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LWR Spent Fuel Approved Testing Materials for Radionuclide Release Studies

J. O. Barner

January 1984

Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
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Richland, Washington 99352

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ABSTRACT

Criteria are defined for the selection of light water reactor (LWR) spent fuels for use as MCC-Approved Testing Materials (ATMs) in radionuclide dissolution and interaction studies. Fuel-related characteristics affecting the release of radionuclides from spent fuel are reviewed and their pertinency evaluated. ATM spent fuel criteria are defined and classes of ATM spent fuels are determined. The available inventory of government-owned LWR spent fuel is identified and current plans for acquisition by the MCC are summarized. The characterization data to be supplied with the spent fuel ATMs are also described.

SUMMARY

This report defines four spent fuel classes for Approved Testing Materials (ATMs) for use in repository-related predisposal tests that measure the radionuclide dissolution characteristics of spent fuel. An approach for selecting the criteria for ATM spent fuel is described which depends upon fuel-related characteristics. Activity by the Materials Characterization Center (MCC) to acquire suitable spent fuel for ATMs is summarized, and fuel characterization procedures are outlined.

Fuel-related characteristics expected to be important with respect to dissolution behavior are 1) fuel material composition, e.g., UO_2, UO_2-PuO_2, UO_2-Gd_2O_3, etc., 2) fuel form, e.g., pellets or packed-particle fuel, 3) burnup level, 4) the extent of radionuclide release from the fuel during irradiation, 5) the decay time, and 6) the mechanical condition of the fuel rod prior to obtaining specimens. The criteria for classification of ATM spent fuels are based upon these important characteristics. The most important class of ATM spent fuel for current testing is a moderate burnup, low-releasing (in-reactor), originally solid-pellet UO_2 fuel. ATM specimens prepared from intact rods are recommended.

The currently-available, government-owned spent fuels from the H. B. Robinson and Turkey Point reactors were determined to meet the criteria for the currently most important ATM spent fuel class. H. B. Robinson spent fuel is currently being obtained by the MCC.

The generic characterization information expected to be supplied with MCC-ATM spent fuel includes data on 1) classification of the ATM, 2) radionuclide inventory, 3) radionuclide distribution within the specimen, and cladding condition. Additional special characterization will be conducted on a "need to know" basis.

1.0 CONCLUSIONS

As a result of this study the following conclusions are made:

1) Criteria for the selection of ATM spent fuel for studying radionuclide dissolution and interaction behavior can be defined by considering the original as-fabricated attributes of fuel, the irradiation-induced characteristics of the fuel, and postirradiation storage/handling-related characteristics.

2) For the base-case spent fuel, i.e., originally solid-pellet UO_2 spent fuel from a LWR, the burnup level and the amount of fission gas (products) released from the fuel during irradiation are the primary criteria for classification as ATM spent fuels. This results in a two-by-two matrix of ATM spent fuel classes, i.e., four spent fuel classes, that vary in burnup level and the degree of in-reactor fission product release. In addition, it is preferable that specimens for dissolution/interaction testing be taken only from intact rods, the operating power (LHGR) or degree of fuel pellet fragmentation of the fuel should be considered, and the fuel should be permitted to decay sufficiently to significantly reduce the quantities of short and moderate half-life radionuclides.

3) The government-owned fuels from the H. B. Robinson and Turkey Point reactors are suitable for classification as ATM spent fuel for the moderate burnup, low-releasing (in-reactor), originally solid-pellet UO_2 class of spent fuels. Fuels in this class should be emphasized for current studies of the dissolution/interaction behavior of radionuclides.

4) The radionuclide dissolution/interaction studies should be flexible enough to permit the addition of additional ATM spent fuel classes if the industry commits to high burnup levels or new fuel types.

5) Characterization information expected to be supplied by the MCC with ATM specimens was selected based upon that data necessary to 1) classify the ATM, 2) describe the radionuclide inventory, 3) describe the radionuclide spatial distribution, and 4) describe the general microstructural characteristics of the fuel and cladding. This information will allow classification of the ATM and permit the calculation of radionuclide release fractions from all or a component of the ATM specimen.

2.0 INTRODUCTION

It will be necessary to adequately predict the migration of radionuclides from proposed waste repositories in order to meet federal regulations concerning the release of these radionuclides to the accessible environment. The initial step in any migration sequence is the release of the radionuclides from the stored waste form. Before disposal of waste in a repository, laboratory and/or pilot-scale testing of the waste form will be required to quantify the initial radionuclide releases for use in predictive models. These predisposal tests must utilize material that is typical of the anticipated waste form(s) that will actually be stored in a repository.

The Materials Characterization Center (MCC) has the responsibility to provide reference and testing materials for use in development of the nuclear waste disposal forms, and for repository-related tests in the several National Waste Terminal Storage (NWTS) projects. Approved Reference Materials (ARMs) are provided by the MCC, as specified in some MCC testing procedures, for use in verifying correct application of the procedures. Approved Testing Materials (ATMs) are provided by MCC for use by participating laboratories in conducting radionuclide dissolution tests in at least a portion of their total repository-related test matrix. ARMs and ATMs provide the common basis for systematic evaluation of test results in different laboratories with various test parameters. The MCC's purpose in providing ATMs is to assure that all of the appropriate laboratories have testing materials that are as nearly comparable as possible. The level of certification to be supplied by the MCC for various materials will vary depending on the intended use of the material and the cost of acquiring additional data. While the ARMs provide calibration of test methods between laboratories, the ATMs provide common testing materials for the different laboratories.

Spent light water reactor fuel[a] is being considered by the Department of Energy (DOE) as a waste form. Therefore, one or more spent fuels need to be defined, both as an ATM for comparison of performance with other waste forms, and as a common testing material. However, because of the inherent inhomogeneity of all spent fuels, only a generic characterization of the composition and properties for each specific fuel source can be provided for individual test specimens rather than the "certified" characterization normally associated with homogeneous reference standards. For this reason, the spent fuel selected by MCC will be provided to users as an ATM, accompanied by the most reliable characterization information available.

The objectives of this study are:

- to develop the criteria for classification and selection of ATM spent fuels that, following further characterization, can be used in hydrothermal materials interaction studies to determine the dissolution and release characteristics of the fuel, i.e., the radiological source term,
- to review the inventory of government-owned LWR spent fuel that might be used in predisposal studies, and document those characteristics of the available fuel that relate to the criteria for ATM spent fuel,
- to identify spent fuels suitable for designation as ATMs from the available inventory, and
- to provide definitive guidelines for the spent fuel characterization information that will be provided with the ATMs by MCC.

(a) The term fuel in this report is used in the generic sense, i.e., all the components of a fuel rod, including the fuel material, cladding, and the radionuclide inventory. The term fuel material, of a specific composition, e.g., UO_2, is used for the oxide fuel with its radionuclide inventory.

The main emphasis here is on the spent fuel material itself, with only secondary attention to the cladding. It is recognized that intact cladding will act as a barrier to fuel-material/ground-water contact, and breached cladding has the potential to delay the release of radionuclides from spent fuel. The significance of the delay of radionuclide release from fuel contained in nonbreached or breached rods can be evaluated by specific, probably very long term, tests using specimens specially prepared from the ATMs. This study does not address individual test specimen forms or specific test methods to be applied to the ATMs for evaluation of spent fuel behavior.

3.0 DEFINITION OF ATM SPENT FUEL

This section describes the approach to definition of ATM spent fuel, the fuel-related characteristics that are expected to affect the release of radionuclides from spent fuel, and the ATM spent fuel definition criteria.

3.1 APPROACH

Many different types of spent fuel with possibly differing radionuclide release characteristics may potentially be disposed of in a geologic repository. One approach to the definition of an ATM spent fuel would be to select the fuel with the "extreme" expected release characteristics and design and model the repository as if it were full of this type of fuel. However, if this "extreme" fuel differed significantly in its release characteristics from "typical" fuel and, especially, if the "extreme" fuel comprised only a small fraction of the actual fuel stored in the repository, such an approach would be excessively conservative. A more reasonable, and probably economical, approach is to 1) identify classes of ATM spent fuels which are "typical", yet conservative[a] with respect to expected dissolution characteristics, and classes which are "extreme", 2) perform the necessary predisposal tests and compare the results to ascertain if there are significant differences between classes, and 3) if there are only minor differences between classes, treat the classes as identical, or, if there are significant differences between classes, take this knowledge into account in design of the repository, e.g., limit the amount of "extreme" spent fuel to be stored or design a better repository. This more reasonable approach will be used to define the ATM spent fuel criteria.

(a) "Conservative", as used here, means towards the higher values of a normal distribution of a characteristic, e.g., higher burnup or more fuel pellet fragmentation.

With the exception of a few gaseous, and possibly, volatile isotopes, the radionuclides in spent fuel are in solid form. In order to be released from the spent fuel, the radionuclides initially must dissolve into the available ground water. For significant dissolution to occur, the radionuclides must be available for contact with the water and the extent of dissolution will primarily depend upon the conditions of this environment and the nature of the specific chemical phases of the radionuclides within the fuel. Environmental factors that can affect the dissolution and/or dissolution rate of the radionuclides into the ground water include whether conditions are oxidizing or reducing, the solubility limit for a specific radionuclide, and whether a dissolution/precipitation mechanism is operative. Several fuel-related factors such as the magnitude of the beta/gamma radiation field can be expected to affect the dissolution characteristics of the radionuclides. Some of these factors can be expected to be dominant, while others can be expected to be of little or no importance. In some cases, the magnitude of an individual factor may be important, especially as it may relate to the amount of material to be stored in a repository.

