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Options Analyses for Back-End of the Fuel Cycle for USHPRR U-Mo Monolithic Fuel

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TABLE OF CONTENTS

1.0 EXECUTIVE SUMMARY

A low enriched uranium-molybdenum (U-Mo) fuel is being developed to replace the high enriched uranium fuel currently used in U.S. High Performance Research Reactors (USHPRR). To support the reactor conversion effort, a study was conducted to identify and analyze various back-end fuel cycle options resulting in the identification of preferred options for further technology development.

A team of DOE complex experts was assembled to conduct the study. The team held two “face-to-face” working meetings in support of the study. In addition to participation in the working meetings, team members developed the detailed descriptions of the options described in this report. These option descriptions provided the basis for the option rating and evaluation process.

The team defined the requirements for the back-end of the fuel cycle for the USHPRR U-Mo fuel. The requirements statement is as follows:

The alternative LEU fuel shall have a viable disposition path.

The team brainstormed to identify possible options to support the back-end of the fuel cycle for the USHPRR U-Mo fuel. The identified options were binned into three categories:

- Treatment with U recovery
- Stabilization treatment without U recovery
- Direct disposal.

A total of 18 unique options were identified. The options were evaluated against screening criteria. The screening process resulted in screening out 10 options. The rationale for screening out these options was documented. Two options were later identified to be technically similar to other options and were not rated but were carried forward in this report.

The team identified rating criteria for the options that were developed in detail. Six criteria categories were identified with sub-criteria determined for each criterion. The criteria were weighted, and a rating and scoring system was developed to rank the options. The options were rated against the criteria collectively by the team. Consensus on the ratings was obtained through discussion and deliberation.

In the evaluation of the option scoring, the team concluded that it was worthwhile to consider the separate groupings of the options that resulted in recovery of uranium and those that did not result in recovery of uranium. In the grouping with uranium recovery, the team concluded that **Pyrochemical Processing, Aqueous Processing with Solvent Extraction** and **Blending with a Commercial Fuel Process** all scored relatively equal and should be considered for further technology development. It should be noted, however, that the value of recovery of the ~13% enriched U from the U-Mo fuel would be essentially lost in the option of blending with a commercial fuel process. For the options in the grouping without uranium recovery, **Direct Repository Disposal to a Domestic Site** was identified by the team as the preferred option that should be further pursued.

The team identified technology gaps for the preferred options. Additionally, the team developed an initial list of technology development activities to address the highest priority technology needs. The next phase of this effort will be to further develop the technology gaps and development activities into a technology development roadmap. The roadmap will identify the research and development initiatives needed to address these technology gaps and provide a basis to develop a life-cycle research and development (R&D) cost analysis.

2.0 INTRODUCTION

The Global Threat Reduction Initiative's (GTRI) U.S. High Performance Research Reactor (USHPRR) program is developing a low enriched U-Mo fuel as a candidate replacement for the Highly Enriched Uranium (HEU) fuel currently being used in the U.S. high performance reactors, which include the Massachusetts Institute of Technology (MIT) reactor, Missouri University Research Reactor (MURR), National Bureau of Standards Reactor (NBSR) at the National Institute of Standards and Technology (NIST), Advanced Test Reactor (ATR) at Idaho National Laboratory, and High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory. The baseline fuel utilizes an aluminum cladding similar to the present generation of fuel; however, the fuel core consists of a low enriched U-10Mo monolithic alloy with a thin layer of zirconium separating the fuel core from the cladding. The GTRI Convert team is working with the national laboratory complex to develop and qualify the LEU fuel and facilitate the research reactor conversions starting with the Nuclear Regulatory Commission (NRC) licensed HPRR reactors.

GTRI's fuel development, qualification and conversion strategy is comprehensive and includes consideration of options for the back-end of the fuel cycle. The goals of this study are to identify and analyze various back-end fuel cycle options leading to development of a roadmap that identifies potential back-end solutions. Further, the task will identify technical gaps and needs for the primary potential back-end solution and associated life-cycle R&D costs.

A team of DOE complex experts was assembled to identify and rate options. Biographical sketches of the team members are provided in Appendix A. Members of the option analysis team included:

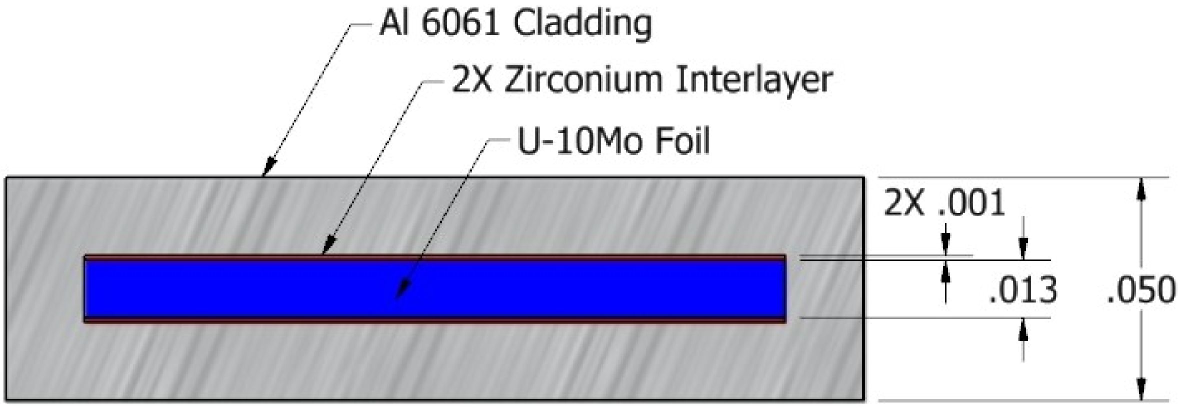
Jim Marra, Savannah River National Laboratory (Study Lead)
George Vandegrift, Argonne National Laboratory
Mark Williamson, Argonne National Laboratory
Steve Frank, Idaho National Laboratory
Steve Herrmann, Idaho National Laboratory
Tracy Rudisill, Savannah River National Laboratory
Bob Sindelar, Savannah River National Laboratory.

This document summarizes the team's analyses of options for the back end of the fuel cycle.

3.0 BACKGROUND INFORMATION FOR OPTIONS STUDY

3.1 U-Mo Monolithic Fuel Description

The U-Mo fuel design is based on a monolithic uranium-molybdenum fuel alloy, enclosed in Al-6061 cladding, with a diffusion/bonding interlayer composed of zirconium. A schematic diagram of a foil-type element under consideration is shown below. The monolithic design can provide for uranium densities much greater than in dispersion fuels which may be required to convert the USHPRRs to a LEU fuel. However, fabrication of the monolithic fuel presents unique challenges especially with regard to consistency in application and performance of the zirconium interlayer.



Schematic diagram of an LEU U-Mo fuel foil (dimensions in inches)

An ATR Full Element demonstration fuel plate was used as an example for this study. This provided a basis of a reduced-enrichment USHPRR fuel element that could be used for comparison of the options. The ATR LEU fuel element is comprised of a U-10Mo alloy (10 wt % Mo) at 19.8 wt % uranium enrichment with a Zr coating at 5 wt %. The fuel is clad in aluminum. The composition of a single fuel element can be approximated as:

- U-10Mo: 460 grams
- Zr: 25 grams
- Al: 245 grams.

3.2 Back-end of the Fuel Cycle Requirements

The requirements for the back-end of the fuel cycle for the USHPRR U-Mo fuel were defined by the team as follows:

> *The alternative LEU fuel shall have a viable disposition path.*

3.3 Assumptions

The following assumptions were made to support definition and rating of the options.

1. LEU conversion starts in 2020-2025 (TBV)

2. Back-end handling is required to start 2025-2030 (30-35 years of storage available). Time from when fuel is removed from the reactor core to when it is discharged from the reactor plant is 5 years.
    a. Disposition path way could start as early as 2025.
    b. Disposition path way would need to start by 2060 to avoid need for additional storage.

3. Extended storage in L-basin at SRS is available from 2010 through 2060.
    a. Foreign Research Reactor (FRR) fuel returns stop in 2019.
    b. Domestic Research Reactor (DRR) fuel returns continue.

4. Treatment and disposition options only consider USHPRR U-Mo LEU fuel.

5. For recycle or stabilization options, new facilities may be required.
    a. Existing facilities could be used for technology development and demonstration purposes.

6. The number of LEU fuel elements to be received is similar to current HEU fuel.

7. The disposition process needs to address a U-10Mo spent fuel discharge rate up to 2 MTHM/year.

8. There will be a disposition path for the treated or untreated U-Mo fuel.

9. There is value to recovered U (i.e. discharged U-10Mo fuel enrichment is expected to range from 10 to 19.8%), and recovered U will meet acceptance specifications (TBD).

10. Pu and other transuranic (TRU) elements will not be recovered and will be dispositioned as part of the waste stream.

11. Engineered waste forms are superior to direct fuel disposal for long-term environmental stewardship.

12. The current HPRRs will operate in the future with at least one HPRR operating until 2035. New HPRRs will be built and commissioned in the future.

13. Any new HPRR fuel design that is significantly different from the U-Mo LEU fuel will be considered in an addendum to this study.

4.0 SCREENING CRITERIA

To facilitate eliminating options deemed to be not feasible and/or of very low maturity, screening criteria were developed. The screening criteria were applied by the study team to the defined options. Those options eliminated in the screening process were not further developed by the study team. However, for completeness, brief descriptions of the screened-out options are included in this report with brief narratives on the screening criteria used to eliminate the options from further consideration.

The screening criteria are as follows:

Maturity (M)

1. Main unit operations not demonstrated beyond lab-scale
2. Reagents and equipment not readily available
3. Undeveloped waste processing methods
4. Undefined wastes and waste by-products

Product Quality/Recoverability (PQ)

1. Unacceptable product quality

Cost (C)

1. Extensive technology development required

Complexity (CX)

1. Highly complex unit operations
2. Multiple recycle/waste streams

Environmental, safety, regulatory and societal acceptance (E)

1. Not acceptable.

5.0 RATING CRITERIA

Six criteria categories were identified with sub-criteria determined for each criterion. The criteria were based on the practicality of the option to satisfy requirements at an industrial scale, the relative maturity of the option, the impact of waste generation, the ability to recycle uranium, and the relative costs and risks to meet programmatic and institutional objectives.

Sub-criteria were developed to further refine the main criteria and provide a means to better distinguish between the options. Careful consideration was given to identify unique criteria (and sub-criteria) to ensure that an option would not be unfairly rewarded (or penalized) by double-counting similar criteria.

The criteria and sub-criteria were defined as follows:

Industrial Practicability

1. Technical practicality
2. Robustness (recovery from process upsets, etc.)
3. Complexity (number of steps, materials handling, etc.)
4. Operating costs
5. Capital costs

Technical Maturity

1. Development costs
2. Schedule to demonstrate
3. Process maturity (includes balance of plant)

Waste

1. High level waste disposition cost
2. Low level waste disposition cost

Uranium recovery

1. Yield
2. Product quality

Cost/Risk to Meet GTRI Objectives

1. Nonproliferation
2. Materials control and accountability
3. Physical security

Cost/Risk to Meet Environmental, Safety and Regulatory Requirements

1. Environmental
2. Safety
3. Regulatory.

The relative ability of the criteria to discriminate among the options was weighted as “high”, “medium,” and “low”. The study team concluded that the criteria weighted as “high” would provide the best ability to discern between the options to identify the most promising. It should be noted that the weighting of the criteria does not directly relate to the *importance* of that criteria but rather to its ability to discriminate among the options.

Industrial Practicability and Technical Maturity were considered to be “high” discriminators. Waste and Uranium Recovery categories were identified as “medium” discriminators. Cost/Risk to Meet GTRI Objectives and Cost/Risk to Meet Environmental, Safety and Regulatory Requirements were identified as “low” discriminators.

The options were rated with a “+” for a positive rating against the criteria, “0” for a neutral rating against the criteria, and “−“ for a negative rating against the criteria. Brief explanation regarding the relative rating was provided in a table for each option. Furthermore, the team worked to obtain consensus on ratings and to ensure consistency of ratings against the criteria.

6.0 OPTIONS

Options to support the back-end of the fuel cycle for the USHPRR U-Mo fuel were identified by the expert team. The identified options were binned into three categories:

- Treatment with U recovery
- Stabilization treatment without U recovery
- Direct disposal.

6.1 Treatment with U Recovery

Consistent with the assumption that "there is value to recovered U," options to reprocess the fuel to separate and recover uranium for re-use were identified. The options involving recovery of uranium were sub-divided into "Non-aqueous Processing" and "Aqueous Processing" categories.

6.1.1 Non-aqueous Processing

In the non-aqueous processing category, five unique options were identified. These options are discussed in the following sections.

6.1.1.1 Pyrochemical Processing

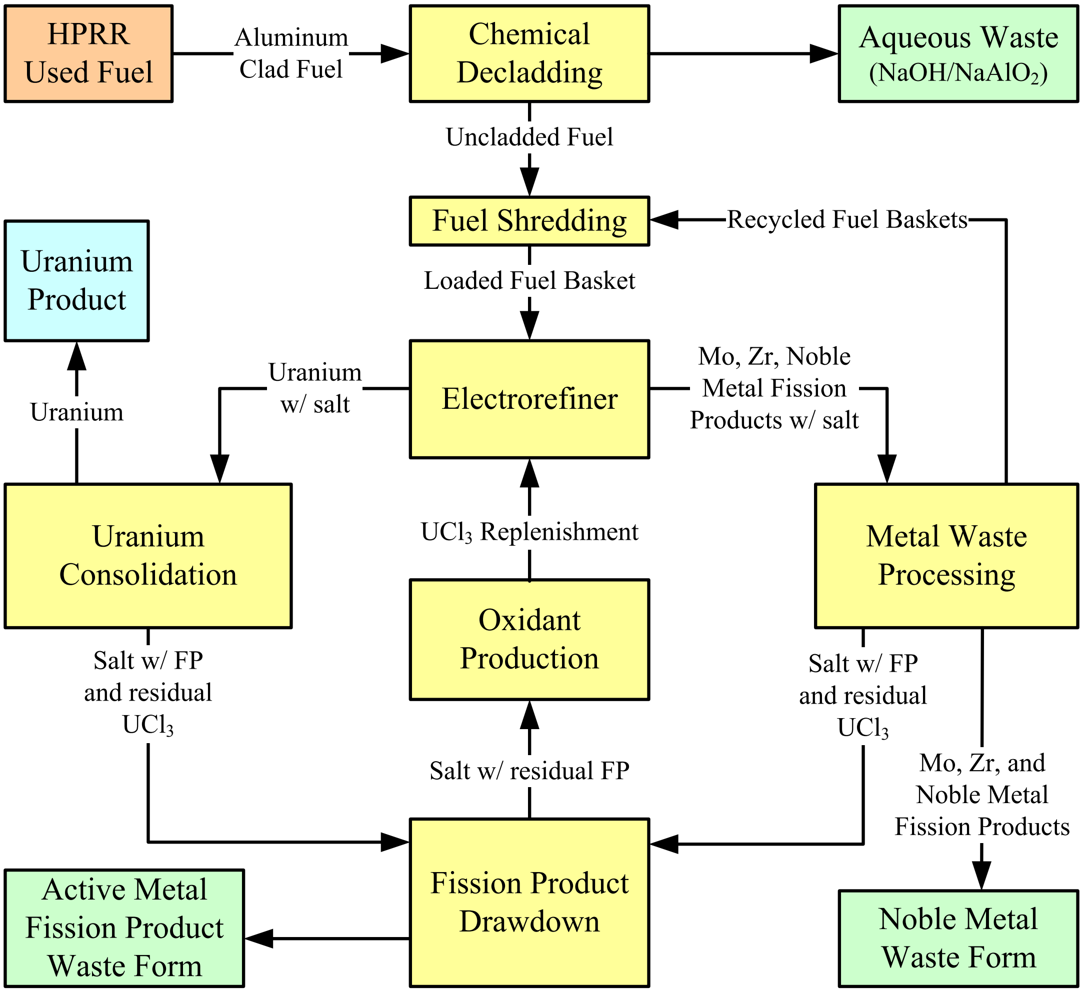
Technology Overview

Pyrochemical processes are well-suited for treating metallic fuel, and application of the technology to actinide recycle has been ongoing since the 1950s [1-4]. In fact, pyrochemical processes are the only means to produce actinide metals. Processes such as melt refining successfully recovered actinides from used Experimental Breeder Reactor II (EBR II) fuel—which, except for the cladding, is similar in constitution to research reactor fuel--for recycle to the EBR II fast reactor. Throughout the 1960s and 1970s a number of pyroprocessing flowsheets based on reductive extraction technologies were developed and tested, demonstrating actinide recovery and fission product decontamination for numerous fuel types, such as oxide and metal fuel. The Integral Fast Reactor (IFR) program resulted in the arrival of electrochemical technologies for decontaminating and recovering actinides from used metallic fuel. Electrochemical technologies such as electrorefining produce far less secondary waste than reagent-based processes, eliminate the need for complex reagent/ solvent recycle systems, are compact, and provide excellent criticality safety features. Electrochemical technologies developed during the IFR program are currently being used to condition fuel discharged from EBR II, and they provide the foundation for the pyrochemical technologies being developed for the Office of Nuclear Energy's Fuel Cycle Research and Development program. Several recent studies explored the application of pyrochemical processing to the recovery of uranium from irradiated research reactor fuel and scrap produced during the fuel manufacturing process.

