Item #1

AIRBORNE RADIOIODINE MEASUREMENTS
LOFT FP-2 TEST

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ABSTRACT

Samples of the Loss of Fluid Test (LOFT) containment air were obtained during the FP-2 test and analyzed for radioiodines. Species differentiation into particulate, elemental, hypoiodous acid, and organic fractions was performed. Decontamination factors (DFs) were measured for several LOFT heating and ventilating (H&V) silver zeolite filters.

Measurements indicated the presence of ^{131}I, ^{132}I, ^{133}I, and ^{135}I. A maximum ^{131}I concentration of 1.45E-4 µCi/cc was reached about 31 hours after reflood terminated the fuel heat up phase of the LOFT test. This concentration corresponds to a total containment air ^{131}I activity of about 1-1/2 Ci. The initial most prominent radioiodine species was elemental. For ^{131}I, ^{133}I, and ^{135}I the elemental fraction decreased with time after reflood while the organic fraction increased. When high volume containment purge was initiated, the elemental fraction returned to approximately its initial level. The elemental fraction of ^{132}I, however, did not exhibit this behavior with time. It remained the major species throughout the measurement period.

In situ measurements of the DF for LOFT H&V cleanup filters indicated that the DF for elemental iodine was the highest, with DFs for the particulate, hypoiodous acid, and organic species being lower. The DFs ranged from 50 to >100,000, depending on filter bank and radioiodine species.

ACKNOWLEDGEMENTS

The valuable assistance of LOFT Operations, LOFT Technical Support, and Health and Safety personnel made this measurement program successful. The financial support and direction of the U. S. Nuclear Regulatory Commission provided by P. Reed and C. Nilson of RES and W. Gammill of NRR made the measurement program possible.

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1.0 INTRODUCTION

The Loss of Fluid Test (LOFT) fission product test number 2 (FP-2) was designed to provide data on the release and transport of radiologically significant fission products (i.e., I, Te, Cs) during accident conditions. The test simulated a V-sequence accident in which a low-pressure injection system line ruptures and the emergency core cooling injection is delayed until severe fuel damage has occurred. This severe damage resulted in the release and subsequent transport of a portion of the fission product inventory in the fuel. Some of the radioiodines released from the fuel were transported to the containment (reactor building) atmosphere. The objective of the measurements reported here was to measure the concentration and chemical species of these radioiodines.

The LOFT center fuel module consisted of new 9.74-wt%-enriched fuel rods that had undergone about 430 MWD/MTU burnup prior to the transient. During the transient, fuel cladding temperatures in the center fuel module reached about 2400 K and exceeded 2100 K for at least 4-1/2 minutes. The cesium-to-iodine mass ratio in the fuel was 4.00 (atom ratio was 3.88). The ^{131}I inventory in the center fuel module at the time of the transient was calculated to be 0.3835 g (4.757E4 Ci).

During the LOFT test, water in the reactor coolant system (RCS) was released into the blowdown suppression tank (BST). The only release into the containment was due to leakage from the LOFT systems. Any radionuclides that got into the containment atmosphere, therefore, had leaked from the LOFT system.

2.0 DESCRIPTION OF EXPERIMENT

2.1 Sample Points

The only adequate sample points available were on heating and ventilating (H&V) systems 8 and 9. H&V 9 consists of a 24-inch diameter line which runs from containment to a silver zeolite (AgX) cleanup filter then either back to the containment (recirculation mode) or to the atmosphere (purge mode). A 2-inch diameter line runs from containment to a 10-inch diameter line in H&V 8. This 10-inch line runs to a AgX cleanup filter then to the atmosphere. H&V 8 is used to slowly vent the containment atmosphere while H&V 9 is used for high volume purge of the containment air.

Both H&V 8 and 9 had sample points just upstream and just downstream of their cleanup filters. These sample points were used for purposes of testing the cleanup filters and had previously been shown to yield representative samples. A helium dilution technique [1] was used in the simple point validation tests.

2.2 Iodine Species Samplers

An iodine species and particulate sampler [1-3] was used to collect samples for analysis. Figure 1 shows the features of the sampler. It consists of five media:

1. Particulate Filter. The particulate filter used is Flanders F-700, HEPA type filter paper that has been waterproofed. This filter is used to retain particulate material which is pulled into the sampler.

2. Cadmium Iodide (CdI_2) on Chromosorb P (60/80 mesh supplied by Applied Science). This is used to adsorb elemental iodine.

3. 4-Iodophenol (IPH) on Alumina (30/60 mesh F-1 activated). This is used to adsorb hypoiodous acid (HOI).

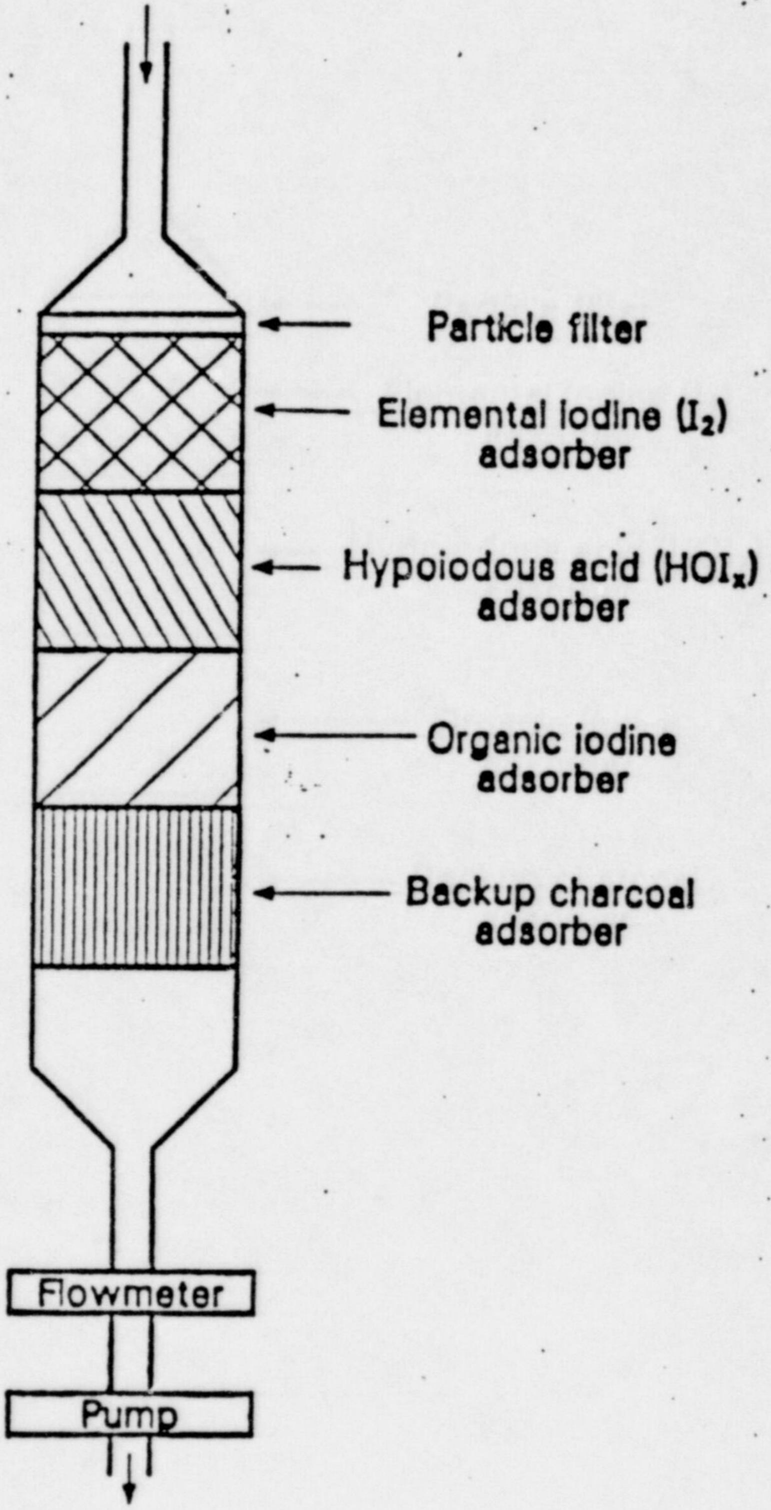

FIGURE 1. IODINE SPECIES SAMPLER

4. Silver Zeolite (AgX) (20/40 mesh). This is used to adsorb organic iodide species such as CH_3I.

5. Charcoal (Barnaby-Cheney 151) or Silver Zeolite (20/40 mesh). This is used as a backup to adsorb any species possibly escaping through the other media. For most samples, AgX was used because of its lower efficiency for adsorbing noble gases.

The media are loaded into aluminum media cups (3/8-inch high cup for the particulate filter; 1-3/8 inch high cup for the CdI_2, IPH, and AgX; and 1-inch high cup for the backup charcoal or AgX). These media cups are loaded into an aluminum sampler body in the order shown in Figure 1. The sampler body is 7-5/8 inches long and 2 inches outside diameter and has a closed and an open end. The open end is threaded to accept a seal cap. Viton 'O' rings are used to insure a seal between the cups themselves and between the cups and the sampler body.

The elemental iodine adsorption efficiency of the CdI_2 has been measured to range from 87% [3] to greater than 99%.[2] Methyl iodide adsorption is less than 1%.[2,3] Adsorption efficiency of the IPH for HOI is about 95% while the efficiency for methyl iodide is less than 1%.[2,3] Efficiency of the AgX for adsorption of methyl iodide is greater than 99%.[3]

2.3 Experimental Setup

Figure 2 shows a schematic drawing of the experimental setup. The sample upstream (downstream) of the cleanup filter was obtained from sample point TSP-1 (TSP-2). The flow went through valve S-4 (S-7), quick disconnect QD-1 (QD-3), valve S-5 (S-8), the iodine species sampler, isolating quick disconnect QD-2 (QD-4), valve S-6 (S-9), pump P-1 (P-2), and then returned to the H&V system through TSP-2. Flow rate was measured by G-3 (G-4). The N_2 tank was used to supply gas for leak testing of the system and to flush radioactive gases out of the system prior to removal of the samplers.