By estimating the relative importance of individual fuel-related factors that may affect the dissolution characteristics of spent fuel and, if necessary, the magnitudes of the factors, the list of factors can be reduced to those which are expected to dominate, and "typical" and "extreme" spent fuel classes can be defined. In the extreme, if the magnitude of a <u>single</u> fuel-related factor is estimated to dominate the dissolution characteristics, a moderate magnitude of the factor would need to be tested for the "typical" condition and a high level for the "extreme" condition (Figure 3.1a). If more than one factor and their magnitudes are expected to be important to dissolution behavior but their relative dominance cannot be estimated, multi-magnitude matrices of the factors are required to define the "typical" and "extreme" conditions. This is shown schematically in Figure 3.1b where the combination of the lower levels of two factors are presumed to represent the majority of the spent fuel that would be placed in a repository and is, therefore, "typical", and the

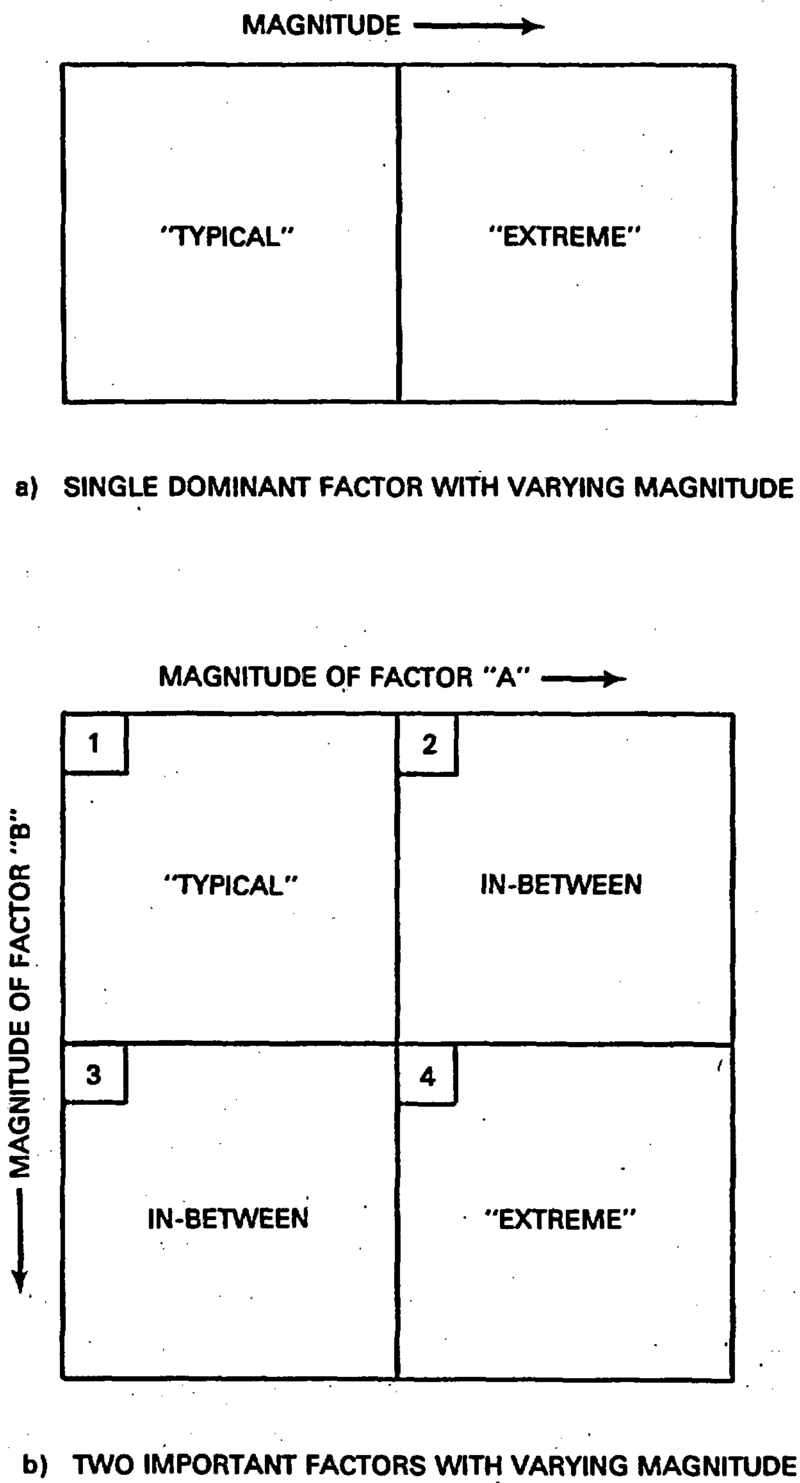

Figure 3.1. Schematic of Factors Affecting Dissolution Characteristics of Spent Fuel

combination of the higher magnitudes is "extreme". For predisposal dissolution testing the "typical" and "extreme" combinations would be tested as ATM spent fuels. If no difference in dissolution behavior between the two combinations was observed, all fuels of the specific type would be treated the same with respect to repository design. If a significant difference was found, the majority of the fuel (box 1) in the repository would be treated as "typical" and the fuel in box 4 would be treated as "extreme". This approach is conservative and, if cost of the repository or some other factor indicated that a more realistic design should be undertaken, the fuel from boxes 2 and 3 could also be tested. In any case, estimating the relative importance of fuel-related characteristics that might affect the dissolution properties reduces the number of spent fuel classes that need to be considered as ATMs during the predisposal tests. The relative importance of fuel-related characteristics that might affect the dissolution of spent fuel is considered in Section 3.2.

In the repository, fuel rods with nonbreached cladding will not permit release of the radionuclides contained from the rod. Therefore, the rods with cladding breaches determine the source term for release and transport of the radionuclides. In consideration of the amount of breached fuel rods in a repository, experience since the inception of commercial nuclear power generation indicates there are two vintages of LWR spent fuel, i.e., that used prior to about 1975 and that used after 1975. Prior to 1975 the in-reactor failure rate of boiling water reactor (BWR) rods was higher than after 1975, primarily because of hydride-induced cladding failures resulting from excess moisture in the as-fabricated fuel rods. Similarly, prior to about 1975 the in-reactor failure rate of pressurized water reactor (PWR) rods was higher than after 1975, in part because of in-reactor fuel densification, cladding collapse, and cladding failure. Fuel rod failure rate data from Bailey and Tokar, 1983, and projected spent fuel inventory data from DOE/NE-0017/2, 1983, were used to perform scoping calculations to estimate which vintage of spent fuel might expose the greater amount of fuel material and radionuclides to the groundwater (Appendix A). If it is assumed that the only fuel material exposed to the groundwater comes from fuel rods that breached in-reactor for the fuel available for disposal in the year 2020, the current vintage (used after 1975) will expose approximately four times more fuel material to the groundwater than will the early vintage fuel. If it is further assumed that during predisposal handling, storage, and packaging that both vintages of fuel experience damage failure at the same rate, an even greater fraction of the current vintage will be exposed to the groundwater than the early vintage fuel, e.g., if the damage failure rate was 0.5%, the current vintage fuel would expose approximately 47 times more fuel than the early vintage fuel. Similar arguments can be made for in-repository failures at a constant rate, with similar conclusions. (The above arguments assume that the geometry of the failures in the two vintages of fuel rods are similar; but assume nothing about rate of transport of the radionuclides from the fuel rod). Of course, it could be argued that the early vintage spent fuel might be more susceptible to ex-reactor failures than current vintage fuel. This assumption would reduce, or even reverse, the exposed-fuel ratio for the different vintages. For instance, it could be argued that there are more incipient cladding defects from moisture or fuel densification effects in the early vintage fuel that might lead to more cladding breaches prior to repository-canister failure. However, this argument could be qualitatively countered by the argument that current vintage fuel is pressurized during fabrication, resulting in higher cladding stresses and a greater susceptibility to stress-corrosion cracking prior to repository-canister failure. Lacking definitive cladding failure rates for the two vintages during handling, storage, packaging, and the early stages of repository disposal, it is expected that the current vintage fuel will dominate the amounts of spent fuel and radionuclides that are available for dissolution and transport. Therefore emphasis is placed on current vintage fuel during the following evaluation.

3.2 FUEL-RELATED CHARACTERISTICS AFFECTING RELEASE OF RADIONUCLIDES FROM SPENT FUEL

There are three categories of inherent fuel-related characteristics that could affect the release of radionuclides from spent fuel: 1) those associated with the as-fabricated fuel; 2) those induced by irradiation; and 3) those associated with postirradiation handling and storage. These fuel-related characteristics are described in the next three subsections and their potential for affecting radionuclide release in a repository is evaluated. The emphasis here is on fuel source-related differences in those characteristics, as well as the magnitude of their effect on repository-related characteristics.

Because of the general lack of information related to the effects of fuel-related characteristics on dissolution and interaction behavior of spent fuel, this evaluation is based upon engineering judgment. It should be kept in mind that the general approach is to attempt to ascertain those characteristics that dominate the dissolution behavior of spent fuel in an effort to reduce the number of testing parameters and to determine if there are major differences between different classes of spent fuel.

With respect to determining the relative importance of each fuel-related characteristic, the following assumptions are made as a base case:

- the fuel is from a LWR, i.e., either a PWR or a BWR,
- the fuel is placed in the repository in a sealed canister,
- when the canister fails, the fuel material from breached fuel rods, the cladding and any radionuclides on the surface of the fuel or cladding are exposed to the groundwater[a],

[a] Note that this assumption says nothing about radionuclide release rates from breached cladding. Evaluation of release rates requires specific test data.

- the fuel material was originally solid-pellet UO_2.

3.2.1 As-Fabricated Characteristics

The as-fabricated fuel-related characteristics that are commonly included in specifications, e.g., ANSI-ASTM C776-76, are listed in Table 3.1. The predicted importance of each characteristic as it may affect the dissolution properties of the radionuclides from spent fuel is noted in the table and is discussed below. The concern here is the difference in dissolution properties which might result from the expected variations in the listed characteristic of as-fabricated fuel obtained from different sources. The characteristic might have a significant absolute effect, of nearly the same magnitude for all fuels because of fabrication quality controls.

- Uranium Content. The base-case material is assumed to be UO_2. The uranium content in spent fuel is essentially invariant in UO_2 at any given burnup.

  Other oxides have been or are being considered for use in LWRs, e.g., UO_2-PuO_2, UO_2-Gd_2O_3 and UO_2-Nb_2O_3. These additions to the UO_2 could affect dissolution characteristics of the fuel materials, especially if they are added by physically mixing the powder feeds and are predominantly located on grain boundaries. Localized attack of the grain boundaries resulting from a composition change would increase the surface area of the fuel material with resultant increased accessibility of the ground water to radionuclides. Fuel additives may be important to the dissolution characteristics.

