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# Standard Guide for Characterization of Spent Nuclear Fuel in Support of Interim Storage, Transportation and Geologic Repository Disposal[1]

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## 1. Scope

1.1 This guide provides guidance for the types and extent of testing that would be involved in characterizing the physical and chemical nature of spent nuclear fuel (SNF) in support of its interim storage, transport, and disposal in a geologic repository. This guide applies primarily to commercial light water reactor (LWR) spent fuel and spent fuel from weapons production, although the individual tests/analyses may be used as applicable to other spent fuels such as those from research and test reactors and mixed oxide (MOX) spent fuel. The testing is designed to provide information that supports the design, safety analysis, and performance assessment of a geologic repository for the ultimate disposal of the SNF.

1.2 The testing described includes characterization of such physical attributes as physical appearance, weight, density, shape/geometry, degree, and type of SNF cladding damage. The testing described also includes the measurement/examination of such chemical attributes as radionuclide content, microstructure, and corrosion product content, and such environmental response characteristics as drying rates, oxidation rates (in dry air, water vapor, and liquid water), ignition temperature, and dissolution/degradation rates. Not all of the characterization tests described herein must necessarily be performed for any given analysis of SNF performance for interim storage, transportation, or geological repository disposal, particularly in areas where an extensive body of literature already exists for the parameter of interest in the specific service condition.

1.3 It is assumed in formulating the SNF characterization activities in this guide that the SNF has been stored in an interim storage facility at some time between reactor discharge and dry transport to a repository. The SNF may have been stored either wet (for example, a spent fuel pool), or dry (for example, an independent spent fuel storage installation (ISFSI)), or both, and that the manner of interim storage may affect the SNF characteristics.

1.4 The values stated in SI units are to be regarded as standard. No other units of measurement are included in this standard.

1.5 *This standard does not purport to address all of the safety concerns, if any, associated with its use. 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.*

1.6 *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.*

## 2. Referenced Documents

2.1 *ASTM Standards:*[2]

C170/C170M Test Method for Compressive Strength of Dimension Stone

C696 Test Methods for Chemical, Mass Spectrometric, and Spectrochemical Analysis of Nuclear-Grade Uranium Dioxide Powders and Pellets

C698 Test Methods for Chemical, Mass Spectrometric, and Spectrochemical Analysis of Nuclear-Grade Mixed Oxides ($(U, Pu)O_2$)

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

C1380 Test Method for the Determination of Uranium Content and Isotopic Composition by Isotope Dilution Mass Spectrometry

C1413 Test Method for Isotopic Analysis of Hydrolyzed Uranium Hexafluoride and Uranyl Nitrate Solutions by Thermal Ionization Mass Spectrometry

C1454 Guide for Pyrophoricity/Combustibility Testing in

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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.

Support of Pyrophoricity Analyses of Metallic Uranium Spent Nuclear Fuel (Withdrawn 2016)[3]
C1553 Guide for Drying Behavior of Spent Nuclear Fuel
E170 Terminology Relating to Radiation Measurements and Dosimetry

2.2 *U.S. Government Documents*[4]
Code of Federal Regulations, Title 10, Part 60 Disposal of High-Level Radioactive Wastes in Geologic Repositories, U.S. Nuclear Regulatory Commission
Code of Federal Regulations, Title 10, Part 63 Disposal of High-Level Radioactive Wastes in a Geologic Repository at Yucca Mountain, Nevada, U.S. Nuclear Regulatory Commission
Code of Federal Regulations, Title 10, Part 71 Packaging and Transport of Radioactive Materials
Code of Federal Regulations, Title 10, Part 72 Licensing Requirements for the Independent Storage of Spent Nuclear Fuel and High-Level Radioactive Waste
Code of Federal Regulations, Title 10, Part 961 Standard contract for the Disposal of Spent Nuclear Fuel and/or High Level Waste
Code of Federal Regulations, Title 40, Part 191 Environmental Radiation Protection Standards for Management and Disposal of Spent Nuclear Fuel, High-Level and Transuranic Radioactive Wastes
Code of Federal Regulations Title 40, Part 197 Protection of Environment: Public Health and Environmental Radiation Standards for Yucca Mountain, Nevada

## 3. Terminology

3.1 *Definitions*—Definitions used in this guide are as currently existing in Terminology C859 or Test Method C170/C170M, or as commonly accepted in dictionaries of the English language, except for those terms defined below for the specific usage of this standard. For consistency, many of the definitions are based on definitions from Federal Regulations in the United States.

3.2 *Definitions of Terms Specific to This Standard:*

3.2.1 *alteration, n*—any change to the form, state, or properties of a material.