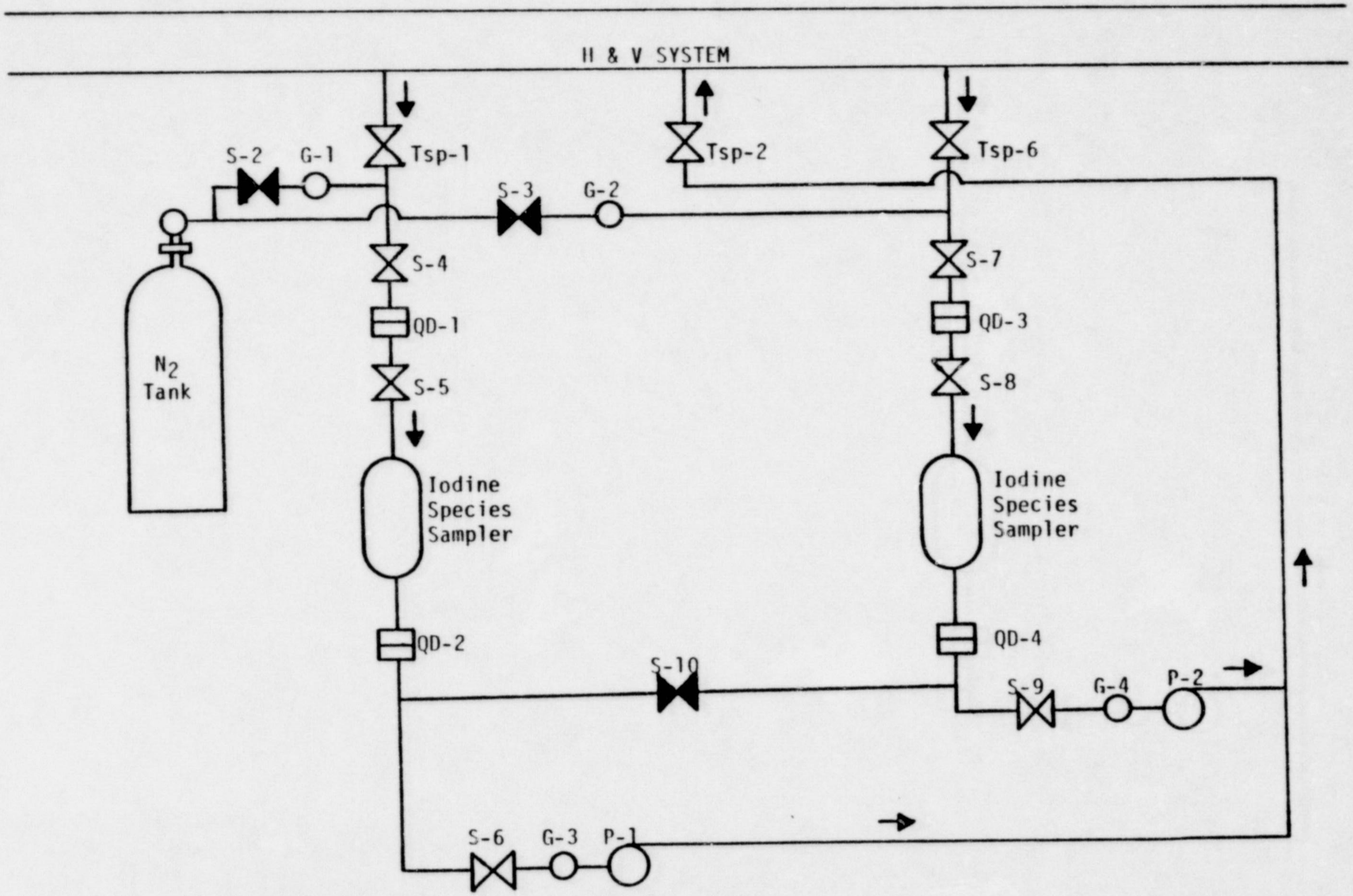

FIGURE 2. SCHEMATIC DRAWING OF SAMPLING SYSTEM

2.4 Sample Counting

After removal of an iodine species sampler from the sampling system, the sample cups were removed and placed in individual vials. These vials were then gamma counted using a hyperpure germanium detector-based spectrometer. The spectrometer was calibrated using NBS standard solutions which were loaded onto the different media.

2.5 Samples Taken

Table 1 contains a summary of the samples taken. Two samples were obtained from H&V 9 while the reactor was at power prior to the test (one on 7/3/85 and one on 7/9/85 about 5 hours before initiation of the test). Six samples were obtained from H&V 8 after the test (3.65h, 5.83h, 7.55h, 31.28h, 42.92h, and 68.32h after reflood terminated the FP-2 test). The 3.65h sample was started after H&V 8 had been running for about 15 minutes. Unfortunately, H&V 8 was secured soon after sample collection began (cf Appendix Table A-1 for the operating history of H&V 8 and 9). This sample, therefore, was taken from a closed H&V system and the measured radionuclide concentrations may not be representative of those in the containment. The iodine species mixture, however, is believed to be representative of that in the containment. Four samples were obtained from H&V 9 beginning shortly after H&V 9 was put into operation (75.82h, 77.67h, 138.75h, and 211.18 h after reflood.

Samples were taken upstream of the H&V cleanup filter for all the times noted in Table 1. In addition, at three sample times (7.55h, 31.28h, and 75.82h) one sample was taken downstream of a H&V filter so that the decontamination factor (DF) for each cleanup filter could be determined.

The 68.32h sample was taken from H&V 8 just prior to initiation of high volume containment purge and the 75.82h sample was obtained from H&V 9 just after initiation of high volume containment purge. The objective of this sample pair was to validate the samples obtained from H&V 8. Since H&V 9 utilizes a large diameter line (24-inch), samples from it would be valid because deposition of iodine from the air onto the walls of the line

TABLE 1 SAMPLE SUMMARY

Sample	H&V System	Type*	Start of sample		Sample Duration (min.)	Sample Volume (cc)	QUENCH Time After Reflood (hrs.)#
			TIME	DATE			
Pretest-1	9	1	0957	7/3/85	39	2.29E5	---
Pretest-2	9	1	0936	7/9/85	60	3.52E5	---
1	8	1	1814	7/9/85	30	1.73E5	3.65
2	8	1	2025	7/9/85	10	5.77E4	5.83
3	8	2	2208	7/9/85	36	2.09E5	7.55
4	8	2	2152	7/10/85	35	2.05E5	31.28
5	8	1	0930	7/11/85	30	2.80E5	42.92
6	8	1	1054	7/12/85	30	1.73E5	68.32
7	9	2	1824	7/12/85	34	1.91E5	75.82
8	9	1	2015	7/12/85	10	5.77E4	77.67
9	9	1	0921	7/15/85	66	3.87E5	138.75
10	9	1	0946	7/18/85	30	1.73E5	211.18

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* Type 1 - Sample taken upstream of H&V cleanup filter.

2 - Samples taken upstream and downstream of H&V cleanup filter.

\# Time between end of FP-2 test (i.e., reflood) and start of sample.

would be negligible compared to the amount of iodine in the air. A similar conclusion for samples from H&V 8 (2-inch line from containment) could not be made a priori. A comparison of the results from the 68.32h and 75.82h samples (cf Sections 3.1 and 3.2) indicates that the measured concentrations and species mixtures are comparable. No major deposition of I_2 (the most reactive species and therefore the most likely species to deposit) was apparent. It was concluded, therefore, that the samples obtained from H&V 8 were valid.

3.0 RESULTS

3.1 Radioiodine Concentrations

Table 2 lists the radioiodine concentrations in the LOFT containment measured before and after the FP-2 test. In all cases, measurements were made using samples taken from the H&V systems and appropriate dilution factors were used to obtain concentrations in the containment. The samples taken from H&V 9 were direct measurements of the containment atmosphere. Radioiodine concentrations measured in the H&V systems and the dilution factors used to obtain the concentrations in the containment are given in Appendix Tables A-2 and A-3, respectively. Note that in Table 2 and in all data tables, the reported uncertainty represents the one standard deviation counting statistics only. An additional uncertainty of about 10% should be added to account for calibration, time, and flow measurement uncertainties.

Prior to the FP-2 test, when H&V 9 was rapidly purging the containment, containment air concentrations of about 1E-9 μCi/cc were measured for ^{131}I and ^{135}I, about 2E-10 μCi/cc for ^{132}I, and about 2E-9 μCi/cc for ^{133}I. Iodine-134 was below detection limits.

The first sample obtained, 3.65 hours after reflood, indicated an increase in the radioiodine concentration in the containment air of about a factor of 4000 for ^{131}I, 8000 for ^{133}I, and 17,000 for ^{135}I. These factors are really minima since the radioiodine concentrations indicated by the 3.65h sample are probably low due to the isolation of H&V 8.(cf Sect.

TABLE 2 RADIOIODINE CONCENTRATION IN LOFT CONTAINMENT AIR #

Sample	Measured Radioiodine Concentration (uCi/cc)				
	^{131}I	^{132}I	^{133}I	^{134}I	^{135}I
Pretest-1	1.38 ± .08E-9	8.6 ± 2.5E-10	4.6 ± .2E-9	<7.3E-9	1.9 ± .7E-9
Pretest-2	9.6 ± .8E-10	2.8 ± 1.0E-10	2.02 ± .08E-9	<7.3E-10	6.4 ± 3.0E-10
1	4.01 ± .03E-6	2.35 ± .03E-6	1.54 ± .01E-5	4.1 ± 1.1E-7	1.11 ± .01E-5
2	8.8 ± .1E-5	3.52 ± .04E-5	3.42 ± .02E-4	<6.3E-6	2.234 ± .009E-4
3	1.13 ± .01E-4	2.93 ± .03E-5	4.04 ± .02E-4	<4.2E-6	2.219 ± .007E-4
4	1.45 ± .01E-4	1.1 ± .5E-6	2.45 ± .01E-4	*	2.44 ± .02E-5
5	6.20 ± .03E-5	2.4 ± .3E-7	7.47 ± .03E-5	*	3.33 ± .06E-6
6	7.41 ± .03E-5	2.1 ± .2E-7	4.09 ± .02E-5	*	2.9 ± .2E-7
7	6.33 ± .03E-5	**	2.80 ± .03E-5	*	**
8	1.434 ± .008E-5	**	6.12 ± .09E-6	*	**
9	9.85 ± .05E-7	1.17 ± .03E-8	6.4 ± .1E-8	*	*
10	5.79 ± .04E-7	3.8 ± .3E-9	4.5 ± .3E-9	*	*

\# Samples taken from H&V systems. Data extrapolated to containment using appropriate dilution factors.

* Radionuclide not detected.

** Radionuclide not measured. Delay between sampling and counting was too long to detect radionuclide.

2.5). The measured concentrations increased with time, reached maxima, and then decreased (cf Figure 3). The time at which the maximum was reached depended on the half-life of the radioiodine (the longer the half-life, the later the maximum was reached). Iodine-132 reached a maximum concentration about 5.83 hours after reflood; ^{135}I peaked between between 5.83 and 7.55 hours after reflood; ^{133}I peaked about 7.55 hours after reflood; ^{131}I reached a maximum concentration approximately 31.28 hours after reflood. After reaching their maxima, and prior to high volume containment purge, the concentrations of all the radioiodine species decreased faster than would be expected due to radioactive decay alone. This indicates that there was a removal mechanism (i.e., deposition onto surfaces) for the airborne radioiodines.