- Fuel Material Form. The base-case fuel form is assumed to be solid-pellet UO_2. A variety of length-to-diameter (L/D) pellets is contained in commercial fuel from different manufacturers. However, because the longer pellets tend to break transversely, the resultant increased surface area of the broken pellets approximates that of

TABLE 3.1. Predicted Effects Due to As-Fabricated Fuel-Related Characteristics

Characteristic(1)	Expected Significant Effect on Spent Fuel Dissolution	Description of Qualifier
Uranium Content		
UO_2	Base-Case	
UO_2-PuO_2	Yes	
UO_2-Gd_2O_3	Yes	
UO_2 Additive	Yes	
Fuel Material Form	Yes	
Open Porosity of Fuel	Yes	
Pellet-Cladding Gap	Yes	
Gas Content of Fuel	Possibly	High N_2 could affect dissolution and will affect ^{14}C production
Fuel Material Density	Possibly	If primarily open porosity
Fuel Rod Dimension	Not directly	Dissolution more relatable to LHGR
Pressurization Level	Possibly	If LHGR high enough for high fission gas release in low pressure rod
BWR vs. PWR	Not directly	Rod characteristics and power history are important
Grain Size of Fuel Material	Not significantly	
Cladding-Type/Mechanical Condition	No	
Impurity Content of Fuel Material	No	
Stoichiometry of Fuel Material	No	
Moisture Content of Fuel	No	
Isotopic Distribution of Uranium	No	
Equivalent Boron Content of Fuel	No	
Pellet Defects	No	

(1) Characteristics commonly listed in fuel specifications.

fuel pellets of shorter length. Thus, the L/D of the pellet should not affect the dissolution properties significantly. Annular pellets only slightly increase the surface area of fuel material and, therefore, can be considered the same as solid-pellets.

Fuel forms significantly different from homogeneous solid-pellet fuel could affect dissolution characteristics by drastically changing the surface area, e.g., sphere-pac or vipac fuel, or the radionuclide distribution, e.g., duplex pellets with radially-graded enrichment.

- Open Porosity of Fuel. If a significant portion of the porosity in the fuel material is open porosity, i.e., the porosity interconnects to the pellet surface, it has the potential for affecting the dissolution properties by increasing the effective surface area of the fuel material. However, the effect on dissolution of this initial open porosity is difficult to evaluate because the amount of open porosity is usually significantly reduced by in-reactor densification in this type of fuel. Current vintage fuels usually have a very low amount of open porosity, i.e., less than 0.5%.

- Pellet-Cladding Gap. The fuel-cladding gap affects the fuel operating temperature, i.e., the larger the gap the higher the temperature. Higher temperatures can possibly increase the release of radionuclides from the fuel material during operation, and the dissolution characteristics of the spent fuel. Fuel materials that release significant quantities of fission products during irradiation are discussed in Section 3.2.2.

- Gas Content of Fuel. Gases that might be included in fuel rods at the time of manufacture include helium, argon, hydrogen, oxygen, carbon monoxide, carbon dioxide, and nitrogen. All except nitrogen are relatively benign in that they are inert or combine rapidly with the cladding during irradiation. Nitrogen has the potential for dissociation in a gamma field and to form nitric acid in the presence of water. However, most of any nitrogen that might be present in the fuel-rod/canister package would be expected to be rapidly dissipated with breach of the canister and, therefore, should be of no importance with respect to the dissolution characteristics of the fuel.

The level of nitrogen gas in the as-fabricated fuel selected as an ATM should be typical of that present in a normal population of fuel rods in order to calculate the ^{14}C inventory.

- Fuel Material Density. The as-fabricated density of the fuel material and its propensity for additional in-reactor sintering should have little effect upon the dissolution characteristics if the porosity contained within the fuel is closed porosity (see third bulleted item).

- Fuel Rod Dimensions. Over the past few years fuel rods have generally become smaller in diameter and the fissile inventory in a fuel assembly has been maintained by increasing the number of rods within the assembly, e.g., BWR assemblies have gone from arrays of 5 x 5 or 6 x 6 to 7 x 7 or 8 x 8, and PWR assemblies have gone from arrays of 15 x 15 to 16 x 16 or 17 x 17. Associated with these size changes has been a general decrease in the linear heat generation rate (LHGR) during irradiation. The fuel rod size, per se, in assemblies containing similar amounts of fissile material should have very little effect on the dissolution characteristics of spent fuel. However, the LHGR, and resultant fuel operating temperature, should have an effect (see Section 3.2.2).

- Pressurization Level. The pressurization level affects in-reactor performance of the fuel in two ways. First, it retards/reduces cladding creepdown, and thereby tends to maintain larger fuel-cladding gaps, higher fuel temperatures, and higher radionuclide release from the fuel material during irradiation. Second, pressurization minimizes the degradation of the fuel-cladding gap conductance caused by fission gas release, which tends to retard/reduce any increases in fuel temperature associated with fission gas release, including thermal feedback.[a] Between these mechanisms,

(a) Thermal feedback is a situation where fission gas release degrades the gap conductance to a point where higher fuel temperatures trigger an increased fission gas release rate. This break-away situation continues until the fission gas is the major fraction of the gas contained within the gas volume of the rod.

which have opposite temperature-release effects, the second is thought to predominate, especially in PWR rods. This is not to say that thermal feedback and high fission product releases from the fuel material cannot occur in pressurized rods during irradiation. Other factors, e.g., operating LHGR, must be considered. Fuel materials that release significant quantities of fission products during irradiation are discussed in Section 3.2.2.

- BWR vs. PWR. There are certain attributes of BWR and PWR fuels that can affect the operating characteristics of fuel rods, e.g., fuel-cladding gap, rod dimensions, and pressurization level. However, there are no as-fabricated fuel-related characteristics, per se, that are different enough to differentiate between the two types of fuel from the standpoint of dissolution of radionuclides.

- Grain Size of Fuel Material. The grain size of the fuel material has the potential for affecting the dissolution/leaching properties of spent fuel, especially if the material is sensitive to grain boundary attack by the leachant. This may be important for fuel materials with additives (discussed earlier) or those that have experienced significant fission product migration to the grain boundaries because of high temperature operation (Section 3.2.2). Most commercial UO_2 fuel materials have grain sizes within a relatively narrow range of from 5 to 15 μm. Therefore, grain size variation, per se, for the base-case UO_2 probaby will not significantly influence the dissolution properties.

- Cladding-Type/Mechanical Condition. Most LWR fuel rods utilize either Zircaloy-2 (BWR) or Zircaloy-4 (PWR) for the cladding. Both the annealed and stress-relieved mechanical conditions have been used. A significantly lesser quantity of LWR fuel with stainless steel cladding has been used. The as-fabricated type or mechanical condition of the cladding is of only secondary importance to the dissolution characteristics of the radionuclides from breached fuel rods; of course different claddings may affect the transport characteristics of specific radionuclides after release from the fuel.

- Impurity Content of Fuel Material. Fuel materials are usually specified to be very pure, i.e., the trace impurities are generally limited to small amounts (ANSI/ASTM C 776-76). In addition, the permitted range for individual impurities is generally quite narrow. Therefore, differences in trace impurity levels, except for nitrogen (p. 15 and 16), between different fuel materials of the same type should not cause significant differences in dissolution characteristics.

- Stoichiometry of Fuel Material. The stoichiometry, i.e., oxygen-to-metal (O/M) ratio, has the potential for affecting the dissolution properties of fuel. However, almost all commercial LWR fuel is manufactured with an O/M ratio of 2.00, or is slightly hyperstoichiometric. The allowable range should not promote significant differences in dissolution characteristics between different fuel materials of the same type.

- Moisture Content of Fuel. Current vintage LWR fuel has very low moisture content (ANSI/ASTM C 776-76). The as-fabricated moisture content is rapidly reduced in-reactor by hydriding and oxidation of the cladding. Thus, after irradiation of current vintage fuel the moisture is no longer present in a form which could affect the dissolution characteristics of fuel material. Similarly, because of the expected similar degrees of hydriding and oxidation of the inner surface of the cladding of different current-vintage fuels, the as-fabricated moisture content should not affect the interaction behavior between the radionuclides and the repository. The latter conclusion may not be true if the interaction behaviors of early and current vintage fuels are compared because of the expected greater oxidation and hydriding of the inner surface of the cladding in the early vintage fuel.

- Isotopic Distribution of Uranium. The isotopic distributions of the uranium used in LWR fuel materials are generally within very limited bounds and are very similar. Thus, any differences in isotopic distribution between different fuel materials of the same type should have no effect on dissolution properties.

- Equivalent Boron Content of Fuel. This characteristic is important for reactor operation in that it combines the cross-sections of impurities into the cross-section of an equivalent amount of boron. It should have no effect, per se, on dissolution characteristics.

- Pellet Defects. Fuel surface chips can affect the operating temperature of the fuel by increasing the effective pellet-cladding gap and resultant radionuclide release during operation. However, fuel specifications do not permit extremely large surface defects which would affect significant volumes of fuel material in any particular fuel rod. In addition, the inclusion of pellets with allowable defects in fuel rods would be expected to be random and any particular rod that might be used for a spent fuel ATM would be just as likely to contain pellet defects as any other rod. Therefore, pellet defects are not expected to significantly affect the dissolution properties.

3.2.2 Effects of Irradiation on Spent Fuel Characteristics

Irradiation characteristics that might affect the dissolution properties of radionuclides from spent fuel are listed in Table 3.2. The importance of each characteristic is noted in the table and discussed below.

