LA-UR-12-23441

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Title: Co-Design: Fabrication of Unalloyed Plutonium

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Intended for: MDI Summer Research Group Workshop Advanced Manufacturing, 2012-07-25/2012-07-26 (Los Alamos, New Mexico, United States)

Co-Design: Fabrication of Unalloyed Plutonium

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MDI Summer Research Group Workshop

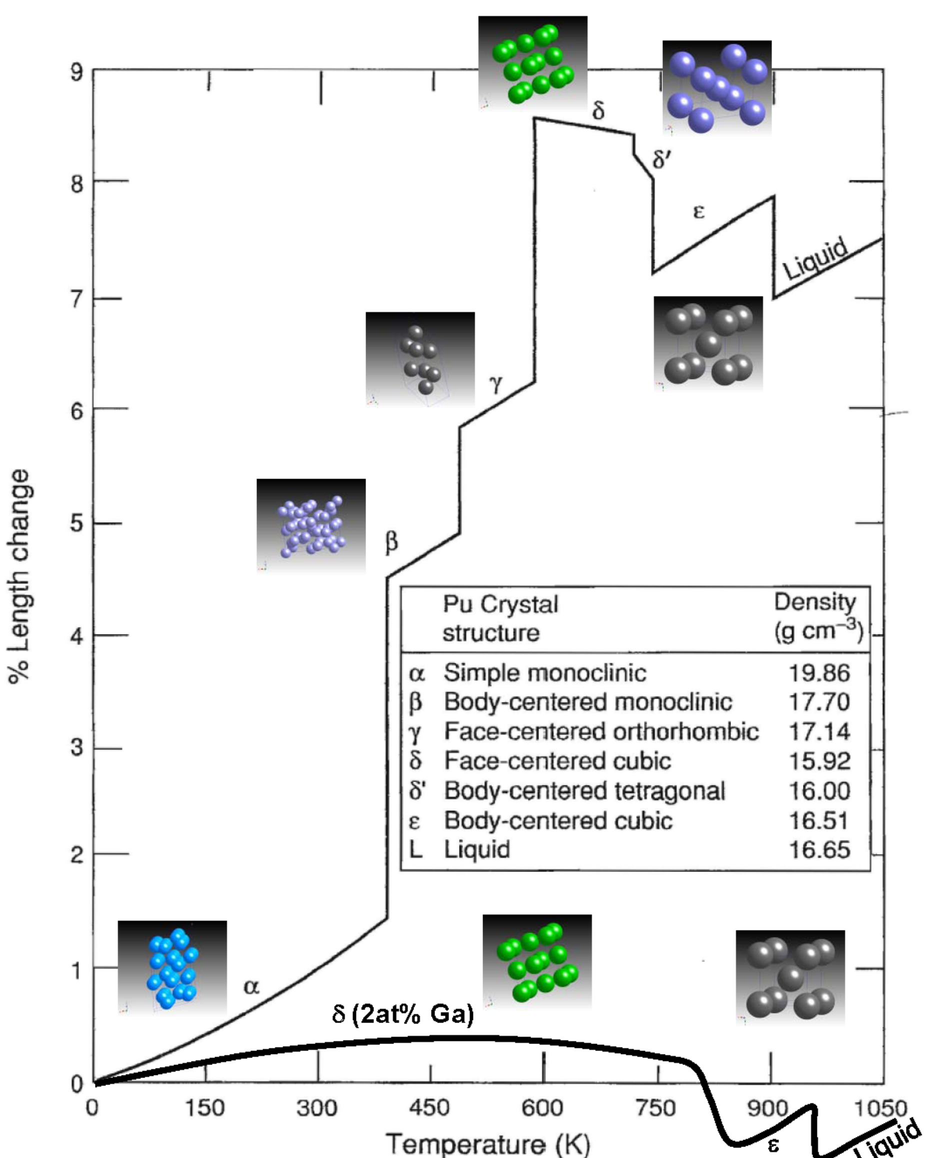
Advanced Manufacturing

July 25 - 26, 2012

Abstract

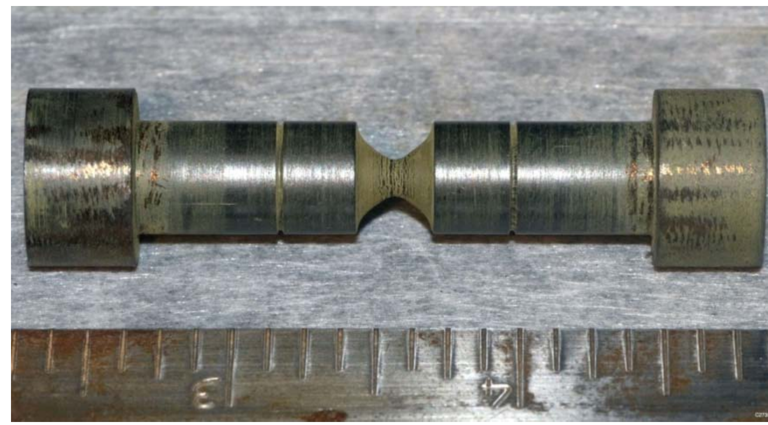
The successful induction casting of plutonium is a challenge which requires technical expertise in areas including physical metallurgy, surface and corrosion chemistry, materials science, electromagnetic engineering and a host of other technologies all which must be applied in concert. Here at LANL, we are employing a combined experimental and computational approach to design molds and develop process parameters needed to produce desired temperature profiles and improved castings. Computer simulations are performed using the commercial code FLOW-3D and the LANL ASC computer code TRUCHAS to reproduce the entire casting process starting with electromagnetic or radiative heating of the mold and metal and continuing through pouring with coupled fluid flow, heat transfer and non-isothermal solidification. This approach greatly reduces the time required to develop a new casting designs and also increases our understanding of the casting process, leading to a more homogeneous, consistent product and better process control. We will discuss recent casting development results in support of unalloyed plutonium rods for mechanical testing.

Plutonium: a fascinating and frustrating element



	Pu Crystal structure	Density (g cm^{-3})
α	Simple monoclinic	19.86
β	Body-centered monoclinic	17.70
γ	Face-centered orthorhombic	17.14
δ	Face-centered cubic	15.92
δ'	Body-centered tetragonal	16.00
ε	Body-centered cubic	16.51
L	Liquid	16.65

Unalloyed plutonium is difficult to cast due to the 25% volume contraction during the delta to alpha transformations. Casting defects include porosity, microcracking, and macrocracking to the point of catastrophic splitting.



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

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

LANL history of casting unalloyed plutonium

1960's 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

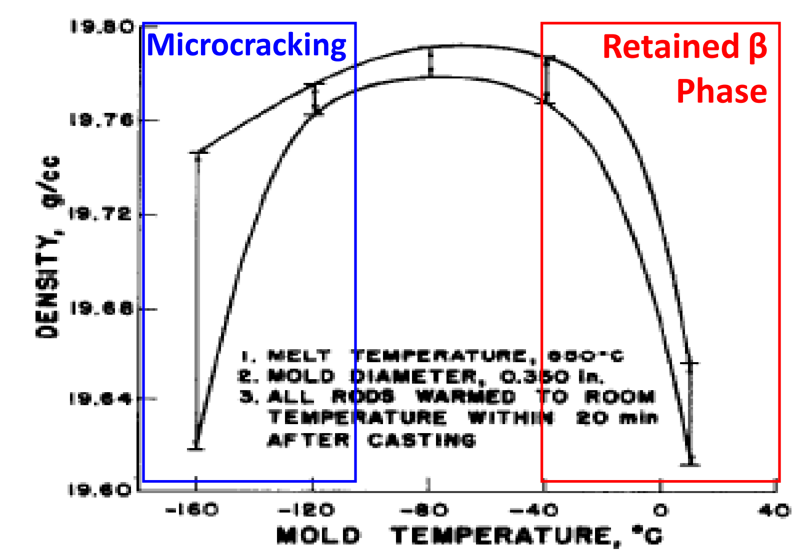
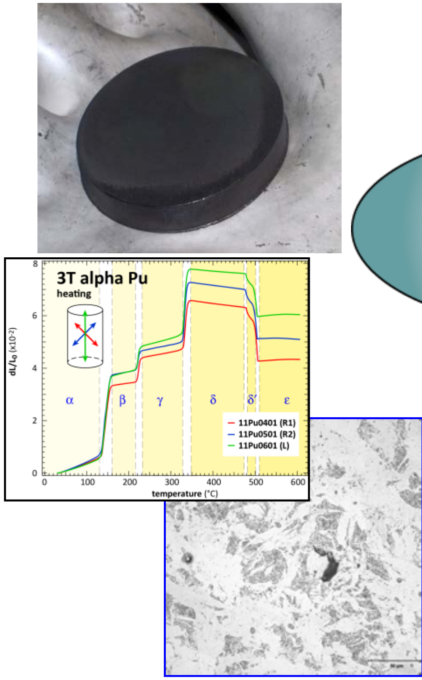


Fig. 3. Effect of mold temperature on ultimate densities of Pu rods.

