

221670

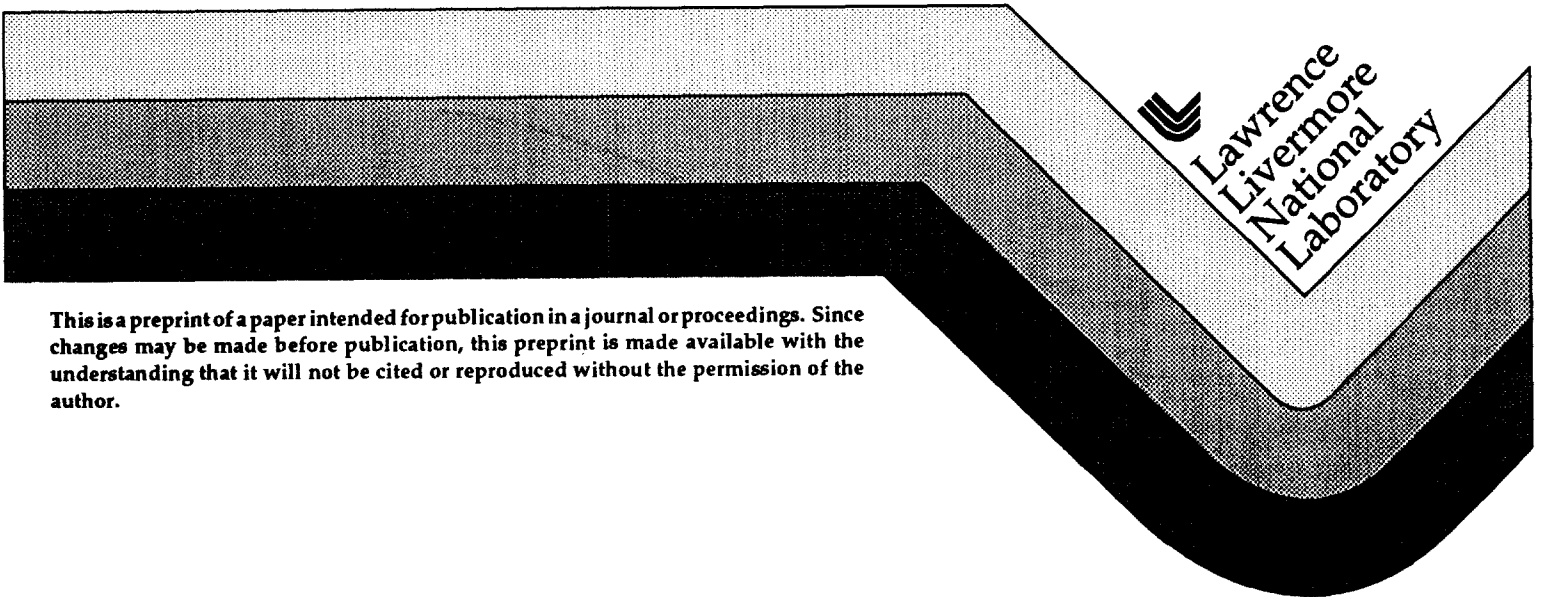
UCRL-JC-126837
PREPRINT

Plutonium Disposition Via Immobilization in Ceramic or Glass

L. W. Gray, T. Kan
H. F. Shaw, A. Armantrout

This paper was prepared for submittal to the
AICHE 1997 Spring National Meeting
Houston, Texas
March 9-13, 1997

March 5, 1997



DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

Plutonium Disposition Via Immobilization in Ceramic Or Glass

by

Leonard W. Gray

Tehmau Kan

Henry F. Shaw

Guy A. Armantrout

Lawrence Livermore National Laboratory

PO Box 5508, L-394

Livermore, CA 94551

Abstract

Safe management of surplus weapons-usable plutonium is a very important and urgent task with profound environmental, national and international security implications. The central, overarching goal is to render surplus weapons-usable plutonium as inaccessible and unattractive for reuse in nuclear weapons, as the much larger and growing stock of plutonium contained in civilian spent reactor fuel. One disposition option considered for surplus Pu is immobilization. Of the 72 waste forms originally considered, five immobilization base case variants, comprising glass and ceramic waste forms, are being evaluated to select the final form for Pu immobilization. Because of the potential reliance on existing facilities, the heterogeneous variants could potentially start more rapidly and be much cheaper than the homogeneous immobilization variants, if they can be shown to meet the non-proliferation objectives of the disposition program. Additional proliferation resistance can be engineered into the canister. The major issue for the can-in-canister approach is to determine whether glass or ceramics will be the plutonium form inside the small cans. A set of criteria and metrics has been established to aid in this determination. An R&D plan has been developed to determine the values of the metrics to be used in the down-selects and to field an operating plant.

