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AEC RESEARCH AND DEVELOPMENT REPORT

**ACTIVITY CONFINEMENT SYSTEM
OF THE
SAVANNAH RIVER PLANT REACTORS**

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Savannah River Laboratory

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Reactor Technology
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ACTIVITY CONFINEMENT SYSTEM
OF THE
SAVANNAH RIVER PLANT REACTORS

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ABSTRACT

A filtration-adsorption system is installed in the ventilation exhaust of each reactor building at the Savannah River Plant for confinement of airborne particulate and iodine vapor activity that might be released in the highly unlikely event of a reactor accident. Air from the process areas of each building is passed continuously through moisture separators, then through particulate filters, and finally through iodine adsorber beds of activated carbon. The system has the experimentally demonstrated ability to confine more than 99% of airborne particulate activity and more than 99.9% of airborne halogen activity, even with allowance for methyl iodide, under emergency conditions that could exist following a reactor accident. A mechanism for methyl iodide formation was developed from published data for general application to reactor confinement. Under Savannah River Plant conditions, less than 0.0001% of the total iodine inventory in the reactor would be converted to methyl iodide.

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ACTIVITY CONFINEMENT SYSTEM OF THE SAVANNAH RIVER PLANT REACTORS

INTRODUCTION

The activity confinement system for the Savannah River Plant (SRP) production reactors is designed to collect airborne particulate and halogen activity and waterborne activity that might be released from the reactor in the highly unlikely event of a major reactor accident. It represents one of many lines of defense that include preventing accidents, limiting the consequences of accidents, and confining activity that may be released from the fuel and reactor.

The confinement system for airborne activity consists of moisture separators, particulate filters, and halogen adsorbers in the exhaust air portion of the ventilation system. Air is exhausted from the reactor process areas through the filters and adsorbers, and discharged to a stack. The system will not contain noble gases. A 50-million-gallon basin is provided near the reactor building to collect waterborne activity. The system has been in full operation in all reactor areas since 1964.

Development work on the confinement system started in 1960 and has been a continuing major effort of both the Savannah River Laboratory (SRL) and Savannah River Plant (SRP) organizations. Considerable laboratory work was required to develop a satisfactory system that would function properly under possible accident conditions. SRL and SRP personnel participated closely in the design, installation, and operation of the system. Comprehensive evaluations of the performance of the operating system have led to several improvements to increase reliability and efficiency. A substantial technical effort is continuing, and further improvements will continue, as more knowledge is gained.

This report presents the results of this extensive program of development and evaluation of the operating system. It covers:

- A brief review of reactor accidents.
- A description of the confinement system.
- A discussion of iodine compound formation and iodine transport.
- An evaluation of the effectiveness of the confinement system.
- Operating experience.
- A review of development programs.

SUMMARY

The SRP activity confinement system has the demonstrated ability to collect over 99% of airborne particulate activity and over 99.9% of airborne halogen activity under emergency conditions that could exist following a reactor accident. These efficiencies are applicable even after the minor deterioration which the units might incur during operation prior to an accident.

The filtration-adsorption system is on-line continuously during normal operation and requires no manual or automatic operating action when an emergency occurs. A negative pressure is maintained in process areas so that air that could be contaminated in an accident is exhausted through the filters and adsorbers. The system would operate at full air flow even after an accident. It is adequately designed so that even if an exhaust fan, normal electrical power, or reactor room seals fail, particulate or iodine activity will be confined without action by the operator.

A large number of actions can be taken by an operator during an emergency to provide backup protection or to counteract additional failures. Control and monitoring are possible either in the reactor central control room or in the remote control station located about 10 miles from the reactors.

Comprehensive and continuing evaluations of each type of unit in the system ensure good performance of the system. Tests of each particulate filter, before and after installation, with dioctylphthalate (DOP) demonstrated that the filters remove 99+% of submicron particules. Representative moisture separators and particulate filters (which had been exposed to service conditions in the SRP facilities for up to 2 years) successfully withstood exposure to steam and steam-air-entrained water mixtures (fog) for 10 days and still maintained a filter efficiency of 99+%. These tests demonstrated that the combination of moisture separator and particulate filter selected for the SRP facilities will withstand the conditions during any accident that does not seriously damage the reactor building.

Tests of activated carbon before use showed that at least 99.99% of elemental iodine is removed consistently under a variety of adverse conditions. Periodic tests have shown that after more than 2-1/2 years of service the efficiency for removing all forms of iodine that might be released from the SRP reactors and transported to the beds remains greater than 99.9%, even with allowance for penetration of methyl iodide and iodine in particulate matter. The expected performance is summarized in the following table:

Average Iodine Penetration of Activated Carbon Beds^(a)

<u>Mechanism</u> →	<u>I₂(b)</u> <u>Vapor</u>	<u>Particulates</u>	<u>Gasket</u> <u>Leaks</u>	<u>Methyl(b)</u> <u>Iodide</u>	<u>Total</u>
Iodine reaching beds, % of total inventory applicable to mechanism shown	50	<0.0003	50	<0.0001	-
Penetration, % of that reaching the beds	0.02	100	<0.03	100	-
Iodine leaving beds, % of total inventory	0.01	<0.0003	<0.015	<0.0001	<0.026

(a) 50% of the iodine was assumed to remain in the reactor. Average refers to four banks on-line.

(b) Assumes steam-air conditions; penetration would be less for air.

The high efficiency of the installed carbon beds is known because:

- The efficiency of representative samples is measured routinely in laboratory tests to establish that the carbon is an efficient adsorber.
- Leak paths of the installed beds are now measured routinely using "Freon-112"* as a tracer.
- A comprehensive analysis and laboratory test program has characterized the mechanism for collection of iodine under normal and accident conditions.

Evaluation showed that the probability of igniting the carbon in the event of a nuclear accident is acceptably low. In the critical period after a possible accident, air flow of less than 10% of normal will remove the decay heat from fission products trapped on the filters and adsorbers and will maintain the carbon temperature below the measured ignition temperature of 340°C. Two exhaust air fans are always operating and a third fan is available. Power for the fans is fed in a loop circuit from two or three power plants that are widely separated.

* "Freon" and combinations of "Freon-" or "F-" with numerals are Du Pont's registered trademarks for its fluorinated hydrocarbons.

A review, analysis, and correlation of published data showed that less than 0.0001% of the total iodine inventory in the reactor fuel core would be converted to nonremovable compounds of iodine under SRP conditions. The conditions that reduce the percentage of elemental iodine converted to methyl iodide include:

1. In Reactor Vessel

- Intense radiation field which decomposes practically all methyl iodide released from molten fuel.
- Rich hydrogen iodide atmosphere which precludes formation of significant amounts of methyl iodide.

2. Outside Reactor Vessel

- Negligible "aging" because of high once-through ventilation flows.
- Low surface-to-volume ratios.
- Low deposition of iodine on surfaces.
- Relatively clean air and building surfaces.
- Low hydrogen content in fuel.
- Oxidizing and/or condensing steam atmospheres.
- Relatively low temperatures.
- High initial gas-phase iodine concentration.

DISCUSSION

I. POTENTIAL REACTOR ACCIDENTS

The activity confinement system is only one of several lines of defense that limit off-site exposure to radiation or radioactive contamination. It is designed for use in the highly unlikely event that a serious reactor accident occurs. Its purpose is to confine 1) airborne particulate and halogen activity after the activity leaves the reactor hydraulic complex but before it leaves the reactor building and 2) waterborne activity after it leaves the building but before it enters the uncontrolled environment. Multiple lines of defense are provided to prevent accidents and to limit the course of accidents.

This section summarizes the conditions under which activity could be released from the reactor and reviews the bases for the design of the activity confinement system. There are basically only two potential reactor accidents that could lead to a substantial release of fission product activity: 1) loss of coolant followed by fuel melting from decay heat, or 2) an increase in reactor power that would cause boiling of the D_2O followed by fuel melting. Theoretically, each of these can occur and can lead to a fairly wide range of activity releases.

The most serious potential accident is a major D_2O leak followed by a significant delay in actuation of the emergency light-water cooling system. Under these conditions, fuel coolant could be lost in a short time and fuel melting would follow. Activity would be released first to the reactor tank and then to the above-grade process room through numerous vents in the top shield and plenum. Activity could be released also to the below-grade process areas through the leak in the hydraulic system. If emergency cooling is not added rapidly after loss of the D_2O coolant, the entire core could melt from the decay heat. Most of the fuel would be contained within the reactor tank or the piping system because heat transfer would be adequate to prevent melting of the reactor or piping. Only if the leak occurred below the bottom of the reactor tank, could a part of the fuel flow into the below-grade areas of the reactor building and release additional activity. The sequence just described is the only known potential accident that could lead to release of 100% of the equilibrium inventory of noble gases, 50% of the iodine, and 1% of the particulates. This potential accident provides one of the bases for design of the activity confinement system.

Loss of cooling could occur if all the D₂O pumps failed (perhaps caused by light-water flooding of the reactor building) or if light-water flow to the process heat exchangers stopped. In neither case would melting occur unless there was also a serious delay in actuation of the emergency cooling system. If that occurred, fuel decay heat would vaporize the D₂O in the reactor before the full core melted. At first, steam would escape through two vacuum breakers into the above-grade process room, and later (as the tank water level decreased) steam would vent through numerous other openings into the above-grade process room. Some fuel melting could be experienced during this stage; hence, the steam could be contaminated with fission product activity, but most of the core will not melt until most of the D₂O has vaporized. Fuel melting then would take about the same course of action as previously described. The added time required to vaporize the D₂O would allow significant decay of fission product activity and would allow an appreciable time to add emergency cooling. Steam could be generated during the course of this accident, but the rate would be significantly lower than in some reactivity accidents.

For possible power-surge accidents, reactor power could increase and cause boiling of D₂O in the reactor. Steam and water would be vented to the above-grade process room through vacuum breakers and other vent paths. After the start of steam generation, fuel would begin to melt because of vapor in the fuel coolant passages. After enough fuel melts, the power surge would stop because of loss in reactivity. Unless fuel cooling also was lost at the same time (this is considered to be unlikely) no further fuel melting would occur. Any activity released from the fuel would be vented to the above-grade process room. Because only part of the core would melt, release of activity would be less than that for a major loss-of-coolant accident.

The filtration-adsorption system was developed specifically for the SRP facilities to achieve high efficiencies for removal of particulate and halogen activity under the most unfavorable combination of accident conditions. As discussed in Section II, filtration-adsorption units were exposed successively to tests that simulated the worst features of accidents involving potential complete core melting.

II. DESCRIPTION OF SRP ACTIVITY CONFINEMENT SYSTEM

A. REACTOR

The SRP production reactors are 1/2-inch-thick stainless steel tanks,⁽¹⁾ 16 feet 2-3/4 inches in diameter*, 15 feet 4 inches high, as shown in Figure II-1. The tank is surrounded on the sides by an iron and light-water thermal shield, and by a concrete biological shield. Thermal shields, containing light water and stainless steel Raschig rings, are built into the top and bottom of the tank. Above the top thermal shield is a 12-inch-thick gas plenum which contains the top surface of the moderator for the reactor and through which helium blanket gas circulates. Above the gas plenum is a 10-inch-thick D₂O plenum.

Heavy water, used as coolant and moderator, enters the reactor through six symmetrically spaced nozzles at the top of the reactor, flows from the plenum through the fuel assemblies and into the bottom of the reactor tank. D₂O is transported in six coolant loops out of the tank to the pumps, heat exchangers, and back into the plenum.

B. REACTOR ROOM

1. Volume, Surface Area, Surface Finishes, and Materials

Information in this section is used in calculating fission product deposition, organic compound formation, and residence time for airborne fission products. The information presented is for the reactor process room, but comparable conditions exist in other process areas.

The reactor room is in the center section of the reactor building and is approximately 125 feet long and 60 feet wide. The height varies from 43 to 61 feet, and the contained volume is about 320,000 cubic feet. The D₂O plenum and six inlet nozzles protrude above the floor of the room as shown in Figure II-2.

Surface materials, finishes, and areas are summarized in Table II-1.

Concrete surfaces are painted with a vinyl coating, "Amercoat" ** 33HB. Floors are painted annually; however, the ceiling and walls have not been repainted since the original application. "Amercoat" 33HB is used because of its hard finish and ease of decontamination.

* The diameter of C reactor is about 28 inches larger.

** Trademark of Amercoat Co.

TABLE II-1

Surface Conditions in Reactor Room

	<u>Material</u>	<u>Finish</u>	<u>Estimated Area, sq ft</u>
Walls, floor, and ceiling	Concrete	Painted	34,500
Crane haunches	Concrete	Bare	2,500
Forest and misc metal	Carbon Steel	Painted	~5,000
Forest and misc metal	Stainless Steel	Bare	~5,000
Forest and misc metal	Aluminum	Bare	~5,000

2. Structural Limitations

During certain accidents, large quantities of steam could be released from the reactor vessel into the reactor room. This steam must be vented to prevent rupture of the reactor building and the confinement system. The maximum containable flow is primarily a function of: 1) the maximum allowable pressure in the reactor room, and 2) the resistance to flow through the vent paths, i.e., exhaust system and the discharge canal between the reactor room and fuel disassembly area. The test pressure and estimated pressures for gross structural failure are shown in Table II-2.

TABLE II-2

Structural Limitations of Activity Confinement System

	<u>Test Pressure, psig</u>	<u>Estimated Pressure for Gross Failure, psig</u>	<u>Max Pressure During Worst Postulated Accident, psig</u>
Reactor building	-	10 to 15	1.9
Filter compartment			
Momentary	3	6 to 8	0.4 to 1.3 ^(a)
Sustained	2	4 to 5	-
Filter bank supports			
Moisture separators	0.22	3	0.1 to 0.8 ^(a)
Particulate filters	0.22	3	0.1 to 0.4 ^(a)
Carbon beds	0.22	3	0.1
Fan casings	0.4	1 ^(b)	0.07

(a) Smaller value is for clean separators and particulate filters; larger value is for worst expected dust loading on units.
 (b) Estimated from test pressure.

C. PRESSURE RELIEF PATHS FROM REACTOR ROOM

Positive pressures could be created in process areas of the reactor building by a rapid steam release from the reactor. This section describes the passages from the reactor room through which the pressure could be relieved and the areas into which flow could occur; these are summarized in Table II-3. Pressure surges in other process areas are much less likely and would be significantly lower than could occur in the reactor room.

TABLE II-3

Reactor Room Vent Paths

<u>Vent Path</u>	<u>Pressure Seal Rating, ΔP</u>	<u>Adjacent Area</u>
Exhaust duct	-	Activity confinement system
Discharge canal	12 inches H ₂ O	Disassembly area
Monorail transfer	2 psi	Assembly area
Crane door	2 psi	Crane wash and maintenance areas
Personnel access door	2 psi	General personnel areas

Normally, ventilation air flows from the reactor room through sixteen 9-inch x 9-inch concrete ducts at elevation 42 feet 7½ inches, and nine 18-inch round ducts at elevation 60 feet 2 inches. These ducts discharge into a single header. The exhaust ports are on one wall of the reactor room at the ceiling. For pressures of up to 12 inches of water these ports would be the only pressure relief paths from the reactor room, other than leakage. The opposite wall contains the same number of exhaust ports, except that under present operating conditions they are closed.

Air is supplied to the reactor room through twenty-four 18-inch-diameter round ducts; these will not act as pressure relief paths because backflow dampers are installed in the system.

At pressures greater than 12 inches of water, the water seal in the discharge canal between the reactor room and the disassembly area would be broken. Flow rates as a function of pressure differential are discussed in Section II,D,3. Typically, the disassembly areas are about twice the size of the reactor room and are connected to the remainder of the building by unsealed corridors, except below-grade process areas. The ventilation system for the disassembly area is separate from the main process area ventilation system. Air is discharged to the atmosphere and no filter or adsorber units are used.

Seals for all other vent paths from the reactor rooms are designed to withstand a differential pressure of 2 psi. All other areas connected to the reactor room by the 2-psi seals are unsealed and interconnected.

Although seals between the reactor room and adjacent areas might be broken during an accident, gases that may escape in the initial pressure surge were assumed to be clean because of the size of the process room, the relatively long distance of the reactor from these openings, the likelihood that fuel melting will not occur until after the initial steam surge, and the relatively short periods when positive pressures may exist in the reactor room. A negative pressure in the process room would be re-established rapidly by the exhaust fans, because the duration of the pressure surge would be only a few seconds.

D. SUPPLY AND EXHAUST AIR VENTILATION

1. Supply Air

a. Fans

The process areas (reactor room, purification, and -20 foot and -40 foot levels) are continuously purged with fresh, outside air supplied by three sets of centrifugal fans. A schematic diagram of the supply and exhaust air systems is shown in Figure II-3. Figures II-4 and II-5 are schematic diagrams of the supply air system and show the location of the reactor room supply fans (902)* and the below-grade supply fans (911). A fan (907) supplies air to the purification area which is then exhausted by the building exhaust system, Figure II-6. The supply air fans and their function are listed in Table II-4.

* Fans and dampers are identified in the text, tables, and figures by equipment piece number: 902, 907, etc., for fans; and A, B, etc., for dampers.

TABLE II-4

Process Area Supply Air Fans

Fan E.P. No.	Service	Air Flow to Confinement System ^(a) , cfm	No. of Fans	
			Operate ^(b)	Spare
902	Reactor room supply	37,700	1	2
907	Purification supply	26,300	1	0
911	-20 and -40 foot levels supply	64,000	1	1

(a) Design value air flow with clean exhaust air filters.

(b) Normal operation.

b. Dampers and Controls

The supply air dampers are designed to control the flow of air to the process areas. Figures II-3, II-4, and II-5 show the location of many of the dampers, and Table II-5 lists the dampers and their type and class of operation. Several groups of dampers are interlocked to perform automatic action under certain conditions. Following is a brief description of the operation of the dampers.

The controls of dampers in the reactor room supply header (C) and in the actuator tower (E and F) are designed to prevent back-flow from the reactor room. The signal source is the ΔP between the discharge plenum of the reactor room supply fans (902) and the reactor room. Normally the reactor room is at a lower pressure than the fan discharge plenum, but should the pressures become equal, the dampers will close and protect against release of possibly contaminated material to the atmosphere without passing through the filtration-adsorption units.

The dampers between each supply fan (902) and the reactor room supply header are interlocked with their respective fan to open or close as the fan starts or stops. If all three supply fans (902) are stopped, all three of the dampers (A) will be opened automatically.

TABLE II-5

Schedule of Supply Air Dampers

<u>Damper</u>	<u>Class(a)</u>	<u>Location</u>	<u>Size, inches</u>	<u>No.</u>
A	C ₁	Between each supply air fan (902) and reactor room supply header	78 $\frac{1}{4}$ x56 $\frac{1}{4}$	3
B	D	Between prefilters and supply air fans (902)	73 $\frac{7}{8}$ Dia	3
C	A	In reactor room supply header	102x60	1
D	F	In fan room ventilation supply header	50x24	1
E	A	In actuator tower supply	63x25	2
F	B	In actuator tower exhaust	42 Dia	2
T	C ₂	Between supply air fans (911) and -40 foot level	105x75 $\frac{3}{4}$	2

(a) Class of Dampers

- A: Rubber-lined butterfly; modulating and quick closing operation.
- B: Rubber-lined butterfly; two position, quick closing operation.
- C₁: Conventional louver blade; modulating and closing operation interlocked with respective fan.
- C₂: Same as C₁, except no modulating action.
- D: Variable inlet vane; fastened wide open.
- F: Louver damper, manual control.

The modulating dampers in reactor room supply header (C) and in actuator tower supply (E) are used to adjust and balance the air flow to the process areas. They are interlocked to be fail-safe (open fully) if the operating air pressure is lost. If air pressure is lost when the dampers are being positioned by another interlock, the dampers will remain in the position dictated by the first interlock signal.

c. Prefilters

On the suction side of the supply air fans are prefilters designed to remove relatively large particles of dust and to reduce the dust burden on the confinement system's high efficiency particulate filters. The existing prefilters are 5 ply and will remove 70% of 1- μ particles and 81% of 2- μ particles. Provisions

are being made to increase the removal efficiency of the pre-filters with the use of 10-ply filters that will remove 90% of 1- μ particles and 96% of 2- μ particles.

d. Air Ducts and Flows

The supply air fans discharge into a system of steel and concrete ducts that convey ventilation air to the process areas. The supply fans create a pressure of ~4 to 5 inches H₂O at the discharge plenum, which is reduced to a negative value in the process areas by throttling the inlet dampers, and by the exhaust fans that discharge at atmospheric pressure.

Ducts downstream of backflow dampers that could be exposed to a 2-psig pressure surge in the reactor room are designed to withstand this pressure without failure. Figures II-4 and II-5 are schematic diagrams of the supply air system and the flows to the process areas.

The reactor room supply air system is designed to produce a flow pattern that will prevent localized activity concentrations. A concrete header 5 feet x 8 feet-6 inches extends the full length of the reactor room (on one side) and provides 24 outlets to disperse 37,000 cfm of air about the room. The outlets are schedule 10 steel pipe, 18 inches in diameter and about 13 to 20 feet long. An 18-inch-long nozzle is on the end of each outlet and tapers the pipe to 14 inches in diameter.

2. Exhaust Air

a. Reactor Room Exhaust Header

Exhaust air is removed from the reactor room through a concrete header duct, 5 feet x 8 feet-6 inches, located on one side of the room. A similar header is on the opposite side of the room, but is closed. Twenty-five nozzles are open to the header in the reactor room and exhaust the air flow. The nozzles (typified by C Area) are of three sizes and include: sixteen 19 inches x 9 inches x 5 feet in length (concrete), six 18 inches in diameter x about 23 feet in length (schedule 10 steel pipe), and three 18 inches in diameter x about 23 feet in length (schedule 10 steel pipe). About 21 feet of 14-inch x 20-inch duct (galvanized) connects the nozzles to the exhaust header. The exhaust header has a volume of about 16,500 cu ft and a surface area of 12,000 sq ft between the reactor room and the filter compartments.

b. Dampers and Controls

Figure II-6 is a schematic diagram of the exhaust air system and shows the location of many of the dampers. Table II-6 lists the dampers and their type and class of operation.

TABLE II-6

Schedule of Exhaust Air Dampers^(a)

<u>Damper</u>	<u>Class</u> ^(b)	<u>Location</u>	<u>Size, inches</u>	<u>No.</u>
G	A	Between purification area and filter compartments	84x48	1
H ₁	A	Upstream side of each filter compartment	54 Dia	5
H ₂	B	Downstream side of each filter compartment	54 Dia	5
I	G	In exhaust fan (903) leak collection system	16x16	3
J	D	Upstream of each exhaust fan (903)	73 $\frac{7}{8}$ Dia	3
K	C ₁	Downstream of each exhaust fan (903)	78 $\frac{1}{4}$ x56 $\frac{1}{4}$	3
M	A	Between -20 foot and -40 foot levels and filter compartments	138x72	1
N	B	Between -20 foot, -40 foot levels and stack	138x72	1
Q	E	Between -40 foot level and exhaust fans (910)	91 $\frac{3}{4}$ Dia	2
R	G	In exhaust fans (910) leak collection system	24x24	2

(a) See Figure II-6 for damper locations.

(b) Class of Dampers

- A: Rubber-lined butterfly; modulating and quick closing operation.
- B: Rubber-lined butterfly; two position, quick closing operation.
- C₁: Conventional louver blade; modulating and closing operation interlocked with respective fan.
- D: Variable inlet vane; fastened wide open.
- E: Variable inlet vane; automatic control.
- G: Blast gate.

Class A and B dampers, which are butterfly-type with a rubber-lined seat, effectively seal the ducts. Following is a brief description of the operation of various dampers.

Several groups of dampers are interlocked to provide automatic actions that take place under certain conditions. The controls of dampers in the exhaust from purification (G) and in the exhaust from the below-grade process areas (M) are interlocked with the upper air plenum pressure (filter compartment inlet pressure). If this pressure should approach atmospheric (normally -1 inch to -2 inches H₂O), the dampers will close to prevent back-flow of air to purification and -20 foot and -40 foot areas. Thus any contaminated material released from the reactor room, even if accompanied by a pressure surge, cannot reach other areas by the exhaust ventilation ducts.

The dampers adjacent to the filter compartments (H₁ inlet damper and H₂ outlet damper) are interlocked with their respective compartments such that both dampers will be closed when the filter compartment is not in its proper position and/or latched. Electrical switches attached to the filter compartment and latch provide the necessary control signals.

The dampers (K) immediately downstream of each main exhaust fan (903) are interlocked with their respective fan to open or close as the fan starts or stops. However, if all three exhaust fans (903) are stopped, all three of the dampers will be opened.

The modulating dampers are used to adjust and balance the air flow from the process areas. All modulating dampers in reactor room supply header (C), in actuator tower (E), in exhaust duct from purification area (G), upstream of each filter compartment (H₁), and in exhaust duct from -20 foot and -40 foot levels are interlocked to be fail-safe (open fully) if the operating air pressure is lost. If air pressure is lost when the damper is being positioned by another interlock, the damper will remain in the position dictated by the first interlock signal.

c. Filter Compartments

1) Description and Location

The filter compartments contain the filtration-adsorption units. Five compartments are on the roof of the reactor building at the +55 foot elevation, and each contains three banks of filters or adsorbers in series, as shown in Figure II-7. The first bank has 20 moisture separators, the second has 32 particulate filters,

and the third has 32 activated carbon beds. Each compartment is connected to the ventilation system between the exhaust air duct and the exhaust fans (903); hence, the compartments operate with a negative pressure. Each compartment is held in place by a latching mechanism attached to the plenum wall and the compartment. If the compartment is not in proper position and latched, it cannot be put on-line because the interlocked inlet and outlet compartment dampers (H_1 and H_2) will not open. The air flow pattern is shown in Figure II-7.

The filter compartments can be remotely detached from the ventilation system should they become highly contaminated.⁽²⁾ All compartments are mounted on tracks and connected to cables for movement away from the plenums to the edge of the reactor building where they can be removed from the roof by a crane and shipped to the burial ground. Sufficient area is also available on the roof to pull the compartments away from the plenum for service. The compartments contain drain lines to the reactor room to dispose of contaminated water, which could collect in the compartment at the moisture separators (described below) if an accident occurs with a release of steam and/or entrained water.

The three types of units in each compartment are described in the following paragraphs and are shown in Figure II-13. Operating experience of the compartment, filters, and adsorbers is presented in Section V.

2) Moisture Separators

The moisture separators ("Demisters"*) are designed to remove about 99% of 1- to 5- μ entrained water particles to protect against significant rupture of the water-repellent particulate filters.⁽³⁾ The separators consist of about 24 mats woven from "Teflon"*** yarn (individual fibers about 0.001-inch diameter) and stainless steel wire forming a pad 24 inches x 24 inches x 2 inches thick. The number of mats in each pad is dictated by the desired removal efficiency which was correlated in terms of a flow resistance requirement of 0.95 ± 0.05 inch H_2O at the rated air flow of 1600 cfm. The case, mat, and support mechanism are designed and tested to withstand flows equivalent to about 10 times the rated air flow through the separator.

Because no information was available in the literature on the performance of commercial separators for removal of water particles in steam-air mixtures (fog), extensive tests were conducted at SRL on candidate units. These tests showed that the

* Registered trademark of Otto H. York Co.

** Registered trademark of E. I. du Pont de Nemours & Co.

specified "Demisters" were the only commercially available units that were satisfactory at the time of installation in the SRP facilities. The "Demisters" successfully withstood the test conditions (wet steam flow to about 10 times the normal air flow and exposure to fog for 10 days) and prevented damage of the particulate filters when exposed to these conditions. On the basis of calculated efficiencies and the observed test results, most of the water particles in the fog were removed by the "Demister" made of "Teflon". The calculated maximum water load on each "Demister" is about 1 pound of liquid H₂O per minute, and the manufacturer's rating is 8 pounds per minute before flooding occurs.

Tests of standard type prefilters and conventional wire-mesh separators showed that they were practically useless in removing the entrained water particles. One type of prefilter was blown from its containing frame after 10 seconds exposure to wet steam at a maximum rate of 7 times the rated air flow.

The minimum temperature at which thermal degradation of the "Demisters" would be expected is about 150°C. At this temperature the neoprene gasket would begin to deteriorate. The gasket is self extinguishing, however. At about 260°C, the "Teflon" would begin to deteriorate, but the strength of the unit essentially would be unaffected because of the stainless steel wires on which the "Teflon" is woven. Radiation effects on "Teflon" have been measured. Calculations showed that at radiation levels associated with the maximum accumulation of fission products in the filter compartments, the reduced fiber tensile strength is far in excess of the actual stress on the fibers for several hours after an accident.

Complete specifications for the "Demisters" are presented in Appendix A. Operating experience is presented in Section V.

3) Particulate Filters

The particulate filters are designed to remove 99+% of all particles of 0.3 μ diameter, or larger. The necessity for this high efficiency is explained in Section IV, B, 1, b. Filter medium is made of 18-mil-thick glass-asbestos felt sheet which is fire resistant and water repellent with high wet strength characteristics. Under accident conditions the filters could be exposed to hot air, wet steam at 100°C, or fog mixtures of steam, air, and entrained water particles.

Commercially available particulate filters with a water repellent medium will remove 99.9+% of particulate matter from flowing air or other dry gases. Such filters were known to be satisfactory for humid air, but were not designed for wet steam and/or fog. The wet strength characteristics of the various media were not known if the water load occurred after the filters accumulated dust from continuous service. To determine the maximum conditions of steam and fog filtration that newly developed filters could withstand with less than 1% loss in efficiency, four types of tests were performed by SRL^(3,4,5):

- Bench-scale tests with air, water, steam, and fog to screen candidate materials.
- Tests on clean filters with air-water spray, steam, and 10 days of fog.
- Tests on clean and dusty filters in series with "Demisters" with air-water spray, steam, and 10 days of fog.
- Dioctylphthalate (DOP) efficiency tests before and after the above tests on full-size units.

The most important tests were on full-size filters with and without "Demisters". The filters were exposed successively to:

- A burst of wet steam to about 10 times the normal air flow of 1000 cfm for 30 seconds.
- A steam flow of twice the normal air flow for 5 minutes.
- Fog flow at about normal flow for 10 days.

The results of these tests showed:

- Filters with non-water-repellent media collapsed when exposed to steam and fog.
- Two types of commercially available filters with clean water-repellent media satisfactorily survived exposure to wet steam flow.
- Only one commercially available filter with clean water-repellent medium survived the entire sequence of tests.
- None of the filters survived the 10-day fog test when loaded with atmospheric dust and without a "Demister".

Based on these tests, the best water-repellent filter was selected and extensively evaluated with clean and dusty media with a "Demister" upstream. These tests demonstrated that the combination of "Demister" and particulate filter selected would withstand all the tests. These tests simulated the exposure conditions for any accident that would not significantly damage the reactor building. More severe conditions would be expected only when there is so much damage to the reactor building that no practical confinement system would work.

Significant design features and specifications for the water-repellent particulate filters include:

- The filter is constructed by folding the felt sheet over corrugated aluminum inserts into closely separated pleats in a steel frame forming a unit 24 inches x 24 inches x 11½ inches thick. The unit is designed for 1000 cfm air at a pressure drop not exceeding 1.00-inch H₂O when clean.
- Each roll of medium must be qualified by the manufacturer by subjecting representative samples to tensile strength and elongation tests, both dry and wet, and with and without a flat fold in the medium.
- One full-size filter assembled from each roll of filter medium and selected randomly by a Du Pont inspector must pass the following sequence of tests:
 - a) ΔP across the filter must not exceed 4 times the normal when exposed to wet steam flow at twice normal flow and with water spray injected upstream of filter for 15 minutes.
 - b) Flow resistance after drying must not exceed normal ΔP at rated air flow.
 - c) The filter must not fail when exposed to a dust loading of 4 pounds at normal air flow.
 - d) The filter must not fail when subjected to a ΔP 6 times normal for 10 minutes.
 - e) Maximum penetration of DOP must not exceed 0.1% of 0.3-μ particles at rated air flow after completion of all sequence tests.

- Each production unit is tested for pressure drop and DOP penetration by the manufacturer. DOP penetration shall not exceed 0.03% for 0.3- μ diameter homogeneous particles at 20% and 100% of rated flow. Individual units are encapsulated during the test to permit detection of leaks in the gaskets and/or in the frame.
- After installation in the filter compartments, the units are tested with 0.6- μ DOP prior to operation of the compartment. Any filter with a detectable leak (greater than 0.05%) is replaced, because small holes may enlarge if the unit remains in service. The in-place DOP test is repeated annually to determine if leaks develop during service.

The gasket, adhesive, and binder in the particulate filter would begin to deteriorate at about 150°C. However, the efficiency of the filters would not be affected because:

- Significant deterioration would not occur until after the fission products were confined.
- All components of the filter are self-extinguishing.
- The strength of the filter is more than adequate to resist damage even without binder.
- Underwriters Laboratory tests showed that damage to the filter is negligible up to 370°C.

Complete specifications for the water-repellent particulate filters are presented in Appendix B. Operating experience is presented in Section V.

The in-place DOP test was modeled after the technique by the Naval Research Laboratory (NRL)⁽⁶⁾ and the Army Chemical Center.⁽⁷⁾ The DOP smoke generator was built to NRL specifications. The generator consists of a DOP container into which compressed air at 30 psig is discharged through orifices located in the liquid. The resulting DOP smoke (polydispersed aerosol with a mass mean diameter of approximately 0.6 μ) is discharged from the generator to the upstream face of the particulate filters. The air-DOP mixture is passed through the filters and then is analyzed for DOP by a Sinclair-Phoenix smoke penetrometer. Both the penetrometer and generator were calibrated at NRL.