- Burnup Level. The burnup level is expected to influence the dissolution properties of the radionuclides from spent fuel because 1) burnup determines the available inventory of the radionuclides and the radiological source term for a repository, 2) burnup affects the

TABLE 3.2. Predicted Effects Due to Irradiation History

Irradiation Characteristic	Expected Significant Effect on Spent Fuel Dissolution
Burnup-Level	Yes
Peak LHGR	Yes
BWR vs. PWR	Possible
High-Releasing vs. Low-Releasing (In-Reactor)	Yes
Crud Deposits	No

radial distribution of the radionuclides within the fuel pellet, and 3) burnup could conceivably affect the residual stress (energy) state within spent fuel. The first point is obvious in that the fission products and transuranic elements are generated within the fuel material and the level of burnup determines the amount of radionuclides that is available for dissolution in a repository. With respect to the second point, there is a natural radial distribution (segregation) of fission products and transuranics that occurs in the fuel. The thermal and epithermal flux levels are higher near the outer edge of the fuel with resultant higher levels of fission products and transuranics in this region. This difference in concentration of radionuclides between the outer region of the fuel material and the inner region increases with burnup. Any preferential dissolution of the outer region of the fuel material would result in higher releases of radionuclides. Thirdly, the outer edge of the fuel may be more accessible to the ground water and may have different dissolution characteristics because of the higher radionuclide loading and higher residual stress level in the atomic structure of the fuel material. (It should be noted that it can be argued that extensive fuel cracking may permit ready access of the groundwater to the interior regions of the fuel material, thus minimizing the effects of the second and third points. However, if there are significant differences in behavior between the inner and outer regions of the fuel material, caution should be taken in selecting fuel fragments for those types of tests that use less than a full cross-section of the fuel material from a fuel rod.) Considering all the points, the burnup level can be expected to influence the dissolution characteristics of spent fuel.

- Peak Linear Heat Generation Rate. The peak linear heat generation rate attained in the fuel during operation can affect dissolution characteristics because the peak LHGR determines 1) the radial temperature gradient in the fuel, the resultant thermal stress levels, and the degree of fuel pellet fragmentation, and 2) depending upon when it occurs, the peak temperatures and any resultant radionuclide release from the fuel material during irradiation. The first point is important because the degree of fragmentation affects the surface area that is available for dissolution of radionuclides from the spent fuel. The second point is important because, if the peak LHGR occurs late-in-life and is sufficiently high, significant release of radionuclides from the fuel material can occur during irradiation; the radionuclides can then migrate to cooler regions of the fuel rod and be available for preferential dissolution. (See second bulleted item below). High temperatures can also result in agglomeration of certain types of fission products, e.g., ruthenium, rhodium, palladium, into nuggets within the fuel material which also may affect dissolution properties.

- BWR vs. PWR. From an operational standpoint there are both similarities and differences between BWR and PWR fuel. The current discharge burnup is usually a little higher in PWR fuel than in BWR fuel, 35 MWd/kgM vs. 30 MWd/kgM, respectively. Although design peak LHGRs are similar for both fuels, BWR fuels often have higher lifetime-average peak-pellet LHGRs and a higher propensity for release of radionuclides from the fuel material during operation. Because current PWR fuel is usually pressurized to significantly higher levels than BWR fuels, PWR fuel has less propensity for thermal feedback than BWR fuel. Considering all the operational characteristics, BWR fuel has a greater propensity for high fission product release during irradiation than PWR fuel. (See next bulleted item).

- <u>High Releasing vs. Low Releasing Fuels (In-Reactor)</u>. Throughout the remainder of this document the terms "high-releasing (in-reactor)" and "low-releasing (in-reactor)" will be used. The term "high-releasing (in-reactor)" refers to fuel material that exhibits a significant amount, e.g., >10%, of fission gas release from the fuel material during irradiation. It is assumed that significant quantities of the volatile fission products are concurrently released from the fuel material and migrate to cooler regions of the fuel rod and are available for preferential dissolution. The release of these fission products is predominantly from the hotter central portion of the fuel material. Usually, fuel material restructuring, porosity agglomeration and, often, noble metal fission product agglomeration, occur concurrently in the central portion of high-releasing (in-reactor) fuel material. "Low-releasing (in-reactor)" fuels are those fuels which have not been exposed to conditions that are sufficient to cause significant release of fission gases (products) during irradiation, e.g., <1% release.

  It has been shown that several as-fabricated characteristics, i.e., open porosity, fuel rod dimensions, pellet-cladding gap, and pressurization level, and the operating fuel rod LHGR can affect the degree of fission gas and fission product release from the fuel material and their subsequent segregation to cooler regions in the fuel rod. It is reasonable to group these characteristics into two categories that address their combined effect on fission product release and the resultant availability of the radionuclides for dissolution in a repository, i.e., high-releasing (in-reactor) and low-releasing (in-reactor). The high-releasing (in-reactor) fuel is expected to enhance dissolution of the radionuclides because of the greater access of the radionuclides to the ground water; high releasing (in-reactor) fuel can be classified in the "extreme" category (Section 3.1). The low-releasing (in-reactor) fuel, which will comprise the bulk of the available fuel for disposal, can be classified in the "typical" category. This categorization reasonably equates several individual variables that may affect the dissolution characteristics and permits a reduction in the number of variables/characteristics that must be considered for defining classes of ATM spent fuel. A similar categorization has been suggested by Jenson, et al, 1982, for characterization of spent fuel.

- Crud Deposits. The deposited crud on the outer surface of fuel rods could be a source of radionuclides for dissolution. The isotopes ^{58}Co, ^{60}Co, and ^{65}Zn are the usual radioisotopes found in crud. The thickness of the crud deposit is dependent upon the reactor coolant chemistry, and therefore, the specific reactor. BWRs tend to have greater, and usually more spallable, crud deposits than PWRs. The commonly found radioisotopes have moderate half-lives and will be of little concern from the standpoint of dissolution of the crud in a repository. Very thick, or very porous, crud can affect fuel operating temperatures (see previous bulleted item).

3.2.3 Storage/Handling-Related Characteristics

Fuel-related characteristics associated with postirradiation storage and handling that might affect the dissolution characteristics of spent fuel are listed in Table 3.3. The importance of each characteristic is noted in the table and discussed below.

- Decay Time. The length of time since discharge of the fuel from the reactor (the decay time) affects 1) the inventory of radionuclides, especially the inventory of short and moderate half-life isotopes, early in the decay period and the beta-gamma to alpha ratio late in

TABLE 3.3. Predicted Effects Due to Storage/Handling-Related Characteristics

Characteristic	Expected Significant Effect on Dissolution
Decay Time	Yes
Rod Condition	Yes
Unusual Incidents	Yes

the decay period, and 2) the amount of decay heating, which affects the temperature of the spent fuel in the repository. With respect to the inventory, the amount of a specific radionuclide that can be taken into solution can sometimes be affected by the presence of an element/compound that has similar dissolution properties. The presence of abundant short half-life isotopes can also interfere with radiocounting results for long half-life isotopes during analysis of experimental data. Thus, those isotopes with short half-lives, for which the concentration is changing rapidly with decay time and which could influence dissolution of chemically similar elements, should be permitted to decay to low levels before a spent fuel is used for predisposal testing. A reasonable decay time might be five years or more before use in tests. This judgment concerning decay time is based upon a balance between using current vintage fuel and permitting significant beta-gamma activity decay. It is not intended that this judgment be based on current estimates of the minimum age of the spent fuel that might be placed in a repository, i.e., ten years with significant possibility of increasing (DOE/NE-0017/2). Fuel that has decayed five years produces only twice as much heat (activity) as fuel that has decayed ten years [approximately 4×10^{-4} and 2×10^{-4} of the original shutdown decay heat, respectively (S. Glasstone and A. Sesonske, 1967)]. However, if fuel with a short decay time is utilized for testing, the dissolution characteristics of the short-lived isotopes must be taken into account.

With respect to decay heating during spent-fuel predisposal testing, this is primarily a testing parameter. Unless shown otherwise, the testing should be done at the temperature/pressure conditions anticipated for the repository.

- Rod Conditions. If it could be shown that only fuel rods that had failed in-reactor or during wet storage were the only rods that would release radionuclides during the lifetime of a repository, only breached rods during these periods would be of concern for predisposal testing. Lacking this information, it is reasonable to assume that the majority of the radioisotopes that will be available for dissolution will come from rods that failed or were damaged during storage (dry), packaging, or after placement in the canister. For this assumption, specimens for use as ATMs are most suitably obtained from intact fuel rods.

Previously cut and stored spent fuel rods may no longer be representative of the class of spent fuel from which they came, because of uncontrolled postirradiation changes. This is especially true if the fuel rod pieces have been stored for long periods of time under unknown environmental conditions. Usually as a minimum, the sections of fuel rods which have been cut apart will have been exposed to oxidizing conditions, i.e., air and/or moisture. Such exposure of itself may not be grounds for rejection as test material; freshly sectioned specimens for predisposal testing will also presumably be exposed to air and, due to the radiation field in a repository, actual spent fuel placed in a repository will probably be exposed to an oxidizing environment. Therefore, it might be argued that previously cut spent fuel rods that have been exposed only to air are adequate for predisposal testing. However, previously cut spent fuel rods which have remained unprotected for long periods, or which might have been exposed to hydrocarbons, e.g., lubricants used during cutting, or to other chemicals commonly found in hot-cells, e.g., acids, cleaning agents, etc., obviously would not be suitable for predisposal dissolution tests. If previously cut and stored spent fuel must be used for predisposal tests, care should be taken to ascertain the environment to which it has previously been exposed.

- Unusual Incidents. Unusual incidents that might occur during handling and storage could affect the dissolution characteristics of spent fuel. Probably the most important incident would be the occurrence of a postirradiation temperature excursion resulting from inadequate cooling. If the temperature rise was sufficient to cause significant radionuclide release from the fuel material, the resultant radionuclide migration and segregation would effectively place such a fuel in a higher releasing (in-reactor) category, as described in Section 3.2.2. Temperatures significantly in excess of 600°C would be required to exceed the minimum fuel material temperatures that occur during irradiation. Such an excursion is very unlikely, but it would be imprudent to use a fuel material exposed to storage temperatures in excess of 600°C for predisposal testing. With respect to unusually high temperatures that might affect the properties of zircaloy cladding, the cladding temperature over extended periods, e.g., up to two weeks, should be maintained at less than 300°C in order to prevent recovery of irradiation-induced damage (Einziger and Fish, 1982).

3.3 ATM SPENT FUEL CRITERIA

Based upon the approach described in Section 3.1 and the fuel-related characteristics that might affect the release of radionuclides described in Section 3.2, the criteria for classification of ATM spent fuel(s) for predisposal testing are:

- Fuel Material Composition. UO_2 is the base-case spent fuel material composition. If the composition of other fuels that might be placed in a repository differs significantly from UO_2, additional ATM spent fuel classes should be determined for these variants.

