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MARTIN MARIETTA

DECOMMISSIONING OF THE MOLTEN SALT REACTOR EXPERIMENT

A TECHNICAL EVALUATION

K. J. Notz

OPERATED BY
MARTIN MARIETTA ENERGY SYSTEMS, INC.
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K. J. Notz

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operated by
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- For measurements of groundwater radioactivity and sump pump operation: Ken Belitz (Operations Division, with direct responsibility for day-to-day surveillance of the MSRE facility).

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DECOMMISSIONING OF THE MOLTEN SALT REACTOR EXPERIMENT - A TECHNICAL EVALUATION

K. J. Notz

ABSTRACT

This report completes a technical evaluation of decommissioning planning for the former Molten Salt Reactor Experiment, which was shut down in December, 1969. The key issues revolve around the treatment and disposal of some five tons of solid fuel salt which contains over 30 kg of fissionable uranium-233 plus fission products and higher actinides. The chemistry of this material is complicated by the formation of elemental fluorine via a radiolysis reaction under certain conditions.

Supporting studies carried out as part of this evalution include (a) a broad scope analysis of possible options for storage/disposal of the salts, (b) calculation of nuclide decay in future years, (c) technical evaluation of the containment facility and hot cell penetrations, (d) review and update of surveillance and maintenance procedures, (e) measurements of facility groundwater radioactivity and sump pump operation, (f) laboratory studies of the radiolysis reaction, and (g) laboratory studies which resulted in finding a suitable getter for elemental fluorine. In addition, geologic and hydrologic factors of the surrounding area were considered, and also the implications of entombment of the fuel in-place with concrete.

The results of this evalution show that the fuel salt cannot be left in its present form and location permanently. On the other hand, extended storage in its present form is quite acceptable for 20 or 30 years, or even longer. For continued storage in-place, some facility modifications are recommended.

To provide a basis for future, permanent disposal, the material should be melted and repackaged into smaller containers, to facilitate handling. During this operation, a fluorine getter can be added, which will eliminate the need for periodic reheating. The repackaged material can then be stored for an additional period of time, in a manner similar to remotely-handled TRU waste, an operation already being conducted routinely at ORNL. In the repackaged and gettered form, interim storage can be done safely for many years. Final disposal can then follow any number of pathways, none of which are actually available today, but all of which are technically sound, including: disposal in a high-level waste repository, disposal in the Waste Isolation Pilot Plant, disposal via intermediate depth disposal or, blending in with a fluoride-based high-level liquid waste from reprocessing at other DOE sites. Utilization of the melt-repackage-dispose concept should save many millions of dollars relative to the original reprocess-dispose concept. In addition, because melt-repackage is much simpler than reprocessing and uses only benign or inert chemicals, it poses a far smaller threat to both the environment and to the operators assigned to the task.

The existing facility was inspected and all cell penetrations cataloged and examined. These were found to be in acceptable condition, but several improvements are recommended: capping off some additional lines (to assure against accidental water entry) and providing additional isolation valves to two instrument lines (to avoid accidental leakage).

During the course of this study an excellent getter for elemental fluorine in a radiation field was discovered. A patent disclosure was submitted and an application is being filed on this invention.

DECOMMISSIONING OF THE MOLTEN SALT REACTOR EXPERIMENT - A TECHNICAL EVALUATION

1. INTRODUCTION

This is the third and final report in a technical study addressing the question of how best to proceed with decommissioning of the MSRE facility; i.e., the hot cells and ancillary facilities of the former Molten Salt Reactor Experiment, housed in Building 7503 at the Oak Ridge National Laboratory. The hot cells contain the reactor itself, the circulating pump and heat exchanger, the drained (and now solidified) fuel salt and flush salt, salt processing equipment, and related equipment. However, the controling factor in disposing of this material is the fuel salt itself — over five tons of fluoride salt eutectic containing 37 kg of uranium (most of it fissionable ^{233}U) plus fission products (beta-gamma emitters) and actinides (alpha emitters). In addition, the frozen salt is also a source of neutrons, from (alpha, n) reactions, and a potential source of elemental fluorine, from radiolysis reactions induced by the radiation field indigenous to the frozen salts. The critical path item in planning for decommissioning of the MSRE is resolution of how to deal with the fuel (and flush) salt. Until this issue is resolved, all planning is highly uncertain and faced with an unrealistic range of options. No major work on this issue can proceed until an acceptable procedure for the fuel salt has been defined.

The earliest work on decommissioning,[15]* done at a time when the facility was still fully operational and salvage of the contained ^{233}U for other uses was a natural assumption, started with the premise that the fuel and flush salts would be reprocessed, first stripping out the uranium by direct fluorination, then converting the fluoride eutectic residue to oxide by some (unspecified) process for disposal at some (unspecified) location. Once the salts were removed, the hot cells and their contained equipment could be dealt with in one of three ways: (1) remove and dispose of the equipment, then use the hot cells for some

*The reference numbers refer to the citation numbers as listed in the Bibliography, Appendix A.

other project; (2) remove and dispose of the equipment, then raze the hot cells and dispose of the scrap metal and rubble; or, (3) entomb the equipment in-place and seal the hot cells permanently. These procedures were estimated to cost as much as $30 million at that time. In today's situation, with higher costs and more stringent controls and QA requirements, the cost would be much higher.** Thus, there is a very high economic incentive to find a more cost-effective approach of dealing with this material. At the same time, the total systems aspects of the overall task must be given due consideration, to assure successful completion of the entire decommissioning project without adverse environmental effects or unnecessary risks to operating personnel.

**The recently completed CEUSP project (see e.g. P. McGinnis, et al., "Development and Operation of a Unique Conversion/Solidification Process for Highly Radioactive and Fissile Uranium," Nuclear Technology, March 1987) cost $29 million and was similar in many ways: it involved elevated temperatures for the chemical solidification of ^{233}U nitrate solution and packaging into small canisters. However, the quantity of material was much less — about 1.4 tons of solid product vs a total of 10 tons of MSRE fuel salt plus flush salt. Nor were the hot cell and contained equipment decommissioned after completion of the CEUSP project. Thus, the original MSRE estimate might double or even triple under current conditions.

2. HISTORY AND PHYSICAL DESCRIPTION

2.1 HISTORY

The Molten Salt Reactor Experiment (MSRE) was concluded in 1969 after several years of well-planned and highly successful work. This homogeneous reactor concept was based on the thorium/U-233 fuel cycle and used a molten fluoride eutectic as the operating medium. This work is thoroughly documented.

The MSRE was a graphite-moderated, homogeneous-fueled reactor built to investigate the practicality of the molten-salt reactor concept for application to central power stations. It was operated from June 1965 to December 1969 at a nominal full-power level of 8.0 MW. The circulating fuel solution was a eutectic mixture of lithium and beryllium fluorides containing uranium fluoride as the fuel and zirconium fluoride as a chemical stabilizer. The initial fuel charge was highly enriched ^{235}U, which was later replaced with a charge of ^{233}U. Processing capabilities were included as part of the facility for on-line fuel additions, removal of impurities, and uranium recovery. A total of 105,737 MWh was accumulated in the two phases of operation. Following reactor shutdown, the fuel salt was drained into two critically safe storage tanks and isolated in a sealed hot cell, along with a third tank containing the flush salt. In addition, there was also coolant salt which was circulated between a heat exchanger (with the fuel salt on the other side) and a radiator which dumped the heat to a forced-air cooling system.

At shutdown, the fuel salt containing most of the uranium and fission products was divided and drained into two separate tanks, thus ensuring criticality safety. The flush salt, containing 1 to 2% of the uranium and fission products, was drained into a third tank. The salts were allowed to cool and freeze, thereby precluding any leakage and decreasing further the already-low corrosion rate. The drain tanks are made of heavy-walled Hastelloy N, a special alloy created for the program, which has superior strength at high temperatures and outstanding corrosion resistance toward the eutectic fluoride system used for the MSRE. These tanks are contained within a hermetically sealed, stainless steel—lined, reinforced-concrete hot cell, located below grade except for a double set of roof plugs which are accessible from a high-bay service area.

When the reactor was first shut down, it was assumed that the facility and the fuel and flush salts would probably be utilized again at some later date. Therefore, the shutdown procedure was essentially a mothballing operation followed by surveillance and maintenance (S&M) procedures. These procedures were designed to ensure safe temporary storage and to maintain the operational capability of the facility. The surveillance and monitoring program, which includes daily and monthly measurements, has been in force since shutdown.[24] There is also an annual reheat (but not hot enough to melt the salt) to recombine any fluorine that might have formed from radiolysis reactions on the fluoride salt. There have been no adverse incidents or releases of radioactivity since the reactor was shut down over 17 years ago.

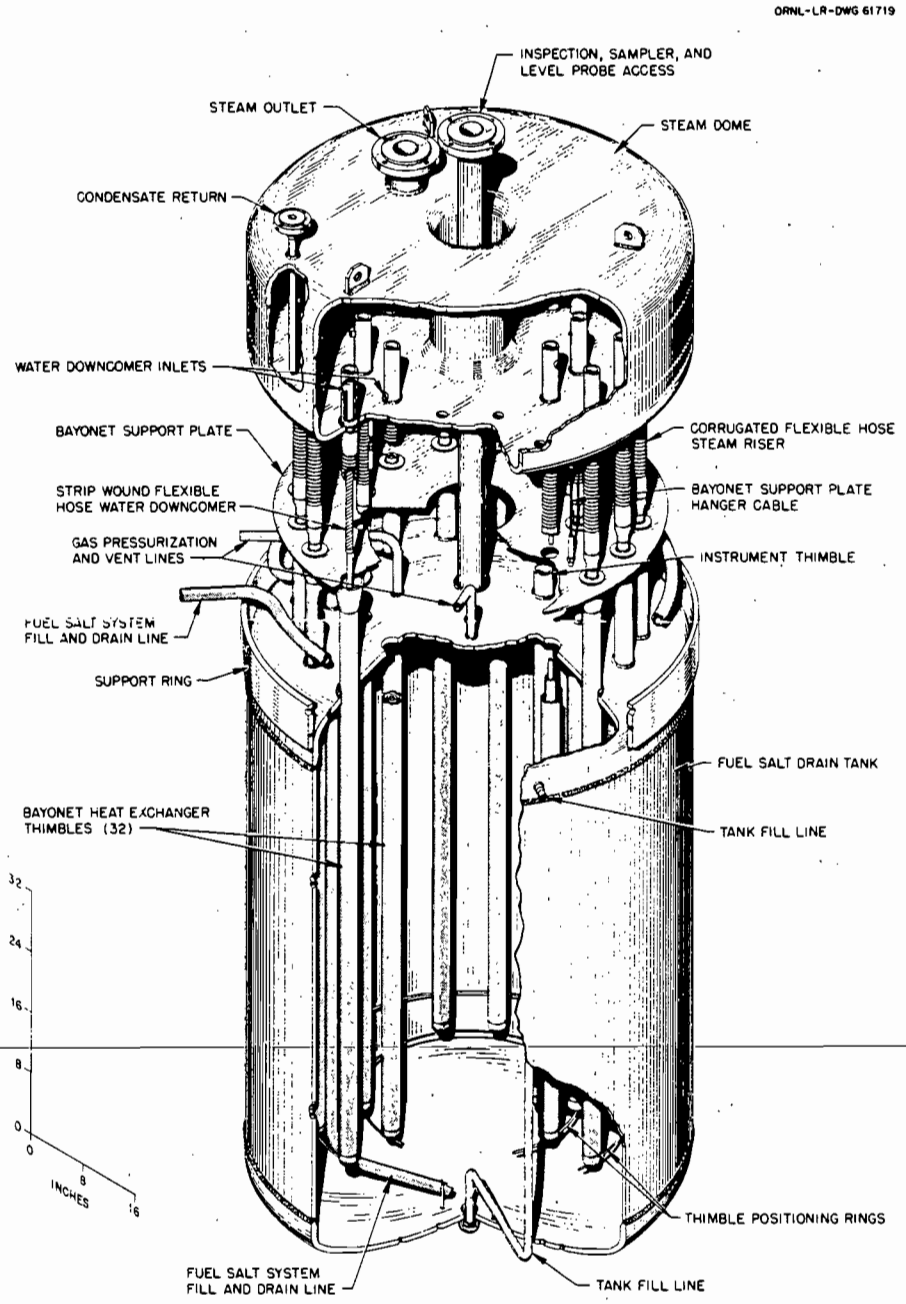
2.2 PHYSICAL DESCRIPTION

The primary reactor and drain system components are contained within two interconnected cells; the coolant and fuel processing systems are located separately within adjoining cells (drawings of these cells are given in ref. 21). The reactor and drain tank hot cells are actually sealed pressure vessels that also provide additional containment for the fuel. The reactor cell is a 24-ft-diam steel tank, while the drain tank cell is a stainless steel—lined reinforced concrete rectangular tank. Each cell has removable roof beams and shield blocks with a welded stainless steel membrane seal that must be cut open for access. The coolant system and fuel process systems are located within shielded hot cells that are kept at a slightly negative pressure and are swept by a containment ventilation system. Access to these cells is gained via removable top shield plugs. The associated support equipment is housed in a steel—concrete— transite structure that has containment features. The hot cells (except for the hermetically-sealed reactor and drain tank cells) and the high-bay area are maintained under negative pressure, with an active ventilation system consisting of centrifugal fans and roughing and HEPA filters that exhaust through a 100-ft tall steel discharge stack. The reactor heat dissipation system included a salt-to-air radiator exhausting through a steel stack and a drain tank

for storage of the coolant salt, where this material now resides. This stored coolant salt is nonradioactive. Ancillary facilities at the site include an office building, a diesel generator house, a utility building, a blower house, a cooling-water tower, and a vapor condensing system. These facilities have been described in detail in other reports.[4,5,15,18] The three hot cells of most concern to this study (reactor, drain, and processing) are described in terms of penetrations and "containment envelopes" in Sect. 4.3.1 of ref. 21. One of the two fuel-salt drain tanks is shown in Fig. 1. The properties and the major components of the material of construction, Hastelloy N (also called INOR-8), are listed in Table 1.[15] This alloy, which was developed for the MSRE, is highly resistant to fluoride corrosion, has high strength at reactor operating temperature (650°C), and is relatively insensitive to neutron embrittlement. These three tanks are each 50 inches in diameter by 86 inches high, with a contained volume of 80 cubic feet.

2.3 FLUORIDE SALTS

The presence of the solidified, stored fuel and flush salts is the most significant aspect of the MSRE decommissioning task. More than 4600 kg of fuel salt and 4300 kg of flush salt, containing about 37 kg of uranium (primarily U-233) and 743 g of plutonium (primarily ^{239}Pu) are present in the drain tanks. Calculated fission product activities (mainly beta-gamma) of these salts, decayed to 1987, total about 30,000 Ci. The alpha activity from transuranic isotopes and their daughters amounts to about 2000 Ci. These isotopes are divided roughly 98-99% in the fuel salt and 1-2% in the flush salt. The total alpha activity of the fuel salt is relatively high, about 400,000 nCi/g, while that of the flush salt is about 6000 nCi/g. The total decay heat at present is about 200 W, with three-fourths coming from the beta-gamma component and the remainder from the alpha emissions. The compositions of the fuel, flush and coolant salts are given in Table 2. The latter salt, which served as a secondary coolant, is not radioactive and is stored in the coolant cell. It is of interest primarily because it contains 338 kg of high-purity lithium-7, which could be recovered if there were sufficient economic incentive.



MSR Primary Drain and Fill Tank

Fig. 1. Fuel-salt drain tank.

Table 1. Composition and properties of INOR-8 (also known as Hastelloy N)

Chemical compositon, %	
Ni	66-71
Mo	15-18
Cr	6-8
Fe, max	5
C	0.04-0.08
Ti + Al, max	0.5
S, max	0.02
Mn, max	1.0
Mn, max	1.0
Si, max	1.0
Cu, max	0.35
B, max	0.01
W, max	0.5
P, max	0.015
Co, max	0.2
Physical properties:	
Density, lb/in.^{3}	0.317
Melting point, °F	2470-2555
Thermal conductivity, Btu/(h•ft•°F) at 1300°F	12.7
Modulus of elasticity at ~1300°F, psi	24.8×10^6
Specific heat, Btu/lb•°F at 1300°F	0.138
Mean coefficient of thermal expansion, 70 to 1300°F range, in./in.•°F	8.0×10^{-6}
Mechanical properties:	
Maximum allowable stress,[a] psi	
1000°F	17,000
1100°F	13,000
1200°F	6,000
1300°F	3,500

[a]ASME Boiler and Pressure Vessel Code, Case 1315.

Table 2. Stored MSRE salts

	Fuel salt[a]	Flush salt[b]	Coolant salt[b]
Total mass, kg	4650	4290	2610
Volume, ft^3 at room temperature	66.4	69.9	42.5
Density, g/cm^3, at RT	2.47	2.17	2.17
at 600°C	2.27	--	--
Composition, mol %			
LiF	64.5	66[c]	66[c]
BeF_2	30.3	34[c]	34[c]
ZrF_4	5.0	--	--
UF_4	0.13	--	--
Uranium content, kg			
U-232	d	d	
U-233	30.82	0.19	--
U-234	2.74	0.02	--
U-235	0.85	0.09	--
U-236	0.04	0.00	--
U-238	2.01	0.19	--
Total	36.46	0.49	
Plutonium content, g			
Pu-239	657	13	--
Pu-240	69	2	--
Other Pu	2	0	--
Total	728	15	--
Lithium composition, %			
Li-6	--	0.009[e]	0.009
Li-7	--	99.991[e]	99.991

[a]Melting point (liquidus temperature) is 434°C (813°F).

[b]Trace-element analyses of 39 batches used for both salts gave 16 ppm Cr, 39 ppm Ni, and 121 ppm Fe. Twelve other analyses of the flush salt gave 38, 22, and 118 ppm, respectively. (Note: Could the Cr and Ni have been interchanged?) In another series of 22 batches, the corresponding values were 19, 25, and 166 ppm.

[c]Reported values. Analytical data for batches 116–161 gave 63 and 37%, calculated from reported values of 12.95 wt % Li, 9.75 wt % Be, and 77.1 wt % F. For batches 101–130, the calculated composition was 64.3 and 35.7%.

[d]Present at 220 ppm, U-basis.

[e]For batches 116–142. The values are 0.0065/99.9935 for batches 143–161.

2.4 RADIATION AND CONTAINMENT

As expected, the radiation hazards associated with the stored fuel are significant. Gamma and neutron dose rates within the reactor and storage cells are in the 10^3 rad/h range. Some of this radiation results from (α,n) reactions with the eutectic salt base. The high radiation field also causes the generation of some fluorine radicals, which slowly accumulate and could, after a suitable incubation period, lead to the presence of free fluorine, i.e. F_2. Since recombination of these radicals (with the metal simultaneously set free in the radiolysis) is accelerated by increased temperature, the salt is reheated periodically; however, it is not melted. The stored salts are in a stable, noncorrosive state, as dry frozen solids.

In addition to the stored fuel, the principal areas of concern at the MSRE are the reactor components and process equipment remaining in the below-grade containment cells. These components are internally contaminated and, in some cases, highly neutron activated. Exposure rates of up to 2200 R/h have been measured in the reactor vessel, attributed primarily to cobalt. The inventory of residual radioactive materials in the reactor and fuel processing cells is estimated to be several thousand curies, with the majority of that activity being associated with fission and activation products. The remaining cells, process piping, and associated operating areas are known to be slightly contaminated. The readily accessible areas of the reactor building (including the reactor bay and office areas) are generally uncontaminated and are being used for laboratory and office space, as well as for storage of various materials.

The MSRE facility appears to be structurally sound and capable of retaining the current radionuclide inventory for many years. No significant spread of contamination or personnel exposure has occurred during the time the facility has been shutdown. The cell ventilation and off-gas system has been operated since shutdown, and the stack is monitored continuously by the ORNL Waste Operations Control Center.

2.5 SURVEILLANCE AND MAINTENANCE

A comprehensive maintenance and surveillance program is provided to ensure adequate containment of the residual radioactivity at the MSRE.[11,24] Routine inspections of the containment systems and building services,

radiological surveillance of operating areas and ventilation exhaust, stored-salt monitoring (temperature and pressure), and periodic testing of safety systems are performed as part of this program. In addition, the fuel and flush salts are reheated periodically in order to facilitate recombination of fluorine and the containment cells are subjected to a leak test, both on an annual basis. Facility maintenance includes general repairs, exhaust duct filter changes, and instrumentation and controls maintenance. Consolidation of the surveillance instrumentation was completed recently. Periodic heater and controls tests are planned as future improvements to the current program.

The salt storage cells have been under a planned program of regular surveillance and maintenance since the reactor was shut down in late 1969. This program includes daily observations of certain parameters by the Waste Operations Control Center and monthly observations by the Reactor Operations Group. The daily observations include measurements of internal cell temperature, building-air radioactivity, pressure differentials of the cell ventilation system, and stack off-gas radioactivity levels in terms of alpha, beta-gamma, and radioiodine. These observations are recorded on log sheets, which have been kept since 1969. The radioactivity data are now also computerized and coordinated through the central Waste Operations Control Center. A primary input point to the Center is located in the control room in 7503.

The monthly checklist involves a complete walk-through inspection of the entire facility plus recordings of in-cell temperatures and sump levels at seven locations. Any necessary maintenance items are noted and added to the list of work to be scheduled. These logs have also been kept since 1969, and they were reviewed by the original author in 1986, who proposed some simplifications, based on the performance since 1969.[28]

On an annual basis, the fuel tanks and the flush salt tank are reheated to recombine any free fluorine that may have formed from radiolysis. The prescribed temperature range for the reheating is >300°F (to ensure that the diffusion rate is fast enough) but <500°F (well below the melting point of 840°F). The last reheat was in October 1987. The reactor and drain tank cells are also leak-tested annually, and a check is made of about 40 equipment items. In addition, special checks are made of the ventilation system, two of the sump pumps, and the DOP efficiency of the ventilation filter.

