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Technologies for Aluminum-Base Spent
Nuclear Fuel (U)

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0	All	Original	12/31/96
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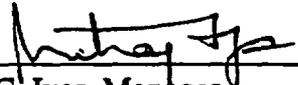
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Title: Task Plan for Development of Dilution Technologies for Aluminum-Base Spent Nuclear Fuel (U)

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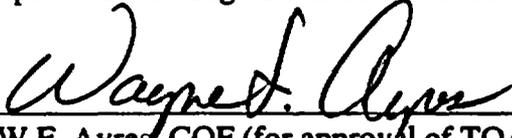
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1.0 Summary

This memorandum provides the task technical and quality assurance plans for the scientific investigation activities necessary to develop melt-dilute and press-dilute technologies for the development of an aluminum spent nuclear fuel (SNF) waste form suitable for disposal in a geologic repository. The focus of this program will be the development of the melt-dilute technology. In addition, the program will also analyze the feasibility of press-dilute technology for developing a diluted waste form. The melt-dilute technology development will focus on two primary issues: (a) melt-dilute process development and (b) diluted aluminum SNF waste form development.

The melt-dilute process development will involve the following scientific investigations:

- (1) review of existing literature to establish acceptable melting and casting approaches for aluminum-uranium alloys,
- (2) development of dilution methods,
- (3) bench-scale testing to determine the behavior of alloys of several different aluminum-uranium compositions, evaluate melt-dilute concepts, and determine alloy characteristics as a function of uranium content, and
- (4) determine the fission product release during the melt-dilution process.

These tasks will culminate with the development of a small-scale furnace and the demonstration of the melt-dilute process for a Materials Test Reactor (MTR) type surrogate fuel.

The aluminum SNF waste form development will include the definition of the optimum alloy compositions to meet the desired performance characteristics such as dissolution, durability, etc.

The press-dilute method will also be analyzed and compared to the melt-dilute process as part of this task plan. If significant advantages warrant, development of a separate technical task plan will be written for the press-dilute method. The task plan for canister and waste form qualification for direct disposal of SNF is covered under a separate test protocol.

2.0 Background

The Research Reactor Spent Nuclear Fuel (SNF) Task Team was established by the DOE - Office of Spent Fuel Management to assist DOE in developing a technical strategy for the interim management and ultimate disposition of the foreign and domestic aluminum-based research reactor spent fuel under DOE's jurisdiction. The team issued a report that evaluated and compared alternatives to reprocessing and concluded that direct (co-disposal package) of the fuel with defense high-level waste canisters was the alternative with the highest score [1]. Under this approach, many of the spent fuel assemblies currently in wet storage at SRS will be prepared for storage in a road-ready package and stored at SRS until a geologic repository is ready for receiving the SNF.

Under the direct disposal approach, the SNF would be dried and placed in a canister for interim storage, then transported to the repository and placed in a waste package for disposal. The direct disposal in co-disposal package option would place the SNF in waste packages which contain High Level Waste glass logs. Major issues associated with the direct disposal of highly enriched uranium (HEU) research reactor fuel in a repository include nonproliferation and criticality control. The Task Team evaluated HEU dilution methods for purposes of reducing the effective enrichment of the waste packages, thereby improving package efficiency, rendering the packages less attractive for diversion, and reducing the need for engineered criticality controls. Melt- and press-dilute methods were recommended by the task team.

Most of the early FRR fuels were manufactured with an aluminum-uranium alloy core that contained 90-93% enriched uranium (^{235}U). By diluting the ^{235}U content of the spent fuel with depleted uranium, enrichments can be lowered. Low enrichments (2 -20%) make the uranium not weapons capable and reduce and perhaps even eliminate criticality concerns during storage when the fuel corrodes and the enriched uranium is rearranged into various configurations. The addition of the diluents increases the volume of the material to be disposed, but the increase in the allowable amount of SNF in each canister may result in a potential decrease in the total number of canisters required.

