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BEHAVIOR OF IODINE IN REACTOR CONTAINMENT SYSTEMS

G. W. Keilholtz and C. J. Barton
Reactor Chemistry Division

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G. W. Keilholtz and C. J. Barton
Reactor Chemistry Division

FEBRUARY 1965

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FOREWORD

This is the fourth report prepared under the auspices of the Nuclear Safety Information Center, which was established in March 1963 at the Oak Ridge National Laboratory under the sponsorship of the U.S. Atomic Energy Commission. The Center serves as a focal point for the collection, storage, evaluation, and dissemination of information in the following areas:

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The present report was prepared at the request of Merson Booth of the Analysis and Evaluation Branch of the AEC Division of Reactor Development. The subject is of particular interest, both because of the extensive research and development in the subject area for a number of years in this country and elsewhere – particularly in the United Kingdom – as well as its potential use in reactor safety evaluations. While this active interest makes the report quite timely, it should be recognized that it is, in a sense, a progress report, because new and pertinent information on the subject is being generated daily. Nevertheless, the authors have attempted to condense, to summarize, and to some extent, to evaluate the present state of knowledge on the behavior of fission-product iodine when it is released into a containment system as a result of a reactor accident. It has not been possible to use or even to refer to all published work on iodine behavior, because of the large mass of the available information. The authors furthermore recognize some bias toward the use of data and references originating in the research and development program at ORNL, but they have tried not to neglect the more pertinent information from other installations.

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Behavior of Iodine in Reactor Containment Systems

G. W. Keilholtz C. J. Barton

ABSTRACT

The properties of radioactive iodine are described through the stages of generation, release, transport, and deposition in a reactor containment system. Trapping systems for different forms of airborne iodine are discussed, and methods of evaluating their efficiency are described. The adsorption of iodine on particulate matter, the rate of deposition of iodine under various possible reactor accident conditions, and the formation of organic iodine compounds are aspects of iodine behavior now considered important in evaluating the hazards of nuclear reactor accidents and in establishing reactor site criteria.

Methods of characterizing the forms of iodine and of inducing their removal from aerosols are compared and evaluated. Information is available on the behavior of iodine in a variety of environments, but few experiments have been performed that possessed all the elements of realism necessary for simulation of conditions likely to exist in loss-of-coolant accidents in water-cooled and/or -moderated reactors. Experiments to be performed in existing and projected facilities described in this report should eventually meet the need for this type of data.

The lack of certainty in regard to the chemical form in which iodine will exist in reactor accident environments makes it inadvisable at present to recommend a change in the factor-of-2 credit presently assumed for natural deposition phenomena. There is need for greater emphasis on large-scale testing of engineered safeguards under the most realistic accident conditions that can be devised in order to define their efficiency and to provide more confidence in their reliability. A broad program of laboratory investigations is also needed to aid in designing large-scale tests and in interpretations of the results.

1. INTRODUCTION

Extensive investigations of the properties of radioactive iodine have been reported in the literature during the last few years. Experience resulting from the Windscale Reactor accident was a major demonstration of the importance of anticipating the behavior of iodine under accident conditions. This has been discussed by Beattie (*1*). There have been both earlier and later demonstrations, though less spectacular. It is the purpose of this report to direct attention to the principal investigations and to outline the properties of radioactive iodine through the stages of generation, release, transport, and deposition or retention in a reactor containment system, and to consider the removal of iodine from containment systems. The adsorption of iodine on particulate matter, the behavior of aerosols, and the formation of organic compounds are processes now considered important in evaluating nuclear safety procedures.

The control of radioiodine is a problem of major importance in the design and siting of nuclear reactors, in which it is produced in high yield. Radioiodine is volatile and can be released from nuclear fuel under certain conditions. The released

iodine can exist in reactor containment systems in various forms, and it is difficult to predict the quantities of the different forms that may be present during reactor accidents. Because it concentrates in the thyroid gland, radioiodine has been assigned (*2*) a very low maximum permissible concentration, 3×10^{-9} μc of ^{131}I per cubic centimeter of air for a 168-hr/week exposure and a maximum permissible body burden of 0.7 μc. Iodine is, therefore, the most important single fission product to be considered in establishing reactor site and containment criteria.

Criteria for the evaluation of reactor sites in the United States were formally established in 1962 with the approval of the Reactor Site Criteria (*3*). This document was supplemented by another (*4*) which illustrated the methods to be used in the calculation of the acceptability of a specific site for a specific reactor. Such calculations involved the determination of the amount of activity that could be released from a pressurized light-water-cooled and -moderated (PWR) reactor system in the event of a loss-of-coolant accident and the subsequent exposure of surrounding persons. This accident is currently considered to be the maximum credible accident (MCA) for PWR systems (*4*). The MCA is defined (*4*) as that for which the "potential hazard would not be exceeded by any other accident considered credible during the lifetime of the facility."

Fission-product release will vary considerably with the reactor system and the nature of the accident. In addition, there are many possible combinations of physical and chemical forms of the radioisotopes released into the containment vessel, and a number of transport phenomena, such as agglomeration, absorption, adsorption, deposition, and steam condensation within the containment structure, which reduce the amounts of iodine available for release to the environment. Therefore, in order to provide a starting point for calculating the potential radiological hazard to the environment, it is now generally assumed (*4*) that the fission-product release into the containment structure includes 1% of the solids, 100% of the noble gases, and 50% of the iodine, and that half of the released iodine will be adsorbed onto internal surfaces of the structure and will not be available to leak into the atmosphere. Applicants for reactor operating licenses are allowed to use their own judgment concerning the applicability of these figures to their postulated accidents.

