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NATURAL TRANSPORT EFFECTS
ON FISSION PRODUCT BEHAVIOR
IN THE CONTAINMENT SYSTEMS EXPERIMENT

R. K. Hilliard and L. F. Coleman

December 1970

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R. K. Hilliard and L. F. Coleman
Fluid and Energy Systems Department
Physics and Engineering Division

December 1970

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BEHAVIOR IN THE CONTAINMENT SYSTEMS EXPERIMENT

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ABSTRACT

The results of six experiments in the 26,500 ft³ Containment Systems Experiment (CSE) containment vessel are reported. In these tests, no active safety feature operated and all transport was by natural passive processes. The time dependence of iodine, methyl iodide, cesium, ruthenium and uranium concentrations was measured at various locations in the vapor space and in the steam condensate. Parameters which were varied from test to test included the ratio of surface area to volume, temperature and pressure of the atmosphere, heat transfer rate to walls, paint age, and initial iodine concentration. Most of the tests were at steady temperature with steam feed to replace heat losses, but in one experiment the steam feed was zero and the temperature and pressure decreased with time.

The results are compared with theoretical predictions with good agreement for iodine behavior and reasonable agreement for particulate materials when suitable particle sizes are used in predictive calculations. The initial rate controlling mechanism for elemental iodine is shown to be gas phase diffusion across boundary layers. For particles, gravitational settling dominated, with turbulent deposition and diffusiophoresis being of secondary importance. Whether the temperature and pressure were held constant or allowed to decrease with time had essentially no effect on the iodine behavior; but particle deposition was significantly slower in the temperature decay case.

The best estimates of the 2-hr dose reduction factor (DRF) due to fission product removal by natural processes

for large PWR containment systems are about 2.6 for inorganic iodine, 1.15 for particulate matter and 1.0 for methyl iodide. At longer times the estimates for DRF are considerably larger.

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I. INTRODUCTION

The safety assessment of nuclear power generating stations is unique from that of conventional plants because of the radioactive nature of the fuel used in nuclear plants. If the fuel were not radioactive, potential damage caused by the worst conceivable accident would be limited to mechanical damage within the confines of the plant, as is the case with conventional plants. However, the nuclear fuel is highly radioactive and an important part of the safety assessment deals with how effectively the radioactive material will be retained within the plant building structures in event of a severe accident. A thorough understanding of the transport behavior of all the radioactive material is essential to reliable prediction of fission product behavior in the unlikely event of an accident.

The Atomic Energy Commission (AEC) and the Nuclear Power Industry have sponsored a considerable amount of work aimed at providing an understanding of the behavior of fission products during accident conditions. Much of the work has been done at laboratory scale or in small-scale facilities. However, the scaling factors are in general uncertain and extrapolation to full-sized plants is questionable. Experiments in large-scale facilities are necessary before confidence can be placed in predictions based on mathematical models.

The Containment Systems Experiment (CSE)⁽¹⁾ is a Commission-sponsored program directed toward obtaining large-scale measurements of systems response and fission product

behavior over a range of possible accident conditions. In some of the experiments in the CSE, active safety features (containment sprays or filter recirculation loops) were operated to study their performance and to verify mathematical models which can be used to predict fission product removal in full size plants. In some tests no active safeguards were operated and all fission product migration occurred solely by natural processes. This report discusses the latter tests.

A study of the behavior of fission products in the absence of active safety features is important for three reasons. First, natural removal processes occur even while active engineered devices are operated and thus should be accounted for in any prediction of overall behavior. This is especially true in small-scale experiments, where natural removal rates may approach or even exceed those due to the engineered device. Validation of the performance of the engineered device thus depends on proper accounting for removal by natural effects. Second, the performance of active safety features such as containment sprays or recirculating filter systems depends strongly on many natural processes such as mixing in the gas space, intercompartment transfer, desorption from surfaces, particle agglomeration, or chemical reactions other than those in the active system. Experimental measurement of such processes can often be done best in the absence of the engineered safety systems. The third reason for studying natural transport processes is that these processes in themselves provide a minimum decontamination rate which will not depend on design or operational reliability of active safety features.

Experiments in the CSE have been underway for over 2 years and progress has been reported in quarterly progress reports.^(2,3,4) Separate topical reports have been issued on the performance of containment sprays systems.^(5,6,7) An

earlier report⁽⁸⁾ described a theoretical model for fission product transport by natural processes and presented preliminary experimental results of CSE natural transport tests. The present report gives a complete discussion of all six experiments performed in the CSE in which only natural processes were involved.

II. SUMMARY AND CONCLUSIONS

The results of six CSE experiments (Runs D-1, D-2, A-1, A-2, A-5 and A-11) are reported in detail. In these experiments no engineered safety features were provided and all fission product transport occurred solely by natural, passive processes. The experiments were performed in two different sizes of containment vessel, the largest being about 1/5 linear scale of a large PWR containment building. This is equivalent to approximately twice that of a typical large PWR for the ratio of total surface area to volume, the most important parameter for iodine deposition.

After representative postaccident conditions were established in the vessel, fission product simulants were injected in essentially an instantaneous manner. The initial iodine concentration exceeded 100 mg/m^3 in three of the tests and was about 1 mg/m^3 in the other three. Initial particle concentrations ranged from 0.1 to 10 mg/m^3 for three types of particles: cesium, uranium (oxide) and particulate iodine. Time-dependent measurements were made for mass concentrations in the vapor space at many locations, in the condensate film on the vessel walls, and in the liquid pools which accumulated due to steam condensation. After 1 to 2 days aging, the vessel was decontaminated to determine the final distribution of simulant materials between gas, liquid and paint.

A major parameter was changed in each experiment. Two vessel sizes were used ($2,286$ and $26,500 \text{ ft}^3$). Temperatures of 180 and 250°F were investigated. Two initial iodine concentrations were used (1 and 100 mg/m^3). The effect of heat transfer rate was measured by performing tests with both uninsulated and insulated walls. Finally, the effect of unsteady temperature and pressure was determined by one test in which temperature and pressure decayed with time.

The following conclusions are based on evaluation of the experimental results presented in this report.

- The experimental values of the initial removal rate for total inorganic iodine were in good agreement with the natural convection model. The effects of surface-to-volume ratio, temperature, and heat transfer were as predicted by the model.
- The measured initial first-order removal rate constant for inorganic iodine varied from 2 to 5 hr^{-1} , averaging 5% lower than predicted assuming all surfaces to be perfect iodine sinks.
- The gas phase limited rate for inorganic iodine lasted only until the gas phase concentration decreased to about 1% of the initial value, after which the concentration continued to decrease at a much slower rate.
- The quasi-equilibrium attained a few hours after iodine release gave a gas-liquid distribution coefficient averaging 6000, with no apparent dependence on temperature, pH or iodine concentration.
- Measurements of fission product transport made in small vessels should not be extrapolated to large vessels by a simple surface-to-volume scaling factor. Vessels under 10 ft in height have laminar boundary layers, while large vessels have predominantly turbulent boundaries.
- Based on these experiments, the natural convection model predicts the 2-hr time-integrated dose reduction factor (DRF) for inorganic iodine in a typical large PWR would be about 2.6. This improves to about 15 after 1 day, with high probability of continued increase with time.
- Methyl iodide was removed slowly from the atmosphere in all tests, with an average overall decontamination factor of 1.7 after 1 day.

- Assuming 5% of the iodine released to a large PWR containment vessel to be methyl iodide and the formation and removal rates of methyl iodide to be equal, the 2-hr DRF for total iodine of all forms is estimated to be 2.5. After 1 day the DRF would increase to about 9.
- These predictions are believed to be conservative because the rapid heat and mass transfer accompanying and immediately following loss-of-coolant (blowdown) is ignored. Forced convection devices would also increase removal rates.
- Cesium and uranium existed in the containment atmosphere as particles associated with relatively large masses of sorbed water. The effective diameter of these fog droplets, as derived from gravity settling measurements, was about 10 μ for the initial 2-hr period, decreasing to about 3 μ after 1 day.
- The cesium and uranium, representing both water soluble and insoluble fission product particles, were removed from the gas space primarily by gravitational settling. Transfer to surfaces by combined diffusion and diffusiophoresis accounted for only about 10% of the total removed.
- Based on these experiments, the 2-hr DRF for all particulate aerosols in a typical large PWR is estimated to be 1.15. After 1 day it would increase to about 3.
- The concentrations of all fission product simulants were essentially uniform throughout the gas space of a single compartment. Large concentration differences between compartments persisted for several hours in these tests where no forced convection was provided.
- Temperature per se had no significant effect on fission product behavior in saturated steam-air atmospheres.

- The heat transfer rate to outer walls is an important influence on iodine transfer. The convection velocity caused by temperature gradients near the walls was effective in providing high mass transfer coefficients at all surfaces exposed to iodine.
- About 50% of the iodine and <10% of the cesium was retained by the phenolic paint and could not be removed by post-test steaming and spraying decontamination efforts.

III. BACKGROUND INFORMATION

The British were pioneers in the study of the behavior of fission products after release into containment vessels.⁽⁹⁾ Their work was largely concerned with iodine behavior in an air atmosphere. They were the first to show that iodine existed in several different forms each with different transport properties.⁽¹⁰⁾ It was their large-scale experiments which provided clues which, when followed by laboratory investigation, resulted in the identification of alkyl iodides as a small but significant fraction of the total iodine released from overheated fuel.⁽¹¹⁾ Their results using the Dido and Pluto reactor containment vessels gave results which are remarkably similar to all those performed elsewhere since then. There was a rapid initial plate out of iodine followed by a slower decrease in iodine concentration with time. The iodine remaining in the gas phase at the longer times was not removable from gas streams passed through caustic scrubbers, absolute filters, or over silver or copper surfaces. It was however, removed by charcoal beds with good efficiency. They showed that the early rapid plateout was due to elemental iodine being deposited on surfaces while the later low concentration material was largely a mixture of alkyl iodides, of which methyl iodide predominated.

At Oak Ridge National Laboratory (ORNL) a large number of experiments have been performed, in which both simulated fission products and fission products released from heated irradiated fuels were released into small vessels.⁽¹²⁾ Four different experimental facilities were used in this work. The Containment Mockup Facility (CMF), 7 ft³; the In-Pile Installation (IPI) 7 ft³; the Containment Research Installation (CRI) 135 ft³; and the Nuclear Safety Pilot Plant (NSPP), 1350 ft³. This work showed that large decontamination factors for all fission product materials except the organic iodides

are obtainable by natural transport processes. A summary report⁽¹²⁾ also concluded that "the use of simulated fission products for studies such as those conducted in the CSE appears to produce valid and realistic results which are applicable to the Nuclear Safety Program."

Experiments have also been performed in the 86 ft³ Contamination-Decontamination Experiment (CDE) at the National Reactor Testing Station, Idaho Falls, Idaho.⁽¹³⁾ The primary objective in these experiments was to demonstrate sampling equipment in support of the Loss-of-Fluid-Test (LOFT), but the information obtained on behavior in the containment atmosphere of the fission products released from short-cooled highly irradiated fuels was consistent with that from tests made elsewhere with simulants and trace-irradiated fuel.

Hilliard, Coleman and McCormack conducted a large number of containment tests with aerosols released from trace-irradiated fuel and with fission product simulants, using small vessels of 32 and 54 ft³ volume.⁽¹⁴⁾ They concluded that the simulant selected for use in the large-scale CSE experiments was a fully satisfactory substitute for actual fission products in containment mass transport tests.

Mathematical models of fission product transport and deposition in containment vessels have been proposed by several workers. Most of these have been limited to the behavior of elemental iodine, but some include transport of particles. Ritzman⁽¹⁵⁾ and Morrision⁽¹⁶⁾ published a model, COVEDEP, which is general in nature and includes terms for leakage and removal by engineered safety features. However they did not provide methods for predicting the several rate constants required. Rosenberg⁽¹⁷⁾ measured the reaction rates of iodine and methyl iodide with typical containment system surfaces.

Ozisik and Hughes⁽¹⁸⁾ used boundary layer theory for predicting molecular iodine flux to surfaces from condensing steam in the presence of noncondensable gases. Their analytical approach was limited to laminar boundaries, however, whereas large containment vessels would very likely have turbulent boundary layers during postaccident conditions.^(19,20)

Watson, et al.⁽²¹⁾ discussed some of the factors influencing mass transport in containment vessels, including vessel size. They outlined a method of predicting mass transfer coefficients in air atmospheres using empirical similarity solutions. However, for the practical case of concern under loss-of-coolant postaccident conditions where a condensing steam and air mixture prevails, they neglected the diffusive flux component, using only the Stephen Flow flux. This is a serious shortcoming for a realistic predictive method, but their discussions provide many useful relationships.

Yuill and Baston⁽²²⁾ have recently published a model for elemental iodine deposition in containment vessels in the presence of condensing steam and air which in essence uses penetration theory. One of their required input terms is the rate of steam condensation which must either be measured or estimated by a separate energy balance. Their predictions compare well with experiments in several different facilities. They also included a method for predicting particle deposition based only on diffusiophoresis and thermophoresis. Results of CSE experiments reported in a later section of this document show that these two removal mechanisms are relatively insignificant and that gravitational settling is the chief factor for particle deposition. It also appears that these authors used an incorrect expression for steam flux (flux relative to the mass average velocity instead of flux relative

to stationary coordinates), which caused their calculations to overestimate removal of particles by diffusiophoresis by a factor of up to 3.

Knudsen and Hilliard⁽⁸⁾ provided a predictive model for deposition of elemental iodine from condensing steam air atmospheres which utilized empirical correlations for heat transfer to vertical plane surfaces by natural convection. They modified the normal mass-heat transfer analogy to allow for the added convection due to increased density differences near cold walls caused by mass transfer of steam.^(23,24) Comparison of their results with experiment are discussed in a later section.

Summarizing, a considerable amount of information has become available on iodine deposition in containment vessels, and the several mathematical models recently developed appear to give accurate predictions when compared with laboratory experiments. Particle behavior in steam-air atmospheres is less well understood.

IV. EXPERIMENTAL FACILITY AND PROCEDURE

A. EXPERIMENTAL EQUIPMENT

A complete description of the CSE experimental equipment is given by Linderoth.⁽²⁵⁾ An abbreviated description is included in this report for the convenience of those who do not need detailed information. Figure 1 is a sectional drawing of the CSE vessel system, showing elevations, radii and wall thicknesses. The outer vessel encloses 30,680 ft³ of space. The drywell (DW) has a 2,286 ft³ volume. The wetwell space has 4,207 ft³ of space, but this was sealed off and not used in the present tests. The carbon steel main vessel is designed for a maximum operating condition of 75 psig at 320 °F. The drywell is carbon steel rated at 150 psig at 330 °F. The wetwell is rated at 25 psi differential in either direction. All interior surfaces are coated with a modified phenolic paint.* The top of the wetwell forms a solid deck which effectively separates the contained gas into what we term the "main room" above the deck and the lower rooms below the deck. In the present tests, the lid of the drywell was raised so that its volume was common to the main room, giving a total volume in the main room of 21,005 ft³. One third of the annular space between the drywell and the main vessel is a small access area we call the "middle room," 2,089 ft³ in volume. Below this is a third space we call the "lower room," 3,380 ft³ in volume. Two 4-ft diameter holes in the deck connect the main room to the middle room; one 4-ft opening connects the middle room to the lower room. A schematic drawing of the arrangement is shown in Figure 2.

Steam condensate accumulates in three locations: the drywell pool, the main vessel pool located in the lower room, and on the flat deck of the main room. The first two are

* Phenoline 302. The Carboline Co., St. Louis, Missouri

FIGURE 1. CSE Vessel Elevations

FIGURE 2. Schematic Drawing of Containment Vessel Arrangement Used in CSE A-Series Tests

stirred, sampled frequently and the liquid volume monitored. The liquid on the deck is not monitored. Condensate running down the main outer vessel walls is caught by a trough near the deck which completely surrounds the vessel. This condensate stream is sampled and the flow rate measured before it is discharged to the lower pool. Condensate is also taken directly from the outer wall at four elevations by troughs 2 ft wide in a vertical array at 10-ft intervals. A line from the plant steam boiler terminating near the bottom of the drywell provided the steam atmosphere. It is a 2-in. ID pipe directed upward on the centerline.

Coleman⁽²⁶⁾ has described the method of generating the fission product simulant in detail. Four materials were released in these tests to permit mass transfer measurements. They were: elemental iodine, methyl iodide, cesium, and particles formed by melting unirradiated Zircaloy-clad UO_2 . Stable elemental iodine was equilibrated with ^{131}I as described by Coleman,⁽²⁶⁾ and when release was desired, the flask containing the iodine was heated electrically and air swept through the flask, carrying the elemental iodine through the hot zone of the UO_2 melting furnace. Some particulate-associated iodine and organic iodides were always produced. In Runs A-5 and A-11 methyl iodide was purposely added through a separate line, not connected to the UO_2 furnace.

Stable cesium was equilibrated with ^{137}Cs in a nickel boat and dried at $<400^\circ\text{C}$. When release was desired, the boat was heated inductively to $\sim 1200^\circ\text{C}$ with an air stream carrying the volatilized cesium and cesium oxide through the UO_2 furnace into the containment vessel.

A steam jet located at the drywell served as a compressor for injecting the volatilized simulant into the pressurized containment vessel. The injection line was a nominal 2-in.

stainless steel pipe, electrically heated to 150 °C. Three samplers took small aliquots from the injection line during release periods.

The suitability of the fission product simulant generated by these methods to represent actual fission products in the containment environment was demonstrated by small-scale tests in the Aerosol Development Facility (ADF).⁽¹⁴⁾ Recently Roberts, et al. at Oak Ridge National Laboratory concluded an extended study of the suitability of the CSE simulant with the conclusion that the use of this simulant is warranted.⁽¹²⁾

The main gas sampling system consists of Maypack samplers arranged in clusters of twelve located at various positions throughout the gas spaces. The system is described by McCormack⁽²⁷⁾ and is depicted schematically in Figures 3 and 4. Each Maypack has its own solenoid valve, with remote operation from a nearby laboratory. Figure 5 shows the arrangement of trapping components within the Maypack.

Samples of gas were taken at all 14 locations concurrently by opening one of the valves in each cluster for 3 min. Each Maypack received only one sample, with twelve sampling times available for each cluster. Flow rate through the Maypack was maintained at $0.50 \pm 0.05 \text{ ft}^3/\text{min}$ (STP) of dry air, with the actual volume sampled later being calculated from knowledge of temperature and pressure at the sample position. Actually, only eleven of the Maypacks in each cluster were used to sample; the twelfth being left as a "blank" for background data.

Liquid sampling was extensive. Samples of 500-ml were drawn from all pools and troughs at many times. Many liquid samples were filtered to determine the insoluble radioactive fraction. The pH was measured on about one third of the samples.

Deposition coupons were exposed to the containment atmosphere at the same locations as the Maypack clusters. Some

CSE SAMPLE TYPES

FIGURE 3. CSE Sampling System Flowsheet

FIGURE 4. Schematic Diagram of CSE Aerosol Sampling System

FIGURE 5. Schematic Diagram of a CSE Maypack Showing Filter and Adsorber Arrangement

were also pressed against the outer vessel wall to provide a condensing surface. The coupons were 2.5 in. diameter by 1/16 in. thick disks of various materials, including Type 304L SS, carbon steel, silver-plated copper, aluminum and carbon steel coated with Amercoat 66* and Phenoline 302** paints. The coupons were exposed to the contained atmosphere for the entire experiment duration.

About 2000 samples were obtained from each experiment. These were analyzed by the CSE staff for iodine and cesium by gamma energy techniques and for uranium by alpha counting. Additional details of the analytical procedures are given by Coleman.⁽²⁶⁾

An important part of these experiments was the accurate measurement of the physical conditions that exist within the containment vessel. About 70 chromel-alumel thermocouples were installed throughout the gas and liquid spaces and attached to surfaces at various locations. Temperature readout was normally by multipoint recorders, though computer data logging is possible. Two absolute pressure gages, accurate to ± 0.1 psi, were provided. Commercially available anemometers[†] were installed at various elevations and radii. The steam supply was monitored by an orifice meter. The heat flux through the wall was measured by a heat flux meter^{††} and by direct measurement of steam condensate from known surface areas. Liquid volumes in the pools were obtained from pressure drop measurements across dip tubes.

