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Standard Guide for Drying Behavior of Spent Nuclear Fuel¹

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1. Scope

1.1 This guide is organized to discuss the three major components of significance in the drying behavior of spent nuclear fuel: discusses three steps in preparing spent nuclear fuel (SNF) for placement in a sealed dry storage system: (1) evaluating the needneeds for drying, drying spent nuclear fuel, and confirmation of drying the SNF after removal from a water storage pool and prior to placement in dry storage, (2) drying the SNF, and (3adequate dryness.) demonstrating that adequate dryness has been achieved.

1.1.1 The guide addresses drying methods and their limitations in drying spent nuclear fuels that have been in storage at when applied to the drying of SNF that has been stored in water pools. The guide discusses sources and forms of water that may remain in the SNF, itsthe container, or both, both after the drying process and has been completed. It also discusses the importance important and potential effects they may have on fuel integrity, and container materials. of the drying process and any residual water on fuel integrity and container materials during the dry storage period. The effects of residual water are discussed mechanistically as a function of the container thermal and radiological environment to provide guidance on situations that may require extraordinary drying methods, specialized handling, or other treatments.

1.1.2 The basic issue issues in drying is are: (1) to determine how dry the SNF must be in order to prevent issuesproblems with fuel retrievability, container pressurization, or container corrosion. Adequate corrosion during storage, handling, and transfer, and (2dryness may be readily achieved) to demonstrate that adequate dryness has been achieved. Achieving adequate dryness may be straightforward for undamaged commercial fuel but may become a complex issue complex for any SNF where cladding damage has occurred during fuel irradiation, storage, or both, prior to or during placement and storage at the spent fuel pools. Dryness issues Challenges in achieving adequate dryness may also result from the presence of sludge, erud, CRUD, and other hydrated compounds connected to the SNF that any other hydrated compounds. These may be transferred with the SNF to the storage container and may hold water and resist drying efforts.drying.

<u>1.1.3 Units are given in both SI and non-SI units as is industry standard. In some cases, mathematical equivalents are given in parentheses.</u>

1.2 This standard <u>only discusses SNF drying and does not purport to address all of the handling and safety concerns, if any,</u> associated with its use. the drying process(es). It is the responsibility of the user of this standard to establish appropriate safety and health practices and determine the applicability of regulatory limitations prior to use to meet regulatory requirements prior to and during use of the standard.

2. Referenced Documents

2.1 ASTM Standards:²

C859 Terminology Relating to Nuclear Materials

C1174 Practice for Prediction of the Long-Term Behavior of Materials, Including Waste Forms, Used in Engineered Barrier Systems (EBS) for Geological Disposal of High-Level Radioactive Waste

C1562 Guide for Evaluation of Materials Used in Extended Service of Interim Spent Nuclear Fuel Dry Storage Systems 2.2 ANSI/ANS Standards:³

ANSI/ANS 8.1-1998 Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors ANSI/ANS-8.7-1998 Nuclear Criticality Safety in the Storage of Fissile Materials

¹ This guide is under the jurisdiction of ASTM Committee C26 on Nuclear Fuel Cycle and is the direct responsibility of Subcommittee C26.13 on Spent Fuel and High Level Waste.

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² For referenced ASTM standards, visit the ASTM website, www.astm.org, or contact ASTM Customer Service at service@astm.org. For *Annual Book of ASTM Standards* volume information, refer to the standard's Document Summary page on the ASTM website.

³ Available from American National Standards Institute (ANSI), 25 W. 43rd St., 4th Floor, New York, NY 10036, http://www.ansi.org.



ANSI/ANS-57.9 American National Standard Design Criteria for Independent Spent Fuel Storage Installation (Dry Type)

2.3 Government Documents:⁴The U.S. government documents listed in 2.3 or referenced in this standard guide are included as examples of local regulations and regulatory guidance that, depending on the location of the dry storage site, may be applicable. Users of this standard should adhere to the applicable regulatory documents and regulations and should consider applicable regulatory guidance.

Title 10 on Energy, Code of Federal Regulations, Part 60, 10 CFR 60, U.S. Code of Federal Regulations, Disposal of High Level radioactive Wastes in Geologic Repositories

Title 10 on Energy, Code of Federal Regulations, Part 63, 10 CFR 63, U.S. Code of Federal Regulations, Disposal of High-Level Radioactive Wastes in Geologic Repository at Yucca Mountain, Nevada

Title 10 on Energy, Code of Federal Regulations, Part 71, 10 CFR 71, U.S. Code of Federal Regulations, Packaging and Transport of Radioactive Materials

Title 10 on Energy, Code of Federal Regulations, Part 72, 10 CFR 72, U.S. Code of Federal Regulations, Licensing Requirements for the Independent Storage of Spent Nuclear Fuel and High-Level Radioactive Waste

<u>Title 10 on Energy, Code of Federal Regulations, Part 961, 10 CFR 961 U.S. Code of Federal Regulations, Standard Contract</u> <u>for Disposal of Spent Nuclear Fuel and/or High-Level Radioactive Waste SFST-IST-1, Damaged Fuel</u>

3. Terminology

3.1 *Definitions*—Terms-For definitions of terms used in this guide are as defined in Practice<u>but not defined herein, refer</u> C1174 or, ifto Terminology C859 not defined<u>or Practice</u> C1174 therein as per their common usage, except where defined specifically for this guide as described as follows.

3.2 Definitions of Terms Specific to This Standard: Refer to SFST-ISG-1 for additional definition details.

<u>3.2.1 breached spent fuel rod, or failed fuel, n</u>—spent fuel rod with cladding defects that permit the release of gas from the interior of the fuel rod. A breached spent fuel rod may also have cladding defects sufficiently large to permit the release of fuel particulate. A breach may be limited to a pinhole leak or hairline crack, or may be a gross breach.

3.2.2 *CRUD*, *n*—*in nuclear waste management*, deposits on fuel surfaces from corrosion products that circulate in the reactor coolant. Compositions <u>of the deposits</u> reflect materials exposed to coolant and activation products formed during irradiation. Term was originally an acronym for "Chalk River Unidentified Deposits."

3.2.3 damaged fuel, <u>SNF</u>, n—in nuclear waste management, nuclear fuel that has been geometrically altered in form/shape to a degree that may affect retrievability from a (licensed) storage system or make it unsuitable for transport in a licensed cask. any fuel rod of fuel assembly that cannot fulfill its fuel-specific or system-related functions.