Laboratory-scale feasibility tests conducted at Argonne National Laboratory demonstrated uranium recovery from irradiated Zr-bonded U-10Mo fuel by pyrochemical methods, specifically by electrorefining [5]. Feed material for the tests was prepared by irradiating sample fuel foils at the Argonne LINAC facility, thereby yielding measurable quantities of fission products from the γ/fission reaction on ^{238}U. Electrorefining the irradiated material yielded a uranium product decontaminated from fission products, bond zirconium and molybdenum. Active metal (e.g., Cs, Sr) and lanthanide fission products partitioned to the molten salt phase in the electrorefiner along with the transuranic elements. Noble metal fission products such as Zr and Mo remained in the anode of the electrorefining cell along with residue from the feed. These tests not only demonstrated the viability of using pyrochemical methods for uranium recovery from used high performance research reactor (HPRR) fuel but also confirmed the behavior of fission products and fuel residues in the treatment system.

Prior to evaluating the feasibility of recovering uranium from irradiated U-10Mo alloy, a series of laboratory-through-engineering-scale tests was conducted to demonstrate recovery of uranium from fuel scrap generated during the HPRR fuel fabrication process [5-8]. Laboratory-scale electrorefining tests confirmed the recovery of uranium from the fuel scrap, leaving the bond Zr and Mo in the anode compartment of the electrochemical cell along with residual feed material. Following the successful laboratory-scale tests, a series of engineering-scale tests was conducted to develop a thorough understanding of the uranium recovery process and begin developing a set of operational guidelines for the system. A prototype module for a commercial-scale electrorefiner was used in the series of engineering-scale tests with U-Mo fuel scrap and simulated fuel materials (e.g., U-10 wt % Zr alloy) as feed. The studies verified recovery of uranium without significant Zr and Mo impurities and that noble metal fuel components within the scrap would remain in the anode of the cell. These tests also explored the effects that different operating parameters had on the performance of the system and uranium purity. In addition to demonstrating uranium recovery by electrorefining, uranium product consolidation studies established that the uranium dendrites collected during the electrorefining process could be treated by distillation and melting to produce a homogeneous uranium ingot for recycle to the fuel fabrication process.

Process Description
A conceptual flowsheet describing the pyrochemical processes needed to treat used HPRR fuel is shown in the following figure. The flowsheet consists of seven principal operations (shown in yellow) that begin with a chemical decladding step to remove the aluminum cladding from the fuel. Otherwise, introduction of aluminum to the pyrochemical process would result in the formation of an aluminum-containing salt phase species that complexes with uranium (and other actinides) and inhibits the deposition of a purified uranium product. Dispersion fuels have been explored as alternatives to the monolithic fuel, and treatment of an Al-based dispersion fuel would lead to similar issues with complex formation.



Conceptual Pyrochemical Process to Treat Used HPRR Fuel

After the fuel decladding process, the material is dried, mechanically chopped or shredded and placed into baskets that are transferred to the electrorefiner. The metallic fuel contained in the basket serves as the anode in the electrorefining cell. A steel mandrel functions as the cathode. The electrolyte used in the cell is a LiCl-KCl eutectic salt containing approximately 6 wt % UCl_3, which is held at 500°C. As current is passed between the anode and cathode of the cell, uranium is anodically dissolved at the anode to produce uranium ions that are soluble in the electrolyte. The uranium ions are transported through the molten salt to the cathode where they are reduced to produce metallic uranium. The transuranic elements present in the feed material are anodically dissolved and form transuranic chlorides that are soluble in the electrolyte. Active metal (e.g., Cs, Sr) and lanthanide fission products partition to the molten salt along with the transuranic elements during the anodic dissolution process. Noble metal (e.g., Zr, Mo, Ru) fission products remain in the baskets along with the bond Zr and Mo from the used fuel. Fission gases are released during the refining process and captured in an off-gas handling system of the facility. Iodine partitions to the salt phase and forms a salt soluble iodide.

Uranium product from the electrorefining process may retain up to 20wt% electrolyte salt on the surface of the dendrites and dispersed in the pores of the uranium. The salt contains transuranic and fission chlorides that must be removed prior to uranium recycle. A distillation process, conducted at approximately 800°C, is used to recover the salt from the dendritic uranium. After salt removal from the surface of the dendrites, the uranium is consolidated to an ingot by heating the dendrites to 1200°C. The consolidation process also removes any salt trapped in pores in the matrix of the dendrites. The resulting uranium product is transferred from the consolidation system to fuel fabrication. If desired, the uranium can be blended down during the consolidation process by adding depleted or natural uranium to the dendritic feed, thereby producing a fuel ingot.

Salt distillation is also performed on the noble metal fission products and residual fuel components, zirconium and molybdenum, that remain in the basket (i.e., metal waste processing). Distillation of salt from the surface of the material occurs at 800°C and is followed by consolidation of the metal into an ingot at ~1600°C to create a metal waste form. It should be noted that the effects of Zr and Mo on the melting process and required melting temperatures would need to be evaluated.

Salt discharged from the uranium consolidation and metal waste processing step is transferred to the fission product draw-down process to remove the salt soluble fission products. This process first recovers residual uranium, contained in the salt phase as UCl_3, for recycle to oxidant production. Next, the process is configured to recover the transuranics and lanthanide fission products for encapsulation in a glass waste form. Alternatively, the transuranics can be separated prior to lanthanide separation from the salt phase if there is a need to do so, for example, if the transuranics will be used in other recycle fuels. Finally, the active metals fission products are recovered along with the iodine for encapsulation in a ceramic waste form or inclusion in the glass waste form containing the lanthanides. The resulting salt is transferred to the oxidant production step where UCl_3 is added to provide a transport pathway for uranium ions in the electrorefiner.

High-level waste forms produced by the process include a metal waste form containing noble metal fission products, bond Zr and Mo; a glass waste form containing the residual transuranic elements and lanthanide fission products; and a ceramic waste form containing the active metal fission products, which can be stored in decay storage prior to disposal in a geologic repository. If desired the active metal fission product waste can be blended with the glass waste form containing the transuranic and lanthanide fission products. A secondary waste generated by the process is the basic aqueous waste stream containing aluminum from the cladding dissolution process.

Identified Technology Gaps

- Work to date has focused on uranium recovery from scrap Zr-bonded U-Mo fuel and simulated materials; additional development is needed at engineering-scale to identify the full range of operating conditions and determine uranium recovery efficiency and product quality as functions of operating conditions. (High Priority)
- Additional testing with irradiated U-Mo fuel is needed to fully assess the impact of fission products, at process-relevant concentrations, on uranium recovery and decontamination. (High Priority)
- A key technology need is development of a method to chop/shred fuel plates to expose the fuel meat for anodic dissolution in the electrorefiner.
- Another need is optimization of uranium product purity through testing and evaluation of U morphology in the electrorefiner, consolidation process improvements, implementation of salt wash. (High Priority)
- The fission product recovery process must be demonstrated at engineering scale to confirm process efficiency and product quality. (High Priority)
- Integrated demonstration of the technology at process relevant scale must be conducted to establish material balance for the system, process efficiency data, and product quality information (i.e., uranium and waste forms).
- The waste forms must be qualified for disposal in a geologic repository.
- The design concept for the integrated fuel treatment facility, including main process equipment, must be developed.
- Potential effects of NaOH dissolution and/or high fuel burn-up on downstream e-chem processing must be determined.

- Alternative decladding techniques (chloride volatility, etc.) also need to be investigated.

Criteria for Evaluating Option

Category	**Rating**	**Comments**
Industrial Practicability		
Technical Practicality	+	Uranium separation from similar fuel (EBR II) has been demonstrated in hot-cell environment, and results from preliminary tests with U-Mo fuel indicate uranium decontamination should be better than achieved with EBR II fuel; auxiliary operations also have been demonstrated in hot cell environment; equipment designs are available to support commercial implementation.
Robustness	+	Pyroprocesses are relatively insensitive to changes in process electrolytes, operating conditions, reasonable temperature fluctuations, etc.; recovery from process upsets are uncomplicated; technology is applicable to treatment of other fuel alloys and cladding types.
Degree of Complexity	0	Plant and equipment are designed for remote operations and maintenance; plug-and-play philosophy can be used in HPRR treatment plant design.
Operating Costs	0	
Capital Costs	-	New facility will need to be designed and built to accommodate pyrochemical treatment of used fuel.
Technical Maturity		
Development Costs	0	Considerable technology development exists from R&D in complementary programs; development activities could begin at engineering-scale followed by pilot-scale demonstration of technology with simulated used fuel; irradiated HPRR fuel tests could be conducted parallel with engineering development activities.
Schedule to Demonstrate	0	
Process Maturity	+	Main unit operations have been identified and demonstrated for similar fuel types at engineering-/ pilot-scale.
Waste		
HLW Disposition Costs	0	Disposition site has not been identified, but HLW disposal costs are not anticipated to be higher than direct disposal of used fuel; engineered waste form performance is superior to direct disposal and potentially requires less space; engineering-scale experience exists in waste form production, and acceptance testing underway.

Category	Rating	Comments
LLW Disposition Costs	0	Disposition site is not identified, but LLW disposal costs are not anticipated to be higher than direct disposal of used fuel; pyrochemical processes are not expected to produce significant quantities of LLW.
<u>U Recovery</u>		Process is specifically designed for recovery and recycle of uranium.
Yield	0	Uranium yield from irradiated fuel needs to be experimentally established; experience in treating unirradiated fuel scrap indicates yield will be acceptable.
Product Quality	+	Tests at engineering-scale with simulated used fuel indicate uranium decontamination would meet recycle requirements; development activities will confirm product quality.
<u>Cost/risk to Meet GTRI objectives</u>		
Nonproliferation	0	Potential benefit is due to uncomplicated nature of process and abundance of available process monitoring information.
Materials Control and Accountability	0	
Physical Security	0	
<u>Cost/risk to Meet Environmental, Safety and Regulatory Requirements</u>		Although no commercial pyroprocessing facility has been built and operated, any barrier to do so is no higher than other technologies of similar maturity; waste disposition pathways are not identified due to lack of specific disposal site.
Environmental	0	
Safety	0	
Regulatory	0	

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6.1.1.2 Justification for Eliminating Molten Salt Extraction

Technology Overview

Molten salt extraction processes have been developed and explored for the treatment of used nuclear fuel. These processes use molten salts and liquid metals to bring about the desired separation of actinides from fission products, typically through a series of oxidation and reduction processes, so that the actinides can be recycled to fuel fabrication. For example, the salt transport process was conceived and developed for the treatment of used oxide fuel [1 and references therein]. The process used a number of liquid metal reagents (e.g., Cu-Mg, Zn-Mg) in combination with a molten salt (e.g., $CaCl_2 – CaF_2$) to decontaminate and recover the uranium and plutonium from the used oxide fuel. In a similar study, the distribution of actinides between liquid cadmium and molten LiCl-KCl was evaluated as a method to decontaminate actinides from fission products as one flowsheet option for the Integral Fast Reactor Program [2]. Numerous other examples of molten salt extraction or reductive extraction techniques can be found through a search of the literature (e.g., Molten Salt Reactor Experiment), but no method specific to the treatment of used HPRR fuel has been developed.

Rejection Criteria

It is conceivable that a process specific to the treatment of used HPRR can be developed based on data from existing studies and properties of the fuel components. However, the technology readiness level of the newly defined process would be low since considerable development work would be required. In addition, waste treatment processes are relatively unknown as are suitable waste forms. Thus, molten salt extraction techniques are not considered viable options for treating used HPRR fuel.

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6.1.1.3 Justification for Eliminating Melt Refining

Technology Overview

Melt refining refers to a pyrometallurgical process in which spent uranium fuel alloys are melted in a ceramic crucible, effecting fission product removal via volatilization and “oxidative slagging” or “drossing.” Specifically, declad spent uranium alloy fuel is melted in a calcia-stabilized zirconia crucible under inert atmosphere at a temperature of 1300 – 1400°C for several hours, after which the molten metal is poured away from the dross. Fuel constituents having high vapor pressures at these temperatures--including xenon, krypton, cesium, cadmium, and iodine (as calcium iodide)--are rapidly removed from the melt by volatilization. Alkaline earth and rare earth fission products react with the crucible to form dross, which adheres to the crucible wall as the molten metal is subsequently poured. Noble metal fission products--including molybdenum, technetium, ruthenium, rhodium, and palladium--are not removed in the melt refining process but form an alloy with the recovered molten uranium metal that is commonly referred to as “fissium” alloy or “fissium” fuel.

The melt refining process enabled a simple, rapid, low-decontamination recycle of metal uranium alloy fuel for the early U.S. fast reactor development program. This process was successfully applied in recycling 2.4 MTHM of spent fuel from Experimental Breeder Reactor – II (EBR-II) in the Fuel Cycle Facility at Idaho National Laboratory from 1964 – 1969 [1-2]. Given the incomplete separation of fission products and losses on the order of several percent, the melt refining process was eventually replaced with an electrorefining process for U.S. fast reactor metal fuel-cycle development, as part of the Integral Fast Reactor program [3].

Rejection Criteria

Melt refining was eliminated as a treatment option for USHPPR fuel based on deficiencies in technical maturity for this fuel type, product quality, cost, and complexity. Even though the melt refining process has been used in past recycle of stainless-steel-clad, sodium-bonded EBR-II fuel elements, its application to aluminum-clad, zirconium-lined U-Mo fuel is not defined. Equipment is not available to support such treatment, and associated waste processing methods are not developed. Given an incomplete separation of fission products, the product quality of the melt refining process for USHPPR fuel would also be considered unacceptable. The investment to adapt this technique to treatment of USHPPR fuel would be extensive and would likely involve highly complex and multiple recycle streams to achieve adequate product quality and suitable waste forms.

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6.1.1.4 Justification for Eliminating Processing using Ionic Liquids

Technology Overview

Room temperature ionic liquid (RTIL) commonly refers to fused or molten salts with melting points at or near room temperature [1-2]. They have been proposed as a replacement for alkali or alkaline earth halides or mixtures thereof in pyrochemical processes. RTILs are organic salts, such as alkylammonium halides or nitrates, mixtures of organic salts, or mixtures of organic salts and inorganic salts, such as N-butylpyridinium chloride – aluminum chloride. Beyond having low melting points, these liquids are excellent solvents due to their adjustable Lewis acid properties, possess low vapor pressures, offer an increased electrochemical window compared to traditional molten salts, and are a non-aqueous alternative for solvent extraction processes because they are immiscible in organic solvents. Early studies focused on chloride-based salt systems, but recently the emphasis is on systems that contain the tetrafluoroborate and hexafluorophosphate ion. They are more robust and, perhaps, could withstand a high-radiation environment. RTILs have been proposed for use in the electroplating industry as solvents for catalysis and synthesis reactions and as the processing media for the treatment of spent nuclear fuels or waste from spent nuclear fuel processing.

A detailed review of the chemistry of RTILs is beyond the scope of this document. However, it is important to mention the status of two research areas. First, knowledge of the chemistry of the actinide elements in RTILs is at an early stage of development. Limited data exist for U, Np, and Pu in RTIL melts. Most efforts are focused on experimentally establishing the electrochemical properties of actinide complexes by voltammetric and other techniques. Spectroscopy is being used to elucidate the speciation of the actinides in the RTILs and to support conclusions drawn from the electrochemical data. Second, the effect of high-radiation environments on these materials is unknown. It is crucial to understand the behavior of RTILs in high alpha, beta, gamma, and neutron environments.

Rejection Criteria

RTIL technology is too immature to consider as an option for treating used HPRR fuel. Conceptual flowsheets have not been developed for treating spent nuclear fuel or partitioning the long-lived fission products. The lack of fundamental experimentally determined data severely inhibits extrapolating the technology to commercial separations systems. Waste issues are relatively unknown. Finally, development of RTIL technology to the plant-scale goes well beyond the current process development timeframe.

