

RECONSTITUTION OF SPENT FUEL TO IMPROVE REPOSITORY

WASTE FORM CHARACTERISTICS

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ABSTRACT

Spent nuclear fuel is a fractured, partially recrystallized material with a radioisotope distribution that depends on the operating history of the fuel. Such material may be difficult to qualify as a repository waste disposal form. Because the fractures increase the surface area of the spent fuel, the leachability of the contained fission products and transuranics could be increased by as much as two to three orders of magnitude once the cladding is breached. The increased leachability along with the variability in fission product content and size distribution of spent fuel will make it difficult to quantify the leach rate of radionuclides and verify that spent fuel will meet the $<10^{-5}$ /yr leach criterion. Reconstituting spent fuel into a crystalline ceramic waste form will not only enhance leach resistance significantly but will also decrease the variability in the waste form so the waste form characteristics can be more easily quantified. This can be accomplished with no material addition to the spent fuel since uranium dioxide, the main component of spent fuel, is a geologically stable mineral. A process is proposed to reconstitute spent fuel using a decladding-pulverization step followed by a reconstitution hot isostatic pressing step. Preliminary economic evaluation indicates the cost of reconstituting spent fuel and disposal is generally greater than the cost of disposal of unconsolidated spent fuel assemblies. Reconstitution and disposal are generally less costly than disposal of consolidated spent fuel providing the waste form is emplaced in the repository before the waste has aged for 30 years. If 10-year-old fuel is emplaced in a repository, reconstitution and disposal are essentially the same cost as disposal of unconsolidated spent fuel.

INTRODUCTION

The Nuclear Waste Policy Act of 1982 establishes the methodology to be used for permanent disposal of commercial nuclear wastes. Disposal of spent fuel elements is one option that is being evaluated. The wide variation in the nature of spent fuel will make this material difficult to characterize as a nuclear waste form.¹⁻⁶ Just developing the tests to verify that spent fuel meets the $<10^{-5}$ /yr leachability criteria for all contained material is a formidable problem. Therefore, in this paper, a cost-effective alternate approach is discussed which will reconstitute the spent fuel into a much more uniform and more leach-resistant material.⁷⁻¹² The nature of spent fuel is discussed followed by the advantages of reconstituting spent fuel. The proposed process to reconstitute spent fuel is outlined and the paper concludes with preliminary economics of this spent fuel management alternative.

NATURE OF SPENT FUEL

Oxide fuel undergoes significant restructuring during irradiation. Near the center of the pellet the high thermal gradient, induces the formation of columnar grains. Equiaxed grain growth occurs adjacent to the outer radius of the columnar grain zone. Grain growth does not take place in the outer, cooler regions of the pellet. The net result of this fuel pellet restructuring is the formation of a highly fractured heterogeneous grain structure.¹⁻³

Accumulation of fission products as burnup increases affects the dimensional and chemical

stability of the fuel, leading to additional fracturing. The burnup, fission product content, and plutonium content of irradiated UO_2 fuel from a thermal reactor will be greatest near the outer regions of the pellet and can vary significantly across the pellet. The exact burnup distribution for a thermal reactor fuel pellet depends upon the type of reactor, type of fuel, position in the core, and amount of burnup.⁴⁻⁶ The net result is that spent fuel is a widely varying, nonhomogeneous, highly fractured material that will be extremely difficult to characterize and qualify as a waste form.

Preliminary evaluations of the leach characteristics of spent fuel show widely varying leach rates, as might be expected.¹ In general, the leach rates of fission products are considerably greater from spent fuel than from borosilicate glass which, in turn, are generally greater than from crystalline waste forms. Thus, with spent fuel disposal, isolation from the biosphere will depend more on the repository than on the waste form. On the other hand, the waste form should be the easiest and surest portion of the isolation matrix to characterize and model the long-term stability and isolation potential.

