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# **Materials Aging Issues and Aging Management for Extended Storage and Transportation of Spent Nuclear Fuel**

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*Protecting People and the Environment*

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# **Materials Aging Issues and Aging Management for Extended Storage and Transportation of Spent Nuclear Fuel**

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## ABSTRACT

An evaluation of potential degradation of materials in the Systems, Structures, and Components (SSCs) of dry cask storage systems (DCSS) for Independent Spent Fuel Storage Installations (ISFSIs) has been performed to identify issues in the technical bases for regulating storage well beyond the present allowable storage time of 120 years that includes 60 years pool storage and 60 years dry storage. The evaluation included low ( $\leq 45$  GWd/MTU) and high burnup (HBU) ( $> 45$  GWd/MTU up to approximately 62.5 GWd/MTU) fuel, and was performed to determine if: 1) degraded conditions could be anticipated to develop in the materials of the SSCs over an extended storage and transportation (EST) period ( $> 120$  years); and 2) the anticipated degradation would challenge functions important to safety during storage and during post-storage transportation including fuel retrieval for final disposition.

The approach followed a general Aging Management Review process by: 1) identifying the materials of construction of the SSCs; 2) evaluating their susceptibility to degradation from environmental conditions of exposure; and 3) evaluating the potential for degradation to impact the safety functions required by the present Code of Federal Regulations for dry storage systems and spent fuel transportation in 10 CFR Part 72 and 10 CFR Part 71, respectively. The evaluation was performed using extrapolation of available data and materials degradation models with consideration of service histories of similar materials systems, and with expert judgment.

With the present materials systems and controls for a DCSS, no materials degradation phenomena were identified that would be expected, with certainty, to be operative and cause an impact sufficient to violate the safety functions for storage and transportation. However, several data/information/technology gaps were identified that would require resolution to provide a sufficient technical basis to demonstrate the continuing integrity of the SSCs and their ability to maintain functions important to safety throughout EST.

Recommendations for activities to resolve the potential materials aging issues are provided. Aging management activities for preventive maintenance and periodic condition assessments through inspections of SSCs are outlined. Additional recommended activities include materials testing and a full cask demonstration project under conditions that challenge the SSCs of the DCSS to verify the expected materials' behavior and provide confidence that the SNF, as is presently conditioned and dry-stored, can continue to be safely stored for greater than 120 years, and handled and transported following storage.



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## EXECUTIVE SUMMARY

Dry storage of spent nuclear fuel (SNF) has been implemented as an alternative to pool storage of fuel that has been removed from a reactor and is awaiting a long-term disposition decision. At this time, there are thousands of spent fuel elements safely stored at Independent Spent Fuel Storage Installations (ISFSI) around the world. Much is known about dry storage – the technical bases to demonstrate safe storage are sound, and the technologies for implementation of dry storage systems are mature. The Nuclear Regulatory Commission (NRC) has responsibility for regulating the dry storage of power reactor SNF in the United States (U.S.).

This document contains an evaluation of age-related degradation of the Systems, Structures, and Components (SSCs) of dry cask storage systems (DCSS) for ISFSIs and identifies gaps in the technical bases needed to demonstrate safe extended dry storage and transportation (EST) of low burnup ( $\leq 45$  GWd/MTU) and high burnup (HBU) ( $>45$  GWd/MTU up to approximately 62.5 GWd/MTU) spent nuclear fuel for its storage beyond the present allowable storage period of 120 years following reactor discharge, up to 300 years, followed by its transportation. The evaluation considers the variety of fuel designs and discharge conditions of SNF including MOX fuels.

There is a large body of information describing degradation mechanisms which can occur during dry storage of low burnup SNF. This information addresses fuels in dry storage for up to about 60 years and provides a firm technical basis for the licensability of dry storage facilities. Several critical investigations have considered the dry storage of this fuel in present storage facilities to 100 years. There is a trend to higher fuel burnup, new fuel compositions and new cladding alloys, and the possibility that the fuel will remain in dry storage for an even longer periods pending final disposition decisions. This report serves to identify information needed to support this trend and extend the understanding of materials degradation to include the expected behavior of SNF and the materials of the DCSS for EST.

An evaluation of the potential age-related degradation of the fuel and materials in the SSCs of DCSS for ISFSIs during EST was made. This evaluation of the information needed to develop the technical basis to demonstrate that safety functions are met during EST, showed four major data/information/technology gaps:

1. A wide variety of alloys are, and have been, used for fuel cladding in the U.S. commercial fleet, and the response of these alloys to EST depends on chemistry, thermal mechanical process history, irradiation history and other fabrication/service variables. The information required to address the response of a specific cladding to a specific age-related degradation process is limited and in many cases simply not available.
2. The conditions required for corrosion-induced degradation processes such as stress corrosion cracking are, and will continue to be, present on the external surfaces of dry storage canisters, especially those fabricated from austenitic stainless steels. The information necessary to assess the time-temperature induced changes in cracking susceptibility is lacking and the technology necessary to inspect (remotely) for such cracking in the canisters has not been developed.

3. Delayed hydride cracking in the Zircaloy-clad fuel may occur and hydrogen embrittlement is anticipated as the fuel cladding temperatures fall during EST. The occurrence of these two phenomena would create cracks in a fuel cladding that has minimal fracture toughness. Limited data is available; however a validated fracture assessment technology is not available to establish the impact of these two degradation processes on assembly retrievability, handling, or transport.
4. Bolted closures with metallic seal materials will experience sealing force reduction and may lose sealing capabilities during EST. The information needed to establish replacement schedules is lacking.

In addition to the four major gaps described above there are several other significant gaps in the available information.

- A) The heat output, attendant radiation, and reactivity of spent MOX fuels significantly exceed those of UO<sub>2</sub> fuels; cask thermal and radiation design must address these conditions to avoid excessive temperatures for long times that would impact fuel and canister degradation.
- B) There is a potential for delayed hydride cracking of fuel cladding and stress corrosion cracking of the canister occurring in the same dry storage cask. The impact of this occurrence on dry storage confinement, retrievability, handling, and post-storage transport has not been well developed.
- C) The concrete pads, and also casks, will deteriorate and a path forward to address the impact of such deterioration has not been defined even though there are some ISFSI's that are built on the only practical site at the reactor.
- D) The lifetime of the ISFSIs, in some cases, will exceed the lifetimes of the corporation using the facility. The information and regulations required for successful transfer of data from a dying or defunct corporation to the new tenant is minimal at best.
- E) The aluminum based baskets and neutron absorbers could degrade during EST because of creep and/or corrosion processes. An evaluation is needed for canister internals with materials subject to creep deformation at anticipated storage temperatures (e.g., aluminum). Data are needed to demonstrate that such material degradation does not compromise the retrievability of the fuel assemblies or change the configuration of the absorber and limit its effectiveness to maintain subcriticality during re-submersion, if required.
- F) The technical basis for any assumptions in both the number of failed fuel rods, and radiological source terms from high burnup (HBU) fuel for normal and accident conditions needs to be developed for EST.

The evaluation that led to the points listed above included HBU (62.5 GWd/MTU) fuel and was performed to determine if degraded conditions could be anticipated to develop over a very long term (up to a nominal 300 years) storage period, and if the anticipated conditions could challenge functions important to safety during storage and/or transportation. It was clear that anticipated materials degradation processes could



cause failures of fuel and SSCs during the extended storage period and/or during post-storage transportation to a facility for final disposition. *However, no materials degradation phenomena were identified that would, with certainty, occur to cause an impact sufficient to violate the safety functions for storage and transportation.*

The approach to the degradation evaluation considered the types of materials and generic designs for the DCSS, and followed, in general, the Aging Management Review process listed in the Standard Review Plan for licensing and license renewal of an Independent Spent Fuel Storage Installation for 40 additional years of storage (NUREG-1927 at <http://www.nrc.gov/reading-rm/doc-collections/nuregs/staff/sr1927/>), with the exception that storage periods up to a nominal period of 300 years followed by handling and transport, was considered.

Additional environmental conditions (e.g., an increase in atmospheric carbon dioxide) due to man-made or natural climatic changes over EST were considered. The impact of off-normal/accident design-basis events such as floods, tornados, projectiles, temperature extremes, cask drop, cask tipover, and other unusual scenarios during EST on the materials of the DCSS were not considered in this report with the exception of uncontrolled air/water ingress into the canister due to failure of the canister.

The materials aging evaluation included a review of published material degradation processes and focused on well-referenced and highly-cited documents. The approach included extrapolation of available constitutive models and expert judgment applied to the materials used in the DCSSs with no specific time limit to the aging phenomenon, except for the general target of 300 years.

There is a wide variety of fuel cladding alloys at various reactor service conditions, and several variations of DCSS design including materials of construction for the SSCs. A reference material and condition was constructed in some cases because of the multitude of materials options. The materials and fabrication of DCSS SSCs were assumed in the as-design condition; and in the case of fuel, the post-discharge condition of the fuel and the effects of drying were considered.

The table ES-1 summarizes results of the assessment of specific degradation processes (types of degradation) and their expected impact to safety functions of the SSCs of the DCSS. An important consideration is the degradation of the fuel cladding. While in the DCSS, a fuel cladding failure causing a release of radioisotopes to the confinement would increase the source term for confinement provided by the canister, but would not compromise confinement. It would impede the retrievability of the fuel by assembly, not canister. Additionally, cladding failure of the higher burnup fuel would be expected to have increased potential source term due to fission gases, volatiles, and fines produced from the rim that formed on the fuel pellets.

Aging management activities were recommended to address information gaps and ensure that the performance of the DCSS meets its safety basis during EST. A periodic monitoring/in-service inspection program is the principal strategy recommended to ensure safety function of the DCSS throughout its desired service. This program would include periodic inspection of the materials and the condition of materials and components of the SSCs of the DCSS. Acceptance criteria for disposition of indications attributed to aging-induced processes and other flaws must be established. Provisions

for repair or replacement (recovery) would need to be available for SSCs that would fall outside the acceptance criteria.

The aging management activities also include preventive maintenance activities such as periodic replacement of gaskets and closure bolts. The gasket materials are expected to relax and lose sealing capacity during long term storage and the bolts may experience stress corrosion cracking during extended service.

General recommendations to obtain additional data/information to support resolution of the information gaps are outlined. The recommendations include those for laboratory tests and for a fuel storage demonstration project that would include storage and testing under challenging (e.g., HBU, partially-degraded fuel) conditions. It is recognized that programs to address information needs, including international programs, are in various stages of completion. The conclusions in this report would be subject to change as additional data and service experience with fuel in dry storage are obtained.

**Table ES-1** Summary of the evaluation of fuel and SSCs of an ISFSI/DCSS for their susceptibility to materials aging degradation under extended storage, and the potential impact of such degradation on the safety functions for EST. All credible degradation mechanisms for EST were considered. Generalized activities are proposed to resolve the issues. The set of degradation mechanisms considered for the fuel are itemized in the table. The primary degradation mechanisms anticipated for each DCSS component are listed in the table.

Fuel (Zircaloy cladding failure mechanisms)	Degradation Under EST	Impact on Safety	Resolution (Recommended)	Report Section
<b>Creep</b>	Creep leading to clad failure is not expected due to low temperature in EST, and the self-relief of the driving force (internal pressure) with creep deformation. If clad failure did occur, it would be a minor breach, not a large rupture.	Clad failure could cause release of gases, volatiles, and some fines to the canister and impact the source term and fuel retrievability from dry storage.	Testing to verify existing creep model(s) applicability to the behavior of HBU fuel cladding, and to the new alloys, as well as to determine the universal applicability of creep failure at a strain of ~2%. Also, evaluate phenomenon as part of cask demonstration project.	Section 2.2.1
<b>Delayed Hydride Cracking (DHC)</b>	DHC, including the development of a through-wall (minor) crack, is possible, if not probable. However, if clad failure did occur, it would produce a minor breach, not a large rupture. Hydrogen diffusion and hydride reorientation are expected to be involved in the DHC processes.	Clad failure could cause release of gases, volatiles, and some fines to the canister. This would impact the source term and retrievability from dry storage. DHC could also create seed flaws that are subject to instability when the storage and/or transport temperatures are below DBTT. Growth of such flaws could cause gross failure during handling and transportation and further impact the retrievability and confinement.	Evaluate phenomenon during the fuel destructive evaluations as part of cask demonstration project. Evaluate impact of drying treatment on hydride microstructure to establish initial fuel condition and assess the potential for DHC and HE during EST.	Section 2.2.3 Section 2.1.6
<b>Hydrogen Embrittlement (HE)</b>	HE will reduce both the tensile ductility and the fracture toughness and shift in DBTT to higher temperatures. Higher burnup fuel has the potential for lower toughness and higher DBTT shifts than low burnup fuel due to additional hydrogen accumulation and hydride precipitation. Hydride orientation with respect to flaw orientation affects the fracture toughness of cladding – flaws with radial hydrides causing low toughness for axially-oriented flaws.	Hydrogen embrittlement, in conjunction with cladding flaws could result in gross failure under high mechanical loads (off normal/ accident conditions and/or post storage handling) that may impact retrievability and cause a large cladding rupture. The cladding rupture would increase the source terms for transportation. Further, HBU fuel would have potentially higher source terms including fines due to rim effects.	Develop models with benchmarked data for tensile and fracture properties. Develop consensus approach to fracture control plan(s) for fuel handling and transportation. Revisit a consensus standard on drying to provide explicit consideration of hydride microstructure reconfiguration during drying and, if practical, modify the standard to reduce potential for hydride reorientation that promotes DHC and HE degradation.	Section 2.2.4 Section 2.2.5 Section 2.1.6

<b>Fuel, CONT'D</b> (Zircaloy cladding failure mechanisms)	<b>Degradation Under EST</b>	<b>Impact on Safety</b>	<b>Resolution (Recommended)</b>	<b>Report Section</b>
<b>Stress Corrosion Cracking</b>	SCC of cladding is not expected because most modern cladding alloys are lined with zirconium to protect against such degradation. If clad failure did occur, it would be a minor breach, not a large rupture.	Clad failure could cause release of gases, volatiles, and some fines to the containment of canister and impact post storage retrievability.	Evaluate, by destructive examination, the occurrence of stress corrosion cracking in fuel cladding after prolonged storage. Such evaluations should be part of cask demonstration program and not conducted simply to assess SCC.	Section 2.2 Section 2.2.7
<b>Helium Pressurization</b>	Additional cladding internal pressurization by helium is not anticipated because the temperature will be decreasing and the increased helium concentration due to alpha decay is insignificant.	No degradation anticipated.	None needed.	Section 2.2.8
<b>Diffusion Controlled Cavity Growth (DCCG)</b>	DCCG is a micromechanism of creep and is not anticipated.	Same as for creep.	Same as for creep.	Section 2.2.9
<b>Off-Normal Air Oxidation of Fuel Cladding</b>	Oxidation of cladding with air in-leakage that could result in metal wastage leading to a large rupture of fuel with time is not anticipated due to the relatively low cladding temperature.	No degradation anticipated.	None needed for breach-free fuel.	Section 2.2.10
<b>Off-Normal Air Oxidation of Fuel Pellet</b>	Oxidation of fuel pellets with air in-leakage because of existing or developed though clad defects could result in pellet swelling and a large rupture of the fuel cladding with time. Air in-leakage could also affect cover gas conductivity, cladding oxidation and emissivity and alter heat transfer conditions. The potential impact of cladding oxidation is minimal compared to fuel pellet oxidation.	Gross failure during storage could cause release of gases, volatiles, and fines to the canister and impact the source term and retrievability. This would be especially significant to HBU fuel because of their higher source terms including fines.	Establish controls or monitoring procedures to ensure inert atmosphere is maintained.	Section 2.2.11

Fuel Canister Internals	Degradation Under EST	Impact on Safety	Resolution (Recommended)	Report Section
<b>Fuel Basket</b>	Loss of geometry of aluminium basket with temperature and time due to creep may occur.	Loss of ready retrievability of fuel.	Evaluate creep of aluminium basket materials for extended storage thermal histories and if excessive, develop fuel recovery strategy.	Section 2.4
<b>Neutron Absorbers</b>	Design of some absorbers (the Boral absorber, B <sub>4</sub> C-Al in SS clad) could have led to inadvertent water ingress and with incomplete drying, and eventual corrosion (of aluminium) and H <sub>2</sub> blistering with absorber crumbling. This is not expected in the newer design absorbers but may be present in the old design absorbers.	Loss of necessary criticality control for wet immersion only (not dry-to-dry transfer).	To assure criticality control during fuel retrieval, even if neutron absorbers have degraded during storage develop plans to: 1) mitigate degradation by modifying drying standards that assure water removal, 2) developing dry-to-dry transfer procedures for fuel retrieval and/or 3) if wet transfer is necessary for shielding, replace the degraded neutron absorbers in cask during fuel retrieval by re-flooding with fluid such as borated water.	Section 2.4 Section 2.1.6

<b>Fuel Canister</b>	<b>Degradation Under EST</b>	<b>Impact on Safety</b>	<b>Resolution (Recommended)</b>	<b>Report Section</b>
<b>Canister Body</b> (stainless steel)	The canister would be expected to experience pitting and cracking that could cause through-wall failure of canister under aggressive conditions. Increased condensation of moisture with time due to cooling exacerbates corrosion condition especially in DCSS located near the sea coast or highly industrial environments that are a source of aggressive species.	Canister/cask is credited as confinement boundary of the DCSS. Only a limited leak path is expected with a pitting/cracking breach of the canister. Release of gases, volatiles, and some fines could occur during storage. Cracks could open under high (e.g., from accident) loadings.	Establish in-service inspection protocol with acceptance criteria for pitting and cracking.	Section 2.6.2
<b>Closure Weldment</b>	The weldment is susceptible to stress corrosion cracking that could cause through-wall failure. Aggressive conditions are the same as for the canister body.	Canister is credited as confinement boundary. Only a limited leak path is expected if a cracking breach should develop.	Establish in-service inspection protocol with acceptance criteria for cracking.	Section 2.6.2
<b>Metallic Seals</b>	Loss of sealing force and seal resiliency is expected and that loss may cause a loss of sealing. Seals are also subject to corrosion including crevice corrosion.	Seal is necessary for confinement boundary of cask in storage.	Perform testing to evaluate sealing capability with loss of sealing force with time. Replacement of metallic seals under EST is expected and therefore facility (pool or dry system) may be needed.	Section 2.8.1
<b>Bolts</b>	Corrosion and stress corrosion cracking leading to breakage may occur.	Multiple adjacent bolt failures would cause loss of sealing force leading to loss of confinement of cask in storage.	Perform periodic bolt replacement as a maintenance item.	Section 2.8.3

<b>Cask and Pad</b>	<b>Degradation Under EST</b>	<b>Impact on Safety</b>	<b>Resolution (Recommended)</b>	<b>Report Section</b>
<b>Steel Cask</b> (carbon steel)	General and pitting corrosion if bare steel. No significant degradation expected contingent on periodic maintenance of coating.	Steel overpack provides shielding and structural stability functions and pitting not typically significant to these functions.	Establish plan for periodic maintenance of steel (e.g., re-coat) as necessary.	Section 2.10.1
<b>Polymeric Shield Material</b>	Medium potential for oxidation-induced mechanical property changes leading to slump within the overpack.	Polymeric materials provide n-shielding function. Shielding function of hydrogenous media would not diminish with structural change unless material re-configures in dimensions and slumps.	Establish plan for periodic monitoring of shielding to check for radiation leakage to augment present monitoring for DCSS.	Section 2.10.2
<b>Concrete Overpack</b>	Corrosion of steel in reinforced concrete leading to concrete spallation and loss of section is expected. Minimal impact to metal-encased unreinforced concrete.	Concrete materials provide gamma and n-shielding function and cooling geometry for maintaining thermal safety function.	Establish in-service inspection protocol with acceptance criteria.	Section 2.12
<b>Concrete Pad</b>	Corrosion of steel in reinforced concrete leading to concrete spallation and loss of structural function for cask and retrieval equipment foundation is expected.	Loss of structural stability of the pad under normal conditions would lead to cask tip-over potential with direct impact to retrievability and indirect impact (unquantified) to other safety functions of DCSS including shielding and thermal functions.	Establish in-service inspection protocol with acceptance criteria.	Section 2.12





## **ACKNOWLEDGMENTS**

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This report is an independent product of the Savannah River National Laboratory and does not necessarily reflect the views or regulatory position of the U.S. NRC.



## FOREWORD

This report is intended for use by the U.S. Nuclear Regulatory Commission (NRC) staff in its consideration of technical information needed to enable regulation of extended storage (up to 300 years) of spent fuel in a dry storage system at a fuel storage site and the subsequent safe transportation of the fuel. Specifically, the report provides information to support a qualitative prediction of material degradation with time for those Systems, Structures, and Components (SSCs) of a Dry Cask Storage System (DCSS) that are important to safety as required in part 72.24 of Title 10 of the Code of Federal Regulations, Part 72, "Licensing Requirements for the Independent Storage of Spent Nuclear Fuel, High-Level Radioactive Waste, and Reactor-Related Greater than Class C Waste." It further provides information to support a qualitative prediction of the fuel condition for post-storage retrieval and transportation.

Additionally, because of the necessity to extrapolate engineering data for far beyond the time frames used for data gathering, some prediction of materials behavior must be based on engineering judgment. For example, several age-related degradation processes (e.g., stress corrosion cracking and delayed hydride cracking) are nucleation and growth processes and the data to demonstrate the lack of nucleation over a 100+ year time frame does not exist. This report provides the evidence and background necessary for realistic, well-informed engineering judgments of SSC materials' aging but does not attempt unrealistic extrapolations of short-term data.

This report also identifies the gaps in the technical bases for prediction of materials performance for extended storage, assesses the impact of the degradation to the safety functions, and provides general recommendations for activities to close the gaps and enable safe extended storage and post-storage transportation. The intent of the report is not to prescribe a detailed approach for issue resolution.

Furthermore, although the report adopts the general approach of the present Standard Review Plan (NUREG-1927 at <http://www.nrc.gov/reading-rm/doc-collections/nuregs/staff/sr1927/>) to consider materials aging in a dry storage system that had an initial license period of 20 years with a 40-year extension, specific guidance for a licensing review is outside the scope of this report.



## ACRONYMS AND ABBREVIATIONS

ALARA	As Low As Reasonably Achievable
AMA	Aging Management Activity
AMP	Aging Management Program
AMR	Aging Management Review
ANL	Argonne National Laboratory
ASME	American Society of Mechanical Engineers
ASTM	American Society for Testing and Materials
BRC	Blue Ribbon Commission on America's Nuclear Future
BWR	Boiling Water Reactor
°C	Degrees Celsius
CC	Criticality Control
CFR	Code of Federal Regulations
CoC	Certificate of Compliance
DCSS	Dry Cask Storage System
DE	Destructive Examination
DHC	Delayed Hydride Cracking
DSC	Dry Storage Canister
DOE	Department of Energy
DOT	Department of Transportation
DSA	Documented Safety Analysis
DWPF	Defense Waste Processing Facility
EIS	Environmental Impact Statement
EM	Environmental Management (DOE)
EPRI	Electric Power Research Institute
ER	Environmental Report

## ACRONYMS AND ABBREVIATIONS (continued)

EST	Extended Storage and Transportation
°F	Degrees Fahrenheit
FSAR	Final Safety Analysis Report
GTCC	Greater-Than-Class-C
GWd	Giga-watt days
HBU	High Burnup (> 45 GWd/MTU)
HE	Hydrogen Embrittlement
HEPA	High Efficiency Particulate Air
HLW	High-Level Waste
HPGe	High Performance Germanium detector
HRS	Hydraulic Ram System (NUHOMS)
HSM	Horizontal Storage Module (NUHOMS)
HT	Heat Transfer
INL	Idaho National Laboratory
INTEC	Idaho Nuclear Technology and Engineering Center
ISFSI	Independent Spent Fuel Storage Installation
ISG	Interim Staff Guidance
ITS	Important to Safety
KSI	Kips per Square Inch (1000 lbs/in <sup>2</sup> )
LWR	Light Water Reactor
MOX	Mixed Oxide (typically plutonium and uranium oxides)
MPC	Multipurpose Canister
MTHM	Metric Ton Heavy Metal
MTU	Metric Tonne of Uranium
MVDS	Modular Vault Dry Storage

## ACRONYMS AND ABBREVIATIONS (continued)

MW	Megawatt
N/A	Not Applicable
NAC	Nuclear Assurance Corporation
NDE	Nondestructive Examination
NRC	U.S. Nuclear Regulatory Commission
PB	Pressure Boundary
PCI	Pellet Cladding Interaction
PIE	Post-Irradiation Examination
PM	NRC Project Manager
PNNL	Pacific Northwest National Laboratory
psig	pounds per square inch, gauge
PVC	Polyvinyl Chloride
PWR	Pressurized Water Reactor
RS	Radiation Shielding
SAR	Safety Analysis Report
SCC	Stress Corrosion Cracking
SER	Safety Evaluation Report
SRP	Standard Review Plan
SSC	System, Structure, and Component
SME	Subject Matter Expert
SNF	Spent Nuclear Fuel
SRS	Savannah River Site
SRNL	Savannah River National Laboratory
TLAA	Time-Limited Aging Analysis
TSS	Terminal Solid Solubility

**ACRONYMS AND ABBREVIATIONS** (continued)

VCC	Ventilated Concrete Cask
VLTS	Very Long-Term Storage (alternate term used to describe consideration of dry storage beyond present licensing basis)
VSC	Ventilated Storage Cask System



## 1. INTRODUCTION

The United States strategy for the nuclear fuel cycle for its nuclear fleet, including the interim options for fuel management and waste form disposition, is under review by the Blue Ribbon Commission on America's Nuclear Future (BRC).<sup>1</sup> The final BRC report to the U.S. Department of Energy including a review of alternatives for storage, processing, and disposal of civilian used nuclear fuel and other materials is expected in January 2012; however, no timetable has been established for decisions by the U.S. Department of Energy on alternatives selection. In addition, the technology development needed for full implementation of any selected alternative may require several additional decades of study and work. Therefore, fuel presently in either wet or dry storage systems may need to be stored well beyond the initial and re-license periods, and the transfer of used fuel from commercial utilities to the U.S. DOE may not occur in the near future.

The U.S. Nuclear Regulatory Commission (NRC) has sponsored an independent evaluation of the bases for predicting materials behavior in dry cask storage systems (DCSS) for extended storage (> 120 years) and transportation (EST) because the spent nuclear fuels may need de facto storage in a DCSS for multiple decades. A time frame up to 300 hundred years is considered a reasonable bound to this need and one purpose of the evaluation is to summarize information, relative to licensing strategies, needed to address such an extended storage schedule. This report describes the results of the material aging evaluation and the aging management activities considered necessary for extended storage and transportation.

### **1.1 Objectives of the Aging Evaluation for EST**

Independent Spent Fuel Storage Installations (ISFSI) must provide for functions important to nuclear safety including thermal performance, radiological protection, confinement, sub-criticality control, and retrievability. These functions are required by the code of federal regulations governing irradiated fuel storage in 10 CFR Part 72 [2], and are provided by the systems, structures, and components (SSCs) of the dry storage systems as directed through the Standard Review Plan process [3, 4]. The original licenses granted by the NRC for DCSS's were for 20 years.

The fact that extended dry storage of spent nuclear fuel in DCSSs may be required was recognized and a technical basis for such storage was developed and summarized in a major report in 2002 [5]. The focus of that work was on age related degradation of the fuel and cask materials. A potential storage period of up to 100 years was considered, and the work concluded that "the review conducted in this report finds no technical reason why extended storage (beyond 20 years) would lead to loss of any of the required safety functions." Recently, formal consideration of the license period (and re-license period) was extended to 40 years through revision of 10 CFR Part 72 and the template of the Standard Review Plan process [6].

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<sup>1</sup> The Blue Ribbon Commission on America's Nuclear Future (the Commission) was established in accordance with the provisions of the Federal Advisory Committee Act (FACA), as amended, 5 USC. App. 2, and as directed by the President's Memorandum for the Secretary of Energy dated January 29, 2010: Blue Ribbon Commission on America's Nuclear Future. This charter establishes the Commission under the authority of the US Department of Energy (DOE). [1]

The storage period for consideration in the current analysis was arbitrarily chosen as 300 years, and includes high burn-up fuels that were not considered in the earlier evaluations. Therefore, one objective of the evaluations included in this present report is to expand the earlier considerations by extending the potential time of storage, including high burn-up fuels, and analyzing previous and emergent data for potential gaps in the data base. For example, the mechanical properties of zirconium based alloys are sensitive to crystallographic texture and texture is sensitive to the thermal-mechanical history of the alloy. These sensitivities raise a question about the relevance of mechanical property data developed on one alloy to another new alloy fabricated by different processing technologies.

The Standard Review Plan [6] for renewal of ISFSI licenses and DCSS CoC's states that "the renewal is primarily a materials engineering effort" and provides a flow chart (Figure A of the SRP [6]) for the license renewal process. An Aging Management Review, the development of Aging Management Programs and the assurance of post-storage fuel retrievability are key elements to the flow chart. Previous evaluations have listed potentially active age related degradation mechanisms, in fuels for example, and evaluated the likelihood that the mechanisms will be active during the initial license period or during extended storage (Table 3-2 from reference [5]).

The primary objective of this document is to identify and discuss materials degradation issues that may impact the safety functions required for the DCSSs to maintain low and high burn-up fuel in EST. The types of materials used in the existing DCSSs [7] were considered. The secondary objective of this document is to provide recommendations for activities (e.g., experiments, lead cask tests, monitoring programs, analyses, or other aging management activities) needed to address the aging issues, reduce uncertainties, and develop a sufficient technical basis for EST.

The 10 CFR Part 72 requirement for fuel performance in a DCSS is for "no gross rupture" of the fuel cladding for design basis conditions. The canister or cask is credited to provide the confinement function in a DCSS, and a defined leak rate of a shipping cask or overpack provides the confinement control for transportation; nevertheless, the viewpoint of the present evaluation is to avoid gross failure of the fuel under an extended dry storage period which is followed by transportation for final disposition, so that retrievability, even following normal conditions of transport, is maintained throughout EST. Transportation of irradiated fuel is governed by 10 CFR Part 71 [8] and the Standard Review Plan (SRP) for Transportation Packages for Spent Nuclear Fuel [9] provides guidance for the review and approval of applications for packages used to transport spent nuclear fuel under 10 CFR Part 71.

## **1.2 Approach**

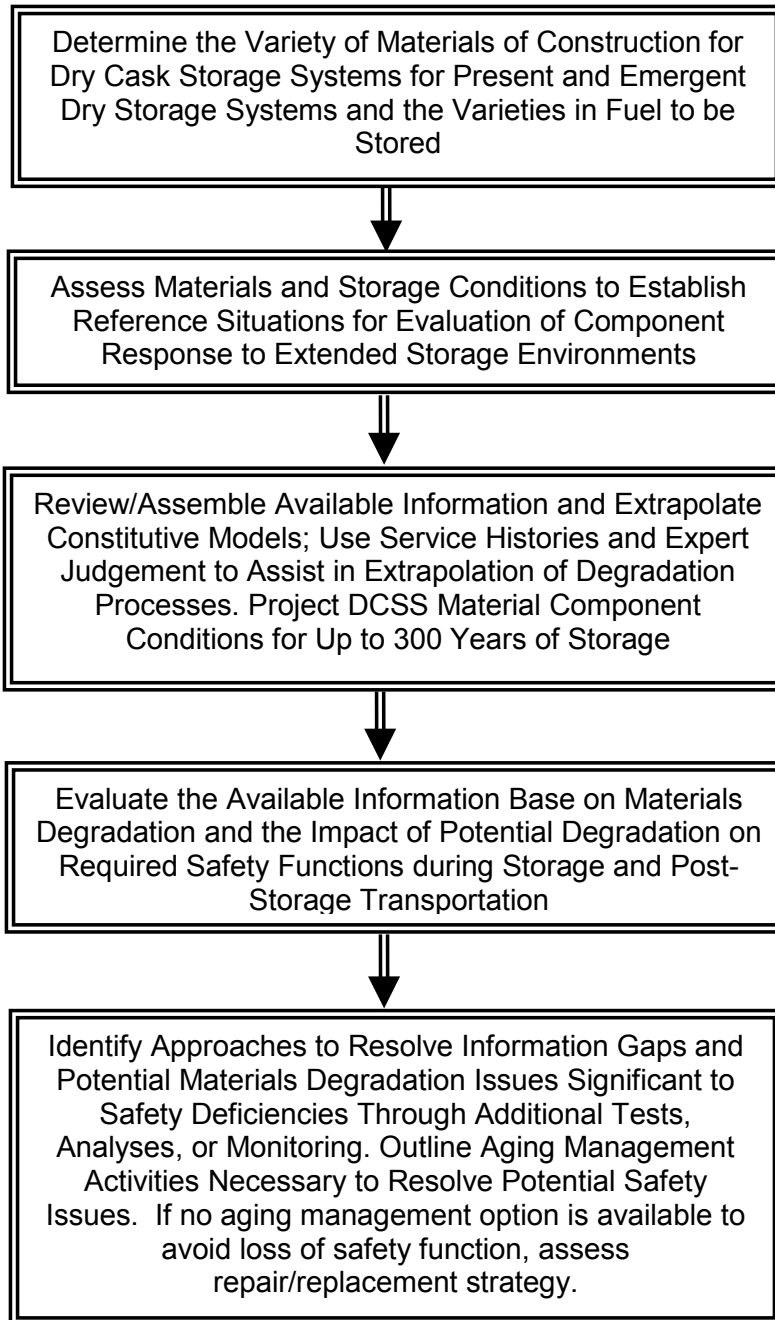
The degradation evaluation considered a set of materials and components currently used in DCSS and included selected emergent materials that may be used in future systems. The information for selecting the materials for evaluation was drawn from the general literature on dry storage systems. The degradation types and mechanisms potentially active for fuel and non-fuel materials in the SSCs under long-term dry storage are drawn from previous degradation analyses contained in key topical reports, and are supplemented with recent literature information and by considerations of the authors of this report. Nuclear power plant aging and aging management documents also provide input to this evaluation. Additionally, emergent information on HBU fuel and extended

storage systems and their impact on materials, components, and degradation evaluations are included.

Except for the fuel, the initial conditions of all other SSCs of the dry storage system are assumed to comply with the applicable national consensus codes and standards for design and construction, and have appropriate controls on workmanship and quality. Therefore, the as-designed, as-built conditions of the SSCs are not susceptible to failure as a result of defects in their initial material's condition or because of poor fabrication practices.

The flowsheet in Figure 1 depicts the approach taken to identify materials issues or gaps in the technical bases necessary to demonstrate the capability of the SSCs to perform their intended safety functions under extended storage and during transportation after storage. Uncertainties in the information base regarding the ability of the SSCs to perform the safety functions are qualitatively discussed and engineering judgment is applied to the evaluation.

ASTM C-1562-10 (*Standard Guide for Evaluation of Materials used in Extended Service of Interim Spent Nuclear Fuel Dry Storage Systems*) [10] is a national consensus guide aimed at aging and degradation of materials of the SCCs of DCSS for a target time of up to 100 years total service. This guide was reviewed and provided many of the inputs for SSCs, their materials of construction, and potential degradation mechanisms that were considered in this report.



**Figure 1** Steps to Identify Materials Aging Issues for EST and Provide Resolution Through Aging Management for SSCs in DCSS.

Section 2 of this report evaluates the potential degradation phenomena and the impact of the degradation processes on the functions important to safety. The authors of this report and other subject matter experts evaluated potential degradation mechanisms and the potential of an operative mechanism to cause effects detrimental to the safety functions. The findings determine whether sufficient bases exist for extended dry storage and, when such bases do not exist, the report makes recommendations for

additional tests, analyses, periodic monitoring, and other activities to support the technical bases for EST.

Emphasis is placed on post-storage retrievability to assure that the 10 CFR Part 72 requirement for retrievability from the dry storage system and the 10 CFR Part 71 requirements for transportation and removal from the storage canister are met. Specifically, 10 CFR 72.122(h)(1) requires that spent fuel cladding must be protected from degradation that leads to gross rupture, or the fuel must otherwise be confined so that degradation of the cladding will not impose operational safety problems. Further, 10 CFR 72.122(h)(1) requires that the storage system must be designed to allow ready retrieval of the spent fuel from the storage system for further processing or disposal.

The odd subsections 2.1, 2.3, etc. in Section 2 of this report describe the component and materials of the DCSS; the even subsections 2.2, 2.4, etc. describe the corresponding potential degradation types and their impact on the safety functions provided by the component. These even subsections include identification of the data needs and recommendations to address the needs. The GALL report [11] identifies Aging Management Programs (AMP) that were previously determined to be acceptable to manage aging effects of systems, structures, and components (SSC) in the scope of license renewal, as required by 10 CFR Part 54, "Requirements for Renewal of Operating Licenses for Nuclear Power Plants," [12]. The template provided by the GALL report provides the foundation for the set of actions for the aging management program that this report recommends for the DCSS.

Section 3 summarizes the activities recommended in section 2 that, along with the body of work referenced in this report and supporting information, would establish the technical bases for enabling EST, and categorizes the activities into Maintenance; Monitoring/In-Service Inspection; and Testing & Analysis bins. Maintenance activities are periodic actions that are pre-emptive to loss of function. Monitoring/In-Service Inspection activities provide for a periodic condition assessment of the component and would disposition conditions found to be below the condition at installation. Testing & Analysis activities are primarily R&D activities to provide materials degradation information and strengthen the technical bases to predict and demonstrate component integrity throughout targeted service life. Section 3 also outlines the type of information to be gained from a test cask demonstration that would, for example, challenge (e.g., partially initially degraded, HBU) the fuel in a DCSS to provide validation of predictions and verification of materials' conditions.

A risk-based evaluation to demonstrate the long-term acceptability of the DCSS is outside the scope of this report.

### **1.3 Assumptions**

The first assumption in the development of this document was that the original placement of spent fuel in the DCSS conformed to the licensing requirements at the time of placement. Most of these requirements were based on an assumption that the time of storage in a dry cask system would be, at most, several decades prior to transport and repository disposal. The recent U.S. Department of Energy decision to review its policies on managing the back end of the nuclear fuel cycle, including alternatives for processing and disposal, has significantly lengthened the potential time requirement for storage. This lengthening of potential storage time prompted additional evaluations to

explore extension of the licensing period for storage of nuclear fuel. The evaluations have focused on fuel discharged from the U.S. commercial fleet and subject to licensing by the U.S. Nuclear Regulatory Commission

To provide a reasonable upper bound on the period for storage, a time of 300 years is considered. Therefore, the evaluations and recommendations are designed to extend the bases for safe storage from the present technical and licensing bases<sup>2</sup> to a time well beyond anticipated needs. Three basic assumptions to provide for safety and security of DCSS are made:

- 1) the personnel and infrastructure necessary to support the storage site will be maintained and available throughout the storage period;
- 2) the SSCs of the DCSS important to safety must be capable of providing/performing their design safety functions throughout the storage period. This includes the assumption that the fuel cladding must be protected from degradation that leads to gross rupture throughout the storage period and also during post-storage, normal conditions of transportation and up to its acceptance for reprocessing or its placement in a repository for ultimate disposition; and
- 3) the potential storage environments and off-normal events outlined in NUREG-1927 [6] represent the range of storage conditions encountered by the DCSS during the storage period with additional consideration of recent reports on general climate change conditions expected in the U.S..

These assumptions establish the foundation that provides the basis for this report to:

- 1) define the potential for various age-related degradation processes, 2) establish the probable impact of potentially operative processes on functions important to safety, and 3) recommend strategies to mitigate and/or repair the negative impacts of materials and system aging. Each of these assumptions is briefly discussed in Sections 1.3.1 through 1.3.3

### **1.3.1 Management Strategy and Infrastructure**

The long-term safety and security of the DCSS requires continual corporate control of the storage site. This control may be maintained using a variety of technologies, all of which will require facilities, trained staff and the resources to support both the personnel and infrastructure. A primary assumption of this report is that staff and facilities are available to provide the support necessary to conduct any operation required to safeguard the dry storage facility and assure nuclear safety throughout the storage period. The staff and/or facilities may be maintained at, or near, the storage site or may be readily mustered to the site to conduct the required operations. Staff functions throughout the storage period must include, but are not limited to:

- 1) implementation of security and controls to preclude unintentional and/or intentional access to the facility by unauthorized personnel and to preclude human theft or sabotage,

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<sup>2</sup> The present technical bases address materials and SCCs fuel storage to enable a regulatory basis for licensing for up to 60 years wet storage at a NPP followed by up to 60 years dry storage at an ISFSI.

- 2) implementation of a maintenance program and a monitoring program for the DCSS; and
- 3) capability to recover and re-pack a canister, repair degrading infrastructure, and/or relocate components and systems to nearby or remote facilities that have, or will have, an impact on the fuel or DCSS components important to safety.

### **1.3.2 Functions Important to Nuclear Safety**

To support of 10 CFR Part 72, NRC's NUREG-1536 [3] and NUREG-1567 [4] the functions important to safety are identified and a template for evaluation of the SSCs to meet the requirement of Part 72 is provided. Nuclear safety functions required by 10 CFR Part 72 and listed in NUREG-1536 include thermal performance, radiological confinement, subcriticality control, and retrievability. These safety functions are the same regardless of the differences among the storage or transport conditions and each of these functions relies on the continual integrity of the materials in the DCSS.

Components of the fuel and container are especially important because of the potential for degradation processes to lead to fuel fragmentation, loss of container integrity, and other structural alterations that could directly impact confinement, sub-criticality control and/or retrievability. Each of these safety functions relies on the continual integrity of the materials in the DCSS.

Additionally, deterioration of associated structures could indirectly impact safety by adversely impacting the monitoring program. Periodic monitoring is essential to assuring the long term nuclear safety because potential degradation mechanisms, including stress corrosion cracking and several of the hydrogen-induced degradation phenomena, are nucleation and growth processes, thus satisfactory behavior cannot be reliably extrapolated beyond the realm of experience. Reliable determinations of the behavior of materials and components during the time frames required for long term storage cannot be assured in the absence of a periodic monitoring program and such a program requires the continuing ability to access, and relocate if necessary, the dry cask, the fuel container and/or the fuel assembly. Therefore, maintenance of nuclear safety functions for dry cask storage system as required by 10 CFR Part 72 would mandate the following under extended storage:

- 1) determine materials degradation processes that could lead to corrosion, cracking, hydrogen induced degradation and other potentially operative age related processes;
- 2) provide verified, time-dependent models for any degradation process of potential concern and continually provide the data necessary to assess the models used. If no suitable models are available, use expert engineering judgment to assess material behavior;
- 3) conduct the monitoring and analysis necessary to verify near-term extrapolations of the models, refine the models for longer term extrapolation and determine materials conditions; and

- 4) establish provisions for satisfactory transfer, if necessary, of corporate responsibilities to other qualified entities even when the time frame of storage exceeds the corporate life of the responsible party.

### **1.3.3 Potential Storage Environments**

The present environmental conditions to which the materials of the DCSS are anticipated to be exposed are essentially the local climate of storage [3]. Additional environments of storage may occur due to climate change over the extended storage timeframe of 300 years. The current predictions of global and local climate changes include only a low (several degrees Fahrenheit) increase in average global temperature, and local changes in total rainfall, in frequency of heavy precipitation; and/or average temperatures [13]. These conditions do not identify any factors which would exceed the current ISFSI design parameters.

The currently predicted climate changes [13] have minimal impact on the general degradation processes because of the range of climate conditions already considered in the current DCSSs. Nevertheless specific features of a global climate change (e.g., increased atmospheric CO<sub>2</sub> concentration) may impact specific materials (e.g., reinforced concrete), and local climate considerations (e.g., local pollutants such as flyash) may impact specific materials (e.g., stainless steel canisters). Prolonged exposure to ambient conditions of a DCSS such as a marine environment may also lead to conditions (e.g., salt deposits on stainless steel canisters) detrimental to the materials of the DCSS. These specific conditions and their impact have been identified in the report.

### **Section 1 References**

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- [3] NUREG-1536, Standard Review Plan for Dry Cask Storage Systems. <http://www.nrc.gov/reading-rm/doc-collections/nuregs/staff/sr1536/>.
- [4] NUREG-1567, Standard Review Plan for Spent Fuel Dry Storage Facilities. <http://www.nrc.gov/reading-rm/doc-collections/nuregs/staff/sr1567/>.
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## 2. AGING OF DRY STORAGE SSCS UNDER EST

Section 2 of this report evaluates the potential degradation phenomena and the impact of the degradation phenomena on the functions important to safety. Subsections 2.1, 2.3, etc. in section 2 describe the component and materials of the DCSS, and subsections 2.2, 2.4, etc. describe the corresponding potential degradation types and their impact on the safety functions provided by the component. Subsections 2.2, 2.4, etc. also include identification of the data needs and recommendations to address the materials aging issues.

The SSCs considered in the EST degradation evaluation include the past and present fuel designs, emergent fuels (e.g., MOX), and the non-fuel SSCs of DCSS that have been listed in references [1] and [2]. The importance to safety of the components of the storage systems and transportation packages are described in reference [3].

The initial or starting condition directly affects the type and extent of the degradation that might occur during long term dry storage. The materials and fabrication assumed in this report are listed as “reference conditions” for the respective SSC and are assumed to comply with applicable codes and standards for design and construction as directed in the SRPs for storage [4] and transportation [5]. Only the fuel is assumed to be in an initially “degraded” condition by virtue of its burn-up in the reactor.

### **Section 2 References**

- [1] ASTM C-1562-10, *Standard Guide for Evaluation of Materials used in Extended Service of Interim Spent Nuclear Fuel Dry Storage Systems*, American Society for Testing and Materials, reapproved 2010.
- [2] Electric Power Research Institute, report 1021048, “Industry Spent Fuel Storage Handbook,” July 2010.
- [3] NUREG/CR-6407, INEL-95/0551, *Classification of Transportation Packaging and Dry Spent Fuel Storage System Components According to Importance to Safety*, J.W. McConnell, Jr., A.L. Ayers, Jr., M.J. Tyacke, February 1996.
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- [5] NUREG-1617, *Standard Review Plan for Transportation Packages for Spent Nuclear Fuel*. <http://www.nrc.gov/reading-rm/doc-collections/nuregs/staff/sr1617/1617/sr1617.pdf>.

### **2.1 Fuel Description**

The fuel is considered as the entire assembly and includes the fuel pellets, fuel cladding, grid spacers, end pieces, and associated hardware. This “fuel” component is handled as an entire assembly in fuel movement and is the system that must be retrievable after EST. Figure 2 and Figure 3, respectively, show versions of PWR and BWR fuel assemblies. While some PWR assemblies have both top and bottom plenums/springs, the majority of the PWR assemblies in storage have only a top plenum and fuel spring.

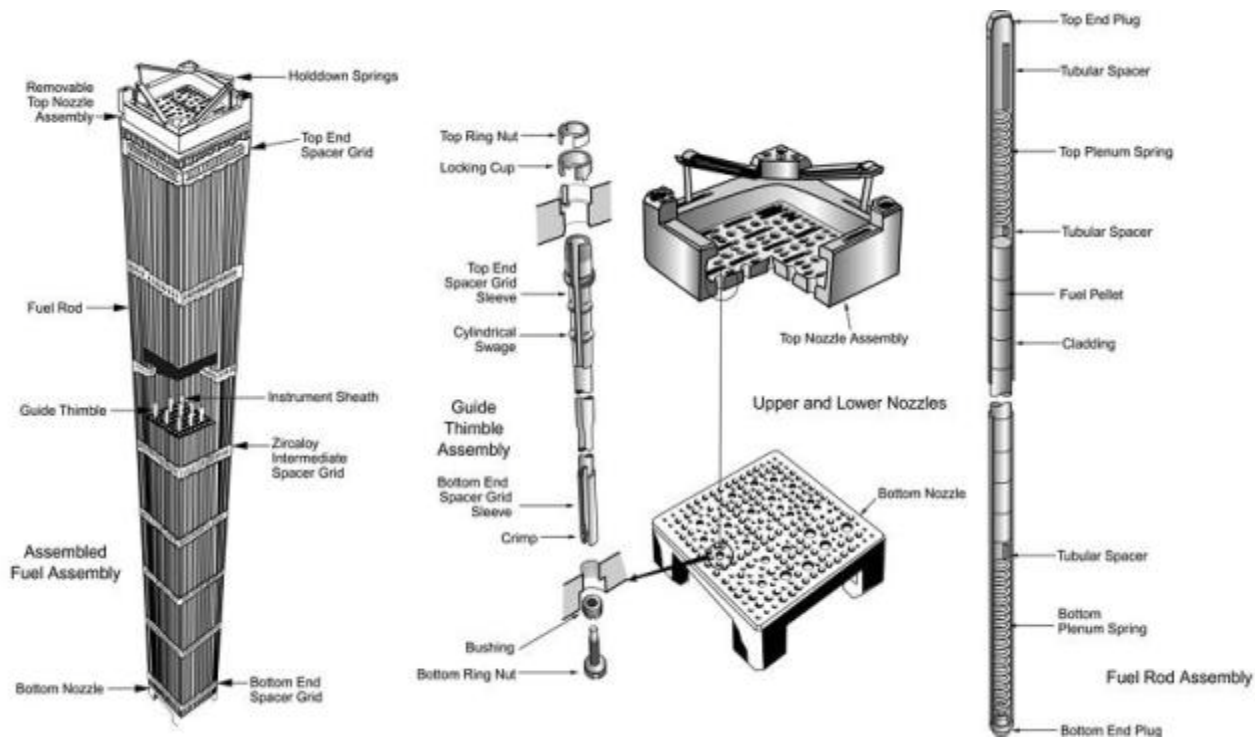


Figure 2 PWR Fuel Assembly.



