SAND2008-1010C

Nickel-Based Gadolinium Alloy for Neutron Adsorption in RAM Packages

PATRAM 2004

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Charles Robino[1], Ron Mizia[2], Paul McConnell[1]

Presented by Doug Ammerman[1]

[1]Sandia National Laboratories, Albuquerque USA

[2]Idaho National Engineering & Environmental Laboratory, Idaho Falls USA

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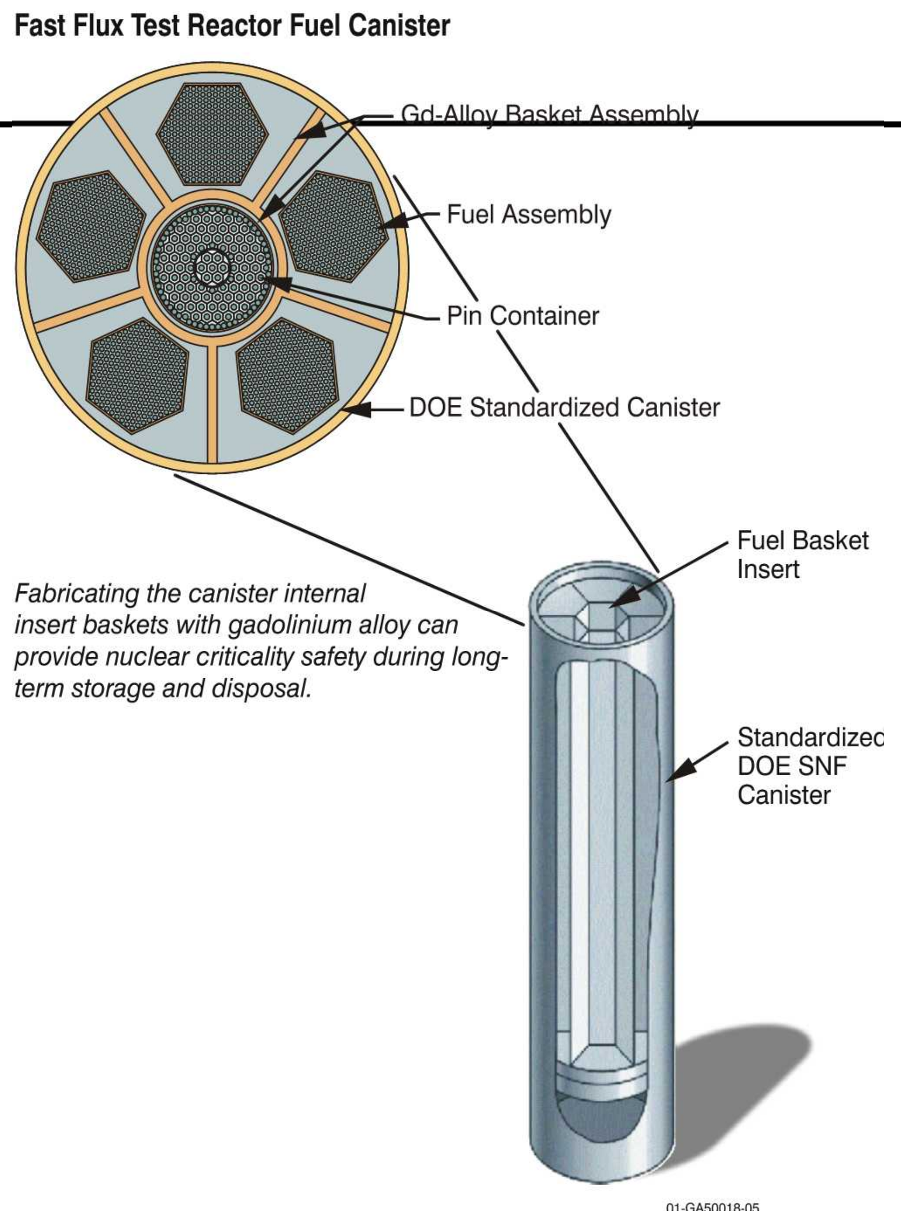
Project Description