Reconstitution of spent fuel into a crystalline waste form will remove the volatile fission products and a significant portion of the cesium, one of the two major heat-generating fission products. This not only simplifies waste management by reducing the heat generated by the waste form but also recovers the cesium as a radiation source. The reconstituted

waste form will be theoretically dense crystalline uranium dioxide monoliths. This crystalline waste form will be much easier to characterize than spent fuel and will have a low uniform leach rate corresponding to the geologically stable uranium dioxide mineral. It is expected that the remaining fission products and actinides will be incorporated in the crystalline uranium dioxide structure. Thus the leach rate of the fission products and actinides will be a function of the uranium dioxide leach rate. Since the waste form can be more adequately characterized, verifying that the waste form meets disposal criteria, and developing a model to predict the long-term isolation of the radionuclides will be more precise and can be accomplished with much greater assurance.

RECONSTITUTION PROCESS DESCRIPTION

Two processes developed for nuclear fuel and waste processing are combined to reconstitute spent nuclear fuel. These two processes are the AIROX (Atomic International Reduction Oxidation) fuel reprocessing method⁷⁻⁹ and the hot isostatic pressing (HIP) waste consolidation method.¹⁰

The AIROX process was developed in the late 1950s and early 1960s to readily reprocess and reconstitute spent fuel for recycle back to nuclear reactors.⁷⁻⁹ The AIROX process is a low decontamination pyrochemical processing method that is proliferation-resistant.⁹ It was investigated for the national and international nuclear fuel cycle evaluation in the late 1970s and is also planned as a head-end step for conventional Purex reprocessing of spent fast breeder fuel.

AIROX processing is a pyrochemical reprocessing technique that is used to simultaneously deacid the fuel, pulverize the fuel, remove the volatile fission products, and restore the fuel to the proper chemical form for reenrichment and recycle. The reprocessing technique utilizes a simple gas-solid oxidation-reduction chemical reaction to form a compound which has a volume much larger (~30%) than that of the original material. The AIROX process oxidizes UO_2 to U_3O_8 and reduces U_3O_8 to UO_2 . When the fuel expands, due to the formation of the less-dense new chemical compound, the cladding ruptures and the spent fuel falls free of the cladding. Repeated oxidation-reduction pulverizes the spent fuel and prepares it for HIP reconstitution into the crystalline uranium dioxide waste form.

Approximately 99% of the tritium, up to 75% of the iodine, and up to 98% of the krypton and xenon can be expected to be released from a single oxidation cycle; with multiple cycles, most of these gaseous components are released. Significant fractions of other fission products such as cesium, tellurium, ruthenium, technetium, cadmium, and indium are also evolved during AIROX fuel processing. The capability to process highly irradiated fuel and reconstitute the product into fuel for recycle to a reactor has been experimentally demonstrated.

The AIROX processed material is a finely divided material that is sufficiently physically active to convert into theoretically dense uranium dioxide monoliths with the application of heat and pressure. The AIROX processed material is placed into a HIPping can which will compress to the desired sized monoliths for incorporation into geologic repository emplacement canisters or overpak units.

Hot isostatic pressing (HIPping) technology has been used in the ceramic, metallurgical, and cermet industries for over 30 years. Radioactive materials and fuel pellets have been consolidated in DOE and commercial HIP units for ceramic immobilization of high-level wastes¹⁰ or fuel fabrication. Most HIP units are or could be remotely operated; many units are automated so that the entire HIPping operation from loading to unloading is automatic and controlled from a remote console. Integration of the two processing steps into a single remotely operated and maintained processing line is required to adapt this technology to reconstitution and immobilization of spent fuel.

ECONOMICS

To obtain a preliminary cost estimate of reconstituting spent fuel, a process layout was developed. The processing equipment was sized to process 60,000 tons of spent fuel in 20 years, i.e., 3,000 tons per year. The processing system is divided into two sections or cells to control contamination. In one cell the spent fuel is deacid, pulverized, and canned. Thus, in this cell there is the potential for contamination outside the processing vessels. However, only canned material is processed in the second cell so the likelihood of contamination outside the cans is much less. The entire system would be designed for remote maintenance, but hands-on maintenance could probably be used for the second cell if desired.