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6.1.1.5 Justification for Eliminating Halide Volatility

Technology Overview

Halide volatility refers to the halogenation, or chemical reaction of fuel with the halogen elements of fluorine, chlorine or bromine to chemically and physically alter the original fuel component. The volatile halide compounds can then be separated from one another based on differing vapor pressures [1]. For example, uranium fuel fabrication involves reacting uranium oxide with fluorine to form volatile uranium fluoride followed by U isotopic enrichment and blending. Fluorination of uranium is applied at the industrial scale throughout the world [2]. Investigations involving chlorination of used nuclear fuel (UNF) have centered on dry chemical de-cladding methods for both zircaloy and aluminum cladding [3], and for electrochemical dissolution of fuel in molten chloride salt [1]. Recently, laboratory-scale bromination studies of surrogate UNF have been investigated as an alternative for chlorination for the separation of volatile components and for electrochemical dissolution in molten bromide salts. With the exception of dry de-cladding, the majority of halogenation processing is performed on oxide materials. Halogenation of HPPR fuel would be performed in the original metallic state of the fuel, or after conversion to another chemical state such as the oxide.

Rejection Criteria

The halide volatility option for the treatment of HPPR fuel was screened out based on operating complexity, incomplete process knowledge and the perceived difficulty in meeting stringent governmental regulations and public safety acceptance. As mentioned, fluorination of non-radiated uranium is performed at the industrial scale as a technologically mature process; however, the technology involved with the halogenation of irradiated materials is far less mature, having been performed primarily at the laboratory and engineering scale. Considerable developmental work would be required to characterize the various product and waste streams and to formulate durable waste forms to immobilize collected fission products. Moreover, processing and material-compatibility constraints involved with the handling of corrosive halogen reagents would add significant complexity and cost to the treatment method. The hazardous nature of halogen reagents also increases regulatory and safety compliance requirements. Removal of the Al cladding by chlorination to separate the fuel, while only in the conceptual stage of development, may be considered as an area for investigation in the pyroprocessing technical gap section.

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6.1.2 Aqueous Processing

6.1.2.1 Use of TBP-Based Solvent Extraction for Uranium Recovery and Purification

Process Description

Following dissolution of high performance research reactor fuels and subsequent clarification of the dissolver solution, the U can be recovered by tributyl phosphate (TBP)-based solvent extraction [1,2]. Two options can be used to dissolve the fuel. Entire fuel plates can be dissolved in nitric acid with sufficient fluoride to dissolve the Al cladding and the U-10Mo alloy, or the Al cladding can first be removed using a caustic solution [1]. The dissolution of the Al cladding is represented by equation (1), which was empirically derived based on laboratory-scale studies of the process used at the Savannah River Site to remove Al cladding from depleted U targets. The $NaNO_3$ is used to suppress the evolution of H_2 by altering the reaction so that primarily NH_3 is produced instead of H_2 [3].

$$\mathrm{Al + 0.7NaOH + 0.58NaNO_3 + 0.11H_2O \rightarrow NaAlO_2 + 0.28NaNO_2 + 0.3NH_3 + 0.009H_2} \quad (1)$$

If the Al is removed using a caustic solution, disposal of the dissolver solution as a low-level waste would be possible based on current regulations. Cladding waste would not be considered a high level waste (HLW) based on the Department of Energy's interpretation of the Nuclear Waste Policy Act and the Nuclear Regulatory Commission's definition of HLW (i.e., the cladding waste stream is generated before the first separation process, so it is not defined as a HLW [4-6]).

During the dissolution of the U-10Mo alloy and the Zr-bonding layer, two limitations must be addressed to ensure safe and efficient operations—(1) the need to dissolve in the presence of fluoride and (2) the limited solubilities of uranyl molybdate and molybdic acid.

The presence of a Zr-bonding layer for the monolithic U-Mo fuel puts a difficulty on aqueous processing of this fuel. The interface between the U-Mo fuel and the Zr layer is likely to contain U-Zr compounds that can react explosively when dissolved in nitric acid alone [1, 7-12]. It is, therefore, common practice to dissolve fuels that contain U-Zr intermetallic compounds in nitric acid containing significant quantities of hydrofluoric acid. Process flowsheets for dissolving and processing this fuel using fluoride have been developed but not demonstrated. The Savannah River Site (SRS) has the capability to reprocess this fuel if the free fluoride ($HF + F^-$) concentration is kept low enough to make corrosion of the dissolver and other stainless-steel equipment manageable. At the free-fluoride concentration required to dissolve this fuel, the corrosion rate is estimated to be in the range of 0.78 mm/year. Determination of an "acceptable" rate of vessel corrosion is an empirical task that depends upon the construction of the vessel, the expected time of contact, and the desired service life of the system, among other factors. Such a determination requires an engineering assessment, although rates below 1.3 mm/ year are considered "low," "mild," or "adequate" in similar contexts [13, 14].

By strongly complexing TRU elements and many of the fission products and thus limiting their extractability, fluoride addition has a benefit to purifying uranium and eliminates concerns about co-extracting plutonium. However, the high fluoride concentration in the process raffinate will be challenging to the formulation of a glass composition to serve as a disposal form; the high fraction of Mo in the raffinate solid adds to the challenge. The raffinate might be treated as was the raffinate waste generated at the Idaho Chem Plant during reprocessing of Navel fuel; in that case, fluoride was precipitated from solution as CaF_2, and the slurry was calcined for long-term storage.

Uranium-molybdenum alloys containing up to 10 wt % Mo dissolve rapidly in nitric acid [15]; however, the formation of uranium-molybdate and/or molybdic-acid solids can be an issue for U concentrations greater than approximately 50 g/L [1]. To prevent the formation of solids following dissolution, ferric nitrate can be added to the dissolver solution to complex the Mo and prevent the precipitation of U [1, 15-22]. In the example flowsheets described below, the fuel has been dissolved at 25 g-U/L without and 75 g-U/L with ferric nitrate addition.

The first step in processing the fuel is its dissolution. The table below shows a series of potential feed compositions (ignoring fission and activation products in the spent fuel) for U-Mo fuel dissolved in nitric acid and hydrofluoric acid. The compositions vary due to (1) whether the Al clad is dissolved with the fuel or dissolved prior to fuel dissolution or (2) whether ferric nitrate is added to the dissolver to increase the solubility of the fuel. A recent publication presented simple, one-cycle flowsheets using 30 vol% TBP in n-dodecane that were modelled for these feeds using the Argonne Model for Universal Solvent Extraction (AMUSE).[1,23]

Potential feed compositions for dissolved fuel plates (excluding fission and activation products) [1,3]

Feed #	U (g/L)	Mo (g/L)	Max. Mo g/L 100 °C	Al-6061 Clad (g/L)	Final HNO_3 (M)	Min. Final HNO_3	Max. Final HNO_3	F^- (M)	Al (M)	Fe (M)	Zr (M)	Si (M)
1	20	2.2	2.8	12.4	3	2.2	5	0.34	4.5E-01	6.2E-04	1.7E-02	3.4E-03
2	50	5.6	10	31.0	2	1	4	0.86	1.1E+00	5.0E-01	4.2E-02	8.4E-03
3	20	2.2	2.8	-	3	2.2	5	0.07	-	-	1.7E-02	-
4	75	8.3	9	-	2	1.5	3.5	0.25	-	5.0E-01	6.3E-02	-

A typical solvent-extraction process has three or more sections--extraction, scrub(s), and strip. In the extraction section, uranium is extracted into the organic phase in a series of counter-current-flow stages. The spent target solution enters at the feed stage--in an 8-stage extraction section, at stage 8--and the fresh solvent enters at the other end of the extraction section at stage 1. As the solvent moves from stage 1 to stage 8, the organic-phase concentration of uranium increases. As the spent-target solution moves from stage 8 to 1, its uranium concentration decreases. The raffinate leaving stage 1 contains very little uranium.

The solvent leaving stage 8 then enters the scrub section. The job of the scrub section is to remove the small amounts of other fuel components from the organic phase. Assuming the scrub section has 6 stages, the loaded solvent enters the scrub section in stage 9 of the full process and travels to stage 15. The aqueous scrub solution enters this section at stage 15. As the solvent travels from stage 9 to 15, its concentration of impurities drops. The aqueous scrub solution, running countercurrently though the scrub section from stage 15 to stage 9, becomes more concentrated in these impurities at every stage. The aqueous scrub solution then travels into stage 8 (the extraction feed stage) where it mixes with the feed. The raffinate flow rate is a combination of the extraction and scrub flow rates.

Following the purification performed in the scrub section, the uranium-loaded solvent then flows to the strip section (stage 16). At the other end of the strip section the aqueous-strip solution is fed into the contactor; for a 15-stage strip section, this would be at stage 30. As the solvent flows from stage 16 to 30, its uranium concentration decreases. As the aqueous-strip solution flows from stage 30 to 16, its uranium concentration increases. The solvent leaving stage 30 is free of uranium and ready to be recycled to stage 1 to repeat its path through the contactor. The aqueous pure-uranium-product solution leaves the contactor at stage 16.

The feed to the scrub section is 0.5M HNO_3 with or without 0.1M acetohydroxamic acid (AHA). Even with the presence of fluoride in the feed, AHA could be added to the scrub feed because it is a powerful complexant for the TRU elements and many transition metals and allows the purification of uranium in a single cycle; it may not be necessary due to fluoride complexation. Because the scrub flows into the extraction section, it acts to complex feed components in both the extraction and scrub sections.

The end result of the solvent-extraction process is a liquid waste raffinate and a solution containing 0.01 M HNO_3, uranyl nitrate, pertechnetate, and, perhaps, iodide and/or iodate. The pertechnetate and iodide can be removed by anion exchange. The resulting uranyl-nitrate product solution should be pure enough for recycle. The U-product solution would be concentrated and thermally denitrated, with the final product leaving the facility being a uranium oxide solid.

Identified Technology Gaps

- Earlier GTRI work performed on processing this fuel was directed toward recycling of fabrication scrap and, therefore, very little emphasis was given to the effects of dissolution conditions on the activation and fission products that would be present in spent fuel. The solubility of rare-earth and some other fission- and activation-product fluorides is quite low. An investigation is required to assure that these fluoride salts will not precipitate during processing. (High Priority)
- Significant data exist for dissolving similar fuels and cold surrogates,[1] but dissolution of highly irradiated fuel of this type has not been done (with the possible exception of some analytical dissolutions at the Idaho National Laboratory). Corrosion tests under conditions necessary to dissolve and process this fuel need to be performed for stainless steel or other potential materials of construction.
- There are two options for dissolution—(1) pre-dissolution of the Al cladding using sodium-hydroxide/nitrate solution or (2) dissolving the intact fuel in a nitric-hydrofluoric acid mixture. Bench-top proof-of-principal experiments have shown the efficacy of both approaches. A technical/economic study needs to be performed to decide on the best approach. (High Priority)
- The addition of ferric nitrate will increase the solubility of this fuel by three-fold (from ~20 to ~75 g-U/L). A technical/economic study needs to be performed to rate the pros and cons of ferric nitrate addition.
- A process flowsheet can be designed using AMUSE; however, this flowsheet needs to be verified by pilot-scale tests using irradiated U-Mo monolithic fuel.
- Although TBP-based processes have been demonstrated with high burn-up spent fuel at Argonne and Savannah River National Laboratories, they have not been demonstrated with solutions containing high fluoride concentrations.
- The raffinate from this process (~300 kg on calcined basis), which contains high-fluoride along with Mo, Zr, Fe, Al, and noble metals, has no easy disposition path. If any aqueous processing is selected, means to prepare a suitable waste form and disposal path must be developed. (High Priority)

Criteria for Evaluating Option Category	**Rating**	**Comments**
Industrial Practicability		Because of the high fluoride concentration needed to dissolve this fuel, addition of AHA (which differentiates UREX from PUREX) is likely not necessary but may increase the robustness of the process in achieving purified uranium. The process will be similar to that used at the Idaho Chem Plant for reprocessing Naval fuel.
Technical Practicality	+	The uranium separation is certainly practical, robust, not complex, and can be performed with reasonable operating and capital costs; however, due to high fluoride content of the raffinate, waste form development is crucial.
Robustness	+	Will be adaptable to a wide variability of fuel specifications.
Degree of Complexity	0	With the possible exception of HLW treatment (due to the presence of high F and Mo), complexity of unit operations is minimal.
Operating Costs	0	Because most of the process is continuous, reagents are inexpensive, and equipment is mature, operating costs should not be significant.
Capital Costs	-	The facility will be expensive due to need for shielded cells, a robust off-gas system, and waste treatment operations.
Technical Maturity		The balance of the plant technologies, with the exception of waste treatment, are mature. Unknowns concerning the preparation and disposition of the HLW form lower its technical maturity.
Development Costs	-	Development and qualification of a suitable waste form will require significant development activities.
Schedule to Demonstrate	0	With the exception of HLW treatment, flowsheets can be demonstrated at the pilot scale in currently available facilities.
Process Maturity	0	All unit operations are mature technologies. Only concerns about the HLW waste form warrant R&D.
Waste		If glass were chosen as a disposal form, the high fluoride and molybdenum content of the raffinate would require development to design its formulation. If it were treated as was the Idaho Chem Plant waste (Ca addition and calcining), a final waste form would need to be developed for disposal.
HLW Disposition Costs	0	It is not clear at this point if waste volume would be significantly less than direct spent-fuel disposal. However, glass waste forms will be an acceptable waste form for a repository.
LLW Disposition Costs	0	More LLW will be generated than for the direct disposal option.
U Recovery		Uranium is recovered in high yield and purity. Discharged U-10Mo fuel enrichment is expected to range from 10 to 19.8%.

Category	**Rating**	**Comments**
Yield	+	Yield will be >99%.
Product Quality	+	Purity will meet recycle requirements.
<u>Cost/risk to Meet GTRI objectives</u>		All GTRI objectives can be met with this technology but for greater cost than for direct disposal.
Nonproliferation	0	Due to the high fluoride concentration in the feed and the possible addition of AHA to the scrub, Pu cannot be extracted in this process.
Materials Control and Accountability	0	Industry has developed means to do so.
Physical Security	0	Industry has developed means to do so.
<u>Cost/risk to Meet Environmental, Safety and Regulatory Requirements</u>		Can be met but at a greater cost than for direct disposal.
Environmental	0	Because this is a mature technology, means to meet criteria are well understood.
Safety	0	Because this is a mature technology, means to meet criteria are well understood.
Regulatory	0	Because this is a mature technology, means to meet criteria are well understood.

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6.1.2.2 Dissolve and blend with commercial fuel for reprocessing

Technology Overview[*]

The French La Hague plant is currently dissolving spent HEU aluminide and a limited amount of LEU silicide research reactor fuel and blending it into their high throughput spent commercial power-reactor-fuel feed to their plutonium extraction (PUREX) plant. In a 2005 publication, Herlet et al. describe experimental work on the dissolution of both irradiated and unirradiated U-Mo dispersion fuel. They also provide a scheme to reprocess the fuel at La Hague by adding the dissolved fuel to the dissolved power-reactor feed to the PUREX process. The spent LEU U-Mo fuel solution would be dissolved to ~15 g-U/L and fed into the conventional stream at a ratio of 1 part per 12 parts ~200g-U/L of dissolved spent power reactor fuel [1, 2].

Assuming a commercial fuel reprocessing plant were available when the monolithic U-Mo spent fuel needs to be dealt with, a similar approach could be taken. However, due to the zirconium barrier, the U-Mo monolithic fuel must be handled quite differently from the U-Mo dispersion fuel in the Herlet et al. study. The presence of a Zr-bonding layer for the monolithic U-Mo fuel puts a difficulty on aqueous processing of this fuel. The interface between the U-Mo fuel and the Zr layer is likely to contain U-Zr compounds that can react explosively when dissolved in nitric acid alone [3-8]. It is, therefore, common practice to dissolve fuels that contain U-Zr intermetallic compounds in nitric acid containing significant quantities of hydrofluoric acid. The following table shows a series of potential feed compositions (ignoring fission and activation products in the spent fuel) for U-Mo fuel dissolved in nitric acid and hydrofluoric acid. The compositions vary due to (1) whether the Al clad is dissolved with the fuel or dissolved prior to fuel dissolution using a sodium hydroxide solution or (2) whether ferric nitrate is added to the dissolver to increase the solubility of the fuel. At the free-fluoride concentration required to dissolve this fuel, the corrosion rate is estimated to be in the range of 0.78 mm/year. Determination of an “acceptable” rate of vessel corrosion is an empirical task that depends upon the construction of the vessel, the expected time of contact, and the desired service life of the system, among other factors. Such a determination requires an engineering assessment, although rates below 1.3 mm/ year are considered “low,” “mild,” or “adequate” in similar contexts [9, 10].