During the course of this study, it became apparent that data on building groundwater were needed. Therefore, two additional items were added to the S&M procedures: a periodic check of the radioactivity of the building sump discharge, and a periodic check of the operating frequency of the building sump pump. The radioactivity measurements, which were initiated in June 1985, have consistently shown no activity above background. The sump pump monitoring was started in August 1985 and readings show an average operating cycle of about 0.9 h/d. These added readings were continued for one year, in order to obtain data over all four seasons, and have now been discontinued (see Section 3.4.1).

2.6 PRIOR STUDIES

Decommissioning of the MSRE presents some unique problems because of the presence of the fuel and flush salts. Plans for site decommissioning will first have to address the issue of disposition of the fuel. In the early studies, it was generally assumed that the stored fuel would be removed from the MSRE cells, with reprocessing (fluorination) to strip out the uranium and some of the fission products, converted to oxide, and then sent to a final repository or to retrievable storage. However, these operations are complex and potentially hazardous and, therefore, also very expensive.[13,15,16,18]

2.7 UTILIZATION OF THE FACILITY

Thus far, no need for the recoverable ^{233}U or for the cell space has been established. Therefore, there has been no incentive from that direction to proceed with decommissioning. However, the MSRE building facilities are being used extensively, particularly the office areas and the storage space in the high-bay and receiving areas. Approximately 50 people from the Health and Safety Research Division are housed in the office complex, and this number is expected to grow. The history of the Laboratory clearly shows an ever increasing need for office space. Therefore, the office facilities should be retained in any event.

The need for storage space for low-level radioactive samples and materials has also continued to grow. At present, the building provides space for soil samples (about 100 55-gal drums) and miscellaneous equipment (e.g., about 20 remote manipulators). Previously, cesium-137 sources

(about 260 in 200-lb lead pigs), were also stored there. These storage facilities need to be retained. Even a conservative estimate suggests an additional 20-year lifetime for the offices and the storage space.

Looking beyond utilization already extant, the hot cells themselves are a valuable asset whose future utility should be considered. Because of activation, the reactor, drain, and processing cells are likely candidates for entombment unless a major project comes along that would justify the expense of refurbishment. Even with entombment of these three cells, however, they could first be filled with activated metal scrap removed from other cells. Entombment is a viable technique because cobalt, the major activation product, has a half-life of only 5.3 years. The remaining cells could then be used for storage purposes or, with modification, for a suitable project.

3. OVERVIEW OF THE EVALUATION

The two previous reports[20,21] in this evaluation include information that bears directly on the overall evaluation and are, therefore, an integral part of the total evaluation. Much of what was covered in the previous reports is still applicable, either as it was reported at that time or with minor modifications. Even those few portions that were subsequently discarded or modified significantly are germane, because they cover aspects that were in fact, evaluated and should therefore be documented. The following sections summarize those factors that were covered in the two previous reports; the most important highlights are summarized, with references to the appropriate sources. New work (first described in this report) is also summarized, with details provided in Appendices B, C, and D.

3.1 RADIOLOGICAL PROPERTIES

The underlying basis for the conclusion that the fuel salt must eventually be removed to a permanent repository location derives directly from its radiological properties; more specifically, from the decay properties of the actinides and their daughters. Figure 2 shows the "double hump" envelope, which is the net result of early decay of several relatively short-lived parents, followed by the long-lived decay chain initiating with ^{233}U. As a consequence, the alpha activity remains relatively constant within one order of magnitude for a half million years. Although the thermal output is quite low (about 50 watts), the radiolysis reaction will also continue for a half million years due to these alpha decays (see later discussion on radiolysis).

The alpha decay chains, starting with uranium or plutonium isotopes, go through a series of six or more alpha decays, each of which contributes decay energy, before attaining a stable state. Decays of this kind are portrayed graphically in a 1982 report.[22]

The flush salt is much lower in radioactivity than the fuel salt, about 1 to 2% as much, but still contains enough alpha activity to exceed the lower limit for TRU waste by a factor of about ten. Therefore, it will also require permanent isolation from the envirosphere, along with the fuel salt.

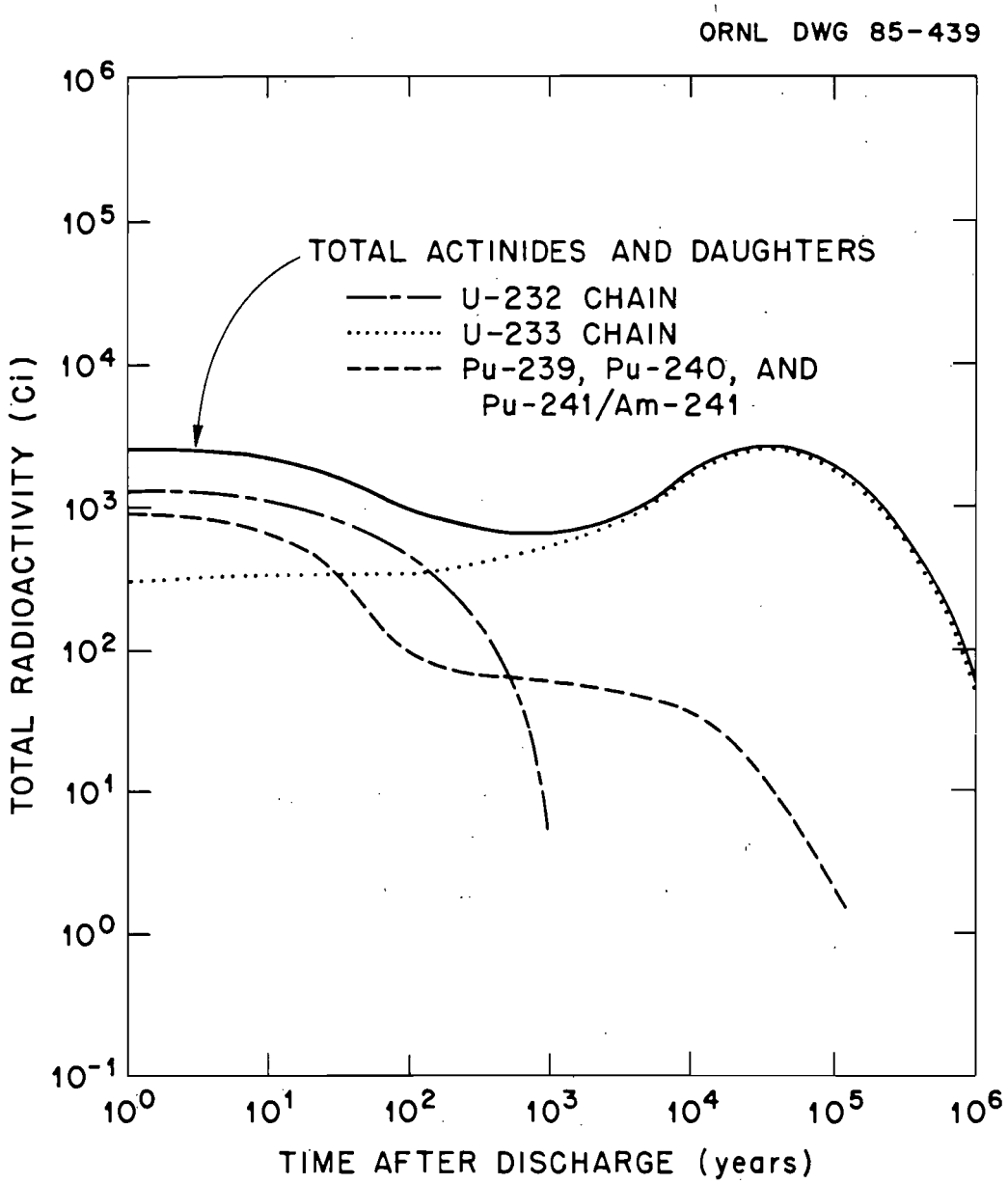


Fig. 2. Actinide and daughter activity of MSRE fuel and flush salts.

The fission product activities decay away in the usual manner (Fig. 3). These exhibit only one unique feature, the long-term residual activity of zirconium-93. This is an activation product not normally noticed in LWR or HTGR spent fuel. It is evident here because of the relatively large amount of zirconium present in the eutectic salt mixture as ZrF_4.

Since radiolysis yields are generally a function of the net energy input to the system (and not the curies of activity), the thermal yields from both categories of activity were calculated (see Fig. 4). Again, the persistent presence of the actinides and their daughters are clearly shown. The thermal output is roughly equivalent to that from aged defense HLW, which is almost an order of magnitude less radioactive than LWR spent fuel.

The neutron activity of the salts from (α,n) reactions is much greater than normal because of the presence of ^{9}Be, ^{19}F, and ^{7}Li. These isotopes, particularly the ^{9}Be and ^{19}F, are excellent targets for (α,n) reactions. At present (17 years after discharge), there are about 6 x 10^{9} neutrons/s from this source. This is about three times the rate from one milligram of ^{252}Cf, a very potent source, which is used as a neutron source. Spontaneous fission also contributes some neutron activity. These values, listed in Table 3, follow the actinide curve with time.[21] Less than 1% of the alphas are converted to neutrons.

Table 3. Neutron activity of fuel salt[a]

Time (years)	(α,n) neutrons/sec			Spontaneous fission neutrons/sec
	^{9}Be	^{19}F	^{7}Li	
Discharge	1.8 E9	4.7 E9	1.6 E7	1.86 E6
1	1.8 E9	4.7 E9	1.6 E7	1.75 E6
10	1.6 E9	4.1 E9	1.4 E7	1.69 E6
100	7 E8	1.8 E9	6 E6	9.30 E5
1000	4 E8	1.1 E9	4 E6	5.23 E5
10,000	1.3 E9	3.5 E9	1.2 E7	1.74 E6
100,000	1.4 E9	3.7 E9	1.3 E7	1.89 E6
1,000,000	4 E7	1 E8	4 E6	4.64 E4

[a]Calculated with ORIGEN2 code.

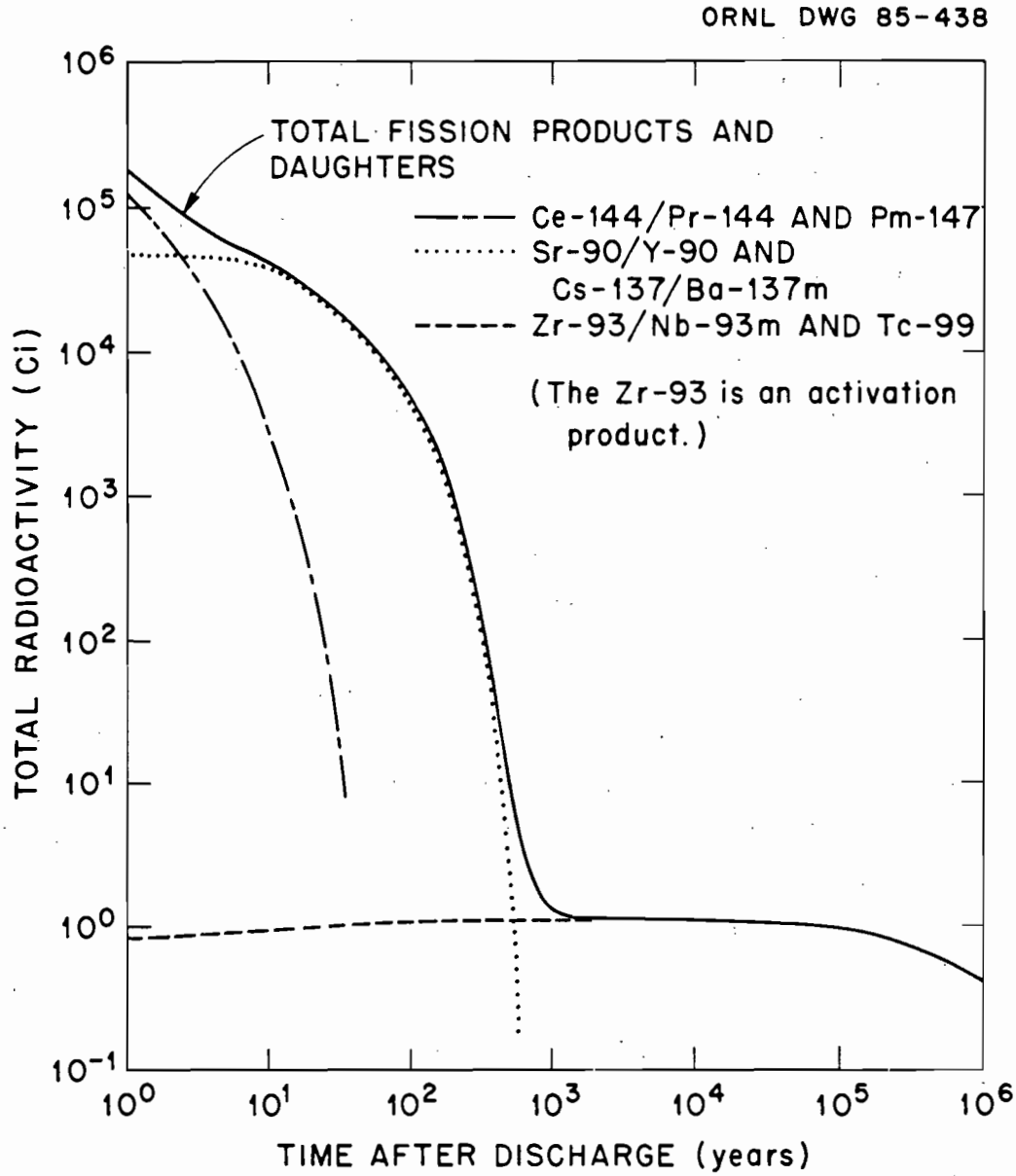


Fig. 3. Fission product activity of MSRE fuel and flush salts.

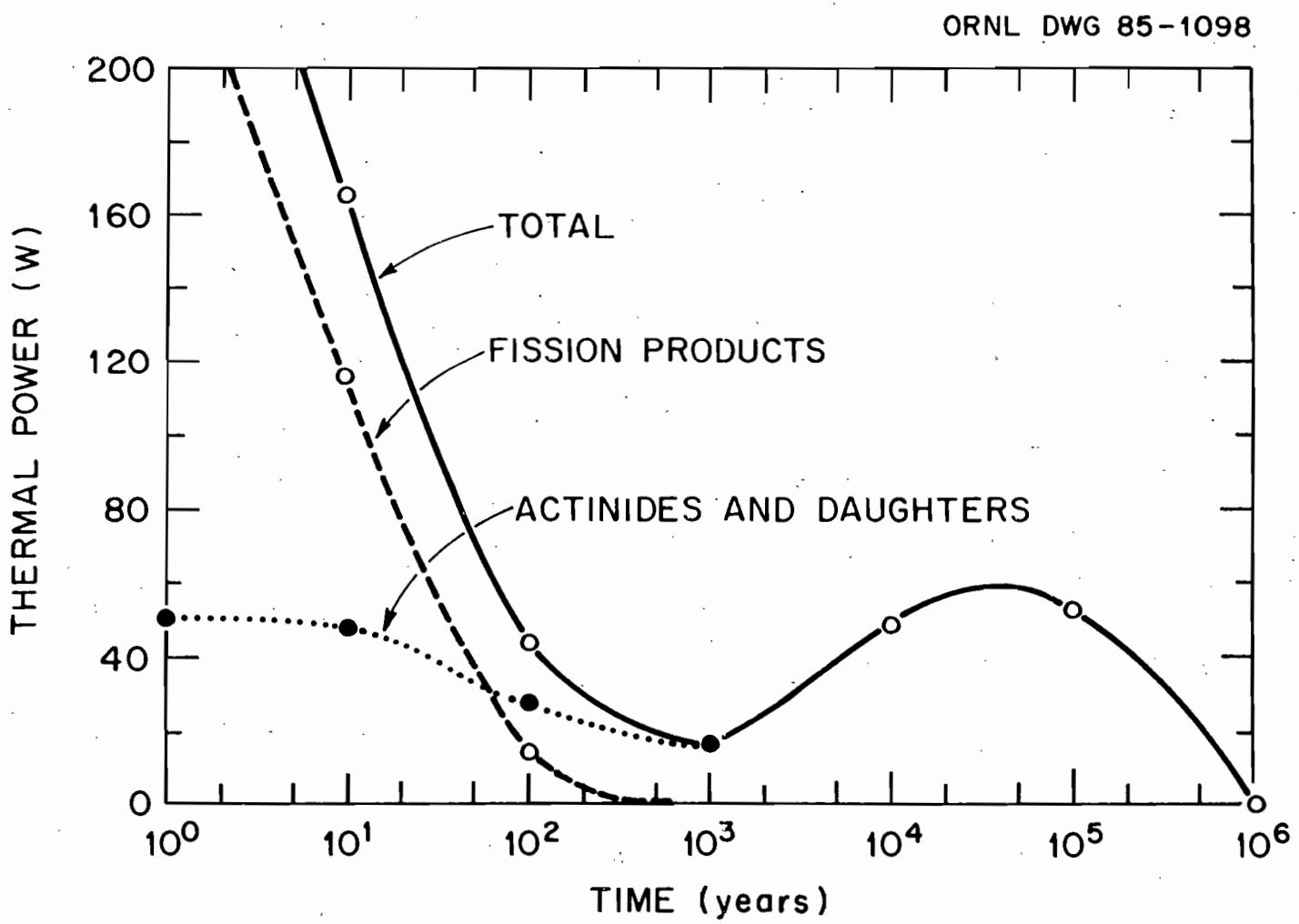


Fig. 4. Thermal power of MSRE fuel salt.

The transuranic alpha activity of the salts is relatively high, about 70,000 nCi/g in the fuel salt and about 1,000 nCi/g in the flush salt. (The total alpha activity, including that from all the daughters, is about six times greater.) Removal of the uranium and neptunium by fluorination could lower the activity of the flush salt below the threshold level for TRU waste, but the fuel salt would still be a TRU waste because of the plutonium content.

These decay calculations were made using the ORIGEN2 code. They are documented in considerable detail in the previous report.[21]

3.2 RADIOLYSIS BEHAVIOR

The predicted formation of free molecular fluorine (F_2) as a consequence of radiolysis of the fluoride (F^-) salts, was thoroughly discussed in the previous report.[21] Earlier work by Haubenreich[10] provided a basis for calculating fluorine production as a function of absorbed radiation energy. An incubation period is involved, during which the initial production of fluorine atoms (F°) builds up the concentration to a level sufficient to drive diffusion to the grain boundaries, where molecular F_2 forms. Formation of fluorine atoms is of course accompanied by the simultaneous formation of free metal atoms. These are available to recombine with the liberated fluorine, and do so more rapidly at higher temperatures, where diffusion within the grains is more rapid. Haubenreich estimated that the reverse reaction would equal the forward reaction at 145°C. From his data, which were obtained at several temperatures, we have calculated an activation energy of 19.4 kcal/mole, which is not unreasonable for a diffusion-controlled process.

Using Haubenreich's factors for the induction period and for fluorine yields, and the thermal power calculated using ORIGEN2, Tables 4, 5, and 6 were prepared.[21] Table 4 shows fluorine production to continue at a significant rate for over 100,000 years. Tables 5 and 6 cover only the period 10 to 100 years. They show that although fluorine yields drop by a factor of four during that time, the integrated total, if allowed to accumulate in the closed drain tanks, would give a fluorine pressure of almost 100 psi, which would be an undesirable condition. On the other hand, because of the induction period, which is

Table 4. Thermal power and calculated fluorine yield rate

Time (years)	Thermal power (W)[a]			Induction period (years)	Fluorine yield (cm^3/h)
	Actinides and daughters	Fission products	Total		
Discharge	49	5,655	5,704	b	b
1	50	682	732	0.9	124
10	49	116	165	4	28
100	29	14	43	16	7
1,000	17	-	17	40	3
10,000	48	-	48	14	8
100,000	51	-	51	13	9
1,000,000	1.3	-	1.3	530	0.2

[a]Calculated with ORIGEN2 code.

[b]Salt is molten; recombination is very fast.

Table 5. Calculated radiolysis yields for the 10- to 100-year period

Time after discharge[a] (years)	Estimated thermal power (W)	Induction period (years)	Fluorine yield (cm^3/h)
10	165	4.2	28
15	140	4.9	24
20	122	5.7	21
30	98	7.0	17
40	83	8.3	14
50	72	9.6	12
60	63	11.0	11
70	57	12.1	10
80	51	13.5	9
90	47	14.7	8
100	43	16.0	7

[a]The reactor was shut down on December 12, 1969.

Table 6. Calculated pressure rise in fuel salt drain tanks, assuming last reheat was in 1985

Time after discharge[a] (years)	Pressure rise rate (psi/year)[b]	Total pressure buildup (psi)[b]
20	2.0	2
30	1.6	20
40	1.4	35
50	1.2	48
60	1.1	59
70	1.0	70
80	0.9	80
90	0.8	88
100	0.7	95

[a]The reactor was shut down on December 12, 1969.
[b]For the drain tanks, with a void volume of 47 ft^3 in each tank.

about five years at present, no fluorine pressure will result until five years after reheating is halted. This is shown in Table 6 for an assumed last reheat in 1985; the fluorine pressure would be only 2 psi by 1990. However, it would continue to rise thereafter, but at a declining rate. Thus, one approach to verify the Haubenreich data and the calculated factors, would be to stop reheating, and start observations to detect fluorine buildup. The obvious shortcomings to this approach are the long induction period and the relatively slow response after that.

Because of the importance of radiolysis to this evaluation, experimental verification of Haubenreich's data was deemed essential. Therefore, an experimental program was carried out by Toth and Felker. A status report on their work is given in Appendix B. A technical report will be issued next year, after work still in process is completed. They repeated the gamma radiolysis work of Haubenreich, but in a much stronger radiation field (provided by spent HFIR fuel elements), so that easily measurable results could be obtained within months, rather than years. They tested several getters, one of which (activated charcoal), proved to be very effective. They also obtained data on aqueous solubility of the eutectic salt because prior data were too limited to be of any real value. Finally, they are now conducting a test using alpha radiation (from a ^{238}Pu source), which is a closer analog to the real case, for predicting long-term behavior, where only alpha activity is present in significant quantity.

