**COMPARISONS OF THE CONTAINMENT
BEHAVIOR OF A SIMULANT
WITH FISSION PRODUCTS
RELEASED FROM IRRADIATED UO_2**

R. K. HILLIARD
L. F. COLEMAN
J. D. McCORMACK

MARCH, 1968

**AEC RESEARCH &
DEVELOPMENT REPORT**

RECORD NO.	PROJECT	SECTION	FILE NUMBER
DUN for 60001 703 JUL 10 1968			
R. L. Etheridge, 5417			

BATTELLE

NORTHWEST

BATTELLE MEMORIAL INSTITUTE

PACIFIC NORTHWEST LABORATORY

3000 STEVENS DRIVE, P. O. BOX 998, RICHLAND, WASHINGTON 99352

3 3679 00060 6998

BNWL-581

UC-80, Reactor Technology

COMPARISONS OF THE CONTAINMENT BEHAVIOR OF
A SIMULANT WITH FISSION PRODUCTS
RELEASED FROM IRRADIATED UO_2

By

R. K. Hilliard

L. F. Coleman

J. D. McCormack

Reactor Engineering Section
Engineering Development Department

March 1968

FIRST UNRESTRICTED
DISTRIBUTION MADE

APR 8 '68

PACIFIC NORTHWEST LABORATORY
RICHLAND, WASHINGTON

BNWL-581

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

COMPARISONS OF THE CONTAINMENT BEHAVIOR OF
A SIMULANT WITH FISSION PRODUCTS
RELEASED FROM IRRADIATED UO_2

R. K. Hilliard

L. F. Coleman

J. D. McCormack

ABSTRACT

Experiments were performed in which either simulant aerosols or fission products released from heated trace-irradiated UO_2 were released into small vessels containing condensing steam-air atmospheres. The behavior of the two different types of aerosol materials was characterized and compared with each other and with transport theory. The conclusion was made that the simulant aerosol developed for the Containment Systems Experiment (CSE) is a fully satisfactory substitute for real fission products in containment transport tests investigating removal by natural processes.

TABLE OF CONTENTS

	LIST OF FIGURES.	v
	LIST OF TABLES	viii
I.	INTRODUCTION.	1
II.	SUMMARY AND CONCLUSIONS	3
III.	THEORETICAL CONSIDERATIONS	5
	Mathematical Models.	6
	Diffusional Models	6
	Well Mixed Models.	7
	Estimation of Mass Transfer Coefficients.	16
IV.	EXPERIMENTAL CONDITIONS	21
	Experiment Structure	21
	Experimental Equipment.	22
	Test Conditions	26
	Irradiated Source Tests.	26
	Simulant Tests.	29
	Methods of Sample Analysis	30
V.	RESULTS AND DISCUSSION	31
	Source Term	31
	Irradiated Source Tests.	32
	Simulant Tests.	34
	Changes in Gas Phase	37
	Maypack Sample Interpretation.	38
	Gas Concentration Time Dependence	39
	Iodine Form in Containment.	40
	Mass Transfer Coefficients.	40
	Gas-Liquid Equilibrium	42
	Distribution Within Containment.	44
	Form in Condensate	47
	Overall Material Recovery.	49
	Miscellaneous Observations	50
	Effect of Mass Concentration	50
	Effect of Aerosol Complexity	50

TABLE OF CONTENTS (contd)

	Effect of UO_2 Cladding Material	51
	Fresh Versus Aged Paint	51
	Sampling Through Long Lines.	52
	Effect of Agitation	52
VI.	ACKNOWLEDGEMENTS.	55
VII.	NOMENCLATURE	56
VIII.	REFERENCES.	58
	APPENDIX A—Tables 1 through 34	A-1
	APPENDIX B—Figures B.1 through B.52	B-1
	DISTRIBUTION	Distr-1

LIST OF FIGURES

1.	Typical Gas Phase Curve from ADF Model	14
2.	Aerosol Development Facility Layout	23
3.	Photograph of SAT Vessel Being Sampled with Maypack Sampler.	25
4.	ADF Maypack (Schematic)	27
5.	Maypack Samples Prepared for Gamma Energy Analysis	28
6.	Comparison of Elemental Iodine Gas Phase Concentration-- Continuous Versus Instantaneous Release	33
7.	Effect of Agitation on Elemental Iodine Gas Phase Concentration	54
B.1.	Iodine Gas Phase Concentration, Run SA 21	B.1
B.2.	Iodine Gas Phase Concentration, Run SA 23	B.2
B.3.	Iodine Gas Phase Concentration, Run IA 26	B.3
B.4.	Iodine Gas Phase Concentration, Run SA 28	B.4
B.5.	Iodine Gas Phase Concentration, Run IA 32	B.5
B.6.	Iodine Gas Phase Concentration, Run IA 33	B.6
B.7.	Iodine Gas Phase Concentration, Run IA 39	B.7
B.8.	Iodine Gas Phase Concentration, Run IA 40	B.8
B.9.	Iodine Gas Phase Concentration, Run IB 41	B.9
B.10.	Iodine Gas Phase Concentration, Run IB 42	B.10
B.11.	Iodine Gas Phase Concentration, Run SA 43	B.11
B.12.	Iodine Gas Phase Concentration, Run SA 44	B.12
B.13.	Iodine Gas Phase Concentration, Run SB 48	B.13
B.14.	Iodine Gas Phase Concentration, Run SB 50	B.14
B.15.	Iodine Gas Phase Concentration, Run SB 53	B.15
B.16.	Iodine Gas Phase Concentration, Run SB 60	B.16

LIST OF FIGURES (contd)

B.17. Iodine Gas Phase Concentration, Run SB 61	B.17
B.18. Iodine Gas Phase Concentration, Run SB 63	B.18
B.19. Iodine Gas Phase Concentration, Run SB 64	B.19
B.20. Iodine Gas Phase Concentration, Run SB 74	B.20
B.21. Iodine Gas Phase Concentration, Run SB 78	B.21
B.22. Gas Phase Concentrations, Run IA 32	B.22
B.23. Gas Phase Concentrations, Run IA 33	B.23
B.24. Gas Phase Concentrations, Run IA 39	B.24
B.25. Gas Phase Concentrations, Run IA 40	B.25
B.26. Gas Phase Concentrations, Run IB 41	B.26
B.27. Gas Phase Concentrations During the Release Period, Run IB 41	B.27
B.28. Gas Phase Concentrations, Run IB 42	B.28
B.29. Gas Phase Concentrations, Simulant Cesium	B.29
B.30. Gas Phase Concentrations, Simulant Tellurium	B.30
B.31. Gas Phase Concentrations, Simulant Ruthenium	B.31
B.32. Gas Phase Concentrations, Simulant Barium	B.32
B.33. Gas Phase Concentrations, Simulant Uranium	B.33
B.34. Mass Transfer Coefficient Versus Steam Flux, Total Iodine	B.34
B.35. Mass Transfer Coefficient Versus Steam Flux, Particulate Iodine	B.35
B.36. Mass Transfer Coefficient Versus Steam Flux, Elemental Iodine	B.36
B.37. Mass Transfer Coefficient Versus Steam Flux, Cesium	B.37

LIST OF FIGURES (contd)

B.38. Mass Transfer Coefficient Versus Steam Flux, Tellurium	B.38
B.39. Mass Transfer Coefficient Versus Steam Flux, Ruthenium	B.39
B.40. Mass Transfer Coefficient Versus Steam Flux, Barium	B.40
B.41. Mass Transfer Coefficient Versus Steam Flux, Uranium	B.41
B.42. Iodine Distribution Versus Series IA	B.42
B.43. Iodine Distribution Versus Series SA	B.42
B.44. Iodine Distribution Versus Series IB	B.43
B.45. Iodine Distribution Versus Series SB	B.43
B.46. Cesium Distribution Versus Series IB	B.44
B.47. Cesium Distribution Versus Series SB	B.44
B.48. Tellurium Distribution Versus Series IA	B.45
B.49. Tellurium Distribution Versus Series SB	B.45
B.50. Ruthenium Distribution, Series SB	B.46
B.51. Barium Distribution, Series SB	B.46
B.52. Uranium Distribution, Series IA	B.47

LIST OF TABLES

1	Series I Experimental Conditions	A.1
2	Series S Experimental Conditions	A.2
3	Fission Product Release from Irradiated Specimens	A.4
4	Iodine Form as Sampled from Delivery Line	A.5
5	Effect of Iodine Mass Loading on Maypack Distribution	A.6
6	Iodine Form in Containment Gas Phase After 10 min Containment Time	A.7
7	Iodine Form in Containment Gas Phase After 5 hr Containment Time	A.8
8	Effect of Mass Released on Iodine Form	A.9
9	Gas Phase Mass Transfer Coefficients--Total Iodine	A.10
10	Gas Phase Mass Transfer Coefficients--Particulate Iodine	A.11
11	Gas Phase Mass Transfer Coefficients--Elemental Iodine	A.12
12	Gas Phase Mass Transport Coefficients--Cesium	A.13
13	Gas Phase Mass Transport Coefficients--Tellurium	A.14
14	Gas Phase Mass Transport Coefficients--Ruthenium	A.15
15	Mass Transfer Coefficients--Barium	A.16
16	Gas Phase Mass Transfer Coefficients--Uranium	A.17
17	Summary of Gas Phase Mass Transfer Coefficients	A.18
18	Comparison of K_G for Various Aerosol Components	A.19
19	Gas-Liquid Partition Coefficients for Elemental Iodine	A.20
20	Iodine Distribution in Containment Vessel After 5 Hr	A.21

LIST OF TABLES (contd)

21	Cesium Distribution in Containment Vessel After 5 Hr	A.22
22	Tellurium Distribution in Containment Vessel After 5 Hr	A.23
23	Ruthenium Distribution in Containment Vessel After 5 Hr	A.23
24	Barium Distribution in Containment Vessel After 5 Hr	A.24
25	Uranium Distribution in Containment Vessel After 5 Hr	A.24
26	Summary of Distribution After 5 hr Containment Aging	A.25
27	Solubility in Steam Condensate After 10 min Containment Time	A.26
28	Solubility in Steam Condensate After 5 hr Containment Time	A.27
29	Iodine Form in Condensate as Measured by Extraction	A.28
30	Overall Material Recovery	A.29
31	Effect of Aerosol Complexity on Iodine Containment Behavior	A.30
32	Effect of Cladding Type on Iodine Behavior	A.31
33	Comparison of Iodine Behavior in Containment Vessels with Fresh Versus Aged Painted Walls	A.32
34	Effect of Long Inlet Line on Sampler	A.33

COMPARISONS OF THE CONTAINMENT BEHAVIOR OF
A SIMULANT WITH FISSION PRODUCTS
RELEASED FROM IRRADIATED UO_2

R. K. Hilliard

L. F. Coleman

J. D. McCormack

I. INTRODUCTION

A major objective of the Containment Systems Experiment (CSE) is to demonstrate the transport behavior of fission products during conditions representative of major accidents to water cooled and moderated nuclear reactors.⁽¹⁾ All reactors have some provision for containing fission products that might escape from the primary reactor vessel during an accident, and the CSE structure includes several different types of safeguard features used in large modern power reactors.⁽¹⁾ Tests will be performed in a manner whereby mass transport models derived from theory and small-scale tests can be confirmed in an engineering-scale facility. The proven mathematical models will then provide reliable means for estimating fission product transport under a wide variety of potential accident conditions in a reactor system.

The experimental program⁽¹⁾ calls for repetitive releases of large masses of fission product aerosols: a requirement well met by the use of simulants but impractical if highly irradiated fuels are used.⁽²⁾ The use of simulants is justified only if the simulants' behavior within the containment environment is identical to, or can be correlated with, that of "real" fission products: that is, fission products released from irradiated power reactor fuel elements.

The Aerosol Development Facility (ADF) is a small-scale containment laboratory operated in support of the CSE at the Pacific Northwest Laboratory (PNL). One of its functions is to compare the behavior of the CSE fission product aerosol with that of fission products released from overheated UO_2 . The findings are the substance of this report. A related study is in progress at the Oak Ridge National Laboratory (ORNL) where the containment behavior of the CSE simulant is being compared with the aerosol released from highly irradiated fuels in the Containment Mockup Facility (CMF) and in the Containment Research Installation (CRI).

This report is one of a series of topical reports on the CSE program. The first⁽¹⁾ was a general description of the experimental program and the planned experimental facility. Other reports have dealt with a mathematical model and computer code for pressure and temperature transients in containment systems,⁽³⁾ and the method of analyzing samples generated in the CSE and ADF for the amounts of each of several fission product radionuclides.⁽⁴⁾

II. SUMMARY AND CONCLUSIONS

Experiments have been performed in a manner whereby a direct comparison could be made between the transport of simulant and reference fission product aerosols in a condensing steam containment atmosphere. The reference fission product aerosol was produced by heating stainless steel clad UO_2 irradiated to 1×10^{17} nvt (~ 0.35 MWd/T) in oxidizing and reducing atmospheres at various temperatures up to that of molten UO_2 . The simulant aerosol was produced by volatilizing stable fission product elements or compounds that were radioactively traced for analytical purposes. The aerosol was injected into a containment vessel filled with an equimolar steam and air mixture. Steam condensed at a steady rate by heat transfer through the vessel surfaces and was replaced by a continuous steam addition to maintain isothermal and isobaric containment conditions.

In order to provide a valid basis for comparing the transport of simulant and reference aerosols, a theoretical model was developed. The model assumes that the bulk gas in containment is well mixed by eddy diffusion and that mass transfer from the gas to the vessel surface is limited by diffusion across a boundary layer. The surface of the vessel is wet by condensing steam. Transport from the gas to the liquid film is limited by the gas phase mass transfer coefficient until equilibrium between gas and liquid is attained. After equilibrium is obtained, transport is limited by the dilution rate of the liquid film by fresh condensate.

The gas phase mass transfer coefficient is a function of the sum of two quantities: the bulk fluid flux to the wall caused by condensation of steam, and diffusion across the boundary layer. In the current tests, the boundary layer was laminar, but in larger vessels it could be turbulent.

Watson recognized that the diffusive contribution to mass transport was neglected, but he assumed that the bulk flow term greatly predominated so that his simplified model gave satisfactory, though conservatively low, results.

The authors, in cooperation with J. G. Knudsen,* made a mathematical analysis of the transport of fission products in a condensing steam-air mixture. Though adaptable for other systems, the simplified case to fit the ADF tests will be discussed here. The assumptions were:

- 1) Fission products were instantaneously injected into a steam-air atmosphere and were well mixed at all times.
- 2) All surfaces were wet by steam condensation that formed a continuous film draining to the sump.
- 3) Fission products were transferred across a gas boundary layer at the liquid surface by a combination of bulk and diffusive flow.
- 4) Fission products were absorbed reversibly by the liquid film, with equilibrium controlled by a suitable gas-liquid partition coefficient.
- 5) After absorption in the liquid, the fission products reacted irreversibly with the vessel walls with a first-order rate constant.
- 6) The concentration in the liquid film was assumed constant throughout, but differed from that in the sump by some proportionality constant. The model is adaptable to unsteady pressure and steam flux conditions, but the discussion will be limited to the ADF experimental condition of thermal equilibrium with steam-feed to make up for losses by condensation.

* Consultant to PNL. Assistant Dean of Engineering, Oregon State University, Corvallis, Oregon.

If we concern ourselves only with changes in the gas phase, the material balance for any single species of fission product or simulant is:

Input = Output + Accumulation

$$0 = K_D A (C_G - C'_G) + A N_B X_i + V \frac{dC_G}{dt}, \quad (4)$$

where K_D = mass transfer coefficient for diffusion through boundary layer, cm/sec

A = vessel surface area, cm^2

C_G = gas phase concentration, mole/ cm^3

C'_G = gas phase concentration in equilibrium with liquid, mole/ cm^3

N_B = flux of bulk gas to walls, mole $\text{sec}^{-1} \text{ cm}^{-2}$

X_i = mole fraction of species "i" in gas,
mole i/mole total

V = vessel volume, cm^3

t = time, sec

The first term on the right side is the rate of transfer from the gas to the liquid by diffusion across the concentration gradient. The second term is the transfer contributed by bulk gas flux to condensing surfaces. The third term is the difference between input and output. Since instantaneous release was assumed, the left term is zero at $t > 0$.

