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**ALTERNATIVE ALUMINUM SPENT NUCLEAR FUEL TREATMENT
TECHNOLOGY DEVELOPMENT STATUS REPORT (U)**

Savannah River Technology Center
Strategic Materials Technology Department
Materials Technology Section

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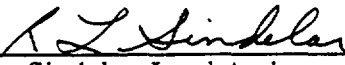
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LIST OF ACRONYMS

<u>Acronym</u>	<u>Definition</u>
Al SNF	Aluminum-clad, aluminum-based Spent Nuclear Fuel
ANL	Argonne National Laboratory
ANSI/ANS	American National Standards Institute/American Nuclear Society
ASME	American Society of Mechanical Engineers
ATR	Advanced Test Reactor
ATTP	Alternative Al SNF Treatment Technology Program
ASTM	American Society for Testing and Materials
CPP	Chemical Processing Plant (INEL)
EIS	Environmental Impact Statement
DOE-EM	U.S. Department of Energy Office of Environmental Management
DOE-RW	U.S. Department of Energy Office of Civilian Radioactive Waste Management (also OCRWM)
DRR	Domestic Research Reactor
F&OR	Functional & Operating Requirements
FRR	Foreign Research Reactor
HEDL	Hanford Engineering Development Laboratory
HEU	Highly-Enriched Uranium (> 20% U-235 in U)
HFIR	High Flux Isotope Reactor
HLW	High Level Waste
ICPES	Inductive Coupled Plasma Emission Spectroscopy
IMNM	Interim Management of Nuclear Materials
INEL	Idaho National Engineering Laboratory
LANL	Los Alamos National Laboratory
LEU	Low-Enriched Uranium (2 to 20% U-235 in U)
LLNL	Lawrence Livermore National Laboratory
MCO	Multi-Canister Overpacks
MPC	Multi-Purpose Canister
MRS	Monitored Retrievable Storage
MTR	Materials Test Reactor
MTRE	Materials Test Reactor Equivalent
NDE	Non-Destructive Examination
NRC	Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
PEIS	Programmatic Environmental Impact Statement
PNNL	Pacific Northwest National Laboratory
RBOF	Receiving Basin for Offsite Fuel
RH	Relative Humidity (vapor pressure of H ₂ O/saturation pressure of H ₂ O)
SCC	Stress Corrosion Cracking
SEM	Scanning Electron Microscope
SI	Structural Integrity
SNF	Spent Nuclear Fuel
SRS	Savannah River Site
TMI	Three Mile Island
SRTC	Savannah River Technology Center
VLEU	Very-Low-Enriched Uranium (< 2% U-235 in U)

CORRELATION OF REPORT SECTIONS WITH TECHNICAL TASK PLANS

The format of this status report is a narrative description of the overall technologies needed to enable storage of aluminum SNF in the federal repository. The sections of the report provide the status of the activities described in the individual task plans. The following shows the relation between the sections of the report and the subtasks of the task plans.

<u>REPORT SECTION</u>	<u>TECHNICAL TASK PLAN</u>
3.0 DIRECT DISPOSAL TECHNOLOGY DEV.	SRT-MTS - 96-2047, Rev. 1
3.1 Requirements, Criteria, and Specifications for Road-Ready Storage	Sub-task 1.1, 1.2 (A2)
3.2 Validation & Verification of Road-Ready Storage	Sub-task 1.3 (A3)
3.3 Waste form Degradation and Release Studies	Sub-task 1.4 (A4), 2.1 (A4, A5)
3.4 Criticality Analysis	Sub-task 2.2 (A6)
4.0 MELT DILUTE PROGRAM	SRT-MTS -96-2063, Rev. 1
4.1 Melt Dilute Technology	Task 1.0 (6)
4.2 Process Development Status	Task 2.0, 3.0 (6,7,8)
4.3 MTR Fuel Assembly Development	Sub-task 5.1 (9)
4.4 Gas Release Analysis and Offgas System Development	Task 4.0 (8,9)
4.5 Fission Product Recovery and Off-Gas System Design	Sub-task 4.5 (9)
4.6 Waste Form Evaluation Status	Task 6.0 (10)
4.7 Furnace Development	Task 2.0 (7)
5.0 SNF CHARACTERIZATION PROGRAM	SRT-MTS - 97-2004, Rev. 0
5.1 Strategy for SNF Characterization	Background (2,3)
5.2 Examination of SNF	Task 1.0 (A2)
5.3 Status of Testing and Test Development	Task 2.0 (A4)
6.0 TEST PROTOCOL	SRT-MTS-96-2064, Rev. 0
6.1 Approach to Test Protocol Development	Background (4)
6.2 Status of Test Protocol Development	Task 4.1 (5,6)

PREFACE

An Alternative Aluminum Spent Nuclear Fuel Treatment Technology Program (ATTP) was initiated at the Savannah River Site (SRS). The technology needed for interim storage and ultimate disposition of aluminum-based research reactor spent nuclear fuel (Al SNF) under the jurisdiction of the U.S. Department of Energy (DOE) will be developed under this program. This report provides the status of the technology development activities.

Aluminum SNF is being consolidated at SRS in Aiken, South Carolina for treatment, packaging, interim storage, and preparation for ultimate disposal in a geologic repository in the United States. Sources of Al SNF are domestic research reactors (DRR), foreign research reactors (FRR), and SRS production reactors. A significant portion of the Al SNF to be consolidated at SRS will contain highly enriched uranium. Hence, although small in quantity compared to the inventory of commercial spent fuel and the high level waste (HLW) to be stored in the repository, the disposition technologies must consider and mitigate the occurrence of a criticality event in the repository. An FY97 program was initiated to evaluate technology alternatives to processing for the ultimate disposition of research reactor Al SNF in a geologic repository. This program is referred to as the Alternative Al SNF Treatment Technology program (ATTP).

These actions followed the DOE Record of Decisions from the Programmatic Spent Nuclear Fuel Environmental Impact Statement (SNF-EIS) [P.1], the Interim Management of Nuclear Materials Environmental Impact Statement (IMNM-EIS) [P.2], and the Foreign Research Reactor Environmental Impact Statement (FRR-EIS) [P.3]. Interim management and preparation for final disposition of DOE-owned Al SNF is programmatically managed by the Office of Environmental Management (EM) of DOE.

The Spent Fuel Storage Division (SFSD) at SRS is responsible for receipt, treatment, packaging, and interim storage of Al SNF and preparation for ultimate disposal in a geologic repository. Criteria for acceptance of off-site spent nuclear fuel for direct basin storage [P.4] and for future drying and interim dry storage of Al SNF for up to 50 years [P.5] were developed previously. The SFSD is responsible for executing the Alternate Al SNF Treatment Technology development program. In addition, SFSD is also expected to initiate and implement a Request for Proposal (RFP) for a Transfer and Storage Facility (TSF) services contract. This facility will provide services to place Al SNF in a road-ready condition, including interim dry storage.

The ATTP is being led by the Savannah River Technology Center (SRTC), coordinating the efforts of a team of scientists and engineers from SRTC, SFSD, and the Projects Engineering and Construction Division (PECD). The team also consists of Argonne National Laboratory (ANL), Pacific Northwest National Laboratory (PNNL), and the DOE-RW Management and Operating (M&O) contractor. This program addresses all aspects of storage and disposition of the Al SNF. It incorporates the recommendations of the Research Reactor Spent Nuclear Fuel Task Team appointed by the Office of Spent Fuel Management of DOE EM-67 [P.6]. The four main program elements of the ATTP are:

- ***Development of Technologies for Direct and Co-Disposal of Aluminum Spent Nuclear Fuel*** - In Direct and Co-disposal, the SNF would be placed into small waste packages, with or without high level waste glass logs, ready for disposal in a repository. The SNF quantities in a package will be limited to satisfy repository

criticality requirements. This program element consists of tasks necessary to qualify the Direct and Co-disposal of Al SNF in a road ready package for the repository.

- ***Development of Dilution Technologies for Aluminum Spent Nuclear Fuel*** - The Melt-Dilute option for treatment of Al SNF consists of melting and diluting the Al SNF with depleted uranium. This program element consists of tasks necessary to develop and qualify the Melt-Dilute process and the diluted waste form.
- ***Characterization of DOE Aluminum Spent Nuclear Fuel*** - This program element consists of identifying the characterization requirements, developing and building the characterization database and the associated characterization technologies needed for both Direct/Co-disposal and Melt-Dilute waste forms.
- ***Development of Test Protocol for Metallic Aluminum Spent Nuclear Fuel Waste forms*** - This program will develop standardized test methods for the evaluation of performance of an aluminum SNF waste form in a repository. It is expected to serve as the test method used to qualify aluminum SNF for repository disposition.

The data and analyses which result from the ATTP will provide the technical bases for qualification of the Al SNF for disposal in the geologic repository. This program will be integrated with the Office of Civilian Radioactive Waste Management (OCRWM) of DOE. OCRWM is ultimately responsible for the development of the license application for the geologic repository and subsequent submittal to the Nuclear Regulatory Commission (NRC).

1.0 SUMMARY

1.1 Direct Disposal Technology Status

The development of direct disposal technology for Al SNF is partitioned into two technology areas per Technical Task Plan SRT-MTS-96-2047 [1.1]. The first area deals with the technology necessary for successful interim storage of the aluminum-based SNF at the Savannah River Site. Acceptance criteria for the drying and storage conditions for the interim storage have been developed to provide for full retrievability of the SNF as a waste form during this storage period. Current plans call for these criteria to be integrated into an ASTM guide for extended dry storage of spent fuel. The guide will aid the designer, the owner/operator, and the regulator in providing for interim dry storage for up to 100 years.

Engineering analyses to support implementation of interim dry storage are underway. Heat transfer analysis codes have been validated for the anticipated low-temperature-differential, convective-flow-driven conditions. Preliminary specifications to achieve the drying criteria for these fuels have been developed. The verification of the drying and storage criteria, as well as the life prediction estimate for the Al SNF in interim storage, will be performed through data collected from storage of spent fuel in special instrumented test canisters. The design and fabrication of two test canisters has been initiated. Two Al SNF assemblies will be loaded into canisters in calendar 1998, prior to the design and construction of a full-scale storage facility at SRS.

The second technology area consists of developing the scientific database necessary to demonstrate that these fuels can be placed in the repository with the HLW waste form (borosilicate glass logs in stainless steel canisters) in an acceptable storage configuration. Corrosion degradation testing and analysis are in progress, and models have been developed for predicting the long-term response of the cladding and fuel materials under direct exposure to the repository. Criticality analysis is being conducted in a joint effort with the DOE-RW M&O. Two design basis fuel types have been selected to bound the criticality conditions for the various types of Al SNF and criticality analysis has been initiated to show that a breached waste package with intact SNF and intact internal structure will meet regulatory requirements for disposal.

1.2 Melt Dilute Technology Status

The dilution of the U^{235} in the high enriched research reactor Al SNF is being evaluated as the back-up option to direct/co-disposal disposal of Al SNF. The primary means of achieving the dilution is by the addition of depleted uranium or uranium oxide during a melting process. This approach is termed as the Melt-Dilution technique. The approach and tasks for the development of the Melt-Dilution technology for Al SNF is detailed in the task plan [1.2]. The focus of these tasks are (1) process development including bench scale testing; off-gas system development and full-scale testing; and (2) waste form development including alloy development and waste form qualification.

A literature review of various dilution technologies was completed and dilution options through a melting process were identified. A process flow diagram was developed. The amount of depleted uranium necessary to achieve various degrees of dilution was calculated and reconciled with the Al-U phase diagram. Several desired microstructures namely eutectic and intermetallic microstructures were identified. Bench scale apparatus

was developed including a resistance furnace and an induction furnaces for the melting of Al-U alloys. Samples consisting of a range of compositions of Al-U alloys were fabricated to achieve the desired microstructures. Preliminary investigation of mold compatibility was also conducted. Corrosion evaluation of these samples in J-13 water environment are currently underway.

The fission product release data as a function of temperature in the melting temperature range (850°C to 1200°C) was also assimilated. ORIGEN code analyses data on research reactor SNF showed that the fission off-gas will primarily comprise of Cs¹³⁷, Kr⁸⁵ and I¹²⁹. Off gas system concepts are currently being developed. The furnace design for a full scale melting demonstration was also completed. This demonstration will consist of melting a full scale surrogate FRR fuel, that is, a surrogate Materials Tests Reactor (MTR) fuel assembly. A resistance furnace has been procured and is currently being installed.

1.3 Characterization Technology Status

The SNF Characterization Program is in the early development stage, with task activities outlined in Task Technical Plan SRT-MTS-97-2004 [1.3]. The SNF characterization program is divided into two primary tasks. The first task involves the review of all pertinent regulatory documents in order to determine the characterization requirements for the Al SNF disposition process. All relevant material property data will then be assimilated into a characterization database for aluminum SNF materials expected to be received, stored, and packaged at SRS into a "road-ready" form. The database will also be used in support of alternate disposition technology development and waste form qualification. As part of this task, deficiencies in the database for specific fuels will also be identified and determined through coordination of experimental activities to be performed at the Savannah River Site, Argonne National Laboratory (ANL) and other institutions as needed.

The review of regulatory documents and drivers is on-going and due to be completed at the end of April 1997, with the issuance of a summary report. A preliminary, high-level format for the characterization database has been outlined, with details to be developed as the program continues and specific requirements are identified. The scope of the database will be modified as needed to meet requirements and waste acceptance criteria (WAC) to be provided by the Office of Civilian Radioactive Waste Management (OCRWM).

In the second task, existing technologies used for the characterization of aluminum SNF are to be reviewed and critiqued, with emphasis placed on those capable of meeting the minimum characterization requirements in the most efficient manner. Existing technologies deemed sufficient to meet applicable requirements shall be identified and included in the preliminary characterization requirements document to be issued as a key deliverable (per the task technical plan). Characterization technology needs shall also be identified under this task, and development activities will be initiated as needed.

1.4 Test Protocol Technology Status

The objective of the tasks presented in this plan is to develop, validate and implement the test methodologies and associated technologies, necessary to assess the suitability of waste forms for repository disposal. The detailed tasks are identified in Technical Task

Plan SRT-MTS-96-2064 [1.4]. Test methodology development is divided into three basic steps: 1) review and analysis of established and emerging test methods that assess commercial nuclear fuels and high level waste glasses; 2) identification of the technology needs that may be specific to waste forms from aluminum based SNF; and 3) modification and/or expansion of the established methods to provide a test protocol that will assess the relative quality of waste forms associated with aluminum-based SNF.

The literature review and identification of the specific physical and chemical property measurements necessary to assess suitability are continuing. The review of available literature suggests that tests required to assess waste form suitability must characterize: 1) the release rate of radionuclides from the waste form, 2) the role of environmental variables and waste form integrity on the release rate, and 3) the extent of transport and relocation of the radionuclides. The anticipated behavior of aluminum-based waste forms is substantially different from the behavior of commercial nuclear fuels and defense waste glasses. Thus, test methods that measure radionuclide release from the aluminum-based materials may differ significantly from test methodologies currently used to assess the more inert waste forms. The mechanisms of radionuclide transport and relocation on the other hand should be consistent with those anticipated for other waste forms.

The physical and chemical property measurements necessary to assess suitability will include evaluation of the oxidation and corrosion resistance of candidate waste forms by chemical and electrochemical measurements in J-13 environment and determination of the mechanical properties of the waste forms by measuring the strength and ductility. Specific measurements will include determinations of 1) the tendency for uniform corrosion and oxidation; 2) the pitting potential; 3) the susceptibility of the waste form to selective leaching of specific elements and/or phases; 4) the tendency for galvanic corrosion; and 5) the impact of radiation fields, temperature and test environment on the oxidation and corrosion processes. Testing has not been initiated in this program.

2.0 INTRODUCTION

2.1 Aluminum-Based Spent Nuclear Fuel

Aluminum-based SNF will account for less than 1% of the total volume of SNF and high level waste that will ultimately require disposal in a geologic repository. However, much of the fuel contains HEU with up to 93% enrichment. The MTR fuel element with Al-clad aluminum-uranium alloy fuel is the dominant design and fuel material for research reactors. In addition, some fuel elements were fabricated from aluminum-uranium silicide alloy or aluminum-uranium oxide. The fuel elements are clad with one of the aluminum alloys 1100, 5052, or 6061 or their foreign equivalents.

Currently this SNF is typically stored under water where corrosion may be severe unless strict control of the water purity is maintained [2.1]. Prolonged underwater storage is not desirable because of the high cost of operating and maintaining a properly controlled water system and the limited space and handling capabilities available at most reactor facilities. Consequently, alternatives to interim storage and ultimate disposition are being evaluated.

2.2 Program Evolution and Technical Strategy:

The United States Department of Energy has selected the Savannah River Site (SRS) as the location to consolidate and store U.S. origin aluminum-based spent nuclear fuel (Al SNF) from foreign and domestic research reactors (FRR and DRR, respectively) [2.2, 2.3, 2.4] through the Environmental Impact Statement (EIS) process. These SNF are currently being irradiated in the research reactors, or are being stored in water basins or dry storage at their sites, or have been transferred to SRS and stored in water basins [2.5]. A portion of this inventory contains highly-enriched uranium (HEU). Since the fuel receipts would continue for several decades beyond projected canyon operations, it is anticipated that alternative disposition technologies to processing will be necessary.

The Research Reactor Spent Nuclear Fuel Task Team was appointed by the Office of Spent Fuel Management of DOE to evaluate the effectiveness, relative merits, costs, and difficulties in implementation of alternative technologies and waste forms for the treatment, packaging, and disposal of aluminum-based SNF [2.6]. The base case for comparison of the several technologies was chemical processing followed by incorporation into borosilicate glass. The principal recommendations of the Task Team were:

- parallel development of direct disposal and dilution technology options including co-disposal with HLW borosilicate glass logs. Co-disposal appears possible in both cases;
- utilization of the SRS canyons for processing of SRS fuel, failed or sectioned fuel, and other selected fuel;
- electrometallurgical treatment as an advanced technology backup to direct and dilution technologies.
- DOE-NRC agreement on requirements for SNF disposal and waste form for HEU; and
- plan, fund, and design a Transfer and Dry Storage Facility at SRS.

None of the alternative waste forms other than borosilicate glass resulting from processing is qualified for disposal in the federal repository in Yucca mountain.

Development and licensing of a geologic repository for ultimate disposal of SNF and high level waste (HLW) is the responsibility of the Office of Civilian Radioactive Waste Management of DOE. At present time, a licensing application for the disposition of SNF and high level waste glass in the repository is expected to be completed by 2002. The licensing application drives the schedule for the repository Performance Assessment and the Viability Assessment to be completed during the 1998 - 2002 period. In order to ensure that Al SNF disposition is part of the DOE-RW licensing application, a technology development program aimed at evaluating non-processing alternatives was initiated in FY97. The purpose of this program was to complete the engineering analyses and develop the science necessary to ensure qualification of an appropriate aluminum SNF waste form for the repository.

3.0 DIRECT DISPOSAL TECHNOLOGY DEVELOPMENT PROGRAM

The DOE-RW is in the process of qualifying the HLW, commercial SNF, and DOE SNF for repository storage. This process involves three basic components:

- 1) Total System Performance Assessment;
- 2) Criticality Assessment; and
- 3) Technical Package for Licensing of the Repository by the NRC

Prior to repository storage, the AI SNF may be stored at SRS road-ready in a licensed dry storage facility.

The experiments and engineering analyses in the direct disposal program are needed to demonstrate the waste form acceptability and support the qualification of the AI SNF for interim and for repository storage. The technology program for direct disposal [3.1] includes the experimental and analytical activities in the following areas:

- Requirements, criteria, and specifications for interim dry storage which is "road-ready" for shipment to the repository;
- Validation of the drying & storage criteria and verification of life prediction in interim dry storage systems;
- Waste form degradation and release studies for interim dry and repository storage; and
- Material reconfigurations and criticality analysis for repository storage

Sections 3.1, 3.2, 3.3, and 3.4, respectively, provide the status of the development activities in these areas.

The configuration of the SNF canisters within the repository waste package, the large canister for co-disposal of the SNF canisters with the defense high level waste canisters (DHLW), has not been determined. The configuration will be based on the maximum fuel loading per waste package as established through materials degradation behavior and criticality studies (sections 3.3 and 3.4).

3.1 Requirements, Criteria, and Specifications for Road-Ready Storage

Requirements for the drying and storage of AI SNF in an interim dry storage system at SRS have been identified. The requirements meet current federal regulations for interim dry and repository storage and are consistent with the SRS-developed requirements for AI SNF. Criteria for drying and storage, derived from engineering analyses, have also been developed to ensure these requirements are met. Preliminary drying specifications have been developed.

Meeting the existing requirements for commercial spent nuclear fuel does not necessarily ensure qualification of the AI SNF waste form for the federal repository. At this point in time, the direct placement of dried AI SNF assemblies in a sealed canister of Type 316L

stainless steel provides a best judgment estimate for an acceptable waste form that could be qualified for repository storage.

3.1.1 Requirements for Interim Dry Storage and Repository Disposal of Spent Nuclear Fuel

The DOE-EM intends to have the interim dry storage facility at SRS licensed by the NRC, who is also the regulatory authority to license the federal repository. Federal regulations have been established for interim storage [3.2], transportation [3.3], and repository storage [3.4] of spent nuclear fuel. In addition, a guide focusing on materials issues for re-licensing dry storage facilities for up to 100 years is being prepared through the ASTM [3.5]. This guide will aid the vendor, regulator, and operator in addressing SNF performance in dry storage for up to 100 years and will cover DOE fuels in addition to commercial SNF. The site report [3.6] providing criteria for interim dry storage of AI SNF will be an integral part of this guide.

A site report on Waste Form Criteria was recently prepared [3.7] to identify current design requirements for repository storage.

Requirements and drying and storage criteria are listed in sections 3.1.2, 3.1.3, and 3.1.4 below. These requirements and criteria are updates to the results contained in reference 3.6 and are recommended to be applied to AI SNF to be stored dry up to 100 years.

3.1.2 Acceptable Fuels and Initial Condition

The AI SNF fuels considered in this analyses include:

- Aluminum-clad, with aluminum-based alloy fuel
- Whole fuel assemblies including those with through-clad pitting corrosion
- Sectioned fuel or fuel pieces, if within an immediate, sealed canister to facilitate handling and retrievability

Aluminum-clad fuels being consolidated at SRS include those with uranium metal as the fuel material. Uranium metal fuel however is not considered for placement in a road-ready storage at SRS. Federal regulations for repository storage state that there must be no pyrophoric events in the repository to affect its performance [3.4]. With uranium metal fuel, the production of UH_3 , a pyrophoric substance, may be feasible under certain repository conditions. Issues are currently being addressed at the Hanford and Idaho sites regarding drying and stabilizing the uranium metal fuel against pyrophoric events for interim storage.

Some of the FRR contain through-clad pitting as a result of basin storage under aggressive chemistry conditions (see section 5.0). Fuels in this condition are acceptable for storage because the cladding is not considered to be a confinement barrier in a sealed canister storage system. Fuels that have been sectioned are also acceptable for storage but must be encapsulated prior to containment in a canister to facilitate handling.

3.1.3 Acceptable Changes in Fuel Condition in Road-Ready Storage

Degradation of the waste form during dry storage is acceptable and is limited to enable retrieveability throughout interim storage. Acceptable degradation during dry storage is defined by the following set of limits to the changes in fuel condition as received from basin storage for conditioning and storage for the duration of the interim dry storage period:

- General corrosion or pitting corrosion of the cladding up to 0.003 inches;
- General corrosion or pitting corrosion of the fuel meat (core) up to 0.003 inches;
- No large rupture of the cladding;
- No plastic deformation at or exceeding the clearance of the fuel assemblies and storage grid in a sealed canister; and
- Release of radionuclides into the canister up to the NRC Maximum Permissible Concentration for air

This set of limits to degradation of the fuel during drying and storage would enable retrieveability (if necessary, to recover the fuel and repackage or treat prior to ultimate disposition), criticality safety, and a level of radionuclide confinement by the fuel. Release of volatile radionuclides into the plenary space of the sealed canister is acceptable. Release during storage to concentrations greater than the Maximum Permissible Concentration as defined by NRC for nuclear facilities is acceptable only if controls are maintained in engineered systems built to store these fuels.

3.1.4 Drying and Storage Criteria for Road-Ready Storage

The fuels in road-ready storage are to be placed in a sealed canister to provide for radionuclide confinement and to limit the amount of species that is available to corrode the cladding and fuel material. The following limits for drying and storage of Al SNF are based upon materials degradation testing and analyses (see section 3.3):

- Cladding Temperature Limit for Drying: 200°C
- Cladding Temperature Limit for Storage: 200°C
- Free Water within Canister Shall Not Cause:
 - 1) Corrosion Clad and Fuel Exceeding the Corrosion Limit; and
 - 2) Hydrogen Build-up Exceeding 4% by Volume of the Canister

Materials testing and analyses have shown that aluminum cladding alloys and aluminum-based fuel is susceptible to hydrogen blistering if corrosion occurs at temperatures above 200°C (see section 3.3). Environments that allow continued corrosion and blistering of the cladding and fuel are not acceptable and therefore drying processes to remove free water are limited to temperatures below 200°C. Analysis has shown that measurable deformation (0.1" deflection of fuel plates) could occur due to creep at temperatures above 200°C for an exposure time of 50 years [3.8]. For this reason, storage

temperatures should be maintained below 200°C. The creep analysis is being extended to predict the response of the assemblies to times greater than 50 years.

The free water within the canister must be limited. Although 10CRF60 does not allow water in the waste package, the above levels of water from free water, adsorbed water, and waters of hydration are or will be bound as hydrated oxides of the aluminum. This water source hence is negligible in consideration of radionuclide transport or in criticality analyses.

The basis for the free water limit is to avoid excessive corrosion and hydrogen gas build-up. The build-up of hydrogen above 4% by volume is acceptable only if controls are maintained in engineered systems built to retrieve these fuels to avoid deflagration concerns. The design of the canister must also be sufficient to withstand the pressure due to hydrogen, water vapor, and back-fill gas at the design storage temperatures. Sections 3.1.6 and 3.3 provide additional recommendations for drying and equations to calculate the hydrogen generation and partial pressure during sealed storage of Al SNF.

3.1.5 SNF Canister Material

The Al SNF waste form is defined as the fuel assemblies together with the canister enclosing the assemblies (SNF canister) and associated internal support structures. The SNF canister material must be compatible with the HLW canister in co-disposed systems. The degradation of the Al SNF and materials interactions within the co-disposal package must consider the canister material and cladding as well as the fuel material.

A preliminary recommendation is for Type 316L stainless steel as the SNF canister material. Type 316L stainless steel is compatible with Type 304 stainless steel, the HLW canister material in aqueous environments, and has superior pitting corrosion resistance. The cost for Type 316L stainless steel is similar to Type 304 stainless steel. Analyses for material interactions of the SNF canister, Al SNF, and HLW canister, etc. need to be performed to allow a final recommendation for the SNF canister and support materials. The internal structure and the potential need for poisons is not identified at this time and will be guided by the materials configuration studies and criticality analyses.

3.1.6 Drying Specifications for a Conditioning System

Vacuum drying at slightly elevated temperatures (~120°F) appears to be an acceptable method for preparation of Al SNF for extended dry storage. Achieving a vacuum below the triple point of water (~4.5 torr) within a heated canister will vaporize all free liquid water. High temperature drying to remove water of hydration from the aluminum oxide on the cladding is not practical, as the temperature required to dehydrate the oxide is high (300°C). Furthermore, the quantity of bound water is insufficient to cause excessive additional corrosion. Negligible amounts of UH₃ are expected to be present on Al SNF as a consequence of water storage; therefore, further conditioning or conversion would not be required in order to eliminate this potential ignition hazard.

The proposed conditions for vacuum drying are based in part on evaluation of SNF drying and stabilization methods under development at the Hanford and Idaho National Engineering Laboratory (INEL) sites. Previous experience at SRS with vacuum drying methods are also being reviewed for applicability to drying Al SNF. The data are summarized in Table 3.1.

Aluminum-Based SNF Drying Concerns

Three sources of water can potentially contribute to corrosion of aluminum-clad SNF:

- 1) Free water on the SNF surface and in pits, crevices, etc.
- 2) Water of hydration within the aluminum oxide surface coating
- 3) Adsorbed water on the surface of aluminum oxide and aluminum metal

The major potential contributor to corrosion is free water on the SNF surface. The existing acceptance criteria of 1 milliliter of water per 0.1 square meter of surface area equates to ~18 milliliters (0.04 pounds) per cropped MTR assembly. This moisture level could produce ~0.0001 inches of uniform aluminum corrosion.

Explosive and Pyrophoric Hazards

Buildup of hydrogen inside a sealed SNF storage container is another potential concern. Water and aluminum react to form Boehmite and hydrogen gas. The explosive range of hydrogen in air is 4% to 75% by volume at room temperature. The ratio of free water to canister volume must remain below 39 to ensure hydrogen does not reach the 4% threshold. This ratio equates to ~17.4 milliliters of water inside a 17-inch diameter by 10-foot long storage canister. Essentially all free water must be removed to satisfy the hydrogen limitation and 10 CFR 60.135 requirements for eventual geologic disposal.

Uranium hydrides are not expected to form during corrosion of aluminum-uranium SNF because the uranium is chemically bound in particles of UAl_x , which are dispersed in a matrix of aluminum which prevents uranium and hydrogen from reacting readily. The aluminum oxide film on the SNF cladding is not a pyrophoric hazard because it forms slowly, is relatively thin, and generates minimal heat during formation.

Aluminum-Based SNF Drying

The initial volume of free water inside a SRS SNF canister cannot be accurately estimated without testing. A 17-inch-diameter by 10 to 15-foot-long SRS co-disposal canister has been proposed. Hanford and INEL SNF canisters of comparable size are expected to contain about one gallon of water that must be removed by vacuum drying.

A test program is highly desirable to quantify drying system performance for SRS SNF. Initial tests should focus on drying dummy test assemblies with various free water contents. The dummy assemblies can be placed into instrumented sealed canisters after drying and monitored to check drying effectiveness.