The different options for management of spent fuel are shown in Fig. 1. These options consist of (1) emplacing the spent fuel in a repository, (2) consolidating the spent fuel prior to repository emplacement and (3) reconstituting the spent fuel prior to repository emplacement. For convenience, it is assumed that the fuel has been aged 10 years prior to repository emplacement. The spent fuel can be emplaced without further cooling. Consolidated fuel requires further storage before emplacement or consolidation into smaller diameter canisters; however, reconstituted fuel can be emplaced directly because the high heat-producing cesium has been removed. In this model it has also been assumed that using canisters greater than 60 cm (24 in.) in diameter for repository emplacement will automatically lead to a cost penalty due to the different technique required to mine the larger emplacement holes. This is reflected by a step function in the waste management costs as shown in Fig. 2.

The model used to evaluate the economic impact of AIROX reconstitution of LWR fuel¹¹⁻¹² is essentially the same as reported previously¹¹ except for the following:

1. The cost of fuel pin disassembly and fuel processing and reconstitution has been added.
2. The cost of recovery and storage of radio-krypton has been added.
3. A limitation on the heat rejection capability of the shipping cask of 50 kW has been added.
4. A decay heat curve for LWR fuel with a user-specified fraction of cesium removed has been added.
5. The cost of disposal of secondary waste from the reconstitution of spent fuel has been added.

