

#### UNITED ST TES NUCLEAR REGULATORY COMMISSION

WASHINGTON, D.C. 20555-0001

March 4. 1998

**MEMORANDUM TO:** N. King Stablein, Acting Branch Chief ENGB/DWM/NMSS

FROM:

Richard A. Weller, Section Leader & RW **Engineering and Material Section** END./DWM/NMSS

Kimberly A. Gruss, General Engineer Kinbuly Am Suss **Engineering and Material Section** ENGB/DWM/NMSS

Narasi Sridhar, Manager N Sndhar **Corrosion Science and Process Engineering Element CNWRA** 

SUBJECT: SUMMARY OF MEETING HELD AT SAVANNAH RIVER SITE, DECEMBER 18, 1997, TO DISCUSS PLANS FOR THE US. NUCLEAR REGULATORY COMMISSION'S REVIEW OF ALUMINUM-BASED SPENT NUCLEAR FUEL

On December 18, 1997, a meeting was held between the U.S. Department of Energy (DOE)-Savannah River (SR), Westinghouse Savannah River Company (WSRC) and the U.S. Nuclear Regulatory Commission staff to facilitate NRC's review of DOE's aluminum-based spent nuclear fuel program, consistent with the August, 26, 1997, Interagency Agreement between NRC and DOE. In this regard, DOE has submitted 12 program reports, to date, for the staff's review. During this meeting, on-going and planned activities for the development of the direct co-disposal and melt and dilute technologies were discussed; the interrelationship of key DOE documents was described; and the expected DOE deliverables, schedules, and level of staff effort requirements for NRC review were discussed. Attachment 1 contains the meeting Agenda; Attachment 2 lists the meeting attendess; and Attachment 3 contains the overheads used during the meeting.

DOE-SR emphasized the need for NRC to focus its review on four main documents, "Evaluation of Co-Disposal Viability for Al-Clad DOE-Owned Spent Fuel" (both Phase I and Phase II documents), and "Alternate Aluminum Spent Nuclear Fuel Development Status Report" (both April 1997 and October 1997 versions). The additional documents provided by DOE-SR, prior to this meeting, contain background information that will aid NRC in its review of these four documents. A roadmap showing the interrelationship of the DOE documents was disseminated at the meeting and is attached (Attachment 4). DOE-SR indicated that it would like the NRC to conduct a topical review of the two disposal technologies, as opposed to an independent review of each document, and requested that NRC's preliminary review of the treatment options be completed in May to facilitate DOE's selection of a treatment option. DOE-SR also agreed to provide NRC with an overview document by the third week in January 1998 which would describe the following: 1) DOE's programmatic needs with respect to NRC's

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N.K. Stablein

review of the documents; 2) the relationship between various DOE documents; 3) expected NRC deliverables pertaining to the melt and dilute and co-disposal technologies; 4) important DOE milestones that may affect NRC's review schedule; and 5) additional documents requiring NRC review.

DOE-SR proposed having monthly teleconference meetings with NRC, starting in January and continuing through the time when a treatment option has been selected. Questions regarding programmatic concerns and the transmission of new documents will be communicated at these meetings and will be handled through the office of A. Lee Watkins in accordance with the Interagency Agreement. Questions concerning the technical content of DOE documents will be directed to WSRC staff. Additional references cited in the DOE documents can be obtained through DOE-SR or WSRC staffs. If needed, an NRC/DOE management coordination meeting will be scheduled following DOE-SR's development and submittal of the aforementioned program overview document.

Attachments: As stated

cc: J. Ridley/DOE-SR (w/o)

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#### NRC and DOE-SR/WSRC Alternative Technology Review December 18, 1997

#### Agenda

#### **OBJECTIVES:**

- 1. Facilitate NRC's review by describing activities completed and planned for developing direct co-disposal and melt and dilute technologies
- 2. Provide the NRC a roadmap/matrix of the interrelationships of the documents
- 3. Establish a working level familiarity of the participants.
- 4. Define deliverables, schedules and resource requirements for the NRC review.

#### LOCATION: Savannah River Site - SRTC

#### SCHEDULE:

- 8:00 8:30 Badging
- 8:30 9:00 Introductions of NRC, DOE & WSRC participants
   include description of roles/responsibilities within WSRC, DOE and NRC
- 9:30 10:00 Overview of Alternate Technology Program and Schedule Status
- 10:00 10:15 Break
- 10:30 11:30 Direct/Co-Disposal Discussion -WSRC review of work completed and planned -Matrix/ Roadmap/Priority of the related reference documents -Comments/Questions from NRC
- 11:30 12:30 Lunch
- 12:30 3:00 Melt/Dilute Discussion -Tour of SRTC lab
  - same topics as above
- 3:00 4:00 Planning for NRC Review
  - Review of WSRC documents and priority
  - NRC deliverables and schedule
  - Assessment of NRC funding and resources
  - Tentative schedule of future meeting

#### MEETING ATTENDANCE

#### Primary:

Jean Ridley, DOE-SR, Program Engineer, NRC Coordinator	(803) 557-3758
Mark Barlow, WSRC, Alt. Technology Program Manager	(803) 557-8021
Natraj Iyer, WSRC, Lead Scientist for Alternative Technology	(803) 725-2695
Harold Peacock, WSRC, Lead Melt & Dilute Technology	(803) 725-1277
Bob Sindelar, WSRC, Lead Direct/Co-Disposal Technology	(803) 725-5298
Bruce Wiersma, WSRC, Test Protocol	(803) 725-5439
Rick Weller, NRC, Section Leader	(301) 415-7287
Kim Gruss, NRC, Materials Engineer	(301) 415-6680
Narasi Sridhar, Southwest Research Institute, Manager	(210) 522-5538
Secondary:	
Randall Ponik, DOE-SR, Program Mgr. Alt. Technology	(803) 557-3236
Howard Eckert, DOE-HQ, EM-67	(301) 903-7173
Scott Gladson, LMITCO, National Spent Fuel Program	(208) 526-9749
Thad Adams, WSRC, Melt & Dilute	(803) 725-5510
Jim Howell, WSRC, Melt & Dilute	(803) 725-5538
Dennis Vinson, WSRC, Direct/Co-disposal	(803) 725-5298
Additional Attendee:	