The gamma radiolysis tests confirmed the Haubenreich results, at least in a qualitative sense. Figure 5 shows pressure readings on a sealed capsule over a 3-month period. An induction period was followed by a pressure rise, which also reflected an increase in rate when the radiation level was increased. Interestingly, the pressure rise leveled off when just under 2% of the contained fluoride has been converted to fluorine. This indicates that at this high degree of conversion the rate of recombination equals the rate of formation, even at ambient temperature. Extension of this work gave data on recombination rates at temperatures of 100, 150, and 200°C, from which an activation energy of 8.74 K cal/mole was calculated. This is about half the value calculated from Haubenreich's data, which is not unreasonable since Toth and Felker obtained their results at a much higher concentration of defects (in the solid crystals), which should enhance diffusion-controlled recombination.

Although it had been suggested earlier[21] that an active metal such as calcium or beryllium might make a good getter for F_2, initial testing was done with graphite. Graphite was selected because of its known compatibility with the fluoride eutectic (from its use in the reactor, where it served as a neutron moderator) and because graphite forms a stable intercalation compound with halogens. It was found to function as a getter, but the capacity was low. Even after steam activation, graphite had a very limited capacity. Activated charcoal was then tested, and found to have both a high capacity and favorable kinetics. The samples tested reacted with 60% of their own weight of F_2, and would subsequently retain this amount of fluorine, even under vacuum.

Finally, the gettering capacity of activated charcoal was tested in a radiation field. Based on the radiolysis experience with the eutectic fluoride salt, dissociation was to be expected. However, on testing the opposite occurred. A fluorine-loaded sample, with a fluorine overpressure added, actually reacted with more fluorine under irradiation. The reaction proceeded to such an extent that a negative pressure (relative to ambient) was attained within the test capsule. The pressure dropped from 7 psig to -10 psig. Activated charcoal is thus an ideal fluorine getter for use in this system, and it appears that radiation enhances the reaction between activated charcoal and fluorine. A patent disclosure has been filed on this process.

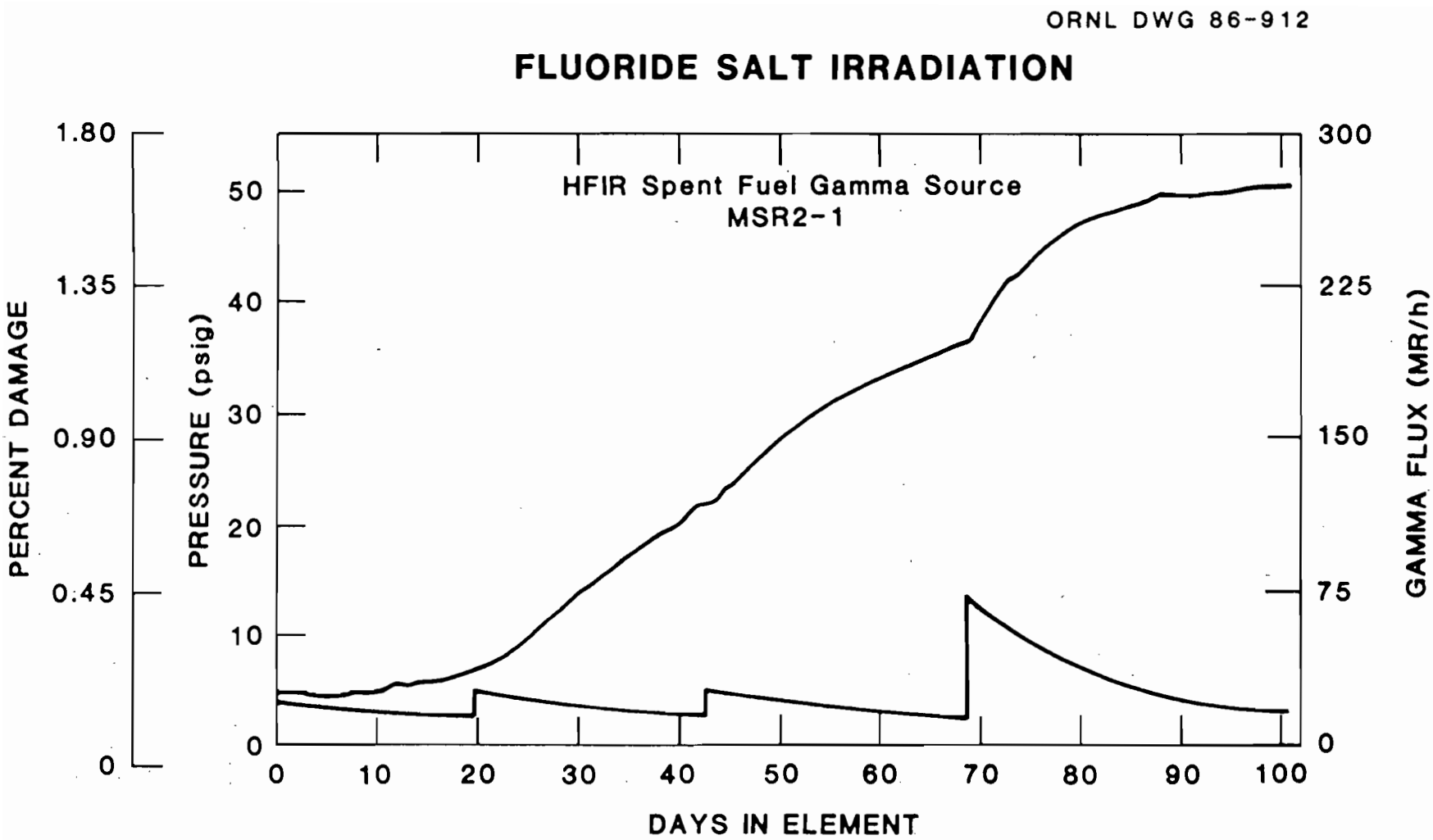


Fig. 5. Fluorine pressure resulting from radiolysis of MSRE eutectic salt.

The real test of radiolysis — in terms of long-term behavior — is in an alpha field, since this is what persists after the beta and gamma radiation have largely decayed away. Such a test requires the in situ addition of an alpha source, because of the very short range of alpha particles. Such a test is now under way, using ^{238}Pu as the alpha source. Since radiolysis effects are generally correlated on the basis of energy input, independent of the form of the radiation, no drastic differences are expected. However, since alpha radiation has a much higher LET — Linear Energy Transfer — than either beta or gamma radiation, some differences could arise. These should however, be differences of degree and not of kind.

3.3 SOLUBILITY BEHAVIOR

In considering alternative treatment possibilities, the solubility of the eutectic salt is of interest. The reported aqueous solubilities of the individual salts varies from "very soluble" for BeF_2 to rather low values for the other components: 2.7 g/liter for LiF and 13.9 g/liter for ZrF_4 (Lange's Handbook of Chemistry). Uranium tetrafluoride also has a low solubility. Limited studies in the early sixties showed the binary mixture of LiF-BeF_2 to dissolve incongruently; i.e. the aqueous composition had a different mole ratio than the solid residue.[1]

Three tests of the solubility of the eutectic were made, all at ambient temperature. Three solvents were tested: water, 4M HF, and 4M HF plus 2M HNO_3. These data are also reported in Appendix B. The two acids were tested because these compositions correspond to solutions used at the Idaho Chemical Processing Plant (ICPP), where fluoride-containing materials are routinely processed. (The fluorides are usually precipitated as CaF_2 prior to calcination.) The results are given in Table 7, part A, expressed as grams/liter. Part B converts these values to moles/liter, and Part C gives the mole ratios for the solutions and also for the starting solid. It is obvious that dissolution is indeed incongruent, and that the composition of the solvent has a strong influence on the relative solubilities. In all cases, the presence of the BeF_2 enhanced the solubilities of the two other salts, presumably by complex formation. The effects of HF and HNO_3 on

Table 7. Solubilities of eutectic salt in various solvents

A - Expressed in grams/liter:

Solvent:	H_2O	4M HF	4M HF + 2M HNO_3
LiF	5.58	2.94	3.33
BeF_2	2.73	5.01	2.47
ZrF_4	25.01	10.40	4.21

B - Expressed as moles/liter:

Solvent:	H_2O	4M HF	4M HF + 2M HNO_3
LiF	0.215	0.113	0.128
BeF_2	0.058	0.107	0.053
ZrF_4	0.150	0.062	0.025

C - Expressed as mole ratios:[a]

	Solid	H_2O	4M HF	4M HF + 2M HNO_3
LiF	12.9	3.7	1.8	5.1
BeF_2	6.1	1.0	1.7	2.1
ZrF_4	1.0	2.6	1.0	1.0

[a]Based on the lowest component in each case.

solubility are due to complex formation and/or hydrolysis effects due to the altered pH. In any event, it appears that the eutectic salt could be put into solution, should that be desired.

In this context, the ICPP flowsheet for high-level waste was reviewed, since they deal with a high-fluoride liquid HLW from the processing of zirconium-clad defense fuel. After calcination, this waste yields a calcine that is one-third CaF_2 and is also high in zirconium.

The fluoride and zirconium from MSRE salts would obviously be compatible with such a mixture, but the chemical compatibility of the lithium and beryellium is an open question. At present, institutional restrictions preclude the ^{233}U itself at the ICPP (because of ^{235}U recovery operations carried out there).[23] There is also concern over the poisonous aspects of the beryllium. Whether or not this restriction and this concern could be circumvented at some future time is a matter of conjecture, but the possibility exists since the waste stream could probably be entered downstream of the recovery operations. This topic is discussed in more detail in Appendix D.

3.4 STATUS OF THE FACILITY

During the course of this evaluation, the status of the physical plant was considered in terms of several time horizons, with distinctive objectives and requirements:

Near-term: About 20 years. The time frame within which present practices of containment, surveillance and maintenance, annual reheating, etc. could reasonably be expected to be continued, under present (and projectable) administrative controls. This is also the time frame within which physical operations could be carried out with a high degree of confidence, while the mechanical components are known to still be functional and personnel with personal knowledge of the facility, based on actual experience, are still available. Physical operations could include modifications to cell penetrations or the building ventilation system, in-cell alterations such as removal of steam domes, addition of more in-cell instrumentation, remelting and repackaging of the salts either with or without addition of a fluorine getter, or actual reprocessing of the fluoride salts to strip out the uranium and convert the large residue to the oxide form.

Mid-term: About 20 to 100 years. The time frame within which the physical integrity of the hot cells themselves can be viewed with confidence, even in the absence of administrative or institutional controls. This is based on the well-known longevity of reinforced concrete structures, even under adverse conditions. This is the time frame within which extended on-site storage of the repackaged

and gettered salts might be required, in the event a final disposal site is delayed more than 20 years from the present. It is also the time frame wherein the most troublesome activation product, cobalt-60, would decay away.

Long-term: About 100 to 1000 years. The time frame within which the hot cell structures can reasonably be expected to survive more-or-less intact, based on historical experience. This is the time frame within which entombment-in-place needs to be evaluated, if it is to be given serious consideration (for entombment of the equipment after removal of the salts). It is also the time frame wherein the fission products decay away; after 1000 years only ^{233}U and some plutonium isotopes contribute significant radioactivity.

As the evaluation developed, it became evident that entombment-in-place of the fuel salt itself could not be acceptable for the long-term, although it does remain an option for the in-cell equipment. It also became clear that any major operations or alterations need to be performed in the near-term, and that the mid-term was quite acceptable for benign storage of gettered salts. Thus, as a rational scenario was evolved, emphasis was shifted to those aspects that were perceived to be germane to the controlling issues and therefore pertinent to the evaluation.

3.4.1 Geologic and Hydrologic Considerations

Initially, before the very long-term (out to be one million years) radiological properties of the fuel salt were calculated, entombment-in-place of the fuel salt itself was given consideration. In this context, long-term geologic and hydrologic factors were briefly reviewed.[20,21] Subsequently, there was no further incentive to pursue this direction. However, the initial review indicated a stable geology, with no tectonic activity in the past 70 to 80 million years, and a low seismic risk (seismic risk zone 2). The nearest significant earthquake epicenters are over 200 miles away, in Missouri and South Carolina. There have been numerous small earthquakes in East Tennessee, but these have been of low or moderate intensity.[25] The latest,[25] on March 27, 1987 was intensity 4.2 on the Richter scale, which is considered moderate. Level 2 is the lowest that can be felt by humans.

Concerning hydrology, the earlier review[20,21] identified a number of areas where further evaluation was desirable, namely:

- Characterization of all earth materials on the site;
- Site design (slopes, placement of fill soils, underdrains, foundations, etc.)
- Site hydrology (water inventory, groundwater flow nets, etc.)

The need for further evaluations of this kind dropped drastically once entombment-in-place of the fuel salt was eliminated as an option. However, four items were pursued further, because they are of potential concern, even for the near-term. These are: more information on building sump pump operation, actual measurements of groundwater radioactivity, an engineering overview of the building drainage system, and an evaluation of flooding potential.

When this evaluation was started, it was learned that the building has a sump, to remove any water accumulation from the foundation, and to pump out internal floor drainage. (Partial flooding occurred one time, during the very severe winter of 1984-5, when water supply lines froze and then ruptured.) There are two sump pumps: the West Pump, at elevation 814', and the East Pump, several feet higher. For this evaluation, a run-time clock was installed on each pump. In addition, for overall safety, an alarm signal was added that would actuate if the East Pump came on, since this condition would indicate either very heavy flow of water (West Pump unable to handle) or loss of redundancy (West Pump not functional). The results of clock readings are tabulated in Table 8, covering a full year. The East Pump never came on. The West Pump apparently operated every day, with operating times of 0.5 to 2.0 hours/day, and a yearly average of 0.88 hours/day. The pump was calibrated and the rate determined to be 2000 gal/hour. The daily/weekly readings were correlated with weather conditions, and the high/low rates matched quite well with high/low rainfall. However, even prolonged dryspells required pump operation of 0.5 to 0.7 hours/day, more-or-less confirming the reported presence of a year-around spring under the building.[26] On the other hand, the relatively limited increase in pumping time during periods of heavy rainfall indicates that the sump does not receive appreciable quantities of roof water or surface drainage.

Table 8. Operational data on West sump pump

Date of observation	Elapsed days	Clock hours	Incremental hours
8-19-85	-	292.6	-
8-20-85	1	294.0	1.4
8-21-85	1	294.9	0.9
8-22-85	1	295.9	1.0
8-23-85	1	296.5	0.6
8-26-85	3	301.0	1.5/d
8-27-85	1	302.4	1.4
8-28-85	1	303.8	1.4
8-29-85	1	304.9	1.1
8-30-85	1	305.7	0.8
9-03-85	4	309.8	1.0/d
9-05-85	2	311.3	0.8/d
9-06-85	1	312.1	0.8
9-13-85	7	315.0	0.4/d
9-20-85	7	320.4	0.8/d
10-02-85	12	329.0	0.7/d
10-11-85	9	335.5	0.7/d
10-18-85	7	339.8	0.6/d
10-25-85	7	344.8	0.7/d
11-01-85	7	349.8	0.7/d
11-08-85	7	358.3	1.2/d
11-29-85	21	378.0	0.9/d
12-14-85	15	395.8	1.2/d
12-27-85	13	406.9	0.8/d
1-03-86	8	413.0	0.8/d
1-20-86	17	427.2	0.8/d
2-11-86	22	450.6	1.1/d
2-21-86	10	470.9	2.0/d
3-07-86	14	486.3	1.1/d
3-21-86	14	504.6	1.3/d
4-14-86	24	525.3	0.9/d
5-02-86	18	538.1	0.7/d
5-07-86	5	541.8	0.7/d
5-16-86	9	547.6	0.6/d
5-29-86	13	557.5	0.8/d
6-06-86	8	563.0	0.7/d
6-11-86	5	566.2	0.6/d
6-13-86	2	567.5	0.7/d
6-20-86	7	571.2	0.5/d
7-11-86	21	582.3	0.5/d
7-16-86	5	586.2	0.8/d
7-18-86	2	587.4	0.6/d
7-25-86	7	591.1	0.5/d
Total time	340	298.5	
Overall average			0.88/d
Minimum rate observed			0.5/d
Maximum rate observed			2.0/d

Apparently the radioactivity of the sump discharge had never been measured. Therefore, this was done first weekly and then monthly, over the same one-year period that sump pump timer readings were taken. Grab samples of discharge water were submitted for beta-gamma analysis. No readings above normal background (200-250 counts) were ever observed. Considering the relatively large flow of sump water, this is a rather insensitive test. Nonetheless, the results obtained are the most favorable possible under the circumstances and indicate no radioactive contamination within the limits of our detection sensitivity.

The building drainage systems were reviewed in 1985 and a description was prepared.[21] This included the overall description, including key discharge elevations, the radioactive liquid waste collection system (which drains into a large tank), the building water collection system (which drains to the building sump), the storm water system (which drains to a catch basin), and the sanitary sewage system (which drains to a septic tank). No problems or potential problems were uncovered during this review. Each of the hot cells contains a sump, from which any water is jetted to the radioactive liquid waste tank. The reactor and drain cells have air jets (to avoid introducing water into these two cells), while the other cells have steam jets. The cell sumps are monitored. No water has ever accidentally entered these cells.

The flooding potential of nearby buildings and facilities have been examined as part of their safety analysis review,[27] since these facilities include a reactor (HFIR, Building 7900) and transplutonium processing facilities (TRU and TURF, Buildings 7920 and 7930). The predicted probable maximum flood crest is 794 ft (above sea level). The MSRE cell floors are at elevations of 814 ft (drain tank cell) and 816 ft (reactor cell), well above the probable flood crest.

3.4.2 Evaluation of Cell Penetrations

Initially, when long-term entombment-in-place of the fuel salt was being considered as a viable option, the condition of the cell penetrations and their projected long-term lifetime was potentially of great importance. There are over 700 penetrations of the drain, reactor, and processing cells, although most of these are for electrical lead wires. The three cells - drain, reactor, and processing - must be treated as a unit in terms of containment, since they are joined by openings through the cell walls (for salt transfer lines, mainly).

Three studies of these penetrations were conducted. The first[20] was a systematic cataloging of the various categories of penetrations and their locations. The second[21] was a detailed description of the physical construction of each of these categorical types and a preliminary classification of options for improved closures (for long-term applications). By the time of the third study (Appendix C in this report) it was apparent that only near-term needs would have to be met, for which the original constructions would be adequate, unless they have suffered damage since shutdown in 1969. Therefore, an actual visual inspection was performed (at the outer cell walls), and each penetration was inspected. Fourteen classes of penetrations were examined. All penetrations were found to be in good condition, with no signs of corrosion. Three recommendations were made:

- to improve the instrumentation for measuring cell pressure;
- to cap the water lines to the drain cell space cooler;
- to cap the air supply line to the reactor cell jet pump.

All of these recommendations are to provide additional safety factors; e.g. cutting and capping water supply lines, rather than relying on a valve (or even two valves in series). All other water lines were cut and capped prior to this evaluation.

3.4.3 Recommended Facility Improvements

The physical condition of the facility was thoroughly examined, with the objectives of identifying areas where a potential weakness might exist, or where improvements could be made. The cell walls contain numerous penetrations, most of them electrical (for power, controls, and instrument readouts). There are also some that provide air, water, inert gas, or other material flows. These penetrations were carefully cataloged, categorized as to type and function, and the closure methods identified that had been utilized.[20,21] Based on this review, it appeared that virtually all of the penetrations were adequately sealed. For example, water lines had been cut or disconnected and capped off, rather than relying on a valve; electrical penetrations had been designed and installed with multiple seals. When first constructed, these cells were pressure-tested and found to hold 15 psi (gage), which verified that all of the penetrations were leak-tight as

installed. However, it was deemed necessary to carry out an actual physical inspection to (1) verify the data as recorded in the penetrations catalog, and also to (2) visually confirm the condition of the penetrations (regarding corrosion, physical damage, etc.). The results of this inspection, which was conducted by an experienced engineer formerly associated with the project and therefore familiar with the facility, are described in Appendix C.

The inspection and evaluation resulted in these conclusions:

- The reactor cell and drain tank cell penetrations are in good condition and provide acceptable containment; no corrosion problems could be identified.
- Several lines which are valved off should be capped.
- The two instrument systems used to monitor pressures in the drain tanks and in the drain tank cell should be improved by adding isolation valves (for times when the instruments are not being used).

3.5 AVAILABLE OPTIONS

At the beginning of this technical evaluation, six broad options were identifiable in principle (Table 9).[20,21] However, there was not enough known about the system to make a definitive choice, although some options were obviously less desirable than others (e.g., because of cost/complexity). For those options that appeared attractive, there were not enough hard data at the time to make these selections, but this did serve to identify the more urgent data needs (e.g. identification of a gettering agent for fluorine).

The six options are not equivalent in terms of end-point, since options 1, 2, and 3 are "final" while options 4, 5, and 6 are "interim." This status derived from the fact that a "final" decision could not be made at the time based on known facts and the "interim" options were therefore forced upon us. The interim options still require a final

Table 9. Options for the decontamination and decommissioning the MSRE

Option	Action to be taken
0	Continue as is, with no decision. Requires continued surveillance and maintenance, introduces maximum uncertainty, and eventually still needs a decision between the "real" options outlined below.
1	Complete dismantlement of hot cells following removal of fuel and flush salts.
2	Entombment of reactor and drain tank cells following removal of fuel and flush salts.
3	Entombment of reactor and drain tank cells, with fuel and flush salts in-place.
4	Enhanced near-term storage of solidified salts (which still leaves all other options available).
5	Enhanced storage in-place, including remelt and addition of getters (which still leaves options 1, 2, 3, and 6 available).
6	Enhanced storage in-place, including remelt, addition of getters, and repackaging (which still leaves options 1, 2, and 3 available).

option selection. With our present knowledge of radiolysis and gettering, we now have a technical basis for making selections from these options in terms of near-term actions that will support final disposition of these fuel and flush salts.

