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FISSION PRODUCT RETENTION IN SODIUM -
A SUMMARY OF ANALYTICAL AND
EXPERIMENTAL STUDIES
AT
ATOMICS INTERNATIONAL

AEC Research and Development Report

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ATOMICS INTERNATIONAL

By
W. P. KUNKEL

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ABSTRACT

A variety of experimental and analytical studies have been performed at Atomics International for the purpose of evaluating the release and transport characteristics of fission products in a molten sodium environment.

Injection methods have included fission product simulants, equilibrium temperature studies using diffusion of fission products from preirradiated fuel, nuclear-transient melting of preirradiated fuel in a sodium environment, and melting by condenser discharge. Sampling methods have included, in addition to simple filters and gas collection devices, application of May pack and diffusion tube sampling techniques. Results indicate that the presence of sodium vapor affects the collection efficiency of some of these devices, and mitigates chemical and physical identification of the fission product effluent.

Equilibrium experimental results for iodine show some agreement when expressed as "distribution coefficients" and compared with a theoretical distribution coefficient derived from $\text{Log } K_d = 4500/T - 5.66$, where K_d is the distribution coefficient, and T is the system temperature in degrees Kelvin. Cesium and strontium experimental results show fairly logical distribution coefficients, although the theoretical correlation for these isotopes is not yet available.

Results of one series of iodine release experiments in which iodine vapor is released under a column of molten sodium show a sharp departure in terms of the distribution coefficient, differing from the equilibrium situation by over four orders of magnitude. The absence of appropriate mixing capability in these cases may be partially responsible.

Extensive plating and deposition on surfaces was observed throughout the experiments. The results of an examination of previously contaminated sodium reactor piping indicated some correlation of fission product activity with carbonaceous impurities in the system.

Application of these results to typical reactor configurations is made with the extrapolation equation developed. Release fractions are suggested as a result of this preliminary model.

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During the latter phases, the project was supported by the Nuclear Safety Research and Development Branch, Division of Reactor Development and Technology of the Atomic Energy Commission, and was under the direction of S. J. Szawlewicz.

This summary report is a compilation of several experimental and analytical efforts. The assistance and contributions of the principal contributors, whose names appear in the text, are gratefully acknowledged. In addition, the detailed review of the draft document by Dr. Hans Hamester of the Research and Development Branch provided invaluable direction to the project effort.

I. INTRODUCTION

In 1959, following the partial meltdown of the Sodium Reactor Experiment (SRE), it was observed that the iodine released from that portion of the core which had undergone melting was not observed in the reactor cover gas plenum.

It was also noted that several other fission products showed a greater reduction in concentration in the cover gas than the radioactive decay would have predicted.

Some experiments were planned to investigate these phenomena and it is from these experiments that a project and the related work at Atomics International has evolved. This project (Fission Product Retention in Reactor Coolants) has primarily focused its attention upon the retention characteristics of sodium.

The earliest studies at Atomics International were accomplished by introducing appropriate fission product simulants into liquid sodium either as salts of cesium nitrate, sodium iodide, or strontium chloride; or as molecular iodine in an inert carrier gas of nitrogen.

In the meanwhile, work by Parker of ORNL,¹ Castleman at BNL,² and others pointed out the significant differences in fission product behavior which arose when fission products are released in different form and into different environments. It was considered advisable, therefore, at an early phase in the project to study the retention characteristics of sodium in a more realistic fashion by injecting fission products into sodium by nuclear heating of foils of U²³⁵ in the KEWB Pulsed Reactor.⁽³⁾ Thus, a variety of experimental injection techniques have been employed ranging from the chemical injection discussed above through in-pile capsule irradiations, and condenser discharge devices to melt and possible vaporize a small uranium foil.

The original sampling activities of the project were conducted with charcoal traps, millipore filters, and to some extent with liquid traps designed to trap fission product contaminants of the sodium vapor-helium stream. As further information became available from the studies underway at ORNL,⁴ UKAEA,⁵ and BNL,⁶ several adaptations of current filtration devices were utilized in the studies. Most of these were adapted from either uranium fuel melting experiments in air, helium, or steam, and were thus not specifically intended to study fission product components in a sodium vapor-helium carrier.

The approach used in these studies was largely of an engineering nature and was intended to demonstrate the retention characteristics of sodium coolant under a variety of conditions such that the resulting data could be employed by those working in the area of safeguards analysis. It is the purpose of this report to correlate and summarize the data and information obtained from the analysis of the experimental and analytical efforts which have been conducted at Atomics International.

A portion of this report also deals with the general problem of radioactivity release and transport in sodium, and the application in a preliminary form of these results to the safeguards problems which exist for the reactor safeguards analyst.

II. SPECIFIC EXPERIMENTS AND RESULTS

A. OUT-OF-PILE STUDIES

1. Introduction of Cesium, Strontium, and Iodine Into Sodium (R. S. Hart, C. T. Nelson)⁷

An experimental effort was initiated to determine the retention ability of sodium for Cs^{137} , Sr^{89} , and I^{131} . These three isotopes were chosen partly because of their importance in assessing reactor hazards resulting from radioactivity release, and partly because of their relatively high volatility.

The apparatus for this experiment was designed to "salt" a given volume of sodium with known quantities of Cs^{137} , Sr^{89} , and I^{131} , raise this mixture to a series of temperature plateaus in a helium atmosphere, and measure the fraction of the radioactivity released from sodium to the helium cover gas.

The sodium handling system consisted of a retort where the actual heating and measuring took place, a drain and fill tank to contain the sodium before and after the experiment, and a helium ballast tank to provide pressure for moving the sodium between the portions of the system. The retort and drain and fill tank both contained a tube into which an eddy-current probe was inserted to measure the sodium level. In addition, the retort was fitted with a sodium sampling facility. Figure 1 shows the sodium handling system before installation of heaters and thermocouples. This entire apparatus was insulated with Kaylo insulation and fiberglass.

The temperature control and measurement system consisted of two main systems heaters with controllers, 17 auxiliary heaters for various parts of the system, and 48 thermocouples read by an auxiliary multipoint recorder.

Fission product salts (CsNO_3 , SrCl_2 , NaI) were obtained as carrier-free aqueous solutions with specific activities of 10 mc/ml. From 0.1 to 0.2 ml of the desired tracer solution was placed in the bottom cup of the sodium sampler and evaporated to dryness.

After the system was thus loaded with the fission product activity, the sodium temperature in the drain and fill tank was elevated to approximately 250°F and the tank was pressurized to force the sodium into the retort to a predetermined level and the fission product cup agitated to introduce the fission products into

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Figure 1. Hart-Nelson Out-of-Pile Fission Product Simulant Apparatus

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the sodium. At this point, the sodium in the system was heated to successive temperature plateaus ranging from 900 to approximately 1500°F in approximately 100°F steps.

Sampling of the sodium was accomplished by raising a sampling rod to which small planchets were affixed at various heights. One cup per plateau was removed from the sodium in the retort and retained in the sealed system until the run was completed, at which time it was removed and the activity level determined.

Gas sampling was accomplished by raising the pressure of the helium cover gas above the sodium level from a nominal 3 psig to 5 psig to prevent a partial vacuum in the gas space when the sample was drawn. The gas sample was then withdrawn through a heated stainless steel 1/4-in. tube into an evacuated flask where it was subsequently counted by NaI scintillator and multichannel analyzer. (An open window Geiger counter was used for counting the Sr^{89} samples).

The data resulting from the cesium, iodine, and strontium experiments are shown in Tables 1, 2, and 3 respectively.

2. Iodine Bubble Experiment (R. J. Begley)⁸

The iodine bubble experiment was not intended to describe or to predict the form of the iodine released from a fuel matrix upon fuel meltdown or vaporization.* It was intended rather as a scoping experiment to simulate the release of a bubble of inert gas from the gas space above the fuel in a typical fuel rod. At the temperature existing in a real reactor, the bubble may contain in addition to xenon and krypton, other volatile isotopes. These would presumably include iodine released from the fuel.

The fraction of iodine absorbed by sodium will be influenced by: (a) sodium temperature, (b) the distance a bubble rises through the sodium which may also be related directly to the reaction time, (c) the fraction of the inert diluent gases in the bubble which may also influence the reaction rate, and (d) the purity of the sodium. Since the sodium coolant in a typical reactor is maintained at an

*Fission-product-produced iodine may exist in either the free state or in a number of complex molecular forms with uranium, other fission products, or with other species present in the fuel or sodium such as alloying metals, cladding materials, impurities, etc.^{1,2}

impurity level of a few ppm by cold traps and other cleanup devices, the purity of the sodium was not included as a variable in this experiment. Sufficient data were obtained to assure a first order correlation of the test variables.

The first test which was performed was a bubble release test in which small capsules of gas were punctured at various levels under a pool of sodium. The capsules, which were fabricated either of 1-3/4 in.- or 1-1/2 in.-diameter tubing were approximately 1 in. long, and were sealed by flat stainless steel top and bottom sheets. To facilitate puncturing, the top sheet was of 0.006-in. shim stock. The capsule was first purged with nitrogen sweep gas through a 1/4-in.

TABLE 1
CESIUM RELEASE FROM MOLTEN SODIUM

	Sodium Temp. (°F)	Total Cs ¹³⁷ in Na* (d/m)	Total Cs ¹³⁷ in He (d/m)	Release Fraction $\frac{\text{Cs}^{137}(\text{He})}{\text{Cs}^{137}(\text{Na})}$
Run C-2	894	3.7×10^7	5.5×10^3	1.5×10^{-4}
	983	3.7×10^7	1.1×10^4	3.0×10^{-4}
	1130	3.7×10^7	1.5×10^4	4.1×10^{-4}
	1222	3.7×10^7	3.8×10^3	1.0×10^{-4}
	1305	3.7×10^7	3.1×10^3	8.4×10^{-5}
Run C-3	927	9.0×10^7	1.4×10^4	1.6×10^{-4}
	1212	9.0×10^7	1.8×10^4	2.0×10^{-4}
	1298	9.0×10^7	1.9×10^4	2.1×10^{-4}
	1446	9.0×10^7	1.1×10^4	1.2×10^{-4}
	1517	9.0×10^7	1.6×10^4	1.8×10^{-4}
Run C-4	968	3.0×10^7	9.7×10^3	3.2×10^{-4}
	1253	3.0×10^7	4.3×10^3	1.4×10^{-4}
	1327	3.0×10^7	2.8×10^3	9.3×10^{-5}
	1457	3.0×10^7	8.9×10^3	3.0×10^{-4}
	1515	3.0×10^7	2.3×10^3	7.7×10^{-5}

*Based on average of sodium samples

TABLE 2
IODINE RELEASE FROM MOLTEN SODIUM

	Sodium Temp, (°F)	Total I ¹³¹ in Na* (d/m)	Total I ¹³¹ in He (d/m)	Release I ¹³¹ (He) Fraction I ¹³¹ (Na)
Run I-1	923	8.8×10^8	9.4×10^2	1.1×10^{-6}
	1112	8.8×10^8	4.5×10^3	5.1×10^{-6}
	1210	8.8×10^8	1.7×10^3	1.9×10^{-6}
	1305	8.8×10^8	3.3×10^3	3.8×10^{-6}
	1410	8.8×10^8	1.5×10^3	1.7×10^{-6}
Run I-3	927	4.8×10^8	6.0×10^2	1.3×10^{-6}
	1212	4.8×10^8	1.6×10^3	3.3×10^{-6}
	1298	4.8×10^8	1.3×10^3	2.7×10^{-6}
	1446	4.8×10^8	1.8×10^3	3.8×10^{-6}
	1517	4.8×10^8	5.7×10^2	1.2×10^{-6}

TABLE 3
STRONTIUM RELEASE FROM MOLTEN SODIUM

	Sodium Temp, (°F)	Total Sr ⁸⁹ in Na* (d/m)	Total Sr ⁸⁹ in He (d/m)	Release Sr ⁸⁹ (He) Fraction Sr ⁸⁹ (Na)
Run Sr-1	927	5.1×10^8	6.7×10^2	1.3×10^{-6}
	1212	5.1×10^8	1.9×10^2	3.7×10^{-7}
	1298	5.1×10^8	1.3×10^2	2.5×10^{-7}
	1446	5.1×10^8	8.6×10^1	1.7×10^{-7}
	1517	5.1×10^8	7.7×10^1	1.5×10^{-7}
Run Sr-2	968	5.2×10^8	1.3×10^2	2.5×10^{-7}
	1253	5.2×10^8	8.6×10^1	1.7×10^{-7}
	1327	5.2×10^8	8.8×10^1	1.8×10^{-7}
	1457	5.2×10^8	8.0×10^1	1.5×10^{-7}
	1515	5.2×10^8	7.2×10^1	1.4×10^{-7}

*Based on average of sodium samples

tube welded in the side, and preweighed iodine crystals were placed in the capsule. The nitrogen pressure in the capsule was reduced by evacuation to the values indicated in Table 4.

Following capsule closure, the iodine-nitrogen capsules were rigidly attached to rings which were in turn welded to a steel frame suspended in the test column. The test column consisted of a 4-in.-diameter pipe, 12 ft long, which included attached heaters, thermocouples, and insulation. A stainless steel rod operated through a sliding seal in the top plate. The bottom end of the rod terminated in a conical plunger which was used to rupture the capsule lid, and release the N_2 + iodine gas to the sodium pool. The depth of sodium in the test column was 10 ft, and the temperatures indicated in Table 4 were maintained constant with controllers.

The iodine- N_2 mixture was released from one capsule at a time by driving the plunger through each thin capsule cap. The released gas traveled up through the sodium column and entered the two-foot deep cover gas region above the sodium pool. Before each capsule was ruptured, sweep gas flow was initiated.

TABLE 4
DESCRIPTION OF PLANNED TEST CONDITIONS

Capsule	Depth Under Surface (ft)	N_2 Gas Diluent (wt %)	Sodium Temperature ($^{\circ}$ F)
1	6	50	500
2	6	50	1000
3	6	90	1000
4	10	50	500
5	10	50	1000
6	6	0	500
7	6	0	1000
8	6	50	500
9	6	50	1000
10	10	50	500

TABLE 5
IODINE BUBBLE TEST RESULTS

Capsule	Initial Iodine (mg)	Iodine in Filter (mg)	Fraction Released (10 ⁻²)
1	0.6	0.009	1.5
2	0.7	< 0.010	< 1.5
3	1.80	< 0.010	< 0.5
4	7.0	0.0013	0.01
5	8.0	< 0.010	< 0.01
6	14.2	0.023	0.2
7	14.6	< 0.010	< 0.01
8	60.0	0.60	1.0
9	60.8	< 0.010	< 0.01
10	59.8	0.14	0.2

The nitrogen cover gas was passed through a charcoal filter, and neutron activation techniques utilized to determine the total iodine in the charcoal filters.* Since the amount of iodine added to the sodium was a total of only 12 ppm, sampling of the sodium for iodine was not considered feasible. The results of the experiment are presented in Table 5.

