**Idaho
National
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Laboratory**

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July 1996

***Initial Evaluation of Dry Storage
Issues for Spent Nuclear Fuels in Wet
Storage at the Idaho Chemical
Processing Plant***

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STORAGE AT THE IDAHO CHEMICAL PROCESSING PLANT**

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Pacific Northwest Laboratory
Richland, Washington 99352

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ABSTRACT

The Pacific Northwest Laboratory has evaluated the basis for moving selected spent nuclear fuels in the CPP-603 and CPP-666 storage pools at the Idaho Chemical Processing Plant from wet to dry interim storage. This work is being conducted for the Lockheed Idaho Technologies Company as part of the effort to determine appropriate conditioning and dry storage requirements for these fuels. These spent fuels are from 22 test reactors and include elements clad with aluminum or stainless steel and a wide variety of fuel materials: UAl_x , UAl_x -Al and U_3O_8 -Al cermets, U-5% fissium, UMo, $UZrH_x$, UErZrH, UO_2 -stainless steel cermet, and U_3O_8 -stainless steel cermet. The study also included declad uranium-zirconium hydride spent fuel stored in the CPP-603 storage pools. The current condition and potential failure mechanisms for these spent fuels were evaluated to determine the impact on conditioning and dry storage requirements. Initial recommendations for conditioning and dry storage requirements are made based on the potential degradation mechanisms and their impacts on moving the spent fuel from wet to dry storage. Areas needing further evaluation are identified.

EXECUTIVE SUMMARY

What are the issues and options for acceptable dry storage of aluminum (Al)-clad, stainless steel (SS)-clad, and unclad uranium-zirconium hydride spent fuel currently in wet storage at the Idaho Chemical Processing Plant (ICPP) in CPP-603 and CPP-666? This is the central question being addressed by Pacific Northwest Laboratory for the Lockheed Idaho Technologies Company (LITCO). This evaluation has focused on obtaining information on past storage histories and fuel conditions, and evaluating failure mechanisms for these spent fuels to determine their vulnerabilities during preparation for and movement to dry storage.

The four primary types of spent fuel evaluated include uranium-aluminum (UAl_x), uranium oxide (UO_2), uranium-fissium metal, and uranium zirconium hydride ($UZrH_x$). The UAl_x fuel is clad in Al; the UO_2 fuel and uranium-fissium metal are clad in SS; the $UZrH_x$ fuel is either clad in Al, SS, or some has had the cladding removed. The three major contributors to this spent fuel inventory, based on the number of units stored, are from the Experimental Breeder Reactor II (EBR-II, ~55%), the Advanced Test Reactor (ATR, ~13%), and the Training, Research and Isotope Production Reactor - General Atomics (TRIGA) spent fuel with Al cladding (TRIGA Al, ~9%). These spent fuels were selected for evaluation because of known corrosion in the CPP-603 for aluminum alloys (ATR, TRIGA-Al, and storage components) and potential issues for other materials (SS and $UZrH_x$).

There are different issues for continued wet storage or future dry storage of each of these spent fuels. Three major issues exist for Al-clad spent fuels.

- The cladding is degrading in the CPP-603 environment, and probably in the CPP-666 pool, based on examinations and open literature information.
- The formation/presence of UH_3 is thermodynamically possible for some of the fuel/environment combinations. This is of concern because low-temperature ignition and combustion of UH_3 could become an issue in drying operations or subsequent dry storage if sufficient quantities form.
- Moisture contained in hydrated corrosion products could contribute to continued degradation if sufficient release occurs; however, the release cannot be presently quantified, even though the amount is expected to be low.

Two primary degradation mechanisms appear to exist for SS-clad spent fuels.

- Intergranular stress corrosion cracking of the cladding is suspected in these spent fuels because the irradiation temperatures were in the range for sensitization and embrittlement. The EBR-II spent fuel is in storage canisters, but the actual environment under which the spent fuel has been exposed (inert or air, moist or dry) has not been established. This perceived issue could affect over half of the spent fuel inventory, depending on its prevalence.
- Formation of UH_3 may be an issue in the EBR-II, ANL-6, and SPEC fuels, although the rate of formation should be slower than for uranium alone.

According to the open literature, the unclad UZrH_x should be relatively inert in the current and potential storage environments, but the material is extremely brittle. Furthermore,

- Special handling and packaging operations could be required due to the extreme brittleness of this material.
- No quantitative data were found for the performance of UZrH_x in these environments to confirm the conclusion on inertness. If the UZrH_x is very inert, then failure of Al-clad or SS-clad spent fuels containing this material will be less important, as long as the fuel rods can be handled.

Solutions to these issues appear possible based on information in the literature on degradation mechanisms, experience with similar fuels or materials at other facilities, and extensive experience with wet and dry storage of commercial spent fuel. A first estimate of the acceptable storage conditions for the Al-clad, SS-clad, and unclad UZrH_x fuels is summarized below and discussed in Section 6:

- For drying operations involving all the spent fuels evaluated, initial temperature limits appear to be below 300°C (572°F) except for SS-clad U alloy fuel, and evacuation to <5 torr static pressure before filling with a cover gas. Limits for the SS-clad U alloy fuel appear to be $<200^\circ\text{C}$ (392°F). Considerations for drying operations include residual moisture for all of the fuels, which is the main reason for the 300°C (572°F) limit, plus concern over uranium oxidation for the EBR-II fuel.
- For dry storage of all spent fuels evaluated, with the exception of the SS-clad UZrH_x and the unclad UZrH_x , initial dry storage limits of $<150^\circ\text{C}$ ($<302^\circ\text{F}$) and 99.995% He, Ar, or N_2 are suggested. If UH_3 is proven not to be an issue, then it might be possible to justify air storage for Al-clad UAl_x and U_3O_8 fuels. The main consideration for these fuels is release of moisture entrained in the fuel or oxide films.
- Dry storage temperature limits for the SS-clad UZrH_x and the unclad UZrH_x are initially proposed to be $<400^\circ\text{C}$ ($<752^\circ\text{F}$) with 99.995% He, Ar, or N_2 cover gas. Considerations in determining these limits include moisture and hydrogen release.
- For a given spent fuel, if UH_3 is detected in significant amounts, conditioning treatments may be justified; drying and canning operations must be conducted to avoid ignitions.

Based on the evaluation thus far on the spent fuel conditions and known or potential failure mechanisms, several recommendations can be suggested to determine whether the initial limits proposed for drying operations

and dry storage are valid.

Recommendations for Al-Clad spent fuels are as follows:

- Determine whether UH_3 is present and in sufficient quantity to warrant conditioning.
- Determine the amount of moisture in corrosion films and evaluate what drying procedures will be required to limit hydrogen and moisture release during long-term dry storage. If UH_3 is not prevalent or is not an issue, determine whether air storage for the Al-clad fuels is feasible.

Recommendations for SS-Clad spent fuels are as follows:

- Determine the former and current storage environment for the EBR-II spent fuel. It is important to know if the fuel has been exposed to moisture, and whether the cladding is significantly sensitized.
- If it appears that the EBR-II fuels are failed, verify whether UH_3 formation in the U-fissium fuel is in sufficient quantity to be a significant problem.

Recommendations for Unclad UZrH_x spent fuel are as follows:

- Verify whether the fuel is stable in water and determine if any conditioning is justified for long-term dry storage. No definitive data were found to resolve these issues, but it would be a very important conclusion to all the fuels containing UZrH_x .
- Verify the brittleness of the UZrH_x fuel to determine whether unclad or clad fuel of this type will present a handling issue.

Evaluation of dry storage for the spent fuels in the CPP-603 and CPP-666 is challenging because of the wide variety of fuel and cladding materials that have been exposed to many different irradiation and storage histories. If evaluation of the fuel conditions and failure mechanisms can be improved to where some or most of the issues are eliminated, then dry storage requirements may be less restrictive than those suggested. The fuels could be segregated into failed and unfailed categories, possibly reducing the extent of any conditioning required. Some Al-clad spent fuels have been moved to dry storage with minimal processing. However, selection of failed and unfailed fuel would require an assessment of the available techniques for making such a selection, and might be a significant task in itself.

It appears that additional information will be required to verify these initial storage limits or to determine how to condition the fuel, particularly for fuel known to have failed cladding. However, dry storage is a mature, widely applied technology for commercial spent fuel. While no failed uranium-bearing fuel subjected to aqueous corrosion has been moved to long-term dry storage, methods have been demonstrated to safely condition fuel that may include uranium hydrides.

This report proposes an approach that includes identifying fuel categories to facilitate efficient and cost-effective movement to dry storage as well as assuring that important issues are identified and properly addressed.

Some fuel types seem likely to require minimal conditioning for safe movement to dry storage. In any event, the stakeholders of this facility may be required to make hard choices among proposed schedules, costs, and adequately understanding the needed conditioning or storage environments.

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ACRONYMS

AEA	Atomic Energy Act of 1954
AGE-2	Atomic General Electric - 2nd Shipment
AI	Atomics International (a SNAP fuel and company)
Al	aluminum
APPR	Army Power Production Reactor
ARMF	Advanced Reactivity Measurement Facility
ATR	Advanced Test Reactor
BMI	Battelle Memorial Institute, Battelle Thermal Reactor
BORAX V	Boiling Reactor Experiment V
CAA	Clean Air Act
CERCLA	Comprehensive Environmental Response, Compensation and Liability Act
CFR	Code of Federal Regulations
CPP	Chemical Processing Plant
CPP-603	Chemical Processing Plant - Building 603
CPP-666	Chemical Processing Plant - Building 666
CS	Carbon Steel
DOE	Department of Energy
DOT	Department of Transportation
DP	Defense Production
EBR II, ANL-6	EBR II Scrap from ANL-6 Test
EBR-II	Experimental Breeder Reactor II
EPA	Environmental Protection Agency
ETR	Engineering Test Reactor
FSA	Fuel Storage Area
Fermi II	Enrico Fermi Atomic Power Plant (SS only)
FFCA	Federal Facilities Compliance Act of 1992
FFTF	Fast Flux Test Facility
f.p.	fission products
FSU	Former Soviet Union
FWPCA	Federal Water Pollution Control Act Amendments of 1977 to the Clean Air Act
GA	General Atomics
GCR	Gas Cooled Reactor
HFBR	High Flux Beam Reactor

HLW	High Level Waste
IASCC	Irradiation Assisted Stress Corrosion Cracking
ICPP	Idaho Chemical Processing Plant
IGA	Intergranular attack
IGSCC	Intergranular stress corrosion cracking
IFSF	Irradiated Fuel Storage Facility located in CPP-603
INEL	Idaho National Engineering Laboratory
ITA	Independent Technical Assessment Team
LANL	Los Alamos National Laboratory
LITCO	Lockheed Idaho Technologies Company
LLW	low level waste
LTS	low temperature sensitization
LWR	light water reactor
MIC	microbially induced corrosion
MOX	Mixed uranium-plutonium fuel
MURR	University of Missouri Research Reactor
mpy	mil per year
$\mu\text{S/cm}$	micro Siemens/centimeter
NEPA	National Environmental Policy Act
NRC	Nuclear Regulatory Commission
NWPA	Nuclear Waste Policy Act
ORR	Oak Ridge Reactor
PFR	Prototype Fast Reactor
PNL	Pacific Northwest Laboratory
RBOF	Receiving Basin for Off-Site Fuel (Savannah River)
RCRA	Resource Conservation and Recovery Act
RH	relative humidity
RIS	radiation induced segregation
SCC	stress corrosion cracking
SM-1A	SM-1A Army Reactor
SNAP	Systems for Nuclear Auxiliary Power
SNF	spent nuclear fuel
SPEC (OMRE)	Organic Moderated Reactor Experiment
SPSS (SPERT)	Special Power Excursion Reactor

SRE	strain rate embrittlement
SRP	Savannah River Plant
SS	stainless steel (austenitic unless otherwise indicated)
TRA	Test Reactor Area
TRIGA	Training, Research and Isotope Production Reactor - General Atomics
TRIGA Al	Kansas State, Cornell, Texas, Hanford, MSU and GA TRIGA SNF with Al cladding
TRIGA Ber II	Berliner Experimentier Reactor II (Germany)
TRIGA SS	MSU, GA, and Berkeley TRIGA SNF w/SS cladding
TRIGA FLIP	SMU, GA, and Berkeley TRIGA SNF w/ss cladding
TRU	transuranic waste
UTS	ultimate tensile strength
UW	University of Washington Argonaut Reactor
UK	United Kingdom
U.S.	United States
VBWR	Vallecitos Boiling Water Reactor
WINCO	Westinghouse Idaho Nuclear Company

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1.0 INTRODUCTION

The Lockheed Idaho Technologies Company (LITCO) established the Spent Fuel Assessment Project at the Pacific Northwest Laboratory (PNL)^(a) to define the basis for moving spent nuclear fuel (SNF) at the Idaho Chemical Processing Plant (ICPP) from wet storage to dry interim storage. The main purpose of this assessment was to identify the current condition of the spent fuels as a basis to assess the most probable degradation mechanisms that may occur during interim dry storage for selected SNF currently in the CPP-603 and CPP-666 storage pools, to provide initial conditioning and storage requirements, and to recommend further work to clarify these recommendations.

To effectively analyze conditioning requirements and fuel behavior in dry storage, it is essential to understand fuel materials, configurations, and historical data. Past efforts at PNL on analyzing dry storage requirements of commercial fuels were facilitated by accessibility of fuel histories and parameters. For essentially all fuels included in this study, the information was fragmentary. Therefore, a major effort of this work involved tracing the information in files that often lacked details; conducting telephone discussions with fuel fabricators, reactor operators, shippers, and storage pools operators; and reviewing relevant literature. Limits on time and resources precluded a full assessment of all fuels in this study. However, bounding fuel conditions were considered sufficiently clear that preliminary analysis of conditioning and dry storage could proceed for most fuels.

Background on this assessment, the scope of the study, and the approach used to make these evaluations are provided in Section 1. A description of the CPP-603 and CPP-666 facilities and the SNF under evaluation is provided in Section 2. These descriptions include known or assumed storage conditions for both the facility and SNF. Sections 3, 4, and 5 include descriptions and evaluations of the conditions and their impact on dry storage for three major categories of SNF: Al-clad spent fuels (Section 3), uranium-zirconium hydride (UZrH_x) spent fuel (Section 4), and SS-clad spent fuels (Section 5). Section 4 deals primarily with UZrH_x spent fuel material since the issues are common to all spent fuels containing this material regardless of whether the material is clad or not. Conclusions derived from this evaluation and suggestions for means of confirming these recommendations are provided in Section 6.

1.1 BACKGROUND

As with many other Department of Energy (DOE) facilities, the mission of the ICPP over the last few years has gradually changed from defense-related operations to waste management and environmental restoration. This has occurred in part because of changing defense requirements, and in part because of

^(a)Operated by Battelle Memorial Institute for U.S. Department of Energy

continued use of facilities that have reached or are reaching their design lives. In many cases, SNF has been stored in the storage pools well beyond original expected durations.

While the number of corrosion-related occurrences for all of DOE's Defense Programs facilities totals only 1.5% of the reported occurrences (DOE 1992), such degradation, if uncorrected, can lead to undesirable results. The need for this evaluation of degradation mechanisms of the materials in the underwater fuel storage facility (CPP-603 or FSF) and the underwater fuel storage area (CPP-666, FSA or FAST) is supported by the known corrosion of storage components in CPP-603 (Dirk 1994). Examination of Al fuel storage cans, Al fuel storage buckets, and other facility equipment indicate that Al has a general corrosion rate of $38.1 \mu\text{m/yr}$ (0.0015 in./yr or 1.5 mil/yr [mpy]), but has preferential attack by pitting. Galvanized carbon steel (CS) in the storage basin for about 40 years has shown corrosion rates of $102 \mu\text{m/yr}$ (4 mpy) and has some pitting. Type 304 Stainless Steel (SS) has had no observable corrosion after 13 years, but galvanic corrosion has been observed on CS welded to SS 304. These couples have had cracking of welds and intergranular cracking that is attributed to the chloride content in the CPP-603 pool and crevices formed during fabrication. Because the CPP-666 is a more recent facility and has modern controls, less evidence exists to support a degrading environment. However, algae observed on nonradioactive specimens in recent examinations of the CPP-666 facility suggests the possibility of microbially influenced corrosion (MIC) (Hoskins et al. 1993).

The projected SNF condition also needs evaluation to support canning operations for SNF in CPP-603 facility (Welland et al. 1993). The underwater fuel drying and canning in the CPP-603 pool or the dry canning operation in the CPP-603 Irradiated Fuels Storage Facility (IFSF) would involve handling the SNF and heating it to drive off excess water prior to repackaging or overpacking. The fuel condition, including the amount of water entrained in hydrated corrosion products, is needed to assist these operations.

1.2 SCOPE

The objective of this assessment is to identify a set of recommended dry storage conditions for the SNF in the ICPP storage basins. The work has focused on obtaining information on the past storage history, fuel condition, and evaluating failure mechanisms for the SNF listed in Table 1.1 to determine potential impacts on dry storage or other operations that may be conducted prior to dry storage, which may last about 50 years. Where possible, an initial set of storage limits is provided for dry storage, as well as anticipated conditioning operations based on available information and expert opinion; however, these recommendations are based on limited information for a number of the spent fuels. Emphasis in this report is on the SNF and not on the storage cans, buckets, racks, or other structural components because these materials have been examined and some information exists on their condition (Hoskins et al. 1993). Interaction of the cans and storage components with the SNF is considered when needed. Areas for future work, including experimental investigations, are outlined where applicable.

While 93 different SNFs have been identified at the Idaho National Engineering Laboratory (INEL) (DOE 1993b), this study addresses only those listed in Table 1.1 that are stored in the CPP-603 and CPP-666 pools and have Al or SS cladding or are UZrH_x from declad SNF. Some degradation mechanisms identified for SNF in the ICPP pools may apply to other SNF stored at INEL, but no specific information has been obtained on such storage to assess any differences that may result from storage other than in the ICPP basins. An exception is information obtained on wet storage of the Advanced Test Reactor (ATR) spent fuels in the ATR canal. The SS-clad Fermi I blanket SNF is in dry storage and is presently evaluated only as to potential failure mechanisms of its fuel and cladding materials without regard for the effects of prior extended dry well storage.

This study excluded SNF with zirconium or Zircaloy cladding, as well as SNF stored in locations other than the ICPP pools. This prioritization results from the more aggressive (high chloride) environment of the CPP-603 pools compared with the dry wells and dry vault storage of other SNF and the well established performance of the zirconium based alloys relative to Al and SS in water environments.

Table 1.1 Types of SNF Being Evaluated

<u>Fuel</u>	<u>Abbreviation</u>	<u>Fuel/Cladding</u>
Atomics International (a SNAP fuel)	AI	UZrH _x /declad
Army Package Power Reactor	APPR	UO ₂ SS/SS
Advanced Reactivity Measurement Facility	ARMF	UAl _x /Al
Advanced Test Reactor	ATR	UAl _x /Al
Battelle Memorial Institute, Battelle Thermal Reactor	BMI	UO ₂ SS/SS
Boiling Reactor Experiment V	BORAX V	UO ₂ SS/SS
Experimental Breeder Reactor II	EBR-II	U-5% Fissium/SS
EBR-II Scrap	EBR-II, ANL-6	U-5% Fissium/SS
Enrico Fermi Atomic Power Plant (SS Clad only)	Fermi I Blanket	UMo/SS
High Flux Beam Reactor	HFBR	UAl _x Al cermet/Al U ₃ O ₈ Al cermet/Al
University of Missouri Research Reactor	MURR	UAl _x /Al
Oak Ridge Reactor	ORR	UAl _x /Al
Pathfinder	Pathfinder	UO ₂ SS cermet/SS
SM-1A Army Reactor	SM-1A	UO ₂ SS/SS
Systems for Nuclear Auxiliary Power	SNAP	UZrH _x /declad
Organic Moderated Reactor Experiment	SPEC (OMRE)	UMo/SS
Special Power Excursion Reactor	SPSS (SPERT)	UO ₂ SS/SS
Kansas State, Cornell, Texas, Hanford, MSU, and GA TRIGA SNF with Al cladding	TRIGA Al	UZrH _x /Al
Berliner Experimenter Reactor II	TRIGA Ber II	UZrH _x /SS
Neutron Radiography Reactor	TRIGA FLIP	UErZrH/SS
MSU, GA, and Berkeley TRIGA SNF w/SS cladding	TRIGA SS	UZrH _x /SS
University of Washington Argonaut Reactor	UW	UAl _x /Al
Vallecitos Boiling Water Reactor	VBWR (GENEVA)	UO ₂ SS/SS

1.3 APPROACH

The approach taken to evaluate dry storage for the SNF fuels in the ICPP involves determining or estimating the current condition of the SNF, applying the existing data base of dry storage experience, and factoring in the guidelines that may be set by local, state, and national governments. Each aspect in this approach is outlined below, although acceptance by governing bodies is not the focus of this particular study.

1.3.1 General Approach

Under ideal situations, the current condition of the SNF would be determined by examination of a statistically adequate selection of fuels and conditions and by using predictions based on detailed historical information. The approach to evaluating future dry storage for SNF in the ICPP basins consists of the following sequence:

- Obtain available historical information to derive an understanding of the configurations, materials, and current wet storage conditions of the SNF identified in Table 1.1.
- Apply information from other sources, such as published corrosion data, to augment this understanding.
- Identify fuel types that are expected to represent the most difficulty in the transition from wet to dry storage.
- Identify fuel conditioning issues that will need to be defined to select processes that are safe and effective for the transition to dry storage.
- Provide preliminary guidance regarding future characterizations of the ICPP fuel inventories that will support demonstration programs and resolve the conditioning issues.

The type of information that is needed for determining appropriate dry storage for the SNF in the ICPP includes data on fuel element design, past irradiation and storage environments, known physical condition of the spent fuel, and projected condition when it is moved to dry storage. Specific information that would prove valuable in making performance predictions is listed below for each phase affecting the SNF at the CPP:

Fuel Fabrication

- Fuel type (metal, oxide, hydride, mixed oxide, cermet); cladding material (Al, SS) and elemental composition; special cladding or fuel treatments prior to irradiation (metallurgical condition); initial surface films; type of end-cap enclosure technique (welded, brazed); fuel vendor and year fabricated; fissile and nonfissile content, enrichment, and chemical composition of fuel; and internal gas pressure, if any.
- Physical dimensions of fuel elements (inside and outside diameters, lengths); general fuel geometry (pellet, vipac, rods); amount and type of oxides formed, and crevices in the design.

In-Reactor Operation

- End-of-life fuel burnup, neutron fluence, operating temperatures for cladding and fuel; coolant type (water, sodium, organic); known condition of cladding (oxides, crud, pitting, cracks); rod gas pressure;

coolant chemical additives; and time, temperature and characteristics of interim storage at the reactor site.

Shipping and Handling

- Duration, condition, type (wet or dry), and effect of shipping and handling on spent fuel elements, including any mechanical damage from these operations.

Wet Storage

- Water chemistry history (water impurity species and concentrations, pH, chemical additives, radiochemical content history to indicate fuel degradation); undissolved solids (sludge accumulation on fuels); storage duration; and temperature history and trends.
- Known spent fuel condition (general and pitting corrosion, number of failed elements, hydriding, surface condition, general nature of any failed cladding); and related corrosion data (include method used to obtain data, e.g. destructive exams, nondestructive exams, visual exams, and coupon data).
- Storage configuration: design of containers used for storage, including physical dimensions, orientation, materials of construction, and degradation.

Because the SNF in the ICPP was received from a variety of sources over a long period of time, the available data range significantly in type and amount. There is little readily available information specific to the condition of the SNF prior to and after each operational phase preceding receipt at the ICPP, such as fabrication, irradiation, at-reactor-storage, and interim storage. This has resulted partly because many of the SNFs were expected to be reprocessed and details on the SNF condition, such as cladding integrity, oxide thickness, irradiation and storage temperatures, and other information were not passed along with the fuel receipt criteria, some for security reasons. This is contrary to most investigations supporting dry storage of commercial SNF for which the fuel types are limited; design and material conditions are known for the fuels being stored, defected fuel is very limited and usually excluded, and the quantity of commercial SNF is so large that testing and demonstrations on dry storage are more cost effective. Thus, evaluation of the condition of SNF in wet storage at the ICPP and its impact on future dry storage must rely on open literature publications, shipping records, and communications with staff from the companies that fabricated, tested, or shipped the nuclear fuels.

Where historical information is not available, estimates of the SNF condition and its impact on dry storage may be possible using computer codes or algorithms to predict the tendency of material reactions and by using other analytical techniques to estimate the possible performance of the SNF in dry storage. Some of these methodologies were developed for commercial spent fuels while other analyses are more generic in nature. The models may require verification for the more complex spent fuels of this study if they are needed for more than estimates of behavior.

1.3.2 Relevant Storage Experience

Dry storage is a widely applied technology for commercial spent fuel, involving numerous fuel types and several dry storage concepts (Schneider et al. 1992). A brief summary of the status of dry storage for UO_2

commercial SNF with Zircaloy cladding is provided in Table 1.2. Representative summaries supporting this experience can be found in Gilbert et al. (1988) and Johnson et al. (1983) for the United States experience and in Schneider and Mitchell (1992) for foreign experience. The basic elements of dry storage have been demonstrated and licensed for commercial spent fuels, including drying, shielding, thermal analysis, criticality, cover gas solutions, and sealing in high-integrity containers. Much of this technology has already been applied at INEL (Anderson and Meyer 1980). However, numerous fuel types now stored in the CPP-603 and CPP-666 facilities do not have prior dry storage precedents and need to be evaluated to determine whether the transition from wet to dry storage has special issues not previously addressed with fuel types for which there is experience.

In order to focus on relevant information, it is important to gain a quick overview of the scope of information needed. This can be accomplished by reviewing the principal facts about the SNF being studied. The fuel elements have uranium-fissium or metal fuels that include uranium (e.g., UAl_x). Some fuel claddings are known to be breached, exposing the metallic fuels to aqueous corrosion that could include generation of uranium oxides and hydrides. Another issue important to dry storage is that some Al cladding has been subject to substantial corrosion, generating corrosion products that contain water of several types: adsorbed, absorbed, waters of hydration, and possibly water entrained in the fuel at cladding breaches. How effectively the water inventories can be removed in drying cycles, and long-term effects of residual water (e.g., hydrogen generation due to radiolysis), need to be understood for all the fuel types. As details of the histories and configurations of the fuel inventories in wet storage at ICPP are defined and understood, other issues important to the wet-dry transition may emerge. Another important source of information is the experience base for related metallic fuels in wet and dry storage being studied at other DOE sites or in foreign countries that are relevant to the issues applicable to the fuels in the ICPP inventories. This experience for wet and dry storage related to metallic fuels is summarized below.

Metallic Fuels in Wet Storage

Facilities that contain spent fuel with metallic uranium in wet storage are listed in Table 1.3. In all of these cases, the fuels are subject to degradation that has exposed the metallic fuels to aqueous corrosion. Based on a broad experience base, potentially pyrophoric uranium hydrides are expected to be included in the corrosion products of these degraded fuels (ITA 1994). However, the hydrides will be dispersed in UAl_x and cermet fuels, mitigating concerns about ignition. Hydride formation in EBR-II fuel depends on leakage of moisture to the inside of the SS cans. No corroding metallic fuel has been moved from wet storage to long-term dry storage thus far, but methods are being investigated to handle and condition the corroding fuels to avoid pyrophoric events and stabilize the fuels for long-term dry storage (ITA 1994).

Table 1.2 Summary of Dry Storage for Zircaloy-Clad UO₂ Commercial SNF

Aspect	Status
Status of fuel in wet storage	Stable in failed and unfailed forms
Relevant dry storage experience	Demonstrations and licensing in Canada, Germany, Switzerland, and U.S.
Handling, emplacement and retrieval	Metal casks, dry wells, concrete canisters, and vaults
Drying technology	Vacuum drying
Cover gases	Commercial purity He (standard and licensed), Ar, N ₂ demonstrations, air under study
Storage of fuel with cladding defects	Avoided, but acceptable in U.S. with restrictions per 10 CFR Part 72
Shipping experience	Fuel shipped dry prior to dry storage demonstrations
Fuel orientation in dry storage	Horizontal and vertical
Maximum cladding temperatures	Age and pressure dependent; 380°C (716°F) nominal (U.S.)
Potential failure mechanisms in dry storage	Rapid stress rupture Stress corrosion cracking Strain rate embrittlement Slow creep rupture Cladding oxidation only under air Hydride Reorientation Fuel oxidation only under air Incipient defects
Consequences of cladding failures in dry storage	Release of fission gas if pinholes; assessments indicate that site boundary limits not exceeded even if all fuel rods fail Large failures that release fuel particles from cask are not credible
Containment barriers (in addition to fuel and cladding)	Metal casks: double lid NUHOMS™: SS canister Vault: SS canister inside sealed storage tube
Fuel failure in dry storage	A few Zircaloy-clad rods in demonstrations
Surveillance procedures	Metal cask: redundant sealed covers NUHOMS™: visual Vault: Cover gas monitoring

Determining levels of conditioning that will be needed for SNF in the ICPP inventory is an essential aspect of managing the transition of the fuels to dry storage.

Metallic Fuels in Dry Storage

Some types of metal fuels have been or are currently in dry storage as shown in Table 1.4. Of particular interest is fuel storage experience in the United Kingdom (UK), France, Japan and United States. How this experience relates to the fuels in this study will be illustrated.

The magnesium-clad Magnox fuel has been stored at the Wylfa power station in the UK since 1972. The fuel goes directly from the gas-cooled (CO_2) reactors to dry storage without being wetted, avoiding aqueous corrosion of the cladding and metallic uranium fuel. However, water in-leakage through the vault roof caused serious corrosion damage to several canisters of fuel (Bindon 1992). A conditioning treatment has been devised to deal with the corroded fuel. At other Magnox reactors, the fuel is discharged to wet storage, except for fuels that fail in-reactor. These frequently are placed in canisters with mechanical seals and are stored in the pools. Occasionally, the seals leak or the cans are penetrated by corrosion, permitting water to enter the cans and attack the exposed uranium. Pyrophoric events have occurred when the cans were opened to air (Fisher and Knight 1993). The integrity of the unfailed fuel is maintained by suitable water chemistry control until the fuel is reprocessed (Tyfield 1988)

Extensive handling of corroded uranium metal fuel under water at Hanford and in the UK has not produced any pyrophoric events. The pyrophoric events that have occurred in the UK have involved exposure of metal fuel with accessible hydrides to air (Fisher and Knight 1993). Hydrides that are occluded in the oxide or metal are not likely to ignite unless they become exposed by some mechanical event or sometimes by drying the fuel in air.

Processes to condition hydrided fuel to decompose the hydrides have been devised in the UK and in France. Currently, the processes are considered proprietary, but initiatives are under way to obtain the process information. Meanwhile, processes are being proposed to condition the corroded N-Reactor fuel (ITA 1994), which could have application in treating certain fuels in the ICPP inventories. However, unless EBR-II fuels have corroded from possible water leakage into the SS canisters or there was sufficient moisture in the canisters during sealing, hydride conditioning may not be an issue with the ICPP spent fuels. In the case of the N-Reactor fuel, one prominent strategy is to treat the fuel as though all elements are failed. This obviates an intensive campaign to separate failed from unfailed fuel (ITA 1994). Some spent fuels in the ICPP are known to be substantially degraded (e.g., ICPP-603 aluminum plate fuels). However, if fuel histories and appropriate inspections indicate that a specific fuel type is not seriously degraded, the fuel could be dry stored with minimal conditioning, e.g., drying and sealing in an appropriate cover gas.

Table 1.3 Examples of Metallic SNF in Wet Storage (ITA 1994)

Site	Fuel Type	Cladding	Fuel Material	Duration ^(a)	Fuel Conditions
Savannah River Plant (SRP)	Defense	Al Alloy	U Metal	~5 yr	Exposed U Corroding
Tomsk/Krasnoyarsk (FSU)	Defense	Al Alloy	U Metal	up to 2 yr	Some Pitting
INEL	Test Reactor	Al Alloy	U Metal and Alloy	up to 20 yr	Exposed U Alloy Corroding
UK Magnox Reactors	GCR	Mg Alloy	U Metal	up to ~1 yr	Exposed U Corroding
Cadarache and Marcoule, France	GCR	Mg Alloy	U Metal	up to 15 yr	Exposed U Corroding
Sellafield, UK	GCR	Mg Alloy	U Metal	up to ~15 yr	Exposed U Corroding
Hanford	N-Reactor	Zircaloy	U Metal	Up to 23 yr	Exposed U Corroding

FSU: Former Soviet Union

(a) As of 1994

Table 1.4 Metallic Uranium Fuel in Dry Storage (ITA 1994)^(a)

Site	Fuel Type	Cladding	Fuel Material	Cover Gas	First Dry Storage	Fuel Condition
Wylfa (UK)	GCR	Mg Alloy	U Metal	CO ₂ and Air	Since 1972	Generally good; some corroded due to water in-leakage
Tokai (Japan)	Test Reactor	Al Alloy ^(b)	U Metal	He	Since 1982	Good condition when placed in inert dry storage
Chalk River (Canada)	Not Available	Al	U Metal	sealed tube ambient air	Since early '60s	Not Available
White Shell (Canada)	Not Available	Al and Zr	U Metal	concrete canisters double concrete sealed + He and Air	Since mid '80s	Some pinhole defects <0.5% of fuel); no indications of f.p. releases
PUREX (U.S.)	N-Fuel	Zircaloy	U Metal	Air and Occasional moisture	Since 1969	Not Available
Lucas Heights (Australia)	Test Reactor	Al Alloy	U Metal	N ₂ ^(c)	Since ~1965	^(d)
UW (U.S.)	Test Reactor	Al Alloy	U Al _x	Not Available	1970 to 1988	Not Available

(a) Personal communication with W.P. Miller of the University of Washington, 1994.

(b) Fuel in wet storage up to 20 yr; air-dried 1 week in hot cell; 45°C (113°F) Max T; RH<20% to prevent Al cladding corrosion; fuel is seal welded in SS canister with He cover gas.

(c) Stored in N₂ atm in sealed cans in fuel storage pool; recent exam indicates much cladding corrosion; sent to BNFL for reprocessing (1990); cans opened under water; no pyrophoric events.

(d) Stored in 15.2-m (50-ft) deep dry wells. Nitrogen backfill gas monitored for ⁸⁵Kr. Indications of ⁸⁵Kr found in one dry well where fuel that had been cut for metallography is stored.

The test reactor fuel at Tokai, Japan represents another interesting case. The fuel was judged free of cladding defects. It was dried in air in a hot cell for one week at 45°C (113°F). It was then seal-welded into SS canisters in a helium cover gas (Adachi 1982; Adachi et al. 1984). This experience has potential relevance to ICPP fuel inventories, if sufficient evidence exists that specific fuels are free from cladding defects and corrosion products with large water contents that could be sources of troublesome amounts of radiolytic hydrogen in long-term dry storage.

The University of Washington (UW) fuel, now stored in CPP-666, had periods of dry storage during reactor outages (water replaced by air in the core) and after removal from the reactor. The burnup level was very low, so decay heats and corresponding temperatures are also low. No evidence of fuel degradation was observed in television inspections prior to shipment to CPP-666. This and the foregoing descriptions of experience with spent fuels illustrates the value of a systematic assessment of fuel history and conditions for comparison with existing experience on wet and dry storage for other relevant fuels. Such an evaluation will assist in defining the current condition of the fuel and its potential impact on moving the spent fuel to dry storage on a basis that is both safe and cost effective.

