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Development of a Gadolinium-Containing Alloy for Spent Nuclear Fuel Disposal in the Yucca Mountain Repository

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- Project is managed by Idaho National Laboratory
- Project has three co-principal contributors
  - Idaho National Laboratory
  - Sandia National Laboratories
  - Lehigh University
- There is a large number of people at all three sites who have contributed to this work

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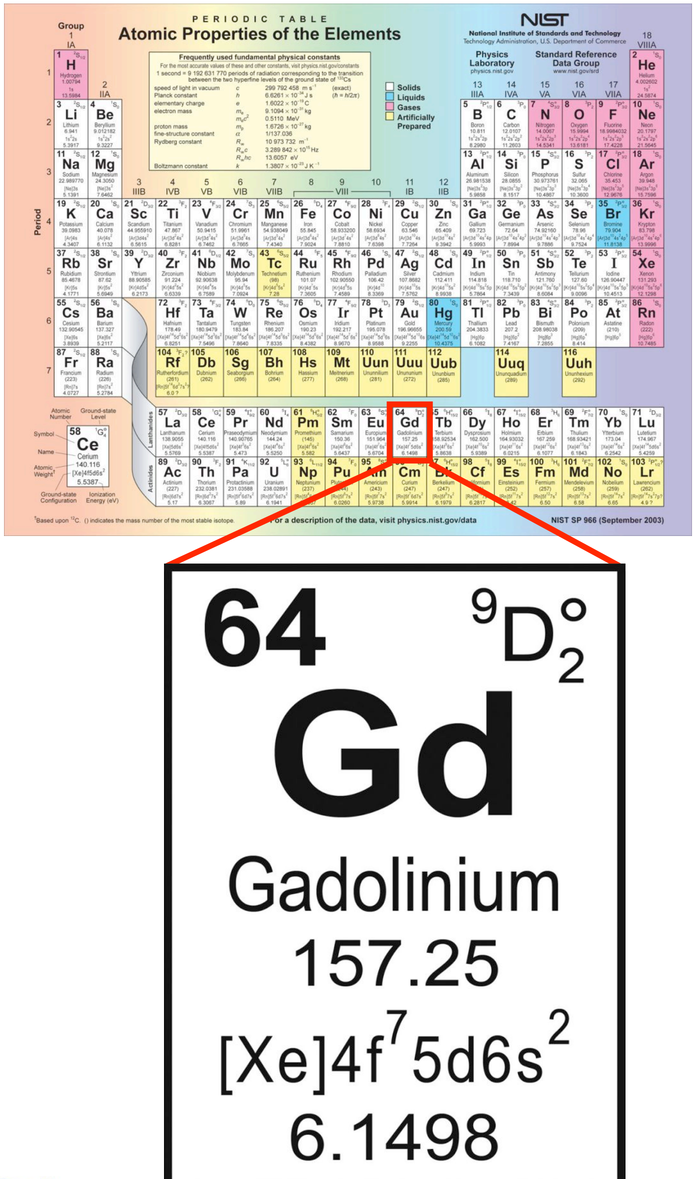
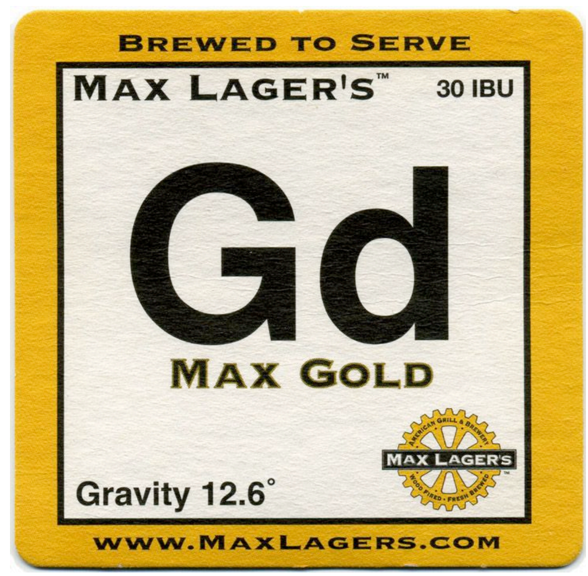
What is Gadolinium?



- Rare earth
- Atomic Number 64
- Lanthanide



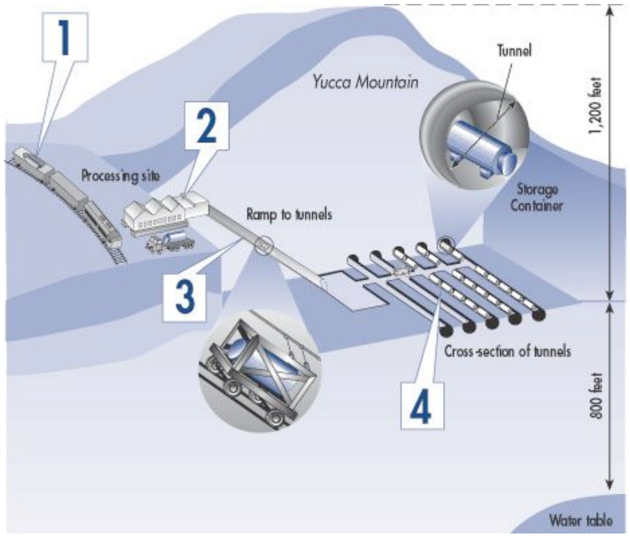
National Nuclear Security Administration

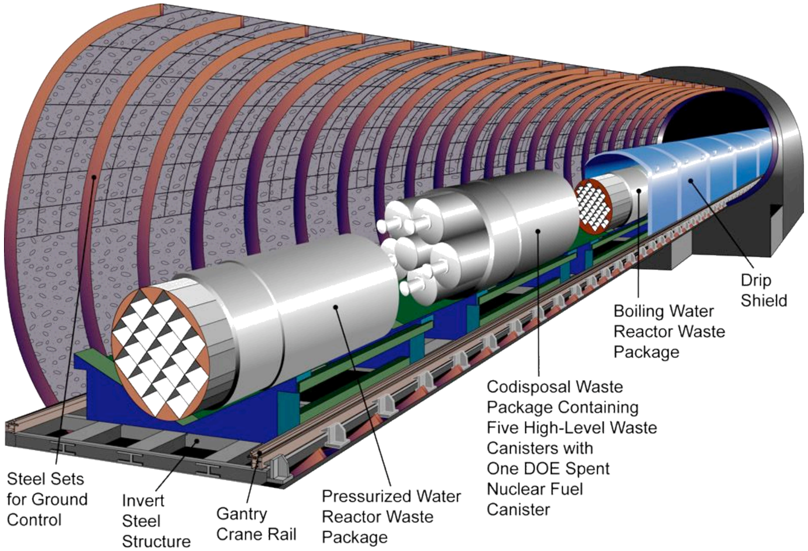
Sandia National Laboratories

What is Yucca Mountain?

- Yucca Mountain is the proposed repository for the nation's military and civilian high-level radioactive waste and spent nuclear fuel

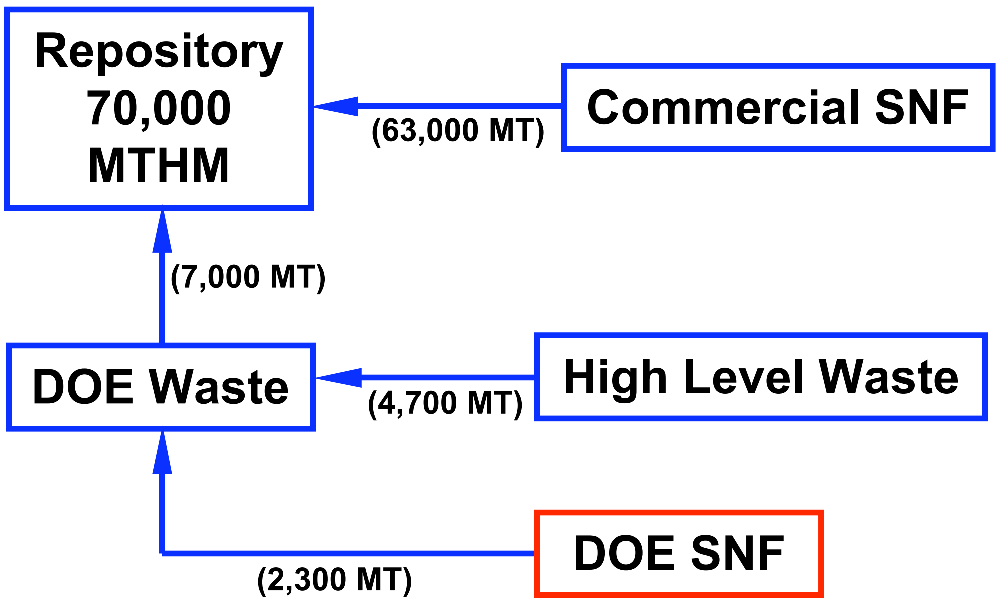
- Located within Nevada Test Site
- Concept relies on engineered barriers rather than geologic containment
- DOE is currently preparing NRC license application to proceed with construction

