

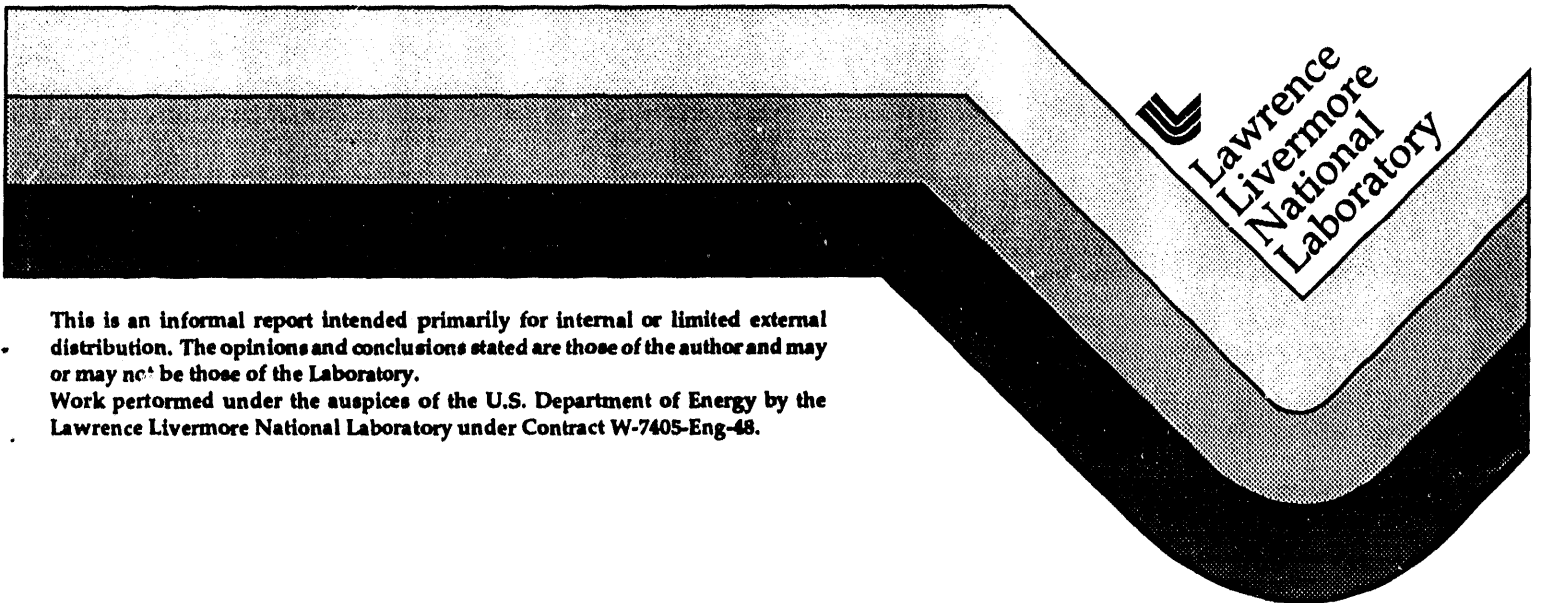
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Rationale for Determining MCC Spent Fuel Acquisitions

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June 1993



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**Rationale for Determining MCC
Spent Fuel Acquisitions**

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June 1993

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Summary

The Yucca Mountain Site Characterization Project of the U.S. Department of Energy (DOE) is investigating the suitability of the Topopah Spring Tuff at Yucca Mountain, Nevada, for use as a disposal site for spent nuclear fuel and other high-level waste forms. The performance of the high-level waste forms and the engineered barrier system at the site must be shown to comply with the requirements in 10 CFR 60. Lawrence Livermore National Laboratory (LLNL) has the responsibility for determining the performance of the U.S. commercial reactor spent nuclear fuels under potential repository conditions. Pacific Northwest Laboratory (PNL) performs testing of these highly radioactive materials in support of the LLNL program.

