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IMMOBILIZATION NEEDS AND TECHNOLOGY PROGRAMS

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ABSTRACT

In the aftermath of the Cold War, the U. S. and Russia agreed to large reductions in nuclear weapons. To aid in the selection of long-term management options, DOE has undertaken a multifaceted study to select options for storage and disposition of plutonium in keeping with U. S. policy that plutonium must be subjected to the highest standards of safety, security, and accountability. One alternative being considered is immobilization. To arrive at a suitable immobilization form, we first reviewed published information on high-level waste immobilization technologies and identified 72 possible plutonium immobilization forms to be prescreened. Surviving forms were further screened using multi-attribute utility analysis to determine the most promising technology families. Promising immobilization families were further evaluated to identify chemical, engineering, environmental, safety, and health problems that remain to be solved prior to making technical decisions as to the viability of using the form for long-term disposition of plutonium. From this evaluation, a detailed research and development plan has been developed to provide answers to these remaining questions.

INTRODUCTION

In the Cold War aftermath, significant quantities of enriched uranium and weapons-grade plutonium have become surplus to national defense needs in both the U. S. and Russia. Nuclear powers are now faced with management of tonnes of Pu in excess to national security needs. These excess stockpiles pose a danger to national and international security, not only in the potential proliferation of nuclear weapons but also in the potential for environmental, safety, and health consequences if these fissile materials (FMs) are not properly managed.

If agreed reductions are implemented, perhaps 100 tonnes of Pu will no longer be needed for military purposes by Nuclear Weapons States. Continued implementation of arms reduction agreements will result in further dismantling of weapons and increasing stockpiles of surplus weapons-usable materials.

Because of the economic and social challenges faced by the new states formed by the transformation of the Former Soviet Union (FSU) into the Commonwealth of Independent States, there is a serious risk of nuclear proliferation from the resulting growing stockpiles. Nuclear weapons or fissile materials could fall into the hands of terrorists or non-nuclear nations through theft or diversion of FMs. The U.S. National Academy of Sciences (NAS) report[1] on the management and disposition of excess weapons plutonium characterized this as a "clear and present danger." This nuclear danger is, in many ways, more diffuse, harder to manage, and more dangerous than the nuclear tensions of the Cold War era.

On January 24, 1994, in response to the President's nonproliferation policy, Secretary O'Leary created a Department of Energy (DOE)-wide project for control and disposition of surplus fissile materials which later became the Office of Fissile Materials Disposition (MD).

MD, through task teams composed of experts from national laboratories, production sites, universities, industry, and other DOE programs, e.g. Civilian Radioactive Waste Management, (RW), have used a process that considered potential long-term storage and disposition options, evaluated them against screening criteria, and identified alternatives reasonable for continued evaluation in a Programmatic Environmental Impact Statement (PEIS) process. Screening criteria, which were developed with input from the public, reflect the President's Nonproliferation and Export Control Policy of September 1993, the January 1994 Joint Statement by Presidents Clinton and Yeltsin on Nonproliferation of Weapons of Mass Destruction and the Means of Their Delivery and the analytical framework established by the NAS.

To aid in selecting long-term management options for surplus weapons Pu, DOE has undertaken a multi-faceted study to select options for storage and disposition in keeping with U. S. policy that excess Pu must be subjected to the highest standards of safety, security, and international accountability. The primary goal is to render weapons-usable fissile materials as inaccessible and unattractive for weapons fabrication as that in commercial reactor spent fuel (i.e. meet the "spent fuel standard") while protecting human health and the environment. Disposition is defined as a process of use or disposal of materials that results in the remaining material being converted to a form substantially and inherently more proliferation-resistant than the original form. Disposition options must take into account technical, nonproliferation, environmental, and economic considerations.

Disposition can be divided into three distinct but overlapping phases—dismantling, intermediate storage, and long-term disposition (Fig. 1). Dismantling of U.S. and FSU weapons and storage of resulting surplus fissile materials (SFM) are already under way. Conversion of residue materials and long-term disposition of all FM will take far longer to accomplish.

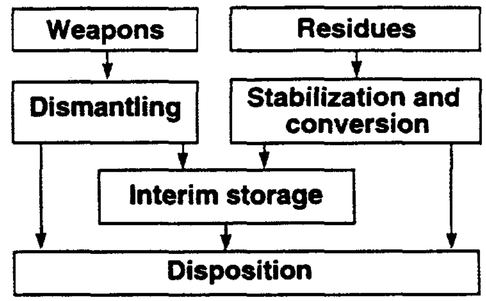


Figure 1. Steps in control and disposition of surplus fissile materials.

Immobilization Program

One class of disposition alternatives is immobilization. In these alternatives, surplus Pu would be immobilized in an acceptable matrix to create a chemically stable form for disposal in a high-level waste repository (or other alternative disposal system). The radiation level of the immobilized form would also meet the "spent fuel standard" in that fissile material would be mixed with high-level wastes or other radioactive isotopes and immobilized to create a radiation field that could serve as a proliferation deterrent comparable to commercial spent nuclear fuel.

MD selected the Lawrence Livermore National Laboratory as Lead Laboratory to study and recommend methods for transformation of SFM into long-term immobilized forms meeting environmental, safety, and security objectives; to provide appropriate input to other Disposition Tasks Teams so as to assess technical feasibility of immobilization as a long-term disposition option; and describe infrastructures required to conduct disposition of SFM. Support laboratories include Savannah River Technology Center, Argonne National Laboratory, Oak Ridge National Laboratory, and Pacific Northwest Laboratory. The team also includes support from US universiti-esand industries.

