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| *Title:* | TRU Waste Determination of MSRE Salt for Disposal Eligibility in WIPP |
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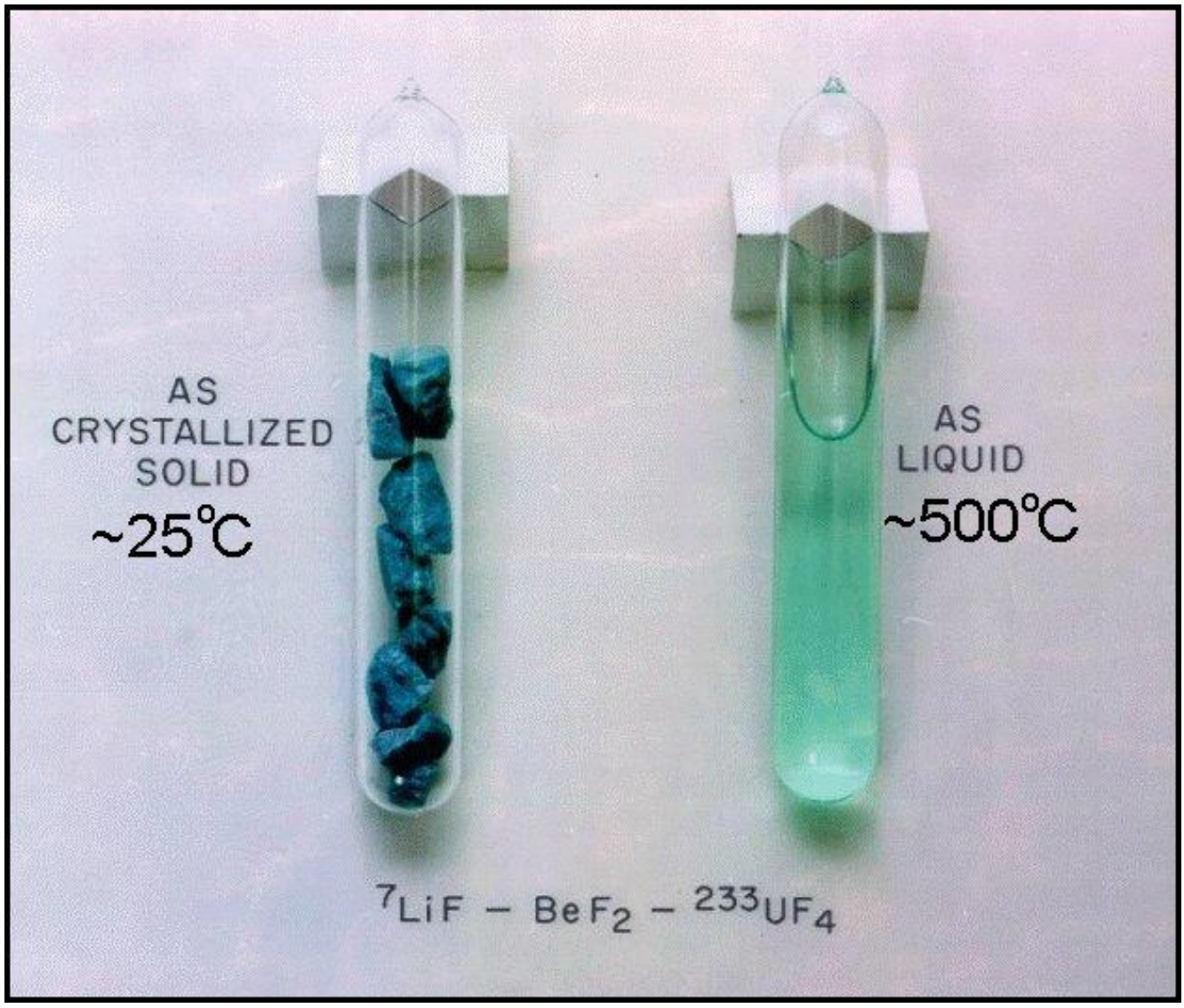
# TRU Waste Determination of MSRE Salt for Disposal Eligibility in WIPP

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## EXECUTIVE SUMMARY

The purpose of this evaluation is to examine the nature of the Molten Salt Reactor Experiment (MSRE) salt, which is currently stored and awaiting a disposition pathway at Oak Ridge National Laboratory (ORNL). This white paper presents evidence, from the best available information, of the candidacy for direct disposition of the MSRE salt as defense transuranic (TRU) waste at the Waste Isolation Pilot Plant (WIPP). An analogy between the MSRE salt and conventional water coolant systems of modern pressurized water reactors (PWR) is also presented.

After MSRE shutdown over 40 years ago, the salt/fuel mixture was stored, and short-lived fission products trapped in the solidified salt were allowed to decay. By 2008, all but a trace of the uranium in the mixture was removed via the process of fluorine sparging. The resulting salt has been safely stored since then, and radioactive decay has continued. During MSRE operation, a significant amount of plutonium was added to the reactor, raising the TRU isotopic concentration. Because the salt functioned as the primary coolant and not a component of the fuel, it is not high-level waste (HLW). In fact, if the plutonium was not present in the salt, the remaining fission products and trace uranium would meet the definition of low-level waste (LLW).

The radiological, chemical, and physical characteristics of the salt are evaluated in this white paper, which demonstrates that the MSRE salt may be considered TRU waste, with a clear defense pedigree, as required for disposal at WIPP. If suitably packaged, the MSRE salt would meet all of the waste acceptance criteria for WIPP disposal as remote-handled (RH)-TRU waste. Analysis and modeling of an MSRE salt waste stream packaged for disposal at WIPP show fissile gram equivalent (FGE) limitations (<305 FGE) for shipment in the U.S. Nuclear Regulatory Commission (NRC)-licensed RH-72B transport package, used for RH-TRU shipments to WIPP, would be easily met. Similarly, transportation dose rates would be well within U.S. Department of Transportation limits and WIPP waste acceptance criteria. Additionally, because there is no hydrogenous material or organics in the salt, flammable gas generation is not a concern. Fluorine would be generated by radiolysis but in such small quantities that container integrity would not be affected. Finally, the final waste form's total activity content is shown to be within WIPP Land Withdrawal Act (LWA) limits (<23 Ci/L).

In conclusion, this evaluation recommends that MSRE salt be packaged for ultimate shipment to WIPP. The unique nature of the salt dictates that special attention focus on demonstrating that this waste stream meets all WIPP waste acceptance criteria. The LWA specifically prohibits spent fuel and HLW disposal at WIPP, and this white paper conclusively shows that the MSRE salt is neither.