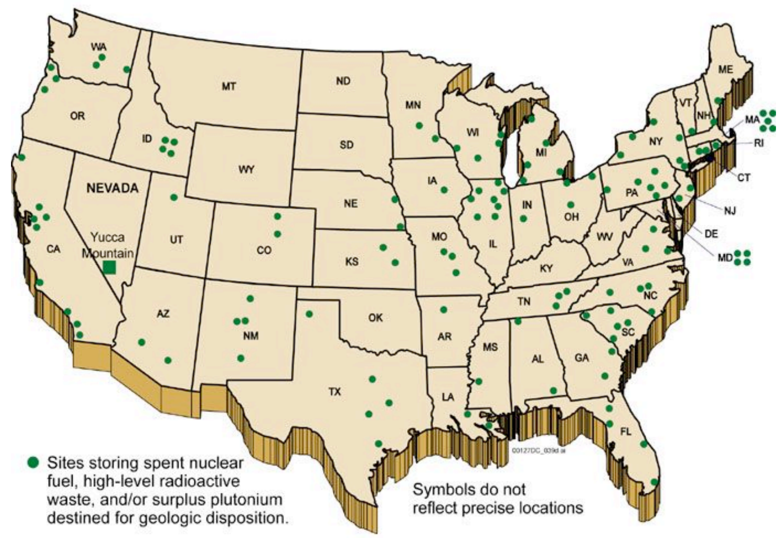




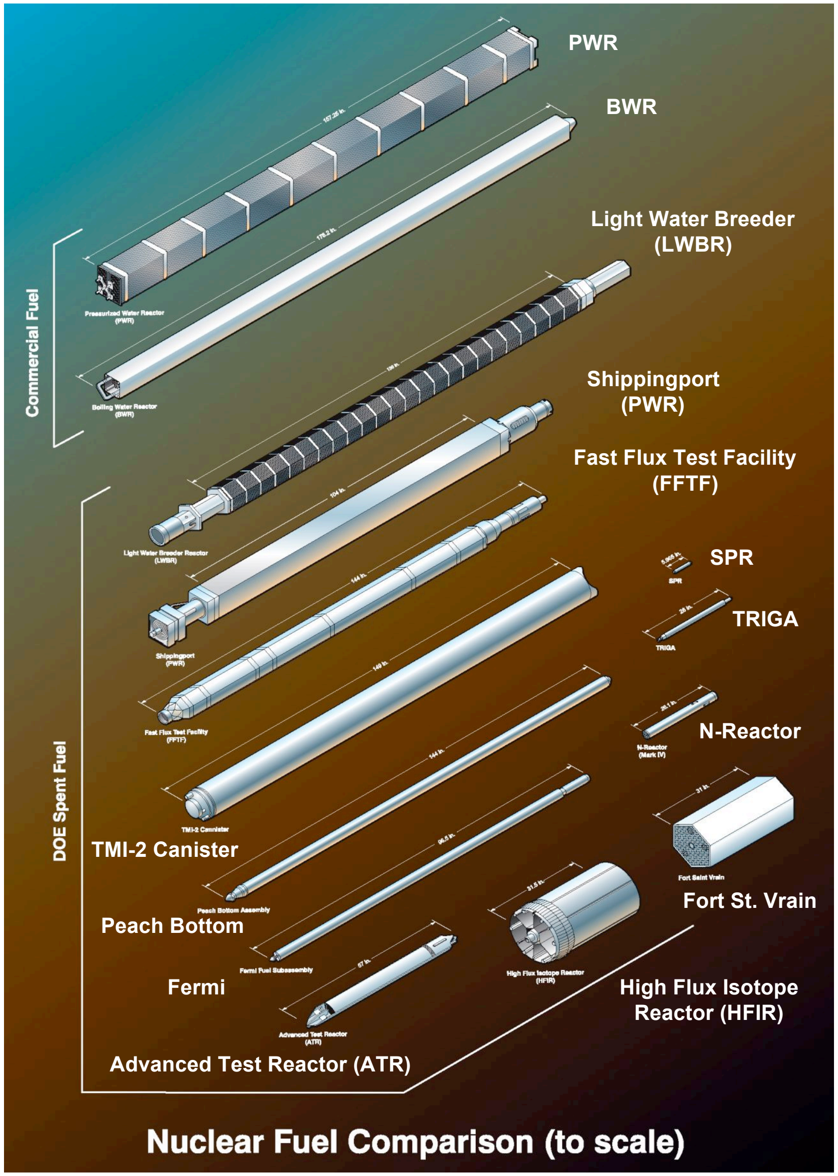
Contents of the Repository

- The Nuclear Waste Policy Act authorized evaluation of a repository to contain 70,000 MTHM





Typical Fuel Assemblies



- DOE Spent Fuel includes a wide variety of fuel types, materials, sizes, and configurations
- Any of these reactors would be an interesting talk in itself

DOE SNF Disposal

Problem

- Some types of US DOE spent nuclear fuel (SNF) contain highly enriched uranium
- Final disposition of this SNF in the repository may require criticality control during the regulatory period

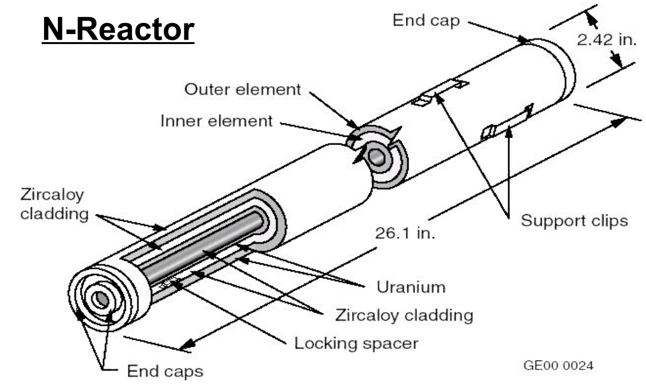
Approach

- SNF will be packaged in standardized canister with baskets fabricated from corrosion resistant neutron absorbing materials

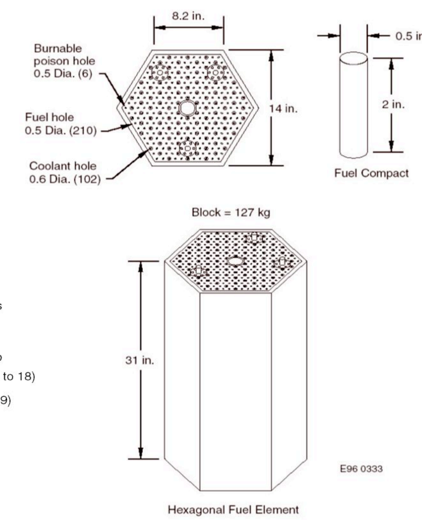
Benefits

- Decreased number of DOE SNF packages going to repository with reduced handling and materials costs.
- Ni-Gd is available for credit in Part 71/72 license if needed