As part of the disposition program, other nations with relevant interests and experience have been invited to participate in the disposition study. Australia, the United Kingdom, France and Russia are participants in the Immobilization Program.

Immobilization would embed Pu in a tailored ceramic, glass, or other suitable material, alone or mixed with radioactive fission products to produce a suitable disposal form. To be viable, the Pu concentration of the form must be in the range of 1.0 to 10 wt% range. To arrive at suitable forms, published information on HLW immobilization technologies was reviewed; 72 uniquely named forms were identified. After prescreening, the 16 surviving forms were screened using multi-attribute utility analysis (MAUA) to determine the more promising technologies. Promising immobilization families (glass, ceramics and metals) were further evaluated to identify and seek solutions for chemical, engineering, environmental, safety, and health (ES&H) problems remaining to be solved prior to making technical decisions as to their viability for long-term disposition of Pu. We are also assessing modifications required to existing U.S. high-level waste immobilization approaches, and modifications required to the DOE response to the Defence Nuclear Facilities Safety Board's Recommendation 94-1, ES&H implications, costs, and schedule.

PROCESSING OPTIONS: Five immobilization base case options comprising glass and ceramic forms are being evaluated in the PEIS/ROD process:

- Vitrification
  - Internal radiation barrier (Fig 2)
    (1) a new greenfield facility that produces a borosilicate glass containing Pu, neutron absorbers, and ^{137}Cs (as a radiological barrier), and then encapsulates this glass in a storage canister;

    (2) an adjunct melter to the existing Defense Waste Processing Facility (DWPF) that produces a glass containing Pu, neutron absorbers, and high level waste (HLW), and then encapsulates this glass in a storage canister;

  - External radiation barrier (Fig 2)
    (3) a "can-in-canister" variant, in which an inner can containing a Pu- and neutron-absorber-bearing glass is surrounded by a glass containing a radiological barrier, which, in turn is contained in an outer storage canister.
- Ceramics
  - Internal radiation barrier (Fig 3)
    (4) a new ("greenfield") facility that produces a ceramic containing Pu, neutron absorbers, and ^{137}Cs and then encapsulates the ceramic in a storage canister;

  - External radiation barrier (Fig 3)
    (5) a "can-in-canister" alternative in which an inner can of a ceramic containing Pu and neutron absorbers is surrounded by a ceramic or glass that contains a radiological barrier, which is in turn contained in an outer storage canister.

Several variant to the base cases are also being examined. These include use of either plutonium oxide (base case) or plutonium nitrate solution as the feed to the melters. The base case for the ceramics is plutonium nitrate feed; the variant is the use of plutonium oxide feed.

FEED STOCKS: When the Cold War unexpectedly and abruptly ended in 1991, DOE stopped production of plutonium for nuclear weapons —it had already in effect stopped production of the warheads themselves in 1988. Facility missions were abruptly changed and budgets were dramatically reduced.

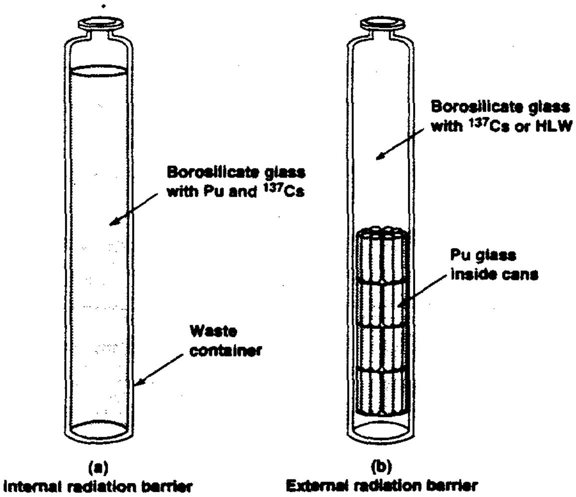
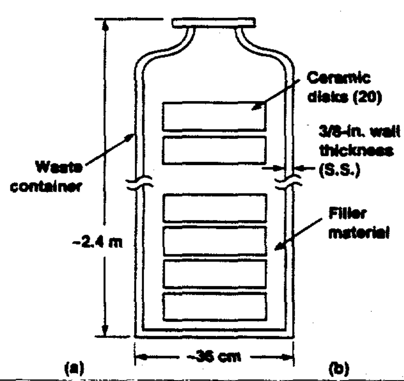


Figure 2. Two options for vitrification of Pu



	Internal radiation barrier	External radiation barrier
Ceramic	Ceramic with Pu and ^{137}Cs	Ceramic with Pu
Filler	Inert material (TiO_2 powder)	Solidified $Na_2Ti_3O_7$ with ^{137}Cs

Figure 3. Two options for immobilizing Pu in a ceramic

Tonnes of plutonium, previously slated for warheads, were left in place, much of it in forms and facilities not suited to long term storage. There are approximately 26 tonnes of Pu, not including amounts contained in assembled and disassembled weapons, located throughout the weapons complex. The alpha decay of Pu results in formation of free radicals which induce chemical reactions within stored solutions and solids containing organics (plastics, paper, cloth, etc.) that make behavior of these solutions and solids difficult to predict. The Defense Nuclear Facilities Safety Board (DNFSB) recognized this when they issued their recommendation 94-1, in May 1994, calling on DOE to:

- bring stored plutonium metal and oxide into conformance with the DOE storage standard
- process the plutonium solutions into forms safer for interim storage
- process possibly unstable plutonium residues into forms suitable for interim storage
- establish a research program to help choose among candidate processes for conversion to interim forms and longer-term disposition.

