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Standard Guide for Corrosion Testing of Aluminum-Based Spent Nuclear Fuel in Support of Repository Disposal[1]

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ε^1 NOTE—Editorial corrections were made throughout in June 2010.

1. Scope

1.1 This guide covers corrosion testing of aluminum-based spent nuclear fuel in support of geologic repository disposal (per the requirements in 10 CFR 60 and 40CFR191). The testing described in this document is designed to provide data for analysis of the chemical stability and radionuclide release behavior of aluminum-based waste forms produced from aluminum-based spent nuclear fuels. The data and analyses from the corrosion testing will support the technical basis for inclusion of aluminum-based spent nuclear fuels in the repository source term. Interim storage and transportation of the spent fuel will precede geologic disposal; therefore, reference is also made to the requirements for interim storage (per 10 CFR 72) and transportation (per 10 CFR 71). The analyses that will be based on the data developed are also necessary to support the safety analyses reports (SARs) and performance assessments (PAs) for disposal systems.

1.2 Spent nuclear fuel that is not reprocessed must be safely managed prior to transportation to, and disposal in, a geologic repository. Placement in an interim storage facility may include direct placement of the irradiated fuel or treatment of the fuel prior to placement, or both. The aluminum-based waste forms may be required to be ready for geologic disposal, or road ready, prior to placement in extended interim storage. Interim storage facilities, in the United States, handle fuel from civilian commercial power reactors, defense nuclear materials production reactors, and research reactors. The research reactors include both foreign and domestic reactors. The aluminum-based fuels in the spent fuel inventory in the United States are primarily from defense reactors and from foreign and domestic research reactors. The aluminum-based spent fuel inventory includes several different fuel forms and levels of ^{235}U enrichment. Highly enriched fuels (^{235}U enrichment levels >20 %) are part of this inventory.

1.3 Knowledge of the corrosion behavior of aluminum-based spent nuclear fuels is required to ensure safety and to support licensing or other approval activities, or both, necessary for disposal in a geologic repository. The response of the aluminum-based spent nuclear fuel waste form(s) to disposal environments must be established for configuration-safety analyses, criticality analyses, PAs, and other analyses required to assess storage, treatment, transportation, and disposal of spent nuclear fuels. This is particularly important for the highly enriched, aluminum-based spent nuclear fuels. The test protocols described in this guide are designed to establish material response under the repository-relevant conditions.

1.4 The majority of the aluminum-based spent nuclear fuels are aluminum clad, aluminum-uranium alloys. The aluminum-uranium alloy typically consists of uranium aluminide particles dispersed in an aluminum matrix. Other aluminum-based fuels include dispersions of uranium oxide, uranium silicide, or uranium carbide particles in an aluminum matrix. These particles, including the aluminides, are generally cathodic to the aluminum matrix. Selective leaching of the aluminum in the exposure environment may provide a mechanism for redistribution and relocation of the uranium-rich particles. Particle redistribution tendencies will depend on the nature of the aluminum corrosion processes and the size, shape, distribution and relative reactivity of the uranium-rich particles. Interpretation of test data will require an understanding of the material behavior. This understanding will enable evaluation of the design and configuration of the waste package to ensure that unfilled regions in the waste package do not provide sites for the relocation of the uranium-rich particles into nuclear critical configurations. Test samples must be evaluated, prior to testing, to ensure that the size and shape of the uranium-rich particles in the test samples are representative of the particles in the waste form being evaluated.

1.5 The use of the data obtained by the testing described in this guide will be optimized to the extent the samples mimic the condition of the waste form during actual repository exposure. The use of Practice C1174 is recommended for guidance. The selection of test samples, which may be unaged or artificially aged, should ensure that the test samples and

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conditions bound the waste form/repository conditions. The test procedures should carefully describe any artificial aging treatment used in the test program and explain why that treatment was selected.