Extended Abstract

The safe-management of surplus weapons plutonium is an important and urgent task with profound environmental, national, and international security implications. In the aftermath of the Cold War, Presidential Policy Directive 13, and various analyses by renown scientific, technical, and international policy organizations have brought about a focused effort within the Department of Energy to identify and implement paths for the long term disposition of surplus weapons-useable plutonium. The central goal of this effort is to render surplus weapons plutonium as inaccessible and unattractive for reuse in nuclear weapons as the much larger and growing stock of plutonium contained in spent fuel from civilian reactors. One disposition option being considered for surplus plutonium is immobilization, in which the plutonium would be incorporated into a glass or ceramic material that would ultimately be entombed permanently in a geologic repository for high-level waste.

Of the 72 waste forms originally considered, only borosilicate glass and a Zr-Ti-based ceramic are under active consideration today. Five different base cases are being evaluated to select the final form for plutonium immobilization:

- A) **Homogeneous Vitrification.** In this variant, plutonium would be mixed with glass frit containing neutron absorbers in a small, geometrically favorable melter to prepare a plutonium plus neutron absorber glass frit. This plutonium plus neutron absorber frit would then be mixed with additional glass frit, additional neutron absorber, and ^{137}Cs , and fed to a larger glass melter. The molten, intensely radioactive, plutonium-bearing glass would be poured into large stainless steel containers and allowed to harden. The resulting glass logs could be stored safely and securely for several decades, pending the availability of a geologic repository. Existing melters for vitrification of high level waste (HLW) are not appropriately designed for handling large amounts of plutonium. Either a new facility could be built on a green field (variant 1) or an additional "adjunct" melter (variant 2) could be added to the building housing the large HLW vitrification plant at the Savannah River Site.
- B) **Homogeneous Ceramic Immobilization.** In this variant, plutonium would be mixed with ceramic precursors, ^{137}Cs for a radiation barrier, and hot-pressed to form a multi-phased ceramic, very similar to the Synthetic Rock or Synroc first prepared by the Australians. While there is much less industrial experience with immobilization of nuclear waste in ceramic forms than there is with vitrification, the ceramic forms are expected to have excellent long-term performance in a geologic repository, as they are designed to be similar to natural minerals that have contained uranium and thorium for millions of years. A new facility would be built for this third variant.
- C) **Can-in-Canister Immobilization.** In the can-in-canister variants, plutonium would be immobilized in small cans of glass (variant 4) or ceramic (variant 5) without the addition of ^{137}Cs . These cans of plutonium glass or ceramic would be arrayed inside a large canister, such as the large canisters currently being used for HLW glass. The canister would then be filled with intensely radioactive waste glass, which contains ^{137}Cs and other radionuclides. The radiation field outside the canister would be similar to that in the homogeneous immobilization cases, but the radioactive fission products would not be mixed directly with the plutonium itself. Design studies are underway to ensure that it would be very difficult to remove the plutonium cans from the larger canisters. These two variants would rely on existing facilities for installation of the plutonium immobilization operations, and the existing vitrification operations for filling the canisters with HLW glass. Because of the simpler chemistry involved with separate Pu disposition forms and the media containing the ^{137}Cs and the reliance on existing facilities, these variants could potentially start more rapidly and be much cheaper than the homogeneous immobilization variants.

There are two major technical decisions that must be made before the immobilization alternative can be fully developed and implemented:

- Choice of the immobilization radiation barrier concept -the choice between a glass or ceramic form containing a homogeneous mixture of plutonium and fission products *versus* the external barrier concept in which cans of glass or ceramic containing plutonium are encapsulated within canisters of HLW glass (*i.e.*, the can-in-canister concept).
- Choice of the specific plutonium-bearing form/technology - the choice between a specially tailored borosilicate glass or a Synroc-like ceramic waste form and the appropriate formation technology for making the selected form (*i.e.*, melter technology for glass and either cold press and sintering or hot pressing for the ceramic form).

Due to their possible earlier start date and their cheaper cost, the can-in-canister variants appear to be the preferred variants if they can be shown to meet the non-proliferation objectives of the disposition program. Additional proliferation resistance can be engineered into the canister either using passive armor or by designing the inner can to be a "disappearing" alpha barrier. Passive armor can be added such as:

- Using depleted uranium rebar cages surrounding inner cans
- Armadillo passive armor around cages (similar to chain-mail armor used by King Arthur's knights and others)
- NATO spaced armor array - similar to Armadillo
- P-2000 Type

The disappearing alpha barrier concept would load ceramic pellets or glass marbles into a low melting temperature alloy container. These containers would then be loaded into the canister and molten HLW glass would be poured around the containers of ceramic pellets or glass marbles. The low melting temperature alloy would be melted by the heat of the molten glass and disgorge its contents, resulting in a more intimate mixture of the plutonium-bearing material and the highly radioactive HLW glass.