3.2.4 *disposal, n—in nuclear waste management,* the emplacement of radioactive <u>materials and</u> wastes in a geologic repository with the intent of leaving itthem there permanently. **10 CFR Part 63.2**

3.2.4 failed fuel, n—in drying of spent nuclear fuel, any breach, such as hairline cracks or holes in a cladding that permits water into a fuel element.

3.2.5 getter, *n*—in nuclear waste management, a material (typically a solid) used to chemically react with certain gases (for example, H_2 , O_2 , H_2O vapor) to form a solid compound of low vapor pressure.

3.2.5.1 Discussion-

A getter may also be used to absorb impurities in chemical and metallurgical processes. Some fuel rod designs include an internal getter to remove residual hydrogen/moisture from the internal rod atmosphere.

<u>3.2.6</u> grossly breached spent fuel rod, n—a subset of breached rods. A breach in spent fuel cladding that is larger than a pinhole leak or a hairline crack and that may permit fuel particulate release.

3.2.7 *independent spent fuel storage installation (ISFSI), n*—a complexsystem designed and constructed for the interim storage of spent nuclear fuel and other radioactive materials associated with spent fuel storage. 10 CFR Part 72

<u>3.2.8 *intact SNF*, *n*—any fuel that can fulfill all fuel-specific and system-related functions, and that is not breached. Note that all intact SNF is undamaged, but not all undamaged SNF is intact, since in most situations, breached spent fuel rods that are not grossly breached will be considered undamaged.</u>

⁴ Available from U.S. Government Printing Office Superintendent of Documents, 732 N. Capitol St., NW, Mail Stop: SDE, Washington, DC 20401, http:// www.access.gpo.gov. The Code of Federal Regulations is available at https://www.gpo.gov/fdsys/browse/collectionCfr.action?collectionCode=CFR. SFST-ISG-1 is available at http://www.nrc.gov/reading-rm/doc-collections/isg/spent-fuel.html.

⁵ The boldface numbers in parentheses refer to the list of references at the end of this standard.

⁶ Considerable work is currently in progress to quantify the effects of radiolysis in both the Pu and U oxyhydroxide systems.

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3.2.9 packaging, <u>or SNF storage container</u>, n—in nuclear waste management, an assembly of components used to ensure compliance with the requirements of Title 10 of the Code of Federal Regulations, (CFR) Part 72 applicable requirements for independent storage of spent nuclear fuel and high-level radioactive waste or 10 CFR Part 71 for transportation of radioactive materials.

<u>3.2.10 pinhole leaks or hairline cracks, n—minor cladding defects that will not permit significant release of particulate matter</u> from the spent fuel rod, and therefore present a minimal concern during fuel handling and retrieval operations. (See discussion of gross defects for size concerns.)

3.2.11 *repository, geologic repository, n— in nuclear waste management*, a disposal site, a permanent location for radioactive wastes.

3.2.12 *spent nuclear fuel (SNF)*, *n*—nuclear fuel that has been irradiated in a nuclear reactor and contains fission products, activation products, actinides, and un-reacted fissionable fuel. Normally spent fuel is contained in a metal cladding whose condition (undamaged, corroded, perforated, etc.) depends upon its original material properties as modified by the conditions during its service life including storage conditions.<u>unreacted fissionable fuel</u>.

3.2.13 *sludge*, *n*—*in nuclear waste management*, a slurry or sediment containing nuclear waste materials, a residue <u>materials</u>; a residue, generally radioactive, that has usually been formed from processing operations, corrosion or water basin storage.corrosion, or other similar reactions.

<u>3.2.14</u> *undamaged SNF, n*—SNF that can meet all fuel-specific and system-related functions. Undamaged fuel may be breached. Fuel assemblies classified as undamaged SNF may have assembly defects.

3.2.15 *waste package, n—in nuclear waste management*, the waste form and any containers, shielding, packing, and other absorbent-materials immediately surrounding an individual waste container. **10 CFR Part 60**

3.2.11.1 Discussion-

The waste package is expected to consist of an overpack (a container) into which commercial SNF, DOE SNF canisters and high level waste are to be placed for disposal at a repository.

3.2.16 water, *n*—in drying of spent nuclear fuel, it refers to the various forms of H_2O present in the fuel storage container. It is the total amount of moisture (specified by weight, volume, or number of moles) present (inin a container)container as a combination of vapor, free or unbound liquid water, H_2O , physisorbed water, H_2O , chemisorbed water, H_2O , and ice. The following specific terms for water are used in this document:

3.2.16.1 bound water, n-bound water includes adsorbed surface layers of water, water and nearly all chemisorbed water.

3.2.16.2 chemisorbed water, *n*—water that is bound to other species by forces whose energy levels approximate those of a chemical bond.

3.2.16.3 *physisorbed water (adsorbed water), n*— water that is physically bound (as an adsorbate, by weak forces) to internal or external surfaces of solid material.

3.2.16.4 *trapped water*, *n*—unbound water that is physically trapped or contained by surrounding matrix, blocked vent pores, cavities, or by the nearby formations of solids that prevent or slow escape. Note: Traps may have varying degrees of reversibility and a trap may be for practical purposes irreversible.the escape of water from the waste package.

3.2.16.5 *unbound_inbound/free water, n*—water, in the solid, liquid, or vapor state, that is not physically or chemically bound to another species.

4. Significance and Use

4.1 This guide provides technical information for use by SNF owners to determine the forms of water usually associated with spent nuclear fuel due to corrosion damage of the fuel, cladding and storage materials during irradiation and in storage pools. Drying may be Drying of the SNF and fuel cavity of the SNF container and its internals is needed to prepare the SNF for sealed dry storage, transportation, and/oror permanent disposal at a repository. This guide provides information for:technical information for use in determining the forms of water that need to be considered when choosing a drying process. This guide provides information to aid in (*a*) selecting a drying system, (*b*) selecting a drying method, and (*c*) demonstrating that adequate dryness was achieved.

