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G. A. Armantrout
L. J. Jardine

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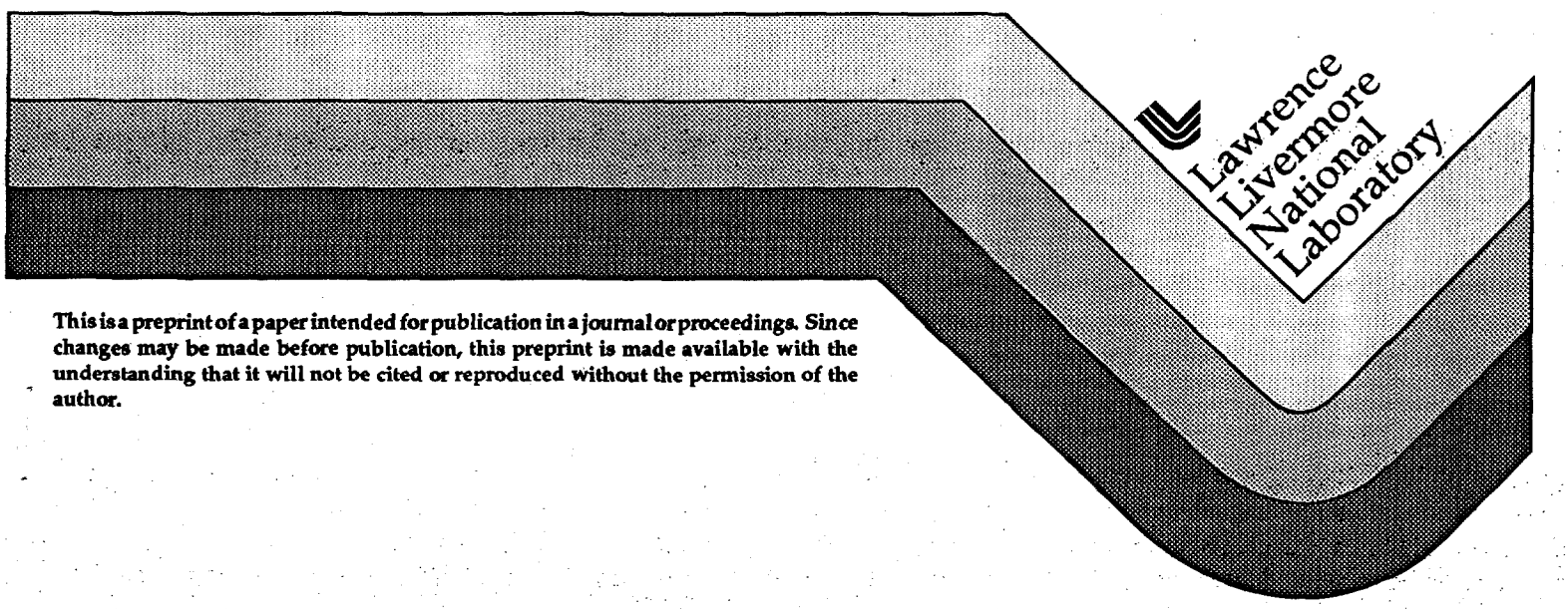

Disposition of Excess Plutonium Using "Off-Spec" MOX Pellets as a Sintered Ceramic Waste Form

Guy A. Armantrout and Leslie J. Jardine
Lawrence Livermore National Laboratory

ABSTRACT

We describe a potential strategy for the disposition of excess weapons plutonium in a way that minimizes (1) technological risks, (2) implementation costs and completion schedules, and (3) requirements for constructing and operating new or duplicative Pu disposition facilities. This is accomplished by an optimized combination of (1) using existing nuclear power reactors to "burn" relatively pure excess Pu inventories as mixed oxide (MOX) fuel and (2) using the same MOX fuel fabrication facilities to fabricate contaminated or impure excess Pu inventories into an "off-spec" MOX solid ceramic waste form for geologic disposition. The key to the combination approach is the use of a sintered ceramic waste form (SCWF) consisting of a UO_2 encapsulating matrix for the excess PuO_2 inventories. The SCWF will have a high probability of being acceptable for geologic disposition because it is similar to an unirradiated MOX fuel matrix without fission products. Because it can be fabricated using the same basic technologies, processing equipment, and facilities used to fabricate the MOX fuel for reactors, there is no need to construct and operate specialized and duplicative facilities using other immobilization technologies such as vitrification.

