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# Standard Guide for Drying Behavior of Spent Nuclear Fuel<sup>1</sup>

This standard is issued under the fixed designation C1553; the number immediately following the designation indicates the year of original adoption or, in the case of revision, the year of last revision. A number in parentheses indicates the year of last reapproval. A superscript epsilon ( $\varepsilon$ ) indicates an editorial change since the last revision or reapproval.

## 1. Scope

- 1.1 This guide discusses three steps in preparing spent nuclear fuel (SNF) for placement in a sealed dry storage system: (1) evaluating the needs for drying the SNF after removal from a water storage pool and prior to placement in dry storage, (2) drying the SNF, and (3) demonstrating that adequate dryness has been achieved.
- 1.1.1 The scope of SNF includes nuclear fuel of any design (fuel core, clad materials, and geometric configuration) discharged from power reactors and research reactors and its condition as impacted by reactor operation, handling, and water storage.
- 1.1.2 The guide addresses drying methods and their limitations 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, the container, or both after the drying process has been completed. It also discusses the important and potential effects 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.
- 1.1.3 The basic issues in drying are: (1) to determine how dry the SNF must be in order to prevent problems with fuel retrievability, container pressurization, or container corrosion during storage, handling, and transfer, and (2) to demonstrate that adequate dryness has been achieved. Achieving adequate dryness may be straightforward for undamagedintact commercial fuel but complex for any SNF where eladding damage has occurred the cladding is breached prior to or during placement and storage at the spent fuel pools. Challenges in achieving adequate dryness may also result from the presence of sludge, CRUD, and any other hydrated compounds. These may be transferred with the SNF to the storage container and may hold water and resist drying.
- 1.1.4 Units are given in both SI and non-SI units as is industry standard. In some cases, mathematical equivalents are given in parentheses.
- 1.2 This standard only discusses SNF drying and does not purport to address all of the handling and safety concerns, if any, associated with the drying process(es). its use. It is the responsibility of the user of this standard to establish appropriate safety safety, health, and healthenvironmental practices and to meet regulatory requirements prior to and during use of the standard determine the applicability of regulatory limitations prior to use.
- 1.3 This international standard was developed in accordance with internationally recognized principles on standardization established in the Decision on Principles for the Development of International Standards, Guides and Recommendations issued by the World Trade Organization Technical Barriers to Trade (TBT) Committee.

<sup>&</sup>lt;sup>1</sup> 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. Referenced Documents

2.1 ASTM Standards:<sup>2</sup>

C859 Terminology Relating to Nuclear Materials

C1174 Guide for Evaluation of Long-Term Behavior of Materials 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:<sup>3</sup>

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

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

2.3 Government Documents: <sup>4</sup>The The 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

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

## 3. Terminology

- 3.1 Definitions—For definitions of terms used in this guide but not defined herein, refer to Terminology C859 or Practice C1174.
- 3.2 Definitions of Terms Specific to This Standard: Standard—Various terms are used internationally for the broad set of definitions related to failed fuel (ref. IAEA Nuclear Energy Series, No NF-T-3.6). In this drying guide, only two fuel conditions that can impact drying behavior are considered (1) intact or non-breached fuel; and (2) breached or failed fuel.—Refer to SFST-ISG-1 for additional definition details.
- 3.2.1 breached spent fuel rod, or failed fuel, n—spent fuel rod with cladding defects that permit the release of gas from the interior of the fuel rod. Arod; a breached spent fuel rod may also have be such that the cladding defects sufficiently largeare sufficient 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.particulate; water could enter a breached spent fuel rod of any severity, and thus may adversely impact the ability to dry the fuel to remove this water.
- 3.2.2 CRUD, n—in nuclear waste management, deposits on fuel surfaces from corrosion products that circulate in the reactor coolant. Compositions of the deposits reflect materials exposed to coolant and activation products formed during irradiation. Term was originally an acronym for "Chalk River Unidentified Deposits."

3.2.2.1 Discussion—

Compositions of the deposits reflect materials exposed to coolant and activation products formed during irradiation.

3.2.2.2 Discussion—

The term CRUD was originally an acronym for "Chalk River Unidentified Deposits."

3.2.3 damaged SNF, n—in nuclear waste management, any fuel rod of fuel assembly that cannot fulfill its fuel-specific or system-related functions.

<sup>&</sup>lt;sup>2</sup> 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.