* Epoxy coating manufactured by Amercoat Corp., South Gate, California

** Phenolic coating manufactured by Carboline Co., St. Louis, Missouri

† Heated thermopile Type S-22AX, power supply Model AM-62R 10X, manufactured by Hastings-Raydist, Inc., Hampton, Virginia

†† Hy-Cal Engineering Co., Santa Fe Springs, California

B. EXPERIMENTAL PROCEDURE

Prior to each CSE experiment a comprehensive "Run Plan" was written which described the physical system, conditions and time profile for sampling and data logging. Then, stepwise:

- The vessel, systems hardware and sampling equipment were readied.
- Shakedown tests were made when possible.
- The vessel was tested for air leakage, by pressurization.
- Air pressure was released until barometric conditions prevailed.
- The air was recirculated through filters until the condensation nuclei concentration was <500 particles/cm³.
- Steam was then fed to the vessel until the desired temperature was attained.
- Once isothermal, isobaric conditions were reached and maintained, the condensate generated during vessel warmup was withdrawn from the liquid pools and discarded.
- The Zircaloy-clad UO₂ fuel pin was brought to a molten state with a continuous air sweep to the containment vessel at 1.8 ft³/min (STP) flow rate.
- After the UO₂ had been molten for 10 min, iodine and cesium release commenced.
- Zero time was defined as the time that iodine and cesium release started. After 10 min, all release was terminated and the valves were closed. Methyl iodide was injected through a separate line during the same 10-min period.
- Steam was added to maintain isothermal conditions throughout all tests except Run A-11. In Run A-11 the steam supply was stopped as soon as the simulant was injected.
- Gas and liquid samples were taken at preselected intervals which were intended to best cover the changes

anticipated to occur in the system; i.e., at short time lapses initially when rapid changes were occurring and less frequently at later times.

- The experiment was terminated after 24 to 48 hr by stopping the steam supply.
- When the temperature dropped to ~ 100 °F the vessel was purged to the stack and the liquid pools drained.
- The injection line and simulant generating apparatus were decontaminated for material balance information.
- The vessel was opened and the Maypacks and deposition coupons were retrieved.
- The Maypacks were disassembled and the components sealed in polyethylene bags or jars. Care was taken to label each sample with its proper identification.
- Liquid samples were filtered, the pH measured, and diluted to standard volumes in 500-ml polyethylene bottles.
- Deposition coupons were sealed in polyethylene bags. Certain coupons were analyzed and returned to the containment vessel to serve as monitors of vessel surface total deposition.
- Gamma analysis began during the test and continued for 1 to 2 weeks on a 24-hr basis.
- Analytical data for all samples were computer reduced and stored on magnetic tape for final computer processing. Final tabulated results were usually available within 1 month after the experiment was terminated.

C. TEST CONDITIONS

Two tests, Runs D-1 and D-2, were made in the intermediate sized drywell vessel. Four experiments, Runs A-1, A-2, A-5, and A-11, were made in the main CSE vessel. In all six of these experiments the fission product simulant material

was released essentially instantaneously and no active counter-measures were operated. All FP transport occurred solely by natural processes. In addition to these six experiments, several other CSE experiments had periods in which only natural processes occurred, and some data from those tests are reported in this document.

The volumes and surface areas associated with the various compartments within the CSE containment vessel complex are shown in Table 4.1. In Runs A-1, A-2, A-5 and A-11 the FP simulant was released into the main room and not much entered the lower rooms. For this reason, the conditions associated with the main room are the most pertinent to mass transport studies, and these are listed in Table 4.2. These are the quantities needed as inputs to mathematical models discussed in Section VI. Some miscellaneous test conditions are listed in Table 4.3.

TABLE 4.1. Compartment Volumes and Surface Areas

	Volume, <u>ft³</u>	Surface Area, <u>ft²</u>	Surface to Volume Ratio, <u>ft⁻¹</u>
Main Room, Above Deck Level Plus Open Drywell	21,005	6,140 ^(a)	0.293
Middle Room	2,089	1,363	0.652
Lower Room	3,384	2,057	0.608
Closed Drywell	2,286	1,042	0.456
Total, All Rooms	26,477	9,560	0.361

a. Includes 242 ft² estimated for experimental equipment.

TABLE 4.2. Summary of Physical Conditions (a)

Parameter (b)	Run D-1	Run D-2	Run A-1	Run A-2	Run A-5	Run A-11
V, ft ³	2286	2286	21,005	21,005	21,005	21,005
D, ft	11	11	25	25	25	25
A _t , ft ²	1042	1042	6140	6140	6140	6140
A _w , ft ²	1042	1042	4500	4500	4500	4500
A _{cs} , ft ²	95	95	490	490	490	490
h _w , Btu/hr ft ² °F	23	23	37	37	0.31	0.31
T _b , °F	252 ± 2	250 ± 2	181 ± 2	185 ± 2	253 ± 3	252 ± 3(c)
T _a , °F	85 ± 5	90 ± 5	110 ± 5	110 ± 5	90 ± 5	85 ± 5
P, psia	48.5 ± 0.5	48.0 ± 0.5	22.5 ± 0.3	23.2 ± 0.3	46.5 ± 0.6	49.1 ± 0.2(c)
L, ft	20	20	40	40	40	40

a. In Runs A-1, 2, 5 and 11 the conditions are for the main room only.

b. Refer to table of Nomenclature for definitions.

c. Initial condition.

TABLE 4.3. Miscellaneous Test Conditions

	<u>D-1</u>	<u>D-2</u>	<u>A-1</u>	<u>A-2</u>	<u>A-5</u>	<u>A-11</u>
Isothermal	yes	yes	yes	yes	yes	no
Thermal Insulation on Walls	no	no	no	no	yes (a)	yes (a)
Previous Steam Exposure, hr	0	100	0	114	586	1500
Aerosol Release Duration, min	7	7	7	10	10	10
Avg Steam Makeup, lb/hr	395 ±25	330 ±25	510 ± 40	650 ± 50	295 ± 25	0
Methyl Iodide Released	no	no	no	no	yes	yes
Avg Temp. Middle Room, °F	--	--	150	163	233	218
Avg Temp. Lower Room, °F	--	--	127	128	210	160
No. of Maypack Clusters Used	7	7	15	11	14	14

a. Type PF-615 Fiberglass, 1 in. thick with aluminum vapor barrier,
 $k = 0.027 \text{ Btu/(hr) (ft}^2\text{) (}^\circ\text{F/ft)}$ at 200 °F. Owens-Corning
 Fiberglas Corp.

Maypack gas samplers were distributed throughout the gas spaces. Table 4.4 gives the coordinates of each sample location. Not every location listed in Table 4.4 was used in a single test, but locations were selected to give an approximate volume weighted average and also to cover several radii and elevations. In one test, Run A-11, seven clusters were installed together at the same location (M26) in order to obtain sampling error information.

TABLE 4.4. Locations of Maypack Clusters Used in Main CSE Vessel

Maypack Cluster Number	Elevation, ^(a) ft	Radius, ft	Azimuth, ^(b) degree	
M 01	0	8.8	225	
02	+12	0	0	0 of Main Room
03	+12	8.8	315	
04	+12	8.8	45	
05	+33	0	0	Top dome
06	+24	8.8	45	
07	0	8.8	45	
08	+12	8.8	135	
09	+12	6	45	
10	+24	8.8	225	
11	+24	8.8	225	
12	-6	4.5	135	In Drywell, near top
13	-18	4.5	135	In Drywell, bottom
14	-12	10	260	Middle Room
15	-24	10	260	Lower Room
16	+24	11	45	
17	+24	11	225	
18	0	11	45	
19	0	11	135	
20	+12	11	315	
21	+24	0	0	
22	+12	6	270	
23	0	11	270	
24	+12	11	45	
25	+24	11	270	
26	+12	9 ± 1.5	0	

a. Main deck is at -5.5 ft.

b. Aerosol injection line enters at 270°.

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V. RESULTS

A. FISSION PRODUCT SIMULANT RELEASE TO THE CONTAINMENT ATMOSPHERE

1. Release Rates

In all six of the experiments the fission product simulant was released over a relatively short period of time compared to the time constants of subsequent events within the containment vessel. The release periods were restricted to 10 min duration, short enough to be considered instantaneous for model fitting purposes. The mass release rates were estimated from dose rate measurements made on the starting mass in the generation apparatus, adjusted to match the material balance made after the release was complete. Figures 6 and 7 show typical curves of release rate versus time for iodine and cesium, respectively.

The volatilization rate of uranium oxide aerosol was measured in several shakedown tests and averaged 0.1 g/min. The uranium oxide fuel pin was heated to the melting point 10 min before the release of iodine and cesium was started, and terminated at the same time as the iodine and cesium. Methyl iodide was released in Runs A-5 and A-11 during the same 10-min interval.

The reference time zero, t_0 , for evaluation and discussion purposes was set at the time iodine and cesium release started.

2. Overall Mass Balances

The procedures for generating and injecting the fission product simulant materials were nearly identical in each of the six experiments. The masses delivered were changed from test to test according to the size of the vessel to which the aerosol was delivered and to the desired initial concentration

FIGURE 6. Typical Release Rate for Iodine

FIGURE 7. Typical Release Rate for Cesium

in the containment vessel atmosphere. Tables 5.1 through 5.6 give the overall mass balances for the six experiments. The top halves of the tables relate to the generation and release to the containment, while the lower halves are based on the masses delivered to the containment vessel.

After the release period was completed, valves were closed to isolate the generation equipment and delivery line from the containment vessel. The amount of material retained in the generation equipment and deposited in the delivery line was determined by decontamination of the equipment and analysis. The sum of the material found exterior to the vessel was subtracted from the known starting mass to determine the amount actually delivered to the containment vessel. The mass of iodine delivered, as reported in Tables 5.1 through 5.6, are believed accurate to $\pm 5\%$. For cesium, less complete release and higher plateout in the delivery line resulted in less certainty of its delivered mass. The values listed are believed accurate to $\pm 10\%$, except for Runs A-1 and A-5 where sample spillage increased the uncertainty to $\pm 20\%$.

The initial gas phase concentration, C_{go} , can be calculated by dividing the mass delivered by the gas volume in the vessel. The Drywell is essentially an empty vessel, so the entire gas space, 65 m^3 , should be used in this calculation. For the experiments in the compartmented main CSE vessel, the proper volume to use for times shortly after release is 595 m^3 . At long times, the space in the middle and lower rooms should be included.

Tables 5.1 through 5.6 show the distribution of iodine and cesium at termination of each experiment. The material found in four locations are reported as (1) the steam condensate in the vessel sumps, (2) material removed in samples,

TABLE 5.1. Overall Mass Balance - Run D-1

<u>Location</u>	<u>Iodine</u>		<u>Cesium</u>	
	<u>mg</u>	<u>% (a)</u>	<u>mg</u>	<u>% (a)</u>
<u>Aerosol Generation</u>				
Starting material	80.0	100.00	515.73	100.00
Generation apparatus	2.99	3.74	84.22	16.33
Injection line	31.81	39.76	109.12	21.16
Injection line samplers	2.03	2.54	17.72	3.44
Accounted for	36.83	46.03	211.06	40.92
Delivered to CV (by difference)	43.17	53.97	304.67	59.08
 <u>Containment</u>				
		(b)		(b)
Delivered to CV	43.17	100.0	304.67	100.00
Condensate, t = 22 hr	32.19	74.56	305.08	100.13
Samples	0.58	1.34	1.80	0.59
Purge to stack	0.10	0.23	0.00	0.00
Decontamination	1.34	3.11	11.92	3.91
Accounted for	34.21	79.24	318.80	104.64
On surfaces (by difference)	8.96	20.76	(-)14.13	(-)4.64
C_{go} , mg/m ³	0.664	-	4.7	-

a. Percent of starting mass.

b. Percent of delivered mass.

TABLE 5.2. Overall Mass Balance - Run D-2

<u>Location</u>	<u>Iodine</u>		<u>Cesium</u>		<u>Ruthenium</u>	
	<u>mg</u>	<u>% (a)</u>	<u>mg</u>	<u>% (a)</u>	<u>mg</u>	<u>% (a)</u>
<u>Aerosol Generation</u>						
Starting material	80.00	100.00	624.20	100.00	183.60	100.0
Generation apparatus	1.17	1.46	69.79	11.18	179.00	97.5
Injection line	16.40	20.50	230.16	36.87	1.46	0.8
Injection line samplers	1.61	2.02	14.95	2.39	1.13	0.6
Accounted for	19.18	23.98	314.90	50.45	181.60	98.9
Delivered to CV (by difference)	60.82	76.01	309.32	49.56	2.00	1.1
<u>Containment</u>						
		(b)		(b)		(b)
Delivered to CV	60.82	100.00	309.32	100.00	2.00	100.0
Condensate, t = 22 hr	35.52	58.41	231.03	74.69	1.57	78.5
Samples	0.34	0.57	0.96	0.31	0.01	0.5
Purge to stack	0.06	0.10	0.00	0.00	0.00	0.0
Decontamination	3.85	6.33	17.75	5.74	0.03	1.5
Accounted for	39.77	65.41	249.74	80.74	1.61	80.5
On Surfaces (by difference)	21.05	34.59	59.58	19.26	0.39	19.5
C _{go} , mg/m ³	0.936	-	4.76	-	0.031	-

a. Percent of starting mass.

b. Percent of delivered mass.

TABLE 5.3. Overall Mass Balance - Run A-1

<u>Location</u>	<u>Iodine</u>		<u>Cesium</u>	
	<u>Grams</u>	<u>% (a)</u>	<u>Grams</u>	<u>% (a)</u>
<u>Aerosol Generation</u>				
Starting material	1.10	100.0	9.36	100.0
Generation apparatus	0.164	14.9	8.1(c)	86.5
Injection line	0.210	19.1	0.842	9.0
Injection line samplers	0.031	2.8	0.056	0.6
Accounted for	0.405	36.8	8.998	96.1
Delivered to containment (by difference)	0.695	63.2	0.367	3.9
<u>Containment</u>		(b)		(b)
Delivered to containment	0.695	100.0	0.367	100.0
In condensate, t = 22 hr	0.284	40.8	0.267	72.5
Samples	0.0006	0.09	0.0004	0.1
Purge to stack	0.0008	0.11	0	0
Decontamination	0.122	17.6	0.082	22.2
Accounted for	0.407	58.6	0.349	94.8
On surfaces (by difference)	0.288	42.4	0.018	5.2
C _{go} , mg/m ³	1.17	-	0.617	-

- a. Percent of starting mass.
b. Percent of delivered mass.
c. Spilled sample. Estimated.

TABLE 5.4. Material Balances - Run A-2

<u>Location</u>	<u>Iodine</u>		<u>Cesium</u>	
	<u>Grams</u>	<u>% (a)</u>	<u>Grams</u>	<u>% (a)</u>
<u>Aerosol Generation</u>				
Starting material	101.4	100.0	10.80	100.0
Generation apparatus	21.5	21.2	4.84	44.77
Injection line	22.2	21.8	2.28	21.10
Injection line samplers	1.5	1.5	0.073	0.68
Accounted for	45.2	44.5	7.193	66.55
Delivered to containment (by difference)	56.2	55.5	3.607	33.45
 <u>Containment</u>				
		(b)		(b)
Delivered to containment	56.2	100.0	3.607	100.0
In condensate, t = 22 hr	29.45	52.40	3.189	88.40
Samples	0.04	0.67	0.0016	0.04
Purge to stack	0.10	0.07	0.00003	0.001
Decontamination	6.18	11.00	0.257	7.12
Accounted for	35.67	63.47	3.448	95.56
On surfaces (by difference)	20.5	36.5	0.159	4.4
C _{go} , mg/m ³	94.5	-	6.06	-

a. Percent of starting mass.

b. Percent of delivered mass.

TABLE 5.5. Overall Mass Balance - Run A-5

<u>Location</u>	<u>Iodine</u>		<u>Cesium</u>	
	<u>Grams</u>	<u>% (a)</u>	<u>Grams</u>	<u>% (a)</u>
<u>Aerosol Generation</u>				
Starting material	101.00	100.00	10.95	100.00
Generation apparatus	9.22	9.15	7.90(c)	72.15
Injection line	5.86	5.80	1.84	16.80
Injection line samples	1.41	1.40	0.11	1.00
Accounted for	16.49	16.35	9.85	89.95
Delivered to Containment (by difference)	84.51	83.65	1.10	10.05
<u>Containment</u>		(b)		(b)
Delivered to Containment	84.51	100.00	1.10	100.00
Condensate, t = 24 hr	18.25	21.60	0.93	84.55
Samples	0.76	0.90	0.003	0.27
Purge to stack	0.30	0.35	0.001	0.05
Decontamination	2.11	2.50	0.06	5.45
Accounted for	21.42	25.35	0.994	90.32
On surfaces (by difference)	63.09	74.65	0.106	9.68
C_{go} , mg/m ³	142.00	-	1.85	-

a. Percent of starting mass.

b. Percent of delivered mass.

c. Spilled sample. Estimated

TABLE 5.6. Overall Mass Balance - Run A-11

Location	Iodine		Cesium	
	Grams	% (a)	Grams	% (a)
<u>Aerosol Generation</u>				
Starting material	101.35	100.00	11.46	100.00
Generation apparatus	0.22	0.21	4.45	38.84
Injection line	1.47	1.45	1.91	16.66
Injection line samples	1.31	1.29	0.08	0.72
Accounted for	3.00	2.95	6.44	56.22
Delivered to Containment (by difference)	98.35	97.05	5.02	43.78
 <u>Containment</u>				
		(b)		(b)
Delivered to Containment	98.35	100.00	5.02	100.00
Condensate, t = 46 hr	30.19	30.69	1.19	23.67
Samples	1.03	1.04	0.011	0.22
Purge to stack	0.35	0.36	0.012	0.24
Decontamination	14.82	15.07	1.72	34.36
Accounted for	46.39	47.16	2.93	58.49
On surfaces (by difference)	51.97	52.84	2.09	41.51
C_{go} , mg/m ³	165.00	-	8.44	-

a. Percent of starting mass.

b. Percent of delivered mass.

(3) material remaining airborne and purged to stack, and
 (4) the surface decontamination liquid. Not measured directly was the material remaining on structural surfaces after decontamination efforts were completed. The latter was estimated by two methods. The method shown in Tables 5.1 through 5.6 was to assume that any materials not found elsewhere were still on the painted surfaces. A second method, discussed in an ensuing section, was to extrapolate the amount found on small deposition coupons located throughout the vessel to an equivalent amount on the total surface area exposed to the vapor phase. Satisfactory agreement was noted between the two methods.

The average distribution between the five locations, expressed as percent of mass delivered to the containment vessel, is given in Table 5.7, along with the standard deviation between tests. Because the test conditions varied so widely, the standard deviations are rather large. However, it is clear that the iodine was about equally divided between the condensate in the vessel sumps and on the painted surfaces, with very little remaining airborne at the termination of the tests. A higher fraction of cesium (~75%) was found in the sumps, and correspondingly less on the surfaces. These results are quite similar to those of experiments where caustic sprays were operated,⁽⁵⁾ except that somewhat higher fractions were found in the sumps for the spray experiments.

TABLE 5.7. Average Distribution of Iodine and Cesium at End of Experiment

	Iodine	Cesium
	Percent of Delivered Mass, $\pm\sigma$	Percent of Delivered Mass, $\pm\sigma$
Condensate	46.1 $\pm$ 19.4	74.6 $\pm$ 26.6
Samples	0.8 $\pm$ 0.4	0.3 $\pm$ 0.2
Gas Phase	0.2 $\pm$ 0.1	0.06 $\pm$ 0.1
Decontamination	9.2 $\pm$ 6.4	13.1 $\pm$ 12.4
Surfaces	43.7 $\pm$ 18.5	12.5 $\pm$ 16.2

3. Iodine Forms in Containment Atmosphere

Iodine was released in three intended forms, consisting of elemental I_2 , methyl iodide, and a small fraction attached to solid particles. Undetermined forms were probably also released in small amounts. The relative proportion of these forms as sampled from the delivery line (before entry into the containment atmosphere) was ~ 0.97 , 0.01 , 0.02 and <0.01 , respectively. As soon as they entered the steam-air containment atmosphere, these relative proportions changed rapidly due to absorption of some elemental iodine in fog drops⁽²⁸⁾ and different rates of deposition on surfaces for the various forms. Table 5.8 lists the percentage in each of the four forms at times 15 min, 2 hr and 24 hr after start of aerosol release.