2. Referenced Documents

2.1 *ASTM Standards:*[2]

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

2.2 *Government Documents*

10 CFR 60 US Code of Federal Regulations Title 10, Part 60, Disposal of High Level Radioactive Wastes in Geologic Repositories

10 CFR 71 US Code of Federal Regulations Title 10, Part 71, Packaging and Transport of Radioactive Materials

10 CFR 72 US Code of Federal Regulations Title 10, Part 72, Licensing Requirements for the Independent Storage of Spent Nuclear and High-Level Radioactive Waste

3. Terminology

3.1 *Definitions:*

3.1.1 Terms used in this guide are defined in Practice C1174, by common usage, by Webster's New World Dictionary, or as described in 3.2, or combination thereof.

3.2 *Definitions:*

3.2.1 *aluminum-based spent nuclear fuel*—irradiated nuclear fuel or target elements or assemblies, or both, that are clad in aluminum or aluminum-rich alloys. The microstructures contain a continuous aluminum-rich matrix with uranium-rich particles dispersed in this matrix.

3.2.2 *aluminum-based spent nuclear fuel form or waste form*—any metallic form produced from aluminum-based spent nuclear fuel and having a microstructure containing a continuous aluminum-rich matrix with uranium-rich particles dispersed in this matrix. This term may include the fuel itself.

3.2.3 *artificial aging*—any short time treatment that is designed to duplicate or simulate the material/property changes that normally occur after prolonged exposure and radioactive decay.

3.2.4 *attribute test*—a test conducted to provide material properties that are required as input to behavior models, but that are not themselves responses to the environment.

3.2.5 *bounding*—a test, sample condition or calculation designed to provide an evaluation of the limits to material behavior under relevant conditions.

3.2.6 *characterization test*—in high-level radioactive waste management, any test conducted principally to furnish information for a mechanistic understanding of alteration.

3.2.7 *corrosion product*—an ion or compound formed during the interaction of the aluminum-based spent nuclear fuel with its storage or disposal environment. The corrosion product may be the result of aqueous corrosion, oxidation, reaction with moist air, or other types of chemical interaction.

3.2.8 *interim storage installation*—a facility designed to store spent nuclear fuels for an extended period of time that meets the intent of the requirements of an independent spent fuel storage installation or a monitored retrievable storage facility, as described in 10 CFR 72.

3.2.9 *melt-dilute process*—a process to lower the fraction of ^{235}U in highly enriched, aluminum-based spent nuclear fuel by melting and adding depleted uranium to the waste from.

3.2.10 *performance assessment*—an analysis that identifies the processes and events that might affect a disposal system, examines the effects of those 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.

3.2.11 *safety analysis*—an analysis to determine the risk to the public health and safety associated with the storage, treatment, transportation, or disposal, or combination thereof, of aluminum-based spent nuclear fuel.

3.2.12 *service condition test*—a test of a material conducted under conditions in which the values of the independent variables characterizing the service environment are in the range expected in actual service.

4. Significance and Use

4.1 Disposition of aluminum-based spent nuclear fuel will involve:

4.1.1 Removal from the existing storage or transfer facility,

4.1.2 Characterization or treatment, or both, of the fuel or the resulting waste form, or both,

4.1.3 Placement of the waste form in a canister,

4.1.4 Placement of the canister in a safe and environmentally sound interim storage facility,

4.1.5 Removal from the interim storage facility and transport to the repository,

4.1.6 placement in a waste container,

4.1.7 Emplacement in the repository, and

4.1.8 Repository closure and geologic disposal. Actions in each of these steps may significantly impact the success of any subsequent step.

4.2 Aluminum-based spent nuclear fuel and the aluminum-based waste forms display physical and chemical characteristics that differ significantly from the characteristics of commercial nuclear fuels and from high level radioactive waste glasses. For example, some are highly enriched and most have heterogeneous microstructures that include very small, uranium-rich particles. The impact of this difference on repository performance must be evaluated and understood.