1.3.3 Regulatory Criteria

While the primary focus of this study is to address the technical issues of dry storage for the ICPP SNFs, it is prudent to mention potential criteria that may affect these operations. Thus, this summary of governmental criteria is provided as a measure of the types of information that may be needed and the requirements that might have to be met for dry storage of SNF at the ICPP.

Protection of the public and the environment is the basis for a body of Public Law that has grown up to address a host of man-made hazardous materials, emissions and wastes, both nonnuclear and nuclear. Taken together, these laws form the basis for regulatory domains of the U.S. Nuclear Regulatory Commission (NRC), the U.S. Environmental Protection Agency (EPA), DOE, and other agencies which govern all the aspects of nuclear materials handling. These major governing laws are:

- Atomic Energy Act of 1954 (AEA). This act specifies the framework for the safety and licensing of nuclear facilities and activities involved in the management of source, special nuclear and byproduct materials; the Energy Reorganization Act of 1974 identifies the functions of the NRC.
- The Clean Air Act of 1963 (CAA and amendments). This act provides regulatory standards for all toxic or hazardous air pollutants under the National Emission Standards for Hazardous Air Pollutants, for which 40 CFR Part 61 is the EPA interpretation and DOE Order 5400.5 is the DOE guideline.
- The National Environmental Policy Act of 1969 (NEPA, Public Law 91-190 and amendments). The basic tenet of NEPA is 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 and amendments). This act regulates waste, which must meet two criteria: 1) it must be solid, and 2) it must exhibit certain hazardous characteristics (interpreted in EPA's 40 CFR 261). RCRA establishes a cradle-to-grave regulatory program for present hazardous waste activities.
- The Clean Water Act (Federal Water Pollution Control Act Amendments of 1977, FWPCA, and amendments). This act is concerned with surface water and, most applicable to waste disposal container and disposal site acceptability, drinking water (and its sources).
- The Nuclear Waste Policy Act of 1982 (NWPAA, Public Law 97-425 and amendments; successor to the Atomic Energy Act of 1954). The NWPAA 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 (CERCLA, Public Law 96-510) of 1980; and the Superfund Amendments and Reauthorization Act of 1986 (SARA) and amendments. This act establishes a comprehensive response program for past hazardous substance activities.
- The Federal Facilities Compliance Act of 1992 (FFCA). This act mandates the development plans for treatment capabilities for mixed wastes at each DOE site where mixed wastes are generated or stored.

The primary interpretation of the governing legislation, as applicable to nuclear materials, is provided in the Code of Federal Regulations (CFR) by NRC, EPA, and the Department of Transportation (DOT). DOE Orders are supplementary to the CFR, frequently written for specific application to DOE tasks.

Note, however, that SNF is in a materials class all its own as far as most of the regulations are concerned. SNF is not "waste" (except in EPA's 40 CFR 261), inasmuch as the determination has not been made that it has no further usefulness and it is not yet "isolated in a disposal system"; it is not HLW unless it has been dismantled and reprocessed (except in NRC's 10 CFR 60). Although it contains TRU and fission products, SNF is not classified as either. The SNF of concern in this study is the property of DOE and is primarily noncommercial; hence, the NRC regulations governing commercial fuel do not strictly apply for the noncommercial SNF. There could be a case for commercial-like SNF falling under these guidelines, which could make their storage easier in some respects. Although SNF contains radioactivity which can be hazardous and may also contain RCRA listed elements such as barium, silver, and selenium, only the RCRA-defined "hazardous waste" portions of the spent fuel material or mixtures of SNF with other hazardous materials have been considered to be regulated; these mixtures continue to be discussed with respect to applicability under RCRA and/or CERCLA. Nevertheless, criteria are needed for activities leading to the interim storage and ultimate disposition of SNF. For this reason, the requirements of the NRC, EPA, and DOE for regulating TRU, HLW, and low-level waste (LLW) have been reviewed for their relevance to dry storage of SNF in the CPP. A systems engineering study has been performed by DOE, resulting in a draft document entitled "Spent Nuclear Fuel Requirements Document (Rev. B)," date July 6, 1994. This document may be the forerunner of comprehensive SNF management mandates.

The regulations providing the most applicable criteria for SNF are given in Table 1.5. Some of the more pertinent details in each cited regulation are given in Appendix A. The summaries and excerpts presented are not exhaustive, however, and should be used as general guidelines only. Many of the regulations are extremely long and complex and contain many exemptions, modifying clauses, and special cases which can be sought directly, as needed.

In general, regulations cover radiation protection for workers and the general public; requirements for transporting various categories of radioactive materials without exposure to the public (with implications that the contents be characterized); requirements for the siting, construction, and monitoring of storage and disposal facilities that will be safe for the foreseeable future and beyond; and requirements that ensure nuclear materials facilities and operations remain benign to the environment. In Appendix A, the citations are grouped according to the following topics, which take the SNF from its present location through any necessary "stabilization" treatment to storage and ultimate disposition: transportation, treatment and storage, disposal, general management, and lists of hazardous materials.

Table 1.5 List of Regulations Reviewed

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 Spent Nuclear Fuel and High-Level Radioactive Waste (NRC)
40 CFR 61	National Emission Standards for Hazardous Air Pollutants
40 CFR 191	Environmental Radiation Protection Standards for Management and Disposal of Spent Nuclear Fuel, High-Level and Transuranic Wastes (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
DOE Order 6430.1A	General Design Criteria

2.0 STORAGE FACILITIES AND SPENT FUEL DESCRIPTIONS

The focus of this assessment is on SNF stored in two facilities, the underwater fuel storage facility, CPP-603, and the underwater fuel storage area, CPP-666. A description of the two facilities, including their general design and storage history is provided in Section 2.1. Details on design, irradiation history, and summary information on the storage experience for the SNF being studied are provided in Section 2.2. This information provides part of the basis for understanding the current condition of the SNF in the ICPP storage pools and is used along with additional information in subsequent chapters of this report to assess potential failure mechanisms that may affect dry storage.

2.1 ICPP STORAGE POOL DESCRIPTIONS

To understand past and future impacts of SNF storage in these facilities on the SNF, it is important to understand the histories, characteristics, and environments of the two storage pools. This information is briefly summarized in this section, including storage and handling impacts on specific fuels in the current CPP-603 and CPP-666 inventories.

2.1.1 Design and Storage History of the CPP-603 Pool

The CPP-603 is the oldest of the two facilities, having stored SNF since 1951. It consists of three unlined concrete pools (north, middle, and south) filled with water and holds 41 different types of SNF received as early as 1959 (DOE 1993b). The CPP-603 pools connect to form an E shaped basin, as shown in Figure 2.1. Two of the pools store fuel in buckets or cans suspended from a monorail system. The other pool stores fuel in Al and SS racks. The fuel types stored in this pool include metals, alloys, hydrides, and oxides in several different fuel matrices. The majority of the SNF is clad with Al, SS, and Zircaloy, but some SNF is unclad or known to be defected.

The major events in the CPP-603 storage pool are shown in a timeline in Figure 2.2. Based on the design and sequence of events in the storage environment, the key considerations in evaluating this environment include the following points:

- Long service time (~40 yr in 1994)
- Unlined concrete surfaces and possibly radionuclide diffusion through the pool wall
- Development of an aggressive pool chemistry due to addition of algicides (up to ~800 ppm chloride); algicides included calcium hypochlorite, chlorine, and iodine
- Nitrate inhibitors introduced to mitigate corrosion (NO_3/Cl^- between 3:1 and 5:1)
- Once-through water flow until 1966

Figure 2.1 Schematic of CPP-603 Fuel Storage Basin (Hoskins et al. 1993)

Figure 2.2 History of the CPP-603 Pool

- Use of reverse osmosis and occasional water replacement to lower the impurity levels
- Ultraviolet lights installed to treat inlet water, mitigating algae growth
- Chloride levels are about 50 ppm in 1994.

Corrosion characteristics of materials exposed to CPP-603 pool waters have been provided by Dirk (1994), DOE (1993b), Hoskins et al. (1993), and Lundberg (1994). Aluminum and CS materials have been susceptible to major corrosion in the aggressive CPP-603 water. Corrosion occurred on the original CS monorail system, but the system is being replaced by SS (Hoskins et al. 1993). Uniform corrosion of aluminum has generally not been excessive, but pitting attack has been a major phenomenon (Dirk 1994); SS and Zircaloy have shown little or no evidence of attack. The materials in use in the CPP-603 basin and their general condition are summarized in Table 2.1. Various storage configurations and conditions important to this study are listed in Table 2.2.

The chloride levels in the pool are of particular interest since high chloride content can be aggressive on many materials. As explained above, the chloride level increased, in part because of an effort to mitigate algae growth, but also because the water system was changed from a once-through to a recirculating system, which allowed the buildup of contaminants. The level of chloride in the water increased significantly in 1976, as shown in Figure 2.3, but the levels were reduced to the current 50 ppm level after 1983 or 1984. A combination of nitrate additives to reduce the effect of chloride on storage components, addition of new water, and use of ultraviolet lights to prevent algae buildup have been used to control water condition (Hoskins et al. 1993). The history of high chloride content in the CPP-603 water is compared in Table 2.3 with the earliest receipt date for the various SNF being evaluated.

Prior storage conditions, facility design, and handling operations all impact the condition of spent fuel. The spent fuels that are stored in the CPP-603 pool developed corrosion films during reactor service that may offer substantial protection from the CPP-603 environment. However, handling is likely to have caused local damage to the films, exposing metal that would be susceptible to pitting. Operations have possibly resulted in mechanical damage, including surface damage (DOE 1993b). For example, the ATR fuel elements are handled with a bayonet device inserted between fuel plates, which results in mechanical damage to fuel plates that seems to result in accelerated local corrosion (Lundberg 1994).

As indicated in Table 2.2, numerous dissimilar metal contacts exist in the CPP-603. Crevices exist both within and between fuel elements and other metal components (buckets, racks). There is evidence of galvanic effects on corrosion in CPP-603 and even between Al-alloys of different compositions in other facilities (Howell et al. 1993).

Table 2.1 Materials in CPP-603 Storage Pool

<u>Fuel/Component</u>	<u>Materials</u>	<u>General Condition</u>
Fuel Cladding	Al Alloys, SS, Zircaloy	Intermittent storage since 1950; Evidence of corrosion in Al alloys and SS; No evidence of corrosion in Zr and UZrH
Fuel Materials	UO ₂ , UZrH, UAl, Metallic U (see Table 1.1 for details)	Some cladding breached, exposing fuel to pool water
Basin Structure	Unlined reinforced concrete walls	wetted since 1951
Fuel Buckets and Storage Cans	Al-Alloys, CS, SS	Severe corrosion of Al-alloy and CS components
Monorails	Original: CS Replacement: SS	Severe corrosion of CS
Hangers	Original: CS	Severe Corrosion
Storage Racks	Al and SS	Severe corrosion of Al-Alloys

Table 2.2 Configurations and Conditions in the CPP-603 Pool
Relevant to Corrosion

Galvanic Couples:	<ul style="list-style-type: none"> a) Al-clad fuel in CS, Al, and SS buckets b) SS fuel in CS, Al, and SS buckets c) Zircaloy-clad fuel in SS- and Al-alloy racks d) CS hangers/SS buckets e) SS cans in Al-alloy racks
Crevice:	<ul style="list-style-type: none"> a) Fuel elements adjacent to cans, buckets, and racks b) Crevices within fuel elements
Mechanical Effects:	Mechanical damage from dropping, bumping, scraping, scratching, etc.
Vibrational Effects:	Fretting or wear from vibrations (if any)

Figure 2.3 Chloride Concentration in CPP-603 Water at Six-Month Intervals (Hoskins et al. 1993)

Because only portions of the storage facility's history and the current condition of the SNF is known, the assumptions shown in Table 2.4 have been made to provide a starting point for the evaluation of the current condition of the SNF. Future characterizations should be conducted to verify some of these assumptions and others that may be needed to confirm the existence of crevices, galvanic couples, and general condition of the fuel and storage components.

In summary, the fuels addressed in this study that are exposed to the CPP-603 waters have been subject to relatively aggressive conditions. Fuels that are sealed in canisters and stored in the pool would offer some protection from the aggressive environment if the canisters have not leaked. Because the fuels are vulnerable to degradation in water or even moist gases, assurances are needed that leakage has not occurred before decisions are made regarding how the fuels are moved to dry storage.

Table 2.3 Apparent Maximum Chloride Content for Storage of
Selected SNF in CPP-603 Based on Receipt Date

Spent Fuel Source CPP-603	Yr Receipt Started	CPP-603 Max. Chloride Content
ORR	1985	50 to 100 ppm
ATR	1985	50 to 100
ARMF	1987	N/A
TRIGA AI	1973	800
MURR	1985	50 to 100
HFBR	1986	50 to 100
TRIGA FLIP	1979	300
SPEC or ORME	1959	800
SM-1A	1971	800
EBR-II	1978	350
EBR-II, ANL-6 Test	1982	350
Pathfinder	1971	800
BMI	1961	800
SPSS or SPERT	1959	800
APPR or AGE-2	1960	800
Fermi I/II SS	1988	N/A
TRIGA SS	1982	200
VBWR	1961	800
BORAX V	1970	800
TRIGA Ber II	1982	N/A
SNAP	1966	800
AI	1974	800

Table 2.4 Assumptions for Spent Fuel Assessment

Pool Chemistry:

Parameters	CPP-603	CPP-666
pH	5.0 to 8.5	5.5
Cl ⁻	up to 800 ppm (see Figure 2.3)	30 ppb
Temperature	20°C (68°F)	20°C (68°F)
Conductivity	600 μ S/cm	1 μ S/cm

Assume that conditions of all pools have the same chemistry in a given building.

Duration of Storage Conditions:

Assume the oldest fuel stored in the pool for a given type to estimate degradation.

Radiation Field:

100 R/hr or less for all spent fuels, unless otherwise indicated.

Fuel Integrity:

All storage containers and cladding are assumed to be failed at some point in wet or dry storage in order to assess the impact of such failures on continued degradation and environmental release issues.

Heat Load:

Currently less than 20 to 30 watts/element, based on ATR elements.

2.1.2 Design and Storage History of CPP-666 Pool

The CPP-666 is a modern underwater fuel storage facility that was placed in operation in 1984 and has six SS-lined storage pools with SS and Al storage racks and equipment for monitoring and treating the water. A plan view of the facility is shown in Figure 2.4. Sixteen types of SNF are stored in CPP-666 with various combinations of SS, Al, and Zircaloy cladding and fuel matrices. This facility will continue interim storage of SNF until other options exist; plans are under way to move portions of the SNF from both the CPP-603 and CPP-666 to dry storage within the next six years (Hoskins et al. 1993).

The pool chemistry in CPP-666 is more benign than that of the CPP-603 as shown in Table 2.4, but the CPP-666 is not free of corrosion. While the base metal and welds of the steel basin liner do not have pitting and have a general corrosion rate of 3.2 μ m/yr (0.13 mpy), some pitting has occurred on Al coupons.

Figure 2.4 Plan View of CPP-666 Storage Facility (Hoskins et al. 1993)

Algae have been observed, which causes concern that MIC is active (Hoskins et al. 1993). This evidence is in contrast to very minor corrosion indications on Al-alloy fuels and components over long-term residence in other benign pools, e.g., the RBOF pool at the Savannah River site described in Lundberg (1994). For example, an Al-alloy spacer that had been adjacent to an irradiated Zircaloy-clad assembly was examined by metallography at PNL and showed only about 25 μm (1 mil) of oxide and similar pit depths after 15 yr in the RBOF pool (Kustas et al. 1981). Investigations are under way to determine whether the unexpected degree of pitting on Al alloys in CPP-666 is due to MIC (Hoskins et al. 1993).

2.2 SPENT FUEL DESCRIPTIONS

The spent fuels that are part of this study and are currently in wet storage are described briefly in this section. This study is concerned primarily with four types of SNF: uranium-aluminum (UAl_x), uranium oxide (UO_2), uranium metal, and uranium zirconium hydride (UZrH_x) (see Table 1.1). The UAl_x fuel is generally clad in Al; the UO_2 fuel and U metal are clad in SS; some of the UZrH_x fuel is clad in Al, SS, or has had the cladding removed. The information most pertinent to the transfer of the SNF to dry storage is compiled into three tables: current wet storage data (Table 2.5), selected fuel design data (Table 2.6), and irradiation history (Table 2.7). In many cases information is not available to place the fuel into interim storage or the repository.

As shown in Tables 2.5 through 2.7, there are numerous types of fuels that need decisions regarding optimum interim storage. Within each SNF type there are further variations, such as in physical configuration, fuel matrix, fuel enrichment, additional materials in the fuel matrix, and cladding thickness. The physical configurations include rods (largely UZrH_x and uranium metal-type fuel); curved or flat plates (UO_2 , UAl_x and uranium metal type fuel); and concentric tubes (UO_2). The rods, plates, or tubes are then grouped in different ways to form the elements (rods, pins), assemblies, or bundles required for each reactor design. The fuel matrix which forms the body holding the fuel particles in the cermet fuels is most frequently SS or Al, although molybdenum is used in one case for uranium metal plates and in one case for rods. Uranium fuel enrichment ranges from depleted to 93% or more ^{235}U . Erbium, samarium, graphite, molybdenum, EuO_2 , ZrB_2 , B_4C , Si, TiO_2 , and fission (a mixture of selected elements typically produced during radiation) are among the materials added to the fuel matrix for specific design purposes. The documented cladding thicknesses range from 124.5 to 762 μm (4.9 to 30 mil). The information is not complete for most of the fuels; additional data will be required for several spent fuels to complete the evaluation of SNF durability in wet and dry storage. The fuels in the ICPP inventory encompass a much more challenging range in fuel designs, materials, and storage conditions than had to be addressed in evaluating dry storage of commercial LWR fuels. However, much is applicable from that experience and more limited experience with metal fuels.

Table 2.5 Information on Current Storage for Selected SNF at ICPP

Fuel Designation	Storage Facility	Storage Location	Earliest Date Received	How Stored	Number Stored	Storage Container Material	Storage Configuration	Container Degradation
ORR	CPP-603	South Basin	1985	bare	17		9 Al buckets	N/A
UW	CPP-666	Pool 6	1990	bare	26	N/A		N/A
ATR Assemblies	CPP-666	Pool 6	1985	bare	760		Al basket	N/A
ARMF	CPP-666	Pool 6	1987	bare	15		Al basket	N/A
TRIGA Al	CPP-603	Middle Basin	1973	bare ?	578	Al buckets	125 Al buckets	N/A
MURR	CPP-666	Pool 6	1985	bare	32	N/A	Al buckets, racks	N/A
ATR Assemblies	CPP-603	South Basin	1985	bare	128		Al basket	N/A
HFBR	CPP-603	South Basin	1986	bare	20		Al buckets	N/A
HFBR	CPP-666	Pool 6	1986	bare	220		Al buckets	N/A
MURR	CPP-603	South Basin	1985	bare	24	N/A	Al buckets, racks	N/A
TRIGA FLIP	CPP-603	South Basin	1979	Al can	2	Al	2 positions in SS racks	corroded
SPEC (OMRE)	CPP-603	Middle Basin	1959	Al can	1	Al		heavy corrosion

Table 2.5 Information on Current Storage for Selected SNF at ICPP

Fuel Designation	Storage Facility	Storage Location	Earliest Date Received	How Stored	Number Stored	Storage Container Material	Storage Configuration	Container Degradation
SM-1A	CPP-603	Middle Basin	1971	SS can	93	SS	47 SS buckets	no visible corrosion
EBR II (ANL-6 Test)	CPP-603	South Basin	1982	SS can	4	SS		no visible corrosion
Pathfinder	CPP-603	Middle Basin	1970	bare	417	N/A	14 SS monorail buckets	N/A
BMI	CPP-603	Middle Basin	1961	SS and/or Al can	3	SS/Al		moderate corrosion; an Al container corroded away
SPSS (SPERT)	CPP-603	Middle Basin	1959	Al can	1	Al	1 bucket	heavy corrosion
APPR (AGE 2)	CPP-603	Middle Basin	1960	CS can	1	CS		heavy corrosion
Fermi I Blanket	CPP-749 (only dry storage in this study)	Dry Well	1974	fuel basket container	510	SS 316		N/A
TRIGA SS	CPP-603	Middle Basin	1982	bare	263		62 SS buckets	N/A
VBWR (Geneva)	CPP-603	Middle Basin	1961	Al can	4	Al		heavy corrosion

Table 2.5 Information on Current Storage for Selected SNF at ICPP

Fuel Designation	Storage Facility	Storage Location	Earliest Date Received	How Stored	Number Stored	Storage Container Material	Storage Configuration	Container Degradation
EBR-II	CPP-603	South Basin	1978	SS 304 can	2164	SS 304		no visible corrosion
BORAX V	CPP-603	Middle Basin	1970	bare	36		13 SS MTR type buckets	N/A
EBR-II	CPP-666	N/A	1978	SS 304 can	1472	SS 304		no visible corrosion
TRIGA Ber II (Germany)	CPP-603	South Basin	1982	bare	21		canister	N/A
SNAP	CPP-603	South Basin	1966	Al can	19	Al		heavy attack and corrosion
SNAP	CPP-603	Middle Basin	1966	Al can	19	Al		heavy attack and corrosion
Al	CPP-603	South Basin	1974	Al can	12	Al		heavy attack

Table 2.6 Design Information for Selected SNF in Storage at ICPP

Fuel Designation	Fuel Type	Fuel Matrix	Other Materials	Form	Cladding Material	Clad Treatment	Fuel Vendor	Year Fab'd	Clad Thickness
ORR	UAl _x			element	Al				
UW	UAl _x	Al		plates in a bundles	Al		Martin Marietta, and Babcock and Wilcox		
ATR Assemblies	UAl _x	Al	B ₄ C, Sm	assembly 19 curved plates	Al 6061, 1100	Metallurgical bond	United Nuclear Corp.		0.04 cm (15 mil), several layers
ARMF	UAl _x	none	Al	plate	Al 1100				
TRIGA Al	UZrH _x	none	Mo, Sm, graphite	cyl. rods	Al	annealed, cold-deformed, work-hardened			0.08 cm (30 mil)
MURR	UAl _x	Al powder		plates in an assembly	Al 6061, 1100				
ATR Assemblies	UAl _x	Al	B ₄ C, Sm	assembly 19 curved plates	Al 6061, 1100	Metallurgical bond	United Nuclear Corp.		0.04 cm (15 mil), several layers
HFBR	UAl _x , U ₃ O ₈	Al powder cermet	none	assembly	Al 6061				
TRIGA FLIP	UErZr H _x		Erbium	3.6 cm (1.42 in.) rod	SS 304				0.05 cm (20 mil)

Table 2.6 Design Information for Selected SNF in Storage at ICPP

Fuel Designation	Fuel Type	Fuel Matrix	Other Materials	Form	Cladding Material	Clad Treatment	Fuel Vendor	Year Fab'd	Clad Thickness
SPEC (OMRE)	U metal or UO ₂	Molybdenum, or SS		subassemblies contained 16 fuel plates	SS		Atomic International (AI)		0.012 cm (4.9 mil)
SM-1A	spherical UO ₂	SS 347 powder	ZrB ₂ , EuO ₂	0.08-cm (30-mil) thick plate; assembly	SS 347		Alco		0.013 cm (5 mil)
EBR-II (ANL-6 Test)	U metal		N/A	chunk, chips and powder	SS				
Pathfinder	UO ₂ cermet	SS 316L	B ₄ C	2 concentric fuel-bearing tubes	SS 316L				0.015 cm (5.9 mil)
BMI	UO ₂	SS		can	SS				N/A
SPSS (SPERT)	UO ₂ type "C" or "P"	SS	0.08-cm (30-mil) thick plate; Al cladding	can - square assembly-made of plates	SS 304 or SS 304L		Phillips Petroleum Co.		0.015 cm (5.9 mil)
APPR (AGE 2)	Geneva UO ₂	302B SS cermet	B ₄ C	0.08-cm (30-mil) thick plate	SS 304		Alco		0.013 cm (5 mil) of 306 SS

Table 2.6 Design Information for Selected SNF in Storage at ICPP

Fuel Designation	Fuel Type	Fuel Matrix	Other Materials	Form	Cladding Material	Clad Treatment	Fuel Vendor	Year Fab'd	Clad Thickness
Fermi I Blanket	depleted ^{238}U w/Mo	N/A	Na bond for cladding	1.12-cm (0.44-in.) dia. rod; 25 rods/assembly	SS 304				0.012 cm (4.9 mil)
TRIGA SS	UZrH_x	none	graphite, Mo, Sm	3.7-cm (1.46-in.) rod	SS 304, 304L	work-hardened ; all cold-deformed			0.05 cm (20 mil)
VBWR (Geneva)	Geneva UO_2 cermet	SS 304L	TiO_2	plate; 6 to 9 plates per assembly	SS 304L				0.013 cm to 0.031 cm (5 to 12 mil)
EBR-II	U metal	none	5% fissium; fuel is Na-bonded to cladding	hexagonal elements containing fuel rods	SS 304L, 316				0.023 cm (9 mil)
BORAX V	UO_2 cermet	SS 304B	Si	Four 0.08-cm (30-mil) thick plates	SS 304L				0.02 cm (8 mil)
TRIGA Ber II (Germany)	UZrH_x		Al powder, graphite	7.6-cm (3-in.) element	SS 347	welded	Euratom		0.06 cm (23.6 mil)
SNAP	UZrH_x		none	can	declad		AI		

Table 2.6 Design Information for Selected SNF in Storage at ICPP

Fuel Designation	Fuel Type	Fuel Matrix	Other Materials	Form	Cladding Material	Clad Treatment	Fuel Vendor	Year Fab'd	Clad Thickness
AI	U ₂ ZrH _x	none	none	can	de clad		AI		

Table 2.7 Irradiation History for Selected SNF Stored at ICPP

Fuel	Burnup	Operating Cladding	Operating Fuel	Cladding Oxide	Corrosion Assessment	Interim Storage
ORR	N/A	?	?	?	?	?
University of Washington	230 elements, <0.3% 87 elements, 0.3-2.6% 19 elements, 2.6-12%	?	66°C (151°F)	< 5 μm (<0.2 mil) estimated	See Table 3.3	about 1 yr dry storage at UW
ATR Assemblies (760)	41%	227°C (440°F) Peak	Reactor Coolant: 76.1°C (169°F) outlet to 54.4°C (130°F) inlet water	?	Corrosion in CPP-603 and CPP-666 pools; breach from pitting corrosion, blistering, penetration of pimple-like surface defects; bayonet mech. dam.; pitting depth 0.05 cm (20 mil) max.	up to 10 yr in ATR reactor and canal
ARMF	?	?	?	?	?	?
TRIGA A1	0.01%	90°C (194°F) (at 100 kW)	Reactor Coolant: Water	None	No pitting; no crud; no cracks.	8 to 20 yr
MURR	1.7 to 2.5 %	N/A	Reactor Coolant: water	0.005 cm (2 mil)	?	160 to 670 days in DI water

Table 2.7 Irradiation History for Selected SNF Stored at ICPP

Fuel	Burnup	Operating Cladding	Operating Fuel	Cladding Oxide	Corrosion Assessment	Interim Storage
ATR Assemblies (128)	41%	227°C (440°F) Peak	Reactor Coolant: 76.1°C (169°F) outlet to 54.4°C (130°F) inlet water	?	In-reactor breach (<0.5%); in-reactor corrosion (>12 m/s, 39 ft/s coolant flow) <10 yr storage in TRA-670 canal; pitting depth 0.05 cm (0.02 in.) max.	up to 10 yr in ATR reactor and canal
HFBR	40.5%	?	?	?	?	?
TRIGA FLIP	0.2% (and 5.7-14.8%)	400 to 1100°C (752°F to 2012°F)	?	?	?	?
SPEC (OMRE)	N/A	427°C (801°F)	?	?	?	?
SM-1A	N/A	316°C to 427°C (601°F to 801°F)	?	?	?	?
EBR-II (ANL-6 Test)	N/A	?	Moderator Water	?	Ends Cut	?
Pathfinder	0.97%	495°C to 760°C (923°F to 1400°F)	?	?	High fuel cladding temperatures can lead to stress corrosion problems	?
BMI	N/A	?	?	?	?	?

Table 2.7 Irradiation History for Selected SNF Stored at ICPP

Fuel	Burnup	Operating Cladding	Operating Fuel	Cladding Oxide	Corrosion Assessment	Interim Storage
SPSS (SPERT)	N/A	485°C (905°F)	?	?	Potential arises for galvanic couple between cladding of fuel and Al can	?
APPR (AGE 2)	33%	316°C to 427°C (601°F to 801°F)	?	?	Fuel elements suffered severe localized attack at brazed joints, inter- and intragranular cracking of SS cladding	?
Fermi I Blanket	0.35%	565°C (1049°F)	?	?	?	?
TRIGA SS	230 elements, 3% 87 elements, 0.3 to 2.6% 19 elements, 2.6 to 12%	400°C to 1100°C (752°F to 2012°F)	?	?	?	8 to 20 yr
VBWR (Geneva)	8%	293°C (559°F)	?	?	Cladding failures occur in all types of SS-clad fuel rods	?
EBR-II	6% (at 25 to 30 GWd/MTHM)	650°C (1202°F) inlet; 2100°C (3812°F) outlet	?	?	?	?
BORAX V	N/A	650°C (1202°F) inlet; 2100°C (3812°F) outlet	?	?	?	?

Table 2.7 Irradiation History for Selected SNF Stored at ICPP

Fuel	Burnup	Operating Cladding	Operating Fuel	Cladding Oxide	Corrosion Assessment	Interim Storage
TRIGA Ber II (Germany)	<0.3% for 230 rods 0.3 to 2.6% for 87 rods 2.6-12% for 19 rods	90°C (194°F) (at 100kW), 60°C (140°F) (at 30 kW), 38°C (100°F) (at 10kW)	115°C (239°F) at 100kW 60°C (140°F) at 30kW 38°C (100°F) at 10kW	?	?	8 to 20 yr in dry storage
SNAP	<1%	?	?	?	?	?
AI	N/A	?	?	?	?	?

In weighing the results of this evaluation of SNF condition, three easily described factors can be considered: 1) the quantity of fuel in storage, 2) the duration of that storage, and 3) the apparent aggressiveness of the storage environment. The quantity of the selected SNF being evaluated in the CPP-603 and CPP-666 storage pools is graphically displayed in Figure 2.5. The maximum duration of storage for these fuels is shown in Figure 2.6 and may be compared with the highest level of chloride that the fuels may have experienced in the CPP-603 storage, as shown in Figure 2.7. The UW plate fuel with Al cladding is a good example of a fuel type with moderate quantities of fuel elements (26) stored in the relatively nonaggressive CPP-666 environment for only four years. In comparison, the EBR-II spent fuel represents slightly over half of the fuel elements/assemblies considered in this study and has been stored for up to 16 years in a fairly aggressive environment of perhaps 350 ppm Cl⁻. The aggressive chloride environment may only be significant if the SS cans containing the EBR-II spent fuel have leaked or had moisture in them at closure. At the other extreme are the SPERT and SPEC fuels that have been stored for up to 35 years in the CPP-603 pool in 800 ppm Cl⁻, but they represent almost a negligible amount of the fuel inventory. These few examples provide an indication of how the quantity, age, and aggressiveness of the fuel environment can be used along with the fuel condition, failure mechanisms, and storage configurations to assess the importance of the fuel conditions on subsequent drying operations and dry storage.

Figure 2.5 Comparison of Quantities of the SNF Being Evaluated in the CPP-603 and CPP-666

Figure 2.6 Comparison of Maximum Storage Duration for SNF in the CPP-603 and CPP-666

Figure 2.7 Maximum Chloride Levels in CPP-603 During Storage of Selected SNF

3.0 ALUMINUM-CLAD SPENT FUELS

Aluminum alloys are represented in many fuels and fuel storage pool components worldwide (Kustas et al. 1981; IAEA 1982, IAEA 1988). Of specific concern in this study are the Al-clad fuels with metallic UAl_x , $UZrH_x$, and U_3O_8 or UAl_x in cermet fuels that have been in extended wet storage (see Table 1.1). These fuels have been stored varying times at the reactors, then transferred to the CPP-603 or CPP-666 pools. The conditioning needed prior to dry storage and the dry storage requirements are driven by the fuel characteristics at the time the fuels will be transferred to dry storage. Thus, current fuel characteristics or estimates of their conditions are made to the extent possible to make judgements on treatment and dry storage requirements.

The review of the condition and failure mechanisms for the Al-clad SNF in the CPP-603 and CPP-666 pools is provided under four major sections: fuel characteristics and failure mechanisms affecting dry storage (Section 3.1), fuel failure mechanisms and their consequences in dry storage (Section 3.2), recommended dry storage conditions (Section 3.3), and recommended spent fuel storage preparation operations (Section 3.4).

3.1 FUEL CHARACTERISTICS AND FAILURE MECHANISMS AFFECTING DRY STORAGE

Available details on the design, storage, and irradiation histories for the seven Al-clad fuels in the CPP-603 and CPP-666 basin inventories that are being reviewed were provided in Tables 2.5 through 2.7. The Al-clad SNF represents about 27% of the fuel inventory addressed in this review and includes the second (ATR) and third [Training, Research, and Isotope Production Reactor - General Atomics (TRIGA) Al] largest quantity of fuel based on the number of units stored (see Figure 2.5). Except for the TRIGA AL fuel, which has been stored for up to 21 years, all of the Al-clad SNF has been stored for less than ten years (excluding storage prior to receipt at the ICPP). As examples, the ATR fuel is typically stored in water for up to ten years prior to shipment to the ICPP, while the UW fuel was stored dry when the reactor was not operational and for about 16 months prior to shipment to CPP-666.⁽¹⁾ The TRIGA Al fuel was stored in the CPP-603 pool during the period of highest chloride levels (800 ppm), while the ORR, ATR, HFBR and MURR fuels have been stored in water with 100 ppm Cl^- or less. Thus, the Al-clad fuels have been subject to broad corrosion conditions.

A significant body of data exists to guide analysis of Al alloys and metallic fuels in wet and dry environments. Information is provided below on the corrosion of Al alloys in water (Section 3.1.1), under reactor conditions (Section 3.1.2), in wet storage facilities (Section 3.1.3), and in the CPP-603 and CPP-666 facilities (Section 3.1.4). This information provides a basis for estimating the condition of the cladding and identifying failure mechanisms for the fuel. Because many of the spent fuels with Al cladding also contain fuel in an Al-alloy matrix, the corrosion characteristics of this metal also provide a basis for evaluation of the fuel material.

⁽¹⁾ Personal communication with W.P. Miller of the University of Washington, July 1994.

3.1.1 General Corrosion Characteristics of Al Alloys

The principal types of cladding and compositions used for spent fuel are indicated in Table 3.1. A review of general corrosion in an aqueous environment of such Al alloys indicated the following characteristics in relatively pure waters (Johnson et al. 1976):

- Aluminum is an active metal, but forms passive films in water and other oxidizing media; oxide spallation is minimized in O₂-saturated water.
- Dissolved oxygen in water generally improves the corrosion resistance.
- Typical steady-state corrosion trends for Al in distilled water under low flow conditions are indicated in Figure 3.1. Comparable data at higher temperatures is required for certainty about the break in the plotted data. However, loss of water by Al-alloy corrosion films does occur and can account for changes in mechanisms. These data are applicable to Al-clad fuels under relatively mild exposure, as experienced by the UW fuel.
- Minimal corrosion rates in the pH range of four to eight in O₂-saturated water at 50°C (122°F) (slightly broader range at lower temperatures).
- Aluminum and most common wrought alloys are essentially immune to stress corrosion cracking (SCC).
- The alloys addressed here are largely immune to intergranular attack (IGA) at low temperatures, but are susceptible in aqueous environments between 125°C to 250°C (257°F to 482°F), depending on alloy composition.
- Crevice corrosion is not generally a major phenomenon with Al alloys, but is accelerated in chloride solutions.
- Pitting corrosion is generally mild on Al alloys in pure water, even with Al-SS couples; pitting was mild in oxygenated distilled water at 70°C (158°F); traces of Cu⁺⁺ (e.g., from corrosion of heat exchangers) can initiate pitting; chlorides and MIC can also promote pitting of Al-alloys (Jones 1992).