It is difficult to prove that the various transport phenomena would reduce release from containment under all circumstances. Therefore, the credit that is presently allowed for such natural phenomena in evaluating the exposure which might result from the maximum credible reactor accident is possibly quite conservative.

Engineered safeguards, such as internal filtering systems, may be accredited under certain stringent conditions. The general conditions (*5*) that such a system must meet are as follows. Systems and equipment must be tested periodically throughout the life of the plant under conditions relevant to accident conditions to ensure their operation when called upon, and they must meet performance specifications under these conditions. Each safeguards system must apply to the entire reactor system, must be protected from the accidents which give rise to its use, and must operate for the duration of any credible accident.

Allowance has been given by the USAEC for the demonstrated capability of certain engineered safeguards. This subject is discussed in another NSIC report (*6*).

2. PHYSICAL AND CHEMICAL PROPERTIES OF IODINE AND IODINE COMPOUNDS

2.1 PHYSICAL PROPERTIES OF ELEMENTAL IODINE

Iodine is a black solid with a slight metallic luster. Gaseous iodine dissociates more readily than the other halogens. Free energy values for the dissociation are given in Table 2.1. Under 1 atm of pressure, dissociation is said (*7*) to become perceptible at 600°C, 5.2% is dissociated at 800°C, 19.7% at 1000°C, and 74.8% at 1400°C.

Table 2.1. Calculated Free Energy Values for the Reaction $\frac{1}{2}I_2 \rightarrow I$ (*7*)

Temperature (°K)	ΔF (kcal/mole)
298	+14.44
500	+11.98
1000	+5.74
1500	−0.62
2000	−7.10

Table 2.2. Isotopes of Iodine Produced in Reactors (*1*)

Isotope	Half-Life	Fission Yield[a] (%)	Activity per Unit Thermal Power (kilocuries/Mw)	
			At Shutdown	1 Day After Shutdown
^{131}I	8d	3.1	25	23
^{132}I	2.3h	4.7[b]	38[c]	0
^{133}I	21h	6.9	54	25
^{134}I	52m	7.8	63	0
^{135}I	6.7h	6.1	55	4.4
^{136}I	86s	3.1	53	0

[a]Fission yield values from Katcoff (*9*) for thermal neutron fission of ^{235}U.

[b]Yield of ^{132}Te.

[c]Amount of ^{132}I generated in the reactor by decay of ^{132}Te and yield of this isotope.

Iodine melts at 113.6°C and boils at 184.4°. The vapor pressure of the solid is represented (*8*) by the equation

$$\log P_{atm} = -3512.8/T + 2.013 \log T + 13.3740 .$$

Natural iodine (^{127}I) has an absorption cross section of 6.3 barns. The principal fission-product isotopes of iodine are listed in Table 2.2. All are beta emitters.

2.2 CHEMICAL PROPERTIES OF IODINE

Information in this section is drawn mainly from reference books on nonradioactive iodine or on the halogen family (*7–8, 10*). Certain aspects of iodine chemistry that are more or less unique to the field of fission-product behavior, mainly because of the radioactivity and the small mass amounts of material involved, are treated in the following sections.

Iodine, along with the other members of the halogen family, is placed in Group VIIA of the Mendeleev periodic chart. Its outermost electron shell has seven electrons, but its *N* shell has only 18 electrons out of the maximum of 32. It has the lowest electronegativity among the naturally occurring halogens, and it exhibits valence states of −1, +1, +3, +4, +5, and +7 in various compounds.

2.2.1 Molecular Iodine, I_2

The chemical activity of iodine is less than that of the other halogens. There is no appreciable reaction with either hydrogen or oxygen at ordinary temperatures. Its reaction with metals is discussed in Sect. 2.2.2. The standard redox potential (E_0, volts) for the reaction

$$I_2 + 2e^- \rightarrow 2I^-$$

is 0.5345, as compared with 1.087 for Br_2, 1.3583 for Cl_2, and 2.85 for F_2.

When I_2 is dissolved in water (0.029 g per 100 g of H_2O at 20°C) slight hydrolysis occurs according to the equation

$$I_2 + H_2O \rightarrow HIO + H^+ + I^- ;$$

the hydrolysis constant for this equation has been calculated to be 4.6×10^{-13} at 25°C.

Iodine oxidizes thiosulfate ion, $S_2O_3^{2-}$, to $S_4O_6^{2-}$ in a bicarbonate-buffered solution or to SO_4^{2-} in a strongly alkaline solution. These reactions are the basis for proposals to add thiosulfate to dousing solutions.

2.2.2 Metal Iodides

Fission-product iodine released in reactor accidents is likely to come into contact with metal surfaces; consequently, the stability of metal iodides is of interest. This subject has been discussed at some length by Rolsten (*8*). The following elements form sufficiently volatile iodides to be amenable to production in a pure form by the iodine process: Y, Ti, Zr, Hf, Th, V, Ni, U, Nb, Ta, Cr, W, Cu, Ag, Fe, B, Ge, Pa, and Si. It will be noted that the three principal components of stainless steel, iron, nickel, and chromium, are included in this list. This is important because stainless steel is a common material of construction in nuclear reactors, especially of primary reactor vessels in pressurized water reactors. The standard heats of formation of anhydrous iron, chromium, and nickel iodides are −30.0, −54.2, and −20.5 kcal/mole, respectively.