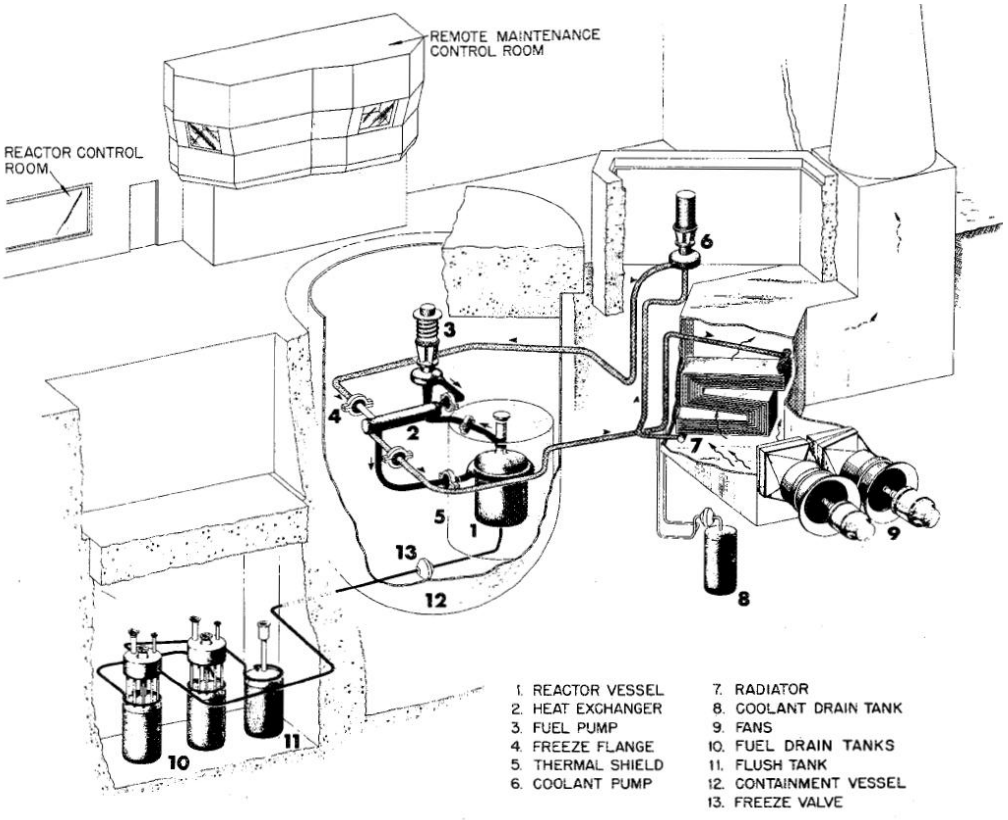
## INTRODUCTION

This white paper examines the nature of the MSRE salt, which is currently stored and awaiting a disposition pathway at ORNL. The historical information and data presented were gathered from sources available in the open literature. A bibliography is presented at the end of this paper. Calculations we performed were reviewed for technical accuracy, and the input for these calculations came from the best available data in the literature. These specific data are referenced directly in the text, with a reference list at the end of this paper.

Based on the best available information and calculations of expected properties of the MSRE salt, this evaluation recommends that the MSRE salt be packaged for ultimate shipment to WIPP for disposal. However, the certified values for physical and radiological properties of this material will have to be determined under a certified program prior to shipment and disposal at WIPP.

<u>Overview of the MSRE Operation</u>

Figure 1. MSRE schematic



The MSRE operated as a defense research reactor for the development of a molten salt breeder reactor. The reactor began operation in 1965 at a power level of approximately 8 MW (thermal) at ORNL and was shut down in 1969. A schematic of the reactor components is shown in Figure 1 (Reference 1).

The reactor operated by circulating uranium tetrafluoride ($UF_4$) fuel, dissolved in a **carrier salt** — composed of a mixture of lithium fluoride (LiF), beryllium fluoride ($BeF_2$), and zirconium fluoride ($ZrF_4$) — through channels in a graphite block located in the reactor vessel. Each time the system was opened for maintenance, the **carrier salt —** containing the fuel and products of actinide fission and transmutation — was drained into the fuel drain tanks, and a **flush salt** — composed of a $LiF/BeF_2$ mixture —was circulated through the reactor circuit.

Because of the unique nature of the molten salt reactor, terminology used to describe the systems, though logical, may be unfamiliar. Below are simplified diagrams of the MSRE and a PWR for comparison (Figures 2 and 3).

There are many similarities in design, with both the PWR and MSRE having two loops that intersect at a heat exchanger. The primary differences for the purposes of this paper are:

- The PWR has self-contained, pelletized fuel, and the MSRE has the fuel dissolved in the **carrier salt**.

- The PWR uses water as the primary and secondary coolant, and the MSRE has a molten $LiF/BeF_2$ salt mixture in each of the loops.

- The PWR secondary loop has steam that powers a turbine, which needs to be condensed for recycle, whereas the MSRE was operated at relatively low power and dumped its excess heat to the atmosphere through a radiator.

Figure 2. MSRE simplified flow diagram

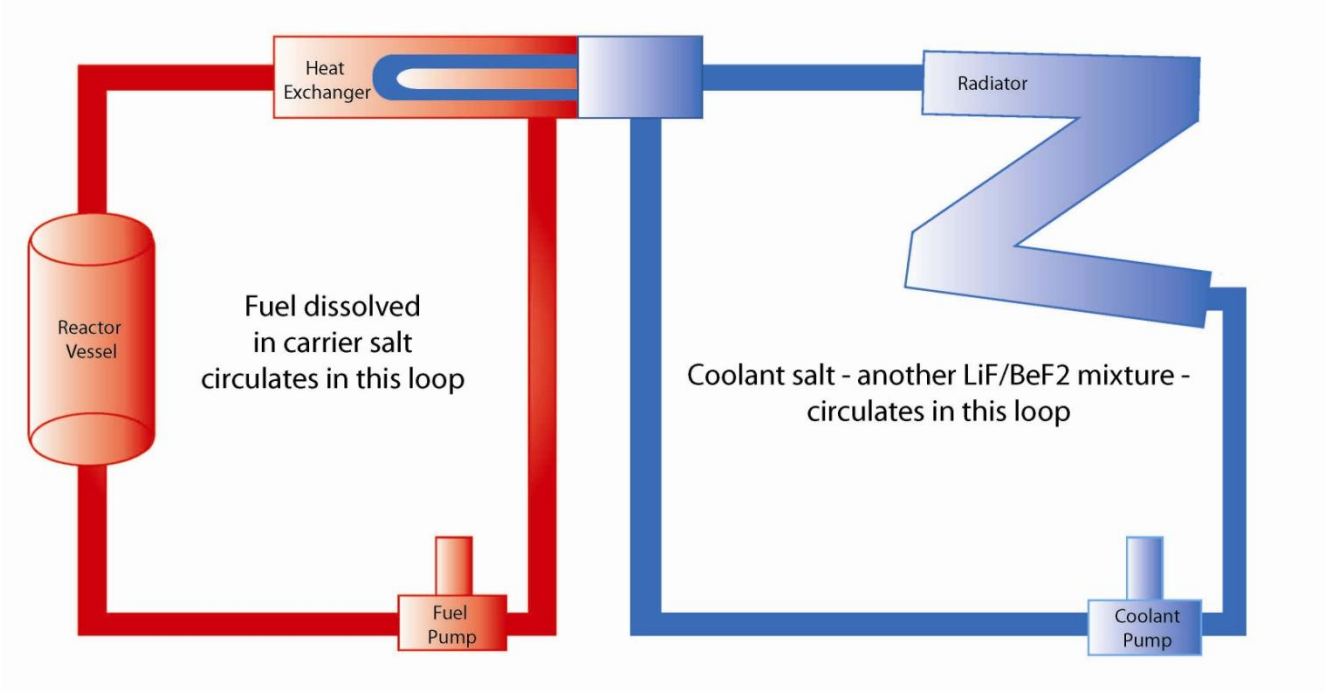
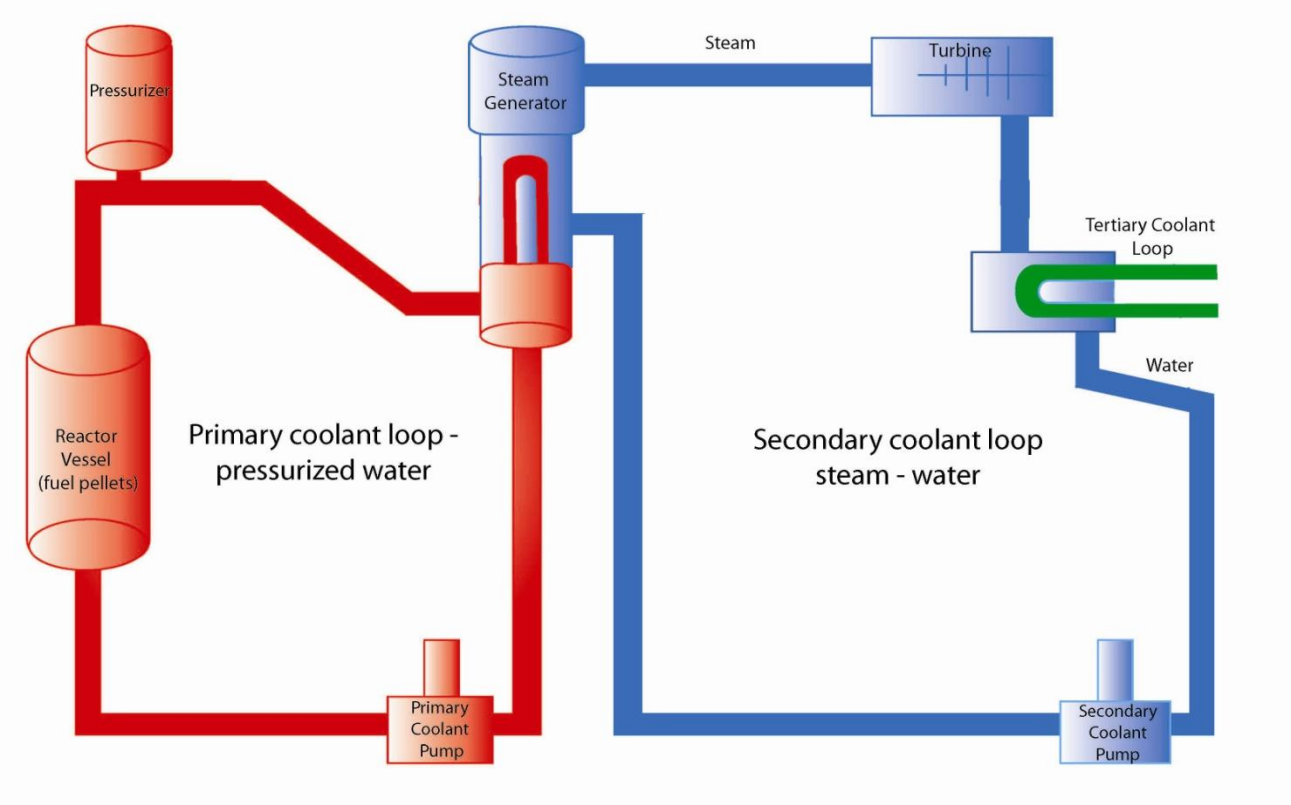