4.3 The U.S. Nuclear Regulatory Commission has licensing authority over public domain transportation and repository disposal (and most of the interim dry storage) of spent nuclear fuels and high-level radioactive waste under the requirements established by 10 CFR 60, 10 CFR 71, and 10 CFR 72. These requirements outline specific information needs that should be

[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.

met through test protocols, for example, those mentioned in this guide. The information developed from the tests described in this guide is not meant to be comprehensive. However, the tests discussed here will provide corrosion property data to support the following information needs.

4.3.1 A knowledge of the solubility, leaching, oxidation/reduction reactions, and corrosion of the waste form constituents in/by the repository environment (dry air, moist air, and repository relevant water) (see 10 CFR 60.112 and 135).

4.3.2 A knowledge of the effects of radiolysis and temperature on the oxidation, corrosion, and leaching behavior (see 10 CFR 60.135).

4.3.3 A knowledge of the temperature dependence of the solubility of waste form constituents plus oxidation and corrosion products (see 10 CFR 60.135).

4.3.4 Information from laboratory experiments or technical analyses, or both, about time dependence of the internal condition of the waste package (see 10 CFR 60.143 and 10 CFR 72.76).

4.3.5 Laboratory demonstrations of the effects of the electrochemical differences between the aluminum-based waste form and the candidate packaging materials on galvanic corrosion (see 10 CFR 71.43) or the significance of electrical contact between the waste form and the packaging materials on items outlined in 4.3.1-4.3.4 (see 10 CFR 60.135), or both.

4.3.6 Information on the risk involved in the receipt, handling, packaging, storage, and retrieval of the waste forms (see 10 CFR 72.3).

4.3.7 Information on the physical and chemical condition of the waste form upon repository placement so that items outlined in 4.3.1-4.3.4 can be evaluated (see 10 CFR 60.135).

4.3.8 Knowledge of the degradation of the waste form during interim storage so that operational safety problems with respect to its removal from storage can be assessed, if such removal is necessary (see 10 CFR 72.123).

4.3.9 Knowledge of the condition of the waste form prior to repository placement so that items outlined in 4.3.1-4.3.4 are properly addressed (see 10 CFR 60.135).

4.4 Conditions expected during each stage of the disposition process must be addressed. Exposure conditions anticipated over the interim storage through geologic disposition periods include dry and moist air, and aqueous environments. The air environments are associated with interim storage and the early stages of repository storage while the aqueous environments arise after water intrusion into the repository has caused corrosion-induced failure of the waste package.

5. Information Relevant to Geologic Disposal

5.1 Tests of the aluminum-based waste forms should, along with applicable and qualified data from the literature, or both, provide data pertinent to (*a*) corrosion or oxidation of waste form constituents in vapor environments, or both, (*b*) corrosion or oxidation of waste form constituents in liquid environments, or both, (*c*) dissolution of waste form constituents, (*d*) oxidation products or corrosion products, or both and (*e*) selective leaching of waste form constituents. Selected tests should provide data concerning:

5.1.1 The effective release rates (as solute or colloidal species) and dissolution rates of waste form constituents and corrosion products in water compositions relevant to repository disposal,

5.1.2 The temperature dependence of, and the effect of radiolysis products on, waste form constituent solubility in repository relevant water compositions,

5.1.3 The corrosion rate or relative corrosion rates of the various constituents in the waste form, or both,

5.1.4 An understanding of the effect of waste form microstructure (the size, shape, distribution, and volume fraction of the uranium-rich particles, for example), corrosion products, and their formation sequence on corrosion and oxidation behavior, and

5.1.5 An understanding of the release of uranium-rich colloids or particles, or both, during storage and disposition.

5.2 Tests conducted to supply the data needs described in 5.1 would ideally provide sufficient information to help establish mechanistic models, or, in any case, empirical correlations, for:

5.2.1 Corrosion rates under the bounding or potential range of repository conditions,

5.2.2 The effective solubility of waste form constituents, including corrosion products, as a function of the temperature and chemistry of the water that may surround the waste form after a canister breach in the repository, and

5.2.3 The selective leaching of the aluminum matrix from the uranium-rich particles with anticipated waste package/repository environments.