The second major issue for the can-in-canister approach is to determine whether glass or ceramic will be used as the plutonium-bearing material inside the small cans. A set of criteria and metrics has been established to aid in this determination and FY97 is being devoted to determining the values for the various metrics that will be used to decide between the two forms. The nine major criteria map directly onto the criteria used to make previous major decisions for the Disposition Program. An R&D plan has been developed to determine the values of the metrics to be used in the downselects and to field an operating plant. The metrics to be used and the progress to date on collection of the metric values will be discussed.

INTRODUCTION

Significant quantities of weapons-usable fissile materials (primarily plutonium and highly enriched uranium) have become surplus to national needs both in the United States and Russia. These stocks of fissile materials pose significant dangers to national and international security. The dangers exist not only in the potential for proliferation of nuclear weapons but also in the potential for environmental, safety, and health (ES&H) consequences if surplus fissile materials are not properly managed.

The Department of Energy (DOE) was directed to complete a comprehensive review of long-term options for surplus fissile materials storage and disposition, taking into account technical, nonproliferation, environmental, budgetary, and economic considerations. In furthering this policy, DOE's objectives included:

1. Strengthening national and international arms control efforts by providing an exemplary model for storage of all weapons-usable fissile materials and the disposition of surplus weapons-usable materials.
2. Ensuring that storage and disposition of weapons-usable fissile materials are carried out in compliance with ES&H standards.
3. Minimizing the prospect that surplus U. S. weapons-usable fissile materials could be re-introduced into the arsenals from which they came, therefore increasing the likelihood of reciprocal measures by Russia and other nuclear powers.
4. Minimizing the risk that surplus U. S. weapons-usable fissile materials could be obtained by unauthorized parties.
5. Accomplishing these objectives in a timely and cost-effective manner.

DOE announced that its evaluations leading to Records of Decision (RODs) would be carried out *via* the process depicted in Figure 1. Also, that any decision to commence implementation of surplus fissile materials disposition would be made in a broad domestic and international context taking into account any arrangements or agreements reached with the Russian government. These decisions will involve other Executive branch agencies in addition to DOE.

Screening Criteria

Screening criteria were developed based on the policy objectives articulated in the President's Nonproliferation and Export Control Policy of September 1993 and the January 1994 "Agreement between the United States and Russia on Nonproliferation of Weapons of Mass Destruction and their means of Delivery," as well as the analytical framework established by the National Academy of Sciences in their study on disposition of surplus plutonium. Before finalizing the original criteria, DOE obtained public input on the screening criteria to be utilized and the options to be evaluated. As an initial step in the NEPA process, and as announced in the NOI, a series of Scoping Meetings were held at twelve locations across the country from August to October 1994. Members of the

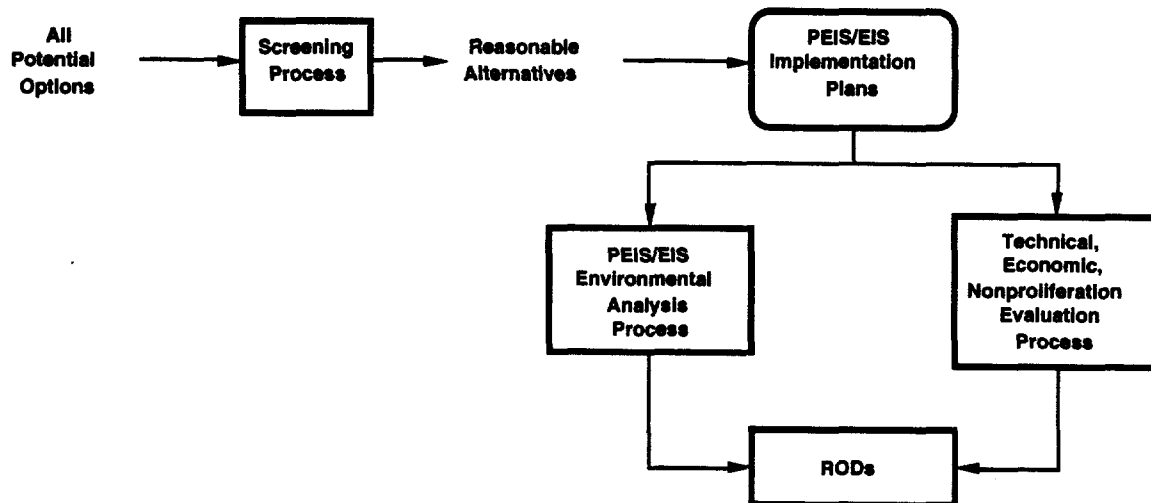


Figure 1. Evaluation Process Leading To Records of Decision.

