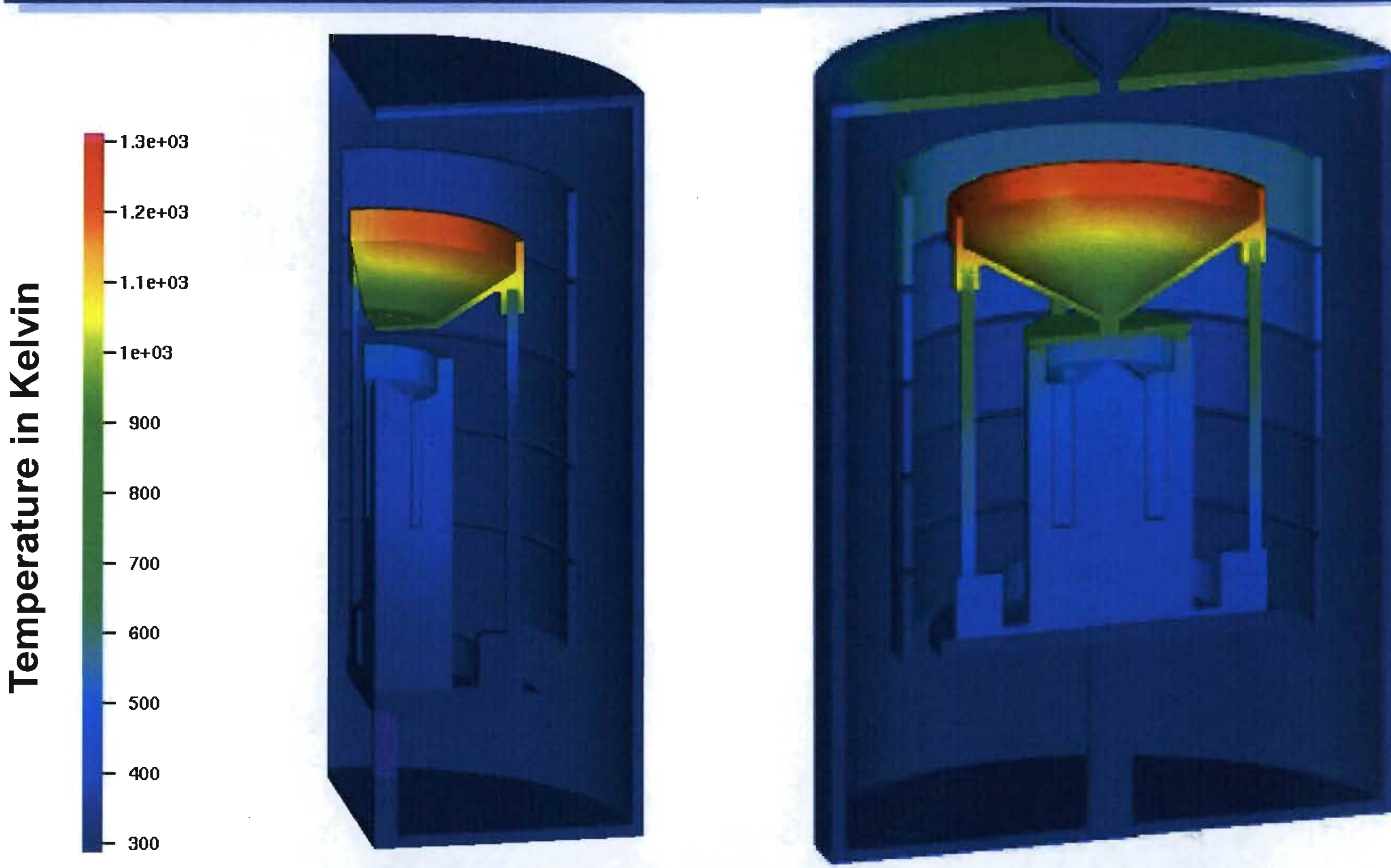
- Computer simulation are only as good as the material property data provided.