The melt-dilute process would involve melting the SNF and achieving dilution through the addition of depleted uranium. The basic melt-dilute process is outlined in Figure 1. The spent fuel assembly will be characterized to determine its uranium composition before melting. The characterized assembly will be melted and depleted uranium metal or oxide will be added to the melt to reduce the ^{235}U content to less than 20 wt%, preferably to less than 2 wt%. The alloy will be analyzed for uranium composition prior to encapsulation in a canister. The canister will be sealed and stored in a road-ready condition at SRS until shipment to the final repository.

Because of density differences between uranium compounds and aluminum, the melt would be mixed by stirring to achieve homogeneity. Due to the homogeneous nature of the product, there is a potential reduction in pre-storage characterization requirements and associated costs and repository storage qualification costs. The melt-dilute process is also compatible with co-disposal technology where the canisters are co-loaded with HLW glass logs in repository overpacks, if this is a desirable option.

In the press-dilute option, the SNF assemblies would be flattened in a mechanical press to eliminate air gaps. Depleted uranium would then be pressed around or between the flattened fuel to reduce the effective enrichment. Several flattened assemblies could be placed into one unit. This unit would be placed into an interim storage canister and later shipped to the repository for disposal.

Under the dilution technology development strategy, several program steps are required for successful completion as shown in Figure 2. Criteria will be developed for the melting, casting, and sealing of SNF through extensive process development. Functional requirements for a treatment system facility will be developed from these criteria. None of the alternative waste forms for disposal, other than the borosilicate glass which is obtained from the base case of

reprocessing, is qualified for the Federal repository. However, activities at the national level are in progress to qualify metallic waste forms for this repository and it is anticipated that direct disposal and co-disposal waste forms including diluted waste forms will be reviewed for qualification.

This technical task plan outlines the approach to be followed for developing and evaluating melt-dilute and press-dilute technologies. The focus of the work will be performed on melt-dilute development. Advantages and disadvantages of press-dilution will be evaluated and compared to melt-dilution. If significant advantages warrant development of press-dilution, a separate technical task plan will be written.

3.0 Reporting and Activity Milestones

The estimated period of performance for development of the dilution process technology is from January, 1997 to January, 1999. The functional requirements for a facility to provide these technologies will be developed by 1999. Each year the task plan will be revised to reflect any changes in scope or schedule. The reporting of major milestones for FY96-97 are:

1. Issue Task Technical Plan for the Dilution Program
Date: December 31, 1996
2. Issue status report on bench-scale studies for melt-dilution technology
Date: October 31, 1997
3. Issue report on the preliminary requirements for the melt-dilution process
Date: October 31, 1997
4. Develop and construct small-scale test furnace for preliminary demonstration of melt-dilution technology
Date: October 31, 1997.

4.0 REFERENCES

1. "Technical Strategy for the Treatment, Packaging, and Disposal of Aluminum-Based Spent Nuclear Fuel," Volume 1 of a Report of the Research Reactor Spent Nuclear Fuel Task Team prepared for the Department of Energy - Office of Spent Fuel Management, May 1996.
2. Technical Task Request, EF&RFSP/SNFP 97-02.