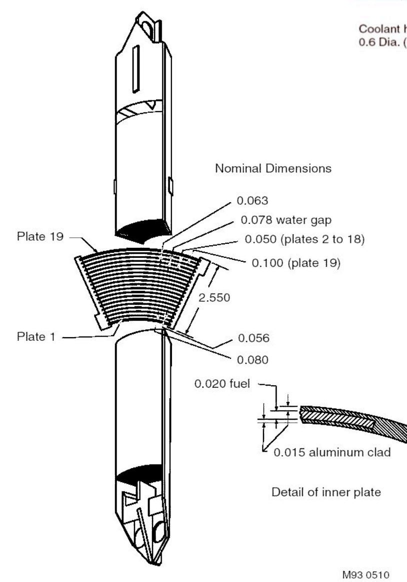
N-Reactor



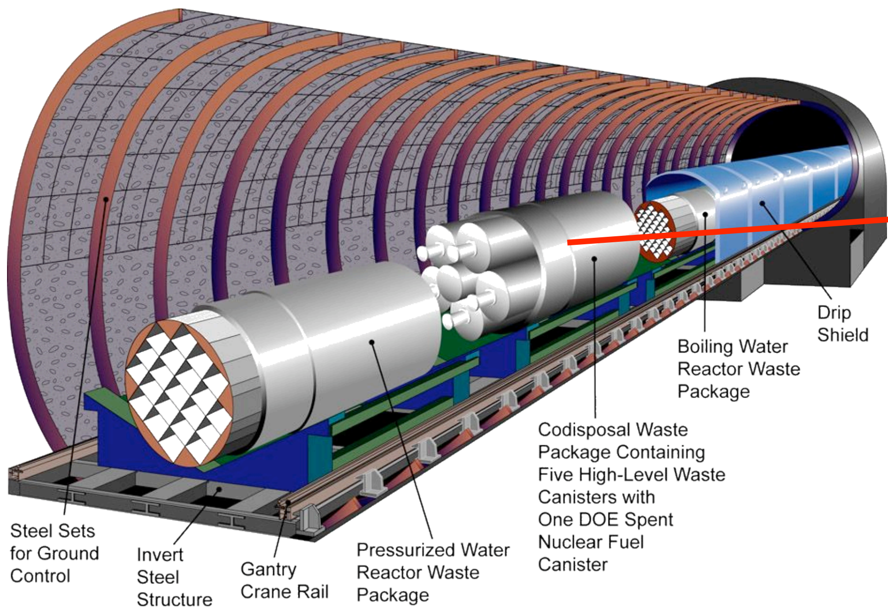
Fort St. Vrain

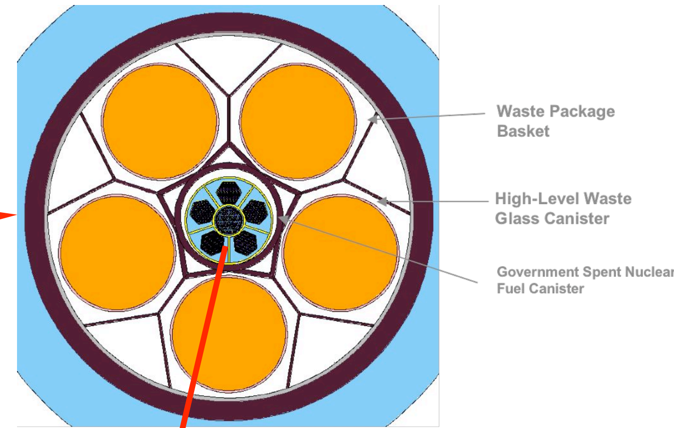


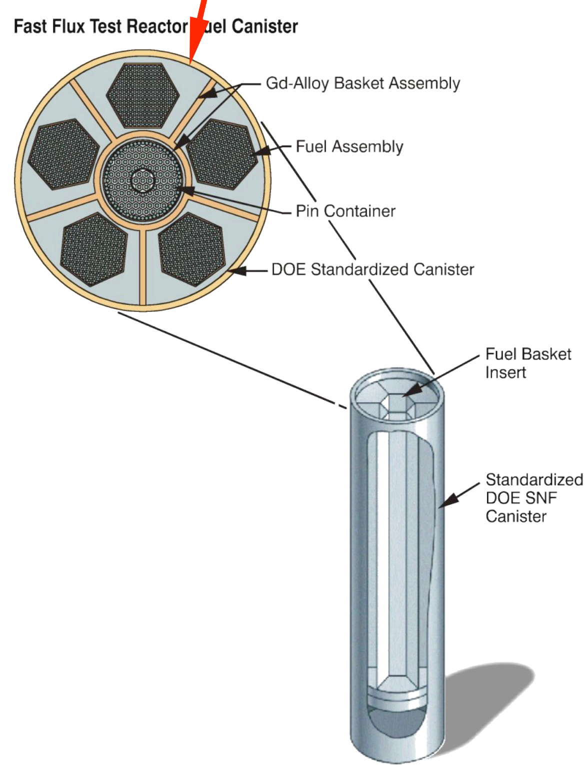
ATR



DOE Standardized Canister







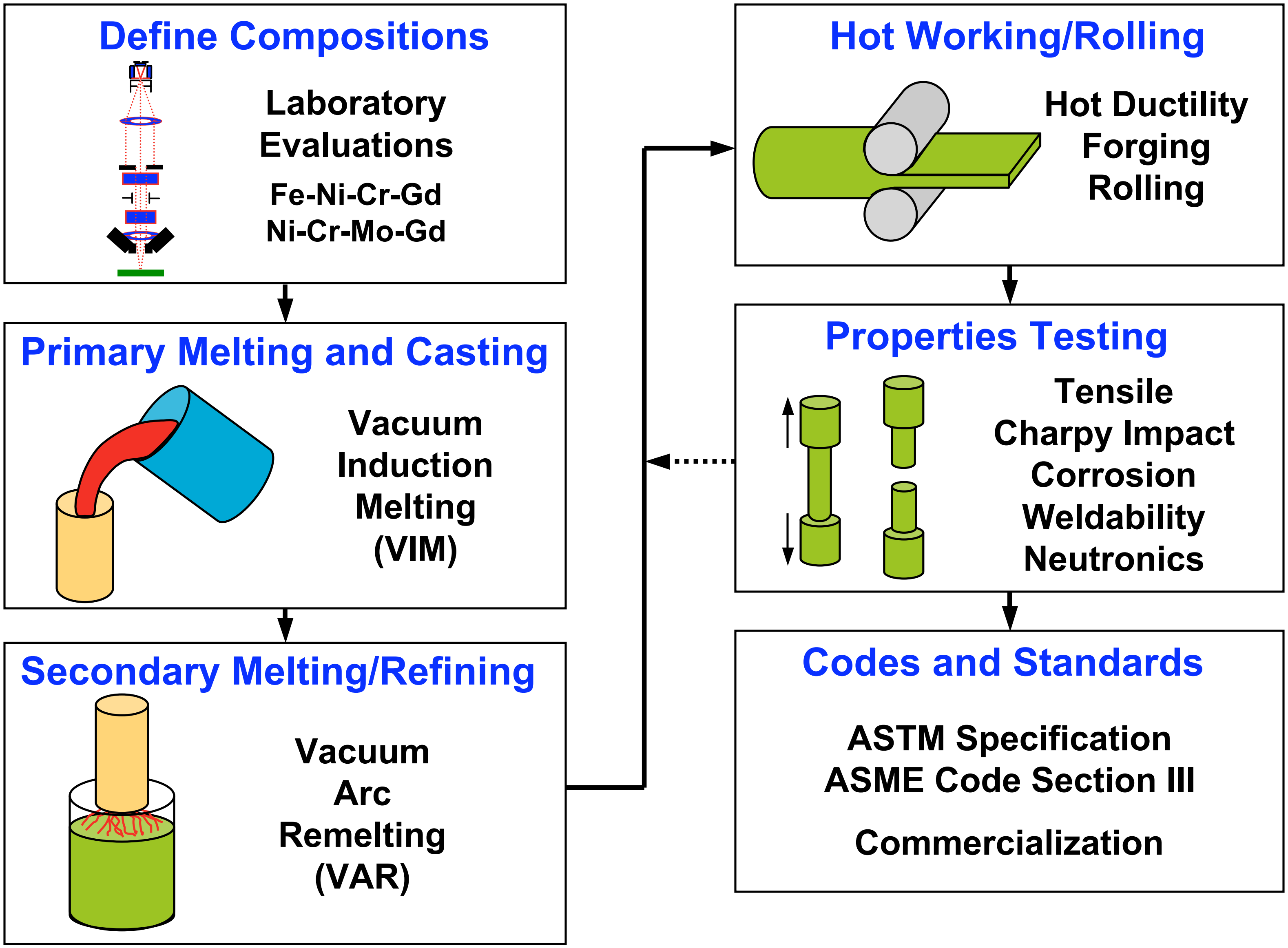
- **Codisposal waste package contains high level waste and SNF (in DOE Standardized Canister)**
- **Preliminary studies indicated that borated stainless steel is not suitable (criticality is concern after degradation)**
- **Gd is a more suitable absorber under these conditions**

Major Alloy Design Constraints

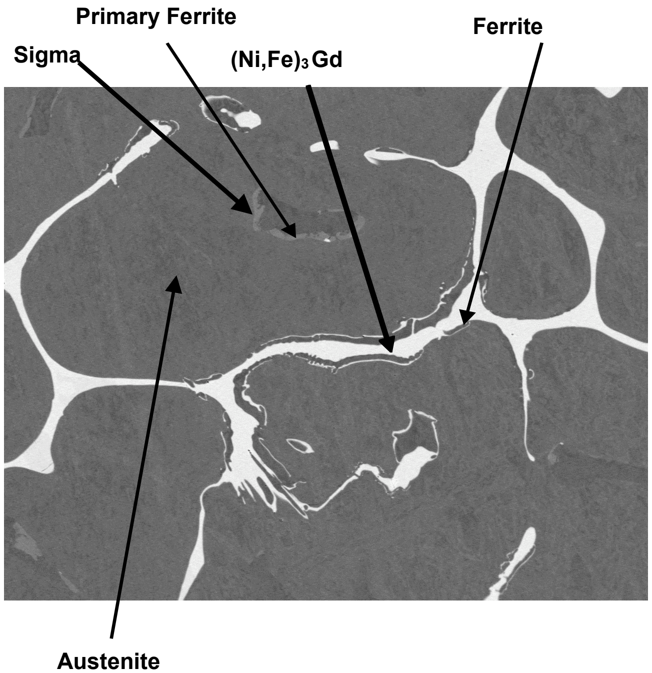
- Gd concentration is essentially fixed by highest enrichment fuel and geometric constraints (and the need to minimize administrative controls)
- Original requirement was 1.5 wt% Gd, current is 1.5 wt% Gd min
- Corrosion requirements
- Produce by conventional routes (e.g. ingot metallurgy, hot rolling, tube forming, etc), approximately 1100 MT needed
- Welding requirements
- Must be acceptable by ASME Boiler and Pressure Vessel Code, Section III, Division 3.

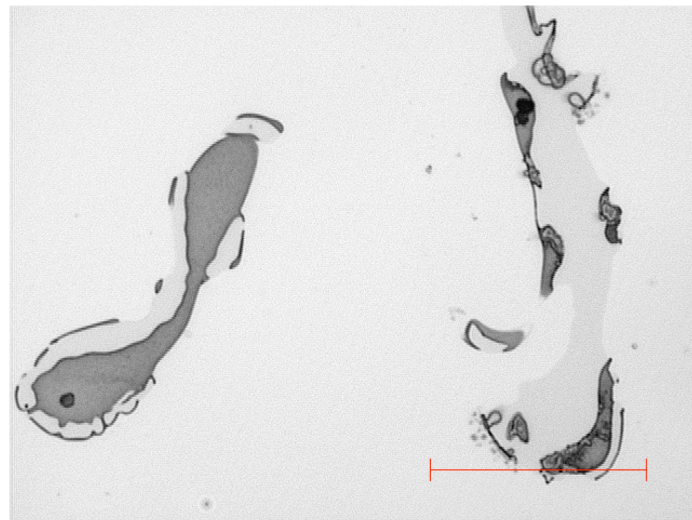
- Original INL concept was Gd-containing austenitic stainless steel (316L)
- Such an alloy was patented by Carpenter in mid-60's

Project Flow

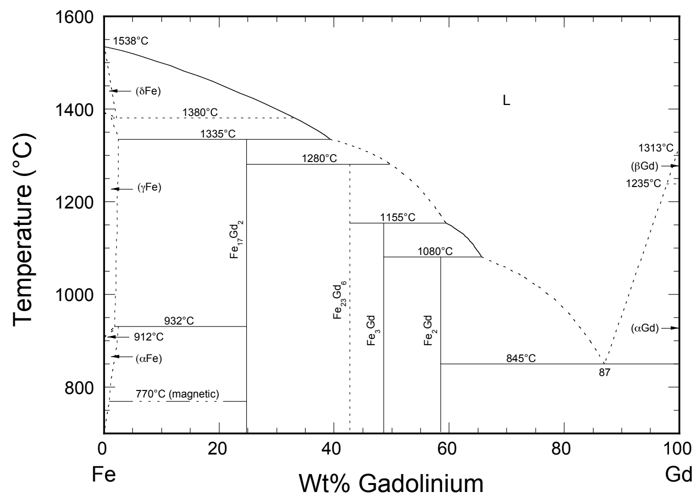


Gd alloyed Stainless Steel



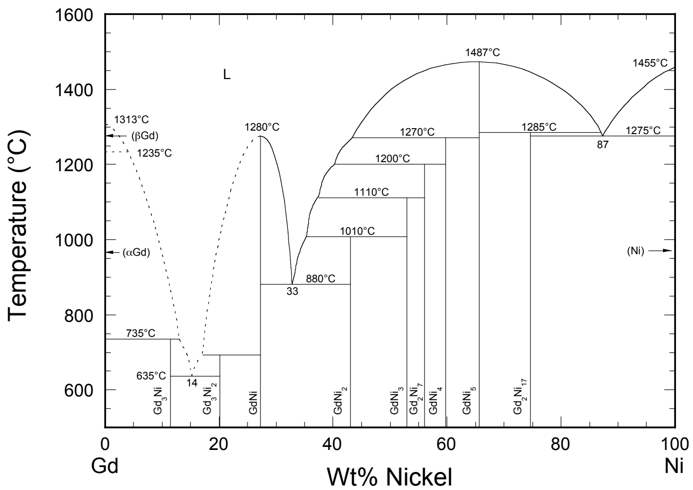


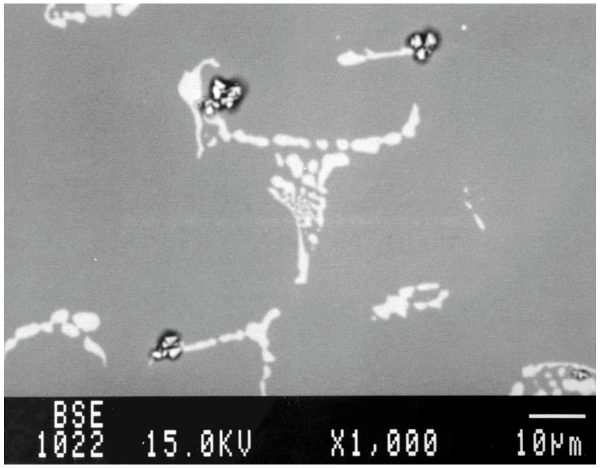
- Complex peritectic solidification for range of Cr_{eq}/Ni_{eq}
- $Gd(Fe,Ni)_3$ terminal constituent



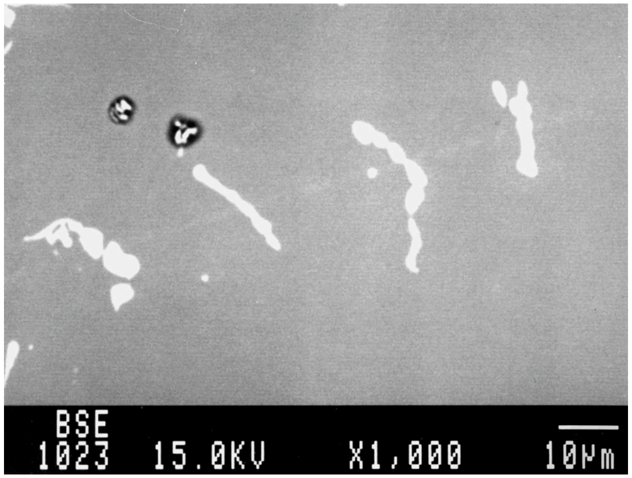
- 316L alloyed with Gd (0 to 6 wt.% nominal) tested
- Large solidification temperature range and complex microstructure caused severe hot workability problems
- Corrosion test data was not encouraging