3.2.2 *attribute test, n*—a test conducted to provide material properties that are required as input to materials behavior models, but are not themselves responses to the materials environment (for example, thermal conductivity, mechanical properties, radionuclide content of waste forms, etc).

3.2.3 *characterization test, n*—any test conducted principally to furnish information for a mechanistic understanding of alteration (for example, electrochemical polarization tests, leach tests, solubility tests, etc).

3.2.4 *combustible, adj*—capable of burning or undergoing rapid chemical oxidation.

3.2.5 *breached fuel, n*—(per Code of Federal Regulations, Title 10, Part 72, Section 122(h)) any spent fuel with extreme degradation or gross rupture, such that fuel particulates or pieces can be released from the fuel rod. ("The spent fuel cladding must be protected during storage against degradation that leads to gross ruptures or the fuel must be otherwise confined such that degradation of the fuel during storage will not pose operational safety problems with respect to its removal from storage," Code of Federal Regulations, Title 10, Part 72, Section 122(h)). It is not expected that minor cladding defects such as pinhole cracks would permit significant release of particulate matter from the spent fuel rod.

3.2.6 *damaged fuel, n*—spent nuclear fuel elements or assemblies that as a result of their irradiation or handling (or both) have significantly altered dimensions or cladding through-wall cracks or penetrations such that it cannot fulfill its direct or indirect regulatory or design function. For example any SNF assembly with rod(s) that are significantly displaced for purposes of criticality calculations (application dependent and function of the stage in the nuclear fuel cycle).

3.2.7 *degraded cladding, n*—spent fuel cladding which has corroded or been physically altered in-reactor or during subsequent interim storage (or both), to the extent that the alteration must be accounted for in the evaluation of its behavior during transport, storage, or disposal (for example, cladding corrosion/thinning, hydride embrittlement, etc.).

3.2.8 *failed fuel (geologic disposal), n*—any significant alteration in the shape, dimensions, or configuration of a spent fuel assembly or fuel element, or through-wall crack in the cladding, that could degrade or open further under long-term exposure to the repository environment.

3.2.9 *failed fuel (interim storage and transport), n*—fuel rods/assemblies whose cladding has been perforated to the extent that powder or pieces of the fuel can relocate or be released from the cladding.

3.2.9.1 *Discussion*—Code of Federal Regulations, Title 10, Part 961, the Standard Contract between the USDOE and the US commercial nuclear utilities defines categories of commercial LWR spent fuel as "Standard," "Non-Standard," and "Failed." These categories are based on the type of handling—normal or special—required for transport and storage of the SNF. The "Standard" classification includes most normal and handle-able LWR (PWR and BWR) spent fuel. "Non-Standard" spent fuel includes non-LWR spent fuel, consolidated fuel, older design fuel, etc. "Failed" fuel includes: Class F-1: (via visual examination) visual failure or damage—"Assemblies which (i) are structurally deformed or have damaged cladding to the extent that special handling may be required or (ii) for any reason cannot be handled with normal fuel handling equipment …" Class F-2: radioactive " leakage" or "any fuel that allows gaseous communication between the inside and the outside of the cladding." Class F-3: Encapsulated—Note that the terms used in this guide for failed fuel, damaged fuel, and degraded cladding can fit the "Failed Fuel" definition of Code of Federal Regulations, Title 10, Part 961. Also, the Code of Federal Regulations, Title 10, Part 961 categories of spent fuel are partially based on the fact that the

[3] The last approved version of this historical standard is referenced on www.astm.org.

[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.

repository is required by statute to accept all commercial LWR spent fuel, including damaged/failed.)

3.2.10 *ignite, v*—to cause to burn and reach a state of rapid oxidation, which is maintained without requiring an external heat source.

3.2.11 *model, n*—a simplified representation of a system or phenomenon, often mathematical.

3.2.12 *performance assessment (PA), n*—an analysis that identifies the processes and events that might affect the disposal system; examines the effects of these processes and events on the performance of the disposal system; and, estimates the cumulative releases of radionuclides, considering the associated uncertainties, caused by all significant processes and events. These estimates shall be incorporated into an overall probability distribution of cumulative release to the extent practicable (see Code of Federal Regulations, Title 10, Part 63 Section 2) and Code of Federal Regulations, Title 40, Part 191 Section 15).

3.2.13 *pyrophoric, adj*—capable of igniting spontaneously under temperature, chemical, or physical/mechanical conditions specific to the storage, handling, or transportation environment.

3.2.14 *sibling sample, n*—one of two or more test samples that are nearly indistinguishable with respect to their chemical and physical properties.

3.2.15 *spent nuclear fuel (SNF), n*—nuclear fuel that has been exposed to, and removed from, a nuclear reactor.

3.2.16 *waste form (WF), n*—(from Practice C1174) the radioactive waste materials and any encapsulating or stabilizing matrix in which it is incorporated.