Charlie Anderson, DOE-SR, Director Spent Fuel Division

**ATTACHMENT 2** 

(803) 557-3828

#### ATTACHMENT 3

## **Alternative Al SNF Treatment Technology Program**

## Development of Direct/Co-Disposal Technology for Aluminum-Based Spent Nuclear Fuel

### R. L. Sindelar

Program Overview Presentation to Nuclear Regulatory Commission Staff December 18, 1997

## Direct/Co-Disposal Technology Development Program Objectives

# • Disposition Path for Al SNF:

- » Interim Dry Storage at SRS for up to 40+ Years
- » Ultimate Disposal in Geologic Repository
- Develop Technical Bases to Meet Interim Dry Storage Requirements
- Develop Technical Bases to Meet Repository Requirements and for Input to PA

## **Interim Dry Storage of Aluminum-Based SNF**

- Established Requirements (Acceptance Criteria) for Drying and Storage Conditions of Al SNF
- Approach:
  - » Limit Changes in SNF Condition Throughout Handling and Storage Period (Enables Retrievability)
  - » Indentify Environmental Conditions such that SNF Changes Remains Within Acceptable
  - » Develop Predictive Models to Demonstrate Acceptable Conditions

## **Interim Dry Storage of Aluminum-Based SNF**

- Acceptable Changes (Enables Retrievability of SNF):
  - » Consumption of Clad/Fuel < 0.003"
  - » No Gross Rupture of Clad
  - » Plastic Deformation < Clearance of
  - » Release of Radionuclides < Max. Permissible Concentrations
- Environmental Limits:
  - » Fuel Temperature: 200°C
  - » Limit Free Water to Avoid Excessive H<sub>2</sub> Build-Up (Flammability) and Consumption by Corrosion
- Requirements in WSRC-TR-95-0347; WSRC-TR-97-00345

# Direct/Co-Disposal Technology Development for Geologic Repository

### Attributes:

- Near term Implementation into Road-Ready Fackage
  - » Interim Storage Technology Developed
- Minimal Conditioning Required



### Challenges:

- NRC and Stakeholder Acceptance of HEU into Geologic Repository
  - » Proliferation Issue
  - » Criticality Issue
- Characterization Requirements
- Qualification of Aluminum SNF Form

## Direct/Co-Disposal Technology Development Program Activities

# **Interim Dry Storage**

- Thermal Analyses
- Degradation Testing & Anal.
  - Corrosion (vapor
  - & aqueous)
  - Creep
  - Volatile species
  - release
  - Hydrogen Generation
- Demonstrate Reqts. are Met

# **Repository Disposal**

- Thermal Analyses
- Degradation Testing & Anal.
  - Corrosion (vapor & aqueous)
  - Creep
  - Volatile species
  - release
  - Hydrogen Generation
  - Particulate formation
  - Material Reconfiguration
- Criticality Analyses
- Input Resalts into PA
- Demonstrate Reqts. are Met

## Direct/Co-Disposal Validation of Interim Storage Criteria

- Storage Criteria
  - » Cladding Temperature Limit: 200°C
  - » Free Water Limited
  - » Sealed Canister; Inert Gas Backfill
- Drying Specfication
  - » Preliminary: Dry to < 5 Torr at RT (Unheated Vacuum Drying)
- Validation of Criteria
  - » Instrumented, Shielded
     Test Canister
  - » Store Live SNF in CY98
  - » Hot Cell Tests



### Direct/Co-Disposal Thermal Analysis



### **Direct/Co-Disposal Thermal Analysis**

### Repository Storage

- Model Using Codes:
   2D FIDAP; CFDS CFX; Closed-Form
- » Fuel T: Dependent on Thermal Loadings & Model; e.g. 175 to 300°C Initial T
- » Complete Parametric and Best-Estimate Analyses



## Direct/Co-Disposal Degradation & Release Analyses

- Degradation Modes & Mechanisms
  - Creep Modeling
  - Corrosion (Vapor & Aqueous) Testing & Modeling
  - Hydrogen Generation Modeling
  - Pyrophoricity Evaluation
- Volatile Fission Product Release (w/ANL) Testing & Modeling
- Fuel Materials Reconfiguration (Chemical/Physical Forms) in Repository
  - Particulate Formation

## Direct/Co-Disposal- Creep Analyses

•CREEP: displacement of fuel elements can exceed 0.1" at temperatures > 200°C

- Materials Test Reactor fuel design

- FE analysis, self-weight loading



- Testing & Modeling of Al Cladding and Fuel Materials
- Effects of Temperature, Relative Humidity, Water Chemistry, Radiolysis
- Alloy Dependency
- Fuel Material and Microstructure Dependency

## TEMPERATURE DEPENDENT PARABOLIC (PARALINEAR) CORROSION of AL6061



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# TEMPERATURE DEPENDENT BREAK-AWAY CORROSION AL5052



### Alloy Dependent Corrosion Mechanisms

### AL1100 Oxide Film (optical)

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### AL5052 (SEM,300X)



#### AL5052 Internal Oxidation (optical)



AL5052 (SEM,4000X)