4.1.1 Evaluating what drying system should be used,

4.1.2 Drying methods, and

4.1.3 Methods to confirm that adequate dryness was achieved.

4.2 The guide can be used to determine: considerations affecting drying processes include:

4.2.1 Drying technologies that are designed to remove most of the unbound water but will not remove all forms of water. Water remaining on and in commercial commercial, research, and researchproduction reactor spent nuclear fuels coming from water basin after removal from wet storage may become an issue when the fuel is sealed in a dry storage system or transport cask. The

movement to a dry storage environment typically results in an increase in fuel temperature due to the decay heat. This temperature change could be significant temperature, which may be sufficient to cause the release of water remaining from the fuel. The water release coupled with the temperature increase in a sealed dry package that container may result in container pressurization, fuel retrievability issues, corrosion of fuel or assembly structures, or both, that could affect retrieval of the fuel, and container corrosion.

4.2.2 Removal of the water associated with the SNF may be accomplished by a variety of technologies including heating, imposing a vacuum over the system, flushing the system with dry gases, and combinations of these and other similar processes.

4.2.3 Water removal processes are time, temperature, and pressure-dependent. Residual water in some form(s) should be anticipated.

4.2.4 Drying processes may not readily remove the water that was retained in porous materials, capillaries, sludge, CRUD, and as thin wetted surface films. Water trapped within damaged SNF may be especially difficult to remove.

4.2.5 A methodology for evaluating drying processes that may not readily remove all forms of water that may be retained in pores in fuel cladding, capillaries, sludge, erud, and thin wetted surface films. Drying techniques are Drying processes may be even less successful in removing bound water. Removal water from the SNF and associated materials because removal of bound water will only occur when the specific-threshold energy is applied required to break the bonds involved and release the water.specific water-material bonds is applied to the system. For spent nuclear fuel this threshold energy may come from the combination of thermal input and ionizing radiation. from decay heat and forced gas flow and from the ionizing radiation itself.

4.2.6 The adequacy of a drying procedure may be evaluated by measuring the response of the system after the drying operation is completed. For example, if a vacuum drying technology is used for water removal, a specific vacuum could be applied to the system, the vacuum pumps turned off, and the time dependence of pressure rebound measured. The rebound response could then be associated with the residual water, especially unbound water, in the system.

4.2.7 <u>How the residual water retained Residual water associated</u> with the SNF, CRUD, and sludge inside a sealed package may become available to react with the internal environment, the fuel, and the package materials as a result of extended time at equilibrium dry storage temperatures, or as the direct result of radiolytic decomposition.under dry storage conditions.

4.2.8 Thermal gradients within the container evolve with time, and as a result water vapor will tend to migrate to the cooler portions of the package. Water may condense in these areas. Condensed water will tend to migrate to the physically lower positions under gravity such as the container bottom.

4.2.9 Radiolytic decomposition of hydrated and other water-containing compounds may release moisture, oxygen and hydrogen to the container.

4.2.10 Extended time at temperature, coupled with the presence of ionizing radiation, may provide the energy necessary to release bound or trapped water to the container.

5. Evaluating the Drying Approach

5.1 Some forms of fuel degradation—such as cladding pinholes or cracks—may form before or during the dry storage period without violating design or licensing requirements. However, damage such as small cladding cracks or pinholes formed during the dry storage period could cause the fuel to be re-classified as failed fuel for repository disposal considerations. The objective in drying commercial LWR SNF containers is to eliminate enough water to preclude "gross" damage to commercial fuel or its cladding during dry storage and transport. The drying process itself must not damage the fuel. For example, the thermal cycling The proper approach to drying SNF is fuel and system-specific, and thus will depend on fuel type, fuel condition, fuel basket design, and associated materials (such as the neutron absorber in the basket). There is no single correct or even preferred approach. Intact commercial fuel may be dried by one approach. SNF with breached fuel rods by another approach, and research and production reactor fuels by yet another approach. Furthermore, the variables that must be considered in selecting a drying approach for one fuel type may differ significantly from those that are important for another fuel type. For example, hydrogen/hydride behavior should be considered in fuel systems clad with zirconium-based alloys but is not important to aluminum or stainless steel clad SNF. The proper drying approach will minimize the potential for damage of the fuel during the drying process in the zircaloy eladding-provides additional information regarding vacuum drying.

5.1.1 Some forms of fuel degradation, such as cladding pinholes or cracks, may form before or during the dry storage period without violating design or licensing requirements. However, damage such as small cladding cracks or pinholes formed during the dry storage period could cause the fuel to be reclassified as failed fuel for disposal. Fuel is classified at the time of loading, so the drying process should be chosen to balance the risks caused by the presence of water in the container and the risks incurred by removing the water.

5.2 Thermal cycling during drying of commercial light water reactor SNF may affect the hydride morphology in the cladding. Heating the SNF during a drying operation may dissolve precipitated hydrides, and subsequent cooling may result in hydride reprecipitation. The hydride orientation and therefore the properties of the fuel cladding may be affected by the dissolution-reprecipitation process.

5.3 DOE research and production reactor Research reactor and other non-commercial SNF that is not treated or reprocessed, will eventually reprocessed may be stored in sealed canisters within dry storage systems that may or may not be regulated by the NRC.



DOE-<u>regulated dry storage systems. Such dry storage canisters are may be expected to contain the SNF through interim storage, transport, and repository packaging. The objectives of drying processes used on this fuel are <u>virtually identical to the drying</u> <u>objectives for commercial fuels and are to:</u></u>

5.3.1 Preclude geometric reconfiguration of the packaged fuel,

5.3.2 Prevent internal components damage to the canister from over-pressurization or corrosion, and,

5.3.3 Minimize hydrogen generation or materialsand material corrosion that could be a problem during transport present problems during storage, transport, or repository handling operations.

5.4 The decision to select selection of the drying methodology for treating fuel for interim dry storage facility or disposition in a geologic repository could be based on the following factors: will involve many factors including the following:

5.4.1 Irradiation and storage history (for example, the decay heat output and the amount of hydrogen in the cladding),

5.4.2 Nature and degree of fuel damage including its storage history, (for example, quantity of breached rods or rods containing water),

5.4.3 FormForms of water in the packaged SNF container, container (for example, absorbed water)

5.3.3 Thermal and radiological environments involved,

5.4.4 Degree of self heating contribution to to which self-heating may contribute to the drying process,

5.4.5 Potential for Impact of residual water on corrosion and radiolytic degradation of the fuel and container material, material during drying, storage, and disposal,

5.4.6 Mechanisms of water interaction with the fuel and container components, and

5.3.7 Interactions that need to be considered to set the bounds for residual water, and

5.4.7 Maximum allowable water based on itemsamount of water remaining in the 5.3.5 throughcontainer after 5.3.7. drying is completed.

