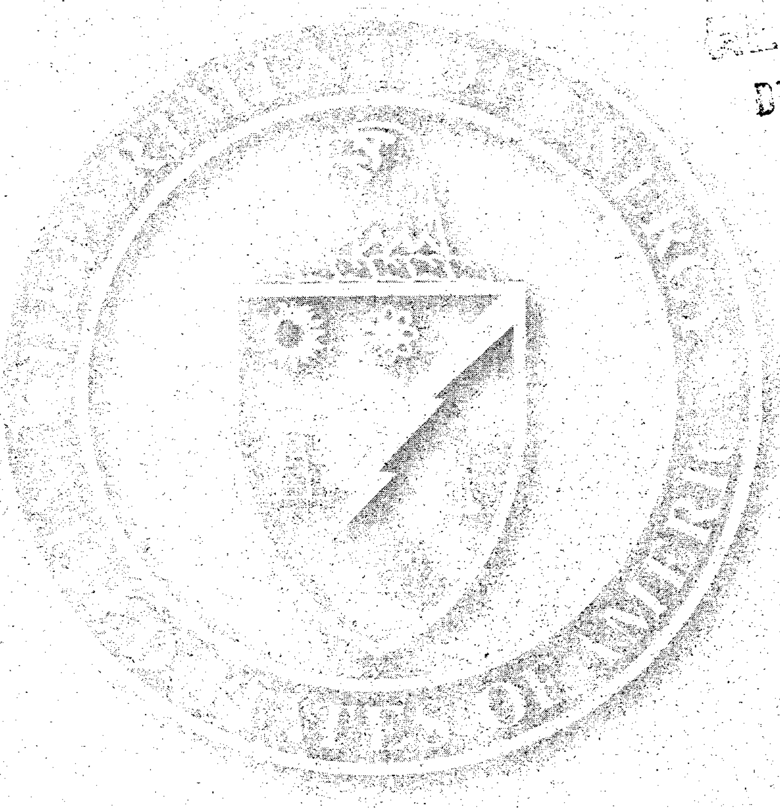
U.S. Department of Energy
Idaho Operations Office
Plutonium Focus Area

DOE/ID-10561

Research and Development Plan



November 1996

DISCLAIMER

DOE/ID-10561

Plutonium Focus Area Research and Development Plan

(Revision 1)

Plutonium Focus Area
Technical Advisory Panel

November 1996

U.S. Department of Energy
Idaho Operations Office
Idaho Falls, ID 83415

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LIST OF ILLUSTRATIONS

1. EXECUTIVE SUMMARY

On May 26, 1994, the Defense Nuclear Facilities Safety Board (DNFSB) issued Recommendation 94-1, expressing the Board's concern about the safety of Department of Energy (DOE) nuclear materials. The DNFSB recommended that DOE stabilize and repackage high-risk material within 2-3 years and the remaining materials within 8 years. The DOE accepted DNFSB Recommendation 94-1 on August 3, 1994. After establishing the Nuclear Materials Stabilization Task Group (NMSTG), DOE issued an implementation plan to address DNFSB concerns ("Defense Nuclear Facilities Safety Board Recommendation 94-1 Implementation Plan," February 28, 1995).

The DNFSB called for the establishment of a research program to fill gaps in the technology base needed to accomplish stabilization. DOE committed to a research and development (R&D) program to support the technology needs for converting and stabilizing its nuclear materials for safe storage. An R&D program plan was prepared by the NMSTG-Chartered Research Committee and published in November 1995. With that publication, the Research Committee was disbanded. To ensure that technology needs for stabilization continue to be addressed and that the R&D Plan is appropriately updated, the Plutonium Focus Area (PFA) was established by DOE in October 1995 under the DOE Idaho Operations Office, with support from Lockheed Martin Idaho Technologies Company (LMITCO) and Argonne National Laboratory (ANL). (See Appendix B for PFA Charter.) The PFA tasked its Technical Advisory Panel (TAP) to update and revise the November 1995 R&D Plan.

The R&D Plan addresses five of the six material categories from the 94-1 Implementation Plan: plutonium (Pu) solutions, plutonium metals and oxides, plutonium residues, highly enriched uranium, and special isotopes. R&D efforts related to spent nuclear fuel (SNF) stabilization were specifically excluded from this plan, and are being coordinated through the Technology Integration Technical Working Group, established by the Office of Spent Fuel Management in June 1993.

This updated plan has narrowed the focus to more effectively target specific problem areas by incorporating results from trade studies. Specifically, the trade studies involved salt; ash; sand, slag, and crucible (SS&C); combustibles; and scrub alloy. The focus was further narrowed by reflecting technology development progress from the Stabilization R&D Program at Los Alamos National Laboratory (LANL). Additionally, new approaches were considered as a result of white papers submitted from the at-large community, and initial interactions between DOE and the Russians have resulted in several proposed joint R&D projects (see Chapter 10).

The plan anticipates possible disposition paths for nuclear materials and identifies resulting research requirements. These requirements may change as disposition paths become more certain. Thus, this plan represents a snapshot of the current progress and will continue to be updated on a regular basis.

The TAP concludes that, in general, the technology needs for stabilizing 94-1 nuclear materials are being addressed by existing DOE programs. The principal programmatic risk identified this year by the TAP is that the technology need dates may not be met in all cases. This year, the

TAP has also identified several research needs that address the path forward to materials stewardship and disposition.

1.1 Status of November 1995 R&D Plan Recommendations

The November 1995 R&D Plan contained 18 recommendations based on technology gaps, technologies that needed to be tracked based on maturity scores, and other issues identified separately by the R&D Committee. These recommendations and their current status are presented in Appendix E. Of the 18 recommendations, 6 have been closed and the other 12 are effectively underway toward closure to meet 94-1 Implementation Plan commitments.

1.2 Programmatic Risk Summary for 94-1 Implementation Plan Commitments

Using the same system engineering approach employed in last year's R&D Plan, the TAP developed programmatic risk assessments based on the technical maturity score and R&D need date of each technology considered. The left frame of Figure 1-1 shows all technologies reviewed by the TAP for this year's R&D plan. The number of technologies reviewed for this plan was 64 compared with 99 for the November 1995 plan. There are 35 medium and high risk technologies among all of the technologies reviewed for this plan compared with 16 medium risk technologies and no high risk technologies identified in the November 1995 R&D Plan. This is because the calendar has advanced one year, but the progress in research and development has not necessarily advanced one year. In general, progress has been made in the research areas of requirements understanding and process and equipment development. Where progress has not yet occurred is in the development of facility, safety, and personnel readiness.

The right frame of Figure 1-1 shows the 38 technologies that are recommended to be pursued as baseline or competitive alternative technologies. Within this context, baseline refers to a technology recommended in this plan for a particular use at a specific site; a competitive alternative is one of two or more technologies that are recommended as candidates for a baseline application. The principal mechanism for these recommendations are the results of the five trade studies. Based on these recommendations, there is only one high risk technology, and there are 12 medium risk technologies that cause some programmatic risk with respect to meeting 94-1 Implementation Plan Milestone dates.

The high risk rating is for the baseline cryogenic crushing of combustibles containing plutonium at Rocky Flats Environmental Technology Site (RFETS). It requires focused commitment by DOE headquarters and the Rocky Flats Field Office to develop this process that was recommended by the trade study. In addition to process development; facility, safety, and personnel readiness are essential for this technology and require a committed effort to meet the November 1998 stabilization date in the 94-1 Implementation Plan.

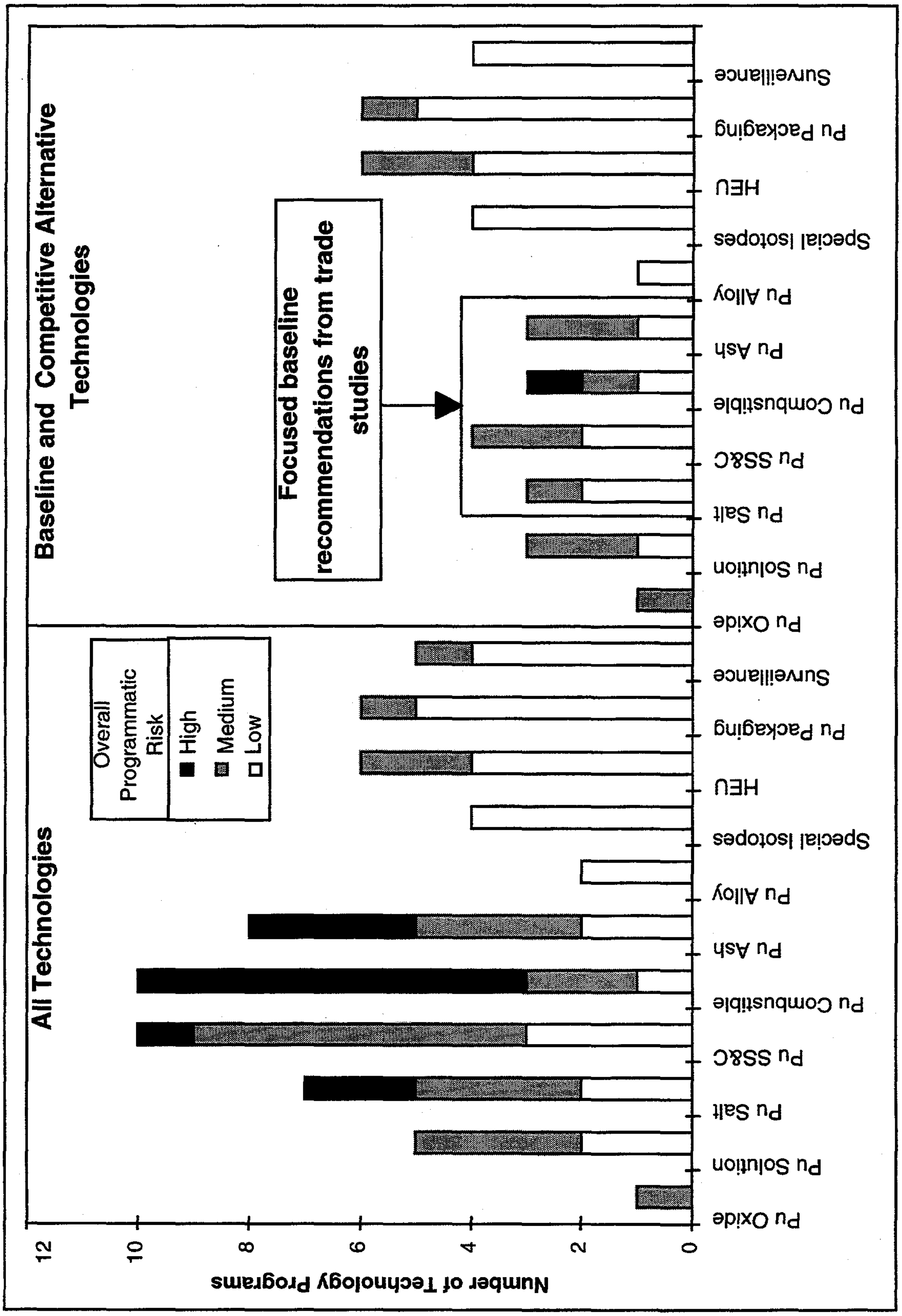

Figure 1-1 Summary Results of Technical Maturity Evaluation

Several baseline and competitive alternative technologies are medium risk and require close tracking by NMSTG. They are the Plutonium Stabilization and Packaging System (PuSPS) stabilization process for plutonium metals and oxides at RFETS; the vertical calciner for plutonium solutions at Hanford; treatment of plutonium solutions at RFETS; distillation for salts at RFETS; cementation for sand, slag, and crucible (SS&C) at Hanford; calcining SS&C for shipment at RFETS; Mediated Electrochemical Leaching of plutonium combustibles at RFETS; Silver Persulfate Dissolution for ash at Hanford; Microwave Vitrification for disposal of RFETS ash at the Waste Isolation Pilot Plant (WIPP); electrochemical treatment and Glass Material Oxidation and Dissolution System (GMODS) immobilization for Molten Salt Reactor Experiment (MSRE) salt at Oak Ridge National Laboratory (ORNL); and the pipe component for shipping salt residue to WIPP from RFETS.

Two gaps were identified for 94-1 stabilization:

1. The need for a comprehensive assessment of the current legacy and anticipated future residue inventories to determine if there are miscellaneous inventories that cannot be treated by baseline and competitive alternative technologies or by straight forward repackaging and disposal. If any residues are found not to have a treatment approach identified, there may be a need to develop new stabilization approaches.

2. The need for a trade study to confirm problems with the RFETS Site Integrated Stabilization Management Plan (SISMP) baseline approach to stabilize pyrochemical crucible residues and evaluate the viability of alternative approaches. R&D activities should be directed to provide data and support analyses in the trade study.

1.3 Technology Gaps Related to Long-term Stewardship and Materials Disposition Interfaces

In comparing R&D activities to the comprehensive set of technical requirements, the TAP identified a small number of new areas of R&D requirements, or "gaps" in the current R&D efforts. These gaps are identified as recommendations in Table 1-1, and are associated with the interface to the Materials Disposition Program and the anticipated Material Stewardship mission that will be pursued as a follow-on to the NMSTG. These provide the path forward for planning core technology in the Stabilization R&D program and PFA-sponsored advanced technology development.

Table 1-1. Recommendations to Address Technology Gaps in Materials Disposition Interface and Stewardship

Recommendations	Comments
Assessment of adequacy of disposal configurations against safe storage criteria to determine if additional measures are required for the safe interim storage of materials awaiting transportation and disposal.	WAC have been developed to address transportation and repository performance issues, but not necessarily interim storage issues.
DOE-EM management of nuclear materials will use decisions derived from the DOE-MD NEPA process. If the stabilization program pursues can-in-canister approaches (currently considered backup technologies with high schedule risk to meet 94-1 commitments), treatment of materials should be closely coordinated with DOE-MD and DOE-RW programs.	The DOE-RW program is evaluating the suitability of the high level waste repository based on reference waste forms. Any new waste forms need to be assessed to ensure that their repository performance characteristics are bounded by the reference waste forms.
Assess which baseline stabilization approaches will not meet safeguards and security requirements.	Treatment methods must be defined or developed to convert materials with plutonium into TRUW forms that allow reduced safeguards and security.
Conduct a trade study to determine the necessity and validity of using automation for surveillance and monitoring, packaging, and handling of storage containers.	The need for automation currently is not integrated into the R&D requirements.

2. INTRODUCTION

2.1 Background

On May 26, 1994, the Defense Nuclear Facilities Safety Board (DNFSB) issued Recommendation 94-1. This recommendation expresses the Board's concern about nuclear materials left in the manufacturing "pipeline" after the United States halted its nuclear weapons production activities and emphasizes the need for remediation of these materials. The Department of Energy (DOE) accepted DNFSB Recommendation 94-1 on August 3, 1994. After establishing the Nuclear Materials Stabilization Task Group (NMSTG), DOE issued an implementation plan to address DNFSB concerns ("Defense Nuclear Facilities Safety Board Recommendation 94-1 Implementation Plan," February 28, 1995).

Recommendation 94-1, Sub-recommendation (2), states that *"...a research program [should] be established to fill any gaps in the information base needed for choosing among the alternate processes to be used in safe interim conversion of various types of fissile materials to optimal forms for safe interim storage and the longer term disposition. Development of this research program should be addressed in the program plan called for by [the Board]."*

Consequently, the NMSTG chartered the NMSTG Research Committee in March 1995 to accomplish the following:

1. Assess the nuclear materials stabilization program outlined in the implementation plan

2. Formulate a research and development (R&D) plan to address the technology and core program needs of the stabilization program

3. Prepare task statements defining R&D activities required to accomplish program objectives.

The committee completed its activities with the issuance of the *94-1 Research and Development Plan* in November 1995 and was disbanded.

To ensure that the technology needs for stabilization continue to be addressed and that the R&D Plan is appropriately updated, the Plutonium Focus Area (PFA) was established by DOE in October 1995 under the DOE Idaho Operations Office, with support from Lockheed Martin Idaho Technologies Company (LMITCO) and Argonne National Laboratory (ANL). (See Appendix B for PFA Charter). As part of its responsibility, the PFA tasked its Technical Advisory Panel (TAP) to update and revise the current R&D Plan.

The TAP reviewed the Implementation Plan and Site Integrated Stabilization Management Plans (SISMP), and visited the Savannah River Site (SRS), the Hanford Site (Hanford), Rocky Flats Environmental Technology Site (RFETS), and Los Alamos National Laboratory (LANL) as part of its review activities. Regular meetings of the TAP, including technical advisors, were also held in conjunction with site visits. Additionally, the TAP reviewed the results of various trade studies directed by NMSTG, as well as white papers submitted from the at-large community.

This plan addresses five of the six material categories discussed in the 94-1 Implementation Plan, namely: plutonium (Pu) solutions, plutonium metals and oxides, plutonium residues, highly enriched uranium, and special isotopes. R&D efforts related to spent nuclear fuel (SNF) stabilization were specifically excluded from this plan. These efforts are being coordinated through the Technology Integration Technical Working Group, established by the Office of Spent Fuel Management in June 1993.

The plan anticipates possible disposition paths for nuclear materials and identifies resulting research requirements. These requirements may change as disposition paths become more certain. Thus, this plan represents a snapshot of the current progress and will continue to be updated on a regular basis.

2.2 R&D Plan Description

The plan format has been modified to focus on each material category. Each material category includes a brief introduction; a brief description of technologies under consideration; a description of any newly proposed R&D; an evaluation of current and proposed R&D against requirements; conclusions and recommendations, and a table showing the maturity scores and status of technologies under consideration.

Revision 0 of the Plan (November 1995) cataloged R&D needs for addressing nuclear materials stabilization issues. Revision 1 (November 1996) narrows the focus of those needs to more effectively target specific problem areas. At the same time, this revision incorporates results from various trade studies and R&D needs identified in various white papers to further focus on stabilization issues. The trade studies targeted salt; ash; SS&C; combustibles; and scrub alloy. Lead Laboratory research results from LANL were also incorporated into the plan to further focus on required R&D needs and technologies. New approaches were considered as a result of submitting white papers from the at-large community. Finally, the NMSTG provided specific direction that the end-state for material stabilization acceptable under the 94-1 program be limited to either transuranic (TRU) waste that meets Waste Isolation Pilot Plant (WIPP) waste acceptance criteria (WAC), or plutonium metal and oxide that meets the 50-year storage standard (DOE-STD-3013). WAC and the storage standard were used to provide a consistent basis for the trade studies.

Selection of Recommendation 94-1 stabilization end-states is also part of ongoing interface efforts between NMSTG and the DOE Office of Fissile Materials Disposition (DOE-MD). Selected end-states are compatible with material input requirements under consideration by DOE-MD. Continued interaction between NMSTG and DOE-MD is facilitated by an interface working group, which was formed in 1996 to pursue common requirements and technologies.

Materials stabilization and other related research activities discussed in this report have been categorized into the following areas, driven by requirements to stabilize and store materials: standards, waste forms, safeguards termination, plutonium oxides, plutonium solutions, plutonium residues, special isotopes, highly enriched uranium, packaging, surveillance and monitoring, engineered systems, and core technology. Each category was linked to appropriate 94-1 Implementation Plan milestones that drive schedule needs for R&D. The schedule need for a specific category of R&D at a particular site was determined by evaluating the programs defined in the SISMP provided by each site to the NMSTG as of August 1996. Subsequent changes in the 94-1 Implementation Plan and the SISMPs may impact the R&D need dates described in this plan and will be reflected in future updates.

3. STORAGE STANDARDS, WASTE FORMS, SAFEGUARDS AND SECURITY

3.1 Standards

3.1.1 Introduction

DOE-STD-3013 stipulates requirements for long-term storage of plutonium metals and oxides in concentrations greater than 50 wt% plutonium. A storage period of up to 50 years is permitted for sealed metal containers.

Plutonium metals must be stabilized in a nonpyrophoric physical form, must be free of loose oxide, and must be stored in sealed metal containers that are free of plastic or other organic materials. Stabilized plutonium oxides must be less than 0.5% Loss on Ignition (LOI) of water to avoid over pressurization of storage containers due to radiolysis of water or vaporization of water under accident conditions (e.g., fire).

Continued R&D is required to ensure adequacy of the standards for 50-year storage of pure metals and oxides. Additional data are required to provide confidence in the adequacy of the standard for long-term storage of impure materials (i.e., between 50 and 85 wt% plutonium). Initial stabilization work at Hanford has identified issue with LOI techniques for impure plutonium: chemical changes in impurities contribute to mass changes in the LOI process. R&D is required to understand the phenomenon and develop methods for validating successful stabilization of impure plutonium.

Interim Safe Storage Criteria for storing residues have been prepared to supplement the 94-1 Implementation Plan. There is no identified research need for support of the Interim Safe Storage Criteria. The NMSTG expects that residues covered under the 94-1 Implementation Plan will be prepared for disposition at WIPP and/or the plutonium metal or oxide will be separated from the residue and stored in accordance with DOE-STD-3013.

3.1.2 Description and Status for Storage Standards Technologies

3.1.2.1 Plutonium Metal & Oxides (>50%) (LANL). Experimental work was performed to determine the relationships between LOI, particle size distribution, surface area and adsorption kinetics versus calcination temperature for pure plutonium oxides. This work provided basic information in support of DOE-STD-3013 on the behavior of plutonium oxides during calcination, packaging, and storage.

The objective of the ongoing work is to characterize and understand the behavior of impure plutonium oxides. Plutonium oxide between 50 and 85 wt% plutonium will be studied to verify that DOE-STD-3013 is appropriate for impure plutonium. The experiments progressively decrease the percentage of plutonium content down to 50 wt% by adding impurities expected to be present in stabilized materials. This information will be used to:

1. Define the stabilization process parameters and LOI testing applicability for impure plutonium

2. Predict potential pressure buildup for pure and impure oxide items in long-term storage (~50 years)

3. Determine the mass fraction of respirable and dispersible particles as a function of processing history and thermal treatment.

Different categories of plutonium oxide are being selected for study; selection criteria are based on process history, as well as plutonium content and impurities.

3.1.2.2 Plutonium Residues (LANL). The 94-1 R&D Plan (Rev. 0), published November 1995, included requirements to develop a technology base for the Interim Safe Storage Criteria. Program guidance was established calling for interim criteria to be used until the year 2002, at which point materials would be packaged to meet either long-term storage or waste disposal criteria. This guidance, coupled with evaluations of diverse materials planned to be placed in interim storage conditions, concluded that developing such a technology base was impractical. Therefore, the safety of materials in interim storage must be ensured through a comprehensive surveillance program, as called for in the Interim Safe Storage Criteria documentation.

3.1.2.3 Shelf Life Program (LANL). Shelf-life efforts are being conducted to fill gaps in the knowledge regarding the behavior of plutonium-bearing materials during storage. The project has studied the extent of gas production and gas recombination for pure plutonium oxide under ambient conditions. Work will continue with kinetics tests and long-term storage tests conducted on pure oxide with known amounts of water and at various temperatures. Similar tests will be conducted on impure oxides. Corrosion effects of material/containment interactions will be analyzed.

The project is evaluating pure and impure metal and oxide materials that exist at RFETS, SRS, Hanford, and LANL. Materials are placed in test containers for PVT measurements. The shelf-life activities are starting with LANL inventory items that are representative of stabilized materials that are anticipated to be placed in 3013 containers. The project will add samples of actual material inventories from RFETS, Hanford, and SRS, including materials that result from residue processing.

Containers have been designed for pressure-volume-temperature measurements using pressure transducers, thermocouplers, and gas sampling ports for mass spectroscopy. Additional R&D on measurement techniques may include Raman spectroscopy, acoustic resonance spectroscopy, and non-invasive, pressure-sensitive devices to enhance shelf-life surveillance capabilities.

3.1.3 Newly-proposed R&D Activities/Technologies

There are no newly-proposed R&D activities or technologies at this time.

3.1.4 Evaluation of Current and Proposed R&D Against Requirements

The storage standard R&D activities described in Section 3.1.2 will provide a comprehensive basis for meeting the requirements of DOE-STD-3013.

Additional R&D may be needed to evaluate the suitability for safe interim storage of disposal forms that meet WAC. WAC have been developed primarily to address transportation and repository performance issues, but not necessarily interim storage issues. An assessment of the adequacy of disposal configurations against safe storage criteria is needed to determine if additional measures are required for safe storage of materials pending transportation and disposal.

3.1.5 Conclusions and Recommendations

Conclusions:

The LANL program supporting plutonium metal and oxide packaging addresses the needs of DOE-STD-3013.

Recommendations:

Surveillance activities should be continued and any resulting R&D needs should be addressed for inventories in interim storage.

3.2 Waste Forms

3.2.1 Introduction

The general strategy for managing plutonium scrap and residue is to (1) stabilize the materials for long-term storage or (2) convert them into an acceptable form for disposal. Secondary wastes from processing and/or any residues treated as waste must meet the WAC of the final disposal sites.

3.2.1.1 High Level Waste Repository Certification. DOE-MD is evaluating disposition options for excess plutonium as part of international arms control agreements. Options being evaluated include using plutonium as a nuclear power reactor fuel, disposal with high-level waste (HLW), and deep borehole disposal. In each case, the final product would be disposed of in a geological repository and would meet repository WACs. After decisions are made on the preferred disposition options, plutonium scrap and residue may be converted to final disposal forms rather than to storage forms.

3.2.1.2 Transuranic Waste. Any plutonium containing waste with transuranic concentrations in excess of 100 nCi/g is classified as transuranic waste (TRUW). These materials are planned to be disposed of in WIPP, located in New Mexico, and must meet the WIPP WAC and safeguards termination requirements before disposal.

3.2.1.3 Low-Level Waste. Low-level waste (LLW) from plutonium facilities is defined as waste containing less than 100 nCi/g of transuranic elements. The WAC for LLW is dependent upon the disposal site.

3.2.2 Description and Status for Waste Form Requirements

There are no current R&D activities that specifically address problems associated with meeting WAC.

3.2.3 Newly-proposed R&D Activities/Technologies

There are no newly proposed R&D activities or technologies at this time.

3.2.4 Evaluation of Current and Proposed R&D Against Requirements

The technical requirements to meet acceptance criteria for waste products from stabilization activities are well understood for TRUW and LLW and integrated into the stabilization R&D projects. However, if the stabilization program pursues can-in-canister approaches (currently considered 'back-up' with high schedule risk), which would ultimately result in emplacement in the high-level waste repository, treatment of the materials should be closely coordinated with the DOE-MD and DOE-Office of Civilian Radioactive Waste Management (DOE-RW) programs.

