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DISTRIBUTION OF FISSION PRODUCT CONTAMINATION IN THE SRE

By

R. S. HART

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ABSTRACT

In the safety analysis of sodium-cooled reactors, a remaining area of significant uncertainty has been the fate of various fission products that may be released to the coolant in the event of a fuel element failure; that is, the degree of their retainment in the sodium, movement to the reactor cover gas system or deposition in other portions of the primary system. During the recovery procedures following the fuel element damage that occurred at the Sodium Reactor Experiment (SRE) in July 1959,[1,2] the opportunity was provided for obtaining such data on a full-scale basis. As a consequence, the collection and analysis of pertinent information from this incident was undertaken as part of the objectives of a concurrent project which involved experimental investigation into the solubility of certain fission products in sodium.[3]

Although the necessity for compatibility with the reactor recovery efforts imposed severe restrictions on certain aspects of the project, a number of conclusions can be drawn from the data. The principal ones are:

1. Although significant fuel melting did not occur, some degree of preferential fission products volatility is involved in the relative release of the various fission products into the primary coolant.
2. Only Xe and Kr isotopes were found in the reactor cover gas (helium).
3. The carbonaceous particulate material, resulting from the decomposition of tetralin which leaked into the primary sodium, proved to be an effective fission product scavenger. A generally non-selective concentration of activity by this material of 10^3 to 10^4 times that of the filtered sodium was measured.
4. The cold trap located in the primary system was effective in removing fission product contamination.

5. Appreciable deposition of fission product contamination occurred throughout the primary piping system. A marked selectivity was evident in this process, with Sr, Ce and Zr-Nb deposition being much greater than that of Cs or I.

6. Although 5,000 to 10,000 curies of fission product activity were unexpectedly released to the primary sodium system, no radiological emergency of any nature occurred. Moreover, recovery operations were completed at a reasonable speed well within accepted personnel exposure limits. The same sodium is still in use during current operation of the SRE, with only Cs^{137} and possibly Sb^{125} remaining as significant, dissolved, long-lived contamination. Even these are now reduced to the same order as the Na^{22} activity ($\sim 10^{-2}\mu c$/gram), and their presence should not appreciably affect future operations of the reactor.

INTRODUCTION

One of the unique qualities of sodium-cooled reactors with respect to radiological safety is the potential fission-product retention ability of the coolant. The principal factors providing this ability are: (1) the occurrence of chemical reactions between the coolant and certain fission products leading to less volatile compounds (e.g. NaI), and (2) the mechanical trapping nature of the liquid coolant for particulate fission products in general. The maintenance of a coolant pool above the core in sodium reactors even with the postulation of severe reactor accidents ensures the continued effectiveness of this capacity.

An opportunity was recently afforded to obtain at least some qualitative data on this aspect of sodium-cooled reactors subsequent to the fuel element damage that occurred in the Sodium Reactor Experiment (SRE) in July 1959. This report is a summary of the fission product release and distribution data which were collected during the interval of modification and cleaning of the reactor prior to the attainment of criticality with the second core loading in September 1960.

I. THE SODIUM REACTOR EXPERIMENT

A. REACTOR SYSTEM DESIGN

The brief description of the SRE presented here is intended only to provide the necessary background for this report. Additional details may be found in Reference 4.

The SRE is a sodium-cooled, graphite-moderated, thermal power reactor. It was built primarily as a development tool with emphasis on investigation of fuel materials. A cutaway view of the reactor is shown in Figure 1.

1. Coolant Systems

Reactor coolant flow is single pass, with sodium flowing up through the core and collecting above it in a top pool. Over this pool, helium, as a blanket gas, is maintained at ~3 psig. Induced Na^{24} activity in the primary loop introduces the desirability for an intermediate heat exchanger in which the reactor heat is transferred to the secondary loop containing nonradioactive sodium.

Figure 2 presents a schematic representation of the cooling system. At full power (20 Mw) the sodium at a temperature of 500°F passes from the lower plenum up through the fuel channels, absorbing heat from the fuel elements, and discharges into the upper pool about 6 ft deep at a mixed mean temperature of 960°F.

Cold traps maintain sodium oxide content at specified low levels in the sodium cooling systems. The cold trap consists of a tank containing stainless steel wire mesh which serves as a filtering agent for the oxide precipitated from the cooled sodium flowing through the trap. These traps are capable of maintaining the oxide content of the sodium at less than 10 ppm.

2. Inert Gas System

Helium is supplied to parts of the system containing a free surface of sodium. In the primary coolant system, the core helium blanket and helium maintained in the primary sodium fill tank are interconnected and vented through the gaseous waste system. The gases discharged from this system are diverted to one of four shielded storage tanks. If necessary, they are held up here for radioactive decay prior to release at permissible concentrations through the building exhaust system.