The mole fraction, X_i , can be expressed as

$$X_i = \frac{C_G RT}{P}, \quad (5)$$

where R = universal gas constant

T = absolute temperature in bulk gas

P = total pressure

Note that P is total pressure in the vessel, not just saturated steam pressure. The bulk flow term then becomes:

$$AN_B X_i = AN_B \frac{C_G RT}{P}, \quad (6)$$

and Equation (4) can be rewritten

$$0 = KD_A (C_G - C'_G) + \left(\frac{N_B RT}{P} \right) AC_G + V \frac{dC_G}{dt}. \quad (7)$$

Now, the equilibrium back pressure, C'_G , is a function of the liquid surface concentration. The relationship is conveniently put in the form of a dimensionless gas-liquid partition coefficient, H

$$C'_G = H C_L. \quad (8)$$

At short containment times, C_L is very small and $C_G \gg C'_G$. For this condition, Equation (7) becomes

$$0 \approx KD AC_G + \left(\frac{N_B RT}{P} \right) A C_G + V \frac{dC_G}{dt}. \quad (9)$$

Rearranging,

$$\frac{dC_G}{dt} = - \left(KD + \frac{N_B RT}{P} \right) \frac{A}{V} C_G. \quad (10)$$

A new transfer coefficient is now defined in Equation (11):

$$K_G = KD + \frac{N_B RT}{P}, \quad (11)$$

where K_G = overall gas phase mass transfer coefficient in cm/sec. Substituting in Equation (10) gives

$$\frac{dC_G}{dt} = -K_G \frac{A}{V} C_G. \quad (12)$$

Separation of variables and integration gives, with the initial condition that $C_G = C_o$ at time zero:

$$\frac{C_G}{C_o} = \exp \left(-K_G \frac{A}{V} t \right). \quad (13)$$

Equation (13) applies only as long as the bulk gas concentration, C_G , greatly exceeds the equilibrium concentration, C'_G . At equilibrium, $C_G = C'_G$ and the diffusive term in Equation (7) goes to zero. Furthermore, though the bulk flow mechanism continues to operate, C_G is forbidden to decrease lower than the equilibrium concentration because of reversibility. Therefore, the gas concentration decreases at a rate fixed by the dilution of the liquid film by fresh condensing steam.

Turning our attention to the liquid phase, we find the material balance is

$$\text{Input} = \text{Output} + \text{Accumulation}$$

$$K_G A (C_G - C'_G) = B C_L S + R_L A C_L + L \frac{dC_L}{dt}, \quad (14)$$

where

C_L = liquid film concentration, mole/cm³

S = steam condensation rate, cm³ liquid/sec

R_L = liquid-solid reaction rate constant, cm/sec

$B = C'_L / C_L$ = factor relating film and sump concentration

C'_L = liquid concentration in vessel sump, mole/cm³

L = volume of liquid film holdup, cm³

The left term is the input to the liquid phase by transfer from the gas. The first term on the right is removal from the film by flow to a sump. The second term on the right is transferred to solid surfaces by irreversible reaction with the walls. The last term is the difference between input and output. At equilibrium,

$$C'_G = H C_{L_{eq}} , \quad (15)$$

and

$$V \frac{dC'_G}{dt} = BS C_{L_{eq}} + R_L A C_{L_{eq}} + L \frac{dC_{L_{eq}}}{dt} , \quad (16)$$

$$VH \frac{dC_{L_{eq}}}{dt} = C_{L_{eq}} (BS + R_L A) + L \frac{dC_{L_{eq}}}{dt} , \quad (17)$$

$$\frac{dC_{L_{eq}}}{dt} = \left(\frac{BS + R_L A}{VH - L} \right) C_{L_{eq}} . \quad (18)$$

Integrating and using the initial condition that

$$\text{at } t = 0, C_{L_{eq}} = \frac{(C'_G)_0}{H} ,$$

$$\frac{H}{(C'_G)_0} C_{L_{eq}} = \exp \left[\frac{BS + R_L A}{VH - L} t \right] . \quad (19)$$

Substitution of Equation (15) gives, for equilibrium,

$$\frac{C'_G}{(C'_G)_0} = \exp \left[- \left(\frac{BS + R_L A}{L - VH} \right) t \right] . \quad (20)$$

For systems with small gas-liquid partition coefficients (e.g., I_2 , HI, particles) and where R_L is low, Equation (20) simplifies to

$$\frac{C_G}{(C_G)_0} = \exp - \frac{BS}{L} t , \quad (21)$$

which states that at equilibrium, the transfer from gas to liquid is controlled by the dilution rate of the liquid film by fresh condensate: the ratio BS/L . The assumption that concentration within the liquid film is uniform may be a poor one at high transfer rates, and liquid diffusion should be considered.

However, on the basis of the described model, a computer program was written that gives the roots of the equations and tabulated values of C_G/C_0 versus time. Expressions for time dependence of concentration in the liquid film and material transferred to solid surfaces were also obtained. Briefly, they predict an initial rapid buildup in liquid concentration followed by a slow decline asymptotically approaching zero. The total mass holdup by both liquid film and solid surfaces rises at a rapid initial, nearly linear rate, reaches a maximum, then decreases with time to an asymptotic value that is the total mass reacted with solid surfaces.

Figure 1 shows a typical gas phase curve from this program obtained by selecting various combinations of model input parameters to obtain the best fit to the experimental results for elemental iodine in ADF Run 63. Without elaborating on experimental results at this point, we should discuss several

FIGURE 1. Typical Gas Phase Curve from ADF Model

important features of the figure. First, the curve of concentration versus time has two segments. At early times the slope is approximately equal to $-K_G A/V$, agreeing with Equation (13). For a given vessel geometry (A/V), the transport depends only on the value of the gas phase mass transfer coefficient, K_G . All other variables are unimportant except as they affect the mass transfer coefficient.

The first segment of the curve holds until gas-liquid equilibrium is approached. A second exponential segment is seen as predicted by Equation (20) and approximated by Equation (21). Extrapolation of the equilibrium curve gives an intercept at time zero of HV/L . At zero condensation rate (i.e., $S = 0$), the equilibrium curve would be constant at $C/C_0 = HV/L$.

Morrison et al. (11,12) derived transport models for both dry and wet systems. Their wet model (COVEDEP) is very similar to the ADF model discussed above. They included terms for source input and for leakage and engineered safeguards (filters and sprays) removal and combined several forms of iodine in the same equations. For the ADF test conditions, where these three terms were zero, their model reduces essentially to the authors' and yields results similar to Figure 1.

British workers (13,14) have released iodine and particles into large air-filled containment vessels and correlated their results by use of an empirical "deposition velocity":

$$v_D = \frac{N_i M}{C_i} , \quad (22)$$

where v_D = deposition velocity constant, cm/sec

M = molecular weight of transported material,
g/mole

C_i = gas concentration near surface, g/cm³

The value of K_G can now be calculated by knowledge of the terms in Equation (11). N_B , T , and P are experimental measurements. R is the universal gas constant. Values for K_D can be predicted by use of the Colburn equation for heat and mass transfer analogy.⁽⁷⁾ Sherwood and Pigford (Reference 7, p. 61) show the Colburn equation as:

$$j_D = \frac{K_D P_{BM}}{U_{av} P} (Sc)^{2/3}, \quad (28)$$

where j_D = dimensionless Colburn factor

P_{BM} = log mean of pressure of inert gas at phase boundary and in bulk of fluid, atm

U_{av} = gas velocity in bulk fluid, cm/sec

P = total pressure, atm

Sc = Schmidt number = $\mu/\rho D$

μ = fluid film viscosity, g/sec cm

ρ = fluid film density, g/cm³

D = diffusion coefficient, cm²/sec.

For conditions of interest in containment studies, P_{BM} is essentially equal to P . The Schmidt number can be calculated with good accuracy for gaseous materials. For aerosols, the diffusion coefficient can be estimated by use of the Einstein particle diffusion equation.⁽¹⁶⁾ For transfer to flat plates, which is approximated in containment vessels, the bulk gas velocity, U_{av} , is the maximum velocity near the walls, as pointed out by Watson.⁽¹⁰⁾ Perry⁽⁸⁾ gives two expressions for the Colburn j_D factor: one for the laminar boundary layer region and another for the turbulent region. For the laminar region,

$$j_D = 0.664 Re_x^{-0.5}, \quad (29)$$

where Re_x is the length Reynolds number,

$$Re_x = \frac{\ell U \rho}{\mu}, \quad (30)$$

where ℓ = length, cm.

For the turbulent region, which might exist in larger vessels,

$$j_D = 0.036 \text{ Re}_x^{-0.2}$$

Some insight can be gained as to which equation applies for a particular system by assuming that transition from laminar to turbulent regions occurs at $\text{Re}_x \approx 10^5$ as suggested by Bird et al.⁽⁵⁾. For the ADF test conditions, the laminar region probably prevailed throughout the vessel.

From the foregoing, it is seen that for any given containment vessel and containment atmosphere specified, all factors required to calculate K_D are known except the bulk gas velocity. Unfortunately, attempts to measure the gas velocity in the ADF were unsuccessful because velocities were generally below the sensitivity limit (30 ft/min) of the heated thermopile anemometers used. To fit the experimental results, velocities in the range of 0.1 to 10 ft/min would have to be used in the Colburn equation.

Because of the absence of direct gas velocity measurements, the assumption was made that the velocity was proportional to the makeup steam feed rate, so that

$$K_D = k (S/A)^{0.5} \quad (32)$$

The proportionality constant, k , was determined by empirical fit of the average experimental values at low steam feed rates. A second assumption was made that some of the steam feed condensed near the inlet wall nozzle before mixing with the fission products, and the term $N_B RT/P$ was adjusted accordingly. After insertion of the appropriate ADF test condition values, Equation (11) becomes

$$K_G = 400 \frac{S}{A} + 6.0 \left(\frac{S}{A} \right)^{0.5} \quad (33)$$

Equation (33) is used in later sections as a basis for discussion of the ADF test results. It applies only to the ADF test conditions of temperature, pressure, and geometry. The validity of this equation and the more general expression of Equation (11) should be checked in experiments where gas velocity profiles can be measured.

IV. EXPERIMENTAL CONDITIONS

The ADF is located in the same building with CSE and is operated by the CSE staff. Much of the aerosol generation and injection equipment is the same as used in the large-scale CSE tests. One of the purposes of the ADF was to provide comparative experiments for confirming the adequacy of simulant aerosols.⁽¹⁷⁾ Results of ADF experiments are especially pertinent to those expected in the CSE because of the similarity of aerosol generation. Over fifty tests have been performed, seven of which were with trace-irradiated UO_2 sources. Not all of the remainder can be used in comparing results because some were made for other purposes, and conditions varied significantly from those used in the irradiated source tests. However, about 20 experiments with simulants permit a reliable comparison to be made.

EXPERIMENT STRUCTURE

The use of simulants for transport experiments is justified technically if either:

- 1) the physical and chemical properties of real and simulant materials are identical or if the effects of differences can be correlated; or
- 2) environmental conditions are transport rate controlling to the extent that differences in material properties do not affect transport significantly.

Information on material characteristics, both for simulants and "real" fission products, is being gathered at PNL and at ORNL. Since our knowledge of the physical and chemical characteristics of fission products apt to be released into containment vessels was incomplete, we chose the second approach for the present study. Tests were made using aerosols of fission products released from heated irradiated UO_2 (Series I). A second series (Series S)

used simulant aerosols. Environmental conditions within the experimental containment vessel were kept as constant as possible in both series so that any differences in transport observed would be attributable to differences in aerosol properties. This objective was only partially successful in that it was found that only slight differences in environmental conditions (notably steam condensation rate) caused significant differences in transport rate. Therefore, Series I tests should not be compared directly with Series S, but allowance must be made for variations in individual test conditions. This is possible by using the mathematical models developed in the previous section.

Steady-state atmospheric conditions were selected for all the tests reported here. This was done so that important transport measurements could be made accurately and reproduced from test to test. Model confirmation was also easily accomplished.

In order to provide more than one environmental condition for comparison, two test subseries were conducted: Subseries A with high heat transfer through the containment walls and consequent large steam condensation flux and convection currents, and Subseries B with relatively low heat transfer, steam flux, and convection.

EXPERIMENTAL EQUIPMENT

A sketch of the major experimental equipment is shown in Figure 2. Materials volatilized by inductive or resistance heating at the aerosol generation station were injected into either of two containment vessels. The PAT vessel (painted aerosol tank) was 1.54 m³ contained volume, 8.0 m² internal surface area, carbon steel painted with Phenoline 300,* a

* Carboline Co., St. Louis, Missouri

FIGURE 2. *Aerosol Development Facility Layout*

bagged separately. Lastly, the granular charcoal was poured into an 8 oz plastic jar with screw lid and shaken before analysis. Thus, each Maypack sample yielded four subsamples for analysis. Supplementary samples were taken occasionally of individual silver screens, nose cone decontaminating agents, and backup traps. Figure 4 is a schematic drawing of the Maypack. Figure 5 shows the packaged samples ready for gamma analysis.

The steam condensate was drawn periodically from the vessel sump directly into 500 ml polyethylene bottles. The sump was drawn dry at each sampling. Condensate samples were filtered, and the filters washed with 100 ml of distilled water. The filtrate and the filter were analyzed separately.

After injection was completed, the generation apparatus and aerosol delivery line to the vessel were decontaminated, and the reagent solutions were analyzed.

TEST CONDITIONS

Irradiated Source Tests

The conditions used for the seven tests with irradiated sources are listed in Table 1 (all tables are given in Appendix A). The sources were sintered natural UO_2 pellets, 95.5% theoretical density, clad in 304L SS, measuring 0.500 in. OD by 3 in. long. The weight of the UO_2 was 68.2 g; the weight of stainless steel was 24.5 g. They were irradiated to about 1×10^{17} nvt (0.35 MWd/T) and allowed to cool from 3 to 5 weeks before using. In each test one of the sources was heated to destruction in a different manner, as listed in Table 1. Temperatures were either the melting point of UO_2 or slightly above the clad melting temperature. Time at temperature was either 6 or 60 min. Heatup time from clad melting to UO_2 melting was on the order of seconds. Cooldown was rapid because of the cooling jacket on the quartz furnace; the temperature dropped

FIGURE 4. ADF Maypack (Schematic)

Neg 45959-16

FIGURE 5. Maypack Samples Prepared for Gamma Energy Analysis

to less than 1000 °C in 1.5 min. The furnace sweep gas was changed from test to test from oxidizing (air or steam-air mixtures) to reducing (pure steam or steam-hydrogen mixture). The sweep gas flow rate was constant at 28 liters (STP) per min. At the end of the heating period the furnace power was shut off, flow through the furnace continued for several minutes, then the valve on the containment vessel was closed. Gas samples were taken from the delivery line near the tank inlet during all injections. "Containment time" is defined as the time after aerosol injection started.

The containment conditions within each test series were almost the same. Prior to release, the vessel was heated to temperature with steam (the vessel had no auxiliary heaters) and permitted to reach thermal equilibrium as shown by constant temperature, pressure, and condensation rate. Steam feed was continued after injection to maintain thermal equilibrium. Usually after about one hour containment time, the steam feed rate required adjustment in order to keep isothermal conditions, and the condensation rate changed accordingly. The two rates are listed in Table 1. A slight vacuum was maintained in the vessel to prevent loss of aerosol by leakage.

The PAT tank was used in all the irradiated source runs, and no mechanical agitation was used. The paint in the PAT had been exposed to 46 hr of steaming at 80 to 100 °C prior to the first irradiated source test and had accumulated 300 hr of steam exposure at the end of the seventh test. Some blistering of the paint was noted at that time.

Simulant Tests

The simulant test conditions are listed in Table 2. Conditions varied more than in the irradiated source tests, chiefly because test objectives other than simulant verification were being pursued. Both the PAT and the SAT vessels

Figure 6 illustrates the great difference in gas phase concentration within containment for the two cases. In Run SB 63 the release was essentially instantaneous, while in Run SB 74 the release rate was constant for 1 hr. Containment conditions were nearly identical, implying that the removal rate constants ($K_G \frac{A}{V}$) were the same. That this was so is shown by the fact that the concentration half lives were the same after release was terminated. For the sake of comparison, the mass released in Run 74 has been normalized to that in Run 63. After 1 hr, the concentration was 200 times greater for the continuous release case than when the same mass was injected instantaneously. After release was terminated in Run 74, the behavior was very similar to the early portion of Run 63.

