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**FISSION PRODUCT TRANSPORT AND BEHAVIOR
IN THE STAINLESS STEEL LINED CONTAINMENT
RESEARCH INSTALLATION (CPI)**

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OAK RIDGE NATIONAL LABORATORY

operated by

UNION CARBIDE CORPORATION

for the

U.S. ATOMIC ENERGY COMMISSION

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STEEL LINED CONTAINMENT RESEARCH INSTALLATION (CRI)

G. W. Parker, G. E. Creek and W. J. Martin

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ABSTRACT

The tests conducted in the ORNL LOFT Assistance Program to date are summarized for 15 release experiments conducted in the CRI with the stainless steel liner. The experiments are listed in chronological order along with the conditions of the release. The runs included direct release of radioiodine tracer, release of the CSE simulant, fission product release from melted irradiated fuel, and prompt release on cladding failure. These release experiments completed the initial phase of the LOFT Assistance Program, and in this report salient facts are presented concerning those phases of the experiments which are thought to be of most interest. Data concerning conditions of temperature, pressure, condensation, and time in the containment vessel together with the results of gas sampling, particle sizing, spraying, and deposition testing are given for the experiments.

i. INTRODUCTION

The Containment Research Installation (CRI) was constructed at the Oak Ridge National Laboratory for investigating fission-product release, transport, and plateout as a function of burnup, fuel and containment temperature, time at temperature, atmosphere, and containment surface composition.

The CRI is operated largely as a test facility to provide information closely coordinated with the objectives of the 50-Mw pressurized-water-reactor (LOFT) destruction test initiated by Phillips Petroleum Company and continued by the Idaho Nuclear Corporation as part of their STEP project.

Fission product behavior in the containment is the type of investigation for which the CRI is ideally suited. With the exception of the subtle effects produced by highly ionizing radiation, essentially all other aspects of the loss-of-coolant accident are readily simulated. It has been shown in several comparative tests that the change in nature of radioiodine in the containment is subject to rather precise measurement by means of the best sampling and fractionation procedures including the classification of submicron particles.

In order to validate the provisional change in the LOFT containment to a stainless clad liner, it was not only practical but within the capability of CRI to make comparisons of this nature with various coated containment surfaces. It has already been shown that the production of organic forms of radioiodine are dependent upon the nature of the plateout surfaces as well as the presence of certain organic materials in the gas phase. Therefore, these tests are likely to be of great interest.

The experimental work to date (Table 1.1) was first concentrated on the stainless steel system with a fuel melt-down type aerosol. Later in the clad-failure type of release in which there was a significant reduction in total mass of solids, Zircaloy clad fuel was used. It was found normal for radioiodine to remain mobile in the stainless steel environment for extended period; although a far more permanent adsorption would be expected to occur with the painted surfaces. The production of methyl iodide could be followed with high precision and any marked differences between the two systems in future runs will be readily detected.

Types of information specifically sought were:

- (1) Particulate and iodine concentration in atmosphere.
- (2) Fission product concentration in condensate and in rainout droplets vs time.

Table 1.1 Summary of Fission-Product Release Experiments
in the CRI with the Stainless Steel Liner Installed

CRI Run No.	Date	Sample	Fuel	Cladding	Atmosphere	Aging (hr)	Maximum Pressure (atm abs)	Maximum Temperature (°C)
100I	7-28-66	^{130}I tracer; I_2 in quartz ampul	None	None	Air	16.9	1.9	25.0
102HI	9-8-66	^{130}I tracer; $\text{I}_2 + \text{H}_2$ (400°C) with platinum catalyst	None	None	Air	19.7	2.0	25.0
102Kr	4-4-67	^{85}Kr tracer; calibration with gaseous ^{85}Kr	None	None	Air		1.0	25.0
103I	5-3-67	^{130}I tracer; I_2 in Pyrex ampul	None	None	Air	20	2.0	25.0
104I	5-30-67	^{130}I tracer; I_2 in Pyrex ampul	None	None	Steam-air	20.9	3.3	125.5
105HI	6-20-67	^{130}I tracer; $\text{I}_2 + \text{H}_2$ (400°C) with platinum catalyst	None	None	Steam-air	21.7	3.6	99.0
106M	7-5-67	$\text{CH}_3^{131}\text{I}$ in stainless steel container	None	None	Steam-air	23.5	3.4	104.0
107S	9-25-67	CSE simulant; I_2 in ampul with filament vaporizer	UO_2	Zircaloy	Steam-air	20.6	3.3	110.5
108sZr	12-5-67	Melted 2490 Mwd irradiated UO_2 ; I_2 in ampul	UO_2	Zircaloy	Steam-air	23.2	3.2	110.0
109SI	2-11-68	Prompt release of $^{130}\text{I}_2$ implant	UO_2	Grooved Zircaloy	Steam-air	22	3.0	107.5
110SI	4-22-68	Prompt release of $^{130}\text{I}_2$ implant	UO_2	Grooved Zircaloy	Steam-air	21.7	2.7	118.0
111SI	5-21-68	Prompt release of $^{130}\text{I}_2$ implant	UO_2	Grooved Zircaloy	Steam-air	23.7	2.7	115.0
112U	6-27-68	U_3O_8 aerosol, 0.0019 mg/ml	$\text{U} + \text{O}_2$	Zircaloy	Air-steam	~72	2.7	109.5
113U	7-30-68	U_3O_8 aerosol, 0.6 mg/ml	$\text{U} + \text{O}_2$	Zircaloy	Air-steam	~72	2.5	114.0
114H	8-21-68	Melted 6000-Mwd irradiated UO_2	UO_2	Zircaloy	Air-steam	20.2	3.6	111.0

- (3) Particle size vs time.
- (4) Fraction of iodine associated with particles as a function of time.
- (5) Chemical state of iodine as a function of time.
- (6) Containment atmosphere pressure and temperature as a function of time.
- (7) Containment wall temperature as a function of time.

Although the program was primarily directed at fulfilling LOFT needs, the data obtained at other than LOFT specific conditions should be of value to the nuclear industry in general.

The CRI (Fig. 1.1) consists of three major parts: (1) a 1200-gal stainless steel containment vessel, (2) a 20-gal stainless steel primary vessel, and (3) a quartz induction furnace. In addition, several recycle and depressurization filters, pressure reduction and decontamination equipment, and various mechanical and electrical components necessary for the operation of such a system are provided. Also it is possible to insert complete gastight liners fabricated of various materials into the containment vessel. However, for the runs reported here, only the stainless steel containment vessel liner was utilized.

The CRI (Figs. 1.1 and 1.2) was built for the LOFT Assistance Program to meet the need for a larger and more versatile facility than those available for studying the behavior of released fission products under LOFT conditions. The CRI also provides for evaluating the performance of engineered safety features with realistically aged fission-product aerosols. Provision is also made for the evaluation of pressure-suppression chemical sprays for the removal of various forms of radioiodine. The readily removable liners in the CRI containment vessel (Fig. 1.3) permit direct comparison of the containment surface environmental effect on fission-product behavior. Stainless steel and typical coated surfaces can be tested.

Fig. 1.1 Schematic of the CRI Facility.

Fig. 1.2 CRI Containment Vessel and Penthouse.

Fig. 1.3 CRI Containment Vessel and Components.

2. DESCRIPTION OF FACILITY AND EQUIPMENT

Construction of the CRI was completed in May, 1966, with the exception of fitting the several liners for the containment vessel that are to be employed in the different tests. After the first two experiments were completed in the unlined stainless steel containment vessel, it was removed from the pent house (Fig. 2.1) in order to expedite the fitting process for the remaining liners. The details of the matching of penetrations in the vessel and the liners and in the sealing flanges are shown in Fig. 2.2. Also visible in this photograph are the matched head rings that connect upper and lower sections of the liners, the support lugs recessed in the major vessel flange, and the lifting rings. The photograph also reveals the very close tolerance in matching diameters of the liners with the pressure shell. The average clearance in the annular space is less than 1/8 in. The liner shown is of stainless steel construction and is one of two to be provided with an interior concrete surface by addition of a 3/4-in. layer of Gunitite that will be painted to simulate the LOFT containment structure surface.

The in-cell primary system and auxiliary equipment are shown in the complex of piped tanks and wiring shown in Fig. 2.3. The insulated vessel is the reactor pressure vessel simulator, and the large valves on its right side are the blowdown outlets (top and bottom). On the left, the 2-in. pipe connected to the bottom of the containment vessel is the cold leg of a thermal-convection loop. It is expected that the natural convection rate through the hot primary vessel will be about 2 volume changes per minute and from the containment vessel about 5%/min. It will be of interest in the future to observe the effect of natural convection on fission-product transport, particularly of radioiodine from the primary vessel.

Fig. 2.1 Interior of Penthouse Showing the Insulated Containment Tank.

Fig. 2.2 Bottom Half of Containment Tank Showing
Liner Penetrations and Sealing Flange.

Fig. 2.3 Interior of Hot Cell Showing Insulated Primary Tank and Connecting Piping.

The provisional limits listed at the top of Table 2.1 define the radiation and safety limitations of the two major vessels of the CRI system. The equipment available for use in these vessels is also listed in the table.

A flow diagram of the CRI equipment is shown in Fig. 1.1. The "primary vessel," which was to simulate a reactor pressure vessel, was not used in the experiments listed, but was bypassed when the induction furnace heating equipment was used.

2.1 Fuel Meltdown Equipment

Induction heating was used to heat fuel samples in the CRI experiments. The copper tubing induction coil was placed around a furnace tube which contained the fuel sample in a quartz boat (Fig. 2.1.1). As pressure was present in the furnace tube and the connected containment vessel during the experiments in which fuel melting and heating was employed, the possibility of a "blow-out" of the furnace tube contents had to be considered. Secondary containment was provided by enclosing the induction coil and the furnace tube in a 6 in. Pyrex pipe, which was pressurized to a slightly greater pressure than that present in the furnace tube and tank during the heating cycle. During fuel heating a steam-air mixture, similar to that present in the containment vessel, flowed over the hot fuel surface and swept the generated aerosol into the containment vessel.

The metal housing shown at one end of the 6 in. Pyrex pipe in Fig. 2.1.1 contains a vaporizer unit which provides a means for heating small pellets to extreme temperatures with electrically heated tantalum and platinum filaments. In experiments CRI 107S, a CSE "simulant" run, it was desired to add the vapor of certain fission elements to the aerosol produced from molten UO_2 , and pellets containing Ba, Te, Ru, and Cs were subjected to heat in the vaporizer unit, and the resulting vapors swept over the molten UO_2 and into

Table 2.1 CRI Operating Conditions and Equipment

	Containment Tank	Primary Tank
Volume, gal	1200	20
Maximum pressure, psi	80	500
Maximum temperature, °F	300	1000
Total ^{131}I activity, curies	50	10^4
Pressure suppression	Fog sprays	Jet sprays
Decontamination	Jet sprays	Flooding
Interior surfaces	Stainless steel Paint on steel Paint on concrete	Stainless steel
Filter systems	Interior recycle Exterior recycle (LOFT)	Pressure letdown
Gas-particulate sampling systems	CRI probe-type filter Pack and dif. tube LOFT gas-particulate samplers	Filter pack and diffusion tube
Liquid sampling systems	Tank condensate Wall condensate Rainout	Tank condensate
Plateout sampling system	Bayonet-type oriented plates LOFT plateout capsules	High-temperature metal plates

Fig. 2.1.1 CRI Melting Equipment with Vaporizer Housing.