After disassembly of the test apparatus, the surfaces enclosing the cover gas region were rinsed with water, etched with sulfuric acid, and rerinsed to remove any residual iodine. The amount recovered was negligible in comparison with the amount of iodine released. One factor which may have influenced the test results, however, is that a small amount of air entered the cover gas during the 1000°F tests, undoubtedly producing an oxidized surface, which may have materially reduced the extent of release from the sodium pool. (During another test in which sodium was heated in a beaker and iodine was allowed to bubble through it as the surface oxidized, the oxide was observed to impede the flow of iodine, at least as far as bubble escape was concerned).

*Sweep gas flow continued until the cover gas volume was displaced five times after the iodine-N₂ bubble reached the surface.

3. Iodine-Sodium Surface Reaction Rate Experiment (R. J. Begley)⁹

Since any iodine released from a vented fuel element or from a damaged fuel element in a sodium column would then be available to a much lesser extent for interaction in the vapor phase above the sodium, an experiment was performed to determine the rate of reaction or more explicitly to measure the rate of iodine loss from the vapor phase. This depletion probably results in large part from iodine absorption while existing in the gas phase above a sodium surface.

The surface reaction rate test was conducted by allowing gaseous iodine to pass from a reservoir through a control valve into a reaction vessel containing liquid sodium. The experimental apparatus, which is shown in Figure 2 is similar to that employed by Longton¹⁰ for studies of the reaction of alkali metals with various gases. This apparatus which was all steel consisted of a reservoir of 373-in.³ volume, a reaction vessel, precision gas gauges attached to the reservoir and to the reaction vessel, piping connecting the reservoir and the reaction vessel, and a flow control valve in the piping. The entire experimental complex was maintained at isothermal conditions to reduce iodine condensation.

A capsule shown in the sketch was evacuated and filled with 14 gms of iodine. This capsule was then welded into the system. It could be punctured from

Figure 2. Reaction Rate Experimental Apparatus

outside the system without contaminating the test atmosphere. The reaction vessel (a 2-in. pipe, 18 in. long) was loaded with 1.2 lb of sodium. An inert gas glove box was used to avoid oxygen contamination.

At room temperature, the entire test apparatus was evacuated to 25- μ pressure. The control valve was then shut and the heaters activated to raise the system temperature to 500°F for a several-hour period. Following temperature equilibration, the capsule containing the iodine was ruptured, thus introducing iodine vapor into the evacuated reservoir and raising the reservoir pressure to approximately 5.8 psia. The control flow valve was then opened and gaseous iodine flowed from the reservoir into the reaction vessel until the reaction vessel pressure reached 2 psia. The valve was manually manipulated throughout the remainder of the experiment to maintain the reaction vessel pressure constant at 2 psia. The rate of decay of pressure in the reservoir corresponds to the rate of reaction of the iodine at the sodium surface of the reaction vessel.

The results of this experiment are shown in Figure 3. Three results are of interest. It may be noted first that the pressure dropped off quickly as the flow control valve was opened and the iodine flowed into the reaction vessel. After this short period, the pressure dropped off linearly with time for approximately

Figure 3. Results of Reaction Rate Experiment

30 min. A pressure decay rate of 0.089 psi/min fits the data obtained. This corresponds to a reaction rate of 4.5 mole/hr-ft². At the conclusion of this experiment, the thermal insulation on the reaction vessel was stripped off and the vessel cooled quickly by blowing compressed air on the outside surface to reduce the probability of further reaction. After the vessel was removed from the apparatus, the contained sodium was analyzed for sodium iodide. Analysis of the sodium in this manner accounted for 9.05 gm of iodine in the sodium. Comparing this result with the value obtained from the pressure decay curve, which indicated that 9.10 grams of iodine were absorbed by the sodium, shows good agreement.

It is also interesting to note that the iodine was not distributed homogeneously through the sodium. The reaction vessel sodium was sectioned into two samples which were analyzed separately. A surface layer of 1.6 gm contained 19.8% iodine, while the remaining 560 gm of mixture contained an average of 1.56% iodine.

If the fate of iodine in the cover gas is controlled by diffusion, then the rate of iodine removal from the cover gas to the sodium can be predicted by solution of the diffusion equation for a diffusing gas through a stationary gas,

$$\frac{\partial}{\partial x} \frac{P_t}{P_s} D \frac{\partial c}{\partial x} = \frac{\partial c}{\partial t}.$$

In the case of this experiment, the concentration of iodine is so low that the ratio of the total gas pressure to the stationary gas pressure P_t/P_s is very near unity. This same condition would prevail in a typical reactor system, since the maximum total weight of iodine contained in all the fuel elements of a 1000 Mwe power plant is of the order of a few pounds, while the sodium plenum surface area is greater than 300 ft².

The diffusion equation then becomes

$$\frac{\partial}{\partial x} D \frac{\partial c}{\partial x} = \frac{\partial c}{\partial t}.$$

If no adsorption occurs on the lid covering the cover gas space, no mass transfer will occur at this boundary. If the concentration of sodium iodide at the surface layer of the sodium is low (or in other words, yielding negligible vapor pressure) then the iodine concentration in the gas phase at the surface would be essentially zero. This condition is assumed to prevail under the temperature conditions of the experiment. Boundary conditions for this one-dimensional transient problem are therefore: no mass transfer at one surface (as indicated by the lack of recovered iodine from the vapor space container) and zero concentration at the other surface. For a uniform initial concentration distribution in the cover gas, the rate of iodine removal is given by

$$F = \frac{DC_o}{(\pi Dt)^{1/2}} \left\{ 1 + 2 \sum_{n=1}^{\infty} (-1)^n e^{-n^2 l^2 / Dt} \right\}$$

and the average concentration is,

$$C_{avg} = C_o \left\{ 1 - 2(Dt)^{1/2} \left(\pi^{-1/2} + 2 \sum_{n=1}^{\infty} (-1)^n \operatorname{ierfc} \frac{n}{(Dt)^{1/2}} \right) \right\}.$$

where D is the diffusion coefficient, C_o is the initial concentration, t is time, and l is the total depth of the cover gas space. For cover gas depths utilized in sodium cooled reactors and times less than about 1200 sec, the exponential terms in the first equation are negligible. During this initial time, the rate of removal may therefore be predicted by

$$F = C_o \left(\frac{D}{\pi t} \right)^{1/2}.$$

The diffusion coefficient for iodine in nitrogen is $0.06 \text{ cm}^2/\text{sec}$ at 0°C . Based on this experimentally determined value,¹¹ D at 500°F was calculated to be $0.225 \text{ cm}^2/\text{sec}$.

For a cover gas depth of 2 ft, pool surface area of 300 sq ft, and an initial iodine content of 10 lbs dispersed evenly through the cover gas, the rate of

diffusive mass transfer predicted from the previous equation becomes less than the reaction rate after the initial 0.04 sec. Therefore, gas-phase diffusion controls iodine removal from the cover gas after a short initial period.

The value of C_{avg}/C_0 for this example is plotted in Figure 4 as a function of time. From this curve, it may be seen that about 3-1/2 hr are required for 90% of the iodine to diffuse to the sodium surface. If the experiment were reaction-rate controlled, the same amount of iodine could be absorbed by sodium in less than 2 min.

It should be pointed out in this experiment also that nitrogen is not normally employed as a cover gas in the primary system of a nuclear plant. The example presented here assumed nitrogen to make use of existing experimental diffusion coefficients. Diffusion of iodine through helium would be quicker than through nitrogen, and the presence of xenon and krypton in the cover gas would also modify the results. However, it is not anticipated that they would change the results by more than a few percent.

4. Temperature Plateau Heating Experiment (W. P. Kunkel, D. M. Elliott)¹²

This experiment was designed primarily to investigate the equilibrium retention characteristics of a sodium system for iodine released directly to the

Figure 4. Rate of Iodine Diffusion to Sodium Surface

sodium from unmelted fuel. It was intended therefore, that the experiment should indicate in a general way the fate of fission products released under conditions where the fuel was not violently dispersed, nor melted to release other fission product activity.

Figure 5 shows a semischematic diagram of the experiment. It consists of irradiating a capsule containing approximately 800 mgm of fully enriched uranium and 50 gm of sodium for 4 hr in a flux of approximately 10^{12} n/cm²-sec, connecting this capsule to an isothermal manifold system which sweeps it with a heated, inert cover gas, raising the capsule to a series of temperature plateaus, and measuring the activity released at each plateau. Because of plating observed in past experiments, the entire apparatus was dismantled and analyzed for iodine and other fission products after the experiment was completed.

The basic capsule was a stainless steel (304) cylinder approximately 4 in. in length by 1-1/4 in. in diameter. Both the top and bottom were closed by stainless steel plugs welded in place. The bottom plug was fitted with a wire stanchion with two hooks to hold the fuel discs. The top plug had two 1/4-in. stainless tubes about 3 in. long, which were flared and fitted with standard

Figure 5. Schematic Diagram of Plateau Heating Experiment

steel fittings designed to accept plugs during irradiation and to mate with the gas manifold for the plateau heating.

The capsule was thoroughly cleaned and baked out in a vacuum in order to remove any traces of oxide or contamination from the surface. After this, it was transferred into the inert atmosphere chamber where two fully enriched 3/4-in. in diameter by 0.004-in.-thick fuel discs were placed on the hooks provided for them. These two fuel discs of uranium metal weighed 0.46 gm and 0.43 gm. A quantity of sodium was then heated to 350°F in a special flask, and 50.91 gm of this was poured directly into the capsule. Following the loading, the top plug and the sleeve were welded in place and the capsule removed from the inert atmosphere chamber. A small (0.0095 gm) natural uranium calibration foil encapsulated in a quartz tube was also attached to the capsule for flux determination.

The principal components of the gas train were in order of flow: a helium supply cylinder with regulator, a flowmeter, the capsule, the primary manifold, the charcoal traps, and the secondary manifold. The capsule and the two valves immediately adjacent to it were enclosed in a glove box to provide an inert atmosphere secondary containment. Critical lines were stainless steel. The remainder of the gas train including the charcoal traps and the secondary manifold were copper.

The manifold was designed to take the sweep gas coming from the capsule and pass it through a separate charcoal trap during each plateau in the heating cycle. Because it was desired to collect any activity which may have been present in the sweep gas in one place, a charcoal trap at liquid nitrogen temperature was used. The tubing from the capsule to and including the primary manifold valves was wrapped with heating wire and instrumented so that it could be maintained at approximately 1000°F. This design resulted in a rather large temperature gradient from the primary manifold valves to the cold-traps to enhance and control the location of the iodine deposition.

Prior to plateau heating of the capsule, the temperature of the system was raised to 1000°F and maintained at this temperature throughout the experiment by means of various controllers.

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Figure 6. Temperature History and Xenon Release, Plateau Heating Experiment

Inside the glove box, the capsule temperature was raised to the series of temperature plateaus shown in Figure 6. These plateaus ranged from 900 to 1400°F and each was maintained for an hour. At the beginning of each plateau, the manifold valves were opened to allow the sweep gas to pass through one of the charcoal traps. At the end, the valves were used to transfer flow to the next trap and the temperature raised to the next plateau and so on.

After the plateau heating had been done and the capsule allowed to cool, it was valved off, removed from the gas train and transferred into the inert atmosphere chamber where the top was cut off and the sodium removed.

The sodium from the capsule was dissolved in butyl cellosolve and aliquots prepared for counting by gamma spectrometer. Both the complete lines and the traps were cut up into small sections and also counted by gamma spectrometry. Table 6 gives the distribution of the recovered iodine.

Only the seventh line showed measurable activity. When the capsule was reheated and the sodium poured out, it was found that the specific activity of the sodium poured from the capsule was $1.92 \mu\text{c}/\text{gm}$, while the specific activity of that which remained on the capsule walls was $122 \mu\text{c}/\text{gm}$.

The only temperature dependence seen in the case of the iodine was indicated by the fact that iodine was also found on the tubing leading to the seventh or highest temperature trap. However, the xenon exhibited quite a marked temperature dependent release as is also shown in Figure 6. This curve shows the xenon following an "S" shaped release curve as the temperature is increased.

The calculated I^{131} inventory of the irradiated fuel discs was 2.35 mc at the time of the experiment. The iodine recovered in the sodium represents 5.3% of this amount. Since the diffusion coefficients of iodine and xenon are of the same order of magnitude, it would seem reasonable to assume a similar level of xenon release.

TABLE 6
LOCATION OF I^{131} ACTIVITY, PLATEAU HEATING EXPERIMENT

Activity in Sodium		Activity in Gas	
Location	Activity (μc)	Location	Activity (μc)
Sodium Poured from Capsule	92.2	Downstream Tubing	0.107
Sodium Plated on Capsule	27.6	Tubing from Valve 7 to Trap	2.43×10^{-4}
Sodium Plated on Transfer Dish	4.19		0.107
Total	123.99		

These data indicate a release fraction of 8.7×10^{-4} .

5. Condenser Discharge Injection Experiments (W. Kunkel, G. Schlesinger)

As described earlier, the possibility of a flexible, out-of-pile technique which rapidly elevates fuel to the melting and/or vaporization temperatures anticipated in certain fast reactor transients is intriguing. A series of condenser discharge experiments was therefore begun. These included several feasibility experiments to evaluate the adequacy of this technique and culminated in three condenser discharge injection studies to study the response of the May pack-diffusion tube sampling system to sodium vapor and to fission products in sodium vapor environment. A block diagram of the condenser discharge device is shown in Figure 7.

Initially, the device was assembled using a plastic box so that the characteristics of the discharge could be evaluated and observed visually. A current shunt and oscilloscope completed the test portion of the apparatus.

Examination of the waveform of the current and voltage pulse generated in a solid copper bar by application of 4,000 volts from the $185\mu\text{fd}$ condenser showed that the electrical energy was delivered to the target shunt in a period approximating $300\mu\text{sec}$. Samples of uranium metal were then melted and

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Figure 7. Block Diagram - Condenser Discharge Apparatus

vaporized by application of the condenser discharge. Upon fragment examination, it was noted that there appeared to be two rather distinct peaks of particulate diameter, one of which ranged up to one micron, and the second of which was 1 to 10 micron.* It was not possible, however, to assess the temperature imparted to the sample from these data.

Provision also existed in the reaction chamber to vaporize a sample of sodium just preceding the uranium discharge. In this manner, a sodium vapor environment of varying density could be provided to ascertain the effect of sodium vapor on the retention characteristics of fission produced iodine and its complex compounds.

The first experiment of this series vaporized 0.236 gm of sodium in the reaction chamber. Immediately following this event, and while the sodium vapor and droplets were still suspended in the vacuum atmosphere of the reaction chamber, the condenser was discharged into a preirradiated, enriched uranium metal foil weighing 53 mgm.