Figure 3 BWR Fuel Assembly [<http://www.nucleartourist.com/images/bwrfuel1.jpg>].

The non-cladding materials for components of the PWR and BWR fuel assemblies are summarized below. It is important to note that the non-cladding materials used will depend on the material used for the fuel cladding.

**PWR Fuel Non-Cladding Materials:** (Grid Spacers, Thimble Guide Tubes)

Fuel with Stainless Steel Cladding: Inconel grids and SS thimbles.

Fuel with Standard Zircaloy-4 Cladding: Inconel and Zircaloy-4 grids and Zircaloy-4 thimbles.

Fuel with Improved Zircaloy-4 Cladding: Inconel and Zircaloy-4 grids and Zircaloy-4 thimbles.

Fuel with ZIRLO Cladding: Some Zircaloy-4 grids and thimble, Mostly ZIRLO grids and thimbles and some Inconel grids.

Fuel with M5 Cladding: Some M5 thimbles and grids and some Zircaloy-4 thimbles and grids.

The assembly end pieces/nozzles are stainless steel and Inconel.

**BWR Fuel Non-Cladding Materials:** (Grid Spacers, Channels)

Fuel with Standard Zircaloy-2 Cladding: Zircaloy channels.

Fuel with Improved Zircaloy-2 Cladding: Zircaloy channels.

The fuel pellets are currently uranium dioxide. However, mixed uranium-plutonium oxide fuel is planned for use in the existing fleet of LWRs.

The fuel material component that is most susceptible to degradation that could lead to a loss of retrievability is the fuel cladding. Although the other components of the fuel experience radiation damage and corrosion, and stress corrosion cracking of guide tube welds has created conditions that required repair to insure retrievability, the fuel cladding exhibits the greatest susceptibility to age related damage in terms of corrosion and hydride embrittlement. This higher susceptibility is due to the hydrogen uptake experienced during reactor service and the higher temperatures encountered during storage.

The wide variety of fuel cladding alloys used by the U.S. commercial fleet is significant because virtually every form of age-related degradation is sensitive to the composition of the alloy. Alterations in alloy chemistry designed to improve the in-reactor corrosion behavior, for example, may also alter the creep resistance, the susceptibility to hydrogen induced degradation and the mechanical properties of the alloy. Thermal/mechanical processing conditions, irradiation level, fuel drying conditions and other fabrication and service variables will also impact the behavior of the cladding during storage. Therefore the response of any fuel during EST will depend on the chemistry, thermal/mechanical and irradiation history and other material variables. Data and models to address, comprehensively across the variety of cladding alloys, the overall impact of these variables on cladding behavior do not exist. Therefore, the age related degradation

behaviors evaluated in this report do not describe the response of any specific cladding but use models and expert judgment to evaluate the generalized behavior of, for example, zirconium-based cladding alloys. Because of the very small inventory of stainless steel clad fuels, and their lack of susceptibility to aging mechanisms in dry storage relative to zircaloy clad fuels, these fuels are not explicitly considered in this report.

Table 1 shows the composition of cladding alloys used at present in the commercial fleet in the U.S. Sections 2.1.1 and 2.1.2 briefly describe these alloys and selected early cladding alloys and section 2.1.3 addresses potential alloys for future applications.

**Table 1** Nominal Compositions of Alloys Used as Nuclear Fuel Cladding

<b>Table 1</b>									
Nominal Compositions of Alloys Used as Nuclear Fuel Cladding									
Alloy	Sn (wt%)	Fe (wt%)	Cr (wt%)	Nb (wt%)	Ni (wt%)	O (wt%)	C (wt%)	Si (wt%)	Zr (wt%)
Zr-2 (BWR)	1.5	0.12	0.1		0.05	0.13			Balance
Zr-2 (BWR) Improved	1.3	0.17	0.1		0.06	0.13			Balance
Zr-4 (PWR) High Tin	1.55	0.22	0.12			0.12	0.015	0.01	Balance
Zr-4 (PWR) Improved	1.3	0.22	0.12			0.12	0.012	0.01	Balance
M5 ®		0.04		1		0.14			Balance
ZIRLO™	1	0.1		1		0.12			Balance
Optimized ZIRLO™	0.7	0.1		1		0.12			Balance
Zircaloy 2 Specification ASTM B811	1.2 - 1.7	0.07 - 0.20	0.05 - 0.15		0.03 - 0.08	0.09 - 0.16	0.027 max	0.012 max	Balance
Zircaloy 4 Specification ASTM B811	1.2 - 1.7	0.18 - 0.24	0.07 - 0.13			0.09 - 0.16	0.027 max	0.012 max	Balance

### **2.1.1 Early Domestic Cladding Alloy Types**

#### **Stainless Steel**

Initially and until the late-1970s, commercial fuel rods used austenitic stainless steel (primarily Type 304) as cladding. Some Type 348H stainless steel was also used [1], and one reactor system, Haddam Neck, used SS cladding through the 1980s [1]. Issues with stress corrosion cracking in the stainless steel cladding resulted in a shift to Zircaloy-2 for BWR applications [2]. In PWR reactors the SS cladding performed well regarding fabricability and corrosion resistance during operation, but the relatively high thermal neutron absorption cross section reduced the neutron economy and stainless steel cladding was phased out and replaced by Zircaloy-4. Therefore, only a small fraction of the commercial spent fuel assemblies are SS clad.

The stainless steel clad fuel may have a higher margin against degradation during long term storage due to lower burnup and fuel duties, and the absence of hydrogen-induced degradation or hydride-related issues. Furthermore, in a dry storage environment with inert gas cover and negligible moisture, stress corrosion cracking of the SS clad fuel in EST is not likely.

## **Zircaloy 2**

Zircaloy 2 (Zr2) was chosen to replace SS in BWR cladding applications. The Zr 2 alloy has about 98% zirconium with alloy additions of:

- Iron, chrome, and nickel to cause second phase precipitates for mechanical strength and improved corrosion resistance.
- Tin for solid solution strengthening and improved corrosion resistance, particularly to offset the adverse corrosion effects related to nitrogen contamination during production and abnormal coolant chemistry during operation.
- Oxygen is also typically added at about a 1200 ppm level for mechanical strength. During reactor operation corrosion of the exterior of the cladding by reaction with the coolant and the interior of the cladding reacts with the fuel to increase the oxygen level at the clad surfaces. At the operating temperatures the diffusion of the oxygen into the clad is relatively slow and most of the oxygen absorbed by the cladding is confined to surface oxide films. Oxygen diffusion is even slower at temperatures below the reactor operation temperature and little or no oxygen relocation is anticipated during long term dry storage.

Zircaloy 2 is generically categorized as grade R60802 in ASTM B811 and B353 and is used extensively for BWR cladding. Prior to 1991 the applicable ASTM specification was B353. This specification was replaced by ASTM B811. Because of the operating conditions the typical uniform oxide layer on BWR fuel is less than that on PWR fuel; however, nodular corrosion of the Zircaloy 2 in a BWR environment could cause local thin spots that can impact cladding behavior during EST.

## **Zircaloy 4 (high tin)**

Zircaloy 4 is defined by ASTM B811 grade R60804, although some vendors produce Zircaloy 4 cladding per their own specifications which provide for more specific controls within the variations allowed by the ASTM specification. Prior to 1991, the applicable ASTM specification for Zircaloy 4 was B353.

Zircaloy 4 is similar to Zircaloy 2 except Zircaloy 4 has the nickel removed to reduce hydrogen absorption from corrosion reactions that occur in the PWR environment. To offset the strength reduction by nickel removal more iron was added to the Zircaloy 4 [2].

The Zircaloy 4 specification has a tin range of 1.25 to 1.7%. The initial versions of Zircaloy 4 were produced with tin levels at 1.55% near the upper tin limit. Carbon is an impurity in zirconium alloys which has positive effects on microstructure control but at high levels has negative effects on corrosion resistance and irradiation-induced growth. Because of the negative effects the maximum carbon level is limited to 270 ppm. However, in the late 1980s, improved versions of Zircaloy 4 were introduced that provided tighter controls on tin and carbon.

The high tin Zircaloy 4 cladding has a lower corrosion resistance than the current low tin version; thus, even though the burn up levels and duties for the early fuels are below the current levels. The lower corrosion resistance could lead to a high hydrogen concentration in the cladding. The clad oxide thicknesses range up to and in some cases exceed the guidelines of 100 microns of oxide. Therefore, because of the higher oxidation levels and the associated hydrogen absorption by the cladding, early fuels may be clad with alloys having higher than anticipated hydrogen contents.

Some of the Zircaloy 4 clad fuel has experienced significant oxide spallation. Although the spallation does not typically result in a clad breach, the spalling oxide presents a particulate source term and can promote localized hydriding that may impact cladding behavior during storage and transportation.

### **2.1.2 Current Domestic Cladding Alloy Types**

There are primarily four cladding alloy types currently used in large quantities and in multiple fuel regions of commercial reactors. These alloys are lined Zircaloy 2, improved Zircaloy 4, M5<sup>®</sup>, and ZIRLO<sup>™</sup>. Optimized ZIRLO<sup>™</sup> is being introduced for very high duty applications and is present as multiple regions in a few reactors.

The typical alloy element levels for these alloys are shown in Table 1, and the alloys are discussed in the following paragraphs. The cladding alloy of the fuel assembly is important to long-term storage considerations because the tendency for zirconium based alloys to experience age-related degradation depends on alloys content, fabrication history, crystallographic texture, thermal history, irradiation level and other materials/environmental variables. The impact of the cladding alloy and the mechanical history of the alloy on material behavior during long-term storage must be considered before the anticipated long term behavior of any specific fuel can be estimated. The degradation processes such as creep, hydrogen-induced degradation, stress corrosion cracking and corrosion resistance are all dependent on the alloy type and processing history but are discussed in a general way in Section 2.2 of this report.

#### **Zircaloy 2 – Lined**

The bulk of the BWR fuels are now clad with lined Zircaloy 2. The Zircaloy 2 clad is similar to the earlier versions but has iron and nickel adjusted towards the upper limits and tin adjusted towards the lower limit of the ASTM range [3].

The ID liner which is “softer” than the base clad is pure zirconium or zirconium with small amounts of alloy additions. The liner is designed to accommodate the strains resulting from pellet expansions that occur when the reactor core power level changes. The lining also aides in mitigating stress corrosion assisted cracking from the clad ID surface; a condition that can lead to PCI (pellet clad interaction) failures.



### **Improved Zircaloy 4 – Low Tin**

The increase in fuel duty and burn up required that the cladding have more margins to the corrosion and hydrogen design limits. For PWRs applications Zircaloy 4 was improved with a reduction in the tin level and improved controls over impurity/alloy elements such as carbon and nitrogen optimized through an annealing process to produce a consistent and desirable microstructure, referred to as A-Time control. The improved versions of Zircaloy 4 were introduced by fuel vendors in the late 1980's. The improved corrosion resistance of this material supported operations to higher burn-up. The higher duties have caused some of the fuel cladding to have oxide and hydrogen levels near the design limits [4, 5, 6] thus clearly impacting the tendencies for hydrogen-induced degradation in this alloy.

### **M5® Alloy**

AREVA introduced the alloy M5® for fuel applications in the 1990's and 16 U.S. commercial reactors have used or are using M5® clad fuel [7]. This alloy is now considered the AREVA PWR reference alloy. M5® is an alloy from the Zr – 1% Nb family and has no additions of tin and chrome like the Zircaloys and only minor amounts of iron (400 ppm) and oxygen (1400 ppm). Sulfur at about the 25 ppm level is also added to improve the creep strength. Performance data indicates that in reactor operation the clad oxidation is significantly reduced compared to Zircaloy 4 for the same burn up conditions. The typical M5® fuel would be expected to have less than 40 microns peak oxide at a discharge burn up of 62 GWD / MTU. At this same discharge burn up the hydrogen levels are also relatively low, and expected to be less than 200 ppm. M5® is also reported to have higher residual irradiated ductility compared to Zircaloy 4.

### **ZIRLO™**

ZIRLO is a Zr -1% Nb - 1% Sn - 0.1% Fe alloy with the typical addition of 0.12% O [8]. Westinghouse introduced ZIRLO in early 1990's as a replacement for Improved Zircaloy 4 for higher duty operations and it has become the reference alloy for Westinghouse fuel cladding with most if not all of the Westinghouse fueled domestic reactors using ZIRLO cladding. ZIRLO has enhanced corrosion resistance and lower hydrogen levels compared to Zircaloy 4 for equivalent burn ups and fuel duties. With the tin and iron additions ZIRLO is a robust alloy that is resistant to coolant chemistry variation effects. ZIRLO is also resistant to oxide spalling which is observed in some high burn up Zircaloy 4 fuel.

### **Optimized ZIRLO™**

Optimized ZIRLO was developed to provide added performance margin for high duty fuel applications [9]. Optimized ZIRLO has the basic characteristics of standard ZIRLO but has a tin level reduced to a range of 0.6% to 0.8% while maintaining the niobium at 1% and iron at 0.1% levels. The reduced tin improves the corrosion resistance compared to standard ZIRLO and the peak oxide thicknesses are less than 40 microns at discharge burn ups of 62 GWD/MTU. In

2005 the NRC approved optimized ZIRLO for use in reactor fuel and as of 2010 the alloy is in use as regions in a few commercial reactors.

### **2.1.3 Potential New Advanced Metal Alloy Cladding**

The fuel vendors are evaluating other advanced alloys for future applications. For example an alloy called GNF-ZIRON™ which is a high iron modification to the improved Zircaloy 2 [10] is being tested and a new family of alloys in the AXIOM program [11] is being evaluated. AREVA has reported test results on a set of alloys which are a modified M5 with increased levels of tin and iron [12].

The diversity in fuel cladding alloy composition and the anticipated emergence of new cladding alloys couples with the alloy-to-alloy difference in cladding behavior to make the estimation of a specific fuels response to EST difficult and make the data requirements for such an estimation extensive, if not prohibitive. Material behavior also varies with numerous process and exposure variables and the data required to predict behavior of new and emergent alloys simply do not exist at this time.

### **Section 2.1.1 – 2.1.3 References**

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- [2] Schemel. J.H., *Zirconium Alloy Fuel Clad Tubing Engineering Guide*, Sandvik Special Metals Corporation, December 1989.
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- [8] Sabol, G. et.al., *Development of a Cladding Alloy for High Burnup* ; ASTM STP 1023 8<sup>th</sup> International Symposium on Zirconium in the Nuclear Industry, 1989 pp. 227-244.

- [9] Wikmark, G. et.al., *Cladding to Sustain Corrosion, Creep and Growth at High Burn-ups*; Nuclear Engineering and technology, Vol. 41 No. 2 March 2009-Special Issue on the Water Reactor Fuel Performance Meeting 2008.
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- [11] Pan, G. et.al., *Advanced Material for PWR Application: AXIOM<sup>tm</sup> Cladding* 2010 LWR Fuel Performance M Top Fuel; September 2010 Orlando Florida.
- [12] Chabretou, V. et.al., *Ultra Low Tin Quaternary Alloys In-Pile Performance Impact of Tin Content on Corrosion and Mechanical Resistance*; ASTM 16<sup>th</sup> International Symposium on Zirconium in the Nuclear Industry; May 2010 Chengdu, China.

#### **2.1.4 Additional Fuel Design - Fort St. Vrain SNF**

The following information describing Fort St Vrain was summarized from reference 1.

The Fort St Vrain reactor, a high-temperature, gas-cooled reactor (HTGR) was the only commercial thorium-fueled nuclear plant in the U.S. This reactor, developed from the AVR in Germany, was operated from 1976 until 1989. It was a high-temperature (700°C), graphite-moderated, helium-cooled reactor with a Th/HEU fuel designed to operate at 842 MWth (330 MWe). The fuel is in microspheres of thorium carbide and Th/U-235 carbide coated with silicon carbide and pyrolytic carbon to retain fission products. Fort St. Vrain spent fuel elements are graphite elements in hexagonal arrays with a 36.0 cm maximum cross section and 79.3 cm in length. The fuel achieved 170,000 MWD/MT burn-up.

A total of 1464 HTGR spent fuel assemblies (15.4 MTHM) have been stored in the Modular Vault Dry Storage (MVDS) since 1991. The fuel is a highly enriched uranium/thorium carbide fuel that operated at a maximum heat output of 150 watts per assembly and an average heat output of 85 watts per assembly. The Fuel Storage Canister is a cylindrical carbon steel tube body with an exterior aluminum spray coating, double metal O ring seals, and a bolted lid. Nitrogen is used to backfill the Fuel Storage Canister (FSC).

The SiC coating of TRISO fuel (Fort St. Vrain) is the primary confinement of fission products and is expected to be stable for >1000 years in a repository environment [2]. This reference, coupled with the design of the fuel supports the conclusion that no fuel-generated problems will result if the fuel is left in dry storage of 300+ years. Therefore, the behavior of this fuel is not discussed any further in this report. Monitoring of the carbon steel storage canisters and the double metal o-ring seals should be performed to detect the onset of any corrosion or leakage and to correct, as necessary.

## **Section 2.1.4 References**

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## **2.1.5 Additional Fuel Design - Mixed Oxide Fuel (MOX)**

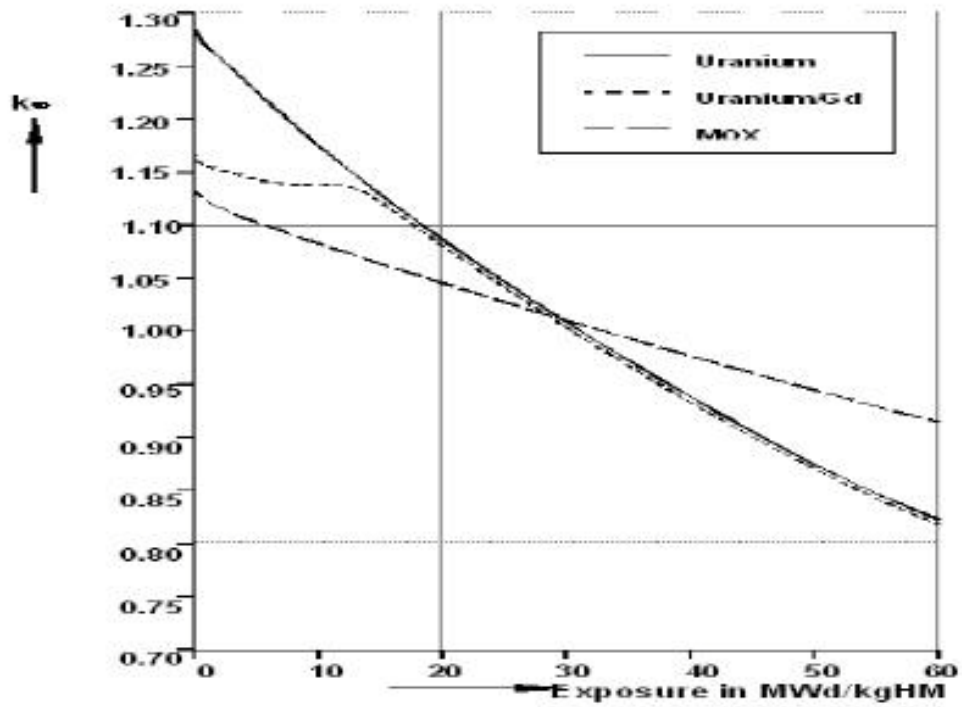
The following information describing Mixed Oxide Fuel was taken primarily from references [1, 2, and 3].

MOX fuel, consisting of about 7% plutonium mixed with depleted uranium, is equivalent to a uranium oxide fuel enriched to about 4.5% U-235, assuming that the plutonium has about two-thirds fissile isotopes. If weapons plutonium is used (>90% Pu-239), only about 5% plutonium is needed in the mix. Under the Plutonium Management and Disposition Agreement, Russia and the U.S. agreed in 2000 to dispose of 34 tonnes of weapons-grade plutonium deemed surplus to military requirements. A Mixed Oxide Fuel Fabrication Facility (MFFF) is being constructed at the Savannah River Site and will convert the U.S. plutonium to MOX fuel. Expected to begin operations in 2016, the MFFF is designed to turn 3.5 t/yr of weapons-grade plutonium into MOX fuel assemblies. This use of MOX fuels in U.S. reactors coupled with the trial irradiation of four MOX fuel lead test assemblies at the Catawba plant will place MOX fuel in the U.S. spent fuel inventory.

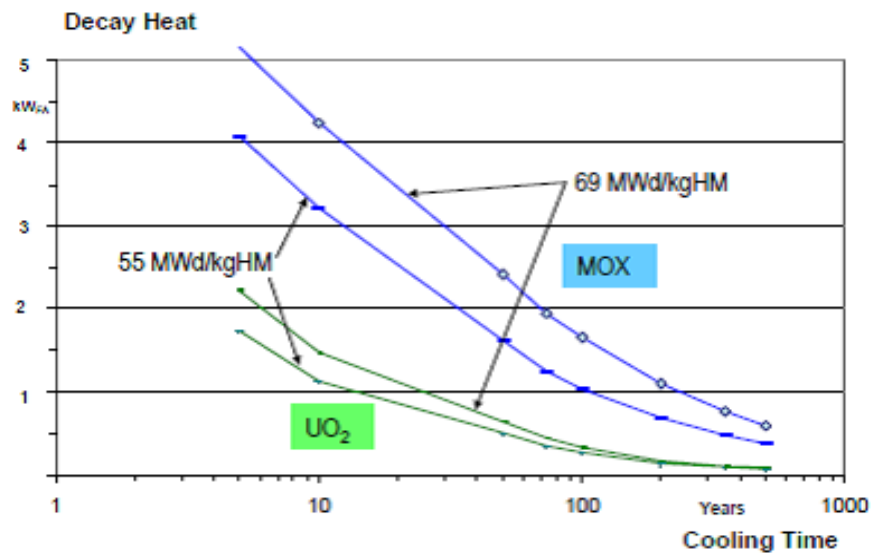
The post irradiation behavior of MOX differs from that of UO<sub>2</sub> fuels including a difference in the reactivity changes with time.

The MOX reactivity curve is flatter than that for Uranium (Figure 4). The point of intersection depends on the relation between U-235 enrichment and Pu fissile content. A high preponderance of the Pu fissile content shifts the point of intersection to the left and leads to a considerably higher thermal load of the MOX-rods at burnups beyond 20 to 30 GWd/MTHM. For dry storage the evolution of the decay heat and the neutron activity with burnup is important to assess cask temperatures and shielding capacities. A burnup increase of 25% from 55 to 69 GWd/MTHM increases the decay heat of both UO<sub>2</sub> and MOX fuel assemblies (FAs) by roughly the same amount (Figure 5). However, the decay heat of a UO<sub>2</sub>-FA with around 2 kW at the beginning of storage after 5 years cooling time is less than a half of a MOX FA of comparable range of burnup. With increasing cooling time the decay heat decreases, however more and more slowly. Whereas for a UO<sub>2</sub>-FA it takes about 10 to 20 years to drop below 1 kW, it takes roughly 100 to 200 years for a MOX FA.

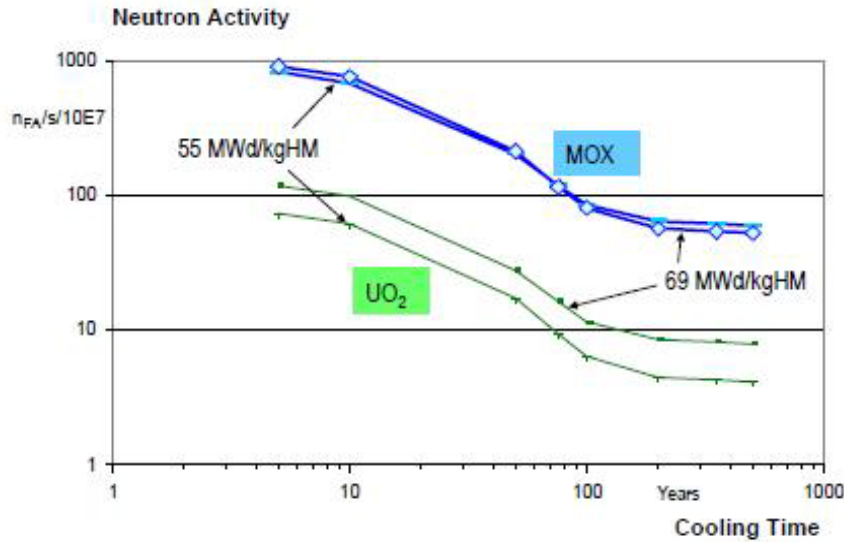
Additional information on weapons grade MOX (WG-MOX that contains Pu-239 only as initial fissile specie) characteristics for extended dry storage compared to UO<sub>2</sub> fuel shows similar results to those below for the U-235/Pu-239 MOX [4].



**Figure 4** Reactivity of MOX and  $UO_2$  fuel depending on burnup [reproduced from ref. 1].



**Figure 5** Decay heat of  $UO_2$  and MOX FAs depending on burnup and cooling time [reproduced from ref. 1].



**Figure 6** Neutron activity of UO<sub>2</sub> and MOX FAs dependent on burnup and cooling time [reproduced from ref. 1].

The neutron activity exhibits somewhat different features than the decay heat (Figure 6). Between 10 and 100 years cooling time it drops by one order of magnitude and above 100 years the reactivity remains relatively constant. The difference between MOX and UO<sub>2</sub> FAs amounts to roughly a factor of ten. With regard to a storage extension, the graphs show that it takes about 100 years cooling time for both decay heat and neutron activity of MOX fuel to reach the UO<sub>2</sub> level at the beginning of storage.

The large difference in the time-heat load profiles of MOX fuel compared to UO<sub>2</sub> fuel mandates thermal management of the MOX fuels if it is to be loaded in a DCSS to avoid high temperatures that exceed design storage limits.

### **Section 2.1.5 References**

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and to be published in the proceedings of the International High-Level Radioactive Waste Management conference, Albuquerque, NM, April 10-14, 2011.

### **2.1.6 Effects of Burnup, Pool Storage, and Drying on Fuel Condition**

Low burn-up fuel has been defined for dry storage as being fuels at burn-up levels of 45 GWd/MTU and lower. High burn-up fuel is above 45 GWd/MTU and nominally up to licensed limit of about 62.5 GWd/MTU. Changes occur in fuel with burnup or fuel duty as is shown pictorially in Figure 7. Both the fuel pellet and the cladding experience significant changes that can impact the performance of the fuel in DCSS and post-storage retrievability and transportation.

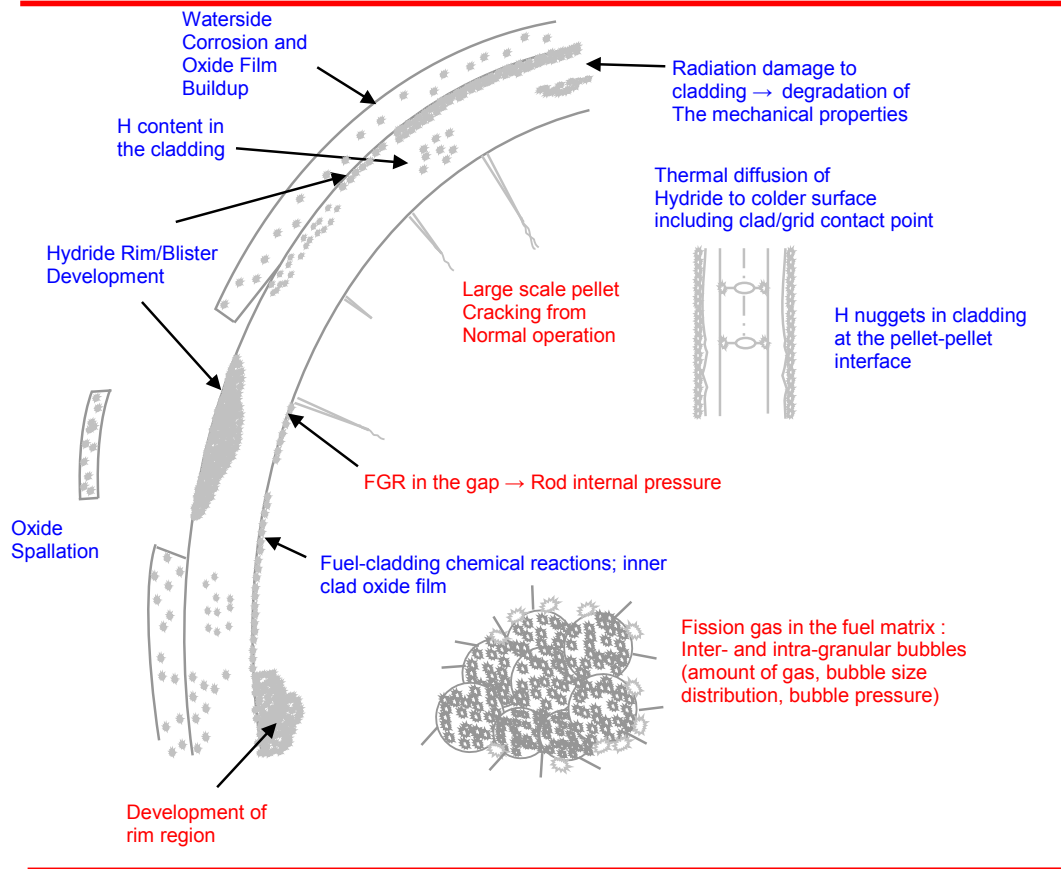
Some generic changes to the fuel rod during reactor operation include:

- a) the fuel pellet develops fines at the rim of the pellet and undergoes cracking to enable fission gas release (FGR) to the cladding; and
- b) the cladding undergoes radiation damage and embrittlement; corrosion and oxide scale formation/spallation; hydrogen ingress, hydriding, embrittlement; and creep.

In addition, fuel selected for placement in dry storage may have been in spent fuel storage pools for very long times and the physical characteristics of the fuel and fuel assemblies may have been altered by the storage and/or drying processes, and for this reason, the drying protocol used for the fuel is important. Pin holes, hairline cracks and other defects that would have released gaseous species but do not present a potential pathway for gross escape of fuel particles are considered minor, and fuel with such damage may be stored directly in the dry storage cask. Fuel with clad penetrations, however, is likely to contain water from the storage pool and needs to be carefully considered because of the potential for oxidation and swelling.

Fuels showing significant damage may also be stored in a cask after the damaged assembly and fuel debris are placed in a metallic fuel can that meets the fuel and system specific functions required for non-damaged fuels. Fuels placed in a dry storage cask, the non-damaged fuels, the fuels with insignificant damage and the degraded and canned fuels must all satisfy the same functions important to safety including, criticality, shielding, thermal and retrievability.

The changes to the pellet and cladding with burnup in the reactor, and the effects of pool storage and drying are summarized in this subsection. Information on the effects of burnup and fuel type on fuel characteristics is drawn from a recent IAEA publication [1] and other listed references in this subsection.



**Figure 7 Fuel Cladding Changes with Burnup/Duty Cycle.**

### **2.1.6.1 Fuel Duty Metric for In-Core Changes to Fuel**

Fuel duty is a general term that is used to describe the relative aggressiveness of operating conditions that tend to damage the fuel over time [2]. These operating conditions include heat flux, irradiation exposure, time-at-temperature, and coolant chemistry. While burn-up is one measure of the fuel condition it does not provide a distinctly defined set of characterizations. For example in determining the clad characteristics, two fuel rods may have the same burn-up but very different levels of surface oxide and clad hydrides depending on heat flux, coolant chemistry and other aspects of the fuel duty.

### **2.1.6.2 Uranium Dioxide Fuel Pellet Changes with Burnup**

Changes to the typical  $UO_2$  fuel pellet occur during operation in a power reactor. For the fuel (pellets) there is both a microstructure change and a fission product build up with increased burn-up. These changes lead to pellet cracking that also occurs with increasing burn-up. The buildup of fission products is one of the major differences between low and high burn-up fuel. The high burn-up fuel will have more fission products, more residual radioactivity and a higher potential for energy release during a dry out anneal and a high temperature during dry storage.



The fuel pellet fractures because of the thermal and internal stresses from the fission product generation. In addition to cracking, the pellet microstructure changes during the high burn up to produce a rim at the outer surface. The rim has high porosity / low thermal conductivity thus leading to the development of higher thermal gradients. As the fuel burn up proceeds more fission products are generated and swelling occurs in the pellet matrix in response to the internal gas bubble formation and coalescence. The continued accumulation of fission gasses in the bubble leads to into high pressures and additional cracking in the matrix.

The pellet matrix changes with burnup have been studied extensively [1, 3, 4, 5, and 6]. The following is a summary of the major characteristics of the changes in the fuel pellet that occur with burnup.

The conclusion of this present evaluation is that pellet aging does not represent a major concern for the fuel clad integrity (which is the primary fuel confinement) but that there is a need to have accurate data to support:

- a) the estimation of the fission gas release and pellet swelling and their impacts on clad stress during storage and
- b) the internal heat generation and its impact on fuel and clad temperatures.

Additionally, the potential release of fines from the rim region in a breached-clad fuel is a consideration that should be evaluated.

Pellet Rim Size – Source Term and Thermal Conductivity - The pellet rim size increases with burn up and is dependent on reactor operating conditions. Any increase in rim thickness increases the potential for the generation of fines and hence the source term for fuels with cladding penetrations. Additionally, the lower thermal conductivity of the rim will result in higher pellet centerline temperatures during storage but since the rim will also restrict heat transfer the clad temperature will not increase.

Pellet Rim Heat Conductivity - The pellet rim size is dependent on the burn up with higher rim thicknesses in the higher burn up fuel. The lower thermal conductivity of the rim will result in higher pellet centerline temperatures during storage but since it will restrict heat transfer to the clad it will not result in a higher clad temperature.

Pellet Rim Grain Size - The grain size in the rim is smaller than the remaining pellet. This smaller grain size needs to be included in storage evaluations of clad breach and fuel dispersion.

Fission gas release - During the fuel operation some of the fission products (FP) generated are elements that are normally in the gas phase. Most of these FPs are captured or contained within the fuel matrix and produce the pellet stress/swelling that is observed with burnup. A small percentage, nominally less than 10%, of the FP gas escapes the pellet and adds to the internal rod pressure. The internal rod pressure, acting in concert with welding and other residual stresses, produce the cladding stresses that are the drivers for age related degradation mechanisms such as creep, DHC and HE. Therefore, the contribution of fission gas release to cladding stresses needs to be well understood throughout the storage period. Measurements exist for fission gas

release during normal operation, but there is insufficient data to assess fission gas release during long term storage.

Pellet Oxidation - During normal storage conditions the pellet is encapsulated in the cladding and is not exposed to oxygen in the environment. In some off-normal conditions such as failed fuel cladding prior to storage or fuel clad breach during storage that protective barrier is lost. When fuel is exposed to air/oxygen and heat there is a thermodynamic driving force for oxidation to occur and convert the  $\text{UO}_2$  to  $\text{U}_3\text{O}_8$  (including some intermediate oxidation states). With the conversion to  $\text{U}_3\text{O}_8$  the fuel will tend to become a powder and be more of a risk for dispersal from the fuel rod. It is reported [7, 8] that higher burn up fuel is more resistant to the oxidation so high burn up fuel should not be a greater concern or risk for pellet oxidation than the low burn up fuel. In addition with a He atmosphere provided in the container during planned storage conditions, even with a cladding breach, pellet oxidation would be precluded. If air or significant levels of humidity are in the container than the risk for pellet oxidation is increased.

Fission Product Generation - The chemical composition of the fuel changes with uranium burn up because a number of elements or fission products are formed from fission and/or neutron absorption. Some of these fission products are radioactive and their presence increases the activity of the fuel. After removal from the reactor, fission product decay causes the activity (and temperature) of the fuel to decrease with time. However, the heat produced by fission product decay necessitates fuel cooling during storage and handling. The heat produced is generally sufficient to necessitate pool storage and water cooling immediately after removal from the reactor. Dry storage systems can only be used after the spent fuel has cooled for a period of time which depends on the fuel and fuel service conditions. The higher burn up fuels will have higher quantities of fission products and thus will have higher heat loads than low burn up fuels and will generally have higher temperature profiles throughout the storage period.

Source Terms for Release - The source terms of HBU fuel are expected to be significantly different than the source terms for low burn up fuels, especially for failed rods. Reference [9] provides a comprehensive description of the features of the pellet rim, fuel grain size, fission gas release (FGR) to the rod plenum, CRUD thickness, cladding oxide thickness, and other fuel parameters on the source terms for HBU fuel for cask evaluation.

### **2.1.6.3 The Differences of Low and High Burn-up Cladding**

Low burn-up fuel has been defined for dry storage as being at burn-up levels of 45 GWd/MTU and lower. High burn-up fuel is above 45 GWd/MTU and nominally up to licensed limit of about 62.5 GWd/MTU.

The following is a discussion comparing major characteristics of the cladding for low and high burn up fuel and the general conclusion is that there is not a clear property characteristic differentiation between low and high burn up fuel for long term storage evaluation. This is not the intuitive conclusion but it is supported by the fact that over time as the burn ups have increased, so have the fuel rod designs and the margins from material improvements increased. From a minor difference comparison, the higher burn

up fuel will nominally have higher internal pressures and more hydride/oxide than the equivalent fuel design with a low burn up.

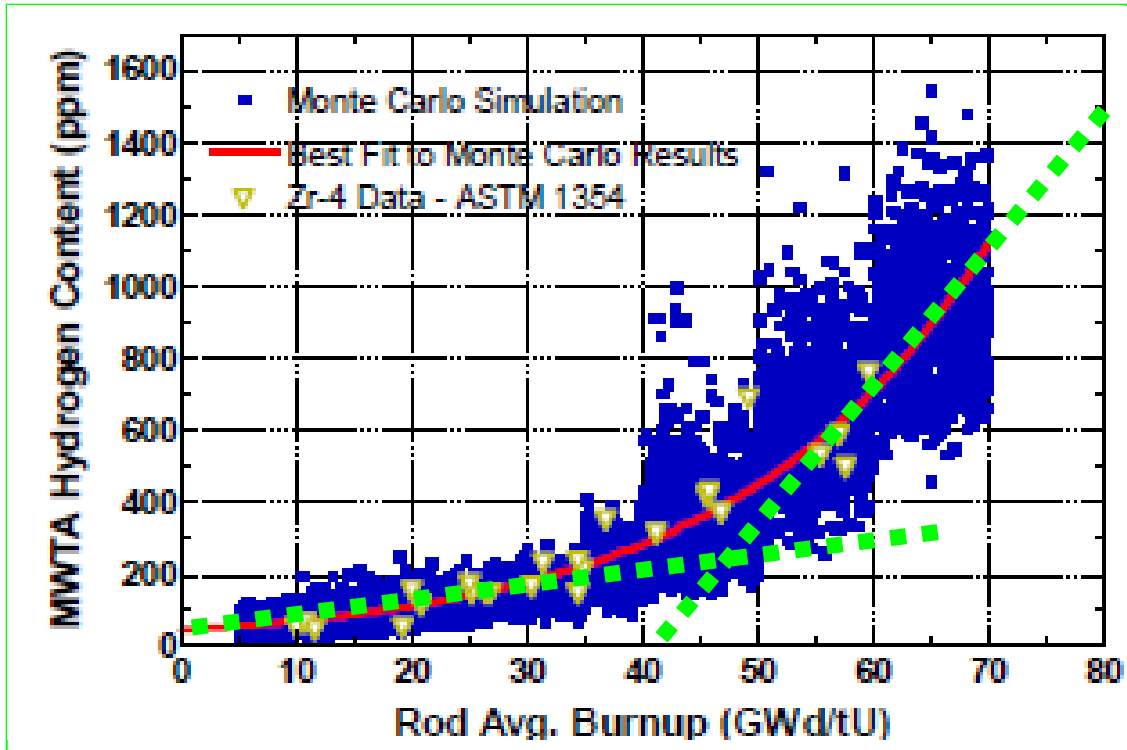
Oxide Thickness: In general it is expected that low burn up fuel will have slightly lower oxide thickness than the high burn up fuel but there is not a clear distinction. Even in a single-clad alloy system such as for low-tin Zircaloy-4 as shown in Figure 2-2 of reference 10, there is a high variability (tolerance interval from 20 to 100  $\mu\text{m}$  at 45 GWd/MT) in oxide thickness at a given fuel burn up.

The continued improvements in clad performance have resulted in the newer or improved clad versions that are more resistant to corrosion such that the newer versions may have less oxide and less hydride at 62 GWd/MTU burn-up than the older fuel rods had a 45 GWd/MTU. There is a driving force to extract as much energy from the fuel as can safely be accomplished. A general limit for both old and more recent fuel is a maximum OD oxide thickness of around 100  $\mu\text{m}$ . The early cladding would reach that limit with burn ups near 45 GWd/MTU while the improved cladding can exceed 62 GWd/MTU before that oxide thickness is reached.

Hydrogen levels: Hydrogen pick up by the cladding during irradiation occurs because the cladding alloy interacts with the water to form zirconium oxide and hydrogen. Some of the hydrogen formed is absorbed by the cladding and the amount absorbed varies with clad alloy. The amount of hydrogen absorbed in any given alloy generally increases with increasing operating temperatures and with the amount of oxide formed. Although the higher burn-up fuels typically have experienced a high duty and trend to see higher hydrogen than low burn up fuels, claddings with similar oxide thicknesses generally have similar levels of hydrogen, regardless of the fuel burn up. Some of the advanced alloys have been developed to minimize hydrogen absorption and thus, even at high burn ups, have a lower hydrogen pickup ratio (hydrogen absorbed into clad versus total hydrogen produced by clad oxidation) and lower hydrogen levels than the older low burn-up fuel [11].

Figure 8 below shows the hydrogen pickup corresponding to the oxide thickness for the low-tin Zircaloy-4 cladding shown in Figure 2-2 of reference 10. The oxide film growth and corresponding hydrogen pick up are not linear with burnup for this cladding system, but rather appear to show a trend that strongly suggests a change in mechanism for the ingress of hydrogen.

A “transition knee” in hydrogen pick-up with burnup at approximately 45 GWd/MTU is suggested using an empirical bi-linear model for hydrogen pickup. It is assumed that a microstructure change or set of changes significant to enhance clad-water interaction and hydrogen pickup may occur with burnup, although no formal model or detailed mechanistic explanation of this effect has been reported to the authors’ knowledge.



**Figure 8** Maximum Wall Thickness Average (MWTA) Hydrogen Content in Low-tin Zircaloy-4 Cladding [Figure reproduced from ref. 10]. A “transition knee” in hydrogen pick-up with burnup at approximately 45 GWd/MTU is suggested.

Hydride Microstructure in Cladding – The hydrogen in the cladding is partitioned as hydrogen in solid solution in the cladding, and, when local concentrations exceed the solubility limit, hydrides form. The formation of hydrides leads to a “hydride microstructure” in the cladding.

A brief description of the cladding microstructure in terms of hydride formation and cracking that is representative of typical hydride structures generated during reactor operation and cool-down of zircaloy cladding follows. However, these microstructures are not expected to reflect the post-dry out microstructures that develop after the hydrogen has been re-dissolved and re-precipitated.

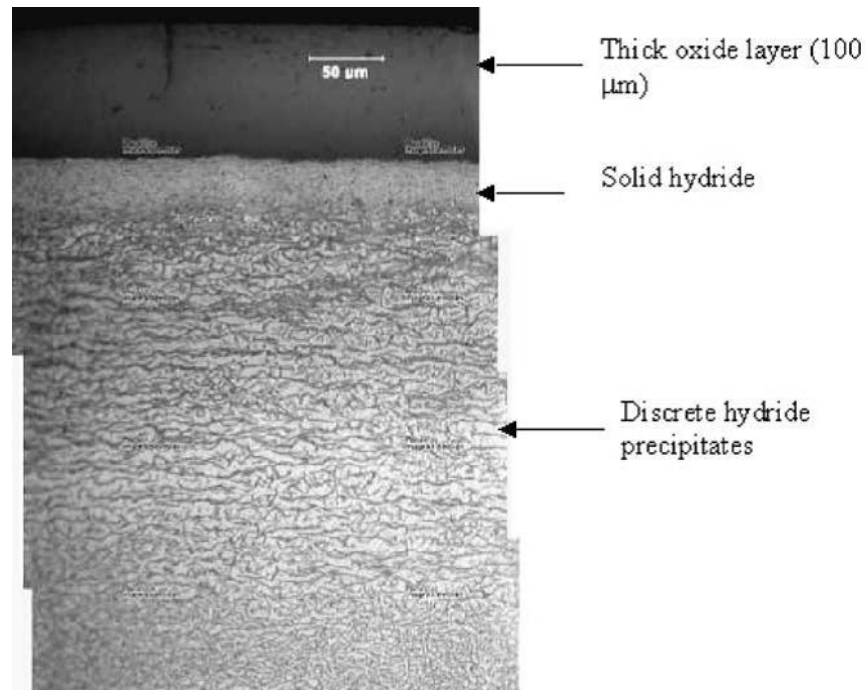
The, as irradiated, hydride microstructures that develop in the fuel clad have been grouped into three major types:

- hydride platelets;
- hydride rim; and
- hydride blisters.

*Hydride platelets:* Hydride platelets are the  $ZrH_x$  phase that precipitates as individual or linked assemblages across the clad thickness. These platelets have preferred orientations depending on the stresses, thermal gradients, grain structure, crystallographic texture and the cooling rates. During reactor operation the bulk of the clad is at a high temperature and it is assumed (at this point in our understanding/evaluation) that in most cases platelets do not exist and that the hydrogen

is in solution for most of the clad wall. However, at the clad OD where the temperature is cooler and the hydrogen solubility lower, a hydride rim may form. Upon cool down, the temperature of the clad decreases and the platelets precipitate across the clad wall.

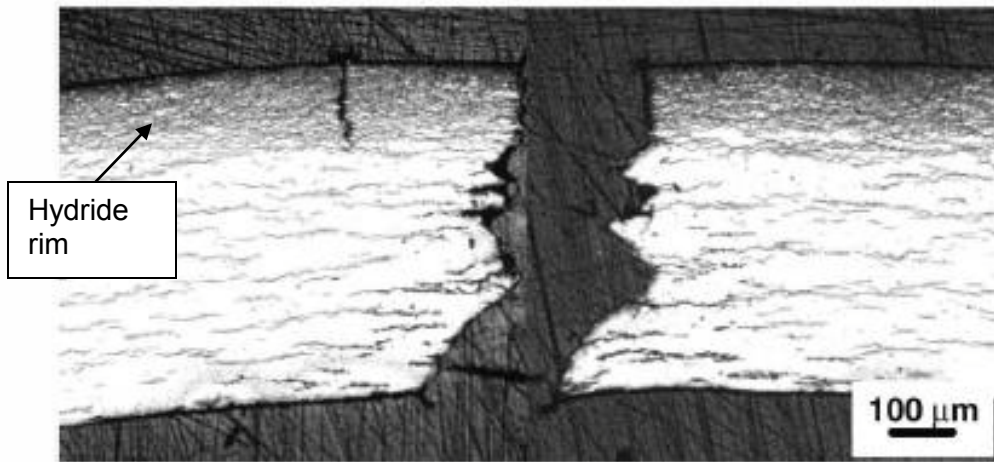
Figure 9 is an optical micrograph showing the Zircaloy-4 cladding microstructure in the clad of a fuel that experienced high burn up.



**Figure 9** A light micrograph showing a hydride layer and associated hydrides in the substrate beneath the layer in irradiated Zircaloy-4 cladding tube (average fuel burnup of 67 GWd/t and fast fluence of  $1.3 \times 10^{22}$  n/cm<sup>2</sup>) (Courtesy of R. Daum, Argonne National Laboratory). [Figure reproduced from ref. 12].

*Hydride rim:* A hydride rim is a semi-continuous hydride structure formed at the clad OD surface. It forms on the clad OD due to the thermal gradient across the clad during reactor operation which lead to an increasing hydrogen content as the OD of the clad is approached. The rim forms when this local hydrogen concentration exceeds the solubility limit for hydrogen in zirconium. Ultimately the local hydrogen content approaches that of solid ZrH<sub>2</sub> and is approximately 10,000 wppm. The rim is of relatively uniform thickness axially and circumferentially proportional to the local oxide thickness. It is thicker at some local areas such as at the pellet to pellet interface because the clad is cooler at that location.