3.2.17 *waste package (WP), n*—(from Practice C1174) the waste form and any containers, shielding, packing and other absorbent materials immediately surrounding an individual waste container.

## 4. Summary of Guide

4.1 The characterization of spent nuclear fuel (SNF)—in support of interim storage, transport, and disposal in a geologic repository—described in this guide includes the examination/testing of such physical attributes as physical appearance, weight, density, shape/geometry, degree and type of cladding damage, etc. It also includes the measurement/examination of such chemical aspects as drying characteristics, water content, radionuclide content, microstructure, zirconium hydride content (of commercial SNF cladding), uranium hydride content (of metallic uranium SNF), and such environmental response characteristics as oxidation rate (in dry air, water vapor, and liquid water), ignition temperature, and dissolution/degradation rates.

4.2 The primary issues involved in the characterization of uranium dioxide-based commercial light water reactor (LWR) SNF are the fraction of fuel rods with non-intact cladding (that is, the amount of "failed fuel" as defined in Section 3 above), the structural integrity of the fuel assembly (that is, the amount of "damaged fuel" as defined in Section 3 above), the amount and structure of zirconium hydride in the cladding (for example, "degraded cladding" as defined in Section 3 above), particularly with respect to high burnup LWR SNF. Also, the radionuclide content of the fuel, the thickness of the zirconium oxide on the external surface of the cladding, and the leaching/dissolution behavior characteristics when in contact with the (repository-relevant) air/water environment are factors that could affect SNF behavior in repository disposal.

4.3 The primary issue involved in characterization of metallic uranium SNF is the extent of damage to the cladding (that is, exposure of metallic uranium to air and water) and its consequently enhanced chemical activity and pyrophoricity/combustibility characteristics. Metallic uranium SNF, largely from plutonium production reactors, has been temporarily stored in water basins in several countries prior to reprocessing or ultimate direct disposal of unreprocessed fuel. In some cases the manner of discharge (for example, those involving physical trauma to the fuel element) of the fuel elements from these reactors, and the type of wet storage environment in which they were emplaced after discharge, has resulted in significant amounts of fuel cladding damage and extensive corrosion of the consequently exposed uranium metal. This corrosion and damage has resulted in alteration of the physical integrity/dimensions of the elements and the chemical reactivity of the material such that the physical and chemical properties of the material no longer straightforwardly resemble, or can be represented by, the properties of the as-fabricated, unirradiated fuel.

## 5. Significance and Use

5.1 In order to demonstrate conformance to regulatory requirements and support the post-closure repository performance assessment information is required about the attributes, characteristics, and behavior of the SNF. These properties of the SNF in turn support the transport, interim storage, and repository pre-closure safety analyses, and repository post-closure performance assessment. In the United States, the interim dry storage of commercial LWR SNF is regulated per the Code of Federal Regulations, Title 10, Part 72, which requires that the cladding must not sustain during the interim storage period any "gross" damage sufficient to release fuel from the cladding into the container environment. In other countries, the appropriate governing body will set regulations regarding interim dry storage of commercial LWR SNF. However, cladding damage insufficient to allow the release of fuel during the interim storage period may still occur in the form of small cracks or pinholes. These cracks/pinholes could be sufficient to classify the fuel as " failed fuel" or "breached fuel" per the definitions given in Section 3 for repository disposal purposes, because they could allow contact of water vapor or liquid with the spent fuel matrix and thus provide a pathway for radionuclide release from the waste form. Also, pinholes/cracks in fuel rods in dry or wet interim storage can also develop into much larger defects (for example, the phenomenon of cladding "unzipping") under long-term repository conditions. Therefore SNF characterization should be adequate to determine the amount of "failed fuel" for either usage as required. This could involve the examination of reactor operating records, ultrasonic testing, sipping, and analysis of the residual water and drying kinetics of the spent fuel assemblies or canisters.

5.2 Regulations in each country may contain constraints and limitations on the chemical or physical (or both) properties and long-term degradation behavior of the spent fuel and HLW in the repository. Evaluating the design and performance of the waste form (WF), waste packaging (WP), and the rest of the engineered barrier system (EBS) with respect to these regulatory constraints requires knowledge of the chemical/physical characteristics and degradation behavior of the SNF that could be provided by the testing and data evaluation methods provided by this guide, using the United States as an example, as follows:

5.2.1 In the United States, for example, Code of Federal Regulations, Title 10, Part 60 Sections 135 and 113 require that the WF be a material that is solid, non-particulate, non-pyrophoric, and non-chemically reactive, that the waste package contain no liquid, particulates, or combustible materials and that the materials/components of the EBS be designed to provide—assuming anticipated processes and events—substantially complete containment of the HLW for the NRC-designated regulatory period.

