EVALUATION OF SPENT NUCLEAR FUEL AS MIXED WASTE

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1.0 INTRODUCTION

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This report assesses whether spent nuclear fuel should be classified as hazardous waste as defined by the Resource Conservation and Recovery Act (RCRA). Disposal of spent fuel is the responsibility of the Department of Energy (DOE). Ultimate disposal will be in a geologic repository licensed by the Nuclear Regulatory Commission (NRC). If spent nuclear fuel were shown to contain RCRA-regulated constituents, it would constitute a mixed waste and therefore be subject to dual regulation by the EPA and the NRC (or DOE). This project was undertaken by the EPA Office of Radiation Programs to assist in a joint determination by these agencies of the correct regulatory regime to be imposed on this type of waste.

The objective of this report was to review the relevant data on spent fuel characteristics and to compare these data with the waste classification scheme in 40 CFR Part 261. The comparison will determine whether spent nuclear fuel meets one or more of the criteria defining a hazardous waste. Interested parties include the EPA, NRC, DOE, state regulatory agencies, nuclear utilities, and the vendors responsible for movement and disposition.

A determination that spent fuel is a hazardous (and therefore "mixed" waste) would have far-reaching implications. The EPA, as well as States with mixed waste regulatory authority, would have to assert authority over the handling of spent fuel and enforce compliance with RCRA regulations. Existing rules and practices authorized by the NRC and the DOE would be complicated by overlaying additional RCRA requirements. Utilities and other reactor operators would become generators with respect to spent fuel as a hazardous waste. Fuel pools and other planned storage facilities might have to seek permits as RCRA storage facilities.

The radioactive properties of spent fuel are obvious, and the responsibilities for control of its radioactive aspects are unambiguouily assigned to the DOE and NRC. The possible classification of spent nuclear

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fuel as a mixed waste hinges on the regulatory definition of hazardous waste and the physical and chemical characteristics of spent fuel. Section 2 summarizes the mixed waste issue. It also discusses the applicable governing documents and places this issue in its historical context. The definitions, criteria, and classification schemes for hazardous waste are presented in Section 3. Section 4 summarizes the relevant data on spent fuel. Section 5 applies the hazardous waste criteria to the existing data for spent fuel. The possibility that fuel pool water constitutes a hazardous waste stream subject to RCRA regulations is discussed in Section 6. Conclusions are presented in Section 7.

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2.0 BACKGROUND

2.1 Radioactive Waste Regulation

The Atomic Energy Act of 1954 (AEA) as amended¹ authorized the Atomic Energy Commission (AEC) to produce special nuclear material in its own facilities, to produce atomic weapons, to research and develop military applications of atomic energy, and to safeguard nuclear materials and information. The AEC was also given the task of both promoting and regulating civilian, commercial applications of atomic energy. The conflict between the roles of promoter and regulator was resolved with the passage of the Energy Reorganization Act of 1974.² This act created the Nuclear Regulatory Commission (NRC) and the Energy Research and Development Administration (ERDA). ERDA was given responsibility for all non-regulatory functions of the AEC. The NRC was assigned the regulatory and licensing responsibilities.

With a few exceptions, the regulatory authority over commercial uses of atomic energy can be delegated to States with qualified technical and administrative programs. The NRC retains responsibility for nuclear power facilities. The NRC was given no regulatory authority over ERDA facilities with two exceptions: the NRC retains regulatory authority over demonstration reactors (including the LMFBR) and over facilities for the receipt and storage of high level radioactive wastes.

The Department of Energy Organization Act³ transferred ERDA's responsibilities and functions to the newly created Department of Energy. This act assigned to the DOE a wide array of waste management responsibilities. Except for those licensing authorities already granted to the NRC, the DOE continued to exercise broad discretion in the management of its own wastes.

Under this regulatory scheme, the agency responsible for protecting public health and safety from hazards associated with handling any given

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radioactive waste stream could be the NRC, the DOE, an Agreement State, or some combination thereof. Radioactive waste is divided into several categories: high level, transuranic, and low level wastes. Regulations for the management of radwaste and responsibilities for its ultimate disposition vary for each waste category. Although the NRC and DOE systems for classifying radioactive waste are not identical, the position of spent fuel is unequivocal. Spent fuel is defined as a high level waste in 10 CFR Part 60.2. Under the Nuclear Waste Policy Act of 1982⁴, the DOE is required to take title to all spent fuel. Spent fuel which is not reprocessed will be disposed of in a geologic repository which will be licensed by the NRC. Thus, while the DOE has formal responsibility for the development of safe handling, transportation, and disposal methods for spent fuel, the disposal is subject to NRC approval under the licensing process for the high level waste repository.

2.2 RCRA - Regulation of Hazardous Wastes

The passage of the Resource Conservation and Recovery Act (RCRA) in 1976⁵ created a "cradle-to-grave" regulatory framework for dealing with the Nation's hazardous waste. In passing RCRA, Congress excluded byproduct, special nuclear material (SNM), and source material from the definition of a solid waste. See Section 1004(27) of RCRA. In addition, Section 1006 (a), stated that RCRA would not apply to any materials or activities regulated under the AEA "... Except to the extent that such application (or regulation) is not inconsistent with the requirement of such Acts."

Notwithstanding Section 1004(27) of RCRA, questions were raised about the applicability of RCRA to the existing LLW disposal sites. The EPA's position was that the disposal facilities and the wastes being received there were not completely exempt from RCRA. On July 3, 1986, EPA published a Federal Register Notice⁶ affirming the EPA's authority to regulate the hazardous component of mixed waste. The notice addressed the requirement that States include the regulation of mixed wastes in their hazardous waste management programs in order to maintain or obtain authorization for their programs. On September 23, 1988, the EPA published a followup notice which clarified interim status qualifications and extended the deadline for owners and operators of facilities handling mixed wastes to obtain interim status.⁷

Thus, mixed wastes are wastes containing materials that are regulated under RCRA as well as materials regulated under the AEA. Compliance with the dual sets of regulations complicates the handling, transport, and storage of such materials. At present, there are no disposal facilities for mixed wastes, but this may change as the various State Compacts begin to construct and operate their own disposal sites under the authority of the Low Level Radioactive Waste Policy Act as amended.⁸ The compact authorities are expected to make specific arrangements for the disposal of mixed wastes either in segregated or wholly separate facilities. Facilities for high level and TRU wastes will address the handling of mixed wastes on a site and wastestream specific basis. See, for example, the Notice of Proposed Decision, Conditional Variance to Department of Energy Waste Isolation Pilot Plant.⁹

With respect to low level wastes, none of the three existing LLW disposal sites will now accept RCRA-regulated (i.e., mixed) wastes. The disposal sites in Beatty, Nevada and Hanford, Washington, chose not to seek the required permits to become authorized disposal facilities for hazardous wastes. South Carolina expressly forbade disposal of mixed LLW at Barnwell. These developments eliminated the only possible disposal sites which might otherwise have existed for disposing of commercial mixed wastes.

On January 8, 1987, the EPA and NRC issued a joint guidance document¹⁰ which defined mixed LLW and put forth a scheme for identifying it. Mixed LLW was therein defined as: "...waste that satisfies the definition of low-level waste in the LLW Policy Amendments Act of 1985 and contains hazardous waste that either (1) is listed as a hazardous waste in Subpart D of 40 CFR Part 261

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FIGURE 2-1

IDENTIFICATION OF MIXED WASTE



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or (2) causes the low-level waste to exhibit any of the hazardous waste characteristics identified in Subpart C of 40 CFR Part 261." Figure 2-1 is a flow diagram for determining whether a waste material is a mixed waste.

In discussing the exclusion of source, special nuclear, and byproduct material from RCRA regulation, the document states that the "... NRC and EPA consider that only the radionuclides themselves are exempt from RCRA."

This same definition was formalized in a final rulemaking published by the DOE on May 1, 1987.¹¹ This rule interprets the AEA definition of "byproduct material" as it applies to DOE-owned or -produced mixed wastes. The key part of this interpretive rule states that when determining the applicability of RCRA to a waste stream "...the words 'any radioactive material'...refer only to the actual radionuclides dispersed or suspended in the waste substance." Under both this rulemaking and the NRC/EPA joint guidance, the non-radioactive fractions of the waste stream would be handled in accordance with RCRA regulations if it were either a listed waste or exhibited the characteristics of a hazardous waste. In effect, these definitions mean that if the radioactive portion of the waste stream has to be handled in compliance with both the EPA's and the NRC's (or the DOE's) programs for implementing RCRA and AEA, respectively.