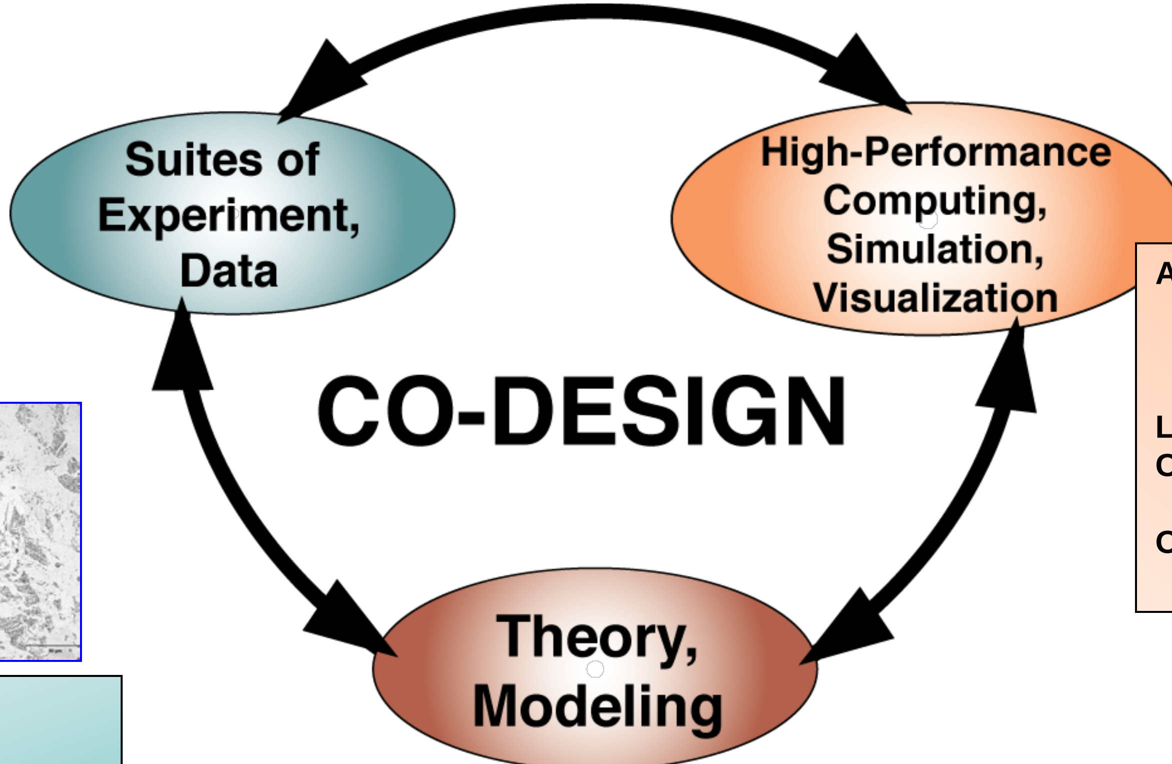
Two conclusions:
1)Rapid cooling, transform directly from ε to α, ε is quite ductile and yields, allowing significant contraction without cracking
2)Transform directionally to α, (edge to center) little or no cracking occurs

LANL currently does not have a chill casting capability. We need to develop an alternate method of achieving the same results.
GOAL: design a mold and process parameters that will have the desired temperature distribution to cast quality unalloyed Pu.

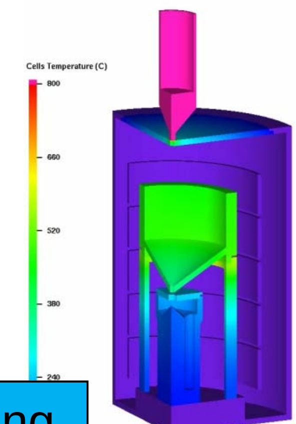
A Co-design Approach to Plutonium Casting Design



Pu castings
Metallography
Physical property data



ASC codes
TRUCHAS (LANL)
CUBIT (SNL)
GMV (LANL)
LANL supercomputers
Commercial codes
FLOW-3D
Open source code
ParaView



Use expert knowledge for initial design. Then use Truchas and FLOW-3D to run through design options and process parameters

Casting modeling represents an informed method for predicting and understanding cast component characteristics.

Development of simulation tools

Typical commercial casting codes offer the following capabilities:

fluid flow with free surface flow
heat transfer with phase change
radiative heat transfer to a T0

TRUCHAS: A simulation tool for casting and other manufacturing operations of interest to the DOE complex.

large scale 3D simulations on unstructured meshes
electromagnetic Joule (induction) heating
solid mechanics contact algorithm
view factor radiative heat transport
phase change strains in solid mechanics

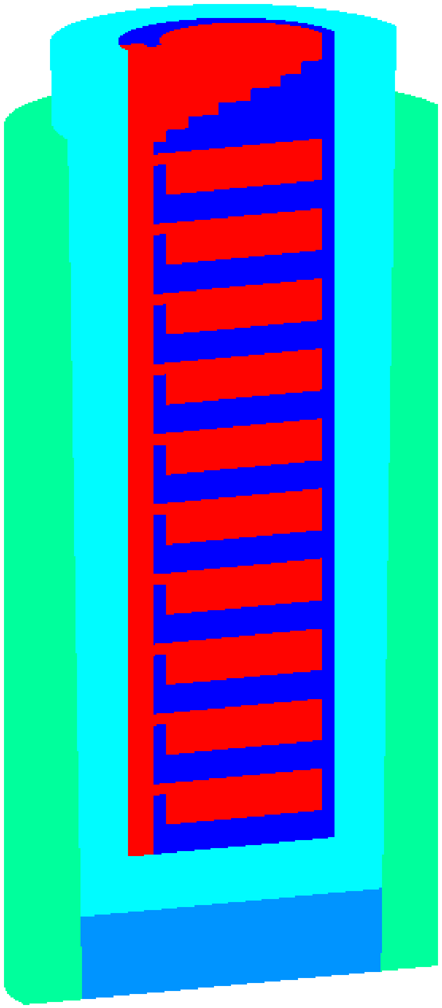
Materials Property data

limited high temperature data is available for Pu-Ga alloys, especially strength data
performed sensitivity studies on variables using TRUCHAS

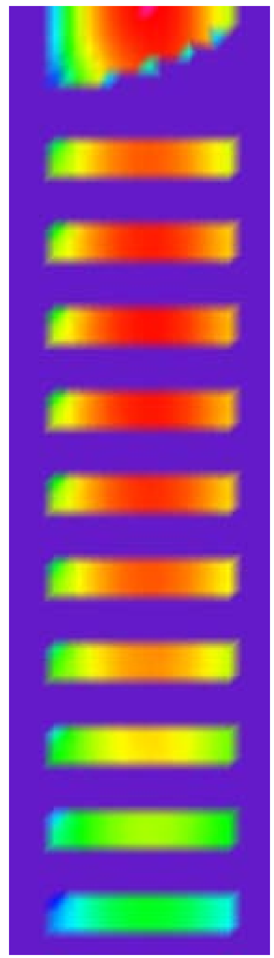
Perform many computer "experiments" and a few actual experiments.
Predict and control microstructures.
Enable rapid development of optimized casting processes.
Perform highly instrumented castings to provide data for process modeling verification.

