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MAYPACK BEHAVIOR IN THE CONTAINMENT
SYSTEMS EXPERIMENT--
A PENETRATING ANALYSIS

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A PENETRATING ANALYSIS

By

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Physics and Engineering Division

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ABSTRACT

The Containment Systems Experiment sampling system is discussed briefly. This system uses Maypacks to sample the simulated fission products which are airborne during tests to study loss-of-coolant type of reactor accidents. The Maypack consists of a particle filter, silver screens, charcoal paper, and a charcoal bed, in order of flow.

The characteristics of the Maypacks to retain and differentiate between the various forms of iodine and particulate materials are reported in detail.

It is concluded that Maypacks can form the basis of a satisfactory sampling system, that the total airborne material is well retained in a properly used Maypack, that useful discrimination of iodine forms can be obtained, but that corrections are necessary at low inlet iodine concentrations, and that some ionic forms are not uniquely identified.

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INTRODUCTION

The Containment Systems Experiment (CSE) is a multiple purpose nuclear safety program conducted by Battelle-Northwest to evaluate the effectiveness of containment vessels and related engineered safety systems.⁽¹⁾ One part of the program is concerned with investigation of the transport behavior of fission products in a containment vessel following a loss of coolant accident to a water-cooled and moderated nuclear power reactor. These fission product transport studies involve large scale experiments in which selected fission product species are released in a 30,000 ft³ containment vessel. The time dependent spatial distribution of the released fission products is determined for postaccident conditions involving operation of engineered safety systems as well as passive removal by the natural processes of steam condensation and deposition. Included in the effectiveness evaluation of fission product scavenging are containment spray systems, recirculating air cleaning systems, and pool scrubbing systems.

Periodic samples of the containment gas phase provide the principal data on the physical or chemical form and the concentration of the fission products. Iodine is the fission product of primary importance because of its large inventory in the cores, high volatility, and biological importance. Iodine is a reactive material possessing an appreciable vapor pressure at temperatures common to postaccident conditions within containment vessels. Hence, in addition to the elemental vapors, iodine can react and be present in the atmosphere as HI or HOI. Organic forms can be produced, and CH₃I and higher molecular

weight compounds have been identified as stable airborne forms.⁽²⁾ Moreover iodine can exist in physical or chemical combination with other solid matter, and can be airborne as small particles. Knowledge of the various iodine forms is necessary for maximum understanding of this subject.

Determination of the iodine form and concentration under various containment conditions is accomplished in the Containment Systems Experiment by use of a sampling system employing Maypacks as its collection and classification device. The Maypack - so named after its originator Fred May of Harwell⁽³⁾ - is a series of selective media for the physical and chemical retention of various forms of iodine. Other fission products are also, of course, retained by the Maypack filters. Thus, the Maypack offers a potentially simple, effective way to obtain the desired information on iodine form in addition to the total concentration of the airborne simulants. Unfortunately, Maypacks are not perfect discriminators of iodine forms, and it has been necessary to experimentally determine the sampling behavior for the conditions anticipated in the CSE studies.

This report presents results of experimental work done to establish the performance characteristics of Maypack media and arrangements for the CSE use, and to establish data to support correction and interpretation of Maypack data from CSE runs.

SUMMARY AND CONCLUSIONS

The use of Maypacks and their performance under conditions expected in the Containment Systems is reviewed. Maypacks are used to sample the atmosphere resulting from simulated MCA conditions for water-cooled reactors. The retention of iodine, methyl iodide, and cesium (oxide) particles by the CSE Maypacks has been measured for the expected containment concentration of 10^5 to 10^{-2} $\mu\text{g}/\text{m}^3$ for iodine, temperatures of 20 to 120 °C, and for atmospheric conditions of dry air to saturated steam-air mixtures.

Normal CSE Maypack sample flow is 1/2 scfm of air for 3 min through 2-in. diam Maypack components for a face velocity of 30 ft/min. The order of components arrangement is:

- An inlet check valve
- Two Gelman Type A glass fiber filters
- (a) Six 100-mesh copper screens, silver plated
- (b) One Selas Corp. silver membrane filter, 5 μ pore
- One Gelman Type AC charcoal-loaded filter
- Sixty grams of activated coconut charcoal of 6 to 14 mesh.

These Maypacks are shown to efficiently retain all forms of iodine and particulate material. The discrimination between forms of iodine is not, however, unequivocal. More than one form may be held on a section of the Maypack. Because of this, correction factors aiding in the interpretation of the Maypack data have been developed.

All Maypacks in a CSE run are subjected to nearly the same temperature-time history, and sample flows and durations are similarly controlled. Hence, while these variables affect the Maypack behavior, any changes in sample distribution within the Maypack during the run is meaningful. The discrimination is good at higher iodine concentrations. Below about 1 mg/m³ of iodine, significant corrections for elemental iodine on the particle filters are necessary in higher temperature steam-air atmospheres. These corrections are less important in dry air or low temperature air.