Under the assumption that long term storage and ultimate disposition of plutonium are not considered to be technologically feasible during this century, current DOE programmatic efforts are focusing corrective actions on stabilizing stored materials in response to the DNFSB recommendations. Materials returning from the nuclear stockpile, together with these existing inventories of fissile materials, which may easily be converted to a weapon usable form, define the scope of materials that have generated a concern for international security. It is assumed that corrective actions will be completed prior to the start of large scale disposition activities and that the source of plutonium for disposition will be materials stored in stabilized forms defined by DNFSB 94-1 recommendations.

Only the very top level of the DOE corrective action plan has been completed. Since the lower level corrective actions have not yet been planned or executed, it is not possible to specify the exact form, quantity, or location of the plutonium contained in the stabilized scrap or irradiated materials which may be available for disposition.

At the end of the cold war, there was also a considerable quantity of material which contained amounts of Pu which were considered to be economically recoverable but which were not considered primary manufacturing feed stocks. These residues, largely in storage at the Rocky Flats site, represent a concern since the fissile inventory is sufficiently large to be of proliferation concern. However, the Pu content for all residues, which is always less than 50%, is only on the order of about 1% on the average overall. The chemical and physical form of these residues presents difficulties for long term storage or use in the weapons complex. As a consequence, these have been declared as waste with the intent to discard them at WIPP when WIPP becomes operational.

Several of the immobilization options, however, may well present a more cost effective approach for the elimination of these residues rather than just placing them in WIPP. In order to consider this question, calculations were done at LLNL which estimated the cost associated with various options for either discarding the Pu directly, or for immobilizing the Pu for disposition as part of the Fissile Materials Disposition Program. These cost calculations considered the cost of packaging and disposing of the residues in WIPP versus the cost of processing the residues, dissolving them in glass, and adding the radiation spike prior to repository emplacement. In doing these calculations, parameters considered included the average Pu cutoff for processing (with the leaner residues always going to WIPP) and the average Pu loading per barrel which could be reasonably expected for WIPP disposition.

Figure 4, shown below, indicates the cost estimates for disposition for the nation's excess Pu using glass immobilization where various quantities of the residues are either included or excluded (based on the Pu cutoff percentage for processing).

As can be seen, the overall cost envelope is reduced substantially if at least some of the residues are processed for glass immobilization disposition. If higher Pu average loadings per barrel can be achieved for the WIPP discard, then the WIPP costs are reduced accordingly and the cost advantages are reduced. A family of total cost curves was calculated for average Pu barrel loadings up to 500 gm/barrel. Note that the current WIPP limit is 200 gms/barrel. An estimate of the advantageous Pu processing cutoff point was then calculated for each of these loading values and the curve shown in Figure 5 was plotted.

As can be seen, there may well be cost advantages in processing a significant portion of the residues for immobilization disposition given the WIPP limits of 200 gms/barrel. Average WIPP loading values will likely be less than 200 gms/barrel.

