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AEC Research and Development Report

LEAKAGE OF FISSION PRODUCTS
FROM ARTIFICIAL LEAKS
IN THE CONTAINMENT SYSTEMS
EXPERIMENT

M.E. WITHERSPOON and A.K. POSTMA

June 1971

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LEAKAGE OF FISSION PRODUCTS FROM ARTIFICIAL
LEAKS IN THE CONTAINMENT SYSTEMS EXPERIMENT

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ABSTRACT

Decontamination of fission products in containment vessel leak paths was measured as part of the Containment Systems Experiment (CSE) program. Most of the leak paths were artificial. Use of artificial leak paths was required because natural leaks identified during air pressure tests plugged when steam was fed to the containment vessel. Both iodine and cesium were retained to an appreciable extent within the leak path. Collection of leaked iodine for 120 minutes following its release to the containment vessel atmosphere gave decontamination factors of 3.1 and 4.0 for leakage through stainless steel needle valves. For a natural leak past a butterfly valve, a decontamination factor of 34 was observed for a collection period of 24 hours. Less than 5% of iodine contained in leaked liquid became airborne when evaporated to dryness.

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LEAKAGE OF FISSION PRODUCTS FROM ARTIFICIAL
LEAKS IN THE CONTAINMENT SYSTEMS EXPERIMENT

M. E. Witherspoon and A. K. Postma

INTRODUCTION

This report describes the results of experiments on the leakage of airborne fission products from a nuclear reactor containment vessel. The work was done as part of the Containment Systems Experiment program, a research effort supported by the USAEC Division of Reactor Development and Technology.

A reactor containment vessel is the primary defense against the leakage of radioactive materials from a nuclear reactor in the unlikely event of fission product escape from the primary reactor vessel. Containment vessels are robust structures, assembled under carefully examined conditions. Leakage rates from assembled containment vessels are very low. Current practice is to measure leakage at a specified pressure to verify that it is below the design value, typically 0.1 volume percent per day. In predicting the radiation doses in the atmosphere which would accompany a hypothetical accident, the source term is calculated as the product of the leakage rate and the airborne concentration inside the containment vessel. This procedure is based on the assumption that airborne fission products pass through the leak paths without depletion or enrichment. This would obviously be true for noble gases, but may not hold for reactive gases such as elemental iodine or for aerosol particles.

The purpose of this work was to provide experimental information on the depletion or enrichment of fission products in leak paths on a large containment vessel. It was recognized at the outset that one could not expect to characterize fission product holdup in all possible leaks because an infinity of leak path situations can be postulated. Rather, several well characterized leaks were studied. The quantity of information obtained here is modest, but provides insight on the degree of conservatism associated with current calculational procedures which neglect the processes which occur in the leak path.

SUMMARY AND CONCLUSIONS

Fission product decontamination was measured for several artificial leak paths installed on the CSE containment vessel. Artificial leaks were used because the natural leaks identified during air pressure tests plugged when steam was fed to the vessel. Individual leak flow rates were varied from 0.1 to 1.0 liters per minute.

Leak path geometries included the following.

- inlet flush with containment vessel, needle valve flow restriction
- inlet protruding into vessel atmosphere, needle valve flow restriction
- inlet protruding into vessel atmosphere, orifice flow restriction
- leak from space between two 8" butterfly valves

The air leaking from these paths was passed through a train to characterize the airborne contaminants, and then through a flow meter. The mass of fission products escaping was compared to the mass calculated from the time integral of the product of the measured flow rate and the measured airborne concentration inside the vessel.

The following conclusions are supported by this work:

- Natural leak paths identified during air pressure tests on the CSE vessel plugged when steam was added to the vessel. Similar behavior is to be expected for PWR containment vessels under accident conditions.
- Appreciable decontamination of airborne iodine and particles occurs in leak paths. For example, in CSE Run A-16, dose reduction factors of 3.1 and 4.0 were observed for iodine passing through a stainless steel leak path. A 24-hour dose reduction factor of 34 for iodine was measured for a natural leak from an 8" butterfly valve. These decontamination factors are in addition to those caused by cleanup of the containment vessel atmosphere. Hence they represent factors of conservatism if fission product deposition within leak paths is ignored.
- Iodine carried from a leak path in water solution will become airborne to only a slight extent. Experiments showed that less than 4% of dissolved iodine was evolved when liquid was evaporated to dryness.

- For fission products in both the gaseous and liquid phase, the leak path acts as a chromatographic chamber. At early times appreciable net holdup occurs by physical retention of liquid and by surface sorption. At longer times the leaked effluent is enriched as retained liquid is released and as sorbed material is desorbed. For long times, the decontamination factor for the sum of airborne and liquid-borne effluent approaches unity.
- The results of this work are not in agreement with an earlier thesis by Philbin⁽¹⁾. Philbin⁽¹⁾ concluded that the leak rate under accident conditions would be larger than predicted from air tests, that iodine does not deposit in leak paths, and that condensing steam transports iodine through containment vessel leaks.

BACKGROUND

Leak testing is required for all operational containment vessels. Most of the effort to date has been devoted to development of test procedures, and application of test results to accident conditions. Very little effort has been directed to evaluating the disposition of fission products once they enter a leak path.