Casting unalloyed Plutonium - Pucks

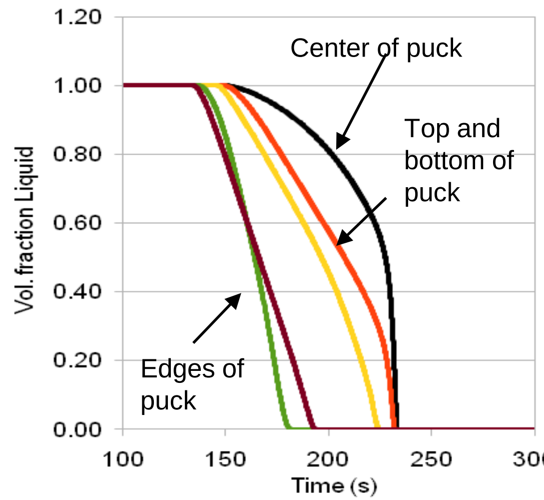
38 mm diameter, 3.2 to 9.6 mm thick
mold temperatures: 600°C
metal temperature at pour: 1000°C



Mold to cast 10 uniform thickness pucks



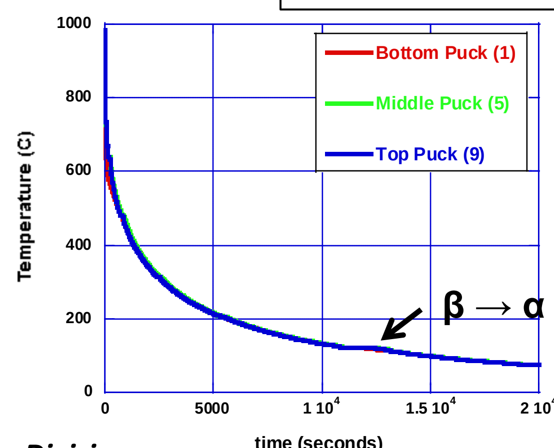
Blue 180 seconds
Red 230 seconds
Simulation of casting showing time to solidify.



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

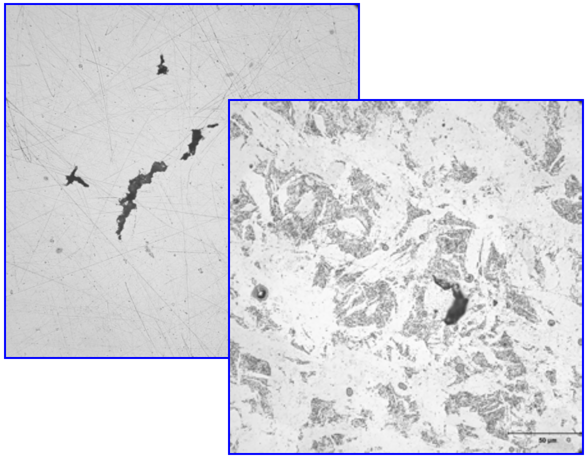
500 ppm Ga alloyed Pu puck

Initial puck densities range from 19.44 to 19.53 gm/cm^3.

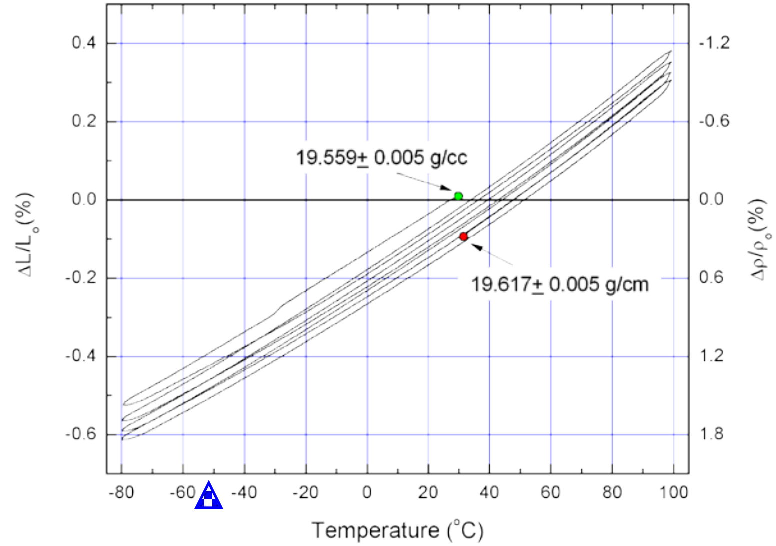


Microcracking and Retained β Phase in cast pucks

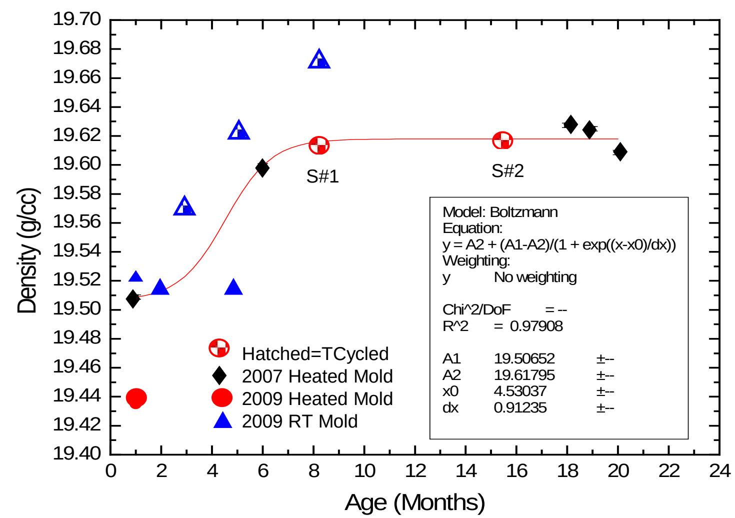
Pucks cast in heated graphite mold



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



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



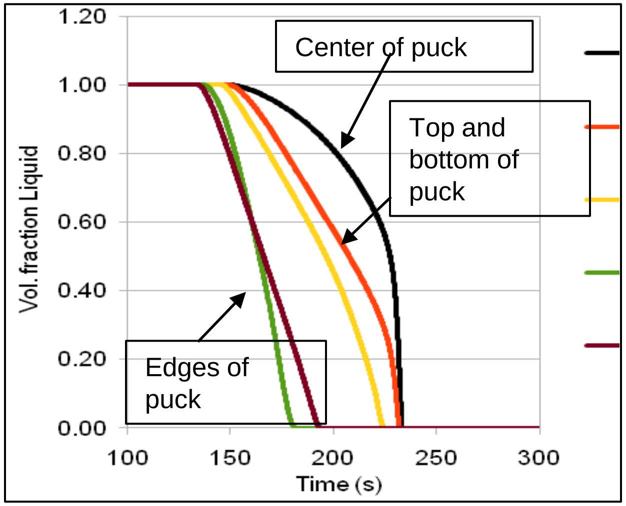
<u>As-cast α-phase unalloyed Pu pucks</u>

- 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.

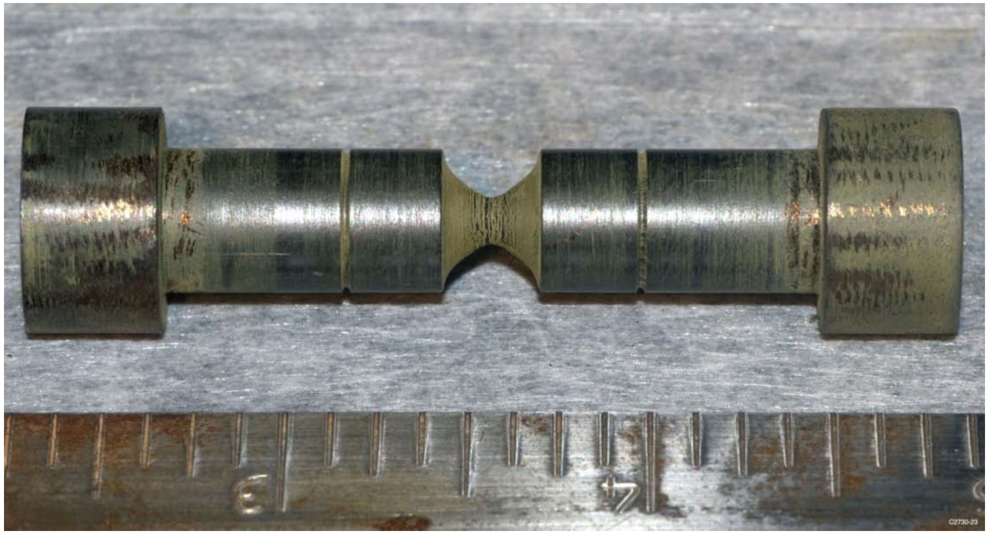
Alpha plutonium for mechanical testing specimens

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.



