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Title: FABRICATION OF UNALLOYED PLUTONIUM

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FABRICATION OF UNALLOYED PLUTONIUM

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Casting of unalloyed plutonium is challenging due to large volumetric phase transformations incurred during cooling, particularly the beta to alpha transformation at 117C. In unalloyed plutonium the beta-alpha transformation at 117C subjects the material to a 9% volume decrease inducing large internal stresses that can result in cracks and voids. Although multiple cast rods of micro-alloyed plutonium have been successfully manufactured using a chill cast procedure and a post-casting treatment (Harbur, et.al 1967). Los Alamos National Laboratory (LANL) no longer has a chill casting capability so we are exploring the casting of micro-alloyed plutonium by slow controlled cooling through the critical phase fields. Two geometries, pucks and rods, have been cast using traditional vacuum induction melting followed by thermal cycling to transform any remaining beta phase to alpha phase.

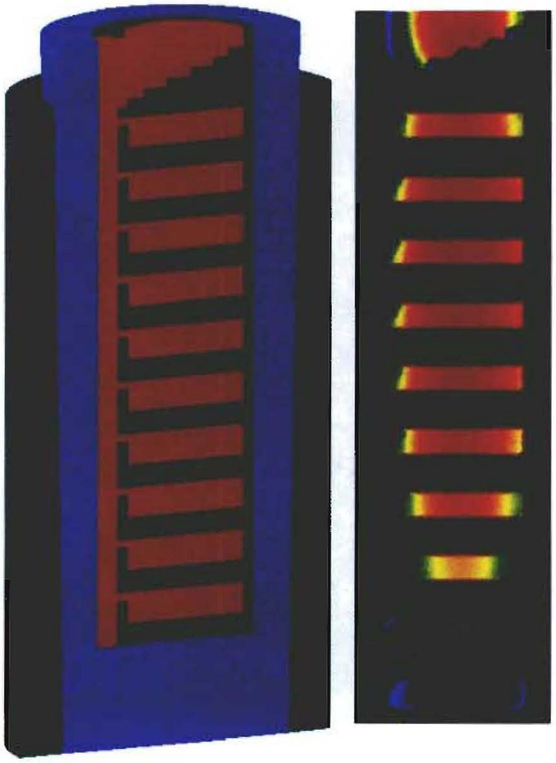

Figure 1: (Left) A cross section of the mold to cast 10 uniform pucks. The red portion represents the puck cavities. (Right) Computer simulation result from TRUCHAS showing non-uniform solidiification times. Red indicates the longest time and blue the shortest time.

The goal of the current process is to control the rate of transformation from the beta-alpha phase and reduce and/or eliminate the damage due to micro-cracking. A stacked puck mold design had been used successfully to cast unalloyed plutonium as shown in Figure 1. The mold contains ten puck cavities of either uniform or varying sizes. The puck cavities range in size from 38 to 50mm diameter and from 3.2 to 9.5 mm thickness. The puck mold design inherently allows a smaller thermal gradient across each mold cavity for accommodation of the internal stresses without cracking so satisfactory castings were produced with minimal thermal control. Unalloyed plutonium has been cast in this geometry producing pucks with as-cast densities of 19.44 gm/cc to 19.52gm/cc. With subsequent thermal cycling, the densities have been increased to values of 19.61 gm/cc to 19.68 gm/cc.

The next challenge is to create castings of unalloyed plutonium large enough for full size mechanical testing and analysis with a cross section of at least 20mm. At this size, the thermal gradient and subsequent transformation gradient across the material is large enough that tight thermal control is necessary to avoid micro-cracking and voids.

This work explores the casting of micro-alloyed plutonium 20mm diameter rods in a vacuum induction furnace using both tantalum and graphite molds. The goal is create a thermal gradient that solidifies directionally from the mold bottom to top and continue to directionally cool the plutonium through the beta phase. 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.

A combined experimental/computational approach has been used to design the rod mold as well as determine the best process parameters needed to produce the desired temperature profiles. Computer simulations are performed using the LANL developed computer coded TRUCHAS. TRUCHAS is designed to simulate the entire casting process starting with heating the mold and metal from room temperature with either electromagnetic or radiative heating. Simulation continues through pouring with coupled fluid flow, heat transfer and non-isothermal solidification. An example is shown in Figure 1. This approach also increases our understanding of the casting process which leads to a more homogeneous, consistent, product and better process control.

The presentation will include results from the both the computer simulations and experiments.

Fabrication of Unalloyed Plutonium

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DA Korzekwa, RM Aikin

Los Alamos National Laboratory

X Workshop on "Fundamental Properties of Plutonium"

RAS, Moscow

July 12 - 16, 2010

Overview

History and challenges of casting unalloyed Pu

Puck casting of 500ppm Ga alloyed Pu

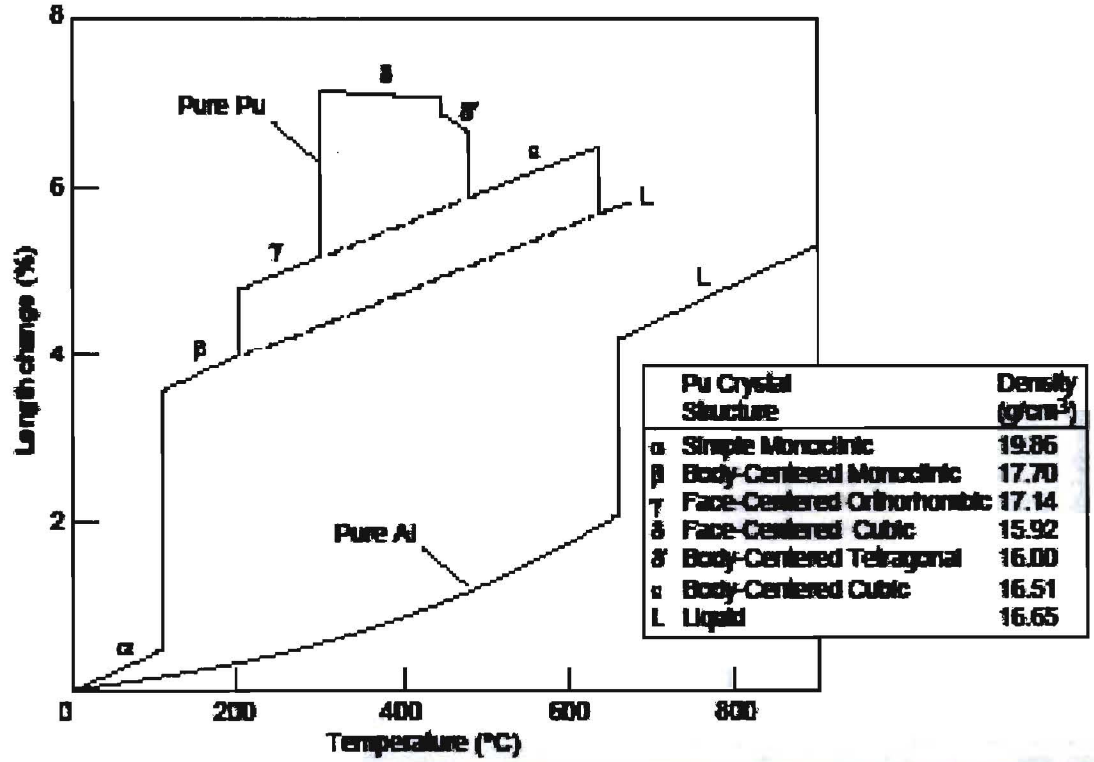
Mold design for rod casting
- Coupled simulation/experimental approach

Casting 500 ppm Ga alloyed Pu Rods

Summary and future work

Problems 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.