- Fuel Material Form. The base-case spent fuel form is solid-pellet UO_2. If fuel material forms that might be placed in a repository differ significantly from solid-pellet UO_2, additional ATM spent fuel classes should be determined for these forms.

- Burnup Level. The burnup level may affect dissolution properties. Therefore, both moderate burnup spent fuel and high burnup spent fuel should be tested to determine if there is a significant burnup effect upon dissolution properties.

  It is recognized that the inventory of spent fuel available for disposal will contain fuel with burnup levels less than the design discharge levels. However, it can be expected that the majority of the fuel assemblies will be near the design discharge level, Therefore, for purposes of these criteria, "moderate burnup" is defined as 25 to 35 MWd/kgM, the currently typical reactor discharge burnup level. "High burnup" is defined as >40 MWd/kgM for BWR spent fuel and >45 MWd/kgM for PWR spent fuel, the current design discharge levels for lead test assemblies.

- High-Releasing vs. Low-Releasing (In-Reactor) Spent Fuels. Low-releasing (in-reactor) spent fuels should be considered "typical" of the majority of the spent fuel that will be placed in a repository. High-releasing (in-reactor) spent fuels are probably the "extreme" type of spent fuel that will be placed in a repository in significant quantity. Both high- and low-releasing (in-reactor) spent fuels should be tested to determine if there is a significant difference in their dissolution characteristics.

  For purposes of these criteria, low-releasing (in-reactor) spent fuel is defined as fuel that has released <1% of the gaseous fission products. High-releasing (in-reactor) spent fuel is defined as fuel that has released >10% of the gaseous fission products. In order to accentuate dissolution behavior differences, the high-releasing (in-reactor) ATM spent fuel selected for testing should have as high an in-reactor release as is practical to obtain.

- Peak LHGR. In order to be conservative with respect to the dissolution behavior of the "typical" low-releasing (in-reactor) spent fuel, fuels should be selected that have operated at LHGRs sufficient to promote significant amounts of fuel material fracture, yet low enough to prevent excessive radionuclide release (in-reactor). The important point for this criterion is the surface area exposed by fuel fragmentation. Therefore, in lieu of knowledge of specific operating LHGRs, fuels that typically exhibit several (≥6) radial cracks in transverse cross-sections of fuel pellets can be selected.

- Decay Time. Spent fuels that have cooled for at least five years are preferable for testing of dissolution behavior. If fresher fuels are used, the effect of dissolution characteristics of the short and moderate half-life isotopes should be considered when evaluating the test results.

- Rod Conditions. It is preferable that reference specimens for dissolution testing be taken from intact fuel rods. If previously cut spent fuel rods are used they must not have been exposed to environments that could affect the test results.

- Unusual Incidents. Specimens for dissolution testing should not be taken from spent fuel that has been exposed to unusual environmental conditions unless it can be verified that such exposure would have no significant affect upon the test results.

4.0 CLASSES OF ATM SPENT FUEL

The criteria for definition of ATM spent fuel for predisposal testing identified in Section 3.3 indicate that a single ATM spent fuel will not suffice unless the results of tests indicate that there is no difference in behavior between classes of spent fuel. Fuel material composition, fuel material form, burnup level, high releasing and low releasing (in-reactor) fuel and the peak LHGR achieved during commercial irradiation (or degree of fuel material fragmentation) are expected to significantly affect dissolution characteristics of the radionuclides. The following two subsections describe the classes of ATM spent fuel for the base-case UO_2 fuel, in detail, and, to a lesser extent, other ATM spent fuel categories, respectively.

4.1 THE BASE-CASE CLASSES OF ATM SPENT FUEL

The base-case spent fuel is originally solid-pellet UO_2 fuel from a PWR or a BWR. The criteria for this base-case spent fuel can be combined in a two-by-two matrix of classes of ATM spent fuel (Figure 4.1). The class in box 1 is "typical", low releasing (in-reactor), originally solid-pellet UO_2 fuel material that has operated at LHGRs sufficient to produce significant fragmentation of the fuel pellets. The class in box 4 is "extreme-case", high-releasing (in-reactor) originally solid-pellet UO_2 fuel material. Referring to the approaches described in Section 3.1, if a repository were to be designed for the "extreme" case, only the ATM spent fuel in box 4 (Figure 4.1) would be tested. If only an insignificant fraction of high-burnup, high-releasing (in-reactor) spent fuel was to be stored in a repository, the tests would only need to use fuel from boxes 1 and 3. If the nuclear industry was currently committed to high burnup fuel and the repository were to be designed prudently with a large fraction of "typical" spent fuel and a small, but significant, fraction of "extreme" spent fuel, the tests would use fuel from both boxes 2 and 4. However, it currently appears that there will be a mixture of moderate burnup fuel with

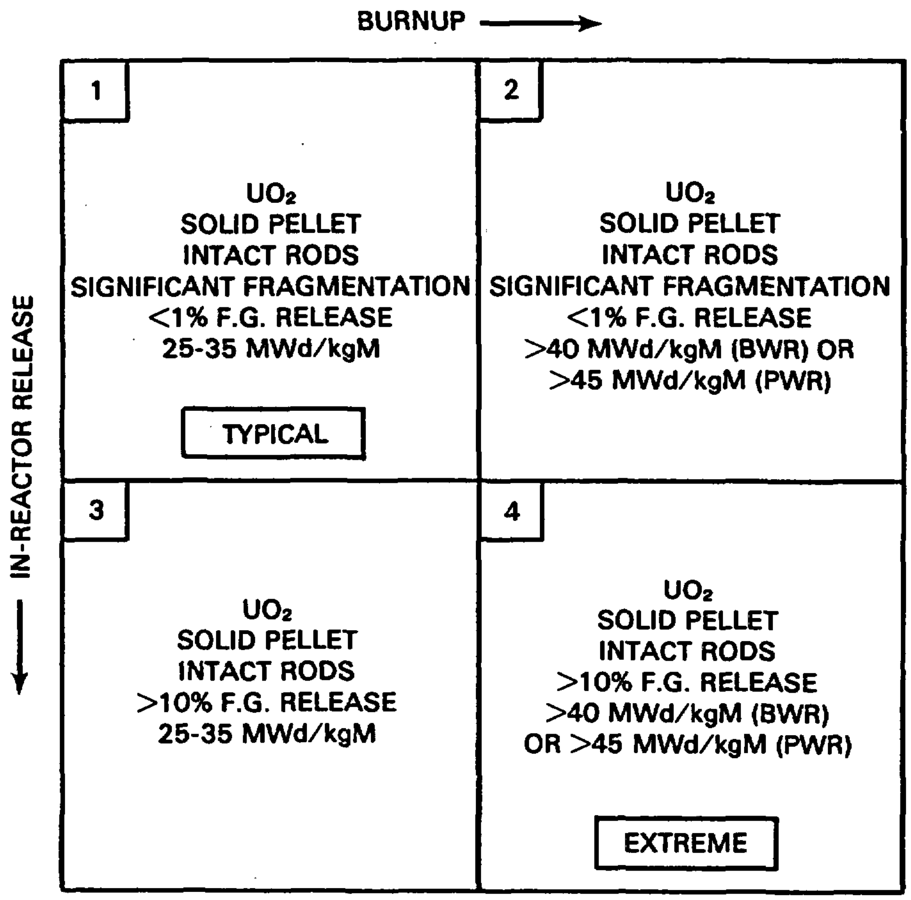

Figure 4.1. Classes of Base-Case UO_2 ATM Spent Fuel (Fragmentation is not listed for boxes 3 and 4 because operating conditions sufficient to cause high release will cause adequate fragmentation).

a lesser quantity of high burnup fuel for placement in a repository. Therefore, the classes of ATM fuel that should be tested are those in boxes 1 and 4.

It should be noted that if there is a general commitment on the part of the nuclear industry to high burnup fuel in the near future, the majority of this fuel material will be UO_2 with a lesser quantity, <5%, of UO_2-Gd_2O_3 fuel. In this event, significant quantities of high burnup fuel could be stored in a repository. However, with the lack of a current commitment on the part of the nuclear industry to high burnup fuel the optimum choice for the emphasis on current testing is fuel from box 1, i.e., moderate burnup, originally solid-pellet UO_2 that has been significantly fragmented and has low fission gas (product) release.

4.2 OTHER ATM SPENT FUELS

As discussed above, a high-burnup UO_2 spent fuel ATM may be required if the nuclear industry commits to utilization of high burnup fuel. If the dissolution properties of UO_2-Gd_2O_3 fuel are significantly different from high burnup UO_2, a UO_2-Gd_2O_3 ATM spent fuel also may be required. The other possible fuel materials, e.g., UO_2-PuO_2 or UO_2-Nb_2O_5, and fuel forms, e.g., sphere-pac fuel, may also require ATM classes depending upon their usage. The program to study the dissolution properties of spent fuel must be flexible enough to accommodate the addition of other ATM spent fuel classes as any changing usage of fuels of varying types may dictate.

5.0 INVENTORY OF SPENT FUEL

In inventorying the available spent fuels for possible use in dissolution tests, emphasis was placed on government-owned material and on privately-owned material that was associated with government-sponsored programs. This emphasis was made because government-owned material can probably be obtained most readily, and because, among the fuel assemblies of commercially-irradiated spent fuel, those involved in government-sponsored programs are often better characterized. While the criteria for definition of ATM spent fuel do not directly address the characterization requirements, the spent fuel ATM needs to be well characterized in order to understand and evaluate the results of the dissolution tests. Table 5.1 lists the currently available intact fuel rods and the currently available characterization references for the fuel rods that 1) are currently available and government owned, 2) are currently available from government-sponsored programs, or 3) are in-reactor and are from government-sponsored programs. The currently available government-owned spent fuels are typically at moderate burnup, are low-releasing (in-reactor), and have decayed for several years. The listed currently available and in-reactor spent fuel rods that come from government-sponsored programs are part of the U.S. Department of Energy Extended Burnup Programs. They will be at high burnup; their release characteristics are not known, and the decay time will be very short. As described in the next section some of the currently available spent fuel with adequate decay time are good candidates for ATM spent fuels for box 1 (Figure 4.2). The high burnup fuels may be adequate for testing for fuels in boxes 2 and 4 (Figure 4.2) after sufficient decay time has accumulated.