5.3 The information needs described in 5.1 and correlations/models described in 5.2 should enable the calculation of the rate of release of radionuclides from the aluminum-based waste forms stored in the repository.

5.4 The information needs described in 5.1 should provide the necessary particle size and leach rate information for an analyst to model the potential for a criticality resulting from the redistribution of uranium-rich particles.

6. Relationship of Aluminum-Based Waste Forms to Other Waste Forms

6.1 The aluminum-based waste forms differ from commercial spent fuels and high level waste glasses in several respects, including homogeneity, reactivity, and the tendency toward galvanic corrosion.

6.2 The position of aluminum in the electromotive series is such that, among the common structural metals, only magnesium and beryllium are more reactive. The corrosion resistance of aluminum and aluminum-based alloys is largely due to the presence of a protective oxide film. Any understanding of aluminum corrosion necessarily incorporates an understanding of oxide film behavior.

6.3 The nature and characteristics of the oxide film will evolve during repository exposure, and the effects of this evolution on subsequent corrosion processes must be considered in the evaluation of waste form behavior, particularly with regard to the tendency for release of microscopic, uranium-rich particles by selective leaching.

6.4 The core of the aluminum-based spent fuels is a two-phase dispersion of uranium-rich particles in a matrix of relatively pure aluminum. Virtually all the uranium in the fuel will be contained in the microscopic, uranium-rich particles. The presence of microscopic particles contrasts the relatively homogeneous nature of the microstructure of commercial nuclear fuels and high-level waste glasses.

6.5 The aluminum matrix is more chemically reactive than the typical particle; thus, the particles may be cathodic to the aluminum matrix. Corrosion of the particle matrix composite may result in selective leaching of the aluminum, thus releasing the particles to the surrounding environment. The tendency for selective leaching may depend on the characteristics of any surface films that are present on the aluminum-based waste form, on the water chemistry, and on the extent of galvanic coupling of the waste form to the waste package or to over pack materials, or both.

6.6 Waste form treatments, such as a melt-dilute process, may alter the typical morphology of the uranium-rich particles and the nature of the surface films and may even remove a substantial fraction of the fission products. However, the treated, aluminum-based waste forms will remain chemically active relative to commercial spent fuels, high level waste glasses, and structural materials for the waste canister package. This difference in chemical activity or corrosion potential provides an additional driving force for galvanic corrosion. Selective leaching of the aluminum matrix could also be promoted by electrical contact with any of the less active materials contained in the waste package.

7. Test Objectives and Requirements

7.1 This guide addresses the information necessary to establish the corrosion behavior of aluminum-based waste forms in repository relevant environments. Such information is required to support the geologic repository disposal of the aluminum-based waste forms. The tests required to develop the necessary information include:

7.1.1 Characterization tests to determine the susceptibility of the waste form to selective leaching and to characterize the resulting release of any microscopic, uranium-rich particles to the repository environment. Such tests could include electrochemical impedance tests, autoclave corrosion tests, flow-through corrosion tests, drip corrosion tests, and numerous other corrosion tests.

7.1.2 Service condition or accelerated tests, or both, to establish the forward dissolution rates of the various microstructural constituents in a waste form exposed to repository-relevant ground water. Such tests could include flow-through tests, autoclave tests, and anodic polarization tests.

7.1.3 Characterization or accelerated tests, or both, to establish the effects of galvanic coupling on corrosion rates, corrosion products, and the release of the radionuclides to the repository environment. Such tests could include anodic polarization tests, electrochemical impedance tests, and autoclave corrosion tests.

7.1.4 Attribute tests provide information for the design and interpretation of characterization tests. The characterization tests establish the effects of pre-exposure waste form condition (nature and characteristics of surface films or deposited radionuclides, or both) on corrosion rates, corrosion products, and the release of radionuclides to the repository environment. The attribute tests could include optical metallography, SEM, Auger analysis, and microprobe analysis

7.2 The tests summarized in 7.1.1-7.1.4 should include provisions to measure the effect of temperature on the corrosion processes and release of radionuclides.