LANL history of casting unalloyed Pu

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

9 to 16mm diameter, 254 mm long
Ta lined Al molds with pre-pour temperatures: -160°C to 10°C
metal temperature at pour: 850°C
mold and metal heated to 25 to 85°C immediately after pour for 2 days

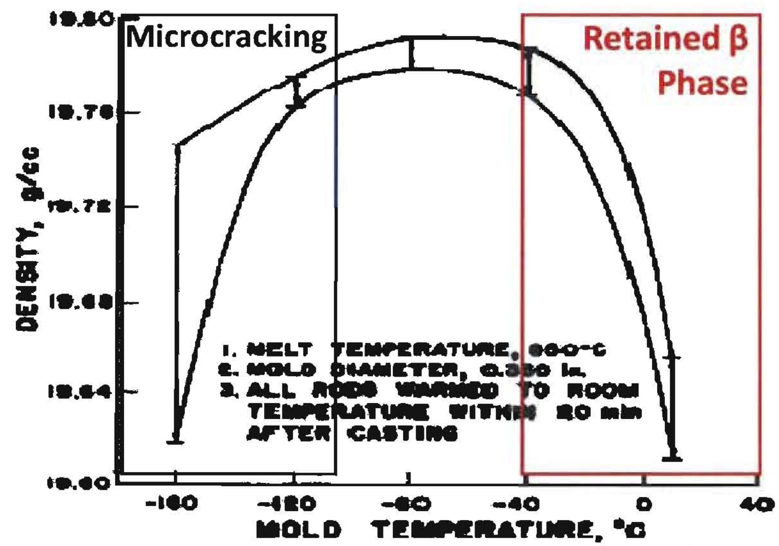


Fig. 3. Effect of mold temperature on ultimate densities of Pu rods.

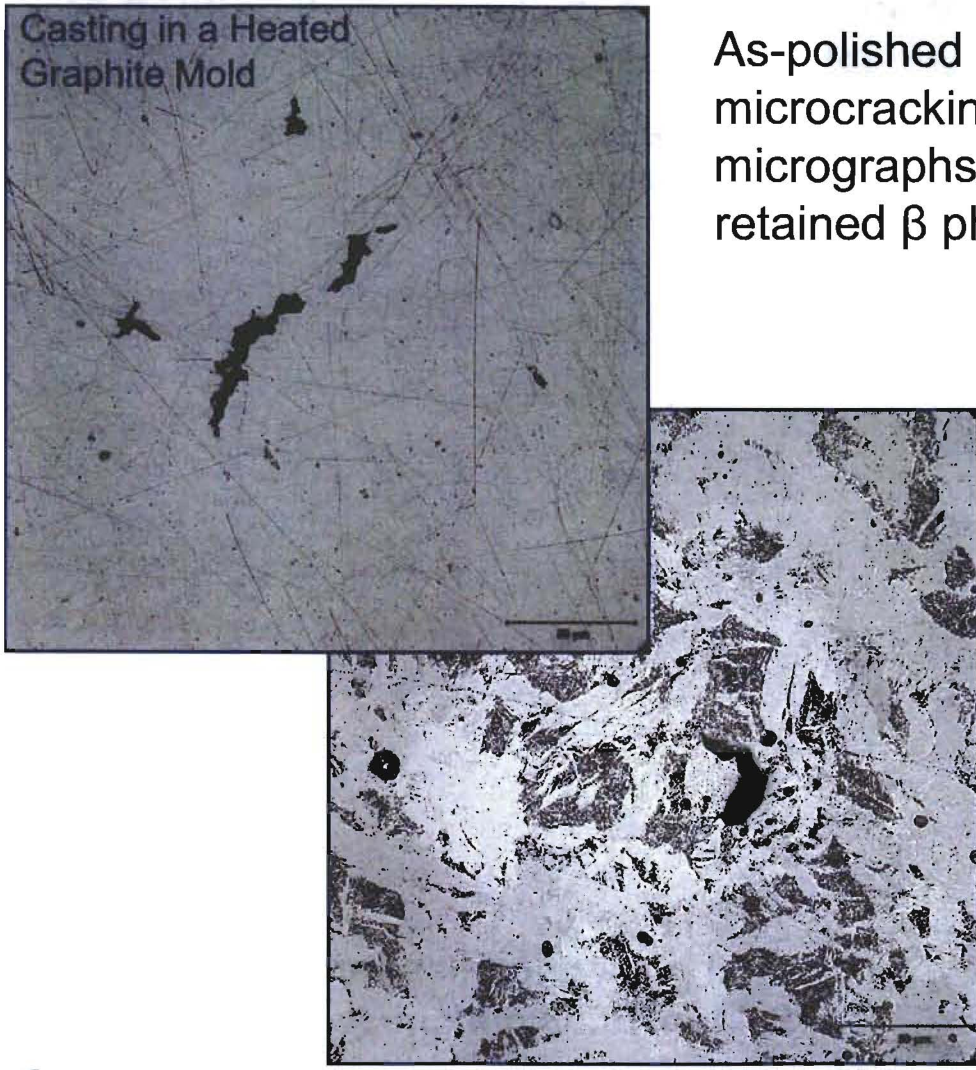
Two conclusions:

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

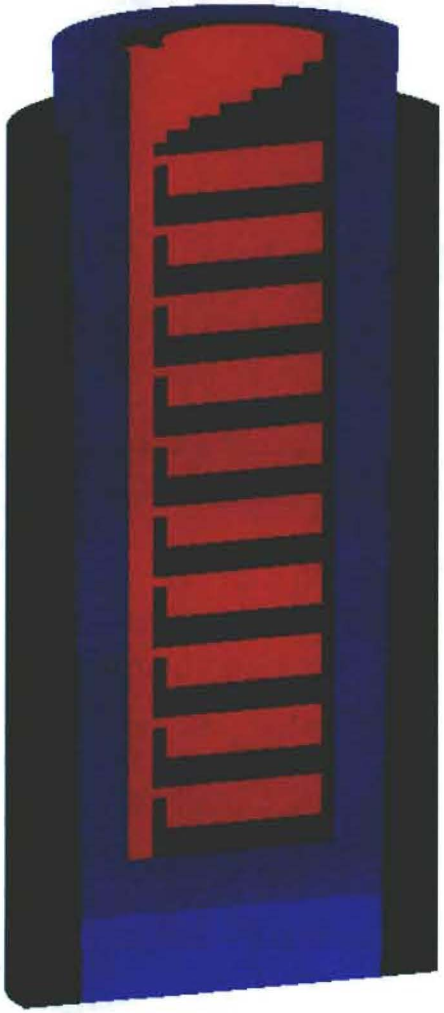


As-polished micrographs reveal microcracking and electro-polished micrographs reveal high temperature retained β phase (marbled microstructure).