interested public were briefed by DOE on the overall long-term storage and disposition considerations, criteria, and evaluation process, including the screening process. During these meetings, questionnaires were provided for public input on (1) the validity and relative importance of the criteria, and (2) additional criteria that should be considered. The responses were evaluated and the revised criteria were documented in the "Summary Report of the Screening Process", DOE/MD-0002. March 29, 1995. Based upon all the available input, the following set of criteria have been used or rating individual options:

1. Resistance to Theft and Diversion by Unauthorized Parties
2. Resistance to Retrieval, Extraction, and Reuse by the Host Nation
3. Technical Viability
4. Environmental, Safety, and Health
5. Cost Effectiveness
6. Timeliness
7. Fosters Progress and Cooperation with Russia and Others
8. Public and Institutional Acceptance
9. Additional Benefits

Immobilization Form Screening

A search of the literature identified 72 waste forms that have previously been considered for immobilizing radioactive wastes. These individual forms were grouped into families that share common chemical and physical characteristics. The families were: (1) calcine, (2) cementitious, (3) ceramic, (4) glasses, (5) glass-ceramic, (6) metallic, (7) multibarrier, and (8) polymeric. Elimination of redundancies (e.g., shape differences, geometric variations) resulted in a list of 45 unique waste forms that were considered in the screening process.

To avoid problems normal with *ad hoc* screening techniques, a balanced approach was selected for Pu Immobilization prescreening that allowed a rapid survey of the waste form alternatives and provided a formal, structured mechanism for selecting candidates for further analysis and consideration. A two-stage approach, based on formal decision analysis techniques, was adopted. Exclusionary (pass/fail) screening (based upon regulatory guidelines, in this case primarily the Code of Federal Regulations (CFR), was used for the first stage. Only waste forms that met all criteria were retained; failure of any criterion resulted in elimination. The second stage evaluated the remaining waste forms more carefully with the goal of selecting a small set of the best forms for further detailed evaluation. This second stage involved a more formal comparison of waste form characteristics using multi-attribute utility analysis techniques. The result of the final stage of screening was to recommend glass, ceramic and electrometallurgical treatment as the preferred options for further study.

Processing Variants

The screening of alternative candidate immobilization forms was documented in "Screening of Alternative Immobilization Candidates for SFM Disposition," UCRL-ID-118819, February 9, 1996. The three immobilization forms can be produced by more than one method. After careful study it was determined that the three immobilization forms could be prepared by at least six different methods. These methods are listed in Table 1 and described in more detail below. These are the variants that were analyzed in the Draft Programmatic Environmental Impact Statement (PEIS).

Table 1. Immobilization Technology Variants

Radiation Barrier Approach	Technology options
Homogeneous or Internal Radiation Barrier	<ol style="list-style-type: none"> 1. Glass (New Facilities) (Greenfield) 2. Glass - Adjunct Melter (Existing / New Facilities) 3. Ceramic -New Facilities (Greenfield) 4. Electrometallurgical Treatment (Existing / New Facilities)
External Radiation Barrier (Can-in-canister)	<ol style="list-style-type: none"> 5. Glass (Existing Facilities) 6. Ceramic (Existing Facilities)