5.5 Categorization of SNF and Defects for Drying Evaluation—Water in SNF storage and transport containers can be a potential eause, result, or both, of fuel cladding damage or "failed fuel." However, there is not a single, universal definition of failure. The NRC ISG-1 Rev 2 defines damaged fuel as "spent nuclear fuel with known or suspected cladding defects greater than a hairline erack or a pinhole leak that have potential for release of significant amount of fuel particles." For the purposes of SNF transport per 10 CFR 71.55, fuel is essentially regarded as "failed" only when the geometric form of the fuel has been "substantially altered." For the purposes of dry cask storage per 10 CFR 72.22, the SNF eladding is required to: "be protected against degradation and gross rupture." Gross rupture is defined as that which could result in the release of significant quantities of fuel materials and fission products to the storage environment. For the purposes of receipt of SNF containers at a geologic repository, 10 CFR 961 defines three categories of commercial LWR "failed fuel:" This standard guide pertains to fuels that contain any cladding breach, such as hairline cracks or holes that may permit water ingress into a fuel element or the release of fuel materials from the fuel element. Such breaches may cause the fuel to be categorized as failed or damaged. Failed fuel may also be categorized as in 10 CFR 961, which separates failed commercial light water reactor fuel into three classes:

Class F-1 failed fuel has visually observable failure or damage

Class F-2 failed fuel has "radioactive leakage"

Class F-3 failed fuel is badly damaged and requires "encapsulation"

Each of these damaged or failed fuels could potentially require different handling/treatment than Drying treatments may depend on the fuel condition. Each category of damaged or breached fuel rods could require handling/treatment operations that differ from those used for non-failedintact fuel. It is, therefore, important when addressing the potential for fuel damage or failure due to moisture in the SNF containers to is important when considering a drying process to be clear about the kind and extent of cladding damage. It is particularly important to note that SNF could be regarded as intact or non-failed class of breached fuel rods in the fuels being dried. A given fuel element could be regraded as intact for the purposes of storage or transport performance but "failed" as having breached fuel rods for the purposes of geologic repository disposal. This potential exists because of the relatively predictable-The classifications differ because the performance of damaged fuel for a dry storage term versus the very difficult prediction of performance in geologic time. Commercial is typically more predictable over dry storage periods than over geologic time. SNF may be characterized as intactdamaged or undamaged or failed through the use of one or more of the following processes: review of operating records (core history and handling), visual examinations, ultrasonic testing, wet or dry sipping, and eddy current testing. Fuel type and the presence/type of defeets involved are essential starting points for analysis of drying. In order to organize the information in this guide, the varieties of spent fuels in existence have been placed into categories with a letter designation for consideration. Similarly, the types of defeets to be considered are identified with a number designation. The identities of these categories are provided below.

Fuel type and the presence and type of defects are essential inputs for evaluating drying operations. In order to organize the information in this guide, selected varieties of spent fuels have been placed into categories with a letter designation for consideration. Similarly, the types of defects to be considered are identified with a number designation. The identities of these categories are provided below.

5.5.1 SNF Grouping—Group—The following groupings for SNF groups are used in this guide to distinguish between the specific SNF types:



A	Commercial PWR/BWR, U oxide or mixed oxide fuel in Zr-alloy cladding
A	Commercial oxide fuel in Zr-alloy cladding
B	Mixed carbide fuel with SiC cladding (in graphite)
В	Carbide or oxide particle fuel with SiC coating (in graphite)
B G	U oxide or mixed oxide fuel in stainless steel cladding
С	Oxide fuel in stainless steel cladding
D	U-ZrH fuel in Zr-alloy or stainless steel cladding
E	U oxide or mixed oxide fuel in Al-alloy cladding
E	Oxide fuel in Al-alloy cladding
F	UAI _x fuel or U-ZrH fuel in Al-alloy cladding
G	U metal and U alloy fuel in Zr-alloy, Al-alloy or stainless steel cladding
G	Metal fuel in Zr-alloy, Al-alloy or stainless steel cladding
H	U carbide fuel with pyrolytic carbon cladding
Н	Carbide or oxide particle fuel with only pyrolytic carbon coating
Ŧ	Unclad U metal or oxide fuel
<u>l</u>	Unclad metal or oxide fuel

5.5.2 *Fuel Conditions*—The following fuel conditions will be used in this document to categorize the needed level of drying required. Fuel Conditions are designated:

1	Fully intact cladding with no known penetration	ons
2	Cladding with known small pinholes or crack	ed cladding that may allow
	entry/exit of moisture (Not considered "failed"	-fuel per 10 CFR 71/72)
	(Class F-1 equivalent)	• • •
2	Cladding with small pinholes or hairline crack	s that may allow entry/exit
_	of moisture	
3	Fuel with visible or suspected sludge, CRUD	or coatings that may hold
	significant water	C
3	Fuel with visible or suspected sludge or coat	ngs that may hold
-	significant water	
4	Cladding with known large cracks or holes th	at will allow free entry/exit of
	moisture (Considered "failed" fuel) (Class F-2	or F-3 equivalent)
4	Cladding with known large cracks or holes the	at will allow free entry/exit of
-	moisture	
5	Fuel with major cladding damage from previo	us fuel oxidation, exposure
	to water, or both (Damaged from previous er	try/exit of moisture)
5	Fuel with major cladding damage from previo	us fuel oxidation, exposure
-	to water, or both	
6	Rubblized fuel that has little or no intact clad	ling, high surface area, and
	previous exposure to water (Includes fuel pin	s or elements that have
	been sectioned for analysis or study) (Class	F-3 equivalent)
6	Rubblized fuel that has little or no intact clad	ling, high surface area, and
_	previous exposure to water (Includes fuel roo	s or elements that have
	been sectioned for analysis or study)	

5.6 Forms of Residual Water in SNF Containers—After drying, residual water in a variety of forms may remain on the fuel, fuel cladding, or internal components of the container. These forms include unbound liquid water, ice formed during drying, physisorbed water, and chemisorbed water.