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

<sup>&</sup>lt;sup>4</sup> 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/doe-collections/isg/spent-fuel.html:from U.S. Government Printing Office, Superintendent of Documents, 732 N. Capitol St., NW, Washington, DC 20401-0001, http://www.access.gpo.gov.

<sup>&</sup>lt;sup>5</sup>Unpublished SRS data on testing of austenitic stainless steel tritium shipping containers used intermittently for 15 years to hold tritium at 1 psig indicated that tritium did diffuse into the steel structure, but to a depth less than that required to cause fracture unless the material was highly stressed.

- 3.2.3 *disposal*, *n*—*in nuclear waste management*, the emplacement of radioactive materials and wastes in a geologic repository with the intent of leaving them there permanently.
- 3.2.4 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.
  - 3.2.4.1 Discussion—

Some fuel rod designs include an internal getter to remove residual hydrogen/moisture from the internal rod atmosphere.

- 3.2.6 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.
- 3.2.5 *independent spent fuel storage installation (ISFSI)*, *n*—a system designed and constructed for the interim storage of spent nuclear fuel and other radioactive materials associated with spent fuel storage.
- 3.2.6 *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.
- 3.2.7 packaging, or SNF storage container, n—in nuclear waste management, an assembly of components used to ensure compliance with the applicable requirements for independent storage of spent nuclear fuel and high-level radioactive waste or for transportation of radioactive materials.
- 3.2.10 pinhole leaks or hairline cracks, n—minor eladding defects that will not permit significant release of particulate matter 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.)
- 3.2.8 repository, geologic repository, n—in nuclear waste management, a disposal site, a permanent location for radioactive wastes.
- 3.2.9 spent nuclear fuel (SNF), n—nuclear fuel that has been irradiated in a nuclear reactor and contains fission products, activation products, activation
- 3.2.10 *sludge*, *n*—*in nuclear waste management*, a slurry or sediment containing nuclear waste materials; a residue, generally radioactive, that has usually been formed from processing operations, corrosion, or other similar reactions.
- 3.2.14 *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.
- 3.2.11 *waste package*, <u>container</u>, <u>n</u>—in <u>nuclear waste management</u>, the waste form and any containers, shielding, packing, and other materials immediately surrounding an individual waste container.
- 3.2.12 water, n—in drying of spent nuclear fuel, 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 in a container as a combination of vapor, free or unbound liquid  $H_2O$ , physisorbed  $H_2O$ , chemisorbed  $H_2O$ , and ice. The following specific terms for water are used in this document:guide:
  - 3.2.16.1 bound water, n—adsorbed surface layers of water and chemisorbed water.
- 3.2.12.1 *chemisorbed water, n*—water that is bound to other species by forces whose energy levels approximate those of a chemical bond.
- 3.2.12.2 *physisorbed water* (*adsorbed water*), *n*—water that is physically bound (as an adsorbate, <u>typically</u> by weak forces) to internal or external surfaces of solid <del>material.</del> material; the binding energy of the first monolayer of water on oxides (for example, ZrO<sub>2</sub>) is strong with reduced binding energy of successive monolayers.
- 3.2.12.3 *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 the escape of water from the waste package.



3.2.12.4 *unbound/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 Drying of the SNF and fuel cavity of the SNF container and its internals is needed to prepare for sealed dry storage, transportation, or permanent disposal at a repository. This guide provides 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 (see Annex A2).
- 4.2 The considerations affecting drying processes include:
- 4.2.1 Water remaining on and in commercial, research, and production reactor spent nuclear fuels 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, which may be sufficient to cause the release of water from the fuel. The water release coupled with the temperature increase in a sealed container may result in container pressurization, 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, physical features that retain water and as thin wetted surface films. Water trapped within damaged breached SNF may be especially difficult to remove.
- 4.2.5 Drying processes may be even less successful in removing bound water from the SNF and associated materials because removal of bound water will only occur when the threshold energy required to break the specific water-material bonds is applied to the system. For spent nuclear fuel this threshold energy may come from the combination of thermal input from decay heat and forced gas flow and heat, externally applied heat, or 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 Residual water associated 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 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 Considerations in Drying Approach