5.2.2 In the United States, for example, Code of Federal Regulations, Title 10, Part 63 Section 113 requires that the EBS be designed such that, working in combination with the natural barriers, the performance assessment of the EBS demonstrates conformance to the annual reasonably expected individual dose protection standard of Code of Federal Regulations, Title 10, Part 63 Section 311 and the reasonably maximally exposed individual standard of Code of Federal Regulations, Title 10, Part 63 Section 312, and shall not exceed EPA dose limits for protection of groundwater of Code of Federal Regulations, Title 10, Part 63 Section 331 during the NRC-designated regulatory compliance period after permanent closure.

5.2.3 In the United States, for example, Code of Federal Regulations, Title 10, Part 63 Section 114 (e), (f), and (g) and Code of Federal Regulations, Title 10, Part 63 Section 115 (c) require that a technical basis be provided for the inclusion or exclusion of degradation/alteration processes pertinent to the barriers of the EBS, and that likewise a technical basis be provided for the degradation/alteration models used in the post-closure performance assessment of the capability of the EBS barriers to isolate waste.

5.3 The enhanced chemical reactivity and degraded condition of corroded/damaged uranium metal-based SNF must be accounted for in both the pre-closure safety analyses and the post-closure performance assessment of the geologic repository. An example of this would be the potential for pyrophoric behavior in uranium metal-based SNF (see Guide C1454). Due to the combustibility of the metallic uranium or uranium hydride (or both), and the enhanced aqueous dissolution rates for the exposed uranium metal, the potential for enhanced chemical activity or pyrophoric behavior must be factored into the repository or interim storage facility safety analyses, and assessments of the potential for radionuclide releases from the repository site boundary after repository closure.

5.4 Characterization of several key properties of SNF may be required to support the design and performance analyses of both repository above-ground SNF receipt and lag storage facilities, the WP into which the SNF is placed, and the subsurface permanent emplacement drift EBS.

5.4.1 Repository waste package design must ensure that the waste to be placed in the repository can be accommodated within the radionuclide and thermal loading ranges of the waste package drift emplacement licensing conditions. To do this the radionuclide content and oxidation rate when exposed to oxygen/water environments should be determined.

5.4.2 The condition of the LWR spent fuel cladding (particularly with respect to hydride content and morphology) could potentially influence the performance of the cladding in interim storage, transportation, and geologic repository disposal. The corrosion and consequent failure rate of cladding with high hydride content may be greater than that of low or no hydride content. If the performance assessment is found to be sensitive to the failure rate of the cladding, it may be necessary to perform zirconium hydride content and orientation testing, particularly for high burnup LWR SNF.

5.4.3 Metallic uranium-based spent fuel introduces aspects of chemical reactivity, such as combustibility and pyrophoricity (see C1454), that should be addressed in WP design and performance assessment, and in safety analyses associated with interim storage and transportation prior to repository emplacement. Metallic uranium-based nuclear fuel has been widely used in nuclear reactors; sometimes for commercial reactors (for example, Magnox) but more often in plutonium and tritium production reactors. The manner of discharge of metallic uranium SNF from these production reactors, and/or the manner of temporary wet storage of that portion of the spent fuel that was not reprocessed has in many instances resulted in significant corrosion and mechanical damage to the SNF assemblies. This damage has resulted in the direct exposure of the metallic uranium to the basin water. The relatively high chemical reactivity of uranium in contact with water can result in significant physical damage to the assemblies as the result of corrosion product buildup, and the creation in the exposed fuel surface and fuel matrix of uranium hydride inclusions which in turn further increase the chemical activity of the material. The reaction of this spent fuel with air, water vapor, or liquid water can introduce a significant heat source term into design basis events. In order to support the evaluation of these events, the physical condition (that is, the degree of optically/visually observable damage), the chemical oxidation kinetics, the ignition characteristics, and radionuclide release characteristics of the SNF should be investigated.

5.4.4 The thermal analysis of the waste package/engineered barrier system requires quantification of the potential chemical heat source. To determine this, the amount of reactive uranium metal in the waste canisters sent to the repository should be provided so the thermal analysis of the waste package/ engineered barrier system can be performed.

5.4.5 Radionuclide inventories and physical/chemical characteristics are required to enable storage canister, transportation package, and WP loading and emplacement configurations to be developed.

5.4.6 Repository WP materials selection and design must account for the potential interactions between the waste and

WP. The potential chemical forms of the wastes must be considered, and the effects of residual water or impurities (or both) should be evaluated.