- **Problem**:
- **Some types of USDOE spent nuclear fuel (SNF) contain highly enriched uranium.**
- **Final disposition of this SNF the repository may require criticality control during the regulatory period.**

- **Approach**:
- **SNF will be packaged in standardized canisters with baskets fabricated from corrosion resistant neutron absorbing materials.**

- **Benefits**:
- **Criticality control will be ensured for DOE SNF under fully flooded / degraded repository conditions.**
- **Decreased number of SNF packages going to repository with reduced handling and materials costs.**

DOE Standardized Canister



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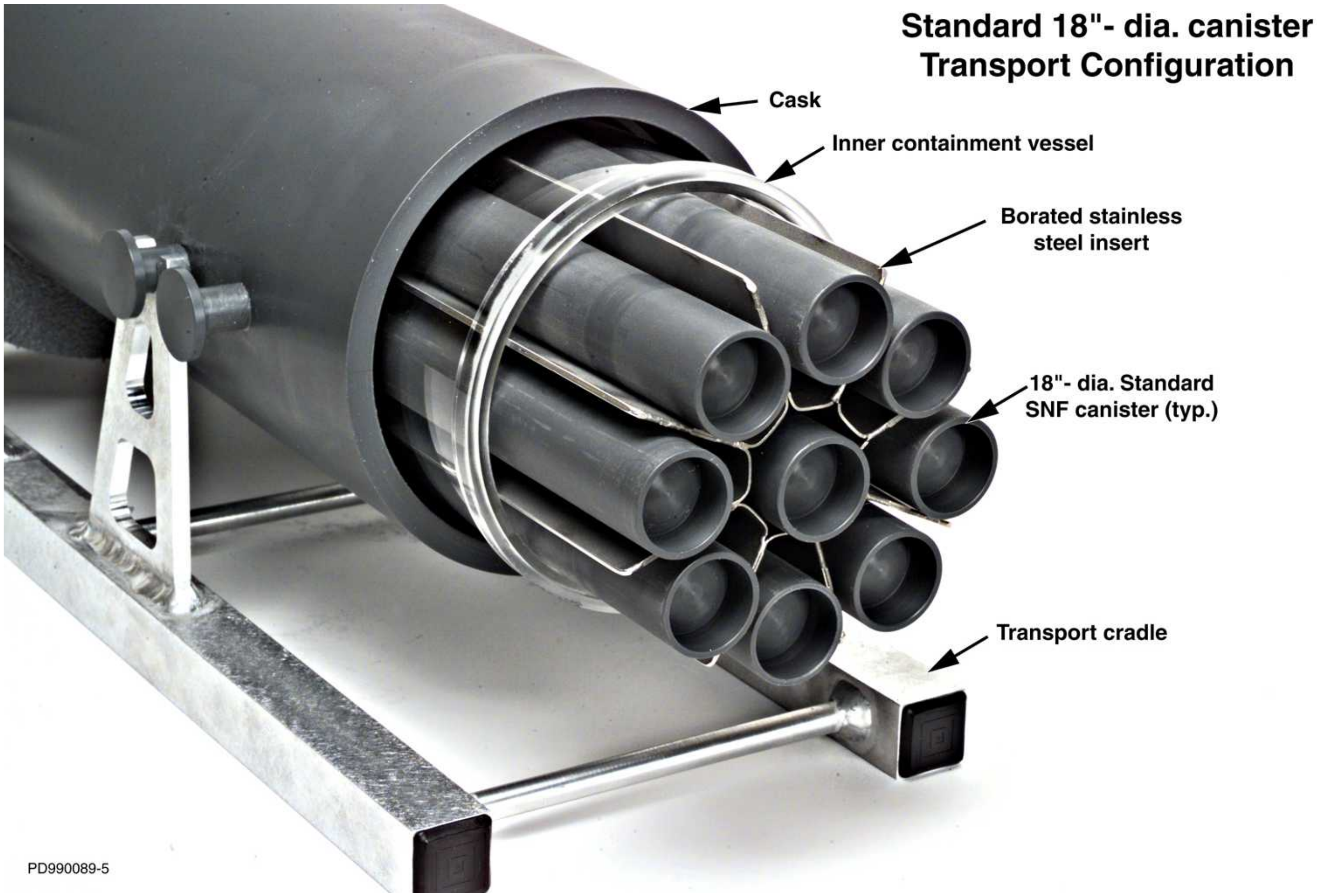
Gd-alloy would replace borated stainless steel in baskets