Diversion protection for the SCWF to meet the "spent fuel standard" introduced by the National Academy of Sciences[1,2] can be achieved in at least three ways. (1) One can utilize the radiation field from defense high-level nuclear waste by first packaging the SCWF pellets in 2- to 4-L cans that are subsequently encapsulated in radioactive glass in the Defense Waste Processing Facility (DWPF) glass canisters (a "can-in-canister" approach). (2) One can add ^{137}Cs (recovered from defense wastes at Hanford and currently stored as CsCl in capsules) to an encapsulating matrix such as cement for the SCWF pellets in a small hot-cell facility and thus fabricate large monolithic forms. (3) The SCWF can be fabricated into reactor fuel–like pellets and placed in tubes similar to fuel assemblies, which can then be mixed in sealed repository containers with irradiated spent nuclear fuel for geologic disposition.

INTRODUCTION

With the end of the cold war, major planning efforts were begun for the management of the excess fissile materials that would result from the reduction of U.S. and Russian nuclear stockpiles. This was motivated by the fear that such a large inventory of weapon-quality fissile material posed a worldwide risk of nuclear terrorism or nuclear proliferation. The United States and Russia are seeking cost-effective methods for the management and ultimate disposition of these materials, with special emphasis on

plutonium, that achieve each country's nonproliferation goals and that can be implemented in a timely manner. The current Russian position strongly favors burning the excess Pu in nuclear reactors. The United States is evaluating options that include, but are not limited to, burning excess Pu in existing reactors or immobilization with radionuclides in glass or ceramic matrices; in either case the final product is buried in a geologic repository. Deep-borehole (2–4 km) burial of Pu in an immobilized form is also being evaluated. All these options have advantages and drawbacks. This paper outlines a potential strategy for disposition of excess Pu in two streams: burning suitably pure Pu (as MOX) in existing reactors, and disposing of contaminated or impure Pu as a sintered ceramic waste form (SCWF) containing 2–10 wt% PuO_2 in a UO_2 matrix, similar in form and concentration to either unirradiated or spent MOX fuel. This alternative approach may minimize capital outlays, repository acceptability issues, and implementation duration schedules.

DISPOSITION CRITERIA

Disposition of excess Pu must satisfy three key objectives to achieve overall safety and nonproliferation goals:

1. Rendering the Pu relatively inaccessible for nuclear weapons use

This objective was one of the principal outcomes of the National Academy of Sciences study,[1] which recommended placing the excess Pu in a form that is roughly as inaccessible for weapons use as are the much larger worldwide stockpiles of Pu contained in spent commercial nuclear fuel (SNF). To achieve this degree of inaccessibility requires measures that would make it difficult for a terrorist group, a non-weapons state, or even the United States or Russia themselves to use (or reuse) the Pu in any kind of nuclear explosive device. The National Academy study observed that greater protection than this for excess Pu would be pointless and expensive, and that less protection would be dangerous. This objective is generally interpreted as meeting the "spent fuel standard."

Access to SNF for weapons use is made difficult by the existence of a high radiation-field barrier, by its having Pu isotopic mixtures different than that of weapons-grade Pu, by dilution of the Pu in a ceramic or glass matrix that requires significant chemical and physical processing for recovery, and by containment of the Pu in large, easily accounted and controlled unit item sealed forms. These factors are being considered by the DOE in developing a spent fuel standard to be used in judging the acceptability of any proposed final disposition form before the implementation of any disposition technology approach.[3,4]

2. Conducting disposition operations so that their health and environmental impacts are acceptable

Any processing activity or final disposition material forms that could adversely affect the environment after geologic disposal must meet myriad environmental, health, and safety regulations. Pu processing operations, geologic repository operations, and final

disposition forms that could affect worker or public radiation doses or exposures to hazardous chemicals must meet or exceed the regulative requirements.

3. Achieving timely, cost-effective disposition

If a given strategy is to achieve timely, cost-effective disposition of excess Pu, all necessary operations must be technologically viable so that R&D schedule times can be minimized. That viability depends on the existence (or ready construction) of appropriate facilities, equipment, and technologies. Pu disposition options requiring high front-end costs (e.g., for construction of new facilities or processes and for the supporting R&D) will not be regarded as desirable by the current balanced budget–minded Congress. Life-cycle costs (which include operational expenses and decommissioning), which are usually discounted, have less immediate, but still non-negligible, impacts and must also be minimized.