Table 3.1 Aluminum-Alloy Compositions (wt. %)

Alloy	Al	Fe	Ni	Si	Mg	Other
1100	99 ^(a)	(b)	--	(b)	--	0.05 to 0.2 Cu
6061	97.9			0.6	1.0	0.25 Cr, 0.25 Cu
6063	98.9	--	--	0.4	0.7	0.1 max Cu
8001	(c)	0.6	1.1	(c)	--	

- (a) Minimum
 (b) Si + Fe = 1% max.
 (c) 1100 Al with Fe & Ni additions

Figure 3.1 Aqueous Corrosion of Aluminum in High-Purity Water Under Low-Flow Conditions (Draley and Ruther 1956)

Aluminum corrosion in relatively pure moist air environments is 0 to 0.1 $\mu\text{m}/\text{yr}$ (0.003 mpy) at ambient temperatures, with maximum pit depths of 10 to 55 μm (0.2 to 2 mil) in 20 yr (Houghton and Ashworth 1992). These conditions would likely be more aggressive than, for example, the helium cover gas used for Japanese fuel at 45°C (113°F), even if the helium had residual moisture (see Table 1.4). In summary, the data for corrosion of Al alloys indicates that Al experiences only mild attack from general, crevice, or pitting corrosion, and is relatively immune to SCC and IGA, unless chloride or MIC is involved.

3.1.2 Corrosion of Aluminum Alloys Under Reactor Conditions

High water flow and a radiation field can be important to corrosion rates, although there is evidence that radiation is not a significant factor for Al-alloy corrosion in some aqueous reactor environments (Dickenson and Lobsinger 1963). Data from in-reactor tests and examinations from reactors, such as the ORR, High Flux Isotope Reactor (HFIR), and ATR provide valuable information on hydrated oxide formations and expected oxide thicknesses of the SNF in the CPP-603 and CPP-666 pools.

Simulated corrosion of Al alloys in test reactors in flowing and static water at pH 5 and 100°C (212°F) indicated corrosion rates of 5 $\mu\text{m}/\text{yr}$ (0.2 mpy) (Griess 1994). Pitting in dissimilar metal crevices was not a major phenomenon. Oxides formed below 70°C (158°F) were indicated to be bayerite ($\beta\text{-Al}_2\text{O}_3\cdot 3\text{H}_2\text{O}$). Between 70°C to 80°C (158°F to 176°F) a duplex oxide of bayerite and boehmite ($\text{Al}_2\text{O}_3\cdot\text{H}_2\text{O}$) was observed. Only boehmite was detected above 80°C (176°F). Thus, reactor operating conditions affect the type of hydration, and the inventory of water in the oxide will vary depending on these conditions.

The Al alloy used for the cladding in the ORR reactor was subject to flow rates of 2.4 to 3.7 m/sec (8 to 12 ft/sec) and to minimal water movement in the storage pool [Griess 1994]. Water in both the reactor and pool storage systems was demineralized (0.75×10^{-6} to 1.25×10^{-6} ohm-cm). The water temperature range in the core was 51°C to 56°C (124°F to 133°F); in the pool it was 38°C (100°F) or less. Corrosion was less than 2.5 $\mu\text{m}/\text{yr}$ (0.1 mpy) in both environments. A few shallow pits (<25 μm , <1 mil) were attributed to surface inhomogeneities. This does not imply that Al-alloy corrosion is the same in flowing and static systems; in this case, both rates were below an apparent detection limit.

Aluminum-alloy plates from the HFIR core, which are not in the ICPP storage facilities, were examined in a hot cell (Griess 1994). Maximum cladding loss was 25 to 40 μm (1 to 1.6 mil) and occasional pits up to 75 μm (3 mil) were observed. The HFIR plates had two oxide layers of about equal thickness, differing from oxides from other test reactor fuels, which had only one layer. Fuels from high performance reactors (HFIR, ATR, HFBR) operate at temperatures where only boehmite would be expected. Based on existing information, it is not clear how fast the boehmite reverts to bayerite under pool storage conditions.

Oxide films up to 50 μm (2 mil) formed on ATR fuel elements that operate up to 227°C (441°F) (Lundberg 1994). Plate thinning averaged 6% but cladding plates decreased from 381 to 150 μm (15 to 6 mil)

in the worst case. Because thicker oxides tend to spall off, part of the material loss may be attributed to the high coolant flow rate in ATR. Based on these and preceding data, test reactors that operate at mild conditions, such as the UW and ORR, are estimated to produce oxide films of 1 to 2 μm (0.04 to 0.08 mil) at the end of fuel life, while high performance cores produce oxides of 25 to 50 μm (1 to 2 mil) on the Al-alloy cladding.

3.1.3 Corrosion of Aluminum-Clad Spent Fuel in Wet Storage

Wet storage prior to receipt at the ICPP is an important factor in the current and future condition of the fuel. Behavior of Al alloys in wet storage has been summarized in several publications (Kustas et al. 1981; IAEA 1982, 1988; Howell et al. 1993; Howell and Zapp 1993; Dirk 1994; Lundberg 1994). Information is provided below on the overall significance of pool chemistry on corrosion rates, examples of excursions in pool chemistry, and the effect of transferring degraded fuel from an aggressive to benign storage environment.

Importance of Pool Chemistry

A comparison of Al alloy behavior in wet storage conditions emphasizes the wide range of durabilities: e.g., pitting rates of 750 μm (30 mil) in 45 days in aggressive water compared to 25 μm (1 mil) in 15 yr in benign water.⁽¹⁾ Al-clad fuels have survived with minimal degradation for over two decades in storage pools where water purity is maintained, e.g., with low chlorides and low conductivities (IAEA 1993). Some Al-alloy fuel storage pool components have remained in service for over thirty years (e.g., at the pool of the National Research Reactor Universal in Canada and the RBOF pool at SRP). The RBOF pool is an example of a benign water environment for Al alloys with the following chemistry: conductivity 1 to 3 $\mu\text{S}/\text{cm}$; pH 6.3 to 7.8; and chloride <0.01 ppm. A corrosion rate of about 1 $\mu\text{m}/\text{yr}$ (0.07 mpy) was reported for an Al-alloy spacer exposed in a radiation field in the RBOF pool for 15 yr. The ORR pool liner (Al 6061) is another example of low corrosion rates in that there has been no evidence of corrosion or pitting over 25 years; results on test specimens confirmed these results (Griess 1994).

The water chemistry for the RBOF is roughly comparable to the CPP-666 chemistry (Table 2.4). However, experience in the CPP-666 pool, with excellent water chemistry control, indicates that Al alloys are not fully immune from pitting under these conditions (Hoskins et al. 1993). Presence of a slime in CPP-666 suggests that MIC may be the cause. The ultraviolet light system was not operational for about two years; however, it is not clear that the UV treatment would have prevented algae growth and bacterial growth may be more important than algae growth.

Aluminum alloys can degrade rapidly in pool waters of poor quality. Pitting completely penetrated coupons 750- μm (30-mil) thick in 45 to 100 days (time was alloy-dependent) in water with the following

⁽¹⁾ Johnson, Jr., A.B. 1994. Presented at the DOE SNF meeting in Salt Lake City, December 1994.

characteristics: conductivity $>130 \mu\text{S/cm}$; pH 6.3 to 7.1; and chloride up to 18 ppm (Howell et al. 1993). The most important factors in the rapid penetration were considered to be 1) high water conductivity, 2) elevated chlorides, 3) sludge (source of corrosive ions, e.g., Fe^{+++} , Cl^-), 4) galvanic couples (Al/SS and Al/Al involving alloys of differing compositions), 5) damage to protective oxides, and 6) relatively stagnant water; although fuels with even mild decay heats are likely to maintain thermal plumes with the attendant circulation, if water flow is not restricted.

Examples of Water Chemistry Excursions

Two cases of excursions have been noted for wet storage pools, one in the K-Reactor basins at Hanford and one in the CPP-603 pool. Corrosion monitoring in the K-Reactor pools illustrates a cycle of low uniform corrosion rates on Al alloys, followed by a sudden increase in rates caused by a biocide injection which increased the levels of chlorides and copper ions (from heat exchanger corrosion), resulting in Al-alloy corrosion increasing from $0.5 \mu\text{m/yr}$ to $5 \mu\text{m/yr}$ (0.02 mpy to 0.2 mpy).⁽¹⁾ More than a year of pool chemistry control was required to restore the low rates. Further water chemistry improvements have reduced Al-alloy corrosion rates to $2 \mu\text{m/yr}$ (0.1 mpy).

Chemistry conditions in the CPP-603 pool represent another case of a major extended water chemistry excursion (see Figure 2.3). The high chlorides resulted from the addition of biocides beginning in 1968 and caused corrosion of Al alloys for the fuel and storage component according to inspections (Hoskins et al. 1993; Lundberg 1994; Dirk 1994). However, the Al-alloy corrosion would almost certainly have been intolerable without addition of nitrate ions as an inhibitor (Dirk 1994). In fact, a case exists where uniform corrosion of Al alloys has proceeded at $38 \mu\text{m/yr}$ (1.5 mpy) in water with a conductivity of over $600 \mu\text{S/cm}$, which seems nominal for the aggressive conditions.⁽²⁾ Thus, pool conditions can greatly affect the performance of Al alloys.

Effect of Transfers from Aggressive to Benign Storage Environments

Because of high corrosion observed in some wet storage pools, it is relevant to consider whether transferring Al materials from aggressive water conditions, where pitting is active, to benign water conditions will reduce the pitting attack. One view maintains that corrosive conditions in a pit that is occluded by overlying

⁽¹⁾Personal communication with S.P. Burke, Westinghouse Hanford Company, July 1994.

⁽²⁾If the $38.1 \mu\text{m/yr}$ (1.5 mpy) rate applies to fuel cladding, a large fraction of the cladding would be consumed over a decade (~70% in the case of ATR fuel elements). However, the oxide films formed in reactor service are likely to provide protection that results in a much lower rate of cladding consumption by uniform corrosion. Pitting at sites where the oxide is damaged remains a prominent issue in CPP-603. While unirradiated Al materials are showing pitting tendencies in CPP-666, if MIC is the principal cause, then biological species that tolerate incident radiation would be required for pitting to proceed on fuel elements. In fact, species that tolerate radiation have been observed. Inspection of fuels in CPP-666 are planned to determine whether radiation suppresses the attack, if in fact it is biological (Dirk 1994).

oxide will not be mitigated by transfer to benign water conditions unless the oxide is removed and the pit is purged of the aggressive solution. While eventual mitigation of the aggressive solution seems likely by reaction, diffusion, and dilution of the corrosive species, a period of continuing pit propagation also seems plausible.

Impacts of fuel transfers from other facilities into CPP-666 must be considered and mitigated where significant. Movement of the spent fuel and containers may transfer or release:

- biological species
- contaminants from other pools (such as Cl⁻)
- corrosion and fission products carried into the pool or released during subsequent corrosion.

Biological species are a prominent concern, resulting in pitting of unirradiated Al alloys. Whether irradiated fuel cladding is susceptible is being investigated. Fuels with low radiation levels, such as the UW, should be included in the MIC surveillance. Transfer of contaminants with the fuel should be mitigated by dilution in the high purity CPP-666 waters; however, chlorides absorbed in the oxide films or occluded in pits are likely to continue to promote corrosion on the transferred fuel. Release of fission products from failed cladding may add detectable radioisotopes to the CPP-666 water, but concentrations will not be sufficient to accelerate chemical corrosion.

Corrosion products released from corroding fuel may affect adjacent fuels. For example, a Hungarian study by Schiller et al. (1987) suggests that aluminum ions can adversely affect Al-alloy corrosion. A significant finding of this study was that pool water conductivities and ionic concentrations doubled from the top to the bottom of the pool. This phenomena should be considered in water sampling and corrosion surveillance programs.

Based on Al-alloy corrosion behavior in other pools with well-conditioned waters (e.g., RBOF), Al-alloy fuels and components should be expected to have excellent durability in the CPP-666 pool. Kustas et al. (1981) provide quantitative data for RBOF. Caskey (1993) and Howell et al. (1993) cite favorable extended performance of Al alloys in RBOF. However, recent reports of pitting attack may require greater surveillance, understanding, and possibly mitigation before long-term satisfactory durability of Al-alloy fuels and components in CPP-666 is fully assured. Because of the recent experience with Al corrosion in CPP-666, transfer of Al-alloy fuels or components to this facility during stages of pitting may not receive the benefit of the more benign environment when the pits are occluded or if the MIC becomes active on cladding, particularly for spent fuel with low radiation levels. Despite the high circulation rate in CPP-666, the water at the fuel is considered stagnant. Transfer of metallic fuel with cladding breaches into CPP-666 would allow continuing corrosion of the metallic fuel, though possibly at lower rates than in CPP-603.

Transfer of mildly pitting fuel into the CPP-666 pool may not result in accelerated pitting rates if the pits are occluded. However, pit initiation is likely, particularly at sites where damage to the oxide film has exposed bare metal. The high pool water conductivity also will tend to promote galvanic and crevice attack. Control of chlorides to more recent values of ~ 50 ppm and continued use of nitrite inhibitor should lessen the impact of accelerated pitting.

3.1.4 Characteristics and History of Aluminum-Clad Fuel Stored in the CPP-603 and CPP-666 Basins

While corrosion data for other spent fuels and components fabricated from Al alloys provide a sound basis for predicting performance, actual evaluation of the characteristics and histories of the specific spent fuels provides even greater insight into the expected failure mechanisms for these materials. Specific inspections of Al exposed in the CPP-603 and CPP-666 also provide direct evidence of corrosion rates and failure mechanisms. This information has been determined for the ATR and UW fuels to provide an example of the extremes of expected fuel conditions. The information is summarized under three topics: analysis of characteristics and histories of specific fuel types, specific inspections of Aluminum-Clad fuel in CPP-603, and cladding failures from reactor service.

Analysis of Characteristics and Histories of Specific Fuel Types

Characteristics and histories of two fuel types, the ATR and UW Al-clad spent fuels, were investigated in detail as representing extremes in reactor service and storage conditions and are summarized in Tables 3.2 and 3.3, respectively. This information provided the basis for developing a detailed list of impacts of storage, shipping, and handling on the vulnerabilities of these two fuels as shown in Table 3.4. The vulnerabilities are summarized in Table 3.5 for all of the Al-clad fuels being evaluated. Details on UZrH_x fuel issues are discussed in Section 4. Only limited information was obtained on irradiation history and past storage experience for most of the Al-clad fuels. Thus, more detail might be added with further review of records and discussions with fuel shippers and vendors.

If the estimation of current conditions for the ATR and UW spent fuels are judged useful, then similar details should be traced for the remaining fuels. Otherwise, the vulnerability analysis for the other fuels addressed in Table 3.5 will be based largely on expected corrosion behavior in the CPP-603 or CPP-666 pools. Storage in these facilities is an important phase in the total fuel histories, but information on only the wet storage period at the ICPP provides incomplete perspectives regarding current fuel condition.

Specific Inspections of Aluminum-Clad Fuel in CPP-603

Inspection of Al-clad fuels and Al-alloy components stored in the CPP-603 basins has shown preferential attack in the form of pitting and crevice corrosion (Hoskins et al. 1993). The degree of attack

depended on the length of exposure. The corrosion appeared to be exacerbated by galvanic couples (Al/Zircaloy or Al/SS).

Cladding Failures From Reactor Service

Of special importance to this study is the nature and incidence of cladding breaches. Exposure of uranium-bearing metal fuels to aqueous reactant results in corrosion products that must be assumed to include uranium oxides and hydrides. If the cladding is failed, then the defective Al-clad UAl_x fuel element would have the characteristics shown in Figure 3.2. Some microstructures of UAl_x fuel have been published (Francis et al. 1962). In one set of UAl_x microstructures, the UAl_x particles were 1.7 to 40 μm (0.07 to 1.6 mil) across and the nominal distance between larger particles was 1.5 to 45 μm (0.06 to 1.8 mil). The dispersion of the UAl_x particles minimizes likelihood that sufficient occluded hydride would be available at a given location to support an ignition event, possibly differing from corroding uranium metal. The cladding and fuel compact are bonded, so corrosion is not likely to develop easy paths between the cladding and fuel where conditions could favor formation of occluded hydrides (local concentrations of hydrogen, limited access of moisture and air). Local pockets, perhaps partially occluded by corrosion products may preserve some hydrides (e.g., at C in Figure 3.2). Again, the offset between UAl_x particles does not seem to favor large local hydride inventories.

There is evidence of interaction layers between the UAl_x particles and the Al-alloy matrix for irradiated materials that may modify corrosion characteristics from simulations that may be conducted in laboratory tests (Richt et al. 1962). Lundberg (1994) describes in-reactor failures of ATR cladding that involved pitting attack. Pitting attack was also reported for Al-clad fuel failure at the SRP (Howell et al. 1993). Lundberg indicated that cladding penetration on the ATR fuel plates is expected to occur first at the hot stripe, where the axial temperature is highest. One ATR plate that operated 19 days after failure developed a pit 0.067 cm (26 mil) across. Another plate had a 0.085-cm (33-mil) pit after six days of operation. The pitted plate with longer operation lost 800 mg of fuel material; the other plate lost 200 mg. Considerable underwelling (loss of material from under the cladding) is indicated, but relative contributions to this loss from corrosion and hydraulic washout is not clear.

Corrosion rates for the UAl_x cermet were reported to be about twice the rates for 8001 Al at 290°C (554°F) in laboratory tests (Caskey 1993). However, the fuel loss rates in-reactor were considerably higher; the average linear fuel loss rate was 38 mg/day through the cladding breaches. Lundberg (1994) observed that conditions in the storage pools are likely to result in lower fuel loss rates. While, aqueous corrosion proceeding inside the pits is more likely to result in occluded hydrides, it does not seem likely that large local inventories of UH_3 would develop.

Table 3.2 Impact Assessment of ATR Fuel

Fuel Name	Reactor Impacts	Storage Impacts at Test Reactor Area (TRA) Pool	Fuel Shipping and Handling	Storage Impacts at CPP-603	Storage Impacts at CPP-666
(ICPP-603) ATR	<p>Conditions:</p> <ul style="list-style-type: none"> • 227°C (441°F) max • Coolant Rate, >12 m/sec (> 39 ft/sec) • pH- N/A • Time in reactor ~10 yr <p>Effects on Fuel:</p> <ul style="list-style-type: none"> • < 0.5% cladding breaches in reactor • Cladding wall thinning: 6% loss (25 μm, ~1 mil) • Pitting: 0.05 cm (20 mil) max • Oxide thickness: up to 50 μm (2 mil) 	<p>Conditions:</p> <ul style="list-style-type: none"> • 20°C (68°F) • pH - 5 to 7 • Cond. - < 5 μS/cm • Time in pool up to 10 yr <p>Effects on Fuel:</p> <ul style="list-style-type: none"> • Some pitting attack 	<p>Conditions/Events:</p> <ul style="list-style-type: none"> • Shipped wet TRA to CPP <p>Effects on Fuel:</p> <ul style="list-style-type: none"> • No known damage 	<p>Conditions:</p> <ul style="list-style-type: none"> • In Al baskets • Storage condition (see tables) • 20°C (68°F) • Cl⁻, 50 ppm • Cond. - >600 μ S/cm <p>Effects on Fuel:</p> <ul style="list-style-type: none"> • Mechanical damage when fuel handled • Pitting attack observed after 4 yr 	<p>Conditions:</p> <ul style="list-style-type: none"> • In Al baskets • 20°C (68°F) • Cl⁻, <0.1 ppm • Cond. - 0.5 μS/cm <p>Effects on Fuel:</p> <ul style="list-style-type: none"> • Mechanical damage when fuel handled • Some pitting on specimens

Fuel Characteristics

Cladding Alloy: 6061 Al/1100 Al (duplex)

Cladding thickness: 0.038 cm (15 mils)

Fuel: UAl_x particles in 8001 Al matrix

Table 3.3 Impact Assessment of UW Spent Fuel

Fuel Name	Reactor Impacts	Storage Impacts at Reactor	Shipping Impacts	Storage Impacts at CPP 666
UW	<p>Conditions:</p> <ul style="list-style-type: none"> • T max: 55°C (131°F) (water) 66°C (151°F) (fuel) • Coolant Rate: 75 L/min (19.8 gal/min) • Time in reactor^(a): 27 yr (1961 to 1988) • Time at temp: 3100 h • Water Conductivity: $\sim 1 \times 10^6 \mu\text{S/cm}$ • Reactor Power: 10 kw to 1967 100 kw to 1988 <p>Effects on Fuel:</p> <ul style="list-style-type: none"> • No evidence of fuel failures • Est. oxide thickness: $< 5 \mu\text{m}$ (0.2 mil) • Pitting: expected to be minimal (i.e., $\leq 25 \mu\text{m}$, $< 1 \text{ mil}$) 	<p>Conditions:</p> <ul style="list-style-type: none"> • Dry Storage^(a): in 9/88 to 2/90 • Storage temp: slightly above ambient • Cover Gas: air • Containment: steel pipes with plastic liners (Note: no specific drying) <p>Effects on Fuel:</p> <ul style="list-style-type: none"> • No evidence of fuel failure • TV inspections indicated no evidence of fuel degradation except few minor scratches; some tarnished areas, and small amounts of oxide • No known mechanical damage in fuel handling • No significant corrosion expected 	<p>Conditions:</p> <ul style="list-style-type: none"> • Shipped in cask with air cover gas <p>Effects on Fuel:</p> <ul style="list-style-type: none"> • No significant corrosion expected • No significant mechanical damage expected 	<p>Conditions:</p> <ul style="list-style-type: none"> • Earliest date: 9/88 • 20°C (68°F) • Cl⁻: 30 ppb • Conductivity: $\sim 1 \mu\text{S/cm}$ • Radiation: 10 R/hr (assumed) <p>Effects on Fuel:</p> <ul style="list-style-type: none"> • No uniform corrosion • Possible mild pitting at scratches

3.11

Fuel Characteristics

Cladding Alloy: 1100 or 1245
Cladding Thickness: 0.038 cm (15 mil)
Fuel: UAl_x particles in 8001 Al alloy matrix
Fuel Plate Thickness: Not Available
Fuel Particle Size: 1.7 to 40 μm (0.07 to 1.6 mil) (Francis et al. 1962)
Distance Between Particles: 1.5 to 45 μm (0.06 to 1.8 mil)
Anodized During Fabrication: Unknown, but would result in $\sim 25 \mu\text{m}$ (1 mil) of compact oxide
Dose Rate: $\sim 1 \text{ R/h}$ @ 1 m (3 ft) in air
Burnup: 14 MWd/MT

(a) The core consisted of 24 assemblies: some fabricated by Martin Marietta; some fabricated by Babcock & Wilcox. Two assemblies were replaced in 1970 and placed in dry storage (total time in dry storage: 20 yr). When the reactor was not operating, the water was removed and the fuel resided in moist air at est. 25°C to 40°C (77°F to 104°F). Portions of information provided by W.P. Miller of the University of Washington.

Table 3.4 Overview of Impacts for ATR and UW Fuels during Reactor Service, Storage, Shipping, and Handling^(a)

Fuel Source	ATR	UW
Reactor Impacts	<ul style="list-style-type: none"> • 0.5% of elements failed in reactor service • Cladding corrosion and wall thinning 	<ul style="list-style-type: none"> • No reactor failures occurred in 27 yr of reactor service
Storage Impacts at-Reactor	<ul style="list-style-type: none"> • Minimal cladding corrosion during ~10 yr at TRA • Corrosion of exposed UAl_x fuel at cladding defects 	<ul style="list-style-type: none"> • No indication of cladding failures during at-reactor dry storage • Minor indications of corrosion and tarnish in TV inspections
Shipping/Handling Impacts	<ul style="list-style-type: none"> • No indication of fuel damage in shipping • Corrosion of exposed UAl_x fuel at cladding defects 	<ul style="list-style-type: none"> • No indication of damage during shipping and handling
Storage Impacts at CPP-603	<ul style="list-style-type: none"> • Pitting attack on cladding observed • Corrosion of exposed UAl_x fuel at cladding defects 	N/A
Storage Impacts at CPP-666	<ul style="list-style-type: none"> • The UAl_x fuel is exposed at reactor-induced cladding defects (0.5% of fuel) • Further exposure of UAl_x fuel may have occurred due to pitting corrosion in FSF 	<ul style="list-style-type: none"> • Minimal corrosion of cladding • Possible pitting at oxide damage sites
Summary of Fuel Vulnerability	<ul style="list-style-type: none"> • High-performance fuel; some failed fuel; up to 50-μm (2 mil) oxide; cladding thinned in service and pitted in CPP-603 	<ul style="list-style-type: none"> • No known fuel failures or serious cladding damage; thin oxide evidence and systematic examination of fuel history indicates that cladding is probably not seriously degraded.

(a) See Tables 3.2 and 3.3 for details regarding reactor, storage, and handling conditions.

Table 3.5 Vulnerabilities of Al-Clad UAl_x Fuels in CPP-603 and CPP-666 Pools

Fuel Type	CPP Facility	Potential Vulnerabilities
ORR	603	<ul style="list-style-type: none"> • High Burnup in reactor service • Pitting corrosion in CPP-603 • No evidence of failed fuel in reactor service
UW	603	<ul style="list-style-type: none"> • No known failures or damage • Possible mild pitting at scratches (see Table 3.3 and 3.4)
ATR	603 and 666	<ul style="list-style-type: none"> • Known cladding failures and damage • Pitting attack in CPP-603 (see Tables 3.2 and 3.4)
ARMF	666	<ul style="list-style-type: none"> • Minimal corrosion, pitting corrosion possible at oxide defects^(a)
TRIGA AI	603	<ul style="list-style-type: none"> • Brittleness, possible UZrH_x solubility
MURR	666	<ul style="list-style-type: none"> • Minimal corrosion, pitting corrosion possible at oxide defects^(a)
MURR	603	<ul style="list-style-type: none"> • Pitting corrosion likely in CPP-603
HFBR	603	<ul style="list-style-type: none"> • Pitting corrosion likely in CPP-603
HFBR	666	<ul style="list-style-type: none"> • Minimal corrosion • Pitting corrosion possible at cladding defects

^(a) Pitting up to 0.3 cm (120 mil) in 4 yr observed on unirradiated Al alloys; MIC is suspected. Inspections of irradiated fuels in CPP-666 are planned to determine whether pitting is active on irradiated surfaces (Dirk 1994).

- A- Partially oxidized UAl_x particles
 B- Fully oxidized UAl_x particles
 C- Fully oxidized UAl_x particles and possible UH₃
 a) Aluminide Compositions:

UAl₂- 6%
 UAl₃- 63%
 UAl₄- 31%

Figure 3.2 Schematic of Corroding UAl_x Spent Fuel

3.2 FUEL FAILURE MECHANISMS AND THEIR CONSEQUENCES IN DRY STORAGE

This section focuses on the current spent fuel condition and its consequences on expected behavior in dry storage. A detailed historical investigation of all of the Al-clad spent fuels could not be accomplished in the allowed time because the available information was very limited and leads on this information could not be pursued. Therefore, the assessment of Al-clad spent fuels focused on two types that seemed to bracket the range of spent fuels included in this study. ATR fuel was selected as representative of the high in-reactor performance type and the UW fuel was selected as representative of the fuel types with low in-reactor performance.

- ATR fuel with higher decay heats, some failed fuel (~ 0.5%) from reactor service and visual evidence of pitting, continuing wet storage in both CPP pools, evidence of mechanical damage.

- UW fuel with low heat ratings, no evidence of breached cladding, periods of storage in moist air and ambient air. Because pitting is occurring during wet storage, a conservative position will be adopted that all ATR cladding has perforations.

The range of fuel element conditions is defined by two classes of spent fuel as shown in Table 3.6. The ATR and UW fuels are used to represent examples of the range of Al-clad fuel characteristics and storage histories. The Class A fuel has a low decay heat and is in good condition relative to the Class B fuel, which has higher decay heat and has fuel cladding breaches. This range of fuel classes will correspond to a range in failure mechanisms that must be analyzed for dry storage and are summarized in Table 3.7.

A comprehensive analysis of potential failure mechanisms was conducted for commercial LWR fuels in dry storage by Gilbert et al. (1990) (see Table 1.2). The dominant mechanism that emerged for commercial spent fuel was stress rupture, driven by internal gas pressures and the generally high storage temperatures resulting from substantial residual decay heats. Neither of these conditions exists in the Al-clad fuel addressed here.

If the cover gas is inert, the sources of reactants will be: impurities carried in the cover gas (<1 vppm H_2O , O_2 , N_2), which will be inconsequential for Al-alloys, and moisture, H_2 , and O_2 released by radiolysis (see Section 3.4.3), which are not aggressive to Al-alloys at moderate-to-low thermal conditions expected in storage for the fuels addressed here. Reactions of H_2O and H_2 with uranium are thermodynamically feasible under expected storage conditions (see Appendix B). However, aluminum alloys performed with no evidence of degradation in moist helium at temperatures up to $\sim 200^\circ C$ ($392^\circ F$) for nearly a decade (Richman and Pollock 1968), consistent with the use of helium for dry storage of Al-clad spent fuel in Japan (Adachi 1982). If air or nitrogen were selected for the cover gas, expected levels of nitric acid would need to be estimated, depending on predicted moisture levels and radiolysis rates.

If cladding breaches exposing uranium-bearing fuels are present in fuels that are candidates for dry storage, possible effects of moisture and hydrogen that build up during storage need to be considered. The impacts of the reactions on uranium metal fuels have been considered in detail elsewhere (ITA 1994). Uranium-aluminum alloy fuels are not likely to undergo substantial reaction.

Table 3.6 Dry Storage and Drying/Conditioning Categories for Al-Clad U Al_x Spent Fuels

Fuel Category	Example(s)	Characteristics	Estimated Storage Conditions	Potential Drying/Conditioning
A	UW	<ul style="list-style-type: none"> • Low decay heat • No cladding breaches • No plenum or significant internal pressure • No significant mechanical damage 	<ul style="list-style-type: none"> • Cladding temperature: <45°C (113°F)^(a) • Inert cover gas^(b) • Single canister 	<ul style="list-style-type: none"> • Drying cycle (conditions TBD)^(c); • Drying temperature to be higher than storage temperature
B	ATR	<ul style="list-style-type: none"> • Higher decay heat • Cladding breaches • No plenum or internal gas pressure • Known mechanical damage • Possible active pitting 	<ul style="list-style-type: none"> • Cladding temperature: TBD • Inert cover gas^(b) • Single canister 	<ul style="list-style-type: none"> • Wash off loose bayerite^(d) • Inert handling • Drying cycle (probably 100°C to 150°C (212°F to 302°F); possibly higher for thicker oxides)^(e)

(a) Estimated from conditions for Japanese Al-alloy fuel

(b) Analysis may justify air as a cover gas

(c) Conditions for Japanese Al-alloy fuel were 40°C (104°F) in air for 1 week, <50% RH

(d) When fuels have loose surface oxides, washing operations may effectively reduce the hydrated oxide inventory that will need to be dried. However, low-temperature oxides are frequently difficult to remove, requiring scraping and wire brushing.

(e) Conversion of residual bayerite probably above 150°C (302°F), close to 140°C (284°F), to remove physisorbed water.

Table 3.7 Potential Failure Mechanisms in Dry Storage for Al-Clad Fuels

Fuel Category	Environment Assumptions	Failure Mechanisms
A	<ul style="list-style-type: none"> • T <45°C (113°F) • Moisture in cover gas: <0.025% • Moisture in oxide TBD • Radiolysis not significant • Thin oxide films (<5μm (0.2 mil)) • Minimal pitting 	<ul style="list-style-type: none"> • Uniform corrosion: not significant • SCC/IGSCC: not significant • Pitting: not significant • Creep rupture: not significant
B	<ul style="list-style-type: none"> • T ~100°C (212°F) max (assumed, needs to be determined) • Moisture in cover gas: <0.025% • Moisture in exposed fuel: TBD • Radiolysis: depends on α / β fluxes • Thicker oxides (25 to 50 μm (1 to 2 mil)) • Active pitting 	<p><u>Cladding</u></p> <ul style="list-style-type: none"> • Uniform corrosion: not significant with effective drying • SCC/IGSCC • Pitting: not significant with effective drying • Creep rupture: not significant <p><u>Fuel</u></p> <ul style="list-style-type: none"> • Uniform corrosion: depends on α / β and moisture • Hydriding: depends on α / β and moisture

3.3 RECOMMENDED DRY STORAGE CONDITIONS

For most of the Al-clad fuels being addressed, low exposures and relatively long decay periods define relatively low storage temperatures. The Japanese experience (Adachi 1982; Adachi et al. 1984) involves a calculated storage temperature of 45°C (113°F) in inert gas. If the cover gas were air, the corrosion penetration in 50 yr would not be expected to exceed $\sim 25\mu\text{m}$ (1 mil) (ASM 1987). The preexisting oxides formed in service would probably limit the penetration in storage to much lower values.

Maximum storage conditions for Al-alloy fuels were addressed by Caskey (1993). The considerations based on 100-yr storage were:

- reduction in ductility between 150°C to 200°C (302°F to 392°F)
- degradation of hydrated oxides above 120°C (248°F); may require 225°C (437°F) for oxides with large H₂O inventories.
- cesium and uranium diffusion through cladding above 200°C (392°F) (currently considered to be a secondary concern)
- corrosion of uranium
- reaction of Al alloys with moist air above $\sim 250^\circ\text{C}$ (482°F)

Based on these considerations a maximum storage temperature of 150°C (302°F) was proposed for the SNF with Al-alloy fuels. Based on knowledge of Al-alloy behavior in the expected dry storage environments, this is regarded as a sufficiently conservative storage temperature for unfailed fuel and probably also for UAl_x fuels with breached cladding. For breached uranium metal fuels, the drying cycle, radiolysis levels, and a degree of acceptable uranium reaction in storage will need further consideration.

3.4 RECOMMENDED SPENT FUEL STORAGE PREPARATION OPERATIONS

The Al-clad fuels involve a range of considerations that must be addressed in decisions that will lead to safe and cost-effective transitions to dry storage. Different fuel categories are likely to require different levels of conditioning, as indicated in Table 3.5. However, these are preliminary perspectives to illustrate issues needing further definition. Further analysis and selected fuel characterizations should be scoped and focused to provide an informed basis for sound decisions regarding levels of conditioning that need to be applied to each fuel type to meet storage and, possibly, repository criteria.

From the fuel durability standpoint, issues that must be addressed include acceptable cladding temperature; required level of dryness, including impacts of corrosion and radiolysis; cover gas selection; and canister materials and design, including whether to seal or vent the canister. Aluminum-clad fuels have been in dry storage, some over extended periods (Table 1.4). Perspectives from that experience provide guidance

regarding the conditions that will provide required durability of the CPP fuels in dry storage. These perspectives are summarized below.