This discussion is intended to alert the reader to expected differences in containment behavior if the release term differs. It should be pointed out that although the case of a constant release rate was demonstrated, similar effects would result with nonconstant (but continuous) release rates.

Irradiated Source Tests

Table 3 lists the percentage of fission products released and the estimated mass injected to containment for the irradiated source tests. The mass released varied from a few hundredths micrograms to a few micrograms of each fission product type. About 80% of the iodine was released in 6 min, 99% in 1 hr when the UO_2 was melted. Only 75% was released in 1 hr when the temperature was held to slightly above the stainless steel clad melting point. In the latter test (Run 40), the iodine gas phase concentration was held at higher values by this continued, but unknown, release rate. Furnace atmosphere, oxidizing or reducing, did not affect the iodine release fraction.

FIGURE 6. Comparison of Elemental Iodine Gas Phase Concentration--Continuous Versus Instantaneous Release

About 65% of the cesium was released in 6 min, 95% in 1 hr when the UO_2 was melted. At clad melting temperature, only 30% was released in 1 hr. No dependence on furnace atmosphere was noted.

Tellurium was also quite volatile in all runs. About 50% was released in 6 min, 95% in 1 hr at UO_2 melting temperatures. No analysis was made for the run at clad melting temperature. Release was independent of furnace atmosphere.

Ruthenium and barium release was too low for analysis in the 6-min heating tests. Later tests used higher irradiation levels and 1 hr heating periods, and enough of these materials was released to permit complete analyses. Ruthenium release averaged 25% and barium 0.13%, with no apparent dependence on temperature or furnace atmosphere.

The deposition of each fission product in the 18-ft long, 0.93 in. ID stainless steel delivery line connecting the furnace to the containment vessel is listed in Table 3. Losses, calculated as percent of material entering the line, averaged about 17%, with ruthenium being the highest and iodine the lowest. The line was heated to approximately 140 °C. Later tests with simulants showed that fractional losses decreased with larger mass concentrations. (See Table 2.) Flow through this line was laminar at a Reynolds number of about 1200. Residence time in the tube was about 5 sec.

Simulant Tests

The details of simulant generation will not be discussed at length here. A separate report on this topic is being prepared. A brief description follows. The simulant sources were generated by volatilization of stable elements or compounds which had been traced with suitable radioisotopes for analytical purposes. Equilibration of stable and radiochemical nuclides was assured before release.

Iodine (equilibrated with ^{131}I) was prepared before the test by fusing the desired mass of KI with $\text{K}_2\text{Cr}_2\text{O}_7$ in a low flow rate helium sweep stream. Iodine released from the fusion tube was passed through a $\text{Mg}(\text{ClO}_4)_2$ drying tube into clean Pyrex U-tubes held at dry ice temperature. A charcoal backup trap caught the penetrating forms. The drying tube and backup trap were analyzed and discarded. When release was required, the U-tube was warmed and an air carrier gas flow was passed through the tube.

Cesium was released by decomposing Cs_2CO_3 (traced with ^{137}Cs) in a nickel boat at approximately 1000 °C in the presence of air. The mixture of cesium oxides and cesium metal was undoubtedly converted to cesium hydroxide particles immediately after entering the steam atmosphere either in the UO_2 melting furnace, delivery line or containment vessel.

Tellurium oxide (traced with either ^{132}Te or ^{127}Te) was prepared by the method of Stinchcombe.⁽¹⁸⁾ The oxide was heated in a nickel boat at approximately 1000 °C in air when needed. Tellurium hydroxide probably was formed when the oxide encountered a steam atmosphere.

Ruthenium oxide (traced with either ^{103}Ru or ^{106}Ru) was volatilized by heating the metal in air at approximately 1200 °C. Higher oxides were probably released but converted to the dioxide in the steam atmosphere.

Barium (traced with $^{140}\text{Ba-La}$) was released by decomposing BaCO_3 in helium on a tantalum filament at greater than 2000 °C. Tantalum at these conditions will reduce barium salts to barium metal, which is volatile at that temperature. Insignificant releases were obtained in the oxide form without reduction. Barium vapors oxidize rapidly in air and would form the hydroxide in steam.

In many of the tests, as shown in Table 2, the volatilized materials were passed through the UO_2 melting furnace and thus were re-exposed to high temperature (estimated at 1000 °C bulk gas temperature) and the presence of other vapor and particulate materials. Residence time in the hot zone of the furnace was about 0.5 sec. In some tests, as shown in Table 2, the UO_2 furnace was omitted. Stainless steel cladding was used for the majority of tests, but Zircaloy-2 was used in later runs.

The form of the iodine injected into containment was measured by sampling from the delivery line near the vessel inlet. A short 1/8 in. 304 SS tube leading to a Maypack sampled from the delivery tube centerline at isokinetic velocity. Table 4 gives the percent retained on each Maypack component. The irradiated test values are averaged as a group; the same was done for the simulant tests. Considerable differences between tests were noted, with no correlation with aerosol source or furnace atmosphere.

Two factors that affect the ratio of particulate-associated and elemental iodine are known. One is the rate of heating or release of simulant iodine, with more elemental form at high release rates. The other is the total mass released, with again more in the elemental form with higher release masses. The fraction penetrating the first three components and retained on the charcoal bed also depended on the total mass. Table 5 shows these relationships. The irradiated sources, with very low iodine mass, averaged 0.58% on the charcoal bed. This is believed to be chiefly methyl iodide. A simulant run (Run 28) with about the same iodine mass gave about the same percent on the charcoal. At higher iodine mass releases, (e.g., Run 63) the organic dropped to 0.01%. The intention in the simulant tests was not to introduce precise amounts of each form, but to determine what happened to each form after release.

Greater than 99% of the noniodine materials was always retained on the Maypack filter. The most trustworthy analytical results gave greater than 99.9% on the filter. Exceptions were noted, of course, when the filter was broken or when steam was permitted to condense on the filter. When the latter happened in a few cases, the soluble particles (Cs and Rb) were partially leached deeper into the Maypack.

CHANGES IN GAS PHASE

Of paramount importance in containment studies is the leakage rate of fission product radioactivity to the surrounding environment. This is the source term for meteorological calculations; it is the end term for containment studies. It is the product of three variables:

$$W_i = L_g C_{gi} f_p, \quad (35)$$

where W_i = efflux of species i , curies/sec

L_g = gas leakage rate, cm^3/sec

C_{gi} = concentration of i in gas, curies/ cm^3

f_p = penetration fraction, ratio activity leaving leak path to activity entering leak path.

Not much is currently known about the decontamination factor, f_p . Until proven otherwise, it must be assumed unity. Both L_g and C_{gi} are time dependent under most assumed accident conditions, and determining this dependence is a prime objective of containment studies. Leakage rate technology was discussed in considerable detail by Maccary et al.⁽¹⁹⁾ Results of air leakage tests in the CSE are in preparation. Generally the leakage rate is amenable to analysis by straight-forward engineering methods. The gas phase concentration of individual fission products is less so. Many graphs and tables of gas concentration time dependence in the ADF vessels are included in the Appendices and discussed in following sections of this report.

diminished very slowly for the duration of the test. Methyl iodide concentration remained constant with time, within experimental accuracy limits.

Iodine Form in Containment

Figures B.1 through B.21 show the gas phase iodine concentration as a function of containment time. From these, the fraction existing in each iodine form was calculated at two times (early and late) and reported in Tables 6 and 7. The percent on each Maypack component 10 min after injection is listed in Table 6. The mean of all irradiated tests and the standard deviation are shown. The same is done for the simulant tests. The reader should recall that the fraction of each iodine form depends on the total mass released. Also, it will be shown in a later section that the removal rate depends on the steam condensation rate, which varied between tests. Therefore, only the trend should be considered. Table 7 gives similar data taken after 5 hr in containment. Table 8 lists the averages in a manner convenient for showing the trend. Clearly, the particulate and elemental forms which predominated at 10 min had been removed to a large extent after 5 hr, leaving methyl iodide as the dominant form.

Mass Transfer Coefficients

Figures B.1 through B.33 were used as a basis for determining the values of the gas phase mass transfer coefficients. As discussed in the Section III, Theoretical Considerations, the slope of the curve at early time depends only on the mass transfer coefficient as long as the gas concentration remains much larger than the surface equilibrium concentration. Expressed mathematically, this is,

$$\frac{d (\ln C_G)}{dt} = - K_G \frac{A}{V} ; C_G \gg C'_G \quad . \quad (36)$$

The vessel surface area to volume ratio, A/V , was known. The surface roughness was ignored, since it should not be important where gas phase resistance is controlling. The slopes were measured directly from Figures B.1 through B.33 and listed in Tables 9 through 16 as concentration half lives. Accuracy was $\pm 15\%$. Mass transfer coefficients then were easily calculated by Equation (37).

$$K_G = \frac{0.693}{t_{1/2} A/V} \quad (37)$$

Tables 9 through 16 report measured values of K_G for total iodine, particulate iodine, elemental iodine, cesium, tellurium, ruthenium, barium, and uranium, respectively, for each run where the material of interest was included in the aerosol. Organic iodides were not included because of their high equilibrium back pressure. The data are grouped according to aerosol source type (UO_2 or simulant) and by Subseries A and B, representing high and low steam condensation rates, respectively. The means and standard deviations are listed for each group.

The steam flux is listed for each run. Where it changed during the measurement period, two values are listed. The theoretical values of K_G as calculated by Equation (33) are also shown. Finally, the ratios of the observed to calculated values are listed.

The dependence of K_G on the steam flux is shown more clearly in Figures B.34 through B.41, where K_G is plotted as a function of S/A on linear paper. For comparative purposes, Equations (3) and (33) are plotted as continuous lines.

The mass transfer coefficients are summarized in Table 17. The averages of each test series and aerosol source type are listed. This table shows that the measured values of K_G were

iodine per liter were measured in the condensate in the present work, where the condensate was removed periodically. It should be noted that in tests where the condensate is allowed to accumulate within the vessel, the liquid concentrations would be higher, with possibly higher values of H and attendant gas phase iodine concentration. However, in a loss-of-coolant accident to a water-cooled reactor, there will probably be sufficient water present to remove elemental iodine to below the concentration of organic forms.

DISTRIBUTION WITHIN CONTAINMENT

By mass balance on the entire containment vessel (for a given FP species) assuming instantaneous release,

$$\frac{d m_t}{dt} = - BSC_L , \quad (39)$$

where m_t = total moles of FP species within vessel. The right term is simply the mass leaving the vessel in the condensate, which was periodically withdrawn from the sump in the experiments. The assumption is made that material in the sump condensate was not involved in further transport processes.

A mass balance within the vessel gives

$$BSC_L + \frac{d m_g}{dt} + \frac{d m_\ell}{dt} + \frac{d m_s}{dt} = 0 , \quad (40)$$

where m_g = moles in gas phase

m_ℓ = moles in liquid film

m_s = moles reacted with solid surfaces

Equation (40) simply states that the fission product must be distributed between four locations: the condensate in the sump, the gas phase, the liquid film, or reacted with solid surfaces.

The time dependence of the first and second terms in Equation (40) was measured experimentally. Typical results are shown in Figures B.42 through B.52, where the first two terms of the equation are shown as curves labeled "in condensate sump" and "in gas phase," respectively. The third and fourth terms were not measured but were lumped and estimated together by using Equation (40). The combined term is plotted as a broken line labeled "on surfaces" in the figures and represents both the material in the liquid film adhering to the vessel surfaces and the amount reacted with the solid surfaces. It should be noted that the total fission product or simulant mass is represented. For example, total iodine is shown, not elemental or particulate only. The material balance was closed at the termination of the test by decontamination of the surfaces. What was not recovered was considered to have reacted with the paint or stainless steel.

The six aerosol materials used in the ADF tests (I, Cs, Te, Ru, Ba, U) can be divided into two groups according to the manner in which they were distributed within containment with time. All were injected into the gas phase, and all showed an initial rapid transfer from the gas to the liquid film covering the vessel surfaces. Differences in water solubility caused a division in the ensuing transport. Iodine and cesium, highly soluble in the condensate film, were transported by the flowing liquid film to the sump and were withdrawn from the vessel. The other materials (Te, Ru, Ba, U) accompanied the liquid film flow to only a slight extent. Filtering the sump effluent showed these latter materials to be largely filterable. It is unlikely that these particulate materials reacted with the solid surfaces of the vessel interior, but rather were held by physical forces. The liquid film, averaging only ~ 0.01 cm (100μ) thick, flowed at an average velocity of 0.5 cm/sec for the Series A runs and

0.07 cm/sec for Series B runs. This is in the laminar region, and viscous drag on particles attached to the wall would be small. Chapman⁽²³⁾ discussed flowing films in uniform steam condensing systems on the basis of mass and momentum transfer and showed that the average film thickness was proportional to the vertical height to the one-fourth power. The average film velocity is proportional to the one-half power of the height.

This suggests that liquid film characteristics are not strongly affected by vertical wall height. For example, at the same uniform condensing steam flux, the average film thickness in a large power reactor containment vessel would be only about twice that in the ADF vessels and film velocity about four times that in the ADF vessels.

Figures B.42 through B.52 show that there were no significant differences between simulant and real fission product behavior in the manner in which they were transported by liquid film flow. Steam flux did affect the relative positions of the curves, as predicted by the ADF model and as shown by differences between Series A and B tests.

Tables 20 through 25 list the percentage distribution between gas phase, condensate sump, and vessel surfaces (liquid film plus solid surfaces) at the end of the 5 hr containment period. Corrections were made to account for the amount sampled. The reader should recall that at 5 hr the gas-borne iodine was largely organic forms (see Table 8), while the last two columns in Table 20 are largely nonorganic forms. Also, the distribution depends on the condensation rate, with more retained on the vessel walls at low steam fluxes.

In obtaining data such as listed in Tables 20 through 25, sample analytical accuracy is very important. Higher confidence is placed on the analyses of simulant samples both because of the simpler gamma energy spectra and because the initial aerosol activity was sufficiently high to permit accurate analyses at long containment times.

Tables 20 and 21 show that Run IA 40 gave anomalous results for iodine and cesium. These two materials were held up on the vessel surfaces to a much greater extent in this run than in any other. It is possible that condensate sample analyses were in error for this test, which would give the reported high value on the vessel surfaces. The aerosol source for this test was irradiated UO_2 heated to 1500 to 1700 °C for 1 hr, and it is conceivable that the normally soluble iodine and cesium were occluded within insoluble UO_2 -stainless oxide particles during the slow, low-temperature release. This point should be studied by further tests. It should be noted, however, that the removal rate from the gas phase was not anomalous.

Table 26 lists the average percent distribution and standard deviation between tests for each isotope. Comparison of irradiated source tests with simulant tests shows the two agree within one standard deviation for the percent in the sump and on the surfaces. Differences in the gas phase are to be expected for iodine because of the different fractions of organic iodine released, as shown in Table 8. Differences between irradiated and simulant noniodine fission products in the gas phase may not be as great as indicated in Table 26 because the analytical error in the irradiated source samples was large for the very low concentrations measured after 5 hr containment time. Analyses of simulant test samples were more accurate because sufficient activity was used to allow accurate measurements after five orders of magnitude decrease in gas concentration.

FORM IN CONDENSATE

Samples of the condensate withdrawn from the vessels at various times were filtered and the filter and filtrate were analyzed separately. The filters were washed with 100 ml of demineralized water. Tables 27 and 28 list the percentage removed by filtering, which is an indication of the materials'

water solubility. Several factors preclude high confidence in these analyses. First, the material in the sump may be more soluble than that remaining on the vessel walls. That is, the sump contains material already washed off the walls, suggesting that remaining materials may be less soluble. Second, some materials were present in very low amounts and presence of other gamma emitters lowered analytical accuracy. Third, a change in procedure resulted in the irradiated source samples being filtered through cellulose acetate membranes (Gelman GM-4, 0.8μ pore size) while fibrous filters (Gelman Type A) were used for simulants. The membrane filters are more efficient for filtering liquids with small particles. In spite of these experimental problems, the trend is clear: iodine and cesium were very soluble, barium and tellurium slightly soluble, and ruthenium and uranium very insoluble.

One test (IA 40) gave results indicating that iodine and cesium were fairly insoluble for that run. This was the test where irradiated UO_2 was held at SS-clad melting temperature for 1 hr. It is conceivable that iodine and cesium were occluded within insoluble Fe-Ni-Cr-U particles and only slowly leached out. The clad- UO_2 fume should have been rich in stainless steel oxides at that temperature, compared with all other tests at the UO_2 melting point where UO_2 is more volatile.