5.1 The properAn effective 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/hydridehydride behavior should be considered in fuel systems clad with zirconium-based alloys but is not important to aluminum or stainless steel clad SNF. The properAn effective drying approach will minimize the potential for damage of the fuel during the drying operation and subsequent dry storage. ReferenceRef. (1) 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 eladding-cladding (2). 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 Research reactor and other non-commercial SNF that is not treated or reprocessed may be stored in sealed canisters within regulated dry storage systems. Such dry storage canisters may be expected to contain the SNF through interim storage, transport, and repository packaging. The objectives of drying processes used on this fuel are virtually identical to the drying objectives for commercial fuels and are to:
- 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 and material corrosion that present problems during storage, transport, or repository handling operations.
- 5.4 The following objectives of drying processes are common to most fuels and containers:
- 5.4.1 Preclude geometric reconfiguration of the packaged fuel,
- 5.4.2 Prevent damage to the container from over-pressurization,
- 5.4.3 Minimize damage to the internal components of the canister from material corrosion,
- 5.4.4 Minimize hydrogen generation that presents problems during storage, transport, or repository handling operations, and
- 5.4.5 Minimize the risk of the formation of significant quantities of potentially pyrophoric hydrides of metallic fuels (particularly uranium metal).
- 5.5 The selection of the drying methodology for treating fuel for interim-dry storage storage, transportation, or disposition in a geologic repository will involve many factors including the following:
- 5.5.1 Irradiation and storage history (for example, the decay heat output and the amount of hydrogen in the eladding), burnup),
  - 5.5.2 Nature and degree of fuel damage (for example, quantity of breached rods or rods containing water),
- 5.5.3 Forms and quantify of water in the container (for example, absorbed water) water),
  - 5.5.4 Degree to which self-heating may contribute to the drying process,
  - 5.5.5 Impact of residual water on corrosion and degradation of the fuel and container material during drying, storage, transportation, and disposal,
- 5.5.6 Mechanisms and kinetics of water interaction with the fuel and container components, and

- 5.5.7 Nature and quantity of adhered sludge on fuel pins and assembly structures, and
- 5.5.8 Maximum allowable amount of water (including both free and bound water) remaining in the container after drying is completed.
- 5.6 Categorization of SNF and Defects for Drying Evaluation—This standard guide pertains to fuels that contain any eladding 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"

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 intact fuel. It is important when considering a drying process to be clear about the 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 as having breached fuel rods for the purposes of geologic repository disposal. The classifications differ because the performance of damaged fuel is typically more predictable over dry storage periods than over geologic time. SNF may be characterized as damaged 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 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 For purposes of drying treatments and evaluation of drying, only two categories of SNF need to be considered are identified with a number designation. The identities of these categories are provided below-depending on whether or not the fuel is exposed. The categories are:

5.6.1 SNF Group—The following groups are used to distinguish between SNF types: Intact or non-breached fuel, and

A	Commercial oxide fuel in Zr-alloy cladding
B	Carbide or oxide particle fuel with SiC coating (in graphite)
е	Oxide fuel in stainless steel cladding
Đ	U-ZrH fuel in Zr-alloy or stainless steel cladding
E	Oxide fuel in Al alloy cladding
https://stancfards.iteh.ai/	eataloo/standards/sis <del>UAI<sub>x</sub> fuel or U-ZrH fuel in Al-alloy cladding</del> Sc_l_de_S32alb(3/astmcl_S53-24.
<del>G</del>	Metal fuel in Zr-alloy, Al-alloy or stainless steel cladding
H	Carbide or oxide particle fuel with only pyrolytic carbon coating
ŧ	<del>Unclad metal or oxide fuel</del>

5.6.2 Fuel Conditions—The following Fuel Conditions are designated: Breached or failed fuel.

4	Fully intact cladding with no known penetrations
2	Cladding with small pinholes or hairline cracks that may allow entry/exit
	of moisture
3	Fuel with visible or suspected sludge or coatings that may hold
	significant water
4	Cladding with known large cracks or holes that will allow free entry/exit of
	moisture
<del>5</del>	Fuel with major cladding damage from previous fuel oxidation, exposure
	to water, or both
6	Rubblized fuel that has little or no intact cladding, high surface area, and
	previous exposure to water (Includes fuel rods or elements that have
	been sectioned for analysis or study)

- 5.7 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 water vapor, liquid water, ice formed during drying, physisorbed water, and chemisorbed water.
- 5.7.1 *Unbound Water*—Unbound water may be present in containers of SNF transferred from a water storage pool. Water retention depends on the condition of the fuel, fuel and its geometry, the container design, and the drying process. Sources of unbound water after vacuum drying may include trapped water and water in capillaries.