Cesium was volatilized as cesium metal and cesium oxide particles of about 0.25μ mass median diameter (MMD), as determined by cascade impactor sampling from the delivery line during shakedown tests. Some growth occurred by agglomeration during the 18-sec transit time to the containment vessel. The rapid fog formation observed within the containment atmosphere is indicative of the rapid absorption of water vapor on the cesium particles to form cesium hydroxide solution droplets. A later section deals with particle sizes in the containment atmosphere.

4. Visual Observations in the Containment Atmosphere

A view of the interior of the containment vessel was possible by means of a 6-in. diameter window and lights located within the vessel. The window was kept free of condensation by a heat lamp mounted externally. The window was not installed for Runs D-1, D-2 and A-1. Visual observations were similar in Runs A-2, 5 and 11, and are summarized as follows.

TABLE 5.8. Iodine Form in Containment Atmosphere

	Percent of Total Airborne Iodine					
	Run D-1	Run D-2	Run A-1	Run A-2	Run A-5 (b)	Run A-11 (b)
Elemental Iodine						
t = 15 min	76.0	59.7	17.0	91.4	82.8	84.5
t = 2 hr	48.3	44.8	43.5	38.5	23.5	16.9
t = 24 hr	19.0	15.0	15.3	8.8	7.9	1.8
Particulate Iodine						
t = 15 min	9.3	33.5	55.5	4.2	12.3	11.3
t = 2 hr	1.0	6.4	18.8	5.5	18.0	41.3
t = 24 hr	6.9	7.2	1.4	0.2	1.7	0.7
Methyl Iodine						
t = 15 min	0.8	0.11	0.27	0.25	1.9	2.0
t = 2 hr	41.2	23.3	4.1	29.9	46.0	28.1
t = 24 hr	55.4	55.5	44.1	78.2	80.2	83.1
Unknown (a)						
t = 15 min	13.9	6.6	27.2	4.2	3.0	2.2
t = 2 hr	9.5	25.5	33.6	26.1	12.5	13.7
t = 24 hr	18.8	22.3	39.2	12.8	10.2	14.4

a. Material deposited on inlet to Maypack and on charcoal paper.

b. One gram iodine added as methyl iodide.

When steam was injected into the cold, sealed vessel, a fog formed which rapidly filled the vapor space and reduced visibility to 6-8 feet. After the desired test temperature was attained and the steam feed was reduced to that required to maintain isothermal conditions, the fog dissipated and the atmosphere was clear except for a very light mist. Convection velocities were noticed by motion of aluminum foil ribbons, slight swaying of sampling lines, and by recording anemometers.*

Not much change was noticed when uranium and Zircaloy oxide fumes were injected, but immediately upon start of cesium and iodine release, a dense cloud issued out of the Drywell and rapidly filled the entire vapor space. The visibility was reduced to 1 to 2 ft by the end of the 10-min release period. Figure 8 is a plot of visibility versus time for Run A-2. A definite violet tinge was evident by the end of the iodine release period, evidence of the presence of relatively high concentrations of elemental iodine. About 3 hr after release, the atmosphere was again clear. Figure 9 shows a series of photographs taken through the window during Run A-11.

B. MIXING IN THE VAPOR SPACE

In experiments in the main CSE containment vessel, Maypack Clusters were hung at various locations throughout the main room, as indicated in Table 4.4. In the Drywell tests, seven clusters were used. In all six experiments the variation between gas concentrations measured at the different locations was essentially the same as the sampling error expected. Table 5.9 lists the standard error as percent deviation from the mean for Run A-1. The other experiments gave similar results.

* Heated thermopile type, manufactured by the Hastings-Radist Co., Hampton, Virginia.

FIGURE 8. Visibility in Containment Atmosphere - Run A2

(a) TIME = t_0 - 19 hours
75 °F, 14.3 psia

(b) TIME = t_0 - 8 min
252 °F, 49.1 psia

(c) TIME = t_0 + 2 min
252 °F, 49.1 psia

(d) TIME = t_0 + 9 min
252 °F, 49.1 psia

(e) TIME = t_0 + 21 min
251 °F, 48.8 psia

(f) TIME = t_0 + 252 min
227 °F, 39.2 psia

FIGURE 9. View of Containment Atmosphere
at Various Times - Run A-11

TABLE 5.9. Typical Variation in Spatial
Concentration in CSE Vessel

Time Sample Taken, min ^(b)	Standard Deviation from Mean Concentration (1 σ ,%) ^(a)				
	Elemental Iodine	Particulate Iodine	Methyl Iodide	Total Iodine	Cesium
5 ^(c)	24	19	32	20	19
11	24	11	30	11	13
26	19	15	33	15	11
41	12	11	26	11	10
61	15	18	25	15	8
81	20	17	28	18	8
101	18	17	26	18	7
121	24	24	23	24	10
181	19	24	25	20	16
300	15	38	38	15	13
1100	20	76	30	20	-

- a. Run A-1, mean concentration is average of 13 sampling locations in the main room plus open drywell.
- b. Midtime of 3-min sample periods.
- c. Sampled during aerosol release period.

Seven Maypack clusters were installed at the same location (M26) in Run A-11, in order to determine the sampling error for single measurements. One of the clusters had an electrical failure, but the results for the remaining six samplers are listed in Table 5.10. Also listed is the average of the samplers at six different locations. The conclusion is that the standard error for an individual sampler was about $\pm 10\%$ at early times due to variations in sampling rate, sampling duration, and gamma analytical measurement error. At longer times, larger deviations

TABLE 5.10. Gas Sample Variation at Same Location
and Between Different Locations in
Main Room - Run A-11

	Avg of 6 Sam- plers at Single Location(a)	Avg of 6 Samplers at 6 Different Locations(b)
	$C_g, \text{mg/m}^3 \pm 1 \sigma$	$C_g, \text{mg/m}^3 \pm 1 \sigma$
<u>Elemental Iodine</u>		
t = 22 min ^(c)	43.2 $\pm 12\%$	43.7 $\pm 12\%$
t = 5 hr ^(c)	0.156 $\pm 16\%$	0.154 $\pm 15\%$
t = 24 hr ^(c)	0.0357 $\pm 43\%$	0.0284 $\pm 50\%$
<u>Particulate Iodine</u>		
t = 22 min	5.64 $\pm 12\%$	5.76 $\pm 11\%$
t = 5 hr	0.425 $\pm 8\%$	0.376 $\pm 23\%$
t = 24 hr	0.0043 $\pm 104\%$	0.0072 $\pm 84\%$
<u>Methyl Iodide</u>		
t = 22 min	1.32 $\pm 6.7\%$	1.41 $\pm 18\%$
t = 5 hr	1.15 $\pm 2.9\%$	1.17 $\pm 3.9\%$
t = 24 hr	0.74 $\pm 7.7\%$	0.70 $\pm 11\%$
<u>Charcoal Paper</u>		
t = 22 min	1.21 $\pm 7.6\%$	1.16 $\pm 12\%$
t = 5 hr	0.238 $\pm 6.2\%$	0.246 $\pm 6.8\%$
t = 24 hr	0.104 $\pm 21\%$	0.100 $\pm 22\%$
<u>Total Iodine^(d)</u>		
t = 22 min	51.6 $\pm 9.9\%$	52.2 $\pm 9.8\%$
t = 5 hr	1.98 $\pm 2.9\%$	1.96 $\pm 5.6\%$
t = 24 hr	0.88 $\pm 7.3\%$	0.83 $\pm 10\%$
<u>Cesium</u>		
t = 22 min	1.50 $\pm 8.2\%$	1.54 $\pm 7.8\%$
t = 5 hr	0.482 $\pm 5.9\%$	0.455 $\pm 13\%$
t = 24 hr	0.135 $\pm 11.6\%$	0.131 $\pm 9.9\%$

a. Operated simultaneously at location M26.

b. Locations M05, M13, M16, M17, M24 and M26.

c. Midtime of 3-min sample periods.

d. Including Maypack inlet deposition.

occurred for elemental and particulate iodine forms, but not for methyl iodide, total iodine and cesium. This was probably due to imperfect Maypack discrimination for the elemental and particulate forms after the concentration of these forms had decreased to <1% of their original values.

The equation of continuity for any airborne species is given by Equation (1).

$$\frac{\partial C}{\partial t} + D_v \left[\frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2} + \frac{\partial^2 C}{\partial z^2} \right] + u \frac{\partial C}{\partial x} + v \frac{\partial C}{\partial y} + w \frac{\partial C}{\partial z} = \frac{G}{V} - \frac{R}{V} \quad (1)$$

where C = concentration in gas at coordinate x, y, z

t = time

D_v = effective diffusivity

u = convection velocity with respect to x coordinate

v = convection velocity with respect to y coordinate

w = convection velocity with respect to z coordinate

G = source term into gas space

R = removal term from gas space

V = gas volume

Knudson and Hilliard⁽⁸⁾ discussed mixing in closed vessels and concluded that the convective terms are much greater than the diffusion term in Equation (1) for conditions pertinent to loss-of-coolant accidents in containment vessels. Thus the conclusion reached in these CSE experiments that the gas space is well-mixed at all times is probably applicable to large PWR containment vessels as well. The CSE experiments were made without the benefit of ventilation or air recirculation systems, which would have increased the mixing rate had they been present.

The above discussion pertains to the single "main room" gas space. Large concentration differences between different compartments existed for fairly long times, however. Figures in the Appendix of airborne concentration versus time show how the concentrations of all fission product simulant forms built up in the lower rooms while the main room concentration decreased. Air recirculation systems, such as those installed in most large nuclear power plants, would have rapidly mixed the iodine to uniform concentration in all three rooms.

C. TIME DEPENDENCE OF CONCENTRATION IN VAPOR SPACE

The equation of continuity, Equation (1), included terms for source and removal. As discussed previously, the release of fission product simulant in these tests was short enough to be considered instantaneous, hence $G = 0$ for times after the release stopped. Measurement of the removal terms was a primary objective of the experiments. The chief tool for accomplishing this objective was the determination of the mass concentrations in the gas space as a function of time. The rate of change of the concentration is a measure of the overall removal rate. Since the gas space was well-mixed, only the mean concentrations are reported in the following sections. Tables A-1 through A-6 in the Appendix list the Maypack data categorized by type of aerosol constituent.

1. Elemental Iodine

Figures B-1 through B-6 in the Appendix show the mass concentrations of elemental iodine in the gas space as a function of time. Straight line segments of the curves on the semi-logarithmic plots are marked with the half life; i.e., the time required to reduce the concentration by a factor of two. Plus and minus one standard deviation from the mean is indicated by the horizontal bars above and below each point on the curve for the main room concentration. The lower rooms had only a single sampler each.

The concentration behavior in all six tests was similar. There was an initial rapid exponential removal lasting until the concentration had decreased to 0.001-0.005 of the initial value, followed by a transition to a second, slower exponential period which lasted until the experiment was terminated. Table 5.11 tabulates the observed half lives, transition times and the decontamination factors (DF) at 2-hr and 24-hr aging times.

Also shown in the figures for the four tests in the main CSE vessel are the concentrations in the middle and in the bottom rooms. Since the iodine was released into the main room, the concentration in the lower rooms started at zero and increased with time as iodine diffused or was convected from the main room.

2. Particulate Iodine

Figures B-7 through B-12 in the Appendix show the concentration of particulate-associated iodine in the gas space as a function of time. It should be pointed out that iodine not only attaches itself to solid particles, but is absorbed by liquid droplets, if such are present. In these experiments, the cesium particles which were introduced along with the iodine probably reacted to some extent in the aerosol injection line to form some cesium iodide. But as soon as the particles were introduced to the steam in the containment vessel they acted as condensation nuclei to form fog drops. Additional elemental iodine was undoubtedly absorbed into these fog drops very rapidly to some equilibrium relationship between gas and liquid. The Maypack samplers sampled the fog drops and any iodine in them was determined to be particulate-associated. If the relative concentrations of gaseous elemental iodine and concentration in the liquid fog drops changed

TABLE 5.11. Early and Late Half Lives for Elemental Iodine in the Gas Space

Run No.	Half Life		Time Start of Transition Period, min	DF (a)	
	Early, min	Late, hr		t = 2 hr	t = 24 hr
D-1	8.0 ± 0.5	9.7 ± 1.5	80	1,000	10,000
D-2	9.5 ± 0.5	10 ± 3	90	1,000	6,000
A-1	15 ± 1	(b)	40	10	500
A-2	9.0 ± 0.5	6.6 ± 1.1	100	1,000	20,000
A-5	13.5 ± 0.5	$\left\{ \begin{array}{c} 22 + 12 \\ - 6 \end{array} \right\}$	100	300	2,400
A-11 (c)	16.0 ± 0.5	$\left\{ \begin{array}{c} 4 \pm 0.5 \\ 18 + 15 \\ - 7 \end{array} \right\}$	150	160	4,000

a. Decontamination factor for elemental iodine. Concentration at t_0 divided by concentration at the indicated time.

b. Indeterminate.

c. Two transitions. $t_{1/2}$ changed from 16 min to 4 hr at $t = 150$ min then from 4 to 18 hr at $t = 14$ hr

due to different removal rates, iodine would redistribute rapidly to maintain equilibrium. Evidence of this behavior can be seen especially clearly in Run A-1 where the low initial iodine concentration ($\sim 1 \text{ mg/m}^3$) probably had a very high gas-liquid equilibrium partition coefficient compared to the higher concentration tests.

Table 5.12 lists the early and late half lives, transition times, and decontamination factors at 2-hr and at 24-hours.

3. Methyl Iodide

Figures B.13 through B.18 in the Appendix show the concentration of methyl iodide in the gas space as a function of time. The concentration is seen to decrease much more slowly with time than for the other iodine forms. Some of the decrease in the main room is due to diffusion into the lower rooms, and this must be taken into account in determining the removal to surfaces. Table 5.13 lists the half life determined by two different methods: by overall decrease in airborne mass, and by the slope of the curve of concentration versus time after the rapid initial diffusion into lower rooms had subsided. The agreement is fair, considering the relatively poor accuracy due to the small decrease in concentration. The first method (total airborne mass) should provide the more accurate estimate for the case of methyl iodide.

4. Iodine Associated with Charcoal Paper

Figures B.19 through B.24 in the Appendix show the concentration in the gas space of the iodine caught on the charcoal-impregnated filter paper of the Maypack. The chief function of the charcoal paper is to ensure that inorganic forms of iodine do not penetrate to the granular charcoal bed section of the Maypack. It probably catches a mixture of

TABLE 5.12. Early and Late Half Lives for Particulate Iodine in the Gas Space

Run No.	Half Life		Time Start Transition Period, min	DF(a)	
	Early, min	Late, hr		t = 2 hr	t = 24 hr
D-1	8.0 ± 0.5	(b)	100	4,000	4,000
D-2	8.7 ± 0.4	(b)	110	25,000	50,000
A-1	10.0 ± 1.0	(b)	40	75	<10,000
A-2	17.3 ± 4.0	5 ± 3	130	1,000	30,000
A-5 (c)	45 ± 4	$\left\{ \begin{array}{c} 9 \pm 13 \\ - 4 \end{array} \right\}$	55	300	1,000
	15 ± 2		140		
A-11	57 ± 5	3.2 ± 0.5	240	4.0	1,000

a. Decontamination factor for particulate iodine. Concentration at t_0 divided by concentration at indicated time.

b. Indeterminate.

c. Two transitions. $t_{1/2}$ changed from 45 to 15 min at $t = 55$, then from 15 to 540 min at $t = 140$ minutes.

TABLE 5-13. Half Lives for Methyl Iodide
in the Gas Space

Run No.	Total Airborne ^(a) mass, mg		DF ^(b)	Effective Half Life, hr	Half Life ^(c) from Slope of curves, hr
	$t = 0$	$t = 24 \text{ hr}$			
D-1	0.031	0.0081	3.84	12	12.3 $\pm$ 2.3
D-2	0.011	0.011	1.0	—	(d)
A-1	0.298	0.249	1.20	93	90 $\pm$ 50
A-2	80.3	59.9	1.34	57	50 $\pm$ 20
A-5	774	287	2.70	17	28 $\begin{cases} + 12 \\ - 4 \end{cases}$
A-11	774	477	1.62	34	25 $\pm$ 12

a. In all compartments.

b. Decontamination factor for methyl iodide. Total airborne mass at 24 hr divided by total airborne mass at t_0 .

c. From Figures B.13 through B.18 in Appendix.

d. Indeterminate.

inorganic forms plus 1 to 5% of the methyl iodide.⁽¹²⁾ It represented only 1 to 2% of the total iodine at early times, but after 24 hr aging in containment it accounted for 10 to 50% of the remaining airborne iodine. Table 5.14 lists the half lives and decontamination factors.

5. Total Inorganic Iodine

Figures B.25 through B.30 in the Appendix show the concentration of total inorganic iodine forms versus time. The data for these figures were obtained by summing the iodine caught on all components of the Maypacks except the granular charcoal section. Iodine which penetrates to the granular charcoal bed has been shown by calibration tests to consist only of methyl iodide.⁽¹²⁾ Thus, the total inorganic iodine

TABLE 5.14. Early and Late Half Lives for Iodine Associated with Charcoal Paper in the Gas Space

Run No.	Half Life		Time Start Transition Period, min	DF ^(a)	
	Early, min	Late, hr		t = 2 hr	t = 24 hr
D-1	9.0 ± 3	12.5 ± 2	60	200	1000
D-2	14 ± 3	37 ± 20	60	40	800
A-1	83 ± 8	5.5 ± 0.5	120	2.6	60
A-2	42 ± 12	10 ± 2	120	4.0	27
A-5	24 ± 12	23 ± 8	100	10	40
A-11 ^(b)	50 ± 10	10 ± 2 60 ± 10	100 960	3.0	12

a. Decontamination factor for iodine associated with charcoal paper. Concentration at t_0 divided by concentration at indicated time.

b. Two transition periods. $t_{1/2}$ changed from 50 min to 10 hr at 100 min, then from 10 to 60 hr at 16 hours.

as defined here consists of I_2 , HI, HOI, particulate-associated iodine and any other ionic species which might be airborne. It also includes a small amount of methyl iodide caught on the charcoal-impregnated paper.⁽¹²⁾

The early and late concentration half lives and decontamination factors are listed in Table 5.15. Comparison with Table 7 shows that total inorganic iodine behaves very nearly like elemental iodine. This is to be expected, since the iodine released to the containment vessel was largely in elemental form for most of the experiments (see Table 5.8).

The overall DF after 24 hr aging was about half that of elemental iodine, however, indicating the presence of other inorganic forms in significant amounts at long aging times.

TABLE 5.15. Early and Late Half Lives for Total Inorganic Iodine in the Gas Space

Run No.	Half Life		Time Start Transition Period, min	DF ^(a)	
	Initial, min	Late, hr		t = 2 hr	t = 24 hr
D-1	8.0 ± 1.0	10 ± 2	70	670	5000
D-2	9.5 ± 0.5	14 ± 5	85	800	3000
A-1	9.0 ± 4.0	5.5 ± 1.5	40	30	1200
A-2	10.8 ± 2.0	7.5 ± 1.0	80	270	3500
A-5	15.2 ± 1.5	22 ± 8	110	120	1000
A-11	21 ± 4	5.2 ± 1.0	140	40	670

a. Decontamination factor for total inorganic iodine. Concentration at t_0 divided by concentration at indicated time.

6. Total Iodine

Figures B.31 through B.36 in the Appendix show the concentration of total airborne iodine in all forms as a function of time. Table 5.16 lists the early and late concentration half lives and decontamination factors. The initial behavior is very similar to that of the total inorganic fraction, but at longer times the presence of methyl iodide can be seen by the lower DF and half life. The behavior is consistent with the combined behavior of the individual iodine forms, as would be expected.