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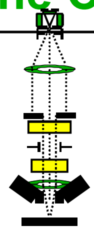
Tasks

- Develop a nickel-based corrosion-resistant alloy with a gadolinium addition. Gadolinium has a high thermal neutron absorption cross section.
- Determine effect of alloy microstructure on corrosion performance, mechanical properties, and thermal neutron absorption.
- Develop an American Society for Testing and Materials specification.
- Perform mechanical properties measurements for acceptance in Section III, Division 3 (NUPACK) of the ASME Code.

Project Workflow

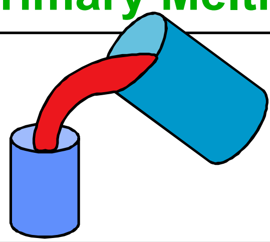
Define Compositions



Laboratory Evaluations

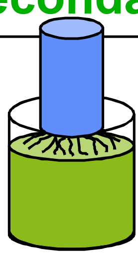
Fe-Ni-Cr-Gd
Ni-Cr-Mo-Gd

Primary Melting and Casting



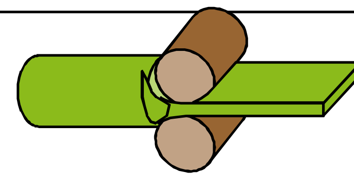
Vacuum Induction Melting (VIM)

Secondary Melting/Refining



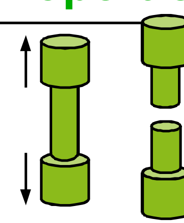
Vacuum Arc Remelting (VAR)

Hot Working/Rolling



Hot Ductility
Forging
Rolling

Properties Testing



Tensile
Charpy Impact
Corrosion
Weldability
Neutronics

Codes and Standards

ASTM Specification
ASME Code Section III
Commercialization

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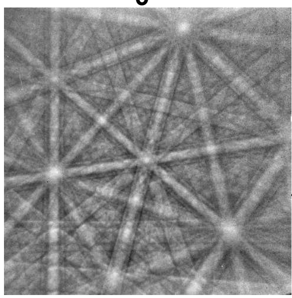
Ni-Cr-Mo-Gd Alloy Design

Element	UNS (N06455)	UNS (N06022)	UNS (N06059)
Mo	14.0-17.0	12.5-14.5	15.0-16.5
Cr	14.0-18.0	20.0-22.5	22.0-24.0
Fe	3.0 max	2.0-6.0	1.5 max
W	– – –	2.5-3.5	– – –
Co	2.0 max	2.5 max	0.3 max
C	0.015 max	0.015 max	0.010 max
Si	0.08 max	0.08 max	0.10 max
Mn	1.0 max	0.5 max	0.5 max
V	– – –	0.35 max	– – –
P	0.04 max	0.02 max	0.015 max
S	0.03 max	0.02 max	0.005 max
Ti	0.7 max	– – –	– – –
Ni	bal	bal	bal
Al	– – –	– – –	0.1-0.4
Cu	– – –	– – –	– – –

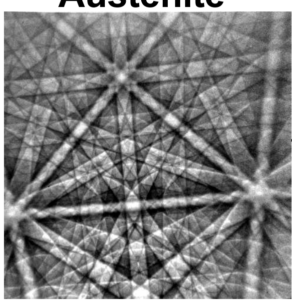
- Three commercial Ni-Cr-Mo alloys selected for initial target compositions.
- Initial target level of 2.25 wt% Gd identified from neutron absorption considerations.
- Final target level is 2.00 wt.% Gd