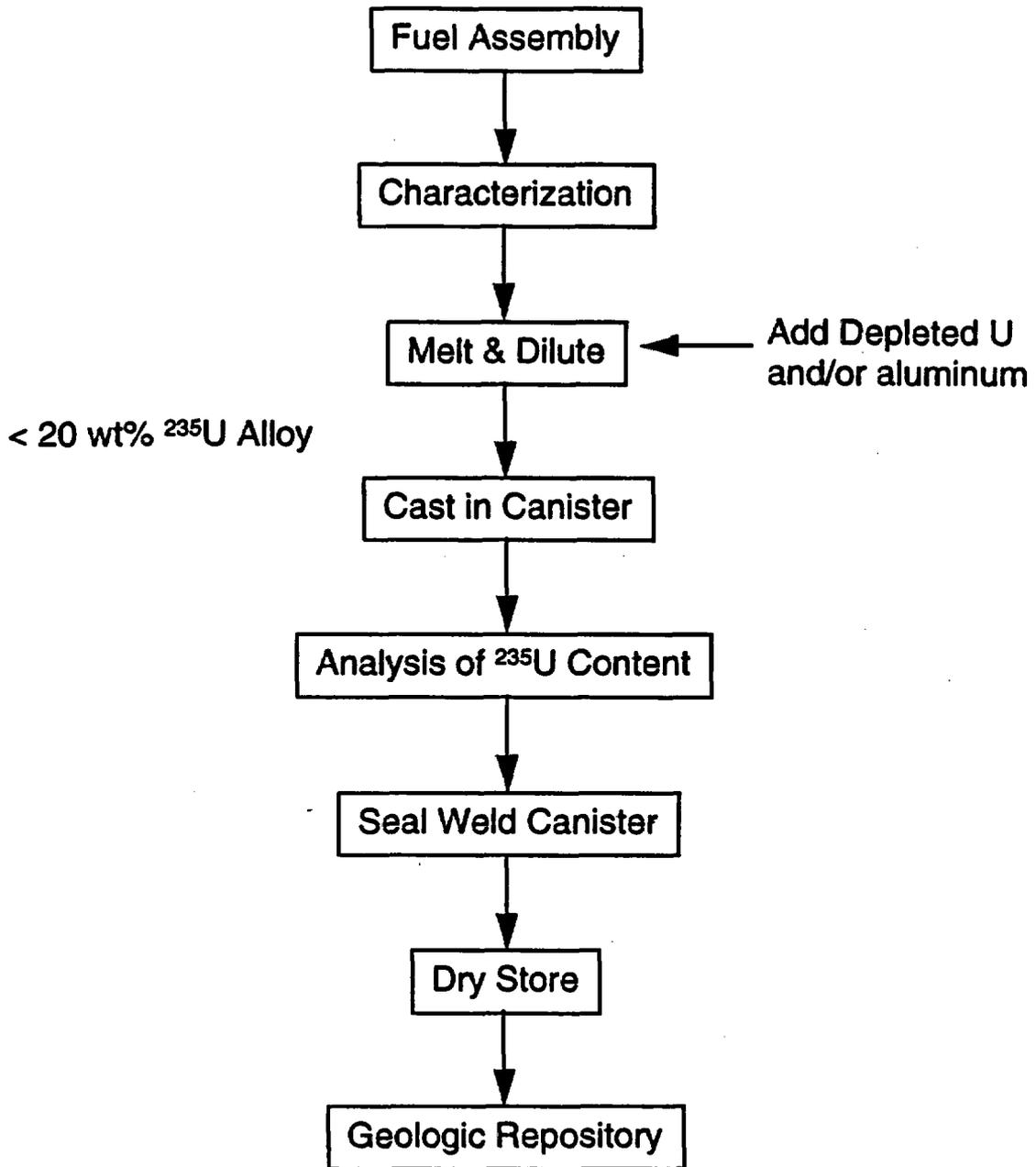


Figure 1. Melt-Dilute Process for Spent Fuel Storage

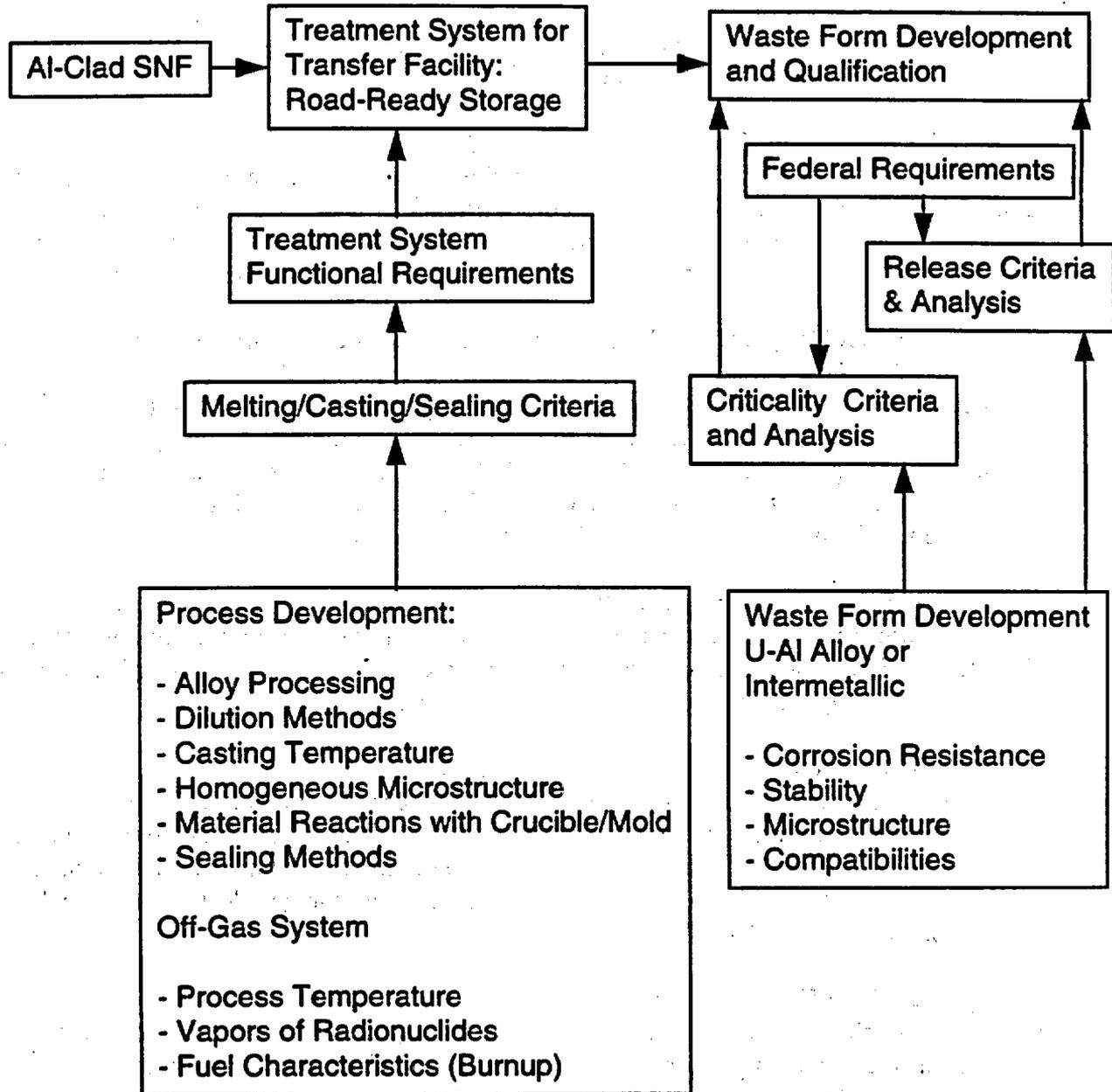


Figure 2. Dilution Technologies Development Strategy

APPENDIX A TASK TECHNICAL PLAN

The following technical tasks are identified as scientific investigations to determine applicable concepts for the melt-dilution program.

Task 1.0 Literature Survey

Literature will be reviewed and used to help define melting and casting techniques for aluminum-uranium alloys at temperatures above about 800 °C.

Subtask 1.1 Acceptable Composition of Dilute Alloys

FRR spent fuel data will be reviewed and desirable alloy-composition ranges determined for diluted fuel. From the results, a specific composition(s) will be selected for the melt-dilute process which allows melting and casting of diluted spent fuel alloys into a waste form that should be acceptable for road-ready and final storage. (Start 1/1/97; Complete 4/30/97)

Subtask 1.2 High Temperature Compatibility of Uranium-Aluminum Alloys with Crucible and Mold Materials

The liquidus temperature for aluminum-uranium alloys varies from 643 °C for the Al-U eutectic to 1620 °C for the intermetallic compound UAl_2 . If the selected alloy compositions require processing at temperatures above ~1000°C molten metal-mold interactions may be significant; thus, acceptable materials and methods for handling the alloy must be determined and applied to the process. (Start 1/22/97; Complete 3/31/97)

Task 2.0 Melt-Dilute Development Activities:

Spent fuel compositions will be made and depleted uranium added to simulate dilution of the ^{235}U content. Initially, scoping tests will be performed to evaluate melting/casting behavior of alloys of several different aluminum-uranium compositions.