Fig. 2.1.2 CRI Melting Equipment with Vaporizer Housing Removed.

the containment vessel. Figure 2.1.2 shows the vaporizer unit with the metal housing removed.

2.2 Containment Vessel

The containment vessel was constructed so that different liners could be inserted to study radioactivity deposition on different materials such as stainless steel, paints, and concrete. Figure 1.3 is a schematic of the containment vessel with a stainless steel liner in place.

The tank walls are 0.95 cm thick stainless steel with a cylindrical section of approximately 1.52 meters in height, with an outside diameter of 1.695 meters. The conical portion of the vessel is approximately 0.76 meters in height with a minor diameter (outside) of 0.19 meters. The dished head is approximately 0.429 meters in height.

The calculated volume is $4.62 \times 10^6 \text{ cm}^3$. The internal surface (without liner), excluding additional surfaces due to internal equipment such as the fan, spray headers, gas samplers, etc., is $1.34 \times 10^5 \text{ cm}^2$.

In the experiments discussed here, the "Recycle Filtering System (Internal)," Fig. 1.3, was not in place. A more detailed discussion of the construction and operation of the CRI can be found in another report.¹

2.3 Pressure Release and Recycle Filters

At the conclusion of an experiment in the CRI, the excess pressure in the containment tank was vented through a filtering system which is referred to as the "pressure release or bypass filter," with a small fraction of the total exhaust gas diverted through a "compound filter" (Fig. 1.1). The compound filter was similar to the Maypack type used as gas samplers, and was used in an effort to determine the different forms of airborne radioactivity (particulate, molecular, etc.).

PHOTO 93253

Fig. 2.3.1 Recycle Filter Components.

When the venting process was complete, the containment tank contents were cycled through a filtering system, referred to as "recycle 1 and recycle 2" filters with "1" and "2" referring to the order of their use.

The pressure release (bypass) and recycle filters (Fig. 2.3.1) were identical in construction and consisted of, in order of exposure, a roughing filter, two absolute filters, three charcoal cartridges, and two back-up charcoal cartridges. Generally, in the same experiment, the same two back-up cartridges were used in both recycle filters. The charcoal cartridges used in the filters were 12.3 centimeters in diameter and 2.86 centimeters deep (Table 2.3.1), and with the charcoal used, having a density of 0.435 g/cc, the surface density (wt of charcoal/cross sectional area) of the cartridges was 1.24 g/cm^2 .

2.4 Sampling Devices

In the CRI experiments, three types of sampling devices were used; gas samplers of the May pack type to determine the forms of airborne activity present, deposition samplers with coupons to determine the deposition velocity of the various radioactivities on different structural materials, and a low pressure cascade impactor to determine the particulate sizes present in the containment tank atmosphere.

2.4.1 Gas Sampler

The gas-particulate sampler is illustrated in Fig. 2.4.1, which shows both the air-lock valve assembly and the component breakdown. The LOFT components provided by Phillips Petroleum Company for their gas-particulate sampler were used. The individual components (filters, silver screens, charcoal, etc.) are contained in stainless steel capsules that are seated by spring loading between Teflon ring gaskets to prevent channeling of the sampled gas. In sampling, a

PHOTO 85375

Fig. 2.4.1 CRI Gas Sampling Unit with Air Lock.

**Table 2.3.1 Diameter and Face Velocity for Components
of the Pressure Release and Recycle Filters**

Component	Diameter (centimeters)	*Face Velocity (Centimeters/second)
Roughing filter	10.39	11.82
Absolute filter	24.0	2.2
Charcoal cartridge	12.3	8.37

$$\text{*Face Velocity} = \frac{\text{(Flow in cm}^3\text{/sec)}}{\text{(component cross sectional area in cm}^2\text{)}}$$

5 liter tank was evacuated and then filled to atmospheric pressure by "pulling" gas from the containment tank through the sampler. In Table 2.4.1, the gas sampler components are listed with the number used and the order of exposure to the sampled gas, for most of the CRI experiments. The silver membranes referred to in the table have a 5 micron pore size and the "treated" charcoal is charcoal that had been impregnated with 2 wt % of KI to exchange with organic iodine compounds. In experiments 100I to 108SZr inclusive, "Flanders" absolute filters were used at the front end of the sampler and "Cambridge" absolute filters at the end. In 109SI, 110SI, 111SI, and 114H the order was reversed and "Cambridge" filters were used at the front of the sampler and "Flanders" at the end. In 109SI, 110SI, and 114H the "odd" and "even" numbered samples have different compositions in an effort to determine the portion of molecular iodine that could be adsorbed on the first absolute filters and lead to erroneous values for particulate iodine, which is assumed to be separated from the other forms at this point.

2.4.2 Deposition Coupons

Figure 2.4.2 shows the deposition sampler used in the CRI experiments. When inserted into the containment vessel through an air-lock, the coupons (in groups of four) are located approximately 11, 21, and 31 centimeters from the wall of the containment vessel. The individual coupons are held in slots in a block of Teflon so that, at each distance, there is a coupon facing upward, two sideways, and one down. Each coupon exposed 6.5 cm^2 ($5 \text{ cm} \times 1.3 \text{ cm}$) of surface area to the containment tank atmosphere.

2.4.3 Low Pressure Cascade Impactor

In the schematic of the impactor (Fig. 2.4.3), the sampled aerosol enters from the top, together with any sweep gas that is being used, and passes through a series of stages

Table 2.4.1 Arrangement and Number of Gas Sampler Components
(The order of exposure of component to sampled gas is from left to right)

Experiment Number	Silver		Filter	Silver		Charcoal		Filter		
	Honey- comb	Scre- ens	Abso- lute	Scre- ens	Honey- comb	Membr- ane	Untre- ated	Trea- ted	Abso- lute	Drier- ite
CRI 100I			2			4	3	1	1	
CRI 101HI (odd numbered samples)			2			4	2	3	1	
CRI 101SI (even numbered samples)			2			8	2	3	1	
CRI 103I			2			4	2	3	1	
CRI 104I			2			4	2	3	1	1
CRI 105HI			2			4	2	3	1	1
CRI 106M			2			4	2	3	1	1
CRI 107S			2			4	2	3	1	1
CRI 108SZr			2			3	2	3	1	1
CRI 109SI (odd numbered samples)			2			4	2	3	1	1
CRI 109SI (even numbered samples)		3	2			4	2	3	2	1
CRI 110SI (odd numbered samples)			2		1	3	2	2	2	1
CRI 110SI (even numbered samples)	1		2			3	2	2	2	1
CRI 111SI		3	2			4	2	2	2	1
CRI 114H (odd numbered samples)			2			4	2	2	2	1
CRI 114H (even numbered samples)		3	2			4	2	2	2	1

PHOTO 85373

Fig. 2.4.2 CRI Deposition Coupon Holder with Air Lock.

Fig. 2.4.3 Low-Pressure Cascade Impactor Mark-I.

to the exhaust side of the apparatus. The amount of sweep gas that is used depends on the pressure to be maintained at the different stages. Each stage of the impactor consists of a "jet" plate with a large number of holes of the same diameter and beneath this "jet" plate there is a metal plate covered with some material (silicone oil or millipore, for example) which will retain any particles that impinge upon it. In succeeding stages, the "jet" holes have decreasing diameters so that any particles present in the gas passing through are accelerated at each stage and particulate separation is accomplished. A complete description of the low pressure cascade impactor is given in another report.²

3. OPERATING PROCEDURE

The experimental procedure in the CRI was, in general, the same for all of the experiments discussed here. The activity was liberated from its source and blown into the containment vessel and, once there, was thoroughly mixed with the tank atmosphere by means of an internal fan. The mixed contents were then isolated for an "aging period" of approximately 20 hours. During this period, portions of the tank atmosphere were withdrawn periodically to be analyzed with a May pack type gas sampler or (in later experiments) with a low pressure cascade impactor. Deposition samples were inserted and withdrawn during the aging period and, accumulated condensates, in cases where steam-air atmospheres were present in the containment vessel, were drained from time to time. At the end of the aging period, the excess pressure in the containment vessel was vented through filters (compound and bypass), and the remaining tank contents cycled through filter systems (recycle 1 and recycle 2) to reduce the radioactivity of the tank atmosphere to a negligible value. When the recycle steps were completed, the interior

surfaces of the containment tank were sprayed with decontaminating solutions to remove deposited activity.

The following is a brief discussion of the methods used to generate aerosols that were used in the containment vessel during the CRI experiments.

CRI 100I, 103I, and 104I. In these experiments the iodine activity (^{130}I) was contained in a glass ampule which was placed in a stainless steel tube, with one end of the tube penetrating the wall of the containment tank (through a flanged port) and the other end connected to an air supply. The iodine activity was released by compressing the stainless tube (around the ampule) until the ampule was crushed. The released iodine was swept into the containment vessel with a stream of air. The walls of the stainless tube were heated to prevent any local deposition of activity.

CRI 101HI and 105HI. In these experiments the HI activity used was prepared by passing hydrogen gas over ^{130}I and combining these two components on a platinum catalyst before sweeping the product (HI) into the containment vessel. The major difference between the two experiments is that in 105HI a steam-air atmosphere was present in the containment tank instead of air as in 101HI.

CRI 106M. In this experiment the methyl iodide used was prepared by combining NaI (tagged with ^{131}NaI) with $(\text{CH}_3)_2\text{SO}_4$ and trapping the resulting CH_3I in a stainless steel cold trap. The methyl iodide was then swept from the trap into the containment vessel with a stream of air.

CRI 102Kr. In order to calibrate the gas sampling equipment used in the CRI experiments, ^{85}Kr was introduced into the containment vessel and thoroughly mixed with the contents. Several gas samples were then taken of the tank contents, and the ^{85}Kr activity (d/m/ml) determined. The contents of the tank were then diluted with a measured volume of air, mixed,

and again sampled. The sample results for the diluted contents were compared to those of the original tank contents and indicated that the sampling equipment removed essentially the same volume (STP) from the containment tank for each sample. Repeated dilutions, with mixing and sampling, confirmed these results.

CRI 107S. An attempt was made to simulate the melting of high-burnup fuel and, since the use of sufficient high-burnup UO_2 to produce realistic fission product concentrations in the containment tank was impractical because of radiation hazards, Ru, Te, and Cs containing tracer activities were vaporized from heated filaments in the vaporizer unit and the resulting vapors combined with a sweep gas of air and steam. This sweep gas was then blown over molten unirradiated UO_2 , with iodine vapor (containing tracer) being added. The resulting aerosol was swept into the containment tank which had been previously filled with an air-steam mixture. The UO_2 that was used to provide the molten surface was clad in Zircaloy prior to melting.

CRI 108SZr. Irradiated Zircaloy was used in this run in an attempt to determine whether the majority of particulates evolved when Zircaloy was melted were tin oxide. Unirradiated UO_2 , in pellet form, was enclosed in Zircaloy which had been highly irradiated and contained induced zirconium, antimony, tin, and cobalt activities. The UO_2 -irradiated Zircaloy specimen was then sealed in a Zircaloy can before melting. The fuel specimen was then melted with induction heating and while molten, irradiated ^{129}I was blown with a steam-air mixture across the melt surface and into the containment vessel. At one time, during the subsequent aging period, the containment tank contents were subjected to a spraying operation with 0.1 M $\text{Na}_2\text{S}_2\text{O}_3$ solutions.