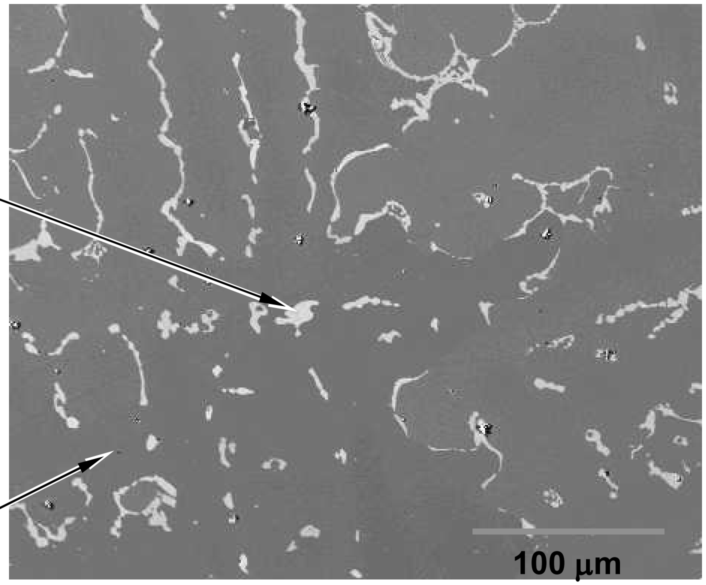
Typical as-cast microstructure

Ni_5Gd



Nickel Austenite





Element	Nominal ASTM Chemistry	Gadolinide
Mo	14.9	0.69
Cr	15.1	2.00
Fe	1.0 max	0.04
Co	0.3 max	---
C	0.010 max	---
Si	0.08 max	0.08
Mn	0.5 max	0.00
P	0.01 max	---
S	0.005 max	---
Ni	bal	63.13
N	0.010 max	---
Gd	2.0	35.26

- Composition of Ni_5Gd (gadolinide) was similar for a range of melt chemistries - no Gd observed in matrix.
- Matrix composition can be controlled by adjustment of bulk chemistry.

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Successful hot working of up to 2.4 wt% Gd ingots

VAR Ingot-as cast

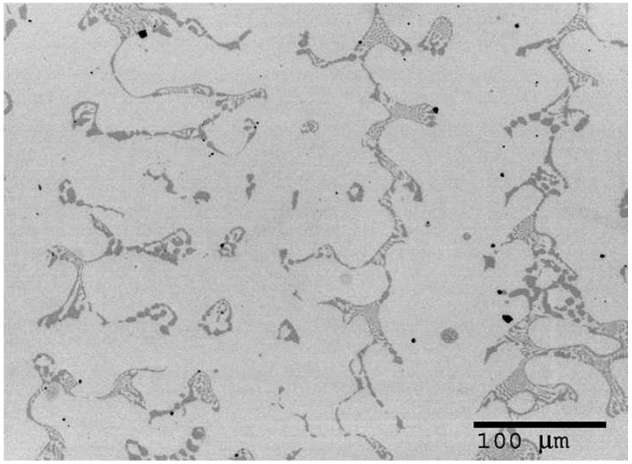
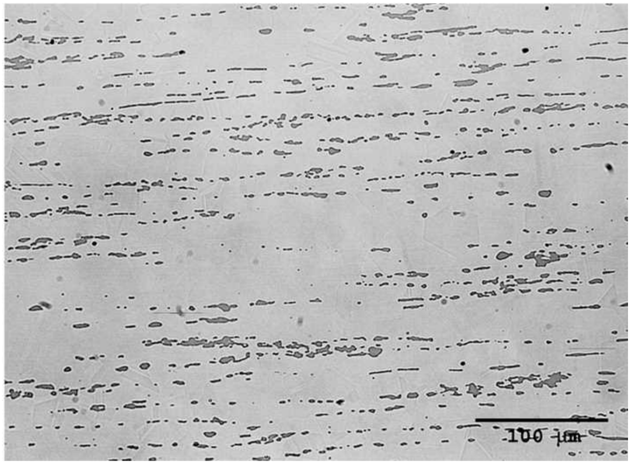


Plate (L-S orientation)



- Gadolinide distribution evolves during hot working.
- Some surface cracking was experienced.
- Differences between various heats have been observed.
- Further optimization of hot working procedures is underway.