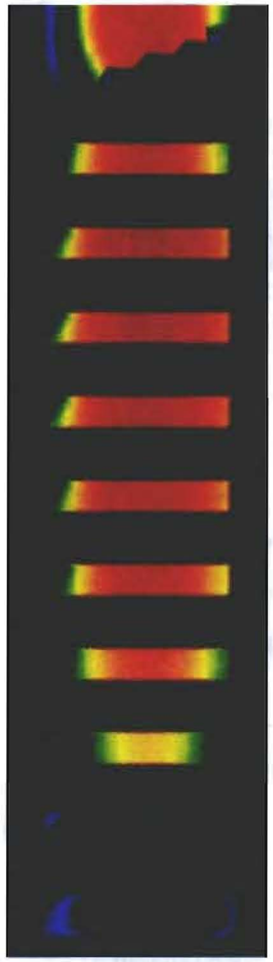
As-cast α-phase Unalloyed Pu

- 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.

Casting Unalloyed Plutonium - Pucks



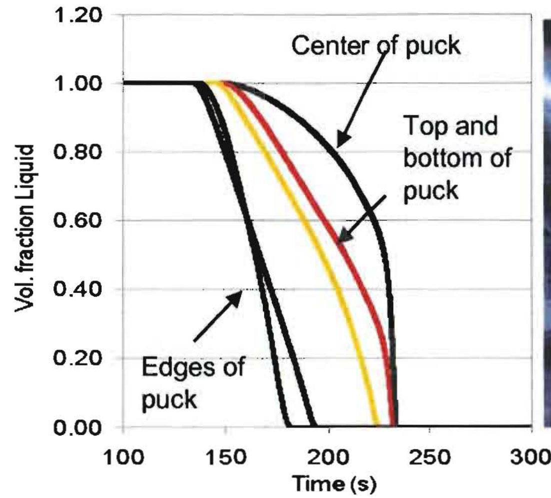
Mold to cast 10 uniform thickness pucks



Blue 180 seconds
Red 230 seconds
Simulation of casting showing time to solidify.

Mold is designed to cast up to 10 uniform or variable thickness pucks

38 mm diameter, 3.2 to 9.6 mm thick
mold temperatures: 600°C
metal temperature at pour: 1000°C

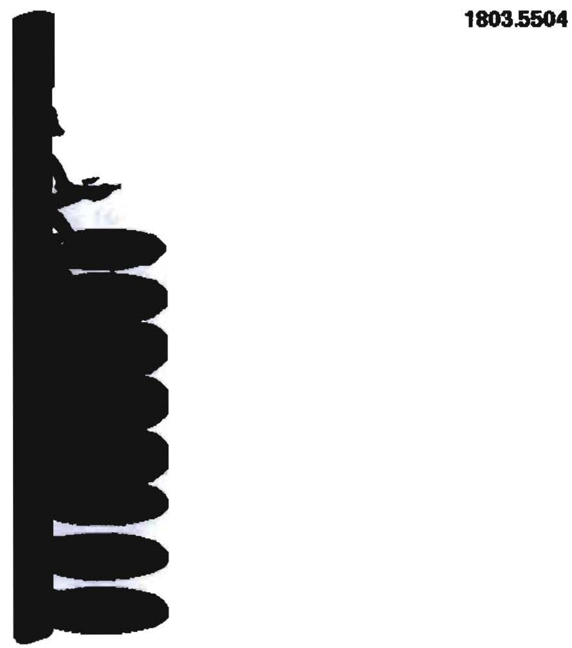


Simulated cooling curves of the middle puck showing volume fraction of liquid.

500 ppm Ga alloyed Pu puck

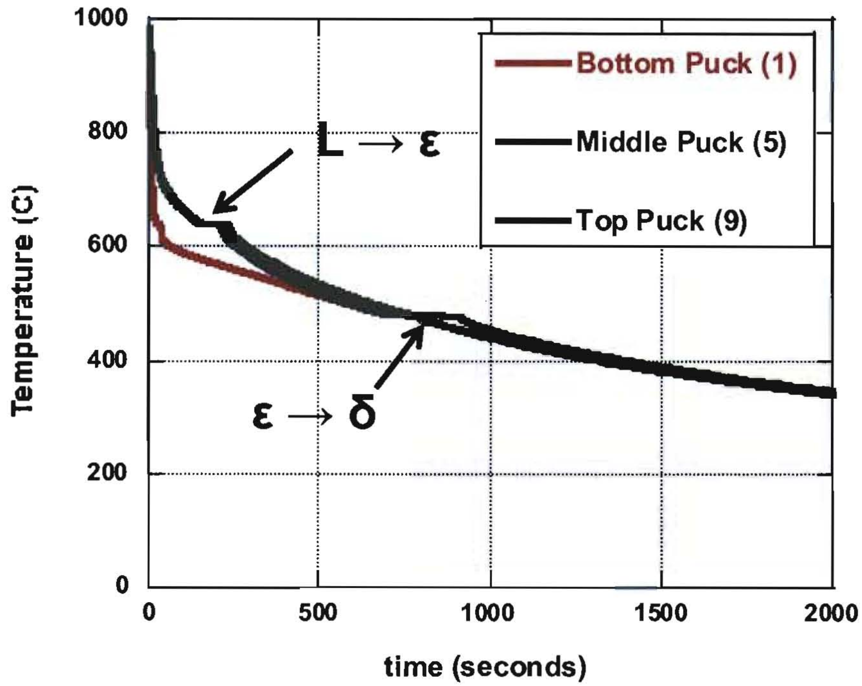
Initial puck densities range from 19.44 to 19.53 gm/cm³.