3.2.5 Conclusions and Recommendations

Conclusions:

Requirements for meeting LLW and TRUW acceptance criteria are well understood; however, requirements for disposition to the high-level waste repository are not well understood.

Recommendations:

1. DOE Office of Environmental Management (DOE-EM) should monitor the decision made by DOE-MD concerning plutonium disposition and identify materials that should be treated in coordination with the DOE-MD program.

2. Close coordination between the stabilization program and waste disposal sites should be maintained to ensure stabilized material meets the appropriate WAC.

3.3 Safeguards and Security Requirements

3.3.1 Introduction

Stabilization activities will generate (1) plutonium forms designed for long-term storage that meet DOE-STD-3013 and (2) waste streams. The stabilized plutonium in storage will require full safeguards and appropriate security. The waste streams also contain some plutonium. In the past, any stream containing significant quantities of plutonium was processed to recover that plutonium. The result was that safeguards could be significantly reduced on the TRUW because the quantities of plutonium in the TRUW were extremely low.

Today, plutonium is in excess supply. There are strong economic and schedule incentives for directly disposing of low-grade plutonium scrap and residue containing higher concentrations of plutonium as TRUW. Similarly, there are economic incentives to not process any materials from decommissioning operations to recover plutonium.

The WIPP facility and associated storage and transport systems for disposing of TRUW are designed with physical protection measures equivalent to the following Category IV requirements from DOE Order 5633.3B:

- WIPP is a property protection area with security controls adequate for plutonium meeting the WIPP WAC
- No materials control and accountability is envisioned for plutonium at WIPP since plutonium forms entering WIPP will meet the safeguards termination/reduction criteria promulgated by the DOE Office of Safeguards ad Security
- Plutonium entering the WIPP will have a measured value either established or confirmed at either the safeguards termination point or upon transfer from the generation/ stabilization/ packaging site(s). No post-emplacement safeguards controls outside property protection for WIPP are anticipated or required by DOE.

3.3.2 Description and Status for Safeguards and Security Requirements

The DOE has recently issued guidance on allowable plutonium concentrations in materials for safeguards termination (DOE memorandum from Edward J. McCallum, Office of Safeguards and Security, NN-51, Subject: *Additional Attractiveness Level E Criteria for Special Nuclear Material (SNM)*, dated July 22, 1996.). The DOE memorandum that provides maximum plutonium concentrations on which safeguards can be terminated, also identifies higher concentrations on which reduced safeguards can be applied. These reduced safeguards include waiving performing measurements and physical inventories requirements that are required for assigning safeguard requirements for materials coming from decommissioning plutonium facilities, stabilizing plutonium scrap and residue, and plutonium operations.

The International Atomic Energy Agency (IAEA) has established its own safeguards criteria for the long-term storage of stabilized plutonium. These criteria do not correspond to the reduction of safeguards and security levels proposed by DOE. For example, the IAEA will likely require more rigorous measurements (e.g., quantitative to include material movement and sampling to address potential material anomalies) versus more qualitative measurements (e.g., confirmation of total mass or gross radiation emission), the latter allowed for under current Departmental policy. At the same time, remote monitoring, incorporating elements of both attribute testing and containment/surveillance (C/S), may be acceptable to the IAEA if these approaches meet long-term IAEA safeguards criteria.

The plutonium stabilization and packaging process provides a good opportunity to exploit process monitoring approaches to meet safeguards requirements. Routine processing activities such as weighing and visual examination by operating personnel can provide a high degree of detection and assessment relative to potential diversion of plutonium. This capability may obviate the need for measurements solely to confirm the presence of plutonium. However, diversion scenarios involving the removal of plutonium and substituting surrogate materials to conceal diversion must be evaluated for the Plutonium Stabilization and Packaging Systems (PuSPS) in order to successfully implement process monitoring.

3.3.3 Newly-proposed R&D Activities/Technologies

3.3.3.1 Mixing TRUW With Secondary Materials Allows for Reduced Safeguards and Security. The objective of this effort is to identify where to mix plutonium-bearing materials with TRUW to create a final form that allows for reduced safeguards and security. Much TRUW inventory must be processed to be acceptable for disposal at WIPP. If plutonium recovery is impractical and safeguards can be reduced, it may be feasible to combine plutonium residues and TRUW with very low concentrations of plutonium to produce a final TRU waste. Such options would meet stabilization requirements for some residue streams, allow for the reduction of safeguards and security, and meet WIPP WAC.

3.3.4 Evaluation of Current and Proposed R&D Against Requirements

Although there are currently no documented R&D requirements, treatment methods must be developed to convert materials with plutonium into TRUW forms that allow for reduced safeguards and security. Technologies in Section 4 provide possible approaches (e.g., vitrification and ceramification). An assessment is needed to determine which baseline stabilization approaches will not meet safeguards and security requirements.

Research and development needs should consider the requirement to measurement plutonium materials being subjected to safeguards termination/reductions. This is necessary (1) to provide assurance that only practicably or practically irrecoverable forms of plutonium are being removed from stringent safeguards controls and (2) to allow for material balances to be closed around stabilization processes. Such R&D needs may include improved material holdup, waste and residue measurement capabilities relative to the PuSPS and waste containers.

DOE-STD-3013-96 (plutonium packaging criteria) identifies specific safeguards and surveillance/inspection requirements for plutonium containers which can define R&D needs.

Decisions regarding the availability and selection of the containerized stabilized plutonium for IAEA safeguards inspection can potentially drive decisions regarding R&D needs, particularly in the area of measurements and containment/surveillance.

3.3.5 Conclusions and Recommendations

Conclusions:

1. Stabilization technologies will be required to design safeguards-compatible waste forms.

2. Options for meeting safeguards and security requirements need further evaluation, since there are significant uncertainties associated with each option. Activities must be coordinated with other DOE organizations and other interested parties.

Recommendations:

1. DOE-EM should monitor decisions regarding the allowable concentration of plutonium in TRU for disposal at WIPP to determine how plutonium residues can be most effectively processed.
2. An Assessment is required to determine which baseline stabilization approaches will/will not meet safeguards and security requirements.

4. PLUTONIUM STABILIZATION

4.1 Plutonium Oxides (> 50%)

4.1.1 Introduction

The end states for plutonium metals and oxides are included in DOE-STD-3013, which states that stabilization systems must be developed to eliminate organics and plastics from storage packages and thermally stabilize metals and oxides prior to repackaging.

The information provided in Table 4.1-1 relates applicable NMSTG milestones identified in the Implementation Plan to the R&D Need Date, as reflected in a site-specific SISMP. The **R&D Need Date** is the date by which a particular technology must be available, including personnel, equipment, facility, and safety readiness. The **DOE Site** is the site responsible for completing the milestone. The **Milestone Text** is a summary of the milestone, as described in the Implementation Plan.

Plutonium metal and oxide (> 50%) R&D Need Dates, driven by 94-1 Implementation Plan milestone, are shown in Table 4.1-1. The RFETS processing line should be available to meet the October 1997 need date.

Table 4.1-1. Driving Implementation Plan Milestone Requirements for Plutonium Metal and Oxide (>50%) Stabilization

NMSTG Milestone Number	R&D Need Date	NMSTG Milestone Due Date	DOE Site	Milestone Text
IP-3.2-022	Oct 1997	Sept 1998	RFETS	New plutonium metal/oxide processing line operational in Building 371 at RFETS.

4.1.2 Description and Status for Plutonium Oxides (> 50%) Technologies.

4.1.2.1 PuSPS Stabilization Process (NMSTG). The NMSTG is procuring an end-to-end automated system from British Nuclear Fuels Limited (BNFL) that will result in stabilized and packaged plutonium oxides and metals in standardized packages that meet DOE-STD-3013 criteria. The prototype will be installed at RFETS in B/707 Module J. Subsequent units will be installed at RFETS in B/371, Hanford, and SRS. The system is comprised of a stabilization process and a packaging process.

Existing plutonium storage containers are transferred from vault storage (using either an existing conveyor or a manual transfer trolley) to a Receipt Area for entry to the stabilization process. The site storage packages from the transfer trolley are then checked for contamination upon opening the outer container and, if the inner container has leaked, transferred into the Material Preparation Area. Otherwise, just the inner container (including packing material, if present) enters the Material Preparation Area through an airlock. Site convenience cans from the existing conveyor are transferred directly into the Material Preparation Area, also through an airlock. In the nitrogen inerted glovebox-like atmosphere of the Material Preparation Area, packing material is cut off and inner containers are opened. To minimize the spread of airborne contamination into other, cleaner areas, this work station area within the Material Preparation Area has entry and exit isolation doors that are normally kept closed. Plutonium oxides and small metal pieces are then exposed to sustained, elevated temperatures in a stabilization furnace under a dry air atmosphere.

A System Design Review was held, and results are documented in the PuSPS System Specification and the PuSPS System Design Document. Completion of end-to-end system testing with a cold surrogate material is scheduled for October 1997. System operational checkout with hot material is scheduled for completion in October 1998.

4.1.3 Newly-Proposed R&D Activities/Technologies

There are no newly-proposed R&D activities at this time.

4.1.4 Evaluation of Current and Proposed R&D Against Requirements

The effort discussed in section 4.1.2.1, along with R&D supporting the storage standard in section 3, provides the needed technology base for meeting the milestone in Table 4.1-1. No additional R&D requirements have been identified at this time.

4.1.5 Conclusions and Recommendations

Table 4.1-2 summarizes the status of a particular technology, in this case the PuSPS Stabilization Process, related to R&D needs identified in this plan. This summary table is repeated throughout the plan as part of the various conclusions and recommendations sections and is used to establish the basis for specific conclusions or recommendations. The **R&D Program** heading refers to the applicable section of this plan that describes the technology. The **R&D Site** reflects the organization responsible for developing a particular technology, while the **End Use Site** indicates where the technology is to be applied. The **94-1 Milestone** is derived from the Implementation Plan. The **R&D Need Date** is the date by which a particular technology must be available, including facilities, equipment, personnel, and safety readiness. For each technology, the **Recommendation** is identified as either baseline, competitive alternative, or backup. Within this context, baseline refers to a technology recommended to be pursued in this plan for a particular use at a specific site and reflect anticipated changes to site baselines that will be documented in the SISMPs. A competitive alternative is one of two or more technologies that are candidates for a baseline application, and a backup technology is one that should be pursued only if baseline or competitive alternative approaches are not successful. Each technology has also been rated relative to its technical maturity.

NOTE: A technology can have significantly different technical maturity scores at different end-use sites, based on varying levels of readiness with respect to facilities, equipment, personnel, and safety. A low technical maturity score represents a mature technology, whereas a high score represents a less mature technology. For a more detailed description on maturity scores, refer to Appendix C, *System Analysis Methodology and Raw Data.*

Conclusions:

By referring to Table 4.1-2 below, it was concluded that the PuSPS Stabilization Process represents a medium programmatic risk.

Recommendations:

The NMSTG should arrange for adequate tracking of the technical and programmatic progress of the PuSPS Stabilization Process to ensure that the need date will be met.

Table 4.1-2. Summary Technology Status for Plutonium Metal and Oxide (>50%) Stabilization

R&D Program	R&D Site	End Use Site	94-1 Milestone	R&D Need Date	Recommendation	Technical Maturity
4.1.2.1 PuSPS Stabilization	NMSTG	RFETS	IP-3.2-022	Oct 1997	Baseline	3.65

4.2 Plutonium Solutions Stabilization

4.2.1 Introduction

Plutonium solutions are acidic, inherently unstable, and corrosive; can leak from containment; and undergo radiolysis. Radiolysis results in loss of acidity, as well as the potential for precipitation from reacting with degradation products and impurities. Radiolysis will also generate hydrogen, which can accumulate in explosive concentrations if not vented or removed. Because these solutions are unstable, plutonium must be separated and stabilized in the near term. Plutonium concentrations vary from less than 1 gram per liter to over 300 grams per liter.

Acid solutions containing plutonium exist in both nitrate and chloride forms at SRS, RFETS, and Hanford. Shipping plutonium solutions to a common processing location could provide a general solution to disposing of the materials; however, shipping kilogram (kg) quantities of plutonium solutions via commercial transport has been expressly prohibited by the U.S. Nuclear Regulatory Commission (NRC) for over 20 years. Unless an exemption is obtained from existing regulations, plutonium solutions must be processed at the sites where they are currently stored. Each site has assessed the facilities, equipment, and technologies necessary to facilitate stabilization. Development of selected processes must be based on the safe and economic stabilization of plutonium and accomplished within the 94-1 schedule.

Implementation Plan milestones for stabilizing plutonium in solutions at SRS, RFETS, and Hanford are given in Table 4.2-1. Technologies related to this stabilization are presented in the following sections.

Table 4.2-1. Driving Implementation Plan Milestone Requirements for Stabilization Processes for Plutonium Solutions**

NMSTG Milestone Number	R&D Need Date	NMSTG Milestone Due Date	DOE Site	Milestone Text
IP-3.1-020A*	Nov 1996	Nov 1996	RFETS	Start draining B771 hydroxide tanks and begin processing
IP-3.1-022	Jun 1997	Jun 1997	HAN	Begin processing solutions at PFP.
IP-3.1-012*	Oct 1999	Oct 2000	SRS	Stabilization operations completed for Pu-239 solutions in SRS's H-Canyon (34,000 liters)
IP-3.1-020D*	Nov 1997	Nov 1997	RFETS	Start draining five (5) B771 high-level tanks and begin oxalate processing

*Proposed 94-1 IP change. ** Refer to Table 4.1.1 for description of table headings.

4.2.2 Description and Status for Stabilization Processes for Plutonium Solutions Technologies

4.2.2.1 Extraction Chromatography Pretreatment (Hanford). Removing interfering elements (e.g., sodium, potassium) from plutonium-bearing solutions facilitates the efficiency of the vertical calciner to convert these solutions to stable, storable solids. The EIChrom extraction chromatography technique pretreats acid solutions of plutonium and other actinides to remove interfering elements. Plutonium and other actinide elements are selectively removed from acid solutions by extracting chromatographic resins. Plutonium is extracted from solutions and passed through a column containing the resin. The plutonium is then eluted from the resin by a solution containing a complexant, such as citrate. Finally, the resulting plutonium solution is converted to solid form by calcination in the vertical calciner.

The Functional Design Criteria (FDC) and the Conceptual Design Review (CDR) report were submitted and approved by the DOE Richland Operations Office (DOE-RL) in September 1996. Advanced Conceptual Design is underway, and the design activity for installation will be complete in December, 1996 to support the milestone IP-3.1-022 start date of June 1997.

4.2.2.2 Vertical Calciner (Hanford). The Plutonium Finishing Plant (PFP) at Hanford has a significant quantity of plutonium-bearing filtrate and product solution from weapons production activities at the Plutonium Uranium Extraction (PUREX) facility and at PFP, as well as miscellaneous other solutions, for a total of 4804 liters of solutions. Even if an exemption to NRC transportation restrictions can be obtained, no approved packaging system is available to ship plutonium-bearing solutions to other sites for disposition. The vertical calciner at Hanford has been designed to directly convert these plutonium solutions to a stable, storable solid that minimizes personnel exposures and waste production.

In the direct denitration processes used in the vertical calciner, small additions of plutonium-bearing solutions are metered into a continuously heated and stirred bed of solids. Calcination proceeds through rapid evaporation of liquid, denitration, and final heat treatment to stable plutonium dioxide. The process is known to work with solutions containing plutonium, thorium, uranium, and mixtures of these elements in concentrations ranging from 15 to 500 grams per liter. The calciner has been successfully hot-tested using high- and low- concentration plutonium solutions.

The FDC and CDR for the Vertical Calciner production unit installation, with the development program data as its bases, were provided to DOE-RL in September 1996 for Project Authorization. Advanced Conceptual Design is also underway. The design activity for the calciner and its supporting systems will be complete in December 1996 to support the milestone IP-3.1-022 startup date of June 1997. The development calciner is being operated in a pre-production mode to gather added operating experience and longevity information. Approximately 350 liters of solution inventory have been stabilized to date during this pre-production mode.

4.2.2.3 Precipitation-Oxalate/Hydroxide (RFETS/LANL). Plutonium-bearing solutions stored at RFETS are an ever-increasing risk to worker safety and will eventually become a risk to the environment and to the public. As such, they are listed as top priority items for treatment in the RFETS DNFSB 94-1 response. Processing and stabilizing these solutions into forms that meet interim or long-term storage criteria will immediately reduce risk at the site.

Simple precipitation techniques have been developed to stabilize plutonium nitrate solutions. The baseline RFETS process includes feed adjustment, reduction of Pu (IV) to Pu (III), precipitation with oxalic acid, filtration, and filtrate removal. This technology has been demonstrated at the Los Alamos Plutonium Facility (TA-55) using actual nitrate solutions and full-scale RFETS equipment. Technology transfer to RFETS has been largely completed, including the development of formal procedures and operator training in a production environment. Once begun, the processing duration at RFETS is expected to take 9 months to stabilize about 85 kg of plutonium in 750 liters of solution.

4.2.2.4 Plutonium Solution Vitrification (SRS). One option for stabilizing plutonium solutions now stored in the SRS H-Canyon involves vitrification after the americium/curium (Am/Cm) and neptunium (Np) materials are stabilized. The process for the plutonium solutions is similar to that used for the Am/Cm solutions, and the same equipment could be used as a pilot demonstration for the vitrification of plutonium solutions. The process is being demonstrated in FY96, and various glass formulations are being examined. Transport of the solutions from H-Canyon to F-Canyon is required for this stabilization option.

4.2.3 Newly-Proposed R&D Activities/Technologies

4.2.3.1 Ceramification of Plutonium Solutions (RFETS). Ceramification as a process was originally developed to provide a fire-safe barrier to weapons parts and prevent the release of plutonium in the event the device was subjected to high heat. The process was used at RFETS for five years. Ceramification has been proposed as a technique for stabilizing high-level plutonium-bearing solutions and plutonium oxides. The process is capable of producing a dense, chemically stable product that has all of the physical properties of a high-fired ceramic monolith. A density that is 98% of the theoretical density can be achieved if desired.

The ceramification process begins with a loose matrix, such as unstabilized plutonium oxide. The oxide is infused with the plutonium-bearing nitrate solutions, and the mixture is heated to about 550C to drive off the water and convert the nitrate to oxide. The nature of this converted oxide is such that it acts as a strong binder for the matrix. The first infusion/calcine step produces a monolith that is about 50% dense with relatively poor physical properties. Subsequent infusions, followed by a calcine step after each infusion, increase the ceramic density and strength.

The amount of processing will vary depending on the quality of the starting matrix and the concentration of plutonium and other heavy metals in the nitrate solutions. However, the process is robust and capable of handling a wide variety of oxides and solutions. All that is required is to vary the number of solution infusions.

4.2.4 Evaluation of Current and Proposed R&D Against Requirements

The technologies described in Sections 4.2.2 and 4.2.3, along with core technology efforts discussed in Section 9.2.1.1, provide an adequate technology base to meet the stabilization requirements outlined in Table 4.2-1. No additional R&D requirements have been identified at this time.

4.2.5 Conclusions and Recommendations

By referring to Table 4.2-2, it was concluded that extraction chromatography represents a low programmatic risk for meeting its R&D need date. The vertical calciner and oxalate/hydroxide precipitation baseline technologies represent medium risk for meeting their R&D need date. As medium risk baseline technologies, the NMSTG should arrange for adequate tracking of their technical and programmatic progress to ensure that the need dates will be met. Plutonium Solution Vitrification (see Section 4.2.2.4) should only be pursued if it is supported by DOE-MD as a plutonium disposition demonstration. Ceramification of plutonium solutions (see Section 4.2.3.1) should not be pursued since it would probably not be timely for the R&D need date and produce a form that would be difficult for the DOE-MD program to treat for disposition. However, ceramification has been considered for stabilizing ash residues (see Section 4.3.5.2).

<u>Conclusions</u>:

1. Extraction chromatography is sufficiently mature to meet R&D need dates.
2. The vertical calciner at Hanford is medium risk and requires adequate tracking to ensure that its need date is met.

<u>Recommendations</u>:

R&D support should be continued for mature technologies as they relate to technology transfer (e.g., transfer of the plutonium oxalate precipitation process from LANL to RFETS) and startup of operations, and NMSTG should arrange for adequate tracking of progress in completing the technology transfer.

Table 4.2-2. Summary Technology Status for Stabilization Processes for Plutonium Solutions **

R&D Program	R&D Site	End Use Site	94-1 Milestone	R&D Need Date	Recommendation	Technical Maturity
4.2.2.1 Extraction Chromatography	HAN	HAN	IP-3.1-022	Jun 1997	Baseline	2.67
4.2.2.2 Vertical Calciner	HAN	HAN	IP-3.1-022	Jun 1997	Baseline	3.44
4.2.2.3 Precipitation - Oxalate/Hydroxide	RFETS /LANL	RFETS	IP-3.1-020A*	Nov 1996	Baseline	3.09
4.2.2.4 Plutonium Solution Vitrification	SRS	SRS	IP-3.1-012*	Oct 1999	Backup	5.70
4.2.3.1 Ceramification of Plutonium Solutions	RFETS	RFETS	IP-3.1-020D*	Nov 1997	Backup	3.79

* Proposed 94-1 IP change. ** Refer to 4.1.5 for description of table headings.

4.3 Plutonium Residue Stabilization

4.3.1 Pyrochemical Salts

4.3.1.1 Introduction. Over 18 metric tons of pyrochemical salt residue are in storage within the DOE weapons Complex. These salts are generally considered unacceptable (high risk) for safe interim storage, as specified in DNFSB Recommendation 94-1. Potential hazards associated with pyrochemical salts include corrosion of containers, gas generation from radiolysis, and the presence of reactive metals. These salts were primarily generated from the following four operations: electrorefining (ER), molten salt extraction (MSE), direct oxide reduction (DOR), and salt scrub. The MSE salts are high in americium and, therefore, have intense gamma radiation levels. About 16 metric tons of salt residue are stored at RFETS and about 2 metric tons are stored at LANL. In the 94-1 Site Remediation Plan, RFETS committed to stabilize 6 metric tons of higher risk salts by the end of August 1997, as shown in Table 4.3.1-1.

The salt stabilization R&D need date, driven by the 94-1 Implementation Plan milestone, is shown in Table 4.3.1-1. The need date appears to be supported by current R&D activities.

Table 4.3.1-1. Driving Implementation Plan Milestone Requirements for Salt Stabilization**

NMSTG Milestone Number	R&D Need Date	NMSTG Milestone Due Date	DOE Site	Milestone Text
IP-3.3-012A*	Aug 1997	Aug 1997	RFETS	Begin stabilization by pyrochemical oxidation and repackage 6,000 kg of higher risk plutonium-containing salts.

* Proposed 94-1 IP change. ** Refer to 4.1.1 for description of table headings.

4.3.1.1.1 Results of Trade Studies—The salt trade study evaluated many options for treating/stabilizing pyrochemical salts. The recommended option was a four-fold processing effort at LANL, Lawrence Livermore National Laboratory (LLNL), RFETS, and SRS. Directly discarding the salts to WIPP was acceptable, but not recommended. Pyrooxidation was only attractive for salts with low plutonium contents. To maximize the probability of success, the trade study recommended that stabilization work be distributed between 3 sites—RFETS, LANL, and LLNL. Additional follow on work on any scrub alloy product would be performed by SRS. The study recommended that (1) RFETS should continue installing furnaces to oxidize the salts, (2) LANL should continue developing and deploying salt distillation technology, and (3) LLNL should continue demonstrating and deploying salt scrub technology and SRS should plan to receive and process scrub alloy from salt scrub.

4.3.1.2 Description and Status for Plutonium Salts Technologies.

4.3.1.2.1 Salt Oxidation (RFETS/LANL)—Salt oxidation eliminates reactive species, bound water, and most chemically-bound water from salt residues. Salt stabilization is performed by adding an oxidant (e.g., sodium carbonate) to the molten salt; any reactive species will then be oxidized. At the high temperatures required to melt the salts, water is driven off, the moisture content is reduced, and the myriad plutonium species are converted to oxide, which then segregates into the lower fraction of the molten salt. This produces an oxide concentrate encased in the salt matrix. The upper portion of the salt may contain a small enough amount of plutonium to allow direct disposal to WIPP.

In FY96, researchers completed demonstrations of salt oxidation (using actual inventories of MSE), ER, and DOR salts at LANL, and optimized the oxidation process for ER salts. The plans for FY97 include optimizing the oxidation process for MSE salts and salt strip salts and supporting technology transfer of the capability to RFETS.

4.3.1.2.2 Salt Distillation (RFETS/LANL)—Salt oxidation converts the salt inventories to a stable form; however, the plutonium content and ease of plutonium recovery may preclude disposal of stabilized salt in WIPP. Salt distillation provides a method for separating plutonium-containing pyrochemical salts into a lean salt fraction (<100 ppm plutonium) and plutonium oxide suitable for long-term storage.

Separation of sodium-potassium chloride (NaCl/KCl) salts from plutonium has been demonstrated on a pilot scale, and new equipment has been designed, fabricated, and installed for production-scale capability. Full-scale testing will establish the reliability of the equipment and the robustness of the process.

Initial testing on the full-scale distillation unit suggests that the process will work well for the NaCl/KCl salts at RFETS and LANL. However, the current design is not suitable for the calcium chloride ($CaCl_2$) salts. The only disadvantages of the process are that it requires two steps (oxidation followed by distillation) and new equipment that is not readily available at the DOE sites.

Development of a suitable processing route for $CaCl_2$ salts was started in FY96. Some of the processing schemes under investigation involve high-temperature (>1200C) distillation of the salt or molten salt filtration to separate the plutonium oxide. Another approach involves aqueous dissolution of the $CaCl_2$ matrix, followed by filtration and then by distillation of the salt solution to recover both the water (for recycle into the process) and a solid $CaCl_2$ waste stream. After development is complete, a full-scale demonstration of the chosen technology will be required in FY97.

In FY97, equipment will be purchased to perform a full-scale demonstration that will mirror the effort for NaCl/KCl salts. Pyrochemical salts at LANL will be used as a feed for a process designed to handle a 3 kg batch size in a 24-hour period. Initial purchase of the equipment will be followed by testing on nonradioactive material, installation, testing on radioactive material, and a production-demonstration run.

4.3.1.3 Newly-Proposed R&D Activities/Technologies.

4.3.1.3.1 Salt Scrub/Oxidation (LLNL)—Automated titration of RFETS salts to collect the actinides in a consolidated metal form and to convert the salt into a form acceptable for WIPP disposal will be demonstrated using actual RFETS salts. The process equipment and associated control system will be developed and demonstrated for use with the RFETS NaCl/KCl based salts. Optimum operating parameters will be established, and the process will be evaluated on the basis of actinide separation efficiency (to validate that the residual plutonium in the salt is low enough for WIPP disposal), process time/conditions, and other factors used in the salt residue trade study analysis.

