

# ENVIRONMENTAL MANAGEMENT

## **Engineering Evaluation of the Proposed Alternative Salt Transfer Method for the Molten Salt Reactor Experiment Oak Ridge National Laboratory Oak Ridge, Tennessee**



BECHTEL JACOBS COMPANY LLC  
OAK RIDGE ENVIRONMENTAL  
MANAGEMENT CLEANUP CONTRACT  
WITH THE UNITED STATES  
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**Engineering Evaluation of the Proposed  
Alternative Salt Transfer Method for the  
Molten Salt Reactor Experiment  
Oak Ridge National Laboratory  
Oak Ridge, Tennessee**

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Prepared by

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Determination***

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## ACRONYMS

BJC	Bechtel Jacobs Company LLC
DOE	U.S. Department of Energy
EDS	Energy Dispersive X-ray Spectroscopy
EPA	U.S. Environmental Protection Agency
FDT	Fuel Drain Tank
FFT	Fuel Flush Tank
FTIR	Fourier Transform InfraRed
MSRE	Molten Salt Reactor Experiment
ORNL	Oak Ridge National Laboratory
Pro2Serve	Professional Project Services, Inc.
RGRS	Reactive Gas Removal System
WIPP	Waste Isolation Pilot Plant



## EXECUTIVE SUMMARY

This evaluation was performed by Pro2Serve in accordance with the *Technical Specification for an Engineering Evaluation of the Proposed Alternative Salt Transfer Method for the Molten Salt Reactor Experiment at the Oak Ridge National Laboratory* (BJC 2009b). The evaluators reviewed the *Engineering Evaluation Work Plan for Molten Salt Reactor Experiment Residual Salt Removal, Oak Ridge National Laboratory, Oak Ridge, Tennessee* (DOE 2008).

The Work Plan (DOE 2008) involves installing a salt transfer probe and new drain line into the Fuel Drain Tanks and Fuel Flush Tank and connecting them to the new salt transfer line at the drain tank cell shield. The probe is to be inserted through the tank ball valve and the molten salt to the bottom of the tank. The tank would then be pressurized through the Reactive Gas Removal System to force the salt into the salt canisters.

The Evaluation Team reviewed the work plan, interviewed site personnel, reviewed numerous documents on the Molten Salt Reactor (Sects. 7 and 8), and inspected the probes planned to be used for the transfer.

**Based on several concerns identified during this review, the team recommends not proceeding with the salt transfer via the proposed alternate salt transfer method. The major concerns identified during this evaluation are:**

- **Structural integrity of the tanks.** The main concern is with the corrosion that occurred during the fluorination phase of the uranium removal process. This may also apply to the salt transfer line for the Fuel Flush Tank. *Corrosion Associated with Fluorination in the Oak Ridge National Laboratory Fluoride Volatility Process* (Litman 1961) shows that this problem is significant.
- **Continued generation of Fluorine.** Although the generation of Fluorine will be at a lower rate than experienced before the uranium removal, it will continue to be generated. This needs to be taken into consideration regardless of what actions are taken with the salt.
- **More than one phase of material.** There are likely multiple phases of material in the salt (metal or compound), either suspended through the salt matrix, layered in the bottom of the tank, or both. These phases may contribute to plugging during any planned transfer. There is not enough data to know for sure.
- **Probe heat trace.** The alternate transfer method does not include heat tracing of the bottom of the probe. There is a concern that this may cool the salt and other phases of materials present enough to block the flow of salt.
- **Stress-corrosion cracking.** Additionally, there is a concern regarding moisture that may have been introduced into the tanks. Due to time constraints, this concern was not validated. However, if moisture was introduced into the tanks and not removed during heating the tanks before HF and F<sub>2</sub> sparging, there would be an additional concern regarding the potential for stress-corrosion cracking of the tank walls.



# 1. INTRODUCTION

The purpose of this evaluation was to evaluate the proposed alternative salt transfer method for the Molten Salt Reactor Experiment (MSRE) per the *Technical Specification for an Engineering Evaluation of the Proposed Alternate Salt Transfer Method for the MSRE at the Oak Ridge National Laboratory (ORNL)* (BJC 2009b) and determine the feasibility of the proposed transfer method. The evaluation was done by Professional Project Services, Inc. (Pro2Serve) for Bechtel Jacobs Company LLC.

The alternate salt transfer method consists of heating the tanks to 600°C, then inserting a cold probe into the open tank, connecting a line from the probe to the existing transfer line, sealing the tank, and pressurizing to 45 psig to force the molten salt out of the tanks through the probe to the transfer lines and then to the staged salt canisters.

A team consisting of two Chemical Engineers (L. Little, P.E., and S. Brady, P.E.), a Metallurgical Engineer (T. Kollie, PhD.), a Mechanical Engineer (K. Roberts, P.E.), and a Team Leader (J. Carlberg) was selected for this evaluation. The evaluation consisted of a review of technical literature provided by Bechtel Jacobs Company LLC (BJC), along with other sources of pertinent information. A list of documents reviewed is contained in Sects. 7 and 8 of this report. Additionally, interviews were conducted with key BJC personnel. A list of personnel interviewed is contained in Appendix A. The Evaluation Team also visited the site and inspected the probes that were constructed for the transfer, toured the High Bay Area, reviewed the Shift Logs from 2004 to 2008, and reviewed videos taken of the Fuel Drain Tank (FDT) and Fuel Flush Tanks (FFT). Some of the specific areas of concern addressed in this evaluation report are:

- The age of the salt and impurities present in the salt that could affect the transfer.
- The age of the tanks and piping and their structural integrity.
- Hastelloy-N embrittlement by Tellurium exposure.
- Hastelloy-N radiation damage.
- Other degradation and corrosion of the Hastelloy-N alloy that may have occurred during operations and subsequent cold storage.
- Evaluation of the blockages associated with the previous failed salt transfers.





## 2. BACKGROUND

The MSRE was an 8 MW reactor that was operated at ORNL from 1965 through 1969 as a demonstration of the technology needed to develop a large-scale Molten Salt Breeder Reactor. The reactor used a unique liquid fuel, formed by dissolving  $\text{UF}_4$  fuel in a carrier salt composed of a mixture of  $\text{LiF}$ ,  $\text{BeF}_2$ , and  $\text{ZrF}_4$ . The fuel salt circulated through a reactor vessel, a fuel salt pump, and a primary heat exchanger at temperatures above  $600\text{ }^\circ\text{C}$  ( $1112\text{ }^\circ\text{F}$ ). In the reactor, the salt was forced through channels of graphite to provide the geometry and moderation necessary for a nuclear chain reaction. Heat was transferred from the fuel salt to the secondary coolant salt in the primary heat exchanger. The coolant salt was similar to the fuel salt, except that it contained only  $\text{LiF}$  (66%) and  $\text{BeF}_2$  (34%). The coolant salt passed from the primary heat exchanger to an air-cooled radiator, a coolant salt pump, and then returned to the primary heat exchanger. Each of the salt loops was provided with drain tanks, located such that the salt could be drained out of either circuit by gravity. The fuel salt drain tanks were provided with a system to remove the intense heat generated by radioactive decay immediately after an emergency reactor shutdown and fuel salt drain. A third drain tank connected to the fuel salt loop was provided for storing a batch of flush salt. This batch of salt, similar in composition to the coolant salt, was used to condition the fuel salt loop after it had been exposed to air and to flush the fuel salt loop of residual fuel salt and contaminants before accessing the reactor circuit for maintenance or experimental activities. All three tanks are located in the drain tank cell, a stainless-steel-lined below-grade cell that could be covered by two layers of concrete shield blocks (see Fig. 1).

The initial plan was to transfer the salt to salt canisters immediately after the removal of the uranium from the tanks via the existing drain line. However, after the removal of the uranium from the FFT, an unsuccessful attempt was made to transfer the salt. It was thought at the time that the transfer was prevented due to blockage in the transfer line that was caused by a mixture of material that would not melt at the melting temperature of the salt. This transfer method was then abandoned and it was decided that the transfer of the salt would be postponed until the uranium was removed from all three tanks. As a result of the concern for the blockage, the Alternative Transfer Method was developed and addressed in DOE 2008.

During the uranium removal operations in 2005–2008, a salt plug was discovered in the piping that did not melt when it was heated to the design process temperature, and a probe used in aged irradiated fission-product-laden salt also plugged unexpectedly with a solid material. That probe plug was found to contain nickel, which may have migrated from the Hastelloy-N components such as tank walls, piping, and fittings. Nickel was present in new salt and in salt after initial criticality, but in lesser concentrations than iron. Several reports and worker witnesses refer to a black tarry substance present in and largely segregated from the general form of salt, possibly in the highest concentration near the bottom of tanks, which has not been identified and thus has unknown melting point(s) and may be involved in the plugging incidents historically experienced. The precise physicochemical causes and mechanisms involved in these plugging events are not known and may never be determined without extensive and expensive sampling and characterization, and possibly experimentation. After the plugging events, the decades-old design involving heating to the process design temperature and transferring liquid molten salt through a probe and small piping has remained the baseline plan for salt removal. The possible existence of materials other than salt in the tanks calls into question whether salt removal may leave some substantial fraction of the fission products behind in a higher-melting-point residue, which seems to reduce or defeat the decommissioning value of salt removal. Due to logistical and equipment problems, the design has

been modified to use a probe that is to be inserted from above into a tank at the process design temperature and that is open to the atmosphere at the time of insertion.

The arrangement of the principal components of the MSRE are shown in Fig. 1.