Data to define the behavior of the CSE Maypacks and sampling system have been taken from bench tests, ADF tests, and full sized CSE runs. In addition, the experiences and observations of other users of Maypacks have been applied where possible. Each installation, however, has developed somewhat different forms of Maypacks. These differences in component arrangement and specifications, in sample flows and durations, and in atmospheric concentrations and composition can greatly affect

the Maypack performance. Consequently, while the performance of the CSE Maypacks can be generally useful elsewhere, the following specific conclusions may apply only to the CSE system:

- Maypacks can form the basis for a satisfactory sampling system.
- Maypacks provide useful discrimination of iodine forms.
- Corrections are necessary at low concentrations to allow for iodine adsorption on the filters.
- Inlet deposition is low, but should be, and is, accounted for.
- Consistent behavior is obtained by sampling at uniform flow and duration.
- Temperature and humidity can affect Maypack behavior.
- Aerosol penetration through the Maypack is small.
- The behavior and importance of ionic forms (as HI and HOI) is not well defined.
- Iodine forms retained on charcoal paper are not clearly identified.

Improvements in Maypack performance could be realized by development of an efficient particle filter media with low iodine retention. Chemical treatment of the glass fibers should be explored further. The sizable corrections now necessary at low iodine concentrations can cause significant errors in the measured amounts of particulate iodine form late in a run. Maypacks currently in use are not suitable for identification of HI as a separate form. Modified Maypacks using ion exchange membranes as suggested in Reference 12 offer a means to distinguish HI.

DISCUSSION

CSE SAMPLING SYSTEM

The purpose of the Maypack sampling system is to measure the concentration and forms of the airborne fission product species within the containment vessel. This measurement should

be accomplished in a simple and effective manner. Knowledge of the transport behavior of aerosol species is necessary for establishing the validity and the applicable range of mathematical models to fission product behavior. Verification of such models will permit more accurate assessment of the consequences of fission product release accidents.

The fission product transport experiments in CSE are conducted by releasing simulated fission product aerosols⁽⁴⁾ into various postaccident environments, generally involving condensing steam-air atmospheres. Sampling of the concentration and form of the various simulants from the accident environment is accomplished by use of clusters of Maypacks located throughout the vapor space in the containment vessel. The present system can use up to 15 clusters of 12 Maypacks each. Hence, samples can be obtained at 12 times during the course of an experiment in 15 locations. The Maypack clusters remain inside the containment vessel throughout the one or two day experiment, and are removed about one day after the end of the run.

Figure 1 is a schematic of the CSE vessel and shows some of the sampling locations. Figure 2 is a photograph of a cluster installed within the vessel. The supporting cables, electrical control cable, and vacuum hoses can also be seen. Hanging the clusters with a cable, pulley, and winch arrangement permits convenient selection of cluster location to suit the particular containment test. In practice, many locations remain fixed from test to test.

The Maypacks within the cluster are connected to a vacuum manifold through individual solenoid valves. The Maypack to be used is selected by energizing the proper solenoid from a control panel. Figure 3 shows the cluster with the rain cover removed. The manifold, valves, and quick-disconnects for the Maypacks can be seen. This unit has been used in 10 steam-air runs. Figure 4 is a photograph of the electrical control panel

Neg PNL0674084-16

FIGURE 1. Schematic of Containment Vessel

Neg 45778-3

FIGURE 2. View of Maypack Clusters Installed in the Containment Vessel

Neg PNL0692095-1

FIGURE 3. *View of Maypack Cluster with Skirt Removed*

Neg PNL0671332-38

FIGURE 4. Maypack Cluster Electrical Panel and Flow Stations

and flow control stations. Selected solenoid valves in all the clusters can be ganged so that the flow is directed through a particular Maypack in each cluster simultaneously. A manually started electrical timer is used to automatically control the duration of the sample flow period by opening and closing the solenoid valves of the selected Maypacks. After each sample is completed, the selector switches are manually advanced to the next Maypack in each cluster.

Flow through each cluster is controlled at the flow control stations shown in Figure 4. Figure 5 is a schematic diagram of the entire system. Flow measurement is based on the volume of air flow after the steam has been condensed. Following the condenser, a pressure reducing valve lowers the sample pressure from containment pressure to near atmospheric. The air is dried by passage through glass beads at -80°C in a dry ice-isopropanol bath. Any activity penetrating the Maypack in the vessel is trapped by the bed of granular activated charcoal, also at dry ice temperatures. Air flow is measured by a rotameter, controlled by a needle valve, and finally is exhausted through a vacuum manifold. Vacuum is not needed for a pressurized test, but is used to provide the required pressure drop for sampling when the containment vessel is not pressurized. Normal sample flow is $1/2$ scfm of air for 3 min. The actual sampled volume is based on containment pressure and temperature measured at each cluster during the sample time. In a saturated system, the steam-to-air ratio is known from the temperature and pressure. Hence, the actual sample volume at containment conditions can be calculated and is the basis for reporting the fission product concentrations. This method avoids apparent changes in aerosol concentration which would occur from steam pressure changes if the vapor volume at standard pressure and temperature were used as a sample basis.

