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Section 1 Contents

1. Introduction

1.1 Overview

This report focuses on radioactive waste form characteristics that will be used to design a waste package and an engineered barrier system (EBS) for a suitable repository as part of the Yucca Mountain Project. The term waste form refers to irradiated reactor fuel, other high-level waste (HLW) in various physical forms, and other radioactive materials (other than HLW) which are received for emplacement in a geologic repository. Any encapsulating or stabilizing matrix is also referred to as a waste form.

This report is divided into three chapters. The first chapter outlines report organization, the second chapter covers properties and data which a design team would use to analyze the mechanical handling, thermal, structural, chemical, and nuclear responses of existing and future waste forms, and the third chapter provides a description of preliminary models which are useful for planning experimental testing and performance assessment activities.

The information in Chapter 1 includes a brief discussion of:

- Design goals.
- Regulatory requirements.
- Interpretation of design goals and regulatory requirements.

Disposal costs for irradiated fuel and other radioactive nuclear wastes are an integral part of energy and national security costs. To proceed with safe disposal, complete characterization information about these wastes must be uniformly and readily available to a variety of different design sub-teams so that their products will interface and integrate consistently into a total disposal system.

In order to quantify preliminary design decisions, an accumulation of waste form characteristic data is required. Chapter 2 contains waste form characteristics along with available analyses for preliminary design.

Chapter 3 expands on the physical, material, structural, chemical, and radiological analytical responses of the waste forms. This subject includes other topics, such as phases, chemical combinations, transport modes, and concentrations. At the present time models for many of the analytical responses of waste forms are still being developed.

1.2 Technical Objectives

When Congress passed the Nuclear Waste Policy Act (NWPA) of 1982 (Public Law 97-425), it began the process to establish a national repository for the permanent disposal of spent fuel and high-level waste. This Act gave the U.S. Department of Energy (DOE) the responsibility for siting, constructing, and operating a repository. It gave the U.S. Environmental Protection Agency the responsibility for developing standards to protect the environment from offsite releases of radioactive material from a repository (40CFR191). The U.S. Nuclear Regulatory Commission (NRC) was responsible for announcing the technical requirements necessary to license all phases of repository operation (10CFR60). In 1987, Congress amended the Act to focus site characterization efforts on a site at Yucca Mountain in Nevada.

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The technical objective of the waste package program is to develop a waste package and an associated EBS, and to demonstrate in an NRC licensing proceeding that the package and system meet all the regulations. NRC rule 10CFR60.113 mandates two specific performance objectives for the waste package and EBS after the repository closes and divides the period after closure into two time periods, referred to as "containment" and "controlled-release." The containment requirement applies primarily to the waste packages, and the controlled-release requirement applies primarily to the EBS:

Containment [10CFR60.113 (a) (1) (ii) (A)]

> . . . the engineered barrier system shall be designed, assuming anticipated processes and events, so that: Containment of HLW within the waste packages will be substantially complete for a period to be determined by the Commission taking into account the factors specified in 60.113(b) provided, that such period shall be not less than 300 years nor more than 1,000 years after the permanent closure of the repository.

Controlled Release [10CFR60.113 (a) (1) (ii) (B)]

> . . . the engineered barrier system shall be designed, assuming anticipated processes and events, so that: . . . The release rate of any radionuclide from the engineered barrier system following the containment period shall not exceed one part in 100,000 per year of the inventory of that radionuclide calculated to be present at 1,000 years following permanent closure, or such other fraction of the inventory as may be approved or specified by the Commission; provided that this requirement does not apply to any radionuclide which is released at a rate of less than 0.1% of the calculated total release rate limit. The calculated total release rate limit shall be taken to be one part in 100,000 per year of the inventory of radioactive waste, originally emplaced in the underground facility, that remains after 1,000 years of radioactive decay.

The requirements relating to postclosure performance of the total repository system [10CFR60.112] place additional requirements on the design and performance of the waste package and EBS as follows:

> The geologic setting and the engineered barrier system and the shafts, boreholes and their seals shall be designed to assure that releases of radioactive materials to the accessible environment following permanent closure conform to such generally applicable standards for radioactivity as may have been established by the Environmental Protection Agency with respect to both anticipated processes and events and unanticipated processes and events.