OVERVIEW OF DISPOSITION STRATEGIES

Many disposition strategies were considered in the NAS report,[1] including space launch and sea dilution. Three of these general Pu disposition strategies are under evaluation by the DOE Office of Fissile Materials Disposition[3,4]: (1) nuclear burning in a reactor, (2) immobilization with radionuclides in solid matrices to achieve the spent fuel standard, and (3) burial in deep (2–4 km) boreholes in ancient rock. The Russians favor nuclear burning as a way of using the energy content of the fissile Pu. Many in the United States, on the other hand, consider the Pu as "waste" to be disposed of by the most expedient and cost-effective method. The draft Preliminary Environmental Impact Statement[3] assumed the use of a single strategy for all excess U.S. Pu inventories—that is, for both relatively pure Pu inventories recovered from weapon components and other, less pure Pu in storage or contained in processing residues or scraps at various U.S. DOE sites. The relative advantages and disadvantages of the three strategies are discussed below.

1. Nuclear Reactor Burning. This approach achieves disposition by burning MOX fuel containing the Pu in a nuclear reactor and then discarding the spent MOX fuel, without reprocessing, in a geologic repository. The major advantages of this approach are its technical maturity (particularly in Europe), its partial favoring by the Russians (who however favor recovery and recycle of the Pu in the spent fuel), and the achievement of the spent fuel standard by the irradiated product form. Both the once-through and recycle approaches reduce the overall weapons Pu inventory and change the plutonium isotopic mixture. The primary disadvantages include possible opposition by opponents of nuclear power, the need to purify impure Pu before MOX fabrication, and reliance on a final geologic repository that is not yet operational. The aqueous technologies used in purification of Pu are expensive and generate significant quantities of secondary low-level and transuranic (TRU) wastes.

2. Immobilization with Radionuclides. This approach achieves the spent fuel standard by incorporating the Pu in a stable, solid matrix containing radionuclides (high-level radioactive defense wastes or ^{137}Cs) whose radiation field, in conjunction with the

chemical dilution, large sizes, and safeguards and security, acts as a deterrent to theft or reuse.[5] The immobilized form is eventually discarded in a geological repository in a manner similar to SNF. Immobilization may be a faster, simpler, and cheaper way to achieve the spent fuel standard than reactor burning. The technology is less mature than reactor burning, however, and shares the same concerns over the lack of a geologic repository.

3. Deep Borehole Burial. This approach depends on the depth of the geological isolation, rather than on a radiation "spike" and on physical and chemical characteristics, as in the first two approaches, to achieve the nonproliferation objectives. Its chief advantages are simplicity and perceived permanence, neither of which is offered by the reactor or immobilization approaches until a geologic repository is operated, filled, and sealed. Significant licensing and siting issues must be resolved before implementation, however.[3,4]

DEVELOPMENT OF SINTERED CERAMIC WASTE FORM STRATEGY

None of the three technology approaches just described can alone provide an optimum disposition strategy. We therefore propose a hybrid strategy, not yet evaluated by DOE, that combines the advantages of two of the technologies to achieve cost-effective, timely disposition.

Because both the United States and Russia possess reactors capable of burning excess Pu, we take reactor burning as a main constituent of our hybrid strategy. And because U.S. Pu policy discourages the development of breeder reactor technology, we assume the use of existing thermal reactors in the United States (or possibly Canada). It remains to be ascertained that there is sufficient reactor capacity which can be made available and that this approach is politically and publicly acceptable.

Reactor burning alone has the disadvantages already described which includes the need to purify much of the feedstock excess Pu to remove impurities incompatible with reactor fuel specifications. The monetary value of Pu as a reactor fuel in the U. S. is negative, given the current cost of low enriched uranium fuel, so the United States has little incentive to recover impure Pu. We suggest instead that a better approach would be to discard any excess Pu that would require significant purification by fabricating an acceptable geologic repository solid form. This would reduce the Pu inventories requiring reactor burning and would shorten the completion schedule (by reducing time-consuming reactor operations); it would probably also reduce the number of reactors needed. For this approach to be attractive, however, there must be a low-cost existing technology for discard of the impure Pu not going to the reactor burning option that meets existing environmental laws, anticipated repository requirements, and nonproliferation objectives.