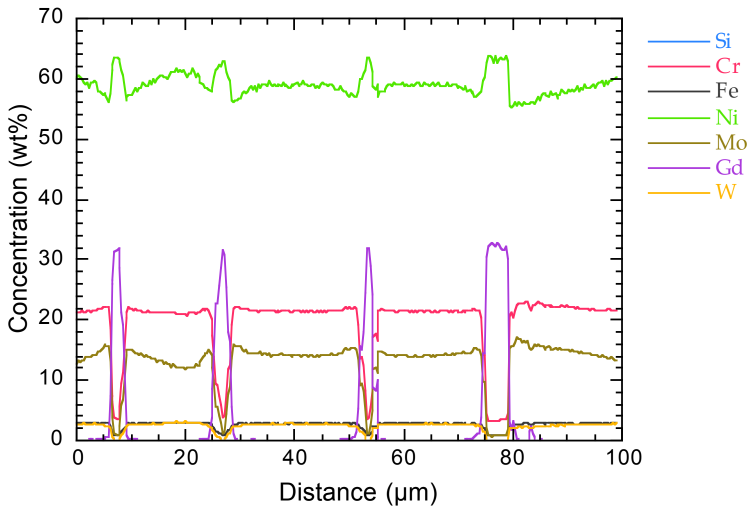
Preliminary Ni-Based Alloy





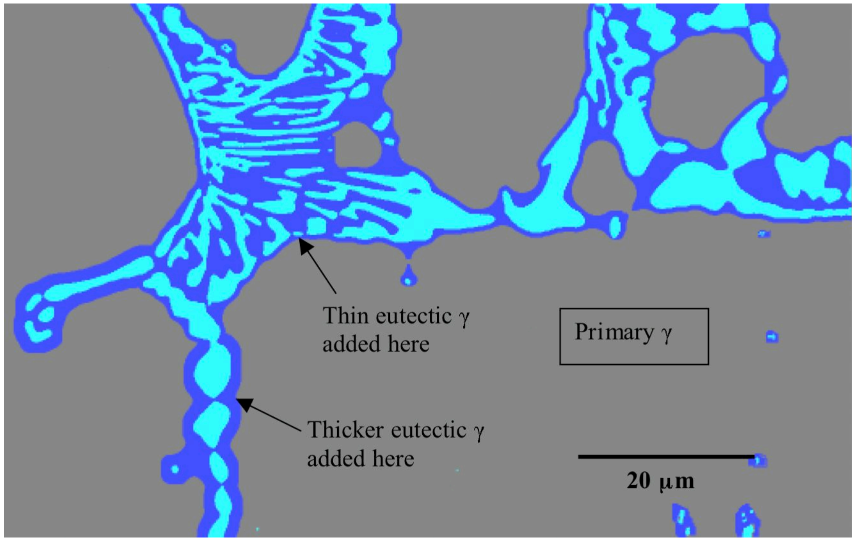
- Eutectic solidification
- $Gd(Ni,Cr,Mo)_5$ terminal constituent

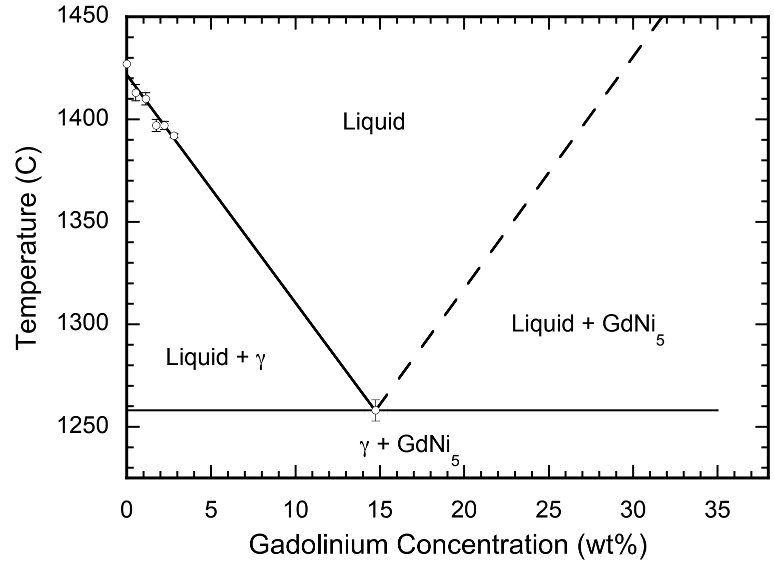


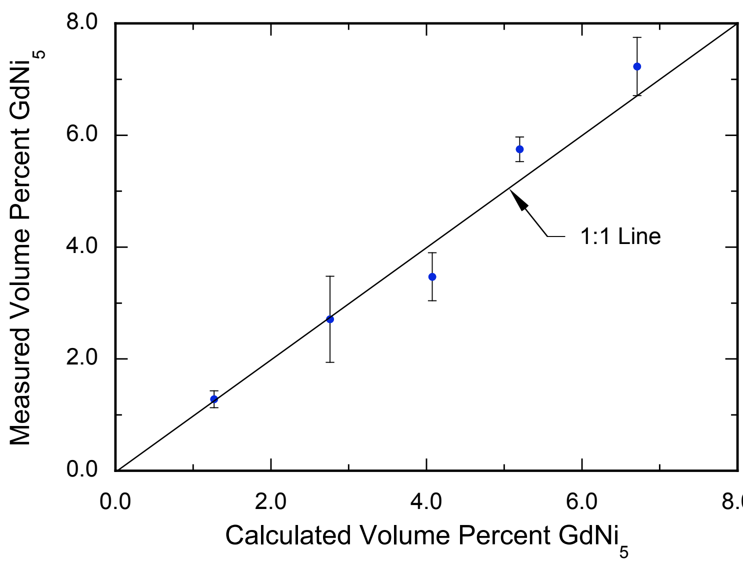


- Elemental partitioning typical of Ni-Cr-Mo alloy solidification
- Essentially no Gd in matrix

Solidification Diagram



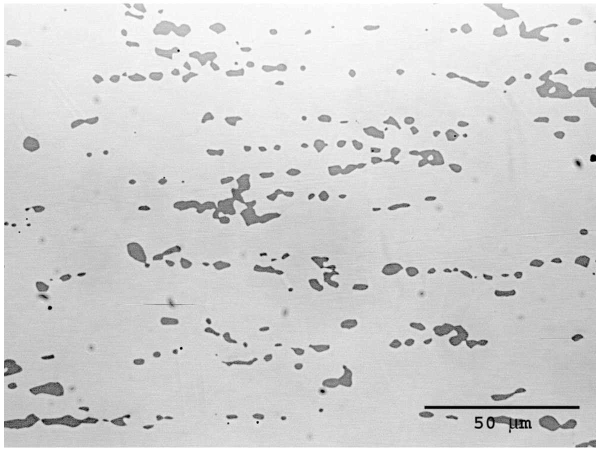




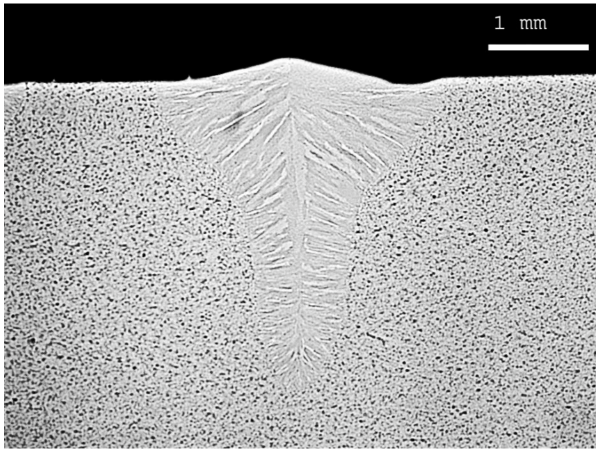
- NiCrMoGd system is complex, but behaves much like a binary eutectic alloy

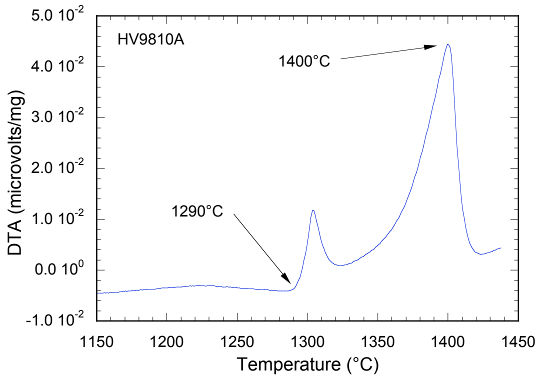
Preliminary Processing Assessments

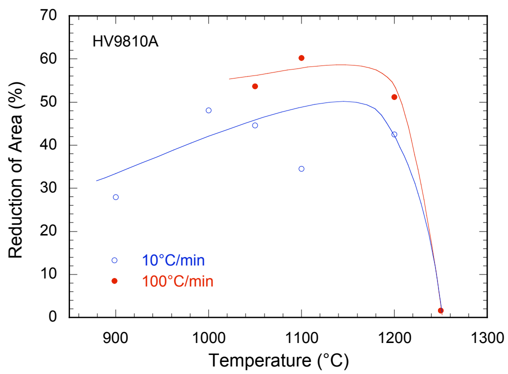
Hot Rolled Plate



Electron Beam Weld



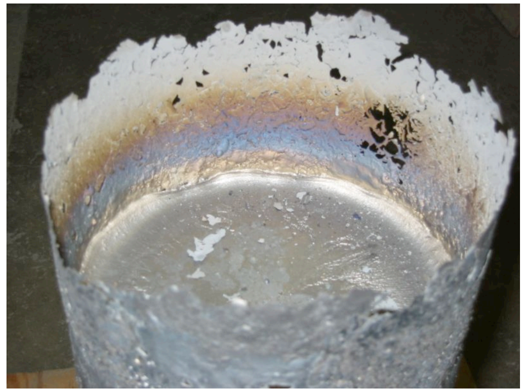


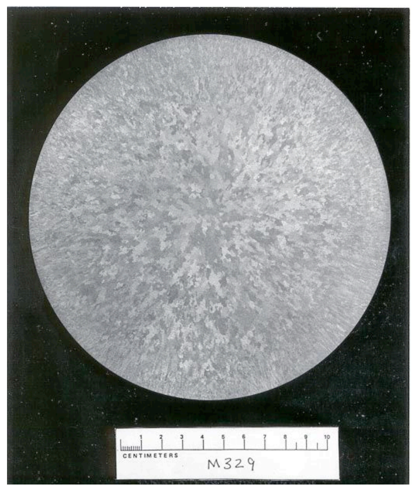


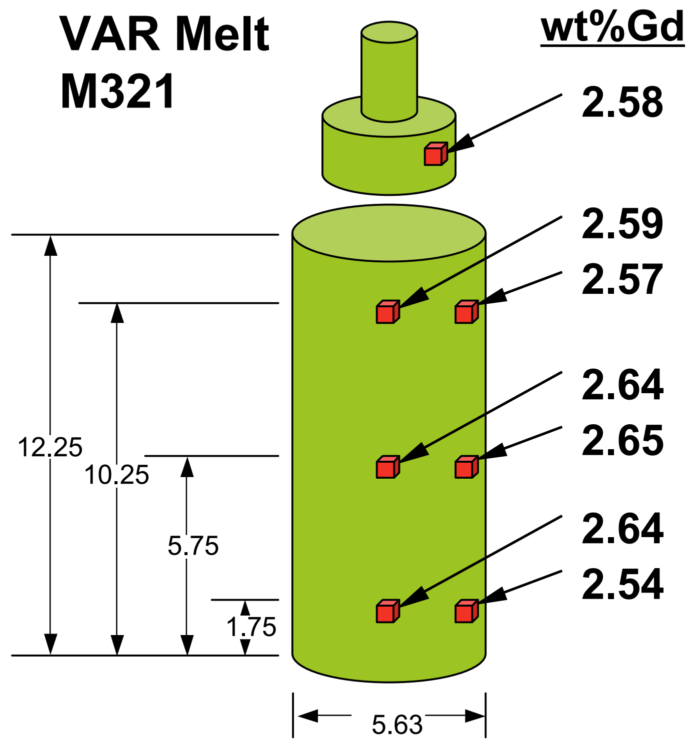
1.6 wt% Gd alloy exhibited promising:

- **Microstructure**
- **Solidification temperature range**
- **Hot ductility**
- **Welding behavior**
- **Mechanical properties (though just acceptable)**

Vacuum Arc Remelting

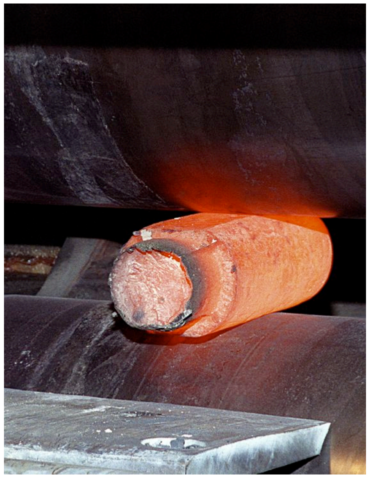






- Gd evaporation and loss effectively controlled
- Gd uniformly distributed throughout ingot

Hot Working



VAR Ingot-as cast

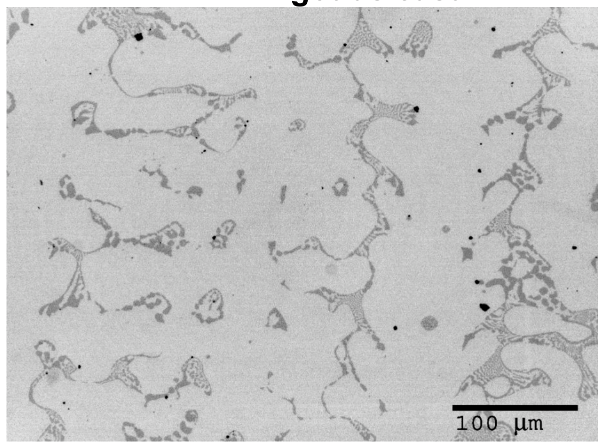
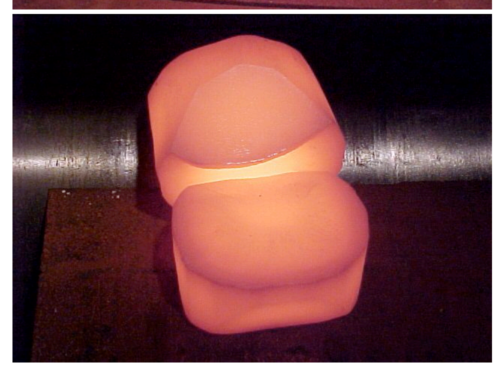


Plate (L-S orientation)

- Alloys up to 2.5 wt% Gd have been successfully hot worked
- Hot working based initially on typical procedures for Ni-Cr-Mo alloys
- Cracking during rolling was sometimes observed
- Transverse lateral expansion was marginal

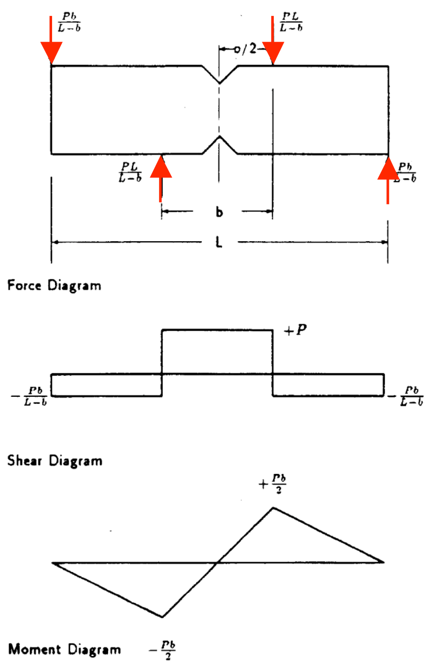
- **Improved understanding and control of hot deformation and resultant properties needed**

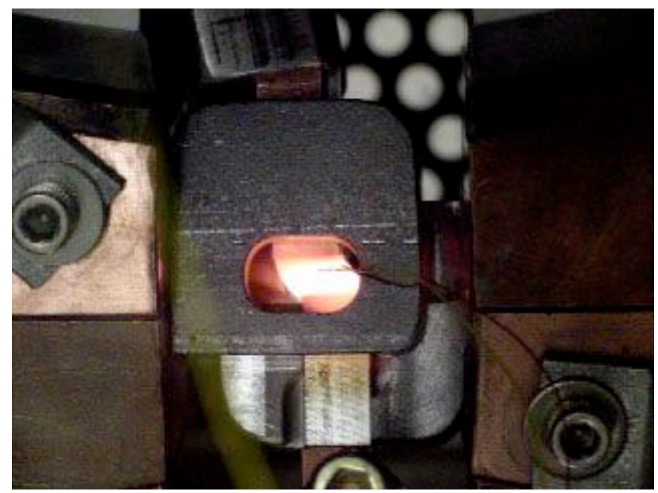


Hot Deformation Analysis

- Understanding microstructural evolution during hot working is crucial to developing appropriate procedures and properties
- This is most easily accomplished for simple deformation conditions

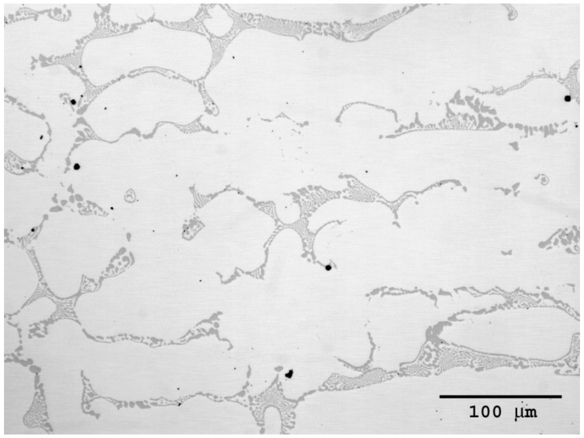
Iosipescu asymmetric flexure concept was adapted to thermo-mechanical test equipment



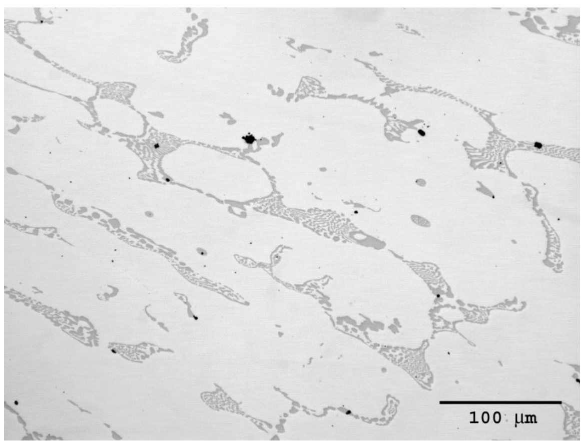


Hot Deformation Analysis

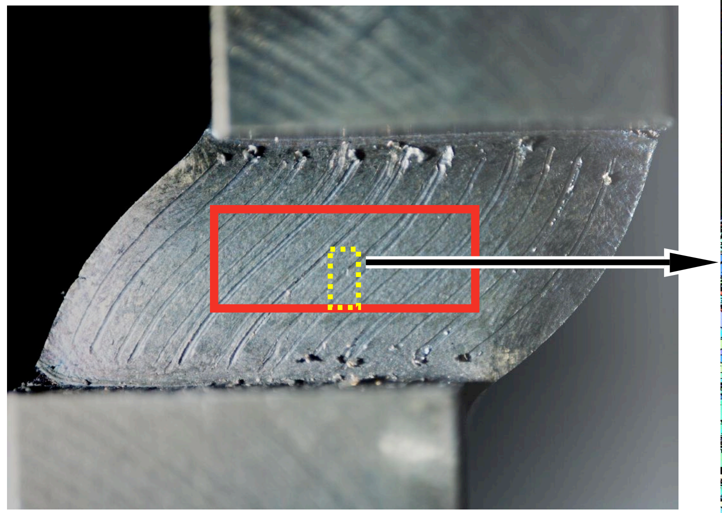
VAR Ingot, As-Cast



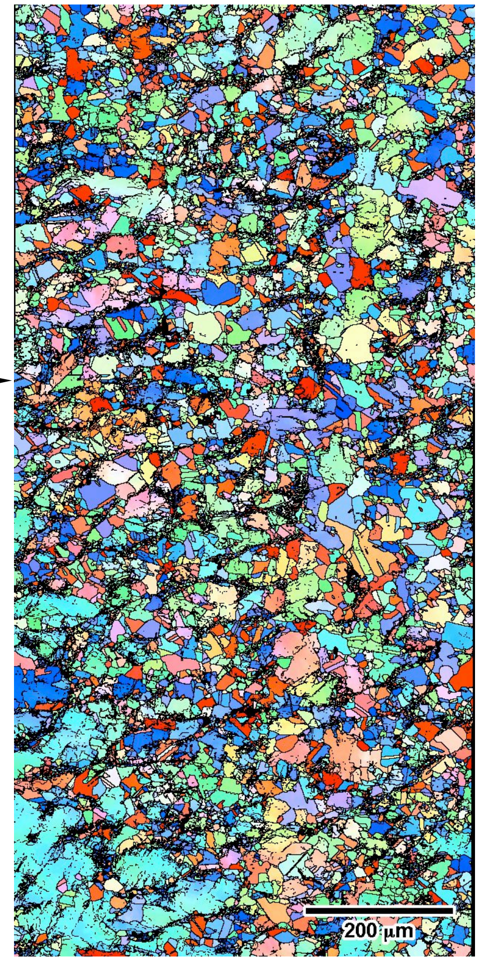
1050°C, τ = 0.97, $\dot{\tau}$ = 0.1/sec



Temperature, deformation, and rate can be controlled in a useful volume of material