- A) **Homogeneous Vitrification.** In this variant, Pu would be mixed with glass frit containing neutron absorbers in a small geometrically favorable melter to prepared a Pu-neutron absorber-glass frit. This Pu-neutron absorber frit would then be mixed with additional glass frit containing neutron absorber, and ^{137}Cs , and fed to a larger glass melter. The molten, intensely radioactive Pu-bearing glass would be poured into large stainless steel containers and allowed to harden. The resulting glass logs could be stored safely and securely for several decades, pending the availability of a geologic repository. Existing melters for vitrification of high level waste (HLW) are not appropriately designed for handling large amounts of plutonium. Either a new facility could be built on a green field (variant 1) or an additional "adjunct" melter (variant 2) could be added to the building housing the large HLW vitrification plant at the Savannah River Site.
- B) **Homogeneous Ceramic Immobilization.** In this variant, Pu would be mixed with ceramic precursors, ^{137}Cs radiation barrier and hot pressed to form a multi-phased ceramic, very similar to the Synthetic Rock or Synroc first prepared by the Australians. While there is much less industrial experience with immobilization of nuclear waste in ceramic forms than there is with vitrification, the ceramic forms are expected to have excellent long-term performance in a geologic repository, as they are designed to be similar to natural minerals that have contained uranium and thorium for millions of years. A new facility would be built for this third variant.
- C) **Electrometallurgical Treatment.** In the Electrometallurgical variant, Pu metal and oxides would be converted to chlorides, dissolved in molten NaCl-KCl, sorbed on zeolites, and then immobilized in a glass-bonded-zeolite (GBZ) waste form. The immobilization operations would be integrated with operations in the ANL- West hot cells to treat DOE-owned spent fuel. The fission products from these fuels would contribute some radiation to the immobilization forms, but ^{137}Cs from the Hanford capsules would provide most of the radiation field needed to create a radiation barrier.
- D) **Heterogeneous (Can-in-Canister) Immobilization.** In the can-in-canister variants, Pu would be immobilized in small cans of glass (variant 4) or ceramic (variant 5) without the addition of ^{137}Cs . These cans of plutonium glass or ceramic would be arrayed inside a large canister, such as the large canisters currently being used for HLW glass. The canister would then be filled with intensely radioactive waste glass, which contains ^{137}Cs and other radionuclides. The radiation field outside the canister would be similar to that in the homogeneous immobilization cases, but the radioactive fission products would not be mixed directly with the Pu itself. Design studies are underway to ensure that it would be very difficult to remove the plutonium cans from the larger canisters. These two variants would rely on existing facilities for installation of the Pu immobilization operations, and existing vitrification operations for filling the canisters with HLW waste. Because of this potential reliance on existing facilities, and the fact that the immobilization of the Pu can be accomplished in glove boxes, rather than hot cells, these variants could potentially start more rapidly and be much cheaper than the homogeneous immobilization variants.

As additional engineering information has become available in the time since the screening process and the writing of the Draft PEIS, the definition and our understanding of how these alternatives might be implemented have matured. Each immobilization variant has been defined for analysis as the beginning-to-end set of operations (*e.g.*, from surplus plutonium to geologic disposal) necessary to address all of the surplus weapons-usable plutonium. We defined and developed the sequence of operations that is necessary to accomplish the immobilization of materials at a much greater level of detail than was used for either the Screening Report or the NAS Report. The following information was assembled for each of the immobilization variants analyzed:

- Block flow diagrams describing process steps for all operations.
- Lists of major equipment and facilities to accomplish each immobilization function.
- Mass balance and rate data for unit operations and facilities.
- Sketches of equipment layouts and plot plans.
- Reviews of regulatory and operational considerations for facilities.
- Estimates of facility sizes, personnel requirements, and facility infrastructure requirements.
- Identification of balance of plant requirements.

This defined the immobilization variants in sufficient detail to permit technical assessments to be performed, and allowed the analysis of the variants with respect to technical, cost, and schedule criteria. The team also performed necessary experimental and development work required to enhance the knowledge base of immobilization, such as:

- Engineering-scale fabrication of ceramic waste forms with plutonium.
- Full-scale "cold" (*i.e.*, without any radionuclides) demonstration of the can-in-canister concept.

Based upon analysis of these six remaining variants, the following four were recommended to be dropped from further consideration:

1. Adjunct Melter
2. Greenfield Vitrification
3. Electrometallurgical Treatment
4. Greenfield Ceramics

The remaining major decision that must be made before the immobilization alternative can be fully developed and implemented is the choice between a specially tailored borosilicate glass or a Synroc-like ceramic waste, and the appropriate formation technology for making the selected form (*i.e.*, melter technology for glass and either cold press and sintering or hot pressing for the ceramic form).

Choice: Glass vs. Ceramics

It is likely that the choice between glass and ceramics will be based primarily on the selection criteria involving Technical Viability and Proliferation Resistance.

Technical Viability

There must be a high degree of confidence that an alternative will be technically successful. It is therefore of interest to rely on technologies that have been proven for similar applications and have a high likelihood of success. New technologies (or new applications of old technologies) may also require an extended period for licensing or regulatory approval due to the immaturity of the process or regulatory framework. This includes the state, readiness and projected lifetime of facilities and infrastructure and the processing/storage/disposal capacity of the facilities.

There are a number of factors that must be considered in evaluating the technical viability of a disposition alternative. These factors include the technical maturity of the processes and technology used in the alternative; the level of risk involved in advancing the existing state of technology of an alternative to a state ready for deployment in a production facility; and the acceptability of a disposition form for disposal in a HLW repository.

The technical maturity of an alternative will be measured by an assessment of the readiness for deployment of the immobilization process(es). Technologies that are less mature may require a number of years to prove themselves and increase the risk that they will not meet technical goals, be adequately mature for deployment, or be more costly and take more time than projected. Readiness for deployment will be assessed based on the stage of development, as measured using the seven stages outlined in Table 2. (This is an adaptation of the gate process used by both the Department of Defense and the DOE EM Office of Science and Technology). The current stage of development is based on whether all of the criteria for completion of the previous stage have been met.