TABLE 5.16. Early and Late Half Lives for Total Iodine in the Gas Space

Run No.	Half Life		Time Start Transition Period, min	DF ^(a)	
	Initial, min	Late, hr		t = 2 hr	t = 24 hr
D-1	9.0 ± 1.0	11 ± 1.5	70	500	2500
D-2	9.0 ± 1.0	30 ± 14	60	1000	2300
A-1	10.5 ± 3.0	14 ± 6	30	40	570
A-2	11.0 ± 1.0	22 ± 7	80	300	1000
A-5	17.0 ± 3.0	26 ± 6	90	67	220
A-11	24 ± 3	11 ± 2	110	28	125

a. Decontamination factor for total iodine. Concentration at t_0 divided by concentration at indicated time.

7. Cesium

Figures B-37 through B-42 in the Appendix show the concentration of cesium in the gas space as a function of time. Table 5.17 lists the early and late half lives and decontamination factors obtained from the figures. Cesium was removed from the main room gas space somewhat more slowly at early times than inorganic iodine, but the removal mechanisms persisted longer, giving overall decontamination factors much larger than for iodine after 24 hr of aging. The one exception was for Run A-11, the temperature decay case, where the DF at 24 hr was only 15. This is discussed in a later section.

TABLE 5.17. Early and Late Half Lives for Cesium
in the Gas Space

Run No.	Half Life		Time Start Transition Period, min	DF ^(a)	
	Initial, min	Late, hr		t = 2 hr	t = 24 hr
D-1	8.5 ± 0.5	(b)	90	1500	~15,000
D-2	10.0 ± 2.5	(b)	60	400	~4,000
A-1	14 ± 4	1.1 ± 0.1	30	16	>35,000
A-2	16.5 ± 2.5	0.67 ± 0.10	80	70	>50,000
A-5	38 ± 4	1.45 ± 0.17	120	8	~15,000
A-11	50 ± 10	7.8 ± 0.5	50	3	15

a. Decontamination factor for cesium. Concentration at t_0 divided by concentration at indicated time.

b. Indeterminate.

8. Uranium

The concentration of airborne uranium particles in the main room gas space is shown in Figures B.43 through B.47 in the Appendix. The early and late half lives and decontamination factors are listed in Table 5.18. The behavior was nearly identical to that of cesium indicating that either cesium and uranium particles had the same size distribution or, more probably, coexisted in the same particles.

9. Ruthenium

Ruthenium was released in only one experiment, Run D-2. Figure B.48 in the Appendix shows the concentration as a function of time. Its half life was the same as that of cesium and uranium, within the accuracy of the measurements.

TABLE 5.18. Early and Late Half Lives for Uranium
in the Main Gas Space

Run No.	Half Life		Time Start Transition Period, min	DF ^(a)	
	Initial, min	Late, hr		t = 2 hr	t = 24 hr
D-2	8.5 ± 3.0	(b)	(b)	1500	>6000
A-1	21 ⁺²⁰ -5	1.25 ± 0.2	40	5.3	>500
A-2	21 ± 5	0.67 ± 0.1	70	23	>1500
A-5	40 ± 14	1.8 ± 0.4	90	6.1	>700
A-11	25 ± 13	7.1 ± 0.5	60	7.0	50

a. Decontaminaton factor for uranium. Concentration at t_0 divided by concentration at indicated time.

b. Indeterminate.

Its concentration decreased to below detection limits within 5 hours. (DF >10,000)

D. CONCENTRATION IN STEAM CONDENSATE

The steam-air mixture in the contained atmosphere was at an elevated temperature in these experiments. The air surrounding the test vessels was 70-160 °F cooler and the vessel walls were not provided with heating devices. Therefore, heat was transferred through the walls at rates dependent on thermal resistance of the wall and gas films surrounding the walls and on the overall temperature difference. Water vapor which diffused from the bulk gas to the cooler surfaces condensed, releasing its latent heat of condensation. Thus, all heat transfer surfaces were wet by a continuous film of liquid condensate in these experiments. This condensate film drained by gravity to lower areas and eventually accumulated in either the main containment vessel (CV) sump or the drywell (DW) sump.

The liquid level in the two sumps was monitored by frequent visual readings of manometer-dip tube level indicators. Figure 10 shows the liquid volumes as a function of time. Individual readings are omitted for sake of clarity, but they were within $\pm 10\%$ of the curves shown by Figure 10.

In Run A-11 the containment pressure and temperature decreased with time, as shown by Figure 11. The rate of steam condensation also decreased with time, as shown in Figure 12.

The liquid in the CV and DW sumps was recirculated through mixer-eductors, and samples were taken periodically for cesium and iodine analysis.

Samples of the condensate film were also obtained from five locations by draining from troughs to collector stations.

1. Concentration in Vessel Sumps

The concentration of iodine and cesium in the liquid accumulated in the CV and in the DW sumps is plotted against time in Figures B.49 through B.54 in the Appendix. In the isothermal experiments the steam condensed at a constant rate while the iodine and cesium transfer rate from gas to surfaces decreased with time. This caused the concentration in the sumps to increase rapidly to a peak at about one hour after release, followed by a gradual decline for the duration of the test.

In the temperature decay experiment where the steam condensation rate decreased with time, Run A-11, the concentration of iodine peaked at a later time and then decreased only slightly. Cesium concentrations continued to increase very slightly throughout the experiment.

Trouble was encountered with the sight glass used in Run D-1, and several partial drains were made of the sump

FIGURE 10. Liquid Volume in Vessel Sumps as a Function of Time

FIGURE 11. Temperature and Pressure History for Run A-11

FIGURE 12. Steam and Heat Flux, Run A-11

liquid. Figure A.49 shows the actual concentration within the sump at all times. The material removed was analyzed and added to the overall mass balance reported in Table 5.1. The DW sump sample line plugged in Run A-1, so no samples were obtained from the drywell sump in that test.

2. Wall Condensate Film

The condensate was sampled from the wall film at five locations. Four of these were from defined local areas, the fifth was from the mixed stream leaving the vertical walls near the deck.

The local wall condensate sampling system is shown schematically in Figure 13. The condensation areas were sections of the main CSE containment vessel wall in vertical array. The sections were defined by vertical fins spaced 24 in. apart and horizontal troughs spaced 10 ft apart. The array extended from near the main deck to about midway up the curved top dome of the containment vessel. Stainless steel sample lines extended down the wall from each trough, penetrating the vessel wall at Nozzle 11 and terminating in a 2-liter receiver in the ADF laboratory. The sample lines had a vertical gradient through-out to minimize holdup. Each receiver was vented back to containment. Table 5.19 lists some pertinent characteristics of the system.

The receivers were drained periodically, the amount collected was weighed and the iodine and cesium concentrations determined for each sample. Figures B.55 through B.57 in the Appendix show the iodine concentration as a function of time. Figures B.58 through B.61 give similar data for cesium.

E. DEPOSITION COUPON DATA

Coupons of various types of materials were suspended at different locations throughout the gas space to measure

FIGURE 13. Local Wall Condensate Sampling System Schematic

TABLE 5.19. Local Wall Condensate Sampling System Details

Item	Elevation, ft	Area Sampled, ft	Volume of Trough and Lines, ml	Line Length, ft
L09 trough ^(a)	+29.7	20.0	2545	100
L10 trough	+19.9	19.7	2340	92
L11 trough	+9.9	20.0	2185	84
L12 trough	(-)0.2	20.2	2000	76
Nozzle 11	(-)19.3			
Receivers ^(b)	(-)29.7			

a. 1 × 1 × 24 in., 400 ml capacity.

b. Stainless steel, 2160 ± 15 ml capacity.

deposition on noncondensing surfaces. Some were also pressed against the outer wall to measure deposition in the presence of heat transfer and steam condensation. The latter coupons extended about 1/16 in. out from the vessel wall, so it is doubtful that condensate from higher elevations ran over them. But since they were attached to the colder wall, condensate did form on them. The coupons were installed before the test and removed after the vessel had been purged. All were oriented vertically.

Table 5.20 lists the average iodine deposition on five types of materials exposed to noncondensing conditions. The standard deviations from the means are also shown and indicate that deposition was fairly uniform throughout the main room. Similar data are presented for cesium in Table 5.21. The relative affinity of each of the five materials for iodine and cesium can be inferred by examining the deposition for a

TABLE 5.20. Iodine Deposition on Various Noncondensing Surfaces

Surface Material (a)	Deposition at End of Experiment $\mu\text{g}/\text{cm}^2$ and 1 σ (%) (b)				
	Run D-1	Run D-2	Run A-1	Run A-2	Run A-5
Silver	0.0093 (30)	0.0144 (15)	0.065 (21)	34.0 (35)	30.0 (33)
Carbon Steel	0.0009 (25)	0.0027 (38)	0.0352 (16)	20.0 (24)	26.6 (27)
Stainless Steel 304 (ASTM A240-63)	0.00011 (24)	0.00034 (36)	0.00075 (84)	0.023 (33)	0.165 (45)
Phenoline 302 (c)	0.0175 (40)	0.0255 (22)	0.0215 (34)	12.0 (34)	36.5 (30)
Amercoat 66 (d)	-	-	0.0755 (21)	17.0 (20)	28.7 (23)
					29.5 (19)

a. 2.5 in. diameter, 1/16 in. thick coupons with 17/32 in. center hole. Total surface area = 64 cm^2 .

b. Mean of 11 locations in the main room. Numbers in parentheses are the standard deviations from mean values, in percent.

c. Phenolic coating manufactured by Carboline Co., St. Louis, Mo. Average of 22 locations.

d. Epoxy coating manufactured by Amercoat Corp., South Gate, Calif.

TABLE 5.21. Cesium Deposition on Various Noncondensing Surfaces

Surface Material (a)	Deposition at End of Experiment Nanograms/cm ² and 1 σ (%) (b)					
	Run D-1	Run D-2	Run A-1	Run A-2	Run A-5	Run A-11
Silver	3.6 (62)	4.6 (47)	0.27 (44)	28 (120)	10 (59)	73 (47)
Carbon Steel	42 (34)	36 (18)	5.3 (43)	100 (25)	25 (64)	115 (75)
Stainless Steel 304 (ASTM A240-63)	1.2 (37)	0.55 (52)	0.28 (42)	2.4 (66)	4.0 (112)	65 (55)
Phenoline 302 (c)	6.8 (18)	9.4 (42)	0.37 (79)	6.1 (73)	38 (45)	70 (72)
Amercoat 66 (d)	-	-	0.28 (97)	3.3 (76)	13 (68)	49 (50)

a. 2.5 in. diameter, 1/16 in. thick coupons with 17/32 in. center hole. Total surface area = 64 cm².

b. Mean of 11 locations in main room. Numbers in parenthesis are standard deviations from mean values, in percent.

c. Phenolic coating manufactured by Carboline Co., St. Louis, Missouri.

d. Epoxy coating manufactured by Amercoat Corporation., South Gate, California.

single experiment. The deposition of iodine was much lower on stainless steel than on the other four materials. Deposition of iodine on silver, carbon steel, Amercoat 66 and Phenoline 302 was about the same, suggesting that deposition rates were limited by gas phase transfer rather than reaction at or within the surface materials. For cesium, deposition on stainless steel was also significantly lower than on the other materials. Deposition on carbon steel was highest, with deposition on silver and the two coatings being about equal. This suggests that cesium reacted with the carbon steel surface but not appreciably with silver or paint.

The deposition on small coupons should not be expected to represent accurately the deposition on large surface areas. Other investigators^(13,29) have shown that orientation, heat transfer and location can affect deposition rates. Gas boundary layer thicknesses are dependent on length along the surface and angle of inclination of the surface to the gas stream.⁽³⁰⁾ However, the data obtained from coupons were extrapolated to the entire CSE vessel surface area for a check on material balance calculations. Since >99% of the interior surfaces in the CSE vessel were coated with Phenoline 302 paint, the data used for this material were prorated between condensing and noncondensing surface as shown in Table 5.22. The two methods of estimating the fractional mass deposited and retained on vessel surfaces are compared in Table 5.23. The coupon data suggest that more iodine was retained by the CSE vessel paint than the overall material balance (Tables 5.1 through 5.6) would indicate. The material balance values are believed to be more accurate, but the coupon data substantiate that the relatively high absorption of iodine by the vessel surfaces, about 44% average of the mass of iodine released, is reasonable.

TABLE 5.22. Deposition on Vessel Surfaces Inferred from Coupon Data (a)

Run	Injected Mass, gram (b)		Fraction of Injected Mass on Vessel Surfaces					
			Heat Transfer Surface (c)		Non Heat Trans. Surface (d)		Total Vessel Surface	
			I	Cs	I	Cs	I	Cs
D-1	0.043	0.305	-	-	0.405	0.0216	0.405	0.0216
D-2	0.061	0.309	-	-	0.406	0.0296	0.406	0.0296
A-1	0.695	0.367	0.043	0.00625	0.075	0.00245	0.118	0.0087
A-2	56.2	3.61	0.140	0.00206	0.520	0.00408	0.660	0.00614
A-5	84.51	1.10	0.955	0.069	1.04	0.084	1.995	0.153
A-11	98.36	5.02	1.09	.033	0.668	0.034	1.758	0.067

- a. Phenoline 302 coupons
b. From Tables 5.1 through 5.6
c. 327 m² in CSE main room
d. 242 m² in CSE main room

TABLE 5.23. Comparison of Deposition by Coupon Data^(a)
with Material Balance Calculations

Run	Fraction of Injected Mass on Vessel Surfaces ^(d)			
	Iodine		Cesium	
	Coupon ^(b) Data	Material ^(c) Balance	Coupon ^(b) Data	Material ^(c) Balance
D-1	0.405	0.210	0.0216	(-)0.0464
D-2	0.406	0.347	0.0296	0.1926
A-1	0.118	0.424	0.0087	0.052
A-2	0.660	0.365	0.00614	0.044
A-5	1.995	0.747	0.153	0.0978
A-11	<u>1.758</u>	<u>0.528</u>	<u>0.067</u>	<u>0.415</u>
Average	0.890	0.437	0.0475	0.126

a. Phenoline 302 coupons.

b. From Table 5.22.

c. From Tables 5.1 through 5.6.

d. After 1 day exposure

F. DISTRIBUTION WITHIN CONTAINMENT VESSEL

Material balances can be made as a function of time by knowledge of the mass injected, mass airborne and mass accumulated in the vessel sumps. Typical examples of such calculations are shown in Figures 14 and 15 for iodine and 16 and 17 for cesium. These figures show that both iodine and cesium were transferred rapidly from the gas phase to structural surfaces and then slowly washed into the sumps by condensing steam. As expected, the rate of washing into the sumps was highest in the experiment with uninsulated walls (Run A-2), and a higher fraction of iodine was irreversibly absorbed by the paint in the experiment with lower steam condensation rates (Run A-5).

The curve for mass on surfaces was calculated by difference and represents the sum of the masses absorbed by the paint and in the flowing condensate film. At long times this is essentially the mass in the paint.

FIGURE 14. Iodine Distribution Within Containment as a Function of Time -- Run A-2

FIGURE 15. Distribution of Total Iodine in Containment-Run A-5

FIGURE 16. Cesium Distribution Within Containment as a Function of Time -- Run A-2

FIGURE 17. Distribution of Cesium in Containment-Run A-5

VI. COMPARISON OF THEORY WITH EXPERIMENT

The adequacy of any theoretical model for general use in predicting the time dependent concentration in the gas space cannot be confirmed by a single experiment or even by a series of experiments unless the model faithfully predicts the effects of changes in all-important parameters. Agreement might be the result of a fortuitous choice of values used for parameters in the mathematical expressions. As an example, Rosenberg, et al.⁽¹⁷⁾ suggest that reaction of elemental iodine with paint might be the limiting rate mechanism for transfer of iodine from the containment atmosphere to vessel surfaces. They published Arrhenius type curves of reaction rate versus temperature for several typical containment vessel coatings. If reaction with paint is the rate limiting step, removal of iodine from the atmosphere should be faster at higher temperatures than at lower temperatures. However, reference to Table 5.15 shows that in the CSE tests the higher temperature experiments (Runs A-5 and A-11) gave a longer concentration half life (slower removal) than did the lower temperature tests (Runs A-1 and A-2). If only the results of Runs A-5 and A-11 had been available for verification, the agreement with the paint reaction limiting model would have been fairly good, as shown in Table 6.1. However, the temperature effect observed in the CSE experiments was just opposite to that predicted if reaction with paint were limiting. This is a strong indication that some other mechanism was controlling. It will be shown later that gas phase mass transfer across the boundary layers is the rate limiting step for all six of the CSE experiments reported here plus the initial periods of five others where only natural effects occurred. The reason for the slower removal at higher temperatures was that thermal insulation was

installed on the vessel for the 250 °F experiments, while the 180 °F tests were with bare walls. Heat transfer rates and mass transfer coefficients were lower in the 250 °F tests, resulting in lower iodine fluxes, in agreement with the natural convection model prediction.

TABLE 6.1. Paint Reaction Model Versus Experiment

	<u>Concentration Half Life, min</u>	
	<u>180 °F</u>	<u>250 °F</u>
Predicted by Paint Reaction Model ^(a)	35	12
<u>CSE Experiment</u>	10 ^(b)	18 ^(c)

a. Based on main CSE room total surface area, Phenoline 302 paint, condensing steam atmosphere (Reference 17, p. 72).

b. Average of Runs A-1 and A-2 for inorganic iodine.

c. Average of Runs A-5 and A-11 for inorganic iodine.

The point of the foregoing example is that correct prediction of trends may be more important than agreement at one particular set of conditions for model verification purposes.

The ultimate objective of all fission product transport models is to predict the amount of radioactive matter that would leak from the containment vessel in event of an accident. The rate of release to the environs is usually calculated by an equation such as Equation (2)

$$Q_i = a L_{avg} V f_p C_{avg} \quad (2)$$

where

Q_i = rate of release of species i from vessel,
Ci/min

a = specific activity of released material, Ci/g

L_{avg} = fractional leak rate of contained gases, min⁻¹

V = volume of contained gas at containment conditions, m^3

f_p = fractional penetration through leak points

C_{avg} = average mass concentration in the contained gas space for species i , g/cm^3 .

If a , L_{avg} , V and f_p are constant and independent of C_{avg} , as is normally assumed, then the rate of release of fission product from the vessel is directly proportional to the concentration in the gas space. Thus the chief objective of most mathematical models of fission product behavior is to predict the concentration in the gas space. Factors which may be important to elemental iodine behavior include:

- heat transfer rate from gas to surfaces,
- vessel volume to surface area ratio,
- total iodine mass released.
- reaction with paint or other surface material,
- gas-liquid equilibrium partition,
- presence of forced convection devices,
- atmosphere temperature and pressure, and
- gas volume to liquid volume ratio.

Additional parameters, such as particle size and electrical charge, should be considered for aerosol behavior.

The present series of experiments in the CSE, though limited in number, covered a range of values for all the above parameters except the type of paint and the ratio of gas volume to liquid volume. The latter was set in all the experiments at an initial value of V/L of about 600, which should give pessimistic results as far as removal of iodine from the gas phase is concerned. After 2 hr this ratio had decreased to about 400 to 500, depending on the particular experiment, due to accumulation of condensed steam. This is still conservative compared to values of about 130 expected in a typical large PWR in the absence of emergency core cooling,

containment spray or other liquid injection systems. Typical PWR values of V/L were not reached in the CSE tests until 15 to 30 hr after release of simulated fission products.

The effect of changing each of the aforementioned parameters is evaluated in the following sections by comparing fission product behavior in different tests and where possible with the model proposed by Knudsen and Hilliard.⁽⁸⁾

A. HEAT TRANSFER

The natural convection model described by Knudsen and Hilliard⁽⁸⁾ was used to predict temperature differences within the vessels resulting from natural heat transfer. Table 6.2 lists the temperature difference across the inside gas film, $T_b - T_i$, and the condensing steam flux, n_s , predicted for each test, along with the measured values when available. Predictions were made for both laminar and turbulent boundary assumptions. A schematic drawing of the temperature profile near and through the outer containment vessel wall is shown in Figure 18. Figure 19 is a schematic illustration of the partial pressure profiles for air and steam near the wall. Heat transfer is not as strongly dependent on whether the boundary is laminar or turbulent as is mass transfer to a perfect sink, but Table 6.2 indicates that the boundaries were turbulent. Agreement ranged from fair to excellent.

