265
7/22/68
+105 FAST REACTOR

363

AI-AEC-12687
COPY

Dr. 363

IR

LMFBR FISSION PRODUCT CONTAMINATION
AND CONTROL STUDIES -
A STATUS REPORT

AEC Research and Development Report

ATOMICS INTERNATIONAL

A DIVISION OF NORTH AMERICAN ROCKWELL CORPORATION

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

Printed in the United States of America
Available from

Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U.S. Department of Commerce
Springfield, Virginia 22151

Price: Printed Copy \$3.00; Microfiche \$0.65

LMFBR FISSION PRODUCT CONTAMINATION
AND CONTROL STUDIES -
A STATUS REPORT

By

W. P. KUNKEL
B. D. POLLOCK
J. GUON
G. B. ZWETZIG
M. SILBERBERG
S. BERGER

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

ATOMICS INTERNATIONAL

A DIVISION OF NORTH AMERICAN ROCKWELL CORPORATION

CONTROL AT(04-3)-701
ISSUED: JUNE 30, 1968

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

leg

DISTRIBUTION

This report has been distributed according to the category "Reactor Technology" as given in the Standard Distribution for Unclassified Scientific and Technical Reports, TID-4500.

CONTENTS

	Page
Abstract	6
I. Summary	7
II. Introduction.	11
III. Source Term Characterization Studies	13
A. Typical Fast Breeder Reactor Characteristics	13
B. Radioisotope Flow Chart	15
1. Sources of Coolant Radioactivity	15
2. Depletion Mechanisms for Coolant Radioactivity	16
C. Relative Importance of Fission Product Isotopes	17
1. Radiological Hazards Index	18
2. After-Shutdown Radiation Aspects	19
3. Relative Concentration of Fission Products and Corrosion-Activation Isotopes	21
D. Fission Product Grouping	26
1. Cesium.	27
2. Barium and Strontium	27
3. Molybdenum, Ruthenium, Antimony, and Silver.	27
4. Rare Earths, Yttrium and Zirconium	30
5. Niobium	30
6. Tritium, Iodine and Tellurium	30
E. Source Model Evaluation	30
IV. Experimental Studies	31
A. Cesium and Iodine Vaporization from Sodium	39
1. Analytical Studies and Supplementary Literature Data.	40
2. Experimental Methods.	44
3. Experimental Results	50
4. Evaluation of Results	54
B. Deposition and Trapping Studies	55
1. Cold-Trapping of Cesium in Sodium.	55
2. Fission Product Behavior Loop.	61
V. System Studies.	63
A. Reduction of Fission Products Observed in Sodium.	63
B. LMFBR System Behavior.	67
References	77

TABLES

	Page
1. Design Characteristics of Reference LMFBR	13
2. Solubility of Metal Elements in Sodium and Free Energy of Formation of Oxides and Carbides at 800°K	28
3. Reported Existence of Binary Compounds of Fission Products and Major Constituents of Type 304 Stainless Steel.	29
4. Analogue Grouping and Expected Fate of Various Fission Products.	29
5. Relative Volatility of Cesium at Low Concentration in Sodium	51
6. Relative Volatility of Sodium Iodide in Sodium	53
7. Activity Coefficients	54
8. Design Parameters for Fission Product Behavior Loop.	62
9. Radioisotopes for Fission Product Behavior Loop Studies	62
10. Time-Dependence of Fission Product Contamination in SRE Primary Sodium	63
11. Total and System Depletion Factors for SRE System.	66
12. Fraction of Available Inventory Removed from Sodium, 30 Years Operation, Continuous Fission Product Release	75
13. Dose Rates 3 ft from Surface of Heat Exchanger Containing Primary Sodium, 10 Days After Shutdown, Following 30 Years of Reactor Operation	76

FIGURES

1. Radioisotope Flow Chart	14
2. Radiological Hazard Index vs Time After Shutdown.	20
3. Radiation Index vs Time After Shutdown.	22
4. Activity Concentrations for Corrosion Products, Fission Products and ²² Na vs Reactor Operating Time	24
5. Primary and Auxiliary Loops for Assumed "Typical" LMFBR	32
6. Relative Coolant Inventory of Hypothetical Isotope vs Time After Shutdown for Case 1 and 2 Release Mechanisms.	36
7. Relative Plated Inventory for a Hypothetical Isotope vs Time After Shutdown for Case 1 and 2 Mechanisms	36
8. Schematic Diagram of Vapor Density Apparatus.	45

FIGURES

	Page
9. Vapor Density Apparatus Removed from Furnace.	45
10. Schematic Diagram of Transpiration Apparatus	47
11. Disassembled Bubbler Used in Transpiration Apparatus	48
12. Cs-Na System, Calculated and Experimental Relative Volatility. . .	51
13. NaI-Na System, Calculated and Experimental Relative Volatility . .	53
14. Schematic Diagram of Cold Trapping Apparatus.	57
15. Temperature Data for Typical Cold Trapping Run	58
16. Radiation Level in Cold Trap vs Cold Trap Inverse Temperature for Various Oxide Concentrations	60
17. Radiation Level in Cold Trap vs Cold Trap Inverse Temperature for Two Cesium Concentrations	60
18. Fission Product Behavior Loop.	62
19. Total Radioactivity Depletion Characteristics for SRE System	65
20. System Depletion Characteristics for the SRE System.	68
21. Radiation Levels 3 ft from Surface of Heat Exchanger — Heat Exchanger Drained	70
22. Radiation Levels 3 ft from Surface of Heat Exchanger — Heat Exchanger Remains Filled with Primary Coolant	70
23. Percent Contribution of Individual Fission Product Isotopes to Total Fission Product Radiation Level	72
24. Estimated Removal Fraction for Significant Fission Products	72

ABSTRACT

The status is presented of studies and experiments being conducted at Atomics International to describe the distribution of fission products and other contamination sources released to the coolant of a Liquid Metal Fast Breeder Reactor (LMFBR). These studies include the formulation of Radiological Hazard and Radiation Indices to aid in determining the most significant fission products, the evaluation of the importance of activated core materials which are subsequently corroded into the coolant stream, the classification of fission products on the basis of physical and chemical properties into analog groupings to reduce the number of isotopes which must be studied in sodium test loops, and a preliminary study of the residual radiation levels which could result from fission product plating on heat exchanger surfaces, thereby limiting access for maintenance or repair.

Experimental results derived from studies of cesium and sodium iodide vaporization from sodium solutions and cesium cold trapping studies are presented. The Fission Product Behavior Loop which will be used for a more comprehensive series of investigations of fission product behavior in sodium is also described.

I. SUMMARY

The status is presented of studies being conducted to describe the distribution of fission product and other contamination sources released to the coolant of a Liquid Metal Fast Breeder Reactor (LMFBR). These studies fall into three major areas: Source Term Characterization, Experimental Studies, and System Studies. The Source Term Characterization effort attempts to define the problem by determination of the important sources of radioactivity; their relative importance within the group; the various processes by which they may influence reactor operation, maintenance or safety; and the effect of parametric variations of the system characteristics on the influence of the various isotopes. The Experimental Studies effort attempts to develop data which will permit quantitative prediction of the behavior of the isotopes determined to be important by the Source Term Characterization Studies. Finally, the Systems Studies goal is to integrate the foregoing experimental and analytical results to determine the radiation and radiological conditions which occur in an overall LMFBR system.

In the area of Source Term Characterization, preliminary but incomplete specifications for a "typical" LMFBR system have been established, and the various sources of radioactivity and their possible fate in an LMFBR have been detailed. Radiological Hazard and Radiation Indices have been calculated for moderate to long-lived fission products to determine the relative importance of the various isotopes relative to these two problems and thus provide a measure of guidance for establishment of priorities for experimental and analytical work. The radioisotope inventory generated by the corrosion of activated stainless steel cladding was estimated (based on 2 mils/year corrosion rate) and found to be about ten times that due to fission products (based on a 10^{-5} release fraction). The physical and thermochemical properties of fission products and the elements in stainless steel, along with various compounds of these elements (oxides, carbides, and intermetallics), were examined to determine chemically similar groupings of elements. By such grouping, it is anticipated that by study of the behavior of one of the elements in the group it will be possible to generally characterize the behavior of all of the elements. Among the groups identified were

those likely to: (1) volatilize, (2) deposit as oxides, (3) diffuse into the stainless steel, and (4) combinations of these. A beginning has been made on the definition of a digital computer program which will calculate the distribution of radioisotopes in the coolant, in the cover gas and on reactor primary system surfaces, as a function of time for various parametric input variables. These variables will include reactor power history, cladding failure rate, fractional release of individual isotopes from fuel, flow rates in the primary system and trapping devices, surface areas, and temperatures, etc. The code will calculate all important sources of radioactivity including ^{22}Na and ^{24}Na and corrosion products activities.

The Experimental Studies effort has concentrated on studies of the partitioning of a volatile fission product between the vapor phase and the liquid sodium in which it is dissolved. This partitioning is expressed in terms of a distribution coefficient, K_d , which is defined as,

$$K_d = \frac{Y_2}{X_2} \quad \dots (1)$$

where

Y_2 = mole fraction of solute in vapor phase

X_2 = mole fraction of solute in liquid phase

and the solute is the fission product of interest. Studies of partitioning have been conducted for the Cs-Na and NaI-Na systems using a transpiration apparatus and tracer techniques. The data from the transpiration experiments yielded the following results:

$$\text{Cs-Na} - \log K_d = [987/T(^{\circ}\text{K})] + 0.490$$

$$\text{NaI-Na} - \log K_d = [1840/T(^{\circ}\text{K})] - 2.34$$

For several possible reasons presented herein, these experimentally derived values exhibit deviations from theoretical predictions. The above relationships may be used to estimate the amounts of Cs and I isotopes that will volatilize to primary system cover gas spaces.

Experimental studies were also performed in a small, forced circulation loop to determine the effectiveness of cold traps in removing cesium. Both Na_2O and Cs concentrations were varied to determine their effects on trapping. The experimental data indicated that precipitated Na_2O was not required for removal of Cs by the cold trap. The removal of Cs was observed to increase with decreasing cold trap temperature (850 to 250°F) and with increased oxygen concentration in the sodium (5 to 105 ppm oxygen). Addition of cesium carrier reduced the apparent trapping efficiency, presumably by displacement of some of the trapped radioactive Cs atoms by stable Cs atoms.

The Systems Studies effort examined data on the rate of isotope removal from the SRE following a fuel failure incident and data on the distribution of fission and corrosion products in the coolant and on primary system surfaces following shutdown of the SNAP 8 Experimental Reactor (S8ER). From the examination of the time-dependent data obtained from the SRE, it was noted that the rate of removal could be reasonably approximated by postulating a system depletion constant for each isotope analogous to the radioactive decay constant. The activity, A , in the coolant could then be represented reasonably well by an equation of the form,

$$A = A_0 e^{-(\lambda+\alpha)t} \quad \dots(2)$$

where

A_0 = initial activity

λ = radioactive decay constant for the isotope

α = system depletion constant for the isotope

t = time

Based on the assumption that the quantity of each isotope removed from the coolant stream was uniformly deposited on primary system surfaces, the depletion constant concept was applied to a "typical" LMFBR system to determine the residual radiation levels 3 ft from a main heat exchanger, 10 days after that loop had been isolated from the rest of the primary system. Dose rates were estimated for the case where the heat exchanger had been drained

and for the case where the heat exchanger contained sodium. In this example, the shielding provided by the sodium (with 10 days decay for Na^{24}) made the dose rate lower for the heat exchanger containing sodium than for the drained heat exchanger.

The principal activities planned for the future are the initial operation of the Fission Products Behavior Loop (FPBL), which is expected to provide much valuable data on fission product behavior in stainless steel systems; the programming of the digital computer code for predicting radioisotope distributions in an LMFBR system; the use of this code to perform preliminary systems studies; and the continued surveillance and application of fission and corrosion product behavior patterns in operating reactor systems such as EBR II, Fermi, and the SNAP 8 Developmental Reactor.

II. INTRODUCTION

The safe and economical operation of a sodium-cooled fast breeder reactor is in part dependent upon reasonable access to the heat exchangers and other large components for maintenance.

To a large extent this accessibility is dependent upon residual radiation levels which, depending on maintenance procedures, determine the waiting time required after shutdown before entry can be effected. An initial waiting time is of course required for the ^{24}Na radioactivity to decrease to acceptable limits. Then, if no other radioisotopes are present ^{22}Na will provide the controlling radiation level. In the event of fuel cladding failure, however, fission products will also be introduced into the coolant. Additionally, radioactive corrosion and erosion products from the core structure, and nonradioactive corrosion and erosion products from the primary loop in general — irradiated as they pass through the core — are transferred to various parts of the plant by the coolant flow. Since these fission products and radioactive corrosion products have been observed to deposit throughout the system or, in the case of more volatile isotopes, to distribute themselves between the cover gas and the sodium coolant, the resulting radiation levels along with those due to ^{22}Na , will largely determine the extent of the accessibility of various portions of the FBR system for maintenance operations. In addition they will establish the requirements for cover gas purification, disposal, and limitations on top-shield leakage.