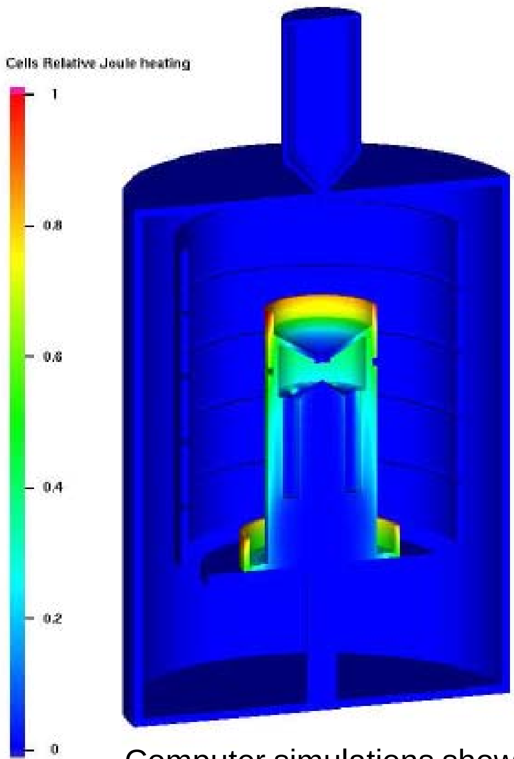
Simulated cooling curves of the middle puck showing volume fraction of liquid.



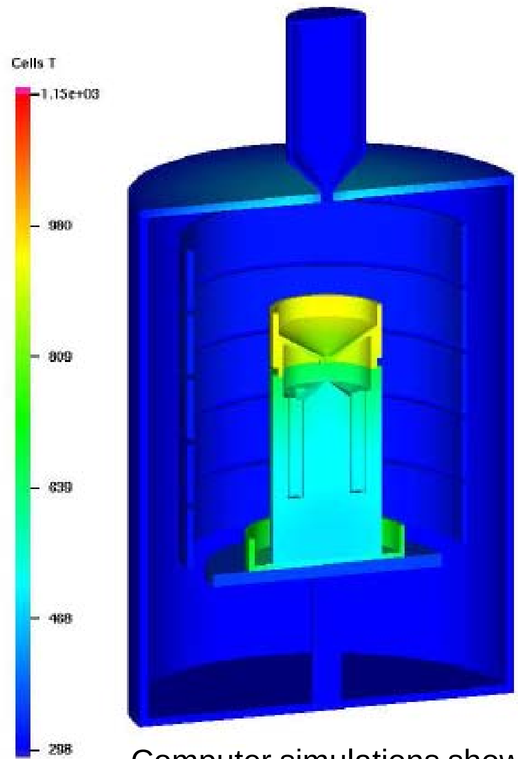
“D” notch 3T (triaxial) specimen machined from delta stabilized Pu.
15.88 mm diameter X 63.5 mm long

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

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.

6 rod mold
20.3 mm diameter
108 mm long

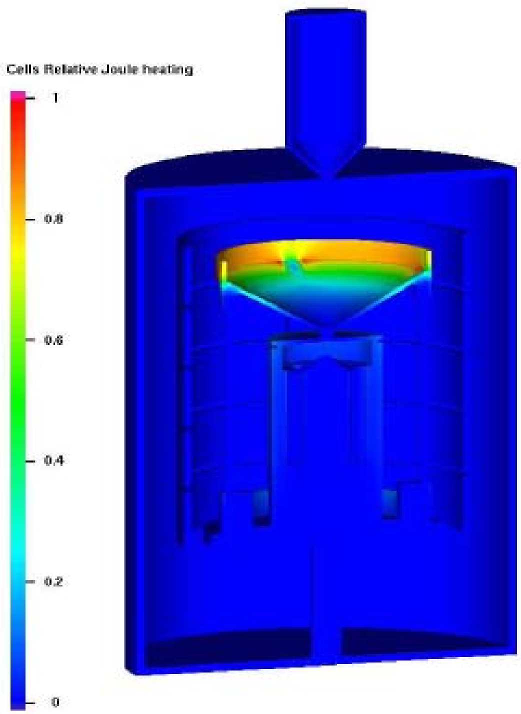
Goal:
Cool from liquid to the β → α as quickly as possible

Cool slow and directionally through the beta to alpha transition.

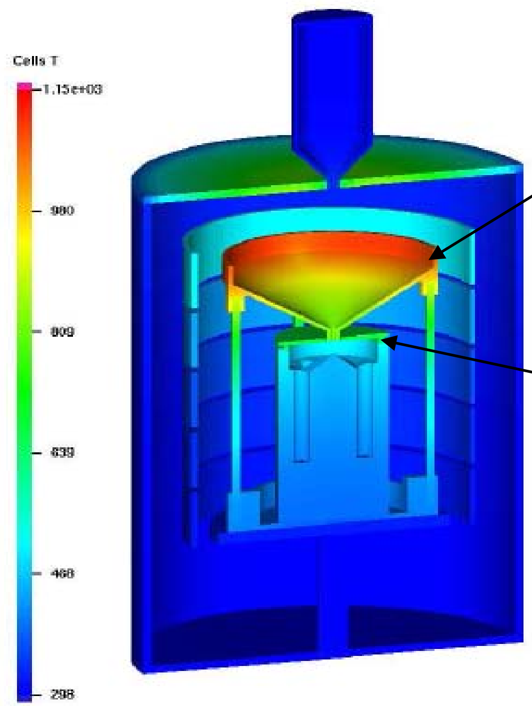
Need to pour into a cold mold to remove heat quickly but keep the funnel hot to prevent cold shuts.

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 re-designed rod mold



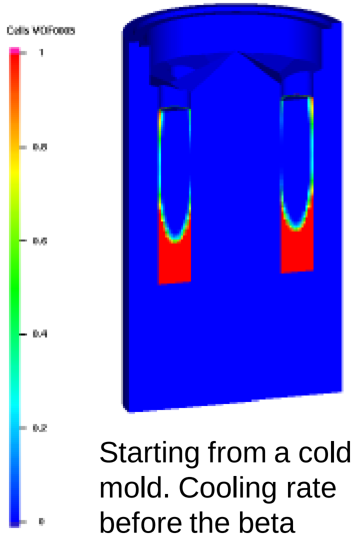
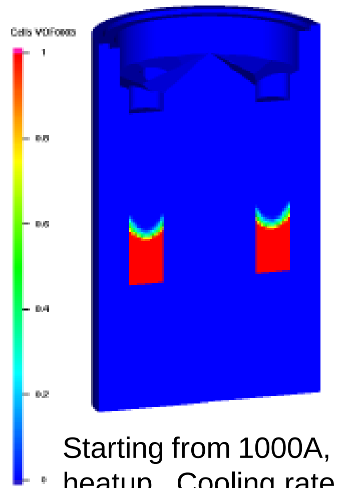
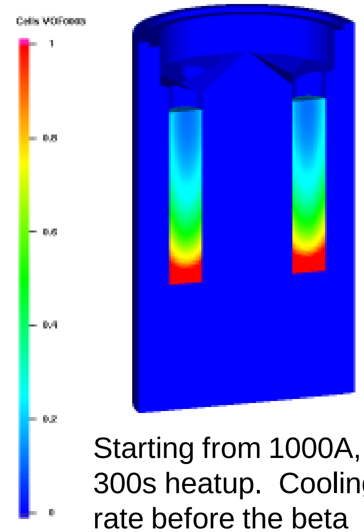
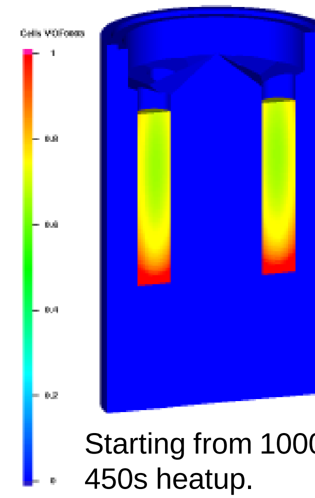
Computer simulations showing joule heating in units of W/m^2.