Viability risks refer to the programmatic risks associated with getting the process from a less mature Stage to Stage 7. Selection of a form or process which is less mature requires acceptance of greater programmatic risk. These include risk that programmatic objectives will not be met, and uncertainties in cost and schedule inherent in readying an immature form or process for deployment. The viability risk will be based upon expert judgment utilizing comparisons to similar unit operations, where available, that are in use in various industries today. The seven stages in Table 2 will be utilized to determine these risks.

The immobilized plutonium must be acceptable for disposal in a Federal geologic repository for HLW. Acceptability of the disposal form for disposal is a fitness-for-purpose criterion that has regulatory and licensing implications as well as long-term, post-

Table 2. Stages of Technology Development

<u>Stage of Development</u>	<u>Criteria for completion</u>
1 Basic research	Benchtop cold experiments are successful.
2 Applied research	Benchtop experiments with radioactive material are successful. Process steps have been successfully demonstrated with expected average feedstock (success means that desired product has been produced - in terms of product quality - with the required loading of Pu; or that the output of the process step - for example, pretreatment - has achieved its desired goals).
3 Exploratory development	Performance and costs of process and product are reasonably established. Product requirements are clearly defined. Integrated demonstrations on a lab-scale with the full range of radioactive feedstocks (of the appropriate chemical and physical forms) expected to be processed have been successfully completed.
4 Advanced development	All showstoppers have been eliminated. Costs are favorable. The program is ready to proceed to develop functional design requirements for the process or form. At the completion of this stage, there should be confidence that the process can be operated reliably in a glove box or remote environment.
5 Engineering development	Functional design requirements have been developed. Site readiness issues are identified, and addressed (Thus, one cannot exit this stage until a site is selected). This includes provision for disposition of off-specification products and for treatment of secondary wastes.
6 Demonstration	The process and form are successfully demonstrated on a large enough scale so that implementation is straightforward (<i>i.e.</i> , there are no scale-up issues). This can be achieved either radioactively or non radioactively, with the radioactive option being preferred. The quality control/quality verification program for the product is validated. No technical issues remain which will impart risk to implementation (<i>e.g.</i> , testing has been sufficiently rigorous so that there is confidence that the process or form will tolerate credible upset conditions).
7 Implementation	Plant operation.

emplacement performance implications. There are a number of regulatory requirements that must be satisfied by the disposal form as pass-fail criteria for acceptability of the disposal form for disposal in a geologic repository. These requirements are listed in Table

3. With the exception of requirement 1.4, which is discussed more extensively below, these pass-fail requirements are the same for all variants, and hence not discriminators.

Table 3. Regulatory requirements for immobilized forms

Criterion	Requirement	Basis
1.1 No free water	"Shall not contain free liquids in an amount that could compromise the waste package"	10CFR 60.135(b)(2)
1.2 Solidification and consolidation	"Shall be... in solid form... [and]... consolidated...to limit the availability and generation of particulate"	10CFR 60.135(c)(1&2)
1.3 Stability	"Shall not contain explosive or pyrophoric or chemically reactive materials in an amount that could compromise the waste package.." and "...shall be noncombustible.."	10CFR 60.135(b)(1)&(c)(3)
1.4 Criticality control	" K_{eff} must ... show at least a 5% margin"	10CFR 60.131(b)(7)
1.5 RCRA metal content	Cannot contain significant quantities of the following free metals: arsenic, barium, cadmium, chromium, lead, mercury, selenium, silver	40CFR 261.24

In addition to minimum regulatory and licensing requirements for repository acceptability set forth in Table 3, there may be substantial differences between the long-term, post-emplacement ES&H performance of different disposal forms. If present, these differences could provide a basis for differentiating between disposal forms and selecting a preferred form. The two main long-term, post-emplacement performance considerations affecting repository acceptability are criticality safety and the potential for contamination of the biosphere by the release and transport of radioactive materials to the accessible environment.

At a minimum, the disposal form emplaced in the geologic repository must remain criticality safe over the regulatory performance period. In the U.S., the current regulatory performance period for HLW and spent fuel in a geologic repository has been specified as 10,000 years. (The pertinent regulations are currently under review, and this may change.) However, the emplaced plutonium and its ^{235}U decay product remain fissile over much longer periods (Pu - hundreds of thousands of years, ^{235}U - billions of years).

Notwithstanding the complexity of the problem, key properties of disposal forms that affect criticality safety can be identified for the purpose of selecting a preferred disposal form. The concentration of neutron absorbers, and neutron absorption properties of the immobilization matrix are key parameters. The characteristics of the degradation mechanisms and rates of the glass and ceramic disposal forms may be very different and are affected by the compositions of the water and disposal form, the solubility of the constituents, the active surface areas available for reaction with water, the compositional and thermal stability of disposal forms, the physical and chemical homogeneity of disposal forms, and radiation effects (radiation damage in the disposal form and radiolysis in water).