In the early 1980s it was recognized that a readily available, well characterized source of spent fuel was needed to operate a technically defensible spent fuel waste form performance program. For this reason, the Materials Characterization Center (MCC) was established by DOE-Headquarters (HQ) as part of its Materials Characterization Organization. The MCC was charged with the tasks of procuring, characterizing and archiving spent fuels of known irradiation history as Approved Testing Materials (ATMs). These characterized fuels provide the baseline for evaluating spent fuel performance under potential repository conditions.

This report summarizes the rationale for selecting additional spent fuels that should be acquired to support the LLNL and PNL testing programs. These programs have identified specific attributes that may affect spent fuel behavior in a repository. To meet the needs of the spent fuel testing programs, a minimum of five additional spent fuel ATMs with the following attributes should be acquired for testing:

- high burnup fuel (> 60 MWd/kgM) with strong rim effect
- two low fission gas release fuels [$< 10\%$ fgr, high burnup (> 50 MWd/kgM) and low burnup (< 10 MWd/kgM)]
- burnable poison fuel (5 to 8 wt% Gd_2O_3) with relatively high burnup
- high fission gas release boiling-water reactor fuel

This spent fuel acquisition plan should be reviewed annually as additional spent fuel performance data are obtained. This will ensure that the MCC is interfacing information with the spent fuel performance research and engineering program and that the MCC is providing the necessary fuels for the performance testing activities.

Contents

Summary	iii
1.0 Introduction	1.1
2.0 Spent Fuel Characteristics For New ATMs	2.1
3.0 Rationale For Acquiring Spent Fuel Testing Materials	3.1
4.0 Conclusions	4.1
5.0 References	5.1

1.0 Introduction

The Yucca Mountain Site Characterization Project (YMPO)^(a) of the U.S. Department of Energy (DOE) is investigating the unsaturated zone at Yucca Mountain, Nevada, as a potential location for a high-level waste repository. To obtain a required U.S. Nuclear Regulatory Commission (NRC) license for the potential repository, the performance of the high-level waste forms and the engineered barrier system proposed for Yucca Mountain must be shown to comply with 10 CFR 60. Lawrence Livermore National Laboratory (LLNL) has accepted responsibility for the high-level waste package design. LLNL has maintained an active research and engineering program to determine the performance of spent nuclear fuels at conditions pertinent to a Yucca Mountain repository. Pacific Northwest Laboratory (PNL)^(b) tests these highly radioactive materials in support of the LLNL program.

In the early 1980s, it was recognized that a readily available, well characterized source of spent fuel was needed to operate a technically defensible spent fuel waste form performance program. For this reason, the Materials Characterization Center (MCC) was established in 1980 by DOE-Headquarters (HQ) as part of its Materials Characterization Organization. The MCC was charged with the tasks of procuring, characterizing, and archiving spent fuels of known irradiation history. These fuels, called Approved Testing Materials (ATMs), would be used by DOE programs for spent fuel performance evaluation at the end of the fuel cycle. These programs included the Basalt Waste Isolation Project, the Salt Repository Project, the Yucca Mountain Repository Project, and the Commercial Spent Fuel Management Office. The ATMs would serve as an established, consistent supply of spent fuels for all DOE laboratories performing repository related fuel studies.

During the early 1980s, it was recognized that many microscopic characteristics (such as grain size/boundary area) could affect the performance of fuel stored in a repository. However, there were little or no directly related experimental data available that demonstrated a significant functional dependence on detailed microscopic characteristics. A systematic examination of the effects of each microscopic characteristic was determined to be both time and cost prohibitive and to require an extensive collection of test materials. Alternatively, based on in-reactor performance surveillance examinations, certain measurable macroscopic operating parameters were identified that could be related to microscopic variables (e.g., radionuclide inventory, fuel microstructure). Two such macroscopic variables are burnup and fission gas release. Based on the microscopic/macroscopic relationship, the