Table 3.1 - Comparison of Drying/Stabilization/Conditioning Treatments for DOE SNF

	Hanford K Reactor Basins	INEL TMI-2	INEL CPP Dry Storage Project	INEL CPP-603 AL-Clad & Sodium-Bonded SNF	SRS
Fuel	100,000 Zirc clad U metal assys 1.25% ²³⁵ U max 2.42" OD, 26.1" long 51.6 lbs per assy	Core rubble 2.95% ²³⁵ U	>4000 TRIGA assemblies, 744 FSV assemblies Shippingport Peach Bottom, and misc SS & Zirc clad SNF	Seven fuel types incl. UAl, UZrH, UO ₂ , U ₃ O ₈ , U metal assys and 20 Kg U flux filter 12" to 50" long plates and 28"-37" long rods 5.3%-93.5% ²³⁵ U	~34,000 Al-clad uranium- aluminum alloy assys
Canister Materials & Dimensions	304L SS MCO with removable baskets 24" OD, 23.25" ID 25.25" OD shield plug 166" long, ~148" cavity	Existing 304L/316L stainless steel cans are 14" OD, 150" long 3 can designs: Fuel Filter Knockout Cans will be stored inside Vectra DPC'S.	304/316SS? Vectra FO-DPC is 67.25" OD, 66" ID, 186.2" long with 167" cavity. Peach Bottom SNF cans are 12.75" OD, 166" long, 161.5" cavity	SS canisters with removable buckets. Canisters are 11 ft. long 18" OD Sch 10 pipe with 17.5" ID	SS canister <17.7" OD <179.9" long
Closure	Threaded shield plug with O-ring and 4 ports Welded cap over plug?	Bolted (1) and welded can lids with 1/4" & 3/8" couplings for venting	Welded	Vented lid secured with lever arm	TBD
Weight	19,142 lbs dry (~288 assemblies max)	2800 lbs max dewatered per can			TBD
Quantity	400 MCO's	343 cans in ~29 DPC's	~78 DPC's	18 to 21 canisters	~1400 canisters
Heat Load	400 watts nominal 800 watts max per MCO	20 watts avg. 80 watts max per TMI can		<300 watts per canister (12 watts per ATR assy) p 21 fsc sar	14.2 kW/canister
Design Rating	ASME Sec III, Sub NB 150 psig, 200 C	ASME Sec VIII, Div I 140 psig, 400 F	DPC not established Peach Bottom cans rated @ 15 psig, 380 C		TBD

Table 3.1 continued

	Hanford K Reactor Basins	INEL TMI-2	INEL CPP Dry Storage Project	INEL CPP-603 AL-Clad & Sodium-Bonded SNF	SRS
Free Water Inside Canister Before Drying	2-4 liters (0.5-1 gal) per MCO expected 16 liters (4.2 gal) max capacity	20-250 lbs (2.4- 30 gal) per "filter" can		3 liters max (0.8 gal) per canister p 104 fsc sar	
Radiation Rate		1500 R/hr @ 1 cm from top of can	300 mR/hr @ top of DPC		850 mR @ 1 ft without shielding
Drying Process	Desludge SNF inside "washing machine" followed by high pressure rinse. Load MCO underwater. Transport MCO inside cask to vac dry station. Purge 150-180 gallons bulk water from MCO via gas injection and eductor. Dry MCO via 4- stage oil free Balzers vac pump. Condenser on pump inlet with 35°F chilled water controls vacuum. Circulate 50°C water in annulus around canister exterior during drying. Declare SNF dry after achieving 0.5 torr, isolating and holding for 1 hr with <3 torr rise. Provisions for 350°C hot conditioning system.	Dewater flooded cans via gas injection. Currently performing cold vacuum drying tests. Considering 300°C hot drying process to fully dehydrate rubble and concrete.	Load DPC & install shield plug inside dry cell. Move DPC out of cell. Install automatic welder and weld plug to shell. Dry DPC via vent & siphon ports with oil- cooled vacuum pump. Perform 22 psia He leak test. 2.3 psig final canister press. Weld caps onto ports. Install and weld outer lid.	Load fuel buckets underwater. Place loaded buckets into canister inside Fuel Handling Cave hot cell. Install temporary vacuum drying lid. Vac dry with Leybold pump throttled to 5 torr. Heat canister internals to 50 - 100°C via nine 1.5 kW external heaters. Declare SNF dry after pressure drops below 3- 5 torr and holds for 1-2 hrs when isolated. Computerized canister temperature and pressure data acquisition & control system. Bake @ 150°C for 2 hrs with 1 scfm purge of 2% O ₂ in Argon to convert hydrides to oxides, i.e. passivate.	120°F heated vacuum drying to below 4.5 Torr

Table 1 continued

	Hanford K Reactor Basins	INEL TMI-2	INEL CPP Dry Storage Project	INEL CPP-603 AL-Clad & Sodium-Bonded SNF	SRS
Drying Time	Est 24 hours per MCO. Prototype tests indicate 4-6 hours per MCO	12-24 hrs hot gas purge?		2-5 hrs drying/heatup ~10 hrs cooling time	
Storage Mode	Canister Storage Bldg similar to Ft St Vrain.	29 DPC's holding 12 TMI cans each. Vented DPC's stored in NUHOMS modules with passive cooling.	78 DPC's stored in NUHOMS modules with passive cooling.	Irradiated Fuel Storage Facility in CPP-603. Exhaust fan online, but not required for cooling 80-90°C inside canister.	Modules or Monolith
Concerns	Self ignition of hydrides on severely corroded assemblies during handling. Hydrides swell cladding allowing evolved H ₂ , O ₂ to escape and pressurize MCO.	Radiolysis of residual moisture may lead to H ₂ , O ₂ generation and pressurization of cans while sealed during transport. Criticality analyses very conservative because there is no record of fissile content per can.	Corrosion & radiolysis lead to DPC pressurization	Uranium hydride is pyrophoric and could ignite U metal SNF. Water of hydration released during storage decomposes into H ₂ , O ₂ leading to hydride formation	Uranium hydriding is not an issue with aluminum-based fuels
Status	Start loading MCO's between 12/97 and 5/98	Contract awarded to Vectra in Summer 1996	Privatize per 1/97 decision	Field installation tested with mockup fuel in Summer 1996. Passivation procedures require revision. Start SNF drying by May-June 1997	Request for Proposal under development
Regulations	DOE Orders	10 CFR 72	10 CFR 72	DOE Orders	10 CFR 72

3.2 Validation of Drying and Storage Criteria in Road-Ready Storage and Verification of Acceptable Storage Using Instrumented Test Canisters

Two identical test canisters are being developed to monitor the response of spent MTR type fuel stored under dry conditions. The objectives of this test program are to provide:

- Verification of the drying and storage criteria
- Verification of dry storage life prediction models (corrosion, creep, specie release)
- Lead assembly monitoring - surveillance program.

The canister design will enable monitoring of temperature, relative humidity, type and quantity of volatile species, and fuel condition. One of the canisters will be loaded with "ideal" fuel, that is, undamaged and nearly free of moisture. The other canister will be a worst case within the acceptable range of fuel elements. The fuel may have some pitting and will contain some moisture.

A conceptual specification on instrumented test canisters [3.9]. The canister design and issues such as loading, handling, and storage of these canisters are currently being addressed. The primary approach is to dry and load the fuel elements in the 105-L disassembly basin, then transport the loaded canisters to the dry cave. This area will provide additional radiological control and will keep the canister isolated from routine basin operations.

In order to estimate radiation dose rate from the SNF and the shielding requirement, preliminary analysis was performed using a High Flux Beam Reactor Fuel Assembly as a reference MTR assembly. The important input parameters for the reference assembly are:

Time in reactor: 52 Days
Pre-Irradiation Total Weight of U: 377 gm
Pre-Irradiation Weight of U-235: 351 gm
Power Level: 2.14 MW/assembly
Cooling Time: 11.9 years
Overall Weight: 4.4 Kg

Preliminary analysis showed that it requires about 8 inches of steel shielding to lower the dose rate to below 200 mrem/hr at the surface of a canister. Without shielding, the dose rate at one foot away from fuel was 850 rem/hr. This analysis will be reviewed and final fuel selection will be made based on reasonably manageable radiation dose for fuel handling and monitoring.

Radiation-hardened camera and viewing window have been identified. Detail specification review is in progress and procurement of these items will be initiated upon approval of the specification..

A preliminary block diagram for the instrumented canister is shown in Figure 3.1. The sensors for quantifying hydrogen, oxygen, water, NOx, Cs, pressure, and temperature have been selected and procurement initiated. Redundant real-time monitoring and recording electronics will be specified and purchased following sensor acquisition. These associated sensors are being incorporated into the canister design.

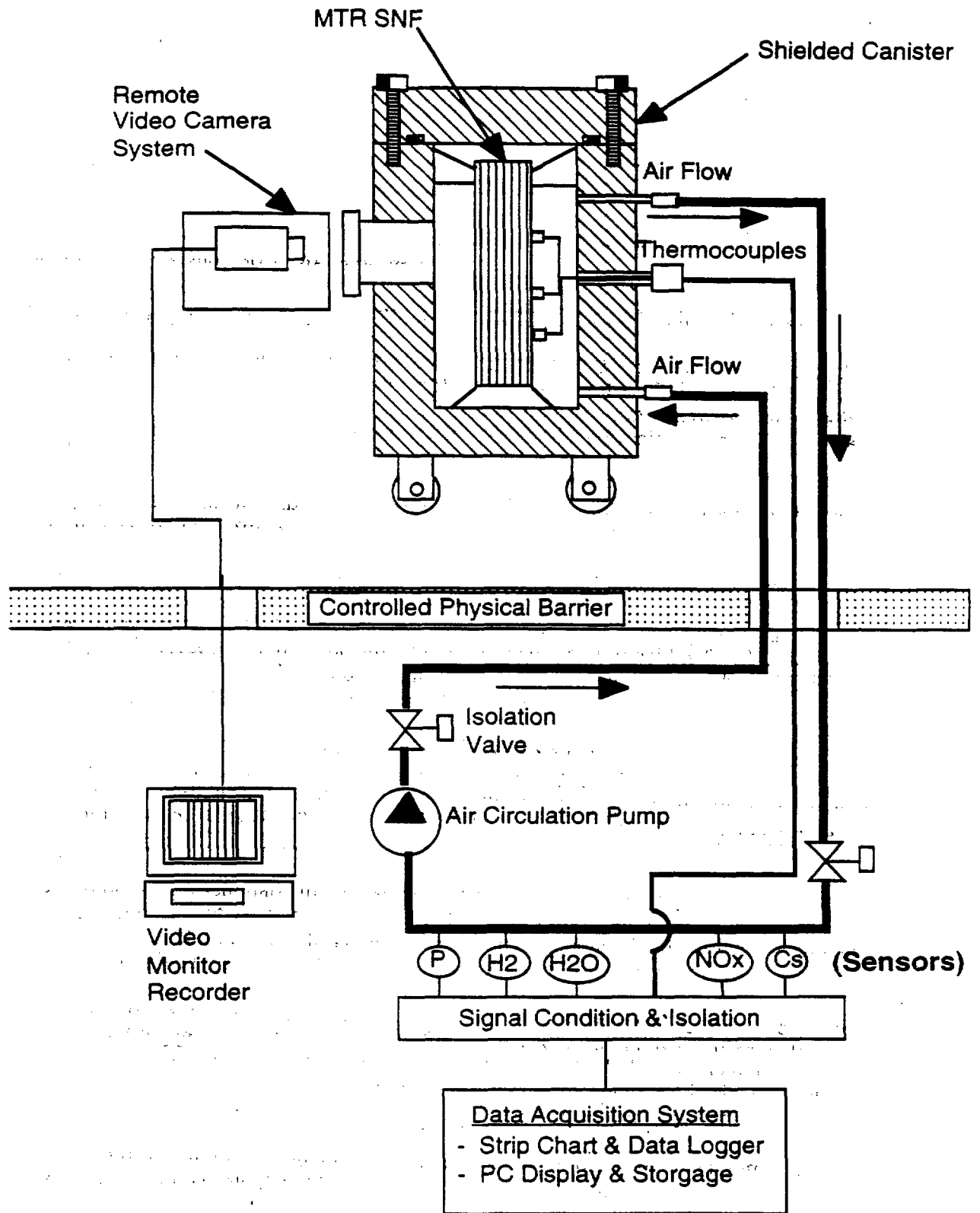


Figure 3.1 - Shielded Test Canister and Data Acquisition System Concept Layout

3.3 Waste form Degradation and Release Studies

The fuel assemblies (structures of fuel elements which themselves are made of fuel cladding and core material), the SNF canister, and the guides within the canister are within the scope of the waste form degradation studies. The initial work has focused on the degradation of the fuel cladding and core materials under storage environments.

The purpose of the waste form degradation studies is to develop information on materials' response (degradation) under potential storage conditions and to develop mathematical models of the degradation processes for input to the repository performance assessment and criticality analyses. Vapor corrosion is the most aggressive form of fuel degradation. A corrosion program to quantify rates and model response to a range of conditions is ongoing. Empirical models have been developed for the corrosion response of the cladding materials to vapor environments. Initial results have been obtained from vapor corrosion tests aluminum-uranium alloy fuel materials.

Corrosion under water vapor conditions is reported. Water for these tests was obtained as condensate from the atmosphere and is relatively free of impurities. It is representative of the water vapor that would surround an AL SNF assembly and its canister during dry storage at SRS. A model for corrosion in high quality water has been developed. It is anticipated that the degradation of the overpack may alter the water chemistry which may affect the corrosion response of the waste form.

Tests to measure the release of volatile species from AL SNF have begun. Results will allow estimation of this release from the fuel under interim dry and repository storage conditions.

3.3.1 Vapor Corrosion Program

This section covers the current status of a portion of Subtask 2.1, Release Studies Program, of the Direct Disposal Technologies. This vapor corrosion test program is a continuation of a previous task on interim dry storage of the aluminum clad spent nuclear fuels.

Aluminum alloys 1100, 5052, and 6061 were chosen for the corrosion tests because their chemical compositions are similar to those of most of the FRR/DRR cladding alloys. The following degradation scenarios have been considered:

- (i) The cladding materials are subject to unlimited corrodant species (water vapor only and water vapor with nitric acid to simulate radiolysis effects) at elevated temperatures. The tests were carried out in an autoclave.
- (ii) The cladding materials are subject to limited corrodant species (water vapor only and water vapor with nitric acid to simulate radiolysis effects) at elevated temperatures. The tests were carried out with specimens enclosed in specially designed stainless steel capsules placed in ovens.
- (iii) The cladding materials are subject to limited corrodant species (water vapor) and exposed to a cobalt 60 high radiation source (~2,000,000 Rad/hour) at elevated temperatures. The tests were carried out with specimens enclosed in stainless steel capsules placed in a cobalt 60 gamma cell.

In addition, preliminary water vapor corrosion tests for a fuel material (aluminum-10 wt% uranium) have been carried out. Some sections of aluminum 8001 clad fuel tubes were also tested for general and pitting corrosion under water vapor environments.

Degradation models for aluminum clad spent nuclear fuels were developed based on the corrosion test results. These models can be used for input to the performance assessment analyses of the national repository. The corrosion models, along with the aluminum oxidation chemical balance equation, will allow a prediction of hydrogen generation in a closed system. The hydrogen generation must be limited to avoid the explosive range that is 4 to 75% by volume in air.

Water analyses were performed before and after the tests to detect any undesired impurities that would adversely affect the test results. The initial impurity concentrations in the condensate water are shown in Table 3.2.

Table 3.2 - Condensate Water Chemical Analysis

pH	6.390 - 7.944
Conductivity	24 to 45.8 $\mu\text{S}/\text{cm}$
Chloride	2.69 - 3.56 $\mu\text{g}/\text{ml}$
Fluoride	0.096 $\mu\text{g}/\text{ml}$
Nitrate	< 1.5 $\mu\text{g}/\text{ml}$
Nitrite	ND
Sulphate	3.97 - 4.28 $\mu\text{g}/\text{ml}$
Ca	0.797 - 4.501 $\mu\text{g}/\text{ml}$
Mg	0.279 - 0.325 $\mu\text{g}/\text{ml}$
Al	0.019 - 0.204 $\mu\text{g}/\text{ml}$
Fe	0.067 - 0.016 $\mu\text{g}/\text{ml}$
Na	2.916 - 4.881 $\mu\text{g}/\text{ml}$

The specimens were prepared with 600-grit surface finish before testing to provide a uniform and consistent surface condition for all specimens. Photographs of the samples were taken before and after tests. Surfaces and cross sections were examined by scanning electron microscopy. The X-ray diffraction (XRD) was used to identify the aluminum oxide film type after exposure. It is expected that Boehmite crystals ($\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$) will form on the sample surfaces because the test temperatures were above 80°C (except for one of the gamma cell tests) throughout the investigation.

3.3.1.1 Vapor Corrosion of Aluminum Claddings with Unlimited Species

This series of corrosion test involves the use of a one-gallon, stainless steel enclosed autoclave. Saturated water vapor (100% relative humidity or 100% R. H.) environment was achieved by adding sufficient condensate water (150 ml) in the bottom of the autoclave. Elevated temperatures (150 or 200°C) were maintained throughout the entire test period. A data acquisition system was developed to automatically record the temperature, pressure, and relative humidity inside the autoclave at 15-minute intervals. This environment represents a severe and limiting spent nuclear fuels storage condition.

The specimens were hung in the middle of the autoclave where temperature distribution was uniform. The galvanic effect between the stainless steel vessel and the aluminum samples was eliminated by using Teflon™ separators and aluminum hangers for the samples.

The specimens were weighed initially before the test. After each test interval, the samples were taken out of the autoclave and dried in a desiccator at room temperature under vacuum for about 30 minutes. This procedure insured that any adsorbed moisture on the surface was removed and only the weight gain due to oxidation of the aluminum was obtained. The weight gain data in $\mu\text{g}/\text{dm}^2$ were obtained by normalizing the total weight gain by the sample surface area (typically, for a test coupon of 2-inch x 0.75-inch x 0.125-inch with a nominal hole of 0.125-inch diameter, the surface area is 0.2395 dm^2). After re-weighing, the samples were put back in the autoclave for the next test interval.

Results of Vapor Corrosion Tests- Aluminum Cladding Alloys, Unlimited Species

Total exposures of 1400 and 5100 hours were accumulated, respectively, for 150°C and 200°C tests. In general, the data for aluminum 1100 and 6061 show a parabolic corrosion behavior. Although the parabolic behavior can also be seen for 5052 initially (<1400 hours at 200°C), break-away corrosion occurred at a weight gain of about 100,000 $\mu\text{g}/\text{dm}^2$ (Fig. 3.2). During post-breakaway behavior there is a linear relationship between the weight gain and time of exposure. Note that in Figure 3.2 the break-away data for 150°C were obtained by exposing the coupon samples which were previously tested at 200°C for 5100 hours (already developed break-away corrosion behavior, at 200°C). It was assumed that the break-away is controlled by the oxide film thickness (which is directly proportional to the weight gain). Therefore, the parabolic corrosion model predicts that for a weight gain of 100,000 $\mu\text{g}/\text{dm}^2$, breakaway corrosion at 150°C would occur after 16,000 hours exposure. This re-use of test coupons is a accelerated study of break-away corrosion for a different temperature assuming that the same corrosion mechanism controls the process.

All the autoclave data can be summarized in a logarithmic plot (Fig. 3.3). Also included in this figure is the data of Godard (1967) from tests at room temperature in a saturated water vapor environment for an aluminum alloy CA-3S which is very similar to alloy 1100 in chemical composition. Note that at room temperature, bayerite or gibbsite ($\text{Al}_2\text{O}_3 \cdot 3\text{H}_2\text{O}$), rather than Boehmite ($\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$), is more likely to form on the aluminum sample surface. Godard assumed in his paper that the oxide form was Al_2O_3 without any water of hydration.

In a radioactive environment, NO_x gas will form by ionization of the air. These gases will further react with water vapor and condense on the fuel clad surface as nitric acid which is an aggressive corrodant to the aluminum alloys. To simulate this radiation effect, a 10 wt% stock nitric acid (HNO_3) was added to the water condensate to achieve a pH value of 1.0 in the solution mixture. The specimens were then tested at 150°C for only one week only because the nitric acid vapor may be harmful to the equipment. Very thick oxide layer was formed and sloughing-off was observed. X-Ray diffraction has confirmed that these oxides were indeed Boehmite. In this case, the weight gain data is not a reliable guide to corrosion of the aluminum. After the oxide on the specimens was chemically removed, aluminum 5052 samples exhibited severe pitting corrosion, while 1100 and 6061 showed general corrosion. It is noticed that the corrosion in an acidic

water vapor environment is about two orders of magnitude higher than in an environment of water vapor only. This implies that radiolysis could have caused a significant increase of corrosion with the presence of water vapor, especially under an elevated temperature.

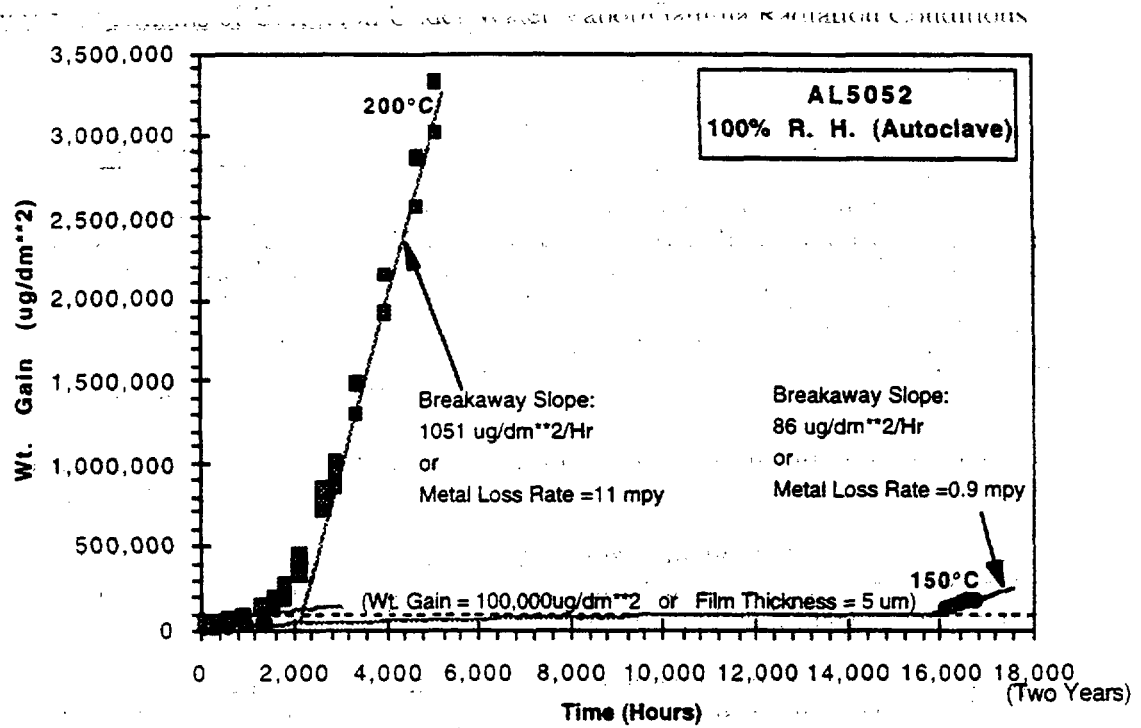


Figure 3.2 - Break-Away corrosion in Aluminum 5052

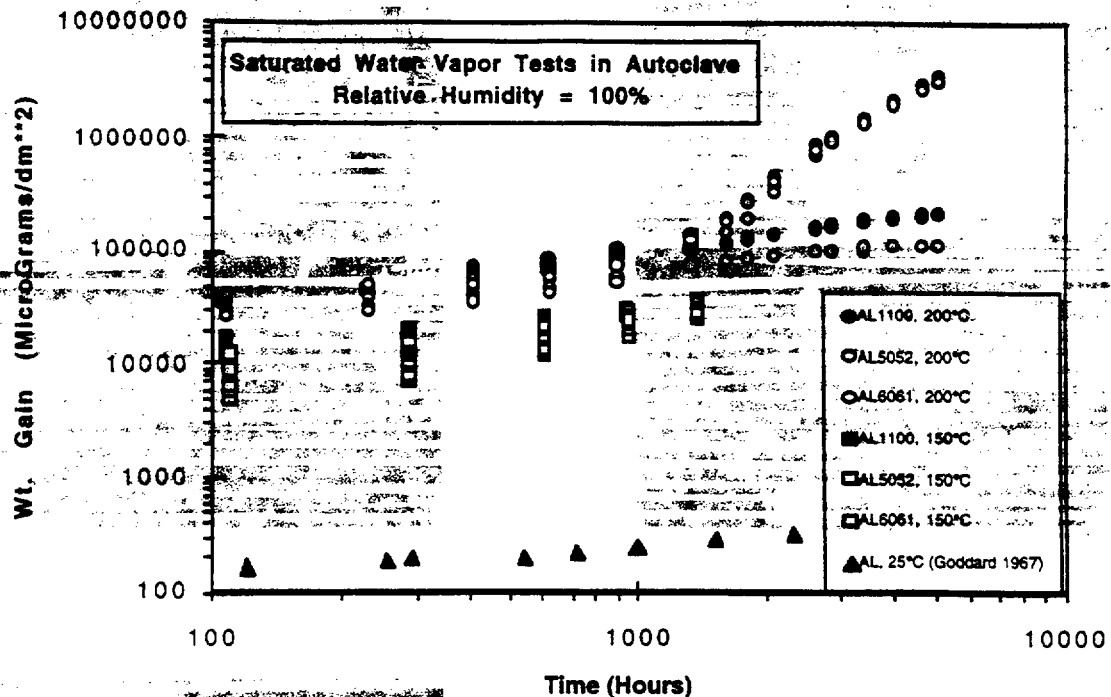


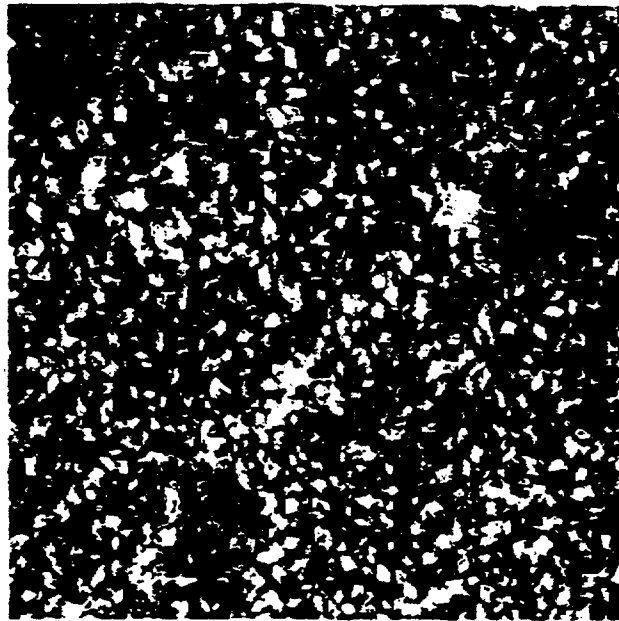
Figure 3.3 - Autoclave Test at 100% Relative Humidity (Saturated Water Vapor)

NOTE: Aluminum Consumed (metal loss in mils) = $1.193E-6 \times$ Weight Gain (in $\mu\text{g}/\text{dm}^2$); Oxide Film Thickness (Boehmite film in nm) = $0.053319 \times$ Weight Gain (in $\mu\text{g}/\text{dm}^2$)

Micromechanisms of Corrosion in High Temperature Water Vapor

Figure 3.4(a)[†] shows a typical Boehmite crystal formation (500X) on the surface of an aluminum specimen after testing. At a magnification factor of 1000X, the Boehmite crystals are shown in Figure 3.4(b). Figures 3.5(a) and 3.5(b) are, respectively, the cross-sections of alloy 1100 and 5052 samples with identical exposure history at which time the break-away corrosion of 5052 has occurred (Fig. 3.2). It can be seen in alloy 1100 (also

[†] Figures 3.4 and 3.5 were obtained at the University of South Carolina at Columbia by Professor Anthony P. Reynolds in the Department of Mechanical Engineering under SCUREF (South Carolina Universities Research and Education Foundation) Task No. 212.



(a) Boehmite Crystals on Aluminum 6061 (2000X)



(b) Boehmite Crystals on Aluminum 6061 (10,000X)

Figure 3.4 Typical Oxide Film On Specimen Surface



(a) Aluminum 1100 Substrate is Intact under the Oxide Film



(b) Internal Oxidation Occurred in Substrate Aluminum 5052

**Figure 3.5 Microstructures Of Pre- And Post Break-Away Corrosion
(Short Dimension of the Photographs is Approximately 180 μm)**

in the case of alloy 6061 which is not shown in this report) that the Boehmite layers protected the metal substrate. However, the optical photograph in Figure 3.5(b) reveals channels and pockets or holes formed by internal oxidation of the metal substrate in alloy 5052. This is in contrast to the general belief that microcracks were formed in the oxide layer itself leading to a direct path from the environment to the bare metal. A 4000X SEM photograph confirmed that Boehmite crystals appeared in the internal oxidation sites. The cause of this internal oxidation in alloy 5052 is unclear, but it appears to be linked to the high magnesium content and possible magnesium segregation to internal boundaries. Other evidence showed that the grain boundaries were under attack prior to the formation of the pockets or holes in Figure 3.5(b). ~~The depth of attack for this sample is about 150 μm . The exposure time was 2909 hours.~~

Vapor Corrosion Models

The oxide growth rate for general corrosion is proportional to the ion concentration gradient. It can be shown that a power law follows:

$$W = (Ct)^n$$

where W is the weight gain (or the oxide film thickness which is proportional to the weight gain, if internal oxidation does not occur), t is the exposure time, n is an exponent ($n = 0.5$ for a parabolic model), and C is related to the concentration of the diffusing species. An Arrhenius relation is used to describe the temperature dependence of the weight gain. Therefore,

$$W = A \cdot \text{EXP}(-Q_1/n/RT) \cdot t^n$$

where A is an coefficient which may be a function of material, relative humidity, corrodant species, etc., Q_1 may be related to the diffusion of oxygen in the oxide film or activation energy of Boehmite, R is the universal gas constant, T is the absolute temperature in Kelvin.

In the breakaway corrosion regime, it is also assumed that an Arrhenius relation exists for the weight gain (W) (the straight line portion of the weight gain in Figure 3.2). Note that in this regime, the oxide film thickness is no longer proportional to the weight gain, but it is still proportional to the metal loss (metal consumption). The Arrhenius relation for the weight gain in the break-away regime is

$$W = B_0 + B \cdot \text{EXP}(-Q_2/RT) \cdot t$$

where B and B_0 are coefficients which may be functions of material, relative humidity, corrodant species, etc., and Q_2 may be related to the diffusion of oxygen through the crystal lattice or along microstructural paths.