- 5.7.2 *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. <u>Additionally, ice can form during water expansion from cracks</u>. 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 formation (see Annex A2.)).
- 5.7.3 *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/mThe mass of a single-molecule thick (monolayer) physisorbed water layer is reported to be between 0.187 and 0.3 mg/m<sup>2</sup> per(3). monolayer. The The layers of physisorbed water may be partial, or multiple layers, depending on the relative humidity. Additionally, 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 orvaries depending on the number of monolayers with the outer layers being less bound than the first monolayer forced(4). gas dehydration. Cracks, open pores, and corrosion products may hinder evaporation and hold also increases the amount of physisorbed water by virtue of additional surfaces.surfaces Corrosion (4products).on exposed U plate fuel are notable for retaining physisorbed water.
- 5.7.4 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. Small quantities can also be retained by hygroscopic species in pool water. 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 of surface hydroxides such as Al (OH)<sup>3</sup> (or Al<sub>2</sub>O<sub>3</sub>·3H<sub>2</sub>O) which begin dehydrating near 100°C to the form AlO(OH) (or Al<sub>2</sub>O<sub>3</sub>·H<sub>2</sub>O) which is stable to >340°C. Zirconium cladding may also form the hydrated oxides ZrO(OH)<sup>2</sup> or Zr(OH)<sup>4</sup> during irradiation. The water content of hydrated zirconium oxides is small, and the water will not be released below 500°C (25). Carbonaceous deposits with varying thicknesses and morphologies can also form on fuel cladding in graphite moderated, gas-cooled reactors, with thicknesses up to approximately 400 µm. See Annex AlAppendix X2 for other hydroxides and hydrates formed from water contact with typical fuel and container materials.
- 5.8 Sources of Water: Water—It is important to understand and consider the source and location of the water in developing an effective drying technique. Appendix X2 cites recent literature for examples in estimation of water sources (free and bound) in commercial SNF-in-canister configurations post-dried condition, and the impact of those water.
- 5.8.1 General Service Environment for Water Reactor Fuel—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:
  - (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. assemblies, and
  - (5) Neutron absorber.

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.

### 5.8.2 CRUD and Sludge:

5.8.2.1 *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 mg/cm<sup>2</sup> 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  $\mu$ m (0.001 in.) but potentially reaching 100  $\mu$ m (0.004 in.) in thickness (36). CRUD deposits on boiling water reactor fuel average 25 to 76  $\mu$ m (0.001 to 0.003 in.) in thickness and may reach a thickness of 250  $\mu$ m (0.010



- in.) (36). 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.
- 5.8.2.2 Sludge in SNF Operations—Sludge may accumulate in SNF water storage systems from twothree 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. Bothetc., (3) biological materials that enter and grow in storage pools. The 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 (47) showed that a variety of aluminum, iron, and uranium hydrous oxides made up over 90 % of the dry weight of the sludge.
- 5.8.3 Water Associated with Specific Fuel Types:
- 5.8.3.1 Commercial SNF, SNF Group A, Fuel Conditions 1-4—SNF—Light water reactor fuel without any through-cladding defects (Fuel Condition 1) will not allow water inside fuel rods. However, even very small pinholes or 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 gap and the rod plenum after drying. Similar 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 (a1) the dashpot drain hole is blocked or partially blocked with sludge or CRUD, (b2) if the discharge point is elevated above the tube bottom or (e3) 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 breached or failed light water reactor rod in Fuel Condition 4-is characterized by a combination of primary and secondary defects. The primary defect is the original penetration, and secondary defects may be located at some distance from it. The secondary defects are normally attributed to local hydride blistering (58). The defects are holes of different sizes that allow water to penetrate and fill the free volume of the rod. The size and location of the defects may retard water removal. Advanced gas-cooled reactor fuel may retain water in the end caps if retained vertically. Failed fuel pins with annular fuel pellets may accumulate a substantial quantity of water within the center bore.
- 5.8.3.2 Clad Metallic U Fuels (SNF Groups F and G), Fuel Conditions 1–4—Fuels—Clad metallic U and most U allowalloy fuels will not allow water inside intact (Fuel Condition 1) cladding. Vacuum drying of such fuels has been performed for intact Zircaloy-clad fuels from Hanford K-basin (69). Drying tests on unirradiated mock-ups have been performed to demonstrate drying capability for Magnox elements from Sella field water basinspools as a contingency for dry storage (710). However, water ingress through even the smallest pinholes (Fuel Condition 2 or 4) may have a noticeable effect on metallic U fuel. Even at basinpool temperatures, water may oxidize U metal sufficiently to rupture or "unzip" fuel cladding (811). If the oxidation processes cause the internal environment to become sufficiently anoxic, hydrogen will be produced, and the U metal will react to form UH<sub>3</sub>. Exposed surfaces of UH<sub>3</sub> may react vigorously with residual moisture or air (912).
- 5.8.3.3 *Mixed Carbide Fuels, SNF Groups B and H—Fuels*—Mixed carbide fuels encapsulated in pyrolytic carbon, graphite, or both, are designed for gas-cooled reactors and should not 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 <u>due</u> to absorption of water in the pores of the graphite or carbon. An aqueous solution can penetrate the graphite matrix of an HTGR fuel element through its open pore system, and under normal conditions a spherical element takes up about 8 mL of solution (1013).
- 5.8.3.4 Miscellaneous Research and Production Reactor Fuels, SNF Groups C, D, and E—Fuels—A wide variety of research reactor fuels have been irradiated. The response of these fuels to water will depend on the fuel composition, cladding alloy, and cladding integrity. Research reactor fuels generally have low decay heat output, which may dictate the use of specialized heating processes to achieve adequate dryness. Dry storage temperatures and radiation levels may be so low that water radiolysis and secondary oxidation reactions are 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.
  - 5.9 SNF Exposure Environments—The dryness required for a given fuel is often related to the duration of exposure and the radiation, temperature, and water chemistry to which it was exposed during reactor operation and storage. Specific fuels typically have an environmental exposure history that provides input into probable drying requirements. The drying process should reliably establish water vapor pressures levels amounts of residual water such that the remaining water is insufficient to cause detrimental