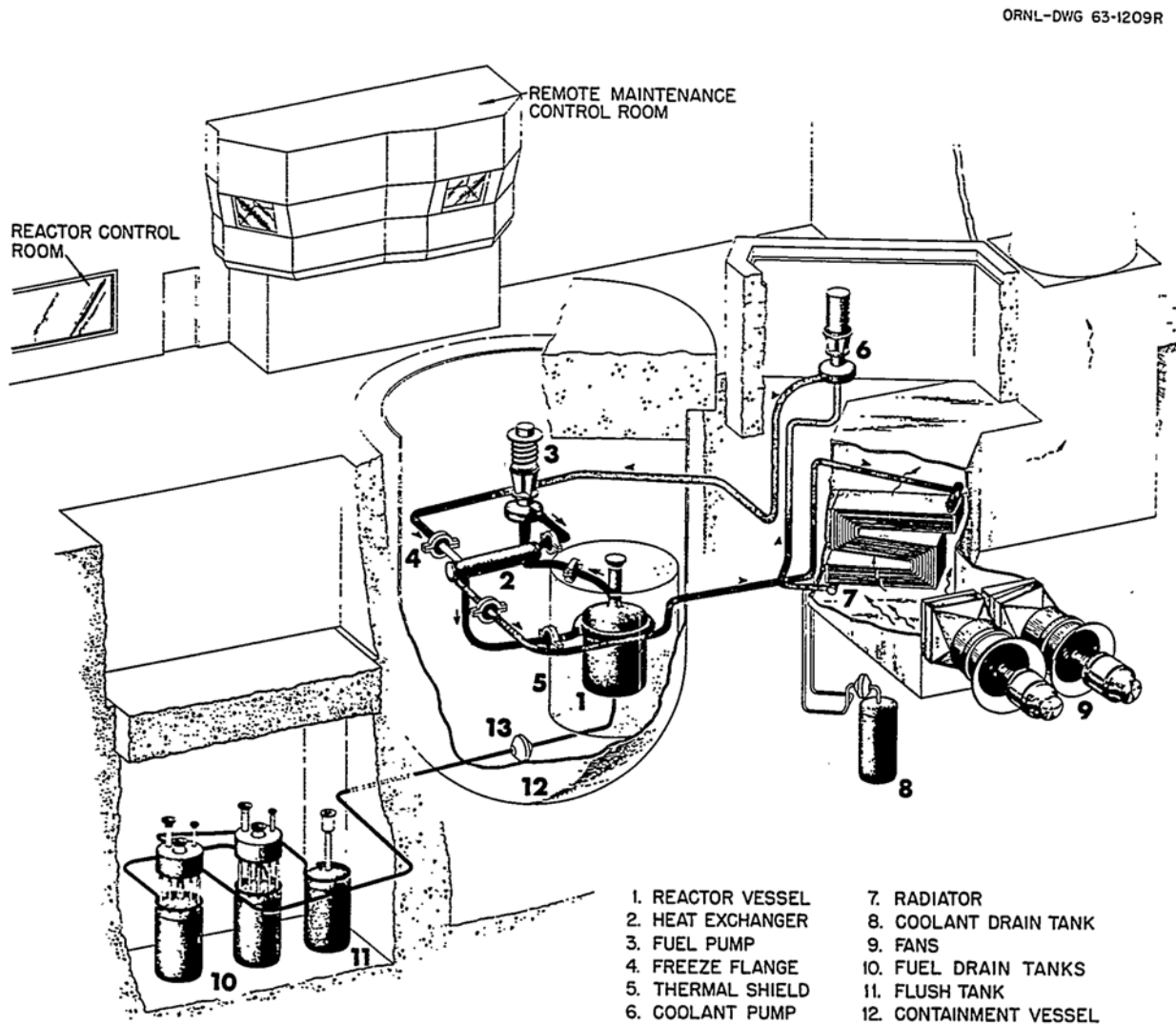


Figure 1. Arrangement of the principal components of the MSRE.

### **3. METALLURGICAL EVALUATION**

#### **3.1 BACKGROUND**

The technical specifications for this project (BJC 2009b) concerning the metallurgy of the Hastelloy-N drain, flush tanks, and piping of the MSRE require Pro2Serve to address:

1. The age of tanks and piping,
2. Hastelloy-N embrittlement by tellurium exposure,
3. Hastelloy-N radiation damage, and
4. Other degradation and corrosion of the Hastelloy-N alloy which may have occurred during operations and subsequent cold storage.

The purpose of this section is to summarize information from the literature on these subjects. Because the probe to be used in the transfer of the salt is new (Muggridge 2009b), the above-mentioned concerns are not relevant to it.

##### **3.1.1 History of the Drain, Flush Tanks, and Piping**

In order to determine the present metallurgical conditions of the drain, flush tanks, and piping, one must know their time-temperature exposure as well as their atmospheric exposures and contents during their lifetime.

###### **3.1.1.1 During reactor operation, 1965–1969**

Figure 3 of BJC 1998 displays the time-temperature histogram of the drain tanks between January 1965 and December 1989. This figure shows 27 heats between about 480°C and about 700°C between January 1965 and December 1969. The reason for the first three heats is not explained, but probably they were associated with checkouts of the drain circuit prior to the reactor going critical. The last 24 heats were due to drains of the fuel from the reactor into the tanks. The reports state that a total of 31,000 h (about 3.5 years) were logged when the system (drain tanks, reactor, and drain system piping) was exposed to fuel salt at temperatures of 480°C or more. Of this time, a total of 12,650 h (about 1.4 years) were accumulated while the reactor was operating.

The paragraph below is a pertinent excerpt from BJC 1998:

“During reactor operation, the salt circuits were pressurized with inert gas, either helium or argon, to protect the Hastelloy-N ... surfaces from corrosion while the salt circulated from the reactor to the heat exchanger ... When the reactor was returned to standby, the fuel salt was routed to the drain tanks by gravity flow and the salt was essentially static in the drain tanks at nearly constant temperatures with the same protective cover gas ... The drain piping was always full and tanks were either full or partially full until fuel salt was transferred to the reactor fuel loop. Depending on operational needs, the piping was maintained at temperatures between 480 and 590°C.”

### 3.1.1.2 During and after anneal cycles, 1970–1998

After reactor shutdown in December 1969 and during the 1970 to 1998 time period covered by BJC 1998, the salt was frozen with an inert cover gas and was kept at ambient temperature except for 20 “annealing cycles.” The annealing cycles were intended to recombine the fluorine that was liberated from the salt by radiolysis and were performed each year from 1970 to 1989. These cycles involved heating the tanks to 150 to 260°C for one to two weeks. Thus, the salt was below its solidus temperature and thus solid during these annealing cycles. The annealing cycles were stopped in 1989 because uranium was found to have migrated to the charcoal bed. At this time, the tanks were isolated from the auxiliary charcoal bed with a blanket gas of helium at 18 psia.

### 3.1.1.3 During the period 1998–2009

The time-temperature history of the tanks from 1998 to the present has not been documented (Muggridge 2009b). Between October 2004 and March 2008, the salt was melted to remove the uranium from the salt and during attempts to transfer the salt from the tanks (Fuel Flush Tank, October 2004 to July 2005; Fuel Drain Tank-2, February 2006 to November 2006; Fuel Drain Tank-1, January 2008 to March 2008 ) (Carlberg 2009a). The actual time-temperature history of the tanks between 2001 and present is stored on computers (Muggridge 2009a.). Unfortunately, the constraints of this project did not permit their retrieval in a timeframe to be useful to this evaluation. Table 1 shows the approximate time-temperature history during HF and F<sub>2</sub> processing and attempts to transfer salt from the tanks. It is thought that the tanks were at ambient temperature from 1998 to 2001.

**Table 1. Approximate time-temperature history of tanks during HF and F<sub>2</sub> processing and attempts to transfer salt from the tanks (Carlberg 2009b)<sup>a</sup>**

Facility	Time Hot	Time HF Processing	Time F <sub>2</sub> Processing
FFT	8 months	28 days	1 day
FDT-2	11 months	48 days	4 days
FDT-1	2 months	32 days	7 days

<sup>a</sup>This information is approximations that were gathered from handwritten logs, and draft data downloaded from Lookout (the monitoring system used during processing).

## 3.1.2 Differences in Exposures of Hastelloy-N of Reactor Vessel and of Tanks and Piping

### 3.1.2.1 Temperature history

From the previous section, the major differences in the temperature exposure of the Hastelloy-N in the reactor vessel and the tanks and piping are the low temperature annealing cycles when the salt was solid, and during the operations to remove the uranium and to remove the salt from the tanks when the salt was liquid.

Reactions between the Hastelloy-N and solid salt would be predicted to be minor, as mentioned in several of the reports provided, for example Notz 1985. Thus, the major concern during the time period 1970 to when the salts were first melted in 2004 is the low-temperature exposure of Hastelloy-N to fluorine and other gases, which will be discussed later.

The approximate temperature-time history of the tanks when the salt was liquid between 2004 and 2008 is listed in Table 1. The major concern during this time period is the exposure of the Hastelloy-N to high

temperatures in the presence of HF and F<sub>2</sub>, possibly with water vapor contamination present. This subject will also be discussed later.

### **3.1.2.2 Neutron fluxes and neutron energies**

Major and obvious differences between the exposure of the Hastelloy-N in the reactor vessel and in the tanks and piping were the energies and magnitudes of the neutron fluxes. In the reactor, the neutrons were moderated by the graphite to reduce their energies to “thermal” levels. In the tanks, only the salt itself was a moderator and the thermalization of the neutrons was minimal, according to Hollenbach and Hopper (Hollenbach 1994). Also, the neutron flux is obviously significantly less in the tanks than in the reactor because of the neutron chain reaction providing fission of the uranium in the reactor. In the tanks, neutrons were produced by spontaneous fission and by (alpha, neutron) ( $\alpha$ , n) reactions on <sup>9</sup>Be, <sup>19</sup>F, and <sup>7</sup>Li; the  $\alpha$ -particles come from the  $\alpha$ -decay of fission products (Notz 1985). Hollenbach and Hopper (Hollenbach 1994) note that very little fission was produced by fast neutrons, those with energies above thermal, in the reactor or the tanks.

Thus, in order to evaluate neutron effects in the tanks, neutron fluxes over the lifetime of the tanks, called the neutron fluences, would need to be calculated as there are no neutron flux measurements available for the tank facility (Muggridge 2009b). Certainly, the energy spectrums of the neutrons in the reactor and tanks are quite different; and, therefore, the interactions with materials would be different. In particular, the effects of fast neutrons on the Hastelloy-N of the tanks and piping would have to be estimated.

## **3.2 AGE OF TANKS AND PIPING**

To evaluate the effect of age on the tanks and piping, the environment to which they have been exposed over their lifetime must be considered. Obviously, the environment of these components is quite different on their insides and outsides. The outsides will be considered first, and the many effects that contribute to the metallurgical condition of the insides of the tanks and piping will be considered next.

### **3.2.1 Outside**

Hastelloy-N has excellent corrosion resistance when exposed to an oxidizing environment, forming a protective oxide layer that reaches essentially a constant thickness with time at a given temperature. The outsides of the tanks and piping were exposed to an oxidizing environment over their lifetime. In their postmortem examination of various parts of the reactor’s fuel circuit, McCoy and McNabb state (McCoy 1972), “Oxide films were formed on all the INOR-8 (an early designation for Hastelloy-N) surfaces exposed to air or the cell environment ...The oxide generally consisted of a uniform layer about 1 mil thick and a selective oxidation front extending to a depth of 4 or 5 mils.” As outlined above, the time-temperature history of the reactor and tanks is quite comparable except for the additional time the tanks were heated to remove the uranium and to attempt to remove the salt. Except for the FFT, this additional time is less than 10% of the 3.5 year exposure in the 1960s; thus, it would be reasonable to assume that an oxide film of about 6 mils exists on the outside of the drain tanks. The FFT outside film may be a mil or so thicker due to its additional nine month exposure.