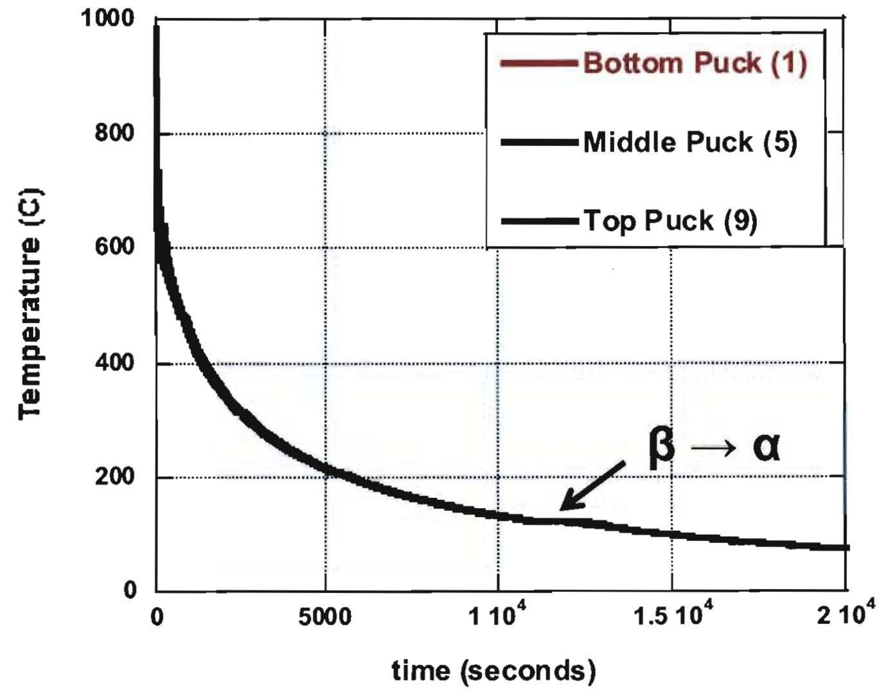
Filling simulation of puck mold



Movie of pucks being filled

Simulation of cooling curves for puck casting

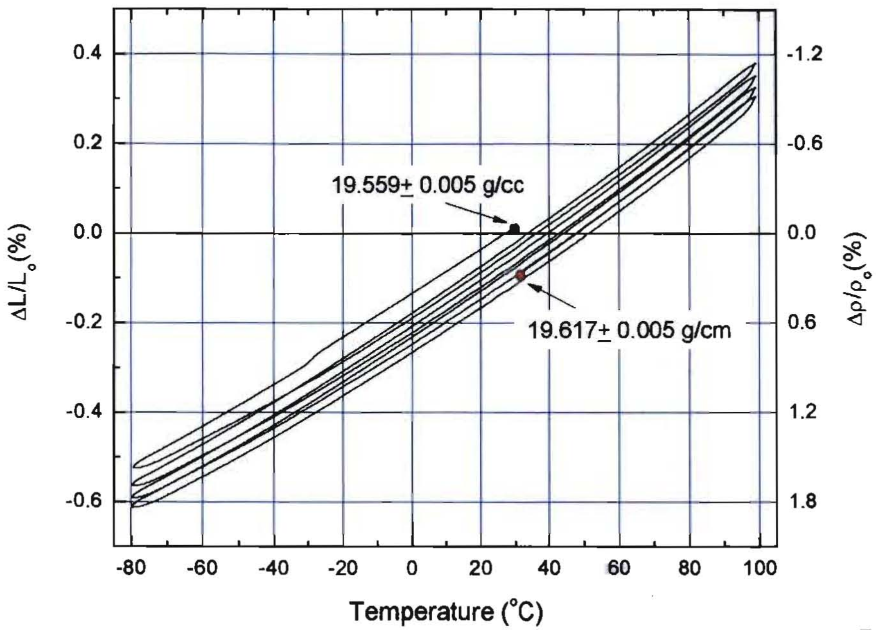




Slow cooling, through all phases
Transforms directionally from edge to center
Top and bottom pucks cool faster than center pucks

Thermal cycling to transform remaining beta phase

Kinetics of β→α Transformation in Cast α-Pu

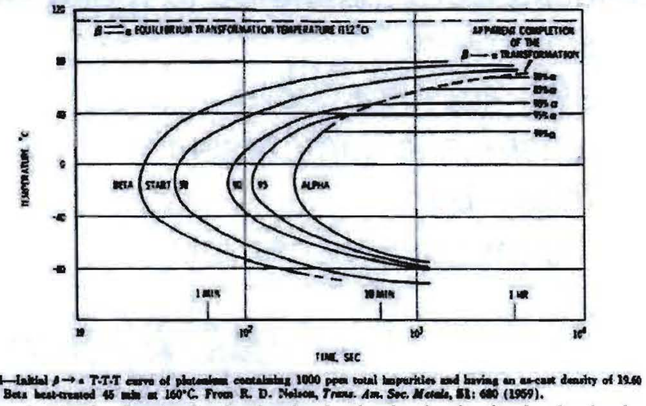


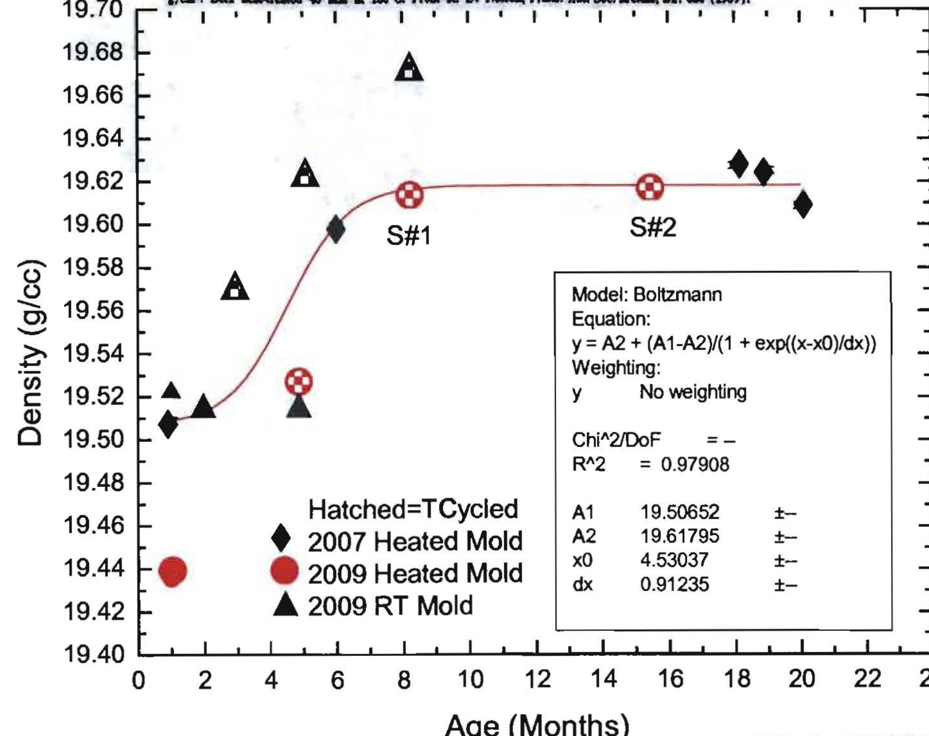
Thermal cycling provides compression stress to drive the β→α transformation:

$$\Delta\sigma_T \approx \alpha_{\alpha Pu} E_{\alpha Pu} \Delta T_{Cycle} - \alpha_{\beta Pu} E_{\beta Pu} \Delta T_{Cycle}$$
$$= (\alpha_{\alpha Pu} E_{\alpha Pu} - \alpha_{\beta Pu} E_{\beta Pu}) \Delta T_{Cycle}$$
$$= 7 \text{x} 10^{-1} GPa$$

10 times the yield stress for β-Pu!

