REMOVAL OF AIRBORNE FISSION PRODUCTS BY RECIRCULATING
FILTER SYSTEMS IN THE CONTAINMENT SYSTEMS EXPERIMENT

J. D. McCormack^(a)

R. K. Hilliard^(b)

A. K. Postma

Mechanical Engineering Department

June 1971

(a) J. D. McCormack is presently with WADCO Corporation,
Richland, Washington.

(b) Chemical Technology Department.

BATTELLE MEMORIAL INSTITUTE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

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ABSTRACT

Five large-scale demonstration tests were carried out in the 26,500 cubic foot Containment Systems Experiment vessel, using an internal recirculating filter loop to remove airborne fission product simulants. Loop components were nominal 2 ft by 2 ft, commercially available units including a heat exchanger, demister, roughing filter, high efficiency particulate filter, and impregnated charcoal adsorbers. Flow through the loop was either 1000 or 1800 CFM and in four of the tests the containment atmosphere was saturated steam-air at 250 °F, 48 psig. This system performed satisfactorily, removing both particulate and gaseous fission products, including methyl iodide. Elemental iodine was removed initially at a rate somewhat faster than predicted for 100% efficiency, due to wall deposition. Particles were removed with an efficiency of 100%, and methyl iodide with an efficiency of 80%. At long times, the airborne concentration of all fission products approached equilibrium levels equivalent to decontamination factors greater than 10,000.

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INTRODUCTION

This report describes the results of large scale experiments on removal of airborne fission products from post-accident containment atmospheres by recirculating filter systems. This work was carried out as part of the Containment Systems Experiment (CSE) program sponsored by the USAEC Division of Reactor Development and Technology. In addition to the filter systems tests described in this report, the CSE program also provided extensive data on fission product removal by natural processes and by sprays. Hilliard^(1,2) has discussed the removal of aerosols and gases by surface deposition which would occur in a large containment vessel following a postulated accident. Hilliard⁽³⁾, and Postma^(4,5) have described the washout of airborne fission products by containment vessel sprays.

The trend in the nuclear power industry is for more reliance on engineered safety features to meet the licensing requirements set forth by the AEC reactor site criteria^(6,7). Filter systems and sprays are two engineered safety systems currently used to reduce the potential for leakage of radioactive materials from a containment vessel following a design basis accident (DBA).

Filter systems operate to reduce leakage potential in two ways. First, they clean the containment vessel atmosphere. This reduces the mass concentration of fission products in leaked gas. Second, the filter system may include a heat exchanger which cools the containment atmosphere and lowers the pressure. This reduces the calculated leak rate, which reduces the mass of fission products leaked to the atmosphere.

Filter systems have an advantage over sprays in that methyl iodide is removed nearly as rapidly as elemental iodine. Sprays are capable of

removing elemental iodine much more rapidly than can be achieved by a practical filter system. Aerosol particles are effectively removed by both sprays and filter systems.

OBJECTIVES OF FILTER SYSTEM EXPERIMENTS

The objectives of the filter system experiments were as follows:

- Measure the rate of removal of fission products from the containment atmosphere and compare with simple models based on a well-mixed atmosphere.
- Determine the loop component efficiencies, using full scale, commercially available components.
- Measure pressure drop and mechanical reliability for a loop operating in a post-accident environment.

SUMMARY AND CONCLUSIONS

Five large scale demonstration tests were carried out as part of the Containment Systems Experiment using a recirculating filter loop to remove airborne fission product simulants. The objectives of the tests were to assess the rate and extent to which the containment atmosphere was cleaned, and to measure loop performance in typical post-accident atmospheres.

Full sized, commercially available filters and charcoal adsorbers were used in a loop operating at 1000 cfm and 1800 cfm. One experiment was carried out at 80 °F and the other four used saturated steam-air at 250 °F. The loop was operated for 24 or 48 hours following injection of fission product simulants.

The results of the five experiments support the following conclusions.

- The filter loop removed airborne fission products with an initial rate which was in agreement with that predicted on the basis of a well-mixed containment volume.
- After the gas phase concentration was reduced to about 1% of initial concentration, desorption of fission products which had deposited by natural processes reduced the net rate at which the loop removed airborne materials.

- The charcoal beds effectively removed iodine-131 which was combined chemically as methyl iodide. At 100% relative humidity, the removal efficiency was about 80%, and at lower relative humidities the removal efficiency was greater than 99%.
- The loop components performed as expected in all atmospheric conditions tested. Deletion of the heat exchanger and moisture separator caused only minor effects on the filter pressure drop.

BACKGROUND INFORMATION

The filter system experiments were designed to give information which could be used in full scale containment vessels. Extrapolation to other situations requires a mathematical model based on theoretical concepts. The theory concepts which were considered important in the experimental design are briefly described in this section of the report.

MACROSCOPIC MODEL OF CLEANUP WITH A FILTER LOOP

The rate at which fission products are removed from the gas phase may be related to the system parameters by making a material balance on the gas phase. The source and sink terms are depicted graphically in Figure 1. The differential equation for a particular species in one of the rooms may be obtained by balancing input and output terms, on the assumption that the gas phase is well mixed.

The input rate is

$$\text{input rate} = G + F C_g (1-E) + R \quad (1)$$

where G = simulant injection rate,

F = flow rate through filter loop,

C_g = airborne concentration in main room,

E = fraction of airborne specie removed by filter in single pass,

R = rate of production of specie by vapor phase reaction.

FIGURE 1. Schematic Diagram of Fission Product Removal by a Filter Loop in the CSE Vessel

The output rate is

$$\text{output rate} = F C_g + k_g A(C_g - C_{gi}) + B_1 (C_g - C_{g1}) \quad (2)$$

where k_g = interfacial mass transfer coefficient,
 A = interfacial area for mass transfer,
 C_{gi} = solute concentration at interface,
 B_1 = inter-room transport coefficient,
 C_{g1} = concentration in gas of adjacent room.

The accumulation rate of a specie in a well mixed volume, V , is

$$\text{accumulation rate} = V \frac{dC_g}{dt} . \quad (3)$$

A mass balance based on Equations (1), (2), and (3) is

$$G + F C_g(1-E) + R = F C_g + k_g A(C_g - C_{gi}) + B_1 (C_g - C_{g1}) + V \frac{dC_g}{dt} \quad (4)$$

This equation is the mass balance for only one room. Similar equations for other rooms would also have to be written. In addition, similar balances on the liquid phase to account for equilibrium are needed to evaluate the quantity C_{gi} . Fortunately, for early and late times, many of the terms in Eq. (4) are small compared to the dominant ones.

Most of the filter systems tests were carried out using short term releases of simulants. Early in the run, the generation rate, R , may be neglected compared to removal. Also, inter-room transport is small compared to removal for early times. For early times then, Eq. (4) may be written as

$$\frac{dC_g}{dt} = - \frac{FE}{V} C_g - k_g A(C_g - C_{gi}) . \quad (5)$$

Thus the initial removal rate is the sum of the cleanup caused by the filter loop, and that due to surface deposition. Hilliard⁽²⁾ has shown that for elemental iodine, the perfect sink assumption, $C_{gi} = 0$, applies until the gas phase concentration falls to 0.01 of the initial concentration. For particles, one would expect the perfect sink assumption to be true for all times. For methyl iodide, the quantity $(C_g - C_{gi})$ is very small so that surface deposition is negligible compared to removal by the

filter system. Thus for early times, the removal rate for elemental iodine and particles is

$$\frac{dC_g}{dt} = -\frac{FE}{V}C_g - \frac{k_g A C_g}{V}. \quad (6)$$

For methyl iodide, surface deposition is negligible, giving

$$\frac{dC_g}{dt} = -\frac{FE}{V}C_g \quad (7)$$

Equations (6) and (7) describe the removal rate in terms of specified or readily calculable parameters. These equations are easily integrated if one assumes that F , E , and V are constants. Equation (6) becomes

$$\frac{C_g}{C_{g0}} = \exp \left[-\left(\frac{FE}{V} + \frac{k_g A}{V} \right) t \right] \quad (8)$$

where C_{g0} = concentration in gas at $t = 0$.

Equation (7) becomes

$$\frac{C_g}{C_{g0}} = \exp \left[-\left(\frac{FE}{V} \right) t \right] \quad (9)$$

DISCUSSION OF ASSUMPTIONS USED IN REMOVAL MODEL

Several assumptions were used to develop the removal rate equations, Eq. (8) and Eq. (9). We will briefly note the basis for the assumptions and the conditions for which the assumptions would apply.

The gas phase of an individual room was assumed to be well mixed. Earlier tests in CSE have shown this a good assumption. Samplers located throughout the gas space indicated a uniform concentration within a few minutes after fission product injection had ceased. This was true where only natural convection currents caused mixing. Filter loops cause mechanical mixing of air, hence the gas space within a room would surely be well mixed.

Inter-room transport was neglected for early times. This will be true if the quantity, FEC_g , is much larger than $B_1 (C_g - C_{g1})$. Initially

the concentration in the connected room, C_{g1} , is 0. Hence inter-room transport at early times will be of minor importance if FE is much larger than the inter-room transport coefficient, B_1 . The latter is surely true for all time periods, and hence it is concluded that inter-room transport will be negligible until $C_g < C_{g1}$. Experience in the CSE has shown that the maximum concentration attained in the connected rooms is never more than a few percent of the initial main room concentration. Thus neglecting inter-room transport would introduce little error until main concentrations were reduced to less than 1% of the initial concentration. For long times, the main room concentration will be reduced to a level such that C_g is less than C_{g1} . At this point inter-room transport will be significant. Models which apply for long times must account for this.

The perfect sink assumption, $C_{gi} = 0$, has been shown to apply for elemental iodine until the main room concentration is reduced to less than 1% of the initial concentration. At longer times, the interfacial concentration, C_{gi} would likely become larger than that of the bulk gas phase. When this happens, the deposited material becomes a source which will limit the removal achieved by the filter loop. For particles, this kind of desorption would not occur, hence the perfect sink assumption should hold for all times.

Although aerosol particles are not reversibly absorbed as are gases, they can become airborne from solution by physical processes which mechanically disperse small liquid drops. Examples of such process include the breaking of bubbles and formation of drops. Both of these processes would occur within containment vessels. The generation rate of non-volatile particles by mechanical dispersion is very small on an absolute scale. The gas phase concentration would have to be reduced to very low levels before this generation rate were important. In summary, particles could become resuspended to a small degree by mechanical dispersion processes. The resuspension rate would be of negligible consequence until the gas phase concentration were reduced to a very low level. The mechanical resuspension would limit the ultimate decontamination factor which could be achieved.

Gas phase generation of fission product types has been neglected. This is a good assumption for CSE tests. For accident conditions, high radiation fields might cause some change. Current calculations by personnel in the AEC Regulatory Branch are based on the assumption that airborne elemental iodine forms methyl iodide. The impact of this formation is minor because the filter system is capable of removing methyl iodide nearly as fast as elemental iodine.

Finally, the flow rate and removal efficiency were assumed constant. Since flow rate is an independent, specified parameter, assumption of constancy is justified. For elemental iodine and particles, removal efficiency is greater than 95%. Since removal efficiency is a linear multiplier in the exponential terms of the removal equation, the removal rate is influenced to only a minor extent as efficiency varies between 95% and 100%. For methyl iodide, removal efficiency is potentially lower and more variable. Performance data will require analysis to justify the assumption of constant removal efficiency.

In summary, it appears that a simple first order theory for fission product removal by recirculatory filter loops should apply until airborne concentrations are reduced to about 1% of the initial concentration. Thus 2-hour dose reduction factors can be reliably predicted from the simple first order, perfect sink theory. For longer time periods, one must account for the approach to equilibrium.