The preirradiation flux level was 10^{16} nvt, and at the time of the experiment, the foil was estimated to contain approximately 0.5 mc of I^{131} . The reaction chamber was then swept with 130 cc/min flow of helium, and the gas flow was conducted through 80 milliliter of KI solution (1 molar), a 10 gm-6/14 mesh ACC charcoal filter, a silver nitrate impregnated absolute filter, and a backup cartridge of charcoal. It was the intent of this experiment not only to evaluate the adequacy of this method of fission product injection, but also to determine the transport and plating characteristics of fission products injected by this technique with no sodium plenum present. The results of this test are presented in Table 7.

Other feasibility experiments were also conducted. It was observed in most of these experiments that a very small fraction of the available fission product activity was transported into the plenum from the reaction chamber. Most of the activity was recovered from the walls of the reaction chamber and from the transfer tube from the reaction chamber to the plenum.

*These results differ from the particle size distribution found in the in-pile tests (See Section III.B.1).

TABLE 7
CONDENSER DISCHARGE INJECTION TEST USING
SODIUM VAPOR.
ACTIVITY (μc)

Isotope	Original Inventory	Plenum	No. 1 Charcoal Filter	Absolute Filter	No. 2 Charcoal Filter
Ba-La ¹⁴⁰	1250	0.017	0.020	0.003	2.4
I ¹³¹	500	0.014	0.09	N.O.*	1.2
Ru ¹⁰³	2800	0.004	0.11	0.001	0.54
ZrNb ⁹⁵	1550	N.O.	0.09	N.O.	0.45

*Not observed.

The purpose of the next two experiments was to characterize the penetration and diffusion characteristics of fission products resulting from the melting and vaporization of uranium with and without a sodium plenum simulator in the system. For this purpose, an arrangement similar to that shown in Figure 7 was employed, except that a form of the May pack sampling device⁵ was used to examine the retention characteristics. The conditions of the system are shown in Table 8. The two experiments were thus identical except that in the one case, a sodium plenum was employed through which the effluent from the reaction chamber was conducted.

TABLE 8
SYSTEM CONDITIONS FOR COMPARATIVE CONDENSER
DISCHARGE STUDIES

Total sodium content in sodium experiment (gm)	13.9
Temperature of sodium in sodium experiment (°F)	800
Helium flow rate through header (or sodium plenum) (cc/min)	200
Total helium flow (cm ³)	3,000
Fuel mass (gm)	0.070
Helium flow through diffusion tubes (cm ³ /min)	50
Temperature of transfer tube to sodium plenum (°F)	500

TABLE 9
TRANSFER TUBE RADIOACTIVITY DEPOSITION

Isotope	Activity (Normalized to BaLa ¹⁴⁰)
BaLa ¹⁴⁰	1.0
Cs ¹³⁷	2.1
ZrNb ⁹⁵	2.7
Ru ¹⁰³	7.7
I ¹³¹	36

The transfer tube between the reaction chamber and the sodium plenum retained a considerable quantity of fission products, even though heated to 500°F. Of particular interest in this respect was the deposition of iodine activity to a much higher extent than any of the other fission products observed. These results are presented in Table 9 where a compilation of the relative deposition of radioactivity in the transfer tube is shown.

A sketch of the May pack filter components is shown in Figure 8. In Tables 10 and 11 are shown a comparison of the May pack component retention for the sodium plenum and the corresponding experiment in which no sodium plenum was employed.

TABLE 10
DISTRIBUTION OF ISOTOPES IN MAY PACK
COPPER SCREENS*

	BaLa ¹⁴⁰	I ¹³¹	Ru ¹⁰⁶	Cs ¹³⁷	ZrNb ⁹⁵
1. With no sodium	1.0	20	6.7	1.9	2.1
2. After bubbling through sodium plenum	1.0	6.6	6.5	2.4	2.3

*Arbitrarily normalized to BaLa¹⁴⁰ to compare the two experiments

TABLE 11
DISTRIBUTION OF ISOTOPES IN MAY PACK
CARBON FILTER PAPER*

	BaLa ¹⁴⁰	I ¹³¹	Ru ¹⁰⁶	Cs ¹³⁷	ZrNb ⁹⁵
1. With no sodium	1.0	17	6.7	1.7	2.2
2. After bubbling through sodium plenum	1.0	8.4	6.5	2.4	2.3

*Arbitrarily normalized to BaLa¹⁴⁰ to compare the two experiments

Although not quantitatively demonstrated, it would appear from these results that the I¹³¹ retention on both the May pack copper screens as well as the carbon filter paper was noticeably modified by the presence of the sodium plenum.

In addition to the May pack components, diffusion tube data were obtained for the two experiments. An attempt was made to identify the iodine components as

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Figure 8. May Pack Filter Components

a function of the diffusion coefficients. For this purpose, the data were compared with diffusion tube data from ORNL¹³ in which no sodium vapor was used. Results are shown in Table 12.

Aside from the deviations of slope in the second components (which as ORNL points out may not be diffusion-limited), the first diffusion tube coefficients in the AI sodium plenum agree to some extent with those reported by ORNL. The S'1 component has been tentatively identified in the ORNL studies as I_2 , but in the AI case, can probably best be characterized as a particle of 7Å diameter.

The last series of experiments with the condenser discharge apparatus was intended to evaluate the possible effect of sodium vapor on the retention characteristics of the May pack components, since even without a sodium plenum the effect of sodium vapor may modify the May pack retention characteristics. In the first of these experiments, the preirradiated uranium foil was vaporized/melted by the condenser discharge in the reaction chamber but with no sodium in the chamber. The effluent and sweep gas (helium) from the reaction chamber were then transmitted directly to the sampling stream.

In the second experiment, the reaction chamber was flushed with helium before the preirradiated uranium foil was melted, and the gas was passed through a sodium plenum. Just before the condenser was discharged to

TABLE 12
COMPARISON OF AI AND ORNL DIFFUSION TUBE DATA*
(Data obtained from silver-plated diffusion tube results)

Apparent Diffusion Coefficient, cm ² /sec		
ORNL Iodine Characteri- zation Studies:	<u>S'1*</u>	<u>S'2</u>
First diffusion tube	0.113	0.002
Second diffusion tube	0.081	0.011
AI Sodium Plenum Studies:		
First diffusion tube	0.108	0.011
Second diffusion tube	0.108	0.002

*S'1 and S'2 represent the two identified components of the diffusion tube radioactivity deposition.

melt/vaporize the foil, the gas stream was diverted from the plenum and was passed directly from the reaction chamber to the sampling network, as in the first experiment. In this manner, the May pack and diffusion tube network were subjected to the action of the sodium vapor before the fission products were present.

The third experiment conducted the off-gas from the reaction chamber through the sampling tube network immediately after the foil had been vaporized, and for a length of time equivalent to that of the sodium vapor transit in the second experiment. Unfortunately, the resulting activity level in the third

experiment was so low due to retention of fission product activity both in the reaction chamber surfaces and in the sodium plenum, that no discernable activity could be measured on the May pack or diffusion tube network.

Results of the first and second experiments, in which the effect of sodium vapor was isolated from the fission product retention characteristics are shown in Figure 9. It may be inferred from the results of the last series of experiments in which the sodium vapor deposition preceded the fission product sampling, that the sodium residue on the copper screens for example, acted as a rather efficient "scavenger" of iodine activity in the gas stream. It was also noted by sectioning one of the larger globules of sodium which had apparently

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Figure 9. Effect of Sodium Vapor on May Pack Characteristics

condensed on the cooler surface of the copper filter screen, that the iodine activity was not homogeneously distributed throughout the sodium globule, but rather was a surface-adhering phenomenon.

6. Capsule Rupture Experiment (W. P. Kunkel, G. Schlesinger)¹⁴

As a result of the recent interest in the retention characteristics of a bubble of iodine and fission gas being transported through a sodium column, the earlier bubble experiment was repeated with a more realistic insertion of fission product activity.

The capsule rupture experiment is shown in schematic form in Figure 10. The large cylinder at the top contained the sodium to fill the plenum simulator shown in the middle of the figure. The plenum simulator was a 304 stainless steel cylinder, with 7.0 in. ID and 19.0 in. inside height. A circular ring was welded in the center of the bottom plate to hold the irradiation capsule in the proper position. The irradiation capsule was also of stainless steel, with an ID of 1.0 in. and an overall inside height of 2 in. A rupture disc mounted on the top of the capsule was calibrated to rupture at 125 psig for ambient temperature and 74 psig at 500°F.

A 1.5-mil-thick disc of uranium metal (93% U^{235}) weighing 129 mg was supported on three pillars at the base of the capsule. The capsule was evacuated inside a helium atmosphere chamber, purged with helium, and loaded with the fuel disc. There was no sodium inside the capsule in this experiment.

Following the placement of the capsule in position, the lid of the plenum simulation chamber was secured and the plenum simulator evacuated and purged with helium several times. The temperature was then elevated to 400°F and 10 lb of liquid sodium was introduced from the fill chamber.

The temperature of the plenum simulator was next stabilized at 1000°F for 2 hr, the flow of helium over the top of the chamber was established at 200 cc/min, and the firing chamber loaded with argon gas.* It is calculated that the

*Argon was used to simulate fission product xenon.

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Figure 10. Capsule Rupture Experimental Arrangement

gas pressure when applied to the smaller capsule represented 100 psig. The rupture disc was opened by this pressure, as was noted by a sudden decrease on the pressure gauge affixed to the firing chamber.

Following the initial burst of the rupture disc, the firing chamber was again charged with argon, and this was introduced to the small capsule. Temperature of the sodium in the plenum simulator was maintained at 1000°F for

2-1/2 hr. at which time the off-gas sampling stream was diverted from both the May pack and the diffusion tube sampler, and the valve closed preparatory to removing filter components and diffusion tube samples for gamma spectrometric analysis.

At the end of the sampling period, the temperature of the sodium in the plenum simulator was lowered to 400°F, and as the sodium was drained from the bottom of the simulator tank, samples of the sodium were taken for analysis. Ten samples of sodium were obtained varying in weight from 7 to approximately 13 gm of sodium.

The May pack components were disassembled, and each filter disc removed for radioactivity analysis. Similarly, the diffusion tube sampling network was disassembled and the tubes were sectioned into approximately ten samples, for gamma counting.

The activity recovered from the various components of the May pack filter device is summarized in Table 13, along with an estimate of the original activity in the uranium fuel disc. Apparently, much of the activity of the fission products in the uranium fuel disc was either not released from the fuel disc as a result of the fissioning in the reactor preirradiation or was released and subsequently attached to the walls of the capsule and the colder surfaces of the experimental configuration.*

The original inventory of the fuel disc at the time of the rupture is estimated to be 48 μc of I^{131} and 2.6 μc of Xe^{133} . By relating the approximate limit of I^{131} detected in the sodium by chemical separation and gamma spectrometry to the amount in the cover gas, a release fraction of 15% was obtained. This number relates to the highest possible I^{131} activity in the sodium and the fraction of this which was released to the gas space. No conclusive data can be presented relating to the fractional release of iodine activity under the conditions of this experiment. However, a much higher release fraction apparently was observed in this experiment than in the capsule experiments described in the next section.

*The presence of silver resulting from activation of the silver component of the rupture disc, as well as the very small radioactivity inventory anticipated in the large sodium volume, made it difficult to estimate sodium activity. A best estimate resulting from an iodine separation is < 0.36 μc of I^{131} .

TABLE 13
ACTIVITY RECOVERED FROM MAY PACK COMPONENTS OF
CAPSULE RUPTURE EXPERIMENT

Component	I ¹³¹ Activity (μc)	Xe ¹³³ Activity (μc)
First copper screen	4.2×10^{-4}	-
Second copper screen	3.3×10^{-4}	-
Third copper screen	3.3×10^{-4}	-
Millipore filter	3.5×10^{-4}	-
First carbon-loaded paper	1.7×10^{-3}	-
Second carbon-loaded paper	8.5×10^{-5}	-
First charcoal trap	2.3×10^{-2}	9.5×10^{-2}
Second charcoal trap	2.2×10^{-2}	9.5×10^{-2}
Total activity recovered	4.8×10^{-2}	1.9×10^{-1}

B. IN-PILE EXPERIMENTAL STUDIES

1. Low Temperature Sodium Studies in KEWB Pulsed Reactor - (W. Kunkel, C. R. Smith, A. Gibson)¹⁵

Early study showed that the KEWB pulsed reactor³ might be an effective tool for rapidly melting uranium fuel in a sodium environment, and for simulating a violent reactor transient with dispersal of fission products in sodium. Therefore, an initial experiment (designated IP-1) was planned and performed to demonstrate the feasibility of using this method to study fission product retention by sodium.

The IP-1 experiment established the usefulness of the KEWB as a means of studying some of the parameters important to the retention of fission products in reactor coolants. It also demonstrated the necessity for preirradiating the uranium fuel prior to the reactor transient. Therefore, the next experiment, IP-2, was preirradiated in the Shield Test Reactor (STF) prior to insertion in the KEWB. This section of the report describes the experimental procedures and results of Experiments IP-1 and IP-2.

It was determined that for a 93%-enriched sample in a stainless steel capsule partially filled with sodium and located adjacent to the KEWB core, during a 1-ms period burst, that approximately 60 to 70% of the total energy released to the fuel would be released during the time of peak temperature rise (6.5 ms) and that an estimated 5% of this heat would be transferred to the sodium during this period. (See Appendix A).

a. Experiment IP-1. This experiment was performed with a 3/4-in. diameter- $\times$ 0.004-in.-thick fuel disc of 93% enriched uranium. This disc was supported within a stainless steel capsule containing 255 cm³ of sodium at 500°F. The remainder of the capsule was filled with a cover gas of helium at atmospheric pressure.

Also included within the capsule were a normal uranium calibration foil of approximately the same mass as the fuel disc (0.49 gms), three thermocouples, and an immersion heater to melt the sodium. A photograph of the components as they were mounted within the capsule is shown in Figure 11.

The experimental equipment was assembled and placed within the irradiation facility of the KEWB. The secondary container surrounding the experiment,

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Figure 11. IP-1 Experimental Capsule

was purged and an inert cover gas of helium maintained. After stabilizing at the desired temperature, the equipment was subjected to a transient neutron burst from the KEWB. The KEWB instrumentation indicated that the transient was approximately 0.94 msec.

After allowing the activity of the sodium to decay for 88 hr, the capsule was removed and the cover gas was flushed through a series of filters and absorbers designed to trap fission products carried as vapors or particulates in the gas stream. The sodium was remelted and filtered through a glass frit, and the capsule was disassembled. Aliquants of the filtered sodium were examined for fission product activity. The residue remaining in the chamber washed free of sodium and its compounds, was collected, weighed, and identified as uranium metal. An aliquant of this material was measured for fission product activity.