Figure 3. PWR simplified flow diagram



When MSRE operations were terminated in 1969, the **flush salt** and coolant salts were drained into their respective tanks (labeled 8 and 11 in Figure 1), where they cooled and solidified. The **carrier salt** (containing the $UF_4$ fuel) was divided between the two fuel drain tanks (labeled 10 in Figure 1), where it cooled and solidified. In 2008, the $UF_4$ was removed by fluorination processes from the **carrier salt,** reducing $^{232\text{-}238}U$ fuel amounts to trace levels. The **carrier salt** is stored in the fuel drain tanks. This paper focuses on the **carrier** and **flush salt** from the primary loop of the MSRE.

## CHEMICAL AND PHYSICAL PROPERTIES

The **carrier salt** for the fuel in the MSRE was initially prepared as 65 mole % LiF, 30 mole % $BeF_2$, and 5 mole % $ZrF_4$. Fuel was added to the **carrier salt** as a 73 mole % LiF - 27 mole % $UF_4$ low-melting, eutectic enriching salt. The composition of the **carrier salt** did not remain constant during the operation of the MSRE because as enriching salt was added to the reactor, LiF was also added to the **carrier salt**. As fuel was burned in the reactor, fission products and plutonium (produced by transmutation of $^{238}U$ in the fuel) were dissolved in the **carrier salt**. Late in the operating campaign, $^{239}PuF_3$ — not produced in the MSRE but manufactured from defense plutonium — was added to the **carrier salt**. Daughter products from radioactive decay of the fuel, fission products, and plutonium also dissolved in the **carrier salt**.

Each time the system was opened for maintenance, the **carrier salt** was drained and the **flush salt** was circulated through the reactor circuit before the **carrier salt** was returned to the reactor. The **flush salt** was initially prepared as 66 mole % LiF and 34 mole % $BeF_2$. It was estimated that about 20 kg of salt remained in the reactor circuit following every **carrier salt** drain; this amount of **carrier salt** was lost to the **flush salt**, and the same amount of **flush salt** was added to the **carrier salt** each time the system was opened. This also changed the composition of the **flush salt**. Different campaigns were conducted in which enriching salt primarily contained $^{233}U$, $^{235}U$, or $^{239}Pu$, respectively. Fluorination removed U nuclides from the **carrier salt**, but an inventory of transmutation and fission products accumulated in the successive campaigns.

$LiF/BeF_2$ mixtures have low melting points and the best neutronic properties of fluoride salt combinations appropriate for reactor use. The liquidus temperature for both the **carrier and flush salt** formulations is approximately 500°C. LiF is a very chemically-stable compound and is only slightly soluble in water (0.29 g/100 ml water). It is most widely used as a flux in the production of ceramics, such as enamels, glasses, and glazes. It is also used in brazing and welding fluxes. The addition of LiF to $BeF_2$ lowers the polymerization of the $BeF_2$ and thus the viscosity of the liquid mixture. $BeF_2$ is also chemically stable. It is very soluble in water but dissolves slowly because the energy of hydration exceeds the lattice energy. All beryllium

compounds are toxic, and $BeF_2$ has a U.S. Occupational Safety & Health Administration (OSHA) permissible exposure limit: time-weighted average of 0.002 mg(Be)/$m^3$. $ZrF_4$, another chemically stable compound, has a solubility of 1.34 g/100 ml in water. It is used in zirconium-based glasses for optical fibers. It was added to the **carrier salt** because zirconium oxides will precipitate before uranium oxides. If the salt became contaminated with water or oxygen, a zirconium oxide precipitate would be visible in the molten salt before precipitation of the uranium fuel occurred.

A container for on-site storage and eventual transport of the MSRE salts has been designed by contractors at Oak Ridge. The design allows the salt container to be shipped inside an RH-TRU waste canister inside a RH-TRU 72-B shipping cask. The transfer of the salt to the containers is currently scheduled for FY12. Based on the volume of the salts, there should be less than 20 containers loaded with salt.

Because the base salt compounds are chemically stable, the primary reaction of concern is the production of fluorine gas by radiolysis. The fluorine gas production rate due to radiolysis has been estimated at 0.02 molecules $F_2$/100 eV absorbed dose (Reference 2). Based on the radionuclide composition of the salts (see Table 3), the **carrier salt** will have the greatest absorbed dose over time. As the fluorine is driven from the salt, the concentration of active metallic sites increases, which increases the rate of metal-to-fluorine recombination (i.e., the back reaction). There is a limit beyond which fluorine is not liberated, called the damage limit, and it occurs after 0.35 mole $F_2$ per kilogram of salt has been driven from the salt. Because of the low decay heat in the salts, the damage limit, for practical purposes, will not be reached. Table 1 shows the calculated $F_2$ generation per kilogram of salt, using the conservative assumption that 100% of the decay energy will be absorbed.

Table 1. Estimated generation of fluorine in the carrier salts

| Year | Generation rate of $F_2$ (mole $F_2$/yr · kg of salt) | Cumulative amount of $F_2$ generated* (mole $F_2$/kg salt) |
|---|---|---|
| 2012 | $7.7 \times 10^{-4}$ | 0.00 |
| 2022 | $6.0 \times 10^{-4}$ | 0.01 |
| 2112 | $1.2 \times 10^{-4}$ | 0.04 |
| 3012 | $2.5 \times 10^{-5}$ | 0.07 |
| 12012 | $1.7 \times 10^{-5}$ | 0.25 |

* Starting $F_2$ accumulation in 2012, which is the planned date for transfer of salt to containers.

At the present time, sodium iodide (NaI) has been selected as the material for sequestering $F_2$ liberated by radiolysis. NaI will be placed on top of the salt monoliths and will react with $F_2$ gas to produce solid sodium fluoride (NaF) and iodine ($I_2$). Under nominal storage conditions, the NaF produced will be stable, and the vapor pressure of the $I_2$ will be very low (about 0.3 mmHg at 25°C), so very little $I_2$ vapor should be present in the headspace. Some of the $F_2$ gas produced will react with the walls of the container before reaching the NaI. If a Hastelloy-N container is used as planned, small quantities of fluorides of both molybdenum and nickel will be produced within the containers. The rate of $F_2$ generation will limit the extent of these reactions and therefore will not impact the integrity of the container during storage, transport, or emplacement time frames.