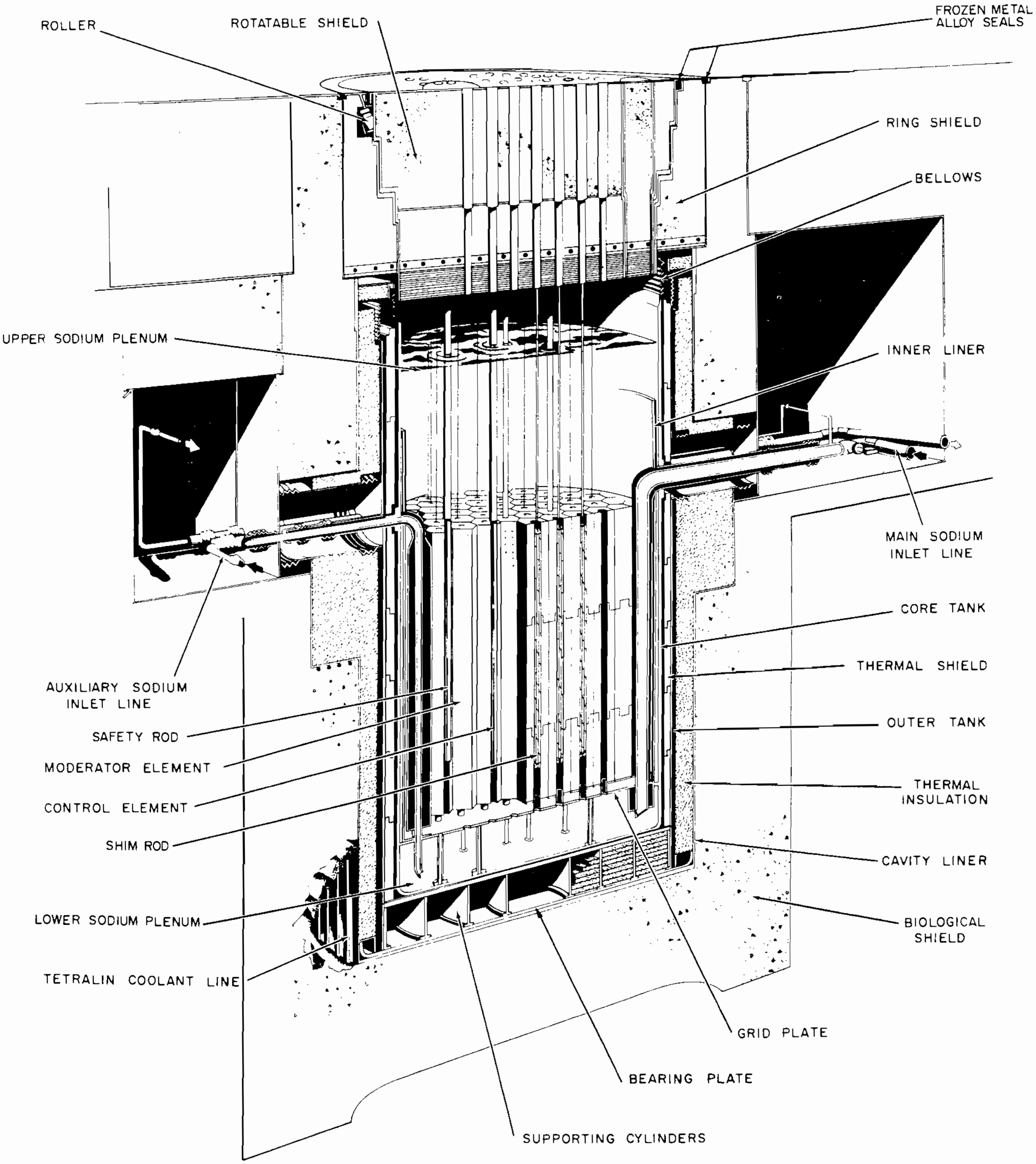

Figure 1. Cutaway View of SRE Reactor

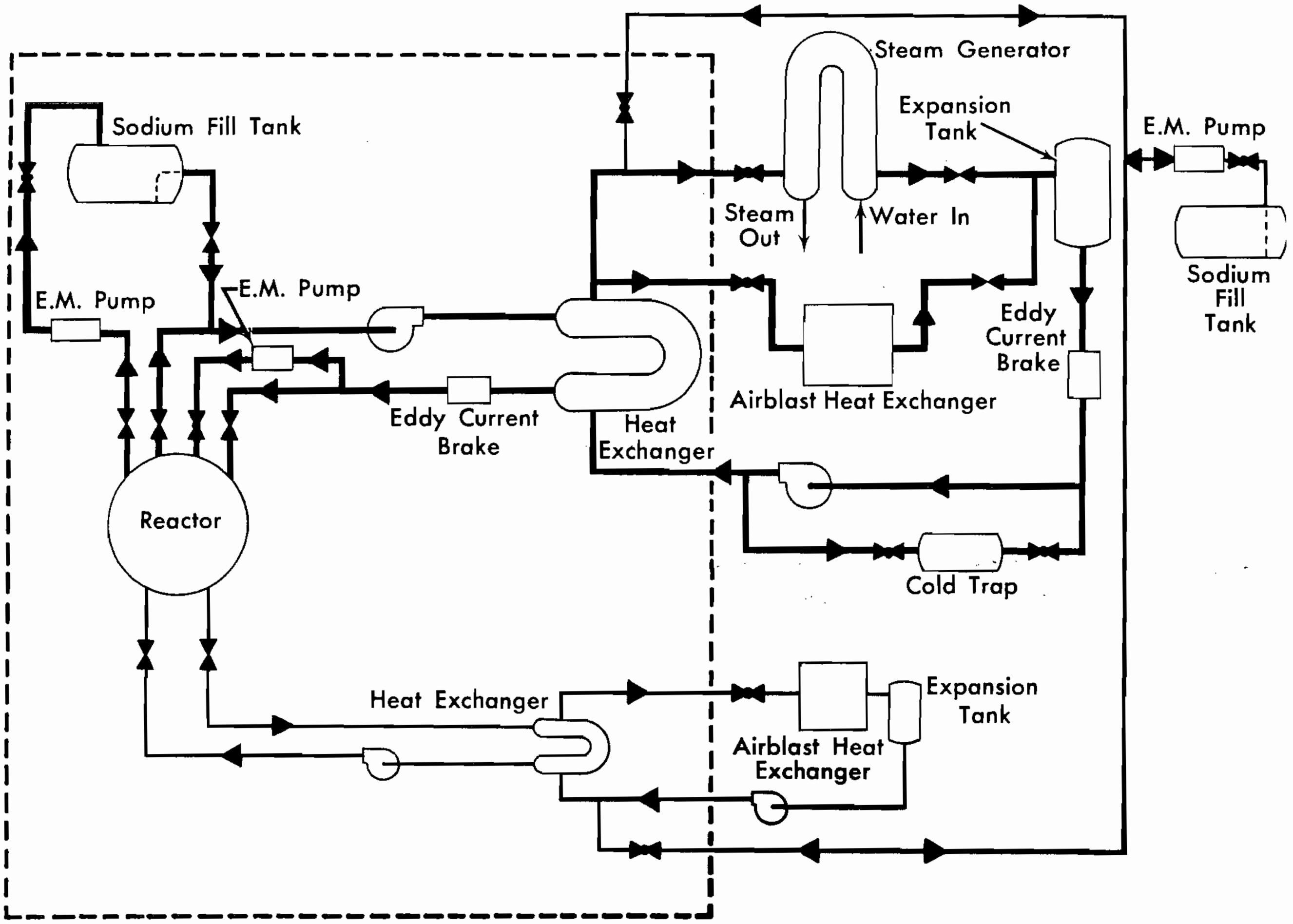

Figure 2. SRE Cooling System

3. <u>Fuel Elements</u>

The original fuel elements (Figure 3) were fabricated in clusters of seven rods, each consisting of a 6-ft-high column of uranium slugs in a thin-walled (0.010-in.) stainless steel jacket tube. The 12 slugs are 0.75 in. in diameter and 6 in. long. They are thermally bonded to the jacket by a 0.010-in. NaK annulus. Above the column of slugs is a void space containing helium. This gas-filled space allows expansion of the bonding NaK and serves as a reservoir for any fission gases escaping from the fuel slugs. The six outside rods of the cluster are spirally wrapped with stainless steel wire to provide spacing between adjacent rods and between rods and the process channel within the moderator can.