A further concern in this option is the effect of the fluoride and Mo being added in this side feed stream to the main oxide-fuel feed stream on the plants major processing line (process chemistry and corrosion of equipment and tanks) and on the waste glass that will be prepared from its raffinate. Under conditions where the bleed stream is diluted by a factor of greater than 100, it may be negligible.

Potential feed compositions for dissolved fuel plates (excluding fission and activation products) [3]

Feed #	U (g/L)	Mo (g/L)	Max. Mo g/L 100 °C	Al-6061 Clad (g/L)	Final HNO_3 (M)	Min. Final HNO_3	Max. Final HNO_3	F^- (M)	Al (M)	Fe (M)	Zr (M)	Si (M)
1	20	2.2	2.8	12.4	3	2.2	5	0.34	4.5E-01	6.2E-04	1.7E-02	3.4E-03
2	50	5.6	10	31.0	2	1	4	0.86	1.1E+00	5.0E-01	4.2E-02	8.4E-03
3	20	2.2	2.8	-	3	2.2	5	0.07	-	-	1.7E-02	-
4	75	8.3	9	-	2	1.5	3.5	0.25	-	5.0E-01	6.3E-02	-

Process Description

A reprocessing plant in the future will likely be designed to treat 500 to 2000 tons of spent commercial fuel per year. It is anticipated that HPRR spent fuel will be generated at ~2 tons/year. At first glance, the option has a

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[*] This write up assumes that the next commercial-fuel reprocessing plant built in the United States will be an aqueous solvent extraction plant (a modified form of PUREX). If the plant were to use pyrochemical processing, the chemically declad U-Mo fuel would need to be shredded and could then be added directly to the electrorefiner with impure metal generated in the first step of the oxide-fuel treatment (oxide reduction). This option would be equivalent in scoring to that given for the aqueous option.

great deal of appeal; the spent HPRR fuel stream would only comprise a fraction of a percent of the total uranium moving through the plant.

In this option, the spent U-Mo monolithic fuel would be dissolved separately from the commercial power-reactor oxide fuel. Depending on economic and technical optimization, the fuel could be dissolved in the four methods depicted in the above table. In the first feed option, the entire fuel plate is dissolved in a nitric-acid/hydrofluoric-acid mixture. It is quite possible that the hydrofluoric acid would be added intermittently during the dissolution of first the Al cladding and then the Zr barrier layer so that the free-fluoride in solution would remain low throughout the process. In the second feed option, ferric nitrate is added to the dissolver feed solution to increase the solubility of the fuel [3, 5, 6, 8]. In the third feed option, the fuel would be chemically declad using a sodium hydroxide solution with or without sodium nitrate (to diminish H_2 generation), leaving only the Zr barrier and the U-Mo fuel to be dissolved. The fourth option is similar to the third except that ferric nitrate is added to increase the solubility of the fuel.

The free fluoride in the dissolver solution must be held low to (1) limit corrosion of the dissolver vessel and (2) to assure that rare-earth and TRU fluorides do not precipitate during dissolution of the fuel. Once the fuel is dissolved, the dissolver solution will need to be filtered to remove any undissolved solids. At this point, the solution will be blended with the dissolved power-reactor fuel to feed the solvent extraction process. The uranium will be stripped from the solvent and a raffinate stream will contain the fission products and all or some TRU elements from both fuel types; it will also contain the fluoride that entered with the U-Mo fuel stream. The fates of the Pu and Np in the solvent-extraction process depend on the design of the commercial plant. It should be noted that in this option the ~13% enriched U from processing the spent U-Mo fuel would not be recovered separately. When blended into the spent power reactor stream at a ratio of 1/200, addition of the ~13% enriched U from the U-Mo fuel would only increase the enrichment of the total U product from ~0.9 to ~0.96.

Identified Technology Gaps

- Earlier GTRI fuel fabrication capability (FFC) work performed on processing this fuel was directed toward recycling of fabrication scrap, so very little emphasis was given to the effects of dissolution conditions on the activation and fission products that would be present in spent fuel. The solubility of rare-earth and some other fission- and activation-product fluorides is quite low. An investigation is required to assure that these fluoride salts will not precipitate during dissolution. (High Priority)
- Significant data exist for dissolving similar fuels and cold surrogates[3], but dissolution of highly irradiated fuel of this type has not been done (with the possible exception of some analytical dissolutions at the Idaho National Laboratory). Corrosion tests under conditions necessary to dissolve and store this fuel need to be performed for stainless steel or other potential materials of construction.
- There are two options for dissolution—(1) pre-dissolution of the Al cladding using sodium hydroxide solution or (2) dissolving the intact fuel in a nitric-hydrofluoric acid mixture. Bench-top proof-of-principal experiments have shown the efficacy of both approaches. A technical/economic study needs to be performed to decide on the best approach. (High Priority)
- The addition of ferric nitrate will increase the solubility of this fuel by three-fold (from ~20 to 50-75 g-U/L). A technical/economic study needs to be performed to rate the pros and cons of ferric nitrate addition.
- In the French study, the dissolved U-Mo stream was conceived to be blended into the main process stream at a ratio of 1/12. A blending ratio for this fuel in a yet-to-be built/conceived reprocessing facility for power-reactor fuel needs to be developed. In this case, because of the significant concentration of fluoride in the U-Mo fuel solution, the blending ratio is far harder to develop. The addition of fluoride (and the additional Mo in this fuel) to the main stream could have a significant effect on both processing and waste treatment.

A thorough investigation is required to learn the efficacy of this approach and to develop a blending ratio that will have a negligible effect on the reprocessing facility and the waste-glass formulation. (High Priority)

Criteria for Evaluating Option

Category	**Rating**	**Comments**
Industrial Practicability		
Technical Practicality	+	Because the dissolved U-Mo fuel is a small fraction of the total plant feed, its effect on operation should be minimal.
Robustness	+	Because the dissolved U-Mo fuel is a small fraction of the total plant feed, its effect on operation should be minimal.
Degree of Complexity	0	The additional U-Mo storage, fuel-dissolution, and blending area will add only a fraction to the complexity of the entire reprocessing plant. It will also be minor compared to that of a complete aqueous processing plant for U-Mo fuel.
Operating Costs	0	Operating costs for this addition will be minor compared to the operating costs for the entire plant and will be less than those of a stand-alone processing plant for U-Mo fuel. However, a significant “users fee” could be imposed for processing this fuel.
Capital Costs	+	Capital costs for this addition will be minor compared to the capital costs for the entire plant and will be far less than those of a stand-alone processing plant for U-Mo fuel.
Technical Maturity		
Development Costs	-	The addition of fluoride and Mo to the main stream may require significant waste form development efforts due to the relative low solubility of these elements in typical glass waste forms. This should be a smaller task than developing a waste form for the stream without dilution. Nevertheless, development of a compatible waste form is required. As with other aqueous processing options, optimizing dissolution of this fuel is required.
Schedule to Demonstrate	0	The development path is clear and should not affect the schedule for disposition of this spent fuel.
Process Maturity	+	A pilot-scale demonstration of the aqueous processing could be run now. Plant equipment could be selected, and the plant could be designed now. Waste-form development will require verification/validation of known parameters.
Waste		
HLW Disposition Costs	0	Should only add fractionally to the HLW cost for the reprocessing plant.
LLW Disposition Costs	0	Should only add a minimal fraction to the LLW cost for the reprocessing plant.
U Recovery		
Yield	+	Recovery would be >99%.

Category	Rating	Comments
Product Quality	0	Purity would be that necessary for recycle. It scored lower because the ~13% enriched U recovered from processing the spent U-Mo fuel separately would not be available; If blended into the spent power reactor stream at a ratio of 1/200, it would only increase the enrichment of the total U product from ~0.9 to ~0.96.
<u>Cost/risk to Meet GTRI objectives</u>		
Nonproliferation	0	This small stream would add minimal additional cost/risk to the large plant.
Materials Control and Accountability	0	This small stream would add minimal additional cost/risk to the large plant.
Physical Security	0	This small stream would add minimal additional cost/risk to the large plant.
<u>Cost/risk to Meet Environmental, Safety and Regulatory Requirements</u>		
Environmental	0	Would add minimum cost/risk to the large plant.
Safety	0	Would add minimum cost/risk to the large plant.
Regulatory	0	Would add minimum cost/risk to the large plant.

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6.1.2.3 Justification for Eliminating Dissolve Fuel and Purify U by Crystallization

Technology Overview
One alternative for the recovery of U from high performance research reactor fuels is the crystallization of uranyl nitrate complexes at low temperatures from nitric acid solution. To utilize this process, the fuels must first be dissolved and clarified to produce a homogenous solution for U recovery. Two options can be used to dissolve the fuel. The entire fuel element can be dissolved in nitric acid with sufficient fluoride to dissolve the Al cladding and the U-10Mo alloy, or the Al cladding can first be removed using a caustic solution [1]. The dissolution of the Al cladding is represented by equation (1) which was empirically derived based on laboratory-scale studies of the process used at the Savannah River Site to remove Al cladding from depleted U targets. The $NaNO_3$ is used to suppress the evolution of H_2 by altering the reaction so that primarily NH_3 is produced instead of H_2 [2].

$$\mathrm{Al} + 0.7\mathrm{NaOH} + 0.58\mathrm{NaNO_3} + 0.11\mathrm{H_2O} \rightarrow \mathrm{NaAlO_2} + 0.28\mathrm{NaNO_2} + 0.3\mathrm{NH_3} + 0.009\mathrm{H_2} \qquad (1)$$

During the dissolution of the U-10Mo alloy and the Zr-bonding layer, two limitations must be addressed to ensure safe and efficient operations. When dissolving U metal that is metallurgically bonded to Zr metal using a nitric acid solution, it is important to maintain at least a 4:1 molar ratio of fluoride to Zr to mitigate the formation of a $U\text{-}Zr_2$ intermetallic compound, which is highly explosive [3-4]. Although U-Mo alloys containing up to 10% Mo dissolve rapidly in nitric acid, [5] the formation of uranium molybdate solids can be an issue for U concentrations greater than approximately 50 g/L [1]. To prevent the formation of solids, ferric nitrate can be added to the dissolver solution to complex the Mo and prevent the precipitation of U [1, 5, 6].

A variety of approaches have been evaluated to recover uranyl nitrate from light water reactor fuel using crystallization techniques. Low-temperature crystallization methods were studied in Germany and Japan [7, 8], and the use of a continuous crystallizer was evaluated in work sponsored by the United States (US) Department of Energy's Advanced Fuel Cycle Initiative [9]. The uranyl nitrate crystallization processes were considered as an alternative to the PUREX solvent extraction system for U recovery. Bench-scale work on uranyl nitrate crystallization in Germany showed hexavalent actinides can be selectively crystallized (95% yield) from cold nitric acid solutions (-20 to -40 °C). Decontamination factors (DFs) of 100-1000 were obtained for Rb, Sr, Y, Zr, Mo, Ru, Rh, Pd, Cd, Te, Cs, Ba, and La after two successive batch crystallizations. The Japanese proposed a process that would cool the dissolver solution to 10 °C and recover approximately 60% of the U as nitrate crystals. In bench-scale test solutions containing thirteen fission product elements, DFs of 2 to 9 were obtained with no crystal washings, and 10 to 100 were obtained after three washes [9].

In the US work, the Los Alamos National Laboratory evaluated the use of a continuous, adiabatic reduced-pressure crystallizer. A circulating loop was used as the crystallizer vessel. The solution was cooled and concentrated by the evaporation of a portion of the nitric acid and water at reduced pressure. Uranyl nitrate hexahydrate was crystallized from the supersaturated solution. The distilled acid would be recycled for use in fuel dissolution, for crystal washing, or for dissolving washed crystals. A model developed for the continuous crystallization process showed improved DFs compared to batch processes; however, the program was terminated before measurements could be performed using prototypical equipment [9].

Rejection Criteria
During the preliminary screening, the dissolution of high performance research reactor fuel and purification of the U by crystallization was screened-out due to low maturity and high complexity of the process. The purification of U by crystallization has not been pursued beyond the laboratory scale, and prototypical equipment has not been demonstrated. Methods for processing the waste streams and the development of final

waste forms have not been addressed. Multiple recycle and waste streams would be generated during U recovery operations, increasing the complexity of the process.

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6.1.2.4 Justification for Eliminating Carbonate Reprocessing

Technology Overview

This process has been proposed and studied as an alternative to solvent extraction for the reprocessing of commercial oxide power reactor fuel. The uranium in the fuel is dissolved by a combination of hydrogen peroxide (to oxidize UO_2 to the carbonate-soluble UO_2^{2+}) and alkaline carbonate solution. It is proposed that the solubility of most other fuel components is far lower than that of uranyl ion, so this method will provide decontamination of the uranium from other fuel components. A short article and the references therein provide a review of the technology [1]. This work has been primarily pursued in Japan, the Republic of Korea, and at the Los Alamos National Laboratory.

The use of carbonate for reprocessing HPRR U-Mo monolithic fuel has never been examined and faces several challenges. The biggest challenge is that this fuel has almost nothing in common (physically or chemically) with UO_2 fuel; the research and development required to even ascertain if it would be applicable would be significant. Further, neither zirconium nor Mo is soluble in carbonate solution, and it is difficult to imagine how the fuel could be pretreated to open the uranium to dissolution.

Rejection Criteria

Since the development of technology would be costly and lengthy, with no clear advantages to other more mature options, this option is not recommended for further consideration.

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6.2 Stabilization Treatment without U Recovery

6.2.1 Melt Processing

Melt Process Technology Overview
Melt Process (MP) technology provides a disposal form (waste form) for the ultimate disposition of fuel discharged from a reactor into a permanent repository without recovery or recycle of the fuel materials or components, but following a melt process, with or without additives, such as depleted uranium, neutron absorbers, and alloying components. The aim of a MP is to improve the waste form that would result in one or more of the following features compared to the direct-disposed fuel:

- reduce the volume of the disposal form
- render the form fully non-attractive
- render the form criticality-resistant even under full reconfiguration and optimal moderation
- create a highly-durable form that may be credited with (slow) dissolution and radionuclide release behavior in a repository performance assessment.

An advantageous feature of MP vis-à-vis the Direct Disposal (DD) option, another option without materials recovery, is that the characteristics of the disposal form are "erased" with the treatment. As an example, the size, shape, and enrichment of uranium in the spent fuel are transformed in a melt process with added depleted uranium to create a new disposal form. As an alternative to (or, as may be required, in addition to) accurate records of production/depletion of isotopes in the fuel as well as off-gas losses during processing, the characteristics of the disposal form (such as the isotope inventory that is expected to be needed for manifestation for repository disposal) can be ascertained in an assay step in the treatment process.

The specific MP treatment for a fuel will be directly tied to the initial composition of the fuel. The actinide and fission product species in the discharged fuel depend on the initial enrichment, burn-up, and cool-down time, but their inventory can be bounded for a MP system design. An example of a reduced-enrichment USHPRR fuel design is that of the ATR Full Element demonstration plate that is comprised of a U-10Mo alloy at 19.8 wt% U enrichment with a Zr coating at 5 wt%. The fuel is clad in aluminum. The composition of a single fuel element is [1]:

- U-10Mo: 460 grams
- Zr: 25 grams
- Al: 245 grams.

This design is amenable to a melt process to create an alloy disposal form. Ceramic forms that could be created (e.g., treatment to form oxides of the fuel materials with addition of glass frit), although conceivable, would require a more involved treatment compared to a melt process to create an alloy.

The full set of component technologies (melt process, furnace system, off-gas bed design, and disposal form or "waste form" characterization) was previously developed for a "melt-dilute" system to produce an alloy disposal form for U-Al, U_3O_8-Al, and U_3Si_2-Al research reactor "dispersion fuels" in aluminum matrices and clad in aluminum. The melt process added aluminum and depleted uranium to the fuel melt [2]. This process was flexible as demonstrated by an option for treating cermet fuel where additives could reduce the cermet fuel (U_3O_8-Al) to allow uranium mixing with depleted uranium (DU) so as to reduce enrichment and create a final alloy disposal form characterized as dispersoids of intermetallic UAl_x in aluminum. The technology readiness was at a medium level, and a facility to demonstrate a batch melt of up to four full-sized actual spent materials test reactor (MTR) assemblies was established. Further development of this technology was suspended [3] based on DOE direction. As part of the technology development, an assessment of the repository "disposability" of the melt-dilute form was prepared [4].