4) Carbon Beds

Activated carbon beds were designed to remove more than 99.9% of elemental iodine vapor that might be released from a reactor if an accident should occur. Numerous tests of small-scale carbon beds showed that more than 99.99% of elemental iodine vapor can be removed consistently with a 1-inch-thick bed of new activated carbon of the type specified for SRP.^(4,8) The tests were run under a wide range of conditions that included:

- Prefiltered fog mixtures (steam-air-entrained water particles) at 65°C and at face velocities to 80 feet per minute (normal face velocity is 65 feet per minute).
- Unfiltered fog mixtures with liquid water entrainment rates up to 5 times that expected downstream of the particulate filters, and with the carbon beds dry or saturated with liquid water.
- Steam at 70 feet per minute and from a wet condition at 100°C to superheated conditions at 130°C.
- Air at 5 times the normal flow and at temperatures to 110°C.
- Iodine loadings to 6 milligrams I₂ per gram of carbon.

Additionally, the tests demonstrated that mass transfer was the controlling mechanism for elemental iodine adsorption under SRP conditions. The results were correlated with the semiempirical equation for critical bed depth developed by Dole and Klotz⁽⁹⁾ and discussed in detail in Reference 8. As shown in Figure II-11, a bed depth of only 0.4 inch is required for an iodine removal efficiency of 99.99% at rated air flow. The design efficiency of 99.9+% for the 1-inch-thick SRP bed is maintained at flows greater than 10 times the rated air flow, as shown in Figure 8 of Reference 8. A 1-inch-thick bed was specified as an added measure of conservatism and to allow sufficient capacity for extended service life in the SRP confinement facilities.

No known commercial adsorber beds could satisfy the test requirements; hence, it was necessary for SRL to design and develop improved beds. The carbon beds developed by SRL and in use at SRP pass all of the above tests. The full-size unit consists of a one-inch-thick bed of activated carbon enclosed between perforated metal sheets, as shown in Figure II-8. The carbon and frame

are subjected to stringent tests by the manufacturer and SRL. Significant design features, shown in Figure II-12, and specifications of the unit currently specified include:

- Type 304L stainless steel construction to retard corrosion. (Beds currently in service are made with frames of cadmium-plated or painted mild steel. These have the same adsorption characteristics as those made of stainless steel, but they have a shorter life (see V,D). Beds made with stainless steel frames are now being installed in all filter compartments.)
- Rigid framing to prevent warping or buckling from mishandling and to withstand steam blasts to at least 10 times the normal air flow. No damage was sustained by either frame or carbon in an 11-day test with flowing steam-air mixtures.
- Spacing fingers external to the carbon bed to maintain the bed thickness at a minimum of 1 inch. Internal spacers are not used because flow channeling at the carbon-metal interface could occur, and spacers may interfere with obtaining a uniformly packed bed.
- 12 separate pleats to retard settling of the carbon during shipment or continuous exposure to flowing air for several years. The pleats are formed by 11 shielded baffles between two continuous sheets of perforated metal. Leaks are possible in units where each pleat is individually removable, but not where the unit is continuous. Shields direct the flow through a tortuous path in the curved portions of the units where minor settling may occur.
- Beds installed with pleats horizontal, to further minimize settling of carbon during service.
- A 1-inch baffle at all sides of the bed formed by omitting perforations at the edge of metal sheet to prevent channeling at the carbon-gasket interface.
- Each carbon bed unit packed with about 57 pounds (minimum) of activated coconut shell carbon that is relatively dust free (97% hardness, ⁽¹⁰⁾ minimum).

- The carbon packed with a gravity system in which the carbon falls a minimum distance of three feet before entering the bed. The bed is vibrated for 1-1/2 minutes after the initial loading operation. This method provides a high packing density and a more uniform distribution of the carbon particles.
- After packing, carbon fines and dust blown from each unit with compressed air at a minimum nozzle velocity of 5000 feet per minute.
- A mesh distribution of 10 x 14, selected because tests showed that occasionally coarse particles could segregate with carbon containing a wider range of carbon particles, i.e., mesh distribution of 6 x 14.
- A perforated face area of about 15 square feet that can pass 1000 scfm of air at a ΔP of about 0.8-inch H_2O . The unit measures 24 inches x 24 inches x 11 $\frac{1}{2}$ inches.
- Physical and chemical tests of representative samples from each batch of carbon made and certified by the vendor.
- Carbon with an ignition temperature greater than 330°C.
- Representative samples of the carbon submitted by the vendor to SRL for analysis. The carbon must remove 99.99% of elemental iodine when exposed to fog at 65°C and flowing at a face velocity of 70 feet per minute and with an iodine loading of 1 milligram per gram of carbon.
- All units inspected by a Du Pont representative for quality construction and freedom from obvious and obscure flaws, before, during, and after charging with carbon. Particular attention is directed to construction details affecting quality of airtight seals and the carbon loading operation.
- Each unit tested for leaks with "Freon" by SRL prior to installation in the confinement system. Beds containing leaks greater than 0.1% of the total flow are unacceptable. Details of the preinstallation "Freon" test⁽¹¹⁾ are given in Section VI, A.

- Carbon beds leak tested in place. The technique was recently developed and demonstrated by SRL. "Freon-112" is used as a tracer to determine if leaks exist in the carbon beds or at gasket surfaces. The test will be conducted on a routine basis. Additional details of the technique are given in Section VI,A.

Activated carbon beds, as well as "Demisters" and particulate filters, have neoprene gaskets which begin to deteriorate at about 150°C. In the ignition test described in Section VI,D,3 on a full-size prototype of the new SRP carbon bed, the neoprene gaskets were not significantly damaged by exposure to an air flow of 70 ft/min at 280°C for 35 minutes.

Complete specifications for the activated carbon bed are presented in Appendix C. Operating experience is presented in Section V.

d. Fans

The exhaust fans (903) are belt-driven centrifugal fans with an electric motor drive. These fans remove the air from the reactor room, purification area, and -20 foot and -40 foot levels during normal operation and discharge it into the stack. During normal operation, two exhaust fans are on-line (in parallel) and one remains on standby. The two operating fans can move the rated flow of 128,000 cfm with a differential pressure of 5.9 inches H₂O. The fan suction pressure is normally -4 to -5 inches H₂O. The fan housings have a positive pressure rating of 10 inches H₂O (about 0.4 psig), but gross failure is unlikely at pressures below 1 psig.

Each exhaust fan has two separate power distribution systems; the electrical distribution systems for the reactor building and for the confinement substation. Either can be selected as a primary source. The systems are supplied by the area powerhouse (except for C reactor) and outside area powerhouses (D₂O-Production Area and South Carolina Electric and Gas). Emergency AC power is also provided to the reactor building distribution system by diesels that start automatically if normal power fails. Automatic switching between normal and emergency power supplies provides a reliable power service to the fans.

e. Stacks

The exhaust air is discharged to a 200-foot-high stack. The stack is of concrete construction with a base I.D. of 17 feet 2 $\frac{1}{4}$ inches and a top I.D. of 16 feet.

3. Flow-Pressure Drop Characteristics

The design air flow for the exhaust ventilation system is 128,000 cfm. The total flow includes 37,700 cfm from the reactor room, 26,300 cfm from the purification area, and 64,000 cfm from the -20 foot and -40 foot levels. Rated flow is achieved with two exhaust fans operating and four filter compartments containing clean filters on-line. Pressure losses in the system are summarized in Table II-7 for the design air flow.

Normally the reactor room is maintained at a negative pressure; however, if an accident should occur a positive pressure could persist for a few seconds. With positive pressure in the reactor room, considerably more air can be vented to the stack through the exhaust header. Figure II-9 shows the flow- ΔP characteristics of the exhaust ventilation system for the reactor room.

The discharge canal has a 12-inch water seal between the reactor room and the fuel disassembly area. If the reactor room pressure exceeds the value of the water seal, significant flow would be vented through the canal. Figure II-10 shows the calculated air flow through the canal for reactor room differential pressures to 5 psi.

TABLE II-7

Pressure Losses in Exhaust Ventilation System at Rated Air Flow

<u>Location of Pressure Losses</u>	<u>ΔP at 128,000 cfm Air Flow, inches H₂O^(a)</u>
Reactor room header	0.77 ^(b)
Filter compartment	.20
Entrainment separators (clean)	.95
Particulate filters (clean)	.83
Carbon beds	.86
Water seal	.44
Fan suction duct	.17
Fan discharge duct	.25
Duct to stack	.20
Stack losses	<u>0.04</u>
Total →	4.71

(a) For 4 filter compartments on-line with clean filters.

(b) For one reactor room header open with an air flow of ~38,000 cfm, measured in R-Reactor Area and confirmed in C-Reactor Area test.

For the worst postulated accident, the maximum pressure in the reactor room would be 1.9 psig. Air would be vented at a maximum rate of about 300,000 cfm each through the exhaust header and the discharge canal. The analysis was based conservatively on air flow through all vent paths, i.e., vent flow would be about 1.4 times greater for steam flow (no credit was taken for heat transfer to the building during the transient). As shown in Table II-2, no damage to the reactor building or activity confinement system would be expected. The exhaust fans would quickly reduce the reactor room pressure to a negative value and no release of activity would be expected, as discussed in Section II,C.

Accumulation of dust on the moisture separators and particulate filters causes an increase in their ΔP and reduces the total ventilation system air flow. Total air flows of about 85,000 to 90,000 cfm have been experienced during normal operation. The increased resistance would not decrease significantly the vent flow during an accident. Reduced air flow during normal operation does not affect the ability to maintain a negative pressure in the process areas. A test in C Reactor showed that a total exhaust system air flow of about 10,000 cfm can maintain the process areas at a negative pressure.

Contaminated air leakage is negligible between the exhaust fans (903) and the base of the stack. Two heavy-duty, flanged joints per fan are located between the fan discharge and the heavy-walled, concrete duct to the stack. Each exhaust fan (903) is in a separate room and the flanges are within the rooms. An air flow of about 2000 cfm is drawn from the room through the blast gates on the suction side of the fans. Air also flows into the fan rooms from the adjacent building corridor. During normal operation, the differential pressure between the fan discharge and the surroundings is less than 0.5 inch H_2O . The reinforced concrete duct leading to the base of the stack is 12 inches thick; it is connected to the stack by steel plates embedded in both the stack and the duct and covered with additional concrete.

E. EQUIPMENT OPERATION

The SRP filtration-adsorption system is continuously on-line during normal operation and requires no manual or automatic operating action when an emergency occurs. Exhaust air from process areas normally passes through the filters and adsorbers and will continue to do so after an accident. In addition,

certain failures can occur but protection from a release of particulate or halogen activity will be maintained without action by the operator. These include:

- Failure of one exhaust fan. Two fans operate continuously, but only one is required to maintain a satisfactory exhaust rate.
- Failure of process room seals. The large exhaust air rate ensures that there will be inleakage of air even if a large number of seals fail.
- Failure of a source of electrical power. The exhaust fans can be operated by electrical power from any of two or three sources of power from outside the reactor building fed through either of two substations (normally each operating exhaust fan is fed from a different source of power and a different substation), or from an emergency, diesel-powered AC generator within the reactor building. Switching among the various sources of power is automatic.

Following an accident, it is planned to operate the exhaust ventilation system at full flow. Careful consideration was given to the possibility of reducing ventilation flow after an accident for the following reasons:

- To increase the decay time of noble gas activity before it is discharged from the stack.
- To increase the length of time during which noble gas activity is released so that if the winds are variable, the activity would be spread over a wider area.

It was decided not to reduce the flow for the following reasons:

- High flow provides more positive assurance of negative pressure in the process areas.
- More cooling is provided for the carbon beds.
- A shorter residence time will minimize deposition of iodine on surfaces and formation of methyl iodide.

- There is additional margin for error at high flow (a fan failure is not as serious).
- There is less likelihood of a mistake being made if no action has to be taken.

A large number of additional actions can be taken by an operator during an emergency to provide backup protection or to counteract additional failures. Control is possible either in the reactor building central-control room or in the remote control station located about 10 miles from the reactors. Some of the possible emergency actions are described below.

The operator has available a large number of instruments and alarms to provide him with the data needed to follow the proper procedure. For the ventilation system some of these located both in the reactor building and in the remote control center are:

- Fan condition (on or off) - an alarm indicates fan failure
- Filter outlet damper position (open or shut)
- Stack total and Kanne activities
- Activity of the effluent from each filter compartment
- Activities on filters and adsorbers
- Various water activities
- Outlet temperature for each filter compartment.

Ventilation system actions that the operator can take in either the reactor building or in the remote control station include:

- Starting or stopping any of the main exhaust (903) fans (one spare is normally available for starting if one of the two operating fans fails).
- Opening or closing the damper downstream of any of the five filter compartments. If a high compartment effluent activity indicates breakthrough or if high effluent temperature indicates fire, the downstream dampers can be closed to prevent loss of activity on the filter or carbon beds.

In addition to the above control actions that can be taken in either control location, any of the many system dampers, supply fans, or other exhaust fans can be controlled from the reactor building.

In addition to the specific controls listed above, a master incident switch in either the reactor central control room or in the remote control center can be manually activated when an emergency occurs. This switch does the following:

- Actuates the light water addition system to provide emergency cooling to the fuel if needed to reduce the likelihood of excessive heating of the ventilation air from fuel decay heat.
- Isolates purification water and gas systems to prevent a spread of activity from the main reactor piping complex. (This action also occurs automatically whenever there is an indication of high activity in the D₂O or helium lines from purification.)
- Stops the supply air fans (902) to prevent an out-leakage of air in case the exhaust fans should fail and to provide better assurance of negative pressure in the process areas. This function has not yet been added, but is planned.
- Actuates various other actions associated with disposition of contaminated water and sources of emergency light water.

F. CONTAMINATED WATER RETENTION SYSTEM

Contaminated water could be discharged into the reactor building from process line breaks, rupture of the reactor tank, or addition of emergency light water. Most of the water released above zero level in the reactor room flows through a floor drain directly to the 50-million gallon basin. Other sources either drain directly or indirectly to the -40 foot level. When sufficient flooding of the reactor building has raised the water level to the -5 foot level (6-7 million gallons for L, K, and C reactor buildings and 15-16 million gallons for P), the water flows by gravity into the retention basin. One basin is located at each reactor area and each basin covers about 4½ acres. The drain line is capable of handling 4000 gpm when only half filled.

The purpose of the retention basin is twofold:

- To retain part of the contaminants in the basin.
- To delay the release of fission products, so that measures can be taken to prevent their release to the ground water or river.

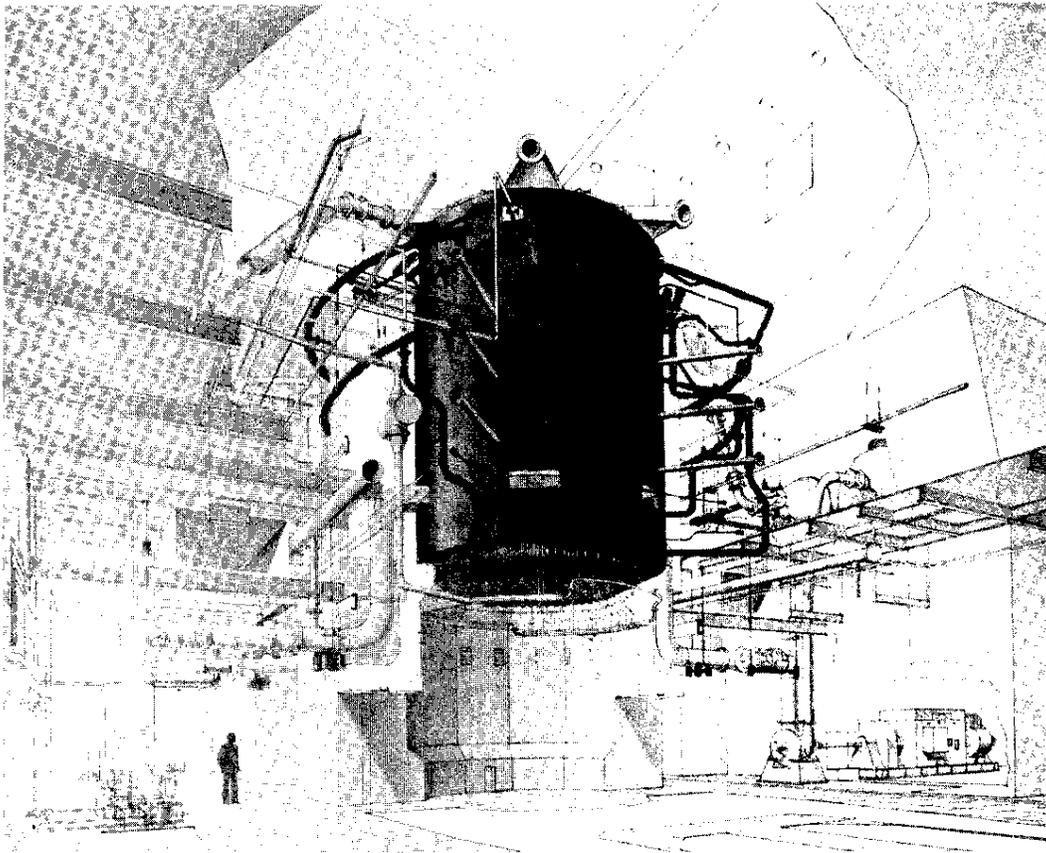


FIG. II-1. SAVANNAH RIVER REACTOR

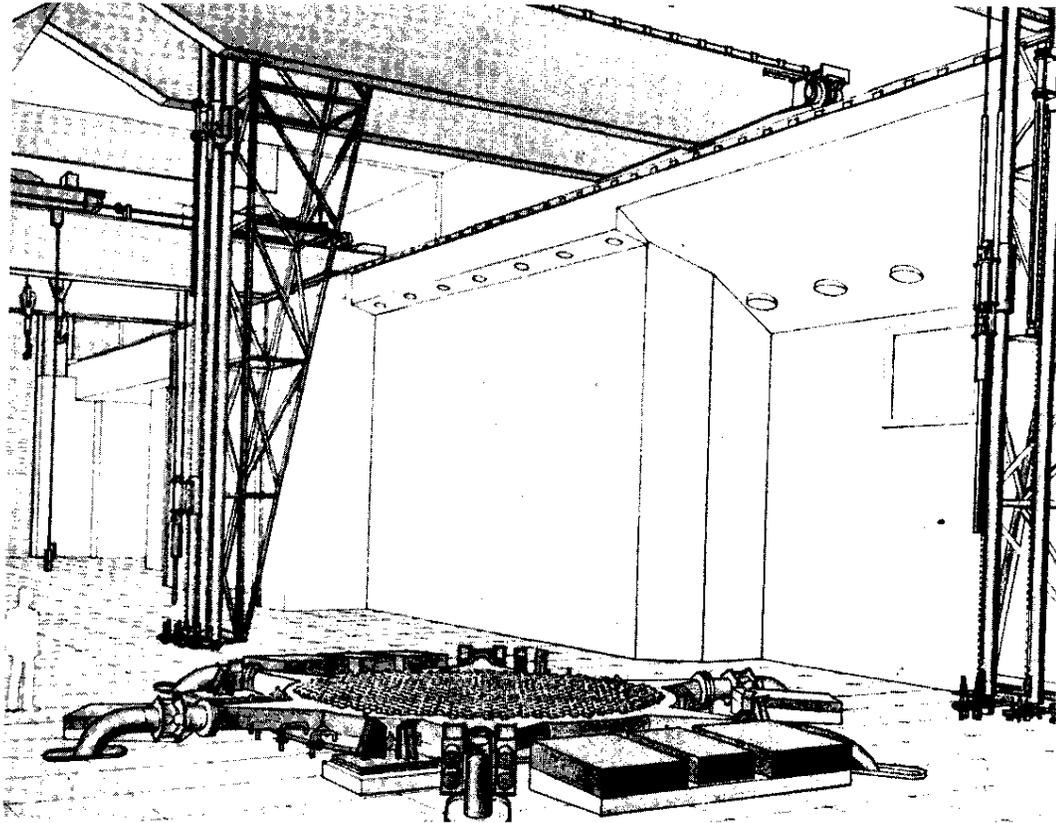


FIG. II-2 SAVANNAH RIVER REACTOR ROOM

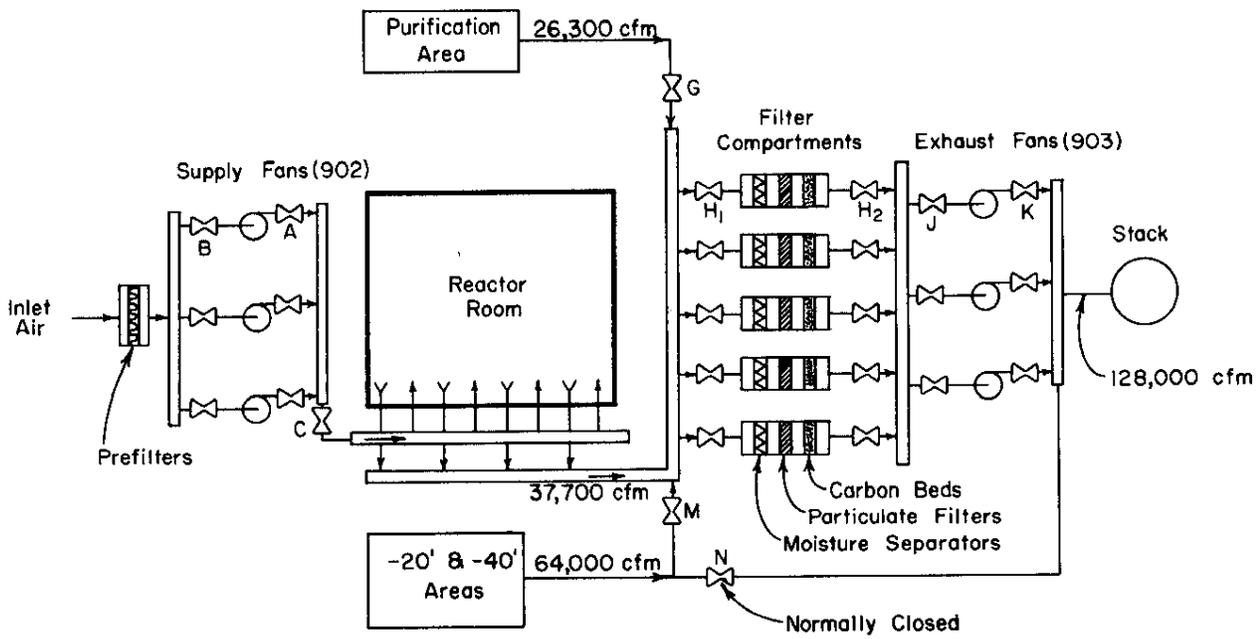


FIG. II-3 FLOW DIAGRAM OF REACTOR VENTILATION SYSTEM

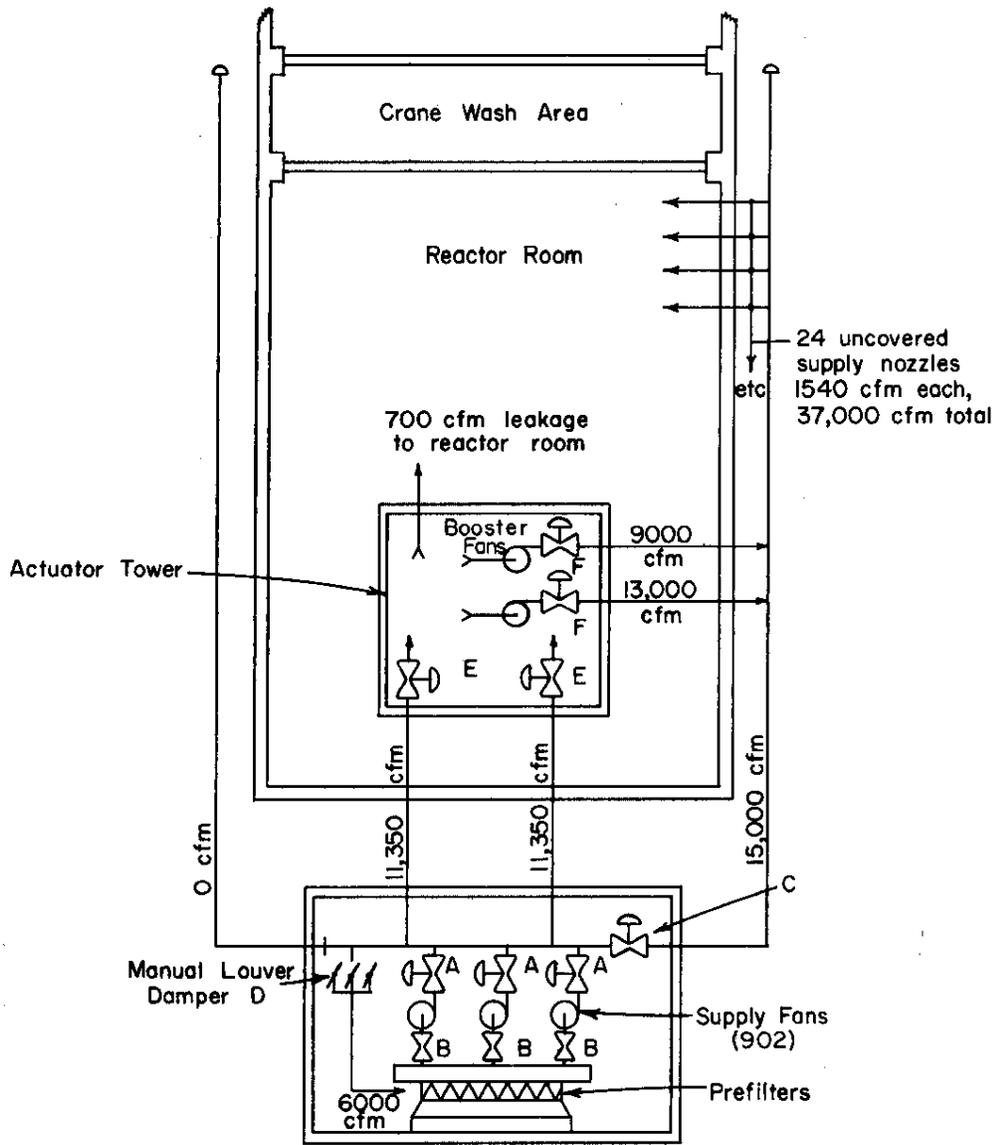


FIG. II-4 REACTOR ROOM SUPPLY AIR SYSTEM

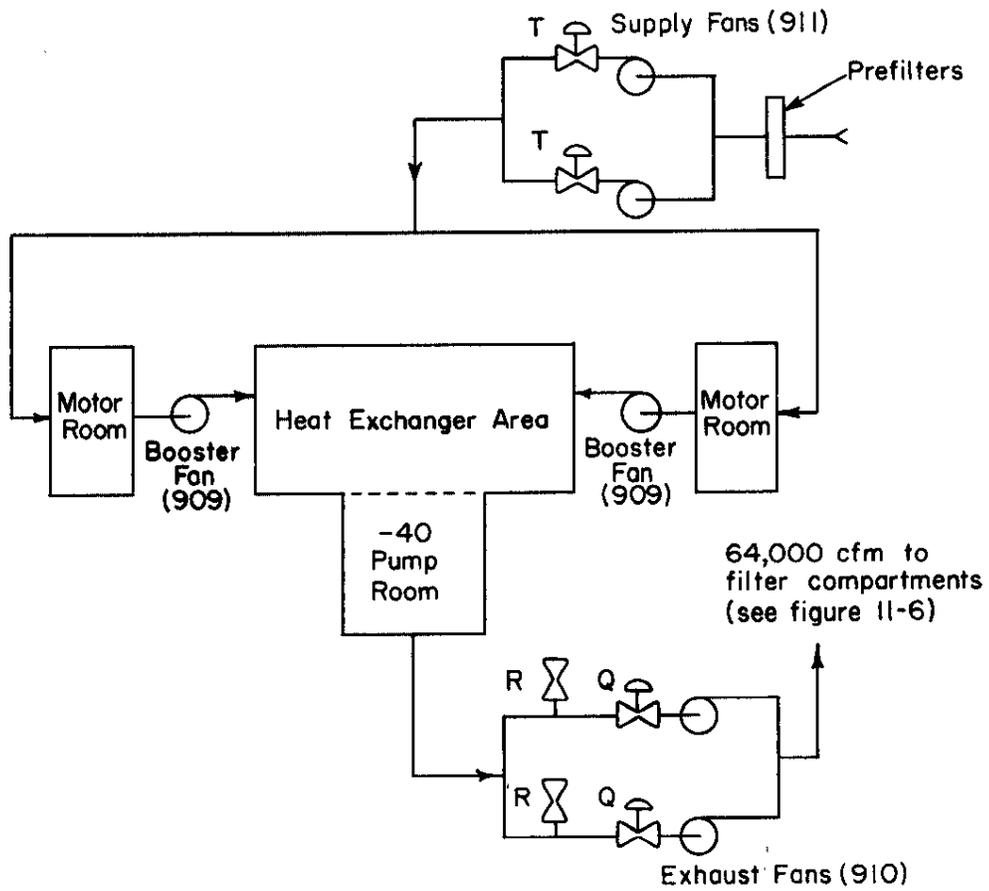


FIG. II-5 VENTILATION SYSTEM FOR -20 FT AND -40 FT LEVELS

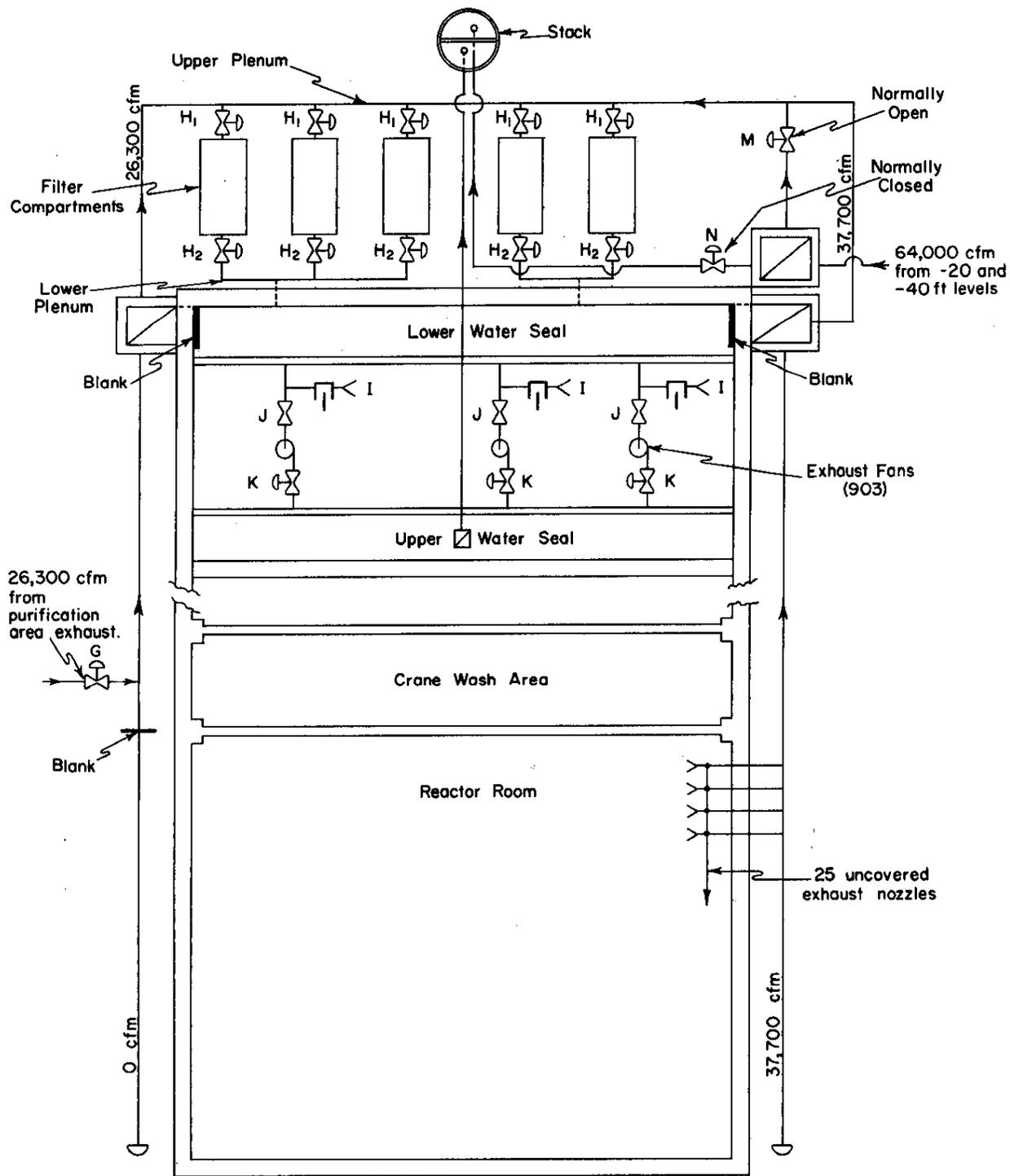


FIG. II-6 PROCESS AREA EXHAUST AIR SYSTEM

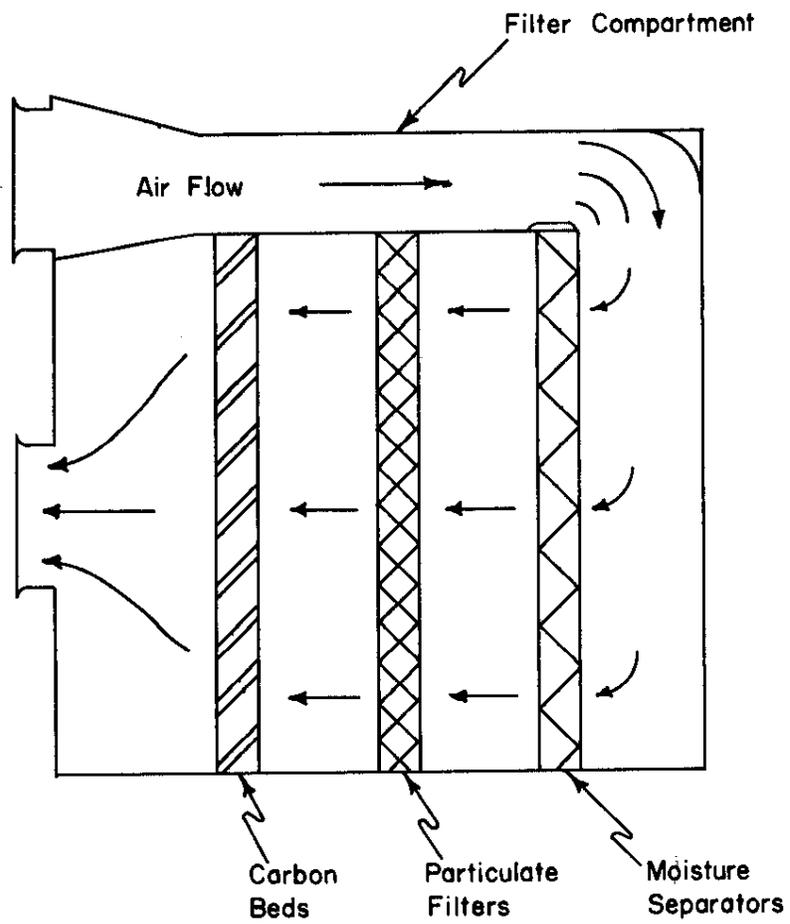


FIG. II-7 FILTER COMPARTMENT
(Elevation Cross Section)