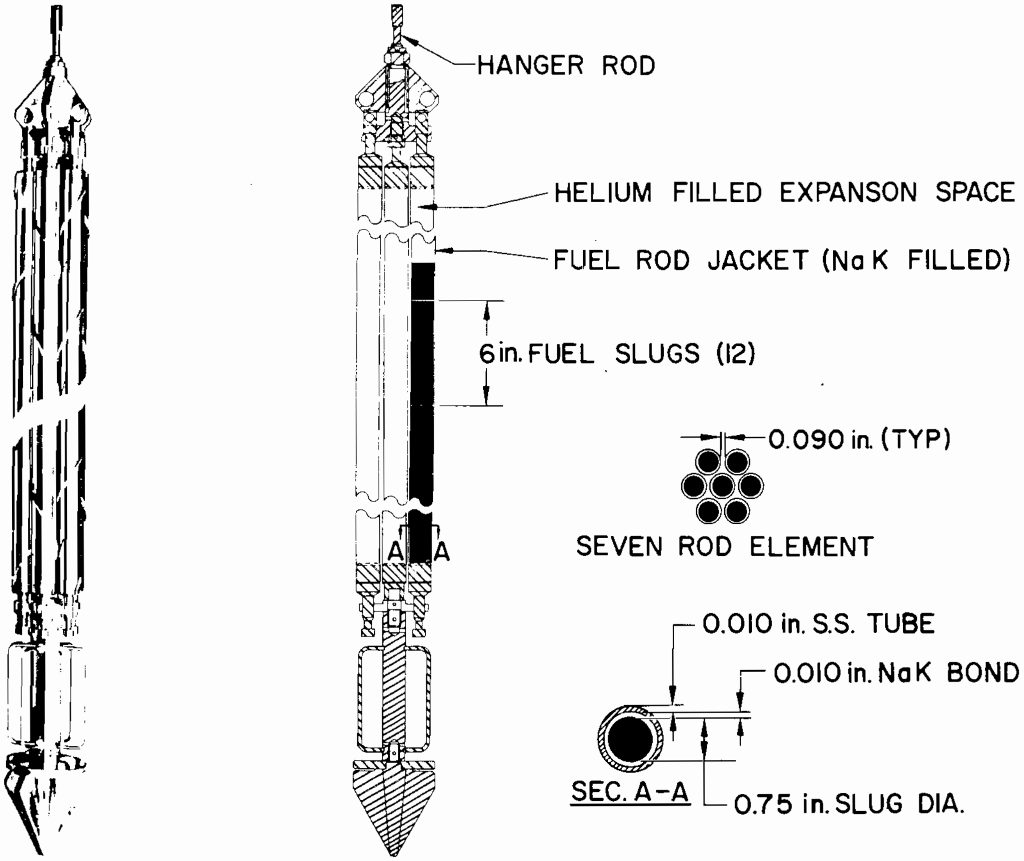

Figure 3. Typical SRE Fuel Element

4. Sodium and Helium Sampling Facilities

The capability for taking primary sodium samples during reactor operation was provided in the Material Evaluations Facility. In practice, however, operating procedures required that the reactor be shut down and primary sodium flow be stopped before taking sodium specimens. In some instances, sodium samples were taken directly out of the primary pool through top shield plugs. In this case, flow could continue, but the reactor had to be shut down.

Connections are provided for taking samples from the gas decay tanks at any time. Direct samples from the primary pool blanket gas may be taken by drawing the helium into an evacuated container from a suitable connection, usually the helium fill line.*

*At the present time, means for obtaining spot samples of the reactor cover gas are provided on the line which supplies the recently installed continuous cover gas monitor.[5]

B. OPERATIONAL HISTORY PRIOR TO JULY 26, 1959

A summary of the operating history of the reactor appears in Table I. A detailed account of this history is given in Reference 1, and only items specifically relating to the subject of this report are repeated here.

1. Primary Sodium Samples

At 20-Mwt power, the saturated Na^{24} activity of the primary sodium is ~0.2 c/g. The resultant radiation levels usually prevent routine access to parts of the primary coolant system for one to two weeks after reactor shutdown. Similarily, radioactive analyses of primary sodium samples were usually delayed by at least this length of time. However, measurements were made periodically, some typical results being shown in Table II. The isotopes listed include those inherent with the sodium activation (Na^{24}, Na^{22}), mass transfer activation (e.g., Cr^{51}), and fission products (e.g., I^{131}). The fission products identified may have originated either from "tramp" uranium in the system (as a sodium impurity) or small fuel cladding imperfections. The analyses were made using a 256-channel gamma spectrometer, either on an untreated sample, or, on occasions, following chemical separation of the major components indicated by the gross scan.

2. Reactor Cover Gas Samples

The helium cover gas was periodically sampled and assayed for radioactive content. Most of the samples were taken during shutdown intervals of reactor operation. Principally because of this delay in obtaining gas samples, the dominant radioactive contamination was Xe^{133} ($T_{1/2}$ = 5.3 days). Attempts to detect iodine invariably gave negative results. The usual irregularity of reactor power and operating time makes any quantitative relationship of cover gas radioactivity difficult, but it is apparent from Table III that the normal Xe^{133} concentration at 20 Mwt was of the order of 10^{-3} to 10^{-4} μc/cc. During reactor operation, the presence of shorter lived rare gas isotopes (e.g., Xe^{135}) may have increased the gross level to perhaps 10^{-2} μc/cc or so as evidenced by the sample taken February 26, 1959.

TABLE I

SUMMARY OF SRE POWER OPERATION
(Prior to July 26, 1959)

Power Run	Time Interval	Actual Operating Days	Average Thermal Power (Mw)	Total Irradiation (Mwd)	Total Irradiation Accumulated Since Startup (Mwd)
1	7/9/57 - 7/15/57				
2	7/25/57 - 7/26/57	5.7	3.93	22.6	22.6
3	11/7/57 - 11/29/57	12.6	6.20	78.2	100.8
4	5/21/58 - 5/28/58	13.3	8.73	116.2	217.0
5	7/18/58 - 8/4/58	11.5	17.70	203.8	420.8
6	8/8/58 - 9/1/58	22.0	17.90	394.0	814.8
7	9/8/58 - 9/25/58	17.2	17.80	306.0	1120.8
8	11/29/58 - 1/29/59	37.0	16.15	597.8	1718.6
9	2/14/59 - 2/26/59	11.5	11.0	126.5	1845.1
10	3/6 /59 - 3/7/59	0.6	5.3	3.1	1848.2
11	3/13/59 - 4/6/59	23.6	12.4	293.5	2141.1
12	5/14/59 - 5/24/59	9.7	15.9	154.3	2295.4
13	5/27/59 - 6/3/59	6.6	17.3	114.3	2409.7
14	7/12/59 - 7/26/59	14.2	1.1	16.1	2425.8