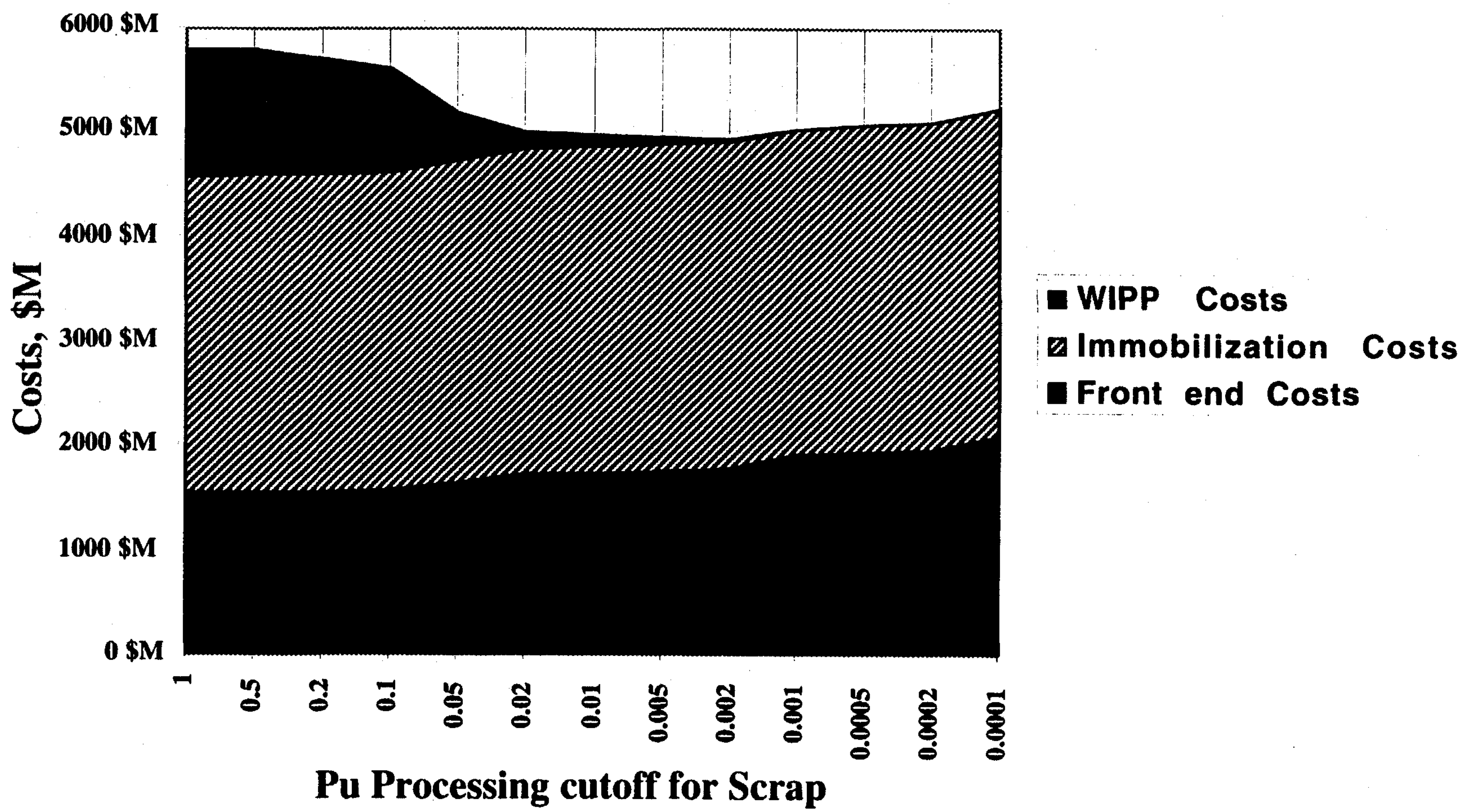
OVERALL R&D KEY FEATURES

Each of these technologies require further research and development to:

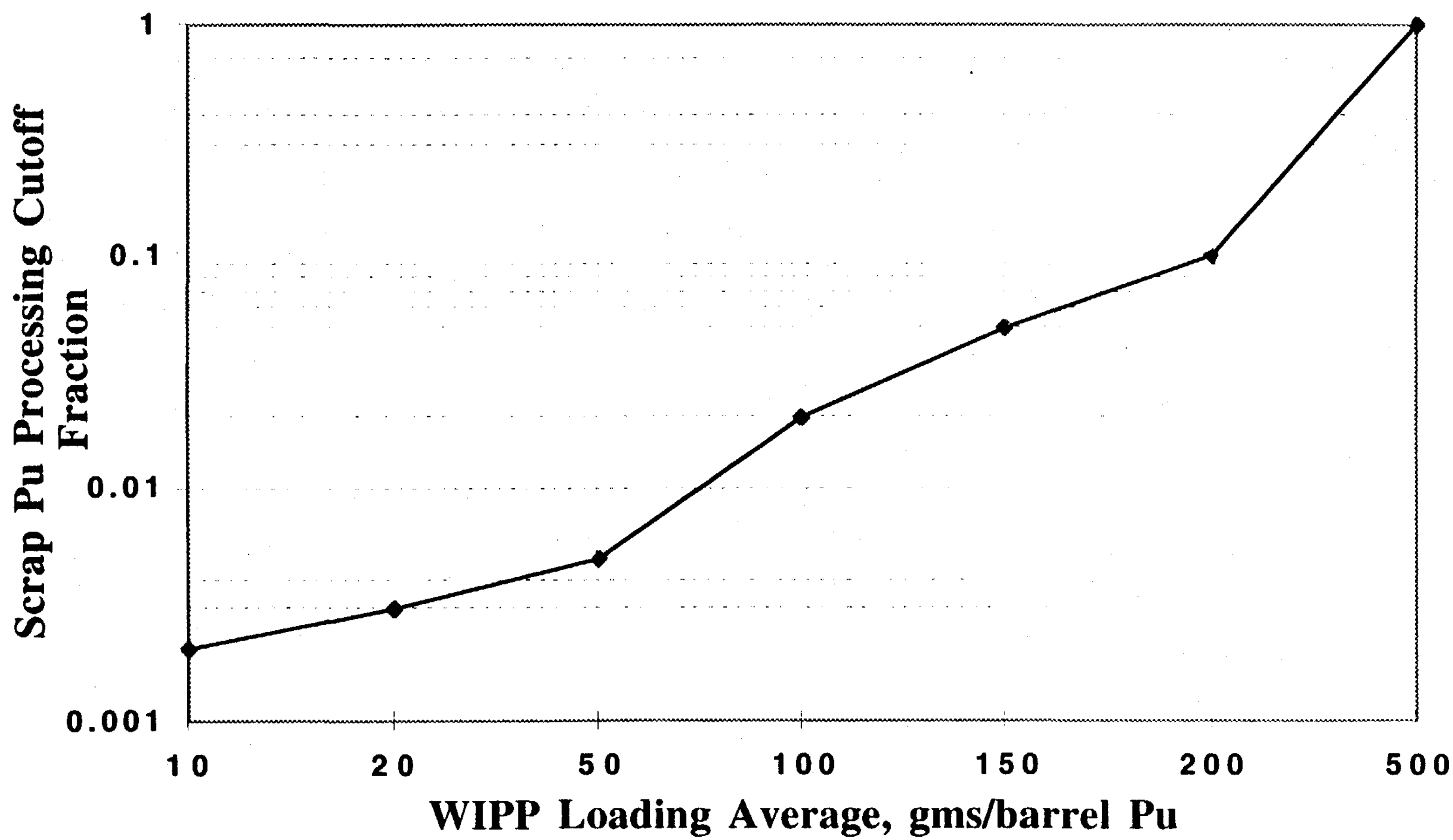
1. identify a material formulation that optimizes processability and long term performance;
2. develop processing equipment, material flow and process controls, operational strategies, and material accountability while minimizing impacts on workers, the environment, cost, and the ability to maintain an acceptable implementatyion schedule;
3. demonstrate on a pilot scale that individual operations or processing steps fit together seamlessly;