The original U^{235} fuel disc had weighed 0.486 gm. Those remnants which were recovered weighed 0.452 gm. These appeared to be a finely divided black powder. However, examination under a 15x binocular microscope showed that the surfaces of the particles had a metallic luster in a rainbow-like spectrum of colors. Subsequent powder x-ray analyses showed that the powder was predominantly uranium metal with only a trace of oxide present. Approximately 93.0% of the fuel was recovered.

Figure 12 illustrates the appearance of the remnants under low power magnification. In general, the particles are in the shape of teardrops and strings. It appears that the original disc was violently dispersed as a multitude of small molten particles and that these were then rapidly refrozen by the relatively cool ambient sodium.

A sample of the recovered fuel was analyzed metallographically, and an attempt was made to etch the specimen without success. The apparent resistance of these particles to conventional etchants has not been explained. It was speculated, however, that rapid freezing of finely divided molten uranium might not have allowed time for the formation of an ordered grain structure. If this were the case, etching would not be able to bring out the grain boundaries. Average particle diameters were estimated to range from 75 to 125 microns although particles larger than 300 and smaller than 40μ were observed. Many of the particles contain small spherical voids which may have been formed by

Figure 12. IP-1 Fuel Fragments, Low Power Magnification

incipient vaporization of the uranium during the excursion followed by rapid refreezing in the surrounding sodium, or by shrinkage during the freezing process.

Analysis of the cover gas, sodium, fuel remnants, metallic residues recovered from the capsule, and finally washings of the capsule surface yielded the data and activity distribution shown in Table 14.

The results of this experiment in which the foil was not preirradiated, led to some interesting speculations on the effect of fission product precursors in determining the release magnitude of various isotopes from the fuel into the sodium. It was noted, for example, that only Ba^{140} was observed in the sodium. An examination of the precursors of Zr^{95} , Ru^{103} , and I^{131} (See Appendix B) indicates that although the boiling points of some are as low as 670°C , it is only in the case of the Ba^{140} chain that two volatile precursors (I^{140} and Xe^{140}) are included. It may be possible therefore, that only very volatile fission products

TABLE 14
FISSION PRODUCT ACTIVITIES FOLLOWING FUEL MELTING IN SODIUM
(d/m x 10⁻⁶ at time 0 for Experiment IP-1)

Radioisotope	Ba ¹⁴⁰	I ¹³¹	Zr ⁹⁵	Ru ¹⁰³	Xe ¹³³
Cover gas	-	-	-	-	trace
Sodium	14.0	-	-	-	-
Fuel remnants on fritted filter	2.64	1.02	1.47	0.136	-
Metallic residues	35.9	27.4	7.68	4.93	-
Chamber washings	4.45	0.378	0.149	-	-
Totals	46.99	28.8	9.30	5.07	trace

were released during the fuel transient in the sodium environment. This seems to be corroborated by the presence of Xe¹³³ in the cover gas. An important consideration of course, is the fact that although calculations indicated that the localized fuel temperatures during the transient exceeded the melting point of uranium and may have reached temperatures between 2300 and 3900°C, the analog studies (Appendix A) demonstrated that the length of time at these temperatures was probably very short in the sodium coolant (20 ms or less to drop 560°C). Thus, the very high thermal conductivity and heat capacity of the sodium may have been responsible for this effect in trapping some of these lower volatility precursors.

b. Experiment IP-2. The capsule used in the IP-2 experiment was similar in construction to that used in Experiment IP-1 except that a "coin-holder" type device was used to facilitate fuel handling and final closure and an external heater was used to save space inside the capsule.

An in-pile gas sampling train was included in this experiment. Directly downstream from the capsule were two sodium-vapor traps connected in parallel. Each trap contained 2 gm of steel-wool. After passing through the sodium vapor traps, the helium cover gas was conducted through a silver-nitrate impregnated fiber filter and a millipore filter. Two activated charcoal traps containing approximately 1.5 gm of Columbia AC charcoal grade 6/14 were then

Figure 13. Block Diagram of IP-2 Experiment

used. The last retention device was a 12-in. stainless steel tube packed with 13.8 gm of ACS charcoal. A block diagram of the overall system is shown in Figure 13.

The "coin-holder" device was designed to provide a method for transferring the preirradiated fuel disc to the capsule, thus facilitating handling and final closure. The fuel disc was secured to the carrier with a stainless-steel snap ring. Prior to welding the lid in place, the natural uranium calibration foil was mounted in a supporting U-frame.

The gas line up to the sodium vapor traps was wrapped with heater tape and insulated to reduce the tendency of fission product or sodium vapor deposition. The fuel disc for this experiment was also highly enriched uranium metal, and of the same approximate dimensions as the first disc. However, this disc was irradiated for a period of four hours at a flux of 1.2×10^{12} n/cm²-sec, and then allowed to decay for eight days to achieve an optimum I¹³¹ ratio to the other fission products present. Table 15 gives the inventory of the various fission products, just before the KEWB burst.

TABLE 15
ISOTOPIIC INVENTORY* AT TIME OF KEWB BURST
FOR EXPERIMENT IP-2

Isotope	Total Activity in Fuel Disc (mc)
Gross fission products	144.0
Zr, Nb ⁹⁵	2.87
Ru ¹⁰³	1.39
I ¹³¹	5.20
Xe ¹³³	11.4
Ba, La ¹⁴⁰	15.3
Cs ¹³⁷	1.6×10^{-2}

*Preburst inventory.

These results were obtained from the analysis of a natural uranium foil irradiated in the same capsule as the test foil.

Following the 8-day fuel decay period, the capsule was pressure-tested and then loaded with 235 gm of sodium. The fuel disc in the coin-holder carrier was then transferred into the capsule and the shoulder of the holder welded to the capsule top. The entire assembly of capsule and gas lines was then inserted into the KEWB. The conditions during the burst are summarized in Table 16.

The gas sampling components were later examined with a multichannel analyzer in the manner described for the preceding experiment. A hole was then bored in the bottom of the capsule flange; the sodium removal was accomplished within a helium atmosphere in a glove box. As the sodium was heated, it was withdrawn through a coarse fritted glass filter into an evacuation flask. Aliquots of sodium were obtained from the evacuation flask as well as the fritted glass filter. The capsule was washed with a mixture of butyl-methyl alcohol and the alcohol was also filtered. Detailed results are shown in Appendix C for both the gas sampling portion and the remainder of the system. (The remainder of the

TABLE 16
CONDITIONS OF SECOND IN-PILE
EXPERIMENT IP-2

Sodium temperature (°F)	500
He flow through capsule (cc/min)	50
Duration of flow*	1 hr
Sodium vapor trap temperature (°F)	300
Period of burst (msec)	1.01

*The upstream solenoid valve was closed for approximately 5 sec during the actual burst to prevent pulse propagation into the helium supply system.

tubing and external sampling system were also examined. Only stainless steel activation products such as Cr⁵¹, Fe⁵⁹, and Mn⁵⁴ were found).

Approximately 75% of the fuel was recovered. Most of this (33%) was obtained from fuel adhering to the holder ring which had apparently been fused to the ring as the fuel melted during fissioning. The fuel fragments accounted for another 7%, and the remainder was observed in two fairly large pieces. One of these was 0.14 gms in mass, and its isotopic activity was measured. The amount of isotopic activity remaining in the fuel then was estimated by extrapolating to the entire fuel mass. These results are shown in Table 17.

TABLE 17
ISOTOPIC INVENTORY REMAINING IN
FUEL AFTER IRRADIATION AND
MELTING (IP-2)

Isotope	Activity (d/min/gm)
ZrNb ⁹⁵	1.58×10^{10}
Ru ¹⁰³	7.75×10^{10}
I ¹³¹	1.70×10^9
Xe ¹³³	Not detected
Xe ¹³⁵	Not detected
BaLa ¹⁴⁰	4.11×10^{10}

As a result of the compilation of isotopes in the gas phase, sodium phase and other portions of the experimental system, it is possible to summarize the fission product behavior of the various isotopes as they were released into 500°F sodium under the conditions of this particular experiment. These data are presented below.

TABLE 18
SUMMARY OF FISSION PRODUCT BEHAVIOR IN 500°F SODIUM
(IP-2 Experiment)

Isotope	Activity Recovered in Sodium (μc)	Gas Activity Recovered in Sampling Line (μc)	Fraction of Sodium Activity (10^{-2}) Released to Gas
I^{131}	52.4	10^{-3}	0.002
Xe^{133}	Not detected	1.29×10^{-3}	11.3*
BaLa^{140}	2.07×10^{-3}	1.8×10^{-3}	0.0001
ZrNb^{95}	58.5	Not detected	0

* Xe^{133} activity in gas based on amount present in fuel

2. High Temperature Sodium Studies in KEWB (W. Kunkel, D. Elliott, A. Gibson)

Two in-pile experiments (designated IP-3, IP-4, etc) which probably have more application toward fast reactor accidents in terms of the simulation conditions, were next conducted in 1000°F sodium. In these in-pile experiments, preirradiated uranium metal fuel discs were melted (and partially vaporized) by neutron burst irradiation in the KEWB reactor. During the time of the fuel melting and the subsequent coolant and resolidification of the sodium, the cover gas was conducted through a series of traps to evaluate the activity release from the sodium. The techniques for maintaining the integrity of the system in the event of a capsule leak, the general procedures, and the recovery technology have been discussed above. The two 1000°F studies were similar to these studies with respect to the technology employed.

A compilation of the radioisotope release fractions from the two 1000°F sodium retention experiments IP-3 and IP-4 is here shown.

TABLE 19
RADIOISOTOPE RELEASE FRACTIONS IN 1000°F SODIUM
IN-PILE EXPERIMENTS

Isotope	Fraction of Isotope Released to Sodium (%)		Fraction of Isotope in Sodium Released to Cover Gas (%)	
	<u>IP-3</u>	<u>IP-4</u>	<u>IP-3</u>	<u>IP-4</u>
ZrNb ⁹⁵	10-24	10.3	Not observed	Not observed
Ru ¹⁰³	0.5 - 0.66	1.9	Not observed	Not observed
I ¹³¹	21-72	41	0.02 - 0.004	0.004
Cs ¹³⁷	Not measured	Not measured	Not measured	0.02
BaLa ¹⁴⁰	2.4 - 5.4	2.1	0.11 - 0.05	0.02

No Cs¹³⁷ was observed in sodium. The fractional release reported in the cover gas is based on pre-burst inventory in total fuel. The activity in the gas may have resulted from the release of short-lived Xe¹³⁷ through the sodium and the subsequent decay to Cs¹³⁷ in the gas space.*

3. High Burn-up UC Fuel Irradiation Studies (W. Kunkel, G. Schlesinger)

Acquisition of the cover gas, sodium bond, and cladding from several high burn-up UC fuel irradiation capsules¹⁶ permitted the observation of fission product distribution and compartmentation under various conditions.

The first capsule was removed from the GETR after approximately 20,000 Mwd/t. A sketch of the capsule is shown in Figure 14. Basically, it contained 71.6 gm of U²³⁵, 644.0 gm of U²³⁸, and 34.1 gm of C. This corresponds to a

*Xe¹³⁷ inventory from the KEWB burst would be approximately 4.4×10^{11} atoms. If all of this were released from the fuel and decayed to Cs¹³⁷ in the gas space, the Cs¹³⁷ activity would be $4 \times 10^{-3} \mu\text{c}$. Actually, $4.5 \times 10^{-4} \mu\text{c}$ of Cs¹³⁷ were measured in gas space.

Figure 14. Sketch of UC Irradiation Capsule

U^{235} enrichment of approximately 11% and a nominal carbon content of 4.6%. Physically, the fuel rod was 18 in. long by 0.5 in. in diameter. Surrounding the fuel was a sodium bond comprising 14.6 gm of sodium, and a 10-mil steel cladding of Type 304 stainless steel. The fuel rod was NaK bonded to a baffle tube and nickel shell about 30 in. long.

Following its removal from the reactor, the capsule activity was allowed to decay for only a short time. It was then shipped to the hot cell while the sodium was still liquid from afterglow heating. The capsule was removed from its shielded shipping container, and the sheath was punctured to obtain gas samples. (The gas sampling system consisted of two charcoal filters in series through which the gas sample was drawn). The filters were subsequently removed and counted on a well-crystal gamma spectrometer with a multichannel analyzer for isotopic content and identification. The second filter contained less than 1% of the activity of the first so only the activity of the first filter is included in the presentation of data.

In obtaining sodium samples, the cladding was cut and the rod was placed in a stainless steel basket and immersed in an oil bath to prevent any sodium reaction. Two sodium samples were obtained from the bond content below the gas space. These were later placed in an inert atmosphere glove box and the outer layer of brown film flecked with black particulate material was physically cut off with a small scalpel. Two "clean" sodium samples were obtained from sodium sample 1, and one was cut from sodium sample 3.

Samples of the cladding were obtained similarly from the area above the sodium bond, towards the middle of the fuel rod, and towards the bottom of the fuel rod. Analysis of the samples was complicated by the high activity levels of the stainless steel constituents which had been activated during the irradiation period. As a consequence, the possible use of autoradiography was eliminated because of the generalized radiation levels from the specimens. It was decided therefore to use etching to remove activity from the cladding and allow determination and identification of isotopic content.

A solution of six parts of water and one part of nitric acid was provided and 80 ml of the solution was poured into each of six 100 ml beakers. Following removal from the shielded shipping cask, the cladding samples were first rinsed in butyl alcohol without agitation for approximately fifteen minutes. After a brief rinse in distilled water, they were placed successively in the etching solutions provided. The initial rinses were of the order of 5 minutes. The final rinse was approximately 18 hours. All rinses were without agitation.

A gamma spectrum of the charcoal trap containing the cover gas activity was obtained approximately 43 days after the capsule was removed from the reactor. As a result of the short decay period, I^{131} was observed in the cover gas.

The data from the rod cover gas sample were extrapolated back to reactor out-time, and were normalized to the longest lived isotope present (Kr^{85}). These data were compared with the estimated inventory of fission products anticipated from fuel under these burnup conditions. The calculated activity ratios in the fuel rod inventory would indicate that there should be approximately 354 times as much Xe^{133} in the cover gas as there would be Kr^{85} . The measurements showed only 11 times as much Xe^{133} as Kr^{85} in the cover gas. There was, therefore, an indicated depletion of Xe^{133} by a factor of 32 relative to the krypton. If it is assumed that one may associate this depletion with half-life, it represents a decay while in transport equivalent to $2^5 = 32$, or five half-lives. Similarly, comparing the Xe^{131m} release, 0.49 half-lives are indicated.

*It should be pointed out that these results may have no direct relationship to the amount actually present in the cover gas, since some of the gas activity was apparently lost due to a leak in a thermocouple weld.

†The choice of Kr^{85} as a normalization point was based on its being the longest lived isotope present.