TABLE II

TYPICAL RADIOACTIVITY LEVELS OF SRE PRIMARY SODIUM PRIOR TO RUN 14

Sample No.	Sampling Location	Date of Sample Removal	Date of Last Reactor Scram	Specific Activity ($\mu c/g$ Na)									
				Na^{22}	Na^{24}	Cr^{51}	$Zr\text{-}Nb^{95}$	Ru^{103}	Ru^{106}	I^{131}	Cs^{137}	$Ba\text{-}La^{140}$	Ce^{141}
R-24	MEF*	10/2/58	9/25/58	6.2(-3)†	1.9(-1)	4.0(-2)	---	5.9(-4)	2.2(-3)	5.1(-3)	5.1(-4)	---	---
R-27	Reactor Pool	2/6/59	1/29/59	1.2(-2)	---	---	5.2(-2)	8.0(-4)	---	1.6(-2)	6.5(-4)	2.8(-4)	5.4(-4)
R-32	MEF	4/14/59	4/6/59	---	---	---	2.9(-3)	---	---	4.0(-2)	1.5(-2)	---	4.3(-2)

*Material Evaluation Facility

†Values in parentheses indicate power of ten by which preceding numbers should be multiplied.

3. Miscellaneous Items of Radiological Interest

a. Primary System Components

Normally access to the main primary gallery is not attempted until at least two weeks after reactor shutdown which permits sufficient decay of the Na^{24}. However, a gradual buildup of long-lived radiation fields in the gallery was noted as operation of the reactor continued. At the last measurement (April 18, 1959) prior to run 14, the radiation levels ranged from 50 to 420 mr/hr. Since at the time of these measurements the primary sodium had been drained to fill tank and no significant decay occurred during the next 5 days, it is probable that some degree of long-lived activity accumulation was taking place on the piping surfaces although piping activation from neutrons leaking from the core was also a factor.

The atmosphere of the main primary gallery (nitrogen) was sampled occasionally for radioactivity. The usual levels were $\sim 1 \times 10^{-5}$ $\mu c/cc$.

Radiation measurements were also taken periodically near the primary system cold trap. Since this trap is not operated continuously, the quantitative significance of these data is difficult to interpret, but it does serve to indicate relative amounts of foreign radioactivity removal from the primary sodium. The levels measured prior to run 14 usually varied from a few mr/hr to about 1 r/hr at ~1 ft from the surface, after the decay of Na^{24}.

TABLE III

TYPICAL RADIOACTIVITY ANALYSIS OF SRE COVER GAS PRIOR TO RUN 14

Date Sample Taken	Specific Activity (μc/cc)
12/11/58	1.9×10^{-3}
12/15/58	1.0×10^{-3}
2/14/59	1.7×10^{-3}
2/26/59	6.0×10^{-2}*
5/26/59	1.7×10^{-3}
6/13/59	1.8×10^{-5}
6/16/59	1.8×10^{-5}
6/20/59	3.8×10^{-5}

*Taken shortly after reactor shutdown

b. Routinely Occupied Areas of the Reactor Building

Radiation levels in routinely occupied areas in and around the reactor building are normally quite low; however, there have been specific instances of temporary relatively high radiation fields and/or airborne radioactivity. Often these have been attributable to some particular operation, such as fuel handling. On several occasions, however, above normal airborne radioactivity was detected in the high-bay area over the reactor loading face during routine operation of reactor. It is likely that some small leakage of the cover gas volume usually existed through the reactor loading face complex into the high-bay area. This slow leakage coupled with the comparatively low levels of radioactivity existing in the cover gas normally precluded detection after mixing with the large volume of air above. Occasionally, increased activity levels in the cover gas, perhaps coupled with increased leakage rates, probably resulted in the appearance of relatively high airborne activity in the reactor floor area.

II. FISSION PRODUCT DATA SUBSEQUENT TO JULY, 1959 SHUTDOWN

During the course of power run 14 on the SRE, a number of fuel cladding failures occurred. As a result of these failures, significant quantities (estimated at 5,000 to 10,000 curies) of fission products were released to the primary coolant system. Following the shutdown of the reactor on July 26, 1959 recovery procedures were immediately undertaken. In addition, numerous system modifications[5] were made during the recovery interval leading to the reattainment of criticality with a thorium-uranium fuel loading on September 7, 1960.

It is apparent the vast majority of the fission product release to the primary coolant took place during run 14, July 12 to 26, 1959. Principally based on evidence of radioactivity in the gas decay tanks and occurrences of high airborne radioactivity in the operating area, it would appear that at least some and perhaps most of the release occurred during the first few days of this period. However, since reactor operation continued until 1120 on July 26, this latter date will be used as the zero decay point for fission product contamination calculations. A complete description and analysis of the events leading up to the fuel element damage and of subsequent recovery operations may be found in the two reports issued by the Atomics International Ad Hoc Committee.[1,2]

A. PRIMARY SODIUM SAMPLES

The Na^{24} activity in the primary coolant delayed the procurement of the first sodium sample until August 2, about 7 days after reactor shutdown. It should be noted that during this interval, circulation and cold-trapping of the primary sodium continued; thus the loss of some portion of the fission product activity originally present prior to obtaining the first sample was certainly possible. Subsequent sodium samples were taken at irregular intervals, principally because of the inconvenient methods available and a necessary compatibility with recovery operations. Table IV presents a tabulation of the results of the fission product analyses of the initial sodium sample and also compares the fraction of each isotope released based on calculations of the total reactor inventory as of July 26, 1959.