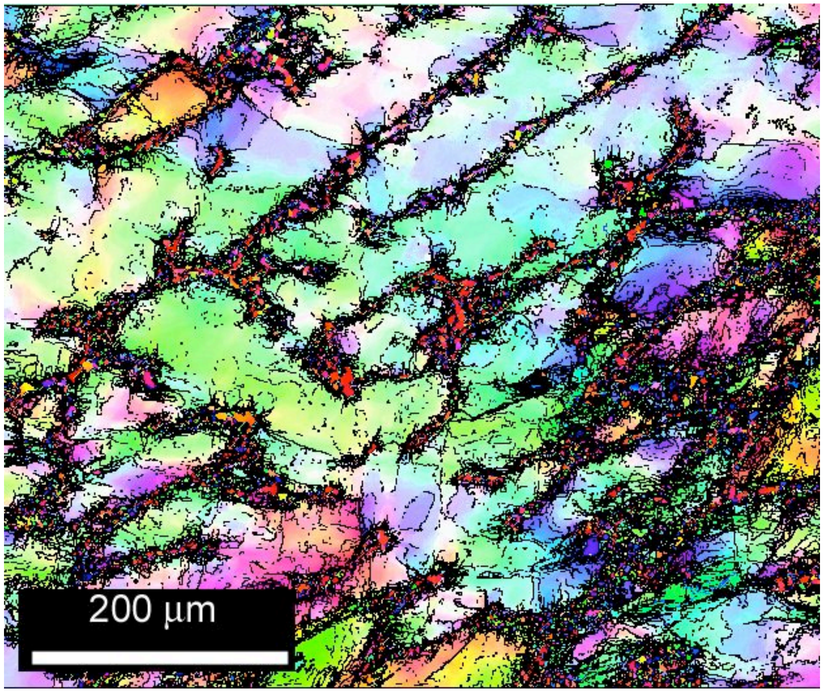


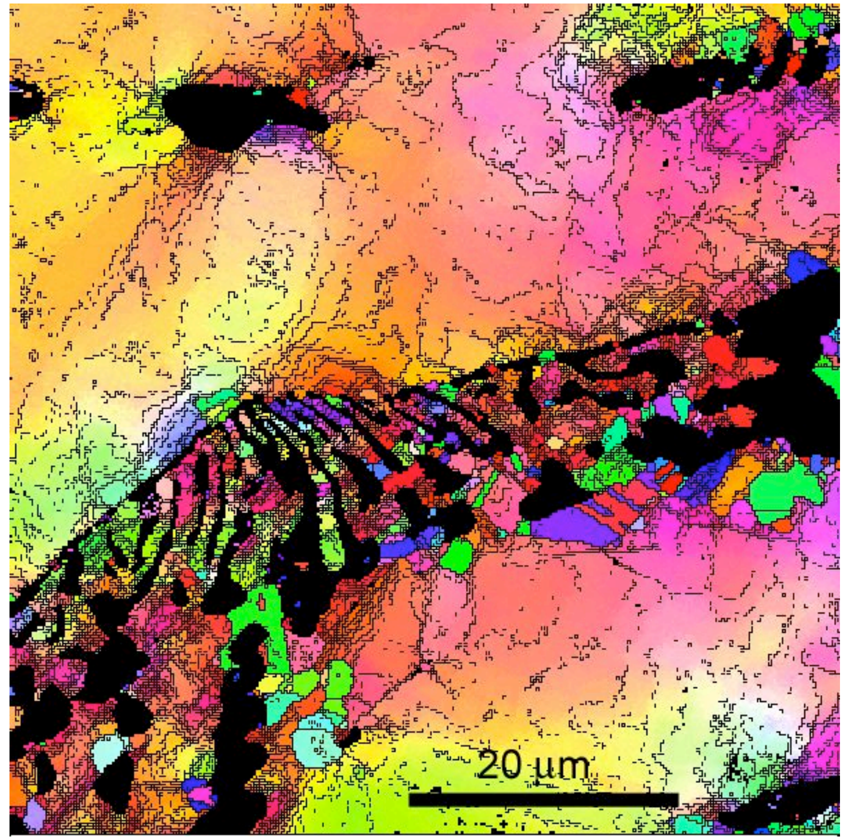
- **Little information can be obtained from optical metallography alone**
- **EBSD can be used to assess deformation localization and recrystallization**



Hot deformation is highly localized

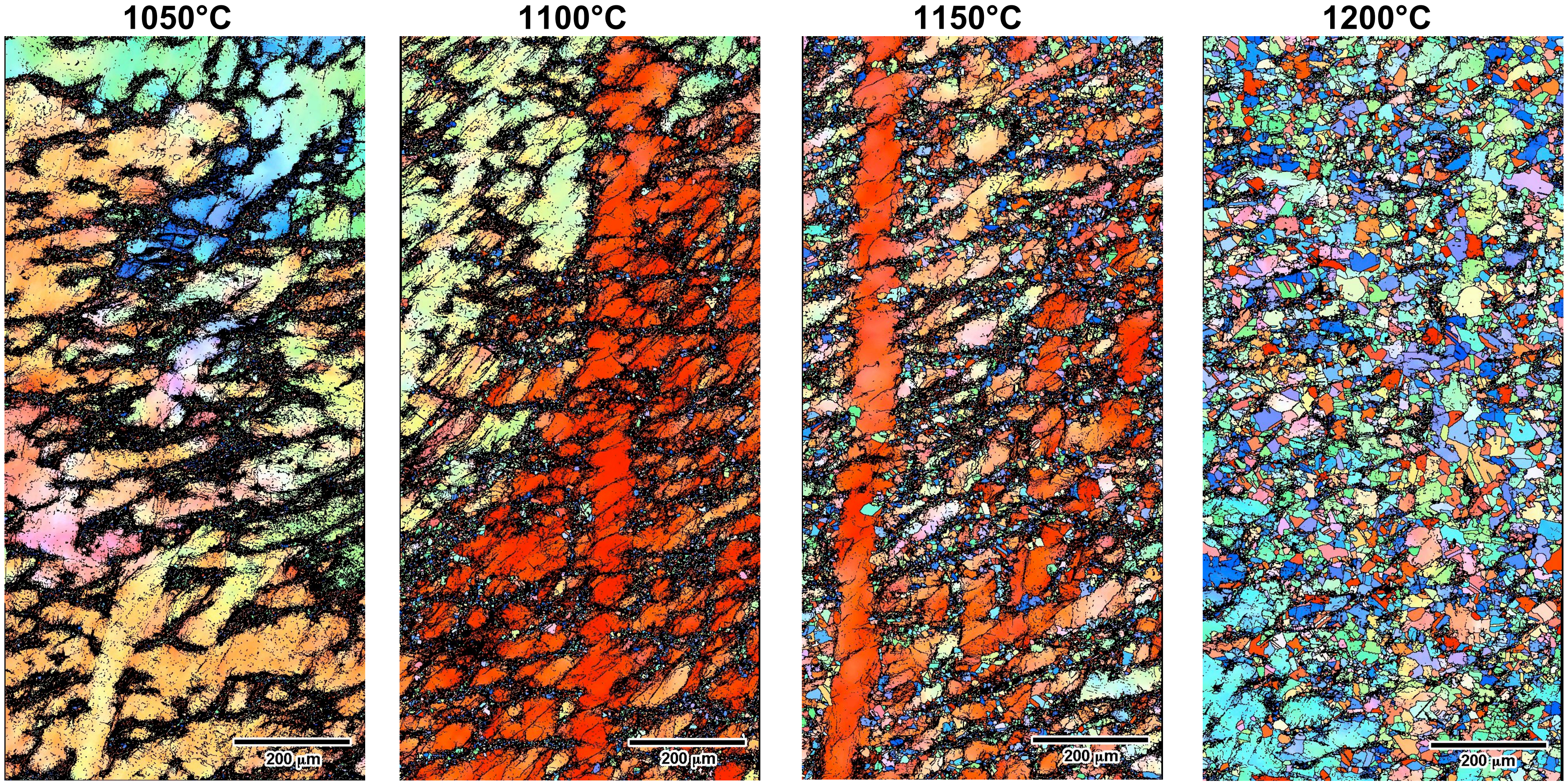
1050°C, τ = 1.0, $\dot{\tau}$ = 0.1/sec





- Deformation is localized near Ni_5Gd particles, some recrystallization within eutectic region
- Gadolinides do not appreciably deform even near solidus temperature

Rolling Temperature



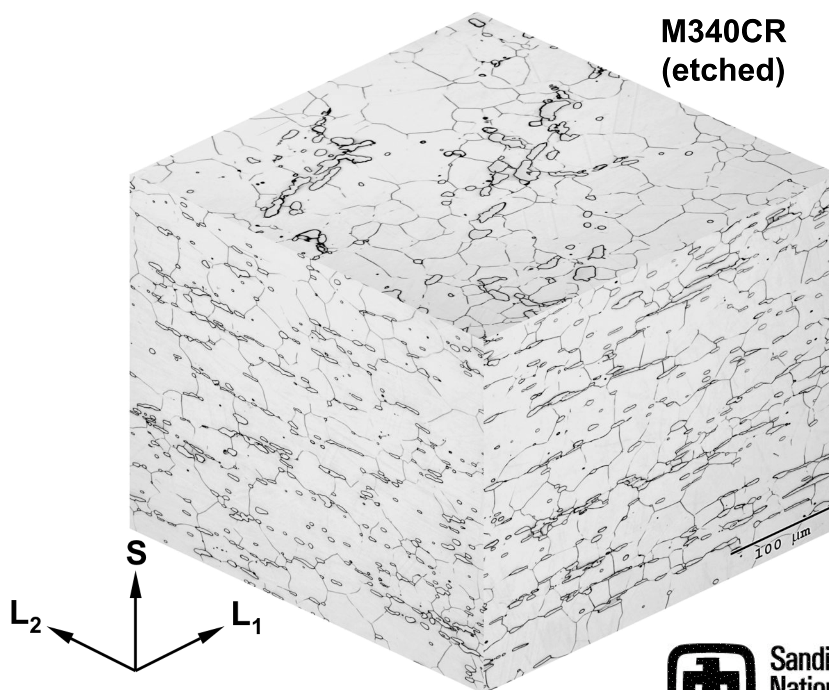
- Minimal recrystallization observed below 1200°C
- Implies that hot working temperatures need to be much higher than conventional Ni-Cr-Mo alloys
- Implementation of higher working temperatures eliminated cracking
- Implementation of cross rolling procedures improved transverse properties