In a few tests, the condensate was heated to boiling and sparged with air to determine whether the iodine could be released. In no case was there a detectable loss of iodine. Samples left open to the atmosphere for several days also did not lose detectable amounts of iodine.

The filtered condensate was extracted with benzene, then the benzene fraction was back-extracted with caustic solution. Table 29 shows a result for two times: an early sample and a later one. Extraction was performed 13 min after the sample

was taken. It appears that iodine in the early condensate was greater than 99% ionic, 0.02% organic, and approximately 0.2% elemental, with later samples having somewhat higher fractions of organic and elemental. Second extractions made on aliquots that were allowed to stand 2 to 3 hr showed no changes. This showed that, once in the condensate, no change of forms occurred.

OVERALL MATERIAL RECOVERY

Obtaining good material balances in containment experiments is difficult because the material is spread over large areas, some of which are impossible to decontaminate without destroying the surface. The painted aerosol tank, for example, had 8 m² of phenolic paint on its interior. The method used to decontaminate the paint was long-term steaming that, since the surfaces were all condensing, gave a washing action. Chemical reagents were not used in order not to change the surface characteristics from test to test.

Table 30 gives the material recovery, expressed as percent of material injected to the containment vessel that was accounted for by analysis. Good recovery of the soluble materials iodine and cesium was obtained. The insoluble materials tellurium, ruthenium, and barium gave poor recovery, since they were left, to a large extent, on the vessel surfaces. It should be noted that percent accounted for would be higher if the calculation had been based on the amount heated rather than that injected.

Table 30 shows that, in general, better material balances were obtained with the simulant materials than with the irradiated. This is partly due to more accurate analyses and partly because some of the simulant tests were conducted in the stainless steel vessels, which was more easily decontaminated. In the authors' opinion, the inability to close the material balance at 100% does not affect the conclusions made in this report.

MISCELLANEOUS OBSERVATIONS

Effect of Mass Concentration

The mass of iodine injected into containment was varied from one microgram (Run SA 28) to 10^5 microgram (Run SB 63). No effect on transport behavior was discernible for any given iodine form. However, the fraction of each iodine form injected did depend on total mass. This is shown in Table 8. Low mass injections gave high organic iodine fractions. The irradiated UO_2 source tests, which were low mass releases, fit the trend very well.

Noniodine aerosol mass injections were not varied appreciably in these tests, except that the simulants were about 10^3 higher than the irradiated source materials. No mass effect was observed.

The ratio of UO_2 to fission product or simulant material heated was varied over a large range. The trace-irradiated sources contained about 1.5×10^8 g UO_2 per gram of iodine. In Run SA 28 the ratio was 7×10^7 , in most of the runs it was approximately 7×10^4 , and in Run SB 63 it was 7×10^2 . The ratio for 10^4 MWd/T irradiated fuel is 1.5×10^4 . No effect on transport behavior was noted.

Effect of Aerosol Complexity

The aerosol released from heated irradiated UO_2 contained the full fission product spectrum. Most of the simulant tests used only one or two components. Table 31 compares the transport behavior of iodine in a simple aerosol consisting of iodine plus SS- UO_2 fume with the average of the irradiated tests and Run SB 53, which had five simulated components plus SS- UO_2 . The agreement is excellent, indicating that a complex aerosol is not required for experiments on iodine transport in containment systems with conditions similar to those used in the ADF tests.

Effect of UO₂ Cladding Material

The cladding used in all the irradiated UO₂ tests was 304 SS. Likewise, all but one simulant test used stainless steel cladding. In Run SB 64, Zircaloy-2 cladding was used. Table 32 compares the transport behavior of iodine in this single test with the average of three comparable simulant tests that used stainless steel cladding. The agreement is good, with no result for the Zircaloy test being outside the range of the stainless steel tests.

Fresh Versus Aged Paint

The carbon steel PAT vessel was coated with Phenoline* 300 orange primer and two Phenoline 300 gray cover coats about three months prior to its first use in a steam atmosphere. Table 33 compares the significant measurements for the first test with two later tests. All conditions were nearly identical except paint age.

The only significant difference seen is that more iodine stayed on the vessel walls during the first test (27% versus 7%). This is substantiated by the lower material recovery (50% versus 100% and 94%) and suggests that the reaction rate of iodine in the condensate film with the fresh paint was higher than with paint previously exposed to steam and low concentrations of iodine. This is in agreement with the work of Genco et al.,⁽²⁴⁾ who postulate a second-order irreversible reaction of iodine on reactive sites in the paint. After the first test there was no further significant change. This is reasonable if only the surface of the paint reacts. Diffusion of iodine to the interior of the paint would be very low in the ADF tests where the concentration of iodine in the steam condensate film decreased very rapidly.

* *Modified phenolic coating, Carboline Company, St. Louis, Missouri.*

It is important to note that no effect can be seen on organic iodine behavior. Methyl iodide was neither formed nor destroyed, within the detection limits of these experiments.

Sampling Through Long Lines

All gas samples reported were taken by Maypacks inserted directly into the containment atmosphere. The steam, air, and aerosol mixture passed directly into the Maypack without traversing an inlet tube or check valve. A few samples were taken through a 20-ft long tube to determine the error introduced by the inlet line. The tube was 0.43 in. ID 304 SS with three 90° bends of 3 in. radius. The line contained one 1/2 in. ball valve. It was electrically heated to slightly above containment atmosphere temperature, as was the Maypack on the downstream end. The flow rate was 0.5 scfm of air, the same as used in the normal sampling procedure. The Reynold's number through the tube was 1600, indicating streamline flow but near the turbulent region.

Table 34 tabulates some typical results along with the normal sample taken concurrently. The agreement is surprisingly good, suggesting that long sampling lines can give usable results if condensation is prevented and the flow is kept in the high streamline region.

Effect of Agitation

As the tests progressed it became apparent that agitating the atmosphere inside the containment vessel had a significant effect on transport from the gas phase to the surfaces. This should not have been surprising in view of the discussion on diffusional transport in Section III. When the agitator was run, the gas velocity past the vessel surfaces was increased, thus increasing the diffusional mass transfer coefficient, K_D (Equation 11). In Subseries A tests (high steam condensation

rates) the effect of agitation would be less noticeable because the bulk fluid flux term, $N_B RT/P$, was much greater than K_D . This can be seen by comparing tests SA 21 and SA 23. Both tests were conducted under essentially duplicate conditions except the agitator was run in SA 21 for the first 43 min; it was not run in SA 23. Table 9 shows that the ratio of measured overall mass transfer coefficient, K_G , to that predicted by the ADF model [Equation (36)] was only 12% higher for the case where the agitator was run.

In Subseries B tests, where the steam condensation rate was low, the effect of agitation was more apparent. In most cases where the agitator was run, K_G was higher than the ADF model predicted.

One test was made where the agitator was run for 27 min, then stopped, then run for the next 153 min, then stopped. Although this test (SO 66) was conducted in an ambient air atmosphere, the rate of decrease of gas phase concentration due to transport to the dry vessel surfaces should reflect the differences in gas velocity caused by agitation. Figure 7 shows the results. Transfer rate out of the gas phase was definitely higher during the times that the agitator was running. Inertial effects prevented sharp time changes in gas velocity, but the effect is clearly displayed.

Tables 1 and 2 show which tests used the agitator. None of the irradiated source tests (Series I) used the agitator. In about half of the simulant tests, the agitator was run, usually only during the sampling period.

The net effect of the inconsistency of agitator usage on comparing irradiated with simulant aerosol behavior in this report is that even closer agreement would have been obtained had the agitator usage been consistent.

FIGURE 7. Effect of Agitation on Elemental Iodine Gas Phase Concentration

VI. ACKNOWLEDGEMENTS

Many members of the staff of the Reactor Safeguards Experiments Unit Staff contributed to the success of this investigation by obtaining and installing the equipment, performing the tests, analyzing the samples, and correlating the results. The authors especially wish to recognize M. F. Dreher, J. L. Manuel, D. A. Nelson and R. M. Tanner, Jr. for their enthusiasm and consistent attention to experimental details. Professor J. G. Knudsen of Oregon State University provided valuable assistance in the development of the theoretical model.

VII. NOMENCLATURE

$\bar{C}$	C/C_0 , dimensionless gas phase concentration
C	concentration, mole/cm ³
C_0	concentration at $t = 0$, mole/cm ³
$\bar{T}$	Dt/a^2 , dimensionless time
D	diffusion coefficient, cm ² /sec
a	vessel characteristic dimension, cm
t	time after release, sec
N_i	fission product flux toward surface, mole sec ⁻¹ cm ⁻²
N_S	steam flux toward surface, mole sec ⁻¹ cm ⁻²
X_i	mole fraction of fission product in bulk gas phase
K_D	mass transfer coefficient for diffusion through gas boundary layer cm/sec
A	vessel surface area, cm ²
C_G	gas phase concentration, mole/cm ³
C_G	gas phase concentration in equilibrium with liquid, mole/cm ⁻³
V	vessel volume, cm ³
R	gas constant, cm ³ atm/mol °K
T	temperature, absolute, °K
P	total pressure, absolute, atm
H	C'_G/C_L , dimensionless gas-liquid partition coefficient
C_L	concentration in liquid film, mole/cm ⁻³
K_G	overall gas phase mass transfer coefficient, cm sec ⁻¹
S	steam condensation rate, cm ³ (liquid) sec ⁻¹
R_L	liquid-solid reaction rate constant, cm sec ⁻¹
B	C'_L/C_L , dimensionless ratio of liquid film to sump concentration
C'_L	sump concentration, mole cm ⁻³
L	volume of liquid film on surfaces, cm ³
v_D	deposition velocity, cm/sec
M	molecular weight, g mole ⁻¹

NOMENCLATURE (contd)

∇C_i ,	gas phase concentration gradient across boundary layer, mole cm^{-3} cm^{-1}
j_D ,	dimensionless Colburn factor
P_{BM} ,	log mean pressure of inert gas at phase boundary and in bulk fluid, atm
U_{av} ,	gas velocity in bulk fluid, cm sec^{-1}
S_c ,	Schmidt number, $\mu/\rho D$
μ ,	fluid film viscosity, $\text{g sec}^{-1} \text{cm}^{-1}$
ρ ,	fluid film density, g cm^{-3}
$Re =$	$\ell U_{av} \rho/\mu$, dimensionless Reynold's number
ℓ ,	characteristic length, cm
k_r ,	aerosol release rate, g mole sec^{-1}
m_t ,	total mass of FP species, mole
m_g ,	mass of FP species in gas phase, mole
m_ℓ ,	mass of FP species in liquid film, mole
m_s ,	mass of FP species reacted with surfaces, mole
N_B ,	flux of bulk gas to walls, $\text{mole sec}^{-1} \text{cm}^{-2}$

VIII. REFERENCES

1. G. J. Rogers. Program for Containment Systems Experiment, HW-83607, September 1964.
2. R. K. Hilliard and J. D. McCormack. "Fission Product Simulation in the Containment Systems Experiment," paper 27, International Symposium on Fission Product Release and Transport, Oak Ridge, April 5-7, 1965, CONF-650407, pp. 588-602.
3. B. M. Johnson. Containment Systems Experiment, Part III, Mathematical Models of Pressure-Temperature Transients, BNWL-233, Pacific Northwest Laboratory, Richland, Washington. May 1966.
4. N. P. Wilburn and L. D. Coffin. The Combination of On-Line Analysis with Collection of Multicomponent Spectra in A PDP-7, BNWL-CC-700, Pacific Northwest Laboratory, Richland, Washington. July 1966.
5. R. B. Bird, W. E. Stewart, and E. N. Lightfoot. Transport Phenomena, John Wiley and Sons, Inc., New York. 1960. 1st Edition.
6. R. E. Treybal. Mass Transfer Operations, McGraw-Hill Book Company, Inc., New York. 1955. 1st Edition.
7. T. K. Sherwood and R. L. Pigford. Absorption and Extraction, McGraw-Hill Book Company, Inc., New York. 1952. 2nd Edition.
8. J. H. Perry. Chemical Engineer's Handbook, McGraw-Hill Book Company, Inc., New York, 1950, pp. 540-547. 3rd Edition.
9. W. B. Cottrell. Nuclear Safety Program Semiannual Progress Report for Period Ending June 30, 1965, ORNL-3843, Oak Ridge National Laboratory, pp. 117-123. September 1965.
10. G. M. Watson, et al. Effects of Containment Size on Fission Product Behavior, ORNL-4033, Oak Ridge National Laboratory. January 1967.

REFERENCES (contd)

11. D. L. Morrison et al. An Evaluation of the Applicability of Existing Data to the Analytical Description of A Nuclear-Reactor Accident, BMI-1779, pp. 155-196. August 1966.
12. D. L. Morrison et al. An Evaluation of the Applicability of Existing Data to the Analytical Description of A Nuclear-Reactor Accident, Quarterly Progress Report for October through December, 1966, BMI-1790. January 1, 1967.
13. W. I. Megaw and F. G. May. The Behavior of Iodine Released in Reactor Containers, AERE-R3781. July 1961.
14. J. F. Croft and R. S. Iles. Experimental Release of Radioiodine in the Zenith Reactor Containment, AEEW-R-172. September 1962.
15. P. Goldsmith et al. The Role of Diffusiophoresis in the Scavenging of Radioactive Particles from the Atmosphere, AERE-R-4097. June 1962.
16. A. Einstein. Ann. Physik, vol. 17, p. 549. 1905.
17. J. D. McCormack and R. K. Hilliard. "Natural Removal of Fission Products Released from UO_2 Fuel in Condensing Steam Environments," paper 8, International Symposium on Fission Product Release and Transport, Oak Ridge, April 5-7, 1965. CONF-650407.
18. R. A. Stinchcombe. Generation of Tellurium Aerosols and Preliminary Filtration Experiments, AERE-M-1130. January 1963.
19. R. R. Maccary et al. Leakage Characteristics of Steel Containment Vessels and the Analysis of Leakage Rate Determinations, TID-20583. May 1964.
20. V. Griffiths. The Removal of Iodine from the Atmosphere by Sprays, AHSB (S) R 45. January 1963.
21. L. C. Watson et al. Iodine Containment by Dousing in NPD-II, CRCE-979. October 1960.
22. A. E. J. Eggleton. A Theoretical Examination of Iodine-Water Partition Coefficients, AERE-R-4887. February 1967.

REFERENCES (contd)

23. A. J. Chapman. Heat Transfer, The MacMillan Co., New York, p. 285. 1960.
24. J. M. Genco et al. Fission Product Deposition and its Enhancement Under Reactor Accident Conditions, BMI-X-10202. July 1967.

APPENDIX A
TABLES 1 THROUGH 34

TABLE 1. *Series I Experimental Conditions*^(a) (Irradiated UO₂ Sources)

Run No.	nvt, n/cm ²	Temp, ^(b) °C	Time at Temp, min	Furnace Conditions				Containment Conditions			
				Atmosphere, %				Temp, °C	Gage Pressure, in. Hg	Steam Flux	
				Air	Steam	H ₂	Ar			t < 1 hr g/sec/m ²	t > 1 hr g/sec/m ²
IA 26	5 x 10 ¹⁶	m.p.	6	100	0	0	0	70 ± 15 ^(c)	0 to -3	0.18	0.12
IA 32	1 x 10 ¹⁷	m.p.	6	0	100	0	0	82 ± 4	-1 to -4	0.45	0.32
IA 33	1 x 10 ¹⁷	m.p.	16	20	80	0	0	82 ± 4	0 to -2	0.40	0.29
IA 39	2 x 10 ¹⁷	m.p.	60	20	80	0	0	82 ± 4	-1 to -3	0.52	0.29
IA 40	2 x 10 ¹⁷	1300- 1800	60	20	80	0	0	84 ± 4	-1 to -3	0.42	0.28
IB 41	2 x 10 ¹⁷	m.p.	60	20	80	0	0	84 ± 8	0 to -2	0.035	0.035
IB 42	2 x 10 ¹⁷	m.p.	60	0	90	1	9	84 ± 8	0 to -3	0.058	0.058

(a) All series I tests made in Painted Aerosol Tank (PAT) with equimolar steam-air atmosphere. No mechanical agitation.

(b) m.p. means UO₂ was melted.

(c) Trouble with inleaking air.