TABLE IV

INITIAL FISSION PRODUCT ANALYSIS OF SRE PRIMARY SODIUM

Isotope	Pri. Coolant Act. (μc/g Na)*	Total Coolant Inv. (curies)*	Total Reactor Inv. (curies)*	Fraction of Inv. Released†
Cs^{137}	1.26	2.77×10^{1}	8.70×10^{3}	3.18×10^{-3}
Cs^{134}	0.02	$\sim 4 \times 10^{-1}$	2×10^{2} §	2×10^{-3} §
Sr^{89}	20.0	4.44×10^{2}	1.60×10^{5}	2.78×10^{-3}
Sr^{90}	0.97	2.14×10^{1}	8.15×10^{3}	2.63×10^{-3}
I^{131}	0.74	1.63×10^{1}	1.68×10^{4}	0.97×10^{-3}
Ce^{141}	4.38	9.65×10^{1}	1.27×10^{5}	$0.76 \times 10^{-\cancel{3}\,4}$
Ce^{144}	5.18	1.41×10^{2}	1.69×10^{5}	~~0.67 x 10~~$^{-3}$ 8.3×10^{-4}
$Ba(La)^{140}$	1.65	3.63×10^{1}	5.61×10^{4}	$0.65 \times 10^{-\cancel{3}\,4}$
$Zr^{95} + Nb^{95}$	13.9	3.06×10^{2}	5.53×10^{5}	$0.55 \times 10^{-\cancel{3}\,4}$
Ru^{103}	0.95	2.09×10^{1}	7.52×10^{4}	0.28×10^{-3}

*As of July 26, 1959

†Multiply values in this column by ~3 to adjust fraction released to average values for those fuel elements which suffered cladding failures (14 of 43 elements failed).

§From neutron capture in Cs^{133}; estimated.

The next sodium sample was obtained on October 16. The fission product analysis of this sample indicated surprisingly large decreases in contamination, in some cases greater than a thousand-fold. However, subsequent samples showed that, for some reason, this second sample was not representative. The data from this sample are listed in the appendix for information only. The third sodium sample was taken on October 31, 1959. Since approximately 3 months elapsed between this sample and the initial one, it is interesting to compare the decrease in fission product contamination beyond that due to radioactive decay. Table V presents such a comparison for these isotopes identified in both samples.

Following the procurement of the October 31 sodium sample, extensive efforts followed in the next few months to cleanse physically the primary sodium of foreign matter, principally of the carbonaceous material formed by the decomposition of tetralin. These procedures included draining the entire primary system into the drain-and-fill tank through a specially constructed wire mesh filter, vacuuming the carbonaceous material from the top of the moderator cans, and prolonged sodium circulation accompanied by cold trapping. The last sodium sample taken for radioactive analysis prior to startup of the reactor with the new fuel loading was obtained on July 26, 1960. The results are given in Table VI.

TABLE V

FISSION PRODUCT CONTAMINATION DECREASE IN PRIMARY SODIUM (August 2 to October 31, 1959)

Isotope	Fission Product Contamination ($\mu c/g$ Na) Aug 2	Oct 31 (predicted)*	Oct 31 (actual)	Ratio $\frac{\text{Oct 31 (actual)}}{\text{Oct 31 (predicted)}}$
Cs^{137}	1.26	1.26	0.45	0.36
Cs^{134}	0.02	0.02	0.006	0.3
Sr^{90}	0.97	0.97	0.060	0.062
Sr^{89}	19.0	5.8	0.25	0.043
I^{131}	0.42	0.00019	0.00012	0.63
Ce^{141}	3.7	0.54	0.000088	0.00016
Ce^{144}	5.1	3.9	0.00031	0.00008
Zr^{95} Nb^{95}	13.0	5.2	0.0067	0.0013
Ru^{103}	0.85	0.19	0.0045	0.024

*On the basis of radioactive decay of August 2 sample.

TABLE VI

FINAL RADIOACTIVITY ANALYSIS OF SRE PRIMARY SODIUM PRIOR TO STARTUP WITH NEW CORE LOADING

Isotope	$T_{1/2}$ (years)	Concentration ($\mu c/g$ Na)*
Na^{22}	2.6	1.2×10^{-2}
Cs^{137}	30	2.8×10^{-2}
Sb^{125}	2.7	6.1×10^{-2}

*As of 7/26/60

The disappearance of the I^{131}, $Ba\text{-}La^{140}$ Ce^{141}, and to a lesser extent, $Zr\text{-}Nb^{95}$, from this final sample would be expected from natural radioactive decay during this interval of about one year. However, the majority of the decrease in the Ce^{144}, Ru^{103} (both undetectable in final sodium sample), and Cs^{137} must be attributable to other causes. Although a strontium analysis was not made on this final sample before startup, its decrease over and above radioactive decay was established by earlier samples (see Table V).

B. COVER GAS SAMPLES

The first helium cover gas sample during run 14 which gave an indication of unusually high radioactivity was taken on July 18. Since the radiation level at the surface of the 2-liter sample chamber was ~30 mr/hr, no further attempt was made to assay the concentration quantitatively. However, assuming that the activity was principally Xe^{133} + Xe^{135}, a rough estimate would place the level at 2 to 10 μc/cc. It is difficult to interpret cover gas samples subsequent to the July 26 shutdown since bleeding and flushing operations to the gas decay tanks and out the stack were almost immediately commenced. A sample taken on August 12, indicated a gross level of about 1 μc/cc. A gamma spectrometer scan on September 14, of this sample identified the principal remaining contributions as Xe^{133} and Kr^{85}. Extrapolating the decay of these two isotopes back to shutdown on July 26, yields ~7.4 μc/cc for Xe^{133} and 0.016 μc/cc for Kr^{85}. The presence of shorter lived rare gas isotopes probably created a higher gross level at shutdown, but there is some evidence that an appreciable time delay exists between release of the fission gases from the fuel and their escape to the cover gas volume. The Xe^{133} value in this sample is in rough agreement with the estimate made from the sample taken on July 18. Using the Xe^{133} and Kr^{85} values obtained from the August 12 sample, an attempt was made to estimate the fraction released from the core. Table VII presents these data.

The estimated release fractions, although comparable to those non-gaseous elements determined from sodium samples, are probably somewhat lower than actually occurred. Through radioactive decay of the Xe^{133} and cover gas purging operations, the fission product contamination level in the reactor cover gas was reduced to 4×10^{-3} μc/cc (Kr^{85}) by September 29, 1959.

TABLE VII

FISSION PRODUCT CONTAMINATION IN COVER GAS

Isotope	Cover Gas Conc.* (μc/cc)	Total Cover Gas Inv.*† (curies)	Total Reactor Inv.* (curies)	Fraction Released
Xe^{133}	7.4	4.7×10^{1}	5.08×10^{4}	0.92×10^{-3}
Kr^{85}	0.016	2.0×10^{-1}	1.1×10^{3}	0.18×10^{-3}

*As of shutdown 7/26/59.
†Includes a factor of two for probable losses due to pressure manipulations on July 18.