Subtask 2.1 Uranium-235 Dilution methods (2-20%)

This task includes the evaluation of different methods for uranium dilution of the molten alloy. Depleted uranium may be added to the melt using common metal and/or uranium oxide additions. Studies are necessary to determine dissolution rates, methods for addition, and waste form characteristics so an optimum dilution technique can be selected for the melt-dilution process. (Start 3/13/97; Complete 7/31/97)

Subtask 2.2 Waste Form Casting Techniques

Graphite crucibles or other suitable materials will be required if the dilution process is accomplished at high temperature. The molten alloy may be melted and solidified in a crucible or directly cast into a mold which will be sealed and will serve as a secondary container for the waste form, or cast into a permanent mold. The use of a permanent mold would require that the casting be removed from the mold after solidification. The alloy casting would then be placed in a secondary container to provide the waste form. Studies are needed to determine the appropriate method for the melt-dilution process. (Start 2/18/97; Complete 6/30/97)

Subtask 2.3 Furnace Requirements

Furnace requirements for melting and casting of the spent fuel alloy will be evaluated. Various melting technologies including induction and resistance melting will be evaluated for process efficiency, waste storage production, flexibility, and ease of approach. Stirring of the melt will be evaluated to determine if stirring is necessary to obtain a homogenous solution and reduce alloy segregation.

(Start 6/1/97; Complete 9/1/97)

Subtask 2.4 Casting Methods

Casting methods will include either direct melting and solidification in a crucible or pouring the alloy into a casting mold using either top or bottom pouring techniques. Other methods such as high-pressure melting and casting or a closed melting and casting system will be evaluated in an effort to reduce the cost for an expensive off-gas system.

(Start 5/5/97; Complete 7/1/97)

Task 3.0 Bench-Scale tests

Bench-scale tests will be conducted to evaluate melt-dilute concepts and to determine alloy characteristics as a function of uranium content. Metallography will be used to determine homogeneity of the waste form and will be a primary evaluation technique for selection and quantifying a composition for the long term storage tests.

Subtask 3.1 Develop Bench-Scale Apparatus

The bench-scale apparatus will be configured to develop and evaluate melt-dilute concepts. Existing equipment in the SRTC Materials Laboratory will be modified, as required, to implement the bench-scale tests.

(Start 1/1/97; Complete 1/31/97)

Subtask 3.2 Feed Samples for Bench-Scale Studies

Extruded rods ~0.700 inch in diameter and containing depleted aluminum-uranium alloy will be used to simulate spent fuel in the bench-scale tests to develop melt-dilute process parameters. The rod samples will be fabricated in the Fabrication Laboratory at SRTC using the 520 ton extrusion press. Several different alloy compositions will be prepared. These samples will be used for the bench-scale melt-dilute experiments.

(Start 2/1/97; Complete 4/30/97)

Subtask 3.3 Eutectic Compositions and Intermetallic Compounds

The uranium-aluminum system contains several eutectic, peritectic, and intermetallic compound reactions. These reactions occur at temperatures varying from about 643 °C to 1620 °C. The characteristics of the waste form will be controlled to a significant extent by the size, shape, number and distribution of the microstructural elements. Therefore, cooling rates, phase transformations and composition will play an important role in the solidification of the molten alloy. An appropriate waste form will be developed for long term storage of spent nuclear fuel. (Start 1/1/97; Complete 8/1/97)

Subtask 3.4 Microstructure Documentation

Bench-scale studies will be performed to correlate microstructural properties of the various selected alloys. Additionally, homogeneity of the various castings will be evaluated. (Start 2/1/97; Complete 9/1/97)

Subtask 3.5 Corrosion Studies

The test protocols for the corrosion studies will be developed under a separate program (*Task Plan for Engineering Test Protocol for Metallic Waste Forms, SRT-MTS-96-2064, To be issued*). The testing per these protocols will be performed under this program.

Corrosion of UAl alloy will occur in the event that the primary canister is breached. Currently this is assumed to occur by corrosion after 1000 years. Corrosion of the core will produce aluminum and uranium oxides which will eventually accumulate at the bottom of the storage facility. For diluted compositions of 20 wt% ^{235}U , there is a limit to the amount that can be safely stored in a given geometry to prevent criticality. However, by diluting to less than ~2% ^{235}U to form intermetallic compounds, criticality becomes significantly less likely. Studies will be done to determine the corrosion behavior of these alloys with 20 wt% and less ^{235}U under storage conditions expected in the repository. (Start 3/1/97; Complete 6/20/98)

Subtask 3.6 Dissolution Studies

The test protocols for the leaching studies will be developed under a separate program (*Task Plan for Engineering Test Protocol for Metallic Waste Forms, SRT-MTS-96-2064, To be issued*). The testing per these protocols will be performed under this program.