TABLE 2. Series S Experimental Conditions^(a) (Simulant Sources)

Run	Aerosol Composition	Mass Injected, mg	Deposited in Deliv. Line, (b) %	Containment Conditions				Agitator Usage
				Vessel Used	Temp, °C	Steam Flux		
						t < 1 hr g/sec/m ²	t > 1 hr g/sec/m ²	
SA 21	I ₂ SS-UO ₂	0.70	0.4	PAT	80 ± 3	0.60	0.28	on 1st 43 min
SA 23	I ₂ SS-UO ₂	0.80	0.3	PAT	79 ± 2	0.34	0.30	None
SA 28	I ₂ SS-UO ₂	5 x 10 ⁻⁴	26	PAT	81 ± 1	0.30	0.30	None
SA 43	I ₂ SS-UO ₂	0.70	7.1	PAT	80 ± 3	0.27	0.27	None
SA 44	I ₂ SS-UO ₂	0.48	2.6	SAT	81 ± 3	0.24	0.28	None
SB 45	Rb ₂ O	19	2.5	SAT	86 ± 6	0.051	0.051	None
SB 46	CS ₂ O	9.3	5.3	SAT	85 ± 3	0.048	0.090	None
SB 47	TeO ₂	2.6	17	SAT	80 ± 3	0.032	0.076	None
SB 48	I ₂ SS-UO ₂	0.49	2.0	SAT	82 ± 1	0.037	0.086	None
SB 50	I ₂ SS-UO ₂	0.47	3.0	SAT	81 ± 2	0.037	0.086	During Sampling
SB 51	BaO	2.2	7.6	SAT	80 ± 1	0.074	0.074	During Sampling
SB 52	RuO ₄	1.0	45	SAT	80 ± 2	0.093	0.080	During Sampling

A.2

BNWL-581

TABLE 2. (Contd)

Run	Aerosol Composition	Mass Injected, mg	Deposited in Deliv. Line, (b) %	Containment Conditions				Agitator Usage
				Vessel Used	Temp, °C	Steam Flux		
						t < 1 hr g/sec/m ²	t > 1 hr g/sec/m ²	
SB 53	I ₂	0.91	3.3	SAT	80 ± 2	0.067	0.090	During Sampling
	CS ₂ O	0.53	11					
	RuO ₄	1.2	15					
	TeO ₂	0.73	3.0					
	BaO	0.70	5.0					
	SS-UO ₂							
SB 60	I ₂	0.70	12	SAT	80 ± 1	0.031	0.067	Steam Jet Injector
	SS-UO ₂							
SB 61	I ₂	1.35	21	SAT	82 ± 2	0.042	0.055	Intermittent
	SS-UO ₂							
SB 63	I ₂	95	0.1	SAT	81 ± 2	0.042	0.042	on entire run
	SS-UO ₂							
SB 64	I ₂	0.97	1.3	SAT	82 ± 2	0.053	0.053	on entire run
	Zr-UO ₂							
SB 74	I ₂	46.8	0.9	SAT	81 ± 2	0.045	0.045	During Sampling
SB 78	I ₂	0.87	4.3	SAT	80 ± 3	0.045	0.065	During Sampling
	Cs ₂ O	2.4	12					

(a) All Series S Tests were with equimolar steam-air containment atmosphere at -1 in. Hg gage pressure.

(b) Percent of mass entering the delivery line.

A.3

BNWL-581

TABLE 1. *Series I Experimental Conditions*^(a) (Irradiated UO₂ Sources)

Run No.	nvt, n/cm ²	Temp, ^(b) °C	Time at Temp, min	Furnace Conditions				Containment Conditions			
				Atmosphere, %				Temp, °C	Gage Pressure, in. Hg	Steam Flux	
				Air	Steam	H ₂	Ar			t < 1 hr g/sec/m ²	t > 1 hr g/sec/m ²
IA 26	5 x 10 ¹⁶	m.p.	6	100	0	0	0	70 ± 15 ^(c)	0 to -3	0.18	0.12
IA 32	1 x 10 ¹⁷	m.p.	6	0	100	0	0	82 ± 4	-1 to -4	0.45	0.32
IA 33	1 x 10 ¹⁷	m.p.	16	20	80	0	0	82 ± 4	0 to -2	0.40	0.29
IA 39	2 x 10 ¹⁷	m.p.	60	20	80	0	0	82 ± 4	-1 to -3	0.52	0.29
IA 40	2 x 10 ¹⁷	1300- 1800	60	20	80	0	0	84 ± 4	-1 to -3	0.42	0.28
IB 41	2 x 10 ¹⁷	m.p.	60	20	80	0	0	84 ± 8	0 to -2	0.035	0.035
IB 42	2 x 10 ¹⁷	m.p.	60	0	90	1	9	84 ± 8	0 to -3	0.058	0.058

(a) All series I tests made in Painted Aerosol Tank (PAT) with equimolar steam-air atmosphere. No mechanical agitation.

(b) m.p. means UO₂ was melted.

(c) Trouble with inleaking air.

A.1

BNWL-581

TABLE 2. Series S Experimental Conditions^(a) (Simulant Sources)

Run	Aerosol Composition	Mass Injected, mg	Deposited in Deliv. Line, (b) %	Containment Conditions				Agitator Usage
				Vessel Used	Temp, °C	Steam Flux		
						t < 1 hr g/sec/m ²	t > 1 hr g/sec/m ²	
SA 21	I ₂ SS-UO ₂	0.70	0.4	PAT	80 ± 3	0.60	0.28	on 1st 43 min
SA 23	I ₂ SS-UO ₂	0.80	0.3	PAT	79 ± 2	0.34	0.30	None
SA 28	I ₂ SS-UO ₂	5 x 10 ⁻⁴	26	PAT	81 ± 1	0.30	0.30	None
SA 43	I ₂ SS-UO ₂	0.70	7.1	PAT	80 ± 3	0.27	0.27	None
SA 44	I ₂ SS-UO ₂	0.48	2.6	SAT	81 ± 3	0.24	0.28	None
SB 45	Rb ₂ O	19	2.5	SAT	86 ± 6	0.051	0.051	None
SB 46	CS ₂ O	9.3	5.3	SAT	85 ± 3	0.048	0.090	None
SB 47	TeO ₂	2.6	17	SAT	80 ± 3	0.032	0.076	None
SB 48	I ₂ SS-UO ₂	0.49	2.0	SAT	82 ± 1	0.037	0.086	None
SB 50	I ₂ SS-UO ₂	0.47	3.0	SAT	81 ± 2	0.037	0.086	During Sampling
SB 51	BaO	2.2	7.6	SAT	80 ± 1	0.074	0.074	During Sampling
SB 52	RuO ₄	1.0	45	SAT	80 ± 2	0.093	0.080	During Sampling

A.2

BNWL-581

TABLE 2. (Contd)

Run	Aerosol Composition	Mass Injected, mg	Deposited in Deliv. Line, (b) %	Containment Conditions				Agitator Usage
				Vessel Used	Temp, °C	Steam Flux		
						t < 1 hr g/sec/m ²	t > 1 hr g/sec/m ²	
SB 53	I ₂	0.91	3.3	SAT	80 ± 2	0.067	0.090	During Sampling
	CS ₂ O	0.53	11					
	RuO ₄	1.2	15					
	TeO ₂	0.73	3.0					
	BaO	0.70	5.0					
	SS-UO ₂							
SB 60	I ₂	0.70	12	SAT	80 ± 1	0.031	0.067	Steam Jet Injector
	SS-UO ₂							
SB 61	I ₂	1.35	21	SAT	82 ± 2	0.042	0.055	Intermittent
	SS-UO ₂							
SB 63	I ₂	95	0.1	SAT	81 ± 2	0.042	0.042	on entire run
	SS-UO ₂							
SB 64	I ₂	0.97	1.3	SAT	82 ± 2	0.053	0.053	on entire run
	Zr-UO ₂							
SB 74	I ₂	46.8	0.9	SAT	81 ± 2	0.045	0.045	During Sampling
SB 78	I ₂	0.87	4.3	SAT	80 ± 3	0.045	0.065	During Sampling
	Cs ₂ O	2.4	12					

(a) All Series S Tests were with equimolar steam-air containment atmosphere at -1 in. Hg gage pressure.

(b) Percent of mass entering the delivery line.

A.3

BNWL-581

TABLE 3. *Fission Product Release from Irradiated Specimens*

Element	Run	Release ^(a) from UO ₂ , %	Delivered to Containment		Deposited in ^(d) Delivery Line, %
			mg ^(b)	% ^(c)	
Iodine	IA 26	(e)	0.07	72	6
	IA 32	82	0.28	68	14
	IA 33	81	0.25	65	20
	IA 39	99	0.45	89	8
	IA 40	75	0.35	67	0.2
	IB 41	99	0.42	83	13
	IB 42	96	0.42	83	12
Cesium	IA 32	65	1.8	58	11
	IA 33	64	1.6	55	15
	IA 39	99	4.7	78	12
	IA 40	30	1.0	18	30
	IB 41	93	4.3	74	12
	IB 42	94	4.5	74	10
Tellurium	IA 32	51	0.2	47	7
	IA 33	50	0.2	41	16
	IB 41	94	0.7	75	13
	IB 42	96	0.8	83	11
Ruthenium	IA 39	14	0.2	6	55
	IA 40	21	0.5	12	38
	IB 41	53	1.5	38	27
	IB 42	18	0.6	15	13
Barium	IA 39	0.22	0.06	0.18	12
	IA 40	0.13	0.03	0.07	39
	IB 41	0.07	0.02	0.05	29
	IB 42	0.10	0.02	0.06	25

(a) *Escaped from fuel-clad UO₂ and granular bed, % of inventory.*

(b) *Estimated from irradiation history.*

(c) *Percent of inventory.*

(d) *Percent of material entering delivery line.*

(e) *Insufficient analyses.*

TABLE 4. Iodine Form as Sampled from Delivery Line^(a)

Run	Maypack Distribution, %			
	Filter ^(b)	Silver Screens ^(c)	Charcoal Impregnated Paper ^(d)	Charcoal ^(e) Bed
<u>Irradiated</u>				
IA 26	29	66	5	0.2
IA 33	17	73	9	1.0
IA 39	69	10	20	0.5
IA 40 ^(f)	9	81	9	1.0
IB 41	16	80	3.4	0.6
IB 42	85	14	0.3	0.2
Average	43	49	8	0.58
(Std. Dev.)	(30)	(33)	(7)	(0.36)
<u>Simulant</u>				
SA 21	3	97	0.1	0.01
SA 23	16	84	0.4	0.08
SA 28	54	40	6	0.7
SA 43	68	30	1.7	0.4
SA 44	3	96	0.6	0.2
SB 48	85	14	0.7	0.04
SB 50	11	88	0.7	0.02
SB 53	88	10	1.3	0.01
SB 60	2	95	3	0.006
SB 61	56	43	1.3	0.01
SB 63	0.7	98	0.4	0.009
SB 64	27	70	2.8	0.03
Average	32	66	1.6	0.14
(Std. Dev.)	(33)	(34)	(1.7)	(0.22)

(a) Sampled at containment vessel inlet.

(b) Indicative of particulate-associated iodine.

(c) Indicative of elemental iodine.

(d) Collects mixture of elemental iodine and heavy organic iodides.

(e) Indicative of methyl iodine.

(f) Filter broken. Only charcoal bed results used in averaging.

TABLE 5. *Effect of Iodine^(a) Mass Loading on Maypack Distribution
(~80 °C, 1 atm pressure, equimolar steam-air, 50 ft/min
superficial velocity through Maypack, 3 min sample period)*

Iodine mass sampled, mg	<u>5.0</u>	<u>0.15</u>
% retained on filter	7.0	40.1
% retained on silver screens	92.4	58.9
% retained on charcoal paper	0.5	1.0
% retained on charcoal bed	0.1	0.05

(a) *Elemental Iodine was prepared by $K_2Cr_2O_7$ fusion method and released by heating the storage cold trap.*

TABLE 6. *Iodine Form in Containment Gas Phase After 10 min Containment Time*

<u>Run</u>	<u>Maypack Distribution %</u>			
	<u>Filter</u>	<u>Silver Screens</u>	<u>Charcoal Impreg. Paper</u>	<u>Charcoal Bed</u>
<u>Irradiated</u>				
IA 26	80	11	7	1
IA 32	73	10	14	3
IA 33	33	35	29	23
IA 39	61	11	19	9
IA 40	73	1	17	9
IB 41	73	10	10	7
IB 42	99	0.4	0.3	0.4
Average	70	11	14.0	4.6
(Std. Dev.)	(20)	(11)	(8.7)	(3.6)
<u>Simulant</u>				
SA 21	34	33	19	14
SA 23	39	58	2	0.6
SA 28	64	22	4	10
SA 43	52	47	0.3	0.3
SA 44	84	15	1	0.1
SB 48	98	1	1	0.2
SB 50	98	1	0.4	0.1
SB 53	85	15	0.5	0.06
SB 60	98	1	0.6	0.1
SB 61	99	0.2	1	0.1
SB 63	5	94	1	0.1
SB 64	95	0.4	4	0.6
SB 74	15	83	1.6	0.9
Average	67	28	2.8	2.1
(Std. Dev.)	(34)	(32)	(5.0)	(4.5)

TABLE 7. *Iodine Form in Containment Gas Phase After 5 hr
Containment Time*

<u>Run</u>	<u>Maypack Distribution %</u>			
	<u>Filter</u>	<u>Silver Screens</u>	<u>Charcoal Impreg. Paper</u>	<u>Charcoal Bed</u>
<u>Irradiated</u>				
IA 26	0.3	4	37	58
IA 32	0.02	0.9	7	92
IA 33	0.5	1	27	71
IA 39	2	0.5	48	49
IA 40	2	0.4	27	70
IB 41	0.2	0.5	30	70
IB 42	1	0.9	25	73
Avg. Irrad.	0.9	1.2	29	69
(Std. Dev.)	(0.9)	(1.3)	(12)	(14)
<u>Simulants</u>				
SA 21	0.03	10	9	81
SA 23	1.0	15	11	73
SA 28	0.2	4	12	84
SA 43	0.2	2	19	79
SA 44	1.4	6	37	56
SB 48	28	11	36	25
SB 50	3	4	32	61
SB 53	0.6	0.6	41	58
SB 60	3	3	63	31
SB 61	0.1	8	11	61
SB 63	13	7	47	33
SB 64	2	2	66	30
SB 74	2	5	28	65
Avg. Sim.	4.2	6.0	32	58
(Std. Dev.)	(7.9)	(4.8)	(19)	(20)

TABLE 8. Effect of Mass Released on Iodine Form

Conditions of Sampling	Iodine Mass Released, μg	Percent Retained on Maypack Component			
		Filter	Silver Screens	Charcoal Paper	Charcoal Bed
<u>Delivery Line</u>					
<u>During Injection</u>					
Irradiated, Avg.	~0.5	43 ± 30	49 ± 33	8 ± 7	0.58 ± 0.36
Simulant, Run 28	0.5	54	40	6	0.7
Simulant, Avg.	10 ³	32 ± 33	66 ± 34	1.6 ± 1.7	0.14 ± 0.22
Simulant, Run 63	10 ⁵	0.7	98	0.4	0.01
<u>Containment After</u>					
<u>10 min</u>					
Irradiated, Avg.	~0.5	70 ± 20	11 ± 11	14 ± 9	4.6 ± 3.6
Simulant, Run 28	0.5	64	22	4	10
Simulant, Avg	~10 ³	67 ± 34	28 ± 32	2.8 ± 5.0	2.1 ± 4.5
Simulant, Run 63	10 ⁵	5	94	1	0.1
<u>Containment After</u>					
<u>300 min</u>					
Irradiated, Avg	~0.5	0.9 ± 0.9	1.2 ± 1.3	29 ± 12	69 ± 14
Simulant, Run 28	0.5	0.2	4	12	84
Simulant, Avg	10 ³	4.2 ± 7.9	6 ± 4.8	32 ± 19	58 ± 20
Simulant, Run 63	10 ⁵	13	7	47	33