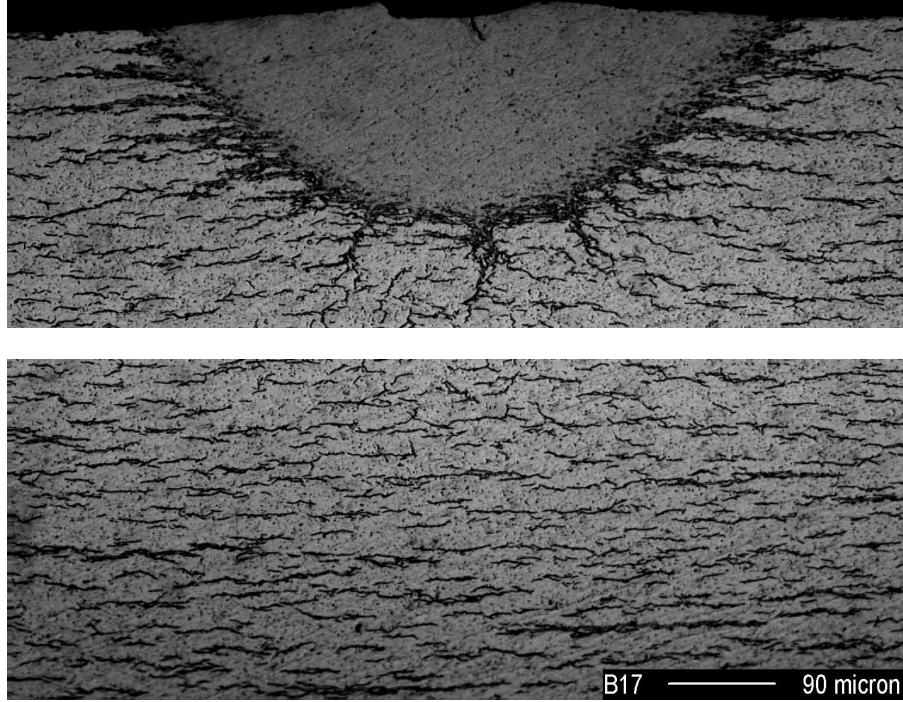
Figure 9 and Figure 10 are examples of a hydride rim condition. The sample in Figure 10 was hydrided ex-reactor to simulate the in-reactor rim formation.



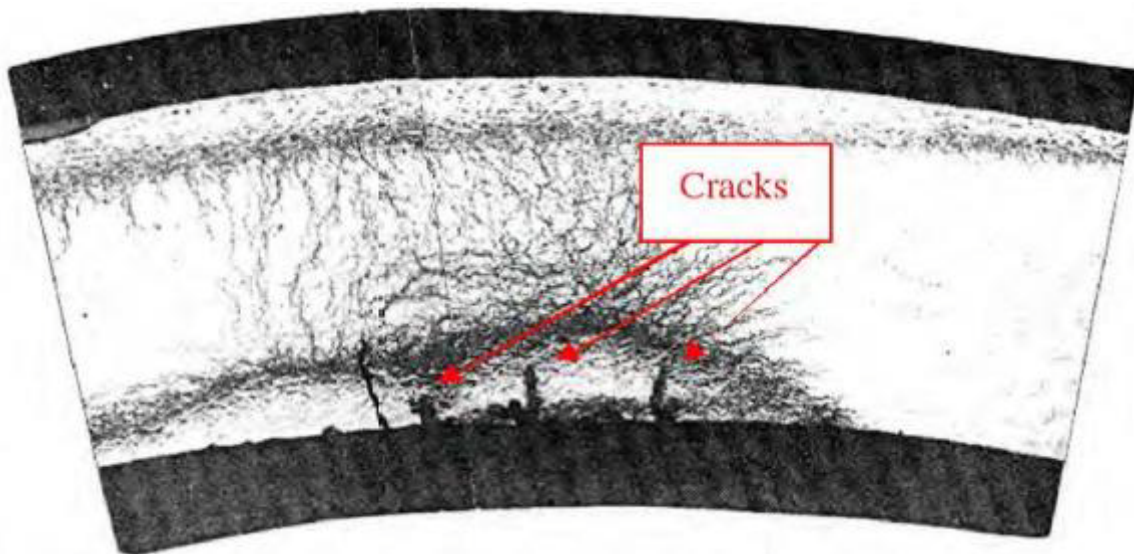
**Figure 10** Example of a hydride rim introduced in an unirradiated clad sample [Figure is reproduced from ref. 13, Fig. 5 - Wall-through crack in sample with hydride rim failed in room temperature burst test with pressurization rate of 1.9 GPa/s].

*Hydride blister:* A hydride blister is a local area of high hydride concentration. It differs from the rim in that it is a local area of significant radial depth. It can be on the OD surface if there is some local corrosion acceleration. It also is associated with spalled oxide which changes the thermal gradient across the fuel and causes a local area of the clad to have a cooler temperature.

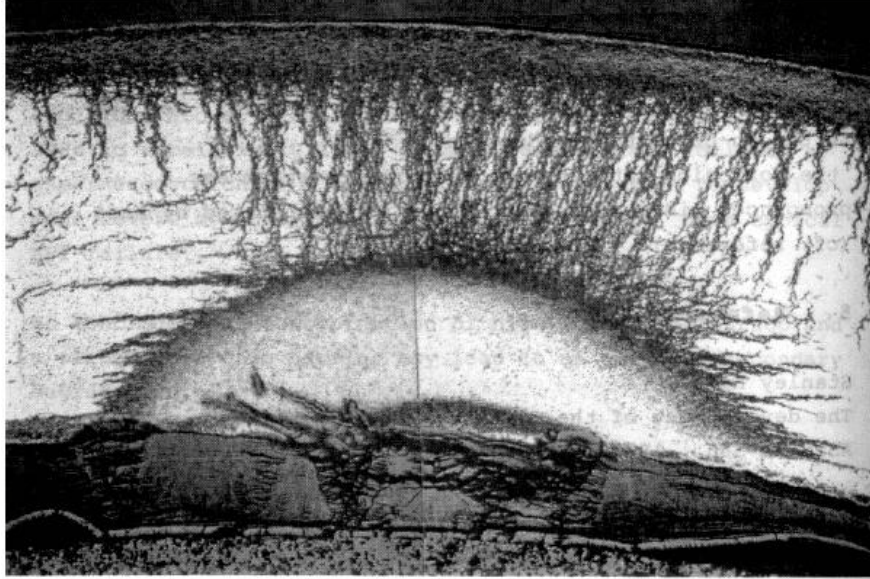
ID blisters are also observed and are associated with internal hydriding either from hydrogenous materials present in the rod or from coolant that has entered the rod due to a cladding breach. Hydride blisters introduce stresses within the matrix due to the significant volume mismatch with the Zircaloy cladding. These stresses interact with the other cladding stresses and promote additional hydride formation along the resulting stress fields. Hydride alignment along the stress fields is manifested as hydride fingers emitting from blisters as shown in Figure 11, Figure 12, and Figure 13. Note that the radial alignment of the hydrides associated with a blister on the OD of the cladding (Figure 11) and the circumferential alignment of the hydrides associated with a blister on the clad ID (Figure 13). The importance of stress during hydride precipitation can be seen in Figure 13 where the hydrides near the clad OD tend to be radial while those near the clad ID tend to be circumferential.



**Figure 11** Example of a hydride blister in an unirradiated sheet of zircaloy [Figure reproduced from ref. 14].



**Figure 12** Example of a hydride blister in a fuel cladding [Figure reproduced from ref. 15].

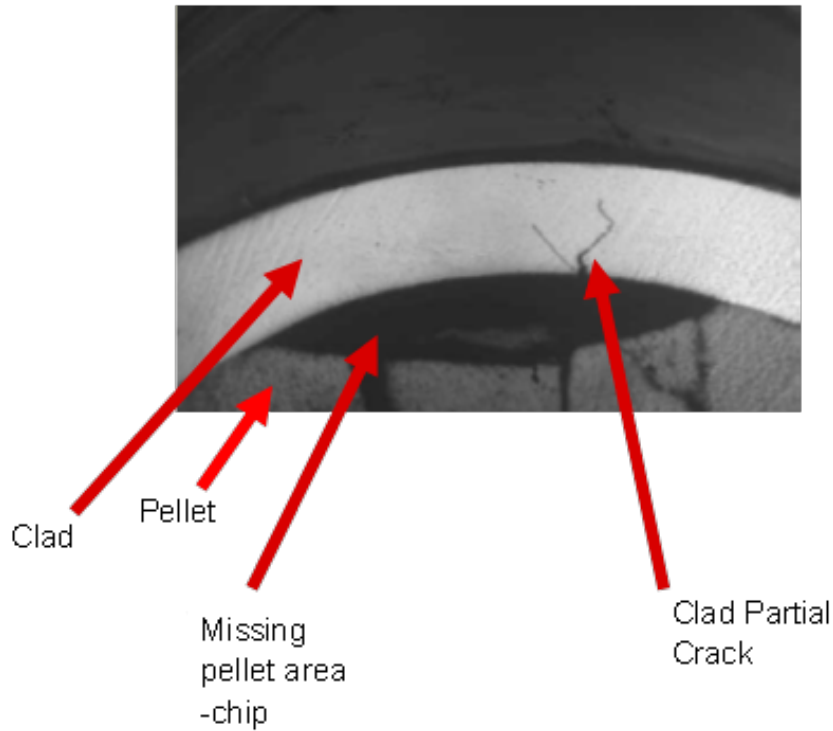


**Figure 13** Example of a hydride blister in a fuel cladding  
[Figure reproduced from ref. 16].

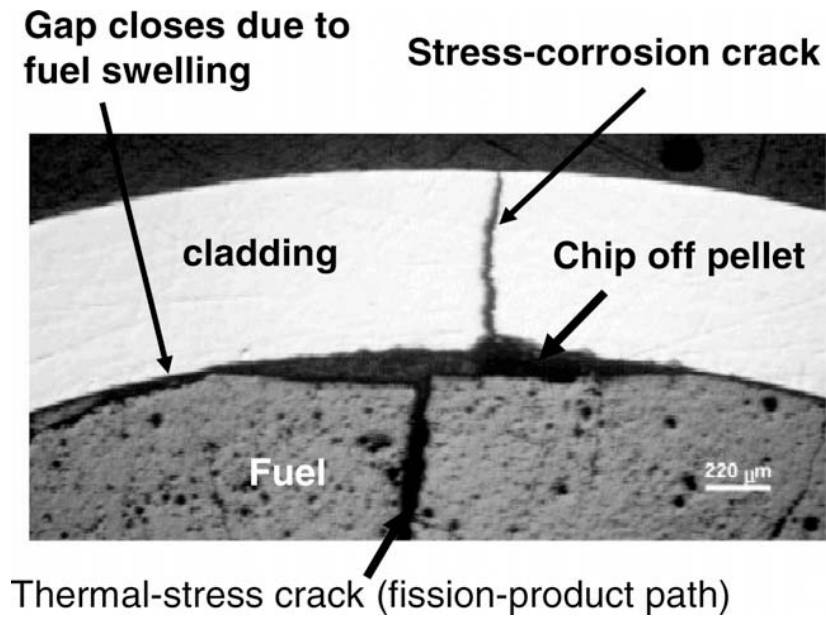
#### Clad Cracks Due to Pellet Chips -

Figure 14 and Figure 15 show radial cracks in an irradiated fuel cladding. These cracks are associated with the as-fabricated-rod missing pellet surface (pellet chip). The cracking is in the general category of Pellet Cladding Interaction (PCI) cracking and can be associated with non-typical loads and cyclic stressing in the cladding due to the lack of contact with the underlying fuel pellet. The lack of contact results from a missing chip in the fuel pellet and is the result of fabrication errors. It is suggested that the operational stresses (coolant pressure, thermal, etc.) produce cyclic stresses in the clad in the region where the clad is not supported by the pellet. Local hydriding and/or iodine-induced stress corrosion cracking may also be contributors to the cracking process. Missing pellet chips have been observed in multiple rods from different vendors. Figure 14 is an example of an ID-initiated clad wall crack that is associated with a missing pellet surface chip. Many early vintage rods will have a potential for the presence of such chips and an associated partial wall crack. Such cracks will concentrate the stresses in the clad wall and may serve as sites for hydrogen induced clad degradation during EST.





**Figure 14** Example of ID crack in fuel cladding [Figure reproduced from ref. 17].



**Figure 15** Example of ID crack in fuel cladding [Figure reproduced from ref. 18].

### Irradiation Hardening and Microstructure Changes

The neutron fluence involved in fuel burn up cause displacement of atoms in the clad matrix and produce damage or a material hardening. This irradiation damage increases with as the fuel burn up increases thus the strength of the clad increases and the ductility decreases with burn up. However, this effect is not linear with the neutron fluence and a near saturation of the irradiation hardening effects is reached typically during the first year of fuel operation [19]. Thus, there is essentially no significant difference in irradiation related clad strength or ductility between low and high burn up fuel.

There are, however, changes in clad and fuel micro structure that occur during irradiation and become more apparent as the fluence increases. Precipitate structures may develop in the cladding and the dislocation substructure is altered. Additionally, the clad density will change with fluence and the magnitude of the change is related to burn-up. The drying operations required to prepare the fuel for dry storage will then alter the irradiation induced changes in microstructure and cladding properties.

### Internal rod pressure, pellet swelling and clad stress

The clad stress states and levels are from a combination of loadings. The stresses are generated by the combination of pellet/cladding interactions, internal gas pressures, the residual stresses resulting from fuel fabrication and handling and the hydrostatic pressure exerted by the cooling water. Storage temperature and fission gas release influence the rod internal pressure and reactor operating conditions influence the free volume inside the fuel rod. Typically, the higher burn up rods will have higher internal pressures due to the combined effects of clad creep down, pellet swelling and fission gas release. Internal gas release from burnable absorbers will occur early in the life of the fuel and should not be significantly different for high burn up versus low burn up fuel. Taken together, the above factors combine to maximize the cladding stresses when the rod temperatures are the highest and before any post irradiation creep has occurred. Although typical cladding stresses decrease as the storage time increases, stresses are anticipated to remain the highest in the high burn up fuels thus making these fuels more susceptible to most operative age-related degradation processes. However, the ultimate susceptibility to degradation is the product of the combined effects of available hydrogen content and hydride characteristics, applied and residual stresses, cladding characteristics, temperature and temperature gradients, time and irradiation.

#### **2.1.6.4 Fuel Drying and Impact on Fuel Initial Condition**

The placement of spent fuel in a dry storage requires removal from pool storage and drying before placement in the DCSS. Drying is necessary fuel to prevent corrosion induced degradation of the fuel and/or canister and radiolytic reactions with moisture that can cause additional canister pressurization. However, the heating of the fuel to the anticipated drying temperature may alter the cladding microstructure by: 1) partially annealing of the microstructural changes induced by irradiation and 2) causing hydride re-resolution during heating and re-precipitation on cooling.

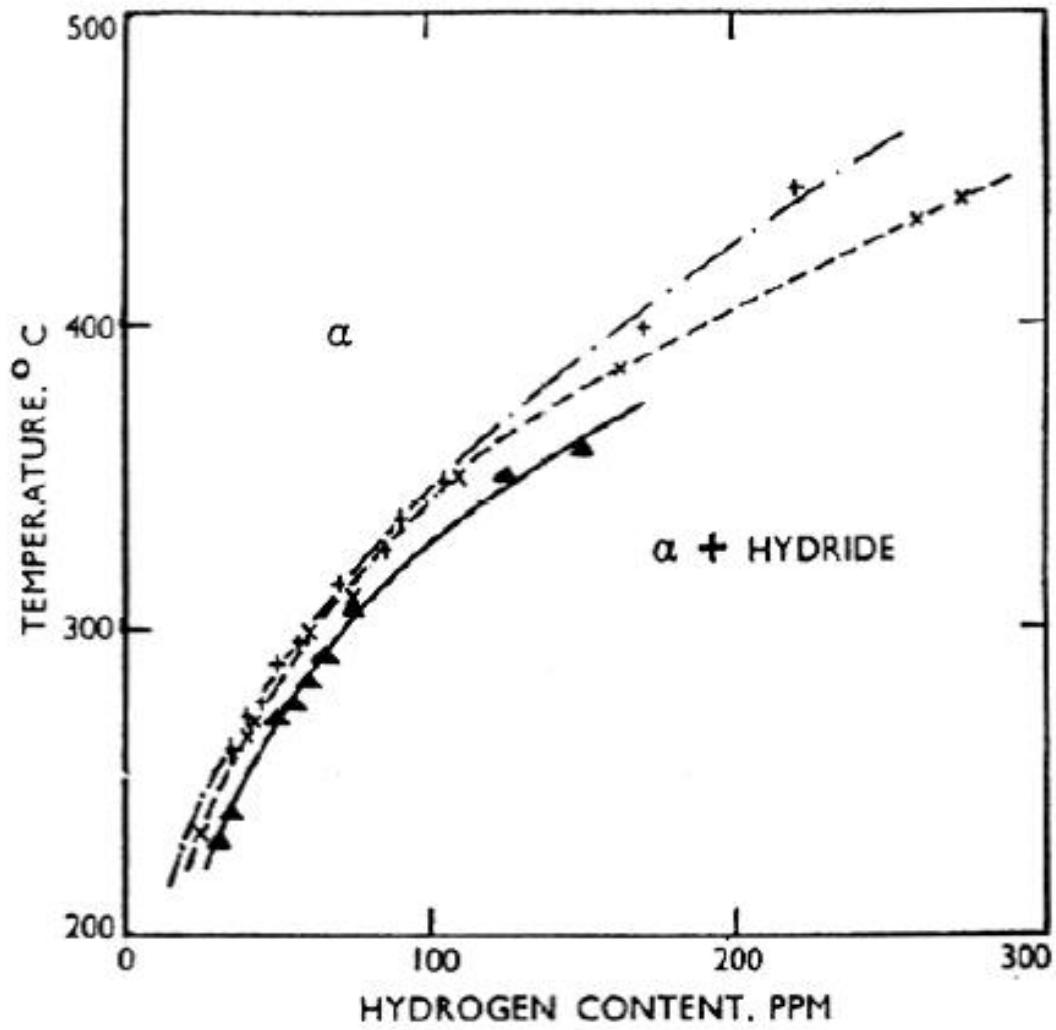
A national consensus standard for drying spent nuclear fuel [20] summarizes the practices for removal of water from fuel systems but does not provide sufficient justification for dryness of water logged rods or the resultant reconfiguration of the

hydride microstructures. It is important to consider the potential for hydrogen redistribution and hydride reorientation in the cladding during the drying process because of the potential for hydrogen embrittlement processes during extended storage. Zirconium is an "exothermic occluder" of hydrogen, which means that hydrogen is more stable in solution in the zirconium matrix than as hydrogen gas and therefore is not removed from the zirconium alloys in a drying process.

Two different solubilities for hydrogen in zirconium are often referred to in the literature; namely the maximum solubility, and the terminal solid solubility (TSS). The maximum solubility is the total or maximum amount of hydrogen that can be absorbed under the exposure conditions (temperature and hydrogen pressure) and may involve the formation of zirconium hydride. The TSS is the amount of hydrogen that can be in solution in the zirconium matrix without the precipitation of a hydride phase. Reference [21] provides a treatise on the hydriding of zirconium alloys.

Figure 16 and Figure 17 below, reproduced from reference [21], show that hydrogen maximum solubility and TSS, respectively for zirconium cladding alloys. Reference [21] summarizes additional literature references showing dependency of TSS on alloy additions, fabrication, and irradiation.

Using the data and data trend from Figure 16, it is seen that the hydrogen solubility is about 100 ppm at 300°C and 200 ppm at 400°C. A high burn up fuel with an assumed clad content of 500 ppm H, including that in the rim, could dissolve up to 200 ppm during a dry out anneal at 400°C. The hydrogen would then precipitate as a hydride as the fuel cools. A time delay for the precipitation process would be expected due to reaction kinetics and the "final" hydride morphology may not develop until the fuel experiences EST.



**Figure 16** Hydrogen (H) solubility for zirconium alloys: (▲) = Zr-2; (×) = Zr-4; and (+) = Zr<sub>2.5</sub>Nb [Figure reproduced from ref. 21].

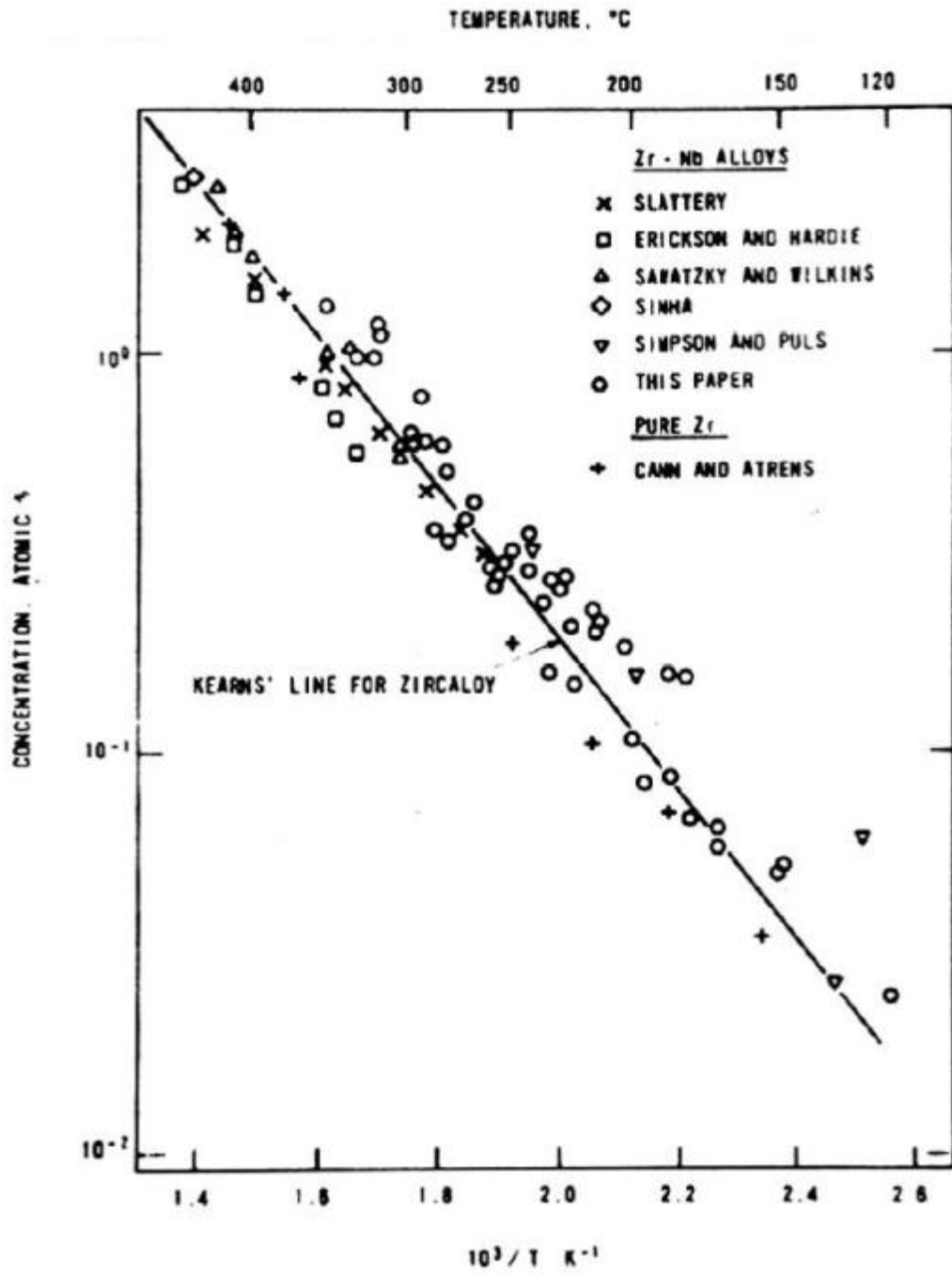


Figure 17 TSS for Pure Zirconium [Figure reproduced from ref. 21].

### **2.1.6.5 Fuel Initial Condition - Fuel Drying Data Needs and Recommendation**

The ASTM standard guide [20] for drying spent fuel describes drying considerations, and lists the standard protocol for drying commercial spent nuclear fuel. The standard provides sufficient details to evaluate remaining free water in systems that do not have major impediments to drying, but it does not provide a quantitative approach to achieve dry-out of water-logged rods. The water from incomplete drying could lead to additional fuel corrosion and/or radiolysis of the water and hydrogen buildup inside the canister. The radiolytically produced hydrogen could be gettered by the cladding and add to its hydrogen content. The contribution of hydrogen from water radiolysis has been discounted [22]; however a parametric evaluation of residual water in a canister free volume and potential hydride microstructure produced during cool-down over EST should be considered.

Most importantly, the present standard guide does not address a thermal drying treatment to minimize the potential for hydride reorientation from the predominantly circumferentially-oriented hydrides at reactor discharge to radially-oriented hydrides after drying. Changes to the fuel initial condition during dry-out can occur and are dependent on the thermal cycles used for drying. These changes may include some recovery of radiation damage and creep, and may alter the hydride microstructure.

1. **Testing & Analysis Activity:** The following topics should be addressed in a revision to the existing ASTM drying standard or in a new drying standard guide. These topics should include explicit methods or controls (e.g., maximum cladding temperatures and appropriate cool-down rates to avoid radial hydrides), to assure water removal and prevent significant hydrogen induced degradation and should be applicable to low and high burn up fuel:
  - Dryout of water-logged rods
  - Hydride microstructure reconfiguration
  - Radiolysis of water with equilibrium back reactions and including gettering of H<sub>2</sub> by fuel components
  - Annealing of radiation damage
  - Creep during dryout

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### **2.1.7 Reference Fuel Assembly**

The fuel assemblies placed in dry storage will be quite variable in terms of their physical characteristics. Typical variables include the fuel cladding alloy and fabrication technique, the fuel duty cycle, time since removal from the reactor, the presence or absence of pinholes or hairline cracks in the cladding, drying times and temperatures and other processing and service variables. These and other fuel assembly variables will alter the response of the fuel and cladding to any given set of dry storage conditions. The anticipated behavior of any given fuel may be very difficult to accurately establish because of the lack of directly applicable fuel/cladding response data. For example, the magnitude of the fuel cladding changes illustrated in Figure 7 will depend on fuel duty cycle and cladding alloy as well as other fabrication and service variables. Fuel cladding alloys progressively evolved to address specific economic and/or service problems (see Sections 2.1.1 and 2.1.2). Therefore, a single fuel cladding alloy was selected, by engineering judgment, to represent the variety of alloys that may be placed in dry storage. However, the evaluation also recognized that the lack of data on the alloy-to-alloy variation in behavior represents a significant gap in the data necessary for accurate estimations of the response of the cladding to the very long term dry storage environment.

The attributes of the single fuel cladding selected as a representative of a reference fuel assembly for consideration of degradation during EST are:

Cladding Material: Standard Zircaloy-4 or Standard Zircaloy-2 (random grain texture, susceptible to hydride reorientation during thermal cycles and during long term exposure).

Oxide Thickness (ZrO<sub>2</sub>): 100 μm including some spalled oxide in local areas.

Hydrogen Content: 600 ppm incorporated in a locally hydrided microstructure that contains predominantly circumferential hydrides.



Cladding Stress (from internal gas at RT): 75 MPa (includes impact of wall loss due to corrosion wastage)

CRUD: 25  $\mu\text{m}$  (low burnup); 100  $\mu\text{m}$  (high burnup). Note that the impact of CRUD due to the predominant radioisotope, Co-60, ( $T_{1/2}$  of 5.5 years) would be expected to be greatly lessened for EST.

Fuel Condition: Fuel in fragments including fines in rim at the cladding/fuel interface.

It is also noted that there are some incipient conditions from both the fabrication (small laps in the clad, pellet chips, weld contamination, etc.) and reactor operation (vibration wear, non-breaching clad cracks, hydride blisters, etc.) that may be present on the fuel.

The detailed descriptions of the physical and material conditions at the beginning of dry storage illustrate two important features:

- 1) plots of fuel characteristics, including rod internal pressure, oxide thickness and hydrogen content as a function of rod average burn-up show considerable data scatter (for example hydrogen content of 400 ppm is found in rods with 35 to 60 GWd/MTU burn-up), and
- 2) internal pressure, oxide thickness and hydrogen content generally increase with increasing burn-up, however, there is no discontinuity in the increase that categorically separates high burn-up from low burn-up conditions.

The data scatter shown in the reference [1] coupled with the lack of a discontinuous, burn-up induced transition in material condition is illustrated by the representative fuel cladding which, according to the specified oxide thickness, could have experienced a burn-up between about 40 and 60 GWd/MTU. The anticipated average hydrogen content in a cladding at these burn-up levels could vary from a low of about 100 ppm to a high of about 600+ ppm while the rod internal pressure could vary from 2.5 to 7.5 MPa. These variations in initial conditions could lead to large variations in the response of the cladding to the dry storage environment.

A discussion of the impact on fuel radiological source terms with burn-up that impact confinement is provided in reference 2.

### **Section 2.1.7 References**

- [1] Electric Power Research Institute, EPRI Synthetics Report 1015048, "Spent Fuel Transportation Applications – Assessment of Cladding Performance", December 2007.
- [2] Einziger, R.E. and Beyer, C., Characteristics and Behavior of High-Burnup Fuel that may Affect the Source Terms for Cask Accidents, Nuclear Technology, Vol. 159, August 2007, pp. 134-146.

## **2.2 Fuel Degradation Phenomena**

There are three basic type processes that control age-related fuel degradation of fuel materials and components:

- 1) processes that involve the diffusion of point defects and defect complexes including dislocation movement in the material or component,
- 2) processes that involve the diffusion of specie atoms, especially hydrogen, and,
- 3) processes that involve chemical interaction among atoms within the materials and/or within the storage environment.

These processes are time-dependent, thermally-activated phenomena that are markedly influenced by alloy chemistry, microstructure, thermal-mechanical history, crystallographic texture, irradiation and other variables that depend on component fabrication and use as well as service and storage conditions. For example, much of sections 2.2.x discuss the effect of zirconium alloy chemistry on material strength, corrosion resistance, in-reactor behavior and other performance measures. Such alloy-to-alloy difference in performance translates directly into differences in the response to age-related degradation processes as shown in those same sections.

In spite of such well-established differences in alloy performance, there is a tendency to use long-term data developed for Alloy “A” to represent the anticipated behavior of Alloy B. For example, reference [1] states that “in creep tests at 250-400°C of Zircaloy cladding irradiated up to burn-up of 64 GWd/MTU, no failures have been observed below 2% strain – therefore a conservative strain limit of one percent has been used...” Similar statements that lump zirconium alloys as a single material system are frequently made, and while true to a very limited extent, alloy-to-alloy variations and burn-up related differences in susceptibility to age-related degradation processes are expected.

The following subsections under section 2.2 discuss the fuel degradation processes for zirconium cladding alloys from published information.

### **General Fuel Data Needs for Fuel in EST; General Recommendation for**

**Resolution:** A wide variety of alloys are, and have been, used for fuel cladding in the U.S. commercial fleet and the response of these alloys to EST will depend on chemistry, thermal mechanical process history, irradiation history and other fabrication/service variables. The information required to address the response of a specific cladding to a specific age-related degradation process is limited and in many cases simply not available. It is recommended that information generated (e.g., fracture toughness of newer cladding alloys with a hydride microstructure) should be explicitly compared to available data and improvements in the basis for constitutive model(s) for alloy performance should be made. Models should then be developed that strive to encompass the spent fuel inventory and these models should be assessed to validate their extrapolation to EST conditions.

**Fuel Condition Data Needs for Storage Retrievability and for Source Term for Transportation; General Recommendation for Resolution:** An important consideration for the stored fuel is the potential release of particulates, especially in evaluation of minor breaches that may occur in storage and impact retrievability during EST. Additionally, failures that may occur during EST should be assessed and the impact of such failures on confinement in transportation should be determined. As discussed, reference [2] provides a comprehensive description of the features of the pellet rim, fuel grain size, fission gas release (FGR) to the rod plenum, CRUD thickness, cladding oxide thickness, and other fuel parameters on the gas, volatile, fines, and CRUD source terms of HBU fuel for cask evaluation. Many data needs related to these source terms were identified and are listed in Table II of reference 2. This present evaluation concurs with these needs. These needs for EST would best be evaluated in a destructive examination of HBU fuel after an extended exposure to dry-storage conditions. This examination could be performed in conjunction with the cask demonstration project described in section 3 of this report.

## **Section 2.2 References**

- [1] Electric Power Research Institute, report 1003416, "Technical Bases for Extended Dry Storage of Spent Nuclear Fuel" December 2002.
- [2] Einziger, R.E. and Beyer, C., Characteristics and Behavior of High-Burnup Fuel That May Affect the Source Terms for Cask Accidents, Nuclear Technology, Vol. 159, August 2007, pp. 134-146.

### **2.2.1 Creep**

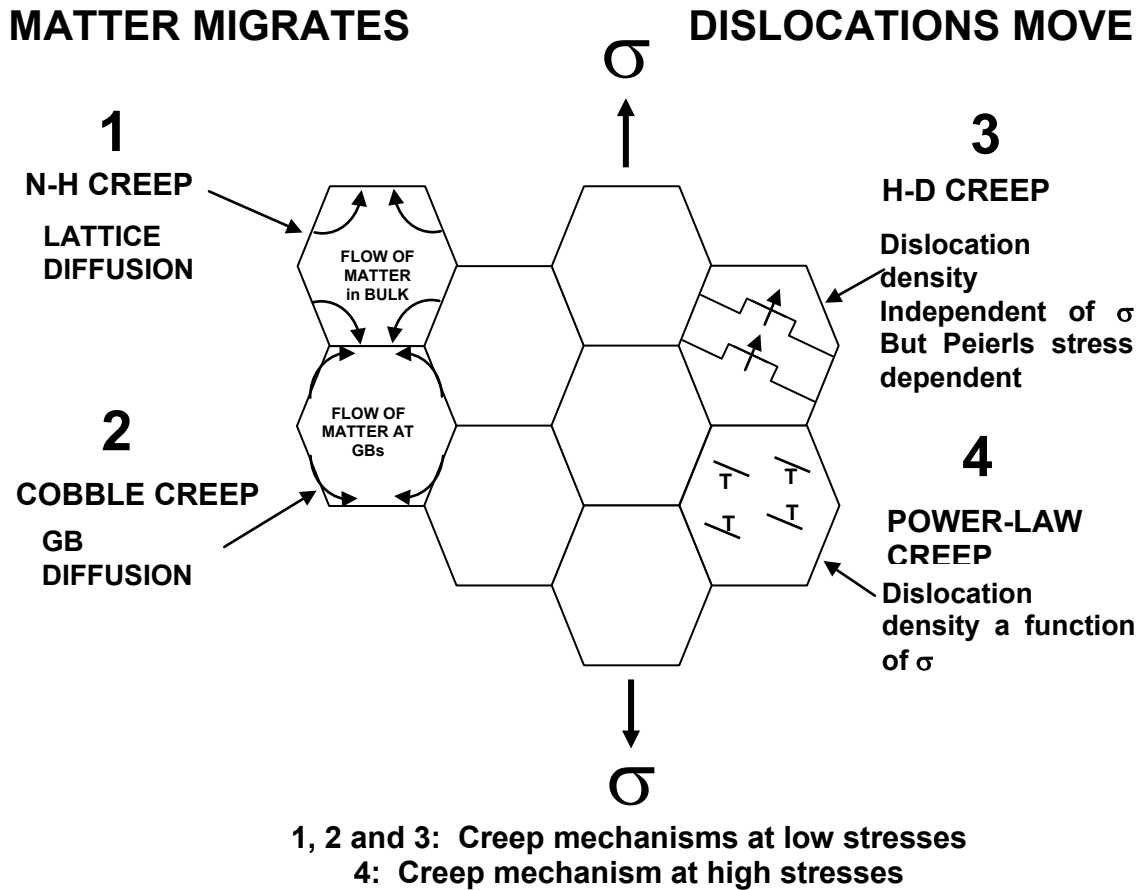
Creep is time dependent plastic deformation that is generally attributed to stress-directed, diffusional processes. The activation energy for diffusion is dependent on the path for vacancy and atom migration. The activation energy for grain boundary diffusion is typically less than the activation energy for lattice diffusion and greater than that for diffusion along an external surface. The variations in the activation energies for diffusion and the relative volumes of the diffusion pathways that exist on a polycrystalline alloy such as the zirconium based cladding alloys frequently causes creep to be divided into several distinguishable but related processes.

Several of the potential creep mechanisms and the associated diffusion pathways are illustrated in Figure 18. Coble creep, for example, involves grain boundary diffusion while lattice diffusion controls the power law creep process. The strain that is associated with any creep process is the sum of the strains caused by the operative creep mechanisms. Models to estimate creep strains or strain rates typically involve the diffusivity of base metal atoms in the lattice,  $D$ , temperature,  $T$ , stress,  $\sigma$ , grain size,  $d$ , material constants such as the elastic modulus,  $E$ , the Burger's vector of dislocations,  $b$ , and constants that are dependent on the operating creep mechanism. For example, the creep strain rate may be expressed as:

$$\dot{\epsilon} = A(DEb/kT)(\sigma/E)^n(b/d)^p \quad [1]$$

where  $A$ ,  $n$  and  $p$  are constants. The constant  $n$  is termed the stress exponent and  $p$  is the inverse grain size exponent. More elaborate creep models are available but these

models also suffer from the weaknesses described and should not be extrapolated into time frames that extend far beyond the data used to develop the model. That is, the use of such an equation to extrapolate short term data into times relevant to long term storage of spent fuels is tenuous at best especially when alloy-to-alloy variations in material behavior are considered. Small changes in alloy composition can cause significant changes in creep rates. Microstructure and heat treatment also have a significant impact on the creep rates of any given alloy as shown in Table 2.



**Figure 18** Creep Mechanisms by atom or defect complex migration (left) and dislocation formation/movement mechanisms (right).

**Table 2** Influences of Heat Treatment on Creep of Zr-2.5Nb Alloy at 545°C (818K) [1]

Specimen	Heat Treatment	Test Temperature	Stress (MPa)	Creep Rate (s <sup>-1</sup> )
A	650°C/3hr, FC	545°C	1.5	8.5x10 <sup>-9</sup>
B	650°C/3hr, FC + Aged at 550°C/10 days	545°C	1.5	2.0x10 <sup>-7</sup>
C	745°C/75hr, FC	545°C	1.5	2.0x10 <sup>-9</sup>

Table 3 lists predicted creep strains, from a creep model benchmarked to short-term pressurized tube testing, for time and temperature conditions that span EST. The creep strain results clearly indicate that most of the strain has occurred within a 50-year period corresponding to the high temperature conditions of fuel storage.

**Table 3** Predicted Hoop Strains of Nb-Modified Zirconium Cladding Alloy Under Extended Dry Storage Conditions [Table reproduced from ref. 2].

**Table 3**

Predicted hoop strains of Nb-Zry clad under dry-storage conditions

Time (year)	T (°C)	Coble (%)	Dislocation (%)	Total strain (%)	
				Time-hardening	Strain-hardening
0.02	295.6	1.66 x 10 <sup>-4</sup>	5.97 x 10 <sup>-6</sup>	1.72 x 10 <sup>-4</sup>	1.41 x 10 <sup>-4</sup>
1.19	330.4	3.12 x 10 <sup>-2</sup>	2.72 x 10 <sup>-3</sup>	3.39 x 10 <sup>-2</sup>	3.40 x 10 <sup>-2</sup>
5.43	330.1	0.160	7.41 x 10 <sup>-3</sup>	0.167	0.180
10.94	321.7	0.294	1.00 x 10 <sup>-2</sup>	0.304	0.338
25.18	298.0	0.483	1.27 x 10 <sup>-2</sup>	0.496	0.559
51.31	265.2	0.598	1.39 x 10 <sup>-2</sup>	0.612	0.696
101.80	222.0	0.638	1.41 x 10 <sup>-2</sup>	0.652	0.750
200.50	193.5	0.650	1.41 x 10 <sup>-2</sup>	0.664	0.767
250.90	186.8	0.652	1.41 x 10 <sup>-2</sup>	0.666	0.770
540.80	168.7	0.658	1.41 x 10 <sup>-2</sup>	0.672	0.777
769.30	159.6	0.660	1.41 x 10 <sup>-2</sup>	0.674	0.779
993.70	153.5	0.661	1.41 x 10 <sup>-2</sup>	0.675	0.780

The pressure loading of the pin-design fuel is the primary driving load for cladding creep. Creep can be significant at high temperatures and/or over long time periods at moderate temperatures [3].

The initial cladding stresses are dependent on burnup. The fission gas inventory in a fuel rod increases with burn-up, while the fuel thermal conductivity decreases and changes of fuel microstructure reduce the ability of the fuel to retain the fission gas. This, together with the rod void volume reduction caused by cladding creep-down (in reactor) and fuel swelling, increases the potential for initial high rod internal pressures with increasing burn-up [3].

**Assessment of Creep Degradation under EST** - Creep-induced degradation of the spent fuel under EST conditions is not anticipated because of the lower cladding stresses due to reduced internal pressure with time, and the limited ability of substitutional atom diffusion at the temperatures anticipated (< 200°C) during extended dry storage.

Furthermore, creep of the cladding in a dry storage system would be self-limiting as the driving force decreases with dimensional changes. For example assuming typical spent fuel rod conditions (ID – 0.83mm, void volume -15 cc) a 1% diametrical creep will produce about a 15% increase in rod internal volume and an equivalent 15% reduction in stress; thus, reducing the creep rate. The internal volume increase with creep and the internal pressure reduction with cooling work to reduce clad stress and hence creep rate.

Should creep rupture occur, the cladding breach is expected to be limited in extent, and would result in gas and volatile specie release, and some small fraction of fines from the rod due to the much reduced internal pressure driving force with time. Axial splitting along the rod length is a beyond unlikely occurrence.

**Creep Degradation Data Needs for EST and General Recommendations for Resolution** - The present temperature limit, applicable to all Zircaloy fuel cladding types for dry storage, is 400°C, and is based on limiting the creep strain total deformation to 1%. This limit is selected to avoid creep rupture that could occur at 2% strain [4, 5]. Primarily fuel at low burnup was used for this determination [citations in 5]. HBU fuel would have higher internal gas pressure and higher cladding stresses at this temperature by virtue of additional gas release to rod with burnup [6].

Other cladding phenomena, primarily hydride reorientation (temperatures affect hydride solubilities and strains affect reorientations) should be considered in creep evaluation and modeling to enable a more accurate prediction of spent fuel performance in very long storage conditions. Specifically,

1. Testing & Analysis Activity: Verification that the creep models, and failure strain limits presently assumed for all zirconium cladding alloys are applicable for new and alloys and for HBU conditions.
2. Testing & Analysis Activity: Consideration of hydrogen effects on creep rate models. Claddings in HBU fuel may contain more hydrogen than LBU fuel claddings.
3. Testing & Analysis Activity: Consideration of higher pressure loading from HBU fuel on creep throughout storage period.
4. Cask Demonstration Activity: As part of a cask demonstration, creep data should be collected on HBU fuel and on new cladding alloys.

These recommended activities are to improve predictive accuracy especially for new alloys and at HBU conditions. Temperature limits for dry storage under short-term (< 60 years at present) are sufficient to protect against creep, contingent upon DCSS thermal management in dry storage to enable credit of low (e.g., < 400°C and falling) temperatures in the EST period.

The probable significance of creep to EST is low considering: 1) that creep rates at the start of EST after 120 years is expected to be low due to very low fuel temperatures (< 200°C); 2) creep is self-limiting in a rod under internal pressure; and 3) any fuel rod breach that would occur due to creep would be a minor breach with a small release within the confinement provided by the canister of the DCSS.

### **Section 2.2.1 References**

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- [6] Manzel, R. and Walker, C.T., “High Burnup Fuel Microstructure and Its Effect on Fuel Rod Performance,” Proceeding of the ANS International Topical Meeting. Light Water Fuel Performance, Park City, Utah, April 2000, American Nuclear Society (2000).

### **2.2.2 Hydrogen-Induced Degradation Processes - Overview**

Hydrogen induced degradation is a generic term used to designate a number of processes or mechanisms by which the load carrying capabilities of metals and alloys are reduced by the presence of hydrogen [1]. Often the hydrogen acts in combination with applied and/or residual stresses on the material to produce the degradation process. The hydrogen may be present in the external environment, absorbed in the metal, precipitated as a second phase or adsorbed on the metal surface. Hydrogen found in the cladding of spent fuels is generated by in-reactor corrosion of the fuel cladding and its ingress increases the hydrogen content in the cladding alloy. The hydrogen induced degradation mechanisms generally considered applicable to the zirconium based alloys in spent nuclear fuel assemblies involve the precipitation of hydride phases and include [2, 3]:

- 1) Delayed Hydride Cracking (DHC),
- 2) Hydrogen Embrittlement (HE) as pertains to reduces tensile ductility, and
- 3) Hydrogen Embrittlement (HE) as pertains to hydrogen/hydride induced shifts in the ductile-to-brittle transformation temperature.

Each of these degradation mechanisms may involve hydride reorientation and hydrogen migration which have occasionally been considered degradation mechanisms [4] but, in reality, are enablers of one of the three mechanisms listed above.

This report demonstrates that each of the three degradation mechanisms listed may cause degradation of zirconium based fuel cladding and assembly components during extended storage and transportation and that such degradation has the potential to impact the safety functions of the DCSS. Because of this potential, a brief background on hydrogen-in-zirconium alloys is presented.

**Background for Hydrogen Effects on Zirconium Alloys** - The adverse effects of hydrogen on the mechanical behavior of zirconium based nuclear fuel cladding alloys are well established and are generally attributed to the transgranular and/or intergranular precipitation of hydrides [5].

Hydrogen solubility is temperature dependent and at temperatures below approximately 100°C, hydrogen is virtually insoluble as illustrated in Figure 19. This lack of hydrogen solubility at 100°C or below contrasts the approximately 200 ppm hydrogen that is soluble at PWR operating temperatures and at the 400°C maximum fuel drying temperature (see Figure 16). The decrease in hydrogen solubility as the cladding cools during dry storage, coupled with the fact that hydrogen was absorbed by the fuel cladding during reactor service provides the opportunity for hydride precipitation during cooling.

Hydrogen remains mobile in many metals, including zirconium alloys, at room temperature, thus hydride precipitation in zirconium alloys may occur throughout the extended storage period. The temperature dependence of hydrogen diffusivity in alpha phase zirconium is [5]

$$D = 2.17 \times 10^{-3} \exp(-8380/RT) \text{ cm}^2 \text{ sec}^{-1} \quad [2]$$

Evaluation of the above equation at  $T = 300 \text{ K}$  (approximately room temperature) gives a value for diffusivity of  $1.7 \times 10^{-9} \text{ cm}^2 \text{ sec}^{-1}$ . The distance that atoms can diffuse,  $x$ , during a time,  $t$ , can be broadly approximated [6] by

$$x = (2Dt)^{1/2} \text{ cm} \quad [3]$$

Thus, even at room temperature, hydrogen in zirconium alloys may migrate, by random walk diffusion, a distance of over 0.05 mm per day. The driving forces for hydrogen migration include concentration, thermal and stress gradients, all of which will exist in spent fuel cladding. Hydrogen will diffuse down a concentration or temperature gradient but up a stress gradient [5]. Therefore, there is tendency for hydrogen to migrate up a stress gradient and concentrate in regions of lattice dilation on the cold side of the fuel cladding [7]. These tendencies can increase the hydrogen flux into regions of lattice dilation and toward colder areas. The influence of the various gradients on hydrogen flux,  $J$ , is given by [5]

$$J = -D(\Delta C) + D(Q^*C/RT^2) \Delta T + \exp(C_2 \Delta s) \quad [4]$$



where  $D$  is the diffusivity,  $Q^*$  is the heat of transport (approximately  $6 \text{ kcal mol}^{-1}$ ),  $C$  is the local hydrogen concentration,  $\Delta C$  is the concentration gradient,  $\Delta T$  is the thermal gradient,  $\Delta \sigma$  is the stress gradient and  $C_1$  and  $C_2$  are constants.

Hydrogen, in solid solution, is a screened proton, occupies interstitial sites in the metal lattice and has a positive partial molar volume. The lattice strains caused by hydrogen absorption interact with other lattice strain such as those produced by imperfections or by stresses acting on the metal [7]. For example, hydrogen will concentrate on the tensile side of an edge dislocation and in the hydrostatic stress field ahead of a sharp crack tip.

There is a tendency for the hydride platelets to develop a preferred orientation during the nucleation and/or growth processes [8, 9]. This preferred orientation depends on the fabrication history of the cladding alloy, post-fabrication thermal-mechanical processing, existing temperature differentials and stress fields, and other material/environmental variables. The typical preferred orientation results from a combination of texture effects, temperature differentials and the tendency of platelets to align parallel to a compressive stress and perpendicular to a tensile stress.