Time-Temperature-Transformation Diagram for β→α





Thermophysical and mechanical Properties

The pucks were machined to make samples for:

- dilatometry
- resonant ultrasound
- quasi-static compression
- Kolsky bar
- 40 mm gas/powder gun

Initial results will be given by Tarik Saleh

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

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.

Coupled modeling/experimental approach

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.

TRUCHAS – LANL developed code

6 rod mold
20.3 mm diameter
108 mm long

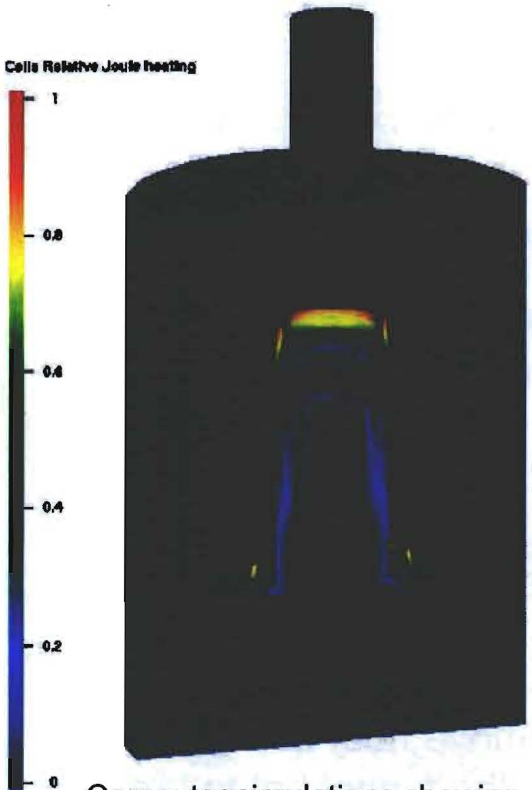
Goal:
Cool from liquid to the $\beta \rightarrow \alpha$ as quickly as possible

Cool slow and directionally through the beta to alpha transition.

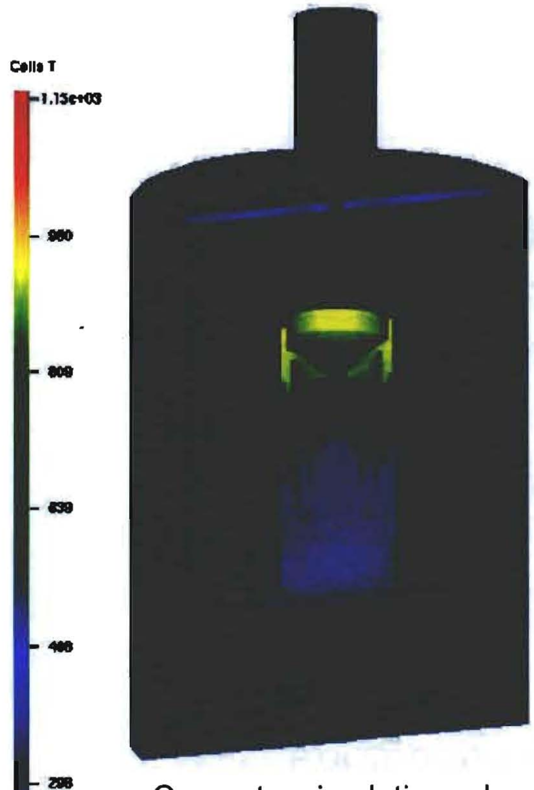
Need to pour into a cold mold to remove heat quickly but keep the funnel hot to prevent cold shuts.

Can we design a mold the will have the desired temperature distribution that is required to cast quality unalloyed Pu?

Heat up simulations of rod mold



Computer simulations showing joule heating in units of W/m^2.



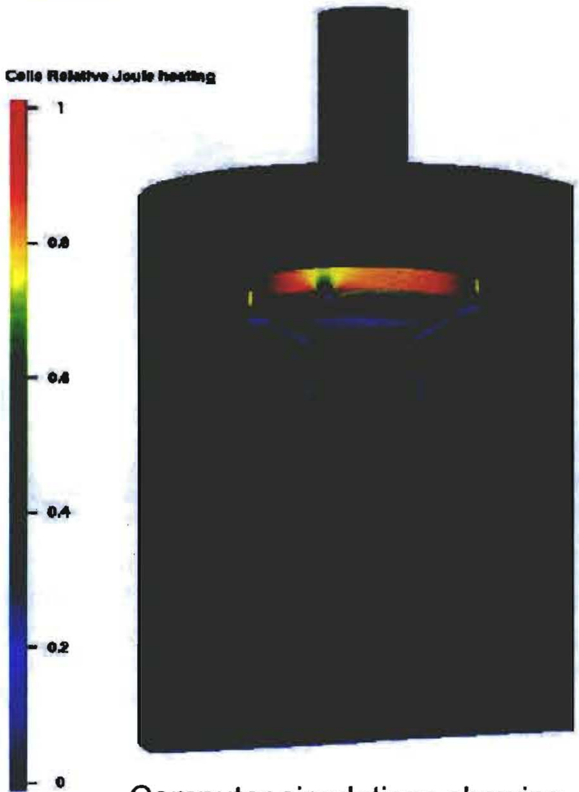
Computer simulations showing temperatures when the funnel outlet reaches 550°C.

TRUCHAS models for preheat studies

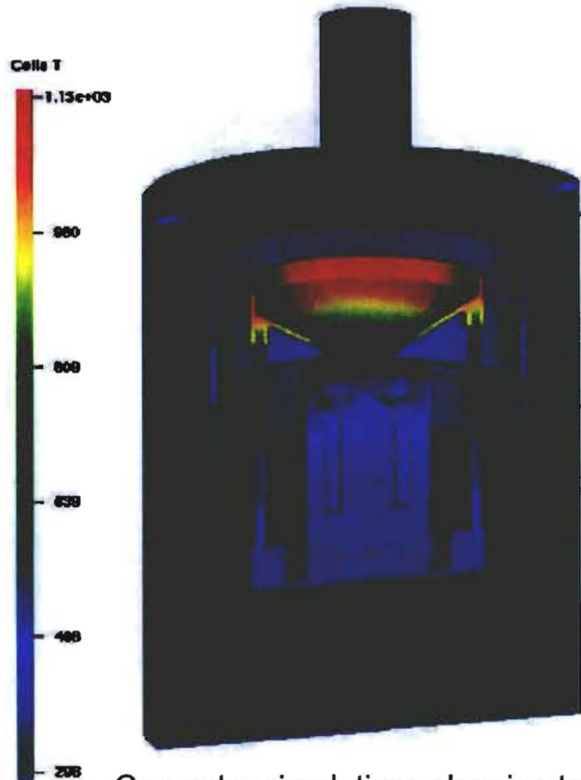
Electromagnetics
Radiative heat transport with view factors
Conductive heat transport

Initial design in which the funnel is supported by the mold.
The funnel and mold are too closely connected in this design making it difficult to control the temperature in the mold.