Some testing of salt scrub to determine effectiveness of the process on aged (i.e., oxidized) salts is needed. Due to extensive studies and actual processing at RFETS, LANL, and LLNL, salt scrub is the easiest process to implement for stabilizing the salt residues. However, it has the disadvantage that the salt product may be too reactive to be easily transported or disposed of at WIPP due to excess reductant in the salt. Carefully titrating the reductant should eliminate the reactivity problem. Alternatively, the salt could be air sparged after the salt scrub process. The titration would require modification to equipment that is readily available at the DOE sites; air sparging would require a minor modification to existing DOE facilities.

4.3.1.3.2 Salt Filtration (LLNL)—Salt filtration is an extremely simple technique that requires no specialized equipment and is not expected to be specific to any particular salt feed type or to the degree of oxidization from extended storage. If shown to be effective, salt filtration could be implemented extremely quickly. One disadvantage of salt filtration is that a plutonium salt/oxide mixture is obtained in the concentrate. Even though the plutonium content may exceed 50%, it is unlikely that it could be stabilized by simple calcination at 950C. It will probably have to be washed with water first.

This task will develop molten salt filtration using a ceramic (zirconia or equivalent) filter medium to condition a major category of pyrochemical salt residues in the DOE Complex. After demonstrating the degree of feasibility with available on-site salt residues at LLNL, other site's residue salts will be used to further define or optimized process parameters. The process will be evaluated on the basis of actinide separation efficiency, process time/conditions, and other factors used in the salt residue trade study analysis.
Upon successful completion of this work, molten salt filtration will be sufficiently developed for use on actual salt residues at RFETS, LANL, or LLNL using standard pyro-

chemical processing equipment. Any final optimizations of the process for efficiency and cycle time can be performed while stabilizing actual salt residues.

4.3.1.3.3 Salt Washing (LLNL)—The purpose of this effort is to develop, demonstrate, and deploy a system to remove residual plutonium from RFETS ER (NaCl/KCl) salts after the bulk of the actinides have been removed by salt scrub or salt filtration. This secondary treatment will allow the salts to be disposed of as certifiable LLW. The true advantage of developing this process is to provide an immediate means to dispose of the depleted salt. This would simplify stabilization of RFETS salts at other sites since the treated salt would not increase TRU waste inventory liability.

The proposed operation would use simple equipment that is modular and readily used in the field. A tri-party effort will be mounted by Rocky Mountain Remediation Services (RMRS), LLNL, and LANL.

4.3.1.3.4 Electroscrub (RFETS/SRS)—The electroscrub process is run exactly the same way as an aluminum reduction (Hall-Heroult) cell. A ceramic crucible is filled with the matrix to be scrubbed and an appropriate alloying agent (Al, Bl, Sn), brought to temperature (~850C), graphite (or thallium-composite) anodes are lowered along with a ceramic-sheathed cathode current collector until the cathode current collector makes contact with the cathode alloy material and the anodes are submerged to about 20mm in the molten matrix. DC power is supplied at about 50A, and the steady-state cell voltage is watched until an increase in the voltage-time slope is observed. The electrodes are removed from the melt and the power to the furnace is cut. The charge is then allowed to solidify and cool. The two products will be the metal alloy, which will contain the actinides and heavy metals, and salt. The salt will be stripped of its actinide and heavy metal content, and any reactive metal which was initially present will be oxidized. The salt will be appropriate for WIPP disposal and the alloy will be acceptable F-canyon feed.

The electroscrub process was proposed since it offers the advantages of using both the salt oxidation process and salt scrub. Since electrochemistry is used to perform both an oxidation and a reduction, the process produces an alloy button form for the plutonium that is ideal for long-term storage and does not leave behind unreacted calcium metal in the salt. In fact, electroscrub will oxidize any light, reactive metals present in the salt at the start. This process has been proven in the aluminum industry as the Hall-Heroult process. More recently, investigators at the Japanese Central Research Institute of Electric Power Industry have used this approach to extract actinides from spent nuclear reactor fuel.

It should be noted that since the electroscrub process is highly tolerant of partially oxidized and hydrated salts, it could be used for a number of residues beyond pyrochemical salts. For example, electroscrub could be used to stabilize and extract actinides from SS&C, pyrochemical crucible residues, incinerator ash, and possibly graphite fines.

4.3.1.3.5 Glass Material Oxidation and Dissolution System (ORNL)—Calcium salts are not easily distilled; therefore, it is proposed that the Glass Material Oxidation and Dissolution System (GMODS) process be used to immobilize these residues for disposition as TRU waste. (see Section 4.3.8.3.1 for a detailed description of the GMODS process.)

4.3.1.4 Evaluation of Current and Proposed R&D Against Requirements. The technologies described in Sections 4.3.1.2 and 4.3.1.3 provide an adequate technology base to meet the stabilization requirements outlined in Table 4.3.1-1; no additional R&D requirements have been identified at this time.

4.3.1.5 Conclusions and Recommendations. By referring to Table 4.3.1-2 below, the following conclusions are made.

Conclusions:

1. If stabilizing pyrochemical salts was the only objective, then salt oxidation would be the only required R&D activity. However, due to long-term storage issues, the plutonium needs to be removed from the salts and the salts processed to meet WIPP acceptance and safeguards termination criteria.
2. Pyrochemical salt scrub/oxidation represent low programmatic risk for R&D need date.
3. Salt distillation represents medium programmatic risk for R&D need date.
4. R&D at LANL for pyro-oxidation is essentially complete.
5. Testing of salt scrub on aged (oxidized) salts to determine process effectiveness is needed.

Recommendations:

1. Salt distillation and salt scrub should continue to be pursued for treating/stabilizing pyrochemical salts in addition to direct discard to WIPP.
2. Reactivity testing of unstabilized salts and scrubbed salts should continue until DOE establishes clear guidance on reactive materials that are unacceptable for WIPP.

Table 4.3.1-2. Summary Technology Status for Salt Stabilization **

R&D Program	R&D Site	End Use Site	94-1 Milestone	R&D Need Date	Recommendation	Technical Maturity
4.3.1.2.1 Salt Oxidation	RFETS/ LANL	RFETS	IP-3.3-012A*	Aug 1997	Baseline	3.02
4.3.1.2.2 Salt Distillation	LANL	RFETS	IP-3.3-012A*	Aug 1997	Baseline	3.44
4.3.1.3.1 Salt Scrub/Oxidation	LLNL	RFETS	IP-3.3-012A*	Aug 1997	Baseline	2.72
4.3.1.3.2 Salt Filtration	LLNL	RFETS	IP-3.3-012A*	Aug 1997	Backup	3.35
4.3.1.3.3 Salt Washing	LLNL	RFETS	IP-3.3-012A*	Aug 1997	Backup	8.23
4.3.1.3.4 Electro-scrub	RFETS/ SRS	RFETS	IP-3.3-012A*	Aug 1997	Backup	3.93
4.3.1.3.5 GMODS	ORNL	RFETS	IP-3.3-012A*	Aug 1997	Backup	7.49

* Proposed 94-1 IP changes. ** Refer to 4.1.5 for description of table headings.

4.3.2 Sand, Slag, and Crucible (SS&C)

4.3.2.1 Introduction. SS&C is a residue that was generated at Hanford, LANL, RFETS, and SRS during the process reduction of plutonium compounds from weapons disposition. The process, metallothermic reduction, generated a large amount of associated material that retained too much plutonium to simply be discarded. SS&C was stockpiled, and since process capacities at RFETS could not keep up with the production, inventories of this material were accumulated. SS&C contains at least 50 wt% of calcium fluoride and, as such, presents problems from a dissolution and corrosion standpoint.

Sand, slag and crucible inventories require treatment to eliminate reactive species and moisture content that have caused storage container failures. The primary objective of the R&D efforts at LANL in FY96 was to provide preliminary data for the SS&C Trade Study. Areas of investigation included thermal stabilization, magnetic separation, aqueous recovery, densification, vitrification, and cementation.

Sand, slag, and crucible R&D need dates, driven by 94-1 Implementation Plan milestones, are shown in Table 4.3.2-1. Although the need dates are supported by current R&D activities, FY97 efforts will be guided by the conclusions of the Trade Study to determine which approaches should be further developed.

Table 4.3.2-1. Driving Implementation Plan Milestone Requirements for Stabilization of Sand, Slag, and Crucible **

NMSTG Milestone Number	R&D Need Date	NMSTG Milestone Due Date	DOE Site	Milestone Text
IP-3.3-014A*	Sep 1997	Sep 1997	RFETS	Begin stabilization of SS&C and graphite fines
IP-3.3-026A*	May 1998	Jan 2000	HAN	On-site stabilization of low-grade reactive solids (SS&C) completed.
IP-3.3-022	Jan 1997	Dec 1997	SRS	Processing of existing inventories of SS&C material completed.

* Proposed 94-1 IP change. ** Refer to 4.1.1 for description of table headings.

4.3.2.2 Description and Status for Sand, Slag, and Crucible Stabilization Technologies.

4.3.2.2.1 Sand, Slag, and Crucible Stabilization (SRS)—SS&C residues at the SRS will be dissolved either in FB-Line or in F-Canyon and the plutonium recovered through the solvent extraction process. The plutonium will be converted to metal in FB-Line. In addition, the recommendation of the SS&C Trade Study is to calcine, repackage, and send the RFETS material to SRS for stabilization. R&D activities have resulted in flow sheets and cycle time that are compatible with F-Canyon operations.

4.3.2.2.2 Carbonate Oxidation (RFETS/LANL)—Oxidation of actinide and reactive metals in pyrochemical salts has already been successfully demonstrated and expanded to include oxidation of actinide and reactive metals in SS&C residues. Thermal stabilization techniques are also being evaluated, but may not oxidize all free metals. Stabilization of SS&C residues is performed by adding an oxidant (e.g., sodium carbonate) to the molten residue. Any reactive species will then be oxidized. At the high temperatures required to melt the residue, water is driven off, the moisture content is reduced, and the myriad plutonium species are converted to oxide. The material will then be transferred to the PuSPS for final stabilization and packaging.

4.3.2.2.3 Nitric Acid Soluble Bags for FB-Line Scrap (SRS)—An alternate flow sheet was studied to permit the processing of SS&C residues in F-Canyon instead of the FB-Line. As part of flow sheet development, studies are being conducted with acid-soluble bag materials, which might replace insoluble polyvinyl chloride (PVC) and polyethylene bags.

The successful candidate material must dissolve in a 35-50% nitric acid solution after about 12 to 24 hours. Out of 13 candidate plastic materials, nylon was chosen as the best candidate material. Once nylon was selected, the effect of the dissolved nylon bags on the F-Canyon evaporator was tested. Nylon bag materials were dissolved in 7 molar nitric acid solutions to an organic concentration of about 1400 ppm. The acid solution was heated in an evaporator for 50 hours at 114C. Samples were obtained throughout the test; their analyses showed little or no change in the total organic concentration in the evaporator acid. Further testing of the nylon material included its suitability to be sealed using a heat sealer. The nylon sealed very well using the FB-Line heat sealer, a Vertrod 14T-1/2 system. No additional R&D is expected for the successful implementation of this technology.

4.3.2.2.4 Cementation (Hanford)—SS&C residues at Hanford comprise approximately 2420kg bulk weight of material in inventory and represents approximately 43kg of plutonium. The process mixes ground SS&C with water to react metallic calcium present in the SS&C and render the SS&C non-reactive for subsequent transfer to WIPP. The water/SS&C mixture is filtered and combined with a quantity of Portland cement sufficient to allow reduced safeguards and security requirements. A portion of the filtrate is used to complete the cementing. The remaining portion of filtrate is chilled and recycled into the next SS&C batch to minimize the volume of cement produced. The cemented SS&C is then packaged in containers, cured, bagged out of the glovebox, and placed into WIPP approved shipping containers. Cementing this residue will be complete in mid 1998, meeting the milestone IP-3.3-026A.

No further development effort is required to cement the SS&C materials at Hanford. Should recovery of the plutonium from the SS&C become a criterion, additional development/implementation actions will be required to affect recovery of plutonium from the SS&C material.

4.3.2.3 Newly Proposed R&D Activities/Technologies.

4.3.2.3.1 Microwave Vitrification for WIPP (RFETS/SRS)—The Sand, Slag, and Crucible Trade Study team recommend pursuing microwave processing as a means to vitrify SS&C residues. The vitrified product might be able to meet the requirement for reduced safeguards and security with a higher plutonium loading than calcined SS&C residues. The only issue that needs to be resolved is to demonstrate that the reactivity of calcium in the virtified product is acceptable under the WIPP WAC. Those vitrified SS&C residues could then be disposed of at WIPP.

4.3.2.3.2 Can-in-Canister (SRS)—An extensive R&D program is required to ensure that vitrification SS&C is compatible with the high-level glass logs certification process for disposal at the high-level waste repository. Such a program includes good characterization of SS&C materials, formulation of an appropriate glass, surrogate and radioactive tests, and certification of the can-in-canister final waste form. If a can-in-canister strategy were to be pursued, it should include a broader suite of residues, such as ash (see Section 4.3.5), to justify the expense of R&D and certification.

4.3.2.3.3 Vitrification for Can-in-Canister (RFETS)—This is the same process described in Section 4.3.5.2.3.

4.3.2.3.4 Electorscrub (RFETS/SRS)—This technology, discussed in section 4.3.1.3.4, has been proposed for stabilization of SS&C.

4.3.2.3.5 SS&C Hydrous Cementation (RFETS)—This technology is essentially the same as the Hanford cementation process (see Section 4.3.2.2.4) except that facilities and personnel readiness are less mature at RFETS.

4.3.2.4 Evaluation of Current and Proposed R&D Against Requirements. The technologies described in section 4.3.2.2 and 4.3.2.3 provide an adequate technology base to meet the stabilization requirements identified in Table 4.3.2-1. No additional R&D requirements have been identified at this time.

4.3.2.5 Conclusions and Recommendations. By referring to Table 4.3.2-2 below, the following conclusions are made:

Conclusions:

1. Stabilization by dissolution at SRS and nitric acid-soluble bags represent low programmatic risk for meeting R&D need dates.

2. Cementation at Hanford and calcining SS&C at RFETS for shipment to SRS represent medium programmatic risk for meeting R&D need dates.

Recommendations:

NMSTG should arrange for adequate tracking of the technical and programmatic progress of Hanford cementation to ensure that need dates will be met.

Table 4.3.2-2. Summary Technology Status for Stabilization of Sand, Slag, and Crucible **

R&D Program	R&D Site	End Use Site	94-1 Milestone	R&D Need Date	Recommendation	Technical Maturity
4.3.2.2.1 SS&C Stabilization	SRS	SRS	IP-3.3-022	Jan 1997	Baseline	0.02
4.3.2.2.1 Calcine for Shipment to SRS	RFETS	RFETS	IP-3.3-014A*	Sept 1997	Baseline	4.70
4.3.2.2.2 Carbonate Oxidation	RFETS/ LANL	RFETS	IP-3.3-014A*	Sept 1997	Backup	4.70
4.3.2.2.3 Nitric Acid Soluble Bags	SRS	SRS	IP-3.3-022	Jan 1997	Baseline	0.86
4.3.2.2.4 Cementation	RL	RL	IP-3.3-026A*	May 1998	Baseline	4.07
4.3.2.3.1 Microwave Vitrification for WIPP	RFETS	RFETS	IP-3.3-014A*	Sept 1997	Backup	4.33
4.3.2.3.2 Can-in-Canister	RFETS/ SRS	RFETS	IP-3.3-014A*	Sept 1997	Backup	4.98
4.3.2.3.3 Vitrification for Can-in-Canister	RFETS	RFETS	IP-3.3-014A*	Sept 1997	Backup	7.58
4.3.2.3.4 Electrochemical Scrubbing	RFETS	RFETS	IP-3.3-014A*	Sept 1997	Backup	3.93
4.3.2.3.5 SS&C Hydrous Cementation	RFETS	RFETS	IP-3.3-014A*	Sept 1997	Backup	4.05

* Proposed 94-1 IP changes. ** Refer to 4.1.5 for description of table headings.

4.3.3 Pyrochemical Crucible Residues

4.3.3.1 Introduction. Pyrochemical processes at RFETS and at LANL required the use of ceramic crucibles to remove product metal. The crucible material was broken into shards after the process contents were removed from the furnace and separated into salt, crucible, and product fractions. The separation of the crucible from the salt was not complete, leaving a significant fraction of crucible shards in the pyrochemical salt residues and salt in the crucible residues.

The pyrochemical crucible residue stabilization need date, driven by 94-1 Implementation Plan milestones, is shown in Table 4.3.3-1. Support of the need date is uncertain based on the outcome of R&D on the three backup technologies now being pursued.

Table 4.3.3-1. Driving Implementation Plan Milestone Requirements for Pyrochemical Crucible Residues *

NMSTG Milestone Number	R&D Need Date	NMSTG Milestone Due Date	DOE Site	Milestone Text
IP-ES-025	Feb 1998	May 2002	RFETS	Repackage all plutonium inorganic oxides and wet/miscellaneous residues (1,113 drums).

* Refer to table 4.1.1 for table headings.

4.3.3.2 Description and Status for Pyrochemical Crucible Residue Technologies. The crucible residue backlog stabilization process, documented in the RFETS SISMP, is to calcine the backlog in the same furnace that the SISMP indicates will be used for ash stabilization. This process has been demonstrated at LANL to be ineffective for crucible residues; therefore, other technologies are required.

4.3.3.3 Newly Proposed R&D Activities/Technologies. There are three proposed technologies: aqueous acid dissolution, molten salt washing, and electroscrub. Aqueous acid dissolution has been used at LANL in the past. However, the reactive metals on the shards have led to a number of process control problems. The molten salt washing process is still in the early stages of development at LANL and appears promising. The electroscrub process, as described in Section 4.3.1.3.4, does not require agitation, and could easily accommodate the shards.

4.3.3.4 Evaluation of Current and Proposed R&D Against Requirements. The technologies discussed in section 4.3.3.3 provide an adequate technology base to meet the stabilization requirements identified in Table 4.3.3-1. No additional R&D requirements have been identified at this time.

4.3.3.5 Conclusions and Recommendations.

Conclusions:

1. Data suggest that the RFETS baseline technology for the stabilization of crucible residues will be ineffective and represents a high programmatic risk.

2. Three technologies have been proposed for the crucible stabilization.

   a. Aqueous acid dissolution technology has been demonstrated; however, reliance on the technology is a high programmatic risk because there are no operable chloride dissolution facilities at RFETS.

   b. Molten salt washing process is a medium programmatic risk. Feasibility studies at LANL have been promising, but more work is needed to define an operation for production.

   c. The electroscrub process has not been demonstrated for stabilization of crucible residue.

Recommendations:

1. A trade study should be conducted to confirm the problems with the RFETS baseline approach and evaluate the viability of alternative approaches.
2. R&D activities should be directed to provide data and support analyses in the trade study.

4.3.4 Combustible Residues

4.3.4.1 Introduction. Potential hazards associated with plutonium-contaminated combustibles include: (1) spontaneous combustion from nitrated combustibles or oxidizer/fuel mixtures, (2) pressure build-up from gas generation due to radiolysis, (3) corrosion due to hydrochloric acid (HCl) generated from radiolysis of chlorocarbons, and (4) the exothermic reaction of plutonium metal and chlorocarbons. Such combustibles include cartridge filter elements, high-efficiency particulate air (HEPA) filters, graphite, rubber (leaded and nonleaded gloves), ion exchange resins, plastics, and cellulose materials (e.g., paper, wipes). Technology requirements for treating the diverse inventories and generating stable waste and/or long-term storage forms include one or more of the following: pretreatment/size reduction, nitrate removal, destruction of the organic matrix, actinide removal, and chlorinated hydrocarbon removal.

Combustible residue stabilization need data, driven by 94-1 Implementation Plan milestones, are shown in Table 4.3.4-1. The polycubes stabilization need date is supported by current R&D activities. Support of the need date for stabilizing high risk combustibles at RFETS is influenced by the outcome of the Trade Study discussed below.

Table 4.3.4-1. Driving Implementation Plan Milestone Requirements for Combustible Stabilization *

NMSTG Milestone Number	R&D Need Date	NMSTG Milestone Due Date	DOE Site	Milestone Text
IP-3.3-017	Nov 1997	Nov 1998	RFETS	Stabilize high risk combustibles (11,000kgs).
IP-3.3-028	Jul 1999	Jul 1999	HAN	Stabilization of polycubes begins.

* Refer to 4.1.1 for description of table headings.

4.3.4.1.1 Results of Trade Study—The Combustibles Trade Study has recently been completed to evaluate incineration and several alternatives to incineration. The trade study suggests that a number of technologies show advantages over the baseline processes being conducted at RFETS. However, a key finding indicates that these technologies, including the baseline, are relatively immature for plutonium contaminated materials and require either R&D or substantial development and testing prior to implementation. Incineration cannot support completing stabilization prior to May 2002 due to the permitting timeline; therefore, it is not recommended by the trade study as a viable stabilization option. Mediated electrochemical oxidation (MEO) is the preferred alternative for stabilization and clearly warrants an accelerated development program. Cryogenic size reduction is the preferred alternative for pre-treatment. The recommended backup technologies are catalyzed chemical oxidation (CCO), mixed acid remediation, and sonication wash/dry. Pyrolysis (except for polycubes) and hydrothermal stabilization were eliminated by the trade study from further consideration.

4.3.4.2 Description and Status for Combustibles Technologies. The primary objectives of the R&D efforts in FY96 were to provide preliminary data for the Combustibles Trade Study and to increase the technical maturity of candidate technologies. The technologies under development are discussed in the following sections.

4.3.4.2.1 Pyrolysis of Polycubes (LANL/Hanford/SAIC)—About 250 polycube containers containing 6kg of plutonium currently reside at Hanford. The stabilization of polycubes is being explored using pyrolysis to destroy the styrene matrix and stabilize the plutonium. Pyrolysis is a pyrochemical technique using a high-temperature inert environment to break down and volatilize polymeric materials, leaving the oxides as free-flowing powders that can be calcined to make them suitable for long-term storage.

An existing furnace that has been successfully used to destroy polycubes is being studied to determine optimum operating parameters and to evaluate downstream technologies to treat the pyrolysis off-gas, allowing it to operate in compliance with environmental regulations. Preliminary tests have shown that pyrolysis allows for nearly 100% removal of polystyrene without forming tars or other constituents that could adversely affect its operation. Three approaches to off-gas treatment are being considered, each designed to oxidize the organic constituents to carbon dioxide and water—catalytic conversion, secondary combustion, and silent discharge plasma.

In FY96, most of the off-gas technology development was completed in support of the decision on off-gas treatment designs in early FY97 and the delivery of treatment equipment to Hanford shortly thereafter.

4.3.4.2.2 Pyrolysis of Other Combustibles (LANL)—Pyrolysis is also being developed to address requirements to stabilize polymeric materials (e.g., cellulose and plastics) commonly generated during nuclear materials applications. The pyrolysis system designed in this effort will destroy the matrix of the polymer, allowing its decomposition products to be removed as a gas. The remaining material containing the radionuclides will be reduced significantly in both mass and volume, and will be in a form suitable for long-term storage, immobilization, or subsequent actinide recovery. The pyrolysis system designed in this effort is being tailored specifically for the glovebox environment. It will be designed to minimize the formation of organic liquids, employ downstream technologies to treat off-gas products, and operate in compliance with environmental regulations. Preliminary tests of nonradioactive materials have shown 80%-100% reduction of mass of selected polymeric materials.

A pyrolysis system is being designed to accommodate a comprehensive array of cellulosic and plastic materials. Design and fabrication efforts were completed in FY96; installation and initial operation will be accomplished in FY97 with DOE-Defense Programs (DOE-DP) funding.

4.3.4.2.3 Catalyzed Chemical Oxidation (RFETS/LANL)—Catalyzed chemical oxidation, developed by Delphi Research, Inc., has been demonstrated to be capable of effectively destroying combustible residues. The process is sufficiently aggressive to oxidize hard-to destroy materials, such as polyethylene and PVC. Surrogate materials have been processed in a 4-liter reactor. Full-scale demonstration with plutonium-contaminated materials is now needed.

Efforts in FY96 included configuring the glovebox system and evaluating the issues associated with disposing of the stabilized products. In FY97, the equipment will be installed and tested in the plutonium facility at LANL or one of the facilities at RFETS.

4.3.4.2.4 Mixed Acid (Nitric/Phosphoric) Remediation and Stabilization (SRS)—This technology is a non-incineration process capable of destroying organic compounds, including plastics and resins, at moderate temperatures and pressures. The technology uses a mixture of an oxidant (nitric acid) and a carrier acid (phosphoric acid). When a metal-contaminated organic material is destroyed, the metals are also dissolved. This final liquid then can be converted to a stable waste form in a single processing step.

The core technologies associated with this process have been demonstrated successfully in the laboratory and are commercially available. Many different organic compounds have been oxidized. Those materials completely oxidized at atmospheric pressure below 180C include neoprene, cellulose, ethylene diaminetetraacetic acid (EDTA), and nitromethane. Polystyrene ion exchange resins have been destroyed effectively at 175C and 5-10 psig. More stable compounds, such as polyethylene and oils, have been decomposed completely below 200C and 10-15 psig. These rates are dependent on operating conditions and can be accelerated by increasing the reaction temperature and pressure.

Recent work has involved the oxidation of the types of ion exchange resins used by the nuclear power industry to deionize water. Studies with radioactively-contaminated resin have exhibited a 2X and 6X volume reduction and demonstrated immobilization of the metal components within a glass matrix. Work is in progress to build and test a small pilot system for FY97. A parallel effort continues toward designing and cost estimating the fabrication of a large pilot system.

4.3.4.2.5 Hydrothermal Treatment (LANL)—Hydrothermal oxidation involves the use of high-temperature, high-pressure hydrogen peroxide as an oxidant to destroy organics. Under this process, organics are converted to carbon dioxide, nitrates to nitrogen, and sulfur to sulfate. The processing equipment used for this technology is compact, operating at about 550C and at pressures of 10,000 psi. The advantages of this system include fast reaction (seconds), small reactor, and complete reaction.

A pilot scale unit has been installed in the Plutonium Facility at LANL. Processing development to date has been with nonradioactive materials. In FY97, treatment studies funded by DOE-DP on various combustible residues will be conducted in the pilot equipment, followed by design and testing of a full-scale system.