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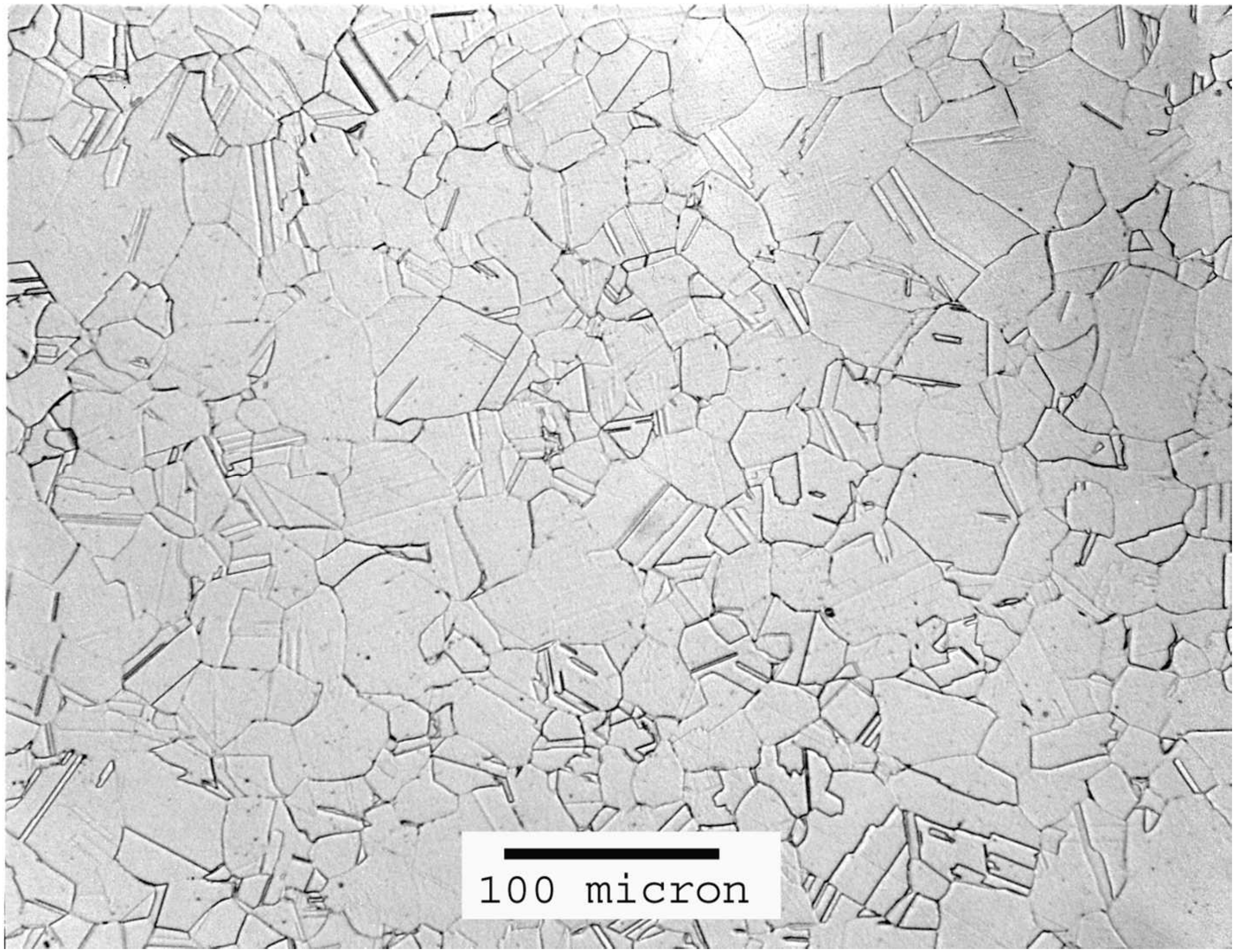
