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Mu*STAR ADSR Fuel Conversion Facility Evaluation and Cost Analysis

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CONTENTS

LIST OF FIGURES ....................................................................................................................................... v
LIST OF TABLES ......................................................................................................................................... v
ACRONYMS ............................................................................................................................................... vii
ABSTRACT .................................................................................................................................................. ix
1. INTRODUCTION ................................................................................................................................. 1
  1.1 OVERVIEW OF THE MU*STAR ADSR ................................................................................ 1
    1.1.1 A Note about Subcritical Operation .............................................................................. 2
  1.2 PURPOSE AND OBJECTIVES ............................................................................................... 3
2. FUEL CONVERSION PROCESSES ................................................................................................... 3
  2.1 CHOP-VOLOXIDIZE-HYDROFLUORINATE PROCESS ...................................................... 5
  2.2 CHEMICAL DECLADDING-VOLOXIDIZE-HYDROFLUORINATE PROCESS ................. 6
  2.3 DISSOLUTION OF FUEL AND CLADDING IN MOLTEN SALT PROCESS ...................... 6
3. SIMULATIONS OF THE MU*STAR ADSR ...................................................................................... 8
4. IMPACTS OF LITHIUM ENRICHMENT ON ENERGY PRODUCTION AND TRITIUM MITIGATION REQUIREMENTS ...................................................................................................... 11
  4.1 EFFECT ON ENERGY PRODUCTION .................................................................................. 11
  4.2 EFFECT ON TRITIUM MITIGATION REQUIREMENTS .................................................... 13
5. FUEL CONVERSION FACILITY COST ANALYSIS ..................................................................... 15
  5.1 PARAMETRIC DATA—COST ESTIMATES FOR RECENT DOE HOT CELL DESIGNS........... 16
    5.1.1 Dry Transfer System Facility ......................................................................................... 16
    5.1.2 Sample Preparation Laboratory .................................................................................... 17
  5.2 PARAMETRIC DATA—ACTUAL COSTS OF PREVIOUS DOE HOT CELL FACILITIES ........ 19
    5.2.1 Fuels and Materials Examination Facility .................................................................... 19
    5.2.2 Recent Nuclear Facility Construction ....................................................................... 21
  5.3 COST ESTIMATE FOR THE ADSR FUEL CONVERSION FACILITY .................................. 21
    5.3.1 Design and Construction ............................................................................................... 22
    5.3.2 Operations and Maintenance Costs ............................................................................ 22
  5.4 FLOW SHEET FUEL CONVERSION EQUIPMENT COSTS ............................................... 22
    5.4.1 Waste Management ....................................................................................................... 22
    5.4.2 End-of-life D&D ........................................................................................................... 23
6. NUCLEAR FUEL CYCLE CONSIDERATIONS .............................................................................. 23
  6.1 COST/BENEFIT ANALYSIS ................................................................................................... 24
    6.1.1 Mu*STAR ADSR Fuel Cycle vs. Open Fuel Cycle with Dry Storage ........................... 24
    6.1.2 Mu*STAR ADSR Fuel Cycle vs. Closed Fuel Cycle with Aqueous Reprocessing .......... 25
    6.1.3 Mu*STAR ADSR Fuel Cycle vs. Closed Fuel Cycle with Pyrochemical Reprocessing ... 27
7. SUMMARY AND CONCLUSIONS ................................................................................................. 28
8. REFERENCES .................................................................................................................................... 29
APPENDIX A. MODELING THE IMPACTS OF LITHIUM ENRICHMENT ON TRITIUM PRODUCTION ................................................................................................................................. A-1
APPENDIX B. IMPACT OF TRITIUM IN USED FUEL CONVERSION FACILITY ....................... B-1
LIST OF FIGURES

Figure 1. Muons Inc. schematic of the Mu*STAR ADSR.................................................................1
Figure 2. Schematic of the proposed chop-voloxidize-hydrofluorinate process flow sheet.................5
Figure 3. Schematic of the proposed chemical decladding-voloxidize-hydrofluorinate process flow sheet......................................................................................................................6
Figure 4. Concept for converting UNF to fluoride salts by molten salt dissolution...............................7
Figure 5. Energy multiplier as a function of the mole fraction of Stream 15 in Stream 99....................9
Figure 6. Energy multiplier as a function of beam energy...................................................................10
Figure 7. Energy multiplier as a function of the $^6$Li fraction in the carrier salt.................................11
Figure 8. Temperature dependence on hydrogen retention in graphite before and after neutron irradiation at fluence of $3.9 \times 10^{23}$ n/m$^2$ (0.047 dpa), temperature of 1273 K and pressure of 10 kPa [Lam, 2017]. ...................................................................................................................14
Figure 9. Proposed schematic for off-gas processing of shear/voloxidation effluent............................15
Figure 10. DTS major components undergoing demonstration activities at INL....................................17
Figure 11. Visualization of the outside of the SPL.............................................................................18
Figure 12. Photograph of the FMEF facility compound......................................................................20
Figure 13. Cutaway image of the FMEF.............................................................................................20
Figure 14. Comparison of Mu*STAR ADSR fuel cycle with the current US fuel cycle......................24
Figure 15. Interim dry storage area for a commercial LWR nuclear-generating station......................25
Figure 16. Comparison of Mu*STAR ADSR fuel cycle with a once-through aqueous reprocessing fuel cycle....................................................................................................................26
Figure 17. La Hague nuclear recycling and reprocessing plant in northern France.............................26
Figure 18. Comparison of Mu*STAR ADSR fuel cycle with a pyrochemical reprocessing fuel cycle..........................................................................................................................27
Figure 19. Advanced Pyroprocess Recycling Facility from Integral Fast Reactor Program..................28

LIST OF TABLES

Table 1. Primary components by mass and activity in the fuel of one PWR fuel assembly (264 fuel rods). ........................................................................................................................................4
Table 2. Separation plant economics for 400 kg/year $^7$Li throughput.................................................13
Table 3. Cost estimate roll-up for the DTS facility.................................................................................18
Table 4. Cost estimate roll-up for the SPL............................................................................................19
Table 5. Nuclear facility design and construction cost comparison ..........................................................22
## ACRONYMS

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
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<tbody>
<tr>
<td>ADSR</td>
<td>Accelerator-driven Subcritical Reactor</td>
</tr>
<tr>
<td>D&amp;D</td>
<td>decommissioning and decontamination</td>
</tr>
<tr>
<td>DOE-NE</td>
<td>Department of Energy Office of Nuclear Energy</td>
</tr>
<tr>
<td>DTS</td>
<td>dry transfer system</td>
</tr>
<tr>
<td>FMEF</td>
<td>Fuels and Materials Examination Facility</td>
</tr>
<tr>
<td>INL</td>
<td>Idaho National Laboratory</td>
</tr>
<tr>
<td>LWR</td>
<td>light water reactors</td>
</tr>
<tr>
<td>ORNL</td>
<td>Oak Ridge National Laboratory</td>
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<tr>
<td>PWR</td>
<td>pressurized water reactor</td>
</tr>
<tr>
<td>SPL</td>
<td>Sample Preparation Laboratory</td>
</tr>
<tr>
<td>UNF</td>
<td>used nuclear fuel</td>
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ABSTRACT

The purpose of the reported effort is the development of a conceptual process for conversion of used nuclear fuel (UNF) from light water reactors (LWRs) to a fluoride salt form to facilitate its use for power generation in a Mu*STAR Accelerator-driven Subcritical Reactor (ADSR). This document evaluates potential processes for converting UNF into a fluoride salt, evaluates the costs/benefits of using enriched lithium in the carrier salt, and analyzes comparable costs of similar facilities to make a rough order of magnitude estimate for the cost to design, construct, and commission a Mu*STAR ADSR fuel conversion facility. This document also makes a high-level comparison of the Mu*STAR ADSR fuel cycle with other important fuel cycles.

The results of the cost analysis show that the Mu*STAR ADSR fuel conversion facility will cost more than $100M, possibly up to around $1B, to design and construct. The cost will depend on the size of the hot cell required, which will depend on the processing options chosen for converting the UNF to a molten salt. Operations and maintenance of the facility will be on the order of $10M per year. The design, fabrication, and installation of the remote process equipment to support the fuel conversion will be an additional cost of more than $50M. Cost surveys of Department of Energy hot cell designs and previous Department of Energy hot cell construction suggest that the cost variance may be large, and the risk of escalating costs may be high in the regulation-mandated atmosphere of nuclear facility design and construction.

The molten salt for the Mu*STAR ADSR contains all of the nonvolatile isotopes in the UNF, and there is never a separation of the plutonium from the fission products. This makes this conversion process much more proliferation resistant than the reprocessing options used for other closed fuel cycles.

When normalized to the amount of electricity produced, the Mu*STAR ADSR fuel cycle’s expected cost might be less than any reprocessing fuel cycle and possibly less than the once-through cycle now used. Simulations show that once the conversion to fluoride is made, that up to seven times as much energy can be extracted from the UNF than was produced by the LWR while making it [Bowman, 2010]. Using once-only, on-site conversion to fluoride means no mining, no enrichment, no manufacturing of fuel rods, and no transportation costs. Typically, the UNF was produced over a 40-year period in an LWR; it can potentially be used for 280 years in Mu*STAR, producing the same power as the LWR did, ultimately requiring disposal of approximately the same volume of material as the LWR generated. Mu*STAR burns most of the higher actinides, which could reduce both the cost of disposal and significantly reduce the longevity of its radiotoxicity.

Mu*STAR ADSR’s ability to demonstrate unique and valuable peripheral benefits are additional arguments for this technology, including (1) turning UNF into a valuable commodity (thus, its disposal will not be such an immediate problem, so continued operation of existing LWRs will be more acceptable); (2) burning fissile or fertile fuel including excess weapons-grade plutonium in subcritical mode; and (3) providing high-temperature process heat to convert natural gas to synthetic diesel fuel, water to hydrogen, or for other process-heat applications.
1. INTRODUCTION

The mission of the Department of Energy Office of Nuclear Energy (DOE-NE) is to advance nuclear power as a resource capable of meeting the nation’s energy, environmental and national security needs by resolving technical, cost, safety, proliferation resistance, and security barriers through research, development, and demonstration. The primary challenge facing expansion of nuclear power generation in the United States and one of the greatest barriers to public acceptance is the safe, cost-effective, and proliferation-resistant dispositioning of used nuclear reactor fuel.

The purpose of the work reported herein was the development of a conceptual chemical process for conversion of used nuclear fuel (UNF) from light water reactors (LWRs) to a fluoride salt form to facilitate its use for power generation in a Mu*STAR Accelerator-driven Subcritical Reactor (ADSR).