3.4.1 Acceptable Cladding Temperatures

It should be possible to control storage temperatures for the Al-clad fuel such that the mechanical integrity of the fuel will not be compromised (see Section 3.3). Therefore, failure considerations will center on prospects for cladding degradation (or degradation to fuel exposed at cladding defects) from reactions with cover gases, their impurities, or species released from the fuel element surfaces during dry storage, as mentioned in Section 3.2.

Low decay heats for several Al-clad fuels have resulted in low temperatures in dry storage. The storage temperature of Japanese test reactor fuel was calculated to be 40°C (104°F) in helium (Adachi 1982). Temperatures for Australian Al-clad fuels are not known, but the fuel cladding has not appeared to fail in ~35 yr of storage in nitrogen, based on gas monitoring.⁽¹⁾ The UW fuel was in dry storage for about 16 months at temperatures estimated to be only slightly above ambient.⁽²⁾

In summary, the Al-alloy fuels are expected to be storable at temperatures far below the regimes for commercial LWR fuels. The maximum allowable temperature of 150°C (302°F) (Section 3.3) is proposed to be sufficiently conservative.

3.4.2 Required Levels of Dryness

For a fuel type with intact cladding, conditioning may comprise only a simple drying cycle and sealing in a canister in an appropriate cover gas. Table 1.4 includes Al-clad test reactor fuel, dried in air at 45°C (113°F) and <50% RH, then welded into SS canisters with an inert cover gas.

For a fuel type with cladding breaches but a relatively non-reactive fuel (e.g., UAl_x , UZrH_x , UO_2), conditioning may also only require drying and sealing in canisters. However, choice of the cover gas may be more restrictive. For example, conditioning of commercial Zircaloy-clad LWR fuel only requires a drying cycle, but the environment must be inerted, due to the possibility of UO_2 exposure to oxygen at a cladding breach. Decay heats of LWR fuel often impose temperatures where oxygen reacting with UO_2 can result in lower density phases (e.g., U_3O_8); this may cause splitting of the cladding. On the other hand, some Zircaloy-clad CANDU fuel has been stored in air in tests because decay heats and corresponding temperatures are sufficiently low. For fuel types with cladding breaches and a reactive fuel type, particularly fuel with metallic uranium or uranium

⁽¹⁾Personal communication with M. Duignan of Westinghouse Savannah River Company, July 1994.

⁽²⁾Personal communication with W.P. Miller of the University of Washington, July 1994.

alloys, more complex conditioning may be required if the fuel is known or suspected to contain uranium hydrides (ITA 1994).

The potential sources of moisture in a canister containing a cover gas, Al-clad fuel elements with corrosion films, and, possibly, fuel with failed cladding are

- Waters of hydration:
 - $\text{Al}_2\text{O}_3 \cdot 3\text{H}_2\text{O}$
 - $\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$
 - $\text{UO}_3 \cdot x\text{H}_2\text{O}$ (for failed fuel with uranium corrosion products)
- Adsorbed waters:
 - oxide films can have large surface areas with several monolayers of adsorbed moisture
- Absorbed waters:
 - thicker oxides have cracks and pores that are penetrated by water during reactor service and wet storage
- Free water:
 - water films on fuel element and canister surfaces and residual water left in the cover gas after the drying cycle
- Water entrained in failed fuel:
 - large fraction of entrained water was removed from failed LWR fuel in vacuum drying at 325°C (617°F) (Kohli et al. 1985)
 - plate fuels with failed cladding will likely retain moisture more than the simpler rod design of commercial LWR fuels
- Water impurity in cover gases:
 - $<1 \mu\text{m}$ of Hg (H_2O water vapor) for commercial purity gases; generally a minor source; will react with U metal, but negligible product in limited gas volumes.

Boehmite decomposition at temperatures above 150°C (302°F) (see Appendix B, Figure B.6) is a potential source of moisture during dry storage of Al-clad SNF. Boehmite decomposition should be a negligible moisture source below 150°C (302°F).

Definition of expected pressure buildup during storage is important for container design. If drying has been conducted above expected storage temperatures, absorbed and adsorbed water and waters of hydration will

be relatively stable when temperature excursions occur. However, as indicated in the following section, radiolysis will generate an inventory of gaseous species, including H_2O .

3.4.3 Radiolysis

For unfailed fuels, the only contribution to radiolysis will come from gammas. For failed fuels, contributions from alphas and betas must be considered, depending on the area of fuel exposed. The alphas and betas have higher energy transfers than gammas per unit energy. The problem of applying radiolysis calculations to water on surfaces was addressed to estimate the importance of alpha, beta, and gamma radiation on release of moisture and hydrogen from the fuel and cladding.

G-values are numbers devised to denote the number of molecules produced per deposition of 100 eV of energy in a "local spur", whose dimensions should be loosely defined by the thermalization distance of secondary electrons. The concept of a "G-value" may not apply to "radiolysis" at interfaces. The thickness of an interface is typically on the order of the mean free path of the secondary electrons impinging or produced by particle impact. Thus, the real issue is a complicated secondary electron scattering problem. Despite this fact, useful approximations can be made concerning radiolytic production and desorption of thin films, such as multilayers of water which may exist at or within the interfaces of spent fuel rods.

One very important question concerns the actual amount of water in the uranium oxide corrosion film, since radiolytic production of H_2 and O_2 and stimulated desorption and diffusion of H_2O does occur. The first overlayer is thought to be double hydrogen bonded to a hydroxylated oxide and the subsequent overlayers are hydrogen-bonded. It will be difficult to completely remove all of this water, and the situation for uranium (tri)oxide is expected to be similar. Though the thickness of this multilayer is short compared to the half-thickness of extinction for water (11.4 cm), particles from decay events within the fuel core can terminate by energy deposition within the uranium (tri)oxide slurry. These medium energy particles, (i.e., Compton electrons at the tail of their energy distribution) can effectively couple to the electron density of the metal oxide and cause electron-hole pair production and secondary electron emission. The fate of these carriers and excess electrons could enhance G-values of the adsorbates. The enhancements can reach a factor of 20 and a few examples of these radiolytic enhancements have been published (Patil and Chiplunkar 1992; Garibov et al. 1982). Recent work at PNL (Kimmel et al. 1994) on the radiolysis of thin-films using low-energy (5 to 50 eV) electron-beam irradiation of amorphous and crystalline ice indicate that H_2 , OH, O, and H desorb from thin-films with reasonably high cross sections (approx. 10^{-18} cm^2). The relative branching ratios observed from 50 layer water films are $\text{H}_2\text{O}/\text{H}_2 = 3$, $\text{H}_2\text{O}/\text{H}_2 = 1.0$, and $\text{O}/\text{H}_2 = 0.9$. Only preliminary information on the production of H and O_2 has been obtained.

An interesting number to keep in mind is that at 10^{-6} Torr a surface will be dosed with background gas at a rate of 1 monolayer/sec. Therefore, at 1% relative humidity (approx. 0.2 Torr of H_2O), a surface will receive a water dose rate of 2×10^5 monolayers/sec.

In general, radiolytic degradation of thin films is facile and should not be overlooked. H_2 and O_2 production and water diffusion will occur; if sufficient quantities exist in the bulk or within the interfacial regions, it is believed pressure build-up and possibly corrosion will occur. It is clear that the situation in terms of pressure buildup and corrosion can be mitigated by removing as much water as possible via heat and/or vacuum treatment. It is also clear that all of the water will not be removed, so the problems will be mitigated but not removed. Unfortunately, sufficient information concerning interfacial effects does not exist to allow a detailed quantitative analysis of the mitigation extent. We assumed radiolysis mainly due to gamma rays only for the sake of simplicity.

Based on a rough estimate of α radiolysis of hydrated films on corrosion products, α radiolysis is not expected to play a role in release of gaseous species from hydrate films on Al. In the case of uranium, the film is expected to be a mixture of hydrated UO_2 and UO_3 , the latter being produced primarily by oxidation by the radiolysis products, OH, HO_2 , and H_2O_2 . The total G-value for these products for α radiation is about 2. The G-value for H_2 is about 1.7. Both values are energy dependent, the latter rising with linear energy transfer and the former decreasing (Draganic and Draganic 1971). The total G value for water destruction may be as high as 4.

An inspection of the composition of 6% ^{240}Pu Mark IV N-Reactor fuel cooled 5 years suggests a total of about 739 Ci/tU, made up of uranium, neptunium, plutonium, americium, and curium nuclides. This has been rounded off at 0.7 mCi per gram of UO_2 . The average energy is 5 MeV which gives 1.3×10^{14} ev/g-s. The range of 5 MeV alphas in air is about 4.25 mg cm^{-2} and about 14 mg cm^{-2} in uranium. This would give a range of about 7 to $8 \text{ } \mu\text{m}$ (0.28 to 0.30 mil) in the metal and probably 10 to $12 \text{ } \mu\text{m}$ (0.39 to 0.47 mil) in the hydrated oxide. Since the UO_x reaction product is reported to have a surface area of $30 \text{ m}^2/\text{g}$, the particles must have a nominal size of less than $5 \text{ } \mu\text{m}$ (0.2 mil). Thus, we can assume the full energy of the alphas will be expended in radiolysis in the thick hydrate film.

A worst-case estimate can be made by assuming all the radiation is absorbed by the water or OH equivalent, and using a G-value of 4,

$$1.3 \times 10^{14} * (3600/100) * 4 = 2 \times 10^{16} \text{ molecules}$$

of water ($6.6 \times 10^{-7} \text{ g}$) will be decomposed per hour per gram of UO_2 . If 20 mg $H_2O/\text{g } UO_2$ is assumed, it amounts to about 38% water decomposition in one year. The water content is based on data for PuO_2

(Stakebake and Steward 1973) which shows about 9 mg per gram for powder of $16.9 \text{ m}^2/\text{g}$ PuO_2 after heating to about 160°C (320°F).

For hydrogen production in the same calculation but using $G = 1.7$ would give $3.9 \times 10^{-4} \text{ mL H}_2/\text{h-g UO}_2$. If a canister contains 200 g of UO_2 , this becomes 0.04 mL/h ($0.0024 \text{ in}^3/\text{h}$) or 0.35 L/yr ($21 \text{ in}^3/\text{yr}$). The direct γ contribution is expected to be no more than 5 to 10% of that of the α and is ignored in this estimate. The pressure buildup will depend on canister volume, but appears to be minimal based on this analysis.

3.4.4 Cover Gas Selection

Aluminum alloys have been stored dry in helium, nitrogen, and air at low temperatures ($<50^\circ\text{C}$ ($<122^\circ\text{F}$)), as indicated in Table 1.4. Aluminum alloys have excellent corrosion resistance in air at 100°C to 200°C (100°F to 392°F). Reactions with impurities in commercial purity gases (H_2O , O_2 , H_2 , H_2) are not expected to have significant impacts on Al-clad fuel elements. However, in moist air or nitrogen, radiolytic formation of nitric acid is a consideration depending on moisture and radiation levels. Helium might be selected if needed for heat transfer.

3.4.5 Canister Selection, Design, and Closure

Selection of canister material must be based on both interior and exterior environments. There are several considerations for the exterior environment. Will the canisters be stored in a dry facility or submerged in a storage pool? What is the expected duration of interim storage? What are expected temperatures and environments?

Stainless steel was selected for storage of Japanese Al-clad fuel (Adachi 1982). Durability of SS canisters holding debris from the Three Mile Island (TMI-2) core was estimated for both wet and dry conditions (Johnson, Lund and Pednekar 1994). At 20°C (68°F) in moist air or in water (2 ppm Cl^-), durabilities will exceed any foreseeable interim storage period. Other canister materials, including CS, have been investigated for repository applications, so data on durabilities are available (e.g., ASM 1987).

Design considerations include the expected maximum pressure during storage. Wall thickness will be driven by projected maximum pressures from thermal and/or radiolytic release of gases (principally H_2O , H_2 , and O_2 if the canisters are sealed. If hydrogen/oxygen concentrations are projected to become excessive in storage, getters or catalysts could provide mitigation if long-term durability can be assured. Venting canisters to relieve pressure buildup has been applied to some canister types. However, when venting can be precluded by an effective drying cycle, the tradeoffs should be evaluated. An effective drying cycle removes sufficient moisture such that impacts during interim dry storage due to radiolysis and corrosion will not be significant. Design should also consider possibly moving the fuel to disposal without recanning.

4.0 URANIUM ZIRCONIUM HYDRIDE SPENT FUELS

The UZrH_x spent fuel in the CPP-603 basin consists of fuel from declad fuel elements, as well as fuel in complete fuel elements with Al and SS cladding that contain the UZrH_x . This chapter focuses on the issues relevant to the UZrH_x in all of the fuel forms stored in the CPP-603. However, issues related to the cladding are considered primarily in Section 3 for Al-clad fuel and in Section 5 for SS-clad fuel.

All UZrH_x fuels at the ICPP are from General Atomics' (GA) TRIGA type reactors. The TRIGA has been under development since 1957 (Simnad et al. 1976). In this time, the reactor design has seen a number of modifications for special applications, such as use in space reactors [Systems for Auxiliary Atomic Power or (SNAP)] and in neutron radiography. Common characteristics of these different types of reactors are their compactness, the use of light water at ambient pressure as the coolant by natural convection for power levels of up to 2 MW per fuel element, and power pulsing capability with peak power levels of up to 12,000 MW. In all types of TRIGA fuel, the fuel is in the form of a rod 25, 36, or 38.1 cm (10, 14, or 15 in.) in length, and 3.6 cm (1.4 in.) in diameter. The fuel is encased in Al or SS cladding.

The CPP-603 basin stores 459 declad fuel elements from SNAP reactors in 19 thin-wall (0.2 cm, 0.083 in.) Al cans (Lords 1994), 578 Al-clad elements in 125 Al buckets, 21 SS-clad elements in one canister, 7 SS-clad elements in 2 Al cans, and 263 SS-clad elements in 62 SS buckets^(a). There is no UZrH_x stored in the CPP-666. As will be shown in later sections, because of likely corrosion of Al cladding and containers, bare SNAP reactor elements and all of the Al-clad TRIGA elements may be assumed to be exposed to the basin water. Information is provided below on the fuel characteristics and failure mechanisms to the extent they are known (Section 4.1), the consequences of these failure mechanisms in dry storage (Section 4.2), recommended dry storage conditions (Section 4.3), and recommended preparations for dry storage (Section 4.4).

4.1 FUEL CHARACTERISTICS AND FAILURE MECHANISMS AFFECTING DRY STORAGE

The present condition of the UZrH_x fuel elements in the CPP-603 basin is dependent on the structure and properties of these elements when they were first put in a reactor and their entire life history. The structure of these elements, their mechanical and chemical properties during and after reactor operation, and the known or estimated fuel condition are described below.

4.1.1 Structure of UZrH_x Fuel Elements

This section provides additional details on the design and fabrication of UZrH_x fuels to assist in the understanding of their present condition. The general procedure for making UZrH_x fuel rods consists of casting a uranium-zirconium alloy into graphite molds, machining the rod to size, hydriding it in a hydrogen atmosphere at temperatures of 600°C to 800°C (1112°F to 1472°F), and grinding it to final size (Stahl 1982). The

^(a)Personal Communication with W.E. Windes, LITCO, July 1994.

resulting fuel rod is 3.8 cm (1.5 in.) in diameter and contains a drilled hole with a 0.64-cm (0.25-in.) diameter for faster and controlled hydriding. The central region was later filled with a zirconium rod, which is in the present CPP-603 SNF with cladding. All rods were slip-fit into a cladding with a thickness of 0.08 cm (0.03 in.) to give an annular gap of 0.05 cm (0.02 in.). Graphite cylinders 10-cm (4-in.) long were placed above and below the fuel rod to serve as reflectors. A disk of samarium or erbium poison was placed between the rod and the top graphite cylinder. The cladding was welded to end fittings of appropriate material (Al alloy for Al-clad and SS for SS-clad elements) to form a fuel rod about 71-cm (28-in.) long. In the case of SNAP reactor fuels, beryllium metal or beryllium oxide disks were used as neutron moderators/reflectors, and SS or Hastelloy cladding was used, and the overall length of an element did not exceed 38 cm (15 in.). Only Al and SS cladding are being addressed in this report because the CPP-603 inventory has only Al and SS cladding. The various types of TRIGA reactors included cladding of Al 1100 or SS Types 304, 304L, and 347.

The TRIGA fuels contained 8.5 or 12 wt.% uranium enriched to 20% ^{235}U . Some SNAP reactor fuels contained up to 20% uranium enriched to varying ^{235}U contents. The H/Zr atom ratio was close to 1.6 in all cases. On the hydrogen-zirconium phase diagram, this composition is in the single-phase, face-centered cubic δ (delta) region (Merten et al. 1958). As uranium is nearly insoluble in zirconium metal or the chemically very stable zirconium hydride (Hansen 1958), it precipitates out during hydriding as finely dispersed inclusions not more than 1 μm (4×10^{-5} in.) in size. Further, because of the much larger specific gravity of uranium compared with $\text{ZrH}_{1.6}$ (19.07 vs. 5.65), the uniformly distributed uranium inclusions in the hydrided fuel occupy from 2.7 vol% in the 8.5 wt.% U fuel to 3.9 vol% in the 12 wt.% U fuel (Simnad 1981).

4.1.2 Mechanical Properties of UZrH_x

The $\text{UZrH}_{1.6}$ alloy is extremely brittle at room temperature. A specimen pulled in tension breaks into multiple pieces by flat brittle fracture. It has a Young's modulus of 1×10^5 MPa (14×10^6 psi) and an ultimate tensile strength (UTS) of no more than 207 MPa (30,000 psi) (Merten et al. 1958). Compressive inelastic deformation of the alloy occurs by massive microcracking rather than by a flow process, with a compressive strength of from 276 to 690 MPa (40,000 to 100,000 psi) depending on the carbon content (Birney 1967). The percentage elongation, which is a measure of the plasticity of a material, is close to zero at temperatures below 600°C (1112°F) (Mueller et al. 1968). These data show that $\text{UZrH}_{1.6}$ is like glass in its properties. For example, the common soda-lime plate glass has a Young's modulus of 7.6×10^4 MPa (11×10^6 psi), a UTS of 240 MPa ($\sim 35,000$ psi), and % elongation of 0.0 (Mantell 1958) at room temperature. The $\text{UZrH}_{1.6}$ alloy therefore may be expected to behave like glass. One consequence of this brittle behavior is that the $\text{UZrH}_{1.6}$ fuel elements need to be handled as if one were handling glass or graphite rods of a similar form; unclad elements would break into pieces if dropped on a hard surface, or bent, or impacted with sufficient force.

4.1.3 Chemical Properties of UZrH_x

An important question in dry storage of UZrH_x fuel elements is the chemical stability and reactivity of this fuel in contact with water in wet storage and in contact with air in dry storage. Also important is the question of the stability of the hydride. These issues are discussed below.

Dissociation Pressure of Hydrogen

An important property in determining the stability of any hydride is the dissociation hydrogen pressure at a given temperature. The relationship between dissociation pressure and temperature for $\text{ZrH}_{1.6}$ is presented in Figure 4.1. Zirconium hydride will dissociate into zirconium metal and hydrogen gas by the following reaction:

Figure 4.1 Hydrogen Dissociation Pressure of $\text{ZrH}_{1.6}$ (based on Simnad 1981)

The higher the pressure of hydrogen gas in equilibrium with the hydride, the more unstable is the hydride. This results because hydrogen dissociates from the hydride and enters the space around the hydride until its pressure in the gas phase equals the equilibrium pressure. For $\text{ZrH}_{1.6}$, the partial pressure is 1 atmosphere at 760°C (1400°F). At this temperature, the hydride would spontaneously dissociate into metal and hydrogen gas at 1 atmosphere ambient pressure. At temperatures between 30°C and 100°C (86°F and 212°F), the partial pressure of hydrogen is very low, between 10^{-6} and 10^{-8} atmospheres. Thus, for storage temperatures below 100°C to 150°C (212°F to 302°F), loss of hydrogen should be negligible. This release will probably be even lower since these numbers do not account for any change in stoichiometry with release or any recombination with zirconium.

Corrosion of UZrH_x in Contact with Water

As noted previously, the structure of UZrH_x consists of small, μm -size inclusions of uranium in a matrix of zirconium hydride containing negligible concentrations of uranium in solid solution. Consequently, in the corrosion of UZrH_x in water, the corrosion performance of the two components of the structure, δ zirconium hydride and uranium inclusions, needs to be considered separately.

Zirconium hydride is a metallic hydride in that its chemical and most physical properties are like a metal. As far as corrosion is concerned, the hydride behaves like zirconium metal (Mueller et al. 1968). Zirconium is a very reactive metal when in contact with water; it dissociates water to form hydrogen and oxygen. However, because of the same high reactivity, it immediately combines with the released oxygen to form zirconium dioxide. The zirconium dioxide forms a tight, electrically insulating film on the zirconium surface, conferring immunity from corrosion to the surface at pHs of 4 to 12.5 (Pourbaix 1974). Zirconium hydride exposed to air forms a protective film of the same oxide and has negligibly small corrosion rates when immersed in water. For this reason, the zirconium hydride matrix in bare UZrH_x fuel will be immune to corrosion by water, whether it has an air-formed film or a reactive, film-free surface when first immersed in water.

The finely dispersed uranium metal inclusions on the fuel surface are another matter. In water with a pH of 8 or lower, uranium will dissolve as trivalent uranium ion, U^{3+} . At pHs >8 , uranium will form hydrated, green uranous hydroxide, $\text{U}(\text{OH})_4$. In either case, the reaction should continue until all metallic uranium has been removed (Pourbaix 1974). However, this will apply only to the surface inclusions of uranium in the UZrH_x fuel. The subsurface inclusions will be protected from coming into contact with water by the overlying zirconium hydride matrix. In addition, it should be noted that the uranium inclusions in the UZrH_x fuel are randomly and finely distributed and that they do not form an interconnected network.

4.1.4. Known Spent Fuel Condition

The UZrH_x fuel in the CPP-603 pools are in Al-or SS-clad rods, as well as declad SNAP fuel that is stored in storage buckets exposed to the pool water. Declad SNAP reactor fuels were canned into thin-walled Al

cans at the ICPP, as noted previously^(a). Considering the chloride excursions experienced in the basin water (see Figure 2.3) and the about 20 years of storage, it is not surprising that the cans have corroded through in places and the pool water has intruded into some of the cans, as observed by LITCO (Lords 1994). However, based on the chemical properties of UZrH_x fuel, the fuel rods ought to be in the same condition as when they were originally placed in the cans, except that the surface may be denuded of exposed uranium inclusions and the surface hydride may have lost some hydrogen.

All the TRIGA elements at ICPP are stored directly in the pool water, not in cans. Many of these elements had been out of the reactor for 8 to 20 years before being shipped to the ICPP^(b). All of these elements have had very low fuel burnups on the order of 0.01%. Because of the low burnups and the long out-of-reactor storage before shipment, decay heat was negligible and the radiation levels were below 15 R/h at contact prior to shipment to the ICPP according to shipment records. These elements apparently had developed punctures in the thin Al cladding due to corrosion, as positive helium leak detection results were reported in the shipment protocols.

An important consideration in evaluating the UZrH_x and cladding condition is the temperature of these fuel elements at shipment and presently at the ICPP. Plots of temperature of individual fuel elements versus operating power density in kW/element for various cooling times in air, supplied by GA, show that the element temperatures were below 100°C (212°F) at the time of shipment to the ICPP for both Al-clad and SS-clad elements. Because the temperature in the fuel during wet storage has probably been lower than at shipment, there are probably no concerns over excessive hydrogen release from the fuel.

If the cladding of the fuel elements containing UZrH_x fuel fail, then water can access the fuel. As explained in Section 3 for Al cladding and later in Section 5 for SS cladding, there are mechanisms that could cause the cladding to fail. Failure of the SS cladding has not been noted, but Al-cladding corrosion has been observed. Aluminum alloy 1100, the cladding for the TRIGA fuel elements, is resistant to corrosion in pure water at temperatures below 100°C (212°F) with general corrosion rates of below 2.5 $\mu\text{m/yr}$ (0.1 mpy). However, it will pit in waters containing chlorides and under deposits (Szklańska-Smiałowska 1986). Considering that the clad UZrH_x fuels have been exposed to 300 to 800 ppm chloride, and chlorides tend to segregate under deposits and in pits, it is conservative to assume that chloride concentrations reached much higher levels locally on the surface of these elements. Recent sludge samples from the CPP-603 pools indicate chloride concentrations are 500 to 800 mg/g of sludge^(c). Maximum pit depths of up to 0.1 cm (40 mil) have

^(a) Since time of writing the SANP fuels have been recanned into stainless steel canisters.

^(b) Personal communication with W.S. Whittemore of General Atomics, June 2, 1994.

^(c) Personal communication with W.E. Windes (LITCO), July 1994. "Final Report for 603 Sludge," by V. Barnes.

been reported for Al 1100 for exposure of five years in very high chloride waters, such as sea water (ASM 1987). As the aluminum cladding on the TRIGA elements is only 0.05-cm to 0.08-cm (20-mil to 30-mil) thick, the probability is very high that most of the elements stored at the ICPP have some through-wall penetrations.

4.2 FUEL FAILURE MECHANISMS AND THEIR CONSEQUENCES ON DRY STORAGE

Given the available information in the literature, it may be concluded that the UZrH_x in the TRIGA and SNAP reactor fuels in storage in the CPP-603 basin only have surface oxidation of the zirconium hydride matrix and complete removal of surface uranium inclusions exposed to the pool water, which probably entered any cladding penetrations long ago. For unclad UZrH_x or UZrH_x in fuel elements with defected cladding, only four issues appear to exist: brittleness, hydrogen diffusion, fuel stability and ignitability.

The only property of the zirconium hydride matrix that is considered detrimental is extreme brittleness, not just for dry storage, but for any handling of the fuel in preparation of dry storage. All TRIGA and SNAP reactor fuels will need to be handled as if they were made of a brittle material such as graphite or glass. If dropped or bent, they are expected to break into pieces, but will not shatter into shard-like fragments unless the forces are highly compressive. Such an event could become a cleanup issue or a criticality concern if extensive enough.

The dissociation hydrogen pressure for $\text{ZrH}_{1.6}$ matrix at the maximum fuel surface temperature of about 100°C (212°F) is 6×10^{-9} atm. Hydrogen from the hydride matrix will diffuse to the outside until there is insufficient driving force. As the released hydrogen will escape to the outside if the cladding has been ruptured by pits, there should be a very small but steady loss of hydrogen to the ambient atmosphere. If the cladding is intact, loss will occur from the fuel until the above pressure has been reached. After that, hydrogen will diffuse through the cladding at a steady but much smaller rate than if the cladding had been compromised. General Atomic's publications indicate that the loss is below 1% for intact SS-clad fuel under full power reactor operation. Even a small loss of hydrogen, will cause a dramatic decrease in the dissociation pressure. At the low temperatures of wet or dry storage, the loss should be exceedingly small and kept even lower by the gettering capacity of the zirconium insert in the hydride fuel and their resistance to hydrogen diffusion presented by zirconium oxide films on the fuel. Neither the escaped hydrogen nor the hydrogen depleted matrix appear to pose any dry storage hazard, although data does not exist to clearly support this conclusion.

The available literature indicates that the UZrH_x fuel is very stable in water and moist environments. Therefore, continued wet or dry storage should not be an issue as far as loss of radionuclides to the environment. However, the conclusions on fuel stability are not based on actual leach testing for the extended times or actual environments experienced in the CCP-603.

It does not appear that the hydride will ignite in air on exposure of bare element. As the solubility of hydrogen in zirconium decreases with temperature, this will only happen at temperatures above which the

hydrogen partial pressure is greater than the spontaneous ignition composition of $\sim 0.04\%$. This temperature for $\text{ZrH}_{1.6}$ is about 660°C (1220°F)^(a). For lower hydrogen contents, as in a hydride that has been losing hydrogen by diffusion over a number of years in storage, the temperature is higher. For example, the ignition temperature will be 700°C (1292°F) for $\text{ZrH}_{1.5}$. Except for an off normal event such as a cask fire, such temperatures are extremely unlikely.

4.3 RECOMMENDED DRY STORAGE CONDITIONS

Based on the previous discussion, it appears that the UZrH_x fuel elements can be safely stored in air without any hazards of radionuclide escape to the atmosphere if their cladding is intact. The acceptability of dry storage in air is supported by the experience for discharged TRIGA elements stored dry at GA and at laboratories such as Cornell University. TRIGA fuels were stored dry out of the reactor by GA for periods ranging from 8 to 20 years before shipment to ICPP^(b). According to GA's calculations, after 8 years of decay the amount of ^{129}I and ^{85}Kr (the two principal gaseous isotopes of concern) are negligibly small ($<1\mu\text{Ci/element}$ for ^{129}I and $0.1\mu\text{Ci/element}$ for ^{85}Kr) for the worst case of 100% burnup. The documentation submitted by shippers of TRIGA fuel indicate that the burnups were much smaller, less than 1% in most cases, and 12% in the worst case. Considering this information, a storage temperature of 100°C to 150°C (212°F to 302°F) with no stringent moisture limitations appears reasonable based on the decay heat and hydrogen release rates and apparent insolubility in water. The performance of the fuel requires data to confirm these expectations.

4.4 RECOMMENDED SPENT FUEL PREPARATION OPERATIONS

The UZrH_x de-clad fuel elements are presently stored bare in thin Al cans which probably are penetrated by pits and contain basin water^(c). The elements also may be present as broken pieces of UZrH_x . The last batch of these elements was put into wet storage at ICPP in September 1974. Because the release of gaseous fission decay products after such a long time is expected to be negligible, it should be possible to remove the cans from water to air without special precautions. As the cans are likely to contain water from the basin, and further aqueous corrosion of the elements cannot occur in the absence of water, the water in the cans should be removed. Instead of recanning the cans, it is suggested that each fuel piece be removed, drained, dried by some convenient means to minimize the water on the outer surface, and placed in dry SS containers. It is not necessary to remove all traces of water from the elements. The limited quantity of water that might be introduced in the final dry-storage containers in this manner does not appear to present a source of significant degradation. The

^(a) Personal communication with C. Wisham, General Atomics, May 1994.

^(b) Letter from L.E. Boysen, Manager, Purchasing, General Atomic Company, to Idaho Chemical Processing Plant, dated December 23, 1978.

^(c) Since time of writing the SNAP fuels have been recanned into stainless steel canisters

containers should be sealed by welding, but could be mechanically sealed to limit the rate of hydrogen loss to the atmosphere. After this operation, the containers may be stored dry with moisture limits set to prevent degradation of the containers rather than the fuel.

5.0 STAINLESS STEEL CLAD SPENT FUELS

Numerous research reactors used SS-clad fuel from the 1950s to the 1980s. The three categories of fuel used with SS cladding in the SNF being studied in this report are UO_2 , uranium metal, and UZrH . The fuel was clad with one of the four austenitic stainless steels: Types 304 (APPR, OMRE, Fermi Blanket, EBR-II, TRIGA FLIP, EBR-II (ANL-6), and TRIGA SS), 304L (SPERT, VBWR, BORAX V, EBR-II, TRIGA FLIP, and TRIGA SS), 316L (Pathfinder), and 347 (SM-1A and TRIGA BER II). The nominal compositions for these alloys are shown in Table 5.1.

Type 304 is the most commonly used general purpose SS alloy in the nuclear industry, as well as for the SNF in the ICPP, due to its low cost and ease of formability. Type 316L is alloyed with less carbon to increase the corrosion resistance in welded structures (less carbon to form chromium carbides). Type 304L also is alloyed with less carbon for better welding characteristics, but along with Type 316, also has a small amount of molybdenum for pitting resistance (ASM 1990a).

5.1 FUEL CHARACTERISTICS AND FAILURE MECHANISMS AFFECTING DRY STORAGE

As explained in Sections 1 and 3, failure mechanisms affecting dry storage will be dominated by the condition of the fuel prior to its transfer to dry storage. Due to the nature of the testing in the research reactors (fuel cladding pressure and temperature threshold testing), the SS-clad fuels

Table 5.1 Nominal Composition of Stainless Steel Used for Cladding								
Alloy	% C	% Mn	% Si	% Cr	% Ni	% P	% S	% Other **
304	0.08	2.00	1.00	18-20	8-10.5	0.045	0.03	...
304L	0.03	2.00	1.00	18-20	8-12	0.045	0.03	...
316	0.08	20..	10..	16-18	10-14	0.045	0.03	...
316L	0.03	2.00	1.00	16-18	10-14	0.045	0.03	2-3 Mo
347	0.08	2.00	1.00	17-19	9-13	0.045	0.03	10 X %C min Nb + Ta

** Balance is Fe

have often been subject to aggressive environments leading to documented accounts of degraded cladding in many fuel types listed in this study (Goslee 1963; Gallagher 1964).^(a) All of the SS-clad fuels, with the exception of the Borax V, Pathfinder, TRIGA BER-II, and TRIGA SST fuels, have been canned with available documentation suggesting that many of the fuel elements were failed prior to storage. The cans used for fuel storage are fabricated from one of three different materials: SS, CS, and Al. There are few data on the integrity of the individual cans, except for some underwater visual inspections and Fermi mock-up can testing. Often, the failure analysis or metallurgical examination, if any, of the failed fuel or storage cans produced inconclusive results as to the definitive mechanisms that caused the failure. Accordingly, it is necessary to examine the potential degradation mechanisms experienced during the life of the fuels to gain insight into the storage behavior of these fuels stored at INEL.

The following sections will address the degradation mechanisms of SS clad fuel in greater detail. These mechanisms were determined to have enough probability of occurring under certain circumstances to warrant discussion. The mechanisms discussed include general corrosion (Section 5.1.1); stress corrosion cracking, intergranular attack, and irradiation-assisted stress corrosion cracking (Section 5.1.2); localized corrosion (Section 5.1.3); rapid stress rupture (Section 5.1.4); strain rate embrittlement (Section 5.1.5); creep failure (Section 5.1.6); hydrogen-induced degradation (Section 5.1.7); helium embrittlement (Section 5.1.8); fission product-cladding interaction (Section 5.1.9); and chemical stability of fuel material constituents (Section 5.1.10).

5.1.1 General Corrosion

Stainless steels are chosen for applications such as fuel cladding because of excellent corrosion resistance the alloy exhibits in many environments, but the SS fuel cladding was found to be vulnerable to many of the aggressive environments produced by reactor service. General corrosion was not found to shorten the cladding life appreciably in-reactor or in storage, but other specialized forms of corrosion, such as SCC, caustic cracking, galvanic corrosion, irradiation assisted stress corrosion cracking (IASCC), pitting, and IGA were implicated in many of the cladding failures. The different mechanisms will be discussed in this section.

The SS-clad fuels considered for this analysis experienced one of three in-reactor coolant environments: 1) light water, 2) liquid metal (sodium), or 3) organic coolant. The degree of degradation experienced by an alloy is often dependent on the time the alloy has been subject to that environment. It was not possible to precisely state from available documentation the time any one fuel element spent in any of the environments of concern, in-reactor or in storage. Consequently, conservative estimates must be used to predict the time any of the fuel resided in the environments considered.