CRI 109SI, 110SI, and 111SI. These experiments were of the "prompt release" type, which involved the release of

fission products as a result of fuel cladding failure due to heat and pressure. In all three experiments, ^{129}I was sealed in glass ampules, irradiated, and then inserted into a hole drilled axially through the center of unirradiated UO_2 pellets. The pellets and enclosed iodine were then sealed in a Zircaloy can and during the experiment heated by induction until the cladding failed and iodine was released to the containment vessel. In 109SI and 110SI the glass ampules were of quartz, which necessitated excessive heating (above the melting point of the Zircaloy cladding) to release their contained iodine activity. In CRI 111SI, the iodine was irradiated in a quartz ampule and then transferred to a Pyrex ampule before insertion into the drilled UO_2 . In CRI 110SI, the Zircaloy capsule containing the UO_2 pellets and inserted iodine was pressurized with 100 psig of He before sealing, and in 111SI the can was pressurized to 50 psig of He before sealing.

CRI 112U and 113U. To generate a U_3O_8 aerosol in CRI 112U, uranium metal was heated to a red heat with induction heating, and a stream of air blown over the hot uranium metal and transferring the resulting "smoke" of U_3O_8 particles to the containment vessel. In CRI 113U, in order to get a greater concentration of U_3O_8 particles in the containment vessel, the sweep air was fortified with oxygen to increase oxidation of the hot uranium, with the resulting increase of U_3O_8 particles to the containment vessel.

In both of these runs, the containment vessel was filled with an air-steam atmosphere prior to the U_3O_8 particle production.

CRI 114H. Forty grams (2 pellets) of Zircaloy-2 clad UO_2 were irradiated to approximately 6000 Mwd/T and, after cooling for approximately 4.5 years, were reirradiated for a short time in the Oak Ridge Research Reactor in order to build up the short-lived fission product inventory. The clad fuel was melted with induction heat, and the resulting aerosol swept into the containment vessel with a steam-air mixture.

4. RUN CONDITIONS

In Table 1.1 the CRI experiments are listed in order with date, sample, fuel, and cladding in addition to the temperature and pressure conditions existing in the containment vessel at the start of the aging period. In Table 4.2, most of the CRI experiments that involved iodine activity are listed with the amounts of iodine used.

4.1 Temperature

If the temperature of the containment vessel contents is expressed as degrees centigrade above room temperature ($t - 30$), and the log of this quantity plotted with time, the resulting curve is approximately a straight line. Such plots are shown for CRI experiments in Figs. 4.1.1 and 4.1.2, with the exception of CRI 112U where the data for the containment tank temperature were incomplete.

Least square fits for straight lines were made for the time-temperature data from the CRI experiments and the results are tabulated in Table 4.1.1. All temperatures in the table are in degrees centigrade above ambient ($t - 30$). Also listed in the table is the error mean square where

$$\text{error mean square} = \sqrt{\frac{(t_c - t_o)^2}{(N - 2)}},$$

where t_c is the computed temperature, t_o is the observed temperature, and N is the number of observations. It should be noticed, in the table, that the error mean square values are generally smaller for experiments 109SI to 114H inclusive. In Figs. 4.1.1 and 4.1.2 it is apparent that in the majority of the experiments that the temperature conditions were somewhat erratic in the early stages. Wall heaters were used on the containment tank walls (Fig. 1.3) to reduce the pre-heating time required to attain the desired starting temperature,

Table 4.2 Mass of Iodine Released in CRI Experiments

Experiment Number	Mass of Iodine Released (mg)	Iodine Concentration in the Containment Tank (gms/cubic meter)
100I	22.0	*4.8 x 10 ⁻³
101HI	10.0	*2.2 x 10 ⁻³
104I	30.0	6.7 x 10 ⁻³
105HI	20.0	4.5 x 10 ⁻³
107S	15.0	3.4 x 10 ⁻³
108SZr	20.0	4.5 x 10 ⁻³
109SI	9.2	2.0 x 10 ⁻³
110SI	2.4	5.0 x 10 ⁻⁴
111SI	0.23	5.0 x 10 ⁻⁵
114H	1.2	2.7 x 10 ⁻⁴

*No liner present in the containment vessel, and with air atmosphere.

Fig. 4.1.1 Temperature Conditions in the Containment Tank During Experiments 104I to 108SZr, Inclusive.

Fig. 4.1.2 Temperature Conditions in the CRI Containment Tank During Experiments 109SI to 114H, Inclusive.

Table 4.1.1 Temperature Conditions of the Containment Vessel
with Time, for the CRI Experiments as Derived
from Least Squares Fit of the Equation:

$$(t - 30.) = A e^{-KT}$$

(t = temperature (°C) of the containment vessel,
T = time in minutes, and A and K are constants)

Exp. No.	Time ¹ (min)	Half Time (min)	Error Mean Square	K ² 10 ⁴ xmin ⁻¹	Temperature at Zero Time ³	
					Computed	Observed
104I	1240	795	2.15	-8.721	92.9	95.0
105HI	1300	752	1.22	-9.214	71.0	69.0
106M	1345	771	1.71	-8.99	76.6	70.0
109SI	1220	770	0.67	-9.001	79.4	77.5
110SI	1223	918	1.18	-7.55	90.1	88.0
111SI	1290	874	0.68	-7.933	88.2	85.0
107S	1220	877	0.56	-7.903	82.2	80.5
108SZr	1210	781	0.49	-8.88	84.6	84.0
114H	1213	938	0.48	-7.393	80.7	81.0
113U	1325	987	0.57	-7.024	82.7	84.0

¹Upper limit of time used in the calculations.

²Slope of temperature-time curve (degrees centigrade/min).

³Temperatures are in degrees centigrade above ambient.
The actual reading of the thermocouples is this listed temperature plus 30 degrees (room temperature).

and thereby reduce the amount of condensation when the steam phase was introduced. These variations from a smooth decay curve could be caused by the wall heaters being activated for too long a period resulting in a temperature "over-ride". The fact that the half-times for the temperature curves are not identical could be due to this unequal heating and variations of steam-air ratios in the containment tank atmospheres.

It has been previously stated that in some of the experiments (CRI 108SZr, CRI 110SI, and CRI 111SI), the efficiency of thiosulfate sprays for reducing the concentrations of airborne activity in the containment tank was investigated. These spraying operations were started rather late in the aging period (CRI 108SZr at 1237 minutes, CRI 110SI at 1223 minutes, and CRI 111SI at 1302 minutes). The spray solution was not preheated to existing containment tank temperatures, but very little cooling of the containment tank contents could be observed in the temperature-time graphs, because of the small volume of spray compared to that of the containment vessel (approximately 1/1000) and, in the case of 111SI, the spraying was at 1302 minutes and the graphs of time-temperature conditions extended only to 1200 minutes.

4.2 Pressure

The pressure-time graphs (Figs. 4.2.1 and 4.2.2) for the CRI experiments have few departures from a smooth curve, except in the early stages of the experiments. Since pressure and temperature are interdependent, those factors causing irregularities in temperature would have a related effect on pressure.

4.3 Condensation

During the runs where a steam-air atmosphere was present in the containment vessel, the condensate was drained from the tank at various intervals and the total condensate

Fig. 4.2.1 Pressure in the Containment Tank During Experiments 104I to 108SZr, Inclusive.

Fig. 4.2.2 Pressure in the Containment Tank During Experiments 109SI to 114H, Inclusive.

produced from the start of the run to the drainage time was recorded. If the values for accumulated condensate (V) are plotted with time (t), the resulting curve can be extrapolated to determine a maximum value for the volume of condensate (V_t) to be produced in the experiment. A typical plot of "V" with "t" is shown in Fig. 4.3.1. When the log of $(1 - V/V_t)$ is plotted for various collection times, the resulting curve (Fig. 4.2.2) approximates a straight line and can be expressed as

$$\ln(1 - V/V_t) = a + bt ,$$

or

$$V = V_t (1 - ke^{bt}) ,$$

where $k = e^a$. The condensation rate (ml/min) is then

$$(dV/dt) = -kV_t be^{bt} .$$

In Table 4.3.1 values for k, b, V_t , and $(dV/dt)_0$ are listed. These values can be used to compute condensate volumes and condensate rates with reasonable accuracy for times from 0 to 1200 minutes inclusive. Ideally "a" should equal zero, but in the CRI experiments where condensation was present, it (a) had small values due to the extreme difficulty of completely draining the containment vessel just prior to time zero.

5. FISSION PRODUCT BEHAVIOR (SAMPLING RESULTS)

The following material has been compiled from the various sampling devices used in the CRI.

5.1 General Distribution of Fission Products in the Containment Vessel

Listed in Table 5.1.1 is the distribution of the fission products in the containment tank atmosphere (from venting,

Fig. 4.3.1 Accumulation of Steam Condensate in the Containment Tank During Experiment 109SI.

Fig. 4.3.2 Steam Condensation Rate in Containment Tank During Experiment 109SI.

Table 4.31 Condensation Rates for the CRI Experiments

Run Number	$V_t^{(1)}$ (ml)	k	b ($10^3 \times \text{min}^{-1}$)	$T_{\frac{1}{2}}$ (min)	$(dV/dt)_0^{(2)}$ (ml/min)	Error Mean Square (3) (ml)
104I	3380	1.02	-2.38	292	8.0	44
105HI	2410	1.08	-2.8	248	6.8	46
106M	2170	1.08	-2.39	290	5.2	40
107S	2370	0.92	-2.09	331	5.0	22
108SZr	1890	1.1	-2.09	331	4.0	24
109SI	2700	0.9	-2.86	242	7.7	51
110SI	2910	0.99	-2.3	300	6.7	24
111SI	3390	0.9	-2.43	286	8.2	26
112U	2180	0.94	-3.47	200	7.6	47
113U	2540	1.0	-1.87	372	4.7	27
114H	2260	1.0	-1.66	417	3.8	39

(1) Value for total volume that results in the best least squares fit for the data.

(2) Condensation rate at zero time.