DOE is considering two discard approaches[3,4]: deep borehole disposition and immobilization with radionuclides followed by geologic disposition. Deep-borehole disposition would require significant effort to modify existing laws, complete the licensing processes, and characterize a potential site. This preparatory effort would be the

same for any borehole disposition strategy, essentially independent of the quantity of Pu to be discarded. New facilities would be required for the borehole operations, increasing up-front costs and possibly introducing implementation time delays. It is unlikely that this approach would be cost- or time-effective for the smaller quantities of impure Pu not destined for burning as MOX in a reactor as part of the proposed hybrid disposition strategy. Thus, deep-borehole disposition does not appear particularly advantageous as part of this strategy.

The immobilization approaches under consideration involve use of a variety of matrix forms for Pu entrainment, including glasses, ceramics, or possibly a glass-bonded zeolite.[3,4] Any waste form intended for repository emplacement must qualify for long-term geological disposal; qualification is a lengthy process and would add to the disposition cost and time for any new waste forms.

Glasses are being developed for disposition of high-level waste from the Defense Waste Processing Facility (DWPF). Since glasses proposed for DWPF high-level waste immobilization were not developed for Pu containment, there are technical problems involving Pu solubility and qualification problems regarding repository acceptability of existing glass waste forms containing high concentrations of fissile materials for Pu disposition. Ceramic or other waste forms under consideration have similar and probably greater concerns.

For a multitude of technical reasons, existing glass immobilization facilities (such as DWPF) cannot be used directly for Pu disposition in glass. In the case of DWPF, for example, criticality considerations would require new melter designs and extensive feed preparation facility modifications and new supporting technology development. Ignoring any issues of delaying the current DWPF mission schedule for HLW vitrification, this would require major front-end investments in research and (eventually) in facility construction and modifications that could exceed the cost and time penalties of simply purifying the impure Pu with current Pu processing technologies for reactor burning.

The acceptability of the Pu disposition waste form within the repository must also be considered. Two waste forms have received significant attention in the United States: DWPF glass and spent commercial nuclear light water reactor (LWR) fuel. DWPF glass, while acceptable for containment of defense high-level waste at the Savannah River Site, is not likely to be directly suitable for Pu containment. This would require the development of alternative glass compositions to serve as the Pu containment systems. Spent LWR fuel is of particular significance in this context, because U.S. policy contemplates direct disposal of such fuel in a geological repository, without reprocessing. In this case, the primary barriers to Pu release to the geologic environment are the containers, any other engineered barriers used to emplace the spent fuel in the repository, the spent fuel assembly structural materials, and the ceramic UO_2 matrix form encapsulating the PuO_2 and fission products in the spent fuel.

To achieve waste form acceptability for the repository, to avoid new facility construction or new process development, and to control costs and schedule, we suggest a hybrid

strategy that produces only one type of material form. The proposed waste form is a UO_2/PuO_2 matrix containing 2–10 wt% PuO_2. Such a matrix is prepared and sintered similar to MOX reactor fuel fabrication and has a chemistry and morphology similar to MOX reactor fuel. Figure 1 shows the proposed strategy, here specifying a light water reactor (LWR) to burn the relatively pure Pu in MOX fuel and using the MOX fabrication technology to fabricate and discard the impure Pu as a sintered ceramic waste form (SCWF). The primary advantages of such a waste form include likely repository acceptability with minimal additional qualification testing for the SCWF and compatibility of SCWF processing and facility requirements with those routinely used in Europe for MOX fuel fabrication. If a permanent discard solution other than a geologic repository for the spent LWR nuclear fuel must be found, that solution would also apply to the discard SCWF waste form proposed here. Disposition of 10 to 20 t of impure Pu in any of the SCWF forms suggested would result in a MOX fuel–like component of repository waste that is less than 1% of the projected U.S. SNF inventory (about 80,000 t).