Earlier work on reactions of iodine with metal surfaces is summarized by Mellor (*10*). This subject is being re-examined at the Oak Ridge National Laboratory by use of tracer techniques, with special emphasis on interaction of gaseous iodine with stainless steel surfaces. Preliminary results of this continuing investigation have been reported (*11–13*). Iodine vapor densities varied from ~0.001 to 1000 mg/m^3, and the temperature of the metal surface (types 302, 304, and 321 stainless steel) varied from 24 to 700°C. No difference in behavior toward iodine vapor was exhibited by the three different types of steel employed. It appears that the reaction probably occurs at points on the steel surface where imperfections exist in the thin oxide coating that normally covers such surfaces. The resulting iodide product deliquesces in the presence of moist air and will eventually lose iodine with the production of oxide. Surface coverages of up to 60 monolayers (a monolayer is approximately 0.3 μg of I_2 per cm^2) were observed, but large variation in the saturation coverage was also noted. Results of exploratory experiments (*11–13*) are summarized in Table 2.3. The information in this table indicates that iodine reacts with stainless steel surfaces over the temperature range 25 to 700°C and that two components exist in the adsorbed phase, one more readily removable than the other. Data (*14*) on the kinetics of iodine sorption by stainless steel are summarized in Table 2.4, and adsorption by preoxidized steel is compared with that by as-received stainless in Table 2.5. The latter data show that the oxidized steel adsorbed only 3% as much as the as-received specimens with a 1000-fold increase in I_2 concentration. Reactions occurred in an essentially oxygen-free atmosphere in this work, and application of the results to accident situations where oxygen and steam will probably be present is not clear. Reactions of iodine with metal surfaces in the presence of oxygen and steam as well as reactions of iodides on surfaces with oxidizing atmospheres will be of more direct interest for hazards analyses.

2.2.3 Hydriodic Acid, HI

Hydrolysis of molecular iodine, mentioned in Sect. 2.2.1 above, results in production of HI and HIO. Properties of hydrogen iodide are considered briefly in this section. It is a strong reducing agent and is extremely soluble in water. Solid HI melts at −50.8°C, and the liquid boils at −35.4°C. A maximum boiling mixture of aqueous HI contains 53% HI at 1 atm and 127°C. Thermal decomposition according to the equation $2HI \rightarrow H_2 + I_2$ is represented (*7*) by the formula

$$\log K_{HI} = -10{,}030/T + 0.5 \log T + 13.001 \ .$$

It is a strong acid but not as strong as HBr or HCl. It reacts with active metals, oxides, hydroxides, and carbonates to form iodides.

2.2.4 Oxy Compounds of Iodine

Hypoiodous acid, HIO, is formed by hydrolysis of I_2 as indicated in Sect. 2.2.1, and it is also formed when iodine is dissolved in cold dilute alkali solutions. On standing or on heating, the hypoiodite ion decomposes to give the more stable iodate ion.

The amphoteric nature of HIO is represented by the equation: $HIO \rightleftharpoons H^+ + OI^- \rightleftharpoons OH^- + I^+$.

Table 2.3. Experiments Conducted on Iodine Adsorption (*11–12*)

Type of Experiment	I_2 Vapor Densities (mg/m^3)	Adsorbent Temperature (^oC)	Principal Findings
Closed loop; helium carrier gas; in situ gamma-ray detection of ^{131}I tracer on type 304 stainless steel	$\sim 10^{-3}$ and 10^2	24–700	Iodine initially reacted irreversibly with SS surfaces and was not further transported, even during 3 weeks' operation.
Metal coupons in evacuated glass apparatus; intermittent removal of coupons and chemical analysis for iodine in leached deposit	$\sim 10^{-3}$ to 10^3	24–500	Water-soluble iodides formed on SS surface up to approximately 60 monolayers. Introduction of air seemed to cause loss of iodine from the surface deposit. The amount of deposit of iodine on the metals varies in the order Cu > Fe > Ni.
Metal coupons in evacuated glass apparatus; in situ gamma-ray detection of ^{131}I tracer on surface	$\sim 10^{-3}$ to 10^2	24–260	Activity sorbed on the glass surface masked activity on the SS at low vapor concentrations. Two components in the adsorbed phase, one readily reversible and one rather irreversible, are observed at higher pressures of I_2. Reversible fraction decreased with time.
Type 304 stainless steel tube coupled to glass vacuum-source system; in situ gamma-ray detection of ^{131}I tracer on SS surface	$\sim 10^{-3}$ to 10^2	300–500	Sorption of only a few monolayers, reversible to some extent, leaving SS surface passive toward readsorption. Eventually passivity was overcome at higher iodine pressures. A second test section was passive toward iodine adsorption from the start of the experiment. Investigation on this sample is in progress. Mass spectrographic analysis of source indicates presence of nonelemental forms of iodine.
Temperature gradient imposed along type 321 stainless steel tube coupled to glass vacuum-source system; in situ gamma-ray detection of ^{131}I tracer on SS surface; detector on moving platform so as to traverse the temperature gradient region of the tube	$\sim 10^{-3}$ and 1	24–380	Maximum adsorption at 150°C; no sorption in hot zone even on cooling (passivation had apparently taken place). On changing the temperature profile along tube, material moved so as to maintain the maximum in the 150°C region of the tube.

Table 2.4. Summary of Experimental Information on Kinetics of Iodine Sorption by Stainless Steel

Surface Temperature (°C)	Iodine Concentration Above Surface (mg/m^3)	Rate Coefficient, a ($monolayers^{-1}$)	Time Range (min)	Number of Monolayers
70	0.006	40	10–100	0.06
		9	100–400	0.26
70	0.166	2.7	100–1000	2.1
180	0.126	0.7	90–400	3.0
180[a]	0.99	9	1–100	0.09
500	0.00270	1.5	10–100	2.3

[a]Surface pretreated to have an oxide layer 200–300 A thick.