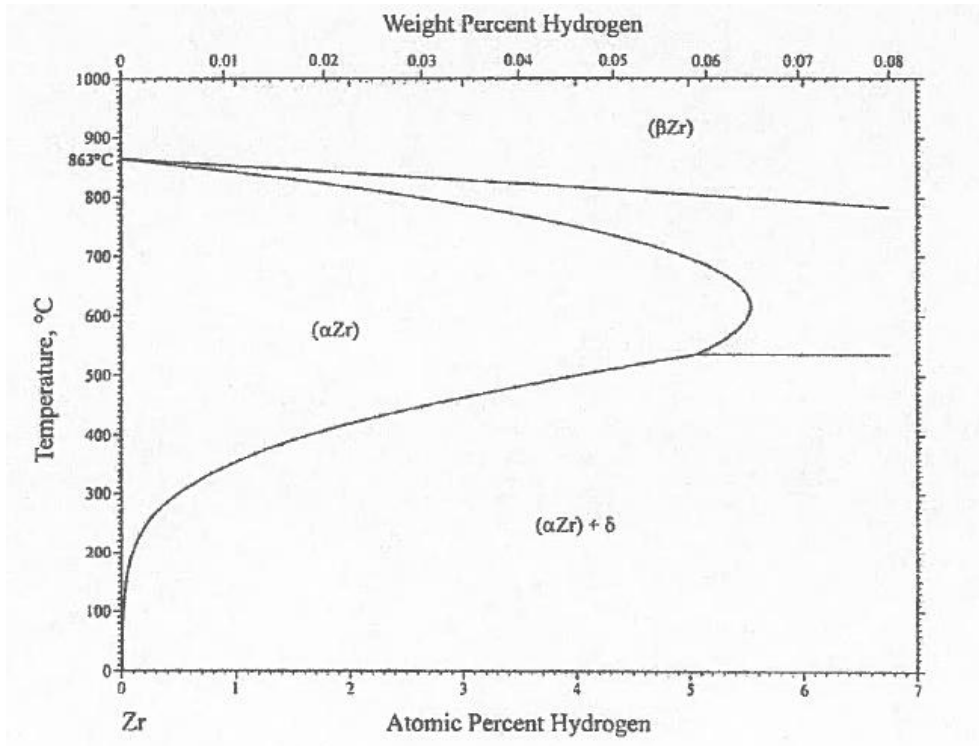
In spent fuel that is discharged from the reactor, the preferred orientation is predominantly dictated by the temperature differential that was present across the clad wall during operation and favors a circumferential alignment of hydride platelets in many cladding alloys. However, as shown above, the mobility of hydrogen in zirconium alloys is sufficient to allow hydride reorientation during spent fuel storage. In fact, the few hours required to conduct a room temperature tensile test and prepare the sample for metallographic examination are sufficient for hydride reorientation as shown in Figure 20. Examination of the hydrides in Figure 20 (magnification 100X) suggests that hydrogen must migrate approximately 0.005 cm during the time required for the tensile test and metallographic preparation. Although this time was not measured for the test specimen shown, typically, at least 24 hours would lapse between the start of the tensile test and the completion of the metallographic examination. The time required for hydrogen to diffuse a distance  $x$ , by random walk processes, is  $x^2/2D$  or approximately 20 hours thus reorientation could occur during the time required to test and prepare the sample. Additionally, hydrogen mobility during the tensile test could be enhanced by dislocation transport and hydrogen diffusion necessary to cause hydride reorientation is not a random walk process but is directed motion toward regions of lattice dilation.

In any event, hydrogen mobility at room temperature is relatively high and this high mobility, coupled with the positive partial molar volume of hydrogen in solution, provide a tendency for hydrogen to accumulate in the cold side of a thermal gradient, and the low density of zirconium hydrides, provides the thermodynamic driving force to cause:

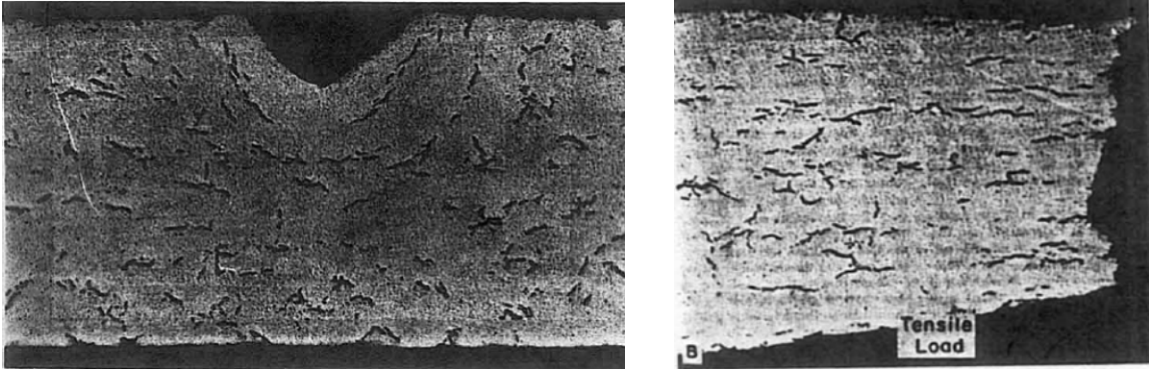
- 1) The development of a hydride rim on the cooler surface of the cladding;
- 2) Reorientation of existing hydrides perpendicular to a tensile stress and the precipitation of stress oriented hydrides as the cladding cools;
- 3) Hydrogen accumulation and hydride precipitation in regions of tensile stress such as those that exist at a stress concentration or at the tip of existing hydride precipitates, and

- 4) Localized hydride concentrations in regions of the cladding adjacent to pellet-to-cladding interaction interfaces.

Figure 15a in reference 8 illustrates how applied stresses during cooling can cause hydride platelets to form aligned clusters and the room temperature stress strain curve for Zircaloy-2 with radially-clustered hydrides shown in reference 11 demonstrates how severe the reduction in load carrying capacity can be when hydride clusters align perpendicular to the stress axis. Data presented in Reference [9] are consistent with the observation that aligned hydrides create low strength, brittle paths within the cladding and therefore may lead to any or all of the three hydrogen-induced degradation mechanisms. Additionally, the present observations are clearly consistent with the conclusion that “spent fuel rods would fail by DHC in dry storage as stress risers are present inside the cladding on cooling to below 180°C” [10] and provide a technical basis for the conclusion of this report that there is a clear potential for hydrogen-induced degradation and failure of spent fuel cladding during extended storage and transportation. Further discussion of the potential cladding degradation mechanisms is given in the subsections below.



**Figure 19** Zr-H Phase Diagram [Figure reproduced from ref. 3]



**Figure 20** Hydride Reorientation in the Near Fracture Region of a Zircaloy-2 Tensile Sample Tested at Room Temperature. A) Before Test, B) After Test (Archival data from M.R. Louthan, Jr.).

### **Section 2.2.2 References**

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### **2.2.3 Delayed Hydride Cracking (DHC) - Potential for DHC during Extended Storage**

Failures of zirconium alloy cladding tubes at end cap welds by delayed hydride cracking (DHC) during long-term storage at room temperature were first reported by Simpson and Ells in 1974 [1]. Subsequently leakage from Zr-2.5Nb pressure tubes in CANDU plants and Russian RMBK reactors has been attributed to DHC [2] and DHC has been observed in other cladding alloys and implicated to be the cause of long splits in some BWR fuel cladding, but the splits may also be attributed to oxidation of the fuel pellets.

Laboratory testing has demonstrated that the temperature dependence of DHC velocities in Zircaloy-4 is similar to those for Zr-2.5Nb (figure 8 in Reference 3). These and other similarities lead to the conclusion that the DHC behavior of Zircaloy-4 fuel cladding will be similar to that of the pressure tubes [3]. In addition to temperature, several material/environmental variables influence DHC velocities including hydrogen content, material chemistry and strength, microstructure, crack or flaw tip stress intensity, thermal history and crystallographic texture [2, 3, and 4]. Although validated crack growth models incorporating these influences are not available, the influence of some of the material/environmental variables has been established.

The temperature dependence of DHC velocities at temperatures below 275°C demonstrates that DHC is a thermally activated process. This temperature dependence does not mimic the activation energy for diffusion because hydride precipitation also depends on the hydrogen concentration reaching/exceeding the local hydrogen solubility limit. Hydrogen solubility is temperature dependent and, at a notch tip, dependent on the applied stress, the sharpness of the notch and the strength of the material [2, 5]. Other considerations that may impact DHC velocities include radiation effects and local variations in alloy chemistry.

Analysis of the impact of material and environmental variables on DHC growth rates in Zr-2.5Nb alloys [6] has shown that DHC growth rates increase with decreasing irradiation temperature, increasing operating flux and increasing operating time. These observations were made on surveillance samples and the experimentally observed axial DHC growth rates showed an Arrhenius-type temperature dependence (Figure 1 from Reference 6) with several orders of magnitude variation in crack velocities. Assessment of DHC in the surveillance samples in CANDU reactors is very prescriptive [7] and designed to provide a leak before break assessment of the pressure tubes [8].

A phenomenological description of DHC [9] concludes that the process proceeds as follows: "Hydrides nucleate and grow slowly in the high stress region of a stress riser such as a crack tip. When they reach a critical condition, probably related to size, they fracture, the crack extends and the process is repeated" [9]. The cracking process thus provides for a stable, time dependent, two-step crack growth mechanism. The first step is the diffusion of hydrogen to the crack tip region coupled with the nucleation and growth of a hydride in a region of high stress concentration, and this step is followed by hydride (or hydride metal interface) fracture and crack extension. Reference 9 concludes that there are two technologically-relevant quantities for DHC: the critical stress intensity for crack initiation and the rate of crack propagation.

Measurements of the threshold (or critical) stress intensity for the initiation of DHC in Zr-2.5Nb alloys [4] suggest values in the range of 9 to 11 MPa m<sup>1/2</sup> for samples tested between 205 and 283°C (from Figure 5 of reference 4). These threshold values are about one third of the critical fracture toughness values for Zircaloy-4 cladding that contains oriented hydrides and is tested at 25°C and much lower than the fracture toughness of the same alloy at 300°C (Figure 7 of reference 10).

The difference between the threshold stress intensity values for DHC and the critical fracture toughness values demonstrates that, if DHC should initiate in the cladding of a spent fuel rod, crack growth would initially be stable. A fracture mechanics evaluation of flaw stability (see subsection 2.2.4 below) demonstrates that the crack would penetrate the cladding before becoming unstable. Clad penetration will relieve the internal pressure in the fuel rod and minimize the driving force for crack growth. This conclusion is consistent with the assessments of DHC in Zr-2.5Nb pressure tubes which have been shown to leak as a result of stable DHC.

Evaluation of DHC from simulated debris fretting flaws showed that the threshold stress intensity for crack initiation was dependent on notch root radius and metallurgical condition as shown in Figure 5 of reference [11]. Additionally, the hydride morphology at the crack tips was markedly dependent on root radius (from Figures 10-12 of reference 11) as was the probability of failure by DHC (from Figure 7 of reference 11) [11]. The difference in failure probability for samples stressed to equivalent applied stress intensities is due to the differences in crack tip shielding caused by hydride precipitation at the notch roots.

Even though most of the DHC assessments have been for Zr-2.5Nb alloys, tests with six versions of Zircaloy-4 fuel cladding representing materials used in PWR, BWR, CANDU and Atucha reactors were basically consistent with results from Zr-2.5Nb alloys [9]. Material strength was a controlling factor for DHC crack velocities. The fastest crack rates were found in the strongest material. One basic difference in DHC in the Zircaloy-4 relative to DHC in Zr-2.5Nb alloys was the lack of striations on the fracture surfaces. Reasons for this difference were not developed.

The observations described above clearly demonstrate the possibility of DHC in fuel cladding during extended storage and transport and also suggest that the DHC behavior of zirconium based alloys is highly variable. The assembled evidence demonstrates that spent fuel rods may fail by DHC during storage upon cooling to temperatures where hydride precipitation may occur. However, such failures would produce small, through wall cracks rather than catastrophic tears in the cladding. Clearly the time/temperature profile necessary for the hydride precipitation required for DHC will depend on the hydrogen content in the cladding alloy which, as an upper bound in the reference fuel, was assumed to be 600 ppm. Reference to Figure 16 in Section 2.2.2 illustrates that hydride precipitation in an alloy containing 600 ppm hydrogen at high temperature would occur continuously as the fuel cools. Typical time/temperature profiles of fuel in dry storage suggest that a 300°C fuel cladding temperature is reached during the first few years of storage and that hydride precipitation should occur in the reference fuel. However, the onset of DHC also requires that several other conditions be satisfied.

The DHC phenomenon is generally subject to an incubation period and the impact of alloy chemistry, temperature, hydrogen content, cooling rate, time, stress intensity and other variables on this incubation period has not been established. For example, in

fatigue pre-cracked samples loaded to an initial stress intensity of approximately  $17.3 \text{ MPa m}^{1/2}$  showed large scatter in the incubation time for crack initiation and no evidence of any temperature dependence (from figure 3.13 of reference 9). Determinations of threshold stress intensities for the initiation of DHC used decreasing load techniques, thus the values determined physically approximated the stress intensity at cessation or arrest of measurable crack growth rather than the stress intensity required for DHC initiation. However, measurements with a variety of metals and alloys show that arrest and initiation  $K_{th}$  values are approximately the same, when measurements are made using reasonable experimental care. Therefore, the arrest threshold stress intensities shown in Figure 5 of reference 4 are assumed to represent the stress intensities required for DHC initiation. Clearly, metallurgical and compositional differences among the cladding alloys may alter the threshold stress intensity. In fact, in-service and laboratory experience with a variety of metals and alloys has shown that hydrogen induced cracking and stress corrosion cracking will occur on samples and components that have such minute defects that estimation of an applied stress intensity is extremely difficult. However, this difficulty may be overcome by evaluations using samples with smooth surfaces.

The behavior of statically loaded, smooth surface samples shows that environmentally induced cracking sequences can be divided into three stages: 1) an incubation stage that represents the time required for crack nucleation, 2) a crack propagation stage that represents the time required for a nucleated crack to grow to a critical size, and 3) final fracture. The effect of applied stress on this cracking sequence is illustrated in Figure 1 of reference 12. This illustration assumes the existence of  $\sigma_{th}$  or stress level below which the degradation process will not initiate. In-situ studies [13] show that a stress of 75 to 80 MPa is required for hydride re-orientation in Zircaloy. Earlier studies [14] showed that the threshold stresses were very dependent on fabrication history with values ranging from 40 to 120 MPa. Stress induced orientation of hydrides is assumed to be a critical component of the DHC process, thus  $\sigma_{th}$  for DHC should be approximately 80 MPa but could be as low as 40 MPa depending on the microstructure, texture, fabrication processes and other material/environmental variables associated with the particular zirconium based cladding.

The fact that smooth specimens, stressed above  $\sigma_{th}$  may be expected to fail by DHC can be conceptually combined with the threshold stress intensity for DHC initiation to estimate a "safe zone" where DHC should not develop. This concept is shown schematically in Figure 61 of reference 12 and demonstrates that a zone of conditions where DHC will not occur depends on both the stresses acting on the fuel cladding and the size of a flaw or crack in the cladding. Technologies for flaw characterization by non-destructive evaluation techniques are well established and have been applied to the identification of DHC induced flaws in zirconium alloys [15].

Interpretation that DHC will not occur in the absence of applied stresses (pressure for example) acting on the cladding must be viewed with caution. Residual stresses from fabrication, handling or in-service situations may provide local stresses that are sufficient for the incubation of DHC. Once initiated, DHC may become a self-sustaining process because of the stresses induced by the volume expansion associated with hydride precipitation. The potential for self-sustaining processes is evident in the appearance of the aligned hydride clusters.

**Summary of Current Observations of DHC during Storage** - There is no evidence that DHC is currently a significant problem in dry storage environments. However, the phenomenon has not been thoroughly studied by evaluating the cladding on either low or high burn-up fuels in dry storage. The evidence presented suggests that DHC is certainly possible and, given the time frames considered, the development of stable, throughwall cracks by DHC could occur. Cracks in the fuel cladding will provide pathways for fission product release and, if oxygen is present in the canister, will allow oxidation of the fuel.

Additionally, cladding, irradiation, and handling variables will impact the potential for DHC and viable models to estimate the time to cracking as a function of storage conditions do not exist. Furthermore, even though there are numerous data for a variety of alloys that suggest, once DHC has initiated, the temperature dependence of DHC crack velocity shows an Arrhenius type behavior, the absolute value of crack velocity is sensitive to a variety of thermal, mechanical, and metallurgical variables. Sufficient information to assess the impact of material and environmental variables on DHC initiation and growth does not exist.

A recent summary report [9] demonstrated that:

- a) DHC has been responsible for several failures in reactor components,
- b) Zircaloy fuel cladding is not immune from DHC,
- c) DHC cracking rates are independent of crack length and stress intensity,
- d) DHC crack velocities are highest in the highest strength materials and the added strength generated by irradiation increased crack velocities by a factor between ten and fifty,
- e) Threshold stress intensities for crack initiation increase with increasing temperature, and
- f) Failure of fuel cladding by DHC should not occur until the cladding has cooled to a temperature where DHC is possible.

The evaluations discussed above show that if cladding cracks have been produced by DHC these cracks should penetrate the wall and allow release of the contained fission product gasses into the spent fuel canister and ingress of the canister environment into the fuel rod. Although the information on the impact of time, temperature, irradiation, hydrogen content etc., on DHC is not sufficient to predict the behavior of any specific fuel assembly, the occurrence of DHC will be stochastic. Only a fraction of the susceptible cladding will experience cracking, much like only a fraction of the stress corrosion cracking susceptible welds in reactors have cracked. As an example, stress corrosion cracking in the near weld regions of austenitic stainless steel primary coolant water piping in the Savannah River Site production reactors had been attributed to sensitization of the heat affected zone of the welds, residual stresses in the near weld region, and the presence of process water at approximately 100°C [16]. There were, literally, thousands of such welds and stress corrosion cracking was observed in less than fifty welds. Additionally, in the welds where cracking was observed, only a fraction of the weld length cracked and most of the weld was not degraded by the exposure.

The tendency for DHC in the cladding of spent fuel rods is anticipated to follow a trend similar to that found for stress corrosion cracking, thus the absence of DHC in a few elements selected for evaluation does little to establish a firm technical basis to predict the behavior of the remaining fuel. An evaluation scheme that includes PIE of both high and low burn-up fuels is needed to maximize the technical gains that result from the limited number of evaluations that will be made during the surveillance of spent fuel during dry storage.

**Assessment of DHC in Fuel Subject to Extended Storage and Transportation** - The fact that DHC has been observed in zirconium based nuclear components coupled with the observation that the conditions that produce DHC may develop as the temperature of the fuel cladding decreases, demonstrate a significant potential for the failure, by DHC, of a number of fuel rods during long term storage. Such failure will only occur after the cladding has cooled down from the initial dryout anneal, during which about 200-300 ppm of hydrogen will go into solution, depending on the temperature reached. As the clad cools the dissolved hydrogen will re-precipitate as hydrides in the clad structure.

There is not sufficient information available to provide models to account for metallurgical, mechanical and chemistry effects on DHC and allow realistic predictions of cladding behavior during EST.

Although additional data may improve modeling capabilities, a more realistic approach to assuring safe storage is to develop the handling and transport technologies necessary for fuels with minor failed cladding and to establish monitoring technologies to detect throughwall DHC. Cladding cracks should be anticipated and, as shown, the ductility and toughness of intact cladding may be significantly reduced by EST, especially at the lower temperatures encountered after decades of storage. The stresses necessary for the initiation of DHC in non-flawed regions may be as low as 40 MPa [14] and can readily be developed by the precipitation of a hydride nugget or rim or exist as residual stresses in the fuel rod.

Cracks in the fuel cladding are expected to be minor and not lead to gross rupture, but it would cause an impact to readily retrieve the fuel. Specifically, failure from DHC would

- 1) increase the source term in the canister/cask,
- 2) lessen the ready retrievability of the fuel during handling, post-storage transport or disposition, and
- 3) create a path to expose fuel pellets and fragments to oxygen if present in the internal canister environment because of bolting failures, seal leakage and/or stress corrosion cracking of the canister.

There would be no impact, by DHC, to cause a loss of confinement provided by the canister/cask of the DCSS.

**DHC Data Needs for EST and General Recommendations for Resolution** - The data and models currently available to predict DHC during long-term dry storage are very limited and are of minimal use to the assessment of DHC in irradiated fuel cladding. Additional data are necessary and as data are accumulated, predictive models for DHC



need to be developed, verified, and modified as appropriate. High burn-up fuels are anticipated to contain more hydrogen than low burn-up fuels, thus the temperature range where hydride precipitation will begin will differ for these two fuel classes. The susceptibility of zirconium alloys to stress orientation is influenced by the hydrogen content and thermal cycling, thus the data/model development schemes must include fuels having a range of burnup and a range of hydrogen content in the fuel cladding. Additionally, concurrent with the development of DHC modeling, it is recommended that improved assembly temperature profile modeling be included. The temperature profiles both within and between stored assemblies from dry out annealing to the very long term storage will be critical parameters in evaluating fuel performance, including the potential for DHC. Specifically,

1. Testing & Analysis Activity: Predictive model(s) for DHC including HBU fuel with high hydrogen contents with respect to thermal equilibrium at low (< 100°C) temperatures should be established for EST.
2. Cask Demonstration Activity: As part of a cask demonstration, DE should be performed with appropriate sampling to check for DHC. Fuel that has undergone a high temperature dryout cycle and has subsequently been at temperatures below approximately 300°C in dry storage and that has experienced in-core failure (to provide “seed” cracks) is judged to be most likely to exhibit DHC.

The impact of DHC to EST is anticipated to be troublesome but manageable considering: 1) that the DHC mechanism has the potential to form flaws in the cladding; 2) any through-clad failure by DHC during dry storage would not be a gross cladding failure but may lead to an increase in source term within the confinement; and 3) flaws formed by DHC (and other existing cladding flaws) may be subject to extension with loading from handling and transportation.

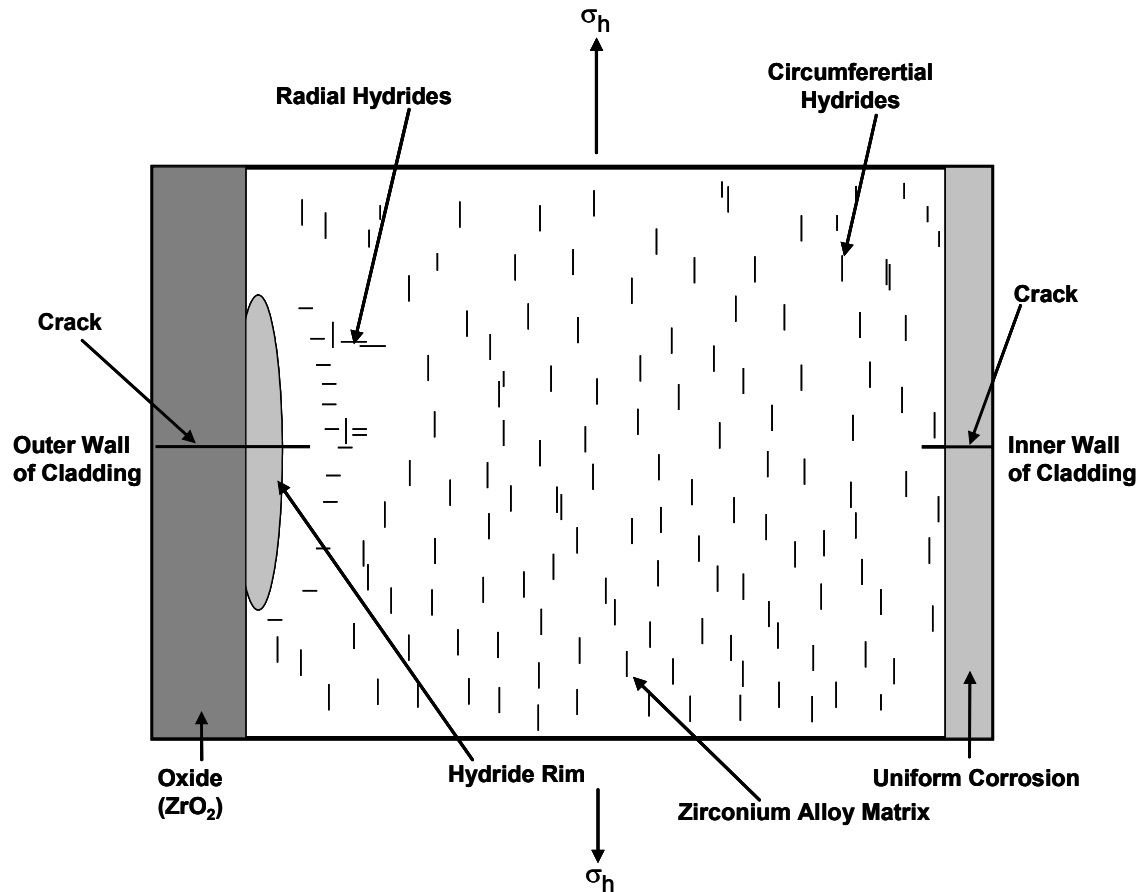
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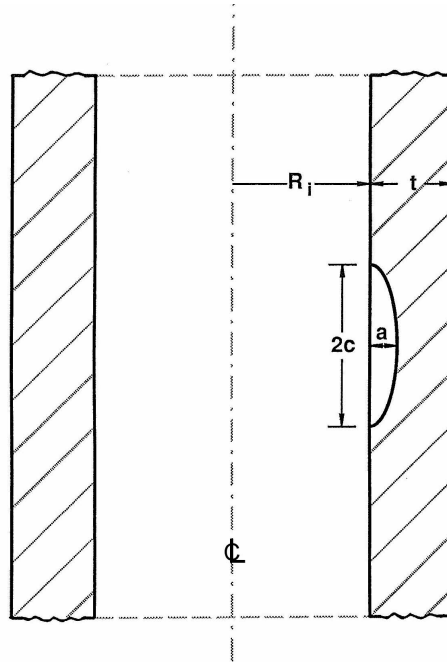
#### **2.2.4 Flaw Stability Assessment in Zircaloy Tube**

A simple assessment of axial flaws was performed to evaluate crack stability in a low-toughness (embrittled) cladding. The evaluation was performed to investigate the potential for a gross cladding axial split to release the fuel materials and fission products from a hydride-embrittled cladding tube under internal pressure while in dry storage. The most limiting configuration under internal pressure loading is cracks aligned to grow axially along the tube while being driven by hoop stresses.



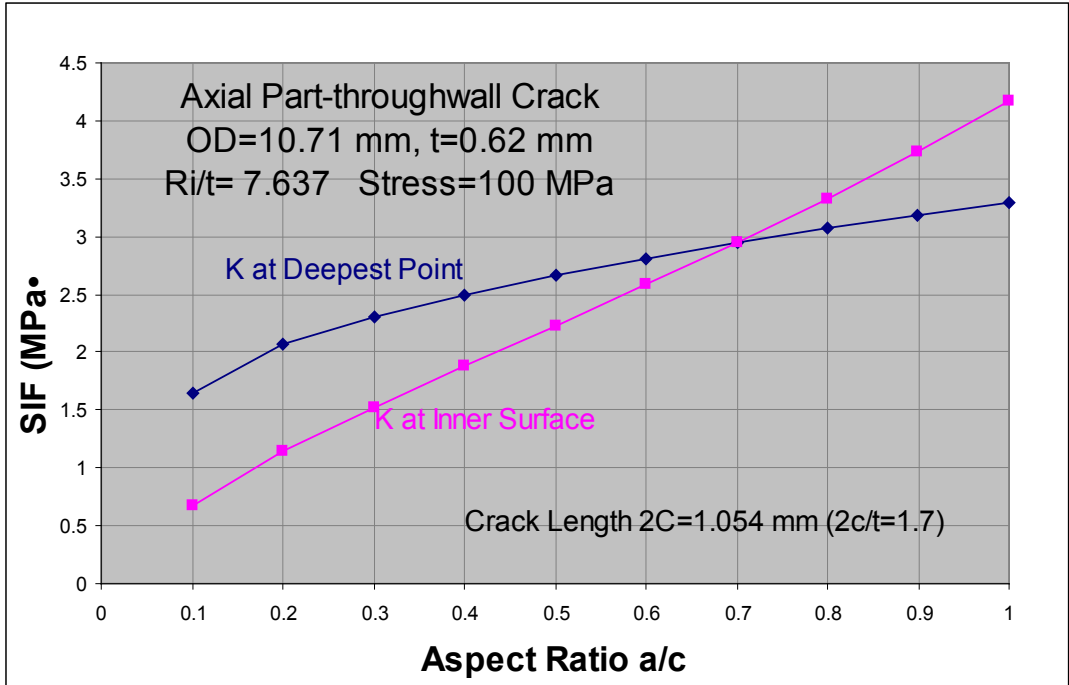
**Figure 21** Postulated Flaws in the Cladding

Cracking emanating from either the ID or OD of the cladding is presumed as schematically shown in Figure 21. An axial thumbnail crack on the inner cladding surface is first considered. The thumbnail crack is simplified as a semi-elliptic crack with length  $2c$  and depth  $a$  into the cladding (aspect ratio  $a/c$ ) as shown in Figure 22 for a fuel rod with inner radius  $R_i$  and cladding thickness  $t$ . The crack is assumed to be loaded with a uniform circumferential stress resulting from fission gas pressure and other mechanical loadings. Linear elastic response is considered through this analysis so the Mode I stress intensity factors (SIF), denoted by  $K$ , can be calculated from handbook solutions [1]. The SIFs were calculated at the deepest penetration point and on the inner fuel rod surface, which provide insight on its potential to penetrate the cladding (by increasing  $a$ ) or the potential to increase its crack length ( $2c$ ). The membrane stress is assumed to be 100 MPa, which allows convenient scaling of the calculated SIFs that are proportional to the stress loading based on the principle of linear elasticity. A cold-worked stress-relieved Zircaloy-4 clad fuel rod was chosen for the fracture analysis with low fracture toughness due to hydriding. This rod has an outside diameter of 10.71mm and a wall thickness of 0.62mm [2].

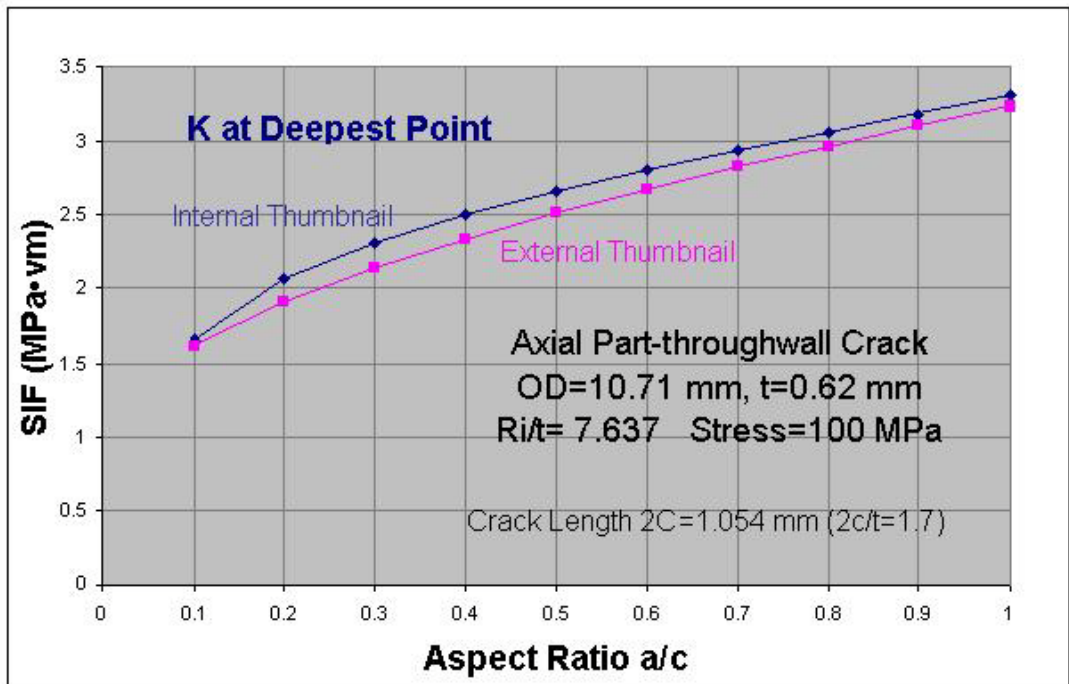


**Figure 22** An axial semi-elliptic crack with aspect ratio  $a/c$  in a fuel rod with inner radius  $R_i$  and cladding thickness  $t$ .

The calculated SIFs for the internal flaw are shown in Figure 23. Figure 24 shows the SIF results at the tip of the external flaw in comparison to the internal flaw as a function of flaw penetration depth. The crack length  $2c$  is 1.054 mm, which is the maximum size limited by the validity of the solution [1]. This crack length is at 1.7 times the cladding thickness. Based on the result, the SIF for this crack configuration is far below the threshold value for DHC to initiate (in the range of 9 to 11 MPa  $m^{1/2}$  based on Zr-2.5Nb alloys which have been widely studied particularly for CANDU reactor systems). It is expected that the DHC threshold for Zircaloy-4 would be in the similar range. In addition, it was also reported in Table 3 of reference [3] that the fracture toughness is about 25 MPa  $m^{1/2}$  at room temperature for Zircaloy-4 sheet with random and mixed hydride platelet. These material property data suggest that it would require about 250 MPa to initiate DHC at room temperature for this semi-circular crack (maximum SIF), and about 600 MPa to propagate this crack in a mechanically unstable manner.



**Figure 23** Stress intensity factors at 100 MPa uniform stress loading for semi-elliptic cracks with length  $2c= 1.7t$  in a typical fuel rod as functions of penetration depth ( $a/c$ ).



**Figure 24** Comparison of stress intensity factors at crack tips of internal and external thumbnail cracks at 100 MPa uniform stress loading.

In the case of an existing flaw which has already penetrated the cladding, the stability of the crack can be analyzed with an axial throughwall crack shown in Figure 25, where the

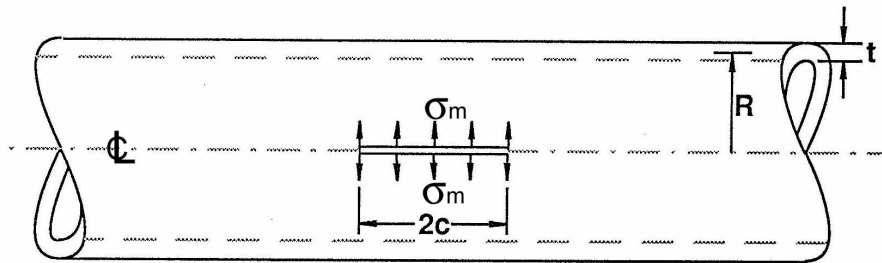
crack is under a uniform stress  $\sigma_m$  through the cladding thickness. The SIFs at a loading of 100 MPa were calculated up to the maximum crack length (17.7 mm or 28.5 times the cladding thickness), allowed by the valid solution [4].

According to the previously quoted data for the fracture toughness of hydrided Zircaloy-4 sheet at room temperature (about  $25\text{MPa m}^{1/2}$  [3]), a throughwall crack must be 6 mm in length (or 10 times cladding thickness) to be split by the 100 MPa loading as indicated by Figure 26. Note that in this calculation, the cladding stress was assumed not to be relieved by the pre-existing crack.

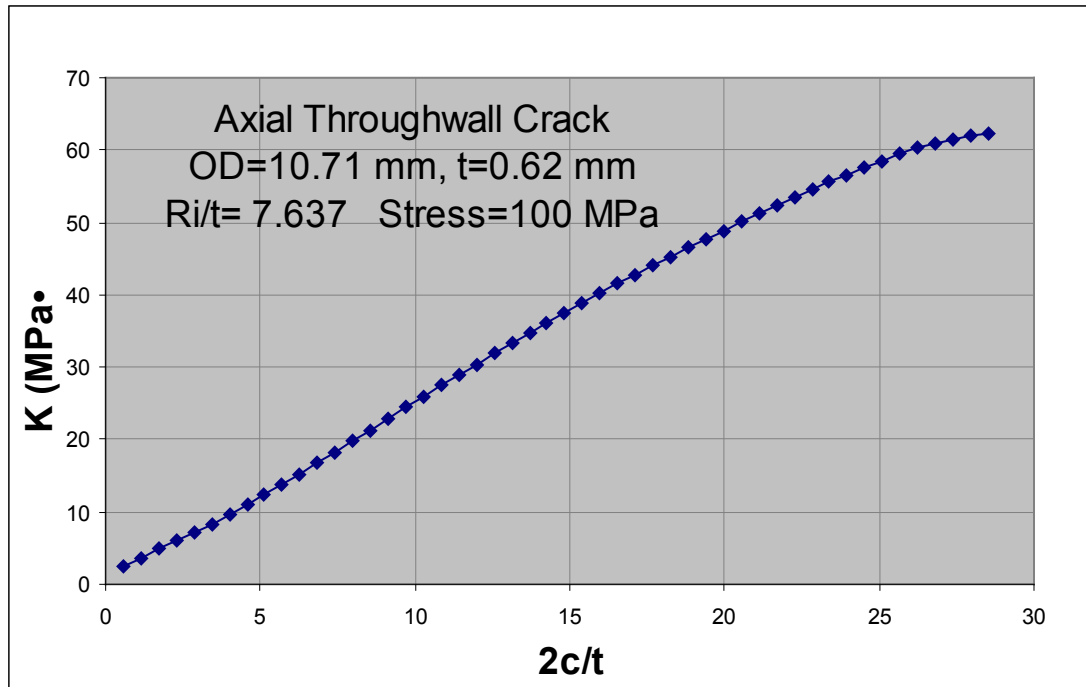
These results for the subcritical flaw growth and stability assessment suggest several conclusions:

- 1) internal pressure on the cladding is not sufficient to drive subcritical cracking; however, residual stresses and stresses generated by hydride precipitation may be sufficient to cause DHC.
- 2) should a subcritical flaw form during the dry storage period, the internal pressure on the cladding would not cause a gross rupture (e.g., axial split) of the cladding.

These conclusions are based on the cited published data for threshold stress intensities for DHC and room temperature fracture toughness value for a heavily hydrided microstructure. These results are consistent with the conclusion in reference [5] for no axial splitting failure of fuel rods. However, the analysis does not consider the development of a clustered, axially oriented hydride distribution. Current data suggest that such distributions may develop during long term storage and lead to stable, throughwall crack growth by DHC (see Section 2.2.3).



**Figure 25** An axial throughwall crack with length  $2c$  loaded by a uniform stress across the thickness  $t$  in a tube with mean radius  $R$ .



**Figure 26** Stress intensity factors for axial flaws with 100 MPa uniform stress loading at various initial crack lengths.

**Flaw Stability Evaluation Data Needs for EST and General Recommendations for Resolution** - The methodology that is used for evaluating stability in Zircaloy rods involves comparison of the strain energy density against the material property of Critical Strain Energy Density (e.g., reference [6]). This methodology has been used in evaluation of fuel resistance to fracture in transportation loading events, but is not currently endorsed by the NRC. This methodology is not readily amenable to flawed materials due to discontinuities imposed by the flaws [5], and alternative methodologies are needed to provide improved accuracy and characterize flaw stability in a hydrided rod with flaws.

It is likely that fuel from EST will be subjected to loads during retrieval, handling, and transportation while the cladding temperature is below its DBTT. If this fuel contains through-clad and/or non-through-clad flaws the fuel may be susceptible to brittle fracture. It is recommended that a consensus standard fracture approach be developed for fuel flaw evaluation. This approach should, consider cladding type, temperature, hydride microstructure, and potential flaws as well as the loads developed during storage, handling, and transportation. Specifically,

1. Testing & Analysis Activity: Develop fracture methodology, with national consensus, for flaw stability in Zircaloy fuel tubes with explicit consideration of subcritical growth leading to mechanical instability, multiple flaws, hydride (second phase) effects, flaw orientation, loading rates, and DBTT, etc. Existing fracture data for Zircaloy would be applied to this methodology and future materials properties and testing methods would be suggested for improvement in accuracy of flaw stability estimation.

The increased susceptibility of fuel to brittle failure due to both HBU (potential higher hydrogen content) and low temperature condition for EST make the fuel vulnerable during handling (with potential off-normal drop) and transportation loading conditions including accident. A comparison of the robustness and accuracy of fracture mechanics methods with the strain energy density method would establish an important technical basis for EST.

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#### **2.2.5 Hydride Embrittlement – Reduction in Tensile Ductility**

Hydride embrittlement will occur in zirconium alloy clad spent fuels placed in extended storage because reactor service has increased the hydrogen content in the cladding and the storage temperature will decrease to the point that hydrides precipitate in the cladding. The fact that hydride precipitation lowers the ductility of zirconium based alloys was well established by the early 1960s [1] and has been confirmed in numerous investigations. For example, Figure 27 shows the changes in the strength and ductility of Zircaloy-4 tensile samples as a function of hydrogen content.

The literature clearly demonstrates that hydrogen uptake and hydride precipitation decrease the ductility of zirconium based alloys. The extent of hydride embrittlement is a function of the hydrogen content and the hydride orientation. Hydride orientation is so important to the embrittlement process that models to estimate the ductility (and strength) of Zircaloy as a function of fluence, burnup, oxide thickness, hydrogen concentration, temperature, strain rate and cold work contain the statement "the models described here apply only to cladding with circumferential hydrides and do not apply to cladding with radial hydrides or significant hydride blisters or spalling" [4]. The precipitation of radial hydrides can reduce the ductility of Zircaloy cladding alloys to

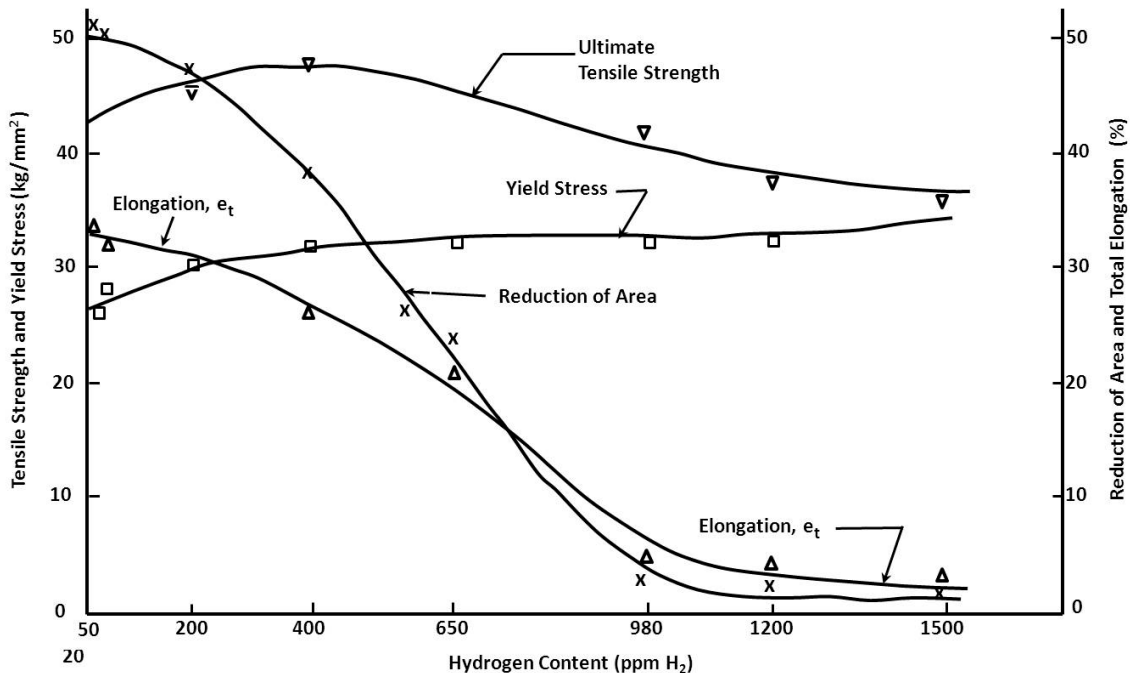


essentially zero and cause normally ductile samples to fail while in the elastic portion of the stress-strain curve [1].

Irradiation also reduces the ductility of zirconium based alloys and the combined effects of irradiation show that when irradiated to a fluence of  $1.6$  to  $9.5 \times 10^{25}$  n/m<sup>2</sup>, Zircaloy-2 specimens lose essentially all their ductility when the hydrogen content reaches 600 ppm (Figure 4 of Reference 4). The hydrogen concentration in irradiated zirconium alloys generally increases with increasing fluence (Figure 2 of Reference 4) although there is considerable scatter in the hydrogen concentration in fuel claddings after irradiation to high fluence. Therefore, although most HBU fuels have a range of fluence between  $9$  and  $11 \times 10^{25}$  n/m<sup>2</sup> the hydrogen content in the cladding alloys on those fuels can be quite variable [4]. For example, the corrosion and hydrogen levels at a burnup of 60 GWd/MTU for low tin Zircaloy-4 can vary between 50 and 100  $\mu$ m of oxide and 400 to 800 ppm hydrogen [4]. Similar estimations for Zircaloy-2 from BWR fuel at the same burnup suggest that the oxide thickness can vary between 20 and 45  $\mu$ m and the hydrogen content between 100 and 220 ppm.

There are numerous literature examples demonstrating that the combination of irradiation and hydride precipitation can significantly reduce the ductility of zirconium base alloys. For example a 1994 summary paper [5] shows irradiation increasing the strength and decreasing the ductility of Zr-2 alloys. Figure 27 illustrates the effects of hydrogen on mechanical properties and reaffirms the hydrogen induced reductions in ductility. Another investigation [6, 7, and 8] of the reduction loss of ductility with hydrogen up to 4500 ppm shows the importance of the hydride microstructure on the ductility in zircaloy claddings.

These reductions are most apparent at low temperatures and therefore will become more serious as the temperature of the fuel cladding decreases during storage. There is not sufficient information available to assess the structural integrity of spent fuel during storage and transport when the cladding has experienced significant hydride embrittlement and fails under elastic stresses.



**Figure 27** Impact of Hydrogen Content on the Tensile Properties of Zircaloy-4 at Room Temperature [Figure reproduced from ref. 2].

**HE Ductility Reduction Data Needs for EST and General Recommendations for Resolution** - Additional data should be developed and models, such as those published in Reference 4, must be modified to include cladding alloys with radial hydrides and also include the emerging cladding alloys that may show behaviors that differ significantly from those of Zircaloy. Emphasis should be placed on the HBU fuels because their hydrogen content is expected to exceed that for low burnup fuels. Specifically,

1. Testing & Analysis Activity: Synthesize tensile data to establish model(s) for properties based on alloy composition, hydrogen content, hydride orientation, static and dynamic loading, bi-axial (mixed mode) loading, radiation damage and hydride microstructures, etc. to enable accurate prediction across fuel varieties.

The increased susceptibility of fuel to brittle failure due to both HBU (potential higher hydrogen content) and low temperature condition for EST make the fuel vulnerable during handling (with potential off-normal drop) and transportation loading conditions including accident.

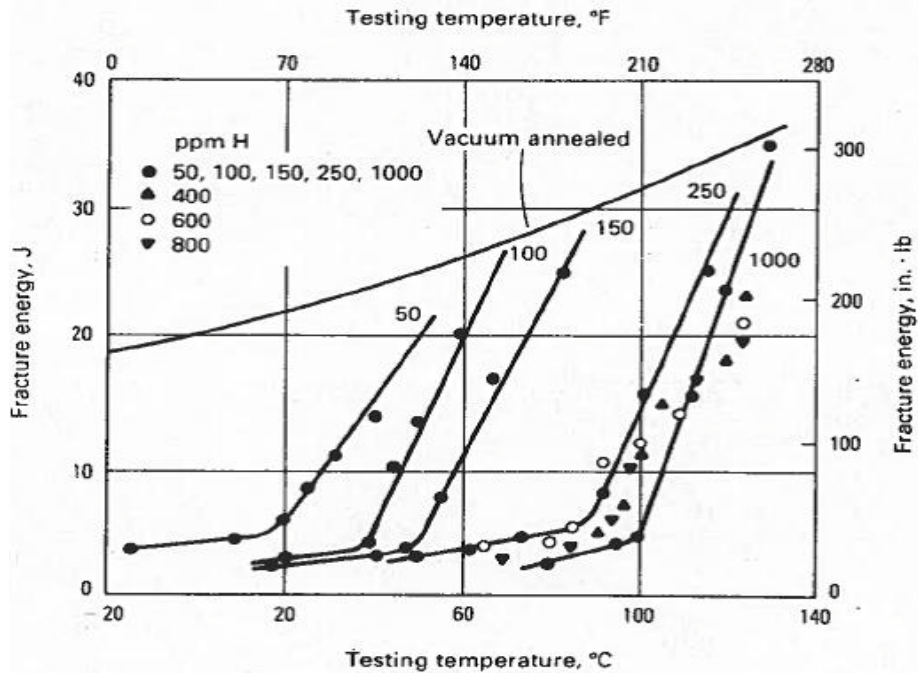
### **Section 2.2.5 References**

- [1] Louthan, M.R., Jr. and Marshall, R.P., "Control of Hydride Orientation in Zircaloy", *Journal of Nuclear Materials*, Vol. 9, pp. 170-184 (1963).
- [2] Cox, B. and Rudling, P., "Hydriding Mechanisms and Impact on Fuel Performance", Advanced Nuclear Technology AB, ANT, Sweden, 2000.