chemical reactions during dry storage. The potential for water adsorption by hygroscopic species derives from pool water chemistry leading to formation of high strength corrosive droplets on cladding or structural components which should be considered where pool water contains significant quantities of potentially aggressive species.

#### 5.9.1 Commercial Reactor Fuels:

- 5.9.1.1 Commercial nuclear fuel is irradiated in a water environment at elevated temperature and pressure. If a breach of the cladding develops while fuel is in-core, the internal gas will be released and water may enter into the fuel rod. Upon removal from the reactor, the fuel is stored in a water <u>basinpool</u> with the water temperature typically less than 40°C. The water pressure acting on the fuel depends on the depth of the fuel in the <u>basinpool</u> water. Both the reactor and <u>basinpool</u> typically have tightly controlled water chemistries that may prevent or at least minimize fuel cladding damage.
- 5.9.1.2 The heat generated by the SNF during storage drops off predictably as the fission products decay. After a suitable cooling time that is dependent on the fuel burnup, decay heat output, system design, and applicable regulations (1114), the SNF may be moved out of basinpool storage (wet storage) and placed into a dry storage system.
- 5.9.1.3 The thermal performance of a cask or package can be modeled to determine the expected temperature profile as a function of time (1215). Design or regulatory requirements may establish short-term temperature limits for maintaining cladding integrity impacted by creep or by embrittlement, for example. The limits may depend on burnup, cladding design and fuel pressurization. Limits from 250 to 570°C (1316) have been suggested. The evaluation of the limits should consider how cladding integrity is affected by hydride dissolution, reprecipitation, and reorientation, creep, delayed, 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.

#### 5.9.2 Research and Non-commercial Reactor Fuels:

- 5.9.2.1 Research reactors have irradiation temperatures and pressures that vary widely but are typically lower than those of a commercial power plant. Fuel lifetimes are also quite variable in 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. Production reactors may provide the opposite extreme as refueling scheduled to provide the optimum isotope abundances, and the total fuel irradiation time may be less than a year.
- 5.9.2.2 Conditions necessary for successful dry storage of research reactor SNF will depend on the total irradiation, fuel type, and decay heat output. The elimination of reprocessing in the U.S. essentially resulted in placing the vast majority of research and production reactor SNF into extended <a href="https://docs.ps...base-necessary">basinpool</a> storage and a few dry storage systems. The primary considerations involved with movement of these fuels into interim dry storage include the lack of significant decay heat, the wide range of fuel cladding materials, and the 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.
- 5.9.2.3 Two primary types of dry storage systems are currently in use for research reactor SNF: Underground <u>dry</u> 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 not sealed in a container 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 even though the decay heat is insufficient to heat the cask 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 (1417).
- 5.9.2.4 Residual water in vented dry storage systems can evaporate or radiolyze over long times, so water can escape from the system. However, canisters containing cool fuels may also aspirate water from the external atmosphere. Water evaporation and aspiration during "dry" storage may significantly change the overall chemisorbed water content of the SNF, especially if it is badly damaged. Characterization of SNF behavior in such vented systems may provide insight into the probable behavior of SNF in alternative dry storage systems.
- 5.10 Potential Effects of Residual Water on SNF and Containers—Residual water in SNF can be released to the container environment by direct, thermally induced thermally-induced vaporization of physisorbed and free water, decomposition of the chemically bonded species, vaporization of physisorbed and free water, and and radiolytic decomposition. The released water and