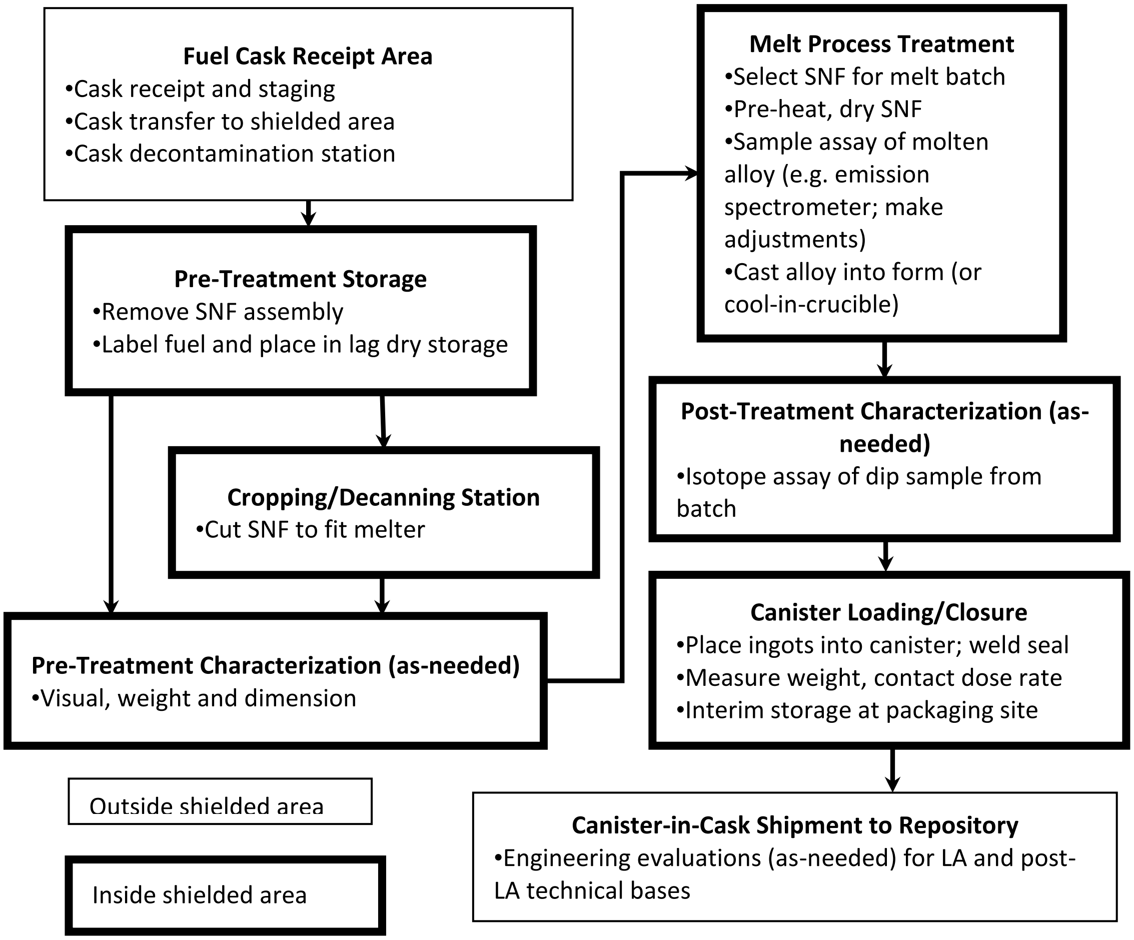
It is conceivable to use the MP treatment for future USHPRR fuel. One option is an alloy based on aluminum and a target process at the Al-13%U eutectic ($T_{eutectic, cooling}$ = 646°C) to allow a low-temperature melt process to limit off-gas challenges. However, the behavior of the Zr interlayer would need to be ascertained to understand the effect in formation of the Al-U alloy or formation of separate phases. Additionally, the effect of Zr on the melting process would need to be understood specifically if it results in an increase in melt temperature. The elimination of attractiveness and criticality issues with a degrading disposal form in a repository could be fully realized with a treatment with added depleted uranium to reduce U-235 enrichment; however, volume reduction would not be achieved for a target alloy disposal form at the eutectic due to the large addition of aluminum needed in the process. Further, the corrosion resistance and overall durability of this disposal form would not be expected to be improved over that of U-10Mo.

Other metal waste forms could be developed with a high durability, perhaps significant to allow credit in a repository Performance Assessment. This would make the MP disposal form superior to the DD form. As an example, work has been done to develop an alloy waste form from primarily the metallic specie waste stream feeds from aqueous and electro-metallurgical reprocessing schemes [5]. Corrosion testing to date has shown this alloy waste form, based on iron with chromium additions, to be very durable. A high temperature (~1600°C) melt process is used for this waste form, but there may be drawbacks; for example the challenges in the variety and magnitude of the species needed to be captured in the off-gas subsystem, in a system to melt process the full spent fuel assembly with its full complement of radioisotopes.

As stated, the technologies needed for a MP treatment for existing research reactor fuel designs were previously established at a medium maturity level. The development of an alloy disposal form for USHPRR fuel is feasible and may be found to have a positive cost-benefit advantage when considering safety, regulatory, and security topics in a repository system for fuel disposal. However, the repository requirements and disposal form characteristics against repository requirements are somewhat uncertain. Further, MP treatment schemes to produce a glass or a cermet form could be explored.

Melt Process Description

A suggested path to take USHPRR fuel and produce a disposal form is shown in figure below.



Basic Steps to Handle and Melt Process Fuel and Load into Road-Ready Canisters that Would Meet Repository Requirements

The MP treatment would be a simple batch process with the following process steps:

1. Remote loading of assemblies in the crucible in an induction furnace[i]
2. Loading of other materials include alloy additions, neutron absorbers, and/or depleted uranium
3. Remote staging of off-gas filter component on top of furnace
4. Heat-to-melt process (TBD) and cool the melt to produce ingot (one-shift operation)
5. Load (stack) ingots into disposal canister
6. Store canisters (like HLW glass canisters at SRS, a mature operation) at storage location until transportation to repository.

To illustrate the concept, the following figure shows the furnace unit used in the laboratory phase of the melt dilute technology development at Savannah River. The two steps to the melt process were heat-to-melt, followed by stirring of the melt pool via magnetic coupling to the oscillating magnetic field of the induction melter operated in a low power mode.

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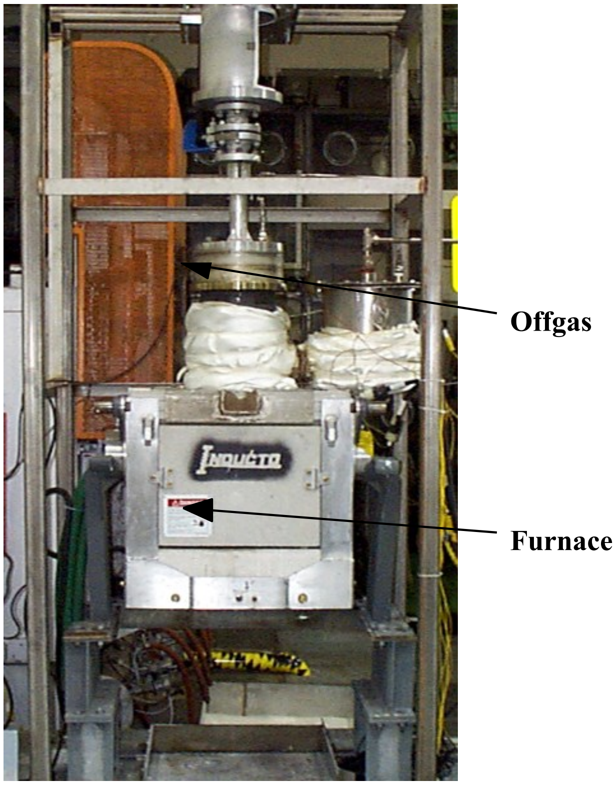
[i] The furnace would have a non-reactive gas lance with a gas stream flow exhaust using a blower pump on the off-gas train. The blower pump would effectively maintain a negative pressure in the furnace chamber and thereby a measure of containment external to the enclosed furnace. The exhaust is through an off-gas system such as a zeolite bed and HEPA filters and/or sand filters. Double confinement is provided by a metal shroud surrounding furnace enclosure which would be at negative pressure to the room.

A key feature was the off-gas bed. The melt system design must consider the process conditions, especially melt process temperatures in the design of the off-gas capture system. [2] A non-reactive gas purge rate and bed used for this 850°C melt process was optimized for cesium capture.



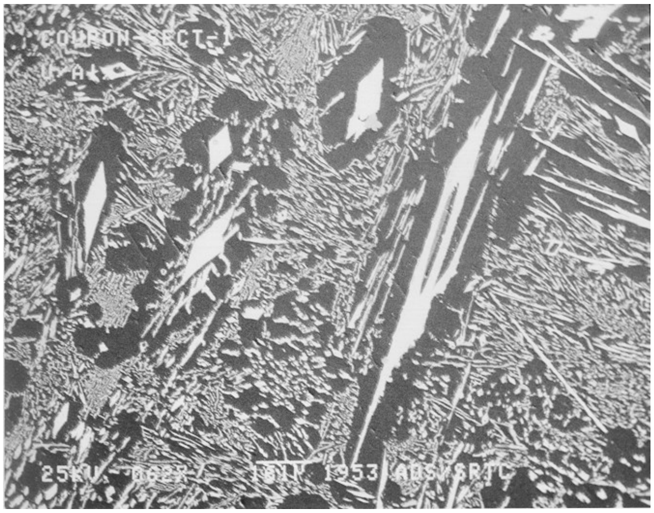
Induction Furnace Used to Produce Surrogate MD-SNF Ingots (DU with aluminum) [2] [negative pressure confinement box removed to view furnace]

The disposal form was an alloy ingot. The first figure below shows an example of a surrogate materials ingot produced in the laboratory. The resultant microstructure that was uniform across the ingot is shown in the second figure below.

Similar to the DD fuel option, the dry-stored ingot would be subject to interim dry storage requirements, if storage longer than lag storage at the canister packaging facility would be needed. The dry storage facility is expected to be an NRC-licensed facility, subject to the requirements under 10 CFR Part 72 [6].

Tests and analyses for the repository disposal may be needed, and performance data such as corrosion behaviour under repository-relevant conditions is expected. Testing and development for the MP disposal form would be common to the USHPRR fuel, regardless of initial fuel condition. That is, the MP disposal form for repository disposal could be produced to be relatively uniform even across the USHPRR fuel feeds.

Surrogate MD-SNF Ingot Produced in an Induction Furnace without Carbon Steel Liner [2]



General Microstructure of the Binary U-Al MD-SNF Form [2]

Identified Technology Gaps

The following are the major technology need areas for furthering the maturity of a MP path for USHPPR fuel disposition.

- The selection of an alloy or other disposal form material system would need to be identified to further identify the technology gaps, especially the processing scheme. If an aluminum-based is selected, a medium technology level of readiness exists. If an iron-based system or other material system is selected for volume reduction and enhanced product durability, a low level of technology readiness exists. Consideration of desired metrics for attractiveness, criticality, and disposal form durability would need to be made for the disposal form of the new design USHPRR fuel.
- The effect of the Zr interlayer on the melt dilute process and alloy selection will also need to be determined. Specifically, the effects of Zr on phase formation and melt temperature and melt processing will need to be ascertained.

- Treatment system design that may require development activities include: selection of furnace melt technology (arc melt; induction furnace; box furnace), and off-gas system design/development (especially needed for volatiles in high temperature (>800°C) MP treatment processes).
- Repository performance information such as characterization tests to determine corrosion/release rates in range of repository environments and attribute tests for interpretation of the characterization tests will be needed for the new disposal form(s) for the MP treatment of USHPRR fuel.

Criteria for Evaluating Option **Category**	**Rating**	**Comments**
Industrial Practicability		
Technical Practicality	+	The full set of component technologies for melt treatment of research reactor fuel has been investigated. A melt process has been demonstrated for aluminum-based RR fuel, and a project was executed, up until full assembly hot demonstration, for an aluminum-based disposal form. The effects of Zr and Mo on phase formation and processing will need to be determined.
Robustness	+	MP could handle various fuel conditions, and the batch process could be tailored to make a consistent disposal form.
Degree of Complexity	+	Alloy fabrication via melt processing is a very mature technology (foundry-like operation). Additionally, off-gas capture in radiological materials processing is well-established.
Operating Costs	0	The processing of radiological materials and the accompanying shielding, confinement, and contamination controls would make operating costs significant.
Capital Costs	-	The costs of design and fabrication of a facility with remote operations and a robust off-gas system and secondary waste handling would be very significant.
Technical Maturity		
Development Costs	-	An alloy system and melt process would be needed to be selected, tested, and demonstrated for the U-10Mo fuel.
Schedule to Demonstrate	0	
Process Maturity	0	
Waste		
HLW Disposition Costs	0	MP can achieve waste volume reduction up to 35% original volume of a materials test reactor assembly if no significant additives are needed. However, it is expected that additives will be necessary and the volume reduction will be much less than 35%.
LLW Disposition Costs	0	Secondary wastes, including those from off-gas beds, may be significant.

Category	**Rating**	**Comments**
U Recovery		Uranium is not recovered.
Yield	-	
Product Quality	N/A	
Cost/risk to Meet GTRI objectives		
Nonproliferation	+	MP can reduce uranium enrichment to any target level. U is not separated.
Materials Control and Accountability	0	Sampling of the melt provides a simple, accurate method for materials inventory that would support the MC&A manifests.
Physical Security	0	
Cost/risk to Meet Environmental, Safety and Regulatory Requirements		
Environmental	0	The submission of the license application (LA) for the Yucca Mountain Project (YMP) provides a template for information needed for a repository. Testing and evaluation of the USHPRR fuel in a MP disposal form is expected to be needed, but the cost/risk to meet requirements should be relatively low with a standardized disposal form.
Safety	0	
Regulatory	0	

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6.2.2 Justification for Eliminating Mechanical Consolidation

Technology Overview
Mechanical Consolidation (MC) can produce a waste form for the disposition of fuel discharged from a reactor with no recovery or recycle of the fuel materials or components. The two basic MC processes are simple mechanical compaction and a more involved process such as chopping and blending the fuel with neutron absorbers prior to mechanical compaction.

The drivers for consideration of MC as a Stabilization Treatment are the following:

- reduce the volume of the disposal form
- render the form fully non-attractive
- render the form criticality-resistant even under full reconfiguration and optimal moderation
- create a highly-durable form that may be credited with (slow) dissolution and radionuclide release behavior in a repository performance assessment.

Mechanical compaction of reactor fuel materials and components alone would likely result in only modest reduction in waste volume. Adding neutron absorbers as part of a chop-and-blend process would achieve criticality resistance in a disposal setting only if congruent dissolution characteristics of the fuel and absorber materials were realized. Furthermore, it is highly unlikely that any credit for reduction in attractiveness or improvement in durability of the MC form could be achieved over that of the DD form.

Rejection Criteria
The simplicity of MC requires limited technology development except for remote crushing and/or chopping methods. The costs and complexity of this treatment would be minimal.

However, MC cannot produce a waste form that is consistent in quality and that would improve the attractiveness compared to the untreated fuel. Also, the safety, environmental, and regulatory issues of a readily dispersible product would be expected to eliminate this disposal form from acceptance. For these reasons, MC to produce a disposal form without fuel recovery is not considered a viable option for the disposition of USHPRR fuel.

6.2.3 Dissolve Fuel and Immobilize Wastes for Disposition

Technology Overview

In this option, one of the aqueous processing routes described above would be employed to dissolve the Al cladding and the U-Mo fuel. The resulting "waste" solutions would be treated separately or combined for immobilization for subsequent disposal in a federal repository. It would appear to be most practical to separately treat the dissolved Al cladding and the dissolved U-Mo fuel. It is likely that the Al cladding product could be disposed of as low-level waste according to current regulations.

There are several immobilization options for the dissolved U-Mo fuel. These generally fall into three categories: glasses, crystalline ceramics, and glass composite materials (GCM). Tailoring the waste form chemistry is done to maximize waste loading, facilitate waste form processing, and ensure long-term durability of the waste form. Final disposition of the immobilized form would be anticipated to be in a federal geologic repository and would need to meet all acceptance requirements for repository disposal.

Process Description

The first step for this option is to dissolve the Al cladding. It is likely that the Al could be removed using a caustic solution and the dissolved solution could be treated for disposal as a low-level waste based on current regulations as described previously.

Once removal of the cladding is completed, the remaining fuel is dissolved using one of the aqueous dissolution methods discussed previously. The resulting (likely acidic fluoride) solution would include U, Mo, Zr and fission products. It is also likely that chemicals may be added during the dissolution process to improve the process. For example, to prevent the formation of solids following dissolution, ferric nitrate could be added to the dissolver solution. Moreover, as discussed in the options above, a significant concentration of fluoride may need to be added to safely remove the Zr interlayer.