***The MSRE salts would meet the WIPP waste acceptance criteria regarding chemical and physical properties and material compatibility.***

**PLUTONIUM IN THE MSRE SALTS**

As fuel was burned in the MSRE, Pu was produced by the transmutation of $^{238}U$ and consumed by fission reactions. Additional $^{239}Pu$ not produced in the MSRE was added to the reactor late in the experiment. Because the Pu remains in the **carrier** and **flush salts**, its origin and purpose is described in greater detail in this section.

In November 1964, the MSRE was prepared for operation and loaded with 452 kg of **carrier salt** and 235 kg of LiF – $^{238}UF_4$ eutectic salt (72.3 - 27.7 mole%). In May 1965, a eutectic mixture of LiF – $^{235}UF_4$ ($^{235}U$ was 93% enriched) was added and criticality was achieved in June 1965. Additions of enriched LiF – $^{235}UF_4$ eutectic salt capsules continued throughout the zero power experiments. At the beginning of power operations on January 23, 1966, there was 4498 kg of **carrier salt** containing 4.628% U by weight and an insignificant amount of $^{239}Pu$. The last powered run with $^{235}U$ fuel was completed and the MSRE drained in March 1968. During this period of operation with the $^{235}U$ mixture, $^{239}Pu$ was formed (bred) by neutron capture on $^{238}U$ more rapidly than it underwent fission (burned) so that the amount of Pu in the **carrier salt** rose steadily to a value of 590 grams (Reference 2, Table C.1.)

Over the summer of 1968, the $^{235}U$ /$^{238}U$ mixture was removed from the **carrier salt** by treatment with $F_2$ gas. Essentially all of the 218 kg of the $^{235}U$ /$^{238}U$ fuel mixture was recovered, and no fission products or inbred Pu were removed. A mixture of LiF – $^{233}UF_4$ was added to the **carrier salt**, and the reactor resumed power operations in October 1968. A small amount of $^{238}UF_4$ was included with $^{233}U$ fuel to facilitate uranium isotopic ratio determinations, which also started breeding $^{239}Pu$. However, the $^{239}Pu$ bred during $^{233}U$

operation of the MSRE was minimal compared to the amount that underwent fission, so the total amount of Pu in the **carrier salt** dropped moderately as the MSRE operated.

At the beginning of the last run (run 19; August 1969) of the MSRE, $PuF_3$ was added as an experiment with the following goals (Reference 3):

- Demonstrate the chemical behavior of $PuF_3$ in a molten salt reactor (MSR).
- Demonstrate that $PuF_3$ could be added to the **carrier salt** without degrading reactor operation (to demonstrate that Pu could be a component of future MSR fuel).
- Demonstrate the use of the isotopic composition of plutonium in the MSR as an accurate measure of power output.

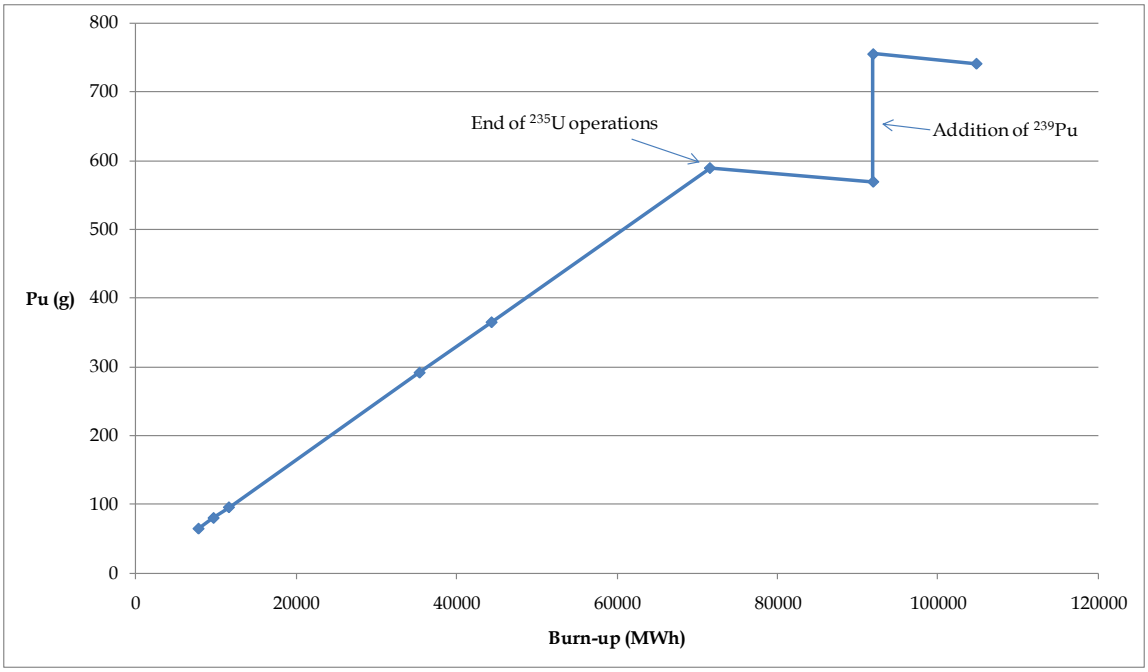
These goals were achieved by the MSRE. The maximum amount of Pu in the **carrier salt** was 756 g, or about 200 ppm, at the beginning of $^{233}U$ operations. Plutonium only accounted for about 4% of the total fissions during the $^{233}U$ operations (Reference 3). The literature shows that Pu was a research tool for proof-of-principle studies and was incidental to the operation of the reactor.

The temporal inventory of Pu as a function of reactor burn-up (Reference 2, Table C.1.) for the MSRE is listed in Table 2 and shown in Figure 4. The rate of production of Pu during $^{235}U$ operations was $8.24 \times 10^{-3}$ g/MWh, and the rate of consumption of Pu during $^{233}U$ operations was $9.85 \times 10^{-4}$ g/MWh.

Table 2. Plutonium inventory as a function of burn-up

| Fuel | Pu addition (g) | Burn-up (MWh) | Total Pu (g) |
|---|---|---|---|
| $^{235}U$ | | 7823 | 65 |
| | | 9700 | 80 |
| | | 11611 | 96 |
| | | 35385 | 292 |
| | | 44362 | 365 |
| | | 71580 | 590 |
| $^{233}U$ | | 91944 | 570 |
| | 185 | 91944 | 755 |
| | | 104816 | 742 |

Figure 4. Plutonium inventory as a function of burn-up




## RADIOLOGICAL PROPERTIES

This section describes the radiological properties of the **carrier** and **flush** salts. These include TRU activity and total activity density. In addition, dose rates, decay heats, and fissile properties are evaluated based on a reasonably-expected packaging configuration.

Activity Density and TRU Activity

The **carrier salt** mass is a combination of salt content from the two fuel drain tanks and the reactive gas removal system, with a reported mass range of 4650-4846 kg (Reference 2, Table 3; Reference 4, Table 2; Reference 5, Table 8) and a density range of 2.47-2.48 g/cm$^3$ (Reference 4, Table 2; Reference 6; Section 2.1). An additional **carrier salt** mass of 226.1 kg is described in the literature (Reference 5, Tables 1 & 2) with "much lower uranium content" and assumed much lower fission-product, actinide, and transactinide (Z>89) content. This additional **carrier salt** is a grouping from six different salts; the largest is 175 kg from the fuel drain tanks.

A reservoir of **flush salt** with a reported mass range of 4265-4290 kg (Reference 6, Section 2.2) and a reported density range of 2.17–2.22 g/cm$^3$ (Reference 4, Table 2; Reference 6, Section 2.2; Reference 7, Table 1) was maintained in a **flush salt** tank to flush the graphite matrix after the **carrier salt** was drained. A small, additional remnant of **flush salt**, with a mass of 20kg, was also identified in the literature (Reference 5, Tables 1 & 2).

Calculations in early 2002 assumed fluorination of the **carrier salt** would be completed in 2002 and would remove gasses trapped in the **carrier salt**, as well as the vast majority of the remaining uranium fuel, leaving less than 10 ppm U in both the **carrier salt** and **flush salt** waste streams. Detailed inventories were reported in the literature for December 2002 and December 2012 (Reference 8, Tables 7 - 19), bounding what was then thought to be the probable disposal time frame.