TABLE 20
RELATIVE ISOTOPIC ACTIVITY INVENTORY OF A
HIGH BURNUP UC CAPSULE

Isotope	Cover Gas		Fuel Rod
	Measured (μc)	Relative*	
Kr ⁸⁵	1.0×10^{-5}	1.0	1.0
Xe ¹³¹	1.2×10^{-5}	1.2	1.7
I ¹³¹	3.6×10^{-7}	0.036	15.3
Xe ¹³³	1.1×10^{-4}	11.0	354.0

*Normalized to Kr⁸⁵

The half-life of Xe¹³³ is approximately 5.3 days, while that of Xe^{131m} is 12 days. Therefore, an apparent delay of xenon release compared to krypton release could account for these anomalies if it is assumed that this delay is within the range of $5 \times 5.3 = 27$ days to $0.49 \times 12 = 6$ days.

The data from the sodium samples were also extrapolated back to reactor "out-time" and are presented here.

TABLE 21
ACTIVITY LEVELS OF SODIUM BOND SAMPLES FROM
UC FUEL ROD TEST
(Extrapolated to Reactor Shutdown)
Activity (μc)

Mass (gm)	Na ²²	Cr ⁵¹	Co ⁶⁰	ZrNb ⁹⁵	I ¹³¹ †	Cs ¹³⁷	BaLa ¹⁴⁰	Ru ¹⁰³
Sodium Sample 14 0.29	*	0.96	0.033	3.22	0.015	*	5.3	7.4
Sodium Sample 18 0.017	0.03	*	*	0.234	*	0.011	0.30	0.22
Sodium Sample 38 0.36	0.04	*	*	5.04	0.0002	0.066	6.0	8.05

*Not observed.

†Because of the chemical separation, I¹³¹ results are presented in terms of $\mu\text{c/gm}$.

Figure 15. Lower Sodium
Bond, UC Capsule
Cladding Etch

Figure 16. Middle Sodium
Bond, UC Capsule
Cladding Etch

Figure 17. Above Sodium
Bond, UC Capsule
Cladding Etch

The sodium bond activity indicates more dispersal of data than the other samples observed. Perhaps part of this problem stems from the difficulty of assuring that a small section of sodium does not contain a fragmented bit of fuel or was contaminated in the hot cell. The general agreement of the sodium bond samples is indicated in Table 22, where normalization has been used to evaluate internal consistency of the data. The choice of the normalization point is again arbitrary and in this case was selected to be BaLa¹⁴⁰ merely because it was present in sufficient amount in each sample. For comparison, these data are presented in Table 22 along with a calculated estimate of the fuel rod inventory of the specific isotopes which has also been normalized to Ba¹⁴⁰.

The results of the cladding sample examination are presented in Figures 15, 16, and 17. The units on these samples are based on the total estimated activity (exclusive of cladding activation) on the cladding surface. As may be noted, a high percentage of the total radioactivity removed was apparently very lightly affixed to the cladding surface. It may also be noted that no BaLa¹⁴⁰ was observed on the middle cladding sample, and CePr¹⁴⁴ was noted only on the sample obtained from the gas space.

4. SRE Deposition and Plating Studies (W. Kunkel, G. Schlesinger)¹⁷

From much of the experimental work described so far, it has been observed that the deposition and plating of fission product isotopes constitutes an important depletion mechanism for the reduction of fission product radioactivity

TABLE 22
INTERNAL CONSISTENCY OF SODIUM BOND SAMPLES

Isotope	Calculated Activity Relative to BaLa ¹⁴⁰	Measured Activity of Sample Isotope Relative to BaLa ¹⁴⁰			
		<u>IA</u>	<u>IB</u>	<u>3A</u>	<u>Average</u>
ZrNb ⁹⁵	1.0	0.76	0.76	0.83	0.73
Ru ¹⁰³	0.4	1.1	0.76	1.3	1.1
BaLa ¹⁴⁰	1.0	1.0	1.0	1.0	1.0
Cs ¹³⁷	0.9	0.03	0.04	0.01	0.03

levels which might exist in a reactor coolant circuit following introduction from fuel element failure or melting.

Therefore, when an opportunity arose during the project activities to acquire information on a typical reactor system into which fission products had been introduced under realistic fuel failure conditions, it was decided to acquire some data related to the fate and ultimate disposition of fission products in a circulating sodium system. It must be realized that only a limited number of specimens were examined, and of course, no opportunity existed for a rigorous control of sodium temperature, flow rate, and other parametric variations which might have been of interest in ascertaining the mechanisms involved. However, the data are related to the fate of fission products released into sodium, and certainly represent a typical reactor system with the various operating devices which are peculiar to sodium systems. They are therefore reported here because of their probable importance in scoping the results of such fission product introduction and in evaluating problem areas for further detailed chemical and engineering experiments and analyses.

The Sodium Reactor Experiment (SRE) is a thermal reactor utilizing sodium as a coolant with typical operating temperatures of about 960°F design outlet temperature. The system pressure is approximately 15 psi at rated flow of 1080 gpm. The design inlet temperature is approximately 500°F. The main primary loop contains 6-in. stainless steel piping through which the sodium is pumped at a velocity of about 13 ft/sec; reactor power is up to 20 Mwt.

The reactor was critical with sodium, initially in April 1957, after which it logged a considerable amount of operating time. In 1958, the SRE operated at power levels of up to 21 Mwt and logged a total of 218 Mwd. In FY 1959, the SRE accumulated 2191 Mwd of operation at power levels of up to 20 Mwt.

In July of 1959, a routine reactor run was terminated because of undesirably high temperature differences between fuel elements. Subsequent examination of the fuel elements revealed substantial damage to approximately 11 of the 43 elements in the core. The conclusions reached as a result of an extensive investigation are documented elsewhere,¹⁸ but indicate that the fuel cladding probably failed as a result of low-melting iron-uranium compound formation which was produced because of partial blockage of the fuel element passages

and consequent overheating of the fuel elements. It was further decided that the fuel coolant channel blockage was initiated by the accumulation of the decomposition products of tetralin which had leaked into the primary system at an earlier date.

The total mass of fission products introduced into the system was estimated to be approximately 0.9 grams. This would correspond to a concentration of fission products in the sodium of about 0.04 ppm.

The SRE was restored to power following the incident and has been operating at powers of up to 20 Mwt for several years. The reactor was last shut down in March 1964 and the primary loop disassembled for modification and high power operation.

Six sodium samples were obtained from the locations indicated in Table 23, and the low and high temperature spool pieces were also made available for study of fission product plating and deposition.

Sodium coring tools were fabricated of stainless steel sleeves with an internal cylinder to eject the sample obtained, and various pipes examined for sodium content. Typically, the samples were approximately 1-3 grams in

TABLE 23
SODIUM ACTIVITY FROM VARIOUS LOCATIONS IN THE
SRE SYSTEM,
ISOTOPIC ACTIVITY (μc)

Location	Mass (gm)	Na ²²	Mn ⁵⁴	ZrNb ⁹⁵	Cs ¹³⁷	Total Activity
L-037	*	1.23×10^{-2}	N.O.	N.O.	4.76	4.78
L-632	4.6	1.35×10^{-3}	N.O.	N.O.	N.O.	1.35×10^{-3}
V-104	*	3.33×10^{-2}	1.41×10^{-1}	6.26	1.0×10^{-1}	6.53
S-4 eductor	1.0	3.83×10^{-2}	N.O.	N.O.	N.O.	3.83×10^{-2}
Pump bowl	0.92	4.14×10^{-2}	N.O.	N.O.	5.0×10^{-3}	4.64×10^{-2}
L-102	1.07	4.95×10^{-2}	N.O.	N.O.	4.29×10^{-3}	5.38×10^{-2}
L-103	1.19	4.55×10^{-2}	N.O.	N.O.	8.52×10^{-2}	1.30×10^{-1}
L-103 spool	36.9	1.6	14.5	1.25	17.7	35.1

*Data were not obtained due to difficulty of handling sample.

weight. These sodium samples were inserted into a dry box and the outer oxide crust was cut away to provide a relatively uncontaminated sample for radiochemical analysis.

The sodium sample activity from various locations within the SRE system is tabulated in Table 23, together with the activity observed for each isotope at the location.

The spools L-103 and L-104 are located in the cold and hot legs of the primary loop adjacent to the reactor.

A residual quantity of sodium in the bottom of the spool pieces appeared to be approximately 2 inches wide and perhaps 1/8 inch deep.

After the sodium had been removed as cleanly as possible, the piping section was rinsed, dried, and autoradiographed.

Although it was obvious from the autoradiograph that a non-uniform deposition of radioactivity existed, a small ring varying in thickness from 1/2 to about 3/4 inch was removed from the end of the spool sample and cut into four quadrants. Each of the quadrant samples was then photographed, autoradiographed, and treated with assorted etching and rinsing solutions to obtain data on the radioactivity existing on its surface as plated or deposited material.

The relative radioactivity measurements of the L-103 spool are shown in Table 24. These results were obtained by measuring both the closed window and the open window radiation levels from the open end of the spool, and represent primarily radioactivity located on the inner aspect of the piping surface.

TABLE 24
RELATIVE RADIATION LEVELS FROM
L-103 PIPE SPOOL*

Condition	Radiation Level
Open window before rinsing	800 mrem/hr/ft
Open window after rinsing	600 mrem/hr/ft
Closed window before rinsing	14 mr/hr/ft
Closed window after rinsing	12 mr/hr/ft

*All readings were taken with a Juno Survey Meter.

The first quadrant was selected for treatment with nitric acid etching. The nitric acid solution was composed of six parts of H_2O by volume and one part of HNO_3 by volume. Six equal volume beakers were used and the sample was successively placed in each etch for a specified length of time with water rinses between each etch. Solutions were sampled, and analyzed for iron content as well. The results of the nitric acid etch are shown in Figure 18. The amount of iron removed is here shown.

TABLE 25
IRON REMOVED BY NITRIC ACID ETCHING

Time of Etching	Total (Integrated) Iron Removed (mg)
30 min	50
85 min	104
220 min	104

A second quadrant was treated by successive etching in a solution containing one part glacial acetic acid and one part distilled water by volume. During the etching procedure, the open window radiation level was decreased by a factor of 38 over that measured before the etching was commenced. The time dependence and identification of the isotopic activity removed with the acetic acid etch is shown in Figure 19.

The third quadrant was treated with hydrochloric acid solution in which six parts of distilled water were used with one part of hydrochloric acid. Similar techniques were employed in the etching analysis, but the HCl etch was more carefully observed by radioactivity readings and autoradiograph at each step than the other samples. Figure 20 presents the radioactivity removal efficiency of the hydrochloric acid etch. Also included in this graph is the relative decrease in dose rate as a function of etching time.

One of the more interesting phenomena observed was the apparent correlation of the radioactivity deposition with the black stain on the quadrant pieces. A typical example is shown in Figure 21, where a visual photograph and an

Figure 18. Relative Isotope Removal Efficiency With Nitric Acid Rinsing

Figure 19. Relative Isotope Removal Efficiency With Acetic Acid Rinsing

Figure 20. Relative Isotope Removal Efficiency With Hydrochloric Acid Rinsing

Figure 21. Visual and Autoradiographic Comparison

autoradiograph are side by side to the same scale. As may be noted, there is almost direct one-to-one correspondence of the radioactivity deposition with the black stain. (The autoradiograph is a negative and the dark areas correspond in this case to maximum radioactive exposure.)

A sample of the black shiny material staining the quadrant piping surface was submitted for fluorescent and x-ray diffraction analysis. The results indicated the presence of Co, Fe, and other stainless steel constituents, but not in sufficient amount to account for the mass of the specimen. No evidence of heavy metals was obtained, but an indication of an amorphous material was observed which could be a carbon compound.

Some data have also been obtained by A. I. Hansen of AI¹⁹ concerning the deposition of fission product and other activity in the cold trap portion of the SRE. The cold trap used in the SRE was of the gas cooled type in which the process coolant enters an internal economizer section and then enters a trapping section. In the trapping section, four rolls of Type 304 stainless steel mesh fill the section in order to provide a settling area for the oxide.

Considerable quantities of carbon were found in the cold trap, which is in general conformity with the expected behavior of carbonaceous material. The cold trap proved also to be efficient in trapping fission product activity. These results, together with the amount of carbon and other impurities noted in the cold trap, are here shown.

TABLE 26
IMPURITY LEVELS IN COLD TRAP AS COMPARED TO
Na COOLANT IN THE SRE

Impurity	Level in Cold-Trap	Level in Na Coolant	Collection Ratio $A_{\text{trap}}/A_{\text{coolant}}$
Carbon	144 - 1550 ppm	18 - 60 ppm	77
Cs ¹³⁷ *	$4.0 \times 10^2 \mu\text{c/g}$	$1.5 \times 10^{-2} \mu\text{c/g}$	2.7×10^4
Sb ¹²⁵ *	$4.3 \mu\text{c/g}$	$0.63 \times 10^{-2} \mu\text{c/g}$	6.8×10^2
Fe†	200 - 500 ppm	50 ppm	10
Si†	200 - 500 ppm	50 ppm	10
Mn†	50 - 500 ppm	5 ppm	100
Pb†	5 - 500 ppm	10 ppm	50
Cr†	5 - 500 ppm	5 ppm	100
Ni†	10 - 300 ppm	5 ppm	60

*Measured by gamma spectroscopy

†Measured by emission spectroscopy

The high collection ratio of the Cs¹³⁷ is particularly interesting and indicates a possible cesium trapping mechanism other than a trapping of carbon particles carrying cesium.

The correlation between radioactive deposition and the dark stains on the pipe spool surface as well as the differences in the removal efficiency of various etching solutions suggested the evaluation of nonacid mechanical agitation to evaluate the tenacity of the deposited radioactivity. The fourth quadrant was therefore autoradiographed and was also photographed. Following this procedure, it was inserted into a solution of detergent and was scrubbed for

four minutes after which it remained in the solution for forty minutes. The sample was removed and again autoradiographed and photographed for comparison. The effect of the detergent rinsing and scrubbing was to reduce the open window dose rate from the sample by a factor of 3.8. No appreciable reduction in the gamma dose rate was observed. The material removed from the sample with the detergent treatment was composed of Cs^{137} , Mn^{54} , and ZrNb^{95} .

The second spool L-101 comprised the main outlet line from the SRE core for the sodium coolant. It was also removed in 1964 when the reactor was overhauled and rebuilt, and an approximate 1-ft section of the pipe was examined for fission product deposition. The section was the first foot of the line, so that one end of it butted up against the core. At the time of its removal, the pipe was so marked that this end could be identified.

It was noted that a thin line of sodium remained adhering to the length of this pipe also. The surface was stained at several locations, but this did not seem to be correlated with the pipe orientation. Radioautographs were then made of each end of the spool.

The estimated 0.1-gram sodium adhering to the inner surface was carefully washed into a small vial with isopropyl alcohol, and the vial was set aside for later analysis. As with the cold-leg sample, the cross-sectional profile of the pipe was arbitrarily divided into quadrants and markings were stamped into the outer surface of the pipe to identify them. A ring of metal 1/8 inch wide was taken from each end and each ring was then divided into quadrants, and examined.