<u>5.6.1 Unbound Water</u>—Unbound water may be present in containers of SNF transferred from a water storage pool. Water retention depends on the condition of the fuel, the container design, and the drying process. Sources of unbound water after vacuum drying may include trapped water and water in capillaries.

5.6.2 *Ice*—Ice formation can be a cause for water retention in SNF containers that have undergone vacuum drying. In vacuum drying the gas pressure is reduced below the vapor pressure of the water to evaporate the liquid phase. The ratio of the heat of vaporization of water (539.6 cal/g) to the specific heat (1 cal/g K) corresponds to a large temperature change; consequently, liquid water may undergo a considerable temperature drop during drying. Since the heat of fusion of water (79.7 cal/g) is relatively small, the energy removed from the liquid by evaporation can cause the remaining water to freeze. Measures may be necessary to prevent the water from freezing in the container or in the vacuum lines. Drying procedures with thermal homogenization steps such as a helium backfill or use of other hot inert gases usually prevent ice formation. It is also important to route vacuum lines to avoid low spots. Throttling of vacuum pumps to slow the rate of vacuum drying may also prevent ice formation. (See Annex A2.)

5.6.3 *Physisorbed Water*—Physisorbed water is found on all external surfaces of the SNF (for example, cladding and assembly hardware) and the container internals (for example container walls, baskets, etc.). Typical water concentrations are about 0.03 to 0.05 g/m² per monolayer. The binding force holding the water to the surface is weak and the water layer can be removed at relatively low temperatures (<50°C) by vacuum drying or forced gas dehydration. Cracks, open pores, and corrosion products may hinder evaporation and hold physisorbed water by virtue of additional surfaces. Corrosion products on exposed U plate fuel are notable for retaining physisorbed water.

5.6.4 Chemisorbed Water—Chemisorbed water may exist in a hydroxide or hydrate in the native oxides or corrosion products on the fuel, cladding, or container materials. The dehydration of hydroxides or hydrates occurs by the reformation of water molecules, which are released when the thermal energy or energy from ionizing radiation equals or exceeds the bonding energy of the hydrated compound. A number of uranium oxide hydrates may be formed as a result of uranium or uranium oxide contact with water. Chemisorbed water may also be found in cladding and container materials. Aluminum metal in water forms a number

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of surface hydroxides such as Al (OH)₃ (or Al₂O₃·3H₂O) which begin dehydrating near 100°C to the form AlO(OH) (or Al₂O₃·H₂O) which is stable to >340°C. Zirconium cladding may also form the hydrated oxides $ZrO(OH)_2$ or $Zr(OH)_4$ during irradiation. The water content of hydrated zirconium oxides is small, and the water will not be released below 500°C (2). See Annex A1 for other hydroxides and hydrates formed from water contact with typical fuel and container materials.

5.7 *Forms of Residual Water in SNF Containers*—Sources of Water: After drying, residual water in a variety of forms may remain on the fuel, fuel cladding, or internal components of the container. These forms and sources are discussed below.

5.7.1 *Forms of Water*—*General Service Environment for Water Reactor Fuel*—The forms of water include unbound water, ice formed during drying, physisorbed water, and chemisorbed water. Water surrounds most SNF assemblies until they are placed in a dry storage environment. The fuel is irradiated in water, stored in water pools, and transferred to dry storage containers while the fuel and the container are both under water. The water may cling to the surfaces it contacts, seep into cracks and crevices, and pool in low places in the storage container. Locations for water that should be considered include:

5.5.1.1 Unbound Water—Unbound water has to be considered as potentially present in SNF containers. In the case of SNF transferred from a water storage pool to a dry container system, unacceptable amounts of unbound water may remain in the eontainer if the drying procedure does not remove the water properly. Sources of unbound water after vacuum drying may include pooling, ice formation, capillaries, trapped water or thin wetted surface films.

(1) Regions beneath the assemblies,

(2) Dash pots in pressurized water reactor guide tubes,

(3) Water rods in boiling water reactor fuel, and

(4) Crevices in grid spacers, baskets, and assemblies.

(5) Neutron absorber.

5.5.1.2 *Ice Formation*—Ice formation can be a root cause for residual water in SNF containers that have undergone vacuum drying. In vacuum drying the gas pressure is reduced below the vapor pressure of the water to evaporate the liquid phase. The heat of vaporization of water (539.6 cal/g) is considerable higher than its specific heat (1 cal/g/°C); consequently, liquid water may undergo a considerable temperature drop during drying. Since the melting heat of ice (79.7 cal/g) is relatively small, the energy removed from the liquid by evaporation can cause the water to freeze. Drying operation design steps may be necessary to prevent the water from freezing in the container or in the vacuum lines. Drying procedures with thermal homogenization steps such as He-backfill or use of other hot inert gases usually prevent ice formation. It is also important to route vacuum lines to avoid low spots. Throttling of vacuum pumps to slow the rate of vacuum drying also prevent ice formation. (See Annex A2.)

Additionally, potential impacts of the drying operation itself should be considered. For example, drying operations could cause blistering and delamination in the neutron absorber if water is trapped in the structures.

5.5.1.3 *Physisorbed Water*—Physisorbed water is found on all surfaces of the SNF and the container. Typical water concentrations are about 0.03 to 0.05 g/m²/monolayer. The binding force is weak and the water layer can be removed at relatively low temperatures with an adequate vacuum drying technique. Cracks and open pores in the surface may act as capillaries that may hinder the evaporation of the water. SNF cladding with cracks, surface corrosion, porosity, or oxide spallation may hold significant physisorbed water. Corrosion products on exposed fuel meat may also retain physisorbed water, especially corrosion products from metallic U fuels.

5.5.1.4 *Chemisorbed Water*—Chemisorbed water may exist as a hydroxide or hydrate in the native oxides or corrosion products on the fuel, eladding, or container materials. The dehydration of hydroxides occurs via the reformation of water molecules, which are released from the lattice at temperatures and/or ionizing radiation corresponding to the specific bonding energy of the compound. A number of uranium oxide hydrates may be formed as a result of uranium or uranium oxide contact with water. These chemical species are discussed in Annex A1. Chemisorbed water may also be found in cladding materials and the materials of eontainer construction. Aluminum metal in water forms a number of surface hydroxides of the form A1 (OH)₃ (or Al₂O₃·3H₂O) which begin dehydrating near 100°C to the form AlO(OH) (or Al₂O₃·H₂O) which is stable to >340°C. Zirconium cladding may form the hydrated oxides $ZrO(OH)_2$ or $Zr(OH)_4$ during irradiation. The potential water content is small and will not be released below 500°C (1).⁵ (See Annex A1 for other hydroxides and hydrates formed from water contact with typical fuel and container materials.)