Option 3 was in fact eliminated as soon as the long-term radiological properties of these salts was identified. If fission products were the controlling radioactivity, this option might be possible, and would be very attractive because of simplicity and low costs. However, the presence of the ^{233}U (and the transuranic species) is a source of very long-term alpha activity that requires truly permanent disposal away from the biosphere. Eventual disposal of these controlling nuclides

could be either a spent fuel/high level waste repository or a TRU waste repository (since the thermal output is very low) or even intermediate depth disposal if it becomes available. In an institutional sense, the present state of uncertainty regarding permanent disposal precludes a final selection in this regard. However, the nature of the waste acceptance criteria for a high-level repository[29] and TRU waste repository[30] are known and can be dealt with for this material. A variant on permanent disposal is to blend the MSRE salts with other waste streams, such as fluoride-bearing high-level waste.

Options 1 and 2 are variants of the same basic "final" option: remove the fuel and flush salts, then either dismantle the cells or entomb them (and their contained but now empty equipment) in concrete. The fundamental problem with these options is that removal of the salts is open ended, as discussed above. There is no site at present that can receive them. Thus, we are forced to some form of interim storage, which should be enhanced as necessary to meet two objectives: to not compromise the continued safe storage of these salts, and to prepare them for final disposal.

Options 4, 5, and 6 are variants on the enhanced storage theme. They reflect increasing sophistication (and costs) of near-term actions, but they also reflect increasing capabilities in dealing with the future final disposal issue. Options 5 and 6 involve remelting, which requires heating above the melting point of 434°C (813°F). Option 4 merely assures continued safety of interim storage, but provides no basis for final disposal.

Option 5 provides a technical basis for extended interim storage without a requirement for reheating (to limit fluorine accumulation). It also provides a minimal concept for disposal, in that the gettered salts could, in principle, be sent to a repository in their present tanks. However, these tanks are large — about twice the diameter presently considered for either a HLW or TRU repository — so that realization of intact tank disposal is highly unlikely.

Option 6 gets around the tank size problem by repackaging. This is not a simple process in-cell, but it is still much simpler than reprocessing, which involves large quantities of fluorine, condensation and

separation of UF_6 and other volatile fluorides, and conversion of the UF_6 to a solid form, most likely the oxide. Option 6 also provides maximum flexibility for exercising of future options. It eliminates only one option: final disposal in the intact tanks, which is highly unlikely to become viable.

Repackaging into smaller containers, along with addition of a fluorine getter while molten, provides these benefits:

- Interim storage can be continued for an extended time period, without the need for reheating periodically.
- During this extended storage, one or more test packages can be carefully monitored, examined, and tested to verify the efficacy of the fluorine getter.
- Once repackaged, the salts are easily transportable to other on-site locations, should that be desirable, and to off-site locations, once that becomes possible.
- By repackaging in corrosion resistant metal (i.e. a nickel-based alloy) the salts would not require additional repackaging thereafter. The individual packages would be small enough that overpacks could be easily added.
- After repackaging, the salts could be removed from Building 7503 (to another on-site location, such as our remotely-handled TRU waste underground storage area), leaving the hot cells and the building available for other purposes.
- By repackaging into canisters that contain about 20 kg each, the fissile content per package would be less than 200 g, the present limit for disposal in WIPP. This would entail about 200 canisters for the fuel salt. The flush salt could be packaged in larger canisters, if desired. The 20 kg canisters would have a contained volume of about 10 liters each (about 3 gal each).
- By including a threaded port, sealed with a removal plug, in the lid of the canisters, the salts could be easily removed from the canisters at some future time by leaching or melting, should this be desired.

4. CONCLUSIONS

1. Calculation of the long-term radiological behavior of the MSRE fuel (and flush) salts shows that these materials must eventually be disposed of in a permanent repository or equivalent.
2. Analysis of the existing containment shows that continued interim storage is acceptable, with only minor enhancements. Extended storage, for 20 or more years, is also feasible, with additional improvements.
3. Laboratory-scale development work has identified a suitable getter for fluorine, whose efficacy is actually increased in a radiation field. This makes possible extended storage without the added burden of periodic reheating. It also makes possible final disposal in the fluoride form, thereby precluding reprocessing, which would be a very complex and costly step. Reprocessing also carries with it the increased probability of radioactive and/or chemically hazardous environmental releases and of operator exposure to these hazards.
4. The maximum benefits of getter utilization can be achieved by also remelting and repackaging into smaller packages. This simplifies continued storage, provides a technical basis for extended storage, opens up options for on-site handling, provides time for verification of getter efficacy, allows time for resolution of open questions concerning final disposal, and is amenable to all realistic final disposal options identifiable at present.
5. Therefore, the overall conclusion is to develop a work plan for decommissioning that embodies the concept of remelt and repackage, with addition of fluorine getter.

Appendix A. BIBLIOGRAPHY AND REFERENCES

Appendix A. BIBLIOGRAPHY AND REFERENCES

The MSRE Project is extensively and thoroughly documented by reports, drawings, photographs, and miscellaneous documents. A computerized listing of these has been prepared by Park Owen and Nancy Knox, of the ORNL Remedial Action Program Information Center. There are approximately 1450 entries in this file, categorized as follows:

Type of entry	No. of entries
Progress report	~140
Technical report	
CF memo	~370
TM report	~240
ORNL report	~ 70
Miscellaneous	14
Journal article	32
Drawing	19
Photograph	~500
Patent	4
Conference paper	~ 50
Correspondence	11
Total	~1450

Copies of most of these documents are in the MSRE file, which is stored in the basement of Building 7503. The Information Center also has copies of many of the reports. These documents are a merger of files previously maintained by individuals who were closely associated with the project. (Unfortunately, several extensive files have been lost over the years.) Most of the photographs were supplied by Luther Pugh. There is also an excellent collection of construction drawings maintained by Martin Marietta Energy Systems, Inc., Engineering on file in Building 1000.

A relatively small number of reports have provided all the background information used for this study. A listing, in chronological order, is provided on the following pages. The chronological numbers are used as reference call-outs in the main body of the report.

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18. F. J. Peretz, Preliminary Decommissioning Study Reports — Volume 5: Molten Salt Reactor Experiment, X-OE-231, Vol. 5, September 1984.

19. T. E. Myrick, The ORNL Surplus Facilities Management Program Long Range Plan, ORNL/TM-8957, September 1984.

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29. Waste Acceptance Preliminary Specifications for the Defense Waste Processing Facility High-Level Waste Form, OGR/B-8, January 1986.

30. TRU Waste Acceptance Criteria for the Waste Isolation Pilot Plant, WIPP-DOE-069, September 1985. See also Appendix C in reference 20.

*The left-hand numbers are used as call-outs in the main body of the report.

Appendix B. Status Report on Recent Radiolysis Studies

AN EXPERIMENTAL STUDY OF RADIATION EFFECTS ON MSRE FLUORIDE SALT MIXTURES

L. M. Toth and L. K. Felker

March, 1987 Status Report

INTRODUCTION

The MSRE fuel salt is presently stored in two drain tanks until a more permanent disposition of the salt can be arranged. The presence of the numerous radioactive fission products in the fuel salt mixture is a matter of some concern because it is known that radiolysis of halide salts produces defect centers with the resultant release of halogen gas. This concern in the MSRE storage tanks is that the release of fluorine (from the fluoride salt) may result in excessive pressures of corrosive gas over a long time period.[1]

It is desirable, therefore, to determine the extent of the fluorine pressure build up and means to abate it. Although there is considerable information in the open literature on the radiolysis of halide salts, not much detail is available on irradiations that have been carried to a steady state limit, ie., where the rate of generation in a radiation environment is matched by the rate of recombination of the free halogen with the active metal centers left behind.

An experimental program was initiated to answer some of these questions. The objective of this work was to determine such a limit, if it exists, and the extent to which certain parameters (such as temperature) affect that pressure. In addition, it was desirable to determine if a suitable getter could be added to the system which would react with the fluorine and thereby sequester it through the formation of a stable chemical compound.

Gamma irradiation is the most practical means of experimentally exposing the halide salts to radiolytic damage. It is, by far, the easiest to use and the results described here were obtained only with this form of radiation. In the final analysis, an alpha source is desirable because it is this source of radioactivity that the fuel salt will experience over the longest period of time.

The dose rate in any experimental approach must be significantly higher than that of the actual salt throughout its projected storage period in order to obtain measurable effects within a reasonable time. Relating experimental results to those in the actual case will therefore require an understanding of the fundamental kinetics and mechanism of the reaction.

EXPERIMENTAL

MATERIALS

The lithium fluoride was obtained from Harshaw Chemical Company as single crystal fragments. The zirconium fluoride and uranium fluoride used in this series of experiments were from ORNL stock material remaining from the MSRE program. The beryllium fluoride, hydrogen fluoride, and fluorine were obtained from Alfa Products, Linde Specialty Gases, and Air Products, respectively. The graphite used in this work was POCO EDM-3 and was obtained from local chemical stores. All materials of construction were resistant to attack by both hydrogen fluoride and fluorine. Nickel vessels and tubing and monel valves, fittings, and gauges were used where possible. A manifold was constructed of resistant materials for use in gas handling and sample preparation. A controlled atmosphere glovebox was used for storage and handling of salt mixtures. A helium atmosphere was circulated through molecular sieves (13X Linde) to remove moisture. A moisture content <10 ppm was maintained in the glovebox.

METHODS

Fluoride Salt Preparation - Two fluoride salt mixtures (200 g each) were prepared for irradiation. The first mixture contained lithium fluoride, beryllium fluoride, and zirconium fluoride (64.6, 30.4, 5.0 mole %): the second contained lithium fluoride, beryllium fluoride, zirconium fluoride, and uranium fluoride (64.5, 30.3, 5.0, 0.13 mole %). Each mixture was weighed into a graphite tube (4.5 cm dia x 15 cm) and loaded into a nickel vessel which was in a vertical tube furnace.

After sealing the nickel vessel, a helium purge was used to sweep the vessel and associated lines. The furnace temperature was increased to 550°C to melt the mixture. (This is approximately 100°C above the melting point of the salt mixture.) A graphite dip leg was inserted into the melt and hydrogen fluoride was added to the helium carrier and bubbled (or "sparged") through the molten salt mixture.

This process of hydrofluorination should remove traces of moisture and convert any oxide to fluoride while the graphite components are used in contact with the fluoride melt to prevent corrosion of the metal pot and dissolution of these metal fluorides in the melt. The hydrogen fluoride flow rate was determined by titration of the exit gas. The first salt mixture was hydrofluorinated for 8 hours with an average rate of .032 L/min hydrogen fluoride. The second salt mixture was hydrofluorinated for ~20 hours with an average hydrogen fluoride rate of .030 L/min. (Previous experience has established that this amount of sparging is sufficient for reducing the oxide content of the salt to <1 ppm.)

The salt mixtures were then He sparged to free the salt of dissolved HF and allowed to solidify. The vessel was put into the controlled atmosphere glovebox where the salt plugs were removed, scraped of surface graphite film that migrates to the salt interface and stored for irradiation. It is important to remove all graphite from the salt in order to be certain that no graphite/fluorine reactions occur unless intended. (See Results and Discussion)

Fluoride Salt Irradiations - The pulverized fluoride salts were irradiated at the High Flux Isotope Reactor (HFIR) at ORNL. Spent fuel elements which are stored in the reactor pool were used as the gamma source. The gamma flux of the elements varies in the range 1.0×10^8 to $<1.0 \times 10^7$ R/hr depending on the age of the element.[2] The storage racks for the elements contain a four inch cadmium sleeve through the center hole of each element to absorb any neutron radiation. Samples are inserted inside this cadmium sleeve. The active irradiation length is 20 inches.

One concern with using the spent fuel elements was that the heat generated by absorption of radiation from the gamma source might produce too high a temperature in the sample. Prior to the irradiation experiments, a lead weight with two thermocouples embedded was used to measure the temperature of a spent fuel element over a 15 day period beginning on the third day out of the reactor. The temperature due to gamma heating varied in the range 58-41°C over this period but appeared to level somewhat after fifteen days in the range 41-38°C. Elements used were generally within this 45 day period after being removed from the reactor and the temperatures in this range were judged to be acceptable.

A nickel vessel was fabricated by the Plant and Equipment Division at ORNL for use as an irradiation vessel (Fig. 1). The vessel, associated tubing, and gauge were passivated with fluorine prior to loading with the fluoride salt mixture in order that the fluorine liberated during the actual irradiation experiments would not be lost due to reaction with these components. The loaded vessel was lowered into the pool and into the center of a HFIR fuel element. The 1/8 inch tube and thermocouple wire extended to the top of the pool to an appropriate pressure gauge and a Doric thermocouple readout. The reactor pool water circulating around the vessel provided sufficient cooling during the course of the irradiation. Pressure and temperature readings were recorded on a daily basis over the period of the irradiation. Test with gettering agents in the irradiation vessel were conducted in a similar manner.

In order to examine the effect of temperature on the salt irradiation, we sought use of the Analytical Chemistry Division "dry tube" because the temperature in it typically ran at 70°C (due to the absence of the cooling influence of the HFIR pool water). This tube consisted of a four inch stainless steel tube which extended from the top of the pool to the center of a spent fuel element. With this arrangement the nickel vessel containing the salt was lowered into the tube and center of the fuel element. However, the availability of the dry tube for experiments became more limited during the course of our measurements so an alternative was sought. A nickel can was fabricated in order to enclose the sample vessel in a water tight environment and thus permit the latter to be resistively heated in a dry atmosphere. This was accomplished by sealing the sample vessel inside the nickel can by soft soldering the lid and tubing lead-throughs. (Fig. 2). The electrical connections for the heating tape extended through a 1/4 inch tube to the top of the pool where a variac was used as a controller.

Preparation of Getter Materials - Various forms of carbon were tested in the course of finding a suitable getter material. In general, the procedure involved taking a suitably prepared getter and loading it with fluorine at 1-2 atm. pressure. A static pressure of fluorine was applied to the carbon specimen that had been sealed in a nickel pot (such as the one shown in Fig. 1) and maintained at pressure (by subsequent additions) until it was apparent that no further consumption of fluorine by the carbon would occur. This amount of loading appeared as an increase in the weight of the carbon which amounted to a fraction of 1% for the POCO graphite to as much as 60% for activated charcoal. (In the case of the activated charcoal, the reaction was so rapid that low pressures of fluorine had to be admitted initially and gradually built up to the limiting pressure of approximately 1.6 atm over the sample to allow dissipation of the large heat of reaction.

Attempts to "activate" the POCO graphite were made according to a steam activation procedure cited in the literature[6]. This involved heating graphite chunks (produced by crushing a 1/2 inch rod and then sieved to 6-60 mesh) at 800°C for 4 h in a stream of water-saturated helium gas. The graphite was heated on a silica frit fused inside a silica tube which, in turn, was positioned vertically in a tube furnace thus permitting the helium and product gases to be vented at the top of the furnace. Because the reaction of water on graphite produces H_2 and CO (the classical water-gas reaction), the combustible products typically ignited on exiting the hot silica tube. The results of the activation process produced a more porous form of the graphite as evidenced by microscopic examination. The net loss of weight due to the activation procedure was 32 and 66%, respectively, for two separate runs.

Fluoride Salt Solubilities - The fluoride salt solubilities were determined by placing approximately 2 g of $LiF-BeF_2-ZrF_4$ (64.6, 30.4, 5.0 mole %) in each of three 120 mL plastic bottles containing 25 mL of aqueous solution. The three solutions that were tested for solubility of the fluoride salts were: (1) H_2O; (2) 4.0 M HF; and (3) 4.0 M HF, 2.0 M HNO_3. These were placed on a wrist shaker and allowed to agitate at ambient conditions (23±1°C). After one day, a sample was taken and sent to Analytical Chemistry Division for an Inductively Coupled Plasma (ICP) scan of the elements and their concentrations. The error in this analysis was no greater than 10%. Additional samples were taken after 6 and 29 days, with more salt addition to solutions #1 and #2 after the 6th day. It was not necesary to make any additions to solution #3 because there appeared to be ample salt in the bottom of the plastic bottle.

RESULTS and DISCUSSION

FLUORINE GENERATION

Even though earlier irradiation experiments have been conducted using a ^{60}Co irradiator, we decided that a much more intense source would be necessary in order to reach a steady state situation (ie., a limiting fluorine pressure) in a reasonable amount of time. The previous study[3] on a ^{60}Co irradiator produced a dose rate in the salt of 0.45×10^{20} eV/h/g for the 35 gram total which, in terms of a flux rate to achieve this, is 0.72 MR/h. Since the previous irradiations took months to generate satisfactory amounts of fluorine, we judged that a more intense source of radiation would be necessary to reach any limiting fluorine pressure in a reasonable amount of time. (The previous study never was carried to a limiting fluorine pressure.)

Therefore, the HFIR spent fuel elements, with a gamma flux of 10 to 100 times the above intensity, was deemed necessary to achieve the limits which we sought. Nevertheless, during the early stages of this work, an experiment on the local ^{60}Co irradiator[4] was performed---mainly to insure that our calculations were correct and controlled experiments were possible in both the irradiator and, especially, in the HFIR spent fuel element. The experiment in the ^{60}Co irradiator showed little, if any, fluorine pressure buildup over an irradiation period of a week in a gamma flux of approximately 0.6 M R/h, convincing us that the much higher fluxes as expected in the HFIR fuel element would not produce disastrous effects.

Several irradiation experiments in HFIR fuel elements have been performed to date at approximately 40°C. The first of these involved 30 g of MSR composition salt in the nickel sample pot. An induction period of about 6 days was observed where there was a slight decrease in the total pressure in the pot followed by a slow increase in the pressure that continued for two weeks following the induction period. Indeed, a higher rate of fluorine pressure generation was expected (by extrapolating the rates previously reported[3] for the fluxes in the ^{60}Co irradiator to values as found in the HFIR spent fuel element) and therefore the experiment was interrupted to examine the contents for any problems. None of any significance were found.

A second experiment was begun to repeat the first one with better control and with more thorough fluorine passivation of the container since it was suspected that the slight drop in pressure during the induction period might be due to further passivation of the container in the radiation field. The results of this experiment are shown in the following two figures which present the data in terms of fluorine pressure as a function of days in the spent fuel element. The first of these figures, Fig. 3, shows the fluorine pressure as it was actually measured with a 15 psig maximum reading pressure gauge. When the fluorine pressure reached the 15 psig limit, it was bled off to 5 psig and allowed to accumulate again. In this manner, a sawtooth appearance of the pressure with time was produced. The pressure continued to build up at a rather steady rate of 0.027 lbs/h until after approximately 100 days, it leveled off.

The same data are presented in Fig. 4, but this time the net pressure as a function of time is plotted. In this figure, it is more obvious how the fluorine generation progressed, regardless of the necessary bleed-off operation. It can be seen that there is a rather constant rate of generation up to the 50 psig limiting pressure. From information in the open literature, it is believed that the bleed-off procedure had little effect on the rate of generation or the limiting value of 50 psig since these factors are more controlled by the situation within the salt lattice itself.

The gamma flux for the particular fuel element is also given in the figure in an effort to demonstrate the effect of flux on the rate of fluorine generation. As can be seen by the discontinuities in this flux curve, several fuel elements were used for the irradiation in an effort to maintain the maximum possible flux. These fluxes have been recently remeasured and documented.[2] Even though there is a noticeable change in the generation rate with changes in the gamma flux, it is not a linear relationship, thus confirming our earlier observations that the rate was not 10-100 times greater in the HFIR fuel element than in the ^{60}Co source even though the fluxes varied by that amount.

Also included on the figure is a scale showing total percent damage to the crystal lattice (as determined by computing the total moles of fluorine appearing in the sample gas space, 40 mL, and subtracting this from the total moles of fluorine available from the 30 g of salt). The limiting value of approximately 2% is greater than that which has been reported for other halide salts using similar, but less intense sources of radiation. (Previously, we had stated that the limiting value was "consistent with that which had been reported"; but recalculation of that reported data revealed, instead, a 0.02% limit.)

The steady-state limit, whatever the actual value, represents a point where the number of defect centers (containing active metals such as Li or Be) are so great that the rate of fluorine atom recombination with them is equivalent to the rate at which the fluorine atoms are generated. Therefore, in the higher radiation field such as available to us, the total amount of radiation damage at steady state should be considerably greater. In addition, the amount of radiation damage at steady state in the actual MSR fuel salt would be considerably less because the radiation density in the stored fuel salt is very much lower.[1]

We believe that the 50 psig pressure limit is dependent on the volume of the gas space above the irradiated salt. If the volume over the salt had been larger, then the limiting pressure would have been smaller. The determining factor on the limiting fluorine pressure should be most dependent on the concentration of defect sites produced. When these sites reach approximately 2% of the salt content for the radiation flux used here, no more generation and release of fluorine should occur. In view of these results and the lower radiation fluxes available in the MSR fuel salt, we conclude that the 50 psig fluorine pressure limit reported here represents the maximum pressure that would ever be possible over the stored fuel salt.

Attempts to reproduce these data have been only partially successful. It is impossible to leave the irradiation vessel undisturbed during the long 100 day term of the experiment because necessary changes in the spent fuel elements must be made by a variety of operations personnel. As a consequence of the handling, leaks frequently appeared

in the system and compromised the validity of the results. Our last attempt to measure the limiting fluorine pressure was interrupted (because of the HFIR shutdown and resulting interruption in the supply of spent fuel elements) after the fluorine pressure had reached 8 psig.

We have also conducted similar irradiations of fluoride salt samples at higher temperatures --- namely in the 70-100°C range. Under no circumstances did we observe any accumulation of F_2 over the salt at these temperatures. Even though the F_2 recombination rate is expected to equal that of the generation at 150°C[3], it is apparent from our results that the recombination rate had approached that of the generation. Nevertheless, these experiments at higher temperatures should be repeated should a high gamma flux source become available again.