TRAPPING OF FISSION PRODUCTS BY FILTER LOOP COMPONENTS

Particle Removal By High Efficiency Filters

The high efficiency filters used in air cleaning systems are built to standards for use in reactor containment systems. Performance specifications require an efficiency greater than 99% for 0.3 μ diameter droplets. For nuclear safety systems, Burchsted⁽⁸⁾ lists performance specifications which require maximum penetration to less than 0.03% when the filter is operated at rated flow. The increase in penetration when tested at 20% of rated capacity must not exceed 0.01%. These efficiency requirements are based on 0.3 μ particles, the most difficult to capture. Hence for

aerosols encountered in a containment system, the removal efficiency should be 99.97% or higher. Thus, one can use $E = 1.0$ in the removal equation and be assured that it applies for all particle sizes for all time periods.

Adsorption of Elemental Iodine by Charcoal

Elemental iodine is efficiently adsorbed by activated charcoal. When exposed as a packed bed of granules, the removal efficiency depends on mass transfer parameters and on the equilibrium adsorption capability of the charcoal. Steps in retention include:

- diffusion across the gas film at the surface of each granule
- diffusion in gas capillaries within the granule (pore diffusion)
- solid phase diffusion along the pore surface or within the solid matrix
- reaction at the solid surface or within the solid matrix.

If the mass of iodine adsorbed is small, the last three steps listed are fast compared to the first. The adsorption is then governed by mass transfer in the gas phase. Dale and Klotz⁽⁹⁾ have published a correlation for adsorption in a packed bed for the case in which gas phase mass transfer limits the adsorption.

$$\log_e \frac{C_1}{C_2} = \frac{1.82 x_0 a}{(Re_p)^{0.51} (Sc)^{0.67}} \quad (9)$$

- where C_1 = concentration in inlet gas,
 C_2 = concentration in effluent gas,
 x_0 = bed depth,
 a = effective area of adsorbent per unit volume of bed,
 Re_p = Reynolds number for a granule, $\frac{d_p U \rho}{\mu}$,
 Sc = Schmidt number, $\frac{\mu}{\rho D_v}$,
 d_p = average carbon particle diameter,
 U = superficial gas velocity,

- ρ = density of gas,
 μ = absolute viscosity of gas,
 D_v = diffusivity of iodine in gas phase.

This predictive equation has been independently verified by Prigge⁽¹⁰⁾ who measured removal efficiencies of elemental iodine in steam-air mixtures as a function of gas velocity, bed thickness, temperature, and iodine concentration, for several types of charcoal. Charcoal beds can be highly efficient for iodine removal. Prigge⁽¹⁰⁾ consistently measured removal efficiencies greater than 99.97% for his test conditions.

The gas phase limited efficiency indicated in Eq. (9) cannot be expected to continue indefinitely in a recirculating filter system. A very small fractional desorption would be expected to occur. After the gas phase concentration had been reduced to a very low level, this desorption should lower the overall removal efficiency. Relatively little data is available^(11, 12) on desorption; hence it is not possible to pin-point the concentration level at which desorption would be important. It is certainly true that desorption would not be a factor until the airborne concentration had decreased by a large factor ($> 10^3$).

Adsorption of Methyl Iodide by Charcoal

Methyl iodide is adsorbed less readily by activated charcoal than is elemental iodine. Experiments reported by Adams, et al⁽¹³⁾ and by Ackley and Adams⁽¹⁴⁾ showed that under dry conditions methyl iodide retention by charcoal was less than for elemental iodine, and that the efficiency approached zero as the relative humidity of the gas stream approached 100%.

It has been found that methyl iodide will exchange iodine atoms while deposited temporarily on activated charcoal. This exchange reaction is currently used to ensure worthwhile removal efficiencies of methyl iodide. The chemical reaction may be written as

Iodine added to the charcoal as an impregnant exchanges with I-131 bound chemically as methyl iodide. This effects a net decontamination even though

methyl iodide is not permanently trapped. Kovach⁽¹⁵⁾ reviewed available information on methyl iodide trapping under accident conditions and concluded that single pass systems would give removal efficiencies of the order of 70%. It was recognized that carbon beds might, under very exceptional conditions, become temporarily flooded with water, which reduces the activity removal efficiency, but that properly designed air handling systems, consisting of moisture separators and HEPA filters, would prevent water droplet carry through the carbon bed.

Heating of Charcoal Beds in Containment Atmospheres

Charcoal bed heating is important from two viewpoints. First, if the bed is warmed initially by flowing steam-air mixtures, one can imagine the bed becoming flooded with condensate. This condensate, if it should form, would inhibit methyl iodide removal. Second, charcoal will burn if heated above the ignition temperature in the presence of oxygen. The following heat transfer mechanisms must be considered.

- heat of adsorption of water vapor
- heat of adsorption of iodine and other trace gases
- heat transfer to the flowing gas
- decay heat of adsorbed fission products.

The CSE tests can be expected to provide information on the first three mechanisms. The last mechanism listed, fission product decay heat, cannot be duplicated in the CSE tests because the radioactive material inventories are low.

EXPERIMENTAL FACILITIES AND PROCEDURECSE CONTAINMENT VESSEL

The CSE experimental equipment comprises a model containment vessel with provision for generating fission product simulants and measuring their behavior under accident conditions. The components of this system are described in detail by Linderoth⁽¹⁶⁾. The containment vessel as arranged for the air cleaning tests is shown in Figure 2.

The containment vessel is composed of three interconnected vessels. An outer vessel, called the main containment vessel, 30,680 ft³, an inner vessel, called the drywell, 2286 ft³, and a vessel composing two-thirds of the annular space between the drywell and the main containment vessel, called the wetwell, 4207 ft³. The main containment vessel is 25 ft in diameter, and 66.7ft in overall height. All interior surfaces are coated with a modified phenolic paint, Phenoline 302*.

The top of the wetwell forms a solid deck which effectively separates the contained gases into what is termed the "main room" above the deck and the lower rooms below the deck. For the filter loop experiments discussed here, the lid of the 11-ft diameter drywell was raised so that its volume was common to the main room. The combined volume of this "main room" is 21,005 ft³. One-third of the annular space between the drywell and main containment vessel is a small access area called the "middle room", 2089 ft³ in volume. Below the middle room, drywell, and wetwell is a third space called the "lower room", 3384 ft³ in volume. The wetwell volumes were sealed off and not exposed to steam or fission product simulants in these experiments. Two 4-ft diameter holes in the deck connect the main room to the middle room; one 4 ft diameter opening connects the middle room to the lower room.

Steam condensate accumulates in two places: the drywell pool and the main vessel pool located in the lower room. These pools were stirred and sampled.

*Manufactured by the Carboline Co., St. Louis, Missouri.

FIGURE 2. Equipment Arrangement Used in CSE Filter Loop Tests

A line from the plant steam boiler, terminated near the bottom of the drywell, provides steam input to heat the vessel and its atmosphere.

Important physical data which apply to all filter loop tests are listed in Table 1.

TABLE 1. Physical Conditions Common To
All Filter Loop Experiments

Volume above deck including Drywell	21,005 ft ³	595 m ³
Surface area above deck including Drywell	6,140 ft ²	569 m ²
Surface area/volume	0.293 ft ⁻¹	0.958 m ⁻¹
Cross section area, main vessel	490 ft ²	45.5 m ²
Cross section area, drywell	95 ft ²	8.8 m ²
Volume, middle room	2,089 ft ³	59 m ³
Surface area, middle room	1,363 ft ²	127 m ²
Volume, lower room	3,384 ft ³	96 m ³
Surface area, lower room	2,057 ft ²	191 m ²
Total volume of all rooms	26,477 ft ³	751 m ³
Total surface area, all rooms	9,560 ft ²	888 m ²
Loop intake centerline elevation	1.3 ft	0.4 m
Loop stack discharge elevation	30 ft	9.4 m
Thermal Insulation	All exterior surfaces covered with 1 in. fiberglass insulation ^(a)	

(a) $k = 0.027 \text{ Btu}/(\text{hr}) (\text{ft}^2) (^\circ\text{F}/\text{ft})$ at 200 $^\circ\text{F}$,

Type PF-615, Owens-Corning Fiberglass Corp.

FILTER LOOP

The filter loop was made of modular units. Each component was located within a 3 ft diameter stainless steel cylindrical shell. The steel cylinders were connected together by flanges. The loop was instrumented to provide measurements of air flow rate, temperature, and fission product removal efficiency. The instrumentation arrangement used in the loop as assembled for Run A-15 is shown in Figure 3.

FIGURE 3. Schematic Diagram of Filter Loop Instrumentation

Pressure difference across each component and across the entire loop was measured to show changes in flow resistance with time. Loop flow was measured using both a calibrated orifice and a pitot tube installed in the discharge duct. A small flow of compressed air was used to ensure that no water condensed in the sensor lines. Absolute pressure was measured to permit evaluation of the thermodynamic state of the contained gas.

Temperature was measured upstream and downstream from each of the loop components. The temperature and flow rate of cooling water fed to the condenser was measured to evaluate the heat removal.

Liquid sampling stations were installed downstream from each component of the loop to ensure that all liquid effluents were accounted for.

Gas samples were withdrawn downstream from each of the loop components. Each sample collector was a Maypack sampling train. Six Maypack gas samples were installed at each location to permit sample withdrawal at 5 times. The remaining sampler was used as a blank to help assess background contamination. A view of the interstage samplers is shown in Figure 4. The samples were all withdrawn from near the center of the loop. One of the pleated charcoal beds is visible behind the sample probes.

The loop components were selected to be typical of those used in power reactor containment systems. Existing and planned reactors include some or all of the following components.

- heat exchanger
- moisture separator
- prefilter
- high efficiency particulate air filter (HEPA filter)
- activated charcoal beds

The CSE loop used nominal 2 ft x 2 ft commercially available components, operating at the design flow of 1000 cfm. At 1000 cfm, the air in the main room of the CSE vessel is turned over about 2.8 times per hour. This is within the range used in PWR containment vessels.

The heat exchanger used in the CSE tests was constructed with finned copper tubes. The fins were 0.75 inches high, with a 0.25 inch spacing

PNL 0692689-4

FIGURE 4. View of a Loop Module Showing Interstage Samplers and Charcoal Filter

along a 1 inch tube. Six tubes made three passes per unit, and two of these units were arranged in series. The face area was 4.1 ft². A concurrent flow of water was adjusted to provide a temperature drop of 2-3 °F in the flowing gas stream. Under typical conditions, heat removal was 3000 Btu/min.

The moisture separator was made from stainless steel and Teflon fibers*. It was 2 in. thick. For 1000 cfm, an 18 in. diameter pad was installed horizontally so that airflow was upward. A 2 ft square pad was installed vertically for the higher airflow test (1800 cfm). Water draining from the moisture separator was sampled.

A prefilter**, designed to protect the HEPA filter from mechanical damage and needless loadings of lint and dust, was installed downstream from the moisture separator. This filter was 2 ft x 2 ft, and made from expanded aluminum sheet. It was 2 in. in thickness.

The high efficiency particle filter was purchased from specifications given by Burchsted and Fuller⁽⁸⁾. This filter[†] had a water-proof glass fiber medium, a cadmium plated steel frame, aluminum separators, and neoprene bond and gaskets. All filters used were tested for pressure drop and particle retention by the USAEC Western Quality Assurance Station after arrival on site.

Pleated charcoal beds were purchased from specifications similar to those listed by Burchsted and Fuller⁽⁸⁾. The units obtained^{††} were loaded with 50 lb of 8-16 Tyler mesh charcoal. The charcoal was impregnated with 5% iodine.

The loop as assembled within the containment vessel just prior to a test is shown in Figure 5. Figure 6 shows the loop as seen through the access door on the containment vessel.

* Type 321-SR Demister, Otto H. York Company, New York, N. Y.

** Type BAA-45, American Air Filter Corp., Louisville, Ky.

† Type A24J6R2, American Air Filter Corp., Louisville, Ky.