Table 2.5. Effect of Stainless Steel Surface Oxidation on Sorption of Iodine Vapor at a Surface Temperature of 150°C

	As-Received Stainless	Preoxidized Stainless
Surface oxide characteristics	Amorphous, undeveloped, probably thin	200–300 A, crystalline
Iodine pressure (mm Hg)	1.4×10^{-5}	1.1×10^{-2}
Time exposed (hr)	8.1	12
Monolayers sorbed	4.5	0.14

Hypoiodous acid is a very weak acid, as shown by its ionization constant, 2×10^{-10}.

Iodic acid, HIO_3, with a heat of formation of 56 kcal/mole, is more stable than either $HClO_3$ (24 kcal) or $HBrO_3$ (12.5 kcal). Iodic acid solutions can be concentrated until crystals precipitate.

2.2.5 Methyl Iodide

A number of organic compounds are apparently formed when radioactive iodine is released in the atmosphere of containment vessels and when molecular iodine is generated in laboratory tests (see Sect. 3.6). Methyl iodide forms the major portion of the group of aliphatic iodides that have been identified, and it is the only one of the group that is considered here.

Methyl iodide (iodomethane) boils at 42.5°C (760 mm) and freezes at –66°. It is available commercially tagged with ^{131}I and possibly can be obtained with other radioiodine isotopes, so it is unlikely that many investigators will make their own tagged methyl iodide. However, several methods for this synthesis are discussed in the literature. Since ^{131}I is most readily available in the form of $Na^{131}I$, possibly the most convenient method is exchange of this compound with $CH_3{}^{127}I$ in ethanol (*15*).

Methyl iodide is slightly soluble in water (1.4 g per 100 g of water at 20°C). The standard free energy of formation of the compound from its elements (ΔF_{f0}) at 25°C is listed (*16*) as –5.3 kcal/mole.

Gover and Willard (*17*) have studied the effect of short-wavelength light (1849 A) on the reaction of hydrocarbons with iodine, but similar studies of the effect of radiation on such reactions have not come to the attention of the authors.

3. GENERATION AND CHARACTERIZATION OF FISSION-PRODUCT IODINE AND OF PARTICLES

The characterization of fission-product iodine from a fuel melt is difficult because of the wide ranges of temperatures, fuels, contaminants, and environments which may exist in a particular accident situation. The physical and chemical forms of the released iodine may change significantly as the result of a change in but one of the many parameters involved. Nevertheless, an understanding of transport and removal phenomena is needed in order to predict the extent of iodine release in a variety of environments as well as the conditions under which it would condense, become adsorbed on particles, react, or remain in the vapor state.

All experimental work relative to fission-product release and transport involves at least some means of releasing either real or simulated fission products and some means of sampling for determining the extent of fission-product release. These minimal requirements are generally supplemented by sufficient sampling to determine an inventory balance on the released fission products in the experimental facility in question. Of increasing recent interest to investigators are sampling techniques which will permit characterization of the released fission products. Completed and proposed facilities for such investigations range from laboratory-sized mockups to very large experimental facilities involving containment systems tests and the meltdown of actual reactor cores. Most of the information in this section concerns the facilities and techniques involved in studies of iodine and other fission products, but a limited amount of experimental data is included in it.

3.1 MAJOR FACILITIES FOR RELEASE AND TRANSPORT STUDIES (*18*)

Most of the work on fission-product release and transport to date has been performed with small-scale laboratory facilities. Such facilities are suited for exploring a wide range of experimental parameters in order to identify the important parameters and to obtain release and transport data under simulated accident conditions. Since it is virtually impossible to simulate all accident conditions, such as heating, materials, environment, and geometry, extrapolations of the results of laboratory experiments are subject to some question when used to predict the results expected of a meltdown

Table 3.1. Major Facilities for Release and Transport Investigations (*18*)

Designation	Installation	Principal Investigator	Contained Volume (ft^3)	Surface-to-Volume Ratio (ft^{-1})	Status	References
Containment Mockup Facility	ORNL	G. W. Parker	6.3	3.4	Operational	*19*
Nuclear Safety Pilot Plant (NSPP)	ORNL	W. B. Cottrell	1,350	0.43	Operational	*20*
Containment Systems Experiment (CSE)	Hanford	E. R. Irish	32,700	0.17	Under construction	*21*
Loss-of-Fluid Test (LOFT)	Phillips	T. R. Wilson	282,000	0.07	Under construction	*22*
Power Burst Facility	Phillips	T. R. Wilson	125,000[a]	0.125[a]	Being designed	*23*

[a]Based upon preliminary design of reactor bay.

Table 3.2. Comparison of Fuel Meltdown Techniques (*18*)

Meltdown Technique	Installation, Designation, and Principal Investigator	Materials Melted	Comments	References
Arc image	ORNL, G. W. Parker	UO_2	Specimen size limited	*24*
Induction heating	ORNL, G. W. Parker Hanford, R. K. Hilliard MSA, S. J. Rodgers BNL, A. W. Castleman, Jr.	UO_2	Requires surrounding conducting material to heat UO_2 up to temperatures at which it becomes conductive	*24, 25* *26* *27* *28*
Resistance heating	ORNL, G. W. Parker Hanford, R. K. Hilliard	U metal, UO_2	When specimen resistance heated, circuit broken at onset of melting; otherwise, temperature limited by furnace materials	*24, 29* *30*
Plasma heating	ORNL, W. B. Cottrell GE-ANPD, P. K. Conn	UO_2 BeO	Has higher temperature capability than any of the above techniques	*31* *32*
In-pile test	ORNL, ORR, W. E. Browning, Jr. ORNL, TREAT, R. A. Lorenz Phillips, PBF	UO_2 UO_2	Nuclear heating but expensive	*33* *34* *23*
Reactor meltdown	Phillips, LOFT, T. R. Wilson Phillips, SPERT, F. Schroeder		Most realistic and most expensive	*22, 35* *36*