The fluorine sparging was not performed in 2002 but was actually completed in 2008. Starting with the 2002 curie inventory reported in the literature, we decayed the **flush salt** inventory to 2005 and the **carrier salt** inventory to 2008. Removal of the fuel from the **flush salt** in 2005 and the **carrier salt** in 2008 were then calculated, and the remaining nuclides were decayed to 2012 (probable packaging timeframe). All decay calculations were done using the MICROSHIELD code. These inventories are listed in Tables 3 and 4, sorted by decreasing activity. (The 2002 inventory (Reference 8, Table 5) and subsequent years are found in the appendix.) Each table is sorted by nuclide in the respective salt. The assumption was made that the U concentration was reduced to 10 ppm in each salt (Reference 8, Tables 7 - 19). Attention to the processing history schedule has little effect upon many final observables, but the precise removal time of $^{232}U$ strongly affects the inventory of its daughters, especially $^{208}Tl$.

Table 3. Carrier salt curie inventory after sparging, decayed to 2012

| **Nuclide** | **Activity (Curie)** | **Decay Heat (Watts)** | **$^{239}$Pu FGE** |
|---|---|---|---|
| Sr-90 | 4.78E+03 | 5.54E+00 | 0.00E+00 |
| Y-90 | 4.78E+03 | 2.64E+01 | 0.00E+00 |
| Cs-137 | 4.09E+03 | 4.52E+00 | 0.00E+00 |
| Ba-137m | 3.86E+03 | 1.52E+01 | 0.00E+00 |
| Sm-151 | 1.04E+02 | 1.21E-02 | 0.00E+00 |
| Th-228 + D | 4.78E+01 | 1.39E+00 | 0.00E+00 |
| Pu-239 | 4.09E+01 | 1.27E+00 | 6.50E+02 |
| Pu-241 | 2.07E+01 | 6.59E-04 | 4.48E-01 |
| Pu-240 | 1.57E+01 | 4.88E-01 | 1.53E+00 |
| Th-229 + D | 6.50E+00 | 1.53E-01 | 0.00E+00 |
| Am-241 | 4.74E+00 | 1.58E-01 | 2.55E-02 |
| Pu-238 | 4.16E+00 | 1.38E-01 | 2.72E-02 |
| All other F.P. | 3.17E+00 | 1.51E-02 | 0.00E+00 |
| U-233 | 3.86E-01 | 1.12E-02 | 3.56E+01 |
| U-232 | 1.43E-01 | 4.60E-03 | 0.00E+00 |
| U-234 | 2.23E-02 | 6.41E-04 | 0.00E+00 |
| Pu-242 | 2.42E-03 | 7.12E-05 | 4.57E-03 |
| U-237 | 5.08E-04 | 1.01E-06 | 0.00E+00 |

| | | | |
|---|---|---|---|
| All other isotopes | 4.20E-04 | 1.02E-05 | 1.55E-04 |
| U-236 | 5.47E-06 | 1.49E-07 | 0.00E+00 |
| U-235 | 2.84E-06 | 7.83E-08 | 8.34E-01 |
| U-238 | 9.55E-07 | 2.42E-08 | 0.00E+00 |
| **Total** | **1.78E+04** | **5.53E+01** | **6.88E+02** |

Table 4. Flush salt curie inventory after sparging, decayed to 2012

| **Nuclide** | **Activity (Curie)** | **Decay Heat (Watts)** | **$^{239}$Pu FGE** |
|---|---|---|---|
| Sr-90 | 8.25E+01 | 9.56E-02 | 0.00E+00 |
| Y-90 | 8.25E+01 | 4.56E-01 | 0.00E+00 |
| Cs-137 | 7.04E+01 | 7.80E-02 | 0.00E+00 |
| Ba-137m | 6.67E+01 | 2.63E-01 | 0.00E+00 |
| Sm-151 | 1.80E+00 | 2.09E-04 | 0.00E+00 |
| Pu-239 | 8.00E-01 | 2.48E-02 | 1.27E+01 |
| Pu-241 | 7.92E-01 | 2.52E-05 | 1.71E-02 |
| Th-228 + D | 5.34E-01 | 1.56E-02 | 0.00E+00 |
| Am-241 | 1.75E-01 | 5.84E-03 | 9.41E-04 |
| Pu-240 | 1.48E-01 | 4.60E-03 | 1.45E-02 |
| U-233 | 8.09E-02 | 2.36E-03 | 7.46E+00 |
| All other F.P. | 5.53E-02 | 2.60E-04 | 0.00E+00 |
| Pu-238 | 4.86E-02 | 1.61E-03 | 3.18E-04 |
| U-232 | 3.02E-02 | 9.67E-04 | 0.00E+00 |
| U-234 | 4.75E-03 | 1.37E-04 | 0.00E+00 |
| Th-229 + D | 3.95E-03 | 9.32E-05 | 0.00E+00 |
| Pu-242 | 2.41E-05 | 7.11E-07 | 4.56E-05 |
| U-237 | 1.94E-05 | 3.86E-08 | 0.00E+00 |
| All other isotopes | 1.36E-05 | 6.94E-08 | 1.01E-05 |
| U-235 | 8.02E-06 | 2.21E-07 | 2.35E+00 |
| U-236 | 3.42E-06 | 9.32E-08 | 0.00E+00 |
| U-238 | 2.82E-06 | 7.15E-08 | 0.00E+00 |
| **Total** | **3.07E+02** | **9.49E-01** | **2.25E+01** |

Activities, activity densities, volumes, and radiological properties were calculated based on the values in Table 3 and 4 and the masses and densities reported in the literature and tabulated in Table 5. The smaller additional **carrier salt**, because of its lower U concentration, and the additional **flush salt** are assumed to have the properties of the **flush salt** and are scaled by total mass.

Table 5. Known and resolved properties of MSRE salts decayed to 2012

| | Carrier Salt | Additional Carrier Salt | Flush Salt | Additional Flush Salt |
|---|---|---|---|---|
| **Activity, (Ci)** | | | | |
| Total Activity in item | 1.77E+04 | 1.63E+01 | 3.07E+02 | 1.44E+00 |
| $^{241}$Am | 4.74E+00 | 9.28E-03 | 1.75E-01 | 8.21E-04 |
| $^{237}$Np | 7.37E-06 | 2.54E-08 | 4.79E-07 | 2.25E-09 |
| $^{238}$Pu | 4.16E+00 | 2.58E-03 | 4.86E-02 | 2.28E-04 |
| $^{239}$Pu | 4.09E+01 | 4.24E-02 | 8.00E-01 | 3.75E-03 |
| $^{240}$Pu | 1.57E+01 | 7.85E-03 | 1.48E-01 | 6.94E-04 |
| $^{242}$Pu | 2.42E-03 | 1.28E-06 | 2.41E-05 | 1.13E-07 |
| total TRU Activity | 6.55E+01 | 6.21E-02 | 1.17E+00 | 5.49E-03 |
| **Density (g/cm$^3$)** | | | | |
| Maximum reported | 2.48 | 2.22 | 2.22 | 2.22 |
| Minimum reported | 2.47 | 2.17 | 2.17 | 2.17 |
| **Mass (kg)** | | | | |
| Maximum reported | 4846 | 226.1 | 4274 | 20 |
| Minimum reported | 4650 | 226.1 | 4265 | 20 |
| **Pu (g)** | | | | |
| $^{238}$Pu (17.15Ci/g) | 0.24 | 1.50E-04 | 2.83E-03 | 1.33E-05 |
| $^{239}$Pu (0.06217 Ci/g) | 657.87 | 0.68 | 12.87 | 0.06 |
| $^{240}$Pu (0.2270 Ci/g) | 69.16 | 0.03 | 0.65 | 3.06E-03 |
| $^{241}$Pu (103.2 Ci/g) | 0.20 | 4.07E-04 | 7.67E-03 | 3.60E-05 |
| $^{242}$Pu (0.003945 Ci/g) | 0.61 | 3.24E-04 | 6.11E-03 | 2.86E-05 |
| total Pu | 728.09 | 0.72 | 13.54 | 0.06 |
| **volume (L)** | | | | |
| Maximum | 1962 | 104 | 1970 | 9 |
| Minimum | 1875 | 102 | 1921 | 9 |
| **Activity Density (Ci/L)** | | | | |
| Maximum | 9.44 | 0.16 | 0.16 | 0.16 |
| **Specific TRU Activity (nCi/g)** | | | | |
| Pu Minimum | 12539 | 233 | 233 | 233 |
| Total Minimum | 13517 | 274 | 274 | 274 |

The majority of the radioactivity remaining after more than 40 years of decay consists of the cesium and strontium fission products (and their immediate daughters: Y-90 and Ba-137m), followed by the TRU isotopes typical of neutron capture on the fissile material that comprised the subsequently fluorine-sparged fuel.