FIGURE 5. Schematic of Sampling System

The components of the CSE Maypacks are arranged in order as follows:

- An inlet check valve
- Two Gelman Type A glass fiber filters^(a)
- (a) Six 100-mesh silver-plated copper screens
- (b) One silver membrane filter 5 μ pore^(b)
- One Gelman Type AC charcoal loaded filter^(a)
- Sixty grams of activated coconut charcoal of 6-14 mesh.^(c)

The Maypack is arranged so that commercial 2-in. diam components are compressed between the end of the inlet nose cone and a stop ring pressed into the stainless steel tubing which forms the Maypack body. The 2-in. diam components permit a reasonable flow rate consistent with good retention. Teflon gaskets prevent bypassing of the filters and provide for easy disassembly. The natural spring in the flat copper screens serves to keep the entire assembly under pressure. Figure 6 is a section of the loaded Maypack. Viton® "O" rings seal the ends and a spring loaded screen prevents movement of the charcoal bed. The normal attitude of the Maypacks is inlet nose down in the clusters. A teflon ball seated by gravity in the inlet nose cone serves as a simple check valve to prevent diffusion of material into the Maypack during the period of exposure to the containment vessel atmosphere.

MAYPACK PERFORMANCE

Maypacks are an effective, simple, sampling device but they are not capable of complete discrimination between the various airborne species used. For instance, while all the particulate material is expected to be removed by the first filter, some is also deposited on the inlet nose and check

-
- a. *Gelman Instrument Co., Ann Arbor, Michigan*
 - b. *Selas Flotronics Co., Spring House, Pa.*
 - c. *Barnebey and Cheney Co., Columbus, Ohio; Type 416 ®E.I. duPont de Nemours Co., Wilmington, Del.*

FIGURE 6. Section of CSE Maypack

valve assembly. The filters will remove some of the elemental iodine passing through them, and some of the methyl iodide will be retained in the charcoal paper. Thus, it can be seen that nearly all the forms sampled from the atmosphere will be trapped to some extent at two or more locations within the Maypack. While the total recovery of the Maypack is excellent, the apparent distribution must be corrected to more accurately determine the true concentration of the forms sampled.

The CSE provides a unique sampling situation, in that the concentration and forms of materials to be sampled are largely determined before the run by selection of the simulants released. In addition, each sample has experienced approximately the same temperature-time history. These circumstances permit testing the Maypack discrimination under actual run conditions. In addition, correction factors may be determined to enable the establishment of the true airborne concentrations from the somewhat imperfect samplers.

Maypack performance information has been obtained by two general methods consisting of 1) duplicating the expected sampling environments and concentrations in the Aerosol Development Facility (ADF) laboratory⁽⁴⁾ and determining the retention of known forms in the Maypack, and 2) analysis of multiple components from Maypacks taken during actual CSE runs. For example, the two particle filters are normally counted as one unit. Individual counting of the two filters can, however, give information on penetration of particles and retention of nonparticle forms.

Particle Retention on Particle Filters

The glass fiber filter media, as purchased, has a stated DOP efficiency of 99.95% at a face velocity of 10 ft/min. The CSE Maypacks operate at about 30 ft/min face velocity. This velocity may change the retention from the catalog values and,

in addition, the possibility of leakage around the filter in the holder could lead to increased penetration.

Diocetyl phthalate (DOP) tests of the filter media⁽⁵⁾ have been made in and out of the Maypack holders and the penetrations are given in Table 1.

TABLE 1. *DOP Penetration of Maypack Filter Media and Assemblies*

<u>Component Arrangement</u>	<u>Face Velocity, ft/min</u>	<u>Penetration Average, %</u>
One Type A filter ^(a)	30	<0.001
Two Type A filters	30	<0.001
Two Type A in CSE Maypack ^(b)	30	1.4
Total Maypack ^(c)	30	<0.002

- a. In a commercial filter holder.*
b. Two thicknesses of Type A media + screens and normal gaskets. After testing, filters were found to be cut by improper assembly.
c. Complete normal Maypack load except for absence of charcoal bed.

Obviously, sampling at a velocity of 30 ft/min does not impair the efficiency of the media. However, the tests run did show some evidence of bypassing of the filter media, as could be seen from the 1.4% penetration in one case. Examination of the filters after the test showed cuts due to improper assembly. It should be noted that leakage in these cases was stopped by the silver membrane filter. The penetration of the entire filter assembly was very low as indicated by the Total Maypack test in Table 1.