or activity release in an actual system. Furthermore, in laboratory-scale systems it is difficult to evaluate the effects of transport phenomena and of countermeasures which might exist or be applied in reactor containment systems. Accordingly, the USAEC has embarked upon an experimental program involving the construction of a variety of experimental facilities which will permit the conduct of release and transport investigations under simulated reactor accident conditions and which will provide the basis for the extrapolation of data already available from laboratory tests. The significant facilities which constitute a part of this program are listed in Table 3.1. It should be noted that programs for large-scale facilities and the laboratory programs are complementary. The former are needed for the reasons indicated above; however, they will never replace the latter, because of the cost and time involved in each test. A more complete listing of past and present programs on fission-product release and transport problems is given in Table 3.2.

3.2 GENERATION OF RADIOACTIVE IODINE

3.2.1 Fuel Meltdown Facilities

Much work in fission-product transport involves the production of selected elements or, if radioactive, selected isotopes as a product of a chemical reaction or from decomposition of a compound upon heating. Thus iodine is frequently generated by the reaction of sodium iodide with molten potassium dichromate or by the decomposition of palladium iodide upon heating, as described in Sect. 3.2.2. Such work is inevitably open to the criticism that the chemical and physical state of the volatilized material and, therefore, its subsequent behavior may be different from what it would have been had the release occurred from the fuel material under realistic environmental conditions. Thus we are concerned here principally with those release techniques which are capable of causing melting or near melting of fuel materials. Several heating

methods, especially induction and resistance, can be used for prolonged periods at temperatures below fuel melting points. All of the more common techniques are listed in Table 3.2 together with the principal investigators who have employed each technique and some comment as to the advantages and disadvantages of each. Facilities used at the Oak Ridge National Laboratory are described in the following sections.

3.2.1.1 Arc-Image Furnace. – The first technique employed to melt irradiated UO_2 for fission-product release determinations involved the use of an arc-image furnace. The equipment used is shown in Fig. 3.1 (*24*). It is possible to obtain very high temperatures by use of this equipment but it was found to have several limitations, such as the need for a BeO holder, very small specimen size, small size of high-intensity beam (~1 cm diam), and small depth of focus of the beam. Consequently, other heating methods such as induction, resistance, and plasma are presently considered preferable to the arc-image furnace.

3.2.1.2 Tungsten-Crucible Method for Induction Melting of UO_2. – A number of advantages were found (*37*) to be associated with the use of tungsten-foil crucibles which were readily heated by rf induction to the melting temperature of UO_2. Particular interest is attached to the fact that a scale-up factor of 1000 was readily attained over the maximum size in the arc-image-furnace method which was previously used for the melting of UO_2 and had been found to be limited to samples of less than 50 mg of highly irradiated UO_2. Tungsten crucibles, shown in Fig. 3.2, made from 5-mil tungsten foil may be used to melt 50 g of UO_2 of any burnup level obtainable. The use of tungsten at high