With the above general models, a curve fitting algorithm was developed using weight gain data from tests at temperatures of 150 and 200°C. The weight gain equations (for corrosion in saturated water vapor environments) for each of the aluminum alloys are:

Alloy 1100:

$$\text{Wt. Gain (in } \mu\text{g}/\text{dm}^2) = 2.1879 \times 10^8 \cdot \text{EXP}[-4941 / T(\text{K})] \cdot (\text{Hours})^{0.4018}$$

Alloy 5052:

(1) Before break-away corrosion

$$\text{Wt. Gain (in } \mu\text{g/dm}^2\text{)} = 2.8072 \times 10^8 \cdot \text{EXP}[-5600 / \text{T(K)}] \cdot (\text{Hours})^{0.5481}$$

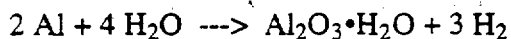
(2) Post-Breakaway

$$\text{Rate of Weight Gain (} \mu\text{g/dm}^2 \text{ per Hour)} = 1.6653 \times 10^{12} \cdot \text{EXP}[-10023 / \text{T (}^\circ\text{K)}]$$

Alloy 6061:

$$\text{Wt. Gain (in } \mu\text{g/dm}^2\text{)} = 3.3010 \times 10^6 \cdot \text{EXP}[-3432 / \text{T(K)}] \cdot (\text{Hours})^{0.4694}$$

Based on the corrosion of aluminum to form Boehmite,



the following conversion formulae in terms of weight gain value can be derived:

$$\text{Aluminum Consumed (or Metal Loss in mils)} = 1.193\text{E-}6 \text{ X Weight Gain (in } \mu\text{g/dm}^2\text{)}$$

$$\text{Oxide Film Thickness (Boehmite film in nm)} = 0.053319 \text{ X Weight Gain (in } \mu\text{g/dm}^2\text{)}$$

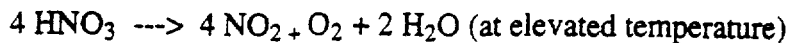
3.3.1.2. Vapor Corrosion of Aluminum Claddings with Limited Species (no radiation)

The autoclave has been used for 100% relative humidity water vapor test. In principal, the autoclave can also be used for tests other than 100% relative humidity by calculating the amount of water condensate that is needed. However, the amount of water vapor that may condense on the cooler parts of the autoclave is uncertain, and the relative humidity cannot be controlled. In addition, long term tests with nitric acid to simulate radiation would cause damage to the stainless steel autoclave. Therefore, specially-designed stainless steel capsules placed within furnaces were used for both the water vapor and acidic water vapor tests. The capsule tests are more realistic than tests in an open system as the fuel assemblies would be in sealed canisters. Under these conditions, corrosion is essentially stopped when the corrodant species, such as water and oxygen, are consumed by the oxidation process, as long as the encapsulating system remains intact. The amount of water or the dryness allowed in the system can be obtained from the test results.

The capsules are made of two 1.5 inch pipe caps (outside diameter is 1.9 inch) with a volume of about 78 ml. Each capsule can hold three test coupons. Normally, aluminum alloy capsules contained one of each type aluminum alloy (1100, 5052, and 6061).

The amount of water condensate to be injected into the sample-containing capsule was calculated based on the saturation water vapor pressure corresponding to the test temperature, the targeted initial relative humidity (e.g., 20, 50, 80, or 100%), and the free volume (air space) of the capsule.

Similar to the nitric acid vapor test in the autoclave, a nitric acid solution with initial pH about 1.0 was prepared for the capsule test. The mixture was made from one part of 10 wt% stock nitric acid and six parts of water condensate. In estimating the initial relative humidity, the small amount of water from the breakdown of HNO₃ at high temperature was also accounted for, that is,



The vapor saturation pressures at 150, 200, and 250°C are, respectively, 69, 226, and 577 psia. Therefore, for testing at high temperature, it is necessary to have capsules pressure-checked. Also, to insure that the system is sealed, a leak-check is performed.

Before the tests, the weights of capsules and test specimens were recorded. The capsules were put in ovens and were taken out according to the assigned test duration. The capsules were then cut open and the samples inside the capsules were weighed.

The test at 150°C has been completed; test intervals were 3 weeks, 3, 6, and 12 months. The tests at 200° and 250°C are in progress.

Results of Vapor Corrosion Tests- Aluminum Cladding Alloys: Limited Species

Figure 3.6 summarizes the capsule test data for aluminum 6061 at 150°C for 12 months. The aluminum 1100 and 5052 behaved similarly. Note that the relative humidity values labeled in this figure are their initial values. As the exposure time increases, the relative humidity decreases due to the consumption of water in the corrosion process. It can be seen that:

- (1) Vapor from the nitric acid solution, or the simulated radiolysis environment, has a significantly increases corrosion.
- (2) Corrosion is more extensive in an environment with higher relative humidity.
- (3) The corrosion rate will decrease and will eventually cease. The capsule is a completely closed system. The amount of air, water, and/or acid is fixed as soon as the capsules were sealed; the corrodant species were depleted due to the oxidation reaction ($2 \text{Al} + 4 \text{H}_2\text{O} \rightarrow \text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O} + 3 \text{H}_2$).
- (4) A critical relative humidity between 40 to 70% exists below which practically no corrosion occurs at room temperature was reported (ref. Vernon, 1931; Ref. 5 in WSRC-TR-95-0345). It appears that in the current capsule test, the threshold relative humidity at 150°C is 20% for 1100 and 6061 because no distinct weight gain was observed up to 9000 hours.
- (5) Based on the present data, the threshold relative humidity for alloy 5052 under a water vapor environment at 150°C is less than 20%. All three aluminum alloys (1100, 5052, 6061) exhibited weight gains in the acidic or simulated radiation environment at 20% relative humidity and therefore observed threshold was observed.
- (6) In a closed system, the corrosion will stop when all the corrodant species (e.g., free water and oxygen) are consumed; therefore, the absence of a threshold value below which no corrosion is detected does not preclude acceptable fuel storage in a completely sealed containment system as long as the acceptance criteria in terms of the allowable corrosion or allowable amount of free water are specified.
- (7) At the end of test, the residual relative humidity, vapor pressure, and hydrogen generation can be calculated based on the weight gain data.

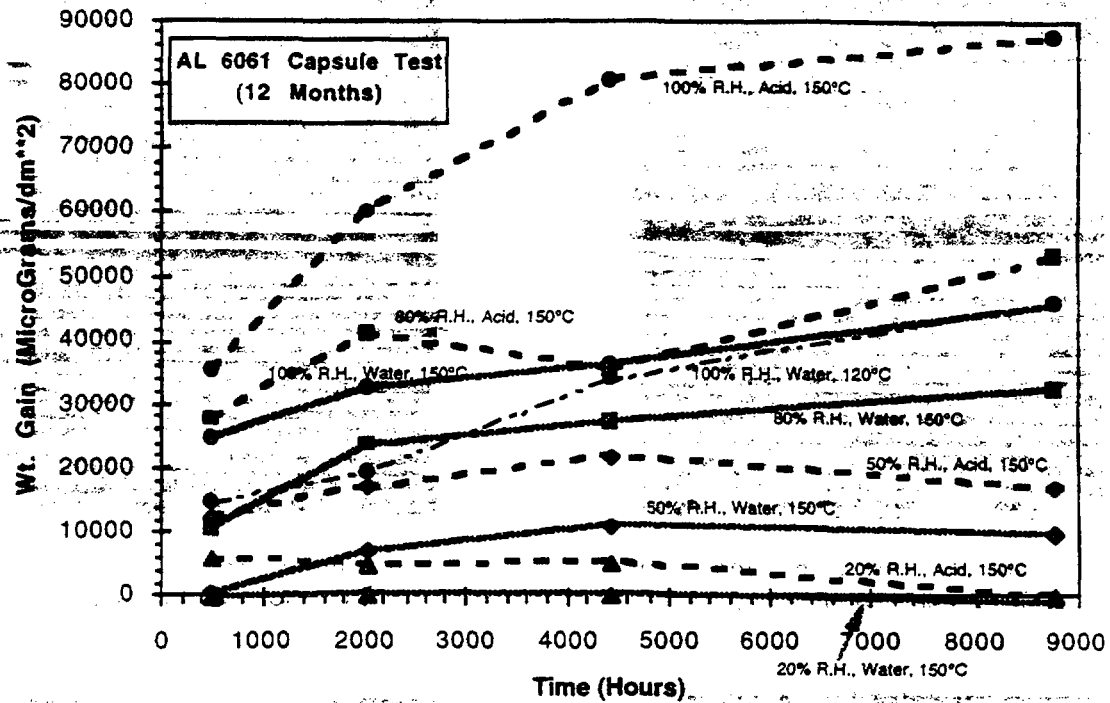


Figure 3.6 Water And Nitric Acid Vapor Corrosion For Aluminum 6061 In Capsules

A verification test was run at 120°C in a separate oven. Four capsules were prepared with 100% R. H. water condensate and were taken out for weight gain measurement at the same time intervals as the 150°C capsules (3 weeks, 3, 6, and 12 months). It was demonstrated that the weight gain curve of 120°C is indeed below that obtained at a 100% relative humidity with a higher temperature, 150°C.

3.3.1.3 Vapor Corrosion of Aluminum Cladding with Limited Species Under Gamma Radiation

Actual storage of spent nuclear fuel will be under a yet-to-be-determined radiation field, which depends on the package configurations, type of fuel, fuel burn-up, etc. As has been demonstrated (nitric acid water vapor environments), the corrosion rates were significantly increased in the tests under the simulated radiation environment. To investigate corrosion behavior under an ionizing radiation field, the stainless steel capsules were again employed.

A rack to hold six capsules was designed. A heating element was placed in the center of the rack. The assembly was put inside a can which is insulated and provides more uniform temperature distribution for the capsules. Building air was introduced to the exterior of the can for cooling purpose so that the gamma cell chamber would maintain a safe operating temperature (below 100°C). The gamma cell has a very limited opening

for installing monitoring instrumentation. Two thermocouple lead wires, a electric cord for the heating element, and an air tube almost occupied the entire entrance opening.

The capsules, each containing an 1100, a 5052, and a 6061 specimen, were filled with water condensate to produce initial relative humidities of 20, 50, 100% and tested at 200°C for 1, 4, 8, and 12 weeks. A thermocouple was attached to a capsule to confirm the test temperature and a separate thermocouple was mounted on the outside of the can (which enclosed the rack and capsules) to prevent overheating the gamma cell. The gamma cell contains a cobalt 60 source and has a fixed dose rate of 1,810,000 Rad/hour (current value). Note that this intensity is very high, and it is higher than any possible radiation field intensity expected during spent nuclear fuel storage in the repository.

In addition, a separate test was carried out at the normal operating temperature of the gamma cell--about 78 to 80°C. This test investigated the low temperature radiolysis effect in a saturated vapor environment. Two capsules, each containing 1100, 5052, and 6061 test coupons, were exposed for periods of one or four weeks. These capsules were filled with excess water condensate to maintain 100% relative humidity throughout the entire test period.

Results of Vapor Corrosion- Aluminum Cladding Alloys, Limited Species, Radiation Environment

A typical result under gamma radiation at 200°C in water vapor for aluminum 6061 is shown in Figure 3.7. The dotted curve corresponds to the test result of unlimited species corrosion test in an autoclave under saturated water vapor condition and is inserted for reference only. It is seen that in the case of initial 100% relative humidity, the radiolysis effects in the water/air environment caused a weight gain about three times as great from that in the water/air environment without radiation in the early stage of the corrosion. After a very short period of time, less than a week, most of the corrosion has occurred and either the usable water vapor corrodant has been depleted or the oxide film has reached a thickness sufficient to significantly decrease the corrosion rate. This is indicated by the flattening of the weight gain curves (of the capsule test in gamma cell). Figure 3.7 also includes a data point for "zero" relative humidity in which case the capsule was loaded with test coupons at ambient room conditions. A small weight gain was observed.

The radiolysis effects at lower temperature are also of interest. The spent fuel is not always subject to the specified upper limit of the repository temperature. A capsule test was carried out at the normal operating temperature of the gamma cell of about 78 to 80°C. Two data points were obtained after exposures of one and four weeks, respectively. These capsules were filled with a sufficient amount of water to maintain a constant 100% relative humidity throughout the test periods. The data points for aluminum 1100 (filled circles) are shown in Figure 3.8. This figure also includes: 1) a reference set of unlimited species test data (autoclave result) at 150°C and its fitted curve (see Section 3.3.1.1 or the Arrhenius equations shown in figures); 2) a set of data obtained by Godard (1967) for CA-3S aluminum which is similar to aluminum 1100; and 3) predicted corrosion responses at 78 and 25°C based on the Arrhenius equations for 1100. It can be seen that the 1100 prediction curve at 25°C closely represent the Godard's CA-3S data. In contrast, the weight gain data for the 78°C test are comparable to the 150°C data without radiation and are at least 5 times the predictive values at the end of the 4-week test at 78°C.

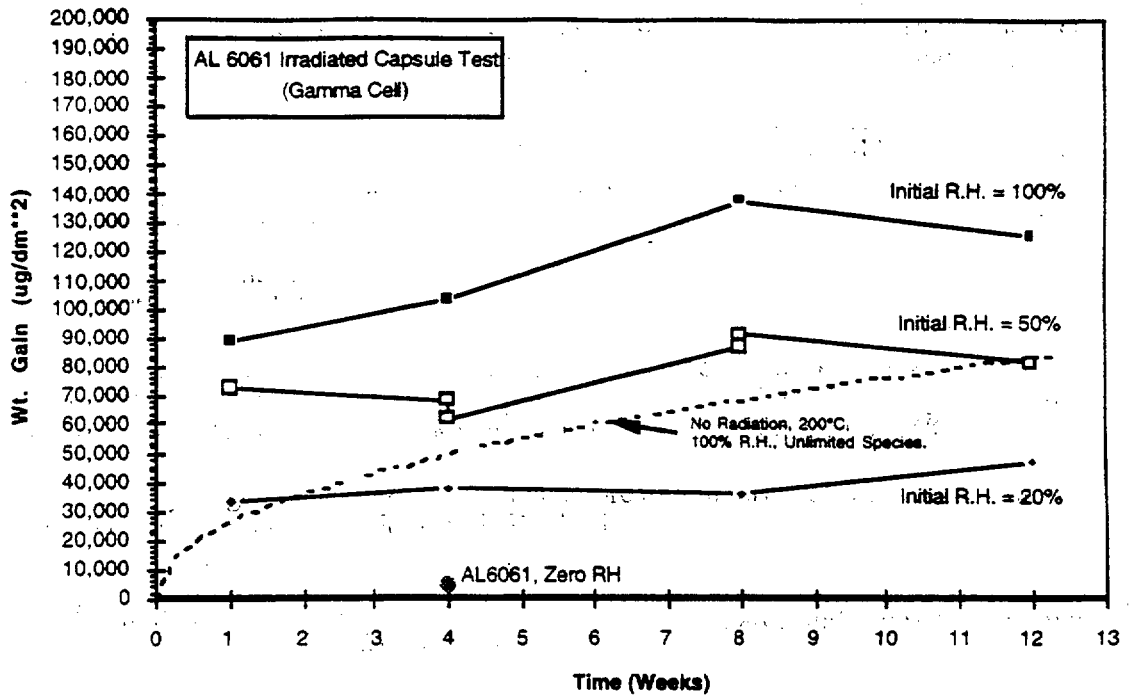


Figure 3.7 Water Vapor Corrosion Of Aluminum 6061 Under Radiation Field At 200°C

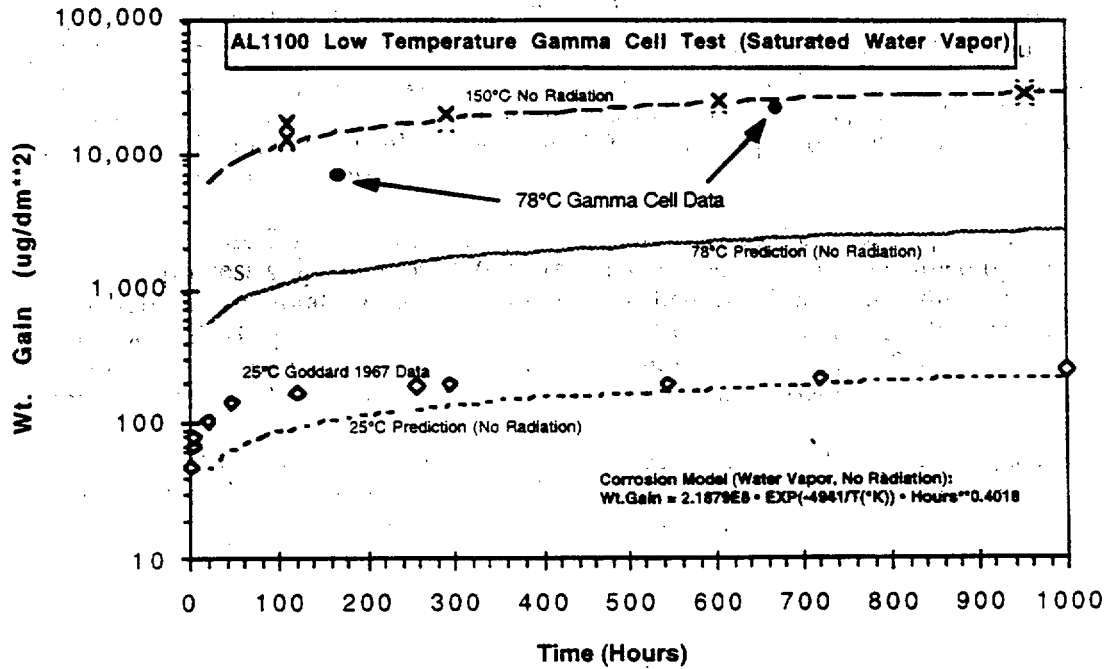


Figure 3.8 - Effects Of Water Vapor Corrosion Under Radiation Field At 78°C

The test showed that the corrosion of aluminum 5052 was more than for 1100 and 6061. The full impact due to radiation is yet to be assessed (e.g., microstructural examination of 5052 in cross-section) because internal oxidation is known now to exist in 5052 alloy in high temperature tests (Figs. 3.2 and 3.5(b)).

It is clear from the test data that a threshold for relative humidity does not exist in gamma radiation field. A noticeable amount of corrosion has occurred at 20% relative humidity. However, in this test with a high dose rate, the water vapor corrodant is consumed quickly in the first week and the corrosion rate is then significantly reduced.

Micromechanisms of Corrosion Under Water Vapor/Gamma Radiation Conditions

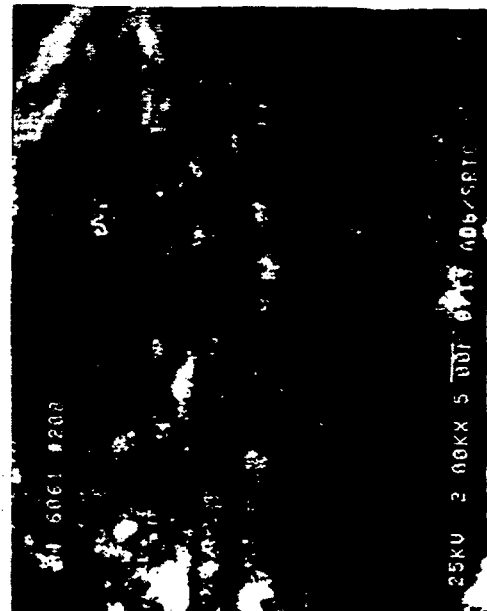
The surfaces of the test coupon were examined by SEM for all 100% relative humidity specimens. Compared to the specimens exposed to water vapor only, a completely different oxide morphology was formed in the presence of a gamma radiation field, as shown in Figure 3.9 for 6061 after only one-week exposure under 1.81×10^6 Rad/hour. The oxide surface (Figure 3.9a) is no longer evenly paved by Boehmite crystals with similar sizes (as in Figure 3.4). The flatter surfaces are still covered by fine crystals (Figure 3.9d). Variable sizes of crystal clusters can be seen in figures 3.9b and 3.9c, along with crater-like opened blisters. Some blisters appeared intact, especially in the one-week specimens. As the exposure time increases, more blisters appeared to be cracked. Some open blisters show the underlying crystalline structure, believed to be Boehmite as determined by XRD.

The following observations can be noted from a careful examination of the morphologies of all three alloys, :

1. AL 1100 The blisters appear as empty rims. Smaller but closed (intact) blisters appear with continued exposure. The 1100 blisters are smaller than those in the other two alloys. The smooth background areas are covered with fine crystalline oxide.
2. AL 5052 The number of blisters and the number of blisters with broken tops increase with exposure time. Very few open blisters have rims. The specimen surface is completely covered by blisters in 8 weeks.
3. AL 6061 The closed, broken, and rimmed blisters are present. Some open blisters have clusters of crystalline oxide inside. Other areas also show crystalline clusters. The specimen surface was not completely covered by blisters.



(a) 500X



(b) 2000X



(c) 10,000X



(d) 10,000X

Figure 3.9 - Aluminum 6061 Exposed To Gamma Radiation Field

3.3.1.4 Vapor Corrosion of Aluminum-Uranium Alloys

Most of the FRR/DRR fuels were made of aluminum-uranium alloys. The corrosion behavior, including general corrosion and pitting corrosion, become important when the aluminum claddings are penetrated or consumed. The corrosion products and their behavior will be input to the waste form performance studies for the direct disposal of spent nuclear fuels.

Aluminum-10 wt% uranium alloy was tested in an autoclave at 200°C under saturated vapor condition. Two types of specimens were used: 1) Coupon specimens which were heavily hot rolled to sheets and then cut to a rectangular shape (2 inches x 0.75 inches x 0.125 inches) similar to the aluminum alloy coupons used throughout this corrosion program; 2) Disk specimens which were made by cutting an extruded rod with diameter 0.624 inches and thickness 0.051 inches. These two sets of specimens were tested separately in the autoclave at 200°C and 100% relative humidity. The specimens were taken out of the autoclave at intervals for weight gain measurement up to exposure times of 1500 to 1700 hours (about 70 days).

Aluminum-18 wt% uranium and aluminum-33 wt% uranium are more representative of the high enriched and low enriched FRR/DRR fuels than the aluminum-10 wt% uranium. Plans have been made to perform tests with alloys of these compositions.

Results of Vapor Corrosion of U-Al Alloy

Figure 3.10 shows the weight gain curves for the coupon specimens and the disk specimens. The initial corrosion was specimen dependent with a large range in weight gain. However, the corrosion rates (the slopes of the curves) appear to be consistent. The corrosion rates differ slightly between the coupon specimens and the disk specimens. Nevertheless, all the weight gains for the aluminum-10 wt.% uranium coupon specimens are much higher than those for aluminum 1100 with the same exposure time at 200°C. The alloy 1100 weight gain is comparable to those for disk specimens No. 7 and No. 8, which are the lower bound of the data shown in Figure 3.10. Reasons for the large range in the initial weight gain are unclear and remain to be investigated. The microstructural inhomogeneity of the alloys may be a factor.

Micromechanisms of Vapor Corrosion of Aluminum-Uranium Alloys

Metallography shows that the hot rolled specimens (coupons) and the extruded specimens (disks) have different microstructures (Fig. 3.11). Large areas of eutectic between elongated bands of primary aluminum can be seen in the extruded specimens. Broken fragments of UAl_x (most likely UAl_4 for the 10 wt% of uranium alloy) are scattered in the aluminum matrix in the hot rolled specimens. These observations account for the different corrosion rates. However, the actual mechanisms causing the discrepancy have not yet been determined.

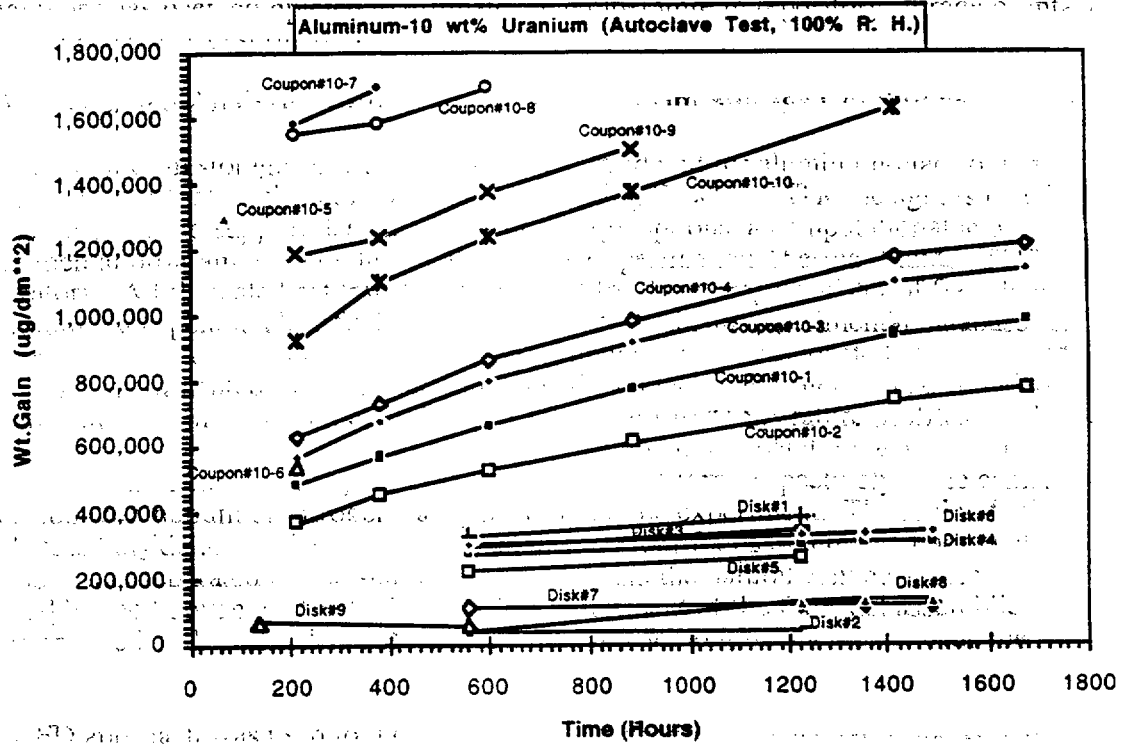
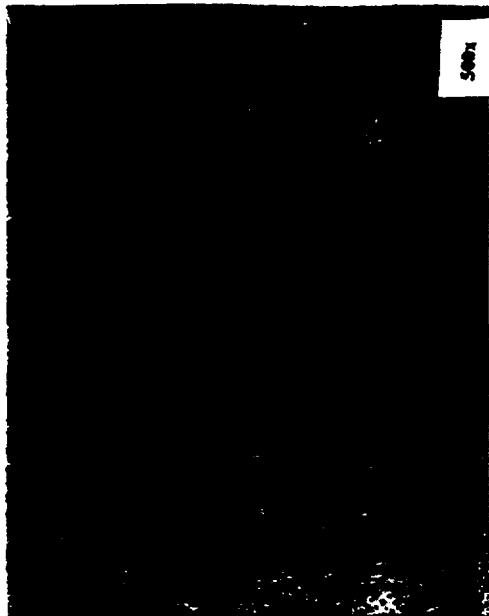
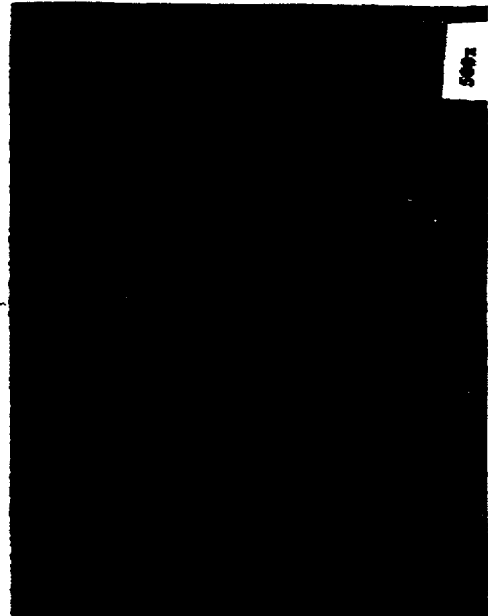


Figure 3.10 Corrosion Of Aluminum-10 Wt% Uranium Alloy At 200°C In Saturated Water Vapor



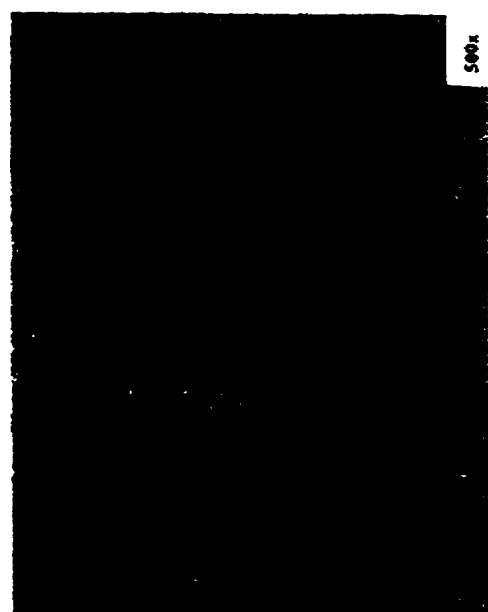
(a) Hot Rolled Aluminum-10 wt% Uranium Alloy in Longitudinal Direction



(b) Hot Rolled Aluminum-10 wt% Uranium Alloy in Transverse Direction



(a) Extruded Aluminum-10 wt% Uranium Alloy in Longitudinal Direction



(b) Extruded Aluminum-10 wt% Uranium Alloy in Transverse Direction

Figure 3.11 Microstructures Of Hot Rolled And Extruded Aluminum-10 Wt% Uranium Alloys

Severe corrosion accompanied by blistering has been noticed in the hot rolled specimens. Optical and scanning electron microscopic photographs were focused on a blistered area in a specimen with only four days of exposure (Figure 3.12). It shows a blistered region covered by a thick oxide layer. In addition, a large number of uranium aluminide particles are scattered in the metal matrix as well as in the oxide layer. This indicates that the uranium aluminide is more stable than aluminum and does not react or reacts very slowly in the 200°C saturated vapor environment.

The aluminum-10 wt% uranium was used for initial testing of Al-U alloys. As noted earlier, typical fuel materials for FRR and DRR are aluminum-18 wt% uranium and aluminum-33 wt% uranium. The microstructures of these two alloys are shown in Figure 3.13. It is obvious that UAl_4 and UAl_3 are the major block-like particles, respectively, in the 18% and 33% uranium alloys. They also notably differ from the aluminum-10 wt% uranium in Figure 3.11. It is expected that the corrosion behaviors will differ. Further tests with these alloys are being planned.

3.3.1.5 Pitting Corrosion of Al-U Alloys in Water Vapor

Artificial pits were milled in the cladding of coextruded fuel tubes (Figs. 3.14(a) and 3.14(c)) of aluminum-18 and -33 wt% uranium fuel clad with 0.030-inch thick aluminum 8001 alloy. Various pit sizes were chosen (1/32, 1/16, and 1/8 inches in diameter) and for each pit size the depths of the pits were 0.010, 0.020, 0.025, and 0.031 (through-clad) inches. Optical metallography was employed to confirm the pit depths.

These ring-shaped specimens were placed in an autoclave at 200°C in saturated water vapor. Another set of the pitting specimens were put in capsules with 20% and 100% relative humidity and tested in 150°C ovens. Nitric acid solution was used for some capsules to simulate the effects of a radiation environment.

Results of Vapor Corrosion- U-Al Alloy, Pit Specimen

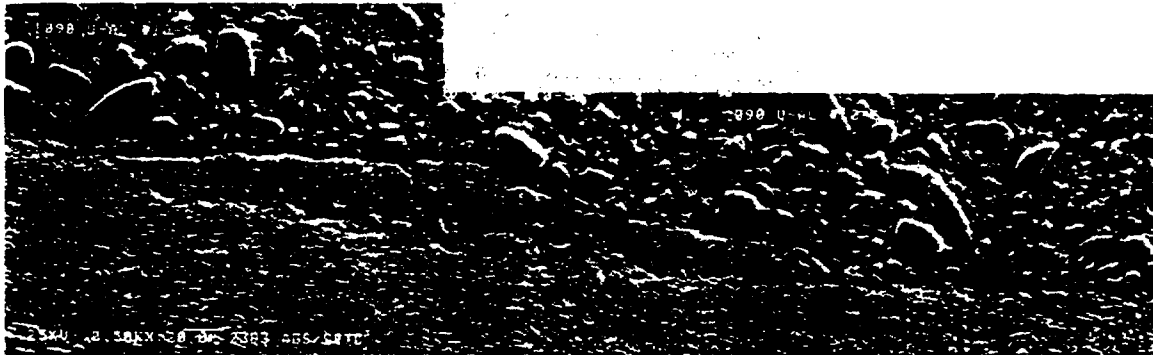
The pitted areas of the 200°C autoclave specimens were sectioned and examined by optical metallography. No penetration by oxidation was found in areas with 0.005 inch residual cladding (corresponding to a pit with a depth of 0.025 inches). For pits machined to penetrate the cladding, uneven oxide was formed in the bottom of the pit, while the oxide layers on the sides of the pits were uniform.