Maccary, et al⁽²⁾ performed detailed analyses of leakage of gases from containment vessels. These authors discuss the available techniques for measuring leak rates and develop formulae for estimating the leak rates under accident conditions from measurements made at ambient temperature.

Zapp⁽³⁾ has reviewed existing information on containment vessel testing associated with water-cooled reactors. Conclusions were presented relative to leakage rate tests results, continuous leakage-rate monitoring, and developments that may affect future testing requirements.

Browning and Ackley⁽⁴⁾ calculated the diffusional deposition of aerosol particles in a leak path for laminar flow in cylindrical and rectangular flow passages. They did not account for the presence of liquid in the flow path. Their calculations showed that the fractional deposition of particles within the leak path depended markedly on the width of the leak path and the particle size. For a rectangular leak path one inch long and having a width of 20

microns, the decontamination factor predicted for 0.05 μ particles was 1.5. For 0.01 μ particles, the predicted decontamination factor was 5000.

Philbin⁽¹⁾ measured the leakage of Kr-85 and I-131 from an aluminum chamber through artificial leak paths. Iodine was collected in a charcoal bed attached to the leak outlet. No discrimination was made between iodine airborne and that dissolved in water. Philbin⁽¹⁾ concluded that the leak rate under accident conditions would be larger than predicted from air tests, that iodine does not deposit in leak paths, and that condensing steam transports iodine through containment vessel leaks. Unfortunately, Philbin⁽¹⁾ did not measure the iodine concentration inside the chamber. Direct comparison of the iodine concentration in the leaked gas with that airborne inside the chamber is thus not possible.

Nelson, et al⁽⁵⁾ measured the penetration of sodium oxide aerosol through concrete specimens which contained cracks. These experiments were designed to simulate leakage through the concrete layer on a containment vessel. Their results showed that leaked gas was decontaminated by factors greater than 150 as it passed through the leak.

For postulated serious accidents, people located downwind will receive radiation doses from materials leaked from the containment vessel. The magnitude of the downwind dose varies linearly with the release rate. This release rate is the product of a gas leak rate, and the gas concentration.

$$Q = LC_0 \quad (1)$$

where Q = radioactive material release rate, curies/time,

L = gaseous leak rate, volume/time,

C_0 = concentration in leaked gas, curies/volume.

The leak rate, L , for a reactor containment vessel, is never measured under accident conditions. The currently used procedure is to measure the leak rate in a pressure test at ambient temperature and extrapolate the measured leak rate to accident conditions on the basis of flow models which account for changes in temperature, pressure, and composition. Maccary, et al⁽²⁾ have outlined the mathematical equations which can be used to do this. Leak paths cannot be expected to be simply characterized. Hence the recommended procedure has been to extrapolate to higher pressures using

laminar flow theory. This is conservative in that laminar flow theory predicts the largest possible leak rate at higher pressure, if geometry remains constant. This is expressed as

$$L_a = L_T F_\lambda \quad (2)$$

where L_a = leak rate predicted for accident conditions,

L_T = leak rate measured at ambient temperature,

F_λ = laminar flow extrapolation factor based on pressure and temperature differences.

If the leak rate is only specified at the highest pressure accompanying an accident, Maccary, et al⁽²⁾ recommend downward extrapolation on the basis of turbulent flow. This again gives a conservative estimate.

These calculations are based on the assumption of constant leak path geometry. This is necessary because one can't predict the changes in geometry which might occur. Witherspoon^(6,7) has carried out experiments on leaks in the CSE containment vessel to assess the change in leak rate when the vessel was pressurized with air at ambient temperature, was heated with dry air, and when it was heated with steam. For air at ambient temperature, the change in leak rate fell between predictions based on laminar and turbulent flow. When the vessel was filled with heated dry air, the leak rate decreased rather than increased as predicted. When steam was injected, the leak rate at all monitored points fell to immeasurably small values. Evidently the steam condensate plugged the leakage paths, invalidating the assumption of constant leak path geometry.

The CSE leak rate measurements reported by Witherspoon^(6,7) were confirmed qualitatively by the leakage tests conducted in the CVTR containment vessel. Bingham⁽⁸⁾ reported a decrease in leak rate from 0.315% to 0.26% as a result of simulated DBA tests, confirming that the tightness of the containment vessel was not lessened by exposure to DBA conditions.

The second factor in the dose source term, Q , is the fission product concentration in the gas which escapes from the leak. The exiting gas concentration is equal to the inlet concentration multiplied by the decontamination factor achieved in the leak path. Mathematically this is

$$C_0 = \frac{C_i}{D} \quad (3)$$

where C_0 = fission product concentration in gas at exit of leak path,

D = decontamination factor in path,

C_i = fission product concentration in gas inside containment vessel.

A pictorial description of fission product leakage is shown in Figure 1.

FIGURE 1. Schematic Representation of Fission Product Leakage From a Containment Vessel

Both gas and liquid enter the leakage path. The liquid and gas become equilibrated as they pass through. In addition, transfer from the liquid to the solid surface will occur. The discharged gas carries with it some fission products. The leaked liquid will also carry fission products, a part of which will be released to the atmosphere. The fraction not released will remain at the site as liquid or possibly as a solid if it becomes evaporated.