The issues of guidance and definition discussed above have generally been addressed in the context of dealing with low level radioactive wastes and not spent fuel. However, the EPA assertions of authority over the hazardous portions of radioactive waste^{6,7} make no distinctions as to the levels or types of activity in the waste. The NRC's or the DOE's classification schemes for the radioactive portions of the waste are irrelevant to the regulation of the hazardous components. To determine the applicability of RCRA, the same arguments are applied to spent fuel as had been applied to low-level waste. By this reasoning, even if spent fuel is defined as a byproduct material, the resulting RCRA exclusion would apply only to the radioactive isotopes. It is

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still necessary to determine if other components of this waste cause it to meet the RCRA definitions of a hazardous waste. The balance of this report seeks to make this determination.

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3.0. HAZARDOUS WASTE

3.1 General Definition

For the purposes of compliance with RCRA, hazardous wastes are defined in 40 CFR Part 261. The flow chart in Figure 3-1 is useful in analyzing whether a waste is hazardous. The probable logical path to be followed in dealing with spent nuclear fuel has been highlighted in this figure.

Section 261.4 enumerates a number of exclusions by which materials avoid regulation under RCRA. The first set (261.4(a)) lists five types of materials that are determined not to be RCRA solid wastes. This includes the exclusion for AEA source, special nuclear material, and byproduct material. As already noted, however, this exclusion applies only to the radioactive isotopes themselves. The second set of exclusions (261.4(b)) lists a series of waste streams that are excluded from the hazardous waste portions of the regulation but are still subject to other parts of the regulations dealing with disposal of non-hazardous wastes.

Once it is determined that a material constitutes a solid waste and is not excluded under Section 261.4, the next question is whether the material constitutes a hazardous waste to which RCRA applies. The EPA has defined hazardous waste in two ways -- listed and characteristic. The RCRA regulations sometimes address listed and characteristic wastes differently, so it is important to recognize this difference.

A waste generator must determine if the waste stream contains any of the RCRA-listed wastes in 40 CFR Part 261.31 through 261.33 (Subpart D). If the

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DEFINITION OF A HAZARDOUS WASTE



waste is found on either of these lists, it is a RCRA-listed waste and must be managed as hazardous waste. If the waste stream is not found on the RCRA lists, then the hazardousness of the waste must still be determined through knowledge of the process that generated the waste (e.g., starting materials and process chemistry) or by testing. EPA provides test methods to determine if a waste is hazardous in the methods manual, "Test Methods for Evaluating Solid Waste Physical/Chemical Methods," SW-846, Second Edition.¹² Throughout this document, references will be made to "equivalent methods." This term refers only to those methods formally submitted to and approved by the Administrator as equivalent, as provided in the regulations. In addition to the initial waste characterization, RCRA requires waste recharacterization whenever the waste-generating process is changed.

Figure 3-1 illustrates the options available in determinating whether a waste fits the definition of hazardous waste.

3.2 Listed Wastes

The first way a waste may be considered hazardous is if it is a "listed" waste as described in Part 261, Subpart D. Listed wastes are waste streams containing hazardous constituents known to be toxic^a. For example, spent halogenated solvents are known to be toxic and are therefore considered hazardous. If a facility generates spent halogenated solvents, it is not necessary to test the waste or even make a determination; the waste is by definition hazardous. Listed wastes are defined and enumerated in 40 CFR Parts 261.31, 261.32, and 261.33. Section 261.31 lists waste streams defined as hazardous wastes that do not arise from any specific industrial source. That is, they may be generated in a variety of typical industrial applications. Chlorinated degreasing solvents are an example of such a waste stream. Section 261.32 lists hazardous waste streams that arise from specific industrial sources. An example would be distillation bottoms from the

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[&]quot;Wastes may also be listed if they exhibit one or more of the hazardous waste characteristics.

production of carbon tetrachloride. Section 261.33 contains a long list of commercial chemicals that become hazardous wastes when discarded. Also defined as hazardous wastes are any off-specification products, manufacturing intermediates, container residues, or spill cleanup residues of the same chemicals. Included are organics such as aldicarb and inorganics such as calcium cyanide.

If a waste appears on any of these lists, it is a hazardous waste unless a specific generator has successfully petitioned to have the waste delisted for his facility. In this case, that specific waste would be listed in Appendix IX of Part 261.

3.3 Characteristic Hazardous Wastes

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If a waste is not listed^b, it may still be considered a hazardous waste if it exhibits one of the characteristics of hazardous wastes described in Part 261, Subpart C. These characteristics are ignitability, corrosivity, reactivity, and toxicity. They are defined in Sections 261.21, 261.22, 261.23, and 261.24, respectively. Specific tests and/or properties are described for each characteristic. A waste that fails any of the characteristic tests is a hazardous waste and subject to RCRA regulation.

3.3.1 Characteristic of Ignitability

A solid waste is defined as having the characteristic of ignitability if it meets any one of four conditions:

1. If a liquid, the waste must have a flashpoint of less than $60^{\circ}C$ (140°F), as determined by prescribed methods. The determination will be made:

^bThe recently promulgated third-third land ban rule requires generators to determine whether their waste exhibits the characteristic (even if the waste is listed) to comply with the land disposal restriction regulations.

- a. with a Pensky-Martens Closed Cup Tester using the test method in ASTH Standard D-93-79 (13) or ASTM Standard D-93-80 (14).
- b. with a Setaflash Closed Cup Tester using the test method in ASTM Standard D-3278-78⁽¹⁵⁾; or
- c. by using a method determined to be and approved as equivalent by the Administrator OR
- 2. If not a liquid, the waste must be capable under standard temperature and pressure, of causing fire through friction, absorption of moisture, or spontaneous chemical changes. When ignited it must also burn so vigorously and persistently that it creates a hazard.

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3. The waste must be an ignitable compressed gas as defined in 49 CFR Part 173.300 and as determined by the test methods in that regulation. Alternate test methods may be approved by the Administrator.

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4. The waste must be an oxidizer as defined in 49 CFR Part 173.151.

Of these four conditions, only one could possibly apply to spent fuel. None of the spent fuel types in the United States will be ignitable compressed gas (3, above) or an oxidizer (4, above). A few reactors use a liquid fuel (see discussion in Section 4.3), but the fuel is an aqueous solution which has no vapor pressure of ignitable compounds. Therefore, the flashpoint condition (1, above) will not be exceeded.

It is possible that some cladding, moderator; or another material integral to one of the more unique reactor types may be capable of ignition under some circumstances. Heat will be generated by the decay of radioactive fission products after the fuel is removed from the reactor. This heat can cause a substantial temperature rise, especially if required cooling were to be removed or inhibited. If the temperature were to rise sufficiently to cause ignition of portions of the fuel assembly which were not excluded under RCRA (i.e., not byproduct material), an interesting regulatory question arises. The temperature at which the ignition would occur is certainly not "standard temperature and pressure." The observed temperature rise results from spontaneous changes in the material, but these changes are occurring in a material specifically not regulated by RCRA. The question of ignitability will be applied to specific spent fuel types in Section 5.

3.3.2 Characteristic of Corrosivity

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A waste will be considered hazardous because it has the characteristic of corrosivity if it meets one of two conditions:

- It must be aqueous and have a pH which is less than or equal to 2 or greater than or equal to 12.5. The pH is to be determined by the EPA method in SW-846, 2nd Edition¹² or by an equivalent method.
 OR
- 2. It must be a liquid and corrode steel (SAE 1020) at a rate greater than 6.35mm (0.250 inches) per year at a test temperature of 55° C. The test method to be used is that specified in NACE standard TM-01-69⁽¹⁶⁾ which has been standardized in SW-846, 2nd Edition ⁽¹²⁾ or an equivalent method.

Since only one type of reactor uses a liquid fuel, only a single type of spent fuel could possibly meet the criteria of corrosivity.

3.3.3 Characteristic of Reactivity

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The waste shows the characteristic of reactivity if it has any one of the following eight properties:

1. The waste is normally unstable and readily undergoes violent change without detonating.

2. It reacts violently with water.

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- 3. It forms potentially explosive mixtures with water.
- 4. When mixed with water, it generates toxic gases, vapors, or fumes in a quantity sufficient to present a danger to human health or the environment.
- 5. It is a cyanide or sulfide-bearing waste which, when exposed to pH conditions between 2 and 12.5, can generate toxic gases, vapors, or fumes in a quantity sufficient to present a danger to human health or the environment.
- 6. It is capable of detonation or explosive reaction if it is subjected to a strong initiating source or if heated under confinement.
- 7. It is readily capable of detonation or explosive decomposition or reaction at standard temperature and pressure.
- 8. It is a forbidden explosive as defined in 49 CFR 173.51, or a Class A explosive as defined in 49 CFR 173.53 or a Class B explosive as defined in 49 CFR 173.88.