The resistance to release and transport by groundwater of disposal form constituents (fissile elements, matrix chemicals/phases and neutron absorbers), and rates and relative timing of releases of different components are also key parameters. However, it is important to recognize that under most conditions, the actual release of a radionuclide or neutron-absorbing element from the waste package will be controlled by the solubility of that element in groundwater, except under conditions of high groundwater flux. (Under high-flux conditions, it is the degradation rate of the disposition form itself that will control release.) To first order, the solubility, and thus the release of elements from a waste package is independent of the composition of the disposition form. Resistance to degradation in the geologic environment is, nevertheless, an important measure of repository acceptability because as long as the form remains substantially in its "as-emplaced" state, it is much easier to assure that the system remains safe with respect to criticality. Once the disposition form has degraded to secondary alteration products, the problem of assuring long-term criticality safety becomes much more complex, and one must be able to model the geochemical behavior of all the important solid and aqueous phase chemical species in the system. Such calculations are routinely carried out as part of repository performance assessment, but require comprehensive sets of thermodynamic data. In many cases, requisite data are lacking, and the resulting calculations are subject to considerable uncertainty.

In addition to remaining criticality safe, the radionuclides present in the disposal form must remain isolated from the biosphere over the performance period. The performance period for isolation of the radionuclides present in HLW and spent fuel in a geologic repository in the U.S. is currently 10,000 years. (The pertinent regulations are currently under review, and this may change.) Contamination of the biosphere can occur through slow dissolution of the emplaced disposal form by flowing groundwater which may transport it towards the biosphere. In a well sited and designed repository, a low groundwater infiltration rate, together with sorption and dispersion of radionuclides during transport through a large geologic barrier, among other factors, are expected to prevent contamination of the biosphere by the radionuclides.

The key properties of a disposal form that affect radionuclide migration can be identified for the purpose of selecting a preferred disposal form without detailed consideration of the complex problem of possible contamination of the biosphere. These factors are largely the same as those necessary for an analysis of criticality safety.

Metrics for determining repository acceptability have been proposed that include consideration of the as-emplaced effective thermal neutron cross section, the active surface area of the as-emplaced form; the maximum possible reaction (degradation) rate of the form with groundwater, the change in reaction rate due to radiation damage of the material, a measure of the sensitivity of the rate of reaction as a function of changes in chemical environment (pH), and the potential for the production of colloidal particles, which could provide a non-solubility-limited release mechanism for radionuclides. Experiments and studies are underway to provide quantitative estimates of the relevant metrics and the resulting data will be used as input to the form downselection process.

Proliferation Resistance

Because many nations and perhaps even some well funded sub-national groups have or could obtain the technical resources to produce a nuclear weapon if they can obtain sufficient fissile materials, the ideal disposition method would be one that would totally eliminate the surplus plutonium from the face of the Earth. Unfortunately, developing the technology to accomplish this would take more time than the world can afford. The U. S. National Academy of Sciences deemed this surplus plutonium a "clear and present danger" to national and international security. The NAS proposed to minimize the risk by transforming the plutonium into a form that is as unattractive and roughly inaccessible as plutonium in the larger and growing stockpile of commercial spent nuclear fuel. This "spent fuel standard" has been accepted by the governments of the U. S. and Russia, and accepted by the governments of the G-7 plus one nations meeting in Moscow in April, 1996, as a practical solution to the excess plutonium stockpile.

There are two primary proliferation concerns: one is reuse of the plutonium by the Host Nation, the other is acquisition of the plutonium by unauthorized parties, *i.e.* rogue nations and sub-national groups. The Host Nation is not considered to be a near term threat because:

- Both nations retain significant stockpiles of weapons
- Both nations retain unverified quantities of Pu in strategic reserves

Furthermore, the initial stages of dismantlement and disposition will be carried out beyond purview of accepted international monitoring capabilities. In addition, breakout for rearming host nation's nuclear weapons stockpile would only be likely should disarmament proceed to the point at which there were no strategic reserves to replenish a

nation's nuclear arsenal. Breakout is arbitrarily defined as recovery of 1.0 tonne of fissile material.