FLUORINE RECOMBINATION WITH SALT

After the fluorine pressure reached the limiting value in the above experiment, we sought to determine the recombination characteristics of the fluorine with the salt. We decided that the best way of examining this was by removal of the sample pot and associated tubing and gauge from the fuel element radiation source and, without opening the container, heating the sample pot for a period of time at ever-increasing temperatures. These results are shown in Fig. 5 for data taken at 100, 150 and 200°C. These give an activation energy of 8.74 Kcal/mole for recombination which is approximately 3X less than that reported by Haubenreich.[5] Even though there would be a finite rate of recombination at 40°C, we compute from the least squares parameters that it would be only 5×10^{-4} psig/h. These data further predict that the temperature at which generation would equal recombination is 150°C --- a value which was suggested in some of the earlier MSR experiments.[3]

FLUORINE GETTERING EXPERIMENTS

During the course of the above experiments, it was realized that the high degree of reactivity of fluorine for graphite might be put to practical use as a gettering reaction. Graphite forms an intercalation compound with fluorine of approximate C_2F stoichiometry at near room temperature. (Higher fluorine contents are possible at higher temperatures.) The most attractive aspect of using graphite as a gettering reagent is that is it most compatible with the MSRE system (having been used extensively in it as a moderator during reactor operations).

We have tested the prospect of using graphite by first loading it with fluorine outside of a radiation environment and then exposing the fluorine-laden graphite to the HFIR gamma flux to determine if the intercalation compound thus formed was stable. A 19 g cylinder of thoroughly outgassed graphite (600°C in a vacuum) was loaded with 0.2 g of fluorine and then moved to the HFIR fuel element in the standard sample pot fitted with a pressure gauge. After two weeks irradiation, no increase in the pressure of the sample pot system was observed that could be attributed to the liberation of fluorine when as much as 23 psi could appear if all of the 0.2 g of loaded fluorine had been driven off.

In an attempt to increase the fluorine capacity of the graphite, we have sought to activate it through a standard steam activation procedure[6]. In spite of a very obvious increase in the porosity of the graphite (as evidenced by microscopic examination), we saw very little increase in the capacity of the graphite for F_2. (Two test samples, one activated and one not activated, showed less than 1 % weight increase --- the activated sample showed 1% increase, while the non-activated showed almost no gain in weight. Nevertheless, both of the samples were reactive to fluorine as shown by a measureable F_2 pressure drop when it was left in contact with the graphite. Very little rise in temperature, however, was noted for either of these samples.

In an attempt to find a carbon form which had a higher capacity for F_2, we tested activated charcoal, as received from the chemical supplier. When it was exposed to F_2 at room temperature, a very exothermic reaction occurred as demonstrated by a sharp rise in the temperature to 30-50°C. The reaction was so rapid that only small amounts of F_2 could be added at a time (an initial F_2 pressure over the charcoal of only 1/10-1/5 atm.) and gradually built up until 1.5 atm. pressure could be left in contact.

We have found that the charcoal samples exposed to F_2 in this fashion experienced a 60% increase in weight --- indicative of the formation of a C/F compound of 2/1 to 3/1 ratio. The F_2 appeared to be irreversibly bound to the charcoal because attempts to pump it off were unsuccessful. The rapid reaction rate and high capacity of the charcoal for F_2 accompanied by the stability of the resultant product led us to consider the activated charcoal more seriously as a getter material. One of the most crucial requirements, however, was the stability of the carbon fluorine product in a radiation field since if it were not stable when exposed to radiation, it would not be satisfactory for actual use with the fuel salt.

To test the radiation stability of the charcoal/fluorine compound, we placed 2 g. of charcoal that had been loaded with 1 g. of F_2 (50% weight increase of sample observed) in the irradiation vessel of Fig. 1 with a F_2 overpressure of 7.0 psig. This system was then placed in a HFIR spent fuel element (flux of approximately 3×10^6 R/h; temperature of 30-35°C). We expected to observe a pressure increase if the charcoal/fluorine compound were unstable and no increase in pressure if it were stable.

The results of the radiation exposure are shown in Fig. 6. An unexpected decrease in the F_2 pressure has been observed indicating that the presence of radiation <u>enhances</u> the reactivity of fluorine with charcoal (most probably through the production of F atoms which would be even more reactive toward the charcoal). Although the limits of the radiation experiment shown in Fig. 6 have not been reached at this time, the present results suggest that activated charcoal is an ideal getter for any fluorine released from the fuel salt.

Assuming a 50% loading capacity as described above and a maximum of 2% damage to the fuel salt, it would take 65 Kg. of activated charcoal over the 4650 Kg. of fuel salt to completely getter the liberated F_2. With a density of 0.531 Kg/L, the charcoal would occupy 244 L in volume. Since the salt storage tank has a void volume of 1331 L over it[1], there is ample space within the present fuel storage tanks to accomodate the required amounts of activated charcoal.

FLUORIDE SALT SOLUBILITY IN AQUEOUS SOLVENTS

In anticipation of reprocessing the fluoride salts as a means of separating fission products, we have examined the solubility of $LiF-BeF_2-ZrF_4$ (64.6, 30.4, 5.0 mole %) in (1) H_2O; (2) 4.0 M HF; and (3) 4.0 M HF, 2.0 M HNO_3. The solubilities of LiF, BeF_2 and ZrF_4 in H_2O at 23°C after 29 days of equilibration were: 5.58, 2.73 and 25.01 g/L, respectively. Likewise for the 4.0 M HF these were: 2.94, 5.01 and 10.40 g/L; and for the 4.0 M HF, 2.0 M HNO_3 these were: 3.33, 2.47 and 4.21 g/L.

The high solubility of ZrF_4 in the less acidic solutions is to be expected since it has the greatest tendency to hydrolyze and form hydrous aggregates in solution. Conversely, in solution #3 its solubility is considerably more like the others.

CURRENT AND FUTURE EXPERIMENTS

A commercial fluorine detector which is capable of detecting fluorine levels up to 100 ppm has been purchased. Calibration checks of it prior to installation in the MSRE facility has revealed satisfactory performance up to approximately 30 ppm. We are presently trying to get the supplier to meet the design specifications of 100 ppm. It is anticipated that a gas stream from the MSR storage tank environment will be routed through this detector to monitor the F_2 pressure. If it is greater than 100 ppm, we plan to sample with standard metal vacuum vials (designed for this purpose by the K-25 analytical group) and send the samples to them for mass spec analysis.

We are currently laying plans for irradiation experiments using a hot alpha source such as ^{238}Pu in order to establish that the results obtained from the gamma experiments are the same as those which would be found using an alpha source. The very elaborate precautions that must presently be exercised in using this kind of material has delayed more timely progress in this area. A safety summary for this work has been approved and the experiment is currently being set up.

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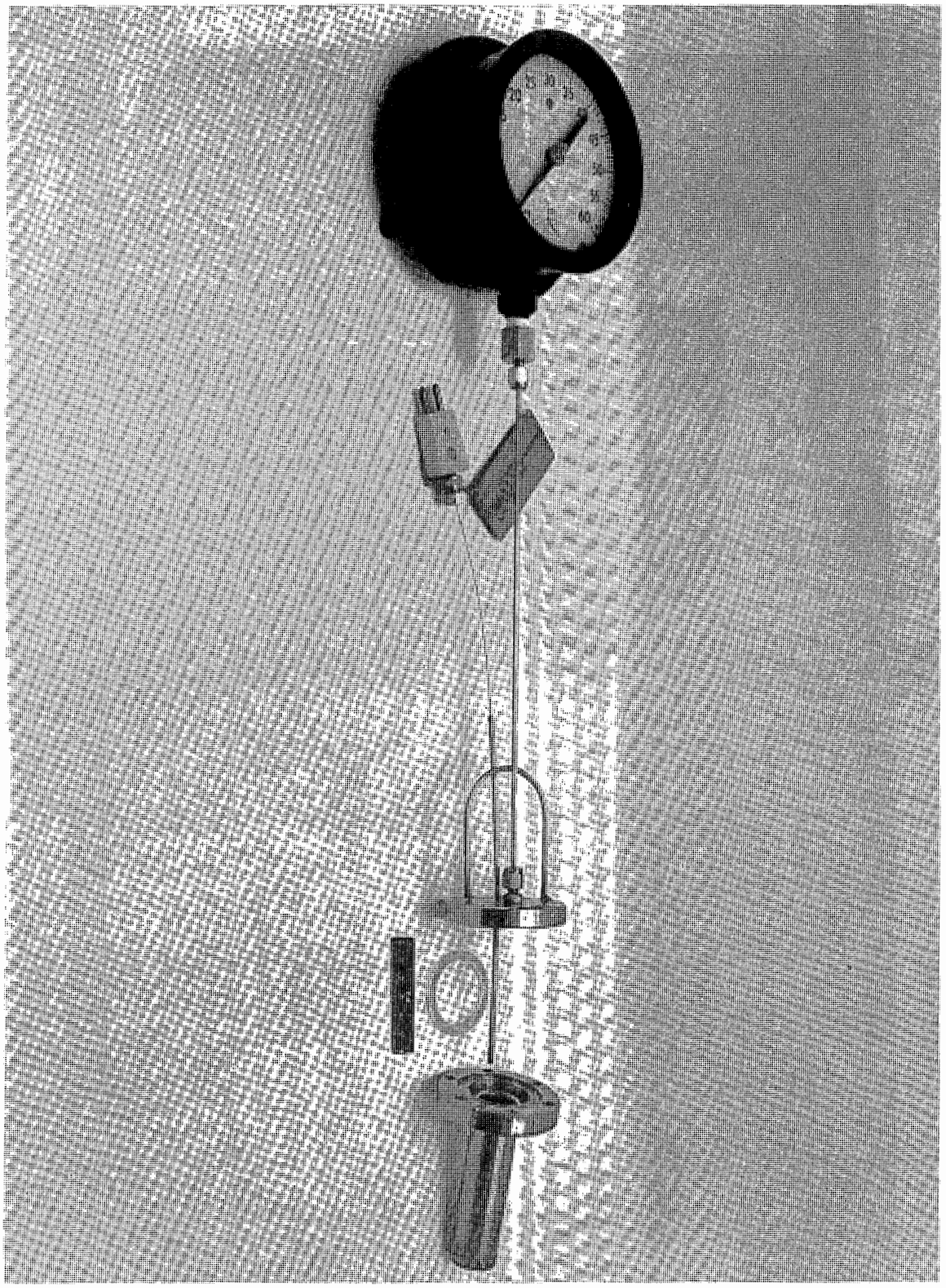

Fig. B.1. Photograph of vessel (sample pot) used for salt irradiations.

ORNL DWG 86-692

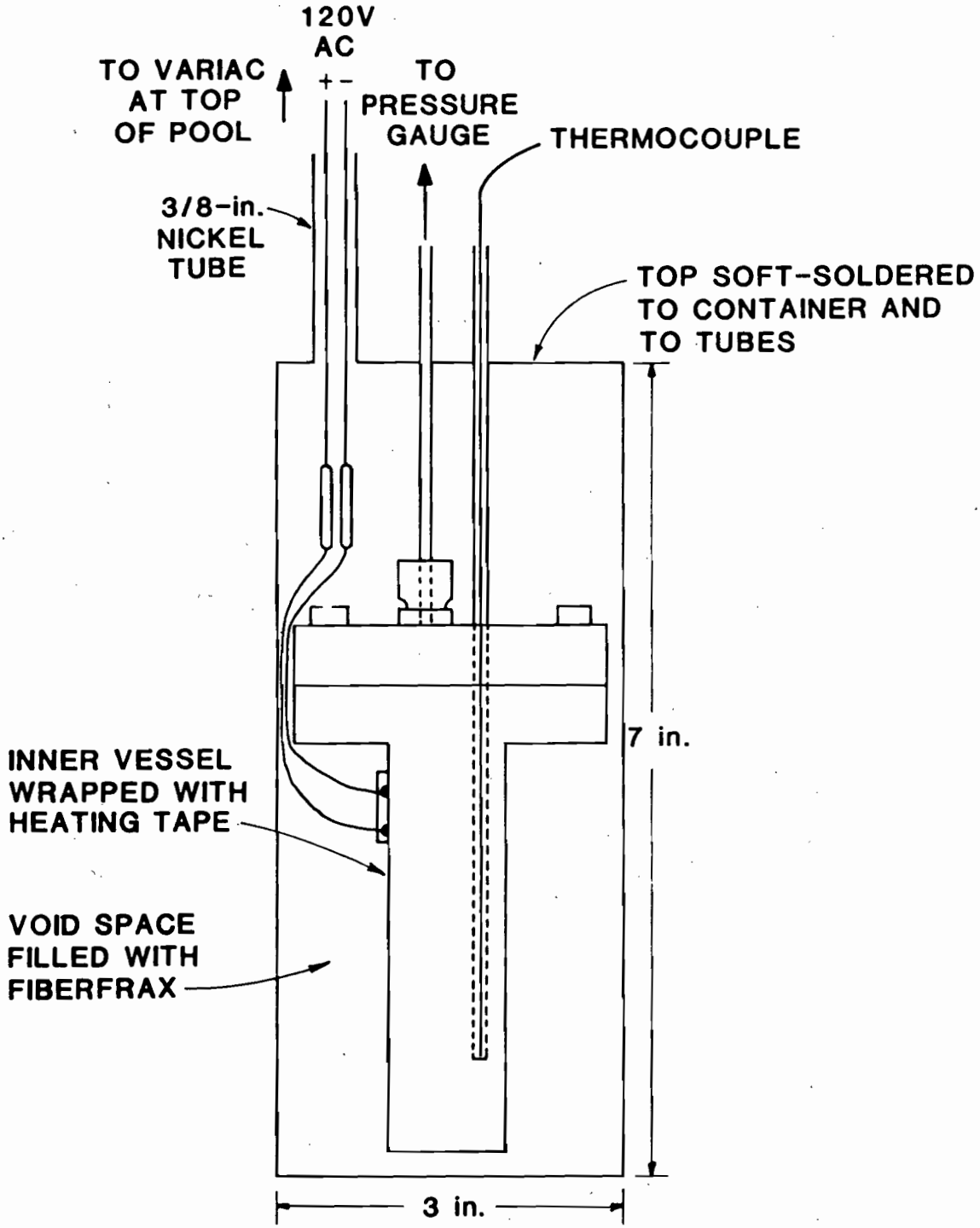

Fig. B.2. Sketch of resistively heated vessel in sealed container.

ORNL DWG 86-693

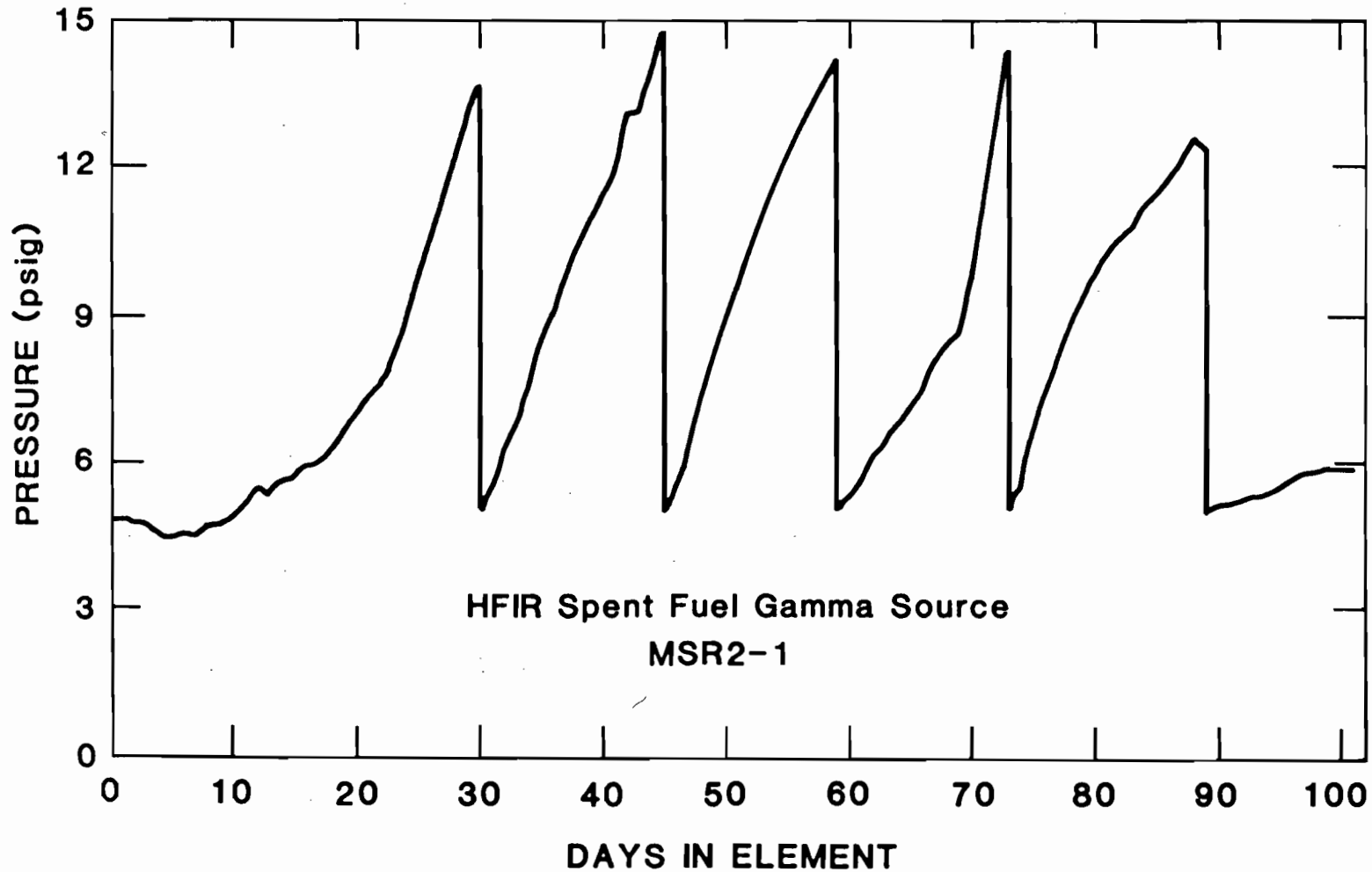

Fig. B.3. Pressure vs. days of irradiation for fluoride salt mixture showing the bleed-off steps.

ORNL DWG 86-912

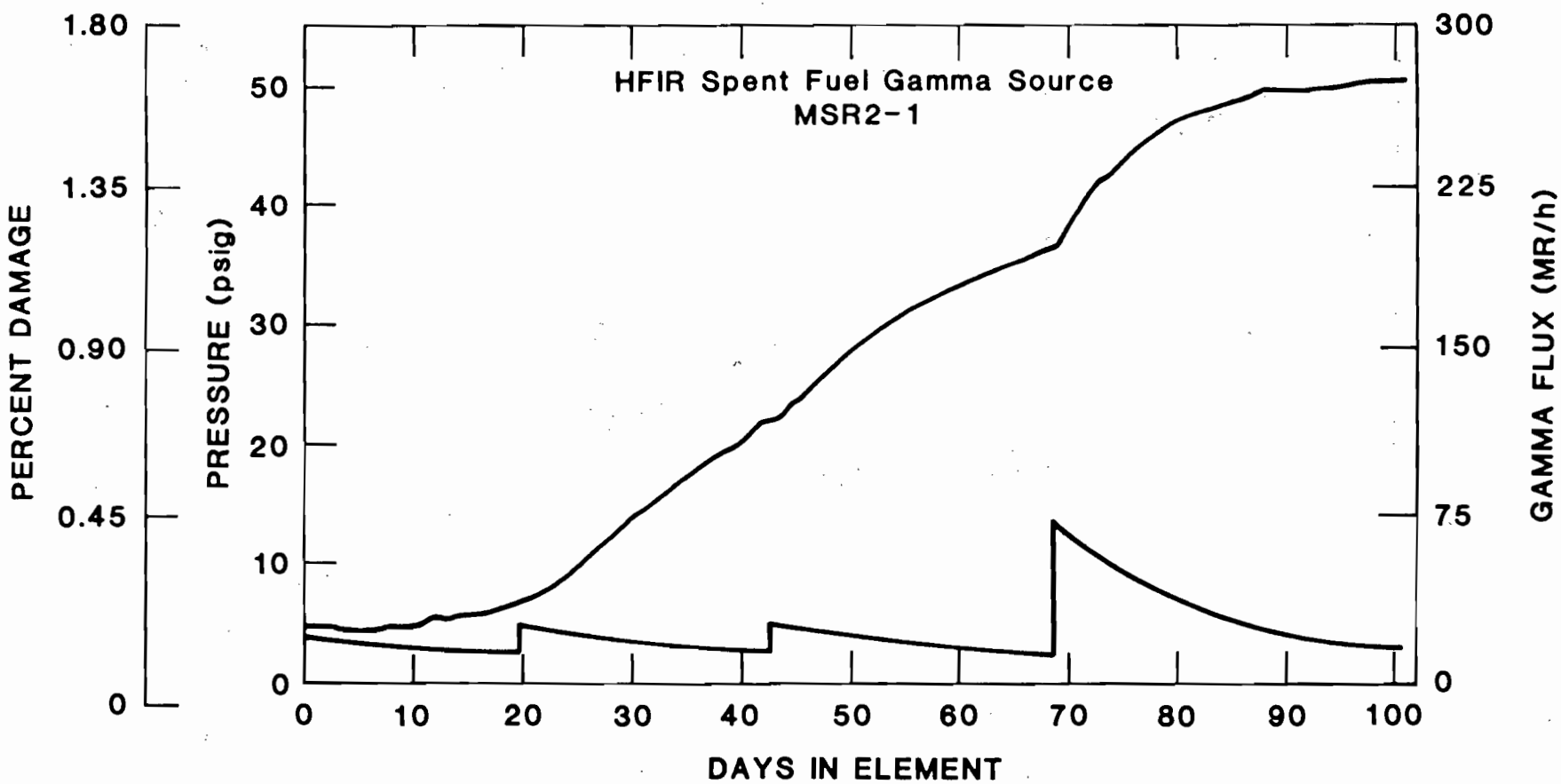

Fig. B.4. Net pressure increase vs. days of irradiation for fluoride salt mixture (the gamma flux is also shown).