A.9

BNWL-581

TABLE 9. Gas Phase Mass Transfer Coefficients--Total Iodine

Run	Steam Flux	Half Life, min	Kg, cm/sec		K _G Measured
	x 10 ⁶ , g/sec-cm ²		Measured	Eq (33)	K _G Eq (33)
<u>Irradiated</u>					
IA 26	18	10	0.022	0.033	0.67
IA 32	45	5	0.045	0.058	0.78
IA 33	40	4.5	0.049	0.054	0.91
IA 39	52	4	0.056	0.063	0.89
Series IA Avg (Std. Dev.)	39 (13)		0.043 (0.015)	0.052	0.83
IB 41	3.5	16	0.014	0.012	1.2
IB 42	5.8	13	0.017	0.017	1.0
Series IB Avg (Std. Dev.)	4.65 (1.6)		0.0155 (0.0021)	0.0145	1.1
<u>Simulant</u>					
SA 21	52	3.1	0.071	0.063	1.12
SA 23	34	4.5	0.049	0.049	1.00
SA 28	30	5	0.045	0.045	1.00
SA 43	27	4	0.056	0.042	1.33
SA 44	24	4.5	0.045	0.039	1.15
Series SA Avg (Std. Dev.)	33.4 (11)		0.053 (0.011)	0.0476	1.12
SB 48	3.7	22	0.009	0.0125	0.72
SB 50	3.7	17	0.012	0.0125	0.96
SB 50	8.6	11	0.018	0.021	0.86
SB 53	6.7	7.5	0.027	0.018	1.50
SB 60	3.1	10.5	0.019	0.011	1.73
SB 61	4.2	14	0.014	0.0135	1.04
SB 63	4.2	5.5	0.037	0.0135	2.74
SB 64	5.3	15	0.013	0.016	0.81
SB 74	4.5	6.5	0.031	0.0145	2.20
SB 78	6.5	9.0	0.022	0.018	1.22
Series SB Avg (Std. Dev.)	5.05 (1.8)		0.0202 (0.0092)	0.0151	1.38

A.10

BNWL-581

TABLE 10. Gas Phase Mass Transfer Coefficients--Particulate Iodine

Run	Steam Flux	Half Life, min	K _G , cm/sec		K _G Measured
	$\times 10^6$, g/sec-cm ²		Measured	Eq (33)	K _G Eq (33)
<u>Irradiated</u>					
IA 26	18	10	0.022	0.033	0.67
IA 32	45	4.2	0.053	0.058	0.92
IA 33	40	4.2	0.053	0.054	0.98
IA 39	52	5	0.045	0.063	0.72
IA 40	28	4.5	0.049	0.043	1.14
Series IA Avg (Std. Dev.)	36.6 (±13)		0.0444 (0.0128)	0.050	0.89
IB 41	3.5	16	0.014	0.012	1.16
IB 42	5.8	13	0.017	0.017	1.00
Series IB Avg (Std. Dev.)	4.65 (± 1.6)		0.0155 (0.0021)	0.0145	1.08
<u>Simulant</u>					
SA 21	52	4.2	0.053	0.063	0.85
SA 23	34	4.5	0.049	0.049	1.00
SA 28	30	5	0.045	0.045	1.00
SA 43	27	4	0.055	0.042	1.31
SA 44	24	4.2	0.047	0.039	1.20
Series SA Avg (Std. Dev.)	33.4 (11)		0.0498 (0.0040)	0.0476	1.04
SB 48	3.7	22	0.009	0.0125	0.72
SB 50	3.7	17	0.012	0.0125	0.96
SB 50	8.6	10	0.022	0.021	1.04
SB 53	6.7	7.3	0.027	0.018	1.50
SB 60	3.1	10.3	0.019	0.011	1.72
SB 61	4.2	14	0.014	0.0135	1.04
SB 63	4.2	21	0.009	0.0135	0.67
SB 64	5.3	14.5	0.014	0.016	0.88
SB 74	4.5	9.5	0.021	0.0145	1.45
SB 78	6.5	8.5	0.023	0.018	1.28
Series SB Avg (Std. Dev.)	5.05 (±1.8)		0.0170 (0.0062)	0.0151	1.13

A.11

BNWL-581

TABLE 11. Gas Phase Mass Transfer Coefficients--Elemental Iodine

Run	Steam Flux x 10 ⁶ , g/sec-cm ²	Half Life, min	K _G , cm/sec		$\frac{K_G \text{ Measured}}{K_G \text{ Eq (33)}}$
			Measured	Eq (33)	
<u>Irradiated</u>					
IA 26	18	7.0	0.031	0.033	0.94
IA 32	45	6.7	0.033	0.058	0.57
IA 33	40	4.5	0.049	0.054	0.91
IA 39	52	4.5	0.049	0.063	0.78
IA 40	<u>28</u>	<u>6.5</u>	<u>0.034</u>	<u>0.043</u>	<u>0.79</u>
Series IA Avg (Std. Dev.)	36.6 (13)		0.0392 (0.0090)	0.0502	0.80
IB 41	3.5	10	0.022	0.012	1.84
IB 42	<u>5.8</u>	<u>13</u>	<u>0.015</u>	<u>0.017</u>	<u>0.88</u>
Series IB Avg (Std. Dev.)	4.65 (1.6)		0.0185 (0.0049)	0.0145	1.36
<u>Simulant</u>					
SA 21	52	3.0	0.074	0.063	1.17
SA 23	34	4.0	0.055	0.049	1.12
SA 28	30	4.5	0.049	0.045	1.09
SA 43	27	3.5	0.063	0.042	1.50
SA 44	<u>24</u>	<u>4.0</u>	<u>0.050</u>	<u>0.039</u>	<u>1.28</u>
Series SA Avg (Std. Dev.)	33.4 (11)		0.0582 (0.0104)	0.0476	1.23
SB 48	3.7	15	0.013	0.0125	1.04
SB 50	3.7	17	0.012	0.0125	0.96
SB 53	6.7	6	0.033	0.0195	1.83
SB 60	3.1	7.5	0.027	0.0110	2.46
SB 61	4.2	12	0.017	0.0135	1.26
SB 63	4.2	5	0.040	0.0135	2.96
SB 74	4.5	5	0.040	0.0145	2.76
SB 78	<u>6.5</u>	<u>8</u>	<u>0.025</u>	<u>0.0180</u>	<u>1.39</u>
Series SB Avg (Std. Dev.)	4.58 (1.3)		0.0230 (0.010)	0.0144	1.60

A.12

BNWL-581

TABLE 12. Gas Phase Mass Transport Coefficients--Cesium

Run	Steam Flux x 10 ⁶ , g/sec-cm ²	Half Line, min	K _G , cm/sec ^(a)		$\frac{K_G \text{ Measured}}{K_G \text{ Eq (33)}}$
			Measured	Eq (33)	
<u>Irradiated</u>					
IA 32	45	5.0	0.045	0.058	0.78
IA 33	50	5.0	0.045	0.054	0.83
IA 39	52	4.0	0.055	0.063	0.87
IA 40	<u>28</u>	<u>4.5</u>	<u>0.050</u>	<u>0.043</u>	<u>1.17</u>
Series IA Avg (Std. Dev.)	41.1 (11)		0.0488 (0.007)	0.0547	0.90
IB 41	3.5	22	0.010	0.012	0.83
IB 42	<u>5.8</u>	<u>15</u>	<u>0.015</u>	<u>0.017</u>	<u>0.88</u>
Series IB Avg (Std. Dev.)	4.65 (1.6)		0.0125 (0.0035)	0.0145	0.86
<u>Simulant</u>					
SB 45	5.1	10.5	0.021	0.016	1.30
SB 46	4.8	10.0	0.022	0.016	1.38
SB 53	6.7	7.2	0.031	0.019	1.59
SB 78	<u>4.5</u>	<u>9.7</u>	<u>0.023</u>	<u>0.018</u>	<u>1.28</u>
Series SB Avg (Std. Dev.)	5.27 (1.0)		0.0242 (0.0046)	0.017	1.40

(a) During first hour of containment time except for Runs 40 through 42 which were for the first hour after release stopped.

A.13

BNWL-581

TABLE 13. Gas Phase Mass Transport Coefficients--Tellurium

Run	Steam Flux x 10 ⁶ , g/sec-cm ²	Half Line, min	K _G , cm/sec		K _G Measured K _G Eq (33)
			Measured	Eq (33)	
<u>Irradiated</u>					
IA 32 ^(a)	45	4	0.055	0.058	0.95
IA 33 ^(a)	40	5	0.045	0.054	0.83
Series IA Avg (Std. Dev.)	42.5 (3.5)		0.050 (0.007)	0.056	0.90
IB 41 ^(b)	3.5	25	0.009	0.012	0.75
IB 42 ^(b)	5.8	14	0.016	0.017	0.94
Series IB Avg (Std. Dev.)	4.65 (1.6)		0.0125 (0.0049)	0.0145	0.84
<u>Simulated</u>					
SB 47 ^(a)	3.2	20	0.010	0.012	0.83
SB 47 ^(b)	7.6	10.5	0.021	0.020	1.05
SB 53 ^(a)	6.7	5.5	0.040	0.019	2.10
SB 53 ^(b)	9.1	9	0.022	0.022	1.00
Series SB Avg (Std. Dev.)	6.7 (2.5)		0.023 (0.012)	0.0182	1.24

(a) During first hour of containment

(b) During second hour of containment

A.14

BNWL-581

TABLE 14. Gas Phase Mass Transport Coefficients--Ruthenium

Run	Steam Flux x 10 ⁶ , g/sec-cm ²	Half Life, min	K _G , cm/sec		K _G Measured
			Measured	Eq (33)	K _G Eq (33)
<u>Irradiated</u>					
IA 39 ^(b)	29	6.0	0.037	0.044	0.84
IA 40 ^(b)	28	4.5	0.050	0.043	1.16
Series IA Avg (Std. Dev.)	28.5 (0.7)		0.0435 (0.0092)	0.0435	1.00
IB 41 ^(b)	3.5	22.5	0.010	0.012	0.83
IB 42 ^(b)	5.8	22	0.010	0.017	0.59
Series IB Avg (Std. Dev.)	4.6 (1.6)		0.010 (0)	0.0145	0.71
<u>Simulant</u>					
SB 52 ^(a)	9.3	10.5	0.021	0.0225	0.94
SB 52 ^(b)	8.0	10.5	0.021	0.0205	1.02
SB 53 ^(a)	6.7	6.5	0.034	0.0180	1.88
SB 53 ^(b)	9.0	10.0	0.022	0.0220	1.00
Series SB Avg (Std. Dev.)	8.2 (1.2)		0.0245 (0.0063)	0.0207	1.21

(a) During first hour

(b) During second hour

A.15

BNWL-581

TABLE 15. Mass Transfer Coefficients--Barium

Run	Steam Flux x 10 ⁶ , g/sec-cm ²	Half Life, min	K _G , cm/sec		$\frac{K_G \text{ Measured}}{K_G \text{ Eq (33)}}$
			Measured	Eq (33)	
<u>Irradiated</u>					
IA 39 ^(b)	29	5.5	0.040	0.044	0.91
IA 40 ^(b)	<u>28</u>	<u>4.5</u>	<u>0.050</u>	<u>0.043</u>	<u>1.16</u>
Series IA Avg (Std. Dev.)	28.5 (0.7)		0.045 (0.007)	0.0435	1.04
IB 41 ^(b)	3.5	22	0.010	0.012	0.83
IB 42 ^(b)	<u>5.8</u>	<u>20</u>	<u>0.011</u>	<u>0.017</u>	<u>0.65</u>
Series IB Avg (Std. Dev.)	4.65 (1.6)		0.011 (0.0007)	0.0145	0.74
<u>Simulant</u>					
SB 51 ^(a)	7.4	7.5	0.027	0.0195	1.38
SB 51 ^(b)	7.4	10.5	0.019	0.0195	0.98
SB 53 ^(a)	7.0	9.5	0.021	0.0190	1.10
SB 53 ^(b)	<u>9.0</u>	<u>9.5</u>	<u>0.021</u>	<u>0.022</u>	<u>0.96</u>
Series SB Avg (Std. Dev.)	7.7 (0.9)		0.022 (0.0035)	0.020	1.10

(a) During first hour

(b) During second hour

A.16

BNWL-518

TABLE 16. Gas Phase Mass Transfer Coefficients--Uranium

Run	Steam Flux x 10 ⁶ , g/sec-cm ²	Half Life, min	K _G , cm/sec		$\frac{K_G \text{ Measured}}{K_G \text{ Eq (33)}}$
			Measured	Eq (33)	
<u>Irradiated</u>					
IA 32 ^(a)	45	6.5	0.034	0.058	0.59
IA 33 ^(a)	<u>40</u>	<u>6.0</u>	<u>0.037</u>	<u>0.054</u>	<u>0.69</u>
Series IA Avg (Std. Dev.)	42.5 (3.5)		0.0355 (0.0021)	0.056	0.64
<u>Simulant</u>					
SB 61 ^(a)	4.2	21	0.0095	0.0135	0.71
SB 61 ^(b)	5.5	15	0.0133	0.0162	0.82
SB 78 ^(a)	4.5	27	0.0074	0.0145	0.51
SB 78 ^(b)	6.5	15	0.0133	0.0180	0.73
SB 79 ^(a)	3.1	29	0.0069	0.0113	0.61
SB 64 ^(a)	5.3	21	0.0095	0.0160	0.60
SB 64 ^(b)	5.3	28	0.0072	0.0160	0.45
SB 79 ^(b)	<u>6.2</u>	<u>21</u>	<u>0.0095</u>	<u>0.0173</u>	<u>0.55</u>
Series SB Avg (Std. Dev.)	5.1 (1.1)		0.0096 (0.0025)	0.0154	0.63

(a) During first hour

(b) During second hour

A.17

BNWL-581

TABLE 17. Summary of Gas Phase Mass Transfer Coefficients

Material	Series	Source	Steam Flux x 10 ⁶ , g/sec-cm ²	Avg K _G cm/sec		K _G Measured	
				Measured	Eq (33)	K _G	Eq (33)
Iodine, Particulate	IA	UO ₂	36.6	0.0444 ± 0.0128(a)		0.050	0.89
	SA	Sim.	33.4	0.0498 ± 0.0400		0.048	1.04
	IB	UO ₂	4.65	0.0155 ± 0.0021		0.0145	1.08
	SB	Sim.	5.05	0.0170 ± 0.0062		0.0151	1.13
Iodine, Elemental	IA	UO ₂	36.6	0.0392 ± 0.0090		0.050	0.80
	SA	Sim.	33.4	0.0582 ± 0.0104		0.048	1.23
	IB	UO ₂	4.65	0.0185 ± 0.0049		0.0145	1.36
	SB	Sim.	4.58	0.0230 ± 0.0100		0.0144	1.60
Cesium	IA	UO ₂	41.1	0.0488 ± 0.007		0.055	0.90
	IB	UO ₂	4.65	0.0125 ± 0.0035		0.0145	0.86
	SB	Sim.	5.27	0.0242 ± 0.0046		0.017	1.40
Tellurium	IA	UO ₂	42.5	0.050 ± 0.007		0.056	0.90
	IB	UO ₂	4.65	0.0125 ± 0.0049		0.0145	0.84
	SB	Sim.	6.7	0.023 ± 0.012		0.0182	1.24
Ruthenium	IA	UO ₂	28.5	0.0435 ± 0.0092		0.0435	1.00
	IB	UO ₂	4.6	0.010 ± 0.0000		0.0145	0.71
	SB	Sim.	8.2	0.0245 ± 0.0063		0.0207	1.21
Barium	IA	UO ₂	28.5	0.045 ± 0.007		0.0435	1.04
	IB	UO ₂	4.6	0.011 ± 0.0007		0.0145	0.74
	SB	Sim.	7.7	0.022 ± 0.0035		0.020	1.10
Uranium	IA	UO ₂	42.5	0.0355 ± 0.0021		0.056	0.64
	SB	Sim.	5.1	0.0096 ± 0.0025		0.0154	0.63

(a) One standard deviation

TABLE 18. *Comparison of K_G for Various Aerosol Components*

<u>Aerosol Component</u>	<u>K_G (cm/sec), Average of All B Series</u>
Particulate Iodine	0.016
Elemental Iodine	0.021
Cesium	0.018
Tellurium	0.018
Ruthenium	0.017
Barium	0.016
Uranium	<u>0.010</u>
Mean (Excluding Uranium)	0.0177