demonstrate that the specific disposal forms meet the spent fuel standard for proliferation resistance.

Immobilization Cost Contributions vs Pu Processing Cutoff for 38 gms/barrel



"Optimum" Scrap Processing Cutoff Points vs Average WIPP Loading Limits



The fundamental features of the overall long term research and development (R&D) plan for plutonium disposition using immobilization technologies include

- Full understanding of criticality safety margins at every stage of plutonium handling and processing
- Practical limits of plutonium concentration from both solubility and kinetic considerations
- Incorporation of ^{137}Cs and its effects on both process operations and final waste form performance and proliferation resistance
- Sensitivity of immobilization process formulation and product performance to impurity concentrations in the feed.
- Process optimization to minimize waste, costs, and time of disposition
- Pilot scale demonstrations with transuranics to confirm viability of the process
- Evolving and characterizing equipment designs and compositions that reliably and safely handle plutonium weight loadings that result in economically effective operations
- Properties that influence performance, reliability, and safety considerations—such as nuclear criticality—must be determined
- Assessment of the impact of plutonium volatility
- Assessment of the impact of the presence of neutron absorbers
- Assessments of the physical durability of the product and the plutonium product phases. Relative durability and leach rate determination
- Assessments of Pu recovery and proliferation resistance of the immobilized plutonium form
- Development of predictive material control and accountability and process controls and models for plutonium immobilization operations.
- Analytical tools and techniques will need to be properly validated

These issues have a large effect on process complexity and limitations on throughput, so it is imperative that a consistent set of baseline data be carefully and fully determined. The experimental work and other assessments identified in the R&D plan are intended to address these issues.

SUMMARY

An International Team was assembled for the purpose of selecting suitable immobilization forms and processing technologies for the Fissile Materials Disposition Program Office. The Task Team use the NAS Study as a reference point for starting the study but was not limited to recommendations of the NAS.

Three basic forms have been selected and the processing options to provide those three forms have been defined. As this conference is for the discussion of glass and ceramics only, metal forms were not discussed. Environmental Data has been supplied to the DOE contractor writing the PEIS for the Disposition Program. The Task Team is now developing cost data for the Record of Decision which is anticipated in the late Summer of 1996. The Task Team is also evaluating these options to determine if they indeed meet the security standard and goals set up by the NAS Study.

APPENDIX
LONG RANGE RESEARCH AND DEVELOPMENT PLAN

The Long Range Research and Development Plan, Immobilization Technologies, summarizes the long term research and development (R&D) requirements for plutonium disposition using immobilization technologies.

In this overview, a brief summary of the status of the immobilization options and the emphasis of the required research and development is presented for each of the immobilization options being considered. The fundamental features of the overall long term research and development (R&D) plan for plutonium disposition using immobilization technologies are presented.

Vitrification Technology

The plan consists of individual standalone plans for each immobilization options. Three alternatives based on vitrification technology are under consideration.

1. Vitrification at a Greenfield site (Internal Radiation Barrier)
2. Vitrification using an External radiation barrier (Can-in-Canister)
3. Vitrification using a DWPF Adjunct melter (Internal Radiation Barrier)

The logical relationship of key milestones for the vitrification alternatives are shown in diagram form in Figure 6.

A critical concern relates to whether a glass for each of the alternatives can be formulated to meet the processability and performance requirements.