Table 2 also shows that iodine continued to be released from the LOFT RCS and/or BST long after the test. This is indicated by the fact that the high volume purge did not remove all the airborne radioiodines from the containment air. Although the purge changed out a containment volume every 1 to 2 hours, the ^{131}I concentration dropped only by about a factor of 100 after 135 hours of purging. Almost half of this reduction can be accounted for by radioactive decay alone. There must, therefore, be a source of iodine to account for this behavior. Only two possible sources are obvious - iodine that had deposited onto surfaces and later became airborne or leakage from the LOFT RCS and/or BST itself. Deposited iodine is ruled out as being the major source because of the high elemental fraction of the ^{131}I after the purge began (of Sect. 3.2). Elemental iodine tends to convert to an organic form with time and resuspended iodine is normally highly organic in form (see Appendix B for a discussion of iodine reaction mechanisms)

The data in Table 2 also indicate that ^{132}I was measured long after radioactive decay should have reduced its concentration to below detection limits. This indicates that the ^{132}I in the containment air was coming from ^{132}Te (78.2 hour half-life). The ^{132}Te was decaying to ^{132}I and this ^{132}I was then released into the containment air.

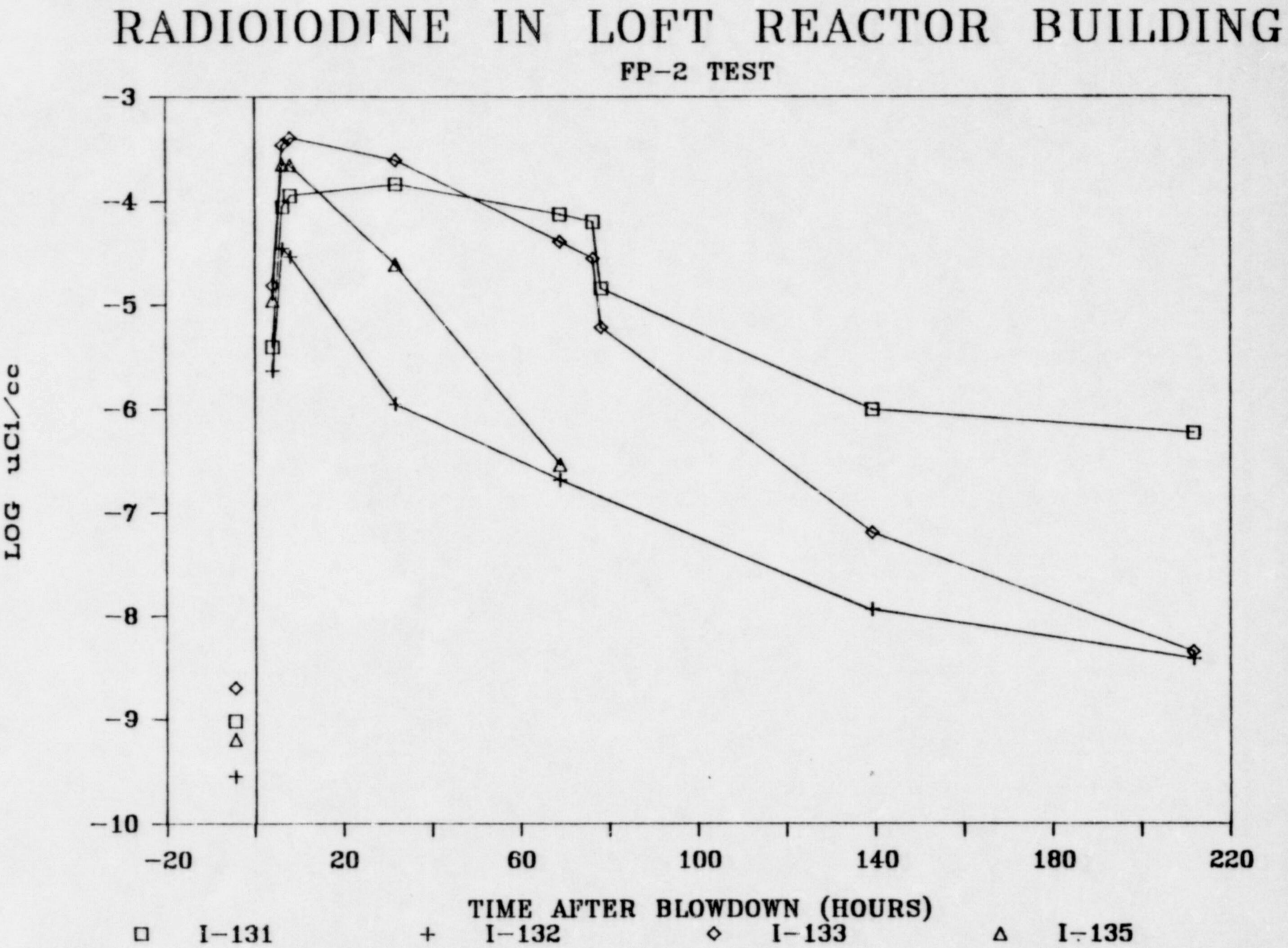

FIGURE 3. RADIOIODINE IN LOFT CONTAINMENT AIR

3.2 Radioiodine Species

Table 3 presents the LOFT containment air radioiodine species mixture measured in each sample. Note that the results of the 42.92h sample appear to be anamalous. The actual species concentrations measured in the H&V systems are contained in Appendix Table A-4. Figure 4 shows a plot of the behavior of the elemental (I_2) and organic fractions of ^{131}I as a function of time. The elemental fraction was high (~70%) early after reflood, dropped with time until it reached about 20% approximately 40 hours after reflood, remained at this level until high volume containment purge was initiated, and then returned to a high value of about 60% (approximately the level measured prior to initiation of the FP-2 test). The organic fraction, on the other hand, started low (~12%), rose with time until it reached about 65% approximately 40 hours after reflood, remained at this level until high volume purge began, and then dropped to about 15% (approximately the level measured prior to the FP-2 test).

The ^{131}I particulate fraction was about 7% in the first sample obtained after reflood. It then dropped with time, reaching a minimum of about 0.3% just prior to initiation of the high volume containment purge. After purge began, the particulate fraction then rose continually and reached about 9% in the last sample taken. The HOI fraction exhibited less dramatic behavior. It started at about 12% in the first sample after reflood, rose to a maximum of 21% at 7.5 hours after reflood, dropped with time and reached a minimum of 9% several hours after high volume purge began, and then rose to the 15-21% range again.

All the species of ^{133}I and ^{135}I behaved in a similar fashion as the corresponding species of ^{131}I. Behavior of the particulate and HOI fractions of ^{132}I was similar to that of the particulate and HOI fractions of ^{131}I. The elemental and organic ^{132}I species, however, exhibited behavior different from that of ^{131}I. Figure 5 shows that the ^{132}I elemental fraction dropped only slightly with time after reflood and then rose back to about its previous level after high volume containment purge began. The ^{132}I organic fraction rose slightly with time after reflood and then dropped back to about its previous value after purge began.

TABLE 3 MEASURED RADIOIODINE SPECIES PERCENTAGES+

Sample (Time after test)	Iodine Species	Radioiodine Species Mixture (percent)			
		^{131}I	^{132}I	^{133}I	^{135}I
Pretest-1	Part	6 ± 2	<20	10 ± 2	25 ± 19
	I_2	55 ± 7	100	63 ± 4	75 ± 43
	HOI	5 ± 2	<16	0.7 ± .1	<30
	Organic	34 ± 5	<41	26 ± 3	<54
Pretest-2	Part	4 ± 2	<15	1.6 ± .7	<32
	I_2	58 ± 12	100	64 ± 5	100
	HOI	20 ± 4	<53	22 ± 3	<43
	Organic	18 ± 4	<27	13 ± 2	<64
1	Part	6.6 ± .2	6.6 ± .3	6.5 ± .2	6.6 ± .2
(3.65 h)	I_2	70 ± 1	71 ± 2	70 ± 2	69 ± 1
	HOI	11.8 ± .2	10.9 ± .6	12.1 ± .2	12.2 ± .3
	Organic	12.0 ± .3	11.1 ± .5	11.8 ± .3	11.3 ± .3
2	Part	2.9 ± .1	2.7 ± .1	2.7 ± .1	2.6 ± .1
(5.83 h)	I_2	64 ± 2	65 ± 2	63.9 ± .8	64.1 ± .6
	HOI	16 ± 1	19.3 ± .7	17.6 ± .2	17.4 ± .2
	Organic	17.0 ± .4	13.3 ± .5	15.8 ± .2	15.9 ± .2

TABLE 3 (Con't) MEASURED RADIOIODINE SPECIES PERCENTAGES+

Sample	Iodine	Radioiodine Species Mixture (percent)			
(Time after test)	Species	^{131}I	^{132}I	^{133}I	^{135}I
3	Part	2.2 ± .1	1.8 ± .1	2.1 ± .1	2.0 ± .1
(7.55 h)	I_2	57 ± 1	63 ± 2	58.9 ± .8	59.7 ± .5
	HOI	21.0 ± .4	19.6 ± .5	20.0 ± .2	19.7 ± .2
	Organic	20.0 ± .3	15.9 ± .4	19.0 ± .2	18.6 ± .2
4	Part	1.04 ± .01	*	1.00 ± .02	1.06 ± .05
(31.28h)	I_2	45.2 ± .7	*	45.1 ± .4	45.7 ± .9
	HOI	16.5 ± .2	*	16.3 ± .2	16.4 ± .4
	Organic	37.3 ± .5	*	37.6 ± .6	36.8 ± .7
5	Part	8.0 ± .1	12 ± 3	7.8 ± .1	7.6 ± .4
(42.92h)	I_2	22.1 ± .2	<19	22.3 ± .3	24 ± 1
	HOI	5.7 ± .1	9 ± 3	6.1 ± .1	6.8 ± .4
	Organic	64.2 ± .8	79 ± 21	63.8 ± .7	62 ± 3
6	Part	0.26 ± .04	<2	0.44 ± .04	<4
(68.32h)	I_2	24.1 ± .2	56 ± 7	24.7 ± .4	30 ± 4
	HOI	13.7 ± .2	17 ± 3	13.7 ± .3	8 ± 2
	Organic	61.9 ± .6	27 ± 4	61.1 ± .9	62 ± 8

TABLE 3 (Con't) MEASURED RADIOIODINE SPECIES PERCENTAGES+

Sample (Time after test)	Iodine Species→	Radioiodine Species Mixture (percent)			
		^{131}I	^{132}I	^{133}I	^{135}I
7	Part	0.64 ± .01	*	0.64 ± .02	*
(75.82h)	I_2	23.5 ± .2	*	24.0 ± .6	*
	HOI	9.8 ± .1	*	9.8 ± .2	*
	Organic	66.1 ± .8	*	66 ± 2	*
8	Part	2.13 ± .04	*	2.2 ± .1	*
(77.67h)	I_2	30.2 ± .4	*	31 ± 1	*
	HOI	9.1 ± .3	*	7.5 ± .2	*
	Organic	58.6 ± .8	*	59 ± 2	*
9	Part	3.62 ± .09	4.6 ± .5	3.8 ± .3	**
(138.75h)	I_2	57.0 ± .7	64 ± 4	55 ± 3	**
	HOI	21.0 ± .3	21 ± 2	21.9 ± .9	**
	Organic	18.4 ± .3	10.4 ± .6	19 ± 1	**
10	Part	9.2 ± .2	3 ± 2	8 ± 2	**
(211.18h)	I_2	60 ± 1	71 ± 12	61 ± 9	**
	HOI	15.2 ± .3	18 ± 4	17 ± 3	**
	Organic	15.1 ± .3	8 ± 3	14 ± 2	**

\+ Species mixture looks suspicious (see Sect. 3.2 text)

* Radionuclide not measured. Delay between sampling and counting was too long to detect radionuclide.