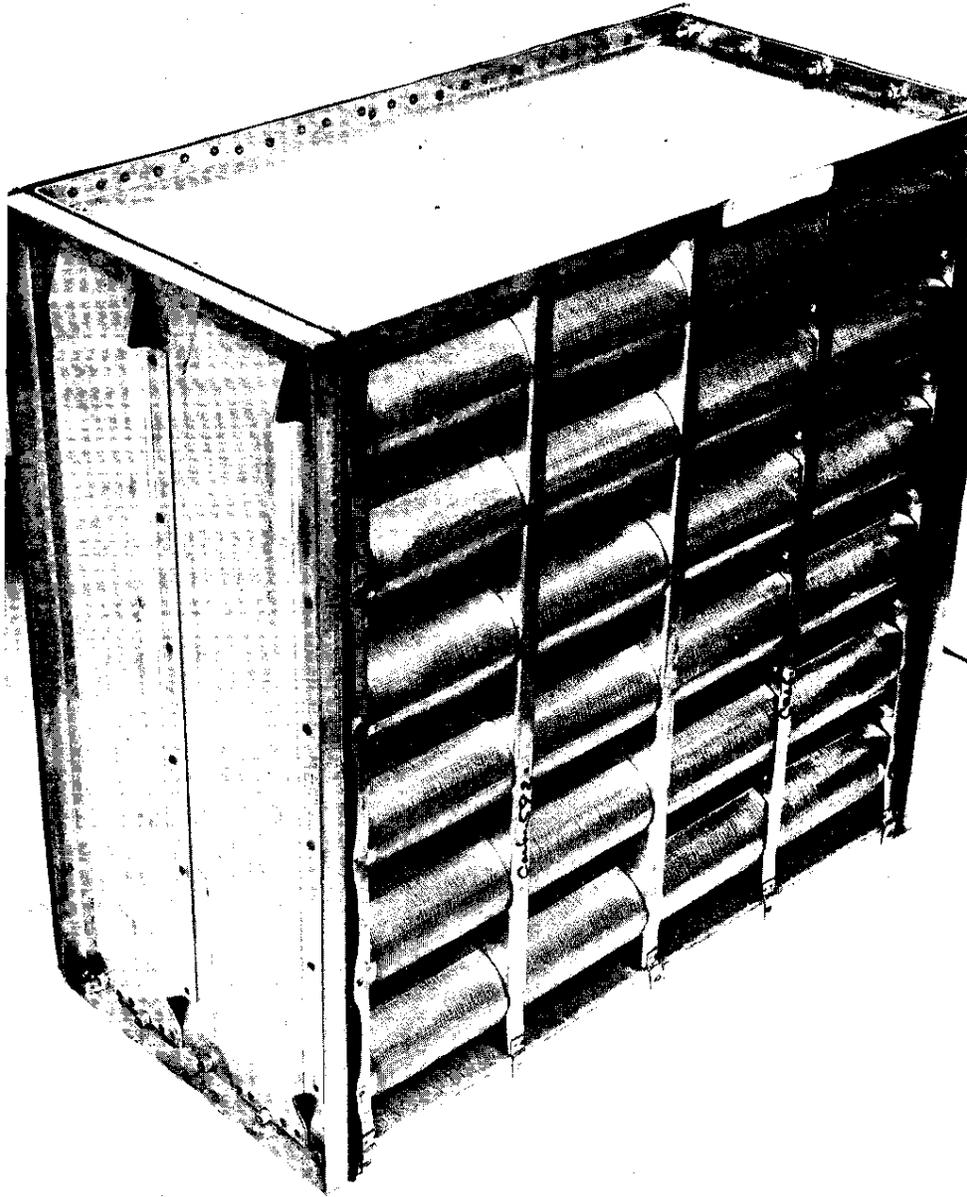
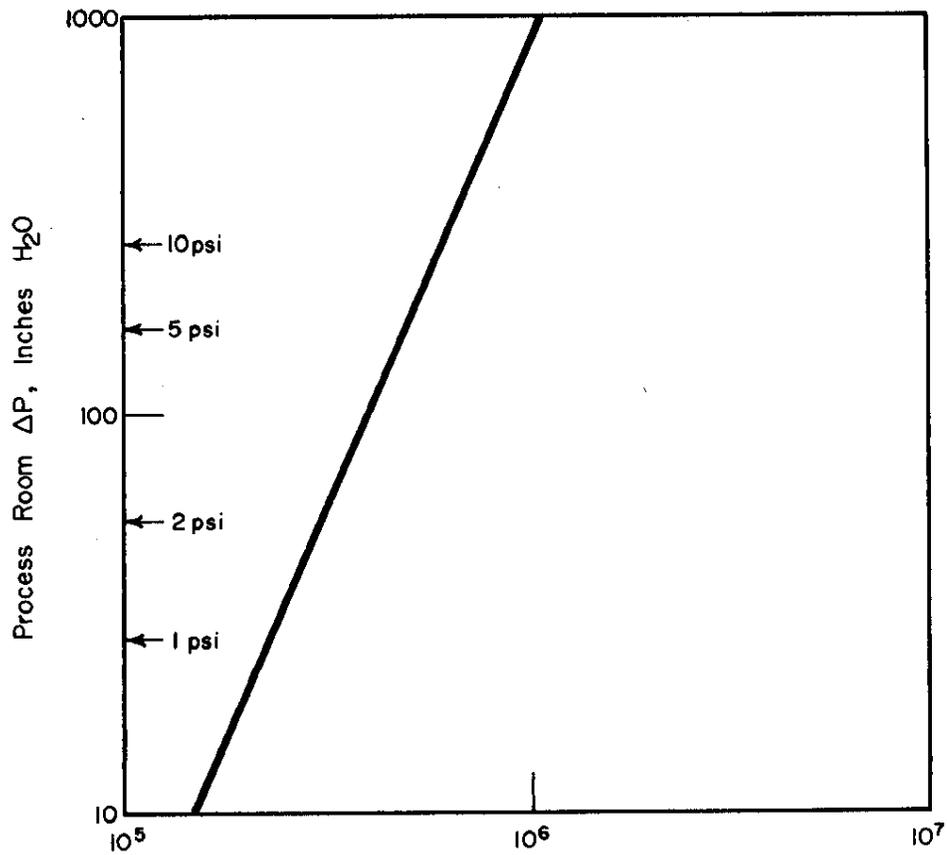


FIG. II-8 ACTIVATED CARBON BED FRAME



Process Room Exhaust Air Flow to Stacks, cfm (steady state)

Bases:

- Air flow at ~15°C & 50% rh and 14.7 psia
- Four filter compartments on line with clean filters
- One reactor room header on line

FIG. II-9 EXHAUST RATE FROM REACTOR ROOM TO STACK

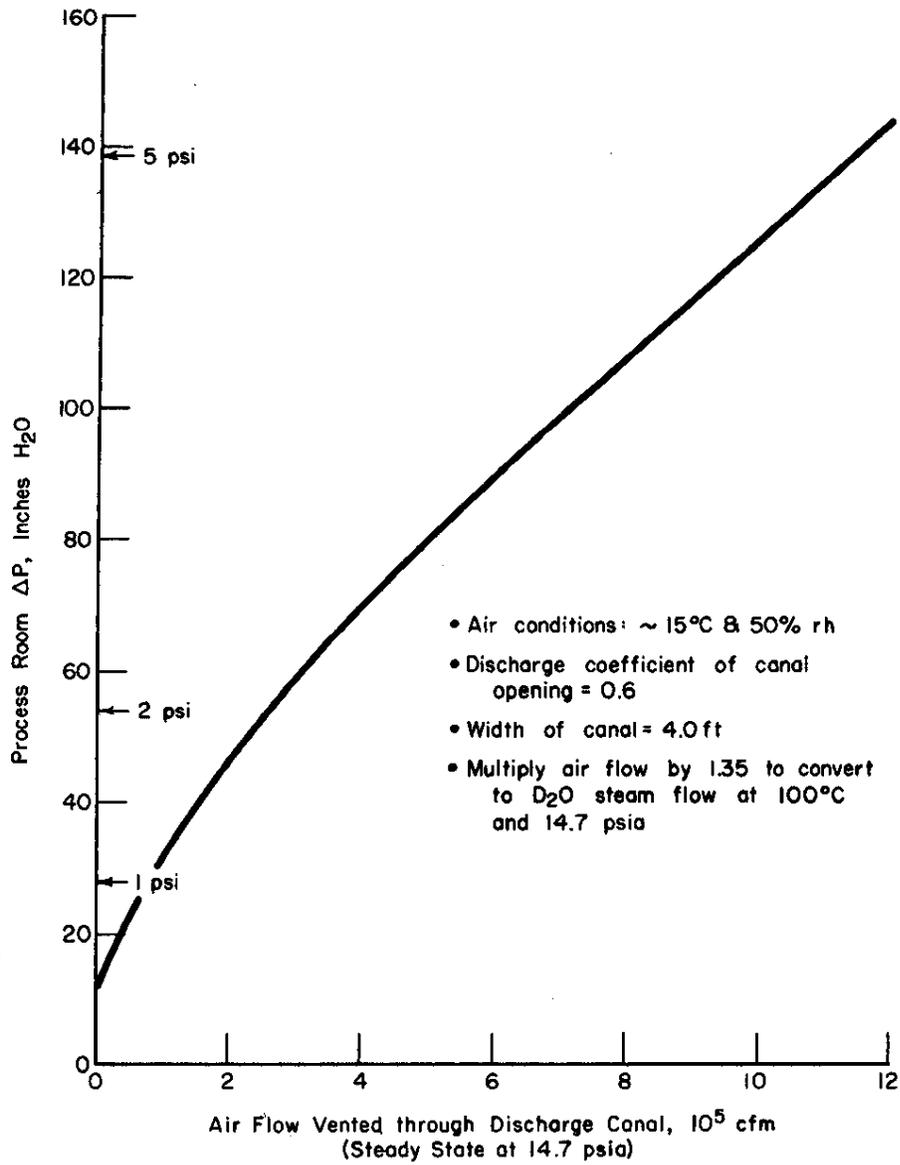


FIG. II-10 VENT RATE THROUGH DISCHARGE CANAL

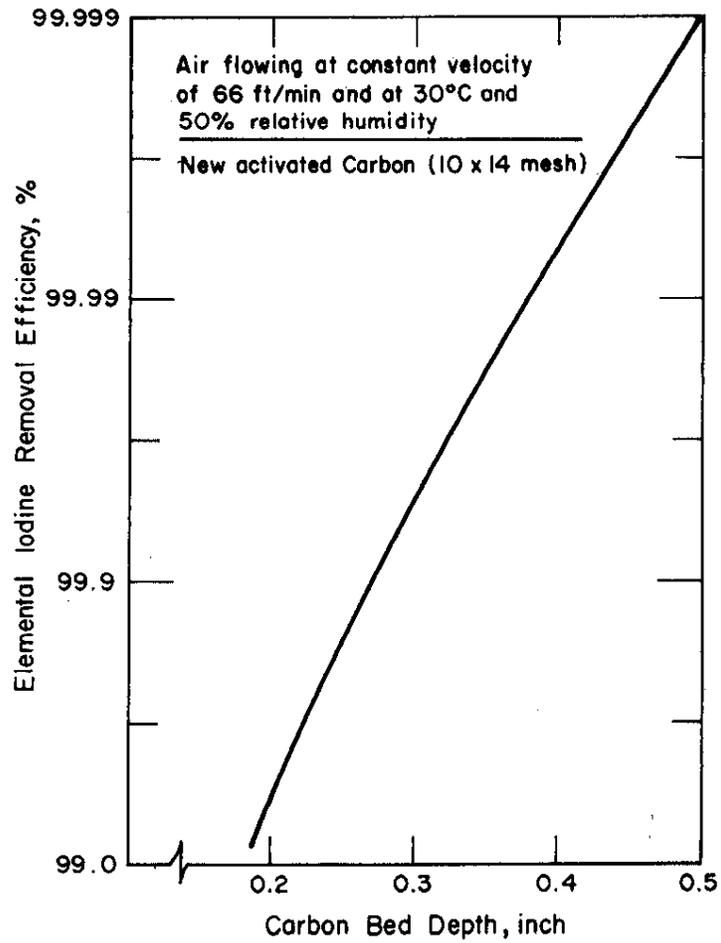


FIG. II-11 EFFECT OF BED DEPTH ON CALCULATED EFFICIENCY OF CARBON BEDS FOR REMOVING ELEMENTAL IODINE VAPOR (Reference 8)

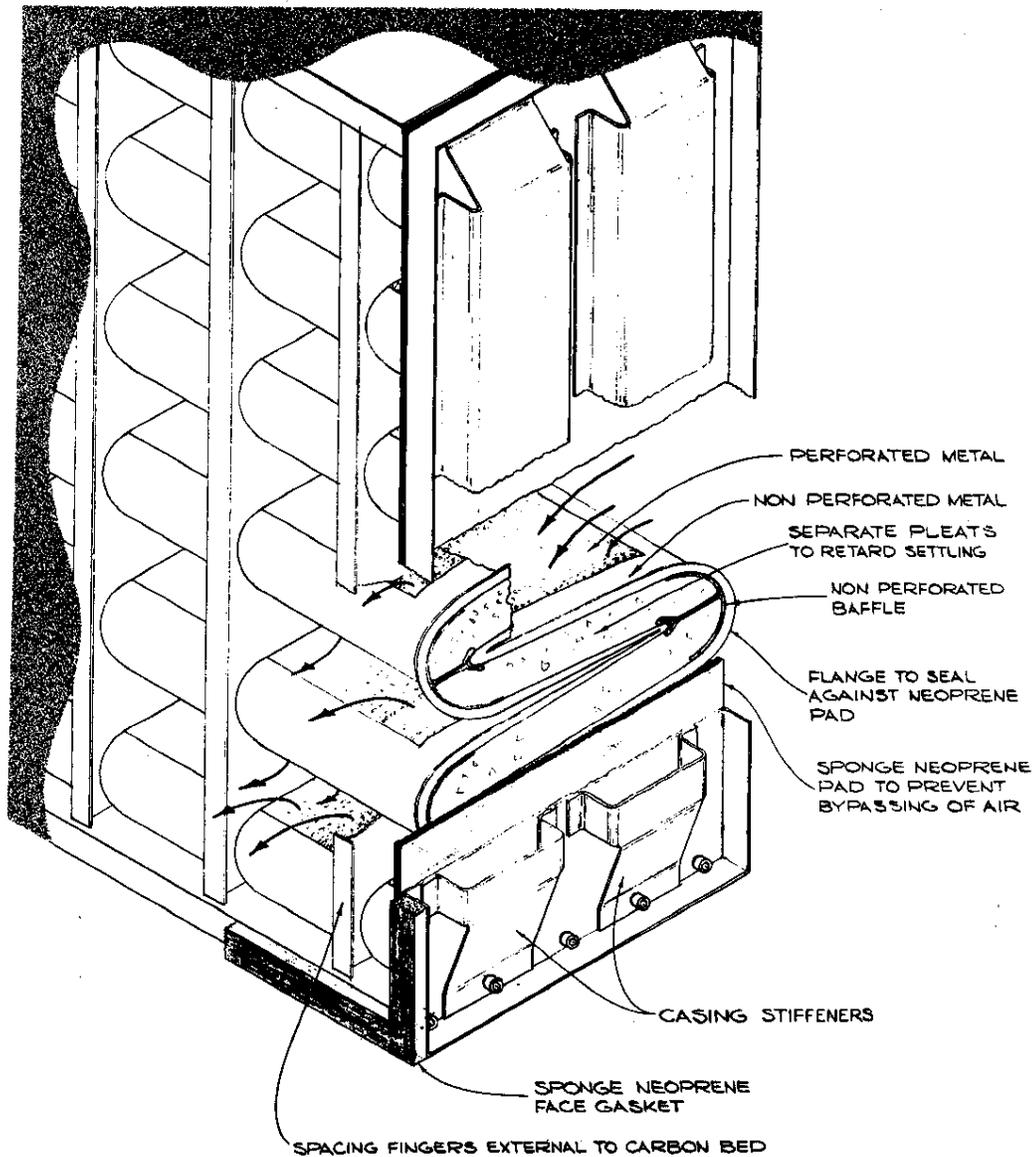


FIG. II-12 SECTIONAL SCHEMATIC OF SRP ACTIVATED CARBON BED

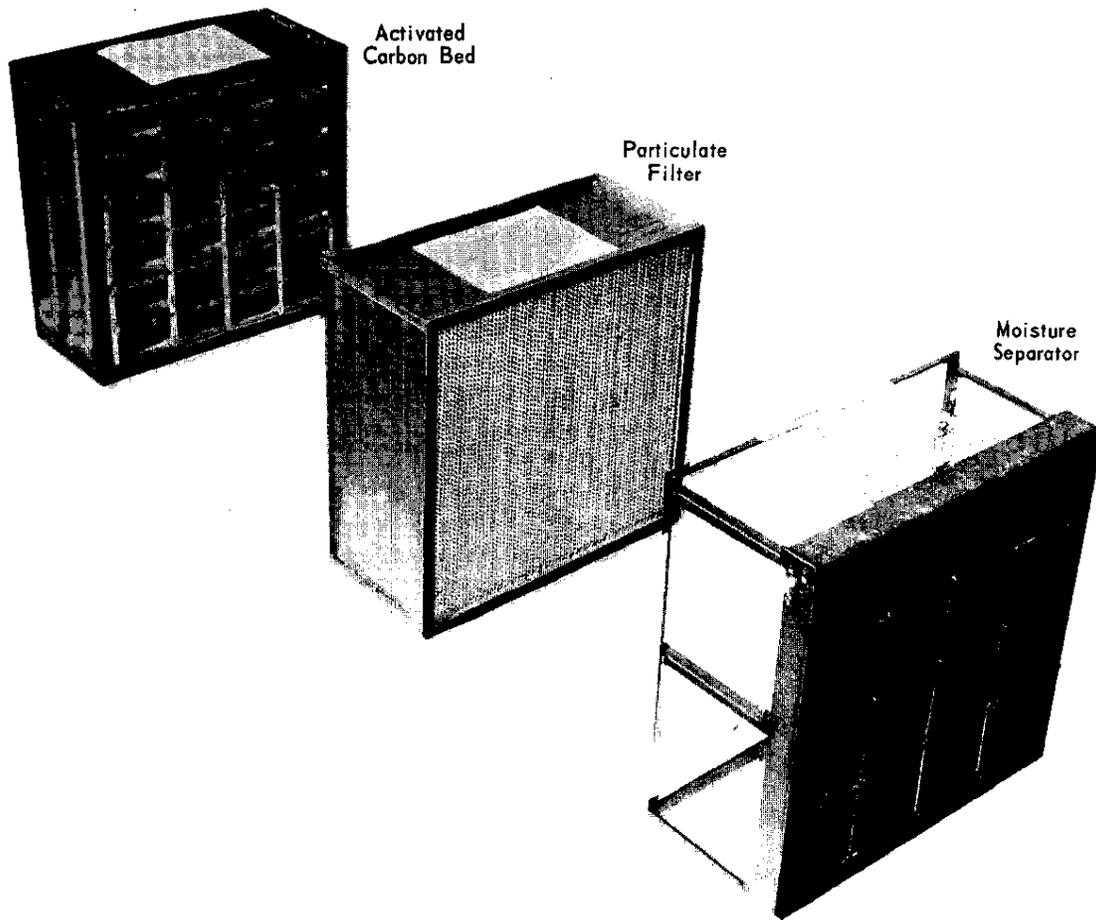


FIG. II-13 SAVANNAH RIVER PLANT FILTRATION-ADSORPTION UNITS

III. KINETICS OF IODINE COMPOUND FORMATION AND IODINE TRANSPORT

In tests at other sites several species of radioiodine were identified in experiments that were designed to determine the chemical form of radioiodine that is released from irradiated fuel under accident conditions.^(12,13,14) Radioiodine in exhaust gases and on surfaces of equipment was identified^(12,13,15,16) as both inorganic compounds, such as elemental iodine (I_2) and hydrogen iodide (HI), and as organic compounds. Several organic compounds were detected, but methyl iodide was the chief organic constituent (about 90%). The fraction of the iodine inventory that appeared as methyl iodide was usually very small, but varied widely under different conditions.

The inorganic species of radioiodine (I_2 and HI) are very reactive and are removed and retained from air streams by activated carbon with extremely high efficiency, even under adverse conditions. On the other hand methyl iodide, which is less reactive than elemental iodine, is removed by activated carbon with less efficiency than iodine, especially from very humid air streams. No penetration of higher homologues of alkyl iodides has been reported in the literature. A knowledge of methyl iodide behavior is necessary therefore for evaluating its likelihood of formation.

The formation and decomposition of methyl iodide under reactor accident conditions are very complex problems. Four aspects of this problem will be treated in greater detail in Section III,A, "Kinetics of Formation of Iodine Compounds":

- Formation of Iodine Compounds in Fuel
- Formation of Iodine Compounds Upon Meltdown
- Formation of Iodine Compounds in the Gas Phase Outside Reactor Vessel
- Formation of Iodine Compounds on Surfaces Outside Reactor Vessel

At present, the precise mechanism for the formation of methyl iodide has yet to be demonstrated, but some experimental evidence exists for the mechanism. This information was combined with thermodynamic and kinetic data in the literature to evaluate the formation of methyl iodide under different conditions. These rates of formation are calculated for ideal conditions. The rates probably would be significantly less under actual reactor conditions.

The information in Section III,A is a general presentation for water-cooled reactors, but the information also has general application for other types of reactors.

Application to the SRP reactors of established theory, existing experimental data, and a correlation developed by SRL for surface reactions involving methane and iodine is presented in Section III,B. In Section III,C, analyses of fission product transport and deposition are presented.

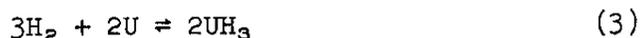
A. KINETICS OF FORMATION OF IODINE COMPOUNDS

1. Formation of Iodine Compounds in Fuel

Uranium metal (U), uranium-aluminum alloy (U-Al), and uranium oxide (UO₂) used as fuel in nuclear reactors contain trace amounts of impurities. Two of the impurities, uranium hydride and uranium carbide, produce some hydrogen gas and carbon during irradiation which can react to form small amounts of organic compounds, such as methane (CH₄), inside the fuel. These compounds can react with iodine, formed during fission, to produce compounds of iodine, such as methyl iodide (CH₃I). In uranium metal used at SRP, the hydrogen content averaged about 1.3 ppm by weight and the carbon averaged about 550 ppm. If all the hydrogen reacted with carbon, 5 ppm methane would form. In uranium oxide evaluated at SRP, the hydrogen content averaged about 0.5 ppm by weight and the carbon averaged 50 ppm. If all of the hydrogen (0.5 ppm) reacted with carbon, 2 ppm methane would form, as follows:



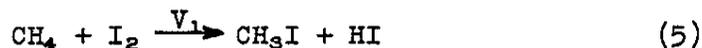
Methane can exist thermodynamically at the conditions inside a fuel element during irradiation. Methane was measured in fission gases that were removed from UO₂ fuel after irradiation at SRP. The methane averaged 0.02 cc per kg UO₂ (0.014 ppm by weight) and ranged up to 0.05 cc per kg UO₂ (0.035 ppm by weight) depending upon the irradiation conditions. These experiments demonstrated that far less than 2 ppm methane would form as expected, because of the immobility of the reactants and because other reactants compete for the carbon and the hydrogen, for example:



The amount of hydrogen and carbon available to react to form methane can be calculated with the knowledge of the various equilibria and equilibrium constants. Methane was not measured in uranium metal nor uranium-aluminum alloy, so the amount of methane in these fuels was inferred from the UO₂ data. Methane

formation in metal fuel is probably lower than in oxide fuel because of reduced mobility of carbon and hydrogen at the lower temperature of irradiation.

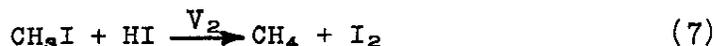
Methane can react with radioiodine formed by fission to produce methyl iodide (CH_3I) and hydrogen iodide (HI) inside the fuel.



The reaction will occur at a velocity V_1 which is equal to the product of the specific reaction rate constant k_1 (a function of temperature) and the concentrations of methane and iodine.

$$V_1 = k_1 [\text{CH}_4][\text{I}_2] \quad (6)$$

Brackets symbolize concentrations of the reactants. Methyl iodide and the hydrogen iodide formed by reaction (5) also react to form methane and iodine,



at a velocity V_2 that is equal to the product of the specific reaction rate constant, k_2 (a function of temperature), and the concentrations of methyl iodide and hydrogen iodide:

$$V_2 = k_2 [\text{CH}_3\text{I}][\text{HI}] \quad (8)$$

At a fixed temperature and at equilibrium, the forward and reverse velocities will be equal,

$$V_1 = V_2 \quad (9)$$

and

$$k_1 [\text{CH}_4][\text{I}_2] = k_2 [\text{CH}_3\text{I}][\text{HI}] \quad (10)$$

so,

$$\frac{k_1}{k_2} = \frac{[\text{CH}_3\text{I}][\text{HI}]}{[\text{CH}_4][\text{I}_2]} \quad (11)$$

At a fixed temperature and at equilibrium, the concentration of each of the reactants and products will be constant, and the ratio of the product of concentrations of products to the concentrations of reactants will be a constant. The latter term is called the equilibrium constant, K :

$$K = \frac{[\text{CH}_3\text{I}][\text{HI}]}{[\text{CH}_4][\text{I}_2]} \quad (12)$$

Thus, with knowledge of the equilibrium constant over the temperature range of interest, one can calculate the concentration of methyl iodide for various concentrations of reactants:

$$[\text{CH}_3\text{I}] = \frac{K[\text{CH}_4][\text{I}_2]}{[\text{HI}]} \quad (13)$$

The equilibrium concentration of methyl iodide was calculated as a function of the average bulk temperature of uranium fuel for a range of methane, iodine, and hydrogen iodide concentrations from measured equilibrium constants.⁽¹⁷⁾ In this calculation, the effect of metals (uranium fuel and aluminum cladding) on the equilibrium was neglected. The effect of metals such as uranium would be to reduce the equilibrium concentration of methane, because uranium metal reacts with carbon to form uranium carbide which is a very stable compound at reactor fuel temperatures (uranium carbide is used for nuclear reactor fuel). Direct combination of carbon with hydrogen to form methane, and diffusion of methane throughout the fuel were assumed. The data are summarized in Figure III-1. The maximum amount of the iodine inventory that theoretically can be converted to methyl iodide based on total conversion of hydrogen to methane is summarized as follows:

Type Fuel	Avg Bulk Temp, °C	CH ₄ , (a) ppm	I ₂ , (a) ppm	CH ₃ I, % I ₂ inventory
U-Al Alloy ^(b)	130	5	2400	0.035

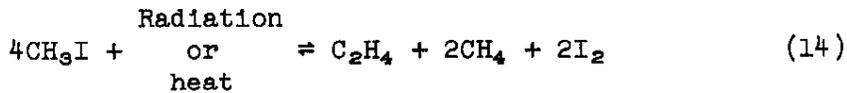
Because of competing reactions (Equations 2, 3, and 4) and the immobility of the reactants, a realistic estimate of methyl iodide in SRP fuel, based on measurements at SRL, as discussed on page III-2, is as follows:

Type Fuel	Avg Bulk Temp, °C	CH ₄ , (a) ppm	I ₂ , (a) ppm	CH ₃ I, % I ₂ inventory
U-Al Alloy ^(b)	130	0.035	2400	0.00067

(a) ppm, based on weight of core material.

(b) For conditions applicable to the Savannah River high flux demonstration, as discussed in Reference 1.

The methyl iodide in fuel also will be reduced by pyrolysis and radiolysis. At high temperature and high radiation fields, methyl iodide decomposes to ethylene, methane, and elemental iodine.^(18, 19)



Upon recombination of these products, some of the iodine will react to form ethylene diiodide:



This reaction not only reduces the methyl iodide concentration, but it has a product that is less volatile than methyl iodide and is more easily adsorbed on carbon.

2. Formation of Iodine Compounds upon Meltdown

The average bulk temperature of uranium fuel would increase rapidly after coolant is lost in a nuclear accident, and melting would occur quickly. The aluminum sheath on the SRP fuel and aluminum-uranium alloy would melt at about 660°C. For off-site dose calculations, it is customary to assume that about 50% of the iodine inventory will be released from the fuel; however, it was conservatively assumed in the analysis for methyl iodide formation that all of the iodine was released into the reactor vessel but only 50% of the iodine inventory was released from the reactor vessel into the reactor building.

At the melting point of uranium, very reactive chemical species exist. These species, free radicals and atoms, ordinarily are only found at elevated temperatures, because a large amount of energy is required to form them. The free radical is frequently indicated as:

$\text{CH}_3\cdot$, methyl free radical

$\text{C}_2\text{H}_5\cdot$, ethyl free radical

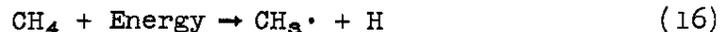
The dot symbolizes the unpaired electron of a free radical.

The atom is indicated as:

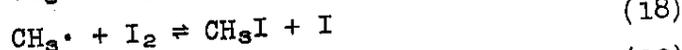
H, hydrogen atom

I, iodine atom

At elevated temperature, organic matter can be decomposed into free radicals and atoms. Intense light and radiation also produce free radicals. For example:



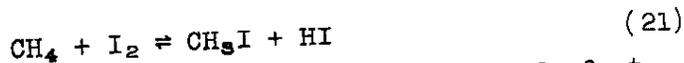
The lifetime of a free radical is very short, about 10^{-9} second, because it reacts so rapidly with its surroundings.



A free radical mechanism has been proposed by Atkins and Eggleton^(12,15) for the formation of organic compounds of iodine. Free radicals as well as iodine atoms can be produced in the vicinity of molten fuel. Although these species could lead to the formation of iodine compounds, the probability is most unlikely because of the low concentration of organic matter in the vicinity of molten fuel and the much greater probability for the reaction of a free radical or iodine atom with other materials (steam, nitrogen, oxygen, or hydrogen) that are present in much higher concentrations. In the presence of oxygen, oxidation of free radicals is the predominant result of intense radiation. It is not necessary that oxygen be present in stoichiometric proportion; as long as any appreciable amount is present in the mixture, oxidation is certain⁽²⁰⁾.

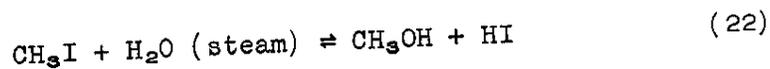
Attempts to prepare methyl iodide by the free radical mechanism were unsuccessful.⁽²¹⁾ Irradiation at 200°C with a 10,000-curie cobalt source formed free radicals, but only trace quantities of methyl iodide were formed.

The equilibrium

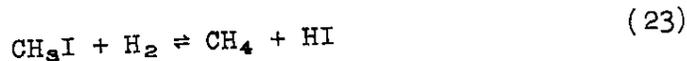


is strongly in favor of methane and elemental iodine. In fact, hydrogen iodide reacts rapidly with methyl iodide to form methane and elemental iodine.⁽²²⁾ To a lesser extent, hydrogen iodide reacts with metal which would shift the equilibrium toward the right.

Upon meltdown, most (if not all) of any methyl iodide formed in the fuel would be decomposed by chemical reaction, radiolysis, and pyrolysis. In water-cooled reactors, the steam resulting from the contact of hot metal and water hydrolyses methyl iodide as follows:



Large excesses of steam will tend to force this equilibrium to the right. Hydrogen produced by the reaction of hot metal and water will also decompose methyl iodide:



A small amount of the molten fuel will react with water inside the reactor tank to produce hydrogen (deuterium) gas.



Uranium oxide does not react with water to form hydrogen; however, aluminum, zirconium, or stainless steel cladding will produce hydrogen by reaction with water. The hydrogen reacts rapidly with molecular iodine to form hydrogen iodide. From published kinetic data⁽²³⁾, it is calculated that the entire inventory of iodine would react with an equivalent amount of hydrogen in about 0.1 second at the temperature of 700°C that is estimated for the atmosphere inside the reactor tank following a major accident with uranium-aluminum fuel. The intense radiation field inside the tank should accelerate Equation 25 so that equilibrium would be attained in even less time than that required for the thermal reaction. Hydrogen, generated by the reaction of less than 0.01% of the metal with water, is sufficient to convert almost all of the iodine inventory to hydrogen iodide.