The known spent fuel forms could possibly exhibit only the second of the eight listed properties. None of the remaining seven apply to spent fuel. The potential applicability of the second property will be discussed in the section dealing with the appropriate spent fuel type.

3.3.4 Characteristic of Toxicity

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Of the four possible characteristic hazardous wastes, the Toxicity Characteristic (TC) has the potential to apply to the broadest number of spent fuels. Hence the discussion of spent fuels will concentrate on this characteristic to a great degree, especially for the commercial LWRs. A solid waste will exhibit the Toxicity Characteristic, if the extract obtained by the appropriate test method contains any of the contaminants in 40 CFR 261.24, Table 1, in concentrations equal to or greater than the respective values shown in that table. A total of 39 constituents are listed, 31 of which are organic compounds. For purposes of this study only the inorganic contaminants are considered. The organic constituents listed will not be contained in spent nuclear fuel. The required test method is Method 1311, the Toxicity Characteristic Leaching Procedure (TCLP).¹⁷ The method provides several options depending on the constituents of possible interest in the waste. Specialized equipment, the Zero Headspace Extractor (ZHE), is required if the volatile organic constituents are targeted. If no volatile constituents are being analyzed, simplified extraction and filtration equipment are adequate. The following is a summary of the method as applied to the inorganic constituents.

A 100 gram, representative sample is pressure filtered through a 0.6 to 0.8 micrometer, borosilicate glass-fiber filter to separate its liquid and solid components. A maximum pressure of 50 psig is used to expedite the filtration. Other methods (e.g. centrifugation) may be used as a separation aid as long as the liquid and solid portions are still filtered through the required filtration apparatus. If the solid portion is less than 0.5% of the original sample weight it is discarded and the liquid is analyzed directly for the constituents of interest. If the sample will obviously yield no liquid under pressure filtration (i.e. it is 100% solid), the filtration step may be skipped.

The solid portion to be analyzed must have a surface area equal to or greater than 3.1 cm/gram or be capable of passing through a 9.5 mm standard sieve. The surface area criterion is applicable to filamentous materials such as paper, cloth or similar materials. Other materials must have their particle sizes reduced by cutting, grinding or crushing so as to pass through the required size sieve. The solid is weighed and placed in a suitable extraction vessel with 20 times its weight of the appropriate extraction fluid. The possible extraction fluids consist of either acetic acid and sodium hydroxide at a pH of 4.93 ± 0.05 or acetic acid alone at a pH of 2.88 ± 0.05 . The choice of extractant is based on a measurement of the neutralizing capacity of the waste. The extractor contents are mixed by end over end rotation for 18 ± 2 hours.

At the end of the extraction, the extraction mixture is separated into its component phases by filtration through a new, acid-washed, glass-fiber filter. The pH of the extract is recorded and the extract preserved for analysis. If compatible, the filtrate is combined with the liquid phase of the original sample. The combined liquid is the extract of interest. This extract is analyzed for the constituents of interest using the appropriate analytical methods from SW-846.⁽¹²⁾ If the extract and the original filtered liquid are not compatible, the liquids must be analyzed separately and volume-weighed average concentrations are calculated.

The test summarized above is referred to as the TCLP. The purpose of the test is to simulate leaching of toxic constituents from a solid waste.

Under improper management conditions, water infiltration could generate a leachate whose subsequent escape could contaminate ground water supplies.

The organic constituents and the ZHE method of the TCLP are of no concern for the characterization of Spent Nuclear Fuel. None of the organic constituents will be present. To evaluate whether Spent Nuclear Fuel exhibits the characteristic of toxicity only the inorganic constituents shown in Table 3.1 need be considered.

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TABLE 3-1

MAXIMUM CONCENTRATION OF INORGANIC CONTAMINANTS FOR TOXICITY CHARACTERISTIC

Contaminant	Maximum Concentration (milligrams per liter)
Arsenic	5.0
Barium	100.0
Cadmium	1.0
Chromium	5.0
Lead	5.0
Hercury	0.2
Selenium	1.0
Silver	5.0

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4.0 SPENT FUEL CHARACTERISTICS

This section describes the various types of Spent Fuel which are in existence in the United States. It includes descriptions of characteristics which are relevant to a determination of whether or not the spent fuel is a hazardous waste. An estimate of the concentration of the relevant elements in spent fuel is presented for commercial scale PWRs and BWRs. Although the analogous data are not presented for non-LWR and special LWR reactors, sufficient chemical and physical descriptions are supplied to extrapolate the conclusions to these other reactor types.

4.1 General Description

Spent nuclear fuel is any fuel which has been irradiated in a nuclear reactor and subsequently removed, but which has not been separated or reprocessed. Spent fuel is generally characterized by reactor type and burnup and decay time. Reactor type determines the chemical and physical form of the fuel while burnup and decay time determine the quantities of new isotopes produced by fission and activation and the amount of radioactivity that has decayed away since removal from the reactor.

The predominant spent fuel in the United States is that removed from the commercial light water reactors (LWRs) used for electrical power generation. Most of the LWRs in this country are Pressurized Water Reactors (PWRs) (about 75%) or Boiling Water Reactors (BWRs) (25%). Although differing in size, physical arrangement, and the construction materials used in the support assemblies, the fuel rods themselves are expected to be similar in their chemical and physical properties, at least insofar as their chemically hazardous constituents.

A variety of reactors exists in educational and research institutions, commercial research facilities, and DOE facilities for research and SNM production. There are also a few non-DOE government reactors. Although the

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total quantity of spent fuel generated by these reactors is small compared to that from the commercial LWRs, there is a broader spectrum of reactor types. The fuels from these reactors differ widely in terms of chemical form and associated matrix materials. The characteristics of the commercial fuels are discussed separately from the characteristics of these miscellaneous reactor types.

DOE has ongoing efforts to inventory and classify all high level wastes and spent fuels. Much of the information in this section is drawn from compilations of those data.^{18, 19} Table 4-1 summarizes the amount and types of spent fuel anticipated to be in inventory in the United States through the year 2020.

Fuel assemblies consist of:

- a. the matrix containing the fissionable materials,
- b. cladding material which confines and protects the fuel material,
- c. support and guide hardware, and
- d. criticality control materials (e.g., poison rods, moderators, control rods, etc.).

This document treats only the fuel matrix itself and the cladding as the "spent fuel" to be considered for RCRA regulation. The non-fuel hardware components will be low level wastes, although the highly activated components may still eventually be disposed of in the geologic repository.

A case could be made for the cladding being a container still serving its intended purpose and therefore not subject to RCRA. However, a small fraction will have defects. Therefore, the conservative assumption that the cladding is part of the waste is made for discussion purposes.

TABLE 4-1 SPENT FUEL INVENTORIES IN THE UNITED STATES PROJECTED TO 2020

Fuel Category	Fuel Description	Number of Reactors	Estimated Number of Fuel Elements ^(a)	MT I HM ^(b)
1	LWR, UO ₂ , Zircaloy clad	117	272,300 ^(c)	77,800
2	HTGR, UC ₂ /ThC ₂ , pyrolytic carbon, graphite clad	2	3853	29.2
3	MTR plate-type highly enriched U-Al alloy fuels	40	20,000	3.23
4	TRIGA U-ZrH ₂ fuels, SS or Al clad, mostly 20% enriched	28	4,500	0.80
5	Homogeneous UO ₂ -polyethylene discs or blocks, 20% enriched	8	87	0.03
6	PULSTAR and other low-enriched UO ₂ pin-array fuels	6	971	1.9
. 7	Liquid fuels (aqueous)	3	-	.015
8	U-Ho alloy highly-enriched fuel	6	d	d
9	FFTF mixed oxide fuel	1	đ	đ
10	Miscellaneous fuel	27	đ	d

a. Total through year 2020, including fuel in reactors at that time.
b. MTIHM-Metric tons initial heavy metal.
c. Number of fuel assemblies.
d. Not determined.