Instantaneous leak rates and decontamination factors are very difficult to determine experimentally. The alternative is to measure these quantities over a time period of several minutes. The total mass of a fission product leaked is

$$\text{measured mass leaked} = \int_0^t C_o L \, dt + \int_0^t C_{LO} L_{liq} \, dt \quad (4)$$

where C_o = gas phase concentration at exit of leak,

L = gas leak rate,

C_{LO} = concentration in leaked liquid,

L_{liq} = liquid leak rate.

The measured mass leaked may be compared to that predicted from the internal gas concentration

$$\text{expected mass leaked} = \int_0^t C_i L \, dt \quad (5)$$

where C_i = fission product concentration inside vessel.

Equation (5) is the formulation currently used in reactor siting calculations. This formulation obviously neglects leaked liquid and interactions within the leak path.

Two decontamination factors can be defined. One is related to decontamination of airborne fission products within the leak path. It is defined as

$$DF_g = \frac{\text{expected mass leaked}}{\text{measured airborne mass leaked}} = \frac{\int_0^t C_i L \, dt}{\int_0^t C_o L \, dt} \quad (6)$$

The second decontamination factor is one which accounts for deposition and holdup within the leak path. The overall decontamination factor is defined as

$$DF_{ov} = \frac{\text{expected mass leaked}}{\text{total leaked mass}} = \frac{\int_0^t C_i L dt}{\int_0^t C_o L dt + \int_0^t C_{LO} L_{liq} dt} \quad (7)$$

The gas phase decontamination factor defined in Equation (6) is the one most applicable to airborne dose calculations. If fission products carried in the leaked liquid do not become airborne, the dose reduction factor due to leak path trapping is

$$(DRF)_{\text{leak path}} = DF_g \quad (8)$$

DESCRIPTION OF EXPERIMENTS PERFORMED

When the leakage experiments were being formulated, the plan was to study leaks from the containment vessel identified during leak tests at ambient temperature. The selected leakage point was to be fitted with a collector and a flow meter. Collection of leaked fission products and measurement of leakage flow would have permitted an evaluation of fission product leakage under accident conditions as compared to that predicted from ambient temperature leak tests and measured fission product concentrations. However, experience showed that the leak points plugged soon after the steam was introduced into the containment vessel. This approach was abandoned and artificial leak paths were used in most tests. Typically, the leak path was a small stainless steel needle valve, adjusted to give the desired leak rate with steam-air atmospheres.

All of the fission product leakage tests were carried out in conjunction with natural transport tests⁽⁹⁾ and air cleaning tests⁽¹⁰⁾ in the Containment Systems Experiment. Two factors favored this. First, the fission products remained airborne for a relatively long period, giving time for leakage measurements. In spray tests, the airborne iodine and cesium concentrations decreased rapidly, so that after a few minutes, the concentrations were low. This limited the detection sensitivity. Second, during spray tests, all surfaces were covered with liquid. This may have interfered with the leak flow.

The airborne fission product concentration was assessed using Maypack samplers suspended within the containment vessel. The containment vessel arrangement used in the natural transport experiments is shown schematically in Figure 2.

FIGURE 2. Containment Vessel Arrangement Used in Natural Process Tests in the Containment Systems Experiment

The Maypack sampler contains a series of adsorbent beds which permit the airborne fission products to be classified as particulate, reactive gas (elemental iodine) easily adsorbed gas (iodine compounds of intermediate volatility), and less easily adsorbing gas (methyl iodide). A cross-sectional view of the Maypack sampler is shown in Figure 3. McCormack⁽¹¹⁾

FIGURE 3. Cross-Sectional View of Maypack Sampler

has given a detailed description of the classification of radio-iodine achieved within a Maypack sampler.

The fission products simulant used in the CSE tests included elemental iodine, methyl iodide, cesium aerosol, and uranium oxide aerosol. Details of the generation methods, and analysis of the radioactive tracers, are given by Coleman⁽¹²⁾.

The leaked fission products were collected by passing the gas first through a glass, water-cooled condenser and then into a Maypack sampler. The collection train typical of that used in most tests is shown schematically in Figure 4. In order to minimize surface deposition, the leakage paths and collection train were made from stainless steel, teflon, and glass.

FIGURE 4. Sampling Train Used to Collect Leaked Fission Products

Several types of leak path geometries were studied. In the first set of experiments, the leak path was a stainless steel needle valve. The inlet tubing was inserted flush with the inner wall of the containment vessel. The geometry for this arrangement is pictured in Figure 5. The flush inlet was used to simulate leaks in which the leak path began at a surface covered with a liquid film.

FIGURE 5. Artificial Leak Path With Flush Inlet

In the second set of tests, the inlet tubes were extended past the inner wall of the vessel as shown in Figure 6. Extension of the inlet tube

FIGURE 6. Artificial Leak Path With Extended Tube Inlet

past the interior surface of the vessel wall prevents direct imbibition of wall liquid films. This type of inlet simulates leaks at penetrations where flowing liquid films would not be present. A third leak path geometry used a tube extended into the vessel. The end of the tube was closed off except for a pin hole in a 0.005" thick steel membrane. This leak path is shown in Figure 7. The orifice size was established by trial to give a flow rate

FIGURE 7. Artificial Leak Path With Orifice Inlet

near one liter per minute. This type of leak probably is not typical of any real leak path from a containment vessel. It does represent a "worst case" from the fission product leakage standpoint in that fission products would have little opportunity to deposit in the leak path.