At conditions inside the reactor vessel this equilibrium is shifted far to the right so that the environment is rich in hydrogen iodide. About 95% of the iodine was calculated to form hydrogen iodide. The equilibrium constant for Equation 25, shown in Figure III-2, was calculated by:

$$K = \frac{[HI]^2}{[H_2][I_2]} \quad (26)$$

The hydrogen also reacts rapidly with atomic iodine to form HI:



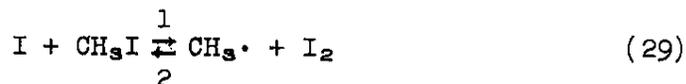
The rate of formation of methyl iodide by the reaction of methane with iodine in a reactor vessel is shown in Figure III-3. The methane concentration was chosen on the conservative assumption that 0.001% of the carbon in the molten U-Al alloy and other components reacted with hydrogen formed by chemical reaction of water with fuel or cladding. The iodine concentration was based on conversion of 95% of the iodine inventory to hydrogen iodide by reaction with hydrogen. Only 5% of the iodine inventory is available to react with methane to form methyl iodide. Under SRP conditions, about 20% of the iodine inventory would be required to convert almost all of the methane in the reactor vessel to methyl iodide. However, as discussed later, the intense radiation

field will decompose essentially all of the methyl iodide. The specific rate constant for the methane-iodine reaction is shown in Figure III-4.

If the concentration of methyl iodide released from the fuel into the tank is greater than the equilibrium concentration, methyl iodide will decompose until the equilibrium concentration is reached. On the other hand, if the methyl iodide concentration is less than the equilibrium concentration methyl iodide will form. These conditions are true provided that the reactants and reaction products (Equation 21) do not change, which would alter the equilibrium relationship, as discussed later.

Hydrogen iodide produced by the above reactions reacts very rapidly and quantitatively with methyl iodide to form methane and elemental iodine. The overall rate constant for the reaction is 1.89×10^{-2} liter/(mole)(sec),⁽²²⁾ at 320°C. This reaction is catalyzed by iodine atoms and involves a free radical. The rate of decomposition of methyl iodide by reaction with hydrogen iodide is shown in Figure III-5.

The rates shown in Figure III-5 were calculated from data reported in the literature.⁽²⁴⁾ The proposed mechanism is:



where, under most conditions step 4 of Equation 30 is negligible. The rate equation for this mechanism (neglecting the reverse of Equation 30) is:

$$-\frac{d[CH_3I]}{dt} = \frac{d[I_2]}{dt} = \frac{k_1 k_3 K_{I_2}^{\frac{1}{2}} [CH_3I][HI][I_2]^{\frac{1}{2}}}{k_2 [I_2] + k_3 [HI]} \quad (31)$$

where:

$$-\frac{d[CH_3I]}{dt} = \text{rate of decomposition of methyl iodide, moles/(l)(sec)}$$

$$\frac{d[I_2]}{dt} = \text{rate of formation of iodine, moles/(l)(sec)}$$

$$\log k_1 = 11.20 - \frac{20,000}{4.575T}, \text{ rate constant for step 1, Equation 29, } 1/(\text{mole})(\text{sec})$$

$$\log k_2 = 10.00 - \frac{1500}{4.575T}, \text{ rate constant for step 2, Equation 29, } 1/(\text{mole})(\text{sec})$$

$$\log k_3 = 9.50 - \frac{2300}{4.575T}, \text{ rate constant for step 3, Equation 30, } 1/(\text{mole})(\text{sec})$$

K_{I_2} = dissociation constant for iodine, moles/l. The terms enclosed by brackets are the concentrations of the compounds.

T = absolute temperature, °K

The effect of a hydrogen atmosphere on the formation of methyl iodide has been demonstrated experimentally.^(13,14,25,26) In one group of experiments, steam was passed over an inductively heated sample of irradiated uranium metal or uranium dioxide. Released fission products and steam passed through a condenser and a series of liquid absorbents. The chemical state of iodine released was determined by chemical analysis of the different liquids. The methyl iodide released from uranium metal (where the metal-water reaction produces a hydrogen-rich atmosphere) was less than 0.3 of that⁽¹⁴⁾ from uranium dioxide (hydrogen-deficient atmosphere).

Methyl iodide also is decomposed very rapidly by radiation. The course of radiolysis⁽¹⁸⁾ is similar to that for pyrolysis that is discussed below.



Because one of the products is ethylene, the usual equilibrium

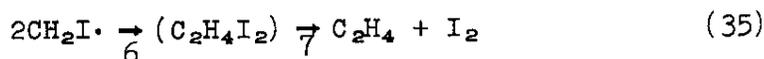
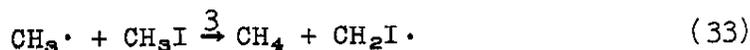


will be shifted toward greater decomposition of methyl iodide. Methyl iodide decays exponentially in a combined oxidizing and radiation environment. The extent of decomposition is independent of the dose rate, but is proportional to the total absorbed dose. Experiments⁽²⁷⁾ with methyl iodide concentration of 10^{-8} to 10^{-6} moles/liter (10^3 - 10^5 $\mu\text{g}/\text{m}^3$) in dry air at 15°C showed that over 99% of the methyl iodide was decomposed at a total absorbed dose of 8×10^5 rads. The radiolysis products observed in these experiments were methyl nitrate and methyl nitrite. The formation of products such as methyl nitrate and nitrite will decrease the equilibrium concentration of methyl iodide. At the high

temperature inside the reactor tank these radiolysis products probably will react with each other and form other new products. These reactions will also shift the equilibrium in the direction of lower methyl iodide concentrations. It is also possible that some materials may slightly reduce the rate of decomposition of methyl iodide in a radiation field. The radiation field from fission products at shutdown is usually greater than 4×10^6 rads/hr per megawatt of thermal power at saturation. Hence, the radiation can be expected to decompose over 99% of the methyl iodide per second per 1000 MW of thermal power.

Methyl iodide is also decomposed by heat. From 300-400°C, saturated alkyl iodides (such as CH_3I) appear to pyrolyze with moderately rapid rates.⁽¹⁸⁾ A study of literature data on the kinetics of the pyrolysis of CH_3I is underway at SRL.

Although no detailed study of the pyrolysis of methyl iodide has been reported, some predictions on this decomposition can be made from recent work by Benson.⁽¹⁸⁾ The important steps should be:



The overall reaction would occur in two stages:



Reactions 36 and 37 will reduce the concentration of methyl iodide. The initial rate of production of CH_2I_2 would have a rate law:

$$\frac{d[\text{CH}_2\text{I}_2]}{dt} = K_{\text{I}_2}^{\frac{1}{2}} k_3 K_{1,2} \frac{[\text{CH}_3\text{I}]^2}{[\text{I}_2]^{\frac{1}{2}}} \quad (38)$$

where, in consistent units:

$$\frac{d[\text{CH}_2\text{I}_2]}{dt} = \text{the rate of formation of } \text{CH}_2\text{I}_2$$

K_{I_2} = the equilibrium constant for the dissociation of iodine

k_3 = the specific reaction rate constant for reaction Equation 33

$K_{1,2}$ = the equilibrium constant for Equation 29

$[\text{CH}_3\text{I}]$ and $[\text{I}_2]$ are concentrations of methyl iodide and elemental iodine,

At 400°C, this reaction would have a half life of about 3 hours when the methyl iodide concentration is 5×10^{-3} mole/liter ($7 \times 10^8 \mu\text{g}/\text{m}^3$). The reaction is expected to be much faster at elevated temperatures, because the rate usually doubles for a 10°C rise in temperature. The activation energy would be about 44 kcal.

The second process would be about one-tenth as fast, and would have an activation energy of about 67 kcal. Its rate law would be of the form:

$$-\frac{d[\text{CH}_2\text{I}_2]}{dt} = \frac{2k_3K_{5,4}^2K_{\text{I}_2}[\text{CH}_2\text{I}_2]^2}{[\text{I}_2]} \quad (39)$$

and would have a half life of 3 hours at about 450°C. Again, the rate would be much more rapid at higher temperatures.

The literature contains data that indicate much higher amounts of methyl iodide present (up to 10%) either in the fuel or upon meltdown than would be expected from the above discussions. Some of these data are summarized in Table III-1.

The high yields of methyl iodide reported can be attributed to poor analytical techniques (discussed in Section III,A,4), impurities that are known to be present in UO_2 , and to impurities in the atmosphere of the test apparatus. In certain experiments, methane was added directly to the apparatus. In addition, some UO_2 fuels may contain more than 0.014 ppm methane (measured for high quality UO_2 fuel irradiated at SRP) which can react with radioiodine to form methyl iodide. UO_2 fuels also may contain more than 0.5 ppm hydrogen, which upon release from the melted fuel could form methane by reaction with graphite that was used in the test apparatus to support the UO_2 sample. A minor source

of organic matter is the compressed air used in the experiments. This air was dried with magnesium perchlorate and filtered through absolute filters before use, but removal of organic matter was omitted. Finally, the experiments were not conducted in an intense radiation field which is expected to decompose essentially all of the methyl iodide as discussed previously.

Experiments in the TREAT⁽²⁸⁾ facility to study the release of fission products from the fuel during a reactor accident in which the fuel melts rapidly as the result of a reactor transient add further confirmation to the insignificant amounts of CH₃I formed. "Stainless-steel and Zircaloy-clad UO₂ fuel pellets were melted under water by transient fission-heat input of approximately 510 cal per gram of UO₂. Most of the water boiled out of the fuel-containing autoclave (about 1.5 inches in diameter by about 7.5 inches long) in 1 minute to simulate a ruptured reactor pressure vessel system, followed by slow boiling to dryness in 10 minutes. Approximately 60% of the ¹³¹I was released from the melted fuel-cladding mixture. About 15% of the total iodine was carried out of the fuel autoclave with the escaping steam. About 40% of the iodine was collected later in distilled-water rinses of the walls of the disassembled fuel autoclave. The following mechanism for transport of iodine was proposed. First, the portion (~15%) that escaped from the fuel autoclave was carried only by steam which was formed at the heated fuel surface and moved out of the autoclave without being condensed. Second, the iodine which entered the liquid water either directly or by entrainment in condensing steam was essentially permanently trapped by chemical reactions to form mostly nonvolatile water soluble compounds such as metal iodides. Less than 0.01% of the total ¹³¹I was found in penetrating forms such as CH₃I."⁽²⁹⁾

3. Formation of Iodine Compounds in the Gas Phase Outside Reactor Vessel

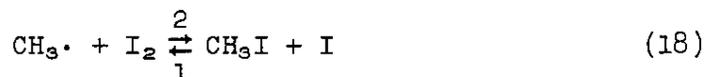
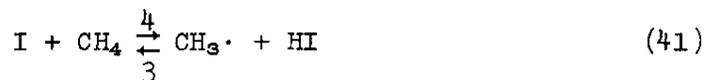
Most of the iodine escapes from the reactor tank as hydrogen iodide, because of the rapid reaction of hydrogen and iodine, Equation 25, and the large equilibrium constant as mentioned previously. As hydrogen iodide escapes, it should react rapidly with oxygen in the air to form elemental iodine.



The literature does not contain sufficient information to calculate rates or equilibrium data for this reaction. In the following discussion, it was assumed conservatively that elemental iodine exists in the gas phase outside the reactor vessel.

Although it appears in some experiments⁽³⁰⁾ that radioiodine has reacted directly in some way with airborne organic matter, the kinetics of the reaction of iodine with hydrocarbons are not favorable because the carbon-hydrogen bond strength (102 kcal/mole) is greater than the carbon-iodine bond strength (54 kcal/mole). However, at trace concentrations (<1 µg/m³) iodine may exist in a form that reacts differently from iodine at macroconcentrations.

The reaction between methane and iodide was studied quantitatively by Benson.⁽³¹⁾ The following mechanism was confirmed by extensive kinetic measurements for the gaseous system $\text{CH}_4 + \text{I}_2 \rightleftharpoons \text{CH}_3\text{I} + \text{HI}$ (Equation 21).



The rate equation for this mechanism is:

$$\frac{d[\text{HI}]}{dt} = \frac{d[\text{CH}_3\text{I}]}{dt} = k_4 K_{\text{I}_2}^{\frac{1}{2}} [\text{I}_2]^{\frac{1}{2}} [\text{CH}_4] \left\{ 1 - \frac{[\text{HI}][\text{CH}_3\text{I}]}{[\text{HI}]_{\text{eq}}[\text{CH}_3\text{I}]_{\text{eq}}} \right\} \quad (42)$$

where, in consistent units:

$\frac{d[\text{CH}_3\text{I}]}{dt}$ = the rate of formation of methyl iodide

k_4 = the specific rate constant for the forward reaction 41, shown in Figure III-4

K_{I_2} = the equilibrium constant for the dissociation of iodine in Figure III-2

$[\text{I}_2]$ = the concentration of iodine

$[\text{CH}_4]$ = the concentration of methane

$[\text{HI}]$ = the concentration of hydrogen iodide

$[\text{CH}_3\text{I}]$ = the concentration of methyl iodide

The righthand term inside the braces can be neglected when calculating the maximum rate of formation of methyl iodide. The rate constant (k_4) and the equilibrium constant (K_T) vary exponentially with temperature; the effect of temperature on these constants can be calculated by the Arrhenius equation:

$$\log K_{I_2} = 15.06 - \frac{37,600}{4.575T}, \text{ equilibrium constant for Equation 27, moles/l}$$

$$\log k_4 = 11.70 - \frac{3400}{4.575T}, \text{ rate constant for step 4, Equation 41, 1/(mole)(sec)}$$

The maximum rate of formation of methyl iodide by the above mechanism in the gas phase was calculated for the following concentrations:

I_2 : 1 and 10 ppm (vol)

CH_4 : 2 and 20 ppm (vol)

The kinetic data⁽³¹⁾ showed a slow rate of reaction between I_2 and simple hydrocarbons to form methyl iodide. Calculated rates of formation are summarized in Figure III-6. These extremely low rates of reaction limit the formation of organic compounds to insignificant concentrations. At a temperature of 100°C, 20 ppm methane, and 10 ppm iodine, the rate of formation of methyl iodide would be negligible, $<1 \times 10^{-14}$ $\mu\text{g}/(\text{m}^3)(\text{min})$. A temperature of about 400°C in the gas phase and at the same concentrations would be required to form about 1% methyl iodide per day. The equilibrium concentration of methyl iodide in the gas phase, shown in Figure III-7, would limit the maximum possible amount of methyl iodide to less than 0.005% of the iodine inventory for the above concentrations and at 100°C. The equilibrium concentration was calculated from the following equation:

$$K = \frac{[CH_3I][HI]}{[CH_4][I_2]} \quad (43)$$

4. Formation of Iodine Compounds on Surfaces Outside Reactor Vessel

The preceding discussions show that it is very unlikely that a significant quantity of methyl iodide will be released from the reactor vessel of most water cooled reactors. Other investigators have reported that the indicated formation of methyl iodide involves an adsorption-desorption activation or local reaction of

iodine with organic materials on surfaces. (30,32) For such a reaction to occur, the iodine vapor must be transferred from the bulk gas to surfaces of the containment structure. The rate of deposition, which is discussed in Section III,C, is a function of the aerodynamic conditions. Once the iodine vapor reaches the surface it may a) chemically react with the surface or with materials on the surface, or b) it may be physically adsorbed. If the first mechanism controls, a layer of reaction product accumulates on the surface; thus, the concentration of iodine on the surface may become much greater than that in the gas phase. If the chemical reaction product of the sorbed molecule has sufficient vapor pressure, the iodine compound (such as methyl iodide) may be desorbed. If the second mechanism controls, physical adsorption of iodine can also lead to concentrations on the surface that exceed that in the gas phase.

The deposition of iodine is rapid on metals such as copper and silver because metal iodides are formed by chemical reaction. Deposition on bare ferrous metals is enhanced by corrosive oxidation especially in a humid atmosphere. Iodine deposition on painted surfaces is generally much slower than on metals, because the protective coating is much less reactive than the bare metal surface.

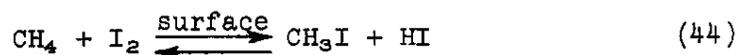
Unfortunately, there is no information in the literature on the catalytic effect of a given surface on methyl iodide formation. SRL therefore undertook a comprehensive review and analysis of the data in the literature on methyl iodide formation in reactor containment test facilities. The pertinent data from this review are summarized in Table III-2.

As shown in Table III-2, the amount of methyl iodide formed in the test facilities varied significantly. Consideration of the differences in the conditions of the tests and the kinetics of the methyl iodide reaction show that the following conditions promote the fraction of iodine that may be converted to methyl iodide:

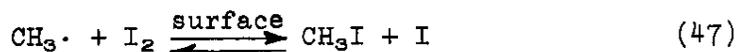
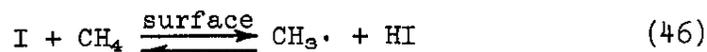
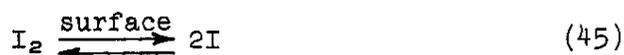
- Bare ferrous metal surfaces
- High deposition rates
- Long aging periods
- High surface to volume ratios
- Low gas-phase iodine concentration
- Moderately high temperatures

- Organic impurities
- CO/CO₂ atmospheres

In order to determine more quantitatively the effects of the above conditions on methyl iodide formation, the available data were correlated. Analysis of the data indicated that the methyl iodide formation varied as the square root of the iodine concentration on the surface, as would be predicted from the gas-phase kinetics. By analogy with the gas phase equation of Benson, discussed in Section III,A,3, a similar expression was developed for the overall reaction on a surface



for the mechanism:



and the rate equation for this mechanism is

$$\frac{d[\text{CH}_3\text{I}]_s}{dt} = k_s \left(K_{\text{I}_2} \right)_s^{1/2} [\text{I}_2]_s^{1/2} [\text{M}]_s \left\{ 1 - \frac{[\text{HI}]_s [\text{CH}_3\text{I}]_s}{[\text{HI}]_{s_{\text{eq}}} [\text{CH}_3\text{I}]_{s_{\text{eq}}}} \right\} \quad (48)$$

An approximate expression of Equation 48 when conversion to CH₃I is relatively small (less than 30%) is:

$$\frac{[\Delta\text{CH}_3\text{I}]_s}{\Delta t} = k_s \left(K_{\text{I}_2} \right)_s^{1/2} [\text{I}_2]_s^{1/2} [\text{M}]_s \left\{ 1 - \frac{[\Delta\text{CH}_3\text{I}]_s^2}{[\Delta\text{CH}_3\text{I}]_{s_{\text{eq}}}^2} \right\} \quad (49)$$

where

$$\frac{[\Delta\text{CH}_3\text{I}]_s}{\Delta t} = \text{specific reaction rate of CH}_3\text{I on a given surface, } \mu\text{g}/(\text{m}^2)(\text{min})$$

$$k_s = \text{specific reaction rate constant for a given surface at a given temperature, } \text{m}^2/(\mu\text{g})(\text{min})$$

$$\left(K_{\text{I}_2}\right)_s = \text{equilibrium constant for Equation 45 at a given temperature, } \mu\text{g}/\text{m}^2.$$

$$[\text{I}_2]_s = \text{concentration of I}_2 \text{ on surface, } \mu\text{g}/\text{m}^2.$$

$$[\text{M}]_s = \text{concentration of CH}_4 \text{ on surface, } \mu\text{g}/\text{m}^2.$$

$$[\Delta\text{CH}_3\text{I}]_{s, \text{eq}} = \text{equilibrium concentration of CH}_3\text{I on a given surface, } \mu\text{g}/(\text{m}^2).$$

Unfortunately, sufficient information is not available from the literature and from the data summarized in Table III-2 to derive accurate values for the specific reaction rate constant (k_s) of Equation 49 for various surfaces. Most data give only the final quantity of methyl iodide formed and iodine deposition on the surface after many hours of aging. In addition, information was not available on the methane concentration $[\text{M}]_s$ on the surfaces (although no significant differences between tests facilities are expected) or on the effect of a given surface on the equilibrium constant $\left(K_{\text{I}_2}\right)_s$ for Equation 45. Consequently, the three parameters k_s , $\left(K_{\text{I}_2}\right)_s$, and $[\text{M}]_s$, which are constants for any given system at a given temperature, were combined into a single constant, and Equation 49 reduces to the following equation for calculating the average rate of formation of methyl iodide:

$$\frac{[\Delta\text{CH}_3\text{I}]_s}{\Delta t} = K_s [\text{I}_2]_s^{\frac{1}{2}} \quad (50)$$

where,

$$K_s = \text{overall rate constant } \left[k_s \times \left(K_{\text{I}_2}\right)_s^{\frac{1}{2}} \times [\text{M}]_s \right], \frac{(\mu\text{g})^{\frac{1}{2}}}{(\text{m})(\text{min})}$$

The terms within the braces in Equation 49 were deleted from Equation 50 because of lack of sufficient data. This does not have a significant effect on the overall rate constant for the experimental data.

All data on methyl iodide formation summarized in Table III-2 were reduced and analyzed in terms of the parameters in Equation 50. This information is shown in Table III-3. In Table III-4, average overall rate constants (K_g) are shown for each surface, atmosphere, aging time, and temperature.

The rate constants (K_g), and therefore the rate of formation of methyl iodide, are one to two decades lower for painted surfaces than for bare stainless steel [paint equivalent to that used in the Battelle Northwest Laboratory (BNWL) Aerosol Development Facility and the United Kingdom Atomic Energy Authority (UKAEA) Zenith Reactor]. However, the combination of a surface with a graphite-base paint coating and an atmosphere of CO-CO₂-air (UKAEA data) produced the highest rate constant, and therefore the maximum rate of formation of methyl iodide, of any surface and environment studied. The rates of formation of methyl iodide in air at 20 to 25°C versus the square root of iodine concentration on painted (nonreactive type) concrete, painted (nonreactive type) steel, and bare stainless steel surfaces are shown in Figure III-8.

The rate constants (K_g) for steam-air mixtures were also about two decades lower for painted surfaces than for bare stainless steel. The rate constants are not significantly different for painted surfaces (BNWL data) in air at 20°C or steam-air at 80°C. However, the rate constant for bare stainless steel [Oak Ridge National Laboratory (ORNL) data] in a steam-air mixture at an average temperature of 50°C is about 5 times higher than in air at about 25°C. As discussed previously, information is not available in the literature for determining the equilibrium constant for dissociation of iodine on a surface. Hence, the data cannot be normalized for temperature. However, the equilibrium constant (K_{I_2})_s and the specific rate constant (k_g) would be expected to increase exponentially with temperature in accordance with the Arrhenius equation for gas phase reactions.⁽³¹⁾

The ORNL data for bare stainless steel in a steam-air mixture indicate that the average rate constant (K_g) increases by a factor of about five for an increase of about 20°C (Table III-3). These results indicate that the rate of formation of methyl iodide in steam would be significantly less than in air at the same temperature.

Although appreciable hydrolysis of methyl iodide in the gas phase would not be expected, hydrolysis of methyl iodide in the liquid film on surfaces may be a significant factor in decreasing the rate of formation of methyl iodide in steam-air mixtures. In the BNWL steam-air tests, the quantity of methyl iodide (fraction of iodine inventory) in the gas phase increased to a maximum value

in about one hour, and after five hours decreased to 40-85% of the maximum value. The BNWL tests provide further evidence that the methyl iodide is hydrolyzed in the liquid film from condensing steam on surfaces. The liquid film also probably depletes the surface of methane and iodine which would further reduce formation of methyl iodide. The rates of formation of methyl iodide in steam-air mixtures at 50 to 80°C versus the square root of iodine concentration on painted steel and bare stainless steel surfaces are shown in Figure III-9.

Data were only available from the BNWL(ADF) and ORNL(NSPP) tests throughout the entire test period. Only the BNWL data have been analyzed to date. The rate constants (K_g) for formation of methyl iodide during the first 20 minutes of these tests are shown in Table III-3, and the average rate for all the tests is shown in Table III-4. A comparison of these figures shows that the rate constants for the first 20 minutes was about one decade higher than the rates for the 300-minute period. In the air tests, formation of methyl iodide was 50 to 95% completed in one hour and almost 100% completed in five hours. As discussed above, the methyl iodide fraction decreased after one hour in steam-air mixtures.

The data and correlation demonstrate that formation of methyl iodide by active surfaces is negligible for most reactor confinement systems in which fresh air flow continuously replenishes the confined air space in structures with a nonreactive-type paint coating. Application of this analysis to the SRP confinement system, discussed in Section III,B, shows that methyl iodide formation on the building surfaces is about $10^{-6}\%$ of the iodine inventory in the fuel.

Care must be taken when applying the rate constants to reactors with containment dome structures in which the fission products are confined for relatively long periods of time. For painted (nonreactive-type) surfaces and for ambient air, the average rate constant derived from the first 20 minutes of exposure in the BNWL tests should be substituted in Equations 48 or 49 and the equation integrated as a function of time. For steam-air mixtures, the rate constant for 20 minutes should be applied over a time interval of one hour. Appropriate rate constants for use in Equations 48 or 49 for bare ferrous metal surfaces are not available as yet, and the rate constants in Table III-4 should not be applied for periods longer than indicated in the table. The rate constants give only the rate of formation of methyl iodide and not the final amount formed when equilibrium conditions are reached. Significantly greater concentrations of methyl iodide can form on surfaces than in the gas phase. Because methyl iodide rapidly desorbs from surfaces

and because the rate of decomposition of methyl iodide to satisfy the equilibrium gas phase relationship is very slow, the gas phase concentration of methyl iodide can exceed the final equilibrium value from one to several hours, depending upon the actual gas phase conditions. However, no significant formation of methyl iodide would be expected in containment domes in which the air is at relatively low temperature and is rapidly recirculated through particulate filters and carbon beds.

Of interest are the data from the UKAEA test facility (Highly Active Test Rig) in CO/CO₂ at 400°C for a period of 30 minutes (Table III-3). At such high temperatures, one would expect that the rate of methyl iodide formation would be controlled by the gas phase reaction. The agreement is good between the indicated rate of formation of methyl iodide in the experiment (about 0.2 μg/(m³)(min) and with an initial iodine concentration in the gas phase of 4 ppm (vol), 4 x 10⁴ μg/m³) with that predicted from Figure III-6 for a methane concentration of 2 ppm (vol).

It was assumed conservatively in this analysis that all of the indicated methyl iodide was formed on surfaces of the containment test facilities and subsequently released to the gas phase. Some fraction of the compounds could have been released from the cooled irradiated fuel used in some of the experiments and/or formed on surfaces in the test furnaces or in the gas phase within the furnaces during the melting operation. Rate constants were based on reported values of iodine deposited on surfaces of the test facilities. Application of Equation 50, including mechanisms that control mass transfer of iodine to surfaces, is discussed in Sections III,B and C.

Additionally, all of the data in Table III-3 on methyl iodide formation involved the use of a May Pack sampling device. Some of the ambiguities that could arise from the use of this device were discussed by Collins,^(32,33) et al. In general, analyses involving May Pack sampling devices are more likely to show higher methyl iodide formation than probably exists. Recent experiments by Collins⁽³²⁾ showed that methyl iodide formation indicated from May Pack data was about 6 times greater on the average than measured by sensitive gas chromatographic techniques (Table III-1). Consequently, the rate constants (K_g) in Tables III-3 and III-4 are probably very conservative (higher than actually exists).

TABLE III-1

UKAEA Estimates of Methyl Iodide
Released from Cooled Irradiated UO₂

Experiment	Gas Composition	UO ₂ Fuel, MWD/T	Percent of Iodine Released from Fuel	Percent of Iodine Inventory Released as Methyl Iodide	
				May Pack	Gas Chromatograph
No. 21	54% CO ₂ , 35% CO plus air, H ₂ , H ₂ O, 200 ppm CH ₄	150	2.8 - 3	1	0.2
No. 22	CO ₂ /CO	150	4 - 9	2	0.5
No. 23	CO ₂ /CO Sat. H ₂ O	150	40	4	5.0
a No. 24	CO ₂ /CO plus CH ₄ (1000 ppm)	150	33	20	1.0
No. 25	CO ₂ /CO	150	27 - 35	10	Not done
No. 26	CO ₂ /air	100	25 - 47	2	0.7
No. 27	CO ₂ /5% CH ₄	150	13 - 27	0.7	0.1
Table 3	Steam-air	0.5	6.2	0.02	-
Table 3	Steam-air	0.5	6.8	0.02	-
Table 3	Steam-air	1.0	2.4	0.01	-
Table 3	Steam-air	2.0	2.4	0.02	-
Table 3	Steam-air	5.0	2.9	0.001	-
Table 3	Steam-air	1.0	13.7	0.1	-
Table 3	Steam-air	2.0	15.4	0.003	-
Table 3	Steam-air	7.0	40.5	0.03	-
b Table 4	Steam	100	28	0.04	-
Table 4	Steam	100	35	0.5	-
Table 4	Steam	100	34	3	-
Table 4	Steam	100	9	2	-
Table 4	Steam	100	27	0.08	-
Table 4	Steam-air	100	60	0.2	-

a - Reference 32.

b - Reference 34.

TABLE III-2

Summary of Literature Data on Methyl Iodide Formation

Test (a) Facility	Surface	I ₂ (b) Deposition, %	Aging Time, hr.	Surface to Volume Ratio, m ² /m ³	I ₂ (c) Conc., μg/m ³	Temp., °C	Atmosphere	CH ₃ I, (b) %
ADF	Painted Steel	20-95	5	5.2	0.03-630	20	Air	0.05-0.7
ADF		50	5		0.2-640	80	Steam-Air	0.1-6.0
CMF	Bare Stainless Steel	25-90	3-4	9.1	5-2200	25	Air	0.3-3.7
CMF		8-60	4-18		2000- 8000	30- 110	Steam-Air	0.5-1.3
CMF		20	5-16		2000	30- 110	Steam-Air- 0.3% CH ₄	2.5-21
NSPP			36-48	1.4	2600-25000	30	Air	6-10
Zenith	Painted Concrete & Steel	90	5	1.4	0.4-0.8	25	Air	0.09-0.6
Zenith		1	Ventilated		0.8	25	Air	<0.003 ^(d)
HATR	Painted Steel	10	0.5	11	40,000	400	CO-CO ₂	0.25
HATR		95	20	7.8	1300	25	CO-CO ₂ -Air	5

NOTES:

- (a) ADF - Aerosol Development Facility, Battelle - Northwest⁽²⁸⁾
 CMF - Containment Mockup Facility, Oak Ridge National Lab. (21, 26, 27)
 NSPP - Nuclear Safety Pilot Plant, Oak Ridge National Lab. (21)
 Zenith - Reactor Facility, United Kingdom Atomic Energy Authority⁽¹⁶⁾
 HATR - Highly Active Test Facility, United Kingdom Atomic Energy Authority^(2a)
- (b) Values shown are maximum reported for test in terms of starting iodine inventory and are not necessarily the final equilibrium value.
- (c) Maximum iodine concentration in gas phase at start of test.
- (d) Limit of experimental measurement.

TABLE III-3

Average Rate Constants for Methyl Iodide Formation on Surfaces of Containment Test Facilities

Test Facility	Test No.	T, °C	Δt, min.	$\frac{[\Delta\text{CH}_3\text{I}]_s}{\Delta t}$ ($\frac{\mu\text{g}}{\text{m}^2 \cdot \text{min}}$)	$[\text{I}_2]_s$ ($\frac{\mu\text{g}}{\text{m}^2}$)	k_s^* ($\frac{\mu\text{g}}{\text{m}^2 \cdot \text{min}}$)
AIR TESTS						
Battelle-Northwest (ss)	16	20	2x10 ¹	2.0x10 ⁻⁶	2.0x10 ⁻²	1.0x10 ⁻⁴
	18	20	2x10 ¹	2.3x10 ⁻³	6.2x10 ⁰	3.7x10 ⁻⁴
Aerosol Development Facility (ADF)	20	20	2x10 ¹	1.1x10 ⁻³	8.8x10 ⁰	1.3x10 ⁻⁴
Painted (Phenoline 300)						
Carbon Steel,	16	20	3x10 ²	1.3x10 ⁻⁷	3.3x10 ⁻²	3.9x10 ⁻⁶
Volume/Surface Area = 1.9x10 ⁻¹ m	18	20	3x10 ²	1.6x10 ⁻⁴	1.1x10 ¹	1.5x10 ⁻³
	20	20	3x10 ²	1.5x10 ⁻⁴	1.1x10 ¹	1.4x10 ⁻³
Iodine Release:						
Tests 16 & 18 - I ₂						
Tests 20, 21, 23, & 28 - I ₂ +UO ₂						
Tests 26 & 33 - Irradiated UO ₂						
STEAM - AIR TESTS						
	21	80	2x10 ¹	5.3x10 ⁻³	7.8x10 ⁰	6.8x10 ⁻⁴
	23	80	2x10 ¹	6.9x10 ⁻⁴	7.0x10 ⁰	9.9x10 ⁻⁵
	28	80	2x10 ¹	1.0x10 ⁻⁴	1.0x10 ⁻¹	1.0x10 ⁻³
	26	80	2x10 ¹	1.3x10 ⁻⁶	3.6x10 ⁻²	3.6x10 ⁻⁵
	33	80	2x10 ¹	3.9x10 ⁻⁶	6.0x10 ⁻²	6.5x10 ⁻⁵
	21	80	3x10 ²	2.6x10 ⁻⁴	7.8x10 ⁰	3.3x10 ⁻³
	23	80	3x10 ²	1.0x10 ⁻⁴	7.0x10 ⁰	1.4x10 ⁻³
	28	80	3x10 ²	3.2x10 ⁻⁶	1.4x10 ⁻¹	2.3x10 ⁻³
	26	80	3x10 ²	1.1x10 ⁻⁷	5.6x10 ⁻²	2.0x10 ⁻⁶
	33	80	3x10 ²	1.6x10 ⁻⁷	7.5x10 ⁻²	2.1x10 ⁻⁶
AIR TESTS						
Oak Ridge National Laboratory (ss, ss, sr)						
Containment Mockup Facility (CMF),	A	25	1.8x10 ²	1.3x10 ⁻⁴	5.3x10 ⁻¹	2.5x10 ⁻⁴
Bare Stainless Steel,	B	25	1.8x10 ²	1.4x10 ⁻⁵	3.9x10 ⁻¹	3.6x10 ⁻⁵
Volume/Surface Area = 1.1x10 ⁻¹ m	C	25	1.8x10 ²	4.0x10 ⁻³	7.8x10 ⁰	5.1x10 ⁻⁴
	D	25	2.4x10 ²	2.5x10 ⁻³	6.5x10 ⁰	3.9x10 ⁻⁴
	E	25	2.4x10 ²	2.6x10 ⁻³	1.4x10 ¹	1.9x10 ⁻⁴
Iodine Release:						
Test E - I ₂						
Tests A, B, D, 6-25, 8-4, 12-9,						
2-24 & 3-25 - I ₂ +UO ₂						
Test C - Irradiated UO ₂ +I ₂	6-25	40	2.4x10 ²	4.6x10 ⁻³	1.1x10 ¹	4.2x10 ⁻⁴
	8-4	40	1.1x10 ²	2.4x10 ⁻³	9.1x10 ⁰	3.8x10 ⁻⁴
	12-9	60	3.3x10 ²	3.3x10 ⁻²	1.7x10 ¹	1.9x10 ⁻³
	2-24	60	3.0x10 ²	2.1x10 ⁻²	1.1x10 ¹	1.9x10 ⁻³
	3-25	60	3.0x10 ²	2.4x10 ⁻²	8.4x10 ⁰	2.9x10 ⁻³
AIR TESTS						
Nuclear Safety Pilot Plant (NSPP) (ss)						
Bare Stainless Steel,	5	30	2.9x10 ³	2.4x10 ⁻²	3.7x10 ¹	6.5x10 ⁻⁴
Volume/Surface Area = 7.0x10 ⁻¹ m	6	30	2.8x10 ³	1.3x10 ⁻³	3.2x10 ¹	4.1x10 ⁻⁵
	7	30	2.2x10 ³	4.1x10 ⁻²	1.2x10 ²	3.4x10 ⁻⁴
Iodine Release:						
Tests 5 & 6 - I ₂						
Test 7 - I ₂ +UO ₂						
AIR TESTS						
United Kingdom A.E.A. (ss)						
Zenith Reactor (ZR),	1	25	3.0x10 ²	7.2x10 ⁻⁷	5.0x10 ⁻¹	1.4x10 ⁻⁶
Painted (chlorinated rubber base)	2		3.0x10 ²	7.5x10 ⁻⁷	7.1x10 ⁻¹	1.1x10 ⁻⁶
Concrete and Carbon Steel,						
Volume/Surface Area = 7.2x10 ⁻¹ m						
Iodine Release - I ₂ (all tests)	3	25	1.6x10 ²	≤5.4x10 ⁻⁸	1.0x10 ⁻¹	≤5.4x10 ⁻⁷
Highly Active Test Rig (HATR) (ss)						
Painted (graphite base) Carbon Steel						
Volume/Surface Area:						
Furnace Vessel = 9.3x10 ⁻² m (Test 1a)	1a*	400	3.0x10 ¹	CO - CO ₂ TEST 2.0x10 ⁻¹	-	-
System Loop = 1.3x10 ⁻¹ m (Test 1b)						
Iodine Release - Irradiated UO ₂ +I ₂	1b	25	1.2x10 ³	CO - CO ₂ - AIR TEST 6.8x10 ⁻²	1.3x10 ¹	5.2x10 ⁻³

NOTES:

$$* k_s = \frac{[\Delta\text{CH}_3\text{I}]_s}{\Delta t} / [\text{I}_2]_s$$

where: k_s = average overall rate constant $\{ k_s \times [\text{I}_2]_s^{\frac{1}{2}} \times [\text{M}]_s \}$, (μg)^{1/2} / (m)(min), for indicated Δt.