(3) Error mean square = $\sqrt{\frac{\Sigma(V_{\text{observed}} - V_{\text{computed}})^2}{(\text{Number of observations} - 2)}}$

Table 5.1.1 Distribution of Activity in the Containment Tank for CRI Experiments
(% of tank inventory)

Run Number	Activity	Deposited on Walls	Removed by Spraying	Removed in Condensated	Removed by Sampling	Venting Containment Tank	Cyclic Filtering
100I	Iodine	97.6			0.25	0.06	2.1
101HI	Iodine (HI)	98.6			0.07	0.06	1.3
103I	Iodine	77.7			0.45	8.5	13.4
104I	Iodine	84.1		15.3	0.03	0.18	0.35
105HI	Iodine (HI)	86.8		13.0	0.02	0.07	0.06
106M	Iodine (CH ₃ I)	5.1		0.39	1.1	53.3	40.1
107S	Iodine	84.0		14.7	0.05	0.55	0.5
	Cesium	62.0		26.7	0.86	4.4	6.0
	Tellurium	86.0		0.27	1.7	2.3	2.4
	Ruthenium	91.0		9.3	0.34	6.3	0.51
108SZr	Iodine	36.6	25.2	36.6	0.1	0.61	0.9
	Antimony	61.6	33.0	1.8	3.4	0.01	0.16
	Tin	91.9	5.4	1.3	1.2	0.002	0.08
109SI	Iodine	51.6		40.0	0.07	2.4	6.0
110SI	Iodine	56.9	20.2	20.9	0.21	0.42	1.3
111SI	Iodine	54.2	18.0	21.2	0.03	0.1	0.32
114H	Iodine	84.2		13.5	0.21	0.76	1.3
	Tellurium	38.6		52.4	1.8	5.0	2.1
	Cesium	72.0		21.3	1.0	2.4	3.3
	Ruthenium	86.1		12.9	0.64	0.002	0.36

cyclic filtering, and gas samples) deposited on the walls, and removed in the condensate. In addition, the amount of activity removed by spraying (CRI 108SZr, 110SI, and 111SI) is included. In all of the listed experiments, the iodine activity deposited on the walls is the major portion of the tank inventory with the exception of CRI 106M, where the activity was in the form of methyl iodide which has a minimal plate-out on metal surfaces. The second largest portion of the activity is generally found in the condensates.

At the conclusion of the aging period, the pressure in the containment vessel is approximately two atmospheres (absolute) (Figs. 4.2.1 and 4.2.2) and when vented to atmospheric, approximately one-half of the tank's atmosphere should be released to the venting filters. From Table 5.1.1 it can be seen that the portion of the iodine activity removed by cyclic filtering is usually greater than that trapped on the exhaust filters. This excess of iodine activity in the cyclic filters is due to desorption from the containment vessel walls.

It should be noticed in Table 5.1.1 that the amount of activity adsorbed on the walls of the containment vessel decreases after 103I. This decrease is caused by the presence of condensing steam in the containment tank with the condensate containing a significant amount of the activity in all runs subsequent to 104I.

Typical distribution of iodine activity in the containment vessel with time can be seen in Fig. 5.1.1. In this figure, the fraction of the iodine inventory in the condensate and airborne are determined by sampling results, while the fraction of the iodine inventory on the walls is assumed to be the remainder. The organic iodide (CH_3I) is, of course, a variable fraction of the total airborne iodine.

Fig. 5.1.1 Distribution of Iodine Activity in Containment Vessel During Run 114H.

5.2 Gas Sampler Results

The activity found on each of the components of the May pack type gas samplers was totaled for each sample and, as the containment vessel was usually under pressure and contained a steam-air atmosphere when sampled, the total in the sample had to be adjusted for pressure and condensation to obtain a value for disintegrations/minute/milliliter (d/m/ml) of the containment tank atmosphere. The values for d/m/ml of containment tank atmosphere, when multiplied by the containment vessel volume and divided by the inventory, became the fraction of the inventory that was airborne and was denoted as " C/C_0 ".

The C/C_0 value for airborne iodine usually approaches a constant value "K" at the end of the aging period in the containment tank (Figs. 5.2.1, 5.2.2, and 5.2.5). In several CRI experiments this constant was estimated from the graphs of the C/C_0 values with time, when the C/C_0 values were not too erratic as is the case for 108SZr and 109SI. The $(C/C_0 - K)$ quantities were then fitted by computer to an expression of the type

$$(C/C_0 - K) = a_1 e^{-b_1 t} + a_2 e^{-b_2 t},$$

where "t" is the time in minutes. The results of the calculations are presented in Table 5.2.1. Experiment 106M was not included, because the iodine activity was in the form of methyl iodide and did not follow the characteristic depletion curve for iodine (Fig. 5.2.1).

The long life component half-times ($0.693/b_2$) listed in Table 5.2.1 are, with the exception of that for 107S, fairly similar having an average value of approximately 31 minutes. The short life component half-times ($0.693/b_1$) show great variation in values; neglecting those half-lives of less than one minute (100I, 101HI, 105HI, and 110SI), the

Fig. 5.2.1 Concentration of Airborne Iodine Activity With Time in CRI Experiments 101I to 106M, Inclusive.

Fig. 5.2.2 Variation in Radioiodine Behavior in CRI Stainless Steel Containment.

Table 5.2.1 Results Obtained by Fitting the Airborne Iodine Concentration in the Containment Tank with Time to the Expression;

$$*C/C_0 = K + a_1 e^{-b_1 t} + a_2 e^{-b_2 t} \quad (t = \text{min})$$

Experi- ment Number	Constant "K"	a_1	Half-time $0.6931/b_1$ (minutes) ¹	a_2	Half-time $0.693/b_2$ (minutes)	**Error Mean Square
100I	0.0035	0.48	0.35	0.52	24.6	0.071
100LHI	0.015	0.91	0.15	0.072	29.8	0.023
103I	0.14	0.37	3.4	0.49	34.0	0.184
104I	0.0058	0.97	1.6	0.023	22.5	0.0053
105MI	0.0018	0.98	0.88	0.02	22.0	0.0035
107S	0.021	0.93	2.8	0.016	208.0	0.049
110SI	0.0165	0.72	0.23	0.26	27.0	0.052
111SI	0.0035	0.97	2.2	0.03	52.2	0.0084
114H	0.016	0.90	2.8	0.082	33.5	0.018

* C/C_0 is equal to airborne concentration at time "t" divided by the airborne concentration at time zero, assuming that the total inventory of the containment vessel is airborne at time zero.

$$** \text{Error mean square} = \sqrt{\frac{\sum (C/C_0 \text{ observed} - \text{computed } C/C_0)^2}{\text{Number of observations} - 2}}$$

Fig. 5.2.3 Variation in Airborne Activity in the CRI Containment Vessel with Time During Run 107S.

Fig. 5.2.4 Concentration of Airborne Activities in the Containment Tank During Experiment 108SZr.

Fig. 5.2.5 Distribution of Activity in the Containment Tank During Run 114H.

remaining have an average value of 2.5 minutes.

Plots of C/C_0 values with time for those experiments where the activity was injected directly into the containment vessel, are shown in Fig. 5.2.1. As would be expected, 106M shows a very slow reduction in concentration of airborne CH_3I . Experiments 104I and 105HI were the only experiments in this group with steam-air atmospheres and a resulting steam flux to the walls. As expected, the presence of this steam flux is reflected in their concentration-time curves, which show a greater initial depletion than that for the other experiments shown.³

In Fig. 5.2.2, the C/C_0 values for iodine activity are plotted with time to a 1200 minute limit for experiments 107S, 108SZr, 109SI, 110SI, and 111SI. From the figure, it is evident that the experiments with the higher iodine concentrations (Table 4.2) have the greatest amount of iodine desorption.

Figures 5.2.3, 5.2.4, and 5.2.5 are plots of airborne activity concentrations (C/C_0) in the containment vessel with time for an aging period of 1200 minutes. In 108SZr and 114H (Figs. 5.2.4 and 5.2.5), the particulate activities (Sb, Co, Sn, Cs, and Te) all are straight lines on a semilog plot with time, while 107S (Fig. 5.2.3) particulate activities (Cs, Ru, and Te), on a similar plot, do not assume a straight line decay until after approximately 300 minutes have elapsed. The average half-time for the straight line portion of the particulate curves in 107S is 8 hours, in 108SZr it is 3.3 hours, and in 114H it is 5 hours. The condensate rates in milliliters/minute at time zero for the experiments 107S, 108SZr, and 114H are 5, 4, and 3.8, respectively (Table 4.3.1). The condensate rates for 108SZr and 114H, as well as the half-times for particulates (3.3 and 5 hours), are somewhat similar. C. 107S is quite puzzling, in that it has a larger condensation rate (5 ml/min), but a longer half-time (8 hours) for the particulates.

The airborne concentration of uranium, as U_3O_8 , during experiments 112U and 113U, is shown in Fig. 5.2.6. The curve for 113U is similar to one by Davis⁴ showing the combined effects of coagulation and settling, diffusiophoresis, and thermophoresis.

The distribution of iodine activity on the gas sampler components provides a rough indication as to the state of the activity. The activity on the filters indicates "particulate," that on silver indicates "molecular" or "reactive," and that on charcoal indicates "organic". The gas samples were taken at different intervals in the different experiments and, in order to list distribution values with time in a table of convenient size, the distributions (fractions of sample activity) were plotted with their times (Figs. 5.2.7 and 5.2.8) and values selected from the smoothed curves for specified times. These values for the various CRI experiments are listed for the selected times in Tables 5.2.2 and 5.2.3. Of interest in Table 5.2.2 is the relatively low values for the fraction of iodine on silver in the early stages of experiments 104I and 105HI, these runs differing from the others listed by having a steam-air atmosphere.

In Table 5.2.3, experiments 109SI and 110SI used two different types of component arrangement in their gas samplers — one type had a silver component (honeycomb or screens) preceding the first absolute filter (Table 2.4.1), and the other was the conventional May pack with the absolute filters being the first components to encounter the sampled gas. Experiments 111SI and 114H also had a silver component preceding the absolute filters, but no "conventional" packs were used in these experiments as a basis for comparison. From Table 5.2.3 it can be seen that in both experiments, 109SI and 110SI, the placing of a silver component before the absolute filters reduced the amount of activity trapped by the absolute filters. This fact would

Fig. 5.2.6 Uranium, as U_3O_8 , Concentration in the Containment Vessel Atmosphere During Experiments 112U and 113U.

Fig. 5.2.7 Distribution of Iodine Species During CRI Experiment 107S.

Fig. 5.2.8 Distribution of Iodine Species During CRI Experiment 114H.