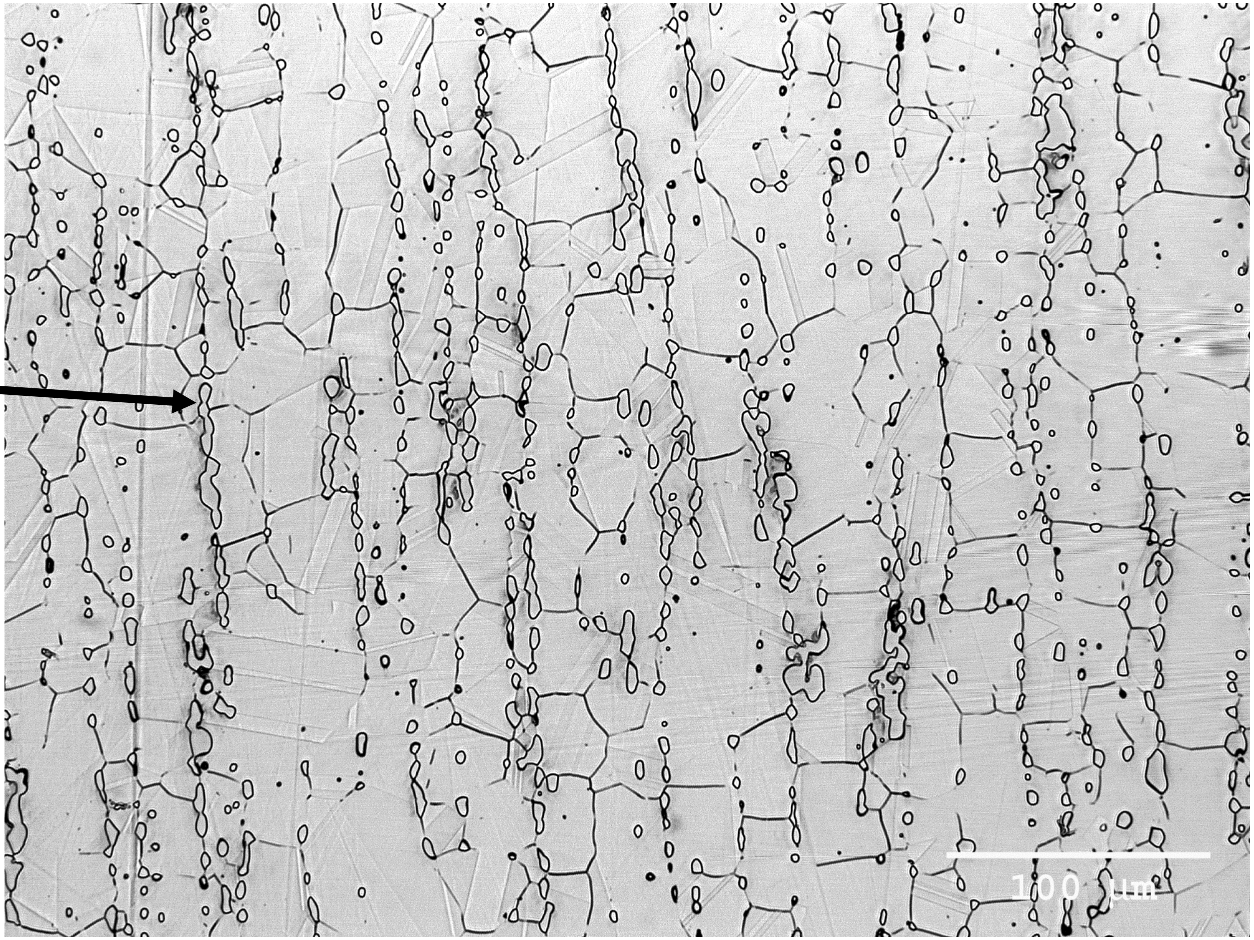
LOM of Nickel Alloy 22 (no Gd)