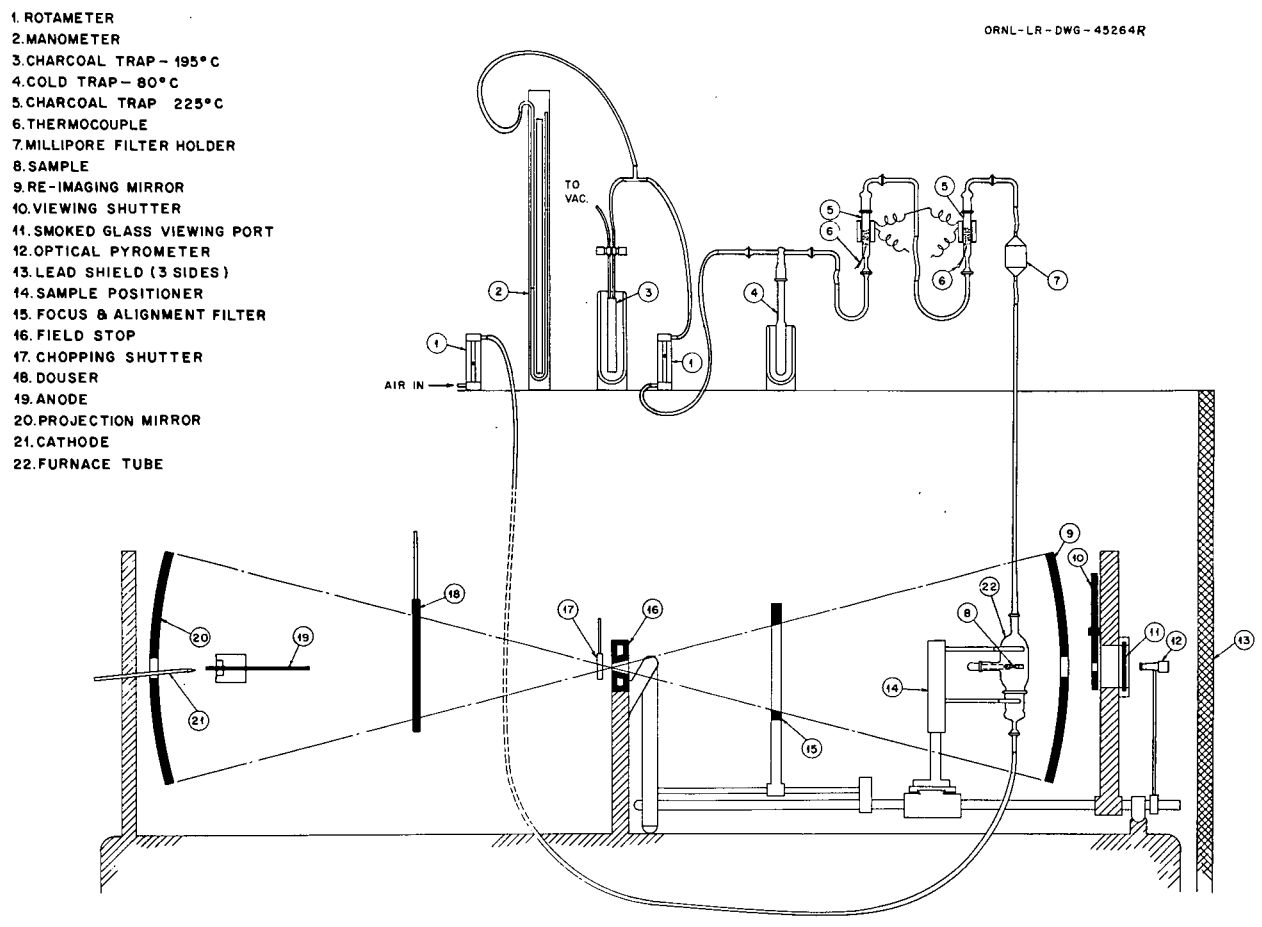

Fig. 3.1. Schematic Representation of Arc-Image Furnace and Collection Train for Fission-Product Release Experiments.

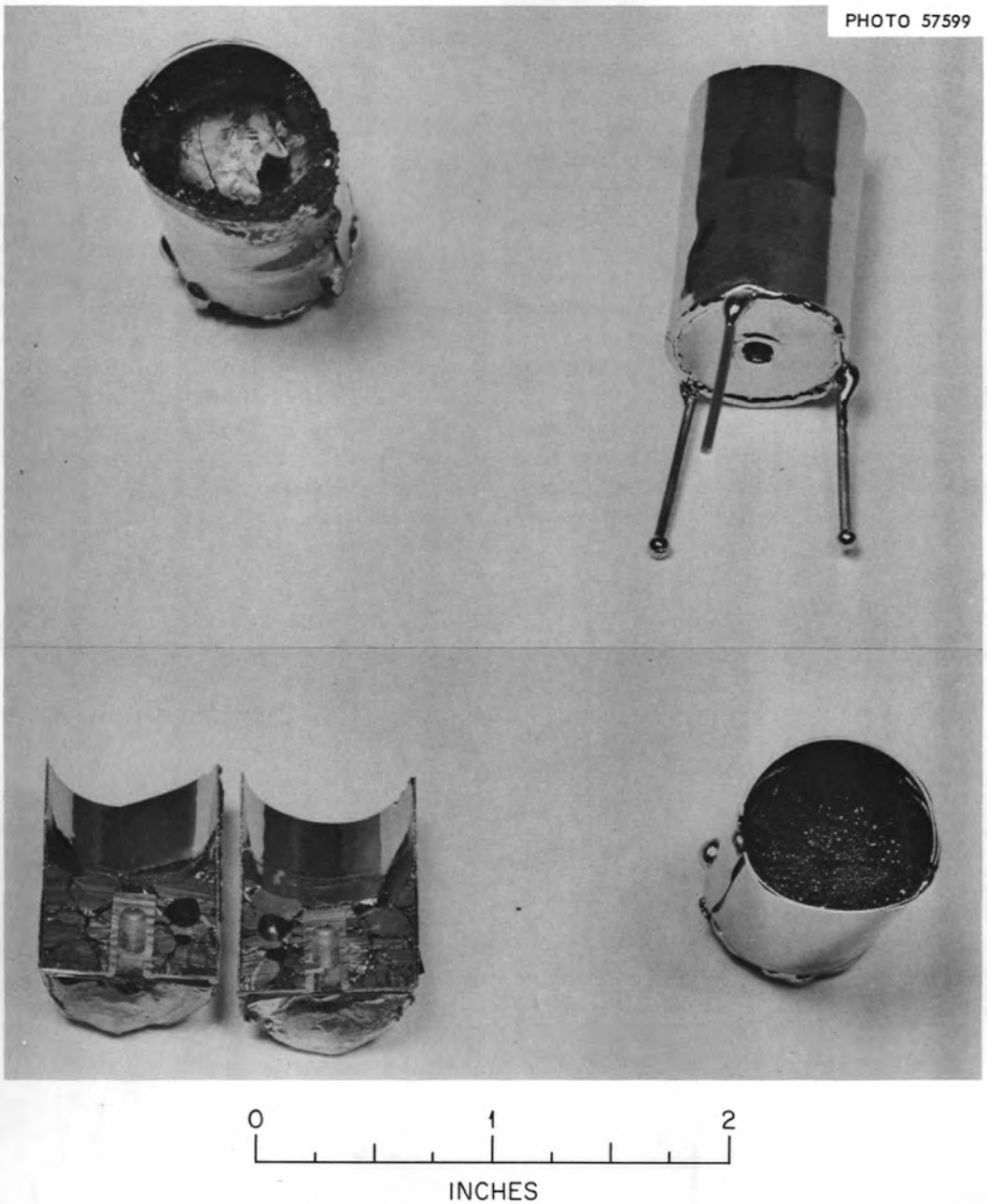

Fig. 3.2. Tungsten Crucibles Showing Varied Appearance of Induction-Melted UO_2. At top left, a melt with smooth top surface heated slightly above melting; at bottom right, a sample with crystal growth on the surface heated slightly below melting. Thermowells for tungsten-rhenium couples are shown at bottom left.

temperatures is limited to noncorrosive atmospheres. Figure 3.3 is a photograph of the furnace and the small fallout chamber used with it.