In addition to the DOP tests, observations of Maypack filtering efficiencies have been made by determining the amounts of ¹³⁷Cs on the filters, screens, and silver membranes in Maypacks used in several CSE runs. Cesium is expected to exist

only as particles or droplets and not as a vapor. Consequently, its location in the Maypack can indicate filter efficiency. Table 2 shows the filter efficiencies as determined by observing the amounts of cesium on the silver membrane. Data from two CSE spray test runs are shown. Run A4 was conducted with a humid air containment atmosphere at 27 °C, and A6 with saturated steam-air at 122 °C. Additional details of the runs may be found in References 6 and 7. The filter penetration measured during actual use of the Maypack is small and not strongly affected by the atmosphere.

TABLE 2. Some Observed CSE Maypack Filter Penetrations for Cesium

Maypack No.	Penetration, %			
	Run A4		Run A6	
	Avg of 4 Clusters		Avg of 4 Clusters	
	Mean	σ	Mean	σ
1	0	0	0.7	0.9
2	0	0	1.2	0.5
3	0.03	0.05	0.2	0.4
4	0.1	0.2	0.05	0.1
5	0.4	0.7	0.06	0.1
6	0.1	0.2	0.3	0.4
7	0.1	0.3	0.01	0.03
8	0.8	1.2	0.4	0.7
9	0.1	0.2	0.1	0.1
10	0.3	0.3	0.8	1.0
11	1	0.4	0.5	0.6
Average	0.3	0.3	0.4	0.5

Other filter media also have the DOP tested. Fluorocarbon fiber filters are of special interest due to the low I₂ retention reported for them.⁽⁸⁾ However, the penetration is higher

humidity, and concentration expected in the CSE tests. Bench tests were made for conditions near ambient temperature. Maypacks loaded with 2, 4, or 6 thicknesses of the filter media were exposed to inlet air of known iodine concentration and the individual filter retention measured by gamma counting for the ^{131}I tracer. Iodine release was by evaporation of $\text{CCl}_4\text{-I}_2$ solutions or from iodine crystals prepared by the $\text{Na}_2\text{Cr}_2\text{O}_7$ fusion method.⁽⁹⁾

Tests at elevated temperatures and pressures with steam-air atmospheres were generally made by sampling from the Stainless Aerosol Tank (SAT) in the ADF Laboratory.⁽⁴⁾ Tank conditions were held constant by adding steam and air as necessary. The natural removal of the I_2 from the vapor into the condensate and removal by repeated Maypack sampling provided a decreasing iodine concentration with time. The Maypacks were preheated to slightly above the test temperatures and then inserted into the tank atmosphere through an air lock and allowed to reach thermal equilibrium before sample flow was started.

Although the first filter (glass fiber) in a Maypack is expected to retain iodine in particulate form, it may additionally adsorb some elemental iodine. The amount of elemental iodine on the first filter was found for these tests by extrapolation (on semilog paper) of the iodine amounts on downstream filters which, it was assumed, were not exposed to particulate forms. Extrapolation on this basis assumes constant removal efficiency for elemental iodine by each filter. The airborne elemental iodine concentration was taken as that on the silver components of the Maypack plus the iodine retained on all but the first two filters. This concentration adjusts for the iodine removed by the extra filters in the test Maypack. The amount of elemental iodine on the first two filters was expressed as a percentage of the adjusted amount on the silver

than for the glass fiber filter. Table 3 shows the DOP penetration at several face velocities for the "fine" grade* filters.

TABLE 3. DOP Penetration of Zitex® Filters

Test No.	Face Velocity, ft/min		
	9	14	28
	Penetration, %		
1	5.5	3.0	0.6
2	8.5	5.0	1.5
3	8.0	5.0	1.0
4	8.0	5.0	2.0
5	12.0	7.0	2.5
Average	8.4	5.0	1.5

The observed penetration was judged to be too high for routine use in the CSE samplers.

Elemental Iodine Retention on Particle Filters

Iodine is a reactive vapor and will deposit to some extent on almost all surfaces it contacts. Retention of the elemental iodine vapors upon the particulate filters is a potential problem with the Maypack for discrimination of iodine forms. The amount of iodine retained depends upon the iodine concentration, type of filter media, temperature and humidity of the atmosphere, flow rate, duration of sample period, and possibly other parameters.

Retention of elemental iodine on the CSE Maypack filter media has been measured for a range of conditions of temperature,

® Chemplast Inc., Wayne, New Jersey.

* The filters were specified as extra fine grade; however, the box received from the vendor was marked as fine. The true grade is not known.

FIGURE 7. Correction for Iodine Retention on Maypack Filters Sampling Air

components. This percentage, then, is a correction to be added to the indicated elemental iodine content on the silver section of the Maypack and thus will allow for iodine losses to the filter components. This same amount should be deducted from the filters to obtain an improved measure of the particulate iodine content of the atmosphere sampled. The total iodine in the Maypack, of course, remains unchanged.