### 3.2.2 Inside

#### 3.2.2.1 Hastelloy-N embrittlement by tellurium exposure

Tellurium (Te) is a fission product produced in the MSRE, as discussed in many of the documents provided. Tellurium has more than 30 isotopes, only a few of which are not radioactive (Lide 1990), so most of the isotopes of Te produced by fission in the reactor will decay away, some quite rapidly.

When exposed to Te in the MSRE, Hastelloy-N becomes brittle, as is widely reported in most of the references provided. McCoy1978 postulates that the embrittlement is probably due to formation of a nickel-tellurium intermetallic compound in the grain boundaries of the alloy. One remark by McCoy in this report (McCoy 1978) is interesting: “Experience with the MSRE indicated that Te was deposited on the metal and graphite surfaces and that only a small amount resided in the salt at any one time.” This assertion raises the question as to how much Te in the salt in the reactor is transferred to the tanks. The following paragraphs explore this question further.

Kedl<sup>1</sup> describes Te as a “noble metal” because it does not form a stable fluoride in the environment of the MSRE. He states that Te and the other noble metals are present in the salt in the metallic state. Because Te melts at 450°C, it would be molten in the reactor and in the tanks during reactor operation. According to Kedl 1972, the noble metals deposit on the Hastelloy-N reactor vessel and the graphite moderator, and migrate to gas-liquid interfaces and adhere to them, thereby being removed from the salt. During operation of the MSRE, Te is being produced constantly by the fissioning of uranium within the reactor. On the other hand, a negligible amount of Te would be produced in the tanks due to the very low level of decay by spontaneous fission. Thus, only the Te present in the salts at the time of reactor shutdown that is drained down to the drain tanks from the reactor need be considered. Once some of this Te has quickly decayed, only a negligible amount would be left or created.

Grimes (Grimes1967) states that the noble metals “are found in the salt at considerably less than the expected concentrations. If calculations of the amounts expected are based upon concentrations of Sr<sup>91</sup> ...about 30% of the Te<sup>132</sup> appears in the melt (salt). It is not yet possible to state with certainty whether these materials are present in the salt as colloidal metal (or alloy) particles or as soluble chemical species, though present evidence suggests the former is more likely.”

Lindauer (Lindauer 1969) notes, “Te exists primarily in the metallic state during reactor operation and, as such, is removed from the salt by deposition and by the off-gas stream. Freeze valve samples showed less than 1% Te remaining in the salt.”

The above discussion leads to the conclusion that Te was present in the tanks at significantly lower levels than in the reactor, maybe 100 times less in the drain tanks, and at even lower levels in the flush tank. The flush tank contains a much smaller amount of all fission products than the drain tanks (BJC 1998). Thus, the Te embrittlement of the Hastelloy-N in the tanks is much less than in the reactor, especially in the flush tank. Lindauer states that the fluorination process will remove all Te from the salt due to its conversion to the volatile TeF<sub>6</sub>, so Te embrittlement would not occur after the first fluorination of the tanks to remove the uranium.

#### 3.2.2.2 Hastelloy-N radiation damage

McCoy1972 states that helium can be produced in Hastelloy-N by two mechanisms: the transmutation by thermal neutrons of tramp B<sup>10</sup> in the alloy and a two-step transmutation by thermal neutrons of Ni<sup>58</sup>, first to Ni<sup>59</sup> then to Fe<sup>56</sup> and He. Helium from both sources collects at the grain boundaries of the Hastelloy-N

and causes reductions in mechanical properties at elevated temperatures, principally in the rupture life and fracture strain.

In the reactor, the neutrons are thermal. Unfortunately, the neutron fluence and energy spectrum of the neutrons in the tanks and pipes are not known. Fast neutrons in the tanks and pipes can cause radiation damage by other mechanisms to produce embrittlement of the alloy as follows (Cunningham 1969, Angeliu 2006):

1. Atomic displacement damage occurs whereby the atoms of a material are dislodged from their lattice positions by fast neutrons; fortunately, this damage reaches a saturation level at low fluences.
2. Fast neutron-radiation-induced solute segregation (precipitation of alloy constituents) can contribute to embrittlement of nickel-based alloys.
3. Fast neutrons can transmute Fe, Cr, and Ni<sup>58</sup> in the Hastelloy-N to He, causing reductions in the mechanical properties as discussed previously.
4. Fast neutrons can cause void formation in the alloy, producing swelling; but this is said to be small in Ni-based alloys.

### **3.3 OTHER DEGRADATION AND CORROSION OF HASTELLOY-N**

The degradation and corrosion effects in the walls and on the inside surfaces of the Hastelloy-N tanks and piping that were not covered during the discussion of Te embrittlement and radiation damage include: aging, selective removal of chromium from the Hastelloy-N by the salt, fluorine, and HF corrosion.

#### **3.3.1 Aging of the Hastelloy-N**

Aging is a phenomenon that occurs in an alloy whereby its properties are modified by heat treating it at an elevated temperature for a period of time. Normally, elements or phases precipitate out of solid solution in the alloy during the heat treatment, thereby altering its microstructure and properties. As mentioned above, radiation also can induce such solute segregation. For Hastelloy-N, McCoy (McCoy 1970) showed that aging Hastelloy-N at 650°C for 20,789 h in a neutron flux significantly reduced the fracture strain. For tests at 25°C, the ductility could be restored by heat treating the alloy at 870°C for 8 h, indicating to him that the loss in fracture strain was caused by precipitation of carbides. For tests conducted at 500°C and 650°C, McCoy said, “the properties of the unirradiated material changes some with aging at 650°C, and we did not have sufficient data to separate the contribution of irradiation and aging to the observed embrittlement.” Because the properties of interest in the transfer of the salt with the probe are at the elevated temperatures noted above, the loss in ductility due to aging should be a minor effect compared to the other property changes discussed herein.

#### **3.3.2 Chromium Removal from Hastelloy-N**

Koger and Grimes (Koger 1972 and Grimes 1967) and others describe the corrosion (oxidation) of the Hastelloy-N of the reactor vessel by the salt, which selectively removed Cr from the hotter reactor region and subsequently deposited it in the cooler heat exchanger region as the salt was pumped through it to remove the heat from the reactor. This mass transfer phenomenon can be explained as follows:

Corrosion starts at the hottest point in the reactor, and the Cr concentration in the salt increases there. When the concentration of Cr in the salt exceeds the “equilibrium” amount at the coldest point in the

loop, the Cr will begin to deposit there. At equilibrium, the corrosion of the Cr at the hottest point will continue at a rate equal to that of the deposition at the coldest point. The rate-limiting step of the corrosion at the hottest point is the diffusion of the Cr from the interior of the Hastelloy-N to its surface in contact with the salt. Diffusion is a relatively slow process at the temperatures of the MSRE.

Grimes (Grimes 1970) states that many samples taken at regular intervals during reactor operation showed Cr concentrations in the salt rose from an initial value of ~ 40 ppm to a final value of ~ 85 ppm. On the other hand, he notes that Fe (120 ppm) and Ni (50 ppm) were present at relatively high levels but showed no perceptible increase with time. The concentration of the other Hastelloy-N component, Mo, was below the detectable limit, ~ 25 ppm. (Note: Mo is a “noble” metal in salt as it does not form a stable fluoride. Some Mo may be removed from the salt by the same mechanisms as described for Te removal.)

In the tanks, the salt is essentially isothermal and static, with only the relatively small temperature gradients in the tanks causing circulation of the salt. Under this condition, one would expect the Cr concentration in the salt to reach an equilibrium value and the removal of Cr from the Hastelloy-N in the tanks to cease. (Without a significant temperature difference, Cr will not deposit and reduce the Cr concentration in the salt.) Once the uranium was removed from the salt, the selective corrosion of Cr from the Hastelloy-N would cease, according to many of the referenced documents. The flush tank would be expected to have little of this type of corrosion because it contained mostly flush salt.

Because the salt had been circulating in the reactor for an extended time before transfer to the tanks, the salt should have had near its equilibrium concentration of Cr when it was drained into the tanks, and little Cr would be removed from the tanks. Conversely, during the early tests of the drain circuit before reactor criticality and any time the tanks were used to melt new salt, Cr leaching from the Hastelloy-N of the tanks would have occurred. Little corrosion would be expected during the annealing cycles due to the low temperatures involved.

### **3.3.3 HF and F<sub>2</sub> Corrosion – General Remarks**

From 1970 to 1989, fluorine gas was released in the drain tanks due to radiolysis of the salt. The drain tanks were heated (annealed) every year to recombine the fluorine and salt. During this period, it was estimated in the general reference (BJC 1998) for this part that dry fluorine corroded the Hastelloy-N to a depth of 1 mil. Between 1989 and 1998, annealing was stopped but the corrosion of the walls also was estimated to be 1 mil additional (BJC 1998).

Because the conditions in the drain tank were about the same from 1989 to 1998 as from 1998 until processing of the salt to remove uranium, it is estimated that the corrosion during the latter period was about 1 mil also.

According to the general reference for this part, the fission product concentrations in the flush tank were very low; thus, radiolysis of the salt in the flush tank is thought to be significantly less in the flush tanks than in the drain tanks. Fluorine corrosion was probably not measurable in the flush tanks until processing to remove the uranium and transfer the salt was undertaken.

Literature searches were performed using the online data bases at ORNL to determine the corrosion rates of Hastelloy-N by HF and F<sub>2</sub> at elevated temperatures. To date, such data could not be found for Hastelloy-N using these data bases. Limited data for the corrosion rates on some nickel-based alloys were found, and these data can be used to estimate corrosion rates for Hastelloy-N, but such estimates include substantial uncertainty as they are not in a MSRE-type salt environment and are not for Hastelloy-N.



Late in this project, the Evaluation Team learned that water vapor may have been present during the HF and F<sub>2</sub> processing. Data were not found for the corrosion of nickel-based alloys by wet F<sub>2</sub>, but limited data were found for HF plus steam corrosion for a couple of nickel-based alloys. Steve Pawel of ORNL (Pawel 2009a) said that “Adding moisture to HF can complicate many things ... the corrosion rate is likely to be wickedly high for a lot of engineering alloys in wet HF at 600°C.” Pawel also stated, “I have never performed testing in wet F<sub>2</sub>, but I’m under the impression wet F<sub>2</sub> is similar to wet HF at high temperature” (Pawel 2009b). The possibility of having moisture in the tanks during HF and F<sub>2</sub> processing adds a high degree of uncertainty to the estimates of the corrosion of the Hastelloy-N during these processes. It is not known, however, whether the moisture that leaked in prior to heating was retained during heating of the tanks.