$\frac{[\Delta\text{CH}_3\text{I}]_s}{\Delta t}$ = specific reaction rate of CH₃I on a given surface, $\mu\text{g}/(\text{m}^2)(\text{min})$.

$[\text{I}_2]_s$ = concentration of I₂ on surface, $\mu\text{g}/\text{m}^2$.

** Reaction controlled by gas phase. I₂ concentration in gas phase was 4x10⁴ $\mu\text{g}/\text{m}^3$ - 4 ppm (vol.); see Figure III-6.

TABLE III-4

Summary of Average Rate Constants
for Methyl Iodide Formation on Surfaces

Facility ^(a)	Surface	Atmosphere	Aging Time, hr	Temp., °C	Average Overall Rate Constant, K_S ^(f)
ADF (BNWL)	Painted Steel ^(c)	Air	<0.5	20	2.0×10^{-4}
ADF (BNWL)	Painted Steel ^(c)	Air	5	20	1.3×10^{-5}
CMF & NSPP (ORNL)	Bare Stainless Steel	Air	5-48	25	3.0×10^{-4}
Zenith(UKAEA)	Painted Concrete & Steel ^(b)	Air	5	25	1.0×10^{-6}
(Estimated) ^(d)	Painted Concrete & Steel ^(b)	Steam-Air	5	80	$<2 \times 10^{-6}$
ADF (BNWL)	Painted Steel ^(c)	Steam-Air	<0.5	80	3.8×10^{-4}
ADF (BNWL)	Painted Steel ^(c)	Steam-Air	5	80	1.5×10^{-5}
CMF (ORNL)	Bare Stainless Steel	Steam-Air	4-18	50	1.5×10^{-3}
HATR (UKAEA)	Painted Steel ^(e)	CO-CO ₂ -Air	20	25	5.2×10^{-3}

(a) Description of test facilities is given in Table III-3.

(b) Chlorinated rubber base paint.^(1a)

(c) Vinyl base paint (Phenoline 300 - Carboline Co.)

(d) Rate constant estimated from Zenith air tests and steam air tests in ADF facility.

(e) Graphite-loaded primer (Strathclyde PA-21)

$$(f) K_S = \left[k, \frac{m^2}{\mu g \cdot min} \right] \left[K_{I_2}, \frac{\mu g}{m^2} \right]^{1/2} \left[M, \frac{\mu g}{m^2} \right]_S$$

B. SRP CONDITIONS WITH RESPECT TO IODINE COMPOUNDS

Discussed in Section III,A are factors which contribute to the formation of iodine compounds. The factors are treated both qualitatively and quantitatively for general application to many reactor confinement applications. In this section, the same information is applied specifically to SRP conditions. Calculations are based on methyl iodide rather than iodine compounds in general, because methyl iodide was reported as being the chief constituent (about 90%) of the compounds associated with radioiodine releases and because other iodine compounds are adsorbed readily by activated carbon even under moist conditions.

Shown in Table III-5 is a qualitative comparison of conditions that promote the fraction of iodine converted to methyl iodide and SRP conditions.

TABLE III-5

Qualitative Comparison of Conditions Affecting
Fraction of Iodine Converted to Methyl Iodide

<u>Conditions that Promote Fraction of Iodine Converted to Iodine Compounds</u>	<u>SRP Conditions</u>
<u>In the fuel</u> <ul style="list-style-type: none">● Organic impurities● High temperatures● High hydrogen content	<u>In the fuel</u> <ul style="list-style-type: none">● High purity fuel● Low temperatures● Low hydrogen content
<u>Upon meltdown</u> <ul style="list-style-type: none">● Low radiation field● High temperatures● CO/CO₂ atmosphere	<u>Upon meltdown</u> <ul style="list-style-type: none">● High radiation field● High temperatures● Steam and/or oxidizing atmosphere
<u>In the reactor room (gas phase)</u> <ul style="list-style-type: none">● Long residence time● High temperatures● Organic impurities● Low gas phase iodine concentration	<u>In the reactor room (gas phase)</u> <ul style="list-style-type: none">● Short residence time● Low temperatures● Relatively clean air● High gas phase iodine concentration
<u>On reactor room surfaces</u> <ul style="list-style-type: none">● Bare ferrous metal surfaces● High surface to volume ratio● Dry surfaces	<u>On reactor room surfaces</u> <ul style="list-style-type: none">● Primarily painted concrete surfaces● Low surface to volume ratio● Condensing steam on surfaces and/or dry

The information in Table III-5 is now treated quantitatively in light of the discussion in Section III,A. Each of the factors affecting the overall kinetic relationship is combined into a single equation; then the individual factors are evaluated and combined to yield the effective reaction rate for converting elemental iodine to methyl iodide. Based on residence time, the conversion rate is expressed as the fraction of initial reactor iodine inventory escaping the activity confinement system as methyl iodide.

Equation (51) is a generalized reaction rate expression for the factors relevant to the SRP system.

$$\begin{aligned}
 \left[\frac{\Delta \text{CH}_3\text{I}}{\Delta t} \right]_{\text{overall}} &= \left[\frac{\Delta \text{CH}_3\text{I}}{\Delta t} \right]_{\text{absorbed radiation}} \left[\left. \begin{array}{c} \text{Prior to} \\ \text{Incident} \\ \left\{ \left[\frac{\Delta \text{CH}_3\text{I}}{\Delta t} \right]_{\text{fuel}} \right\} \end{array} \right\} \right. \\
 &+ \left\{ \begin{array}{c} \text{Molten Fuel in Environment in Reactor Tank} \\ \left[\frac{\Delta \text{CH}_3\text{I}}{\Delta t} \right]_{\text{tank}} + \left[\frac{\Delta \text{CH}_3\text{I}}{\Delta t} \right]_{\text{hydrolysis}} + \left[\frac{\Delta \text{CH}_3\text{I}}{\Delta t} \right]_{\text{chemical reaction with HI}} + \left[\frac{\Delta \text{CH}_3\text{I}}{\Delta t} \right]_{\text{pyrolysis}} \end{array} \right\} \\
 &+ \left\{ \begin{array}{c} \text{Outside Reactor Tank} \\ \left[\frac{\Delta \text{CH}_3\text{I}}{\Delta t} \right]_{\text{gas phase}} + \left[\frac{\Delta \text{CH}_3\text{I}}{\Delta t} \right]_{\text{surface}} \end{array} \right\} \left[\frac{\Delta \text{CH}_3\text{I}}{\Delta t} \right]_{\text{absorbed radiation}} \quad (51)
 \end{aligned}$$

1. Formation of Methyl Iodide in Fuel

High purity fuel used at SRP and low metal temperatures do not favor conversion of elemental iodine to methyl iodide. The average impurity level for hydrogen and methane in uranium-aluminum alloy is 1.2 ppm H₂ and 0.035 ppm CH₄. These values are inferred from measured values on uranium oxide fuel. From Figure III-1 and the above methane concentrations, the following amounts of methyl iodide in SRP fuel were determined:

Type Fuel	Avg Bulk Temp, °C	CH ₄ , ^(a) ppm	I ₂ , ^(a) ppm	CH ₃ I	
				Mass, µg	% of Total Iodine Inventory
U-Al Alloy ^(b)	130	0.035	2400	8400	0.00067

(a) ppm, based on weight of core material.

(b) For conditions applicable to the Savannah River high flux demonstration, as discussed in Reference 1.

2. Formation of Methyl Iodide Upon Meltdown

The following factors relating to formation and decomposition were considered in calculating the net methyl iodide that could be released from the reactor tank following a full-core meltdown:

- methyl iodide release from the fuel
- gas-phase formation in tank
- surface reaction
- chemical reaction with hydrogen iodide
- hydrolysis
- pyrolysis
- radiolysis

Those factors which contribute to methyl iodide formation include the material released from the fuel, gas-phase reaction with impurities in the tank, and reaction catalyzed by metal surfaces. Should an accident occur involving a full-core meltdown, most of the liquid water would be rapidly drained or ejected from the tank. The remaining coolant would be vaporized, and molten fuel would collect in the bottom of the tank.

As discussed in Section III,A,4, analysis of UKAEA tests^(sa) at 400°C for 30 minutes showed that surface reaction is inconsequential compared to the gas-phase reaction; therefore, the mechanism for reaction is recognized, but not included because of the relative magnitude of the results compared to the gas-phase reaction in the tank. The gas-phase equilibrium concentration was determined from Equation 43 to be 400 µg/m³ (0.004% of total iodine). The following conditions were used for the determination:

Iodine concentration: 1.8 x 10⁷ µg/m³ (100% of iodine inventory released into reactor vessel).

Methane concentration: 50 µg/m³ (0.001% of carbon in fuel and other components conservatively assumed to react with hydrogen at surface of molten fuel mass to form methane).

Temperature: 700°C

Because the equilibrium concentration of methyl iodide in the reactor vessel (400 µg/m³) is more than that which would be caused by release from the fuel (120 µg/m³), Equation 42 for rate of formation of methyl iodide would apply. Because of the very rapid reaction rate at 700°C, the equilibrium value would be obtained in less than one second.

The remaining factors — hydrolysis, pyrolysis, and radiolysis favor decomposition of methyl iodide. Radiolysis is considered quantitatively, but because of insufficient data, no credit is taken for the effects of hydrolysis and pyrolysis. Pyrolysis, however, is being studied further.

For conditions applicable to the Savannah River high flux demonstration, (i) radiolysis is expected to decompose 99% of methyl iodide per 1.2 seconds, as discussed in Section III,A,2. The quantitative effects of radiolysis are considered independently of chemical reaction and are summarized in Table III-6.

TABLE III-6

Decomposition of Methyl Iodide in
the Reactor Vessel by Radiolysis

<u>Residence Time, sec</u>	<u>Maximum Initial CH₃I Concentration in Reactor Vessel, μg/m³</u>	<u>Remaining CH₃I Concentration in Reactor Vessel, μg/m³</u>	<u>Net CH₃I, μg</u>
1	400	9.4x10 ⁰	6.6x10 ²
2	400	2.2x10 ⁻¹	1.5x10 ¹
3	400	5.4x10 ⁻³	3.8x10 ⁻¹
4	400	1.3x10 ⁻⁴	9.1x10 ⁻³
5	400	3.2x10 ⁻⁶	2.2x10 ⁻⁴

The net amount of methyl iodide that could escape the tank after the effects of radiolysis is less than 10⁻³ μg (10⁻¹²% of reactor iodine inventory) for a residence time of about 5 seconds. Because there is little driving force to rapidly remove vapor from the tank prior to the addition of light water, most of the iodine would remain in the tank until water was added.

Radiolysis alone is adequate to reduce the concentration of methyl iodide to an inconsequential value in the reactor vessel. Because of the effectiveness of the decomposition mechanisms, the initial concentration of methyl iodide in the tank would have no bearing on the off-site ingestion dose.

3. Formation of Methyl Iodide in Reactor Room

Formation of methyl iodide in the reactor room was calculated for two cases: (1) formation in the gas phase, and (2) formation on surfaces.

The gas phase reaction is controlled by the temperature in the reactor room. Because the reactor room pressure is essentially atmospheric, the highest temperature that might be expected would be 100°C or less in the immediate vicinity of the reactor tank during the initial phase of a major accident. After light water addition has begun and steam generation has stopped, the room temperature would drop to about 25°C. During the first 30 minutes of an accident, an average temperature of about 40°C was calculated. As shown in Figure III-6, at a temperature of about 40°C, the rate of formation of methyl iodide in the gas phase would be about 10⁻²⁰ μg/(m³)(min), which is inconsequential.

The controlling mechanism for formation in the reactor room would be reaction on surfaces. The reaction rate for surfaces is expressed by Equation 49. In the SRP ventilated system, however, the iodine concentration is continuously diminishing, as expressed by Equation 52 in Section III,C,1. Therefore a precise determination of the reaction rate would be calculated by integrating a combined form of Equations 49 and 52. In the interest of time, the reaction rate is estimated conservatively (Equation 50) from the maximum iodine concentration as determined by the procedure described in Section III,A,4. This maximum concentration corresponds to a reaction rate of $5 \times 10^{-5} \mu\text{g}/(\text{m}^2)(\text{min})$. If it is assumed that methyl iodide desorbs upon formation, the total amount reaching the adsorption beds is negligible, about $10^{-6}\%$ of total reactor iodine inventory in the first hour. Inconsequential formation of methyl iodide on the surfaces would occur after one hour because of desorption of elemental iodine as demonstrated by the Zenith reactor tests.

4. Decomposition of Methyl Iodide Release to the Atmosphere

Although a radiation field in the order of 10^6 R/hour could exist on the beds, no credit was taken in the analysis for decomposition of methyl iodide on the beds, because the retention time of methyl iodide on SRP carbon is unknown. Tests are planned to determine this value.

5. Summary of Methyl Iodide Release to the Atmosphere

The net amount of methyl iodide that could be formed in the SRP system if an accident should occur is inconsequential; it would contribute less than 1 Rem to the off-site ingestion dose. Although the iodine compound formation was calculated to be about $10^{-6}\%$ of total reactor iodine inventory, a value of 10^{-4} is used. This conservatism allows for possible differences between methane concentration on surfaces at SRP, variation in results from tests from which K_s was derived, precision of factors used in deposition calculations, and conditions on the surfaces, i.e., dry or condensing steam.

C. TRANSPORT AND DEPOSITION OF FISSION PRODUCTS IN THE SRP SYSTEM

Transport and deposition of fission products within the reactor building must be determined to permit calculation of the disposition of the fission products, methyl iodide formation, and heat loads on the filters and adsorbers. Transport of fission products was considered from two aspects: removal of material from the reactor room through the exhaust ventilation system and removal in water solution or suspension to the 50-million-gallon emergency basin.

1. Transport and Deposition through Exhaust Ventilation System

In a once-through, high-volume ventilation system such as the SRP system, removal of fission products from the proximity of the reactor to the activity confinement system was designed to be very rapid. Most of the material is carried through the exhaust ducts to the filter compartments; only a small part is deposited on the surfaces of the reactor room and ventilation ducts. The reduction in iodine vapor activity in the reactor room and the amount of material deposited on surfaces was calculated from an exponential equation and deposition constants developed primarily by the British. (1a, 3a)

$$C = C_0 \exp - \left[\left(\frac{vA}{V} + \frac{F}{V} \right) t \right] \quad (52)$$

where:

C = quantity of fission products in reactor room at time t

C₀ = initial quantity of fission products in reactor room at time zero

v = deposition velocity

A = surface area

V = room volume

F = exhaust air flow

t = time after incident

The amount of material deposited on surfaces of the reactor room and ventilation ducts was calculated with use of the IBM 704 computer as the difference between Equation 52 when the deposition velocity was equal to zero, and when the deposition velocity was equal to the experimentally determined value. Instantaneous release of fission products into the reactor room was assumed. An air flow of 38,000 cfm through the reactor room and an effective deposition velocity of 4.7×10^{-3} cm/sec were used in the calculation. (3a)

For SRP application, maximum iodine deposition of 1% of the airborne inventory was calculated to occur on surfaces of the reactor room and exhaust ducts. Compared to unventilated facilities 1% deposition is low, but the SRP surface to volume ratio and the residence time for deposition are very low. Also,

"Amercoat" 33HB, which is the paint used on concrete at SRP, had the lowest deposition of several paints tested at ORNL⁽²¹⁾. TID-14844⁽⁴⁰⁾ suggests a deposition of 50%, but for application to reactors with containment shells this value may be low, and higher deposition factors may be applicable. Although the initial mass of iodine calculated to reach the carbon beds was almost doubled when 1% deposition was used rather than the suggested 50%, the potential for iodine compound formation is greatly reduced, as discussed in Section III,B. The results of the transport and deposition calculations are shown in Figures III-10 and III-11. About 99% of the material would be removed from the process room in 40 minutes at normal exhaust ventilation flow.

For practical purposes, iodine is the only fission product of concern in the deposition determination. Noble gases deposit on reactor room surfaces in negligible quantities and then are retained only temporarily. The amount of noble gas that dissolves in the full reactor inventory of D₂O plus emergency light water that might be added to the system is negligible. Likewise the SRP activated carbon beds detain noble gases for only a few seconds. Therefore, no deposition is assumed, and 100% of the noble gases leaving the reactor escape to the atmosphere.

Only one percent of the nonvolatile fission product activity was assumed to escape the reactor⁽⁴¹⁾. Half of this amount was estimated to settle out in the reactor room or deposit on surfaces. Because of the small fraction released and the 99% efficiency of the particulate filters, the off-site radiation and ingestion doses are negligible.

2. Transport to Emergency Storage Basin

As discussed in Section II-F, any waterborne fission products would be transported to the 50-million-gallon storage basin. With regard to rapid off-site contamination, the only group of fission products of concern would be the halogens, some of which could enter the basin and escape into the atmosphere. All noble gases would be released to the ventilation system because of their high volatility and low solubility. Nonvolatile particulate matter would either remain in the water, settle to the bottom of the basin, or soak into the soil. If the reactor iodine inventory were transported to the storage basin, less than half the 300 rem stated in the 10 CFR 100 guidelines could be released to the atmosphere.