A fourth major objective is to perform a "comparative evaluation of alternatives to the major design features that are important to waste isolation, with particular attention to the alternatives that would provide longer radionuclide containment and isolation" [10CFR60.21 (c) (1) (ii) (D)].

A number of other requirements apply to the waste package and EBS before the permanent closure of the repository. These requirements include radiological protection [l0CFR 60.111 (a)], retrievability [10CFR60-111 (b)], and geologic repository operations area design criteria [10CFR60.131].

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Finally, 10CFR60.135 sets forth specific design criteria for the waste package and its components that must be met. These criteria include constraints on the general performance of the package, its chemical reactivity, and provisions for its handling and labeling, as well as design criteria for the waste forms.

1.3 Quality Objectives

All information for the final design, design analysis, testing, and performance assessment of the waste package and EBS that will form a basis of the license application will be acquired or developed under an NQA-l quality assurance program based on the criteria of Appendix B of 10CFR50. All participants in the project have developed or adopted quality assurance program plans (QAPPs) that reflect all requirements of the Project Office Quality Assurance Plan, which incorporates the provisions of the Office of Civilian Radioactive Waste Management (OCRWM) Quality Assurance Requirements (QARs). For waste package and EBS work, a system of quality procedures (QPs) are used to implement the QAPP. A software quality assurance plan (SQAP), which specifically addresses implementing the QAPP requirements to computer software, supplements the QAPP and QPs.

QPs establish methods to control scientific investigations, testing activities, design activities, and performance assessments that are described in the technical planning sections of the *OCRWM Yucca Mountain Project Waste Package Plan.* For example, the QPs describe how scientific investigations and design analyses are planned, controlled, and documented. They also describe which documents are quality assurance records and how these records are created, maintained, and stored. They also cover how documents are reviewed and how the document content is verified.

In the case of the present documentation of preliminary waste form characteristics, the data and analytic response models provided were taken primarily from the open literature. These data and models are considered as best available and their quality and suitability cannot be assured for waste disposal design purposes except as noted in this report.

1.4 Types of Waste Forms

Waste forms in this report are divided into three categories: spent fuel, glass, and other waste forms. This division is selected because each category may have different design constraints and may require distinct solutions for the EBS.

1.5 Spent-Fuel Waste Forms

In this report, spent fuel is understood to mean elements from the entire inventory of existing and future fuel assemblies from nuclear reactors. When spent-fuel properties were compiled, it was assumed that issues central to waste package design must be considered, beginning when fuel is discharged from the reactor and ending after the “controlled release” time period.

Spent reactor fuels originate mainly from civilian nuclear power reactors. The vast majority of these fuels are from light-water reactors (LWRs), which are either boiling-water reactors (BWRs) or pressurized-water reactors (PWRs).

Many varied issues and operations are involved in waste package design, and the importance of different spent-fuel properties depends greatly on the particular aspect of design under consideration. Some properties, such as physical dimensions and masses, are

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well defined. The distribution of such properties must be considered in any arbitrary design intended to transport, handle, and to maintain containment of intact fuel assemblies or a combination of consolidated fuel pins and segregated assembly hardware. Given the relative range of assembly and fuel designs that are accommodated in existing power reactors, we must extrapolate properties with reasonable confidence from the present inventory to the total expected inventory that will be contained in the repository.

Most, if not all, of the information that must be available for the containment and EBS designs for the spent reactor fuels must be available for the other categories of waste forms. Therefore, we will discuss the information once and will present the available data for each waste form.

1.6 Physical Inventory

The spent-fuel properties needed are included schematically in Figure 1-1 and are given in detail in Table 1-1. We assume that the entire inventory is comprised solely of LWR, BWR and PWR fuel assemblies. We also assume that the characteristics of the assemblies are available at their arrival at the repository. After arrival, it is likely that the assemblies will be placed into temporary storage and, possibly, subjected to tests to verify external physical dimensions, to determine their contribution to reactivity of an array, their thermal power, etc. If a design accommodates intact fuel assemblies, they can then be transferred directly to a container. If a design accommodates consolidated fuel pins and associated assembly hardware, additional handling is needed, and additional measurements may be required. For this purpose, the term container design is broadly interpreted to include the actual design of the container, its cover, means for handling individual spent-fuel assemblies, and, if necessary, means for removal of fuel pins from the assemblies.