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4.2 Light Water Reactor Fuel Characteristics

Both commercial boiling water reactors (BWRs) and commercial pressurized water reactors (PWRs) contain fuel that consists of pellets of uranium dioxide (UO_2) most of which are contained in Zircaloy tubing (the cladding). Some early fuel was contained in stainless steel tubing. The elemental composition of zircaloy, an alloy of zirconium, is shown in Table 4-2. Of the elements shown, the only one of interest from a RCRA standpoint is chromium (Cr).

In addition to the fuel and cladding, there are structural components (e.g., spacer grids) which may be made out of Hastelloy, Inconel, or stainless steel. Again, the only material of interest in these alloys is chromium. Note that these structural components have a much higher chromium content (15-20 %) than Zircaloy.

Operating the reactor produces fission, decay, and activation products. Most of these are radioactive, but there are some stable isotopes of the elements of interest here. To determine the potential for spent fuel to exhibit the characteristic of toxicity, it is necessary to estimate the quantities of these elements produced in the reactor during operation and through the subsequent decay of radioactive isotopes. The ORIGEN computer code is a program used to calculate the inventory of fission and activation products (both stable and radioactive). We examined ORIGEN runs for a PWR and for a BWR. Table 4-3 shows the inventory of stable isotopes of interest for an 849 MWe PWR with a burnup of 36,000 megawatt-day/metric ton (MWD/MT). Because the values given here are on a per metric ton basis, they are reasonably representative of PWR plants in general (except that the higher burnup in newer plants will lead to somewhat larger values for these isotopes).

The first column in Table 4-3 gives the initial amount of each isotope present in the core before it starts operating. Of the eight elements of interest here, chromium is the only one initially present. The others are

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Table 4-2

ELEMENTAL COMPOSITION OF ZIRCALOY

Element	<u>Weight %</u>		
Zr	98.0		
Sn	1.5		
Cr	0.05-0.15		
Ni	0.05-0.15		
Fe	0.05-0.15		
0	0.15		

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either fission products, decay products, or activation products and begin to form only after the plant starts operating. The values in the second column are the amounts present after the fuel has been burned up to 36,000 MWD/MT and has been stored for four years.

Table 4-4 is similar to Table 4-3 except that the data are for a BWR and the ORIGEN output is for 5 years of storage. The ORIGEN run for the BWR was not as complete as for the PWR in that no values are given for a number of isotopes. However, except for Cr, the BWR values are similar to those for the PWR. The difference in the Cr content is probably the result of different structural materials.

The values in Tables 4-3 and 4-4 are based on grams of isotope per metric ton of uranium (ppm based on the weight of uranium). The value of interest is ppm of element based on the total weight of spent fuel (fuel plus cladding plus whatever structural components are part of the spent fuel). The following procedure is used to convert these values to the appropriate units.

First, 1 metric ton of U is equivalent to 1.13 metric tons of UO_2 . Next, for every metric ton of fuel (as UO_2), there are about 0.24 metric tons of cladding and structural materials in both PWR and BWR cores. Thus, the total values for each element in Tables 4-3 and 4-4 are multiplied by 1/(1.13 x 1.24) = 0.71 to get the grams of element per metric ton of fuel bundle. The results are given in Table 4-5 for each element of interest.

The data contained in Tables 4-3 through 4-5 represent an inventory of the elements of concern to the RCRA toxicity characteristic. These data can be expected to vary as a function of fuel configuration, burnup, decay time and other parameters. An exhaustive study of this relationship is beyond the scope of this report, however, some generalizations can be made. Increased

TABLE 4-3

PWR INVENTORY OF RCRA ISOTOPES (GRAMS/MTU) AT INITIAL LOADING AND 4 YEARS AFTER REMOVAL OF FUEL Burnup = 36000 MWD/MT

	•	After & Voc
<u>Stable Isotope</u>	<u>Initial</u>	of Decay
As-75 Total As	0 0	9.1×10 ⁻² 9.1×10 ⁻²
Ba-132 Ba-134 Ba-136 Ba-138 Total Ba	0 0 0	N ⁴ 267 31 1340 1638
Cd-106 Cd-108 Cd-110 Cd-111 Cd-112 Cd-113 Cd-114 Cd-116		N 8.5x10 ⁻⁷ 64 23 12 0.2 15 5
Total Cd Cr-50 Cr-52 Cr-53 Cr-54	0 614 12400 1440 366	118 591 12400 1410 423
Pb-204 Pb-206 Pb-207 Pb-208 Total Pb	0 0 0 - 0 0	N 2.3x10 ⁻¹² 3.5x10 ⁻⁹ 1.1x10 ⁻⁵ 1.1x10 ⁻⁵
Hg	no Hg reported	on ORIGEN run ^b
Se-74 Se-76 Se-77 Se-78 Se-80 Se-82 Total Se	0 0 0 0 0 0	N 2.4×10 ⁻³ 1 3 11 34 49
Ag-107 Ag-109 Total Ag	0 0 0	1.5x10 ⁻⁴ 73 73

 a N indicates isotope was not included in ORIGEN run.
 b Fission products with atomic weights greater than 167 were neglected in the ORIGEN run because their yield is so small.

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burnup should result in increases in the concentrations of all elements created, but variations in design and composition of structural components between reactors will dwarf this effect. This is particularly true of chromium.

A comparison of some of the values in Table 4-3 with an ORIGEN run for another PWR gives the following results:

<u>Stable Isotope</u>	PWR A (36.000 MWD/MT)	PWR B (50.000 MWD/MT)
	(g/MT)	(g/MT)
Ba-134	267	278
8a-138	1340	1937
Cd-108	63	67
Cr-52	12400	4172
Cr-53	1410	482
Pb-206	2.3×10^{-12}	2.2×10^{-26}
Pb-207	3.5×10 ⁻⁹	2.6×10^{-14}
Pb-208	1.1x10 ⁻⁵	5.3×10^{-5}
Ag-109	73	105

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Except for the Cr and Pb, the results are comparable. The lower Cr content for PWR B probably reflects the use of Zircaloy control rod guide tubes (PWR B is a more modern plant) versus stainless steel for PWR A. The generally higher numbers for the other isotopes are likely the result of the higher burnup for PWR B. The discrepancies between the decay products Pb-206 and Pb-208 probably result from roundoff. (The solution to the differential equations describing decay chains involve small differences between large numbers and hence are subject to roundoff errors when the quantities are small.)

The fission and activation processes will be fundamentally the same for the non-LWR and special LWR reactors. The relationship between burnup and construction materials and the ultimate concentrations of the elements of interests allows us to conjecture about the potential applicability of RCRA to

TABLE 4-4

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BWR INVENTORY OF RCRA ISOTOPES (GRAMS/MTU) AT INITIAL LOADING AND 4 YEARS AFTER REMOVAL OF FUEL Burnup = 50000 MWD/MT

<u>Stable_Isotope</u>	<u>Initial</u>	<u>After 4 Yrs</u> <u>of Decay</u>
As-75 Total As	0	N ^a N
Ba-132 Ba-134 Ba-136	0 0 0	N 284 N
Ba-138 Total Ba	0 0	1938 2222
Cd-106 Cd-108 Cd-110	0	N N 66
Cd-111 Cd-112 Cd-113	0	N N N
Cd-114 Cd-116 Total Cd	0 0 0	N N 67
Cr-50 Cr-52 Cr-53 Cr-54 Total Cr	N 711 N N 711	N 704 N N 704
Pb-204 Pb-206 Pb-207 Pb-208 Total Pb	0 0 0 0	N 4.1x10 ⁻¹² 9.9x10 ⁻⁹ 5.6x10 ⁻⁵ 5.6x10 ⁻⁵
Hg	no Hg reported	on ORIGEN run ⁶
Se-74 Se-76 Se-77 Se-78 Se-80 Se-82 Total Se	0 0 0 0 0	N N N N N
Ag-107 Ag-109 Total Ag	0 0 0	N 106 106

a N indicates isotope was not included in ORIGEN run.
 b Fission products with an atomic weight greater than 167 were neglected in the ORIGEN run because their yield is so small.

^aCalculated from data in Table 4-4, assuming a uranium to bundle weight ratio of 0.71. Reactor is a BWR with a burnup of 50,000 MWD/MT and a decay time of 5 years.

^bCalculated from data in Table 4-3, assuming a uranium to bundle weight ratio of 0.71. Reactor is a PWR with a burnup of 36,000 MWD/MT and a decay time of 4 years.

^cN indicates isotope was not included in ORIGEN run.