ORNL DWG 86-911

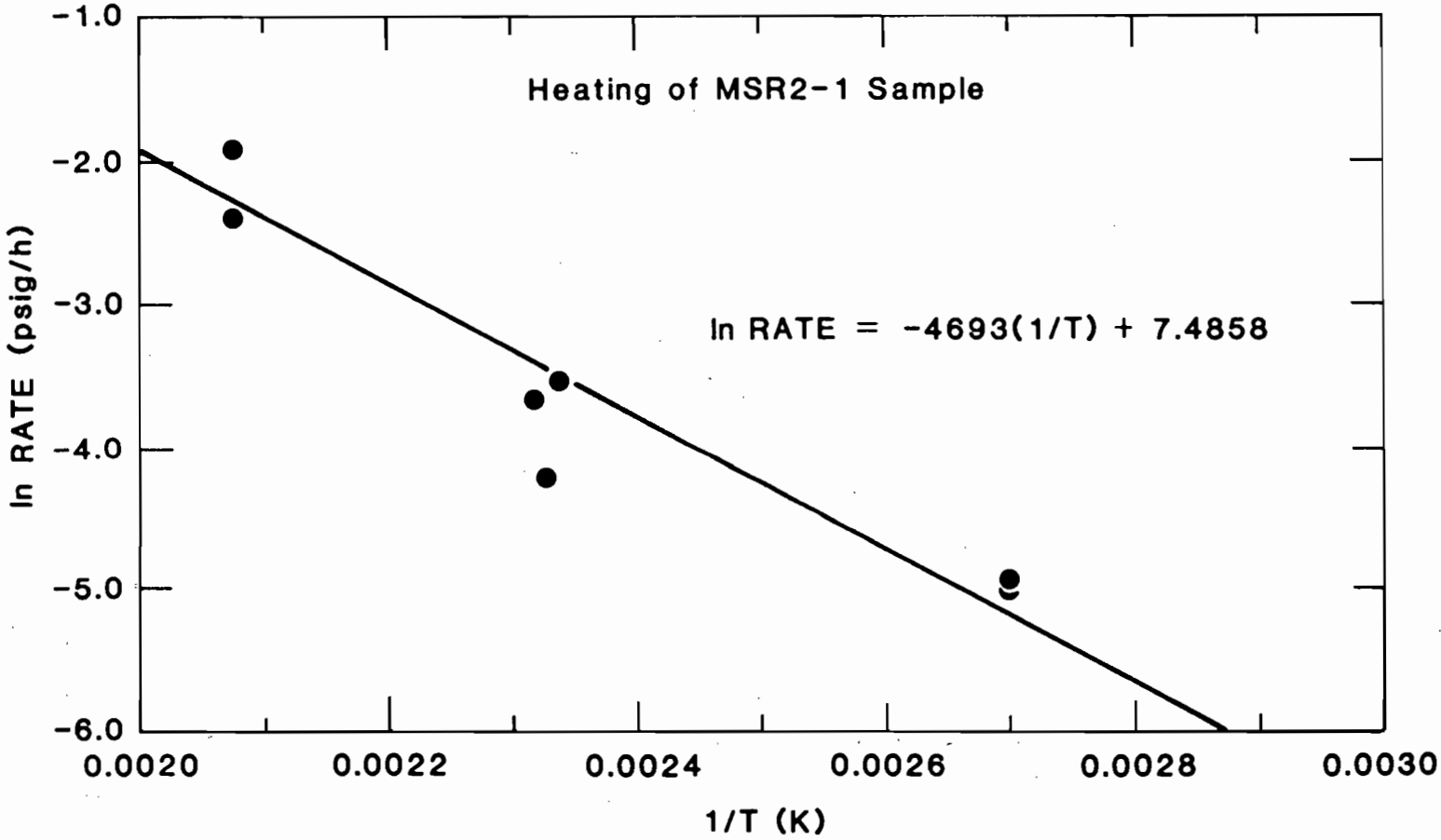

Fig. B.5. Arrhenius plot for fluorine-salt recombination.

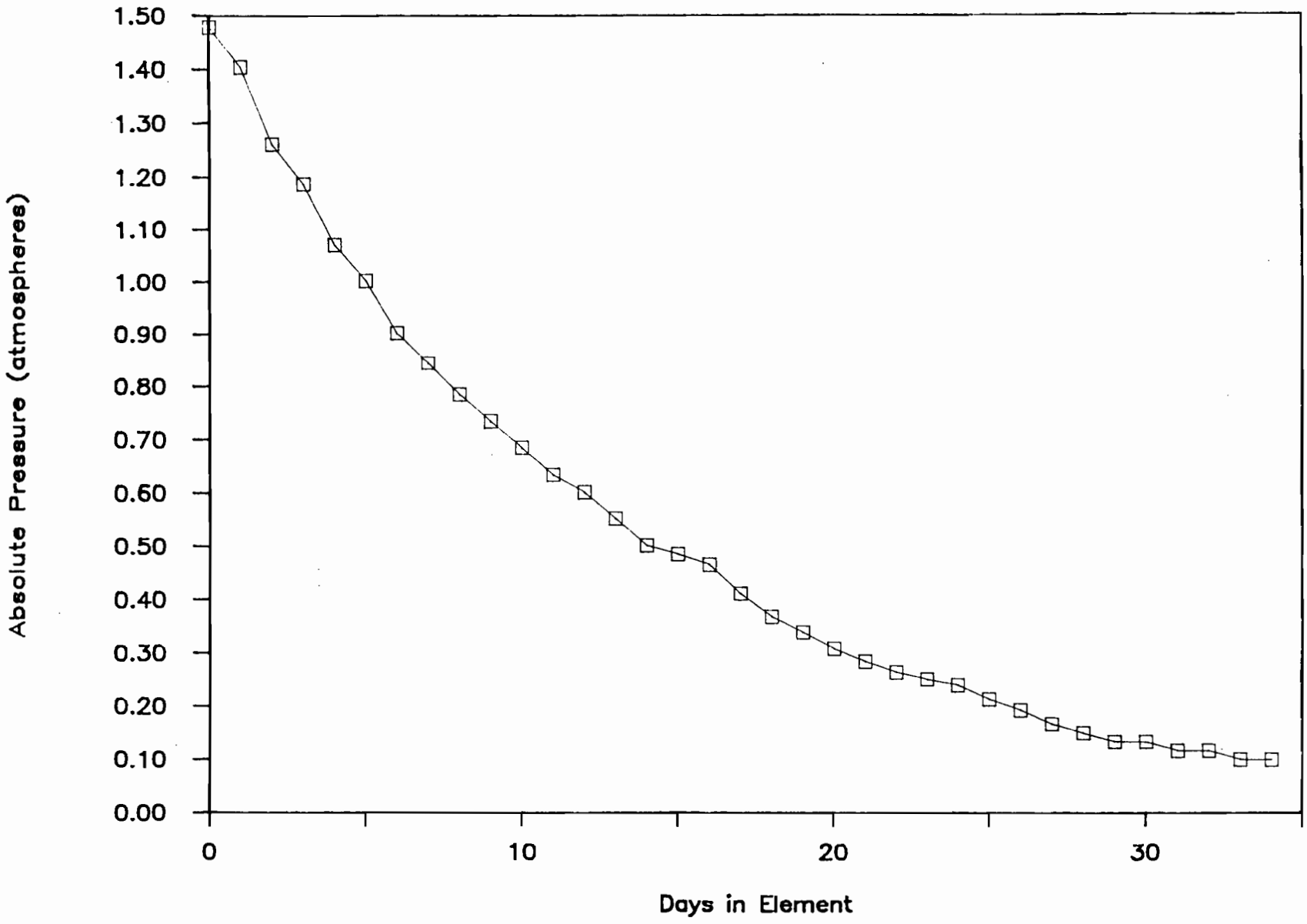

Fig. B.6. Pressure drop during irradiation of fluorine-loaded charcoal.

APPENDIX C: EXAMINATION OF MSRE PENETRATIONS

CONTENTS

Examination of MSRE Penetrations

R. H. Guymon

1. Introduction

The MSRE was shut down in 1969 and in 1971 was put into condition for short-term storage of the fuel salt. Periodic checks of reactor cell and drain tank cell leak rates since then have indicated that all of the penetrations are intact. However, in view of the fact that 15 years have now passed, it seemed prudent to visually examine these to verify their physical condition and to recommend any changes that might be needed. This report describes the results of that examination.

2. Background

The MSRE reactor and drain tank cells were designed to withstand the maximum credible accident which postulated that with the reactor at full power, the molten salt would be drained to a pool of water in the reactor cell. Under these conditions the pressure would increase to 39 psig and the temperature would rise to 260 F.[1]

One strength test at 48 psig was made of the cells in 1962 before many of the penetrations were installed. Prior to power operation, a leak test was made at 30 psig and annual leak tests were made at about 20 psig until the reactor was shut down in 1969.[2] After shutdown, annual leak tests were made at about 5 psig.[3] These were well within the limits of 400 standard cubic feet per day. The 1984, 1985, and 1986 leak rates were 290, 204, and 175 standard cubic feet per day. Periodic repairs have been made to the penetrations based on the cell leak rates. In addition, several water lines have been blanked off.

The reactor cell and drain tank cell penetrations are listed in Tables 1 and 2 from the MSRE Design Report.[4] Drawings showing the locations of most of the penetrations were made recently by the Design department.[5] These tables, figures, and the MSRE flowsheets[4] provided useful information during the review.

Descriptive information on the various types of penetrations is given in Part I[4] and Part II A[6] and B[7] of the MSRE Design and Operations Report. Figure 1 [8] shows the major types of penetrations and the methods used for containment.

3. Examination of Penetrations

Each accessible penetration from the reactor cell and drain tank cell was visually examined. No unacceptable corrosion or uncorrected

physical damage was found. Details of the results of the examinations are given below and are separated into related groups. Reasons are given for not inspecting some penetrations.

3.1 Penetration of Lines Connected to the Drain Tanks

There are only 2 lines on each drain tank which penetrate into the fuel salt (or flush salt). (1) The freeze valves (FV-105 and FV-106) between the reactor and the two drain tanks (FD-1 and FD-2) were removed in 1970. The ends of the lines were sealed inside the drain tank cell using tapered INOR-8 plugs held in place mechanically and sealed with RTV cement.[9] Containment is considered adequate. (2) The salt transfer lines to the fuel storage tank have freeze valves (filled with frozen salt) inside the drain tank cell and are backed up by freeze valves in the fuel processing cell. These could not be examined since the fuel processing cell is closed. Freeze valves were used during operation to prevent transfer of salt, and containment is considered acceptable.

One of the two lines which penetrate the gas space of each drain tank is a spare and has been blanked off inside the drain tank cell since construction. The other line on each drain tank separates into three lines. The first is the helium supply line which contains two check valves and is also capped inside containment enclosures in the north electric service area.[9] Pressure transmitters tie into the drain tank side of the check valves. The transmitters are reactor grade and, therefore, containment is considered adequate. A flange in the second line, which was the equalizer line to the fuel pump, has been blanked in the drain tank cell.[9] The third line is jacketed until it gets to the vent house where it is blocked by hand valves. Leakage through the hand valves would present no problem since the gas would discharge through the charcoal beds.

3.2 Other Helium Cover Gas and Off-Gas Lines

All other helium lines into the cells are capped inside containment enclosures. The off-gas lines are jacketed until they get to the vent house where they are blocked by hand valves. Leakage through the hand valves would present no problem since the gas would discharge through the charcoal beds. Containment is considered adequate.

3.3 Instrument and Instrument Air Lines

Most instrument and instrument air lines contain fail-closed block valves on the supply and vent lines or they contain a reactor grade instrument. When block valves or instruments have been removed the lines have been sealed using plugs or pipe caps. The above containment is considered acceptable.

Improvements are needed in two systems presently used for monitoring the facility. (1) Some of the gauges in the transmitter room used to measure the cell pressure do not appear to be in good condition and they are connected to the cell with PVC instrument tubing. (2) The gauge in the vent house used to measure the pressure in the drain tanks also connects to two glass rotameters which could be broken. It is suggested that unnecessary lines and instruments be removed and defective gages be replaced. The use of PVC instrument tubing should be minimized. It is also suggested that the procedures be changed to require closing an isolation valve after the periodic reading of the pressures.

3.4 Thermocouples

The thermocouples are routed through the penetration sleeves in 1/4" copper tubing which have seals inside and outside the cell. The copper tubing is soldered to the penetration sleeves. The outside seals appear to be in good condition, and the penetrations are considered acceptable.

3.5 Electrical

Electrical wires are routed through the penetration sleeves in magnesium-oxide-copper sheaths which have seals inside and outside the cell. The sheaths are sealed to the penetration sleeves by two compression-type fittings. During early operation a number of small leaks were encountered at the electrical penetrations, and these were coated with epoxy resin. The epoxy resin and the outside seals appear to be in good condition, and the penetrations are considered acceptable.

3.6 Water Lines

In August, 1985, water lines entering the cells were sealed to prevent water from accidentally getting into the cells and to enhance containment. Screwed caps or blank flanges were used. These were reexamined at this time and were found in good condition. Containment is considered acceptable.

The supply and return lines to the drain tank cell space cooler were not capped. Block and check valves provide adequate containment. However, even though the space cooler may be needed when the fuel is eventually melted for repackaging and/or removal from the drain tanks, it is suggested that these also be sealed with screwed caps or blank flanges.

3.7 Drain Tank Steam Drums

The steam domes and bayonettes inside the drain tank cell are closed systems. They are connected to feedwater tanks and condensers in the west tunnel which are vented to the vapor

condensing system. Sometime during the storage period, control valves were removed from the feedwater lines to the steam domes (lines 806 and 807), and the lines were not resealed. Since the system is closed inside the drain tank cell, containment is considered acceptable. However, if the steam domes are removed, all flanges inside the cell will need to be blanked.

3.8 Leak Detector Lines

Leak detector lines were provided to indicate leakage of in-cell "O" ring seals on flanges. Approximately sixty lines of 1/4 in. OD X 0.083 in. wall thickness tubing run from penetrations in the south electric service area to headers in the transmitter room. Shut-off valves are located at these headers. Since all valves are closed, containment is considered acceptable.

3.9 Ventilation and Component Cooling Systems

Line 930 which is a 30-in. diameter pipe runs from the reactor cell to the ventilation system, where the line contains two butterfly valves in series for containment. These valves are locked closed with chains as are two 2-in. valves which bypass the butterfly valves. This is considered to be acceptable containment.

A 10-in. line connects to line 930 between the cell and the butterfly valves. This line connects to the vapor condensing system. Containment is provided by a 10-in. rupture disc having a bursting pressure of 20 psi and a parallel 3-in. rupture disc with a bursting pressure of 15 psi. Since these are high-quality rupture discs made of stainless steel and teflon, containment is considered acceptable.

The component coolant system which is presently shutdown takes its suction from line 930 and discharges to the reactor cell for freeze valve and other cooling. The component coolant pumps are located in large containment tanks, and the system is totally closed. Therefore, containment is considered acceptable.

3.10 Fuel Sampler

The fuel sample line has two ball valves between the fuel pump and the sampler. These are closed and the sampler access port is closed. The vent line from the access port is blocked by closed hand valves. Containment is considered acceptable.

3.11 Coolant Salt Lines

The coolant salt lines from the heat exchanger to the radiator have been welded closed. This is considered acceptable containment.

3.12 Sump Lines

The air supply lines to the reactor cell and drain tank cell sump jets contain closed hand valves and the one to the drain tank cell sump has been sealed with a screwed pipe cap. Containment is considered acceptable. However, these lines are located outside on the west side of the building which is easily accessible. Therefore, it is suggested that the line to the reactor cell sump also be capped.

Each sump jet discharges to the liquid waste storage tank through two fail closed valves in series. Since the waste cell is closed, these valves could not be inspected. However, this containment is considered acceptable.

The bubbler level indicators contain check valves near the penetration and hand valves which are opened periodically to check the sump levels. Containment is considered acceptable.

3.13 Nuclear Instrument Thimble

The nuclear instrument thimble or well is a 3-foot diameter by 25-foot long welded stainless steel tube which extends from the high bay to the thermal shield. It is filled with water for shielding. The water contains a corrosion inhibiter. Containment is considered acceptable.

3.14 Spares

All spare penetrations which could be examined were in good condition and containment is considered acceptable. Those in the fuel storage cell could not be checked.

Conclusions and Recommendations

It was concluded that all of the reactor cell and drain tank cell penetrations are in good condition and containment was considered acceptable. There are essentially no corrosion problems which would cause future problems.

Three recommendations are listed below:

(1) The quality of two instrumentation systems presently used to monitor the drain tank and drain tank cell pressures should be improved (see 3.3).

(2) The lines to the drain tank cell space cooler should be capped (see 3.6).

(3) The reactor cell sump jet air supply line should be capped (see 3.12).

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4. R. C. Robertson, "MSRE Design and Operations Report, Description of Reactor Design," ORNL-TM-728, Part I, January 1965.

5. K. J. Notz, "Extended Storage-in-Place of MSRE Fuel Salt and Flush Salt," ORNL/TM-9756, September 1985. See also, K. E. Lott, "MSRE Cell Penetrations Study," J3E20794 (E100 to E104) and J3D20794 (E015), April 1986.

6. J. R. Tallackson, "MSRE Design and Operations Report, Nuclear and Process Instrumentation," ORNL-TM-729, Part II A, February 1968.

7. R. L. Moore, "MSRE Design and Operations Report, Nuclear and Process Instrumentation," ORNL-TM 729, Part II B, September 1972.

8. P. N. Haubenreich, J. R. Engle, C. H. Gabbard, R. H. Guymon, and B. E. Prince, "MSRE Design and Operations Report" (Safety Analysis of Operations with ^{233}U), Part V-A, ORNL-TM-2111, February 1968.

9. MSR Program Semiannual Progress Report, February 26, 1971, ORNL-4676.

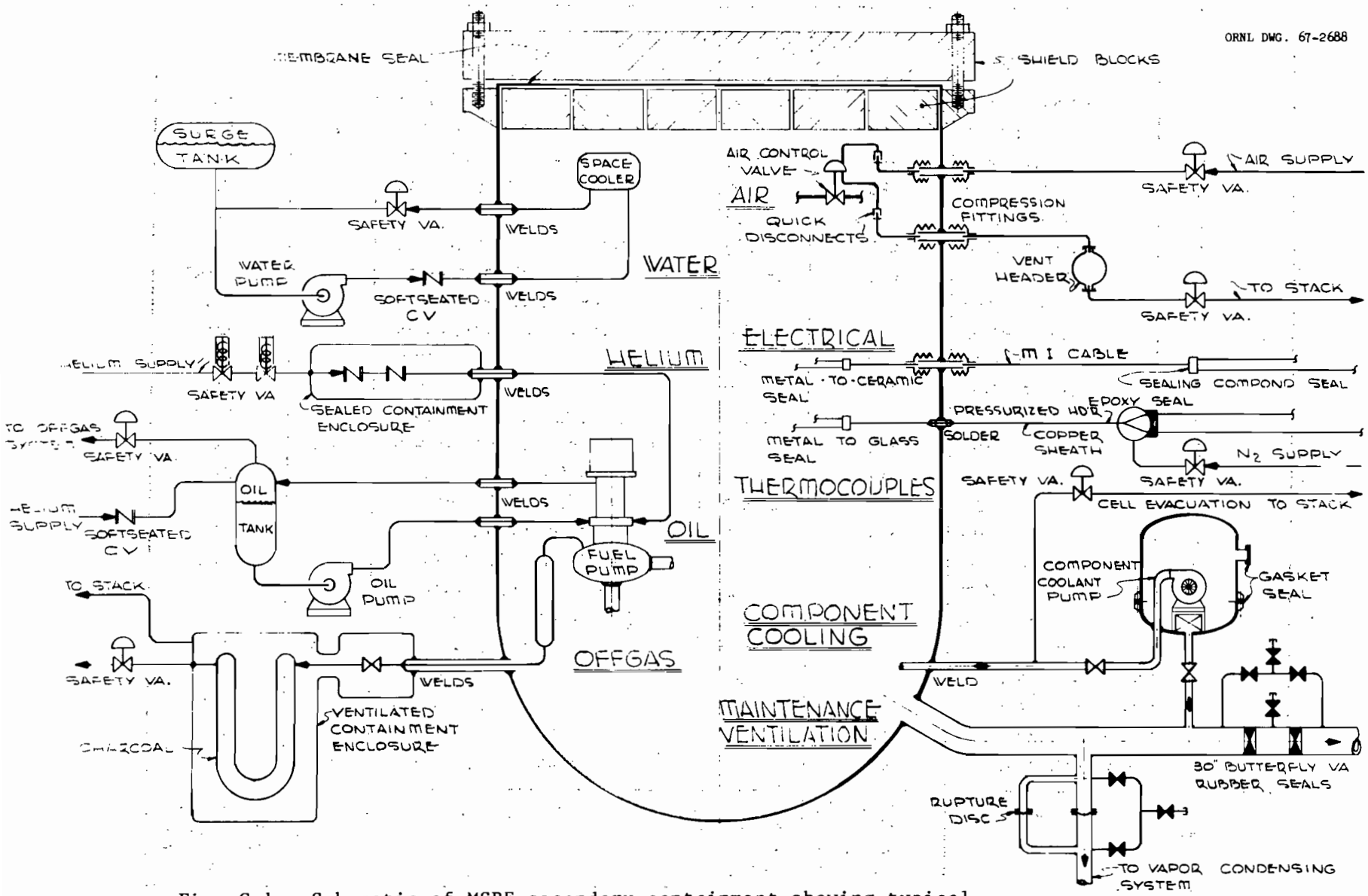

Fig. C.1. Schematic of MSRE secondary containment showing typical penetration seals and closures.