It is instructive to compare the concentration of the fission products in the **carrier salt** with the activity concentration limits set by the NRC in its regulations for disposal of LLW, promulgated at 10 CFR Part 61.55. The Class C limits for Sr-90 and Cs-137 found in Table 2 of 10 CFR Part 61.55 are 7 Ci/L and 4.6 Ci/L, respectively. While not subject to NRC regulation, the MSRE **carrier salt** total volume of about 1900 liters — with activity concentrations of Sr-90 and Cs-137 at 2.5 and 2.1 Ci/L, respectively — is well below the Class C activity concentration limit and would otherwise be suitable for disposal as LLW in an NRC-licensed near-surface disposal facility, except for the high Pu concentration.

Similarly, the **flush salt** activity concentrations of Cs-137 and Sr-90 of about 0.04 Ci/L also are well within the Class C limits and would otherwise be suitable for near-surface disposal in an NRC-licensed LLW disposal facility.

Therefore, the activity concentration of the fission products remaining in the MSRE salt (if subject to NRC license requirements) does not demonstrate the need for greater confinement, under 10 CFR Part 61.55. However, the TRU isotopes (primarily Pu-239) clearly exceed the Class C limits, and because the MSRE salt is a defense-related waste, WIPP may be considered a primary disposition option.

***The first result of these calculations is that the Pu-specific activities of all carrier and flush salts exceed 100 nCi/g TRU and the total activity concentration for the salt is less than 23 Ci/L.***

Radiation Dose Rate

**Carrier** and **flush salts** were examined in a configuration for shipment in RH-TRU 72-B shipping casks to determine the magnitude of the dose-equivalent rate (DER) at the surface of an RH-TRU 72-B canister, at the outer surface of an RH-TRU 72-B cask, and at 2 meters from the outer surface of an RH-TRU 72-B cask. The actual configuration may provide more shielding, depending upon the container design, but the default design evaluated here is conservative. The DER is the sum of neutron (n-DER) and gamma (γ-DER) contributions. Shipping requirements for the RH-TRU 72-B include an external-surface DER not exceeding 200 mrem/hr and a DER at 2 meters not exceeding 10 mrem/hr. Each cask was assumed to hold an RH-TRU 72-B canister completely filled with salt and no other materials.

The MICROSHIELD code was used to obtain the γ-DER values given in Table 6. Both **carrier and flush salts** would result in surface dose rates from unshielded packaging greater than 200 mrem/hr and would thus be considered RH-TRU waste, as defined in the LWA.

The γ-DER beyond the shield is dominated by the penetrating 2.61 MeV γ of 3.05-min. $^{208}Tl$ decaying in equilibrium with 1.91-year $^{228}Th$.

Table 6. Calculated dose equivalent rates for 12/2012

| Location | Carrier Salt | | Flush Salt | |
|---|---|---|---|---|
| | γ-DER | n-DER | γ-DER | n-DER |
| RH canister @ surface | 412 rem/h | 13.4 mrem/h | 8.06 rem/h | 0.10 mrem/h |
| RH-TRU 72-B **@ surface** | 5.3 mrem/h | 8.25 mrem/h | 0.05 mrem/h | 0.06 mrem/h |
| RH-TRU 72-B **@2meters** | 0.9 mrem/h | 1.72 mrem/h | 0.01 mrem/h | 0.001 mrem/h |

The neutron sources associated with the MSRE salts are products of spontaneous fission (SF) decays of actinide and transactinide nuclides, (α,n) reactions of decay alpha particles, and (γ,n) reactions from high-energy gamma rays. The photonuclear neutron source was evaluated using a gamma flux obtained with MICROSHIELD and an average 0.6 mb (γ,n) cross section (Reference 9) above the 1.66-MeV threshold of $^{9}Be$, yielding 7.5 n/cm$^2$-s in **carrier salt** and 0.10 n/s-cm$^2$ in **flush salt** with average energy <1 Mev.

The earlier work (Reference 8) used ORIGEN-S values for (α,n) source strengths originating from associated SOURCES code results; we have used SOURCES to evaluate the (α,n) sources at 30.8 n/cm$^2$-s and 0.20 n/cm$^2$-s in the **carrier** and **flush salts**, respectively, with average neutron energy >3 Mev. SOURCES was also used to calculate spontaneous fission neutron production, which is due essentially to $^{240}Pu$ at .04 n/cm$^2$-s and 1E-4 n/cm$^2$-s in the two salts. Total neutron source rates then are 38.3 n/cm$^2$-s in the **carrier salt** and 0.30 n/cm$^2$-s in the **flush salt**.

Conservative estimates of n-DER values were obtained assuming no attenuation of neutrons within the salt, the steel, stainless steel, and lead shielding materials of the RH-TRU 72-B assembly, a subcritical multiplication of 2, and radial emission of all neutrons. A **carrier salt** canister surface n-DER of 8.25 mrem/hour resulted from the combination of the surface flux and a conservative response of $3x10^{-9}$ rem-cm$^2$/n.

***The second result of these calculations shows that the MSRE salt would result in a maximum dose rate at the surface of a canister of less than 1000 rem/h, thereby meeting the LWA limit. The LWA also limits the amount of RH-TRU waste that may exceed 100 rem/hr to <5% of the total RH-TRU waste volume permitted (about 354 cubic meters). The total volume of the carrier salt packaged as RH-TRU waste >100 rem/hr would be less than approximately 2 cubic meters, again well within the limit. In addition, these calculations show that the RH-TRU***

***72-B can safely transport the MSRE salts, as they exist without dilution, with maximum surface DER of approximately 13 mrem/hour and 2-meter DER of approximately 3 mrem/hour.***

Decay Heat of MSRE Carrier and Flush Salts

Activities of major radionuclides have been combined with the specific decay heat (w/g) data from the WIPP Contact-Handled TRU Waste Authorized Methods for Payload Control (CH TRAMPAC) to evaluate the 2012 decay heat properties of the salts. Detailed results are given in watts in Tables 3 and 4 for the **carrier** and **flush salts**. The total salt decay heat from the entire salt volume was found to be less than 56 watts, which is much less than the 300 watt maximum allowed for a single RH-TRU 72-B shipment of solid, inorganic, non-hydrogenous matrix.

***The third result of these calculations shows that the RH-TRU 72-B can safely transport the MSRE salts, as they exist, without exceeding decay heat limits.***

$^{239}$Pu Fissile Gram Equivalence (FGE)

The RH-TRU 72-B cask has an internal waste canister, which has an inner diameter of 25.5 inches and an internal length of 112 inches (approximately 937 L). Bechtel Jacobs has designed a container for molding, on-site storage, and eventual transport of the MSRE salts that will fit inside of an RH-TRU 72-B waste canister. To bound the amount of material that could be loaded in a RH-TRU 72-B canister, it will be assumed that the volume of loaded salt is 937 L, which is the entire usable volume of the RH-TRU 72-B canister.

The salts, if placed within an RH-TRU 72-B canister, must not exceed 305 $^{239}$Pu FGE because the salt has greater than 1% by wt chemically-bound Be (RH TRAMPAC 2006). Activities of the major radionuclides in MSRE **carrier** and **flush salts** have been combined with the $^{239}$Pu FGE data from the WIPP CH TRAMPAC to determine the $^{239}$Pu FGE properties of the salts.