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^dFission products with an atomic weight greater than 167 were neglected in the ORIGEN run because their yield is so small.

TABLE 4-5								
INVENTORY	OF	RCRA	ELEMENTS	IN	REPRESENTATIVE	LWR	FUEL	BUNDLES

Grams/MT PWR^b

	(ppm)	(ppm)
As	Nc	6.5x10 ⁻²
Ba	1600	1100
Cď	48	85
Cr	500	11000
РЬ	4.0x10 ⁻⁶	7.8x10 ⁻⁶
Hg	0 ^d	0 ^d
Se	N	35
Ag	73	52

Grams/MT BWRª

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Element

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these other reactors based on the physical and chemical properties of their fuel assemblies. The next section discusses those properties.

4.3 Non-LWR And Special LWR Spent Fuels

Although the non-LWR and special LWR spent fuels are less than 0.1% of the total U.S. inventory, they comprise an array of chemical forms, physical configurations, enrichments, and burnups. Included in this category are the fuels from two HTGRs (Fort St. Vrain and Peach Bottom I) and a variety of research and test reactors. The research and test reactors fall into four general categories:

- 1. Reactors at universities or other educational institutions
- 2. Privately-owned research and test reactors
- 3. Reactors owned and operated by U.S. agencies other than the DOE
- 4. Reactors owned and operated by the DOE

Within these categories, different reactor types are used. Fuel types break down into these eight categories:

- 1. Materials Test Reactor, plate type
- 2. TRIGA
- 3. UO₂-polyethylene
- 4. PULSTAR, UO₂ pin type
- 5. Liquid fuels
- 6. Uranium-molybdenum metal
- 7. Fast Flux Test Facility UO₂-PuO₂ Fuel

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8. Miscellaneous

Table 4-6 gives a breakdown of the numbers of reactor types in the various owner/operator categories. It seems certain that some of this fuel will be placed in the repository without reprocessing. It is also highly probable, though not certain, that fuel in several of the above categories

will be reprocessed. Some fuels, if not reprocessed, will require special handling and preparation before being placed in the geologic repository. The nature of this processing is not known at present. Any determination of RCRA characteristics of these fuels at this stage would apply only during the storage period while decisions are made on the proper and safe treatment, recovery or disposal of this material.

4.3.1 High Temperature Gas Cooled Reactors

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There have been two high temperature gas cooled reactors (HTGR) operated in the United States. These were Fort St. Vrain, located in Platteville, Colorado, and Peach Bottom I, located in Delta, Pennsylvania.

Fort St. Vrain operated from January 1979 until it was shut down in August 1989. Fuel elements were hexagonal graphite blocks drilled with holes for fuel and coolant flow. The fuel particles were uranium and thorium carbide, coated with several layers of pyrolytic carbon and one coating of silicon carbide. The particles were bonded into right circular cylinders with a low-density graphite matrix. Fuel rods were 0.5 inches in diameter by 1.94 inches long. The initial core loading consisted of 774 kg of 93.5% ²³⁵U and 15905 kg of thorium. Its final core loading will be shipped to the Idaho National Engineering Laboratory (INEL) for storage; INEL is presently storing 720 spent fuel assemblies from this reactor.

Fuel blocks were constructed of nuclear-grade graphite, which had a very low level of impurities. None of the toxic metals listed in the Toxicity Characteristic are in the specifications for maximum concentrations of impurities. Impurity specifications for the metals other than iron, silicon, calcium, and aluminum are typically less than 70 ppm.²⁰ Impurity specifications for the fuel rods themselves were similar. Again, there were no reported concentrations for the elements of interest. The organic constituents of the TC are not expected to be a concern in either the cladding or fuel.

TABLE 4.6

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NUMBER OF RESEARCH AND TEST REACTORS IN EACH FUEL TYPE CATEGORY

	Fuel Type	University/ Educational	Private Research And Test	Government Owned (DOE)	Government Owned (non-DOE)
1.	MTR plate type, U-Al alloy, high enrichment	20	4	16	1
2.	TRIGA (U-ZrH ₂ Fuel)	18	5	2	3
3.	UO ₂ -polyethylene discs or blocks	7	0	0	0
4.	PULSTAR and other low- enriched pin type	3	2	1	0
5.	Liquid fuels (aqueous solutions)	2	0	1	0
6.	U-Mo alloy, high-enrich (93.2%)	ed O	0	4	2
? .	FFTF (UO ₂ -PuO ₂ pellets)	0	0	1	0
8.	Kiscellaneous	0	0	27	-
	Totals	50	11	52	6

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Peach Bottom Unit I was a smaller HTGR (115 MW(t)) operated between 1966 and 1974. Its fuel assemblies consisted of 3.5-inch diameter, 12-foot long cylindrical elements. These assemblies were mostly graphite which served as moderator, reflector, cladding, fuel matrix, and structure. The fuel was contained in annular compacts over a graphite spine. Fuel was in the form of uranium (93.15% 235 U) and thorium carbides coated with pyrolytic carbon. Most of the spent fuel (1,598 elements) from Peach Bottom I is in storage at INEL. 12 elements were shipped to the Oak Ridge National Laboratory (ORNL), and 28 elements are unaccounted for. Impurity data were not available for the Peach Bottom material, but the data quoted for Fort St. Vrain were for nuclear-grade graphite and should set an upper bound on the contaminants. Chemically, the two fuels would be expected to behave similarly. Both graphite and pyrolytic carbon are essentially insoluble in water, acid and alkali.²¹

The very low impurity levels in the graphite and the resistance of graphite to attack by water or acid makes it extremely unlikely that these materials would fail the Toxicity Test. Contaminant levels are lower and resistance to the acetic acid solution is probably greater for the graphitebased fuel compared to the metallic cladding and UO, fuels discussed above.

4.3.2 Materials Test Reactor (MTR) Plate-Type Fuels

Used by 41 reactors, fuels of this type are made of an aluminum-uranium alloy with aluminum cladding. Fuel elements are generally 7.6 x 7.6 x 90 centimeters, although the length can vary considerably. Enrichment ranges from 90 to 93%. The in-core inventory is approximately 360 kg of 235 U. The annual refueling requirement is approximately 100 kg of 235 U. This type of fuel has frequently been reprocessed in the past. It is unlikely to be shipped to the repository as spent fuel if any reprocessing at the Savannah River Plant (SRP) or INEL continues. Its absorption into the reprocessing system will generate an infinitesimal increase in the solidified HLW compared to defense HLW. Because of the high likelihood that this fuel will be

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reprocessed, any determination that it also constitutes a RCRA regulated waste will likely only affect storage requirements.

4.3.3 TRIGA Fuels

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The most common reactor type among research and test reactors is the TRIGA, built by General Atomics. The fuel used in TRIGA reactors is a uranium-zirconium hydride. The usual enrichment is 20%, although the enrichment could be as high as 70% for some fuels. The cladding of this fuel may be aluminum, stainless steel, or Incoloy-800, with stainless steel now being the standard. Because the economics are not favorable for reprocessing, this fuel will be disposed of in a repository.

The burnup (HWD/HT) is expected to be about two to four times that of commercial light water reactors. Based upon extrapolation of the time behavior of fission, activation, and decay products in the PWR A ORIGEN run, we estimate that the inventory of these isotopes will vary between the first power and the cube of the burnup. Thus, the inventory of fission, activation, and decay products for the TRIGA reactors will be between 2 to 64 times the values given in Table 4-3. This estimate is not borne out by the comparison given above for the two PWRs with different burnups. (We do not understand the reason for this.) The chromium content will be considerably higher because of the stainless steel or Incoloy cladding.

Elements are generally 30 inches long and 1.5 inches (for SS or Al) or 0.54 inches (Incoloy) in diameter. The density of the uranium-zirconium hydride is about 8 gm/cc, and that of stainless steel or Incoloy-900 is about 7 gm/cc. Thus, chromium constitutes about 1 percent of the fuel rod weight (10,000 ppm).

Of 31 reactors in the U.S., 27 are still operating. Approximately, 3,000 fuel rods are in use or in inventory, and almost 900 spent fuel rods are in storage. Reprocessing of these fuels is an open question since the

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4.3.4 UO2-Polyethylene Fuel

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Seven reactors use fuel in this form. The fuel material is an homogeneous mixture of UO_2 in polyethylene. Six reactors use the fuel in the form of disks, and one reactor uses a block form. Disks are 25.75 cm in diameter and range in thickness from 1 cm to 3.9 cm. The block form is 7.3 x 7.78 x 12.7 cm. Fuels are enriched to approximately 20% ²³⁵U. Neither reprocessing nor treatment technologies have yet been reported for this fuel form.