Of course, currently privately-owned fuel could also be considered for use as spent fuel ATMs, e.g., fuel from Commonwealth Edison's Zion reactor that is being characterized under the sponsorship of the Electric Power Research Institute. However, use of this type of material will require the approval of both the fuel vendor and the utility.

TABLE 5.1. Summary of Intact Rods Potentially Available for ATMs

Fuel Source, Identification	No. of Assemblies or (Rods)/Type	Nominal Burnup, MWd/kgM	High/Low Releasing (In-Reactor)	Location[1]	Characterization, Type/Reference
I. Inventory as of February 1983--Government-Owned[2]					
Turkey Point, B17	(196)/PWR	27	Low	BCL	Rod description (general); power history (general); NDT, sipping, visual, metrology, and gamma scanning./HEDL-TME-79-68. Destructive PIE: fission gas release, cladding metallography, fuel ceramography, autoradiography, burnup, hydrogen, and microprobe./ HEDL-TME-80-85.
Turkey Point, D01	(2)/PWR	27	Low	BCL	Same as above.
Turkey Point, D04	(4)/PWR	27	Low	BCL	Same as above.
Turkey Point	(5)/PWR[3]	27	Low	BCL	Same as above.
Turkey Point,	16/PWR	25-27	Low	EMAD	Same as above.
H. B. Robinson, B0-5	(1)/PWR	30	Low	BCL	Overall characterization: rod description (general); burnup, power history; transportation/ storage history; PIE, visual, profilometry, gamma scanning, eddy current, fission gas release, cladding metallography, and fuel ceramography./ NUREG/CR-2871 (HEDL-TME-82-27).
H. B. Robinson	(4)/PWR[4]	30	Low	BCL	Same as above.

TABLE 5.1. Summary of Intact Rods Potentially Available for ATMs (con't)

Fuel Source, Identification	No. of Assemblies or (Rods)/Type	Nominal Burnup, MWd/kgM	High/Low Releasing (In-Reactor)	Location[1]	Characterization, Type/Reference
H. B. Robinson, BO-5	(∿130)/PWR	30	Low	EG&G	Same as above.
Peach Bottom 2, PH462	(39)/BWR	12	Intermediate	EG&G	Overall characterization: rod description (general); burnup; power history (general); PIE visual, eddy current, fission gas release, cladding metallography, and fuel ceramography./ NUREG/CR-2871 (HEDL-TME-82-27). (Some UO_2-Gd_2O_3 rods).
Peach Bottom 2, PH006	(∿45)/BWR	9	Intermediate	EG&G	Presumably similar to above.
Shippingport, 0074	(116)/PWR	18	Low	SRP	Overall characterization: rod description (general); power history (general); storage; PIE, visual, gamma scanning, eddy current, profilometry, leak testing, fission gas release, burst tests, cladding metallography, fuel ceramography, hydrogen, and burnup./ PNL-3921. (Short rods, blanket material).
Shippingport, 0551	(11)/PWR	4	Low	SRP	Presumably similar to above with much lower burnup.
Dresden, UN0064	(36)/BWR	24	Unknown	EG&G	No reference.
Dresden, E00161	(∿30)/BWR	20	Unknown	EG&G	No reference.

TABLE 5.1. Summary of Intact Rods Potentially Available for ATMs (con't)

Fuel Source, Identification	No. of Assemblies or (Rods)/Type	Nominal Burnup, MWd/kgM	High/Low Releasing (In-Reactor)	Location[1]	Characterization, Type/Reference
II. Irradiation Completed--Government-Sponsored--Possibly Available[5]					
Oconne 1	4/PWR	40	Probably Low	Reactor Owner	Characterization in progress.
Fort Calhoun	20/PWR 1/PWR	33 52	Probably Low Probably Low	Reactor Owner	Characterization in progress.
Big Rock Point	(59)/BWR	39	Probably Low	Reactor Owner	Characterization in progress.
Monticello	5/BWR 2/BWR 2/BWR	36 42 46	Probably Low, but Possibly High	Reactor Owner	Characterization in progress.
Surry 2	1/PWR	43	Probably Low	Reactor Owner	Characterization in progress.
III. Irradiation in Progress--Government-Sponsored--Possibly Available[6]					
Oconee 1	1/PWR	50 (1983)	Probably Low	In-Reactor	Characterization after irradiation.
ANO2	1/PWR	25 (1984)	Probably Low	In-Reactor	Characterization after irradiation.
Oyster Creek	4/BWR	34 (1983)	Probably Low, Possibly High	In-Reactor	Characterization after irradiation.
ANO1	4/PWR	50 (1986)	Possibly High	In-Reactor	Characterization after irradiation. (Annular pellets).
ANO2	(42)/PWR	55 (1986)	Possibly High	In-Reactor	Characterization after irradiation. (Annular pellets, graphite-coated cladding).

TABLE 5.1. Summary of Intact Rods Potentially Available for ATMs (con't)

Fuel Source, Identification	No. of Assemblies or (Rods)/Type	Nominal Burnup, MWd/kgM	High/Low Releasing (In-Reactor)	Location(1)	Characterization, Type/Reference
Oconee 1	5/PWR	50 (1989)	Possibly High	In-Reactor	Characterization after irradiation. (UO_2-Gd_2O_3 fuel).
BR-3	(28)/PWR	60-80 (1987)	Some High, Some Low	In-Reactor	Characterization after irradiation. (Includes some annular fuel and some UO_2-Nb_2O_3 fuel).

1. BCL = Battelle Columbus, West Jefferson Laboratory, Ohio.
   EMAD = Nevada Test Site, Nevada
   EG&G = EG&G, Idaho Falls, Idaho
   SRP = Savannah River Laboratories, South Carolina
   Reactor Owner - Reactor location or fuel is under jurisdiction of reactor owner.
   In-Reactor - Fuel is being irradiated in reactor listed.

2. Fuel is owned by USDOE or USNRC, but some sources require approval from previous owner(s) for release of data.

3. Rods are sectioned. Equivalent of 3+ rods from originally five rods.

4. Rods are sectioned. Equivalent of approximately three rods from originally four rods.

5. Irradiation recently completed, i.e., very fresh fuel. Fuel is (will be) better characterized than standard fuel because of being included in USDOE-sponsored Extended Burnup Program. Some rods may have been removed for PIE. Fuel belongs to reactor owner; will require negotiations with owner and USDOE for possible usage.

6. Irradiation in progress. Fuel will be better characterized than standard fuel because of being included in the USDOE-sponsored Extended Burnup Program. Fuel belongs to reactor owner; will require negotiations with owner and USDOE for possible usage.

6.0 ASSESSMENT OF APPLICABILITY OF THE CURRENT INVENTORY OF SPENT FUEL TO THE ATM CLASSES

6.1 APPLICABILITY OF AVAILABLE MODERATE-BURNUP, LOW-RELEASING UO_2 SPENT FUELS

All the available spent fuels listed in Section 1 of Table 5.1 were discharged several years ago. The spent fuels from the H. B. Robinson and Turkey Point reactors meet the criteria for the Class 1, base-case ATM spent fuel (box 1, Figure 4.1). The characterization of these fuels, including fragmentation of the fuel pellets, is described by R. B. Davis (1980), R. B. Davis and V. Pasupathi (1981), and R. E. Einziger and R. L. Fish (1982). These fuels can be considered as candidates for ATM spent fuels for the class consisting of moderate burnup, low-releasing (in-reactor), originally solid-pellet spent fuel that have adequate pellet fragmentation. There are several hundred intact rods available from the Turkey Point reactor and about 130 intact rods available from the H. B. Robinson reactor. A temperature excursion occurred during postirradiation handling of the H. B. Robinson fuel. However, it was not sufficient to produce temperatures in excess of 300°C in the cladding during horizontal transport of the BO-5 assembly, as judged by microhardness measurements that showed no evidence of annealing of the irradiation damage to the cladding (R. E. Einziger and R. L. Fish, 1982).

The spent fuel from the Dresden reactor may be suitable for the Class 1, or possibly Class 3, high-releasing, UO_2 fuel (boxes 1 or 3, Figure 4.2). However, fission gas release measurements will be required to properly classify this fuel.

The spent fuel from the Shippingport reactor, as described by Bradley et al (1981), is too low in burnup to qualify as an ATM spent fuel. The spent fuel from the Peach Bottom 2 reactor is also too low in burnup to qualify as an ATM spent fuel. However, in the absence of a high-releasing (in-reactor), moderate-burnup fuel this fuel may possibly be used as a fill-in because the fission gas release is about 3% as described by Einziger and Fish (1982).

The MCC is currently acquiring H. B. Robinson fuel as an ATM for the Class 1, moderate-burnup, low-releasing (in-reactor) spent fuel. Turkey Point fuel is a potential backup spent fuel ATM. Current acquisition plans are summarized in Appendix B. Examples of the characterization proposed for these fuels are discussed in Section 7.3.

Because of schedular and shipping cost considerations, the first lot of H. B. Robinson fuel to be used as an ATM required sectioning of the individual rods into three segments subsequent to fission gas collection and prior to shipment. It is recognized the sectioning will expose the interior of the fuel rod to air and moisture. However, efforts have been made to minimize the air available to the fuel and fission product compounds by sealing the rod segments in limited-volume air-filled storage tubes. In all likelihood this exposure will be similar to any other spent fuel rod that is sectioned in an air environment. Every attempt will be made to procure only intact spent fuel for subsequent ATMs.

6.2 APPLICABILITY OF OTHER AVAILABLE SPENT FUELS

The listing of high burnup rods in Sections II and III of Table 5.1 was obtained from Lang (1982). Sufficient information is not currently known about these fuels to classify them appropriately. However, it is anticipated that some of the fuels will meet criteria for ATM spent fuels as they become available and are documented and characterized. These fuels should be relatively well characterized, compared to "the run-of-the-mill" discharged commercial fuel, and should be considered for testing as priorities are set for other classes of ATM spent fuel (see Sections 4.1 and 4.2). Some UO_2-Gd_2O_3 fuel is included in the extended burnup programs.