If the alloy is exposed to J13 ground water conditions, radionuclides may be leached from the alloy oxide. Redistribution and accumulation of the radionuclides including ^{235}U could occur and present hazardous conditions. To determine the leach rate for hazardous radionuclides, leaching studies will be performed for simulated alloy compositions. (Start 6/1/97; Complete 6/20/98)

Subtask 3.7 Alloy Additions to Enhance Properties and Criticality resistance

When the spent fuel is melted, alloying elements can be easily added to the melt. Per customer requests, studies will be done to determine if the corrosion resistance can be improved, and if the propensity for criticality can be reduced by additions of nuclear poisons. (Start 9/1/97; Complete 2/28/98)

Task 4.0 Fission Product Release

Fission products will be released when the spent fuel is melted. The FRR fuel has been stored in wet basins for many years, and decay of radioactive elements has occurred and will continue throughout the storage period.

Subtask 4.1 Determination of Radioactive Fission Products

Data from the ORIGEN or similar validated computer code will be used to determine expected fission product inventory for spent FRR fuels. (Start 2/1/97; Complete 9/30/97)

Subtask 4.2 Determination of Expected Radioactivity

Typical radioactive elements will be determined for FRR fuels in subtask 4.1. Radioactive decay will reduce radionuclide content and will be determined as a function of time from irradiation from ORIGEN data. The worst case will determine the quantities expected during the melt-dilution process and will be used as a design basis for the off-gas system.
(Start 2/1/97; Complete 9/30/97)

Subtask 4.3 Survey Literature of Fission Gas Release during Melting.

There have been numerous studies at SRS and other DOE laboratories of fission gas release from non-irradiated (containing simulated products) and irradiated aluminum-uranium fuel during melting. These reports will be reviewed to assess the release of radionuclides during melting and casting.
(Start 1/1/97; Complete 4/30/97)

Subtask 4.4 Initiate Fission Product Release Tests, if required

If the data in subtask 4.3 is insufficient, additional studies will be made to determine fission product releases during the melt-dilution process. (Start 6/1/97; Complete 12/31/97)

Subtask 4.5 Development of Off-Gas System

A small-scale off-gas system will be developed and laboratory tested for the melt-dilution technique. The system will incorporate filters and traps to contain volatile and hazardous radioactive products. The test will include irradiated or simulated fuel sections to mock-up fission product release. (Start 6/1/97; Complete 7/30/98)

Task 5.0 Small-Scale Test using MTR Type Fuel Assemblies

Small-scale test will be done to further develop and demonstrate methods for the melt-dilute process.

Subtask 5.1 Fabricate and/or Purchase MTR Type Assemblies with Aluminum and Depleted Uranium-Aluminum Alloy Fuel Plates

MTR type fuel assemblies with both aluminum and aluminum-uranium fuel plates will be fabricated for testing the melt-dilute concept. The aluminum assemblies will be used to develop the process and to determine operational variables, while depleted uranium assemblies will be used for process demonstration and verification. The fuel plates will contain typical uranium compositions. The fuel cores will be made from cast alloys and inserted in aluminum picture frames with top and bottom aluminum plates. The plate assembly will be hot rolled at about 500 °C to simulate the original fabrication process. The plates will be assembled into MTR type fuel assemblies using appropriate techniques.
(Start 2/1/97; Complete 11/30/97)

Subtask 5.2 Small-Scale Melt-Dilute Furnace

Design and build a small-scale prototype furnace and begin laboratory testing of the concept using MTR type fuel assemblies fabricated in subtask 5.1.
(Start 9/1/97; Complete 10/1/97)

Subtask 5.3 Optimization of the Small-Scale Furnace

The small-scale furnace operations and control will be optimized using MTR type fuel assemblies.