- [3] Huang, J.-H. and Huang, S.-P., "Effect of Hydrogen Contents on the Mechanical Properties of Zircaloy-4", J. Nucl. Mater. Vol. 208, pp. 166-180, (1994).
- [4] Geelhood, K.J., Luscher, W.G., and Beyer, C.E., "PNNL Stress/Strain Correlation for Zircaloy", PNNL-17700, July 2008.
- [5] Zirconium Alloys in Nuclear Applications, Chapter 7 in Materials Science and Technology, Vol 10B, Nuclear Materials, Part II, VCH New York.
- [6] Arsene, S., Bai, J.B., and Bompard, P., "Hydride Embrittlement and Irradiation Effects on the Hoop Mechanical Properties of Pressurized Water Reactor (PWR) and Boiling-Water Reactor (BWR) ZIRCALOY Cladding Tubes: Part I. Hydride Embrittlement in Stress-Relieved, Annealed, and Recrystallized ZIRCALOYs at 20°C and 300°C," Met. Mat. Trans. A, Vol. 34A, Number 3, p. 553 (March 2003).
- [7] Veleva, M., Arsene, S., Record, M., Bechade, J.L., and Bai, J.B., "Hydride Embrittlement and Irradiation Effects on the Hoop Mechanical Properties of Pressurized Water Reactor (PWR) and Boiling-Water Reactor (BWR) ZIRCALOY Cladding Tubes: Part II. Morphology of Hydrides Investigated at Different Magnifications and their Interaction with the Processes of Plastic Deformation," Met. Mat. Trans. A, Vol. 34A, Number 3, p. 567 (March 2003).
- [8] Arsene, S., Bai, J.B., and Bompard, P., "Hydride Embrittlement and Irradiation Effects on the Hoop Mechanical Properties of Pressurized Water Reactor (PWR) and Boiling-Water Reactor (BWR) ZIRCALOY Cladding Tubes: Part III. Mechanical Behavior of Hydride in Stress-Relieved Annealed and Recrystallized ZIRCALOYs at 20°C and 300°C," Met. Mat. Trans. A, Vol. 34, Number 3, p. 579 (March 2003).

### **2.2.6 Hydrogen Embrittlement - Hydrogen Induced Shifts in the DBTT**

Numerous references, including those highlighted in Section 2.2.5, show that the fracture resistance of zirconium based alloys decreases with increasing hydrogen content. The decrease includes a concomitant shift in the DBTT, as illustrated in Figure 28 and the extent of the shift will vary with alloy composition, hydride orientation, temperature and irradiation level as well as hydrogen content. Although the hydrogen induced decrease in fracture resistance is well established, the database to analyze the influence of hydride orientation, hydride rims, alloy type and irradiation on fracture resistance is not comprehensive and virtually non-existent for newer cladding alloys. High burn-up fuel may be especially susceptible to hydrogen embrittlement and hydrogen induced shifts in the DBTT because of the higher hydrogen contents that result from the longer in-reactor time frames [1].



**Figure 28** Ductile-to-brittle transitions in hydrided zirconium [Figure reproduced from ref. 2].

The shift in DBTT may become critical to the behavior of fuel cladding during post storage handling and transport. Significant reductions are possible if the combination of alloy type, thermomechanical history, hydrogen content and storage conditions lead to the formation of clustered, radial hydrides. Such hydride formations can make the cladding brittle in a room temperature tensile test, even when the hydrogen content is only 100 ppm and the alloy has not been irradiated [3].

The elevation of the DBTT is generally associated with the presence of hydrides in the microstructure and should therefore become increasingly important as the storage temperature decreases. Therefore the potential for such a shift should increase with increasing storage time (decreasing storage temperature) and reach its maximum at the end of the fuel storage period. This potential for a hydrogen induced increase in the DBTT will generally increase as the fuel burnup increases because the hydrogen content of the cladding alloys tends to increase with increasing fuel burnup. However, the database necessary to analyze the time/temperature/burnup dependence of the DBTT for cladding alloys during very long term storage is insufficient and the data necessary to couple the DBTT with other cladding variables are minimal, at best.

These evaluations illustrate a significant potential for hydrogen induced reductions in the fracture resistance of the cladding on stored fuel assemblies and also illustrate that there are no models to accurately assess the magnitude of that potential. Additionally, even if the potential for hydrogen induced increases in the DBTT could be established, the models to assess the impact of DBTT increases on the handling and retrieval of spent

nuclear fuels after very long term storage are not currently available, as discussed in Section 2.2.4.

**Data Needs for Hydrogen Induced Increases in the DBTT** - The data and models currently available are not sufficient to assess the impact of long term storage on the DBTT of cladding alloys. Data are needed to establish the combined effects of alloy content, hydrogen content, irradiation, temperature and other cladding variables on the DBTT (and other measures of fracture resistance) of fuel cladding. Emphasis should be placed on HBU fuel [1] and efforts should be made to establish the singular and combined effects of irradiation and hydrogen content on fracture resistance. Specifically,

1. Testing & Analysis Activity: Synthesize existing fracture data and develop the additional fracture toughness data necessary to establish model(s) for properties based on temperature, alloy composition, hydrogen content, hydride orientation (with respect to crack orientation), static and dynamic loading, bi-axial (mixed mode) loading, combined radiation damage and hydride microstructures, etc. The models developed should enable accurate prediction across fuel varieties and be validated using a variety of fuel claddings.

The increased susceptibility of fuel to brittle failure due to both HBU (potential higher hydrogen content) and low temperature condition for EST make the fuel vulnerable during handling (with potential off-normal drop) and transportation loading conditions including accidents.

Measurements of the fracture resistance of irradiated cladding requires significant resources and, before such resources are allocated, attempts should be made to model the behavior of flawed fuel assemblies during postulated post storage handling and transport conditions when the fracture resistance of the cladding is minimal (represented by the lower shelf fracture toughness at room temperature). A satisfactory outcome for a validated model using lower shelf fracture toughness properties could mitigate the need for data on irradiated, high burn-up fuel cladding. However, if a satisfactory outcome does not develop, data gathering for HBU spent fuel cladding should become a data need focus.

### **Section 2.2.6 References**

- [1] Very High Burn-ups in Light Water Reactors, ISBN 92-64-02303-8, Nuclear Energy Agency, NEA 6224, 2006.
- [2] Fig. 55, Environmentally Induced Cracking, p 175, ASM Metals Handbook, Vol. 13, Corrosion: Fundamentals, Testing and Protection, ASM International, Materials Park, OH (2003).
- [3] Marshall, R.P., and Louthan, M.R., Jr. "Tensile Properties of Zircaloy with Oriented Hydrides", Trans. ASM, Vol. 56, pp. 693-698, 1963.

### **2.2.7 SCC of Fuel**

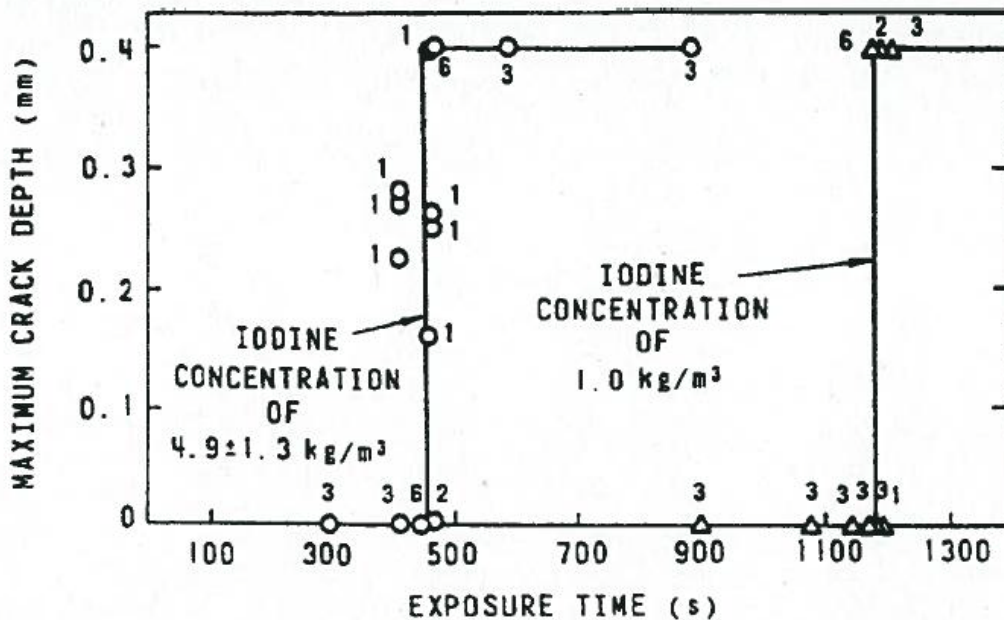
Stress corrosion cracking of Zircaloy clad SNF due to the presence of fission product iodine was observed during the 1960's and continued through the early 1980's. This cracking followed reactor power transients, particularly in BWRs. The stress corrosion process was identified as Pellet Cladding Interaction (PCI) and the cracks developed from the clad interior. To address this problem, a layer of pure zirconium was added to the inner surface of the cladding, effectively eliminating the susceptibility to SCC while the fuel was in the reactor [1].

However, many of the fuel rods without the pure zirconium layer have been placed into dry storage and the potential for recurrence of SCC as a result of radiolysis of CsI has not been thoroughly investigated. Additional studies of the potential for SCC in zirconium based fuel cladding alloys placed in long term storage are needed because even though the mechanism of cracking is well established, the failure propensities still remain unresolved for spent fuel rods. Although a number of works have been conducted on iodine SCC of Zircaloy in relation to the pellet clad interaction induced failures of LWR fuel rods, most of the experimental works in which Zircaloy was exposed to iodine and other chemical elements are within the range of operating temperatures below about 320°C and exposure times shorter than 259 ks [2]. There is little data related to longer time exposures or other temperature conditions.

Most of the fission-product iodine is tied up in compounds with zirconium and with fission-product cesium and exists in the fuel as  $ZrI_4$  or CsI. Experimental evidence indicates radiolysis can create an iodine partial pressure approximately 9 orders of magnitude greater than would be otherwise present in a spent fuel rod at 400°C [3]. The presence of this free iodine may lead to SCC in fuel with appropriate cladding stress/strain conditions. Additionally, as seen in Figure 29, the incubation time for cracking is highly dependent on the iodine vapor concentration.

In addition to the consideration of the classic PCI–SCC related cracking, the fission products present on the clad ID surface may synergistically contribute to crack propagation by interacting with the DHC mechanism. Most if not all of the DHC tests were done with de-fueled clad samples. These tests did not have the added parameter of the SCC factor from the I or Cs. Thus, future DHC testing should also consider the potential contribution of the elements/compounds that are present at the clad ID surface.

**Top Nozzle – Stainless Steel Connecting Tubes Stress Corrosion Cracking** - NRC issued Information Notice 2002-09 to illustrate the fact fuel handling events had resulted in the separation of the top nozzle from a fuel assembly. This separation caused the subsequent drop of the assembly during fuel movement [4]. The fuel assemblies were identified as Westinghouse fuel assemblies supplied in the early to mid-1980s that contained materials that were susceptible to intergranular stress-corrosion cracking (IGSCC). Several methods to allow this fuel to be loaded into dry storage have been used [5]. Fuel placed in dry storage should be evaluated for such cracking [4].



**Figure 29** Exposure Time Dependence of Iodine-Vapor-Induced SCC [Figure reproduced from ref. 2].

**Fuel SCC Data Needs for EST and General Recommendations for Resolution:**

Failures due to PCI-SCC have occurred with earlier generation Zircaloy cladding but have been generally mitigated in later generation fuel. Nevertheless, fuel susceptible to SCC is in dry storage, and radiolysis and exposure time may be sufficient for incubation of this degradation phenomenon. Fuel should be examined for SCC to provide additional information. Specifically,

1. Cask Demonstration Activity: As part of a cask demonstration, DE of the cladding should be performed with appropriate sampling to check for SCC. Considerations include zirconium-lined and non-lined fuel. HBU fuel that may be susceptible to DHC should be evaluated to check for potential interactive effects.

The significance of cladding SCC to EST is considered minimal considering: 1) the issue for present fuel has been resolved with the addition of zirconium to the ID surface; 2) any fuel rod breach that would occur due to SCC would be a minor breach with a small release to the confinement provided by the DCSS.

**Section 2.2.7 References**

[1] Armijo, J.S., Coffin, L.F., Rosenbaum, H.S., Development of zirconium barrier fuel cladding, Zirconium in the Nuclear Industry – 10th International Symposium, ASTM STP 1245, Eds, A.M. Garde and E.R. Bradley, ASTM, West Conshohocken, PA, (1994), pp. 3–18.

- [2] Mayuzumi, M., Onchi, T., and Matsuo, Y., "Failure Propensities Of Pressurized Zircaloy Tube Containing Iodine And Other Chemical Species," Journal Of Nuclear Science And Technology, 30 [12], pp. 1288-1298 (December 1993).
- [3] Konashi, K., Kamimura, K., and Yokouchi, Y., "Estimation of Irradiation Induced Iodine Pressure in an LWR Fuel Rod," J. Nucl. Mater. Vol. 125, pp. 244-247, (1984).
- [4] Information Notice 2002-09, "Potential for Top Nozzle Separation and Dropping of a Certain Type of Westinghouse Fuel Assembly", on February 13, 2002.
- [5] Industry Spent Fuel Storage Handbook EPRI 1021048 Final Report, July 2010.

### **2.2.8 Fission Product and Helium Gas Pressurization**

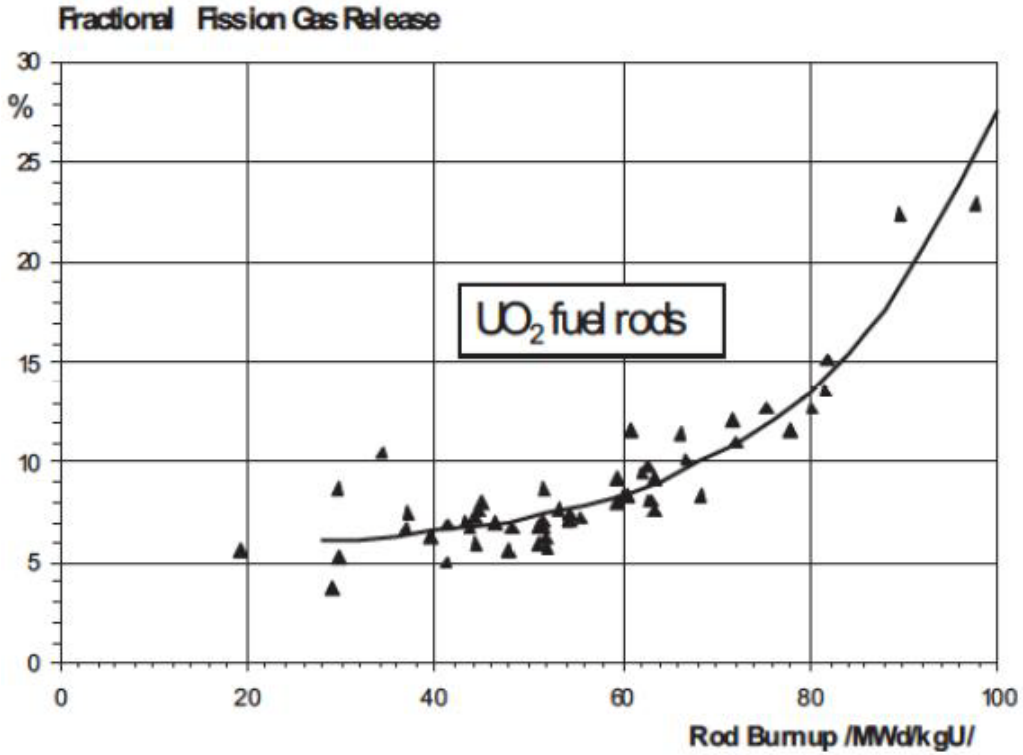
The stress acting on the cladding is an important factor in many of the age related degradation processes that have the potential to impact the performance of the spent fuel during long-term storage. For example, stress corrosion cracking, creep and hydrogen induced degradation require the presence of a tensile stress, and the potential for significant degradation increases as the stress increases. The dominant, non-localized stress on the fuel cladding results from the internal pressure in the fuel pin. This pressure will increase with fuel burnup due to increased fission gas release as suggested in Figure 30.

The fuel pin internal pressure decreases with decreasing temperature and therefore will generally decrease with time during storage. However, the release of fission product gases, including helium and iodine, and the production of helium by alpha decay may occur during dry storage and could increase the internal pressure. There are, however, two factors that demonstrate that cladding stresses due to internal pressurization will not increase significantly during the storage period:

1. the production of fission product gasses will have essentially ceased when the fuel was removed from the reactor and the most of the release of these gasses from the fuel into the fuel pin interior occurred before the onset of dry storage, and
2. the buildup of helium by alpha decay will not be significant until after thousands of years of storage.

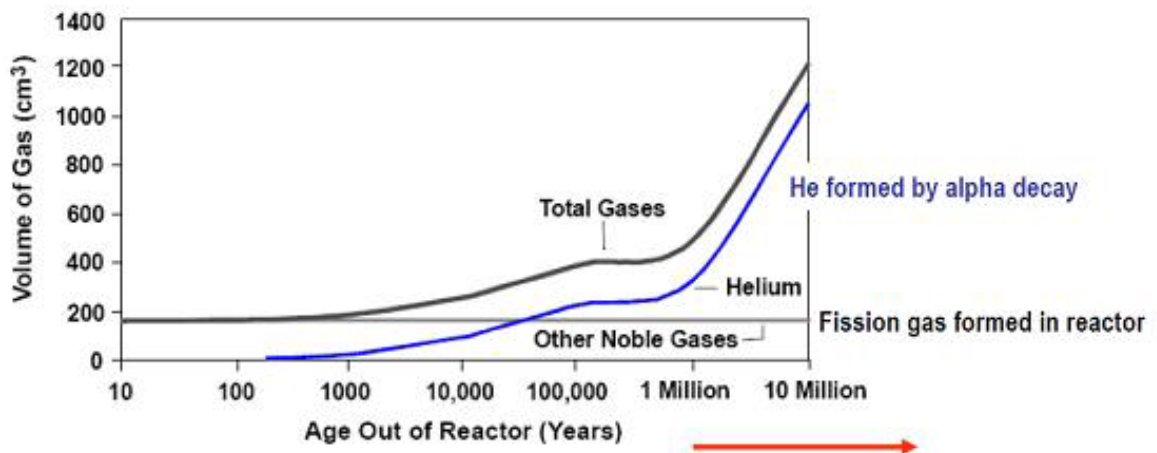
These two factors, coupled with the decreasing temperatures within the dry storage cask system, lead directly to the conclusion that changes in helium pressurization of spent fuel pin in dry storage will not be significant during a 300-year storage period. This conclusion is consistent with the information presented in Figure 31 for CANDU fuel which illustrates that no significant increase in gas volume occurs until approximately 800-years of storage.





**Figure 30** Fission gas release from  $UO_2$  fuel with burnup [Figure reproduced from ref. 1].

## BUILD UP OF HELIUM GAS PRESSURE DUE TO ALPHA DECAY PROCESSES IN THE FUEL



**Figure 31** Helium Build-up in Fuel Pin (CANDU Fuel) [Figure reproduced from ref. 2].

## **Section 2.2.8 References**

- [1] IAEA Nuclear Energy Series, No. NF-T-3.8, Impact of High Burnup Uranium Oxide and Mixed Uranium-Plutonium Oxide Water Reactor Fuel on Spent Fuel Management, IAEA, 2011.
- [2] UNENE, McMaster University, Hamilton, Ontario, CANADA.

## **2.2.9 Creep and Diffusion Controlled Cavity Growth (DCCG)**

Diffusion-Controlled Cavity Growth is a micromechanism for creep degradation of the fuel cladding. Coble creep and other creep mechanisms that rely on atom migration by low activation energy diffusional processes can also cause measurable creep at temperatures below  $T_m/2$ . Studies with zirconium based alloys suggest that the lower temperature limit for significant creep in zirconium based alloys at realistic stress levels is approximately 350°C. Therefore, because the temperature decreases continually during dry storage, creep is not anticipated to produce significant strain after the initial storage period. However, there are significant alloy-to-alloy differences in creep rates (for example low tin SRA Zircaloy 4 creeps twice as fast as regular RXA Zircaloy 2) and estimates of creep strains during the initial storage period should be based on data and analysis directly applicable to the cladding alloy of concern and data gaps certainly exist in this area.

Creep processes will provide characteristic microstructural evidence that cladding damage is occurring prior to the onset of failure. However, collection of such evidence requires destructive evaluation of the material of interest. SEM examinations of the fracture surfaces and cross sections of the near fracture surfaces of the zirconium alloys that were undergoing creep revealed extensive cavitation damage. These results, combined with an examination of the initial material condition and the condition of samples during steady-state creep, suggest that failure of Zircaloy under conditions relevant to the dry storage of spent nuclear fuel occurs by a cavity nucleation, growth, and interlinkage mechanism [1].

Chin and Gilbert [2] constructed a fracture mechanism map based on Ashby from which they concluded that creep under the initial dry storage conditions would occur by diffusion controlled cavity growth (DCCG). They used the Raj and Ashby [3] model for DCCG fracture mechanism to predict the fracture lifetimes for the anticipated temperature–time variations during the initial period of dry storage. However, the acceptance of this model is not universal.

Zircaloys are generally considered to be resistant to cavity formation and there is limited evidence of creep-induced grain boundary cavity formation in unirradiated or irradiated Zircaloy. Keusseyan et al.'s [4] work does show evidence for cavity formation and a potential for creep induced damage.

The vast majority of the literature, however, clearly shows that cavity formation is unlikely in Zircaloy clad spent fuels. Even high neutron radiation exposures do not provide evidence of voids or cavities in Zircaloys. This behavior is unlike that of stainless steels and other metals which show extensive evidence of creep-induced voids and cavities. Many investigators attempted to find cavities and reported black-dot

microstructures comprising dislocation loops [5]. Yoo [6] presented a model based on the anisotropy of Zircalloys to rationalize the absence of voids and cavities.

Based on the proposal by Chin and Gilbert [2], many investigators made detailed calculations assuming the predominance of DCCG. However, Pescatore and Cowgill [7] point out the probable lack of applicability of this model to creep in dry storage based again on the lack of sufficient experimental evidence for cavities in deformed Zircaloy [8].

#### **Assessment of DCCG Degradation in Fuel Subject to Very Long-Term Storage -**

The pressure loading on the cladding of the pin-design fuel is the driving load for clad creep. Creep can be significant at high temperatures and/or over long time periods at moderate temperatures with moderate to high stresses [9].

The fission gas inventory in a fuel rod increases proportionally with burn-up, while the decreasing fuel thermal conductivity and changes of fuel microstructure reduce the ability of the fuel pellet to retain the fission gas. This, together with the rod void volume reduction caused by cladding creep-down during reactor operations and fuel pellet swelling due to irradiation, increases the potential for high rod internal pressures with increasing burn-up [9]. Therefore, although creep damage is not anticipated, the potential for creep, especially when high burn-up fuels are stored and during high temperature processing such as dry out anneals is considered in the present technical bases for DCSS. However, potential creep concerns are basically related to the initial storage period and are not anticipated to be significant during long term dry storage.

#### **DCCG Degradation Data Needs for EST and General Recommendations for**

**Resolution** - The degradation mechanism of DCCG, if operative would cause local rupture of the cladding on fuel rods. This failure phenomenon is preceded by creep deformation and the driving force for deformation in the cladding is reduced as creep deformation takes place, as discussed in section 2.2.1. Under EST, creep damage is not considered likely due the low temperatures after 50-60 years of storage and the resulting low driving forces for creep. The recommendations for creep evaluations in section 2.2.1 address DCCG with the additional note to verify the presence or absence of triple boundary cavitation in materials that is thought to have undergone creep.

#### **Section 2.2.9 References**

- [1] Hayes, T. A. and Kassner, M.E., "Creep of Zirconium and Zirconium Alloys," Metallurgical and Materials Transactions A, Volume 37A, August 2006.
- [2] Chin, B.A., Gilbert, E.R., 1989. Prediction of Maximum Allowable Temperature for Dry Storage of Zircaloy-Clad Spent Fuel in Inert Atmosphere. Nucl. Tech. 85, pp. 57–65.
- [3] Raj, R., Ashby, M.F., 1975, Intergranular Fracture at Elevated Temperature. Acta Met. 23, pp. 653–667.
- [4] Keusseyan, R.L., Hu, C.P., Li, C.Y., 1979. Creep Damage In Zircaloy-4 At LWR Temperatures. J. Nucl. Mater. Vol. 80, pp. 390–392, (1979).
- [5] Wolfenden, A., Farrell, K., 1972, On The Question Of Void Formation In Neutron Irradiated Zirconium, Scripta Met. 6, pp. 127–129.

- [6] Yoo, M.H., 1994, Suppression of Void Formation in Zirconium. In: Zirconium in Nuclear Applications, ASTM STP 551, pp. 292–307.
- [7] Pescatore, C., Cowgill, M.G., 1994. Temperature Limit Determination for the Inert Dry Storage of Spent Nuclear Fuel, EPRI Report TR-103949.
- [8] Short-term rupture studies of Zircaloy-4 and Nb-modified Zircaloy-4 tubing using closed-end internal pressurization, Y.Zhou, B.Devarajan, K.L.Murty, Nuclear Engineering and Design 228 (2004).
- [9] Light Water Reactors, ISBN 92-64-02303-8, Nuclear Energy Agency, NEA 6224, 2006.

### **2.2.10 Air Oxidation of the Cladding – Off-Normal Condition**

An off-normal condition that may occur during EST is air-ingress due to failure of canister seals or weldments, or failure of the canister body itself. There is a potential for stress corrosion cracking of the canister (see section 2.6.2) and throughwall cracks will provide a pathway for air ingress as would seal or weldment failure. The ingress of air into the canister will provide an oxygen rich environment in the canister interior. This oxidizing environment may interact with the fuel cladding and, if the cladding is breached, with the fuel pellet. Both fuel pellet and cladding oxidation are strongly dependent on temperature, available oxygen supply and time at temperature.

Excessive oxidation of the Zircaloy cladding combined with an internal stress can potentially breach the cladding. Oxidation of the Zircaloy [1] is a thermally induced process. Dry storage under inert gas conditions leads to no further increase in the oxide layer that existed on the cladding at final discharge from the reactor. Pool storage conditions (water temperature) mitigate the continued growth of the oxide and the absence of oxidizing species in the as-designed DCSS does not enable continued oxidation. However, if the canister is breached while the internal temperature is above 300°C oxidation of the cladding may occur. This oxidation will reduce the effective wall thickness of the cladding and can lead to rupture if the combination of oxygen induced wastage and cladding stress is sufficient. Constitutive relationships for cladding oxidation are contained in reference [2].

Oxidation of the pellet will cause swelling and may cause the cladding to split locally and the fuel pin to fragment (see section 2.2.11). However, cladding oxidation at the low temperature (< 200°C) of EST is not considered significant and any air oxidation impact will be due to oxygen interactions with the fuel pellet.

**Assessment of Air Oxidation of Cladding - Impact to Safety Function** - Extensive cladding failure could result with cladding wall loss through continued oxidation of the cladding with long-term exposure to air at high cladding temperature (>> 300°C); however the occurrence of such oxidation is extremely unlikely during EST because of the low temperatures (< 300°C) of exposure.

**Cladding Air Oxidation Degradation Data Needs for EST and General Recommendations for Resolution** - Oxidation of the fuel cladding in low temperature considerations of EST is not a significant degradation mechanism. An evaluation should be made for air oxidation of fuel in a demonstration program if the fuel is stored at the

off-normal condition of air ingress to provide verification of lack of significant changes to the cladding as a result the event.

### **Section 2.2.10 References**

- [1] Rothman, A. J., "Potential Corrosion and Degradation Mechanisms of Zircaloy Cladding on Spent Nuclear Fuel in a Tuff Repository," Report Attachment 10 to MRB-0418, JUCID-20172, Contract W-7405-Eng-48, Lawrence Livermore National Laboratory, Livermore, CA, 1984.
- [2] Einziger, R.E., Baldwin, D.L., and Pitman, S.G., Data Needs for Long-Term Dry Storage of LWR Fuel, EPRI Report TR-108757, April 1998.

### **2.2.11 Air Oxidation of the Fuel Pellet Material – Off-Normal Condition**

Oxidation of the fuel,  $\text{UO}_2$ , is possible only if the fuel is exposed directly to air in the dry storage cask system. Such exposure is possible as illustrated in section 2.2.10 and a comprehensive review of the mechanisms and kinetics of fuel oxidation has been performed [1]. Typical spent fuels oxidize by first forming either  $\text{U}_3\text{O}_7$  or  $\text{U}_4\text{O}_9$ , phases that are denser than the original  $\text{UO}_2$ . This first transition is accompanied by a net contraction of the fuel, relieving mechanical stress on the cladding. Oxidation typically occurs first along the fuel grain boundaries. Thus, the release of fission gases, such as  $^{85}\text{Kr}$ , to the DCSS relatively early during this transition is possible. Upon further exposure to air, the fuel is oxidized to  $\text{U}_3\text{O}_8$ , a phase which is approximately 36% less dense than the original fuel, thus oxidation ultimately results in swelling of the pellet.

The swelling of the fuel as  $\text{U}_3\text{O}_8$  forms has been shown to supply sufficient mechanical stress to split the cladding [2].  $\text{U}_3\text{O}_8$  formed by oxidation of the fuel is a fine powder that spalls from the fuel surface. The release of fines and/or fuel relocation from split cladding must be evaluated if  $\text{U}_3\text{O}_8$  formation is suspected. The extent of oxidation of irradiated  $\text{UO}_2$  is a time and temperature dependent phenomenon. In addition, the oxidation of spent fuel to  $\text{U}_3\text{O}_8$  has been shown [3] to be burnup-dependent. As burnup increases, the soluble fission products and actinides stabilize the matrix and inhibit the formation of  $\text{U}_3\text{O}_8$ . For example, at 229°C, the reported incubation time for crack propagation due to fuel oxidation was approximately 1000 h for a rod with an assembly average burnup of about 12 MWd/kg U, whereas a rod with an assembly average burnup of about 30 MWd/kg showed no sign of splitting or dilation after 5962 h [2]. At the lower temperatures expected during extended storage, oxidation of even low-burnup fuels will take sufficiently long, at least months or years, that detection and mitigation of the off-normal condition should prevent fuel oxidation from being a concern. At higher burnup levels, under the conditions of extended storage, sufficient oxidation to propagate cladding defects should not occur and no mitigative measures may be needed to protect against pellet oxidation and swelling which might lead to cladding rupture.

The potential for air oxidation of MOX fuels is likely to be higher than that for  $\text{UO}_2$  fuels because the flatter temperature profile during cooling will increase the time that the fuels and cask are at higher temperatures. This increase may promote stress corrosion of the cask and should stress corrosion occur promote oxidation of the fuel. Additionally, the handling difficulties associated with MOX fuel fragments must be evaluated.

**Assessment of Air Oxidation of Fuel - Impact to Safety Function** - There is a potential to load some fuel into the canister that initially has a pin hole type clad breach. Also, during long term storage some degree of clad breach may occur from operating mechanisms like DHC. Coupled with a failure of the canister and air ingress (loss of confinement), there is a potential to have gross clad failures from fuel oxidation. The oxidation process would expand the fuel and stress the cladding and could lead to gross rupture of the cladding. Additionally, the dimensional changes in the fuel oxide may cause a loss of fuel retrievability. The potential air oxidation rates will be coupled with the air ingress rates and will depend on the temperature of the fuel at the time of container compromise, the specific cladding alloy in the compromised container, the fuel type (uranium dioxide or MOX) and the irradiation history. Qualitatively, a loss of confinement provided by the canister and an increase in source term within the canister will result. The data necessary to evaluate the impact of these potential issues are not available; however, this assessment concluded that the impact to safety, if any, will involve an increase in the source term and a loss of retrievability.

**Data Needs for Air Oxidation and Recommendations** - Air oxidation of the fuel and fuel cladding will only occur if the cover gas in the containment system is compromised. Air ingress could occur if corrosion or stress corrosion cracking occurred in the canister or, for bolted closures, if the bolts failed or the seals relaxed. The rate of oxidation will depend on the fuel and cladding temperatures and physical conditions at the time of air ingress. The development of the data necessary to assess these quantities will require extensive resources and the return on investment will be minimal. Therefore, periodic monitoring for cover gas and containment efficacy is recommended throughout EST to assure that the inertness of the cover gas environment and avoid air oxidation.

1. Monitoring/In-Service Inspection Activity: Periodic monitoring for cover gas and confinement efficacy in necessary throughout EST and acceptance criteria should be developed as should remediation plans for action should confinement be lost.
2. Testing & Analysis Activity: Evaluate the potential for MOX fuel pellet oxidation (as was done for UO<sub>2</sub> fuel with burn up in PNNL 11929).

A very small fraction of the cladding on fuels in EST will contain minor breaches and only a small fraction of the canisters are anticipated to allow air ingress. Thus, air oxidation of fuel in EST is not likely but is clearly possible. However, since such oxidation could lead to cladding loading and large ruptures, if left unmitigated for a long time, an inspection protocol and mitigation path forward should be established.

### **Section 2.2.11 References**

- [1] McEachern, R.J., and Taylor, P., "A Review of the Oxidation of Uranium Dioxide at Temperatures below 400°C," JNM 254, 1998, pp. 87-121.
- [2] Einziger, R.E. and Cook, J.A., "Behavior of Breached Light Water Reactor Spent Fuel Rods in Air and Inert Atmospheres at 229°C," Nuclear Technology, Volume 69, 1985, pp. 55-71.
- [3] Hanson, B.D., "The Burnup Dependence of Light Water Reactor Spent Fuel Oxidation;" PNNL 11929, 1998.

### **2.3 Baskets and Neutron Absorbers Description**

Fuel transport and storage canisters incorporate baskets of aluminum alloys, carbon steel, or stainless steel that provide geometric configuration control of assemblies within the canister [1, 2]. The primary function of the basket is to maintain spacing during wet loading and unloading operations and ensure fuel retrievability. The fuel basket also provides structural support for the canister and conduction pathways for decay heat dissipation.

Neutron poisons are added to the fuel canister to provide criticality control. These neutron poisons are absorber plates that are held in place by the fuel basket or are integrated into the metallic basket matrix material. Criticality control within fuel canisters is required only during canister loading and unloading operations conducted in reactor spent fuel pools [3] and during transportation in the event of an accident that could lead to flooding. During dry storage, the inert atmosphere of the canister provides insufficient moderation to support criticality and subcriticality is maintained by moderator exclusion.

There are several neutron poisons that are in use in licensed dry storage casks. These include the following:

#### **Aluminum Boron Carbide Cermets: BORAL<sup>®</sup>**

BORAL<sup>®</sup> is widely used as a neutron absorber in fuel pool storage racks and in some dry storage casks. “Boral<sup>®</sup> is made by mixing boron carbide granules and aluminum powder inside an aluminum box, heating the box and its content to form an “ingot”, and then hot-rolling the “ingot” to form a plate consisting of a coarse core of B<sub>4</sub>C-Al composite material bonded between two thin sheets of aluminum cladding” [4]. The application of BORAL in basket designs is the predominant component for criticality control during fuel retrieval in basins. The BORAL<sup>®</sup> remains with the fuel in dry storage and is expected to be present for fuel retrieval, as necessary. BORAL<sup>®</sup> has excellent boron loading capabilities and is a very effective neutron absorber, but has very poor mechanical and thermal performance.

#### **Metal matrix composites: Metamic<sup>®</sup>, Bortec<sup>®</sup>**

Metamic<sup>®</sup> is a fully-dense, discontinuously-reinforced, metal matrix composite material. It consists of high-purity Type 6061 aluminum (Al 6061) alloy matrix reinforced with Type 1 ASTM C-750 isotopically-graded boron carbide (B<sub>4</sub>C). The boron carbide loading can be varied and includes concentrations up to 33% by weight. Metamic<sup>®</sup> is used as a neutron absorber in wet and dry storage of used nuclear fuel. Metamic is weldable and has favorable mechanical properties up to 400°F (204°C), making it suitable as a structural material in fuel storage racks and in low heat emitting fuel baskets in dry storage casks [5]. The material is produced at essentially maximum theoretical density with no internal interconnected porosity.

BORTEC<sup>®</sup> MMC is produced in rolled or extruded shapes and in B<sub>4</sub>C contents up to 32% by weight using various 5000, 6000, and 1000 series aluminum alloys as matrix materials. The resulting composites have outstanding properties for structural or non-structural design applications, and are lightweight and adaptable for neutron capture in both wet and dry environments. BORTEC<sup>®</sup> MMC is produced with no interconnected porosity and is used in high areal density neutron absorber applications.

### **Borated Aluminum Alloys: BorAluminum®**

BorAluminum® alloys are true alloys developed by incorporating boron as a second phase in standard aluminum compositions using alloys 1100 and 6351. BorAluminum® alloys are used in several spent fuel storage and transportation applications. Non-structural applications typically use 1100 alloy + boron, while applications requiring structural alloys most frequently use 6351 + boron that maintains the strength and thermal conductivity of a standard alloy.

### **Borated Stainless Steels: NeutroSorb® Plus, Neutronit®**

NeutroSorb Plus is a sintered borated stainless steel material, typically ASTM A887 Types 304B3 through 304B6 [6] while Neutronit® is an austenitic stainless steel containing boron. Neutronit can be characterized as a 316 stainless steel with a boron addition. Both borated stainless steel alloys have good mechanical properties and fabrication capabilities, but are challenged by limited boron loading capability [7].

### **Section 2.3 References**

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- [5] Personal communication, J. Russell, Sales and Marketing Manager, Holtec International, to M. E. Dupont, September, 2010.
- [6] Pasupathi, V., Waste Package Neutron Absorber, Thermal Shunt, and Fill Gas Selection Report, B00000000-01717-2200-00227 REV 00, January 2000.
- [7] White Paper on the Proposed Design, Development, and Implementation of a Monitored Retrievable Storage Module and the Siting Criteria for Spent Nuclear Fuel, B. Villarreal and D. Knobloch, Los Alamos National Laboratory, LA-13001, UC-721 January 1996.

### **2.4 Baskets and Neutron Absorbers Degradation Phenomena**

Degradation of the fuel baskets could impact the ability of the fuel baskets to maintain the geometry of fuel assemblies, neutron poison plates, and integral neutron absorbers. Deformation of the baskets could cause fuel assemblies to physically bind within the basket, thereby impacting retrievability of the spent fuel assemblies. Creep is one



mechanism potentially leading to basket deformation. Additionally, should the storage canister become breached (see section 2.2.10 for example) air ingress could lead to oxidation of basket material.

For metallic materials, creep generally only becomes important for homologous temperatures greater than  $0.4T_m$  where  $T_m$  is the absolute melting temperature [1]. Though carbon steel and stainless steel would not be subject to creep at expected service temperatures and conditions, the temperature within the canisters may be above the temperature at which aluminum baskets experience creep.

Fuel baskets may experience limited corrosion associated with residual moisture within the canister as a result of incomplete drying or waterlogged rods. This residual water would be consumed very early in the storage period through corrosion processes, after which time, the corrosion would cease. Corrosion of the fuel baskets by corrosion induced by residual water is expected to be higher for the aluminum and carbon steel baskets than for stainless steel baskets. Because this is a phenomenon that is limited to the early storage period, there is no additional impact to basket structural integrity for very long-term storage by corrosion of fuel baskets. However, air ingress into a breached canister could lead to continued oxidation of the basket material. This may be particularly significant to the aluminum based basket material and, after prolonged exposure to elevated temperature air the aluminum in the assembly may be converted to aluminum oxide. Such conversion would degrade the mechanical properties of the aluminum based materials to the point that structural integrity could not be assured and the basket/poison plate material may lose dimensional stability. This would impact the retrievability of the fuel and could impact re-immersion of the DCSS, if needed.

Degradation of the fuel basket would affect its thermal characteristics. Thinning of the baskets walls by corrosion will decrease the cross section of the conductor and therefore decrease the heat conducted by the basket. Deformation of the basket material will increase the defect concentration in the material and decrease its thermal conductivity. Oxidation of the basket surface will affect the emissivity of the basket surface, thereby impacting the radiative heat transfer characteristics within the canister.

Neutron poisons that are fixed within the fuel basket, in conjunction with soluble neutron absorbers, provide criticality control during flooding associated with loading and potential unloading operations in a spent fuel pool. During the storage period, fixed neutron poisons are not necessary to maintain criticality control, except for their contribution to moderator exclusion. The degradation of fixed neutron poisons within the canister may specifically impact retrievability of spent fuel assemblies by physically impeding the removal of spent fuel assemblies from the canister or by redistribution of neutron absorbers within the canister leading to a decreased effectiveness as a neutron absorber during flooding.

BORAL<sup>®</sup> is used extensively in dry spent fuel storage applications. However, there are indications that BORAL<sup>®</sup> could form blisters between the boron carbide matrix and the aluminum sheet cladding as a result of infiltration of spent fuel pool water into the porous boron carbide matrix. The blisters may likewise develop during vacuum drying of the casks by vaporizing water which had infiltrated into the plates or from corrosion during long-term storage at temperatures on the order of 500°F (260°C). Under these conditions, it is possible the material may crumble or otherwise relocate in storage, or may be physically damaged by reflooding associated with retrieval operations [2].

Similar degradation may result if a canister with BORAL became breached during EST. Data are needed to demonstrate that aluminum clad/boron carbide absorbers with core porosities between one and three-percent will not develop blistering that impacts the effective multiplication factor during cask reflooding associated with cask retrieval (or loading) operations [3] and during transportation in the event of an accident that could lead to flooding. Similar data are needed to assess the performance of BORAL in a breached dry storage system. The effect of the degradation on criticality control will depend on the physical condition and location of the neutron poison material, thus, calculations of the effects require significant post storage information. To eliminate the expenditure of the resources needed for the collection of such information, it is recommended that, fuel retrieval be achieved dry-to-dry or by flooding the canister with a fluid such as borated water.

The metal matrix composites, Metamic<sup>®</sup> and Bortec<sup>®</sup>, are not subject to blister formation like the aluminum clad/boron carbide absorbers, because these composites are produced with a homogenous material at effectively the maximum theoretical density, and with “no interconnected porosity in which corrosion can occur causing hydrogen generation, internal pressure buildup and deformation of the matrix” [4]. Likewise, the borated aluminum and stainless steel poison materials are not subject to the mechanisms of blister formation that have been observed in BORAL<sup>®</sup>. Because of the general lack of ductility characteristic of clad absorbers, similar to BORAL<sup>®</sup>, there is a need to evaluate the potential for embrittlement and cracking, and associated change in reactivity, of clad neutron absorber plates due to thermal cycling of the canister during the drying, storage, and retrievability period. However, Bortec should be evaluated for its performance in a breached canister.

Exposure of neutron absorber materials to neutron radiation generated within a spent fuel storage cask during the service life of the cask is expected to produce negligible effects on the efficacy of the neutron absorber. The flux of neutrons within the canister is decreasing with storage time and the total neutron fluence, even after extended storage, is insignificant compared to the number of absorber atoms in the neutron absorber material. The effect of extending the storage times will, therefore, not affect the susceptibility of the absorber materials to absorber isotope depletion or radiation induced embrittlement or cracking.

**Assessment of Degradation, Impact to Safety Function** – Excessive creep deformation of the aluminum basket and absorber casing materials would potentially impact the retrievability of the fuel from the canister as would excessive oxidation in a breached canister. Even though the temperature of the fuel basket would decrease with storage time, creep is viable for aluminum baskets and neutron absorbers due to relatively high temperatures expected in the fuel basket relative to the homologous temperature of  $0.4T_m$  for creep in aluminum, or approximately  $100^{\circ}\text{C}$ . Additionally, the long term (100+ years) performance of aluminum alloys at anticipated EST temperatures is not known. Deformation and slump analyses would be needed on a structure-specific basis to evaluate and demonstrate that creep does not impair load-bearing components of aluminum fuel baskets to perform their safety functions during very long-term storage. Similar analyses are needed to assess performance in breached canisters.

**Basket/Absorber Data Needs and Recommendations:** The neutron absorbers should be evaluated for retrievability of fuel and efficacy of absorber material after water/dryout cycling and long-term exposure to air at anticipated storage temperatures. Deformation and slump analyses are routinely done in the initial SAR and need to be extended, on a structure-specific basis to demonstrate that creep and oxidation, time dependent processes, do not impair load-bearing components of aluminum fuel baskets to perform their safety functions during very long-term storage.

1. Testing & Analysis Activity: Because of the general lack of ductility characteristic of clad absorbers, similar to BORAL<sup>®</sup>, there is a need to evaluate the potential for embrittlement and cracking, and associated change in reactivity, of clad neutron absorber plates due to thermal cycling of the canister during the drying, storage, and retrievability period. The impact of long-term exposure to air should also be evaluated.
2. Testing & Analysis Activity: Evaluate basket-specific designs of aluminum baskets for slump over EST.

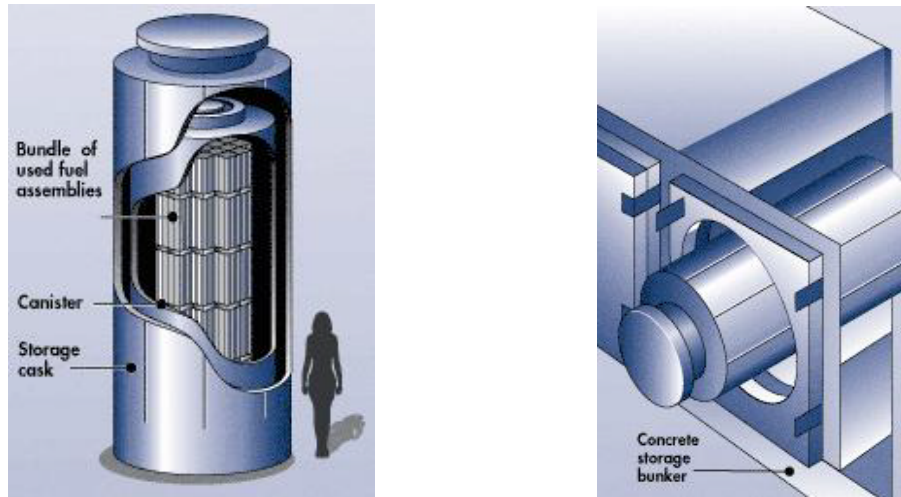
Basket degradation during EST and the impact of the water/dryout degradation for BORAL has been addressed, and the creep damage of aluminum basket materials is not anticipated to have a high negative impact on retrievability since engineering methods could be developed and implemented to free a slumped basket from a canister or remove fuel that may be stuck in a basket. It is not anticipated that reconfiguration of the basket would be severe to violate criticality safety during re-immersion in a pool, if necessary. However, the impact of long-term exposure to air at anticipated storage temperatures should be established.

### **Section 2.4 References**

- [1] Cadek, J., Creep in Metallic Materials, Elsevier Science Publishing Company, Inc., 1988.
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### **2.5 Canisters and Weld Description**

A variety of DCSSs have been approved by the NRC for storage only, and, if approved, for both storage and transport, the DCSS is termed a dual-purpose dry storage system [1, 2]. The two basic configurations for storage canisters in a DCSS are shown in Figure 32.



**Figure 32** Typical Dry Cask Storage Systems. The cask may be an integrated cask with confinement and shielding in an integrated structure. The dry storage cask may include a separate canister (dual-purpose canister) over which an overpack is placed to provide shielding for storage or a cask is placed for transportation [<http://www.nrc.gov/waste/spent-fuel-storage/diagram-typical-dry-cask-system.html>].

The weld-sealed canister is the component of the DCSS that constitutes the confinement boundary and is essential for fuel handling and retrievability in the DCSS design with a canister and overpack that includes an air gap for passive cooling between the canister and overpack. The canister is sometimes termed a “multi-assembly sealed basket” or “multi-purpose canister” (MPC) in DCSS designs [2]. A cask with a metallic gaskets and bolted lid closure constitutes the confinement boundary in a DCSS design of an integral basket and cask.

For transportation, the shipping cask or overpack is credited with a defined leakage rate and is the primary confinement boundary. In some cases, the MPC is desired to provide moderator exclusion during transportation, and therefore it must continue to remain as a leak-tight component in the transportation package.

The canister (MPC) is typically weld-sealed as illustrated in Figure 33. Also, the canister is constructed from rolled plate, with a typical plate thickness of 0.5 inches (1.3 cm). The plate is rolled into a cylinder and seam welded. Two or more cylinders are stacked and joined to form the canister, and a bottom plate is welded at the canister base.

The cask, as an integrated container with confinement and shielding, is typically closed with a bolted, gasket seal.

Several materials are used for construction of the canister and dry storage casks including [2]:

- Austenitic stainless steel
- Carbon steel (coated for corrosion protection)

- Ductile cast iron (coated for corrosion protection)

The shell and bottom plates are joined by welding and the joined plates may be one to several inches thick [2]. The carbon steel and ductile cast iron canisters use a bolted closure with metallic gasket seals. The austenitic stainless steel canisters are closed with either a double seal weld or bolted closure with two concentric gaskets.

Most of the dry storage canisters that are placed in concrete overpacks are fabricated from austenitic stainless steels [2]. In general, the canisters will be marginally protected from the environment but will be exposed to the storage site atmosphere. As discussed in Section 2.6, pitting and stress corrosion cracking are the only expected type of degradation anticipated in canisters during EST.

### **Section 2.5 References**

- [1] Source: U.S. Nuclear Regulatory Commission, <http://www.nrc.gov/waste/spent-fuel-storage/designs.html>.
- [2] Industry Spent Fuel Storage Handbook, EPRI Report 1021048, July 2010.