SPENT FUEL MANAGEMENT ALTERNATIVES

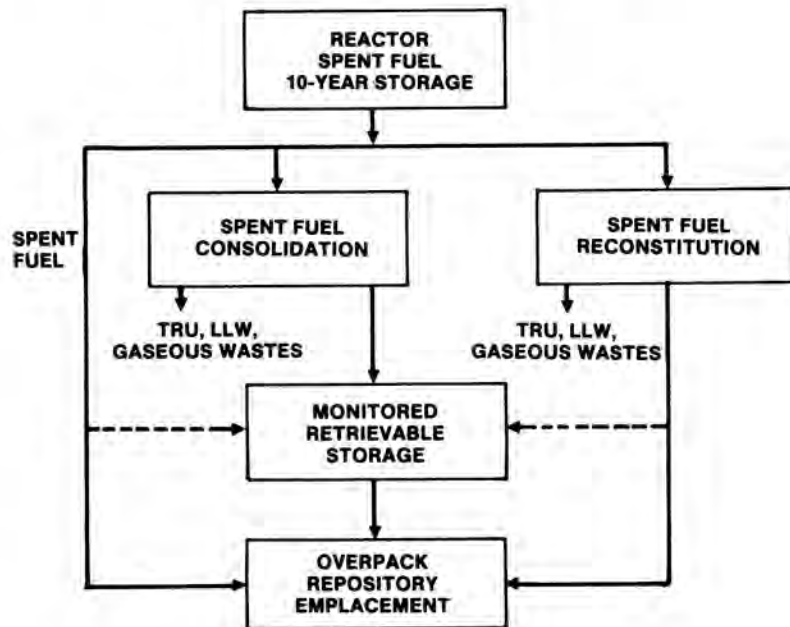


Fig. 1. Spent Fuel Management Alternatives

COST FOR PROCESSING AND/OR DISPOSAL OF SPENT FUEL

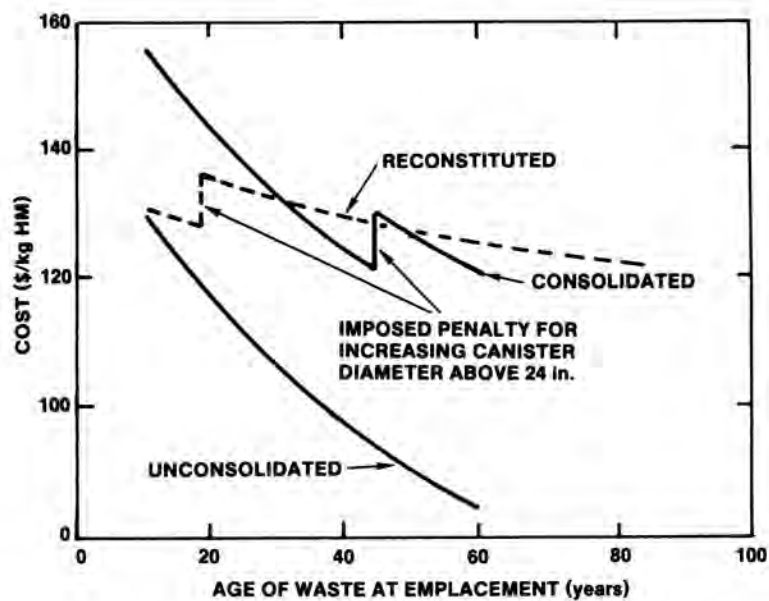


Fig. 2. Cost for Processing and/or Disposal of Spent Fuel

6. The model assumes reconstituted spent fuel is an acceptable waste form; therefore, no buffer is added to the waste package.

Fig. 2 shows the cost comparison among unconsolidated spent fuel, consolidated spent fuel, and reconstituted spent fuel for the waste management system in which 10-year-old fuel is the waste being processed and the immediate backfill alternative is used in a basalt repository. The unconsolidated spent fuel alternative assumes no krypton is stored and no secondary TRU waste is generated. The consolidated spent fuel alternative assumes consolidation costs are \$2.3/kg U; krypton must be stored at \$23.8/kg U; and the disposal cost for secondary TRU waste is \$2.2/kg U. In order to meet the leachability criterion, spent fuel canisters are assumed to contain a clay buffer. The temperature limit for the clay is assumed to be 300°C. The reconstituted fuel alternative assumes reconstitution costs are the same function of waste form diameter as used for ceramic waste forms from high-level waste^(11,12) plus \$23/kg U for disassembly of spent fuel; krypton storage costs are \$31.4/kg U, and the cost of disposing of secondary TRU waste is \$9.4/kg U.

From Fig. 2, and as previously reported,⁽¹¹⁾ the disposal of consolidated spent fuel is more expensive than unconsolidated spent fuel disposal. The driving force for fuel consolidation is not waste disposal cost but the ability to increase the spent fuel storage capacity in fuel storage pools. For fuel emplaced without use of a monitored retrievable storage (MRS) to age the fuel, fuel reconstitution is economically competitive with emplacement of unconsolidated spent fuel. This is caused by the significant economic penalty associated with restricting the amount of unconsolidated fuel in each waste package due to the buffer temperature limit. The repository space used to emplace reconstituted fuel is significantly less than that used for consolidated fuel. Therefore, reconstitution offers comparable economics to the disposal of unconsolidated fuel without using the large amount of repository space associated with the unconsolidated spent fuel alternative.

For the reconstitution alternative, cost is a weak function of fuel aging. Therefore, if an MRS is used to age unconsolidated or reconstituted fuel prior to emplacement in a repository, the consolidation alternative becomes the economically desirable alternative for fuel aged more than 30 years.

CONCLUSIONS

A process has been proposed to reconstitute spent fuel to improve its repository characteristics. This converts the spent fuel into a superior waste form as an alternative to disposal of spent fuel or consolidated spent fuel. The proposed process consists of two steps, AIROX decladding and HIP reconstitution into monoliths. A preliminary assessment of the economics of reconstitution has also been made and indicates that reconstitution is cost competitive with fuel consolidation. Reconsti-

tution will improve fission product and actinide leach resistance and provide a waste form that will be much easier to characterize than spent fuel. Modeling the reconstituted waste form for projection of long-term environmental isolation will also be much easier and more precise than modeling either the unconsolidated or consolidated spent fuel waste forms. Therefore, it appears that fuel reconstitution is a feasible, cost-effective alternative method to improve repository characteristics when compared with disposal of either spent fuel or consolidated spent fuel.

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