(Start 10/1/97; Complete 6/1/98)

Subtask 5.4 Remote Operations

Remote handling requirements will be determined from the small-scale tests and will be incorporated into the facility design.

(Start 10/1/97; Complete initial phase by 5/31/98)

Task 6.0 Waste Form Qualification

Subtask 6.1 Containment Requirements

Candidate canister materials will be selected based on independent work by Lawrence Livermore National Laboratory (LLNL) and the compatibility of these materials with the melt-dilute waste form will be evaluated.

(Start 6/1/97; Complete initial studies by 12/30/97)

Subtask 6.2 Waste Form Characterization

The waste form characteristics (e.g. dissolution behavior, leaching, corrosion, etc.) will be evaluated and analyzed with respect to the performance characteristics desired for geologic repository storage. (Start 7/1/97; Complete initial studies by 6/30/98)

Task 7.0 Evaluate Press-Dilute Technology for Waste Form

Press-dilution technology produces a lamella macrostructure containing alternate layers of aluminum, aluminum-uranium alloy, and depleted uranium metal/oxide. The aluminum-enriched uranium alloy is not diluted directly as with melt-dilution, but maintains its original microstructure and enrichment. However, the average ^{235}U for the assembly is less.

Subtask 7.1 Comparison of Press-Dilute vs Other Technologies

Advantages and disadvantages of press-dilution will be analyzed and compared to melt-dilution technology. Waste form stability will be assessed and compared to direct disposal technology prior to process development. If significant advantages warrant development of a separate technical task plan will be written. (Start 3/1/97; Complete 8/29/97)

APPENDIX B QUALITY ASSURANCE PLAN

The task activities are governed by the requirements of the WSRC 1Q and WSRC E7 manuals and the implementing procedures of the WSRC-SRTC L1 manual (latest revisions of each). The technical task activities are scientific investigations and will be conducted in accordance with 1Q QAP 2-3 and RW-0333P Supplement III. Per the TTR (EF&RFSR/SNFP 97-02), the task is classified Critical Protection. Revisions will be made, as necessary, to reflect programmatic and/or technical changes.

The activities described are not expected to affect any established technical baselines of SRS. However, the data and results produced may affect the creation of a new baseline. Consequently these activities are non-baseline and are designated as Research and Development tasks per 1Q QAP 2-3, Rev. 1.

The control of the task activities is shown below. Measurements and testing will be performed by technical sections (Materials Technology, Applied Technology, Equipment Engineering) of SRTC. Existing guides for routine functions and new special procedures, technical guides, or both are anticipated and will be developed and used, as appropriate.

Customer approval will be secured for all technical and programmatic reports.

Training to RW-0333P requirements will be documented. All participants in the task and subtasks shall read this task plan.

Engineers performing the subtasks or their designees are responsible for maintaining their own records for subtasks in progress. A copy of the records of completed tasks will be stored by Harold Peacock or a designee until program completion. At that time, the task records will be transferred to SRS site records for permanent retention. The task records include items 1-3 per Section 3 of page 3 of this document and those documents as listed in QAP 2-3 Section 5.

Experimental tasks and subtasks will be carried out by issuing experimental task requests, laboratory work requests and observational sheets. They will be collected and stored in a folder until stored permanently. Tests and pertinent observations will be indicated and referenced in laboratory notebooks. Information concerning the melt-dilute program will be reported in laboratory notebooks unless otherwise specified.

Test samples will be tracked using the work request ID number along with an additional unique 1-5 digit and/or alphabetic numbering system. Identification will be included in laboratory notebooks.

This task was assessed per L1, 7.10 Rev. 1 Att. 3 for impact of failed equipment or technology on the programmatic cost and schedule. Multiple paths are being used to minimize effect on cost and schedule.