Closure Weld Sealed

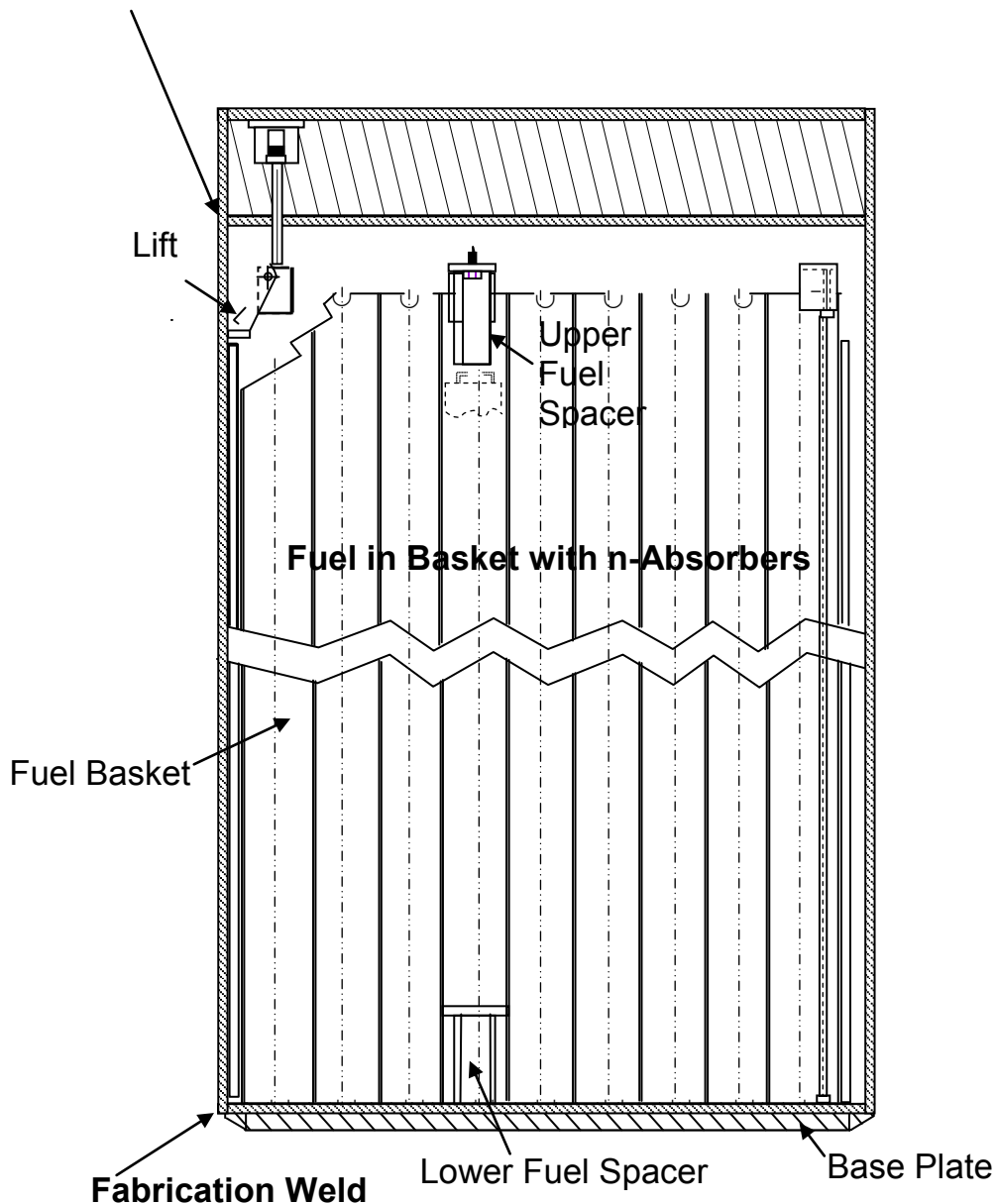


Figure 33 Generic Canister Sketch Showing Basket Internals.

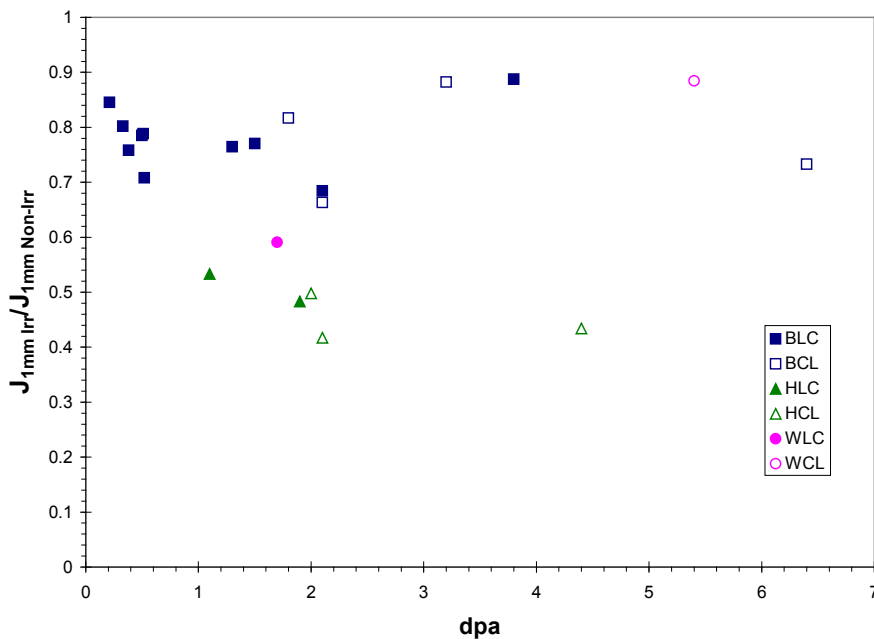
## 2.6 Canister Degradation Phenomena

The storage environment (external) for the canister in EST will include mildly elevated temperatures, ambient air, low irradiation doses, and the ‘industrialized’ atmosphere of the storage site. A gamma field of  $\sim 10^5$  R/hr and a neutron flux ( $E_n > 1$  MeV) of  $10^4$ - $10^6$  n/cm<sup>2</sup>-s [1] are typically assumed [2] for the irradiation field of a DCSS for extended storage, but the gamma field would be expected to decrease with age of the fuel and the increase with burnup. The radiation fields for the MOX fuel on a per fuel assembly (FA) basis, as discussed in section 2.1.5, would be above that for UO<sub>2</sub> fuel. Irradiation from

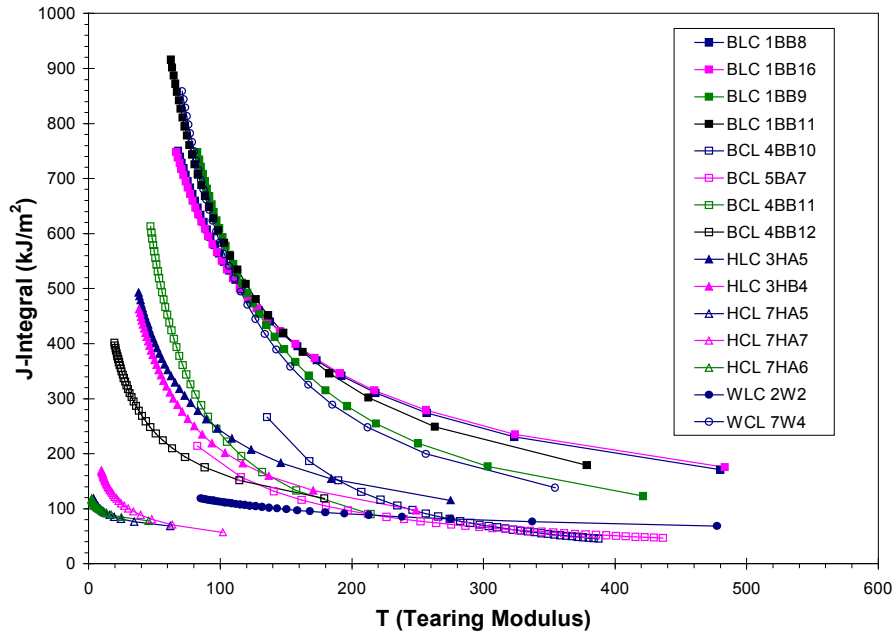
spontaneous fission neutrons and/or ( $\alpha$ , n) neutrons will not be significant to impact the properties of the canister. The very low dose, even after several hundred years of exposure, is insignificant compared to dose levels needed to manifest changes to the mechanical properties at the low temperature of irradiation. Even then, high residual fracture toughness levels are observed in the canister materials [3].

The stainless steel canisters would not suffer significant radiation damage over EST. Figure 34 shows the dose dependence of the normalized (unirradiated/irradiated) fracture toughness for Base (B), Heat-Affected-Zone (H), and Weld (W) materials as obtained from Compact Tension (C(T)) specimens oriented with the notch/crack parallel (CL) or perpendicular (LC) to the axis of the large diameter pipe (rolling direction of the initial plate) source material. Figure 35 shows the irradiated fracture toughness of the materials in the form of J-T curves. The lowest toughness was in the heat-affected zone region when a crack ran parallel to the rolling direction of the original plate used to fabricate the large diameter piping; this toughness remained high even up to dose levels much higher than anticipated in the canisters.

Thermally-induced changes in the canister material are also not plausible due to the low temperatures anticipated for EST.



**Figure 34** Residual Fracture Toughness After Low Temperature ( $T_{irr} \sim 150^\circ\text{C}$ ) Neutron Irradiation on Type 304/304L Stainless Steel and Type 308 Stainless Steel Weld Metals [Figure reproduced from ref. 3].

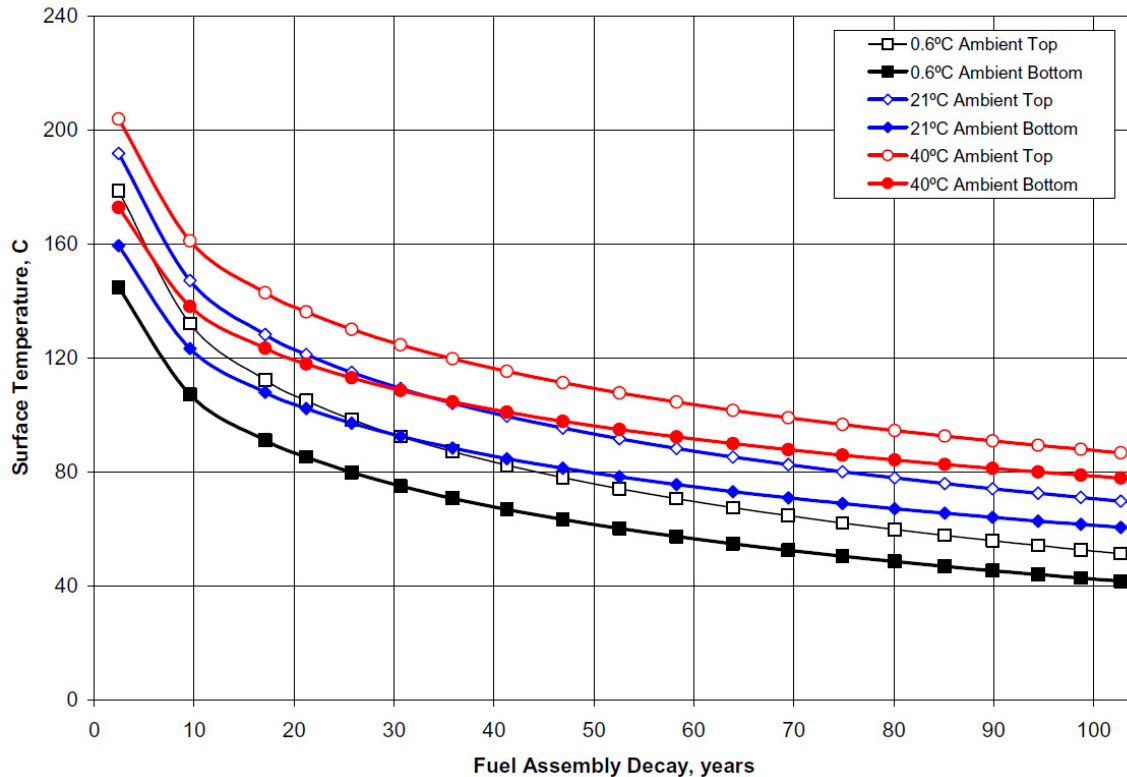


**Figure 35**  $J_{\text{deformation}}-T$  Curves for Base (BLC, BCL), Weld (WLC, WCL), and HAZ (HLC, HCL) Components of Low-Temperature-Irradiated ( $T_{\text{irr}} \sim 150^{\circ}\text{C}$ ) Type 304 Stainless Steel. The CL crack orientation is parallel to the rolling direction of the plates used to fabricate the stainless steel tank [Figure reproduced from ref. 3].

The fuel canister is generally exposed to the external environment at slightly elevated temperatures due to the heat generated by the stored fuel assemblies. The surface temperatures of the canister depend on the temperature of the surrounding environment, the canister heat load and the design of the overpack system providing for canister cooling.

The temperature-time profile illustrated in Figure 36 provides an example of possible conditions at the surface of a spent fuel storage canister and demonstrates that the surface temperatures are in the regime for corrosion induced degradation of the canister but are too low to cause significant metallurgical changes. However, canister surface temperature vs. time data for specific applications should be determined using actual canister heat loads and atmospheric conditions because, as discussed below, time – temperature profiles will impact the tendency for corrosion.





**Figure 36** Example of Canister Surface Temperature vs. Time  
[Figure reproduced from ref. 4].

Austenitic stainless steels are susceptible to pitting, crevice, and stress corrosion cracking under conditions anticipated during EST. The tendency for cracking when exposed to marine environments is dependent on salt chemistry and content, temperature, relative humidity and time of exposure [5, 6, and 7]. Additionally, chloride leaching from paints, adhesives, tapes and labels is known to cause cracking of such steels at room temperature [8].

### **Section 2.6 References**

- [1] Johnson, A.B. and Gilbert, E.R., "Technical Basis for Storage of Zircaloy-Clad Spent Fuel in Inert Gases," PNL-4835, Pacific Northwest National Laboratory, 1983.
- [2] Einziger, R.E., Baldwin, D.L., and Pitman, S.G., "Data Needs for Long-Term Dry Storage of LWR Fuel," Electric Power Research Institute report TR-108757, April 1998.
- [3] Sindelar, R.L., Lam, P.S., Duncan, A.J., Wiersma, B.J., Subramanian, K.H., and Elder, J.B., "Development and Application of Materials Properties for Flaw Stability Analysis in Extreme Environment Service," invited paper in Engineering Safety, Applied Mechanics & NDE – A Symposium in honor of Dr. Spencer H. Bush, Proceedings of ASME Pressure Vessels and Piping Conference, San Antonio, TX, Paper No. PVP2007-26660, 2007.

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### **2.6.1 Corrosion of Canister Materials**

If the storage conditions were truly dry, corrosion, which is an electrochemical process, would neither degrade the materials nor compromise the intended safety function of a system or component. Corrosion can only occur in the presence of an electrolyte and in a dry storage system the electrolyte is anticipated to be moisture (water) that has condensed or fallen (rain) on the external surfaces of DCSS components. Initially, the materials selection processes were based on a license period of twenty years and the system design life, although not formally stated, was approximately fifty years. The radiation levels and temperatures of material exposure decrease with time and the initial exposure conditions were generally considered to be the most aggressive. Therefore, if no significant degradation had occurred during the initial storage period, extension of the "design life" and license period was thought to be relatively straightforward. This thought is basically reflected in ASTM C1562-10 [1] which states that "--- the demands on materials properties for an additional 20 to 80 years of storage may be reduced ---". The review also concluded that "corrosion of metal components exposed to air and humid environments will occur." The outer surfaces of the canister, with its welds, seals and attachments, therefore represent the area most likely to experience corrosion induced degradation during extended dry storage.

### **Section 2.6.1 References**

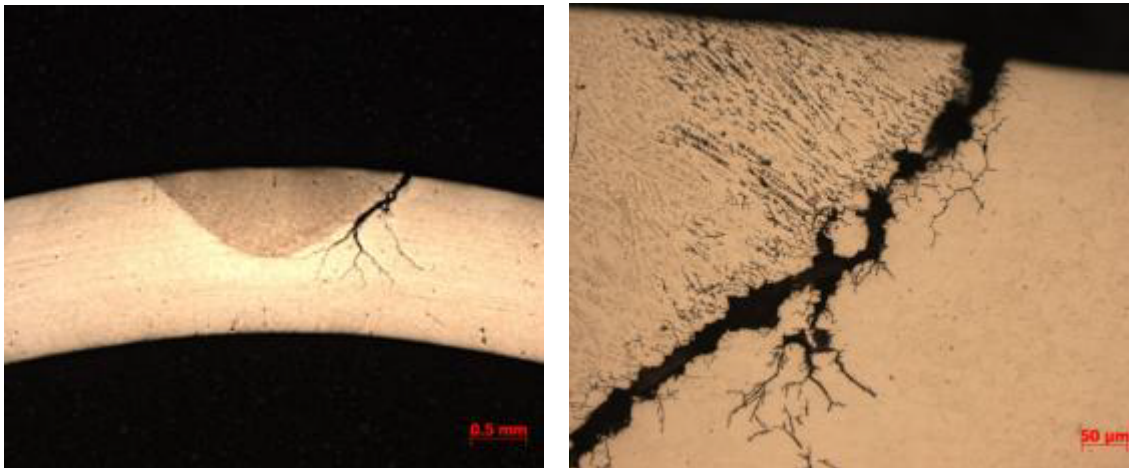
- [1] ASTM C-1562-10, *Standard Guide for Evaluation of Materials used in Extended Service of Interim Spent Nuclear Fuel Dry Storage Systems*, American Society for Testing and Materials, reapproved 2010.
- [2] Technical Bases for Extended Dry Storage of Spent Nuclear Fuel, EPRI report number 1003416, Final Report, December 2002.

## **2.6.2 Stress Corrosion Cracking and Pitting of Austenitic Stainless Steel Canisters**

The austenitic stainless steel canisters are anticipated to experience corrosion-induced degradation during long-term dry storage. Stress corrosion cracking at welds is probable, especially in canisters exposed to marine environments and both stress corrosion cracking and pitting are anticipated at non-weld regions. Additionally, if crevices that intersect the canister surface are present, crevice corrosion may occur.

Canisters constructed from austenitic stainless steels (Types 304 and 304L) will be susceptible to stress corrosion cracking at ambient temperatures in EST. Legend formally held that stress corrosion cracking of austenitic stainless steel did not occur at temperatures below 40°C (the legend was so strong that 1970's versions of ASM Handbooks published this as a lower temperature limit for SCC) but experience has now demonstrated that cracking occurs at room temperature. Welded tanks, forged rings for climbing equipment, bolts and piping made of austenitic steels have cracked during room temperature service.

Recently, a welded, Type 304L stainless steel coupon test as part of a qualification program for plutonium storage containers developed stress corrosion cracks when exposed, at room temperature, to salt mixtures that contained only about 0.6 wt% moisture [1]. The stress corrosion cracks initiated in the heat affected region of the weld and propagated along the HAZ and into the base metal (Figure 37). Such observations are clearly relevant to the welded canisters placed in dry storage.



**Figure 37** SEM Micrographs of Stress Corrosion Crack in the Heat-Affected Zone in a Type 304L Stainless Steel Coupon with an Autogenous Weld. The specimen was exposed, at room temperature, to salt mixtures that contained about 0.6 wt% moisture [SRNL Micrograph].

Stress corrosion cracking involves the synergistic action of three variables: a tensile stress, a susceptible material, and a semi-specific environment (chloride ions on austenitic stainless steel for example). Remove one of the three variables and cracking will not occur.

The tensile stress necessary to cause stress corrosion cracking may be either residual or applied. The residual stresses developed by operations such as forming, welding, and grinding are more than sufficient to support cracking in austenitic stainless steel. This is illustrated in Figure 38, which shows multiple throughwall cracks in the bottom of a welded, stainless steel container. There were no applied stresses acting on the container during the cracking process.



**Figure 38** ThroughWall Stress Corrosion Cracks in a Welded Stainless Steel Container not Subjected to Applied Stresses [SRNL Micrograph].

The chloride ions and the humidity necessary to cause cracking in a Type 304 stainless steel container in a DCSS may come from a variety of sources. The most apparent source of chlorides is a sea coast environment. The importance of humidity to the cracking process is apparent from studies [1] that demonstrated the necessity of deliquescence to the cracking process. These studies were conducted in sealed containers and showed that no cracking would occur in the absence of deliquescence.

This observation is consistent with more recent results from in salt fog tests on U-bend specimens which showed that all the 304 and 304L stainless steels maintained at 43°C experienced cracking while all specimens tested at 85 and 120°C showed no SCC [2]. These observations, coupled with time-temperature profiles shown in Figure 36, suggest that the austenitic stainless steel dry storage canisters may be more susceptible to SCC during long term storage than during the initial storage period. The high surface temperatures during the initial storage period could prevent deliquescence of the salt and therefore prevent cracking of the canister.

The elements necessary to cause stress corrosion cracking to initiate on the external surfaces and propagate through an austenitic stainless steel component will be present during long-term dry storage and the tendency for cracking may increase with time. Furthermore, stress corrosion cracking is a nucleation and growth process, thus the absence of cracking at time  $t$ , provides no assurance that cracking will not occur at time  $t +$  one day. The potential for SCC to produce a throughwall penetration in a dry storage canister provides a potential for oxygen exchange with the helium environment and could adversely impact the performance of the fuel cladding. If the cladding has been breached, the fuel pellets would be exposed to oxygen and could be oxidized and swell.

Pitting and/or crevice corrosion could also occur on the canister if an electrolyte develops on the canister surface. Both pitting and crevice corrosion could lead to a throughwall breach in the canister.

Although the preceding discussion mentioned a sea coast environment as a potential source of chlorides, other stress corrosion cracking causing species may contact the external surface of the canister. Figure 39 shows a pit and an associated stress corrosion crack that resulted from fly ash and the associated chlorides depositing on the surface of a weld-closed stainless steel drum. Industrial pollutants including flyash are well recognized to have the potential to cause SCC. Although the applied stresses on dry storage canister walls are minimal, residual stress (fixed displacement stresses) are clearly present and likely to be driving force for the cracking process. This was the case for the cracks shown in Figure 39.



**Figure 39** Pit and Stress Corrosion Crack due to Fly Ash Contact with Austenitic Stainless Steel [SRNL photograph].

The cracks shown in Figure 39 are typical of cracks driven by residual or fixed displacement stresses. Such cracks tend to be tight and when the crack propagated from the outside in, the tightness of the crack increases as the inner surface of the container is approached.

**Conditions for Pitting/Cracking on Stainless Steel Canisters:** Aggressive ionic species and water are required for cracking of the austenitic stainless steel canisters. It is assumed that surface tensile stresses are present due to weld residual stresses or fabrication-induced stresses and the material is known to be susceptible to SCC.

Recently, key work has been conducted on conditions for stress corrosion cracking and pitting susceptibility of the canisters in a dry storage system [2]. Welded and unwelded, single and double U-bend specimens of Types 304, 304L, and 316L stainless steel were subjected to two types of environmental tests, salt spray and salt fog, using simulated sea salt. Complementary tests with NaCl and MgCl<sub>2</sub> salt droplets were also conducted. All specimens exhibited pitting and cracking within a 2-month exposure in the salt spray tests that included heated specimens at temperatures of 25°, 93°, and 176°C. In the case of salt fog test, all specimens showed cracking at the specimen test temperature of 43°C whereas specimens tested at 85 and 120°C showed no SCC, consistent with the inability of the salt to deliquesce.

Experimental and modeling work (high temperature Pitzer ion interaction thermodynamic data base model) were performed to evaluate salt deliquescence for repository dust assemblages [6]. Specific to the system, the modeling results predict that mixtures of NaCl, KNO<sub>3</sub>, and/or NaNO<sub>3</sub> can show deliquescence when the relative humidity is as low as 20% at 160°C. Reversed deliquescence experiments were conducted in NaCl-NaNO<sub>3</sub>-H<sub>2</sub>O and KNO<sub>3</sub>-NaNO<sub>3</sub>-H<sub>2</sub>O systems at 90°C to 120°C to determine relative humidity and solution composition. The modeling results were in moderate disparity at high temperatures (120°C) with the experimental results – the model underestimated relative humidity by as much as 8% and solution composition by as much as 50% in the KNO<sub>3</sub>- NaNO<sub>3</sub>- H<sub>2</sub>O system.

Two important observations are made: 1) salt assemblages can have a lower deliquescence relative humidity than single-component systems, and 2) assemblages can form brines above 100°C and at standard atmospheric pressure of 1.01325 bar. Although the deliquescence relative humidity of most pure salt minerals is known over a range of temperature, the behavior of salt mixtures at elevated temperatures is largely unknown.

With an assumed presence of marine salts or industrial pollutants such as flyash on a canister surface during EST, it is concluded that the stainless steel canister is susceptible to pitting and cracking when canister temperatures are below 100°C and that cracking can occur at more elevated temperatures if exposure conditions allow deliquescence of the salts.

**SCC Growth Rates – Salt Deposits:** Work done by CRIEPI on SCC growth in marine environments is included in reference [5]. This work, coupled with thermal analyses of temperature profiles anticipated in canisters (Figure 40) indicate that the canister could be at temperatures that readily support SCC (at 100°C) within several decades of storage. Test results for 3-point bend specimens such SCC crack growth rates for salt deposit conditions (Figure 41) are high, approximately up to 10 cm/year. These data suggest that once cracking is initiated, the time for a crack to grow through the wall of a 0.5 inch thick canister will be within a one year timeframe.

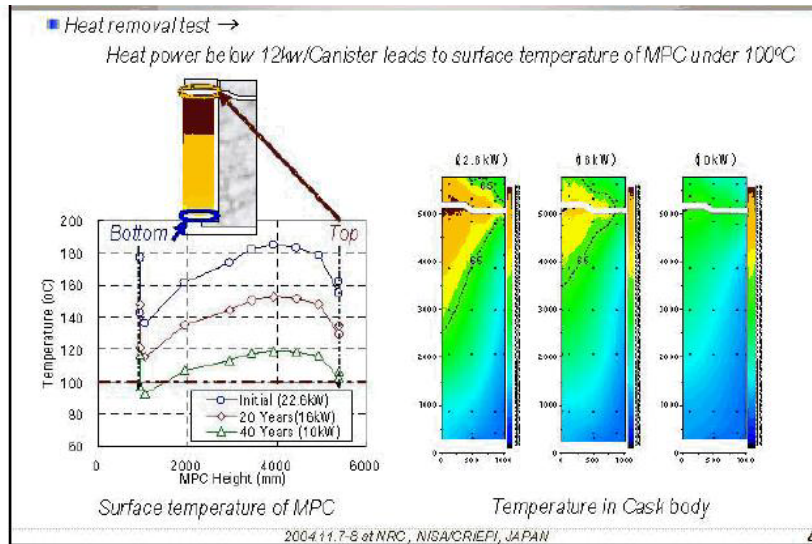


Figure 40 Temperature Profile on MPC [Figure reproduced from ref. 5].

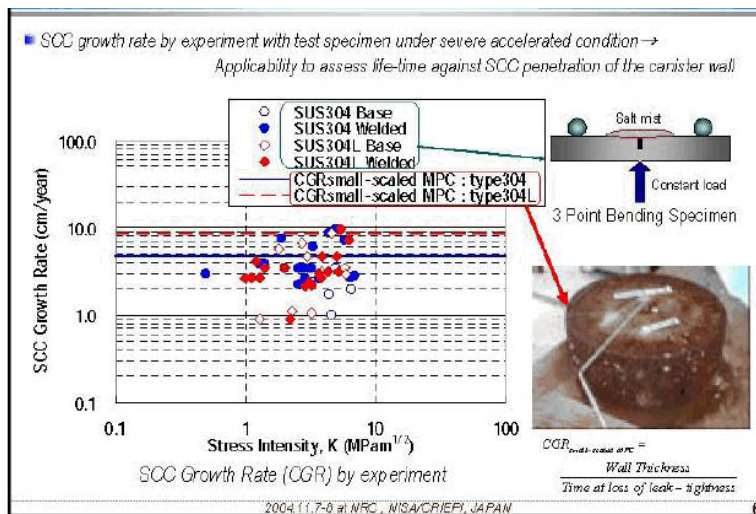


Figure 41 SCC Growth Rate on Weldment Components of MPC [Figure reproduced from ref. 5].

**Impact of Cracking on Confinement** - Fixed displacement stress corrosion cracks can be so tight that in some instances liquids do not leak from the container, even when the crack is throughwall [5]. This observation suggests that radioactive or contaminated particulates would be unlikely to pass through a stress corrosion crack in a canister wall unless the crack were opened by mechanical forces such as might be imposed during handling and transportation. Additionally, such transport will generally require a fluid or gas stream be available for particulate transport. Therefore, the loss of confinement would be limited to gases and volatiles that would be in the volume space of the

canister. However, as previously mentioned, oxygen ingress through a SCC could significantly alter the condition of the stored fuel.

**Residual Stress Distribution on Large Diameter Canisters:** The residual stress field of multipass metal inert gas welds used to fabricate canisters is expected to exhibit both throughwall and azimuthal variation. Typically, tensile stresses will be found on both the inside diameter and outside diameter of 0.5 inch plate that is joined with multipass welds. Sufficient stresses to initiate and propagate a stress corrosion crack are therefore assumed to be present on canister external surfaces. This assumption is consistent with industrial experience that includes SCC failures in systems with no (very small) applied stresses on the failed component.

**Effects of Low-Temperature Neutron Irradiation on SCC:** Previous studies on SCC in fully aqueous conditions were performed to support the low temperature production reactors at the Savannah River Site. Corrosion testing of sensitized Type 304SS, including Type 304SS samples that had been neutron-irradiated at low temperature were performed [7]: Constant extension rate tests (CERT) were conducted to study of environmental effects on intergranular stress corrosion cracking of Type 304 stainless steel. Specimens of both furnace sensitized and annealed material were irradiated to fluence levels of 1 to  $2 \times 10^{21}$  n/cm<sup>2</sup> ( $E > 0.1$  MeV) at 150°C in a beryllium reflector position of the High Flux Isotope Reactor. The CERT test results showed the same pattern of dependence on test variables (high importance of chloride ion and temperature) as the non-irradiated specimens in an associated study and the annealed and irradiated specimens showed no evidence of irradiation assisted stress corrosion cracking. These results demonstrate that the low fluence that the canisters will receive will not significantly change the SCC behavior and suggest that industrial experience with austenitic stainless steel systems is relevant to long-term canister behavior.

Experimental work to characterize stress corrosion cracking in water systems (reactor grade pure water from 40 to 100°C, Cl < 40 ppb) in the Savannah River Site production reactors piping system [8] showed stress corrosion crack growth rates that varied from  $10^{-4}$  to  $10^{-7}$  in/hr ( $2.2$  to  $2.2 \times 10^{-3}$  cm/yr). These results show that cracking can occur at very low chloride concentrations and suggest that stress corrosion cracking of the canisters is possible even in high quality water if water is pooled on the canister in regions where tensile stress are present at the pool-canister interface.

**Assessment of Corrosion Degradation of Canister under EST:** It has been demonstrated in laboratory and in field studies plus observed in field experience with actual and simulated components that the stress corrosion cracking (SCC) of austenitic stainless steel can occur at low temperatures (down to room temperature) in marine-type environments. As discussed in detail in reference [5], the SCC propensity of austenitic stainless steels in marine environments is affected by many variables including the amount of moisture and time of wetness, airborne contaminants (e.g., chlorides, sulfur dioxide, carbon dioxide, etc.), distance, elevation, orientation and shelter, temperature, sunlight and wind [9].

Austenitic stainless steels are very tough, fracture resistant materials. The toughness of the steel should be sufficient to minimize the tendency for mechanically-induced crack growth even during post storage handling and transport operations. Tabulated fracture toughness data are available for Type 304, 304L, 316 and 316L stainless steel and, if stress corrosion crack lengths in a canister were established by non-destructive



inspections techniques, available fracture data could be used to establish safe handling and transport protocols.

ASME code cases N-513 and N-705 provide consensus methods to disposition planar and non-planar flaws (cracks) as would occur from SCC and pitting. These code cases also provide consensus crack growth rates for high (> 200°F (93.3°C)) and low (< 200°F (93.3°C)) temperature water to support flaw disposition in high purity water conditions. The growth rates observed in samples exposed to salt water condition appear to exceed by an order of magnitude those for similarly stressed samples (common crack tip stress intensities) exposed to high purity water at a common stress intensity loading. Additional data are needed to establish consensus crack growth rates for canister materials exposed to conditions anticipated during EST.

**Assessment of SCC and Pitting in Canister in EST, Impact to Safety Function:** The potential for stress corrosion crack and/or pit development presents several challenges to the safety functions of the DCSS:

1. a loss in confinement function of the storage canister,
2. the loss of retrievability of the fuel assemblies due to degradation of the fuel by oxygen ingress and the potential for structural weakness in a canister containing cracks,
3. a loss of confinement for transport of a cracked or pitted canister.

The toughness of the steel, combined with the tightness of potential stress corrosion cracks suggest that non-destructive inspection techniques could safely be used to identify crack sites and that actions could be taken to mitigate further risks and safely recover the canister and contents. However, it must be recognized that the potential for stress corrosion cracking in canister walls clearly exists. The time to initiate SCC in the stainless steel is highly variable. Low temperatures of the canister would allow moisture condensation and would be increase the potential for SCC. Therefore, SCC is anticipated to be significant to canister integrity in EST.

Reference 6 reports that deliquescence of a salt mixture of NaCl–NaNO<sub>3</sub>–KNO<sub>3</sub> can occur at a 20% relative humidity at 160°C. This data is used to suggest that if occluded areas are established, sufficient chemistry (aggressive species and water) could form, even at temperatures well above 100°C, on the canister surface to lead to pitting and cracking. This condition could adversely impact the canister integrity and confinement function.

#### **Canister Corrosion Degradation Data Needs for EST and General**

**Recommendations for Resolution** - Available data and analyses demonstrate a very real potential for stress corrosion cracking of the austenitic stainless steel canisters. Additional information concerning this potential is unlikely to provide practical technologies to mitigate or even minimize the tendency for cracking. Therefore any research to address stress corrosion cracking susceptibility should focus on developing 1) technologies to determine (remotely if practical) the location and size of any cracks that may occur during dry storage, and 2) analytical methodologies to assure that identified and characterized cracks do not compromise the handling and transport of the canister system. Specifically,

1. Monitoring/In-Service Inspection Activity: Prescribe in-service inspection for canister and develop inspection methods, as practical for canister to monitoring for pitting and cracking. Develop acceptance criteria for disposition of detected flaws.
2. Testing & Analysis Activity: Develop structural and fracture methodologies with consensus for disposition of canister flaws in terms of acceptance leakage, acceptable flaw size, structural resistant to handling and DBE loadings and other anticipated activities.
3. Testing & Analysis Activity: Develop methods to effectively flush surface contaminants to minimize potential for deleterious salt and debris buildup on canister surface.
4. Cask Demonstration Activity: As part of a cask demonstration, canister should be monitored for:
  - a. buildup of surface debris including salts and particles (e.g., a “swipe” of the surface taken to evaluated particulate deposits);
  - b. canister surface temperature and relative humidity should be monitored;
  - c. NDE methods should adapted/developed to identify and characterize pitting and cracking, remotely through vents in the overpack, as practicable.
5. Maintenance Activity: Development of a wet or dry transfer system with protocols for canisters that develop flaws that need remediation

The distinct probability that SCC will occur in a canister during EST illustrates the importance of the above activities because 1) the canister provides confinement of the DCSS; 2) the canister is susceptible to pitting and cracking with raising of ambient relative humidity and capability for salts to deliquesce on the canister surface as temperature decreases; and 3) the necessity for remediation of a flawed canister will be adversely impact cost and safety in the management of DCSS in EST.

### **Section 2.6.2 References**

- [1] Zapp, P.E., et al “Relative Humidity and the Susceptibility of Austenitic Stainless Steel to Stress Corrosion Cracking in an Impure Plutonium Oxide Environment”, p.4 Vol. XXXVIII, Number 3, Journal of Nuclear Materials Management, 2010.
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- [5] Effects of Marine Environments on Stress Corrosion Cracking of Austenitic Stainless Steels, EPRI 1011820, 2005.
- [6] Carroll, S., Craig, L., Wolery, T., "Deliquescence of NaCl–NaNO<sub>3</sub>, KNO<sub>3</sub>–NaNO<sub>3</sub>, and NaCl–KNO<sub>3</sub> salt mixtures from 90 to 120°C," *Geochem. Trans.* 2004; 6(2):19.
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- [8] WSRC-RP-90-95, "Reactor Materials Program – A Derivation of a Nominal IGSCC Growth Rate for SRS Reactors (U)," Westinghouse Savannah River Company Report, March 1990.
- [9] Climatic Corrosion Considerations for Independent Spent Fuel Storage Installations in Marine Environments EPRI 1013524, June 2006.
- [10] ASME Code Case N-513, Evaluation Criteria for Temporary Acceptance of Flaws in Moderate Energy Class 2 or 3 Piping.
- [11] ASME/BPVC CASE N-705: N-705 Evaluation Criteria for Temporary Acceptance of Degradation in Moderate Energy Class 2 or 3 Vessels and Tanks Section XI, Division 1 Supp 4.

## **2.7 Bolted Closures Description (Bolts and gaskets)**

A component of the confinement boundary for some canister designs or for casks that are an integrated unit includes bolted closures using metallic seals [1, 2, and 3]. A variety of metallic gasket materials, including Inconel X730, Al-jacketed Nimonic 90, and Al-jacketed Inconel X750, are used. Bolt materials include 304L SS, 193 Grade B7, 320 Grade L43, SB637 Grade NO7718, SA564 Type 630, and H1150. Bolts can fail if given an improper heat treatment (temper embrittlement) however that event is not aging-related.

Loss of confinement could occur if either the sealing capability of the gaskets was compromised due to viscous flow or creep, or the bolt closure stresses were reduced by processes such as stress corrosion cracking. The potential for these degradation processes is discussed in the following section of the report.

Polymeric seals are used in DCSS to provide secondary confinement and leak detection capability, and are used in transportation packages to provide confinement. A variety of polymeric compounds are used as listed in reference [3].

## **Section 2.7 References**

- [1] Source: U.S. Nuclear Regulatory Commission, <http://www.nrc.gov/waste/spent-fuel-storage/designs.html>.
- [2] Industry Spent Fuel Storage Handbook, EPRI Report 1021048, July 2010.
- [3] ASTM C-1562-10, Standard Guide for Evaluation of Materials used in Extended Service of Interim Spent Nuclear Fuel Dry Storage Systems, American Society for Testing and Materials, reapproved 2010.

## **2.8 Bolted Closures Degradation Phenomena**

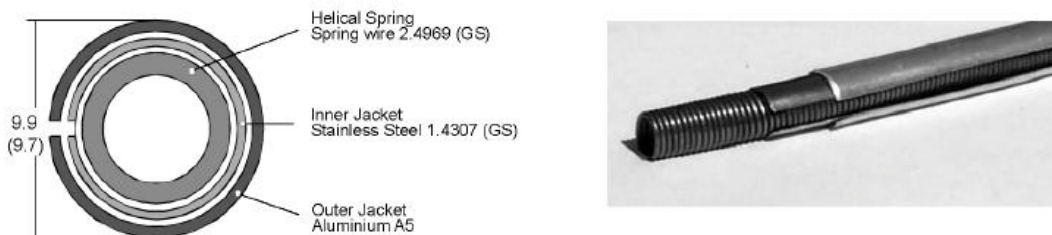
Metallic seals and bolts are anticipated to be subject to degradation in EST. Given that these components are SSC and part of the confinement boundary, it is important to assess the sealing capability of a system under EST.

### **2.8.1 Metallic Seal Degradation**

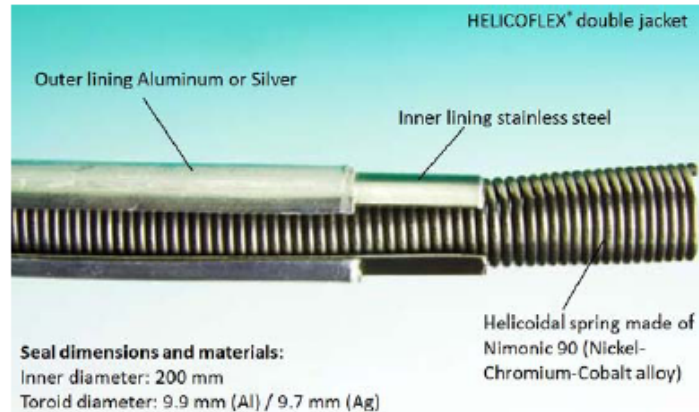
Metallic gaskets are an essential component in the confinement of a bolted closure, and are subject to aging [1]. The degradation of a closure seal could occur if the gasket relaxed to the point that no sealing forces were present on the closed joint. The ability to forecast century-long performance from 25,000-hour-long experiments is a difficult process thus accurate assessments of gasket behavior during EST are not available although several experiments are in progress.

As an example of an approach to demonstrate metallic seal efficacy over long-term storage, an experimental program to quantify the relaxation of four types of metallic Helicoflex® seals for their use in spent nuclear fuel storage casks is being conducted [2].

The Helicoflex® seals (see Figure 42 and Figure 43) are constructed of a helical spring, surrounded by an inner jacket of stainless steel and an outer jacket of a malleable material, such as aluminum or silver. These seals are capable of sealing to leakage rates of less than  $10^{-7}$  atm cc s<sup>-1</sup>.



**Figure 42** General structure of 'Helicoflex' type metal seals  
[Figure reproduced from ref. 2].



**Figure 43** Construction principles of HELICOFLEX® double jacket seals [Figure reproduced from ref. 2].

Two types of linings were studied: aluminum and silver [3]. Tests longer than 10,000 hr were implemented only for silver. For each type of lining, two different section diameters were investigated. A primary goal of the work [3] was to determine the residual linear load that can be guaranteed for a seal after a particular time of relaxation. This relaxation depends on the seal temperature and time. To test this process, seals were tightened between two flanges and exposed to constant temperatures, including 100 and 200°C. Residual load and ‘useful’ recovery was measured after the exposure. Results were interpreted according to two methods: a time extrapolation, and a time–temperature equivalence parameter. Both methods are based on linear relationships and were assessed through a statistical analysis (calculation of scatter) which is also used to determine a minimum guaranteed residual closure load. Finite element simulations of the relaxation of a seal were also performed in order to show that the time extrapolation method is appropriately conservative.

For silver lining seals, the authors of the referenced report conclude that use of a time–temperature equivalence parameter equal to  $T(11 + \log_{10}(t))$  is justified. This relationship allows one to assess the maximum temperature at which seals can be ‘safely’ used. However, the use of this relationship assumes that a residual linear load of at least 100 N/mm will maintain a seal after closing the cask. This time/temperature equivalence may not be a universal relationship for all materials and designs. Additional work is needed to validate this assumption or to determine bounding (minimal) residual loads required to maintain leaktightness.

Studies completed and on-going at BAM indicate that relaxation of the sealing force of metallic seals is expected to occur over time because of plastic deformation of the outer jacket of the seal. Greater deformation is expected at higher temperatures. The extent of the relaxation is determined by the construction of the seal and the temperature of the sealing surfaces. Table 4 shows the extent of relaxation which occurs in Helicoflex® seals with outer jackets of aluminum or silver at two different temperatures after 1 year.

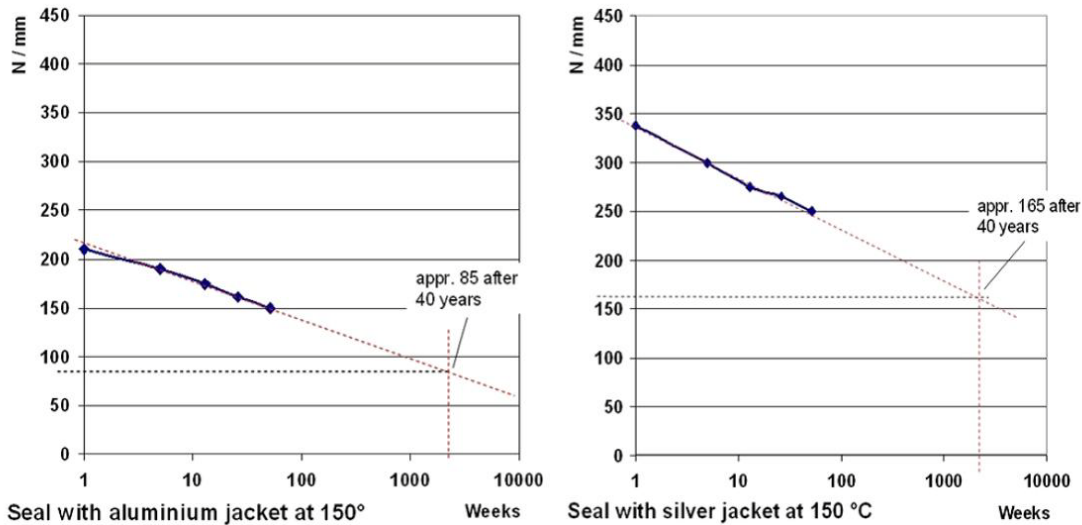
In addition to the potential problems with stress relaxation, the crevices formed by gasket joint are subject to crevice corrosion conditions including potential galvanic effects. Moisture is assumed to be able to condense and build up in the crevice of the joint. The kinetics of crevice corrosion are difficult to predict reliably but crevice

corrosion is supported by the same environmental conditions that support SCC and as indicated in section 2.2.6 those conditions will exist on canisters during EST.

**Table 4** Sealing Force after Temperature Exposure for 1 Year [3]

Seal jacket material	Reduction of initial sealing force after 1 year (% of initial force remaining)	
	150 °C	20 °C (room temperature)
Aluminum, Al	39%	78%
Silver, Ag	51%	81%

Figure 44 shows an extrapolation of the 150 °C data indicates the decrease in the sealing force due to stress relaxation would be expected to continue leaving but even after 40 years approximately 22% (aluminum) or 33% (silver) of the initial sealing force will remain. Further extrapolation of these data indicate that at least 10,000 weeks (192 years) is required to reach the 100 N/mm threshold value assumed above. Linear extrapolation goes well beyond the 10,000 week point, and is closer to 20,000-30,000 weeks (384-576 years). Linear extrapolation over this range is tenuous but indicates significant lifetimes are possible, perhaps up to and beyond 300 years if no significant changes in the relaxation mechanism are observed and crevice corrosion does not occur.



**Figure 44** Extrapolation of seal force decrease over 40 years under constant conditions [Figure reproduced from ref. 3].

Elastic rebound or resiliency is another parameter that is suggested [3] to be linked to sealability. This parameter is also subject to Arrhenius aging behavior or property reduction thus these parameters also depend on temperature and time.

**Assessment of Metallic Seal Degradation – Impact to Safety:** As a consequence of the continued reduction in sealing force during EST, replacement of the metallic seals may be required at some point to insure the integrity of the primary confinement boundary of the cask. Otherwise, undetected loss of the seal could lead to loss of the inert cover gas in the cask and to infiltration of outside atmosphere. Such infiltration could potentially lead to degradation of the fuel and release of radioactive material.

**Metallic Gasket Degradation Data Needs for EST and General Recommendations for Resolution:** Metallic seals should not be assumed to remain leak tight throughout the entire 300 years target for EST. Available data are simply not sufficient to support that assumption. However, the gaskets may continue to provide a sufficient barrier to leakage for multiple decades even after seal forces have been greatly reduced (e.g., due to viscous flow of the seal). It is recommended that a plan for replacement of the metallic seals for EST be developed and included as part of periodic maintenance. This could involve a re-wet or a dry system to effect seal replacement. Specifically,

1. Monitoring/In-Service Inspection Activity: Periodic surveillance for leakage at the seal joint would be needed under EST with acceptance criteria for leakage detection.
2. Testing & Analysis Activity: The metallic leakage tests in progress (BAM) should be continued to evaluate the sealing force at which leakage occurs and also evaluate the tendency for crevice corrosion degradation. The effect of temperature cycling, service environment (air, moisture, chlorides, etc.) and other possible relevant parameters should be accounted for in the testing. The response of aged/relaxed metallic seals under transient excursion conditions (if deemed possible, such as an extended abnormally cold period) should be considered. Depending on cask/seal design and actual seal temperatures, such excursions may or may not pose a concern.
3. Maintenance Activity: Development of a wet or dry system with protocols for replacement of seals that develop leakage or experience crevice corrosion and need to be replaced.

Metallic seal degradation during EST is likely to be significant due to the confinement function provided by metallic seals, and the continuing loss of sealing force with time. The potential for crevice corrosion also contributes to the significance.