Late in this review, a reference (Litman 1961) was discovered in a Google search of “fluorine sparging” that contains corrosion data for INOR-8 (the experimental version of Hastelloy-N) during F<sub>2</sub> sparging of MSRE-type salts. Due to time and funding constraints, cross-references contained in this report were not pursued to obtain more of similar references. These new data also will be discussed in Sect. 3.3.3.1.

### **3.3.3.1 Corrosion by F<sub>2</sub>**

In hot F<sub>2</sub>, the reaction rate between Ni and F<sub>2</sub> is parabolic, so the rate decreases with time (CEB 1968). Thus, short-time corrosion tests will give significantly higher corrosion rates than longer term tests. For example, corrosion data in F<sub>2</sub> for Nickel-200 Alloy, a 99.5% commercially-pure Ni alloy, yields a rate at 538°C of 24.5 mils/year for a 5-h test, which drops to 13.8 mils/year for a 120-h test. Also, as predicted theoretically, the corrosion rates are very temperature sensitive. At 370°C, the above rates drop to 1.7 mils/year for a 5-h test and 0.4 mils/year for a 120-h test.

Nickel forms a dense nickel fluoride reaction product in hot F<sub>2</sub>, which is protective to the Ni substrate up to reasonably high temperatures. Only when the vapor pressure of the fluoride becomes high enough does the fluoride film begin to sublime, and the film is no longer protective.

As mentioned previously, data are not available for Hastelloy-N (70% Ni, 17% Mo, 7% Cr) corrosion by hot F<sub>2</sub>. Data for Inconel-600 (76% Ni, 15.5% Cr, 8% Fe) are available but only for 5-h tests. Using the Inconel-600 data to estimate the corrosion rate of Hastelloy-N is suspect because Inconel-600 does not have Mo as a major constituent and has a higher concentration of Cr. Also, the time the Hastelloy-N tanks were exposed to F<sub>2</sub> was of the order of days, not 5 h, and the testing on the Inconel-600 was not performed in a MSRE-type salt environment.

At 370°C, the corrosion rate for Inconel-600 is 78 mils/year and increases to 3450 mils/year at 538°C. Using the Arrhenius rate equation (Sinnott 1958), the corrosion rate for Hastelloy-N at 450 to 475°C (462°C was used) is estimated to be 850 mils/year or 2.3 mils/day, which is a quite high rate. It was noted (Muggridge 2009b), however, that the Fourier Transform InfraRed (FTIR) Spectrometer data did show large amounts of MoF<sub>6</sub> when uranium was being removed from the salt, so these high corrosion rates could be in the ballpark. Additionally, Kedl (Kedl 1972) noted that during processing to change the uranium isotope in the fuel using other facilities than the tanks in question, corrosion of the process tank was estimated to be 2.4 mils/day in the presence of the salt. He states, “The tank was constructed of Hastelloy-N and the corrosion products were NiF<sub>2</sub>, FeF<sub>2</sub>, and CrF<sub>2</sub>. Following the fluorination step, it was therefore necessary to remove the corrosion products from the salt.”

Thus, it is obvious that the fluorination step not only badly corroded the Hastelloy-N process tank, but it also contaminated the salt. Table 2 is adapted from Table 11 of Lindauer’s report (Lindauer 1969). This table shows a factor of two to three increases in the concentrations of Cr and Fe in the salt and over a factor of 20 increases in the concentration of Ni. Of course, the Ni concentration in Hastelloy-N is a least

a factor of ten greater than the Cr and Fe, so these large differences are roughly in agreement with their concentrations in the alloy. Molybdenum, the major alloying element in the nickel-based Hastelloy-N alloy, is not seen in the salt as its fluoride,  $\text{MoF}_6$ , is removed as a gas with the  $\text{UF}_6$  during fluorination.

**Table 2. Concentrations of Hastelloy-N components in salt before and after fluorination to remove uranium from the salt (adapted from Lindauer 1969)**

	Cr (ppm)	Fe (ppm)	Ni (ppm)
Before Fluorination	170	131	36
After Fluorination	420	400	840

The data in the report (Litman 1961) found are for corrosion during  $\text{F}_2$  sparging in INOR-8 (the experimental version of Hastelloy-N) vessels. These sparging tests were performed for 50 hours at 450 and 600°C in salts with chemistries somewhat different than that of the actual MSRE salt, and only one of the four salts tested had  $\text{UF}_4$  in it. The authors found that the  $\text{UF}_4$  in the salt increased the corrosion rate of the INOR-8 by a factor of at least 3.2 under otherwise identical conditions; the presence of uranium is known to significantly increase Cr corrosion by the salt as mentioned in a number of the references used for this report. The 150-degree temperature difference between the tests increased the corrosion rate of the INOR-8 by a factor of at least 7.5 under otherwise identical conditions; such large temperature effects would be expected from the Arrhenius-rate theory (Sinnott 1958).

The most important finding of this report (Litman 1961) is that the corrosion was not uniform over the surface of the vessel. The authors describe four regions of corrosion of the INOR-8 vessel: the region in contact with the salt, the region near the vapor-salt-interface, the “middle-vapor” region just above the salt level, and the “upper-vapor” region well above the salt level.

Two of the four salt solutions tested had the same chemistries; one was tested at 450°C and the other at 600°C. A third salt solution had the same chemistry as the aforementioned two except for the addition of  $\text{UF}_4$  and was tested at 450°C. In the tests without the  $\text{UF}_4$ , the vapor-salt interface region had the highest corrosion rate (0.9 mils/day at 450°C) as compared to that of the other three regions, which had identical corrosion rates at 450°C (0.4 mils/day). When the  $\text{UF}_4$  was added, the middle-vapor region had the highest corrosion rate (2.9 mils/day at 450°C), the upper-vapor and vapor-salt-interface regions had identical corrosion rates (2.4 mils/day at 450°C), and the salt region had the lowest corrosion rate (1.7 mils/day at 450°C).

For the tests with the salt containing  $\text{UF}_4$ , photomicrographs showed that the surface at the vapor-salt interface region was very undulated, but the other three areas seemed to be as smooth as the INOR-8 surface before the tests. The photomicrographs of the salt region of the fourth corrosion test, which was conducted at 600°C in a salt having a different chemistry than the other three and did not contain  $\text{UF}_4$ , shows a very irregular surface in the salt region, which could be interpreted as pitting corrosion.

The corrosion rates quoted above from Litman (Litman 1961) for the  $\text{UF}_4$ -containing salt at 450°C are the best estimates for the corrosion in the MSRE drain tanks. On the other hand, the corrosion rates measured at 450°C without the  $\text{UF}_4$  in the salt are the best estimates for the MSRE flush tank. To compare these rates with those estimated from the Inconel-600 corrosion rates at 450 to 475°C (462°C), the Arrhenius rate-equation was used to obtain a multiplication factor of 1.6 for the 12-degree increase in temperature. For the flush tanks, the calculated corrosion rates near the vapor-salt interface were 1.4 mils/day and for the other three regions it was 0.64 mils/day. For the drain tanks, the calculated corrosion rate in the middle-vapor region was 4.6 mils/day, in the upper-vapor and vapor-salt-interface regions it was

3.8 mils/day, and in the salt region it was 2.7 mils/day. These corrosion rates span the estimates obtained from the Inconel-600 data of 2.3 mils/day in  $F_2$  gas and the 2.4 mil/day corrosion rate Kedl (Kedl 1972) obtained during  $F_2$  sparging to change the uranium isotope in the fuel during operation of the MSRE.

Table 1 shows that the tanks were exposed to  $F_2$  for periods of 1 day to 7 days. Using the above estimated corrosion rates of Litman (Litman 1961), removal of 1.4 mils is estimated for the FFT near the vapor-salt interface, and 0.6 mils in the other three regions; 18 mils for the middle-vapor region, 15 mils for the upper-vapor and vapor-salt-interface regions, and 11 mils for the salt region of FDT-2; and 32 mils for the middle-vapor region, 27 mils for the upper-vapor and vapor-salt-interface regions, and 16 mils for the salt region for FDT-1. The effect of water vapor or an oxidizing atmosphere during  $F_2$  processing of the salt will not be considered as data do not exist for this type of atmosphere for nickel-based alloys; however, corrosion would be expected to be increased drastically for a water-vapor- $F_2$  mixture. In addition, stress-corrosion cracking could occur in a water-vapor- $F_2$  mixture (Pawel 2009b). Also, the corrosion within the piping has not been estimated due to time constraints and the uncertainty as to its overall environment.

### 3.3.3.2 Corrosion by HF

Hastelloy-N is much more resistant to corrosion in HF than in  $F_2$ . Unfortunately, corrosion data in HF for Hastelloy-N (70% Ni, 17% Mo, 7% Cr) could not be found; it was located, however, for Hastelloy-C (59% Ni, 16% Mo, 15.5% Cr, 5.5% Fe, and 4% W) (CEB 1968). These two alloys are similar but the “C” alloy is lower in Ni and has more Cr than the “N” alloy.

The corrosion rate of Hastelloy-C in HF was measured at 0.3 mils/year at 550°C in a 36-h test. This test temperature is about 75°C higher than the HF processing temperature. Using this value and the times given in Table 1 for the processing in HF, the corrosion of Hastelloy-N would be estimated to be insignificant in HF. The only question about this estimate is how the HF corrodes the Hastelloy-N in the presence of salt. Because of time limitations and the lack of information, this question was not pursued.

If water vapor was in the tank during the HF processing, the corrosion rates would have been much higher. Because of time limitations and the difficulty of manipulating FTIR information in its existing computer formats, this question was not pursued.