The fourth and last type of leak path was perhaps more typical of expected leaks. The gas leaked past the first valve of a double butterfly valve installed on a ventilation line. The collection train used in this case was different in that no water-cooled condenser was used. A condenser was not needed because the volume between the two valves was considerably cooler than the containment vessel and was at about atmospheric pressure. The blades of

the butterfly valves were made from stainless steel and the seats from an ethylene-propylene elastomer rated for steam service. These valves were installed in an 8" ventilation line made from carbon steel. The interior surface of the ventilation pipe was likely covered with a thin layer of rust. This leak point and collection train is pictured in Figure 8. The charcoal

FIGURE 8. Collection of Fission Products Leaked From a Butterfly Valve

trap installed downstream from the Maypack was added to ensure that all methyl iodide would be retained during the long (1 day) sampling period. The wet test meter was used to provide an accurate integrated flow.

An important related question is the degree to which absorbed fission products can become released when the liquid is evaporated. In one run, liquid from the vessel sump was continuously evaporated, and the off gas was analyzed. The apparatus used for the liquid evaporation is shown in Figure 9.

FIGURE 9. Apparatus For Measuring Fission Product Release From Sump Liquid

The liquid evaporated was made up of 20 ml aliquots withdrawn at 60-minute intervals from the sump of the containment vessel. Ten such samples were added to the evaporating flask as they were withdrawn. A second set of these aliquots was added to a second flask which was held at room temperature for 24 hours before being evaporated. The purpose of this step was to determine if storage at room temperature influenced the fraction which was reversibly absorbed. The amount of material remaining in the evaporating flask was compared to the amount found in the traps.

RESULTS OF EXPERIMENTSLeak Inlet Flush With Containment Vessel Wall

In CSE Run A-5, the airborne fission products were attenuated by natural deposition only. The leak point was located in the wall of the vessel some 10 ft. above the floor of the main room. The leak path geometry was as shown in Figure 5, and the collection train was as shown in Figure 4. The leakage flow was maintained constant at 1 liter/minute. Constant attention was required to keep the flow rate near the target rate. The amounts of iodine and cesium leaked are compared with the amounts expected in Table 1.

Table 1. Iodine and Cesium Leakage Measured For
Inlet Flush With Wall In CSE Run A-5.

<u>Sampling Time(a) Minutes</u>	<u>Leak Rate ℓ/min.</u>	<u>Mass Collected Micrograms</u>	<u>Mass Expected(b) Micrograms</u>	<u>Decontamination Factor(c) (Total)</u>	<u>% Leaked Activity in Condensate</u>	<u>Decontamination Factor For Airborne Activity(d)</u>
<u>Total Iodine</u>						
23-28	1.0	450	745	1.65	95	33
48-63	1.0	211	252	1.19	90	12
73-103	1.0	173	71	0.41	76	1.7
168-308	1.0	<u>98</u>	<u>25</u>	<u>0.26</u>	51	<u>0.53</u>
Overall	1.0	932	1093	1.17		8.1
<u>Total Cesium</u>						
23-28	1.0	9.86	10.9	1.10	> 99	> 110
48-63	1.0	5.97	7.40	1.24	> 99	> 124
73-103	1.0	6.10	9.13	1.50	> 99	> 150
168-308	<u>1.0</u>	<u>3.74</u>	<u>1.38</u>	<u>0.37</u>	> 99	> <u>37</u>
Overall	1.0	25.67	28.81	1.12		> 112

(a) time after iodine injection.

(c) calculated using Eq. 7.

(b) calculated using Eq. 5.

(d) calculated using Eq. 6.

Two conclusions are apparent from these measurements. First, there is a chromatographic type holdup of fission product activity in the leak path. At early times, the amount of activity penetrating the leaks is less than expected. At longer times (after the airborne concentration within the containment vessel had decreased to very low levels), a fraction of the fission products retained in the leak path is evolved, causing the collected activity to be higher than expected. The second important conclusion is that most of the leaked activity was retained by the leaked liquid. Hence, for real leaks in which heat transfer causes condensation, most of the fission products will be retained by the condensate and will not be available for airborne transport. Virtually all of the cesium was retained by the liquid.

Measurements for a similar leak were also obtained during CSE Run A-16. The sample periods were longer. The data obtained are shown in Table 2.