The "waste" composition would need to be specifically known to tailor a waste form to immobilize the fuel dissolution product. For this study, a general knowledge of the major constituents of the waste can be used to assess the various waste form options and chemistries.

Borosilicate glass (BSG) is the current baseline for HLW immobilization in the U.S. and in several other countries [1]. Glasses have been developed and tested and fabrication processes have been demonstrated for a wide variety of applications including:

- High-level wastes resulting from defense and commercial fuel processing
- Surplus plutonium
- Am and Cm solutions

For this application, a BSG composition could likely be tailored for the waste. However, issues with the relative solubility of U, Mo and F could significantly limit waste loading.

Iron-phosphate glass (IPG) could be an alternative for this application. The IPG is an attractive option due to the relatively high solubility of components, such as molybdate and fluorine in the phosphate liquid. While most phosphate-based glasses are relatively low in chemical durability and are not suitable for nuclear waste forms, two phosphate-based glass families have shown superior durability: the alkali-alumino-phosphate family and the alkali-iron-phosphate families. Due to the relatively low maturity of this waste form compared to BSG, a greater effort may be required for qualification of the IPG glass for repository disposition. Additionally, phosphate glass can present processing challenges due to the corrosiveness of these compositions.

In the vitrification process, the feeds are prepared for vitrification by mixing the feeds with glass frit or appropriate glass forming chemicals, melted in a high temperature melter, cast into a container, and stored. The gases generated from feed preparation and melting processes are treated appropriately. The Joule Heated Melter (JHM) and Cold Crucible Induction Melter (CCIM) are the primary melter technologies used presently. Joule-heated ceramic melters are currently used in radioactive operations for treatment of HLW in the U.S. The CCIM technology is currently receiving increased interest due to their ability to process at higher temperatures, minimize melter corrosion by use of a skull layer to contain the melt, and allow processing of crystalline inclusions in the melt. The CCIM technology is in use in France, Russia and South Korea.

Ceramic-based (or more appropriately, crystalline) waste forms retain the radionuclides by tailoring the composition to create certain minerals (i.e., unique crystalline structures) that will host the radionuclides by binding them in specific crystalline networks [1]. These synthetic minerals are made in a manner to reflect natural minerals that host natural radioactive species such as uranium, nonradioactive isotopes of the fission products, or both.

The primary ceramic-based materials suitable for consideration are from the Synroc and monazite families of compositions. In general, Synroc (i.e., synthetic rock) is an advanced synthetic crystalline ceramic comprised of geochemically stable titanate-based minerals, which have immobilized uranium, thorium, and other natural radioactive isotopes in the environment for millions of years. The mineral monazite, $(Ce,La,Nd,Th,U)PO_4$, is a mixed lanthanide orthophosphate that is found as either a pure monoclinic or tetragonal single crystal or as a specific mineral in an assemblage of phases in granites and pegmatites. Several other ceramic forms could be considered for immobilization of the U-Mo fuel waste stream. These include: murataite ($[Y,Na]_6[Zn,Fe]_5[Ti,Nb]_{12}O_{29}[O,F]_{14}$) and brannerite ($[U,Ca,Ce][Ti,Fe]_2O_6$). Difficulty is often encountered in ceramic waste forms when trying to accommodate a wide variety of waste components into the crystalline structure. Ensuring partitioning of the elements in the correct mineral phase is often challenging. For this application, partitioning of Mo, F and fission products such as Cs in the ceramic phases could be problematic.

Ceramic materials can be formed using a wide range of processes including cold pressing and sintering, hot uniaxial pressing (HUP), hot isostatic pressing (HIP), and CCIM. Of these various processing routes, HIP is the most developed for production of Synroc-type materials. Throughput limitations have hampered the utility of ceramic processes for large-scale waste treatment operations. The use of the CCIM technology to produce ceramic forms may provide a solution to this limitation.

Glass composite materials (GCM) describe polyphase materials where an amorphous matrix and crystalline phases comprise the waste form. These waste forms take advantage of the attributes of a glassy waste form to accommodate a wide range of components into the amorphous structure and the attributes of ceramic phases to immobilize specific elements of concern into a highly stable crystalline form. Recent work by Crum, et al. has demonstrated the ability of a GCM to immobilize a surrogate commercial fuel reprocessing stream [2]. The GCM composition was able to accommodate typical low glass solubility phases such as Mo into a powellite crystalline phase and Zr into a cerianite phase. A GCM composition may be highly suitable for the dissolved U-Mo fuel waste stream if a suitable host phase for U could be identified.

GCMs can be processed similarly to glasses by combining the waste stream with the appropriate additives and melting. However, it may be necessary to include a post-melting step to either cool the GCM according to a prescribed schedule to form the targeted crystalline phases or to post-heat treat the resulting glass product. Neither of these crystallization processing routes has been demonstrated at an appropriate scale for radioactive waste materials.

Identified Technology Gaps

- The appropriate waste form and waste form chemistry will need to be defined based on the actual composition of dissolved U-Mo fuel waste stream. A GCM appears to be the most attractive waste form candidate, however, incorporation of a high concentration of U into the waste form could be challenging.
- The qualification of the waste form for geologic repository disposal would be required. Qualification of GCM or ceramic waste forms would require an additional effort since no basis for qualification of these waste form-types currently exists.
- A waste form process would need to be developed and demonstrated to ensure that a consistent, high quality waste form could be produced.

Option Evaluation

Category	**Rating**	**Comments**
<u>Industrial Practicability</u>		
Technical Practicality	+	Although HLW streams are currently immobilized using a glass waste form, the U-Mo fuel waste stream could present unique challenges.
Robustness	+	Although HLW streams are currently immobilized using a glass waste form, the U-Mo fuel waste stream could present unique challenges.
Complexity	+	Multiple unit operations involving dissolution and waste treatment.
Operating Costs	0	Multiple unit operations required.
Capital Costs	-	High construction cost of dissolution and waste treatment facility.
<u>Technical Maturity</u>		
Development Costs	-	Fairly significant waste form and process development efforts would be required but there is some previous work that can be leveraged.
Schedule to Demonstrate	0	Fairly significant waste form and process development efforts would be required but there is some previous work that can be leveraged.
Process Maturity	0	Likely no show stoppers but development and demonstration is required.
<u>Waste</u>		
HLW Disposition Costs	0	The disposition of multiple liquid and solid waste streams is required and volumes are expected to be large compared to other options.
LLW Disposition Costs	0	To optimize the process, new waste forms may need to be developed and will require qualification.
<u>U Recovery</u>		
Yield	-	No U will be recovered.
Product Quality	N/A	

Category	**Rating**	**Comments**
Cost/risk to Meet GTRI objectives		
Nonproliferation	+	With no separation of U, it is expected that nonproliferation goals will be easily satisfied.
Materials Control and Accountability	0	No separation of U.
Physical Security	0	No separation of U.
Cost/risk to Meet Environmental, Safety and Regulatory Requirements		
Environmental	0	Construction of a new reprocessing facility will likely require eliminating essentially all emission which will increase cost.
Safety	0	Dissolution and waste form processing has been safely conducted at production scale.
Regulatory	0	Regulatory issues are expected to be easily met based on previous similar industrial processes

References

1. D. Gombert II, “Global Nuclear Energy Partnership Integrated Waste Management Strategy Waste Treatment Baseline Study - Volume I,” GNEP-WAST-AI-RT-2007-000324, U.S. Department of Energy, 2007.
2. J.V. Crum, L. Turo, B. Riley, M. Tang and A. Kossoy, “Multi-Phase Glass-Ceramics as a Waste Form for Combined Fission Products: Alkalis, Alkaline Earths, Lanthanides, and Transition Metals,” Journal of the American Ceramic Society, **95** [4], 1297-1303, 2012.

6.2.4 Justification for Eliminating Metal Oxidation (w/ or w/o immobilization)

Technology Overview

The thought behind this approach is that Al, U, and Zr may be considered active metals by the repository waste acceptance criteria and therefore ineligible for disposal. If this is the case, the spent fuel could be combusted in a controlled manner to put the metals of the fuel into stable oxide forms. The oxide could then be compacted for direct disposal or fed to a glass melter.

The main components of the oxide product would be Al_2O_3, UO_2/U_3O_8, MoO_3, and ZrO_2. Most of the fission products also would be oxidized. For this operation, the off-gas system would need to be equivalent to that of a waste-glass melter. Radio-Xe and –I, some or most of the Tc, Cs, Ru, and Mo also would volatilize during combustion.

The ignition temperature of uranium and uranium alloys is in the range of 500-600°C [1]. Al and Zr ignition will be at a higher temperature [2]. Zr sponge has been burned successfully in a laboratory-scale rotary kiln [3].

It is envisioned that the fuel plates would be chopped into ~1-2 cm slices and fed into the rotary kiln. The kiln would be heated to ~700°C to initiate the combustion of uranium. As the uranium burned, the temperature would increase inside the kiln to the ignition temperatures of Zr, Al, and Mo. Once the kiln cooled, the oxide powder would be removed from the kiln and configured into a final waste form.

Rejection Criteria

This process is fairly simple compared to aqueous processing but needs significant development. Controlled ignition and burning of radioactive material does trigger a large number of safety and environmental concerns.

References

1. J. G. Schnizlein, L. Baker, Jr. and J. D. Bingle, The Ignition of Binary Alloys of Uranium, Journal of Nuclear Materials, 20 (1966) 39-47.
2. P. L. Harrison and A. D. Yoffe, The Burning of Metals, Proc. R. Soc. Lond., A 1961, 261, 357-370.
3. Paul M. Lemieux and David W. Pershing, Design and construction of a rotary kiln simulator for use in studying the incineration of hazardous waste, Rev. Sci. Instrum. **60**, 2768 (1989); doi: 10.1063/1.1140656.

6.3 Direct Disposal

In the direct disposal category, no fuel treatment (other than to stabilize the fuel for storage) or attempt to recover uranium is made. Two unique options were identified in this category and are described in the sections below.

6.3.1 Direct Disposal—Domestic Site

Direct Disposal Technology Overview
Direct Disposal (DD) is the ultimate disposition of fuel discharged from a reactor into a permanent repository without recovery or recycle of the fuel materials or components. This path would involve component technologies for interim wet or dry storage, drying and/or treatment for disposal, packaging into standard disposal canisters, transportation, and placement in a repository meeting the acceptance criteria for the waste package. The packaging of the fuel may require neutron absorbers internal to the fuel canister to meet criticality control requirements for recovery, if needed, during storage; accident conditions in transportation, and/or avoidance of criticality in the repository as part of the waste acceptance criteria. Most of the component technologies for DD of USHPRR fuel leading up to the step to package the fuel for ultimate repository disposal are mature. However, the repository requirements and spent nuclear fuel (SNF) characteristics against repository requirements are somewhat uncertain. With regard to non-proliferation consideration, the spent fuel, albeit LEU (<20%), would be retrievable in storage and in the pre-closure repository disposal period.

The practice for the USHPRR to date (present fuel design) has been to store the discharged fuel in good quality water at the reactor site, and then to transfer and to continue to store the fuel (aluminum-clad) in wet storage with the exception that the spent ATR fuel at Idaho is dried and placed in non-sealed dry storage in the Fuel Storage Building. It is important to maintain good water quality in interim wet storage since aluminum fuel is susceptible to rapid localized corrosion attack if exposed to aggressive or poor quality water [1].

Due to the irradiation of the fuel and post-discharge storage, the fuel is altered from its as-fabricated condition both physically and isotopically. The characteristics of the spent fuel at this point are important to transportation, storage, and disposal systems so as to address system operation, safety, and environmental impact. At present, quantification of research reactor spent fuel attributes for receipt and storage are formally required by the U.S. Department of Energy for receipt and storage when transporting the fuel from the reactor site to the storage site; the information is compiled in an “Appendix A” document [2]. The Appendix A document includes a detailed description of the elements that make up the fuel assembly including geometric and mass information for the fuel core and the cladding, and the balance of hardware that comprises the assembly. Additional information includes fuel irradiation history such as power level, burn-up, and cooling time. The mass of uranium and plutonium are also specified as well as activity and decay heat levels. Appendix A documentation for the USHPRR fuel (present fuel design) is available.

Shipment of spent nuclear fuel in the U.S. is subject to federal regulation by the U.S. Nuclear Regulatory Commission (NRC) [3]. The requirements address structural integrity, radiation shielding, thermal analysis, criticality analysis, and containment analysis of the shipment. Criticality control is a particular challenge for large fuel inventories in a cask, especially for highly-enriched fuel, and credited neutron absorbers, moderator exclusion, and/or burn-up credit are strategies (moderator exclusion and burn-up credit pending NRC approvals) to demonstrate criticality control under required considerations of normal and accident conditions.

Storage of research reactor fuel at a U.S. Department of Energy facility is not subject to NRC license requirements that are driven by federal regulation [4]. Nevertheless the general safety functions of criticality control, retrievability, confinement, and thermal control are maintained, and a limit in the alteration of the fuel during interim storage is also suggested to allow a full set of options for ultimate disposition [5]. A summary of the full set of technologies involved in management of spent nuclear fuel prior to final disposition in a repository is given in reference 6.

The last step or the readiness for ultimate disposition of the USHPRR fuel is less certain. The License Application for the federal repository at Yucca Mountain, which included commercial SNF, High-Level Waste, and the DOE SNF inventory including the USHPRR fuel, was submitted to the NRC in 2009 [7]. Subsequently, the Department withdrew, with prejudice, the application.

The present activities in the DOE-Nuclear Energy, Fuel Cycle Technologies program, Used Fuel Disposition – Disposal Research (DR) campaign include revisiting and analyzing repository types. The DOE-Nuclear Energy Fuel Cycle R&D program – Separations and Waste Form campaign is revisiting the concept of commercial fuel recycling and development of repository waste forms. The DR campaign does not explicitly include the DOE SNF, and, the DOE SNF may be handled and disposed in a separate waste disposal system. Regardless whether the DOE SNF would be co-disposed with commercial fuel, or would be disposed of separately in the repository, the associated waste disposal system will be subject to regulatory review.

Repository waste disposal system interface specifications, and waste acceptance criteria would be expected. These would establish requirements for the spent USHPRR fuel, and its container for compatibility with surface handling and disposal system infrastructure, safety, and the licensing basis. Considerations such as free liquids, heat loads, criticality safety, characterization of fines, are just a few of the attributes of the fuel that may be subject to limits or controls. Evaluation of the SNF-in-canister to the Features, Events, and Processes (FEPs) of the disposal system would be expected. Consistency of the SNF as a waste form as it relates to repository performance may also be a consideration. Footprint minimization (volume reduction) for storage and repository placement may be other drivers of the storage and disposal systems that would translate into life-cycle costs.

As an example, considerations for the Yucca Mountain repository disposal of aluminum-based fuel DOE SNF, in the direct disposal form, and in a treated, volume-reduced form, is documented in reference 8.

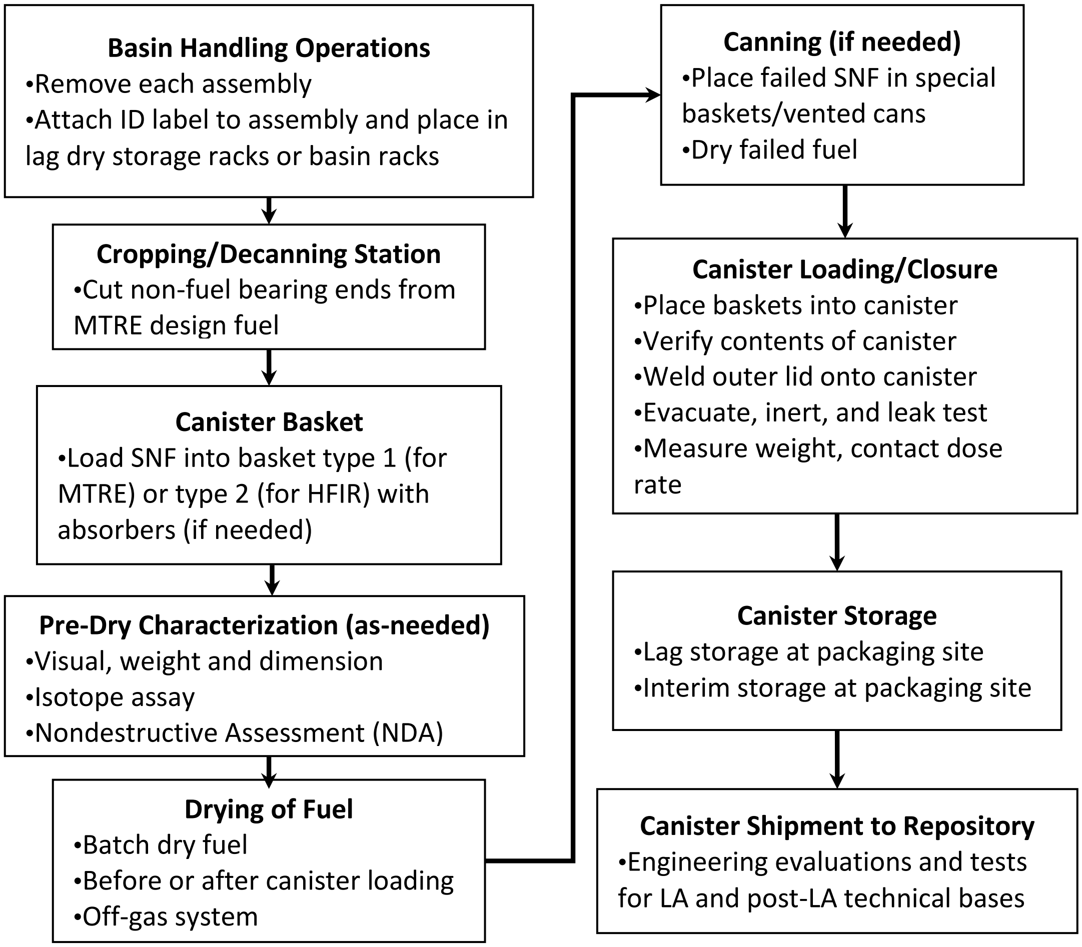
The above shows the important need to have the attributes of the USHPRR fuel post-discharge and storage, well-characterized to meet future repository requirements.