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- (a) The Yucca Mountain Project is managed by the Yucca Mountain Site Characterization Project Office as part of the U.S. Department of Energy Civilian Radioactive Waste Management Program.
 - (b) Pacific Northwest Laboratory is operated for the U.S. Department of Energy by Battelle Memorial Institute under Contract DE-AC06-76RLO 1830.

initial approach for ATM fuel selection subdivided the spent fuel population space into a 2 x 2 x 2 cubic matrix (Figure 1.1) with attribute variables of fission gas release, burnup, and reactor fuel type [boiling-water reactor (BWR), pressurized-water reactor (PWR)], recognizing that some fuels, such as burnable poison fuels, would not completely fit into this matrix (Barnes 1984; Mendel 1989).

The ability of the MCC to acquire these fuels was limited by utility cooperation, questions of ownership, minimum amounts of fuel the utilities would release, cost of transportation, and the utilities' lack of knowledge regarding operating and fabrication parameters. Once a large quantity of fuel covering a limited number of conditions was obtained (see Table 1.1), a small amount of the fuel was extensively characterized using proflometry, gamma scanning, ceramography (fuel), metallography (cladding), analytical transmission electron microscopy, burnup analyses, and fission gas analyses. Other data were documented such as the conditions under which the fuel was irradiated and handled, as-fabricated design and characteristics, in-reactor radiation history, and postirradiation handling and storage. Archive samples were maintained following characterization.

Other than ATM-101, which was received from Idaho National Engineering Laboratory (INEL), spent fuel ATMs have been provided by power utilities. To obtain fuel from the utilities, the MCC has had to be willing to accept either full fuel assemblies or equivalent quantities of mixed amounts of fuels. Generally, the utilities prefer to provide full assemblies, which allows them to efficiently manage spent fuel storage space at their storage facilities.

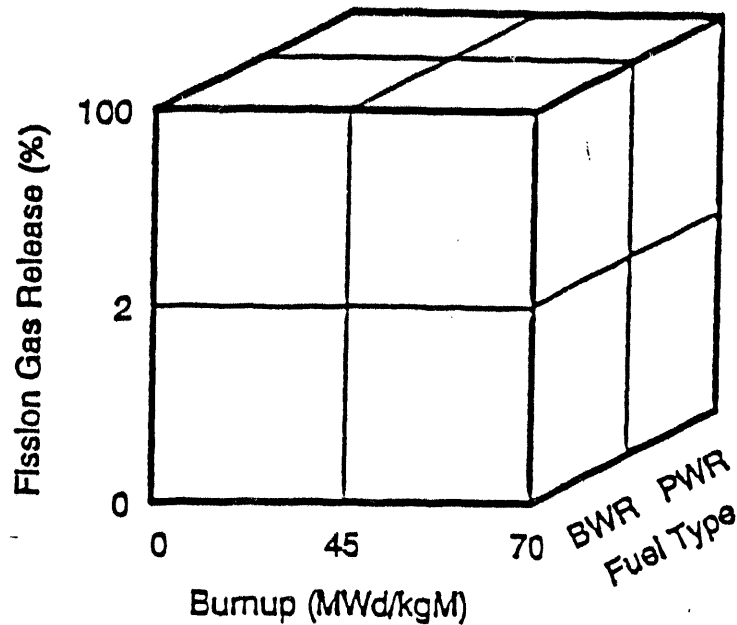


Figure 1.1. Matrix Used to Simplistically Define Attribute Variables of the Spent Fuel Population Space

Table 1.1. Characteristics of Current Spent Fuel ATMs and Quantity of Rods Used-to-Date