C. PRIMARY COOLANT PIPING SAMPLES

Since it became apparent soon after the reactor shutdown that large amounts of fission product contamination in the primary sodium were being removed by means other than radioactive decay, deposition on piping walls was investigated. Some of the deposition appeared to involve simple settling in low velocity portions of the sodium system, as evidenced by radiation surveys following the draining of the primary system. Of perhaps more interest was the deposition which involved a "plating" type mechanism that could not be removed by simple flushing. This type of activity deposition could have an important bearing on the accessibility of certain areas for some time to come. In general, the piping samples were obtained from hot and cold trap areas and consisted of short sections of 1-1/2 in., schedule 40, Type 304 stainless steel pipe which were removed by hack-sawing while filled with frozen sodium. In the analyses the frozen sodium was dug from the pipe as well as possible. Next, any residual sodium was dissolved in methanol and water. Finally, the pipe was subjected to a series of hydrochloric acid etches, with dimensional and weight checks between each etch. The sodium, methanol wash, and etch solutions were each analyzed using a 256-channel gamma spectrometer. Chemical separations were made on some of the etch solutions for strontium analysis by beta counting. Since the amount of fission product deposition in any one region of piping is certainly some function of the exposure conditions (e.g., flow rate and temperature, total volume of flow, position in system, etc.) no attempt to extrapolate quantitatively to the total piping system from the few isolated samples taken was made. However, the relative deposition between isotopes of different elements is also of interest and probably less sensitive to variance in flow conditions. An example of an etch solution analysis for the "fixed" (i.e., not removed by the methanol wash) pipe contamination is shown in Table VIII.

TABLE VIII

EXAMPLE OF "FIXED" SRE PRIMARY-SODIUM FISSION-PRODUCT PIPING CONTAMINATION

Isotope	Contamination Level ($\mu c/cm^2$)*	Ratio*† $\frac{\mu c/cm^2 \text{ of pipe surface}}{\mu c/cc \text{ of primary sodium*}}$
Sr^{89}	15.2	0.76
Sr^{90}	0.78	0.80
$Zr\text{-}Nb^{95}$	2.7	0.19
Ce^{144}	2.1	0.41
Cs^{137}	0.022	0.018

*Corrected for radioactive decay since July 26, 1959 for comparative purpose. The actual contamination levels of Sr^{89} and $Zr\text{-}Nb^{95}$ may never have been this high.

†Sodium contamination level as determined in first sample after reactor shutdown.

It should also be noted here that certain non-fission product contamination on the piping walls was also identified, principally Mn^{54}, Co^{60}, and Fe^{59}. The levels of these isotopes were roughly comparable to that of the Cs^{137} given in Table VIII ($\sim 10^{-2}$ $\mu c/cm^2$ as of 7/26/59). Figure 4 is a plot of the fission product data obtained from this same piping sample as a function of the depth of successive etches.

D. OTHER MEASUREMENTS AND SAMPLING DATA

1. Uranium

Since it was planned to continue the use of the primary sodium for future reactor operation, it was important to ascertain the amount of uranium which

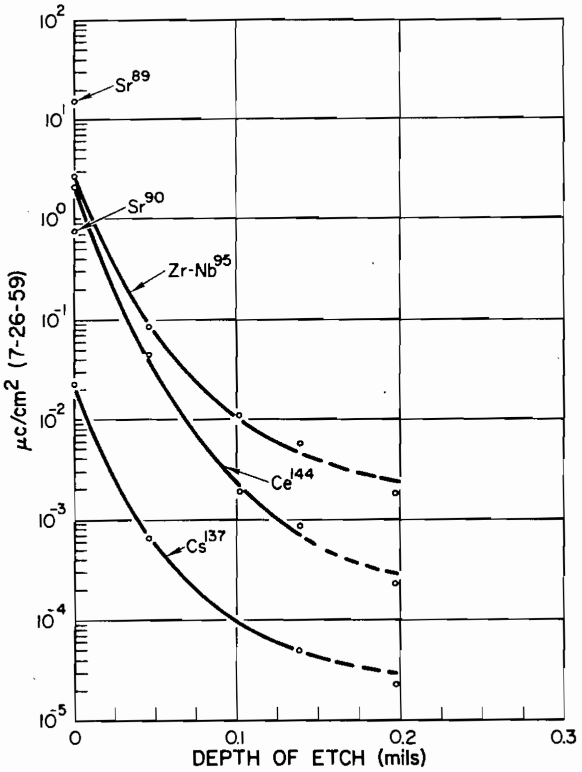

Figure 4. Fixed Fission Product Contamination Remaining on SRE Primary Piping vs Depth of HCL Etch

may have contaminated the sodium as a result of the fuel element damage. The principal concern was over the effect that uranium contamination might have on the future accessibility of the primary system during periods of reactor shutdown. Such contamination could result in high level fission product deposition in the piping system. A secondary concern was the possibility of creating an abnormally high Xe and Kr background activity level in the helium cover gas, thus limiting the sensitivity of the continuous cover gas monitor, which was installed during the recovery and modification effort. Preliminary calculations indicated that while 10 ppm of uranium (2.8% U^{235}) probably could be tolerated, 1 ppm or less would be highly desirable. An analysis conducted by Argonne National Laboratory showed a concentration of less than 0.2 ppm, which was considered negligible.

2. Non-Fission Product Radioactivity

The Mn^{54}, Co^{60}, and Fe^{59} contamination on the primary piping system was mentioned earlier. After appreciable decay had taken place, small quantities of Sb^{125} were also identified. Since the fission yield of this isotope is quite small (0.024%), the primary source is probably due to neutron activation of Sn^{124} impurity in the sodium and its subsequent decay to Sb^{125}. The only other significant non-fission product activity was that due to the formation of Na^{22} in the primary sodium (~1 x 10^{-2} μc/g Na as of October 1959).

3. Carbonaceous Material

It has been estimated that from 7 to 70 lb of carbon or carbonaceous material may have been deposited in the primary sodium as the result of tetralin leakage prior to run 14. A large fraction of this amount maintained its existence as solid carbon particles throughout the primary system, with some settling occurring in low velocity flow areas. Almost all of the sodium samples obtained were contaminated with small flecks of carbon. These particles were filtered from the sodium samples after dissolving and analyzed separately. In some cases, carbonaceous samples were obtained directly from the top of the moderator cans after lowering of the sodium pool level. Radioactivity analysis of this material showed a much higher concentration of fission product contamination per unit weight than in the filtered sodium. Table IX gives the results from a typical sample and, for comparison purposes, also indicates the corresponding contamination of the accompanying sodium sample.