SRTC 4/30/97

## Aluminum 6061 Exposed to 250°C Water Vapor (6 Months, 100% R. H.)



- Degraded in WP -Failure Mechanisms Evaluated; Geochemistry Analysis using EQ3/6; Criticality Analysis using MCNP4A
- Parametric Analysis (water, geometry, U-235 volume/distribution, absorbers, materials, reflectors)
- Use GdPO4, otherwise solubility issues
- Results: Fully Loaded Can of MIT, Flooded
  - » Intact canister, intact basket, degraded fuel: 1kg Gd in plates
  - » Intact canister, degraded C steel basket: 0.12 kg Gd
  - » Intact canister, degraded SS basket: 0.25 kg Gd
  - » Degraded in WP, fuel on top: 0.20 kg Gd with fuel, no Fe credit
  - » Degraded in WP, fuel on bottom: 0.1 kg Gd with fuel, no Fe credit
  - » Degraded in WP, fuel homogenized with clayey material, subcritical w/o credit for Gd and Fe
- Qs: 1) Absolute or, no impact on PA?;
  2) Time period of demonstration of keff?

## **FY98** Activities

- Al SNF Form Performance
  - Materials Degradation Testing and Modeling for Repository Conditions
  - Criticality Analysis of Direct and Melt-Dilute

Forms

- ASTM Test Protocols for Metallic Waste Forms
- Performance Assessment
- SNF Form Qualification
  - Assemble Initial "Technical Package" for DIS for

License

- Interim Storage Criteria
  - Validation Testing & Analysis
  - ASTM Standard Guide for Extended Dry Storage

# MELT-DILUTE PROCESS TECHNOLOGY FOR SPENT NUCLEAR FUEL STORAGE

H.B. PEACOCK

## Alternative SNF Treatment Melt-Dilute Technology

#### Attributes:

- Dilute HEU to LEU Levels
- Waste Form Homogenous
  - Minimal Characterization Requirements
- Volume Reduction up to 70%

#### **Challenges:**

- Integrated Process Requires Validation
- Wasteform Qualification
  - Isotopic Dilution
  - Ternary Constituents Effect
- Off-Gas System and Waste Streams





Fig. 1. Several fuel element configurations.

# Irradiated MTR Type Fuels





U-Al<sub>x</sub>



U<sub>3</sub>Si<sub>2</sub>-Al

# Melt-Dilute Fow Diagram



# Melt-Dilution Process Dilution Mechanism

- Dilute to 2 to 20%
- Dilute by Addition of
   Depleted U and Al
- Dilute to Target Compositions:
  Eutectic (13.8 wt% U)
  - UAl4 or UAl3: Intermetallics



# Melt-Dilute Process Options



## Melt-Dilute Al-SNF Form Development Process Cycle Option



Time

# **Process Conditions**

	Low Temperature	High Temperature		
Temperature	800 - 1000	1000 - 1600		
Composition, wt	.% 13*, 30	30, 67+		
Crucible Option	s Steel/Graphite	Graphite		

\* Eutectic Composition
+ UAl<sub>4</sub>, Intermetallic Composition

## Melt-Dilute Al-SNF Form Development Casting Techniques



Furnace Cooling

Slow Cool ~5 Hrs Crucible - One Time Use

Tilt and Pour



Compatibility with Automation Compatibility with Off-Gas System

#### Bottom Pour



Commercial Technology

# Crucible - Melt Interactions

Steel Crucibles - Low Temperature Graphite - High Temperature Ceramic - High Temperature



50 wt% 1400 °C

Carbon Steel Crucible Experiments Uranium-Aluminum Alloys 13.2-50 wt%U



Steel - Metal Interactions

# **Casting Compositions**

- Eutectic (13.2 wt% Uranium Alloy)
  - Low Liquidus Tempe5ratuare
  - •No Segregation during Solidification
  - •Potential for Natural Galvanic Couping (two phases)
- •30 wt% Uranium Alloy
  - Experience with Cast U-Al SRS Reactor Fuel
  - •Complex Microstructure
  - •Liquidus Temperature Below 1000 °C
- 67 wt% Uranium Alloy
  - Homogeneity -- Monolith
  - Corrosion Resistance (expected)
  - High Liquidus Temperature

# Microstructure of Cast U-Al Alloys


# Aluminum-Silicon-Uranium Ternary Phase Diagram

950° C ISOTHERM OF THE U-AI-SI TEPNARY SYSTEM



## Effect of Terrnary Constituents

- Aluminum Cladding Alloys and Uranium
  - 1100, 5052, AND 6061
  - Major Impurities: Si, Mg, Fe, Mn
- •Activation Products (Si, Fe, Mn, Zn...)
- •Fission Products (Cs, Sr, Tc, Rb, ...)
- •Partioning of Ternary Elements
- •Phase Stability

## Microstructure of U-Al Alloys



Uranium-30 wt% Aluminum

Iron Partitions to UAl<sub>4</sub>



### Melt-Dilute Process Process Simplicity

Process Scale Up

- Apparatus for Full Scale MTR
- Resistance Furnace
- Induction Furnace
- Stirrer
  - Mechanical
  - Induction
- Start-Up Tests of M-D Process Using Resistance Furnace



Resistance Furnace Assembly Melt-Dilute Process for Full-Scale MTR

### Melt-Dilution Process Off-Gas System

CARACTER CONTRACTOR

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## Melt-Dilution Process Off-Gas System Concept

SCHREEK CONTRACTOR



PROCESS SCHEMATIC

# Offgas Approach

- Provide cold surfaces for solids deposition
- Filter remaining aerosols
- Dissolve these isotopes in aqueous solution and send to HLW facilities
- Release gases (Dose has been shown to be much less than permissible limits)