TABLE 6.2. Predicted Versus Measured Heat Transfer

CSE Run	$(T_b - T_i) \text{ } ^\circ\text{F}$			Steam Condensation Flux, lb/hr ft ²		
	Predicted		Measured	Predicted		Measured
	Laminar	Turb.		Laminar	Turb.	
D-1	8.9	7.8	(a)	0.28	0.30	0.38
D-2	8.9	7.8	(a)	0.275	0.29	0.32
A-1	19.0	7.9	(a)	0.11	0.097	0.096
A-2	19.0	7.9	5.5	0.12	0.105	0.11
A-5	2.2	1.6	1.4	0.045	0.035	0.032
A-11	2.2	1.6	(a)	0.045	0.035	0.046

a. not measured

FIGURE 18. Schematic Temperature Profile Through Containment Vessel Wall

FIGURE 19. Partial Pressures Near Wall

B. INITIAL REMOVAL RATE OF IODINE FROM THE ATMOSPHERE

The natural convection model⁽⁸⁾ was used to predict the initial removal rates of elemental iodine from the atmosphere. The value of the exponential removal rate constant, λ , depends on whether laminar or turbulent boundaries are assumed. In large containment vessels, including the CSE, a transition probably occurs from laminar to turbulent, with very large vessels being nearly entirely turbulent. Hales⁽²⁰⁾ shows that transition should occur at distances of 5 to 10 ft from the leading edge of a cold wall immersed in an atmosphere typical of postaccident conditions in a PWR containment vessel. Values predicted for transition from laminar to turbulent at $x = 10$ ft are listed in Table 6.3. It appears that the Drywell tests, Runs D-1 and D-2, were largely laminar and the main CSE mostly turbulent. The predictions using a transition from laminar to turbulent at 10 ft down from the top of the vertical walls gave the best overall agreement.

TABLE 6.3. Comparison of Predicted Initial Removal of Inorganic Iodine with CSE Experiments

Run No.	Measured ^(a) Initial Half-Life, min	Predicted Half Life, min ^(b)			Ratio Predicted to Measured
		Laminar	Turbulent	Transition at L = 10 ft	
D-1	8.0 $\pm$ 1.0	9.6	5.2	7.7	0.963
D-2	9.5 $\pm$ 0.5	9.7	5.2	7.8	0.821
A-1	9.0 $\pm$ 4.0	19.0	8.9	10.0	1.111
A-2	10.8 $\pm$ 2.0	18.9	8.9	9.9	0.917
A-5	15.2 $\pm$ 1.5	33	14.4	16.1	1.059
A-11	21 $\pm$ 4	33	15.0	16.8	<u>0.800</u>
Average					0.945

a. Inorganic iodine, Table 5.15.

b. Using method of Reference (8) and atmospheric properties of Appendix C.

Several experiments were made in the main CSE vessel in which either containment sprays or recirculation filter loops were operated after an initial period in which only deposition by natural effects occurred. Table 6.4 lists the measured concentration half lives for the initial period of seven tests conducted under essentially identical conditions. The average experimental value was 15.0 min with a standard deviation of 1.9 min. The natural convection model predicts 16.1 min, in good agreement.

TABLE 6.4. Reproducibility of Initial Removal Rate for Elemental Iodine in CSE Tests (a,b)

<u>Run No.</u>	<u>Measured Concentration Half Life, m/n</u>
A-5	13.5
A-6	12.0
A-7	16.5
A-8	17.0
A-9	14.0
A-11	16.0
A-14	<u>16.0</u>
Average	15.0
Std. Dev.	1.85

- a. All experiments in main CSE vessel, 250 ± 5 °F, 47 ± 4 psia, 1 in. of thermal insulation, $C_{go} \sim 100$ mg/m³, and 10-min release period.
- b. Predicted half life by natural convection model is 16.1 min for these test conditions.

C. IODINE EQUILIBRIUM

The initial exponential removal rate of inorganic iodine lasted only until the concentration had decreased to about 1% of its initial value, after which the concentration decreased at a much slower rate. This is in agreement with the natural convection theory⁽⁸⁾ assumption of approach to equilibrium between gas and liquid, in accordance with Equation (3).

$$\frac{C_g}{C_{go}} = \left(1 - \frac{V}{HL}\right) e^{-\lambda_1 t} + \frac{V}{HL} e^{-\lambda_2 t} \quad (3)$$

where C_g = concentration in bulk gas at time t
 C_{go} = concentration at time zero
 V = gas volume
 H = equilibrium gas-liquid distribution coefficient
 L = liquid volume
 λ_1 = gas mass transfer limited rate constant
 λ_2 = slow chemical reaction rate constant
 t = time.

Methods for evaluating the rate constants λ_1 and λ_2 are presented in Reference (8). The most uncertain parameter is H , the iodine equilibrium gas-liquid distribution coefficient.

$$H = \frac{C_L}{C_g} \text{ equilibrium} \quad (4)$$

Eggleton⁽³²⁾ presented theoretical predictions for the value of H , based on known rapid hydrolysis reactions. He predicted that H would depend strongly on iodine concentration in the liquid and on pH, and weakly on temperature. He also pointed out that slow reactions which he ignored would tend to increase the magnitude of H with time.

It was difficult to measure H in the CSE tests at very early times because the iodine distribution between gas and liquid phases did not attain equilibrium immediately after release of iodine to the gas phase. However, estimates of the values of H were obtained by measuring liquid and gas concentrations and plotting the ratios as a function of time. When the value of the ratio became essentially constant it was assumed that a pseudo-equilibrium was attained. Table 6.5 lists the concentrations and ratios for the three low iodine concentration experiments (D-1, D-2 and A-1) and Table 6.6 the same data for the high iodine concentration experiments (A-2, A-5 and A-11). The total inorganic iodine in the gas phase is used rather than elemental iodine, since this gives conservative values of H .

The average value of H for all four of the main CSE tests was about 6000, with very little dependence on iodine concentration, pH or temperature. Values at $t = 2$ hr and at 24 hr are compared with Eggleton's predictions in Table 6.7. Comparison with theory was poor at the early time, as expected, but improved with time. Run A-1 was the only experiment which gave results lower than theory. This may have been due to some methyl iodide being included as inorganic iodine. Considering the complexity of the system, the overall agreement must be considered satisfactory, though the predicted effects of iodine concentration and pH were not observed.

TABLE 6.5. Iodine Gas-Liquid Distribution Coefficients for Low Concentration Experiments (a)

Time, hr	Run D-1			Run D-2			Run A-1		
	$C_L^{(b)}$	$C_g^{(c)}$	C_L/C_g	$C_L^{(b)}$	$C_g^{(c)}$	C_L/C_g	$C_L^{(d)}$	$C_g^{(c)}$	C_L/C_g
1	5.3 (1)	4.0 (-3)	1.3 (4)	6.6 (1)	5.5 (-3)	1.2 (4)	1.1 (2)	2.6 (-2)	4.2 (3)
2	4.6 (1)	5.8 (-4)	7.9 (4)	5.6 (1)	4.0 (-4)	1.4 (5)	2.7 (1)	1.0 (-2)	2.7 (3)
4	3.4 (1)	3.7 (-4)	9.2 (4)	4.2 (1)	2.8 (-4)	1.5 (5)	8.4 (0)	3.6 (-3)	2.3 (3)
8	2.6 (1)	2.8 (-4)	9.3 (4)	2.3 (1)	2.6 (-4)	8.8 (4)	3.2 (0)	1.9 (-3)	1.7 (3)
12	1.1 (1)	1.8 (-4)	6.1 (4)	1.5 (1)	1.9 (-4)	7.9 (4)	2.3 (0)	1.2 (-3)	1.9 (3)
16	6.0 (0)	1.4 (-4)	4.3 (4)	1.2 (1)	1.6 (-4)	7.5 (4)	1.7 (0)	7.0 (-4)	2.4 (3)
20	3.3 (0)	1.0 (-4)	3.3 (4)	1.0 (1)	1.3 (-4)	7.7 (4)	1.3 (0)	4.1 (-4)	3.2 (3)
24	1.9 (0)	7.7 (-5)	2.5 (4)	9.0 (0)	1.0 (-4)	9.0 (4)	1.0 (0)	2.5 (-4)	4.0 (3)
Avg. Ratio	-	-	6.1 (4)	-	-	6.3 (4)	-	-	2.6 (3)
Avg. pH	(e)			5.1 ± 0.1			7.1 ± 0.1		

- a. Numbers in parenthesis are exponents of base 10.
b. Iodine concentration in liquid pool, $\mu\text{g/l}$.
c. Total inorganic iodine concentration in gas space, $\mu\text{g/l}$.
d. Iodine concentration in wall film, $\mu\text{g/l}$.
e. Not measured.

6.10

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TABLE 6.6. Iodine Gas-Liquid Distribution Coefficients for High Concentration Experiments (a)

Time, hr	Run A-2			Run A-5			Run A-11		
	C_L (b)	C_g (c)	C_L/C_g	C_L	C_g	C_L/C_g	C_L	C_g	C_L/C_g
1	4.5 (4)	3.0 (0)	1.5 (4)	1.0 (5)	9.0 (0)	1.1 (4)	1.5 (5)	1.5 (1)	1.0 (4)
2	2.3 (4)	2.7 (-1)	8.5 (4)	3.0 (4)	7.8 (-1)	3.8 (4)	8.0 (4)	2.5 (0)	3.2 (4)
4	7.5 (3)	1.3 (-1)	5.8 (4)	3.5 (3)	2.9 (-1)	1.2 (4)	1.3 (4)	9.4 (-1)	1.4 (4)
8	1.7 (3)	8.0 (-2)	2.1 (4)	1.0 (3)	1.8 (-1)	5.6 (3)	3.0 (3)	5.6 (-1)	5.4 (3)
12	7.0 (2)	5.6 (-2)	1.3 (4)	6.5 (2)	1.6 (-1)	4.1 (3)	2.0 (3)	3.4 (-1)	5.9 (3)
16	3.9 (2)	4.0 (-2)	9.8 (3)	5.6 (2)	1.3 (-1)	4.3 (3)	1.5 (3)	2.1 (-1)	7.1 (3)
20	2.5 (2)	2.7 (-2)	9.3 (3)	5.0 (2)	1.2 (-1)	4.2 (3)	1.2 (3)	1.6 (-1)	7.5 (3)
24	1.7 (2)	1.9 (-2)	9.0 (3)	4.5 (2)	1.0 (-1)	4.5 (3)	1.1 (3)	1.5 (-1)	7.3 (3)
Avg. Ratio	-	-	2.0 (4)	-	-	5.8 (3)	-	-	7.9 (3)
Avg. pll	6.4 $\pm$ 0.4			5.4 $\pm$ 0.4			7.1 $\pm$ 0.3		

a. Numbers in parenthesis are exponents of base 10.

b. Iodine concentration in wall film, $\mu\text{g}/\text{l}$.

c. Total inorganic iodine concentration in main gas space, $\mu\text{g}/\text{l}$.

TABLE 6.7. Comparison of Measured Values of H with Eggleton's Predictions^(a)

Run No.	t = 2 hr			t = 24 hr		
	C_L/C_g Expt.	H Ref (32)	Ratio Expt/ Ref (32)	C_L/C_g Expt.	H Ref (32)	Ratio Expt/ Ref (32)
D-1	7.9 (4)	2.3 (3)	34	2.5 (4)	5.0 (4)	0.5
D-2	1.4 (5)	1.8 (3)	78	9.0 (4)	1.0 (4)	9.0
A-1	2.7 (3)	5.0 (4)	0.05	4.0 (3)	1.0 (6)	0.004
A-2	8.5 (4)	3.0 (1)	2800	9.0 (3)	1.0 (3)	9.0
A-5	3.8 (4)	3.2 (1)	1200	4.5 (3)	1.3 (3)	3.5
A-11	3.2 (4)	1.2 (2)	265	<u>7.3 (3)</u>	1.0 (4)	0.73
Avg. of all tests				2.3 (4)		
Avg. of 4 main CSE tests				6.2 (3)		

a. Based on Reference (32) and Tables 6.5 and 6.6.

D. EFFECT OF VESSEL SIZE

A macro mass balance for iodine in the gas space gives:

$$-\frac{dC}{dt} = \left(\frac{A}{V}\right)(n_I)_{\text{avg}} \quad (5)$$

where A = effective surface area for deposition
 V = gas volume on which mass balance made
 $(n_I)_{\text{avg}}$ = average mass flux of iodine to surfaces

Equation (5) shows the familiar dependency of removal rate on geometry. If the iodine flux is independent of size, the rate of concentration decrease is directly proportional to the surface area and inversely proportional to the gas volume. However, n_I is not necessarily independent of vessel size. Small vessels, such as those often used for mass transfer studies, probably have laminar boundaries, in which case the flux decreases with length of surface according to Equation 6.

$$n_I = b C_g x^{-1/4}, \text{ laminar} \quad (6)$$

where n_I = flux at distance x down vertical wall
 b = a constant

At a distance of about 5 to 10 ft down the vertical wall a transition occurs to turbulent boundaries⁽²⁰⁾ for natural convection conditions in a condensing steam atmosphere. The rest of the surface has a turbulent boundary where the iodine flux is independent of distance:

$$n_I = c C_g, \text{ turbulent} \quad (7)$$

where c = a constant

The point of this discussion is that experimental measurements of mass transfer rates made in small vessels will not give a correct estimate of removal rate in large vessels by applying the simple scaling factor A/V . It is best to combine both laminar and turbulent calculations, with a transition at a distance of about 10 ft down the wall, as was done in Table 6.3.

Another question concerning the effect of vessel size is how much of the vessel surface to use. All calculations made in this report used the entire surface exposed to the steam-air-iodine vapors in the gas space of concern. Other authors^(15,16,17) choose to use only the outer wall area in order to be conservative. The CSE experiments clearly show that all surfaces collect iodine at about the same rate (Tables 6.3 and 5.22). Only 3530 ft² of the CSE main room surface is exposed to the outside air, out of a total of 6140 ft². If the lower figure is used in predictive models, much poorer agreement with theory would be obtained.

The two sizes of vessels used in the present work gave about the same agreement with theory, using transition to turbulent boundary at $x = 10$ ft, even though the smaller Drywell vessel had all surfaces exposed to outside air and the larger vessel had about 40% of its total area as internal, nonheat transfer areas.

Although these observations indicate that all surfaces exposed to iodine should be used in predictive model calculations, it is difficult to derive a proper mass transfer coefficient for the interior nonheat transfer surfaces. Calculations made in later sections of this report used the same mass transfer coefficient for all surfaces, but this is an area which needs additional study.

E. COMPARISON OF EXPERIMENTAL CONDITIONS TO THOSE ANTICIPATED IN A PWR LOCA

The atmosphere within the containment vessel of a large PWR prior to a loss-of-coolant accident (LOCA) would normally be air at about 100 °F and 1 atm pressure, with <100% relative humidity. All surfaces exposed to this atmosphere would be at about the same temperature. Immediately after rupture of the primary piping system the containment atmosphere would consist of a mixture of steam, air and liquid fog drops at a higher temperature and pressure and at essentially 100% relative humidity, but the surfaces would still be at their initial temperature. Heat would be transferred rapidly to all surfaces by three mechanisms: (1) radiation, (2) conduction of sensible heat, and (3) mass transfer of water vapor to the surfaces with subsequent condensation and release of latent heat. It can be shown that the heat of condensation of steam greatly exceeds that of sensible heat transfer for early times, and prediction of heat transfer is therefore largely a mass transfer problem.

Shortly after the LOCA all surfaces would have a film of steam condensate which would drain by gravity to lower regions. This film would be replenished by more steam condensation as long as the surface was at a lower temperature than the atmosphere. Whether steam continues to condense on any specific surface or the liquid film evaporates depends on the local geometry, the overall vessel geometry and whether additional steam is released from the primary system. If the emergency core coolant system (ECCS) functions properly, there will not be a further release of steam. Heat will continue to be transferred to those surfaces which act as heat sinks, mostly the outer walls. As steam is condensed, the temperature and pressure of the atmosphere would decrease. Ultimately, some of the interior surfaces which are surrounded by the atmosphere would be warmed to equilibrium with the atmosphere. Further cooling of the atmosphere by continuous heat sink surfaces would then cause the atmosphere temperature to decrease to below the temperature of the interior surfaces. Any liquid film on the interior surfaces would then evaporate. Thus, the general state a few seconds after LOCA is one of all surfaces wet by condensate, but a short time later (perhaps about 10 min) some of the interior surfaces would be dry while condensation would continue on exterior walls and other heat sinks. The CSE natural effects experiments were conducted under conditions similar to the latter case. Steam was fed to the vessel at just the rate required to maintain a constant temperature in most of the experiments, but conditions were otherwise very similar to those which probably would exist in a large PWR containment vessel several minutes after coolant loss.

Knudsen and Hilliard⁽⁸⁾ developed equations and published charts for predicting iodine deposition to surfaces in a condensing steam-air atmosphere. For early times after an instantaneous release, they predict an exponential decrease in gas phase concentration.

$$\frac{C_g}{C_{go}} = e^{-\lambda_{nc} t} \quad (8)$$

where λ_{nc} = removal rate constant for natural convection
 t = time after release of iodine.

They showed that the removal rate constant, λ_{nc} , was the sum of two distinct effects:

(1) the sweep effect of condensing steam, and

(2) the diffusion of molecular iodine across the gas boundary due to a concentration gradient.

Their equations can be put in the form

$$\lambda_{nc} = \lambda_{sweep} + \lambda_{diff} = \frac{k_{sw} A_w}{V} + \frac{k_d A_t}{V} \quad (9)$$

where

k_{sw} = mass transfer coefficient due sweep effect
of steam condensation

A_w = surface area on which steam condenses

V = total gas volume of chamber

k_d = mass transfer coefficient due to diffusion

A_T = total surface area of chamber

The removal rates by the two effects can be expressed in terms of concentration half lives, the time required for the concentration to be reduced by half.

$$t_{1/2 \text{ sweep}} = \frac{0.693 V}{k_{sw} A_w} \quad (10)$$

$$t_{1/2 \text{ diff}} = \frac{0.693 V}{k_d A_t} \quad (11)$$

In Table 6.8 the predicted half lives for elemental iodine due to each effect by itself is listed for each of the six experiments. Also shown are the measured values. It is

TABLE 6.8. Comparison of Predicted Removal Rate for Elemental Iodine by Steam Condensation and by Diffusion

Run No.	Predicted $t_{1/2}$, min		Measured Half Life, min ^(c)
	By Steam Sweep ^(a)	By Diffusion Across B.L. ^(b)	
D-1	36	7.7	8.0 ± 1.0
D-2	38	7.8	9.5 ± 0.5
A-1	130	10.0	9.0 ± 4.0 ^(d)
A-2	122	9.9	9.0 ± 0.5
A-5	475	16.1	13.5 ± 0.5
A-11	500	16.8	16.0 ± 0.5

a. Calculated by Equation (10)

b. Calculated by Equation (11), using a transition from laminar to turbulent at $x = 10$ ft.

c. From Table 6.11.

d. For total inorganic iodine.

very clear that the sweep effect of condensing steam was relatively unimportant compared to diffusion in these experiments.

The presence of condensing steam in the containment vessel has other effects than that of increasing transfer rates across gas boundaries. The liquid water films and pools provide a sink for iodine due to the large gas-liquid partition coefficient. This causes the concentration difference across boundaries to be large for long time periods and the removal from the atmosphere to be essentially a first-order process. The liquid water also continues to react slowly with dissolved elemental iodine to produce nonvolatile iodine species,⁽³²⁾ which aids greatly in reducing the gas phase concentration.

In Figure 20 the steam condensation flux is plotted as a function of time for the six CSE experiments. Also shown is the predicted condensation flux for a typical large PWR, taken

FIGURE 20. Steam Condensation Flux as a Function of Time

from Reference (33). This figure shows that the CSE tests were conducted under conservative conditions of low condensation rates except for long times--times beyond those where most of the removal would have occurred in a PWR.

F. REACTION WITH PAINT

All interior surfaces of the CSE containment vessels were coated with a phenolic paint.* This coating was selected for its relatively long service life under the strenuous conditions of repeated cycles of exposure to steam and air at temperatures to 260 °F. Its behavior toward iodine is about average of the ten commercial coatings tested by Rosenberg, et al.⁽¹⁷⁾ Rosenberg measured the velocity of deposition of elemental iodine to Phenoline 302 painted coupons. The reaction rates of iodine with the paint is sufficiently rapid that it does not limit the transfer rate, as discussed previously and in Table 6.1.