Table 2. Iodine and Cesium Leakage Measured For
Inlet Flush With Wall in CSE Run A-16

<u>Sampling Time Minutes</u>	<u>Leakage Flow Rate Liters/min.</u>	<u>Micrograms Collected</u>	<u>Micrograms Expected</u>	<u>Decontami- nation Factor</u>	<u>% Leaked Activity in Condensate</u>	<u>Decontamination Factor For Airborne Activity</u>
<u>Total Iodine</u>						
69-90	1.0	73	67	0.92	94.8	17.6
99-120	1.0	14	14	1.00	90.6	10.4
155-275	1.0	9	7	0.78	78.2	3.5
313-1350	0.6	<u>9</u>	<u>16</u>	<u>1.78</u>	71.4	<u>6.1</u>
Overall		105	104	0.99		10.8
<u>Total Cesium</u>						
69-90	1.0	124	186	1.5	99.9	1500
99-120	1.0	145	187	1.3	99.9	1300
155-275	1.0	28	90	3.2	100.0	> 3200
313-1350	0.6	<u>9</u>	<u>1.4</u>	<u>0.16</u>	100.0	> <u>160</u>
Overall		306	464	1.5		> 1500

Results are similar to those obtained in Run A-5. The holdup of total activity in the leak path was slightly less than for Run A-5. The iodine remaining airborne after the leaked gases had passed through the ambient temperature condenser was only about 10% of the total iodide leaked. This agrees well with the results listed in Table 1. The distribution of the airborne iodine within the Maypack sampler is shown in Table 3.

Table 3. Maypack Analysis of Leaked Fission Products
For Inlet Flush With Wall In CSE Run A-16.

Sampling Time Minutes	<u>Percent of Maypack Activity in Component</u>			
	<u>Filter (Particulate)</u>	<u>Silver (Elemental Iodine)</u>	<u>Charcoal Paper (Molecular Iodides)</u>	<u>Charcoal Bed (Methyl Iodide)</u>
69-90	1.0	2.9	38.5	57.6
99-120	0.4	2.5	28.7	68.4
155-275	0.8	4.4	17.0	77.8
313-1350	1.6	6.3	16.8	75.3

The Maypack data presented in Table 3 show that most of the iodine penetrating the condenser is methyl iodide. Elemental iodine is effectively trapped by the liquid water.

Cesium, present as an aerosol particle, was effectively trapped by liquid in the leak path. Results for this test showed decontamination factors near unity when based on gas-borne plus liquid-borne effluent. The decontamination factor based on gas-borne cesium particles was greater than 1500.

Leak Inlet Protruding Into Containment Vessel Atmosphere

As a variation of leak path geometry, two stainless steel tubes (0.18" ID) were extended into the containment vessel through a blind flange. The leak restriction for each tube was a stainless steel needle valve located just outside the flange. The leak path type is shown in Figure 6. The two leak paths were identical in geometry. One was adjusted to give a leak rate of 1 liter per minute, and the other was adjusted to give a leak rate of 0.1 liter

per minute. The masses of iodine and cesium collected for these two leaks are compared to the masses expected on the basis of the airborne concentration in Table 4. These results for the protruding inlet are similar to those

Table 4. Fission Products Leaked From a Protruding Inlet Path In CSE Run A-11.

<u>Sampling Time Minutes</u>	<u>Leakage Flow Rate Liter/min.</u>	<u>Mass Collected Micrograms</u>	<u>Mass Expected Micrograms</u>	<u>Decontamination Factor</u>	<u>% Leaked Activity in Condensate</u>	<u>Decontamination Factor For Airborne Activity</u>
<u>Total Iodine</u>						
15-30	1	136	829	6.1	88	51
50-80	1	187	411	2.2	89	20
240-480	1	<u>779</u>	<u>421</u>	<u>0.54</u>	<u>46</u>	<u>1</u>
Overall		1102	1661	1.51		3.6
15-30	0.1	11.6	82.9	7.2	16	8.6
50-80	0.1	6.6	41.1	6.2	70	20.6
240-480	0.1	<u>48.3</u>	<u>42.1</u>	<u>0.9</u>	76	<u>3.8</u>
Overall		66.5	166.1	2.5		4.1
<u>Total Cesium</u>						
15-30	1	9.8	22.1	2.3	> 99	> 230
50-80	1	17.3	22.8	1.3	> 99	> 130
240-480	1	<u>349.4</u>	<u>433.0</u>	<u>1.2</u>	> 99	> <u>120</u>
Overall		376.5	477.9	1.3		> 130
15-30	0.1	5.7	2.2	0.4	> 99	> 40
50-80	0.1	0.2	2.3	11.5	> 99	> 1150
240-480	0.1	<u>3.0</u>	<u>43.3</u>	<u>14.4</u>	> 99	> <u>1440</u>
Overall		8.9	47.8	5.37		> 537

for the flush inlet. Iodine is held up in the leak path at early times and

is then re-evolved at longer times. The smaller leak evolved comparatively less fission products. The longer holdup time associated with the lower flow rate accounts for this.

Cesium was removed more effectively than iodine. This high degree of removal for cesium agrees with the measurements shown in Tables 1 and 2.

Similar data for a protruding inlet were obtained in A-16. For the latter test, the leak rate was set just prior to iodine injection, and then allowed to vary naturally for the first 120 minutes after iodine injection. This operational procedure more nearly duplicates that for a real leak. Results of these measurements are shown in Table 5.

Table 5. Iodine and Cesium Leakage From A Protruding Inlet Path in CSE Run A-16.