TABLE IX

RADIOACTIVITY ANALYSIS OF TYPICAL SAMPLE OF CARBONACEOUS MATERIAL FROM PRIMARY SODIUM SAMPLE

Isotope	Activity of Carbonaceous Material* (μc/g of particles)†	Activity of Sodium (μc/g Na)†
Cs^{137}	1.1 x 10^{3}	0.45
Cs^{134}	2.9 x 10^{1}	0.006
Ce^{141}	2.3 x 10^{2}	0.0007
Ce^{144}	2.9 x 10^{2}	0.00039
$ZrNb^{95}$	2.2 x 10^{2}	0.018

*Obtained from reactor on 10/31/59
†As of 7/26/59

Since the weight of the carbon particles in the above sample was quite small (0.1 to 0.2 mg), appreciable error may exist in the specific activity determination. A larger sample taken from the top of the moderator cans about two months later gave even higher results as shown in Table X.

TABLE X

RADIOACTIVE ANALYSIS OF CARBONACEOUS MATERIAL TAKEN FROM TOP OF MODERATOR CANS

Isotope	Activity ($\mu c/g$)*
Zr Nb^{95}	6.8×10^3
Cs^{137}	5.0×10^3
Cs^{134}	1.3×10^2
Ce^{144}	3.6×10^3

*As of 7/29/59

4. Radiation Surveys of Primary Piping System and Components

Radiation surveys of the primary system during reactor operation and for a week or two after shutdown were not feasible because of the intense Na^{24} radiation. Consequently, no data on these areas were available until about the second week in August. One of the more interesting locations is the primary cold trap. Although radiation measurements are not routinely taken in this area, considerable data prior to run 14 are available. The cold trap is usually operated when the oxygen content in the coolant exceeds 10 ppm. Cold trapping was initiated during run 14. The first radiation measurement after the reactor shutdown from this run was not made until August 8. It was immediately apparent from this measurement that significant quantities of radioactivity were being deposited in the cold trap since measurements from other areas in the primary system were considerably lower. The variation of the cold trap dose rate was followed thereafter until its removal from the system. Table XI summarizes these measurements. Assuming an average gamma ray energy of 0.7 mev, it was estimated that the cold trap contained of the order to 150 to 200 curies at the time of the maximum radiation level (~80 r/hr at 2 in. from surface).

Following the draining of the sodium from the primary system into the drain-and-fill tank, radiation surveys were made of various portions of the primary piping galleries. It was found that the general radiation levels increased somewhat upon sodium draining, thereby indicating the dominance of deposited radioactivity over that actually contained by the sodium. A closer inspection of the radiation fields also showed a marked non-uniformity along sections of piping. The levels ranged from a few hundred mr/hr to a few r/hr.

TABLE XI
RADIATION LEVELS FROM PRIMARY COLD TRAP

Date (1959)	Dose Rate* (r/hr)
April 5	1†
August 8	63
August 12	63
August 13	81
August 17	72
August 18	68
August 25	80
August 31	70
September 2	73
September 9	65
September 11	63
September 14	65
September 16	69
October 5	50

*At ~2 in. from surface of cold trap

†Prior to April 5, 1959, all dose rates measured were less than 1.5 r/hr, usually a few hundred mr/hr.

III. ESTIMATING FISSION PRODUCT RELEASE TO PRIMARY COOLANT AND COVER GAS

Not only is the absolute magnitude of the gross fission product release from the fuel of interest, but also the relative release of the different elements. This latter aspect could produce some insight into the type of activity release mechanism(s) involved, that is, a higher release percentage of the more volatile elements would suggest the dominance of a diffusion mechanism, while a more evenly spread release would favor a non-temperature dependent mode.

If we examine the data from the first sodium sample (Table V) and neglect for the moment the gas samples, some degree of volatility-release fraction ranking seems to exist; that is, Cs, Sr, and I have the highest release fractions, with the less volatile Ce, Ba, Zr, and Ru at somewhat lower values. In the cases where two isotopes of the same element were identified (Cs, Sr, and Ce), the release fractions are in reasonably good agreement. However, some anomalies do exist. One would expect the release fraction of iodine to be considerably higher than that of cesium rather than one-third of it as actually found. Also, on a volatility basis, the barium component would rank appreciably higher than the cerium and zirconium. A possible answer to the low iodine value would be the escape of this element from the primary sodium to the cover gas system. However, iodine has never been found in the cover gas of the SRE either after this incident or in any of the many other gas samples taken during the operation of the reactor. Although it may be possible that small amounts of iodine could exist undetected in the gas when accompanied by high concentrations of xenon and krypton, certainly not the quantities necessary to produce such relatively low values as actually found in the sodium.

In consideration that the first sodium sample was obtained about 1 week after the reactor was shutdown (possibly 3 weeks after the major part of the fission product release), and further, that large amounts of activity deposition occurred in the primary system, this sample may not present a true picture of the initial fission product release distribution. With the limited data available and the varying conditions of primary sodium flow that existed, it is not feasible to adjust this first sample quantitatively for deposition prior to its obtainment. However, it is apparent that the relative deposition of Ce, Zr, Ru, and Sr was much greater than that of Cs or I. This appears reasonable since cesium is quite soluble in sodium, as is NaI, the most probable product of any iodine released to the sodium. A qualitative allowance for this deposition prior to the first sodium sample would tend to decrease the actual elemental spread in the release fractions and bring them closer to the cesium value.

Unfortunately, the role of carbonaceous material in removing activity from the sodium is even more difficult to assess in a quantitative sense. It would appear that it is relatively non-selective in an elemental sense, concentrating the fission products to a degree roughly proportional to their concentration in the sodium. Taking the upper weight estimate of the carbonaceous material ($\sim$70 lb) and assuming an average gross activity of about 1 x 10^3 μc/g at the time of obtaining the first sodium sample, the total activity contained in this manner is $\sim$32 curies. An estimate of the total longer-lived fission product activity dissolved in the sodium from the initial sample is $\sim$1000 to 2000 curies; thus the carbon removal would not seem to be a dominant factor as far as accounting for the total activity initially released to the sodium is concerned.

Adjusting the fission product release fractions to consider only the estimated inventory of those fuel elements which actually suffered cladding damage and applying somewhat arbitrary factors to account for deposition prior to obtaining the first sodium sample, the following estimate of the initial average fission product release from the damaged elements was derived: Cs and Sr - 1%; I, Ba-La, and Ce - 0.7%; Zr-Nb - 0.4%; Ru - 0.2%.