1.1 OVERVIEW OF THE MU*STAR ADSR

The proposed Mu*STAR ADSR will use a high-power superconducting radio-frequency proton accelerator to allow reactor operation without the need for a critical core, fuel enrichment, or reprocessing. Used commercial nuclear fuel, natural uranium, depleted uranium, thorium, or excess weapons-grade plutonium may be converted into a molten salt for use as the reactor fuel. The Mu*STAR reactor will use a superconducting proton accelerator, which has the capability to generate enough neutrons to generate 500 MW\textsubscript{thermal} in a subcritical molten salt fueled reactor core. Turning off the accelerator beam will immediately stop the fission reactions, thereby facilitating rapid, safe shutdown. Figure 1 shows a simplified schematic of the Mu*STAR ADSR [Muons, 2018].

![Mu*STAR ADSR Conceptual Elements](image)

**Figure 1. Muons Inc. schematic of the Mu*STAR ADSR.** Figure courtesy of Muons Inc.
While larger cores can be designed the 500MW\textsubscript{thermal} design is sufficiently small that passive air cooling can dissipate decay heat when the accelerator and reactor are off-line, making the reactor “walk-away safe.”

Mu*STAR is based on the GEM*STAR system described in Bowman et al. [2010]. Development of GEM*STAR by Accelerator Driven Neutron Applications Inc. is ongoing. The two technologies are similar in that they derive from Bowman’s 2010 Handbook of Nuclear Engineering article, which is based on a graphite-moderated, thermal spectrum, molten-salt, accelerator-driven, internal spallation target design. The analysis performed in this Gateway for Accelerated Innovation in Nuclear (GAIN) project is applicable to both reactor designs. Muons Inc. is now focused on taking advantage of several patented enhancements to very high-power proton accelerators based on superconducting radio-frequency cavities, which allow new approaches to burning spent nuclear fuel. It is anticipated that multiple cores, and possibly multiple accelerators, will be configured at an existing reactor site to generate the same amount of electrical power as the LWR that would ultimately be replaced at the end of its life cycle.

1.1.1 A Note about Subcritical Operation

The Mu*STAR ADSR operates in subcritical mode. That is, the neutrons produced from fission processes in the core are insufficient to maintain the chain reaction. The accelerator that drives the reactor generates an enormous flux of spallation neutrons to which the reactor responds: The power of the beam is “amplified” to yield the thermal power of the reactor:

\[
\text{(Thermal Power)} = (\text{Beam Power}) \eta \frac{1}{1 - k_{\text{eff}}}
\]

Here $\eta$ is a factor related to the efficiency of the spallation process, accounting for the number of neutrons produced and their spectrum after moderation; typical values for a 1,000 MeV proton beam are between 3 and 4. The effective neutron ratio from generation-to-generation in the fission chain reaction is $k_{\text{eff}}$, which depends on the geometry and fuel composition but is independent of the beam. On average, each neutron from the beam generates $k_{\text{eff}}$ fission neutrons, and each of those generates $k_{\text{eff}}$ more, etc., in a series that terminates in less than a second (because $k_{\text{eff}} < 1$). The reactor will be configured and operated so $0.7 < k_{\text{eff}} < 0.985$. Note that a critical reactor (e.g., an LWR) must maintain $k_{\text{eff}} = 1.0$ to within about a part per million, so the chain reaction is both stable and self-sustaining. Mu*STAR has much more flexibility here, permitting it to burn many more types of fuel, including low-reactivity fuels that most other technologies cannot use. Control of the reactor is achieved via the composition of the liquid fuel and the accelerator power; no delayed neutrons are required, giving additional flexibility in fuel selection, without redesign of the core. As the fuel is burned, fission products build up and absorb neutrons, causing $k_{\text{eff}}$ to decrease over time—to maintain a constant thermal power, the beam power must be increased. When $k_{\text{eff}}$ decreases to $\sim 0.7$ it becomes uneconomical to operate the accelerator with any more power, so the fuel load is finished and must be replaced; the replacement interval is estimated at about 280 years [Bowman, 2010].

This flexibility in $k_{\text{eff}}$, and therefore in fuels, has a striking consequence: It is estimated that there is enough uranium out of the ground today to supply 100% of the current U.S. electricity usage for more than 1,000 years using the Mu*STAR ADSR technology. Much of it is depleted uranium that cannot be used by most other technologies. Using depleted uranium, or thorium, in the Mu*STAR ADSR would result in no power production initially, and then slowly increasing production until the concentration of fissile elements reaches equilibrium, in about 2 years. Adding natural uranium to the depleted uranium would speed up the approach to equilibrium [Bowman, 2010].
1.2 PURPOSE AND OBJECTIVES

The purpose of this document is to provide a preconceptual design and cost estimate for the installation, operation, and maintenance of a fuel conversion facility to supply fluoride-salt fuel for a Mu*STAR ADSR. This document does not explore any costs related to the actual reactor or accelerator to be used for power generation. The cost estimate also explores potential costs related to waste management and end-of-life decommissioning of the facility because these costs are important to stakeholders involved in the siting and licensing processes for such a nuclear facility.

The Mu*STAR ADSR fuel conversion facility is at the preconceptual design stage, so the objective of this document is to provide an evaluation of potential processes for producing the fluoride salt, a description of the equipment that would be required, and a level of magnitude cost estimate based on historical data, as well as recent cost estimates for comparable facilities. This simplified cost analysis will make assumptions about what components of the fuel conversion facility drive the cost of the facility.

Additionally, in a separate section within this document, comparisons are explored between the fuel cycle involving the Mu*STAR ADSR and other established fuel cycles, such as the current open fuel cycle in the United States, and the two closed fuel cycles that involve either aqueous or pyrochemical recycling of spent nuclear fuel.

2. FUEL CONVERSION PROCESSES

For the purposes of this document, it is assumed that the molten salt fuel for the Mu*STAR ADSR will be converted from used commercial LWR fuel. A typical pressurized water reactor (PWR) assembly weighing 657.9 kg contains 26.1 kg (4.0 w%) of hardware (mostly stainless steel) and 108.4 kg (16.5 w%) of Zircaloy (cladding). A boiling water reactor (BWR) assembly weighing 275.7 kg contains 12.4 kg (4.5 w%) of hardware and 99.5 kg (36.1 w%) of Zircaloy. In each case, the balance of the mass is almost all UO$_2$, 523.4 kg for a PWR assembly and 163.8 kg for a BWR assembly. PWR assemblies account for about two-thirds of accumulated inventory in the United States, and BWRs account for the remaining one-third of the inventory.

To produce a molten salt fuel from the used commercial fuel, a hot cell facility for performing remote fuel handling and pyrochemical operations will be required. Depending on the chemical process chosen, the fuel conversion facility will include some, or all, of the following unit operations to convert the used LWR fuel to molten salt fuel:

- LWR fuel assembly dismantlement equipment
- Fuel chopping equipment
- A voloxidation system for decladding
- A chemical decladding reactor
- An oxide reduction unit
- A hydrofluorination unit
- Off-gas treatment including a stack
- A metal waste form fabrication unit.

Used fuel from a PWR was used to determine the composition of the fluoride fuel salt. We selected a Catawba I type fuel, with an initial enrichment of 4% and a burnup of 40 GWD/MTHM, that was cooled for 40 years. The primary isotopes by mass and activity are shown in Table 1. A relatively high burnup was chosen as a worst case, even though most older fuel has a lower burnup. Using spent fuel that has cooled for 40 years (or more) has many advantages. The radiation dose is much lower, $^{85}$Kr and $^3$H have
decayed substantially which will reduce off-gas treatment needs, and the decay of $^{241}$Pu means that more $^{239}$Pu, and less $^{243}$Am and $^{244}$Cm will be produced when used in a reactor.

Table 1. Primary components by mass and activity in the fuel of one PWR fuel assembly (264 fuel rods).

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Mass (g)</th>
<th>Isotope</th>
<th>Activity (Ci)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-238</td>
<td>3.96E+05</td>
<td>Cs-137</td>
<td>2.12E+04</td>
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<tr>
<td>U-235</td>
<td>4.36E+03</td>
<td>Sr-90</td>
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<td>Pu-239</td>
<td>2.54E+03</td>
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<td>9.30E+03</td>
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<td>U-236</td>
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<td>Am-241</td>
<td>1.81E+03</td>
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<tr>
<td>Xe-136</td>
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<td>Pu-238</td>
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<td>Pu-240</td>
<td>1.06E+03</td>
<td>Kr-85</td>
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<tr>
<td>Xe-134</td>
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<td>Cs-133</td>
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<td>Cs-133</td>
<td>5.76E+02</td>
<td>H-3</td>
<td>2.97E+01</td>
</tr>
</tbody>
</table>

During the conversion of the used fuel to fluoride salts, most of the volatile fission products (H, Kr, Xe) are removed. The fluoride salt product from the conversion process is labeled as Stream 15 in the discussions below.

The fuel salt will be mixed with a carrier salt, consisting of 65% LiF, 30% BeF$_2$, 5% ZrF$_4$ (mole %), which is based on the carrier salt used in the Molten Salt Reactor Experiment that was operated at Oak Ridge National Laboratory (ORNL) from 1964 to 1969 [Thoma, 1971].

As currently envisioned, the Mu*STAR ADSR reactor would contain 69.4 t (36.3 m$^3$) of molten salt, of which 17.6 t would be fluoride salts of the UNF. The overall composition (in mole%) of the carrier plus fuel salts would be 62.3% LiF, 28.8% BeF$_2$, 4.8% ZrF$_4$, and 4.1% UNF, the latter being mostly UF$_4$. This mixture is labeled as Stream 99 in the discussions below. The fuel conversion facility will need to be sized to produce the initial UNF salt inventory within a reasonable time span, possibly a couple of years. In order to be economical, the facility would then need to start preparing UNF salt for a new Mu*STAR ADSR reactor every few years for the life of the facility.

For each of the processes, it is assumed that used commercial nuclear fuel is transferred into the fuel conversion facility as full fuel assemblies. The hot cell facility will need to have cask handling features and the ability to unload a fuel assembly payload from a cask into the hot cell.

In addition to the process unit operations equipment within the hot cell, the fuel conversion facility will include offices for operations and support staff, a truck lock for cask receipt, a cask handling area, a hot cell dedicated to decontamination, hot cell manipulators, hot cell shielded windows, hot cell inert atmosphere controls, a control room, hot cell cranes, security systems, and nuclear facility safety basis documentation.

Three processes were evaluated for converting the UNF to fluoride salts and are described below.
2.1 CHOP-VOLOXIDIZE-HYDROFLUORINATE PROCESS

Figure 2 shows the proposed flow sheet for the first conversion process. All of the steps in this process are well developed and have been deployed in full-scale facilities. The shear is very large, which could increase the required hot cell space. The Zircaloy cladding hulls will need to be disposed of as high-level waste or extensively cleaned for low-level waste disposal.