4.3.5. PULSTAR Type Reactors

These reactors are categorized by having low enriched UO_2 fuels arranged in pin-type arrays similar to commercial LWRs. Six reactors are in this category, three educational, two commercial, and one DOE-owned. The DOE-owned reactor is the Loss of Fluid Test (LOFT) Reactor at INEL. Fuel for this reactor is 4% enriched UO_2 in Zircaloy-4 cladding. Two of the educational reactors, at State University of New York and North Carolina State, are PULSTARS. The fuels are 4% and 6% enriched UO_2 in Zircaloy-2 cladding. These two reactors will generate approximately 1,600 kg of spent uranium fuel by the year 2020. The third educational reactor is a zero power reactor at Cornell University. Its fuel is 2.1% enriched UO_2 in aluminum pins. At its very low burnup rate, the total spent fuel generated by this reactor will be its current core load plus about 10 elements for a total of 1,700 kg of uranium.

The two private reactors, both owned by Babcock and Wilcox, have been shut down. Their fuel consisted of 2.5% and 4.0% enriched UO_2 with alterinum and stainless steel cladding, respectively. Spent fuel from these reactors has been forwarded to the Savannah River Plant for reprocessing.

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The fuels from these reactors are very similar to those used in commercial LWRs. If not reprocessed, the fuels can be expected to have the same RCRA status as commercial LWR fuels.

4.3.6 Aqueous Liquid Fuels

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Three reactors fall into this category, two educational and one DOE-owned. The two educational reactors use a solution of uranyl sulfate in water at enrichments of 20% and 80%. The total combined volume is 70 L. The DOE-owned reactor is the Solution High Energy Burst Assembly reactor at Los Alamos. This reactor utilizes approximately 80 L of 4.95% enriched uranyl fluoride. The total quantity of uranium in all three reactors is about 2.7 kg. These cannot be disposed of as free liquids. In view of the very small volumes involved, this fuel will likely be blended into a reprocessing campaign.

4.3.7 Uranium-Molybdenum Alloy Fuels

Six government-owned reactors use fuels consisting of highly enriched (93.2%) uranium-molybdenum alloy clad with nickel or aluminum.

Data on dimensions and physical characteristics or quantities of these fuels have not been obtained. The DOE currently handles the reprocessing of these fuels. It is assumed that this practice will continue and that the resulting waste will be included with DOE high-level waste.

4.3.8 UO2-PuO2 Fast Reactor Fuel

The Fast Flux Test Facility (FFTF) at Hanford is a sodium-cooled 400 MW(t) sodium-cooled fast reactor fueled with mixed oxide UO_2 -PuO₂ pellets contained in 0.23-in diameter stainless-steel-clad fuel pins. The pins are assembled into hexagonal assemblies, 12 ft in length, each containing 217 pins.

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4.3.9 Miscellaneous Fuels

DOE or its predecessors are operating or have operated some 27 miscellaneous reactors. Information on many of these reactors is incomplete. No fuel information was available for 14. Most of the rest used metal, metal alloy, or fuels similar to those discussed above. Some fuel from experimental breeder reactors (e.g., Fermi I and EBR-II) has a sodium or sodium-potassium metal bond between the cladding and fuel. The Fermi-I fuel consists of 510 assemblies (34.17 MTHM) in storage at INEL. A total of 0.281 MTHM from the EBR-II fuel are stored at Hanford, ANL-West, and Los Alamos.

Also in storage at INEL is a fuel from the Molten Salt Reactor Experiment (MRSE). The circulating fuel solution was a eutectic mixture of lithium, beryllium, uranium, and zirconium fluorides. The experiment was concluded in 1969, but the fuel has remained in the facility since that time. There is a total of 36.95 kg of U and 0.74 kg of Pu in this storage facility.

Other miscellaneous materials are in storage at each of the major DOE facilities. It is beyond the scope of this study to attempt classification of each of these materials for RCRA applicability.

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5.0 APPLICATION OF HAZARDOUS WASTE TESTS TO SPENT FUEL

The determination of whether spent fuel is a hazardous waste subject to regulation under RCRA can be made by tracing the appropriate logical path. It has been determined that we are dealing with a solid material that has served its intended purpose and is intended to be discarded or recycled. The exclusion from 261.4(a) for byproduct material applies, but only to the radioactive constituents. Therefore, the balance of the matrix is a RCRA solid waste.

Spent fuel is not excluded from being a hazardous waste under 261.4(b). It is not listed nor does it contain any components listed in Part 261, Subpart D. The key question becomes this -- does spent fuel exhibit any of the four characteristics of hazardous waste? Table 5-1 summarizes the findings of hazardous waste characteristics for each of the fuel types examined.

5.1 Ignitability

For a solid material to be considered ignitable, it must be capable of causing fire through friction, absorption of moisture, or spontaneous chemical changes and burning vigorously enough to create a hazard. This is not the case for the bulk form, UO_2 fuel in Zircaloy, of interest. Nor are most of the other spent fuel types ignitable under the RCRA definition. Because of the high heat conditions for which reactor components are designed, it is not surprising that the materials are not ignitable.

Of the spent fuel types examined, only two have combustible materials in their makeup. The first type is the homogeneous UO_2 in polyethylene spent fuel. Polyethylene is a combustible material; however, it is not ignitable under standard conditions of temperature and pressure. The reactors which use UO_2 -polyethylene fuel are very low power reactors used for educational purposes. The largest of this type is only 15 watts. With the very low thermal output from these fuels, there is no danger of spontaneous ignition.

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TABLE 5.1

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SUMMARY OF PROBABLE RESULTS OF RCRA CHARACTERISTICS TESTING

		Characteristic				
	Fuel Type	Ignitability	Corrosivity	Reactivity	EP Toxicity	
1.	LWR, UO ₂ , Zircaloy clad	. N	N	N	N	
2.	HTGR, UC ₂ /ThC ₂ , Pyrolyticarbon, Graphite clad	c N	N	N	N	
3.	MTR plate type, U-Al alloy, high enrichment	N	N	N	N	
4,	TRIGA (U-ZrH ₂ Fuel)	N	N	N	N	
5.	UO ₂ -polyethylene discs or blocks	N	N	N	N	
6.	PULSTAR and other low- enriched pin type	N	N	N	N	
7.	Liquid fuels (aqueous solutions)	· N	?	N	?	
8.	U-Mo alloy, high-enriche (93.2%)	ed N	N	N	N	
9.	FFTF (UO ₂ -PuO ₂ pellets)	N	N	N	N	
10.	Hiscellaneous	N	N	Y	N	

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N - does not have this characteristic
Y - shows this characteristic
? - unknown, but generator will have sufficient data to determine

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The second reactor type utilizing a combustible material is the HTGR. The graphite is theoretically combustible but has a very high ignition temperature. The Fort St. Vrain fuel is safe for storage in air with only passive convective cooling after it has been out of the reactor or shut down for 100 days.²² At that point, it can be shipped in a closed container in air without the decay heat causing ignition. Since Fort St. Vrain has been shut down since August 1989, its fuel has already cooled sufficiently to present no ignition hazard. These same arguments hold for the fuel from Peach Bottom I, which has been shut down since 1974. Its fuel has had longer to cool and presents no spontaneous ignition hazard. Thus, none of the existing forms of spent fuel in the United States are likely to exhibit the characteristic of ignitability.

5.2 Corrosivity

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Since corrosivity is defined only for liquids under RCRA, this excludes the spent fuel from all but the three liquid fuel reactors. These reactors have total quantities of spent fuel of less than 35 L for the two educational reactors and less than 80 L for the DOE reactor. No evidence of a specific test for corrosivity could be found; however, it seems likely that the generator will be able to make a determination based on the design basis for the reactor vessel. In the worst case, even if the waste is found to be hazardous by virtue of being corrosive, the quantities of waste are such that the generators would be subject only to the regulations for small-quantity generators.

5.3 Reactivity

This is the only characteristic definitely exhibited by some types of spent fuel. Certain fuels were manufactured with an active metal bonding between the fuel and the cladding. This metal was sodium or a sodium-potassium eutectic. Since these metals will undergo a violent reaction with water, the fuels containing them exhibit the characteristic of

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reactivity. The fuels in question are those from Fermi I, EBR II, and possibly other miscellaneous wastes stored at ANL-West. The total quantity of this spent fuel will be on the order of 34.5 MTU.