<u>Sampling Time Minutes</u>	<u>Average Leakage Flow Rate Liter/min.</u>	<u>Mass Collected Micrograms</u>	<u>Mass Expected Micrograms</u>	<u>Decontamination Factor (Total)</u>	<u>% Leaked Activity in Condensate</u>	<u>Decontamination Factor For Airborne Activity</u>
<u>Total Iodine</u>						
0-120	0.5	300	350	1.2	61.9	3.1
155-264	1.6	<u>23</u>	<u>90</u>	<u>3.9</u>	81.8	<u>22</u>
Overall		323	440	1.4		3.73
<u>Total Cesium</u>						
0-120	0.5	210	485	2.3	97.6	96
155-264	1.6	<u>94</u>	<u>125</u>	<u>1.3</u>	97.4	<u>51</u>
Overall		304	610	2.0		81

The distribution of activity deposited within the Maypack for this leak is given in Table 6.

For the 120 minute period, a 2-hour dose reduction factor of 3.1 was achieved for iodine airborne downstream from the condenser. The leaked material was mostly methyl iodide. The second sample gave a higher decontamination factor. This is apparently due to the presence of a higher

Table 6. Maypack Analysis of Iodine Leaked From
A Protruding Inlet Path in CSE Run A-16.

<u>Sampling Time Minutes</u>	<u>Filter (Particulate)</u>	<u>Silver (Elemental Iodine)</u>	<u>Charcoal Paper (Molecular Iodides)</u>	<u>Charcoal Bed (Methyl Iodide)</u>
0-120	0	0.5	1.6	97.5
155-264	4.4	28.6	10.5	57.5

proportion of elemental iodine as indicated by the Maypack analysis in Table 6. It should be pointed out that in Run A-16 elemental iodine was released at a constant rate for the time interval 0 to 60 minutes. Cesium was again efficiently collected by the leaked liquid.

Orifice Inlet Leak

One set of experiments was carried out with the orifice inlet shown in Figure 7. This leak geometry was designed to provide comparative data for a well defined leak path representing a limiting case. The results are tabulated in Table 7.

Table 7. Iodine and Cesium Leaked From
An Orifice During CSE Run A-16.

<u>Sampling Time Minutes</u>	<u>Average Leakage Flow Rate Liters/min.</u>	<u>Mass Collected Micrograms</u>	<u>Mass Expected Micrograms</u>	<u>Decontami- nation Factor</u>	<u>% Leaked Activity in Condensate</u>	<u>Decontami- nation Factor Airborne Activity</u>
<u>Total Iodine</u>						
0-120	1.6	640	1190	1.9	53.2	4.0
155-275	1.5	22	86	3.9	89.4	37
313-1345	0.7	<u>34</u>	<u>19</u>	<u>0.56</u>	46.6	<u>1.0</u>
Overall		696	1295	1.85		3.81
<u>Total Cesium</u>						
0-120	1.6	560	1550	2.8	62.0	7.3
155-275	1.5	44	132	3.0	96.2	77.5
313-1345	0.7	<u>430</u>	<u>34</u>	<u>0.08</u>	68.4	<u>0.25</u>
Overall		1034	1716	1.7		4.9

Results for iodine are not greatly different than those obtained for the other leak geometries. For cesium, however, appreciably less decontamination occurs in the leak path. This result is reasonable because the orifice would not be an effective trap for aerosol particles. The distribution of iodine within the Maypack trap is shown in Table 8.

Table 8. Maypack Analysis of Iodine Leaked From
An Orifice In CSE Run A-16.

Sampling Time Minutes	<u>Percent of Maypack Activity in Component</u>			
	Filter (Particulate)	Silver (Elemental Iodine)	Charcoal Paper (Molecular Iodides)	Charcoal Bed (Methyl Iodide)
0-120	34.2	0.6	6.4	58.8
155-275	10.4	9.5	17.9	62.2
313-1345	7.7	49.2	6.8	36.3

The Maypack analyses show that more of the leaked material is in the form of an airborne particle than in previous tests. This is consistent with the relatively low decontamination factors observed for cesium. Overall, the Maypack analyses are consistent with previous data in that most of the airborne iodine which escaped from the condenser is methyl iodide.

Leak Past Ventilation Isolation Valve

A final leak geometry studied involved leakage past a butterfly valve as shown in Figure 8. The leak rate was not adjusted during the day-long collection period. Also, the leaked gas was not passed through a condenser, but merely through a glass condensate trap. A condenser was unnecessary because condensation had already occurred within the piping between the two butterfly valves. The leaked fission product masses are compared with the predicted masses in Table 9. The total decontamination of iodine and cesium is much larger than observed for the stainless steel leak paths. Two factors combine to cause this greater decontamination. First, the leaked liquid was not collected, but remained within the valve chamber. Second, the iron surfaces of the valve chamber would be

Table 9. Iodine and Cesium Leaked From A
Butterfly Valve in CSE Run A-16.

<u>Sampling Time Minutes</u>	<u>Average Leakage Flow Rate Liter/min.</u>	<u>Mass Collected Micrograms</u>	<u>Mass Expected Micrograms</u>	<u>Decontami- nation Factor</u>	<u>% Leaked Activity in Liquid Trap</u>	<u>Decontamination Factor For Airborne Activity</u>
<u>Total Iodine</u>						
0-1368	1.0	24	820	34	0	34
<u>Total Cesium</u>						
0-1368	1.0	7	1350	190	10	210

expected to sorb iodine more effectively than the much smaller stainless steel surfaces exposed in the artificial leak paths.

The character of the leaked iodine, as shown by a liquid trap and a Maypack sampler, is shown in Table 10.