In the autoclave specimens with two-month exposure to saturated water vapor at 200°C, corrosion occurred only in the initially exposed aluminum-uranium core, that is, in the two ends of the samples (Fig. 13(b)) and in the through-the-clad pits (Fig. 13(d)). The black corrosion product was stripped from the pits and was determined by XRD as a mixture of U_4O_9 and Boehmite ($Al_2O_3 \cdot 3H_2O$).

The capsule test at 150°C in a water vapor environment for 12 months showed similar result. In the case of capsules with 20% relative humidity, the degree of oxidation was markedly reduced. However, more pits were found to contain the black oxide in the capsule containing nitric acid and 100% initial relative humidity.



(a) Optical Microscopy



(b) Scanning Electron Microscopy

Figure 3.12 Optical And Scanning Electron Photographs Of A Blistered Area In Aluminum-10 Wt% Uranium Hot Rolled Alloy In Saturated Water Vapor At 200°C For Four Days. (500x)



(a) Aluminum-18 wt% Uranium in Longitudinal Direction (500X)



(b) Aluminum-18 wt% Uranium in Transverse Direction (500X)



(c) Aluminum-33 wt% Uranium in Longitudinal Direction (500X)

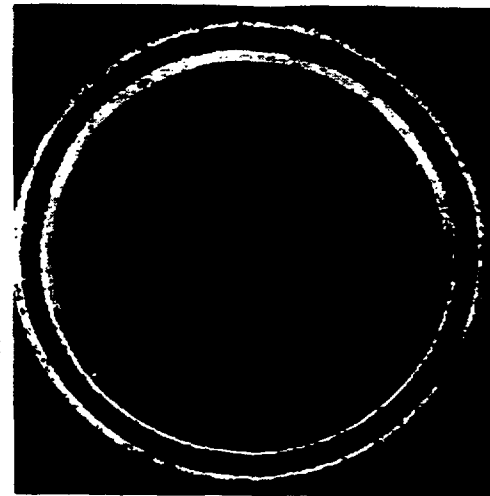


(d) Aluminum-33 wt% Uranium in Transverse Direction (500X)

Figure 3.13 Microstructures of Aluminum-Uranium (18 & 33 Wt%) Alloys



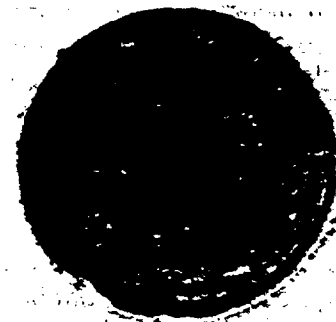
(a) As Received Fuel Tube showing Aluminum 8001 Cladding and Aluminum 18 wt% Uranium Core



(b) Corroded Specimen in a Capsule with Initial 100% R. H. Water Vapor and Nitric Acid at 150°C after 12-Month Exposure



(c) Typical Ring Specimen (Fuel Tube) with Artificial Pits



(d) Black Oxide Formed in a Pit with 1/8" Diameter Exposed to 100% R. H. Water Vapor

Figure 3.14 Typical Ring Specimen (Fuel Tube) And Pitting Corrosion

3.3.1.6 Hydrogen Generation in a Closed System

When the aluminum cladding alloys interact with water vapor, hydrogen gas is generated along with the aluminum oxides (for example, Boehmite). The reaction equation can be found in Section 3.3.1.1. The generation of hydrogen will stop if the corrodant species (water) is depleted. Therefore, in a closed system such as in a test capsule or in the spent nuclear fuel canister, the total amount of hydrogen that can be produced by corrosion and the corresponding hydrogen pressure can be estimated by the ideal gas law and the weight gain equations. It can be shown that

$$P_{\text{hydrogen}} = \frac{1}{V} \frac{m_h}{2} RT$$

where P_{hydrogen} is the pressure of the hydrogen gas, V is the air space or free volume of the closed system, m_h is the mass of hydrogen generated, R is the universal gas constant ($R = 8.314 \text{ J/gmoleK}$ or $\text{N} \cdot \text{m/gmoleK}$), and T is the absolute temperature (K).

The amount of hydrogen generated, m_h , can be expressed in terms of the weight gain of the specimen. Assuming the oxide form is Boehmite,

$$m_h = (6/66) (\text{Weight Gain}), \text{ and} \\ \text{Weight Gain} = (\text{weight gain equation in terms of unit area and time}) \times (\text{specimen surface area})$$

For example, the weight gain equations in Section 3.3.1.1 for alloys 1100, 5052, and 6061 (or equivalently, their pure power law corrosion models) were used to generate Figure 14 for a test capsule with 70 ml volume and containing three standard specimens at 150°C with initial relative humidity of 100%. Figure 3.15 also includes the change of water vapor pressure and relative humidity. The calculation of water pressure requires the input of steam table (vapor pressure as a function of specific volume). The relative humidity is defined as the percentage of vapor pressure in terms of the saturation pressure at the temperature. For example, the saturation pressure for 150°C vapor is 69.046 psia. The total pressure inside the capsule is the sum of the water vapor pressure, the hydrogen gas pressure, and the air pressure.

It can be seen in Figure 3.15 that the hydrogen pressure rises as the water vapor pressure decreases. The process stops in about 5.6 years when all the water is consumed by the corrosion reaction. The total pressure is actually less than its initial value which is the saturation (100% R. H.) pressure of water vapor at 150°C. This calculated decrease in total pressure may not be valid when nitric acid or radiolysis is present. The gas generation in systems other than in water vapor must be characterized.

In the capsule tests that have been carried out, it was observed that the opened capsules after exposure were dry or contained very little moisture. This is a direct evidence of water depletion as a result of oxidation of the aluminum.

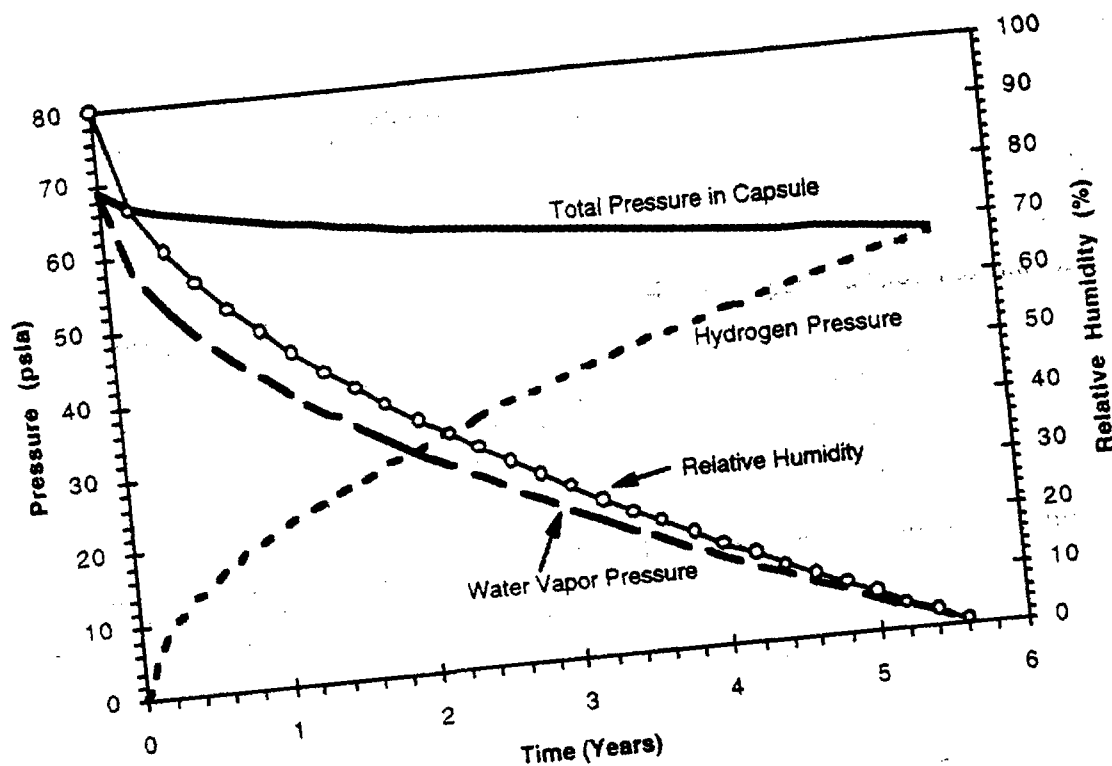
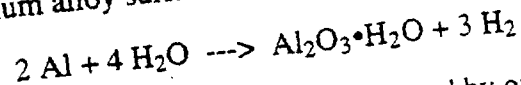


Figure 3.15 Hydrogen Generation And Pressure Changes In A Closed System At 150°C

3.3.1.7 Applications to Storage Systems

(1) Application to Interim Dry Storage or Road Ready Package:

The fuel is most likely to be stored in a sealed system. Limiting the hydrogen generated in a sealed system to 4% by volume will prevent the potential for explosion. The amount of water or the degree of dryness for this system can be calculated based on the chemical reaction equation which conservatively assumes that Boehmite is formed on the aluminum alloy surface:



Assuming all the water will be consumed by oxidation, the maximum allowable water (W_{water} , in grams) is expressed as a function of the free volume (V , in cm^3) of the container:

$$W_{\text{water}} = 3.873 \times 10^{-5} V$$

in which the molar volume of hydrogen, 24789.2 cm^3 , was used.

With the allowable amount of water calculated, the initial relative humidity of the system can be obtained at the specified operating temperature.

In theory, the result of a capsule test at that relative humidity can be used to estimate the cladding corrosion rate. However, the application of the corrosion models (Section 3.3.1.1) developed with the autoclave test data at a constant 100% relative humidity will lead to a conservative estimate of cladding corrosion. The time required for a complete loss of cladding materials is expected to be much longer than the specified time allowed for interim dry storage or in a road ready package. Additional credit can be taken for the integrity of the cladding materials.

(2) Application to the Repository:

For a breached overpack and SNF canister, and under vapor and/or aqueous conditions, the aluminum cladding would be expected to be eventually convert to hydrated oxides. Next the core would be expected to convert to hydrated oxides of aluminum with the fuel particles (e.g. UAl_x) possibly remaining stable. Thermodynamic analysis will be performed for expected chemistry and environmental conditions to predict the range of possible forms of the corroded fuel. Experimental investigation of the stability of the intermetallic fuel particles will also be performed in the future corrosion testing of the fuel materials.

3.3.1.8 Corrosion Program

Tests have been carried out for aluminum alloys 1100, 5052, and 6061 in an autoclave with 100% relative humidity at 150° and 200°C, in completely enclosed capsules at 150°C with various relative humidities, and in a gamma cell at 200°C with various relative humidities, all using SRS atmospheric condensate as the water source. Capsule tests at 200° (with water and nitric acid solution) and at 250°C (with water only) are in progress.

For the fuel materials, aluminum-10 wt% uranium was tested in an autoclave at 200°C and in capsules at 150°C. Weight gain data show specimen-to-specimen variations larger than expected considering the aluminum alloy results. Metallographic examinations are needed to explain the possible role of microstructural variations between the hot rolled and extruded specimens on the corrosion mechanism. Tests for typical fuel materials such as aluminum-18 and -33 wt% uranium alloys should be carried out to obtain relevant corrosion data for FRR/DRR fuels.

Additional cladding and fuel materials will be tested at several relative humidities in a gamma radiation field with the J-13 well water chemistry as the water source. Constant 100% relative humidity capsules should be included to establish the upper bound on the corrosion rate for an air/water environment in the presence of a radiation field. The correlation between the nitric acid test and the gamma cell test will be investigated. The species resulted from the radiolysis of water vapor should be identified for input to radioactive performance assessments and for criticality analyses.

The corrosion models should be refined to include the humidity and radiation effects, if sufficient data are available. This will broaden the applicability of the model to cover both the interim dry and repository environments. The corrosion products will be examined to identify stable phases of aluminum-uranium alloys so that performance assessment and criticality analysis may benefit from the information.

3.3.2 Dissolution Rate in Water

The dissolution of metal fuels in aqueous systems is directly related to their rate of corrosion. A corrosion model for uranium- and uranium alloy-based spent fuel was used in Sandia 1994 performance assessment [Reichard, 1995]:

$$M = A e^{-B/T} (t_2^c - t_1^c) D E S, \text{ where}$$

M is the metal loss due to corrosion from time t_1 to time t_2 , A is an empirical coefficient, B is an Arrhenius activation energy term, c is a power law exponent, D is a function of saturation, E is a function of oxygen concentration, and S is the surface area of the metallic spent fuel. In the INEL PA, the equation was simplified to a linear relationship by assuming that $c=1$ (linear in time), $D=1$ (fully saturated system for conservatism), $E=0.2$ (approximated by the volume concentration of oxygen in dry air. This model is not appropriate for aluminum-based metals which do not have a significant temperature dependence in corrosion rate for temperatures below 100°C and which exhibit passive film formation under neutral pH chemistries similar to aluminum.

From SRTC corrosion tests in storage basin water (high purity), preliminary data using linear polarization testing showed that the consumption rate for both a cladding alloy, 8001 aluminum, and a fuel alloy, aluminum-10%uranium is 0.2 mpy (mills per year) for a 600-grit fresh surface. For an 1100 aluminum specimen with a Boehmite film approximately 1 μm thick, the immersion testing indicated that the rate is 0.02 mpy. No dissolution tests has been carried out for aluminum-uranium alloys. Based on the vapor corrosion test of the extruded aluminum-10 wt% uranium disk specimens (Section 3.3.1.4 and Figure 3.10), the rate of metal consumption is in the order of 0.5 mpy. The reaction in the higher temperature vapor environment is more severe than that of the lower temperature water environment.

Figure 3.10 also includes the weight gain data for hot rolled specimens. It can be seen that the weight gain (or, equivalently, metal consumption) rate is reduced as exposure time increases consistent with the formation of a passive film. This behavior is similar to with that of the SRTC aluminum cladding in tests in a vapor environment and the literature data for aluminum corrosion in high purity water at temperatures less than 100°C [Thomas and Ondrejcin, Journal of Nuc. Mat., 199 (1993), pp. 192-213]. The corrosion in water proceeds in accordance with [Thomas and Ondrejcin]:

$$W = A + B \ln(t), \text{ where}$$

W is the metal consumption, t the exposure time, and A, B are the empirical coefficients.

It is recommended that water dissolution tests should be carried out in J-13 chemistry water. Test protocols for this testing are being established (see section 6.0).

3.3.3 Volatile Species Release at Moderate Temperatures (< 300°C)

Testing to measure the release of volatile species from Al SNF has been initiated at ANL. This testing is being conducted using a specially-constructed furnace which contains controlled-temperature plates to condense gas vapors. A plate type fuel specimen of uranium silicide fuel with cropped ends has been exposed to a temperature of 275°C for 1

month. The preliminary results did not indicate detectable fission products on the collection plates.

The furnace design and preliminary results are being transmitted to SRTC by ANL.

3.4 Spent Fuel Thermal Analysis

A program to validate a heat transfer model and analysis tools for thermal analysis for interim dry storage conditions had been previously completed. An overview of this analysis and the principal results and conclusions are described in section 3.4.1 below. Initial work has been completed to estimate the temperature in repository storage of intact fuels. This work is described in section 3.4.2 below.

3.4.1 Thermal Analysis of Intact Fuel Under Interim Storage Conditions

One of the interim storage configurations being considered for aluminum-clad foreign research reactor fuel, such as the Material and Testing Reactor (MTR) design, is in a dry storage facility. To support design studies of storage options, a computational and experimental program was conducted at the SRS Experimental Thermal Fluids (ETF) Laboratory. A full-scale heat transfer experiment for dry storage canister of flat-plate aluminum-clad spent nuclear fuel was conducted to obtain an experimental database for the verification of computer codes. In the experiment, which was located inside a wind tunnel, the instrumented fuel canister with embedded electrical heater was surrounded by five unheated dummy canisters. Radial and axial heat flux/temperature profiles inside the fuel canister, air velocity outside the canister, and ambient temperature were measured. The canister diameter and height was 0.4064m and 0.9144m, respectively. The canister was filled with helium or nitrogen gas depending on the experiment. The sealed fuel can was located inside the canister and designed to store four fuel assemblies. Each fuel assembly was separated by the stainless steel grid, and the natural convective flow induced by the buoyancy effect within each compartment of the four fuel assemblies communicated with each other only through the top and bottom slot holes inside the canister.

The CFD approach was used to model the three-dimensional convective velocity and temperature distributions within a dry storage canister of MTR fuel elements. A 90° sector of the canister was modeled as a computational domain for the present analysis as illustrated by Figure 3.16. CFDS-FLOW3D code has been used as a CFD tool to simulate the detailed conjugate heat transfer model for the complicated internal geometry of a dry storage canister. The simulation results and analyses were made for the cases with $q''=85$ to 138 watts per MTR fuel element (equivalent to 22 to 35 kW/m³) using various different boundary conditions around the canister wall and different cooling gases (N₂ or He). The CFD approach methodology and benchmarking results against the experimental database have been reported. [3.12]. Figure 3.17 shows temperature contour plot for a typical He-cooled canister. Overall gas flow pattern over the entire flow domain within the canister is illustrated in Figure 3.18 from the simulation results for He- or N₂-cooled canister. The benchmark results provide the verification that the model can predict reasonably accurate buoyancy-driven gas flow and thermal behavior of a typical foreign research reactor fuel (thin aluminum-clad, flat-plate fuel) stored in various storage configurations.

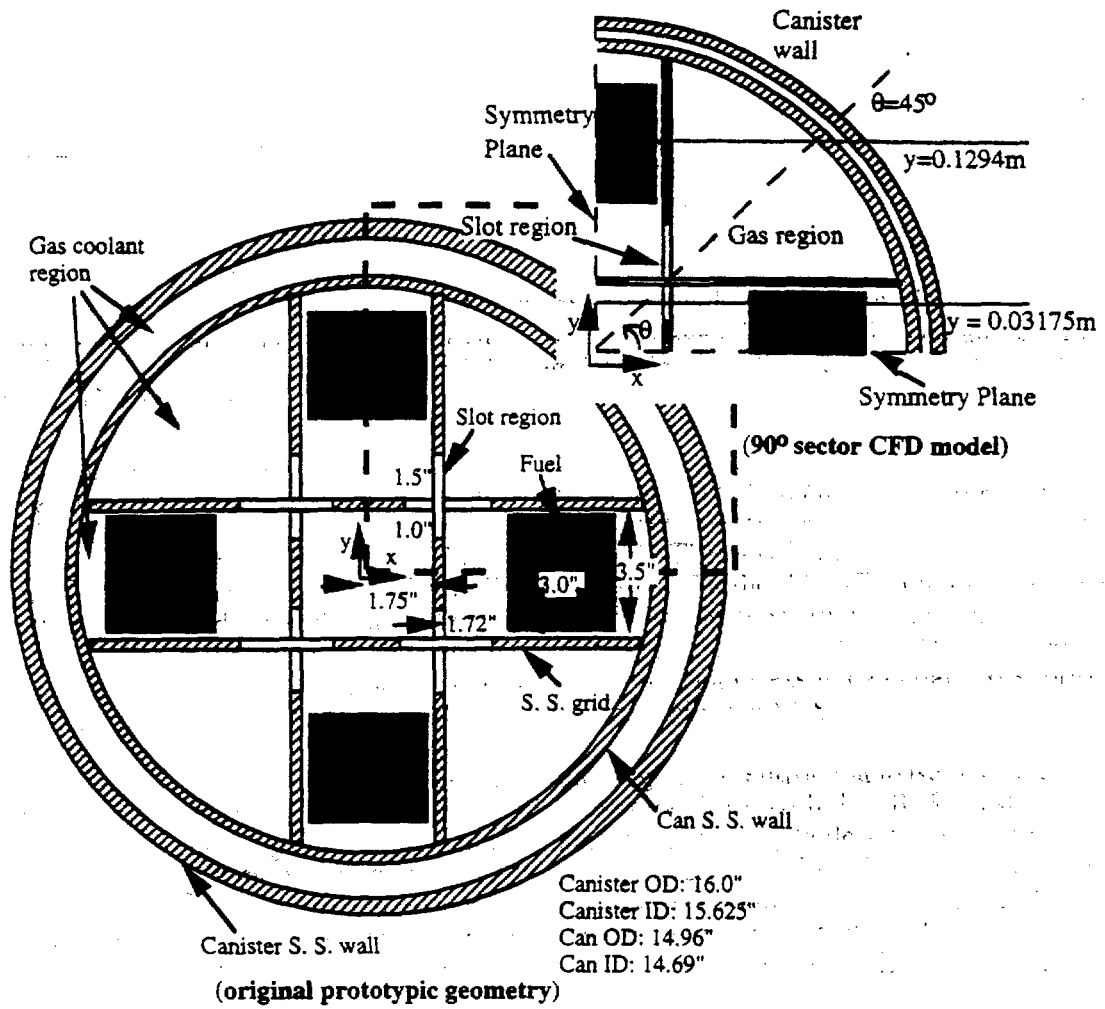


Figure 3.16 - Cross-sectional view of 90° sector model (slot is open only near top and bottom regions)

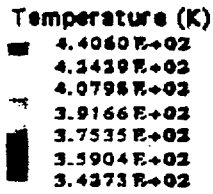


Figure 3.17 - Surface temperature contour plot for a typical He-cooled canister

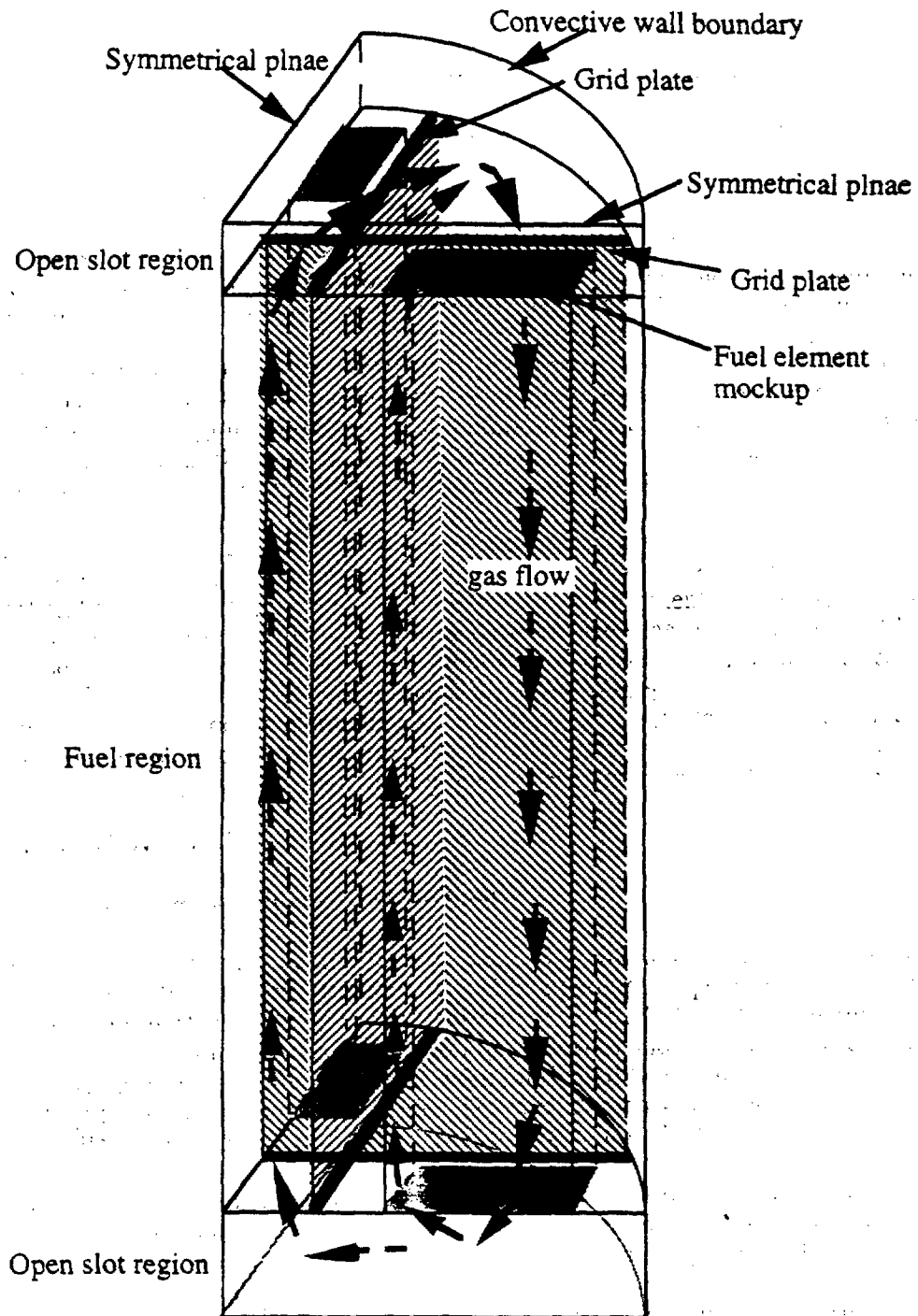


Figure 3.18 - Overall gas flow pattern due to buoyancy-induced natural convection in the 90° sector model of an enclosed canister

3.4.2 Thermal Analysis of Intact Fuel Under Repository Storage Conditions

The thermal performance of the aluminum-clad fuel inside its SNF canister which is stored inside a large co-disposal canister with defense high level waste (DHLW) glass canisters has a strong influence on the kinetics of the aluminum fuel degradation over its storage period in the repository. The number of spent fuel assemblies, their heat loads, and any special conditioning inside a spent fuel canister are still largely unknown while initial studies of criticality, fuel degradation, transportation, and other issues are underway. Thermal loading will have an impact so that an iterative process is needed to come out with the final arrangement in the SNF canister and the co-disposal canister. Consequently, initial thermal studies will be performed over a range of possible conditions and scenarios to narrow down the choices for the acceptable configurations.

The thermal study will use two previous studies as its starting point, referred to here as Study 1 and Study 2. Study 1 analyzed several configurations of SNF and DHLW canisters [3.13]. The two main configurations consisted of 1) a 17-inch diameter SNF canister surrounded by 5 DHLW glass canisters, all contained in a 1754 mm ID co-disposal canister; and 2) five 8-inch diameter SNF canisters at the periphery of the co-disposal canister, together with 5 DHLW canisters. The round peripheral canisters have other variations of square and triangular shapes which affect capacity. The 17-inch central SNF canister has the advantage of minimizing the number of SNF cans to be handled, plus it affords a wider separation of SNF canisters for an array of co-disposal canisters for criticality control. This design is also a potential interim dry storage concept which would then greatly reduce handling. The 8-inch peripheral canisters have the advantage of better heat dissipation but suffers on the basis of increased handling. Study 2 re-analyzed the 17-inch diameter SNF canister based on the use of an internal basket with divider plates to help dissipate the heat [3.14].

Assuming the decay heat load of each of five DHLW glass canisters to be 460 watts and a repository temperature of 150°C, the maximum SNF temperature at the center of the co-disposal canister is 217.4°C [3.14]. The calculation used a 2D FIDAP model for 16 SNF fuel assemblies of 10 watts each inside the SNF canister (highest rated FRR fuel assembly with 10 years of cooling), with stainless steel divider plates in the co-disposal canister. An alternate analytical method based on homogenizing the configuration (which neglects radiative exchange with the divider plates) resulted in a maximum SNF temperature of 285.5°C. Thus with innovative canister design, the 17-inch diameter canister concept can be maintained as a viable design.

The present study will continue the previous studies focusing on the 17-inch and 8-inch diameter concepts. The objective will be to provide possible thermal environments for the fuel as a function of repository storage time. The heat transfer problem will be divided into two separate problems: 1) the internal temperature distribution inside the co-disposal canister based on an assumed canister exterior heat transfer coefficient and repository temperature, and 2) the exterior canister heat transfer. The interior heat transfer problem will be analyzed to determine the effects of

- structural design changes (to improve heat transfer),
- a range of fuel decay heats,
- changes in fuel geometry (due to effects of creep), and

- addition of depleted uranium or other neutron absorbing materials (which may be needed to resolve criticality issues).

The exterior canister heat transfer will be affected by the canister heat loading, canister emplacement design, repository ventilation, and canister location inside the repository, which at this time is not known. Consequently, the following parameters will be varied to determine the sensitivity of the spent fuel temperature on these factors:

- canister array heat loading and spacing,
- repository air flow,
- canister support design and use of backfill material,
- contribution of heat radiation on cooling of waste package.

The location inside the repository will determine if the effect of water evaporation and its transport inside the repository, which is an extremely important effect on the canister heat transfer, must be taken into account. This work will be integrated with ongoing work by other DOE agencies or contractors.

3.5 Criticality Analysis

The criticality safety analyses needed to support the direct disposal option for SNF assemblies in a long-term repository are being conducted in conjunction with the DOE-RW M&O. The scope of work, recently initiated, consists of two phases.

Phase I criticality analyses will evaluate the degree of subcriticality of 8 or 17 inch storage canisters filled with typical highly enriched, uranium-aluminum, plate-type, Material Test Reactor (MTR) fuel assemblies. These calculations will involve analyzing the reactivity of intact fuel assemblies stored inside these canisters assuming various water moderation and reflection conditions which are credible during long-term storage.

Phase II is a degraded mode analysis and will be a series of evaluations that models the degradation of the SNF and the internal structure of a breached waste package. That is, analysis will be performed to determine the degree of subcriticality of these same fuel assemblies assuming some degree of physical, chemical, and metallurgical decomposition and deterioration inside the storage canister. This phase will involve contributions from individuals in a number of disciplines to determine the most appropriate fuel degradation scenarios to consider, along with criticality evaluations of these scenarios.

The approach will be to assemble degradation modes for the fuel and identify worst cases of a redistributed material configuration (based on solution thermodynamics of aluminum, stainless steel, water, overpack materials system) under the appropriate moderator considerations and debris configurations within the canister and within the overpack and perform analysis thereof. The geochemistry code will predict compounds formed during the degradation. This will be done for several initial configurations, e.g. Al SNF in middle of HLW logs, Al SNF in four canisters within overpack with HLW logs, etc. Poisons or additions of depleted uranium or both would be considered in this activity if critical configurations are credible.