Comparison of mold preheat: smaller Pu Rods



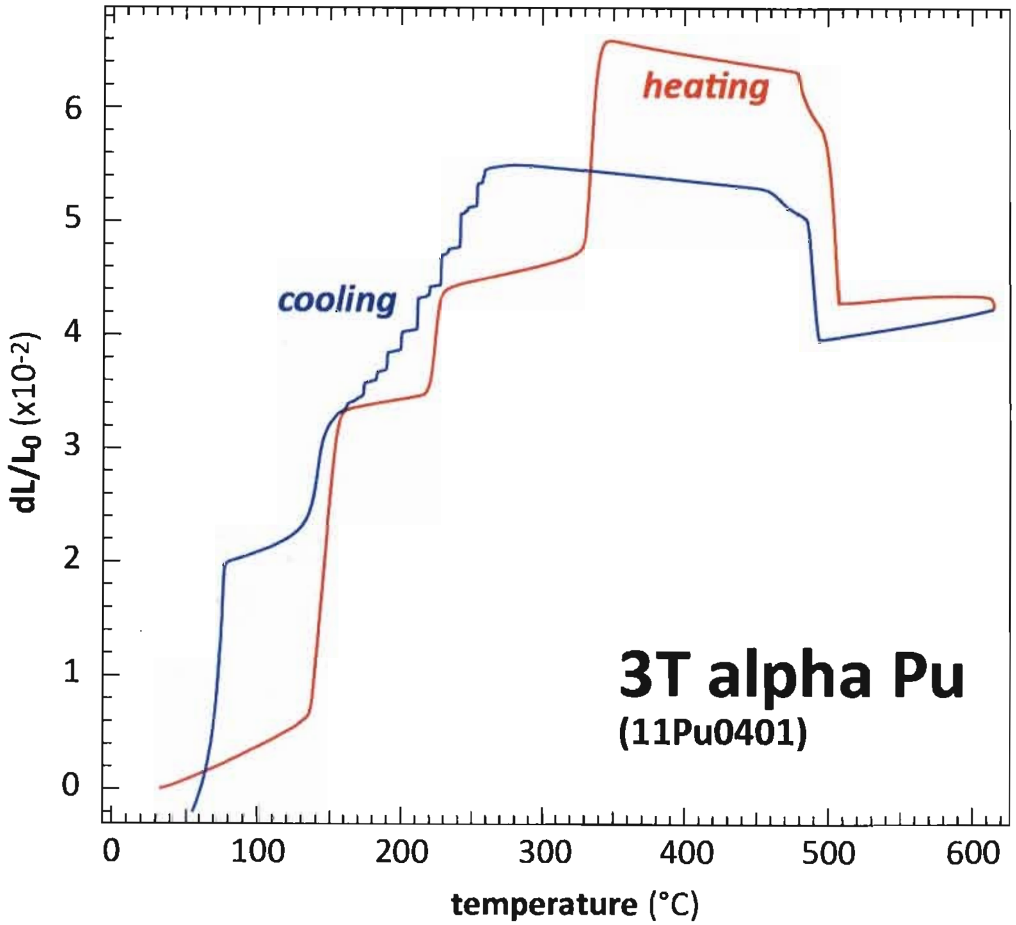
750 Amps for 5 minutes | **1000 Amps for 3 minutes**

Comparison of two molds: preheat



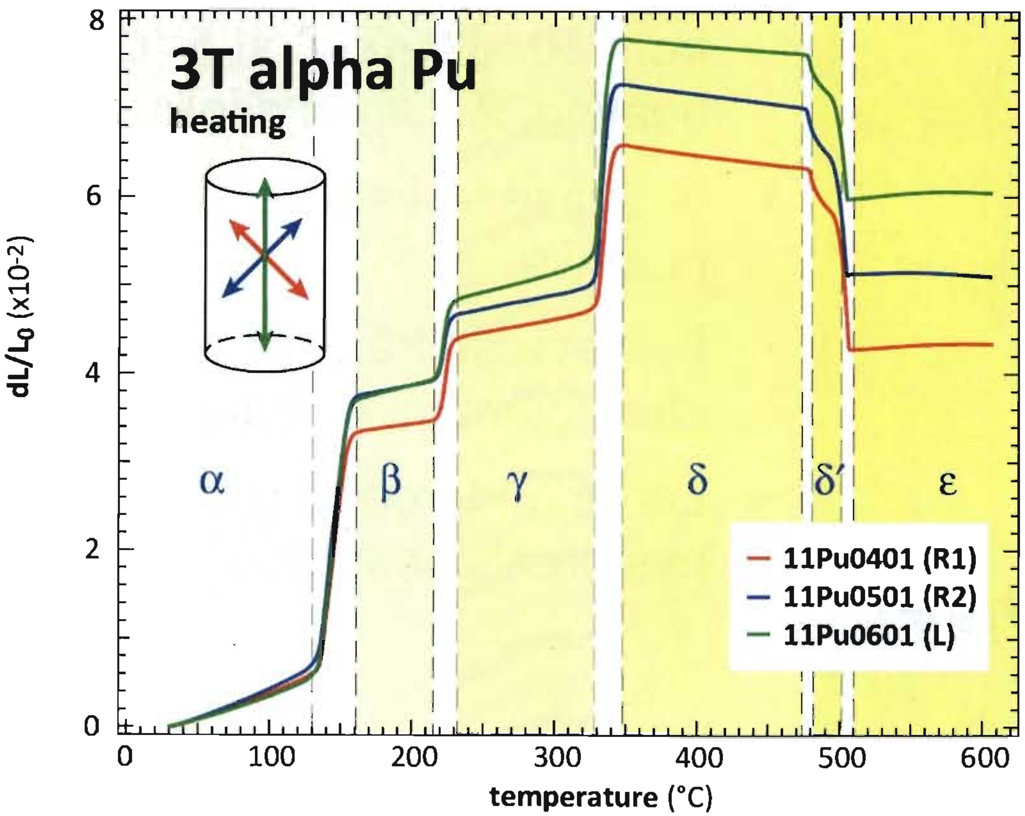
What about the material properties?