WSRC IQ Manual Section	Applies to Task (Y, N, AR)	Applicable Procedures	Procedures Used
Organization	AR	IQ; 1-2 Stop Work	
QA Program	Y	L1; 8.21 Supplemental QA Requirements for DOE/RW-0333P	
-Training and Qualification	AR AR	IQ; 2-2, Personnel Training & Qual. L1; 5.03, SRTC Training Records	
-R&D Activities	Y Y Y Y AR	L1; 7.10, Control of Technical Work E7; 3.12, Non-Baseline Tech. Ass. Requests IQ; 2-3, Control of R&D Activities L1; 4.19 Technical Notebook Use E7; 3.70, Qualification of Existing Data	
Design Control			
-Initiation	Y N	E7; 2.02, Baseline Technical Task Request E7; 2.05, Plant Modification Traveler	
-Design Control	AR AR N N N AR N	E7; 2.10 Func. Performance Requirements E7; 2.11, Function Design Criteria E7; 2.13 Task Requirements and Criteria E7; 2.15, Alternative Studies E7; 2.25, Functional Classifications L1; 1.13, Process Hazards Reviews E7; 2.37, Design Change Form	
-Calculations	N Y	E7; 2.16, Technology Risk Assessment E7; 2.31 Engineering Calculations	
-Reviews	AR N N	E7; 2.40, Design Verification and Checking E7; 2.60, Plant Mod. Technical Review E7; 3.14, Design Authority Tech. Reviews	
-Outputs	N N Y N AR	E7; 2.12, Fac. Des. Descrip. and Sys. Des. Descrip. E7; 2.41, Interface Coordination E7; 3.60, Technical Reports L1; 1.22, SRTC Green Letters (Tech. Rec.) for SRS L1; 4.01, Preparing Scien. and Tech. Rep. and Papers	
Procurement Document Control	AR AR AR	WSRC-3E, Procurement Spec. Manual 7B; 1.1, Purchase Requisitioning IQ; 4-1, Procurement Document Control	
Instructions, Procedures, and Drawings	AR AR N	E7; 2.30 Drawings L1; 1.01, Procedure Adm. (Field) L1; 4.02, Generation and Rev. of Process Tech. Manuals	
Document Control	AR N AR	E7; 1.20, Engr. Doc. Numbering System E7; 2.03, Tracking and Turnover of Tech. Baseline Tasks 1B; 3.11, Doc. and Corr. Numbering System	
Control of Purchased Items and Services	AR N N	IQ; 7-2, Control of Purchased Items & Services E7; 3.42, Replacement Item Eval. E7; 3.46, Commercial Grade Item Dedication and Material Upgrade	
Identification and Control of Items	N	E7; 1.30, Component Numbering System	

Control of Processes	AR AR AR N N	1Q; 9-1, Control of Processes 1Q; 9-2, Control of NDE 1Q; 9-3, Control of Welding and Other Joining Processes E7; 2.06, Temporary Modification Control E7; 2.38, Design Change Package	
Inspection	N N	E7; 2.35, Quality Assessment E7; 2.38, Quality Inspection Plan	
Test Control	N AR	E7; 2.26, Post-Mod. Acceptance Criteria 1Q; 11-1, Test Control	
Control of Measuring and Test Equipment	Y AR	1Q; 12-1, Control of M&TE 1Q; 2-7, QA Program Req'ts. for Analytical Measuring Systems	
Packaging, Handling, Shipping, & Storage	AR AR	1Q; 13-1, Packaging, Handling, Shipping and Storage L1; 2.17, Procurement, Labeling, Handling, and Disposition of Hazardous Material	
Inspection, Test, and Operating Status	AR AR	1Q; 14-1, Inspection, Test, and Operating Status L1; 3.03, Off-Shift Coverage of Experimental Equipment	
Control of Nonconforming Items & Action	AR AR	1Q; 15-1, Control of Non-conf. Items 1Q; 15-2, Control of Non-conf. Activities	
Corrective Action System	AR	1Q; 16-1, Corrective Action System	
Quality Assurance Records	Y Y	L1; 8.17, QA Records Management 1Q; 17-1, Quality Assurance Records Management	
Audits	AR AR N N	1Q; 18-1, Quality Assurance Internal Audits 1Q; 18-2, Quality Assurance Surveillances L1; 1.07, Management Assessments L1; 1.08, Self Assessment	
Quality Improvement	N N	1Q; 19-1, Quality Assurance Trending 1Q; 19-2, Quality Improvement	
Software Quality Assurance	Y	1Q; 20-1, Software Quality Assurance	
Environmental Quality Assurance	N	1Q; 21-1, Environmental Quality Assurance	

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