4.3.4.2.6 Mediated Electrochemical Oxidation (LANL/LLNL)—MEO uses electrochemically-generated silver (Ag II) as an oxidizing agent for plutonium oxide and organic materials. Extremely high destruction efficiencies for certain organics have been achieved in full-scale tests using an industrial-sized electrochemical cell.

It has been demonstrated that MEO can be used to "leach" the plutonium from the combustible matrix. Chlorinated hydrocarbons are efficiently converted to carbon dioxide at room temperature; however, other materials, such as polypropylene and polyethylene, are fairly inert to MEO treatment. Thus, it would be an ideal technology for treating residues like full-flow filters that are contaminated with carbon tetrachloride and plutonium. The solvent would be converted to carbon dioxide and HCl, the plutonium dissolved and recovered, and the filter rendered free of significant organic or radioactive contamination and suitable for compaction and disposal.

Some combustible materials, such as cheese cloth, paper, and cardboard, are not inert to MEO treatment. The MEO process results in matrix destruction, resulting in a stabilized produce.

Both bench-scale and pilot-scale capabilities are already in place in a nonradioactive laboratory environment. A pilot-scale apparatus has been installed in the LANL Plutonium Facility. Treatability studies were initiated in FY96 and will continue into FY97 to support implementation as the baseline process at RFETS.

4.3.4.2.7 Sonication Wash/Dry (RFETS/LANL)—Washing is not a matrix destruction technology, but it does provide a potentially simple solution to the four storage concerns—oxidizer/fuel mixtures, hydrogen generation, reactive plutonium metal, and corrosion. In addition, washing could meet Resource Conservation and Recovery Act (RCRA) extraction criteria for debris treatment and could result in TRUW instead of transuranic mixed waste, benefiting disposal or additional waste treatment.

Washing has been used for nitrate removal from residues at most DOE sites. Industrial detergents require evaluation and testing for specific treatment applications. In addition, sonication during an aqueous wash physically removes plutonium particles and may allow up to 90% plutonium removal from the matrix by simple gravity separation and filtration.

In FY96, bench-scale experiments using surrogate materials were conducted; radioactive experiments will be conducted in FY97.

4.3.4.2.8 Ion Exchange Resin Denitration (RFETS/LANL)—Nineteen drums of ion exchange (IX) resin containing substantial quantities of plutonium currently reside at RFETS. Salicylate has a high affinity for the anion exchange site on exchange resins and can displace the nitrate ion. Exchangeable nitrate and plutonium can be readily removed with a solution containing salicylate.

The development of this approach has been completed; however, the current RFETS plan is to add the resin a little at a time to cement solutions.

4.3.4.2.9 Cryogenic Size Reduction (LANL)—Pretreatment or size reduction is necessary for many categories of combustibles before matrix destruction or actinide removal technologies are used. In particular, size reduction can enhance or enable pyrolysis, hydrothermal processing, catalyzed chemical oxidation, aqueous washing, and electrochemical oxidation.

Coupling cryogens that embrittle many combustible materials with size reduction systems produces desired particle size distributions and robust operations. Other benefits include increased worker safety (as compared to workers using knives or hacksaws), exposing trapped nuclear materials from combustible forms, and making handling of materials easier.

Cryogenic-based size reduction has been tested fully on several combustible matrices. Various cryogenic system applications are being optimized, including crushing polycubes, grinding filters, and shredding soft matrix materials. FY97 effort will continue with glovebox installations and demonstrations on combustibles with radioactive contamination.

4.3.4.3 Newly-proposed R&D Activities/Technologies. Steam reforming has been proposed as an alternative technology by the Scientific Ecology Group (SEG) and warrants further analysis through a demonstration phase with plutonium-contaminated materials. A key element of this analysis is determining the plutonium material balance in the system, which has yet to be established.

4.3.4.4 Evaluation of Current and Proposed R&D Against Requirements. The technologies discussed in Section 4.3.4.2 and 4.3.4.3 provide an adequate technology base to meet the stabilization requirements identified in Table 4.3.4-1. No additional R&D requirements have been identified at this time.

4.3.4.5 Conclusions and Recommendations. By referring to Table 4.3.4-2, the following conclusion is made:

Conclusions:

1. Polycube pyrolysis represents a low programmatic risk for meeting its R&D need date. The technical maturity scores for the technologies proposed to meet IP-3.3-017 (stabilization of high risk combustibles at RFETS) all have maturity scores that indicate a medium to high risk of achieving an R&D need date of November 1997, in support of a November 1998 stabilization completion date. Technology development efforts require special attention, tracking, and support by the NMSTG.

2. The recommended baseline technology for combustible treatment at RFETS is MEO (leach) and is a medium risk item.

3. The baseline for pretreatment at RFETS is cryogenic crushing and is high risk. Research at LANL shows promising results, but there is inadequate progress on technology transfer and operational readiness at RFETS.

Recommendations:

1. The risk rating for stabilizing combustibles at RFETS requires commitment by DOE headquarters and the RFETS field office to develop MEO and cryogenic crushing processes recommended by the trade study.

2. Gas generation rate data are required on dry combustibles in order to quantify the cost benefit of matrix destruction technologies. RFETS is currently conducting such experiments and is encouraged to continue toward the completion of this study in early FY97.

Table 4.3.4-2. Summary Technology Status Combustible Stabilization **

R&D Program	R&D Site	End Use Site	94-1 Milestone	R&D Need Date	Recommendation	Technical Maturity
4.3.4.2.1 Polycube Pyrolysis	LANL	HAN	IP-3.3-028	Jul 1999	Baseline	4.40
4.3.4.2.2 Pyrolysis	LANL	RFETS	IP-3.3-017	Nov 1997	Backup	6.19
4.3.4.2.3 Catalyzed Chemical Oxidation	RFETS/ LANL/ SRS/ LLNL	RFETS	IP-3.3-017	Nov 1997	Backup	6.00
4.3.4.2.4 Nitric/Phosphoric Digestion	SRS	RFETS	IP-3.3-017	Nov 1997	Backup	5.79
4.3.4.2.5 Hydrothermal	LANL	RFETS	IP-3.3-017	Nov 1997	Backup	6.05
4.3.4.2.6 Mediated Electrochemical (Leach)	LLNL/ LANL	RFETS	IP-3.3-017	Nov 1997	Baseline	4.53
4.3.4.2.6 Mediated Electrochemical (Destruction)	LANL	RFETS	IP-3.3-017	Nov 1997	Backup	6.14
4.3.4.2.7 Sonication Wash/Dry	RFETS/ LANL	RFETS	IP-3.3-017	Nov 1997	Backup	6.91
4.3.4.2.8 IX Resin Denitration	RFETS/ LANL	RFETS	IP-3.3-017	Nov 1997	Backup	3.44
4.3.4.2.9 Cryogenic Size Reduction	RFETS/ LANL	RFETS	IP-3.3-017	Nov 1997	Baseline	5.93
4.3.4.3 Steam Reformation	SEG	RFETS	IP-3.3-017	Nov 1997	Backup	6.21

** Refer to 4.1.5 for description of table headings.

4.3.5 Ash

4.3.5.1 Introduction. Approximately 20 metric tons of incinerator ash residues from various plutonium operations exist within the DOE weapons Complex. Most of this residue is stored at RFETS. One metric ton is stored at Hanford and much smaller quantities are at LLNL, LANL, and SRS. These ash residues are generally considered unacceptable (high risk) for safe interim storage, as specified in DNFSB Recommendation 94-1. Potential hazards associated with ash include corrosion of containers and gas generation from radiolysis. In the 94-1 Site Remediation Plan, RFETS committed to stabilize all of its ash before May 2002.

Ash stabilization R&D need data, driven by 94-1 Implementation Plan milestones, are shown in Table 4.3.5-1. The need date for ash residues at LLNL should be addressed by current R&D activities; however, ash stabilization at Hanford is uncertain pending a technology decision by DOE-RL. Meeting the R&D need date for RFETS ash residues is of concern since baseline and backup technologies are somewhat immature.

Table 4.3.5-1. Driving Implementation Plan Milestone Requirements for Stabilization of Ash *

NMSTG Milestone Number	R&D Need Date	NMSTG Milestone Due Date	DOE Site	Milestone Text
IP-3.3-027	Apr 1998	Jan 2002	HAN	Stabilization and repackaging of interim-stabilized materials completed.
IP-3.3-041	Mar 1997	Apr 1998	LLNL	Stabilize and package all containers of ash/residue.
IP-ES-025	Feb 1998	May 2002	RFETS	Repackage all plutonium inorganic oxides and wet/miscellaneous residues (1,113 drums).

* Refer to 4.1.1 for description of table headings.

4.3.5.1.1 Results of Plutonium Ash Residues Trade Study—The Plutonium Ash Residues End-States Trade Study evaluated numerous options for treating/stabilizing plutonium-bearing ash residues. Four main options were examined—*no action; separate and store; treat, stabilize, and store/dispose; and repackage and store/dispose*—each consisting of several approaches. For Hanford, LLNL, and LANL, the suggested path forward is to separate and store the plutonium per DOE-STD-3013 long-term storage criteria. Each site would use a separation technology that is peculiar to the ash at that site and to the processing systems available. Hanford would use silver persulfate dissolution, LLNL would use a mild acid wash followed by calcination, and LANL would use standard nitrate or chloride dissolution and recovery technology. For RFETS, the path forward is not as simple. The most cost effective and preferred option for RFETS ash was vitrification of all the residues at 5 wt% plutonium for disposal at WIPP. The second option is to vitrify the ash at 5 wt% for HLW repository disposal using a can-in-canister developed by SRS. The third option is to dilute the ash to 2 wt% and ceramify it for disposition to WIPP. The fourth option is to separate the plutonium from the ash. This would require RFETS to install and operate an aqueous plutonium recovery line or SRS to start up the New Special Recovery (NSR) Facility.

4.3.5.2 Description and Status for Ash Technologies.

4.3.5.2.1 Pretreatment/Thermal Stabilization (LLNL)—This year, LLNL created three small plutonium ash batches (100 to 130 g each) in recovery operations. Plutonium metal was removed from the contaminated portion by dissolving in concentrated HCl, then precipitated by adding concentrated NaOH. The precipitate was filtered and calcined at about 500C for several hours; LOI at 500C was less than 1%. This is the same procedure used in the past to generate most of LLNL ash material. To test the effectiveness of washing, the calcined ash was rinsed with 1 liter of deionized water. The remaining solids were filtered and the remnant calcined again at about 500°C for several hours. LOI at 500C was less than 1%. From these preliminary wash tests, it was found that between 60 and 70% of the material was soluble. As a result, a simple wash process appears to be adequate to upgrade LLNL ash material from an average of 24% to approximately 70% plutonium.

4.3.5.2.2 Silver Persulfate (Hanford)—Hanford has a considerable inventory of incinerator ash generated from onsite and off site processing. Approximately 1530 containers of ash representing 80.4kg of plutonium are in storage awaiting disposition. The Ash Residues End-States Trade study concluded that silver persulfate dissolution was the preferred option for Hanford ash. Silver persulfate dissolution is similar to the MEO process described in Section 4.3.5.3.2, where silver (II) is used to oxidize the plutonium in the ash. The persulfate process simultaneously converts the dissolved plutonium to plutonium sulfate, which is easily converted to plutonium oxide for storage; the silver is not recovered. The MEO process differs in that the solubilized plutonium is recovered by ion exchange, precipitation, and calcination to the oxide; and silver is recovered for reoxidation to silver (II) and recycle to the dissolver. The selection of which of these two processes is to be employed at the site was left to the discretion of the DOE-RL Field Office.

The use of a glovebox-sized silver persulfate dissolution process or an existing cementing capability is dependent upon Field Office direction. There is ample time for this decision to be made and the processing to be completed before the milestone IP-3.3-027 due date of Jan 2002.

4.3.5.2.3 Vitrify Ash for Can-in-Canister Disposition (SRS)—An R&D program to demonstrate the vitrification of RFETS incinerator ash is currently in progress at SRS. Initially, surrogate ash representing a range of potential compositions is being used to demonstrate the borosilicate glass formulation used in the Ash Trade Study's vitrification material balances. A demonstration using RFETS ash has been delayed until calculations are accepted by the South Carolina Department of Health and Environmental Control showing that the offgas from vitrification will not generate a downstream secondary mixed waste. Once this issue is resolved, demonstrations with the RFETS ash using the borosilicate glass and a new soda-lime silica glass formulation are planned. Initial surrogate experiments focused on vitrifying the ash without pretreatment. Future experiments will investigate the merit of a dilute nitric acid wash to remove the alkali and alkaline-earth elements, which may allow higher ash loadings in the glass.

In addition to the vitrification demonstration, experiments are also being performed to determine if the incinerator ash can be dissolved in an alkali flux to create a mineral product that is soluble in nitric acid. Currently, sodium and potassium oxides in various combinations with surrogate ash (thorium is the surrogate for plutonium) are being used to investigate the feasibility of this strategy. Experiments have demonstrated that surrogate ash can be dissolved in the sodium/potassium oxide at approximately 1000C. However, analysis of the resulting minerals by x-ray diffraction has yet to show the formation of a sodium/potassium/thorium oxide. There is some evidence that the thorium may form an oxide mineral in combination with potassium and aluminum. Future experiments will investigate this possibility and the potential recoverability of the thorium by nitric acid dissolution. If the surrogate experiments are successful, demonstrations using the RFETS ash will be performed.

4.3.5.2.4 Ceramify Ash for WIPP Disposal (RFETS)—Ceramification offers a dramatic reduction in processing and waste generation and combines the efforts required to stabilize plutonium-bearing solutions and oxide generated by the corrosion of plutonium metal and its alloys. Ceramification is a process whereby a plutonium nitrate solution is infused into a suitable substrate to saturation. The saturated mass is then fed to a muffle furnace and heated to 500C. During the heating, the solution is denitrated, converted to hydroxide, and finally incorporated into the ceramic structure of the substrate and resulting oxide. These two steps are repeated until the porosity of the resultant mass is reduced to the required level. The product would be suitable for disposal at WIPP.

4.3.5.3 Newly-proposed R&D Activities/Technologies.

4.3.5.3.1 Vitrification for Ash Disposal at WIPP (RFETS)—The objective of this activity is to develop and demonstrate a vitrification technology adequate for treating the current range of ash residue compositions to a form suitable for disposal at WIPP. The technology will be as simple and robust as possible to avoid the need for excessive characterization of the ash input and the need for detailed manipulation of the frit additive for the changing ash input stream. Two tasks need to be performed: first, the development of a semi-empirical model for selecting frit compositions; and second, glass fabrication work on real and simulated ash compositions. The fabrication work will be used to examine both the process of making glass from ash residues and the adequacy of the semi-empirical model. It will likely be necessary to synthesize and test glasses in compositional ranges not previously investigated to ensure the adequacy of this model. The frit model will then be combined with the rest of the vitrification process (pretreatment, calcination, pouring) to synthesize glasses from real ashes to demonstrate the method. The method will then be scaled up using a 3.5 liter tilt-pour furnace.

The major advantage of this process is the decrease in volume compared to other stabilization processes, such as ceramification. Also, it is possible to vitrify plutonium oxides and evaporate and denitrate plutonium nitrate solutions simultaneously.

4.3.5.3.2 Mediated Electrochemical Oxidation for Ash (SRS)—SRS proposes to integrate the MEO technology into the existing NSR Facility to process incinerator ash and ash heels. The resulting plutonium solution will then be processed in F-Canyon to yield a plutonium product that is compatible with DOE-STD-3013 for the long-term storage of plutonium metal and oxide. The MEO technology has been demonstrated as an effective method for recovering plutonium from high-fired incinerator ash similar to the ash and ash heel residues stored at RFETS. Silver (II) ions will be used as the oxidizing species. The process rate is expected to be about 3kg of residue per hour at 200 amps. SRS has modified the process to eliminate the secondary catholyte waste associated with silver (II) oxidation; a common electrolyte system uses the same solution to satisfy both the anode and cathode reactions. Extensive development of the MEO prototype system using a plutonium surrogate has been done at LLNL. Successful implementation of this ash treatment strategy will require both cold and hot demonstrations of the prototype system at SRS.

4.3.5.3.3 Can-in-Canister (SRS)—This technology is described in Section 4.3.2.3.3.

4.3.5.4 Evaluation of Current and Proposed R&D Against Requirements. The technologies discussed in Section 4.3.5.2 and 4.3.5.3 provide an adequate technology base to meet stabilization requirements identified in Table 4.3.5-1. No additional R&D requirements have been identified at this time.

4.3.5.5 Conclusions and Recommendations. By referring to Table 4.3.5.-2, the following conclusions are made:

Conclusions:

1. The pretreatment/thermal stabilization baseline at LLNL represents low programmatic risk for meeting the R&D need date.
2. The baselines of silver persulfate dissolution at Hanford and vitrification at RFETS for WIPP disposal represent medium programmatic risk for meeting their R&D need dates.

Recommendations:

1. NMSTG should arrange for adequate tracking of the technical and programmatic progress of silver persulfate dissolution and vitrification for disposal at WIPP to ensure that need dates will be met.
2. The resolution of which path to pursue for RFETS ash requires that RFETS and DOE resolve issues regarding WIPP limits, aqueous plutonium separation operations at RFETS, or shipment of RFETS ash to others sites for treatment. However, technology development need to continue concurrently with the decision process, at least until the path forward is chosen; otherwise, the process will not be ready to implement. With regard to the other sites, the continued support of development activities at Hanford and LLNL will provide the technology necessary to complete the stabilization and disposition of the ash residues at these sites.

Table 4.3.5-2. Summary Technology Status for Stabilization of Ash **

R&D Program	R&D Site	End Use Site	94-1 Milestone	R&D Need Date	Recommendation	Technical Maturity
4.3.5.2.1 Pretreatment/ Thermal Stabilization	LLNL	LLNL	IP-3.3-041	Mar 1997	Baseline	0.70
4.3.5.2.2 Silver Persulfate Dissolution	HAN	HAN	IP-3.3-027	Apr 1998	Baseline	4.91
4.3.5.2.3 Vitrify in MPPF	SRS	SRS	IP-ES-025	Feb 1998	Backup	5.60
4.3.5.2.5 Vitrify at RFETS	RFETS	RFETS	IP-ES-025	Feb 1998	Backup	7.58
4.3.5.2.4 Ceramify for WIPP	RFETS	RFETS	IP-ES-025	Feb 1998	Backup	7.49
4.3.5.3.1 Microwave for WIPP	RFETS	RFETS	IP-ES-025	Feb 1998	Baseline	4.44
4.3.5.3.2 MEO in NSR	SRS	RFETS Material at SRS	IP-ES-025	Feb 1998	Backup	4.00
4.3.5.3.3 Can-in-Canister	SRS	SRS	IP-ES-025	Feb 1998	Backup	2.81

** Refer to 4.1.5 for description of table headings.

4.3.6 Metal Alloys and Intermetallic Compounds

4.3.6.1 Introduction. Metal alloys and intermetallic compounds refer to the scrub alloy created at RFETS during the 1980's, anode heels, and a few unique alloy formulations. The bulk of the scrub alloy buttons were composed of about 70 wt% aluminum and 30 wt% plutonium. The scrub alloy contained plutonium and americium recovered from MSE salt residues. The scrub alloy was shipped to SRS for further processing to produce purified plutonium metal (F-Canyon and FB-Line process flow sheets). The purified plutonium was returned to RFETS; the americium was not recovered, but was transferred to the SRS HLW tanks. When plutonium operations were curtailed at RFETS in 1990, about 300 scrub alloy items were in storage awaiting shipment to SRS. In addition to the aluminum/magnesium scrub alloy at RFETS, there are some test buttons from experimental programs at RFETS and at LLNL. Those test buttons are calcium/gallium alloy and were generated as part of a RFETS development program to implement a new scrub process to minimize waste and radiation exposure.

In 1995, the NMSTG directed that a Scrub Alloy Trade Study Team be formed to determine the best option for stabilizing the scrub alloy items. The Study Team reviewed several options for scrub alloy stabilization, including: No Action, Ship-As-Waste to the WIPP, Repackage and Store at RFETS, Calcine-to-an-Oxide, and Ship-to-SRS. Based on the consideration of several performance measures, the Scrub Alloy Trade Study concluded that the Ship-to-SRS option should be implemented. This option requires that the scrub alloy be repackaged for shipment to SRS, shipped to SRS, processed in F-Canyon and FB-Line, and the resulting metal stored in accordance DOE-STD-3013 for the long-term storage of plutonium metal and oxides. If the SRS option is not available, the backup option is to package the material for storage at RFETS.

4.3.6.2 Description and Status for Metal Alloys and Intermetallic Technologies.

4.3.6.2.1 Scrub Alloy Processing—The standard dissolution/purification flow sheet for F-Canyon and FB-Line will be used to process the scrub alloy.

4.3.6.3 Newly-proposed R&D Activities/Technologies.

4.3.6.3.1 Purification by Impurity Chlorination (LLNL)—Livermore has demonstrated a non-aqueous process that will separate the plutonium in anode heels from the salt and other materials. The process produces a purified plutonium metal that can be stored in long-term storage containers and a waste salt that will meet WIPP disposal criteria. The process is known as Purification by Impurity Chlorination (PIC) and consists of the following three unit operation: (1) the anode heel is hydrided, converting the metal to a hydride powder; (2) the powdered anode heel is chlorinated at about 400C, vaporizing most impurities and converting the plutonium to chloride; and (3) the product chloride is reduced to metal, resulting in a metal/salt separation.

The PIC process has undergone a limited feasibility demonstration in which several tests were performed on impure plutonium metal alloys, including one test with an anode heel. Additional development is needed to establish the overall operational envelope. All the equipment needed to perform this process exist at LLNL. Once the operational envelope is established, the equipment at LLNL could be used to process the anode heels being stored at RFETS.

4.3.6.4 Evaluation of Current and Proposed R&D Against Requirements. The technologies described in Section 4.3.6.2 and 4.3.6.3 provide an adequate technology base to meet stabilization requirements. No additional R&D requirements have been identified at this time.

4.3.6.5 Conclusions and Recommendations. By referring to Table 4.3.6-1 the following conclusions are made:

Conclusions:

1. Scrub alloy processing and impurity chlorination both represent low programmatic risk and should be adequate to support the R&D need date, which is proposed by SRS as part of the canyon utilization recommendation to DOE.

2. No R&D is required on the magnesium/aluminum alloy in storage at RFETS.

3. A scrub alloy technique has been demonstrated at RFETS for processing anode heels into a stable storage form. However, this technology would require RFETS to perform the alloy scrub and ship the alloy to SRS for aqueous processing. The PIC technology being developed at LLNL would simplify processing and generate less waste.

Recommendations:

1. Magnesium/Aluminum alloy in storage at RFETS should be sent to SRS and processed in F-Canyon and FB-Line.

2. The LLNL calcium/gallium scrub alloy should be sent to SRS for testing to determine if this alloy is compatible with F-Canyon operations.

3. The RFETS calcium/gallium scrub alloy should be sent to SRS for processing in F-Canyon upon successfully completing the compatibility tests at SRS.

Table 4.3.6-1. Summary Technology Status for Scrub Alloy Processing *

R&D Program	R&D Site	End Use Site	94-1 Milestone	R&D Need Date	Recommendation	Technical Maturity
4.3.6.2.1 Scrub Alloy Processing	SRS	RFETS Material at SRS	N/A	July 1997	Baseline	1.07
4.3.6.3.1 Impurity Chlorination	LLNL	RFETS	N/A	July 1997	Backup	1.81

* Refer to 4.1.5 for description of table headings.

4.3.7 Miscellaneous Plutonium Residues

4.3.7.1 Introduction. Small quantities of miscellaneous residue items are currently in storage throughout the DOE Complex. Miscellaneous plutonium residues include insulation, ceramics, contaminated scrap metals, fire brick, and LECO crucibles. Although combustible constituents are usually not present, these solids may have been packaged in plastic.

In addition, there are a number of actinide stabilization issues that were not considered by the DNFSB Recommendation 94-1. The materials of interest are various neutron sources, analytical actinide standards or sources, and small quantities of R&D materials that are located at several DOE, contractor, or university sites. The Small Sites/Small Holdings committee will review the materials and determine whether or not a path forward exists for disposing of the material. Where no path exists, the committee will formulate a remediation plan that includes a possible technical partnership with a larger site for implementation.

4.3.7.2 Description and Status for Miscellaneous Plutonium Residue Technologies. There are no technologies currently under development for miscellaneous plutonium residues.

4.3.7.3 Newly-Proposed R&D Activities/Technologies. The multi-purpose process technologies described in Section 4.3.8 would be applicable to miscellaneous plutonium residues.

4.3.7.4 Evaluation of Current and Proposed R&D Against Requirements. A comprehensive assessment of the residue inventories is needed to determine if there are miscellaneous inventories that cannot be treated by technologies being developed for the major inventory items discussed in Section 4.3.1 through 4.3.6 or by straight forward repackaging and disposal. If any residues are found not to have a path forward, there may be a compelling need to develop new stabilization approaches.

4.3.7.5 Conclusions and Recommendations.

Conclusions:

1. There may be miscellaneous residue items with no identified path forward.
2. The multi-purpose process discussed in Section 4.3.8 may play a role in stabilizing any miscellaneous residue items.

Recommendations:

An inventory assessment should be completed to determine if new approaches are needed for any miscellaneous residues.

4.3.8 Multi-Purpose Processes

4.3.8.1 Introduction. Multi-purpose processes have the capability to stabilize more than one type of plutonium scrap and residue and to avoid the need to develop, deploy, operate, and decommission multiple separate processes. This can reduce costs, worker radiation exposures, and schedule. Such processes are particularly useful for addressing miscellaneous plutonium-bearing residues where there are small quantities of many different types of materials and where development of separate processing technologies for each form may become prohibitively expensive. Such technologies also create the long-term capability to process future waste, scrap, and residue streams with undefined compositions.

Better definition of requirements have identified two categories of materials that may require general purpose processes. The first category consists of miscellaneous plutonium-bearing residues, where small quantities exist of many different materials. The second category is a general purpose backup option in the event any of the mainline treatment options fail.

4.3.8.2 Description and Status of Multi-Purpose Process Technologies.

4.3.8.2.1 F-Canyon Dissolution (SRS)—A general purpose process that involves the use of F-Canyon to treat materials from other sites is under consideration. The F-Canyon dissolution process is designed to dissolve a variety of plutonium scrap and residue forms, with purification and conversion to plutonium metal. F-Canyon could be used for feed materials compatible with the dissolver, including oxides and metals. The facility can not process salts and certain other specific residues.