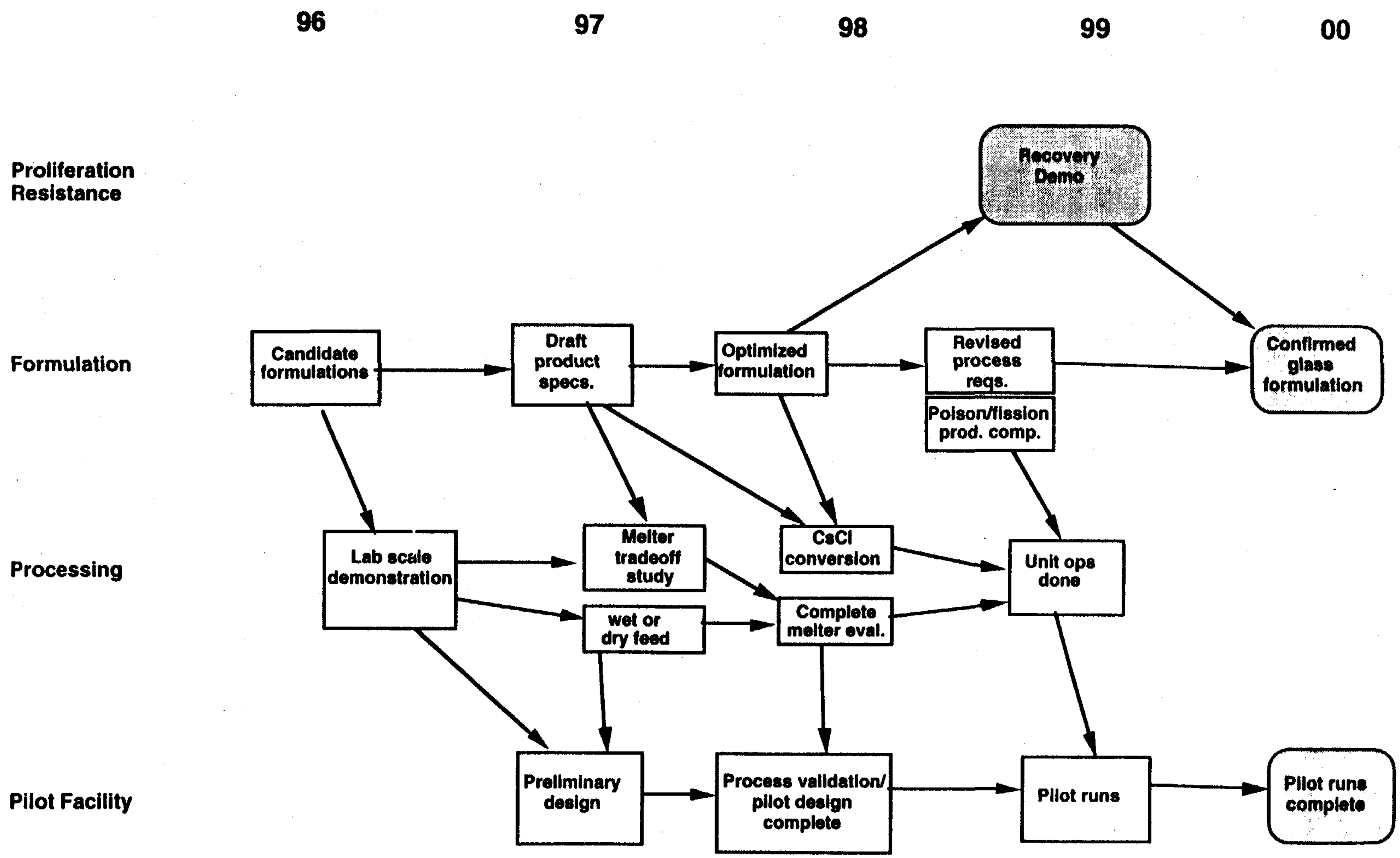
Development and Characterization of Optimized Glass Formulations for Plutonium Immobilization

The specific glass formulation selected will strongly influence the design and cost of the immobilization facility, as well as the extent of characterization necessary for facility and wasteform licensing, with a direct impact on implementation schedule. For example, the maximum allowable plutonium loading, which will be set by the limits of process safety and long term performance of the wasteform, may determine facility size and throughput.

Project Objective: To formulate, through testing and analysis, a Pu-containing glass optimized for safety, performance, processability, and cost effectiveness. To achieve adequate understanding of the glass behavior, both during processing and at long times, so that Title 1 design can be initiated.

Project Description: Detailed glass requirements for both processing anlong term performance will be documented. Guided by existing data and analysis, a range of glass formulations will be prepared and characterized for Pu solubility, the influence of required additives such as neutron absorbers, tolerance for process and compositional variations, processability, resistance to radiation damage, and long-term chemical durability. Models for Pu processing parameters and release rates will be developed and used to optimize the formulations. These formulations will be characterized, material response models modified as necessary, and long term performance testing initiated. This effort will be closely coupled with the Processing and Equipment Technologies effort to insure that candidate formulations meet processability and process safety requirements. Completion of this task is not required prior to beginning Title 1 design.

Glass Immobilization - Key Milestones



Key Information Generated/Key Issues Resolved:

- Pu solubility as a function of glass composition
- neutron absorber solubility as a function of glass composition
- physical and chemical requirements for feedstocks
- effect of radiation damage on stability of wasteform
- composition of final wasteform
- information needed for long-term degradation/radionuclide release models
- relative release rates of fissile materials and neutron absorbers

Process Development, Controls, and Equipment Technologies

Immobilization with large weight fractions Pu has never been implemented in a production setting. Optimization of the facility for safety, performance, ES&H and cost effectiveness will require background development for feed stock preparation, process flow, melter design, material accountability, and process and criticality control. These efforts will work in close concert with the Glass Formulation R&D tasks.

Project Objective: To understand facility unit processes and production flow well enough to optimize facility and equipment design, performance, and cost effectiveness (while maintaining adequate safety and performance margins) during Title 1 and 2 design.

Project Description: Glove box scale operations will be set up, evaluated and modeled for each unit operation. Process schemes will be developed, analyzed and, tested comparing wet and dry melter feed streams. For the greenfield alternative, the process for converting $^{137}CsCl$ from Hanford to a form suitable for a glass feed will be demonstrated and off-gas handling approaches developed. In combination with glass formulation efforts and nuclear criticality analysis, glass melter designs will be evaluated and modifications proposed. Prototype melter performance will be documented. A Process/Product Control System will be developed consistent with the glass processability models determined in the formulation effort. The technology for tracking accountable materials will be demonstrated.