†† Type FE, with BC-727 charcoal, Barneby Cheney Company, Columbus, Ohio.

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FIGURE 5. Air Cleaning Loop Assembled in CSE Vessel

BNWL-1587

PNL 0694495-5

FIGURE 6. Air Cleaning Loop in CSE Vessel

GAS PHASE SAMPLING

The airborne concentration of iodine, cesium, and uranium was measured using Maypack samplers. Twelve individual Maypack samplers were clustered together and placed at selected positions in the containment vessel. Eleven of the twelve samplers within each cluster were used to obtain samples at 11 discrete time periods. The remaining sampler was retained as a blank to assess background contamination levels. The 10 cluster locations used in the filter loop tests are shown in Figure 7.

In addition to the cluster samples, "thief" samples were obtained by inserting individual Maypack samplers into the vessel atmosphere through an airlock. These thief samples provided independent data for conditions where background contamination was very low.

The Maypack sampler is designed to characterize airborne iodine according to its chemical identity. It is composed of several adsorbents in series, as depicted in Figure 8. Briefly stated, the iodine distributes within the sampler as follows. Particles are captured by the inlet filter. Elemental iodine deposits on the silver components which follow the particulate filter. Heavy organic iodides, and perhaps HOI, deposit on the charcoal impregnated filter paper. Methyl iodide is retained within the charcoal granule bed. This idealized deposition sequence is not fully achieved in practice. A small part of elemental iodine is retained by the inlet filter. Also, a small fraction of the methyl iodide is retained by the adsorbents upstream from the charcoal beds. In addition, the disposition of HOI within the Maypack has not been studied. Although the CSE Maypack is not a perfect sampling tool, it has been highly useful in the CSE program. McCormack⁽¹⁷⁾ has given a detailed analysis of the design rationale and performance of this sampler.

Airborne particles were collected with a cascade impactor which had been preheated and then inserted into the vessel atmosphere. As Postma⁽⁴⁾ has noted, the process of sampling by this technique may cause droplet evaporation and thereby alter the particle size.

FIGURE 7. Location of Maypack Clusters in CSE Filter Loop Tests

Neg. No. 702507-1

FIGURE 8. Schematic Diagram of a CSE Maypack Showing Filter and Adsorber Arrangement

LIQUID PHASE SAMPLING

Sample delivery lines were installed to allow periodic sampling of the drywell sump, the main vessel sump, wall condensate, condensate formed within the loop heat exchanger, and the liquid removed by the mist eliminator. All liquid, except that in the drywell sump, eventually was combined in the main vessel sump. Condensed moisture from between loop sections was sampled during loop disassembly after completion of a test run.

GENERATION OF FISSION PRODUCT SIMULANTS

Coleman⁽¹⁸⁾ 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⁽¹⁸⁾, and when release was desired, the flask containing the iodine was heated electrically and air swept through the flask. This carried the elemental iodine through the hot zone of the UO_2 melting furnace. Some particulate associated iodine and organic iodides were always produced. Methyl iodide was in addition purposely added through a separate line not connected to the UO_2 furnace.

Stable cesium was equilibrated with ^{137}Cs as CsCO_3 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 in the drywell served as a compressor for injecting the volatilized simulant into the pressurized vessel. The injection line was a nominal 2 in. stainless steel pipe, electrically heated to 150°C .

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)⁽¹⁹⁾. Roberts, et. al.⁽²⁰⁾ at Oak Ridge National Laboratory completed an extended study of the suitability of the CSE simulant with the conclusion that the use of this simulant is warranted.

Two types of aerosol releases were used: a short term "puff" release, and a longer "linear" release. The puff release simplifies the fission product generation equipment and the mathematical models of removal. The longer linear release provides higher aerosol concentrations of cesium and uranium to be attained and permits evaluation of the source term in the removal model.

Iodine concentrations, based on the released iodine, of 160 mg/m^3 were used in all filter loop tests. Maximum airborne concentrations were about 100 mg/m^3 . The release concentration corresponds closely to those predicted for hypothetical "design basis accidents" in pressurized water reactors. Methyl iodide equal in mass to about 5% of the total iodine was released. Cesium and uranium aerosol concentrations were appreciably lower than predicted for the DBA.

EXPERIMENTAL TEST SEQUENCE

The work sequence for all filter loop tests was substantially as follows.

1. The loop was loaded with new components (Demister was re-used).
2. The loop was tested for leaks and components were tested for penetration of methyl iodide and DOP aerosol.
3. Maypack samplers were installed in the containment vessel.
4. The containment vessel was leak tested.
5. The containment vessel was brought to operating temperature with boiler house steam.
6. Fission product simulants were released.
7. Sampling of gas and liquids was begun.
8. Filter loop blower was turned on.
9. The test run was carried out as per run plan.
10. Steam was shut off and vessel allowed to cool.
11. Loop flow was stopped.
12. Loop components tested for DOP aerosol penetration.
13. Gas and liquid samples within containment vessel were recovered.
14. Loop was disassembled and components removed for sampling.
15. Containment vessel and loop housing was decontaminated by additional steaming.

In four of the five tests, the loop was started 30 to 40 minutes after completion of the fission product release. This delay allowed measurement of airborne removal due to surface deposition. This is essential for assessing the removal by the loop alone. In Run A-16, an extended linear release was used. This prevented measurement of the rate of fission product removal by natural processes.

The test runs typically lasted for two days--one day at steady temperature and pressure, and one day of decreasing temperature and pressure. The loop flow noted was kept constant for the whole run by manually adjusting the flow control damper.

EXPERIMENTAL RESULTSNUMERICAL VALUES OF TEST PARAMETERS

The five filter loop tests completed in CSE covered a range of conditions and component arrangements. These are described in Table 2.

TABLE 2. Summary of Air Cleaning Run Conditions

	CSE Run Number				
	A-13	A-14	A-15	A-16	A-17
Date	8/19/69	10/14/69	11/11/69	1/20/70	3/10/70
Temp. °F	96	250	250	245	246
Press., psia	14.5	48	48	48	48
Components in order of flow	Prefilter HEPA Charcoal Beds (3)	Heat Exchanger Demister Prefilter HEPA Charcoal Beds (3)	Prefilter HEPA Charcoal Beds (3)	Heat Exchanger Demister Prefilter HEPA Charcoal Beds (3)	Heat Exchanger Demister HEPA Charcoal Beds (3)
Flow Rate, cfm	1,000	1,000	1,000	1,000	1,800
Turnover/hr (main room)	2.8	2.9	2.8	2.8	5.2
Heat Exchange ΔT , °F	None	2	None	2	1
Aerosol Release, min	10	10	10	120	10
Initial Main Room Conc., mg/m ³					
I	105	100	100	11 ^(a)	105
CH ₃ I	5	5	5,5 ^(b)	2.5 ^(a)	6
Cs	7	3	2.6	9 ^(a)	2.5
U-Zr	4	1.6	1.6	0.1 ^(a)	1.5

(a) Max. Conc. reached. Loop operated during release.

(b) Two methyl iodide releases were made.

REMOVAL OF AIRBORNE FISSION PRODUCT SIMULANTS

The purpose of a filter loop system is to remove fission products which might become airborne as a result of hypothetical accidents. Thus the most important measurements made in the CSE experiments involved the rate of decrease in gas phase concentration of the several fission product forms as a result of loop operation. In this section of the report, the observed rates at which airborne fission products were removed will be summarized. The measured removal rates will be compared to theory in the discussion section of this report.

Iodine

Airborne iodine was assayed by Maypack samples withdrawn at 8 locations in the main room of the containment vessel, and from samplers suspended in the other two connected containment volumes. A typical time-concentration history is presented in Figure 9. These data are for Run A-17, which used a loop flow rate of 1800 cfm. Several features of this plot are important and typical of other runs. These will be briefly noted.

The airborne concentration attains a maximum at the end of the fission product generation period. The concentration of elemental iodine then decreases due to wall deposition, prior to loop operation. Methyl iodide is removed only very slowly by wall deposition, as expected.^(3,4,5) When the loop fan is turned on, both elemental iodine and methyl iodide are rapidly removed. Elemental iodine is removed more rapidly than methyl iodide due to wall deposition and due to the fact that the removal efficiency for methyl iodide is appreciably less than 100%. After the airborne concentration is reduced to about 1% of the initial concentration, the removal rate decreases. This decrease in removal rate is presumably due to a source term caused by desorption of iodine from containment surfaces.

The removal of airborne iodine is summarized in Table 3.

FIGURE 9. Iodine Concentrations in the Main Room Gas Space in CSE Run A-17

TABLE 3. Early Removal Half-Times for Airborne Iodine In Filter Loop Experiments

Removal Half-Time, Minutes																
CSE Run No.	A-13			A-14			A-15			A-16			A-17			
Loop Flow Rate, cfm	1000			1000			1000			1000			1820			
	N	N+L	L	N	N+L	L	N	N+L	L	N	N+L	L	N	N+L	L	
Elemental Iodine	26	7.3	10.1	16	4.9	7.1	27	6.0	7.7	15	7.3	14.3	13	3.8	5.4	
Particulate I	50	11.0	14.1	27	9.0	13.5	27	10.3	16.7	36	15.0	25.7	25	6.3	8.7	
AC Paper Form	50	16.0	21.0	39 ^(a)	9.0	11.8	58	10.0	12.5	39	13.7	14.2	20	7.7	12.5	
Methyl Iodide	long	15.0	15	long	14.3	14.3 ^(c)	long	14.7	14.7 ^(c)	long	16.0	16.0	long	11.0	11 ^(b)	
								19.0	19.0 ^(b)							

N = Natural, L = Loop Only

(a) Natural t_{1/2} not measured this run. Value given is avg. of 8 CSE runs under similar conditions.

(b) Bed equilibrated with 100% relative humidity air.

(c) "Dry" bed.

A measure of the degree to which iodine was removed may be obtained by comparing the iodine airborne at the end of the test to that initially released. The decontamination factor measured for each of the filter loop tests is given in Table 4.

TABLE 4. Iodine Decontamination Factors Achieved in Filter Loop Tests

Run No.	Loop Flow Duration, min	Overall Decontamination Factor ^(a)	
		Total Inorganic Iodine ^(b)	Methyl Iodide
A-13	2730	5.3×10^4	1.5×10^3
A-14	1000	1.8×10^3	2.7×10^3
A-15 ^(c)	2560	1.1×10^4	1.8×10^3
A-16	2730	1.0×10^5	1.1×10^4
A-17	2700	3.6×10^4	4.5×10^4

(a) Overall decontamination factor = $\frac{\text{Mass of Iodine Released to Containment}}{\text{Mass of Iodine Airborne at Run Termination}}$

(b) Total inorganic iodine defined as total iodine minus methyl iodide.

(c) In Run A-15, methyl iodide was released at t=0 and at t=1350 min.

The decontamination factors achieved in all runs are large, showing that gas cleanup by the loop is effective. Methyl iodide was removed to a slightly lesser extent than total inorganic iodine in 3 of the 5 runs. Although the observed decontamination factors are large, they are much smaller than would be predicted if the initial removal rate had continued for the whole run duration. The numerical value of the two hour dose reduction factor depends only on the initial removal rate. For times longer than about 24 hours, the slower removal rate observed at low concentrations would reduce the dose reduction factor calculated on the basis of the initial exponential removal.

Gas phase concentrations of iodine in the main room of the CSE vessel are shown in Figures A-1 through A-5 in the Appendix to this report.

Samples withdrawn from the connected containment volumes permitted the concentration to be determined for these rooms. Typical results are shown in Figure 10, where the iodine concentration in the middle room in Run A-14 is presented. The airborne concentrations increase rapidly following fission product injection into the main room, and attain a maximum at about 120 minutes. They then fall off with the main room concentration. These results are typical of those obtained in all CSE experiments, hence we will not list the data in detail.