A.19

BNWL-581

TABLE 19. *Gas-Liquid Partition Coefficients
for Elemental Iodine*

Run	$H \times 10^7, (a)$	
	$\frac{g \text{ I}_2/cm^3 \text{ gas}}{g \text{ total iodine/cm}^3 \text{ liquid}}$	
<u>Irradiated</u>		
IA 26	3	
IA 32	0.6	
IA 33	4	
IA 39	0.6	
IA 40	4	
IB 41	12	
IB 42	0.6	
Average	3.5	
(Std. Dev.)	(4)	
<u>Simulant</u>		
SA 21	1	
SA 23	1	
SA 28	2	
SA 43	0.6	
SA 44	0.5	
SB 48	11	
SB 50	1	
SB 53	1	
SB 60	0.5	
SB 61	0.5	
SB 63	3	
SB 64	2	
SB 78	0.5	
Average	2	
(Std. Dev.)	(3)	
Theoretical, pH 7, 80 °C ^(b)		
$C_L = 10^{-5} \text{ g I}_2/\text{liter}, H = 10^{-5}$		
$C_L = 10^{-6} \text{ g I}_2/\text{liter}, H = 10^{-6}$		
$C_L = 10^{-7} \text{ g I}_2/\text{liter}, H = 10^{-7}$		
$C_L = 10^{-8} \text{ g I}_2/\text{liter}, H = 10^{-8}$		

(a) *Calculated from Figures B.1 through B.21 by
Equation (38)*

(b) *Estimated from Reference 22*

TABLE 20. *Iodine Distribution in Containment Vessel After 5 Hr*

Run	Percent of Mass Injected		
	Gas Phase	Sump	Surfaces
<u>Irradiated</u>			
IA 26	0.3	80	20
IA 32	0.4	87	13
IA 33	1	63	36
IA 39	0.3	60	40
IA 40	1	11	88
IA Average ^(a)	0.6	60	39.4
(Std. Dev.)	(0.37)	(29)	(29)
IB 41	2.3	61	37
IB 42	0.2	67	33
IB Average	1.2	64	35
(Std. Dev.)	(1.5)	(4.2)	(2.8)
<u>Simulant</u>			
SA 21	0.1	73	27
SA 23	0.04	93	7
SA 28	0.9	92	7
SA 43	0.02	93	7
SA 44	0.07	97	3
SA Average	0.23	90	10
(Std. Dev.)	(0.37)	(9.5)	(9.5)
SB 48	0.1	60	40
SB 50	0.1	81	19
SB 53	0.03	77	23
SB 60	0.05	90	10
SB 61	0.1	62	38
SB 63	0.06	90	10
SB 64	0.6	77	22
SB 74	0.05	77	23
SB Average	0.14	77	23
(Std. Dev.)	(0.18)	(11)	(11)

(a) *If Run IA 40 is omitted:*

<i>Series IA Avg</i>	<i>0.5</i>	<i>72</i>	<i>27</i>
<i>(Std. Dev.)</i>	<i>(0.33)</i>	<i>(13)</i>	<i>(13)</i>

TABLE 21. *Cesium Distribution in Containment Vessel After 5 Hr*

Run	Percent of Mass Injected		
	Gas Phase	Sump	Surfaces
<u>Irradiated</u>			
IA 32	1×10^{-3}	92	8
IA 33	2×10^{-3}	98	2
IA 39	1×10^{-3}	86	14
IA 40	1×10^{-2}	39	61
IA Average (a)	3.5×10^{-3}	78	22
(Std. Dev.)	(4.5×10^{-3})	(27)	(27)
IB 41	3×10^{-2}	82	18
IB 42	1×10^{-2}	69	31
IB Average	2×10^{-2}	75	25
(Std. Dev.)	(1.4×10^{-2})	(9.2)	(9.2)
<u>Simulant</u>			
SB 46	4×10^{-5}	76	24
SB 53	1×10^{-4}	100	0
SB 78	1×10^{-3}	87	13
SB Average	4×10^{-4}	87	13
(Std. Dev.)	(5.4×10^{-4})	(12)	(12)

(a) *If Run IA 40 is omitted:*

<i>Series IA</i>			
Avg	1.3×10^{-3}	92	8
(Std. Dev.)	(5.4×10^{-4})	(6)	(6)

TABLE 22. *Tellurium Distribution in Containment Vessel After 5 Hr*

Run	Percent of Mass Injected		
	Gas Phase	Sump	Surfaces
<u>Irradiated</u>			
IA 32	2×10^{-2}	18	82
IA 33	2×10^{-2}	30	70
IA Average	2×10^{-2}	24	76
(Std. Dev.)	(0)	(8.5)	(8.5)
IB 41	1×10^{-3}	6	94
IB 42	1×10^{-1}	2	98
IB Average	5×10^{-2}	4	96
(Std. Dev.)	(7×10^{-2})	(2.8)	(2.8)
<u>Simulant</u>			
SB 47	1×10^{-2}	19	81
SB 53	3×10^{-5}	5	95
SB Average	5×10^{-3}	12	88
(Std. Dev.)	(7×10^{-3})	(10)	(10)

TABLE 23. *Ruthenium Distribution in Containment Vessel After 5 Hr*

Run	Percent of Mass Injected		
	Gas Phase	Sump	Surfaces
<u>Irradiated</u>			
IA 39	4×10^{-5}	52	48
IA 40	1×10^{-4}	13	87
IA Average	7×10^{-5}	32	68
(Std. Dev.)	(4×10^{-5})	(28)	(28)
IB 41	6×10^{-2}	3	97
IB 42	4×10^{-2}	7	93
IB Average	5×10^{-2}	5	95
(Std. Dev.)	(1.4×10^{-2})	(2.8)	(2.8)
<u>Simulant</u>			
SB 52	5×10^{-2}	22	78
SB 53	3×10^{-4}	5	95
SB Average	2.5×10^{-2}	14	86
(Std. Dev.)	(3.5×10^{-2})	(11)	(11)

TABLE 24. Barium Distribution in Containment Vessel
After 5 Hr

Run	Percent of Mass Injected		
	Gas Phase	Sump	Surfaces
<u>Irradiated</u>			
IA 39	1×10^{-3}	(a)	(a)
IA 40	8×10^{-3}	25	75
IA Average (Std. Dev.)	4×10^{-3} (5×10^{-3})		
IB 41	2×10^{-2}	14	86
IB 42	1×10^{-2}	15	85
IB Average (Std. Dev.)	1.5×10^{-2} (7×10^{-3})	14 (1)	86 (1)
<u>Simulant</u>			
SB 51	3×10^{-4}	34	66
SB 53	3×10^{-5}	7	93
SB Average (Std. Dev.)	1.6×10^{-4} (2×10^{-4})	20 (19)	80 (19)

(a) *Insufficient analyses*

TABLE 25. Uranium Distribution in Containment
Vessel After 5 Hr

Run	Percent of Mass Injected		
	Gas Phase	Sump	Surfaces
<u>Irradiated</u>			
IA 32	<0.1	71	29
IA 33	<0.1	10	90
IA Average (Std. Dev.)	<0.1	41 (42)	59 (42)
<u>Simulant</u>			
SB 78	<0.1	3	97
SB 79	<0.1	2	98
SB Average (Std. Dev.)	<0.1	2.5 (0.7)	97.5 (0.7)

TABLE 26. *Summary of Distributions After 5 hr Containment Aging*

<u>Material</u>	<u>Series</u>	<u>Average Percent of Mass Injected</u>		
		<u>In Gas Phase</u>	<u>In Sump</u>	<u>On Surfaces</u>
Iodine	IA	0.5 ± 0.33	72 ± 13	27 ± 13
	SA	0.23 ± 0.37	90 ± 9	10 ± 9
	IB	1.2 ± 1.5	64 ± 4	35 ± 3
	SB	0.14 ± 0.18	77 ± 11	23 ± 11
Cesium	IA	(13 ± 5 x 10 ⁻⁴)	92 ± 6	8 ± 6
	IB	(2 ± 1.4 x 10 ⁻²)	75 ± 9	25 ± 9
	SB	(4 ± 5 x 10 ⁻⁴)	87 ± 12	13 ± 12
Tellurium	IA	(2 ± 0 x 10 ⁻²)	24 ± 9	76 ± 9
	IB	(5 ± 7 x 10 ⁻²)	4 ± 3	96 ± 3
	SB	(5 ± 7 x 10 ⁻³)	12 ± 10	88 ± 10
Ruthenium	IA	(7 ± 4 x 10 ⁻⁵)	32 ± 28	68 ± 28
	IB	(5 ± 1.4 x 10 ⁻²)	5 ± 3	95 ± 3
	SB	(2.5 ± 3.5 x 10 ⁻²)	14 ± 11	86 ± 11
Barium	IA	(4 ± 5 x 10 ⁻³)	25	75
	IB	(15 ± 7 x 10 ⁻³)	14 ± 1	86 ± 1
	SB	(1.6 ± 2 x 10 ⁻⁴)	20 ± 19	80 ± 19
Uranium	IA	<0.1	41 ± 42	59 ± 42
	SB	<0.1	2.5 ± 0.7	97.5 ± 1

A.25

BNWL-581

TABLE 27. *Solubility in Steam Condensate After
10 min Containment Time*

		Percent of Material in Condensate Removed by Filtering					
<u>Run</u>		<u>I</u>	<u>Cs</u>	<u>Te</u>	<u>Ru</u>	<u>Ba</u>	<u>U</u>
<u>Irradiated</u>							
	IA 26	0.5					
	IA 32	2	0.6	98			>94
	IA 33	0.7	2	98			>98
	IA 39	0.5	50		6		
	IA 40	70	75		99	90	
IA	Average	0.92(a)	17.5(a)	98	53	90	>96
	IB 41	3	3	80	99	20	
	IB 42	0.1	3	98	99	50	
IB	Average	1.6	3	89	99	35	
<u>Simulant</u>							
	SB 46		5				
	SB 47			18			
	SB 48	0.1					
	SB 50	1					
	SB 51					1	
	SB 52				>16		
	SB 53	1	1	50	97	40	
	SB 60	1					
	SB 61	0.1					
	SB 63	0.6					
	SB 64	0.2					
	SB 78		4				
SB	Average	0.6	3.3	34	>56	20	

(a) *Excluding Run IA 40*

TABLE 28. *Solubility in Steam Condensate After
5 hr Containment Time*

Run	Percent of Material in Condensate Removed by Filtering					
	I	Cs	Te	Ru	Ba	U
<u>Irradiated</u>						
IA 26	2					
IA 36	3	1	95			
IA 33	5	6	90			
IA 39	5	50		90		
IA 40	35	25		90	70	
IA Average ^(a)	3.7	19	92	90		
IB 41	5	30	80	90	30	
IB 42	1	5	88	99	20	
IB Average	3	17	84	95	25	
<u>Simulant</u>						
SB 46		1				
SB 47			8			
SB 48	0.2					
SB 50	0.03					
SB 51						
SB 52				50		
SB 53	0.3	5	50	99	80	
SB 60	0.1					
SB 61	0.5					
SB 63	0.2					
SB 64	0.04					
SB 78		0.3				
SB Average	0.2	2	30	75	80	

^(a) *Excluding Run IA 40*

TABLE 29. *Iodine Form in Condensate as Measured by Extraction*^(a)

Time of Sampling, min	Time of Extraction, min	Percent of Total		
		<u>Aqueous (b)</u>	<u>Benzene (c)</u>	<u>Caustic (d)</u>
5	18	99.78	0.024	0.20
5	175	99.76	0.020	0.22
90	103	98.11	0.19	1.7
90	196	98.15	0.18	1.67

(a) *Run 65, simulant iodine*

(b) *Probably ionic iodine*

(c) *After caustic back extraction. Probably organic iodine.*

(d) *Back extraction of benzene fraction. Probably elemental iodine.*

TABLE 30. Overall Material Recovery

Run	Mass Accounted for (Percent of Injected)				
	I	Cs	Te	Ru	Ba
<u>Irradiated</u>					
IA 26	82				
IA 32	88	100	25		
IA 33	153	99	32		
IA 39	65	93		65	104
IA 40	18	33		18	71
IB 41	92	93	34	20	31
IB 42	82	79	16	34	43
IA Average	83	83	27	34	62
<u>Simulant</u>					
SA 21	50				
SA 23	100				
SA 28	170				
SA 43	94				
SA 44	102				
SB 46		87			
SB 47			89		
SB 48	96				
SB 50	91				
SB 51					40
SB 52				109	
SB 53	88	103	23	16	20
SB 60	80				
SB 61	100				
SB 63	95				
SB 64	94				
SB 74	91				
SB Average	96	95	56	62	30

TABLE 31. *Effect of Aerosol Complexity
on Iodine Containment Behavior*

	<u>Simple (a)</u> <u>Aerosol</u>	<u>Complex (b)</u> <u>Aerosol</u>
Maypack Analysis at Vessel Inlet, % Iodine		
Filter	46	50
Silver Screens	52	43
Charcoal Paper	1.8	7
Charcoal Bed	0.2	0.6
Ratio $\frac{K_G \text{ Measured}}{K_G \text{ Calculated}}$		
Total Iodine	1.03	1.00
Particulate Iodine	1.04	1.02
Elemental Iodine	1.19	1.07
Distribution after 5 hr, %		
Gas Phase	0.2	0.7
Vessel Sump	81	63
Surfaces	19	36
H, Gas-Liquid Distribution Coefficient	2×10^{-7}	3×10^{-7}
Percent Removed from Condensate by Filtering		
at 10 min	0.4	1.1
at 5 hr	0.2	3
Overall Recovery, % of Injected	96	84

- (a) $I_2 + SS-UO_2$, average of Runs 28, 43, 44, 48, 50, and 61.
(b) Average of all Series I plus Run SB 53.

TABLE 32. *Effect of Cladding Type on Iodine Behavior*

	$\text{I}_2 + \text{Zr-UO}_2^{(a)}$	$\text{I}_2 + \text{SS-UO}_2^{(b)}$
Deposited in Delivery Line, %	1.3	8.7
Maypack Analysis at Vessel		
Inlet, %		
Filter	27	51
Silver Screens	70	48
Charcoal Paper	2.8	0.9
Charcoal Bed	0.03	0.02
Ratio $\frac{K_G \text{ Measured}}{K_G \text{ Calculated}}$		
Total Iodine	0.81	0.91
Particulate Iodine	0.88	0.91
Distribution after 5 hr, %		
Gas Phase	0.6	0.1
Vessel Sump	77	68
Surfaces	22	32
H, Gas-Liquid Distr. Coef.	2×10^{-7}	4×10^{-7}
Percent Removed from Condensate		
by Filtering		
at 10 min	0.2	0.4
at 5 hr	0.04	0.25
Overall Recovery, % of Injected	94	96

(a) *Run SB 64*

(b) *Average of Runs SB 48, SB 50, and SB 61*

A.31

BNWL-581

TABLE 33. *Comparison of Iodine Behavior in Containment Vessels with Fresh Versus Aged Painted Walls(a)*

	SA 21 Fresh Paint	SA 23 Aged Paint	SA 43 Well-Aged Paint
Previous Steam Exposure, hr ^(b)	0	24	462
Maypack Analysis at Vessel Inlet, %			
Filter	3	16	68
Silver Screens	97	84	30
Charcoal Paper	0.1	0.4	1.7
Charcoal Bed	0.01	0.08	0.4
Ratio $\frac{K_G \text{ Measured}}{K_G \text{ Calculated}}$			
Total Iodine	1.12	1.00	1.33
Particulate Iodine	0.85	1.00	1.31
Elemental Iodine	1.17	1.12	1.50
Distribution after 5 hr, %			
Gas Phase	0.1	0.04	0.02
Vessel Sump	73	93	93
Surfaces	27	7	7
H, Gas-Liquid Distr. Coef.	1×10^{-7}	1×10^{-7}	6×10^{-8}
Overall Recovery, % of Injected	50	100	94

(a) *PAT vessel, Phenoline 300 paint, Carboline Company, St. Louis, Missouri.*

(b) *Exposed to steam-air mixture at 80 to 100 °C, 1 atm.*

TABLE 34. Effect of Long Inlet Line on Sampler

Sample	Filter	Iodine Retained, %			Gas Concentration (arbitrary units)			Ratio, $\frac{C_g \text{ Remote}^{(a)}}{C_g \text{ Normal}^{(b)}}$		
		Silver Screens	Charcoal Paper	Charcoal Bed	^{131}I	^{137}Cs	^{129}Te	^{131}I	^{137}Cs	^{129}Te
<u>Irradiated</u>										
IA 26-1	73	22	4	1	1.9			1.1		
IA 26-1R(c)	80	12	7	1	2.1					
IA 32-1	72	10	15	3	6.27	1.77	4.17	1.35	1.22	1.25
IA 32-1R	92	4	2	2	8.45	2.16	5.22			
IA 32-7	0	0.9	7	92	2.07	4.3	1.3	1.02	0.28	2.23
IA 32-7R	0.4	6.0	16	78	2.11	1.2	2.9			
IA 33-4	1.4	16	40	42	1.62	11.5	18.0	1.70	0.27	0.47
IA 33-4R	0.3	10	54	36	2.77	3.05	8.7			
IA 33-7	0.5	1	27	71	1.83	1.3	11.0	0.97	0.78	0.62
IA 33-7R	0.3	10	28	62	1.77	1.0	6.8			
Average								1.23	0.64	1.14
<u>Simulant</u>										
SA 23-2	23	48	9	20	1.35			1.15		
SA 23-2R	22	16	18	44	1.55					
SA 23-5	1.3	38	11	50	3.34			1.35		
SA 23-5R	2.2	20	34	44	4.53					
Average								1.25		

(a) Remote sample was a heated Maypack with 20 ft long inlet tube connecting to containment vessel.