### **Section 2.8.1 References**

- [1] “Ageing of metallic gaskets for spent fuel casks: Century-long life forecast from 25,000-h-long experiments”, H. Sassoulas, L. Morice, P. Caplain, C. Rouaud, L. Mirabel, F. Beal, Nuclear Engineering and Design 236 (2006) pp. 2411–2417.
- [2] General structure of ‘Helicoflex’ type metal seals [ref. D. Wolff, et al, “Safety Aspects of Long-Term Dry Interim Storage of Type-B Spent Fuel and HLW Transport Casks” RAMTRANS Vol. 15, Nos.3–4, pp. 207–213 (2004).]
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## **2.8.2 Polymeric Seal Degradation**

Elastomeric seals are not credited for confinement in storage, though they may be part of the transport/storage cask design for leak testing purposes. Therefore, degradation of elastomeric seals is considered a relatively low priority item from a DCSS confinement perspective for EST. Elastomeric seals are used in post-storage transportation, and are important to safety if the elastomeric seals have to provide a confinement function. In that case, it may be prudent or necessary to evaluate elastomeric seals used in the DCSS design to predict when such seals may no longer provide the regulatory function.

The primary degradation mechanism for elastomeric seals exposed to an oxygen-bearing environment is oxidation and the oxidation processes are enhanced by elevated temperature and radiation. The general literature strongly suggests that elastomers used as seals have lifetimes of no more than several decades [1-8], and therefore would require periodic replacement for continuing integrity during EST, and as a minimum, replacement must be made prior to post-storage transportation where integrity must be assured.

The lifetime of elastomeric seals is dependent on many factors, including the seal material, storage conditions, the acceptance criterion used to define lifetime, design parameters and sensitivity to oxidation. Seals that only have to maintain minimal sealing integrity at well-controlled ambient conditions may function for many years. For example, some studies indicate that it may be possible to use of EPDM seals from 150 years to several hundred years at well-controlled conditions (25°C) [3]. Seals that are exposed to higher temperatures or radiation doses will likely fail sooner. Accelerated-aging studies based solely on leak rate measurements or monitoring of a mechanical parameter such as compression set or compression stress-relaxation are useful for prediction purposes, but may overlook slower degradation mechanisms such as diffusion-limited oxidation. Oxygen consumption analysis and other techniques are therefore proving useful in providing more reliable lifetimes for elastomeric seals [5, 6].

Elastomeric seals are not credited for confinement directly during storage; however, there may be a need to credit such seals for post-storage transportation. Therefore, the aging of elastomeric seals may still be an issue to consider for the very long-term dry storage period. The length of time the seal needs to function and what acceptance criterion should be established (helium leaktight per ANSI N14.5 or other) are not well-defined.

The seals may be credited with being leak tight defined per ANSI N14.5 as  $1 \times 10^{-7}$  ref cc/sec or  $2 \times 10^{-7}$  cc/sec He. This is the typical requirement for elastomeric seals in a Type B shipping package such as the Model 9975 [7]. A common approach in such designs is to use dual O-rings, with one seal credited for confinement and the other one for leak testing of the volume in between the two seals. For transportation, such packages are generally demonstrated to be leak tight prior to loading, then the packages are loaded and given a post-assembly verification test, often using a pressure drop criteria with  $1 \times 10^{-4}$  cc/sec sensitivity. For short-term transport purposes, it is not necessary or required to verify helium leaktightness during the shipping period as the seals are replaced during maintenance. However, if the shipping package is then used for interim storage, it may be difficult if not impossible to verify that the seals are still leaktight after assembly unless the system is repackaged.



The basis for elastomer seal qualification in shipping packages or casks is typically to prove that a new seal can be leaktight at room temperature and perhaps down to low temperatures (such as -40 °C) that might be encountered during transport, depending upon requirements specific to the package. Leak tests after elevated temperature exposure might also be performed as part of the package qualification, but usually the aging time involved is quite limited. It is important to note that the technical basis for “continuous” service temperature limits often quoted by elastomer seal manufacturers is often very limited and is typically based on test results from relatively short aging periods (e.g. 1000 hrs. or so). Validation of long-term seal performance, regardless of temperature, is usually left to the end-user to perform.

Many polymers, including most elastomers, also exhibit a significant difference in radiation and temperature resistance between oxidizing and non-oxidizing or vacuum environments, particularly if the radiation dose rates are high. There is a strong synergistic relationship between radiation, temperature, and oxygen that will impact the performance of any polymer. While this relationship varies in significance among polymer types, the general observation is that materials exposed to radiation and/or temperature in limited oxygen environments or vacuum exhibit significantly higher resistance to degradation, as defined by measurement of certain mechanical properties such as tensile strength and elongation than materials exposed to high oxygen concentrations. Such properties are usually measured for convenience and relative comparison, but are not necessarily the most relevant for all applications such as O-rings or gaskets. In addition, the relative change in some properties may be higher or lower than the changes observed in other properties, making complete evaluation of environmental resistance more complex. Therefore, critical properties should be evaluated based on the nature of the application when possible. Nevertheless, a review of the common elastomeric seals in a transportable storage cask concludes that “All elastomer seals are unacceptable for exposures of  $10^8$  rads” [8].

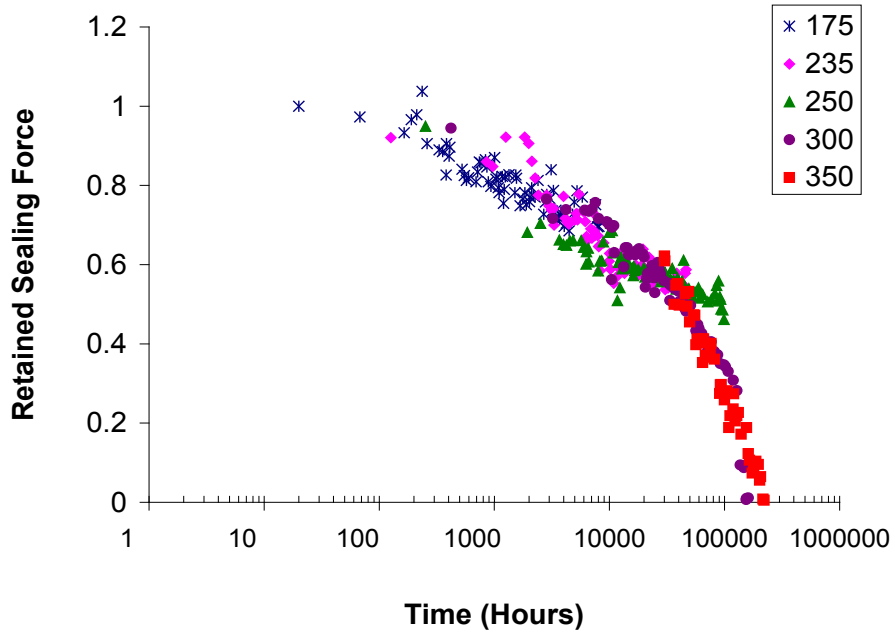
The general aging methodology is to assume Arrhenius-type behavior, which assumes a single activation energy for the degradation mechanism over the entire temperature/dose rate range anticipated in service. Though useful for evaluation purposes and general prediction of service life, many studies over the years have shown evidence of non-Arrhenius aging behavior, diffusion-limited oxidation effects and other factors that complicate the life prediction of elastomeric materials [1-9].

Using the general Arrhenius-type relationship for reaction rate as a function of temperature, lifetime to failure (of varying definition) can be extrapolated for varying temperatures. Figure 45 and Figure 46 show examples of the trend of loss of sealing force using time-temperature superposition principles. All experimental data at various aging temperatures are superposed into a “master” curve using shift factors that can be translated to other service temperatures of interest.

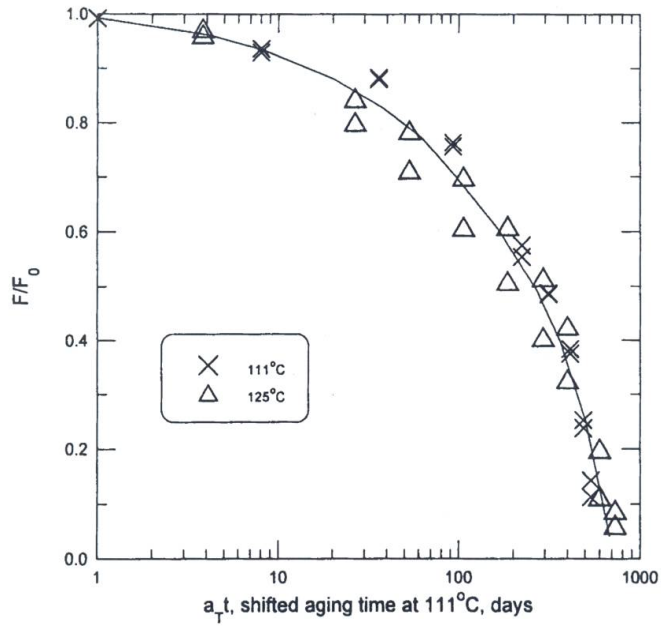
Among the limitations of these techniques are: 1) the use of a constant activation energy over the temperature range anticipated may not be valid, and 2) extreme extrapolation of higher temperature results to lower temperatures does not always provide an accurate result due to competing degradation mechanisms. This is particularly true for materials experimentally aged at temperatures much higher than anticipated in service.

Due to the synergy of all competing mechanisms, variation in compound formulation, and specific service conditions, accelerated aging can never completely duplicate true long-term aging data in actual service. A useful approach is to periodically leak test sealed mock or surrogate vessels that are aged at relevant temperatures for the storage environment [7, 9-11].

The correlation between leak tightness and mechanical parameters such as elongation, compression set or even compression stress-relaxation (CSR) is not well-defined and likely varies with each design. Even with the same seal compound, some designs may be more or less sensitive to seal material property changes than other designs.



**Figure 45** Time-Temperature Superposition of Compression Stress Relaxation Data for Viton GLT O-rings (Parker V0835-75) at 80 °C [Figure reproduced from ref. 7].



**Figure 46** Time-Temperature Superposition of CSR data for EPDM seals [Figure reproduced from ref. 2].

**Assessment of Polymer Seal Degradation – Impact to Safety:** The function of sealing by elastomeric (polymeric) seals is typically not credited for long-term confinement in DCSS designs due to known active degradation. They are used as secondary seals to provide a leak check of the primary seal, and thus are not credited directly with a safety function.

Most polymeric materials are subject to degradation of their physical properties by oxidation, radiation, heat and/or hysteresis effects. However, the metrics of compression set and compression stress relaxation (CSR) are often used for predicting the service life and sealing capability of the elastomers. Oxygen consumption analysis and dynamic mechanical analysis can also be useful, particularly to identify possible non-Arrhenius behavior. Such metrics do not provide direct time to leakage failure, as leak rate depends on many design parameters, but do provide insight into long-term material performance.

No requisite data needs for EST are established. It is assumed that polymeric seals will not meet leak tightness for desired service after, at most several decades in DCSS service. Additional research may be warranted to predict seal lifetime in order to maximize seal longevity and minimize seal replacement efforts, during storage and for post-storage transportation. When seals are replaced (either due to degradation or at a prescribed maintenance period), it is recommended that the removed seals be retained for further analysis and characterization. Such data may be useful for other nuclear materials packaging applications and non-nuclear applications where the same elastomer compounds may be used.

### **Section 2.8.2 References**

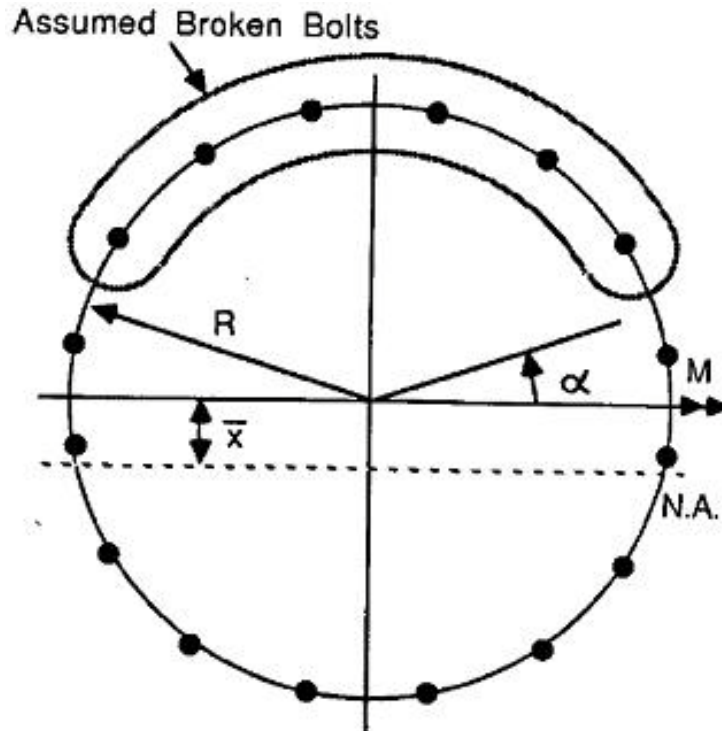
- [1] “Validation of Improved Methods for Predicting Long-Term Elastomeric Seal Lifetimes from Compression Stress-Relaxation and Oxygen Consumption Techniques”, Polymer Degradation and Stability 82 (2003), K.T. Gillen, M. Celina, R. Bernstein.
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- [6] “Predicting and confirming the lifetime of O-rings”, K.T. Gillen, R. Bernstein, M.H. Wilson, Polymer Degradation and Stability 87 (2005) pp. 257-270.

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- [10] "Seal life of EPDM O-rings at high temperature determined by unique method", R. Marlier, R. Andre, P. Malesys and H. Issard, Packaging, Transport, Storage & Security of Radioactive Materials, 2006, VOL. 17 NO 1.
- [11] "First tests results for determination of seal life of EPDM O-rings at high temperature (determined by unique method)", Packaging, Transport, Storage & Security of Radioactive Material, 2010, VOL. 21 NO 1.

### **2.8.3 Bolt Degradation**

Carbon and low alloy steel bolts are subject to general corrosion and creep in EST. With time, bolt failure can occur, and if sequential (Figure 47), a loss of gasket compression and leak tightness would be expected. A failure of one or several bolts may not cause a sealing (confinement) failure unless the failed bolts are adjacent to each other in the bolt pattern.

Bolting is considered a maintenance item that would be replaced. Corrosion and stress-cracking are common degradation phenomena in bolts, and a schedule for bolt replacement is suggested for aging management. Bolts would not be expected to remain viable for EST.



**Figure 47** Loss of Sealing Force Can Occur with Multiple Adjacent Bolt Breakage.

**Bolt Degradation Data Needs for EST and General Recommendations for**

**Resolution:** Bolts are not expected to remain capable of providing a sealing force throughout the entire 300 years target for EST. Single bolt failures are not expected to cause a significant loss of sealing capacity of the gasket/bolt joint. However, multiple sequential bolt failures would be expected to enable a leakage path and loss of confinement. It is recommended that a plan for periodic monitoring of the bolts (e.g., check torque) and/or bolt replacement be performed for EST as part of periodic maintenance. Specifically,

1. Monitoring/In-Service Inspection Activity: Periodic surveillance for leakage at the seal joint would be needed under EST with acceptance criteria for leakage detection.
2. Testing & Analysis Activity: Analysis of cask to determine the impact of sequence of bolt failure on leak tightness of gasket/bolt/cask system.
3. Maintenance Activity: Development plan for periodic bolt replacement.

The need for bolt replacement during EST is high due to the confinement function provided by metallic seal/bolt system, and the potential for bolt degradation and breakage to go undetected unless actively checked.

## **2.9 Casks Description**

The casks of a licensed DCSS may be of several different designs [1, 2, and 3]. Figure 48 shows a generic overpack on a canister that comprises the cask. Other cask designs are an integral container with shielding rather than the overpack. Cask materials are listed in the table in Annex 3 of reference [4].

Casks bodies are made of ferritic or ferritic-martensitic steel; stainless steel; cast iron and graphite. Some casks contain lead that, as a component of the system, provides shielding, confinement, and thermal performance functions.

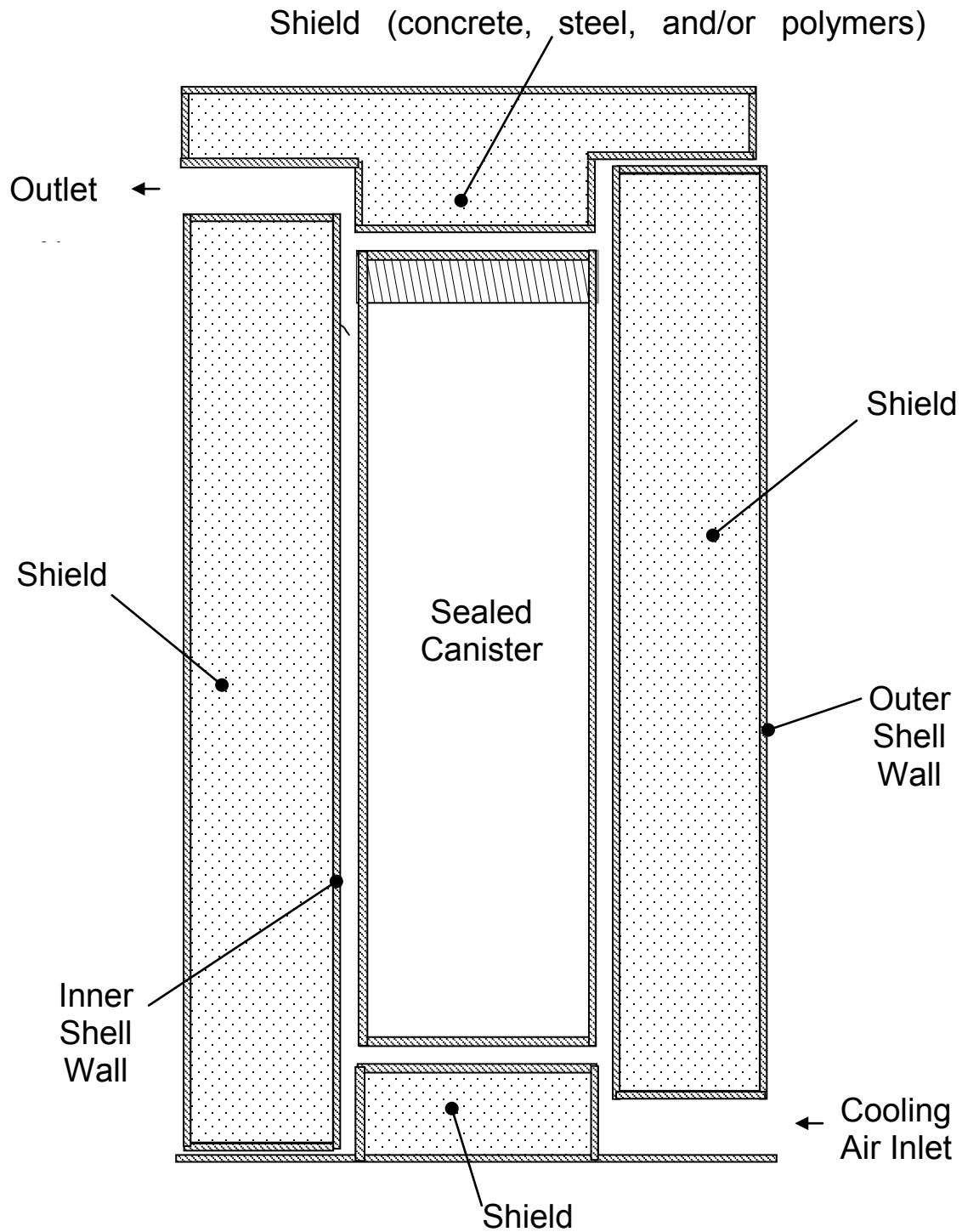
Casks can be made of concrete, with or without reinforcement and with or without steel shells and lead. These casks provide a shielding and thermal performance functions and confinement is provided by the stainless steel canisters within the cask

Casks may also contain components that are layers of neutron absorber and/or gamma shields as part of the cask structure. Such layers provide an important safety function. Radiation shielding is incorporated into dry storage systems as a means of providing integral personnel protection. For example, polyethylene or other low Z material is often placed in a cask as a neutron shield. Similarly, gamma shielding is provided in dry storage systems, either as an add-on material (e.g., lead), or by appropriate selection of the dry storage system construction materials. For example, heavy steel or concrete casks, by their very nature, provide a measure of radiation shielding.

A variety of designs are used for neutron shielding [1]. Neutron shielding in the Transnuclear Inc. TN-40 cask is provided by a resin compound surrounding the body. The resin compound is enclosed in long, slender aluminum containers. The array of resin filled containers is enclosed within a smooth outer steel shell. A disk of polypropylene is attached to the cask lid to provide neutron shielding during storage.

Polyethylene shielding rods are used as a neutron shield in CASTOR-V/21 PWR spent fuel storage cask. For the NAC-STC, gamma shielding is provided by a lead wall and neutron shielding by a layer of a solid borated synthetic polymer, which surrounds the outer shell along the cavity region.

The Westinghouse MC-10 cask body is made of low-alloy steel and has forged steel walls and bottom that provide radiation (gamma) shielding and structural integrity. Neutron shielding is provided by a layer of BISCO NS-3 cured in the cavity between the cask wall and outer protective skin. In concrete dry storage systems (casks and vaults), neutron and gamma radiation shielding are provided, in part, by the concrete structure.



**Figure 48** Generic Dry Storage Cask Sketch for an Overpack on a Canister.



## **Section 2.9 References**

- [1] Source: U.S. Nuclear Regulatory Commission, <http://www.nrc.gov/waste/spent-fuel-storage/designs.html>.
- [2] Industry Spent Fuel Storage Handbook, EPRI Report 1021048, July 2010.
- [3] Data Needs for Long-Term Dry Storage of LWR Fuel, EPRI report number TR-108757, Interim Report, April 1998.
- [4] ASTM C-1562-10, Standard Guide for Evaluation of Materials used in Extended Service of Interim Spent Nuclear Fuel Dry Storage Systems, American Society for Testing and Materials, reapproved 2010.
- [5] IAEA TECDOC 1100, Survey of Wet and Dry Spent Fuel Storage, July 1999.

## **2.10 Casks Degradation Phenomena**

Excessive degradation of the materials of the casks could lead to failure to provide a shielding function, structural function, loss of environmental control and/or loss of configuration control for the fuel storage.

### **2.10.1 Carbon Steel Material Corrosion**

Non-stainless metallic systems may be subject to corrosion over EST. The metal shell for thick-walled monolithic metal canister systems provides gamma radiation shielding which is provided by lead in multi-shelled metal casks and by the concrete in thin-walled metal casks. Carbon steel will require periodic maintenance to avoid general and pitting corrosion. This is a low priority data need due to the ease of detection and maintenance.

### **2.10.2 Neutron Shielding Degradation Phenomena**

Polymeric materials and resins are used in transport and storage casks as neutron shields due to their high hydrogen contents. These materials are subject to degradation and possible relocation within the casks as a result of thermal and radiation exposures, as well as the passage of time. Chain scission can result in a reduction in molecular weight and an increased tendency of the polymer to creep. Crosslinking can increase the molecular weight of the polymer, increasing its hardness and resistance to creep. These radiation effects are sensitive to the applied dose, the dose rate and the presence of oxidants and other reactive species that make polymer performance application specific.

Polymeric materials and resins used for neutron shielding are subject to degradation and possible relocation as a result of thermal and radiation exposures, as well as the passage of time. Loss of shielding will be detectable during required periodic radiation monitoring and must be addressed as part of the life management of the dry storage facility. Materials for neutron shielding include polyethylene, polypropylene, and borated polymer. During very long-term storage, these shielding materials are subject to thermal, radiation, and corrosion degradation phenomena that may lead to physical

changes in the materials that may impair the ability of the neutrons shield to perform its intended safety function.

Periodic survey of radiation fields from the cask should be performed and administrative controls could be readily implemented if a loss of shielding was detected. This is a low priority item, especially since the radiation fields in a cask will reduce with time during EST.

## **2.11 Concrete Pad Description**

A concrete pad is used as the foundation to stage the casks in the DCSS. The concrete pad is not generally within the scope of the materials degradation analysis for present licensing consideration unless the pad provides a safety function during a seismic event [1]. Several mechanisms for concrete degradation described below in section 2.12 of this report are anticipated to be significant to cause loss of compressive strength and/or reinforcement bar corrosion leading to spallation and further loss of material section over a very long term exposure period. These degraded conditions of the pad may impact the stability of the foundation for the casks under normal conditions. Additionally, concrete is also used in overpacks and is susceptible to the degradation mechanisms addressed.

**Reference Concrete Pad Design:** The concrete pad design and construction is assumed to be in accordance with the specifications and codes of the American Concrete Institute, ACI 318 [2].

Placement of concrete is important in its susceptibility to degradation. It is important to prepare concrete with 1) the correct positioning of the steel reinforcement, 2) an appropriate water/cement ratio and the water chemistry being low in detrimental constituents such as chloride species, 3) the correct use of aggregates and 4) other considerations in weathering regions. It is assumed that the as-fabricated condition of the pad provides for high-quality concrete in its initial condition. Guidance to assure a dense and well-cured concrete is provided in references 2-4.

The concrete is considered to have a compressive strength of 3000 psi. The reinforcement bar is considered to be #3 (0.375 inch (0.953cm) diameter) with a yield strength of 60 ksi. A cover of concrete of 1 inch is assumed over the reinforcement bar for the purpose of this assessment of degradation.

### **Section 2.11 References**

- [1] Section 2.4.3 of NUREG-1927, Standard Review Plan for Renewal of Independent Spent Fuel Storage Installation Licenses and Dry Cask Storage System Certificates of Compliance. <http://www.nrc.gov/reading-rm/doc-collections/nuregs/staff/sr1927/>.
- [2] ACI-318-08, Building Code Requirements for Structural Concrete and Commentary, American Concrete Institute, 2008.
- [3] ACI-301-10, Specifications for Structural Concrete, American Concrete Institute, 2010.

[4] ACI 201.2R-08, Guide to Durable Concrete, American Concrete Institute, 2008.

## 2.12 Concrete Pad Degradation Phenomena

The physical properties and/or physical condition of concrete can degrade and impact the ability of concrete structures to perform their desired function. The modeling of concrete degradation that occurs during the EST exposure requires a coupled chemical-physical model to accurately describe the evolution of the degrading concrete. For example, the degrading cement phase in concrete may involve concomitant features including micropore and microcrack evolution, and transport/equilibrium of multiple species including species ingress (e.g.,  $O_2$ ,  $CO_2$ ,  $Cl^-$ ) and leaching (e.g.,  $Ca^{2+}$ ) may even include further hydration of unreacted cement constituents.

Several of the phenomena involve corrosion of the reinforcement bar. Typically, the highly alkaline ( $pH > 13$ ) nature of the cement provides an environment around embedded steel and steel reinforcement, which protects them from corrosion [1, 2]. If the pH is lowered (for example,  $pH < 10$ ) due to carbonation, intrusion of acidic materials through microcracks, and/or leaching of alkaline products through cracks, corrosion of the reinforcement bar may occur causing high internal stresses due to volume mismatch between the steel rebar removed (corroded) and the corrosion products formed as shown in Figure 49. The high stresses can lead to extensive cracking of the concrete and degradation such as that shown in Figure 50.

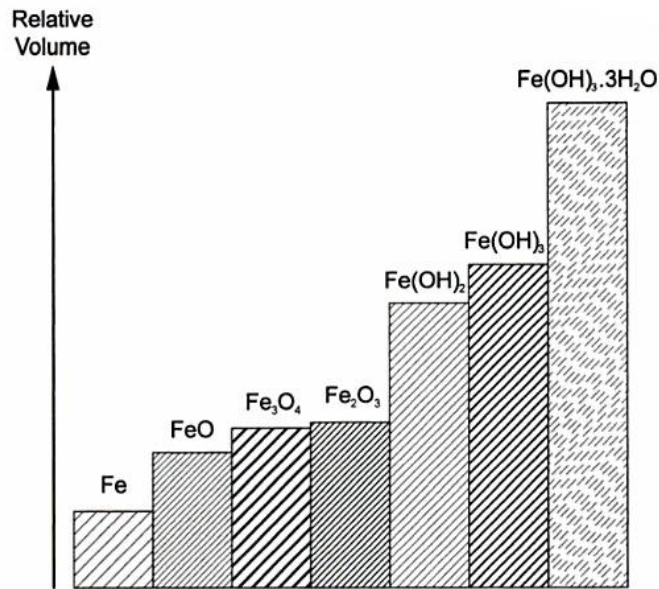


Figure 2.26 Relative volume of possible rebar corrosion products.

**Figure 49** Relative volume of possible rebar corrosion products  
[Figure reproduced from ref. 3].



**Figure 50** Spallation of Concrete Induced by Rebar Corrosion [SRNL Photograph].

Loss of strength may occur with leaching of species (e.g., Ca) that increase porosity, or by ingress of other species (e.g., sulfates) that cause alteration of calcium-silicate-hydrate (CSH) and calcium hydroxide (CH) paste and replacement with a weak paste [2].

The degradation mechanisms relevant to concrete pads and concrete casks in a DCSS are described, and degradation models used for estimation of the condition of the concrete with service exposure and time are listed. An assessment of the significance of degradation in terms of affecting the physical properties or physical condition of the concrete over EST is made with regard to a single operative mechanism (e.g., CO<sub>2</sub> diffusion) unless noted otherwise. An assessment of the potential impact to the safety functions of the DCSS concrete pad or concrete cask from the mechanism is provided. Ideally, the assessment would involve the use of a coupled-phenomena model but a model applicable to the times involved in extended storage is not currently available.

Resolution of concrete degradation issues will primarily be through monitoring/in-service inspection.

### **Section 2.12 References**

- [1] Troxell, G. E., Davis, H. E., and Kelly, J. W., *Composition and Properties of Concrete*, Second Edition (J. W. Kelly), McGraw-Hill, 1968.
- [2] Walton, J. C. et al. "Models for Estimation of Service Life of Concrete Barriers in Low-Level Radioactive Waste Disposal," NUREG/CR-5542, July, 1990.
- [3] Figure from page 159 of Piere R. Roberge, *Handbook of Corrosion Engineering*, McGraw Hill Professional, 2000.

### **2.12.1 Carbonation of Concrete**

Carbonation or the ingress of CO<sub>2</sub> into the concrete to alter its chemistry can lead to concrete degradation by reducing the high alkalinity of the pore water in the material in which reinforcement bars are embedded. Subsequent oxygen ingress into the pore water feeds the corrosion of the steel and the local expansion due to formation of corrosion products can cause large splitting (cracking > ~ 1 mm opening) exposing additional rebar to the ambient external air/water environment with eventual spallation of the concrete cover. In as-prepared concrete, the remaining water in the pores is at a high pH (above 13) from residual solutions saturated in Ca, K, and/or Na hydroxides.

Reaction of CO<sub>2</sub> from the atmosphere with cement can occur to form calcium carbonate and lower pH from basic to neutral so that reinforcing steel becomes susceptible to corrosion [1]. A summary description of carbonation processes in concrete is given in Reference 1 and is consistent with the experiences at the Savannah River Site. Mechanistically-based models show that both time and CO<sub>2</sub> atmospheric concentration increase the depth of carbonation in concrete. The depth of carbonation is proportional to the square root of time.

$$X_c = A\sqrt{t} \quad [5]$$

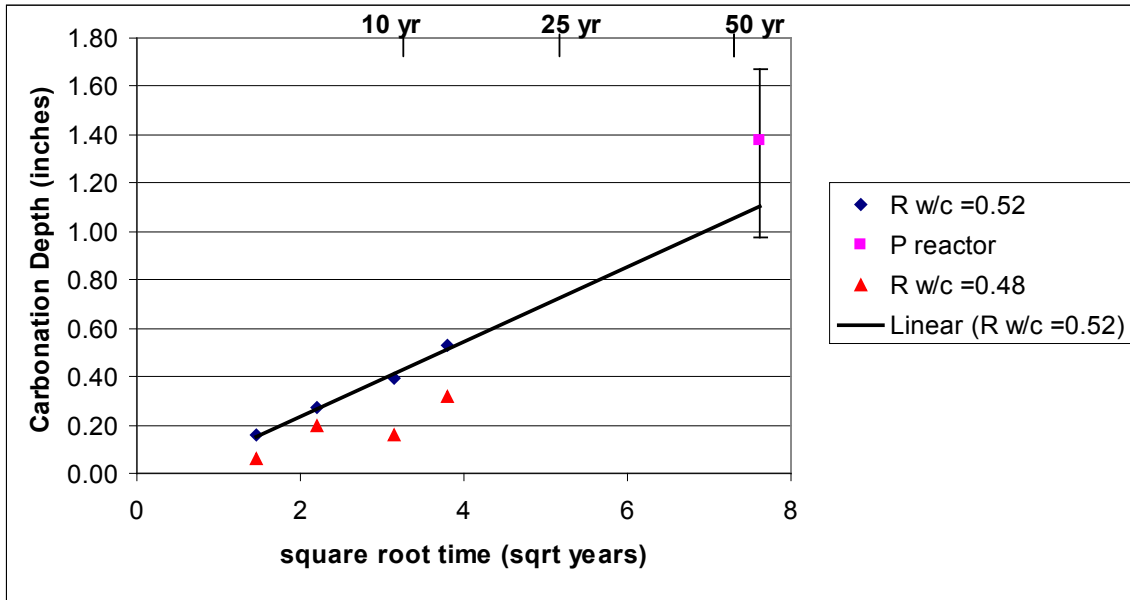
X<sub>c</sub> is the distance from the external surface of the concrete to the carbonation front, t is the time and A is a material constant. Papadakis *et al.* [2] developed an expression for A from theoretical basis.

$$A = \sqrt{\frac{2[CO_2]^0 D_{eCO_2}}{[Ca(OH)_2(s)]^0 + 3[CSH]^0}} \quad [6]$$

D<sub>eCO<sub>2</sub></sub> is the effective diffusivity of CO<sub>2</sub> in the concrete, while [CO<sub>2</sub>]<sup>0</sup>, [Ca(OH)<sub>2</sub>(s)]<sup>0</sup>, where Ca(OH)<sub>2</sub> is the solid calcium hydroxide, and [CSH]<sup>0</sup>, where CSH is the gelatinous calcium silicate hydrate, are the molar concentrations in the concrete.

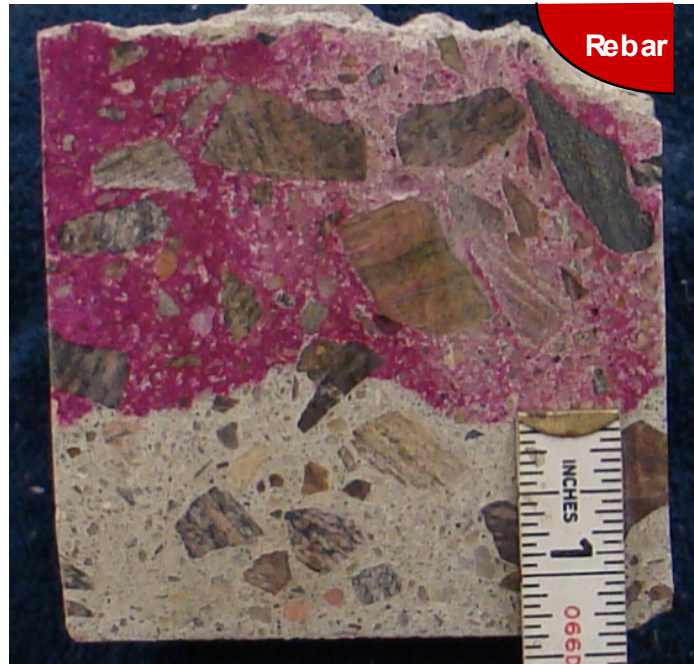
The effective diffusivity of CO<sub>2</sub> is dependent on pore size. Pore size can be affected by several factors including the carbonation reaction itself, as well as moisture, and it must be accounted for in the relationship.

With the assumptions stated above, the carbonation depth can be predicted based on the known age and environmental conditions in service. Figure 51 shows one example of this for concrete exposed in indoor service conditions with data for two cases (w/c ratios of 0.28 and 0.52) from reference 2, and from Savannah River Site (SRS) data from the P-reactor building. For 10 years of service, the carbonation depths vary between 0.5 and 1 cm for the 2 cases shown.



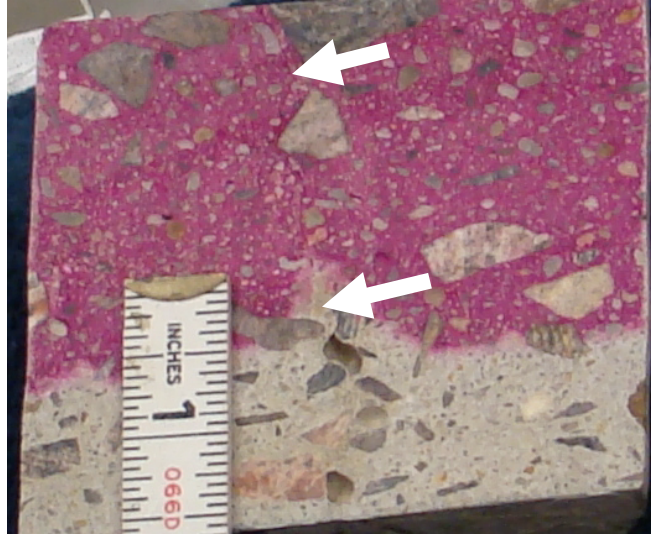
**Figure 51** Carbonation depth with time for normal atmospheric CO<sub>2</sub> exposure indoors of P-reactor building at the Savannah River Site plotted with experimental data by Nagataki et al. [Figure reproduced from ref. 3].

The depth of carbonation can be measured with a simple technique using a concrete core removed from the structure. When a 1% phenolphthalein solution is applied to normal concrete it will turn bright pink. If the concrete has undergone carbonation, no color change will be observed. An example of this is shown in Figure 52. This concrete core was removed from the P-reactor building, an above-ground facility at SRS after approximately 60 years of service. Extrapolation of the linear fit of the literature data [3] to 60 years of service would predict a lower carbonation depth than was actually measured in the sample from the inside wall from the SRS P-reactor as shown in Figure 51. The difference could be attributed to different starting materials or environmental conditions.



**Figure 52** Cross-section of a 3.7 inch (9.4cm) diameter concrete core taken from the inside wall of the SRS P-reactor building. A phenolphthalein solution has been applied to indicate the extent of concrete carbonation. Lack of color indicates a pH below 10.2 showing that carbonation has progressed ~ 1.4 inches (3.6cm) in from the surface of the inner wall. [Figure reproduced from ref. 10].

Cracking in concrete can lead to a short circuit in the depth of carbonation. Figure 53 shows carbonation from the faces of a microcrack in concrete that is deeper than the average carbonation depth from the surface exposed to atmosphere.



**Figure 53** Tight crack (see arrows) from surface (bottom) to interior of concrete providing a pathway for enhanced diffusion. Evidence of enhanced diffusion of  $\text{CO}_2$  adjacent to crack is observed. [Figure reproduced from ref. 10].

**Assessment of Degradation of Concrete Due to Carbonation under Extended Storage:** Carbonation is expected to occur in concrete and penetrate to depths of the reinforcement steel well within the exposure time of 300 years. For example, the data in Figure 51 show carbonation to a depth of one inch within 50 years. The carbonation of concrete can adversely impact concrete integrity by creating a low pH environment at the rebar. In conjunction with oxygen and pore water, this condition promotes reinforcement bar corrosion [1, 3, and 4]. Oxygen diffusivity is greater than carbon dioxide diffusivity in cement [5], and the onset of general corrosion of the reinforcement bar would be controlled by carbonation. However, the rate of corrosion would be controlled by oxygen diffusion to the reinforcement bar. Both  $\text{CO}_2$  and oxygen diffusion can be short-circuited by microcracks in the concrete.

With the increase in volume of the oxide product from that of the metal removed, tensile stresses, cracking and the opening of larger (~ 0.3 to 1 mm) cracks [6-8] expose additional rebar to the ambient external air/water environment leading to additional corrosion and cracking. Eventually, spallation of the concrete cover would be expected.

As stated, the depth of carbonation increases parabolically with time and  $\text{CO}_2$  level. Atmospheric concentration of  $\text{CO}_2$  is projected to rise from 380 ppm to several thousand ppm within a hundred years [9] and that increase would increase the rate of carbonation. Although the rate of carbonation would be concrete-condition-specific and environment-specific (e.g., low RH conditions tend to increase carbonation rate), the time for carbonation to depths of the reinforcement bar should be well-within a 300-year period.



### **Assessment of Concrete DCSS Structures with Carbonation - Impact to Safety**

**Function:** There is a potential for extensive cracking of a concrete pad from carbonation and rebar corrosion. The ultimate result of the corrosion will be spallation of the concrete cover and could result in an unstable foundation for a cask. This would create a cask tip-over hazard with a loss of its thermal cooling configuration and its shielding function. Additionally retrieval of the casks for transportation would also be impacted with an unstable pad, even if tip over did not occur.

There is a potential that extensive cracking of a concrete cask containing reinforcement bar but without an encasement shell could result in concrete spallation with a loss of net section and shielding function. Concrete casks without reinforcement bars would not be subjected to carbonation-induced spallation damage.

### **Concrete Degradation by Carbonation, Data Needs for EST and General**

**Recommendations for Resolution:** The concrete in the pad and cask is subject to degradation by carbonation throughout EST. There is presently no program for monitoring carbonation and technologies to support a demonstration that the pH condition at the embedded reinforcement bars in the pad and shell is benign to corrosion is needed. Specifically,

1. Monitoring/In-Service Inspection Activity: Develop monitoring/in-service inspection protocol for carbonation with acceptance criteria.
2. Testing & Analysis Activity: Investigate new technologies for NDE for carbonation in concrete.
3. Cask Demonstration Activity: As part of a cask demonstration, establish baseline and periodically monitor for carbonation and other concrete degradation mechanisms.

Carbonation degradation of the concrete is important because: 1) carbonation is expected throughout EST; 2) aging management for carbonation is not established; and 3) remediation of carbonation degradation would be costly.

### **Section 2.12.1 References**

- [1] Walton, J.C. et al. "Models for Estimation of Service Life of Concrete Barriers in Low-Level Radioactive Waste Disposal," NUREG/CR-5542, July, 1990.
- [2] Papadakis, V.G., "A Reaction Engineering Approach to the Problem of Concrete Carbonation, AIChE Journal Vol. 35, No. 10 p. 1639, (1989).
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**2.12.2 Chloride Ingress**

Chloride ingress can cause localized de-stabilization of the passive iron oxide film that is normally present on reinforcement bars embedded in the concrete. Such destabilization will lead to pitting corrosion. If carbonation, as described in section 2.12.1, would also reach the depth of the reinforcement bars, the oxyhydroxide film on steel would tend to dissolve, and gross steel corrosion would be expected leading to eventual spallation. That is, chloride ingress alone would cause localized (i.e., pitting) attack, but with carbonation, gross steel corrosion would occur and would be accelerated by the presence of chlorides.

The following discussion is drawn primarily from reference 1. Chloride attack has been modeled as a 2-step mechanism [1]. The first step is the time to initiation of corrosion due to chloride diffusion to the rebar. This initiation time can be given by a simplistic empirical model [2]:

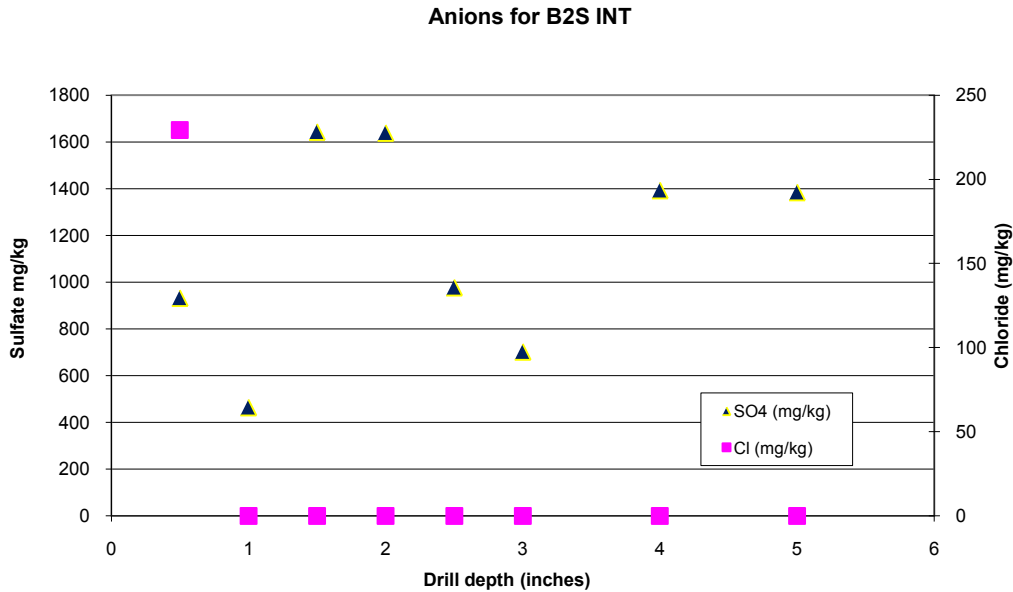
$$t_c = \frac{129X_c^{1.22}}{(WCR)[Cl^-]^{0.42}} \quad [7]$$

- $t_c$  = time to onset (yr)
- $X_c$  = thickness of concrete cover (in.)
- WCR = Water-to-cement ratio (mass)
- $Cl^-$  =  $Cl^-$  ion concentration at surface of concrete (ppm)

With an assumed concrete cover of 1 inch (2.54cm), a WCR of 0.45, and a chloride ion concentration of 10 ppm, a time of 109 years would be expected for chloride penetration to the rebar. With microcracks in the cover and the concomitant short-circuit transport compared to bulk diffusion, this time would be lessened.

Figure 54 shows the results of a chloride ingress characterization taken from the P-reactor building at SRS after approximately 60 years of service. With the assumptions above for surface chloride ion concentration and WCR, the predicted result of approximately 0.4-inch (1.0cm) ingress with the empirical model is congruent with the measurement of high chloride at the depth of 0.5 inches (1.3cm).

The second step is the corrosion rate of rebar is controlled by oxygen diffusion. Reference 1 describes reaction kinetics for iron corrosion and loss of reinforcement bar net section in terms of oxygen diffusion.



**Figure 54** Chloride ingress (left to right) limited to enhanced near-surface concentration in a concrete specimen from the SRS P-Reactor Building with 60 year's service [Figure reproduced from ref. 10].

**Assessment of Degradation of Concrete Due to Chloride Ingress under Extended Storage:** With time, the diffusion of chloride and its ingress through the cover of concrete over the reinforcement bar can lead to pitting corrosion of the bar. Additionally, general corrosion of the rebar and potential spallation of the concrete through corrosion product expansion could occur because of the carbonation and oxygen diffusion that will also be taking place. A lowering of pH at the steel/concrete interface can be caused by diffusion of CO<sub>2</sub> through the concrete cover. An oxygen feed and water are necessary conditions for the active degradation of the reinforcement bar by general corrosion. Such corrosion can lead to loss of tensile strength and concrete spallation. An oxygen/water supply can develop through microcracks in the concrete which can result from mechanical stresses and/or the combined effects of CO<sub>2</sub>, Cl<sup>-</sup>, and O<sub>2</sub> species. The

penetration of any species depends on the surface concentration of that species, thus, the driving force for chlorination in marine environments will be higher than in a non-marine environment due the chloride concentration at the surface of the concrete. Clearly, the nature of the exposure environment will have a significant impact on the rate of concrete degradation.

Although the rate of chloride ingress would be concrete-condition-specific, the time to effect carbonation to depths of the reinforcement bar in a concrete pad should be well-within a 300-year period as shown in Figure 42.

#### **Assessment of Concrete DCSS Structures with Chloride Ingress - Impact to Safety**

**Function:** The potential for extensive cracking of the concrete from chloride ingress with rebar corrosion and spallation of the concrete cover is significant and could result in an unstable pad foundation for a cask. The degraded concrete would create a cask tip-over hazard with the associated loss of thermal cooling configuration and shielding function. Additionally retrieval of the casks for transportation would also be impacted by an unstable pad.

The potential for extensive cracking of a concrete cask containing reinforcement bar, but without an encasement shell, could result in concrete spallation with a loss of net section and a reduction in the shielding function. Concrete casks without reinforcement bars would not be subjected to chloride ingress induced spallation damage but would be subjected to freeze-thaw damage and would lack the strength of reinforced concrete structures.

**Recommendations for Resolution:** Age related degradation should be anticipated in any concrete structure used in a dry storage system. Models to accurately predict such degradation as a function of concrete and rebar specifications and exposure environment are not available. The resources required to develop and validate such models are extensive and investment of such resources is not likely to be cost effective. Additionally, concrete degradation generally initiates at the surface and can be noted by visual examination. Therefore, it is recommended that visual inspection and evaluation protocols be developed and applied as necessary.

#### **Section 2.12.2 References**

- [1] Walton, J. C. *et al.* "Models for Estimation of Service Life of Concrete Barriers in Low-Level Radioactive Waste Disposal," NUREG/CR-5542, July, 1990.
- [2] Clear, K. C., Time to Corrosion of Reinforcing Steel in Concrete Slabs, Vol. 3. Performance After 830 Daily Salt Applications, Federal Highway Administration Report No. FHWA-RD-76-70, NTIS PB-258 446, 1976.