In summary, the technology for Direct Disposal is mature up through interim storage including wet and dry (non-sealed) storage. Sealed dry storage will require resolution of the safety issue of radiolytic gas generation from aluminum oxyhydroxides [9].

The disposal system interface specification and waste acceptance criteria for repository disposal, and the corresponding characteristics of the USHPRR fuel against those requirements, is not established. Nevertheless, general criteria can be gleaned by analogy to the preparation of the License Application (LA) for the Yucca Mountain Project (YMP) [7], and the expected post-application activities that would be needed for USHPRR fuel in a repository.

Process Description

A suggested processing path to transition fuel from a wet storage system, to a canister, “road-ready” to be transported to and handled at a repository for direct disposal placement, is shown in the following figure.



Basic Process Steps to Transition Wet-Stored-Fuel to Road-Ready Canisters that Would Meet Repository Requirements

Several steps in the transition of wet-to-dry and the readying of the fuel for repository disposal are highlighted.

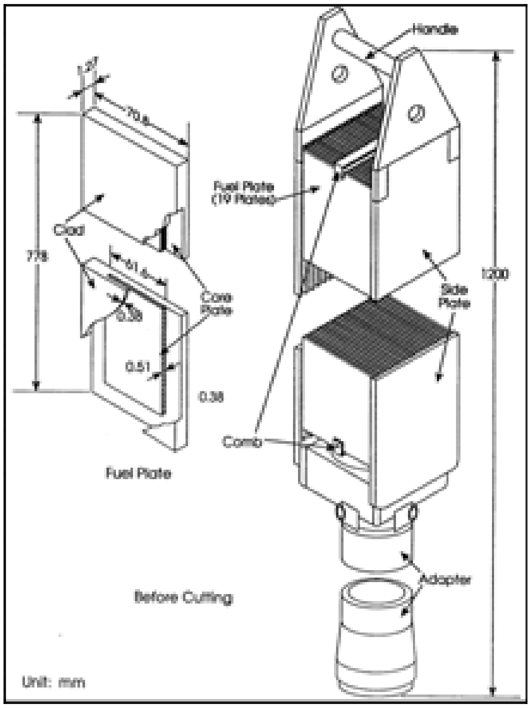
Two basic fuel designs are expected for the USHPRR fuel, namely the Materials Test Reactor Equivalent design for the ATR, MIT, MURR, and NIST fuel, and the HFIR fuel. The MTRE fuel would be cropped and placed in a basket (“Type 1”) for loading in a multi-purpose storage, transportation, and disposal canister. The HFIR fuel would be placed in a basket (“Type 2”). See figures below.

An important step involves the manifestation of the physical and isotopic condition of the fuel for disposal. It is expected that the information provided in Appendix A documentation, coupled with standard isotopic production/depletion methods and codes (e.g. ORIGEN-S) to evaluate other radioisotopes not provided in the Appendix A documentation, would be sufficient to provide the required information set. If Appendix A information is not sufficient or records would not be able to be assembled, the pre-dry characterization activities would be needed. This information is needed, for example, for storage, transportation, and repository surface facility analyses (e.g. release of fines); and for repository evaluation, for example, for heat load of the fuel in the repository.

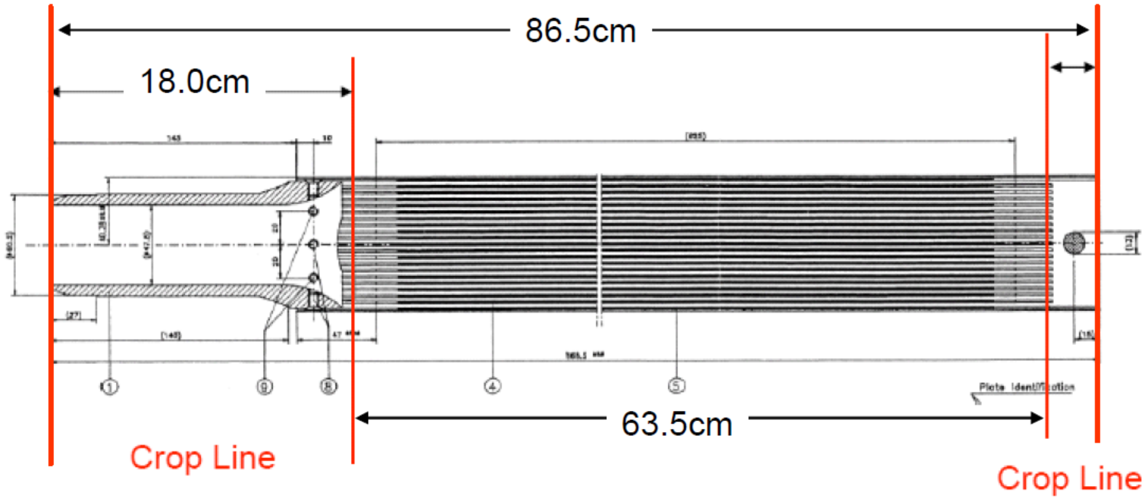
The actual drying protocol would need to be established. The drying could be performed pre-canister loading or following canister loading where the fuel in the basket is loaded underwater and then drained and dried. There are trade-offs in either approach.

The dry-stored fuel would be subject to interim dry storage requirements, if storage longer than lag storage at the packaging facility would be needed. The dry storage facility is expected to be an NRC-licensed facility, subject to the requirements under 10 CFR Part 72 [4].

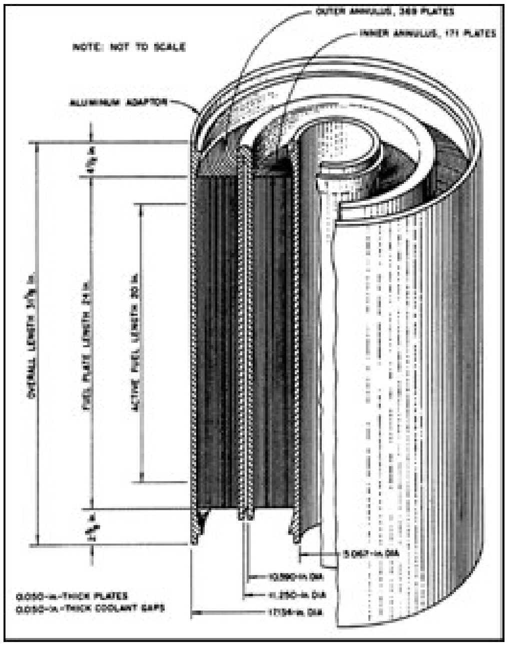
Tests and analyses for the repository disposal would be needed, and fuel performance data such as corrosion behavior under repository-relevant conditions is expected. The engineering evaluations and tests could be performed as part of the technical bases for a license application, or could be completed during the license review process.



Schematic of a Typical (Boxed-Type/Flat-Plate) Material Test Reactor Equivalent (MTRE) Aluminum-Based Fuel Element



Dimensions of MTRE showing the cropped length (63.5 cm) for L Basin bundled fuel storage



High Flux Isotope Reactor Core

Identified Technology Gaps

- A base of information related to the present design of USHPRR fuel exists. The post-discharge and the post-storage (physical condition and isotopic) characteristics of the fuel need to be developed for storage, transportation, and repository disposal safety- and regulatory-driven needs (High Priority)
- Drying protocols to remove free water are system-specific. Failed fuel may have additional drying challenges. An additional issue is radiolysis of aluminum oxyhydroxides in generation of hydrogen and potential oxygen (safety issues for sealed dry storage). (High Priority)
- Repository performance information such as characterization tests to determine corrosion/release rates in range of repository environments and attribute tests for interpretation of the characterization tests will be needed for the new design USHPRR fuel. That is, information on USHPRR fuel as a waste form needs to be developed.

Criteria for Evaluating Option Category	**Rating**	**Comments**
Industrial Practicability		
Technical Practicality	+	Commercial dry and non-sealed dry storage of DOE SNF are being performed at present.
Robustness	+	Process of DD takes the fuel “as-is” no matter what its condition is. May need secondary canning for handling/retrievability, as-needed, for highly damaged fuel.
Degree of Complexity	+	Straightforward packaging operation and weld-seal envisioned for a canister system for DD
Operating Costs	+	Minimal packaging operations costs and interim dry storage costs prior to repository disposal
Capital Costs	0	A packaging facility with remote handling and welding operations would be needed.
Technical Maturity		
Development Costs	+	Commercial dry and non-sealed dry storage of DOE SNF are being performed at present and systems to place USHPPR in a road-ready canister are anticipated to be directly adaptable.
Schedule to Demonstrate	+	Short-term schedule to demonstrate anticipated.
Process Maturity	+	Straightforward project. Little RD&D anticipated to address drying and safety topics (H2 generation from oxyhydroxides)
Waste		
HLW Disposition Costs	-	This disposal form has a “full footprint” that may adversely impact disposition cost in the repository
LLW Disposition Costs	+	Minimal secondary wastes from a packaging operation of the fuel are expected for DD
U Recovery		
Yield	-	Uranium is not recovered
Product Quality	N/A	
Cost/risk to Meet GTRI objectives		
Nonproliferation	+	Materials are not separated
Materials Control and Accountability	+	Fuel assemblies are readily “trackable” and “countable.” Computer codes mature to give accurate isotopics.
Physical Security	-	Uranium albeit LEU (<20%) would be retrievable in storage and in the pre-closure repository disposal period

Category	Rating	Comments
Cost/risk to Meet Environmental, Safety and Regulatory Requirements		
Environmental	+	The submission of the LA for YMP provides a template for information needed for a repository. Testing and evaluation of the USHPRR fuel is expected to be needed, but the cost/risk to meet requirements should be relatively low.
Safety	+	The present design USHPPR was part of the YMP construction LA submission and no significant safety issues were identified.
Regulatory	+	The present design USHPPR was part of the YMP construction LA submission and no significant regulatory issues were identified.

References

1. Recommended practices for water quality management in research reactors & spent fuel storage facilities," International Atomic Energy Agency, IAEA Nuclear Energy Series No. NP-T-5.2, July 2011.
2. Dunsmuir, M.D., Spieker, T.J., "Return of research reactor spent fuel to the country of origin: Requirements for technical and administrative preparations and national experiences," International Atomic Energy Agency, IAEA-TEC-DOC-1593, 2008.
3. U.S. Nuclear Regulatory Commission, 10 CFR Part 71, Packaging and transportation of radioactive material.
4. U.S. Nuclear Regulatory Commission, 10 CFR Part 72, Licensing requirements for the independent storage of spent nuclear fuel, high-level radioactive waste, and reactor-related greater than class C waste.
5. Sindelar, R.L., Vinson, D.W., Iyer, N.C., and Fisher, D.L., "Overview of Criteria for Interim Wet & Dry Storage of Research Reactor Spent Nuclear Fuel," SRNL-STI-2010-00688, Savannah River National Laboratory, Savannah River Nuclear Solutions, LLC, 2010.
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9. Fisher, D.L., Westbrook, M.L., and Sindelar, R.L., "Test Results from Gamma Irradiation of Aluminum Oxyhydroxides," SRNL-STI-2011-00602, Savannah River National Laboratory, Savannah River Nuclear Solutions, LLC, 2011.

6.3.2 Justification for Eliminating Direct Disposal – Foreign Disposition

Technology Overview

The back-end handling of USHPPR fuel following discharge and cool-down from U.S. reactors by shipment to a foreign country with the foreign nation taking title to the fuel and providing for its ultimate disposition defines this option path. With contract negotiations, it would be stipulated that the foreign nation would not reprocess the fuel but would package and dispose of it in its own repository or package it for disposal in a third-party country.

This path would have no technology development or costs, and thus would be expedient for disposition of the USHPRR fuel.

Rejection Criteria

The shipment of the fuel to a foreign country for ultimate disposition does not require any U.S. investment in technology development or in repository acceptance evaluation for a disposal form. There are essentially no considerations other than a (high expected) cost and contract negotiations with a foreign country for this arrangement.

It is fully expected that this is abundantly non-acceptable from the U.S. or the foreign country societies, and especially for potential violation of the non-proliferation policy of the U.S. government since the fuel is out of U.S. ownership and control with this option. For these reasons, foreign disposition is not considered an option for USHPRR fuel.

6.4 Outsource

In the outsource category, it is assumed that the irradiated fuel is directly transferred for a fee to a commercial entity for treatment and disposition. Further, it is assumed that development and operation of the dissolution and waste treatment processes are the responsibility of the commercial entity. The reprocessing options defined above cover the practicality, relative maturity, and technology development requirements needed for the outsource options; therefore, the options in this category are not rated. It is recommended that continued assessment of world-wide reprocessing efforts be made to evaluate the potential for these options as the USHPRR conversion program moves forward.

6.4.1 Outsource for commercial domestic fuel reprocessing

Technology Overview

Reprocessing commercial domestic fuel in the United States is again being considered. Research efforts are underway to evaluate limited-recycle and full-recycle options. The goal of a limited-recycle option would be to use uranium resources more efficiently while utilizing limited processing to minimize wastes. In a full-recycle option, nuclear fuel would be repeatedly reprocessed and recycled so that TRU elements were completely consumed and wastes minimized. The desired outcome of the research efforts is to identify and develop a preferred sustainable nuclear fuel cycle to facilitate the use of nuclear power as a major contributor toward enabling the nation to achieve energy security and greenhouse gas emission reduction.

The current program is designed as a long-term effort to ensure that a sound scientific basis underpins the selected recycling option. Therefore, the current program implementation plan includes distinct research and technology demonstration phases prior to commercial implementation. This results in a timeline that includes engineering-scale demonstration testing in 2030-2050 and technology transfer for commercial implementation after 2050.

It is envisaged that this developed, demonstrated and deployed future commercial fuel processing option would be able to reprocess hundreds to thousands of tons of spent fuel per year so that the relatively small inventory of U-Mo fuel (assumed to be approximately 2 tons/year) would be easily accommodated. However, issues identified with reprocessing U-Mo monolithic fuel with a zirconium barrier layer (as discussed in the other evaluated technology options) would need to be resolved prior to introduction into a commercial reprocessing facility. Furthermore, any impacts of processing the U-Mo monolithic fuel on commercial fuel reprocessing operations would need to be explicitly identified and vetted.

Process Description

In this option, the spent U-Mo monolithic fuel would be handled and processed in a facility designed and operated to process other fuels (likely commercial-power reactor oxide fuels). The U-Mo monolithic fuel could either be handled and processed separately in a campaign manner or mixed in with processing of other fuel types. Due to the unique make-up of the U-Mo fuel, it is expected that a separate approach would be utilized.

Identified Technology Gaps

- The U-Mo monolithic fuel incorporating a barrier layer is a unique design that is expected to be significantly different from fuel being processed in a domestic reprocessing facility. All technology gaps to process this fuel using the reprocessing technology employed in the commercial facility will need to be addressed.
- It is anticipated that the U-Mo monolithic fuel will represent a minor fraction of the fuel inventory to be reprocessed in the commercial facility. However, the reprocessing of this unique fuel could disrupt commercial fuel processing operations. Therefore, any impacts of reprocessing the U-Mo monolithic fuel will

need to be identified and vetted. Moreover, specialized reprocessing schemes or capabilities may need to be added to the reprocessing facility.