Table 5.2.2 Distribution of Iodine Activity on Gas Sampler Components
(Fraction of total activity in the gas sample)

Experiment No.	Components	Time (minutes)										
		0	50	100	150	200	300	400	600	800	1000	1200
100I	Filter	0.091	0.085	0.074	0.07	0.074	0.09	0.14	0.30	0.44	0.575	0.59
	Silver	0.90	0.91	0.92	0.924	0.917	0.903	0.848	0.67	0.46	0.354	0.33
	Charcoal	0.009	0.005	0.006	0.006	0.004	0.0076	0.012	0.03	0.055	0.071	0.081
101HI	Filter	0.20	0.064	0.046	0.04	0.037	0.034	0.033	0.034	0.039	0.043	
	Silver	0.77	0.848	0.88	0.908	0.923	0.941	0.948	0.952	0.947	0.942	
	Charcoal	0.03	0.088	0.073	0.052	0.040	0.021	0.019	0.014	0.014	0.015	
103I	Filter	0.021	0.021	0.021	0.021	0.021	0.020	0.019	0.018	0.017	0.017	0.016
	Silver	0.975	0.973	0.97	0.968	0.966	0.96	0.955	0.943	0.937	0.933	0.93
	Charcoal	0.004	0.006	0.009	0.011	0.013	0.02	0.026	0.039	0.046	0.051	0.054
104I	Filter	0.76	0.58	0.44	0.35	0.28	0.19	0.14	0.09	0.067	0.053	0.043
	Silver	0.23	0.315	0.38	0.40	0.42	0.44	0.42	0.36	0.30	0.287	0.277
	Charcoal	0.009	0.105	0.18	0.25	0.30	0.37	0.44	0.55	0.63	0.66	0.68
105HI	Filter	0.42	0.38	0.35	0.31	0.28	0.25	0.21	0.16	0.13	0.11	0.09
	Silver	0.58	0.60	0.60	0.61	0.61	0.6	0.6	0.58	0.56	0.54	0.54
	Charcoal	0.001	0.02	0.05	0.08	0.11	0.15	0.19	0.26	0.31	0.35	0.37

Table 5.2.3 Distribution of Iodine Activity on Gas Sampler Components
(Fraction of total activity in gas sample)

Experiment No.	Components	Time (minutes)										
		0	50	100	150	200	300	400	600	800	1000	1200
107S	Filters	0.94	0.55	0.48	0.45	0.42	0.40	0.38	0.38	0.37	0.36	0.34
	Silver	0.05	0.31	0.33	0.32	0.33	0.32	0.32	0.31	0.34	0.39	0.46
	Charcoal	0.005	0.14	0.19	0.23	0.25	0.28	0.30	0.31	0.29	0.25	0.20
108SZr	Filters	0.73	0.71	0.68	0.65	0.63	0.58	0.54	0.47	0.40	0.34	0.29
	Silver	0.24	0.26	0.29	0.32	0.33	0.38	0.41	0.46	0.51	0.54	0.54
	Charcoal	0.027	0.03	0.032	0.03	0.04	0.04	0.05	0.07	0.09	0.12	0.17
109SI	Filters	0.81	0.79	0.77	0.74	0.72	0.70	0.69	0.63	0.61	0.60	0.59
	Silver	0.17	0.19	0.21	0.23	0.25	0.27	0.30	0.33	0.35	0.36	0.36
	Charcoal	0.02	0.02	0.24	0.025	0.027	0.03	0.032	0.036	0.04	0.043	0.046
(1) 109SI	Filters	0.35	0.31	0.28	0.26	0.24	0.22	0.20	0.135	0.18	0.18	0.18
	Silver	0.62	0.66	0.69	0.71	0.74	0.76	0.78	0.79	0.79	0.80	0.79
	Charcoal	0.03	0.03	0.03	0.03	0.02	0.02	0.02	0.02	0.02	0.02	0.03
110SI	Filters	0.88	0.90	0.90	0.91	0.92	0.93	0.93	0.93	0.93	0.91	0.89
	Silver	0.115	0.10	0.09	0.08	0.07	0.06	0.05	0.05	0.05	0.06	0.07
	Charcoal	0.002	0.0044	0.0062	0.008	0.0092	0.012	0.015	0.021	0.026	0.032	0.039
(1) 110SI	Filters	0.69	0.71	0.77	0.74	0.74	0.75	0.76	0.77	0.77	0.77	0.77
	Silver	0.28	0.26	0.25	0.23	0.22	0.21	0.20	0.18	0.17	0.17	0.17
	Charcoal	0.03	0.03	0.03	0.03	0.04	0.04	0.04	0.05	0.06	0.06	0.06
(1) 111SI	Filters	0.25	0.26	0.26	0.26	0.25	0.23	0.21	0.16	0.12	0.09	0.07
	Silver	0.71	0.67	0.65	0.63	0.61	0.58	0.54	0.50	0.45	0.39	0.33
	Charcoal	0.04	0.07	0.09	0.11	0.14	0.19	0.25	0.34	0.43	0.52	0.60
114H	Filters	0.08	0.42	0.43	0.44	0.44	0.43	0.41	0.33	0.27	0.21	0.15
	Silver	0.92	0.55	0.53	0.50	0.49	0.48	0.48	0.55	0.59	0.64	0.69
	Charcoal	0.001	0.03	0.04	0.06	0.07	0.09	0.11	0.12	0.14	0.15	0.16

(1) The usual arrangement of components was filters, silver, and charcoal, but in these samples the arrangement was silver, filters, silver, and charcoal (Table

indicate that the component arrangement used in the early CRI experiments could lead to inflated values for "particulate" iodine which is supposedly trapped on the filters, and the amounts of particulates trapped by honeycomb and screens⁵ are negligible.

5.3 Deposition Samples

The deposition velocity (V_g) for an activity on a surface was computed according to:

$$V_g = \frac{\text{Total activity deposited on surface (d/m)}}{\text{Time} \times \text{area} \times \text{concentration}}$$

where

Time = Time surface is exposed (sec)

Area = Area of surface exposed (cm^2)

Concentration = Average gas phase concentration during exposure (d/m/cc).

The deposition coupons are described in section 2.4.2 of this report. It is possible to calculate a value for V_g of the containment vessel walls by considering the wall as a deposition coupon and using the value for activity removed in the tank washes as the amount deposited. A source of error in the wall calculations is the fact that iodine activity tends to desorb from the walls during the recycling procedure, and results in a lesser value for V_g than that found from deposition coupons. In Table 5.3.1 only in two experiments, 103I and 114H, are the deposition velocities computed for the tank walls larger than those calculated from deposition coupons. The average value for the eight V_g 's listed for iodine, as determined from deposition coupons, is 1.45×10^{-2} cm/sec, and the V_g 's computed from the tank walls have an average value of 8×10^{-3} cm/sec.

In some of the later CRI experiments, two sets of deposition coupons were used; one set being exposed during the first part of the aging period and a second set exposed during

Table 5.3.1 Deposition Velocity (V_g) of Iodine
on Stainless Steel in the CRI Experiments
(V_g values listed in the table are to be
multiplied by 10^{-2})

Experiment Number	V_g in cm/sec	
	*Deposition Coupons	Tank Walls
100I	-	0.36
101HI (as hydroiodic acid)	0.035	2.4
103I	0.03	0.08
104I	2.5	2.1
105HI (as hydroiodic acid)	12.3	8.2
106M (as CH_3I)	0.00072	-
107S	1.5	1.1
108SZr	0.63	0.14
109SI**	3.0	0.5
110SI**	0.05	0.4
111SI**	2.5	1.1
114H**	0.9	1.1

* V_g value is the average for all of the coupons.

**Two sets of coupons used in the experiment.
Average value listed.

the latter part of the aging period. The coupons were of three types: stainless steel, stainless steel painted with Amercoat, and stainless steel painted with Phenoline. The deposition velocities for iodine on the different surfaces are presented in Table 5.3.2. From the table, it is evident that the deposition velocities from coupons used in the early part of an experiment were larger than those used in the later part of the same experiment. The main differences between the two sets being the exposure times (about twice as long for the second set) and the average iodine concentration in the gas phase over the coupon surfaces. From Table 5.3.2 the deposition velocities found in experiment 109SI are larger than the others listed in the table. Table 4.2 shows that 109SI had the largest total iodine inventory in the four experiments compared here (109-114). The deposition velocities listed in the table for Phenoline and Amercoat surfaces are in a ratio of about 2 to 1, the Phenoline V_g 's being about twice as large as the Amercoat V_g 's for the individual experiments. The average for all V_g 's listed for a particular surface are: 1.82×10^{-2} cm/sec for stainless steel, 6.7×10^{-2} cm/sec for Amercoat, and 11.1×10^{-2} cm/sec for Phenoline.

In Table 5.3.3, the V_g 's for HI in a steam-air atmosphere (105HI) are much greater than those for HI in an air atmosphere (101HI). The average V_g for 105HI is 12.3×10^{-2} cm/sec, and the average V_g for 101 HI, excluding the "standard surface" coupons,⁶ is 2.5×10^{-2} cm/sec. Table 5.3.3 also indicates that an air-steam atmosphere increases the deposition velocity for iodine on stainless steel.⁷ The V_g 's listed for CRI 106M in Table 5.3.3 are those for CH_3I and, as one would expect, very small with most of them with the exception of those situated 11 centimeters from the tank wall, being in the neighborhood of 10^{-7} cm/sec.

In those experiments with elemental iodine, the attitude of the coupons seems to have little effect with any excess of

Table 5.3.2 Deposition Velocities for Iodine on Various Materials

(All values are to be multiplied by 10^{-2})

Experiment Number	Sample* Set	Exposure Time (minutes)	Average Values for Deposition Velocities (cm/sec)		
			Stainless Steel	Amercoat	Phenoline
109SI	1	5 - 401	4.3	12.0	22.8
109SI	2	414 - 1215	2.1	10.9	15.8
110SI	1	10 - 435	0.7	4.4	8.7
110SI	2	439 - 1214	0.6	1.1	1.8
111SI	1	1 - 470	2.51	6.0	14.8
111SI	2	475 - 1280	2.5	4.3	7.1
114H	1	8 - 338	1.5	10.5	15.0
114H	2	342 - 1180	0.35	4.5	2.9

* Denotes the first or second set of deposition samples used.

Table 5.3.3 Effect of Distance from Tank Wall and Attitude of Stainless Steel Deposition Coupons, on the Deposition Velocity of Various Forms of Iodine Activity

(All values of V_g are to be multiplied by 10^{-2})

Run No.	Distance of Coupon From Tank Wall (cm)	Coupon Exposure Time (min)	Average Gas Phase Concentration During Exposure (d/m/ml)	Deposition Velocity (V_g) According to Direction of Exposed Surface			
				Up	Side	Down	Side
103I	11	1221	3.1×10^3	0.034	0.029	0.035	0.031
	21			0.04	0.031	0.024	0.037
	31			0.025	0.019	0.037	0.033
104I	11	1254	1.3×10^3	(1)	(1)	(1)	(1)
	21			6.0	0.37	0.61	0.34
	31			7.6	1.2	2.0	1.7
107S	11	1233	375	3.7	3.7	9.7	2.9
	21			0.41	0.74	1.4	0.55
	31			0.58	0.5	0.73	0.61
108SZr	11	1219	630	0.44	0.53	0.68	0.3
	21			1.1	0.33	0.6	0.68
	31			0.75	0.95	0.51	0.68
101HI	11	1124	3.1×10^3	3.3	0.8 ⁽²⁾	3.4	3.4
	21			1.8	1.1 ⁽²⁾	2.4	2.0
	31			2.2	0.4 ⁽²⁾	2.0	1.1
105HI	11	1303	79	19.9	6.4	6.2	15.6
	21			21.3	4.4	11.9	7.3
	31			18.1	12.0	12.2	12.9
106M	11	1348	1.2×10^4	4×10^{-3}	3×10^{-3}	2×10^{-5}	1×10^{-3}
	21			3×10^{-4}	3×10^{-5}	6×10^{-5}	3×10^{-5}
	31			2×10^{-4}	3×10^{-5}	3×10^{-5}	7×10^{-5}

(1) Sampler stuck in the containment tank and remained until after the tank was decontaminated. These samples (11 cm) were washed with the decontaminating solution and are not recorded.

(2) Special treated "Standard Surface" stainless steel.⁽⁵⁾

activity on the surfaces "facing upward" indicating the presence of iodine activity on particulates. In general, it can be said that the coupons closest to the tank wall (11 cm) have V_g 's larger than those in the other two positions (Figs. 5.3.1, 5.3.2, and 5.3.3).