Heat up simulations of rod mold



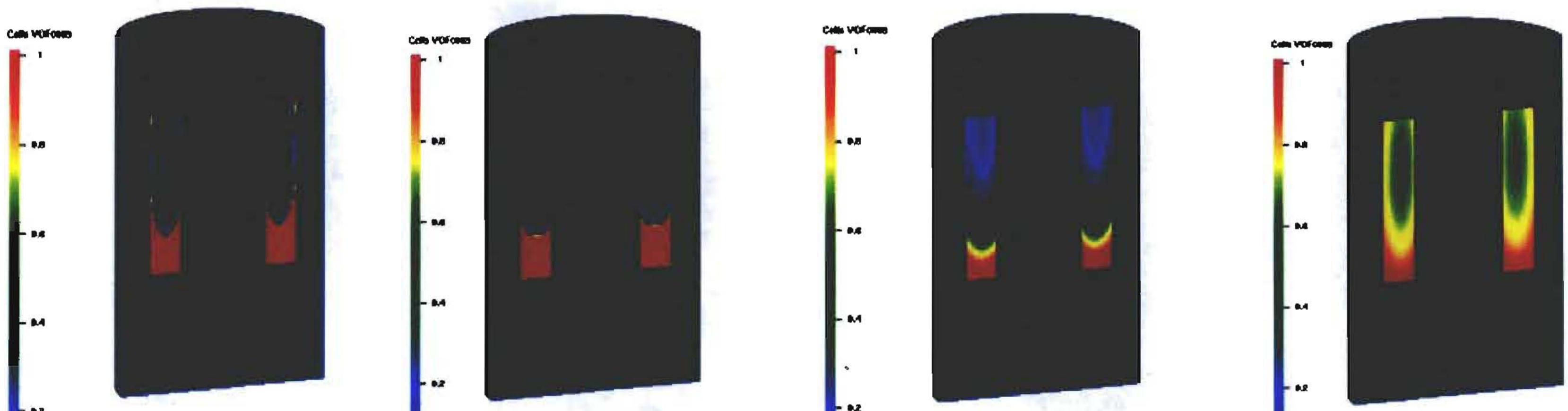
Computer simulations showing joule heating in units of W/m².



Computer simulations showing temperatures when the funnel outlet reaches 550°C.

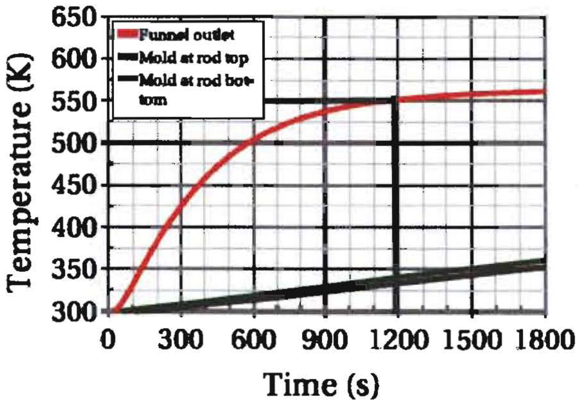
New design in which the funnel is supported by ceramic rods.
With the new design, the funnel couples with the induction field allowing better temperature control in the mold.

Effect of mold preheat on β to α transformation

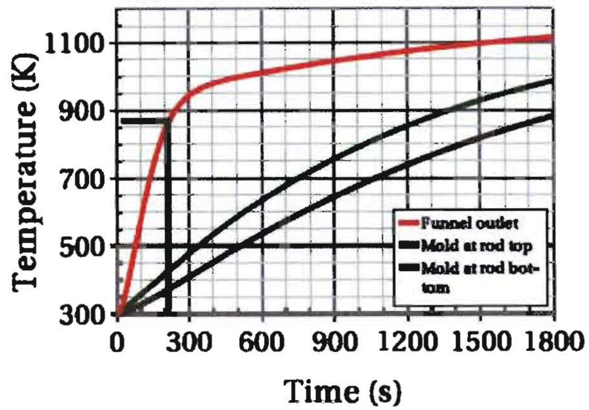


Process conditions	Starting from a cold mold. Cooling rate before the beta formation was approximately 1.6K/s.	Starting from 1000A, 150s heatup. Cooling rate before the beta formation was approximately 0.4K/s.	Starting from 1000A, 300s heatup. Cooling rate before the beta formation was approximately 0.04K/s.	Starting from 1000A, 450s heatup. Cooling rate before the beta formation was approximately 0.03K/s.
β to α transformation behavior	Radially directional, rapid (under 10 minutes to onset of alpha formation)	Axially directional, moderate (approximately 1 hour to the onset of alpha formation)	Non-directional, slow (approximately 2 hours to the onset of alpha formation)	Non-directional, very slow (on the order of 10 hours to the onset of alpha formation)

Heat up scenario of rod mold



a) 250A setting. Ultimate temperature is 575K or 200°C



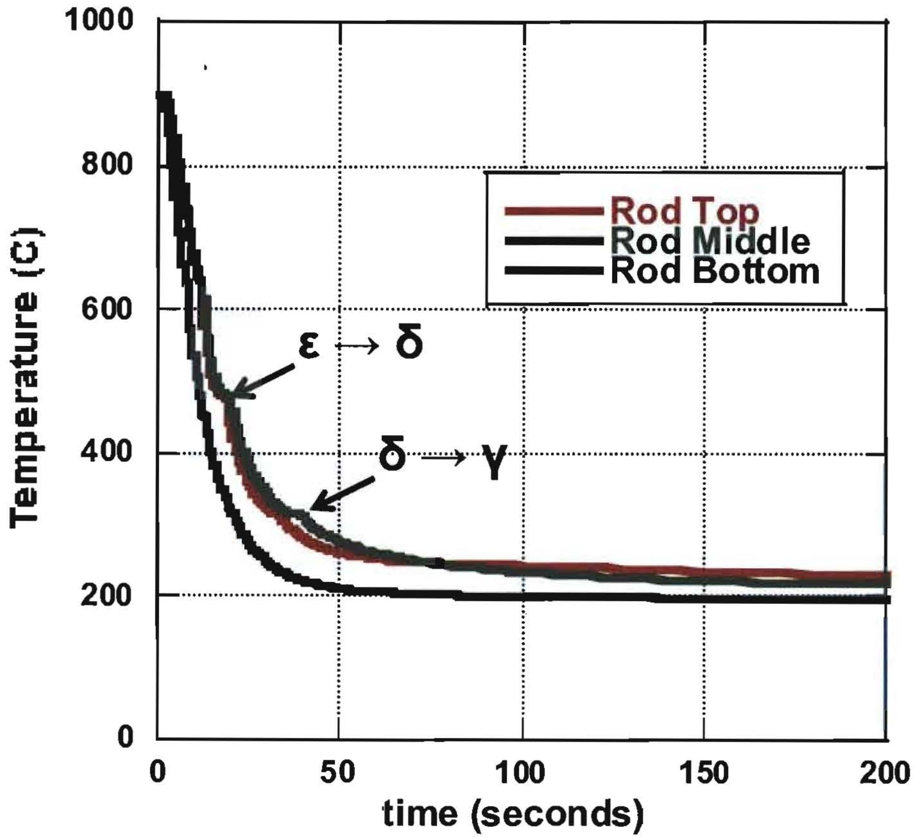
b) 1000A setting. Ultimate temperature is 1150K or 875°C