Aside from observations which have been made following the release of fission products into reactor systems such as the SRE,⁽¹⁾ BR-5,⁽²⁾ DFR,⁽³⁾ Fermi,⁽⁴⁾ and the SNAP 8 Experimental Reactor (S8ER),⁽⁵⁾ very little fundamental research has been directed at understanding the behavior of fission products in a reactor system.^(6,7)

The objective of the present project is to elucidate the behavior of fission products released to the coolant in a sodium-cooled LMFBR during normal operation. This information will be useful in more realistically assessing the probable consequences of potential accidents. It may also indicate the necessity for developing fission product trapping techniques which will reduce the effects of clad failures under normal operating conditions.

Experimentally, the project has studied vaporization equilibria for the volatile fission products, cesium and iodine. It also includes studies of the rate and extent of trapping by cold and hot traps, and stainless steel surfaces.

The analytical effort accompanying the fundamental experiment program has been directed towards determining which isotopes are most significant and thus most profitable to examine. Typical inventories have been evaluated with parametric release fractions from fuel elements. (Development of fission product release fractions from fuel are not part of the scope of this project.) A major goal of this portion of the effort is to derive a "source term" which is amenable to computer programming. In this manner, the information acquired from fundamental studies and from reactor operating experience can be inserted, and the analytical expressions relating to fission product location and radiation levels can be used to describe the probable results of any fission product injection into the coolant.

A study of existing reactor systems and the manner in which fission products have been distributed in reactor accidents has comprised the remaining area of analytical evaluation.

It is the purpose of this report to summarize the present state of knowledge with regard to fission product distribution, and to outline briefly the next steps by which the project will fulfill its desired objectives.

III. SOURCE TERM CHARACTERIZATION STUDIES

A. TYPICAL FAST BREEDER REACTOR CHARACTERISTICS

For a project of this nature, it is of value to establish a set of conditions which are more or less typical of those to be found in an LMFBR environment. For data acquired under conditions which are greatly different, an extensive investigation into the validity of extrapolation techniques must also be made; otherwise, the ultimate purpose for which the data are acquired, i. e., to assist in reactor design evaluations, is not achieved, and the project data are essentially of no value to the reactor designer. Thus in all cases where possible, it is considered advisable to employ experimental temperatures and other parameters which are characteristic of an actual reactor system. Table 1 shows a typical set of reactor conditions upon which activation and fission product inventories were based. It is recognized that there may be variances in many of these parameters; they are used in this project as interim numbers until more detailed information is available on the system configuration of a specific reactor.

TABLE 1
DESIGN CHARACTERISTICS OF REFERENCE LMFBR

Rated Core Power (Mwt)	2120
Sodium Inventory in Tank, Total (lb)	1.1×10^6
Flux, Estimated (n/cm^2 -sec)	5×10^{15}
Coolant Mean Circuit Time Through Primary (sec)	120
Velocity of Coolant Through Core (ft/sec)	11.1
Active Length of Core (ft)	2

Another parameter which is of importance in evaluating fission product deposition in a fast breeder reactor system is the surface-to-volume ratio, i. e., the surface of piping in contact with the primary sodium divided by the volume of the primary sodium. An examination of available data indicates that a typical surface-to-volume ratio for a loop type LMFBR might be 0.05 cm^{-1} . It is of interest to note that, for most of the systems examined, nearly 83% of

FUEL	HEAT TRANSFER SPACE	CLAD	COOLANT	COVER GAS	STRUCTURAL	CONTAINMENT	ENVIRONMENT
------	---------------------	------	---------	-----------	------------	-------------	-------------

7-S18-231-14

Figure 1. Radioisotope Flow Chart

the available piping surface area is in the associated heat exchangers themselves; another 11% is in the accessible piping associated with the heat exchanger; and about 5% is in inaccessible piping in the primary coolant circuit.

Less data are available for the "pot" geometry FBR in which the entire primary loop is submerged in a large sodium pool. However, since most of the surface area results from the heat exchanger, it seems probable that the surface-to-volume ratio under these circumstances will be approximately the same.

B. RADIOISOTOPE FLOW CHART

Because of the complexity of the system in a typical LMFBR with regard to the fate of fission and corrosion products released into the coolant, it is necessary to delineate as simply as possible the paths by which these products are injected, transported, and depleted. A block diagram which illustrates these processes is shown in Figure 1, and the various components and processes are described below.

1. Sources of Coolant Radioactivity

The radioactivity content of the coolant arises from several sources as may be noted in Figure 1. For example, the fission product inventory in the core may be introduced through diffusion or recoil into the fuel pin bond. From the bond it may, if volatile, be released into the plenum of the individual fuel pin, be deposited or plated on the internal surface of the cladding, or be released through clad failure or deliberate venting into the coolant. The venting process releases only the noble gases, but subsequent decay into particulate daughters may occur during transit of the noble gases through the upper sodium plenum, as for example in the case of the short-lived xenon-cesium chain. Other processes also contribute activity from relatively nonvolatile fission products into the bond. These processes include knock-on collisions, diffusion, and grain boundary or pore diffusion. Normally these fission products are contained within the fuel element cladding unless a defect develops.* The presence

*The release of fission products from fuel to coolant is outside the scope of this project. These categories are included only because this information may be required in a more sophisticated analysis of the source term in the coolant.

of fuel contamination on fuel element surfaces or in the coolant provides a mechanism by which a broad spectrum of fission products may be introduced into the coolant.

The corrosion of the steel surfaces in an LMFBR will result in the activation of several elements which are constituents of the steel. Some of these, such as the longer-lived isotopes, are also significant in determining the after-shutdown radiation levels and are thus appropriate contributors to the coolant source term.

The presence of various impurities in sodium will result in their activation and contribution to the overall system radioactivity. One example is lithium which, through the (n, α) reaction, can produce ^3H in the coolant. Another source of ^3H is ternary fission. In this case the ^3H , following its release from the fuel, may diffuse directly through the cladding into the coolant stream.

2. Depletion Mechanisms for Coolant Radioactivity

There are several mechanisms (described below) within an LMFBR system which tend to limit the buildup of fission products in the sodium.

a. Radioactive Decay

Radioactive decay is the most obvious depletion mechanism. This process generally limits the isotopes of importance to those having a half life comparable to, or greater than, that of ^{24}Na .

b. Vaporization and Release from Coolant

A mechanism by which the more volatile fission product elements or compounds are removed from the coolant is by means of escape or evaporation into the cover gas. The presence of these radioisotopes in the cover gas may lead to subsequent difficulties in cover gas storage; effluent disposal; or personnel exposure, in the event of top shield leakage. Some of the program effort has therefore been directed towards developing a quantitative method for estimating the extent to which various isotopes will transfer from the sodium coolant to the cover gas. The source term study correlates these data.

c. Deposition and Plating

This process refers to the removal of coolant activities by means of sorption mechanisms occurring at the various primary system surfaces. This type of removal is of particular importance in determining post-shutdown radiation levels throughout the reactor system. Since it would also account for a proportionate depletion of radioactivity in the coolant, an accurate study of this process is necessary.

d. Trapping

Various types of impurity traps are used to remove coolant activities. Experience in the effectiveness with which coolant activities have been removed by cold traps or other system trapping techniques has varied widely, and, early in the project, it was considered of importance to study this characteristic for various fission products, not only to understand the mechanisms involved in present trapping technology but also to obtain fundamental information upon which the design of future trapping systems could be developed.

C. RELATIVE IMPORTANCE OF FISSION PRODUCT ISOTOPES

There are three main ways in which fission product contamination can affect the design, operation, and maintenance of an LMFBR. First, the release of volatile or gaseous fission products into the main primary coolant volume may ultimately result in the release of these fission products into the cover gas region over the reactor core. As a consequence, radioactivity in the contaminated cover gas may be released into the reactor high bay or other operating spaces if any large leaks or malfunctions of the gas seals develop. This occurrence has been observed in Dounreay Fast Reactor (DFR), Fermi, and other sodium cooled reactor systems. With regard to design characteristics, the requirement for disposal and/or clean up of the contaminated cover gas would obviously require more complicated designs and operational or maintenance procedures.

The second way in which fission product contamination of the coolant may affect safety or design considerations arises from the observed tendency of fission products to deposit or to plate out at various locations in the reactor system, or to be trapped in coolant purification devices such as cold traps, etc.

Finally, the coolant inventory itself is an important consideration in the design and shielding of piping and external components, in the specification of reactor shutdown and coolant removal procedures, and in accident hazard analyses. Two of these effects have been qualitatively evaluated in terms of indices to determine the relative significance of the various isotopes, and the evaluations are presented below.

1. Radiological Hazards Index

A convenient measure of the radiological hazard imposed by fission product contamination of the cover gas is the Maximum Permissible Concentration (MPC) criteria specified by the Atomic Energy Commission.⁽⁸⁾ In effect, these suggested MPCs, which relate to physiological effects, are already used throughout the industry to assess the relative hazard of various fission products with regard to personnel ingestion or inhalation.

Thus it was assumed that the relative importance, Radiological Hazard Index (RHI), evaluated for each nuclide should include an expression inversely proportional to the MPC. Other factors which are directly proportional to the Index include the inventory or the amount of the nuclide present, the release fraction from the fuel, and an expression of volatility relating to the probable concentration of the particular isotope in the cover gas. Normalized to Kr⁸⁵, the Radiological Hazard Index is given by:

$$RHI(t,i) = \frac{(C_i)_i}{(C_i)_{Kr^{85}}} \times \frac{(MPC_a)_{Kr^{85}}}{(MPC_a)_i} \times \frac{f_i}{f_{Kr^{85}}} \times \frac{v_i}{v_{Kr^{85}}}$$

where

RHI(t,i) = Radiological Hazard Index of the ith isotope after decay time t

C_i = radioactive inventory of the isotope in the fuel

MPC_a = Maximum Permissible Concentration of the isotope in air

f = release fraction of the isotope from the fuel to the sodium

v = volatility of the isotope (or fugacity)

The subscripts i and Kr⁸⁵ refer to the ith isotope and Kr⁸⁵ respectively.

The inventory of each isotope in the fuel was calculated in the usual way, using the recent Los Alamos plutonium fast fission yields. An operating period of one year, and a reactor thermal power of 2120 Mwt were also assumed to provide an initial estimate of the RHI. The choice of Kr^{85} as a normalization point was prompted by its long half-life, high release fraction from the fuel, and its volatility, which implies a high release fraction to the cover gas. For lack of additional information at this time, the release fractions of all isotopes were assumed equal to Kr^{85} , as were the fugacity expressions. Thus: $f_i = f_{\text{Kr}^{85}}$, $v_i = v_{\text{Kr}^{85}}$, and

$$\text{RHI}(t,i) = \frac{(C_i)_i}{(C_i)_{\text{Kr}^{85}}} \times \frac{(\text{MPC}_a)_{\text{Kr}^{85}}}{(\text{MPC}_a)_i}$$

These results are shown in Figure 2 as a function of time after shutdown.

2. After-Shutdown Radiation Aspects (The Radiation Index)

A second evaluation was performed to determine those fission product isotopes whose inventory, half-life, and gamma energy provide contributions to the after-shutdown radiation levels for maintenance or repair of the LMFBR system. Again, no convenient quantitative basis is presently available. For example, although it seems obvious that the hazard is proportional to the gamma energy, the effect of the gamma energy upon dose rate is a complex function of geometry, material, energy absorption characteristics, and thickness of piping and gamma ray shielding material. Therefore to minimize subjective aspects of the evaluation, dose rates were based upon an assumed unshielded point source. Using the above assumption, a dose rate was calculated for each isotope considered, based on the inventory of the isotope per unit volume of sodium. The resulting values were then normalized by dividing by the corresponding ^{22}Na dose rate. This isotope was selected as a normalization base because of its long half-life and because it will obviously remain with the sodium regardless of the volatility or deposition of fission products.

Figure 2. Radiological Hazard Index vs Time After Shutdown

Thus:

$$RI(t,i) = \frac{D_i}{D_{Na^{22}}} = \frac{(C_i)_i \times E_i}{(C_i)_{Na^{22}} \times E_{Na^{22}}} \quad \dots(3)$$

where

$RI(t,i)$ = radiation index for the i^{th} isotope at time t

$D_i, D_{Na^{22}}$ = dose rate at a distance of 1-ft from 1-cm³ of sodium containing isotope i and Na^{22} , respectively

$(C_i)_i, (C_i)_{Na^{22}}$ = inventory of i^{th} and Na^{22} in coolant respectively (curies)

$E_i, E_{Na^{22}}$ = energy of equivalent photons emitted from i^{th} isotope and Na^{22} respectively

v = coolant volume (cm³).

These results are shown in Figure 3 which presents the RI for each isotope as a function of the time after shutdown. Due to ^{24}Na radioactivity levels, most LMFBFR operating plans anticipate a waiting period of approximately two weeks following reactor shutdown before maintenance can be accomplished on the primary system. This initial period is indicated in the figure.