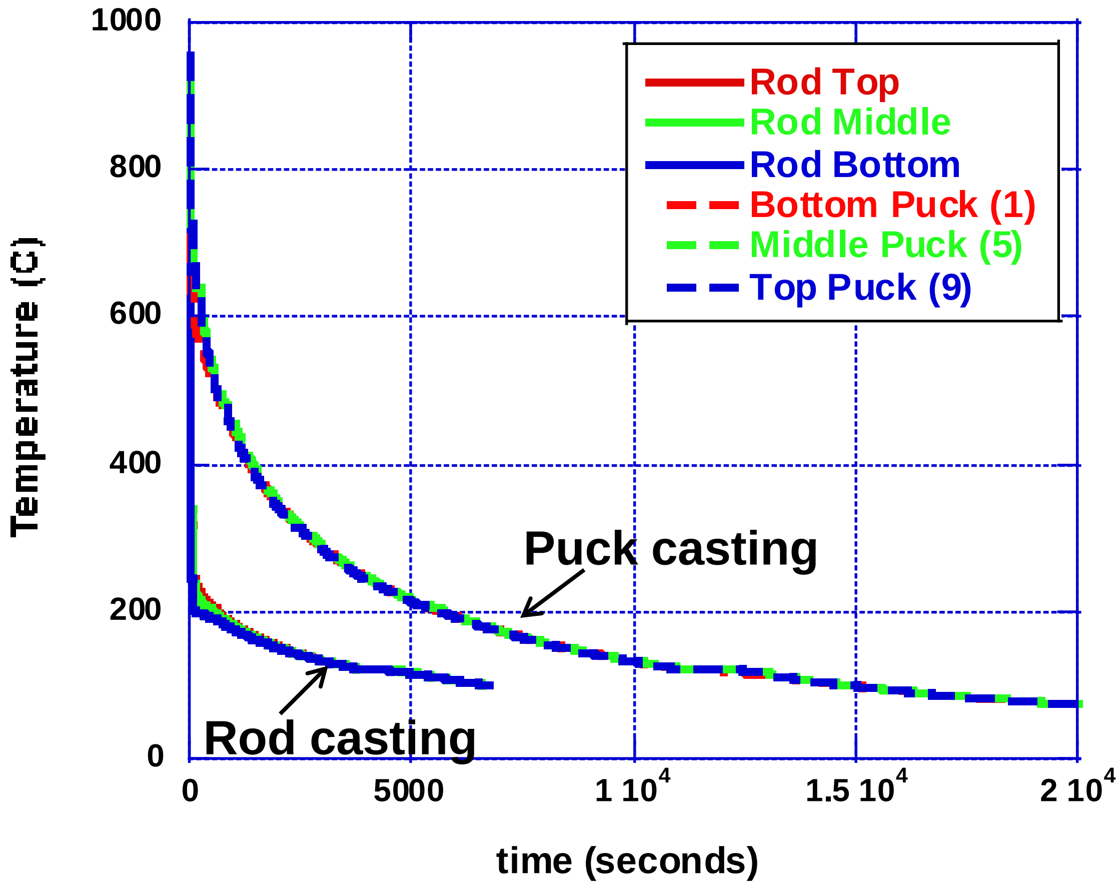
Computer simulations showing temperatures when the funnel outlet reaches 550°C.

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

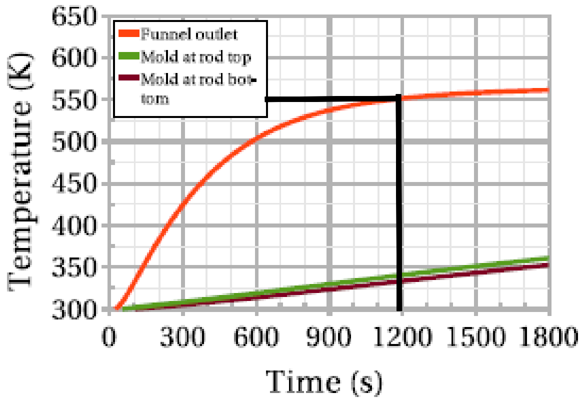
				
Process conditions	Starting from a cold mold. Cooling rate before the beta formation was approximately 1.6K/s.	Starting from 1000A, 150s heatup. Cooling rate before the beta formation was approximately 0.4K/s.	Starting from 1000A, 300s heatup. Cooling rate before the beta formation was approximately 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)

Comparison of simulated of cooling curves

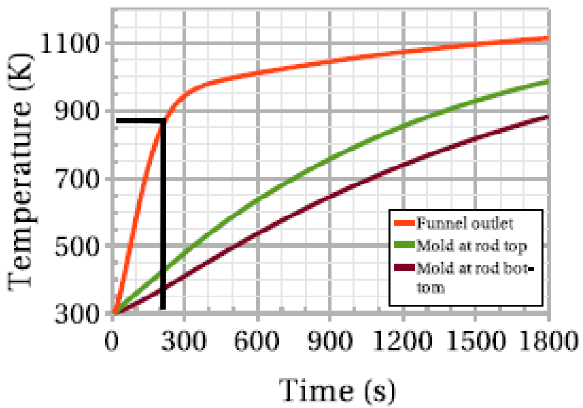


The pucks cool much slower through all phase transformations.

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.

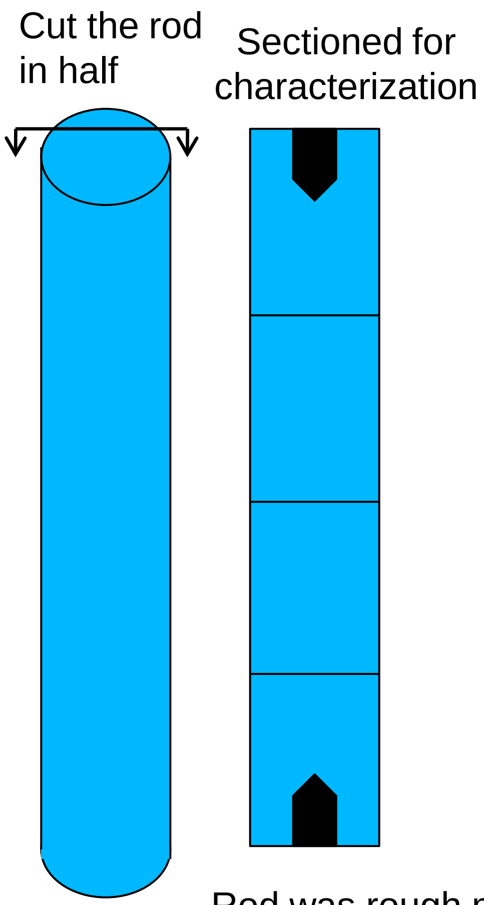
Casting of low Ga alloyed Pu in rod mold

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

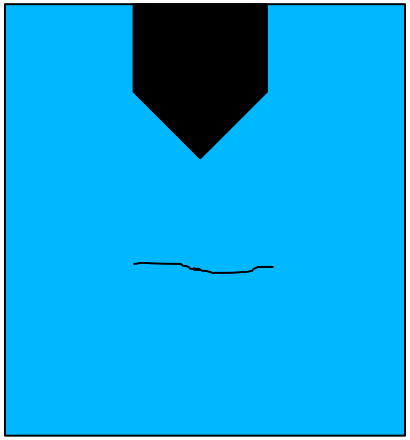


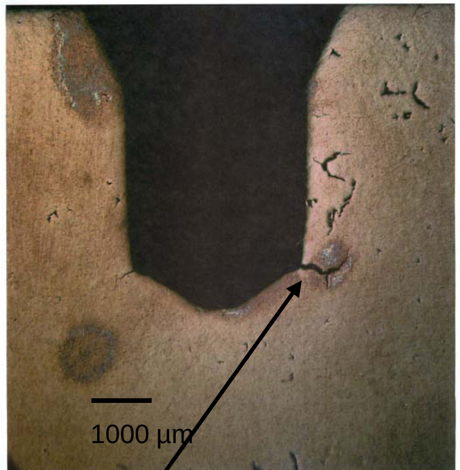
Rod was rough machined
to clean up outer surface
Center drilled for machining

One half :
Metallographic analysis of all sections

Other half:
Thermo dynamic properties on samples from middle section:
Dilatometry
Differential Scanning Calorimetry
quasi static specimens
Three remaining sections reserved for thermal cycling studies (not yet performed)

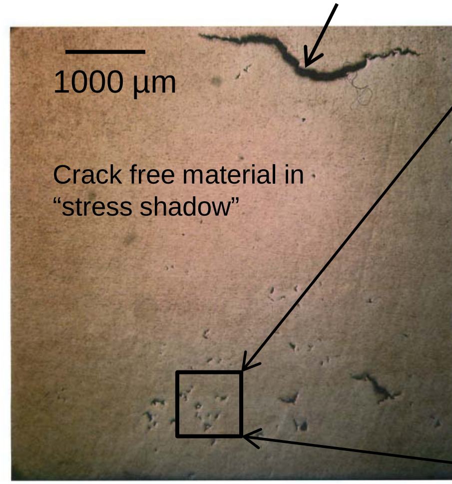
Metallographic characterization

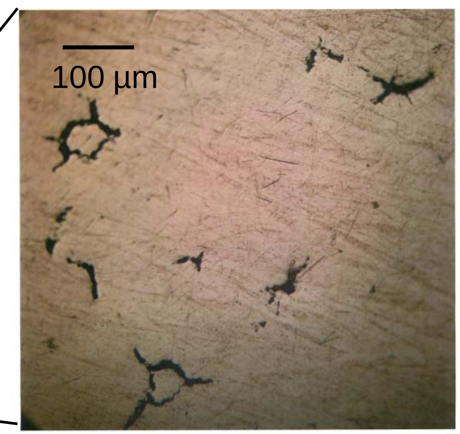




Machining induced cracks:
The center drilling operation heated the material into the beta phase. Upon subsequent cooling, cracks developed during the transformation back to the alpha