Figure 2. Schematic of the proposed chop-voloxidize-hydrofluorinate process flow sheet.

Once within the facility hot cell, the fuel assemblies will be transferred via hot cell crane to the disassembly equipment to remove all of the structural hardware from the fuel assemblies so that the fuel pins may be easily accessed (also called “debundling”). The irradiated metal fuel hardware will then be transferred to a hot cell furnace to fabricate a metal waste form. Recycle of the metal is also an option, however most stainless steels are not valuable enough to warrant the cost. The metal waste form furnace is not shown in the Figure 2 flow sheet. The metal waste form will then be stored in a container in a storage area in the hot cell to await removal and disposal.

Following fuel assembly dismantlement, the fuel pins will be delivered to a shear for sectioning into small pieces. Voloxidation of the fuel segments with air converts the UO$_2$ to U$_3$O$_8$, or with NO$_x$ the UO$_2$ may be converted to UO$_3$. The conversion renders the higher oxides into powders that can be separated from the cladding pieces by sieving. The cladding can be compacted for disposal as high-level waste or potentially cleaned and mixed with the fuel assembly structural hardware for disposal as low-level waste. Off gas from both the chopping and voloxidation operations will be treated and exhausted through the hot cell ventilation system and stack.

The fuel powder from the voloxidation unit is then transferred to the oxide reduction unit where the U$_3$O$_8$ or UO$_3$ powder is converted to UO$_2$ powder. The product from the reduction vessel is then transferred to the hydrofluorination unit where the uranium is converted into uranium tetrafluoride, and the other fission products and transuranic metals in the UNF are converted to fluorides. At this stage, the salt is solidified in a form that can be sent to the reactor core and again heated to a molten state.
2.2 CHEMICAL DECLADDING-VOLOXIDIZE-HYDROFLUORINATE PROCESS

Treatment with either anhydrous chlorine (Cl₂) or anhydrous hydrogen chloride (HCl) at 350°C–400°C converts the Zircaloy cladding to volatile ZrCl₄, leaving the fuel essentially untouched and ready for the voloxidation and hydrofluorination steps. This process constitutes the primary difference between the flow sheet shown in Figure 3 with the one shown in Figure 2. As a result of the chemical decladding process, tritium trapped in the cladding will be released, and an off-gas treatment system will be required to remove the tritium from a chloride-bearing gas stream. As before, debundling allows for the recycling or low-level waste disposal of the stainless-steel hardware, which is 4–5 wt% of each fuel assembly. Chemical decladding separates the Zircaloy from the fuel meat upfront. At present, ORNL is studying chemical decladding by high temperature chlorination (350°C–400°C) using HCl or Cl₂ and vaporizing the ZrCl₄.

A parallel effort by a University of Tennessee–ORNL team is looking at a promising low-temperature alternative decladding chemistry using thionyl chloride or sulfur chloride to remove the Zircaloy, also as ZrCl₄. The goal is to make the ZrCl₄ clean enough for disposal as low-level waste or for reuse. ZrCl₄ could also be used as one of the salt components of the Mu*STAR ADSR molten salt fuel, replacing some of the LiF.

Figure 3 shows the three chemical steps being performed in three reactors. However, the three steps can potentially be done sequentially in just one reactor vessel by changing the gaseous reagents and still use a common off-gas treatment system.

2.3 DISSOLUTION OF FUEL AND CLADDING IN MOLTEN SALT PROCESS

Another approach to converting UNF to fluoride salts involves hydrofluorination in a molten salt [Cathers, 1957]. The processes described in the paper involve dissolution of the zirconium-uranium–based fuel elements in a molten fluoride salt at 600°C–700°C as the oxides are converted to fluorides.
using anhydrous HF. The reference also describes fluorination of the melt with \( F_2 \) to remove uranium as volatile \( UF_6 \); this operation is not applicable to the current application since uranium removal is not desirable. Two molten salts were used in the reported experiments, NaF–ZrF\(_4\) and LiF–NaF. At 600°C the NaF–ZrF\(_4\) remained liquidus for compositions ranging from 40–60 to 60–40 mol%, and the molten salt could accommodate “considerable” amounts of uranium solute. The 57–43 mol% LiF–NaF has a liquidus temperature of 675°C and could accommodate zirconium concentrations up to 55 mol%. It is very likely that other salts could be used, but experimental work is required to verify their efficacy.

Converting UNF to fluoride compounds in the molten salt is attractive because the \( UO_2 \) reacts readily with HF to produce water vapor and \( UF_4 \):

\[
UO_2 + 4HF \rightarrow UF_4 + 2H_2O
\]

Fission product oxides are also converted to fluorides, and HF does not volatilize uranium or other metal oxides in the fuel. The cladding can also be converted to fluoride, for example,

\[
Zr + 4HF \rightarrow ZrF_4 + 2H_2.
\]

A simple process can be envisioned, as shown in Figure 4, in which only the stainless-steel hardware is removed and the fuel pins are converted to fluoride salts. Several variations of the process are possible. Every variation may include a fuel pin cutting step to make more easily handled lengths.

![Figure 4. Concept for converting UNF to fluoride salts by molten salt dissolution.](image)

Three variations of molten salt dissolution are elaborated as examples.

1. Co-conversion of fuel and cladding—This is essentially the process shown in Figure 4. The first step is disassembly or debundling to remove the steel hardware. The Zircaloy-clad fuel is then converted to fluorides in a molten salt bath. Segmenting the fuel pins before salt dissolution can reduce the
required size of the molten salt bath. Fluoride salts to make the proper ratio of carrier salt and used LWR fuel may be added to act as the hydrofluorination flux. Depending on the characteristics of a UF$_4$–ZrF$_4$ melt, it may be possible to avoid adding a fluoride salt carrier and bootstrap a melt consisting of only the clad fuel. The presence of the Zircaloy will result in considerable quantities of hydrogen in the off-gas stream, which will be contaminated with released tritium. Flammable hydrogen poses a risk that will require stringent engineering controls. The fluoride salt product (Stream 16) will contain much more ZrF$_4$ than for the fuel and carrier salt (Stream 99) from the other conversion processes shown above.

2. Conversion of fuel after chemical decladding—A chemical decladding step (see Section 2.2) can be used to remove the zirconium, the primary constituent of the cladding. The predominantly UO$_2$ fuel pellets can be converted to UF$_4$ with HF, without the concern of hydrogen production. Fission product oxides will also be converted to fluorides. The addition of carrier salt is required to provide a fluxing agent. Tritium and other volatile fission products released to the off-gas will have to be sequestered from a fluoride-bearing gas stream. For this variation, Stream 16 will have the same composition as Stream 15 in the other two conversion processes, and combining the fuel salt with the carrier salt will produce the same Stream 99 composition.

3. Conversion of fuel after voloxidation decladding—Chopping the fuel pins into short lengths is required to improve the efficiency of voloxidation (see Section 2.1). Voloxidation of the fuel with air converts the UO$_2$ to U$_3$O$_8$, and with NO$_x$, the UO$_2$ may be converted to UO$_3$. The conversion renders the higher oxides into powders that can be separated from the cladding pieces by sieving. Neither U$_3$O$_8$ nor UO$_3$ are converted to UF$_4$ with HF; usually an oxyfluoride is produced. However, hydrofluorination using both HF and H$_2$ converts the higher oxides of uranium to UF$_4$ [Shaffer, 1971]. Considerable water vapor will be released during the hydrofluorination and dissolution of the fuel powder in a molten carrier salt, but it will not be contaminated with tritium because the tritium and other volatile fission products will have already been removed by the voloxidation process. Because the voloxidized fuel is in powder form, the dissolution step will be rapid. For this variation, Stream 16 will have the same composition as variation 2 above.

In all cases, the carrier or fluxing salt should be chosen such that a fuel salt mixture is produced, or a master mixture is produced that can be later blended with additional carrier to provide the desired fuel.

There are alternative chemical processes that could be used to dissolve both the cladding and the fuel pellets, but they were not evaluated for this report.

3. SIMULATIONS OF THE MU*STAR ADSR

Computational simulations of the Mu*STAR ADSR were performed to evaluate the operation of the molten-salt fuel converted from an LWR’s UNF. Monte Carlo N-Particle (MCNP) 6.1 was used, executing on the Romulus and Remus clusters at ORNL; a typical job uses 256 cores and occupies them for 1–4 hours of clock time.

The basic approach used was to take Stream 15 from the conversion processes described above and add it to the carrier salt to form Stream 99, which is the fuel salt for the reactor. The fraction of Stream 15 in the fuel salt is varied, as are the kinetic energy of the beam and the fraction of $^6$Li in the carrier salt.

The carrier salt is the same as that used during most of the Molten Salt Reactor Experiment at ORNL: 65% LiF, 30% BeF$_2$, and 5% ZrF$_4$ (mole %). The $^6$Li/$^7$Li ratio is varied; all other elements use their natural isotopic ratios.
As Mu*STAR operates subcritically and is driven by a proton beam, the primary figure of merit is the energy multiplier:

\[ \text{Energy multiplier} = \frac{\text{(Total thermal energy per proton)}}{\text{(Kinetic energy of each proton)}} \]

The total thermal energy was tallied for 1 second after the beam; the simulations use a beam pulse of a few picoseconds. The energy multiplier does not include energy from decays that happen more than 1 second after the beam pulse. The tally does include all of the fissions and contains >90% of the total thermal energy when totaled forever, and >95% of the total for 1 minute. For a given beam energy, the energy multiplier is determined by the geometry of the reactor and the composition of the fuel salt; it is also a function of beam energy. All of these simulations use the current Mu*STAR ADSR geometry, varying other parameters.

From the discussion in Section 1.1.1 above, we also have:

\[ \text{Energy multiplier} = \eta \frac{1}{1 - k_{\text{eff}}} \]

where \( \eta \) is typically between 3 and 4, and \( 0.7 < k_{\text{eff}} < 0.985 \). The energy multiplier rises rapidly as the composition of the fuel salt is varied to approach criticality \( (k_{\text{eff}} = 1) \).

Figure 5 shows the energy multiplier as a function of the mole fraction of Stream 15 in Stream 99. It is expected that the Mu*STAR ADSR will operate with an energy multiplier around 200 \( (k_{\text{eff}} \sim 0.98) \).

Figure 5. Energy multiplier as a function of the mole fraction of Stream 15 in Stream 99.
Figure 6 shows the dependence of the energy multiplier on the kinetic energy of the beam. Note that a 650 MeV beam has a reduction of only 8% compared to a 1,000 MeV beam—this is significantly less reduction than predicted from simply counting the difference in spallation neutrons. Evidently, the differences in their spectra are important. Note that for this scale the statistical fluctuations are much more visible.

Figure 6. Energy multiplier as a function of beam energy.