Additionally, little information was given on the fuel condition prior to its arrival at INEL. One

^(a)Letter from E.P. Mondok (Exxon Nuclear Idaho) to M.J. Painter, titled "Results of SM-1A and BORAX-V Fuels Leak Tests," dated July 23, 1982.

exception is the TRIGA SS and TRIGA FLIP fuel shipped from GA. This fuel was stored dry at GA's facility in 55-gallon drums for periods ranging from 8 to 20 years before shipment to INEL. Qualitative and semi-quantitative measurements (visual examination with a surveyor's telescope and internal swipes of the empty storage drums) performed by GA on the fuel cladding prior to shipping to INEL suggest that the fuel tolerated the dry storage with little to no degradation evident.^(a)

For the other fuels considered, published accounts of fuel environments or conditions are used to conservatively evaluate the status of the fuels prior to placement in dry storage. Because no measured data exist on the corrosion of the specific SS-clad fuels in water storage at INEL, a survey of corrosion of SS in water, sodium, and organic environments is provided below; where possible, direct experience with the cladding in reactor and storage environments are given.

General Corrosion in Water Environments

Stainless steels are highly resistant to general corrosion with the formation of a protective oxide and can exhibit excellent integrity in environments that do not promote localized corrosion. Stainless steel (tie-plate) coupon data from the CPP-603 indicates a negligible loss of material over 15 years.^(b) The corrosion rate for the coupons was less than $3.3 \mu\text{m}$ (0.129 mil/yr) with a confidence level about 95% (Thomas 1993). The low general corrosion rates cited in the INEL coupon studies are supported by several investigations discussed below that evaluate corrosion of SS and SS SNF and indicate that general corrosion of the cladding should not be of concern.

Corrosion rate studies using specimens of Types 304 and 348 SS indicate that irradiation during reactor operation acts to slightly accelerate the uniform corrosion rate, but it seems unlikely that the radiation fields ($\leq 200 \text{ R/hr}$) in the CPP-603 and CPP-666 pools would influence the corrosion rate. Corrosion rates of Types 304 and 348 specimens exposed in the Engineering Test Reactor (ETR) were 4 and $2.8 \mu\text{m/yr}$ (0.16 mpy and 0.11 mpy), respectively (Johnson 1975). The reactor operating conditions were 270°C to 280°C (518°F to 536°F), pH 10 NH_4OH , $<0.05 \text{ ppm O}_2$, and a neutron flux range from 9×10^{19} to $1.5 \times 10^{21} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$). The corrosion rate for a nonirradiated control Type 348 specimen was $1.3 \mu\text{m/yr}$ (0.05 mpy). Additionally, the oxide layer correlated to 60% of the total corrosion penetration. If the process is not taken to be diffusion-limited, the oxide layer penetration corresponds to a corrosion penetration of 1.6 to $2.4 \mu\text{m}$ while in reactor. The Type 304 SS oxide layer measurements correlated well with oxide layer thickness measurements of $6 \mu\text{m}$ (0.23 mil) from a Type 304L LWR assembly that had been irradiated for three cycles in the Haddam Neck reactor (Langstaff et al. 1982). This oxide layer appeared tenacious and stable after a five-year period of storage in a

^(a)Personal communication with C. Wisham and W. Stout of General Atomics, May 1994.

^(b)Memo from W.J. Dirk (Westinghouse Idaho Nuclear Company) to G.E. McDannel, Dirk-19-93, titled "Semi-annual Corrosion Inspection of the CPP-603 Stainless Steel 'Tie-Plate' Specimen," dated April 29, 1993.

PWR spent fuel pool. The SS-clad fuels from the water-cooled reactors considered for this study are not expected to fail in water storage or dry storage by general corrosion due to the protective nature of the oxide film.

General Corrosion in Organic Environments

The only SNF at the CPP-603 that was irradiated in an organic moderated environment was the OMRE fuel. Stainless steel is largely resistant to organic coolant environments in test reactors, but reactions with oxygen and water contaminants in the coolant result in magnetite formation. Chloride in the organic coolants can have a particularly deleterious effect on corrosion rates. Schleicher (1963) found that coupon weight changes were 30 times higher for an increase in the chloride level from 3 ppm to 100 to 200 ppm at 450°C (842°F) (Schleicher 1963). Corrosion rates are similar for tests with both irradiated and unirradiated coupons. The Cl⁻ levels that the OMRE fuel was exposed to are not known.

Corrosion particles consisting primarily of Fe₂C₉, Fe₂C, and Fe₃O₄ were found in the OMRE coolant from corrosion of the steel reactor vessel. Due to inadequate coolant purification, these corrosion particles plated out on the fuel element surfaces, causing fuel element plugging and gross fouling of the fuel elements. This also led to high temperature gradients in the fuel and to hot spots and warpage in the fuel elements, which was especially evident in stagnant regions such as slots in the fuel plates. The heat and the radiation induced chemical changes produced tenacious surface films on the fuel cladding that interfered with the heat distribution along the cladding, which could be controlled but not eliminated (Simnad 1971). Hot spots, warpage, and other temperature-induced defects results in compromised cladding, which offers less resistance to other degradation mechanisms while in storage. It is not known if the OMRE fuel element stored in CPP-603 has cladding defects due to nonhomogeneous heat distribution while in-reactor caused by corrosion particles in the coolant.

General Corrosion in Liquid Sodium Environments

The general corrosion rate of SS in liquid sodium test reactor environments is dependent on the purity of the liquid sodium. In a low oxygen environment (≤ 10 ppm), the corrosion rate of austenitic SS ranges from 7.6 to 38 $\mu\text{m}/\text{yr}$ (0.3 to 1.5 mpy) at temperatures to 704°C (1299°F) (Berry 1971). Irradiation appeared to have little effect on the corrosion rate. A 14-month study of Type 304 SS coupons in the EBR-II core showed essentially no weight change at temperatures to 530°C (986°F) (ANL 1967). As with the fuels in the water-cooled environments, general corrosion was not the predominant failure mode. The SS cladding was found to be very susceptible to caustic cracking, IGA, SSC, and pitting, which will be discussed in the following sections.

5.1.2 Stress Corrosion Cracking, Intergranular Attack, and Irradiation Assisted Stress Corrosion Cracking

The cladding alloys for this study can be categorized as austenitic SS that are susceptible to sensitization, characterized by the formation of chromium carbides at the grain boundaries that deplete the

chromium in the areas directly adjacent to the grain boundaries and render the material susceptible to environmentally induced corrosion. In order for sensitization to be an issue, the material must be held for a minimum amount of time at a temperature above the threshold temperature for diffusion to occur to deplete the chromium levels adjacent to the grain boundaries. These zones of chromium depletion can become prone to corrosion when exposed to a sufficiently aggressive environment that results in IGA, or if stresses are present in this aggressive environment above some threshold value, IGSCC. Chromium depletion can also occur without carbides being formed as the result of radiation induced segregation, which leads to IASCC (Andresen et al. 1989). Stress corrosion cracking requires the confluence of a susceptible microstructure (i.e., sensitized), a tensile stress (even residual stresses from welding operations), and an aggressive environment (although SCC is exhibited in high-temperature, high-purity reactor water conditions). Bruemmer et al. (1984) identified conditions necessary for IGSCC in sensitized Type 304 SS in water at 32°C (90°F). The conditions were a combination of 1) chlorides ≥ 0.5 ppm; 2) bulk carbon content ≥ 0.06 wt%; and 3) applied stress ≥ 1.4 times the yield strength. Higher temperatures, such as fuel storage temperatures, increase the aggressiveness of the environment and promote the initiation of IGSCC. Sensitization can occur during irradiation at temperatures as low as 427°C (800°F) (Long and Michelson 1964) and at temperatures as low as 482°C (900°F) in the absence of neutron radiation (Brick et al. 1965).

Low-temperature sensitization (LTS) is also a possible degradation mechanism for austenitic SS in wet or dry storage that had been exposed to short periods of high-temperature exposure in the reactor. Small carbides are precipitated, but the chromium level in the adjacent area is not markedly reduced due to the small size of the precipitates. Sensitization can subsequently occur during long periods at temperatures below that required to sensitize in a single brief isothermal exposure. Brief high-temperature fuel cladding experiments have been performed for many of the fuel types considered in this analysis, and would be the normal mode of operation for the "pulse" reactors (TRIGA). The temperature limit to prevent LTS of the cladding during dry storage can be extrapolated from data presented by Andresen et al. (1981) that indicates an upper limit of 255°C (490°F) for up to 50 years of storage. This temperature only holds true if there is no prior degradation from reactor service and water storage. There is insufficient information about the cladding temperatures during water storage to evaluate the potential for LTS during the water storage period for the fuels. Due to the age and low burnup experienced by much of the fuel, low fuel temperatures (upper bound $<150^\circ\text{C}$) are expected in dry storage^(a), so LTS in dry storage does not appear to be a credible degradation mechanism for temperatures at that level.

It is apparent from the list of cladding temperatures at normal operating conditions shown in Table 5.2 that most of the SS-clad fuel considered for this study was operated at temperatures where sensitization was

^(a)Personal communication with W.E. Windes of LITCO, July 1994.

possible. A sensitized microstructure can be detrimental to the integrity of the fuel cladding in the presence of elevated (>0.5 ppm) levels of chlorides. The maximum levels of chlorides experienced by the individual fuels based on the date of receipt are given in Figures 2.3 and 2.7, and are high enough to cause concern as to the status of the fuel cladding. Water environments and moist air environments have produced rapid corrosion and cracking in sensitized SS even at spent fuel pool temperatures (Long and Michelson 1964). Specific to the fuels in CPP-603, intergranular cracking was identified as a failure mechanism for APPR (Age 2), EBR-II, SM-1A, and later cores of the VBWR (later than the receipt date given for the fuels in the CPP-603). Since all of the SNF in the CPP-603 has been exposed to >0.5 ppm Cl^- , if they have been exposed directly to the basin water or if they have water from leaking cans, the potential for IGSCC is high for the fuels identified as potentially sensitized.

This susceptibility to IGSCC is related in an example of fuel cladding degradation experienced with gas-cooled reactor fuel. Extensive intergranular corrosion resulted in SS Type 304 cladding on prototype instrumented fuel elements for the Experimental Gas-Cooled Reactor during storage in a high-humidity environment or underwater following irradiation in the helium-cooled ORR Loop 1 at cladding temperatures up to 816°C (1500°F) (Long and Michelson 1964). Extensive IGA of SS Type 304 was also detected in components operating adjacent to the test elements, but at lower temperatures of 427°C to 650°C (801°F to 1202°F). Each element was irradiated for reactor cycles of 50 or 100 days in helium at 2.1 MPa (300 psia). This range of temperatures is consistent with the range of 427°C to 816°C (800°F to 1500°F) indicated in ASTM (1993) for sensitization of nonstabilized austenitic SS. Elements 1) loaded into a storage tube containing moist air and pressurized with 0.2 MPa (20 psig) nitrogen and placed in the reactor pool for 15 to 44 days or 2) stored in pool water prior to examination were found to have extensive IGSCC. Special efforts to store subsequent elements in a dry helium-nitrogen-oxygen environment for only three to ten days resulted in less SCC. These results led to the conclusion that storage of sensitized SS Type 304 in a high humidity environment or in water may lead to extensive intergranular corrosion.

In addition to the potential for sensitization of the cladding, the use of liquid sodium as a coolant and moderator led to additional causes of failure in SS cladding. It was noted by Smith et al. (1968) that cycles of draining the sodium and exposing the fuel cladding to air, followed by a subsequent refilling and operation at temperature, led to IGA in unstressed material and cracking in stressed austenitic SS. Na_2O and NaOH form on the metal surface as a direct result of the reaction of the air with the sodium. Intergranular attack will result from the interaction of the Na_2O with unstressed SS at room temperature and dilute NaOH with stressed material at temperatures from 250°C to 300°C (482°F to 572°F) (Andresen et al. 1989).

Mixed oxide $[(\text{U},\text{Pu})\text{O}_2]$ fuel with SS 316 cladding and irradiated in sodium in the EBR-II and the Fast Flux Test Facility (FFTF) in the temperature range of 370°C to 650°C (698°F to 1202°F) has degraded during

Table 5.2 Cladding Temperatures at Normal Operating Conditions

Fuel	Cladding Temperature, °C (°F)	Cladding Material	Reference
SM-1A	316 to 427 (600 - 800)	347 SS	Goslee (1963)
PATHFINDER	495 to 760 (923 - 1400)	316L SS	IAEA Record (1961)
BMI	?	?	
SPERT	485 (905)	304L SS	Wing (1964)
APPR (AGE 2)	316 to 427 (600 - 800)	304 SS	Goslee (1963)
VBWR	293 (560)	304L SS	Joseph (1966)
BORAX V	650 to 2100 (1202 - 3812)	304L SS	Cerchione (1966)
OMRE	427 (800)	304 SS	Trilling (1959)
FERMI BLANKET	565 (1050)	304 SS	Simnad (1971)
EBR-II	477 to 566 for Mark 1A (890 - 1050)	304, 304L, 316 SS	Simnad (1971)
TRIGA FLIP	400 to 1100 (752 - 2012)	304, 304L SS	Pers. Comm. w/ GA 1994
EBR-II (ANL-6)	660 (1220)	304 SS (?)	Simnad (1971)
TRIGA SST	400 to 1100 (752 - 2012)	304, 304L SS	Pers. Comm. w/ GA 1994
TRIGA BER-II	400 to 1100 (752 - 2012)	347 SS	Pers. Comm. w/ GA 1994

air storage.

The phenomenon, referred to as "hot cell rot," has been commonly observed when fuel pins irradiated in EBR-II or the FFTF are handled after a period of air storage.^(a) The phenomenon has also been observed by staff at Los Alamos National Laboratory (LANL) (Dowler 1983) after receiving fuel pins irradiated in EBR-II. When the shipping tube was opened the fuel pin cladding fell apart during handling.

^(a)Personal communication with B.J. Makenas of Westinghouse Hanford Company, March 30, 1994.

One of the mechanisms postulated^(a) for the "hot cell rot" experienced at FFTF and EBR involves formation of Na₂O and NaOH on the cladding surface, as mentioned above in the General Corrosion in Liquid Sodium Environments Section (Smith et al. 1968). When a fuel pin is removed from the washing station, it is generally quite bright, except for a few lightly oxidized spots. During storage in air or inert gas, a white "fuzz" forms on the surface, which in time "melts" to form a dull whitish coating on the surface of the pin. This sequence is interpreted as the formation of NaOH or sodium oxide from the residual sodium in the grain boundaries, followed by the hydrolyzation of the sodium hydroxide. Rust and pitting of the cladding often follow. The failure of the cladding is attributed to the following mechanism (though confirmatory experiments have not been performed):

"...a form of caustic stress corrosion cracking (SCC), or environmentally assisted cracking (EAC). Irradiated fuel pin cladding is generally thought to be highly stressed. Irradiation tends to reduce stress gradients through thermal and irradiation induced creep while the fuel is under irradiation in the reactor core. However, when the reactor is shut down, thermal gradients that are present during the irradiation period are relieved, creating high residual thermal stresses in the irradiated cladding and duct materials. These residual thermal stresses, as well as those resulting from irradiation induced microstructural damage, create the moderate to high stress levels needed for SCC or EAC to occur".^(b)

Embrittlement and accelerated attack of SS in the presence of caustic compounds (predominantly NaOH) is well documented (Peckner and Bernstein 1977).

Once the cladding has failed, reactions take place with the mixed oxide fuel. The fission product cesium is thought to form highly reactive cesium uranate in the region of the fuel cladding gap when the fuel is exposed to the inert hot cell atmosphere. This results in a volume expansion of the reactants which adds stress to the cladding, enlarging the breach.

"A similar reaction occurs when the MOX fuel is exposed to liquid sodium or air at elevated temperatures. The resulting reaction has been observed to cause failure propagation along the length of the fuel column, resulting in release of fuel materials and fission products."^(c)

Karnesky and Chastain have pointed out that the spent fuel from the British Prototype Fast Reactor has not experienced "hot cell rot" because it has been reprocessed and recycled, thereby avoiding the need for interim storage (Clottes et al. 1981). Also, the French successfully use monitored water storage of spent fuels (Walter 1992), recording no cladding breaches and very low cladding corrosion rates in the storage pool. The

^(a)Memo by Karnesky, R. A. and S. A. Chastain titled "The Need for Development of Improved Spent LMR Fuel Storage Methods," attached to memo from A. E. Waltar to D. M. Lucoff, titled "Transmittal of White Paper on the Long Term (Interim) Storage of Spent LMR Fuels," Westinghouse Hanford Company, Richland, Washington, dated January 29, 1993.

^(b)Memo by Karnesky, R. A. and S. A. Chastain titled "The Need for Development of Improved Spent LMR Fuel Storage Methods," attached to memo from A. E. Waltar to D. M. Lucoff, titled "Transmittal of White Paper on the Long Term (Interim) Storage of Spent LMR Fuels," Westinghouse Hanford Company, Richland, Washington, dated January 29, 1993.

United States is the only country operating a liquid metal reactor (LMR) that is known to be using dry storage of spent LMR fuels for extended periods.

Insufficient studies have been performed to definitively identify the mechanisms of the phenomenon, but carbide precipitates have been observed at grain boundaries of affected cladding. Degradation during short-term storage (a few weeks) has sometimes been attributed to trace contaminants, e.g., chlorine or chloride. Longer-term degradation (one or two years) has sometimes been attributed to residual stress and residual sodium compounds remaining after an incomplete water wash during sodium removal operations. EBR-II fuel assemblies were given a quick wash with approximately 95 L (25 gal) of water prior to the canning process, which leaves a strong possibility of sodium adhering to portions of the outside of the assemblies. EBR-II fuel will be susceptible to these degradation mechanisms attributed to the reaction of sodium with air and trace contaminants, and will require special handling to prevent rapid degradation if removed from the storage cans. In addition to SCC, IASCC, IGA, and "hot cell rot", other degradation mechanisms discussed in the following sections may operate independently or concurrently.

5.1.3 Localized Corrosion

Austenitic SS are susceptible to localized forms of corrosion such as pitting, crevice corrosion, and galvanic corrosion. Pitting of SS is found in solutions containing high levels of chlorides, as the CPP-603 has experienced during the time of the fuel storage (Figure 2.2), and is accelerated by higher temperatures. For Cl⁻ levels above 10 ppm, Types 304 and 304L SS are susceptible to pitting over long periods of water storage or in humid environments that contain those Cl⁻ levels (ASM 1990b). Visual underwater examinations of the SS clad fuel have not indicated degradation by pitting,^(a) even though the levels of the chloride have been high. This may be due to the sensitivity of the underwater examination method to finding small pits.

Pitting has been observed on LMR fuel SS cladding observed during postirradiation handling. This pitting is attributed to the caustic compounds formed when the fuel is exposed to oxygenated environments. Macroscopic examination of EBR-II fuel subject to "hot cell rot" documented by K. Dowler of LANL showed the surface of the cladding to be lightly pitted (Dowler 1983).

Galvanic corrosion arises from the galvanic coupling of SS-clad fuel with Al or CS cans used for storage as listed in Table 5.3. The SS will be noble to the Al and CS, so corrosion of the cans will accelerate preferentially to the corrosion of the SS. Large cathode to anode ratios will serve to further accelerate the corrosion of the less noble material. Corrosion due to galvanic couples potentially exists in the storage arrangements for APPR (Age 2), BMI, SPERT, VBWR, OMRE, and TRIGA FLIP fuels.

^(a)Memo from W.J. Dirk (Westinghouse Idaho Nuclear Company) to G.E. McDannel, Dirk-19-93, titled "Semi-annual Corrosion Inspection of the CPP-603 Stainless Steel 'Tie-Plate' Specimen," dated April 29, 1993.

Crevice corrosion may be exhibited for tight crevices in the fuel rod design, and occluded cells set up by crud adhering to the outside of the cladding that produce an oxygen gradient with respect to the metal. Crevice corrosion for austenitic SS is accentuated by the presence of aggressive anions such as Cl^- (Farmer et al. 1991). At high Cl^- concentrations, the fuel that has been in water storage would be vulnerable to varying amounts of crevice corrosion dependent on the tightness of the crevice. Plate geometry (exhibited by SM-1A, APPR, SPERT, VBWR, BORAX V, and OMRE) seems to be a design that would provide an opportunity for crevice formation.

5.1.4 Rapid Stress Rupture

Deformation and fracture maps for austenitic SS alloys Types 304 and 316 were applied to evaluate the allowable cladding temperatures to prevent cladding breach by stress rupture of intact cladding (cladding without incipient defects). Although deformation maps are also available for Al, only the EBR-II, MK-Ia, and MK-IIb driver fuel cladding were evaluated for stress rupture, because pressure stresses were not identifiable in other types of SNF in this study that did not employ a plenum area to accommodate fission gas releases. The TRIGA fuels do have fuel-cladding gaps where fission gas could build up, but the literature suggests that these fuels have a low fission gas release (Simmad 1981). For this reason, stress rupture for TRIGA fuels was not evaluated.

Rapid stress rupture requires hoop stresses in excess of the yield stress, approximately 210 MPa (30,000 psi) at 400°C (752°F) in Types 304 and 316 SS. This mechanism is not generally of concern during dry storage because there usually are no credible sources of primary stress sufficiently high to generate this type of cladding breach in crack-free cladding during storage. However, high localized stresses may develop from pressure stresses on regions of incipient cladding defects that are formed during irradiation. Therefore, fuel rods with incipient cladding defects have a higher susceptibility to cladding breach during dry storage. Propagation of an incipient crack to a small pin hole vents the internal gas pressure to the storage system. Venting of the fission gas relieves the pressure and terminates the cracking process. Releases of ^{85}Kr from Zircaloy-clad LWR SNF during dry storage tests at the Nevada Test Site, Morris-Illinois, and INEL were attributed to opening of pin hole breaches at sites of incipient defects (Johnson et al. 1980). Fuel particulates would not be expected to be released through a small pin hole breach. Also, stresses may approach the range for rapid stress rupture during handling operations. Some mechanical distortion of fuel assemblies (bowing and twisting) has been observed. These high stresses may propagate incipient defects. Fission gas releases observed during SNF storage tests have generally been more frequent following handling and rod consolidation operations.

The presence of incipient defects and thru-wall cracks in EBR-II SNF is well documented (Koenig and Hayner 1985a, 1985b; Olson and McClellan 1985; Olson et al. 1985). These investigations indicate a reasonably large number of EBR-II driver fuel elements have been irradiated to the failure threshold and SNF with cracked and breached fuel cladding may be common.

Table 5.3 Galvanic Couples from Storage Arrangements

FUEL	CAN MATERIAL	GALVANIC COUPLE	CONTAINMENT
CANNED			
APPR (AGE 2)	SS 304	no	none
BMI	SS	no	none
EBR-II	CS	yes	none
EBR-II (ANL-6)	SS/Al	no/yes	none
OMRE	SS	no	SS buckets
SM-1A	Al	yes	none
SPERT	Al	yes	none
TRIGA FLIP	Al	yes	none
VBWR	Al	yes	SS racks
UNCANNED			
BORAX V			SS buckets
PATHFINDER			SS buckets
TRIGA BER-II			SS MTR bucket
TRIGA SS			canister

The bounding plenum pressure prior to failure has been estimated to be 13.8 to 20.7 MPa (2000 to 3000 psi) (Olson et al. 1976). For Mark IA driver fuel, these pressures can generate cladding hoop stresses up to 189 MPa (27,500 psi).

Stresses other than pressure stresses in fuel cladding were not assessed. Other stresses, such as from in-reactor fuel swelling or fuel swelling from chemical reactions during pool storage, tend to stress the cladding beyond the yield stress and produce plastic deformation. Reactions of spent fuel constituents that could result in fuel swelling and cladding breach are presented in Appendix B, Table B.1.

5.1.5 Strain Rate Embrittlement

Strain-rate embrittlement (SRE) and triple-point cracking occur at stresses much higher than those that occur during dry storage. Based on fracture maps for SS Type 316 (Miller and Langdon 1979), maximum shear stresses higher than 105 MPa (15,200 psi) (210 MPa (30,450 psi) maximum hoop stress) would be required for SRE, whereas maximum shear stresses estimated for EBR-II Mark IA driver fuel are only 94 MPa (13,630 psi) (189 MPa (27,400 psi) maximum hoop stress). Failure strains tend to remain in excess of 0.01, even for

irradiated SS Type 316 in the triple-point cracking regime (Gilbert et al. 1987), and EBR-II cladding should not have such strains with the above stresses.

5.1.6 Creep Failure

The primary driving force for cladding failure by creep rupture is the pressure differential between the external environment and the rod internal gas pressure. This cladding failure mechanism was concluded to be the principal failure mechanism for Zircaloy-clad LWR rods (Blackburn et al. 1978; Cunningham et al. 1987). Deformation and fracture maps are particularly useful for evaluating dry storage conditions because they permit a systematic evaluation of the expected creep response in regions where the mechanisms have been identified, based on deformation and fracture theory, but where experimental data may not be available because of lengthy and expensive testing requirements.

The deformation and fracture maps for SS are presented in terms of normalized shear stress and shear strain rate (Frost and Ashby 1982; Miller and Langdon 1979). Deviatoric shear stresses and shear strain rates are defined in Frost and Ashby (1982) as follows:

For deviatoric shear strain rate,

$$\dot{\gamma} = \sqrt{\frac{2}{3}[(\dot{\epsilon}_1 - \dot{\epsilon}_2)^2 + (\dot{\epsilon}_2 - \dot{\epsilon}_3)^2 + (\dot{\epsilon}_3 - \dot{\epsilon}_1)^2]}$$

For pressurized fuel cladding where $\epsilon_1 = \epsilon_h$ and $\epsilon_2 = \epsilon_3 = 0$ (Gilbert and Blackburn 1977),

$$\dot{\gamma} = 2 \frac{\dot{\epsilon}_h}{\sqrt{3}}$$

For deviatoric shear stress,

$$\tau = \sqrt{\frac{1}{6}[(\sigma_1 - \sigma_2)^2 + (\sigma_2 - \sigma_3)^2 + (\sigma_3 - \sigma_1)^2]}$$

For pressurized fuel cladding where $\sigma_1 = \sigma_h = 2\sigma_2$ and $\sigma_3 = 0$,

$$\tau = 0.5 \sigma_h$$

where

$\dot{\gamma}$ = shear strain rate

$\dot{\epsilon}_i$ = strain rate in principal i th direction

$\dot{\epsilon}_h$ = strain in hoop direction

τ = shear stress

σ_i = principal stress in i th direction

σ_h = hoop stress

Cladding hoop stress is the principal stress used for deformation/ fracture map analysis and may be computed from pressures using the midwall formula:

$$\sigma_h = 0.5 \cdot \Delta P \cdot D_m / t$$

where σ_h = hoop stress

ΔP = difference between cladding internal and external pressures

D_m = midwall cladding diameter
= $D_o - t$

D_o = outer cladding diameter

t = cladding thickness

Bounding cladding stresses for EBR-II MK-IA fuel have been estimated by assuming a cladding external pressure of 1 atmosphere during dry storage and a maximum cladding internal gas pressure of 13.8 to 20.7 MPa (2,000 to 3,000 psi). For a cladding wall thickness of 0.023 cm (0.009 in.) and an outer diameter of 0.44 cm (0.174 in.), the peak value of 20.7 MPa (3,000 psi) generates a peak cladding shear stress of 94.34 MPa (13,680 psi) at a dry storage temperature of 400°C (752°F).

An approximate strain rate is required to identify from a deformation map the predominant deformation mechanism during storage. Investigations of mechanical properties of austenitic SSs irradiated in thermal and

test reactors show a reduction in failure strain, with a residual strain ductility of usually at least 0.01 (Olson et al. 1976; Gilbert and Harding 1969; Standring et al. 1969; Van der Schaaf 1982; Harries 1979; Beeston 1981). Tensile and crush ductility measurements of SS Type 304 from Assembly S004 irradiated in Haddam Neck (Connecticut Yankee) were on the order of 0.01 (Langstaff et al. 1982). Failure tensile strain in irradiated SS Type 304 rod cluster control assembly cladding was also on the order of 0.01 (Matsouka et al. 1993). Fracture strains above 1% (as high as 2 to 3%) are reported by Olson et al. 1976. Based on a 0.01 hoop strain or 0.0115 shear strain ductility to avoid cladding breach in less than 50 yr, the relevant steady-state hoop strain rate is $<6.3 \times 10^{-12} \text{ s}^{-1}$ and the corresponding steady-state shear strain rate is $<7.3 \times 10^{-12} \text{ s}^{-1}$.

The combination of the average shear strain rate over a 50-y dry storage period, the value of 0.00117 normalized shear stress (94.34 MPa (13,680 psi) cladding shear stress to 80,700 MPa (11.7×10^6 psi) elastic shear modulus ratio at 400°C (752°F)) identifies a position on the deformation map for Type 304 SS cladding for 100- μm (3.9 mil) average grain diameter, which corresponds to an allowable dry storage temperature of approximately 420°C (788°F). For an average grain diameter of 50 μm (2 mil), the temperature limit is approximately 400°C (752°F).

Based on the fracture map for Type 316 SS with a 40- μm (1.6 mil) average grain diameter, the temperature limit is over 500°C (932°F). These estimates of dry storage temperatures required to cause the cladding of EBR-II fuels to breach by stress rupture are considered conservative for defect-free cladding, because they assume that the temperature and pressure do not decline with the decline in decay heat.

5.1.7 Hydrogen-Induced Degradation

Most literature indicates that hydrogen embrittlement in SS is not a prevalent degradation mechanism (Forcey et al. 1988; Nelson 1974). Hydrogen is thought to have a low solubility and high mobility in austenitic SS, which reduces the trapped hydrogen in the material. This makes the austenitic SS resistant to hydrogen cracking (Johnson 1977). However, Jones (1992) has observed hydrogen-induced crack growth in irradiated 316 SS at 100°C (212°F). Therefore, the issue of hydrogen-induced crack growth has not been clearly resolved in the literature.

5.1.8 Helium Embrittlement

Helium is generated in SS in thermal reactors by (n, α) reactions with boron, nitrogen, and certain reaction products of nickel, such as ^{59}Ni and ^{58}Co . In the case of Ni, an (n, γ) reaction transmutes ^{58}Ni to ^{59}Ni which decays to ^{56}Fe by releasing an n and an α . Considering bounding LWR SS spent nuclear fuel assembly average and assembly peak fast neutron fluences of 1.0 and $1.2 \times 10^{22} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$) and total neutron fluences of 3 and $4 \times 10^{22} \text{ n/cm}^2$ ($E > 10^{-10} \text{ MeV}$), the helium content could be in excess of 100 to 200 ppm (McElroy and Ferrar 1971). The helium is mobile above 400°C (752°F) (Bauer and Wilson 1972). For irradiation temperatures above 400°C (752°F), these helium atoms cause high temperature embrittlement by

combining to form gas bubbles along dislocations and grain boundaries. The effects of irradiation on decreased ductility are factored into the evaluation of allowable temperatures to prevent creep rupture during dry storage.

5.1.9 Fission Product-Cladding Interaction

Fission product-SS cladding interaction effects have been observed in irradiated LMR fuels (Hamilton et al. 1983; Adamson et al. 1985), but has not been noted for commercial SS-clad LWR fuels (Johnson 1977). Analysis of the spent mixed uranium-plutonium fuel (MOX) used in the EBR-II reactor has shown that in the presence of oxygen cesium hydroxide and tellurium compounds will form on the inside cladding surface, and will cause degradation of the fuel cladding (Tani et al. 1986). The fission product cesium is located predominantly in the fuel-to-cladding gap and is thought to be in the form of a cesium uranate, which is highly reactive with trace amounts of oxygen. Breaches in the cladding provide an additional source of oxygen enabling the cesium hydroxide and tellurium compounds to form, even with trace amounts of oxygen and moisture as found in an "inert" hot cell environment.

5.1.10 Chemical Stability of Fuel Material Constituents

Potential or acknowledged degradation of the cladding suggests that the storage environment can interact freely with the fuel material, and more importantly, fuel material can be released to the storage environment. This indicates that the chemical stability of the actual fuel material constituents is an important issue that needs to be addressed.

For the UO_2 fuel considered for this study, the following SNF in the CPP-603 have referenced accounts of the cladding degradation: SM-1A, SPERT, APPR, and VBWR. The Pathfinder and BORAX V fuels were operated in temperature regimes that would increase the potential for sensitization and subsequent IGSCC of the cladding material. For fuel elements, rods, or plates that are failed, oxidation of the UO_2 fuel in air may occur by the following sequence: U_3O_8 powder forms, the fuel swells, the cladding splits, and the radionuclides are dispersed beyond the primary containment of the fuel cladding. Oxidation of the UO_2 fuel through the breached cladding was observed for a commercial SS-clad fuel assembly during shipment to a hot cell for analysis without a cover gas (Klingensmith 1980). The maximum cladding surface temperature during that shipment was estimated to be $\sim 300^\circ\text{C}$ (572°F). The oxidation of the fuel resulted in the requirement for an inert cover gas for future fuel shipments. The lower bound in temperature for air oxidation of UO_2 has not been determined.

In some cases the metallic and ceramic components of the fuel material may interact or react with reactive gases or water during irradiation or during periods of dry storage to cause degradation of the SNF or compounds that may compromise extended wet or interim dry storage. An example of a concern may be the formation of pyrophoric UH_3 , as discussed in Section 3. If sufficient UH_3 by reaction with the SNF pool water were to form during pool storage, then exposure to air during handling or placement into dry storage could potentially result in ignition and burning of the fuel. Thermodynamic calculations can show if there is a

chemical potential for certain products to form. However, because they provide no information on kinetics of reactions or the effects of protective films, their usefulness is very limited as a tool to predict the quantity of reaction products.

Additionally, anecdotal experience with SS-sheathed metallic uranium shielding blocks used to shield the fuel transfer port at the FFTF suggested that the reaction of the irradiated uranium with air in places on the blocks where the sheathing was compromised created a lower density reaction product that permanently deformed the blocks.^(a)

It is apparent from Appendix B, Table B.1 that there is little probability of a reaction of UO_2 from the breached SS-clad fuel cladding in the storage pools for the following fuels: APPR, BMI, BORAX V, VBWR, PATHFINDER, SM-1A, and SPERT. This indicates that fuel materials exposed to a pool storage environment would not have a tendency to form U_3O_8 powder and swell, further damaging the cladding.

As discussed in Section 4.2, UZrH fuel materials have been shown to be stable in both air and water environments over long periods of time. Degradation of the cladding of the TRIGA fuels is not expected to lead to gross deterioration of the fuel (Simnad 1981).

5.2 FUEL FAILURE MECHANISMS AND THEIR CONSEQUENCES ON DRY STORAGE

Based on our review of available information and open literature on SNF, there are ten categories of credible failure mechanisms that could operate on the SS-clad SNF in the CPP-603 and CPP-666. In an effort to prioritize the probable mechanisms the following mechanisms are likely the most problematic for the fuels stored in the basins: SCC (IGSCC and IASCC), IGA, caustic cracking ("hot cell rot"), crevice corrosion, pitting, and galvanic corrosion. All of these mechanisms would be mitigated to some degree by the selection of an inert cover gas for the long-term storage environment. As discussed in previous sections, the mechanisms listed would result from the in-reactor environment combined with the pool storage time at the reactor and at INEL, followed by a period of dry storage. The galvanic corrosion would arise from the choice of storage medium in the pool at INEL. Accordingly, the damage experienced by the fuel cladding and fuel materials must be taken into consideration with regards to fuel movement to dry storage.

The most immediate consequence of the pre-dry storage fuel failure mechanisms is the potential for fuel degradation during handling operations necessary prior to placement in dry storage. Handling operations introduce stresses not present during the water storage environment, allowing cracks to propagate through precracked surfaces. The consequences of this degradation would be loss of the primary containment of the fuel

^(a)Memo by Karnesky, R. A. and S. A. Chastain titled "The Need for Development of Improved Spent LMR Fuel Storage Methods," attached to memo from A. E. Waltar to D. M. Lucoff, titled "Transmittal of White Paper on the Long Term (Interim) Storage of Spent LMR Fuels," Westinghouse Hanford Company, Richland, Washington, dated January 29, 1993.

and loss of the structural integrity of the fuel.