#### **2.12.3 Shrinkage of Concrete**

Excess water is typically added to the concrete mix to improve its workability during forming of a concrete structure. Shrinkage of concrete occurs initially during curing as the result of the excess water leaving the concrete. This curing period typically continues several months after placement. As excess water evaporates into the surrounding environment, tensile stresses are induced in the concrete due to internal

pressure from the capillary action of water movement, and cracks develop. Subsequent drying and shrinkage occurs in concrete for up to about 30 years. Over 90% of the shrinkage occurs during the first year and about 98% in the first five years [1]. Therefore, shrinkage cracking is not a significant aging degradation mechanism for extended storage.

**Assessment of Degradation of Concrete Due to Shrinkage Cracking under Extended Storage:** Microcracking due to shrinkage cracks start as surface cracks in concrete that can grow in depth with time [2]. It is assumed that the DCSS concrete structures were designed and fabricated with a high degree of workmanship, and that shrinkage cracking would be limited to minor superficial cracking.

However, degradation due to carbonation, chloride ingress, and chemical leaching can be promoted by surface and internal cracking in concrete.

**Assessment of Concrete DCSS Structures with Shrinkage Cracking - Impact to Safety Function:** Shrinkage cracks themselves are not significant to concrete structural performance. No impact to the safety function for either a concrete pad or a concrete cask is expected under EST.

**Data Needs and Recommendation for Resolution:** Shrinkage can cause surface cracking. Periodic monitoring of surface cracks to evaluate and trend distribution and opening width is recommended as part of a monitoring & in-service inspection program.

### **Section 2.12.3 References**

- [1] ACI 209R-92, Prediction of creep, Shrinkage, and Temperature Effects in Concrete Structures (Reapproved 2008), American Concrete Institute, 2008.
- [2] ACI 224R-01, Control of Cracking in Concrete Structures, American Concrete Institute, 2001.

### **2.12.4 Freeze-Thaw of Concrete**

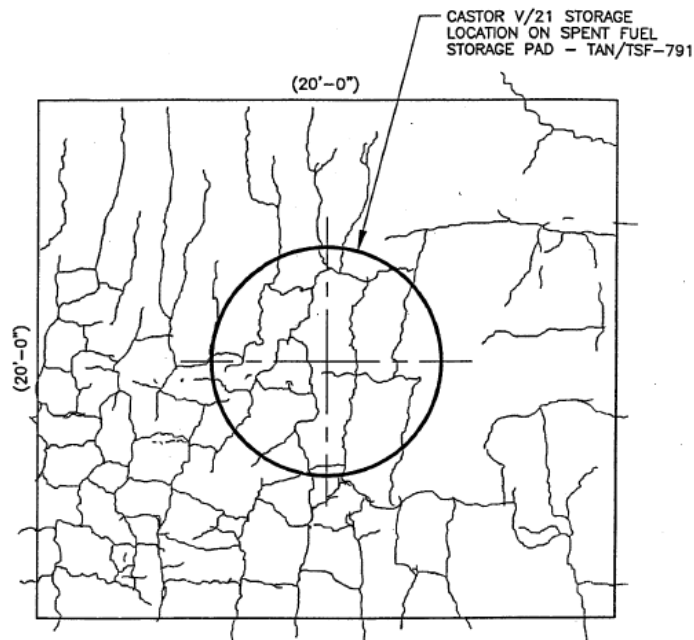
Repeated cycles of freezing and thawing may change the mechanical properties and physical form of concrete [1]. The durability of concrete to a freeze-thaw damage mechanism depends primarily on the porosity characteristics of aggregates, the presence of moisture to saturate the fines pores in aggregates, and the permeability of the hardened cement mortar matrix to the passage of water. A mathematical model for the basic processes involved in freezing and thawing of concrete is available [2], however, there is no rational model for crack growth in a freeze-thaw cycle, and any such model would be dependent on these many factors including concrete and rebar specifications, environmental conditions. Because of the large number of variables involved, such a model would be of little value to the assessment of dry storage facilities.

Freeze-thaw damage typically occurs on relatively flat surfaces such as pavement, where water remains in contact with the concrete. Concrete structures or components located in areas of the country that experience numerous freeze-thaw cycles with significant winter rainfall are more likely to exhibit damage than areas in milder climates. Specification C 33 [2] groups the areas of the U.S. into severe, moderate, and negligible

weathering regions on the basis of the weathering index. This index is the product of the average annual number of freezing cycle days and the average annual winter rainfall in inches. However, climate change over many decades is expected to result in changes including local annual precipitation [3].

Reference 1 claims that for concrete structures or components located in a geographic region subject to negligible weathering conditions, that is, a weathering index of less than 100 day-inches per year, the freeze-thaw is not a significant aging degradation mechanism and requires no further evaluation. This claim would not be expected to be applicable to EST at any specific site because of the anticipated climate change during the storage period.

Figure 55 shows a sketch of the face map indicating cracks on the pad for the Idaho cask demonstration program at Test Area North. These cracks are assumed to be a combination of shrinkage and freeze-thaw damage.



**Figure 55** Example of surface cracking on concrete pad at Test Area North in Idaho [Figure reproduced from ref. 4].

**Assessment of Degradation of Concrete Due to Freeze-Thaw under Extended Storage:** Concrete structures designed in accordance with the ACI 318-63, or a later version of ACI 318 [5], or ACI 349-85 [6], and constructed in accordance with ACI 301-66 or later [7] and using materials conforming to ACI and ASTM standards were suggested [1] to not be subject to the freeze-thaw aging degradation mechanism.

Virtually all concrete contains pore water and surface/rain water can, with time, ingress into minor discontinuities which can be enlarged during freeze-thaw cycles. Therefore, it is expected that, with time and continued exposure to freeze-thaw cycles over the many decades of EST, freeze-thaw damage would initiate. Freeze-thaw damage originates at the surface of a concrete structure and propagates as surface defects are opened. The extent and depth progression of freeze-thaw cracking in a concrete pad or even a bare

concrete cask shell would be expected to increase with repeated exposure cycles and therefore it is not self-limiting. Climate change can cause regions that have been initially classified as negligible in weathering to become severe, so freeze-thaw cannot be dismissed categorically for DCSS in EST.

Minor freeze-thaw cracking is localized on a structure and will typically not affect the strength of the concrete. Continued freeze-thaw cracks may cause “pop-outs” or local areas of spalled concrete and extensive cracking of the concrete would be expected with time under repeated freeze-thaw cycles.

In addition, degradation due to carbonation, chloride ingress, and chemical leaching can be accelerated by short-circuiting the diffusion process through surface cracking in concrete caused by freeze-thaw induced cracking.

**Assessment of Concrete DCSS Structures with Freeze-Thaw Cracking - Impact to Safety Function:** Minor freeze-thaw cracks themselves are not significant to concrete structural performance unless they continue to progress and cause “pop-outs” and extensive cracking. “Pop-outs” or local areas of spalled concrete can impact a component’s safety function as previously discussed.

There is a potential for extensive cracking of a concrete pad from freeze-thaw damage and such cracking could result in an unstable foundation for a cask creating a cask tip-over hazard with a loss of its thermal cooling configuration and its shielding function. Additionally retrieval of the casks for transportation would also be impacted with an unstable pad.

There is a potential for extensive cracking of a concrete cask without an encasement shell which could result in concrete spallation with a loss of net section and shielding function.

**Data Needs and Recommendation for Resolution:** Freeze-thaw can cause surface cracking and more extensive damage with time. Additionally, freeze-thaw processes may interact with other concrete degradation mechanisms to accelerate age related damage. Periodic monitoring of the concrete pad and DCSS components for cracking and results trending is recommended as part of a monitoring & in-service inspection program. Acceptance criteria would need to be established for the concrete inspection. The condition at which the concrete would need to be repaired or a new concrete pad would need to be constructed would be identified.

**Concrete Degradation by Freeze-Thaw, Data Needs for EST and General Recommendations for Resolution:** The concrete in the pad and cask is subject to degradation by freeze-thaw throughout EST, especially if climate change cause normal storage environments to produce many additional freeze-thaw cycles than typical for the present climate. Freeze-thaw degradation can be readily monitored in a periodic in-service inspection program. Specifically,

1. Monitoring/In-Service Inspection Activity: Develop monitoring/in-service inspection protocol for freeze-thaw damage with acceptance criteria.

2. Testing & Analysis Activity: Investigate new technologies for NDE for characterizing freeze-thaw cracking in concrete to better evaluate crack depth rather than present methods of surface opening and mapping.
3. Cask Demonstration Activity: As part of a cask demonstration, establish baseline and periodically monitor for freeze-thaw cracking.
4. Maintenance: Develop/implement a repair technology to seal surface penetrations from continued freeze-thaw damage.

The significance to EST for freeze-thaw degradation of the concrete structures is low. However, it is noted that: 1) freeze-thaw damage may occur due to climate changes in EST and may accelerate other degradation mechanisms, 2) aging management for freeze-thaw damage is not established; and 3) remediation of freeze-thaw degradation should be simple with surface sealing.

#### **Section 2.12.4 References**

- [1] Class I Structures License Renewal Industry Report; Revision 1, EPRI TR-103842, July 1994.
- [2] "Mathematical Model for Freeze-Thaw Durability of Concrete," Z.P. Bazant, J.-C. Chern, A.M. Rosenberg, and J.M. Gaidis, *J. Am. Ceramic Soc.*, 71 [9] (1988) pp. 777-783.
- [2] Specification C-33: ASTM C 33, Figure 1, "Location of Weathering Regions."
- [3] *Global Climate Change Impacts in the United States*, Thomas R. Karl, Jerry M. Melillo, and Thomas C. Peterson, (eds.). Cambridge University Press, 2009.
- [4] Dry Cask Storage Project – Phase 1: CASTOR V/21 Cask Opening and Examination, NUREG/CR-6745, INEEL/EXT-01-00183.
- [5] ACI-318-08, *Building Code Requirements for Structural Concrete and Commentary*, American Concrete Institute, 2008.
- [6] ACI-349-01/349R-01, *Code Requirements for Nuclear Safety-Related Concrete Structures and Commentary*.
- [7] ACI-301-10, *Specifications for Structural Concrete*, American Concrete Institute, 2010.

#### **2.12.5 Calcium Leaching of Concrete**

Cement components can be leached from concrete only when water passes over the surface or through cracks. Specifically, calcium compounds in the concrete can be dissolved in unsaturated water and washed away. Since the most readily soluble calcium compound in concrete is calcium hydroxide (lime), water can leach lime from concrete. When calcium hydroxide in the lime in concrete is leached away, other



cementitious constituents become exposed to chemical decomposition, eventually leaving behind silica and alumina gels with little or no strength [1].

The extent of the leaching during any given exposure to water depends on the salt content and temperature of the water [2]. Both of these variables are associated with the atmospheric conditions at the storage site which are likely to change significantly during the storage period. Therefore, exposure criteria cannot be developed and modeling concrete behavior is tenuous, at best. Because such difficulties dominate the concrete degradation arena, visual inspection of the storage sites is the most applicable current technology for site evaluation.

However, in spite of the lack of an ability to quantify concrete behavior, leaching over long periods of time can increase the porosity and permeability of concrete, making it more susceptible to other forms of aggressive attack and reducing strength. Leaching can also lower the pH of the concrete and affect the integrity of the protective oxide film of reinforcement steel [2], similar in effect to carbonation.

The dissolving and leaching actions of the percolating water are related to permeability of concrete [3]. Resistance to leaching action can be enhanced by using a concrete with low permeability [2]. A dense concrete with a suitable cement content that has been well cured will be less susceptible to leaching of calcium hydroxide because of its low permeability and low absorption.

Models for the several coupled processes involved in calcium leaching from cement hydrates have been developed [3, 4]. A recent model [4], benchmarked by immersion tests, includes a multi-component hydration model, a moisture transport/equilibrium model, and a micro-pore structure development model. The effects of w/c ratio, flyash content, and temperature on calcium leaching were assessed. Concrete with a low w/c ratio (0.25) and fly ash content of 20% were highly resistant to long-term calcium leaching showing a calcium-loss region of 5 cm after 10,000 years exposure.

**Assessment of Degradation of Concrete Due to Calcium Leaching under Extended Storage:** The requisite conditions of refreshed water and the overall kinetics of the coupled processes involved in calcium leaching renders it an insignificant degradation mechanism for the concrete in the pad or cask of the DCSS for EST. Calcium leaching is a slow process compared to the carbonation mechanism in the environment for the DCSS structures and is not rate limiting because it is not required for carbonation and other degradation processes to be operative.

**Assessment of Concrete DCSS Structures with Calcium Leaching - Impact to Safety Function:** Calcium leaching would reduce the strength and would increase micropore size in concrete; however, it is not considered to be a significant degradation mechanism so it would not be significant to impact the safety functions of the DCSS structures.

**Calcium Leaching Data Needs and Recommendation for Resolution:** Calcium leaching is not considered a significant degradation processes and therefore does not present any issues requiring resolution. Furthermore, the periodic monitoring of the concrete pad and DCSS components recommended as part of a monitoring & in-service inspection program to resolve other degradation issues are applicable to calcium leaching.

### **Section 2.12.5 References**

- [1] Troxell, G. E., Davis, H. E., and Kelly, J. W., Composition and Properties of Concrete, Second Edition (J. W. Kelly), McGraw-Hill, 1968.
- [2] Class I Structures License Renewal Industry Report; Revision 1, EPRI TR-103842, July 1994.
- [3] Bentz, D.P. and Garboczi, E.J., "Modeling the Leaching of Calcium Hydroxide from Cement Paste: Effects on Pore Space Percolation and Diffusivity," Materials and Structures 25, pp. 523-533 (1992).
- [5] Nakarai, K., Ishida, T., and Maekawa, K., "Modeling of Calcium Leaching from Cement Hydrates Coupled with Micro-Pore Formation," Journal of Advanced Concrete Technology, Vol. 4, No., 3, pp. 395-407, October 2006.

### **2.12.6 Sulfate Ingress**

The ingress of sulfate ions from the environment can cause degradation of concrete structures and is termed sulfate attack. Damage due to sulfate interactions can cause softening and cracking of the concrete leading to spallation. The cracking is the result of expansive attack of calcium hydroxides and calcium-aluminates in the concrete. The sulfate ions are present in water on the concrete surface because of dissolved sodium, potassium or magnesium sulfates from the environment.

Modeling of the rate of degradation due to sulfate ingress shows that the rate is directly dependent on the diffusivity of the sulfate ion in concrete and increases as the sulfate concentration in the exposure environment increases [1]. Other models for sulphate and freeze-thaw damage coupled model calculate the penetration depth based on sorptivity coefficient at current concrete surface temperature, current "duration" of time-of-wetness, and concrete porosity [2]. These models, using sorption as the dominant transport mechanism, show sulphate attack is due to reaction with cement to form expansive ettringite that leads to spallation of concrete, as described in reference [3].

Because the sulfate concentration in any water on the concrete pad will be low and the time of exposure generally short, this degradation mechanism is not considered significant to long term storage of spent fuel. Clearly, acid rain and industrial environments will increase the sulfate concentration and promote sulfate ingress. However, even in heavy industrial environments, sulfate ingress is slow relative to carbonation and therefore it is not considered to be the primary degradation mechanism to concrete degradation during EST.

**Assessment of Degradation of Concrete Due to Sulfate Ingress under Extended Storage:** The requisite conditions of water transport of the sulphates dissolved in water renders sulphate ingress an insignificant degradation mechanism for the concrete in the pad or cask of the DCSS for EST. Sulfate ingress is a slow process compared to the carbonation mechanism in the environment for the DCSS structures.

**Assessment of Concrete DCSS Structures with Sulfate Ingress - Impact to Safety Function:** Sulfate ingress would reduce the strength and in the absence of carbonation could cause degradation of the concrete leading to spallation. However, it is not

considered to be a significant relative to carbonation and will not impact the safety functions of the DCSS structures.

**Sulfate Ingress Data Needs and Recommendation for Resolution:** Sulfate ingress is not considered significant to cause concrete degradation to impact a safety function. However, tests to evaluate carbonation should include evaluation of sulfate ingress at the time of the test as a verification of limited sulphate ingress.

### **Section 2.12.6 References**

- [1] Walton, J. C. *et al.* "Models for Estimation of Service Life of Concrete Barriers in Low-Level Radioactive Waste Disposal," NUREG/CR-5542, July, 1990.
- [2] CONCLIFE: Service Life Predictions for Concrete Pavements and Bridge Decks Exposed to Sulfate Attack and Freeze-Thaw Deterioration, Software prepared for the FHWA by Building and Fire Research Laboratory 2001, National Institute of Standards and Technology.
- [3] Atkinson, A., and Hearne, J.A., "Mechanistic Model for the Durability of Concrete Barriers Exposed to Sulphate-Bearing Groundwaters," MRS Symp. Proc., Vol. 176, pp. 149-156, 1990.

### **2.12.7 Temperature Effects and Radiation Effects on Concrete**

Concrete has been successfully used as a structural material in nuclear facilities for over 50 years. These facilities include storage pools for spent fuel, reactor buildings, fuel reprocessing canyons and other areas that may be exposed to elevated temperatures, and that have received doses of  $\alpha$ ,  $\beta$ ,  $\gamma$ , and n radiation. The structures are typically in very good condition.

A recent publication [1] that contains an extensive literature survey of the effects of temperature on the short-term mechanical and physical properties of concrete for the nuclear industry shows that elevated temperature properties are dependent on temperature (level), heating rate, loading, external sealing of the specimen (affects moisture loss during the test), and concrete composition including its moisture state, chemical structure, and physical structure. The publication [1] also compiled the available literature on laboratory testing and specimens harvested from service-experienced structures, albeit limited, on long-term aging. A rigorous test program conducted by Central Research Institute of Electric Power Industry in Japan show results [2] that indicate, in the worst case, there is only a small decrease in compressive strength (up to 25%) and a decrease in elastic modulus (up to 50%) in concrete exposed for 8 years for temperatures up to 110°C.

An earlier publication [3] evaluated the results of the "long-term" exposure (5000 days) exposure of concrete to 65°C temperatures. The results showed an increase in both the compressive strength and elastic modulus. Similar increases were seen in concrete samples exposed for shorter times (< 5000 days) at temperatures up to 121°C. However, exposure of concrete to temperatures above 149°C decreased the compressive strength and elastic modulus [3].

Reference 1 provides sufficient data to demonstrate that the present limit for long-term, steady-state state temperature exposure of concrete should be greater than the American Society of Mechanical Engineers Boiler and Pressure Vessel Code limit of 65°C [1]. The primary mechanism of long-term, low elevated temperature (up to approximately 100°C) exposure that can affect the properties of concrete is the loss of pore water [1]. The data presented in references 1-3 suggest a threshold for thermal degradation during EST of approximately 90°C which exceeds the anticipated temperature of the concrete in DCSS service.

The only plausible degradation is water loss from the near surface regions. Shrinkage due to water loss may lead to some surface cracking that would be readily detectable. Surface cracking can promote other degradation mechanisms such as freeze-thaw cracking and the ingress of aggressive ions.

Generally, concrete is considered stable until  $10^{20}$  neutrons/cm<sup>2</sup> or  $10^{10}$  rads of gamma. A literature review [4] confirms the stability under compressive loads, but suggests that the tensile properties begin to degrade after about  $10^{19}$  neutrons/cm<sup>2</sup>. However, it is worth noting that the review also stated that “all effects on concrete due to radiation, per se, were too slight to reliably measure because of the gross effects of the increased temperature during exposure.” Additionally, the total exposure anticipated for concrete shielding structures containing high burnup MOX fuel is less than  $10^{20}$  neutrons/cm<sup>2</sup>.

The literature, as summarized in references 1-4, suggests that exposure of concrete to low level irradiation of a DCSS and low, < 90°C, temperatures, even under long times, will not significantly degrade the properties of concrete.

Additional temperature limits for concrete have been recommended in the literature [5] to protect concrete against water loss and retention of shielding capability. For neutron shields, the maximum internal concrete temperature is suggested to be 88°C, and for gamma shields, the maximum internal temperature is suggested to be 177 °C. These temperature limits are consistent with the 90°C suggested for limiting mechanical property impact during EST.

**Assessment of Concrete DCSS Structures Exposed to Temperatures and Radiation Levels under EST:** The impact of the temperature below 90°C and low radiation fields of both gamma and neutron expected during EST is expected to be insignificant. Surface cracks may form on the exposed surfaces due to loss of pore water, and these cracks could serve to enhance other forms of age related degradation which require the ingress of moisture or aggressive species. However, the exposure temperatures and radiation levels per se should not significantly impact the bulk properties of concrete associated with a DCSS.

**Temperature and Radiation Effects on Concrete - Data Needs and Recommendation for Resolution:** Long-term exposure data do not exist, and because temperature and irradiation effects have been observed during (albeit very high dose) short-term exposures, relevant data should be collected during the aging management program activity of periodic monitoring of the physical condition of the concrete. Periodic radiation monitoring would provide a measure of loss of the shielding function.

The significance to EST for high temperature and radiation degradation to concrete is low. There is no significant degradation due to temperature or radiation effects expected

to concrete structures under EST. Periodic monitoring of radiation fields during EST would identify loss of shielding function; however administrative controls could be readily established if shielding loss became significant.

### **Section 2.12.7 References**

- [1] Naus, D.J., "A Compilation of Elevated Temperature Concrete Material Property Data and Information for Use in Assessments of Nuclear Power Plant Reinforced Concrete Structures", NUREG/CR-7031; ORNL/TM-2009/175, published December 2010.
- [2] Kanazu, T., Matsumura, T., and Nishiuchi, T., "Changes in the Mechanical Properties of Concrete Subjected to Long-Term Exposure to High Temperatures," Report No. U95037, Abiko Research Laboratory, Central Research Institute of Electric Power Industry, Japan, March 1996.
- [3] Kassir, M.K., Bandyopadhyay, K.K., and Reich, M., "Thermal Degradation of Concrete in the Temperature Range from Ambient to 315°C", BNL-52384, 1996.
- [4] Fillmore, D.L., "Literature Review of the Effects of Irradiation and Temperature on the Aging of Concrete", INEL/EXT-04-02319, 2004.
- [5] Hungerford, H.E., Hönig, A., Desov, A.E., Du Bois, F., and Davis, H.S., "Concretes, Cements, Mortars, and Grouts," Engineering Compendium on Radiation Shielding," Vol. II, p. 117, R.G. Jaeger (Editor), Springer-Verlag, Berlin, Germany, 1975.

### **2.12.8 Alkali-Silicate Reactions**

Alkali-silica reactions, although a major cause of concrete degradation, are not anticipated in concretes relevant to long term dry storage, because these concretes are assumed to have been constructed with aggregates that are compatible with the hydroxyl ions in the concrete mixture. There is a simple, non-destructive field test to demonstrate the lack of alkali-silica reactions in concrete structures [1] and it is recommended that, if necessary, tests to confirm this assumption be conducted.

Chemical reactions may develop between certain mineral constituents of aggregates and alkalis that compose the Portland cement [2]. These alkalis are largely introduced in the concrete by cement, but may also be present from improper admixtures and salt-contaminated aggregates [3]. Seawater and solutions of deicing salt can also inject alkalis into concrete by action of penetration. Three types of chemical reactions may occur depending upon the composition of the aggregates. They are alkali-aggregate reaction, cement-aggregate reaction, and expansive alkalis-carbonate reaction [3].

Alkali-aggregate reaction, also known as alkali-silica reaction, occurs when aggregates that contain silica get in contact with alkaline solutions. All silica minerals have the potential to react with alkaline solutions, but the degree of reaction and ultimate damage incurred can vary significantly [3]. Alkali-aggregate reactions can cause expansion and severe cracking of concrete structures [4]. A map and data showing geographic areas known to yield natural aggregates suspected of, or known to be capable of, alkali-silica reaction is included in ACI 201.2. The reactivity of such aggregates might not be

recognized until the structures are over 20 years old, even if used in combination with high alkali cement [3]. It is recommended that if an NPP is close to one of the areas shown on the ACI 201.2 map, that the associated concretes be reviewed and, if necessary, evaluated to assure the absence alkali-silicate reactions.

Cement-aggregate reaction, which is a second type of reaction similar to alkali-aggregate reaction, occurs between the alkalis in cement and siliceous constituents of the aggregates, and is complicated by environmental conditions that produce high concrete shrinkage and alkali concentrations on the surface due to drying (aging) of the concrete [3].

Expansive alkali-carbonate reaction is a third type of reaction between certain carbonate aggregates and alkalis, which in some instances produces expansion and cracking of concrete. Certain limestone aggregates have been reported as reactive [3]. Aggregates that react with alkalis can induce expansion stress in the concrete to a varying degree of severity, and in some cases, the expansion stresses may be high enough to produce cracking of the concrete structures. This cracking is irregular and has been referred to as “map cracking” [3].

The operating histories of nuclear power plant concrete structures do not indicate that structural integrity of these concrete structures is significantly affected by alkali-aggregate reactions. Aggregates used in the concrete can be tested and petrographically examined in accordance with ASTM Practice C 295 and Test Method C 227 to determine the potential for reactivity with alkalis. When aggregate reactivity is considered a possibility, accepted construction practices place limitations on cement alkalis used during construction and/or require an additive such as an effective pozzolan to be used in combination with the cement. Reaction with aggregates is not a significant aging degradation mechanism for concrete structures designed and constructed in accordance with provisions given in the ACI 201.2. However, in the unlikely event that concretes used in construction did not meet accepted practices, the inspection and monitoring protocols be modified to increase the frequency of evaluation.

**Assessment of Degradation of Concrete Due to Alkali-Silicate Reactions under Extended Storage:** Alkali-silicate reactions are caused by incompatibilities between the aggregate and cement in the concrete mixture. The assumption made in this evaluation is that the concrete was comprised of and fabricated with aggregates, cement, and admixture constituents that are compatible. No fundamental aging mechanism, including environmental effects, is involved in this degradation mechanism, and it is judged as insignificant to EST.

**Assessment of Concrete DCSS Structures with Alkali-Silicate Reactions- Impact to Safety Function:** Alkali-silicate reactions can be insidious to impact strength by increasing micropore/microcrack size in concrete; however, it is not considered to be a significant degradation mechanism, so it would not be significant to impact the safety functions of the DCSS structures.

There is a simple, non-destructive field test to demonstrate the lack of alkali-silica reactions in concrete structures [1] and it is recommended that, if necessary, such a test be conducted to confirm this assumption.

### **Section 2.12.8 References**

- [1] Colored Dye Field Test to Detect Alkali Silicate Reaction, ASR Detect\*, U.S. Patent 5,739,035 Los Alamos National Laboratory.
- [2] Suoulikidis, T., Tsakopoulos, A., and Moropoulos, T., "Accelerated Rebar Corrosion When Connected to Lightning Conductors and Protection of Rebars with Needle Diodes Using Atmosphere Electricity," Contained in Publication ASTM STP 906, "Corrosion Effects of Stray Currents and the Techniques for Evaluation Corrosion of Rebars in Concrete."
- [3] Class I Structures License Renewal Industry Report; Revision 1, EPRI TR-103842, July 1994.
- [4] Stanlon, T. E., "Expansion of Concrete Through Reaction Between Cement and Aggregate," American Society of Civil Engineers, Proceedings, Vol. 66, December 1940.





### 3 RECOMMENDATIONS FOR ACTIVITIES TO ESTABLISH THE TECHNICAL BASES TO ENABLE EST

Recommendations are provided for activities to resolve materials aging issues in the technical bases needed to demonstrate the continuing integrity of SSCs and their ability to maintain functions important to safety during EST. Aging management activities, such as monitoring and preventive maintenance, are essential to provide condition assessments through the storage period. Other activities are suggested to strengthen the set of information to evaluate materials reliability in the technical bases for EST.

The recommended activities are grouped in the following four areas:

1. **Monitoring Program/In-Service Inspection** – Periodic actions to provide a condition assessment of the SSCs through examinations and non-destructive testing are recommended. Acceptance criteria for service-induced degradation of the components should also be established. The decision to keep in service or repair/replace components and/or recover the fuel for repackaging would follow identification and characterization of service-induced flaws or degraded materials conditions.
2. **Maintenance Activities** – Periodic actions to provide preventive maintenance required to maintain the DCSS throughout the EST are required. These actions should address items identified in the inspection programs.
3. **Testing and Analysis** – Laboratory full-scale separate-effects testing and/or additional modeling and analysis of degradation phenomena to provide verification of the presence or absence of age-related degradation should be conducted. If degradation is expected, data to support constitutive model(s) for the exposure-time dependence of materials properties should be obtained.
4. **DCSS Demonstration** – Integrated testing of SSCs of DCSS in storage setting. The demonstration would:
  - a. include a fuel safe storage demonstration, including those at challenging conditions (high burn-up fuels, for example)
  - b. confirm degradation and aging models
  - c. demonstrate NDE techniques needed for an ISI Program
  - d. provide a test bed for new remote sensors
  - e. provide information from fuel DE removed from the demonstration cask

Additionally, activities for EST storage will require plan development and potential new dry-transfer system design for fuel recovery, including for degraded fuel and SCCs, and especially for envisioned orphan sites.

### **3.1 Maintenance Program**

Polymeric and metallic seals installed on casks with bolted closures are expected to deteriorate over time. The integrity of these gasketed seals will need to be periodically confirmed by leak test and provisions must be made to replace degraded seals. This will not be a trivial exercise as it may require removing the cask lid, replacing the seal(s), reinstalling the lid, and reestablishing the cask gas fill, all while maintaining personnel radiation exposure ALARA.

The surfaces of metal casks may need periodic re-coating. Bolts installed on casks with bolted closures may need periodic replacement.

The selection of items for preventive maintenance and the preventive maintenance schedule should be integrated with the monitoring program.

### **3.2 Monitoring & In-Service Inspection Program**

Monitoring and in-service inspection provides a condition assessment of the DCSS SSCs, and enables trending of material condition with time. A summary of domestic and international experience in DCSS surveillance (monitoring) is provided in chapter 4 of reference [1]. For EST, a standing program is recommended to be developed and implemented.

The inspection program recommended for EST should be consistent with the requirements of ASME Section XI, with scope components consistent with the Safety Analysis Reports, Certificates of Compliance, and Technical Specifications of the individual licensees. With storage times expected to extend to greater than 100 years, it is clear these inspection programs will have to include mechanisms for maintenance and repair of the ISFSI components which would be expected to degrade over time. Additionally, the inspection protocols will change as new data are obtained and new inspection technologies emerge.

The element of an ISI program for DCSS would follow those in an ASME SCXI-type program and include the scope, extent, NDE methods, frequency, acceptance criteria, and records. Through acceptance criteria of an SCC that is identified and characterized with service-induced degradation, decisions for repair/replacement and also for fuel recovery for repackaging would be determined.

Several SSCs are suggested to be subject to in-service inspection. Employment of inspection programs has shown to be an effective tool to ensure that confinement structures in the commercial nuclear industry meet design life requirements [2].

The canister and especially its closure weld are also subject to failure from stress corrosion cracking as a result of the interaction of environmental conditions with residual stresses associated with the welds. In general, the gas fill of welded containers is not monitored, so detection of an SCC failure (wall penetration) may be very difficult. Insipient cracking of containers could be detected by penetrant or ultrasonic interrogation of the canister welds, but would require access to the welds which is not currently available. Methods of remotely performing periodic inspections of container welds should be developed. Obtaining the access to perform necessary inspections *in situ* may require modification or redesign of the ISFSI.

As discussed, the concrete in casks, vaults, and pads will be expected to degrade as a result of environmental exposure, chemical attack, etc. This could lead to cracking, spalling, and/or cement/aggregate reactions, potentially leading to corrosion of any reinforcing steel members. Inspection of the concrete structures will primarily be performed by visual inspection, but may also include inspection by sonic or ultrasonic techniques, impact tests, radiography, radar, and thermal techniques.

Periodic visual inspection of exposed concrete cover to monitor for signs of degradation (i.e., cracking, spalling, and alkali aggregate reactions) will be the primary foundation of this program element. ACI 201.1R (Guide to conducting a visual inspection of concrete in service) will serve as the basis by which these inspections are performed. A frequency will be selected to ensure the changes in condition are noted prior to degradation proceeding beyond the point of mitigation. Reports will detail the condition of the concrete in a manner such that further inspections will be able to discern if any change has occurred since previous inspections. If necessary, non-destructive (e.g., ultrasonic techniques for crack measurement) and destructive (e.g., core sampling) evaluation will be performed as part of the inspection to determine if degradation is in an active or dormant state.

### **3.3 Testing and Analysis**

Several gaps in the technical bases for materials aging that may be addressed with testing and analysis include the following:

**DHC testing and modeling of fuel cladding alloys:** There are many factors that affect delayed hydride cracking in the zirconium cladding alloys. These factors, combined with the very long time of storage, make it very challenging to realistically test and predict DHC performance. Hydride reorientation is a large factor in DHC. The hydride orientation is influenced by the alloy composition, temperature differentials, texture, grain boundary and microstructure, presence of hydrides (preference to continue growth of existing versus nucleate new hydrides), plastic strains and stress states, to list the major contributors. To accurately characterize DHC requires that all of these and other factors such as stress raisers from partial throughwall cracks and flaws need to be considered. Thus, it is suggested that initially a multiple approach be taken to develop basic data on DHC that includes a) worst case analysis of large quantity of clad breaches during long term storage, b) characterization of the morphology of a DHC throughwall crack and the risk for fuel relocation, and c) general characterization of the relative time frames when DHC has the most risk of occurring. Expansion of these three data needs suggests that:

a) In considering the worst case condition one approach could be to assume a short hairline crack exists in all or most of the fuel rods and determine what impacts the internal gas release would have on the safety aspects of the storage system. Additionally, one could determine how the release would affect retrieval and transport and what mitigating actions could be taken.

b) To characterize the DHC, crack samples of the various cladding types could be tested using existing pressurized tube samples to represent the nominal crack growth geometry. Relatively high stresses and temperatures near, but below, the DBTT could be used to enhance the probability of generating a DHC. Care should be taken to limit the volume or path of the pressurization gas so that there is not excessive stored energy in the system. Testing of different alloys in different conditions could be done and the

resultant OD surface crack morphology then documented and evaluated for risk of fuel relocation.

c) At temperatures above the DBTT, the zirconium microstructure has some ductility even with the hydrides present. The ductility is strongly related to the irradiation damage, also, and the DBTT is affected by the hydrogen content as discussed in a previous section. Hydride reorientation also is a very important factor for consideration and is very alloy, stress, strain, microstructure and temperature dependent, and other factors also play a part as listed above. The DHC testing plan should consider using both internally pressurized clad samples and some fracture toughness-type samples with controlled cracks present. The samples should be representative of the microstructure condition present after a typical dry out anneal, including residual radiation damage. This is important, because if microstructure changes and some annealing of irradiation damage occur, these changes will affect the hydride orientations and basic material matrix properties. Testing temperatures should be chosen to map particularly the conditions around the DBTT. Data from these tests can be used to 1) estimate the probability for long term DHC failure rates and frequencies, 2) determine if there are temperatures above which the probability for DHC is very small and that handling and transport has an acceptably low risk, and 3) determine the effects of various parameters which can enhance future testing by allowing a focus on variables of consequence and limiting the testing scope by excluding variables that are not significant.

**Hydrogen Embrittlement testing and modeling of fuel cladding alloys:** Hydrogen embrittlement is expected to an extent in all zircaloy cladding alloys in EST. With temperature decrease, the cladding will transition from ductile tearing behavior to brittle fracture as the failure mode. Deformation mode will also be expected to change with temperature. There is a need to establish fracture behavior for a set of alloys, establish fracture methodology, and to establish analysis protocol for transport of fuel in a transport cask.

The hydrogen embrittlement concern is partially related to the DHC issue, but while DHC is a longer term crack velocity-type of mechanism, hydrogen embrittlement is associated with the general reduction of ductility of the clad metal matrix and the risk for mechanical failure due to excessive stresses/strains associated predominately with handling and accident conditions. After operation in the reactor, the fuel assembly cladding has both extensive radiation damage and a buildup of precipitated hydrides in the microstructure. Like other cladding material characteristics, the degree of hydride embrittlement will be affected by a number of variables, including alloy chemistry, microstructure, level of hydrogen in the clad, hydride orientations, test temperatures, irradiation history, and inherent cladding flaws, both from manufacture and from operation.

Embrittlement testing should focus on testing in the circumferential (hoop) direction using ring sample testing, for example, from which a lot of past data has been generated. The testing scope should also include axial testing to generate data for conditions where axial loading may be predominating. Some ring crush testing and expansion testing should be performed using expanding mandrels from which there is also some data generated in the past. The basic tests (ring and maybe axial) should encompass the range of expected temperature exposures from room temperature to 400°C and address the major variables of alloy, hydrogen level, hydride orientation,

microstructure (including crack / flaws) and strain rate. Data on tensile strengths, ductility, elastic constants, and hardness can be generated.

Data from these tests will support the analysis of fuel performance during both normal and off-normal storage, handling and transport conditions. Tests of samples from the same rod, but at different axial elevations such that there are significant differences in hydrogen levels, will also provide data on the relative effects of radiation and hydrogen on clad ductility.

**Creep Testing:** There are two major regions where gaps exist in creep data for long-term fuel storage; during high temperature dry out anneal where the temperatures and pressures are relatively high and during the long term storage where temperature and pressures are decreasing and relatively low for significant creep to occur. Creep data generated in these two regions can be used to characterize and predict the creep condition of the fuel cladding during long-term storage.

The typical creep testing procedures used for testing of unirradiated and as fabricated cladding are applicable to the spent fuel testing also. The techniques are based on an accurate measurement of the cladding OD to a 2 micron or better accuracy, typically using laser micrometers. There are additional problems with spent fuel samples since they will have an oxide coating that will have varying dimensions and also may experience minor spalling during the creep test. Consideration should be given to removal of small sections of the oxide to allow accurate dimensional data on exposed metal surfaces. The creep samples would be defueled fuel rod sections approximately 25 cm long. For accurate creep measurements the gage length should be at least 10 times the diameter to remove end fixturing effects. The clad sample would have end fittings that seal one end and allow internal pressurization through the other end. The creep samples would be placed in a vacuum or inert gas furnace and pressurized via a manifold that can serve multiple samples and maintain a constant pressure.

Testing parameters of time, pressure (hoop stress) and temperature would be chosen to encompass the predicted exposure conditions. For example if the dry out temperature can reach 400°C then testing should be done, for example, at temperature internals of 350°C, 400°C and 450°C to cover the potential exposure ranges. Since the dry out time at these temperatures is relatively short the test times should be from a few hours to a few days. The samples can be measured after each time interval and then returned to the furnace for additional exposure. The pressures would be determined based on the range of representative fuel rod internal pressures at the test temperatures.

For the longer term lower temperature creep representing conditions after the dry out anneal similar samples and test procedures could be used except that the test conditions would be different. The test times may need to extend to 3 or 5 years. It is important in projecting the creep rate to the long storage times to have confidence in obtaining a quasi-steady state creep rate for the condition, or if still in primary creep to have an understanding of that (upper bounding) creep rate.

Creep rates will be strongly alloy and microstructure dependent. Other factors that will affect the creep rates will be the hydride content and morphology, the degree of anneal during dry out, variations in oxygen levels and, maybe, the irradiation damage, although that will approach saturation early in the reactor operation.

**DCCG:** The relative influence of diffusion controlled cavity growth (DCCG) on cladding performance during long term dry storage is unknown, but there are opportunities to develop information without expending a lot of additional effort by using samples from other testing programs such as the creep, DHC, and hydrogen embrittlement tests. For example, after the creep tests are completed a section of the creep sample can be examined under SEM to determine the presence or lack thereof of cavities in the microstructure. Likewise, the fracture surface of DHC samples and of some hydrogen embrittlement samples can be examined to assess the presence of cavities. The examination scope should include different alloys, microstructures and temperatures.

**SCC Testing and Model of Canister:** The stainless steel canister is a critical component of the DCSS that provides the confinement barrier. Stainless steel is subject to stress corrosion cracking, and may be subject to pitting and crevice corrosion. For containers that are exposed to outside air, humid oxidation occurs on the outside of the container; especially in marine environments. With longer storage times, temperatures drop to lower values and condensation on the canister would be more likely. Condensation allows the wet corrosion mechanisms to occur on the outside of containers exposed to outside air.

A fabricated prototypic canister(s) is recommended for the testing. Tests and analysis would consider the following materials and environments: The critical regions for SCC consideration are the welded joints of the canister—these welded joint regions provide sites for residual stresses combined with either humid air or condensate to potentially lead to incidents of SCC in both the weld and weld HAZ regions. SCC at the weld regions of the DCSS could potentially impact both DCSS confinement integrity and fuel retrievability.

SCC testing of prototypic weld region materials should be conducted under the following conditions: 1) environment—a) humid air, to include chlorides for marine environment simulation and additional germane atmospheric contaminants and, b) condensate, also to include chlorides for marine environment simulation and additional germane atmospheric contaminants, 2) temperature—isothermal temperature testing over a range of temperature to bound the anticipated canister wall temperatures, 3) coupled radiation—some testing should be conducted with coupled gamma irradiation in order to understand any impact of gamma radiation on the SCC corrosion processes and to assess the impact of radiolysis decomposition products on the SCC processes, and 4) test configuration—SCC testing should also include U-bend weld samples configured to provide varying levels of stress and to be tested in a controlled environment capsule test for the humid environment testing in radiation fields to capture radiolysis effects in air/moisture system. Testing for condensate effects can also be performed using weld u-bend samples immersed in shallow condensate vessels—the goal of these u-bend tests is to provide a go-no-go measure of SCC response of the weld region materials and also to provide an indication of the incubation time to crack initiation. Finally, testing to determine accurate growth rates for materials/stress levels that indicate a cracking response through u-bend testing can be conducted using fully instrumented bolt-loaded compact tension specimens.

For long-term predictive capability, data from the aforementioned testing must be combined to produce a model to represent the effect of the various test variables on SCC response of the DCSS weld region materials.

### **3.4 Cask Demonstration Considerations**

An integrated, full cask demonstration project could provide important degradation information and/or confirm degradation models to support establishment of the technical bases for EST. A cask demonstration could involve three types of monitoring and data collection: 1) in situ monitoring of cask internals with sensors; 2) periodic removal of fuel and other internals for destructive examination; and 3) external NDE of cask, canister, and pad components. The specification of functions and performance requirements for a cask demonstration project are outside the scope of this report. The following discussion lists considerations for a cask demonstration.

Cask external and internal parameters could be monitored. Environmental parameters important to degradation are: temperature, environment (including gas atmosphere), radiation, and chemical effects on the fuel in storage and all storage system components.

Monitoring the conditions inside the storage canister or cask would include, thermocouple probes to determine the temperature gradient along and across the fuel bundles, and periodic or continuous gas monitoring for pressure and gas composition that would correspond to fuel cladding breaches and fission gas/volatiles release.

It is not generally straightforward to obtain cladding material properties *in situ* during storage. An exception may be cladding emissivity that would change with oxide thickness and structure on the cladding. With development of remote technologies, monitoring the SNF *in situ* could provide information about changes in cladding ductility and the potential for cladding creep; the formation, orientation, and distribution of zirconium hydrides in the cladding; and possibly information about the condition of the fuel pellets. Periodic removal of the fuel for destructive examination would validate the remote technologies.

As an example, an instrumented dry storage cask could include sensors to monitor for:

- temperature (of fuel and cask internals),
- radiation fields (gamma and spontaneous neutron fluxes),
- “air-borne” contamination (monitor for gaseous and volatile fission products),
- gas composition (monitor for initial cover gas and air contamination),
- gas conductivity,
- gas pressure,
- gas moisture, and
- camera(s) (for fuel cladding and cask internal conditions including corrosion, swollen fuel, other evidence of materials degradation).

Redundant thermocouples, passive sampling tubes, and fiber optics provide a robust, long-term platform for information-gathering. Information from the instrumented cask would serve to provide a real-time surveillance of the materials and also data to compare against predicted materials performance models.

External cask examinations could also be performed. For example, a bolt could be retrieved for destructive testing and a new bolt installed. Destructive examinations could be ultimately performed on a cask removed from service.

The cask demonstration would also provide a test bed for new NDE methods and remote sensor technologies. New NDE methods, new sensor technology and remote telemetry of data, and new DE methods could be demonstrated and deployed in the cask demonstration program. Adaptation of these new technologies would be expected to follow national consensus of their efficacy and consensus for their application in DCSSs.

### **Section 3 References**

- [1] Technical Bases for Extended Dry Storage of Spent Nuclear Fuel, EPRI report number 1003416, Final Report, December 2002.
- [2] NUREG/CR 4652 "Concrete-Component Aging and Its Significance Relative to Life Extension of Nuclear Power Plants," D. J. Naus, September 1986.



#### 4. OFF-NORMAL AND ACCIDENT CONSIDERATIONS

Normal conditions of operation include those environments specified in the Environmental Report, Site Characteristics, and/or the Operating Procedures for the local site.

Off-normal conditions/events are considered to include those events that may reasonably be expected to occur during the life of the cask system and that exceed normal conditions. Types of off-normal conditions include loads associated with equipment malfunction or inadvertent loads during equipment transfer and environmental conditions such as low or high extreme ambient temperatures. Events due to climate change, such as local heavy/low rainfalls and temperature increase/decreases discussed in [1] would be expected in EST for a DCSS. The extent of these changes will be dependent on geographic location and may be categorized under off-normal conditions. Even under climate change conditions for the next 300 years, no conditions are expected to fall outside of the design space for normal operation environmental conditions (e.g., only several degrees change in average temperature would be expected) with the exception of increases in atmospheric CO<sub>2</sub> concentration and industrial pollution.

Accident analyses are ISFSI-specific. The types of accidents considered are seismic, tornado (wind and missile), cask tip-over, fire, explosion, and ice/snow overload. These events should be at the same magnitude and frequency for both present license periods and EST period.

Explicit consideration of the impact of off-normal and accident events on dry storage system performance is given in references 2 and 3. The impact of off-normal or accident events on material condition of the SSCs of the dry storage systems is outside the scope of this report.

Materials must be considered in their degraded (if any) condition as required by reference 4 to withstand design-basis normal, off-normal, and accident conditions. The potential for materials to degrade and impact safety functions during normal conditions of EST were discussed in Section 2 of this report. Degraded conditions, that could impact the performance of the SSCs of the DCSS for normal conditions, would also impact off-normal and accident conditions.

The following list highlights the potential impact of low burnup and HBU fuel in EST with respect to off-normal and accident conditions. The specific response of an SSC in a degraded condition is dependent on the specific off-normal or accident event, and is outside the scope of this report.

- HBU fuel contains a fine-grain size rim region on the fuel pellet that would be expected to cause a larger fraction of a fines source term for accident conditions in transportation.
- Both low burnup and HBU fuel are susceptible to brittle fracture and may fail at low temperature, high loading off-normal or accident conditions.
- Degraded (cracked) canisters with tight hairline cracks may exhibit large crack openings if sufficiently loaded under off-normal or accident conditions.

- Degraded reinforced concrete pads and overpacks may not have requisite capacity in off-normal or accident events, as would undegraded concrete.

The aging management activities described in Section 3 of this report could provide resolution to these materials issues. For example, a crack identified and characterized in an inspection of a canister before its transportation could be dispositioned with acceptance criteria that include off-normal and design basis loading events.

#### **Section 4 References**

- [1] Global Climate Change Impacts in the United States, Thomas R. Karl, Jerry M. Melillo, and Thomas C. Peterson, (eds.). Cambridge University Press, 2009.
- [2] NUREG-1536, Standard Review Plan for Dry Cask Storage Systems. <http://www.nrc.gov/reading-rm/doc-collections/nuregs/staff/sr1536/>.
- [3] NUREG-1567, Standard Review Plan for Spent Fuel Dry Storage Facilities. <http://www.nrc.gov/reading-rm/doc-collections/nuregs/staff/sr1567/>.
- [4] NUREG-1927, Standard Review Plan for Renewal of Independent Spent Fuel Storage Installation Licenses and Dry Cask Storage System Certificates of Compliance. <http://www.nrc.gov/reading-rm/doc-collections/nuregs/staff/sr1927/>.

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An evaluation of potential degradation of materials in the Systems, Structures, and Components (SSCs) of dry cask storage systems (DCSS) for Independent Spent Fuel Storage Installations (ISFSIs) has been performed to identify issues in the technical bases for regulating storage well beyond the present allowable storage time of 120 years that includes 60 years pool storage and 60 years dry storage. The evaluation included low ( $\leq 45$  GWd/MTU) and high burnup (HBU) ( $> 45$  GWd/MTU up to approximately 62.5 GWd/MTU) fuel, and was performed to determine if: 1) degraded conditions could be anticipated to develop in the materials of the SSCs over an extended storage and transportation (EST) period ( $> 120$  years); and 2) the anticipated degradation would challenge functions important to safety during storage and during post-storage transportation including fuel retrieval for final disposition. The approach followed a general Aging Management Review process by: 1) identifying the materials of construction of the SSCs; 2) evaluating their susceptibility to degradation from environmental conditions of exposure; and 3) evaluating the potential for degradation to impact the safety functions required by the present Code of Federal Regulations for dry storage systems and spent fuel transportation in 10 CFR Part 72 and 10 CFR Part 71, respectively. The evaluation was performed using extrapolation of available data and materials degradation models with consideration of service histories of similar materials systems, and with expert judgment.

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