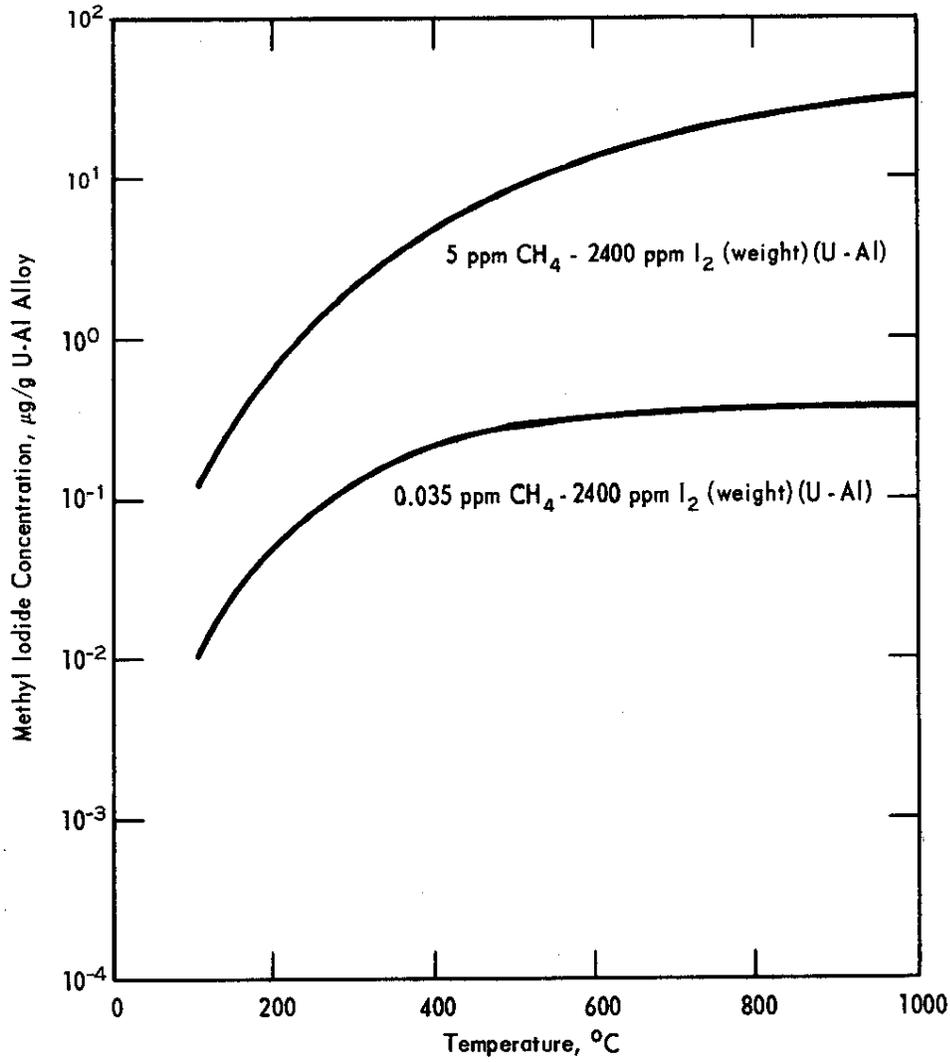


FIG. III-1 MAXIMUM POSSIBLE EQUILIBRIUM AMOUNT OF METHYL IODIDE IN IRRADIATED URANIUM-ALUMINUM ALLOY

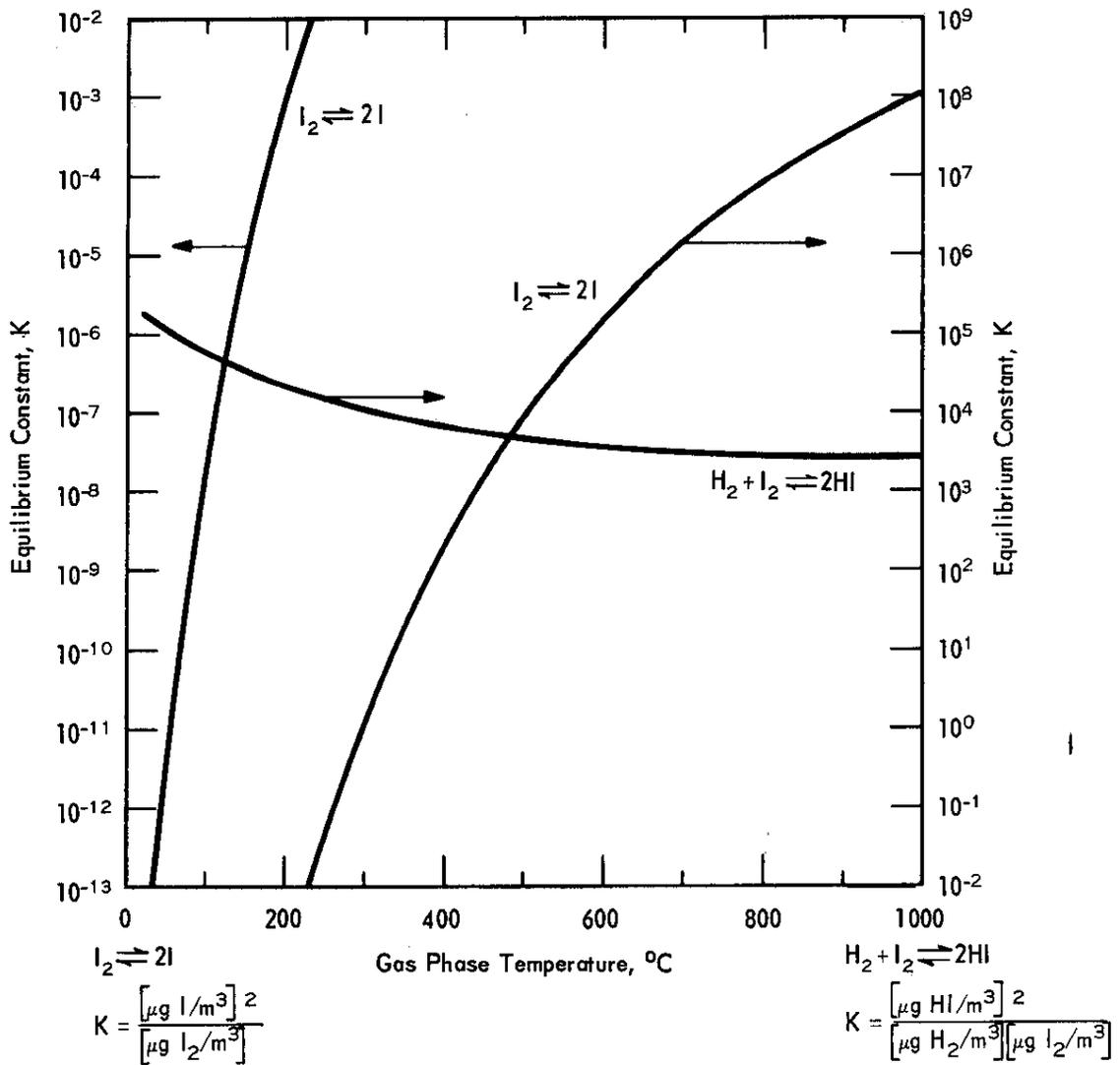


FIG. III-2 EQUILIBRIUM CONSTANTS FOR IODINE AND HYDROGEN IODIDE

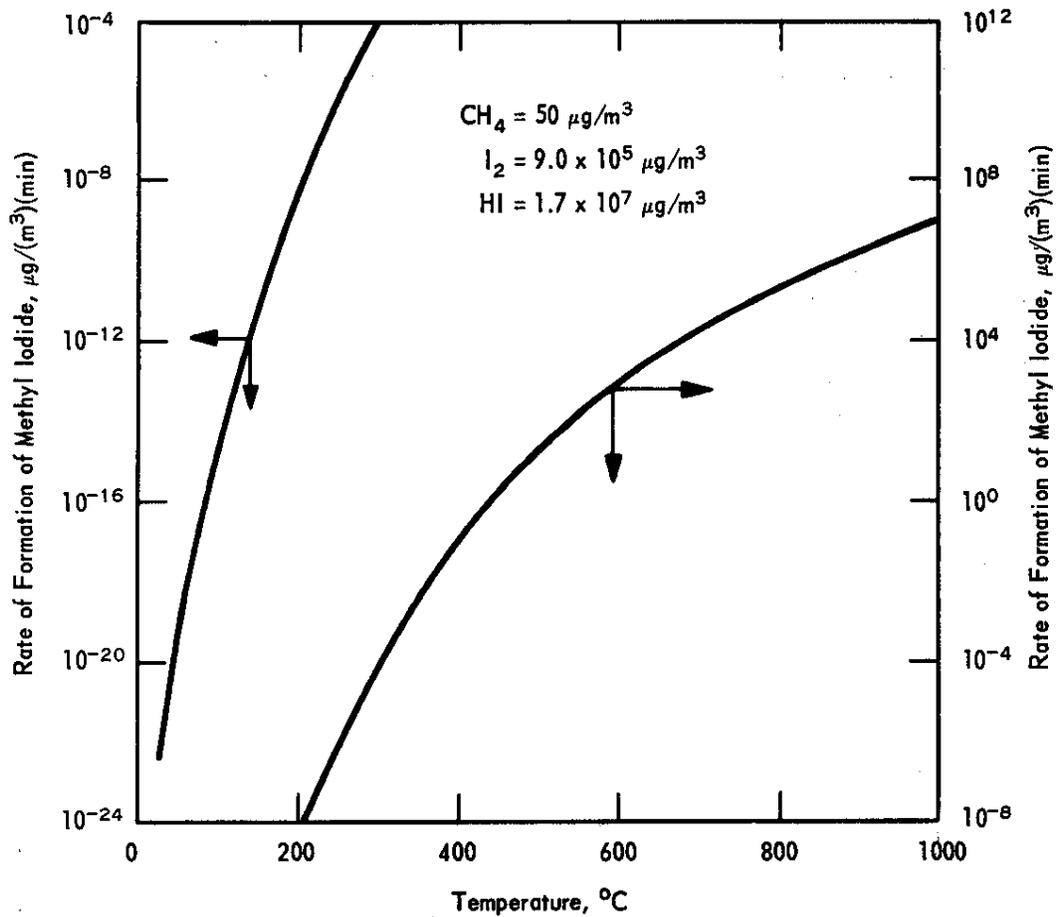


FIG. III-3 RATE OF FORMATION OF METHYL IODIDE IN HYDROGEN IODIDE - RICH ATMOSPHERE

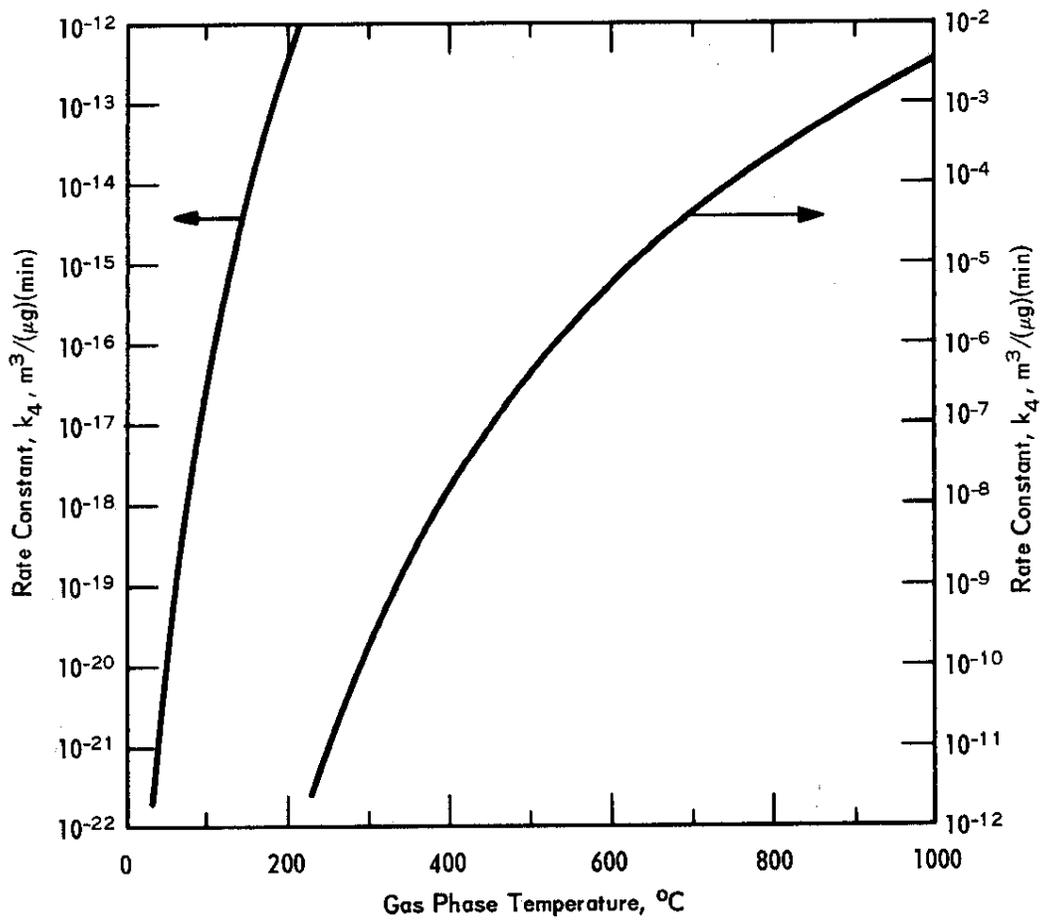


FIG. III-4 SPECIFIC RATE CONSTANT FOR METHANE - IODINE REACTION

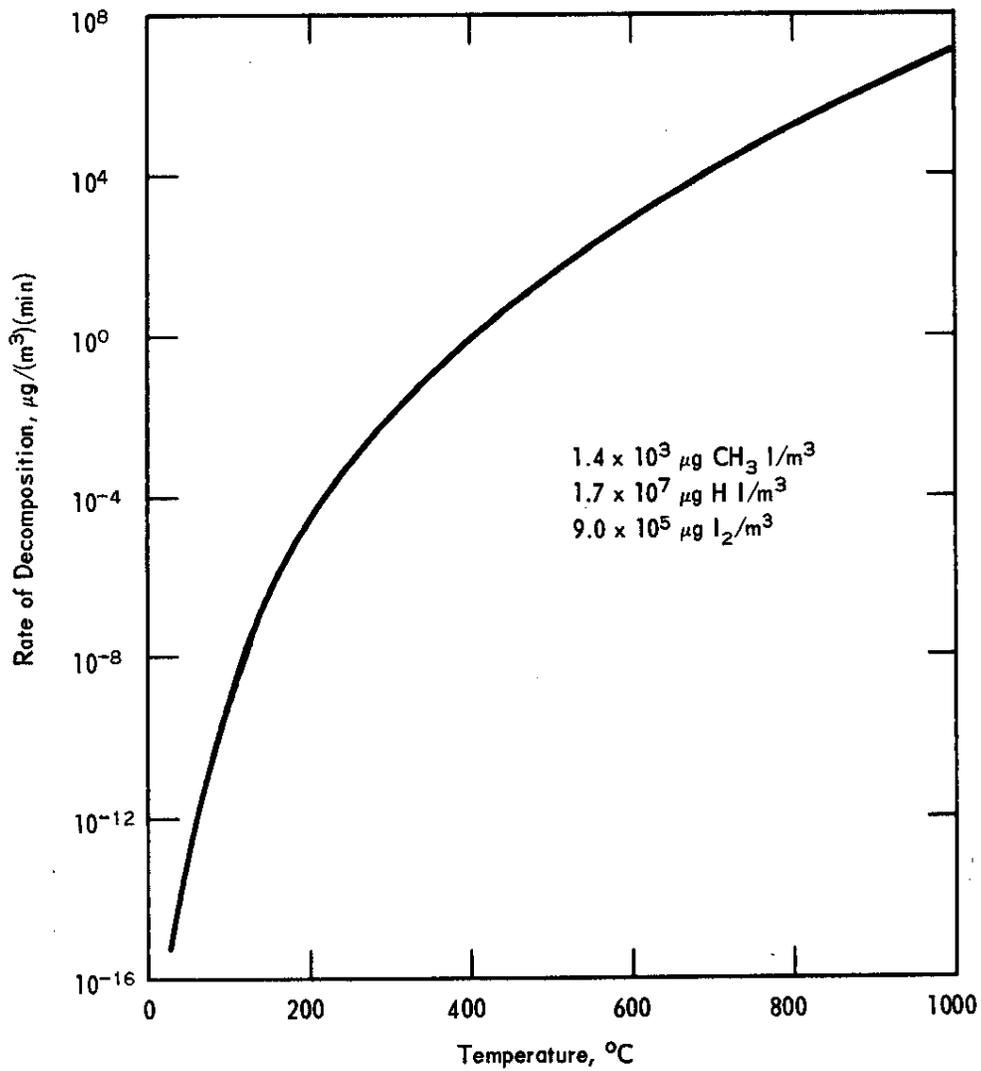


FIG. III-5 RATE OF DECOMPOSITION OF METHYL IODIDE BY REACTION WITH HYDROGEN IODIDE

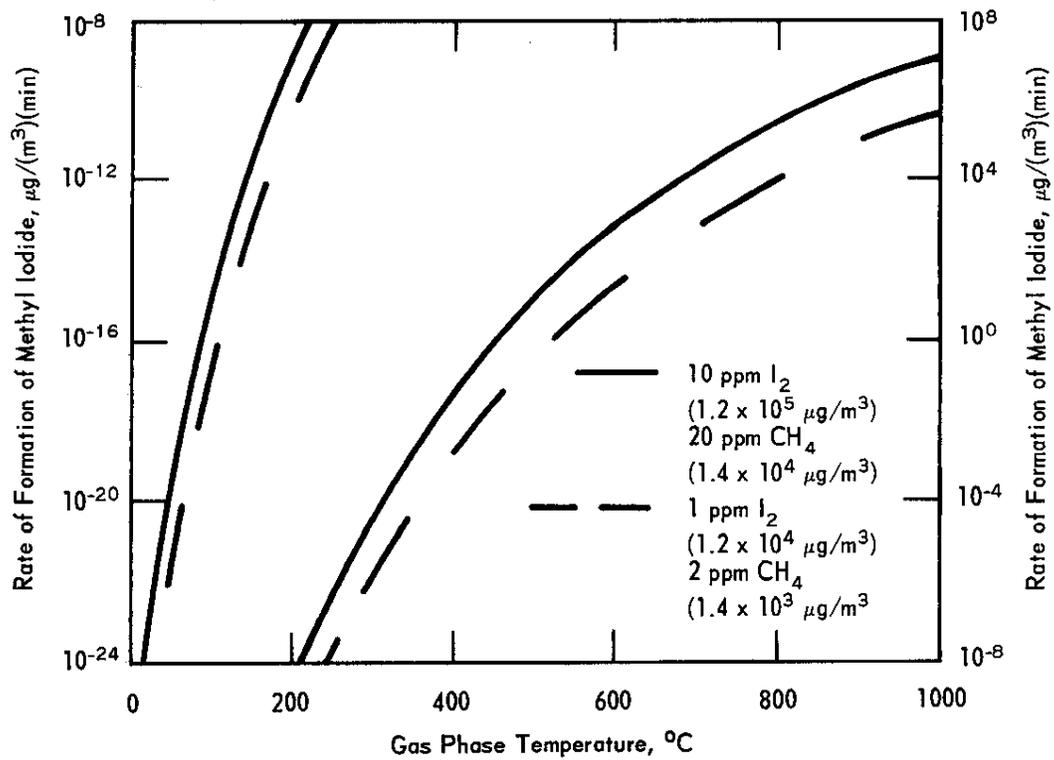


FIG. III-6 RATE OF FORMATION IN GAS PHASE OF METHYL IODIDE IN CONTAINMENT BUILDINGS

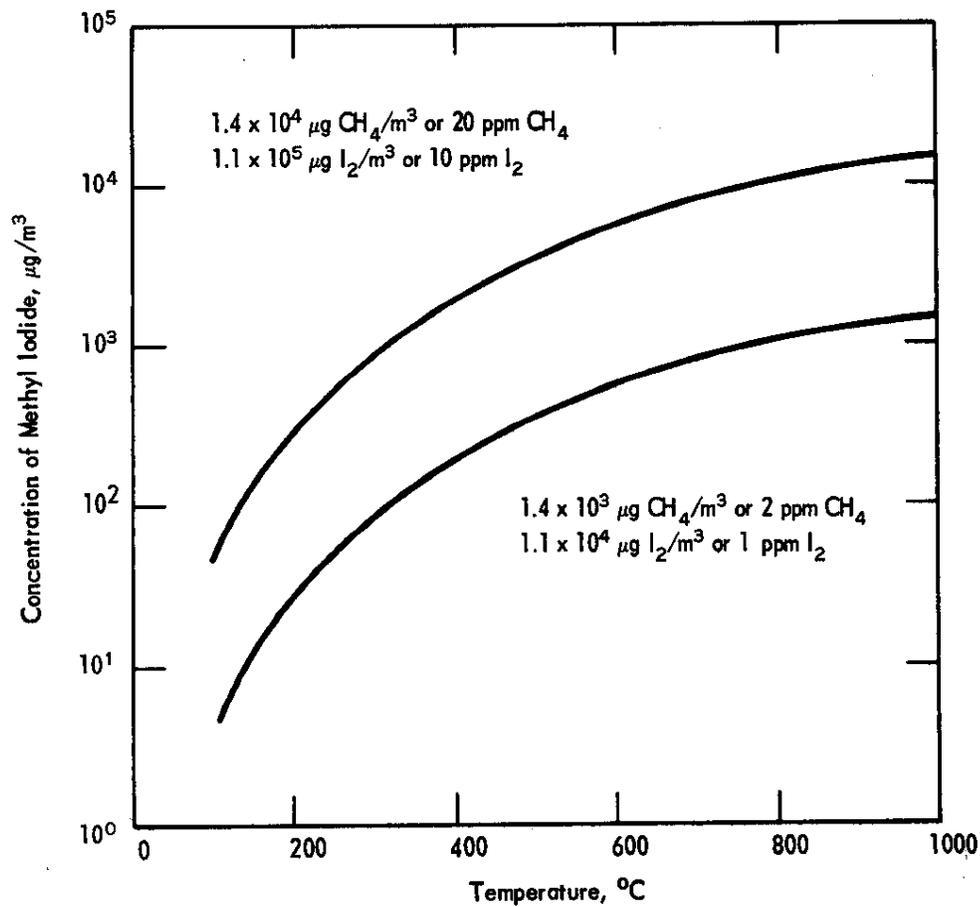


FIG. III-7 EQUILIBRIUM AMOUNT IN GAS PHASE OF METHYL IODIDE IN CONTAINMENT BUILDINGS

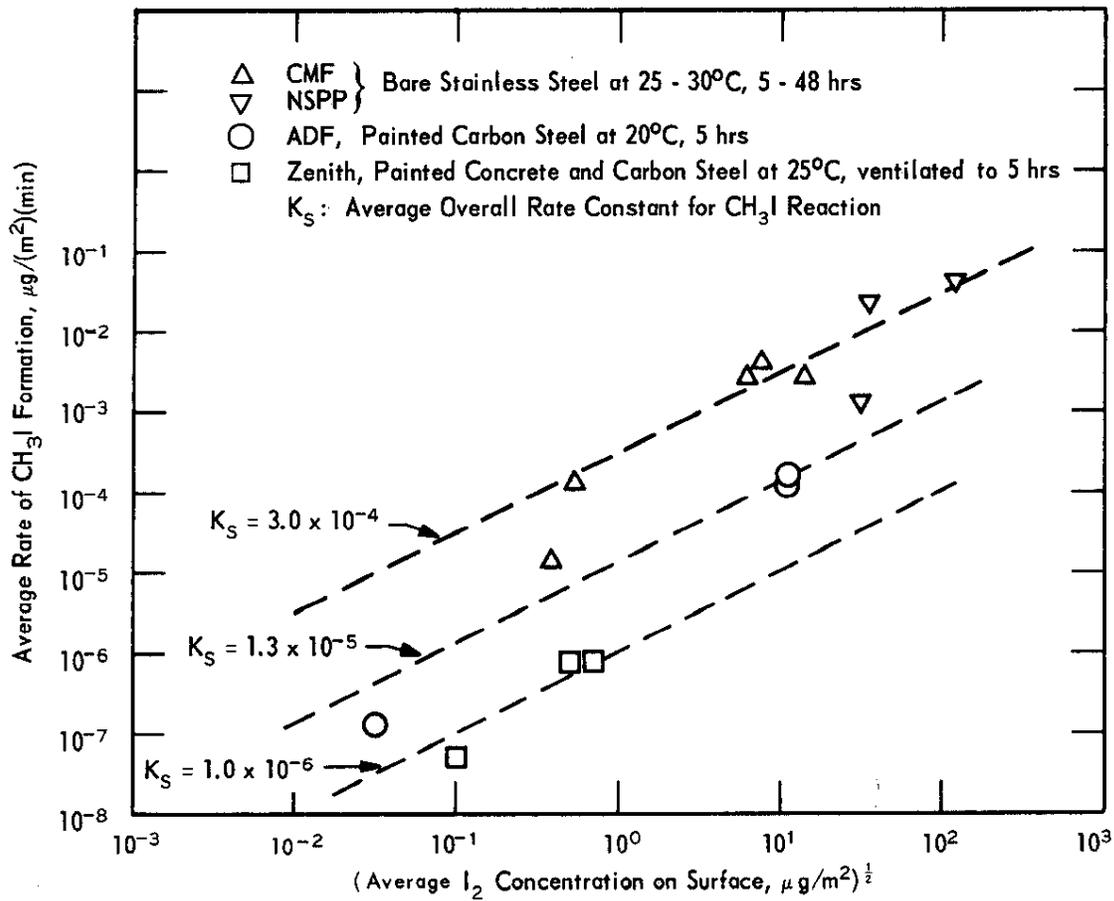


FIG. III-8 METHYL IODIDE FORMATION ON SURFACES IN AIR ENVIRONMENT

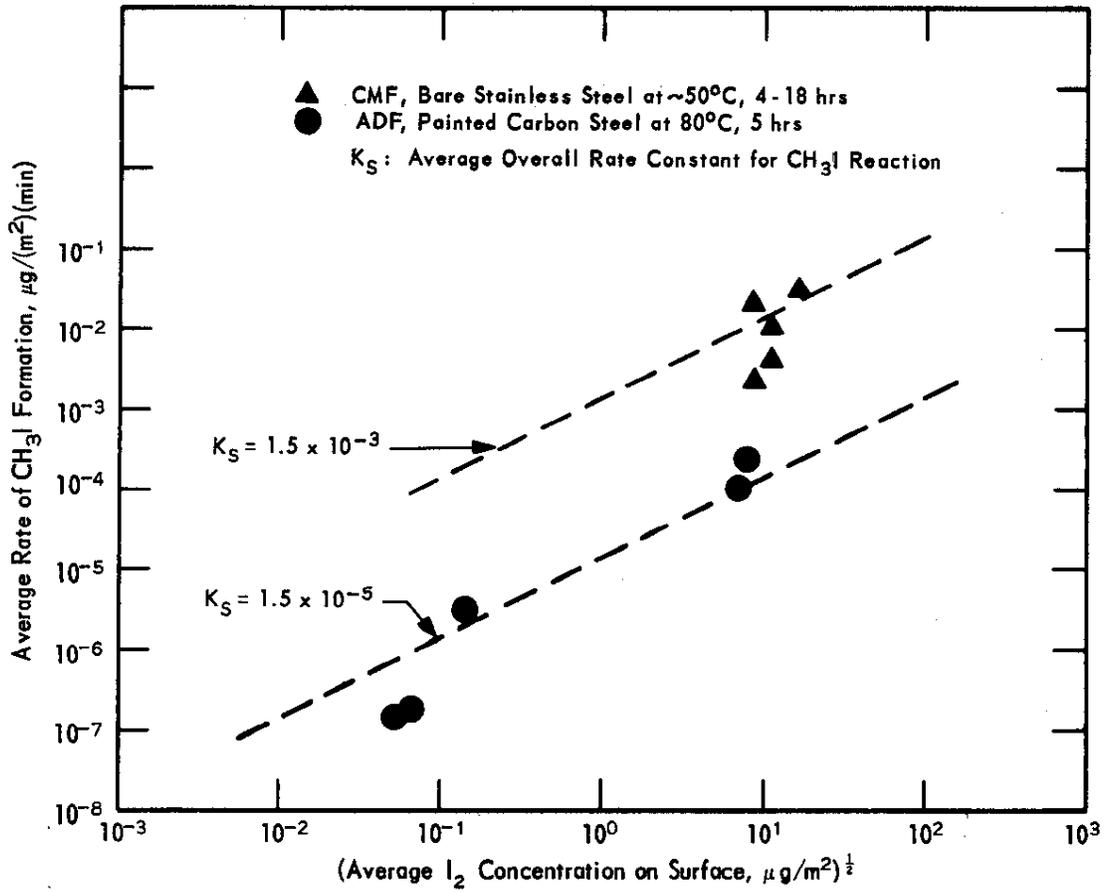


FIG. III-9 METHYL IODIDE FORMATION ON SURFACES IN STEAM - AIR ENVIRONMENT

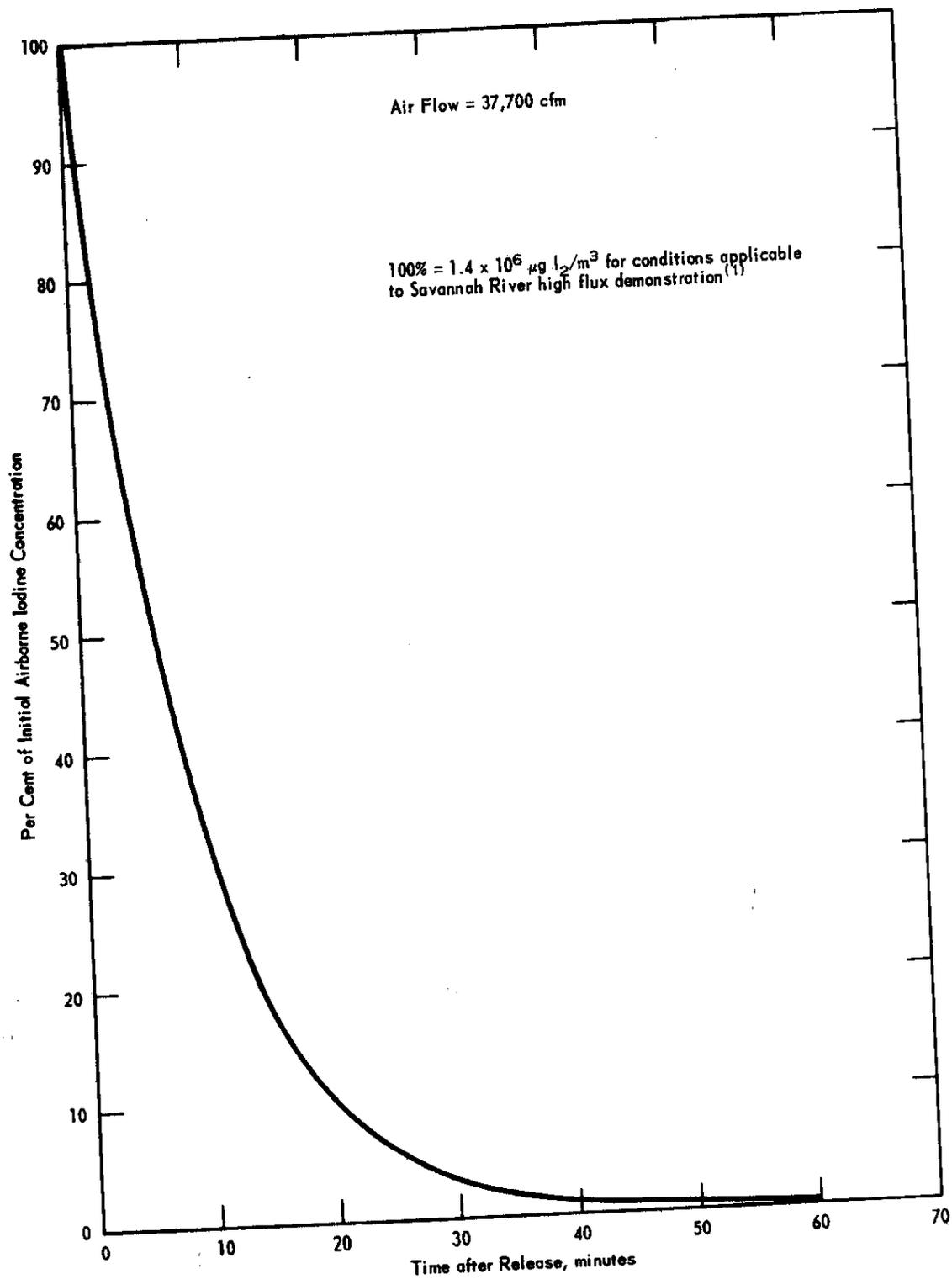


FIG. III-10 AIRBORNE IODINE CONCENTRATION IN SRP REACTOR ROOM

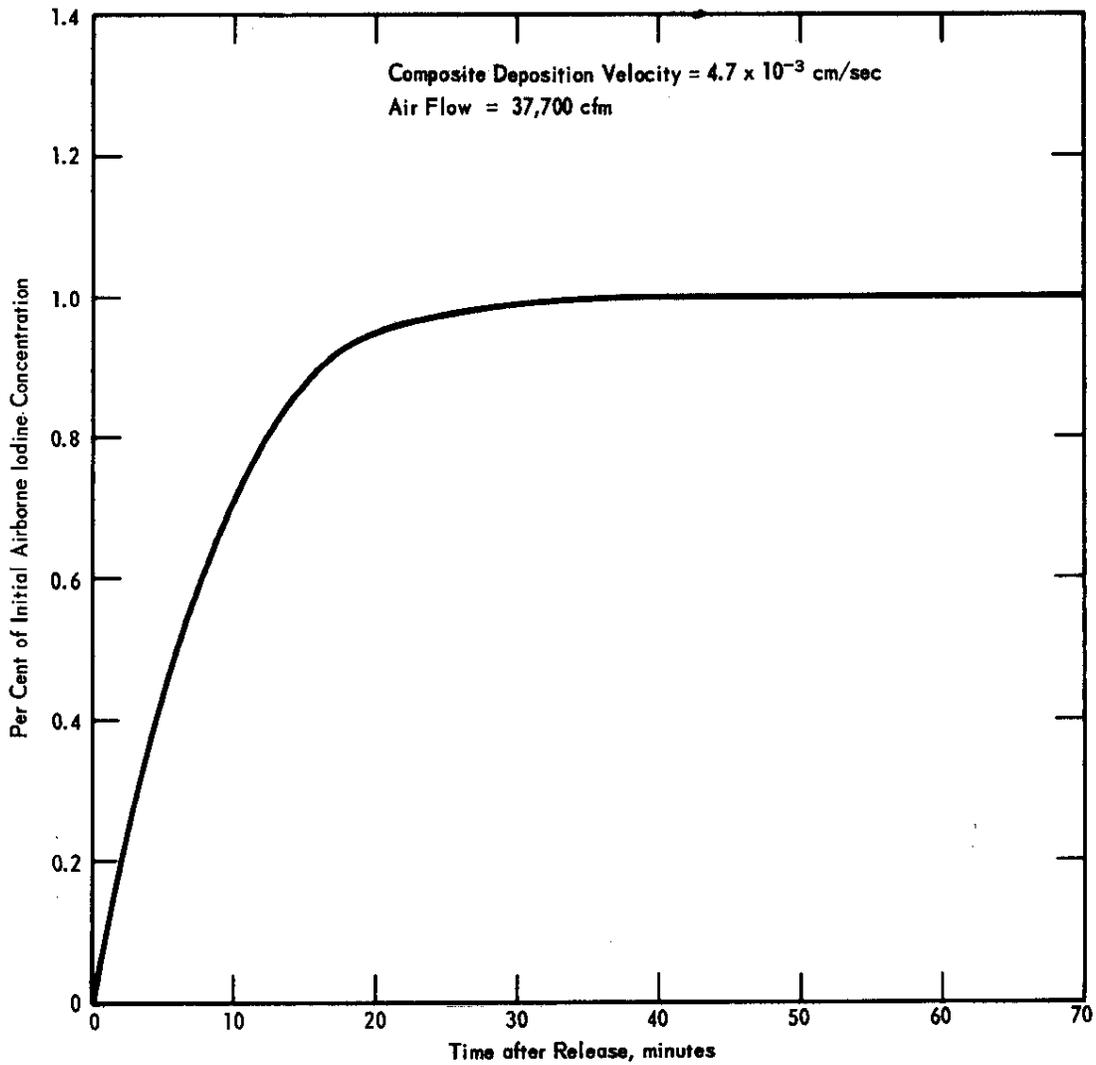


FIG. III-11 AMOUNT OF IODINE DEPOSITING ON SRP REACTOR BUILDING SURFACES

IV. FISSION PRODUCT COLLECTION

This section is concerned primarily with the relationship of the efficiency of the SRP activity collection system to the guidelines in 10 CFR 100⁽⁴²⁾. Information presented is based on operating conditions in the Savannah River high flux demonstration⁽¹⁾ and is summarized with respect to the type of fission products released; particulate matter, halogens, and noble gases.

A. PARTICULATE MATTER

Although the gamma radiation from particulate matter (the relatively nonvolatile fission products) is approximately equal to that from noble gases, the effect of particulate matter is of small concern in relation to ingestion and whole body dose because of the relatively small amount of material that would reach the plant boundary. SL-1,⁽⁴¹⁾ British,⁽⁴³⁾ and ORNL^(21,22,36,44) experience indicated that an assumption of 1% released from the reactor is reasonable. About 50% of the material released was estimated to deposit on building or containment surfaces prior to reaching the particulate filters. Based on a design efficiency of 99% (in-place DOP tests demonstrated an efficiency of 99.9+%, Sections II,D,2 and V,C), the fraction of particulate matter which would escape the reactor building even for the highly unlikely case of full core meltdown would be less than 0.00005, which is inconsequential compared to noble gas release.

B. HALOGENS

The primary concern with halogen activity is thyroid ingestion. The criterion in 10 CFR 100 is a dose of 300 rem at the plant boundary integrated over the time of passage of the cloud. In the highly unlikely event of a full core meltdown with a full inventory of fission products and under adverse meteorological conditions using conservative calculational methods, the average efficiency of the adsorbers must be equal to or greater than 99.42% (maximum penetration of 0.58% of the material reaching the banks) to remain within the 10 CFR 100 guidelines.

1. Factors in Evaluating Adsorber Units

Four factors were considered in evaluating the effectiveness of the iodine adsorption system.

- The efficiency of used carbon for removing elemental iodine vapor.
- Penetration of iodine in particulate matter not retained by particulate filters.

- Leak paths through the bed or gaskets.
- Iodine compound formation and retention.

The maximum and average penetrations that were measured or calculated to occur by the above mechanisms are summarized in Table IV-1.

TABLE IV-1

Iodine Penetration of Activated Carbon Beds (a)

Mechanism	I_2 (b) Vapor	Particulates	Gasket Leaks	Methyl Iodide (b)	Total
<u>Average (c)</u>					
Iodine Reaching Beds, % of total inventory	50	<0.0003	50	<0.0001	-
Penetration, %	0.02	100	<0.03	100	-
Iodine Leaving Beds, % of total inventory	0.01	<0.0003	<0.015	<0.0001	<0.026
<u>Maximum (c)</u>					
Iodine Reaching Beds, % of total inventory	50	0.0025	50	<0.0001	-
Penetration, %	0.04	100	0.05	100	-
Iodine Leaving Beds, % of total inventory	0.02	0.0025	0.025	<0.0001	<0.048

(a) 50% of the iodine was assumed to remain in the reactor.

(b) Assumes steam-air conditions; penetration would be less for air. Some of the iodine may reach the carbon beds as HI. As discussed in Section III, A,3, insufficient information is available to permit determination of the rate of oxidation of HI in air. Because HI is as readily adsorbed on activated carbon as I_2 , no attempt has been made to distinguish between the materials.

(c) Average refers to a minimum of three banks on line; maximum refers to a single bank.

The maximum penetration corresponds to an ingestion of 50 rem, well below the 10 CFR 100 guideline. The penetration was based on 50% of the iodine inventory in the reactor being released

into the reactor room as vapor and 0.5% in particulate matter. A 50% release from the fuel was assumed as is customary in reactor safety analysis⁽⁴⁰⁾, and a review of literature indicated that 50% was a typical value in experiments^(28,38) that were conducted under conditions favoring the release of iodine vapor. Less than a 50% release would be expected at SRP because temperatures and reaction times would be less favorable to iodine release than in the experiments. No credit was taken for solubility of iodine in D₂O, although the entire reactor inventory of elemental iodine is soluble in a small fraction of the D₂O in the reactor tank. Most of the iodine will appear in the reactor vessel, as hydrogen iodide, which is much more soluble in water than elemental iodine⁽⁴⁵⁾. Solutions of hydrogen iodide, like other solutions containing iodide ion, can dissolve large quantities of elemental iodine. For example, a solution saturated with iodine at 25°C contains 72.85% iodine, 18.1% hydrogen iodide, and 9.05% water, with a specific gravity of 3.28⁽⁴⁶⁾. Therefore the assumption of 50% release is considered highly conservative.

The four mechanisms are discussed in more detail in the following paragraphs.

a. Elemental Iodine Efficiency of In-Service Activated Carbon

The efficiency of new activated carbon in steam-air tests is greater than 99.99% for elemental iodine vapor⁽⁸⁾. However, as discussed in Section V,D, the efficiency is reduced by exposure to atmospheric impurities. The minimum efficiency that was measured on carbon from an operating reactor area was 99.96%⁽⁴⁷⁾. The carbon was removed from one of the original cadmium-plated frames of beds from K Area. The average efficiency, though, was 99.98%. In air tests the minimum measured efficiency was greater than 99.99%.

Efficiencies lower than 99.96% were measured on one carbon bed removed from a filter bank in R-reactor area after 14 months of operation. The minimum measured efficiency was 99.89% in a steam-air test. The R-reactor beds corroded (see Section V) much faster than those in other reactor areas; hence, there appeared to be a real difference between R reactor and the other areas in the environment that the carbon beds were exposed to. Because R reactor is no longer operating, the R-reactor experience was not used in calculating the maximum penetration for Table IV-1.

b. Penetration of Carbon Beds by Iodine in Particulate Matter

Iodine can be associated with particulate matter in two forms: physically or chemically adsorbed to the particle surface after release from the reactor, or trapped in the particle at the time the particle separates from the fuel. Based on 1% of the mass of fuel released from the reactor as particles, only about $5 \times 10^{-7}\%$ of the iodine released from the reactor as vapor could adsorb on particulate surfaces as a monomolecular layer. About 1000 times this amount would be required before this mechanism would be of any consequence. If it is assumed that the particulate matter escaping the reactor retains 50% of its initial iodine content, the second mechanism becomes important in specifying and maintaining high efficiencies for the particulate filters. In this case, it is assumed that half of the released particulates would settle in the reactor room so that only 0.5% of the total particulate matter (0.25% of the total iodine inventory) would reach the particulate filters. At an efficiency of 99% (the specified minimum), 0.0025% of the total iodine inventory would pass through the filters; at an efficiency of 99.9% (average measured) only 0.00025% would pass through. Because no filtering benefits can be claimed for the carbon beds, 100% penetration of beds by particulates was assumed.

Although a much lower efficiency for the particulate filters is required to remove nonvolatiles released from the reactor, the higher efficiency of 99% is specified to retain iodine associated with particulate matter.

c. Gasket Leaks

Bypassing of the exhaust stream around the activated carbon beds is detected by the in-place "Freon-112" test discussed in Section VI,A. The average leakage flow in the five compartments of C reactor was less than 0.03% of the total air flow through the compartments. A maximum leak of 0.2% was detected in one compartment. Retesting of the compartment after bolts holding the frames in place were tightened showed a leak of 0.04%. Compartments 6 and 5 had leaks of less than 0.01%. Compartments 4, 3, and 2 had leaks of 0.01, 0.04, and 0.05%; respectively.

The maximum for Table IV-1 was taken as 0.05% penetration and the average as 0.03%. The 0.2% value was not used because it represents a one-time situation that existed because of the absence of a leak test when the carbon beds were installed. With the recently developed "Freon-112" leak test and with improved installation and maintenance techniques, 0.05% represents the maximum for future use.

d. Iodine Compound Penetration

Details on iodine compound formation are presented in Section III and are summarized here. The quantity of methyl iodide that might be formed in the SRP system was conservatively determined to be equal to or less than 0.0001% of the total iodine inventory. Because the first contaminated vapor to reach the carbon beds probably would be in a steam-air environment; a zero efficiency for iodine compounds was assumed. The emergency light-water addition system in the reactor would rapidly stop steam generation; however, no credit was taken for any increase in iodine compound adsorption efficiency which might occur as the relative humidity of the exhaust air decreased. Additional drying of the carbon would occur from decay heat released from adsorbed iodine.

Under certain conditions, however, activated carbon has a high efficiency for methyl iodide. Activated carbon removes iodine compounds efficiently from a dry air stream. Typical tests that demonstrated this were run at Oak Ridge for 5 hours, at an air velocity of 25 feet per minute on a 1.5-inch-deep bed and at an iodine compound concentration of 0.05 milligram of iodine per cubic meter⁽³⁸⁾. The efficiency was dependent also upon the type of activated carbon used. Coconut carbon was more effective than either bituminous coke carbon or Whetlerite, a carbon impregnated with salts of silver, chromium, and copper. The coconut carbon had an efficiency of 99.89% for methyl iodide at room temperature and 4% relative humidity. At 100°C and about 0.1% relative humidity, efficiencies ranged from 99.91-99.99%⁽⁴⁸⁾.

Under wet steam-air conditions, the efficiency of activated carbon for removing methyl iodide was reduced. At room temperature and a relative humidity of 75%, the efficiency ranged from 50 to 97%, with coconut carbon being superior to the other types of carbon⁽⁴⁸⁾. Under steam conditions at temperatures from 95 to 130°C, the efficiency was reduced to 5 to 78%.

2. Removal of Decay Heat from Filter-Adsorber Units

Evaluation showed that the probability of igniting the carbon in the unlikely event of a nuclear incident is acceptably low. In the critical period after a possible incident, air flow less than 10% of normal will remove the decay heat from fission products trapped on the filters and adsorbers and maintain the carbon temperature below the ignition temperature of about 340°C.⁽⁴⁹⁾

As explained in Section II,D,2,d, two exhaust air fans are always operating and a third fan is available. The ignition temperature of the carbon could be approached only if all electrical power to the fans is lost after several minutes of normal operation following an accident. Power for the fans is fed in a loop circuit from two or three power plants that are widely separated. In the very unlikely event that all power plants should fail, power would be supplied by a diesel generator in each reactor area to maintain full system air flow. The exhaust fan system has never experienced an unscheduled stoppage in over 10 years of operation.

C. NOBLE GASES

Fission products in the fuel elements of nuclear reactors include radioactive noble gases (principally krypton and xenon). The existing SRP activity confinement system has no provisions to collect or attenuate noble gases. The only dose reduction credit for the existing system arises from dispersion due to release of the noble gases from a 200-foot-high stack rather than at ground level. Thus, in the very unlikely event of an accident in which the entire core is melted, the total equilibrium noble gas content was assumed to be released from the reactor building to the environs.