The consequences of cladding and fuel failure would also dictate retrieval processes: handling, moving, repackaging, and cladding integrity verification tests. The later consequences of the failure mechanisms that degraded the fuel and fuel cladding while in the reactor, as well as in subsequent water storage, is the potential for structural integrity failure during handling operations necessary for retrieval and repackaging the fuels while in dry storage for final repository storage. This structural integrity failure can lead to slumping of the fuel assembly during handling, mechanical breakage of the fuel assembly, and releasing of fuel particulates into the storage environment. As detailed earlier, EBR-II fuel handled in air in a hot cell at LANL had lost so much structural integrity that it actually crumbled apart during handling operations. It was noted from staff communications at Los Alamos and Hanford that the only method of handling and storage that protects the structural integrity of fuel from sodium cooled reactors is confining the operations to inert gas environments (Dowler 1983).^(a)

Many of the SS clad fuels considered for this study have been canned at some point after their receipt at INEL. In many cases, the failure mechanisms discussed in Section 5.1 have produced degraded cladding that requires a secondary barrier in addition to the cladding to reduce the possibility of the fuel leakage. In the case of EBR-II (ANL-6) test, the fuel material inside the can is in the form of fragments. The concern at this point is whether the cans must be overpacked prior to storage. More concrete information about the integrity of the storage cans would be necessary to evaluate this issue. If repacking or overpacking were chosen as a preliminary step before long-term storage, it would be of value to eliminate the galvanic couples that exist between the fuel cladding and storage cans as much as is reasonable by choosing galvanically compatible materials or liners to separate the materials within the couple.

Communication with W.E. Windes of LITCO revealed that the EBR-II fuel was canned at ANL-W before it was shipped to ICPP. The atmosphere the fuel was exposed to during the canning process depended on the age of the fuel, with the older fuels canned in an air atmosphere and the later fuels canned in an argon atmosphere. The argon atmosphere contained water and oxygen levels of 60 ppm each. Formation of cesium hydroxide, a strong caustic, from a cesium-uranium-oxygen compound on the fuel surface had been observed even in argon-atmosphere hot cells with oxygen and water vapor in the ppm level.^(b) The cans were sealed with a high integrity swagelock endcap, but no leak testing has been performed on the swaged caps. Based on the information given, no conclusion can be drawn as to the integrity of each can. Due to the risk imposed on this

^(a)Memo by Karnesky, R. A. and S. A. Chastain titled "The Need for Development of Improved Spent LMR Fuel Storage Methods," attached to memo from A. E. Waltar to D. M. Lucoff, titled "Transmittal of White Paper on the Long Term (Interim) Storage of Spent LMR Fuels," Westinghouse Hanford Company, Richland, Washington, dated January 29, 1993.

^(b)Letter report by W.F. Brehm, Westinghouse Hanford Company, titled "Analysis of Material Degradation During Dry Storage of FFTF Fuel," dated February 18, 1992.

fuel from caustic cracking caused by oxygen in the fuel environment, it would be prudent to verify the canning procedure and documentation for each of the 3636 elements stored in the CPP-603 and CPP-666 pools. Additionally, the integrity of the cans will have to be investigated to determine the need for overpacking or repackaging the fuel.

Documentation from INEL states that all 93 assemblies of the SM-1A fuel were canned wet into individual SS cans fabricated from 12.7-cm (5-in.) diameter tubing. Additional documentation addresses the failed condition of significant quantities of the SM-1A fuel prior to the canning operations.^(a) Based on this information, the storage cans should be drained of the water inside the can in preparation for dry storage. This task must be undertaken with caution, to avoid draining particulate fuel matter, if it exists, into an open environment such as the CPP basins. Canisters containing other fuels may have experienced in-leakage of water, so the can drainage procedure would be an issue for those canisters also.

5.3 RECOMMENDED DRY STORAGE CONDITIONS

Corrosion rates of SS in a controlled (inert), dry environment are very low, so the storage duration would rely on the projected stability of the fuels in direct contact with the storage environment (air or cover gas). The use of a cover gas, as opposed to using an air environment, would be required to reduce the interaction of the sodium with oxygen for the liquid metal fuels, but would be prudent for the UO_2 fuels also. The UZrH fuels appear to tolerate an air environment without further degradation, so air storage would be an option. The storage environment that would provide the best protection against further degradation of the fuel cladding and fuel materials would incorporate three characteristics: a low-humidity environment; an inert atmosphere such as argon, helium, or nitrogen; and a low level of contaminants such as Cl^- .

5.4 RECOMMENDED SPENT FUEL PREPARATION OPERATIONS

The following operational issues should be considered to prepare SS-clad fuel for dry storage: 1) fuel preparation through vacuum drying, 2) choice of a drying methodology in cases where pyrophoricity is a concern, 3) choice of a cover gas for fuels identified as susceptible to rapid degradation in air, 4) overpack of leaking cans, and 5) redundancy in handling operations for fuels that are not canned and have potentially compromised cladding.

Vacuum drying is routine for Zircaloy-clad fuels prepared for dry storage and should be considered for those fuels that have been stored bare in the ICPP pools. Beyond standard preparations that are recommended for use in commercial dry storage operations (McKinnon and Deloach 1993), the issue of pyrophoricity must be considered for uranium metal fuels with degraded cladding, including the OMRE, Fermi Blanket, and EBR-II

^(a)Letter from E.P. Mondok (Exxon Nuclear Idaho) to M.J. Painter, titled "Results of SM-1A and BORAX-V Fuels Leak Tests," dated July 23, 1982.

fuels. The drying operations discussed in Section 3 for Al-clad fuels would be necessary to reduce the potential for pyrophoric reactions during fuel preparation for storage. Additionally, to reduce the potential for "hot cell rot" of the sodium-cooled reactor fuel (Fermi Blanket and EBR-II), the preparations for storage should be carried out in an inert cover gas to reduce the formation of Na_2O and NaOH on the surface and in crevices.^(a)

Bursting or blistering during the drying process is assumed to be credible only if the drying temperature causes expansion of an entrapped internal gas or liquid. This condition would require a quantity of entrapped gas or liquid in excess of that present during irradiation, because drying temperatures and times are less than irradiation temperatures and times. There does not appear to be any credible mechanism for gas generation in intact SS-clad SNF. Water that ingresses during pool storage should be released during drying without significant pressure generation. Degradation by blistering would require a chemical reaction to be activated at the drying temperature. This response requires the presence of reactants that were not present during irradiation. Sufficient hydrogen gas accumulation to blister the cladding is not expected because the permeability of hydrogen through SS is too high. Oxygen and nitrogen diffusion is too slow to lead to blistering, and hydrogen from a sodium-water reaction should have already escaped through the breach that permitted water ingress to feed the reaction. Thus, these issues do not seem significant.

If the integrity of the cans containing the fuel is suspect, the fuel should either be repackaged or the cans should be overpacked. If the fuel was canned originally due to degraded cladding and loss of structural integrity, the prudent choice would be to overpack the can to reduce the potential for particulate release during the recanning operation. Choice of the overpack can should be made with a focus on future retrieval and/or long-term disposal. Standardization of cask size and other transportation issues may need to be evaluated for a prudent choice of can overpacks to be made.

Redundancy in handling the bare fuels and suspect cans should be considered during the fuel preparation operations. Use of a secondary containment, such as buckets, should be evaluated case-by-case to reduce the potential for loss of fuel integrity and fuel materials during handling operations involving potentially degraded materials. The fuels that have been canned prior to or during their storage in the INEL storage basins should be handled with caution due to their potentially degraded state. Caution should be used in handling Al cans used to store SS-clad fuel, as the galvanic couple would cause an acceleration of the corrosion rate of the Al can. The corrosion of the Al can will affect the ability of the can to withstand stresses imposed on it during handling operations.

^(a)Memo by Karnesky, R. A. and S. A. Chastain titled "The Need for Development of Improved Spent LMR Fuel Storage Methods," attached to memo from A. E. Waltar to D. M. Lucoff, titled "Transmittal of White Paper on the Long Term (Interim) Storage of Spent LMR Fuels," Westinghouse Hanford Company, Richland, Washington, dated January 29, 1993.

6.0 CONCLUSIONS AND RECOMMENDATIONS

This study was conducted to assess the conditions of several selected spent fuels in wet storage at the CPP-603 and CPP-666 pools. Understanding the characteristics and current condition of the fuels establishes an essential basis for informed decisions regarding safe, efficient, and cost-effective methods of long-term interim storage. Some fuel types, principally the Zircaloy-clad water reactor fuels, can remain in wet storage over periods of decades without degrading. Other fuels, principally Al-clad types, are observed to be degrading in wet storage. Some fuel types are known to have breached cladding that is exposing uranium-bearing metal fuels to aqueous corrosion. Because only limited data were available on the actual condition of the fuel, estimates on the fuel condition were made in order to make an initial assessment of potential conditions for drying operations and dry storage. The conclusions drawn from the current review are provided in Section 6.1. Recommendations to improve the validity of these conclusions are provided in Section 6.2

6.1 CONCLUSIONS

The information available on factors affecting the present condition of the SNF in storage in the CPP-603 and CPP-666 was used to estimate the current condition of the spent fuel and potential failure mechanisms that would affect drying operations and dry storage of these fuels. The mechanisms that appeared significant were evaluated with the available information and ranked as shown in Table 6.1 and summarized below. The current condition of the fuel may be a result of past or ongoing mechanisms listed in Table 6.1 that may have been initiated as a result of irradiation in reactor, previous interim storage, handling, or ongoing wet storage. Because only limited information was available for many of the fuels, the validity of these rankings and their absolute levels of importance will require verification. A summary of the conclusions for the Al-clad, SS-clad, and unclad UZrH_x fuels is provided below.

The most significant cladding degradation mechanism for the Al-clad SNF is corrosion in reactor or during wet storage. Uniform corrosion, IGSCC, and the formation of incipient defects from reactor operation can also lead to embrittlement, which is considered the second most significant cladding degradation mechanism. Embrittlement in Table 6.1 is defined for all the fuels to encompass brittle failures as well as the general loss of structural integrity from significant material loss due to corrosion, which can lead to loss of fuel containment. Hydrate instability should be a minimal source of moisture during dry storage because storage temperatures are expected to be less than the instability temperature limit of 150°C (302°F). If the drying cycle has been effective, subsequent pressure buildup in the canister will be limited by the total moisture inventory available for radiolytic release during interim dry storage. If residual moisture is sufficient to cause pressures exceeding canister mechanical design, then long-term interim dry storage may result in pressures that require venting. The

Table 6.1 Degradation Mechanisms and Issues That May Impact Movement of Selected SNF from Wet to Dry Storage

Fuel (Cladding)	Cladding Degradation Mechanisms											Fuel Degradation Mechanisms/Issues			
	Corrosion	Stress Rupture	Incipient Defects	Embrittlement	Sensitization*	LTS	IGSC	IASCC	FAE	Hydrate Release	Crud Spallation	Corrosion	Pyrophoric	Fission Product Release	Fracturing
ARMF (AI)	Y1*	N		>150°C	N	N	N	N	N	>150°C	Y2	Y1	Y2	Y1	
ATR (AI)	Y1	N	Y1	>150°C	N	N	N	N	N	>150°C	Y2	Y1	Y2	Y1	
MURR (AI)	Y1	N		>150°C	N	N	N	N	N	>150°C	Y2	Y1	Y2	Y1	
ORR (AI)	Y1	N		>150°C	N	N	N	N	N	>150°C	Y2	Y1	Y2	Y1	
UW (AI)	Y1	N		>150°C	N	N	N	N	N	>150°C	Y2	Y1	Y2	Y1	
HFBR (AI)	Y1	N		>150°C	N	N	N	N	N	>150°C	Y2	Y1	Y2	Y1	
TRIGA AI (AI)	Y1	N		>150°C	N	N	N	N	N	>150°C	Y2	N	N	N	Y1
EBR-II (SS)	N	>400°C	Y7	Y1	Y1	>200°C	Y1	Y6	Y5	N	N	N	Y1	Y2	Y2
ANL-6 (SS)	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	Y2	Y1	Y2	Y3
SPEC (SS)	N		Y2	Y1	Y1	>200°C	Y1	Y3	Y1	N	Y1	N	Y1	Y1	
Fermi Cores 1 & 2 (SS)	N	NA	Y2	N	Y2	N	Y2	Y2	Y2	N		Y1	Y1	Y1	Y1
APPR (SS)	N	N			Y1	>300°C				N		N	N	N	N
BMI (SS)	N	N		Y1	Y1	>200°C				N		N	N	N	N
BORAX V (SS)	N	N		Y1	Y1	>200°C				N		N	N	N	N
VBWR (SS)	N	N	Y2	Y1	Y1	>200°C				N		N	N	N	N
Pathfinder (SS)	N	N	N	N	Y1	>200°C		N	N	N		N	N	N	
SM-1A (SS)	N	N	N	N	Y1	>200°C	Y1	N	N	N		N	N	Y1	
SPSS (SS)	N	N	N	N	Y1			N	N	N		N	N	N	
TRIGA FLIP (SS)	N	N		Y1	N	>200°C	N	N	N	N		N	N	N	Y1
TRIGA SS (SS)	N	N		Y1	N	>200°C	N	N	N	N		N	N	N	Y1
TRIGA Ber II (SS)	N	N		Y1		>200°C				N		N	N	N	Y1
AI (Unclad)	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	N	N	N	Y1
SNAP (Unclad)	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	N	N	N	Y1

The table is being developed to show the correspondence between degradation mechanisms and the affected fuel types. The table is not complete, and new information may change some of the entries that are based on limited information. The table assumes that there are no EBR-II oxide fuels in LITCO storage. FAE = Fuel Adjacency Effect; IASCC = Irradiation-assisted stress corrosion cracking. * N indicates mechanism is of low concern; Y indicates mechanism is of concern; lower numbers indicate higher order of importance. NA = Not applicable. * Y indicates that the cladding may have been sensitized during irradiation. Temperature limits indicate levels above which degradation is of concern.

need to attain adequate dryness must be accommodated in canister design and considered in selecting drying conditions. If fuel confinement is lost because of corrosion or cracking of the cladding during handling, the exposed fuel is susceptible to corrosion in wet storage or in moist environments with subsequent fission product release. Pyrophoricity of potential UH_3 appears to be of secondary concern for the Al-clad fuels because the fuel in the matrix is expected to be too dispersed to form ignitable UH_3 . Corrosion, pyrophoricity, and fission product release are considered to be unlikely for the Al-clad TRIGA fuel with UZrH_x , although the brittleness of these fuels may cause them to be susceptible to fracturing.

The most significant cladding degradation mechanism for the SS-clad SNF is IGSCC, because the irradiation temperatures for these fuels were in the range for sensitization and embrittlement. Intragranular stress corrosion cracking and the formation of through-wall as well as incipient defects is documented for some of these fuels. These degradation phenomena were not ranked high for the Fermi Cores 1 and 2, because this fuel was degraded during irradiation to the extent that no credit can be taken for the cladding, i.e., it should be treated as declad fuel. Low temperature sensitization is not considered an important degradation mechanism during storage of SS-clad SNF, because the expected dry storage temperatures are less than the 200°C (392°F) required for LTS. Furthermore, the SS types of cladding have already been exposed to sensitizing conditions during irradiation.

The fuels with a SS matrix are expected to be too dispersed to form ignitable UH_3 . The metallic EBR-II, ANL-6, and SPEC fuels may be susceptible to UH_3 formation, although the rates of formation will be slower than for uranium because of the benefits of alloying on retarding corrosion. Corrosion, pyrophoricity, and fission product release are considered to be unlikely for the SS-clad UZrH_x TRIGA SNF, although the brittleness of these fuels may cause them to be susceptible to fracturing.

Corrosion, pyrophoricity, and fission product release are considered to be unlikely for the unclad UZrH_x TRIGA SNF, although the brittleness of these fuels may cause them to be susceptible to fracturing as explained above for UZrH_x fuel with Al or SS cladding.

6.2 RECOMMENDATIONS

Based on the apparent degradation mechanisms and their importance to each of the spent fuels, an initial list of environmental conditions for acceptable pool storage, drying operations, and dry storage was made. These conditions are summarized in Table 6.2. Related information on the three categories of SNF evaluated is provided below.

6.2.1 Aluminum-Clad Fuel

The characteristics and histories of the ATR and UW Al-clad fuels were traced in some detail since these two fuels seem to bracket the Al-clad fuels addressed in this study. The ATR fuel represents a high-performance type, with some known cladding breaches, considerable corrosion in service, and storage in the aggressive CPP-603 pool water. The UW fuel represents fuel with a mild reactor service, no known cladding breaches, low decay heat, a period of at-reactor dry storage, and wet storage in the more controlled CPP-666 pool environment.

Table 6.2 Preliminary Recommendations for Storage Conditions and Limits for Selected ICPP SNF

Fuel Type	Recommended Conditions		Considerations
	Environment	Temperature, °C (°F)	
Al-Clad UAl _x , U ₃ O ₈ ● Pool Storage ● Drying ● Dry Storage	<1ppm impurities <5 torr static 99.995% He, Ar, N ₂ , or air?	<40 (<104) <300 (<572) <150 (<302)	Al corrosion residual H ₂ O corrosion, dehydration
Al-Clad UZrH _x ● Pool Storage ● Drying ● Dry Storage	<1ppm impurities <5 torr static 99.995% He, Ar, N ₂ , or air?	<40 (<104) <300 (<572) <150 (<302)	Al corrosion residual H ₂ O corrosion, dehydration
SS-Clad U Alloy ● Pool Storage ● Drying ● Dry Storage	<1ppm impurities <5 torr static 99.995% He, Ar, N ₂	<40 (<104) <200 (<392) <150 (<302)	fuel dissolution, UH ₃ U oxidation, residual H ₂ O fracturing, UH ₃
SS-Clad Cermet ● Pool Storage ● Drying ● Dry Storage	<1ppm impurities <5 torr static 99.995% He, Ar, N ₂	<40 (<104) <300 (<572) <150 (<302)	fracturing residual H ₂ O fracturing
SS-Clad UZrH _x ● Pool Storage ● Drying ● Dry Storage	<1ppm impurities <5 torr static 99.995% He, Ar, N ₂	<40 (<104) <300 (<572) <400 (<752)	fracturing residual H ₂ O fracturing, H ₂ release
UZrH _x ● Pool Storage ● Drying ● Dry Storage	<1ppm impurities <5 torr static He, Ar, N ₂ , air	<40 (<104) <300 (<572) <400 (<752)	fracturing residual H ₂ O fracturing, H ₂ release

Based on this review, candidate conditioning steps for dry storage include

- a drying cycle - This should exceed the expected storage temperature and include special provisions for failed fuel or fuel with thick films if radiolysis is expected to be a significant factor;
- a hydride conditioning cycle - If failed uranium-bearing metal fuels are involved; some analysis and characterizations are needed to define whether corroding UAl_x fuels need conditioning treatments;
- sealing in inerted containers - This includes single or double containment depending on the potential presence of pyrophoric species.

The principal considerations in the level of drying are

- to preclude long-term degradation of cladding and/or exposed fuels
- to minimize pressure buildup from radiolysis products of residual moisture, principally H_2 and O_2 .
- to minimize additional fuel damage due to excessive or aggressive drying.

The Al alloys have good corrosion resistance in moist environments and in air at elevated temperatures. If air or nitrogen cover gases are attractive, impacts of nitric acid formed by radiolysis must be forecast.

An interim maximum storage temperature of 150°C (302°F) is recommended for intact Al-clad fuels, based on mechanical property, corrosion, and fission product diffusion considerations. For uranium-bearing fuels with breached cladding, the maximum storage temperature will depend on residual moisture levels and their long-term effects on exposed fuels.

Most Al alloy fuels addressed in this study have low decay heats suggesting low storage temperatures. A reference point involves intact Japanese Al alloy test reactor fuel, removed from wet storage, dried one week in air in a hot cell, sealed in SS canisters with a helium cover gas, and stored at an estimated temperature of 45°C (113°F). Whether there is significant hydrogen buildup is not known, but gamma radiolysis would be the only significant energy source for fuel with intact cladding. If failed uranium-bearing metal fuels are involved, alpha and beta radiolysis effects must be considered.

Based on the evaluation thus far on the spent fuel conditions and known or potential failure mechanisms, two specific recommendations can be made to determine whether the initial limits proposed for the drying operations and dry storage of Al-clad SNF are valid.

- Determine whether UH_3 is present in Al-clad spent fuels and in sufficient quantity to warrant conditioning.
- Determine the amount of moisture in corrosion films and evaluate what drying procedures will be required to limit hydrogen and moisture release during drying or dry storage. If UH_3 is not prevalent or is not an issue, determine whether air storage for the Al-clad fuels is feasible.

6.2.2 Stainless Steel-Clad Spent Fuels

A large portion of the SS-clad SNF is stored in canisters. High-purity, neutral pool water is essential for future pool storability of SNF packages for up to an additional 50 yr. Radiolysis is not of concern because the irradiation fields are $<200\text{ R/h}$; however, reactive impurities such as chlorine and microbes, accelerate corrosion of Al and SS canister alloys, as well as the SNF.

Maintenance of pure and neutral pool water will prevent the accumulation of troublesome reactants. However, degradation of some canisters may require recanning. Further analyses would be required to identify the affected canisters.

The formation of UH_3 by reaction of uranium alloys with water is of concern because of its low temperature of ignition and combustion. The dispersed configuration of UH_3 that forms in the UAl_x fuels should reduce the potential for sustained combustion. The effect of the uranium-fissium alloy in reducing the significance of UH_3 formation is not known but should reduce the concern over this issue. Preparations for drying and dry storage should consider the potential for UH_3 formation to ensure that this is not an issue.

The moisture level in drying operations will be important to the spent fuel condition. It was estimated that approximately 10 g of moisture could remain after drying SNF in a 7 m³ (247 ft³) internal free volume storage system. This moisture could form approximately 70 g of UH_3 if it reacted only with uranium metal. If it is assumed that moisture would likely react at more than 100 different locations, each pocket of UH_3 should be too small (>1 g UH_3 required for ignition) for ignition. In a small fraction of the fuel inventory (i.e. 1 in 10,000), the residual moisture may react at fewer than 100 sites, and a pyrophoric pocket of UH_3 may form.

Although thermodynamic calculations show that it is energetically favorable, the extent to which UAl_x alloys react with H_2O to form UH_3 has not been experimentally verified. No specific information was obtained on the validity of uranium reaction with moisture for the fuels in the ICPP, but the uranium reaction is well known for other fuels (ITA 1994). Moisture and reactive gases will also react with surfaces of structural materials and will not be as available to react with fuel.

Primary degradation mechanisms during dry storage of SS-clad SNF were embrittlement from sensitization and SCC. This could lead to fission product release and fracturing during retrieval. Except for the metallic uranium fuel alloys, formation of UH_3 was secondary because of the dispersed configuration of the uranium. Radiolysis is not of concern because the irradiation field is <200 R/h. The potential for galvanic couples should be considered during the design of the fuel packages.

Given that the EBR-II spent fuel comprises a large portion of the spent fuel inventory, the following information should be determined to help resolve the uncertainties in the limits set for this type of spent fuel.

- Determine the former and current storage environment for the EBR-II spent fuel. It is important to know if the fuel has been exposed to moisture, and whether the cladding is significantly sensitized.
- If it appears that the EBR-II fuels are failed, verify whether UH_3 formation in the U-fissium fuel is in sufficient quantity to be a significant problem.

6.2.3 UZrH_x Spent Fuels

TRIGA and SNAP reactor fuels stored in the CPP-603 are expected to be at surface temperatures below 100°C (212°F), have insignificant radioactive gaseous product release, and, owing to the chemically inert nature of the zirconium hydride matrix, their mechanical shape should be intact. For this reason, it should be possible to store these elements safely in a dry environment after they have been removed from present wet storage, drained of water adhering on the surface, and placed inside a sealed SS canister.

The only concern in preparation for and placement in dry storage of these elements arises from the well-documented brittleness of the zirconium hydride matrix. These elements need to be handled with great care to avoid breakage. All operations concerning their retrieval from present wet storage, preparation for dry storage, and placement into dry storage need to be carefully planned with this possibility in mind. Finally, most of the data regarding the chemical and mechanical properties of these elements and regarding the safety of dry storage of these elements comes from one source, the publications and statements of the GA personnel. Though no reason exists to doubt the veracity of these data, an independent confirmation of the ease of handling these elements and the safety of their dry storage is desirable and recommended as indicated below.

- Verify whether the UZrH_x is stable in water and determine if any conditioning is justified for long-term dry storage. No definitive data were found to resolve these issues, but it would be a very important conclusion to all the fuels containing UZrH_x .
- Verify the brittleness of the UZrH_x fuel to determine whether unclad or clad fuel of this type will present a handling issue.

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APPENDIX A

**EXPANDED SUMMARY OF CURRENT GOVERNMENTAL REGULATIONS
RELATED TO SNF**

APPENDIX A

Section 1.3.3, Regulatory Guidelines, provided a brief overview of the NRC, EPA, DOT, and DOE regulations applicable to the transfer of spent nuclear fuel from wet storage to long-term (interim) dry storage. An expanded overview of these regulations is provided below, with specific pertinent citations in Tables A.1 and A.2.

Transportation: The basic ruling document is 10 CFR 71 (NRC), "Packaging and Transportation of Radioactive Material", which states that no licensed material (originally, commercial nuclear material) may be moved without authorization from the NRC, for which the requirements are set forth. These requirements include details of categories of fissile material and levels of radioactivity of the contents, with extensive instructions on determining the permissible levels for each category of package (requiring adequate knowledge of the composition of the contents); acceptable radiation levels outside the package; and packaging requirements to prevent leakage under a wide variety of scenarios, for which prescribed tests must be passed. The DOT regulation 49 CFR 173, "Shippers - General Requirements for Shipments and Packagings", has a section devoted to radioactive materials, which includes much of the contents of 10 CFR 71, but in some details goes beyond the latter. DOE 5480.3, "Safety Requirements for the Packaging and Transportation of Hazardous Materials, Hazardous Substances, and Hazardous Wastes", is based on excerpts from 10 CFR 71 and 49 CFR 173.

Treatment: The EPA regulations 40 CFR 264 and 265, respectively, "Standards for ...", and "Interim Standards for Owners and Operators of Hazardous Waste Treatment, Storage and Disposal Facilities", (almost identical documents), provide general administrative requirements from manifest systems to finances; give design, operations and leakage rates for a variety of land disposal options (tanks, landfills, incinerators); determine emergency preparedness and prevention requirements; and give details on permissible air emissions. The requirement that detailed chemical analysis be performed before any waste can be treated, stored, or disposed of is found in 264.13 and 265.13.

Storage: Two documents specifically address SNF storage, NRC 10 CFR 72, "Licensing Requirements for the Independent Storage of SNF and High-Level Radioactive Waste" and EPA 40 CFR 191, "Environmental Radiation Protection Standards for Management and Disposal of SNF, High-Level and Transuranic Radioactive Wastes". The definitive 10 CFR 72 addresses independent spent fuel storage installations (ISFSIs) for all persons in the United States and monitored retrievable storage (MRS) facilities for DOE. The EPA document 40 CFR 191 emphasizes limits on exposure to the public from any storage or disposal facility for the foreseeable future, with extensive details on estimating releases.

Disposal: NRC regulation 10 CFR 60, "Disposal of High-Level Radioactive Wastes in Geologic Repositories", prescribes rules for high-level waste disposal in a repository such as the Yucca Mountain facility, covering aspects such as licensing and license application requirements, criteria for the waste package and its components, package and repository performance and performance confirmation, stakeholder participation, training, and administration. NRC regulation 10 CFR 61, "Licensing Requirements for Land Disposal of Radioactive Waste", prescribes these aspects for LLW land disposal facilities. EPA 40 CFR 268, "Land Disposal Restrictions", lists general (RCRA-list) hazardous wastes that may not be disposed of in land facilities, and provides exemptions for materials such as farm pesticides and small quantities of various wastes. It is applicable to dry storage primarily in connection with the RCRA portion of "mixed wastes". DOE 6430.1A, "General Design Criteria", provides for such requirements as safety structures for avoiding criticality, secondary confinement barriers, temporary provisions for storage of leaky assemblies, filtration of discharge systems, and incorporation of the provisions of DOE LA10294-MS on Radiological Accident Considerations for Siting and Design..., 10 CFR 72 for storage licensing requirements, among other documents.

General Management of Hazardous Materials: The basic document establishing radiation exposure limits is the NRC document 10 CFR 20, "Standards for Protection Against Radiation". This details occupational dose limits and monitoring, control of access, respiratory protection, security of stored material, administrative procedures, and enforcement penalties. DOE 5633.3 "Control and Accountability of Nuclear Materials", describes full procedures for materials accountability and materials control. DOE 5400.3, "Hazardous and Radioactive Mixed Waste Program", provides the interpretation that whenever any hazardous waste identified in 40 CFR 261 is inadvertently mixed with any source, special nuclear, or byproduct material, the hazardous waste component is subject to regulation under RCRA. DOE 5820.2, "Radioactive Waste Management", consists of six chapters which cover High-Level Waste, Transuranic Waste, Low-Level Waste, Waste Containing Naturally Occurring and Accelerator Produced Radioactive Material, Decommissioning of Radioactively Contaminated Facilities, and a Waste Management Plan Outline.

Lists of Hazardous Materials (RCRA): The EPA 40 CFR 261, "Identification and Listing of Hazardous Waste", is the definitive document identifying the non-nuclear solid wastes which are subject to regulation under RCRA. The four characteristics which form the criteria for determining a hazardous waste are described. Extensive tables of hazardous chemicals from nonspecific sources, such as halogenated solvents used in degreasing, and from specific sources, such as bottom sediment from wood preserving processes are provided. The important information in these tables for SNF mixed waste includes the elements considered hazardous, the maximum concentration allowable for any single composite sample, and a table on Maximum Concentration of Contaminants for the Toxicity Characteristic. Sections of the Clean Air Act are codified in 40 CFR 61. This

regulation lists the substances that have been designated as hazardous air pollutants. Subpart H, "National Emission Standards for Emissions of Radionuclides Other Than Radon from Department of Energy Facilities" is the most pertinent section. Subpart H includes procedures for emissions monitoring, compliance reporting, and exemption claims for the named facilities. DOT 49 CFR 172, "Hazardous Materials Table, Special Provisions, Hazardous Materials Communications, Emergency Response Information and Training Requirements", lists and classifies the materials the DOT has designated hazardous for purposes of transportation and prescribes the requirements for shipping papers, package marking, labeling, and transport vehicle placarding for these hazardous materials.

Table A.1 Some Typical and Essential Regulations for Storage of SNF

<u>Reg. No.</u>	<u>Org.</u>	<u>Regulation Title</u>	<u>Purpose and Scope (and limitations)</u>	<u>Applicability</u>
<u>Transportation</u>				
<u>10 CFR 71</u>	NRC	Packaging and Transportation of Radioactive Material	Licensed Material > Type A; Fissile Material	All those NRC-licensed to possess, receive, store or dispose of licensed material.
<u>49 CFR 173</u>	DOT	General Requirements for Shipping and Packaging	Definitions and Classifications of Haz Mat; Requirements; Tests.	All hazardous materials transported by air, rail, water and/or highway.
<u>DOE 5480.3</u>	DOE	Requirements/Packaging and Transport of Haz Mat.	Fissile Classification; Transport Index.	All DOE entities and contractors packaging and shipping hazardous materials and hazardous waste.
<u>Treatment (stabilization)</u>				
<u>40 CFR 264</u>	EPA	Standards for Owners/Operators of Hazardous Waste Treatment, Storage and Disposal (TSD) ¹ Facilities	Minimum national standards re: disposal of hazardous waste.	RCRA primarily; radioactivity not mentioned; owner or operator of facilities
<u>40 CFR 265</u>	EPA	Interim standards for owners/operators of hazardous waste TSD facilities	Same as for 40 CFR 264	Same as for 40 CFR 264 for facilities awaiting certification of closure
<u>Storage</u>				
<u>10 CFR 72</u>	NRC	Licensing requirements for independent storage of SNF and HL Rad Waste	Requirements for license to receive, possess, transfer radioactive materials related to SNF.	Wet and dry modes of storage of SNF in an ISFSI (all US citizens) and SNF and HLRadW in an MRS (DOE only)
<u>40 CFR 191</u>	EPA	Environmental Radiation Protection Standards for Management/Disposal of SNF/HLW and TRU	Ensure that radiation doses and releases from storage/transportation of SNF are as low as reasonably achievable (ALARA).	Facilities operated by DOE and not regulated by the NRC or by Agreement states (1) Radioactive materials released into the accessible environment as a result of the disposal of SNF or high-level or TRU radioactive wastes; 2) Radiation doses received by members of the public as a result of such disposal; and 3) Radioactive contamination of certain sources of groundwater in the vicinity of disposal systems for such fuel or wastes.
<u>Disposal²</u>				
<u>10 CFR 60</u>	NRC	Disposal of HLW in Geologic Repositories	Provides rules governing licensing of DOE to receive and possess source and special nuclear material at a geologic repository (NWSA 1982)	Activities not licensed in other parts of the 10 CFR series

¹ TSD = Treatment, Storage and Disposal

² Disposal (10 CFR 60.1) means the isolation of radioactive wastes from the accessible environment

Table A.1 Some Typical and Essential Regulations for Storage of SNF

<u>Reg. No.</u>	<u>Org.</u>	<u>Regulation Title</u>	<u>Purpose and Scope (and limitations)</u>	<u>Applicability</u>
<u>10 CFR 61</u>	NRC	Licensing requirements for land disposal ¹	Procedures/criteria for land disposal of rad waste containing byproduct, source and special nuclear material from "other persons"	Disposal of waste generated by an individual is covered in 10 CFR 20. Applies to all persons in the US; not to disposal of HLW (10 CFR 60), byproduct (10 CFR 40), or to licensed material (10 CFR 20).
<u>40 CFR 268</u>	EPA	Land Disposal Restrictions	Wastes restricted from land disposal; exception to restrictions.	Persons who generate or transport hazardous waste and owners/operators of hazardous waste TSD facilities.
<u>DOE 6430.1A</u>	DOE	General Design Criteria	Requirements for various details of repository design	DOE land disposal facilities
<u>General Management of Hazardous Wastes</u>				
<u>10 CFR 20</u>	NRC	Standards for Protection against radiation	Protects against hazardous materials from NRC-licensed activities	Officially only NRC licensees; however, serves as guideline for all radioactive material activities.
<u>DOE 4633.3</u>	DOE	Control and Accountability of Nuclear Weapons	Guidelines for DOE Control and Accountability	Parallels NRC control and accountability for DOE, which is technically exempt from NRC regulations.
<u>DOE 5820.2</u>	DOE	Radioactive Waste Management	Guidelines for DOE management of HLW, LLW, TRU and radioactive mixed waste	All those subject to Atomic Energy Act and RCRA.
<u>Lists of Hazardous Materials</u>				
<u>40 CFR 61</u>	Cong	National Emissions Standards for Hazardous Air Pollutants	Lists air pollutants under various facilities categories	DOE (Subpart H) and non-DOE Federal (Subpart I) Facilities; NRC-licensed facilities
<u>40 CFR 261</u>	EPA	Identification and Listing of Hazardous Waste	Identifies RCRA solid wastes and waste characteristics	RCRA hazardous wastes (not trash); "spent waste" (contaminated and no longer usable until reprocessed); "byproduct"
<u>49 CFR 172</u>	DOT	Haz Mats Table, special provisions. Prescribes requirements for paperwork.	Lists and classifies DOE hazmat or combined means.	Each person and carrier transporting hazardous materials by air, rail, highway, water.