Although certain anomalies in this ranking exist on an element volatility basis (e.g., the high release fraction of strontium, the relatively low release fraction of iodine), the overall order suggests some degree of volatility dependency. It was initially felt that since examination of the recovered fuel slugs from damaged elements showed no evidence of significant melting, that the majority of the fission product contamination may have originated from the loss of the NaK bonding fluid from these elements. Previous fission product contamination measurements made of this bonding fluid from undamaged SRE elements have indicated a level no greater* than one would expect from fission recoil process alone. That is, diffusion or other possible modes of contamination

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*Actually, only the Cs^{137} and I^{131} contamination was found in approximately the theoretical amount. Zr^{95}, Ce^{141}, and Sr^{90} were present at about 10%, 1% and 0.1%, respectively, of the calculated values, thus indicating a deposition or plating inside the fuel rod cladding of a nature similar to that which was observed in the primary sodium system.

apparently were not significant. On this basis, the release fraction from the 3/4 in. diameter metallic uranium slugs is calculated at about 8×10^{-4}. Thus, it would appear that this source could account for less than 10% of the estimated Cs^{137} activity in the primary sodium, and at best, about 40% of the Ru^{103}. Moreover, such a spread in release fractions by elements as observed in the sodium sample would not be expected if recoil processes were the primary sources of contamination.

Severe overheating of some of the fuel elements is known to have existed. Some melting of uranium involved in the Fe-U eutectic formation also occurred. Many of the fuel slugs were badly swollen, cracked and spongy. It may well be that these effects were sufficient to allow the release of the majority of the measured amounts of fission product contamination by diffusive processes. Moreover, the conditions conducive to diffusion may have persisted over a fairly long time during the last reactor run, perhaps up to two weeks. It is also possible that the fission product release from a few of the badly damaged elements raised the overall average release appreciably.

Extrapolations of measurements made of fission product contamination in the helium cover gas seem of little additional value in determining release fractions. The more mobile nature of the cover gas system (i.e., flushings, pressure changes, etc.) over the period prior to sampling makes any corrective factors applied highly uncertain. However, the amounts actually measured, even with a liberal allowance for losses and dilution, seem significantly lower than would be expected on a relative volatility basis. This result may be due in part to the capture of the gases in the void space provided at the top of the fuel rod cladding enclosure. Since most of the failed fuel elements' claddings parted at or below the mid-point, not all of the previously accumulated gas would have escaped. If such a capture did occur, it would be more important for Kr^{85} than Xe^{133} since more than 93% of the Kr^{85} inventory was produced prior to the last run. Thus, lower release fraction measured for Kr^{85} ($\sim$20% of that for Xe^{133}) may be partially a result of such an event.

IV. SUMMARY AND CONCLUSIONS

The data obtained subsequent to the SRE fuel damage incident have provided considerable insight on the movement and retention of certain fission products which were released to the primary sodium. The limited sampling means* and the overall necessity for compatibility with the reactor recovery efforts imposed severe restrictions on much of the quantitative value of the data. Nevertheless, a number of conclusions can be drawn from the experience.

a) Although significant fuel melting did not occur, the release fractions of the various fission products to the primary sodium indicate that some degree of volatility preference was involved.

b) Only Xe and Kr isotopes were identified in the reactor cover gas system. This confirms the previously held premise that the sodium coolant forms an effective trapping agent for all but the rare gas isotopes.

c The iodine release fraction seems unusually small on a volatility basis relative to the other elements. Since neither deposition in the primary system nor escape to the cover gas occurred in detectable quantities, the low sodium value for iodine remains unexplained.

d) The carbonaceous particulate material resulting from the decomposition of the tetralin leakage proved to be an effective fission product scavenger. Specific activities of this material ranged from 10^3 to 10^4 times that of the filtered sodium in which it was contained. This concentration appears to increase with exposure time and to be generally non-selective with regard to those fission products measured.

e) The cold trap located in the primary system was effective in removing fission product contamination. Whether this removal occurred principally by the collection of the contaminated carbonaceous material or of dissolved fission products directly is not presently known, but both processes appear likely.

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*The inadequacy of the sodium and cover gas sampling methods was recognized and this situation has been corrected by the installation of a continuous cover gas monitor and a much improved primary sodium sampler at the SRE.[5]

f) Appreciable deposition of fission product contamination occurred throughout the primary piping system. In this type of activity removal, a marked element selectivity is evident. Sr, Ce, and Zr-Nb deposition (relative to their estimated original concentrations in the sodium) being respectively about 500, 25, and 12 times greater than that of Cs and I. On an absolute basis, the gross depletion of fission product activity in the primary sodium by all modes other than radioactive decay is approximated as follows:

Element (Isotope)	Approx. Decontamination Factor From Na
I (131) Cs (134, 137)	Small (<10)
Ru (103) Zr-Nb (95)	Large (10^2 - 10^3)
Ce (141, 144) Sr (89, 90)	Very Large ($>10^3$)

g) In the overall analysis of the consequences of the fission product release to the primary system, it is important to recognize that:

(1) Although about 5,000 to 10,000 curies of fission product activity was unexpectedly released to the primary sodium system, no radiological emergency of any nature occurred. The activity release from the primary containment system (core vessel and piping) was restricted to very nominal amounts of rare gas isotopes. Neither operating personnel nor the reactor environs were subjected to any significant radiation exposure.

(2) Recovery operations subsequent to reactor shutdown were completed at a reasonable speed well within the accepted personnel exposure limits.

(3) The fission product contamination level of the primary sodium decreased to an acceptable level during the recovery period by natural radioactive decay and deposition, without the employment

of extensive decontamination operations (other than certain techniques used to remove the particulate carbonaceous material).

(4) In spite of rather extensive fission product deposition in the primary system, future access to this area (following the normal waiting period for Na^{24} decay) is not expected to be seriously hampered by long-lived radiation fields. However, this deposition, especially the Sr^{90} component, may well affect future procedures which involve opening the primary sodium system. As for long-lived activity still dissolved in and moving with the primary sodium, only Cs^{137} and possibly Sb^{125} still are present in significant quantities. These exist in concentrations of the same order as the unavoidable Na^{22} activity ($\sim 10^{-2}$ $\mu c/g$) and thus their presence should not appreciably affect future operations.

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APPENDIX

The fission product analysis of the sodium sample taken from the reactor on October 16, 1959 is presented in Table XII. The variance of this sample from the contamination trend of all the other samples is not understood, but probably reflects the difficulty in obtaining representative coolant samples under the conditions existing at the time.

TABLE XII

FISSION PRODUCT ANALYSIS OF SRE PRIMARY SODIUM
(10-16-59)

Isotope	Concentration in Primary Coolant (μc/g Na)*
Sr^{89}	2.9×10^{-3}
Sr^{90}	1.7×10^{-4}
$Zr^{95} + Nb^{95}$	3.3×10^{-2}
Cs^{137}	4.0×10^{-2}
Ce^{141}	1.1×10^{-2}
Ce^{144}	1.2×10^{-2}

*As of July 26, 1959