Key Information Generated/Key Issues Resolved:

- determination of whether to use a wet or dry feed for melter
- demonstration of the process for converting CsCl and other potentially incompatible feed materials to suitable form
- establishment of processing requirements on glass formulation
- development of process criticality control, process quality, and materials accountability systems
- processing equipment and melter design

Pilot Facility for Plutonium Vitrification

Prior to completing detailed facility design, it is essential to demonstrate that full scale (glove box) processes and control strategies work together seamlessly and predictably. For these operations surrogates will be used in place of radiation barrier (e.g., ^{137}Cs) materials.

Project Objective: Demonstrate processes for making Pu-containing glass logs using realistic equipment and procedures.

Project Description: Using facilities, melter and procedures developed in other tasks, demonstrate full scale operations, initially using surrogates and later using Pu-containing glass but no radiological barrier. Demonstrate process control and monitoring systems. Develop operational experience, characterize product and compare to model, and modify unit operations and Process Control System as appropriate.

Key Information Generated/Key Issues Resolved:

- demonstration of compatibility of unit processes
- demonstration of lack of Pu or neutron absorber segregation in product glasses

Determination of Proliferation Resistance of Plutonium Glasses

Prior to implementation, it is important to demonstrate that Pu glasses meet the spent fuel standard.

Project Objective: To document the proliferation resistance of candidate Pu glasses.

Project Description: Process flow sheets for dissolution and recovery of Pu from vitrified wasteforms will be determined. Time and cost estimates will be made. Limited validation testing will be performed.

CERAMICS TECHNOLOGY

Two different immobilization alternatives based on ceramics technology are under consideration:

1. Ceramics Greenfield site (Internal Radiation Barrier)
2. Ceramics External radiation barrier (Can-in-Canister)

The logical relationships of key milestones for the ceramic alternatives are shown in diagrammatic form in Figure 7.

A critical concerns relate to whether a ceramic can be formulated that meets the processability and performance requirements

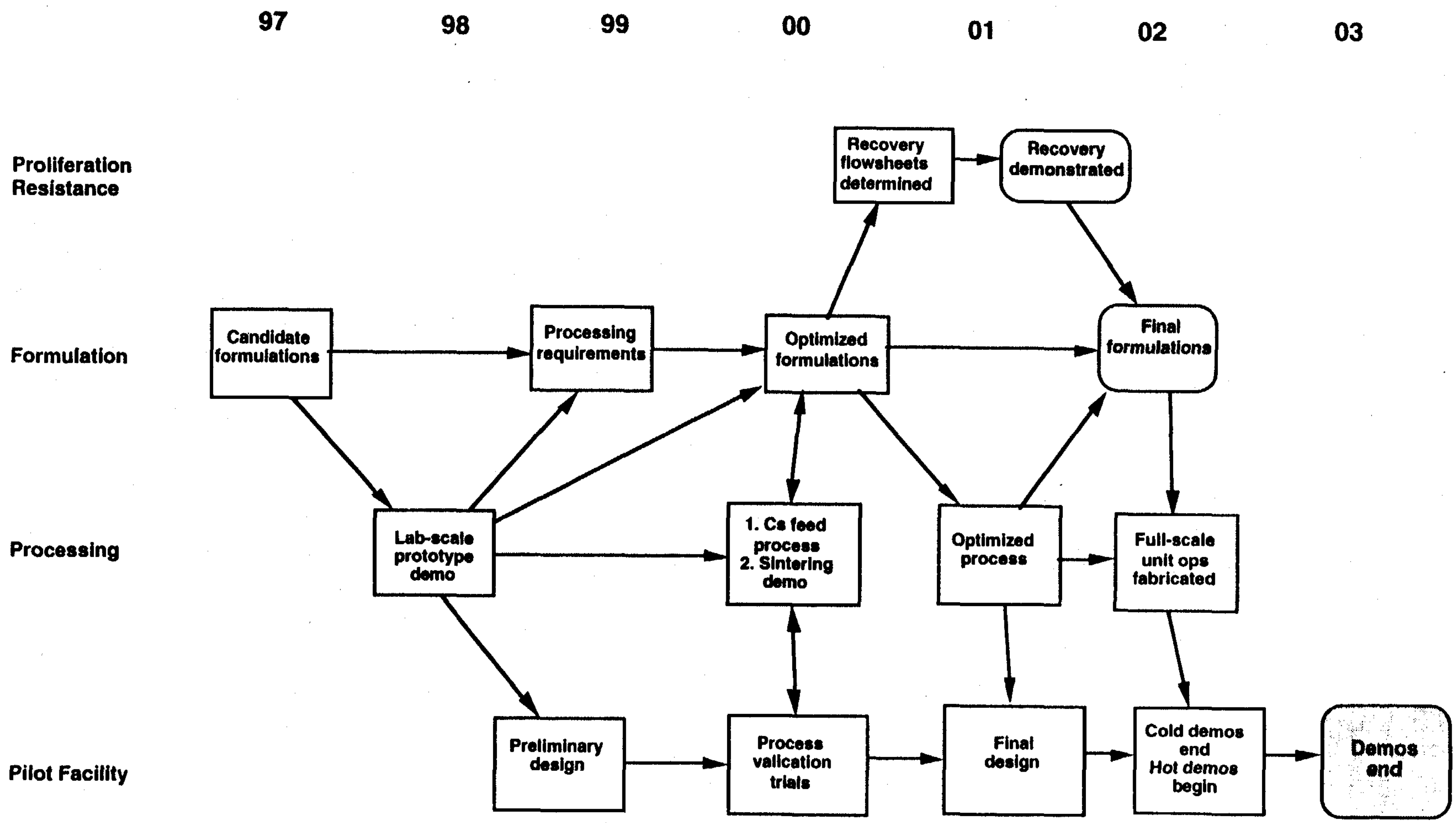
Development and Characterization of Optimized Ceramic Formulations for Plutonium Immobilization