Cesium and Uranium Aerosols

Cesium and uranium aerosols were removed by the loop with essentially a 100% efficiency. These materials, as aerosol particles, were also removed by wall deposition and gravitational settling. A typical airborne depletion curve is shown in Figure 11. Both cesium and uranium are removed at about the same rate after the loop is turned on. This rate is similar to the removal rate for iodine. At long times, the removal rates decrease. This net decrease in removal rate is explained in terms of small sources. Aerosol particle could be formed, for example, by breaking of bubbles in the sumps.

The observed removal rates for cesium and uranium aerosols are summarized in Table 5.

FIGURE 10. Concentration of Airborne Fission Products in the Middle Room of the CSE Vessel

FIGURE 11. Cesium and Uranium Concentrations in the Main Room Gas Space in CSE Run A-17

TABLE 5. Early Removal Half-Times for Cesium
and Uranium in Filter Loop Experiments

CSE Run No.	Removal Half-Time, Minutes														
	A-13			A-14			A-15			A-16			A-17		
	1000			1000			1000			1000			1820		
Loop Flow Rate, cfm	N	N+L	L	N	N+L	L	N	N+L	L	N	N+L	L	N	N+L	L
Cesium	62	11.0	13.7	42 ^(a)	12.3	17.4	70	13.0	16.0	47	9.3	11.5	33	7.7	10.0
Uranium	80	14.0	17.0	85	13.0	15.4	70	12.0	14.5	31	16.0	33.0	41	8.3	10.4

N = Natural, L = Loop Only

(a) Natural t_{1/2} not measured this run. Value given is avg. of 8 CSE runs under similar conditions.

The long term removal of cesium particles is summarized in Table 6.

TABLE 6. Decontamination Factors
Achieved in Filter Loop Tests

Run No.	Loop Flow Duration, min	Overall Decontamination Factor ^(a)
A-13	2730	1.3×10^5
A-14	1000	1.5×10^5
A-15	2560	$> 2.7 \times 10^4$
A-16	2730	2.0×10^5
A-17	2700	1.6×10^5

(a) Overall decontamination factor = $\frac{\text{mass released}}{\text{mass airborne at run termination}}$

These results show that aerosol particles are removed to a greater extent than gaseous iodine. This is reasonable because gaseous fission products can desorb from liquid solution, whereas particles can be resuspended only by mechanical disintegration processes. The lower analytical sensitivity for uranium prevented reliable detection of the fraction airborne at the termination of the tests, hence results for uranium aerosols could not be included in Table 6. For the earlier part of each test, cesium and uranium were removed at comparable rates.

The time-concentration histories for cesium and uranium aerosols are shown in Figures A-6 through A-10 in the Appendix to this report.

FISSION PRODUCT DISTRIBUTION WITHIN CONTAINMENT COMPONENTS

Concentration in Liquid Condensate

Liquid condensate formed in three places: the drywell sump, the containment vessel sump, and the heat exchanger. The drywell sump collected liquid formed by condensation on the vertical wall of the drywell vessel and liquid drops which fell from the surfaces above the drywell opening. The main vessel sump accumulated condensate formed on the outer walls of the vessel. The third place condensate formed was within the heat exchanger which was incorporated as a loop component in several experiments. The condensate from the heat exchanger was allowed to drain into the main vessel sump.

Typical condensate concentrations for iodine are shown in Figure 12. These data show that the concentration in the heat exchanger condensate closely parallels the airborne iodine concentration. The drywell concentration peaks soon after iodine injection at a level about ten times higher than main vessel sump. This higher concentration results from absorption of iodine close to the injection point which is located within the drywell. Also, the volume of liquid which accumulates within the drywell is small compared to the liquid in the main sump. The concentration in both sumps decreases with time due to dilution. Similar behavior was observed for cesium. The liquid concentrations for the five filter loop tests are shown in detail in Figures A-11 through A-20 in the Appendix.

Liquid Volumes

The volumes of liquid were measured as a function of time. In the experiments in which the heat exchanger was used, the condensate flow rate was measured. Results of these measurements are shown in Figure 13 for Run A-17. The main vessel sump accumulates liquid at a constant rate until the steam is turned off. Since the condensate formed on the outer walls of the vessel drains to the main sump, the main sump volume accumulates most of the condensate. The heat exchanger condensate

FIGURE 12. Iodine Concentration in Liquid Sumps
in CSE Run A-14

FIGURE 13. Water Volumes Accumulated in CSE Run A-17

flow is essentially constant after the fan was turned on, and up until the time the steam flow was turned off.

At the time the steam flow was terminated, the vessel contained about 8000 liters of water. This volume represents about 1% of the volume of the gas space. This amount of liquid is relatively less than for PWR plants which use containment spray as well as a filter loop. One would expect a liquid/gas volume of about 0.03 for full scale PWR nuclear plants.

The liquid volumes which accumulated in the four filter loop experiments are shown in detail in Figures A-21 through A-24 in the Appendix. Data for Run A-13 are not included because no steam was added. For the latter run, the liquid volumes were established before the run at 330 ℓ in the main sump and 400 ℓ in the drywell.

Retention by Loop Components

After each run, the loop was disassembled and the components analyzed to assess the quantity of fission products retained. The moisture separator was decontaminated and the washings analyzed. For the other components, samples were taken, and the total fission product load estimated from the sample analysis. The amounts of iodine on each loop component for the five tests are listed in Table 7.

TABLE 7.- Post Run Loop Iodine Inventory

	<u>Iodine Retained in the CSE Filter Loop, g</u>				
	<u>Run No.</u> <u>13</u>	<u>Run No.</u> <u>14(a)</u>	<u>Run No.</u> <u>15</u>	<u>Run No.</u> <u>16</u>	<u>Run No.</u> <u>17</u>
Heat Exchanger Liquid	----	0.036	----	2.82	0.90
Moisture Separator Washings	----	0.43	----	9.32	0.17
Prefilter	0.052	0.019	0.0053	0.40	----
HEPA Filter	2.39	2.90	5.53	8.62	6.07
1st Char Bed 1/2	18.5	5.52	9.81	4.88	9.3
2/2	2.46	2.47	4.07	1.87	2.9
2nd Char Bed 1/2	1.61	0.086	2.00	1.23	1.3
2/2	1.12	0.036	1.56	0.76	0.93
3rd Char Bed 1/2	0.61	1.09	1.38	0.38	0.60
2/2	0.34	0.546	0.99	0.23	0.47
Total, g	27.08	13.13	25.36	30.50	22.6

(a) Corrected for Post Run CH_3I Test.

As one would expect, most of the iodine was retained on the first charcoal bed, except for Run A-16. In Run A-16 the mass of cesium released was large compared to the iodine mass and as a result, the cesium reacted with much of the airborne iodine to form an aerosol of CsI. For this reason more than 50% of the iodine was retained by the filter components of the loop.

The retention of cesium by the loop components is given in Table 8. Interestingly, an appreciable fraction of the cesium was trapped by the

TABLE 8. Post-Run Loop Cesium Inventory

<u>Component</u>	<u>Cesium Retained in the CSE Filter Loop, g</u>				
	<u>Run No.</u> <u>13</u>	<u>Run No.</u> <u>14</u>	<u>Run No.</u> <u>15</u>	<u>Run No.</u> <u>16</u>	<u>Run No.</u> <u>17</u>
Heat Exchanger Liquid	----	0.031	----	3.4	0.05
Moisture Separator Washings	----	0.76	----	16.0	0.50
Prefilter	0.021	0.0002	0.011	0.91	----
HEPA Filter	<u>2.88</u>	<u>0.43</u>	<u>0.735</u>	<u>10.67</u>	<u>0.41</u>
Total, g	2.90	1.22	0.746	30.98	0.96

moisture separator. This demonstrates that the aerosol particles in the containment atmosphere were larger than one micron in size. Sub-micron aerosols do not have sufficient inertia to be captured as was observed.

Fission Product Material Balances

After the Maypack samplers and loop components had been removed from the vessel following cool-down, the vessel was decontaminated by steaming. The decontamination procedure removed the contaminants which were reversibly attached to surfaces. Material balances were then made, accounting for the masses contained in liquid pools, that remaining airborne, and that within the loop. The quantity of material on surfaces was taken as the difference between the mass injected and that accounted for.

The disposition of iodine in Run A-17 is shown in Figure 14. These data show that after 30 minutes, most of the iodine is attached to wall surfaces. This occurs because a major fraction of the iodine injected mass deposits on wall surfaces by natural transport before the loop flow begins. Much of the deposited iodine was irreversibly retained by the wall. Similar behavior was observed in all five tests. The disposition of iodine in the containment vessel is given in Figures A-25 through A-29 in the Appendix.

Cesium is retained less effectively by wall surfaces than is iodine. This is shown in Figure 15 where the cesium mass distribution is given for Run A-17. Most of the cesium is retained by the loop filters for times longer than 50 minutes. The distribution of cesium within the containment vessel for the five filter loop tests is presented in Figures A-30 through A-34 in the Appendix.

The final material balance for iodine in the filter loop tests is summarized in Table 9. These results show that about half of the iodine initially deposited on the walls is removable by steaming. The mass of iodine remaining on surfaces is in agreement with that predicted from small deposition coupons exposed to the containment atmosphere.

The final cesium material balances for the filter loop tests are summarized in Table 10. Cesium is retained by wall surfaces to a lesser extent than is iodine. This is reasonable since it is present as a salt and is thus highly soluble in water.

FIGURE 14. Total Iodine Material Balance vs. Time for CSE Run A-17

FIGURE 15. Cesium Material Balance vs. Time for CSE Run A-17

TABLE 9. Iodine Material Balances

Location	Run A 13		Run A 14		Run A 15		Run A 16		Run A 17	
Aerosol Generation	Grams	% ^(a)	Grams	% ^(a)	Grams	% ^(a)	Grams	% ^(a)	Grams	% ^(a)
Starting Material	105.5	100.00	100.05	100.00	110.2	100.00	130.05	100.00	105.00	100.00
Generation Apparatus	4.19	3.99	2.216	2.125	4.537	4.11	11.97	9.20	3.581	3.41
Injection Line	7.52	7.16	2.193	2.192	1.34	1.22	8.39	6.45	2.733	2.60
Injection Line Samplers	0.51	0.49	0	0	0	0	0	0	0	0
Accumulated Mass	12.21	11.62	4.319	4.317	5.877	5.33	20.36	15.66	6.314	6.01
To Containment (By Difference)	92.84	88.38	95.731	95.683	104.323	94.67	109.69	84.34	98.686	93.99
Containment	Grams	% ^(b)	Grams	% ^(b)	Grams	% ^(b)	Grams	% ^(b)	Grams	% ^(b)
Delivered to Containment	92.84	100.00	95.731	100.00	104.323	100.00	109.69	100.00	98.686	100.00
In Liquid Pools ^(c)	10.28	11.07	17.869	18.666	14.747	14.14	39.81	36.29	17.112	17.34
Samples	0.9	0.97	0.532	0.556	0.541	0.520	0.54	0.49	0.633	0.64
Purge to Stack	0.008	0.00	0.0119	0.012	0.014	0.01	0.0004	0.0004	0.0009	0.001
Decontamination (Steaming)	30.15	32.48	10.20	10.66	5.966	5.721	21.47	19.57	6.88	6.98
On Loop	26.98	29.06	13.11	13.69	25.34	24.29	20.31	18.52	21.77	22.06
Accounted For	68.32	73.59	41.72	43.58	46.61	44.68	82.13	74.87	46.40	47.01
On Surfaces (By Difference)	24.52	26.41	54.011	56.42	57.72	55.32	27.56	25.13	52.29	52.99
On Surfaces (Deposition Data)	14.63	15.76	52.66	55.01	63.57	60.94	14.38	13.11	51.04	51.72