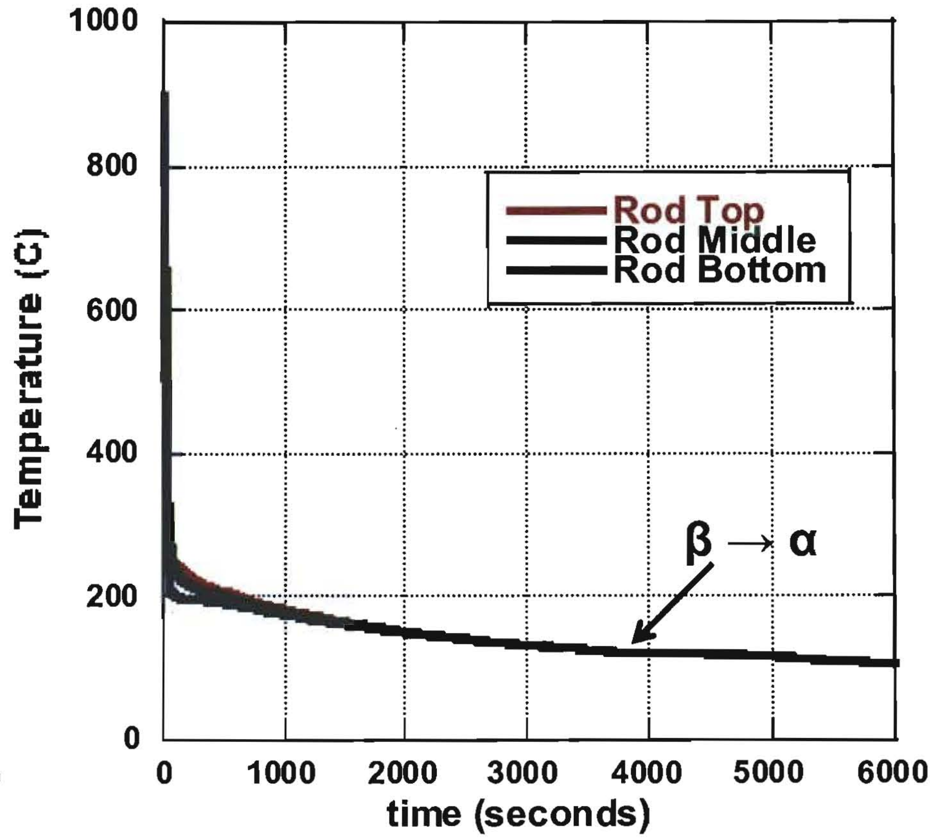
Based on computer simulations, the optimal mold heat up scenario involves a low-current (250A), long-time (1200 second) initial phase which heats the funnel without heating the mold significantly.

Then apply a high-current (1000A), short time (200 second) phase that rapidly brings the funnel outlet to the final temperature of 600°C.

By using similar methods, it was determined that a metal temperature of 900°C provide the most favorable cooling without a risk of freezing in the funnel.