The specific ceramic formulation selected will strongly influence the design and cost of the immobilization facility, as well as the extent of characterization necessary for facility and wasteform licensing, with a direct impact on implementation schedule. For example, the maximum allowable Pu loading, which will be set by the limits of process safety and long term performance, may determine facility size and throughput.

Project Objective: To formulate, through testing and analysis, a Pu-containing ceramic optimized for safety, performance, processability, and cost effectiveness. To achieve adequate understanding of the ceramic's behavior, both during processing and at long times, so that Title 1 design can be initiated.

Project Description: Detailed ceramic requirements for both processing and long term performance will bemented with all relevant agencies and departments. Guided by existing data and analysis, a range of ceramic formulations will be prepared and characterized for Pu solubility, the influence of required additives such as neutron absorbers, tolerance for process and compositional variations, processability, resistance to radiation damage, and long-term chemical durability. Models for Pu processing parameters and release rates will be developed and used to optimize the formulations. These formulations will be characterized, material response models modified as necessary, and long-term performance testing initiated. This effort will be closely coupled with the Processing and Equipment Technologies effort to insure that candidate formulations meet processability and process safety requirements. Completion of this task is not

Ceramic Immobilization - Key Milestones



required prior to beginning Title 1 design.

Key Information Generated/Key Issues Resolved:

- Pu solubility as a function of ceramic composition
- neutron poison solubility as a function of ceramic composition
- physical and chemical requirements for feedstocks
- effect of radiation damage on stability of wasteform
- composition of final wasteform
- information needed for long-term degradation/radionuclide release models
- relative release rates of fissile materials and neutron poisons

Process Development, Controls, and Equipment Technologies

Immobilization using ceramics with large weight fractions of Pu has never been implemented in a production setting. Optimization of the facility for safety, performance, ES&H, and cost effectiveness will require background development for feed stock preparation, process flow, calciner and press design, material accountability, and process and criticality control. These efforts will work in close concert with the Ceramic Formulation R&D task.

Project Objective: To understand facility unit processes and production flow well enough to optimize facility and equipment design, performance, and cost effectiveness (while maintaining adequate safety and performance margins) during Title 1 and 2 design.

Project Description: Glove box scale operations will be set up, evaluated and modeled for each unit operation. Process schemes will be developed, analyzed and, possibly, tested comparing wet and dry melter feed streams. The effect of impurities in feed materials on ceramic processability will be evaluated, and if needed, preprocessing procedures established. For the greenfield alternative, the process for converting $^{137}CsCl$ from Hanford to an acceptable feed for ceramics will be demonstrated and off-gas handling approaches developed. In combination with ceramic formulation efforts and nuclear criticality analysis, calciner and press designs will be evaluated and modifications proposed. A Process/Product Control System will be developed consistent with the ceramic processability models determined in the formulation effort. The technology for tracking accountable materials will be demonstrated.

Key Information Generated/Key Issues Resolved:

- determination of whether to use a wet or dry Pu feed
- demonstration of process for converting $^{137}CsCl$ and other potentially incompatible feed materials to suitable form
- establishment of processing requirements on ceramic formulationdevelopment of process criticality control, process quality, and materials accountability systems
- processing equipment (e.g., hot press, calciner, slurry tank) design

Pilot Facility for Immobilization of Plutonium Using Ceramics

Prior to completing detailed facility design, it is essential to demonstrate that the full scale (glove box) processes and control strategies work together seamlessly and predictably. For these operations will culminate in the pilot-scale production of Pu-containing ceramics with surrogates used in place of the radiation barrier materials.

Project Objective: Demonstrate the integration of the slurrying, calcining, and hot-pressing operations needed to make

Pu-containing ceramic wasteforms using realistic equipment and procedures.
Project Description: Using facilities and procedures developed in other task demonstrate full scale operations, initially using all surrogates and later using Pu and non-radioactive surrogates for the radiation-barrier. Demonstrate process control and monitoring systems. Develop operational experience, characterize product, compare to models, and modify unit operations and Process Control System as appropriate.

Key Information Generated/Key Issues Resolved:

- demonstration of compatibility of unit processes
- demonstration of homogeneity of materials in the slurrying and calcination processes

Proliferation Resistance of Plutonium Ceramics

Prior to implementation, it is important to demonstrate that Pu ceramics meet the spent fuel standard.
Project Objective: To document the proliferation resistance of candidate Pu ceramics.
Project Description: Process flow sheets for recovery of Pu from ceramic wasteforms will be determined. Time and cost estimates will be made. Limited validation testing will be performed.

ACKNOWLEDGMENT

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