** Radionuclide not detected.

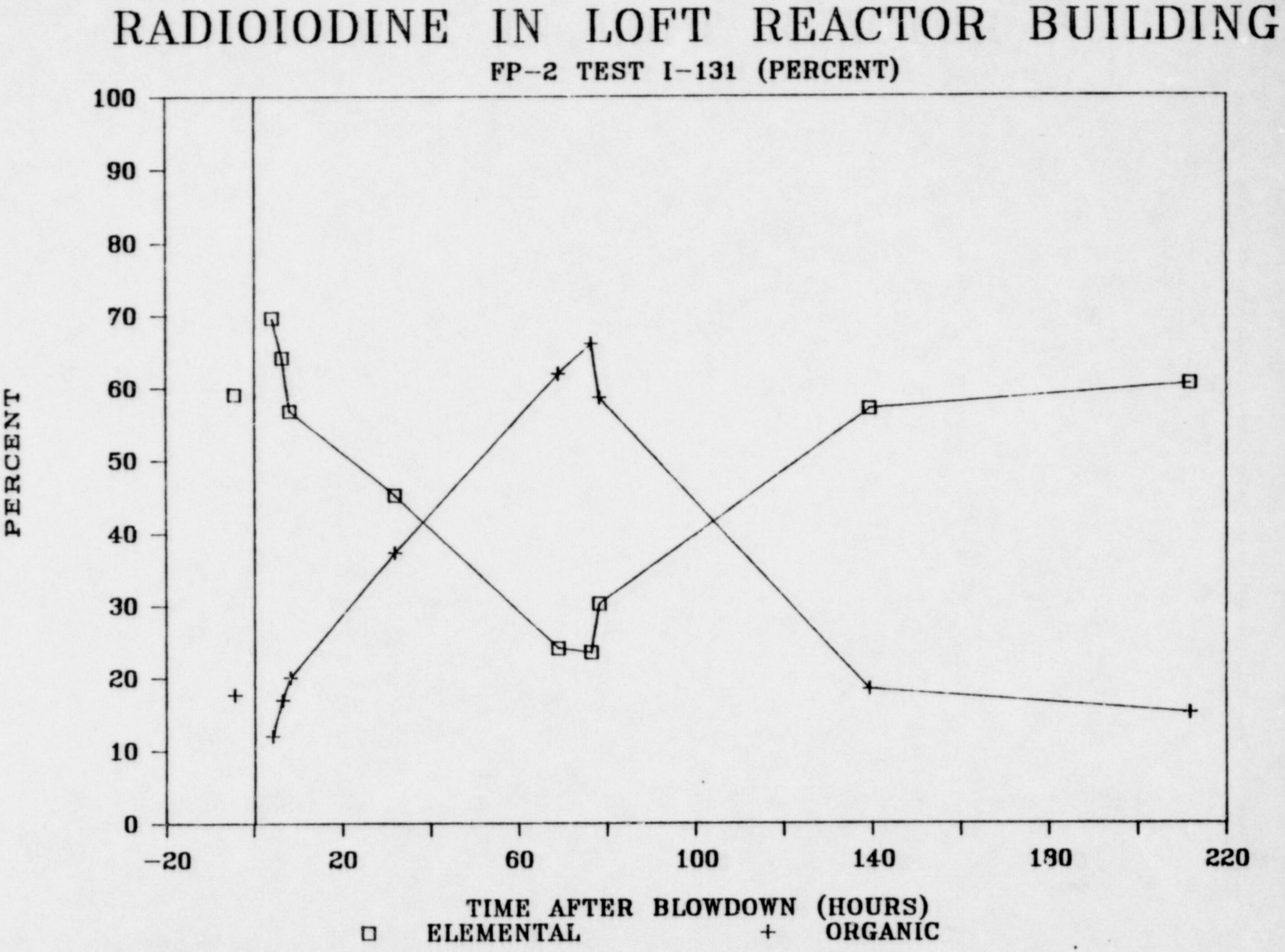

FIGURE 4. ELEMENTAL & ORGANIC ^{131}I FRACTIONS

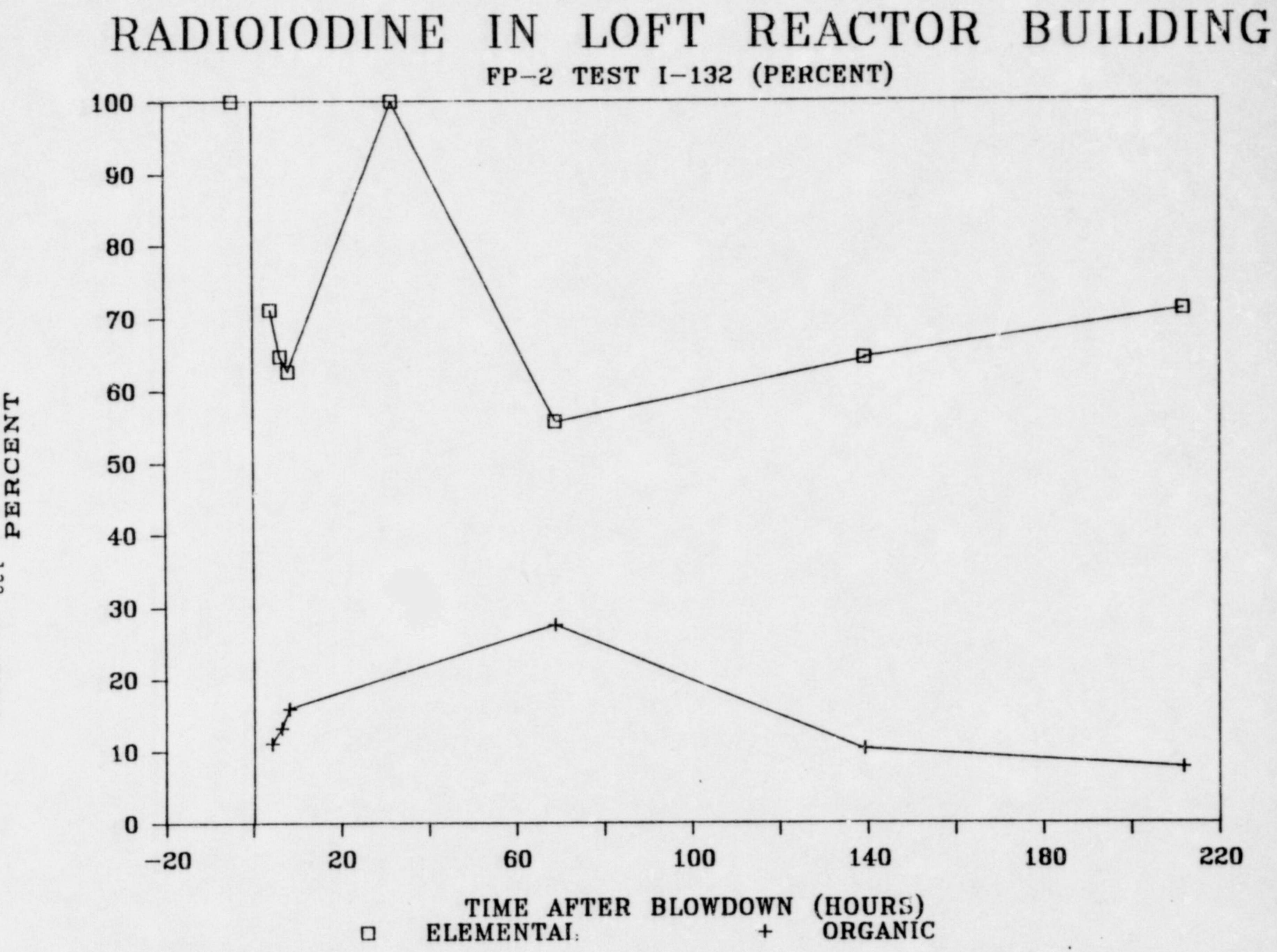

FIGURE 5. ELEMENTAL AND ORGANIC ^{132}I FRACTIONS

Figures 6 and 7 show the dramatic difference in the behavior of the elemental and organic species in ^{131}I and ^{132}I. The reasons for this difference are (1) the half-lives of ^{131}I and ^{132}I (8.04 days and 2.30 hours, respectively), (2) the fact that the ^{132}I was coming from ^{132}Te (78.2 hours half-life), and (3) because the iodine that was released into the containment air began in predominently an elemental form and was converted to an organic form with time. Initially, both ^{131}I and ^{132}I were released into the containment air and this release continued, but at a much lower rate. Upon release into the containment air, both ^{131}I and ^{132}I were highly elemental. With time the ^{131}I was converted to an organic form. The ^{132}I, however, decayed before much of it could be converted to an organic form, The observed ^{132}I was fresh (i.e., recently released, mostly elemental) ^{132}I.

When the high volume purge was put into operation, the ^{131}I became highly elemental. This occurred because the older (mostly organic) ^{131}I was swept out rapidly and the freshly released elemental ^{131}I was also swept out before it had a chance to be converted to organic.

Figures 6 and 7 also show that after high volume containment purge was initiated, the elemental and organic fractions of ^{131}I approached but never reached that of ^{132}I. The elemental fraction of ^{131}I remained lower while the organic fraction remained higher than those of ^{132}I. This behavior is attributed to resuspension (from surfaces) of organic ^{131}I. The ^{132}I had only one source - fresh (highly elemental) ^{132}I from the LOFT RCS and/or BST. The ^{131}I, however, had two sources - fresh (highly elemental) ^{131}I from the LOFT system and old (highly organic) ^{131}I resuspended from surfaces on which elemental ^{131}I had deposited earlier.