The correction is shown for room air and humid air at 20 to 25 °C in Figures 7 and 8. During these bench tests, the inlet airborne iodine concentration was kept nearly constant by uniform release from the iodine source. The flow duration was 5 or 6 min at 1/2 cfm. Material balances were made for each release and the average inlet concentrations were corrected for iodine not released or deposited prior to reaching the Maypack. Figure 7 shows the filter percentage correction for room air to be low at high iodine concentrations and increases at lower concentrations. This increased percent retention at low iodine concentrations has been observed by others.⁽¹⁰⁾ Figure 8 gives the correction for iodine on the Maypack filters when sampling humid air at 24 °C. The behavior seems anomalous in the reduction of retained iodine at concentrations below about 30 $\mu\text{g}/\text{m}^3$. This behavior remains unexplained. However, the method of determining the correction factors assumes that elemental iodine is the species retained. If some of the iodine were to react in the presence of the moist air to form a compound retained by silver but not by filters, the general nature of Figure 8 could be explained. This behavior is similar to that noted for HOI in References 11 and 12, but the role of HOI in this case has not been established and this explanation must be regarded as a conjecture at this time.

Similar data for saturated steam-air at 120 °C (at 35 psig) and 80 °C (at about 7 psig) are shown in Figures 9 and 10. These

FIGURE 8. *Correction for Iodine Retention on Maypack Filters Sampling Humid Air*

FIGURE 9. *Correction for Iodine Retention on Maypack Filters Sampling Steam-Air at 80 °C*

FIGURE 10. Correction for Iodine Retention on Maypack Filters Sampling Steam-Air at 120 °C

conditions have been used in several CSE tests. The typical range of iodine concentration in a CSE test is from 10^5 to 10^1 $\mu\text{g}/\text{m}^3$, hence the correction for filter loss is small during much of the test duration. Work is currently in progress to extend the data in Figure 9 to lower concentrations, about $0.1 \mu\text{g}/\text{m}^3$ of elemental iodine.

The scatter of data in Figure 10 creates some uncertainty about the correction at lower concentrations. However, nearly all the data are within a factor of two of the indicated correction. The uncertainty becomes important only late in a run. Figure 11 shows airborne concentrations versus time for CSE Run A-5 (steam-air 120°C , 35 psig) and illustrates the effect of the correction and the resulting errors. Because of the large decrease in airborne concentration during the initial period of a run, the correction would cause only minor changes in the slope but would reduce the amount of particle associated iodine by about a factor of two late in the run. Table 4 shows the changes caused by correcting for filter errors to be small. Thus, although the low precision corrections are necessary, their application does not significantly affect the interpretation of most of the data from a typical experiment.

TABLE 4. *The Effect of Filter Corrections on Early Half Times, Run A-5*

	<u>Airborne Concentration Half-Times, Minutes</u>	
	<u>Corrected</u>	<u>Uncorrected</u>
Elemental Iodine	20	19
Particle Associated Iodine	23	30

Elemental Iodine Retention on Other Filter Media

During the development and testing of the CSE Maypack, other media were examined for good particle removal ability combined with low iodine retention. Some of the results are

FIGURE 11. Effect of Filter Corrections on Typical CSE Run

shown in Table 5 for dry and humid atmospheres. In addition, treatment of the Gelman Type A fiber filter media was examined briefly. The pretreatment of the filter by drawing chlorine gas or Dri Film[®] vapors through it reduces the iodine retention, probably by filling some of the reaction sites. This process shows some promise, but has not been pursued.

Zitex filters exhibit desirably low iodine retention but are not used as a normal Maypack component because of the high particle penetration shown in DOP tests (see Table 3). The other filter material examined did not show improved behavior with the possible exception of the 1- μ pore size Nuclepore[®] filter, an irradiated, etched plastic film. The surface area of such a filter should be low, and this may be responsible for the reduced iodine pickup. Polypropylene fiber filters and 0.8- μ pore triacetate membrane filters retained large amounts of the elemental iodine and were not considered for routine use in the CSE Maypacks.

Elemental Iodine Retention by Silver and Charcoal Paper Components

The silver plated copper screens and silver membrane filters are expected to retain the elemental iodine reaching this section of the Maypack. The 5- μ pore silver membrane was included as a part of the Maypack because of its superior iodine retention. In addition, it serves as a backup filter for particles in the event of cut or broken glass fiber filters. Figure 12 shows the increased iodine retention of the silver membrane filter material over the silver plated screens at several flow rates in dry air.

The charcoal loaded filter paper downstream of the silver section of the Maypack serves to mechanically separate the silver from charcoal granules in addition to adsorbing heavy organic

[®] General Electric Co.