The first step in fabricating such a compatible waste form for the impure Pu would be to make a compressed PuO_2/UO_2 mixture and pellet similar to the starting form for MOX fuel, as indicated in Fig. 1, but without the dimensional tolerances and feed Pu purity requirements applicable to MOX destined for a LWR. Impurities introduced with the impure Pu feed would in most cases probably be contained in the waste form and would likely not decrease repository acceptability any more than the fission products normally contained in spent LWR fuel. Such a compressed pellet form would then be sintered in a reducing atmosphere to increase grain size and intergranular bonding to form a sintered matrix, which would constitute the final Pu containment waste form. This sintered ceramic waste form (SCWF) pellet would then be encapsulated in one of two types of container, as suggested in Fig. 1: (1) relatively small (2 to 4 L) cans, which would then be sealed and installed in the glass can-in-canister configuration for addition of a radiation barrier such as DWPF glass containing defense HLW, or (2) relatively large DWPF type canisters (0.6 m o.d. × 3 m long), to which would be added an encapsulating grout matrix containing ^{137}Cs. A third, hypothetical approach could be to fabricate canisters of SCWF pellets in geometries like fuel assemblies and mix these canisters with irradiated SNF in the multipurpose canister (MPC)-like repository container[6] for final emplacement. Radiation fields calculated for mixing five fuel-like SCWF assemblies with 12 SNF assemblies from pressurized water reactors (PWRs) are on the order of 300 rem/hr at 1 m from the centerline 30 yr after MPC container loading; this is similar to the radiation fields for the PWR spent fuel assemblies alone.[5] Such fuel-like SCWF assemblies would require a level of protection similar to MOX assemblies until they were mixed with the spent fuel, at which time the essence of the spent fuel standard would be achieved.

Clearly, such an approach has the advantage that the SCWF waste form would be produced with the same technology used for MOX reactor fuel production. If integration of the private and government sectors can be accomplished, it may be possible to utilize the same facilities for both processes, as indicated in Fig. 1. If such integration of the MOX fuel and SCWF fabrication is not economically or politically feasible, duplication of the MOX fuel fabrication technology in the Pu recovery facility for SCWF fabrication

would very likely be faster and cheaper than development of a new immobilization technology and processing capability.

FEASIBILITY OF THE SINTERED CERAMIC WASTE FORM (SCWF) IMPLEMENTATION

The feasibility of the approach shown in Fig. 1 was assessed by assuming that the 50 t of projected excess Pu feed material consists of 40 t of relatively pure Pu suitable for reactor burning and 10 t of impure Pu to be discarded in the SCWF. The number of reactors required to burn 40 t of relatively clean Pu would depend on the reactor types selected and on the MOX Pu loadings, but should be within the range of available existing reactors. The 10 t of impure Pu would be blended and sintered with depleted UO_2 to form the SCWF pellets—essentially, "off-spec" unirradiated MOX pellets..

Figure 2 indicates the number of DWPF cans that would be required to hold the 2- to 4-L SCWF cans in the DWPF can-in-canister option, as a function of average Pu loading in the SCWF matrix. Since 5000 to 6000 DWPF canisters are projected for completion of the defense high-level waste immobilization mission, discard of the impure Pu within these canisters, with only a modest increase in the total of DWPF canisters, using the can-in-canister approach is possible with average Pu loadings of 0.5 wt% or higher in the SCWF. This loading is substantially lower than that in new MOX fuel and is comparable to that in normal spent LWR fuel. Higher Pu loadings (up to 5 wt%) would substantially reduce the number of 2- to 4-L cans that would have to be handled in the can-in-canister option and would reduce the disposition cost. Plutonium loadings of a few weight per cent should not have a significant repository impact as compared with SNF assemblies from the standpoint of criticality concerns, particularly during the post-closure period. In fact, the use of depleted UO_2 as a matrix material virtually eliminates long-term criticality concerns as the ^{239}Pu decays to ^{235}U. If the ^{239}Pu were contained in a non-uranium matrix, the possibility may occur for ^{235}U geological reconcentration as a result of long-term migration and waste form degradation; this could not occur after the ^{239}Pu decays with the use of a depleted UO_2 matrix.

Figure 2 also indicates the number of containers similar to the proposed multipurpose containers (MPCs)[6] that would be required to discard 10 t of impure Pu if the SCWF pellet products are mixed with a ^{137}Cs-loaded grout in a DWPF-like container or fabricated as canisters looking like MOX fuel assemblies and loaded five at a time into an MPC-like unit. No more than a few hundred MPC-like containers would be required for Pu loadings of a few weight per cent. This is consistent with the normal Pu loadings in both spent MOX fuel and normal SNF. Even at loadings as low as 0.5 wt%, fewer than 1000 MPC-like containers would be required.