5.4.7 The history of the SNF interim storage and transportation conditions prior to delivery to the repository is important whenever the storage conditions may have altered the degradation characteristics of the SNF (for example, with respect to hydride content and morphology in high burnup LWR SNF cladding). Interim dry storage of commercial SNF requires that the fuel cladding should not sustain gross damage during the storage period to the extent that fuel is released from the fuel rods into the canister. Small pinholes or cracks may form in the cladding during the storage period without violating this interim storage requirement, but may cause the fuel to be classified as failed fuel for repository disposal purposes. The objective of drying commercial SNF fuel is thus to preclude gross damage for interim storage purposes. If the conditions of transport or interim storage are such that there is a significant potential for further degradation of the SNF or change in properties important to the repository pre-closure safety or post-closure performance analyses, the characterization should provide sufficient information to evaluate these changes.

## TEST METHODS—IN SUPPORT OF LICENSING

### 6. Testing Requirements and Scope

6.1 The tests described below are designed to support license application safety analyses from the appropriate governing body (an example from the United States is SNF transport per Code of Federal Regulations, Title 10, Part 71, interim dry storage per Code of Federal Regulations, Title 10, Part 72, and the repository disposal per Code of Federal Regulations, Title 10, Parts 60 and 63). They also support the repository performance assessment by providing EBS component behavior models in conformance with methods described in Practice C1174. (Testing and analysis in support of EBS performance confirmation are described in Section 6.10.)

6.2 Physical Appearance, and Extent and Distribution of Visible Cladding Damage Optical, videotape, and other visual examinations of the spent fuel assemblies would be performed to assess the degree of damage to the assemblies, the amount and condition of corrosion product, the condition of the cladding (including pinholes or cracks in LWR spent fuel cladding, in the United States this would be sufficient to classify the SNF as "failed fuel" per the requirements of Code of Federal Regulations, Title 10, Part 961), and the basic ability to transfer, handle, and emplace the assemblies into canisters for drying and long-term storage. These examinations would be used to determine the initial conditions for the spent fuel assemblies for further conditioning and storage, such as evaluating the adequacy of previous storage, designing and conducting packaging operations, and evaluating any need for further treatment of the spent fuel prior to long-term storage. The examinations could also aid in distinguishing "failed fuel" for the purposes of interim storage (that is, gross damage) from "failed fuel" for the purposes of repository disposal (that is, pinholes and small cracks). A number of NDE technologies may be utilized to locate failed fuel. This includes utilizing fuel bundle sipping technology to identify fuel bundles with a breach fuel rod and various and Eddy Current technology to identify the particular breached fuel rod. The information obtained in these examinations could also be used to sort the spent fuel assemblies into categories of damage. These categories can be then used for the purposes of sorting the fuel assemblies for emplacement in long-term storage canisters, or selecting certain kinds of damaged fuel elements for further conditioning prior to canister emplacement (or both).

6.2.1 (Typical test method and data obtained) The mechanical integrity of the SNF elements and fuel cladding can be determined by several methods including crush tests or ring compression tests. Scanning electron microscopy (SEM) and optical microscopy can be used to measure the resulting particle sizes.

6.2.2 (Typical test method and data obtained.) In the Transfer of SNF from wet storage to a storage or transportation canister adherent sludges may need to be removed. The settling time of the sludge in a graduated cylinder will be measured and the particle sizes of the sludge could be used to determine filtration needs during sludge movement.

6.2.3 (Typical test method and data obtained.) SNF relocation in canisters could be detected by visual observation in a hot cell. A detailed visual inspection of the SNF elements may be done in the hot cell to detect intact or broken elements, cracks, and visible pinholes.

6.2.4 Determination of the integrity of the LWR cladding can be aided by visual examination of whole assemblies or individual rods extracted from assemblies (example: governed in the United States by Code of Federal Regulations, Title 10, Part 961 failed fuel classification F-1). In addition cladding integrity can be examined using ultrasound or Eddy current testing to determine the existence of pinholes or cracking not visible to the eye (example: governed in the United States by Code of Federal Regulations, Title 10, Part 961 failed fuel classification F-2).

6.2.5 (Typical data requirement) Fuel rod profilometry measurements can be used to indicate where fuel rod internal stress limits have been exceeded, resulting in excessive cladding creep and consequent ballooning of the fuel rod.

6.2.6 (Typical data requirement) Cladding "crud" deposits thickness and quantity.

6.2.7 (Typical data requirement) Fuel rod and cladding thermal history as provided by the utility owner of the SNF.

6.3 *Weight/Size/Dimensions*—The weight, size, physical dimensions, or combinations thereof, of the SNF assemblies from their as-fabricated condition would be measured in order to support storage canister, transportation canister, or waste packaging design. The difference between the as-fabricated weight and dimensions, and those existing after long periods of pool storage should be established. These determinations could be made through visual examination of the assemblies in the storage pool, or on representative examples of fuel assemblies transported to hot cells specifically for these measurements.