Dilatometry of 300 ppm Ga alloyed Pu rod



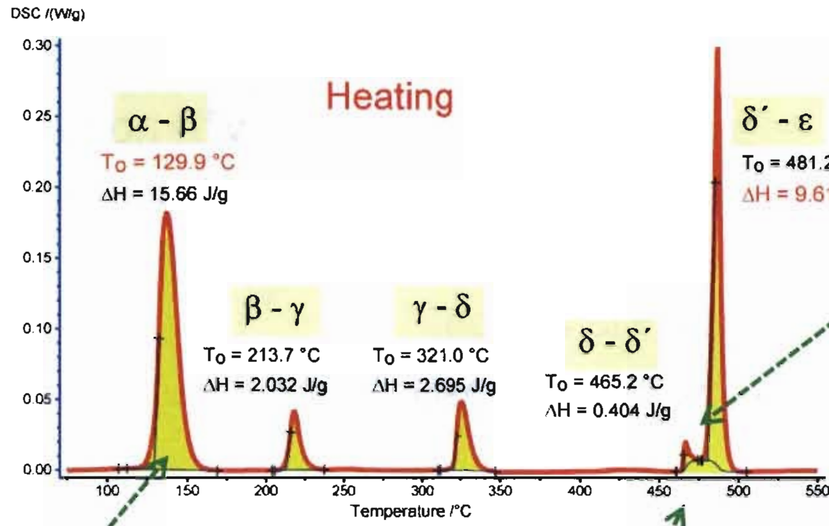
- thermal expansion measured from 30 to 620 °C at 5 °C/min heating, 2 °C/min cooling
- δ′ phase still stable at 300 ppm Ga
- δ-γ reverse transformation characterized by bursting
- Large undercooling and hysteresis observed

Thermal expansion differences in cast rod

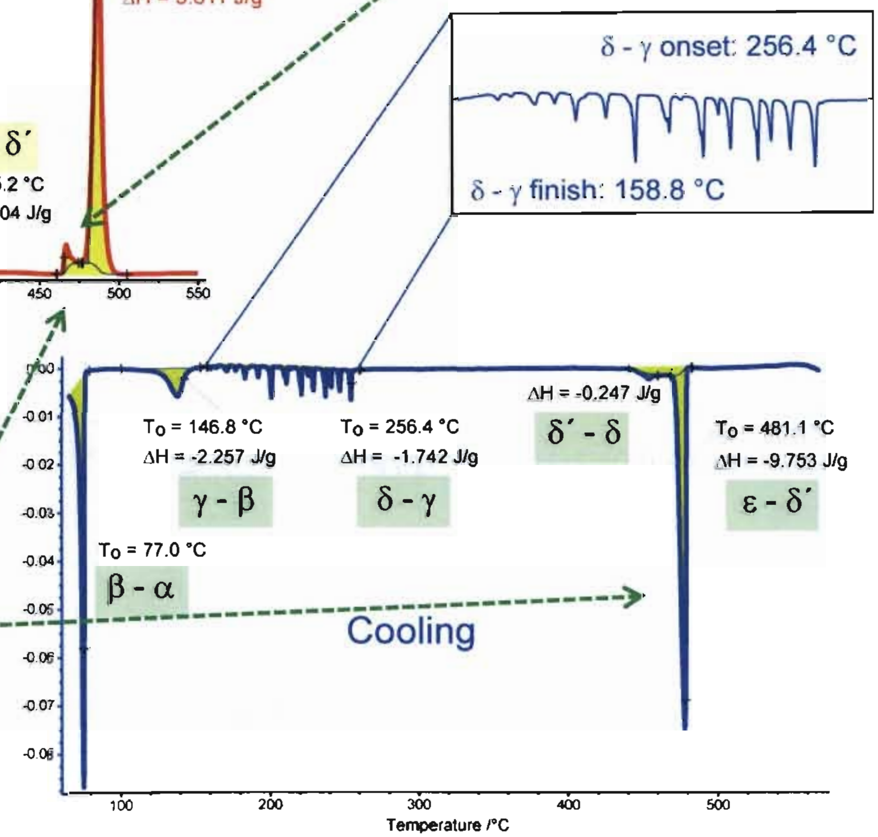


- three samples were cut from rod
- each dilatometry run represents a different orientation within the rod
- differences in curves probably reflect preferred orientation caused by anisotropic cooling in mold