Samples from the bottom and top of each end of the pipe were selected and were etched in successive baths of 2.5 N nitric acid. Although aliquots of each bath were drawn and counted on a 400-channel pulse height analyzer counting system, it was found that virtually all activity was removed in the first two baths. The results are herewith presented.

TABLE 27
SRE HOT-LEG SPOOL RADIOLOGICAL
EXAMINATION RESULTS (L-101)
(Microcuries per 100 g of Metal)

	Cs ¹³⁷	Ru ¹⁰⁶	Mn ⁵⁴	Co ⁶⁰
<u>"Close End"*</u>				
Bottom				
1st rinse	0.36	--	--	0.72
2nd rinse	0.0054	0.0072	--	--
Top				
1st rinse	0.81	0.41	0.52	0.25
2nd rinse	0.022	0.022	0.015	0.030
<u>"Far End"†</u>				
Bottom				
1st rinse	0.030	0.21	0.46*	0.16*
2nd rinse	0.0076	--	--	--
Top				
1st rinse	0.40	0.15	0.45*	0.17*
2nd rinse	0.13	--	--	--
Sodium Sample §	140	--	--	13

*Adjacent to reactor vessel

†Downstream side of spool

§Total content

III. ANALYTICAL STUDIES

Most of the experimental studies reported in the project have been paralleled by analytical tasks to interpret and apply the results which have been obtained. In addition however, there have been more generalized studies performed to either scope the area which was to be experimentally evaluated, to attempt an explanation of the possible mechanisms involved in the fission product behavior, or to provide some application of these results.

A. A Thermodynamic Analysis of Fission Product Behavior in Sodium (S. Stachura)

In studying fission product release under the abnormal conditions of a nuclear excursion or loss of coolant flow, it is necessary to examine the behavior of both stable and radioactive species under the conditions of steady state and transient heating. That is, the steady state behavior largely determines the course of events during an accident.

There are many factors which could play an important role in the release of fission products and there is presently little basis on which to ascribe relative importance. Among those which should be considered are the following.

- 1) Description of fission product levels and concentration profiles during steady state operation including disposition of volatile fission products and possible solution or compound formation with fuel, bond, clad or other reactor materials - this description would require some knowledge of the importance of recoil, and the effects of temperature and structural condition of fuel on diffusivities.
- 2) Behavior of fission products in sodium at temperatures up to and exceeding the normal boiling point.
- 3) Consequences of sodium boiling and cladding failure.
- 4) Volatilities and possible decomposition of fission product compounds.
- 5) Possible recondensation of fission products and fission product compounds due to cooling (by bond, coolant of upper plenum sodium or adsorption for reaction) on solid materials.
- 6) Biological importance of released fission products.

The above factors are extremely general and the details of evaluating some of them in a quantitative fashion (for example, No. 1) could be prohibitive. Nevertheless, the assumptions that are made in this type of analysis could be extremely critical and no factor should be carelessly discarded.

The considerations which follow are based on a sodium cooled thermal reactor which has metallic fuel, sodium bonded to stainless steel cladding in unvented fuel elements. Other reactor materials would include graphite canned in zirconium or zircalloy. Where burnup conditions are important, a flux of 10^{13} neutrons/cm²-sec for 10^7 sec (slightly more than 100 days) has been assumed.

The most difficult problem in the study of fission product chemistry is the large number of elements present. Little is known for example, regarding the effect of one fission product on the solubility (in sodium) of another. In addition, the thermodynamic calculations must be approximations to the extent of dealing with one or two fission products at a time and neglecting the effects of others. This ties in with the assumption that is made that the activities are equal to the molar concentrations. These limitations are not likely to be lifted at the present because they would require very large numbers of careful experiments.

Another problem of importance is the matter of small concentrations. Isotopes which have concentrations less than 10^{-6} atoms/atom U²³⁵ have been neglected. This has resulted in the elimination of seven minor elements (As, Gd, Tb, Dy, Zr, Ga & Ge).

The details of the analysis are presented here to indicate the reasoning and the assumptions which were employed. The concentration levels for all stable and radioactive fission products were tabulated for conditions of flux equal to 10^{13} neutrons/cm²-sec and time of operation of 10^7 sec (no cooling-off period). Data were taken from the graphical presentation of Blomke & Todd.²⁰

Properties of the fission products were compiled primarily to indicate those likely to escape from the fuel because of their volatilities. Biological significance is also indicated so that fission products important in this regard are given careful attention.

Of the 37 fission products, those which are regarded as electronegative elements are iodine, bromine, selenium and tellurium. Of these, iodine and bromine are of greater important because of their volatilities. The escape of Se and Te (bp of 958 and 1260°K) may be further reduced by the formation of compounds in the uranium. Because the iodine concentration is about two orders of magnitude greater than bromine, the behavior of iodine is of greatest importance with respect to compound formation for the system under consideration.

Free energies of formation of all pertinent iodides at temperatures up to 1500°K were compiled, together with the melting points and boiling points of the iodides. This information then provides a basis for calculating the equilibrium behavior of the system.

In order to simplify the calculation of the chemical equilibria, the analyses were confined to a consideration of reactions of the form

to determine relative stabilities and approximate values for concentration levels. Activities were assumed to be equal to the molar concentrations and in most cases, total release of the fission product from the fuel was assumed. The calculations could readily be modified to account for release fractions. Modifications could also be made if activity data were available, however, the complexity of the system does not lend itself to straightforward measurement of the relevant thermodynamic functions.

Once the relative stabilities and approximate equilibria are established for the fission products and fission product compounds, some assessment can be made regarding the transport of fission products in a typical fuel rod. Volatile inert elements and volatile stable compounds favor release, while stable compounds having high boiling points improve retention. Some considerations along these lines were treated.

Tables 28 and 29 constitute a fairly complete representation of the basic information relevant to fission product release for the system under consideration. The references from which these data have been obtained appear in a list following the cited literature.

TABLE 28
BASIC INFORMATION, FISSION PRODUCT INVENTORY

Element	M.P. °K	B.P. °K	(Atoms/Atom U ²³⁵) Concentration			Biological Importance		Fission Product Iodide		
			Stable	Total	Percent of F.P.	Total	Modified	-ΔF _{100°K} (kcal/mol I ₂)	M.P. °K	B.P. °K
Xe	161	165	0.011	0.015	15	-	-	-	-	-
Kr	116	120	0.002	0.0023	-	-	-	-	-	-
I	387	456	6 x 10 ⁻⁴	8 x 10 ⁻⁴	1	3	1	-	-	-
Br	266	331	1 x 10 ⁻⁵	1 x 10 ⁻⁵	-	-	-	IBr	309	389
Cs	302	955	5 x 10 ⁻³	8.9 x 10 ⁻³	10	-	-	136	894	1553
Rb	312	967	2.2 x 10 ⁻³	2.2 x 10 ⁻³	2	-	-	135	911	1577
Cd	594	1040	-	3.3 x 10 ⁻⁵	1	-	-	32	660	1069
Se	490	958	-	7.5 x 10 ⁻⁵	-	-	-	Se ₂ I ₂	Unstable	-
Te	723	1260	6 x 10 ⁻⁵	3 x 10 ⁻⁴	-	-	-	-	532	Unstable
Sb	903	1908	1.6 x 10 ⁻⁵	3.7 x 10 ⁻⁵	-	-	-	-	444	700
Sn	505	2896	5.6 x 10 ⁻⁵	5.7 x 10 ⁻⁵	-	-	-	-	593 419	987 621
In	429	2343	-	6 x 10 ⁻⁶	-	-	-	-	624 7483 403	988 11000 773
Sr	1043	1648	0.002	0.007	10	1	2	117	788	1850
Ba	983	1895	0.003	0.0036	-	4	4	121	984	(2000)
Zr	2125	4688	0.017	0.020	18	5	5	71	772	784
Mo	2890	4924	0.010	0.011	10	-	-	0	(690)	(695)
Ru	2700	4392	0.0035	0.0045	2	8	-	+	d400	(1150)
Rh	2239	4000	-	0.001	-	-	-	+	d700 d800	(1700) (1100)
Pd	1823	3020	-	6.5 x 10 ⁻³	-	-	-	-	(700)	(1100)
Ag	1234	2437	2 x 10 ⁻⁵	2.1 x 10 ⁻⁵	-	-	-	22	830	1779
Nb	2740	5007	-	8 x 10 ⁻⁴	-	-	6	-	(600)	(620)
Y	1803	3577	0.0012	0.003	5	-	-	72	1273	1580
Tc	(2400)	(4900)	-	0.0035	-	-	-	-	-	-
La	1193	3634	0.003	0.0031	28	7	-	88	1034	1675
Ce	1077	3742	0.003	0.008	-	2	3	86	1025	(1670)
Pr	1200	3295	0.002	0.008	-	-	-	85	1006	(1650)
Nd	1297	3335	-	0.009	-	-	-	84	1048	1640
Pm	(1300)	(3000)	-	0.0013	-	-	-	83	1070	(1640)
Sm	1345	2076	-	0.0011	-	-	-	79	1093	d
Eu	1099	(1764)	-	1.6 x 10 ⁻⁴	-	-	-	68	1150	d
Other elements of interest										
Na	371	1163	-	0.1 to 1.0	-	-	-	112	935	1577
U	1405	4473	-	20 to 30 solid	-	-	-	60 50	1030 779	1700 1032
Fe	-	-	-	-	-	-	-	15	860	(1100)
Cr	-	(2938)	-	-	-	-	-	26	1066	(1100)
Ni	(1728)	-	-	-	-	-	-	3	1070	(1020)

TABLE 29

BASIC INFORMATION, FISSION PRODUCT PHYSICAL AND CHEMICAL CHARACTERISTICS

Reaction No.	Iodine 1 (I ₂)	Uranium 2	Sodium 3	Cesium 4	Rubidium 5	Strontium 6	Barium	Zirconium 7	Bromine 8 (Br ₂)	Chromium 9
-(F-H)/T Element										
298	62.28	12.03	12.24	20.16	18.1	12.5	16.0	9.32	58.65	5.68
500	63.29									
1000	66.82									
1500	69.58									
Component	I	UI ₂	NaI	CsI	RbI	SrI ₂	BaI ₂	ZrI ₄	NaBr	CrI ₂
-ΔH ₂₉₈	36	137	76.7	91.3	88.4	151	159.5	160	90.3	58
-ΔH ₂₉₈ /T ₂₉₈		460	257	306	296	506	535	536	302	194
500	-	274	153.4	183	177	302	319	320	181	116
1000	-	137	76.7	91.3	89.4	151	159.5	160	90.3	58
1500	-	91	51.1	61	59	100	106	106	60	38.6
-(F-H)/T										
298	43.18	55	23.55	28	28.1	38	38	60	20.1	34
500	43.75	58	25	29	(29.7)	40	40	64	246	36
1000	45.72	69	(30.8)	35	(35.11)	50	50	(~70)	26.7	44
1500	47.24	78	(37.0)	43	(41.22)	58	58	-	32.9	53
-RlgK										
298	-97	409								
500	-49	224								
1000	-11.5	90	81.6	11	-0.7	4	-	-142	7.7	-87
1500	-0.43	46								

The following reactions were considered.

- 1) Dissociation of iodine,

- 2) Formation of uranium iodides,

- 3) Reaction of sodium with uranium iodide,

- 4) Reduction of sodium iodide by cesium,

- 5) Reduction of cesium iodide by rubidium,

- 6) Reduction of sodium iodide by strontium,

- 7) Reduction of sodium iodide by zirconium,

- 8) Reduction of sodium iodide by bromine,

The standard states for iodine and bromine at 298°K were taken as the perfect diatomic gas at 1 atm; the conventional standard state is employed for all other elements and iodides. The typical calculational technique is shown here for the reaction.

$$F = -RT \ln \left(\frac{a_{\text{CsI}}}{a_{\text{NaI}}} \right) \frac{(a_{\text{Na}})}{(a_{\text{Cs}})}$$

$$R = 1.99 \frac{\text{cal}}{\text{g mol} \cdot ^\circ\text{K}} F \text{ in units of cal/mol}$$

$$T = 1000^\circ\text{K}$$

In the sodium bond, the sodium activity is unity and the Cs, NaI and CsI activities are taken to be their molar concentrations. This is rationalized by the known levels of mutual solibilities of the metals and their salts which exceed the levels under consideration.

The mole fraction of I is on the order of 10^{-3} ; the mole fraction of CsI $< 10^{-3}$, and $.008 < a_{\text{Cs}} < .009$. Rather than solving the quadratic, let $a_{\text{Cs}} = 0.008$, then

$$\frac{a_{\text{CsI}}}{a_{\text{NaI}}} = 0.0085 e^{-\frac{\Delta F}{RT}} = 0.0085 e^{5.53} \approx 2.$$

This shows that the iodine would be present as 67% CsI and 33% NaI. These levels become modified on consideration of the other equilibria (5) and (6), for example.

Since the value of $\ln K$ for each reaction appears in Table 29, the implications of these values will now be discussed for each of the reactions.

1) At 1000°K, dissociation at 1 atm is very small (<5%). Higher pressures (which would be the case in the bond after burnup) further suppress dissociation.

2) UI_3 is the most stable species at the temperatures under consideration. At $1000^\circ K$ it is unlikely that there would be appreciable vapor pressure of I_2 or UI_3 (or UI_4) and therefore, the release of iodine could be low. On the other hand, the higher temperature of the fuel ($>1000^\circ K$), could result in the escape of precursors which would decay to iodine, and thus appreciable release might occur before the formation of UI_3 . It would seem therefore, that the temperature history of the accident is of utmost importance. If iodine is mostly retained in the fuel during steady state reactor operation, then reaction kinetics and the details of the flow patterns could largely dictate the form of the iodine and its behavior.

3) With $\ln K = 45$ at $1000^\circ K$, the thermodynamics strongly favor the formation of sodium iodide.

4) This calculation shows that $a_{CsI}/a_{NaI} \cong 2$ together with reactions (5) and (6) ($K_5 \cong -0.35$ & $K_6 \cong 2$), which indicate $a_{CsI}/a_{RbI} \cong 1/5$ and $a_{NaI}/a_{SrI_2} \cong 10$ giving an approximate distribution:

CsI - 50%, NaI - 30%, RbI - 10%, SrI_2 , BaI_2 each 5%.

7) Since zirconium is a major fission product and because its iodide is volatile, its stability was examined. The value for $\ln K$ is -70, which is equivalent to no reaction.

8) Though $NaBr$ is more stable than NaI , the small amount of bromine generated makes it less important. In addition, the properties (b.p.) of the bromides closely resemble those of the important iodides.

9) To examine possible reactions with container materials, the formation of CrI_2 was considered since it is more stable than the iron and nickel iodides. The result indicates that no reaction is likely.