TABLE 5. Retention of Elemental Iodine on Various Filters^(a)

Iodine Retention, % of Total in Maypack														
		Humid Air ~99% RH					Dry Air							
Maypack Component	Type A	Zitex	Dri Film A	Type A	Cl ² A	Dri Film A	Zitex	5 μ Nuclepore	1 μ Nuclepore	Poly-Prop.	0.8 μ Memb.			
1 Filter	8.5	1.56	1.97	5.8	2.5	4.1	0.74	20 ^(b)	4.8 ^(b)	47.2	22.1			
2 Filter	7.2	0.54	1.17	4.4	5.7	0.65	0.19	3.8	0.59	29.8	14.2			
3 Filter	6.4	0.33	1.23	4.9	2.4	0.24	0.12	7.6 ^(b)	2.2 ^(b)	14.5	7.1			
4 Filter	6.6	0.40	1.28	4.1	2.1	0.26	0.19	--	--	3.8	5.5			
Ag Screens	52.8	76.4	76.7	74.3	77.7	89.3	77.4	64.6	86.6	1.3	46.2			
Ag Membrane	4.5	5.6	4.4	3.9	7.6	2.5	7.1	3.9	2.2	0.8	3.7			
AC Paper	13.5	14.6	12.8	2.4	3.1	2.5	14.2	3.5	2.9	2.1	1.4			
Charcoal Bed	0.4	0.5	0.5	0.1	0.1	1.0	0.2	0.6	0.5	0.6	0.6			

a. 1/2 cfm, 25 °C, for about 6 min. Inlet iodine concentration ~60 $\mu\text{g}/\text{m}^3$.

b. Normal Type A filter.

FIGURE 12. Elemental Iodine Retention on Silver Surfaces Versus Air Flow

forms or ionic forms which may penetrate the silver. Because iodine is reactive and is known to form small amounts of organic compounds such as CH_3I which will penetrate the silver and the charcoal paper, assay of this section of the Maypack for absolute efficiency for iodine is difficult. Although adventitious penetrating forms may be present during the tests, assignment of a minimum efficiency to the combined Ag + charcoal paper is possible by assuming the penetration to, and retention on, the charcoal bed of only elemental iodine. This assumption is obviously very conservative. Figure 13 plots the penetration for a range of concentrations and for several atmospheric conditions. The penetration given is the maximum, as it quite possibly includes organic species. The expected true penetration of elemental iodine to the charcoal bed is thought to be much below this. It can be seen that, although the atmosphere is not highly important, the penetrating fraction may increase somewhat at lower iodine concentration. The observed penetration through the silver-charcoal paper combination is small, amounting to less than about 0.4% in most cases for elemental iodine concentrations above $1 \mu\text{g}/\text{m}^3$.

The efficiency of silver for iodine removal has been measured by others^(7,8) at a few atmospheric conditions and iodine concentrations. However, the flow and component arrangements were not the same as in the CSE Maypack. Figure 14 is typical of the iodine retention on the CSE Maypack screens at low concentrations in dry filtered air, where the conversion to other forms is expected to be small. Note that each screen shows a constant removal efficiency, as evidenced by the constant slope, down to an incident iodine concentration of about $0.02 \mu\text{g}/\text{m}^3$ at the last screen. The observed efficiency of 73% for each screen and 99.95% for six screens is evidenced that silver can effectively remove elemental iodine at low concentrations.

FIGURE 13. Iodine Penetration to the Maypack Charcoal Bed

FIGURE 14. Iodine Retention on a Series of Silver Plated Screens

References 8 and 10 both indicate increased penetration of the silver with decreasing total iodine concentration in humid atmospheres, as do our tests (Figure 15). As before, the maximum possible penetration was shown because all activity retained on the backup charcoal paper was assumed to be elemental iodine when, in fact, some may be other forms. Should humidity and temperature per se reduce the efficiency of silver for iodine, the same results should be noticeable also at higher concentrations of iodine. Examination of Figure 15 shows no significant change. Hence, we can attribute the apparent loss of silver efficiency at low concentrations to the formation of other iodine species, either organic or inorganic. The conversion of small amounts of the iodine at these low concentrations would produce the results shown in Figure 15.

Other Ionic Forms on Silver

The distribution of HI in CSE Maypacks has been reported by Howes⁽¹³⁾ at a sample flow rate of 3 l/min. He observed high retention in glass filters when sampling humid air, but lower amounts on the filters in steam and air at higher temperatures (155 °C). Good retention on the silver screens was noted.

Limited studies by the author confirm the BMI studies. Table 6 summarizes the HI retention in air and steam. Retention of I₂ at similar conditions is shown for comparison.

Note the similar behavior of the HI and I₂ in steam-air atmosphere and also the low penetration to the charcoal bed of the Maypack. Large concentrations of HI are not expected in the wet, oxidizing CSE test atmospheres. Any small airborne amount possibly present in equilibrium with the iodine and the water in the system will be incorporated into the filter correction factors developed for iodine in the various atmospheres. HI sampled in this manner will lose its identity and will be reported as iodine in the silver section of the Maypack.