The entire **carrier salt** was found to have a 688 $^{239}$Pu FGE and the **flush salt** a 22.6 $^{239}$Pu FGE; these $^{239}$Pu FGE values are essentially unchanged in the 10-year decay period. Using the average values for the volumes of the salts (to avoid compounding conservative assumptions), the **carrier salt** has a 0.359 $^{239}$Pu FGE/L and the **flush salt** a 0.0116 $^{239}$Pu FGE/L. An RH-TRU 72-B canister completely filled with **carrier salt** would thus have a 336 $^{239}$Pu FGE, and a similar canister of **flush salt** would have a 10.9 $^{239}$Pu FGE.

A full canister of **carrier salt** would exceed the 305 $^{239}$Pu FGE limit. Thus, the amount of **carrier salt** packed in an RH-TRU 72-B canister would need to be limited to 92% of the available volume. This option is easily manageable, both technically and operationally.

***The fourth result of these calculations shows that the RH-TRU 72-B can safely transport the MSRE salts, as they exist, without exceeding $^{239}$Pu FGE limits.***

**CONCLUSION**

The defense pedigree of the MSRE salt is clear. The MSRE generator site documents state that the MSRE was operated to demonstrate that this type of reactor could breed weapons-usable plutonium for defense programs. Also, weapons grade $^{239}PuF_3$, manufactured from defense plutonium at Los Alamos National Laboratory and shipped to ORNL, was added to the MSRE carrier salt. The comingling with defense plutonium or the direct mission documents alone could provide justification for the defense pedigree necessary to dispose of the MSRE carrier and flush salts at WIPP.

Additionally, the MSRE salt is eligible for disposal at WIPP since it is neither spent nuclear fuel nor HLW, which are prohibited at WIPP under the LWA. Spent nuclear fuel is defined in the Nuclear Waste Policy Act (NWPA) as fuel that has been withdrawn from a nuclear reactor following irradiation, the constituent elements of which have not been separated by reprocessing. HLW is defined in the NWPA as the highly-radioactive material resulting from the reprocessing of spent nuclear fuel, including liquid waste produced directly in reprocessing and any solid material derived from such liquid waste that contains fission products in sufficient concentrations.

The MSRE flush salt is neither HLW nor spent nuclear fuel as it was simply circulated through the reactor after the carrier salt was removed. The carrier salt also does not fit the NWPA definitions, which reference a self-contained fuel that can be mechanically withdrawn from a reactor, such as in a more traditional PWR. The MSRE was designed such that the fuel was dissolved in the carrier salt; the carrier salt alone did not serve as fuel. Additionally, fluorine sparging was used to extract the uranium fuel, which was converted to uranium oxide for disposal, as opposed to extracting the uranium for reprocessing. This method of removing the fuel left the plutonium and remaining fission products in the carrier salt, with the intention to dispose of this salt as is.

In fact, the carrier salt was chosen for its neutronic, thermal, and chemical properties, which allowed it to serve the same purpose as the coolant water in a PWR. The coolant water in a PWR is not considered HLW or spent nuclear fuel, even when contaminated with fission

products and plutonium. In a PWR, the primary coolant water travels from the reactor to a steam generator, where water in a secondary coolant loop is heated. In the MSRE, the carrier salt traveled from the reactor to a heat exchanger, where salt in a secondary loop was heated. Also, both the primary coolant water in a PWR and the MSRE carrier salt serve a moderating function in the reactor.

Finally, the activity concentrations of residual fission products in the salt are below the Class C limits for disposal of LLW, thereby demonstrating that the salt contains activity levels less than those otherwise associated with HLW. Therefore, the MSRE salt is neither HLW nor spent nuclear fuel and is not prohibited from disposal at WIPP.

Also, based on the calculations presented in this paper, the MSRE salt would meet the TRU activity level (>100 nCi/g TRU), surface dose rate (<1000 rem/hr after packaging), and total activity concentration (<23 Ci/L) requirements of the LWA. In addition, there are no radiological properties that would prohibit transportation in WIPP-approved shipping containers (NRC licensed). The salt can be packaged in such a way as not to exceed any transportation or emplacement limits. There are also no chemical or physical incompatibilities with the waste acceptance criteria for WIPP. Therefore, the MSRE salt can reasonably be expected to meet all of the WIPP waste acceptance criteria and be certified to meet the requirements of the WIPP waste analysis plan.

It is concluded that the MSRE salt is defense TRU waste and when packaged to meet all applicable criteria, will be eligible for disposal at WIPP.

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# Appendix
## Carrier and Flush Salt Isotopic Composition