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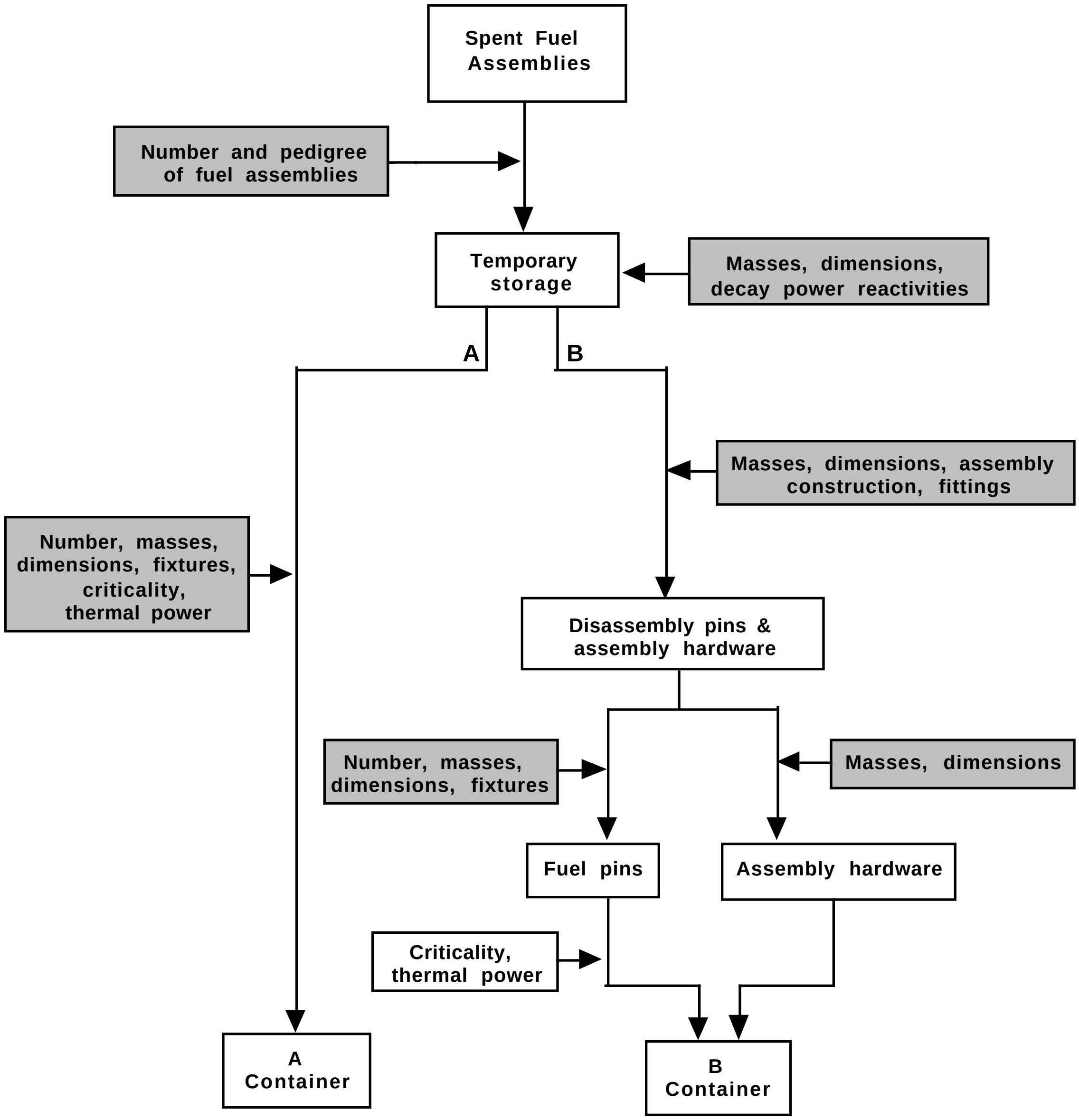

Figure 1-1 **The effect of spent-fuel properties on container design: mechanical, handling, thermal, criticality, and shielding**