The results which have been presented imply that during the steady state operation of a sodium bonded-metallic fuel rod, fission products can be divided into these three groups.

1) Inert gases which are partly retained in the fuel but which are more likely to concentrate in the fuel rod gas space and develop pressure

2) Volatile elements and compounds (I, Cs, Rb, UI) which on release could (in the case of metals and iodine) form compounds, and/or dissolve in the sodium bond

3) Nonvolatile elements and those which form nonvolatile compounds in the fuel and remain in the fuel.

Under these circumstances, it is possible that many of the fission products are present as compounds which would not leave the reactor under conditions of transient heating and even uranium melting (1405°K). The most volatile stable (chemically) iodides NaI, CsI and RbI all have boiling points >1500°K. If fuel temperatures were much greater than 1500°K, the release would depend on the course of the vaporized materials. It is notable that the bonded elements tend to keep the fission products in solution form and this permits reaction to the stable iodides. This appears to be an inherent safety feature of bonded fuel elements.

In summarizing the results of this study, the following conclusions may be noted.

1) Fission product generation was calculated for both stable and radioactive species to arrive at typical concentration levels resulting from several months of steady state operation of the reactor.

2) The possibility of compound formation was investigated and it was shown that the compounds of greatest importance were iodides of the electro-positive fission products, sodium and uranium.

3) The free energies of formation for the iodides were compiled. The order of chemical stability was found to be:

4) The equilibrium: $\text{UI}_3 + 3\text{Na} \rightleftharpoons 3\text{NaI} + \text{U}$ was found to be displaced strongly to the right at typical reactor operating temperatures, that is, the released iodine would be expected to be present in the bond as NaI. The properties of sodium and the more stable iodides were examined. The most volatile iodides were those of cesium, rubidium and sodium, all of which have normal boiling points in excess of 1500°K.

5) Analysis of the equilibrium: $\text{NaI} + \text{Cs} \rightleftharpoons \text{CsI} + \text{Na}$ for typical cesium concentrations (assuming most of it is released to sodium) indicated that CsI is present in concentrations somewhat greater than NaI.

6) In order to obtain a quantitative estimate of the iodine concentrations, several other equilibria were examined (CsI-RbI , NaI-SrI_2). These gave iodide concentrations of approximately 50% - CsI, 30% - NaI, 10% - RbI, and 5% each Ba and Sr iodides (assuming total release from fuel).

7) The formation of ZrI_4 was determined to be negligible. This is an important result because of the high Zr concentrations and the volatility of its iodide. Reaction with the steel was examined (NaI-CrI_2 equilibrium) and also proved to be negligible.

These results indicate that the mechanism of iodide retention by bonded, metallic fuel elements may be iodide formation with fission products and sodium. The fact that the most chemically stable iodides have high boiling points (exceeding the melting point of uranium) provides an inherent safety factor.

B. RADIOLOGICAL EVALUATION OF THE MOST SIGNIFICANT ISOTOPES (W. Kunkel)

There are several factors which must be considered in evaluating the response of a sodium reactor system to an accident condition. Among these are the physical inventory of fission products present (the yield and production characteristics of these isotopes), their volatility, and any special characteristics which may make them amenable to chemical or physical adsorption, reaction, or other retention mechanisms. All of these factors, therefore, determine to some extent the behavior conditions of the isotopes under the special conditions of environmental pressure, temperature, and other physical and chemical factors which may exist during a reactor malfunction.

To describe the behavior of all of the fission products produced in a nuclear incident, or the fate of those in the core inventory under all of these conditions may be enlightening, but superfluous. It may indeed require only the systematic demonstrations of fission product retention for a few key isotopes to modify considerably the environmental results (and the siting requirements) of a reactor under these circumstances.

It was therefore considered advisable in the project to describe a "relative hazard index" of the various fission products to be considered. This would describe the level at which the controlling isotope (in terms of hazard or environmental effect) is no longer the determining factor in assessing the reactor radiological results.

The first attempt at this task was completed by evaluating the inventory in the core as a function of time and operating conditions. Each isotope was then described in terms of its relative hazard by multiplying the inventory factor by the reciprocal of the MPC* (which would then represent a hazard factor).

Assuming the complete inventory suggested to be released to the environment, and further assuming that all the isotopes described show equal tendency to deposit or fall out enroute, Figure 22 is obtained. The data obtained are normalized to I^{131} for two reasons. First, iodine represents the primary hazard index as far as radiological criteria are concerned, and secondly, the project is

Figure 22. Radiological Hazard Evaluation

*The Maximum Permissible Concentration (MPC) was obtained from those suggested for non-occupational exposure. Specifically, the number relates to the values recommended by the ICRP Committee on Permissible Dose for Internal Radiation and therefore represents an inhalation exposure recommendation.

directed primarily towards an evaluation of iodine retention in sodium. For this reason, it is advisable to determine at which point the I^{131} is no longer the controlling isotope.

It should be obvious that the assumption of 100% release fractions for all isotopes is not valid, either for the iodine release fractions as derived experimentally in this project, or for the lower release fractions of the nonvolatile elements. In Figure 23, therefore, are presented the same curves as modified by including the release fraction data suggested by the USAEC report²¹ TID-14844, March 1962. These fractions are strictly appropriate only for pressurized and boiling water reactors, but are presented to show the extent to which these curves become modified, in a reactor situation.

The results of these data would seem to imply that if a nominal iodine release fraction from the fuel is assumed (i.e., 40%), and if a release fraction of 10^{-2} or less could be demonstrated in every instance, then I^{131} would no longer be the controlling isotope, at least with regard to the hazard index postulated here. It should be pointed out, however, that these situations may be modified by consideration of both the submersion dose aspects of xenon and krypton, as well as the different factors which must be considered in assessing the environmental dose under accident conditions. The latter factor relates to the whole body vs thyroid dose criteria evolved for reactor site acceptability, and becomes more complex and less generalized due to shielding, wind conditions, deposition, etc.

Figure 23. Modified Radiological Hazard Evaluation

IV. EVALUATION OF RESULTS

A. IODINE-SODIUM SYSTEMS

The experiments which have been conducted at Atomics International concerning fission product retention have encompassed a wide variety of experimental conditions. It is difficult therefore, to correlate the results in a meaningful manner without arriving at a common method of expressing the activity release from sodium.

Recently, it has been suggested* that a "distribution coefficient" to express the fraction of radioactivity in the gas phase above a sodium pool to the radioactivity in the sodium. This expression has been amplified and expanded by Pollock (see Appendix D), and provides such a basis of comparison for most of the equilibrium release studies.

The distribution coefficient as defined at AI can be expressed as follows. $\text{Log } K_d = 4500/T - 5.66$ for iodine release from sodium, where T refers to the temperature of the system in °K.

Subject to the approximations and limitations assumed, the results of the project experimental studies at Atomics International have been expressed in terms of the distribution coefficient and are compared with the calculated value of K_d for iodine and cesium results. As will be discussed later, the concepts are valid for equilibrium conditions only, and thus not all of the data included in this report are applicable for comparison.

To express the experimental release fractions in terms of a distribution coefficient, the following equation was used:

$$K_d = \frac{A_g / \text{Na}_g}{A_l / \text{Na}_l},$$

where A_g and A_l = measured radioactivity in the gas and liquid, respectively, since the radioactivity can be related to the number of atoms present through the decay constant. Na_l then refers to the amount of sodium in the liquid phase,

*Private communication, A. E. J. Eggleton, AERE, HARWELL; also DRDT, AEC

and Na_g refers to the amount of the sodium as vapor in the gas phase. It should be pointed out, of course, that all of the values were measured experimentally with the exception of Na_g . This was obtained from the gas law relationship.

As may be noted from the arrangement of the equation terms,

$$K_d = \frac{A_g}{A_l} \times \frac{Na_l}{Na_g}$$

and since A_g/A_l is merely the release fraction, the remaining terms include the geometry relationships, and temperature of the system, i.e.,

$$\frac{Na_l}{Na_g} = \frac{MRT}{KPV},$$

where

M = mass of sodium in grams

R = the gas constant

T = system temperature (°K)

K = number of grams of material/mole (23 for sodium)

P = vapor pressure of sodium (atmospheres) at temperature, T

V = volume of gas above sodium pool (liters).

Therefore:

$$K_d = 3.56 \times 10^{-3} \frac{MT}{PV} \times (\text{release fraction}).$$

The values of these experimental parameters, as well as the release fractions are summarized in Table 30 for the iodine studies of Hart and Nelson.

In a similar manner, the experimental parameters of the out-of-pile experiment OP-1, the 500°F in-pile capsule experiment (IP-2) and the 1000°F in-pile capsule experiments (IP-3 and IP-4) are shown in Table 31.

TABLE 30
SUMMARY OF EXPERIMENTAL PARAMETERS FOR
IODINE RELEASE EXPERIMENTS
OF HART AND NELSON

	T (°F)	M (gm)	V (liters)	Release Fraction
I-1	923	4.3×10^3	4.77	1.1×10^{-6}
	1112	4.3×10^3	4.77	5.1×10^{-6}
	1210	4.3×10^3	4.77	1.9×10^{-6}
	1305	4.3×10^3	4.77	3.8×10^{-6}
	1410	4.3×10^3	4.77	1.7×10^{-6}
I-2	927	4.3×10^3	4.77	1.3×10^{-6}
	1212	4.3×10^3	4.77	3.3×10^{-6}
	1298	4.3×10^3	4.77	2.7×10^{-6}
	1446	4.3×10^3	4.77	3.8×10^{-6}
	1517	4.3×10^3	4.77	1.2×10^{-6}

TABLE 31
EXPERIMENTAL PARAMETERS OF OP-1, IP-2, IP-3
AND IP-4 EXPERIMENTS

Experiment	Sodium Mass (gm)	Cover Gas Vol. V (liters)	Temperature T (°K)	Release Fraction
OP-1	50.9	3.0	1033	8.9×10^{-4}
IP-2	217	3.0	533	1.9×10^{-5}
IP-3	217	4.5	811	$4.0 \times 10^{-5} - 1.4 \times 10^{-4}$ *
IP-4	217	30	811	4.3×10^{-5}

*Lower Release Fraction includes activity rinsed from capsule walls after sodium draining. Higher release fraction refers only to iodine activity recovered in the molten sodium.

Figure 24. Distribution Coefficients, Iodine-Sodium System

The iodine distribution coefficients obtained experimentally from the Hart-Nelson studies, as well as the OP-1, IP-2, IP-3 and IP-4 experiments are presented in Figure 24 as a function of the reciprocal temperature in $^{\circ}\text{K} (\times 10^4)$. Included for comparison is the plot of the theoretical distribution coefficient computed by Pollock from the equation:

$$\text{Log } K_d = \frac{4500}{T} - 5.66.$$

As may be noted from the figure, there is in general rather good agreement with this preliminary theoretical model.

It should be pointed out however, that the 500°F data from IP-2 represents only one experimental point and that the agreement may therefore be fortuitous. Similarly, in both IP-3 and IP-4 experiments, the gas was being circulated during the KEWB burst, and it is quite possible that both experiments might thus reflect the presence of radioactive iodine which may have been physically entrained in the gas stream. One might expect therefore, that these experimental values might be higher than the theoretical value predicted.

The release fractions obtained from the iodine experiments of Begley in which a bubble of iodine vapor was released from a submerged capsule have similarly been expressed as distribution coefficients, although as will be noted presently, they are over four orders of magnitude higher than the equilibrium conditions in which mixing of the iodine and sodium vapor was not mitigated by the presence of an inert gas carrier. These results are shown in Table 32.

As pointed out by Begley (Sec. III A-2), the presence of oxygen in the tests conducted at 1000°F casts doubt upon the experimental results at that temperature. Thus, these values are not included in the summary discussion.

It is not immediately apparent from the scatter of these data that there is any strong dependence of distribution coefficient upon either the capsule pressure,

TABLE 32
EXPERIMENTAL PARAMETERS AND DISTRIBUTION COEFFICIENTS
FOR BEGLEY IODINE RELEASE EXPERIMENTS (500°F)

<u>Sodium Depth 6.0 ft</u>				
	<u>Capsule</u>	<u>Capsule Pressure (atmos)</u>	<u>N₂ Conc.</u>	<u>K_d</u>
	1	1.46×10^{-2}	0.5	6.22×10^6
	6	6.05×10^{-2}	0	8.30×10^5
	8	1.46	0.5	4.16×10^6
<u>Sodium Depth 10.0 ft</u>				
	4	1.71×10^{-1}	0.5	4.16×10^4
	10	1.46	0.5	8.3×10^5

or the depth of the sodium. However, the average of the tests at the six ft depth results in a higher distribution coefficient than those at the ten ft depth. It can be more definitively stated however, that there does appear to be a much higher distribution coefficient resulting from the conditions of this type of test in which iodine vapor is released as a bubble through a column of sodium than from any of the equilibrium conditions discussed above.

B. CESIUM-SODIUM SYSTEMS

The experimental results of the cesium-sodium studies are not delineated as clearly as in the case of the iodine studies. It is of interest, however, to present the results of the Hart-Nelson data on cesium release from sodium systems in terms of distribution coefficients. This has been done by using the same technique as the iodine studies. These results are presented in Figure 25. The theoretical curve drawn in Figure 25 was obtained merely by using a Raoult's Law Calculation to obtain the number of moles of cesium in the vapor phase for a given concentration of cesium in the sodium and thus may be a very crude model. The agreement differs markedly above a temperature of 980°K. The three parallel slopes of the theoretical distribution coefficients were obtained by multiplying the effective vapor pressure of cesium at the temperatures involved by an activity coefficient (γ) of 2, 3, or 4.

Figure 25. Distribution Coefficients, Cesium-Sodium System

Figure 26. Distribution Coefficients, Strontium-Sodium System

Hart and Nelson also point out (Sec. III A-1) that there appeared to be cesium activity in the reaction vessel at the higher temperatures even after the sodium was dumped. It is quite possible therefore that this resulted from cesium plating, and that the data at the higher temperatures may be somewhat suspect for this reason. It may also be noted that there is considerable scatter of the data even for temperature points differing only by a few degrees.

Studies are under way on another project at Atomics International in which the evaporation behavior of cesium in a sodium system will be studied extensively. Until further data are available, these cesium results therefore should be used cautiously.*

C. STRONTIUM-SODIUM SYSTEMS

The strontium data have similarly been expressed as distribution coefficients and are presented in Figure 26. The theoretical correlation which seemed partially appropriate for the sodium-cesium system because of their similar chemical characteristics, does not provide any comparable curve for the strontium distribution coefficients. The figure shows rather good distribution of the experimental points, however. Until a suitable analytical model is

*The relative stability of cesium nitrate as compared to metallic cesium in sodium might also be important in determining the extent of retention.

developed, therefore, the best fit to the experimental points has been drawn.