# Fission Product Forms Expected

- Cs: Cs metal, Cs oxides, CsI, Cs<sub>2</sub>Te
- Kr: Kr gas
- Tc: Tc metal
- I: CsI, I<sub>2</sub>

### Al-SNF Form Stability Radionuclide Inventory

Contraction -



\* Reference FA after 10 yrs wet storage

## **Fission Product Quantities**

•	Yield, M	<u>laximum m</u>	ole fraction	<u>, max g/g Al</u>
•	Cs+Rb:	0.22	0.0031	0.016
•	Tc:	0.06	0.0008	0.003
•	RE:	0.53	0.007	0.04
•	Sr:	0.09	0.0013	0.004
•	I:	0.009	0.0001	0.0006
۲	Te + Se:	0.05	0.0007	0.003

## Amount of Fission

- Mole fraction U in fuel as fabricated
  - 0.005 to 0.020
- Burnup and mole fraction fissions
  - Up to 70% (0.0035 to 0.014)
- Fission Product quantities (mole fraction)
  - = FP yield x mole fraction fissions

### Volume Reduction

V



Reference: Direct Disposal 1400 Canisters

### **Proliferation Resistance**



Melt-Dilute => Isotopic Dilution Because of Liquid Phase Processing • Ready Separation of Enriched Uranium not Possible

### Criticality



U-Al-B Ternary Phase Diagram

Interstitial Solid Solution with B

- Boron Will Form Compounds of U or UAlx
- Efficacy of Poison Related to UAlx Degradation Rate

### Al-SNF Form Stability Chemical Stability

Preliminary Results of Electrochemical Tests

Corrosion/Dissolution Resistance of Al + UAl4 > Al + UAl3 + UAl4



As Fabricated MTR Assembly

M-D Waste Form



Tailored Microstructure to Optimize Chemical Stability



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Preferential Corrosion of UAl3 to UAl4



Irradiated Microstructure Al + UA13 + UA14



M-D Waste Form Al + UAl4



Preferential Dissolution of Al UAl4 Stringers Intact

# In Process Melt Characterization

- Total Uranium Glow Discharge or Density Measurements
- Waste Form Composition Glow Discharge

Density Measurement:As CastPressed1924.7 (24.3 wt%)

Glow Discharge Emission Spectroscopy Method:

- Measure U, Al, major trace elements
  - Qualitative & Quantitative Technique
  - $\sim$ 5% accuracy

# In Process Melt Characterization

- U235 or Burn-Up
- Total Uranium
- •Waste Form Composition

#### Process Strategy:

- Assume No Burn-Up
   => Consistent Composition but Variable Dilution (<20%)</li>
- Measure Burn-Up or U235: Characterization Costs

### Melt-Dilute Al-SNF Form Process Development Activities

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- Casting Technique Development
- Off-Gas System Development

- Evaluation of Stirring Techniques
- Surrogate Melts and Fission Product Partitioning
- Process Adaptation for Oxide and Silicide SNF
- Evaluation of Crucible Life
- Effects of Ternary Constituents (Cladding Alloys)
- Evaluation of Porosity Effects
- Evaluation of Secondary Waste Streams
- Evaluation of Melt Slag
- Process Control Techniques
- Evaluation of Melt Sampling Techniques
- Evaluation of Poison Additions
- Waste Form Characterization
  - Microstructural Evolution
  - Performance Evaluation

### **Alternative Al SNF Treatment Technology Program**

### **Development of Direct/Co-Disposal Technology for Aluminum-Based Spent Nuclear Fuel**

R. L. Sindelar

Program Overview Presentation to Nuclear Regulatory Commission Staff December 18, 1997

### Direct/Co-Disposal Technology Development Program Objectives

- Disposition Path for Al SNF:
  - » Interim Dry Storage at SRS for up to 40+ Years
  - » Ultimate Disposal in Geologic Repository
- Develop Technical Bases to Meet Interim Dry Storage Requirements
- Develop Technical Bases to Meet Repository Requirements and for Input to PA

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Attributes:

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#### Challenges:

- NRC and Stakeholder Acceptance of HEU into Geologic Repository
  - » Proliferation Issue
  - » Criticality Issue
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- Qualification of Aluminum SNF Form

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  - Volatile species

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- Hydrogen Generation
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  - Material Reconfiguration
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### Direct/Co-Disposal Validation of Interim Storage Criteria

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#### Direct/Co-Disposal Thermal Analysis



#### **Direct/Co-Disposal Thermal Analysis**

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- Testing & Modeling of Al Cladding and Fuel Materials
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- Alloy Dependency
- Fuel Material and Microstructure Dependency

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## TEMPERATURE DEPENDENT BREAK-AWAY CORROSION AL5052



#### Alloy Dependent Corrosion Mechanisms

#### AL1100 Oxide Film (optical)



#### AL5052 (SEM,300X)



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#### AL5052 Internal Oxidation (optical)



AL5052 (SEM,4000X)



### Aluminum 6061 Exposed to 250°C Water Vapor (6 Months, 100% R. H.)

#### Exposed Specimen



#### Chipped Area



#### Section in the Chipped Area



#### **Direct/Co-Disposal-** Criticality Analysis

- Degraded in WP -Failure Mechanisms Evaluated; Geochemistry Analysis using EQ3/6; Criticality Analysis using MCNP4A
- Parametric Analysis (water, geometry, U-235 volume/distribution, absorbers, materials, reflectors)
- Use GdPO4, otherwise solubility issues
- Results: Fully Loaded Can of MIT, Flooded
  - » Intact canister, intact basket, degraded fuel: 1kg Gd in plates
  - » Intact canister, degraded C steel basket: 0.12 kg Gd
  - » Intact canister, degraded SS basket: 0.25 kg Gd
  - » Degraded in WP, fuel on top: 0.20 kg Gd with fuel, no Fe credit
  - » Degraded in WP, fuel on bottom: 0.1 kg Gd with fuel, no Fe credit
  - » Degraded in WP, fuel homogenized with clayey material, subcritical w/o credit for Gd and Fe
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  - Materials Degradation Testing and Modeling for Repository Conditions
  - Criticality Analysis of Direct and Melt-Dilute