V. OPERATING EXPERIENCE

A. FILTER COMPARTMENTS

After 3 years of service, operating experience with the filter compartment shell has been excellent.⁽⁵⁰⁾ Only minor corrosion on the aluminum walls of compartments in one reactor area has been observed. The maximum depth of pitting, resulting from trace nitrogen compound attack, is about 0.005 inch and is of no consequence to the structural integrity of the compartments. Minor electrical problems in the remote delatching mechanism were experienced but were readily corrected.

B. MOISTURE SEPARATORS

Because the moisture separators are fabricated of stainless steel and "Teflon", no corrosion from trace acidic compounds has been observed. Also, no degradation of the "Teflon" has been detected.

The only problem with the separators has been an accumulation of an oily substance and particulate matter on the upstream face. Units are cleaned with steam at a nozzle pressure of 15-20 psi to restore flow-pressure drop characteristics to values equal to those of new units without damaging the mat. The front face is steam-cleaned for three minutes in a vertical position, and then the back face is steam-cleaned by blowing steam downward onto the back for three minutes with the separator in a horizontal position. The separators are removed from the compartments for cleaning.

No units have been replaced because of mechanical damage. The stainless steel and "Teflon" mat is quite tough and flexible and is not easily damaged from handling during installation or maintenance.

With periodic steam-cleaning about once per year, an indefinite service life is expected.

C. PARTICULATE FILTERS

Particulate filters have a relatively short life compared to moisture separators because there is no practical method for cleaning the medium; 1 to 2 years is typical. Wet fly ash from the powerhouse stack is the primary constituent in the exhaust air that accumulates on the filters. More efficient prefilters in the

supply air system currently are being installed to reduce the fly ash loading on the particulate filters as well as the separators. Although removal of the fly ash from the high-efficiency particulate filters is not practical, drying with electrical heaters at reduced flow and use of more efficient prefilters are expected to prolong the filter life. Experimental heaters in C Area proved the feasibility of drying the filters, and permanent installation of 80-kw heaters in each filter compartment has been authorized. Other benefits from the heaters are discussed in Section VI.

Early models of the particulate filters had no protection against puncture of the medium during installation, and the rejection rate was about one percent based on dioctylphthalate (DOP) tests of each unit after installation. To protect against puncture, use of galvanized hardware cloth face guards⁽⁵¹⁾ is now specified.

The ability of the particulate filters to withstand the severe conditions which might exist under emergency conditions was demonstrated in typical tests on two filters that were exposed for 13 months to flowing, normally dust-laden exhaust air in the P-Reactor Area ventilation system. Moisture separators had not been installed in the reactor areas at the time of this test. Tests at SRL showed that moisture separators remove at least 30% of atmospheric dust; thus the 13 months of service of the filters in the reactor area was equivalent to about 18 months of design service with separators. The filters, with moisture separators upstream, were subjected to flowing fog. Although very minor damage of one of the filters was observed, the efficiency of each filter for removing 0.6-micron particles of DOP was 99.9+% after 12 days of exposure to the flowing fog.

A test was performed also with both a dusty separator and two dusty particulate filters that were exposed for 34 weeks to flowing air that contained normal atmospheric dust. After 10 days of exposure to a fog mixture, both filters had an efficiency of 99.9+% for 0.6-micron DOP. Migration of the dust cake through the medium was negligible. Samples sectioned before and after the test showed a penetration of about 10% of the medium thickness.

The required efficiency of the filters for 0.3-micron* or larger particles is 99%, although measured efficiencies are greater than 99.9%^(s). The high rated efficiency can be main-

* Tests by the vendor and at the Oak Ridge Filter Testing Facility are with 0.3-micron aerosols; the tests at SRP are with 0.6-micron aerosols.

tained even though the filters may contain a considerable number of holes. For example, four banks of 128 clean particulate filters (about 30,000 square feet total surface area) can contain about 1100 $\frac{1}{4}$ -inch-diameter holes, yet allow only 1% of the air stream to pass through the holes⁽⁵²⁾ at normal flow.

D. ACTIVATED CARBON BEDS

The efficiency of activated carbon after more than 2 $\frac{1}{2}$ years of continuous exposure to exhaust ventilation air from Savannah River Plant reactor buildings remains greater than the design objective of 99.9% for the removal of elemental iodine. Details of the performance of the beds are presented in DP-1028⁽⁴⁷⁾ and are summarized herein. Shown in Table V-1 are the results of elemental iodine tests on carbon exposed to reactor building exhaust ventilation air for about 2-1/2 years⁽⁵⁰⁾.

Although the beds continue to fulfill their function of providing protection from halogen activity, two problems have arisen. The first is that the mild steel frames which contain the carbon are corroding from nitrogen and sulfur compounds that are in the building exhaust air in trace quantities. The corrosion is intergranular which results in severe embrittlement of the metal. Both the original cadmium-plated units and the later painted frames have been attacked. Failure of the perforated metal screen of only two units has been detected, and these failures resulted in a negligible loss of carbon. New units fabricated of Type 304L stainless steel have been ordered, and replacement will begin during the summer of 1966. Details of the stainless steel units are in the Appendix.

The second problem is that of slow degradation of the carbon from adsorption of impurities, primarily NO₂. NO₂ is formed by radiolytic decomposition of atmospheric nitrogen and subsequent reaction with oxygen in the space between the reactor tank and the biological shield. The 200-Area separations plant and nearby industrial plants also contribute NO₂ to the atmosphere. Analysis of used carbon shows only the presence of NO⁽⁴⁷⁾, so it is assumed that because free NO would not normally exist in air, NO₂ oxidizes the carbon and the residual NO sorbs on available sites. NO can be removed from the carbon by drying the bed in-place at about 60°C. The drying operation partially restores the iodine adsorption efficiency to about 99.98%, and the service life of the carbon will be improved. Less than 0.01 g NO₂/g carbon would be formed in the reactor during a major accident. No effect on the efficiency of the carbon for iodine would be expected for this concentration.

Contaminants in the carbon that reduce efficiency migrate through the bed even at room temperature. Initially, the material is sorbed in greatest concentrations on the front face of the bed. Although the efficiency at the front face may be low, the overall efficiency of the bed still remains high. This was demonstrated by tests on carbon beds from K Reactor Area, as shown in Table V-1.

As would be expected, the efficiency of used carbon in air tests was higher than in steam-air tests.

As mentioned above, all carbon beds are scheduled for replacement starting in the summer of 1966. Tests of the carbon will be made periodically to ensure that beds will be replaced when efficiencies begin to fall below a satisfactory level.

TABLE V-1

Iodine Tests of Used Carbon Samples from SRP Reactor Areas

Test No.	Area	Bed Serial No.	Months of Service	Type Test	Efficiency %	Bed Loading mg I ₂ /g carbon	Notes
113	R	24	14	Steam-air	99.95	1.3	(1,2)
114	R	24	14	Steam-air	99.92	0.9	(1,2)
115	R	24	14	Air	99.99	0.8	(1,2)
117	R	24	14	Steam-air	99.89	0.6	(1,2)
119	R	24	14	Steam-air	99.99+	0.5	(1,3)
122	R	23	14	Steam-air	99.98	0.5	(1,2)
123	R	23	14	Steam-air	99.98	0.4	(1,4)
124	R	23	14	Steam-air	99.98	0.4	(1,5)
111	P	114	15	Air	99.99+	1.6	(1,2)
112	P	114	15	Steam-air	99.99+	1.2	(1,2)
118	P	114	15	Steam-air	99.99+	1.2	(1,2)
164	P	613	15	Steam-air	99.99+	0.6	(2,15)
187	P	91	28	Steam-air	99.97	0.4	(1,2)
189	P	91	28	Steam-air	99.96	0.3	(2,15)
202	P	91	28	Steam-air	99.98	0.6	(2,15)
203	P	91	28	Steam-air	99.92	0.4	(2,15)
204	P	91	28	Steam-air	99.98	0.5	(2,15)
205	P	91	28	Steam-air	99.98	0.5	(2,15)
206	P	91	28	Air	99.98	0.7	(2,15)
133	L	132	20	Steam-air	99.97	0.4	(1,2)
134	L	132	20	Steam-air	99.99+	0.5	(1,6)
135	L	132	20	Steam-air	99.99+	0.5	(7,2)
136	L	132	20	Steam-air	99.99+	0.6	(8,2)
137	L	132	20	Steam-air	99.99	0.6	(9,2)
138	L	132	20	Steam-air	99.94	0.6	(10,2)
139	L	132	20	Steam-air	99.99	0.6	(11,2)
185	L	40	28	Steam-air	99.96	0.5	
140	K	76	20	Steam-air	99.99+	0.6	(1,2)
141	K	76	20	Steam-air	99.89	0.7	(8,2)
143	K	76	20	Steam-air	99.60	0.4	(8,2,12)
144	K	76	20	Steam-air	99.93	0.6	(8,2,13,14)
145	K	76	20	Steam-air	99.96	0.6	(1,2)
146	K	76	20	Steam-air	99.92	0.6	(10,2)
180	K	185	28	Air	99.99+	0.7	(2,15)
184	K	185	28	Steam-air	99.98	0.5	(2,15)
120	C	668	7	Steam-air	99.99	0.6	(1,2)
121	C	668	7	Steam-air	99.99+	0.7	(1,2)
158	C	650	17	Steam-air	99.98	0.5	(1,2)
201	C	650	17	Air	99.99+	0.9	(2,15)

Notes:

- (1) Test bed was packed with 3 layers of carbon taken from the front, middle, and back of the 1-in.-thick layer in the full-size bed.
- (2) Carbon was not dried.
- (3) Carbon was dried for 20 hr at 95°C and 70 fpm air flow.
- (4) Carbon was dried for 24 hr at 95°C and 70 fpm air flow.
- (5) Carbon was dried for 24 hr at 65°C and 70 fpm air flow.
- (6) Carbon was dried for 16 hr at 65°C and 70 fpm air flow.
- (7) Test bed was packed with carbon from the front 1/2" of full-size carbon bed.
- (8) Test bed was packed with carbon from the front 1/8" of full-size carbon bed.
- (9) Test bed was packed with carbon from the back 1/2" of full-size carbon bed.
- (10) Test bed was packed with carbon from the center 1/3" of full-size carbon bed.
- (11) Test bed was packed with carbon from the back 1/8" of full-size carbon bed.
- (12) Carbon removed from full-size bed 1 wk after carbon removed for test No. 141.
- (13) Carbon for tests No. 143 and 144 identical.
- (14) Carbon was dried for 16 hr at 95°C and 70 fpm air flow.
- (15) Test bed was packed with a thoroughly mixed carbon sample removed from full-size bed.

Conditions for air test:

Duration of test: 2-1/2 - 3 hr
 Face velocity: 70 ft/min
 Temperature at test bed: 35°C (95°F)
 Isotopic dilution: 1.5 mCi ¹³¹I per 100 mg ¹²⁷I

Conditions for steam-air test:

Duration of test: 2-1/2 - 3 hr
 Face velocity: 70 ft/min
 Steam flow: 1 ft³/min
 Temperature of test bed: 68°C (155°F)
 Isotopic dilution: 1.5 mCi ¹³¹I per 100 mg ¹²⁷I

VI. EXPERIMENTAL AND DEVELOPMENT PROGRAM

A. NONDESTRUCTIVE LEAK TEST OF CARBON BEDS

The efficiency of installed beds of carbon is determined by a combination of two tests: 1) a nondestructive test to measure the magnitude of leaks past the carbon in the entire bank of beds, and 2) a destructive test of representative samples of the carbon to ensure that the carbon has a high efficiency for adsorption of iodine. The nondestructive leak tests are described in this section and the destructive iodine tests are described in Section VI,B.

1. "Freon-12" Leak Tests before Installation of Carbon Beds

The carbon beds of the SRP activity confinement system were designed to remove 99.9+% of elemental iodine vapor. To achieve this high efficiency, the units were carefully designed and tested, as discussed in previous sections of this report. After manufacture of the carbon bed unit and before installation in the confinement system, evaluation of the units for absence of leak paths was desired. A nondestructive test was developed by SRL⁽¹¹⁾ to detect leaks caused by settling or voids in the carbon beds. "Freon-12" was used as a tracer to detect any leak paths.

The "F-12" leak test was used to evaluate over 800 new carbon beds before installation in the confinement systems. The average leakage flow for all the beds was less than 0.01%. Only one bed had a leak greater than 0.1%; inspection showed that the carbon had either settled during shipment or was not completely packed by the manufacturer during the loading operation. Carbon was added to the bed, and an "F-12" retest showed that the leakage flow was reduced to less than 0.01%. These "F-12" tests demonstrated the adequacy of the SRL design of carbon beds to prevent leaks that could bypass the carbon and reduce the iodine adsorption efficiency of the bed to less than 99.9%. The tests showed that "Freon-12" was satisfactory for new and relatively dry carbon at air velocities to 20 feet per minute.

Measurement of any leaks in a carbon bed required knowledge of three facts: 1) the concentration of the tracer in the flowing air upstream of the test bed, 2) the concentration of the tracer in the flowing air downstream of the test bed, and 3) assurance that the carbon at test conditions would remove essentially all (about 99.99%) of the tracer if all the air flow passed through the 1-inch-thick bed of carbon. Therefore, any tracer that bypassed the carbon bed would be caused by leaks in the bed and/or around gasket seals. Numerous tests demonstrated that the "F-12" technique would detect leaks less than 0.01% of the total air flow through the beds. "F-12" downstream of the bed was measured by an electron-capture-type detector with a gas chromatograph⁽¹¹⁾.

"F-12" was readily desorbed by normal exposure to the exhaust air flow; however, tests showed that "F-12" adsorbed on the carbon did not interfere with the iodine adsorption characteristics of the carbon. The "Freon" compounds offer other desirable characteristics as a tracer, i.e., they are nontoxic, easy to handle, noncorrosive, and readily available.

2. "Freon-112" In-Place Test of Installed Carbon Beds

During the design of the confinement system, it was recognized that a method of testing the carbon beds in-place would be required to establish that air flow around gasket seals is insignificant. Although each carbon bed was evaluated before installation, gaskets on the carbon beds that seal the units in mounting racks of the compartment could be tested only in-place. The "F-12" technique discussed in the preceding section was unsatisfactory for used carbon beds, because impurities and moisture adsorbed on the carbon from continuous exposure to exhaust air flow interfered with "F-12" adsorption.

Because of the above limitations, the Savannah River Laboratory developed a nondestructive test that is generally applicable for in-place leak testing of installed carbon beds, both new and used. The limitations on air velocity and sorbed water when testing new carbon were reduced significantly by substituting the less volatile "F-112" ($\text{CCl}_2\text{F}-\text{CCl}_2\text{F}$)^(53,54,55) for "F-12". This work is being sponsored by the AEC Division of Operational Safety.

Techniques for in-place leak testing of a complete bank of installed carbon beds with "F-112" were developed and demonstrated.^(56,57) Tests of used carbon beds in five SRP filter compartments showed that the average leakage flow (flow that bypasses the carbon) was less than 0.03% of the total air flow through the compartments. Compartments 6 and 5 had leaks of less than 0.01%; compartments 4, 3, and 2 had leaks of 0.01, 0.04, and 0.05%, respectively, in C reactor. The results of the tests are shown in Figure VI-1. In compartment 3, an initial leakage flow of about 0.2% was reduced to the acceptable value of 0.04%, as discussed on page IV-4. "F-112" leak testing of carbon beds in all SRP reactors is in progress.

The average leakage flow for the five banks of carbon beds corresponds to an average efficiency greater than 99.96% for removal of elemental iodine vapor, because small-scale tests (in which there were no detectable leak paths) showed that the iodine adsorption efficiency of representative samples of the carbon from C-reactor was greater than 99.99%. The beds that were

tested had been exposed continuously for about 2 years to exhaust air flow in the confinement system prior to the "F-112" and iodine tests.

The "F-112" leak test required only one minor modification of the compartment. A mixer was installed downstream of the carbon beds to adequately mix the total compartment flow prior to sampling for "F-112" detection. Adequate mixing was essential to prevent possible channeling of any "F-112" from leak paths adjacent to the compartment walls. The effectiveness of the mixer was demonstrated by tests in one of the compartments. The device was removed before returning the compartment to normal service. A schematic of an SRP compartment and equipment used for the "F-112" leak test is shown in Figure VI-2.

The in-place "F-112" test requires a minimum knowledge of the actual conditions in the test system. The precise values of air flow, "F-112" injection rate, and calibration of the "F-112" detectors are not required. Only the relative sensitivities of two detectors for "F-112" are required. Gas chromatographs with electron-capture-type detectors are used for measuring "F-112" concentrations in the air samples. The detectors have a sensitivity of 1 ppb (vol.) for "F-112" in air. Complete details on the "F-112" detectors are discussed in References 11 and 56.

New carbon is leak tested with "F-112" at air velocities to 70 feet per minute and with sorbed water to about 20%. When testing used carbon beds in place, air velocity and sorbed H₂O are reduced for a quantitative leak test. Used carbon is dried in place prior to the test by preheating the inlet air (at about 1/10 normal flow) to about 60°C.

Exposure of carbon beds to normal exhaust air flow desorbs over 95% of the "F-112" in about three weeks; however, tests showed that at least 4 times the maximum concentration of "F-112" adsorbed on the carbon does not affect its efficiency for iodine removal.

The "F-112" test has been standardized for general application for in-place testing of carbon beds in other reactors in addition to those at SRP^(5a).

B. IODINE TESTS

1. Small-Scale Tests

Small-scale tests with elemental iodine are discussed in detail in References 8 and 47. A photograph of the equipment is shown in Figure VI-3. Elemental $^{127}\text{I}_2$ and tracer $^{131}\text{I}_2$ (prepared to avoid iodine compound formation⁽⁸⁾) are deposited on a fritted glass tube, called a cold finger, and reduced in temperature with dry ice to retard sublimation. After the cold finger is mounted in the test apparatus, a small stream of warm air vaporizes the iodine and carries it into a main stream of air or steam and air. The material then passes through a moisture separator, particulate filter, test carbon bed, a second particulate filter, and three backup carbon beds. Condensate is removed at seven locations. The efficiency of a test carbon bed is calculated from the ^{131}I disintegrations measured on the test bed compared with the disintegrations measured on all units downstream.

This equipment currently is being used for continuing tests of the iodine removal efficiency of SRP carbon beds⁽⁴⁷⁾, for evaluation of high ignition temperature materials⁽⁵⁹⁾, for determination of effects of "Freon" on iodine adsorption efficiency⁽⁵⁸⁾, and for testing with methyl iodide.

2. Full-Size Tests

A method of determining the iodine removal efficiency of full-size carbon beds is being developed. The primary purpose of the experiment is to correlate the short term "F-112" adsorption efficiency and the iodine adsorption efficiency of full-size beds. The test facility (Figure VI-4) used for development of the "F-112" in-place leak test will be used also for the iodine tests. The proposed test method will use ^{127}I (nonradioactive) as a tracer to minimize safety hazards and utilize existing facilities. The ^{127}I detection method will be activation analysis, as discussed in Section VI,B,3, of iodine traps located in sample lines upstream and downstream of the test carbon bed. A gas chromatograph will be used also to detect any methyl iodide that might be formed.

3. Analytical Techniques

The development of tests for iodine adsorption by full-size carbon beds requires a sensitive analytical determination for elemental iodine. Neutron activation analysis offers a rapid, sensitive method.

An existing procedure⁽⁵⁰⁾ will be used to determine iodine in samples of activated carbon. Sealed capsules containing known amounts of HIO_3 are embedded in the samples of iodine-containing carbon that are to be analyzed. The samples are irradiated for 1 hour at a flux of $2 \times 10^{11} \text{ n}/(\text{cm}^2)(\text{sec})$. Iodine is leached from the carbon with $\text{NH}_4\text{OH}-\text{H}_2\text{O}_2$, separated from interfering radionuclides by three cycles of oxidation-reduction and extraction into CCl_4 , and then precipitated as AgI . The induced ^{128}I is determined by gamma pulse height analysis of the AgI , and iodine in the carbon is calculated on the basis of comparative counting of the irradiated encapsulated HIO_3 . The sensitivity is 1 microgram I_2 per 100 grams of carbon.

Any methyl iodide formed during tests of full-size carbon beds will be detected by an electron capture detector used in conjunction with a gas chromatograph. The detector has a sensitivity of about 0.1 ppb (wt) for methyl iodide in air. Details are presented in References 11, 61, 62, and 63.

C. METHYL IODIDE TESTS

Testing of the efficiency of activated carbon for methyl iodide is in progress at SRL. The same apparatus (Figure VI-3) used for small-scale elemental iodine tests is being used. Initial results are shown in Table VI-1.

Tests 178 and 194 confirmed that under dry conditions, the efficiency of coconut shell activated carbon for methyl iodide is in the order of 99% and is a function of iodine loading.

Tests 196 and 197 are significant because the results indicate that for the test $\text{CH}_3 \text{ }^{131}\text{I}$ was subjected to both exchange reactions and chemical decomposition. Penetrations of the test bed by $\text{CH}_3 \text{ }^{131}\text{I}$ was not detected. The results confirm the reactions discussed in Section III,A (Equation 22). About 8 times more $^{127}\text{I}_2$ and $^{129}\text{I}_2$ than $^{131}\text{I}_2$ would be released during an incident, and the iodine may be released either in water or a condensing steam atmosphere that would favor decomposition of $\text{CH}_3 \text{ }^{131}\text{I}$. Further testing with methyl iodide is in progress.

TABLE VI-1

Small-Scale Methyl Iodide Tests of SRP
Activated Carbon^(a)

No.	Material and Quantity	Loading CH ₃ I, mg/g C	Loading I ₂ , mg/g C	Efficiency, %
178	CH ₃ ¹³¹ I 0.11 mc	0.06	0	93.4
	CH ₃ ¹²⁷ I 5.7 mg			
194	CH ₃ ¹³¹ I 0.002 mc	0.008	0	99.4
	CH ₃ ¹²⁷ I 0.5 mg			
196	CH ₃ ¹³¹ I 0.001 mc	-	1.0	99.99+
	CH ₃ ¹²⁷ I 0.5 mg			
	¹³¹ I ₂ 0.24 mc			
	¹²⁷ I ₂ 100 mg			
197	CH ₃ ¹³¹ I 0.0013 mc	-	0.8	99.99+
	CH ₃ ¹²⁷ I 10.0 mg			
	¹³¹ I ₂ 0.13 mc			
	¹²⁷ I ₂ 100 mg			

(a) Test medium: Air - precleaned by passage through a 2-foot-deep bed of Type 416 carbon.

Face velocity: 70 feet per minute.

Bed temperature: 40°C.

Test carbon: Barnebey-Cheney, Type 416, activated coconut shell.

Relative humidity: 10%.

Iodine carrier: 1½ milliliters water.

Time of test: 1 hour.

D. IGNITION TEMPERATURE OF HALOGEN ADSORBERS

The temperature of the units of a filtration-adsorption system will increase after a major accident because of heat generated by radioactive decay of trapped fission products. The increase depends primarily upon:

- Temperature and air flow through the system
- Quantity of trapped fission products
- Heat loss from the system

The distribution of the fission products among the units and the heat capacity of the system components will also affect the temperature of individual units, but to a lesser extent than the above factors. If air flow stops, the carbon beds conceivably might be heated to the ignition temperature in some confinement applications.

Studies are being made at SRL on halogen adsorbers to reduce the probability of ignition.^(59,64) A standardized procedure is being developed to measure ignition, to evaluate new adsorber materials, and to determine the effects of such variables as air flow, particle size, internal surface area, and catalysts on ignition temperature. This work is being sponsored by the AEC Division of Operational Safety.

1. Literature Survey of Adsorptive Materials

Published data indicate that the iodine adsorptive properties of existing types of noncombustible substitutes for activated carbon are not satisfactory for most reactor confinement applications. Hence, work at SRL has been concentrated on raising the ignition temperature of coconut shell activated carbon. Although coconut shell carbon has a lower ignition temperature than carbon prepared from coal, the coconut shell carbon is preferred because it is more adsorptive for elemental iodine and is more resistant to erosion and abrasion.

Published ignition temperatures are inconsistent, apparently because of variations in the measuring apparatus and procedures. Conditions that affect the ignition temperature have been reviewed⁽⁵⁹⁾.

2. Small-Scale Ignition Temperature Apparatus

A small-scale apparatus was developed for measuring ignition temperature of carbon beds with diameters and depths up to 3 inches, as shown in Figure VI-5. The 3-inch diameter was chosen to simulate a full-size bed.

An ignition temperature of 340-350°C was measured for a 1-inch-thick by 3-inch-diameter bed packed with activated carbon now used in the SRP confinement system (Type 416* coconut shell). Air velocity had little effect on the ignition temperature. Exposure in the confinement system for 18-25 months increased the ignition temperature about 60°C. This increase is attributed to the increased concentration of iodine, chlorine, and phosphorous in carbon, as shown by mass spectroscopic analysis. These materials (products of the combustion of coal in the vicinity) inhibit the oxidation of carbon and increase the ignition temperature.

An ignition temperature of about 550°C was measured for Type 592,* a new high temperature coconut shell carbon, packed in a 1-inch-thick bed. Detailed evaluation of the iodine adsorption and the ignition characteristics of this promising new carbon is in progress at SRL. Preliminary tests indicate that Type 592 carbon meets SRP specifications outlined in Appendix C. The ignition temperatures of several other commercial carbons were measured also. Type BPL** (bituminous carbon) ignited at 470°C, and Type PCB** (coconut carbon) ignited at 340°C. An ignition temperature of 250°C was measured for Whetlerite carbon.

3. Ignition Tests of Full-Size Carbon Bed

The ignition temperature of carbon was measured in tests at the Lawrence Radiation Laboratory (LRL) in a prototype of the new SRP bed (described in Reference 47) framed with stainless steel. Figure VI-6 is a schematic diagram of the apparatus. The measured ignition temperatures were 340°C for Type 416 carbon at an air velocity of 70 ft/min and 530°C for Type 592 carbon at an air velocity of 105 ft/min. These values are in good agreement with those obtained from the small-scale apparatus.

* Product of Barnebey-Cheney Company.

** Product of Pittsburgh Activated Carbon Company.

E. REMOVAL OF IODINE AND NOBLE GASES BY ISOTOPIC EXCHANGE

Experiments at ORNL⁽⁶⁵⁾ showed that carbon impregnated with iodine, potassium iodide, potassium bromide, or potassium bromate removed methyl iodide efficiently from moist air. Isotopic exchange is the proposed mechanism for the efficient removal of methyl iodide. However, carbon impregnated with potassium iodide removed five times more methyl iodide than would be expected by an exchange mechanism. Furthermore, molecular sieve and alumina impregnated with these salts removed methyl iodide from moist air at reduced efficiency. Therefore the mechanism is probably more complex than simple isotopic exchange.

Isotopic exchange of organic iodides has been described in the literature^(66, 67, 68, 69, 70, 71) and has been applied in the confinement of iodine. The kinetics have been studied in liquid phase for the exchange of n-butyl and n-propyl iodide with zinc iodide⁽⁶⁶⁾; ethyl iodide^(67, 68) and isopropyl iodide⁽⁶⁹⁾ with iodine; methyl iodide with silver iodide⁽⁷⁰⁾; and ethyl iodide with aluminum triiodide⁽⁷¹⁾. In the work most nearly related to confinement^(67, 68, 69) the exchange proceeded more rapidly with decreasing iodine concentration. The half-time of the exchange was predicted to be 5×10^{-2} second in 10^{-11} mole per liter of solution but 5×10^3 seconds in 10^{-6} mole per liter of solution. Additional study of the published literature is in progress.

The isotopic exchange of noble gases with noble gas compounds may have an application for confinement. No work has been reported on this exchange in the literature. In preliminary work at SRL, the exchange of xenon with sodium perxenate was tested at room temperature in a helium atmosphere. No exchange was detected.

The planned work on isotopic exchange at SRL includes completion of the literature survey, selection and testing of promising compounds for exchange of iodine and noble gases, experiments designed to clarify the mechanism of the retention of radiiodine atoms of methyl iodide on activated carbon, and an engineering evaluation of the feasibility of impregnating carbon beds with xenon compounds. This work is being sponsored by the AEC Division of Operational Safety.

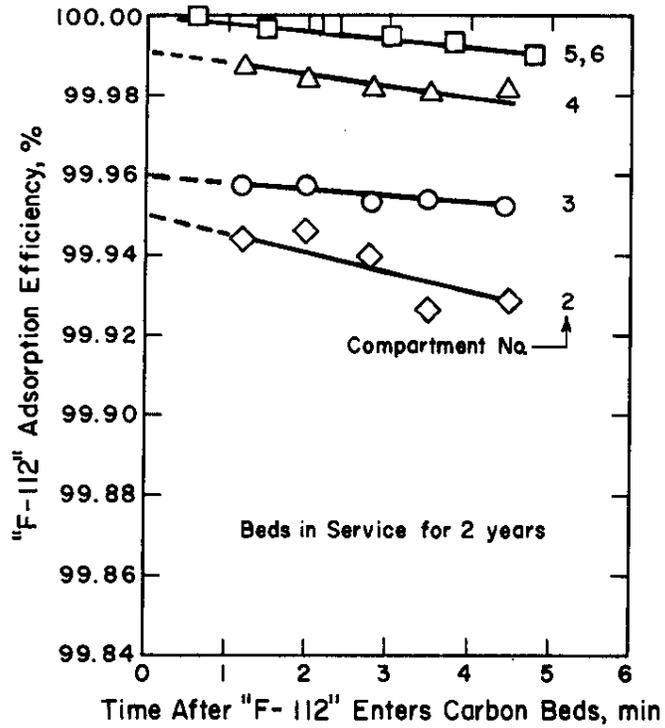


FIG. VI-1 "F-112" ADSORPTION EFFICIENCY OF BANKS OF CARBON BEDS INSTALLED IN C REACTOR. The per cent of flow bypassing the carbon beds (leak path) equals 100% minus the "F-112" adsorption efficiency at zero time.