The variation of deposition of iodine on coupons with distance from the tank wall and orientation for experiments 109SI, 110SI, and 111SI, is graphically depicted in Figs. 5.3.1, 5.3.2, and 5.3.3, and again the coupons closest to the wall have the larger deposition velocities. In Table 5.3.1, one set of the coupons orientated parallel to the tank wall ("side") have the lesser V_g 's for these coupons could be caused by currents (swirling) in the containment tank atmosphere, with a resulting lower concentration of activity over the exposed surfaces.

In Fig. 5.3.4, the deposition of particulate activities (Sb and Sn) as well as iodine is pictured. It can be seen that the iodine activity is rather evenly distributed over the different coupon orientations or attitudes, while the particulates show a marked preference for the "top" coupons (facing upward). This preference of particulates for the "facing up" orientation is again depicted in Fig. 5.3.5 where cesium activity deposited on the coupons (CR! 114H).

5.4 Impactor Samples

When the activity collected on the plates of the Low Pressure Cascade Impactor is plotted with the plate number, the results resemble the ones shown in Fig. 5.4.1. The activity collected on the plates is added with that collected on "back-up" systems that are used to trap any activity penetrating the impactor proper, and the total is used to compute the "% penetration" for each stage of the impactor. When the "% penetration" of a stage is plotted with the "stage constant,"² on log-probability paper (Fig. 5.4.2), it

ORNL-DWG 68-6154

Fig. 5.3.1 Results of Plateout of Radioiodine on Oriented Coupons in Run 109SI.

ORNL-DWG 68-8828

Fig. 5.3.2 Analyses of Radioiodine on Plateout Coupons in CRI Run 110SI.

ORNL-DWG 68-8829

Fig. 5.3.3 Analyses of Radioiodine on Plateout Coupons in CRI Run 111SI.

Fig. 5.3.4 Average Distribution of Activity on Deposition Coupons According to Orientation, Run 108SZr.

OPNL-DWG 69-941

Fig. 5.3.5 Cesium Deposited on Plateout Samples
From CRI 114H.

is possible to evaluate a "mass median diameter" (MMD or D_{50}) and a "sigma" (σ) value. In Fig. 5.4.2 the "particle size (μ)" is the same as the "stage constants". The D_{50} is the diameter, in microns, found at the intersection of the 50% penetration and the generated lines. By definition⁸ the sigma value for the sample is

$$\text{Sigma} = \frac{50\% \text{ size (microns)}}{15.87\% \text{ size (microns)}}$$

The results obtained by using the Low-Pressure Cascade Impactor in the CRI experiments are listed in Tables 5.4.1, 5.4.2, and 5.4.3. In Table 5.4.1 the experiments listed (109SI, 110SI, and 111SI) are of the prompt release type where the Zircaloy encapsulated UO_2 and iodine tracer were heated until the cladding failed and released the iodine activity to the containment vessel. In Table 5.4.2, experiments 108SZr and 114H had molten UO_2 and Zircaloy cladding as a source of particulate material for the generated aerosols, with 108SZr having an additional amount of high burnup Zircaloy present. In Table 5.4.3, experiments 112U and 113U generated aerosols containing U_3O_8 particles by "burning" uranium and blowing the resulting particulates (smoke) into the containment vessel.

In experiments 108SZr and 114H (Table 5.4.2), those D_{50} 's obtained by using iodine activity, decrease with a corresponding increase of sigma, with time. The D_{50} values for 112U and 113U (Table 5.4.3) also decrease (sigma increases) with time. The above seems to indicate that the experiments with a high particulate content in their aerosols have a decreasing mass median diameter with time.

Disregarding the sampling times, and averaging the D_{50} 's resulting from iodine counting, as listed in Tables 5.4.1 and 5.4.2, the average values for experiments 109SI, 110SI, 111SI, 108SZr, and 114H are 0.12, 0.12, 0.11, 0.21 and 0.27 microns, respectively. Those experiments having molten metal

Fig. 5.4.1 Low-Pressure Impactor Plate Distribution of Radioactivity in CRI 114H.

Fig. 5.4.2 Size Distribution of Particles Carrying ^{134}Cs .

Table 5.4.1 Mass Median Diameters and Sigma Values
Found by Using the Low-Pressure Cascade Impactor
During the "Prompt-Release" Experiments

(Particle density assumed to be 6 grams/cc)

Experi- ment No.	Sample	Activity ¹	Sampling ² Time (min)	Mass Median Diameter (cm x 10 ⁵)	Sigma
109SI	1	Iodine	33	0.98	2.2
109SI	2	Iodine	161	0.96	2.2
109SI	3	Iodine	427	1.6	2.0
110SI	1	Iodine	37	0.85	2.3
110SI	2	Iodine	452	1.6	1.7
111SI	1	Iodine	36	0.85	2.8
111SI	2	Iodine	486	1.3	2.0

¹Activity counted to determine total material on each impactor plate.

²Midpoint of the sampling interval.

Table 5.4.2 Particle Sizes (MMD's) Found in Aerosols
Generated in CRI Experiments 108SZr and 114H

(Particle density assumed to be 6 gm/cc)

Experi- ment No.	Sample	Activity ¹	Sampling ² Time (min)	Mass Median Diameter ($\mu m \times 10^5$)	Sigma
108SZr	1	I	14	2.4	2.04
108SZr	1	Sb	14	4.0	1.74
108SZr	1	Sn	14	4.7	1.74
108SZr	2	I	83	1.7	2.5
108SZr	2	Sb	83	5.5	1.82
108SZr	2	Sn	83	5.5	1.59
114H	1	I	36	3.2	1.45
114H	1	Te	36	3.1	1.32
114H	1	Cs	36	3.0	1.42
114H	1	Ru	36	2.8	1.82
114H	1	Sb	36	3.0	1.45
114H	2	I	220	3.2	2.57
114H	2	Te	220	3.0	1.74
114H	2	Cs	220	2.6	1.95
114H	2	Ru	220	3.2	2.34
114H	2	Sb	220	2.6	1.44
114H	3	I	753	2.2	2.38
114H	3	Te	753	2.0	2.0
114H	3	Cs	753	2.3	2.04
114H	3	Ru	753	2.5	2.35
114H	3	Sb	753	2.0	2.23
114H	4	I	1135	2.3	3.0
114H	4	Te	1135	2.0	2.0
114H	4	Cs	1135	2.5	1.94
114H	4	Ru	1135	2.0	2.34
114H	4	Sb	1135	2.6	2.24

¹Activity counted to determine total material on each plate.

²Midpoint of sampling time.

Table 5.4.3 Mass Median Diameters and Sigma Values
Found by Using the Low-Pressure Cascade Impactor
During Experiments 112U and 113U

(Particle density assumed to be 8.3 grams/cc)

Experiment No.	Sample	Sampling ¹ Time (min)	Mass Median Diameter (cm x 10 ⁵)	Sigma
112U	1	21	1.2	2.3
112U	2	55	1.3	1.8
112U	3	178	1.3	2.2
112U	4	323	1.2	2.3
112U	5	428	1.5	2.5
112U	6	1455	0.73	4.3
112U	7	1880	0.5	3.8
113U	1	15	3.7	2.0
113U	2	61	7.5	4.0
113U	3	154	4.2	2.0
113U	4	445	2.1	2.6
113U	5	672	2.9	2.1
113U	6	1250	1.9	3.2
113U	7	1523	1.8	3.5
113U	8	2652	1.7	6.5

¹Midpoint of sampling interval.

present (108SZr and 114H) have mass median diameters (average) about twice as large as the others.

In Table 5.4.3 for experiments 112U and 113U, the D_{50} values for the particles in 113U are larger (average = 0.32 microns) than those in 112U (average = 0.11 microns). The initial concentration of airborne uranium (U_3O_8) in 113U was about 200 times ($0.62/0.003$) that for 112U (Fig. 5.2.6).

5.5 Sprays

In Table 5.5.1 and 5.5.2 the time, duration, percent activity removed, and volume of spray used are tabulated for CRI experiments 108SZr, 110SI, and 111SI. The data in Table 5.5.3 indicates that in each of these experiments approximately 19% of the iodine removed by spraying came from something other than the airborne. This 19% was probably washed from the walls and floor of the tank. In each of the experiments listed in Table 5.5.3, the values of airborne iodine were reduced to about the same level by spraying (108SZr to 1.1%, 110SI to 1.5%, and 111SI to 0.4%). In all of the experiments in the CRI, where spraying was a part of the operation, the spraying occurred after about 1200 minutes had elapsed (Tables 5.5.1 and 5.5.2), and the concentration of the airborne iodine was nearly in equilibrium.

In Fig. 5.5.1 the effects of spraying on the airborne iodine, antimony, tin, and cobalt in experiment 108SZr are shown. The first spraying ($\frac{1}{2}$ min) is the one with the most effect at reducing the concentrations of iodine.

In Figs. 5.5.2 and 5.5.3 the effects of spraying on the various radiation monitors (Fig. 1.1), as well as on the gas samples, are shown. The Geiger tube and ion chamber values have been normalized to their maximum value during the experiment, while the results of the gas samples are expressed as the ratio of the amount of airborne iodine to the containment tank inventory (C/C_0).

Table 5.5.1 Spraying Operations in CRI Experiment 1083Zr
(Spraying solution = 0.1 M Na₂S₂O₃)

Spray number	1	2	3	4
Time after start of run (min)	1237	1262	1327	1362
Duration of spraying operation (min)	0.5	1.0	2.0	2.0
Volume of spray (tank volumes)*	1.3×10^{-4}	2.6×10^{-4}	5.2×10^{-4}	5.2×10^{-4}
Activity removed (% of tank inventory)				
Iodine	12.9	9.0	0.7	2.6
Antimony	4.2	0.5	27.1	1.2
Tin	0.9	1.4	2.8	0.3

*Tank volume is equal to 4.46×10^6 ml.

Table 5.5.2 Sprays (0.1 M $\text{Na}_2\text{S}_2\text{O}_3$, 0.1 M H_3BO_3) Used in Runs 110SI and 111SI

	110SI		111SI	
Spray number	1	2	1	2
Time after start of run (min)	1223	1253	1302	1345
Duration of spraying operation (min)	2	3	2	3
Volume of spray (tank volumes)*	5.3×10^{-4}	7.6×10^{-4}	5.3×10^{-4}	8.5×10^{-4}
Activity removed (% of tank inventory)	14.9	5.3	13.3	4.7

*Containment tank volume is 4.46×10^6 ml.

Table 5.5.3 Iodine Activity Removed by Spraying

Experiment No.	Total ¹ Removed by Sprays	Airborne ²			Total Minus Airborne
		Before Spraying	After Spraying	Net Removed	
108SZr	25.2	7.25	1.1	6.15	19.0
110SI	20.2	2.3	1.5	0.8	19.4
111SI	18.0	0.46	0.36	0.1	17.9

¹From Table

²Determined from gas samples.

Fig. 5.5.1 The Effect of Spraying on the Airborne Activity in the Containment Vessel, Run 108SZr.

Fig. 5.5.2 Effect of Spraying on Airborne Iodine in the Containment Vessel, Run 110SI.

Fig. 5.5.3 Effect of Spraying in Experiment CRI 111SI.

5.6 Condensates

As stated in Section 3 of this report on "operation," the condensates were drained from the CRI containment vessel at various times during the experiments. The accumulated activity in the condensates, with time, was plotted for each experiment and the values for specified times were taken from the smooth curves drawn through the data. These data are recorded in Table 5.6.1.