Properties of assemblies in the present pool of spent fuel are generally contained in the *Integrated Data Base for 1990,* DOE/RW-0006, Rev. 6, and in the Characteristics Data Base of ORNL. These include masses and physical dimensions, materials of construction, physical characteristics of fuel pins, etc. Assembly drawings of varying degree of complexity are also given in the latter. The data sets appear to be adequate for the majority of PWR fuel assemblies but are very sparse for BWR assemblies. Many of the details that are relevant to handling and disassembly of fuel manufactured by the General Electric Company are presently unavailable. (Characteristics of these are only inferred by reference to those of other manufacturers.) Some additional effort may be needed to ensure sufficiency and proprietary aspects of information to meet the needs of handling and the preliminary stages of design of containers and other facilities.

Table 1-1 Spent-fuel physical characteristics

	Number and physical dimensions	Handling hardware	Assembly drawings	Special properties	As-fabricated fuel characteristics	As-irradiated fuel characteristics
Fuel assemblies	Total width, length, and mass	Design of end plates for locating assembly and handling	Dimensions of end plates, spacers, and other hardware; fastener characteristics	Failed pins, control rods, etc.		
Fuel pins	Total and active lengths, location of active length, O.D., total masses	Design of end plugs for locating pin		Failed pins, control rods, etc.	Total fuel mass, enrichment, pressurization dimensions of fuel pellets	Estimates of changes in pressurization and physical dimensions from as- fabricated conditions

For waste forms such as glass, the physical inventory can be described simply in terms of container dimensions and mass, and the total amount of each waste form. Some forms, e.g., spent-fuel hardware, may be difficult to characterize. However, these forms will probably be consolidated before being placed in the repository, and it may be necessary to do research to determine the compacted density of these materials. For these forms, the most important factors are the waste form mass density and the physical description.

1.7 Radionuclides

The inventory of radionuclides in any waste form is important for several reasons. It determines the amount of heat generated per unit mass of the waste form, and it determines the background radiation created by the waste form, in terms of intensities, energies, and kinds of radiation.

The potential release of radionuclides from a spent-fuel element into the immediate surroundings of the element depends on a large number of factors, many of which are interrelated. The irradiation history of the fuel, measured by the burnup, together with the initial, as-built inventory of elements in the fuel, determine what the inventory will be when the fuel is discharged from the reactor and any time thereafter. The same factors also can be useful in estimating the amount of physical damage that has been found in the fuel pellets and in the fuel cladding. The operating temperatures and the chemistry of the reactor coolant water are also dependent factors in this estimating process.

A spent-fuel assembly may have failed fuel pins. Presently, a detailed characterization of failed pins has not been completed. In any case, the dissolution and transport of radionuclides out of the fuel pin requires a breach in the fuel cladding.

For spent fuels, the mixture and amount of radionuclides within a pellet enclosed inside the cladding depends primarily on assembly burnup, which translates into the number of fission events per cubic centimeter of the pellet. At the pellet dimensional scale, there is some slight variation in the number of fission events in the radial direction across a pellet. This is termed the pellet rim effect, but it is not presently considered a significant variation for spent

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fuels with burnups less than 60 gigawatt days per metric ton of uranium (Gwd/MTU). At the assembly dimensional scale, the burnup rate can vary spatially in both the radial and axial directions within the core volume of an operating nuclear reactor; thus, there are burnup variations across an assembly and axially along an assembly. For preliminary/conceptual design purposes, the variations within the set of fuel rods of an assembly are not considered significant. The concentrations of gaseous nuclides and the potentially volatile nuclides within a fuel pellet depend primarily on the fission gas released (FGR) during reaction operation. At present little detailed information is available on the FGR spent-fuel attribute.

For the HLW forms, which are a mixture of by-products from spent-fuel reprocessing plants, the radionuclide content will be measured by nuclide and radioactivity per unit volume. For the most part, the HLW have three phases, liquid, sludge, and salt cake. A stream of these three phases will be mixed and incorporated into a glass waste form. It is during the production of the glass waste form that the radionuclide content will be measured and recorded in the data base.

Fortunately, the radionuclide inventory is readily calculable or measurable. Computer codes are available which calculate the fuel inventory, and the industry generally uses these codes for fuel management. We must assume that measurements will be used to determine radionuclide inventories in other waste forms.