5.7.2 Sources of Water: CRUD and Sludge:

5.7.2.1 *CRUD on Commercial SNF*—CRUD deposits on commercial SNF may include corrosion products from reactor coolant system materials or other materials/chemicals from the system inventory. The amount and type of the deposits are dependent on the reactor type, operating fuel duty, and water chemistry. Characteristic CRUD area density for pressurized water reactor fuel is $<5 \text{ mg/cm}^2$ with an inhomogeneous distribution over the fuel surface, typically deposited on the upper/hotter portions of the fuel rod as a layer averaging less than 25 µm (0.001 in.) but potentially reaching 100 µm (0.004 in.) in thickness (3). CRUD deposits on boiling water reactor fuel average 25 to 76 µm (0.001 to 0.003 in.) in thickness and may reach a thickness of 250 µm (0.010 in.) (3). Depending on CRUD type and fuel pool chemistry, CRUD levels may be reduced during pool storage. The contribution of CRUD to the water content on the surface of commercial SNF is typically small.

5.7.2.2 CRUD and Sludge: Sludge in SNF Operations-

(1) CRUD on Commercial SNF—Surface CRUD deposits on commercial SNF are corrosion products from reactor coolant system materials or other chemicals from within the system inventory. The amount and type of the deposits are dependent on the



reactor type and its water chemistry. Characteristic CRUD areal density for PWR fuel is $<5 \text{ mg/cm}^2$ with an inhomogeneous distribution over the fuel surface, typically deposited on the bottom half of the fuel rods as a layer averaging less than 25 µm (<0.001 in.) but potentially reaching 100-µm (0.004-in.) thick (2). CRUD deposits on BWR fuel average 25 to 76 µm (0.001 to 0.003 in.) and may reach 250 µm (0.010 in.) maximum thickness (2). The contribution to the source of water from CRUD on the surface of commercial SNF appears to be small.

(2) Sludge in SNF Operations—Sludge may accumulate in SNF wet storage systems from two primary sources: (1) water corrosion of the SNF and other materials in the storage pool, (2) dirt and dust entering from loading doors, HVAC systems, etc. Both sources of sludge are similar in that they may hold significant quantities of water and could get transferred with the fuel into dry storage containers unless appropriate cleaning operations are employed. For groups A-D fuels in defect conditions 1 and 2 from well-controlled wet storage systems, sludge is probably not a concern. For fuel group A-D with defect conditions 3-6 and groups E-I fuels, especially with defect conditions 3-6, the quantity and characteristics of the sludge carryover and its impact on drying needs to be evaluated. Analyses of sludge accumulated from wet storage of damaged DOE metallic uranium fuels (3) showed that a variety of aluminum, iron, and uranium hydrous oxides made up over 90 % of the dry weight of the sludge.Sludge may accumulate in SNF water storage systems from two primary sources: (1) corrosion of the SNF and other materials in the storage pool, (2) dirt and dust entering from loading doors, ventilating systems, etc. Both sources of sludge are similar in that they may hold significant quantities of water and could get transferred with the fuel into dry storage containers unless the fuel is appropriately cleaned. Sludge is probably not a concern for fuels in SNF Groups A-D that have Fuel Conditions 1 or 2 and are from well-controlled wet storage systems. However, the quantity and characteristics of the sludge carryover and its impact on drying should be evaluated for fuels in SNF Groups A-D that have Fuel Conditions 3-6 and for SNF Groups E-I, especially those with Fuel Conditions 3–6. Analyses of sludge accumulated from wet storage of damaged metallic uranium fuels (4) showed that a variety of aluminum, iron, and uranium hydrous oxides made up over 90 % of the dry weight of the sludge.

5.7.3 Water in SNF Packages: Associated with Specific Fuel Types:

5.7.3.1 Commercial SNF, <u>SNF Group A</u>, <u>Defect Conditions 1–4–Fuel Conditions 1–4–EWR-Light water reactor</u> fuel without any through-cladding defects (<u>Fuel Condition 1</u>) will not allow water inside fuel rods. <u>Even However, even very small pinholes</u> or eracks, however, cracks (Fuel Condition 2) may result in water penetrating the cladding during reactor operations and pool storage, and being held in the fuel to cladding fuel-to-cladding gap and the rod plenum after drying. <u>PWR fuel may also trapSimilar</u> water retention may also occur for Fuel Condition 4. Pressurized water reactor fuel in Fuel Condition 3 may also retain water in guide tubes if (a) the dashpot drain hole is blocked or partially blocked with sludge or <u>CRUD</u>. <u>CRUD</u>, (b) if the discharge point is elevated above the tube bottom or (cA typical LWR rod defect) in some designs if there are spaces such as in the tube-in-tube design. Adequate removal of the residual water will depend primarily on the temperature–pressure conditions at the specific location within the fuel assembly. For example, the water associated with the thimble tube dashpots is at the bottom of the assembly which in most drying scenarios is the cooler region during the drying process. A typical light water reactor rod in Fuel Condition <u>4</u> is characterized by a combination of primary and secondary defects. Whereas the <u>The</u> primary defect is regarded to be the location of the original penetration, the original penetration, and secondary defects may be located <u>at</u> some distance from the primary defect andit. The secondary defects are normally attributed to local hydride blistering (45). The defects may retard water removal.

5.7.3.2 Clad Metallic U fuels (Groups Fuels (SNF Groups F and G), Defect Conditions 1 - 4—Fuel Conditions 1 - 4—Clad metallic U and most U alloyallow fuels will not allow water inside intact eladding. Even-(Fuel Condition 1) cladding. Vacuum drying of such fuels has been performed for intact Zircaloy-clad fuels from Hanford K-basin (6). Drying tests on unirradiated mock-ups have been performed to demonstrate drying capability for Magnox elements from Sella field water basins as a contingency for dry storage (7). However, water ingress through even the smallest pinholes with water egress, however, (Fuel Condition 2 or 4) may have a noticeable effect in metallic U. Water, even on metallic U fuel. Even at basin temperatures, will-water may oxidize U metal sufficiently to rupture or "unzip" fuel cladding (58). If the oxidation processes cause the internal environment becomes to become sufficiently anoxic, hydrogen will be produced, and the U metal will start forming react to form UH₃. Free reactive Exposed surfaces of UH₃ may impact safety considerations for drying operations in the presence of react vigorously with residual moisture or air (69).