3.2.1.3 Hot-Cell Confinement Mockup Facility. – The hot-cell confinement mockup facility (CMF) shown in Fig. 3.4 has been used (*19*, *25*) to melt irradiated UO_2 specimens clad with stainless steel. A 25-kw, 5-Mc rf power source is employed; as indicated in the photograph, Fig. 3.5, it was necessary to place part of the equipment within the hot cell, because it did not appear to be practical to transmit the needed power the required distance

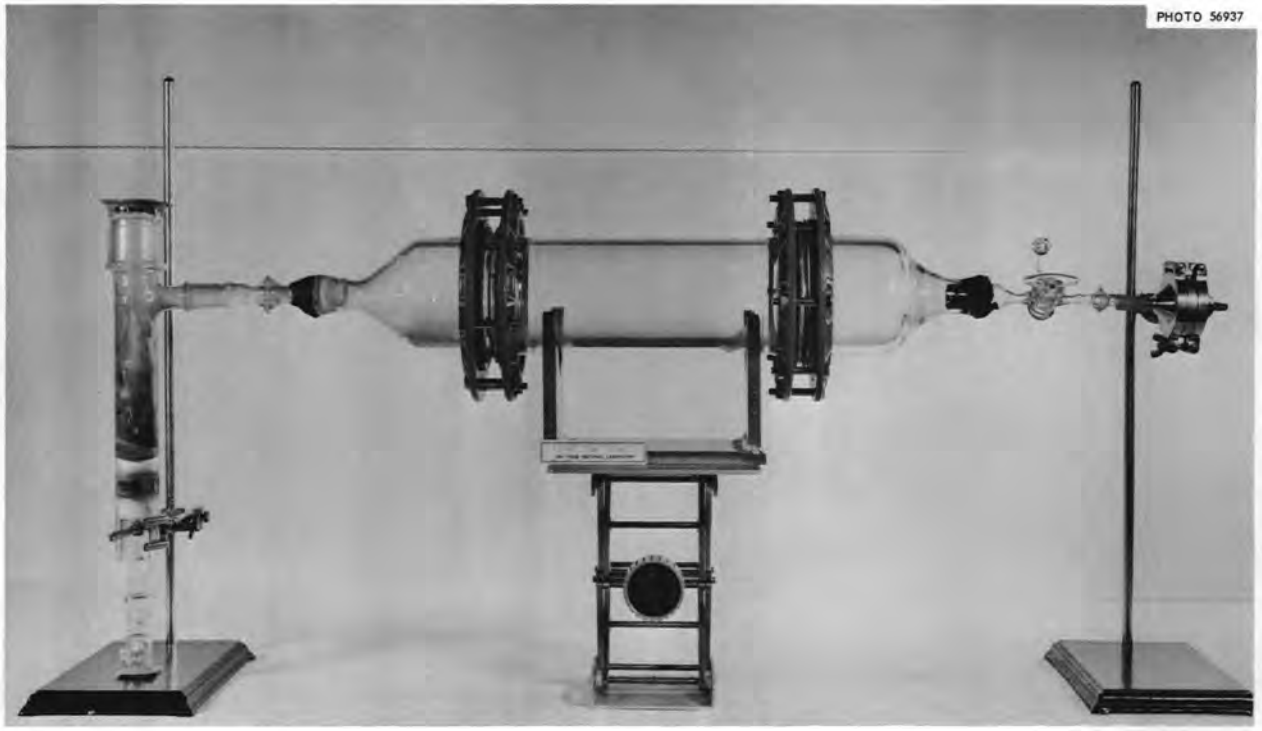

Fig. 3.3. Induction-Furnace Tube and Miniature Fallout Chamber Used with Tungsten-Crucible Method for Melting UO_2.

for out-of-cell locations. Melting of the specimen is accomplished by coupling first to the stainless steel cladding and then to the UO_2 specimen itself after the cladding melts (UO_2 becomes a conducting material at high temperatures). The specimen is contained in a zirconia crucible, and the duration of melting is limited by the time required for the molten UO_2 to break through the bottom of the crucible ($\sim$1 min). A quartz "chimney" carries released fission products through the large valve at the bottom of the confinement vessel.

3.2.1.4 Centered Tungsten Resistor. – Center melting has been one of the most difficult fuel conditions to simulate in a non-nuclear heating experiment. A simple mockup of these conditions was produced (*29*) by the construction of the resistance-heated assembly shown in Fig. 3.6. A metal-clad capsule containing cored PWR UO_2 fuel pellets, through which a tungsten rod is passed, is surrounded by a glass container which can be evacuated and filled with an inert gas. Electric heating is accomplished by passing a low-voltage current through the tungsten rod from water-cooled terminals.

The most nearly realistic, accident-oriented, fission-product-release experiment will involve an assembly of fuel pins into a cluster so that the heat radiation from the outer pins is conserved to effect complete melting of the inner pins. In five- or seven-pin clusters, complete melting occurs only in the center pin, which is the only one that is irradiated. A demonstration of fission-product release from an assembly of this type includes in one experiment the simulation of the important effects of size and physical arrangement. This will permit an extrapolation to a large-scale release with confidence.