1.8 Decay Heat and Criticality

The nuclide inventory, types of isotopes, and their amounts determine how the waste may be stored in the individual waste package and in the repository as a whole. The properties we must know are shown in Figure 1-1 and are also listed in Table 1-2.

Table 1-2 Spent-fuel characteristics associated with thermal, criticality, and shielding subtask

	History	Composition and thermal properties	Physical dimensions	Detailed geometry	As-fabricated fuel composition
Assembly hardware					
Thermal effects	Irradiation and post-irradiation stroage and conditions	Construction materials and their thermal conductivities after irradiation	Total width, length, and mass	Distribution of materials in space, detailed geometry of fuel-pin placement	
Criticality/ shielding effects	Irradiation and post-irradiation stroage and conditions; burnup	Construction materials and their neutron interaction characteristics	Total width, length, and mass	Distribution of materials in space, detailed geometry of fuel-pin placement	

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	History	Composition and thermal properties	Physical dimensions	Detailed geometry	As-fabricated fuel composition
Fuel pins					
Thermal effects	Irradiation (burnup) and post-irradiation storage and conditions	Cladding material and irradiated fuel and their thermal conductivities	Total length, outer diameter, cladding thickness, fuel massq	Distribution of materials in space, including active dimensions of fuel pins	
Criticality/ shielding effects	Irradiation (burnup) and post-irradiation storage and conditions	Construction materials and their neutron interaction characteristics	Total width, outer diameter, cladding thickness, fuel mass	Distribution of materials in space, including active dimensions of fuel pins	Enrichment and components added or deposited onto the fuel for reactivity control during reactor operation

It is assumed that guidelines or standards are established which define, for any EBS design, temperature limits and their spatial distributions, and the maximum effective neutron multiplication factor that can be achieved by any given container loading or geometry. It is also assumed that an approved methodology exists for determination of the isotopic and elemental composition of the spent fuel and the types and spectra of ionizing radiation emitted by fuel pins and assembly hardware as a function time and irradiation history. (The latter define the source terms for thermal and shielding calculations and the content of fissile and fertile material needed for criticality calculations.)

Reactivity and thermal properties of the assemblies and fuel pins are needed both for meeting temperature and reactivity limitations during temporary and long-term storage. These parameters essentially define the number and arrangement of fuel assemblies or consolidated fuel pins plus separated assembly hardware that can be accommodated in any single container and meet design requirements for the repository as a whole.

1.9 Radiation Field

The radiation field surrounding a given amount of any waste form is a determining factor in how the waste form and its container must be handled. The amount of shielding necessary during handling, storage, emplacement, and disposal is based on the radiation field and repository operational and performance requirements.

The important spent-fuel properties here are all those that will ultimately impact the rates and quantities of radionuclides which can be released and transported from failed fuel pins to the container and beyond. These properties also cover external radioactive deposits which may be important during the handling of the spent fuel prior to loading into the waste containers. These properties are shown schematically in Figure 1-2 and listed in detail under subtopic Spent Fuel Characteristics Associated with Radioactivity Release and Radiation

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Field. We have assumed that an approved methodology exists to obtain the detailed elemental and isotopic composition of the fuel and cladding, as well as the spectrum and types of radiations emitted from fuel pins and assembly materials.