The final disposition of this fuel has not been determined but it is unlikely that fuel containing reactive metal will be acceptable for disposal at the geologic repository. Regardless of the disposal plans, this fuel is subject to RCRA regulations for storage of hazardous waste until the fuel has been treated and no longer exhibits the characteristic of reactivity.

5.4 Toxicity

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Determining whether the spent fuels exhibit the characteristic of toxicity was the most difficult part of this assessment. TCLP tests have not been explicitly performed on the spent fuel. Therefore, attempts were made to infer the fuel's probable behavior based on other data. These attempts concentrated on the LWR fuel, which is by far the largest quantity of spent fuel. Based on the values in Table 4-5, we can develop an upper limit estimate of how much of each of the eight hazardous elements will be released into 2 liters of water in 24 hours from a 100 gram sample of a fuel rod. Two approaches were used.

A computer code, ORIGEN, was used to predict the quantities of the stable elements of interest in typical spent fuel assemblies. In the absence of corrosion data for the preferred conditions, an estimate of material removal under reactor conditions was used as a conservative estimate. This was treated as the amount of "spent fuel" dissolved in the TCLP. The elements of interest were then scaled by their concentrations in the fuel to determine theoretical concentrations in TCLP leachate. The values obtained easily pass the maximum contaminant level criteria. The problems with this method are difficulties in relating the corrosion figures to dissolution in the TCLP. Also, the calculations assume that concentrations of the elements are uniform throughout the spent fuel and that the fuel and cladding undergo equivalent

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corrosion.

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We were not able to find any information on the corrosion rate of Zircaloy; however, a rule of thumb is that the corrosion release rate for stainless steel or Inconel at reactor conditions (2000 psi, 600° F) is 1 mg/dm²-mo. This is likely to be an overestimate for the Zircaloy cladding, which during its time in the reactor acquires a protective oxide film that does not corrode significantly. Nevertheless, we will use this value to estimate an upper limit for the amounts of the elements that would be released to the water.

The characteristics of the fuel rods are shown below for both a PWR and a BWR.

	PWR	BWR
Outside Diameter, cm	0.914	1.430
Clad Thickness, cm	0.0572	0.0813
Pellet Diameter, cm	0.769	1.237

Zircaloy has a density of 6.5 gm/cc, and the 95% theoretical density of UO_2 is 10.4 gm/cc (typical pellets are 95% of theoretical density). Given these values, we can calculate the surface areas of 100-gram samples of fuel rods. The results are 0.49 and 0.30 dm², for PWR and BWR fuel rods, respectively. The corrosion release rates are then calculated as follows:

PWR gm/day = 1 mg/dm²-mo x 0.49 dm² x 1/30 mo/day x 0.001 gm/mg = $1.6x10^{-5}$ gm/day BWR gm/day = 1 mg/dm²-mo x 0.30 dm² x 1/30 mo/day x 0.001 gm/mg = $1.0x10^{-5}$ gm/day

Releasing these amounts of fuel rod into 2 liters of water leads to the concentrations given in Table 5-2.

TABLE 5-2

CONCENTRATION OF RCRA ELEMENTS IN 2 LITERS OF WATER AS THE RESULT OF CORROSION RELEASE FROM PWR AND BWR FUEL RODS

	Concentration, ppm		
Element	PWR	BWR	
As	5.2x10 ⁻⁸	Nª	
Ba	8.8×10 ⁻⁴	8.2x10 ⁻⁴	
Cđ	6.8x10 ⁻⁵	2.4x10 ⁻⁵	
Cr	8.8x10 ⁻³	2.5x10 ⁻⁴	
РЪ	6.2x10 ⁻¹²	2.0x10 ⁻¹²	
Hg	0 ⁶	0 ^b	
Se	2.8x10 ⁻⁵	N	
Ag	4.2x10 ⁻⁵	3.6x10 ⁻⁵	

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N indicates isotope was not included in ORIGEN run. Fission products with an atomic weight greater than 167 were neglected in the ORIGEN run because their yield is so small. b

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An alternative approach to this estimate is to look at fuel solubility only. An ongoing study has reported bare fuel dissolution rates in the groundwater from the Nevada Test Site.^{23, 24} These studies have shown that the uranium concentration tends to approximate 1 to 2 mg/L under semi-static conditions. The maximum concentrations which were measured early in the tests were around 10 mg/L. These tests exposed bare fuel particles to the groundwater for cycles of up to 6 months. Periodic samples were analyzed for solution chemistry parameters. These showed chromium to be below the detection limit of 10 ppb in most samples. In high temperature tests ($85^{\circ}C$), chromium was found in the final test solution at 0.03 to 0.06 ppb after 180 days of exposure. (These tests were run in stainless steel containers so there may be doubt as to the origin of the chromium.)

The possible concentrations of the toxic metals can be estimated if it is assumed that the 10 mg/L figure for uranium represents a rapid dissolution figure for the exposed fuel. Since solution concentrations decrease after some time, it can be inferred that less soluble surfaces have been exposed or some surface protective reaction takes place. (The references contain a detailed discussion of the formation of the various mineral phases present.) Using the apparent concentrations of the RCRA-related elements in Table 4-5, and the 10 mg/L rapid solution value for uranium, we obtain the values shown in Table 5-3. The water used in these experiments had a final pH of 8.0 to 8.6. Therefore, it does not closely approximate the conditions of the TCLP. However, the cited fuel dissolution studies included an overnight soak in 8 H nitric acid. This would be a very aggressive leach relative to the TCLP. A 1 M solution would have a pH of approximately 0.1 compared to a TCLP leachate pH of 2.88 to 4.93. The analysis of this rinse solution indicated a uranium release in the acid rinse which averaged 2.7 times the total uranium released in the final solution. This small a release into such an aggressive leach implies that the extrapolation from the final solution data to a TCLP leachate is not likely to be off by as much as an order of magnitude.

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The comparison in Table 5-3 shows that the worst case value (PWR, Cr) is less than the maximum contaminant level permitted by a factor of 33. Even this is extremely conservative, since this analysis assumes that all the elements of interest are in the fuel when it is known that most of the chromium will be in the cladding.

A review of Tables 4-3 and 4-4 shows that the concentration of chromium in the fuel assembly changes very little from its initial value. Most of the chromium is present in the cladding and other structural materials. If one were to consider a waste stream composed of cladding hulls from which the fuel had been dissolved, chromium would be the key constituent in predicting the results of the TCLP. Since irradiation is unlikely to affect the bulk solubility of these materials, one would expect that the irradiated cladding and structural components would not be significantly more likely to fail the TCLP than the unirradiated material. Corrosion data tables for the metals of interest (aluminum, stainless steel, Zircaloy, etc.) all show excellent resistance to acetic acid in tests ranging from 5% to 95%.⁽²⁵⁾ Unfortunately, the data are not presented in a way useful for quantitatively predicting TCLP performance.

Elements which are mostly in the fuel include the fission products and various decay products. As shown in Tables 5-2 and 5-3 these are likely to be in very low concentrations. The lead isotopes result directly from decay through the uranium decay chains. To reach a concentration of lead in the fuel which could conceivably fail the TCLP would require approximately 1×10^6 years. It then would fail only if the fuel were 100% dissolved.

In summary, extremely conservative calculations indicate that the materials in spent fuel should easily pass the TCLP, but the argument could be strengthened by a few more directly relevant tests.

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TABLE 5-3	
ESTIMATED CONCENTRATION OF RCRA ELEMENTS	IF
URANIUM DISSOLUTION REACHES 10 MG/L	•••

	Concenti	<u>Concentration. ppm</u>	
Element	PWR	BWR	
As	9.1x10 ⁻⁷	Nª	5.0
Ba	1.6x10 ⁻²	2.2x10 ⁻²	100.0
Cđ	1.2x10 ⁻³	6.7x10 ⁻⁴	1.0
Cr	1.5x10 ⁻¹	7.0x10 ⁻³	5.0
РЬ	1.1x10 ⁻¹⁰	5.6x10 ⁻¹⁰	5.0
Hg	0 ⁶	0 ⁶	0.2
Se	4.9x10 ⁻⁴	N	1.0
Ag	7.3x10 ⁻⁴	1.1x10 ⁻³	5.0

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 a N indicates isotope was not included in ORIGEN run.
 b Fission products with an atomic weight greater than 167 were neglected in the ORIGEN run because their yield is so small.