The previous figures use pure $^7\text{Li}$ in the carrier salt. This is unrealistic as the production of isotopically pure $^7\text{Li}$ is impossible: Figure 7 shows the effect of adding small fractions of $^6\text{Li}$ (keeping the rest of the salt composition fixed and a 1,000 MeV beam). Clearly very pure $^7\text{Li}$ is required.
4. IMPACTS OF LITHIUM ENRICHMENT ON ENERGY PRODUCTION AND TRITIUM MITIGATION REQUIREMENTS

There are two main impacts of lower lithium enrichment\(^1\) on the operation of the Mu*STAR ADSR. Reducing \(^7\)Li enrichment (increasing \(^6\)Li) decreases the energy multiplier and increases the amount of tritium formed from the initial \(^6\)Li charge.

4.1 EFFECT ON ENERGY PRODUCTION

The amount of energy that can be produced by the Mu*STAR ADSR is directly related to the energy multiplier. Section 3 provides a detailed analysis of Mu*STAR simulations on the effect of the UNF fraction, beam energy, and lithium enrichment on the energy multiplier. Focusing on the isotopic lithium composition in Figure 7, for \(^6\)Li fractions greater than 0.0001 (0.9999 \(^7\)Li enrichment, 99.99\%) the energy multiplier is less than 35–40. This is much lower than the energy multiplier of 200 described in Section 3. However, Figure 5 shows that increasing the fraction of the UNF salt in the total molten salt above 0.04 greatly increases the energy multiplier. Additional simulations may be able to optimize the energy multiplier as a function of both \(^7\)Li enrichment and UNF salt fraction.

There are no US commercial or government producers of enriched \(^7\)Li. The main global source of enriched lithium, NCCP in Novosibirsk, Russia, advertises on its website a standard enrichment of

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\(^1\) In this context the term “lithium enrichment” refers to increase in the isotopic concentration of \(^7\)Li with the balance being \(^6\)Li. The natural abundance of \(^7\)Li is about 92.5\% with 7.5\% \(^6\)Li. In other contexts, such as nuclear weapons and fusion energy research, the term can refer to increasing the concentration of \(^6\)Li.
99.95% $^7\text{Li}$ for BWRs and a higher enrichment of 99.99% for molten salt reactors and PWRs [NCCP, 2018], but no prices are listed.\(^2\)

More importantly, it is uncertain that there will be enough enriched $^7\text{Li}$ to supply the first Mu*STAR ADSR. Besides the Russian producer, the only other producer of commercially available enriched $^7\text{Li}$ is in China. In 2013 the Government Accountability Office investigated the issue of $^7\text{Li}$ supply and issued a report on the subject that states:

Little is known about lithium-7 production in China and Russia and whether their supplies can meet future domestic demand. According to industry representatives, China and Russia produce enough lithium-7 to meet demand from U.S. pressurized water reactors, a type of commercial nuclear power reactor that requires lithium-7 for safe operation. However, China’s continued supply may be reduced by its own growing demand, according to an expert that is familiar with China’s plans. Specifically, China is building several pressurized water reactors and developing a new type of reactor\(^3\) that will require 1,000s of kilograms of lithium-7 to operate, rather than the 300 kilograms needed annually for all 65 U.S. pressurized water reactors. Relying on two producers of lithium-7 leaves U.S. pressurized water reactors vulnerable to lithium-7 supply disruptions. [Government Accountability Office, 2013]

Trade sanctions and tariffs will increase the risk of disruptions in supply from foreign sources.

In a 2010 book chapter on GEM*STAR, a precursor to the current Mu*STAR concept, it is stated that ADNA Corporation has developed economically practical alternative means to reach the desired purity levels of $^7\text{Li}$ (0.9999) at the production rates required (stated as 1 ton/year per 200 MWe GEM*STAR) [Bowman, 2010]. No additional information on development of that technology or progress toward its commercialization has been found.

A 2012 report from the University of California at Berkeley describes possible options for domestic $^7\text{Li}$ enrichment and provides an estimate for construction of a plant to enrich $^7\text{Li}$ to 99.95% at a rate of 400 kg/year using crown ether aqueous organic contacting in mixer-settlers [Ault, 2012]. The capital cost in 2012 was estimated to be $11,500,000 as shown in Table 2. (Inclusion of the estimate is not an endorsement nor indication that any of the information is accurate.) This cost estimate has not been critiqued and is presented here simply as an example of some of the only available work that has been reported on lithium enrichment.

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\(^2\) The website claims NCCP supplies 70% of $^7\text{Li}$ world consumption.

\(^3\) This refers to China’s plans for thorium molten salt reactors.
Table 2. Separation plant economics for 400 kg/year $^7$Li throughput

<table>
<thead>
<tr>
<th>Capital Costs</th>
<th>Qty</th>
<th>Cost (each) $</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reverse osmosis water systems (GPM)</td>
<td>5</td>
<td>$100,000</td>
<td>$500,000</td>
</tr>
<tr>
<td>Dryers (each)</td>
<td>2</td>
<td>$100,000</td>
<td>$200,000</td>
</tr>
<tr>
<td>Facility (ft)</td>
<td>10,000</td>
<td>$120</td>
<td>$1,200,000</td>
</tr>
<tr>
<td>Electrical equipment (MCC)</td>
<td>1</td>
<td>$500,000</td>
<td>$500,000</td>
</tr>
<tr>
<td>Electrical equipment (Substation)</td>
<td>1</td>
<td>$180,000</td>
<td>$180,000</td>
</tr>
<tr>
<td>Other equipment</td>
<td>1</td>
<td>$500,000</td>
<td>$500,000</td>
</tr>
<tr>
<td>Refrigeration system</td>
<td>300</td>
<td>$300,000</td>
<td>$300,000</td>
</tr>
<tr>
<td>Mixer-settler cells</td>
<td>260</td>
<td>Size dependent</td>
<td>$1,400,000</td>
</tr>
<tr>
<td>Analytical lab</td>
<td>1</td>
<td>$200,000</td>
<td>$200,000</td>
</tr>
<tr>
<td>Organic recovery system</td>
<td>2</td>
<td>$100,000</td>
<td>$200,000</td>
</tr>
<tr>
<td>Benzo-15-crown-5 (kg)</td>
<td>370</td>
<td>$1,000</td>
<td>$370,000</td>
</tr>
<tr>
<td>Chloroform (L)</td>
<td>6,200</td>
<td>$2</td>
<td>$12,400</td>
</tr>
<tr>
<td>Engineering</td>
<td>1</td>
<td>$3,000,000</td>
<td>$3,000,000</td>
</tr>
<tr>
<td>Installation</td>
<td>1</td>
<td>75% equipment</td>
<td>$2,800,000</td>
</tr>
<tr>
<td><strong>Total capital cost</strong></td>
<td></td>
<td></td>
<td><strong>$11,500,000</strong></td>
</tr>
</tbody>
</table>

The same Berkeley report includes the following statement: “In 2010, the estimated price for Chinese enriched Li-7 was $15,000/kg for 99.99% enrichment level. Since prices for 99.92% enriched lithium were not found, the price was estimated to be roughly in the $10,000/kg range (taken from the higher enrichment prices).” The referenced source for the estimate was an email from P. Peterson, professor of nuclear engineering at University of California, Berkeley. A search for a more recent or more definite price found several mentions of $10,000/kg referencing the 2012 Berkeley report or with no reference at all.

4.2 EFFECT ON TRITIUM MITIGATION REQUIREMENTS

As shown in Appendix A Figure A-2, tritium will be produced in the molten salt even with 100% $^7$Li. Tritium production from sources other than the initial $^6$Li begins at a rate of about 150 Ci/day and increases to about 300 Ci/day after 10 years. The same figure shows that with 99.99% enriched $^7$Li, the tritium production rate will initially be about 16,000 Ci/day but will decrease as the $^6$Li is consumed over 15 to 20 years. The concern is that at high temperatures (≥700°C) hydrogen isotopes, including tritium, diffuse readily through typical materials used for reactor vessels and heat transfer systems that perform well at 300°C. These materials include stainless steels and corrosion-resistant nickel alloys. As an example, the hydrogen (tritium) permeability of austenitic stainless increases by a factor of over 100 between 300°C and 700°C [Causey, 2012]. The permeation will increase with increasing $T_2$ concentration in the molten salt.

The tritium produced will not all be immediately available for permeation and release from Mu*STAR. Tritium will adsorb on the graphite moderator and diffuse into its pores. Calculations of maximum tritium inventories in the graphite moderator vary greatly with time and the type of graphite, with a large spread even among different nuclear grades of graphite. Radiation damage to graphite creates trapping sites for tritium and greatly increases the tritium retention in the graphite as seen in Figure 8.
Figure 8. Temperature dependence on hydrogen retention in graphite before and after neutron irradiation at fluence of $3.9 \times 10^{23}$ n/m$^2$ (0.047 dpa), temperature of 1273 K and pressure of 10 kPa [Lam, 2017].

The formation of HF/TF occurs with the production of tritium. Tritium is initially produced as a T$^+$ ion, and the conversion of the Li leaves uncombined Li$^-$ ions. To prevent the formation of TF and the accompanying corrosion issues, the reduction-oxidation (redox) potential of the salt must be controlled. One way to do this is to provide an excess of Be in the salt. The results of an experiment that bubbled HF gas through molten salt with varying concentrations of dissolved Be indicated that the HF/TF could be maintained as low as 0.020 ppb in the salt [Simpson, 2006].

To prevent tritium buildup in Mu*STAR, the reactor will have a helium cover gas and purge. To enhance removal of the tritium and fission gases, the helium should be bubbled through a portion of the molten salt and captured by a stripping system outside of the reactor. The most common method of removing tritium from a nonhydrogen gas stream is oxidation on a catalyst and absorption on zeolite [Hsu, 1997]. Figure 9 shows the tritium removal system in context with removal of metal vapors and fission noble gases. Removal of condensable solids may include return of fluoride salts to the reactor. This schematic shows treatment of off-gas from both the conversion facility and the Mu*STAR ADSR in one treatment train; however, separate trains may be used for each facility.
One of the most effective ways to improve removal of tritium from the reactor is dilution with H\(_2\) before off-gas treatment. This can be easily done by adding H\(_2\) to the He purge gas. At steady state, the tritium inventory in the reactor is reduced by the ratio of the rate of hydrogen addition to the rate of tritium production. From Appendix A Figure A-2 the tritium production will be about 350 Ci/day (135 STP cc/day) for the majority of the life of the reactor. Adding 13.5 STP L/day of hydrogen to the purge gas and then stripping it out with the tritium will reduce the total reactor inventory of tritium to 1% of what it would be without hydrogen addition.