Rosenberg reported that the capacity of Phenoline 302 for absorbing elemental iodine is about 0.6 mg/cm^2 for the 2-mil thick coupons he used, and proportionally greater for thicker paint films. Thus, the CSE paint, with 25-mil thickness, has a capacity for about 4×10^4 g of iodine. Only about 150 g were sorbed irreversibly in the four large-scale CSE tests, far below predicted saturation. This is borne out by Table 5.23 which shows about as much sorption in later tests as in early ones. Also, tests with fresh paint (Runs D-1 and A-1) had no differences in behavior, compared to subsequent tests, which could be attributed to the effect of paint age.

* Two coats of Phenoline 302 over 1 coat Phenoline 300 Orange primer. Total thickness 25 mils.

G. EFFECT OF TEMPERATURE AND PRESSURE

The natural convection model⁽⁸⁾ predicts that temperature and pressure will not have much effect on iodine removal rates in containment vessels. This is partly due to the fact that as the temperature is increased the pressure also increases in confined volumes of saturated steam-air mixtures (refer to Appendix C). Temperature and pressure have opposite effects on diffusivity, the chief property for gas phase controlled transfer. Figure 21 shows the dependence of the gas phase mass transfer coefficient, k_d , on bulk gas temperature. This figure shows that k_d is fairly insensitive to temperature over most of the range of interest for water reactor containment systems. Figure 22 shows that the half life of elemental iodine is likewise rather insensitive to atmospheric temperature, except for low temperatures near ambient.

The six CSE experiments do not permit a direct demonstration of the effect of temperature, since tests at different temperature also were accompanied by changes of area to volume ratio or wall thermal resistance. Both of these latter two factors have a stronger influence on half life than temperature. However, the general agreement with the natural convection model (Table 6.3) is evidence that the temperature effect is properly accounted for by the model.

H. METHYL IODIDE BEHAVIOR

The partition coefficient for methyl iodide-water systems is very low, being on the order of unity for typical containment postaccident conditions.⁽³⁴⁾ Therefore, the transfer from gas to liquid is liquid-phase controlled. A lower limit to the absorption rate in the wall film can be estimated by assuming that the liquid film is laminar and that no methyl

FIGURE 21. Effect of Atmosphere Temperature on Gas Phase Mass Transfer Coefficient for Elemental Iodine

FIGURE 22. Effect of Atmosphere Temperature on Elemental Iodine Half Life in CSE

iodide leaves the liquid at the liquid-paint interface.

Postma⁽³⁴⁾ gives the following expression for this lower limit case.

$$n_{MI} = C_{Li} \sqrt{k D_\ell} \tanh \left(\sqrt{k/D_\ell} \delta \right) \quad (12)$$

where n_{MI} = mass flux of methyl iodide to liquid surface

C_{Li} = mass concentration in liquid at the gas-liquid interface

k = first order reaction rate constant

D_ℓ = diffusivity of methyl iodide in liquid

δ = thickness of film.

The film thickness, δ , can be estimated from laminar flow theory. Experimental measurements in the PAT vessel in the Aerosol Development Facility (ADF)⁽¹⁴⁾ lead to the conclusion that the average value of δ for the main CSE vessel is about 0.1 cm, and is independent of steam condensation rate over a wide range of conditions. Solving Equation (12) for CSE Run A-5 conditions, using $H = 1.2$ and $k = 3.3 \times 10^{-3} \text{ sec}^{-1}$, gives a predicted concentration half life of 165 hr for methyl iodide in the gas space.

An upper limit to the absorption rate can be made by assuming that the liquid film is well-mixed. Postma gives the solution⁽⁶⁾ as

$$\text{Absorption Rate} = k C_{Li} V_{WF} + C_{Li} L_F \quad (13)$$

where

V_{WF} = volume of liquid held up on walls and deck

L_F = volumetric flow rate of liquid film.

Solving Equation (13) for Run A-5 conditions gives an estimated half life of 85 hr.

The observed half life for Runs A-5 and A-11, taken from Figures B-17 and B-18, averaged 26 ± 12 hr. This is significantly faster removal from the gas phase than predicted even when using the optimistic assumption of well-mixed liquid films. Three explanations are possible. Methyl iodide may react with Phenoline paint. The first-order reaction rate may be faster than the value used in the calculation, which is extrapolated from data at lower temperatures.⁽³⁵⁾ Or the liquid holdup in the main room might be higher than calculated, based on 0.1 cm average film thickness. The latter is the more probable error, since the drywell pool and the thick layer of condensation on the horizontal deck certainly were much above 0.1 cm thickness.

The practical conclusion regarding methyl iodide behavior is that removal from the gas space is insignificant during the first 2 hr after release, but can be significant after several days.

I. PARTICULATE AEROSOL BEHAVIOR

Most of the fission products which would be released from an overheated nuclear reactor core have very low vapor pressures at the temperature which would exist in the containment vessel atmosphere. Only the rare gases and halogens can exist as gases; all other fission products and fuel substances would very rapidly condense to form either liquid or solid particles. Although the mean primary particle size is probably $<0.1 \mu$ immediately after condensation from the vapor state, the particles rapidly grow in size by agglomeration and by condensation of moisture from the steam-air atmosphere in the containment vessel.

Postma⁽⁶⁾ has discussed the growth of soluble and insoluble particles in humid environments and concluded that both types would act as condensation nuclei for water vapor. The equilibrium size would depend on the relative humidity of the atmosphere and the mass of the particulate substance serving as the nucleus. Growth to large sizes are possible for design basis accident (DBA) conditions. Visual observations of the atmosphere in the CSE experiments confirm that relatively large fog droplets form when the aerosol was injected into the saturated steam-air atmosphere (Figure 9).

If fission product matter is held within the fog drops, the effective size of the particle in the containment atmosphere is that of the fog drops, not of the primary fission product particles produced in the reactor core. If the mass of the primary particle is small compared to the mass of water condensed on it, as seems probable from Postma's calculations, then the density of the resultant particle is nearly that of liquid water at the containment conditions.

Particles can be removed from the atmosphere in closed vessels by a number of mechanisms, among which are

- gravitational settling,
- diffusion to surfaces,
- diffusiophoresis,
- thermophoresis,
- electrical attraction to surfaces, and
- removal by active engineered safety systems.

In the absence of active engineered safety features, only the first three are significant for the conditions of the CSE tests. A material balance on the gas space of a single chamber, with instantaneous release and a well-mixed atmosphere gives

$$\frac{C_g}{C_{g_0}} = \exp \left[- \left(\frac{u_t A_{cs}}{V} + \frac{k_w A_T}{V} + \frac{A_w N_s RT}{18 PV} \right) t \right] \quad (14)$$

where u_t = terminal settling velocity of particles
 A_{cs} = horizontal cross-sectional area of vessel
 V = gas volume
 k_w = effective mass transfer coefficient due to
 turbulent and Brownian diffusion
 A_w = heat transfer surface
 A_T = total surface area
 N_s = condensing steam flux
 R = gas constant
 T = absolute temperature in bulk gas
 P = total absolute pressure
 t = time after release.

The first term in the exponent represents the removal by gravitational settling, the second term is diffusion to surfaces, and the third term is an approximation of the diffusiophoretic effect. Thus the first term is the rate of transfer to the floor and the second and third terms predict the transfer to all surfaces. Equation (14) can be used to predict the concentration of particulate matter in the containment atmosphere if the values of all the terms are known beforehand. All parameters except u_t and k_w are known from the structural design features and by heat transfer calculations made for the conditions of interest. Unfortunately, the magnitudes of u_t and k_w depend strongly on the particle size.

Attempts were made during the CSE experiments to measure the particle sizes as they existed in the containment atmosphere. Samples of the atmosphere were drawn through cascade impactors inserted directly into the vessel through air locks. The impactors were preheated to 10 to 20 °F above the containment atmosphere temperature to avoid condensation within the instrument. As Postma⁽⁶⁾ has discussed, it is very likely that the fog droplets containing the fission

product particulate matter were partially evaporated due to the change in relative humidity caused by heat transfer and pressure changes as the gas passed through the impactor. The mass median diameter (MMD) determined by this technique, assuming a particle density of unity, ranged from a high of 4 μ in Run A-5 at early times to 0.3 μ late in Run A-11. The data were erratically variable with time, but generally decreased with longer containment times. It is concluded that the measurements are unreliable and at best give a minimum size range.

The effective particle size in the CSE tests can be estimated by measurements of concentration changes in the gas space and by collection rate in the wall condensate troughs. Table 6.9 lists the fractions of cesium collected on walls and on the floor. It shows that 75 to 92% of the cesium settled to the floor. This information is used in Table 6.10 to correct the observed concentration half life to that which was due only to gravitational settling. The effective terminal settling velocity was then calculated by Equation (15).

TABLE 6.9. Cesium Distribution Between Walls and Floor

	<u>Run A-1</u>	<u>Run A-2</u>	<u>Run A-5</u>	<u>Run A-11</u>
Mass to Wall Condensate, mg ^(a)	46.2	450	31.7	287
Mass Lost from Gas Space, mg ^(b)	367	2950	419	1120
Fraction to Walls	0.126	0.152	0.0756	0.256
Fraction to Floor	0.874	0.847	0.924	0.744

a. Calculated from wall trough liquid concentration and flow rate

$$\text{Mass} = \int_0^{24h} \frac{C_L N_s A_w}{\rho_L} dt$$

b. From Figures B.39 through B.42.

$$(u_t)_{\text{eff}} = \frac{0.693 V}{(t_{1/2})_{\text{settling}} A_{\text{cs}}} \quad (15)$$

Once the terminal settling velocity is known, the particle size can be calculated from Stoke's Law, which holds for particles of unit density with diameters less than about 40 μ .

$$u_t = \frac{g d_p^2 (\rho_p - \rho_f)}{18 \mu_f}$$

where

g = acceleration due to gravity

d_p = particle diameter

ρ_p = density of particle

ρ_f = density of fluid through which particle settles

μ_f = viscosity of fluid

Using the properties of the steam-air atmosphere as reported by Knudsen,⁽³¹⁾ the relation between particle size and terminal velocity was calculated and plotted in Figure 23 for 180 °F and for 250 °F. The resulting effective particle diameters, listed in Table 6.10 for cesium and 6.11 for uranium show that early in the experiments the particles were 10 to 15 μ in diameter, decreasing to 3 to 7 μ after a few hours. This is significantly larger than the cascade impactor measurements showed. This method should give a fairly accurate estimate of the effective particle sizes, since the diameter varies only as the square root of the concentration half life, which is believed to have been measured to within about $\pm 40\%$. Thus the values listed in Table 6.10 and 6.11 should be accurate to about $\pm 20\%$.

The size of the uranium and cesium particles were about the same, in agreement with the hypothesis of uranium and cesium both existing in liquid water fog droplets.

FIGURE 23. Terminal Settling Velocity of Fog Drops in Steam-Air Atmosphere

TABLE 6.10. Effective Cesium Particle Diameters in Containment Atmosphere

	Run D-1	Run D-2	Run A-1		Run A-2		Run A-5		Run A-11		
			Early	Late	Early	Late	Early	Late	Early	Middle	Late
Observed $t_{1/2}$ (a) In Gas, min	8.5	10.0	14	66	16.5	40	38	105	50	360	540
$t_{1/2}$ due to Settling, min(b)	10.5(c)	12.5(c)	16	75	19.5	47	40	112	67	484	725
v_t , ft/min(d)	1.60	1.34	1.86	0.396	1.52	0.632	0.743	0.265	0.443	0.0614	0.0410
d_{eff} , ν (e)	16.5	15.0	18.5	8.6	16.0	10.3	11.2	6.7	8.6	3.2	2.7

a. From Figures B-37 through B-42 in Appendix.

b. Estimated by deducting mass transferred to walls.

c. Assuming 20% of transfer was to walls.

d. Calculated by Equation (15).

e. From Figure 23.

TABLE 6.11. Effective Uranium Particle Diameters in Containment Atmosphere

	Run D-2	Run A-1		Run A-2		Run A-5		Run A-11	
		Early	Late	Early	Late	Early	Late	Early	Late
Observed $t_{1/2}$									
In Gas, min ^(a)	8.5	21	75	21	40	40	108	25	425
$t_{1/2}$ Due to Settling, min ^(b)	10.5 ^(c)	24	86	25	47	43	115	34	570
u_t , ft/min ^(d)	1.60	1.24	0.345	1.19	0.632	0.690	0.258	0.874	0.052
d_{eff} , μ ^(e)	16.4	15.0	6.8	14.7	10.8	10.8	6.6	12.0	2.9

a. From Table 5.18.

b. Estimated by deducting mass transferred to walls.

c. Assuming 20% of transfer was to walls.

d. Calculated by Equation (15).

e. From Figure 23.

Further evidence that gravitational settling was responsible for the major portion of the removal of particles from the atmosphere is given in Table 6.12 and 6.13. A stainless steel panel measuring 2 by 2 ft was placed in a horizontal position near the deck in Run A-11. A similar panel was placed in a vertical position nearby. Both were sheltered by a 3 by 3 ft panel to prevent condensate from dripping on them. The panels were exposed for the entire experiment and then removed and decontaminated. Table 6.12 shows that extrapolation of the deposited cesium to the entire vessel floor could account for 65% of the removal, in fair agreement with the 74% calculated by wall condensate in Table 6.9.

TABLE 6.12. Fraction of Cesium Settling to Floor in Run A-11

Deposition on 4-ft ² horizontal panel	5.95 mg
Deposition density	1.49 mg/ft ²
A _{CS} , floor area	490 ft ²
Deposition on entire floor	730 mg
Loss from gas	1120 mg
Fraction to floor	0.652

TABLE 6.13. Effect of Orientation on Cesium and Iodine Deposition

<u>Panel Surface Orientation</u>	<u>Relative Deposition^(a)</u>	
	<u>Cesium</u>	<u>Iodine</u>
Upward facing (horizontal)	1 (b)	1 (b)
Vertical	0.043	0.038
Downward facing (horizontal)	0.00165	0.0077

a. 2 by 2 ft stainless steel panels near deck, non-condensing surface. Run A-11.

b. Extrapolation to entire floor area accounts for 65% of cesium loss from gas; 2.3% of iodine loss from gas.

Table 6.13 shows that only minor amounts of both iodine and cesium deposited on downward and sideway facing surfaces. Extrapolation of the iodine deposition to the entire vessel floor indicates that only 2.3% of the iodine was removed from the gas space by gravitational settling. This small value is in agreement with the Maypack identification of >90% of the iodine in gaseous forms.

J. COMPARISON OF NATURAL REMOVAL RATES WITH REMOVAL BY SPRAYS AND FILTER RECIRCULATION

A total of 19 large-scale experiments were performed during the CSE fission product transport study, of which only the six natural effects tests are discussed in this report. Eight spray experiments are reported separately,⁽⁵⁻⁷⁾ and five tests with a recirculation mode air-cleaning loop will be documented later.

The relative effectiveness of the three fission product removal systems depends on the vessel size and on the manner in which the active safety features (spray or filter loop) are operated. However, a direct comparison for several specific cases is possible by comparing the results of CSE tests. In Figure 24 the concentration of total iodine in the main room gas space is plotted for three experiments conducted under similar conditions except that in one (Run A-11) only natural removal effects occurred, in another (Run A-10) a caustic-borate spray was operated essentially continuously for 24 hr, and in the third (Run A-14) a filter-adsorber air-cleaning loop ran continuously for 24 hr, starting 30 min after the beginning of the aerosol release. The fission product simulant was released during the first 10 min in all three tests. In order to make the time scales coincide, the 30-min initial period in Run A-14 was ignored and time zero was set at the beginning of the loop operation. The pertinent conditions for the three experiments are listed in Table 6.14.

TABLE 6.14. Test Conditions for Runs A-10,
A-11 and A-14

	Run A-11	Run A-10	Run A-14
Type of Test	Natural (Temp. Decay)	Spray	Filter Loop
CV Temperature, °F	250	250	250
Pressure	49	48	48
C_{go} , mg/m ³			
Total Iodine	165	160	100
Methyl Iodide	1.5	5	5
Cesium	8	10	3
Uranium	3	2	1.5
Spray Solution	-	NaOH + H ₃ BO ₃ pH 9.4	-
Spray Flux, gpm	-	0.1	-
Fresh Spray Period, min	-	0 to 40	-
Recirculation Spray Period, min	-	50 to 1400	-
Spray Drop Size, μ	-	1200, MMD	-
Filter Loop Components	-	-	Condenser, Demister Rough Filter Absolute Filter 3 Charcoal Filters
Filter Recirculation Rate, Main Room Volumes/hr	-	-	2.9

Figure 24 shows that, as expected, the sprays were the most effective at very early times. However, after the first half hour the filter loop was the most effective, due to its ability to remove methyl iodide more rapidly than the other two systems.

The real test for effectiveness of removal systems is the reduction in the fraction of fission products escaping the vessel. Assuming that the gas leakage rates were identical in the three tests, the relative amount of iodine which

FIGURE 24. Comparison of Total Iodine Removal Rates By Natural Effects, Sprays and Recirculation Filter Loop

leaked can be calculated by numerically integrating the concentration over the time period of interest. This can be expressed as a "dose reduction factor" (DRF). It is the ratio of the concentration if no reduction occurred to the average concentration during the specified periods.

$$DRF_t = \frac{t C_{go}}{\int_0^t C_g dt} \quad (17)$$

The DRF for the three tests is listed in Table 6.15 for two time periods--the critical initial 2-hr period and the first 24 hr. The $DRF_{2 \text{ HR}}$ were 4.2 for natural processes, 8.0 for the filter loop and 30 for sprays. It should be pointed out that natural effects undoubtedly contributed to removal during the spray and filter loop tests. The superiority of sprays diminished with time, and at 24 hr the $DRF_{24 \text{ HR}}$ were 30.5 for natural, 79 for filters and 101 for sprays. It is possible that at long containment times the DRF would become approximately equal for all three systems.

TABLE 6.15. Relative Effectiveness of Natural Effects, Sprays and Filter Loop Against Total Iodine

	Run A-11	Run A-10	Run A-14
Type	Natural	Spray	Filter Loop
C_g/C_{go} at $t = 2 \text{ hr}$	0.039	0.024	0.0065
Average C_g/C_{go} during first 2 hr	0.239	0.0334	0.126
$DRF_{2 \text{ HR}}^{(a)}$	4.2	30	8.0
C_g/C_{go} at $t = 24 \text{ hr}$	0.008	0.002	0.0015
Average C_g/C_{go} during first 24 hr	0.0327	0.0099	0.0126
$DRF_{24 \text{ HR}}^{(a)}$	30.5	101	79

a. Dose reduction factor. $DRF_t = \frac{t C_{go}}{\int_0^t C_g dt}$

VII. APPLICATION TO LARGE POWER REACTOR CONTAINMENT SYSTEMS

All large PWR nuclear plants built in the USA are equipped with active engineered safety features which are intended to limit the consequences of accidental release of radioactive material from the primary system. These engineered systems are highly reliable and all evidence indicates that, in the unlikely event of a major accident, they would rapidly reduce the rate of leakage of radioactive matter from the containment vessel.

An estimate of the maximum amount which would escape containment in the absence of any active engineered safety system is possible by use of mathematical models based on the results of the CSE natural effects experiments. An illustration of how the models can be applied to a typical PWR follows, but it should be emphasized that this is merely to obtain an estimate of the lower limit of decontamination. Operation of containment sprays or a recirculation air cleaning loop would certainly increase the decontamination appreciably above that calculated in the illustration.

The postulated conditions and values of parameters used in the calculations are listed in Table 7.1. The basic assumptions are:

- instantaneous release of fission products to the containment vessel, with no more release
- 5% of the total iodine released as methyl iodide
- the vapor space is well mixed at all times
- the liquid films and pool are well mixed at all times
- all heat transfer surfaces are wet by a liquid film, other surfaces are dry
- the vapor temperature is in quasi-equilibrium with the surfaces

- the steam-air atmosphere is saturated with water vapor
- all surfaces are reactive toward iodine with rate constant listed in Table 7.1
- the particles are 8 μ diameter
- the only removal is by natural effects.