## 3.4 CONCLUSIONS AS TO METALLURGICAL CONDITIONS OF TANKS AND PIPING

The tanks and piping are **not** reactors! Understandably, the focus of most reports and data acquisition during MSRE operation was on the reactor itself and not the tanks. Lessons learned about the metallurgy of the Hastelloy-N in the reactor can only be applied to the tanks and piping on a mechanism-by-mechanism basis after careful evaluation. There are many unknowns concerning the variables affecting these mechanisms in the tanks and piping and the salt therein. Neutron fluxes in the tanks and pipes are assumed to be significantly below that of the reactor. This is probably a good assumption, but neither measurements nor calculations of the neutron fluxes in the tanks have been made to support it. It stands to reason, however, that the neutron fluxes in the tanks are far below that of the reactor, as sustained fission reactions did not occur in the tanks. Nuclear fission was insignificant in the tanks and piping because the neutrons in the tanks and piping were not thermal neutrons and fast fission did not occur, so the only fission mechanism was through spontaneous nuclear decay. With very little fission in the tanks and piping, fission products such as Te would not be produced in more than negligible quantities in the tanks and piping. Also, very small amounts of Te were transported from the reactor to the tanks and pipes when the tanks were filled because the vast majority of the Te was either deposited in the reactor or lost in the off-gas, so Te embrittlement of the tanks and pipes would be extremely low. Low neutron fluxes

would also create extremely small amounts of helium in the Hastelloy-N, so this form of radiation damage would be far less than that occurring in the reactor. Other fast neutron radiation damage of the Hastelloy-N should also be minimal; i.e., atomic displacement, solute segregation, and void formation.

Because the tanks and piping were held at operating temperatures for times comparable with the reactor, the aging effects in the Hastelloy-N tanks and pipes should be about the same as the reactor. The degradation of the mechanical properties of these components due to aging alone would be acceptable insofar as structural integrity for heating, pressurizing, lifting, etc.

The reduction in mechanical properties due to Cr corrosion would be very low in the drain tanks, and because the salt in the flush tank did not contain an appreciable amount of uranium, Cr corrosion should be negligible in the flush tank.

Fluorine corrosion of the walls of the drain tanks and piping between 1970 and the removal of the uranium between 2005 to 2008 should be about 3 mils, which is acceptably small. Because the flush tank contained only small amounts of fission products, radiolysis of the salt to produce fluorine should be minimal. Consequently, fluorine corrosion in the flush tank should be negligible.

Before the F<sub>2</sub> sparging to remove the uranium from the salt, it appears from the foregoing discussion that the tanks and piping would probably have been suited for the heating, pressurizing, lifting, etc., prior to the operations of the past. The tanks were subjected to an engineering analysis and nondestructive testing in 2000 that concluded: “The results of the examination indicated that the mechanical integrity of the three drain tanks is more than adequate for the intended FSD process operations. There is enough margin in the tank construction to qualify the tanks for pressures higher than 80 psia at 600°C (the design process operating conditions are 65 psig and 600°C)” (BJC 2008). This engineering analysis and nondestructive testing were performed before the F<sub>2</sub> sparging to remove the uranium from the salt.

The corrosion that occurred during the processing to remove the uranium was significant and was the dominant degrading metallurgical effect uncovered in this study. For example, it is estimated that the FDT-1 suffered the most corrosion by the F<sub>2</sub> gas sparging, namely an estimated loss of up to 32 mils. It is predicted that FDT-2 suffered up to 18 mils loss by corrosion, whereas the FFT lost up to 1.4 mils by corrosion in the fluorination process. In a worst case scenario, pitting corrosion, which possibly occurred in one of the high temperature tests of Litman (Litman 1961), and stress-corrosion cracking, which could occur if water vapor were present during HF and F<sub>2</sub> gas sparging, would significantly increase the damage reported herein. After fluorination, the two drain tanks are not likely to be suitable to withstand the higher operating pressures and temperatures of the salt transfer process because of the degradation of their mechanical integrity by F<sub>2</sub> corrosion, but the FFT would probably be suitable.

In conclusion, the corrosion and structural degradation caused by hot fluorination operations would by far dominate the other property-degrading mechanisms discussed, such as Te embrittlement or radiation damage, so further investigation of these and other metallurgical mechanisms is not warranted.

### **3.5 RELATIONSHIP – METALLURGICAL DEGRADATION OF HASTELLOY-N AND BLOCKAGE DURING SALT TRANSFER**

The mechanism of degradation of the Hastelloy-N tanks and piping discussed above are in two categories: those that occur internally in the walls of the Hastelloy-N components and those that happen at the surfaces of the components. The internally occurring mechanisms are Te embrittlement, radiation damage due to helium production at the grain boundaries, aging, and the fast neutron radiation damage effects of atomic displacement, solute segregation, and void formation. With the exception of void

formation, which is judged to be minor, none of these mechanisms result in a measurable increase in the dimensions of the component, particularly of the piping. Thus, *per se*, these damages could not cause a blockage during salt transfer.

Protective oxides form on the outside surfaces of the components; and, of course, they would not contribute to the blockages during salt transfer. Two mechanisms that occur on the inside surfaces of the components are salt and fluorine corrosion. Both of these mechanisms remove material from the surfaces of the components. Thus, the inside diameter of the piping would be increased by these mechanisms and would not contribute, *per se*, to the blockage during salt transfer.

On the other hand, the corrosion of the tanks, piping, and reactor vessel do add impurities to the salt, particularly Ni, Fe, and Cr. In addition, nuclear fission in the reactor added many fission product elements to the salt. While this subject is best handled in sections of this report dealing with impurities in the salt, the following is offered for completion of this section.

Some of these impurity elements may form many different phases with each other in the salt. These phases could be solids at the molten salt temperatures. If they are much denser than the salt, these solid phases could fall to the bottom of the tank. If density and buoyancy permit, these solid phases could be distributed through the salt. As the salt is pushed through the piping during salt transfer, these solid phases could agglomerate in the small-diameter piping, causing a blockage. In the previous attempts of salt transfer, the drain tube was at the bottom of the tanks. Thus, any solid phases that had settled to the bottom of the tanks would have been pushed into this drain tube early in the transfer and could have caused a blockage by agglomerating.

### 3.6 RECOMMENDATIONS

The major uncertainty in the evaluation of the metallurgical state of the tanks and piping is the degradation that occurred during processing to remove the uranium. The corrosion due to fluorine was judged to be significant to the point that the mechanical integrity of the tanks could be compromised under the process conditions during salt transfer. Therefore, it is recommended that salt transfer not be attempted unless an engineering analysis and nondestructive testing, similar to that performed in 2000 (BJC 1998) but with some added features, has been performed on the tanks in their present condition, particularly on the drain tanks.

In addition, if a part of the drain tanks that has been exposed to the salt, fluorine, and HF could be removed and evaluated extensively, then the metallurgical condition of the tanks could be determined. In the calculations of corrosion, it was shown from the work of Litman (Litman 1961) that the corrosion was not uniform across the surface of the Hastelloy-N tanks. If pitting corrosion or stress-corrosion cracking occurred, the mechanical integrity of the tank could easily be compromised. Thus, this recommended metallurgical examination should be designed so that it determines whether pitting and stress-corrosion cracking corrosion occurred. The previous engineering analysis and nondestructive testing probably would not have detected pitting corrosion or stress-corrosion cracking.

If the decision is made to attempt the transfer of salt without the above two recommendations being accomplished with acceptable results, which is not advised, it is recommended that the tanks be heated to temperature slowly, with frequent pressure tests on the way up to operating temperature. These pressure tests should be conducted at a constant temperature, and the pressure should be increased slowly. During pressurizing, the pressure increase should be stopped and the pressure in the tank monitored to see if the tank is leaking. Thus, pinhole size leaks in the walls of the tanks or piping could be demonstrated. Also,

it is recommended that this pressure testing regimen be performed even if the above two recommendations are performed with successful results.

The piping in the new probe to be used to transfer the salt has not been used. Thus, it is obviously clear of any blockages. Every effort should be made to insure that the existing piping to be used in the transfer of salt is clear of any blockage.

## 4. CHEMICAL EVALUATION

The chemical aspects associated with the MSRE fuel salt and flush salt transfer from the fuel drain cell to the ultimate disposal container have been briefly reviewed. Creation of solids and corrosion of containment vessels have been the immediate focus of this work. Both the MSRE operating period, 1965–1969, and the storage within the FDT-1, FDT-2, and FFT are discussed in this section. A considerable volume of information has been accumulated on the operating period, and the information has been studied in support of other reactor programs and has been extremely helpful in the current review activities.

### 4.1 SALT CHEMISTRY CONSIDERATIONS

The original fuel salt charged to the system contained metal fluorides with the ability to withstand the requirement of the reactor and the compatibility with the structural components. The flush salt included the major fluoride compounds without the uranium or the zirconium. The actual fuel composition was dependent upon the amount of uranium required to bring the system to the critical, and then to the operating, condition. Fissionable uranium comprised about one-third of the uranium inventory; the balance, as nonfissionable  $U^{238}$ , was included for chemical purposes. Zirconium was a constituent of the fuel mixture to prevent the precipitation of  $UO_2$  and resultant criticality hazards in the event that oxide contamination of the fuel occurred. At a concentration of 5 mole %  $ZrF_4$  in the fuel, significant and recognizable quantities of  $ZrO_2$  would be preferentially precipitated prior to loss of any uranium from the fuel solution as  $UO_2$ . The table below provides the basic composition of the fuel and flush materials in terms of mole percentages.

**Table 3. Basic composition of the fuel and flush materials (mole percentages)**

	Fuel Salt		Flush Salt	
	Estimate 12/2002 <sup>a</sup>	Initial Charge <sup>b</sup>	Estimate 12/2002 <sup>a</sup>	Initial Charge <sup>b</sup>
LiF	64.59	64.7	65.88	63.6
BeF <sub>2</sub>	30.46	30.1	33.94	36.4
ZrF <sub>4</sub>	4.90	5.2	0.179	
UF <sub>4</sub>	0.0331		0.00166	
PuF <sub>3</sub>	2.51E-03		4.43E-05	
Other Fission Products	0.0187		3.06E-04	

<sup>a</sup>Huxtable 2002a

<sup>b</sup>Thoma 1971

Today the fuel and flush salts are mixtures of LiF, BeF<sub>2</sub>, ZrF<sub>4</sub>, UF<sub>4</sub>, and small quantities of fluorides of corrosion products, fission products, actinides, and transuranics. During the MSRE operational period various materials were added to the salt to control the reactor activity and the behavior of the salt. The fission products can be roughly divided into three different material: volatile materials that exited the system (e.g., Xe, Kr), fluoride-forming elements that remain within the circulating salt (e.g., Rb, Cs, Sr, Ba, Y, Zr, and lanthanides), and noble metals that do not form fluorides that are plated out on the equipment or remained suspended in the salt matrix (e.g., Mo, Ru, Te, Tc, Ag, Sb, and sometimes Nb). The volatile materials are not of current concern since they have exited the process during the operating period.

Removal of the salt from the tanks demands consideration of the potential for solids deposited within the tanks and the corrosion of the tanks during the processing history. Histories of the operating period

suggest the formation of solids, the plating of noble metal fission products, and the formation of coating on sample elements, all indicating that the salt contained solid-forming compounds. Radiolytic reduction of the salt and the generation of fluorine were recognized during the post-operating period and initiated the practice of creating conditions to recombine the fluorine on a regular frequency. Corrosion concerns have centered on the impact of chromium and tellurium.