Materials selected for possible treatment operations in F-Canyon will require samples for R&D studies at SRS to determine if the product materials are compatible with F-Canyon dissolution operations.

4.3.8.3 Newly Proposed R&D Activities/Technologies.

4.3.8.3.1 Glass Material Oxidation and Dissolution System (GMODS)—GMODS is designed to directly convert plutonium scrap and residue in any chemical form into a high-quality plutonium glass suitable for long-term storage or disposal. The GMODS process (1) converts metals, ceramics, and amorphous solids to glass; (2) oxidizes organics to carbon oxides, with the noncombustibles residues converted to glass; and (3) converts halides to a low-halide glass and a secondary clean sodium-halide stream.

Feed materials are directly fed to a glass melter to form a special, molten, lead-borate, dissolution glass. Metals and organics are oxidized by lead oxide in the molten glass with metal oxides dissolving into the glass, carbon dioxide exiting the melter via the off-gas system, and byproduct lead metal separating from the glass. Halides fed to the melter form volatile lead halides that enter an aqueous off-gas scrubber, forming soluble halide salts (e.g., sodium chloride) and insoluble lead hydroxides. Lead hydroxides from the offgas system are recycled to the melter in the subsequent feed batch. Byproduct lead is also reoxidized and recycled to the melter in the subsequent feed batch. Glass forming components are added after feed dissolution to form a high-quality borosilicate glass product. If desired, the borosilicate glass can be lead-free glass by adding carbon during final processing to remove any lead oxide from the glass.

Experiments to date have converted uranium and cerium (plutonium surrogates), stainless steel, aluminum, various oxides, and other materials to glass; oxidized carbon; and separated chlorides into a separate stream. Although equipment options have been identified, significant development effort is required.

4.3.8.3.2 Enhancing Cemented Waste Forms (LANL)—Supercritical carbon dioxide treatment decreases the porosity and permeability of cements while increasing the hardness and transforming the cement into a geologically stable calcium carbonate. The process removes water that is not chemically bound and constituents that will leach out in a disposal facility. Removal of the organics eliminates the potential classification of the waste as RCRA materials due to the presence of the organics. For residues that are treated for disposal by cementation, this process can significantly enhance the integrity of the waste form and reduce hydrogen generation rates that are constrained by shipping regulations.

The process has been successfully scaled up to 6" by 12" cement cylinders in existing equipment to treat nonradioactive inventories. It is proposed to evaluate the capabilities of this technology using plutonium residues that are cemented for disposal.

4.3.8.3.3 Plasma Hearth Process and Plasma Arc Process (ANL, et al.)—The plasma hearth process is designed to heat scrap and residues to very high temperatures (significantly above their melting points) to consolidate the materials as a glass/ceramic mixture. The process uses a high-temperature plasma arc to melt all components in the feed stream. Oxygen can also be added during processing to oxidize feed materials; several equipment configurations are possible. The plasma arc process is being developed to convert transuranic waste on a large scale; however, the process may not handle salts and certain other feeds where volatile plutonium compounds are formed.

4.3.8.4 Evaluation of Current and Proposed R&D Against Requirements. Requirements need to be further defined to determine if any miscellaneous inventories have no path forward (see Section 4.3.7.4).

4.3.8.5 Conclusions and Recommendations.

Conclusions:

There may be miscellaneous residue items with no path forward.

Recommendations:

An inventory assessment should be completed to determine if multi-purpose processes are needed for any miscellaneous residues.

5. SPECIAL ISOTOPES STABILIZATION

5.1 Introduction

Special isotope R&D need dates, driven by 94-1 Implementation Plan milestones, are shown in Table 5-1. Special isotopes of Am/Cm and neptunium in solution at SRS will be stabilized, shipped to the Oak Ridge National Laboratory (ORNL), and stored. Am/Cm mixtures contain other actinides, including plutonium. Pu-238 and Pu-242 are handled separately and are stored at SRS and LANL.

Need dates are presently supported by existing R&D activities. However, recent melter problems may impact the NMSTG milestones for stabilizing Am/Cm solutions.

Table 5-1. Driving Implementation Plan Milestone Requirements for Stabilization of Special Isotopes **

NMSTG Milestone Number	R&D Need Date	NMSTG Milestone Due Date	DOE Site	Milestone Text
IP-3.4-015*	Jun 1999	Jun 1999	SRS	Start vitrification of Am/Cm Solutions.
IP-3.4-019*	Apr 2000	Sept 2001	SRS	Begin stabilization of Np-237 Solutions HB-Line, Phase II.
IP-3.4-017	Oct 1996	Mar 1997	SRS	Begin stabilization of Pu-242 Solutions at HB-Line, Phase III.

* Proposed 94-1 IP change. ** Refer to 4.1.1 for description of table headings.

5.2 Description and Status of Special Isotope Stabilization Technologies

5.2.1 Americium/Curium Solution Stabilization (SRS)

Approximately 15,000 liters of solution containing isotopes of Am/Cm are currently stored in the F-Canyon Tank at SRS. These isotopes were recovered during plutonium-242 production campaigns in the mid and late 1970's. Vitrification will be used to stabilize the Am/Cm for eventual transport to the heavy isotope programs at ORNL. The Multi-Purpose Processing Facility (MPPF) in F-Canyon will be used for the vitrification process. Pretreatment operations will be performed in canyon vessels to separate the actinides and lanthanides from other impurities—primarily iron, aluminum, and sodium—before subsequent vitrification.

Research and development activities to support the vitrification process include development programs focused on the behavior of tank contents during pretreatment operations, glass formulation and physical property measurement, and equipment development and demonstration for the Am/Cm melter and supporting equipment. The pretreatment development program is complete. During an oxalate precipitation of the Am/Cm solution, specific volumes and settling rates of the precipitate slurry were measured. The results of these experiments are documented in WSRC-TR-96-0074 and WSRC-TR-96-0116.

Studies in support of the formulation and physical properties of the high lanthanide glass specified for the vitrification process are also complete. Process demonstration runs using prototype melters and support equipment are currently in progress at the SRS facilities Mock-up, and demonstration of the process racks that will be installed in the MPPF is scheduled for June 1997. Recent melter problems, however, may impact this schedule. The melter problems are a result of operational issues and do not reflect a need for additional R&D.

The scope of R&D activities is currently limited to items directly associated with stabilization (pretreatment and vitrification). Many issues associated with the temporary storage of the glass at SRS, transportation to ORNL, and recovery of the Am/Cm are not yet being addressed. Funding independent of the project will be requested from appropriate DOE Offices to resolve these issues and ensure successful recovery and utilization of these materials.

5.2.2 Neptunium Stabilization (SRS)

Neptunium solutions are stored in the SRS H-Canyon. These solutions are highly corrosive and have a significant penetrating gamma dose. Neptunium has a relatively high solubility in basic solutions compared with plutonium. This solubility effectively precludes transferring the neptunium solutions to the HLW tank farm since the soluble neptunium would ultimately be processed into saltstone. To meet LLW criteria, saltstone cannot accept the radiation levels that the neptunium would create.

Facilities and neptunium stabilization techniques have been assessed. Results indicate that the development of selected processes must be based on safe and economic stabilization and must begin in the near term. Processing the neptunium into an oxide or a glass in the SRS canyon is a viable option, since processing the neptunium solutions at another site would require an exemption from existing transportation regulations.

The baseline flow sheet for neptunium vitrification involves transferring the material to F-Canyon, where the solution will be concentrated to approximately 100 grams/liter. The neptunium solution will be delivered into MPPF and fed simultaneously with glass frit into a bushing melter. The solution will then be evaporated on the cold cap, the neptunium and frit will be melted, and the resulting melt will be drained into small stainless-steel cans.

5.2.3 Pu-238 and Pu-242 Process Support (SRS)

Stabilization of plutonium-238 at SRS was completed in FY96. The plutonium-242 solutions must be processed through H-Canyon and HB-Line and converted to oxide to meet the DOE-STD-3013 for long-term storage. The plutonium-242 stabilization will be completed in FY97. Since the baseline technology for stabilizing plutonium-242 is the same as for stabilizing plutonium-238, the required flow sheet development has been completed and only troubleshooting support will be required. The stabilized material will be stored in vaults at SRS until it is needed.

5.3 Newly Proposed R&D Activities/Technologies

There are no newly proposed R&D activities that need to be addressed at this time.

5.4 Evaluation of Current and Proposed R&D Against Requirements

The activities described in Section 5.2 will provide an adequate technology base for meeting the requirements outlined in Table 5-1. No additional R&D requirements have been identified at this time.

5.5 Conclusions and Recommendations

By referring to Table 5-2 below, the following conclusions are made:

Conclusions:

Special isotope stabilization technology development represents low programmatic risk with adequate time to meet the R&D need dates.

Recommendations:

Site baseline stabilization technologies should be pursued as indicated in Table 5-2..

Table 5-2. Summary Technology Status for Stabilization of Special Isotopes **

R&D Program	R&D Site	End Use Site	94-1 Milestone	R&D Need Date	Recommendation	Technical Maturity
5.2.1 Am/Cm Product Test	SRS	SRS	IP-3.4-015*	Jun 1999	Baseline	0.51
5.2.1 Am/Cm Vitrification	SRS	SRS	IP-3.4-015*	Jun 1999	Baseline	4.51
5.2.2 Neptunium Flowsheet	SRS	SRS	IP-3.4-019*	Apr 2000	Baseline	5.95
5.2.3 Pu-242 Stabilization	SRS	SRS	IP-3.4-017	Oct 1996	Baseline	0.00

* Proposed 94-1 IP change. ** Refer to 4.1.5 for description of table headings.

6. HIGH-ENRICHED URANIUM (HEU) STABILIZATION—MSRE REMEDIATION PROJECT

6.1 Introduction

The HEU stabilization activities addressed herein are necessary to meet the needs defined in DNFSB Recommendation 94-1. This excludes added activities identified by the ongoing DOE vulnerabilities studies.

Molten Salt Reactor Experiment (MSRE) stabilization R&D need dates, driven by 94-1 Implementation Plan milestones, are shown in Table 6-1. R&D for trapping uranium hexafluoride is complete. Approximately one and a half years are available to remove deposits from the charcoal bed. The time available to meet 94-1 Implementation Plan milestones regarding the removal of uranium salts is sufficient to allow a timely decision on the removal technology.

In its shutdown configuration, the MSRE at ORNL presents several potentially serious problems. The reactor fuel, a mixture of beryllium and lithium fluoride salts, uranium fluorides, and fission products, was a liquid at high-temperature reactor operating conditions. Today it is a solid stored in drain tanks. Uranium-233 hexafluoride ($^{233}UF_6$) and fluorine (F_2) generated by radiolysis in the drain tanks are migrating to the off-gas system, which contains a charcoal bed of unsafe geometry. This poses a potential criticality problem as the uranium builds up.

The MSRE Remediation Project will mitigate or correct problems that now exist at the MSRE. This includes: (1) trapping $^{233}UF_6$ and F_2 and removing them from the off-gas system; (2) removing uranium and carbon fluorine compounds from a charcoal bed; (3) removing or stabilizing the bulk of the fuel salt, with likely separation of the uranium from the salt matrix followed by stabilization of the salt for disposal; and (4) converting the recovered uranium to triuranium oxide for storage. The stabilization of salts containing the uranium includes options requiring R&D of new technologies and options not requiring R&D, such as blending to low concentrations with HLW and converting the salt to HLW glass. Regulatory and economic factors will determine whether new technologies or alternative disposition options will be used for salt disposal.

Table 6-1. Driving Implementation Plan Milestone Requirements for Stabilization of Uranium in MSRE **

NMSTG Milestone Number	R&D Need Date	NMSTG Milestone Due Date	DOE Site	Milestone Text
IP-3.5-004B*	Jun 1996	Jun 1997	ORNL	Complete Off-gas System Reactive Gas Removal
IP-3.5-005A*	Sep 1997	Feb 1999	ORNL	Complete Uranium Deposit Removal (Charcoal Bed)
IP-3.5-011A*	Sep 1999	May 2002	ORNL	Complete MSRE fuel salt removal.

* Proposed 94- IP change. ** Refer to 4.1.1 for description of table headings.

6.2 Descriptions and Status for MSRE Remediation Technology

6.2.1 Trapping of Uranium Hexafluoride

A system for trapping uranium hexafluoride has been developed and is being installed at ORNL. R&D is complete to meet this requirement.

6.2.2 Direct Fluorination of MSRE Salt (ORNL)

Uranium can be removed from the salt by sparging molten salt with fluorine gas, producing uranium hexafluoride that can be trapped, and converting the uranium into triuranium octoxide for long-term storage. This process was used in 1968 to remove the initial charge of U-235. R&D is complete to meet this requirement.

6.2.3 Electrochemical Treatment of MSRE Salt (ANL)

Argonne National Laboratory (ANL) is examining a competitive alternative electrolytic ion displacement process as a potential solution to convert the salt to an acceptable waste form while removing uranium. The ion displacement process sequentially displaces salt constituents using a lithium anode and then deposits them on either a solid cathode or liquid bismuth cathode.

6.3 Newly Proposed R&D Activities/Technologies (ORNL)

6.3.1 Glass Material Oxidation and Dissolution System (GMODS)

The GMODS is being examined as a competitive alternative for direct conversion of the salt to a borosilicate waste glass and a secondary clean sodium fluoride stream. GMODS is a general purpose waste treatment process where recent developments indicate its potential to process fluoride-salt wastes. See section 4.3.8.3.1 for a detailed description of GMODS.

6.3.2 Packaging with Getter (ORNL)

Packaging the salt for long-term storage is also being examined as a competitive alternative. Packaging would require a catalytic getter within the package to prevent buildup of $^{233}UF_6$ or F_2.

6.3.3 Charcoal Treatment (ORNL)

Alternative treatment options for processing charcoal containing absorbed $^{233}UF_6$, F_2, and other compounds have been identified and are being experimentally investigated at ORNL. The objective is to treat the charcoal bed in place to reduce the potential for exothermic chemical reactions between absorbed compounds on the charcoal and the charcoal itself. After reduction of chemical reactivity, the charcoal will be removed from the bed and oxidized for recovery and storage of 233-U in an oxide form. All of the treatment options involve purging the charcoal bed with helium and one or more gases designed to chemically inert the charcoal bed.

6.4 Evaluation of Current and Proposed R&D Against Requirements

The technologies described in Section 6.1.2 and 6.1.3 provide an adequate technology base to meet the requirements outlined in Table 6-1. No additional R&D requirements have been identified at this time.

6.5 Conclusions and Recommendations

Conclusions:

1. Trapping Uranium Hexafluoride, Direct Fluorination, Packaging with Getter, and Charcoal Treatment represent low programmatic risk for meeting the R&D need date.
2. Electrochemical Treatment and GMODS for MSRE Salt Immobilization represent medium programmatic risk for meeting the R&D need dates. These technologies for salt processing are in competition with alternative disposition options with fewer technical issues but other institutional or economic issues.

Recommendations:

The NMSTG should arrange for adequate tracking of the technical and programmatic progress of Electrochemical Treatment and GMODS for MSRE Salt Immobilization to ensure that the need dates will be met.

Table 6-2. Summary Technology Status for Stabilization of Uranium in MSRE **

R&D Program	R&D Site	End Use Site	94-1 Milestone	R&D Need Date	Recommendation	Technical Maturity
6.2.1 Trapping of Uranium Hexafluoride	ORNL	ORNL	IP-3.5-004B*	Jun 1996	Baseline	0.42
6.2.2 Direct Fluorination of MSRE Salt	ORNL	ORNL	IP-3.5-011A*	Sep 1999	Comp Alt	3.23
6.2.3 Electrochemical Treatment of MSRE Salt	ANL	ORNL	IP-3.5-011A*	Sep 1999	Comp Alt	7.49
6.3.1 GMODS	ORNL	ORNL	IP-3.5-011A*	Sep 1999	Comp Alt	7.49
6.3.2 Packaging with Getter	ORNL	ORNL	IP-3.5-011A*	Sep 1999	Comp Alt	4.28
6.3.3 Charcoal Treatment	ORNL	ORNL	IP-3.5-005A*	Sep 1997	Baseline	3.02

* Proposed 94-1 IP changes. ** Refer to 4.1.5 for description of table headings.

7. STORAGE TECHNOLOGIES

Storage technology R&D focuses on methods and equipment needed to package materials in containers that meet the DOE-STD-3013 long-term storage criteria and on technologies and procedures for performing surveillance on items in storage. The surveillance activities also play a role in enhancing materials characterization capabilities to better understand stabilization processes and priorities.

7.1 Packaging Technologies

7.1.1 Introduction

Plutonium and other actinide materials are currently stored in a variety of packages, including tanks, piping, plastic containers, and metal containers. When production operations were terminated in the early 1990's, these materials were being held for staging into the production systems. The major sites in the Complex now require the capability to (1) stabilize these inventories into material forms appropriate for long-term storage (metal or oxide) and (2) package these materials according to DOE-STD-3013, which requires that plutonium metal and oxides be stored in dry atmospheres in hermetically sealed containers that do not contain organic materials.

Plutonium storage technologies R&D need dates, driven by 94-1 Implementation Plan milestones, are shown in Table 7.1-1. Need dates are being supported by existing R&D activities; however, there is some uncertainty on meeting the need date for IP-3.3-012A due to remaining development studies and the time needed to approve the pipe component.

Table 7.1-1. Driving Implementation Plan Milestone Requirements for Packaging Technologies **

NMSTG Milestone Number	R&D Need Date	NMSTG Milestone Due Date	DOE Site	Milestone Text
IP-3.2-014	Oct 1997	May 2002	LANL	Thermally stabilize and repackage all plutonium oxide to meet the metal and oxide storage standard.
IP-3.2-027	Sep 1997	Sep 1997	SRS	Modifications to the FB-Line facility (installation of a bagless transfer system) completed.
IP-3.2-022	Oct 1997	Oct 1998	RFETS	New plutonium metal/oxide processing line operational in Building 371 at RFETS.
IP-3.3-012A*	Aug 1997	Aug 1997	RFETS	Begin stabilization by pyrochemical oxidation of 6,000kg higher risk plutonium salts.
IP-3.2-045*	April 1998	April 1998	LLNL	Begin repackaging material to meet the metal and oxide storage standard

* Proposed 94-1 IP change. ** Refer to 4.1.1 for description of table headings.

7.1.2 Description and Status for Packaging Technologies

There are multiple efforts underway that will provide this packaging capability. The main effort involves a standardized design and procurement, managed by DOE Headquarters through a contract with BNFL, to provide a capability based on a bagless transfer system. This system is currently planned to be installed at several sites. LANL is continuing to develop an electrolytic decontamination approach as part of DOE-DP and DOE-MD program activities. This approach also serves as a back-up technology if there are problems identified in the BNFL prototype demonstration. SRS and LLNL are pursuing the implementation of bagless transfer capabilities to address unique constraints.

7.1.2.1 PuSPS Packaging Process (NMSTG). The NMSTG is procuring an end-to-end automated system from BNFL that will stabilize and package plutonium oxides and metal into standardized packages that meet DOE-STD-3013 criteria. The prototype will be installed at RFETS in B/707 Module J. Subsequent units will be installed at RFETS in B/371, Hanford, and SRS. The system is comprised of a stabilization process and a packaging process. The packaging process is being integrated by BNFL.

A System Design Review was held, and the results are documented in the PuSPS System Specification and the PuSPS System Design Document. Completion of end-to-end system testing with a cold surrogate material is scheduled for October 1997. System operational checkout with hot material is scheduled for completion in October 1998.

Loss on Ignition testing is performed on an oxide sample to verify sufficient stabilization. Stabilized oxides are then transferred to the Tipping/Dispense/Fill Area of the nitrogen inerted packaging process, where the tray of oxide is tipped into an empty convenience can and its screw top is replaced. Empty trays are returned to the Material Preparation Area for another batch of oxides.

Later in the packaging process, the convenience cans of either metal or oxide are inserted into an inner can, followed by the insertion of a sealed hollow plug (or bung) that serves as the cap for the inner can. Helium from the glovebox atmosphere fills the space between the convenience can and the inner can. The inner can and plug are laser welded together, then the inner can is cut off from a "stub" end piece by a laser. This allows a "clean" removal of the sealed inner can, with the stub maintaining the boundary seal of the inerted glovebox.

This inner/convenience can assembly is then transferred into an outer can and leak tested. The outer can is evacuated, filled with helium, and a lid is added prior to weld sealing. The final storage package is then checked for external contamination and leak tested prior to moving it to the Lag Storage Trolley for transfer to storage in site vaults.

7.1.2.2 Electrolytic Decontamination Transfer System. For this system, the glovebox is separated into contaminated and uncontaminated sides by a wall with a built-in decontamination fixture. The packaging, can welding, and leak checking of the inner 3013 container are performed in the contaminated side of the glovebox prior to decontamination. The electropolish can is then placed into the decontamination fixture, thereby removing nearly all of the contamination, and is checked to verify that it meets decontamination specifications. After fulfilling these criteria and undergoing a confirmatory leak-check, the can is removed from the glovebox line. Subsequent outer containers can be welded outside the glovebox.

7.1.2.3 Bagless Transfer System for Plutonium Materials. SRS has completed the design and fabrication of a bagless transfer system that uses the same approach as the PuSPS process, but addresses unique facility constraints in FB-Line.

A series of welding tests are being performed to qualify the closure weld. Parameters being evaluated include nominal arc gap, weld current, cut height, and rotation speed. Weld samples will be destructively tested (metallurgical evaluation or burst test) to determine the weld quality. The weld tests will be completed prior to the installation of the bagless transfer unit into the SRS facility.

LLNL is working with SRS to establish an inexpensive bagless transfer system for LLNL materials that will meet DOE requirements. This system will generate welded stainless-steel containers that are nearly identical to those produced at the larger sites.

7.1.2.4 Dustless Oxide Transfer (LLNL). In FY96, LLNL developed a system to transfer plutonium oxide powder within a glovebox without generating dust. This system was first tested with surrogate materials, and with plutonium in FY97. This system was developed to support the Fissile Materials Disposition Program but is applicable for 94-1 operations throughout the DOE Complex. It provides a means to transfer oxide powders without generating dust, which coats gloveboxes, collects in HEPA filters, and increases background radiation dose.

7.1.2.5 Study on the Use of Radioactive Contaminated Scrap Metal for Storage Containers (PFA). The PFA was tasked by DOE-EM-66 to evaluate the potential use of radioactively contaminated metal in fabricating storage containers. The objective of this study was to evaluate important issues related to the use of recycled radioactively-contaminated scrap metal (RSM) for fabricating type 3013 storage packages. The issues addressed were:

1. Life cycle costs of using RSM-based alloys as compared to "virgin" steel
2. The impact of using RSM-based containers on accountancy measurement based on nuclear radiation (spectrometry and neutron coincidence)
3. Availability of scrap metal to produce the 10,000 canisters necessary for plutonium storage
4. Any impacts of the RSM-based materials on transportability of the storage containers using Department of Transportation (DOT)-certified shipping containers
5. The effects of the radioactive constituents on container material behavior
6. The ability to provide container materials that meet national standards.

As a result of this investigation, the following conclusions have been reached:

1. Data to complete a life-cycle cost analysis for fabricating of RSM-based 3013 packages are not available.

2. The impact of using recycled RSM to fabricate 3013 packages is expected to have a negligible impact on material accountancy measurements because the quantity of radionuclides in the recycle product is expected to be less than 1 ppm for any contaminant radionuclide.

3. There is adequate RSM available to produce the 10,000 3013 packages.

4. The presence of contaminant radionuclides in the bulk of the 3013 packages is not expected to impact the transportability of the empty or loaded packages.

5. The effects of contaminant radionuclides in the container material will have a negligible impact on the container material behavior.

6. It is possible to provide RSM-based container materials that meet American Society of Testing and Materials (ASTM) standards for 304 L and 316 L type stainless steels.

Based on the results of this investigation and on a DOE-HQ directive for the beneficial reuse of radioactive steel when possible, it is recommended that RSM steel be recycled to fabricate type-3013 packages.

7.1.2.6 Pipe Component (RFETS). The original intent of the RFETS pipe component was to develop an additional layer of containment that could be used within a Department of Transportation (DOT)-17C drum to optimize the use of the TRUPACT II shipping container. The potential benefits of the pipe component include:

1. Providing radiation shielding to allow handling of 200 fissile grams equivalent (FGE) of high-content pyrochemical salts as contact-handled waste

2. Meeting WIPP immobilization criteria (Note: These criteria were deleted in WIPP WAC Revision 5)

3. Providing a critically favorable geometry that may allow increasing the TRUPACT II fissile material limit from 325 to 2800 grams of plutonium.

Performance testing of the component design (a combination of 30 foot drop tests) was completed in the fall of 1995, and the request for modification to the TRUPACT II Safety Analysis Report for Packaging (SARP) was submitted to the NRC during the spring of 1996. Approval of the SARP is pending.

In January 1996, the *Criteria for Interim Storage of Plutonium Bearing Materials* was released. This document provides material form and packaging criteria for plutonium-bearing residues that are under the scope of DNFSB Recommendation 94-1. It became evident that the pipe component design would meet the storage container requirements of the recommendation and could be modified to provide a structurally enhanced container equivalent to a DOT-Type B container. The pipe component could also be used to address DNFSB Recommendation 94-3 concerns at RFETS. These design modifications were implemented in the spring of 1996, with additional changes to allow both a welded bottom and formed bottom design for the component. Fabrication of the test units was completed, and testing began in early September 1996. To show Type

B equivalency for RFETS storage, both designs are being subjected to the IAEA dynamic crush and DOT 30 minute fire tests. The formed bottom design will receive the 30 foot drop tests previously performed on the welded design for use with the TRUPACT II.

7.1.2.7 Pu-239 Standard Container (NMSTG). The NMSTG is procuring a standard container design from BNFL, which will deliver a test lot of containers. Then a competitive procurement will be made of a large number of containers to meet DOE's need for a standard container designed to meet DOE-STD-3013.

7.1.3 Newly Proposed R&D Activities/Technologies

7.1.3.1 Transportation Container Certification. There may be a need to ship plutonium residues between sites, depending on future decisions on residue stabilization options. DOE-DP has nearly completed certification of the SAFEKEG for shipment of DOE-DP items. It is proposed that the NMSTG provide incremental funding to extend the certification to include shipping the residues.

7.1.4 Evaluation of Current and Proposed R&D Against Requirements

The technologies described in Sections 7.1.2 and 7.1.3 provide an adequate technology base to meet the requirements outlined in Table 7.1-1. No additional R&D requirements have been identified at this time.