ATM	Rod Design	Fuel	Cladding	Nominal ^(a) Burnup (MWd/kgM)	Fission Gas Release (%)	Acquired Inventory (rods)	Number of Rods Used in Testing
101	PWR, WEC ^(b) 15 x 15	UO ₂	Zircaloy-4	32	0.2 to 0.3	9	1 (partially)
103	PWR, CE ^(c) 14 x 14	UO ₂	Zircaloy-4	33	0.25	176	1
104	PWR, CE 14 x 14	UO ₂	Zircaloy-4	44	0.4 to 1.1	128	1
105	BWR, GE ^(d) 7 x 7	UO ₂	Zircaloy-2	34	0.6 to 7.9	98	2 (partially)
106	PWR, CE 14 x 14	UO ₂	Zircaloy-4	44	7.8 to 18	20	1
108	BWR, GE	UO ₂ + 3-4% Gd ₂ O ₃	Zircaloy-2	~26	~9	included in ATM-105	< 1

- (a) Variations in burnup and fission gas release exist from rod to rod.
 (b) WEC - Westinghouse.
 (c) CE - Combustion Engineering.
 (d) GE - General Electric.

The procurement of full assemblies has several drawbacks: 1) Negotiating the transfer details of fuel ownership with the utilities requires a large amount of time. 2) Because shipping a full assembly requires full-size transportation casks, all transportation regulations for spent fuel must be met. 3) While general operational characteristics of the fuel have often been established by the utilities, the type of data generated by detailed destructive examination of the assembly is unavailable before acquisition. Therefore, extensive and costly characterization must be performed before using the fuel in research programs. 4) Shipping fuel cross-country is expensive, costing over \$100K per assembly. 5) A full assembly contains far more material than needed for the repository programs (see Table 1.1). The excess fuel presents a storage problem since the fuel must be held during destructive examination. Maintenance of the hot cell space used to store the excess fuel becomes more expensive each year.

Although most of these drawbacks can be overcome if time and funds are available, there is a significant complication associated with procuring full assemblies that have not been completely characterized. When the fuel is received, only macroscopic characterization (i.e., burnup, linear heat generation rate, operating history) may have been performed. The microstructural characteristics and fission gas release that make the fuel useful for the repository programs are unknown; these parameters must be defined during destructive examinations. Therefore, when a fuel is purchased, shipped, and characterized, its usefulness to the repository program has not been determined. Experience indicates that over \$500K is invested in an assembly before it is possible to determine whether the fuel is useful to the research programs. Such an investment was not a problem when the MCC initially began acquiring fuel, since nearly any fuel provided data useful for developing a baseline knowledge of spent fuel performance. However, subsequent research has resulted in a well developed data base on spent fuel

behavior, and because of funding and time constraints, the MCC must now ensure that only fuels with very specific characteristics are procured. Thus, the established method of acquiring uncharacterized full assemblies is no longer justifiable.

2.0 Spent Fuel Characteristics For New ATMs

The proposed new method for obtaining spent fuel ATMs requires the examination of parameters known to affect spent fuel performance. The performance response of spent fuel may be represented by the following equation:

$$\text{Response} = \sum R(\mu_i)P(\mu_i) \quad (2.1)$$

where R and P are the response and population of each attribute variable, μ_i . In the early 1980s, it was proposed that understanding spent fuel behavior under repository conditions would require evaluation of any variable that might influence that behavior. A long, complex list of variables that could potentially affect fuel performance was compiled. It included variables such as grain size, cladding type, reactor type, crud deposits, and storage conditions. The list was extensive enough that a program to investigate these variables would be extremely expensive.

When the repository programs began testing spent fuel, fuel was randomly selected with justification based on its location within the cubic matrix shown in Figure 1.1. As described earlier, this matrix was developed to limit the list of potential performance-affecting attributes to those that could be measured and related to other, more obscure attributes. Though useful during the early repository research activities, fuel selection using this population-based method is inadequate for selecting future ATMs. Using the existing ATMs, test results have identified a number of fuel characteristics that potentially influence the spent fuel radionuclide release performance. Thus, new ATMs will be selected with these specific characteristics in mind, so the functional and bounding impact of those characteristics on spent fuel performance in a repository can be determined.