¹ Land Disposal (40 CFR 268.2[c]) means placement in or on the land and includes, but is not limited to, placement in a landfill, surface impoundment, waste pile, injection well, land treatment facility, salt dome formation, salt bed formation, or underground mine or cave, or placement in a concrete vault or bunker intended for disposal purposes.

Table A.2 Specific Citations Related to SNF

Transportation

10 CFR 71 - Packaging and Transportation of Radioactive Material

71.31-39 Application for Package Approval: The application to ship a package must include the following information about the package: Identification and maximum radioactivity of radioactive contents; Identification and maximum quantities of fissile constituents; Chemical and physical form; maximum normal operating pressure; maximum weight; maximum amount of decay heat; identification and volumes of any coolants; Extent of reflection, the amount and identity of nonfissile materials used as neutron absorbers or moderators, and the atomic ratio of moderator to fissile constituents.

71.47 External Radiation Standards for All Packages: A package must be designed and prepared for shipment so that the radiation level does not exceed 200 millirems per hour at any point on the external surface of the package and the transport index does not exceed 10 (see 71.4 definitions).

71.83 Assumptions as to Unknown Properties: When the isotopic abundance, mass, concentration, degree of irradiation, degree of moderation, or other pertinent property of fissile material in any package is not known, the licensee shall package the fissile material as if the unknown properties have credible values that will cause the maximum nuclear reactivity.

71.97 Advance Notification of Shipment of Nuclear Waste: Written notification of the intent to ship licensed material "to, through, or across the boundary of the state" must be sent to the governor of the state (or his designee), postmarked at least seven days prior to the seven-day period in which the shipment is expected to take place. Notable among the types of information to be provided is the description of the material, as required by DOT document 49 CFR 172.202 and 172.203 (subpart C, description of hazardous materials on shipping papers). See 71.97 (b) for radioactivity limits covered by this rule.

49 CFR 173 - Shippers - General Requirements for Shipments and Packaging

173.2 Hazardous Materials Classes and Index to Hazard Class Definitions: The class or division numbers or names and the definitions for classifying hazardous materials, including forbidden materials, are given in this section. "Radioactive materials" are Class 7.

173.2a Classification of a Material Having More Than One Hazard: (a) In general, a material having more than one hazard shall be classed according to the highest applicable hazard, except for such highly hazardous materials as explosives, peroxides, highly infectious materials and all but very small quantities of radioactive materials, which must be regulated according to both hazards, depending on the specific combination and type of hazards. See 173.421-.422.

173.3 Packaging and Exceptions: The packaging of hazardous materials for transportation by any air, rail, highway, or water must be as specified in this section.

173.21 Forbidden Materials and Packages: Details on materials that may not be shipped.

173.24 General Requirements for Packagings and Packages: Details on limits on releases, types of packaging, conformance to specifications of DOT and U.N., etc.

173.421 Limited Quantities of Radioactive Materials: Radioactive material whose activity does not exceed the limits specified in 173.423 (Table of limits based on fractions of A_1 and A_2 values) are excepted from the specification packaging, certification, marking, and labeling required by this subpart, if the package is strong, will not leak, does not exceed a radiation limit at any point on the surface of 0.5 millirems per hour, has surface contamination no greater than the limits provided in 173.443(a), and is otherwise prepared for shipment as specified in 173.421-1.

173.401-478 Subpart I - Radioactive Materials: 173.401 Scope The requirements for radioactive materials are in addition to, not in lieu of, the requirements of other sections of 49 CFR 173 and of 10 CFR 71.

173.431 Activity Limits for Type A and Type B Packages: The limits of radioactivity permitted in Type A_1 (special form radioactive material) and Type A_2 (normal form) packages shall not exceed the limits designated in 173.435 or 173.433, to which the reader is referred.

173.433 Requirements for Determination of A_1 and A_2 Values for Radionuclides: Refer to the document for tables, equations and rules for their application.

173.441 Radiation Level Limitations: Each package of radioactive material offered for shipment shall be designed and prepared so that the radiation level does not exceed 200 millirems per hour at any point on the external surface of the package and the transport index no greater than 10.

Table A.2 Specific Citations Related to SNF

173.442 Thermal Limitations: Each package of radioactive material shall be designed, constructed, and loaded so that: (a) the heat generated within the package because of the radioactive contents will not affect the integrity of the package under normal shipping conditions, and (b) the temperature of the accessible external surfaces will not exceed 50°C for other than an exclusive use shipment or 82°C for an exclusive use shipment.

173.443 Contamination Control: The limits for removable contamination from β - γ emitting radionuclides and specified uranium and thorium radionuclides must be less than 10^{-5} $\mu\text{Ci}/\text{cm}^2$ and for all other α emitting radionuclides must be less than 10^{-6} $\mu\text{Ci}/\text{cm}^2$

Various useful details may be found in the following sections, as the reader finds the need:

173.461-469 Tests - preparation for tests; tests for packaging and shielding integrity; tests for packages designed for liquids, for gases, for fissile radioactive materials, for special form materials, and for withstanding accidents.

173.474-475 Quality control for construction of packaging and required prior to each shipment of radioactive materials.

173.476 Approval of Special Form Radioactive Materials.

173.478 Notification to Competent Authorities for Export Shipments.

DOE 5480.3 - Safety Requirements for the Packaging and Transportation of Hazardous Materials, Hazardous Substances, and Hazardous Wastes

5480.3 Section 8, Package Standards - Requirements include: a) General Standards as given in 10 CFR 71 and 49 CFR 173. b) Load resistance must be such that the package can withstand a static load, normal to and uniformly distributed along its length, equal to 5 times its fully loaded weight, without generating stress in excess of its field strength. c) All considerations required to prevent criticality of the package contents. d. The package must be able to withstand reasonable transportation accident conditions.

5480.3 Section 12, Tests for Special Form Material a) Free drop through 30 feet onto an unyielding surface; b) Impact of the flat circular end of 1-inch diameter steel rod weighing 3 pounds dropped through a distance of 40 inches; c) Heating in air to 1475° Fahrenheit for 10 minutes, and d. Immersion for 24 hours in water at room temperature, water with a pH between 5.0 and 8.0 and a maximum conductivity of 10 microhms per centimeter.

Treatment and Storage

40 CFR 264 (and 40 CFR 265) Standards (and interim standards) for Owners and Operators of Hazardous Waste Treatment, Storage and Disposal Facilities: Establishes minimum national standards defining management of hazardous waste.

264.13 General Waste Analysis: Before an owner/operator treats, stores, or disposes of any hazardous (and non-hazardous under 264.113(d)) waste, he must obtain a chemical and physical analysis of a representative sample, which must contain all the information required to treat, store, or dispose of the waste. The information may be obtained from existing published or documented data on waste generated by the same or similar processes.

264.17 General Requirements for Ignitable, Reactive, or Incompatible Wastes. Precautions must be taken to prevent fire, explosions, or violent reactions; toxic mists, fumes, dusts, or gases in quantities sufficient to cause harm; damage to structural integrity of the facility; and any other situation which could threaten human health or the environment. Compliance must be documented.

264.94 Concentration Limits: The concentration of hazardous constituents in the ground-water must be below the background level and below the level given in Table I of this section, if the level in the table is below the background level. Values are given for the following elements: Arsenic, Barium, Cadmium, Chromium, Lead, Mercury, Selenium, Silver, Endrin, Lindane, Methoxychlor, Toxaphene, 2,4-D (2,4-Dichlorophenoxyacetic acid), and 2,4,5-TP Silvex (2,4,5-Trichlorophenoxypropionic acid).

264.97 General Groundwater Monitoring Requirements: A groundwater monitoring system must be put into operation, including a sufficient number of wells, situated at appropriate locations and distances apart, to provide representative samples of the ground-water content and allow for early detection of contamination. Appropriate records, statistical methods, and analysis must be applied to control contaminants and protect human health and the environment. and 264.98 Detection monitoring program: Details of ground-water detection requirements. and 40 CFR 264.99 Compliance Monitoring Program: More compliance requirements.

264 Subpart I - Use and Management of Containers - 264.171-178: Containers must be in good condition, must be inspected weekly, and must not interact with the contents or with the residue in a previously used and unclean container (compatibility requirement). Containers holding ignitable or reactive waste must be located at least 15 meters (50 feet) from the facility's property line. Storage containers containing incompatible materials must be separated from each other by a berm, dike, wall or other device.

Table A.2 Specific Citations Related to SNF

264 Subpart K - Surface Impoundments - 264.220-231: Each new or replacement impoundment must be equipped with at least two liners with leachate collection systems between the liners. Leakage limit is specified and a plan for response if the leakage rate is exceeded must be in place before waste can be accepted. Special requirements are stipulated for wastes which are ignitable, reactive or incompatible.

264 Subpart AA - Air Emission Standards for Process Vents - 264.1030-1-37: This subpart applies to process vents associated with distillation, fractionation, thin-film evaporation, solvent extraction, or air or steam stripping operations that manage hazardous wastes with organic concentrations of at least 10-ppmw, if these operations are conducted in, among other things, hazardous waste recycling units that are located on hazardous waste management facilities...Detailed requirements for emission limits for a variety of emission control devices (open- and closed-vent) are listed. Auxiliary software (which is not included here) is used for heating value and maximum allowed velocity of the gases. Requirements are given for monitoring the input to the process.

10 CFR 72 - Licensing Requirements for the Independent Storage of Spent Nuclear Fuel and High-Level Radioactive Waste

72.24 Contents of application: Technical information: The application must include a Safety Analysis Report (SAR) with a description of the design and operating characteristics of the ISFSI or MRS in full detail, as outlined in this section.

72.26-34 Additional contents of application include qualification of the application, training of the personnel, decommissioning planning, emergency planning, and an environmental report meeting the requirements of 10 CFR 51, Part A.

72.44 License Conditions: (a) License conditions are based on the SAR, and pertain to design, construction, operation and any additional conditions the Commission finds appropriate. (c) The license must include technical specifications, which must in turn include: (1) Functional and operating limits and monitoring instruments and limiting control settings; (2) Limiting conditions - lowest operating performance levels of equipment required for safe operation; (3) Surveillance requirements; (4) Design features; (5) Administrative controls - full management procedures.

72.46-52 Requirements for public hearings; Changes, tests and experiments; Transfer of license; and Credit regulations.

72.70-86 Subpart D - Records, Reports, Inspections, and Enforcement required - See full details.

72.90-108 Subpart E - Siting Evaluation Factors: See in particular 72.100 Defining potential effects of the ISFSI or MRS on the region; 72.104 Criteria for Radioactive Materials in Effluents and Direct Radiation from an ISFSI or MRS; and 72.106 Controlled Area of an ISFSI or MRS; also:

72.96 Siting Limitations: (a) An ISFSI owned and operated by DOE must not be located at any site within which there is a candidate site for a HLW repository. (b) An MRS must not be located in any State in which there is located any site approved for site characterization for a HLW repository. (c) If an MRS is located within 50 miles of the first HLW repository, the quantity of spent fuel or HLW that may be stored in both the repository and the MRS must be limited to 70,000 metric tons of heavy metal or the quantity of HLW resulting from the reprocessing of that quantity of spent fuel until the second repository is in operation.

72.120 General Design Criteria: The design criteria must be included in the application, and must include the design, fabrication, construction, testing, maintenance, and performance requirements for the entire system.

72.122 Overall Requirements: Quality Standards; Protection against environmental conditions and natural phenomena, fires and explosions; testing and maintenance of systems and components; Emergency capability; confinement barriers and systems; Instrumentation and control systems.

72.124 Criteria for Nuclear Criticality Safety;

72.126 Criteria for Radiological Protection;

72.128 Criteria for Spent Fuel, HLRadW and Other Radioactive Waste Storage and Handling.

40 CFR 191 - Environmental Radiation Protection Standards for Management and Disposal of SNF, HLW and TRU

(This regulation does not apply to the Yucca Mountain site, but it can be used as guidance while the regulations for Yucca Mountain are being written.)

40 CFR 191.13 - Containment Requirements: Releases to the environment for 10,000 years after disposal shall have a likelihood of less than one chance in 10 of exceeding the listed quantities for the listed radionuclides or one chance in 1000 of exceeding 10 times the quantities listed in Table 3 (of 101-13).

Table A.2 Specific Citations Related to SNF

40 CFR 191.14 - Assurance Requirements:

(a) Active institutional controls over disposal sites should be maintained for as long a period of time as is practicable after disposal; however, performance assessments that assess isolation of the wastes from the accessible environment shall not consider any contributions from active institutional controls for more than 100 years after disposal...(b) Disposal systems shall be monitored after disposal to detect substantial and detrimental deviations from expected performance until there are no significant concerns. (d) Both natural and engineered barriers shall be used to isolate the wastes from the environment.

40 CFR 191.15 - Individual Protection Requirements: "Disposal systems for spent nuclear fuel or high-level or transuranic radioactive wastes shall be designed to provide a reasonable expectation that, for 1,000 years after disposal, undisturbed performance of the disposal system shall not cause the annual dose equivalent from the disposal system to any member of the public in the accessible environment to exceed 25 mrem to the whole body or 75 mrem to any critical organ. All potential pathways (associated with undisturbed performance) from the disposal system to people shall be considered, including the assumption that individuals consume 2 liters per day of drinking water from any significant source of ground water outside of the controlled area."

40 CFR 191.16 - Groundwater Protection Requirements: Ground water protection requirements, with 1,000-year pCi/L limits, have been remanded and are being rewritten.

40 CFR 191.17 - Alternative Provisions for Disposal: Alternative provisions must be submitted for public comment which must be fully considered in developing the final version of the alternative.

40 CFR 191 Appendix B Guidance for Implementation of Subpart B: This Appendix provides details on the procedures to use in determining acceptable limits for emission releases.

Disposal

10 CFR 60 - Disposal of High-Level Radioactive Wastes in Geologic Repositories

This regulation prescribes rules governing the licensing of the U.S. DOE to receive and possess source, special nuclear, and byproduct material at a geologic repository operations area in accordance with the Nuclear Waste Policy Act of 1982. Spent nuclear fuel, as defined in 10 CFR 72.3, "includes special nuclear material, byproduct material, source material and other radioactive materials associated with fuel assemblies" and is specifically defined as High-Level Radioactive Waste in 10 CFR 60.2

60.15-18 Site Characterization: Full details are given.

60.21-23 Content of Application: Pages of details are given.

60.43 License Specification: The ability to obtain a license requires certain characterization, as there are restrictions and conditions on the physical and chemical form and radioisotope content of the licensed material.

60.46 Particular Activities Requiring License Amendment, and

10 CFR 111 (b) Retrievability of waste. Disposal under 10 CFR 60.46 (a)(1) and 60.111 (b) requires that the material be retrievable, inasmuch as a license amendment is required for any action that would make the emplaced HLW irretrievable or difficult to retrieve during the first 50 years after emplacement.

60.102 (e) Isolation of waste and

60.113 (a)(1) Engineered barrier system. The licensed materials must be contained within the disposal package without leaking for at least the first 300 years.

60.131 General design criteria for the geologic repository operations area. (b)(7) Criticality Control All systems involving the materials to be placed in the repository must be designed to avoid criticality. This implies knowledge of the characteristics of the material to be deposited.

60.135 Criteria for the waste package and its components (b) Specific criteria for HLW package design -- (1) The waste package shall not contain explosive or pyrophoric materials or chemically reactive materials in an amount that could compromise the ability of the underground facility to contribute to waste isolation ...(2) The waste package shall not contain free liquid in an amount that could compromise the ability of the waste packages to contain the HLW. (4)(c) All radioactive wastes shall be in solid form. Particulate wastes shall be consolidated into an encapsulating matrix...Combustible radioactive wastes shall be reduced to non-combustible form unless it can be demonstrated that a fire would not compromise the waste package integrity...

Table A.2 Specific Citations Related to SNF

60.140 General Requirements: There must be a performance confirmation program, started during site characterization and continued until permanent closure. It will involve in situ monitoring and laboratory tests to ensure that changes in the environment due to the repository are within license specifications. The monitoring likewise must not adversely affect the performance requirements.

60.143 Monitoring and testing waste packages: A program for monitoring waste packages under the conditions of the repository, consistent with safe operation of the repository and including laboratory tests focussing on the internal condition of the packages, shall continue until the time of permanent closure.

10 CFR 61 - Licensing Requirements for Land Disposal of Radioactive Waste

61.3 License required: (a) No person may receive, possess, and dispose of radioactive waste containing source, special nuclear, or byproduct material at a land disposal facility unless authorized by a license issued by the Commission pursuant to this part...

61.7 Concepts: This applies to disposal in the top 30 meters of the earth, unless deeper burial is deemed satisfactory in the future. The purpose is to protect the general population from releases of radioactivity and from intrusion and to provide stability of the waste and the disposal site to minimize the access of water to the waste. Waste that will not decay to acceptable hazard levels to an intruder within 100 years is Class C and will be buried at greater depth. The effective life of intruder barriers should be 500 years, at which time the waste will have decayed to a level that is an acceptable hazard.

61.10 Content of Application: An application to receive from others, possess and dispose of wastes containing or contaminated with source, byproduct or special nuclear material by land disposal must consist of general, specific technical, institutional, and financial information as set forth in Sections 61.11 through 61.16 (which see).

Containing pages of details with which the licensee must comply are:

61.23 Standards for Issuance of a License:

61.24 Conditions of License:

61.41 Protection of the General Population from Releases of Radioactivity: Concentrations of radioactive material which may be released to the general environment in ground water, surface, water, air, soil, plants, or animals must not result in an annual dose exceeding an equivalent of 25 millirems to the whole body, 75 millirems to the thyroid, and 25 millirems to any other organ of any member of the public. Reasonable effort should be made to maintain releases of radioactivity in effluents to the general environment as low as reasonably achievable.

61.52 Land Disposal Facility Operation and Disposal Site Closure. This section provides details of the depth and segregation required for waste packages according to general waste classifications A through C. These classifications are defined in detail in 10 CFR 61.55.

40 CFR 268 - Land Disposal Restrictions

268.2 Definitions Applicable in this Part. Land disposal includes concrete vaults or bunkers.

268.3 Dilution Prohibited as a Substitute for Treatment. Dilution is not acceptable as a substitute for treatment.

268.30-35 Subpart C Prohibitions on Land Disposal. Specific definitions of hazardous non-radioactive waste types are given with correlated specific restrictions for land disposal and standards for treatment. These restrictions might apply to wastes from certain separation, leaching and other disposal processing steps.

Table A.2 Specific Citations Related to SNF

268.40-45 Subpart D Treatment Standards. Treatment standards are expressed in a variety of ways.

268 Appendices. Appendices give Toxicity Characteristic Leaching Procedure (I), A list of Regulated Organic Compounds (III), and Recommended Technologies to Achieve Deactivation of Characteristics (VI), to name the most pertinent-looking of them.

10 CFR 20 - Standards for Protection Against Radiation

This section sets forth the standards to be used in NRC licensed facilities for protecting the public.

20.1201 Occupational Dose Limits for Adults. Given in great detail. A few highlights follow. (a) The licensee shall control the occupational dose to individual adults, except for planned special exposures under Section 20.1206, to the following dose limits: (1) An annual limit, which is the more limiting of-- (i) The total effective dose equivalent being equal to 5 rems (0.05 Sv); or (ii) The sum of the deep-dose equivalent and the committed dose equivalent to any individual organ or tissue (other than the lens of the eye) being equal to 50 rems (0.5 Sv).

(2) The annual limits to the lens of the eye, to the skin, and to the extremities, which are: (i) An eye dose equivalent of 15 rems (0.15 Sv), and (ii) A shallow-dose equivalent of 50 rems (0.50 Sv) to the skin or to any extremity.

20.1202 Compliance with Requirements for Summation of External and Internal Doses.

20.1203-1208 Determination of External Dose from Airborne Radioactive Material; Internal Exposure; Planned Special Exposures; Occupational Dose Limits for Minors; and Dose to Embryo/Fetus.

20.1301 Dose Limits for Individual Members of the Public. (1) The total effective dose equivalent to individual members of the public from the licensed operation does not exceed 0.1 rem (1 mSv) in a year, exclusive of the dose contribution from the licensee's disposal of radioactive material into sanitary sewerage in accordance with Section 20.2003; and 2) The dose in any unrestricted area from external sources does not exceed 0.002 rem (0.02 mSv) in any one hour; b) If the licensee permits members of the public to have access to controlled areas, the limits for members of the public continue to apply to those individuals; and c) A licensee or license applicant may apply for prior NRC authorization to operate up to an annual dose limit for an individual member of the public of 0.5 rem (5 mSv). For the information to be included in the application and additional details, the reader is referred to the document.

20.1501 General. (a) Each licensee shall make or cause to be made, surveys that-- (1) May be necessary for the licensee to comply with the regulations in this part; and (2) Are reasonable under the circumstances to evaluate -- (i) The extent of radiation levels; and (ii) Concentrations or quantities of radioactive material; and (iii) The potential radiological hazards that could be present. For the details, the reader is referred to the document.

20.1801 Security of Stored Material.

20.1802 Control of Material Not in Storage.

20.2001(b) General Requirements. A licensee must be specifically licensed to receive waste containing licensed material for: 1) treatment prior to disposal, 2) treatment or disposal by incineration, 3) decay in storage, 4) disposal in a land disposal facility licensed under 10 CFR 61 or 5) disposal at a geologic repository under 10 CFR 60.

20.2002 Method for Obtaining Approval for Proposed Disposal Procedures "...for approval...to dispose of licensed material generated in the licensee's activities. Each application shall include: (a) A description of the waste containing licensed material to be disposed of, including the physical and chemical properties important to risk evaluation, and the proposed manner and conditions of waste disposal;..."

20.2005 Disposal of Specific Wastes Material used for liquid scintillation counting may be disposed of as if it were not radioactive if it contains 0.05 μCi or less of ^3H or ^{14}C per gram of medium.

20 APPENDIX B This appendix introduces and defines the complex terms ALI and DAC, which may be useful in the dry storage context.

Table A.2 Specific Citations Related to SNF

DOE 5633.3 Control and Accountability of Nuclear Materials

DOE 5633.3 Chapter I provides minimum requirements for the control and accountability of nuclear materials. The level of control and accountability shall be consistent with the economic and strategic value of these materials. The minimum reportable quantities are given in Table 8 of this chapter.

DOE 5633.3 Chapter I Section Graded Safeguards. "Graded safeguards" is the concept of providing the greatest relative amount of control and effort to the types and quantities of SNM that can be most effectively used in a nuclear explosive device. The grade considerations of specific configurations are provided in Figures 1-2, to which the interested reader is referred.

DOE 5633.3 Chapter II Materials Accountability, Section 4 Measurements and Control provides details on the inventory measurement methods, statistics and controls.

DOE 5820.2 Radioactive Waste Management

DOE 5820.2 Chapter I, High-Level Waste, Section 3 Requirements HLW will be considered to be radioactive mixed waste and subject to AEA and RCRA. Most requirements listed are concerned with tanks.

DOE 5820.2 Chapter II, Management of Transuranic Waste, Section I Concerned with Waste Isolation Pilot Plant. TRU also assumed to be radioactive mixed waste. Section 3 - Radioactive wastes with quantities of TRU in concentrations of 100 nCi/g or waste or less shall be considered to be LLW.

DOE 5820.2 Chapter III, Management of LLW, Section 3.d LLW must be characterized with sufficient accuracy to permit proper segregation, treatment, storage, and disposal, to ensure that the actual physical and chemical characteristics and major radionuclide content are recorded and known during all stages of the waste management process.

Section 3.i.5a For all types of containers, void spaces within the waste and between the waste and its packaging shall be reduced as much as practical.

Section 3.i.5b Liquid wastes, or wastes containing free liquid, must be converted into a form that contains as little freestanding and noncorrosive liquid as is reasonably achievable, but in no case shall the liquid exceed 1% of the volume of the waste in the disposal container, or 0.5% of the volume of the waste processed to a stable form.

DOE 5820.2 Chapter IV, Management of Waste Containing AEA Section 11e(2) Byproduct Material and Naturally Occurring and Accelerator Produced Radioactive Material, Section 3 Waste containing RCRA-defined hazardous materials shall be managed in accordance with RCRA.

DOE 6430.1A - General Design Criteria

Coverage of many aspects of storage facilities requirements. The reader should consult the entire document. Some aspects are not covered in 10 CFR series and other regulations; e.g.:

DOE 6430.1A, Section 1320, Irradiated Fissile Material Storage Facilities (IFMSFs) specifies the additional requirements above and beyond those in other DOE and NRC guidelines.

Section 1320-5 mandates confinement requirements, similar to those in 10 CFR 72(h)(1).

Lists of Hazardous Materials

40 CFR 61 - National Emission Standards for Hazardous Air Pollutants

40 CFR 261 - Identification and Listing of Hazardous Waste

49 CFR 172 - Hazardous Materials Table, Special Provisions, Hazardous Materials Communications, Emergency Response Information, and Training Requirements

APPENDIX B

THERMODYNAMIC CALCULATIONS AND FACTORS FOR SELECTING A COVER GAS

APPENDIX B

THERMODYNAMIC CALCULATIONS AND FACTORS FOR SELECTING A COVER GAS

Whether the SNF is degraded or not, control of the storage environment is important to ensure acceptable dry storage of SNF. Selection of an appropriate cover gas and control of the moisture and temperature of the storage environment are important operational requirements. Thermodynamic calculations have been made in this study to assess a variety of potential reactions of the SNF with moisture and other storage media. However, because the chemistry equilibrium calculations used for this study do not provide information on kinetics or the effects of microstructural details such as reaction inhibiting films, their usefulness is very limited as a tool to predict the quantity of reaction products. A review of cover gases is also presented to summarize past experience with commercial spent fuel that provides some insight on conditions that affect acceptable storage.

B.1 CHEMICAL STABILITY OF SNF CONSTITUENTS

Some metallic and ceramic components of SNF may interact or react with reactive gases or water during irradiation or during periods of dry storage to cause degradation of the SNF or compounds that may compromise extended interim storage. An example of a concern may be the formation of pyrophoric UH_3 . If sufficient UH_3 by the reaction of uranium or UAl_x with water or hydrogen were to form during pool storage, then exposure to air during handling or placement into dry storage could potentially result in ignition and burning of the fuel. However, if oxygen were present in the storage system, there is a reverse reaction, as shown by Equation B27 for the UH_3 to react with the oxygen to form UO_2 . In addition to the reaction of UH_3 with oxygen to form UO_2 , Equation B26 indicates that UH_3 also reacts with water to form UO_2 . Consequently, when it is expected that a metallic uranium alloy or UAl_x fuel has been exposed to water, some characterization of the fuel may be required to verify its condition.

While the thermodynamic calculations do not account for kinetics, formation of oxide films, or the effects of physical or chemical changes in the fuel material during irradiation or storage, these calculations have been made to provide a starting point for this evaluation. The thermodynamic calculations were performed with the thermodynamic code, Outokumpu HSC Chemistry for Windows, Version 1.10 (Outokumpu 1993). One of the outputs of this code is the free energy of formation. A negative free energy of formation indicates that a reaction is thermodynamically favorable. A positive free energy indicates that the reaction is not favorable. The free energy of formation provides no information on the kinetics of the reaction. This version of Outokumpu does not provide information on the kinetics or rate of reaction. Very little product may form from a reaction with a negative free energy of reaction if the kinetics are negligibly slow or if a protective barrier film is formed to separate the reactants.

Results of thermodynamic calculations are summarized in Table B.1. Also included in Table B.1 are

columns to show the reactants, potential products of the reaction, and potential concerns. The reaction equations are shown at the end of this appendix and are identified by the reaction equation numbers shown in brackets; these equation numbers are also shown in Table B.1. Thermodynamic calculations for the reaction of zirconium-clad UO_2 commercial SNF with water indicate that UH_3 formation is thermodynamically unfavorable. In agreement with this calculation, and as shown for Equation B2 in Table B.1, UH_3 has not been a problem during pool storage of commercial UO_2 SNF clad with zirconium alloys. Furthermore Equations B2, B3, B4, B5, applicable to the APPR, BMI, Borax V, VBWR, Pathfinder, SM-1A, and SPSS fuels, indicate that reactions of UO_2 with water to oxidize the UO_2 or with water or hydrogen to form UH_3 are not energetically favorable. These calculations also agree with experience in storing commercial fuel with breached cladding.

Calculations for exposure of ARMF, ATR, MURR, ORR, UW, and HFBR fuels in water indicate in Table B.1 that the free energy of reaction to form UH_3 is negative. Although it is expected that the microstructural features of these alloys would prevent pockets of UH_3 to form that are large enough to support a pyrophoric reaction, some characterization work may be needed to verify that it does not form or that its presence is not a concern.

Table B.1 SNF Reactants and Potential Products Based on Calculations with Outokumpu Computer Code

Fuel Type	Equation/ Reactants	Products Evaluated	Potential Concern
Commercial UO ₂ SNF clad with Zirconium alloy	[B1, B2, B3, B4, B5] UO ₂ , Zr, H ₂ O	UH ₃ , UO _{2+x} , H ₂ , O ₂	$\Delta G > 0$; No Concern
APPR, BMI, BORAX V, VBWR, PATHFINDER, SM-1A, SPSS	[B2, B3, B4, B5] UO ₂ , H ₂ O, H ⁺	UH ₃ , UO _{2+x} , H ₂ , O ₂	$\Delta G > 0$; No Concern
ARMF, ATR, MURR, ORR, UW, HFBR	[B13, B17, B21] UAl ₄ , UAl ₃ , UAl ₂ , H ₂	UH ₃ , Al	$\Delta G > 0$; No Concern
ARMF, ATR, MURR, ORR, UW, HFBR	[B11, B15, B19] UAl ₄ , UAl ₃ , UAl ₂ , H ⁺	UH ₃ , Al	$\Delta G < 0$; Pyrophoric
ARMF, ATR, MURR, ORR, UW, HFBR	[B12, B16, B20] UAl ₄ , UAl ₃ , UAl ₂ , H ₂ O	UH ₃ , Al ₂ O ₃ , H ₂	$\Delta G < 0$; Pyrophoric H ₂ Concentration
ARMF, ATR, MURR, ORR, UW, HFBR	[B13, B17, B21] UAl ₄ , UAl ₃ , UAl ₂ , H ₂	UH ₃ , Al	$\Delta G > 0$; No Concern
ARMF, ATR, MURR, ORR, UW, HFBR	[B14, B18] UAl ₃ , UAl ₂ , H ₂ O	UH ₃ , Al ₂ O ₃ , UO ₂ , H ₂	$\Delta G < 0$; Pyrophoric
ARMF, ATR, MURR, ORR, UW, HFBR	[B6] UO ₂ , Al, H ₂ O	UH ₃ , Al ₂ O ₃	$\Delta G < 0$; Pyrophoric
ARMF, ATR, MURR, ORR, UW, HFBR	[B7, B8, B9, B10] UO ₂ , Al	UAl ₄ , UAl ₃ , UAl ₂ , U, Al ₂ O ₃	$\Delta G < 0$; Swelling
HFBR	[B22] U ₃ O ₈ , Al	UAl ₃ , Al ₂ O ₃	$\Delta G < 0$; Swelling
EBR-II, ANL-6, SPEC, FERMI Cores I & II	[B23] U, H ₂ O	UH ₃ , H ₂ , UO ₂	$\Delta G < 0$; Pyrophoric H ₂ Concentration
ARMF, ATR, MURR, ORR, UW, HFBR,	[B28] Al ₂ O ₃ •H ₂ O	Al ₂ O ₃ , H ₂ O	$\Delta G > 0$; No Concern
ARMF, ATR, MURR, ORR, UW, HFBR,	[B29] Al ₂ O ₃ •3H ₂ O	Al ₂ O ₃ , H ₂ O	$\Delta G < 0$ for T>150°C Moisture Release

A graphical view of the temperature dependence of the free energy of formation is shown in Figures B.1 through B.6. Positive values for the free energy of formation indicate that the reaction is unlikely. Energetically favorable reactions have negative values for the free energy of formation.

Thermodynamic computations indicate that UO_2 and U_3O_8 , potential products of reaction of ARMF, ATR, MURR, ORR, UW, and HFBR fuels with water, may further react with Al to form UH_3 (Equation B6) or to form other uranium-aluminum intermetallic compounds and Al_2O_3 (Equations B7, B8, B9, and B10.). The change in free energy for Equations B7, B8, B9, and B10 indicate that the reactions are only slightly favored; oxide films and kinetics may further inhibit all of these reactions. In-reactor fuels testing has verified some of these reactions (Francis et al. 1962). Again it should be noted that these calculations do not show the quantity of reaction product nor do they show synergistic effects with reverse chemical reactions.

Thermodynamic calculations for the water reaction with metallic uranium in EBR-II, ANL-6, SPEC, FERMI Cores I & II fuels indicate in Table B.1 that the formation of UH_3 is favorable. Whether UH_3 has formed or not will depend on the extent of water exposure of the fuels and the kinetics of reaction. Some characterization work may be required to resolve the questions unless the knowledge of the storage conditions can rule out the possibility that these fuels were exposed to water.

B.2 FACTORS FOR SELECTING A COVER GAS AND IMPACT ON DRY STORAGE

The three most common gases for inerting spent nuclear fuel are helium, argon, and nitrogen (Wheeler 1984). Less common gases that have been used are carbon dioxide and neon. Helium is popular because of its high thermal conductivity and its high susceptibility for detection with mass spectrometers. These features reduce temperature gradients in storage systems and provide a high sensitivity for detecting and locating leaks. Argon is easier to contain than helium, but offers a lower thermal conductivity and lower sensitivity for leak detection. Nitrogen is the most common gas, is easy to contain, but compared to helium provides a lower thermal conductivity and lower sensitivity for leak detection. A summary is provided below on the source of moisture in dry storage and potential degradation during dry storage in cover gases with residual moisture.

Figure B.1 Free Energies of Formation for Uranium in Water

Figure B.2 Free Energies of Formation of UAl_2 with Water and Hydrogen

Figure B.3 Free Energies of Formation for UAl_3 with Water and Hydrogen

Figure B.4 Free Energies of Formation for UAl_4 with Water and Hydrogen

Figure B.5 Free Energies of Formation for UO_2 with Water, Hydrogen, and Zirconium and U_3O_8 with Aluminum

Figure B.6 Free Energies for Decomposition of $\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$ and $\text{Al}_2\text{O}_3 \cdot 3\text{H}_2\text{O}$ Versus Temperature

B.2.1 Sources of Moisture in Dry Storage

Moisture in dry storage containment is of concern because it is a potential source of reactants that can degrade SNF and may generate pyrophoric UH_3 under certain conditions. The source of moisture in a dry storage system can be residual impurities, impurities from the backfill gas, outgassing of structural and fuel assembly materials, fuel surface hydrates, and water-logged fuel. Each of these sources is discussed below.

Estimates considered in this section are based on residual moisture introduced from SNF and associated hardware and a total gas volume of $<7 \text{ m}^3$ at $<1.5 \text{ atm}$ absolute pressure in dry storage system types listed in Table B.2. These estimates were provided for a commercial dry storage cask system (Knoll and Gilbert 1987). Actual numbers will vary with the storage system as well as with the design and configuration of the SNF packages. The quantity of gas contained in a 7-m^3 volume at a pressure of 1.5 atm at 300 K is 426 moles. Reactive impurities present in the helium cover gas include O_2 , H_2 , CO_2 , CO , and H_2O . In dry storage cask tests containing commercial Zircaloy-clad SNF, each of the above impurities is typically present at concentrations $<0.1 \text{ vol}\%$ and the total impurity gas concentration was $<0.2 \text{ vol}\%$. Typical cover gas composition in three different operating casks is shown in Table B.2.