TABLE 7.1. Postulated Conditions for a Large PWR

Parameter	Assumed Value
D, vessel diameter	125 ft
L, vertical height	150 ft
V, contained gas volume	$2 \times 10^6 \text{ ft}^3$
A_T , total surface area	$3.5 \times 10^5 \text{ ft}^2$
A_w , heat transfer surface	$1.0 \times 10^5 \text{ ft}^2$
A_{CS} , cross-sectional area	$1.2 \times 10^4 \text{ ft}^2$
h_w , wall heat transfer coefficient	$0.15 \text{ Btu}/(\text{hr})(\text{ft}^2)(^\circ\text{F})$
T_b , bulk vapor temperature	250 $^\circ\text{F}$
P, total pressure	48.6 psia
T_a , surrounding air temp.	80 $^\circ\text{F}$
H, iodine gas-liquid distribution coefficient	5000
V_{wf} , liquid wall film holdup	500 ft^3
L_o , liquid pool volume	$1 \times 10^4 \text{ ft}^3$
k_p , iodine-paint reaction rate constant	$3 \times 10^{-4} \text{ ft/hr}$

A. INORGANIC IODINE

The method of Knudsen and Hilliard⁽⁸⁾ is used to predict inorganic iodine removal. Figure 3 of Reference (8) predicts an inside gas film temperature difference of 1.0 °F. Figure 21 of the present report gives the gas phase mass transfer coefficient, k_d , as 8.1 ft/hr for turbulent boundary conditions and the mass transfer coefficient due to sweep of condensing steam, k_{sw} , as 0.4 ft/hr. Figure 14 of Reference (8) gives the condensing steam flux, n_s , as 0.023 lb/hr ft². The total rate of condensation is $n_s A_w = 2300$ lb/hr. We should emphasize at this time that these are conservatively small estimates, since thermal quasi-equilibrium is assumed. Actually, initial transfer coefficients and condensing steam flux would be much larger (see Figure 20).

The top dome of the vessel is assumed to have a laminar boundary, but a turbulent boundary exists at all other interfaces. The effective value of k_d is then 8.0 ft/hr.

The initial half life for inorganic iodine is

$$t_{1/2} = \frac{0.693 V}{k_d A_T + k_{sw} A_w} = \frac{(0.693)(2 \times 10^6)}{(8.0)(3.5 \times 10^5) + (0.4)(10^5)}$$

$$t_{1/2} = 0.488 \text{ hr} = 29.3 \text{ min}$$

It is necessary to know how long this initial half life will last. Equation (3) can be used to obtain long-term concentration dependence.

$$\frac{C_g}{C_{go}} = \left(1 - \frac{V}{HL_o}\right) e^{-\lambda_1 t} + \left(\frac{V}{HL_o}\right) e^{-\lambda_2 t} \quad (3)$$

We have already calculated the magnitude of λ_1

$$\lambda_1 = \frac{0.693}{t_{1/2}} = \frac{0.693}{0.488} = 1.42 \text{ hr}^{-1}$$

Knudsen and Hilliard⁽⁸⁾ give the following estimate for λ_2 .

$$\lambda_2 = \frac{n_s A_w}{\rho_L L_o} + \frac{k_p A_T}{L_o} = \frac{2300}{(60)(10^4)} + \frac{(3 \times 10^{-4})(3.5 \times 10^5)}{(10^4)}$$

$$\lambda_2 = 0.0038 + 0.0105 = 0.0143 \text{ hr}^{-1}$$

The value of the term $\frac{V}{HL_o}$ is

$$\frac{V}{HL_o} = \frac{2 \times 10^6}{(5000)(10^4)} = 0.040$$

Therefore, from Equation (3)

$$\frac{C_g}{C_{go}} = 0.960 e^{-1.42t} + 0.040 e^{-0.0143T}$$

where t is in hours.

This equation is plotted in Figure 25.

B. METHYL IODIDE

Methyl iodide will be destroyed at a slow rate by chemical reaction in paint and water films. All CSE tests, including nearly 100 small-scale tests in the Aerosol Development Facility, have shown this trend. However, these experiments were conducted in very low radiation fields compared to those expected in a design basis accident in a large PWR.⁽¹⁷⁾ It is possible that some methyl iodide could be produced by a free radical mechanism⁽³⁶⁾ in the presence of high radiation fields. For lack of specific rate information, the assumption is made that depletion and formation rates would be equal, and the concentration of methyl iodide will remain constant in the containment atmosphere.

FIGURE 25. Prediction of Iodine and Particle Mass Concentration in the Containment Atmosphere of a Typical Large PWR

C. PARTICLES

Equation (14) can be used to predict the concentration of particulate mass in the containment atmosphere. The conservative assumption is made that laminar and turbulent diffusion to surfaces is negligible and only gravitational settling and diffusiophoresis would occur.

$$\frac{C_g}{C_{go}} = \exp \left[- \left(\frac{u_t A_{cs}}{V} + \frac{n_s RT A_w}{18 P V} \right) t \right] \quad (14)$$

From Figure 23, the terminal settling velocity, u_t , is 0.34 ft/min for 8 μ diameter particles of unit density.

Therefore

$$\frac{C_g}{C_{go}} = e^{- \left[\frac{(0.34)(1.2 \times 10^4)(60)}{(2 \times 10^6)} + \frac{(0.023)(10.73)(710)(10^5)}{(18)(48.6)(2 \times 10^6)} \right] t}$$

$$\frac{C_g}{C_{go}} = e^{-(0.1224 + 0.0100) t} = e^{-0.132 t}$$

where t is in hours. This equation is plotted in Figure 25.

Iodine is excluded in the calculation of particle behavior, since the CSE experiments have shown that elemental iodine and particulate iodine concentrations decrease at about the same rate. The value of H , the iodine gas-liquid equilibrium coefficient, includes the particulate iodine as part of the total inorganic iodine.

D. DOSE REDUCTION FACTORS

The dose reduction factor (DRF) can be estimated from Figure 25 if the additional assumption is made that the gas leakage rate is invariant with time.

$$DRF_t = \frac{t C_{go}}{\int_0^t C_g dt} \quad (17)$$

The gasborne concentrations for total iodine and for particles were integrated numerically and the resulting DRF's are listed in Table 7.2 for several aging times. The table shows that a significant reduction in total iodine occurs for the initial, important 2-hr period ($DRF_{2HR} = 2.5$), but only a very small reduction occurs for particles ($DRF_{2HR} = 1.14$). These values are believed to be realistic and conservative, based on CSE experiments and the discussions in earlier sections of this report.

TABLE 7.2. Calculated Dose Reduction Factors for a Typical Large PWR^(a)

Time, hr	DRF ^(b)			
	Inorganic Iodine ^(c)	Methyl Iodide	Total Iodine	Particles
1	1.72	1.0	1.71	1.07
2	2.63	1.0	2.50	1.14
4	4.5	1.0	3.92	1.3
8	7.8	1.0	5.83	1.6
12	10.3	1.0	7.0	2.0
24	15.5	1.0	9.0	3.3

a. Refer to Table 7.1 for assumed conditions.

b. Dose reduction factor for constant gas leakage rate, excluding radiodecay.

c. Includes particulate-associated iodine.

At 24 hr after release, DRF's have improved to 9.0 for iodine and 3.3 for particles. At longer times the DRF for iodine would approach 20, due to the assumption of constant methyl iodide concentration at 5% of the initial total iodine release.

VIII. ACKNOWLEDGMENTS

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IX. NOMENCLATURE

a	specific activity of radioactive mass, Ci/lb
A	surface area, ft^2
A_{CS}	horizontal cross-sectional area of vessel, ft^2
A_T	total surface area exposed to fission products, ft^2
A_w	surface area on which steam condenses, ft^2
C	mass concentration, lb/ft^3
C_g	mass concentration in the gas phase, lb/ft^3
C_{go}	mass concentration in gas at zero time, lb/ft^3
C_{avg}	average concentration in vessel, lb/ft^3
C_L	mass concentration in liquid phase, lb/ft^3
d_p	diameter of particle, ft
D	diameter of vessel, ft
D_l	diffusivity in the liquid, ft^2/hr
D_v	diffusivity in the gas, ft^2/hr
f_p	fractional penetration through leak points
g	acceleration due to gravity, ft/hr^2
G	source term, lb/hr
h_w	composite wall heat transfer coefficient, $\text{Btu}/(\text{hr})(\text{ft}^2)(^\circ\text{F})$
H	equilibrium gas-liquid distribution coefficient
k	first-order reaction rate constant, hr^{-1}
k_d	gas phase mass transfer coefficient due only to diffusion, ft/hr
k_{sw}	gas phase mass transfer coefficient due to sweep effect of condensing steam, ft/hr
k_w	effective mass transfer coefficient for particles due turbulent and Brownian diffusion, ft/hr
L	overall height of chamber, ft

L_{avg}	fractional leak rate of contained gas, hr^{-1}
L_f	volumetric flow rate of liquid film, ft^3/hr
n_I	mass flux of iodine to surface, $lb/hr\ ft^2$
n_s	mass flux of steam to surface, $lb/hr\ ft^2$
n_{MI}	mass flux of methyl iodide to surface, $lb/hr\ ft^2$
p_b	partial pressure of steam in bulk gas, psia
p_i	partial pressure of steam at the gas-liquid interface, psia
p_a	partial pressure of air, psia
P	total pressure, psia
Q_i	rate of release of species i from vessel, Ci/hr
R	removal term from gas space, lb/hr , or gas constant, $psia\ ft^3/lb-mole\ ^\circ R$
t	time after beginning of release into containment vessel, hr
$t_{1/2}$	time required to reduce concentration by half, hr
T_a	Temperature of air outside of vessel, $^\circ R$
T_b	temperature of bulk gas inside vessel, $^\circ R$
u	convection velocity with respect to x coordinate, ft/hr
u_t	terminal settling velocity of particle, ft/hr
v	convection velocity with respect to y coordinate, ft/hr
V	gas volume of single chamber, ft^3
V_{wf}	liquid volume held up as wall film, ft^3
w	convection velocity with respect to z coordinate, ft/hr
x	distance down vertical wall, ft

Greek Symbols

λ_1	gas phase mass transfer limiting rate constant, hr^{-1}
λ_2	slow chemical reaction rate constant, hr^{-1}
λ_{diff}	removal rate constant due to diffusion, hr^{-1}
λ_{nc}	removal rate constant due to natural convection, hr^{-1}
δ	effective thickness of liquid wall film, ft
ρ_f	density of fluid through which particle settles, lb mass/ft hr
ρ_L	density of liquid phase, lb/ft^3
ρ_p	effective density of particle, lb/ft^3
μ_f	viscosity of fluid through which particle settles, lb mass/ft hr

X. REFERENCES

1. G. J. Rogers. Program For Containment Systems Experiment, HW-83607. Available from Clearinghouse for Federal Scientific and Technical Information, Springfield, Virginia, September 1964.
2. Staff of Battelle-Northwest. Nuclear Safety Quarterly Report for July-October, 1967, BNWL-754, pp. 2.1-2.30. Battelle-Northwest, Richland, Washington, June 1968.
3. Staff of Battelle-Northwest. Nuclear Safety Quarterly Report for August-October, 1968, BNWL-926, pp. 2.1-2.18. Battelle-Northwest, Richland, Washington, December 1968.
4. Staff of Battelle-Northwest. Nuclear Safety Quarterly Report for May-July, 1969, BNWL-1187, pp. 2.6-2.16. Battelle-Northwest, Richland, Washington, September 1969.
5. R. K. Hilliard et al. Removal of Iodine and Particles from Containment Atmospheres by Sprays - Containment Systems Experiment Interim Report, BNWL-1244. Battelle-Northwest, Richland, Washington, February 1970.
6. A. K. Postma and L. F. Coleman. Unpublished Data. Battelle-Northwest, Richland, Washington. (Report in Preparation: Effect of Continuous Spray Operation on the Removal of Aerosols and Gases in the Containment Systems Experiment)
7. A. K. Postma. Unpublished Data. Battelle-Northwest, Richland, Washington. (Report in Preparation: Effect of Flow Rate on Spray Removal of Aerosols and Gases in the Containment Systems Experiment)
8. J. G. Knudsen and R. K. Hilliard. Fission Product Transport by Natural Processes in Containment Vessels, BNWL-943. Battelle-Northwest, Richland, Washington, January 1969.
9. W. J. Megaw and F. G. May. The Behavior of Iodine Released in Reactor Containers, AERE-R-3781. Atomic Energy Research Establishment, Harwell, England, July 1961.

10. D. H. F. Atkins and A. E. J. Eggleton. Iodine Compounds Formed on Release of Carrier-Free Iodine-131, AERE-M-1211. Atomic Energy Research Establishment, Harwell, England, May 1963.
11. A. C. Chamberlain. Report of Aerosol Group, Part I. Physical Chemistry of Iodine and Removal of Iodine From Gas Streams, AERE-R-4286. Atomic Energy Research Establishment, Harwell, England, April 1963.
12. B. F. Roberts, S. H. Freid, G. W. Parker, L. F. Parsly and T. H. Row. Evaluation of Various Methods of Fission Product Aerosol Simulation, ORNL-TM-2628. Oak Ridge National Laboratory, Oak Ridge, Tennessee, September 1969.
13. W. A. Freeby et al. Fission Product Behavior Under Simulated Loss-of-Coolant Accident Conditions in the Contamination-Decontamination Experiment, IN-1171. Idaho Nuclear Co., Inc., Idaho Falls, Idaho, January 1969.
14. R. K. Hilliard, L. F. Coleman and J. D. McCormack. Comparisons of the Containment Behavior of a Simulant with Fission Products Released from Irradiated UO₂, BNWL-581. Battelle-Northwest, Richland, Washington, March 1968.
15. R. L. Ritzman et al. "Analytical Description of Fission-Product Transport and Deposition in Containment Vessels," Trans. Am. Nucl. Soc., vol. 10, no. 2, p. 714. 1967.
16. 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. 136-200. Battelle Memorial Institute, Columbus, Ohio. August 1966.
17. H. S. Rosenberg et al. Fission Product Deposition and Its Enhancement Under Reactor Accident Conditions: Deposition on Containment-System Surfaces, BMI-1865. Battelle Memorial Institute, Columbus, Ohio, May 1969.

18. M. N. Ozisik and D. Hughes. "Effects of Condensation on the Transport of Matter from Vapor and Noncondensable Gas Mixtures," Nucl. Sci. Eng., vol. 35, pp. 384-393. 1969.
19. T. W. Horst. A Review of Particle Transport in a Condensing Steam Environment, BNWL-848. Battelle-Northwest, Richland, Washington, June 1968.
20. J. M. Hales. Aerosol Transport in a Condensing Steam Environment--Status Report, BNWL-SA-2359. Battelle-Northwest, Richland, Washington, February 1969.
21. G. M. Watson, R. B. Perez and M. H. Fontana. Effects of Containment System Size on Fission Product Behavior, ORNL-4033. Oak Ridge National Laboratory, Oak Ridge, Tennessee, January 1967.
22. W. A. Yuill, Jr. and V. F. Baston. "A Model for Fission-Product Deposition Under Natural-Response Conditions in Containment Buildings," Nuclear Safety, vol. 10, no. 6, pp. 492-498. 1969.
23. H. Schlichting. Boundary Layer Theory. McGraw-Hill Book Co., Inc., New York, 1962. p. 334.
24. M. Jakob. Heat Transfer. John Wiley and Sons, New York, 1949. vol I, p. 592.
25. C. E. Linderorth. Containment Systems Experiment, Part I, Description of Experimental Facilities, BNWL-456. Battelle-Northwest Laboratories, Richland, Washington, March 1970.
26. L. F. Coleman. Preparation, Generation, and Analysis of Gases and Aerosols for the Containment Systems Experiment, BNWL-1001. Battelle-Northwest, Richland, Washington, April 1969.
27. J. D. McCormack. Maypack Behavior in the Containment Systems Experiment--A Penetrating Analysis, BNWL-1145. Battelle-Northwest, Richland, Washington, August 1969.

28. Staff of Battelle-Northwest. Nuclear Safety Quarterly Report for November-December, 1968, January 1969, BNWL-1009, pp. 2.23-2.27. Battelle-Northwest, Richland, Washington, March 1969.
29. W. B. Cottrell. Nuclear Safety Program Annual Progress Report for Period Ending December 31, 1968, ORNL-4374, p. 28. Oak Ridge National Laboratory, Oak Ridge Tennessee, June 1969.
30. J. G. Knudson and D. L. Katz. Fluid Dynamics and Heat Transfer. McGraw-Hill Book Co., Inc., New York, 1958. Chapter 11.
31. J. G. Knudsen. "Properties of Air-Steam Mixtures Containing Small Amounts of Iodine," BNWL-1326. Battelle-Northwest, Richland, Washington. April 1970
32. A. E. J. Eggleton. A Theoretical Examination of Iodine-Water Partition Coefficients, AERE-R-4887. Atomic Energy Research Establishment, Harwell, England, February 1967.
33. H. S. Rosenberg et al. Fission Product Deposition and Its Enhancement Under Reactor Accident Conditions - Development of Reactive Coatings, BMI-1874, p. 15. Battelle Memorial Institute, Columbus, Ohio, November, 1969.
34. A. K. Postma. Absorption of Methyl Iodide by Aqueous Hydrazine Solutions Within Spray Chambers. Oregon State University, Corvallis, Oregon, 1970. (Ph. D. thesis)
35. L. C. Schwendiman et al. The Washout of Methyl Iodide by Hydrazine Sprays. Final Report, BNWL-935. Battelle-Northwest, Richland, Washington, November 1968.
36. L. L. Burger. Unpublished Data. Battelle-Northwest Laboratory, Richland, Washington, 1967. (Personal Communication)

APPENDIX A

TABLES OF MAYPACK DATA

The mean concentrations of the various airborne materials in the main room gas space are tabulated in Tables A-1 through A-6 for each of the six experiments. The standard deviation from the mean is also listed as percent. In calculating the mean value, judgment was used to eliminate anomalies caused by occasional equipment malfunction or operator error. Less than 5% of the data was discarded, and this only after careful checking.

The values tabulated are those obtained after correcting for "blank"--the value obtained by the twelfth Maypack in each cluster which was never used to draw a sample, but nevertheless picked up traces of iodine and cesium. Corrections were also applied to convert iodine on silver and filters to elemental and particulate concentrations.⁽²⁷⁾

TABLE A-1. Concentration in the Main Vapor Space--Run D-1 (a)

Time, min	Mean Concentration, $\mu\text{g}/\text{m}^3 \pm 1 \sigma$ (%) (b)										
	15	30	40	50	70	90	150	300	600	1100	1340
Elemental Iodine	94.4 (12)	33.5 (11)	10.9 (29)	4.15 (17)	0.964 (8)	0.53 (9)	0.42 (24)	0.22 (20)	0.12 (12)	0.065 (8)	0.041 (21)
Particulate Iodine	11.6 (20)	2.43 (19)	0.96 (15)	0.36 (18)	0.056 (20)	0.010 (32)	0.011 (52)	0.0086 (65)	0.011 (59)	0.0080 (98)	0.0154 (33)
Iodine Associated with Charcoal Paper	4.81 (35)	2.31 (37)	0.775 (38)	0.318 (59)	0.161 (22)	0.104 (13)	0.0856 (15)	0.0737 (13)	0.047 (15)	0.033 (22)	0.027 (23)
Iodine on Maypack Inlet	12.3 (57)	4.4 (42)	1.21 (54)	0.35 (72)	0.090 (84)	(c)	0.027 (140)	0.012 (175)	0.016 (102)	0.002 (620)	0.014 (143)
Total Inorganic Iodine	123 (7)	42.6 (10)	13.9 (28)	5.17 (18)	1.28 (10)	0.647 (11)	0.543 (11)	0.311 (13)	0.197 (10)	0.108 (8)	0.097 (26)
Methyl Iodide	8.11 (31)	2.81 (28)	1.13 (22)	0.63 (13)	0.443 (14)	0.453 (17)	0.405 (11)	0.349 (13)	0.279 (11)	0.177 (7)	0.119 (8)
Total Iodine (All Forms)	131 (5)	45.4 (9)	15.0 (28)	5.8 (18)	1.72 (9)	1.10 (10)	0.948 (11)	0.660 (12)	0.476 (8)	0.285 (7)	0.216 (26)
Cesium	577 (18)	183 (16)	59.6 (25)	22.9 (26)	5.70 (28)	1.49 (11)	0.916 (75)	0.394 (88)	0.375 (46)	0.048 (235)	0.348 (107)

a. Average of 7 sampling locations.

b. Numbers in parentheses are standard deviation from the mean, percent.

c. Not detectable.