7.1.5 Conclusions and Recommendations for Packaging Technologies

By referring to Table 7.1-2, the following conclusions are made:

<u>Conclusions</u>:

All packaging technologies except the pipe component are low risk.

<u>Recommendations</u>:

The NMSTG should arrange for close tracking of pipe component development progress.

Table 7.1-2. Summary Technology Status for Packaging Technologies **

R&D Program	R&D Site	End Use Site	94-1 Milestone	R&D Need Date	Recommendation	Technical Maturity
7.1.2.1 PuSPS Packaging	NMSTG	RFETS	IP-3.2-022	Oct 1997	Baseline	3.02
7.1.2.2 Electrolytic Decon	LANL	LANL	IP-3.2-014	Oct 1997	Baseline	3.09
7.1.2.3 FB-Line and LLNL Bagless Transfer	SRS	SRS/ LLNL	IP-3.2-027	Sep 1997	Baseline	2.77
7.1.2.4 Dustless Oxide Transfer	LLNL	LLNL	IP-3.2-045*	Apr 1998	Baseline	2.09
7.1.2.5 Recycled Metal Study	INEL	NMSTG	IP-3.2-022	Oct 1997	Project Complete	Not applicable
7.1.2.6 Pipe Component	RFETS	RFETS	IP-3.3-012A*	Aug 1997	Baseline	3.93
7.1.2.7 Pu-239 Standard Container	NMSTG	RFETS	IP-3.2-022	Oct 1997	Baseline	2.67

* Proposed 94-1 IP changes. ** Refer to 4.1.5 for description of table headings.

7.2 Surveillance and Monitoring

7.2.1 Introduction

Surveillance of storage containers is required to ensure their integrity and their ability to contain nuclear materials safely. Surveillance technologies can also help characterize materials prior to stabilization and indicate if there are unique hazards associated with individual inventory items. Nonintrusive techniques are required to allow monitoring of storage conditions without violating the integrity of the packaging configuration or producing waste.

Generally, the R&D need date for surveillance and monitoring technologies is October 1998, when the PuSPS becomes operational at RFETS. Surveillance and monitoring technologies R&D need dates, driven by 94-1 Implementation Plan milestones, are shown in Table 7.2-1.

Table 7.2-1. Driving Implementation Plan Milestone Requirements for Surveillance and Monitoring Technologies **

NMSTG Milestone Number	R&D Need Date	NMSTG Milestone Due Date	DOE Site	Milestone Text
IP-3.2-022	Oct 1998	Oct 1998	RFETS	New plutonium metal/oxide processing line in Building 371 at RFETS.
IP-3.2-027	Sep 1997	Sep 1997	SRS	Modifications to the FB-Line facility (installation of a bagless transfer system) completed.

** Refer to 4.1.1 for description of table headings.

7.2.2 Description and Status of Technologies Currently Under Development

One component of the surveillance effort involves the development of nonintrusive techniques to allow monitoring of the long-term storage containers. The other component is the Shelf-Life program, which will assemble a number of inventory items that represent classes of materials in storage from around the Complex. The materials will be placed in containers with special instrumentation to allow detailed measurements of the conditions of the materials and containers, and to help identify problems that may affect an entire class of items in the storage inventory. The information gained will also provide guidance on the adequacy of the long-term storage criteria. This component of the program is discussed in Section 3.1.2.3.

7.2.2.1 Digital Radiography (LANL). Real-time digital radiography is under development to nondestructively gain information on metal, oxide, and residues in storage throughout the DOE Complex. Development efforts focus on gaining experience with commercial equipment and documenting the capabilities of this technique. This method will determine the formation of plutonium corrosion products, assess container corrosion, and detect pressurization.

A real-time radiography system was installed in the LANL Plutonium Facility in FY96. It is being used to inspect residue packages and determine the extent of knowledge that can be gained with this approach before the container is opened for stabilization.

For long-term storage surveillance applications, image resolution studies are determining the sensitivity of this approach for detecting container deformation to identify containers that are pressurizing in storage. In addition, the ability to detect surface corrosion of storage containers is being evaluated, along with the ability to detect changes of the contained materials and identify problems developing in storage.

Real-time digital Radiography is also being evaluated as a means of measuring storage container bulging and/or changes in a pressure-sensitive bellows (see Section 7.2.2.5) to detect pressure buildup before container deformation.

7.2.2.2 Tomography (LANL). Researchers are investigating the feasibility of using x-ray tomography as a noninvasive method for determining the contents and condition of nuclear materials in storage containers.

Using surrogate materials, researchers are evaluating commercially-available x-ray tomography equipment for its ability to distinguish between plutonium compounds in sealed metal containers. The capabilities of this approach to identify plutonium corrosion products, such as plutonium hydride and plutonium oxide, are being investigated. The efforts involve investigating measurement techniques that are complementary to computerized tomography, such as gamma-ray backscattering and alpha-n neutron emission, including algorithms for integrating results with computerized tomographic data. Coupling digital radiography with tomography would provide a very useful surveillance capability.

In FY96, existing tomography equipment was installed and tested in PF-4 at LANL to evaluate the approach and establish specifications for a prototype system. The prototype system will be installed in FY97 and tested in FY98. Tomography standards are being used to evaluate commercially-available equipment.

7.2.2.3 Laser Sampling (LANL). A sampling system has been developed to enable researchers to obtain gas samples from within a sealed plutonium storage container. This method uses a laser to make a small penetration in the container and release a sample of the gas inside. The laser then reseals the container, eliminating the requirement to repackage and requalify materials after testing. This sampling system was completed for the SRS primary containment vessel. The LANL research project is complete, and the system is ready for delivery to SRS.

7.2.2.4 Acoustic Resonance Spectroscopy (LANL). Acoustic resonance spectroscopy is being developed as a non-intrusive means of detecting changes in the pressure and composition of gases inside long-term storage containers. Specific resonances due to longitudinal and radial gas modes for argon and helium have been identified. The behavior of these gas modes as a function of temperature, gas pressure, and gas mixture has been studied. Changes in gas pressure and composition can be readily seen in the acoustic spectrum, and quantitative predictions of the gas composition can be made. With current instrumentation, the detection limit for a single container is less than 1 pound per square inch.

Studies will continue using surrogate materials in cans to represent plutonium metal and oxide. Measurement of partial pressures for hydrogen, oxygen, and helium components will also be researched. Experiments are being conducted to study the effects of temperature, data acquisition rate, frequency range, and step size on the ability to detect changes within a can. Areas needing investigation include (1) the ability to detect changes in pressure, gas composition, and in the can itself, and (2)the ability to observe changes in a can within a second can. In FY97, efforts will continue to establish the applicability of this technology to long-term surveillance requirements.

7.2.2.5 Pressure Sensitive Devices (LANL). Pressure-sensitive devices installed in storage containers will provide the capability to detect gas generation and determine if restabilization and repackaging are required for safe storage. There are two devices under development—aneroid bellows and micro-electronic hydrogen sensors.

The development of aneroid bellows has resulted in the successful demonstration of a commercial bellows. Pressure changes (or bellows failure) are determined by detecting length changes in the bellows via radiography because the bellows are inexpensive enough to be installed in a large number of containers. This technology is available for application in both the Shelf-Life efforts and more general surveillance efforts.

This activity also involves developing hydrogen sensors and a data telemetry system to monitor the presence of hydrogen in nuclear material storage containers. Knowing the amount of hydrogen gas that has evolved in the containers is an important factor for improving the understanding of the chemical processes that occur during long term storage. This system will be capable of measuring hydrogen levels from the part-per-million (ppm) level to 100% hydrogen. Telemetry data transmission will preserve the integrity of the storage container by avoiding breaching the container with wires or cables.

In early FY97, a sensor system will be delivered for testing at LANL. The sensors will be evaluated for use in the Shelf-Life program. If the initial work demonstrates the utility of this approach, follow-on work will include developing an oxygen sensor and a total pressure sensor.

7.2.2.6 Risk-based Prioritization for Plutonium Residues (LANL). In FY96, a methodology was developed to help personnel prioritize the remediation of legacy materials and evaluate the impacts of schedule modifications. The effort was first directed at addressing issues at LANL, and then generalized to provide criteria for similar needs at other sites.

The probability of failure of one of the primary containment barriers was developed from a statistical sampling of vault items for six different material categories. From this information, estimates were developed on the probable number of items with one contamination barrier rendered ineffective for each material category. These estimates were used to calculate probabilities on risk event trees, which provide a way to assess the relative impacts of changing stabilization priorities and schedules.

Workshops with other sites were conducted to discuss how this methodology could be applied to provide a more rigorous basis for prioritization. All sites have already developed components of this methodology. Complex-wide criteria will allow DOE to establish inter-site priorities.

7.2.2.7 Non-Destructive Assay—Surveillance of Plutonium (LANL). Work will focus on the evaluation of nondestructive assay methods for a variety of plutonium forms. Special emphasis will be given to those difficult-to-measure materials common to many DOE sites. In general, the approach will be the application of existing nondestructive assay methods and investigation of new and promising technologies. Specific areas of investigation will be:

- The gamma ray analysis of heterogeneous materials such as molten salt extraction residues will be given high priority. The ability to accurately determine plutonium isotopic distribution and americium concentration is required to support both calorimetric assay and neutron measurements. Efforts will build on existing technology already in use at LANL and extend that technology to difficult to measure materials.

- Neutron assay is also a promising method for assay of difficult materials and matrices. Although calorimetric assay is often more accurate, neutron assay is faster and may be advantageous for some materials. Investigations will emphasize those methods which will improve measurements for difficult to measure materials.

- Calorimetric assay is often the most precise and accurate method of nondestructive assay. It is insensitive to chemical matrix and this calorimetric assay is an excellent choice for measurement of many difficult materials such as pyrochemical processing residues. However, calorimetric measurements take a long time and calorimeters are bulky, large instruments. Work will apply finite element analysis to obtain a model that will predict calorimeter performance and permit the design of smaller, faster calorimeters.

The objective is to optimize nondestructive assay methods and to investigate promising technologies for the measurement of various materials, to include the capability to perform accountability and waste measurements of nuclear material essential for recovery and clean up efforts.

7.2.3 Newly-proposed R&D Activities/Technologies

7.2.3.1 Integrated Surveillance System (INEL). An integrated data acquisition, storage, and retrieval system is to be developed for use throughout the life of plutonium-bearing material to ensure safeguards and accountability requirements are met. This work will identify a consistent set of surveillance requirements and process functions using a proven systems engineering process. The process will define customer needs for an integrated system to survey and monitor stabilized plutonium packages during storage. Detailed major functions of the system will cover the following areas:

1. Characterization of stabilized plutonium-bearing material packages

2. Surveillance and monitoring of the containers and contents during storage

3. Tracking containers and contents throughout the DOE Complex until final disposition.

7.2.3.2 Non-Intrusive Measurement of Water on Plutonium Oxide using Neutron Interrogation. Measurement of absorbed moisture has the potential for presenting a significant problem in packaging plutonium oxide. Two methods involving interrogation of the sample with neutrons may provide a means of measuring the moisture content of an entire batch of oxide after it is packaged. Both techniques are non-intrusive and interrogate the sample with Californium-242 neutrons which are captured by hydrogen species in the sample. One method being considered is based on the analysis of moderated neutrons. The second method, prompt-gamma neutron activation analysis (PGNAA), analyzes gamma-rays released during the capture of neutrons by hydrogen.

This work will evaluate the sensitivity of both techniques and determine their potential for replacing the conventional LOI method.

7.2.3.3 Thermal Modeling of the Temperature Distribution in Oxide and Metal Containers under Various Storage Configurations. DOE-STD-3013 requires that the temperature of unalloyed plutonium metal not exceed 100C during storage. In addition, the Standard assumes the maximum storage temperature for oxide would not exceed 204C. There are no physical measurements of temperatures or theoretical temperature calculations for these materials under storage conditions. Such information is needed to perform shelf-life storage studies and design storage vaults.

This work involves preparing a theoretical thermal model to predict temperatures of plutonium metal and oxide under various storage configurations and to make confirmatory physical measurements.

7.2.4 Evaluation of Current and Proposed R&D Against Requirements

The technologies described in Section 7.2.2 will provide an adequate technology base to meet the requirements. No additional R&D requirements have been identified at this time.

7.2.5 Conclusions and Recommendations

Referring to Table 7.2-1, the following conclusions are made:

Conclusions:

1. Nonintrusive surveillance systems need to be developed to monitor plutonium packages in long-term storage at RFETS.
2. All surveillance technologies represent a low programmatic risk for meeting R&D need dates.

Recommendations:

1. Other systems (e.g., digital radiography/tomography, acoustic sampling, and pressure-sensitive devices) appear viable and should be developed and evaluated as part of an integrated systems approach for nonintrusive surveillance. The results of these evaluations should be used to minimize the potential for personnel radiation exposures and to maximize adherence to safeguards and security requirements.
2. Other areas of investigation include (1) the ability to detect changes in pressure, gas composition, and in the can itself; and (2) the ability to observe changes in a can within a second can.

Table 7.2.-1. Summary Technology Status for Surveillance **

R&D Program	R&D Site	End Use Site	94-1 Milestone	R&D Need Date	Recommendation	Technical Maturity
7.2.2.1 Digital Radiography	LANL	RFETS	IP-3.2-022	Oct 1998	Comp Alt	1.14
7.2.2.2 Tomography	LANL	RFETS	IP-3.2-022	Oct 1998	Comp Alt	3.88
7.2.2.3 Laser Sampling	LANL	SRS	IP 3.2-027	Sept 1997	Backup	4.93
7.2.2.4 Acoustic Resonance Spectroscopy	LANL	RFETS	IP-3.2-022	Oct 1998	Comp Alt	4.23
7.2.2.5 Pressure-Sensitive Devices	LANL	RFETS	IP-3.2-022	Oct 1998	Comp Alt	3.95
7.2.2.6 Risk-Based Prioritization of Residues	LANL	All	IP-3.3-034	Sep 1995	Project Complete	Not applicable
7.2.3.1 Integrated Surveillance	INEL	RFETS	IP-3.2-022	Oct 1998	Baseline	Not Applicable

** Refer to 4.1.5 for description of table headings.

8. ENGINEERED SYSTEMS

8.1 Introduction

Each of the processes outlined in Sections 3 through 7 and in Section 9 may require or involve systems engineering, probability risk assessment, human reliability assessment, end-to-end (local and remote) automated systems, and (advanced) human interfaces for safeguards and security. Development of automated systems may require a trade study or need assessment to define what systems best meet 94-1 needs, to eliminate any possibility of duplicating efforts, and to evaluate what requirements may still need to be developed. The hardware and software implemented should be of the latest technology currently available on the market.

Two white paper proposals involving robotics automation (and currently funded by EM-50 Robotics) have been submitted to the PFA for review and proposed inclusion in the PFA FY97 program.

8.2 Description and Status for Engineered Systems Technologies

8.2.1 Modular Concepts (LANL)

The objective of this effort is to develop and optimize integrated, skid-mounted, modular processing systems to identify where this approach has cost or schedule advantages over baseline processing plans. The fundamental aspect of the modular approach is to establish an authorization basis for operating the equipment that minimizes the reliance on aging systems in existing major facilities. The costs for such an approach may be projected to be more expensive than relying on existing buildings, but the schedules for implementation are more predictable. In addition, modular systems provide the flexibility to process different materials with minimal impact on the over-all system.

In FY96, efforts focused on the development of conceptual designs for addressing several RFETS stabilization requirements. Efforts will continue in early FY97 to support a decision on whether or not to pursue the implementation of this approach.

8.2.2 PuSPS Integration (NMSTG)

The NMSTG is procuring an end-to-end automated system from BNFL to stabilize and package plutonium metals and oxides into a standard package that meets DOE-STD-3013 criteria. The prototype will be installed at RFETS in B/707, Module J, and subsequent units will be installed at RFETS B/371, Hanford, and SRS. The system is comprised of a stabilization process, integrated by Raytheon Constructors, and a packaging process, integrated by BNFL.

Overall system integration is the responsibility of BNFL and requires hardware and software items in addition to the stabilization and packaging processes. The integration hardware includes:

- A Lag Storage Trolley
- Furnace Trays
- Storage Packages
- Receipt Hood/Fume Cabinet/Glovebox Atmosphere
- Electrical Interface
- Support Structure and Platforms
- Fire Protection
- PuSPS Control Hardware
- Process Radiometric Instrumentation
- Automation Control

The integration software requirements include:

- Stabilization Control
- Packaging Control
- Data Management
- Data Highway Communication
- Process Radiometric Instrumentation Software
- Human-Computer Interface
- Automation Control

A System Design Review was held, and results are documented in the PuSPS System Specification and the PuSPS System Design Document. Completion of end-to-end system testing with a cold surrogate material is scheduled for October 1997. System operational checkout with hot material is scheduled for completion in October 1998.

8.3 Newly-Proposed R&D Activities/Technologies

There are no newly-proposed R&D activities that need to be addressed at this time.

8.4 Evaluation of Current and Proposed R&D Against Requirements

8.4.1 Automation

The need for automation is not integrated into the *Baseline R&D Technical Requirements* document at this time. A trade study is required to evaluate the necessity and validity of using automation in surveillance and monitoring, packaging, and storage container handling.

8.5 Conclusions and Recommendations

Conclusions:

The need for automation is currently not integrated into the *Baseline R&D Technical Requirements* document.

Recommendations:

1. Automation baseline requirements need to be established (to include such areas as Systems Engineering, Human-Computer Interfaces, and Task Performance).

2. A trade study or need assessment is recommended to examine and develop baseline hardware, software, and human factors requirements to evaluate the necessity and validity of using automated technology that may or may not currently exist at DOE sites (or in the commercial realm).

9. CORE TECHNOLOGY

9.1 Introduction

In accepting DNFSB Recommendation 94-1, DOE developed an Implementation Plan that included a Core Technology Program. The objectives of the Core Technology Program are (1) to augment the knowledge base about general chemical and physical processing and about storage behavior of actinide-containing materials, and (2) to ensure safe interim nuclear material storage until disposition policies are formulated. Each Core Technology Program activity relates to one or more technology-specific R&D areas (i.e., standards, packaging, surveillance, transportation, and stabilization).

Although the fundamental chemical and physical data base to support weapons production and nuclear fuel development is extensive, new requirements for stabilization and storage call for better understanding of many areas of actinide science. The Core Technology Program is needed to develop a detailed understanding of the processes that affect stabilization, to provide technology for predicting the long-term behavior of nuclear materials in storage, and to develop and maintain capabilities to address unforeseen problems associated with managing these materials over time.

Maintenance of these capabilities is necessary to further develop state-of-the-art technologies applicable to DOE Complex-wide needs. Such technologies are extremely desirable because technologies within the DOE Complex are not designed to current waste reduction and worker safety requirements. Further, this effort is designed to educate and retain personnel essential to key 94-1 missions and to provide hands-on training for scientists who will be needed to address unforeseen problems. The development of a strong effort in fundamental actinide science will also enhance DOE's ability to ensure the success of other DOE programmatic missions.

9.2 Description and Status of Current Core Technologies

A series of R&D Tasks have been developed to enhance detailed understanding of technical problems associated with the stabilization and long-term behavior of plutonium-containing materials. In developing these tasks, the NMSTG has solicited input across all scientific disciplines. The coordinated set of tasks is a multidisciplinary approach to the understanding and control of these complex materials over time.

9.2.1 Molecular Characterization and Chemical Behavior

As nuclear materials degrade due to their radioactive or chemical nature, the chemistry of the remaining material becomes more complex. Understanding the chemical behavior of complex materials requires characterization beyond simple elemental analysis. A detailed description at the molecular level is needed if the chemistry is to be understood and controlled, thus allowing more informed decisions to be made concerning material stabilization.

9.2.1.1 Actinide Solution Chemistry. Because of the complex nature of plutonium solutions that have been stored for extended periods of time, the chemistry of these solutions must be well understood for proper and safe stabilization. More generally, there will be a continuing need to maintain a core competence in actinide solution chemistry to address unforeseen issues with current solution inventories and with new solutions generated through stabilization activities. Separating actinides from other elements and from each other generally relies on partitioning the actinides into a different phase from the bulk of the material. Important chemical factors in separations processes include: oxidation state, thermodynamic formation constants, solubility, and sorption characteristics.

This task focuses on plutonium solution chemistry because major solution chemistry issues are associated with applying traditional acid solution separations processes to largely neutralized and alkaline residues and wastes. Initial work will complete experiments aimed at understanding the chemistry of Pu (IV) in nitric acid, a chemical system of central importance to traditional plutonium separations processes. Surprisingly, this system is poorly understood at the molecular level, especially at high-acid concentrations. In particular, the effects of halide and alkali metal ions on plutonium complexation are incompletely understood and will be explored.

Besides stabilizing higher oxidation states, such as Pu (V), carbonate complexation at high pH can dominate over nitrate and phosphate complexation. Therefore, carbonate complexation and solubilization of plutonium in alkaline solutions are being measured. While good room temperature data is available for carbonate reactions under environmental conditions, little work has been done on the temperature or ionic-strength dependence under conditions relevant to existing residues or likely residue treatment processes.

Chloride complexation of plutonium under conditions relevant to residue treatment is another significant and fundamental area of plutonium chemistry that is not sufficiently understood. Chloride appears to compete with carbonate for plutonium coordination in alkaline solutions, if the chloride concentration is high enough. It is also important to understand how reduction-oxidation chemistry and the solubility of plutonium is affected by coprecipitation with hydroxides of other metals that may be made less soluble by treatment. The limited literature that exists on plutonium under these conditions shows that these conditions are poorly understood.

Structure-specific electronic absorption bands of Pu (V) shift as plutonium is complexed. These changes will be measured and correlated with parallel extended x-ray absorption fine structure (EXAFS) experiments on well characterized referenced materials.

9.2.1.2 Chemical and Physical Interactions of Actinides with Surfaces. Actinide species interact with surfaces through a number of mechanisms (e.g., surface complexation, physiosorption, matrix diffusion, and entrainment) to yield a wide range of surface-bound species. Stabilizing residues by removing actinides from surfaces, while minimizing secondary waste, is strongly influenced by actinides-surface interactions. Molecular spectroscopic probes will be employed on several prototypical actinide/substrates to obtain detailed characterization and imaging of the surface interaction between the actinide species and the substrate. This information can then be folded into existing and new stabilization technologies to optimize cost and efficiency.

Substrate systems have been chosen to represent residues found in ash and combustibles. Silicates will be used to represent the dominant phases found in incinerator ash and ash-heel. As prototypes for combustibles, LANL will examine ion-exchange resins and cellulosic and plastic materials. These material classes provide a diverse range of surface chemical interactions, while representing a significant fraction of the total residue mass within the DOE Complex. The chemical and structural characteristics of the resulting actinide-substrate complexes will be determined using a combination of diffuse reflectance, luminescence, photoacoustic, Raman, and x-ray spectroscopies. This suite of spectroscopic tools will provide oxidation state, elemental composition, and molecular or phase composition of surface-bound actinide species. While the principle focus will be on substrate interactions with plutonium, U (IV) and U (VI) species will be used to provide baseline information in the initial studies. Actual residues from each material class will be used to confirm the correlation in actinide-substrate interactions between synthetic controlled samples and real residues.

9.2.1.3 Changes in Chemical State of Plutonium. Large volumes of plutonium-containing materials that are not pure oxide or metal are being (and will be) stored. Storage durations of decades are anticipated, yet the chemical behavior of plutonium-containing materials over this time-scale is not known. These materials contain constituents that can lead to gas generation and container corrosion. The time-dependent chemical behavior of these materials, especially when in direct contact with plutonium, needs to be studied to ensure that any chemical changes that do occur do not adversely affect the storage characteristics.

The oxidation state of plutonium is especially vulnerable to aging effects. It has been shown that x-ray techniques can determine plutonium valence and chemical structure. Since x-ray absorption spectroscopy is sensitive to the local chemical environment of a target atom, it is an outstanding analytical technique to study changes in plutonium-containing materials. X-ray diffraction and photoelectron spectroscopy will also be applied in this task. Residues such as ash, salts, alloys and carbides will be targets. This effort is conducted in collaboration with research on accelerated aging effects, which is separately supported under the Shelf-Life Program.

Early identification of constituents in stored materials and understanding of chemical changes in those materials is necessary for assuring that stabilization objectives are met. This information may also guide future disposition technologies. A comprehensive understanding of the chemical changes that occur in materials will ensure the safety of the storage media and aid in the environmental assessment of sites where the materials will be disposed.

9.2.1.4 Actinide-Organic Interactions. Proposed treatment schemes for combustible residues include thermal (pyrolysis), electrochemical (MEO), and chemical (hydrothermal, catalyzed chemical oxidation) methods to destroy the host organic matrix and leave the actinide in a form suitable for stabilization. The nature and effect of actinide interactions with organic residues, although fundamental to each of these treatment methods, is unfortunately not well developed. A number of problems associated with combustibles involve actinides entrained, complexed, and/or sorbed within organic matrices, with uncertain affect on residue treatment, storage, stabilization, and disposition. Improved understanding of the underlying actinide-organic chemistry should result in better performance, predictability, and understanding of matrix destruction technologies.

The actinide-organic core technology task focuses on the chemistry associated with the interaction of organic moletics with actinides. This involves understanding the interaction of actinides entrained or sorbed on solid organic matrices, or the complexation of organic species in the presence of actinide ions in aqueous solution. Factors that can affect process technologies include heterogeneous organic speciation, surface and solution kinetics, and chemical reactions under non-equilibrium conditions. FY97 work will address the solid-state characterization of plutonium on one or more representative organic matrices (e.g., plastics, flow-through filters) and the chemical speciation of plutonium with one or more organic acids in aqueous media. The surface work will focus on the nature of actinide entrainment, sorption, complexation, diffusion, or deposits within surface imperfections. The solution work will focus on chemical speciation of actinide-organic complexes, structure and solution equilibria, and oxidation states.

The second activity in the actinide-organic interaction task involves the synthesis and characterization of new organo-metallic compounds of plutonium, americium, and neptunium. Several new classes of compounds having varied ligand sets will be prepared and their dynamic solution chemistry investigated. An understanding of ligand exchange processes in these model systems will allow the modeling and prediction of behavior in more complex and/or new systems. Such knowledge is relevant to the development of new separation and stabilization technologies for actinides and could lead to the development and testing of new characterization and analysis processes.

9.2.2 Efficient Separations

The technology baseline requirements of the November 1995 94-1 R&D plan included actinide removal from aqueous solutions in several areas of stabilization process development. Robust actinide removal processes are needed for a variety of aqueous streams ranging from low-to-high pH and from dilute-to-high salt concentrations.