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

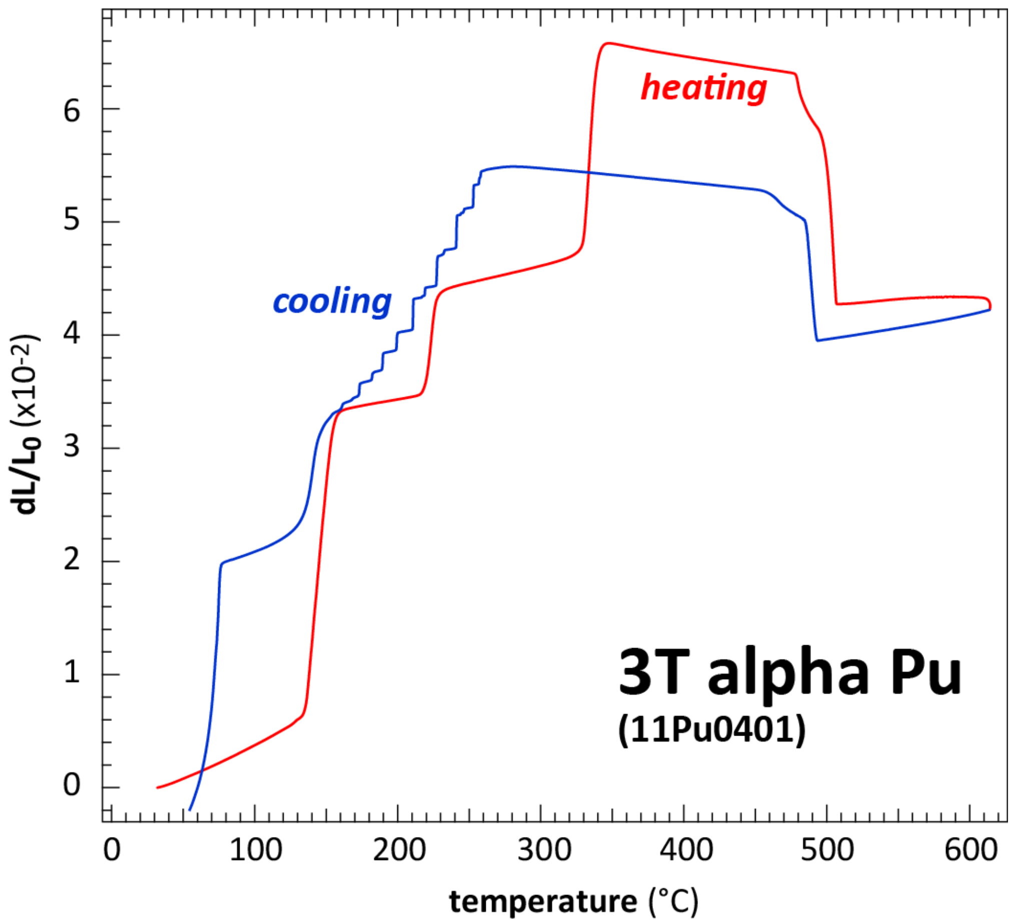




Cracks form during beta to alpha transformation most likely nucleating at triple points.

300 ppm Ga alloyed Pu rod

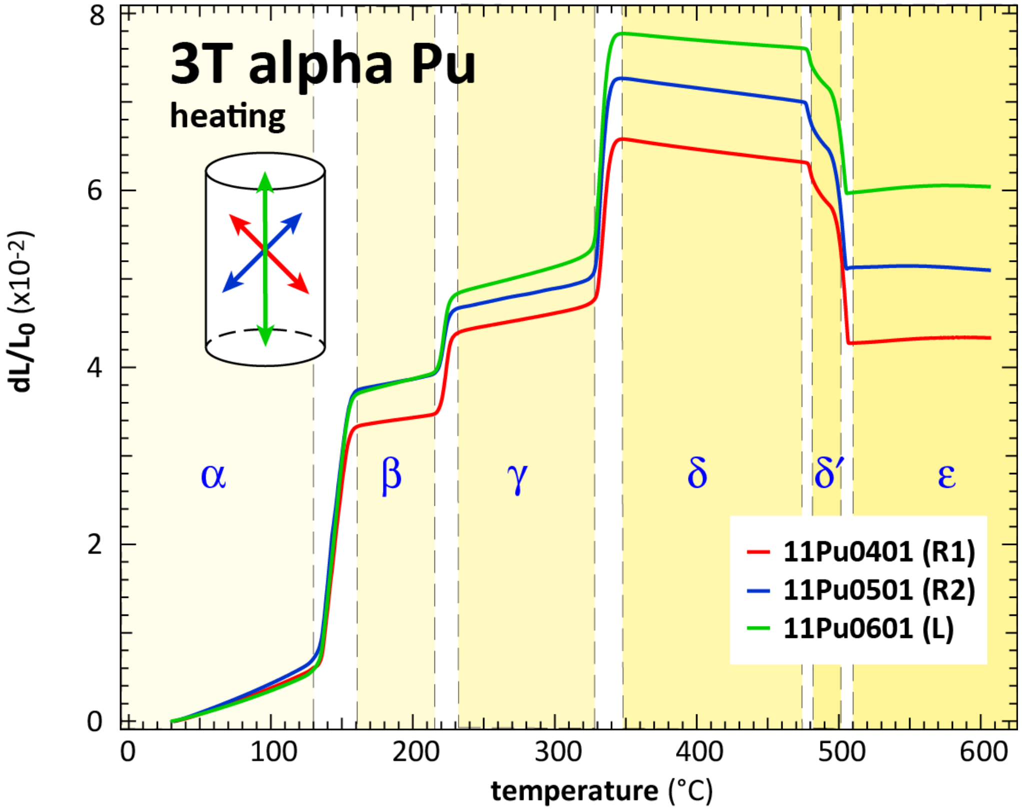
Dilatometry



- 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

300 ppm Ga alloyed Pu rod

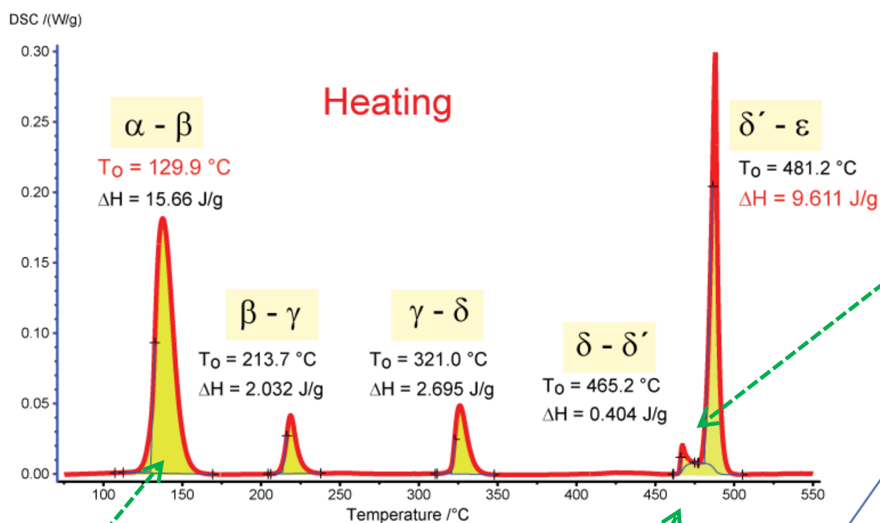
Thermal expansion differences in cast rod