5. FUEL CONVERSION FACILITY COST ANALYSIS

Because of the radioactivity of spent nuclear fuel, all fuel conversion operations will be performed remotely, requiring a large hot cell facility. The costs of building and operating a hot cell facility include design and construction, operations and maintenance, waste management, and end-of-life decontamination and decommissioning. Because the Mu*STAR ADSR is currently in the preconceptual design stage, enough data to complete a detailed cost estimate for the fuel conversion facility does not exist. This section discusses the historical costs of similar facilities as well as cost estimate data for recently designed DOE hot cell facilities.

Because of the proprietary nature of hot cell and reprocessing facility cost estimates performed by private companies, details regarding commercial facilities have been excluded from this document. The data is implicit in some fuel cycle costs where the data was used as input to fuel cycle costs discussed in Section 3 of this document. Specific values that exist in some DOE Next Generation Nuclear Plant project studies are also not included because they are considered business sensitive and are thus designated as Official Use Only information Verbal conversations with individuals involved in development of those DOE Next Generation Nuclear Plant studies confirm that the order of magnitude for facility construction costs provided in this report are in the range of those estimates [private communication from Joe Carter, Savannah River National Laboratory (SRNL) to Steve Warmann, INL, August 2018].

Key assumptions about the cost estimate for the fuel conversion facility are listed below.

- Spent nuclear fuel is cooled at the LWR reactor site in the fuel storage pool and then transferred to dry storage casks. LWR fuel assemblies are dry-transferred via a cask cart to a hot cell with shield walls, shield windows, and remote manipulators. The fuel conversion facility unit operations equipment includes fuel assembly dismantlement equipment, a voloxidation unit, an oxide reduction...
unit, a hydrofluorination unit, off-gas treatment and a stack, and a metal waste form fabrication unit. This configuration is consistent with the first conversion operation alternative considered, and is depicted in Fig. 2.

- Any required maintenance of the molten fluoride salt fuel is assumed to occur in the Mu*STAR ASTR facility. If an additional process line for pyrochemical fuel salt maintenance is required in the conversion facility, it would significantly increase the facility cost. The fuel salt will be burned for up to 280 years and then dispositioned, presumably as a ceramic waste form in a separate waste form fabrication unit, possibly within the fuel conversion facility, and replaced with fresh molten salt fuel. Costs for ceramic waste form fabrication are not considered in this fuel conversion facility estimate.

- The fuel conversion facility is adjacent to an interim LWR dry storage facility or a commercial nuclear power–generating station so that major transportation costs do not exist, such as the capital needed for receiving cask-laden rail cars.

- Security staffing and major security systems are already existing as part of a nuclear site within which this fuel conversion facility will be sited.

5.1 PARAMETRIC DATA—COST ESTIMATES FOR RECENT DOE HOT CELL DESIGNS

Two cost estimates were used to establish an order of magnitude cost estimate for a fuel conversion facility for the Mu*STAR ADSR. Firstly, a cost estimate was performed by Idaho National Laboratory (INL) on a dry transfer system (DTS) facility that was intended to perform dry transfer operations on commercial spent nuclear fuel assemblies [Wasley, 2016]. Such dry transfer mechanisms will be required in the fuel conversion facility. The DTS facility design is comparable to a similar facility already constructed at the INL site. Additionally, portions of the design underwent proof-of-concept prototyping and testing.

Secondly, a small hot cell facility called the Sample Preparation Laboratory (SPL) has been designed for construction at INL. A detailed cost estimate that includes all aspects of a modern hot cell facility is applied here directly to develop the order of magnitude estimate for the fuel conversion hot cell facility. The SPL cost estimate includes operations and maintenance costs, which are all developed based on actual operational costs of DOE hot cell facilities at the INL [Wasley, 2016a]. Additionally, the SPL estimate includes decommissioning and decontaminating (D&D) costs for the end of life of the facility [Wasley, 2016b].

The Mu*STAR ADSR fuel conversion facility will include most aspects of both the DTS facility and the SPL.

5.1.1 Dry Transfer System Facility

In 2016 a cost estimate was performed for the design, construction, and commissioning of a DTS facility at the Advanced Test Reactor at INL [Wasley, 2016a]. The purpose of the DTS was to demonstrate a DOE commitment to pursue a demonstration of a spent fuel transfer system that could be deployed at decommissioned reactor pools after shutdown. The system design allowed for the use of large storage-only casks for temporary on-site storage of fuel assemblies as well as transfer of fuel assemblies from storage casks to shipping casks. Figure 10 shows a major component of the DTS being tested at INL.
Figure 10. DTS major components undergoing demonstration activities at INL.

The facility was scoped to include an 8,300 ft² DTS including concrete shield walls, a drive through truck bay, a 150 ton crane, a 10 ton crane, a cask preparation area, a fuel element transfer area, an operator control room, shielded windows, master-slave manipulators, a crane maintenance area, an electrical equipment room, a cask wash down area and liquid collection system, a criticality safety alarm system, HEPA filtered ventilation system, asphalt roadways, two cask transfer carts, a rail system between the cask preparation area and the fuel element transfer area, an inert purge gas system, radiological surveillance systems, camera viewing systems, canister welding systems, a Par remote manipulator extending from a crane trolley, and security systems. Based on the description of the Mu*STAR ADSR fuel conversion facility, it is expected that most of these facility items would be required to handle a large input of spent LWR fuel for conversion into molten salts.

The cost estimate for this facility was based on a conceptual design and thus was a rough order of magnitude estimate. The total final cost estimate for the DTS facility was $91M in 2016 dollars. For an 8,300 ft² facility, this equates to approximately $11K/ft² of hot cell floor area. No value was provided for the expected total square footage of the facility. Table 3 shows the breakout for the cost elements.

5.1.2 Sample Preparation Laboratory

The SPL (Figure 11) is a proposed hot cell facility designed for advanced post-irradiation examination at the Materials and Fuels Complex at INL. The formal cost estimate performed for this facility in 2016 is thorough and based on a complete design of the facility [Wasley, 2016a]. The cost estimate included the full project lifecycle from design to construction to commissioning. A separate formal cost estimate was developed at INL to cover the cost to operate and maintain the facility over a lifetime of 40 years [Wasley, 2016b].
Table 3. Cost estimate roll-up for the DTS facility

<table>
<thead>
<tr>
<th>Project element</th>
<th>Estimated cost in 2016 dollars</th>
</tr>
</thead>
<tbody>
<tr>
<td>Project development</td>
<td>$11.4M</td>
</tr>
<tr>
<td>Construction management</td>
<td>$8.8M</td>
</tr>
<tr>
<td>Preliminary/final design</td>
<td>$11.5M</td>
</tr>
<tr>
<td>AE support during construction</td>
<td>$1.8M</td>
</tr>
<tr>
<td>Project management</td>
<td>$8.0M</td>
</tr>
<tr>
<td>Project turnover and closeout</td>
<td>$8.3M</td>
</tr>
<tr>
<td>DTS construction contract</td>
<td>$40.9M</td>
</tr>
<tr>
<td><strong>Total cost for design, construction, and commissioning</strong></td>
<td><strong>$90.7M</strong></td>
</tr>
</tbody>
</table>

The SPL facility will be three stories consisting of 900 ft$^2$ of hot cell area and 43,500 ft$^2$ of total floor area. The facility design includes shielded hot cell windows, remote manipulators, hot cell concrete with a stainless-steel liner, a cask transfer system, decontamination equipment, and hot cell research equipment. The facility includes all other standard nuclear facility equipment including radiological monitoring equipment, HEPA filtered ventilation and an exhaust stack, cask handling features, etc.

The total design, construction, and commissioning cost for the SPL is estimated at $75M in 2016 dollars. This equates to approximately $83K/ft$^2$ of hot cell area or $2K/ft^2$ of total building square footage.

Table 4 shows the breakout for the cost elements for the SPL.
Table 4. Cost estimate roll-up for the SPL

<table>
<thead>
<tr>
<th>Project element</th>
<th>Estimated cost in 2016 dollars</th>
</tr>
</thead>
<tbody>
<tr>
<td>Project management, design/construction integration</td>
<td>$13.0M</td>
</tr>
<tr>
<td>Supporting documentation including nuclear safety basis</td>
<td>$2.8M</td>
</tr>
<tr>
<td>Site-work and utilities</td>
<td>$2.1M</td>
</tr>
<tr>
<td>Facility structures</td>
<td>$14.5M</td>
</tr>
<tr>
<td>Interior systems</td>
<td>$11.7M</td>
</tr>
<tr>
<td>Research environment including remote handling features</td>
<td>$16.9M</td>
</tr>
<tr>
<td>Research hardware</td>
<td>$8.7M</td>
</tr>
<tr>
<td>Commissioning</td>
<td>$3.2M</td>
</tr>
<tr>
<td><strong>Total cost for design, construction, and commissioning</strong></td>
<td><strong>$74.7M</strong></td>
</tr>
</tbody>
</table>

The operations and maintenance costs are estimated at $1.22B over the 40-year life of the facility. This cost includes escalation over a 40-year time period to account for inflationary effects. The first year (2016) operations and maintenance estimate was $8M, which is the approximate budget for the existing operations and maintenance activities at the two major INL hot cell facilities, the Hot Fuel Examination Facility and the Fuel Conditioning Facility. The cost for operations and maintenance at year number 40 in the cost estimate is $36M. The operations and maintenance budgets include the minimum staffing required to ensure safe and compliant operations of the nuclear facility as well as general facility maintenance, spare parts, system upgrades, and utility costs.

Decontamination and decommissioning at end of life was estimated at $300M in 2016 dollars. This value was adopted directly from a D&D project cost for a similarly sized facility that underwent D&D by the Idaho Cleanup Project prime contractor at the INL site.

5.2 PARAMETRIC DATA—ACTUAL COSTS OF PREVIOUS DOE HOT CELL FACILITIES

In addition to some of the latest parametric cost data for recent DOE hot cell designs, it is valuable to consider historical costs of previous hot cell facility construction. For the purposes of this document, the best data that exists for a facility that is comparable to the proposed Mu*STAR ADSR fuel conversion facility is the Fuels and Materials Examination Facility (FMEF) that was constructed at the Hanford Site outside of Richland, Washington. The SPL analyzed above is a relatively small hot cell facility, whereas FMEF hot cell is approximately 10 times the proposed size of the SPL hot cell. The FMEF hot cell size correlates well with what would be required for the magnitude of remote operations and pyrochemical processing that would be required for a full-scale Mu*STAR ADSR fuel conversion facility.