Simulation of cooling curves using selected parameters



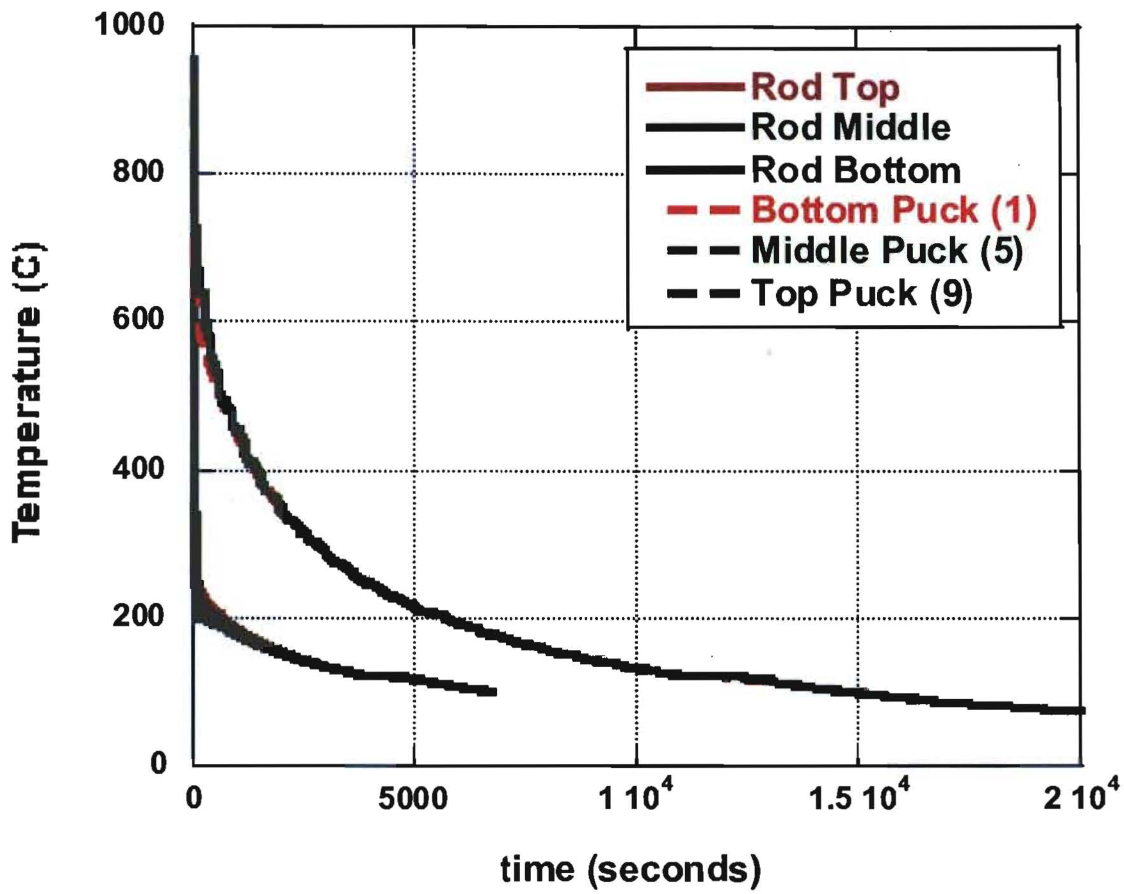


Initial cooling is rapid to ~ 225°C

γ → β transition is at ~215°C

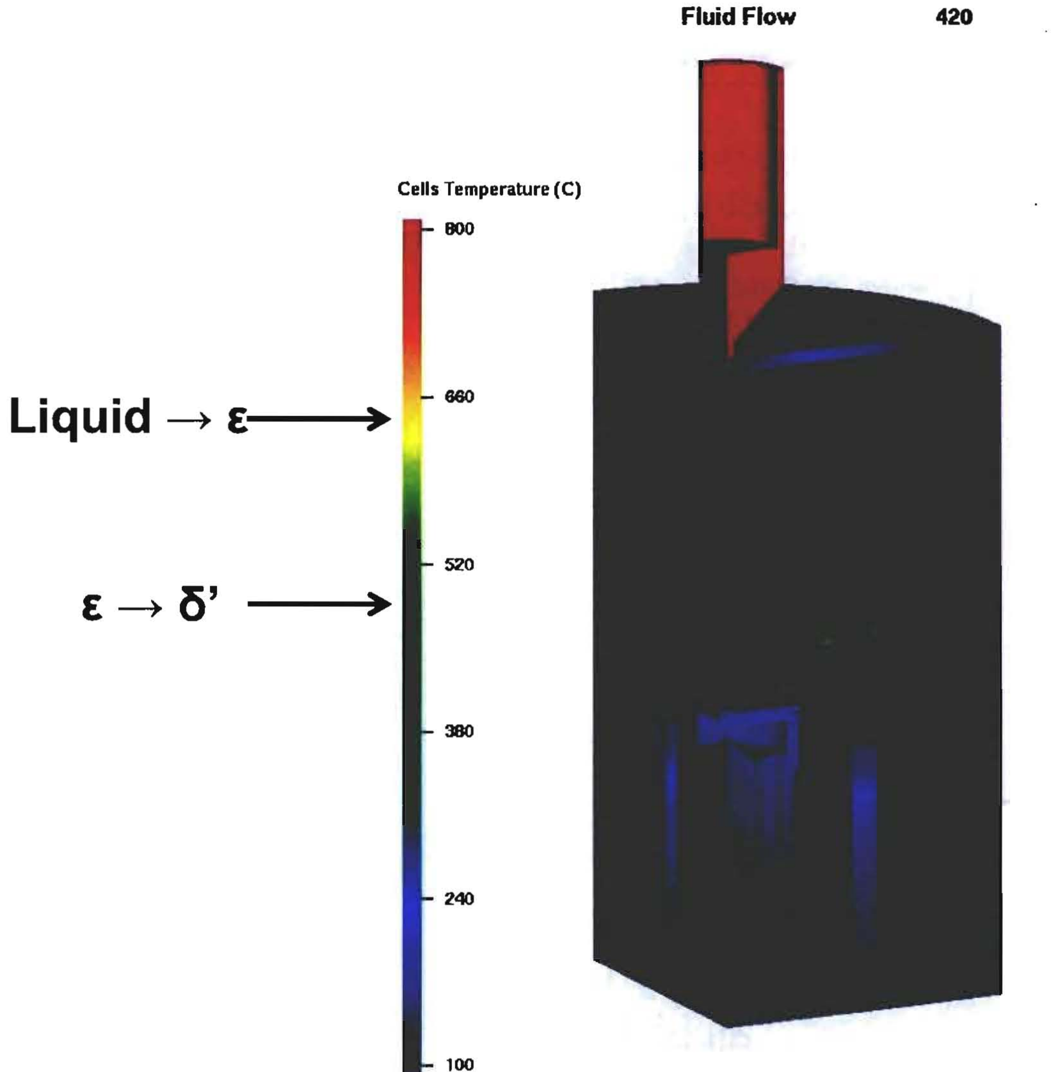
Subsequent cooling is slow through the beta phase and the beta to alpha transformation.

Comparison of simulated of cooling curves



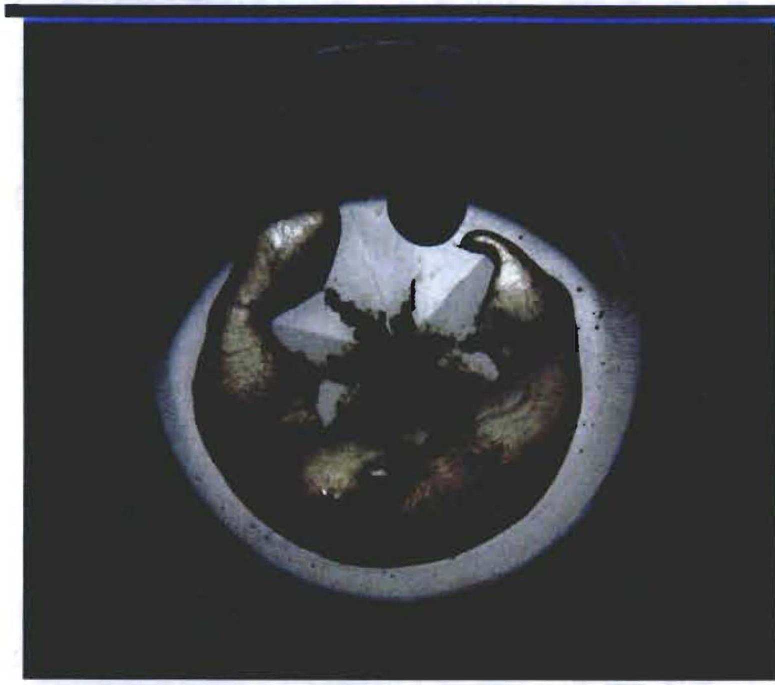
The pucks cool much slower through all phase transformations.

Filling simulation of rod mold



Movie of rods being filled

Casting of 500ppmGa 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 metal contained 500 ppm Ga.

The casting was very non-symmetric. The cause is still unknown.

Initial as-cast density measurement from the first rod was 19.6 gm/cm^3 indicating a good casting. The rod will now be thermal cycled to transform any retained beta phase. The material will be characterized as-cast and post thermal treatment.

Conclusions and Future Work

Conclusions

- Single phase unalloyed α-Pu is difficult to produce due to microcracking and retained β-Pu, but computer simulations help drive mold and process design and subsequent thermal cycling and/or long term soak at ambient temperatures reduces retained β-Pu;
- Equilibrium thermodynamics is not sufficient to define phase transformations in plutonium - kinetics, microstructure, and impurity composition are important;

Future Work

- Cast, fabricate and characterize microstructure of high density unalloyed Pu with and without Ga.
- Measure temperature dependence of thermal expansion, specific heat, and elastic moduli for Ga soluble (~0.15 at%) in α-Pu
- Measure thermomechanical properties of low-alloy Pu.