The iodine species data provide more evidence that the elemental radioiodines were undergoing deposition with time. Figure 8 shows the behavior of elemental and organic ^{131}I as a function of time. Prior to inititaion of the high volume purge, both species exhibited an increase in concentration, peaking at 31 hours after reflood. The organic concentration, however, increased faster than did the elemental

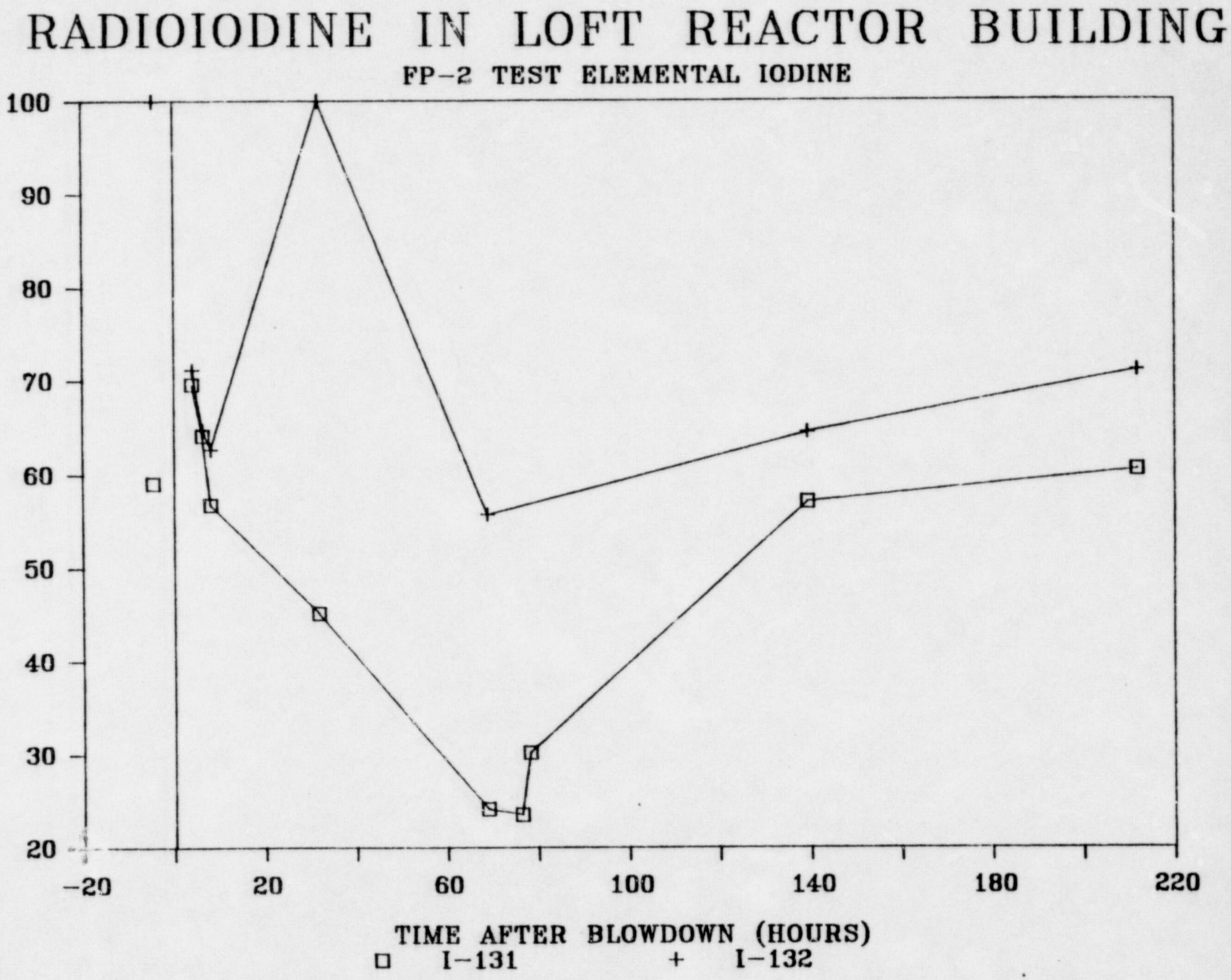

FIGURE 6. ELEMENTAL ^{131}I AND ^{132}I FRACTIONS

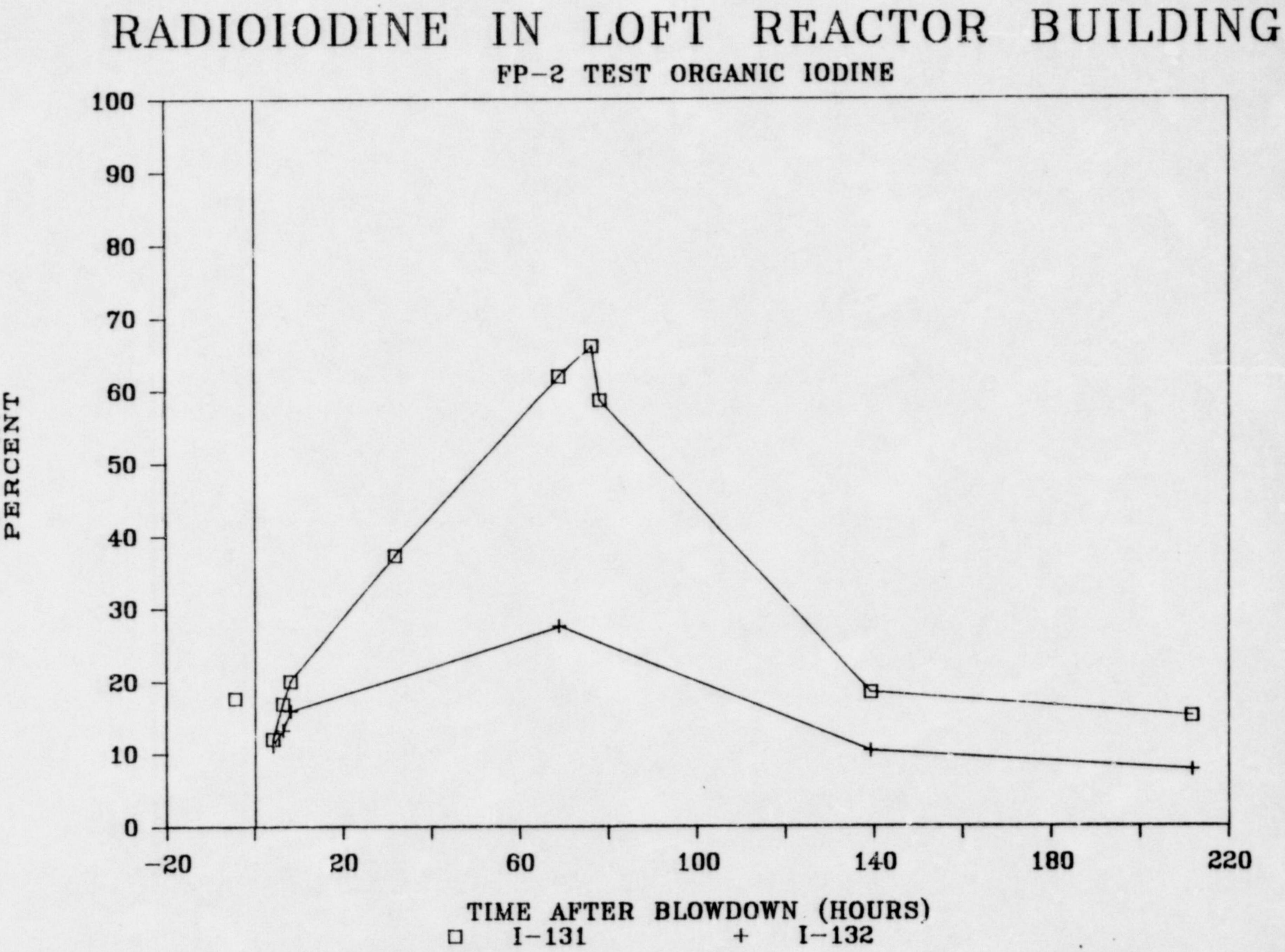

FIGURE 7. ORGANIC ^{131}I AND ^{132}I FRACTIONS

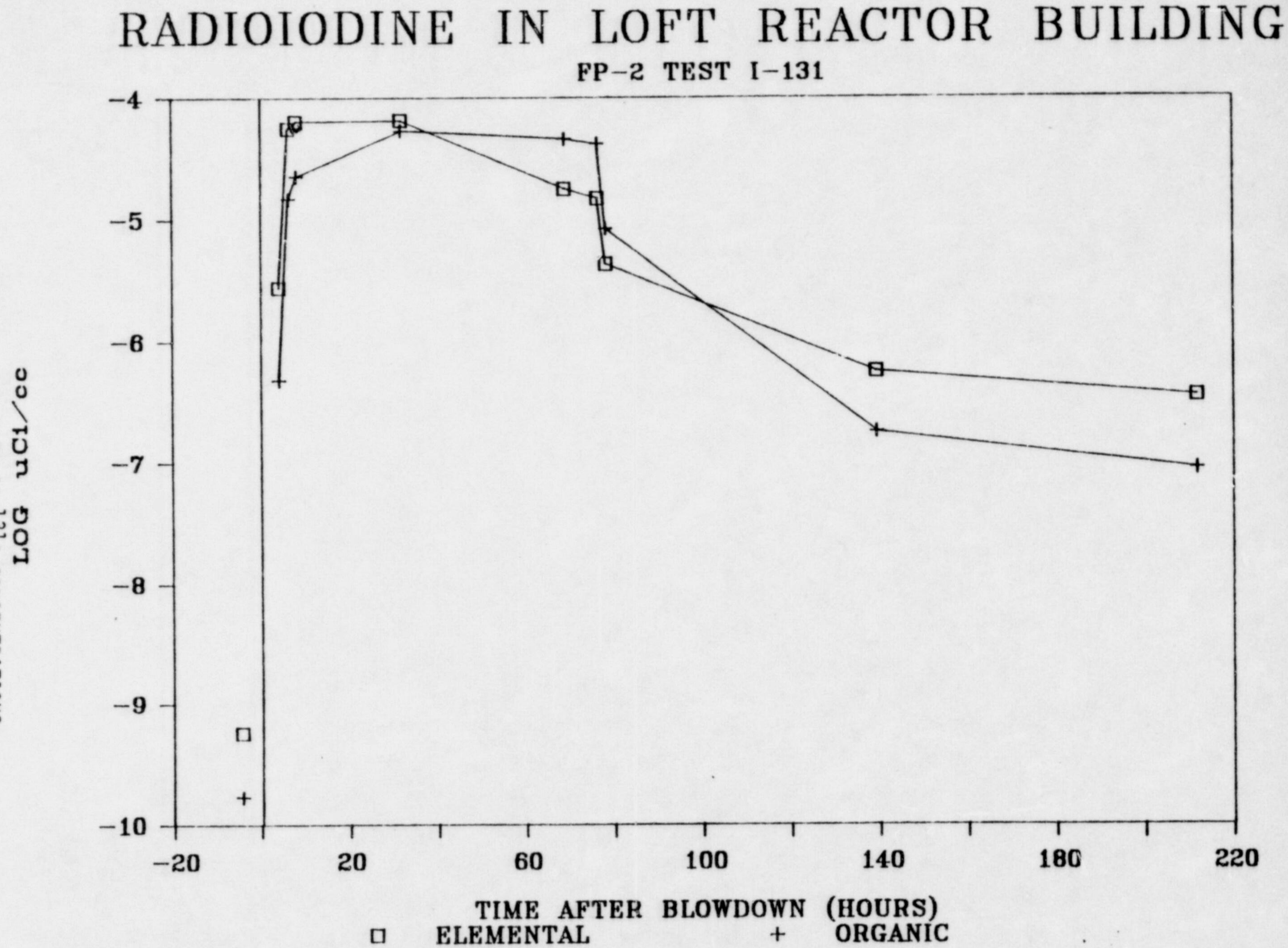

FIGURE 8. ELEMENTAL AND ORGANIC ^{131}I CONCENTRATIONS

concentration. After peaking, both concentrations decreased, but at vastly different rates. While the elemental concentration decreased at a rate much faster than can be accounted for by radioactive decay, the organic concentration decreased at a much slower rate (approximately equal to the natural decay rate). The elemental ^{131}I, therefore, was removed from the containment air (probably by deposition) more rapidly than was the organic ^{131}I. Both ^{133}I and ^{135}I exhibited this same behavior. Iodine-132 did not exhibit this behavior due to its short half-life, i.e., it decayed before it could deposit.