(a) % of starting mass

(b) % of delivered mass

(c) Includes spray solutions and steam condensate

TABLE 10. Cesium Material Balances

Location	Run A 13		Run A 14		Run A 15		Run A 16		Run A 17	
Aerosol Generation	Grams	% (a)	Grams	% (a)	Grams	% (a)	Grams	% (a)	Grams	% (a)
Starting Material	11.54	100.00	11.228	100.00	10.989	100.00	445.40	100.00	11.222	100.00
Generation Material	4.04	35.01	1.428	12.718	6.873	62.54	209.30	46.99	7.450	66.39
Injection Line	0.50	4.33	2.058	18.329	1.530	13.92	20.81	4.67	1.287	11.47
Injection Line Samplers	0.07	0.61	0	0	0	0	0	0	0	0
Accumulated Mass	4.61	39.91	3.486	31.047	8.403	76.46	230.11	51.66	8.737	77.86
To Containment (By Difference)	6.93	60.05	7.742	68.953	2.585	23.54	215.29	48.34	2.485	22.14
Containment	Grams	% (b)	Grams	% (b)	Grams	% (b)	Grams	% (b)	Grams	% (b)
Delivered to Containment	6.93	100.00	7.742	100.00	2.585	100.00	215.29	100.00	2.485	100.00
In Liquid Pools (c)	1.226	17.69	2.992	38.646	0.685	26.50	85.85	39.88	1.227	49.38
Samples	0.032	0.46	0.039	0.504	0.016	0.62	0.80	0.37	0.0146	0.59
Purge to Stack	0.00005	0.00	0.00002	0.0003	0.000004	0.00	0.002	0.0009	0.0001	0.003
Decontamination (Steaming)	1.027	14.82	1.248	16.12	0.512	19.81	40.23	18.69	0.5218	21.00
Loop Components	2.92	42.12	1.209	15.616	0.746	28.86	16.77	7.79	0.9032	36.35
Accounted For	5.205	75.11	5.488	70.886	1.959	75.78	143.65	66.72	2.667	107.31
On Surfaces (By Difference)	1.725	24.89	2.254	29.114	0.626	24.22	71.64	33.28	-0.182	-7.31
On Surfaces (Deposition Data)	0.074	1.07	0.058	0.75	0.032	1.24	0.353	0.16	0.0211	0.85

(a) % of starting mass

(b) % of delivered mass

(c) Includes spray solution and steam condensate

REMOVAL EFFICIENCIES FOR FILTER LOOP COMPONENTS

Interstage samples were taken to provide a direct measurement of removal efficiency for individual loop components. Six individual Maypack samplers were installed to withdraw samples between the several components. One of these was used as a blank. The other 5 samplers were operated at 5 discrete time step during the run. The decontamination factors measured from these samples are summarized in Table 11 for Run A-17.

TABLE 11. Measured Loop Decontamination Factors For Run A-17

Component	Decontamination Factor at Stated Time								
	35 minutes			60 minutes			80 minutes		
	Cs	I	CH ₃ I	Cs	I	CH ₃ I	Cs	I	CH ₃ I
Heat Exchanger	1.2	1.3	-	1.2	1.6	-	1.2	1.4	-
Demister	6.2	1.3	-	3.7	1.1	-	2.4	2.5	-
HEPA Filter	44	1.6	-	40	85	-	-	>13	-
1st Charcoal Bed	-	470	1.7	-	-	1.7	-	-	1.7
2nd Charcoal Bed	-	>7	-	-	-	-	-	-	-
3rd Charcoal Bed	-	-	-	-	-	-	-	-	-
Overall	27,000	>8000	12	1900	>120	10	520	>180	7.5

Aerosol particles and elemental iodine are removed with very high efficiencies early in the run. Methyl iodide is removed with lower efficiency, but still is effectively removed. The removal efficiency for cesium, elemental iodine, and methyl iodide decreases with time, though from a practical standpoint, this decrease is negligible until the concentration decreases to very low levels. The overall decontamination factor listed in Table 11 is the experimentally measured value taken from samples of the air entering and leaving the loop. The overall decontamination factor must equal to the product of the decontamination factors for the individual components. The analytical sensitivity limits prevented measurement of the decontamination factor for each loop component so it is not possible to compare the overall loop efficiency with that predicted for the components in series.

Although one would like to know the removal efficiency for each component at long times (24-48 hours), the very low airborne concentration which existed at these times effectively prevented their reliable detection. Much more sophisticated sampling and analytical techniques would be required to do this.

The measured loop component efficiencies for the five filter loop tests are summarized in Tables A-1 through A-5 in the Appendix.

PARTICLE SIZE MEASUREMENTS

Aerosol particle size measurements were made during the experiments using a round jet cascade impactor.* Use of a cascade impactor to sample saturated steam-air mixtures is less than ideal, because of thermodynamic changes which occur within the impactor. In order to avoid water condensation on the impactor stages, the impactor was preheated to about 300 °F prior to use. Warming of the saturated steam-air gas stream may be expected to cause evaporation of water associated with aerosol particles. This evaporation would cause a decrease in the particle size compared to that in the containment atmosphere. Postma⁽⁴⁾ has discussed evaporation of "wet" particles within a cascade impactor.

The particle sizes measured by the impactor are listed in Table 12. These particle diameters are similar for the ambient temperature run, A-13,

TABLE 12. Cesium Particle Size in Filter Loop Tests

<u>Run No.</u>	<u>Mass Median Aerodynamic Diameter, μm</u>	
	<u>10-20 min</u>	<u>> 100 min</u>
A-13	0.9	0.6
A-14	0.9	< 0.3
A-15	0.5	0.5
A-16	1.0	0.5
A-17	0.5	< 0.4

and the other tests made at 250 °F. For Run A-13 the impactor was not heated prior to use, hence the uniformity of the results leads to the

*Model CI-S-8, Scientific Advances Inc., Columbus, Ohio.

conclusion that use of a heated impactor did not change the indicated particle sizes. The particle sizes shown in Table 12 are in agreement with those obtained in previous CSE experiments.^(1,2,3,4)

VISUAL OBSERVATIONS OF CONTAINMENT ATMOSPHERE

The CSE vessel is equipped with a 6" diameter window and interior lights so that the containment atmosphere can be observed visually. The time sequence of observations in a typical run is briefly as follows. When steam is introduced into the cold vessel, a dense fog is formed. Mixing of the steam with the cool air initially present leads to supersaturation and condensation within the gas phase. The fog clears when the temperature approaches equilibrium. Uranium oxide aerosol is then injected into the vessel, and this results in a very slight fog, barely visible in the 25 ft diameter vessel. When cesium and iodine are injected, a very dense fog forms, reducing the visibility to a few feet. This fog clears gradually by natural processes before the loop fan is started.

Within a few minutes after the loop flow was started, the fog disappeared completely. Two factors explain this. First, iodine and particles were removed from the atmosphere which would clean the atmosphere directly. Second, the charcoal beds adsorbed significant amounts of water vapor, which caused slight superheating of the vessel atmosphere. This caused fog drops to evaporate. The vessel atmosphere remained clear for the remainder of the test period.

MECHANICAL ASPECTS OF LOOP OPERATION

Operational Tests

Several operational tests were made on the loop before and after each test to verify that the loop components were undamaged and that leakage past seals was within expected tolerance. Prior to each run, the HEPA filters were tested for penetration using a standard DOP aerosol test. This was done after assembly of the loop. Also, the charcoal beds were tested for penetration using methyl iodide. The methyl iodide pretest was of short duration and showed: (1) that direct leakage of air through the bed was within tolerance limits, and (2) that the charcoal installed

possessed a high degree of retentivity toward methyl iodide, and by implication toward elemental iodine.

The loop operated at a slight negative pressure compared to its environment so that in-leakage between stages was a potential problem. Before each run the ductwork was tested at the operating differential pressure for in-leakage. This was done by sealing the inlet and outlet openings, and evacuating the duct to the operating pressure differential. The in-leakage rate was calculated from the rate of pressure buildup within the duct.

Following each test, the DOP penetration test was applied to the HEPA filter. In one experiment, Run A-14, the methyl iodide penetration test was carried out after the run.

Results of the operational tests are summarized in Table 13. The HEPA filter was not degraded by the containment test. The charcoal beds

TABLE 13. Operational Tests on Loop and Components

	<u>Run No.</u> <u>A-13</u>	<u>Run No.</u> <u>A-14</u>	<u>Run No.</u> <u>A-15</u>	<u>Run No.</u> <u>A-16</u>	<u>Run No.</u> <u>A-17</u>
Pre Run					
Duct Leakage, % ^(a)	0.01	0.01	0.02	0.02	0.002
DOP Penetration, %	<0.03	<0.03	<0.03	0.01	<0.03
CH ₃ I Penetration, % ^(b)	<0.002	0.02	0.001	0.0003	0.03 ^(c)
Post Run					
DOP Penetration, %	<0.03	<0.03	<0.03	<0.03	<0.03
CH ₃ I Penetration, %		~0.2			

(a) As % of loop flow at estimated operating ΔP

(b) Penetration in 3 min at ambient temp and humidity, 1000 cfm

(c) Same as (b) except 1800 cfm.

were somewhat less effective for methyl iodide after the containment test, but were still within an acceptable range.

Loop Flow and Pressure Drop

The temperature and pressure of the vessel atmosphere was measured for the entire test, along with the loop flow rate. Typical results are shown in Figure 16. The atmosphere temperature and pressure were essentially constant until steam flow was halted, and then decayed over the next 24 hours. The loop flow rate was adjusted using a damper to maintain it constant. In Run A-17, the increase in pressure drop caused the loop flow to decrease with the damper completely open. This was expected because the flow rate for this run was 1800 cfm or about double the design capacity of the system.

The pressure drops across loop components for Run A-17 are shown in Figure 17. The most notable feature of the data shown is that the pressure drop across the HEPA filter increased from about 2" to 4.5" some 15 hours after the loop flow was started. For times from 15 hr to 24 hr, the pressure drop limited the loop flow rate, which fell to 1600 cfm from 1800 cfm. The HEPA filter returned to the pre-run flow resistance soon after steam injection was terminated.

Data on loop flow rate, thermodynamic conditions, and pressure drop for all 5 filter loop tests are given in Figures A-35 to A-44 in the Appendix.

Heat Removal By Finned Tubes

Although the primary objective of the filter loop tests was to assess the removal of airborne fission products, the loop instrumentation allowed the heat transfer coefficient to the finned tubes to be evaluated. The heat transfer coefficient was found to be sensitively dependent on the air/steam ratio. Results for Run A-16 are shown in Figure 18. The outside heat transfer coefficient, based on total outside area, is shown as a function of the steam/air ratio of the inlet gas. During the steady state temperature period of Run A-16, the heat exchanger removed 44,000 BTU/min, including both sensible and latent heat.

FIGURE 16. Thermodynamic Conditions and Loop Flow for CSE Run A-17

FIGURE 17. Pressure Drop Across Loop Components in CSE Run A-17

FIGURE 18. Heat Transfer Coefficient to Finned Tubes
For CSE Run A-16

Iodine and Steam Adsorption by Charcoal Beds

After each experiment the charcoal beds were reweighed to measure moisture pickup and charcoal samples were analyzed for total iodine, to look for any losses of iodine impregnant. Table 14 gives the amount of stable iodine found.

TABLE 14. Stable Iodine on Charcoal Beds After Use, Wt %

<u>Bed and Location</u>		<u>Run No. A-13</u>	<u>Run No. A-14</u>	<u>Run No. A-15</u>	<u>Run No. A-16</u>	<u>Run No. A-17</u>
1st	1st half	4.1	4.6	4.0	3.7	3.4
	2nd half	5.1	6.0	3.2	4.0	3.7
2nd	1st half	4.1	4.3	3.7	3.5	3.8
	2nd half	3.9	4.3	3.8	3.9	4.0
3rd	1st half	5.3	4.1	3.8	3.8	3.7
	2nd half	4.4	4.4	3.8	4.1	4.4
Average		4.5	4.9	3.6	3.8	4.0

The average level of impregnation does not change significantly with the run conditions. The average iodine content on a dry basis would be 5.4%, which compares well with the vendors specifications of 5% to 5.5%. This shows that little, if any, loss of iodine occurred during the containment tests.