Forms

- ASTM Test Protocols for Metallic Waste Forms
- Performance Assessment
- SNF Form Qualification
  - Assemble Initial "Technical Package" for DIS for License
- Interim Storage Criteria
  - Validation Testing & Analysis
  - ASTM Standard Guide for Extended Dry Storage
# MELT-DILUTE PROCESS TECHNOLOGY FOR SPENT NUCLEAR FUEL STORAGE

H.B. PEACOCK

## Alternative SNF Treatment Melt-Dilute Technology

#### **Attributes:**

- Dilute HEU to LEU Levels
- Waste Form Homogenous
  - Minimal Characterization Requirements
- Volume Reduction up to 70%

#### Challenges:

- Integrated Process Requires Validation
- Wasteform Qualification
  - Isotopic Dilution
  - Ternary Constituents Effect
- Off-Gas System and Waste Streams





Fig. 1. Several fuel element configurations.

# Irradiated MTR Type Fuels





U-Al<sub>x</sub>



U<sub>3</sub>Si<sub>2</sub>-Al

# Melt-Dilute Fow Diagram



# Melt-Dilution Process Dilution Mechanism

- Dilute to 2 to 20%
- Dilute by Addition of Depleted U and Al
- Dilute to Target Compositions:
  - Eutectic (13.8 wt% U)
  - UAl4 or UAl3: Intermetallics



# Melt-Dilute Process Options



## Melt-Dilute Al-SNF Form Development Process Cycle Option



Time

# **Process Conditions**

	Low Temperature	High Temperature
Temperature	800 - 1000	1000 - 1600
Composition, v	vt% 13*, 30	30, 67+
Crucible Option	ns Steel/Graphite	Graphite

\* Eutectic Composition
+ UAl<sub>4</sub>, Intermetallic Composition

## Melt-Dilute Al-SNF Form Development Casting Techniques

#### Furnace Cooling



Slow Cool ~5 Hrs Crucible - One Time Use

#### Tilt and Pour



Compatibility with Automation Compatibility with Off-Gas System

#### Bottom Pour

121 - Carlos Car



#### Commercial Technology

# Crucible - Melt Interactions

Steel Crucibles - Low Temperature Graphite - High Temperature Ceramic - High Temperature



50 wt% 1400 °C

Carbon Steel Crucible Experiments Uranium-Aluminum Alloys 13,2-50 wt%U



Steel - Metal Interactions

# **Casting Compositions**

- Eutectic (13.2 wt% Uranium Alloy)
  Low Liquidus Tempe5ratuare
  No Segregation during Solidification
  Potential for Natural Galvanic Couping (two phases)
- •30 wt% Uranium Alloy
  - Experience with Cast U-Al SRS Reactor Fuel
  - Complex Microstructure
  - •Liquidus Temperature Below 1000 °C
- 67 wt% Uranium Alloy
  - Homogeneity -- Monolith
  - Corrosion Resistance (expected)
  - High Liquidus Temperature

# Microstructure of Cast U-Al Alloys



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#### DOCUMENTS FROM THE ALTERNATE TECHNOLOGY PROGRAM--ROADMAP FOR THEIR RELATIONSHIP TO THE PROGRAM (12/97)

The following table provides information regarding the relationship of several technical documents to the Savannah River Site Alternate Technology Program.

These documents were part of the initial package of information sent to the NRC and are listed in order of importance in the evaluation of technologies (direct/co-disposal and melt-dilute) being developed under the ATP.

Title	Relationship to SRS ATP
Evaluation of Codisposal Viability for Al-Clad DOE Owned Spent Fuel: Phase 1, Intact Codisposal Canister	This is a technical report to evaluate the reactivity of the direct-disposed Al SNF form within an intact (non-degraded) DOE SNF co-disposal canister. The purpose was to demonstrate that the 10CFR60 requirements for criticality are met for this disposal configuration.
	Structural integrity analysis of the canister against drop loads and thermal analysis of the co-disposal configuration were also part of the report. Additional criticality analyses are in progress and include degradation and its impact of the direct-disposed Al SNF form within a waste package and similar analyses for the melt-dilute form. Additional thermal analyses of disposal configurations of the direct-disposed and melt-dilute Al SNF forms in the WP are in progress.
Alternate Aluminum SNF Treatment Technology Development Status Report (WSRC-TR-97-00345, October 1997)	This is the second and latest technical report to provide the status of the development activities under the ATP technical programs. The four main task technical programs are:1) Direct/Co-Disposal Technology Development.; 2) Melt-Dilute Technology Development.; 3) Test Protocol Technology Development; and 4) Characterization Technology Development. Task technical programs 3 and 4 have been combined in FY98.
Acceptance Criteria for Interim Dry Storage of Al Alloy Clad SNF	This is a technical report prepared prior to the ATP and provides the requirements, with technical bases, for the dry storage of Al-clad SNF. The SRS-developed requirements are an expansion of those 10CFR72 requirements for maintaining fuel integrity throughout handling and storage. These requirements were adopted in the ATP as part of the preliminary requirements for road-ready storage which were listed in the above status report.
Evaluation of Corrosion of Al- Based Reactor Fuel Cladding Materials During Dry Storage	This is a technical report providing part of the technical bases for safe interim dry storage of Al SNF. It describes tests and initial development of models for vapor corrosion degradation of Al SNF materials in pure water vapors. The results and conclusions have been updated in the following report.
Vapor Corrosion of Aluminum Cladding Alloys and Aluminum- Uranium Fuel Materials in Storage Environments	This is a technical report containing part of the bases for safe interim dry storage of Al SNF. It updates the data and models of the corrosion response of Al cladding and Al-based fuel materials to repair/water vapor/radiation environments. Additional testing is in progress to investigate the effects of J-13 water chemistries on cladding and fuel materials and on the melt-dilute forms in air/water vapor/radiation environments.
Plan for Development of Technologies for Direct Disposal of Al SNF	This document contains the Task Technical and Task Quality Assurance Plans for the activities under the Direct/Co-Disposal Technology Development Program. The plan is revised/superseded at least annually to reflect changes in the program. A new plan is in final approval for the continuation of the program in FY98.
Task Plan for Development of Dilution Technologies for Al Based SNF	This document contains the Task Technical and Task Quality Assurance Plans for the activities under the Melt-Dilute Technology Development Program. The plan is revised at least annually to reflect changes in the program. A revised plan is in final approval for the continuation of the program in FY98.