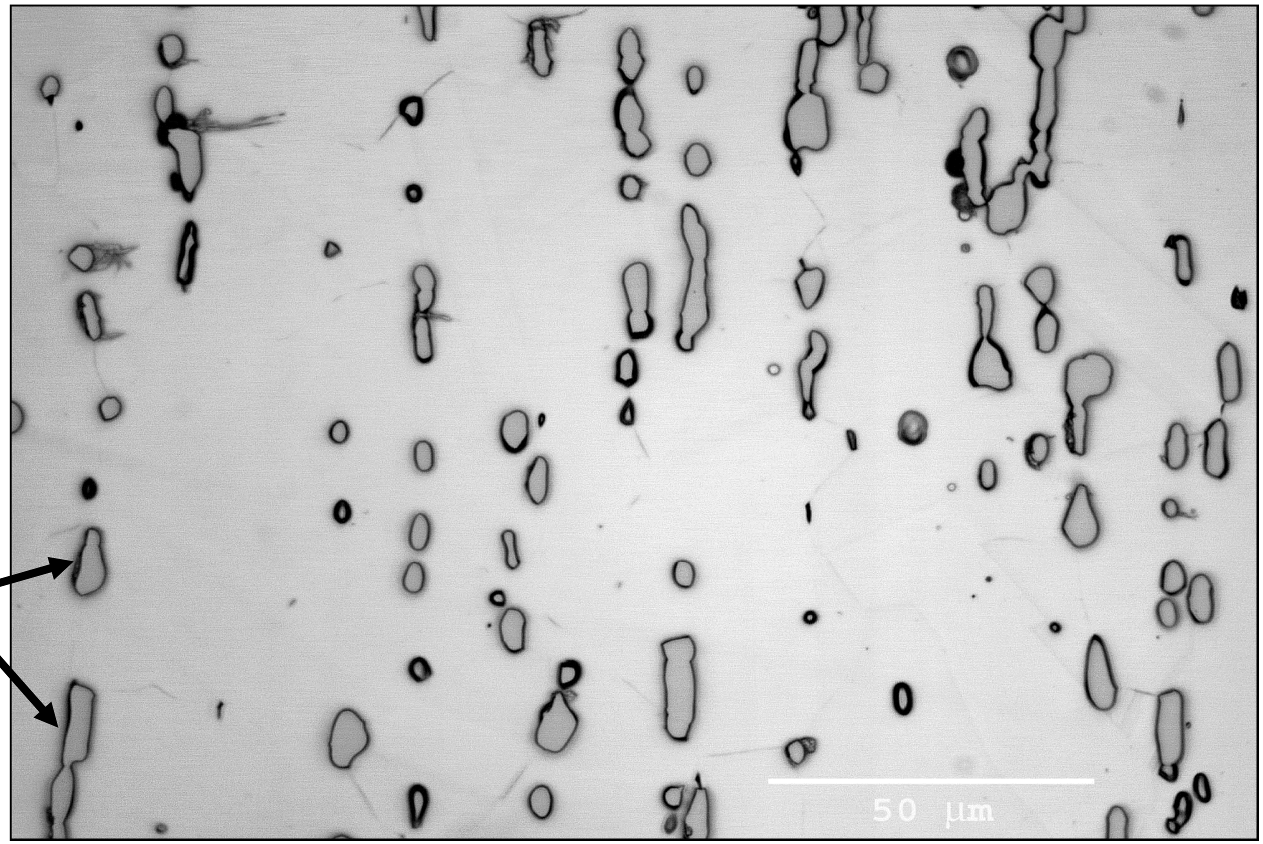
LOM of Heat M327

$(Ni,Cr)_5Gd$



LOM of Heat M327

$(Ni,Cr)_5Gd$



YMP Waste Package Criticality Control

- The Waste Package System for the YMP repository is designed so that the effective neutron multiplication factor (k_{eff}) is less than or equal to 0.95.

- For defense-in-depth, supplemental neutron absorber material in the form of Ni-Cr-Mo-Gd plates is required.

- The material must be resistant to leaching of neutron absorbing material through localized corrosion processes.