The results of the 42.92h sample were not included in any of the above analyses as it provided suspicious results. This may have been due to operations that were performed in containment prior to the time the sample was collected.

3.3 Particulates

In addition to the radioiodines, nine other radionuclides (i.e., ^{91}Sr, ^{92}Sr, ^{91m}Y, ^{92}Y, ^{136}Cs, ^{137}Cs, ^{138}Cs, ^{140}Ba, and ^{140}La) were detected on the particulate filter. The concentrations of these particulate radionuclides measured in the H&V systems are presented in Appendix Table A-5. To convert these values to concentrations in containment air, they must be multiplied by the dilution factors in Appendix Table A-3.

Using the measured ^{137}Cs, ^{131}I, ^{132}I, ^{133}I, and ^{135}I concentrations observed on the particulate filter and the half-lives of the radionuclides, a cesium-to-iodine ratio was determined. The calculations resulted in cesium-to-iodine ratios of 10, 8, 2.6, 2.6 for the 5.83h, 7.55h, 31.18h, and 75.82h samples, respectively. These high ratios do not preclude the particulate iodine being CsI. They do, however, indicate that there was more cesium than could be accounted for if it was all in the form CsI.

3.4 Cleanup Filter Decontamination Factor

The H&V systems contain silver zeolite cleanup filters for iodine removal. H&V 8 contains two cleanup filters, only one of which is operated at a time. H&V 9 contains one cleanup filter. The DF of each cleanup filter was evaluated once (cf Samples 3,4, and 7).

Table 4 presents the results of the DF measurements. In all cases the DF for elemental iodine was the highest, with the DFs for the other species being lower. The cleanup filter on H&V 9 exhibited approximately the same DF for particulate iodine as did filter 12 on H&V 8. The DFs for all other species were much higher for the filter on H&V 9 then filter 12 on H&V 8. Filter 21 on H&V 8 exhibited lower DFs than the filter on H&V 9 regardless of species.

All the cleanup filters exhibited lower DFs than were expected. The filter on H&V 9 had been tested about one month before the FP-2 test and had exhibited a DF of 39,000 for CH_3I. Filter 12 and 21 on H&V 8 had exhibited DFs of approximately 1200 to 1500 for CH_3I about 9 months before the FP-2 test.

In the course of the DF analysis an anomaly was discovered. The amount of radioiodine on the backup filter is always examined for each sample to insure that there was no radioiodine breakthrough. It was noticed that the amount of radioiodine on the backup filter was approximately the same for samples obtained both upstream and downstream of the H&V cleanup filter (even though the iodine concentration in the inlet stream was 2 to 3 orders of magnitude higher than in the outlet stream). In addition, the ratio of the radioiodine on the backup filter to that on the main AgX filter was several hundreths of a percent for samples obtained upstream of the H&V cleanup filter but was several percent for samples obtained downstream of the H&V cleanup filter. The above indicate that there may be an iodine species (probably a higher order organic than CH_3I) that is not effectively removed by AgX or charcoal.

TABLE 4 DECONTAMINATION FACTORS MEASURED FOR LOFT AgX CLEANUP FILTERS

Sample	H&V System	Iodine Species	Decontamination Factor ^{131}I	^{132}I	^{133}I	^{135}I
3	8+	Part	1.2 ± .2E3	>3.5E2	1.29 ± .09E3	>6.8E2
		I_2	2.7 ± .1E3	2.8 ± .7E3	3.21 ± .09E3	3.1 ± .2E3
		HOI	4.6 ± .1E1	5.8 ± .8E1	4.65 ± .09E1	4.6 ± .1E1
		Organic	6.0 ± .1E1	5.9 ± 1.0E1	6.1 ± .1E1	5.9 ± .1E1
		Total	1.22 ± .02E2	1.6 ± .2E2	1.31 ± .02E2	1.32 ± .02E2
4	8++	Part	3.5 ± .3E2	*	3.3 ± .3E2	>6.0E1
		I_2	3.0 ± .1E3	*	3.1 ± .3E3	>1.4E3
		HOI	4.56 ± .07E1	*	4.6 ± .1E1	4.2 ± .6E1
		Organic	6.26 ± .06E1	*	6.5 ± .1E1	6.0 ± .8E1
		Total	1.03 ± .01E2	*	1.05 ± .01E2	1.0 ± .1E2
7	9	Part	1.1 ± .4E3	*	>1.1E3	*
		I_2	>2.1E5	*	>1.4E4	*
		HOI	3.8 ± .2E3	*	4 ± 2E3	*
		Organic	1.07 ± .02E3	*	1.05 ± .07E3	*
		Total	1.43 ± .03E3	*	1.43 ± .09E3	*

\+ Cleanup filter no. 12 was in use.

++ Cleanup filter no. 21 was in use.

* Decontamination factor not measured. Time between sampling and counting was too long to detect radionuclide.

4. SUMMARY AND CONCLUSIONS

Samples of containment air were obtained before and as a function of time after the LOFT FP-2 test. Ten samples were obtained during the period 3.65 to 211.18 hours after the test. For all radioiodine isotopes (i.e.. ^{131}I,^{132}I,^{133}I,^{135}I), the measured concentrations increased with time, reached maxima, and then decreased with time. Maximum concentrations reached were 1.45E-4 μCi/cc, 3.52E-5 μCi/cc, 4.04E-4μCi/cc, and 2.23E-4μCi/cc for ^{131}I, ^{132}I, ^{133}I, and ^{135}I, respectively. Based on a containment free volume of about 1E10 cc, the maximum radioiodine activity in the containment air was about 1-1/2 Ci, 1/3 Ci, 4 Ci, and 2-1/4 Ci for ^{131}I, ^{132}I, ^{133}I, and ^{135}I respectively. This is a small fraction of the radioiodine inventory present in the fuel at the time of the LOFT test (e.g., 4.75E4 Ci of ^{131}I and 2.147E5 Ci of ^{133}I).

Species measurements indicated that the radioiodine species mixture changed with time. Prior to the LOFT test, when high volume purge was rapidly sweeping any released radioiodines out of the containment, the radioiodines were predominantly elemental (~60%), with smaller components of organic (~15%), HOI (~20%), and particulate (a few percent). Shortly after reflood terminated the LOFT test, the elemental fraction of all the observed radioiodines except ^{132}I rose to about 70%, dropped with time until it reached about 20-25% prior to high volume containment purge, and then returned to approximately the level (~60%) measured prior to the LOFT test. The organic fraction, on the other hand, was low (~15%) prior to the test, dropped to about 12% shortly after reflood, rose with time until it reached a maximum of about 65% prior to high volume purge, and then dropped to about 15% (approximately the level measured prior to the LOFT test).

This behavior was caused by three factors; (1) a continuous release of radioiodines (primarily in the elemental form) into the containment atmosphere, (2) deposition of elemental iodine onto surfaces, and (3) conversion of elemental iodine to an organic form. Initially during the LOFT test, radioiodines were released into the containment air. This release continued, but at a lower rate, long after the LOFT test ended (at least as long as the containment air samples were taken). The released iodine was primarily in the elemental form. With time, the highly reactive elemental species underwent deposition onto surfaces and chemical reactions that converted it to an organic form. When high volume containment purge was initiated, most of the older radioiodines were rapidly swept out, and the detected radioiodines were those that were continuously being released from the LOFT system.

The elemental and organic ^{132}I species, however, did not exhibit behavior like that of the other radioiodine isotopes. Both changed only slightly with time after reflood, the elemental fraction remaining high (~60-70%) and the organic fraction remaining low (~10-25%). This occurred because the ^{132}I was continuously being produced by decay of ^{132}Te (78.2 hour half-life) and was continuously being released into the containment atmosphere in a highly elemental form (like the other radioiodine isotopes). The ^{132}I, however, underwent radioactive decay before much of it could be converted to an organic form.

The particulate fraction of all the radioiodines was about 7% in the first sample obtained 3.65 hours after reflood. It then dropped, reaching a minimum of about 0.3% just prior to initiation of high volume containment purge, then rose to about 5-9% (depending on isotope) during high volume purge.

Behavior similar to that of the elemental and organic species of ^{131}I and the other long-lived radioiodines at LOFT has been observed at operating nuclear power plants. High elemental and low organic ^{131}I fractions have been observed in the air near primary coolant sampling systems [4-6], indicating that freshly released radioiodine is primarily in the elemental form. Evidence that the elemental species converts with time to organic species comes from observations that (1) the elemental fraction decreased while the organic fraction increased with time after containment was isolated and purging ceased [7,8] and (2) the radioiodine in ventilation air far removed from the source of radioiodine leakage was highly organic.[4,6]. In contrast to the results at LOFT, the HOI fraction observed at operating nuclear power plants was sometimes much higher. [5-8] At times it was the predominent species.