(b) Normal sample was a Maypack inserted directly into containment atmosphere.

(c) R signifies remote sample taken concurrently with normal sample.

APPENDIX B

FIGURES B.1 THROUGH B.52

FIGURE B.1. Iodine Gas Phase Concentration, Run SA 21

FIGURE B.2. Iodine Gas Phase Concentration, Run SA 23

FIGURE B.3. Iodine Gas Phase Concentration, Run IA 26

FIGURE B.4. Iodine Gas Phase Concentration, Run SA 28

FIGURE B.5. Iodine Gas Phase Concentration, Run IA 32

FIGURE B.6. Iodine Gas Phase Concentration, Run IA 33

FIGURE B.7. Iodine Gas Phase Concentration, Run IA 39

FIGURE B.8. Iodine Gas Phase Concentration, Run IA 40

FIGURE B.9. Iodine Gas Phase Concentration, Run IB 41

FIGURE B.10. Iodine Gas Phase Concentration, Run IB 42

FIGURE B.11. Iodine Gas Phase Concentration, Run SA 43

FIGURE B.12. Iodine Gas Phase Concentration, Run SA 44

FIGURE B.13. Iodine Gas Phase Concentration, Run SB 48

FIGURE B.14. Iodine Gas Phase Concentration, Run SB 50

FIGURE B.15. Iodine Gas Phase Concentration, Run SB 53

FIGURE B.16. Iodine Gas Phase Concentration, Run SB 60

FIGURE B.17. Iodine Gas Phase Concentration, Run SB 61

FIGURE B.18. Iodine Gas Phase Concentration, Run SB 63

FIGURE B.19. Iodine Gas Phase Concentration, Run SB 64

FIGURE B.20. Iodine Gas Phase Concentration, Run SB 74

FIGURE B.21. Iodine Gas Phase Concentration, Run SB 78

FIGURE B.22. Gas Phase Concentration, Run IA 32

FIGURE B.23. Gas Phase Concentrations, Run IA 33

FIGURE B.24. Gas Phase Concentrations, Run IA 39

FIGURE B.25. Gas Phase Concentrations, Run IA 40

FIGURE B.26. Gas Phase Concentrations, Run IB 41

FIGURE B.27. Gas Phase Concentrations During the Release Period, Run IB 41

FIGURE B.28. Gas Phase Concentrations, Run IB 42

FIGURE B.29. Gas Phase Concentrations, Simulant Cesium

FIGURE B.30. Gas Phase Concentrations, Simulant Tellurium

FIGURE B.31. Gas Phase Concentrations, Simulant Ruthenium

FIGURE B.32. Gas Phase Concentrations, Simulant Barium

FIGURE B.33. Gas Phase Concentrations, Simulant Uranium

FIGURE B.34. Mass Transfer Coefficient Versus Steam Flux, Total Iodine

FIGURE B.35. Mass Transfer Coefficient Versus Steam Flux, Particulate Iodine

FIGURE B.36. Mass Transfer Coefficient Versus Steam Flux, Elemental Iodine

FIGURE B.37. Mass Transfer Coefficient Versus Steam Flux, Cesium

FIGURE B.38. Mass Transfer Coefficients Versus Steam Flux, Tellurium

FIGURE B.39. Mass Transfer Coefficient Versus Steam Flux, Ruthenium

FIGURE B.40. Mass Transfer Coefficient Versus Steam Flux, Barium

FIGURE B.41. Mass Transfer Coefficient Versus Steam Flux, Uranium

FIGURE B.42. Iodine Distribution, Series IA

FIGURE B.43. Iodine Distribution, Series SA

FIGURE B.44. Iodine Distribution, Series IB

FIGURE B.45. Iodine Distribution, Series SB

FIGURE B.46. Cesium Distribution, Series IB

FIGURE B.47. Cesium Distribution, Series SB

FIGURE B.48. Tellurium Distribution, Series IA

FIGURE B.49. Tellurium Distribution, Series SB

FIGURE B.50. Ruthenium Distribution, Series SB

FIGURE B.51. Barium Distribution, Series SB

FIGURE B.52. Uranium Distribution, Series IA

No. of
Copies

7

Argonne National Laboratory

C. E. Dickerman

S. Fistedis

R. O. Ivins

P. Lottes

R. C. Vogel

LMFBR Program Office

A. Amorosi

L. Baker

56

Atomic Energy Commission, WashingtonAdvisory Committee on Reactor Safeguards

F. R. Fraley (17)

Division of Reactor Development and Technology

R. S. Brodsky (2)

J. W. Crawford

R. L. Ednie

H. L. Hamester

H. G. Hembree

J. A. Lieberman

R. R. Newton (5)

R. E. Pahler (2)

A. J. Pressesky

I. C. Roberts

E. E. Sinclair

S. A. Szawlewicz

M. J. Whitman

Division of Compliance

L. Kornblith, Jr.

Division of Operational Safety

H. Gilbert

Division of Production

G. B. Pleat

Division of Reactor Standards

M. Bolotsky

E. G. Case (10)

A. B. Holt

Division of Compliance, Region IV

J. W. Flora

1

Atomic Energy Commission, San Diego

R. H. Ball

No. of
Copies

1	<u>Atomic Energy Commission</u> USAEC Technical Representative H. J. Reynolds
1	<u>Atomic Energy Research Establishment,</u> Harwell, England W. J. Megan
2	<u>Atomics International</u> H. Morewitz R. Vollmer
1	<u>Auburn University</u> School of Engineering and Engineering Experiment Station G. H. Nix
2	<u>Babcock & Wilcox Co.</u> <u>Virginia</u> R. Wascher <u>Washington</u> L. R. Weissert
7	<u>Battelle Memorial Institute,</u> Columbus A. R. Duffy D. L. Morrison (2) R. L. Ritzman (1) S. Paprocki (2) D. N. Sunderman (2)
1	<u>Battelle-Frankfort</u> Frankfort, Germany N. Henzel
1	<u>Bechtel Corporation</u> R. F. Griffin
1	<u>Brookhaven National Laboratory</u> A. W. Castleman

No. of
Copies

1	<u>Canadian General Electric Company</u> Peterborough, Ontario, Canada S. Davies
1	<u>Chalk River Nuclear Laboratories</u> Chalk River, Ontario, Canada Station 3 F. C. Boyd G. Hake
1	<u>Chicago Operations Office</u> Atomic Energy Commission D. M. Gardiner
1	<u>Combustion Engineering</u> M. F. Valerino
1	<u>Consolidated Edison Company</u> J. J. Grob
1	<u>Dilworth, Secord, Meagher, and Associates</u> Ltd., 4195 Dundas St. Toronto, Ontario, Canada I. J. Billington
265	<u>Division of Technical</u> <u>Information Extension</u>
4	<u>Douglas United Nuclear</u> T. W. Ambrose N. R. Miller J. W. Riches J. R. Spink
3	<u>General Electric Company,</u> San Jose S. Vandenberg G. E. Wade E. Zebroski
4	<u>General Electric Corp</u> APD San Jose P. Bray M. Siegler W. A. Sutherland B. Wolfe

No. of
Copies

2	<u>General Atomic</u> A. J. Goodjohn
1	<u>Harvard Air Cleaning Laboratory</u> F. J. Viles
2	<u>Idaho Nuclear Corporation</u> J. A. Buckham C. M. Slansky
2	<u>Idaho Operations Office</u> <u>Atomic Energy Commission</u> D. Williams
1	<u>Idaho Nuclear Corporation,</u> <u>Idaho Falls, Idaho</u> C. M. Slansky
2	<u>ITT Research Institute</u> E. Gallagher T. A. Zaker
2	<u>Atlantic Richfield Hanford Company</u> O. F. Hill G. R. Kiel
1	<u>Los Alamos Scientific Laboratory</u> J. H. Russel, K. Division
1	<u>MPR Associates, Inc.</u> T. Rockwell III
1	<u>National Bureau of Standards</u> C. Muehlhause
1	<u>Naval Ordnance Laboratory</u> J. Proctor
1	<u>North Carolina State University</u> M. N. Ozisik
1	<u>Nuclear Fuels Services</u> R. P. Wischow

No. of
Copies

13	<u>Oak Ridge National Laboratory</u> R. E. Adams C. J. Barton R. Blanco W. E. Browning J. Buchanan W. B. Cottrell (4) D. Ferguson M. H. Fontana S. Friede C. E. Miller G. W. Parker L. F. Parsley T. H. Rowe D. B. Trauger (2) G. M. Watson
4	<u>Oak Ridge Operations Office</u> Atomic Energy Commission D. Cope (2) E. Delaney W. L. Smalley
1	<u>Oregon State University</u> James G. Knudsen
1	<u>Pacific Gas and Electric Company</u> W. Nutting
14	<u>Phillips Petroleum Company</u> G. O. Bright G. F. Brockett S. Forbes (2) C. Haire G. B. Matheny W. E. Nyer (2) F. Schroeder (3) N. K. Sowards T. R. Wilson (2)

No. of
Copies

8	<u>Richland Operations Office</u> <u>Atomic Energy Commission</u> P. G. Holsted (3) J. Kitchen C. L. Robinson R. K. Sharp A. S. Waterhouse Technical Information Library
1	<u>San Francisco Operations Office</u> <u>Atomic Energy Commission</u> Lt. Col. J. B. Radcliffe
1	<u>Savannah River Laboratories</u> A. H. Peters
2	<u>TRW Incorporated</u> <u>TRW Systems Group</u> D. B. Langmuir S. M. Zivi
2	<u>University of California</u> <u>Institute of Engineering</u> <u>Research</u> H. A. Johnson V. E. Schrock
1	<u>University of Washington</u> Wells Moulton
5	<u>Westinghouse Electric Co. (APD)</u> E. Beckjord D. Fletcher H. Graves F. M. Heck R. A. Wieseemann
75	<u>Battelle-Northwest</u> F. W. Albaugh R. T. Allemann J. M. Batch G. E. Benedict R. H. Bond L. L. Burger J. J. Cadwell

Battelle-Northwest (contd)

L. F. Coleman
D. L. Condotta
F. G. Dawson
D. R. de Halas
J. R. Fishbaugher
R. F. Foster
J. C. Fox
J. J. Fuquay
W. A. Haney
H. Harty
R. K. Hilliard (30)
J. F. Honstead
R. T. Jaske
R. L. Junkins
A. R. Keene
C. E. Linderoth
J. D. McCormack
A. J. McElfresh
J. M. Nielson
R. E. Nightingale
H. M. Parker
R. S. Paul
L. T. Pedersen
A. K. Postma
D. L. Reid
W. D. Richmond
G. J. Rogers
L. C. Schwendiman
G. A. Sehmel
D. E. Simpson
J. C. Spanner
W. C. Townsend
E. E. Voiland
R. G. Wheeler
N. P. Wilburn
N. G. Wittenbrock
M. E. Witherspoon
D. C. Worlton
Patent Section (2)
Technical Information Files (5)
Technical Publications (1)

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60
61
62
63
64
65
66
67
68
69
70
71
72
73
74
75
76
77
78
79
80
81
82
83
84
85
86
87
88
89
90
91
92
93
94
95
96
97
98
99
100
101
102
103
104
105
106
107
108
109
110
111
112
113
114
115
116
117
118
119
120
121
122
123
124
125
126
127
128
129
130
131
132
133
134
135
136
137
138
139
140
141
142
143
144
145
146
147
148
149
150
151
152
153
154
155
156
157
158
159
160
161
162
163
164
165
166
167
168
169
170
171
172
173
174
175
176
177
178
179
180
181
182
183
184
185
186
187
188
189
190
191
192
193
194
195
196
197
198
199
200
201
202
203
204
205
206
207
208
209
210
211
212
213
214
215
216
217
218
219
220
221
222
223
224
225
226
227
228
229
230
231
232
233
234
235
236
237
238
239
240
241
242
243
244
245
246
247
248
249
250
251
252
253
254
255
256
257
258
259
260
261
262
263
264
265
266
267
268
269
270
271
272
273
274
275
276
277
278
279
280
281
282
283
284
285
286
287
288
289
290
291
292
293
294
295
296
297
298
299
300
301
302
303
304
305
306
307
308
309
310
311
312
313
314
315
316
317
318
319
320
321
322
323
324
325
326
327
328
329
330
331
332
333
334
335
336
337
338
339
340
341
342
343
344
345
346
347
348
349
350
351
352
353
354
355
356
357
358
359
360
361
362
363
364
365
366
367
368
369
370
371
372
373
374
375
376
377
378
379
380
381
382
383
384
385
386
387
388
389
390
391
392
393
394
395
396
397
398
399
400
401
402
403
404
405
406
407
408
409
410
411
412
413
414
415
416
417
418
419
420
421
422
423
424
425
426
427
428
429
430
431
432
433
434
435
436
437
438
439
440
441
442
443
444
445
446
447
448
449
450
451
452
453
454
455
456
457
458
459
460
461
462
463
464
465
466
467
468
469
470
471
472
473
474
475
476
477
478
479
480
481
482
483
484
485
486
487
488
489
490
491
492
493
494
495
496
497
498
499
500
501
502
503
504
505
506
507
508
509
510
511
512
513
514
515
516
517
518
519
520
521
522
523
524
525
526
527
528
529
530
531
532
533
534
535
536
537
538
539
540
541
542
543
544
545
546
547
548
549
550
551
552
553
554
555
556
557
558
559
560
561
562
563
564
565
566
567
568
569
570
571
572
573
574
575
576
577
578
579
580
581
582
583
584
585
586
587
588
589
590
591
592
593
594
595
596
597
598
599
600
601
602
603
604
605
606
607
608
609
610
611
612
613
614
615
616
617
618
619
620
621
622
623
624
625
626
627
628
629
630
631
632
633
634
635
636
637
638
639
640
641
642
643
644
645
646
647
648
649
650
651
652
653
654
655
656
657
658
659
660
661
662
663
664
665
666
667
668
669
670
671
672
673
674
675
676
677
678
679
680
681
682
683
684
685
686
687
688
689
690
691
692
693
694
695
696
697
698
699
700
701
702
703
704
705
706
707
708
709
710
711
712
713
714
715
716
717
718
719
720
721
722
723
724
725
726
727
728
729
730
731
732
733
734
735
736
737
738
739
740
741
742
743
744
745
746
747
748
749
750
751
752
753
754
755
756
757
758
759
760
761
762
763
764
765
766
767
768
769
770
771
772
773
774
775
776
777
778
779
780
781
782
783
784
785
786
787
788
789
790
791
792
793
794
795
796
797
798
799
800
801
802
803
804
805
806
807
808
809
810
811
812
813
814
815
816
817
818
819
820
821
822
823
824
825
826
827
828
829
830
831
832
833
834
835
836
837
838
839
840
841
842
843
844
845
846
847
848
849
850
851
852
853
854
855
856
857
858
859
860
861
862
863
864
865
866
867
868
869
870
871
872
873
874
875
876
877
878
879
880
881
882
883
884
885
886
887
888
889
890
891
892
893
894
895
896
897
898
899
900
901
902
903
904
905
906
907
908
909
910
911
912
913
914
915
916
917
918
919
920
921
922
923
924
925
926
927
928
929
930
931
932
933
934
935
936
937
938
939
940
941
942
943
944
945
946
947
948
949
950
951
952
953
954
955
956
957
958
959
960
961
962
963
964
965
966
967
968
969
970
971
972
973
974
975
976
977
978
979
980
981
982
983
984
985
986
987
988
989
990
991
992
993
994
995
996
997
998
999
1000