CONCLUSIONS

Using the hybrid strategy of existing thermal reactors to burn clean excess Pu and fabricating a sintered ceramic waste form (SCWF), largely of depleted UO_2, for discard of impure excess Pu has the following advantages:

1. The SCWF mimics the chemical and morphological forms of unirradiated MOX LWR fuel and should have similar repository impacts over geological times. The SCWF should be as acceptable for repository disposal as spent MOX LWR fuel, or even spent commercial low enriched uranium LWR fuel.

2. The proposed SCWF pellet fabrication for impure Pu utilizes facilities and technologies virtually identical to those routinely used in Europe to fabricate mixed oxide (MOX) fuel, thus minimizing new required facilities or new technology development and demonstrations.

3. The proposed use of SCWF pellets to dispose of impure excess Pu eliminates the need to purify it, as would be required in any MOX reactor-based disposition strategy.

4. The SCWF provides a direct avenue for disposition of TRU scraps of impure Pu generated during the recovery of Pu from excess pits and for residues from MOX fuel fabrication. This avoids the need for additional processing facilities or operations for recovery and minimizes disposal of these Pu-containing residues.

5. Radiation barriers and the achievement of the spent fuel standard can be readily implemented by (1) using the can-in-canister approach with DWPF as the source of a glass containing a high-level waste radiation barrier, (2) encapsulating the SCWF pellets in a grout matrix containing ^{137}Cs in a DWPF type can in a small hot-cell facility, or (3) by mixing the SCWF pellets, packaged as a fuel-like assembly (or similar size container), with SNF from normal power reactor operations.

6. Use of a single waste form and fabrication process similar to unirradiated MOX for impure excess Pu and for MOX fabrication of the pure Pu for burning in existing reactors would reduce up-front costs for capital construction, research, development, and demonstration.

7. Because existing reactor capacity in the United States (or possibly Canada) would be utilized, and because no new technology development is required, this strategy would minimize the time required to achieve the spent fuel standard for excess Pu.

8. Adoption of the reactor burning approach may assist in developing effective linkages with reactor disposition approaches for pure excess Pu currently favored by the Russians.

9. MOX fuel fabrication and reactor burning can be time phased with the production of the SCWF fabrication to best utilize facility capacity and to optimize schedules. Multiple-

line MOX plants could be considered for simultaneous processing and fabrication of pure and impure excess Pu forms.

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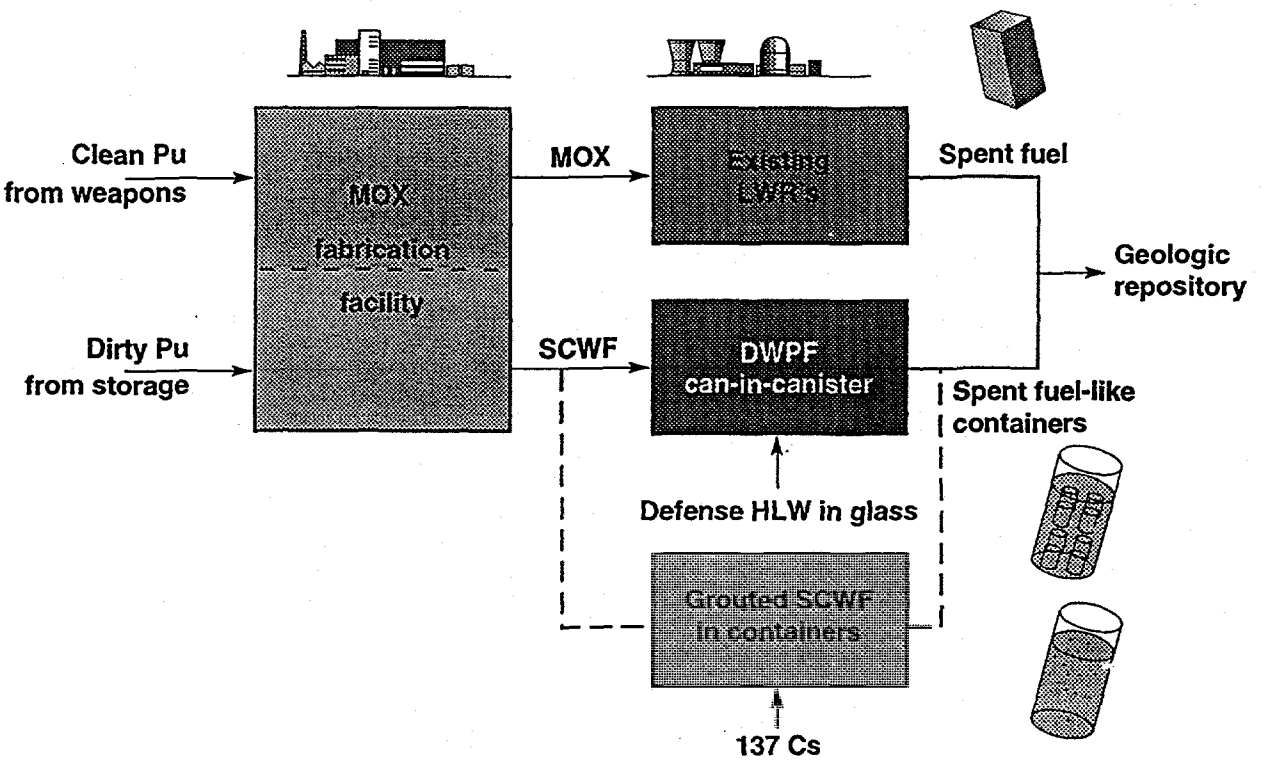