5.7.3.3 *Mixed Carbide Fuels, <u>SNF</u> Groups B and H*—Mixed carbide fuels encapsulated in pyrolytic carbon, graphite, or both, are designed for gas-cooled reactors and <u>areshould</u> not designed to be exposed to water. If such fuels become soaked with water for any reason (dry storage mishaps, incursion of water into dry wells, etc.), drying may be quite difficult due to potential adsorption <u>absorption</u> of water in the pores of the graphite or carbon. <u>An aqueous solution can penetrate the graphite matrix of an</u> HTGR fuel element through its open pore system, and under normal conditions a spherical element takes up about 8 mL of solution (10).

5.7.3.4 Miscellaneous Research Fuels, and Production Reactor Fuels, SNF Groups C, D, and E—A wide variety of research reactor fuels have been irradiated. These fuels will respond to water according to their fuel meat, cladding composition, and The response of these fuels to water will depend on the fuel composition, cladding alloy, and cladding integrity. Research reactor fuels generally have relatively low burnup and low decay heat. The low decay heat low decay heat output, which may dictate the use of a specialized heated drying process. specialized heating processes to achieve adequate dryness. Dry storage temperatures and

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radiation levels may be so low that water radiolysis and secondary oxidation reactions may not occur or may be insignificant. <u>are</u> insignificant. However, many of the research and production reactor fuels have been damaged during storage and, therefore, may be difficult to dry. Each group of these should be evaluated separately because of the wide variations in type and condition.

5.8 SNF <u>Exposure</u> Environments—The dryness required for a given nuclear–fuel willis often relate directly related to the radiation, time, <u>duration of exposure and the radiation</u>, temperature, and water chemistry environment-to which it was exposed during reactor operation and pool-storage. Specific fuels typically have an environmental exposure history that provides input into probable drying requirements. The drying process mustshould reliably establish water vapor pressures levels such that the remaining water is insufficient to cause detrimental chemical reactions are limited for the proposed lifetime in <u>during dry</u> storage.

5.8.1 Commercial Reactor Fuels:

5.8.1.1 Commercial nuclear fuel is irradiated in a water environment at elevated temperature and pressure. If a breach of the cladding occurs develops while fuel is in-core, the internal gas will be released and the pressurized water may enter into the fuel rod. Upon removal from the core, reactor, the fuel is stored in a water basin with the water temperature typically less than 40°C and water pressure equivalent only to that resulting from its depth 40°C. The water pressure acting on the fuel depends on the depth of the fuel in the basin water. Both the reactor core water and basin water-typically have tightly controlled chemistry and purity water chemistries that may prevent fuel or at least minimize fuel cladding damage.

5.8.1.2 The decay heat generated by the SNF during storage in water basin drops off predictably as the fission products from irradiation decay away. decay. After a suitable cooling time (aged for at least 1 year) that is dependent on the fuel burnup, decay heat output, system design, and applicable regulations (711)), the SNF may be moved out of basin storage (wet storage) and placed into a dry storage cask system.

5.8.1.3 The expected thermal performance of an SNF a cask or package can be modeled to determine the expected dry storage temperature profile of the system as a function of time (812). Design or regulatory requirements may establish short term short-term temperature limits for SNF cladding. The limits to ensure cladding integrity are in part a function of maintaining cladding integrity impacted by creep or by embrittlement, for example. The limits may depend on burnup, cladding design and fuel pressurization. Limits from 400°C250 (9)to 570°C (1013) have been suggested. The evaluation of the limits should consider how cladding integrity is affected by hydride dissolution, reprecipitation, and reorientation, creep, delayed, hydride cracking, and thermal annealing of radiation damage. The impact of the hydrogen concentration and morphology on the cladding properties, such as the ductility transition temperature, will affect the temperature limits.

5.8.2 Research/DOE Research and Non-commercial Reactor Fuels:

5.8.2.1 Research reactor fuel and most other DOE fuels are also irradiated in water. The reactors have irradiation temperatures and pressures tend to that vary widely, widely but they are typically lower than those of a commercial power plant. Fuel lifetimes are also quite variable in DOE production and test reactors. Research reactors may operate with little or no change in fuels for many years, and the fuel may be exposed to stagnant water or a humid air environment between operating cycles. DOE production Production reactors may provide the opposite extreme as the fuel is changed out on a schedule refueling scheduled to provide the optimum isotope production desired, with a abundances, and the total fuel irradiation lifetime time may be less than a year.

5.8.2.2 SNF storage for DOE fuels varies somewhat on the basis of Conditions necessary for successful dry storage of research reactor SNF will depend on the total irradiation, fuel type, and decay heat. heat output. The elimination of reprocessing in the U.S., however, U.S. essentially resulted in a basic trend toward placing all types of DOE placing the vast majority of research and production reactor SNF into extended basin storage and a few dry storage systems. The primary considerations involved with movement of largely metallic SNF and research/DOE SNF into dry storage are these fuels into interim dry storage include the lack of significant latent decay heat, the wide range of fuel cladding materials, and the eladding integrity involved.lack of cladding integrity in many fuels. One possible approach to determining the necessary dry storage conditions may include demonstrating that, because of prior damage to the fuel, any anticipated in-storage degradation would not compromise subsequent disposition options.

5.8.2.3 A wide variety of environments are possible in dry storage of DOE SNF due to the types of dry storage employed. Three Two primary types of dry storage systems are currently in use: Underground well storage, active vented storage, and passive use for research reactor SNF: Underground well storage and vented storage. Underground well storage and interior facility storage typically operate at temperatures between ambient and 60°C, and the SNF is typically not sealed in a small confined space because containmentcontainer because confinement is provided by the well or the facility itself. Exterior cask storage systems may be very similar to those used for commercial SNF except thateven though the latent decay heat is typically-insufficient to heat the cask above exterior ambient conditions.significantly. The experience and expertise gained in operating the current dry storage systems for production reactor SNF should be carefully considered if the research reactor SNF is to be transferred to alternative dry storage systems for storage or disposition. The Irradiated Fuel Storage Facility at the Idaho Nuclear Technology and Engineering Center uses a forced ventilation system with high-efficiency particulate air filtration for dry storage of research reactor fuel in unsealed canisters (14).