- The high-fluoride raffinate that would be expected from aqueous dissolution has no easy disposition path. If any dissolution process is selected, means to prepare a suitable waste form and disposal path must be developed.

References

1. “Separations and Waste Forms Campaign Implementation Plan,” U.S. Department of Energy Office of Nuclear Energy - Fuel Cycle Research and Development, FCRD-SWF-2012-00123, June 2012.

6.4.2 Outsource for fuel reprocessing at foreign facility

Technology Overview

Commercial reprocessing has been conducted over the past several decades in France, Japan, Russia, India and the United Kingdom. Advances in separations processes in these countries have resulted in significant improvements in process efficiency, materials control and accountability, and waste management. Both uranium and plutonium are being recycled into fresh reactor fuel, thereby making use of more of the original fuels' potential to generate energy. The highly radioactive wastes, including the minor actinides, are stabilized into waste forms suitable for deep geologic disposal.

Recently in France, next generation reprocessing technologies have been developed to avoid the separation of pure plutonium. These processes use variants of the PUREX process, changing conditions to maintain some uranium with the separated plutonium in what has been called the COEX, NUEX or UREX co-extraction process.

South Korea is a technology leader in developing electrochemical (or pyrochemical) separation processes to reprocess metal fast reactor fuel. In the electrochemical process, a molten salt (heated to about 500°C) is used to dissolve the fuel and separate a relatively pure uranium stream and a combined uranium/TRU stream.

Process Description

In this option, the spent U-Mo monolithic fuel would be packaged and shipped overseas. Upon receipt, the fuel would be handled and processed in a facility designed and operated to process other fuels (likely commercial-power reactor oxide fuels). The U-Mo monolithic fuel could either be handled and processed separately in a campaign manner or mixed with other fuel types. Due to the unique make-up of the U-Mo fuel, it is expected that a separate approach would be utilized.

It is expected that the selected foreign fuel processing facility would be able to reprocess hundreds to thousands of tons of spent fuel per year so that the relatively small inventory of U-Mo fuel (assumed to be approximately 2 tons/year) would be easily accommodated. However, issues identified with reprocessing U-Mo monolithic fuel with a zirconium barrier layer (as discussed in the other evaluated technology options) would need to be resolved prior to introduction into a commercial reprocessing facility. Furthermore, any impacts of processing the U-Mo monolithic fuel on commercial fuel reprocessing operations would need to be explicitly identified and vetted.

Identified Technology Gaps

- The U-Mo monolithic fuel incorporating a barrier layer is a unique design that is expected to be significantly different from fuel being processed in a commercial reprocessing facility. All technology gaps to process this fuel using the reprocessing technology employed in the commercial facility will need to be addressed.
- It is anticipated that the U-Mo monolithic fuel will represent a minor fraction of the fuel inventory to be reprocessed in the commercial facility. However, the reprocessing of this unique fuel could disrupt commercial fuel processing operations. Therefore, any impacts of reprocessing the U-Mo monolithic fuel will need to be identified and vetted. Moreover, specialized reprocessing schemes or capabilities may need to be added to the reprocessing facility.
- Although not a technology gap, transporting US-origin fuel for reprocessing in a foreign facility would require that programmatic, institutional, and regulatory issues are addressed.
- Waste processing and development of an acceptable waste form would be necessary. This could be complicated since the resulting form would likely be required to be shipped to the U.S. for final disposition.

Reference

1. “Separations and Waste Forms Campaign Implementation Plan,” U.S. Department of Energy Office of Nuclear Energy - Fuel Cycle Research and Development, FCRD-SWF-2012-00123, June 2012.

7.0 OPTIONS EVALUATION

The process to evaluate the options included weighting the criteria, determining a scoring scale, rating each option against each criterion, tallying the scores for the options, and reviewing the scoring to ensure accuracy and consistency. The team performed these analyses in a “face-to-face” working meeting, which facilitated discussion and resulted in the team reaching consensus on the option scoring and in the identification of the preferred options.

7.1 Criteria Weighting

To begin deliberations on criteria weighting, each team member individually weighted the 6 criteria with the total of the criteria summing to 100% and reported their results to the group. Following discussion among the team, consensus was obtained on the criteria weighting as follows:

- Industrial Practicability = 35%
- Technical Maturity = 25%
- Waste Byproducts = 15%
- Allows for Recovery of U = 15%
- Cost/Risk to Meet GTRI Objectives = 5%
- Cost/Risk to Meet Environmental, Safety and Regulatory Requirements = 5%

It should be noted that the weighting of each criterion does not necessarily directly relate to the *importance* of that criterion but rather in its ability to discriminate among the options. For example, it is assumed that the cost/risk to meet GTRI objectives is highly important. However, it is realized that all viable options will meet GTRI objectives (or they would not be considered) and the relative cost/risk to meeting the objectives is not a large discriminator.

7.2 Rating against the Criteria

As previously discussed, a relatively simple rating scale was developed by the team to evaluate each option against the criteria. The scale consisted of three scores denoted as:

- + = option rated positively against the criterion
- 0 = option rated neutral against the criterion
- − = option rated negatively against the criterion.

Each option was rated using this scale against the sub-criteria identified for each criterion (Section 5.0). No attempts were made to weight the sub-criteria under each criterion. That is, each sub-criterion was equally weighted with respect to the criteria it rolled up to.

7.3 Determination of Option Score

Each element in the rating scale was assigned a numerical or “point” value as follows:

- + = 2
- 0 = 1
- − = 0

Using this numerical scale, a sub-score was determined for each criteria by adding the "points" awarded to the sub-criteria divided by the maximum points possible multiplied by the weighting factor percentage. A final score was determined for each option (out of a maximum of 100) by adding up all the criteria sub-scores.

7.4 Option Score Results and Identification of Preferred Options

The scores for the rated options are provided in the table below.

	Option					
	With U Recovery			**Without U Recovery**		
Rating Criteria	**Pyrochemical**	**Aqueous/SX**	**Commercial Fuel Blend**	**Melt/Dilute**	**Dissolve/ Immobilize**	**Direct Disposal – Domestic**
Industrial Practicality (35)	21	21	28	24.5	24.5	31.5
Technical Maturity (25)	16.7	8.3	12.5	8.3	8.3	25
Waste (15)	7.5	7.5	7.5	7.5	7.5	7.5
Uranium Recovery (15)	11.3	15	11.3	0	0	0
GTRI Objectives (5)	2.5	2.5	2.5	3.3	3.3	3.3
Environmental Safety Regulatory (5)	2.5	2.5	2.5	2.5	2.5	5
TOTAL	61.4	56.8	64.3	46.2	46.2	72.3

In the evaluation of the option scoring, the team concluded that it was worthwhile to consider the separate groupings of the options with recovery of uranium and without recovery of uranium. In the grouping with uranium recovery, the team concluded that **Pyrochemical Processing, Aqueous Processing with Solvent Extraction,** and **Blending with a Commercial Fuel Process** all scored relatively equal and should be considered for further technology development. It is noted that the Blending with a Commercial Fuel Process is related to the other options resulting in uranium recovery from a technology development standpoint.

For the options in the grouping without uranium recovery, **Direct Repository Disposal to a Domestic Site** was identified by the team as the overwhelmingly preferred option and should be pursued further. The Melt/Dilute and Dissolve/Immobilize options do not appear to provide any significant advantages and, thus, it is recommended that no further technology development be pursued in support of these options.

8.0 CONCLUSIONS

A team of DOE complex experts completed a study to identify and analyze various back-end fuel cycle options resulting in the identification of preferred options for further technology development. A total of 18 unique options were identified. The options were evaluated against screening criteria. The screening process resulted in screening out 10 options. Two options (i.e. outsource for commercial reprocessing – domestic and foreign facilities) were later identified to be technologically similar to other options and were not rated but were carried forward in this report.

The team identified rating criteria for the options that were developed in detail. Six criteria categories were identified with sub-criteria determined for each criterion. The criteria were weighted, and a rating and scoring system was developed to rank the options. The options were rated against the criteria collectively by the team. Consensus on the ratings was obtained through discussion and deliberation.

The team concluded that identification of the preferred options could best be accomplished by grouping the rated options into options that resulted in recovery of uranium and options that did not result in recovery of uranium. In the grouping with uranium recovery, the team concluded that **Pyrochemical Processing, Aqueous Processing with Solvent Extraction** and **Blending with a Commercial Fuel Process** all scored relatively equal and should be considered for further technology development. It should be noted, however, that the value of recovery of the ~13% enriched U from the U-Mo fuel would be essentially lost in the option of blending with a commercial fuel process. For the options in the grouping without uranium recovery, **Direct Repository Disposal to a Domestic Site** was identified by the team as the preferred option that should be further pursued.

The team identified technology gaps for the preferred options and an initial list of technology development activities to address the highest priority technology needs. The next phase of this effort will be to further develop the technology gaps and development activities into a technology development roadmap. The roadmap will identify the research and development initiatives needed to address these technology gaps and provide a basis to develop a life-cycle R&D cost analysis.

LIST OF ACRONYMS AND ABBREVIATIONS

AFIP	ATR Full Size Plate In center flux trap Position
ALARA	As Low As is Reasonably Achievable
ANL	Argonne National Laboratory
ANS	American Nuclear Society
ANSI	American National Standards Institute
ATR	Advanced Test Reactor at Idaho National Laboratory
ATRC	ATR's associated critical assembly
BFD	Base Fuel Demonstration
BFQ	Base Fuel Qualification
CFR	U. S. Code of Federal Regulations
DDE	Design Demonstration Experiments
DF	Decontamination factor
DOE	U.S. Department of Energy
DU	Depleted uranium
EBR	Experimental Breeder Reactor
F	Function
F&R	Functions and Requirements
FA	Fuel Assembly
FE	Fuel Element
FFC	Fuel fabrication capability
FR	U.S. Federal Register
GTRI	Global Threat Reduction Initiative
HEU	Highly Enriched Uranium
HFIR	High Flux Isotope Reactor at Oak Ridge National Laboratory
HLW	High level waste
HPPR	High Performance Research Reactor
HVAC	Heating, Ventilation, and Air Conditioning
IAEA	International Atomic Energy Agency
IFR	Integral Fast Reactor
LA	License Application
LEU	Low Enriched Uranium
LINAC	ANL's Linear Accelerator
LLW	Low-level waste
MIT	Massachusetts Institute of Technology
MURR	Missouri University Research Reactor
NBSR	National Bureau of Standards Reactor
NIST	National Institute of Standards and Technology
NNSA	National Nuclear Security Administration
NRC	U.S. Nuclear Regulatory Commission
NUREG	NRC Regulation
ORRR	Oak Ridge Research Reactor
PUREX	Plutonium Extraction
QA	Quality Assurance
R	Requirement
R&D	Research and Development
RERTR	Reduced Enrichment for Research and Test Reactors

SAR	Safety Analysis Report
SNF	Spent Nuclear Fuel
SNM	Special Nuclear Material
TBD	To Be Determined
TRU	transuranic
U-Mo	Uranium-Molybdenum
UNF	Used Nuclear Fuel
USHPRR	U.S. High Performance Research Reactor
YMP	Yucca Mountain Project

APPENDIX A – BIOGRAPHICAL SKETCHES

The team of DOE complex experts who identified and analyzed the various back-end fuel cycle options described in this report included:

Jim Marra, Savannah River National Laboratory (Study Lead)
George Vandegrift, Argonne National Laboratory
Mark Williamson, Argonne National Laboratory
Steve Frank, Idaho National Laboratory
Steve Herrmann, Idaho National Laboratory
Tracy Rudisill, Savannah River National Laboratory
Bob Sindelar, Savannah River National Laboratory.

Pertinent biographical sketches follow.

Dr. James (Jim) Marra is an Advisory Engineer in the Materials Science and Technology Directorate of the Savannah River National Laboratory. He has over 24 years of work experience. Dr. Marra has extensive experience in development and performance testing of waste forms for nuclear and hazardous waste stabilization. He also has expertise in waste form production processes. Dr. Marra has conducted research on materials for use in extreme environments including materials for high temperature and radioactive operations. Dr. Marra received his Ph.D. in Ceramic and Materials Engineering from Clemson University. He obtained an M.S. in Materials Engineering from Worcester Polytechnic Institute. He also obtained a B.S. degree in Ceramic Science and a B.A. degree in Mathematics from Alfred University.

Dr. George Vandegrift is an Argonne Distinguished Fellow in the Chemical Sciences and Engineering Division of Argonne National Laboratory, where he has been for 35 years. Over that period, he has served as group leader, section head, department head, and associate division director. He is considered a world expert in the areas of (1) separation processes for radioisotope production, radioactive waste treatment, and industrial applications, (2) development of technology to convert Mo-99 production from high-enriched uranium to low-enriched uranium as part of the Global Threat Reduction—Conversion Program, and (3) development of processes for treating spent nuclear fuel in support of the Global Nuclear Energy Partnership. He has 200 journal articles, book chapters, reports, and patents in basic chemistry and applied topics associated with these areas.

Dr. Mark Williamson leads a multidisciplinary research and development organization with programs in nuclear fuel reprocessing, radioactive waste management, medical isotope production, process modeling and simulation, and safeguards in the Chemical Sciences and Engineering Division at Argonne National Laboratory. Dr. Williamson's technical expertise is in developing pyrochemical processing technologies for sustainable nuclear energy systems. He has extensive experience in actinide thermodynamics; pyroprocess design, development and demonstration; equipment engineering; and facility design. His work includes the transformation of unit operations from concept development to early-stage engineering with a focus on technology commercialization.

Dr. Steven (Steve) Frank is a lead scientist at the Idaho National Laboratory in Pyroprocessing Technology group. Dr. Frank has been at the INL for twenty years and has been involved in numerous research projects involving electrochemical and aqueous processing, waste form development, material separation, and materials characterization. Dr. Frank received his Ph.D. in Chemistry from Washington State University in 1989.

Mr. Steven (Steve) Herrmann (P.E. Nuclear Engineering, 2001; P.E. Chemical Engineering, 2000; MBA, Idaho State University, 1997; Nuclear Power School, Naval Nuclear Propulsion Program, 1990-91; B.S. Chemical Engineering, Brigham Young University, 1990) is a senior researcher at Idaho National Laboratory (Fuel Cycle

Science and Technology Division, Pyroprocessing Technology Department). His areas of expertise have involved research, development, and implementation of processes for treatment of sodium-bonded metal fuel, spent oxide fuel, and next generation research reactor fuel. In particular, he has led development efforts for the pyrochemical processing of oxide fuels at Idaho National Laboratory (INL) since 1995. These efforts have included development and bench-scale demonstration of integrated oxide reduction and electrorefining processes with used light water reactor fuel and fast reactor MOX fuel at INL and various associated process development activities. These efforts have involved collaborations with Argonne National Laboratory researchers and other international researchers, including a previous International Nuclear Energy Research Initiative project and a current Cooperative Research and Development Agreement to collaborate with researchers at the Korea Atomic Energy Research Institute on pyrochemical processing of oxide fuels.

Mr. Tracy Rudisill is a Senior Fellow Engineer in the Separations and Actinide Science Programs at the Savannah River National Laboratory (SRNL). In this position, Tracy has performed R&D activities associated with most uranium, neptunium, plutonium, americium, and curium processing activities at the Savannah River Site. His principle interests include separation processes associated with the nuclear fuel cycle and processes used to recover, purify, and isolate actinide materials. Tracy has worked as a principle investigator for the Fuel Cycle R&D Separations Campaign since the early stages of the program in 2001. In 2002, he worked with a team of SRNL scientist and engineers to perform the first successful demonstration of the UREX solvent extraction process. Tracy received his M.S. in chemical engineering and B.S. in chemistry from North Carolina State University.

Dr. Robert (Bob) Sindelar is a Senior Advisory Engineer in the Materials Science & Technology Directorate at the Savannah River National Laboratory. He has 27 years of experience at the laboratory in research, development, and deployment (RD&D) activities in materials for the nuclear fuel cycle and structural integrity of aging materials systems. Dr. Sindelar was a lead investigator in previous program activities to comprehensively evaluate repository disposability of aluminum-based research reactor fuels in a direct form, and following melt-dilute treatment. He presently supports the safe management of research reactor fuel in wet and dry storage systems at the Savannah River Site and worldwide through consultancies with the International Atomic Energy Agency, and the extended dry storage of commercial reactor fuel for the Nuclear Regulatory Commission. Dr. Sindelar received a Ph.D. in Nuclear Engineering in 1985 from the University of Wisconsin.