#### CONT'D

Title	Relationship to SRS ATP
Task Plan for Characterization of DOE AI SNF	This document contains the Task Technical and Task Quality Assurance Plans for the activities under the Characterization Program in FY97. The Characterization Program has been combined with the Test Protocal Development Program in FY98 and the plan for the combined program is in final approval.
Task Plan for Engineering Test Protocol for Metallic Alloy Waste Forms	This document contains the Task Technical and Task Quality Assurance Plans for the activities under the Test Protocol Development Program. The plan is revised at least annually to reflect changes in the program. A revised plan combining the Test Protocol Development Program with the Characterization Program for FY98 is in final approval.
Material Issues in Interim Storage & Direct Disposal of Al Clad SNF	This is a technical report preceding the development of the Acceptance Criteria for Interim Dry Storage (WSRC-TR-95-0347) and the ATP. It identified several of the degradation processes that effect Al SNF in storage and disposal environments. The results and conclusions from this report, providing technical bases for storage and disposal, have been updated in the development of the dry storage acceptance criteria and in the testing and analysis activities under the ATP.
Creep Analysis for Materials Test Reactor (MTR) Fuel Assemblies in Dry Storage	This is a technical report providing part of the technical bases for interim dry storage of Al SNF. It describes the analytical model and data input to predict thermally-induced deformation and slump of the MTR fuel design for temperatures up to 250°C and times up to 50 years. The creep model is being improved and range of time/temperature parameters is being extended to allow prediction fuel deformation under short-term repository disposal conditions (pre-environmental intrusion).

#### ATTACHMENT 3

### **Alternative Al SNF Treatment Technology Program**

## Development of Direct/Co-Disposal Technology for Aluminum-Based Spent Nuclear Fuel

R. L. Sindelar

Program Overview Presentation to Nuclear Regulatory Commission Staff December 18, 1997 Direct/Co-Disposal Technology Development Program Objectives

- Disposition Path for Al SNF:
  - » Interim Dry Storage at SRS for up to 40+ Years
  - » Ultimate Disposal in Geologic Repository
- Develop Technical Bases to Meet Interim Dry Storage Requirements
- Develop Technical Bases to Meet Repository Requirements and for Input to PA

## **Interim Dry Storage of Aluminum-Based SNF**

- Established Requirements (Acceptance Criteria) for Drying and Storage Conditions of Al SNF
- Approach:
  - » Limit Changes in SNF Condition Throughout Handling and Storage Period (Enables Retrievability)
  - » Indentify Environmental Conditions such that SNF Changes Remains Within Acceptable
  - » Develop Predictive Models to Demonstrate Acceptable Conditions

## **Interim Dry Storage of Aluminum-Based SNF**

- Acceptable Changes (Enables Retrievability of SNF):
  - » Consumption of Clad/Fuel < 0.003"
  - » No Gross Rupture of Clad
  - » Plastic Deformation < Clearance of</p>
  - » Release of Radionuclides < Max. Permissible Concentrations
- Environmental Limits:
  - » Fuel Temperature: 200°C
  - » Limit Free Water to Avoid Excessive H<sub>2</sub> Build-Up (Flammability) and Consumption by Corrosion
- Requirements in WSRC-TR-95-0347; WSRC-TR-97-00345

## Direct/Co-Disposal Technology Development for Geologic Repository

#### Attributes:

- Near term Implementation into Road-Ready Package
  - » Interim Storage Technology Developed
- Minimal Conditioning Required



#### Challenges:

- NRC and Stakeholder Acceptance of HEU into Geologic Repository
  - » Proliferation Issue
  - » Criticality Issue
- Characterization Requirements
- Qualification of Aluminum SNF Form

## Direct/Co-Disposal Technology Development Program Activities

## **Interim Dry Storage**

- Thermal Analyses
- Degradation Testing & Anal.
  - Corrosion (vapor
  - & aqueous)
  - Creep
  - Volatile species
  - release
  - Hydrogen Generation
- Demonstrate Reqts. are Met

## **Repository Disposal**

- Thermal Analyses
- Degradation Testing & Anal.
  - Corrosion (vapor
  - & aqueous)
  - Creep
  - Volatile species
  - release
  - Hydrogen Generation
  - Particulate formation
  - Material Reconfiguration
- Criticality Analyses
- Input Results into PA
- Demonstrate Reqts. are Met