Table C.1. Reactor cell penetrations

MSRE Reactor Cell Penetration Number	Former Number	Identification	Approximate Location in the Reactor Cell		Access Area	Penetration Size, ID (in.)
			Elevation (ft)	Arc (N = 0° Ref.)		
I	R-4	Reactor Leak Detectors	836	15	S. Elec. Serv. Area	24
II	R-3	Electrical	834	30	S. Elec. Serv. Area	24
III	R-2	Electrical	836	45	S. Elec. Serv. Area	24
IV	R-1	Thermocouples	834	60	S. Elec. Serv. Area	24
V	R	Instrumentation	836	75	S. Elec. Serv. Area	24
VI		Sampler Offgas (918, 542)	847	110	High Bay	4
VII		Sampler (999)	847	115	High Bay	6
VIII	S-1	FP (590, 703, 704, 706)	836-9"	125	Service Tunnel	18
IX		Neutron Instrument Tube	834-5"	145	High Bay	36
X		FP Level (592, 593, 596)	844-6"	155	SER	4
XI	S-3	FP (516, 519, 524, 606)	836-9"	160	SER	18
XII	S-4	Component Coolant Air (917)	829-10"	165	SER	6
XIII		Coolant Salt to HX (200)	840-10"	170	Coolant Cell	24
XIV		Water Lines (830, 831)	839-9"	185	Coolant Cell	8
XV		Spare	839-9"	200	Coolant Cell	8
XVI		Water Lines (844, 845)	839-9"	205	Coolant Cell	8
XVII		Water Lines (838, 846)	839-9"	210	Coolant Cell	8
XVIII		Water Lines (840, 841)	839-9"	220	Coolant Cell	8
XIX		Coolant Salt to Radiator (201)	837	220	Coolant Cell	24
XX		Offgas (522)	839-9"	225	Coolant Cell	6
XXI		Offgas (561)	839-9"	230	Coolant Cell	6
XXII		Cell Exhaust Duct (930)	824-10"	245	CDT Cell	30
XXIII	R-7	Thermocouple	836	325	West Tunnel	24
XXIV		Drain Tank Cell Interconnection (103, 333, 521, 561, 920)	825-2"	330	Drain Tank Cell	36

Table C.1. (continued)

MSRE Reactor Cell Penetration Number	Shielding	Reference Drawings (General References: EGGD-40704, 41487, 41489, 41490)
I	Magnetite grout	DKKD-40976, EBBD-41863, EBBD-41864, DJJD-55494, DJJD-40495
II	Magnetite grout	DKKD-40976, EBBD-41863, EBBD-41864, EMMZ-56230, EMMZ-56246
III	Magnetite grout	DKKD-40976, EBBD-41863, EBBD-41864, EMMZ-56230, EMMZ-56246
IV	Magnetite grout	DKKD-40976, EBBD-41863, EBBD-41864, DHHB-55567
V	Magnetite grout	DKKD-49976, EBBD-41863, EBBD-41864, DHHB-55567
VI	Fuel Sampler	DKKD-40973, DKKD-40974
VII	Steel plates	DKKD-40973, DKKD-40974, DBBC-41339
VIII	Sand and water from annulus	DKKD-40717, EKKD-40735
IX	Water in penetration	DKKD-40716, EKKD-40715, EHHA-41796
X	Tube filled with magnetite grout	DKKD-40973, DKKD-40975, EGGD-55411, EJJD-55428
XI	Tube filled with sand and water from annulus	DKKD-40718, EKKD-40737, EGGD-55411
XII	Lead in annulus	DKKD-40714, EGGD-55411
XIII	Steel except for pipe and heaters	EKKD-40711, EGGZ-55498
XIV	Steel shot except for straight-through pipes	DKKD-40740, DKKD-40741
XV	Steel shot except for straight-through pipes	DKKD-40740, DKKD-40741
XVI	Steel shot except for straight-through pipes	DKKD-40740, DKKD-40741
XVII	Steel shot except for straight-through pipes	DKKD-40740, DKKD-40741
XVIII	Steel shot except for straight-through pipes	DKKD-40740, DKKD-40741
XIX	Steel except for pipe and heaters	DKKD-40712, EGGZ-55498
XX	Steel shot except for straight-through pipes	DKKD-40740, DKKD-40741
XXI	Steel shot except for straight-through pipes	DKKD-40740, DKKD-40741
XXII	Steel plate in reactor cell	DKKD-40710, EKKD-40749
XXIII	Magnetite grout	DKKD-40976, EBBD-41863, EBBD-41864, DHHB-55567
XXIV	None needed	EKKD-40713

Table C.2. Drain tank cell penetrations

MSRE Drain Tank Cell Penetration Number	Identification	Location in Drain Tank Cell	Access Area	Penetration Size, I. D.	References (General References: EGGD-40708 40709, 40882, 41512, 41513)
1	Steam (804)	South Wall	South Elec. Serv. Area	3" in 4"	DKKD-40948, EGGD-55425
2	Steam (805)	" "	" "	3" in 4"	" "
3	Condensate (806)	" "	" "	3" in 1"	" "
4	Condensate (807)	" "	" "	3" in 1"	" "
5	Water (837)	" "	" "	3" in 2"	DKKD-40948, DKKB-41253
6	Water (836)	" "	" "	3" in 2"	DKKD-40948
7	Spare	" "	" "	3" in 1"	" "
8	Spare	" "	" "	3" in 1"	" "
9	Spare	" "	" "	1/2" in 1"	" "
10	Spare	" "	" "	1/2" in 1"	" "
11	Spare	" "	" "	1"	" "
12	Spare	" "	" "	1"	" "
13	Spare	" "	" "	1/2" in 1"	" "
14	Spare	" "	" "	1/2" in 1"	" "
15	Spare	" "	" "	1/2" in 1"	" "
16	Spare	" "	" "	1/2" in 1"	" "
17	Sump Discharge (333)	" "	Waste Cell	3/4"	DKKD-40948, DKKB-41280, DKKB-41281
18	Sump Discharge (343)	" "	" "	3/4"	" " "
19	Air to Sump (332)	West Wall	W. of Bldg.	3/4"	DKKD-40948, DKKB-41280
20	Air to Sump (342)	" "	" "	3/4"	" "
21	Spare	North Wall	F. Proc. Cell	1-1/2"	DKKD-40949
22	Spare	" "	" "	1-1/2"	"
23	Spare	" "	" "	1-1/2"	"
24	Spare	" "	" "	1-1/2"	"
25	Spare	" "	" "	1-1/2"	"
26	Spare	" "	" "	4"	"
27	Spare	" "	" "	1-1/2"	"
28	Spare	" "	" "	1-1/2"	"
29	Spare	" "	" "	1-1/2"	"
30	Salt Transfer (110)	" "	" "	14"	"
A-1 to 36	Instrumentation	East Wall	N. Elec. Serv. Area	3/4"	DKKD-40947, DHHB-55567
B-1 to 36	Instrumentation	" "	" "	3/4"	" "
C-1 to 36	Thermocouples	" "	" "	3/4"	" "
D-1 to 36	Thermocouples	" "	" "	3/4"	DKKD-40947
E-1 to 36	Thermocouples	" "	" "	3/4"	"
A-37 to 60	Electrical	" "	" "	3/16 in 3/4	DKKD-40947, EMMZ-51656
B-37 to 60	Electrical	" "	" "	" "	" "
C-37 to 60	Electrical	" "	" "	" "	" "
D-37 to 60	Electrical	" "	" "	" "	" "
E-37 to 60	Electrical	" "	" "	" "	" "
F-37 to 60	Electrical	" "	" "	" "	" "
G	Spare	" "	" "	6"	DKKD-40947
H	Spare	" "	" "	6"	"
I	Cover Gas (572, 574, 576)	" "	" "	6"	DKKD-40947, EGGE-41884
J	Spare	" "	" "	6"	DKKD-40947
K	Spare	" "	" "	6"	"
L	Spare	" "	" "	6"	"
M	Component Coolant Air (911, 912, 913)	" "	" "	6"	DKKD-40947, EGGE-41884, DJJA-41879, DJJA-41880
N	Component Coolant Air	East Wall	N. Elec. Serv. Area	6"	DKKD-40947, EGGE-41884, DJJA-41879, DJJA-41880
O	Component Coolant Air (920)	" "	" "	6"	DKKD-40947, EGGE-41884, DJJA-41879, DJJA-41880
P	DP Cell (LT 806 and 807)	" "	" "	6"	DKKD-40947, EGGE-41884, DJJA-41879, DJJA-41880
Q	Leak Detector	" "	S. Elec. Serv. Area	6"	DKKD-40947, EBBD-41863, EBBD-41865, DJJD-55494, DJJD-55495
R	Leak Detector	" "	" "	6"	DKKD-40947, EBBD-41863, EBBD-41865, DJJD-55494, DJJD-55495

APPENDIX D: BLENDING MSRE SALTS WITH LIQUID HLW

APPENDIX D. BLENDING MSRE SALTS WITH LIQUID HLW

D.1 INTRODUCTION AND OVERVIEW

Three DOE installations reprocess defense-generated spent nuclear fuel, producing liquid high-level waste: Savannah River, Hanford, and Idaho Operations. The first two deal mainly with aluminum-clad plutonium production fuel, and produce an acidic waste which is neutralized and stored in tanks where it resides awaiting vitrification into borosilicate glass.

The Idaho facility reprocesses zircaloy-clad naval fuel, which requires the use of hydrofluoric acid (as well as nitric acid) to achieve dissolution.[1] This has been done using the Zirflex process, but recently the Fluorinel process was introduced. The resulting solution is then converted to a dry calcine via a large spray calciner, and this calcine is stored in bins awaiting final disposal. Recent studies have shown that this calcine can be incorporated into a high-density ceramic for final disposal.[2] The calcine itself (Table D.1) is very high in both fluoride (33% as CaF_2) and zirconium (18% as ZrO_2). It does not contain significant amounts of either lithium or beryllium, but it does contain some monovalent cations (3.4% as K_2O plus Na_2O) and additional divalent cations (14% as CaO, in addition to the CaF_2 cited above). The MSRE fuel and flush salts should be chemically compatible with such a mixture. This suggests the possibility of blending the MSRE salts with the Idaho high-level waste stream. The solubility of the MSRE salts appears adequate to allow this. Such an approach could serve as an alternative to direct disposal of the gettered salt in repackaged containers.

D.2 DEFENSE HLW PROCESSING

Relatively large amounts of liquid HLW have been produced at the three defense reprocessing sites. The current inventories and projected quantities are tabulated in Tables D.2, D.3, and D.4 for Savannah River, Hanford, and Idaho, respectively.[3] The following tables (D.5, D.6, and D.7) give the thermal outputs and the radioactivities.[3] Tables D.8, D.9, and D.10 give the chemical compositions for these three generators.[3]

Table D.1. Composition of calcine at ICPP*

Component	Wt%
K_2O	0.7
Na_2O	2.7
Al_2O_3	18.6
ZrO_2	18.2
CaF_2	32.9
CaO	13.7
NO_3-	7.5
Misc.	5.7

*From R. S. Baker, B. A. Staples, and H. C. Wood, "Development of Ceramic-Based Waste Form to Immobilize ICPP High-Level Waste," WINCO-1044, September 1986.

Table D.2. Current and projected volumes of HLW at SRP

End of calendar year	Volume, 10^3 m^3				
	Liquid	Sludge	Saltcake	Glass	Total
1986	72.90	13.80	41.20	0.00	127.80
1987	53.17	9.17	46.49	0.00	108.83
1988	44.74	9.52	50.83	0.00	105.09
1989	42.37	7.13	49.78	0.00	99.28
1990	43.76	7.45	47.47	0.06	98.74
1991	41.89	7.76	46.77	0.32	96.74
1992	39.75	5.81	43.35	0.58	89.49
1993	36.41	6.13	42.63	0.84	86.01
1994	40.34	6.38	38.83	1.10	86.65
1995	39.99	3.74	36.71	1.36	81.80
1996	31.70	4.03	36.49	1.62	73.84
1997	29.87	4.33	34.90	1.88	70.98
1998	34.42	3.71	31.37	2.14	71.64
1999	34.82	3.78	31.40	2.40	72.40
2000	36.12	2.54	30.03	2.66	71.35
2001	33.12	2.66	27.49	2.80	66.07
2002	33.41	1.12	26.37	2.94	63.84
2003	37.60	1.39	23.05	3.08	65.12
2004	38.79	0.61	20.85	3.22	63.47
2005	36.68	0.88	22.26	3.36	63.18
2006	39.44	1.16	24.20	3.50	68.30
2007	39.44	1.16	24.20	3.56	68.36
2008	39.44	1.16	24.20	3.62	68.42
2009	39.44	1.16	24.20	3.68	68.48
2010	39.44	1.16	24.20	3.74	68.54
2011	39.44	1.16	24.20	3.80	68.60
2012	39.44	1.16	24.20	3.86	68.66
2013	39.44	1.16	24.20	3.92	68.72
2014	39.44	1.16	24.20	3.98	68.78
2015	39.44	1.16	24.20	4.04	68.84
2016	39.44	1.16	24.20	4.10	68.90
2017	39.44	1.16	24.20	4.16	68.96
2018	39.44	1.16	24.20	4.22	69.02
2019	39.44	1.16	24.20	4.28	69.08
2020	39.44	1.16	24.20	4.34	69.14

[a]Source: Chandler 1987.

Table D.3. Hanford Operations. Historical and projected volumes of single-shell tank waste, double-shell tank waste, and borosilicate glass[a]

End of calendar year	Single-shell tanks cumulative volume, m^3			Double-shell tanks	Borosilicate glass	
	Liquid	Sludge	Salt cake	Cumulative volume, m^3	Number of canisters	Cumulative volume, m^3
1982	3.3E+04	4.7E+04	9.7E+04	3.6E+04	0	0
1983	3.0E+04	4.7E+04	9.4E+04	5.9E+04	0	0
1984	2.9E+04	4.6E+04	9.3E+04	5.7E+04	0	0
1985	2.8E+04	4.6E+04	9.3E+04	5.5E+04	0	0
1986	2.8E+04	4.6E+04	9.3E+04	6.5E+04	0	0
1987	2.7E+04	4.6E+04	9.3E+04	6.3E+04	0	0
1988	2.7E+04	4.6E+04	9.3E+04	5.4E+04	0	0
1989	2.7E+04	4.6E+04	9.3E+04	5.7E+04	0	0
1990	2.6E+04	4.6E+04	9.3E+04	5.7E+04	0	0
1991	2.4E+04	4.6E+04	9.3E+04	5.7E+04	0	0
1992	2.3E+04	4.6E+04	9.3E+04	5.7E+04	0	0
1993	1.8E+04	4.6E+04	9.3E+04	5.9E+04	0	0
1994	1.1E+04	4.6E+04	9.3E+04	6.4E+04	0	0
1995	7.4E+03	4.6E+04	9.3E+04	6.0E+04	0	0
1996	7.2E+03	4.6E+04	9.3E+04	5.3E+04	145	90
1997	7.2E+03	4.6E+04	9.3E+04	4.3E+04	290	181
1998	7.2E+03	4.6E+04	9.3E+04	3.1E+04	435	271
1999	7.2E+03	4.6E+04	9.3E+04	2.8E+04	508	316
2000	7.2E+03	4.6E+04	9.3E+04	2.5E+04	653	407
2001	7.2E+03	4.6E+04	9.3E+04	2.4E+04	798	497
2002	7.2E+03	4.6E+04	9.3E+04	1.7E+04	870	542
2003	7.2E+03	4.6E+04	9.3E+04	1.3E+04	1,015	632
2004	7.2E+03	4.6E+04	9.3E+04	9.4E+03	1,160	722
2005	7.2E+03	4.6E+04	9.3E+04	2.3E+03	1,305	812
2006	7.2E+03	4.6E+04	9.3E+04	2.8E+03	1,378	858
2007	7.2E+03	4.6E+04	9.3E+04	3.4E+03	1,523	948
2008	7.2E+03	4.6E+04	9.3E+04	3.9E+03	1,668	1,038
2009	7.2E+03	4.6E+04	9.3E+04	4.5E+03	1,740	1,083
2010	7.2E+03	4.6E+04	9.3E+04	5.1E+03	1,860	1,158
2011	7.2E+03	4.6E+04	9.3E+04	5.6E+03	1,860	1,158
2012	7.2E+03	4.6E+04	9.3E+04	6.2E+03	1,860	1,158
2013	7.2E+03	4.6E+04	9.3E+04	2.9E+03	1,860	1,158
2014	7.2E+03	4.6E+04	9.3E+04	3.5E+03	1,860	1,158
2015	7.2E+03	4.6E+04	9.3E+04	4.0E+03	1,860	1,158
2016	7.2E+03	4.6E+04	9.3E+04	5.2E+01	1,860	1,158
2017	7.2E+03	4.6E+04	9.3E+04	5.2E+01	1,860	1,158
2018	7.2E+03	4.6E+04	9.3E+04	5.2E+01	1,860	1,158
2019	7.2E+03	4.6E+04	9.3E+04	5.2E+01	1,860	1,158
2020	7.2E+03	4.6E+04	9.3E+04	5.2E+01	1,860	1,158

[a]Source: Coony 1987. Volumes shown are total cumulative volumes at end of year. It was assumed that there is no fuel reprocessing after CY 2001. Each additional year of fuel reprocessing after CY 2001 was estimated to generate an equivalent of 50 canisters per year and an equivalent of 35 m^3 of borosilicate glass per year after CY 2010. The amount of residual volume remaining in double-shell tanks after CY 2015 was assumed to be 0.05% of the tank capacity.

Table D.4. INEL. Historical and projected volumes of interim and immobilized high-level wastes[a]

Calendar year end	Liquid $10^3 m^3$	Calcine 10^3 m^3	Glass-ceramic 10^3 m^3	Glass-ceramic Cumulative number of canisters	Total 10^3 m^3
1986	6.5	3.0	0.0	0	9.5
1987	7.6	3.1	0.0	0	10.7
1988	7.5	3.4	0.0	0	10.9
1989	7.8	3.7	0.0	0	11.5
1990	6.8	4.0	0.0	0	10.8
1991	6.6	4.2	0.0	0	10.8
1992	6.2	4.5	0.0	0	10.7
1993	5.6	4.9	0.0	0	10.5
1994	6.1	4.9	0.0	0	11.0
1995	5.2	5.2	0.0	0	10.4
1996	5.8	5.3	0.0	0	11.1
1997	6.3	5.8	0.0	0	12.1
1998	7.1	6.3	0.0	0	13.4
1999	7.9	6.8	0.0	0	14.7
2000	8.6	7.4	0.0	0	16.0
2001	8.8	7.9	0.0	0	16.7
2002	9.2	8.4	0.0	0	17.6
2003	9.6	8.9	0.0	0	18.5
2004	8.3	9.4	0.0	0	17.7
2005	6.1	9.9	0.0	0	16.0
2006	7.4	9.9	0.0	0	17.3
2007	8.5	9.9	0.0	0	18.4
2008	4.7	10.5	0.0	0	15.2
2009	4.4	11.2	0.0	0	15.6
2010	4.5	11.9	0.0	0	16.4
2011	3.9	12.15	0.28	500	16.33
2012	3.1	12.30	0.63	1100	16.03
2013	2.7	12.36	1.03	1800	16.09
2014	3.0	12.05	1.60	2800	16.65
2015	3.3	11.74	2.17	3800	17.21
2016	3.7	11.53	2.74	4800	17.97
2017	3.1	11.32	3.31	5800	17.73
2018	1.3	10.70	3.88	6800	15.88
2019	1.6	10.49	4.45	7800	16.54
2020	1.7	10.28	5.02	8800	17.00

aSource: Berreth 1987. Quantities shown are based on the assumptions that immobilization starts in year 2011 and that the glass-ceramic form is used. Each canister is assumed to contain 0.57 m^3 of ceramic (1825 kg), with a calcine loading of 70 wt% of 1277 kg; this is the equivalent of 0.91 m^3 of calcine prior to immobilization. The reader is cautioned that these projections are based on estimates and assumptions that are subject to change.

Table D.5. Radioactivity and thermal power of HLW at SRP[a]

End of calendar year	Radioactivity, millions of curies					Thermal power (1000 W)
	Liquid	Sludge	Salt Cake	Glass	Total	
1987	83.9	619.4	163.7	0	867.0	2666
1988	80.0	590.1	163.8	0	833.9	2553
1989	88.0	686.8	167.0	0	941.8	2976
1990	87.6	696.9	156.7	6.1	947.3	2995
1991	93.7	747.1	154.2	27.0	1022.0	3284
1992	97.5	796.2	139.5	50.8	1084.0	3509
1993	93.2	758.0	129.6	74.2	1055.0	3374
1994	90.3	712.3	120.0	98.4	1021.0	3238
1995	94.6	738.9	106.6	127.9	1068.0	3418
1996	91.9	706.9	89.7	156.5	1045.0	3324
1997	93.8	724.0	87.5	171.7	1077.0	3443
1998	91.3	694.4	77.5	192.8	1056.0	3358
1999	93.2	707.8	68.7	214.3	1084.0	3462
2000	85.7	620.7	55.6	242.0	1004.0	3148
2001	91.6	678.4	55.0	249.0	1074.0	3425
2002	84.2	612.9	54.6	251.3	1003.0	3144
2003	87.4	639.4	54.3	256.9	1038.0	3288
2004	87.7	643.4	54.5	261.4	1047.0	3319
2005	87.5	640.5	54.5	268.5	1051.0	3336
2006	87.0	636.1	54.2	276.7	1054.0	3345
2007	87.0	637.7	54.4	277.9	1057.0	3353
2008	87.0	635.7	54.9	281.4	1059.0	3359
2009	86.6	635.7	54.9	283.8	1061.0	3366
2010	86.6	633.9	55.4	287.1	1063.0	3372
2011	86.5	635.4	55.4	287.7	1065.0	3378
2012	86.1	637.4	55.2	288.3	1067.0	3383
2013	86.0	638.9	55.2	288.9	1069.0	3388
2014	86.0	640.4	55.2	289.4	1071.0	3393
2015	82.7	643.9	54.2	291.1	1072.0	3398
2016	82.3	645.7	54.0	292.0	1074.0	3403
2017	82.2	647.2	54.0	292.6	1076.0	3408
2018	82.1	648.8	54.0	293.1	1078.0	3413
2019	81.7	649.9	53.7	293.7	1079.0	3418
2020	81.6	651.5	53.6	294.3	1081.0	3422

[a]Source: Chandler 1987. Values shown are for fission products only. Radioactivity will be about 1% greater and thermal power about 6% greater when actinides are included.