Table 10. Maypack Analysis of Iodine Leaked From
8" Butterfly Valve in CSE Run A-16.

<u>Sampling Time (Minutes)</u>	<u>Liquid Trap</u>	<u>Filter (Particulate)</u>	<u>Silver (Elemental Iodine)</u>	<u>Charcoal Paper (Molecular Iodine)</u>	<u>Charcoal Bed (Methyl Iodide)</u>
0-1368	0	0.9	28.4	5.6	65.1

Most of the leaked activity is methyl iodide. This leak path is the most realistic one studied. The observed decontamination factor is the largest observed for a similar time period, which re-enforces the decontamination factors observed for the artificial leaks.

Retention of Fission Product By Leaked Liquid

An important factor in the leakage measurements is the degree to which fission products are retained by leaked liquid. A test was carried out in Run A-16 to determine what fraction of iodine and cesium could be made airborne by evaporating liquid to dryness.

Liquid was collected from the vessel sump and added to a continuous evaporation flask as pictured in Figure 9. Sump liquid was formed by steam condensation, hence should closely represent condensate exiting from a leak path. Duplicate samples were accumulated in a glass flask and stored at room temperature for 24 hours. This liquid was then evaporated in a similar way to determine the influence of storage time on release of iodine. Results of this evaporation are given in Table 11.

Table 11. Iodine and Cesium Carry-Over
From Evaporated Liquid.

Collection Train Component	<u>Continuous Aliquot Addition</u>		<u>Stored Cumulative Aliquot</u>	
	<u>Mass Iodine Micrograms</u>	<u>Mass Cesium Micrograms</u>	<u>Mass Iodine Micrograms</u>	<u>Mass Cesium Micrograms</u>
Residue in Flask	457	573.0	458	593
Condensate	12.0	8.3	12.1	0.29
Filter	0.9	0.02	1.8	0.002
Silver	0.3	--	2.0	0.02
Charcoal Paper	0.9	--	1.1	0.01
Charcoal Bed	<u>3.3</u>	<u>0.04</u>	<u>3.6</u>	<u>0.04</u>
Total	474.4	581.36	478.6	593.362

Two important facts are apparent from the results of the evaporation. First, most of the fission products are retained in the solid residue remaining in the flask. Less than 4.5% of the iodine and less than 1.5% of the cesium contained in the liquid was released to the gas phase. Since cesium is non-volatile, the observed carry-over of cesium must have been the result of mechanical entrainment. Thus iodine loss via volatilization is estimated to have been 3.2% for the continuous aliquot and 4.2% for the stored aliquot. Second, the one-day storage time had little influence on the degree of retention. Chemical reactions which bound the iodine in an irreversible manner occurred by the time the first sample was withdrawn.

DISCUSSION OF RESULTS

The transport of fission products through leak paths in nuclear reactor containment vessels is important because only leaked materials can be carried off-site. Since fission products are not generated within a leak path, the issue at question is "how fission products are held up in the leak path."

The degree of retention would be expected to depend on the geometry and materials of construction of the leak path, as well as on the chemical and physical forms of fission products. Since geometry is a factor, it is difficult to generalize to predict leakage under accident conditions where leak path geometry is not known. Thus one cannot expect to arrive at definitive predictions of leak path decontamination for postulated accidents. One can, however, expect to obtain information for special cases which are believed to represent potential leak situations under accident conditions.

When this work was planned, the goal was to identify natural leaks in the CSE containment vessel during the air leak tests prior to each major test. These leaks were to be fitted with collection apparatus so that the volumetric leak rate and the leak rate of fission products could be measured throughout the CSE test. Attempts to do this were unsuccessful. The leaks simply plugged when steam was introduced to heat the vessel atmosphere. Hence it early became obvious that leakage data could be obtained only by using artificial leak paths which could be adjusted to assure a measurable flow rate. This process of providing artificial leaks obviously overturns a large measure of conservatism naturally associated with leaks. Whereas the leak would naturally plug, we simply enlarged it enough so that it would not remain plugged. The results of the tests with artificial leaks can thus be viewed as conservative. The one test with a natural leak path (butterfly valve) verified the contention that decontamination in natural leaks would be greater than that in artificial paths constructed of stainless steel.

It should be mentioned that the measurements made here do not agree with the conclusions of Philbin⁽¹⁾, who concluded that iodine was not retained within the leak path, and that condensed water could greatly enhance the leak rate of iodine. Neither of these conclusions is borne out by the present work. Philbin⁽¹⁾ did not measure the iodine concentrations within his chamber, but apparently calculated it as though it was saturated vapor. It is anticipated

that experiments with unrealistically high iodine concentrations would give results such as those obtained by Philbin⁽¹⁾. The experiments conducted in the CSE vessel should be more representative of LOCA in light water reactors.

The measurements reported here were subject to errors, the largest of which was the measurement of the leak rate as a function of time. It was estimated that the total error in assessing the leaked material was no larger than $\pm 10\%$. The expected concentrations were calculated from gas phase concentration data which were also subject to errors of $\pm 10\%$. Thus it was concluded that the calculated decontamination factors were subject of up to $\pm 20\%$.

Physical conditions for the CSE tests applicable to the leakage experiments are given in the Appendix.