9.2.2.1 Polymer Filtration. Polymer filtration uses chemically modified water-soluble polymers to selectively bind radioactive metals, such as plutonium, americium, thorium, and uranium. The solution containing the metal-loaded polymer is concentrated using commercial ultrafiltration equipment with the filtered water meeting all regulatory requirements. The metal-loaded polymer can go directly to disposal, or the metals can be recovered and the polymer recycled.

The polymer filtration process is designed to remove actinides to very low levels from solutions produced by stabilization operations. Advantages of polymer filtration, a technology based on water-soluble metal binding polymers used with commercially available ultrafiltration equipment, over current actinide separations technologies include rapid metal-binding kinetics, a single aqueous phase, high throughput, high selectivity and capacity, low energy and capital costs, and a small equipment footprint.

The objective of this core technology task is to optimize the polymer filtration process by understanding key properties of the polymers at the molecular level as a function of important parameters. Properties include solubility, filterability, binding strength, and capacity. Parameters include metal ion loading, pH, polymer concentration, metal ion separation, and ionic strength. Since the metal-binding polymer is key to the process, understanding the properties of the polymers is of utmost importance.

The main 94-1 application areas include aqueous process stream polishing of plutonium, americium, uranium, and/or neptunium; mixed waste stream treatment; and preconcentration for analysis purposes. The research includes the following:

1. Development of direct probing techniques to characterize metal-bonding polymers
2. Correlation of direct structural results with physical and chemical properties of the polymer
3. A rapid survey to evaluate many structural effects and to obtain a structure-function relationship in the polymers
4. Improved synthetic procedures for optimized polymers
5. Evaluation of polymers for designed applications, including selectivity studies.

This project benefits from strong collaboration with several university and industrial partners.

9.2.2.2 Polymer Foams. Ion-exchange and chelating resins, typically in bead form, are currently used to remove nuclear materials and other solutes from processing and waste streams. However, relative to foams, bead materials have problems that tend to lower their efficiencies, including: packing instability, liquid channeling through the bed, slow sorption or desorption kinetics, and smaller effective surface area. The goal in this research is to develop advanced microcellular polymeric foams to overcome these problems and to demonstrate the viability of polymer foams for plutonium and americium separation from liquid streams.

Several types of foams containing different function groups will be used. The first series will contain phosphonic acid groups that have been attached to chloromethylstyrene foams. The second series will contain multifunctional phosphonic acid or phosphonate groups that have even higher binding constants and selectivity. Another series of foams will incorporate polyvinylpyridine units in which the pyridinium group provides an anion-exchange site in acidic media. The foams will be engineered into a column design, and their flow properties will be optimized to maximize available surface area for a given mobile-phase throughput rate. Plutonium and other metal ion uptake will be investigated, including kinetics and capacities as a function of pH. Collaboration on graft polymerization experiments with separations experts at the University of Tennessee will support this task.

9.2.3 Materials Science

The behavior of common engineering materials is being studied to provide a technical basis for materials specification for stabilization technologies as well as for predicting the behavior of long-term storage systems.

9.2.3.1 High Temperature Thermodynamics and Kinetics. The primary objective of this activity is to maintain state of the art competency in experimental actinide chemical thermodynamics and kinetics. This competency is crucial because thermodynamic and kinetic properties ultimately define all actinide material processes and end states.

A number of characteristics establish the high temperature properties of a chemical reaction system including: enthalpy and entropy change, activities of the components, reaction rates and mechanisms, and identity of the species involved in the reactions. Techniques available to evaluate these system characteristics include: high-temperature Knudsen effusion mass spectrometry, differential thermal analysis, differential scanning calorimetry, thermogravimetric analysis (TGA), x-ray diffraction, and electrochemical techniques. Ultimately, the thermodynamic, kinetic, and chemical reaction information can be used to construct models for thermal processes relevant to many stabilization and storage issues.

FY96 technical work was focused on the vaporization behavior of plutonium oxychloride to better understand the vaporization chemistry and kinetics of the salt distillation process. In FY97, these studies will continue and extend to americium oxychloride and alkali halide/actinide oxyhalide salt mixtures. Thermodynamic and kinetic support will also be provided to other 94-1 tasks including other halide and oxide systems and waste form work on actinide bearing-minerals and glasses.

9.2.3.2 Corrosion. The effects of crystallography on localized corrosion (pitting) and general corrosion (dissolution of surfaces) are being examined by coupling the scanning reference electrode technique with orientation imaging microscopy. The objectives of this study are:

1. To examine the effects of crystallographic orientation on the electrochemical behavior of engineering materials to discern the effect of surface structure on pitting
2. To gain insight into the stochastic nature of corrosion
3. To improve corrosion resistance of common materials through intelligent surface processing or texturing.

These issues are pertinent to immediate stabilization and storage aspects of the 94-1 mission and to maintaining longer-term materials core competencies.

Previous corrosion studies have focused on the corrosion behavior of single crystals with exposed low index planes. This study will enable examination of actual polycrystalline engineering materials containing crystalline phases examined previously. Also, all crystallographic orientations will be examined, instead of a few chosen orientations. Depending on the results, subsequent tests could incorporate electrochemical studies on single crystals to further detail the electrochemical behavior of individual orientations of interest.

In FY96, the focus was on characterizing the bulk effects of texture on selective attack. In FY97, the focus will be on characterizing local grain orientation effects on the initiation and propagation of surface pitting. In FY98, the effort will focus on processes for optimizing material texture for corrosion prevention.

This project has resulted in a strong collaboration with SRS, which has technological interest in the effect of crystallographic orientation on the corrosion of piping systems that handle hazardous waste, specifically salt systems. Consequently, in FY96, a portion of the overall corrosion research program was leveraged with funds provided by SRS. The program will be leveraged in FY97 by surveillance and enhanced surveillance funding. Collaborations with Carnegie Mellon University and the University of California-Berkeley also strengthen this task.

9.2.3.3 Diffusion. Diffusion of plutonium into structural and storage materials may be a limiting factor for 94-1 issues, such as long-term storage and contamination control. This work seeks to measure the diffusion coefficients of plutonium in engineering materials, such as stainless steel at room temperature, to reliably predict behavior over long time periods.

Plutonium diffusion coefficients have been measured at high temperatures for some materials. These diffusion coefficients are known to be highly dependent upon temperature and concentration. Extrapolation of these diffusion coefficients to the temperatures relevant to long-term storage have not been verified experimentally. Use of extrapolated diffusion coefficients (when available) in current models can lead to errors of many orders of magnitude. Therefore, experimental measurements of diffusion coefficients near room temperature are necessary. Such measurements require experimental techniques that can measure very small plutonium concentration in very thin material. This study will use an array of surface characterization methods to determine the plutonium diffusion coefficients for various forms of plutonium into different engineering materials at ambient temperature. This information will result in more accurate predictions of plutonium diffusion for specified time periods near room temperature.

In FY96, materials for study and surface science techniques were selected. Experimental work will begin in FY97. This work will integrate historical information from nuclear weapons literature with direct measurements on available aged material and well-characterized models.

9.2.3.4 Vitrification. The objective of this task is to develop, install, and demonstrate a glovebox-scale vitrification process that can be used to study the application of vitrification as an alternative to cementing certain plutonium-contaminated waste materials. In addition, vitrification may play a role in converting materials to an acceptable disposal form.

Glasses are an internationally-accepted waste form for fission product wastes, but vitrification has not been equally developed for actinide residues or wastes. Tests will begin with materials known to be suited for vitrification, such as incinerator ash, and progress to more challenging types.

Cementation, a current plutonium waste management practice, significantly increases waste volume. Vitrification results in a decrease in waste volume, which may translate to reduced disposal costs. Vitrification of residues also decreases the attractiveness of recovering the plutonium, which enables a reduction in safeguards requirements.

Planning and evaluation of vitrification equipment was completed in FY96. In FY97, equipment will be procured and cold testing completed. In FY98, the equipment will be installed, tested with plutonium-bearing materials, and made available for a series of vitrification viability studies.

9.2.3.5 Mineral Waste Forms. The purpose of this task is to enhance the technical basis for alternative waste forms for stabilizing plutonium for both intermediate and long-term storage. The FY97 goal is to determine the preparative conditions and solubilities of plutonium and americium in binary actinide-silicate or ternary actinide-zirconium-silicates. These systems were selected because of the well known stability of orthosilicate materials, such as zircon ($ZrSiO_4$).

To prepare actinide-zirconium silicates, high-purity oxide powders will be mixed with silicon dioxide powder and pressed into pellets at high temperatures and pressures. The product will be ground into powder and subjected to x-ray powder diffraction and chemical analyses to determine product phase and composition. Once the actinide silicate has been synthesized, it will be mixed with commercial high-purity zircon in various proportions and then pressed into pellets. The resulting material will be analyzed for equilibrium phases and actinide content. Once phase equilibria have been established and optimum loadings have been estimated, further experiments will be conducted to study the effects of temperature, pressure, and starting material characteristics on the reaction rate, yield, and microstructure of the products.

This effort will be leveraged by related work, funded through the LANL Laboratory-Directed Research and Development (LDRD) program. The comparison of plutonium americium behavior will provide not only fundamental phase data, but also may offer insights as to differences in the long-term stability of these waste forms. This project benefits from collaborations with the University of New Mexico and Pennsylvania State University.

9.3 Newly-Proposed Core Technology Activities

9.3.1 Pu (III), (IV), and (VI) Phosphates

This is a collaborative effort between LANL, Pennsylvania State University, and the University of Maryland. This task will develop the understanding of plutonium phosphate chemistry in solution and solid states using synthetic efforts coupled with structural, spectroscopic, and analytical characterization. The correlation between observed structural features and spectroscopic signatures will be determined to develop a fundamental understanding of structure/spectroscopy relationships and a database of actinide phosphate compounds. This knowledge will be applicable to developing selective phosphate ligands for separation and remediation purposes, and will provide insight into phosphate-mediated actinide behavior in waste streams and in the environment.

Phosphate coordination plays a significant but poorly understood role in actinide chemistry. Phosphate minerals have been proposed as secondary barrier or backfill materials to retard canister corrosion and enhance radionuclide adsorption in underground repositories. Organic phosphates have been used widely in plutonium processing in which insoluble plutonium phosphate salts have been reported but not identified. In contrast to hundreds of structurally analyzed transition metal phosphates, only seven actinide phosphates (mostly involving uranium) have been fully characterized and reported. Thermodynamics of plutonium phosphate compounds have been reported by a few groups, but the results often have been contradictory to studies with other actinides. Factors governing the formation, structure, and bonding of plutonium phosphate compounds are not clear.

Initial efforts will focus on the synthesis and structural characterization of Pu (III), Pu (IV), and Pu (VI) compounds, especially those that show promise as waste forms. A variety of synthetic strategies will be investigated, including sol-gel, hydrothermal, and simple precipitation methods. Soluble complexes will be characterized in solution by various spectroscopic techniques. Solid compounds will also be characterized by x-ray diffraction, infrared and Raman spectroscopy, electronic absorption spectroscopy, x-ray absorption, and nuclear magnetic resonance. Resistance to radiation damage and corrosion will be investigated. Thermodynamic properties, such as solubility products and formation constants, will be determined by conventional methods.

9.3.2 Nonaqueous Actinide Electrochemistry

A critical step in many actinide separation processes is adjusting actinide valence states. In this task, the electrochemical transfer reactions of selected actinides and actinide complexes will be studied in non-aqueous solvent systems that heretofore have not been investigated with actinides. By using the unique properties of room temperature molten salts and liquid sulfur dioxide, it should be possible to stabilize oxidation states that are not accessible in aqueous acidic media. This will allow, for the first time, an accurate measure of thermodynamic reduction/oxidation potentials and heterogeneous electron transfer kinetics of these elements in various oxidation states. Compilation of this data will add to fundamental knowledge of the actinide elements and serve as a technical basis for next generation stabilization and separation processes.

Missing from current knowledge of actinide elements is information about heterogeneous electron transfer reactions (i.e., the electron transfer between the solvated solution and an inert electrode). Dynamic electrochemical techniques have been used to characterize the kinetics of electron transfer for most elements, but data is conspicuously missing for the actinides. This task will investigate the use of recently developed ultramicroelectrodes that can measure electron transfer rates several orders of magnitude faster than previously possible.

9.3.3 Actinide Self-Fluorescence

The self-fluorescence signatures of plutonium, uranium, and other actinides will be investigated systematically as a basis for developing analytical methods and instrumentation for nondestructive assay and evaluation. The intent is to develop and experimentally validate a model of the various mechanisms that give rise to self-fluorescence for potential surveillance applications.

X-rays arise from the self-fluorescence of actinides, particularly plutonium and uranium, which can be observed using high-resolution gamma-ray spectroscopy. Energetic alpha particles, beta particles, and gamma rays produced by the decay of plutonium isotopes, for example, induce self-fluorescence. The rate of self-fluorescence is a strong indication of actinide concentration and the relative probabilities for the various decay processes. The fluorescent x-ray intensities are also dependent upon radiation transport properties of the matrix. For example, alpha particles have a very short range in most materials of interest, and consequently are important only for materials with high actinide concentrations (e.g., metals and oxides). In contrast, gamma rays can induce fluorescence over a much larger volume.

Computational techniques will be developed to model gamma, beta, and alpha transport processes, as well as second order compositional and matrix effects that underlie x-ray self-fluorescence signatures. A Monte Carlo model will be developed and validated experimentally using uranium solutions, mixed-oxide solids, and plutonium oxide samples of various shapes, sizes, and densities. The model then will be used as a basis for interpreting self-fluorescence signatures.

9.3.4 Extractive Scintillators for Alpha Counting

This is a collaborative effort between Tennessee Tech, LANL, and Ordela, Inc.. The traditional analytical method for determining alpha emitters, such as plutonium, is alpha spectroscopy. This method offers excellent energy resolution, but requires time-consuming sample preparation to eliminate interfering radiological and chemical components. In this task, researchers will investigate the use of a Photon Electron Rejecting Alpha Liquid Scintillation (PERALS) spectrophotomer in combination with extractive scintillators to develop faster, more sensitive procedures for quantifying alpha emitters in aqueous media.

The PERALS system is a commercially available instrument that offers a number of advantages for alpha liquid scintillation. Using electronic pulse-shaped discrimination, the PERALS system can reject beta and gamma pulses with 99.9% efficiency, offers very low background, and has sufficient energy resolution to quantify isotopes such as U-234 and U-238.

In close collaboration with LANL staff, the initial phase of the project would test new extractive scintillators and solid exchangers on uranium and thorium at Tennessee Tech. Promising systems would then be tested with plutonium and americium at National Laboratory sites. Ordela, Inc. of Oak Ridge, Tennessee, will serve as an industrial partner.

9.3.5 Environmentally Assisted Cracking of 304 Stainless Steel

This is a collaborative effort between LANL and UC Berkeley. The objective of this task is to determine the environmentally assisted cracking (EAC) susceptibility of 304 stainless steel (304SS) as a function of nitric acid, halide concentration, and sensitization level. This research, in conjunction with ongoing research on electrochemical corrosion of 304SS in environments relevant to plutonium stabilization and storage, will allow a comprehensive determination of safe operating conditions for processing radioactive salts in nitric acid processing streams. This research will also enhance core competency in corrosion science.

The proposed research is a natural extension of current research on electrochemical corrosion properties. Establishing electrochemical behavior is required for EAC testing. In addition to determining conditions that promote EAC susceptibility, a goal of this research is to examine the mechanisms that control cracking to understand EAC on a fundamental level.

Container materials for nitric acid process streams, as used to process actinide salts, are commonly comprised of 304SS. Although corrosion behavior of 304SS exposed to nitric acid and chloride environments have been studied separately, little work has focused on the combination of the two. The results of the proposed research will delineate a regime where the 304SS has a minimal overall corrosion rate in the combination environment.

Environmentally assisted cracking 304SS in acidic halide environments has been well documented; however, little study of nitric acid/halide environments has been reported. Since nitric acid is a powerful passivating agent, due to its strong oxidizing power, and since halide ions are powerful depassivators, the expected EAC tendency cannot be extrapolated from tests reported in the literature. It is, therefore, imperative to examine the EAC susceptibility of 304SS to relevant actinide processing environments.

9.3.6 Plutonium Chemistry in Aqueous Sulfate/Phosphate Media (SRTC)

Contaminated organic materials can be destroyed by "wet ash" oxidation in phosphoric or sulfuric acid with nitric acid, hydrogen peroxide, ozone, etc. In this task, the chemistry of plutonium in aqueous phosphate and sulfate media will be investigated with the goal of developing a process for recovering plutonium from these solutions.

Pu (IV) is reported to be quite soluble in 85% phosphoric acid at room temperature, but a literature search has revealed no quantitative data. The initial approach will be to determine the solubility of plutonium in 85% phosphoric acid from room temperature to the boiling point. A consideration of possible processes suggests that a precipitation method would require less equipment than solvent extraction and, thus, be more adaptable to small-scale operation. The study will explore possible complex phosphates with low solubility that may be useful in a recovery process. In addition, the study will pursue the possibility of decontaminating phosphate solutions for disposal as LLW.

In contrast to phosphoric acid, the solubility of plutonium sulfates has been studied extensively, and a number of complex sulfates have been prepared and characterized. In general, literature reports on the solubility of complex sulfates indicate that alkali metal ion-Pu (IV) sulfates should have low enough solubility at room temperature to provide a basis for a practical recovery method. However, decontamination of the solutions for disposal as LLW requires additional study.

For purposes such as simple dissolution, the destruction of organic materials by oxidation can be incomplete, since high production rate is a common goal and there is little incentive to continue the process once dissolution has been achieved. Incomplete oxidation leaves soluble organic acids, such as oxalic acid, acetic acid, etc., which may affect the plutonium chemistry. Therefore, the effect of partially-oxidized organic acids on plutonium chemistry will be investigated during development of a plutonium recovery process. The effect of organic acids on decontamination of glove box waste will also be considered.

9.4 Core Technology Requirements

There are no additional requirements identified at this time, other than those captured in the recommendations in Section 9.5.

9.5 Conclusions and Recommendations

Conclusions:

1. Since publication of the November 1995 R&D Plan, there has been a substantial effort to develop a Core Technology program. As such, a comprehensive picture of the important gaps in the underlying science of materials stabilization and storage, and of the core capabilities that need to be maintained is evolving. It is still unclear, however, which program elements contribute to the underlying science and which maintain core competencies.

2. The Vitrification task (see Section 9.2.3.4) is establishing a technology-specific capability. Therefore, it is not appropriate for this task to be part of the Core Technology program.

Recommendations:

1. The Lead Laboratory should develop a more comprehensive, systematic picture of the breadth of expertise that needs to be maintained, along with an assessment of directed R&D activities that currently meet some of these needs. The resulting gaps covered within the Core Technology program framework should also be systematically defined.

2. The Lead Laboratory should clearly identify those Core Technology activities that fill in important needs in the underlying science of stabilization and storage versus those activities that are primarily maintaining core competencies.

3. The Core Technology aspects of the Vitrification task should be captured within the Mineral Waste Forms task, and the rest of the Vitrification task should be moved to a technology-specific application.

10. PROPOSED DOE/RUSSIAN TECHNOLOGY EXCHANGE PROJECTS

On August 27 and 28, 1996, the PFA (primarily TAP members) met with a Russian delegation of senior technical scientists in Seattle, Washington. The purpose of this meeting was to openly discuss technical issues related to the post "Cold War" management of plutonium and other fissile materials with the intent of identifying areas for potential collaborative projects.

The TAP provided background information and a technical summary of the plutonium management issues that were unique or of concern to DOE. The Russian delegation provided a summary perspective of similar issues in their country. For the most part, the Russians have avoided the plutonium management/remediation issues that were created in the U.S. from an immediate shutdown of production facilities by continuing to operate until their facilities were made ready for shutdown. The Russian technical discussions summarized the techniques they believed could help solve the U.S. waste treatment and plutonium management issues. After several iterations, a preliminary list of proposed projects developed. The rather large number of potential topics underlined the success of the meeting, and the productivity of an open discussion and flexible agenda. Points of contact for continued discussions were established. The U.S. contact person is Dr. Yevgeny Macheret, DOE-ID, and the Russian contact is Dr. Albert Aloy, Khlopin Radium Institute.

The following projects were proposed by the Russian attendees at the Plutonium Workshop (the names and associations of each presenter appear in italics below each item):

1. Russia's experience in handling plutonium-bearing waste. Description of approaches and analysis of applied processes on various operational stages for both accumulated and current waste.

   Institute of Inorganic Materials: L. M. Borisov
   MAYAK Production Association: V. I. Kuzmenko

2. Latest technologies for plutonium-bearing waste processing. Evaluation of the application in the USA developing a Russian perspective of dealing with the problems at one of the sites (i.e., RFETS). Description of processes and results of their applications in Russia, their applicability to the RFETS waste, and recommendation on the waste processing.

   Institute of Inorganic Materials. L. M Borisov
   Institute of Chemical Technology: A. K Nardova
   MAYAK Production Association: V. I. Kuzmenko

3. Design and production of the equipment for various stages of plutonium waste processing, including (1) dissolution of the waste containing metal plutonium, (2) dissolution of slags and residues, and (3) pretreatment of the solution prior to chemical processing.

   Sverdlovsk Research Institute of Chemical Machine Building: I. M. Balakin
   MAYAK Production Association: V. M. Kuzmenko

4. Design and production of the equipment for processing solutions with various plutonium contents by means of oxalate precipitation with further generation of calcined precipitate. Additional experience in developing and operating small, simple, reliable, efficient machines.

   Sverdlovsk Research Institute of Chemical Machine Building: I. M Balakin
   MAYAK Production Association: E. G. Dzekun

5. Special materials for fabricating the equipment to handle nitric acid solutions containing fluoride and chloride ions.

   Sverdlovsk Research Institute of Chemical Machine Building: I. M. Balakin

6. Evaluation of high-temperature self-sustaining synthesis of mineral-like matrices for immobilizing plutonium and transuranic waste.

   Federal Scientific Center of Theoreticla and Experimental Physics: V. V. Drozdov

7. Plutonium immobilization into inorganic matrices (silica gel) from liquid waste by means of high-temperature absorption. A method has been developed to immobilize plutonium containing waste by impregnating into porous inorganic matrices (silica gel). The silica gel capacity for plutonium reaches 45%. Depending on the process conditions, the generated plutonium solid forms are suitable for:

   - interim safe storage and transportation with a possibility of plutonium recycling into the nuclear fuel cycle, if required

   - final disposal.

   Nuclear criticality safety of the plutonium solid forms can be ensured if neutron poisons are inserted into the inorganic matrices. The generated plutonium solid forms are chemically and thermally stable, as well as radiation resistant.

   Institute of Chemical Technology: A. K. Nardova

8. Stabilization and immobilization of plutonium-bearing waste by means of synthesis of superstable crystalline matrices, and plasmachemical pretreatment of the waste and its hot gas static pressing applied for the RFETS waste.

   Khoplin Radium Institute: E. B. Anderson

9. Thermodynamic and thermochemical studies of high-temperature processing of plutonium-bearing waste (e.g., at RFETS), evaluation of plutonium volatility depending on its forms.

   Khlopim Radium Institute: A. S. Aloy
   Institute of Silicare Chemistry (St. Petersburg)
   Institute of Inorganic Materials

10. Development of a matrix with a high chlorine content for transportation and storage of RFETS salts.

    Institute of Inorganic Materials: G. B. Borisov

11. Development of data base about compositions and generation of plutonium-bearing waste for one of the U.S. sites (i.e., RFETS). This work is required to evaluate applicability of the Russian technologies for U.S. needs.

12. Processes and equipment for plutonium-bearing waste evaporation to recover and recycle nitric acid, thereby reducing the waste volume.

    Institute of Inorganic Materials: L. M. Borisov
    Khlopin Radium Institute: B. Ya. Zilberman
    Research Institute of Chemical Machine Building: S. N. Filippov

13. Improvement and adjustment of the Russian technologies for dissolution of plutonium-bearing waste or various types, filtration of the generated solutions and their processing by means of oxalate precipitation at RFETS, thereby making the waste processing more cost-effective and efficient.

    Institute of Inorganic Materials: L. M. Borisov

14. Plutonium-bearing waste processing by means of sorbents that provide nuclear criticality safety and prevent any fire or explosion hazards in the presence of highly concentrated nitric acid, as well as under the conditions of sorbent heating and drying.

    Institute of Inorganic Materials: L. M. Borisov
    Institute of Chemical Technology: A. K. Nardova

15. Plutonium-bearing waste solidification and immobilization technology applying ceramic materials on natural clays.

    Khlopin Radium Institute: A. S. Aloy
    MAYAK Production Association: G. Medvedev.

GLOSSARY

NMSTG Milestone Number - The unique identifier for a milestone based on the 94-1 Implementation Plan.

R&D Need Date - Date when the technology needs to be ready for operation according to a *Site Integrated Stabilization Management Plan*. This includes training operations personnel, safety readiness, and facility readiness.

NMSTG Milestone Due Date - The date corresponding to the NMSTG Milestone Number.

DOE Site - The location responsible for completing the milestone.

Milestone Text - The action required to complete the milestone.

R&D Program - The R&D plan section number and title of a specific R&D project.

R&D Site - The organization responsible for completing the R&D.

End Use Site - The location where the technology is to be applied.

94-1 Milestone - The unique identifier for a milestone based on the *94-1 Implementation Plan.*

Technology Status - The technology status is identified as baseline, competitive alternative, or backup. Within this context, baseline refers to a technology designated for a particular use at a specific site; a competitive alternative is one of two or more technologies that are candidates for a baseline application; and a backup technology is one that could be pursued if baseline or competitive alternative approaches are not successful.

Technical Maturity - A technology can have significantly different technical maturity scores that range from 0 to 10, based on varying levels of readiness with respect to facilities, equipment, personnel, and safety at different end-use sites. A low technical maturity score represents a mature technology, whereas a high score represents a less mature technology. As a rule of thumb, the technical maturity score divided by two is an estimate of the maximum time (in years) to complete operational implementation of an R&D project.