Evidence that airborne radioiodine undergoes deposition comes from observations that the ratio of ^{131}I to ^{3}H was much lower (even after correction for radioactive half-life) in containment air than it was in the reactor coolant which was the source of both. [7-9] Resuspension of radioiodine was observed at TMI-2 when the airborne ^{129}I concentration increased to approximately its previous level after purging ceased. [10] A very simple model developed to predict airborne radioiodine deposition and resuspension predicts that about 90% of the ^{131}I will be on surfaces when equilibrium is reached. [11]

In situ measurements of the DF for LOFT H&V system silver zeolite cleanup filters indicated that the DF for elemental iodine was the highest, with the DFs for the particulate, HOI, and organic species being lower. The DFs were relatively low (about 1000-3000 for particulate, 3000 for I_2, and 50-60 for HOI and organic) for the cleanup filters in H&V 8. For the cleanup filter in H&V 9, the DF for particulates was similar (~1000) but those for the other species were much higher (>100,000 for I_2, about 4000 for HOI, and 1100 for organic). Because of the iodine species admixtures at the time of the measurements, the effective total DFs were 100-120 for the H&V 8 filter and 1400 for the H&V 9 filter.

The DF measurements also indicated that there may be a species of iodine, probably a heavy organic, that is not effectively removed by silver zeolite filters. Charcoal filters probably are no more effective for this species than are silver zeolite filters.

Only several particulate radionuclides besides the radioiodines were detected in the containment air after the LOFT FP-2 test. Maximum airborne particulate concentrations observed in containment air were $10^{-8}\mu Ci/cc$ for $^{136,137}Cs$; $10^{-7}\mu Ci/cc$ for ^{92}Sr, ^{92}Y, ^{138}Cs, and ^{140}La; and $10^{-6}\mu Ci/cc$ for ^{91}Sr, ^{91m}Y, and ^{140}Ba. The presence of ^{140}Ba-^{140}La was not expected and may indicate that the LOFT fuel got quite hot.

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APPENDIX A. SAMPLE DATA

Sample data obtained from H&V systems during the measurements of the radioiodines in LOFT containment air are presented in the following tables.

TABLE A-1

H&V HISTORY

Date	Time	Action
7/09/85	1409	H&V 9 secured.
	1758	H&V 8 turned on. Filter unit 21 used.
	1814	H&V 8 secured.
	2019	H&V 8 turned on. Filter unit 21 used.
7/10/85	0145	H&V 8 secured.
	1833	H&V 8 turned on. Filter unit 12 used.
7/11/85	1107	H&V 8 secured.
	1655	H&V 8 turned on. Filter unit 12 used.
7/12/85	1820	H&V 9 turned on recirc. with partial exhaust to stack (1200 cfm). H&V 8 still on.
7/13/85	0215	H&V 9 full exhaust to stack. H&V 8 secured.

TABLE A-2 RADIOIODINE CONCENTRATION IN LOFT H&V SYSTEMS

Sample	Measured Radioiodine Concentration (uCi/cc)				
	^{131}I	^{132}I	^{133}I	^{134}I	^{135}I
Pretest-1	1.38 ± .08E-9	8.6 ± 2.5E-10	4.6 ± .2E-9	<7.3 E-9	1.9 ± .7E-9
Pretest-2	9.7 ± .8E-10	2.8 ± 1.0E-10	2.02 ± .08E-9	<7.3 E-10	6.4 ± 3.0E-10
1	4.18 ± .03E-7	2.45 ± .03E-7	1.60 ± .02E-6	4.3 ± 1.1E-8	1.16 ± .01E-6
2	1.37 ± .02E-5	5.50 ± .07E-6	5.35 ± .03E-5	<9.9 E-7	3.49 ± .01E-5
3a#	1.55 ± .02E-5	4.02 ± .04E-6	5.54 ± .03E-5	<5.7 E-7	3.04 ± .01E-5
3b#	1.26 ± .01E-7	2.5 ± .2E-8	4.23 ± .04E-7	*	2.31 ± .03E-7
4a	2.84 ± .02E-5	2.2 ± .9E-7	4.80 ± .02E-5	*	4.78 ± .04E-6
4b	2.77 ± .02E-7	<9.4 E-9	4.55 ± .03E-7	*	4.8 ± .4E-8
5	7.85 ± .04E-6	3.0 ± .4E-8	9.46 ± .04E-6	*	4.21 ± .07E-7
6	6.62 ± .03E-6	1.8 ± .1E-8	3.65 ± .02E-6	*	2.6 ± .1E-8
7a	6.33 ± .03E-5	**	2.80 ± .03E-5	*	**
7b	4.42 ± .06E-8	**	2.0 ± .1E-8	*	**
8	1.434 ± .008E-5	**	6.12 ± .09E-6	*	**
9	9.85 ± .05E-7	1.17 ± .03E-8	6.4 ± .1E-8	*	*
10	5.79 ± .04E-7	3.8 ± .3E-9	4.5 ± .3E-9	*	*

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a denotes sample upstream of H&V cleanup filter; b denotes sample downstream of H&V cleanup filter.

* Radionuclide not detected.

** Radionuclide not measured. Delay between sampling and counting was too long to detect radionuclide.

TABLE A-3 DILUTION FACTORS

Sample	Dilution Factor *
Pretest-1	1.0
Pretest-2	1.0
1	9.6
2	6.4
3	7.3
4	5.1
5	7.9
6	11.2
7	1.0
8	1.0
9	1.0
10	1.0

* Factor used to multiply concentration measured in H&V system to obtain concentration in containment air.

TABLE A-4 RADIOIODINE SPECIES CONCENTRATIONS MEASURED IN LOFT H&V SYSTEMS

Sample	Iodine Species	Radioiodine Species Concentration (uCi/cc) in H&V Sample				
		^{131}I	^{132}I	^{133}I	^{134}I	^{135}I
Pretest-1	Part	8.4 ± 2.7E-11	<1.7E-10	4.7 ± .8E-10	<1.2E-9	4.7 ± 1.8E-1
	I_2	7.6 ± .5E-10	8.6 ± 1.4E-10	2.9 ± .1E-9	<2.8E-9	1.4 ± .3E-9
	HOI	6.4 ± 2.8E-11	<1.4E-10	3.1 ± .5E-11	<1.5E-9	<5.6E-10
	Organic	4.7 ± .4E-10	<3.5E-10	1.21 ± .08E-9	<6.5E-9	<1.0E-9
Pretest-2	Part	3.5 ± 1.5E-11	<4.3E-11	3.2 ± 1.3E-11	<7.9E-11	<2.0E-10
	I_2	5.7 ± .7E-10	2.8 ± .5E-10	1.29 ± .06E-9	<2.7E-10	6.4 ± 1.3E-
	HOI	1.9 ± .2E-10	<1.5E-10	4.4 ± .4E-10	<1.8E-10	<2.8E-10
	Organic	1.7 ± .2E-10	<7.8E-11	2.6 ± .3E-10	<6.4E-10	<4.1E-10
1	Part	2.76 ± .06E-8	1.65 ± .06E-8	1.04 ± .02E-7	<5.3E-9	7.7 ± .1E-8
	I_2	2.91 ± .03E-7	1.75 ± .03E-7	1.11 ± .02E-6	4.3 ± .5E-8	8.07 ± .09E-
	HOI	4.91 ± .06E-8	2.7 ± .1E-8	1.94 ± .02E-7	<1.3E-8	1.42 ± .02E-
	Organic	5.02 ± .08E-8	2.73 ± .09E-8	1.89 ± .03E-7	<1.5E-8	1.37 ± .03E-
2	Part	3.97 ± .03E-7	1.47 ± .05E-7	1.43 ± .02E-6	<3.9E-8	9.1 ± .1E-7
	I_2	8.8 ± .1E-6	3.56 ± .06E-6	3.42 ± 0.3E-5	<7.6E-7	2.24 ± .01E-
	HOI	2.2 ± .2E-6	1.06 ± .03E-6	9.40 ± .06E-6	<4.4E-7	6.07 ± .04E-
	Organic	2.32 ± .02E-6	7.3 ± .2E-7	8.47 ± .08E-6	<4.5E-7	5.54 ± .04E-

TABLE A-4 (Con't) RADIOIODINE SPECIES CONCENTRATIONS MEASURED IN LOFT H&V SYSTEMS

Sample	Iodine Species	^{131}I	^{132}I	^{133}I	^{134}I	135
		Radioiodine Species Concentration (uCi/cc) in H&V Sample				
3a	Part	3.42 ± .04E-7	7.1 ± .2E-8	1.154 ± .007E-6	<2.3E-8	6.18 ± .04E
	I_2	8.8 ± .1E-6	2.52 ± .04E-6	3.26 ± .03E-5	<5.4E-7	1.815 ± .0
	HOI	3.25 ± .03E-6	7.9 ± .1E-7	1.106 ± .008E-5	<1.4E-7	5.99 ± .03E
	Organic	3.11 ± .02E-6	6.39 ± .09E-7	1.055 ± .007E-5	<1.4E-7	5.64 ± .03E
3b	Part	2.9 ± .3E-10	<2.0E-10	8.9 ± .6E-10	*	<9.1E-10
	I_2	3.21 ± .08E-9	9.1 ± 2.1E-10	1.02 ± .02E-8	*	5.8 ± .4E
	HOI	7.1 ± .1E-8	1.4 ± .2E-8	2.38 ± .03E-7	*	1.29 ± .02E
	Organic	5.13 ± .05E-8	1.1 ± .1E-8	1.71 ± .03E-7	*	9.5 ± .1E-
4a	Part	2.96 ± .02E-7	<8.7E-9	4.81 ± .08E-7	*	5.1 ± .2E
	I_2	1.28 ± .01E-5	2.2 ± .6E-7	2.17 ± .01E-5	*	2.18 ± .03E
	HOI	4.68 ± .04E-6	<8.5E-8	7.84 ± .05E-6	*	7.8 ± .1E
	Organic	1.059 ± .008E-5	<9.7E-8	1.80 ± .01E-5	*	1.76 ± .02E
4b	Part	8.3 ± .5E-10	<2.2E-9	1.4 ± .1E-9	*	<8.4E-10
	I_2	4.3 ± .1E-9	<3.8E-9	6.9 ± .2E-9	*	<1.6E-9
	HOI	1.027 ± .008E-7	<1.3E-8	1.70 ± .03E-7	*	1.9 ± .2E
	Organic	1.66 ± .01E-7	<1.8E-8	2.73 ± .03E-7	*	2.9 ± .3E

TABLE A-4 (Con't) RADIOIODINE SPECIES CONCENTRATIONS MEASURED IN LOFT H&V SYSTEMS (Con't)