| Nuclide | Carrier Salt | | | | Flush Salt | | | |
|---|---|---|---|---|---|---|---|---|
| | Activity 2002 (Ci) | Activity 2012 (Ci) | Decay Heat 2012 (W) | Pu239 FGE 2012 | Activity 2002 (Ci) | Activity 2012 (Ci) | Decay Heat 2012 (W) | Pu239 FGE 2012 |
| Ac-225 | 7.69E-01 | 8.12E-01 | 2.78E-02 | 0 | 5.49E-03 | 4.94E-04 | 1.70E-05 | 0 |
| Am-241 | 4.39E+00 | 4.74E+00 | 1.58E-01 | 2.55E-02 | 1.61E-01 | 1.75E-01 | 5.84E-03 | 9.41E-04 |
| At-217 | 7.69E-01 | 8.12E-01 | 3.40E-02 | 0 | 5.49E-03 | 4.94E-04 | 2.07E-05 | 0 |
| Ba-137m | 4.85E+03 | 3.86E+03 | 1.52E+01 | 0 | 8.39E+01 | 6.67E+01 | 2.63E-01 | 0 |
| Bi-210 | 1.00E-05 | 2.00E-05 | 4.60E-08 | 0 | 6.48E-08 | 1.12E-07 | 2.59E-10 | 0 |
| Bi-212 | 5.47E+01 | 6.83E+00 | 1.12E-01 | 0 | 8.00E-01 | 7.63E-02 | 1.26E-03 | 0 |
| Bi-213 | 7.69E-01 | 8.12E-01 | 3.21E-03 | 0 | 5.49E-03 | 4.94E-04 | 1.96E-06 | 0 |
| Bi-214 | 3.65E-05 | 5.70E-05 | 9.37E-07 | 0 | 2.39E-07 | 2.47E-07 | 4.06E-09 | 0 |
| Cs-137 | 5.14E+03 | 4.09E+03 | 4.52E+00 | 0 | 8.87E+01 | 7.04E+01 | 7.80E-02 | 0 |
| Eu-152 | 9.71E-01 | 5.83E-01 | 4.42E-03 | 0 | 1.68E-02 | 1.01E-02 | 7.66E-05 | 0 |
| Eu-154 | 2.42E+00 | 1.10E+00 | 9.84E-03 | 0 | 4.18E-02 | 1.90E-02 | 1.70E-04 | 0 |
| Eu-155 | 2.78E+00 | 6.87E-01 | 5.00E-04 | 0 | 4.82E-02 | 1.19E-02 | 8.68E-06 | 0 |
| Fr-221 | 7.69E-01 | 8.12E-01 | 3.08E-02 | 0 | 5.49E-03 | 4.94E-04 | 1.87E-05 | 0 |
| H-3 | 6.20E+02 | 0 | 0 | 0 | 1.07E+01 | 0 | 0 | 0 |
| I-129 | 2.74E-03 | 0 | 0 | 0 | 4.75E-05 | 0 | 0 | 0 |
| Nb-93m | 2.26E-01 | 6.04E-02 | 1.07E-05 | 0 | 3.85E-03 | 1.57E-03 | 2.78E-07 | 0 |
| Np-237 | 3.07E-05 | 7.37E-06 | 2.16E-07 | 1.55E-04 | 1.10E-06 | 4.79E-07 | 1.40E-08 | 1.01E-05 |
| Pa-233 | 3.07E-05 | 7.21E-06 | 1.70E-08 | 0 | 6.60E-05 | 4.73E-07 | 1.11E-09 | 0 |
| Pb-209 | 7.69E-01 | 8.12E-01 | 9.51E-04 | 0 | 5.49E-03 | 4.94E-04 | 5.79E-07 | 0 |
| Pb-210 | 1.00E-05 | 2.00E-05 | 5.07E-09 | 0 | 6.48E-08 | 1.12E-07 | 2.85E-11 | 0 |
| Pb-212 | 5.47E+01 | 6.83E+00 | 1.30E-02 | 0 | 8.00E-01 | 7.63E-02 | 1.45E-04 | 0 |
| Pb-214 | 3.65E-05 | 5.70E-05 | 2.59E-07 | 0 | 2.39E-07 | 2.47E-07 | 1.12E-09 | 0 |
| Pd-107 | 2.86E-03 | 2.86E-03 | 1.57E-06 | 0 | 4.92E-05 | 4.92E-05 | 2.71E-08 | 0 |
| Pm-147 | 5.97E+00 | 4.24E-01 | 1.55E-04 | 0 | 1.03E-01 | 7.33E-03 | 2.69E-06 | 0 |
| Po-210 | 9.44E-06 | 1.92E-05 | 6.13E-07 | 0 | 6.15E-08 | 1.10E-07 | 3.51E-09 | 0 |
| Po-212 | 3.49E+01 | 4.37E+00 | 2.28E-01 | 0 | 5.12E-01 | 4.88E-02 | 2.55E-03 | 0 |
| Po-213 | 7.52E-01 | 7.94E-01 | 3.95E-02 | 0 | 5.37E-03 | 4.82E-04 | 2.40E-05 | 0 |
| Po-214 | 3.65E-05 | 5.70E-05 | 2.59E-06 | 0 | 2.39E-07 | 2.47E-07 | 1.12E-08 | 0 |
| Po-216 | 5.47E+01 | 6.83E+00 | 2.75E-01 | 0 | 8.00E-01 | 7.63E-02 | 3.07E-03 | 0 |
| Po-218 | 3.65E-05 | 5.70E-05 | 2.03E-06 | 0 | 2.39E-07 | 2.47E-07 | 8.80E-09 | 0 |
| Pu-238 | 4.51E+00 | 4.16E+00 | 1.38E-01 | 2.72E-02 | 5.27E-02 | 4.86E-02 | 1.61E-03 | 3.18E-04 |
| Pu-239 | 4.09E+01 | 4.09E+01 | 1.27E+00 | 6.50E+02 | 8.00E-01 | 8.00E-01 | 2.48E-02 | 1.27E+01 |
| Pu-240 | 1.57E+01 | 1.57E+01 | 4.88E-01 | 1.53E+00 | 1.48E-01 | 1.48E-01 | 4.60E-03 | 1.45E-02 |
| Pu-241 | 3.36E+01 | 2.07E+01 | 6.59E-04 | 4.48E-01 | 1.28E+00 | 7.92E-01 | 2.52E-05 | 1.71E-02 |
| Pu-242 | 2.42E-03 | 2.42E-03 | 7.12E-05 | 4.57E-03 | 2.41E-05 | 2.41E-05 | 7.11E-07 | 4.56E-05 |
| Ra-224 | 5.47E+01 | 6.83E+00 | 2.31E-01 | 0 | 8.00E-01 | 7.63E-02 | 2.58E-03 | 0 |
| Ra-225 | 7.69E-01 | 8.12E-01 | 5.76E-04 | 0 | 5.49E-03 | 4.94E-04 | 3.50E-07 | 0 |
| Ra-226 | 3.65E-05 | 5.70E-05 | 1.64E-06 | 0 | 2.39E-07 | 2.47E-07 | 7.12E-09 | 0 |
| Rn-220 | 5.47E+01 | 6.83E+00 | 2.55E-01 | 0 | 8.00E-01 | 7.63E-02 | 2.85E-03 | 0 |
| Rn-222 | 3.65E-05 | 5.70E-05 | 1.85E-06 | 0 | 2.39E-07 | 2.47E-07 | 8.04E-09 | 0 |
| Sb-125 | 1.46E-01 | 1.20E-02 | 3.76E-05 | 0 | 2.53E-03 | 2.06E-04 | 6.48E-07 | 0 |
| Sm-151 | 1.12E+02 | 1.04E+02 | 1.21E-02 | 0 | 1.94E+00 | 1.80E+00 | 2.09E-04 | 0 |
| Sr-90 | 6.08E+03 | 4.78E+03 | 5.54E+00 | 0 | 1.05E+02 | 8.25E+01 | 9.56E-02 | 0 |

| Nuclide | Carrier Salt | | | | Flush Salt | | | |
|---|---|---|---|---|---|---|---|---|
| | Activity 2002 (Ci) | Activity 2012 (Ci) | Decay Heat 2012 (W) | Pu239 FGE 2012 | Activity 2002 (Ci) | Activity 2012 (Ci) | Decay Heat 2012 (W) | Pu239 FGE 2012 |
| Tc-99 | 5.01E-01 | 0 | 0 | 0 | 8.68E-03 | 0 | 0 | 0 |
| Te-125m | 3.57E-02 | 2.94E-03 | 2.56E-06 | 0 | 6.17E-04 | 5.08E-05 | 4.43E-08 | 0 |
| Th-228 | 5.43E+01 | 6.79E+00 | 2.22E-01 | 0 | 7.98E-01 | 7.61E-02 | 2.49E-03 | 0 |
| Th-229 | 7.69E-01 | 8.12E-01 | 1.60E-02 | 0 | 3.39E-05 | 4.94E-04 | 9.74E-06 | 0 |
| Th-231 | 5.95E-04 | 2.84E-06 | 3.43E-09 | 0 | 1.87E-04 | 8.02E-06 | 9.69E-09 | 0 |
| Th-234 | 2.15E-04 | 9.55E-07 | 1.42E-10 | 0 | 6.60E-05 | 2.82E-06 | 4.19E-10 | 0 |
| Tl-208 | 1.96E+01 | 2.46E+00 | 5.77E-02 | 0 | 2.88E-01 | 2.74E-02 | 6.44E-04 | 0 |
| Tl-209 | 1.61E-02 | 1.75E-02 | 3.62E-04 | 0 | 1.15E-04 | 1.07E-05 | 2.20E-07 | 0 |
| U-232 | 3.55E+01 | 1.43E-01 | 4.60E-03 | 0 | 7.76E-01 | 3.02E-02 | 9.67E-04 | 0 |
| U-233 | 8.69E+01 | 3.86E-01 | 1.12E-02 | 3.56E+01 | 1.89E+00 | 8.09E-02 | 2.36E-03 | 7.46E+00 |
| U-234 | 5.01E+00 | 2.23E-02 | 6.41E-04 | 0 | 1.11E-01 | 4.75E-03 | 1.37E-04 | 0 |
| U-235 | 5.95E-04 | 2.84E-06 | 7.83E-08 | 8.34E-01 | 1.87E-04 | 8.02E-06 | 2.21E-07 | 2.35E+00 |
| U-236 | 7.31E-04 | 5.47E-06 | 1.49E-07 | 0 | 7.92E-05 | 3.42E-06 | 9.32E-08 | 0 |
| U-237 | 8.06E-04 | 5.08E-04 | 1.01E-06 | 0 | 3.07E-05 | 1.94E-05 | 3.86E-08 | 0 |
| U-238 | 2.15E-04 | 9.55E-07 | 2.42E-08 | 0 | 6.60E-05 | 2.82E-06 | 7.15E-08 | 0 |
| Y-90 | 6.08E+03 | 4.78E+03 | 2.64E+01 | 0 | 1.05E+02 | 8.25E+01 | 4.56E-01 | 0 |
| Zr-93 | 2.99E-01 | 2.99E-01 | 8.69E-05 | 0 | 5.10E-03 | 5.10E-03 | 1.48E-06 | 0 |
| Total | 2.35E+04 | 1.78E+04 | 5.53E+01 | 6.88E+02 | 4.06E+02 | 3.07E+02 | 9.49E-01 | 2.25E+01 |