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APPENDIX A

SELECTED ACRONYMS AND ABBREVIATIONS

APPENDIX A: SELECTED ACRONYMS AND ABBREVIATIONS

Am	americium
ANL	Argonne National Laboratory
ASTM	American Society of Testing and Materials
BNFL	British Nuclear Fuels Limited
CCO	Catalytic Chemical Oxidation
C/S	containment/surveillance
CDR	Conceptual Design Review
Cm	curium
DNFSB	Defense Nuclear Facilities Safety Board
DOE	Department of Energy
-DP	DOE Defense Programs
-EM	DOE Office of Environmental Management
-MD	DOE Office of Materials Disposition
-RL	DOE Richland Operations Office
-RW	DOE Office of Civilian Radioactive Waste Management
DOR	Direct Oxide Reduction
DOT	Department of Transportation
EAC	Environmentally Assisted Cracking
EDTA	ethylene diaminetetraacetic acid
ER	electrorefining
EXAFS	Extended X-Ray Absorption Fine Structure
FDC	Functional Design Criteria
FGE	Fissile Grams Equivalent
FY	Fiscal Year
GMODS	Glass Material Oxidation and Dissolution System
HCl	hydrochloric acid
HEPA	High-Efficiency Particulate Air
HEU	High Enriched Uranium
HLW	High-Level Waste
IAEA	International Atomic Energy Agency
INEL	Idaho National Engineering Laboratory
LANL	Los Alamos National Laboratory
LDRD	Laboratory Directed Research and Development
LLNL	Lawrence Livermore National Laboratory
LLW	Low-Level Waste
LMITCO	Lockheed Martin Idaho Technologies Company

LOI	Loss on Ignition
MEO	Mediated Electrochemical Oxidation
MPPF	Multi-Purpose Processing Facility (SRS)
MSE	Molten Salt Extraction
MSRE	Molten Salt Reactor Experiment
NEPA	National Environmental Policy Act
NMSTG	Nuclear Material Stabilization Task Group
NRC	Nuclear Regulatory Commission
NSR	New Special Recovery Facility (SRS)
ORNL	Oak Ridge National Laboratory
PERALS	Photon Electron Rejecting Alpha Liquid Scintillation
PFA	Plutonium Focus Area
PFP	Plutonium Finishing Plant (Hanford)
PGNAA	Prompt-Gamma Neutron Activation Analysis
PIC	Purification by Impurity Chlorination
ppm	parts per million
psi	pounds per square inch
PUREX	Plutonium Uranium Extraction
PuSPS	Plutonium Stabilization and Packaging System
PVC	Polyvinyl Chloride
RCRA	Resource Conservation and Recovery Act
R&D	Research and Development
RFETS	Rocky Flats Environmental Technology Site
RMRS	Rocky Mountain Remediation Services
RSM	Radioactively-contaminated Scrap Metal
SAIC	Scientific Applications International Corporation
SARP	Safety Analysis Report for Packaging
SEG	Scientific Ecology Group
SISMP	Site Integrated Stabilization Management Plan
SNF	Spent Nuclear Fuel
SRS	Savannah River Site
SRTC	Savannah River Technology Center (SRS)
SS&C	Sand, Slag, and Crucible
TAP	Technical Advisory Panel
TGA	Thermogravimetric Analysis
TRU	Transuranic
TRUPACT II	transuranic waste package
TRUW	transuranic waste

U	uranium
WAC	Waste Acceptance Criteria
WIPP	Waste Isolation Pilot Plant
wt%	weight percent
304SS	Stainless Steel, 304 alloy

APPENDIX B

PLUTONIUM FOCUS AREA
CHARTER

Department of Energy
Washington, DC 20585

October 2, 1995

MEMORANDUM FOR MR. JOHN M. WILCYNSKI
MANAGER
IDAHO OPERATIONS OFFICE

FROM: THOMAS P. GRUMBLY
ASSISTANT SECRETARY FOR
ENVIRONMENTAL MANAGEMENT

SUBJECT: Plutonium Focus Area

A Plutonium Focus Area is hereby chartered and will be managed by the Idaho Operations Office with support from Lockheed Martin Idaho Technologies (LMIT) and Argonne National Laboratory (ANL). Management of the focus area will not bring fissile or waste materials to Idaho.

The Plutonium Focus Area scope provides for peer and technical reviews of research and development in plutonium stabilization activities and includes the establishment of an Executive Panel to advise the Plutonium Focus Area Manager. The charter is attached. The Plutonium Focus Area Manager reports to the Director, Nuclear Materials Stabilization Task Group.

My congratulations to the Idaho National Engineering Laboratory for being selected to manage the Plutonium Focus Area.

Attachment

Charter
Plutonium Focus Area
September 1995

The Implementation Plan (IP) for Defense Nuclear Facilities Safety Board (DNFSB) Recommendation 94-1 identifies a requirement for research and development to meet short term technological needs plus a longer term core program. To assist the Nuclear Materials Stabilization Task Group (NMSTG) in meeting this requirement, a Plutonium Focus Area (PFA) is chartered under the DOE-ID field office with support from Lockheed Martin Idaho Technologies (LMIT) and Argonne National Laboratory (ANL).

The PFA will help identify and recommend solutions to both site specific and complex-wide technical and operational issues associated with plutonium stabilization and storage, while seeking opportunities for industry participation. The scope encompasses Plutonium-bearing materials within the DOE complex, excluding TRU wastes and final-form weapons components. The technology-specific program will be focused on treating and storing materials safely, with concomitant development of storage criteria and surveillance requirements, initially concentrated on the 3 and 8-year stabilization targets. The core technology program will augment the knowledge base about general chemical and physical processing and storage behavior to assure safe material storage until disposition.

The focus area activities will be coordinated with the Office of Technology Development (EM-50) programs and other focus areas to ensure effective use of resources and to prevent duplication of efforts. The PFA will apply systems engineering methods in pursuing the following activities:

1. Provide peer and technical review of research activities directed towards resolution of stabilization issues and identify needs for new technologies;

2. Encourage commercial industry to participate in partnerships with the DOE toward development of needed technologies;

The structure of the proposed PFA includes three key components:

1. *Executive Panel* will provide advice to the manager of the Focus Area in coordination and integration of plutonium stabilization and research activities. It will consist of federal employees representing the Offices of Fissile Materials Disposition, Defense Programs, Environmental Safety and Health, and the Albuquerque, Savannah River, Rocky Flats, and Richland Operations Offices. The purpose of the panel is to ensure that the interests of affected sites and program offices will be represented at the management level of the Focus Area.

2. A *Technical Advisory Panel* will provide technical evaluation of R&D and technology proposals and evaluation of the technical progress of the lead laboratory carrying out the NMSTG-sponsored R&D. The Technical Advisory Panel will support the NMSTG by

analyzing stabilization needs and identifying candidate topics for concerted R&D effort. This would include such aspects as preparing technical input to task statements that define the technical shortcomings upon which the lead laboratory is expected to focus and work, as well as evaluating the technical approach and content of proposals submitted by the lead laboratory addressing identified R&D tasking.

3. A *Systems Engineering* group will perform requirements and functional systems definition, trade studies, and systems performance evaluations for stabilization and research related activities.

APPENDIX C

SYSTEMS ANALYSIS METHODOLOGY AND RAW DATA

APPENDIX C: SYSTEMS ANALYSIS METHODOLOGY AND RAW DATA

This appendix discusses updates to the methodology used to quantify technical maturity and to assess programmatic risk for each of the technologies considered in this R&D plan. The methodology and its use was presented in its entirety in Appendix D of the November 1995 R&D Plan. The results captured in this appendix represent the state of technical maturity for each assessed technology at the time this plan was published. The maturity scores presented here will change as the technologies evolve; accordingly, these scores should be used with caution. The NMSTG plans to update these scores on a regular basis to keep program managers and other technical professionals apprised of the current status of each technology.

C-1. Technical Maturity Assessment Methodology Updates

Attachment 1 of this appendix shows changes to last year's methodology for Process Maturity Scoring and Consequences of Failure Assessment.

Chart 2 of Attachment 1 show changes to the Process Maturity scale (underlined and italicized), which clarify that prototype status for a process is achieved only if the prototype is demonstrated at the end-use site. This is consistent with the interpretation of this maturity level used by the NMSTG-chartered trade studies during FY 1996.

Charts 3 and 4 of Attachment 1 show an updated to assessment of the consequence of failure. The November 1995 methodology calculated this score as a simple average of the performance and schedule consequence components. The schedule consequences component was a discrete function of the priority of the project (3 vs. 8 years) and the status (Baseline vs. Backup). This year, the performance component is dropped, and the schedule component is a continuous function of the R&D Need Date derived from the Site Integrated Stabilization Management Plans (SISMP), which are traceable to the 94-1 Implementation Plan Milestone Dates. The example curves on Chart 4 of Attachment 1 show the overall risk scores for theoretical cases where a technology scheduled to be completed in 5 years remains behind schedule for a fixed number of years as the program progresses. An on-time project never exceeds an overall programmatic risk of 0.25. Consequently, a project whose score does not exceed 0.25 should be considered low risk. Conversely, a project with a score greater than 0.25 should be reviewed by management to determine if any further action is necessary.

C-2. Results

Results of the technical maturity assessments conducted by the TAP are presented in Attachment 2. Baseline and competitive alternative technologies with an overall programmatic risk of 0.40 or greater are high risk and require special management attention. Those baseline and competitive alternative technologies with an overall programmatic risk score from 0.25 to 0.40 are medium risk and require close tracking.

NMSTG Technical Maturity Assessment Changes

Appendix C, Attachment 1, Chart 1

Process Maturity (PM) Scale

Level	Maturity Assessment Criteria
10	No currently identified solutions that meets requirements
9	Design concept and/or technology application formulated.
8	Cold feasibility demonstrated
6	Hot feasibility demonstrated
5	End-to-end design (flowsheet) complete
4	Cold prototype demonstrated *at end-use site*
2	Hot prototype demonstrated *at end-use site*
0	Process integrated into operations

Appendix C, Attachment 1, Chart 2

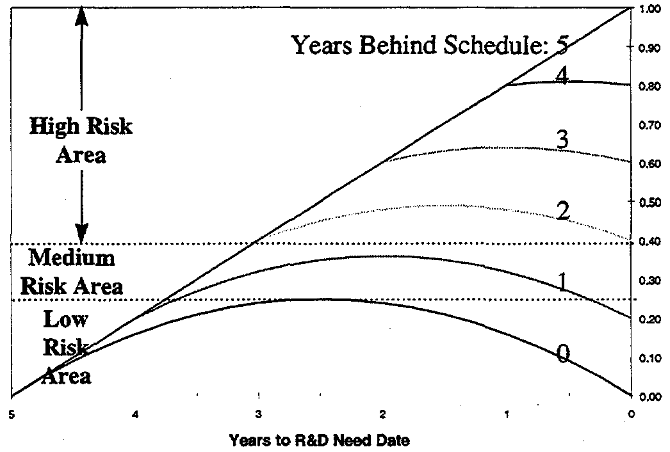
New Basis for Consequences of Failure Assessment

- Technical Maturity (TM) score is proportional to time to completion for an R&D project
  - Typically, the TM score divided by two is the number of years remaining
- R&D Need Dates can be extracted from end-user project plans
  - Consequence of Failure is proportional to the time remaining until the Need Date

Appendix C, Attachment 1, Chart 3

Schedule-Driven Consequence of Failure Calculation

- Cf = MIN {1, MAX { 0, 1- (Years from Now to Need Date / 5) }}
- TM = 2 * MIN {5, Years Behind Schedule + Years to R&D Need Date}
- Overall Programmatic Risk Score = TM * Cf / 10



Appendix C, Attachment 1, Chart 4

Table C-1. Technical Maturity/Raw Data

Technology	R&D Site	End-Use Site	RM	PM	EQ	FAC	SAFT	PER	SCH	R&D Plan Section	R&D Need Date	Status	Tech Maturity Score	Cs	Overall Programmatic Risk Score
PuSPS - Stabilization	NMSTG	RFETS	2	5	4	7	0	1	2	4.1.2.1	Oct 1997	Baseline	3.65	0.78	0.29
Extraction Chromatography	HAN	HAN	3	2	1	3	5	1	2	4.2.2.1	Jun 1997	Baseline	2.67	0.85	0.23
Vertical Calciner	HAN	HAN	2	2	1	7	5	1	2	4.2.2.2	Jun 1997	Baseline	3.44	0.85	0.29
Precipitation - Oxalate/Hydroxide	LANL	RFETS	0	5	0	0	9	1	2	4.2.2.3	Nov 1996	Baseline	3.09	0.97	0.30
Pu Solution Vitrification	SRS	SRS	4	6	6	3	9	5	4	4.2.2.4	Oct 1999	Backup	5.70	0.38	0.22
Pu Solution Ceramification	RFETS	RFETS	3	4	1	3	9	1	0	4.2.3.1	Nov 1997	Backup	3.79	0.77	0.29
Pyrochemical Salt Oxidation	LANL	RFETS	0	5	0	3	5	1	4	4.3.1.2.1	Aug 1997	Baseline	3.02	0.82	0.25
Pyrochemical Salt Distillation	LANL	RFETS	3	5	1	3	5	1	4	4.3.1.2.2	Aug 1997	Baseline	3.44	0.82	0.28
Salt Scrub/Oxidation	LLNL	RFETS	3	2	2	2	5	3	2	4.3.1.3.1	Aug 1997	Baseline	2.72	0.82	0.22
Salt Filtration	LLNL	RFETS	6	6	0	2	5	3	2	4.3.1.3.2	Aug 1997	Backup	3.35	0.82	0.27
Salt Washing	LLNL	RFETS	8	9	4	10	10	3	10	4.3.1.3.3	Aug 1997	Backup	8.23	0.82	0.67
Electrochemical Scrubbing	RFETS	RFETS	4	5	0	3	9	1	1	4.3.1.3.4	Aug 1997	Backup	3.93	0.82	0.32
GMODS	ORNL	RFETS	4	9	8	7	9	1	4	4.3.1.3.5	Aug 1997	Backup	7.49	0.82	0.61
SS&C Stabilization	SRS	SRS	0	0	0	0	0	1	0	4.3.2.2.1	Jan 1997	Baseline	0.02	0.93	0.00
Calcine for Shipment to SRS	RFETS	RFETS	6	6	1	3	9	1	4	4.3.2.2.1	Sep 1997	Baseline	4.70	0.80	0.38
Carbonate Oxidation	LANL	RFETS	4	6	1	5	3	4	9	4.3.2.2.2	Sep 1997	Backup	4.14	0.80	0.33
Nitric Acid Soluble Bags	SRS	SRS	0	4	0	0	0	1	0	4.3.2.2.3	Jan 1997	Baseline	0.86	0.93	0.08
Cementation	HAN	HAN	3	4	3	3	7	1	4	4.3.2.2.4	May 1998	Baseline	4.07	0.67	0.27
Vitrification for WIPP	RFETS	RFETS	6	4	1	3	9	3	4	4.3.2.3.1	Sep 1997	Backup	4.33	0.80	0.35
Can-in-Canister	SRS	SRS	1	2	1	0	10	1	0	4.3.2.3.2	Sep 1997	Backup	2.81	0.80	0.23
RFETS Vitrification for Can-in-Canister	RFETS	RFETS	6	9	9	5	9	2	6	4.3.2.3.3	Sep 1997	Backup	7.58	0.80	0.61
Electrochemical Scrubbing	RFETS	RFETS	4	5	0	3	9	1	1	4.3.2.3.4	Sep 1997	Backup	3.93	0.80	0.31
SS&C Hydrous Cementation	RFETS	RFETS	3	4	3	7	7	3	4	4.3.2.3.5	Sep 1997	Backup	4.95	0.80	0.40
Polycube Pyrolysis	LANL	HAN	4	5	3	5	5	3	4	4.3.4.2.1	Jul 1999	Baseline	4.40	0.43	0.19
Pyrolysis	LANL	RFETS	6	6	6	3	10	5	6	4.3.4.2.2	Nov 1997	Backup	6.19	0.77	0.47
Catalyzed Chemical Oxidation	RF/LA/SR/LL	RFETS	4	8	4	3	10	3	6	4.3.4.2.3	Nov 1997	Backup	6.00	0.77	0.46
Nitric/Phosphoric Digestion	SRS	RFETS	4	5	6	3	10	3	6	4.3.4.2.4	Nov 1997	Backup	5.79	0.77	0.44
Hydrothermal	LANL	RFETS	4	8	4	3	10	5	6	4.3.4.2.5	Nov 1997	Backup	6.05	0.77	0.46
Mediated Electrochemical	LANL	RFETS	4	2	3	3	10	3	6	4.3.4.2.6	Nov 1997	Baseline	4.53	0.77	0.35
Mediated Electrochemical (Destruction)	LANL	RFETS	6	6	6	3	10	3	6	4.3.4.2.6	Nov 1997	Backup	6.14	0.77	0.47
IX Resin Denitration	LANL	RFETS	3	5	4	0	5	1	4	4.3.4.2.8	Nov 1997	Backup	3.44	0.77	0.26
Cryogenic Size Reduction	LANL	RFETS	6	8	6	3	7	3	6	4.3.4.2.9	Nov 1997	Baseline	5.93	0.77	0.45
Steam Reformation	SEG	RFETS	4	5	6	5	10	3	6	4.3.4.3	Nov 1997	Backup	6.21	0.77	0.48
Pretreatment/Thermal	LLNL	LLNL	4	2	0	0	0	0	0	4.3.5.2.1	Mar 1997	Baseline	0.70	0.90	0.06
MEO for Ash	SRS	SRS	3	2	1	3	10	1	6	4.3.5.2.2	Feb 1998	Backup	4.00	0.72	0.29
Silver Persulfate Dissolution	HAN	HAN	4	4	4	3	9	1	6	4.3.5.2.2	Apr 1998	Baseline	4.91	0.68	0.34
MPPF Vitrification for Can-in-Canister	SRS	SRS	4	5	4	5	10	1	4	4.3.5.2.3	Feb 1998	Backup	5.60	0.72	0.40
RFETS Vitrification for Can-in-Canister	RFETS	RFETS	6	9	9	5	9	2	6	4.3.5.2.3	Feb 1998	Backup	7.58	0.72	0.54
Ceramification for WIPP	RFETS	RFETS	8	8	8	5	10	1	6	4.3.5.2.4	Feb 1998	Backup	7.49	0.72	0.54
Vitrification for WIPP	RFETS	RFETS	6	2	1	5	9	2	6	4.3.5.3.1	Feb 1998	Baseline	4.44	0.72	0.32
Can-in-Canister	SRS	SRS	1	2	1	0	10	1	0	4.3.5.3.3	Feb 1998	Backup	2.81	0.72	0.20
Scrub Alloy Processing	SRS	SRS	1	3	1	0	0	1	2	4.3.6.2.1	Jul 1997	Baseline	1.07	0.83	0.09
Impurity Chlorination	LLNL	RFETS	3	5	0	2	0	0	2	4.3.6.3.1	Jul 1997	Backup	1.81	0.83	0.15
Am/Cm Vitrification Process	SRS	SRS	1	4	6	3	7	5	2	5.2.1	Jun 1999	Baseline	4.51	0.45	0.20
Am/Cm Product Test	SRS	SRS	1	2	0	0	0	1	0	5.2.1	Jun 1999	Baseline	0.51	0.45	0.02
Neptunium Flowsheet	SRS	SRS	8	9	6	3	7	1	2	5.2.2	Apr 2000	Baseline	5.95	0.28	0.17
Pu-242 Stabilization	SRS	SRS	0	0	0	0	0	0	0	5.2.3	Oct 1996	Baseline	0.00	0.98	0.00
Trapping of Uranium Hexafluoride	ORNL	ORNL	0	1	1	0	0	0	0	6.2.1	Jun 1996	Baseline	0.42	1.00	0.04
Direct Fluorination of MSRE Salts	ORNL	ORNL	1	2	0	3	9	1	3	6.2.2	Sep 1999	Comp	3.23	0.40	0.13
Electrochemical Treatment of MSRE Salts	ANL	ORNL	4	9	8	7	9	1	4	6.2.3	Sep 1999	Comp Alt	7.49	0.40	0.30
GMODS	ORNL	ORNL	4	9	8	7	9	1	4	6.3.1	Sep 1999	Comp	7.49	0.40	0.30
Packaging with Getter	ORNL	ORNL	3	5	1	3	9	1	4	6.3.2	Sep 1999	Comp	4.28	0.40	0.17
Charcoal Treatment	ORNL	ORNL	3	5	1	3	3	1	4	6.3.3	Sep 1997	Baseline	3.02	0.80	0.24
PuSPS - Packaging	NMSTG	RFETS	2	5	1	7	0	1	2	7.1.2.1	Oct 1997	Baseline	3.02	0.78	0.24
Electrolytic Decontamination	LANL	LANL	3	2	1	5	5	1	2	7.1.2.2	Oct 1997	Baseline	3.09	0.78	0.24
Bagless Transfer System - FB	SRS	SRS	2	4	3	5	0	5	0	7.1.2.3	Sep 1997	Baseline	2.77	0.80	0.22
Dustless Oxide Transfer	LLNL	LLNL	0	5	4	0	0	3	2	7.1.2.4	Apr 1998	Baseline	2.09	0.68	0.14
Pipe Component	RFETS	RFETS	4	5	0	3	9	1	1	7.1.2.6	Aug 1997	Baseline	3.93	0.82	0.32
Pu239 Standard Container	NMSTG	RFETS	1	5	1	5	0	1	2	7.1.2.7	Oct 1997	Baseline	2.53	0.78	0.20
Digital Radiography	LANL	RFETS	3	2	1	0	0	1	4	7.2.2.1	Oct 1998	Comp	1.14	0.58	0.07
Tomography	LANL	RFETS	6	9	6	0	0	2	4	7.2.2.2	Oct 1998	Comp	3.88	0.58	0.23
Laser Sampling	LANL	SRS	3	5	3	3	10	2	4	7.2.2.3	Oct 1998	Backup	4.93	0.58	0.29
Acoustic Resonance	LANL	RFETS	6	8	8	0	0	2	6	7.2.2.4	Oct 1998	Comp	4.23	0.58	0.25
Pressure Sensitive Devices	LANL	RFETS	4	8	8	0	0	2	4	7.2.2.5	Oct 1998	Comp	3.95	0.58	0.23
		Weights	0.3	0.9	0.9	0.9	0.9	0.1	0.3						

APPENDIX D

LIST OF PFA MEMBERS

APPENDIX D: LIST OF PFA MEMBERS

This Research and Development Plan was prepared for the Plutonium Focus Area by the PFA Technical Advisory Panel. The TAP consists of the following members:

Dr. Douglas C. Crawford (TAP Chairman)
Manager, Materials Technology Section
Engineering Division
Argonne National Laboratory

Mr. Mark C. Bronson
Associate Program Leader
Strategic Materials Program
Lawrence Livermore National Laboratory

Mr. Louis H. Rodgers
Consultant
Westinghouse Hanford Company

Dr. William A. Averill
Principal Materials Scientist
Rocky Flats Environmental Technology Site
Kaiser Hill Company, L.L.C.

Dr. Rowland E. Felt
Office of Environment, Safety and Health
U.S. Department of Energy

Dr. Nathan A. Chipman (TAP Deputy Chairman)
Consulting Engineer
Idaho National Engineering Laboratory
Lockheed Martin Idaho Technologies

Dr. Clinton R. Wolfe
Manager Chemical Process Technology Department
Savannah River Technology Center
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Dr. Randall M. Erickson
Program Manager for Disposition
and Immobilization
Los Alamos National Laboratory

Dr. Charles W. Forsberg
Research Scientist
Oak Ridge National Laboratory

An ad hoc committee was formed to correlate and produce this document under the direction of the TAP. The committee consisted of the following members:

Dr. Alenka Brown-VanHoozer
(R&D Plan Coordinator)
Research Scientist
Argonne National Laboratory

Dr. C. Robert Kenley
NMSTG Systems Engineer
Lockheed Martin Idaho Technologies

Dr. Randall M. Erickson
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Los Alamos National Laboratory

Dr. Nathan A. Chipman
Consulting Engineer
Idaho National Engineering Laboratory
Lockheed Martin Idaho Technologies

Dr. Bernard R. Kokenge
Consultant
BRK Associates, Inc.

APPENDIX E

STATUS OF NOVEMBER 1995 R&D PLAN RECOMMENDATIONS

APPENDIX E: STATUS OF NOVEMBER 1995 R&D PLAN RECOMMENDATIONS

Table E-1. Status of November 1995 R&D Plan Recommendations

Recommendations	Comments	Status
Develop standards for stabilizing and storing each of the special isotopes (Pu-238, Pu-242, and isotopes of Np and Am/Cm).	Site-specific actions are underway.	Underway
Develop analytical methods for determining moisture content, gas composition from radiolysis, and reactive metals present in pyrochemical salts.	Trade study recommendations obviate the need for this requirement.	Closed
Develop a flowsheet for stabilizing neptunium solutions at SRS.	Flowsheet under development and will meet need date.	Underway
Develop large-volume storage containers for low-assay (<10%) plutonium residues.	Trade study recommendations obviate the need for this requirement.	Closed
Evaluate the need for corrosion-resistant containers for halide salts and other corrosive residues.	Trade study recommendations obviate the need for this requirement.	Closed
Develop a surveillance system for monitoring Am/Cm and Pu-238 in storage.	Activity underway at SRS.	Underway
Complete the timely development and startup of the vertical calciner at Hanford.	Calciner development is proceeding with close tracking by NMSTG.	Underway
Continue the concurrent development of multiple processes for stabilizing all categories of hazardous combustibles containing plutonium.	Combustible trade study recommended specific technologies to pursue and revised schedule.	Closed
Continue the development of the modular concept at LANL as a means to eliminate startup of processing facilities at various sites.	A LANL team was to develop the modular concept.	Underway
Continue the development of technologies to address U-233 criticality safety issues at the MSRE facility.	Multiple technologies are being evaluated to address criticality safety.	Underway
Continue development of the bagless transfer system as a baseline technology and of electrolytic decontamination as a close-coupled backup technology for plutonium packaging.	The PuSPS is on schedule for October 1997 delivery and electrolytic decontamination is being developed at LANL.	Underway
Complete development of digital radiography and/or digital radiography/tomography for monitoring plutonium packages in storage.	Under development at LANL.	Underway
Evaluate the institutional issues—particularly, negative public perceptions—that prevent deployment of incineration technology, which the RC believes to be the most viable method for treating combustible residues.	Incineration was evaluated by the combustibles trade study, and it was determined that incineration cannot be developed to meet the need dates.	Closed
Maintain an ongoing core technology program to support stabilization technologies, to address unforeseen problems associated with long-term storage, and to provide technology for predicting the long-term behavior of nuclear materials.	Core technology program is underway at LANL.	Underway
Provide R&D to identify and support the development and implementation of an interim storage standard for residues.	Trade study recommendations obviate the need for this requirement.	Closed
Continue basic R&D studies of plutonium oxide behavior in support of DOE-STD-3013 (>50 and <80 wt % plutonium.)	Studies underway at LANL.	Underway