7.3 The tests summarized in 7.1.1-7.1.4 should include provisions to establish the effects of ground water chemistry on the corrosion processes and the release of radionuclides.

8. Selection of Test Samples

8.1 The test samples should be carefully identified/selected based on the intended use of the test data. Examples of selection criteria could include: bounding or typical characteristics such as macrostructure and microstructure, damaged versus undamaged fuel, level of enrichment or dilution, level of burn-up, availability, typical size of uranium rich particles, and particle-volume to matrix-volume ratio.

8.2 The position of the aluminum-based waste form material in the electromotive series will virtually ensure that electrical contact with waste package materials or waste forms codisposed with the aluminum-based waste form, or both, will cause the aluminum-based waste form to act as a sacrificial anode. Test samples should be selected to reflect this potential for galvanic corrosion.

8.3 Fuel claddings, if present on the waste form being evaluated, should be removed from at least one surface of the test samples prior to testing.

8.4 Sample configuration should be consistent with the requirements of the test method and the use of non-standard samples should be avoided.

9. Test Methods and Constraints

9.1 Test methods used to establish the chemical stability and rate of radionuclide release from the waste form may include, but are not limited to:

9.1.1 Standard electrochemical test methods, such as controlled potential techniques, controlled current techniques, and electrochemical impedance spectrometry,

9.1.2 Coupon testing in controlled static or dynamic environments, or both, such as provided by a drip, vapor hydration or flow tests, under both repository-relevant and accelerated conditions,

9.1.3 Coupon testing in the environments selected to simulate those anticipated for interim storage, and

9.1.4 Confirmatory field testing in repository relevant environments or at repository facilities, such as those at the Yucca Mountain Project, or both.

9.2 The test method, or combination of methods selected, should provide the data necessary to assess the oxidation-dissolution behavior of the waste form or various phases present in the waste form, or both.

9.2.1 The test method should provide measurements of the relevant electrolyte composition prior to contact with the waste form, if practical; during contact with the waste form, if practical; and after contact with the waste form, if a flow-through environment is used. These measurements of electrolyte composition should be compared to repository relevant environments to ensure that the proper, and potentially bounding, environments have been selected and used.

9.2.2 The suite of tests (or test plan) should also include provisions:

9.2.2.1 Measure temperatures in the test environment(s),

9.2.2.2 Determine the presence, or absence, of alteration phases. If such phases are present, the composition and structure of the phases should be assessed,

9.2.2.3 Determine release of particles and isolate (filter, for example) and characterize particles released from the waste form by the oxidation-dissolution process,

9.2.2.4 Determine the chemical stability of waste form during exposure to water vapor, and

9.2.2.5 Characterize the radionuclide and fissile species distribution in the sample.

10. Use of Test Data

10.1 The data from the corrosion tests program should support the development of material performance models for aluminum-based spent nuclear fuel waste forms in the repository environment(s) and the use of those models to assess the performance of aluminum-base waste forms in the repository. The models will ideally be as mechanistically based as practical, and the tests should, to the extent possible, support mechanistic interpretation of the material behavior. However, in many cases, only empirical interpretations of the data or empirically-based performance models may be practical.

10.2 The waste form performance is determined by the rate of release of radionuclides to the repository environment and by the changes in mineralogy or physical coherence of the waste form. Thus, the test data should provide the parameters necessary to calculate the time and temperature dependence of radionuclide release and exposure-induced alterations in the waste form.

10.3 The test data should provide information on waste form stability and radionuclide release rates that can be used to compare and contrast the anticipated repository behavior of waste forms from aluminum-based spent nuclear fuels with other waste forms, such as glass and spent commercial nuclear fuels.

10.4 The test data should provide information that can be used to understand, and model, the fundamental modes of waste form degradation.

10.5 The test data should provide information that can be used to test, and validate, models of waste form behavior in anticipated repository environments.

10.6 The test data should provide information that can be used to model the release, transport, and redistribution of uranium-rich particles for inclusion in the criticality analyses.