7.0 MCC CHARACTERIZATION OF SPENT FUEL ATMs

The spent fuel ATM specimens to be provided for dissolution/interaction studies will require characterization in order to interpret the test results. The following subsections 1) define the types of characterizations that are required, 2) select the types of examinations that will provide the characterization data, 3) provide an example of a characterization plan that will provide the necessary data, and 4) describe the ATM spent fuel archive system. The flow of spent fuel ATMs and their associated characterization data are illustrated in Figure 7.1.

The characterization plan described is given as a detailed guideline for optimum documentation of properties of ATMs and similar test materials; it should not be viewed as a commitment by MCC to the specific details shown. Rather, it is an illustration of the type of information and methods to be applied in providing the appropriate level of characterization for MCC certification of each spent fuel ATM for repository-related test programs. A detailed characterization plan will be established for each ATM individually. It will be based on the requirements of the specific class of spent fuel, the quantity and quality of previously published data, and the expected significance of the possible measurements relative to the cost and effort required to obtain the data.

7.1 CHARACTERIZATION OF SPENT FUEL ATMs

Characterization of spent fuel ATMs will be divided into two general categories; 1) generic information, including characteristics that are important for any repository condition or test method; and 2) special characteristics that are specific to a particular repository, a unique type of testing, or aimed at clarifying specific test results (Table 7.1). The generic characterization data will be supplied from the MCC data bank with the spent fuel ATM (Figure 7.1). The MCC will collaborate with experimenters in individual repository test programs in obtaining the special characterization on a "need to know" basis. Characterization classification is summarized in detail below.

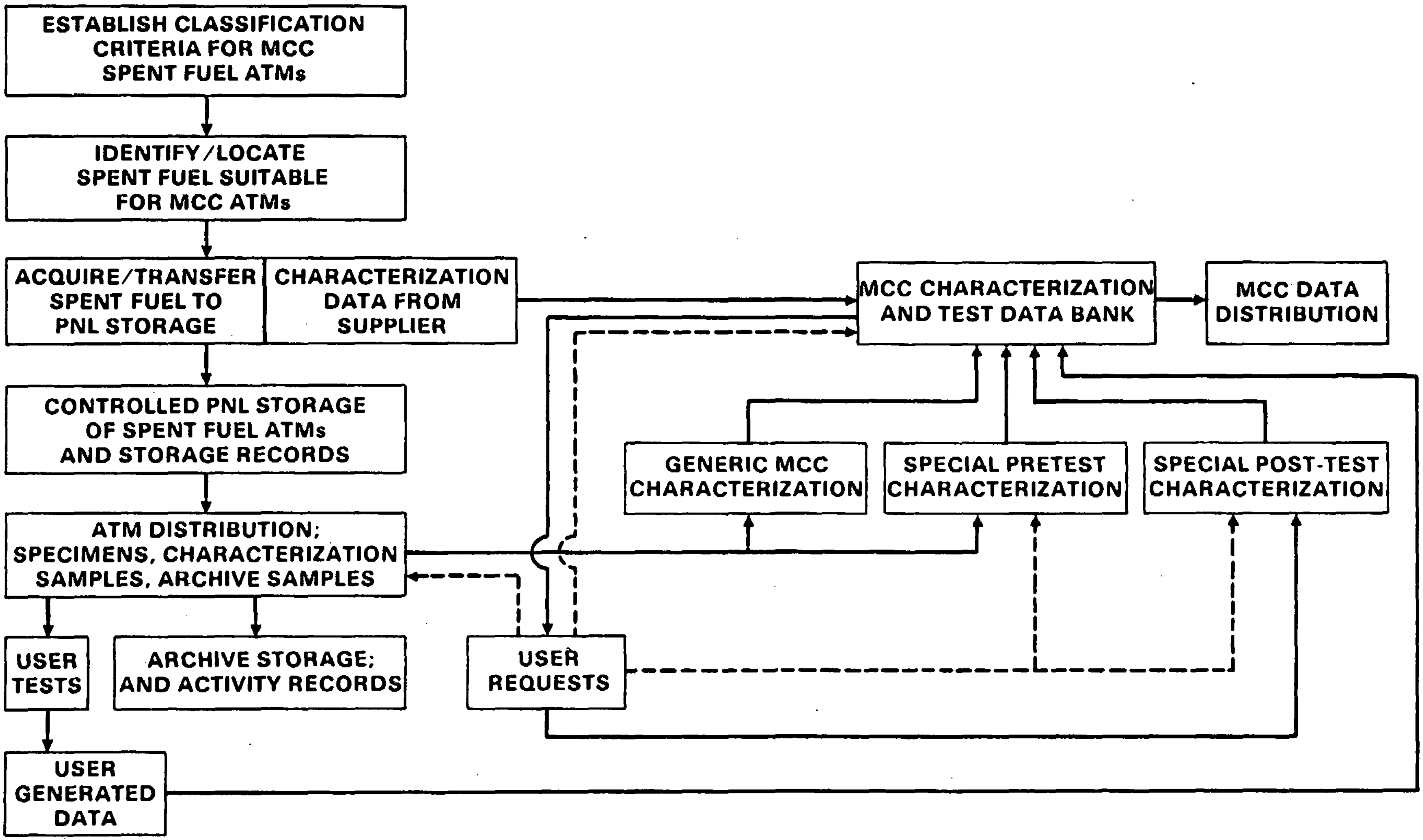

Figure 7.1. Flowchart for Acquisition and Characterization of MCC Spent Fuel ATMs and Characterization Data

Table 7.1. Spent Fuel ATM Characterization Data Classification

GENERIC INFORMATION	DATA SOURCE OR MEASUREMENT METHOD[a]
1. Related to ATM Classification	
a. Original fuel composition and form	a. Fabrication data
b. Approximate burnup level	b. Reactor power history
c. Fission gas release (in-reactor)	c. Measurement of gas content, gas composition, and rod void volume
d. Degree of pellet cracking	d. Transverse and longitudinal ceramography
e. Decay time	e. Computation from reactor shutdown date
f. Fuel rod integrity	f. Fission gas release measurement. Controlled storage of the penetrated cladding
g. Description of unusual incidents	g. Fabrication, operation, and handling/storage histories
2. Total Radionuclide Inventory	2. Burnup analyses, gross gamma scan, and ORIGEN calculation
3. Radionuclide Distribution	
a. Axial	a. Gamma scans and ORIGEN calculation
b. Radial	b. Calculation and/or microprobe/SEM examination
4. General Microstructure Characteristics	
a. Fuel material grain size and porosity distribution	a. Ceramography and quantimet
b. Cladding condition	b. Etched metallography
SPECIAL CHARACTERISTICS	**DATA SOURCE OR MEASUREMENT METHOD**
1. Related to Radionuclide Chemical Interaction with Repository Environment	
a. Radionuclide chemical species, e.g., phase analyses	a. Microprobe and SEM/EDX
b. Chemical species distribution, e.g., grain boundaries, radial location, second-phase inclusions	b. Ceramography, microprobe, and SEM/EDX
c. Fuel density, grain size or porosity distributions (quantitative)	c. Density measurement and image analysis
d. Specimen surface area	d. Photographs and direct measurement
2. Nonfuel Components	
a. Incipient cladding flaw description	a. Eddy current or ultrasonic examination and metallography
b. Crud characterization	b. Chemistry or SEM/EDX

(a) The terms ceramography and metallography are used herein to include photomicrographs from optical (light) systems and/or SEM.

GENERIC INFORMATION	
ATM classification data	Required to properly classify the spent fuel.
Quantifying radionuclide inventory	Required to determine overall release fraction for dissolution study.
Location of radionuclides within the specimen	Required to describe general distribution of radionuclides in the fuel material, in the gas space within the rod, on rod component surfaces, and those that were lost during specimen preparation.
Microstructure of fuel and cladding	Required to provide physical basis for interpretation of dissolution test results.
SPECIAL CHARACTERISTICS INFORMATION	
Data related to radionuclide chemical interaction with the repository environment	Required to develop detailed chemical models for spent-fuel/repository interactions.
Nonfuel material components	Required to permit accounting for effect of cladding, etc. on radionuclide release.

Spent fuel characterization related to special characterization information associated with chemical interactions between the spent fuel and the repository environment will be conducted on a case-by-case "need-to-know" basis after the specific problems have been identified. The examinations for test-related characterizations on nonfuel material components should be performed prior to dissolution testing and will be selected to provide information specifically for the intended spent fuel test.

7.2 SOURCES OF GENERIC CHARACTERIZATION DATA FOR MCC SPENT FUEL ATMs

The generic characterization data for MCC spent fuel ATMs are related to classification of the spent fuel, and to radionuclide inventory and distribution. The following describes the sources of the information/data

(Table 7.1) and the general philosophy for how it will be obtained for any particular group of spent fuel rods which will comprise a spent fuel ATM.

Several types of characterization data listed in Table 7.1 will be obtained from available records: i.e., 1a, fuel form and composition; 1b, approximate burnup level, 1e, decay time; and 1g, description of unusual incidents.

The fission gas release from the fuel must be known to properly classify each rod of a spent fuel ATM. The fission gas release measurement will consist of 1) the determination of the amounts of the common gases normally found or suspected to be formed in a fuel rod, i.e., Xe, Kr, He, Ar, N_2, O_2, CO, CO_2, and CH_4, 2) the volume of gas present in the fuel rod, and 3) the gas (open) volume of the fuel rod. The probable radioactive species in the plenum gas are ^{85}Kr and ^{14}C compounds. The ^{85}Kr will be determined from an isotopic analysis of the fission gas. Because all the fuel rods in an ATM group will be at a similar burnup level, the isotopic compositions of the fission gases in the rods will be similar and, therefore, the isotopic composition need be only spot-checked for each group, e.g., perhaps only for 1 or 2 rods.