ASME Code Case

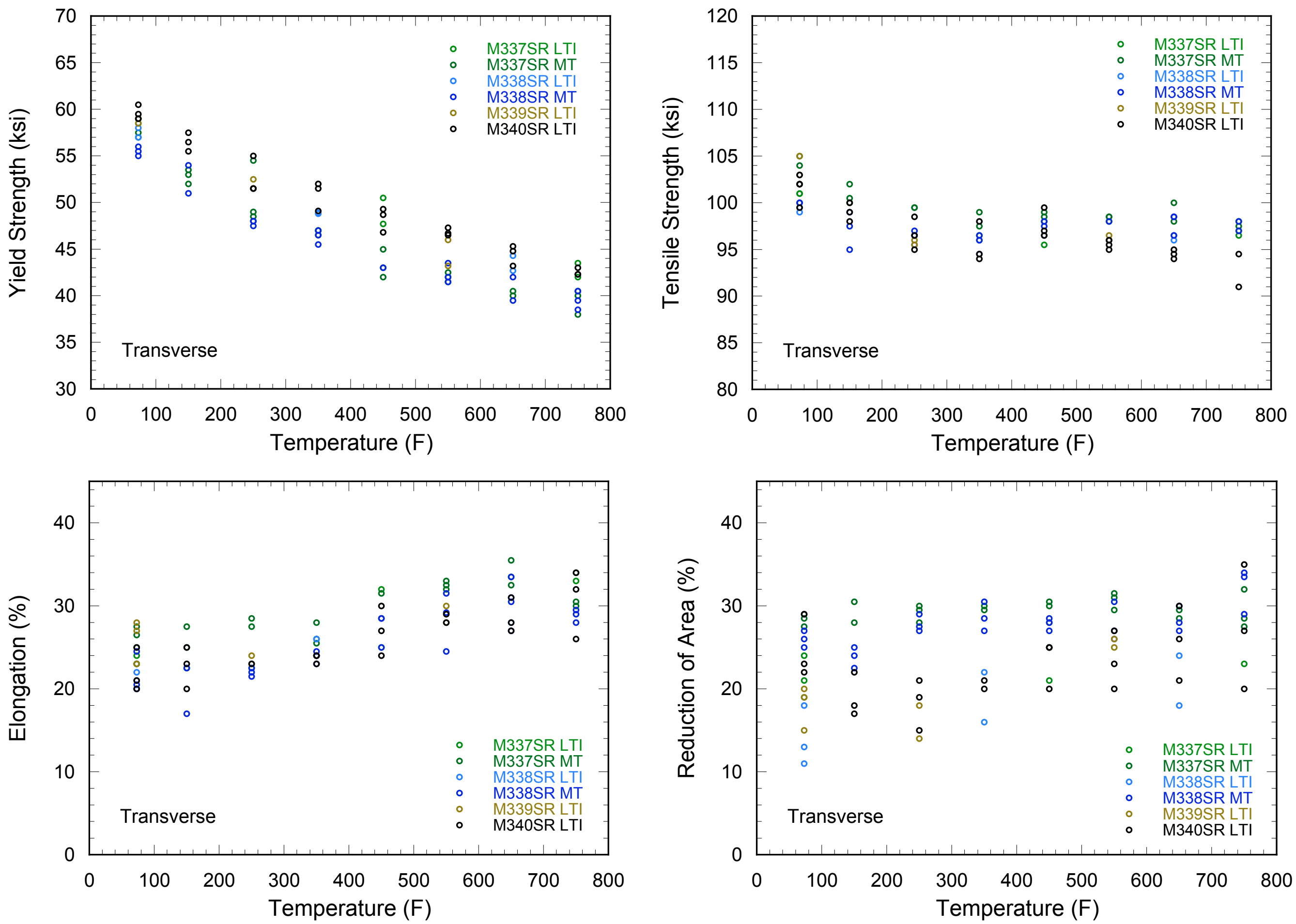
- **ASTM coverage applied for, defended, and received (ASTM B 932-04)**

ASTM B932-04 Composition Limits (%)	
Element	Alloy N06464
Molybdenum	13.1 to 16.0
Chromium	14.5 to 17.1
Iron	1.0 max
Cobalt, max	2.0
Carbon, max	0.010
Silicon, max	0.08
Manganese, max	0.5
Phosphorous, max	0.005
Sulfur, max	0.005
Nickel	Remainder
Oxygen	0.005
Nitrogen, max	0.010
Gadolinium	1.9 to 2.1

- **2 wt% Gd**
- **Heats produced by VIM/VAR and hot rolling to 1/2" thick plate**
- **Straight and cross rolled versions produced**

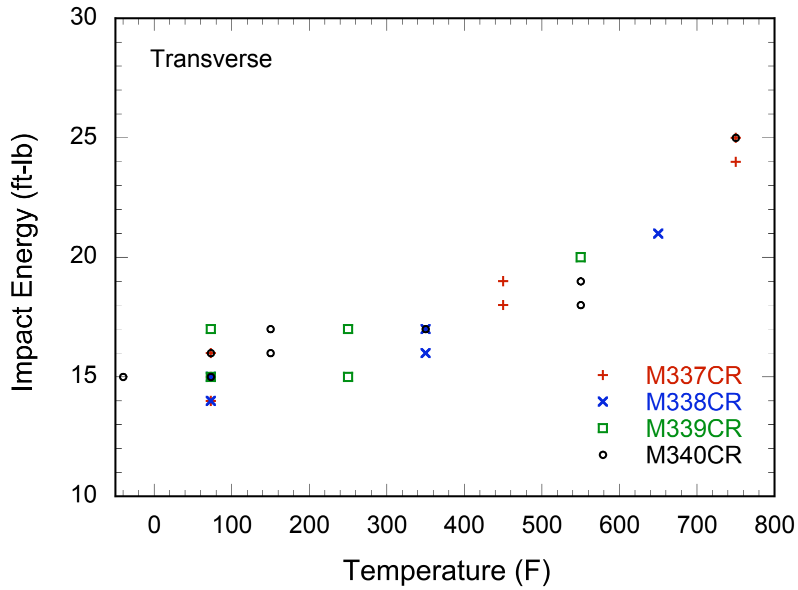


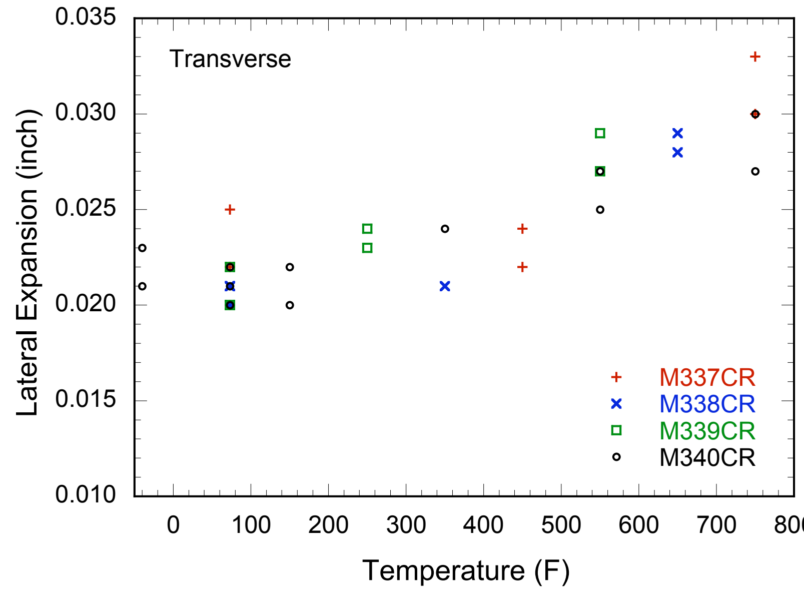
Tensile Properties



- Tensile properties are used by Code Committees to set allowable stress limits
- Strength and trend curves are similar to UNS N06455 (Hastelloy C-4)
- Gadolinium reduces ductility
- Thermophysical properties also measured

Impact and Fracture Toughness

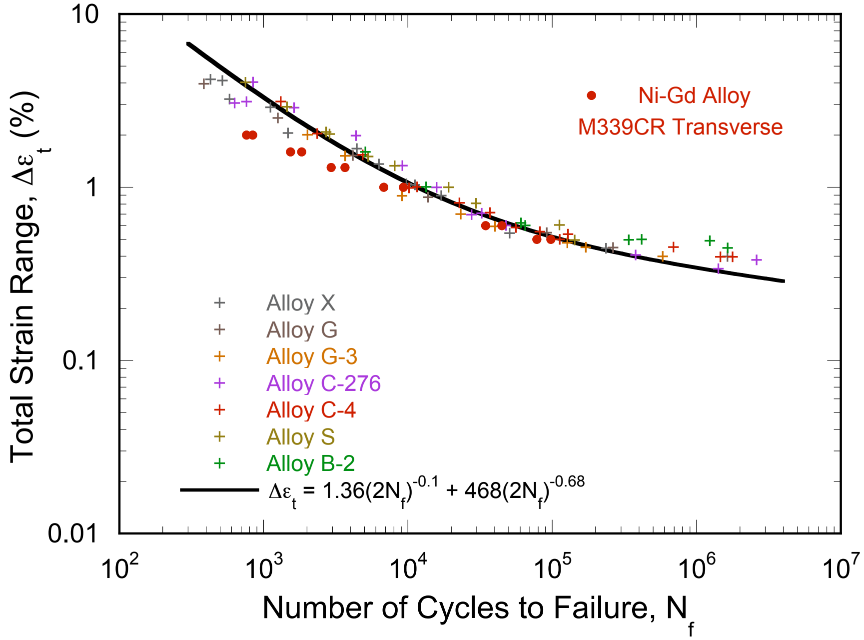




Alloy	J_{IC} (in-lb/in2)	K_{JIC} (ksi-in1/2)
M339CR	209.9*	84.2
M339CR	221.0*	86.4
M339SR	172.7*	76.4
M339SR	193.3	80.8

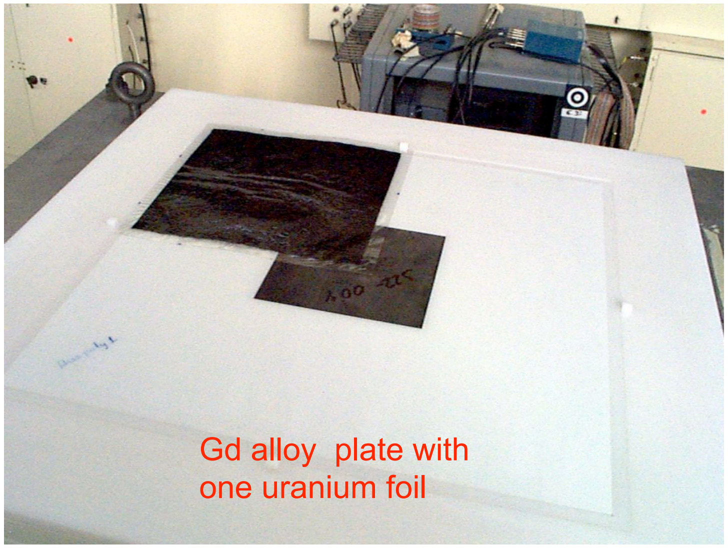
- ASME Code minimum imposed for borated stainless steel is 15 ft-lb and 0.015" lateral expansion
- Code Case N-728 initially approved without this restriction

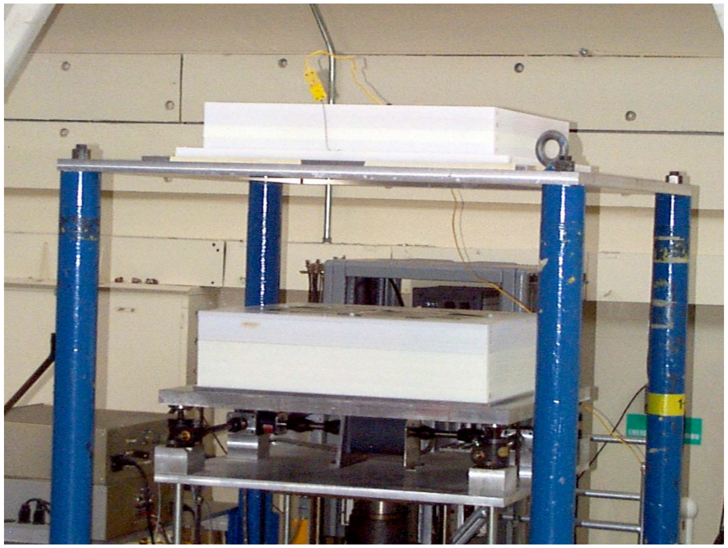
Low Cycle Fatigue



- Fatigue behavior similar to other Ni-Cr-Mo alloys especially at low reversed strains