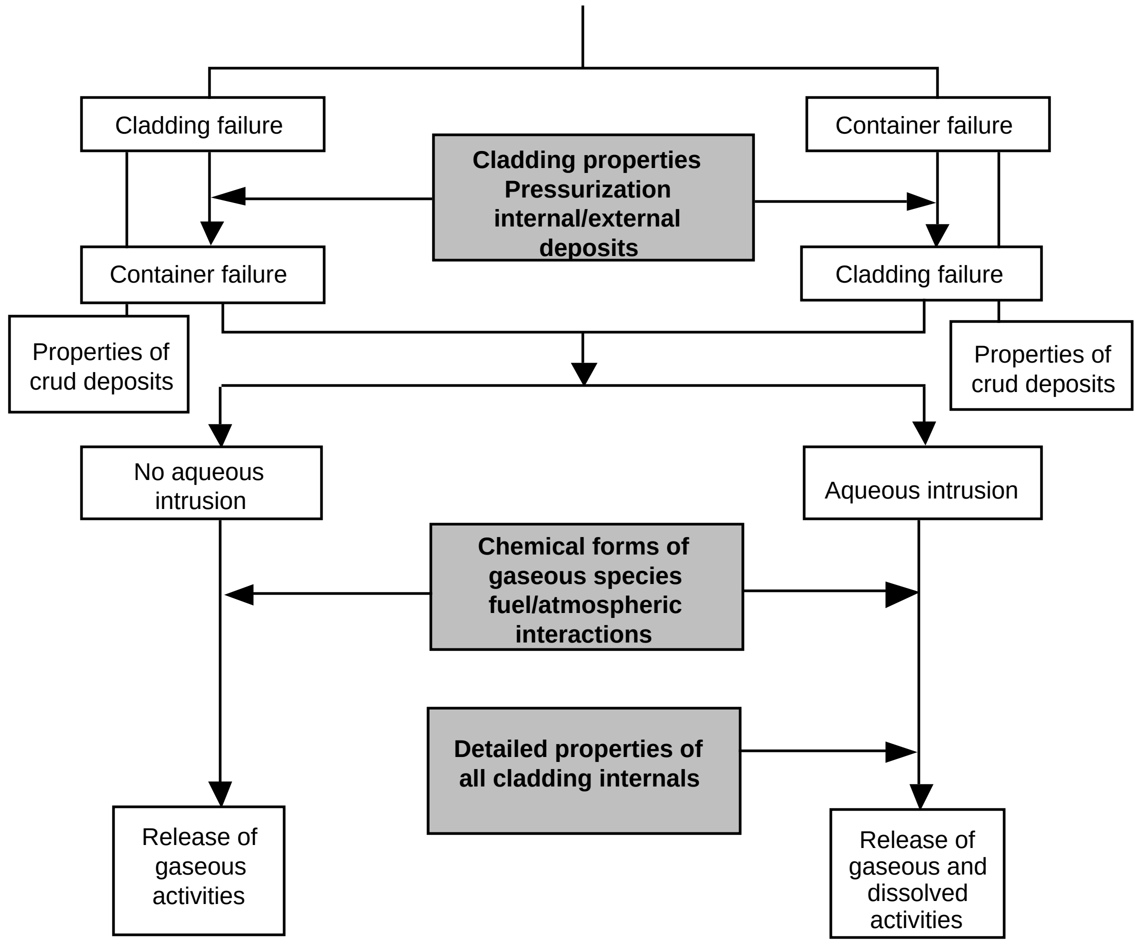

Figure 1-2 Radioactivity release

We have already discussed the importance of all of the properties of irradiated fuel and its cladding. However, external crud deposits on the assembly hardware and cladding are worthy of a few comments. Crud deposits typically contain radioactivity (e.g., ^{60}Co from neutron activation) with short half-lives compared to the time periods of interest in waste disposal. However, the length of these half-lives must be considered during handling. Their physical and chemical properties and the extent and content of radioactivities will be considered in design of the intermediate storage facilities, for assessing the extent of contamination possible throughout handling procedures, etc.

The majority of spent-fuel properties necessary for characterization of the radiation field are provided through the studies underway at the Materials Characterization Center (MCC) of the Pacific Northwest Laboratories (PNL). The principal information deficiencies are:

- Modern, high-burnup fuel from PWRs and BWRs that span the range of spent-fuel properties likely to be found in the inventory of a repository are not yet included within the approved testing materials (ATM) presently analyzed or on hand for measurement.

- Knowledge of the distribution of fission gas release to a reasonable degree of accuracy is not now available nor is it clear that a means has been provided to assess accurately the nature of this distribution for the larger fission gas releases. An activity is underway at MCC to elicit information from fuel vendors. However, it is not certain that this information will lead to sufficient confidence in the distribution function in the high burnup region to mitigate the need for further experimental measurements.

Spent-Fuel Characteristics Associated with Radioactivity Release and Radiation Field

Fuel assemblies have the following properties:

- As-fabricated properties
  - Materials composition: materials of construction, masses, and chemical compositions, including major and minor constituents.
- As-irradiated properties
  - History: irradiation and post-irradiation storage.