decomposition products may cause corrosion, pressurization, and possibly embrittlement, although such degradation is not generally anticipated to have a significant impact on the condition of fuel and storage system (1518).

#### 5.10.1 Radiolysis:

- 5.10.1.1 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 (1619, 1720). 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 on temperature, time, the presence of hydrated oxides, and the cover gas including the amounts of residual air and water; recent experimental results show the profound effects of these parameters on radiolytic yield temperature, (21, 22time,). and the amounts of residual air and water. One calculation for an SNF container with one litre of water (1720) showed that the concentration of hydrogen remained well below the flammability limit for hydrogen/air mixtures after 300 years of storage.
- 5.10.1.2 Neutron radiolysis is important during reactor operation but diminishes rapidly after fuel removal from the active core, and is insignificant by the end of pool (basin or wet) (wet) storage.
- 5.10.1.3 Gamma interactions with water and hydroxyl groups may affect both the fuel and other hydrated compounds inside the cask. Gamma radiolysis of hydrated uranium oxides will occur in fields of 1000 Gy/h (1720). Hydrogen production from dry (no free of physisorbed water) oxyhydroxides of aluminum has been reported (1823). Gamma activity in SNF decreases over time, and the levels of hydrogen, oxygen, and nitric acid developed during storage are generally considered inconsequential even after 300 years (1720). (See also Appendix X2.)
- 5.10.1.4 Beta radiolysis of water occurs only in close proximity to the decay event because of the limited travel of the beta particle. However, if hydrated corrosion products are 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.
- 5.10.1.5 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 known and additional work is needed (1924).
- 5.10.2 Hydrogen—Fuel, Cladding, and Packaging Reactions: (1552.)
- 5.10.2.1 Hydrogen is generated by the radiolytic decomposition of water and by most metal corrosion reactions. In order to ensure that flammable gas cannot be flammable mixture is not present in the event a welded canister needs to be opened, the hydrogen content in SNF containers is usually limited to below 4-%;4 volume%, the lower flammability limit for hydrogen in air (20). 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 An alternative limit for flammability control in a container is oxygen control below 4 volume% in a mixture of any balance gas composed of hydrogen and a non-reactive gas (20, 25). Hydrogen generation rates can be predicted with reasonable accuracy from the temperature, radiation levels, types of materials present, and water content (1720, 2126). See Appendix X2 for a further discussion on hydrogen generation from residual free and bound waters.
- 5.10.2.2 Hydrogen may be a problem should also be considered for SNF container materials over long storage times, although analysis—one calculation has shown (1720) 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, and it may embrittle certain steels, especially high strength ferritic and martensitic steels. The effects of hydrogen in steels are fairly well established, and numerous ASTM test methods are available for evaluating hydrogen effects (2227). Hydrogen may also be absorbed by zirconium-alloy cladding and make it more susceptible to fracture. fracture, although compared to hydrogen picked up in reactor operation, any additional hydrogen absorbed in the cladding will be small in comparison and have a small effect. In general, these effects increase with increases in the hydrogen concentration or in the strength of a metal. Hydrogen content increases with increasing hydrogen fugacity, which is generally greater at a surface during corrosion by aqueous environments than during exposure in gaseous atmospheres and thus is minimal in dry storage systems compared to in-core (2328). Hydrogen entry into fuel or container materials may also be driven by galvanic corrosion. High-pressure hydrogen effects data, in general, should not be used to predict the impact of hydrogen on SNF storage eontainers containers (see Note 1). Austenitic stainless steels and low-strength ferritic and ferritic-pearlitic steels are relatively insensitive to low-pressure hydrogen exposures.