Moisture gain by the beds at the time of recovery is given in Table 15, where the weight gain is expressed as a percentage of the net weight of dry charcoal in each bed.

TABLE 15. Moisture Pickup by Charcoal at End of Run,^(a) wt%

<u>Bed</u>	<u>Run No.</u> <u>A-13</u>	<u>Run No.</u> <u>A-14^(b)</u>	<u>Run No.</u> <u>A-15</u>	<u>Run No.</u> <u>A-16</u>	<u>Run No.</u> <u>A-17</u>	<u>Aver-</u> <u>age</u>
1	37	34	32 ^(b)	21	35	32
2	37	30	39	33	38	35
3	39	31	37	34	39	36

(a) Based on vendors net wt. when filled

(b) Net wt. not recorded. Assumed 50.5 lb.

The average 34% weight gain measured at the end of the run is lower than would be expected for charcoal saturated with moisture at 100% relative humidity. A charcoal with 1400 m²/g area would be expected to pick up about 50 wt% moisture⁽¹⁵⁾ if at equilibrium. These beds have had many hours to re-equilibrate with the lower humidity air used to purge the vessel before sample recovery. This likely accounts for the lower than expected moisture content.

Mechanical Reliability of Loop

In a design basis accident, the filter loop is required to run for weeks in the humid hot containment atmosphere. Each CSE run involved about 150 hours of operation including the vessel warmup and steam decontamination after each run. The total exposure to elevated temperature and humidity amounted to about 30 days. The motor, fan, and sample inlet valves were exposed to the containment atmosphere for this total time. The filter loop components, however, were changed after each experiment.

The motor was a 10 horsepower, encapsulated winding, totally-enclosed, fan-cooled unit with class H insulation. It was completely trouble free. All bearings on the fan and motor were regreased with a high temperature lithium-silicone grease before the series of loop tests. Bearing performance was adequate. except that grease washed out of one fan bearing, possibly as a result of a damaged seal. No serious deterioration of other

loop components was noted. The galvanized protective screen at the face of the HEPA filter showed loss of zinc due to oxidation and flaking.

The loop sections were assembled with asbestos gaskets. Components were installed using neoprene sponge gaskets and no problems were experienced with either type.

The fan was driven by three V-belts. The flow in Run A-14 was stopped early because of belt slippage. Type L light duty belts had been unknowingly used and these stretched beyond use in the steam air environment. All subsequent runs used premium grade B section V-belts without problems.

EFFECT OF HUMIDITY ON METHYL IODIDE REMOVAL BY CHARCOAL BEDS

The temperature of air exiting from the charcoal beds was measured during the runs. Significant heating of the air was recorded when the beds were adsorbing moisture. This heating is due to heat of adsorption as the water vapor condenses in the charcoal pores. Temperature increases up to 35 °F were recorded. Figure 19 is typical, and similar heating was measured in all runs. The net effect is that the air flowing through the charcoal bed is dehumidified, both by heating and by transfer of moisture to the charcoal. The relative humidity due to heating only is shown in Figure 18. If allowance is made for moisture removal, the humidity would be a few percent lower. This is a transient effect. After a few hours, the bed is saturated and the exit air is also saturated.

It is well documented that the ability of charcoal to remove methyl iodide from air is dependent on the humidity of the air.^(12,13,14,15,21) To measure this humidity effect, Runs A-13 and A-14 were made with "dry" (no previous exposure to steam) charcoal and in Runs A-16 and A-17 the charcoal beds were pre-equilibrated with the saturated steam/air atmosphere. Run A-15 involved two methyl iodide releases, one before saturation of the beds and one 22 hr later. Figure 20 gives the main room methyl iodide concentrations measured in Run A-15. The dry charcoal removed the methyl iodide at nearly 100% efficiency until the concentration was reduced to about 0.002 of that at the start. The saturated charcoal beds following

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FIGURE 19. Heating Due to Moisture Adsorption on Charcoal Beds in CSE Run A-15

FIGURE 20. Methyl Iodide Removal by Impregnated Charcoal in CSE Run A-15

the second release, removed methyl iodide at a reduced efficiency of about 78%. The final concentration of methyl was about the same as achieved by the "dry" beds. This is a graphic demonstration of the behavior of a recirculating system, wherein good cleanup is accomplished even though operating at less than 100% efficiency.

The primary removal of methyl iodide was by isotopic dilution of the radioactive iodine with the stable iodine on the impregnated charcoal. This was borne out by analyses of the airborne methyl iodide concentration using a gas chromatograph. The total concentration of airborne methyl iodide (radioactive and non-radioactive) decreased only about a factor of ten whereas the radioactive methyl iodide was reduced in concentration by more than three orders of magnitude.

DEPOSITION OF FISSION PRODUCTS ON EXPOSED SURFACES

Deposition coupons were attached to the Maypack sample clusters housing to evaluate the sorption of fission products by several surfaces. The coupons were 2-1/2 inches in diameter, 1/16 inch thick, and had a 17/32 center hole. Both sides were exposed to the atmosphere. Thus heat would transfer to the specimens only during temperature transients.

Results for iodine are summarized in Table 16. It is obvious that stainless steel picks up much less iodine than the other surfaces.

Results for cesium are presented in Table 17. Cesium retention is essentially the same for all of the surfaces tested.

TABLE 16. Iodine Deposition on Various Noncondensing Surfaces

Surface Material ^(a)	Deposition at end of Experiment $\mu\text{G}/\text{cm}^2$ and $\pm 1\sigma$ (%)				
	Run A-13	Run A-14	Run A-15	Run A-16	Run A-17
Silver ^(b)	31.9 ±29%	25.2 ±39%	28.4 ±52%	6.60 ±20%	27.1 ±30%
Carbon Steel ^(b)	31.6 ±37	23.0 ±54	30.3 ±49	5.83 ±23.5	0.152 ±100%
Stainless 304 ^(b) (ASTM A240-63)	0.588 ±80%	0.317 ±23%	0.184 ±53%	0.690 ±43%	32.3 ±36%
Phenoline 302 ^(b,c)	6.31 ±54%	22.7 ±22%	27.4 ±35.6%	6.20 ±17%	22.0 ±37%
Amercoat 66 ^(b,d)	2.85 ±52%	26.4 ±32%	28.1 ±49%	7.05 ±22%	29.0 ±36%

(a) 2 1/2 in. diameter, 1/16 in. thick, 17/32 in. center hole.
Total surface = 64 cm².

(b) Average of 11 locations in main room.

(c) Coating manufactured by Carboline Company, St. Louis, Missouri.
Average of 22 locations.

(d) Coating manufactured by Amercoat Corporation, South Gate,
California.

TABLE 17. Cesium Deposition on Various Noncondensing Surfaces

<u>Surface Material</u> ^(a)	Deposition at end of Experiment <u>$\mu\text{g}/\text{cm}^2$ and $\pm 1\sigma$ (%)</u>				
	<u>Run A-13</u>	<u>Run A-13</u>	<u>Run A-15</u>	<u>Run A-16</u>	<u>Run A-17</u>
Silver ^(b)	0.054 ±52%	0.030 ±24%	0.0045 ±88%	0.38 ±69%	0.022 ±58%
Carbon Steel ^(b)	0.049 ±40%	0.047 ±58%	0.023 ±82%	0.027 ±61%	0.012 ±61%
Stainless 304 ^(b) (ASTM A240-63)	0.027 ±37%	0.033 ±60%	0.062 ±56%	0.024 ±175%	0.022 ±50%
Phenoline 302 ^(b,c)	0.032 ±37%	0.025 ±38%	0.014 ±37%	0.152 ±24%	0.009 ±31%
Amercoat 66 ^(b,d)	0.031 ±18%	0.021 ±51%	0.007 ±40%	0.206 ±67%	0.013 ±29%

(a) 2 1/2 in. diameter, 1/16 in. thick, 17/32 in. center hole.
Total surface = 64 cm².

(b) Average of 11 locations in main room.

(c) Coating manufactured by Carboline Company, St. Louis, Missouri.
Average 22 locations.

(c) Coating manufactured by Amercoat Corporation, South Gate,
California.

DISCUSSION OF RESULTS

MIXING OF GAS SPACE

All practical models of fission product transport in containment vessels are based on the assumption that the bulk of the volume within a single room is well mixed. All prior CSE experiments have shown this to be a good assumption, and it was expected that gas phase mixing in the air cleaning runs would be complete. Mixing can be assessed by noting the standard deviation of the measured concentration levels, such as shown in Figure 21. Early in Run A-14, standard deviations are of the order of 20% of the mean. This degree of variation is about equal to the variation expected as a result of differences in sampled volumes and component analysis. Hence, it is concluded that the gas phase of the main room is well mixed.

At longer times, the concentrations are much lower, and the standard deviations are relatively larger. This larger standard deviation is the result of background contamination which becomes important at the lower concentrations.

Mixing between adjacent rooms is relatively slow, as shown by the gas phase concentration data shown in Figure 10. The middle room concentration remains below the main room concentration for about 2 hours. These results are similar to earlier spray and natural transport tests in CSE as expected. In all CSE tests, inter-room mixing has been due solely to natural convection.

These observations show that the "well-mixed gas space" assumption would be valid within a single room, but that separate rooms can have different concentrations.

COMPARISON OF MEASURED REMOVAL RATE WITH THEORY

A simple model of removal by an air cleaning loop showed that the removal rate was equal to the sum of that due to the loop, and that due to surface deposition:

FIGURE 21. Iodine Concentration in the Main Room Gas Space in CSE Run A-14

$$\underbrace{-\frac{dC_g}{dt}}_{\text{overall}} = \underbrace{\frac{FE}{V}C_g}_{\text{loop}} + \underbrace{k_g A(C_g - C_{gi})}_{\text{natural deposition}} \quad (5)$$

The natural, or surface deposition removal rate, $k_g A(C_g - C_{gi})$ was evaluated by measuring $\frac{dC_g}{dt}$ with the loop fan off ($F = 0$). Such a measurement was made at the beginning of four of the five filter loop experiments. From the total observed removal rate, one can obtain that due to the loop:

$$\text{loop removal rate} = \text{overall rate} - \text{natural deposition rate} \quad (10)$$

In writing Eq (10) it has been assumed that the surface deposition rate is not influenced by the operation of the fan. In reality, the fan operation will agitate the gas phase, causing an increase in the mass transfer coefficient. Initially the loop also dries the air which would reduce the particle size, and potentially reduce the deposition rate of aerosols. Thus Eq (10) will overpredict the loop removal rate by a small increment.

Predicted removal rates are compared to measured rates in Table 18. The removal rates are presented in terms of removal half-times measured early in the run. The measured removal rates are in agreement with predicted in 4 of the 5 runs. This is believed due to enhanced surface deposition of elemental iodine as a result of flow induced mixing.

The removal rate was increased by increasing the flow rate in Run A-17. The increase in removal rate was slightly less than predicted, due apparently to a lowering in removal efficiency for methyl iodide.

Run A-16 used an extended release of fission products, with the loop operating from the beginning. The observed efficiency is some 25% lower than observed in prior runs. This decrease is attributed to drying of the containment atmosphere, which caused a reduced natural deposition rate. Thus it appears that the loop operated at 100% efficiency, but that our method for correcting for natural transport removal was faulty.