5.2.1 Fuels and Materials Examination Facility

The FMEF (Figure 12) was the last large inert-atmosphere hot cell facility constructed with DOE funding. The facility construction was initiated in 1978 and was completed in 1984. FMEF was constructed for fuel system development and irradiated materials analysis. The facility includes approximately 9,000 ft² of hot cell area and 215,000 ft² of support and laboratory area. The facility included three remotely operated process hot cells including a main hot cell, a decontamination hot cell, and an upper process hot cell. The facility included truck and rail car unloading mechanisms. The facility also includes an automated special nuclear material vault. Figure 13 shows a cutaway of the FMEF [FMEF Design Information, 2004].
By the end of construction in August 1984, the actual cost of the facility was $234M in 1984 dollars. Assuming that construction costs have increased by a factor of approximately 2.5 (the US dollar inflation rate has averaged about 2.6% since 1984), the cost to build an FMEF today would be $585M. This equates to about $65K/ft$^2$ of hot cell area or $3K/ft^2$ of total facility square footage.
5.2.2 Recent Nuclear Facility Construction

It should be noted that some nuclear fuel cycle cost-estimating experts within the DOE complex believe that the FMEF was built for a cost that would not be possible today. The FMEF was constructed before the regulatory impacts to the nuclear industry driven by the Three Mile Island accident, which occurred in March 1979.

According to experts at Savannah River National Laboratory:

All major DOE nuclear material processing projects since the 1980's at all sites translate today to costs in the range of $20,000 per ft$^2$. This database includes 1980 and 1990 vintage projects like the Defense Waste Processing Facility (DWPF) at the Savannah River Site, the Replacement Tritium Facility (RTF) and the Tritium Extraction Facility (TEF) also at the Savannah River Site, the Fuel Processing Restoration (FPR) Facility at INL, and several smaller facilities. In this decade it is seen in the plutonium disposition projects at [the Savannah River Site, the Chemistry and Metallurgy Research Replacement Nuclear Facility (CMRR-NF) at Los Alamos, the Uranium Processing Facility (UPF) and the Highly Enriched Uranium Material Facility (HEUMF) at Y-12, and the Waste Treatment Plant (WTP)] at Hanford. This cost profile is not exclusively a DOE artifact—it is also in the same range as the world’s newest commercial reprocessing plant at Rokasho, (Japan) [private communication from G. Teske, INL, to B. Dixon, SRNL, June 2010].

Along these lines, Module R7 (molten salt reactors) of the DOE Advanced Fuel Cycle Cost Basis document states that

...non-direct construction factors other than inflation have driven up capital costs since 1970. Among these factors would be regulatory, financial, schedule, start-up, quality assurance, environmental safety and health, and security related requirements and factors which have changed from 1970 or did not even exist in 1970 ... For most of today’s nuclear projects, these “people-related” indirect costs are now a much higher fraction of or are equal to the direct construction costs. This is a result of today’s regulator-mandated requirements for quality control, design verification, inspections, etc. [DOE, 2017]

Additionally, the Integrated Waste Treatment Unit at the INL is a hot cell facility currently undergoing commissioning activities. It is a 53,000 ft$^2$ facility whose mission is to treat sodium-bearing liquid nuclear waste. The facility has surpassed the $1B mark for design, construction, and commissioning. Assuming the final cost for the Integrated Waste Treatment Unit will be approximately $1B, the cost per square footage for the facility will be approximately $19K/ft$^2$, which is in agreement with the number proposed by the SRNL cost-estimating experts.

5.3 COST ESTIMATE FOR THE ADSR FUEL CONVERSION FACILITY

The recent cost estimates for DOE hot cell facility designs and analysis of historical costs of DOE facilities allows development of an estimated level of magnitude cost for the proposed design and construction of the Mu*STAR ADSR fuel conversion facility. The following sections provide a summary of the previous analyses and provide high and low ranges for the potential costs for the design, construction, and operation of the fuel conversion facility.
5.3.1 Design and Construction

Table 5 presents a comparison of the design, construction, and commissioning costs discussed earlier in this document.

<table>
<thead>
<tr>
<th></th>
<th>SPL (estimate)</th>
<th>DTS (estimate)</th>
<th>FMEF (actual)</th>
<th>SRNL Advisory (survey)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$(900 \text{ ft}^2 \text{ hot cell and } 43,500 \text{ ft}^2 \text{ total})</td>
<td>$(8,300 \text{ ft}^2 \text{ hot cell})</td>
<td>$(9,000 \text{ ft}^2 \text{ hot cell and } 215,000 \text{ ft}^2 \text{ total})</td>
<td></td>
</tr>
<tr>
<td><strong>Total cost</strong></td>
<td>$74.7M</td>
<td>$90.7M</td>
<td>$585M</td>
<td>—</td>
</tr>
<tr>
<td><strong>Hot cell area cost</strong></td>
<td>$83K/ft$^2$</td>
<td>$11K/ft$^2$</td>
<td>$65K/ft$^2$</td>
<td>—</td>
</tr>
<tr>
<td><strong>Total facility area cost</strong></td>
<td>$2K/ft$^2$</td>
<td>—</td>
<td>$3K/ft$^2$</td>
<td>$20K/ft$^2$</td>
</tr>
</tbody>
</table>

Based on Table 5, and assuming that the square footage of the Mu*STAR ADSR fuel conversion facility is between that of the SPL and FMEF, the total design, construction, and commissioning cost will be above $100M. Depending on the size and complexity of the facility that would be provided in a final design, a risk does exist that the cost of the fuel conversion facility could be around $1B, if the cost per total square footage is actually 7–10 times that shown for SPL or FMEF in Table 1.

5.3.2 Operations and Maintenance Costs

Based on the actual budgets for the operations and maintenance of the major hot cell facilities at INL, it is estimated that these costs would be on the order of $10M/year in 2018 dollars. With escalation over a lifetime of 40 years, the cost would total approximately $1B to operate and maintain the facility.

5.4 FLOW SHEET FUEL CONVERSION EQUIPMENT COSTS

Since 2016, INL has installed a series of unit operations equipment in the Hot Fuel Examination Facility main hot cell for performing pyrochemical operations on the kilogram scale. This fully operational equipment includes LWR fuel decladding equipment, an oxide reduction system, an electrorefining system, two salt distillation systems, and a metal fuel casting system. The total cost for design, fabrication, and installation of this equipment was approximately $5M/each. Additionally, a metal waste form furnace is being designed for installation in the Fuel Conditioning Facility hot cell in 2020. The current total project budget for this kilogram-scale system is $6M.

These INL systems are all designed to function on the kilogram scale, or a scale of approximately 1–10 fuel rods per batch. The scale of the Mu*STAR ADSR fuel conversion equipment would be larger by an order of magnitude assuming the equipment will work with full fuel assemblies during operational batches. It is likely that design, fabrication, installation, and commissioning of each unit operation system will be greater than $10M. Given the assumed flow sheets in Figures 2–4, this would equate to at least $50M in process equipment.

5.4.1 Waste Management

Waste management costs for a fuel conversion facility will be significant. Potential waste streams for the facility that will require packaging and shipment to a repository include:

- A large volume of low-level radioactive waste from routine nuclear operations
• A large volume of highly radioactive waste derived from the metal fuel assembly hardware

• A metal waste form derived from fuel cladding that could be more expensive to dispose of, due to the potential transuranic loading in the metals

• And a ceramic waste form derived from the fuel salt, which will be an expensive waste form because of its transuranic loading and high fission product content.

Estimating the costs of disposal of such wastes from the Mu*STAR ADSR fuel conversion facility is not within the scope of this document, but it is discussed here because it is an operational cost that should not be overlooked. The geological disposal costs for high-level waste are estimated to be $500–$2,000/kg of UNF, whereas the disposal costs for low-level waste in the United States is about $20/kg.

The waste management for a pyrochemical-based fuel conversion facility should not be underestimated. Waste form development for pyrochemical reprocessing is a major research field within the DOE complex because of the known challenges. The regulatory and technical challenges of dealing with the by-products of converting LWR fuel to a molten salt fuel will be similar to the technical challenges faced in both the pyrochemical nuclear fuel reprocessing field and the molten salt reactor field.

5.4.2 End-of-life D&D

For purposes of completeness, a cost for D&D of the fuel conversion facility has been included based on D&D costs of legacy hot cell facilities at INL. It is assumed that D&D costs for the Mu*STAR ADSR fuel conversion facility will be greater than $300M.

6. NUCLEAR FUEL CYCLE CONSIDERATIONS

This section explores the nuclear fuel cycle needed for the Mu*STAR ADSR and compares this fuel cycle to the following three fuel cycles:

1. The current US fuel cycle including dry storage of spent LWR fuel and proposed shipment of this spent fuel to a future geologic repository

2. A once-through fuel cycle with aqueous reprocessing where mixed oxide fuel is fabricated and used in an existing fleet of LWR reactors

3. A continuous-recycle fuel cycle with pyrochemical reprocessing where metal fuel is fabricated and used in a metal-fueled fast reactor

The primary references for this discussion are the publicly available DOE Advanced Fuel Cycle Cost Basis Report [DOE, 2017] and the DOE Nuclear Fuel Cycle Evaluation and Screening [DOE, 2014]. These documents are available on the web at https://fuelcycleoptions.inl.gov/SitePages/Home.aspx and are the most detailed and all-encompassing review of existing and proposed nuclear fuel cycles. Modules R6 and R7 of the Advanced Fuel Cycle Cost Bases Report cover ADSRs and molten salt reactors, respectively. The Mu*STAR ADSR is a hybrid of these two technologies.

The DOE fuel cycle analysis documents include high, low, and median cost estimates for many fuel cycle options. Within these documents, the fuel cycles are usually discussed in cost per metric ton of material processed for reprocessing and cost per megawatt electricity generated for reactors. The documents include a comprehensive economic comparison of fuel cycle options. The cost basis document includes a
fuel fabrication module (Module D), but it does not include molten salt fuel fabrication specifically. It does touch on this though in the R7 module, which, as stated, is specific to a molten salt reactor itself.

The second most important reference used in this document is the 2003 Harvard study titled “The Economics of Reprocessing vs. Direct Disposal of Spent Nuclear Fuel” [Bunn, 2003]. Because fabricating, or converting, a molten salt fuel from LWR fuel is essentially a reprocessing activity, most of the discussion in this Harvard study is applicable to the Mu*STAR ADSR fuel cycle.

For the discussion in the following subsections, a brief comparison of fuel cycles is performed using the data from these primary references.

6.1 COST/BENEFIT ANALYSIS

Using the cost data provided in both the DOE cost basis documents [DOE, 2014 and 2017] and the Harvard study [Bunn, 2003], a high-level cost/benefit analysis is performed below. The Mu*STAR ADSR fuel cycle is compared with each of three major fuel cycle options.

6.1.1 Mu*STAR ADSR Fuel Cycle vs. Open Fuel Cycle with Dry Storage

Figure 14 shows a block diagram comparison of the Mu*STAR ADSR Fuel Cycle with the current US fuel cycle. The Mu*STAR ADSR fuel cycle begins at the point of dry storage (Figure 15) of LWR fuel from the open fuel cycle. The fuel cycle cost listed in green for the open cycle is solely for the interim dry storage portion of the fuel cycle from the DOE cost basis report Module I. It does not include the geologic repository cost of $600/kgHM (Module L) because waste disposal costs have been neglected in the reprocessing fuel cycle diagrams below.