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NOMENCLATURE

C_i	concentration of radioactive material in contained gas, curies/volume
C_{LO}	concentration of radioactive material in leaked liquid, curies/volume
C_0	concentration of radioactive material in leaked gas, curies/volume
D	decontamination factor for material passing through a leak, dimensionless
DBA	design basis accident
DF_g	decontamination factor for airborne material passing through a leak, dimensionless
DF_{ov}	overall decontamination within a leak path, including gas borne and liquid borne effluent, dimensionless
DRF	dose reduction factor = $\frac{\text{mass leaked if concentration remained constant}}{\text{actual mass leaked}}$, dimensionless
F_ℓ	laminar flow extrapolation factor, dimensionless
L	gaseous leak rate from containment volume, volume/time
L_a	leak rate predicted for accident conditions, volume/time
L_{liq}	liquid leak rate, volume/time
L_T	leak rate measured at ambient temperature, volume/time
Q	rate of release of radioactive material from a containment vessel, curies/time
t	time

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APPENDIX

SUMMARY OF CONDITIONS FOR CSE TESTS
RELATED TO FISSION PRODUCT LEAKAGE EXPERIMENTS

APPENDIX

SUMMARY OF CONDITIONS FOR CSE TESTS
RELATED TO FISSION PRODUCT LEAKAGE EXPERIMENTS

The following data are intended to provide the reader with an overall view of the test conditions for CSE Runs A-5, A-11, and A-16. The fission product leakage tests were carried out in conjunction with these CSE runs. Additional details on Runs A-5 and A-11 are given by Hilliard in Reference 9. McCormack gives detailed information concerning Run A-16 in Reference 10. The simulant generation techniques and analytical methods are described in detail by Coleman⁽¹²⁾. McCormack⁽¹¹⁾ has provided a detailed account of iodine sample classification by the CSE Maypack samplers.

Several of the important test conditions are summarized in Table A-1. The concentrations of airborne fission products are listed in Tables A-2, A-3, and A-4.

Table A-1. Summary of Test Conditions for CSE Experiments

Parameter	<u>Value For CSE Runs</u>		
	A-5	A-11	A-16
Containment Atmosphere	Saturated Steam-Air	Saturated Steam-Air	Saturated Steam-Air
Test Type	Natural Transport Steady Temperature	Natural Transport Decaying Temperature	Filter Loop Cleanup
Initial Temperature °F	253	252	245
Initial Pressure, psia	47	49	48
Release Period for Elemental Iodine	0-10 min	0-10 min	0-60 min
Release Period for Cesium	0-10 min	0-10 min	0-120 min
Release Conc.* for Elemental Iodine, mg/M ³	140	162	176
Release Conc.* for Cesium	1.85	8.44	361
*release conc. = $\frac{\text{mass released}}{596 \text{ M}^3}$			

TABLE A-2. 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.

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TABLE A-3. Concentration in the Main Vapor Space--Run A-11^(a)

Time, min	Mean Concentration, $\mu\text{g}/\text{m}^3 \pm 1 \sigma$ (%) ^(b)										
	22	52	60	90	120	181	300	480	960	1440	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)	9.51 (104)
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)	3.80 (165)
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)	76.9 (18)
Iodine on Maypack Inlet	203 (27)	284 (89)	340 (43)	175 (45)	120 (32)	335 (45)	162 (76)	162 (168)	(c)	(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)	90.2 (20)
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)	443 (20)
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)	533 (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)	15.4 (39)
Uranium	1,650 (11)	678 (19)	553 (22)	462 (13)	368 (29)	383 (35)	284 (28)	247 (18)	87 (30)	55 (44)	(c)

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-4. Concentration in the Main Vapor Space--Run A-16^(a)

Time, min	Mean Concentration, $\mu\text{g}/\text{m}^3 \pm 1 \sigma$ (%) ^(b)										
	10	30	61	86	111	141	181	301	1441	1801	2760
Elemental Iodine	2170 (19)	3310 (17)	5350 (7.5)	314 (31)	123 (28)	66 (31)	27 (37)	24 (36)	4.8 (85)	c	c
Particulate Iodine	2430 (26)	4060 (15)	4110 (15)	1660 (14)	442 (7)	94 (16)	4.9 (94)	c	2.8 (72)	1.7 (119)	c
Iodine Associated with Charcoal Paper	125 (36)	98 (13)	93 (39)	23 (27)	11 (23)	7.9 (52)	2.4 (75)	2.1 (77)	0.7 (101)	0.2 (530)	0.4 (239)
Iodine on Maypack Inlet	297 (42)	370 (43)	520 (44)	67 (25)	19 (76)	8.0 (89)	c	2.0 (260)	c	c	c
Total Inorganic Iodine	5020	7840	10,070	2064	595	176	34.3	28.1	8.3	1.9	0.4
Methyl Iodide	2620 (14)	2470 (7)	629 (14)	217 (14)	88 (6)	34 (16)	14 (17)	11 (57)	1.9 (43)	1.4 (61)	0.8 (78)
Total Iodine (All Forms)	7640	10,310	10,700	2280	683	210	48	39	10	3.3	1.2
Cesium	4310	8320	7945	9175	10,100	1700	122	c	c	c	c

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

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

c. Not detectable.

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