3. Relative Concentration of Fission Products and Corrosion-Activation Isotopes

Calculations have also been performed to estimate the level of radioactivity introduced into the coolant as a result of structural component activation and subsequent corrosion into the coolant stream. A preliminary estimate of these contributions indicates that, with an assumed average corrosion rate of 2 mils per year, the fuel element cladding will constitute the most significant source of activation-corrosion activity. With this assumed corrosion rate, the loss of cladding is $W = 4.6 \times 10^{-2}$ g/sec; and the incremental loss of cladding during a short time interval, dt , is Wdt . The activity produced in this increment of cladding up to the time, t , of its entrance into the coolant stream is:*

*Assuming steady power operation

Figure 3. Radiation Index vs Time After Shutdown

$$d\bar{A}_i = \phi \Sigma_i \frac{W dt}{\rho} \left[1 - e^{-\lambda_i t} \right] \quad \dots (4)$$

where

$\bar{A}_i$ = activity of the i^{th} isotope (sec^{-1})

ϕ = average neutron flux in the cladding ($\text{cm}^{-2} \text{sec}^{-1}$)

Σ_i = effective macroscopic cross section for the reaction (cm^{-1})

W = corrosion rate (g/sec)

ρ = cladding density (g/cm^3)

The decay of the initial activity in the increment of cladding (assuming it escapes the core and is no longer irradiated) gives an activity at a later time T , measured from startup of:

$$d\bar{A}_i = \phi \Sigma_i \frac{W dt}{\rho} \left[1 - e^{-\lambda_i t} \right] e^{-\lambda_i (T-t)} \quad \dots (5)$$

The total activity at time T , of all the cladding removed by corrosion up to that time is:

$$\int_0^T d\bar{A}_i = \int_0^T \phi \Sigma_i \frac{W}{\rho} \left(1 - e^{-\lambda_i t} \right) e^{-\lambda_i (T-t)} dt \quad \dots (6)$$

which when integrated yields:

$$\bar{A}_i = \phi \Sigma_i \frac{W}{\rho} \left[\frac{1}{\lambda_i} - \left(\frac{1}{\lambda_i} + T \right) e^{-\lambda_i T} \right] \quad \dots (7)$$

In performing the above calculations, the resonance integral for each isotope was evaluated over the flux spectrum of a typical LMFBR. The activity contribution from each of the significant stainless steel components with sufficient half-life and energy to constitute an appreciable after-shutdown radiation

Figure 4. Activity Concentrations for Corrosion Products, Fission Products and ^{22}Na vs Reactor Operating Time

contribution was then obtained, and results are shown in Figure 4. This figure also shows the growth of ^{22}Na activity concentration (assuming fresh sodium) and the concentration of some of the more important fission products, assuming one year of reactor operation, 10^{-5} release fraction at the end of one year and no decay or depletion.

Figure 4 indicates that, for the assumed corrosion rate of 2 mils/year (which is probably high) and the assumed fission product release fraction, the activity of corrosion products in the coolant, if not depleted by some mechanism, will predominate over all of the other activities. Moreover, since the corrosion product activity is directly proportional to the corrosion rate, as can be seen from the above equation, Figure 4 shows that, even if the corrosion rate were one-tenth the assumed value, the corrosion product activity, if not depleted, would still be dominant. Evidence obtained from post-operational examination of the S8ER, however, indicates that most all of the activated corrosion products are, in fact, depleted from the coolant stream by deposition on primary system surfaces.⁽⁵⁾ Indeed, a similar finding was also made relative to the nonvolatile fission products — thereby lending support to the basic depletion factor concept.

In connection with the corrosion product activity, it is interesting to note that after about 2-1/2 years the ^{60}Co activity becomes dominant. This is interesting because Co is only an impurity in stainless steels, and this activity arises from an assumed Co impurity concentration of 0.05%.

Two other activities which may arise from impurities are ^{134}Cs and ^{86}Rb . These were found in the S8ER coolant and were attributed to activation of stable Cs and Rb impurities present in the NaK coolant.

The ^{22}Na activity is also of interest since it rapidly builds up to significant concentrations and cannot be selectively removed from the coolant. Further, although there are some uncertainties involved in calculating the ^{22}Na activity due to uncertainties in cross sections and fluxes, these uncertainties are less than those associated with prospective corrosion rates, and clearly less than the speculative uncertainties associated with fission product release fractions. Accordingly, it is apparent that, after extended operation, ^{22}Na will be one of

the most important activities in the coolant stream. Further, this activity will not be substantially reduced without replacing a major fraction of the primary system sodium.

The greatest uncertainty is associated with the calculated fission product concentrations. This arises because of the uncertainties associated with the extent of fuel cladding failure and the limited understanding of the mechanisms of fission product migration out of the fuel. Throughout these studies we have arbitrarily assumed that the cladding simultaneously fails on 1% of the fuel pins and that 0.1% of the fission product inventory existing at that time escapes from the fuel. Both of these values, however, are highly arbitrary, as is the assumed instantaneous release. Further, no accounting is made for depletion of the fission product concentrations by various mechanisms currently under study. The result is that the above estimates of fission product activity are highly speculative, are based on a rigid release model which may be very unrealistic, and are incomplete in that removal mechanisms are not considered. The development of the Source Term Model (Section II-E below) is undertaken in an effort to improve realism, flexibility, and accuracy.

D. FISSION PRODUCT GROUPING

Definition of the behavior of fission products in sodium is necessary in order to evaluate the radiation hazard associated with plant operation and maintenance. By successfully grouping fission products as to their chemical behavior, the analytical effort required for the prediction of radiation levels is reduced in proportion to the number of chemical species and the number of analogue groupings.

As shown in Figure 1, fission products may ultimately remain in solution in the sodium, vaporize into the various gas spaces, or deposit on system walls. In order to establish an initial analogue grouping, the following potentialities were considered:

- 1) Do the isotopes form oxides that are stable relative to sodium oxide?
- 2) Do the isotopes form intermetallic compounds with the major constituents of Type 304 stainless steel?

3) Are the isotopes volatile?

4) To what extent are the isotopes soluble in sodium?

Table 2 presents data on solubility in sodium and vapor pressure for the elemental forms of various fission products, fuel materials and major constituents of Type 304 stainless steel (iron, nickel and chromium). The Free Energies of Formation of the most stable oxide and carbide for the same elements are also reported. Prediction of fission product behavior based on carbide data has not been made because of the uncertainty as to the chemical nature of carbon in sodium. Table 3 presents information on the ability of the various fission products to form binary compounds with iron, nickel and chromium.

Based on the above considerations and data, fission products have been divided into six analogue groups (Table 4).

1. Cesium

Volatilization should be the only removal mechanism for cesium. The most stable reported oxide of cesium is less stable than sodium oxide; cesium is miscible with sodium; cesium is not reported to form intermetallic compounds with iron, nickel or chromium; and elemental cesium has a high vapor pressure. However, cesium has been experimentally studied relative to its deposition tendencies (the only one so studied), and it has been shown that cesium can be cold-trapped from sodium. The need for experimental confirmation of all analogues is therefore clear.

2. Barium and Strontium

It is expected that these isotopes will deposit as oxides or be mechanically separated (filtered or centrifuged), assuming a low solubility for the oxides. Barium and strontium form stable oxides, have suitably low vapor pressures, and are not reported to form intermetallic compounds with iron, nickel or chromium.

3. Molybdenum, Ruthenium, Antimony and Silver

It is expected that these fission products will deposit on system walls, e. g., piping, vessels, hot and cold traps. They do not form stable oxides, have low

TABLE 2
SOLUBILITY OF METAL ELEMENTS IN SODIUM AND FREE ENERGY OF
FORMATION OF OXIDES AND CARBIDES AT 800°K

	Elemental Solubility (ppm, unless otherwise stated)*	Elemental Vapor Pressure (mm Hg)†	Oxide	Free Energy of Formation§ (kcal/g atom O)	Carbide	Free Energy of Formation§ (kcal/g atom C)
Ag	25 w/o	Low	Ag ₂ O	+5	-	+
Ru	(< 10)	Low	RuO ₂	-12	-	(0)
Te	35 w/o	1.4	TeO ₂	-21	-	-
Sb	30 w/o	8 x 10 ⁻³	SbO ₂	-35	-	+
Ni	< 1	Low	NiO	-40	NiC ₃	(0)
Co	< 1	Low	CoO	-43	CoC ₃	(0)
Cs	High	140	Cs ₂ O	-45	Cs ₂ C ₂	+
H	High	High	H ₂ O	-48	-	-
Mo	(< 1)	Low	MoO ₂	-49	MoC ₂	(0)
Fe	5	Low	Fe ₃ O ₄	-51	Fe ₃ C	+2
Cr	(< 5)	Low	Cr ₂ O ₃	-74	Cr ₄ C	-18
Nb	(< 10)	Low	NbO	-80	Nb ₂ C	-39
Na	-	7	Na ₂ O	-82**	Na ₂ C ₂	(-1)
Zr	< 1	Low	ZrO ₂	-112	ZrC	-45
Pu	?	Low	Pu ₂ O ₃	-113	PuC	-10
U	(10 to 100)	Low	UO ₂	-113	UC	-23
Ba	3 w/o	7 x 10 ⁻⁴	BaO	-115	BaC ₂	(-4)
Y	(10 to 100)	Low	Y ₂ O ₃	-120	YC	(-10)
Sr	(3 w/o)	8 x 10 ⁻³	SrO	-122	SrC	(-5)
Ce	(10 to 100)	Low	Ce ₂ O ₃	-124	CeC	(-20)
La	(10 to 100)	Low	La ₂ O ₃	-124	LaC	(-20)
Nd	(10 to 100)	Low	Nd ₂ O ₃	-124	NdC	(-20)

*References 10 and 11

†Reference 12

§References 13, 14, 15, 16, and 17

**At 5 ppm oxygen in sodium. Free energy of formation in oxygen in saturated sodium is -73 kcal/g atom O.

NOTE: Values in parentheses are estimated.

TABLE 3
REPORTED EXISTENCE OF BINARY COMPOUNDS OF
FISSION PRODUCTS AND MAJOR CONSTITUENTS
OF TYPE 304 STAINLESS STEEL (18, 19)

	Iron	Nickel	Chromium
Ag	(soluble)	Yes	Yes
Ba	Probably No	N. D.†	N. D.
Ce	Yes	Yes	No
Cs	No	N. D.	N. D.
I	Yes*	Yes*	Yes*
La	Yes	Yes	Probably No
Mo	Yes	Yes	Yes
Nb	Yes	Yes	Yes
Nd	Yes	Yes	Probably No
Ru	(soluble)	Yes	Yes
Sb	Yes	Yes	Yes
Sr	No	N. D.	N. D.
Te	Yes*	Yes*	Yes*
Zr	Yes	Yes	Yes

*Less stable than the corresponding sodium compound.

†No data.

TABLE 4
ANALOGUE GROUPING AND EXPECTED FATE OF
VARIOUS FISSION PRODUCTS

Analogue Grouping	Expected Fate in Stainless Steel System
Cs	Volatilization or deposition as complex oxide
Ba, Sr	Deposition as oxide
Mo, Ru, Sb, Ag*	Diffusion into stainless steel
Ce, La, Nd, Y, Zr	Deposition as oxide or diffusion into stainless steel
Nb	Deposition as oxide or diffusion into stainless steel
H ³ , I, Te	Vaporization or diffusion into stainless steel

*May not be a successful analogue grouping.

vapor pressure, and form intermetallic compounds with stainless steel. Isotopic dilution may be most successful with this analogue group in reducing radiation levels in operating equipment.

4. Rare Earths, Yttrium and Zirconium

These fission products may be removed by deposition on the system walls or by mechanical separation (filtering or centrifuging), assuming the oxides have low solubilities in sodium. They form stable oxides, intermetallic compounds with iron, nickel and chromium, and have low elemental vapor pressures.

5. Niobium

The stability of niobium oxide relative to sodium oxide depends upon the oxygen concentration. In regions where sodium is nearly saturated in oxide (such as the cold trap), niobium oxide will probably form, and niobium may therefore be removed by deposition as oxide or diffusion into stainless steel. In regions of low oxygen concentration (oxide-gettering hot traps), metallic niobium will probably be deposited on the system walls.

6. Tritium, Iodine and Tellurium

These fission products have high solubility in sodium and, at least at low concentrations, probably exist as the corresponding sodium compound (experimentally verified for iodine). These compounds have moderate vapor pressures, so vaporization should be one of the active removal processes. Deposition on the system walls should also occur.

E. SOURCE MODEL FORMULATION

A generalized source model is currently under development. It is the object of this model to provide a quantitative, flexible accounting of all of the important processes which affect the radioactivity of the sodium coolant of an LMFBR, the radioactivity of the cover gas, and the contamination-induced, residual radiation levels of equipment for which contact maintenance may be required. Many of the constants and functional relationships for some of the processes in the model remain to be determined. However, such a model will be of considerable value in assessing the relative importance of the various processes which are known and in establishing limits of importance for the processes under study.