4.0 MELT-DILUTE PROGRAM

Major issues associated with the direct disposal of research reactor fuel in a repository include nonproliferation and criticality control, both of which may be a concern for HEU Al SNF. Consideration must be given to the potential desirability and/or regulatory necessity of diluting the HEU SNF to below 20% enrichment. The probability of criticality could be further lowered by reducing the enrichment. One method of achieving dilution is through a melt-dilution process. The Melt-Dilute (MD) technology program is focused on qualifying a diluted waste form for the repository. The technology program consists of the following key elements:

- **Melt-Dilution Process Development** - this task consists of:
 - (i) developing process flow sheets;
 - (ii) bench scale development including process parametric experiments;
 - (iii) evaluation of mold materials;
 - (iv) development of off gas system.

This task will culminate with the development of a full-scale Melt-Dilute system and demonstration of the Melt-Dilute process using full-scale surrogate MTR type elements. Sections 4.1, 4.2, 4.3, 4.4 and 4.5 provide the status of the process development efforts.

- **Waste form Development and Waste form Assessment:** The process development will be driven by the ability to produce waste form microstructures which would meet the repository waste acceptance criteria. Specifically, corrosion, leachability and durability test protocols will be used to guide the waste form qualification. Section 4.1.4 and 4.6 provide the status of these activities.

4.1 Melt-Dilute Technology

Development of a Melt-Dilute process and the waste form assessment for aluminum-base fuels is outlined in Technical Task Plan SRT-MTS-96-2063. Most of the early Foreign Research Reactor (FRR) fuels were made with an aluminum-uranium core containing 90-93 % enriched uranium-235. These fuel assemblies have been in water storage basins for up to 40 years.. The addition of depleted uranium to a molten alloy could reduce the ²³⁵U content to less than 20 % and greatly reduce proliferation concerns while lowering the need for engineered criticality controls. The fundamental melt-dilute process is shown in Figure 4.1.

As a first step in the process, the uranium content of incoming spent fuel is determined through characterization. From this data, the amount of depleted uranium needed to dilute the alloy is calculated. Finally, the spent fuel element is placed in a crucible and melted. Depleted uranium metal (or in oxide form) is added to the crucible, and in some cases, aluminum may also be added to adjust the composition to obtain specific concentrations of uranium in the alloy. Once diluted, the melt is sampled, and the sample analyzed, preferably in-situ, for uranium and/or ²³⁵U content. Once the desired composition has been obtained, the molten alloy is either solidified in the crucible or poured into a mold. The mold is sealed and is ready for dry storage. Eventually canisters containing the diluted fuel can be packaged and shipped to the geologic repository.

The melt-dilute technology is relatively simple and versatile process that can resolve both nonproliferation and criticality concerns for long term storage of HEU spent nuclear fuel. Potential options available for melt-dilute technology include:

1. Reduction in enrichment of the spent fuel from 93% ^{235}U to 20% or less which can reduce criticality concerns in the canister and the repository.
2. Reduction in volume of spent fuel resulting in reduction in the number of canisters needed for repository storage.
3. Minimal need for characterization vis-a-vis direct/co-disposal disposal technology,
4. Potential for Microstructural enhancements (i.e., intermetallic compounds of UAl_4 and/or UAl_3 and the eutectic composition) to improve long-term physical and chemical properties of the waste form.
5. Poison additions added directly to the alloy for enhanced criticality safety.
6. Potential to vary waste form geometry to utilize space and to address criticality issues within the canister.

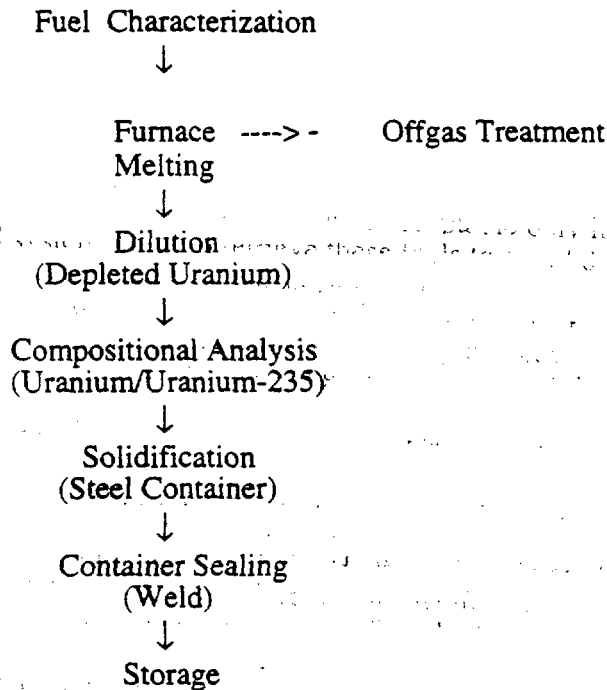


Figure 4.1 - Melt-Dilute Process Flow Diagram

4.1.1 Feed Material - MTR Fuel Assemblies

MTR type elements are representative of many research reactor fuel designs. These elements have typically up to 25 fuel plates which are clad with an aluminum alloy. Additionally, the elements have aluminum side plates with top and bottom fittings. Reactors may have variations of the basic design such as geometry and fuel loading specifications. Initially, development studies will utilize starting alloys of about 10 wt % uranium in aluminum which is the approximate composition of the melted fuel assembly. MTR assemblies are being fabricated to test the final concept and manufacturing technique. Assembly manufacturing will be discussed later.

4.1.2 Melt-Dilute Process Description

Melting the Fuel: Melting the fuel is the easy part of the process. A crucible that is slightly larger in diameter than the greatest dimension of the fuel cross section and high enough to contain all of the melted fuel plus the material needed to dilute the uranium is positioned in the hot zone of a furnace. Although the fuel may be over three feet long, the crucible will be less than one foot tall. The crucible is heated to a temperature above 800°C and the fuel melts, bottom first, into a pool of molten alloy. The rate of melting is a function of the furnace temperature or energy input.

Because alloy density varies with the uranium content, it is possible that the higher density material will be on the bottom of the pool with the less dense aluminum-uranium alloy on top. This occurs especially when the melt temperature is below the liquidus and the uranium metal dissolves slowly in the melt.

Stirring: When all of the fuel has melted, it will be necessary to stir the alloy to obtain a uniform mixture. If the alloy is above the liquidus temperature, the melt will be a homogenous solution. Stirring can be either mechanical or induction, but induction stirring involves no mechanical equipment, so it is easier to accomplish uniformity during solidification particularly in a closed system.

Dilution: The uranium in the solution contains a known high percentage of ^{235}U atoms relative to ^{238}U atoms. Dilution of the uranium with ^{238}U atoms is required to prevent the possibility of nuclear criticality during storage. This is accomplished by the addition of uranium that has been depleted of nearly all of its ^{235}U atoms. For the sake of this discussion, it is assumed that depleted uranium metal will be used for dilution. (The possibility of using depleted UO_3 will be discussed later.) Uranium metal does not melt at temperatures below $1,135^\circ\text{C}$ but will dissolve in aluminum alloy. The rate of dissolution depends on temperature and how rapid the alloy is stirred.

The uranium dissolution reaction results in a measurable alloy temperature increase. As the uranium content increases the alloy liquidus temperature increases, and if the alloy temperature is below the liquidus temperature, a mixture of solid UAl_3 and liquid alloy is present. The solids content increases as the uranium content of the liquid increases. As the per cent solid UAl_3 increases, it becomes more difficult to stir the mixture and dissolve more uranium metal. When the solid content of the mixture reaches approximately 50%, the mixture behaves as a solid and it is not possible to insert or remove a thermocouple probe from the alloy.

Although it is possible to dissolve uranium in alloy at a temperature well below the liquidus temperature, it may be desirable to bring each batch of alloy to a temperature above the liquidus temperature to assure a uniform solution before cooling to a solid. Additional tests will be done to resolve this issue.

If the amount of depleted uranium required to dilute the enriched uranium in the fuel results in an alloy with a high liquidus temperature, it is possible to add additional aluminum to lower the liquidus temperature. The cost of the resulting volume increase will have to be compared to the process difficulty of high temperature operation to determine if this option is worth consideration.

Processing evolved gas: Because it is likely that some radioactive gasses will be released during the fuel melting process, it is necessary to contain the system so the gasses can be collected and processed. This process is described in more detail in another section of the report. The need to process evolved gas has an impact on both the equipment design and the operating temperatures. Gas evolution is a function of melt temperature, so the amount of evolved gas can be minimized by operating at low temperatures. It may not be possible to operate the process at the temperature of the lowest gas evolution rate, but gas evolution will be considered when defining the process temperatures.

Because the melt crucible must be enclosed to capture evolved gas, special remotely operated equipment will have to be designed to perform operations inside of the

containment vessel, such as taking alloy samples, uranium addition and mechanical stirring. Although much of the early bench scale testing will be done with an open crucible, the process will have to be designed with an enclosed system in mind.

4.1.3 Analysis of Dilution of HEU Reactor Fuel Elements

The application of the melt-dilute technology for MTR spent fuel elements is shown in Table 4.1 for 20% dilution of several spent fuel elements. Initial data for these calculation were obtained from Argonne National Laboratory [4.1]. The calculations consider burnup and are based on the final ²³⁵U content of the fuel. The final ²³⁵U of the fuel is used so that the actual amount of depleted uranium can be determined and added to the melt for the desired dilution.

Table 4.1 -- Dilution Calculations for Several FRR Fuels

Calculations for the dilution with aluminum and depleted uranium

FRR Fuel Data		MTR	Dilution =		0.2		20 %		All weights are in grams									
Reactor	country	(%) initial enrich.	gm Int. U235 per FA	Int. U per FA	gm U235 Burned	U235 left per FA	gm KG U - Al per FA	initial Wt% U Alloy	final Wt% U Alloy	initial wt% U235U Alloy	Added Uranium gms	U Added dec3	Alloy Diluted U Wt%	Al Added gm	Al Added dec3	Weight Fraction U in Alloy	Dilution Factor	
Orpheus	France	93	840	903	252	588	9000	10.0	7.2	9.3	2280	0.12	28.4	0	0.00	0.284	0.20	
Orpheus	France	93	630	877	189	441	8000	8.5	6.1	7.9	1717	0.09	23.0	0	0.00	0.230	0.20	
HFR Patien	Netherlands	91	420	462	214.2	205.8	4800	10.0	5.4	9.1	782	0.04	19.5	0	0.00	0.195	0.20	
SAR-GRAZ	Austria	90	351	390	0.4	350.6	4500	8.7	8.7	7.8	1383	0.07	30.6	0	0.00	0.306	0.20	
Silow/Mel	France	93	340	366	153	187	5900	6.2	3.6	5.8	722	0.04	14.3	0	0.00	0.143	0.20	
Silovette	France	93	330	355	1.7	328.3	5900	6.0	6.0	5.8	1288	0.07	23.2	0	0.00	0.232	0.20	
JMTR	Japan	45	323	718	87.2	235.8	6000	12.0	10.5	5.4	548	0.03	22.4	0	0.00	0.224	0.20	
JMTRC	Japan	45	323	718	0.3	322.7	6000	12.0	12.0	5.4	896	0.05	28.1	0	0.00	0.281	0.20	
ASTRA	Austria	45	322	716	208.3	112.7	5300	13.5	9.6	6.1	57	0.00	13.1	0	0.00	0.131	0.20	
Saphr(ed)	Switzerland	45	320	711	211.2	108.8	4100	17.3	12.2	7.8	44	0.00	16.3	0	0.00	0.163	0.20	
Scarabee	France	93	307	330	3.1	303.9	5500	6.0	5.9	5.6	1192	0.06	23.0	0	0.00	0.230	0.20	
UTR300	UK	90	300	333	6	294	6500	5.1	5.0	4.6	1143	0.06	19.7	0	0.00	0.197	0.20	
HFR Patien	Netherlands	91	290	319	0.6	289.4	4500	7.1	7.1	6.4	1129	0.06	26.2	0	0.00	0.262	0.20	
JMTR	Japan	93	285	306	59.8	225.1	5500	5.6	4.5	5.2	679	0.06	17.9	0	0.00	0.179	0.20	
JMTRC	Japan	93	285	306	0.3	284.7	5500	5.6	5.6	5.2	1117	0.06	21.8	0	0.00	0.218	0.20	
Saphr(ed)	Switzerland	93	282	303	194.6	87.4	3900	7.8	2.8	7.2	328	0.02	10.5	0	0.00	0.105	0.20	
ASTRA	Austria	93	281	302	179.8	101.2	4900	6.2	2.5	5.7	384	0.02	9.7	0	0.00	0.097	0.20	

When the various fuel elements are melted, the composition of the melt is expected to vary between 2% and 12% uranium. The amount of depleted uranium added to the melt depends on the enrichment and burnup of each assembly. Adding depleted uranium produces an alloy composition typically between 9% and 30% uranium in aluminum for 20 % dilution of the ²³⁵U. The composition is within a range generally cast for nuclear reactor fuel and presents little challenge to manufacturing. Some of the spent fuel elements diluted to 20 % are in the range of the eutectic composition (14-15 wt % U). Segregation of the primary aluminides is not a concern at the eutectic because the liquid forms a solid at a constant temperature of 641 °C. However, at compositions higher than the eutectic, separation of the high density intermetallic compounds UAl₃/UAl₄ will occur from the liquid and cause alloy segregation.

When uranium metal is added at 850 °C and the composition is within the two phase region, the uranium dissolves slowly in the liquid. The uranium-235 composition of the primary intermetallic compounds will be diluted as precipitation occurs from the melt. On a microscopic scale, the ²³⁵U content across the dendritic arms may vary somewhat as the uranium concentration of the melt changes. This phenomenon is related to coring or compositional gradients found in dendrites of two component cast alloys.

No aluminum is added to the melt for the examples in Table 4.1. However, if the diluted composition is greater than the eutectic composition and if the specification requires an

alloy of the eutectic composition, then aluminum must be added to the melt. The addition of aluminum increases the volume of waste because aluminum adds about 370 cc/kg of aluminum whereas the addition of uranium adds only 50 cc/kg of uranium.

The enrichment of the alloy may be reduced further, on the other hand, by diluting to lower enrichment values below 20% uranium-235. This produces an alloy containing higher uranium concentrations. As the uranium content increases at 850°C, the per cent solid increases so that above approximately 50% solid, a continuous structure of solid material exists in the crucible. This phenomena reduces the effect of stirring of the melt. Further solidification of the liquid effects gravity segregation because the liquid will be trapped within the solid matrix, thus producing a more homogenous microstructure.

Theoretically, intermetallic compounds of either UAl_3 or UAl_4 can be made by continually adding uranium to the crucible. Formation of the solid phase, would eliminate segregation and produce an uniform microstructure of intermetallic compounds for the waste form.

4.1.4 Microstructure Development

The equilibrium phase diagram is used to predict phases present in cast alloys even though the casting process is non-equilibrium. The phase diagram for aluminum-uranium system is shown in Figure 4.2. A series of Al-U alloys with composition 10, 20, 30, and 70 wt % uranium were prepared for the purpose of studying the microstructure evolution. These samples were prepared by sequentially adding depleted uranium to the melt at 850 °C. Samples were taken from the melt and slow cooled. They were examined using the SEM. Instrumental analysis using Inductive Coupled Plasma Emission Spectroscopy (ICPES) was done to determine uranium content, and the results are given in Table 4.2 for each composition analyzed. The following discussion will demonstrate the flexibility of the process to tailor microstructures to maximize the performance of the waste form in the repository environment. The ensuing discussion will refer to the calculated composition with the measured composition in parentheses.

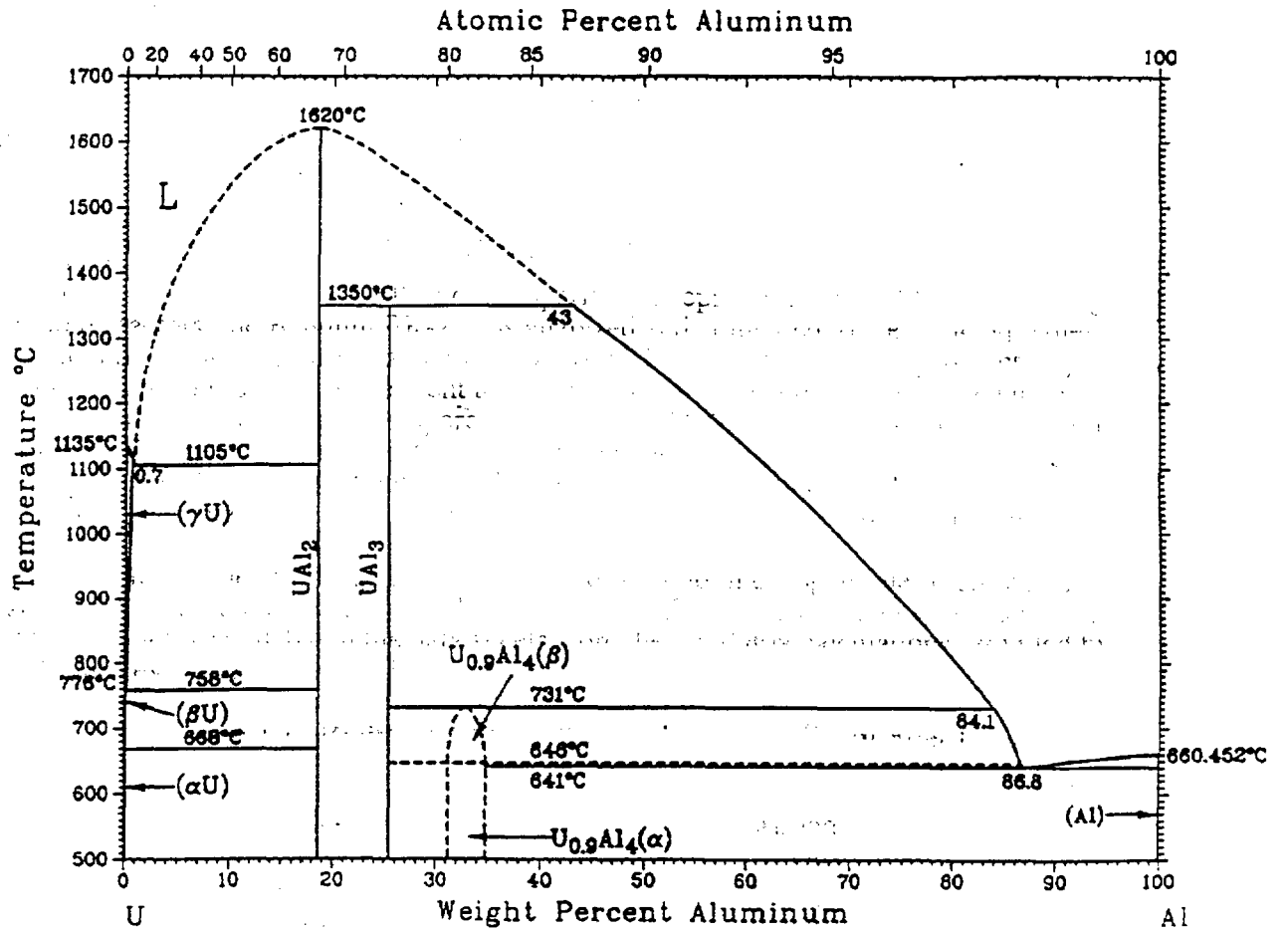


Figure 4.2 - Uranium Aluminum Phase Diagram

Table 4.2 - ICPEs Analyses of Test Samples

Sample wt % Uranium	Sample ID Number	Measured wt % Uranium
10	86178	10
20	86179	18
30	86180	26
70	86184	--

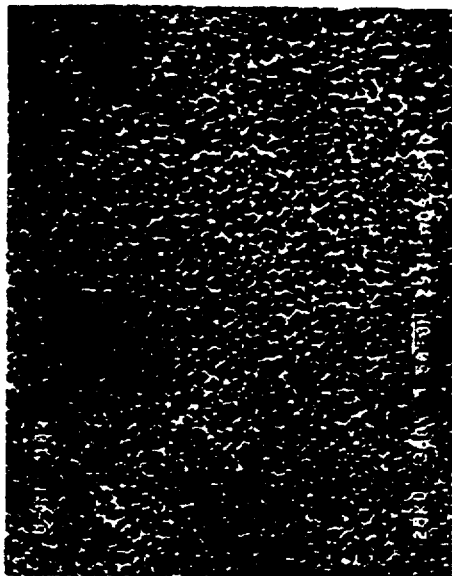
Photomicrographs of 10, 20, 30, and 70 wt% uranium in aluminum are shown in Figure 4.3. When a FRR fuel assembly is melted, the composition is about 10 wt % uranium in aluminum. A typical microstructure is shown in Figure 4.3a which consists of primary aluminum dendrites and the eutectic. The primary aluminum phase is uniformly distributed in this microstructure as indicated by the black dendritic arms in the figure. The eutectic is a lamellar structure of UAl_4 and aluminum.

The 20 wt % uranium alloy (18%) is shown in Figure 4.3b. At 850°C, only the liquid-phase is present. As the melt cools below about 800°C, primary UAl_4 precipitates from the liquid solution as the alloy enters the two-phase region of the phase diagram. At the eutectic temperature the remaining liquid, now at the eutectic composition, solidifies as the eutectic composition. The final microstructure consists of primary UAl_4 , a "diamond" shape phase, and the lamellar eutectic. Because of the higher density of UAl_4 , gravity segregation of primary UAl_4 can occur during solidification when the melt is not agitated or stirred.

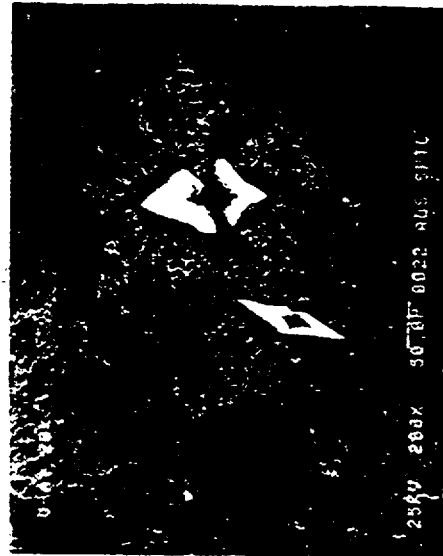
According to the phase diagram, alloys above 22 wt % uranium and at 850°C consist of solid and liquid phases. Uranium added to the alloy must first dissolve in the liquid since the melting point of uranium is much higher, 1135 °C. The dissolution kinetics is somewhat slow, requiring several minutes for completion. Growth and number of primary aluminides occurs as the uranium content increases from 30 to 70 wt %. The UAl_3 phase exhibits the characteristic "blocky" microstructure and can be easily distinguished from UAl_4 . For compositions greater than 22 wt % uranium, a homogenous liquid solution is obtained only by heating above the liquidus temperature. Microstructures of 30 and 70 wt % uranium-aluminum alloys are shown in Figures 4.3c and 4.3d, respectively.

Separation of the primary phases from the liquid during solidification causes uranium segregation in slow cooled alloys. However, stirring of the mixture during solidification prevents separation of the solid high density phase. Stirring is expected to be effective until about 50% solid is formed.

Generally intermetallic compounds show chemical and physical properties different from the base materials. Both UAl_3 and UAl_4 intermetallic compositions are being investigated to determine corrosion behavior and stability of these compounds for long term storage. The microstructure in Figure 4.3d has about 95% UAl_3 with remaining 5% consisting of aluminum and the eutectic. Screening test are being done to assess the corrosion behavior of cast 10, 30 and 74 wt % uranium microstructures and will be discussed in section 4.6



(a) Aluminum-10-wt% Uranium



(b) Aluminum-20-wt% Uranium



(c) Aluminum-30-wt% Uranium



(d) Aluminum-70-wt% Uranium

Figure 4.3 - Photomicrographs of Aluminum-Uranium Alloys

4.2. Process Development Program

The Melt Dilute process is a simple basic concept. An MTR fuel element will be positioned with its bottom end in a heated crucible. When heated, the bottom end of the element will melt and the fuel assembly will slowly slump into the pool of molten alloy. When completely melted, the uranium content of the alloy will be about 10 weight per cent uranium in aluminum. At this point, depleted uranium will be added to the alloy to dilute the percentage of ^{235}U . Dissolved fission gas will be released from the molten fuel and will be collected and treated as required to meet EPA standards. The alloy will be solidified and sealed in the container which will serve as a secondary container for the alloy.

Although the basic concept is simple, there are many technical issues that must be resolved before the system can be implemented. A series of experiments is in progress to define equipment, materials and procedural details to make the process a production operation.

4.2.1 Furnace design

The basic types of furnaces that will be evaluated in the experimental program include: 1) resistance heated, 2) induction heated and 3) vacuum, induction heated.

Resistance Heated Furnace: Resistance heated furnaces are relatively inexpensive and can be made to order and delivered within eight to ten weeks. They consist of high temperature heating elements contained in a "box" of thermal insulation. Temperature is measured by a thermocouple and controlled by a conventional control system. Although resistance furnaces can operate at any temperature required for the melt dilute process, the cost and size of the furnace is related to the maximum operating temperature.

The primary disadvantage of the resistance furnace for use in the melt dilute process is that any stirring of the molten alloy must be done mechanically. This is possible, but it adds to the complexity of the system. In addition, the crucible in a resistance furnace is heated primarily by thermal radiation, a process that is relatively slow and could be a potential problem for a process where time at temperature must be kept to a minimum.

Induction Furnace: Induction furnaces heat electrically conductive materials by means of a high frequency alternating magnetic field. The furnace itself is not heated directly. (The inside of the furnace is heated indirectly by radiation of heat from the hot crucible.) Because a high amount of energy can be converted to heat directly in the crucible, it is possible to increase temperature at a rapid rate in an induction furnace. In addition to heating, molten alloy can be stirred by induction, an attribute that makes the induction furnace attractive for the melt dilute process.

The high frequency generator and transformers required for induction melting and stirring make this equipment larger, more complex and more expensive than resistance furnaces. Delivery time is also likely to be longer for an induction furnace.

Vacuum Induction Furnace: A vacuum induction furnace is an induction furnace enclosed in a chamber capable of holding a vacuum. The chamber can be operated at a vacuum or back filled with an inert gas. Any dissolved gasses in the fuel that are melted will be contained in the chamber or can be directed to a treatment facility.

Vacuum induction furnaces are the most expensive option and have the longest delivery time because of the vacuum chamber and associated pumping equipment.

4.2.2 Selection of Crucible Material

The crucible material should 1) be electrically conductive if it is used in an induction furnace, 2) survive the process cycle with enough remaining wall thickness to serve as a container for the alloy, and 3) be relatively inexpensive.

Carbon Steel: Carbon steel is the material of choice at this time. Crucibles can be made by welding a plate on one end of a section of pipe. Schedule 40 pipe is inexpensive, and although there is some dissolution of iron by the molten alloy, it should be possible to complete the melt dilute cycle before more than half of the wall has been dissolved. If necessary, the wall thickness can be increased by using schedule 80 pipe. The carbon steel is quickly heated by induction if an induction furnace is used. After the heating cycle is complete, a lid can be welded onto the crucible.

Stainless Steel: Although stainless steel is known for its corrosion resistance, it will not survive exposure to molten aluminum for even a short time. Because of this problem, it can not be used as a crucible material in this process.

Graphite and/or Silicon Carbide: These are good materials that are often used as crucibles for induction melting of aluminum-uranium alloy in induction furnaces. If the melt-dilute process requires prolonged operation at temperatures above 1,200°C, it may be necessary to use one of these materials. They are not well suited for use as a final container, however, because it is not possible to seal weld the top directly to the crucible after the alloy has cooled. The high cost of these materials would not be a problem if the alloy were poured into a carbon steel container and the crucible used again to melt another fuel tube. However, it may be possible to solidify the molten alloy in this type crucible and then put the crucible and contents into a steel secondary container for encapsulation.

Other corrosion resistant alloys e.g. titanium alloys may also be considered if necessary.

4.2.3 Temperature Effects

There are at least six variable parameters that must be fixed to completely define the melt dilute process: melt temperature, crucible material, maximum wt% uranium in the alloy, acceptable gas evolution, crucible and final container material, and acceptable alloy volume. Each of these variables is related to some or all of the others. The table below shows the relation of each of these variables to the range of temperatures being considered.

Table 4.3 -- Temperature Effects

Melt Temperature °C	Crucible Material	Expected Maximum % U	Gas Evolution	Final Container	Additional Aluminum Required?
800	Carbon Steel	35 to 40	Low	Crucible	Likely
1000	Carbon Steel	50	Medium	Crucible	Perhaps
1,200	Carbon Steel Graphite SiC	60 to 70	High	Crucible or Mold	No
1,400	Graphite SiC	75	Highest	Mold	No

4.2.4 Bench Scale Experiments - Test Results

Tests have been conducted in a small resistance furnace and in an modified air melt induction furnace. The purpose of these tests was to develop an understanding of the behavior of alloys and crucible materials under conditions that simulate a broad range of possible melt-dilute process parameters. The results of these tests are the basis for some of the observations made in the preceding paragraphs. Many of the early test results are qualitative observations that will be quantified in future tests as the melt-dilute process parameters are better defined. Some of the significant results are summarized below:

Resistance Furnace Tests: Aluminum was melted in a crucible made from 3-inch carbon steel pipe. Uranium was added and the molten alloy was stirred with a 1/2-inch diameter carbon steel rod.

- This furnace required one hour to reach 800°C.
- At 800°C, the uranium required to increase the alloy content to 20% U was dissolved in about 10 minutes, but the uranium required to increase the concentration from 20% to 40% did not dissolve after ten minutes.
- The reaction between uranium and aluminum caused the crucible temperature to increase above the furnace temperature.
- When the furnace temperature was increased to 1,200°C to encourage reaction of the undissolved uranium, the crucible temperature increased to over 1,300°C and the crucible wall failed. There was a relatively uniform thinning of the crucible wall resulting from reaction of the alloy with the iron.

Induction Furnace Tests: Several tests have been run in the laboratory induction furnace using 6-inch diameter schedule 40 carbon steel pipe.

- The induction field couples with the carbon steel crucible, rapidly heating it to over 1,000°C. Although there may also be some induction heating of the aluminum in the crucible, the mechanism for melting the aluminum appears to be primarily conduction

and radiation from the hot crucible wall. The induction process heats the crucible and melts aluminum in much less time than required in the resistance furnace.

- The temperature increase resulting from reaction of uranium with aluminum that was observed in the resistance furnace test was also observed in all induction furnace tests.
- Induction stirring with 60 hertz resulted in good mixing of the molten pool. Induction stirring is based on the same principle used for magnetic pumping of liquid metals.
- With induction stirring at 850°C the uranium required to increase the alloy content to 10 wt % uranium will react completely in less than two minutes. At 1,000°C the alloy content was increased from 30 wt % to 50 wt % in less than five minutes.
- At 800°C, alloy that is composed of 50% solid and 50% liquid behaves as a solid. The surface can not be penetrated with an iron shaft, and rods that were immersed in the alloy at a higher temperature can not be moved.
- Although some loss of the carbon steel crucible wall thickness results from melting and alloying up to 60 wt % uranium alloy at temperatures below 1,200°C, the rate of loss is slow enough to conclude that the proposed melt dilute process can be completed before wall thickness loss is excessive.