![Figure 14. Comparison of Mu*STAR ADSR fuel cycle with the current US fuel cycle.](image-url)
The Mu*STAR ADSR fuel cycle avoids the geologic repository cost for LWR UNF found in the current US open fuel cycle. Nevertheless, the Mu*STAR ADSR fuel cycle generates its own waste concerns and costs in the form of the waste disposal of hardware, cladding hulls for some options, and eventually the fuel salt, all of which are difficult and expensive wastes to dispose from a regulatory perspective. The benefit of such wastes, though, is that typically the actinides are bound in a condensed form that takes up much less volume than an LWR fuel assembly in a geologic repository.

The Mu*STAR ADSR adds an accelerator and a molten salt reactor to the fuel cycle, in addition to a fuel conversion facility as discussed in the earlier sections of this document. Such facilities involve large capital and operating costs that would need to be balanced by benefits of the Mu*STAR ADSR such as additional electricity generation, the ability to burn excess weapons-grade plutonium, and the use of excess generated heat to convert natural gas to synthetic diesel fuel.

6.1.2 Mu*STAR ADSR Fuel Cycle vs. Closed Fuel Cycle with Aqueous Reprocessing

The advantage of comparing any fuel cycle to a fuel cycle containing aqueous reprocessing is that industrial aqueous reprocessing is being demonstrated on a daily basis and real cost data is available. This fact is contrary to the case of pyrochemical reprocessing or sending spent nuclear fuel to a national repository site. Figure 16 shows a comparison between an aqueous reprocessing fuel cycle and the Mu*STAR ADSR fuel cycle. The cost shown for the aqueous reprocessing fuel cycle is the sum taken from Module F1 (aqueous reprocessing) and Module D1-2 (LWR mixed oxide fuel fabrication) of the DOE cost basis documents [DOE, 2014 and 2017] The costs do not include any reactor capital or reactor operating costs nor any waste disposal costs.
Figure 16. Comparison of Mu*STAR ADSR fuel cycle with a once-through aqueous reprocessing fuel cycle.

Figure 16 makes apparent that a large quantity of waste is generated because UNF and the by-products of reprocessing remain for disposal in this aqueous reprocessing fuel cycle. Despite the significant waste management issue, a potential benefit of this once-through fuel cycle is that it uses an existing fleet of LWR reactors to burn the recycled mixed oxide fuel. Figure 17 shows a photograph of the major aqueous reprocessing facility at La Hague in northern France.

Figure 17. La Hague nuclear recycling and reprocessing plant in northern France.

One of the arguments against the cost benefits of reprocessing is that uranium costs have decreased, whereas in the early stages of the nuclear industry, it was assumed that uranium resources were scarce and
that the cost of fabricating new LWR fuel would grow to be unmanageable. The Harvard reprocessing economics study [Bunn, 2003] states:

The margin between the cost of reprocessing and recycling and that of direct disposal is wide, and is likely to persist for many decades to come. In particular, at a reprocessing price of $1000/kgHM, and with our other central estimates for the key fuel cycle parameters, reprocessing and recycling plutonium in existing light-water reactors (LWRs) will be more expensive than direct disposal of spent fuel until the uranium price reaches over $360 per kilogram of uranium (kgU)—a price that is not likely to be seen for many decades, if then. At a uranium price of $40/kgU (comparable to current prices), reprocessing and recycling at a reprocessing price of $1000/kgHM would increase the cost of nuclear electricity by 1.3 mills/kWh. Since the total back-end cost for the direct disposal is in the range of 1.5 mills/kWh, this represents more than an 80% increase in the costs attributable to spent fuel management. . . World resources of uranium [are] likely to be economically recoverable in future decades at prices far below the breakeven price amount to tens of millions of tons, probably enough to fuel a rapidly-growing nuclear enterprise using a once-through fuel cycle for a century or more.

Such an argument could also be applied to the Mu*STAR ADSR fuel cycle because it too ventures into the realm of reprocessing, although there is no separation of plutonium from the fission products in the Mu*STAR fuel. But as stated above, the additional peripheral benefits to the Mu*STAR ADSR may balance some of the expensive cost elements of its fuel cycle compared to the open fuel cycle.

6.1.3 Mu*STAR ADSR Fuel Cycle vs. Closed Fuel Cycle with Pyrochemical Reprocessing

Figure 18 shows a comparison of a pyrochemical reprocessing fuel cycle and the Mu*STAR ADSR fuel cycle. The pyrochemical fuel cycle shown is considered continuous recycle, where the spent metal fuel from the fast reactor could be recycled back through the pyrochemical reprocessing facility. The fuel cycle cost shown in green is taken from module F2/D2-1 of the DOE Cost Basis documents [DOE, 2014 and 2017] and includes both reprocessing and metal fuel fabrication.

Figure 18. Comparison of Mu*STAR ADSR fuel cycle with a pyrochemical reprocessing fuel cycle.
The two fuel cycles in Figure 18 are the most directly comparable because of the similar waste output from the processes and because both require a newly constructed nuclear reactor. The potential benefit of the Mu*STAR ADSR fuel cycle is shown in yellow, where the reprocessing and fuel fabrication are essentially performed in one step. The pyrochemical “reprocessing” or “conversion” step for the Mu*STAR ADSR does not require an additional remote-handled solid-fuel fabrication step like the typical pyrochemical reprocessing fuel cycle.

Pyrochemical recycling has been demonstrated at the INL at the engineering/kilogram scale for the recycling of oxide fuel at the Hot Fuel Examination Facility and at a larger scale for the processing of metal fuels at the Fuel Conditioning Facility. Figure 19 shows a rendering of the Advanced Pyroprocess Recycling Facility of the Integral Fast Reactor Program. This DOE program developed the technology for a complete system including the reactor, the full fuel cycle, and all necessary waste management technologies. Nevertheless, this technology and fuel cycle has not yet been industrialized. Many of the challenges related to molten salt fuel fabrication applicable to the Mu*STAR ADSR fuel conversion process have been learned through the pyrochemical research and operations at INL as well as through the activities on molten salt reactors at ORNL.

Figure 19. Advanced Pyroprocess Recycling Facility from Integral Fast Reactor Program.

A large benefit of both the pyrochemical and Mu*STAR ADSR fuel cycle is their ability to concentrate actinides and fission products in waste forms through low volume chemical separations. Such benefits can relieve the burden that siting and designing a geologic repository may entail. And as discussed previously, the potential peripheral benefits of the Mu*STAR ADSR may, over time, outweigh the challenges of the upfront nuclear facility capital costs.

7. SUMMARY AND CONCLUSIONS

In conclusion, the Mu*STAR ADSR fuel conversion facility will cost more than $100M, possibly up to around $1B, to design and construct. Operations and maintenance of the facility will be on the order of $10M per year. The design, fabrication, and installation of the remote process equipment to support the fuel conversion will cost more than $50M and should be included in the total cost to design and construct the fuel conversion facility. Cost surveys performed on DOE hot cell designs and previous DOE hot cell
construction suggest that the cost variance may be large, and the risk of escalating costs may be high in the regulation-mandated atmosphere of nuclear facility design and construction.

When normalized to the amount of electricity produced, the Mu*STAR ADSR fuel cycle’s expected cost is likely to be less than any of the other reprocessing fuel cycles that have been developed and possibly less than the once-through cycle now used. Simulations show that once the conversion to fluoride is made, that seven times as much energy can be extracted from the UNF than was produced by the LWR while making it. Using once-only, on-site conversion to fluoride means no mining, no enrichment, no manufacturing of fuel rods, and no transportation costs. Typically, the UNF was made over a 40-year period in an LWR, and it can be used for up to 280 years in Mu*STAR producing the same amount of power as the LWR did, ultimately requiring disposal of approximately the same volume of material as the LWR generated. Mu*STAR burns most of the higher actinides, which reduces both the cost of disposal and significantly reduces the longevity of its radioactivity.

The Mu*STAR ADSR’s ability to demonstrate unique and valuable peripheral benefits are additional arguments for this technology, including (1) turning UNF into a valuable commodity (thus its disposal will not be such an immediate problem, so continued operation of existing LWRs will be more acceptable); (2) the ability to burn any fissile or fertile fuel including excess weapons-grade plutonium in subcritical mode; (3) providing high-temperature process heat to convert natural gas to synthetic diesel fuel, to convert water to hydrogen, or for other process-heat applications.

8. REFERENCES


APPENDIX A. MODELING THE IMPACTS OF LITHIUM ENRICHMENT ON TRITIUM PRODUCTION

The fluoride salt in the Mu*STAR ADSR core will be irradiated with the fuel, and one of the major concerns for source term release will be the tritium generated in the salt. Tritium will be produced even if it were possible to get the lithium to be pure $^7$Li. There are five methods for tritium to be produced in the fluoride salt consisting of LiF and BeF$_2$ as shown below:

\[
\begin{align*}
^6\text{Li} + n &\rightarrow ^4\text{He} + ^3\text{H} \\
^7\text{Li} + n &\rightarrow ^4\text{He} + ^3\text{H} + n \\
^7\text{Li} + n &\rightarrow ^5\text{He} + ^3\text{H} \\
^9\text{Be} + n &\rightarrow ^4\text{He} + ^6\text{Li} \\
^{19}\text{F} + n &\rightarrow ^{17}\text{O} + ^3\text{H}
\end{align*}
\]

Ternary fissions can produce tritium also, but this impact is much smaller and is not impacted by the lithium enrichment. Tritium has a decay half-life of 12.32 years, while its specific activity is 9650 Ci/g, or $3.57 \times 10^{14}$ Bq/g. The reaction between a neutron and $^7$Li requires that the energy of the neutron is above 0.546 MeV. And for the reaction between $^{19}$F and a neutron, the neutron energy required is above 9.5 MeV, which is rare in a nuclear reactor. Although the reaction between $^9$Be and a neutron does not generate tritium directly, it produces $^6$Li.

The tritium generated in fluoride salt exists initially in the charge state T+, and it combines with F− or T to form TF or T$_2$ molecules. The ratio of TF and T$_2$ depends on the chemical redox potential of the primary coolant [Rosenthal et al., 1970]. As TF is corrosive to structure materials, it must be removed or converted to less corrosive chemicals as soon as possible after its generation in the reactor core [Ingersoll et al., 2004].

Because of the design of the Mu*STAR ADSR, the production rate of tritium cannot be lowered to the level of other reactor types. The tritium release from the Mu*STAR ADSR must be mitigated with a tritium management system.