In general, a model is needed which will account for the processes in Figure 1, which significantly affect the distribution of radioisotopes. Basically, this involves quantitative treatment of radioisotope production processes, and of transport and removal processes. With respect to the coolant, a general equation of the following form is being used as a starting point,

$$\dot{N}_i = \sum_j R_{i,j} - N_i \sum_k \alpha_{i,k} \quad \dots(8)$$

where

N_i = number of atoms of isotope i in coolant

$R_{i,j}$ = rate of introduction of isotope i into coolant by process j

$\alpha_{i,k}$ = fraction of i removed from coolant per unit time by process k .

The production term $\sum_j R_{i,j}$ includes all important processes by which radioactivity is introduced into the coolant, including: fission product leakage from failed elements; absorption of gaseous fission product leakage from vented elements; activation of core materials and subsequent corrosion; activation of the coolant itself, and of coolant impurities; fuel contamination of the coolant stream; and fission products resulting from fission of the fuel in the coolant stream.

The removal term $N_i \sum_k \alpha_{i,k}$ implies that all removal processes are concentration dependent. While this is true for radioactive decay (where $\alpha_{i,1} = \lambda_i$, the radioactive decay constant) and for diffusion removal processes, and appears consistent with the "depletion factor" concept discussed in Section V, it may or may not be correct for a number of other processes, such as adsorption, surface chemical reactions, loss to cover gas, and various filtering and trapping processes. Even where a concentration-dependent process is appropriate, there is the problem of evaluating the various $\alpha_{i,k}$. This is because, with the exception of the radioactive decay constant, most of the $\alpha_{i,k}$ are likely to be functions of temperature, surface area, material composition, surface condition, etc. However, as more data become available it will be possible to make quantitative estimates of the $\alpha_{i,k}$ for various systems and add additional functional relationships to the removal term.

In order to obtain the desired analytical tool it is planned to apply the above generalized equation, including the constants derived for the various depletion processes, to a typical LMFBR primary system as depicted in Figure 5. The resulting equations will then be programmed for digital computer solution so that the effect of a variety of parameter variations can be quickly and economically investigated.

Figure 5. Primary and Auxiliary Loops for Assumed "Typical" LMFBR

Inspection of Figure 5 shows the principal components and systems that will be considered in the model. These are: (1) the main primary coolant loop, (2) the auxiliary by-pass loops, and (3) the radioactive gas handling system. As indicated above, the value of α or the controlling functional relationship will have to be determined or estimated for each of the components in each of these systems, and for each of the important isotopic species. These can then be inserted in the source model code to obtain estimates of system behavior. It is anticipated, however, that even before approximate values are available for all components and processes, the computer program will still provide valuable program guidance as to the general behavior of the system and the relative importance of various processes.

Within the framework of the above preliminary model some initial study has been directed toward determining the effect of the manner of release of the fission product inventory to the coolant. This study has assumed that the fission products enter the coolant by way of cladding defects and are removed by radioactive decay and by a concentration-dependent plating process wherein the fission products are deposited on the steel surfaces of the primary system. For this study we have let $\alpha_{i,1} = \lambda_i$, the radioactive decay constant, and $\alpha_{i,2} = \alpha_i$, the depletion constant. For simplicity we have also dropped the subscript "i."

We consider three means by which fission products can be released to the coolant:

- Case 1. After a period of operation of time T , there is, due to an over-power situation, an instantaneous failure of the cladding in a fraction, f_1 , of the fuel pins and a fraction, f_2 , of the inventory of the isotope of interest in the failed pins is instantly released to the coolant stream. The reactor is then shut down, but circulation of the coolant is continued.
- Case 2. The reactor starts operation with cladding failures already present in a fraction, f_1 , of the fuel pins, and a fraction, f_2 , of the isotope of interest in the failed pins is instantly released to the coolant as soon as it is produced. The reactor continues to operate until time, T , when it is shut down, but circulation of the coolant is continued.
- Case 3. The reactor commences operation with no cladding failures, but failures occur progressively until at time, T , cladding failures have occurred in a fraction, f_1 , of the fuel pins. With the onset of a cladding failure, a fraction, f_2 , of the isotope of interest in the failed fuel pin is instantly released to the coolant stream and a fraction, f_2 , of all of the isotope of interest produced thereafter in the failed pins is released to the coolant as it is produced. The reactor is shut down at time, T , but circulation of the coolant is continued.

Of the three mechanisms described above, the first two have been analyzed, and the results are presented below.

In the absence of a chemical depletion process and for a constant rate of production, R , the instantaneous rate of production of the isotope of interest, $\dot{N}$ (atoms/sec), is given by the familiar expression,

$$\dot{N} = R - \lambda N \quad \dots(9)$$

where N is the number of atoms of the isotope of interest. After a period of time, T , $N(T)$ is given by,

$$N(T) = N(T)_{w/o} = \frac{R}{\lambda}(1 - e^{-\lambda T}) \quad \dots(10)$$

where the subscript w/o denotes the absence of a depletion mechanism. Since there is no depletion mechanism, Equation 10 corresponds to the basic inventory of the isotope of interest that would be subject to release to the coolant should simultaneous cladding failures occur at time, T . This, then, corresponds to the situation in Case 1, above. The number of atoms of the subject isotope released to the coolant when the cladding fails at time T , $N_{Na,w/o}(T)$, is then given by,

$$N_{Na,w/o}(T) = \frac{f_1 f_2 R}{\lambda}(1 - e^{-\lambda T}) \equiv N_1(T) \quad \dots(11)$$

where $N_1(T)$ is introduced to simplify subsequent discussion.

For the mechanism described under Case 2, the instantaneous rate of production of the subject isotope in the coolant for the same basic production rate is given by,

$$\dot{N}_2 = f_1 f_2 R - (\lambda + \alpha)N_2 \quad \dots(12)$$

where α is the depletion constant. Thus

$$N_2(T) = \frac{f_1 f_2 R}{(\lambda + \alpha)} \left[1 - e^{-(\lambda + \alpha)T} \right] \quad \dots(13)$$

If $\lambda T > 5$, so that the exponential terms in Equations 11 and 13 are negligible, the ratio of the number of atoms of the subject isotope in the coolant at time T for Cases 1 and 2 is given by $N_1(T)/N_2(T)$,

$$N_1(T)/N_2(T) = \frac{\lambda + \alpha}{\lambda} \quad \dots(14)$$

After time, T , the coolant inventories denoted by $N_1(T)$ and $N_2(T)$ will be reduced according to $e^{-(\lambda+\alpha)t}$. The results for $\alpha = \lambda$ and $\alpha = 10\lambda$ are shown in Figure 6. From these results it is clear that, when there is a depletion mechanism operative in the primary system, it is necessary to specify not only the release fractions but also the temporal character of the release.

Building upon the preceding development it is also of interest to examine the plated inventory, i. e. the number of atoms of the subject isotope adhering to the primary system surfaces. This inventory will be denoted by N_{1w} for the Case 1 release mechanism and N_{2w} for the Case 2 release mechanism.

For Case 1, $N_{1w} = 0$ until cladding failure occurs at a time, T . After this

$$\dot{N}_{1w} = \alpha N_1(t) - \lambda N_{1w} \quad \dots(15)$$

where $N_1(t) = N_1(T)e^{-(\lambda+\alpha)t}$ and t is measured from time, T . Equation 15 has the solution,

$$N_{1w} = N_1(T)[e^{-\lambda t} - e^{-(\lambda+\alpha)t}] \quad \dots(16)$$

and N_{1w} reaches a maximum of

$$N_{1w}(\text{max}) = N_1(T) \left(\frac{\lambda}{\lambda + \alpha} \right)^{\lambda/\alpha} \cdot \left(\frac{\alpha}{\lambda + \alpha} \right)$$

when

$$t = \frac{1}{\alpha} \log_e \left(\frac{\lambda + \alpha}{\lambda} \right).$$

$N_{1w}(t)/N_1(T)$ is plotted versus time in Figure 7 for $\lambda = 1 \times 10^{-2}$ where $\alpha = \lambda$ and $\alpha = 10\lambda$. It is to be noted that $N_1(T)$ is the coolant inventory of the subject isotope upon the occurrence of fuel failure. A value of $\lambda T > 5$ was also assumed.

6-10-68 UNCL

7702-4588

Figure 6. Relative Coolant Inventory of Hypothetical Isotope vs Time After Shutdown for Case 1 and 2 Release Mechanisms

6-10-68 UNCL

7702-4589

Figure 7. Relative Plated Inventory for a Hypothetical Isotope vs Time After Shutdown for Case 1 and 2 Mechanisms

For Case 2, isotope deposition on system surfaces commences at the beginning of reactor operations. Thus, for $t < T$, the instantaneous rate of change of the wall inventory is given by,

$$\dot{N}_{2w} = \alpha N_2(t) - \lambda N_{2w} \quad \dots(17)$$

where, $N_2(t)$ is given by Equation 13, with t replacing T . Equation 17 has the solution

$$N_{2w}(t) = f_1 f_2 R \left[\frac{\alpha}{\lambda(\lambda + \alpha)} - \frac{1}{\lambda} e^{-\lambda t} + \frac{1}{\lambda + \alpha} e^{-(\lambda + \alpha)t} \right] \quad \dots(18)$$

Again, if $\lambda T > 5$, the exponential terms become insignificant and,

$$N_{2w}(T) \approx \frac{\alpha f_1 f_2 R}{\lambda(\lambda + \alpha)} \quad \dots(19)$$

After the time, T , the instantaneous rate of change of N_{2w} is given by:

$$\dot{N}_{2w} = \alpha N_2(T) e^{-(\lambda + \alpha)t} - \lambda N_{2w} \quad \dots(20)$$

where $N_2(T)$ is a constant as indicated in Equation 13 and time is now measured from T .

Equation 20 has the solution,

$$N_{2w}(t) = N_2(T) \left[e^{-\lambda t} - e^{-(\lambda + \alpha)t} \right] + \frac{\alpha f_1 f_2 R}{\lambda(\lambda + \alpha)} e^{-\lambda t} \quad \dots(21)$$

The value of $N_{2w}(t)/N_1(T)$ for $t > T$ is also plotted in Figure 7. Inspection of the figure indicates that the time behavior of the release also has an important effect on the plated fission product activity, particularly during the first few weeks after shutdown, which may be a critical period if contact maintenance is desired.

The above relationships permit a more accurate assessment of coolant and deposited fission product activity levels as a function of time and therefore will be included in the Source Term Model. The relationships for the Case 3 release mechanism will also be added as soon as they are completed.

IV. EXPERIMENTAL STUDIES

A. CESIUM AND IODINE VAPORIZATION FROM SODIUM

The purpose of this section is to summarize experimental and analytical work performed as part of a fundamental study of the distributional behavior of fission products released to the sodium coolant of a fast reactor during normal operation. The information obtained is intended to permit evaluation of normal operating hazards and to support efforts to develop trapping or other fission product contamination control devices.

It was concluded from the analysis conducted for the initial source term studies that fission products released could correspond to concentration increments of the order of 10^{-6} to 10^{-9} mole fraction, and that of these cesium would be among the more abundant. It was on this basis, and because of its relative radiation index after prolonged shutdown, that cesium was selected for study. The selection of iodine for study was based on its radiological importance in the event of its release by leakage through the top shield.

It should be noted that, with the exception of the noble gases and rubidium and perhaps tellurium, cesium would be the only element likely to be sufficiently volatile that vaporization would be an important distributional mechanism. It is much more likely that, at the low concentrations likely to be found in the reactor coolant, various plating mechanisms and/or interactions with particulate matter or corrosion products would account for the ultimate fate of the isotopes. The primary objective of this task is to investigate such interactions, including the effect of oxygen.

A method of investigation well-suited to the study of cesium and iodine interactions is to observe changes in vapor equilibrium at given nominal concentrations as a function of the amount of additive or surface. For this method to be feasible, however, requires that the experimental procedure be sufficiently precise to detect changes that may be relatively small. It has therefore been necessary to select and perfect a technique having the required precision. The criterion in this respect was taken as 10% or less experimental uncertainty. It might be noted that a secondary, but scientifically valuable, result of this work was that development of data for vapor-liquid equilibria for solutes in sodium would permit deduction of solution thermodynamic properties.

Two experimental techniques were selected for use. The first was the transpiration method, which is well developed and quite suitable for the anticipated temperature range. The second was a vapor density method which was selected to serve as an alternate because of its potential simplicity. To ensure that the solutes would behave according to Henry's Law (in order to have a thermodynamically well-defined system) and to minimize extrapolation in later work, it was desirable to work at fairly low solute concentrations, i. e. mole fraction of the solute in the range from about 10^{-6} to 10^{-4} . Initial analysis, however, made it quickly apparent that tracer techniques would be necessary for this concentration regime. Finally, the range of coolant temperatures expected according to present LMFBR concepts is from about 650 to 900°K, and it was considered desirable to conduct experiments within this range, where possible.