Note 1—Unpublished SRS data on testing of austenitic stainless steel tritium shipping containers used intermittently for 15 years to hold tritium at 1 psig indicated that tritium did diffuse into the steel structure, but to a depth less than that required to cause fracture unless the material was highly stressed.

- 5.10.2.3 The hydrogen concentration in a sealed container depends on the extent of reaction of water with fuel and container materials. Assuming that free and physisorbed water are removed by drying, the issue of adequate dryness may relate directly to:
  - (1) The mass of hydrogen in the chemisorbed water within the system,
  - (2) The potential for thermal or radiolytic decomposition of the compounds holding the water, hydrogen, or both,
  - (3) The rate of hydrogen generation by corrosion from chemisorbed water that is released from the compounds,
  - (4) The hydrogen diffusion, venting, corrosion, gettering, and recombination rates during interacting with the system,
  - (5) Free volume within the container,
  - (6) The rate of hydrogen reaction with metals such as zirconium-based alloys, and
  - (7) Mass of metal that may absorb hydrogen.
- 5.10.2.4 Hydrogen gettering may be an effective technique to mitigate hydrogen buildup in storage containers if the radiation levels are low and the hydrogen is not radiolyzed from the getter material. However, effective gettering may require high temperatures, so getters may have limited utility for long-term storage.
- 5.10.3 Water Corrosion Reactions:
- 5.10.3.1 The expected quantities of residual water expected after drying are typically small relative to the substantial internal surface area of typical dry storage containers and the mass of the fuel and cladding, so water corrosion damage to the structural materials and SNF should not be significant in establishing adequate dryness. However, there are potential exceptions to the anticipated lack of corrosion damage, including:
  - (1) Small containers of badly damaged fuel materials previously exposed to water,
  - (2) Fuels that may contain large quantities of water that cannot be removed with drying processes,
- (3) Fuels that would be expected to release aggressive fission products and reach a temperature sufficient to allow corrosion cracking of container welds, and
  - (4) Fuels with significant chloride contamination.

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- 5.10.4 Fission Product Reactions:
- 5.10.4.1 Some fission products could be released from fuel during storage. These could react with residual water and increase the corrosiveness of the storage environment. Cesium, rubidium, and iodine are the fission products of primary concern. Krypton and Xenon may add to internal container pressures, and decay of krypton to rubidium may help spread rubidium throughout the container. Cesium and rubidium may react with residual water to form caustic hydroxides that could lead to caustic cracking of stainless steel weldments at elevated dry storage temperatures (>110°C). Iodine would be expected to behave similarly to chlorine in attacking stainless steel packaging components if sufficient residual tensile stress and ion concentrations are present. Fission product interactions are not expected to present major problems, but they should not be overlooked when eriteria-dryness criteria are established.
- 5.10.5 Galvanic Coupling with Aluminum Clad Fuel:
- 5.10.5.1 Internal water corrosion is a primary concern for the storage of aluminum components if residual <u>water</u> is present. Large quantities of stainless steel are typically present in storage containers, and galvanic coupling between the stainless steel and aluminum can occur if sufficient electrolyte is present. Galvanic coupling will result in accelerated corrosion of the aluminum components. This process is especially important with relatively cold aluminum clad fuel in vented storage systems where water ingress is possible. Considerations for magnesium-clad fuels are similar.
  - 5.10.6 Carbide Fuel—Water Reactions:
  - 5.10.6.1 Carbide fuels, which represent only a small fraction to the SNF inventory, are irradiated in a gas atmosphere and are normally not stored in water. However, many carbide fuels have come into contact with water due to reactor or storage incidents. The majority of carbide fuels are coated spheres of uranium carbide, thorium carbide, or both, that are dispersed in a carbonaceous matrix. The coating material is often SiC. The fuel particles are dispersed in porous compacts of pyrocarbon and typically encapsulated in a graphite sheath or block. If the SiC is penetrated, the reaction of the fuel with moisture may be quite rapid.
  - 5.10.6.2 The intrinsic rate of hydrolysis of  $ThC_2$  and  $UC_2$  in moisture proceeds at a penetration rate as fast as 24  $\mu$ m/day. Bulk



samples of  $ThC_2$  powder hydrolyzed completely in ambient laboratory air within 12 hoursh (2429). Uranium and thorium carbides react with water or water vapor to form hydrogen and low molecular weight hydrocarbons:

$$(Th, U)C_2 + H_2O \rightarrow (Th, U)O_2 + H_2 + hydrocarbons$$
 (1)