## Direct/Co-Disposal Validation of Interim Storage Criteria

- Storage Criteria
  - » Cladding Temperature Limit: 200°C
  - » Free Water Limited
  - » Sealed Canister; Inert Gas Backfill
- Drying Specfication
  - » Preliminary: Dry to < 5 Torr at RT (Unheated Vacuum Drying)
- Validation of Criteria
  - » Instrumented, Shielded Test Canister
  - » Store Live SNF in CY98
  - » Hot Cell Tests



### **Direct/Co-Disposal Thermal Analysis**



## **Direct/Co-Disposal Thermal Analysis**

#### • **Repository Storage**

- Model Using Codes:
   2D FIDAP; CFDS-CFX; Closed-Form
- » Fuel T: Dependent on Thermal Loadings & Model; e.g. 175 to 300°C Initial T
- » Complete Parametric and Best-Estimate Analyses



## Direct/Co-Disposal Degradation & Release Analyses

- Degradation Modes & Mechanisms
  - Creep Modeling
  - Corrosion (Vapor & Aqueous) Testing & Modeling
  - Hydrogen Generation Modeling
  - Pyrophoricity Evaluation
- Volatile Fission Product Release (w/ANL) Testing & Modeling
- Fuel Materials Reconfiguration (Chemical/Physical Forms) in Repository
  - Particulate Formation

•CREEP: displacement of fuel elements can exceed 0.1" at temperatures > 200°C

- Materials Test Reactor fuel design
- FE analysis, self-weight loading



- Testing & Modeling of Al Cladding and Fuel Materials
- Effects of Temperature, Relative Humidity, Water Chemistry, Radiolysis
- Alloy Dependency
- Fuel Material and Microstructure Dependency

## TEMPERATURE DEPENDENT PARABOLIC (PARALINEAR) CORROSION of AL6061



## TEMPERATURE DEPENDENT BREAK-AWAY CORROSION AL5052



### Alloy Dependent Corrosion Mechanisms

#### AL1100 Oxide Film (optical)



#### AL5052 (SEM,300X)



#### AL5052 Internal Oxidation (optical)



#### AL5052 (SEM,4000X)



SRTC 4/30/97

## Aluminum 6061 Exposed to 250°C Water Vapor (6 Months, 100% R. H.)



- Degraded in WP -Failure Mechanisms Evaluated; Geochemistry Analysis using EQ3/6; Criticality Analysis using MCNP4A
- Parametric Analysis (water, geometry, U-235 volume/distribution, absorbers, materials, reflectors)
- Use GdPO4, otherwise solubility issues
- Results: Fully Loaded Can of MIT, Flooded
  - » Intact canister, intact basket, degraded fuel: 1kg Gd in plates
  - » Intact canister, degraded C steel basket: 0.12 kg Gd
  - » Intact canister, degraded SS basket: 0.25 kg Gd
  - » Degraded in WP, fuel on top: 0.20 kg Gd with fuel, no Fe credit
  - » Degraded in WP, fuel on bottom: 0.1 kg Gd with fuel, no Fe credit
  - Degraded in WP, fuel homogenized with clayey material, subcritical w/o credit for Gd and Fe
- Qs: 1) Absolute or, no impact on PA?;
  2) Time period of demonstration of keff?

## **FY98** Activities

- Al SNF Form Performance
  - Materials Degradation Testing and Modeling for Repository Conditions
  - Criticality Analysis of Direct and Melt-Dilute Forms
  - ASTM Test Protocols for Metallic Waste Forms
  - Performance Assessment
- SNF Form Qualification
  - Assemble Initial "Technical Package" for DIS for License
- Interim Storage Criteria
  - Validation Testing & Analysis
  - ASTM Standard Guide for Extended Dry Storage

# MELT-DILUTE PROCESS TECHNOLOGY FOR SPENT NUCLEAR FUEL STORAGE

H.B. PEACOCK

## Alternative SNF Treatment Melt-Dilute Technology

#### Attributes:

- Dilute HEU to LEU Levels
- Waste Form Homogenous
  - Minimal Characterization Requirements
- Volume Reduction up to 70%

#### Challenges:

- Integrated Process Requires Validation
- Wasteform Qualification
  - Isotopic Dilution
  - Ternary Constituents Effect
- Off-Gas System and Waste Streams




Fig. 1. Several fuel element configurations.

# Irradiated MTR Type Fuels







U<sub>3</sub>Si<sub>2</sub>-Al

# Melt-Dilute Fow Diagram



# Melt-Dilution Process Dilution Mechanism

- Dilute to 2 to 20%
- Dilute by Addition of

Depleted U and Al

- Dilute to Target Compositions:
  - Eutectic (13.8 wt% U)
  - UA14 or UA13: Intermetallics



## Melt-Dilute Process Options



#### Melt-Dilute Al-SNF Form Development Process Cycle Option



#### Time

# Process Conditions

	Low Temperature	High Temperature
Temperature	800 - 1000	1000 - 1600
Composition, w	t% 13*, 30	30, 67+
Crucible Option	ns Steel/Graphite	Graphite

\* Eutectic Composition+ UAl<sub>4</sub>, Intermetallic Composition

### Melt-Dilute Al-SNF Form Development Casting Techniques

#### Furnace Cooling



Slow Cool ~5 Hrs Crucible - One Time Use

Tilt and Pour



Compatibility with Automation Compatibility with Off-Gas System

#### Bottom Pour



Commercial Technology

# Crucible - Melt Interactions

Steel Crucibles - Low Temperature Graphite - High Temperature Ceramic - High Temperature