TABLE A-2. Concentration in the Main Vapor Space--Run D-2 (a)

Time, Min	Mean Concentration, $\mu\text{g}/\text{m}^3 \pm 1 \sigma$ (%) (b)									
	5	11	21	31	46	61	81	121	181	301
Elemental Iodine	90.2 (11)	84.2 (18)	58.0 (22)	25.8 (16)	11.1 (25)	3.50 (14)	1.24 (19)	0.296 (34)	0.214 (21)	0.192 (25)
Particulate Iodine	139 (47)	108 (45)	39.6 (26)	22.8 (15)	6.33 (27)	1.45 (22)	(c)	(c)	(c)	(c)
Iodine Associated with Charcoal Paper	3.05 (61)	1.90 (33)	1.75 (47)	0.865 (42)	0.404 (18)	0.260 (38)	0.143 (10)	0.111 (14)	0.083 (15)	0.069 (51)
Iodine on Maypack Inlet	15.8 (87)	11.6 (61)	20.7 (57)	5.21 (39)	1.34 (90)	0.340 (33)	0.11 (47)	0.0366 (13)	0.040 (53)	0.0274 (38)
Total Inorganic Iodine	248.0 (31)	205.7 (23)	120.0 (15)	54.7 (7)	19.2 (11)	5.55 (7)	1.49 (11)	0.444 (17)	0.337 (12)	0.288 (25)
Methyl Iodide	0.176 (39)	0.221 (23)	0.226 (34)	0.174 (12)	0.175 (20)	0.154 (20)	0.152 (20)	0.135 (16)	0.109 (17)	0.182 (14)
Total Iodine (All Forms)	248.2 (31)	205.9 (23)	120.2 (15)	54.9 (7)	19.3 (10)	5.70 (6.6)	1.65 (10)	0.579 (16)	0.446 (12)	0.470 (14)
Cesium	767 (43)	1135 (32)	733 (13)	354 (6.3)	156 (10)	63.6 (4.4)	24.9 (6.1)	5.66 (15)	1.15 (25)	0.581 (40)
Uranium (d)	660	2060	2140	1250	123	36.0	19.2	3.8	(c)	(c)
Ruthenium	5.7 (40)	14.2 (45)	9.4 (11)	5.5 (11)	2.75 (26)	1.4 (11)	0.60 (5)	0.115 (9)	0.0095 (48)	(c)

a. Average of 7 sampling locations.

b. Numbers in parentheses are standard deviation from the mean, percent.

c. Not detectable.

d. One location.

TABLE A-3. Concentration in the Main Vapor Space--Run A-1 (a)

Time, min	Mean Concentration, $\mu\text{g}/\text{m}^3 \pm 1 \sigma$ (%) (b)									
	5	11	26	41	61	81	100	120	180	300
Elemental Iodine	43.9 (24)	34.3 (24)	20.0 (19)	13.4 (12)	9.84 (19)	6.72 (21)	5.62 (19)	4.68 (24)	2.58 (19)	1.07 (16)
Particulate Iodine	189 (20)	112 (11)	43.4 (15)	24.3 (11)	10.9 (18)	6.31 (17)	3.39 (17)	2.02 (24)	(c)	(c)
Iodine Associated with Charcoal Paper	7.91 (17)	8.45 (7.8)	7.42 (10)	6.28 (7)	5.48 (8)	4.41 (15)	4.02 (15)	3.53 (15)	2.65 (16)	1.85 (17)
Iodine on Maypack Inlet	67.5 (34)	46.4 (26)	9.26 (29)	3.50 (37)	0.94 (47)	0.475 (97)	0.13 (65)	0.085 (53)	0.012 (343)	0.015 (323)
Total Inorganic Iodine	308.3 (12)	201.1 (8)	80.1 (12)	47.5 (8)	27.2 (12)	17.9 (14)	13.2 (15)	10.3 (15)	5.24 (20)	2.93 (15)
Methyl Iodide	0.529 (34)	0.574 (31)	0.513 (34)	0.473 (27)	0.480 (26)	0.457 (29)	0.473 (27)	0.441 (24)	0.423 (26)	0.400 (40)
Total Iodine (All Forms)	308.8 (12)	201.7 (8.7)	80.6 (12)	48.0 (8)	27.7 (12)	18.4 (14)	13.6 (14)	10.8 (14)	5.66 (21)	3.33 (15)
Cesium	268 (19)	218 (14)	109 (10)	72.5 (9.7)	48.8 (8)	34.3 (8.3)	27.2 (7.4)	21.4 (10)	10.3 (17)	3.09 (13)
Uranium	432 (30)	364 (31)	250 (20)	178 (41)	143 (40)	125 (42)	86 (71)	85 (13)	71 (86)	13.5 (56)

a. Average of 13 sampling locations in the main room, including drywell.

b. Numbers in parentheses are standard deviation from the mean, percent.

c. Not detectable.

TABLE A-4. Concentration in the Main Vapor Space--Run A-2 (a)

Time, min	Mean Concentration, $\mu\text{g}/\text{m}^3 \pm 1 \sigma$ (%) (b)									
	5	12	25	48	74	114	151	210	300	720
Elemental Iodine	18,020 (72)	51,910 (10)	23,580 (13)	4,476 (13)	558 (41)	156 (10)	129 (9)	83.4 (8.5)	54.5 (7)	25.8 (50)
Particulate Iodine	1,960 (35)	2,375 (22)	1,672 (20)	789 (22)	363 (18)	22.9 (16)	(d)	(d)	(d)	(d)
Iodine Associated with Charcoal Paper	263 (44)	340 (8)	410 (39)	277 (30)	184 (15)	102 (11)	88.6 (10)	70.2 (9)	54.0 (9)	31.0 (23)
Iodine on Maypack Inlet	1,460 (33)	2,029 (27)	1,553 (54)	455 (34)	110 (22)	4.87 (112)	(d)	(d)	(d)	(d)
Total Inorganic Iodine	21,700 (30)	56,650 (9)	27,214 (12)	5,997 (11)	1,215 (12)	286 (6)	218 (7)	153 (10)	109 (10)	56 (15)
Methyl Iodide	97.6 (21)	140 (13)	136 (15)	145 (16)	124 (26)	115 (14)	134 (15)	121 (14)	118 (13)	110 (15)
Total Iodine (All Forms)	21,798 (31)	56,790 (8.8)	27,350 (12)	6,142 (11)	1,339 (11)	401 (5.1)	352 (7)	274 (9)	227 (10)	167 (14)
Cesium	2,258 (35)	3,144 (14)	2,098 (14)	720 (18)	267 (13)	76.2 (8.7)	38.0 (6)	12.7 (18)	3.25 (32)	(d)
Uranium ^(c)	1,380	870	500	460	137	79	44	9	2	(d)

a. Average of 9 sampling locations in the main room, including drywell.

b. Numbers in parentheses are standard deviation from the mean, percent.

c. One location.

d. Not detectable.

TABLE A-5. Concentration in the Main Vapor Space--Run A-5 (a)

Time, min	Mean Concentration, $\mu\text{g}/\text{m}^3 \pm 1 \sigma$ (%) (b)										
	13	25	45	71	111	151	211	301	531	1000	1426
Elemental Iodine	60,400 (21)	35,400 (28)	12,290 (29)	2,680 (40)	445 (14)	246 (13)	141 (17)	93.7 (18)	67.2 (22)	54.3 (16)	43.0 (19)
Particulate Iodine	8,900 (26)	7,600 (21)	5,400 (23)	2,530 (18)	340 (26)	62 (43)	22.9 (76)	32.9 (92)	17.4 (157)	5.1 (200)	9.1 (185)
Iodine Associated with Charcoal Paper	1,970 (24)	1,860 (19)	1,360 (19)	670 (16)	204 (14)	143 (14)	127 (18)	101 (17)	81 (21)	60 (12)	51 (25)
Iodine on Maypack Inlet	111 (40)	120 (35)	122 (54)	53 (32)	34 (74)	14.2 (61)	10.3 (59)	9.1 (77)	9.9 (66)	13 (78)	4.7 (110)
Total Inorganic Iodine	72,380 (18)	45,000 (22)	19,100 (19)	5,945 (16)	1,021 (13)	465 (15)	261 (18)	237 (18)	176 (20)	132 (17)	108 (20)
Methyl Iodide	1,320 (17)	1,320 (11)	1,204 (12)	1,055 (9)	869 (13)	855 (9)	785 (12)	697 (10)	630 (12)	527 (10)	438 (8.4)
Total Iodine (All Forms)	73,800 (18)	46,300 (22)	20,300 (19)	6,990 (16)	1,890 (8)	1,320 (7)	1,046 (9.5)	903 (8)	806 (11)	659 (9)	546 (8)
Cesium	630 (20)	506 (13)	410 (29)	238 (39)	125 (15)	83 (8)	52 (7)	25.1 (18)	6.70 (13)	0.362 (88)	0.257 (140)
Uranium	510 (35)	390 (33)	350 (12)	226 (21)	132 (27)	102 (19)	53 (23)	38 (12)	7.3 (36)	(c)	1.1 (173)

a. Average of 12 sampling locations in the main room, including drywell.

b. Numbers in parentheses are standard deviation from the mean, percent.

c. Not detectable.

TABLE A-6. Concentration in the Main Vapor Space--Run A-11 (a)

Time, min	Mean Concentration, $\mu\text{g}/\text{m}^3 \pm 1 \sigma$ (%) (a)									
	22	52	60	90	120	181	300	480	960	2770
Elemental Iodine	43,700 (12)	12,000 (20)	9,020 (3.2)	2,450 (46)	647 (35)	202 (20)	154 (15)	79.1 (12)	32.8 (33)	28.4 (50)
Particulate Iodine	5,760 (11)	4,650 (18)	3,520 (13)	2,930 (13)	1,600 (24)	985 (47)	376 (23)	194 (30)	24.2 (55)	7.15 (84)
Iodine Associated with Charcoal Paper	1,160 (12)	1,170 (15)	962 (19)	590 (16)	405 (19)	310 (12)	246 (7)	191 (14)	113 (13)	100 (22)
Iodine on Maypack Inlet	203 (27)	284 (89)	340 (43)	175 (45)	120 (32)	335 (45)	162 (76)	162 (168)	(c)	(c)
Total Inorganic Iodine	50,820 (10)	18,100 (13)	13,840 (20)	6,140 (16)	2,770 (12)	1,832 (30)	938 (8)	626 (10)	170 (20)	135 (25)
Methyl Iodide	1,410 (18)	1,280 (8.5)	1,290 (7)	1,210 (7)	1,090 (6.3)	1,110 (10)	1,160 (4)	1,090 (8)	771 (8)	696 (11)
Total Iodine (All Forms)	52,200 (10)	19,300 (13)	15,100 (19)	7,340 (16)	3,870 (12)	2,840 (28)	2,098 (5.6)	1,716 (7)	941 (6)	831 (10)
Cesium	1,540 (8)	1,010 (10)	935 (12)	807 (15)	640 (18)	606 (27)	455 (13)	372 (3.4)	177 (6)	131 (10)
Uranium	1,650 (11)	678 (19)	553 (22)	462 (13)	368 (29)	383 (35)	284 (28)	247 (18)	87 (30)	55 (44)

a. Average of 12 sampling locations in the main room, including drywell.

b. Numbers in parentheses are standard deviation from the mean, percent.

c. Not detectable.

APPENDIX B

FIGURES OF CONCENTRATION VERSUS TIME

The concentration of the several forms of iodine and of cesium and uranium in the gas and liquid phases are plotted as functions of time in the following figures. Individual measurements or averages of several samples taken at the same time are shown as points. The curves are the best fit of the experimental measurements. Where exponential rates are observed, the slopes are measured in terms of "half life," or time to reduce the concentration by a factor of two.

FIGURE B-1.
Concentration of Elemental
Iodine in Gas Space, Run D-1

FIGURE B-2.
Concentration of Elemental
Iodine in Gas Space, Run D-2

FIGURE B-3.
Concentration of Elemental
Iodine in Gas Space, Run A-1

FIGURE B-4.
Concentration of Elemental
Iodine in Gas Space, Run A-2

FIGURE B-5.
Concentration of Elemental
Iodine in Gas Space, Run A-5

FIGURE B-6.
Concentration of Elemental
Iodine in Gas Space, Run A-11

FIGURE B-7.
Concentration of Particulate
Iodine in Gas Space, Run D-1

FIGURE B-8.
Concentration of Particulate
Iodine in Gas Space, Run D-2

FIGURE B-9.
Concentration of Particulate
Iodine in Gas Space, Run A-1

FIGURE B-10.
Concentration of Particulate
Iodine in Gas Space, Run A-2

FIGURE B-11.
Concentration of Particulate
Iodine in Gas Space, Run A-5

FIGURE B-12.
Concentration of Particulate
Iodine in Gas Space, Run A-11

FIGURE B-13.
Concentration of Methyl
Iodide in Gas Space, Run D-1

FIGURE B-14.
Concentration of Methyl
Iodide in Gas Space, Run D-2

FIGURE B-15.
Concentration of Methyl
Iodide in Gas Space, Run A-1

FIGURE B-16.
Concentration of Methyl
Iodide in Gas Space, Run A-2

FIGURE B-17.
Concentration of Methyl
Iodide in Gas Space, Run A-5

FIGURE B-18.
Concentration of Methyl
Iodide in Gas Space, Run A-11

FIGURE B-19.
Concentration in Gas Space
of Iodine Associated
with Charcoal Paper, Run D-1

FIGURE B-20.
Concentration in Gas Space
of Iodine Associated
with Charcoal Paper, Run D-2

FIGURE B-21.
Concentration in Gas Space
of Iodine Associated
with Charcoal Paper, Run A-1

FIGURE B-22.
Concentration in Gas Space
of Iodine Associated
with Charcoal Paper, Run A-2

FIGURE B-23.
Concentration in Gas Space
of Iodine Associated
with Charcoal Paper, Run A-5

FIGURE B-24.
Concentration in Gas Space
of Iodine Associated
with Charcoal Paper, Run A-11

FIGURE B-25.

Concentration of Total Inorganic Iodine in Gas Space, Run D-1

FIGURE B-26.

Concentration of Total Inorganic Iodine in Gas Space, Run D-2

FIGURE B-27.

Concentration of Total Inorganic Iodine in Gas Space, Run A-1

FIGURE B-28.

Concentration of Total Inorganic Iodine in Gas Space, Run A-2

FIGURE B-29. Concentration of Total Inorganic Iodine in Gas Space, Run A-5

FIGURE B-30.

Concentration of Total Inorganic Iodine in Gas Space, Run A-11

FIGURE B-31. Concentration of Total Iodine in Gas Space, Run D-1

FIGURE B-32. Concentration of Total Iodine in Gas Space, Run D-2

FIGURE B-33.
Concentration of Total
Iodine in Gas Space, Run A-1

FIGURE B-34.
Concentration of Total
Iodine in Gas Space, Run A-2

FIGURE B-35.
Concentration of Total
Iodine in Gas Space, Run A-5

FIGURE B-36.
Concentration of Total
Iodine in Gas Space, Run A-11

FIGURE B-37.
Concentration of Cesium
in the Gas Space, Run D-1

FIGURE B-38.
Concentration of Cesium
in the Gas Space, Run D-2

FIGURE B-39.
Concentration of Cesium
in the Gas Space, Run A-1

FIGURE B-40.
Concentration of Cesium
in the Gas Space, Run A-2

FIGURE B-41.
Concentration of Cesium
in the Gas Space, Run A-5

FIGURE B-42.
Concentration of Cesium
in the Gas Space, Run A-11

FIGURE B-43.
Concentration of Uranium
in the Gas Space, Run D-2

FIGURE B-44.
Concentration of Uranium
in the Gas Space, Run A-1

FIGURE B-45.
Concentration of Uranium
in the Gas Space, Run A-2

FIGURE B-46.
Concentration of Uranium
in the Gas Space, Run A-5

FIGURE B-47.
Concentration of Uranium
in the Gas Space, Run A-11

FIGURE B-48.
Concentration of Ruthenium
in the Gas Space, Run D-2

FIGURE B-49. Concentration of Iodine and Cesium in Vessel Sump, Run D-1

FIGURE B-50. Concentration of Iodine and Cesium in Vessel Sump, Run D-2

FIGURE B-51. Concentration of Iodine and Cesium in Vessel Sump, Run A-1

FIGURE B-52. Concentration of Iodine and Cesium in Vessel Sumps, Run A-2

FIGURE B-53. Concentration of Iodine and Cesium in Vessel Sumps, Run A-5

FIGURE B-54. Concentration of Iodine and Cesium in Vessel Sumps, Run A-11

FIGURE B-55. Iodine Concentration in Wall Condensate as a Function of Time, Runs A-1 and A-2

FIGURE B-56. Iodine Concentration in Wall Condensate Film, Run A-5

FIGURE B-57. Iodine Concentration in Wall Condensate Film, Run A-11

FIGURE B-58. Cesium Concentration in Wall
Condensate Film, Run A-1

FIGURE B-59. Cesium Concentration in Wall
Condensate Film, Run A-2

FIGURE B-60. Cesium Concentration in Wall Condensate Film, Run A-5

FIGURE B-61. Cesium Concentration in Wall Condensate Film, Run A-11

APPENDIX C

ATMOSPHERIC PROPERTIES

A knowledge of the fundamental properties of the gases and particles comprising the containment atmosphere is needed for mathematical modeling. Knudsen⁽³¹⁾ has tabulated values for many of the properties over the range of temperature and pressure pertinent to containment post-accident conditions. An excerpt of some of his data is presented in Table C-1. The pressures associated with each temperature in the table are those which would result if steam were added to a closed vessel initially at 80 °F, 14.7 psia containing air at 100% RH. Saturated conditions and uniform concentrations are assumed for all cases.

TABLE C-1. Atmospheric Properties for Various Temperatures in the Containment Atmosphere

Property	Value in the Bulk Gas Mixture					
	80	140	180	220	250	280
T_b , vapor temp., °F	14.70	18.717	24.393	35.124	48.555	68.640
P , total press., psia	0.507	2.889	7.510	17.186	29.825	49.204
P_s , partial press. steam, psia	15.183	15.828	16.883	17.938	18.730	19.436
P_a , partial press. air, psia	0.0345	0.154	0.308	0.489	0.616	0.717
X_s , mol fraction steam	0.9655	0.846	0.692	0.511	0.384	0.283
X_a , mol fraction air	28.63	27.31	25.61	23.62	22.23	21.11
MW, molec. wt of mixture	0.0729	0.0786	0.0910	0.1145	0.1434	0.1826
ρ , density of mix., lb/ft ³	0.0437	0.0449	0.0439	0.0423	0.0412	0.0407
ν , absolute viscosity, lb/ft hr	0.599	0.571	0.482	0.369	0.287	0.223
ν , kinematic viscosity, ft ² /hr	0.0150	0.0158	0.0172	0.0175	0.0177	0.0167
k , thermal conduct., Btu/hr ft °F	0.2498	0.2649	0.2869	0.3185	0.3461	0.3678
C_p , heat capacity, Btu/lb °F	0.728	0.754	0.735	0.767	0.802	0.896
Pr, Prandtl No.	1.004	0.971	0.734	0.589	0.463	0.354
D_s , molec. diffusivity steam, ft ² /hr	0.346	0.326	0.278	0.211	0.165	0.126
D_{I_2} , molec. diffusivity of I ₂ , ft ² /hr	0.439	0.415	0.353	0.271	0.212	0.162
D_{MI} , molec. diffusivity of methyl iodide, ft ² /hr	0.598	0.572	0.572	0.566	0.564	0.564
Sc_s , Schmidt No., steam	1.743	1.735	1.741	1.750	1.756	1.736
Sc_I , Schmidt No. I ₂	1.370	1.361	1.361	1.361	1.357	1.348
Sc_{MI} , Schmidt No., Methyl Iodide						

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