5.7 Methyl Iodide

In the gas and impactor samplers, as well as the bypass and recycle filters, the charcoal beds were arranged in an order such that the filtered gas passed through untreated and then treated charcoal. Treated charcoal was impregnated with 2% KI before use to provide ^{127}I so that exchange could take place with any radioactive iodine present as organic (CH_3I).

In the following discussion, "saturated air" will denote air that is saturated with water vapor (100% relative humidity).

Numerous investigators have reported on the inability of untreated charcoal to retain absorbed CH_3I when subjected to a flow of saturated air. This fact has been used by this group as a method for determining CH_3I in the presence of molecular iodine. It was observed that the elution characteristics of CH_3I varied with the time between the end of the adsorption (loading) and the start of the elution treatment. This time period is referred to as the residence time.

In a series of tests, charcoal cartridges identical to those used in the CRI and LOFT gas samplers were exposed to a flow of saturated air containing 1 to 3 mg/meter^3 of tagged CH_3I . The build-up of activity (from $\text{CH}_3^{131}\text{I}$) was monitored during this "loading" operation which lasted for approximately

Table 5.6.1 Total Activity in CRI Condensates with Time
(% of containment vessel inventory)

Run No.	Activity	Time (minutes)										
		50	100	150	200	300	400	600	800	1000	1200	
104I	Iodine	2.4	5.00	6.7	8.0	10.2	11.5	12.7	13.6	14.0	14.1	
105HI	Iodine (HI)	2.7	6.0	7.8	9.0	10.3	11.1	11.8	12.6	12.8	12.9	
106M	Iodine (CH ₃ I)	0.06	0.12	0.17	0.21	0.27	0.3	0.33	0.35	0.36	0.37	
107S	Iodine	3.9	6.0	7.3	8.3	9.5	10.6	11.9	12.9	13.6	14.1	
107S	Tellurium	0.05	0.07	0.09	0.1	0.14	0.16	0.18	0.2	0.21	0.22	
107S	Cesium	0.24	0.5	0.73	1.0	1.5	1.9	2.3	2.5	2.6	2.7	8
107S	Ruthenium	0.95	1.9	2.6	3.4	4.3	4.9	5.5	5.8	5.9	6.0	3
108SZr	Iodine	2.5	5.5	8.0	10.5	15.3	19.5	26.5	31.0	34.2	36.1	
108SZr	Antimony	0.06	0.15	0.24	0.35	0.64	1.0	1.4	1.6	1.7	1.8	
108SZr	Tin	0.05	0.1	0.14	0.2	0.3	0.41	0.86	1.0	1.2	1.3	
109SI	Iodine	16.7	19.2	21.0	22.2	23.9	25.5	28.9	32.1	35.2	37.7	
110SI	Iodine	4.8	8.5	11.1	13.0	15.5	17.0	19.0	19.7	20.6	20.9	
111SI	Iodine	12.0	16.1	18.5	20.5	23.1	24.4	25.9	26.6	27.0	27.2	
114H	Iodine	1.4	2.9	4.3	5.7	8.4	10.0	11.6	12.5	13.0	13.2	
114H	Tellurium	1.5	4.1	6.9	10.5	18.6	25.7	37.3	44.6	48.8	50.5	
114H	Cesium	0.5	1.5	2.8	4.4	8.5	12.5	16.6	18.5	19.6	20.6	
114H	Ruthenium	0.43	1.1	1.9	3.4	6.4	8.1	10.3	11.6	12.3	12.8	

one minute. The loaded cartridges were set aside for a period of time ("residence time") before being subjected to a flow (6.6 liters/min) of saturated air. This sweeping operation was termed the "elution step" and during this step the loss of activity from the cartridges was monitored. In Fig. 5.7.1 the "residence time" of CH_3I on the charcoal is plotted with the percent of the CH_3I that is eluted with a 3 hour sweep of 6.6 liters/minute of saturated air. As can be seen from the figure (5.7.1), the longer the "residence time," the less the amount of CH_3I removed from the charcoal by sweeping with saturated air.

The "elution" method (above) for the estimation of CH_3I was used in CRI 107S and subsequent CRI experiments. After sampling, the charcoal cartridges, after some delay or residence time, were removed from the gas samplers and those containing untreated charcoal were subjected to a flow (6.6 liters/minute) of saturated air for 3 hours and the amount of eluted activity noted. From the elapsed time between sampling and elution (residence), a certain "percent eluted" was selected from Fig. 5.7.1 (i.e., 2 hours residence time corresponds to 70% eluted). Using the percentage corresponding to the residence time (P_e), the activity eluted (A_e) from the cartridge was converted to the amount of CH_3I present when the sample was taken $(\text{CH}_3\text{I})_0$.

$$(\text{CH}_3\text{I})_0 = \frac{A_e \times 100}{P_e}$$

where

$(\text{CH}_3\text{I})_0$ = methyl iodide activity present at time of sampling,

A_e = eluted activity, and

P_e = % eluted (from Fig. 5.7.1).

Using this elution technique on gas sample charcoals, it was possible to determine the methyl iodide concentration with time for all runs containing iodine activity following CRI 107S. The results are shown as a log-log plot in Fig. 5.7.2.

Fig. 5.7.1 Elution of CH₃I From CRI Type Stainless Steel Charcoal Holder.

Fig. 5.7.2 Methyl Iodide Concentration in the Containment Tank with Time for Various CRI Experiments.

6. DISCUSSION OF RESULTS

6.1 Forms of Radioiodine Present in CRI Condensate

Since most of the iodine collected in the CRI condensate was exposed to relatively large amounts of stainless steel surface, it would be logical to assume that most of it would undergo electrochemical reduction and would remain as iodide ions in the collected fractions. The presence of much organic iodide in the condensate was improbable because of its low solubility. In order to demonstrate that this was indeed the case, fresh samples of condensate were extracted with CCl_4 , which would be expected to remove both free iodine and organic iodides. By re-extracting the I_2 with a dilute NaHSO_3 solution, the distribution between the two forms was obtained. Details of this ORNL procedure are reviewed in a LOFT-support document.¹⁰ In Run 109 the partition between the three forms was as follows (sample 3, pH = 8):

Reduced iodine (I^-)	99.95%
Organic iodides (CH_3I)	0.035%
Free iodine (I_2)	0.015%

In Run 110 the partition was even more in favor of reduced iodine, since less than 1 part in 1400 extracted. The pH was 7.2.

6.2 Airborne Radioiodine Species in CRI

The most significant results obtained from the series of aerosol release and deposition behavior tests in the CRI under the LOFT Support Program are probably contained in the information on radioiodine distribution between the postulated chemical state and the fraction of iodine remaining airborne as a function of time. Perhaps unique to this series is the consistent indication of the residual mobility in the radioiodine in equilibrium between the containment atmosphere and

the stainless steel vessel walls. Obviously, then, the deposition did not result in a complete, irreversible conversion to a metallic salt such as FeI_2 although this is probably the species recovered in the condensate. At any rate, from the filter pack sampling and impactor data, the desorbed radioiodine species was mainly reactive toward silver (indicating I_2 or HI) and amounted to as much as several percent of the original inventory. A comparison of most of the runs covering a total iodine concentration range of over a factor of 100 is given in Table 6.2.1. An additional comparison of the fraction of radioiodine existing as particulates is given in Table 6.2.2. The presence of the particulate species equal to about half of the total amount airborne was confirmed by the ultimate test: collection on a plate by aerodynamic inertial impaction. The range in size of the radioactive species corresponded to those found by the electron microscope and to the expected rate of settling according to Stokes' theory. A summary of the radioiodine particulate analyses and some similar data for particulate tellurium and cesium is presented in Fig. 6.2.1. It is not obvious from a comparison of individual run conditions why the radioiodine particulate fraction varied by as much as a factor of ten.

6.3 Rate of Production of Methyl Iodide in CRI

The rate of increase of methyl iodide in the various CRI runs as given in Fig. 5.7.2; Table 6.2.1 gives only the final concentration. The method of detection of CH_3I is unambiguous and involves the established procedure of partial elution by aeration of unimpregnated carbon.

The data in Fig. 5.7.2 and in more detail in Figs. 5.2.7 and 5.2.8 show that a significant portion of the CH_3I is formed rapidly if not promptly. The simulant runs all showed about the same rate of increase or slope; however, in the highly irradiated specimen the rate of increase was significantly greater but had started at a lower point. Thus, the end

Table 6.2.1 Airborne and Desorbed Iodine in CRI Runs

Run No.	Mass of Iodine Released (mg)	Total Time of Run (hr)	Tank Pressure After Aging (atm)	Radioiodine Collected After Aging (% of inventory)				
				Pressure Release	Filter Recycle-1	Filter Recycle-2	Total Airborne and Desorbed ^a	CH ₃ I
104I	30	20.9	1.6	0.18	0.29	0.06	0.53	0.22
105HI	20	21.7	2.4	0.074	0.06	None	0.13	0.04
107S	15	20.6	2.2	0.54	0.42	0.04	1.0 ^b	0.32
108SZr	20	23.2	2.2	0.6	0.62	0.39	1.6 ^c	0.26
109SI	9.2	21.0	1.8	2.3	4.4	1.4	8.1	0.04
110SI	2.4	21.7	1.5	0.39	1.1	0.08	1.6	0.06
111SI	0.23	23.1	1.4	0.08	0.24	0.04	0.4	0.16
114H	1.2	20.2	2.1	0.88	1.11	0.23	2.2	0.12

^aDesorbed iodine may be estimated by subtracting pressure release fraction from total airborne iodine.

^bValue of 3.5% in Fig. 6. either in error or reduced by line loss to the filter.

^cReduced by chemical spray from 7.2% before filtering.

Table 6.2.2 Radioiodine Particulates in CRI Runs from Impactor Analyses

Run	107		108		109			110		111		114			
Sampling time ^a	10	60	15	80	35	160	430	35	450	35	480	35	220	750	1130
Radioiodine present															
Percent airborne at sampling time	40.0	6.9	17.0	12.0	3.5	2.2	3.9	17.0	5.4	2.9	0.86	8.0	3.7	2.3	2.3
Fraction on impactor plates	0.17	0.16	0.76	0.41	0.46	0.41	0.22	0.71	0.88	0.43	0.2	0.54	0.50	0.31	0.23
Percent on particles ^b	6.8	0.8	13.0	4.9	1.6	0.9	0.85	12.1	1.7	1.2	0.18	4.3	1.8	0.71	0.52

^aDuration of sampling was 5 to 15 min; table value represents mid-point.

^bAs measured with the Low Pressure Cascade Impactor.

Fig. 6.2.1 Particulate Iodine, Cesium, and Tellurium from Impactor Samples of Runs 107 and 114.

point for all runs was in the same range of 0.1 to 0.3% of the containment inventory. There is only one point for CSE simulant Run 107, since it was the first in the series and the method for measurement of rate of increase of CH_3I had not been developed; this value is 0.32%. The mechanism of formation suggested is that of a surface reaction involving the iodine plated out on the tank walls.