50 wt% 1400 °C

Carbon Steel Crucible Experiments Uranium-Aluminum Alloys 13,2-50 wt%U



Steel - Metal Interactions

# **Casting Compositions**

- Eutectic (13.2 wt% Uranium Alloy)
  - Low Liquidus Tempe5ratuare
  - •No Segregation during Solidification
  - •Potential for Natural Galvanic Couping (two phases)
- •30 wt% Uranium Alloy
  - Experience with Cast U-Al SRS Reactor Fuel
  - •Complex Microstructure
  - •Liquidus Temperature Below 1000 °C

the sand

- 67 wt% Uranium Alloy
  - Homogeneity -- Monolith
  - Corrosion Resistance (expected)
  - High Liquidus Temperature

# Microstructure of Cast U-Al Alloys



# Aluminum-Silicon-Uranium Ternary Phase Diagram



## Effect of Terrnary Constituents

Aluminum Cladding Alloys and Uranium

- 1100, 5052, AND 6061
- Major Impurities: Si, Mg, Fe, Mn
- •Activation Products (Si, Fe, Mn, Zn...)
- •Fission Products (Cs, Sr, Tc, Rb, ...)
- •Partioning of Ternary Elements
- •Phase Stability

# Microstructure of U-Al Alloys



Uranium-30 wt% Aluminum

Iron Partitions to UAl<sub>4</sub>



#### Melt-Dilute Process Process Simplicity

Process Scale Up

- Apparatus for Full Scale MTR
- Resistance Furnace
- Induction Furnace
- Stirrer
  - Mechanical
  - Induction
- Start-Up Tests of M-D Process Using Resistance Furnace



Resistance Furnace Assembly Melt-Dilute Process for Full-Scale MTR

### Melt-Dilution Process Off-Gas System

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#### Melt-Dilution Process Off-Gas System Concept



# Offgas Approach

- Provide cold surfaces for solids deposition
- Filter remaining aerosols
- Dissolve these isotopes in aqueous solution and send to HLW facilities
- Release gases (Dose has been shown to be much less than permissible limits)

# Fission Product Forms Expected

- Cs: Cs metal, Cs oxides, CsI, Cs<sub>2</sub>Te
- Kr: Kr gas
- Tc: Tc metal
- I: CsI, I<sub>2</sub>

### Al-SNF Form Stability Radionuclide Inventory

Radionuclide Inventory	Irradiated MTR	M-D Waste Form	
Cesium	37.3 gms	7.46 gm (20%)	
Krypton	0.63 gms	-0	
Iodine	5.54 gms	0.55 gm (10%)	
Tç.	33.3 gms	33.3 gm	
Rb	19.5 gms	3.9 gms	
Те	0	0	

\* Reference FA after 10 yrs wet storage

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# Fission Product Quantities

•	Yield, M	<u>laximum m</u>	ole fraction	<u>n, max g/g Al</u>
•	Cs+Rb:	0.22	0.0031	0.016
•	Tc:	0.06	0.0008	0.003
•	RE:	0.53	0.007	0.04
٠	Sr:	0.09	0.0013	0.004
•	I:	0.009	0.0001	0.0006
٠	Te + Se:	0.05	0.0007	0.003

# Amount of Fission

- Mole fraction U in fuel as fabricated
  - 0.005 to 0.020
- Burnup and mole fraction fissions
  - Up to 70% (0.0035 to 0.014)
- Fission Product quantities (mole fraction)
  - = FP yield x mole fraction fissions

**Volume Reduction** 



Reference: Direct Disposal 1400 Canisters

V

#### **Proliferation Resistance**

Melt-Dilute Process: Isotopic Dilution to <20% Enrichment



Melt-Dilute => Isotopic Dilution Because of Liquid Phase Processing • Ready Separation of Enriched Uranium not Possible

## Criticality

CASE OF STREET, STREET,

Boron-Doped UAI

- Potential for Criticality Lower
- Poisons If Necessary are Integral to the Waste Form



Aluminue CONTRACTOR CONTRACT

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U-Al-B Ternary Phase Diagram

Interstitial Solid Solution with B

- Boron Will Form Compounds of U or UAlx
- Efficacy of Poison Related to UAlx Degradation Rate

### Al-SNF Form Stability Chemical Stability

#### Preliminary Results of Electrochemical Tests

Corrosion/Dissolution Resistance of Al + UAl4 > Al + UAl3 + UAl4



As Fabricated MTR Assembly

M-D Waste Form





Preferential Corrosion of UAl3 to UAl4



Irradiated Microstructure Al + UAl3 + UAl4



M-D Waste Form Al + UAl4



Preferential Dissolution of Al UAl4 Stringers Intact

# In Process Melt Characterization

- Total Uranium Glow Discharge or Density Measurements
- Waste Form Composition Glow Discharge

Density Measurement:As CastPressed1924.7 (24.3 wt%)

<u>Glow Discharge Emission Spectroscopy Method:</u>

- Measure U, Al, major trace elements
  - Qualitative & Quantitative Technique
  - ~5% accuracy

# In Process Melt Characterization

- U235 or Burn-Up
- Total Uranium
- •Waste Form Composition

Process Strategy:

- Assume No Burn-Up => Consistent Composition but Variable Dilution (<20%)
- Measure Burn-Up or U235: Characterization Costs

#### Melt-Dilute Al-SNF Form Process Development Activities

- Casting Technique Development
- Off-Gas System Development

- Evaluation of Stirring Techniques
- Surrogate Melts and Fission Product Partitioning
- Process Adaptation for Oxide and Silicide SNF
- Evaluation of Crucible Life
- Effects of Ternary Constituents (Cladding Alloys)
- Evaluation of Porosity Effects
- Evaluation of Secondary Waste Streams
- Evaluation of Melt Slag
- Process Control Techniques
- Evaluation of Melt Sampling Techniques
- Evaluation of Poison Additions
- Waste Form Characterization
  - Microstructural Evolution
  - Performance Evaluation