4.2.5 Bench Scale Experiments - Path Forward

The initial tests yielded qualitative information about the behavior of the alloy under conditions that simulate potential melt dilute process conditions. Future tests will quantify many of these observations so it will be possible to specify actual operating conditions. Some of the tests already scheduled are outlined below:

Effect of aluminum temperature on stirring rate: When molten metal is stirred by induction, the alloy in the center of the crucible rises so the top surface forms a "crown". The height of this crown relative to the liquid level when it is not being stirred is a measure of the stirring rate. The stirring rate will be measured in pure aluminum at temperatures ranging from 850°C to 1200°C.

Effect of uranium content on stirring rate: The stirring rate will be measured at the same temperature for uranium alloy concentrations ranging from 10 wt % to 30 wt %.

Crucible wall thickness: Although the final process has not been defined, each future test will record the time at temperature history of the crucible wall so the rate of wall thickness loss can be better understood.

Source of depleted uranium: All of the tests to date have been done with depleted uranium metal. There is a large supply of excess depleted UO₃ on site which is available for use in this process if it could be made to react with aluminum to form uranium aluminum alloy. The reaction is theoretically possible, but little is known of the temperature at which it will take place or the efficiency of the reaction. Tests are planned to determine if UO₃ is a practical source of depleted uranium.

4.3 Melt-Dilute Process Demonstration

The bench scale experiments will guide the development of a full-scale melt-dilute furnace capable of melting a full-size MTR type element. It is expected that a furnace system will be developed and the melt-dilute process simplicity demonstrated using full size surrogate MTR type element during calendar 1997. Surrogate MTR type element will be fabricated to support the process demonstration.

4.3.1 MTR Fuel Assembly Development

Non-irradiated, surrogate MTR assemblies will be used to demonstrate the melt-dilute process. The assemblies will be fabricated at SRS by substituting depleted uranium for enriched uranium. Plate and assembly dimensions will remain the same as FRR fuel.

Powder metallurgy (PM) and casting are typical methods of producing compacts for MTR fuel plate manufacturing. After compaction, PM compacts are inserted into an aluminum frame with cover plates and hot rolled to completely clad the element. Since SRS is better equipped to extrude than to hot roll flat plates, extrusion techniques will be used to manufacture fuel plates containing cast UAl cores. The aluminum side plates and end fittings for the assembly will be made in the SRS machine shops.

MTR fuel plates have cladding thicknesses varying from 0.025 inch to 0.050 inch thick. This characteristic will be reproduced by coextruding an uranium-aluminum alloy billet with an aluminum sleeve around it. Proper die design will ensure that the aluminum cladding is uniform around the U-Al center core. The U-Al alloy will be cast with 20 wt % uranium, so the resulting composition of the melt will be about 10 wt % uranium in aluminum, the composition of a melted FRR assembly.

The uranium-aluminum alloy billets will be cast using air induction melting. Approximately eighty castings about 3 inches long are needed for 12 complete fuel elements. The billets will be cast as long as possible to reduce the total number of castings needed. Figure 4.4 gives the dimensions of a typical MTR fuel assembly.

4.4 Gas Release Analysis and Offgas System Development Status

The melting of reactor fuel and reactor core components and the release of volatile fission products from the molten fuel have been studied extensively because of their importance for severe nuclear reactor accidents. Some of these studies have analyzed the release of fission products from aluminum-clad UAl fuel, including the measurement of fission product release from irradiated fuel and unirradiated fuel using simulated radioactive elements. Of these studies, many were done on SRS uranium-aluminum alloy fuels similar to the FRR fuels which will be the feed material for the melt-dilute process. A literature review on the release of fission products from the melting of irradiated aluminum-clad, uranium-aluminum alloy nuclear fuel was initiated. This review is being conducted under Subtask 4.3 of the technical task and quality assurance plan.

The objectives of this review are to determine the radioactive species generated during the irradiation of aluminum-clad, uranium alloy research reactor fuels and identify the fission gases and products which would be released from spent nuclear fuel during the melt-dilute process. The information from this survey will provide baseline data for design of a furnace and off-gas system to process research reactor fuel before shipment to a repository.

In the melt-dilute concept, enough uranium would be added to dilute the uranium enrichment to the desired level. In doing this, the ideal melting point of diluted uranium-aluminum alloy would be near 850°C. Stirring constraints could drive this temperature up to 1050°C or higher. The concern in designing an off-gas system for use with the furnace would be in identifying the elements which would be released from the melt process and the ability to trap them. The review of the literature will concentrate on these areas.

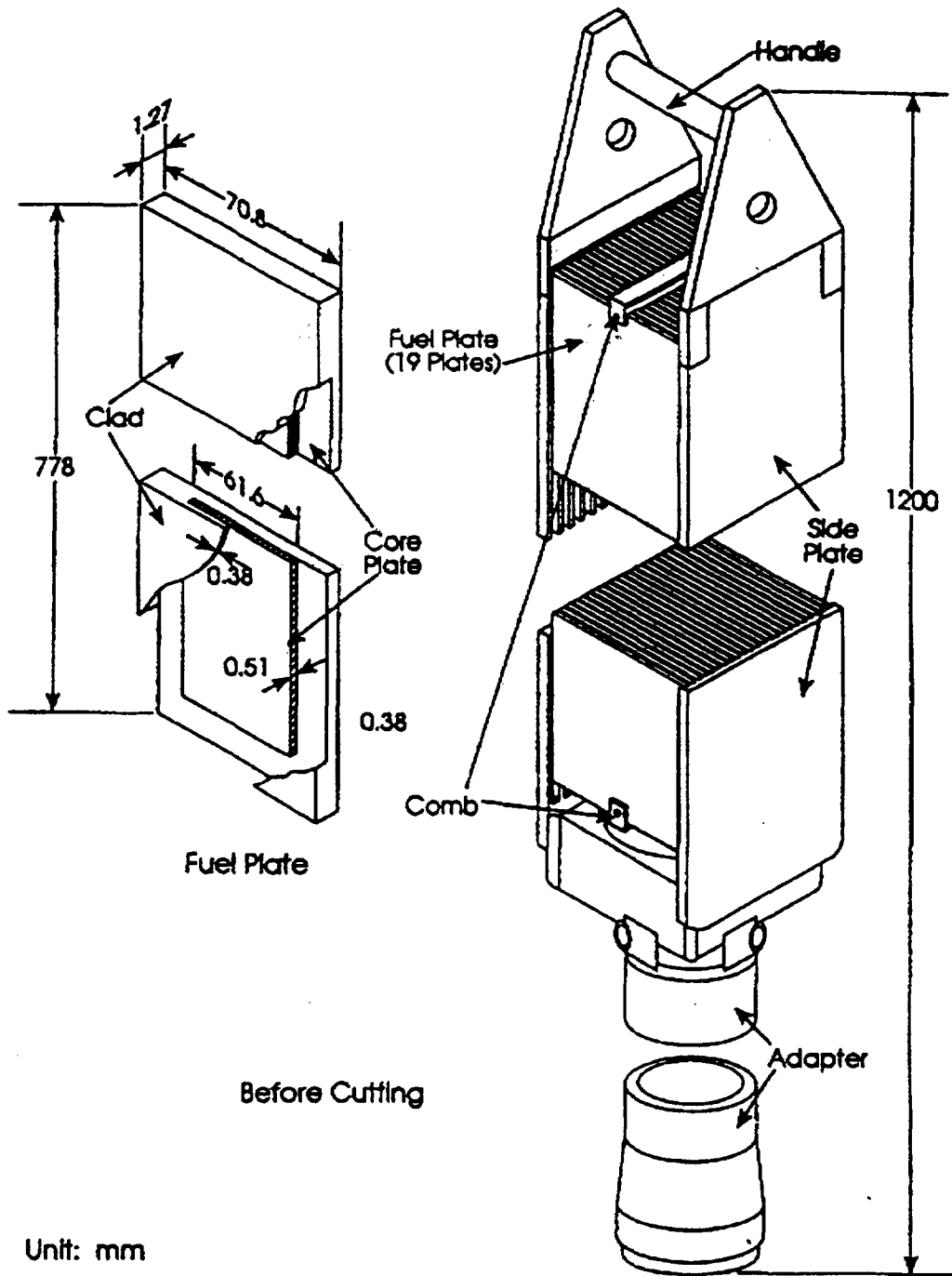


Figure 4.4 - Schematic Representation of Typical MTR Fuel Assembly

4.4.1 Determination of Radioactive Fission Products in Spent Fuel

Radioactive nuclides, including actinide isotopes and fission products, are generated in the fuel during reactor operation. These nuclides are contained in the fuel core, and further contained by the aluminum cladding. The ORIGEN (4.1) computer code, widely used within the DOE complex, can be used with the fuel history to calculate the expected inventory of radioisotopes.

The basis for this preliminary review is the ORIGEN code analysis of an ATR spent fuel element at 367.2 MW-days power. This SNF element is a bounding case for the FRR fuel and was used in the FRR EIS. For comparison, a representative fuel from the foreign research reactors, the Belgium BR-2, was analyzed and compared to the ATR type fuel on a nuclide by nuclide basis, each after 3 years of decay time. The total radioactivity content of an ATR spent fuel element after a 3 year decay period was calculated to be approximately 23,300 curries compared to about 11,000 for the BR-2 assembly, a factor of about 2X. The actinide content of the ATR element was found to be about 4X the activity of the BR-2, but the total assembly activity was dominated by the fission products. These were about 2X higher in the ATR fuel than in the BR-2 assembly.

A list of some of the most important radionuclides found in the spent fuel is shown in Table 4.4. A complete listing of all the actinides and fission products in spent fuel for the ATR type fuel is available in the data sheets from the ORIGEN calculations provided by INEL and reviewed by Bickford [4.2].

Table 4.4 - Radionuclides in Research Reactor Spent Fuel

H-3 (oxide)	Th-232
Kr-85	Pa-233
I-129	U-232
I-131	U-235
Co-60	U-236
Sr-90 (Y-90)	U-238
Zr-95	Np-237
Nb-95	Pu-236
Tc-99	Pu-238
Ru-106	Pu-239
Ag-110m	Pu-240
Sb-125	Pu-241
Cs-134	Pu-242
Cs-135	Am-241
Cs-137 (Ba-137m)	Am-243
Ce-144	Cm-242
Pm-147	Cm-243
Eu-154	Cm-244
Eu-155	Cf-252
Th-228	

Graphs showing the activity levels of these radionuclides in an ATR fuel assembly during up to 40 years of storage are seen in Figures 4.5.

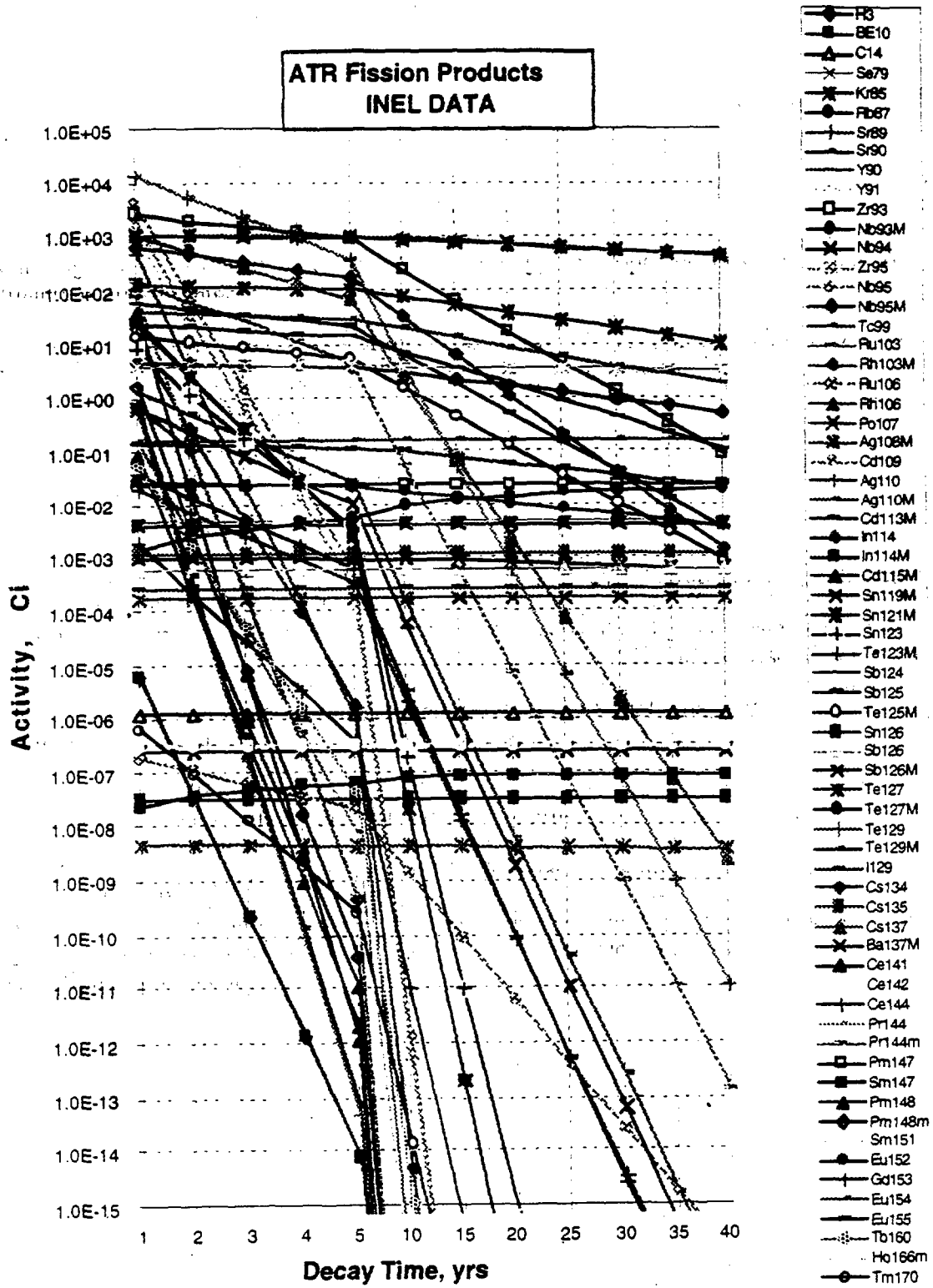


Figure 4.5 - Activity Levels for Radionuclides During 40 years of Storage

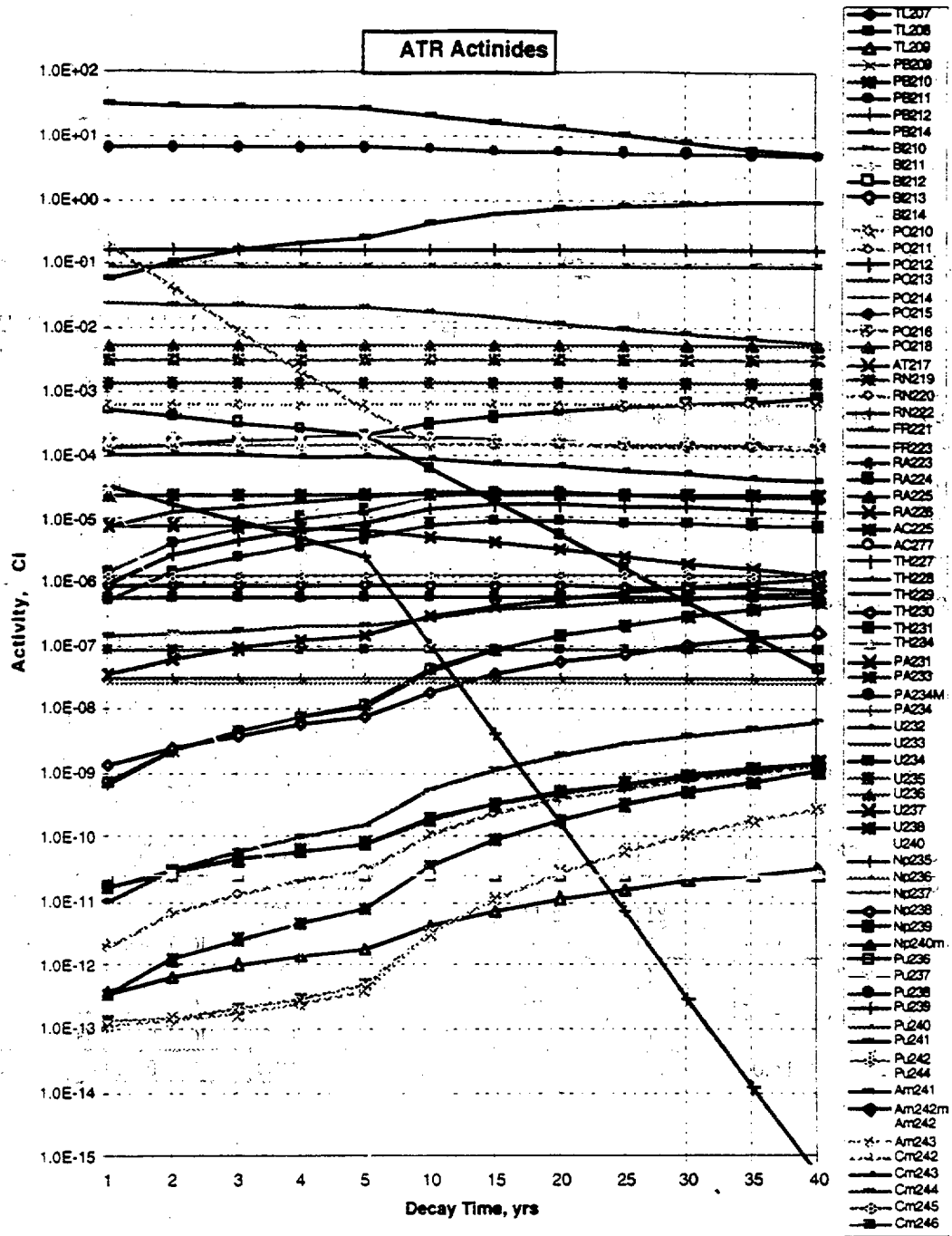


Figure 4.5 - Activity Levels for Radionuclides During 40 years of Storage

4.4.2 Fission Product Release During Melting

The release of radionuclides during reactor severe accident scenarios have been extensively described in the literature on the basis of both experiment and calculation. The published documents provide insights on the release of radionuclides from the spent nuclear fuel during the proposed melt-dilute process. Many of these studies were conducted by SRS under the Severe Reactor Accident Program and for the New Production Reactor at Savannah River (4.2).

The studies showed that the release of noble gases, and other fission products from the fuel matrix in an accident situation primarily occurred as the result of fuel melting. During fuel melting, all the noble gases are assumed to be released. The release of other fission products however is not as easily defined. Many of these fission products are only liberated from the molten fuel at extremely high temperatures, while others are retained in the fuel indefinitely.

A large number of radionuclides with widely varying release characteristics exist in the reactor core at the end of an operating cycle. These fission products may be grouped into classes that exhibit similar release characteristics. The thermophysical characteristic of the radionuclides that provides the greatest insight into fission product evolution from the molten fuel is the elemental boiling point.

Five classes can be used to group fission products with similar boiling points:

- Class A: Noble gases and tritium
Boiling point range < 27 °C (300 °K)
- Class B: Iodine, Cesium, and Rubidium
Boiling Point Range 27 °C to 927 °C (300 °K to 1200 °K)
- Class C: Tellurium
Boiling Point 927 °C (1200 °K)
- Class D: Strontium and Barium
Boiling Point Range 927 °C to 1727 °C (1200 °K to 2000 °K)
- Class E: All other radionuclides
Boiling point > 1727 °C (2000 °K)

The elements listed above will represent the source terms. The elemental boiling points are used as the delimiting characteristic for estimating the magnitude of release, because the primary release mechanism is vaporization of volatile species. In the metallic fuel, all these species are expected to exist in their elemental forms. Since the melting point for the UAl alloy fuel is quite low (approx. 900°K), the mobility of fission product vapors in the fuel above the melting point of the fuel is quite high. Therefore, very little time is required for fission products to be released from the molten fuel once the boiling point of the fission products is surpassed. The release mechanisms identified here are time and temperature, or rather, time-at-temperature. Once the boiling point of a species is surpassed, the species will begin to evolve from the molten pool. If the molten pool

remains at a given temperature for a long period of time, elements with boiling points below this temperature will eventually totally evolve.

Core meltdown calculations for the HWR-NPR indicated minimal release of radionuclides other than the noble gases from fuel during the assembly melting and in-tank relocation phase (15 minutes from the onset of melting). The entire inventory of noble gases is expected to be released. As the molten materials accumulate on the bottom of the reactor tank and heat up to higher temperatures, substantial quantities of fission products evolve. Some of the higher boiling point species in Class D are only released by entrainment in other isotope vapors that are more readily released. This is a minor effect.

4.4.3 Experimental Determination of Release of Fission Products from Irradiated Al Based Fuel at Elevated Temperatures

In an effort to upgrade SRS source terms for severe accident studies, fuel melting experiments were conducted under contract with Hanford Engineering Development Laboratory (HEDL) in 1986-1987 to determine the fission product release for melt temperatures of 700 °C, 1100 °C, 850 °C, and 1000 °C [4.3].

Many variables can affect the release of fission products from melted fuel. These variables include fuel type, fuel burn-up, exposed surface area, temperature, time at temperature and atmosphere. The HEDL experiments investigated the effect of these variables on the release of fission products from SRS fuel under fuel melting conditions.

Temperature and atmosphere were varied in the experiments. Fuel burn-up was fixed by the fuel assemblies at 52% with time at temperature set at 2 minutes. The fuel surface area was set by the apparatus at 0.5 inch round disks or 0.5 inch squares. The final melt temperatures were chosen to be 700 °C or 1100 °C. The lower temperature is slightly above the melting point of aluminum (660 °C) while the upper temperature is slightly below the melting point of uranium (1130 °C). Steam and air were chosen for the atmosphere as they would be present in the reactor room following fission product release. Air provides a chemically oxidizing atmosphere while steam produces a reducing atmosphere due to the generation of hydrogen from the metal/steam reaction. Argon was used for a comparison with an inert atmosphere.

Nine fission products were analyzed using gamma spectrometric methods or chemical separation techniques. These include -154, Ce-144, Sb-125, Ru/Rh-106, Cs-134, Cs-137, Zr-95, Kr-85 and I-129. Of the isotopes measured, only Cs-134/137, I-129 and Kr-85 were released in significant amounts. The other releases were less than 1 percent of the total release. The data for the release of Kr-85 shows that the quantity is likely to be total on core melting, irrespective of atmospheres. The U₃O₈/Al fuel was an exception. When melted in air at 700 °C less than 10% of Kr-85 was released.

The release of Kr-85 is total when the release of cesium exceeds about 15-20% cesium. Kr-85 was also found to be released in about 45 seconds of reaching the maximum temperature. For cesium, about 80% was released from the U-Al alloy heated in argon at 1100 °C. In steam, about 72% of the cesium inventory was released. Temperature was the most important variable, but atmosphere did have an effect. Cesium releases were higher in argon than in air or steam at the same temperature. In the 700 °C experiments, the bulk of the cesium remained in the fuel. Usually less than 30% was released. Most of the cesium plated out on the thermal gradient tube and the adjacent alumina furnace

tube as elemental cesium or CsOH. Of the 80% released, about 64% was gaseous and 16% was particulate.

Iodine release at a temperature of 1100°C for 2 minutes was nearly complete, irrespective of the atmosphere or the fuel type. CsI and elemental cesium were found deposited on the tubes. Evidence suggested that CsI production (as the result of the reaction of cesium with iodine vapor) may be significant. A complete release of iodine from the melt is possible when the temperatures reach 1100°C and remain there for two minutes.

Because of experimental limitations, not all of the elements releases were analyzed. Rubidium was not determined, yet it has chemical properties similar to cesium and its boiling point is only 30 degrees higher. Similarly, tellurium was not analyzed, but because of its relatively low boiling point, some tellurium releases may have occurred.

The tests showed that the oxide and metal alloy fuels behaved similarly with respect to fission product release. The fission product release was significantly lower from U₃O₈/Al than from U/Al up to 1000 °C. At this point, the oxide fuel release rises abruptly to a comparable level to the alloy fuel. This appears to coincide with the thermite reaction between aluminum and the oxide. The Iodine release data of Tests 1 and Test 2 compare favorably with results earlier reported by ORNL, with the exception of iodine release fractions in inert atmospheres. SRS show that iodine release in inert gas is the same as that in air or steam, while ORNL results show a significant variation. Both ORNL and SRS iodine release data for air, steam, and steam/air mixtures show nearly identical results. This comparison shows that iodine release fractions for U/Al fuel with greater than 23.6% burn-up are significantly affected by temperature and time, and to a lesser extent by atmosphere. The report says that both iodine and tellurium were released in significant quantities. Chemical forms of tellurium were not identified, while iodine was known to form CsI and I₂.

4.4.4 Additional Reviews

A large number of additional studies have been reviewed and will be detailed in a final report scheduled to be completed by May 1, 1997. Based on these reviews, a table of information containing elemental melting point, boiling point, half-lives, and vapor pressure has been formulated. Considering that all elements whose boiling points are about the melting point of the uranium-aluminum alloy fuel (850 °C-1050 °C) will likely vaporize and be released during melting, a preliminary list of elements is shown below.

Table 4.5 -- Elements with Boiling Points Below 1000 °C

<u>Element</u>	<u>Symbol</u>	<u>BP (°C)</u>
Tritium	H-3	-248
Krypton	Kr-85	-153.2
Xenon	Xe-133	-108.0
Iodine	I-129, 131	184.4
Cesium	Cs-134,135,137	671
Rubidium	Rb-137	688
Tellurium	Te-90	988

The literature will continued to be reviewed for information on radioactive release during melting of the uranium-aluminum alloy fuels and for techniques which can be applied to

the SRTC experimental program. The SRS Criticality Technology Group will provide fissile material burnup and depletion calculations, using the ORIGEN computer code for estimation of fission product inventories in spent Material Test Reactor (MTR) fuel assemblies.

The analyses will involve selection of several representative MTR fuel assembly types received (and expected to be received) at the Savannah River Site over the next 30 years. After selection of the candidate assemblies, Appendix A fuel composition and burnup record data on each of the fuels will allow reconstruction of the burnup history of the fuel assembly. With this input data, the ORIGEN computer code will be used to calculate the expected fission product inventories as a function of fuel U-235 content, burnup, and decay time. The results of these calculations will serve as design input to researchers developing a conceptual fission product recovery system for the Melt-Dilute process.

4.5 Fission Product Recovery and Off-Gas System Design

The requirements for an off gas system will depend on determination of the isotopes that can be released from the operation. Cs-137 will be the major concern, because of its longevity and high concentration in spent fuel. The requirements for some of the other isotopes, in particular Kr-85 and I-129, will be set mainly by regulatory requirements. In past operations at SRS it has not been necessary to retain Kr-85 because of its low radiation energy and minimal biological effect. Similarly, I-129 has not been a concern, because of its very low specific activity, although much of it was retained by offgas treatment systems. Retaining these isotopes could add substantially to the cost and complexity of the offgas treatment system. The need for retention of these radionuclides must be resolved with regulatory authorities. The retention of cesium and other particulate radioactivity requires efficient filtration. This is established technology at SRS and other nuclear sites. Both HEPA and sand filters have been used successfully for filtering particulate radioactivity. New technology, such as metal high efficiency filters, appears very promising for future use, as the metal filters can be cleaned and reused. Established technology for retaining iodine uses silver-coated solids or carbon beds to absorb iodine from the air. Both are effective for the low levels of radioiodine in long-cooled spent fuel. The krypton-85 released from irradiated fuel can be adsorbed using cooled zeolite beds. This technique was developed at SRS for possible application in reactor offgas systems. This is a more expensive offgas treatment process, but it should work very well at the low gas volumes associated with the Melt-Dilute process. The principal development activity associated with offgas treatment will be in developing and qualifying the forms and processes required for the ultimate disposition of these long-lived volatile isotopes. Several alternatives are available for each.

4.6 Waste Form Evaluation - Current Status and Path Forward

The waste form characteristics will guide the process development effort for the Melt-Dilute process. The test protocols for the waste form characterization and assessment is being developed as detailed in Section 6.0, however corrosion resistance measurements using electrochemical tests will be used for screening the characteristics of the melt-dilute alloys. The corrosion resistance will provide an indication of the long-term integrity of the waste forms. The program will consist of a series of accelerated electrochemical tests that yield data on the waste form stability in an aqueous environment, a general corrosion rate, and a measure of pitting susceptibility. The test technique, the test apparatus, analysis procedure and the test samples and environment will be discussed in this section.

4.6.1 Corrosion Test Technique and Apparatus

The study consists of a series of accelerated electrochemical tests and will provide data on the stability of the alloys in an aqueous environment, the general corrosion rate, and the susceptibilities of the alloys to pitting in this environment. Electrochemical testing is a standard approach used in the field to study material corrosion. Since corrosion involves electrochemical reactions, i.e. metal oxidation and reduction of aqueous species, these reactions can be understood by the current-potential relationships of metallic samples in the aqueous environment. The tests that will be used in this study involve applying a sequentially changing potential to a sample (polarizing) and measuring the responding current. The current-potential data, which is normally displayed graphically, is analyzed to establish the active corrosion phenomenon.

The electrochemical tests will be performed using a potentiostat controlled by a personal computer with commercially available software for test control and data acquisition and analysis. The potentiostat applies the potential to the sample and measures the responding current. The actual setup consists of an EG&G Princeton Applied Research (PAR) Model 273 potentiostat controlled by an IBM PS/2 Model 70 computer. The software is PAR Model 352 Softecorr®. The proper equipment functioning will be verified by conducting ASTM G5-87, "Standard Reference Test for Making Potentiostatic and Potentiodynamic Anodic Polarization Measurements." This reference is used for checking the technique and instrumentation with data obtained under standardized conditions against similar data obtained in other laboratories.

The U-Al or UAlx sample for these tests will be exposed to the test solution in a glass test cell that has several ports. The complete cell consists of a working, counter, and standard reference electrodes. The working electrode is the metal specimen which is placed in a teflon holder. The electrical contact is made at the back of the sample. The counter electrode, which must be inert to the solution, will be a pair of graphite rods. The standard reference electrode that is used to measure the sample potential will be a saturated calomel electrode (SCE). The potential of this reference electrode is 0.241 V versus a normal hydrogen electrode. To minimize the effect of solution resistance on the potential measurement the SCE will be in a Luggins capillary, which locates the measurement point near the sample surface. The Luggins capillary is a glass cylindrical container with a small frit end and holds the SCE in a conductive salt solution. The test cells and necessary electrodes are being obtained and prepared.

4.6.2 Corrosion Tests And Surface Analysis Procedures

A series of three different tests will be performed on each sample after being immersed into the test solution. These tests are corrosion potential monitoring, linear polarization resistance, and cyclic potentiodynamic polarization. Corrosion potential monitoring involves measuring the potential of the sample for 24 hours. The stable corrosion potential of the alloy will be determined from this data. This test will also verify that the sample has reached a steady state prior to the linear polarization resistance test. During this test the sample is not polarized. ASTM G69-81, "Standard Practice for Measurement of Corrosion Potentials of Aluminum Alloys" will be used as guidelines for the potential monitoring.