Microstructural Features and Mechanical Properties/Corrosion Performance

- The Ni-Cr-Mo alloys are resistant to corrosion by oxidizing and reducing environments. Repository waste package outer barrier material is also a nickel alloy.
- There is no solubility of Gd in austenite matrix of Ni-Cr-Mo alloys.
- A Gd rich, eutectic, secondary phase forms - $(Ni,Cr)_5Gd$.
- The two phase structure differentiates these alloys from other Ni-Cr-Mo alloys.
- This second phase reduces the ductility of the alloy. The strength of the alloy is increased.
- This second phase may be selectively attacked in some projected YMP in-drift environments.

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Codes, Standards, and Patent Status

- ASTM Specification B932-04 approved and published in ASTM Volume 2.04, May 2004.
- Alloy neutronics data (LANL) incorporated into International Handbook Of Evaluated Criticality Safety Benchmark Experiments.
- Intellectual Property covered under US Patent # 6,730,180 B1, “Neutron Absorber Alloy” and IDR #611, “Modified Neutron Absorbing Alloy”.
- ASME Code Case submitted August 2004 to ASME Section III, Division 3.

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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

Mechanical Properties for ASME

Alloy	Orientation	Tensile at 23°C YS (ksi)	UTS (ksi)	Elongation (%)	RA (%)	Charpy Impact -40°C	(Energy, ft-lb)	300°C
Melt 340	Long*	58.1	111	39	30	22.5	24	-
Melt 340	Trans*	58.8	104.3	28.6	22.6	15	15.7	-
Melt 322	Long	82.5	127.5	42.6	29.2	23.0	23.0	28.0
Melt 322	Trans	83.3	100.0	7.4	5.5	7.5	7.8	9.0
Melt 326	Long	60.9	118.5	46.1	35.2	—	27.3	—
Melt 326	Trans	60.0	104.7	22.3	18.6	—	14.1	—
Melt 327	Long	60.7	115.7	51.1	38.5	—	33.1	—
Melt 327	Trans	61.4	108.1	24.1	21.0	—	16.3	—
HV-0182 (as-rolled)	Long[a]	80.7	125.0	39.3	35.3	—	19.7	—
HV-0182 (as-rolled)	Trans[a]	86.3	116.7	22.6	21.3		15.7	—
HV-0182 (CR) (1093ºC/4hr./WQ)	Long[a]	53.8	114.3	44.3	35.3	—	38.7	—
HV-0182 (CR) (1093ºC/4hr./WQ)	Trans[a]	54.8	104.0	29.0	22.0	—	23.3	—

a. Cross rolled relative to primary rolling direction.

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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 wrought and as-welded condition (transportation issue).

  - Alloy is weldable.

  - Criticality control during regulatory period is ensured based on corrosion tests.
    - Thermal neutron absorption performance of prototype alloys is exceptional and consistent with published data.

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Project Summary (cont'd)

- ASTM Material Specification (B932-04) for Ni-Cr-Mo-Gd Alloy has been issued.

- Mechanical and physical properties data set for ASME Code Case (Section III,Division 3, NUPACK) submitted to ASME.

2005 Alloy Development Activities

- Continue ASME Code activities.
  - Additional mechanical property measurements may be required
    - fracture toughness, fatigue, creep.

- Further work on chemistry modifications.

- Complete welding program.
  - Qualify weld procedure.
  - Complete post-weld annealing studies.

Scale-Up issues for production-size heats

- Identify company to produce large plate.
  - Technical capability
  - Financial resources
- VIM Issues – form of gadolinium addition.
  - Metallic gadolinium will oxidize easily.
  - The use of a master melt feedstock ingot of eutectic composition (87% Ni, 13% Gd) has been demonstrated.
- VAR Issues
  - Technology transfer from SNL Liquid Metal Processing Facility.
- Conversion from round VAR ingot to slab for hot rolling.
  - VAR molds of various sizes (17” to 36”).
  - Minimum width requirement on rolling mill.

Scale-Up Issues (cont'd)

- **Scrap Control – Suppliers may be concerned about Gd contamination.**

- **Possible Suppliers**
  - ***Special Metals Corporation* – has provided VIM ingots and rolling services.**
  - ***Allegheny Technologies***
  - ***Santoku America* – has supplied VIM ingots and "master melt" starting feedstock ingots .**

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