The test results have indicated how several spent fuel attributes may affect the spent fuel performance in a repository (Einziger et al. 1992; Thomas, Einziger, and Buchanan 1992; Gray and Thomas 1992; Gray, Thomas, and Einziger 1993). A limited number of tests have been performed to evaluate the effect of fuel burnup over a range of 27 to 48 MWd/kgM. Results showed that the fuel oxidation and dissolution responses are insensitive to burnup over this range. However, studies performed in the U.S. and Canada suggest that at lower burnup values (< 10 MWd/kgM), fuel oxidation may more rapidly progress to higher oxide phases. To quantify this effect, fuel of a suitable lower burnup value should be acquired for testing purposes.

Additionally, extremely high burnup fuels (> 60 MWd/kgM), which may eventually compose a significant part of the spent fuel inventory, have not been tested by the repository programs. However, fuels extended to higher burnups can experience microstructural restructuring. Restructuring of fuel grains is understood to begin at the fuel pellet-gap interface and to progress inward toward the centers of the fuel pellets. This restructuring results in an extremely fine grain size and a significant increase

in the grain boundary surface area. The effect of restructuring on the oxidation and dissolution responses of spent fuel is unknown. Thus, a high burnup fuel with significant restructuring should be obtained for testing purposes.

Oxidation tests have been performed on a limited number of PWR fuels over the range of 0.3% to 18% fission gas release. The oxidation responses of these fuels were similar, indicating no apparent effect of fission gas release on oxidation behavior. However, these results have not been confirmed for BWR fuels, since only low fission gas release BWR fuels have been tested. Thus, a higher fission gas release BWR fuel(s) should be acquired for testing.

Limited dissolution tests have been conducted to determine if a relationship exists between the fission gas release and the inventories of radionuclides concentrated within the fuel-cladding gap of U.S. nuclear fuels. If established, such a relationship would be useful because it would relate an easily determined fuel parameter (fission gas release) to an important source term parameter (gap inventory, or instant release term). To date, however, only one type of fuel, ATM-106, has been tested. Additional fuels must be tested to determine if a relationship can be established that applies to a wide range of U.S. light-water reactor (LWR) fuels.

Related dissolution tests on these same ATM-106 fuels have indicated that the inventories of radionuclides concentrated within the grain boundaries are small (generally < 1% of the total inventories of Cs, Tc, or Sr). Again, additional fuels must be tested to determine if this observation is applicable to a wide range of U.S. LWR fuels. Grain boundary inventories are important source term parameters because they are commonly grouped together with the fuel-cladding inventories to yield what is often called the instant release term.

Burnable poison fuels have not been tested. Data from the mixed-oxide programs indicated at higher levels of burnable poison (> 10%) that actinides tend to stabilize the UO_2 matrix and that stabilization increases with increased levels of actinides. These data suggest that gadolinia-doped fuel may behave more favorably (with respect to oxidation behavior) than undoped UO_2 -based fuels. Preliminary studies of oxidation response have been initiated using the low-gadolinia fuel (ATM-108) currently available from the MCC (studies to determine the dissolution response of this fuel have been delayed). A modern, high-burnup, high-gadolinia (~ 8% Gd_2O_3) spent fuel should be acquired for testing.

In addition to the fuel parameters, the acquisition plan must address fuel cladding and hardware behavior, though cladding and failure response studies are currently suspended. However, it is believed that the cladding attributes of internal pressure, hydrogen content in Zircaloy, and oxide film thickness are important input parameters for evaluating cladding failure response under repository conditions. The current fuel supply is judged adequate for initial studies to determine which of these attributes will be most important to the cladding response. While these studies are in progress, the need for additional ATMs can be determined based on cladding properties. Also, it can be investigated whether or not population statistics are needed from other research programs, utilities, and fuel vendors.