TABLE 18. Comparison of Observed and Predicted
Fission Product Removal Rates By Filter Loop

CSE Run No.	Early Concentration Half-Times, $t_{1/2}$, minutes														
	A-13			A-14			A-15			A-16			A-17		
	<u>N</u>	<u>N+L</u>	<u>L</u>	<u>N</u>	<u>N+L</u>	<u>L</u>	<u>N</u>	<u>N+L</u>	<u>L</u>	<u>N^(a)</u>	<u>N+L</u>	<u>L</u>	<u>N</u>	<u>N+L</u>	<u>L</u>
Elemental I	26	7.3	10.1	16	4.9	7.1	27	6.0	7.7	15	7.3	14.3	13	3.8	5.4
"Particulate" I	50	11	14.1	27	9	13.5	27	10.3	16.7	36	15	25.7	25	6.3	8.7
AC Paper	50	16	21	39 ^(a)	9	11.8	58	10	12.5	39	13.7	14.2	20	7.7	12.5
Char Bed	-	15	15 ^(c)	-	14.3	14.3 ^(c)	-	14.7	14.7 ^(c) 19.0 ^(b)	-	16	16 ^(b)	-	11	11 ^(b)
Cesium	62	11	13.7	42	12.3	17.4	70	13	16.0	47	9.3	11.5	33	7.7	10
Uranium	80	14	<u>17.0</u>	85	13	<u>15.4</u>	70	12	<u>14.5</u>	31	16	<u>33</u>	41	8.3	<u>10.4</u>
Average			15.15			13.25			14.4			19.1			9.7
Expected ^(e) , min			14.8			13.5			14.6 ^(d)			14.2			7.9
Ratio: Exp./Calc.			.98			1.02			1.01			.743			.81

N = Natural L = Loop Only

(a) The natural removal half-time is an average of 8 CSE runs under similar conditions.

(b) Bed moisture equilibrated.

(c) "Dry" Bed.

(d) Excludes (c) above.

(e) $t_{1/2} = \frac{0.693 V}{F}$, F Includes steam condensed. Assumed loop efficiency = 100%.

APPROACH TO EQUILIBRIUM GAS CONCENTRATION

The initial removal rate continues until several vessel volumes have passed through the loop. The removal rate then decreases and asymptotically approaches zero. The net removal rate does not vanish altogether, however, so that for all CSE loop tests the gas phase concentration was decreasing when the loop flow was terminated.

The change in iodine removal rate with time is depicted in Figure 22, where the normalized removal rate (observed rate/rate for 100% loop efficiency) is shown as a function of the number of vessel volumes passed through the loop. The removal rate remains constant at a value 50% to 110% higher than the theoretical value until the atmosphere has been turned over 1.5 to 2.5 times. The net removal rate then falls with time. The observed removal rate is equal to the theoretical rate at 2 to 3 turnovers. The removal rate continues to fall with time and is less than 10% of the theoretical value for times equivalent to more than 10 turnovers.

Similar data for cesium are shown in Figure 23. For cesium, the initial rapid removal rate persists for a longer time. The removal rate does not diminish from the initial value until 3.5 to 5.5 vessel volumes have passed through the air cleaning loop. At long times, the removal rate approaches zero.

The decrease in removal rate may be due to two factors. According to the material balance equation, the net removal is equal to the sum of loop removal and wall absorption:

$$-\frac{dC_g}{dt} = \underbrace{+\frac{FE}{V}C_g}_{\text{loop removal}} + \underbrace{k_g A(C_g - C_{gi})}_{\text{wall removal}} \quad (5)$$

A decrease in net removal rate would be observed if the loop removal efficiency, E , decreased markedly. Also, the net removal rate would decrease if the wall removal term changed sign and became a source term. This would be expected to take place as the gas concentration decreased below the concentration at absorption interfaces:

FIGURE 22. Comparison of Iodine Removal Rate With Theory

FIGURE 23. Comparison of Cesium Removal Rate With Theory

$$C_g \leq C_{gi}$$

The loop removal efficiency was measured directly by interstage sampling. The most extensive data were obtained in Run A-16. These are presented in Table 19. The interstage samplers show that the removal efficiencies decrease slightly with time, but the decrease is of a negligible magnitude and could not account for the observed lowering in removal rate. On this basis we must conclude that surface desorption is the controlling factor in causing the removal rate to fall.

An experimental observation which re-enforces the conclusion that surface desorption is the controlling factor, is that in Run A-14, the iodine gas phase concentration appeared to increase when the loop flow was terminated. The airborne iodine concentration for Run A-14 is shown in Figure A-2 of the Appendix. An increase in cesium concentration was unobservable, because the concentrations were too close to background level.

The plausibility of release from surfaces was examined by estimating the generation rates which would be needed to maintain a steady state, low level, concentration. For cesium, the concentration at long times was of the order $0.1 \mu\text{g}/\text{M}^3$. For a loop flow of 1000 cfm, the cesium aerosol generation rate needs to be only 3 micrograms per minute. Detachment of drops and bubbles could account for this low generation rate. For iodine, the concentration levels were about 100 times higher, and a generation rate of 300 micrograms per minute would be needed. This, however, amounts to only 3 parts in one million, per minute compared to the amount of iodine injected. This is plausible.

A simple model which accounts for desorption may be constructed as pictured in Figure 24.

TABLE 19. Measured Loop Decontamination Factors,^(a) Run A-16

Time, Minutes ^(d)	30			85			110			180		
	Cs	I	CH ₃ I	Cs	I	CH ₃ I	Cs	I	CH ₃ I	Cs	I	CH ₃ I
HEX + ME	3	1	1	4	1	1	4	1	1	3	1	1
Prefilter	3	4	1	1	2	1	1	2	1	1	2	1
HEPA	>200	18	1	1000	3	1	700	2	1	>15 ^(b)	1	1
1st Char Bed		10	2		15	2		20	2		10	2
2nd Char Bed		6	5		8	4		10	3		>2 ^(b)	1
3rd Char Bed												
Overall ^(c)	>2000 ^(b)	4300	53	4500	1000	30	2900	>1000 ^(b)	20	>40 ^(b)	>100 ^(b)	30

(a) DF = Conc. in/Conc. out; Efficiency = 1 - 1/DF.

(b) Measured downstream conc. was less than the blank.

(c) Best estimate of all components.

(d) Loop flow started at (-10) min. Aerosol release started at (0) min.

FIGURE 24. Model of Surface Desorption Influence

If equilibrium between the gas and liquid phase is assumed, the input rate to the gas phase

$$\text{input rate from liquid} = V_L \frac{dC_L}{dt} \quad (11)$$

The output rate from the gas phase is

$$\text{output rate} = F C_g \quad (12)$$

and the accumulation rate within the gas phase is

$$\text{accumulation rate} = V \frac{dC_g}{dt} \quad (13)$$

From a material balance, equations (11), (12), and (13), lead to

$$V_L \frac{dC_L}{dt} = F C_g + V \frac{dC_g}{dt} \quad (14)$$

Using the equilibrium relation, $C_L = H C_g$ equation (14) may be written as

$$-\frac{dC_g}{dt} = \frac{-F C_g}{V(1 + \frac{V_L H}{V})} \quad (15)$$

Thus the removal rate is diminished from the ideal, non-desorption case by a factor of $(1 + \frac{V_L H}{V})$. For the CSE experiments, V_L/V varied with time. For long times, it was about 0.01. Thus with a partition coefficient of 1000, the loop removal rate would be reduced to 10% of the value for no surface effects. This simple model thus appears to reasonably fit the observations.

In summary, surface desorption effects play an important role in describing removal of airborne fission products by filter loops. Neglect of surface effects is conservative for short times. At longer times, the surface desorption represents a source term which appreciably slows the removal rate.

EFFECT OF HEAT EXCHANGER AND MIST ELIMINATOR ON LOOP OPERATION

Deletion of the heat exchanger and mist eliminator from the loop during Run A-15 did not significantly reduce the loop effectiveness, but stressed the system in two ways. First, the HEPA pressure difference increased more during this run than in the runs which used the omitted components. The observed pressure drops for Run A-15 are shown in Figure 25. The total loop ΔP decreased to nearly normal values during the DOP test performed after the run.

Moisture may have carried over to the first charcoal bed, as an increase in ΔP was noted there also. Both iodine and cesium removal efficiencies remained high for the loop which did not incorporate the heat exchanger and mist eliminator.

The second effect of deleting the mist eliminator is to increase the fission product loading on the HEPA filter and charcoal. The moisture eliminator is an effective prefilter, and together with the heat exchanger

FIGURE 25. Effect of Heat Exchanger and Mist Eliminator on Loop Pressure Drop

can remove 50% to 70% of the cesium from the inlet gas. For iodine, this effect is smaller. Only 3% to 4% of the iodine is removed by these two components. The efficiency of the moisture separator for cesium particles is somewhat less than expected, however. Hilliard⁽²⁾ estimates the Cs to be present as droplets about 10 microns in diameter, and Peters⁽²²⁾ calculates the separator to be 98% efficient for 10 micron particles. We measure about 50 to 70%, which is significantly less than estimated from Peters⁽²²⁾ work. Although we can postulate no reason for it, it is possible that drops may evaporate during passage through the mist eliminator and the measured efficiency would then be explained by the smaller particle sizes. Cascade impactor sampling during the runs indicated that the cesium particles were only 0.5 to 1 μ , which is more consistent with the low removal efficiency for the mist eliminator.

INFLUENCE OF RELATIVE HUMIDITY ON METHYL IODIDE ADSORPTION

The efficiency of charcoal for removal of methyl iodide from humid containment atmospheres is important in determining the overall removal of iodine. The CSE tests were all made under conditions of 100% relative humidity. The atmospheric conditions were established by addition of saturated steam. All vessel heat loss was at the outer surfaces and no significant sources of internal heat (except the fan motor and lamps which amounted to about 10 KW maximum compared to $\sim$ 100 KW of external losses) existed, hence it may be expected that the vessel atmosphere was saturated with water.

As noted in an earlier section, charcoal heating due to moisture adsorption had been measured. Two types of charcoal tests were made. In one type of test the charcoal was exposed to steam and methyl iodide simultaneously. In the second the charcoal was exposed to the steam air atmosphere first, allowing the bed to become saturated with water vapor at the test conditions prior to exposure to the methyl iodide. Good removal of methyl iodide by the iodine impregnated charcoal was observed in both cases. Table 20 gives the charcoal efficiencies as determined by the observed main room half times.

TABLE 20. Methyl Iodide Removal By Charcoal

Run	Calc ^(b) $t_{1/2}$ min	Dry (Self-Heated)		Saturated		Integrated ^(c) Efficiency, %
		$t_{1/2}$, min	Eff ^(a) %	$t_{1/2}$ min	Eff ^(a) %	
A-13	14.8	15	99			99.2
A-14	13.5	14.3	94			99.98
A-15	14.6	14.7	99	19	77	99.96
A-16	14.2			16	89	99.992
A-17	7.9			11	72	99.993
Average			97.3%		79.3%	

$$(a) \text{ Efficiency} = \frac{t_{1/2} \text{ calculated}}{t_{1/2} \text{ observed}}$$

$$(b) \ t_{1/2} \text{ calculated} = 0.693 \text{ V/F.}$$

$$(c) \text{ Integrated Efficiency, \%} = \frac{\text{CH}_3\text{I Removed}}{\text{CH}_3\text{I Released}} 100.$$

In the tests where fission products and steam were exposed simultaneously, the self-heating of the charcoal prevents condensation in the bed. However, use of beds which had been previously saturated with water vapor also show good removal. The integrated removal of methyl iodide as shown in Table 20 is essentially the same for both types of experiments.