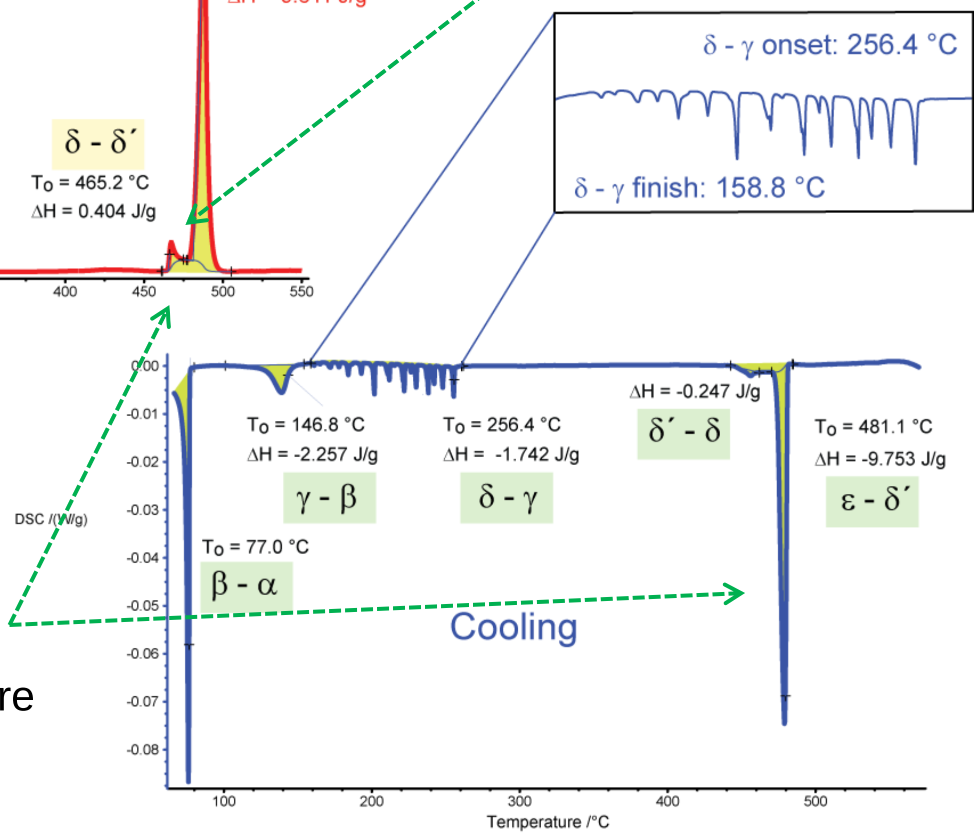
- three samples were cut from rod
- each dilatometry run represents a different orientation within the rod
- differences in curves probably reflect preferred orientation caused by anisotropic cooling in mold

300 ppm Ga alloyed Pu rod

Differential Scanning Calorimetry



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



- **Ga content shifts α → β transformation to higher T_o**

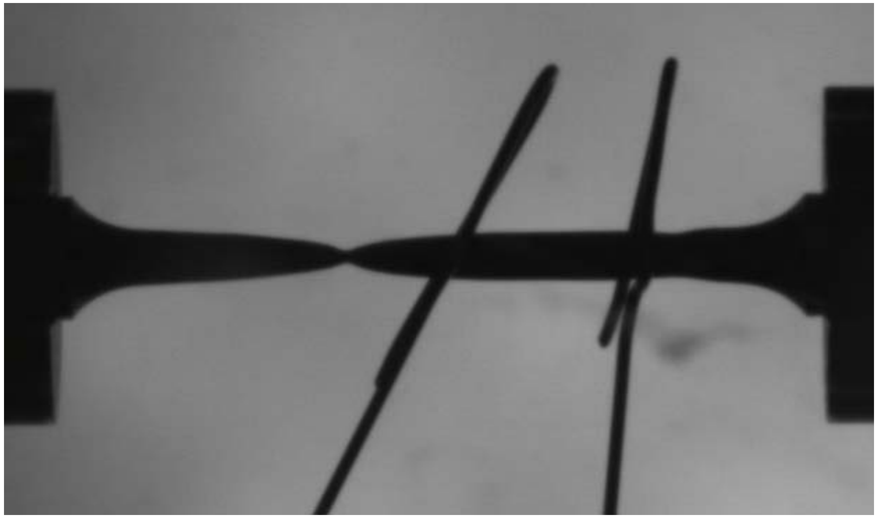
  130°C vs. 126°C

- **Some unexpected features**

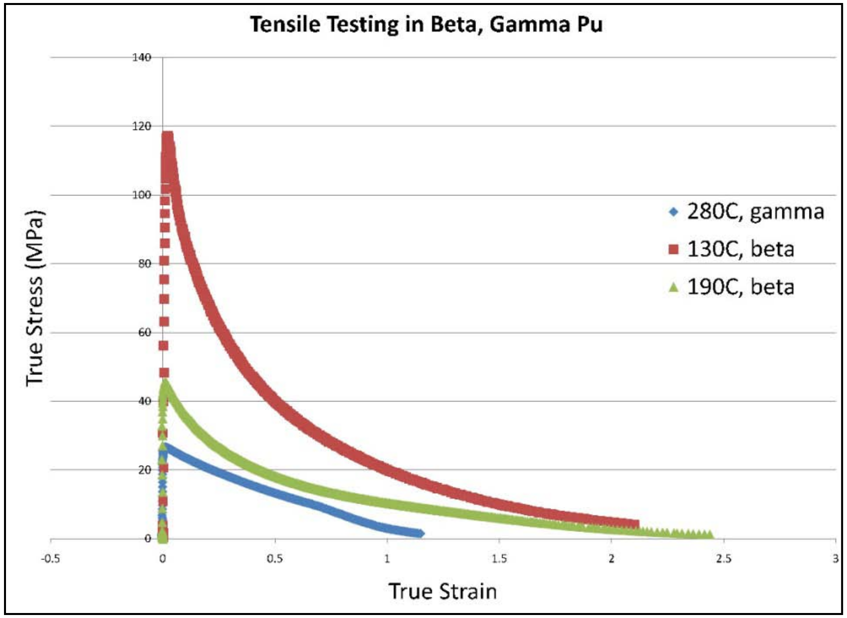
  δ´→ε transformation is ~35% more energetic than in pure α-Pu, and very sharply defined

300 ppm Ga alloyed Pu rod

Mechanical testing



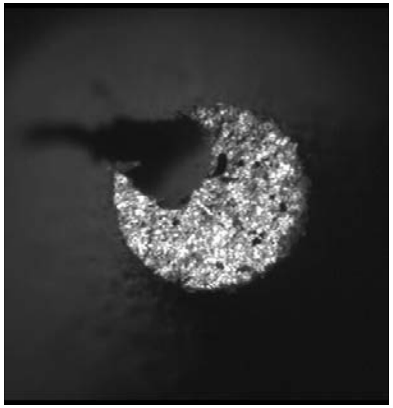
Smooth tensile bar tested in the high beta, 190 º C, 10^{-1}/min



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

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.



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

What went wrong?

Large undercooling and hysteresis observed

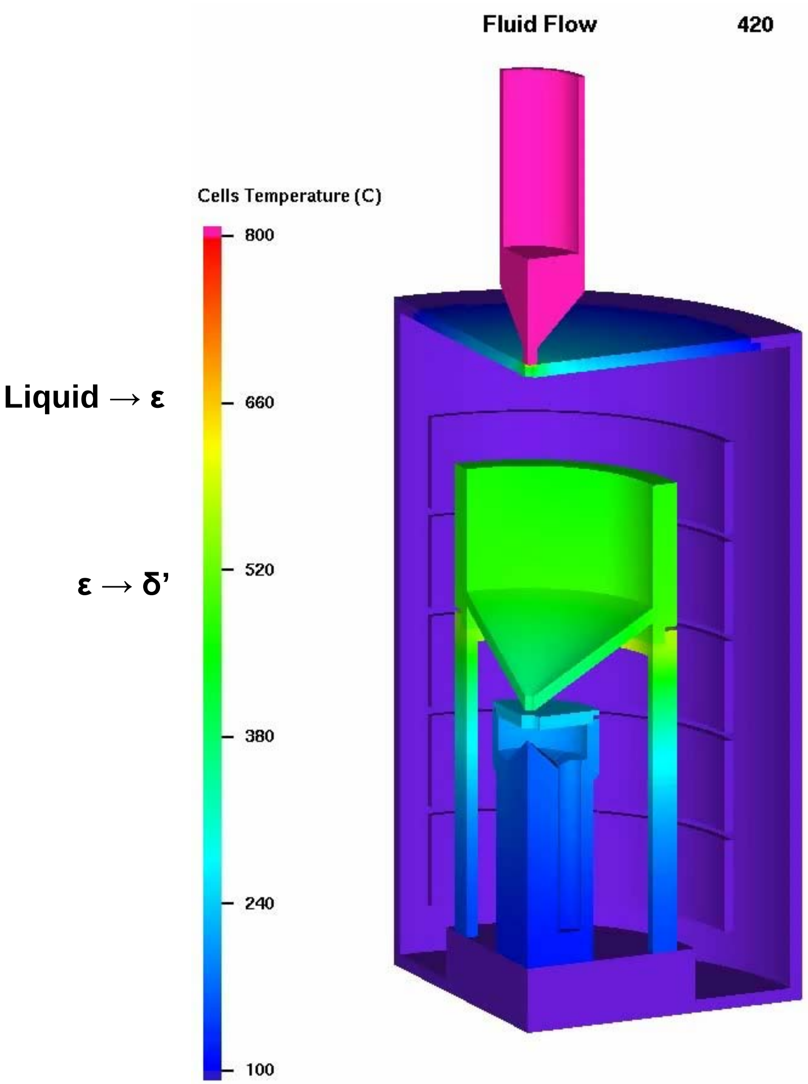
- Undercooling severely impacts phase transitions
- Phases transform from outside in
- Lack of thermal gradient results in lack of directionality for transformations
- Transformation is almost completely uniform

Simulations were performed using the new transformation temperatures and heats of transformation. The results indicate that the metal and mold became isothermal in the low gamma phase rather than the beta phase as planned.