The tritium generation rate depends on the initial $^6$Li inventory in the salt. Therefore, enriching the salt to reduce the $^6$Li concentration is required. Even after enriching the salt to 99.99% $^7$Li, the initial $^6$Li contributed greatly to the total tritium generation, as shown below in Figure A-1.
Figure A-1. Total tritium produced with 99.9% $^7\text{Li}$ content in LiF.

The effect of the ratio of $^6\text{Li}$ to $^7\text{Li}$ in the LiF salt was calculated based upon the rate equations of the various isotopes in the salt:

$$\frac{dN_T}{dt} = \Phi \sigma_T^{^7\text{Li}} N_{^7\text{Li}} + \Phi \sigma_T^{^6\text{Li}} N_{^6\text{Li}}$$

$$\frac{dN_{^6\text{Li}}}{dt} = -\Phi \sigma_T^{^6\text{Li}} N_{^6\text{Li}} + \Phi \sigma_\alpha^{^9\text{Be}} N_{^9\text{Be}}$$

$$\frac{dN_{^7\text{Li}}}{dt} = -\Phi \sigma_T^{^7\text{Li}} N_{^7\text{Li}}$$

$$\frac{dN_{^9\text{Be}}}{dt} = -\Phi \sigma_\alpha^{^9\text{Be}} N_{^9\text{Be}}$$

The tritium production from $^{19}\text{F}$ and ternary fissions was neglected in these calculations as their contribution to total tritium production is minimal compared to the other production terms. The key parameters used in the calculations are shown below in Table A-1.
Table A-1 Key parameters used for tritium production calculations

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value 1</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average thermal neutron flux [Cacuci, 2010]</td>
<td>1 × 10^{14}</td>
<td>n/cm^{2}s</td>
</tr>
<tr>
<td>Core salt content (Stream 15) [Roberts, 2018]</td>
<td>17.6</td>
<td>MT</td>
</tr>
<tr>
<td>LiF salt % [Roberts, 2018]</td>
<td>65%</td>
<td>—</td>
</tr>
<tr>
<td>BeF_{2} salt % [Roberts, 2018]</td>
<td>30%</td>
<td>—</td>
</tr>
<tr>
<td>Li-6 (n, T)</td>
<td>150</td>
<td>bn</td>
</tr>
<tr>
<td>Be-9 (n, alpha)</td>
<td>3.5 × 10^{-3}</td>
<td>bn</td>
</tr>
<tr>
<td>Li-7 (n, T)</td>
<td>1.0 × 10^{-3}</td>
<td>bn</td>
</tr>
</tbody>
</table>

The generation rate of tritium from the salt changes from the beginning of life to the steady state production rate because of the consumption of $^6$Li. At the beginning of life, the tritium generation rate is dominated by the consumption of $^6$Li. As the $^6$Li is consumed, the reaction rate decreases until the contributions from the $^6$Li generated from $^9$Be reaction reaches equilibrium rate. With the quantities of $^7$Li and $^9$Be initially in the salt, these create a quasi–steady state generation rate, and they can be essentially be treated as infinite sources. This effect can be seen below in Figure A-2.

![Figure A-2. Comparison of the production rates of tritium with varying $^7$Li content of the salt.](image)

Shown in Table A-2 below is the total tritium produced during the operation with varying $^7$Li concentration from 99%–100%. For the quantity of tritium produced during the Mu*STAR ADSR operating life, these results show a definite separation between 99.9% and only 99% $^7$Li in the salt.
Table A-2 Total tritium production with varying $^7$Li salt content

<table>
<thead>
<tr>
<th>$^7$Li salt content</th>
<th>1 year</th>
<th>5 year</th>
<th>10 year</th>
<th>20 year</th>
<th>50 year</th>
<th>100 year</th>
<th>200 year</th>
</tr>
</thead>
<tbody>
<tr>
<td>100%</td>
<td>0.007</td>
<td>0.05</td>
<td>0.10</td>
<td>0.21</td>
<td>0.55</td>
<td>1.1</td>
<td>2.2</td>
</tr>
<tr>
<td>99.9%</td>
<td>0.05</td>
<td>0.16</td>
<td>0.23</td>
<td>0.35</td>
<td>0.7</td>
<td>1.3</td>
<td>2.4</td>
</tr>
<tr>
<td>99.95%</td>
<td>0.24</td>
<td>0.6</td>
<td>0.8</td>
<td>0.9</td>
<td>1.2</td>
<td>1.8</td>
<td>2.9</td>
</tr>
<tr>
<td>99.9%</td>
<td>0.482</td>
<td>1.2</td>
<td>1.4</td>
<td>1.5</td>
<td>1.9</td>
<td>2.4</td>
<td>3.6</td>
</tr>
<tr>
<td>99%</td>
<td>4.8</td>
<td>11.9</td>
<td>13.3</td>
<td>13.5</td>
<td>13.9</td>
<td>14.4</td>
<td>15.6</td>
</tr>
</tbody>
</table>

Reducing the initial $^6$Li content of the salt is important to minimize the production of tritium over the life of the Mu*STAR ADSR. In addition, any additional $^6$Li in the salt could reduce the power output of the Mu*STAR ADSR. Mitigation of the initial surge in production rate will need to be handled by capturing and cleaning the purge gas to remove the tritium to minimize the release source term for the Mu*STAR ADSR.

**APPENDIX A REFERENCES**


APPENDIX B. IMPACT OF TRITIUM IN USED FUEL CONVERSION FACILITY

As indicated in Table 1, each fuel assembly will contain 30 Ci of tritium after 40 years based on Catawba 1 17 × 17 array fuel assemblies. The current Mu*STAR ADSR concept uses 17.6 t of used nuclear fuel (UNF) (about 40 assemblies) containing about 1,190 Ci of tritium. This amount of tritium may require cleanup of the waste gas before it is released to the atmosphere.

Atmospheric release regulations vary greatly by location, regulatory body, and facility type. Some states (e.g., California) are much more restrictive than federal requirements. Although the release of 1,190 Ci tritium to the atmosphere in 1 year is on the same order as atmospheric tritium releases from most PWRs, the possibility for more stringent regulations should be analyzed. Also, fresh UNF without the 3+ half-lives of tritium decay (12.3 years) would contain about 10 times as much tritium (11,900 Ci).

Tritium Capture in Conversion Processes That Use Voloxidation Decladding

Conventional Tritium Capture: Breaching of the Zircaloy cladding by chopping/shearing may release elemental tritium (HT, T$_2$) as a gas. This can be converted to oxide (water vapor) by a small amount of precious metal catalyst (Deoxo®) or an oxygen supplying catalyst (hopcalite, CuO + MnO). The fuel conversion methods that use oxygen or dry-air voloxidation (as described in Sections 2.1 and 2.3, option 3.) will oxidize the tritium and hydrogen to form tritiated water vapor. The tritiated water vapor can be easily removed from the effluent stream by absorption on a desiccant material, typically zeolite (type 3A, 4A, 5A, or 13X). These materials are relatively inexpensive and available from several commercial vendors. This is the method used in almost all tritium handling facilities, usually in combination with an oxidation catalyst, for stripping tritium from air, nitrogen, helium, or argon [Hsu, 1997]. Since the tritium released from voloxidation will already be oxidized, an oxidation catalyst will not be necessary.

Greater than 90% of the tritium will be removed in the voloxidation step, and the majority of the remaining tritium will be bound in the Zircaloy cladding as very stable zirconium tritide [International Atomic Energy Agency, 2004]. In a suitable waste package, that tritium will remain bound in the Zircaloy hulls without concern for tritium release. For this reason, there should be very little tritium remaining to be released during subsequent fluoridation. According to a recent ORNL paper, the tritium in the cladding is estimated to vary from 0% to 96%, and correlations with burnup and other parameters are not well characterized [Robinson, 2015]. This indicates that the specific fuel to be used needs to be characterized as early as the beginning of the project design process.

Although the focus of this section is tritium removal, examples of the removal of other gas stream contaminants are included in Figure B-1 to show the tritium removal system in context. Metal wools of copper, bronze, and surface-modified stainless steel (functionalized with Au nanoparticles) were demonstrated to effectively trap Zn metal vapor for the Tritium Extraction Facility at the Savannah River Tritium Enterprise [Korinko, 2017, and Murph, 2017]. Metal wools may be capable of trapping other metal vapors from the voloxidizer off-gas stream.

Capture of xenon and krypton by novel modified mordenite sorbents was studied and reported by Idaho National Laboratory as part of a Department of Energy (DOE)-sponsored Off-Gas Sigma Team tasked with a multilaboratory collaborative research and development effort [Garn, 2013]. The purpose of the effort was to investigate and evaluate emissions and immobilization control technologies for the volatile radioactive species generated from commercial UNF reprocessing. The sorbent pellets were fabricated from type AgZ and HZ mordenite powders and a microporous binder. The materials were found to have a
high degree of sorption for xenon and an acceptable level for krypton at a temperature of 190 K. Warming to ambient temperature released the noble gases, possibly allowing for their recovery if desired.

Since there are no halogens in the voloxidation process, this system can be fabricated from stainless steel like typical tritium handling processes.

**Figure B-1. Proposed schematic for off-gas processing of sheer/voloxidation effluent.**

**HCl/TCl and/or HF/TF Capture and Tritium Recovery Problem in Chemical Decladding and Molten Salt Dissolution Processes**

There are no examples of tritium facility or tritium laboratory processes, past or current, that involve the quantities and concentrations of halides that will be present in Chemical Decladding or Molten Salt Dissolution of UNF. Two DOE directives covering tritium handling recommend against using chlorine- and fluorine-containing materials in contact with tritium [DOE, 2014 and 2015]. Even materials that are relatively stable are degraded by tritium exposure into products that can cause chloride-, or fluoride-induced stress corrosion cracking in stainless steels.

However, the tritium will be in dilute form, so the off-gas system can be viewed as an HCl capture system with a small amount of tritium. It still needs to be contained for personnel protection, but in this dilute form, the tritium beta decay is much less a factor for material degradation than the presence of Cl⁻.

That said, ORNL has tested a small-scale system to handle off-gas from the chemical decladding process described in Section 2.2 [Spencer, 2016]. The flow sheet of this process is shown in Figure B-2.
Figure B-2. Conceptual liquid scrubbing process design for recovery of HCl from dilute off-gas streams by chemical conversion.

For molten salt dissolution of UNF, the HF/TF in the off-gas should be controlled in the same way it is controlled in the Mu*STAR itself (i.e., by redox control with excess Be to convert the HF/TF to H₂, HT, and T₂ and to keep the fluoride out of the off-gas stream). In addition, it may be necessary to add a redox-controlled molten salt scrubber/bubbler as part of the off-gas system as shown in Figure B-3. The molten salt could be drained to the dissolver when it is saturated.
Figure B-3. Conceptual redox-controlled molten salt scrubber/bubbler for HF/TF removal.

APPENDIX B REFERENCES