1. Analytical Studies and Supplementary Literature Data

The vaporization behavior of these systems is conveniently described by the relative volatility, designated here by the symbol " K_d " and defined as the ratio of the mole-fraction of the solute in the vapor, Y_2 , to its mole-fraction in the liquid, X_2 , or $K_d = Y_2/X_2$. Experimentally this ratio may be determined using tracer techniques by determining the ratio of specific activity, A , in the vapor to that in the liquid, $K_d = Y_2/X_2 = A_g/A_l$. The derivation of an analytical expression for iodine (or more accurately, NaI, is given below.

Referring again to the definition for K_d ,

$$K_d = Y_2/X_2, \quad \dots(1)$$

we may note that at low solute concentrations,

$$Y_2 = \frac{P_{NaI}}{P_{NaI} + P_{Na} + P_{Na_2}} \cong \frac{P_{NaI}}{P_{Na}^o}, \quad \dots(2)$$

where

P_{Na}^o = the pressure of sodium vapor (containing about 3 to 7% dimer)
over essentially pure sodium liquid

P_{Na} = partial pressure of sodium monomer in the vapor

P_{Na_2} = partial pressure of sodium dimer in the vapor

P_{NaI} = partial pressure of sodium iodide in the vapor.

It should be noted that the iodine would be expected to be predominantly in the form of NaI in the vapor, with negligible dissociation or mixed halides. Expressing P_{NaI} as a function of concentration.

$$P_{NaI} = \gamma X_2 P_{NaI}^0, \quad \dots(3)$$

where

γ = activity coefficient

P_{NaI}^0 = vapor pressure of sodium iodide

and substituting in (1),

$$K_d = \frac{\gamma X_2 P_{NaI}^0}{X_2 P_{Na}^0} \cong \frac{\gamma P_{NaI}^0}{P_{Na}^0} \quad \dots(4)$$

Since the NaI concentration of interest in this work will be in the "Henry's Law" region, the activity coefficient will be a function of temperature only. In order to use literature data, it is convenient at this point to take logarithms:

$$\text{Log } K_d = \text{Log } \gamma + \text{Log } P_{NaI}^0 - \text{Log } P_{Na}^0 \quad \dots(5)$$

These quantities are given fairly accurately over moderate temperature ranges by the familiar linear expression,

$$\text{Log } P \text{ (or } \gamma) = A/T + B,$$

where

A and B = constants

T = absolute temperature.

Data for sodium have been taken from the compilation of Stull and Sinke⁽²⁰⁾ for the elements by calculating the total pressure due to sodium monomer and dimer at 700 and 900°K and determining (by the "two point" method) the expression,

$$\text{Log } P_{\text{Na}}^{\circ} = 5373/T + 4.65 \quad .$$

Bauer and Porter⁽²¹⁾ give data for the free energy of vaporization of alkali halides in the form of free energy functions and standard heats of vaporization at 298°K,

$$\Delta F_{\text{T}}^{\circ}(\text{vap}) = - \left(- \frac{\Delta F_{\text{t}}^{\circ} - \Delta H_{298}^{\circ}}{T} \right) T + \Delta H_{298}^{\circ} = -4.576 T \log P^{\circ} \quad \dots (6)$$

Free energy function differences do not change rapidly with temperature, so that the value for 800°K may be substituted, and, solving for $\text{Log } P^{\circ}$,

$$\text{Log } P_{\text{NaI}}^{\circ} = 10,500/T + 7.51 \quad . \quad \dots (7)$$

To evaluate the activity coefficient of sodium iodide, one may note that NaI is not very soluble in sodium below about 900°K. If it is assumed that the activity is equal to the fractional saturation,

$$\text{activity} = \frac{\text{concentration}}{\text{solubility}} = \gamma X_2 \quad \dots (8)$$

where concentration and solubility are expressed as mole-fractions (X_2 and X_2° respectively); then $\gamma = 1/X_2^{\circ}$. The solubility data of Bredig, Johnson, and Smith⁽²²⁾ for NaI in sodium for the temperatures below melting have been treated by least squares to give,

$$\text{Log } X_2^{\circ} = -9300/T + 8.19 = -\text{Log } (1/\gamma) \quad . \quad \dots (9)$$

Substituting for the logarithms on the right hand side of Equation 5 and simplifying, the relative volatility of NaI in sodium can be expressed as,

$$\text{Log } K_{\text{d}} = 4173/T - 5.33 \quad . \quad \dots (10)$$

The derivation of an expression for K_d for cesium in sodium is much the same as for NaI, the difference being in the manner in which the activity coefficient is evaluated. Thus,

$$K_d = \frac{Y_2}{X_2} \cong \frac{P_{Cs}}{P_{Na}^0 X_2} = \frac{\gamma P_{Cs}^0}{P_{Na}^0},$$

and

$$\text{Log } K_d = \text{Log } \gamma_{Cs} + \text{Log } P_{Cs}^0 - \text{Log } P_{Na}^0. \quad \dots(11)$$

As is true with other alkali metals, cesium vapor in equilibrium with liquid cesium contains appreciable dimer; however, at low concentrations in sodium, there would be very little dimer in comparison to the monomer, so that only the partial pressure of the monomer must be considered. Using the data for monatomic cesium from Stull and Sinke,⁽²⁰⁾ it is possible to obtain the expression,

$$\text{Log } P_{Cs}^0 = -3795/T + 3.900. \quad \dots(12)$$

The vapor pressure of sodium liquid is from Stull and Sinke.⁽²⁰⁾

The cesium-sodium system differs from the sodium iodide-sodium system in that cesium and sodium are essentially completely miscible above about 100°C. To evaluate the activity coefficient (which incorporates departures from ideal solution behavior), one may express the activity coefficient in terms of the excess free energy of mixing:

$$4.576T \text{ Log } \gamma = \Delta F^E = \Delta H_{\text{mix}}^E - T\Delta S^E. \quad \dots(13)$$

Yokokawa and Kleppa⁽²³⁾ have measured heats of mixing of cesium and sodium over the composition range by calorimetric means, at 111°C, and have stated that for cesium mole-fractions less than about 0.5, the excess entropy of mixing is zero (i.e., the solutions are "regular"). An extrapolation of their data to zero mole-fraction (infinite dilution) cesium gives for the partial molar heat of

mixing of cesium a value of approximately 1900 cal/mole. The activity coefficient for cesium should therefore be given by,

$$\text{Log } \gamma_{\text{Cs}} = 1900/4.576T = 415/T \quad \dots(14)$$

Combining the expressions for P_{Cs} , P_{Na}° , and γ as for NaI gives

$$\text{Log } K_d(\text{Cs}) = 1945/T - 0.668 \quad \dots(15)$$

In using the relationship for estimating the relative volatility of NaI in sodium, it should be remembered that the assumption of Henry's law behavior is only an approximation, the uncertainty of which is unknown. Further, in using the solubility data for NaI in sodium at low solubilities, caution should be exercised since a small systematic error in the measurements would considerably affect the temperature dependence. In the case of cesium, the chief uncertainty appears to be the extrapolation of the heats of mixing, which may lead to an uncertainty of about 20%, corresponding to an uncertainty of about 300 calories in the heat of mixing.

2. Experimental Methods

With the exception of a few trial experiments conducted with a very simple vapor density apparatus, all vaporization experiments were performed with what proved to be a simplified version of a transpiration apparatus. Some comments, however, are pertinent to the vapor density apparatus, which is illustrated in Figures 8 and 9. Two stainless steel chambers are connected by stainless steel fittings through a stainless steel diaphragm-type valve. Provisions also were made to evacuate the chambers. The lower, or liquid, chamber, of about 45 to 50 ml capacity, was normally charged with about 25 ml of sodium. The upper, or vapor, chamber was given a preliminary hydrogen treatment in order to remove or reduce error due to presence of oxidation products on the surfaces. Usually, the apparatus was first charged and assembled in a glove box, then was attached to the various lines. The furnace could be pulled down over the assembly after being brought to temperature.

6-29-67 UNC

7702-4042A

Figure 8. Schematic Diagram of Vapor Density Apparatus

6-13-68 UNCL

7702-4020

Figure 9. Vapor Density Apparatus Removed from Furnace

The transpiration apparatus used in both Cs-Na and Na-NaI studies is illustrated in Figures 10 and 11. In this work, one of the concerns was that the solutions be adequately stirred in order to maintain uniform concentration. Otherwise, concentration gradients would be established which would give rise to a diffusion-limiting effect on one or another of the solution components and thus invalidate the results. The bubbler tube design was therefore adopted in order to maintain good mixing.

The carrier gas was 99.999% argon. Volume measurements could be made by the combination of the calibrated 3 liter stainless steel ballast tank and calibrated pressure gauge, or by the gas collector burette; the results of both methods were in good agreement. A NaK bubbler was in the line to ensure carrier gas purity.

To ensure establishment of the equilibrium vapor concentration in the carrier gas, the usual procedure was followed of making a series of runs at various flow rates. These runs showed that saturation with sodium occurred at least between 20 and 160 ml/min. At higher flow rates (260 ml/min), saturation also occurred but mechanical entrainment became a problem. In practice, flow rates between about 20 to 100 ml/min were used.

a. Cesium - Sodium System

In this work, the transpiration apparatus of Figure 10 was operated with the furnace in a horizontal position. To agitate the liquid, the furnace was slightly rocked manually at frequent intervals.

To permit addition of a reasonably well-defined, but rather small, concentration of cesium (usually at mole fraction levels of 1.0×10^{-5}), a few grams of a "master batch" of cesium-sodium alloy in the atomic ratio of 1 to 10 was made up and stored in a plastic vial within an inert-atmosphere glove box. This allowed use of pieces sufficiently large to permit easy handling and weighing on a small semi-micro balance kept within the glove box. These small weighed samples, sufficient to dope about 175 to 200 grams of sodium solution, were transported in sealed polyethylene vials and irradiated in the Shield Test and Irradiation Reactor (STIR) to a level corresponding to about 1 Mwt-hr of reactor operation. The ^{24}Na was then allowed to decay over a period of several days before being brought back to the laboratory for incorporation in the main batch of sodium. After the activated alloy was added to

6-13-68 UNCL

7702-4044A

Figure 10. Schematic Diagram of Transpiration Apparatus

6-13-68 UNCL

Figure 11. Disassembled Bubbler Used in Transpiration Apparatus

7702-4021A

the sodium, a sample was removed for analysis and specific activity determination. Several transpiration cells were loaded with charges of about 23 gm each. About 5 mgm Na_2O_2 was added to provide an oxygen concentration of 125 ppm. Each cell was used for only a single run, to minimize concentration changes and to have well-defined specific activities of the liquid from run-to-run in a given series.

The runs were made in the temperature range 650 to 900°K to simulate the expected range of coolant reactor temperatures. Carrier gas volumes were selected to give approximately the same amount of condensed sodium in each run, i. e., 10 to 25 gm. The transported vapor was condensed in a water-cooled collector tube and the amount of sodium content was determined by a titrimetric or flame photometric method. The cesium activity was determined with a scintillation detector, pulse height analyzer system using the same geometry as for the liquid metal reference solution. Carrier gas volumes were measured and theoretical sodium transports calculated for comparison with measured quantities as a check for gross experimental errors. The agreement was usually fairly good.

b. Sodium Iodide - Sodium System

The furnace shown in Figure 10 was used in a vertical position in these runs. To minimize the effect of surface absorption and exchange, a small quantity of carrier iodine, of the order of 1 to 2 atomic ppm was added. Since sodium iodide is only slightly soluble in sodium, especially at the sampling temperatures that could be safely used in the glove box, the following procedure was used to provide a sample that would be typical of a reactor coolant. About 10 mc of ^{131}I , in the form of 1 ml aqueous sodium iodide, were washed into a small beaker using 0.5 mgm of sodium iodide carrier and sufficient de-ionized water to give about 25 ml total volume. One ml was pipetted into a 100-ml volumetric flask, together with a few mgm of sodium iodide, and was then diluted to the mark. Of the remaining iodide activity, 20 ml was then pipetted directly into the vapor chamber of the vapor density apparatus or into the bubbler part of the transpiration apparatus; a drop of 5% NaOH was added to prevent loss due to oxidation, and the solution was then evaporated to dryness. The chamber or bubbler was then transferred to the glove box,

a weighed amount of sodium metal was added, and the apparatus was then assembled. In this way a known concentration of iodine was added to the sodium (usually 1 atom-ppm), and an aqueous solution representing a known fraction of the liquid metal was obtained.

In performing the iodine transpiration runs, the furnace was first brought to temperature, the bubbler was inserted and, as soon as the sodium became melted, a very low flow-rate of gas was begun in order to prevent back-flow of sodium and resultant plugging. When the sodium reached the desired experimental temperature, the flow-rate was increased to 20 to 75 ml/min, until an estimated quantity of about 10 to 25 mgm of sodium had volatilized. The flow-rate was then reduced to the earlier value. The bubbler was then removed from the furnace and allowed to cool. When the system had cooled enough to handle, gas flow was stopped and the cell with its collector and lead-in tube was removed. Sodium transport was determined with standard HCl and the specific activity of the iodine counted and compared against the reference.