6.4 Particulate Iodine in CRI from Impactor Analyses

In Table 6.2.2 and Fig. 6.2.1 the directly measured particulate species of radioiodine are summarized for each sample taken. The change in concentration is rather marked in the very early portion of some runs but is relatively consistent with the Stokes settling rate of the solid aerosols (Cs, Te) where these were available to measure. The more striking observation was made in Run 110, which was a prompt burst release at 1100°C, that the particulate iodine fraction was above 10% of the tank inventory (60% release from the capsule). The variations between runs do not suggest any special effect, except perhaps that in the lower temperature release (Run 111) a somewhat lower particulate fraction was observed.

6.5 Effect of Natural Deposition Processes for Radioiodine in CRI Steam-Air Runs

Table 6.5.1 summarizes the effect of two modes of reduction of the airborne mixed species of radioiodine generated in CRI: (1) an initial fast plateout process, and (2) a secondary effect of natural settling of particles that is partly offset by desorption. This distinct change in the radioiodine behavior, which is also shown in Fig. 5.2.2 for all the CRI steam-air runs, presents a strong argument that the controlling step in deposition process in the stainless steel system is a chemisorption phenomenon that is not especially concentration sensitive and is partially reversible. It is also suggested that there is little or no influence of condensation at the

Table 6.5.1 Effect of Natural Deposition Processes for Radioiodine in CRI Steam-Air Runs

Run No.	Conditions	Initial Halftime (min)	Amount Deposited (%)	Secondary Halftime (min)	Amount Deposited (%)	Amount Not Deposited ^a (%)
107S	Simulant + UO ₂ melt	4.5	95.0	260	1.5	3.5
108SZr	I ₂ + SnO ₂ aerosol + UO ₂ melt	4.0	87	132	5.5	7.2 ^b
109SI	I ₂ + partial Zircaloy melt	3.0	93.5 ^c	-	-	6.4
110SI	I ₂ + Zircaloy rupture	3.9	82	186	15.7	2.1 ^b
111SI	I ₂ + Zircaloy rupture	3.0	96.5	145	3.0	0.4 ^b
114H	6000-Mwd/MT UO ₂ melt	4.6	93.0	159	4.3	2.6

^aIncludes all iodine remaining airborne at end of aging period (Figs. 5.2.2-5.2.5).

^bChemical spray used after aging period reduced iodine in Run 108SZr to 1.6% and in Run 110SI to 1% but gave no reduction in Run 111SI.

^cMaximum value of 97% was reached, but desorption rate increased later.

low steam flux available. In this respect the CRI studies are decidedly different from the ADF containment studies¹⁰ at the Pacific Northwest Laboratory, which are strongly affected by a steam condensation process that is one or two orders of magnitude greater.

From the simplest analysis of the plateout process, presented in Fig. 5.2.2, it is evident that there is a small concentration effect at the level of Run 111 (0.04 mg/m^3) which perhaps reduces the desorption process to a minimum. This concentration effect, however, is only realistic at LOFT conditions and cannot be used for full-scale reactor projection.

The intent of the LOFT oriented study,¹¹ however, is to predict both levels of behavior. Since the full-scale reactor would have a painted environment, it is felt that the range of concentrations pertinent to the artificial environment provided by the stainless steel-lined containment vessel has been tested.

The next series of CRI runs will be conducted with painted liners in the containment vessel. It is expected that the steam condensation rate will play a somewhat more important role in these runs, particularly if there is a tendency toward more complete wetting. However, if the non-wetted areas are in proportion to those in the stainless steel-lined vessel, a more complete natural deposition reaction and significantly less desorption may be expected. The production of particulate iodine, however, will probably not be affected, since it appears to be determined only by the generation process.

6.6 Comparison of Deposition Behavior of "Real" and Simulated Fission-Product Aerosols in CRI

A number of the tests described above were conducted to determine the time-space behavior of iodine, cesium, tellurium, and other isotopes with the point of view of qualifying or

validating the "so-called" CSE simulant. In each case the containment vessel was prepressured to about 34 psig with an air-steam atmosphere prior to introducing the simulated fission product tracers. The pressure was then permitted to decay to atmospheric in about 24 hr by normal heat losses through the liner walls. The specific conditions for these tests are described in a separate report.¹²

While we saw difference in the behavior of fission products in these experiments (compare Runs 107S and 114H), we are convinced that these were not due to whether real or simulated fission products were present. Rather, such factors as the chemical environment in which melting occurred and the degree of melting achieved appear to have had great influence.

We concluded that the chemical environment in the melt zone determines the chemical state of the fission products in the fuel and, to a large extent, the percentage of each element which is released. This environment is principally influenced by the composition of the gases present and by the nature of the cladding metal.

6.7 Chemisorption Behavior of Radioiodine in CRI

In the CRI, a typical run shows an initial extremely rapid radioiodine removal process. The total reduction in airborne radioiodine by this initial process, however, may be as little as a single factor of 10 or may approach a factor of 100 (e.g., Runs 107, 108, and 114 in Fig. 5.2.2; in Run 107 the CSE simulant was used; in Run 108, a 2490-Mwd/MT UO_2 clad in Zircaloy; and in Run 114, a 6000-Mwd/MT UO_2 clad in Zircaloy). The identifications of radioiodine species present during the rapid chemisorption period showed that essentially all the airborne iodine was in the molecular reactive state. The predominant form after the initial period was a loosely adsorbed particulate. The radioiodine found in

the condensate was essentially all (99.9%) in the reduced state. Further, it was noted that a relatively small fraction (10 to 30%) was washed off the containment surfaces by the condensate. Both facts tend to support a conclusion that the rapid gas-phase depletion is mainly by chemical adsorption on the moist (not saturated) exposed metal surfaces. Direct visual observation by way of the tank windows also revealed that a light non-uniform accumulation of water was present on the surfaces, and more bare metal was seen without an apparent water film than with it. Variations between runs in the CRI were therefore probably controlled by the unique surface reactivity of the exposed stainless steel, which should be variable. The rate of iodine desorption is therefore believed to be more related to the aerosol source characteristics and to the tank wall temperature than to the partial removal by the water film drainage.

6.8 Comparison of Particulate Natural Removal Rates in CRI and ADF (Stainless Steel Tank)

In order to emphasize this comparison between facilities, the observations made in one of the ADF runs (No. 53)¹⁰ and one of the CDE¹³ runs are combined with those of CRI Run 114 (6000-Mwd/MT UO₂) in graphic form in Fig. 6.8.1. The data have not been adjusted for the fact that ADF has only about one-half the height of CRI, which would double the settling time observed in ADF. On the graph the halftime for natural settling of tellurium and cesium in CRI is about 5 hr, while the observed halftimes for removal in ADF are 7-1/2 to 9 min. This value is also typical for radioiodine in ADF, while in CRI it is significantly shorter (Table 6.5.1) or 4 to 5 min. This indicates that gas-phase removal of iodine is slightly faster on the semi-dry metal wall than on the flooded wall. In the CDE, the deposition rate initially conforms to a high steam flux rate, but then decreases to a rate comparable to CRI.

Fig. 6.8.1 Comparison of Aerosol Removal Rates in CRI (Low Steam Flux) with ADF and CDE (High Steam Flux).

6.9 Comparison of Steam Condensation Rates in LOFT, CSE, NSPP, CRI, and CMF

With the exception of the general model by Watson, Fontana, and Perez,³ most fission-product radioiodine removal models, such as those by Morrison and his associates,¹⁴ Hilliard, Coleman, and McCormack,¹⁰ Ozisik and Hughes,¹⁵ and Yuille,¹⁶ are based on a sufficiently large steam condensation rate to form a continuous water film over the surface of the containment vessel. It is not surprising then that for a given set of conditions in this regime these models predict about the same results. The assumptions made in each case are that the removal of radioiodine is principally due to the dissolution of iodine (I_2) in the water film at a rate that is not too far from equilibrium and which obeys a range of partition coefficients such as those given by Eggleton.¹⁷

A comparison of the actual steam flux rates in several of the major nuclear safety facilities is given in Fig. 6.9.1 from which it may be seen that there are differences of up to several orders of magnitude. This suggests that there should also be a considerable difference observed in the removal rates for each facility and that different processes may actually be involved. For this reason, when comparisons within the same facility between the fission-generated species and the simulant aerosols are made, good correlation is observed; on the other hand, comparisons of the fundamental behavior between the facilities leaves a very different impression.

Fig. 6.9.1 Comparison of Steam Condensation Rates in Nuclear Facilities.

7. CONCLUSIONS

The series of tests in the CRI stainless steel-lined containment vessel indicate that radioiodine is depleted initially from the gas phase by a fast chemisorption process that may leave up to 10% of the radioiodine in a relatively stable airborne state. This is most likely to be a particulate form with a mean mass diameter of 0.1-0.25 microns. The solid aerosols of cesium and tellurium are not depleted by adsorption or diffusion processes nor by the limited CRI steam condensation rate but obey Stokes settling for the particle size distribution according to stirred-settling theory.

The fraction of organic iodide observed is low and variable and appears to be formed at a rapidly decreasing rate, probably in a surface reaction on the walls. Significant amounts of radioiodine may be desorbed from the stainless steel. Up to several percent have been observed, with the desorbed species being reactive molecular iodine. The amount of desorption appears to be related to both the amount of iodine deposited and the amount of solids in the aerosol.

The natural effects phenomena affecting the behavior of real fission-product radioiodine, cesium, and tellurium in CRI show about the same behavior as that observed for the CSE-type simulant. However, the effect of steam condensation transport of particulates (diffusiophoresis) is hardly measurable in CRI because of the very low magnitude of the rapidly decreasing steam flux.

In the CRI the natural removal processes are limited to chemisorption and gravimetric settling. A much higher steam condensation rate would be expected to enhance particle removal by nucleation and diffusive transport to the tank walls; however our criteria for heat transfer did not approach this limit.

Deposition coupons indicated that the iodine activity showed a marked preference for the two painted surfaces (Amercoat and Phenoline) used in the experiments, with the average deposition velocities for stainless steel, Amercoat, and Phenoline being in the ratio of about 1, 4, and 6. Concentration vs time curves derived from gas sampling indicated that all aerosol activities, other than iodine, existed as particulates, and this was substantiated by data from deposition coupons which showed that these activities tended to concentrate on those coupons which were positioned so that they faced upward.

The particulate sizing measurements with the Low-Pressure Cascade Impactor showed that the particles had mass median diameters in the neighborhood of 0.2 microns, with those experiments having molten metal present as a source of aerosol particulates, having slightly larger mass median diameters. Also, in those experiments having the molten material, the mass median diameter of the airborne particles decreased with time. In a given experiment, all radioactive species had about the same particle size distribution.

The spraying operations, using a boric acid-sodium thiosulfate solution, markedly reduced the concentration of iodine activity in the sprayed medium in one to two minutes. The first spray removed the major portion of the activity (iodine), with subsequent sprays removing significantly smaller amounts. There was evidence that particulate iodine was reversibly adsorbed since it was reduced by the chemical sprays, however the particulate solids (Cs & Te) were not removed by spraying.

Methyl iodide concentration in the containment tank atmosphere increased linearly on a log-log plot with time. The maximum value found for any experiment for CH_3I concentration, after 1200 minutes of the aerosol aging time had elapsed, was approximately 0.3% of the iodine inventory. The slopes of the log concentration of CH_3I with log of time curves, with one exception (CRI 114H), were very similar.

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