FIGURE 15. Maximum Iodine Penetration Through Maypack Silver Components

TABLE 6. Retention of HI in CSE Maypacks at 96 °C^(a)

Concen. mg/m ³	Atmosphere	Form Released	% of Total			
			Type A Filter (2)	Silver Screens (6)	Charcoal Paper	Charcoal Bed
33	Air	HI	10.5	36.8	52.2	0.5
10	Air	HI	25.7	74.2	0.1	0.01
2	Air	HI	54.9	43.2	1.8	0.02
1	Steam-Air	HI	76.7	23.2	0.1	0.01
0.25	Steam-Air ^(b)	HI	85.5	12.4	2.1	0.02
2	Air	I ₂	0.6	97.6	1.8	0.3
1	Steam-Air	I ₂	40.1	58.9	1.0	0.05

a. Flow duration 5 min at 1/2 cfm air; steam-air tests (50 vol% each) 1 cfm total flow.

b. Saturated steam-air at 80 °C and 15 min flow duration.

The presence of HOI during some containment conditions has been suggested from observations at the Contamination Decontamination Experiment (CDE) at NRTS.⁽¹⁴⁾ Work by this group at NRTS indicates this form to be in equilibrium with I_2 in aqueous systems, and the amounts are concentration and pH dependent.⁽¹¹⁾ The material seems to penetrate the filters well, and to be moderately well retained by silver membranes (80-98%), and charcoal papers (~70%).⁽¹²⁾ Hence, the bulk of this iodine species should be retained by the silver and charcoal paper sections of the Maypack, but added information on the identification and behavior of this species is needed.

Methyl Iodide Retention

Charcoal is known to have poor retention of large amounts of methyl iodide under humid operating conditions when exposed to a continuing flow.⁽¹⁵⁾ The retention of methyl iodide by charcoal can be enhanced by decreased loadings, increased residence time in the bed, and by use of specially treated (impregnated) charcoals. Reference 16 provides a good review of these factors.

The CSE Maypacks use a 120 cm^3 charcoal bed 6 cm deep. The usual sample flow is such that the residence time is 0.5 sec. A normal sampling duration of 3 min will result in a methyl iodide loading of about $1 \text{ } \mu\text{g/g}$ of charcoal when sampling an inlet concentration of 1 mg/m^3 . It is expected from data in Reference 17 that 99.9% should be retained even at 75°C and 100% relative humidity. Lower concentrations would be more effectively retained, and much higher concentrations are not expected to be used in the CSE.

The normal CSE Maypack does not use impregnated charcoals. Tests showed that when the run was terminated and the sampling equipment cooled, condensation in the charcoal bed leached out some of the iodide impregnant causing corrosion of the stainless

steel Maypack bodies. To avoid this corrosion, impregnated charcoals will be used only when conditions or anticipated concentrations warrant.

The distribution of methyl iodide on the charcoal paper and beds has been investigated for several conditions and is given in Table 7. The retention of methyl iodide on the particle filter media and silver components was less than 0.04% and is not shown.

TABLE 7. *Retention of Methyl Iodide in CSE Maypacks*

Temp., (a) °C	Sample Duration, min	Char- coal Paper	% Retained			
			Charcoal Bed			Backup Charcoal (-70 °C)
			1st 20 g	2nd 20 g	3rd 20 g	
30	2	1.1	98.9	0.04	0.005	0.0006
80	2	1.3	76.2	13.4	8.5	0.6
120	2	2.6	59.4	22.3	13.4	2.5
120 ^(b)	2	3.5	87.2	8.3	0.7	0.2
120 ^(b)	20	3.4	65.0	22.6	8.1	0.9

a. Saturated steam-air. Inlet methyl iodide concentration was 2 mg/m³ (ADF Run 83).

b. I₂-KI impregnated charcoal. (BC727, 8 × 20 mesh).

Examination of Table 7 shows increasing methyl iodide penetration with increasing sample temperature and increasing sample duration, as expected. The observed penetration of 2.5% to the refrigerated charcoal backup is judged to be the maximum acceptable for normal CSE runs. Obviously, more extreme conditions of temperature, time, or concentration may require the use of impregnated charcoals.

Retention of methyl iodide on the type AC charcoal-loaded filter increases from 1% to over 3% as the temperature increases. This increase is consistent with a small amount of chemisorption which would increase with temperature. The limited physical adsorption of methyl iodide on charcoal paper permits this effect to be noticeable.

Iodine penetration of the Maypacks to the backup traps has been measured in the CSE under actual sampling conditions. The amount in the traps would also include that from leaks in the sampling system. Table 8 shows penetrations observed during Run A-6, with saturated steam-air at 37 psig and 122 °C, and with a methyl iodide concentration of about 1.5 mg/m³. The average penetration of 11 clusters in the main vessel is given.