Information regarding the availability of ^{14}C is important in repository evaluation, but analysis of the ^{14}C content in fission gas and solid fuel is difficult. A reasonable attempt will be made by the MCC to obtain ^{14}C information for each spent fuel ATM. Carbon monoxide, carbon dioxide and methane are the sources of ^{14}C in the plenum gas. Only a small fraction of the total carbon present in these gases can be expected to be ^{14}C. The amount of the individual gases in the plenum gas is often below the limit of detectability for the mass spectrometric measurements, and the sum of the amounts of the three gases, using the limit of detectability if gas is not detected, is usually less than 0.001 of the total gas, as described by R. E. Einziger and R. L. Fish (1982). A separation of the carbon-containing gases from the other gases will be required to concentrate the ^{14}C for determination by a technique such as beta-sensitive scintillation counting. Such a separation could be accomplished by oxidation of the carbon-containing gases to CO_2 and subsequent trapping in an alkaline trap.

The degree of pellet cracking must be known to assure that fuel fracture is typical of the intended spent fuel ATM class. Because the fuel rods in an ATM group will have had similar power/temperature histories, the degree of pellet fragmentation can be expected to be similar. Therefore, a ceramographic sampling along the length of one of the rods in the group, plus spot-checking at one location in the other rods, will be sufficient to characterize the degree of pellet cracking. The ceramographic specimens can also be used to estimate grain size, porosity, and specimen surface area. Both transverse and longitudinal ceramography will be required to assess the pellet cracking.

The integrity of each fuel rod prior to puncturing can be verified from the fission gas composition. Controlled handling/storage of ATM spent fuel at all times will be required during and after puncturing and sectioning of the fuel rods to prevent exposure of the material to water or other liquids and to minimize exposure of the material to air. Such exposures could modify the radionuclide inventory or change the dissolution characteristics of the spent fuel ATM.

Initial characterization of ATM radionuclide inventory and axial distribution will be conducted by a combined sampling/calculational/normalization procedure. The procedure comprises the following steps:

1) Perform gross and spectral gamma scans along the length of all rods within an ATM group, using the same counting procedure and geometry. This scan provides a relative activity/burnup indication along the length of each rod, and an inter-rod comparison of activity/burnup.

2) Select one rod from the ATM group and remove about five samples from locations having different gamma activities for burnup analysis.

Correlate the burnup results with the gross, or spectral, gamma scan results for the rod and predict the burnup distributions in the remaining rods. Spot-check the prediction by performing a burnup analysis on a sample from at least one additional rod in the ATM group.

3) Using the general power history, the burnup results, and the decay time, calculate the predicted radionuclide inventory of the spent fuel ATM specimens using a code such as ORIGIN or the later ORIGEN-II version.

4) Verify the results of the inventory calculation by performing radiochemical analyses for selected radionuclides, using the original burnup specimens from step 2. Compare the analytical results to the computed results and correct the calculation or normalize the analytical/calculation results as necessary. The radionuclides listed in Table 7.2 are recommended for the comparison. They represent radionuclides from the three groups of products, i.e., fission, activation, and transuranic, and also represent radionuclides that may be of special concern with respect to transport from a repository environment, e.g., ^{14}C and ^{99}Tc, or of special concern to the health of humans, e.g., the actinides and ^{14}C.

In principle the radial distribution of the radionuclides in the fuel can be determined using a similar procedure. However, the calculational techniques and computer software will probably require a lengthy and expensive development and, therefore, are not planned at this time. Instead, radial distribution of radionuclides will be limited to semi-quantitative examinations of the ceramographic specimens using SEM/EDX and/or microprobe methods. Quantitative determinations of radial distributions of radionuclides will be conducted on the "need-to-know" basis of the special characterizations, as described in Section 7.1.

TABLE 7.2. Recommended Radionuclides for Verification of Inventory Calculations

Type of Product	Isotope
Transuranic	^{235}U ^{239}Pu ^{237}Np
Activation	^{14}C
Fission	^{99}Tc ^{137}Cs ^{233}Pa

The general microstructure of the spent fuel ATM will also be determined by ceramography and SEM/EDX examinations. For grain size, fuel cracking, and similar fuel material characteristics, the ceramographic specimens will be used. The general cladding condition will also be determined by etching these specimens, and by SEM examination of the surfaces of representative fuel rod segments. Detailed evaluation of the cladding and characterization of crud deposits will be conducted only as a special characterization on a "need-to-know" basis, as described in Section 7.1.

7.3 EXAMPLE OF GENERIC CHARACTERIZATION PLAN FOR AN MCC-ATM SPENT FUEL

An example of a characterization plan for a group of eight fuel rods comprising a MCC spent fuel ATM is summarized in Table 7.3.

7.4 ARCHIVE SAMPLES FOR SPENT FUEL ATMs

Archive samples from the spent fuel ATMs will be retained in order to perform any special post-test characterizations on archive material (Figure 7.1) and for any other purposes as they may arise. Depending upon the quantity of the ATM samples supplied to the user and the variation in

TABLE 7.3. Example of a Generic Characterization Plan for a Group of Eight Spent Fuel Rods for Use as an MCC-ATM

	Number of Rods (No. of samples from each rod) Receiving Characterization									
			Ceramography							
No. Rods	Historical Records[(a)]	Fission Gas Release[(b)]	Transverse	Longitudinal	Burnup	Gross Gamma Scan	Inventory Calculation	Analytical Spot-Check Inventory	Radial Distribution Calculation[(i)]	Analytical Spot-Check Radial Distribution[(i)]
8	8	8[(c)]	1(5)[(d)] 7(1)[(e)]	1(5)[(d)] ---	1(5)[(f)] 1(1)[(g)]	8	8	1(5)[(h)]	8	1[(c)]

a) Includes characterization of fuel composition and form, approximate burnup level, decay time, and description of any unusual incidents from available records.

b) Includes gas composition, gas pressure, and rod void volume.

c) In addition, two rods to receive an isotopic analyses of the Kr and Xe and one rod to receive an evaluation of the ^{14}C in the plenum gas.

d) Adjacent transverse and longitudinal samples to be taken from five locations having different burnup levels.

e) Spot-check near mid-length of the fuel column.

f) Burnup samples taken adjacent to transverse ceramographic samples.

g) Burnup spot-check on a second rod.

h) Analyses of burnup specimens for radioisotopes listed in Table 7.2.

i) Methods to be determined; microprobe, SEM/EDX or other techniques that use the ceramography specimens where possible.

location of the pieces of ATM supplied, between 10 and 20% of the spent fuel ATM material will be retained as archive material. This material will be subject to controlled storage under an appropriate quality assurance/control system in order to preserve the material.

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APPENDIX A
ESTIMATES OF FAILED FUEL AVAILABLE FOR DISPOSAL

Table A.1. Estimates of Failed Fuel of Different Vintage for Disposal in 2020

Reactor Type	Fuel Rod Failure Rate[a], %		Fuel Inventory in 2020[b], MTHM		Inventory of Failed Fuel Based Upon In-Reactor Failures[c], % of Total		Inventory of Failed Fuel Based Upon In-Reactor and Handling/Storage Induced Failures[d], % of Total	
	Prior to 1975	After 1975	Prior to 1975	After 1975	Prior to 1975	After 1975	Prior to 1975	After 1975
BWR	0.76	0.02	772.4	45,291.8	0.004	0.006	0.007	0.159
PWR	0.005 to 0.45 (Ave. ~0.15)	0.02	815.5	100,937.6	0.001	0.014	0.004	0.355
				Totals	0.005	0.020	0.011	0.514

(a) Summarized from NUREG/CR-3602 (PNL-4817) for U.S. fuel vendors.

(b) Summarized from DOE/NE-0017/2 assuming no future reprocessing.

(c) Assumes all spent fuel available in 2020 is stored in a repository.

(d) Assumes a 0.5% fuel rod failure rate for predisposal handling and storage damage. This assumption is made only as an example; actual rate may actually be higher or lower.

APPENDIX B

SUMMARY OF MCC ACTIVITY FOR ACQUISITION OF SPENT FUEL ATMs

Table B.1. Summary of MCC Activity for Acquisition of Spent Fuel ATMs[(a)]

ATM Source	ATM Class	No. Rods for ATM	Rationale for Selection
H. B. Robinson, Assembly B-05	1 [moderate-burnup, low-releasing (in-reactor)]	8 to 10	Results can be related to other programs. Rods are immediately available. Rods are intact. Rods meet all selection criteria for ATM (see Sections 4.0 and 6.0). Sufficient number of rods having same original enrichment and at similar burnup for current usage projections.
Peach Bottom-2, Assembly PH-006	3 [moderate-burnup, high-releasing (in-reactor)]	0 to 2	Results can be related to other programs. Rods are available if fuel vendor approves of MCC use. Rods are intact. Rods are closest, of available fuels, to meeting criteria for Class 3 ATM.
Turkey Point, Assembly B-17	1 [Backup for HBR for Class 1; moderate-burnup, low-releasing (in-reactor)]	10 to 20	Results can be related to other programs. Rods may be available, pending cost for removal from assembly and approval of fuel vendor. Rods are intact. Rods meet all selection criteria for ATM (see Sections 4.0 and 6.0). Sufficient number of rods are available.
Turkey Point, Assemblies at EMAD	1 [Backup for HBR for Class 1; moderate-burnup, low-releasing (in-reactor)]	--	Results can be related to other programs. Rods meet all selection criteria for ATM (see Sections 4.0 and 6.0). An entire fuel assembly would have to be acquired which would be costly and provide excess material which will later cause disposal problems. Use of this material would be considered only if both HBR, Ass'y B-05, and Turkey Point, Ass'y B-17, rods were unavailable.

Table B.1. Summary of MCC Activity for Acquisition of Spent Fuel ATMs[a] (con't)

ATM Source	ATM Class	No. Rods for ATM	Rationale for Selection
Zion	1, 2, or possibly 4	As required.	Results can be related to other programs. Rods will meet all the selection criteria for an ATM. Rods will be available at several burnup levels from fuel manufactured in a single campaign. Costs, schedules, availability to be determined. (Fuel is privately owned and will require utility/vendor approval for usage).

(a) The acquisition of nine H. B. Robinson fuel rods as a spent fuel ATM has been completed and characterization is underway. Acquisition of additional spent fuel is under consideration. Characterization of each ATM will be based upon procedures described in Section 7.0.

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