Linear polarization resistance consists of slowly ramping the sample potential over a small range, ± 0.02 V versus the corrosion potential. The scan rate will be 0.2 mV/sec. ASTM G59-91, "Conducting Potentiodynamic Polarization Resistance Measurements," will be followed. Over this small range, the potential and current approach a linear relationship, which is referred to as the polarization resistance, R_p . This value can be used to calculate a general corrosion rate. At the conclusion of this test, the sample will be allowed to restabilize prior to performing cyclic potentiodynamic polarization.

Cyclic polarization consists of ramping the sample potential over a large potential range. The test will be initiated at a potential -0.050 V versus the corrosion potential up to a vertex potential of 0.800 V versus the SCE potential, and reverse scanned to the initial corrosion potential. The scan rate will be 0.5 mV/sec. ASTM G61-86, "Conducting Cyclic Potentiodynamic Polarization Measurements for Localized Corrosion Susceptibility of Iron-, Nickel-, or Cobalt-Based Alloys," will be used as guidelines for this test. Pitting susceptibility is determined by the presence of a pitting potential and a hysteresis in the potential-current graph. The pitting potential, if it exists, will be chosen where a significant increase in current occurs on the forward scan. The closer this value is to the corrosion potential the more susceptible the material is to pitting. The presence of a hysteresis also indicates the material is susceptible to pitting. A hysteresis occurs when the current on the reverse scan is larger than that on the forward scan. A greater amount of pitting generally produces a larger hysteresis.

After each test, the samples will be evaluated to characterize the degradation of the surfaces including the pit population and depth, microstructural features, and preferential corrosion sites. Light and electron microscopy will be used for these evaluations. The samples will be resurfaced to similar initial conditions and the series of tests repeated.

4.6.3 Test Materials and Test Environment

Several U-Al alloys are being investigated as compositions for the final waste form. These alloys contain 10, 30, and 50 wt % uranium and also include several intermetallic compositions. The samples for the corrosion study were made during the bench scale melt-dilute process development. Two heats of the 10 and 30 wt % U alloys were made, then either slow cooled or quenched. The 50 wt % U-Al alloy was slow cooled so that the UAl_3 would form and precipitate out of the bulk melt. All the cylindrical ingots were 3.5-4 inch lengths and 5/8-inch diameter, which was the diameter necessary to fit the sample holder. Disc shaped samples, 0.004 inch thickness, were cut from each cylinder. For the 10 and 30 wt % U-Al alloys, discs were taken from two locations, the bottom of the poured cylinder and approximately 1 inch from the top. The middle piece was reserved for possible future use. The discs from the 50 wt % U-Al alloys were cut only from the bottom of the cylinder. Each disc was numbered uniquely. For the 10 and 30 wt % U-Al alloys, two disc from each melt will be tested, one each from the bottom and top.

The discs will be placed in epoxy mounts for ease of handling and test set-up. The mounting procedure is currently being developed. The process will involve attaching a metal bolt to the back of each disc using either a conductive epoxy or paint. The disc and bolt will be encased in a nonconductive epoxy following standard metallographic procedure. The end of the bolt will be at one end of the mount for electrical contact to the sample and the disc will be exposed at the other end of the mount for exposure to the test solution. The disc will be ground with 800 grit paper prior to immersion in the test solution.

The representative ground water chemistry for the repository, J-13 well water, was chosen for the test solution. The composition of the test solution is shown below. Trial batches will be prepared and analyzed prior to making those necessary for corrosion testing.

Table 4.6 - Component Concentration of J-13 Well Water

<u>Component</u>	<u>Concentration (mg/L)</u>
Ca	13
K	5.3
Mg	1.9
Na	44
Si	33
Cl	7.1
F	2.3
NO ₃	8.1
SO ₄	18
HCO ₃	120
pH	8.5

5.0 SNF CHARACTERIZATION PROGRAM

The characterization program is currently focused on the Direct/Co-Disposal approach for aluminum SNF disposition, with additional support anticipated for alternate disposition technology development such as melt-dilution. This task will establish and/or determine the properties of aluminum SNF necessary to meet all applicable requirements for the receipt, handling, conditioning, packaging, interim dry storage, transportation, and final disposition of aluminum SNF in the geological repository. This program was initiated in January '97 and the current status is provided herein.

Section 5.1 defines the characterization task and Section 5.2 identifies the Technical Approach in the context of the ATTP program. Section 5.3 provides the status of the Al SNF characterization database development and Section 5.4 provides the status of the review of Al SNF characterization requirements.

5.1 Task Definition

The characterization task is currently limited to the determination of the inherent properties of the fuel, rather than behavioral characteristics such as corrosion resistance or radionuclide release mechanisms, which are dependent upon environmental conditions. Determination of these characteristics or responses are to be determined under the separate but related Test Protocol task for final waste form qualification. Properties of aluminum SNF to be determined and/or established under the characterization program are expected to include but may not be limited to: fuel condition upon receipt, cladding/fuel alloy compositions and microstructures, burnup/fissile material content, isotopic inventory, density, cladding surface/oxide characteristics (thickness, microstructure, composition, porosity, etc.), heat load, ignition/burning temperature, moisture content, hydride content, cladding integrity, etc.

Inherent fuel properties will be determined through either review and assimilation of existing data or through experimental activities to establish a characterization baseline or database. The fuel properties necessary to meet the applicable requirements of the disposition process will then be determined upon receipt and incorporated into the database which can then be used to facilitate the evaluation of the behavioral characteristics or responses of the final waste form such as corrosion resistance and radionuclide release under varying environmental conditions. Fuel or waste form response to environmental degradation can thus be determined and correlated as a function of these properties.

5.2 Technical Approach

As outlined in the attached Task Technical Plan [5.1] the strategy of the characterization program is currently divided into two separate but related tasks: (i) characterization data requirements; and (ii) review of characterization technology requirements.

The objectives of the first task (Task 1.0 of the TTP) are to review existing regulatory documents governing all pertinent aspects of Al SNF, to determine and/or establish the applicable characterization requirements for the disposition process, to review and assimilate existing characterization data necessary to meet the outlined requirements, and to identify and determine through experimental activities those properties needed that are currently unknown or insufficiently characterized. This objective will involve the review of existing literature, regulatory documents, experimental data, and fuel history, as well

as the monitoring and coordination of experimental activities to be performed at the Argonne National Laboratory (ANL), SRS, Pacific Northwest National Laboratory (PNNL), or other institutions as necessary.

The objectives of the second task (Task 2.0) are to review existing technologies applicable for the characterization of aluminum SNF, to screen and identify technologies and techniques most applicable based on criteria yet to be determined, and to recommend characterization technologies, techniques, and equipment for the disposition process. For those characterization data requirements established under the first objective for which current acquisition technologies are either inadequate or non-existent, characterization technology needs will also be identified. Technology development activities will also be initiated as appropriate.

The primary year-end deliverables of this program are: 1) Preliminary characterization data needs and technology requirements, and 2) Topical Report on the SNF Characterization Database. All of the activities outlined above are to be performed as input for these two deliverables.

5.3 SNF Characterization Database Development

5.3.1 Al SNF Inventory

Based on the DOE-Owned Spent Nuclear Fuel Technology Integration Plan published by the Environmental Management (EM) Spent Fuel Management Office in May 1996, the inventory of DOE-owned SNF projected to 2035 was tabulated [5.1]. Data was obtained from the Integrated Spent Nuclear Fuel Database System (ISNFDS) release of September 1995. The inventory is currently separated into 55 sections according to five main characteristics of fuel materials: enrichment (high or low), fuel type, matrix material, cladding, and burnup.

Only aluminum SNF will be received at SRS for disposition, with non-aluminum fuels to be transported to the INEL (Idaho National Engineering Laboratory) for disposition. Existing defense production SNF currently at the Hanford Site will remain at Hanford under the current program. Based on the ISNFDS data, a listing of the aluminum fuels to be received at SRS are given in Table 5.1 on the following pages for reference. The primary focus of the characterization task and database is on the determination of interest materials properties and characteristics of the aluminum fuels necessary to meet the disposition requirements.

Table 5.1 - Aluminum SNF Inventory Projected to 2035

Site	Area/Facility	Fuel Name	Mass (kg)	Vol. (m3)	No.items	U. Mass (kg)	Enrich.	Fiss.Mass (kg)
HEU, Uranium Oxide Fuel, Al-Matrix, Al-Clad, High Burnup								
FRR	FRR	FRR/East	694	8	6720	556.01	48%	268.8
Non-DOE	NBSR	NBSR	3848.8	9.995	880	117.18	81%	94.9
ORNL	7900 Pools	HFIR	22663.8	19.261	162	1635.68	93%	1523.3
SRS	H-Area/RBOF	Sterling	741	3.705	226	102.08	93%	94.6
HEU, U-Alx fuel, Al-Matrix, Al-Clad, Low Burnup								
ANL-E	Chicago Pile 5	CP-5	2	0.02	2	1.23	93%	1.1
BNL	BNL-BLDG. 750	HFBR	28645.1	23.64	6537	1673.47	77%	1287.1
BNL	BNL-BLDG.491	BMRR	304.1	0.247	68	6.44	84%	5.4
FRR	FRR	FRR West	27136	31	5080	4221.48	20	828
INEL	ICPP-603	ORR	85	0.067	17	3.25	80%	2.6
INEL	ICPP-666	ARMF	3.6	0.019	15	0.2	90%	0.2
INEL	ICPP-666	Univ.of Wash.	71.2	0.078	26	3.9	93%	3.6
INEL	TRA-660	MARK I	314.7	0.379	56	11.19	93%	10.4
INEL	TRA-660	MARK I LL	9	0.014	2	0.24	93%	0.2
INEL	TRA-660	MARK II	37.2	0.054	8	1.16	81%	0.9
INEL	TRA-660	MARK III	2.6	0.003	4	0.1	92%	0.1
INEL	TRA-660	Misc.	5	0.001	1	1.5	93%	1.4
LANL	TA3-BLDG.29	Omega West	507.4	0.602	86	14.88	86%	12.8
Non-DOE	GE-Pleasanton	GENTR	80	0.05	32	8	93%	7.4
ORNL	ORNL-BSR	BSR Reactor	184.5	0.214	41	7	86%	6
ORNL	ORNL-TSF	TSR	182	0.1	1	9.2	93%	8.6
Sandia	KAFB/SNM	SPR II	20	0.1	1	12	83%	10
Sandia	Tech.Area V.	SPR I,II,III	60	0.1	1	31	97%	30
Sandia	Tech.Area V.	SPR III	1080	1.8	18	227	97%	220
SRS	H-Area/RBOF	ATSR	20	0.135	4	3.22	93%	3
SRS	H-Area/RBOF	JMTR	200	0.506	15	16.73	88%	14.8
SRS	H-Area/RBOF	ORR	34.8	0.067	2	5.46	11%	0.6
SRS	H-Area/RBOF	ORR	243.3	0.472	14	90	16%	14.4
SRS	H-Area/RBOF	ORR	999.6	1	17	20.51	82%	16.9

Table 5.1 (continued) - Aluminum SNF Inventory Projected to 2035

Site	Area/Facility	Fuel Name	Mass (kg)	Vol. (m3)	No.items	U. Mass (kg)	Enrich.	Fiss.Mass (kg)
HEU, U-Alx Fuel, Al-Matrix, Al-Clad, High Burnup								
FRR	FRR	FRR/East	83934.1	100	14484	12123.1	22%	2679.5
INEL	ICPP-603	ATR	1102.1	1.129	128	99.2	79%	79.1
INEL	ICPP-603	HFBR	87.6	0.074	20	4.9	78%	3.8
INEL	ICPP-603	MURR	153.1	0.559	24	16.1	89%	14.3
INEL	ICPP-666	ATR	6956.9	7.129	808	674.7	82%	558.1
INEL	ICPP-666	HFBR	964	0.817	220	58.5	79%	46.5
INEL	ICPP-666	MURR	204.2	0.745	32	21.73	87%	19
INEL	TRA-670	ATR	25520	26.152	2964	2711.2	87%	2353.1
SRS	A/SRTC	SRS Driver	5.3	0.005	4	0.83	31%	0.3
SRS	H-Area/Canyon	SRS Driver	430	0.42	13	67.6	61%	41.5
SRS	H-Area/RBOF	ANLJ	30	0.135	4	2.79	93%	2.6
SRS	H-Area/RBOF	Foreign/Relief	341.5	0.067	153	18.66	73%	13.6
SRS	H-Area/RBOF	MIT	200	0.12	16	15.93	81%	12.9
SRS	H-Area/RBOF	MURR	3900	1.754	52	70.18	87%	61.2
SRS	H-Area/RBOF	Ohio State	200	0.3	4	3.28	94%	3.1
SRS	H-Area/RBOF	RHF	408	0.502	4	25.51	81%	20.8
SRS	H-Area/RBOF	RINC	65	0.438	13	8.51	90%	7.7
SRS	H-Area/RBOF	SRS	50	0.1	5	6.12	32%	1.9
SRS	H-Area/RBOF	Sterling	97.9	0.8	34	24.6	85%	20.9
SRS	H-Area/RBOF	Univ. of Va	160	0.27	8	6.86	88%	6.1
SRS	K-Basin	SRS Driver	20400	29.144	902	3237	65%	2113.5
SRS	L-Basin	SRS Driver	17250	16.672	516	2593	60%	1546
SRS	P-basin	SRS Driver	10700	14.507	449	1391.2	66%	921.3
Univ.	GA TECH	GA TECH	290	0.314	27	5	90%	4.5
Univ.	IOWA STATE	IOWA STATE	99.4	0.099	14	4.1	95%	3.9
Univ.	MIT	MIT	1884	1.172	471	211.95	89%	187.7
Univ.	PURDUE	PURDUE	186.	0.574	124	2.19	91%	2
Univ.	UNIV. OF FLA	UNIV. OF FLA	313	0.655	256	4	92%	3.7
Univ.	U.Mass-Lowell	U.Mass-Lowell	230	0.153	26	4.3	93%	4
Univ.	U. of Missouri	MURR-Col.	7551.6	6.494	1218	916.2	90%	828.2
Univ.	U. of Missouri	MURR-Rolla	173.6	0.161	28	4.8	90%	4.3

Table 5.1 (continued) - Aluminum SNF Inventory Projected to 2035

Site	Area/Facility	Fuel Name	Mass (kg)	Vol. (m3)	No.items	U. Mass (kg)	Enrich.	Fiss.Mass (kg)
LEU, Uranium Oxide Fuel, Al-Matrix, Al-Clad								
Univ.	U. of Missouri	MURR-Rolla	173.6	0.161	28	26.4	20%	5.2
LEU, U-Al Fuel, Al-Matrix, Al-Clad								
SRS	H-Area/RBOF	NEREIDE	500	0.042	8	35.42	20%	7
SRS	H-Area/RBOF	U. of Michigan	300	0.27	8	33.85	15%	5.1
SRS	K/L/P-Basins	K/L/P Non-U	2478.4	3.36	104	0.04	0%	4.2
UNIV.	Iowa St.	Argonaut	169	0.092	13	20.2	20%	4
UNIV.	Ohio State	Ohio State-LEU	200	2.136	414	26.1	20%	5.1
UNIV.	Rhode Island	RINSC	979	1.037	178	246.23	19%	47.5
UNIV.	U. of Michigan	U. Of Michigan	598	0.745	131	98.5	17%	16.6
UNIV.	U. of Virginia	U. Of Virginia	261	0.265	45	56.43	20%	11
UNIV.	Technology	Polytech	156.6	0.165	27	22.73	20%	4.5
UNIV. LEU, Uranium-Silicon Fuel, Aluminum Cermets, AL-Clad								
Non-DOE	Hot Cell (GA)	GA-RERTR	14.9	0.023	2	3.04	12%	0.4
ORNL	ORNL-BSR	ORR	142.7	0.185	32	52	20%	10.3
UNIV.	GA TECH	MTR-SI	1428.4	1.545	133	151.1	20%	29.9
UNIV.	PURDUE	MTR-SI	35	0.102	22	25	20%	5
UNIV.	U. Of Florida	MTR-SI	137.5	0.064	25	28.4	20%	7.4
UNIV.	U. Mass-Lowell	U.Mass-MTR	248	0.165	28	37.5	20%	7.4
UNIV.	U. of Michigan	U. of Michigan	3031	3.14	552	593	20%	117.5

5.3.2 Database Structure

As currently defined, characterization of aluminum SNF shall be limited to those activities necessary to establish the inherent material properties of the fuel necessary to meet the characterization requirements interpreted from review of existing regulatory documents and drivers. As requirements for repository acceptance of the final waste form are developed and established by the OCRWM, the scope of characterization activities may increase or decrease as necessary. For each fuel type to be received and dispositioned, a minimum degree of characterization is anticipated as necessary, depending upon the fuel condition, final waste form, and applicable requirements. Because the operation history and storage conditions between participating facilities and countries is known to vary, some degree of characterization of fuels from each point of origin is expected to be required. For highly degraded or "failed" fuels, or those lacking in adequate documentation, a greater degree of characterization may be necessary.

A preliminary outline of the characterization database format for a given fuel type is shown in Table 5.2. The database is expected to contain several individual but related sections for a given fuel type, with fuel information and material property data falling into one of the following four categories: Receipt Inspection/Operation History, Visual Examination/NDE&A, Destructive Evaluation, and Computational Analysis. This database of material properties is not considered all-inclusive at this point and is subject to change as the disposition requirements are established.

Table 5.2 - General SNF Characterization Database Structure

Aluminum SNF Characterization Database			
Receipt Inspection/ Operation History	Visual Examination/ NDE&A	Destructive Analysis	Computational Analysis
QA/Product Verification	Fuel Condition (high-resolution macrophotography)	Cladding/Fuel Composition (SEM/EDX)	Criticality/Fissile Content
Initial Enrichment	"Failed Fuel Criteria"	Cladding and Fuel Microstructure (metallography)	Heat Transfer/Heat Load
Fuel Burnup History	Fuel Integrity (ultrasonics, radiography)	Oxide Characteristics	Isotopic Inventory/ORIGEN
Dimensional Measurement	Burnup (Gamma Scan/Spectroscopy)	Radionuclide Inventory	
Nuclide Inventory	Cladding Breaches (SIP test)	Hydride Content (metallography)	
	Radioactivity (Scintillation)	Heat Load (Calorimetry)	
	FORK/neutron detection	Ignition Temperature	

5.3.3 SNF Material Properties

The majority of DOE-owned fuel materials to be received and dispositioned under the Direct/Co-Disposal approach are of the MTR (Materials Test Reactor) design, constructed of aluminum-uranium alloy core materials with aluminum alloy cladding. A small percentage of fuel elements were fabricated from aluminum-uranium silicide alloys or aluminum-uranium oxide alloys. Cladding alloys are generally either alloy 1100, 5052, 6061, or their foreign equivalents. The fuel materials are either classified as highly enriched uranium (HEU) fuels, with ²³⁵U content up to 90%, and low enriched uranium (LEU) fuels, with ²³⁵U between 2-20% of the total uranium content. Cladding alloys are used primarily for containment of radionuclides and fission products, but also provide corrosion protection, reaction moderation, radiation shielding, and structural integrity.

The nominal elemental composition and mechanical properties of the aluminum alloys used for cladding are given in Table 5.3 [5.2]. Although the determination of corrosion resistance of the cladding alloys is not considered part of the characterization program, it should be pointed out that the corrosion resistance of the fuel core materials is considered to be nearly equivalent to that of the cladding materials, with the primary differences observed attributed to cladding alloy composition, oxide formation, and the effects of radiation exposure (measured as a function of burnup).

Table 5.3 - Nominal Elemental Compositions for SNF Cladding Alloys

Alloy	Nominal Elemental Composition (Wt%)				
1100	0.12 Cu	bal. aluminum			
5052	2.5 Mg	0.25 Cr	bal. aluminum		
6061	0.6 Si	0.28 Mn	1.0 Mg	0.20 Cr	bal. aluminum

Table 5.4 - Typical Mechanical Properties of Cladding Alloys

Alloy/Temper	Typical Mechanical Properties (Room Temperature)			
	T.S. (ksi)	Y.S. (ksi)	Hardness	Elongation (%)
1100/0	90 MPa (13)	35 MPa (5.1)	23 BHN	35
5052/0	195 MPa (28)	90 MPa (13)	47 BHN	25
6061/0	125 MPa (18.1)	55 MPa (8.0)	30 BHN	25

0 - annealed, recrystallized

Oxide Film Characterization

Passive aluminum oxide films form on the outer surface of the cladding alloys due to oxidation and radiation-induced growth during fission processes and at elevated temperatures. The thickness of these films can vary and have been observed up to 50 μm . The composition varies depending upon service/storage conditions and burnup history, but the oxide is generally of the more protective form Boehmite ($\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$), with the Bayerite form ($\text{Al}_2\text{O}_3 \cdot 3\text{H}_2\text{O}$) present to a lesser degree. A representative photomicrograph of aluminum oxide formation is shown in Figure 5.1 below.

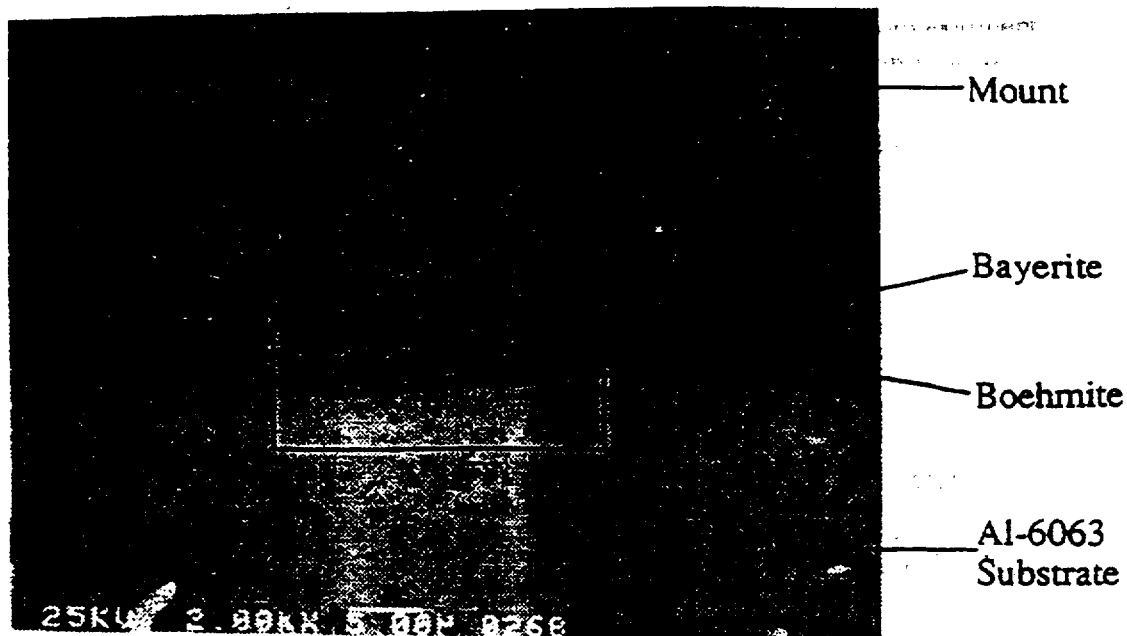


Figure 5.1 - Microstructure of aluminum oxide film on surface of tube specimen of aluminum alloy exposed to hot water

Fuel Meat/Core Compositions and Microstructures

Most of the DOE-owned aluminum spent nuclear fuels consist of a core or meat composition clad with the one of the alloys previously discussed. The actual fuel compositions mostly consist of aluminum-uranium alloys, with the uranium dispersed in the form of uranium aluminides throughout an aluminum matrix. The uranium composition may range from that of depleted uranium to highly enriched uranium or HEU (^{235}U content greater than 20% up to 90%). Enriched fuels of ^{235}U content between 2 and 20% are considered to be low enriched uranium (LEU) fuels. A smaller fraction of the fuels are constructed of aluminum-uranium silicide or aluminum-uranium oxide alloys in which the uranium is in the form of U-Si_x or UO_x particles, respectively. The typical microstructures of highly-enriched aluminum-uranium alloys are shown in Figure 5.2 below, with aluminum-uranium silicide and aluminum-uranium oxide microstructures shown in Figures 5.3 & 5.4, respectively.

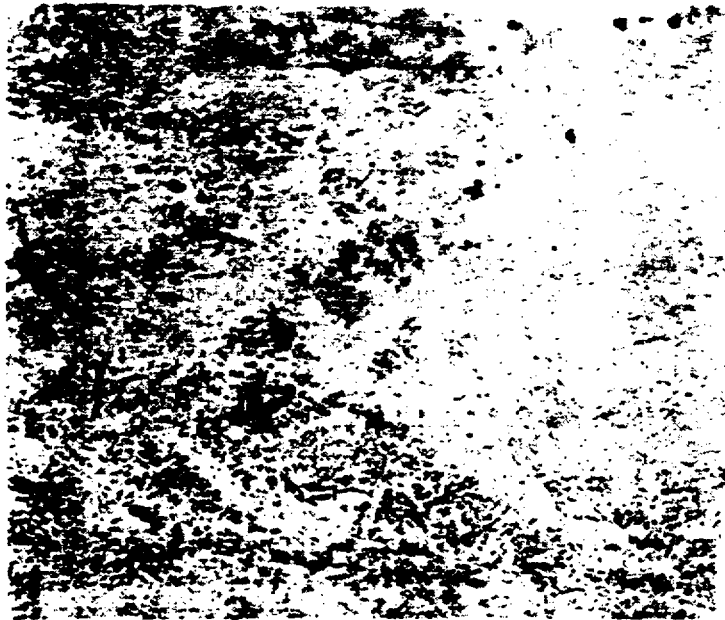


Figure 5.2 - Typical Microstructure of Irradiated HEU Aluminum-Uranium Alloy (33.9 wt% U-Al) (100X) [5.3]

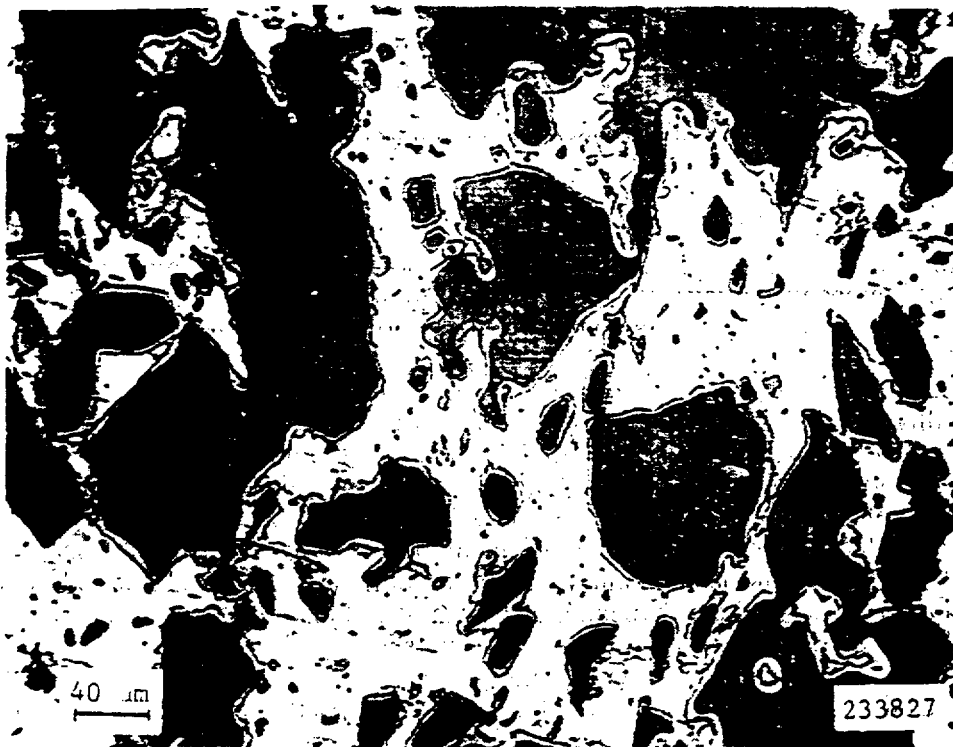


Figure 5.3 - Typical Microstructure of Irradiated (90% burn-up) Aluminum-Uranium Silicide (U_3Si_2) Alloy (500X) [5.3]



Figure 5.4 - Typical Microstructure of Irradiated Aluminum-Uranium Oxide (38 wt. % U_3O_8 - Al) Alloy (50X) [5.3]

5.3.4 Examinations/Observations

Research reactor Al SNF is presently stored in water basins of various degrees of water quality. An on-site examination of Brazilian SNF was performed in July 1996 [5.4]. The Brazilian fuel is assumed to represent some of the "worst case" fuel conditions expected to be received in the aluminum SNF disposition process. Photographs of the fuel assemblies highlighting the worst observed cases of pitting corrosion is shown in Figure 5.5.



Figure 5.5 - Close-up photograph of MTR Assembly IEA-53 from the IEA-R1 Reactor at the IPEN, São Paulo, Brazil.

The corrosion nodules were subsequently removed to reveal through-clad pits. Fuel in this condition would need to be encapsulated for storage in the SRS basins [5.5]. Fuels in

this condition (some through-clad pitting) are acceptable for interim dry and repository storage (see section 3.0).

5.4 Characterization Requirements - Program Status

5.4.1 Regulatory Review

Currently, the review of regulatory documents and drivers is in progress in order to determine the minimum characterization requirements for the disposition of aluminum SNF. A summary of the review is due to be completed at the end of April 1997 as one of the deliverable reports outlined in the task technical plan. A list of major governing laws is given here in chronological order:

- *The Atomic Energy Act of 1954 (AEA)* outlines the framework for the safety and licensing of nuclear facilities and activities involved in the management of source, special nuclear and byproduct materials.
- *The Clean Air Act of 1963 (CAA)* provides regulatory standards for all toxic and/or hazardous air pollutants under the National Emission Standards for Hazardous Air Pollutants (NESHAPs), for which 40 CFR 61 (Code of Federal Regulations) is the EPA (Environmental Protection Agency) interpretation and DOE Order 5400.5 is the DOE guideline.
- *The National Environmental Policy Act of 1969 (NEPA, Public Law 91-190)* states that Federal plans, functions, programs, and resources must be used to achieve six general goals, including the assurance of "safe, healthful, productive, and aesthetically and culturally pleasurable surroundings" for all Americans.
- *The Resource Conservation and Recovery Act of 1976 (RCRA, Public Law 94-580)* regulates waste that meets two criteria: i) waste must be a solid, and ii) it must exhibit certain hazardous characteristics (interpreted in 40 CFR 261). RCRA establishes a "cradle-to-grave" regulatory program for current hazardous waste activities.
- *The Clean Water Act (Federal Water Pollution Control Act Amendments of 1977, FWPCA)* is concerned with surface water, drinking water, and its sources.
- *The Nuclear Waste Policy Act of 1982 (NWPA, Public Law 97-425)* is a successor to the AEA and provides environmental protection standards for management and disposal of SNF, high-level waste (HLW), and transuranic waste (TRU) and specifies the requirements for characterization and licensing of a federal HLW repository.
- *The Comprehensive Environmental Response, Compensation, and Liability Act of 1980 (CERCLA, Public Law 96-510)* and the *Superfund Amendments and Reauthorization Act of 1986 (SARA)* and amendments establish a response program for previous hazardous substance processing and disposal activities.
- *The Federal Facilities Compliance Act of 1992* mandates the development of plans for treatment capabilities for mixed wastes for each DOE site where mixed wastes are generated and/or stored.