3. Experimental Results

a. Cesium - Sodium System

One of the difficulties encountered during much of the work with cesium was inconsistent liquid metal specific activity data. For example, on occasion the specific activity of the residue from a run might show a very substantially higher specific activity than found for the (apparent) initial specific activity. Part of the difficulty may have been due to inadequate mixing of the tracer batch; however one definite source of trouble was traced to segregation during the freezing with the resultant appreciable inhomogeneities existing in the cesium distribution activities. This effect was corrected (after Series D) by dissolving the entire sodium sample in butyl alcohol and taking an aliquot (instead of cutting and weighing a piece of the residue as before). When residues from a number of the Series E runs were corrected for cesium activity transport, the calculated initial activities agreed well (1 to 2%) with the measured initial activities.

Results of the Series D and E transpiration runs at nominal oxygen levels of 150 ppm and cesium mole-fractions of 1×10^{-5} and 8×10^{-5} are summarized in Table 5 and Figure 12 (Series E only). Much of the earlier work tends to cluster about the

TABLE 5
RELATIVE VOLATILITY OF CESIUM AT
LOW CONCENTRATION IN SODIUM

Run No.	Temperature (°K)	Calculated Na Carried (mgm)	Observed Na Carried (mgm)	Vapor ($\frac{\text{cpm}}{\text{gm Na}}$)	Liquid ($\frac{\text{cpm}}{\text{gm Na}}$) _{avg}	K _d
Series D						
D-2	898.5	50.4	34.3	6.18×10^7	8.38×10^5	74
D-4	822.5	17.6	14.4	8.26×10^7	8.58×10^5	96
D-5	892.7	46.1	40.9	3.84×10^7	8.50×10^5	45
D-6	836.0	25.0	22.4	7.33×10^7	8.47×10^5	87
D-7	769.0	29.2	30.0	9.6×10^7	8.21×10^5	117
D-8	698.0	30.7	32.6	13.6×10^7	7.87×10^5	137
Series E						
E-1	731.8	18.2	19.8	6.45×10^6	0.99×10^5	65
E-2	775.0	16.5	21.3	5.18×10^6	1.00×10^5	52
E-4	868.5	28.8	34.7	3.65×10^6	0.99×10^5	37
E-5	680.0		14.7	7.60×10^6	1.00×10^5	76
After loading sample 5, 500 ppm normal Cs was added to give $X = 8 \times 10^{-5}$, and another specific activity determination made, giving 0.92×10^5 cpm/gm						
E-6	736.5	18.9	28.0	6.16×10^6	0.98×10^5	63
E-7	773.5	15.1	18.4	5.85×10^6	1.00×10^5	59
E-9	868.5	30.1	31.8	3.82×10^6	0.99×10^5	39
E-10	684.0	16.3	19.1	9.1×10^6	0.99×10^5	93

Figure 12. Cs-Na System, Calculated and
Experimental Relative Volatility

experimental $\log K_d$ vs $1/T$ curve, but is not shown because of various difficulties, including those mentioned above. In the table, the column headings need little explanation. In computing the K_d values, the measured sodium transport was used in calculating vapor specific activity in order to reduce effects of gas measuring and temperature errors. In addition, it should be noted that, although rather small fractional sodium transports were used, i. e., about 0.1%, the resulting changes in cesium activity (or concentration) in the liquid were sometimes as high as 15 to 20%. Accordingly the average liquid metal specific activity was calculated, based on the initial specific activities of the batches and transports during the run.

The slopes of the curves obtained in the two series are in very good agreement. Least squares treatment of the data for Series D gave $\log K_d = +997/T + 0.750$, while similar treatment of the Series E data gave $\log K_d = +987/T + 0.490$. A second feature shown in Figure 12 is the excellent agreement between the two groups of runs in the E series, that is, the group with $X_{Cs} = 1 \times 10^{-5}$ and the group with $X_{Cs} = 8 \times 10^{-5}$. The latter fact is evidence that Henry's law is obeyed, as was expected. Apparently the experimental precision within a group or series is quite good, but this fact, and the vertical displacement between Series D and Series E, suggests some error in the determination of the liquid metal specific activities.

b. Sodium Iodide - Sodium System

The results of the transpiration series, designated here as the iodine Series F, are summarized in Table 6 and plotted in Figure 13. The temperatures listed in the first column of Table 6 are average temperatures estimated from visual inspection of temperature-time data and are subject to uncertainty of 6 to 7°C. The difference between the observed and calculated sodium transports is in part the result of the temperature and gas volume uncertainties, and in part the result of extraneous transport due to diffusion during the heating and cooling phases of the experiments. Such differences do not affect the results since the effects would apply to both sodium and iodide and would thus cancel each other. Because of the short half-life of I-131 as compared to Cs-134, it was necessary to count the reference solution at the same time as the experimental transports, and these counts are shown in the table. The K_d values were

TABLE 6
RELATIVE VOLATILITY OF SODIUM IODIDE IN SODIUM
(SERIES F)

Run No.	Average Temperature (°K)	Sodium Transport (mgm)			Measured Condensate (cpm)	cpm/47.5 mgm Liquid Sodium	K_d
		Calculated	by Titration	by Flame Photometer			
F-2	754	9.1	16.2	16	82,720	229,200	1.06
F-3	863	31.8	41.0	44	156,800	229,200	0.79
F-4	695	5.9	10.4	11	123,400	224,600	2.50

Figure 13. NaI-Na System, Calculated and
Experimental Relative Volatility

AI-AEC-12687

calculated using the experimental values of sodium transports, for the same reasons as for the cesium work.

A "best line" through the three available points gives for K_d :

$$\text{Log } K_d = 1840/T - 2.34 \quad \dots(15)$$

4. Evaluation of Results

Experience to date indicates that the vapor density method is not well adapted for the investigations involving components which are very active chemically at the low temperatures (and therefore low vapor densities) of interest in this work. The effects of extraneous and side reactions are difficult to control and are of sufficient magnitude to obscure the desired effects. In contrast, the transpiration method appears to be much less inherently susceptible to such errors, and the experimental procedures are capable of the desired precision although there is an unidentified systematic error in the cesium determination. (This matter is discussed below.) The transpiration data for the iodine system are too few to permit drawing estimates of precision, although such data as are available seem to be thermodynamically credible.

In examining the data for the two systems, it is convenient to compare the experimentally derived activity coefficients with those estimated in Section IV-A-1. These values are given for 605 and for 900°K in Table 7. The experimental values were calculated from the defining Equation 4 using the experimental values chosen from the curves of Figures 12 and 13. The calculated activity coefficients were obtained from Equations 9 and 12. The data for Cs

TABLE 7
ACTIVITY COEFFICIENTS

Temperature (°K)	Cs-Na		NaI-Na	
	Calculated	Experimental	Calculated	Experimental
650	4.4	1.9	1.2×10^6	3×10^5
900	2.9	3.5	140	420

show a discrepancy in that the activity coefficients would ordinarily tend toward unity with increase in temperature, as is the case for the NaI. One possible explanation for the trend may be due to the level of oxygen (e. g. 125 ppm) in the sodium for the cesium runs. That there does seem to be some kind of interaction between cesium and oxygen in sodium has been observed in cold trapping studies which are discussed in Section IV-B-1. Additional data on Cs vaporization obtained with sodium having low oxygen levels (e. g. <5 ppm) could confirm this effect.

The data for NaI, although exhibiting somewhat lower temperature dependence than predicted, seem to be quite reasonable and in fair agreement with work of Saroul,⁽²⁴⁾ Clough,⁽²⁵⁾ and Koontz.⁽²⁶⁾ The data for the NaI-Na system is not in agreement with the experimental and theoretical work of Castleman.⁽²⁷⁾ As shown in Figure 13 the BNL curve (for the region 940 to 1200°K) has a considerably different temperature dependence for K_d and, although agreement with the present work is obtained at 1000°K, the departure between the two investigations is considerable when the BNL curve is extrapolated to 700°K. The BNL experimental equilibrium vaporization data for the NaI-Na system were obtained with a carrier-free NaI-Na system, whereas in the AI work 6 ppm of NaI carrier was present. The difference between the results of the two investigations may be due to the fact that the present study was conducted at concentrations approaching the NaI solubility in sodium at the lower temperature.⁽²⁸⁾ However, at the lowest temperature studied, 700°K, the reported solubility of NaI in Na is $\sim 1.2 \times 10^{-5}$ mole fraction, compared with the 1×10^{-6} mole fraction of NaI employed in the present studies. Thus it is not apparent that solubility limitations have affected the results, unless Henry's law does not hold at this concentration (~ 0.1 saturation), so that the activity coefficient is not concentration-independent as assumed.

B. DEPOSITION AND TRAPPING STUDIES

1. Cold Trapping of Cesium in Sodium

Traps are used in liquid metal systems to control corrosion by oxygen and embrittlement of stainless steel by carbon. However, cold-trapping also appears to offer possibilities for control of fission product contamination. Hart⁽¹⁾

and Hansen⁽²⁹⁾ examined parts of the SRE after shutdown and found a high degree of preferential segregation of fission products in the cold trap. (See Section V-A for amplification.)

The apparent scavenging of cesium in the SRE cold trap cannot be readily explained by thermodynamic equilibrium considerations. In the analogues Na - K - O (or NaK - O), the sodium and potassium may be assumed to be at comparable activities of the order of, but less than, unity. Since sodium oxide is more stable than is potassium oxide, the sodium oxide might be expected to precipitate first as the oxygen concentration is increased; or if the temperature is lowered, the solution would first become saturated with respect to Na₂O. This behavior has been observed. In such experiments, there is no significant solid solubility of K in the Na₂O. Inspection of the data in Table 2 shows that Na₂O is also considerably more stable than Cs₂O and, since the solubilities of K and Cs in Na₂O should be similarly low, similar behavior of Cs with respect to oxygen might be expected. Unless the distribution coefficient for Cs between the Na (liquid) and Na₂O phases is markedly different at very low Cs concentrations, there should be no stabilization of Cs in the Na₂O lattice and hence no preferential removal of Cs from liquid Na in a cold trap. As was noted above, a high distribution coefficient is actually found. It seems reasonable therefore to assume that some other mechanism exists other than simple coprecipitation by oxygen.

One possibility is adsorption on surfaces, presumably oxide surfaces. To test this hypothesis, cesium cold-trapping studies were initiated. In the first of these loop experiments, the objective was to study the relationship between the radioactivity deposition in the cold trap and the amount of sodium oxide removed from the coolant stream by the cold trap and presumably deposited on surfaces. The apparatus consists of a small loop of stainless steel tubing of 98 ml capacity (Figure 14). The loop includes a cold-trap section, a loading and thermal expansion chamber at the top for insertion of oxide and radioactive material, and a small electromagnetic pump to provide circulation. An immersion heater of 500 watt capacity is provided in the leg adjacent to the cold trap. The remainder of the loop system is traced with resistance heaters, appropriately insulated; and thermocouples are provided at key points in the circuit.

at 900°F. The equilibrium radiation levels were then observed as the cold trap was successively cooled in finite temperature increments from the isothermal system temperature of 900 to 250°F. At each temperature level, the system was equilibrated for approximately 30 minutes. The temperature data for a typical run are shown schematically in Figure 15. These data show that operation of the cold trap caused a slight decrease in the temperatures in the balance of the loop.

1-15-68 UNCL

7702-4563

Figure 15. Temperature Data for Typical Cold Trapping Run

In principle, data on the solubility of oxygen in sodium permits estimation of the amount of oxide which can be retained in solution at each temperature, and thus deduction of the quantity of oxide deposited on the cold-trap surfaces. Comparison of the radiation curve and the oxide deposition curve for various oxide levels should then provide some indication of the $\text{Cs}_2\text{O}-\text{Na}_2\text{O}$ relationship.

Early experiments of this type, while showing correlation between temperature and radiation, were only partially successful in determining the correlation between amounts of cesium and sodium oxide trapped. Obviously to

investigate the effect of adsorption on oxide deposition, the effect of oxide concentration (at various starting concentrations) should also be evaluated. This was done in the next series of experiments.

The lowest oxygen concentration (as oxide) was approximately 5 ppm and was limited by the purity of the sodium employed. Following a typical temperature plateau run as described above, a sufficient quantity of oxygen was added to raise the level to 25 ppm. Similar procedures were employed in 25 ppm increments until the cesium trapping had been observed in sodium containing up to 150 ppm of additional oxygen. The results for 5 to 105 ppm of oxide are shown in Figures 16 and 17. The data in Figure 17 were obtained by increasing the amount of stable cesium in the loop after the 155 ppm oxygen run from 65 to 330 ppm and indicate, to some extent at least, a probable exchange or dilution phenomenon between the radioactive cesium removed by the cold trap and the additional stable cesium added.