6.4 *Radionuclide and Fissile Content*—The elemental and isotopic inventory in the spent fuel should be determined in order to support the geologic repository requirements, including EBS radionuclide release characteristics, and support the repository thermal analyses by providing the radionuclide decay heat source term. Elemental and isotopic inventories can be determined using such radiochemical techniques as inductively coupled mass spectrometry, spark source mass spectrography, alpha and gamma spectrometry, etc (see Test Methods C696, C698, C1380, C1413). Redistribution of the elements/isotopes could be determined by taking samples from different sections of the fuel elements.

6.4.1 (Typical data requirement) Application of ORIGEN, SCALE, or similar validated fissile depletion and radionuclide inventory calculation code.

6.4.2 (Typical data requirement) Dissolution of SNF into nitric acid and subsequent application of the spectrochemical techniques described in Test Methods C1380 or C1413 to measure the elemental or isotopic (or both) inventory in the samples.

6.5 *Metallography/Optical Microscopy*—Metallographic examinations of small sections of LWR spent fuel can be used to reveal (*1*) the amount and morphology of zirconium hydride in the spent fuel cladding, (*2*) the microstructural characteristics (for example, grain size, distribution of intermetallic particles, porosity, etc.), and (*3*) the amount and morphology of uranium hydride or carbide inclusions in metallic uranium spent fuel (which could affect such other properties of the fuel as its pyrophoricity/combustibility, oxidation, dissolution, and drying characteristics).

6.5.1 *Zirconium or Uranium Hydride (or both) Content*—Zirconium hydride inclusions in SNF cladding, formed primarily as a product of the in-reactor corrosion of the zircaloy, can degrade the mechanical properties (such as ductility, rupture strength, etc) of the cladding. Uranium hydride, a product of the corrosion of uranium metal in water, can be a source of pyrophoric behavior in uranium metal-based spent fuel. Knowledge of the quantity, distribution, and morphology of hydrides can be important in evaluation the chemical reactivity and integrity of the spent fuel. Hydrogen content can be determined from the type of metallographic examination described above, and also by gas effusion or thermogravimetric techniques.

6.5.2 (Typical test method and data obtained.) Use LECO-type gas effusion analyzer to heat zircaloy, uranium metal, or SNF samples (or combinations thereof) to high temperature in an inert gas (such as argon) and measure the quantity/volume of hydrogen gas released.

6.5.3 (Typical test method and data obtained.) Microscopically examine cut-and-polished sections of the metallic uranium SNF. Polishing and etching techniques are used on sectioned samples of the SNF to reveal microstructural features and highlight uranium hydride and other inclusions in the uranium metal matrix. Uranium hydride inclusions are generally identified after mechanical polishing, heat tinting, and acid etching as needles/stringers (light brown or silver under bright field light or gray under polarized light) in the uranium matrix. This technique enables an estimate of the quantity of uranium hydride within the uranium metal fuel matrix, as well as a determination of the location of hydride concentrations, for example, near the corrosion layer, underneath cladding, etc.

6.6 *Water Content and Drying Characteristics:*

6.6.1 Knowledge of the residual water content of canisters containing uranium metal-based SNF is necessary to perform criticality analyses, analysis of the potential pressurization of the canisters, estimates of the potential for generating combustible materials, and the development of drying techniques for the SNF canisters prior to sealing for interim storage. The water retained by the SNF may be composed of free water, physisorbed water, or chemisorbed water (or combinations thereof). The amount of residual water retained in LWR SNF assemblies or in SNF canisters can be determined through direct measurement, or implementation of a validated drying procedure, or a combination of both.

6.6.2 The required drying temperatures and process times must reflect the condition of the SNF and the expected amounts of free and chemically bound water that need to be removed. The rate of drying is a function of the applied temperature, the composition of the materials in the system, the ambient pressure, and the specific convection/diffusion restrictions imposed by the materials and the drying system geometry.

6.6.3 Free water removal is primarily limited by the geometry of the drying system and the physical location of the water in the system. Free water that is isolated from the vacuum source, trapped inside fuel cladding, or within fuel basket structures can still be removed with adequate time and temperature applied.

6.6.4 Removal of physisorbed water depends on the relative humidity in the system that relates directly to the number of superficial water layers that must be desorbed. Dry air should desorb the superficial physisorbed water layers in 10 to 30 hours. Less time is required with a vacuum at 20°C.

6.6.5 Removal of the interfacial monolayer of physisorbed water is much more difficult, requiring heating in dry air or in a vacuum at 100–200°C. However, there is some evidence that certain complexed physiosorbed waters will only desorb at temperatures above 350°C. Water physisorbed into $UO_2$ has been shown to desorb starting about 150°C with the reaction essentially complete at 230°C. Removal of chemisorbed water depends on the specific chemical species and purity of chemical species involved.