 $(Th, U)C+H_2O\rightarrow (Th, U)O_2+H_2+hydrocarbons$ 

 $(Th, U)C_2 + H_2O \rightarrow (Th, U)O_2 + H_2 + hydrocarbons$  $(Th, U)C + H_2O \rightarrow (Th, U)O_2 + H_2 + hydrocarbons$ 

5.10.6.3 The low molecular weight hydrocarbons are primarily methane  $(CH_4)$ , ethylene  $(C_2H_4)$ , and ethane  $(C_2H_6)$ , with minor amounts of acetylene  $(C_2H_2)$  and the  $C_3H_x$ to  $C_6H_x$  alkanes, alkenes, and alkynes (25, 25-30-3035). The reported product distributions vary substantially and reflect the effects of impurities in the carbides and variations in analysis techniques. Careful consideration should be given to the flammability of the hydrocarbons if damage to the SiC cladding on the carbide spheres is established or anticipated.

5.10.6.4 The reaction of graphite or pyrocarbon with water <u>producesproducing</u> hydrogen, CO, and CO<sub>2</sub> is extremely slow at temperatures below 200°C, so that the reaction is considered to be of no consequence at temperatures anticipated for dry storage (31-36-3338).

5.10.6.5 Silicon carbide reacts with water vapor to form silica ( $SiO_2$ ), carbon dioxide, carbon monoxide, and hydrogen at temperatures above 600°C, and silica, methane, hydrogen, carbon dioxide, and carbon monoxide below 600°C (3439, 3540). However, the reaction is extremely slow at temperatures below 500°C (2126) and is therefore not considered to be important.

5.10.7 Water-Oxide (Fuel) Reactions and Consequences:

5.10.7.1 Cladding damage may lead to water ingress into the fueled fordsrods and subsequent water retention. Residual water may oxidize the fuel pellets toward a low density UO<sub>3</sub> hydrate and may subsequently rupture or "unzip" the fuel cladding (see Annex A1 for fuel oxide reaction data). The cladding rupture is a direct result of the volume expansion from hydrated compound formation. For example, the hydrated compound UO<sub>3</sub>-2H<sub>·</sub> 2H<sub>2</sub>O has a volume 2.6 times that of the starting UO<sub>2</sub>. Evaluations of the reaction process indicate that UO<sub>2</sub> begins to form hydrated phases within six weeks if exposed to moisture at fuel storage temperatures (3641). Additionally, sintered UO<sub>2</sub> forms metaschoepite when reacted with deionized water. The peroxide phases, studite and meta-studtite, may form if radiolysis of residual water produces hydrogen peroxide (3742). The formation of low-density hydrated compounds and resulting cladding rupture may impactaffect the handling and transport of SNF.

https://standards.iteh.ai/catalog/standards/sist/32780937-b3f2-4536-bc61-5c1de532abf3/astm-c1553-2

#### 5.10.8 *Gas Pressurization:*

5.10.8.1 Pressure inside the storage container is increased both by reactions that form gas as a reaction product and by failure of cladding and the subsequent release of gas from spent fuel. However, reaction of these gases with constituents inside the storage container can decrease the pressure build up. The internal pressure of the storage container will be determined by the gas generation and reaction processes. Quantitative estimates of the pressure will need to consider the following variables:

- (1) Free volume of the container,
- (2) Quantity of free, physisorbed, and chemisorbed water,
- (3) Location of the water relative to high radiation fields,
- (4) Inventory of gaseous radioactive decay products,
- (5) Radiogenic gas formation rates, rates (including He formation from alpha particle emissions),
- (6) Inventory and available surface area of solid decay products,
- (7) Inventory of dust, CRUD, and sludge,
- (8) Decrease in temperature as decay heat output declines,
- (9) Initial pressure and composition of the fill gas,
- (10) Container and basket materials corrosion rates,
- (11) The number of damaged fuel rods and the fission product content of the rods, and
- (12) Corrosion, including galvanic corrosion, of fuel cladding and assembly materials.

5.10.8.2 The inability to accurately predict pressure and hydrogen concentration within a sealed system affects the design criteria for the container. Regulators and designers typically assume that only reactions that increase pressure are occurring. Such calculations yield pressures up to 0.35 MPa (50.8 psi) for a commercial fuel container with hydrated uranium oxides heated to 250°C by decay heat after sealing. See example calculation in Appendix X1.