Table B.2 Typical Cover Gas Composition in Casks Storing Commercial SNF

Cask Type	Concentration, vol%							
	He	N_2	O_2	Ar	CO_2	CO	H_2O	H_2
TN-24P ^(a)	>99.8	<0.06	<0.02	<0.01	<0.01	<0.01	~ 0.10	<0.01
MC-10 ^(b)	>99.8	<0.10	<0.02	<0.01	<0.01	0.01	<0.01	<0.01
MSF IV ^(c)	>99.7	<0.25	<0.06	<0.01	<0.01	<0.01	0.04	--

^(a) Creer et al. 1987

^(b) McKinnon et al. 1986

^(c) McKinnon et al. 1987

Because there are numerous sources for gas impurities, including moisture, the application of a certified high purity cover gas such as high-purity helium (99.995%) may not be required. Gas impurities and moisture from outgassing of the numerous surfaces as well as the residual moisture contained in the fuel packages will be larger than the contaminants introduced from using welding-grade helium (99.995 but uncertified). The total oxidizing gas concentration (O_2 , CO_2 , and CO) in the containment system from all sources will likely be $<0.25 \text{ vol}\%$ after implementing a typical industrial type procedure for drying and implementing SNF into an inerted dry storage system.

In addition to residual impurities remaining in the cask after evacuation, impurity gases including residual moisture are introduced into the containment after evacuation and backfill because of outgassing from the structural materials and fuel assemblies, the backfill gas contains residual impurity gases, and impurities are introduced during gas transfer operations. Gaseous impurities from inleakage during storage is negligible (Knoll and Gilbert 1987).

Residual Impurities

Cask operating manuals (Kingsley 1985; Transnuclear 1985; Westinghouse 1985) specify a minimum pressure that must be sustainable during evacuation of the loaded casks after the vacuum drying operations are completed and before the final helium or cover gas backfill. This pressure varies from 4×10^{-4} MPa (0.004 atm) for the MC-10 cask to about 1×10^{-4} MPa (0.001 atm) for the TN-24P cask. The higher pressure is associated about 1.2 moles residual gas in a 7-m^3 (247 ft^3) cask volume. Only about half of this gas consists of reactive gases and the other half consists of inert gases remaining from purging with inert gas. Therefore the actual reactive gas concentrations are $<0.2\text{ vol}\%$, corresponding to ~ 0.6 moles (0.14 vol%) in a 7-m^3 (247 ft^3) cask volume. A key point is that this estimate requires that the pressure be stable. An increasing pressure may indicate that drying is incomplete and additional gases would be released to the storage atmosphere over an extended time period.

Impurities from Backfill Gas

A welding-grade helium (purity of 99.995% but uncertified) may introduce up to 0.005 vol% (50 ppm by volume) of gas impurities. Only a fraction of these impurities are reactive gases (Air Products 1987).

Outgassing of Structural and Fuel Assembly Materials

Cask materials are designed and specified to minimize surface porosity, cavities, and roughness that are trapping sites for adsorbed gasses. This is consistent with the observation from Table B.2 that the measured impurity levels appear to consist primarily of the residual impurities and very little margin is remaining for impurities from backfill gas or outgassing.

Fuel Surface Hydrates

Decomposition of hydrates in the surface oxides of stainless steel cladding is considered to be a negligible potential source for moisture in dry storage containment. Moisture release from decomposition of hydrates in Al-clad SNF should be negligible according to Equations B28 and B29 of Table B.1, if dry storage temperatures are maintained below 150°C (302°F). Because of the low decay heat remaining in the Al-clad fuels, dry storage below 150°C (302°F) may be a reasonable expectation.

Water-Logged Fuel

Investigations on the release of water by nonirradiated UO_2 pellets by Srivastava and Olander (1978) showed that temperatures above 600°C (1112°F) were required to release the bound moisture. Therefore, this mechanism is not expected to contribute to moisture release during dry storage at the lower anticipated dry storage temperatures.

Investigations on the release of water from water-logged commercial spent fuel rods under dry storage conditions (Kohli and Pasupathi 1986) indicated that most uncombined water in the fuel rod plenum region was released within 1 h during heating in the range of 200 to 400°C (392°F to 752°F) and the moisture was completely removed within 4 h. This moisture should be removable during vacuum drying of the SNF packages. Water that reacted with the UO_2 was not released at temperatures up to 400°C (752°F), in agreement with studies by Peehs and Fleisch (1986).

B.2.2 Degradation of SNF by Residual Gases

The potential quantity of reactive residual gases in a 7-m^3 (247 ft^3) dry storage system is estimated to be approximately 0.6 moles of O_2 and approximately 0.6 moles of H_2 (see Table 3 in Knoll and Gilbert 1987). If these were in the form of moisture to react with U- or UAl_x -alloy in accordance with Equations such as B12 and B23, then the maximum theoretical quantity of UH_3 that could be formed in a uranium alloy is approximately 0.3 moles or up to 70 g. Because of the dispersion of the U in the UAl_x , the UH_3 would be expected to be distributed over a large surface area and located in small isolated pockets. Other factors reducing the potential for UH_3 formation are the favorable reaction of UH_3 with water or oxygen to form UO_2 . Because a mass of at least 1 g may be required for ignition^(a), and individual masses approaching this size may not be credible, UH_3 ignition is not considered to be a concern in the UAl_x SNF types.

In contrast, the uranium in the metallic fuel types is not dispersed. Formation of UH_3 is energetically feasible per Equations B23 and B25. Although it is expected that the reactive gases would uniformly react with U at numerous cladding breach sites throughout the storage system, if it all were to react with metallic U as a single location, the approximately 70 g of UH_3 that could be formed is considerably more than the quantity that may be required for ignition during subsequent fuel handling, packaging, or drying operations.

Similar estimates and results can be made for the UAl_x metallic fuels using Equations B20, B16, and B12; however the total calculated quantities of UH_3 that could theoretically be generated by reaction with UAl_2 , UAl_3 , and UAl_4 are approximately 48, 32, and 24 g, respectively.

^(a)Personal communication with A. B. Johnson, Jr., Pacific Northwest Laboratory, July 1994.

Table B.3 Reaction Equation Results for Various Fuel and Environmental Constituents Using Outokumpu HSC Chemistry for Windows

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	103.230	94.123	1.001E-069
50.00	102.688	93.381	6.921E-064
75.00	102.143	92.681	6.527E-059
100.00	101.595	92.021	1.258E-054
125.00	101.041	91.398	6.703E-051
150.00	100.482	90.810	1.243E-047
175.00	99.909	90.255	9.588E-045
200.00	99.320	89.732	3.539E-042
225.00	98.708	89.241	6.990E-040
250.00	98.072	88.782	8.084E-038
275.00	97.406	88.354	5.890E-036
300.00	96.710	87.956	2.873E-034

H₂O Extrapolated from 500 K

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	33.615	27.797	4.195E-021
50.00	33.443	27.316	3.346E-019
75.00	33.268	26.848	1.395E-017
100.00	33.090	26.394	3.469E-016
125.00	32.906	25.951	5.675E-015
150.00	32.717	25.520	6.580E-014
175.00	32.519	25.101	5.730E-013
200.00	32.310	24.692	3.922E-012
225.00	32.087	24.296	2.188E-011
250.00	31.849	23.910	1.024E-010
275.00	31.595	23.537	4.120E-010
300.00	31.322	23.175	1.453E-009

H₂O Extrapolated from 500 K

[B3]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	42.815	34.888	2.655E-026
50.00	42.659	34.230	7.047E-024
75.00	42.505	33.584	8.246E-022
100.00	42.351	32.948	5.022E-020
125.00	42.195	32.324	1.802E-018
150.00	42.035	31.709	4.185E-017
175.00	41.868	31.103	6.769E-016
200.00	41.693	30.508	8.077E-015
225.00	41.505	29.921	7.442E-014
250.00	41.304	29.345	5.494E-013
275.00	41.088	28.778	3.349E-012
300.00	40.855	28.222	1.728E-011

H₂O Extrapolated from 500 K

[B4]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	331.372	314.281	4.046E-231
50.00	330.911	312.866	2.442E-212
75.00	330.435	311.488	2.808E-196
100.00	329.948	310.144	2.172E-182
125.00	329.451	308.834	2.904E-170
150.00	328.945	307.555	1.380E-159
175.00	328.426	306.306	4.083E-150
200.00	327.892	305.087	1.168E-141
225.00	327.341	303.896	4.601E-134
250.00	326.769	302.733	3.314E-127
275.00	326.177	301.599	5.512E-121
300.00	325.561	300.491	2.565E-115

H₂O Extrapolated from 500 K

[B5]

T, °C	delta H kcal/mol	delta G kcal/mol	K	E vs. SHE V
25.00	228.900	201.327	2.577E-148	2.9121
50.00	229.241	199.001	2.526E-135	2.8784
75.00	229.569	196.649	3.501E-124	2.8444
100.00	229.889	194.274	1.609E-114	2.8101
125.00	230.203	191.877	4.648E-106	2.7754
150.00	230.512	189.461	1.375E-098	2.7405
175.00	230.820	187.027	6.092E-092	2.7052
200.00	231.127	184.576	5.454E-086	2.6698
225.00	231.435	182.108	1.254E-080	2.6341
250.00	231.744	179.625	8.998E-076	2.5982
275.00	232.055	177.127	2.360E-071	2.5620
300.00	232.370	174.615	2.580E-067	2.5257

H⁽⁺⁾ Extrapolated from 398.15 K

H⁽⁺⁾ Extrapolated from 398.15 K

[B6]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	-135.761	-126.796	8.943E+092
50.00	-136.302	-126.022	1.726E+085
75.00	-136.831	-125.206	4.022E+078
100.00	-137.349	-124.354	6.895E+072
125.00	-137.860	-123.466	5.995E+067
150.00	-138.366	-122.547	1.989E+063
175.00	-138.873	-121.597	2.016E+059
200.00	-139.385	-120.620	5.240E+055
225.00	-139.907	-119.615	3.034E+052
250.00	-140.442	-118.583	3.492E+049
275.00	-140.993	-117.526	7.275E+046
300.00	-141.563	-116.443	2.540E+044

H₂O Extrapolated from 500 K

[B7]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	-38.633	-36.372	4.607E+026
50.00	-38.705	-36.179	2.954E+024
75.00	-38.782	-35.981	3.880E+022
100.00	-38.862	-35.777	9.036E+020
125.00	-38.945	-35.568	3.351E+019
150.00	-39.029	-35.353	1.823E+018
175.00	-39.114	-35.133	1.364E+017
200.00	-39.200	-34.909	1.336E+016
225.00	-39.286	-34.680	1.645E+015
250.00	-39.373	-34.447	2.464E+014
275.00	-39.461	-34.209	4.370E+013
300.00	-39.550	-33.968	8.983E+012

[B8]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	-34.933	-32.752	1.023E+024
50.00	-35.008	-32.566	1.063E+022
75.00	-35.088	-32.374	2.111E+020
100.00	-35.173	-32.176	7.029E+018
125.00	-35.261	-31.973	3.562E+017
150.00	-35.351	-31.763	2.551E+016
175.00	-35.444	-31.549	2.436E+015
200.00	-35.539	-31.329	2.966E+014
225.00	-35.636	-31.104	4.438E+013
250.00	-35.735	-30.874	7.924E+012
275.00	-35.836	-30.639	1.649E+012
300.00	-35.941	-30.400	3.917E+011

[B9]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	-31.233	-28.983	1.766E+021
50.00	-31.317	-28.791	2.974E+019
75.00	-31.404	-28.592	8.918E+017
100.00	-31.493	-28.387	4.242E+016
125.00	-31.584	-28.176	2.935E+015
150.00	-31.676	-27.960	2.766E+014
175.00	-31.769	-27.737	3.371E+013
200.00	-31.864	-27.510	5.105E+012
225.00	-31.959	-27.277	9.293E+011
250.00	-32.056	-27.040	1.982E+011
275.00	-32.154	-26.798	4.846E+010
300.00	-32.253	-26.551	1.334E+010

[B10]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	-7.633	-5.400	9.097E+003
50.00	-7.726	-5.209	3.338E+003
75.00	-7.815	-5.011	1.400E+003
100.00	-7.901	-4.807	6.540E+002
125.00	-7.983	-4.597	3.338E+002
150.00	-8.062	-4.382	1.834E+002
175.00	-8.136	-4.162	1.071E+002
200.00	-8.207	-3.939	6.598E+001
225.00	-8.273	-3.711	4.250E+001
250.00	-8.336	-3.481	2.846E+001
275.00	-8.394	-3.247	1.972E+001
300.00	-8.449	-3.011	1.407E+001

[B11]

T, °C	delta H kcal/mol	delta G kcal/mol	K	E vs. SHE V
25.00	0.600	-14.345	3.281E+010	-0.2075
50.00	0.967	-15.613	3.631E+010	-0.2258
75.00	1.342	-16.910	4.129E+010	-0.2446
100.00	1.725	-18.234	4.788E+010	-0.2637
125.00	2.113	-19.584	5.631E+010	-0.2833
150.00	2.507	-20.958	6.689E+010	-0.3031
175.00	2.906	-22.356	8.002E+010	-0.3234
200.00	3.311	-23.776	9.620E+010	-0.3439
225.00	3.722	-25.218	1.160E+011	-0.3648
250.00	4.139	-26.681	1.403E+011	-0.3859
275.00	4.562	-28.163	1.697E+011	-0.4074
300.00	4.992	-29.665	2.055E+011	-0.4291

H^(+a) Extrapolated from 398.15 K

H^(+a) Extrapolated from 398.15 K

[B12]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	-390.310	-402.470	1.104E+295
50.00	-391.734	-403.431	7.372E+272
75.00	-393.101	-404.284	6.442E+253
100.00	-394.430	-405.041	1.768E+237
125.00	-395.738	-405.709	5.216E+222
150.00	-397.039	-406.295	7.274E+209
175.00	-398.356	-406.804	2.528E+198
200.00	-399.709	-407.238	1.319E+188
225.00	-401.115	-407.599	6.887E+178
250.00	-402.587	-407.889	2.587E+170
275.00	-404.138	-408.106	5.336E+162
300.00	-405.778	-408.250	4.834E+155

H₂O Extrapolated from 500 K

[B13]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	0.600	13.591	1.087E-010
50.00	0.448	14.687	1.165E-010
75.00	0.301	15.794	1.215E-010
100.00	0.161	16.912	1.242E-010
125.00	0.026	18.038	1.252E-010
150.00	-0.103	19.173	1.249E-010
175.00	-0.227	20.316	1.235E-010
200.00	-0.345	21.465	1.214E-010
225.00	-0.457	22.620	1.189E-010
250.00	-0.563	23.781	1.160E-010
275.00	-0.664	24.947	1.129E-010
300.00	-0.757	26.117	1.097E-010

[B14]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	-342.417	-360.028	8.502E+263
50.00	-343.620	-361.455	2.994E+244
75.00	-344.771	-362.791	5.751E+227
100.00	-345.886	-364.046	1.720E+213
125.00	-346.983	-365.227	3.122E+200
150.00	-348.073	-366.339	1.673E+189
175.00	-349.178	-367.386	1.508E+179
200.00	-350.318	-368.371	1.465E+170
225.00	-351.506	-369.294	1.074E+162
250.00	-352.755	-370.156	4.450E+154
275.00	-354.077	-370.957	8.212E+147
300.00	-355.482	-371.696	5.550E+141

H₂O Extrapolated from 500 K

[B15]

T, °C	delta H kcal/mol	delta G kcal/mol	K	E vs. SHE V
25.00	-3.100	-17.965	1.478E+013	-0.2598
50.00	-2.730	-19.226	1.009E+013	-0.2781
75.00	-2.352	-20.517	7.590E+012	-0.2968
100.00	-1.965	-21.834	6.156E+012	-0.3158
125.00	-1.571	-23.179	5.297E+012	-0.3353
150.00	-1.171	-24.548	4.780E+012	-0.3551
175.00	-0.764	-25.940	4.482E+012	-0.3752
200.00	-0.350	-27.356	4.335E+012	-0.3957
225.00	0.072	-28.794	4.302E+012	-0.4165
250.00	0.501	-30.253	4.361E+012	-0.4376
275.00	0.938	-31.733	4.500E+012	-0.4590
300.00	1.383	-33.233	4.713E+012	-0.4807

H^(+a) Extrapolated from 398.15 K

H^(+a) Extrapolated from 398.15 K

[B16]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	-296.283	-302.075	2.786E+221
50.00	-297.385	-302.515	4.084E+204
75.00	-298.444	-302.872	1.387E+190
100.00	-299.472	-303.153	3.701E+177
125.00	-300.482	-303.367	3.435E+166
150.00	-301.483	-303.517	5.950E+156
175.00	-302.494	-303.608	1.184E+148
200.00	-303.529	-303.642	1.841E+140
225.00	-304.601	-303.621	1.646E+133
250.00	-305.719	-303.544	6.581E+126
275.00	-306.894	-303.413	9.593E+120
300.00	-308.132	-303.226	4.304E+115

H₂O Extrapolated from 500 K

[B17]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	-3.100	9.972	4.898E-008
50.00	-3.249	11.074	3.236E-008
75.00	-3.392	12.187	2.233E-008
100.00	-3.529	13.311	1.597E-008
125.00	-3.658	14.443	1.178E-008
150.00	-3.781	15.584	8.924E-009
175.00	-3.897	16.731	6.918E-009
200.00	-4.006	17.885	5.472E-009
225.00	-4.107	19.044	4.407E-009
250.00	-4.201	20.209	3.606E-009
275.00	-4.288	21.377	2.993E-009
300.00	-4.367	22.549	2.517E-009

[B18]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	-248.390	-259.782	2.759E+190
50.00	-249.266	-260.701	2.134E+176
75.00	-250.105	-261.553	1.595E+164
100.00	-250.919	-262.347	4.639E+153
125.00	-251.719	-263.086	2.652E+144
150.00	-252.514	-263.776	1.766E+136
175.00	-253.320	-264.418	9.111E+128
200.00	-254.151	-265.014	2.638E+122
225.00	-255.018	-265.566	3.307E+116
250.00	-255.928	-266.073	1.456E+111
275.00	-256.891	-266.535	1.895E+106
300.00	-257.914	-266.952	6.323E+101

H₂O Extrapolated from 500 K

[B19]

T, °C	delta H kcal/mol	delta G kcal/mol	K	E vs. SHE V
25.00	-6.800	-21.734	8.560E+015	-0.3144
50.00	-6.422	-23.001	3.608E+015	-0.3327
75.00	-6.036	-24.298	1.797E+015	-0.3515
100.00	-5.645	-25.623	1.020E+015	-0.3706
125.00	-5.248	-26.975	6.428E+014	-0.3902
150.00	-4.846	-28.351	4.408E+014	-0.4101
175.00	-4.439	-29.752	3.238E+014	-0.4303
200.00	-4.025	-31.175	2.519E+014	-0.4509
225.00	-3.605	-32.621	2.054E+014	-0.4718
250.00	-3.178	-34.087	1.744E+014	-0.4931
275.00	-2.745	-35.575	1.531E+014	-0.5146
300.00	-2.304	-37.082	1.383E+014	-0.5364

H⁽⁺⁾ Extrapolated from 398.15 K

H⁽⁺⁾ Extrapolated from 398.15 K

[B20]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	-202.255	-201.828	9.041E+147
50.00	-203.031	-201.760	2.911E+136
75.00	-203.778	-201.634	3.848E+126
100.00	-204.504	-201.454	9.984E+117
125.00	-205.217	-201.226	2.917E+110
150.00	-205.924	-200.954	6.281E+103
175.00	-206.636	-200.640	7.152E+097
200.00	-207.363	-200.286	3.314E+092
225.00	-208.113	-199.892	5.065E+087
250.00	-208.892	-199.461	2.153E+083
275.00	-209.708	-198.991	2.214E+079
300.00	-210.565	-198.483	4.903E+075

H₂O Extrapolated from 500 K

[B21]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	-6.800	6.203	2.837E-005
50.00	-6.940	7.299	1.157E-005
75.00	-7.077	8.406	5.284E-006
100.00	-7.209	9.522	2.646E-006
125.00	-7.335	10.647	1.430E-006
150.00	-7.456	11.780	8.229E-007
175.00	-7.571	12.920	4.999E-007
200.00	-7.681	14.066	3.180E-007
225.00	-7.784	15.218	2.104E-007
250.00	-7.880	16.374	1.442E-007
275.00	-7.971	17.536	1.018E-007
300.00	-8.054	18.701	7.388E-008

[B22]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	-295.233	-284.930	7.523E+208
50.00	-295.603	-284.051	1.326E+192
75.00	-295.978	-283.143	5.710E+177
100.00	-296.354	-282.208	1.995E+165
125.00	-296.731	-281.248	2.475E+154
150.00	-297.109	-280.264	5.804E+144
175.00	-297.488	-279.258	1.575E+136
200.00	-297.869	-278.231	3.361E+128
225.00	-298.253	-277.183	4.136E+121
250.00	-298.641	-276.116	2.286E+115
275.00	-299.034	-275.030	4.623E+109
300.00	-299.434	-273.927	2.888E+104

[B23]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	-76.535	-75.333	1.680E+055
50.00	-76.766	-75.223	7.559E+050
75.00	-76.992	-75.095	1.395E+047
100.00	-77.215	-74.951	7.975E+043
125.00	-77.437	-74.792	1.143E+041
150.00	-77.660	-74.619	3.490E+038
175.00	-77.889	-74.433	2.004E+036
200.00	-78.126	-74.234	1.958E+034
225.00	-78.374	-74.022	3.005E+032
250.00	-78.636	-73.797	6.789E+030
275.00	-78.914	-73.559	2.142E+029
300.00	-79.208	-73.309	9.033E+027

H₂O Extrapolated from 500 K

[B24]

T, °C	delta H kcal/mol	delta G kcal/mol	K
0.00	-122.317	-132.382	8.482E+105
25.00	-122.670	-133.287	5.128E+097
50.00	-123.001	-134.163	5.542E+090
75.00	-123.319	-135.015	5.783E+084
100.00	-123.629	-135.844	3.706E+079
125.00	-123.938	-136.652	1.038E+075
150.00	-124.250	-137.441	9.813E+070
175.00	-124.574	-138.211	2.553E+067
200.00	-124.915	-138.962	1.558E+064
225.00	-125.279	-139.695	1.962E+061
250.00	-125.672	-140.409	4.591E+058
275.00	-126.097	-141.104	1.834E+056
300.00	-126.558	-141.778	1.165E+054

[B25]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	-30.400	-17.380	5.507E+012
50.00	-30.531	-16.283	1.031E+011
75.00	-30.665	-15.176	3.367E+009
100.00	-30.800	-14.059	1.716E+008
125.00	-30.936	-12.932	1.257E+007
150.00	-31.071	-11.798	1.241E+006
175.00	-31.205	-10.655	1.573E+005
200.00	-31.338	-9.505	2.460E+004
225.00	-31.469	-8.348	4.602E+003
250.00	-31.600	-7.185	1.004E+003
275.00	-31.730	-6.015	2.503E+002
300.00	-31.859	-4.839	7.005E+001

[B26]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	-92.270	-115.907	9.313E+084
50.00	-92.470	-117.881	5.376E+079
75.00	-92.654	-119.839	1.718E+075
100.00	-92.829	-121.785	2.159E+071
125.00	-93.002	-123.720	8.260E+067
150.00	-93.180	-125.643	7.905E+064
175.00	-93.369	-127.556	1.623E+062
200.00	-93.577	-129.457	6.334E+059
225.00	-93.810	-131.347	4.263E+057
250.00	-94.072	-133.224	4.572E+055
275.00	-94.367	-135.089	7.327E+053
300.00	-94.699	-136.939	1.663E+052

H₂O Extrapolated from 500 K

[B27]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	-228.900	-229.263	1.171E+168
50.00	-228.722	-229.301	1.235E+155
75.00	-228.529	-229.353	9.712E+143
100.00	-228.326	-229.419	2.395E+134
125.00	-228.116	-229.499	9.674E+125
150.00	-227.903	-229.593	3.895E+118
175.00	-227.687	-229.699	1.063E+112
200.00	-227.471	-229.817	1.452E+106
225.00	-227.256	-229.946	7.782E+100
250.00	-227.042	-230.087	1.344E+096
275.00	-226.830	-230.237	6.371E+091
300.00	-226.620	-230.397	7.259E+087

[B28]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	10.285	6.689	1.250E-005
50.00	10.437	6.381	4.832E-005
75.00	10.614	6.061	1.567E-004
100.00	10.812	5.727	4.421E-004
125.00	11.029	5.379	1.114E-003
150.00	11.263	5.017	2.561E-003
175.00	11.515	4.641	5.451E-003
200.00	11.785	4.250	1.088E-002
225.00	12.075	3.845	2.056E-002
250.00	12.386	3.424	3.710E-002
275.00	12.718	2.988	6.434E-002
300.00	13.072	2.537	1.078E-001

$\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$ Extrapolated from 500 K

H_2O Extrapolated from 500 K

[B29]

T, °C	delta H kcal/mol	delta G kcal/mol	K
25.00	12.855	4.260	7.540E-004
50.00	13.553	3.510	4.225E-003
75.00	14.226	2.708	1.996E-002
100.00	14.872	1.858	8.164E-002
125.00	15.491	0.965	2.952E-001
150.00	16.084	0.035	9.594E-001
175.00	16.658	-0.930	2.842E+000
200.00	17.221	-1.927	7.763E+000
225.00	17.777	-2.953	1.975E+001
250.00	18.331	-4.007	4.720E+001
275.00	18.888	-5.087	1.068E+002
300.00	19.451	-6.193	2.300E+002

$\text{Al}_2\text{O}_3 \cdot 3\text{H}_2\text{O}$ Extrapolated from 500 K

H_2O Extrapolated from 500 K

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APPENDIX C

SUMMARY OF ADDITIONAL INFORMATION FOUND ON SNF WITH SS CLADDING

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Summaries of additional information found on the SNF with SS cladding are provided below. The information is provided for the APPR, BMI, BORAX V, EBR-II, EBR-II (ANL-6 TEST), FERMI BLANKET, OMRE, SM-1A, SPERT, TRIGA BER II, TRIGA FLIP, and VBWR spent fuels.

APPR (Age 2)

Age 2 refers to the shipper's number of this particular fuel. Age probably refers to Atomic General Electric; 2 refers to their second shipment to the ICPP. For the fuel to have been received at INEL in 1960, the fuel had to belong to core 1 at SM-1. From this core, fuel elements suffered severe localized attack at the brazed joints, intergranular and intragranular cracking of the SS cladding, and fairly extensive rippling of the outer fuel plates; although the cladding was severely cracked, it released only nominal quantities of fission products to the primary coolant and withstood ^{235}U depletion up to 57% without matrix cracking (Gallagher 1964).

One SS-clad element is contained in a CS can in the CPP-603. In a moist or wet environment caused by water intrusion into the can, the two alloys will form a galvanic couple which will cause corrosion of the CS to accelerate.

BMI

The fuel history is unknown except that it was received at INEL between 1961 to 1962. Thirteen elements of SS-clad UO_2 fuel are currently canned in three cans of SS or Al in the CPP-603 according to the INEL Fuel Inventory.

BORAX V

"Leak-testing of the BORAX-V ... has been completed as you requested ... the entire BORAX-V inventory ... has been tested to determine if these fuels are suitable for continued storage in the basin or if they should be canned ... BORAX-V fuels have been determined to be comparatively low contributors, amounting to

only 0.1% of the total leakage for the entire inventory ... the maximum leakage rate for any single BORAX-V element was 0.016 millicuries/day total activity, and a total of 0.122 millicuries/day was measured for the entire inventory of 32 intact elements ... four disassembled sections of a BORAX-V element(s) were not tested due to difficulty in handling."^a The 36 elements of fuel are not canned but contained in 13 stainless steel material test reactor buckets in the CPP-603.

EBR-II

The EBR-II fuel is thought to have been canned in the HFEF hot cell at Argonne. The normal cover gas in this cell is argon or nitrogen, and the fuel is thought to have been canned in argon. The cans that were used contain swagelock closures of high integrity, so water in-leakage was not expected during the pool storage.^b The 3636 elements of canned fuel are stored in both the CPP-603 and CPP-666 basins.

EBR-II (ANL-6 TEST)

The fuel and cladding is in the form of chunks, chips, and powder. According to Ronald Damn of ANL East, the material contains no sodium but may be pyrophoric (how this was determined was not addressed). The fuel was stored prior to 1977 in an 800 series cask at the TRA Hot Cell with the fuel material doubly encapsulated in watertight capsules. The inner capsule consisted of a galvanized pipe nipple [5.08-cm (0.82-in.) diameter, (6-in.) 15.24-cm long], sealed at each end with galvanized pipe caps. The outer capsule consisted of SS pipe [7.62-cm (3-in.) diameter, 55.88-cm (22-in.) long, 0.64-cm (0.25-in.) thickness) with a welded bottom closure and a pipe cap top enclosure. The pipe cap threads were sealed with Teflon tape. The capsules were felt to be watertight upon assembly, but leak testing of the outer capsule was recommended in 1977 prior to transferring the capsules to the CPP-603 basin.

^a Letter from E. P. Mondok (Exxon Nuclear Idaho) to M. J. Painter dated July 23, 1982; "Results of SM-1A and BORAX-V Fuels Leak Tests."

^b Private communication with Al Christensen, LITCO, July 1994.

FERMI BLANKET

The Fermi 1 subassembly was carried out of the reactor in a sodium-filled pot destined for spent fuel storage. The subassembly was then washed in a cleaning machine that reacts the sodium on the subassembly with a stoichiometric amount of dry steam. The subassembly was washed with hot demineralized water, then lowered into the water-filled cut-up pool. The subassemblies were stored for 180 days in the pool for decay, then the blanket was typically cut from the subassembly. For shipping, the blanket assemblies were loaded into a cylindrical steel basket container, and the container was then capped. The shipping container and cask were removed from the pool, and internally drained of any pool water. There was no cover gas for the cask mentioned in the cask shipping plan for the assemblies. It may then be implied that the blanket assemblies were exposed to air during the shipment, and subsequently when the assemblies were stored in CPP-749 (Dry Wells) after receipt of the blankets by INEL. At this time, there are 510 blanket assemblies stored in 14 Type 316 SS baskets in the CPP-749.

The fuel performance was critiqued in an Energy Research and Development Administration report. They stated that plant operation after 1970 was limited by irradiation-induced creep and SS swelling. These problems were more significant in the Fermi 1 reactor than in any other operating fast reactor due to the 50% greater flux level.

OMRE

"Failures occurred in three of the fuel elements removed from the first core loading of the OMRE ... (one) was caused by improper seating of the element in the core ... temperatures had become sufficiently high on the other side to carbonize the coolant and deform the fuel box, preventing it from passing through the grid plate (Trilling 1959).

One element of fuel is currently stored in an Al can in the CPP-603 basin. It is unknown whether the fuel was canned dry or in water. With the ingress of moisture or water into the can,

the Al will form a galvanic couple with the SS fuel cladding which will increase the corrosion rate of the Al.

SM-1A

In 1983, all the SM-1A fuel was canned into SS cans fabricated of 12.7-cm (5-in) tubing 12.4-cm (4.88-in ID) with overall length of 100 cm (39.38 in.). The fuel was canned wet.^c At that time, one-third of SM-1A fuel elements at CPP-603 had been tested to determine if the fuels were suitable for continued storage in the basin or if they should be canned. Data revealed that SM-1A fuel elements were significant contributors to the leakage of total fission products into the basin. Calculated leak rates extrapolated for all SM-1A elements amounted to 50% of total leakage into the basin water. Every one of the SM-1A elements tested at a leakage greater than 0.08 mCi/day with the highest about 3.4 mCi/day. Most SM-1A elements at the time of receipt were known leakers. Comparison of leak rates measured recently and loading plan information provided earlier revealed that the integrity of almost all of the elements has degraded over the 11-year storage. It should be noted that even the highest leakage level measured during this testing, 3.4 mCi/day, would have been well below background levels and could not have been measured in leak rate tests performed when the SM-1A casks were received (because of lower basin water activity levels in 1982).^d

Of the 32 elements tested (61 elements not tested):

1982

13 heavy leakers
19 moderate leakers

Prior to 1971

5 heavy leakers
10 moderate leakers
17 non leakers

SPERT

Limited damage to the SS-clad fuel was observed for three

^c Letter from S. R. Boulton (INEL) to G. T. Paulson dated October 1, 1990, "CSE Request for Rack Storage of Borax-V and SM-1A."

^d Letter from E. P. Mondok (Exxon Nuclear Idaho Co.) to M. J. Painter, dated July 23, 1982, "Results of SM-1A and BORAX-V Fuels Leak Tests."

SPERT nuclear reactor cores. The cores were used in SPERT I and SPERT III reactors. The damage was exhibited as blistering and rippling was observed macroscopically on the cladding surface. This damage occurred during power excursion testing rather than steady-state operation. Post-damage investigations noted that the temperatures experienced by the cladding were well above the sensitizing range for nonstabilized austenitic SS and the plates were most certainly susceptible to IGSCC. The blistering was thought to be a result of the nonuniformity of the distribution of the UO_2 in the fuel matrix, and the rippling from the gross deformation due to high stresses induced due to localized thermal gradients (Heffner 1961). Most of the fuel was accepted for chemical processing, so only one element remains. This element has not been positively identified as to type, but it is suspected that due to the SS cladding, it is a Type "C" or Type "P" element.

The element of fuel currently stored is in an Al can in the CPP-603 basin. It is not known if the element was canned wet or dry. There is a potential for a galvanic couple between the cladding of the fuel and the Al can if there is moisture inside the can, leading to an increased corrosion rate for the Al. Containment is listed as a bucket in the CPP-603 basin.

TRIGA BER-II

There are currently 21 elements stored uncanned in the CPP-603 basin, according to the INEL Fuel Inventory. The containment device is listed as a canister (no alloy designation given). The shipment documentation from the source of the fuel, TRANSNUCLEAR GMBH, stated that the elements were intact at the time of shipment so they did not can the elements.

TRIGA FLIP

The seven elements of TRIGA FLIP fuel are presently canned in two Al cans placed in SS racks in the CPP-603 basin. It is unknown whether the fuel was canned dry or in water. With the ingress of moisture or water into the can, the Al will form a galvanic couple with the SS fuel cladding, which will increase

the corrosion rate of the Al. The Al cans in the SS racks will also form a galvanic couple, accelerating the corrosion rate of the Al can.

VBWR

Until May 1962, thin-wall SS clad fuel rods with wall thicknesses ranging from 0.013 cm (0.005 in.) to 0.030 cm (0.012 in.) had operated successfully in VBWR tests. Some rods had attained peak burnups of approximately 6000 MWD/MT with no apparent performance problems. Eventually, cladding failures occurred in essentially all types of SS-clad fuel rods under irradiation in the VBWR. These included thin-wall and freestanding cladding, pellet, and powder fuel, and cold-worked and annealed cladding (Pashos 1965).

There are currently four elements of VBWR fuel canned in four Al cans in the CPP-603 basin. It is unknown whether the fuel was canned dry or in water. With the ingress of moisture or water into the can, the Al will form a galvanic couple with the SS fuel cladding which will increase the corrosion rate of the Al.

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