6.6.6 There are several methods for determining the amount of residual water in the SNF (SNR—Scanning Neutron Radiography, NMR—Nuclear Magnetic Resonance, Gamma Radiography). The particular method or methods chosen will depend on the manner in which residual water will be determined. A database for the amount of residual water remaining in small samples after a drying procedure could be used to certify the drying procedure, and conformance to the drying technique under QA controls could be taken as certification that dryness has been achieved. Such drying parameters as temperatures, vacuum level, time, and the number of backfill-and-evacuate cycles must reflect the condition of the SNF and the types of residual water that must be removed. On the other hand, a technique for the actual nondestructive measurement of residual water on canisters containing the SNF right after sealing of the canister could be performed.

6.6.6.1 (Typical test method and data obtained.) See Guide C1553 Annex B for drying parameters particular to the various kinds of SNF (commercial $UO_2$-based, metallic uranium-based, and fuel rubble) covered by this guide.

6.7 *Oxidation Kinetics*—Knowledge of the oxidation kinetics of both LWR and metallic uranium-based spent fuel is important in evaluating the potential need for drying or conditioning treatment (or both) of the spent fuel, the potential for rupturing ("unzipping") of LWR SNF cladding as a result of the oxidation of the spent fuel matrix, and also for supporting the thermal analyses associated with fuel handling and storage safety evaluations. This would include evaluating the potential for pyrophoric reactions in metallic uranium-based SNF. A widely used method for determining the oxidation kinetics of samples of uranium metal-based spent fuel is thermogravimetric analysis. In this method the oxidation rate of the spent fuel sample is determined by measuring the weight gain of the sample continuously as it is exposed to an oxidizing gas atmosphere. The weight gain is recorded along with the sample, enabling data to be collected that can be regressed to an oxidation rate model.

6.7.1 (Typical test method and data obtained.) Thermogravimetric Analyzer (TGA) measures the rate of weight change versus time at a given temperature. It can be used to measure the rate of weight gain attributable to oxidation of the SNF samples.

6.7.2 (Typical test method and data obtained.) Differential Scanning Calorimetry (DSC) can be used to measure the increase in temperature of an SNF sample as a result of the heat given off due to the exothermic reaction of the oxygen with the uranium oxide or uranium metal. The heat of reaction can then be used to calculate the rate of reaction of the SNF with the oxygen gas in the furnace.

6.8 *Ignition Characteristics*—Knowledge of the ignition characteristics of uranium metal-based spent fuel in oxidizing atmospheres is required to support safety analyses for transportation, interim storage, and final repository disposition. This is due to the fact that uranium metal and uranium metal-based spent fuel has exhibited pyrophoric behavior in the past (commercial reactor spent fuel is uranium dioxide-based and has not in the past shown pyrophoric behavior). Three methods widely used to characterize the ignition characteristics of uranium and other metals are Burning Curve Ignition, Isothermal Ignition, and Thermogravimetric Ignition testing. None of these tests provide a direct measurement of the ignition temperature of a particular storage or disposal configuration (size and shape of assembly/rod pieces, total uranium metal in storage container, atmosphere, etc.) of the spent fuel. But each of these tests can provide ignition data that can be combined with the storage/disposal configuration to support code predictions of pyrophoric behavior (see Guide C1454).

6.9 *Dissolution Characteristics*—The purpose of dissolution rate tests would be to support the evaluation of the radionuclide release performance of the SNF as part of the repository performance assessment of the EBS. Exposure of samples of the SNF to the flow of simulated groundwaters of the repository (termed flow-through dissolution testing) and the measurement therein of dissolution/degradation rates would support the generation of a degradation model suitable for inclusion in the evaluation of the EBS. The rates determined from dissolution testing of the SNF should be based on relevant conditions (or potential conditions) and water chemistries. Examples include ambient, undisturbed water chemistry to thermally altered dilute water chemistry to concentration groundwaters expected after contacting EBS components. Data obtained from such dissolution tests (as defined in Practice C1174) can be used to develop models of spent fuel dissolution as functions of water chemistry parameters. As pointed out in Practice C1174, development of a purely mechanistic model may not be possible, and semi-empirical models represent a practical compromise. Thus, mechanistically, temperature is expected to affect dissolution rates according to an Arrhenius relationship. Oxygen concentration is expected to influence the dissolution rate of $UO_2$ because it is known to involve an oxidative dissolution mechanism. Carbonate concentration may affect the dissolution rate because it is known to form soluble complexes with uranium, and so on. These relationships can be represented by the generalized form of the kinetic rate law:

$$Rate = k[A]^a\,[B]^b\,[C]^c\ldots exp(-E_a/RT)$$

where the parameters [A], [B], etc. are test variables such as pH, and carbonate concentration, and where the constant $k$ and the reaction orders, a, b, c, etc. plus the activation energy, $E_a$, are determined by an empirical fit to the data obtained by the characterization tests. This type of equation can be used to calculate expected dissolution rates under potential repository conditions. However, because specific dissolution rates are normalized to surface area, the rate equation must be multiplied by the effective surface area of the waste form being considered when using the data.