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6.0 FUEL POOL WATER

In addition to the issue of spent fuel's status under RCRA, there is also a question as to whether the fuel pool water constitutes a hazardous waste stream subject to RCRA regulation.

The fuel storage pit of a typical reactor is used to receive and store fresh fuel for refueling operations, to store spent fuel, and to store various irradiated components removed from the reactor. The water in the fuel pool is highly purified and is used for heat removal and shielding. A simplified block diagram is shown in Figure 6-1.

Total volumes, capacities, and flow rates vary significantly between plants. The total volume of the water in the fuel pool can be expected to be on the order of 5x10⁵ gallons. Flow through the heat exchangers could range from less than 100 to thousands of gallons per minute. High-flow systems may divert a fractional bleed of 5 to 10% of this flow through the demineralizers. Exhausted filters are changed and disposed of as radwaste. Spent resins are pumped to spent resin storage tanks. Eventually, these are also processed and disposed of as radwaste. The spent resins may or may not be mixed with other resins prior to disposal.

The fuel pool water itself is continuously recycled back to the fuel pool. Connections with other systems may allow diversions to or from other systems for specific purposes. Should any of the water itself become "waste," it would most likely be routed through plant treatment systems to be discharged under an NPDES permit and plant environmental technical specifications. Small amounts may be entrapped in spent filters and resins or evaporated and lost through gaseous discharge systems. These amounts and their contamination levels are not significant for waste classification purposes. FIGURE 6-1 SIMPLIFIED FLOW DIAGRAM OF A SPENT FUEL PIT



6-2

To determine whether fuel pool water is a hazardous waste subject to RCRA regulations, one must determine whether the fuel pool water is a solid waste. It seems clear that fuel pool water does not meet the definition of a solid waste. It is not abandoned, nor is it inherently waste-like. It is not recycled as specified in 40 CFR 261.20. Fuel pool water is a combination coolant and radiation shield in a closed, continuous flow process. Any portion removed from the process would either be transferred for use in another process or would ultimately be discharged as a CWA point source effluent and therefore be excluded from RCRA regulation.

Two waste streams, exhausted filter media and spent resin, may be generated from processing the fuel pool water. Within the context of nuclear power plant operations, neither of these waste streams is unique. Both filters and spent resin will be generated from multiple other sources within the plant. These waste streams would be classified for disposal as low-level wastes. The types of contamination picked up by these systems are similar to the contamination removed by other filters and resin beds used in other plant systems. The applicability of RCRA regulations would be determined by the generator's (i.e., power plant operator's) knowledge of the waste composition and characteristics or by a measurement program. In any case, the status and disposition of these treatment system wastes are not relevant to the issue of spent fuel as a potential mixed waste.

6-3

7.0 CONCLUSIONS

11. 1

This study has reviewed the characteristics of spent fuel in order to evaluate the possibility that spent nuclear fuel may be classifiable as a hazardous waste and subject to RCRA regulation. The conclusions may be summarized as follows:

- Spent fuel does not fit the definition of a listed hazardous waste. None of the spent fuel types are listed in either the general waste stream list (261.31) or the industry-specific waste stream list (261.32). In addition, none of the fuels or the associated materials are contained in the commercial chemical list (261.33). Therefore, a spent nuclear fuel could only be a hazardous waste by virtue of exhibiting one of the characteristics of a hazardous waste.
- 2. None of the spent fuels are likely to exhibit the characteristic of ignitability.
- 3. None of the solid spent fuels exhibit the characteristic of corrosivity. Data need to be presented to verify this finding for the three liquid spent fuels. However, it is unlikely that they will be corrosive; and, even if they were, the quantities involved are trivial.
- 4. More than 99.9% of the spent fuels will not exhibit the characteristic of reactivity. One specific type of fuel contains a reactive metal which will result in the classification of these fuels as hazardous waste by virtue of the characteristic of reactivity. Although treatment of this waste before placement in the repository is virtually guaranteed, this classification affects storage requirements under RCRA.
- 5. Calculations and some experimental evidence suggest that it is highly unlikely that any of the spent fuel types will exhibit the toxicity characteristic. Although the assumptions in these methods are

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conservative, they are still subject to some questions. Generator knowledge of specific fuel properties and related tests on unirradiated materials would buttress these preliminary conclusions considerably.

6. The overwhelming majority of the spent nuclear fuel anticipated to be generated in the United States will not be classified as mixed waste. Questions remain about a number of the specialized and one-of-a-kind fuels. Sufficient generator knowledge probably already exists to determine the status of these fuels if the historical records relating to the material can be located.

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9.0 GLOSSARY

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"Byproduct material" means (1) any radioactive material (except special nuclear material) yielded in or made radioactive by exposure to radiation incident to the process of producing or utilizing special nuclear material, and (2) the tailings or wastes produced by the extraction or concentration of uranium or thorium from any ore processed primarily for its source material content.

"Container" means any portable device in which a material is stored, transported, treated, disposed of, or otherwise handled.

"Disposal facility" means a facility or part of a facility at which hazardous waste is intentionally placed into or on any land or water, and at which waste will remain after closure.

"EPA Kazardous waste number" means the number assigned by EPA to each hazardous waste listed in 40 Part 261, Subpart D, and to each characteristic identified in 40 Part 261, Subpart C.

"Equivalent method" means any testing or analytical method approved by the Administrator under 40 CFR 260.20 and 260.21.

"Generator" means any person, by site, whose act or process produces hazardous waste identified or listed in 40 CFR 261 or whose act first causes a hazardous waste to become subject to regulation.

"Geologic repository" refers to a planned type of disposal facility wherein high-level wastes will be placed in mined cavities in stable geologic strata for isolation from the biosphere.

"Hazardous waste" means a hazardous waste as defined in RCRA either because it is specifically listed or because it exhibits one of the hazardous waste characteristics.

"Hazardous waste characteristic" means one of four characteristics by which a solid waste is defined as hazardous. These are ignitability, corrosivity, reactivity, and toxicity. If a solid waste possesses one of these characteristics, it is hazardous regardless of the specific constituent which is responsible for the property.

"Hazardous waste constituent" means a constituent that caused the Administrator to list the hazardous waste in 40 CFR Part 261, Subpart D, or a constituent listed in Table 1 of 40 CFR 261.24, or Appendix VIII of Part 261.

"High Level Waste" means certain highly radioactive wastes, including spent fuel and specified wastes from the reprocessing of spent fuel.

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"Leachate" means any liquid, including any suspended components in the liquid, that has percolated through or drained from hazardous waste.

"Low-Level Waste" means radioactive waste which is suitable for near surface disposal. Classified differently by DOE and NRC, it is generally defined by what it is not, i.e. not high-level, not spent fuel, not transuranic.

"Mixed waste" means waste containing both hazardous and radioactive constituents and therefore subject to regulation under both AEA and RCRA.

"Representative sample" means a sample of a universe or whole (e.g., waste pile, lagoon, ground water) which can be expected to exhibit the average properties of the universe or whole.

"Reprocessing" means the mechanical and chemical treatment of nuclear fuel to separate uranium, plutonium and other heavy metal from the fission products.

"Small Quantity Generator" means a generator who generates less than 1000 kg of hazardous waste in a calendar month.

"Solid waste" means any garbage, refuse, sludge from a waste treatment plant, water supply treatment plant, or air pollution control facility and other discarded material, including solid, liquid, semisolid, or contained gaseous material resulting from industrial, commercial, mining, and agricultural operations, or from community activities, but does not include solid or dissolved materials in irrigation return flows or industrial discharges which are point sources subject to permits under section 402 of the Federal Water Pollution Control Act, as amended (86 Stat. 880), or source, special nuclear, or byproduct material as defined by the Atomic Energy Act of 1954, as amended, (68 Stat. 923).

"Source material" means (1) uranium, thorium, or any other material which determined by the Atomic Energy Commission (AEC) pursuant to the provisions of section 61 of the AEA to be source material, or (2) ores containing one or more of the foregoing materials, in such concentration as the AEC may by regulation determine from time to time.

"Special nuclear material" means (1) plutonium, uranium enriched in the isotope 233 or in the isotope 235, and any other material which the AEC, pursuant to the provisions of Section 51 of the AEA, determines to be special nuclear material; or (2) any material artificially enriched by any of the foregoing, but does not include source material.

"Spent Nuclear Fuel" means any nuclear fuel which has been emplaced in a reactor, irradiated, and has not been reprocessed, chemically separated or destructively analyzed.

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