Table D.6. Hanford Operations. Cumulative historical and projected radioactivity of single-shell tank wastes, double-shell tanks wastes, and borosilicate glass[a]

End of calendar year	Single-shell tanks			Double-shell tanks (Ci)	Borosilicate glass (Ci)
	Liquid (Ci)	Sludge (Ci)	Salt cake (Ci)		
1982	3.4E+07	1.4E+08	1.5E+07	2.3E+05	0
1983	2.9E+07	1.4E+08	1.4E+07	3.8E+06	0
1984	2.8E+07	1.3E+08	1.4E+07	1.4E+08	0
1985	2.6E+07	1.3E+08	1.4E+07	1.7E+08	0
1986	2.6E+07	1.3E+08	1.3E+07	1.8E+08	0
1987	2.4E+07	1.2E+08	1.3E+07	1.6E+08	0
1988	2.3E+07	1.2E+08	1.3E+07	1.8E+08	0
1989	2.3E+07	1.2E+08	1.2E+07	2.0E+08	0
1990	2.1E+07	1.2E+08	1.2E+07	2.1E+08	0
1991	1.9E+07	1.1E+08	1.2E+07	2.2E+08	0
1992	1.8E+07	1.1E+08	1.2E+07	2.4E+08	0
1993	1.4E+07	1.1E+08	1.1E+07	2.6E+08	0
1994	8.6E+06	1.1E+08	1.1E+07	2.8E+08	0
1995	5.5E+06	1.0E+08	1.1E+07	2.8E+08	0
1996	5.2E+06	1.0E+08	1.0E+07	2.4E+08	4.7E+07
1997	5.1E+06	9.8E+07	1.0E+07	1.7E+08	7.6E+07
1998	5.0E+06	9.6E+07	1.0E+07	1.3E+08	1.0E+08
1999	4.8E+06	9.3E+07	9.8E+06	1.0E+08	1.1E+08
2000	4.7E+06	9.1E+07	9.5E+06	6.4E+07	1.3E+08
2001	4.6E+06	8.9E+07	9.3E+06	3.2E+07	1.6E+08
2002	4.5E+06	8.7E+07	9.1E+06	1.2E+07	1.7E+08
2003	4.4E+06	8.5E+07	8.9E+06	2.5E+05	1.7E+08
2004	4.3E+06	8.3E+07	8.7E+06	2.1E+05	1.7E+08
2005	4.2E+06	8.1E+07	8.5E+06	1.7E+05	1.6E+08
2006	4.1E+06	7.9E+07	8.3E+06	1.5E+05	1.6E+08
2007	4.0E+06	7.7E+07	8.1E+06	1.1E+05	1.5E+08
2008	3.9E+06	7.5E+07	7.9E+06	9.8E+04	1.5E+08
2009	3.8E+06	7.3E+07	7.7E+06	9.0E+04	1.4E+08
2010	3.8E+06	7.2E+07	7.5E+06	7.3E+04	1.4E+08
2011	3.7E+06	7.0E+07	7.4E+06	7.1E+04	1.4E+08
2012	3.6E+06	6.8E+07	7.2E+06	7.0E+04	1.3E+08
2013	3.5E+06	6.7E+07	7.0E+06	6.8E+04	1.3E+08
2014	3.4E+06	6.5E+07	6.9E+06	6.6E+04	1.3E+08
2015	3.3E+06	6.4E+07	6.7E+06	6.5E+04	1.3E+08
2016	3.3E+06	6.2E+07	6.6E+06	6.3E+04	1.2E+08
2017	3.2E+06	6.1E+07	6.4E+06	6.2E+04	1.2E+08
2018	3.1E+06	5.9E+07	6.3E+06	6.0E+04	1.2E+08
2019	3.1E+06	5.8E+07	6.1E+06	5.9E+04	1.1E+08
2020	3.0E+06	5.6E+07	6.0E+06	5.8E+04	1.1E+08

[a]Source: Coony 1987. No fuel reprocessing operations are assumed after CY 2001; however, some planning scenarios have been identified at Hanford that will extend reprocessing beyond CY 2001. Each additional year of fuel reprocessing generates an equivalent of 50 canisters per year and an equivalent borosilicate glass activity of 7.3E+6 curies per year after CY 2010. The amount of residual activity remaining in double-shell tanks is assumed to be 0.05% of the original activity.

Table D.7. INEL. Historical and projected radioactivity and thermal power of interim and immobilized high-level wastes[a]

Calendar year end	Liquid		Calcine		Glass-ceramic		Total	
	10^6 Ci	kW	10^6 Ci	kW	10^6 Ci	kW	10^6 Ci	kW
1986	12.9	38.5	47.7	137.4	0.0	0.0	60.6	175.9
1987	10.6	31.0	52.3	151.1	0.0	0.0	62.9	182.1
1988	17.7	52.4	54.2	156.7	0.0	0.0	71.9	209.1
1989	26.0	79.4	57.4	165.9	0.0	0.0	83.4	245.3
1990	15.5	46.3	62.6	181.2	0.0	0.0	78.1	227.5
1991	9.2	27.0	75.0	217.6	0.0	0.0	84.2	244.6
1992	16.5	50.1	77.3	224.4	0.0	0.0	93.8	274.5
1993	10.5	30.9	89.3	268.0	0.0	0.0	99.8	298.9
1994	11.1	32.5	85.0	246.9	0.0	0.0	96.1	279.4
1995	3.6	11.6	90.0	262.0	0.0	0.0	93.6	273.6
1996	13.3	42.5	98.1	285.5	0.0	0.0	111.4	328.0
1997	18.0	54.0	107.8	313.3	0.0	0.0	125.8	367.3
1998	27.2	81.5	121.4	353.4	0.0	0.0	148.6	434.9
1999	28.4	83.9	133.0	387.0	0.0	0.0	161.4	470.9
2000	47.3	144.7	139.0	403.9	0.0	0.0	186.3	548.6
2001	55.1	164.4	146.9	426.5	0.0	0.0	202.0	590.9
2002	63.2	189.5	158.7	460.9	0.0	0.0	221.9	650.4
2003	73.1	225.7	172.5	501.4	0.0	0.0	245.6	727.1
2004	64.9	200.0	183.1	532.1	0.0	0.0	248.0	732.1
2005	52.5	160.6	189.9	552.1	0.0	0.0	242.4	712.7
2006	57.7	176.3	184.0	535.0	0.0	0.0	241.7	711.3
2007	54.1	163.7	185.0	538.2	0.0	0.0	239.1	701.9
2008	39.8	122.2	199.2	579.9	0.0	0.0	239.0	702.1
2009	55.2	177.4	217.3	634.6	0.0	0.0	272.5	812.0
2010	58.9	189.6	242.9	715.5	0.0	0.0	301.8	905.1
2011	53.3	172.2	248.0	736.6	20.0	56.0	321.3	964.8
2012	46.7	151.9	244.2	728.6	44.0	124.0	334.9	1004.5
2013	41.5	133.6	242.1	724.2	70.0	202.0	353.6	1059.8
2014	45.0	145.4	226.2	676.9	107.0	313.0	378.2	1135.3
2015	47.0	151.6	208.2	621.2	143.0	421.0	398.2	1193.8
2016	46.6	149.5	190.0	560.7	177.0	526.0	413.6	1236.2
2017	26.9	82.9	174.4	514.2	210.0	624.0	411.3	1221.1
2018	18.1	53.6	134.8	383.1	242.0	726.0	394.9	1162.7
2019	20.4	60.7	121.5	340.5	272.0	819.0	413.9	1220.2
2020	22.7	68.1	108.9	300.4	301.0	908.0	432.6	1276.5

[a]Source: Berreth 1987 and ORNL calculations based on the assumption that two-thirds of the glass-ceramic produced each year is made from fresh calcine (average age 5 to 8 years) and the other third is made from old calcine. Each canister is assumed to contain 0.57 m^3 of ceramic (1825 kg), with a calcine loading of 70 wt% or 1277 kg; this is the equivalent of 0.91 m^3 of calcine prior to immobilization. The reader is cautioned that the assumptions and estimates used here are subject to change.

Table D.8. Savannah River Plant.
Estimated chemical compositions of interim and immobilized HLW.[a]

Liquid		Sludge		Salt cake		Glass	
Component	Wt %	Component	Wt %	Component	Wt %	Component	Wt %
Ag	Trace	$Fe(OH)_3$	11.8	$NaNO_3$	65.4	SiO_2	45.6
Hg	Trace	MnO_2	2.0	$NaNO_2$	0.9	Na_2O	11.0
Pb	Trace	$UO_2(OH)_2$	1.3	NaOH	3.4	B_2O_3	10.3
U	Trace	$Al(OH)_2$	13.7	$NaAl(OH)_4$	7.8	Fe_2O_3	7.0
F	0.003	AlO(OH)	5.2	Na_2CO_3	2.7	Al_2O_3	4.0
Fe	Trace	$CaCO_3$	1.5	Na_2SO_4	9.4	K_2O	3.6
Cl^-	0.023	$CaSO_4$	0.2	Na_3PO_4	Trace	Li_2O	3.2
OH^-	1.63	CaC_2O_4	0.2	NaF	0.2	FeO	3.1
NO_2^-	1.10	$Ni(OH)_2$	0.8	$Na_2C_2O_4$	0.1	U_3O_8	2.2
NO_3^-	9.63	HgO	0.4	Insoluble	3.7	MnO	2.0
$AL(OH)_4^-$	4.54	SiO_2	0.2	H_2O	6.4	Other	8.0
$CO_3(_2^-)$	0.72	ThO_2	1.8				
$CrO_4(_2^-)$	0.014	$Ce(OH)_3$	0.2			Total	100.0
$SO_4(_2^-)$	0.22	$ZrO(OH)_2$	0.2				
$PO_4(_3^-)$	0.12	$Cr(OH)_3$	0.2				
NH_4^+	Trace	$Mg(OH)_2$	0.2				
Na	11.0	$NaNO_3$	1.1				
H_2O	71.0	NaOH	1.3				
		Zeolite	1.5				
		Others	1.2				
		H_2O	55.0				

[a]Source: Chandler 1987.

Table D.9. Hanford Operations. Representative chemical compositions of tank wastes[a]

Component	Single-shell tanks			Double-shell tanks
	Liquid	Sludge	Salt cake	
$NaNO_3$	20.8	25.3	81.5	14.8
$NaNO_2$	15.8	3.8	1.7	5.6
Na_2CO_3	0.6	2.2	0.5	1.9
NaOH	6.2	5.3	1.5	7.0
$NaAlO_2$	12.5	1.2	1.4	6.0
NaF	--	--	--	0.4
Na_2SO_4	--	1.0	1.3	0.3
Na_3PO_4	2.3	15.8	1.6	0.8
KF	--	--	--	0.4
FeO(OH)	--	1.3	--	0.2
Organic carbon	0.17	--	--	1.2
NH_4^+	--	--	--	0.08
$Al(OH)_3$	--	2.9	--	4.9
$SrO \cdot H_2O$	--	0.1	--	--
Na_2CrO_4	1.3	--	--	--
$Cr(OH)_3$	--	0.2	--	0.02
$Cd(OH)_2$	--	0.1	--	--
$Ni(OH)_2$	--	--	--	<0.1
$BiPO_4$	--	0.5	--	--
Cl^-	--	0.1	--	--
$Ni_2Fe(CN)_6$	--	0.6	--	--
$P_2O_5 \cdot 24WO_2 \cdot 44H_2O$	--	<0.1	--	--
$ZrO_2 \cdot 2H_2O$	--	0.5	--	0.2
Fission products	--	--	--	<0.01
H_2O	38.8	33.6	10.5	56.2
Other	<0.1	5.5	--	<0.01
Hg^+	--	0.12 ppm	--	--
	100	100	100	100
Density, g/mL	1.6	1.7	1.4	~1.3

[a]Source: Coony 1987. Unless stated otherwise, all values are weight percent.

Table D.10. INEL. Representative chemical compositions of interim high-level wastes[a]

A. Chemical composition of HLW liquid, wt %

Component	Zirconium fluoride	Sodium bearing	Nonfluoride	Fluorinel
Al	1.3	0.8-1.6	1.51	0.742
B	0.15	0.005-0.01	0.003	0.241
Ca	-	0.03-0.2	0.27	-
Cl^-	-	0.06-0.1	0.023	-
Cd	-	-	-	1.42
Cr	-	-	0.036	0.0087
F^-	3.4	0.005-0.06	0.032	5.99
Fe	0.04	0.05-0.09	0.19	0.023
H^+	1.12	0.03-0.15	0.12	0.18
K	-	0.5-0.7	0.33	-
Mg	-	-	0.062	-
Mn	-	-	0.048	0.0004
Na	0.12	2.1-4.0	1.31	-
Ni	-	-	0.016	0.0049
NO_3^-	13.7	19.4-23.3	23.1	11.47
SO_4^{2-}	-	0.33-0.5	0.65	1.52
Zr	2.47	-	-	3.80
H_2O	78.7	76.6-69.2	72.3	74.6
Total	100.0	100.0	100.0	100.0
Density, g/mL	1.2	1.2-1.3	2	1.2

Table D.10. (continued)

B. Chemical composition of HLW calcine, wt %

Component	Alumina	Zirconium fluoride	Zirconium-sodium blend	Stainless steel sulfate	Fluorinel-sodium blend
Al_2O_3	82-95	13-17	10-16	4.4	6.5-7.5
$Al_2(SO_4)_3$	-	-	-	81	-
B_2O_3	0.5-2	3-4	2-3	-	3.0-3.2
CaO	-	2-4	13-17	-	3.3-3.6
CaF_2	-	50-56	33-39	-	46-49
Cd	-	-	-	-	6.0-6.5
Cr_2O_3	-	-	-	2.0	0.05
Fe_2O_3	-	-	-	7.0	0.2-0.3
Na_2O	1.3	-	6-8	-	1.5-4
NiO	-	-	-	0.9	0.02-0.03
NO_3^-	5-9	0.5-2	7-9.5	-	10-15
SO_4^{2-}	-	-	-	-	-
ZrO_2	-	21-27	16-19	-	19-20
Miscellaneous	0.5-1.5	0.5-1.5	0.5-1.5	4.4	-
Fission products and actinides	0.2-1	0.2-1	0.2-1	0.2-1	<0.1%
Density, g/mL	1.1	1.4	1.8	1.2	1.4

[a]Source: ORNL 1986.

The physical categories differ for the three sites, reflecting differences in operating flowsheets. However, it is obvious that the quantities involved are very large relative to the MSRE fuel and flush salts, whether measured in terms of volume, thermal output, or radioactivity. It therefore seems reasonable to consider blending in the MSRE salts with one of these defense HLW streams. Consideration of the chemical compositions shows that only the Idaho flowsheet produces a waste stream high in fluoride, the major component of the MSRE salts.

D.3 IDAHO CHEMICAL PROCESSING PLANT

The Idaho Chemical Processing Plant (ICPP) is operated by Westinghouse Idaho Nuclear Company, Inc. for the DOE Idaho Operations Office; the facility is adjacent to the Idaho National Engineering Laboratory.[1] The ICPP operates a production-scale reprocessing facility for naval fuel. They also operate laboratories and pilot plants which are capable of reprocessing a variety of other, speciality fuels. The liquid HLW from all of this reprocessing goes to their tank farm (as stream CPP-02, "process waste"), for subsequent conversion to calcined solids. The latter are transferred to an engineered underground storage facility. The liquid waste stream contains mixed fission products and actinides (referred to as "MFP" in their literature). The generation rate is about 7 million liters per year. The calcination is carried out in their New Waste Calciner Facility (NWCF), which was placed in operation about seven years ago. The volume of waste generated annually is projected to increase four-fold during the 1985-2010 time frame.[4] The ICPP (along with the Savannah Plant) stores miscellaneous spent fuels from test and research reactors, and reprocesses those for which process flow sheets suitable for their pilot-scale lines can be designed.[5,6] It would appear, based on chemical compositions, that the MSRE salts should be chemically compatible with the ICPP liquid HLW stream. However, this requires careful, detailed review and perhaps laboratory testing before a decision on chemical compatibility can be reached. Even if this should be favorable, institutional and site-specific issues need to be considered. These latter points were flagged in communications with Idaho;[7] two concerns cited by them were criticality (because of the 30-odd kg of ^{233}U) and hazard (because of the well-known toxic properties of airborne beryllium).

The ICPP has at their disposal an exceptionally wide range of fuel dissolution capabilities:

- Aluminum-alloyed fuel (facility CPP-601): dissolution using nitric acid and a mercuric nitrate catalyst.
- Zirconium-alloyed fuel (facility CPP-601): dissolution in hydrofluoric acid, using boron as a nuclear poison.
- Custom fuel (facility CPP-627): custom processing in their hot chemistry lab which houses the multicurie cell; each fuel processed uses its own specially-developed flowsheet and equipment.
- Electrolytic (facility CPP-640): accelerated anodic oxidation of stainless steel clad fuel (mainly EBR-II) in nitric acid, using gadolinium as a poison.
- Graphite fuel (facility CPP-640): for reclaiming uranium from ROVER fuel; dissolution in $HF-HNO_3$; no plans for future operation.
- Fluorinel and Storage Facility, FAST (facility CPP-666): provides underwater storage for zircaloy-clad fuels; dissolution in $HF-HNO_3$, using cadmium as the nuclear poison. Liquid wastes high in fluoride go to the HLW tanks.

After dissolution, the usual next step is recovery of uranium via liquid-liquid extraction. Both TBP and hexone facilities are used at ICPP. For the MSRE salts, where the ^{233}U has no salvage value, this step would, of course, be bypassed and the total solution sent to the HLW tank farm and thence to the calciner. In a general sense, neither the ^{233}U nor the beryllium should create problems - the ^{233}U because it is dilute and would be diluted even further, and the beryllium because it is a hazard only when airborne, but hot-cell facilities provide exceptional control over airborne contaminants. A process flow-sheet for a modified zirflex process is shown in Fig. D.1, indicating the composition of the liquid HLW.[8] This is the stage at which MSRE salts might be considered for blending in. Other flowsheets and HLW compositions have also been described.[9]

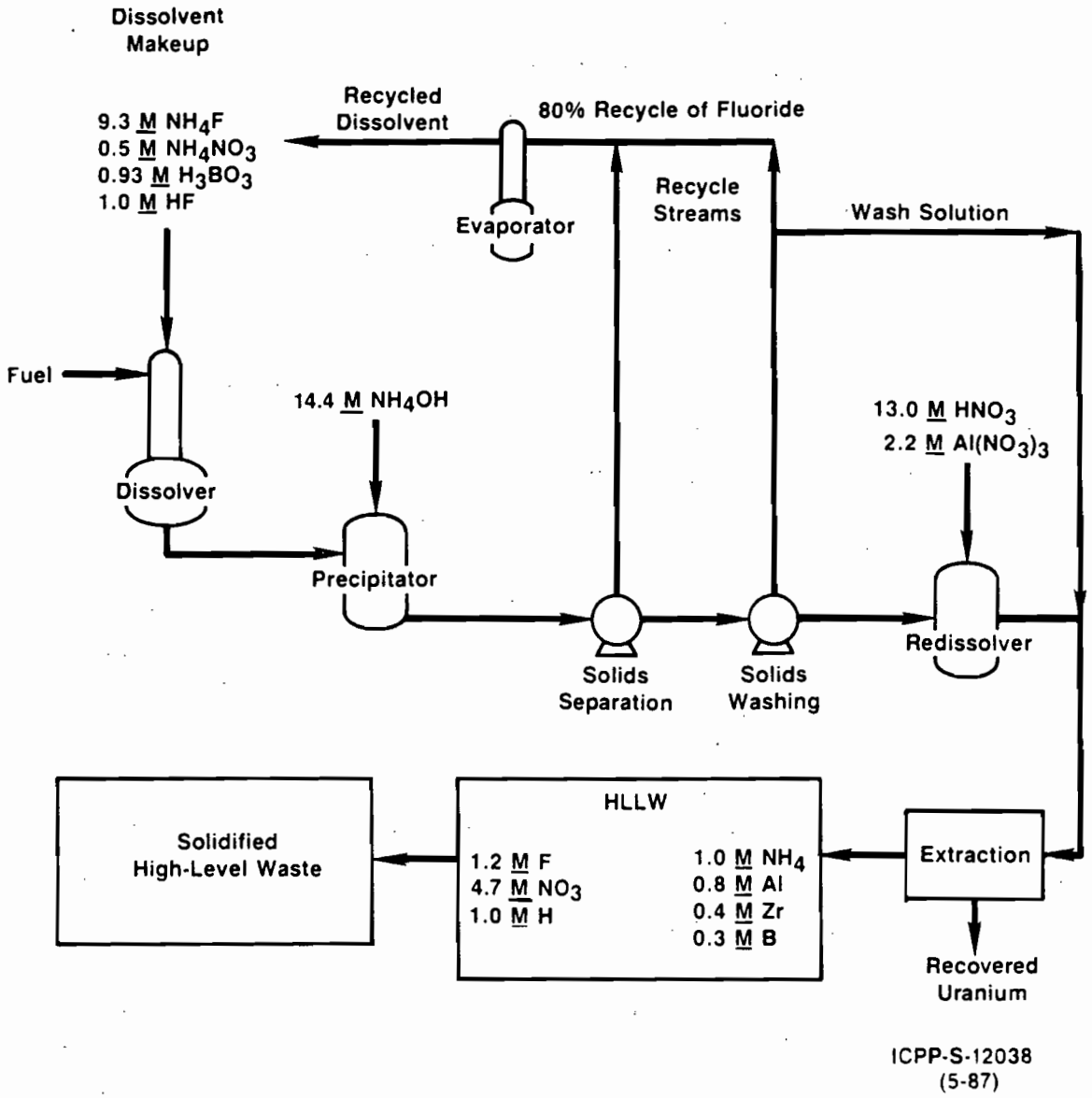

Fig. D.1. Simplified conceptual flowsheet for the modified Zirflex fuel dissolution process.

Blending MSRE salts in with ICPP wastes appears promising, but any further evaluation should involve ICPP personnel. Aside from specific concerns about criticality and chemical hazard, there must be assurance that the MSRE chemical components do not disrupt operation of the ICPP processes.

D.4 REFERENCES

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