6.9.1 (Typical test method—Flow Through Testing) In this test, the specimen size and solution flow rates are adjusted so that the steady-state dissolution rates are independent of the precise flow rate (over a range of flow rates that includes the particular flow rate being used) and are directly proportional to the ratio of surface area to flow volume of the specimen. Required measurements are the solution concentrations of the SNF radionuclides of interest, the test solution flow rate, and the effective surface area of the test specimen. The concentrations of radionuclides within the SNF test specimen must also be known (concentrations are required only for those radionuclides of interest). Test solutions of any desired composition can be used. Characterization Tests, for example, would commonly employ test solutions of various compositions and at various temperatures to provide specific dissolution rates for the given fuel as functions of temperature and water chemistry parameters such as pH and carbonate concentration.

6.9.2 (Typical test method and data obtained—Unsaturated or Drip Tests) Water contact with SNF waste form surfaces can occur with both bare SNF surfaces and with SNF surfaces partially protected by the fuel cladding. Contact can be in several modes: (*1*) continuous contact between the waste form and water vapor and the associated formation of surface water films on the waste form; (*2*) intermittent contact between water and waste form as water periodically flows into and passes through the container; and (*3*) slow ingress and accumulation of water in the container. Each of these modes may result in different processes controlling the rate of reaction and the ultimate release of radionuclides. Unsaturated tests should use water compositions similar to the repository groundwaters. An example test focusing on Yucca Mountain local geology in the United States typically uses the J-13 well near Yucca Mountain, Nevada that is treated by heating in contact with Topopah Spring tuff at 90°C for 3 weeks, termed EJ-13 water. An initial charge of 0.5 mL of EJ-13 is placed in the bottom of each vessel at startup (and after each sampling); thereafter 0.075 mL (about 3 drops) of EJ-13 is dripped onto the test assembly (typically in each 3.5 days). Approximately 0.25 mL of additional air is also injected into the test vessel following the water. The injections are done using a calibrated syringe. The oven temperature is continuously monitored by a data logger, which records the temperature twice per 24 hour period and alarms for deviations greater than ±2°C. The test assemblage used in the tests consists of a sample of the SNF contacted on the top and bottom by two perforated retainer plates made from sensitized 304L stainless steel, which are held in place by two wire posts, also made from 304L stainless steel. The entire test apparatus is enclosed in a 90°C oven except when samples are taken and observations made. Water drips down the sides of the sample. The solution then accumulates at the bottom of the test assemblage, where the glass and sensitized 304L stainless steel contact. Eventually the water drips from the test assemblage to the bottom of the vessel. The unsaturated tests are typically sampled at intervals of not less than 26 weeks for N2 and N3. During sampling, the test assemblage is examined visually to qualitatively ascertain the degree of reaction, including evidence of alteration phase formation and possible spalling. After observation, the test assemblage is transferred to another test vessel, the vessel is rinsed with a measured volume of deionized water and the resulting "vessel rinse" solution removed for analysis, and the just-used vessel is acid-stripped to determine sorbed species.

6.10 *Deliquescence and Advective Flow*—Dissolution of the SNF cladding and fuel matrix may be affected by water that forms on the fuel surfaces due to the precipitation of vapor phase water on certain kinds of deliquescent salts that could adhere to the SNF surfaces. The liquid water formed by deliquescence could also serve as a source for the advective flow of radionuclide-containing liquid water away from the fuel surfaces and into the rest of the EBS. Information on the EBS and WP performance can be found in Practice C1174. Waste Form and EBS materials confirmation testing should provide sufficient data to show, by the standards set by the appropriate governing body, that the components of the EBS are functioning as intended (in the United States by Code of Federal Regulations, Title 10, Part 63, Section 131(a)) and that the waste package monitoring program include laboratory experiments that address the internal conditions of the waste package (in the United States by Code of Federal Regulations, Title 10, Part 63, Section 134 (c)).

## 7. Keywords

7.1 cladding; corrosion; damaged fuel; degraded cladding; engineered barrier system (EBS); failed fuel; geologic repository; interim dry storage; SNF transport; spent nuclear fuel (SNF); waste package performance; waste form degradation; zirconium hydride