D. APPLICATION TO REACTOR SAFEGUARD ANALYSIS

With the expression derived for the distribution coefficient, and the apparent agreement of most of the experimental data with the theoretical iodine distribution coefficient, it appears possible to check these small scale results with different system configurations in which there are variations in the system geometry, temperature, and volume ratios. In this manner, it is possible to solve for the ratio of iodine radioactivity in the gas and liquid phase, by using a scaling factor which involves only the vapor pressure of sodium, temperature, distribution coefficient, and the ratio of the cover gas volume (liters) to the sodium mass (gm).

Therefore, after inserting the applicable constants, the release fraction of radioactivity in the system from the sodium pool to the cover gas may be expressed as

$$\frac{(A)_{\text{gas}}}{(A)_{\text{sodium}}} = \text{release fraction} = (K_d)_T \times 2.8 \times 10^2 \frac{PV}{MT},$$

where

$(A)_{\text{gas}}$ and

$(A)_{\text{sodium}}$ = radioactivity in cover gas and sodium, respectively,
(curies)

$(K_d)_T$ = theoretical distribution coefficient at temperature T,
(°K)

P = vapor pressure of sodium at temperature T (atmos),

V = cover gas volume (liters), and

M = mass of sodium in pool (gm).

This expression is plotted in Figure 27 as a function of the inverse temperature for V/M = 1.0. It is suggested that as a first approximation it may represent a model from which the release fraction of any equilibrium system in this

Figure 27. Extrapolation Equation, Iodine-Sodium System

temperature range may be determined merely by multiplying the curve values by the appropriate ratio of the cover gas volume (liters) to the sodium mass (gm).

Application of the equation to typical reactor conditions can also be made, although in this case, the mode of release may apparently influence the extent of iodine release, judging from the one experiment involving bubble release in which data were obtained.

System conditions for the Sodium Reactor Experiment are of course not typical for many of the larger sodium graphite reactor installations (such as the HNPF, for example) nor are they typical of fast reactor conditions. However, they are used here to demonstrate the general applicability of the extrapolation equation, and to indicate the order of magnitude release figures which may be obtained from a reactor malfunction releasing fission products into the sodium, but in which the integrity of cover gas over the sodium in the core tank is maintained. These system parameters are shown in Table 33.

TABLE 33
SRE SYSTEM CONDITIONS FOR TYPICAL
IODINE-SODIUM RELEASE ESTIMATE

Mass of sodium in core tank	47,000 lb
Volume of cover gas	400 ft ³
Temperature of upper plenum bulk sodium	1200°F

If one assumes that the fission product release into the SRE sodium would be gradual (as a result of a slow power ramp which might cause some cladding ruptures, for example), the equilibrium conditions of the system would be assumed to apply. In this case, the radioactivity release of three of the most significant isotopes (radiologically) can be estimated as shown in Table 34. (Note that these release values do not include release from fuel.)

The iodine release numbers are considered more substantive than the cesium and strontium, since the iodine release values agree both experimentally and theoretically in small-scale studies. These estimates represent only the application of the preliminary model however, and may obviously require modification.

If a more violent dispersion of fission products is assumed, then perhaps the bubble simulation is more realistic. No data are available on the strontium and cesium release conditions in this case. However, the iodine release number might approximate 1.3×10^{-2} .

TABLE 34
TYPICAL FISSION PRODUCT RELEASE
FROM SODIUM AS ESTIMATED BY
EXTRAPOLATION EQUATION
FOR THE SRE

<u>Isotope</u>	<u>Estimated Release Fraction</u>
Sr ⁸⁹	1.7×10^{-7}
I ¹³¹	2.1×10^{-6}
Cs ¹³⁷	5.1×10^{-4}

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V. SUMMARY AND CONCLUSIONS

The complexity of the fission product-sodium-fuel environment needs no emphasis. This summary report has attempted to describe only the salient details of the experiments which have been conducted in this area at Atomics International as well as some of the analytical studies and techniques. Further detailed information on each of the experiments is available in the reference literature.

A general criticism of the studies is that there are few related parameters or duplications of experiments so that a cohesive comparative survey of the results is difficult. Similarly, the range of experimental error is difficult to evaluate. In spite of these inadequacies however, the following conclusions are drawn.

1. With regard to the experimental techniques, it appears that both in-pile and out-of-pile studies are necessary for the simulation of various conditions. The general agreement of the iodine release distribution coefficients for both in-pile and out-of-pile experiments under equilibrium conditions is gratifying, but unfortunately not enough variables have been included to provide more than a preliminary indication. For example, the effect of the bubble transport appears to present anomalous data in all of the studies performed.
2. The May pack and diffusion tube sampling technique in their present form appear to have limited application to the study of fission product form after transit through a sodium plenum. The May pack response was seen to change with the preconditioning of the May pack components by the passage of sodium vapor. The problem of the diffusion tube arises because of the condensation of sodium vapor on colder tube walls and the consequent obscuring of iodine specific response media used in the diffusion tube arrangement. It may be that the quadrupole mass spectrometer, which was being investigated late in the project, or the temperature gradient deposition technique⁶ may be required to ultimately supply the answer to the important corollary questions as to the form of the fission product species, and the extent to which it is present in the cover gas after passing through the sodium medium.
3. In SRE and fuel capsule studies, the effect of plating and deposition of iodine and other fission products from the sodium environment was noted to play an

important part in source depletion. For this reason, it is suggested that a major part of any future effort in fission product retention studies should relate to the role of geometry and plating factors in varying the behavior of fission product transport.

4. The results of the plateau heating experiment and the sodium surface reaction experiment suggest that there is a noticeable inhomogeneity of fission product activity in the sodium. It is not clear as to whether this is a phenomenon arising from the cooling and stratification of activity, or is due to convection or other causes. There is considerable disparity noted, however, in comparing samples of various locations.

5. There appears to be definite indication from the SRE studies and the work of Hansen at AI that the presence of carbon (and perhaps other impurities in the sodium) may be important in the depletion of fission products circulating in the sodium coolant after release. In this respect, the correlation of the carbonaceous deposit and the radioactivity as well as the SRE cold trap results might bear further investigation. This problem is also under attack presently in another project at Atomics International.²³

6. The distribution coefficient model presented by Pollock provides a preliminary model for the release of iodine from an equilibrium sodium system. It is suggested that this be applied to further experimental conditions in which iodine release has been measured to determine its validity over a wider range of situations.

7. One might have expected the violent dispersion of the IP-2, IP-3, and IP-4 experiments to have produced results tending toward the non-equilibrium aspects of the bubble transport experiments. The reasons that they did not do so are not completely clear, but may be associated with the gas transport of either inert gas or iodine vapor. Since the effect of bubble transport has presented anomalous data, it is suggested that this particular aspect of the problem be studied extensively in future studies because of its obvious application to reactor safeguard analysis.

APPENDIX A

ANALOGUE STUDIES OF KEWB TRANSIENT

Figure 28 shows a portion of the results of some analog studies using a simplified analytical model describing the transient energy input into a fuel disc during a KEWB burst. The model assumed no film resistance to heat transfer between uranium and sodium, no phase change of sodium, and no dispersal of uranium during the transient. These assumptions are incorrect, but the analog studies were useful in providing an estimate of the transfer of heat to sodium and in providing an estimate of the rate of change of temperature in the fuel disc. From this figure, it can be seen that the time from the initiation of the burst to the maximum temperature point is 6.5 msec and the time for the average fuel temperature decrease would probably be reduced because of the dispersion of uranium into the sodium heat sink. The fact that very little of the heat is lost to the sodium during the period of peak temperature rise and that the rate of temperature drop after the burst is quite steep are useful in explaining some of the fission product release phenomena noted during the experiment.

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Figure 28. Transient Energy Input, KEWB Burst

There is a variety of parameters which affect energy release within the sample; e.g., enrichment, geometry, location, alloying elements, and reactor period. Consequently, extrapolation of previously obtained test data and analog results to the expected conditions resulted in a range of anticipated sample temperature. The range varied from 1470°C (360°C above melting) to 3900°C (the boiling point).

APPENDIX B

ISOTOPE DECAY CHAINS AND PHYSICAL CHARACTERISTICS

In Figures 29 through 32 are summarized the decay chain, isotope characteristics, and probable generation resulting from the KEWB transient irradiation of Experiment IP-1. These results have been obtained by using the system conditions and applying the idealized curves²⁴ of USNRDL-456.

Figure 29. Characteristics of Zr^{95} and Precursors

Figure 30. Characteristics of Ru^{103} and Precursors

Figure 31. Characteristics of I^{131} and Precursors

Figure 32. Characteristics of BaLa^{140} and Precursors

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APPENDIX C
RESULTS OF SECOND IN-PILE EXPERIMENTS

TABLE 35
COVER GAS ACTIVITY (EXPERIMENT IP-2)

Component	Isotope	Total Activity (μc)
Sodium vapor trap No. 1	Xe ¹³³	6.3×10^{-3}
	BaLa ¹⁴⁰	1.4×10^{-3}
	I ¹³¹	Not detected
Sodium vapor trap No. 2	Xe ¹³³	6.3×10^{-3}
	BaLa ¹⁴⁰	2.5×10^{-3}
	I ¹³¹	Not detected
Millipore and AgNO ₃ filter	Xe ¹³³	Not detected
	BaLa ¹⁴⁰	1.8×10^{-3}
	I ¹³¹	Not detected
Charcoal trap No. 1	Xe ¹³³	5.7
	BaLa ¹⁴⁰	1.6×10^{-2}
	I ¹³¹	Not detected
Charcoal trap No. 2	Xe ¹³³	9.0
	BaLa ¹⁴⁰	7.7×10^{-3}
	I ¹³¹	Not detected
Charcoal tube	Xe ¹³³	240
	BaLa ¹⁴⁰	Not detected
	I ¹³¹	Not detected
Evacuation flask	Xe ¹³³	1.04×10^3
	BaLa ¹⁴⁰	Not detected
	I ¹³¹	Not detected

TABLE 36
SUMMARY OF RECOVERED FISSION PRODUCT ACTIVITY FOR
EXPERIMENT IP-2

Sample	ZrNb ⁹⁵	Ru ¹⁰³	I ¹³¹	Xe ¹³³	Cs ¹³⁷	BaLa ¹⁴⁰
No. 1 sodium*	5.68×10^3	+	1.78×10^5	+	1.55×10^4	3.34×10^5
No. 2 sodium*	6.56×10^3	+	2.03×10^5	+	1.75×10^4	3.79×10^5
Filter §**	1.30×10^8	1.11×10^8	7.27×10^7	+	+	3.40×10^9
Alcohol rinse §	5.10×10^6	1.49×10^4	1.18×10^8	+	5.23×10^3	4.87×10^7
No. 3 sodium*	Not meas.	+	2.13×10^6	+	Not meas.	3.10×10^6
No. 4 sodium*	Not meas.	+	2.17×10^6	+	Not meas.	3.53×10^6

*dpm/gm

+below limits of sensitivity

§total in dpm

**amount left on fritted glass

APPENDIX D

ANALYSIS OF SODIUM-IODINE SYSTEM

An analytical study of the evaporation of sodium iodide from sodium has been made using information available in the literature. For purposes of this work, the vaporization of iodine (as iodide) from sodium solutions can be meaningfully expressed in the form of a "distribution coefficient," K_d , which is defined in terms of molar concentrations in the vapor and liquid, $K_d = X_{2,g}/X_{2,l}$, where "X" refers to the mole fraction, the subscript "2" refers to the solute NaI, and g and l refer to the gas and liquid phases. Recalling the gas law, one may express $X_{2,g}$ in terms of partial pressures,

$$X_{2,g} = P_{\text{NaI}} / (P_{\text{Na}}^{\circ} + P_{\text{NaI}}) \cong P_{\text{NaI}} / P_{\text{Na}}^{\circ},$$

since the partial pressure of the NaI would be much less than that of sodium. Further, one may express the partial pressure of the sodium iodide over sodium in terms of its vapor pressure and concentration as follows:

$$P_{\text{NaI}} = \gamma X_{2,l} P_{\text{NaI}}^{\circ},$$

where γ is the "activity coefficient." Inasmuch as the experiments which have been performed, as well as typical reactor accidents represent dilute solutions, it is reasonable to make the assumption that the solution will be in the "Henry Law" region, that is, that the activity coefficient is approximately constant with concentration in the temperature range of interest. It is therefore possible to evaluate the activity coefficient in terms of the solubility of sodium iodide in sodium, i.e., $\gamma = 1/X_{2,l}^{\circ}$, where $X_{2,l}^{\circ}$ is the solubility of NaI in Na. This is of course a function of temperature. Making the indicated substitutions, one finds for K_d ,

$$K_d = \frac{X_{2,l} P_{\text{NaI}}^{\circ}}{X_{2,l}^{\circ} P_{\text{Na}}^{\circ}} \Bigg/ X_{2,l} = \frac{P_{\text{NaI}}^{\circ}}{P_{\text{Na}}^{\circ} X_{2,l}^{\circ}}.$$

It will be convenient to use the logarithmic form,

$$\text{Log } K_d = \text{Log } P_{\text{NaI}}^{\circ} - \text{Log } P_{\text{Na}}^{\circ} - \text{Log } X_2^{\circ}, (T).$$

The solubility data of Bredig, Johnson and Smith²⁷ for NaI in Na may be used and are well expressed at temperatures below the melting point of NaI by the expression, $\text{Log } X_2^{\circ} = -9,300/T + 8.19$. Bauer and Porter²⁶ give data for the vaporization of NaI in the form of ΔH_{298}° and free energy functions. An excellent approximation good over a range of 200-300 degrees can be made because of the fact that free energy functions do not change rapidly with temperature. The free energy of vaporization can therefore be expressed by a simple equation:

$$\begin{aligned} \Delta F_T^{\circ}(\text{vap}) &= - \left[- \frac{\Delta F_T^{\circ} - \Delta H_{298}^{\circ}}{T} \right]_{800} T + \Delta H_{298}^{\circ} \\ &= -34.38T + 47,900. \end{aligned}$$

Recalling that $\Delta F_T^{\circ} = -RT \ln P$, and solving for $\text{Log } P$,

$$\begin{aligned} \text{Log } P &= \frac{34.38}{4.576} - \frac{47,900}{4.576T} \\ &= - \frac{10,500}{T} + 7.51. \end{aligned}$$

In a similar manner, the partial pressure of the sodium monomer can be derived from the data in Stull and Sinke's compilation,²⁷

$$\text{Log } P_{\text{Na}}^{\circ} = \frac{-5,700}{T} + 4.98.$$

Combining the expressions for the pressures and solubility data leads to the equation for K_d ,

$$\text{Log } K_d = \frac{4,500}{T} - 5.66.$$

Note that the contribution of Na_2 to the vapor composition has been neglected, however, this involves only a few percent error and does not materially affect the calculated results. According to this treatment, $K_d = 1.0$ at about 800°K (or rather 796°K), and becomes larger with decreasing temperature.

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