The third series of experiments was performed to verify the effects and trends observed in the other two. For example, the flow rate was varied for the low oxygen concentration run and showed little change in results. A "blank" run was also made with no radioactivity in the sodium to verify that the temperature variations of the cold trap had not been affecting the detector response.

The results of the screening studies performed on the small loop seem to indicate at least an observable trend of trapping characteristics for cesium under various levels of oxygen concentration in the sodium. These preliminary experiments imply a trapping mechanism which is operative even at temperatures at which the oxygen would presumably be in solution. This might indicate that deposited Na_2O in the cold trap may not be the primary mechanism related to the trapping. Speculation may imply a mechanism related to the reaction of the oxygen with steel constituents and a consequent effect on the trapping characteristics. The experimental data obtained for the two cesium concentrations are too limited to allow a conclusive evaluation. However, the data do indicate that the introduction of stable Cs provided a possible mechanism for exchange of stable Cs with radioactive Cs, thus possibly explaining the decrease in radioactivity and relative trapping efficiency. Further studies may explore this possibility. In summary, the purposes of the existing loop were fulfilled in outlining

1-15-68 UNCL

7702-4565

Figure 16. Radiation Level in Cold Trap vs Cold Trap Inverse Temperature for Various Oxide Concentrations (65 ppm cesium)

1-15-68 UNCL

7702-4564

Figure 17. Radiation Level in Cold Trap vs Cold Trap Inverse Temperature for Two Cesium Concentrations (155 ppm oxygen)

problem areas which will be more amenable to solution in the more sophisticated and flexible loop presently being prepared (see Section IV-B-2).

2. Fission Product Behavior Loop

The Fission Product Behavior Loop (Figure 18) is intended to study the deposition tendencies of the various fission products. The loop consists of a surge tank; 3/8-in. diameter interconnecting tubing; two stainless steel packed traps capable of operating independently over the temperature range of 300 to 1200°F; and a series of by-pass stations to be used to inject individual fission products, to condition loop sodium, and to collect samples for chemical analysis. With the exception of Type 316 stainless steel valves, the loop is constructed entirely of Type 304 stainless steel. Design parameters for the loop are presented in Table 8. Gamma counting will be used to determine, in situ, the distribution of the fission products. Table 9 presents the radioisotopes to be used for the initial set of experiments. These experiments are designed to provide insight into the following problems:

- 1) In thoroughly cold-trapped sodium: the reversibility of deposition of fission products on system walls and the effect of system temperature gradients on this deposition.
- 2) In sodium containing appreciable oxygen: fission product deposition sites, the reversibility of the deposition, and the effect of system temperature gradients on deposition.
- 3) Evaluation of the effectiveness of stainless steel in hot and cold traps for the removal of fission products from sodium.

The loop is presently in the final stages of preparation for initial tests.

TABLE 8
DESIGN PARAMETERS FOR FISSION PRODUCT
BEHAVIOR LOOP

Loop Temperature (°F)	300 to 1200
Trap Temperature (°F)	300 to 1200 (independently variable)
Sodium Volume (cc)	200
Sodium Flow Rate (ft/sec)	
Loop Tubing	0.3 to 3.0
Annular Section of Trap	0.3 to 2.6
Unpacked Trap Section	0.04 to 0.4
Pump Throat	0.7 to 6.2
Material of Construction	Type 304 stainless steel (except valves, Type 316 stainless steel)

6-10-68 UNCL

7702-4065A

Figure 18. Fission Product
Behavior Loop

TABLE 9
RADIOISOTOPES FOR FISSION
PRODUCT BEHAVIOR
LOOP STUDIES

Analogue Group	Radioisotope
Cs	^{134}Cs
Ba, Sr	^{133}Ba
Mo, Ru, Sb, Ag	^{103}Ru , ^{124}Sb
Ce, La, Nd, Y, Zr	^{141}Ce
Nb	^{182}Ta
^3H , I, Te	^{75}Se

V. SYSTEM STUDIES

A. REDUCTION OF FISSION PRODUCTS OBSERVED IN SRE SODIUM

The deposition of fission product radioactivity at various points in an LMFBR system may result either from operation with defective fuel elements or, in the case of a vented fuel design, may occur as a result of the solution in sodium of the radioactive daughter products of volatile or gaseous fission products.

The concept of a "system depletion factor" provides an interim method for estimating this deposition. The preliminary observations which form the basis of this study were obtained as a result of evaluating the time-dependent fission product concentration in the coolant of the SRE, and the Fermi reactor.⁽⁴⁾

Table 10 shows the decrease in radioactivity with time for certain isotopes in the SRE primary sodium, as reported by Hart.⁽¹⁾

TABLE 10
TIME-DEPENDENCE OF FISSION PRODUCT CONTAMINATION
IN SRE PRIMARY SODIUM

Isotope	Fission Product Contamination ($\mu\text{c/g Na}$)			Fraction of Original Radioactivity Remaining in Coolant*	
	August 2, 1959	October 31, 1959	July 26, 1960	October 1959	July 1960
Cs ¹³⁷	1.26	0.45	1.2×10^{-2}	3.6×10^{-1}	9.55×10^{-3}
Cs ¹³⁴	0.02	0.006	Not observed	3.0×10^{-1}	-
Sr ⁹⁰	0.97	0.060	Not observed	6.2×10^{-2}	-
Sr ⁸⁹	19.0	0.25	Not observed	1.32×10^{-2}	-
I ¹³¹	0.42	0.00012	Not observed	2.86×10^{-4}	-
Ce ¹⁴¹	3.7	0.000088	Not observed	2.38×10^{-5}	-
Ce ¹⁴⁴	5.1	0.00031	Not observed	6.08×10^{-5}	-
Zr ⁹⁵ and Nb ⁹⁵	13.0	0.0067	Not observed	5.15×10^{-4}	-
Ru ¹⁰³	0.85	0.0045	Not observed	5.3×10^{-3}	-

*On the basis of the radioactivity in the August 2 sample.

These data are plotted in Figure 19 as a function of time after the first measured concentration. Since the three Cs¹³⁷ points represent a fairly straight line on semi-log paper it was theorized that the radioactivity remaining in the sodium coolant at any time (t) may be represented by an equation of the form:

$$A = A_o e^{-(\lambda+\alpha)t} = A_o e^{-\beta t} \quad \dots(16)$$

where

A_o = radioactivity present at t_o ($\mu\text{c/g}$) (in this case August 2, 1959)

A = radioactivity present at some later time t ($\mu\text{c/g}$)

t = time (days)

α = system depletion factor (days^{-1})

λ = radioactive decay constant (days^{-1})

β = total depletion factor (days^{-1}), ($= \alpha + \lambda$)

It was next postulated that, since the radioactivity decay constant is known for each isotope, the SRE may be viewed as a system having isotopic depletion characteristics associated with only the system depletion factor α . (It is recognized that this coefficient may be associated with trapping, deposition, plating, combinations of these parameters, or other physical/chemical processes which would modify the radioactive concentration in the coolant.)

Table 11 presents the total and system depletion factors for the specific isotopes which were measured in the SRE as well as the radioactive decay constants associated with each. The last column presents the system depletion factor which was obtained from the exponential expression,

$$\alpha = \left[\frac{\log_e A_o / A}{t} \right] - \lambda \quad \dots(17)$$

4-18-67 UNC

7702-2518A

Figure 19. Total Radioactivity Depletion Characteristics for SRE System

TABLE 11

TOTAL AND SYSTEM DEPLETION FACTORS FOR SRE SYSTEM

Isotope	Depletion Factor (A/A_0)	Time After Initial Measurement (days)	Exponential Expression, βt	Total Depletion Factor (days ⁻¹)	Radioactive Decay Constant (days ⁻¹)	System Depletion Factor (days ⁻¹)
¹³⁷ Cs	0.5	60	0.693	1.16×10^{-2}	7.2×10^{-5}	1.16×10^{-2}
¹³⁴ Cs	0.5	51.5	0.693	1.35×10^{-2}	0.095×10^{-2}	1.26×10^{-2}
⁹⁰ Sr	0.5	22.0	0.693	3.14×10^{-2}	1×10^{-5}	3.14×10^{-2}
⁸⁹ Sr	0.5	13.7	0.693	5.05×10^{-2}	1.3×10^{-2}	3.75×10^{-2}
¹³¹ I	2.86×10^{-4}	90.0	8.16	9.07×10^{-2}	8.62×10^{-2}	2.8×10^{-3}
¹⁴¹ Ce	2.38×10^{-5}	90.0	10.65	1.18×10^{-1}	2.13×10^{-2}	9.69×10^{-2}
¹⁴⁴ Ce	6.08×10^{-5}	90.0	9.72	1.08×10^{-1}	2.43×10^{-3}	1.05×10^{-1}
⁹⁵ Zr-Nb	5.15×10^{-4}	90.0	7.08	8.4×10^{-2}	1.07×10^{-2}	7.34×10^{-2}
¹⁰³ Ru	5.3×10^{-3}	90.0	5.23	5.81×10^{-2}	1.73×10^{-2}	4.10×10^{-2}

Figure 20 presents a plot of the system removal characteristics for each isotope obtained by plotting the curve

$$A/A_o = e^{-\alpha t} \quad \dots(18)$$

where α now represents the system removal factor for the isotope of interest.

B. LMFBR SYSTEM BEHAVIOR

With the above observations on the behavior of solid fission products in sodium it is of interest to apply the depletion factor concept to a typical operating reactor. To do this, a set of operating conditions is specified together with some (hopefully) realistic assumptions. Specifically, a 2120 Mwt FBR which has operated for an extended period of time with one percent fuel pin cladding failures is examined. A design objective for this reactor would be to allow entrance to one of the three intermediate heat exchanger cells (IHX) shortly after reactor shutdown. This is to be accomplished by valving off the particular IHX cell. The reactor continues to operate at something like 2/3 of full-power for a pre-determined length of time and is then shut down. It is desired to enter the IHX cell immediately.

The radiation level within the IHX just prior to personnel access, is assumed to be due to the following sources:*

- 1) Na^{24} and Na^{22} Activity — From sodium-filled heat exchangers and piping; or, in the event the heat exchanger is drained, activity will be due to the sodium which adheres to the metal surfaces.
- 2) Fission Product Activity — From fission products which escape from failed fuel pins and subsequently plate out on metal surfaces and/or remain dispersed in the sodium.

*While it is shown in Section III-C-3 that corrosion products may be of major importance as regards coolant contamination, they are neglected in this example since we are merely concerned with illustrating the use of the depletion factor concept.

4-18-67 UNC

7702-2519A

Figure 20. System Depletion Characteristics for the SRE System

The additional assumptions used in the analysis are as follows:

- 1) The fission products considered most significant from the standpoint of after-shutdown radiation are Zr^{95} - Nb^{95} , Ba^{140} - La^{140} *, and Cs^{137} - Ba^{137m} .
- 2) The quantity of these fission products released into the coolant is based on cladding failures in 1% of the fuel pins and release of 0.1% of the isotope inventory from the failed pins.
- 3) When the fission products are assumed to plate out, they plate out uniformly on all metal surfaces outside the reactor vessel.
- 4) When the fission products are assumed to remain in the sodium, they remain uniformly dispersed in the primary coolant. The quantity of sodium adhering to metal surfaces after draining is assumed to be 0.017 lb/ft^2 .
- 5) The total surface area available for fission product plating or for sodium adherence is $80,000 \text{ ft}^2$. The corresponding surface area of a heat exchanger (one of three) is $25,000 \text{ ft}^2$.
- 6) A heat exchanger is assumed to be 35-ft long by 5-ft in diameter. The radiation levels indicated in Figure 21 are based on a line source located in the center of the heat exchanger without any self-absorption of the gamma-rays. Due to the vast quantities of sodium present when the IHX is not drained, the radiation levels shown in Figure 22, are based on a line source with sodium attenuation for the plated fission products and on an infinite pool of sodium for the fission products in sodium.

*Because the Ba^{140} activity had decayed below the limit of detection in the SRE measurements, it was not possible to determine a depletion factor for this isotope from these data. However, two measurements were recently obtained for Ba^{140} radioactivity in the sodium coolant of the Fermi reactor. While it is recognized that gross system differences exist between the two reactor systems, the Fermi depletion factor for Ba^{140} ($\alpha = 2.61 \times 10^{-1} \text{ day}^{-1}$) has been used in this example, because of the importance of this isotope.

Figure 21. Radiation Levels 3 ft from Surface of Heat Exchanger - Heat Exchanger Drained

7-026-243-8

Figure 22. Radiation Levels 3 ft from Surface of Heat Exchanger - Heat Exchanger Remains Filled with Primary Coolant

7-026-243-9

- 7) The equilibrium specific activity of the sodium due to Na^{24} and Na^{22} is as follows:

$$\text{Na}^{24} - 0.0774 \text{ curies per cc}$$

$$\text{Na}^{22} - 2.49 \times 10^{-6} \text{ curies per cc}$$

- 8) The reactor has operated for 30 years with the fuel failures specified above. The isotopes ^{24}Na , ^{22}Na , ^{95}Zr - ^{95}Nb , and ^{140}Ba - ^{140}La have reached their equilibrium activities. ^{137}Cs , with a half-life of 30 years, has reached 1/2 its equilibrium concentration.