Assembly hardware is another source of radionuclide release. It is well known from reactor performance studies that the properties of the steels, Inconels, and Zircalloys in the hardware are established at relatively low burnup, therefore, the current supply of hardware should be adequate for testing. In any case, cladding and hardware characterization activities will require cooperative interfaces with the fuel vendors, utilities, and others.

3.0 Rationale For Acquiring New ATMs

There are three means to obtain fuel samples for the DOE testing programs: 1) Small quantities of fuels can be obtained from DOE- or Electric Power Research Institute (EPRI)-sponsored programs. 2) Fuel may be obtained from foreign sources for studies of particular fuel characteristics. 3) Full-size assemblies can be obtained directly from commercial utilities, although for reasons discussed earlier, this option should be considered a last resort.

Some of the DOE- or EPRI-sponsored fuel testing programs use modern fuels irradiated to high burnups, as well as new fuel cladding designs intended for the next generation of fuels. These programs present an excellent opportunity to obtain small quantities of fuel. Since there is always the chance that the microscopic characteristics of the acquired fuels will be unsuitable, acquisition of small amounts of fuel from the testing programs would minimize risk and cost in the following ways:

- These fuels have undergone extensive characterization to develop an in-reactor data base for NRC licensing.**
- Since these fuels have already been sectioned, small quantities could be easily obtained at low cost (about \$50K for the acquisition). If the fuel is determined to be unacceptable, much less funding will have been invested than for a full assembly.**
- Limiting the quantity of fuel purchased would reduce the shipping costs since small casks could be used instead of full-size casks.**
- Smaller quantities of fuel would minimize storage and disposal costs.**

The only problem that could be encountered by procuring fuel from DOE- or EPRI-sponsored programs is the need to verify that rod segments were stored under conditions that preserve the fuel characteristics. The MCC would also want to verify the quality assurance of the postirradiation data (such as fission gas release) obtained from these programs for use by the YMPO.

When domestic sources are unable to supply samples for testing of particular fuel characteristics, the MCC may have to purchase fuel from foreign sources. For example, no U.S. source has been identified to supply samples of a high burnup fuel (>65 MWd/kgM). If utilities eventually decide to increase the average core burnup to about 60 MWd/kgM, it will be important to have tested a fuel with high enough burnup to cause grain restructuring (which is representative of peak burnup fuel). The cost of obtaining samples from foreign sources has not been evaluated. Further investigation of possible U.S. sources should be performed before foreign sources are pursued.

Of the three options, the MCC recommends obtaining fuels from DOE, EPRI, or other vendor programs that can provide small quantities of partially or fully characterized fuels. Foreign sources for small quantities of fuel should be pursued as a second option. Acquiring spent fuel from U.S. utilities should only be considered when the first two options cannot provide a fuel with the desired characteristics for testing because of the long lead-time, high cost, and waste liability.

4.0 Conclusions

As knowledge of spent fuel performance increases, new fuel types will be required. To meet the new spent fuel sample requirements of the YMPO testing program, a minimum of five additional spent fuel ATMs should be acquired for testing. These are:

- high burnup fuel (> 60 MWd/kgM) with significant rim effect
- two low fission gas release fuels [$< 10\%$ fgr, high burnup (> 50 MWd/kgM) and low burnup (< 10 MWd/kgM)]
- burnable poison fuel (5 to 8 wt% Gd_2O_3) with relatively high burnup
- high fission gas release BWR fuel.

These fuels should be obtained from DOE, EPRI, or other vendor programs that can provide small quantities of partially or fully characterized fuels. Foreign fuel sources should also be investigated if small quantities are required of a special type of fuel unavailable in the U.S. This spent fuel acquisition plan is recommended for annual review as more spent fuel performance data are obtained. A review will ensure that the MCC interfaces information with the spent fuel performance research and engineering program and that the MCC provides the necessary fuels for the performance testing activities.

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