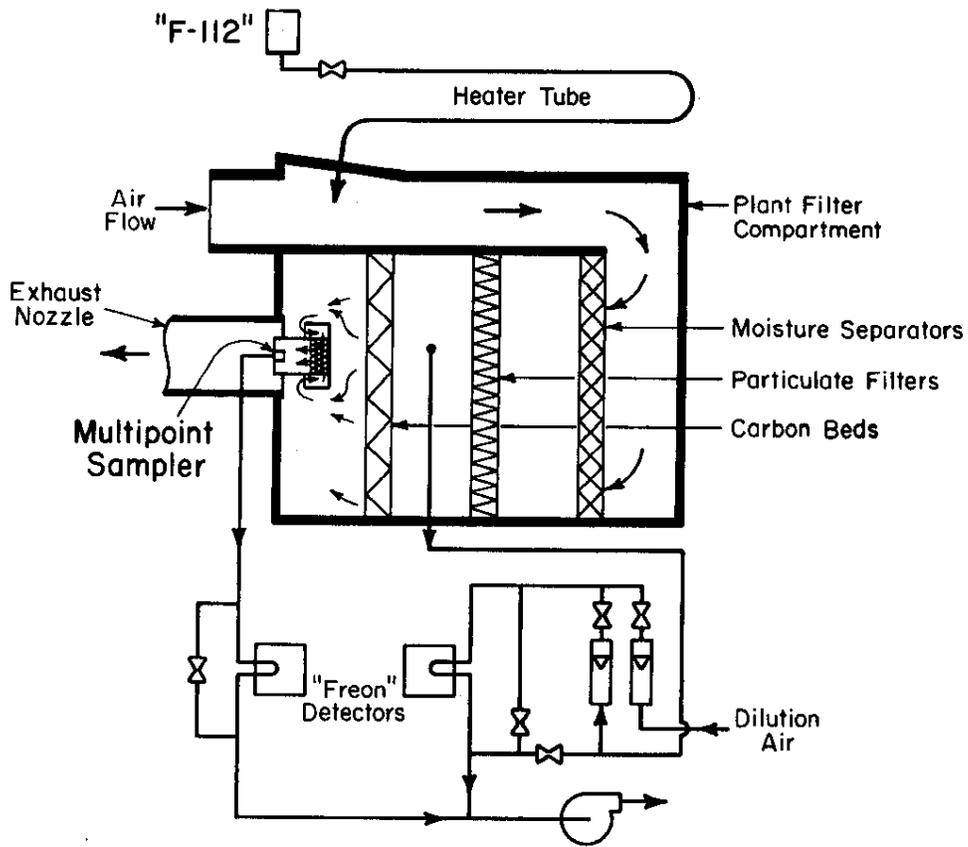
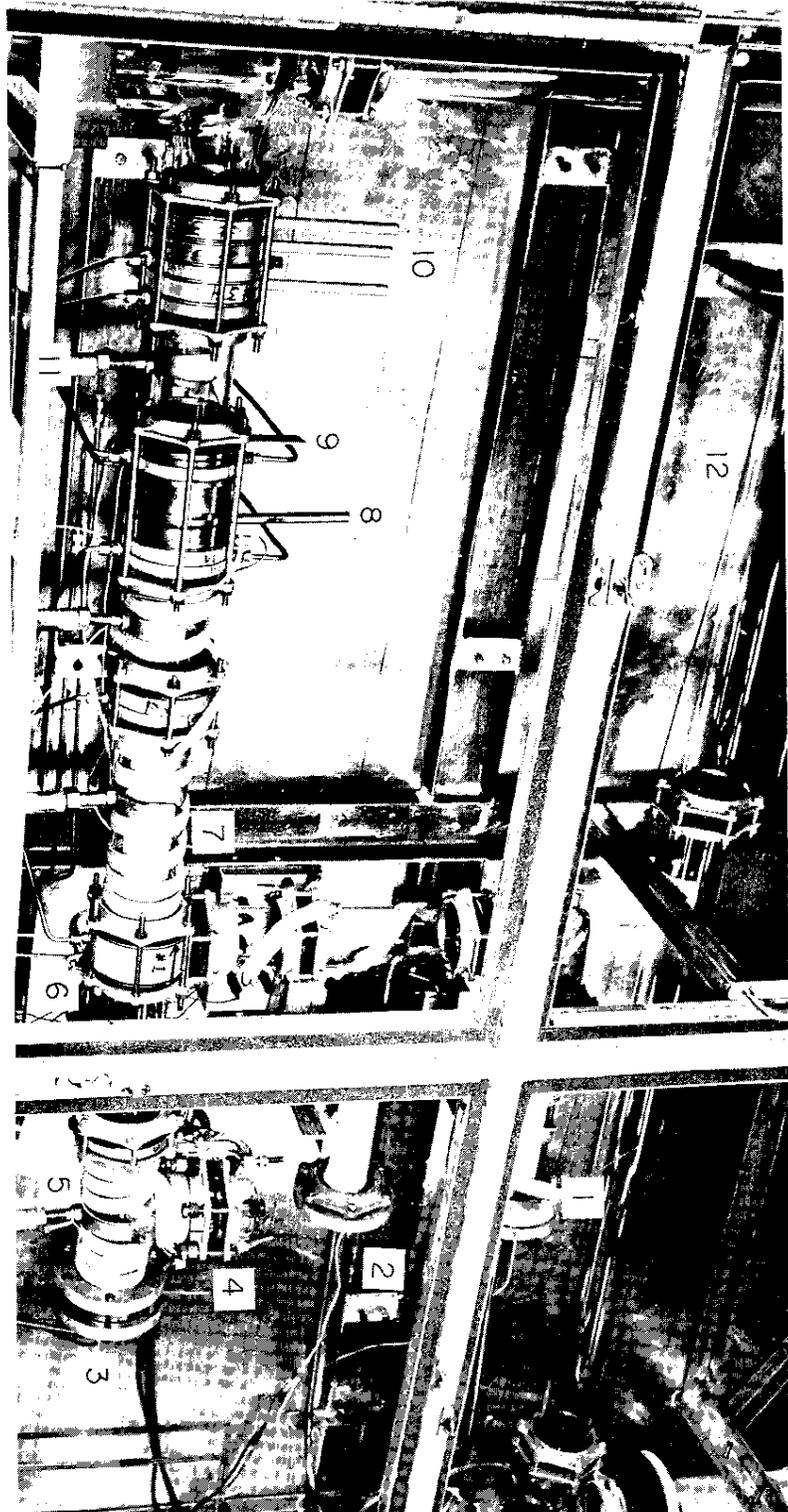


FIG. VI-2 EQUIPMENT FOR IN-PLACE TESTING OF PLANT CARBON BEDS



- 1. Steam supply
- 2. Main air supply
- 3. Bypass air supply
- 4. Iodine source
- 5. Typical heating tape
- 6. Moisture separator
- 7. Particulate filter
- 8. Test carbon bed
- 9. Particulate filter
- 10. Backup carbon beds
- 11. Typical of 6 condensate drains
- 12. To vacuum pump

FIG. VI-3 IODINE-131 TEST FACILITY

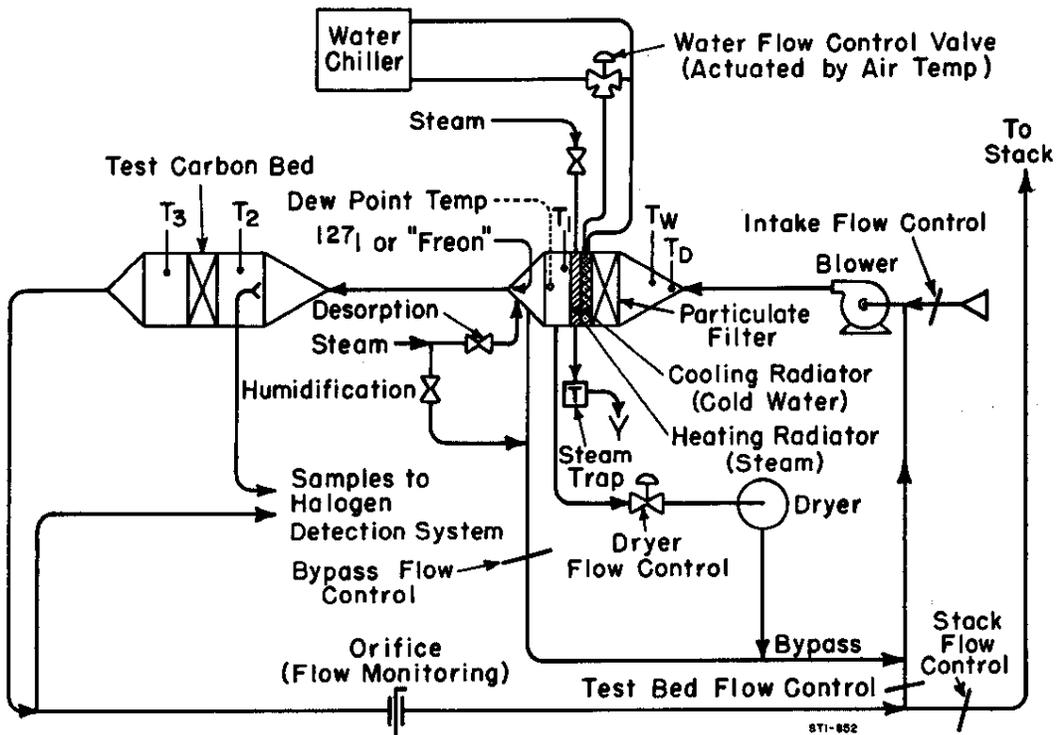


FIG. VI-4 FLOW DIAGRAM OF TEST DEVELOPMENT FACILITY FOR FULL-SIZE IODINE ADSORBERS

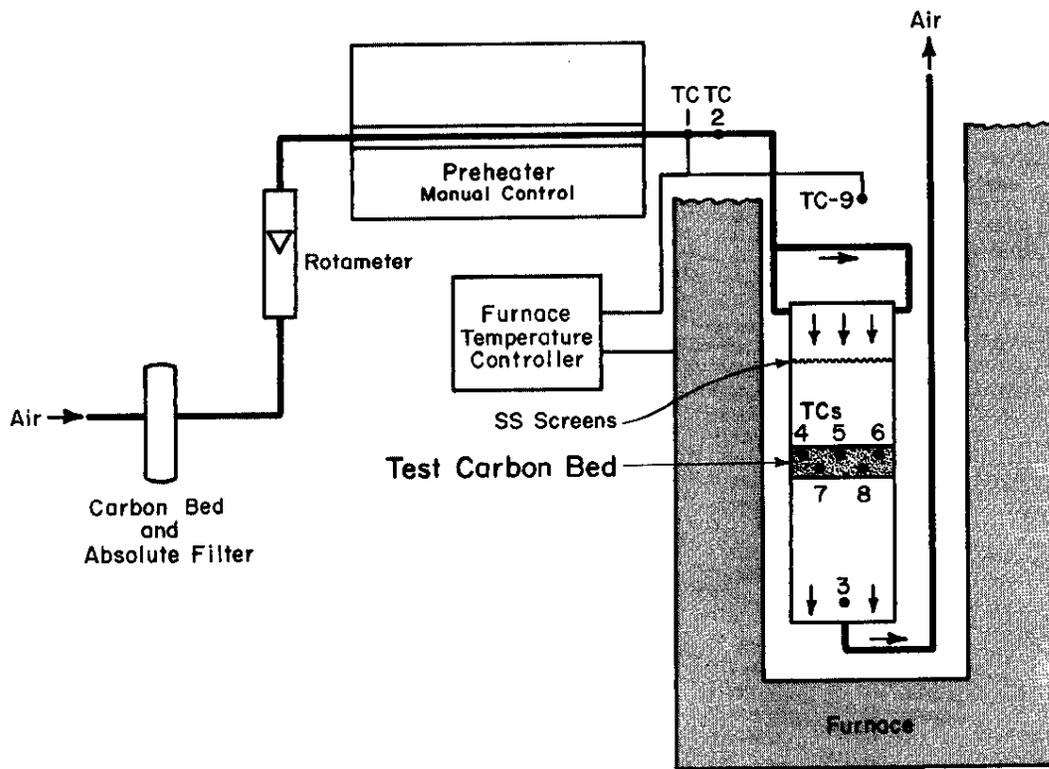


FIG. VI-5 SMALL-SCALE IGNITION TEST EQUIPMENT

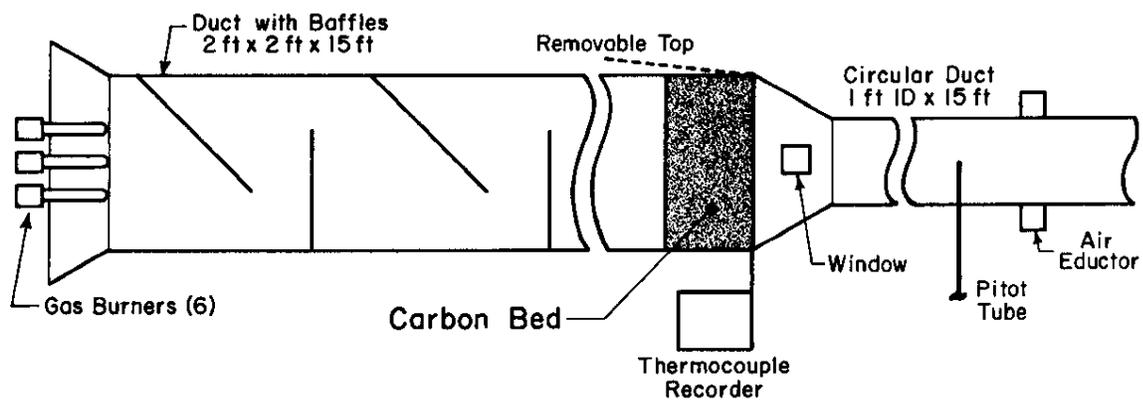


FIG. VI-6 FULL-SIZE IGNITION TEST EQUIPMENT

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APPENDIX A

SPECIFICATIONS FOR MOISTURE SEPARATORS FOR REACTOR CONFINEMENT FACILITIES

I. GENERAL

1.1 DRAWINGS

Fabrication of the moisture separators shall be in accordance with these specifications and drawing number W-234059, E. I. du Pont de Nemours & Co.

1.2 APPLICATION

1.2.1 Moisture separators are installed in casings that are erected in a process ventilation system to remove entrained liquid water particles from steam-air mixtures. Separator efficiency shall be adequate, about 99%, to prevent the blinding of a water repellent particulate filter (described in Appendix B) by 1 to 5 micron entrained water particles when exposed in series to the standard steam-air mixture test described in Section III.

II. CONSTRUCTION

2.1 SUPPORT MECHANISM AND CASING

2.1.1 General

The units shall be built in accordance with the highest standards of workmanship and finish and shall be free from obscure and obvious flaws.

2.1.2 Materials

2.1.2.1 Casing

Unless otherwise specified on drawing number W-234059, all material shall be type 304 stainless steel. The support mechanism must be capable of withstanding a ΔP of 12 in. H_2O across the separator.

APPENDIX A (Continued)

2.1.2.2 Separator Material

The separator material shall be fabricated from "Teflon"* yarn** and stainless steel wire woven into a mat. Separator material shall be composed of a number of layers, each approximately 24 x 24 inches. Minimum and maximum edge compression of the material when installed in the case shall be 3/8 and 1/2 inch, respectively, at each edge. The number of layers of material used in each nominal 2-inch-thick unit shall be determined by the number of layers necessary to meet the flow resistance requirement. Layers shall be held together by 16 gauge (approximate) stainless-steel wire for adequate support when separator is in vertical position for horizontal air flow.

2.1.2.3 Gaskets

Flange gasket on the upstream face of the separator case shall be 1/4 x 3/4 inch neoprene sponge (15-20 durometer). Gaskets shall have tight-butted joints at corners and shall be firmly and continuously cemented to flange with a neoprene-base adhesive. Gasket material shall maintain its integrity and resilient properties when subjected to air at 120°C (250°F).

2.1.2.4 Adhesive

The adhesive used for the gasketing shall be neoprene base and cured. It shall be resilient, water resistant, and resistant to a minimum temperature of 120°C for 8 hours when cured. If capable of ignition, it shall be self-extinguishing and shall meet all operating conditions without change in physical properties and without loss of seal or strength. Adhesive shall not show cracks, checks, "alligatoring", or separation.

* Registered trademark of E. I. du Pont de Nemours & Co.

** "Teflon" yarn or equal.

APPENDIX A (Continued)

III. TESTING AND INSPECTION

- 3.1 All units shall be inspected by purchaser's representative for quality of construction and freedom from obvious and obscure flaws. Particular attention will be directed to the flow resistance test, 3.2.
- 3.2 Each assembled separator shall exhibit a resistance to flow of 0.95 ± 0.05 inch H_2O , at an air flow of 1600 cfm at 22 to 25°C. Each unit shall be individually tested by the manufacturer.
- 3.3 In addition to the test described in Section 3.2, the manufacturer must conduct a steam-air mixture test (at his own expense) on one representative separator to qualify under this specification. The test consists of exposing a dust-loaded particulate filter (described in Appendix B) (with the moisture separator upstream) to wet steam flowing at a maximum rate of 8000 cfm for 30 seconds, following by 10 days continuous exposure to steam-air mixtures at flows up to 1000 cfm. Details of the equipment and procedures for conducting such a test are available on request.

IV. MARKING AND SHIPPING

- 4.1 Each unit shall have indelibly stamped on the installation case the following information:
 - a) Model number
 - b) Serial number
 - c) Resistance to flow
 - d) Flow rate and conditions
 - e) Material run number
- 4.2 Each unit shall be individually packed for shipment in substantial containers completely enclosing and protecting the units from physical damage during storage and shipping. They shall be palletized for convenience and economy in handling and shipping.

APPENDIX B

SPECIFICATIONS FOR WATER-REPELLENT PARTICULATE FILTERS FOR REACTOR CONFINEMENT FACILITIES

I. CONSTRUCTION

1.1 GENERAL

This specification is for high efficiency particulate filters with a high strength, water-repellent medium. The filters are used in confinement facilities for removing radioactive particles that might be released in the event of a reactor accident. Under accident conditions, the filters could be exposed to air at 120°C (250°F); wet steam at 100°C; or mixtures of steam, air, and entrained water particles.

Complete filter assembly shall be of fire-resistant construction for operating temperatures up to 120°C and shall pass all tests described in Section II.

In addition to tests described in Section II, the manufacturer must conduct a steam and steam-air mixture test (at his own expense) on one representative filter to qualify under this specification. The test consists of exposing a dust loaded filter (with a moisture separator upstream) to wet steam flowing at a maximum rate of 8000 cfm for 30 seconds, followed by 10 days continuous exposure to steam-air mixtures at flows up to 1000 cfm. Details of the equipment and procedures for conducting such a test are available on request.

The filter shall be constructed by pleating a continuous sheet of filter medium into closely spaced pleats separated by corrugated inserts. Pleated medium and separators shall be enclosed in a flanged, steel box and fastened at all boundaries with adhesive so that under test and rated conditions gas stream will pass through medium without bypassing.

Filter shall be assembled in accordance with highest workmanship standards to provide uniformity of materials and construction, finish, cleanliness, and freedom from obscure or obvious flaws.

APPENDIX B (Continued)

1.2 FRAME

Cadmium-plated 16-gauge steel sheet with double turned inlet and outlet flanges. Dimensions, exclusive of gaskets, shall be 24 by 24 by 11-1/2 inches plus or minus 1/16 inch, and square within 1/8 inch measured diagonally.

1.3 FILTER MEDIUM

Glass-asbestos fibers containing a minimum of 7% asbestos fibers, and with a minimum thickness of 18 mils. No substitution allowed.

Filter medium shall contain not more than 5% combustible matter and shall not contain holes, cracks, slits, or other physical imperfections. In completed filter unit the medium may not contain more than one splice. All splicing must be from same lot of filter medium. Splices must conform in all respects to paragraphs 3.3.2 of Military Standard MIL-F-51068A and 3.3 of MIL-F-51079.

Medium binder shall conform to Military Standards MIL-F-51079 and shall withstand a minimum temperature of 120°C for 8 hours.

Medium shall be treated with a maximum of 1% silicone-base water-repellent material to achieve wet strength characteristics sufficient to pass tests of Sections 2.2.5 and 2.3.

Patching of medium not allowed.

1.4 SEPARATORS

Full-hardened aluminum with 0.0015 inch minimum thickness.
Ref: Military Standard MIL-F-51068A.

1.5 ADHESIVE

Neoprene base, cured. Adhesive shall be resilient and water resistant and shall resist a minimum temperature of 120°C for 8 hours. If capable of ignition, it shall be self-extinguishing and shall meet all general operating conditions without change in physical properties and without loss of seal or strength. Adhesive shall not show cracks, checks, "alligating", or separation.

APPENDIX B (Continued)

1.6 GASKETS

1/4 inch thick by 3/4 inch wide, ozone-resistant neoprene, 15-20 durometer, in gasket pieces which shall be die-cut for dovetail mating. Mating surface of die-cut ends shall be coated with rubber-based adhesive and interlocked to present a workmanlike appearance. Gaskets shall be attached firmly and continuously to upstream flange surface of frame with adhesive and shall provide a continuous seal around filter face.

1.7 FACE GUARDS

Fabricated from galvanized hardware cloth, 23 gage 4 x 4 mesh, 1/4 inch opening, shall be installed on both faces of each filter. Face guards shall be fitted flat against aluminum separators but shall not contact filter medium.

II. TESTING

2.1 GENERAL

These tests control quality, reliability, uniformity, and performance characteristics of filters procured by this specification.

Purchaser's representative shall witness all factory tests. The vendor shall notify the purchaser when tests are to be run.

Each roll of medium is qualified by subjecting representative samples to tests described in Section 2.2. Acceptable rolls shall be identified with a code legend and cross-referenced to a document with the following information: (1) date and results of tensile tests, (2) serial numbers of filter assembled from roll of medium, (3) signatures of tester and witness, (4) medium manufacturer's name, lot and order numbers, and (5) any other pertinent background information describing history of medium. Rolls that do not pass tests shall be rejected.

A group of filters shall be assembled from each roll of medium that successfully passed tests; each filter shall be marked with appropriate code legend and serial number.

APPENDIX B (Continued)

A sample filter shall be selected randomly by purchaser's representative from each group. Sample filter shall be destructively tested in accordance with sequence of tests described in Section 2.3. Results of these tests shall be recorded on appropriate document, dated, and signed by tester and witness. This filter may then be discarded. Reject any group of filters whose samples fail any tests.

Each unit of remaining groups of filters qualified for production by passing sequence of tests, shall be subjected in turn to Flow Resistance Tests and Penetration Efficiency Test (DOP), both witnessed, described in Section 2.4. Reject any units that fail either test. Mark each filter as directed in Section 3.3 and pack for shipment in accordance with Section 3.2.

Test results shall be recorded on appropriate document adjacent to respective serial number of each filter, dated, and signed by tester and witness.

2.2 MEDIUM TENSILE STRENGTH

- 2.2.1 Samples of each roll of medium shall be prepared for each of the following witness tests. Each test result shall be the average of three samples selected across width of the medium at free end of roll.
- 2.2.2 Filter medium shall exhibit a minimum tensile strength of 2.5 pounds per inch of width when tested with tensile force perpendicular to normal direction of fibers in medium ("Across Machine Direction").
- 2.2.3 After a flat fold is made in sample perpendicular to normal direction of fibers in medium, sample shall be subjected to a tensile test parallel with normal direction of fibers in medium ("With Machine Direction"); minimum tensile strength shall be 1.5 pounds per inch of width.
- 2.2.4 Minimum elongation before rupture in both of above tests shall be 1.0%.
- 2.2.5 Soak samples of medium for 2 hours in room temperature water and repeat tests of Section 2.2.2 and 2.2.3. Wet medium shall exhibit minimum tensile strengths of 60% of dry medium readings.

APPENDIX B (Continued)

2.3 SEQUENCE TESTS ON RANDOM SAMPLE FILTERS

2.3.1 Moisture Resistance

One random sample filter assembled from each roll of filter medium shall be exposed to 2000 cfm of wet steam at 14.7 psia (measured upstream of the filter) and with water injected about 15 ft upstream of the filter, at a rate of 8 lb H₂O per minute.

Water shall be injected with nozzle No. 1/8G-6SQ or 1/8GG/6SQ, manufactured by Spraying Systems Company, 3201 Randolph St., Bellwood, Illinois. Only unentrained moisture shall be drained from test facility upstream of filter. No devices for removing entrained moisture or particulate matter from stream, such as roughing filters, separators, etc., shall be used in test facility upstream of filter.

Only one filter shall be tested in test facility at any one time. Filter shall be exposed to these conditions for 15 minutes and ΔP across filter obtained under these conditions shall not exceed 4 inches H₂O.

Filters that successfully pass the above test shall be dried with air between 20 and 25°C.

2.3.2 Flow Resistance

Dried samples from the test of Section 2.3.1 shall be individually tested for air-flow resistance in calibrated, standard DOP test apparatus without generation of DOP aerosol. Initial resistance of filters (when clean) shall not exceed 1.00 inch H₂O when passing rated air flow of 1000 cfm.

2.3.3 Dust Capacity

A dust capacity test shall be conducted on sample filters that passed moisture and flow resistance tests of Sections 2.3.1 and 2.3.2. Test shall be conducted using NBS Standard Dust Loading Test with NBS Cottrell Precipitate (without linters) while air is flowing through filter at rated capacity of 1000 cfm.

APPENDIX B (Continued)

Filter must hold a minimum of 4 lb dust per filter, before initial resistance (1.00 inch H₂O or less at 1000 cfm) doubles.

2.3.4 Ultimate Strength

Sample filters that passed dust capacity test shall be subjected to an Ultimate Strength Test with deposited dust still on filter. A sufficient air flow shall be passed through filter to increase and maintain for ten minutes a ΔP of 6 inches H₂O across filter.

2.3.5 Efficiency-Penetration (DOP)

Vendor must perform an encapsulated-Type dioctylphthalate (DOP) test to detect frame, gasket, or filter medium leakage.

Sample filters that passed Ultimate Strength Test, still carrying its load of dust, shall be tested according to Military Standard MIL-STD-282, May 28, 1956.

Penetration shall not exceed 0.1% for 0.3-micron diameter homogeneous particles of dioctylphthalate.

2.4 PRODUCTION TESTS

For final acceptance, each filter in the groups that are acceptable (because the sample from the group successfully qualified by passing sequence tests 2.3.1-2.3.5) shall pass the following two tests:

2.4.1 Flow Resistance Test

This test is identical to that described in Section 2.3.2.

2.4.2 Efficiency-Penetration Test (DOP)

This test is identical to that described in Section 2.3.5 except that DOP penetration shall not exceed 0.03%. Also filters shall be tested at 20% of rated flow as well as 100% of rated flow.

III. SHIPPING

3.1 PACKAGING

Each filter shall be packed for shipment in substantial hardboard or wooden containers, completely enclosed with extra protection for both open faces of the cell. Package shall support and completely protect filter whether handled in vertical or horizontal position. Container shall be designed so that it may be opened and cell removed without damage to container or filter cell, so that container may be reused for shipment to ultimate destination.

3.2 PALLETIZING

Containers shall be shipped as individual units without palletizing or fastening together in any multiple.

3.3 MARKING

Results of test penetration, test resistance, test flow rate, together with direction of test air flow, manufacturer's name, model number, serial number of the filter cell, and code legend of roll of medium used in production of cell shall be registered legibly and indelibly on the top of the frame of the filter unit. Serial number of unit shall be so registered by machine.

On top of frame opposite serial number stencil FOR REACTOR CONFINEMENT USE ONLY. Letters to be 3/4" high.

APPENDIX C

SPECIFICATIONS FOR ACTIVATED CARBON BEDS FOR REACTOR CONFINEMENT FACILITIES

I. GENERAL

1.1 DRAWINGS

Fabrication of the activated carbon beds shall be in accordance with these specifications and drawing number ST5-10545 (Figure C-1). Savannah River Plant, E. I. du Pont de Nemours & Co.

1.2 APPLICATION

- 1.2.1 Carbon beds are installed in casings that are erected in process ventilation exhaust systems. They are mounted with pleats horizontal on a vertical holding frame which is arranged with supporting and clamping devices so that 100% of the horizontal air stream flows through the beds.
- 1.2.2 The carbon beds are used in confinement facilities for radioactive iodine vapor that might be released in the event of a reactor accident. The units are to be constructed with airtight joints so that all of the air contacts the one-inch-thick carbon beds. Slight by-passing of the beds, permissible in conventional applications for odor control, is unacceptable for the subject carbon units.
- 1.2.3 The assembled carbon unit, when fully charged with activated carbon, shall pass 1000 scfm of air with a pressure drop of about 0.8 in. H₂O. The direction of flow shall be indelibly marked on one of the casing members that is permanently welded to the pleated section. This side will be designated TOP and so marked for shipping and installation.

APPENDIX C (Continued)

II. CONSTRUCTION

2.1 CASING

2.1.1 General

The units shall be built in accordance with the highest standards of workmanship and finish and be free from obscure and obvious flaws. Welds at flange and gasket surfaces shall be ground flush. Particular care shall be taken when installing gaskets that are required to prevent air from bypassing the carbon beds. Spacers shall not be installed within the carbon bed except as shown on drawing ST5-10545 (Figure C-1).

2.1.2 Airtight Seals

To prevent air from bypassing the carbon beds, airtight seals shall be provided at all edges of the carbon bed in contact with the enclosing casing as follows:

Where the two end legs of the pleated carbon bed join their respective casing surfaces, the bed walls shall not be perforated. They shall be lapped into a flange and spot welded to the casing surface before assembling gaskets and neoprene sealing pads.

The two side casing surfaces shall be furnished with internal neoprene pads that are cemented to the casing flanges with the specified adhesive. These pads shall bear jointly on the flanged edges of the pleated carbon bed walls and on the mating casing flanges such that when assembled and bolted, compression of the pads will form the required airtight seals.

2.1.3 Materials

2.1.3.1 Casing

The casing shall be fabricated of either type 304L stainless steel or cold-rolled carbon steel, whichever is specified on the purchase order. Stainless steel should be specified for use in areas where NO_2 or SO_2 are present, even in trace quantities.

APPENDIX C (Continued)

2.1.3.2 Gasketing Materials

Gasket material shall maintain its integrity and resilient properties when subjected to air at 120°C (250°F). The flange gasket shall be 1/4-inch thick by 3/4-inch wide, ozone-resistant neoprene sponge (15-20 durometer) of identical material specified below for the sealing pads. It shall be formed from die-cut gasket pieces for dovetail mating. Mating surfaces of die-cut ends shall be coated with the specified adhesive and interlocked to form a flawless joint. Gaskets shall be firmly and continuously cemented to the upstream flange surface with the specified adhesive.

The sealing pads, cemented to the 24 inch by 11-1/2 inch casing flanges, shall be fabricated of neoprene sponge 3/8-inch thick. They shall be completely cemented to the inner surfaces of their casing pieces with the specified adhesive.

2.1.3.3 Adhesive

The adhesive used for the gasketing and sealing pads shall be neoprene base, cured. It shall be resilient, water resistant, and resistant to a minimum temperature of 120°C for 8 hours when cured. If capable of ignition, it shall be self-extinguishing and shall meet all operating conditions without change in physical properties and without loss of seal or strength. Adhesive shall not show cracks, checks, "alligatoring", or separation.

2.1.3.4 Finish

This section is applicable only to carbon steel units. Dip all subassemblies in phenolic coating (Tithcoat LC24 or Hanna XB8081, or equal), air dry, and then subsequently dry parts in oven. All parts are then sprayed with at least two coats of moisture-resistant paint (Reliance 150-HB1-63 or Hanna XB-4357 Epon Black, or equal) and baked in oven until dried. (Exception: mask areas which are to be spot welded or arc welded in subsequent operations.)

APPENDIX C (Continued)

After final assembly of parts, clean and neutralize welds, brush-coat bare spots with phenolic coating, and then apply at least two additional coats of moisture resistant paint and bake in oven until dried.

2.2 ADSORBENT MEDIA

The adsorbent shall be new activated carbon made from coconut shells in accordance with the following:

2.2.1 Physical Properties

- a. Particle size*: 10-14 range, Tyler mesh size, as per following distribution:

<u>Mesh</u>	<u>Retention</u>
8	0.1% Maximum
10	10.0% Maximum
14	88.9% Minimum
Through 14	1.0% Maximum

- b. Packing Density: 34 lb/ft³ nominal, bone dry carbon based on apparent density measurement method (laboratory measurement, 32-36 lb/ft³ "free fall").
- c. Charge per unit: 57 lb (actual filled weight based on bone dry carbon) minimum weight to be adjusted for water content measured at time of filling units - one test at the beginning of the use of each batch and repeated at the beginning of each day when a batch is used that was held over from the work the preceding day.

*As per methods described in Military Specification, MIL-C-13724A, dated May 4, 1960.

APPENDIX C (Continued)

- d. Fines: After packing, carbon fines and dust shall be blown from the unit with compressed air at a minimum nozzle velocity of 5000 ft/min. Compressed air supply shall be free of oil, dust, and other contaminants. Nozzle orifice area shall be 0.8-in² minimum. Nozzle shall be fishtail-shaped and arranged so that the upstream screen surface of the entire pleated carbon bed, including the crevice at the apex of each pleat, can be blown with air. The direction of flow shall be the same as the installed air flow as marked on the top of the unit and specified in Section 1.2.3.
- e. Hardness*: 97% minimum.

2.2.2 Chemical Properties**

- a. Iodine number: Minimum of 1050 mg per gram of carbon.
- b. Activity for carbon tetrachloride: 50% by weight, minimum.
- c. Activity, Standard Accelerated Chloropicrin Test: 50 minutes minimum.
- d. Retentivity for carbon tetrachloride: 30% by weight, minimum.
- e. Ash content: 4% by weight, maximum.

2.2.3 Loading

The purpose of this section is to ensure that each carbon bed is packed firmly and with a uniform distribution of the carbon particles. The hopper used for loading the carbon unit shall provide a minimum of 3 ft of "free fall" for the carbon granules (excluding the height of the unit). There shall be at least three dispersal screens or baffles for scattering the falling granules in this 3 ft "free fall".

* As per methods described in Military Specification, MIL-C-13724A, dated May 4, 1960.

**For definition of tests, see Adsorption, C. L. Mantell, McGraw-Hill Book Co., New York, N. Y., 2nd Edition, 1951.

APPENDIX C (Continued)

The feed rate of the carbon through the hopper shall be as small as practically possible. Other loading methods are acceptable provided that the new method is proven to purchaser's satisfaction to provide equal or better distribution of the carbon particles throughout the bed.

During the initial filling operation, the unit shall be completely filled but not vibrated. The final step of the loading operation shall be vibration of the carbon unit on a packing machine built in accordance with MIL STD 282, Method 105.9. The packing machine shall operate at a frequency of 200 cycles/min with an amplitude of 3/4 inch and with the lifting cams operated in phase. The carbon unit shall be vibrated for 1.5 minutes while mounted rigidly on the packing machine platform. During the complete time of vibration, the unit must be kept full of carbon and in addition, the uppermost layers of carbon must be subjected to a packing force equivalent to a minimum height of 6 inches of carbon that is maintained above the top carbon layers in the unit. As an alternative, the packing force may be provided by a spring loaded device (with a force equivalent to 6 inches of carbon) to force carbon into the unit while it is vibrated. As stated in Section 4.1, this operation shall be witnessed by purchaser's representative.

III. TESTING

- 3.1 Physical and chemical tests specified in Sections 2.2.1 and 2.2.2 need not be witnessed by purchaser's representative. Tests confirming the specified properties shall be performed by the vendor on a representative sample from each 2000-lb carbon lot, and the certified results submitted to purchaser.
- 3.2 The carbon must have a high efficiency for the removal of iodine. The units are used in a confinement facility for removing radioactive iodine that might be released during a reactor incident. Under incident conditions, the carbon could be exposed to air at 120°C, wet steam at 100°C, or mixtures of steam and air. In addition to the tests specified in Sections 2.2.1 and 2.2.2, the vendor shall submit a 5 lb

APPENDIX C (Continued)

representative sample of carbon to the purchaser for testing of iodine removal efficiency and ignition temperature. This carbon shall be certified as being representative of that carbon which the vendor will use in the carbon beds. The carbon shall remove 99.99+% of the iodine when exposed to a steam-air mixture at 70°C and flowing at a face velocity of 70 ft/min and with an iodine loading of 1 mg per gram of carbon.

The carbon must have an ignition temperature greater than 330°C (625°F) under the following conditions:

Air velocity: 70 ft/min
Rate of heating: 2-4°C/min
Bed thickness: 1 inch
Bed diameter: 3-1/2 inches with 1/4-inch-wide baffle on front and back sides
Apparatus: Stainless steel construction

- 3.3 All units shall be tested for leaks with "Freon"*, or equal, by the purchaser prior to installation in the confinement system. Testing shall be in accordance with that specified in AEC Research and Development Report, DP-870, "Nondestructive Test of Carbon Beds for Reactor Containment Application, Progress Report, June 1962 - December 1963". Those units which are not acceptable (leaks greater than 0.1% of total bed flow) shall be returned to the vendor at the vendor's expense, for repair.

IV. SHIPPING

Each carbon unit shall be packed for shipment in substantial containers completely enclosing and protecting the units from physical damage during storage and shipping. They shall be palletized for convenience and economy in handling and shipping, and shall be stored and shipped with the TOP side up, as defined in Section 1.2.3. The carbon weight, carbon lot number, and bed serial number shall be indelibly marked on the casing.

4.1 INSPECTION

All units shall be inspected by purchaser's representative for quality construction and freedom from obvious and obscure flaws, before, during, and after charging. Particular attention will be directed to construction details affecting quality of airtight seals and the loading operation.

* Du Pont's registered trademark for its fluorinated hydrocarbons.

FIG. C-1 ASSEMBLY AND SECTIONS OF SRP ACTIVATED CARBON BED FRAME