The results obtained using the above assumptions are shown in Figures 21, 22, 23, and 24.

Figure 21 shows, as a function of time after isolation, the calculated radiation level 3 ft from a heat exchanger which has been drained of sodium and isolated from the primary coolant flow. The "fission products plated" curve depicts the calculated radiation levels assuming 100% of the fission products have been removed from the sodium and deposited on the piping surfaces. The "fission products remain in sodium" curve shows the estimated radiation levels assuming there is no fission product deposition or plating, so that the entire dose is due to the 0.017 lb/ft^2 of Na adhering to the heat exchanger surfaces. By comparing these two curves it is seen that the effect of complete plating is to increase the radiation level from a drained heat exchanger by a factor of about 10^3 .

The ^{24}Na and ^{22}Na activities are also shown in Figure 21. As would be anticipated ^{24}Na is a matter of concern only for the first few days. It is also seen, however, that ^{22}Na could become important approximately one month after isolation, but only if all of the fission products were to remain in the sodium.

Figure 22 shows the expected radiation levels assuming the heat exchanger is not drained. Here it is seen that the effects of fission product plating is much less marked, causing an increase in radiation levels of only a factor of 3 to 4 over the no-plating case.

Figure 23. Percent Contribution of Individual Fission Product Isotopes to Total Fission Product Radiation Level

Figure 24. Estimated Removal Fraction for Significant Fission Products

As indicated, the above curves provide dose rates only for the cases of 100% or 0% fission product deposition. The depletion factor concept, however, theorizes that the degree of fission product deposition is a function of time and of the isotopic species involved. To apply this concept, it is therefore necessary to identify the isotopes responsible for the radiation levels given in Figures 21 and 22 and to apply the time factor as it affects deposition.

Figure 23 provides the identification of the isotopic species primarily responsible for the fission product radiation levels and their relative contributions to the dose rate as a function of time. These numbers were directly derived from the dose rate calculations.

Finally, the degree of deposition of each of the isotopic species identified above, assuming continuous release of fission products from the fuel, is given by Figure 24. The plotted quantity in Figure 24 is the fraction of the available inventory removed from the sodium, f_i . This quantity was obtained in the following manner: Let the number of atoms of isotope "i" entering the coolant stream per unit time be a constant R_i . Then, in the absence of a depletion mechanism, the number of atoms of "i," $N_{i,w/o\ depl.}$ is

$$N_{i,w/o\ depl.} = \frac{R_i}{\lambda_i} (1 - e^{-\lambda_i t}) \quad \dots(19)$$

With a depletion mechanism present, the number of atoms of "i," $N_{i,w\ depl.}$ is,

$$N_{i,w\ depl.} = \frac{R_i}{\lambda_i + \alpha_i} (1 - e^{-(\lambda_i + \alpha_i)t}) \quad \dots(20)$$

The fraction of "i" removed from the coolant is then the number of atoms that would be present in the absence of depletion minus the number that are present with depletion, the difference divided by the total inventory, or

$$\begin{aligned}
f_i &= \frac{N_{i,w/o\ depl} - N_{i,w\ depl}}{N_{i,w/o\ depl}} \\
&= \frac{\frac{R}{\lambda_i} (1 - e^{-\lambda_i t}) - \frac{R_i}{\lambda_i + \alpha_i} (1 - e^{-(\lambda_i + \alpha_i)t})}{\frac{R}{\lambda_i} (1 - e^{-\lambda_i t})} \\
&= 1 - \frac{\lambda_i}{\lambda_i + \alpha_i} (1 - e^{-(\lambda_i + \alpha_i)t}) / (1 - e^{-\lambda_i t}). \quad \dots(21)
\end{aligned}$$

This is the relationship plotted in Figure 24. Figure 24, however, covers operation up to only 1000 days. For the assumed 30 years of operation, f_i essentially reaches its asymptotic value,

$$f_i(\infty) = \frac{\alpha_i}{\lambda_i + \alpha_i} \quad \dots(22)$$

for all important isotopes except ^{137}Cs . In the case of ^{137}Cs the reactor has operated for one half-life, so that, with $\alpha_{^{137}\text{Cs}} \gg \lambda_{^{137}\text{Cs}}$,

$$f_{^{137}\text{Cs}} \approx 1 - \frac{2\lambda_{^{137}\text{Cs}}}{\lambda_{^{137}\text{Cs}} + \alpha_{^{137}\text{Cs}}} = \frac{\alpha_{^{137}\text{Cs}} - \lambda_{^{137}\text{Cs}}}{\alpha_{^{137}\text{Cs}} + \lambda_{^{137}\text{Cs}}}. \quad \dots(23)$$

The resulting removal fractions are then as shown in Table 12.

Based on the foregoing discussion, it is possible to estimate the dose rate 3 ft from a heat exchanger, 10 days after it has been isolated and drained. Considering ^{95}Zr - ^{95}Nb , the total dose rate due to deposited fission products (assuming 100% deposition) 10 days after isolation is, from Figure 21, 120 r/hr. Of this total, however, Figure 23 indicates that only 40% of this dose, or 48 r/hr, is due to ^{95}Zr - ^{95}Nb . Furthermore, Table 12 shows that only 87% of the ^{95}Zr - ^{95}Nb is deposited, so that the dose due to the deposited fraction of these isotopes

TABLE 12
FRACTION OF AVAILABLE INVENTORY REMOVED FROM
SODIUM, 30 YEARS OPERATION, CONTINUOUS
FISSION PRODUCT RELEASE

Isotope	f_i	$1 - f_i$
$^{95}\text{Zr}-^{95}\text{Nb}$	0.87	0.13
$^{140}\text{Ba}-^{140}\text{La}$	0.83	0.17
$^{137}\text{Cs}-^{137\text{m}}\text{Ba}$	0.99	0.01

is 42 r/hr. Since the heat exchanger is drained, the dose due to $^{95}\text{Zr}-^{95}\text{Nb}$ in the residual sodium will be down by a factor of at least three orders of magnitude and so may be neglected. A similar assessment for the other two isotopic species gives values as follows,

$$^{140}\text{Ba}-^{140}\text{La}: 120 \text{ r/hr} \times 0.48 \times 0.83 = 48 \text{ r/hr}$$

$$^{137}\text{Cs}-^{137\text{m}}\text{Ba}: 120 \text{ r/hr} \times 0.12 \times 0.99 = 14 \text{ r/hr}$$

The total dose rate 10 days after isolation is therefore 104 r/hr.

Referring to Figure 22, a similar calculation performed for the case of a heat exchanger filled with sodium 10 days after isolation gives the dose rates shown in Table 13.

In both of the above examples the deposited fission products make a dominant contribution to the residual radiation levels. The limitations of the example, however, should be reiterated. It is presented merely to introduce and illustrate the potential usefulness of the depletion factor concept. Insofar as accuracy is concerned, this is probably very low at the present time, because the depletion constants, α , were based on very limited data from two reactor systems which bear little detailed resemblance to projected LMFBR's. Further, the assumption of uniform plating seems highly questionable, since one would normally expect some preferential plating in either the cold or hot

TABLE 13
DOSE RATES 3 ft FROM SURFACE OF HEAT EXCHANGER
CONTAINING PRIMARY SODIUM, 10 DAYS AFTER
SHUTDOWN, FOLLOWING 30 YEARS OF
REACTOR OPERATION

Isotope	Dose Rates (r/hr)		Total Dose Rate (r/hr)
	From Plated Fission Products	From Activity in Sodium	
$^{95}\text{Zr}-^{95}\text{Nb}$	$65 \times 0.40 \times 0.87 = 23$	$17 \times 0.40 \times 0.13 = 0.88$	23.9
$^{140}\text{Ba}-^{140}\text{La}$	$65 \times 0.48 \times 0.83 = 26$	$17 \times 0.48 \times 0.17 = 1.39$	27.4
$^{137}\text{Cs}-^{137m}\text{Ba}$	$65 \times 0.12 \times 0.99 = 8$	$17 \times 0.12 \times 0.01 = 0.02$	8.0
^{24}Na		7.0	7.0
^{22}Na		3.0	3.0
	57	12.29	69.3

legs of the loop or in cold traps or other parts of the system. Finally, the question of corrosion product activities has been neglected and, as shown in an earlier section, these activities might ultimately be dominant. Nevertheless, the depletion factor concept is considered very useful as an interim or preliminary tool for the assessment of the effect of deposited activity.

REFERENCES

1. R. S. Hart, "Distribution of Fission Product Contamination in the SRE," NAA-SR-6890 (March 1962)
2. A. I. Leipunsky, et. al., "Five Year Operation Experience on Reactor BR-5," Third United Nations International Conference on the Peaceful Uses of Atomic Energy, A/CONF/ 28/P/312 (May 1964)
3. R. R. Mathews, et. al., "Performance and Operation of the Dounreay Fast Reactor," Third United Nations International Conference on the Peaceful Uses of Atomic Energy, A/CONF/28/P/130 (May 1964)
4. R. E. Mueller, et. al., "Results of Fission-Product Analysis to Determine Extent of Fuel Failure in Fermi Accident," Trans. Am. Nucl. Soc. 10: 334 (1967)
5. K. M. Broom, Jr., C. D. Bingham, T. B. Crockett and N. M. Trahey, "Post-Irradiation Chemical Analysis of SNAP-8 Experimental Reactor Coolant," Nuclear Applications, Vol. 2, p. 519 (December 1966)
6. W. P. Kunkel. "Fission Product Retention in Sodium - A Summary of Analytical and Experimental Studies at Atomics International," NAA-SR-11766 (August 1966)
7. J. C. Clifford, et. al., "Behavior of Fission Products in Sodium," presented at IAEA Symposium on Alkali Metal Coolants, Vienna, Austria (November 28 to December 2, 1966) also available as LA-DC-8094
8. Code of Federal Regulations, Title 10, Chapter I, Part 20, "Standards for Protection Against Radiation"
9. C. A. Anderson, Jr., "Fission Product Yields from Fast (1 Mev) Neutron Fission of Pu-239," LA-3382 (December 1965)
10. C. B. Jackson, Ed., "Liquid Metals Handbook, Sodium (NaK) Supplement," 3rd ed., Supt. Doc., U. S. Gov't Printing Office (1955)
11. R. L. Eichelberger and R. L. McKisson (both at Atomics International), personal communication
12. A. V. Nesmeyanov, "Vapor Pressure of the Elements," Academic Press, New York (1963)
13. J. P. Coughlin, "Heats and Free Energy of Formation of Inorganic Oxides," NSB Bulletin 542 (1954)
14. A. Glässner, "The Thermochemical Properties of the Oxides, Flourides and Chlorides to 2500°K," ANL-5750 (No Date)

15. D. R. Stull, et. al., "JANAF Thermochemical Tables," PB 168370, (August 1967)
16. "NASA-AEC Liquid Metals Corrosion Meeting," Vol. I, Lewis Research Center, Cleveland, Ohio, p 56 (October 2-3, 1963)
17. C. E. Wicks and F. E. Block, "Thermodynamic Properties of 65 Elements- Their Oxides, Halides, Carbides and Nitrides," Bu. Mines Bull. 605 (1965)
18. R. P. Elliot, "Constitution of Binary Alloys, First Supplement," McGraw-Hill, New York (1965)
19. M. Hansen, "Constitution of Binary Alloys," 2nd ed., McGraw-Hill, New York (1958)
20. D. R. Stull and G. C. Sinke, "Thermodynamic Properties of the Elements," Am. Chem. Soc., Washington, D. C. (1956)
21. M. Blander, ed., "Molten Salt Chemistry," Chapter by S. H. Bauer and R. F. Porter titled "Metal Halide Vapors." Interscience Publishers, New York (1964)
22. N. A. Bredig, J. W. Johnson and W. T. Smith, J. Am. Chem. Soc., 77, 307-312 (1955)
23. T. Yokokawa and O. J. Kleppa, J. Chem. Phys., 40, 46-54 (1964)
24. J. Saroul, "Investigation of the Behavior of Fission Products in Sodium and Argon - PIRANA Experiments," Int. Conf. on the Safety of Fast Reactors, Aix-en-Provence (September 1967)
25. W. S. Clough in "A Summary of Research and Development Work Carried Out in 1966 in Health and Safety Departments of UKAEA," AHSB (RP) M 46 (September 1967)
26. R. L. Koontz, Atomics International, Unpublished data (available upon request)
27. A. W. Castleman, Jr. and I. N. Tang, "Thermodynamics of Fission Product - Sodium Solutions," BNL-11611 (November 1967)
28. A. W. Castleman, Brookhaven National Laboratory, private communication with M. Silberberg (Atomics International)
29. A. I. Hansen, "The Effects of Long-Term Operation on SRE Sodium System Components," NAA-SR-11396 (August 1965)