Fuel pins have the following properties:

- As-fabricated properties
  - Physical dimensions and masses: total length, outer diameter, cladding thickness, as-fabricated fuel-pellet dimensions, plenum dimensions or total void volume of fuel pin, and fuel and cladding masses.
  - Actual compositions of fuel and cladding: includes cladding type and any liners added to enhance cladding performance during in-core service, fuel enrichment, and the composition and location of any components added or deposited onto the fuel for the purposes of reactivity control during in-core operation.
  - Fill gas composition and pressure.
  - Fuel morphology: average grain size, porosity.
- As-irradiated properties
  - History: irradiation (burnup) and post-irradiation storage.
    - Identification of failed fuel pins as delivered.
    - Cladding composition and morphology: including external crud deposits, thickness of external and internal oxide layers, hydride content, deposits of fission products and other fuel components.
    - Composition and pressures of gases, including fission gases and helium.
    - Fuel morphology and composition, including surface deposits; extent, properties, and composition of periphery or rim region; grain sizes and their radial distributions; fuel phases and their radial distributions; characteristics of grain boundaries, including nature and extent of materials segregated along grain boundaries; fuel-fragment size distributions; and estimates of total surface area per unit fuel mass.

1.10 Hardware

In discussing waste forms, the term hardware refers to the material contained in a fuel assembly, with the exception of the fuel pins. The amount of hardware and the specific components differ for PWRs and BWRs, and these differences extend even within a fuel class. Generally, the hardware will remain part of the fuel assembly, unless both are consolidated.

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When hardware is part of the assembly, the radioactivity of the hardware is small, compared with the activity of the fuel within the assembly. However, relatively short-lived hot spots of 60Co may be found in some assemblies. Thus, when the hardware is part of the assembly, the hardware reduces the specific emissions (number of emissions per unit mass or volume) of the fuel assembly.

If the fuel is disassembled or if large amounts of fuel assemblies are consolidated, we must know more specific details about the hardware: physical properties of the hardware, its radiological characteristics, and whether or not it is greater than Class C (GTCC).

1.11 Modeling

Models are necessary to predict future thermal, structural, chemical, and nuclear responses of waste forms placed in the expected environment of a suitable repository when it is not practical to experimentally measure such responses. Several methods are employed in modeling. Physical models can be built to a certain scale, or even built with components on a scale which is distorted from the rest of the model. Models can be based on knowledge of mathematical relationships governing the factors being studied. In some cases a computer model may combine some of the same procedures used on the physical models together with numerical processing. In other cases it may be possible to use an analog model, i.e., model the phenomenon being studied by using a different phenomenon which obeys the same mathematical laws.

In all cases we must know what laws or relationships govern the interactions of the physical variables and functions being studied. Thus, to do modeling we must first know what factors are involved and how they interact. We must plan and carry out experiments to gather sufficient data to calculate or deduce relationships. With sufficient data, we can construct and run models. The models must be tested and validated, and only then can we use them to make predictions.

For spent-fuel waste forms, we must model rates for cladding failure, oxidation, and dissolution of many materials. At this time we are planning or executing experiments. From the experiments that have been run, some relationships have been deduced, and the data has been presented in tabular, graphic, or empirical form.

1.12 Burnup Models

Models exist which predict radionuclide concentrations. These models can be used to calculate other properties which are directly dependent on the concentrations. However, more specific attributes, such as fission gas release, grain size, and pellet fragmentation are changes in fuel characteristics, which depend on the burnup and the thermal history of the fuel.

Although dependent on burnup and thermal history, fission gas release has been named as a criterion in selecting spent fuel ATMs. Investigations are underway to determine distributions of burnup and fission gas release in the present and future LWR spent-fuel inventory.

No models exist to predict the total effect of exposure in a reactor core on fuel or other materials.

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1.13 Glass Modeling Status

Models are being developed to predict the behavior of the glass waste forms in a Yucca Mountain repository during the period of regulatory concern. Information from these models will be used in performance assessments to calculate the release of radionuclides from breached glass waste containers over time. These assessments are required to demonstrate compliance with the containment and controlled release requirements of 10CFR60.113, and to find the fractional contribution of the glass waste form in the cumulative release limits of 40CFR191.13.