There are combinations of events that have suggested solids formation within the system either as a dispersed material with the capability to settle out or the creation of a separate phase. There have been reports of lubricant leakage into the salt pump and creating sludge materials (p. 33, Thoma 1971). A discussion on tellurium (Compere 1975) suggests the presence of a separate phase circulating within the salt during the reactor operations:

“The viewpoint that emerges with respect to noble metal behavior in salt is that what we see is due to the appearance of highly dispersed but undissolved material in the salt, a mobile separate phase, presumably solid, which bears much higher noble-metal fission product concentrations than the salt.

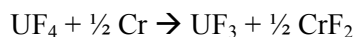
The nature and amount of the mobile phase are not established with certainty, but several possibilities exist, including (1) graphite particles, (2) tars from decomposed lubricating oil from the pump shaft, (3) insoluble colloidal structural metal in the salt, (4) agglomerates of fission products on pump bowl surface and/or bubbles, (5) spalled fragments of fission product deposits on graphite or metal. As we shall later see, at least some of the material deposits on surfaces, and it are also indicated that some is associated with the gas-liquid interface in the pump bowl.”

Should the dispersed phase have a density greater than the salt, there is the potential for the material to accumulate in the bottom of the drain tanks once the system is drained to FDT-1 and FDT-2 but before the salt is solidified. During investigation of the vessel integrity, there was indication from the radiation probe within the thimbles that the radiation level was substantially higher in the lower portion of the tank. Also it was reported during interviews with MSRE operating personnel that resistance was encountered during the insertion of the probe to hydrofluorinate the salt in preparation for the removal of the  $U^{235}$ .

The salts contained within the fuel drain/flush tanks have withstood the processing demands of the last four and half decades. The actual utilization within the reactor was limited to only a half decade, followed by radiolytic and salt disposition processes that continue to utilize the salt as a media for transfer. The basic fuel salt similarity across the time span can be attributed to the ability to monitor the composition and to make necessary adjustments in the composition.

## 4.2 CORROSION EFFECTS

General corrosion of the Hastelloy-N during the operation of the reactor seems limited. Levels of chrome, nickel, iron, and molybdenum, the primary constituents of Hastelloy-N, in the salt remained surprisingly low during the operation of the reactor. The primary corrosion mechanism in the system can be assigned to the removal of chromium through the reaction with  $UF_4$  to form  $CrF_2$  by the following reaction:



During the operating period, the Cr level was monitored and adjustments to the  $UF_4$ - $UF_3$  ratio were made to limit the oxidative conditions. There were approximately 30 additions, 238.6 g total, of reductants to the fuel salt during the operation of the reactor in order to maintain the  $U^{3+}$ /Total U. Once the salt was



discharged to the drain tanks and flushed, the ability to make adjustment and/or monitor the oxidative state was lost.

The release of fluorine from the salt in the cold state suggests the potential to form reduced chemical species that may not be soluble within the carrier salt. While these species will not be mobile upon formation during the melting of the salt, their movement can be expected. The generation of fluorine will continue at some level for the immediate future whether the salt remains in the tanks or in the final disposal package. The alternatives to handle the fluorine include the removal of the fluorine as generated, heating the salt to 250°C to promote the recombination of the fluorine with the metals in the salt, or adding a finely-divided getter material to react with the fluorine as it is generated.

Based on the work reported by Lindauer (Lindauer 1969), a significant level of corrosion took place during the removal of the  $U^{235}$  from the fuel salt in preparation for the  $U^{233}$  operating campaign. The work was performed in the fuel processing cell and does not impact the condition of the tanks to be emptied by this effort. During the fluorination to remove the uranium, the level of nickel and chromium in the salt increased greatly from 36 ppm Ni and 170 ppm Cr to 840 ppm and 420 ppm, respectively. The nickel and chromium levels in the fuel salt were reduced prior to the  $U^{233}$  operation, but the metal was removed from the wall of the vessel of the fuel storage tank. It would be instructive to learn the wall thickness before and after the uranium removal in the fuel storage tank. The nickel and chromium was reduced with hydrogen and Zr and the metallic materials filtered prior to using the salt for the  $U^{233}$  fuel runs.

Recent activities have been directed to the removal of the uranium by fluorine vaporization. During the timeframe 2005–2008 each of the tanks was hydrofluorinated under controlled conditions to avoid the creation of a criticality condition. Once the hydrofluorination was completed, the bath was maintained at molten conditions and the salt was fluorinated to generate  $UF_6$ . The  $UF_6$  vaporizes at approximately 60°C and was removed from the tanks and collected on NaF chemical traps. During the fluorination the  $MoF_6$ ,  $CrF_4/CrF_5$ ,  $IF_7$  and fluorides of tellurium, niobium, ruthenium, and antimony were also volatilized (Lindauer 1969). Based upon the DSA, the uranium level after fluorination is less than 50 ppm, and may be even lower (BJC 2009a).

While the removal of the uranium has many advantages in handling the salt in the future, the process for accomplishing the task has a potential to increase the corrosion of the fluorination vessel, in this case each of the salt tanks. Experience gained during the removal of the  $U^{235}$  charge from the fuel salt resulted in the corrosion of the Hastelloy-N at a rate of approximately 0.1 mil/h. Fluorination times are currently unknown for the 2005–2008 effort, but are at least several days. If the salt in the FDT-1 and FDT-2 experience the same increase in Ni and Cr that was experienced in the 1968 work, a nickel loss of 3.74 kg and a Cr loss of 1.16 kg would be expected. Since the fuel drain tanks have been of questionable integrity, this suggests the need to confirm the level of metal loss during the recent processing. Further, the metal fluorides formed from the tank structure add to the material within the salt mixture.

During the recent uranium removal activities a sample of material was obtained from the probe and submitted for X-ray diffraction and scanning electron microscopy analysis (MCLinc 2009). The bulk chemistry results from this analysis indicate the presence of fluorine, zirconium, and nickel. A specific portion of the sample was selected for further examination and gave the relative proportions of these elements. As would be expected, the major component of the energy dispersive X-ray spectroscopy (EDS) spectrum of Region A is fluorine, with lesser amounts of zirconium and nickel. Since the sample was collected from the lower portion of the tank, and also during the fluorination of the system, it might be expected that nickel fluoride would be present based on the discussion above. The solubility of the nickel fluoride in the salt system is unknown, but it could have been rejected from the solution if the local concentration is elevated. The area under the curve for the nickel appears to be approximately one-half

that of the zirconium, and the zirconium is present at 5 mole %. The appearance of nickel metal in the Region A sample suggests a separate phase suspended in the surrounding material. This would support the concern that a separate solid phase is present with the potential to accumulate in isolated locations for the plugging of flow paths.

#### **4.3 CHEMICAL RECOMMENDATIONS**

From a chemical impact standpoint it is recommended that the following actions be taken prior to the transfer of the salt from the FDT-1, FDT-2, and FFT vessels:

- Heat tanks to liquidus and homogenize and collect a representative sample, testing the tank heaters in the process.
- Evaluate the salt sample for corrosion product concentrations (Ni, Cr) to verify the final uranium content and to quantify the current potential to generate fluorine radiolytically.
- Evaluate the thickness of the vessels similar to the evaluation performed in BJC 1998.
- Evaluate the condition of the Fuel Storage Tank relative to wall thickness degradation.

#### **4.4 PROCESS DESIGN CONSIDERATIONS**

As described in Sect. 3, there are three salt drain tanks located in a shielded pit roughly 38 ft below grade. Two of these tanks contain the MSRE reactor fuel salt; the third contains flush salt, which is less contaminated than the fuel salt. All three tanks have had their uranium content reduced to 50 ppm or less (BJC 2009a) by first injecting HF into each tank to rebalance the fluorine chemistry, then by fluorinating the molten salt and collecting the uranium as  $UF_6$  from the off gas. Still, the radioactivity levels remain high in the salts due to the fission products and decay products from both the uranium and the plutonium fuels.

An unsuccessful attempt to transfer the flush salt was made in 2005. In summary, the following steps were taken (Little 2009, Muggridge 2007):

- The flush salt was heated to 600–650°C, and
- The FFT headspace was pressurized to roughly 51 psia with an inert gas, with the intent to push the molten salt through a ½-in transfer line to a series of canisters located in a nearby cell.

When the original transfer did not occur, several steps were taken, including reversing pressure in the transfer line and attempting to install new heat tracing blankets on portions of the transfer line. The salt transfer line between the canisters and the FFT was fluorinated for a period of six days in June, but no specific details of this procedure (e.g. temperature, concentrations) were investigated. After a little more than two months of trying to achieve salt transfer, a decision was made to defer further efforts at salt transfer in favor of concentrating available resources on the more urgent need to remove the uranium from all three tanks. This was completed in the Spring of 2008.

A revised method of removing the salt from the tanks has been proposed, which includes:

- Heating the salt tank to approximately 600°C;
- Inserting a modified probe through the ball valve on top of the tank;
- Connecting this probe to the existing transfer line inside the shielded pit;
- Sealing the tank, then pressurizing the tank headspace with an inert gas to a pressure not to exceed 65 psia (BJC 2009a); and
- Pushing the molten salt through the ½-in transfer line to the same series of canisters located in a nearby cell, as before.

#### **4.5 CONCERNS WITH THE PROPOSED METHOD**

There are numerous concerns associated with the proposed method of salt transfer. First, the integrity of the tanks must be proven to be able to withstand the proposed operating pressures. Next, the pressure drop through the transfer line must be reviewed, and a thorough review of the line heat tracing must be made. Finally, several recommendations are made to improve the likelihood of a successful salt transfer. Each of these topics are addressed in more detail below.

Due to the severity of the recent operational history of all three tanks, it is imperative that all possible precautions be taken to demonstrate that the structural integrity of the tanks is able to withstand the proposed operating conditions. The tanks were originally designed to be pressurized and were operated under pressure during the reactor operations in the 1960s. In preparation of the system to remove uranium, the tanks were subjected to a thorough visual check, including ultrasonic wall thickness and eddy current measurements (BJC 1998, BJC 2000). However, since that time, each of the tanks has been exposed to both HF and F<sub>2</sub> gas during the uranium removal steps while the salt was in a molten state (~450°C). As described in the chemical and metallurgical evaluation sections, and as experienced in the fluorination of the fuel salts in 1968 when the uranium assay was shifted from U<sup>235</sup> to U<sup>233</sup>, even short exposures to fluorine gas under elevated temperatures can result in tank corrosion on the order of 0.1 mil/hour or more (Haubenreich 1970, Lindauer 1969, Notz 1985). The corrosion rates would be exacerbated in the presence of any moisture at all.