Differential Scanning Calorimetry of 300 ppm Ga alloyed Pu rod

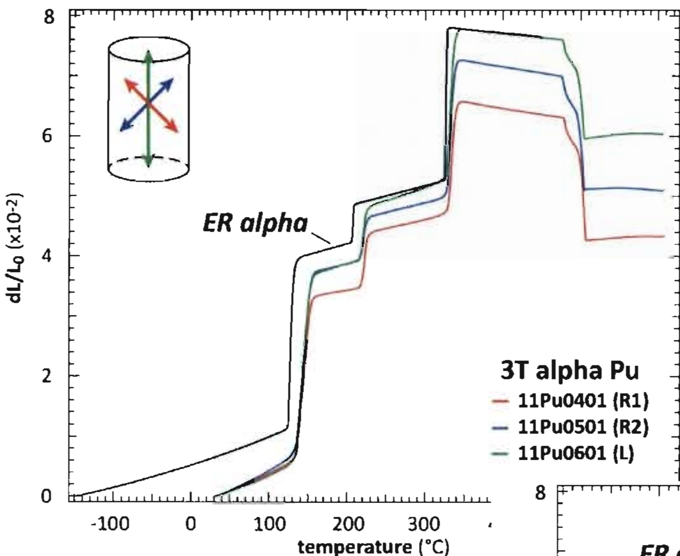


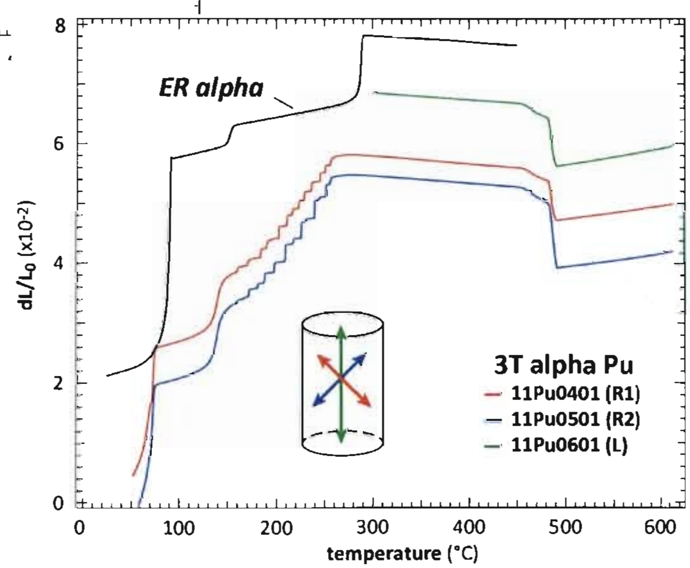
- Ga content does not suppress δ′ phase (as other impurities do)



- **Ga content shifts α → β transformation to higher T_o**
  130°C vs. 126°C
- **Some unexpected features**
  - δ′→ε transformation is ~35% more energetic than in pure α-Pu, and very sharply defined

Comparison with other ER alpha Pu





- electro-refined (ER) a Pu with no Ga has
  - lower transition onset temperatures
  - little to no bursting during δ-γ reverse transformation
  - narrower transition temperature fields

transition start-finish	no Ga	300 ppm Ga
α–β (°C)	124, 131	134, 154
β–γ (°C)	207, 209	218, 227
γ–δ (°C)	325, 327	328, 339

Summary and Future Work

Summary

- Single phase unalloyed α-Pu is difficult to produce especially in large cross sections
- Care must be taken to ensure quality material
- Video extensometry has successfully been used to test microalloyed Pu in three phases. The techniques allows accurate non-contact strain measurements while normalizing higher temperature phase specimen area from room temperature measurements.
- Computer simulations are very helpful in guiding mold and process parameter design for casting processes BUT accurate material property data is essential to produce quality simulations.

Future Work

- Complete thermal cycling studies on 300 ppm Ga alloyed Pu
- Perform quasi static compression testing in three orthogonal directions.
- Cast Pu using the new mold design for smaller 3T rods
- Measure thermomechanical properties using new, smaller rods.