Sample	Iodine Species	Radioiodine Species Concentration (uCi/cc) in H&V Sample				
		^{131}I	^{132}I	^{133}I	^{134}I	^{135}I
5	Part	6.27 ± .06E-7	3.8 ± .5E-9	7.43 ± .08E-7	*	2.50 ± .08E-
	I_2	1.735 ± .007E-6	<5.7E-9	2.11 ± .02E-6	*	1.01 ± .03E-
	HOI	4.48 ± .04E-7	2.7 ± .7E-9	5.75 ± .08E-7	*	2.9 ± .1E-8
	Organic	5.04 ± .04E-6	2.4 ± .3E-8	6.04 ± .04E-6	*	2.59 ± .06E-
6	Part	1.7 ± .3E-8	<4.4E-10	1.6 ± .1E-8	*	<1.1E-9
	I_2	1.597 ± .008E-6	1.03 ± .05E-8	9.03 ± .09E-7	*	7.8 ± .6E-9
	HOI	9.09 ± .09E-7	3.1 ± .4E-9	5.01 ± .06E-7	*	2.0 ± .5E-9
	Organic	4.10 ± .03E-6	5.1 ± 1.1E-9	2.23 ± .02E-6	*	1.6 ± .1E-8
7a	Part	4.04 ± .04E-7	**	1.79 ± .05E-7	*	**
	I_2	1.490 ± .007E-5	**	6.7 ± .1E-6	*	**
	HOI	6.18 ± .05E-6	**	2.75 ± .04E-6	*	**
	Organic	4.18 ± .03E-5	**	1.84 ± .02E-5	*	**
7b	Part	3.5 ± 1.3E-11	**	<1.6E-10	*	**
	I_2	<7.0E-11	**	<4.8E-10	*	**
	HOI	1.61 ± .06E-9	**	7.6 ± 3.3E-10	*	**
	Organic	3.92 ± .06E-8	**	1.7 ± .1E-8	*	**

TABLE A-4 (Con't) RADIOIODINE SPECIES CONCENTRATIONS MEASURED IN LOFT H&V SYSTEMS

		Radioiodine Species Concentration (uCi/cc) in H&V Sample				
Sample	Iodine Species	^{131}I	^{132}I	^{133}I	^{134}I	^{135}I
8	Part	3.05 ± .03E-7	**	1.35 ± .06E-7	*	**
	I_2	4.33 ± .03E-6	**	1.88 ± .05E-6	*	**
	HOI	1.30 ± .03E-6	**	4.60 ± .06E-7	*	**
	Organic	8.40 ± .07E-6	**	3.65 ± .07E-6	*	**
9	Part	3.57 ± .07E-8	5.3 ± .4E-10	2.4 ± .1E-9	*	*
	I_2	5.61 ± .04E-7	7.5 ± .3E-9	3.5 ± .1E-8	*	*
	HOI	2.06 ± .02E-7	2.4 ± .1E-9	1.40 ± .03E-8	*	*
	Organic	1.81 ± .02E-7	1.2 ± .1E-9	1.24 ± .04E-8	*	*
10	Part	5.35 ± .08E-8	1.2 ± .6E-10	3.8 ± .5E-10	*	*
	I_2	3.50 ± .04E-7	2.7 ± .2E-9	2.7 ± .3E-9	*	*
	HOI	8.8 ± .1E-8	6.9 ± 1.0E-10	7.7 ± .7E-10	*	*
	Organic	8.73 ± .09E-8	2.9 ± .8E-10	6.1 ± .7E-10	*	*

* Radionuclide not detected.

** Radionuclide not measured. Delay between sampling and counting too long to detect radionuclide.

TABLE A-5 CONCENTRATIONS (uCi/cc) OF OTHER RADIONUCLIDES MEASURED IN LOFT H&V SYSTEM

	Sample				
Nuclide	Pretest-1	Pretest-2	1	2	3
^{91}Sr	<2.6E-10	<8.2E-11	5.0 ± .4E-9	1.05 ± .05E-7	8.3 ± .2E-8
^{92}Sr	<3.3E-10	<6.6E-11	1.7 ± .4E-9	2.6 ± .4E-8	1.2 ± .1E-8
^{91m}Y	<9.6E-10	<7.2E-11	1.5 ± .5E-8	2.7 ± 1.0E-7	2.4 ± .2E-7
^{92}Y	<1.1E-9	<5.0E-10	4.5 ± 1.5E-9	4.7 ± 1.3E-8	3.8 ± .7E-8
^{136}Cs	<6.5E-11	<5.4E-11	<3.7E-10	<2.9E-9	2.3 ± .5E-9
^{137}Cs	<3.6E-11	<6.1E-11	<4.5E-10	4.2 ± .8E-9	2.7 ± .4E-9
^{138}Cs	<7.0E-9	6.9 ± 1.5E-10	1.6 ± .3E-8	<6.8E-7	<1.7E-7
^{140}Ba	<9.5E-11	<1.1E-10	6.7 ± .4E-9	1.54 ± .04E-7	1.38 ± .03E-7
^{140}La	<4.6E-11	<2.5E-11	<1.1E-9	9.9 ± 1.3E-9	1.23 ± .06E-8

	Sample				
Nuclide	4	5	6	7	8
^{91}Sr	<3.5E-9	<1.8E-9	<3.5E-10	<3.8E-8	<1.6E-7
^{91m}Y	*	2.4 ± 1.2E-8	1.1 ± .5E-9	*	*
^{92}Y	2.1 ± .8E-8	<9.3E-9	<1.9E-9	*	*
^{136}Cs	<7.5E-10	<8.1E-10	<3.7E-11	<2.9E-10	<7.7E-10
^{137}Cs	6.7 ± 1.6E-10	<6.5E-10	<9.9E-11	8.2 ± 1.1E-10	<7.0E-10
^{140}Ba	2.55 ± .09E-8	1.8 ± .1E-8	2.5 ± .4E-9	1.89 ± .08E-8	5.8 ± .9E-9
^{140}La	7.1 ± .6E-9	<1.1E-8	1.4 ± .1E-9	<6.4E-9	*

TABLE A-5 (Con't) CONCENTRATIONS (uCi/cc) OF OTHER RADIONUCLIDES MEASURED IN LOFT H&V SYSTEM

	Sample	
Nuclide	9	10
^{136}Cs	<5.2E-11	<9.6E-11
^{137}Cs	<6.6E-11	<1.2E-10
^{140}Ba	<1.5E-10	3.2 ± 1.2E-10
^{140}La	<9.8E-11	2.3 ± .5E-10

APPENDIX B. IODINE REACTIONS

The source of iodine in the LOFT containment from the FP-2 test can only be surmised. It is not known what fraction of the airborne radioiodine came from leaks in the system during the blowdown and heatup phase of the test where the iodine species could be found at high temperatures, or what fraction came from leaks in the reactor system after reflood and a return to ambient conditions.

Iodine can exist in many oxidation states, -1 to +7. Table B-1 provides a summary of the physical attributes of selected species of iodine.

The three species of non-organic iodine that probably would exist after the evaporation of boric acid containing reactor coolant are HI, I_2, HOI, and some HIO_3. HIO_3 present would be found on the particulate filter of the species sampler. The sampler would also separate I_2 and HOI. However, the retention of HI has not been tested and it is not known which sampler media bed would retain this species.

The following reactions would occur in the containment atmosphere. However, the amounts of each product at any time would depend upon the kinetic rate for each reaction, which is a function of a number of variables (temperature, pressure, concentration, surface, catalysis activation energy, etc.)and the equilibrium concentration for reversible reactions.

TABLE B-1

Physical Properties[12] of Iodine Bearing Substances

Oxidation State	Chemical Formula	Name	Melting Point °C	Boiling Point °C	Solubility In H_2O
-1	HI	Hydroiodic Acid	-50.8	-35.8	Very Soluble
0	I_2	Iodine	113.5	184.35	0.03g//100g @ 25° C
+1	HOI	Hypoiodous Acid	NM	NM	Decomposes
+3					
+5	HIO_3	Iodic Acid	Decomposes @ 110°C		Very Soluble
+7	HIO_4	Periodic Acid	Sublimes @ 110°C		Very Soluble
Covalent Bond	CH_3I	Methyl Iodide	-66.4	42.4	NM
Covalent Bond	CH_3CH_2I	CH_3I Ethyl Iodide	-108	72.3	NM

NM= Not Measured

Reference

$I_2 \overset{h\nu}{\rightleftharpoons} 2I\bullet$ [13]

$I_2 + CH_4 \longrightarrow CH_3I + HI$ [14]

$I_2 + R\text{-}CH_3 \longrightarrow R\text{-}CH_2I + HI$ [14]

Where R is any organic radical (CH_3. C_2H_5, etc.)

$HI + CH_3OH \longrightarrow CH_3I + H_2O$ [14]

$I_2 + H_2 \longrightarrow 2HI$ [13]

$nI_2 + 2M \longrightarrow 2MI_n$ [13]

Where M is almost any metal

There have been a number of reports that have attempted to define the kinetics and interactions of the various iodine forms under expected accident conditions.[15,16,17] Very little information is available on the expected concentrations of airborne HOI or the possible reactions that may occur.

The following reactions of HOI with CH_4 have not been reported in the literature but would be expected to occur based on similar organic reactions of this kind [14].

$$HOI + CH_4 \begin{cases} \longrightarrow CH_3I + H_2O \\ \longrightarrow CH_3OH + HI \end{cases}$$

One other reaction that may be important for the production and loss of HOI that has not been studied is the reaction of I_2 and H_2O in the gas phase as follows:

$$I_2 + H_2O \underset{r}{\overset{f}{\rightleftharpoons}} HI + HOI$$

Either the forward (f) or reverse (r) reaction will be the predominate one which would affect the relative concentration of these three iodine species.

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I tem #2

Iodine Chemical Form in the PBF-SFD Tests

A luminescence microprobe developed specifically for analysis of SFD test samples was used to investigate deposited fission products. Reagent grade CsI was examined under ambient conditions and found to exhibit a strong luminescence spectrum, but with time (min) it collects waters of hydration that quench any luminescence. Heating of the CsI samples restored the luminescence spectrum. The same technique was used to examine SFD 1-3 deposition samples where ^{131}I had been measured. No CsI was identified on the SFD 1-3 samples examined. By the time of the luminescence examination ^{131}I had decayed away, but based on the fact that significant stable and long-lived iodine isotopes are also present in the SFD 1-3 fuel and the assumptions that all iodine behaves similarly, and that the presence of other elements does not cause quenching of the luminescence, significant iodine should have found on the samples at the time of the examination.

Iodine behavior in the SFD-ST was notably different from the other three SFD tests. Iodine was observed to transport readily through the effluent sample system during the high-steam flow conditions of the scoping test, but iodine held up readily during the SFD 1-1, 1-3, and 1-4 tests diminishing greatly the transported fraction. In all four tests the iodine partitioned strongly to the liquid stream leaving the condenser/separator part of the effluent sample system.

Thermochemical equilibrium calculations supported by chemical kinetics calculations have been used to describe the observed behavior of I and Cs in the PBF tests. This analysis suggests that HI is the dominant form of iodine generated in the steam/H_2 effluent in the trace irradiated fresh fuel tests (ST and 1-1) and that HI will convert to CsI in the PBF sample system as the effluent temperatures are lowered. On the other hand, for the tests with higher burnup fuel (SFD 1-3 and 1-4) the realistic I and Cs concentrations may have been high enough to favor CsI formation in the effluent rather than HI, however, this analysis considers only a 4-component system (H-O-Cs-I).