The heightened concern over the possibility of unauthorized parties acquiring nuclear materials has developed because:

- There is a developing global market for Special Nuclear Materials (SNM)
- Non-weapons states have declared a desire to achieve nuclear power status
- Political, ethnic and religious realignments have changed the balance of power. This has increased the resolve of some nations to acquire the status of nuclear powers
- Economic disruptions have occurred within some nuclear powers. This has improved conditions for an illicit global market for SNM
- Multiple insider threat is more credible than in the past
- Lack of knowledge is no longer an enduring barrier to unauthorized threats
- Sub-national groups have access to billion-dollar resources
- Disarmament is downsizing the nuclear weapons design, development and manufacturing community
- There is an ever-increasing proliferation of the detailed information needed to acquire nuclear materials and to fabricate weapons
- Car/truck bombs, and chemical/biological agents have become credible terrorist tools
- A willingness to die or kill for a cause has been frequently demonstrated by the suicidal attacks of dedicated terrorist groups

Proliferation resistance can generally be thought of as the barrier that must be overcome to acquire nuclear weapons. Larger barriers typically impose greater risks to the proliferants, require greater resources to achieve the desired outcome, require longer timelines to accomplish the goal, and in general require a greater level of effort on the part of the proliferant. Unfortunately, proliferation resistance is not a directly measurable or unambiguously calculable quantity. Unequivocal techniques for determining the adequacy of a system's proliferation resistance do not exist.

When one looks at the "spent fuel standard" in the context of the real world, one must realize that plutonium in all stages of all the disposition alternatives could be made weapons usable, should sufficient fissile materials be obtained. One must also realize that the technology for the recovery of plutonium from spent fuel is documented in the open literature in all of the major languages. This technology is readily adaptable for other material forms of plutonium and that, if the environmental, safety, and health standards of the western world were not followed, the resources required for plutonium recovery would be relatively modest. There are, however, three major discriminators between plutonium-containing materials:

1. The presence of a radiation barrier sufficient to require shielding during theft and processing to prevent death.
2. The presence of a radiation barrier sufficient to force mechanical manipulation of the material behind shielding.

The extent of the chemical processing necessary for recovery and purification of the plutonium prior to its reuse in weapons.

There are a number of energetic materials that are available as tools to terrorist group trying to acquire nuclear materials by covert actions. These include the list shown in Table XX.

Table XX. Energetic Materials Available for use in stealing nuclear materials

<u>Material</u>	<u>Primary use</u>
Conical shaped charge	Penetration / perforation
Linear shaped charges	Cutting steel
High explosives	Breaching charges
Low explosives	Lifting charge or bursting
Burn bars	Thermite cutting of metals and concrete

Energetic materials work best when attacking simple targets, e.g., perforation of homogenous materials. Proper design of the energetic attack requires knowledge of the design of the target and the materials of construction.

Knowledge of possible energetic attack, also allows the construction of the package in a way that is protective of energetic attack; this would of course delay any terrorist group trying to steal nuclear materials. The heterogeneous nature of the can-in-canister option adds to the complexity of designing an energetic attack. A breaching charge designed to cut the steel casing of the DWPF canister and crush the HLW glass, would not cut the rebar used to make the support and cages surrounding the cans. To cut these, a second phase of energetic cutting would be necessary. For example, using depleted uranium rebar to make the support and cages would greatly extend the time necessary to remove the cans from the canister. Passive armor such as Armadillo type armor (similar to chain mail), or type 2000 used by the Israelis military services would shred the energetic jets much like shredding cheese or carrots on a kitchen shredder.

Another way to guard against an energetic attack is to prepare the form either as small pellets for ceramics or as marbles for glass and load these into a low melting alloy can. (This low melting can would be used only as an alpha barrier while the cans were being prepared and loaded into the canister.) When the molten HLW glass is then poured around the immobilized plutonium, the alpha barrier will melt and disgorge the immobilized forms into the molten glass matrix. An energetic attack would result in further disintegration of the immobilized forms and further mixing with the HLW glass.

A vulnerability risk analysis will be used to determine just how far and what protective measures should be taken to assure that the surplus fissile material is sufficiently difficult to retrieve.

Conclusions

The Excess Fissile Materials Disposition Program has published both the PEIS and a ROD which selected a dual path of Immobilization and MOX fuel as the preferred routes to manage the growing stockpile of excess weapons-usable plutonium. The Immobilization portion of this program has gone through a series of downselects to arrive at a heterogeneous immobilization form in which the plutonium is immobilized in small cans in a matrix of either ceramics or glass. These cans will be surrounded by HLW glass at either Hanford or Savannah River. A September downselect is planned to select between glass or ceramics. A R&D plan has been developed and is being worked to collect sufficient technical data to allow a technically based selection to be made between ceramics and glass. DOE/MD will publish a second EIS which will select between Hanford and Savannah River with Savannah River being the preferred site. The overall program is on a fast track schedule to bring an immobilization facility on-line at the end of FY2004 or early FY2005.

Technical Information Department • Lawrence Livermore National Laboratory
University of California • Livermore, California 94551