TABLE 8. *Average Iodine Penetration for Run A-6*

<u>As % Iodine Held By</u>	<u>% Penetrating to and Retained in</u>		
	<u>Condensate Pot 20 °C</u>	<u>Glass Beads -70 °C</u>	<u>Charcoal Backup -70 °C</u>
Cluster Charcoal Bed (CH ₃ I)	0.11	0.0015	1.0
Cluster Total (Iodine)	0.0075	0.0001	0.069

The actual performance of the Maypacks in use, somewhat better than expected from the data in Table 7, indicates no serious differences between the laboratory evaluation and large-scale application of the Maypacks in clusters.

Xenon Retention

The noble gas xenon has been used as a tracer in the CSE for leak testing and in the ADF for eddy diffusion measurements. The normal means of xenon collection is with a dry ice-cooled charcoal trap located at the flow control station in the laboratory. Because xenon possibly may be sampled through a line containing a Maypack, the retention of xenon in Maypacks has been examined briefly for dry air and steam-air mixtures at several temperatures. Tables 9 and 10 summarize the measurements.

TABLE 9. Retention of Xenon in Maypacks Sampling Dry Air

<u>Component</u>	<u>% of Inlet Xenon Retained^(a)</u>		
	<u>25 °C</u>	<u>76 °C</u>	<u>96 °C</u>
Charcoal Paper	0.001	--	--
40-g Charcoal Bed ^(b)	9.1	1.2	0.2

- a. Air flow at 12 l/min for 5 min; dew point ~60 °F; $P_p \text{Xe} \sim 8 \times 10^{-10}$ atmos.
 b. BC-416, 6-14 mesh.

TABLE 10. Retention of Xenon in Maypacks Sampling Saturated Air

<u>Component</u>	<u>% of Inlet Xenon Retained^(a)</u>	
	<u>32 °C</u>	<u>82 °C</u>
40-g Charcoal Bed	1.5	0.03

- a. Air flow at 1/2 cfm for 5 min.

Only when sampling air at 25 °C is any significant correction for xenon adsorption in the Maypack necessary.

PRACTICAL ASPECTS OF MAYPACK SAMPLING

In actual use, all Maypacks within the clusters are exposed to the test atmosphere for the entire run duration. The concentration of the sampled material can decrease several orders of magnitude. Hence, any leakage, diffusion, or "breathing" into the later samples will give deceptively high concentrations. Two steps are taken to reduce these problems:

- A simple gravity operated teflon ball check valve stops diffusion into the Maypack.
- One of the 12 Maypacks is not used to sample the atmosphere and serves instead as a blank to permit evaluation of average leakage and breathing effects.

This latter method has been used also with good success at the NSPP.⁽¹⁸⁾ Subtraction of the amounts in the average

blank from the activity in the later Maypacks gives the corrected amount sampled. The blank is usually significant only for the last few Maypacks.

Inlet Losses

Use of an inlet check valve not only reduces entry of material into the Maypack, but also causes greater deposition of material from the sampled atmosphere. The amount of deposition is measured for each Maypack by separate decontamination of the internal surfaces of the nose cone and teflon ball. While the amount is known and is included in the Maypack total, the exact form remains undetermined. For iodine, the elemental and particulate forms are most likely to deposit. The cesium is assumed to be deposited as particles or droplets. The relative amount of inlet deposition is given in Table 11 for seven CSE runs encompassing a range of conditions. The values reported are the averages for the first three Maypacks and the last three Maypacks in all the clusters in the main containment vessel. It can be seen that the amount of material on the inlet is low for iodine and cannot cause substantial changes in the forms collected. Although the amount of cesium deposited is larger, no uncertainty can result because the form is known in this case to be particulate. Abnormally high cesium deposition was noted in Run A-2. The cause, while unknown, may be related to temperature increases during the aerosol release. The inlet deposition for Runs A-3 and A-4 was low during the early period of the run. Both of these runs started with air at 27 °C and about 70% relative humidity. All other runs in Table 11 were made with saturated steam-air atmospheres. Moisture condensing on the inlet sections possibly accounts for the differences seen.

TABLE 11. *Typical Maypack Inlet Deposition*
Average % of Total Found on Inlet^(a)

CSE Run No.	Avg. Temp., °C	<u>1st Three Maypacks</u>		<u>Last Three Maypacks</u>	
		<u>Cs</u>	<u>I</u>	<u>Cs</u>	<u>I</u>
A1	82	27	21	1.1	0
A2	85	67	5.7	58	7.8
A3	27	1.9	0.1	0.9	7.2
A4	27	2.1	0.2	9.7	3.0
A5	122	12	0.4	35	14
A6	122	8.6	0.5	14	13
A7	122	8.6	1.4	--	23

a. Total includes inlet, filters and screens only.

Manpower Requirements

The rather simple Maypack design and the small number of components permits rapid loading and disassembly of the Maypacks. Two persons can assemble and load three clusters (36 Maypacks) per hour. Disassembly, bagging, and marking the Maypack components for analysis requires about two man-hours per cluster.

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