Criticality Experiments



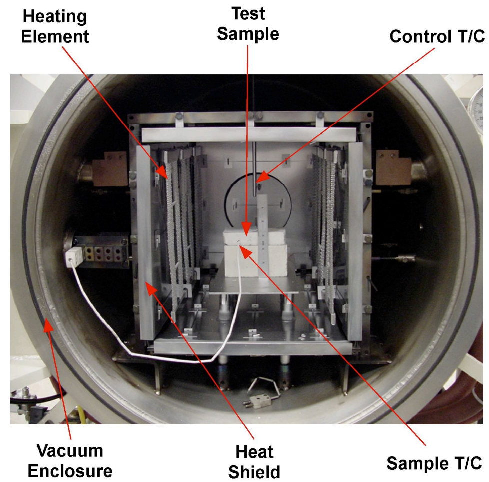


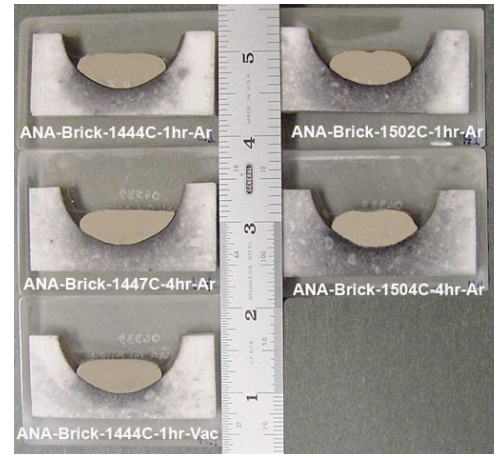
- Experiments were performed at the Los Alamos National Laboratory Criticality Experiments Facility
- The experiment consisted of interlaying highly enriched uranium foils with polyethylene and Gd alloy plates that simulate a fully moderated configuration in a critical system
- Initial measurements and calculations suggest that the negative worth of Gd alloy plates is about 8.8$ of reactivity
- Calculated negative worth of an equivalent volume of borated stainless steel plate (1.7% B) is 6.4$ of reactivity

Scale-up Issues

- Generic Issues:
  - Price and availability of gadolinium
  - Master alloy pilot study
  - Melt chemistry control – composition limits, virgin materials, scrap recycling/recovery
  - Chemistry standards
  - Safety
- VIM Studies - lining interactions, large ingot segregation (in progress)
- Secondary Melt Refining
- Ingot Reduction (in progress)
- Surface Conditioning
- Material Acceptance Standards
- Welding Development (in progress)
- Product Scale-up Confirmatory Analysis

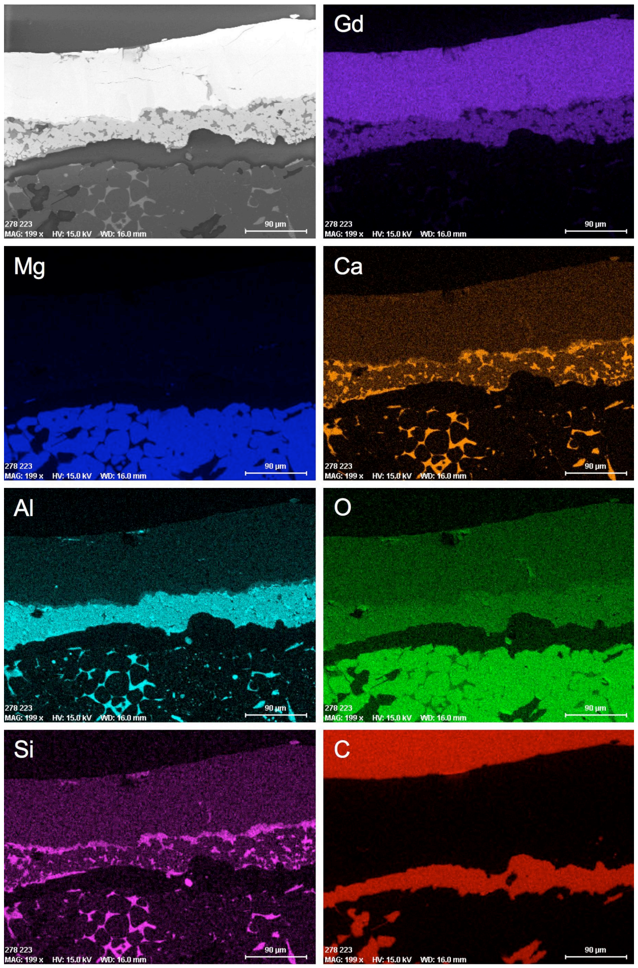
Furnace Lining Interactions

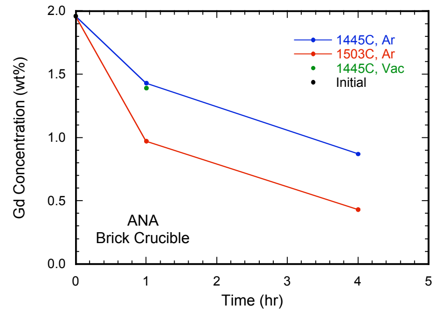


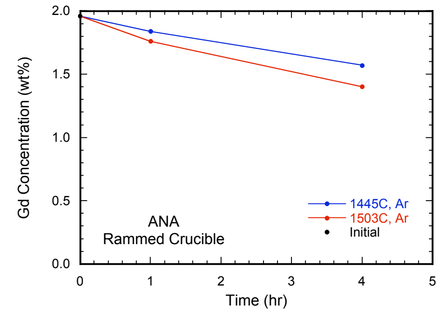


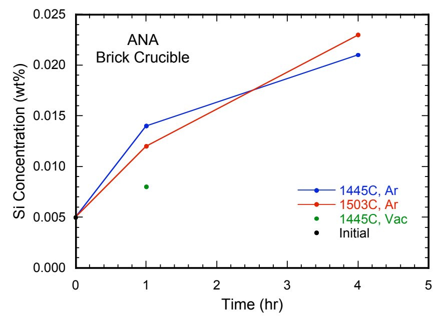
- **Basic approach was to fabricate crucibles from refractories of interest and expose them to ANA, and UNS N06455 at temperatures above the liquidus to various times**
- **Refractories included a typical brick and a typical rammed crucible**
- **Analysis consisted primarily of measurement of**
  - **the species liberated from the melt or refractory**
  - **the composition of the melt button**
  - **the nature and extent of the interfacial reactions**

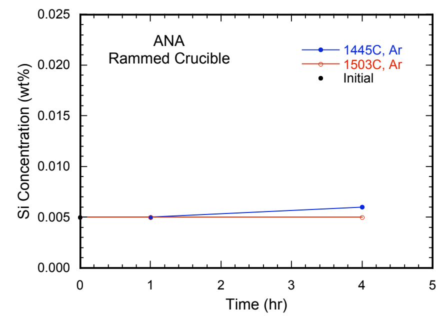
Furnace Lining Interactions









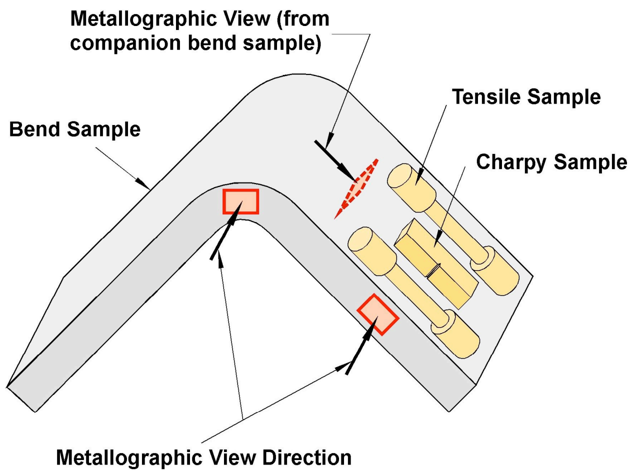


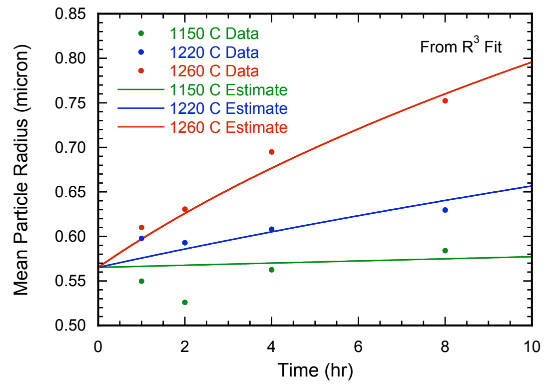
- **Results showed significant loss of Gd for both refractories**
- **Brick showed generally higher Gd loss and refractory uptake than rammed crucible**
- **Results were qualitatively in agreement with FACTSAGE estimates, but experiments were not taken to completion**

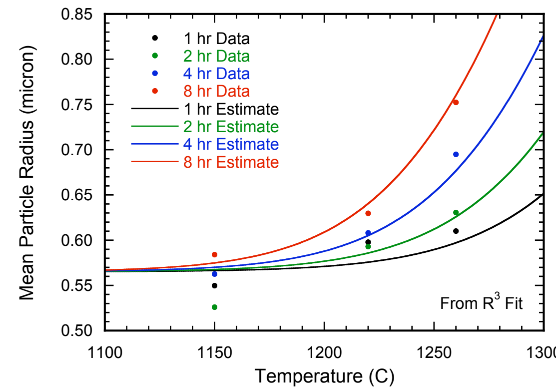
National Nuclear Security Administration

Sandia National Laboratories

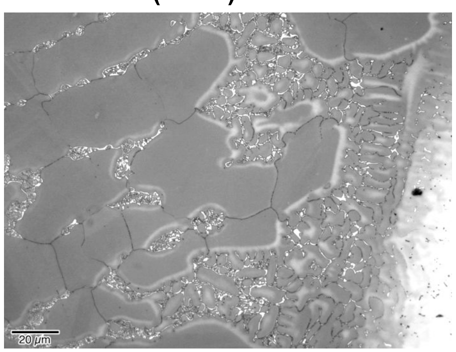
Bend Testing of Plate and Weldments



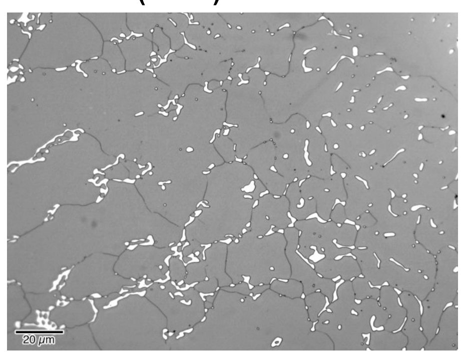




RMGD-2 (M326) As Welded



RMGD-2 (M326) PWHT 1163C/8hr



- **Base metal and weld bend tests were analyzed in detail experimentally and through kinetic modeling**
- **Of particular interest were the effects of additional annealing and PWHT**

Size Scale-Up - Reduction of 250 lb Ingots

Summary

- Ni-Cr-Mo-Gd alloys can be made with conventional ingot metallurgy techniques
- The alloys will meet all performance requirements
  - Mechanical properties will meet ASME requirements in post-welded heat treated condition (transportation issue)
  - Thermal neutron absorption performance of prototype alloys is consistent with published data
- Currently in LA for Commercial and DOE SNF
- Production material quantities ~24 months out