Normal applications of charcoal beds for CH_3I removal use 2" of charcoal at a face velocity of about 45 feet per minute. The residence time in the bed is thus about 0.22 seconds. Matching of the flow of the HEPA filter and a single 2 x 2 face area charcoal unit in the CSE loop required use of three 1" deep beds in series. The residence time at 1000 cfm was about 0.21 sec., which is nearly equal to the recommended practice. Thus the charcoal performance should be equivalent to a 2" deep bed operated at 45 fpm. In addition the three individual layers provide a better opportunity to examine the iodine loading vs depth. The measurements made indicate that most of the iodine is trapped on the first 1/2" of the first bed. Only small amounts of iodine were found in the downstream charcoal, as shown in Table 7.

FILTER LOOP CLEANUP IN PWR CONTAINMENT VESSELS

PWR containment vessels are much larger than the CSE vessel, hence the removal rate data obtained in the CSE tests do not apply directly. Following are general recommendations for analyzing fission product removal by filter loops.

First, the removal efficiency for all fission product species except methyl iodide may be taken as 100%. For methyl iodide the removal efficiency depends on the relative humidity. The lowest efficiency observed in CSE tests with 100% relative humidity was 77% at 1000 cfm, for three beds in series (residence time of 0.21 sec). Thus for methyl iodide a loop removal efficiency of 75% is justified for containment vessel usage.

Initially, the gas phase removal rate will be equal to the loop removal rate and surface deposition term as described in equation (6). The surface deposition rates may be estimated from data presented by Knudsen⁽¹⁾ and Hilliard⁽²⁾.

After two volumes have been turned over by the loop, surface deposition of iodine should be ignored, and a reversible source, such as described by Eq (15) should be factored in. Generally, the partition coefficient must be predicted. For this we recommend CSE data be used.⁽²³⁾ Selection of CSE data must be based on a particular solution type. For example, the partition coefficient of a liquid pool containing only boric acid would be much lower than a solution of NaOH having a much higher pH. The higher pH solutions will minimize the iodine desorption as the airborne concentrations decrease. For particulate materials, the 100% removal rate will persist until four containment volumes have passed through the loop. For longer times a source term should again be factored in. The source strength may be estimated from conservative use of data shown in Figure 23.

If reactive sprays are used in conjunction with a filter loop, one would conclude that elemental iodine removal will be governed almost entirely by spray washout. Methyl iodide will, on the other hand, be

removed only by the filter loop. Particulate materials will fall between, being removed by both the sprays and filter loop.

ACKNOWLEDGEMENTS

These experiments required the efforts of many persons and would not have been successful without their full cooperation. We are pleased to acknowledge the help of the entire CSE staff. In addition, we wish to recognize the contributions of G. J. Rogers, Manager of Reactor Safeguards Experiment Section, B. M. Johnson, Manager of Fluids and Energy Systems Section, L. F. Coleman, and C. E. Linderoth of the RSE staff, in the completion of this work.

NOMENCLATURE

a	= effective surface of adsorbent per unit volume of bed
A	= interfacial area for mass transfer
B_1	= inter-room transport coefficient
C_g	= mass concentration in gas of main room
C_{gi}	= solute concentration at interface
C_{go}	= mass concentration in gas at $t = 0$
C_{gi}	= concentration in gas of adjacent room
C_1	= concentration in inlet gas
C_2	= concentration in effluent gas
C_L	= concentration of solute in liquid phase
d_p	= average carbon particle diameter
D_v	= gas phase diffusivity
E	= fraction of airborne species removed by filter in a single pass
F	= flow rate through filter loop
G	= fission product mass injection rate
H	= iodine gas-liquid partition coefficient
k	= thermal conductivity
k_g	= interfacial mass transfer coefficient
R	= rate of production of a species by a vapor phase reaction
Re_p	= Reynolds number for a granule = $\rho d_p U/\mu$
Sc	= Schmidt number = $\mu/\rho D_v$
t	= time
U	= superficial gas velocity
V	= main gas volume
V_L	= liquid volume
x_0	= depth of charcoal bed

Greek Symbols

ρ	= density of gas
μ	= absolute viscosity of gas

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APPENDIX

Tables and Figures of data from the five air cleaning runs are attached. Adequate information is given to support the conclusions drawn in the report and to present detailed information for the use of other workers.

TABLE A-1. Measured Loop Decontamination Factors, Run A-13

	60 Min.			100 Min.		
	<u>Cs</u>	<u>I</u>	<u>CH₃I</u>	<u>Cs</u>	<u>I</u>	<u>CH₃I</u>
Prefilter	2	-	1	1	-	1
HEPA	>5000	-	1	>2000	-	1
1st CB	-	270,000	4	-	170	4
2nd CB	-	-	16	-	-	1.5
3rd CB	-	-	28	-	-	2.7
Overall	>10,000	>270,000	1800	>2000	>170	17

Loop Flow started at 40 min.

DF = Conc in/conc out, efficiency = $1 - \frac{1}{DF}$

TABLE A-2. Measured Loop Decontamination Factors, Run A-14

	35 Min.			90 Min.		
	<u>Cs</u>	<u>I</u>	<u>CH₃I</u>	<u>Cs</u>	<u>I</u>	<u>CH₃I</u>
Hex + Dem.	3.5	1.4	-	3	2.2	-
Prefilter	1.7	1.6	-	1	1	-
HEPA	6800	1.7	1	2500	1	1
1st CB	-	>1500	4.2	-	>70	2
2nd CB	-	~10	120	-	~3	4
3rd CB	-	-		-	-	4
Overall	41,000	>55,000	420	7700	>176	32

$$DF = \text{Conc In/Conc out, efficiency} = 1 - \frac{1}{DF}$$

Loop started at 30 min.

TABLE A-3. Measured Loop Decontamination Factors, Run A-15

	35 Min			90 Min		
	<u>Cs</u>	<u>I</u>	<u>CH₃I</u>	<u>Cs</u>	<u>I</u>	<u>CH₃I</u>
Prefilter	1	1	-	-	1	-
HEPA	-	1.6	1	-	2	1
1st CB	-	1400	2.5	-	>60	1.7
2nd CB	-	-	80	-	-	4.6
3rd CB	-	-	10	-	-	4
Overall	6000	>250,000	2000	2000	>1000	34

Loop started at 30 min.

TABLE A-4. Measured Loop Decontamination Factors,^(a) Run A-16

Time, Minutes ^(d)	30			85			110			180		
	Cs	I	CH ₃ I	Cs	I	CH ₃ I	Cs	I	CH ₃ I	Cs	I	CH ₃ I
HEX + Dem	3	1	1	4	1	1	4	1	1	3	1	1
Prefilter	3	4	1	1	2	1	1	2	1	1	2	1
HEPA	>200	18	1	1000	3	1	700	2	1	>15 ^(b)	1	1
1st Char Bed		10	2		15	2		20	2		10	2
2nd Char Bed		6	5		8	4		10	3		>2 ^(b)	1
3rd Char Bed												
Overall ^(c)	>2000 ^(b)	4300	53	4500	1000	30	2900	>1000 ^(b)	20	>40 ^(b)	>100 ^(b)	30

(a) DF = Conc. in/Conc. out; Efficiency = 1 - 1/DF.

(b) Measured downstream conc. was less than the blank.

(c) Best estimate of all components.

(d) Loop flow started at (-10) min. Aerosol release started at (0) min.

TABLE A-5. Measured Loop Decontamination Factors,^(a) Run A-17

Time, minutes ^(b)	35			60			80		
	<u>Cs</u>	<u>I</u>	<u>CH₃I</u>	<u>Cs</u>	<u>I</u>	<u>CH₃I</u>	<u>Cs</u>	<u>I</u>	<u>CH₃I</u>
HEX	1.2	1.3		1.2	1.6		1.2	1.4	
Dem	6.2	1.3		3.7	1.1		2.4	2.5	
HEPA	44	1.6		40	85			>13	
1st Char Bed		470	1.7			1.7			1.7
2nd Char Bed		>7							
3rd Char Bed									
Overall ^(c)	27,000	>8000	12	1900	>120	10	520	>180	7.5

(a) DF = Conc. in/Conc. out; Efficiency = 1 = 1/DF

(b) Loop flow started at t = 30 min.

(c) Best estimate of all components.

FIGURE A-1. Iodine Concentration in the Main Room Gas Space in CSE Run A-13

FIGURE A-2. Iodine Concentration in the Main Room Gas Space in CSE Run A-14

Neg. No. 700315-4

FIGURE A-3. Iodine Concentration in the Main Room Gas Space in CSE Run A-15

FIGURE A-4. Iodine Concentration in the Main Room
Gas Space in CSE Run A-16

FIGURE A-5. Iodine Concentration in the Main Room Gas Space in CSE Run A-17

FIGURE A-6. Cesium and Uranium Concentrations in the Main Room Gas Space in CSE Run A-13

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FIGURE A-7. Cesium and Uranium Concentrations in the Main Room Gas Space in CSE Run A-14

FIGURE A-8. Cesium and Uranium Concentrations in the Main Room Gas Space in CSE Run A-15

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FIGURE A-9. Cesium and Uranium Concentrations in the Main Room Gas Space in CSE Run A-16

FIGURE A-10. Cesium and Uranium Concentrations in the Main Room Gas Space in CSE Run A-17

FIGURE A-11. Iodine Concentration in Liquid Sumps in CSE Run A-13

FIGURE A-12. Iodine Concentration in Liquid Sumps in CSE Run A-14

FIGURE A-13. Iodine Concentration in Liquid Sumps in CSE Run A-15

FIGURE A-14. Iodine Concentration in Liquid Sumps in CSE Run A-16

FIGURE A-15. Iodine Concentration in Liquid Sumps in CSE Run A-17

FIGURE A-16. Cesium Concentration in Liquid Sumps in CSE Run A-13

FIGURE A-17. Cesium Concentration in Liquid Sumps in CSE Run A-14

FIGURE A-18. Cesium Concentration in Liquid Sumps in CSE Run A-15

FIGURE A-19. Cesium Concentration in Liquid Sumps in CSE Run A-16

FIGURE A-20. Cesium Concentration in Liquid Sumps in CSE Run A-17

FIGURE A-21. Water Volumes Accumulated in CSE Run A-14

FIGURE A-22. Water Volumes Accumulated in CSE Run A-15

FIGURE A-23. Water Volumes Accumulated in CSE Run A-16

FIGURE A-24. Water Volumes Accumulated in CSE Run A-17

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FIGURE A-25. Iodine Material Balance Versus Time for CSE Run A-13

FIGURE A-26. Iodine Material Balance Versus Time for CSE Run A-14

FIGURE A-27. Iodine Material Balance Versus Time for CSE Run A-15

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FIGURE A-28. Iodine Material Balance Versus Time for CSE Run A-16

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FIGURE A-29. Total Iodine Material Balance vs. Time for CSE Run A-17

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FIGURE A-30. Cesium Material Balance Versus Time for CSE Run A-13

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FIGURE A-31. Cesium Material Balance Versus Time for CSE Run A-14

FIGURE A-32. Cesium Material Balance Versus Time for CSE Run A-15

FIGURE A-33. Total Cesium Material Balance Versus Time for CSE Run A-16

FIGURE A-34. Cesium Material Balance Versus Time for CSE Run A-17

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FIGURE A-35. Thermodynamic Conditions and Loop Flow for CSE Run A-13

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FIGURE A-36. Pressure Drop Across Loop Components in CSE Run A-13

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FIGURE A-37. Thermodynamic Conditions and Loop Flow for CSE Run A-14

FIGURE A-38. Pressure Drop Across Loop Components in CSE Run A-14

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FIGURE A-39. Thermodynamic Conditions and Loop Flow for CSE Run A-15

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FIGURE A-40. Pressure Drop Across Loop Components in CSE Run A-15

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FIGURE A-41. Thermodynamic Conditions and Loop Flow for CSE Run A-16

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FIGURE A-42. Pressure Drop Across Loop Components in CSE Run A-16

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FIGURE A-43. Thermodynamic Conditions and Loop Flow for CSE Run A-17

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FIGURE A-44. Pressure Drop Across Loop Components in CSE Run A-17

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