5.8.2.4 Vented well-type and facility storage systems have the advantage that residual water in and around the SNFResidual water in vented dry storage systems can evaporate or radiolyze over long times and times, so water can escape from the system. However, this situation may also work in reverse because canisters containing cool fuels may also aspirate water from the external atmosphere as a function of external temperature and dew point. Such water ingress atmosphere. Water evaporation and aspiration during "dry" storage may significantly increase change the overall chemisorbed water content of the SNF over the storage period,

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<u>SNF</u>, especially if the <u>SNF it</u> is badly damaged. <u>Characterization of SNF behavior in such vented systems may provide insight into</u> the probable behavior of SNF in alternative dry storage systems.

5.9 Potential Effects of Residual Water on SNF and Containers as a Function of Environment—Containers—The presence of water in storage containers for dry storage of Residual water in SNF can be released by direct the container environment by direct, thermally induced decomposition of the chemically bonded species, vaporization of the physisorbed and the free water, and radiolytic decomposition. These forms of The released water and decomposition products eouldmay cause corrosion, pressurization and possibly embrittlement issues for the storage of spent fuel.pressurization, and possibly embrittlement, although such degradation is not generally anticipated (15).

5.9.1 Radiolysis:

5.9.1.1 Radiolysis occurs as a result of gamma, beta, neutron, or alpha particle interaction with residual water or oxyhydroxides. Radiolysis within a sealed spent fuel package releases free oxygen and hydrogen which may promote corrosion or produce a flammable atmosphere (16, 17). These radiolysis products, in the absence of residual air, recombine and persist at very low concentrations; however, as little as 0.1 % residual air can impede recombination and increase the concentration of radiolysis products. The specific concentration of radiolysis products depends temperature, time, and the amounts of residual air and water. One calculation for an SNF container with one litre of water (17) showed that the concentration of hydrogen remained well below the flammability limit for hydrogen/air mixtures after 300 years of storage.

5.9.1.2 The radiolysis of residual water within a sealed spent fuel package releases free oxygen and hydrogen for internal corrosion reactions. Radiolysis occurs as a result of gamma, beta, neutron or alpha particle interaction with the water molecule. Neutron radiolysis is a major factor important during reactor operation but diminishes rapidly after fuel removal from the active core, and is insignificant by the end of the pool storage term.pool (basin or wet) storage.

5.9.1.3 Gamma radiolysis that may act on interactions with water and hydroxyl groups that are external and internal to the fuel is an issue for may affect both the fuel itself and other hydrated compounds within the radiation field (inside the eask). inside the cask. Gamma radiolysis of hydrated uranium oxides will occur in fields of 100 000 R/h1000 Gy/h or(17). greater. Such radiolysis has resulted in HHydrogen production from dry (no free of₂ pressure increases of up to 10 psi in an enclosed system physisorbed water) oxyhydroxides of aluminum has been reported (11, 1218). Gamma activity in SNF also decreases relatively rapidly as a function of time; therefore, the potential for gamma radiolysis will decrease during the fuel storage term, and should become inconsequential after about 15 years.decreases over time, and the levels of hydrogen, oxygen, and nitric acid developed during storage are generally considered inconsequential even after 300 years (17).

5.9.1.4 Beta particles radiolysis of water occurs only in close proximity to the fuel surfaces and hydrated species. In the case where decay event because of the limited travel of the beta particle. However, if hydrated corrosion products are fairly distributed in sludges, uniformly distributed in sludges or if sludges are in contact with fuel surfaces, the contribution by the beta emitting isotopes to water radiolysis could be significant.

5.9.1.5 Alpha particles have a low linear-energy-transfer rate but alpha emitters are long lived and represent the longest term issue for water radiolysis. Alpha radiolysis occurs only when the alpha emitter is in direct contact with the hydrated species. Therefore, alpha radiolysis is generally limited to hydrated fuel compounds or fuel-bearing sludges within the container. The actual rates for alpha radiolysis are not well definedknown and moreadditional work is needed on the issue (1319).

5.9.2 Hydrogen—Fuel, Cladding, and Packaging Reactions:

5.9.2.1 Radiolytie Hydrogen is generated by the radiolytic decomposition of water and by most metal corrosion reactions with water generate hydrogen. reactions. In order to ensure that a-flammable environmentgas cannot be present in the event of a container rupture accident, a welded canister needs to be opened, the hydrogen content in SNF containers is usually limited to below 4 %, the lower flammability limit for hydrogen in air (1420). The hydrogen generation When the hydrogen-containing volume is otherwise inert, flammability will not be an issue, but hydrogen may cause embrittlement, and a flammable mixture may form when the container is breached or opened. This is especially true for non-intact fuel assemblies where, for example, the French Competent Authority has requested measurement of the hydrogen generation rate before transport of packages transporting at least one breached fuel rod (20). Hydrogen generation rates can be predicted by performing a material and energy balance using with reasonable accuracy from the temperature, radiation levels, materials, and water contained in the package types of materials present, and water content (1517, 21).

5.9.2.2 Hydrogen may be a problem for SNF container materials over long storage times. times, although analysis has shown (17) that after 300 years of storage in a container with one litre of residual water, the hydrogen concentration reaches only 2.3 %. Hydrogen tends to collect in steels at locations of high stress and surface discontinuity. Hydrogen may also be absorbed into Zircaloy cladding making it more susceptible to fracture. discontinuity, and it may embrittle certain steels, especially high strength ferritic and martensitic steels. The effects of hydrogen in steels are fairly well understoodestablished, and numerous ASTM test methods are available for evaluating thesehydrogen effects in materials (1622). Hydrogen may also be absorbed by zirconium-alloy cladding and make it more susceptible to fracture. In general, these effects increase with increases in the hydrogen fugacity, which is generally greater in at a surface during corrosion by aqueous environments than during exposure in gaseous atmospheres (1723). Hydrogen entry into fuel or container materials may also be driven by galvanically coupled dissimilar metals if an electrolyte is present. Low-pressure gaseous hydrogen acting on materials for very long times may involve different mechanisms than those