What this could lead to?

- Severe centerline cracking
- Rod could pull itself apart
- High residual β retention

New mold design for Pu rods – will it work?



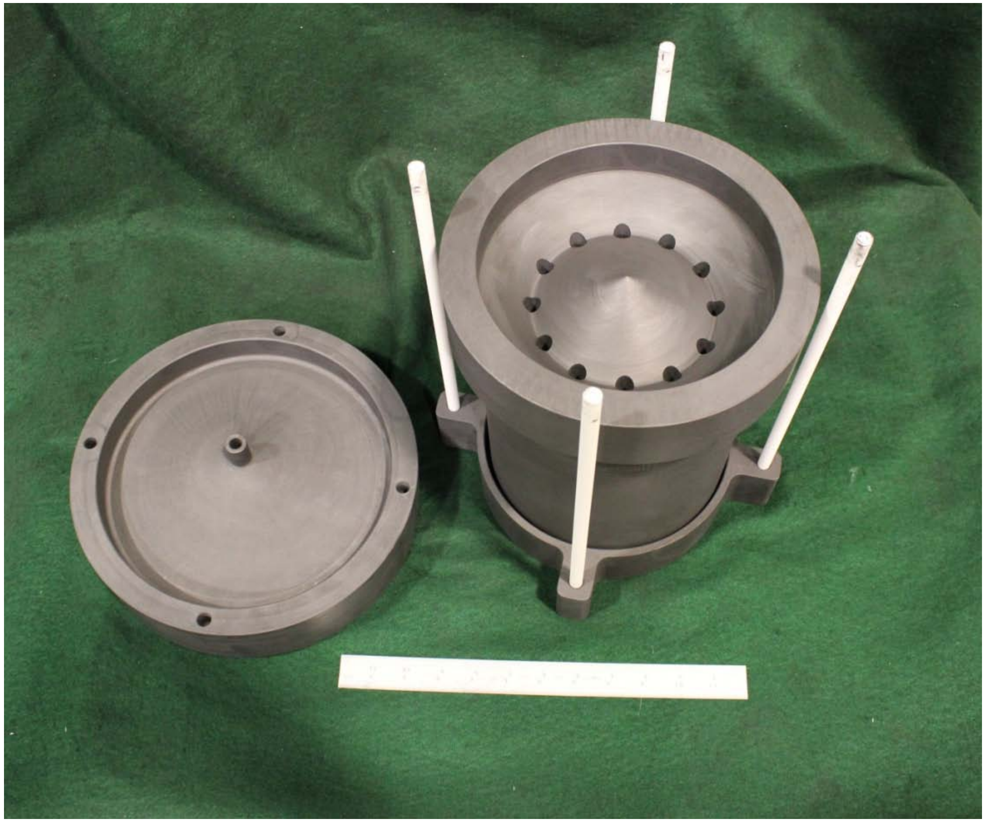
Simulations of new mold design with new material property data indicate that we will not be successful casting large diameter rods. We cannot get the isothermal temperature low enough without chill casting.

Smaller rods and therefore 3T samples are needed.

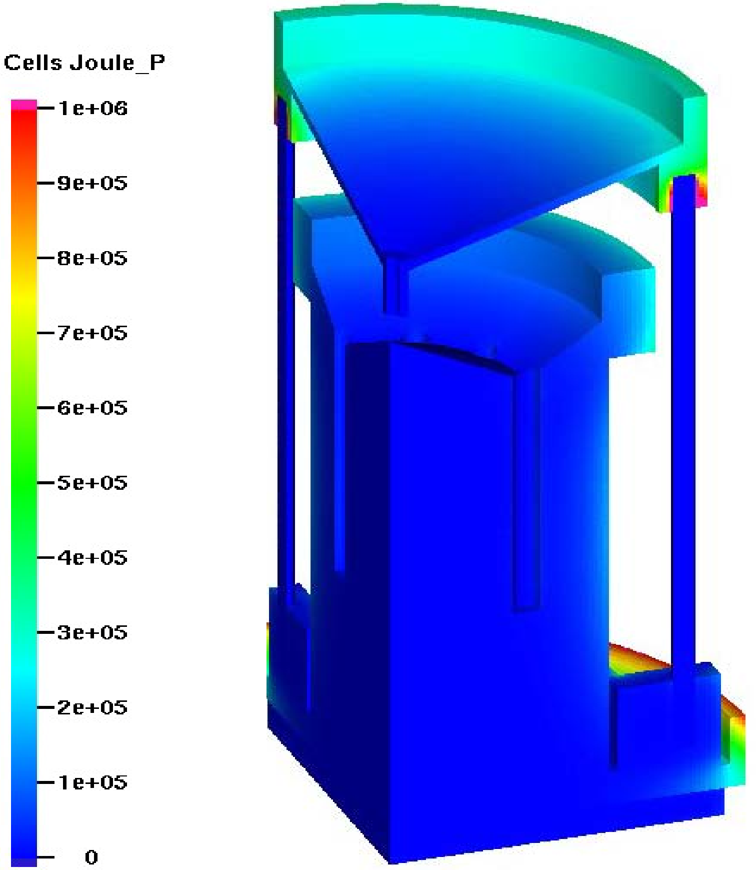
Smaller plutonium rod design

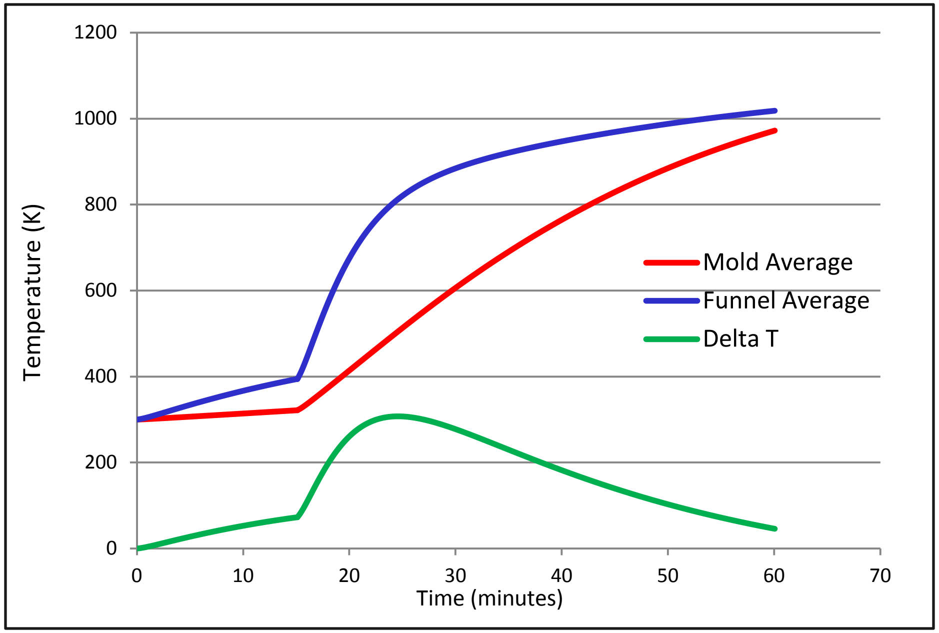
12 rod mold
10 mm diameter
127 mm long

Goal:
Cool from Liquid to β rapidly(<20 seconds)
Cool from β to α slowly from bottom to top (~5-10 minutes)
Keep β to α transition front flat and relatively narrow
This will avoid the β coring phenomenon



Smaller plutonium rod design





Where do we go from here?

Maintain modeling competency
- computers (secure and open)
- continual improvement to models and codes
- personnel to understand processes and codes
  - coupled experiment/manufacture and simulation

Manufacturing innovation

Collaboration with universities, community colleges, and industry
- talent pipeline
- next generation workforce

Generate and utilize high quality material property data
- MaRIE – 1st experiments
- molecular dynamics