Fig.1 Conceptual disposition of the sintered ceramic waste form (SCWF) using a common MOX fuel fabrication facility

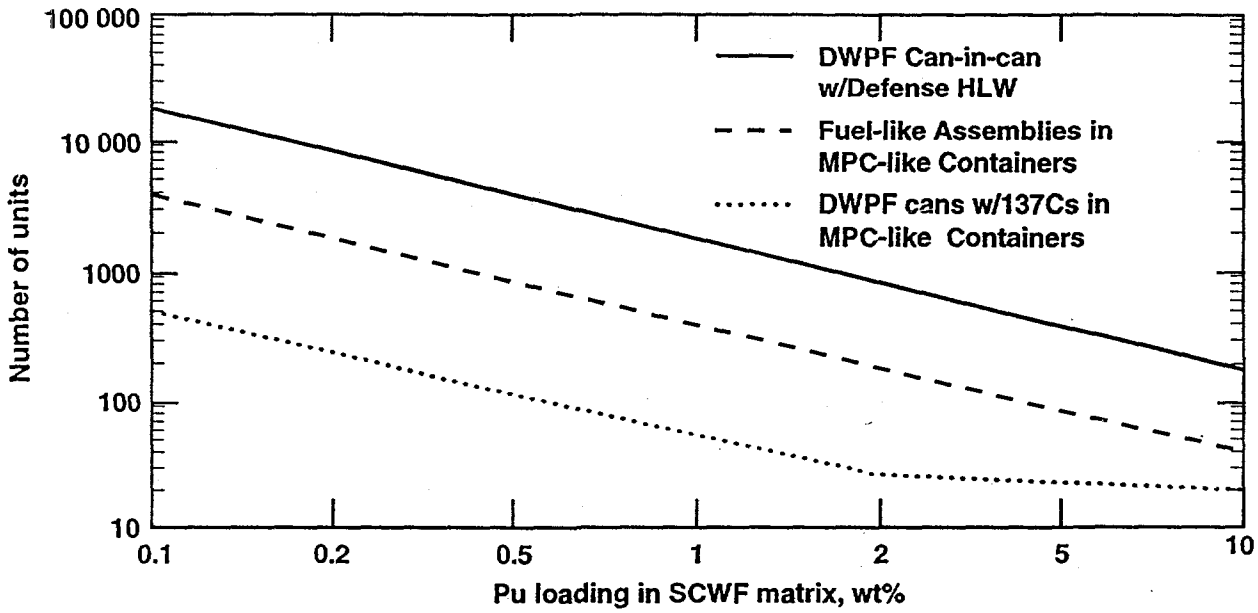

Fig. 2 Number of DWPF or MPC-like containers required to dispose of 10 t impure Pu vs Pu loading in SCWF matrix