Primary interpretation of these governing laws is given in the Code of Federal Regulations (CFR). DOE orders are supplementary to the CFR and are written for

specific application to DOE tasks. Because aluminum SNF is not technically classified as either HLW, TRU, or fission products, but exhibits characteristics of all defined waste types, guidelines specific to the handling, transportation, storage, and disposition of aluminum SNF are subject to open interpretation. It is anticipated, however, that the requirements for commercial nuclear fuel to be dispositioned in a geological repository will be followed with some exceptions made to account for the physical, chemical, metallurgical, and volume differences between DOE-owned and commercial fuels.

A list of regulations and orders believed to be the most pertinent to the disposition of aluminum SNF is provided in Table 5.5. These documents are currently under review for direct applicability to aluminum SNF characterization. From these documents, interpretation of the requirements will be made, and preliminary characterization requirements will be established. Because these documents are subject to open interpretation, the minimum degree of characterization required will be assumed until dictated otherwise by regulatory drivers or repository waste acceptance criteria (WAC) expected to be provided by the Office of Civilian Radioactive Waste Management (OCRWM). These requirements will be determined in conjunction with the preliminary data needs previously issued by the OCRWM in August 1996 [5.6, 5.7].

Table 5.5 - List of Regulations Currently Under Review

10 CFR 20	Standards for Protection against Radiation (NRC)
10 CFR 60	Disposal of High-Level Radioactive Wastes in Geologic Repositories (NRC)
10 CFR 61	Licensing Requirements for Land Disposal of Radioactive Waste (NRC)
10 CFR 71	Packaging and Transportation of Radioactive Material (NRC)
10 CFR 72	Licensing Requirements for the Independent Storage of SNF and High-Level Radioactive Waste (NRC)
40 CFR 61	National Emission Standards for Hazardous Air Pollution
40 CFR 191	Environmental Radiation Protection Standards for Management and Disposal of SNF, HLW, and TRU (EPA)
40 CFR 261	Identification and Listing of Hazardous Waste (EPA)
40 CFR 264	Standards for Owners and Operators of Hazardous Waste Treatment, Storage, and Disposal Facilities (EPA)
40 CFR 265	Interim Status Standards for Owners and Operators of Hazardous Waste Treatment, Storage, and Disposal Facilities (EPA)
40 CFR 268	Land Disposal Restrictions (EPA)
49 CFR 172	Hazardous Materials Table, Special Provisions, Hazardous Materials Communications, Emergency Response Information, and Training Requirements (DOT)
49 CFR 173	Shippers - General Requirements for Shipments and Packaging (DOT)
DOE Order 5400.3	Hazardous and Radioactive Mixed Waste Program
DOE Order 5480.3	Safety Requirements for the Packaging and Transportation of Hazardous Materials, Hazardous Substances, and Hazardous Wastes
DOE Order 5633.3A	Control and Accountability of Nuclear Materials
DOE Order 5820.2A	Radioactive Waste Management

6.0 TEST PROTOCOL

The objective of the tasks under the test protocol program is to develop, validate and implement the test methodologies, and associated technologies, necessary to assess the suitability of waste forms for storage, handling, transfer and repository disposal. Test methodology development is divided into three basic steps: 1) review and analysis of established, and emerging, test methods that assess commercial nuclear fuels and high level waste glasses, 2) identification of the technology needs that may be specific to waste forms from aluminum based SNF, and 3) modification and/or expansion of the established methods to provide a test protocol that will assess the relative quality of waste forms associated with aluminum based SNF. Although the specific waste form for repository storage has not been established, the characteristics anticipated for waste forms developed for aluminum based SNF will differ significantly from the characteristics of either commercial nuclear fuels or waste glasses.

Evaluation of the suitability of a waste form for "road ready" storage and subsequent geologic disposal must include tests and analysis that assess the potential for nuclear criticality and environmental consequences. Ultimately, the results of the assessment must demonstrate that inclusion of the waste forms will not adversely impact the performance assessment of the repository. The waste forms developed from the aluminum based fuels may differ significantly from direct disposed commercial nuclear fuels and from glasses manufactured in the Defense Waste Processing Facility (DWPF). These differences include:

- a) ^{235}U enrichment, which may be as high as 93% if the waste form is created by direct disposal, and
- b) waste form stability (i.e. chemical durability, mechanical integrity and radiation and thermal stability).

The waste forms for the aluminum based fuels are likely to be more reactive than other waste forms placed in the repository. Additionally, both the aluminum cladding and aluminum-uranium core material in the aluminum based fuels are more reactive than the alloys anticipated for use in the "road ready" canister. The Zircaloy cladding and uranium oxide fuel core from the commercial fuels and the glasses from the DWPF process will generally be less reactive than the canister materials. These differences, coupled with the potential for a higher ^{235}U content in the waste form, suggest that the compatibility of the waste form with the anticipated storage and repository environments must be established and the technologies used to establish that compatibility must be well understood. The understanding is necessary to provide a technical basis to extrapolate the short term test data into the long term regimes of geologic disposal. The initial effort to establish the test protocol to assess waste forms developed for aluminum based SNF was to review the literature associated with testing, analysis and qualification of other waste forms.

6.1 Review of Literature

6.1.1 Background

Disposal of the aluminum based SNF will be a staged process that includes:

- a) continued, interim, wet storage,
- b) transfer to interim, dry storage, (The transfer may involve drying, canning, melt-dilute, and/or any of a variety of other possible treatments to provide the dry storage, waste form.), and
- c) disposal in a repository.

The test methodologies developed through the test protocol program will be designed to assess the waste forms after treatment and prior to transfer to interim, dry storage. The waste form will include both the treated SNF and the packaging required to make the treated fuel "road ready" for transfer to, and disposal in, the repository. The repository is assumed to be located in the Topopha Spring Member of the Paintbrush Tuff under Yucca Mountain. Yucca Mountain is approximately 100 miles northwest of Las Vegas, Nevada, and in the Nevada Test Site (1). The repository horizon is anticipated to be well above (100+ meters) the static water table, thus, in-repository corrosion of the waste package will involve interaction with water which permeated from the surface, through the mountain and through the repository back-fill before contacting the waste package. The chemical composition of that water will change during permeation by dissolution of selected constituents from the soil, rock and other components in the permeation/repository environment. Radiolysis may also effect the chemistry of the water. The probable types of corrosion (uniform, pitting, stress corrosion cracking, etc.) and the corrosion rate for any specific type will depend, to a large extent, on the water composition. Although the exact composition cannot be established until the materials of construction for the repository are specified, testing for suitability for repository service has generally involved the use of water having a chemistry similar to that of J-13 well water, as summarized in Table 6.1.

Table 6.1 - Concentration Ranges for Chemical Species in J-13 Well Water
(References for Table 1 are included in Reference 6.1 of this report.)

Environmental Variable	Nominal Concentration of Well Water (mg/l)	Concentration Range of Groundwater Heated to 90-250°C with Tuff Rock (mg/l)	Concentration Range of Groundwater Heated to 90-150°C in Presence of Radiation (mg/l)
pH	7.6	5.8 - 8.3	7.6 - 9.0
SiO ₂	58	29 - 394	1.05 - 3.10
HCO ₃ ⁻	125	45 - 195	8.1 - 21
F ⁻	2.2	2.2 - 4.4	0.38 - 1.4
Cl ⁻	6.9	6.5 - 8.9	2.5 - 5.8
NO ₃ ⁻	9.6	8.5 - 16.8	2.5 - 18.8
SO ₄ ²⁻	18.7	13.3 - 22	1.8 - 6.2
NO ₂ ⁻	---	0.7 - 1.5	1.2 - 3.8
H ₂ O ₂	---	---	0 - 4.8
Al ³⁺	0.012	0.016 - 4.8	<0.15 - 0.18
Fe ²⁺	0.006	---	<0.02 - 0.04
Ca ²⁺	12.5	0.21 - 13.2	2.7 - 9.8
Mg ²⁺	1.9	0.009 - 2.0	0.6 - 1.1
K ⁺	5.1	3.2 - 19.4	2.4 - 4.8
Na ⁺	44	35 - 74	2.8 - 36

The initial placement of the waste package to the repository environment may involve exposure to near ambient temperature water. The temperature should rise soon after repository closure, causing the water to boil and exposing the waste package to steam. After some period of time the repository should dry out and the only exposure of the waste package will be to water that may penetrate the environment through cracks, fissures and other defects in the rock formations and drip or flow toward the waste package. The temperature of the dripping water will rise as it progresses through the repository and, depending on the position of the waste package, may be turned to steam before the contact is made. The specific evolution of time-temperature profiles will depend on repository loading. High heat loads, 114 kW/acre, will produce the time-temperature profiles shown in Figure 6.1.

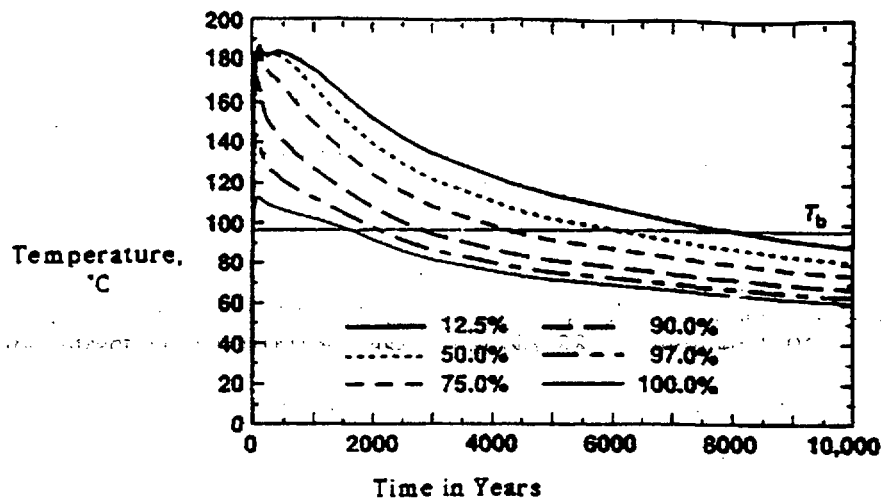


Figure 6.1 - Calculated Effect of Position and Time on Repository Temperature. The boiling temperature of water in the repository is T_b and the % values estimate the fraction of the repository considered with the center being zero (Reference 6.2)

The hot dry environment at the center of the repository will significantly decrease the tendency for corrosion induced degradation during the first 10,000 years of exposure. Because of this tendency, repository loading becomes a major consideration in the assessment of potential package failure rates. Estimates of package failure frequencies suggest that, if the outer package is constructed from alloy 825, "most of the containers in the 55 kW/acre case will fail (by pitting corrosion), but most of the containers in the 110 kW/acre case will not, except for those at the edge" [6.3]. These and other similar analyses coupled with the emerging acceptance criteria of approximately 200°C for direct disposal of aluminum based SNF, suggest that if the direct disposal option is used, the aluminum based fuels will be relegated to the repository edge where the tendency for corrosion is maximized [6.4]. Such selective placement would not be necessary in a colder repository and could be avoided if a disposal technology such as melt-dilute were used. However, regardless of disposal technology, the repository temperature profile, and/or the material of construction for the outer canister of the waste package, radionuclide release from the waste package is not anticipated until the outer canister is penetrated and the "road ready" waste form is exposed to the repository environment. The time frame for the initiation of this event will depend on: canister material, repository thermal loading, permeability of the repository site, and the chemistry of the water ultimately contacting the outer surface of the canister. The post penetration release of radionuclides, including ^{235}U , will depend on the condition and behavior of the "road ready" waste form.

The large scale, ongoing efforts to determine the time for the penetration of canisters containing commercial nuclear fuels should be directly applicable to the disposal of aluminum based waste forms. However, the chemical activity of the aluminum based waste form may be higher than the chemical activity of either commercial nuclear fuel or waste glass. This difference, coupled with the high ^{235}U enrichments in many of the aluminum based fuels, suggests that post canister penetration behavior may differ significantly from that anticipated for commercial nuclear fuels and waste glasses. The

technical basis to assess the magnitude and importance of this difference to the repository performance assessment will be developed through an understanding of the pre-breach condition of the "road ready" waste form and the post-breach interactions of the waste form with the evolving environment. Figure 6.2 is a schematic illustration of the characteristics that must be measured to make this assessment.

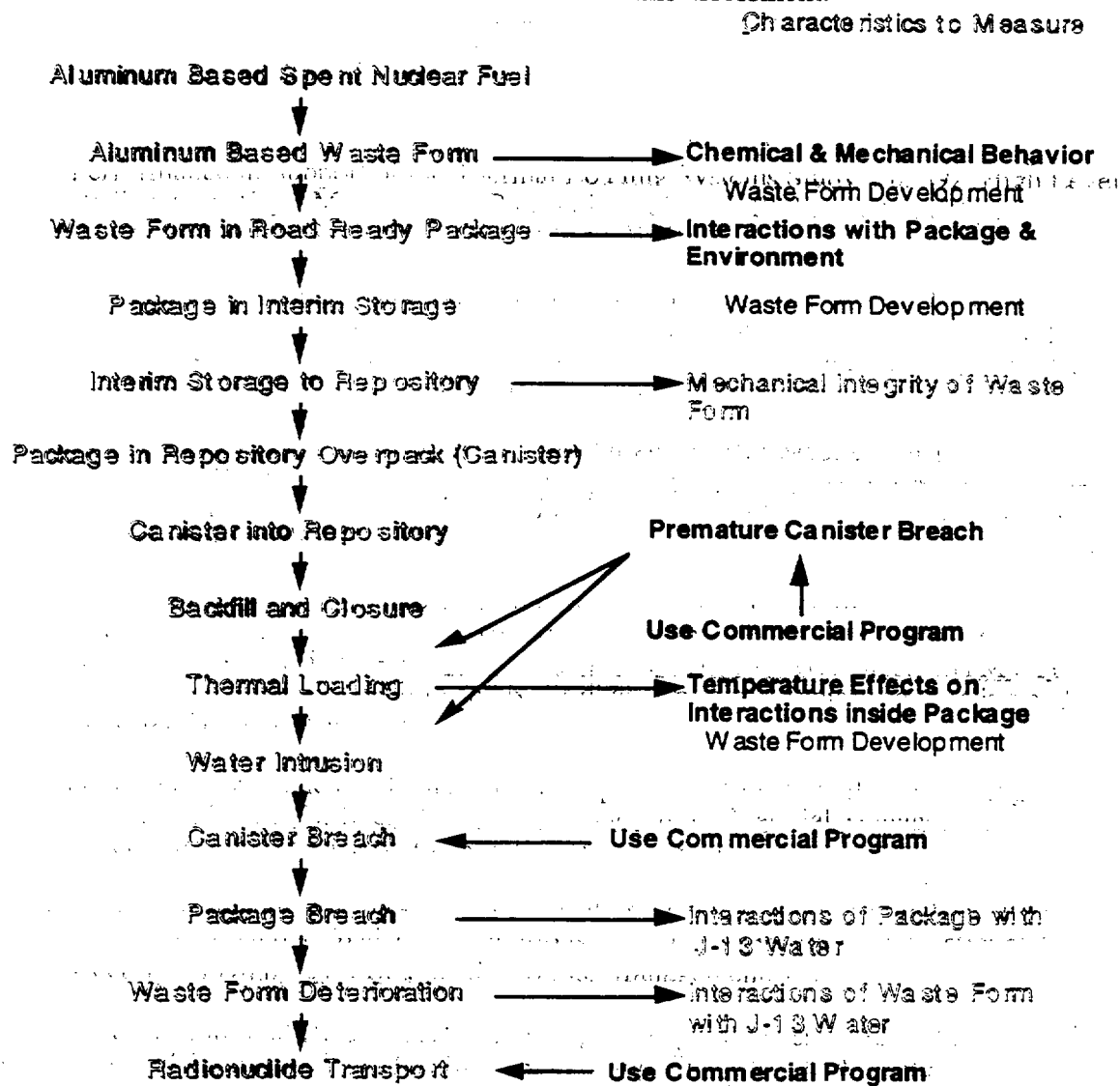


Figure 6.2 - Waste Form Characteristics Required to Assess Suitability for Repository Storage

The waste form/environmental interaction characteristics outlined in Figure 6.2 are similar to the characteristics currently measured during the evaluations of nuclear waste glasses and commercial spent fuels. These characteristics may be determined through a single, very broad, test protocol or through several, highly focused test protocols.

The on-going review of available literature suggests that a minimum of three basic types of tests are required to assess waste form suitability. The three tests should provide

techniques to establish: 1) the release rate of radionuclides from the waste form, 2) the role of environmental variables and waste form integrity on the release rate, and 3) the extent of transport and relocation of the radionuclides. The anticipated behavior of aluminum based waste forms is substantially different from the behavior of commercial nuclear fuels and defense waste glasses. Thus, test methods to measure radionuclide release may differ significantly from tests currently used to assess the suitability of more inert waste forms. The mechanisms of radionuclide transport and relocation should be consistent with those anticipated for other waste forms and work in this area will basically involve fitting corrosion and release data from the aluminum based wastes into the assessment methods developed for other waste forms.

6.1.2 Predicting Deterioration and Corrosion

Knowledge of the condition of the waste form, prior to placement in the "road ready" package, will be established through the characterization program and/or the treatment processes used to convert the spent nuclear fuel into a waste form. The assessment of this initial condition, including chemical reactivity and mechanical properties, must be determined through established through approved and validated test methods and consistent with the overall approach to life prediction used to assess the repository emplacement of commercial nuclear fuels and defense waste glasses. There are basically three types corrosion evaluations that have been made to assess repository lifetimes:

- 1) evaluation of the behavior of the over-pack or canister material in anticipated repository environments,
- 2) evaluation of the behavior of the waste form in anticipated repository environments, and
- 3) evaluation of interactions among the over-pack, waste form and/or the packaging material.

The corrosion studies have, since potential sites for the US repository were reduced to Yucca Mountain, emphasized corrosion evaluations in J-13 or modified J-13 water. These evaluations are supplemented with more basic corrosion evaluations necessary to provide the technical basis for extrapolation of the short term data.

The as-placed or initial condition of the waste form may deteriorate with time because of interactions with components of the environment and/or material associated with the "road ready" package. This deterioration, or lack thereof, must be assessed with reasonable confidence over time spans exceeding the span of recorded history. A requirement of EPA standard 40CFR191 is that the repository system isolate radionuclides from the accessible environment for 10,000 years. Additionally, the assessment be based on relatively short term tests (years at most) and applicable to a variety of repository emplacement strategies. The repository canister or over-pack may be a corrosion resistant material such as alloy 825 or a corrosion allowance material such as low carbon or mild steel. The thermal loading may be above or below the boiling temperature of water and the back-fill may include any of a variety of materials. Uniform corrosion, pitting, crevice corrosion, galvanic corrosion, stress corrosion cracking and perhaps even microbiological corrosion could develop and promote early release of radionuclides. Corrosion of the waste form could initiate prior to a canister breach if conditions inside the "road ready" package do not inhibit corrosion and oxidation during interim storage and/or during the initial stages of repository storage. The

U. S. Nuclear Regulatory Commission requirements for geologic disposal of high-level nuclear waste, as contained in 10 CFR 60.113, specify that waste packages shall be designed such that containment of radionuclides will be substantially complete for a period of at least 300 to 1,000 years [6.5]. This time span is significantly longer than the 50 to 100 year times typically sighted as the interim storage period.

The behavior of canister materials in the repository environment has been, and is being, extensively studied. This Office of Civilian Radioactive Waste Management work will provide predictions of the time to canister breach and the anticipated failure mode. Pitting corrosion is generally considered the most probable failure mode for corrosion resistant canister materials while failure by uniform corrosion is considered probable for corrosion allowance materials. A corrosion-induced canister breach will expose the "road ready" package to the repository environment. The waste form will also be exposed if corrosion and/or oxidation compromised the integrity of the package prior to canister breach. Exposure of the waste form to the repository environment will provide a pathway for release of radionuclides.

The rate of release of radionuclides from the waste form to the repository environment will depend on several factors including:

- a) the physical and chemical condition of the waste form (phases present, surface-to-volume ratio, crevice or crack morphology, etc.)
- b) the degree of electrical contact between the waste form and surrounding package and canister materials (galvanic corrosion),
- c) the chemistry and temperature of the surrounding water, and
- d) the flow of water across the exposed waste form.

This release, or loss of containment, is the first stage in the spread of radionuclides from the immediate repository environment and could lead to the formation of actinide deposits in the repository. Both the spread of radionuclides and actinide deposit formation are being actively studied in ongoing programs in the Office of Civilian Radioactive Waste Management. For example, the possibility of a critical event in Yucca Mountain is "chiefly dependent on geologic and geochemical constraints on the accumulation of tens to hundreds of kilograms of plutonium or highly-enriched uranium in volumes of ten cubic meters" [6.6]. There are three distinct stages to this process: release from the waste form, transport to void volumes in the rock, and localized deposition from solution. Significant differences between the aluminum based waste forms and the commercial spent fuels include: the reactivity of the waste form and, potentially, the ^{235}U enrichment. The primary potential impact of these differences is on the release rate of radionuclides from the waste form. This release rate must be evaluated before aluminum based waste forms can be effectively included in assessments of materials behavior in the repository. Therefore, one of the primary requirements of the test protocol will be to provide validated test methodologies to measure radionuclide leaching under simulated repository conditions.

6.1.3 Techniques for Testing

A single pass, flow through technique has been used to determine the dissolution rate of the spent fuel. This technique requires a relatively simple, four major component system which consists of: a feed water container, a controlled flow pump, a specimen column and a sample/waste collection unit, as illustrated in Figure 6.3.

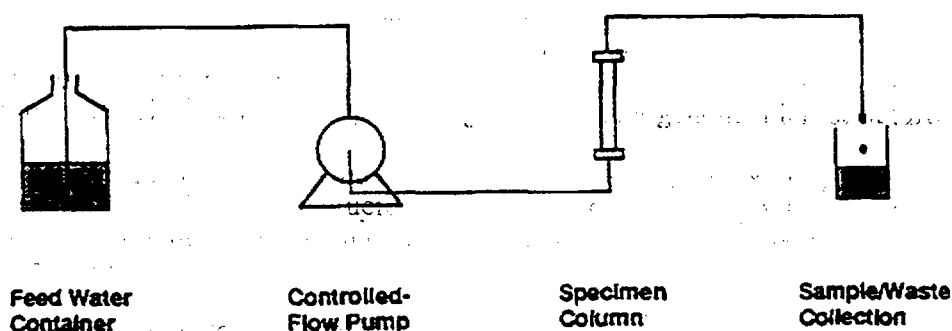


Figure 6.3 - Schematic illustration of the various units used in the flow through technique for determining the spent fuel dissolution rate kinetics

The feed water containers store the simulated repository groundwater and are vented to provide an equilibrium between the water and the atmospheric air. The flow rate of water from the containers to the specimen column is adjusted to the desired value with a controlled flow pump. The flow rates are measured by measuring (weighing) the time rate of release of water from the specimen chamber. The walls of the specimen columns are Type 316 stainless steel and the internal surface of the container is mirror polished. The internal dimensions of the specimen column is 6.35 mm diameter and 50.8 mm long. Fritted stainless steel filters (2 μm) are attached at both ends of the specimen column. The waste form (spent fuel) is placed in the specimen chamber and processed so that the particle size approximates that anticipated when the waste form is exposed to the repository environment. For example the particle size for commercial spent fuels range from 1 to 3 mm diameter because this size range is typical of fuel particle sizes after thermal cracking during irradiation in a light water reactor. The appropriated particle size range can be obtained by crushing and screening the waste form. The small particles adhering to the surfaces of larger particles are rinsed from the surfaces before the properly sized particles are conditioned, or tested in zero flow water, before the flow through experiments are initiated. The purpose of conditioning is to (a) get an estimate of the maximum steady state uranium concentration in the water and (b) initiate the alteration of the waste form under static conditions before using them in the flow through or dynamic testing procedure. Single flow through experiments on non-irradiated UO_2 pellets have shown that a U concentration of 700 ng/mL can be reached under static steady state conditions when testing in synthetic J-13 water [6.7].

The sample/waste collection chamber is used to collect water samples for determining the uranium concentration as a function of either flow rate and/or the time of testing. The main purpose of varying the flow rate to determine if a flow independent dissolution rate was achieved [6.7]. However, nonlinear relationship between the uranium concentration and inverse flow rate of synthetic J-13 water was observed. Similar experiments in deionized water produced linear plots for the dissolution rate as a function of the inverse flow rate. The results suggest that the relatively high concentrations of carbonates

produced a higher rate of dissolution. One aspect of the test protocol program is to obtain contracts with established waste form testing laboratories, such as PNNL, to develop the procedures, data and analysis necessary to formulate a test protocol.

The importance of test protocol is readily apparent from the effect of specimen column material on test conditions. The use of polymethylpentene cells with Teflon tubing and cell filters allowed the diffusion of oxygen through the plastics and made it very difficult to control the amount of dissolved oxygen in the system [6.8]. This effect was not observed in a stainless steel system. Many other tests have been used to characterize the durability of spent nuclear fuels and other nuclear waste forms and these tests must also be evaluated before a test protocol is established.

6.2 Pyrophoricity

The ASTM Committee C-26 is working to develop a standard guide for pyrophoricity testing of metallic spent nuclear fuels. The ASTM effort is directed toward metallic uranium fuels but the potential for pyrophoric reactions in the aluminum based waste forms must be considered. Initial evaluations indicate that the potential for pyrophoric reactions in the aluminum based spent fuels is minimal. However, because of the potential impact of the emerging standard guide for metallic spent nuclear fuels to the test protocol(s) for aluminum based fuels and because of the general applicability of the ASTM Committee C-26 activities to the development of any test protocol(s) for nuclear waste forms, membership on, and interaction with, the committee has been obtained.

6.3 Property Measurements

The development of a test protocol requires the identification of the specific and collective physical and chemical properties of the form that are important to safe and efficient storage in the repository environment. Review and evaluation of the literature demonstrate that general corrosion of the waste form and/or selective leaching of specific constituents from the waste form are of primary importance. Prediction of corrosion rates will require a knowledge of the active and the dominant corrosion mechanisms. This will require a knowledge of the metallurgical condition of the waste form, the evolution of that condition during interim and repository exposure and the physical condition and integrity of the waste form. For example, the leach rate will depend on the surface-to-volume ratio and will generally increase as the surface-to-volume ratio increases. However, importance of waste form surface-to-volume ratio may be insignificant relative to the importance of material-environmental effects that promote galvanic corrosion or initiate crevice or microbiologically induced corrosion. Measurement of "properties" to define the operative modes or submodes of corrosion is therefore of significant importance to the test protocol.

Corrosive "properties" are frequently measured as a function of operational or system variables. Pourbaix diagrams, Figure 6.4, define corrosion tendencies as a function of

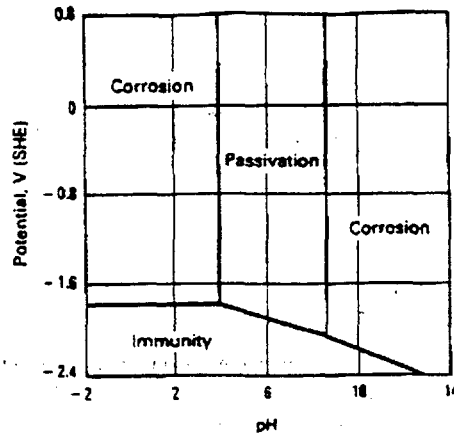


Figure 6.4 - Pourbaix diagram for aluminum with a hydrates oxide film

potential and pH. Polarization diagrams, Figure 6.5, provide indications of the tendencies for stress corrosion cracking (zones 1,2 and 3) pitting (above zone 3), general corrosion (E_{corr}), hydrogen affected processes (zone 1) and passivity (between zones 2 and 3).

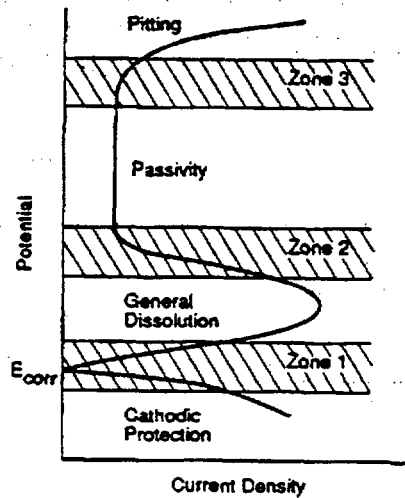


Figure 6.5 - Illustration of polarization diagram showing regions of general corrosion, cathodic protection, pitting, and stress corrosion cracking

Electrochemical measurements to determine the corrosion potential and the pitting potential as a function time and environment have been coupled with pit growth kinetics and the stochastic nature of the pitting process to assess the long term behavior of canister materials in repository environments, Figure 6.6.

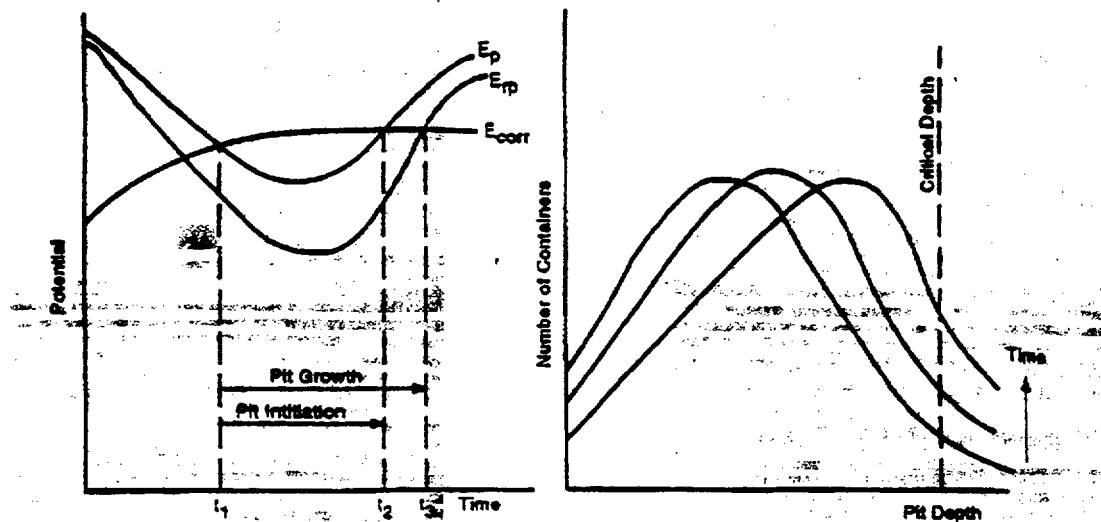


Figure 6.6 - A schematic illustration of predicting pitting failure in canister materials (After Reference 6.5)

In addition to measurement of the electrochemical behavior of the waste form, a test methodology that will be based on ASTM standards, test methods must also measure properties such as leach rate, galvanic effects and determine the effect of environmental variables on electrochemical behavior, leaching and other corrosion processes.

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