At a minimum, nondestructive tests should be performed that are designed to gain an understanding of any recent corrosion damage, including pitting and stress-corrosion cracking damage to the tanks which occurred during fluorination. The tests performed previously in 2000 are primarily visual, pertaining to the wall thickness and/or weld integrity, and will not reveal the historic damage associated with embrittlement due to thermal neutrons or grain boundary defects due to fission product exposure (McCoy 1978). There is still a risk of tank rupture or leakage, and care should be taken if the tanks are pressurized or lifted.

Similar tests should be performed on the salt transfer line between the FFT and the salt canisters, especially if the fluorinating treatment performed in June 2005 was under elevated temperatures.

If, after testing, it is decided to apply pressure to the tank headspace, a step-wise approach is recommended. Prior to heating the salt, isolate the tank and attempt to pressurize the headspace with inert gas, holding it for some period of time to demonstrate leak tightness. Then, if it is still a viable option,

heat up the tank, insert the probe, and apply pressure to the headspace in a gradual manner so as to not overstress the tank structure.

A detailed calculation was performed in 2000 to estimate the required pressure drop that would be expected across the distance of the salt transfer lines (Huxtable 2002b). Embedded in this calculation are the inherent assumptions that: 1) the irradiated aged salt properties remain the same as the original “clean salt” fed to the MSRE in the 1960s; 2) the salt temperature remains at 600°C throughout the transfer process; and 3) the pipeline transfer lengths for all three salt tanks (FFT, FDT-1, and FDT-2) are identical. With these simplifying assumptions, the estimated differential pressure needed between the tanks and the salt canisters is shown in Table 4.

**Table 4. Estimated differential pressure needed between the tanks and the salt containers, given the simplifying assumptions**

	<b>Initial pressure differential required (psi)</b>	<b>Final pressure differential required (psi)</b>
Fuel Flush Tank (FFT)	36.9	41.6
Fuel Drain Tank (FDT-1)	42.7	45.8
Fuel Drain Tank (FDT-2)	43.1	45.8

From a review of the shift supervisor logbooks during the attempted salt transfer operation, the salt canisters were held at a pressure of approximately 6 psia (Little 2009). The FFT headspace pressure was increased to a maximum value of 51.23 psia during the same timeframe, which equates to a pressure differential of 45 psi, about 8 psi above the value estimated in CAJ-02MSRE-A005 (Huxtable 2002b). In theory then, this should have been sufficient pressure to transfer the molten salt. However, there was no evidence of salt transfer at this time. This is likely due in part to the uncertainties associated with using “clean salt” physical property values—an assumption hard to avoid, but difficult to justify given the chemistry differences that have occurred over time. Another key assumption is that no heat loss (e.g., change in salt temperature) occurred during the transfer, which is highly unlikely based on interviews with MSRE personnel regarding the status of heat trace continuity and viewing of videotapes of the tank pit interior.

A rough estimation of the expected heat transfer behavior of the flush salt—again, based on properties of “clean salt” (overall heat coefficients and specific values) (Haubenreich 1970)—supports the premise that the salt would not freeze solid in the transfer line under steady state conditions, even with damaged heat tracing circuits. However, there is no doubt that a reduction in temperature will increase both the density and viscosity of the salt, which causes the Reynold’s number to decrease. This indicates that the salt transfer may fall into the laminar flow regime for the proposed flow rates. This in turn requires a greater pressure drop to effect flow. If one looks at the relative mass of salt and the mass of metal in a given cross-section of the transfer line, one discovers that the metal mass is approximately four times that of the salt. If the metal is cold relative to the salt—as it probably was initially, where the heat tracing was absent or damaged—then the salt will cool as the metal is heated. In order for the salt to remain liquidus, the metal must be approximately 400°C prior to initial entry of the salt into the pipe section. Once flow is established, the salt will maintain the metal at temperature, as described above. This emphasizes the need for effective heat tracing of the lines prior to salt transfer. At best, a non-steady state calculation would be needed to predict actual conditions in the transfer pipeline during the transient period before steady state is achieved.

The flush salt is the least likely of the salts to have solid precipitates that would inhibit transfer.

## 4.6 PROCESS DESIGN RECOMMENDATIONS

Since the actual heat transfer properties of the aged salt are not known, it is difficult to reach a conclusive recommendation, but there are two modifications to the salt transfer system which could increase the probability of success:

- 100% of the transfer line must be heat traced (and tested) to maintain the molten salt temperature while transferring; and
- If the tank integrity will allow, the ability to increase the head space pressure above the current tank pressure of 65 psia stated in the DSA (BJC 2009a) would provide additional process flexibility in the event of difficulties in transfer.

In addition to ensuring full heat trace on the salt transfer line, there are several other process improvements recommended:

- The proposed probe configuration has end caps with four holes drilled into the side, similar to the discharge pipe for the fluorination probe used to inject HF / F<sub>2</sub> (BJC 2005). It is recommended to remove these end caps from the probes in order to reduce the pressure drop losses associated with entry effects and to reduce any issues associated with solids in the tanks.
- The proposed probe has no heating capabilities for its tip. The previous probe had multiple heating zones on its tip, which worked well. It is recommended that the new probe have tip heating capabilities to mitigate any potential problem with inserting a cold probe into a molten bath.
- If possible, it would be beneficial to provide an inert purge flow through the probe during its insertion. This will aid in resisting any plug formation (such as that which occurred in FDT-1 in 2008 (Little 2009, Muggridge 2007) and also aid in mixing the stratified tank contents prior to transfer.
- It will be difficult to remotely connect and subsequently inspect the pipe segments in the pit while the salt is molten, especially the connection nearest the tank ball valve. Since the larger access plate in the pit shielding panel must be removed in order to lower the probe into the pit, it is recommended that the probe be pre-assembled to the extent possible to limit the number of field connections to one: the final connection to the existing transfer line. This will also limit the exposure of personnel to the radiation field of the opened pit.
- It is recommended to add sufficient getter material (Be, Ca, Mg, etc.) to the receiving salt canisters (if planning to transfer the salt) or to the existing salt tanks while the salt is molten (if the transfer does not proceed as planned). In this manner, the long term concern of continued fluorine generation by radiolysis can be mitigated.

## 4.7 CONSIDERATION OF FUTURE OPTIONS

In the course of reviewing the wealth of information pertaining to the MSRE operation and subsequent history, it is clear that many talented individuals have spent a significant effort at both trying to understand the system as it now exists and to project the most responsible means of long-term care of its

residues. The removal of the uranium from the salts was successfully accomplished under a demanding environment, resulting in the major long-term hazard of the MSRE salts—the bulk of the  $U^{233}$  and  $U^{232}$ —being separated out to the ORNL Materials Disposition Program for further care.

The remaining salt at the MSRE still has a high activity level, primarily due to fission products in the “near term” (over the next 1000 years) (Notz 1985). Thereafter, the plutonium and associated daughter products should dominate the total radioactivity at a level approaching 100 Ci, then gradually decay to an estimated 46 Ci in 10,000 years (Peretz 1996). A more explicit estimation of both the long-term radioactivity levels and associated radiolysis impacts should be calculated (via ORIGEN or other means) based on the current composition of the fuel and flush salts.

At this time, the final disposition destination for the MSRE salts (without uranium) is still undecided. The Waste Isolation Pilot Plant (WIPP) may still be a viable alternative, but details (such as acceptable packaging, pretreatment requirements, etc.) remain to be finalized. The CERCLA Record of Decision (EPA 1998) addressed the interim actions required to remove the principal threats of criticality and/or uncontrolled release of contaminants to the environment. These immediate actions have been completed. The same document also states:

*“The salt with the uranium removed will be moved from the drain tanks into storage containers. The salt, which still contains a large quantity of radionuclides, will then be stabilized/packaged to capture fluorine gas which may be generated. (The waste containers will be placed in shielded casks for interim storage.) The casks will be set in an existing storage facility at ORNL and managed there until final disposition is arranged.”*

This is in agreement with the feasibility study conclusions (Peretz 1996) and has traversed through the public comment cycle with mostly positive reviews.

Given the reservations associated with the proposed method of removing the salts from the MSRE drain and flush tanks—tank integrity, salt chemical content, salt physical characteristics, and as low as reasonably achievable considerations—it is recommended that a review be made of the decision to remove the salts from the tanks. It may prove more cost effective (and safer) to heat the tanks, add sufficient (or even an excess) of getter material for addressing the calculated fluorine generation over the years, then either to lift the tanks—in a structurally approved manner—into an above-grade shielded cask or to entomb the tanks as-is, containing the salt in-situ in the cell where they now reside. These alternatives merit a closer look, now that the more immediate concerns have been addressed. It would be prudent, given the failed transfer attempts and the uncertainties highlighted in this evaluation, to review the changes to the situation before proceeding with a recommendation made ten years ago. A review of what has not changed (e.g., uncertainties associated with the decision on permanent disposal options) is also merited.

## **5. SYSTEM EVALUATION**

### **5.1 INTRODUCTION**

This evaluation addresses the conceptual approach for the removal of salt from the Flush Tank and the Drain Tanks at the Molten Salt Reactor described in DOE 2008. This information does not include details, so additional information was developed through interviews with plant personnel (Appendix A), literature searches and reviews (Sects. 7 and 8), reviews of plant drawing (Sects. 7 and 8), and walkdowns of the plant. This additional information was critical to an understanding of exactly how the conceptual approach described in DOE 2008 would actually be implemented. It should be noted here that the limited amount of time available for the evaluation precluded a more thorough evaluation and the results of the evaluation are limited to the information developed and reviewed. There are indications that other factors not fully explored or even identified could negatively impact the proposed removal of salt.

### **5.2 BACKGROUND**

The salt in the Flush and Drain Tanks at the Molten Salt Reactor has been treated to remove most of the fuel. That process was originally intended to include the transfer of the treated salt to canisters, but problems developed when this was initiated on the first tank, i.e., the FFT. The discharge piping from the FFT was blocked and attempts to establish flow through this piping were not successful. The best information identified/developed during the evaluation suggests that solidified salt in this line was never melted, and the suspected cause is an area of pipe adjacent to the tank wall where the heating elements and insulation had been “peeled back.” An attempt was made to remedy this by installing a “box with heating elements” around the pipe and heating it. Other efforts included pressurizing the blockage from the downstream side as well as the tank side, all to no avail. Since a similar situation existed and exists on the next tank (FDT-1), salt transfer was not attempted on that tank. On the last tank to be processed for removal of the fuel (FDT-2), the heating elements and insulation are intact and functional, but there was no attempt to transfer salt.

### **5.3 ADDITIONAL INFORMATION IDENTIFIED**

The proposed salt removal probes (BJC 2005) are similar to the probes used to remove fuel from the molten salt. Several of these probes were purchased and are stored at the plant. An inspection of these probes and the probe drawings revealed the following:

- The probe tip has a cap on the bottom with four 0.31-in-diam radially-oriented openings similar to the fuel removal probes. It is worth noting here that the proposed Work Plan says that “The lower end of this assembly will be equipped with a strainer to prevent sludge from entering the transfer line and causing a plug.”
- There are no provisions for heating the bottom section of the probe.
- The probe discharge piping is heated in sections, using heating cable for two sections and a heating blanket for a section (between the other two sections).
- There are three Locring fittings connecting the sections of probe discharge piping.

- There are two different versions of the probe, with different overall lengths and different lengths of the outer tubing to accommodate the different heights of the flush and drain tanks.

The proposed salt removal process would require inserting the probe into molten salt all the way to the bottom of the tank, securing the probe to the tank nozzle flange, and connecting the probe discharge piping to the canister transfer piping before beginning the transfer (removal) of molten salt. This would be done without the benefit of any flow through the proposed probe into the molten salt (because the discharge of the probe would be open to the atmosphere until it is connected to the canister transfer piping). All of this activity would take place with the head space above the molten salt in the tank exposed to atmosphere and the personnel on the operating floor above exposed to the heat, off gases, and (some) radiation through the openings in the shield plates needed to insert the probe.

The proposed transfer of molten salt from the tanks to the canisters would be powered by putting a gas pressure of 50 psig on the tank head space to push the molten salt up through the 0.622-in inside-diameter probe to the transfer piping to the canisters. Based on previous calculations (Huxtable 2002b), the minimum pressure required to push the molten salt to the canisters is 41.6 psig.

The FFT does not have thermowells like FDT-1 and FDT-2, so the temperature and temperature gradient of the salt cannot be monitored in that tank.

The salt in the drain and flush tanks was not melted by the tank heaters during the fuel removal process. The tank heaters were used to bring the salt up to just below the melting point (450°C), and the heated probe was then used to progressively melt an expanding pool of salt as the probe was lowered into the tank while being accompanied by a flow of gas through the probe into the molten salt. The flow of gas served to mix the molten salt and to combine with and remove fuel from the salt.

The plugging of the probe during the fuel removal process was most likely due to an interruption of the gas flow through the probe, which allowed backflow of molten salt into the probe tip through the 1/8-in-diam radially-oriented openings. Subsequent efforts to clear the openings, i.e., unblock the probe, were unsuccessful. Analysis of material from the blocked orifices revealed the presence of molybdenum, nickel, zirconium, and iron, which would require temperatures of 950 to 1050°C to melt. This would explain why efforts to clear the blockage were unsuccessful.

The review of available information strongly indicates that the salt in the tanks is stratified vertically and may be stratified radially. This stratification of the material could affect the melting point and in turn impact whether salt could be removed or how much salt could be removed from the tanks. Ideally, samples of the salt from multiple points both vertically and radially should be taken and analyzed to determine exactly what the composition is and what its physical properties are. This would provide data to support whatever course of action is chosen regarding the disposition of the salt in the tanks.

## **5.4 AGE OF TANKS AND PIPING**

The primary concerns regarding the age of the tanks and the salt transfer piping is the extent of corrosion that has taken place and if embrittlement has weakened these components. The tanks are the main concern because of their exposure to higher temperatures in the presence of fluorine gas. There is some evidence that very rapid corrosion could have occurred. This could locally weaken the tank walls to the point that the maximum internal pressure applied (50 psig) to push the molten salt (at 660°C) to the canisters could cause a failure which would spill molten salt into the tank vault. This possibility needs to be addressed by surveying the tank wall thicknesses, as was done prior to the fuel removal (Fig. 3.1 in



Peretz 1996). It would also be prudent to do incremental pressure/leak tests at different stages of the salt melting process for each tank. The existing piping to be used for the salt transfer was not exposed to the same conditions, and the review of literature did not identify any other factors of concern, so this piping should be adequate to the task.

## **5.5 CONCLUSIONS**

The proposed approach to removal of salt from the drain and tanks at the Molten Salt Reactor has a number of potential problems:

1. The proposed insertion of an unheated probe at room temperature into the molten salt at a nominal temperature of 600°C runs the risk of locally cooling the salt enough to cause blockage.
2. The proposed (existing) probe discharge path heating configuration between the top of the tank (nozzle) and the connection to the existing transfer line is not continuous. This could result in a blockage if the molten salt cools enough in the areas where the heating is not continuous. Even if there is no blockage, the salt may cool enough to restrict the flow through the areas.
3. The unheated probe is to be inserted all the way to the bottom of the tank before being secured to the flange on the top of the tank nozzle. This presents several problems, including the risk of putting the bottom end of the probe into or against material that is not molten (fluid), which could block flow through the probe. Other problems could result from exposing the head space of the tank to atmosphere during the process and during the period needed to connect the probe discharge piping to the existing transfer piping to the canisters.

## **5.6 RECOMMENDATIONS/SUGGESTIONS**

1. Add instrumentation to monitor the temperature along the full length of the probe and its discharge line.
2. Add heaters and insulation such that the entire length of the probe and discharge line can be heated.
3. Use a probe design such that the tank containment can be maintained during insertion (similar to the probes used for fuel removal).
4. Ensure that the fitting used to connect sections of the probe discharge piping and this piping to the existing canister transfer piping is tightened without constraints on piping, i.e., the connector needs to be free to center itself and its sealing surfaces during the tightening process.
5. Complete a comprehensive survey of the FDT and FFT wall thickness and integrity to determine if the vessel is adequate to contain the proposed pressures without leaking or rupturing.
6. Remove the “cap” from the bottom of the probe to eliminate any restriction in the flow path. Perhaps consider retaining part or most of the side openings at the bottom of the probe to prevent solid contact of the probe bottom with the tank bottom from blocking flow. Additionally, recommend not installing the strainer mentioned in the Work Plan.

7. Consider attempting transfer of FDT-1 salt using the original existing system before using the alternative probe since the intact system may be operational.

## 6. CONCLUSIONS

### 6.1 KEY RECOMMENDATIONS

**Based on several concerns identified during this evaluation, the Evaluation Team recommends not proceeding with the salt transfer by the proposed alternate salt transfer method. The major concerns identified during this evaluation are:**

- **Structural integrity of the tanks.** The main concern is with the corrosion that occurred during the fluorination phase of the uranium removal process. This may also apply to the salt transfer line for the Fuel Flush Tank. *Corrosion Associated with Fluorination in the Oak Ridge National Laboratory Fluoride Volatility Process* (Litman 1961) may show that this problem is significant.
- **Continued generation of Fluorine.** Although the generation of Fluorine will be at a lower rate than experienced before the uranium removal, it will continue to be generated. This needs to be taken into consideration regardless of what actions are taken with the salt.
- **More than one phase of material.** There are likely multiple phases of material in the salt (metal or compound), either suspended through the salt matrix, layered in the bottom of the tank, or both. These phases may contribute to plugging during any planned transfer. There is not enough data to know for sure.
- **Probe heat trace.** The alternate transfer method does not include heat tracing of the bottom of the probe. There is a concern that this may cool the salt and other phases of materials present enough to block the flow of salt.
- **Stress-corrosion cracking.** Additionally, there is a concern regarding moisture that may have been introduced into the tanks and not removed during the heating of the tanks before HF and F<sub>2</sub> sparging. Due to time constraints, this concern was not validated. However, if moisture was introduced into the tanks, there would be an additional concern regarding the potential for stress-corrosion cracking of the tank walls.

### 6.2 SUPPLEMENTAL RECOMMENDATIONS

There are three major hurdles to cross before the team can recommend salt transfer:

1. Tank integrity must be proven
  - Evaluate the corrosion that occurred during the fluorination of the salts during uranium removal.
  - Check the tanks again in a manner similar to that done in 2000. Even though these NDT tests will not detect stress-corrosion cracks or embrittlement, it will be an additional data point as to the corrosive damage done to the tank walls during fluorination. If feasible, removal of a tank metal sample, followed by destructive tests, could provide a measure of the damage not detected

with NDT. The team also recommends that a cold pressure drop test of the tanks and piping with inert gas be considered prior heating the salt. Additionally, consideration to perform pressure testing during the heat-up, as addressed in Sect. 3, may help to indentify a small leak before it gets bad.

- Ensure that the tanks are pressurized slowly, both during testing and for the transfer.

2. Transfer of salt (line pressure drop)

- Little cushion exists on pressure drop calculation. The team recommends evaluating the tanks for a higher pressure rating based on selected NDT data, then raising the safety analysis limit of the maximum tank pressure to 80 psia, so the operating procedure can use the higher pressure if needed.

3. Heat tracing of transfer lines and probe

- Ensure that all lines are 100 % heat traced. This may have been the cause of the failed transfer from the FFT.

If all of these hurdles are cleared, the team also recommends the following:

1. Modify the end of the probe to remove the cap. The end of the current cap has the same small hole design that the old probes had. These probes experienced clogging during uranium processing. It is thought that an open pipe would be better than small radial holes.
2. Heat the whole probe prior to immersion to keep from forcibly injecting molten salt up the probe's inner diameter.
3. While inserting probe, blow inert gas through the probe to aid in mixing the salt and to ensure the probe doesn't plug during insertion.
4. The fitting used to connect the probe discharge piping to the existing canister transfer piping needs to be tightened without constraints on piping, i.e., the connector needs to be free to center itself and its sealing surfaces. The type of fitting that is planned to be used to connect the probe discharge piping to the existing transfer line to the canisters has experienced leakage incidents when it was not installed properly on past projects at ORNL.
5. Consider installing instrumentation on the probe to monitor the probe temperature along the full length of the probe and its discharge line.
6. A more explicit estimation of both the long-term radioactivity levels and associated radiolysis impacts should be calculated (via ORIGEN or other means) based on the current composition of the fuel and flush salts. This will quantify the need for adding getter material to address fluorine generation.

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## **APPENDIX A. PERSONNEL INTERVIEWED**





## APPENDIX A. PERSONNEL INTERVIEWED

The personnel interviewed were:

<b>Name</b>	<b>Title</b>
Gene Muggridge	Facility Manager
James Hylton	Process Engineer
John Bufis	Electrical Engineer
Phil Clark	Shift Supervisor



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