Power-input requirements for a 19-pin array are expected to be about 100 kw, or 20 w per g of UO_2. While a rare gas is the only practical atmosphere for use at present, there is a possibility that steam may be used in later experiments if it is found to be compatible with this system. Platinum-group metals or alloys may be used to produce fuel temperatures in the 1500–1800°C range, and graphite rods should give higher temperatures, but useful service lives under oxidizing conditions remain to be demonstrated.

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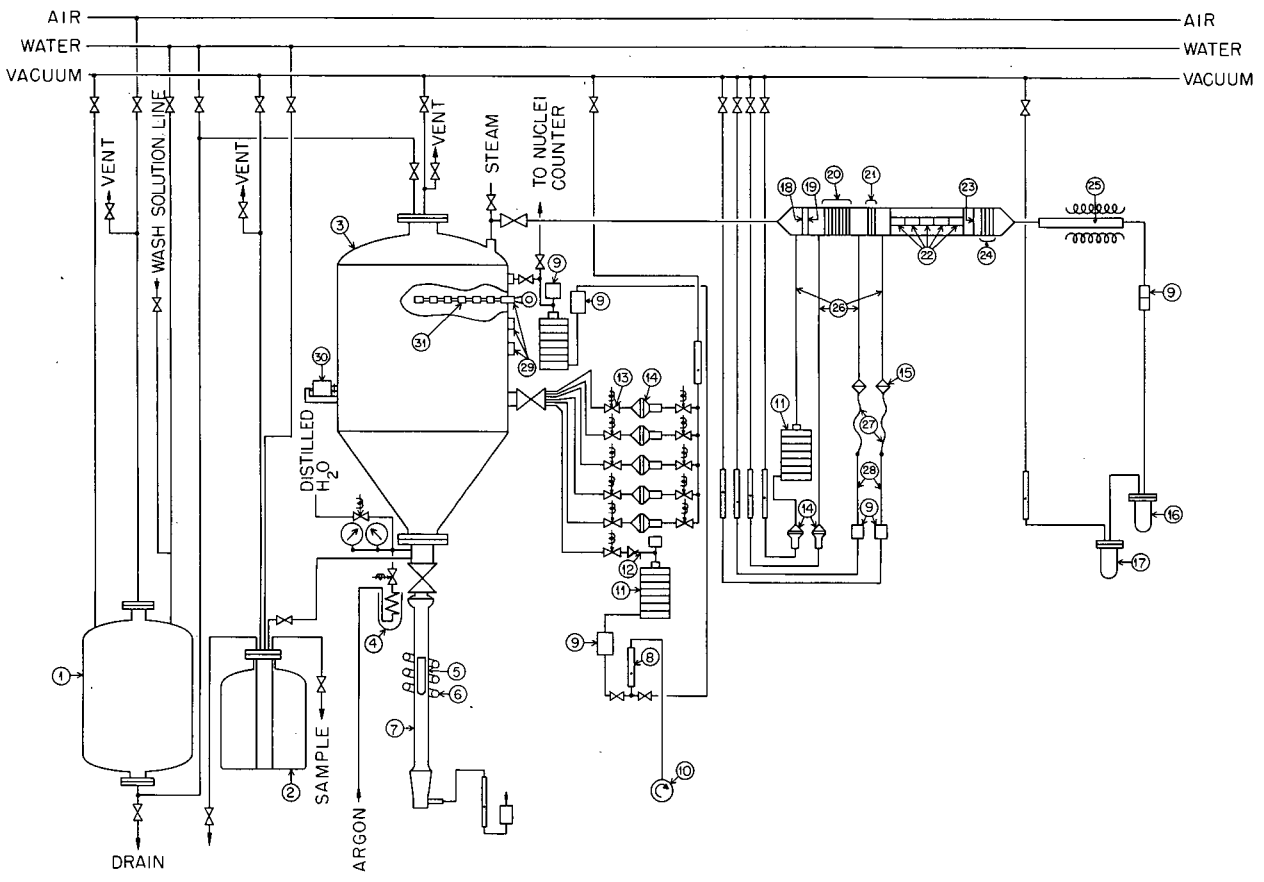

① WASH TANK
② 20-LITER CARBOY
③ 50-GAL. S.S. HOLD-UP TANK
④ COOLING BATH
⑤ CRUCIBLE AND SAMPLE
⑥ INDUCTION HEATER COIL
⑦ QUARTZ TUBE
⑧ ROTAMETER
⑨ CHARCOAL TRAP
⑩ VACUUM PUMP
⑪ ANDERSON "IMPACTOR" SAMPLER
⑫ ORIFICE
⑬ SOLENOID VALVE
⑭ MILLIPORE FILTER AND CHARCOAL
⑮ MILLIPORE FILTER
⑯ RASHIG RING TRAP
⑰ CHARCOAL TRAP
⑱ ROUGHING FILTER
⑲ ABSOLUTE FILTER
⑳ COPPER SCREENS (8)
㉑ CHARCOAL PAPER (3)
㉒ CHARCOAL CARTRIDGES
㉓ ABSOLUTE FILTER
㉔ COPPER SCREENS (4)
㉕ PLATINIZED ALUMINA
㉖ DIFFUSION TUBE (Ag-lined Cu tubing)
㉗ DIFFUSION TUBE (rubber tubing)
㉘ DIFFUSION TUBE (charcoal-lined S.S. tube)
㉙ SAMPLER HOLES
㉚ FAN
㉛ CUTAWAY, SHOWING PLATE-OUT SAMPLER

Fig. 3.4. Hot-Cell Confinement Mockup Facility.

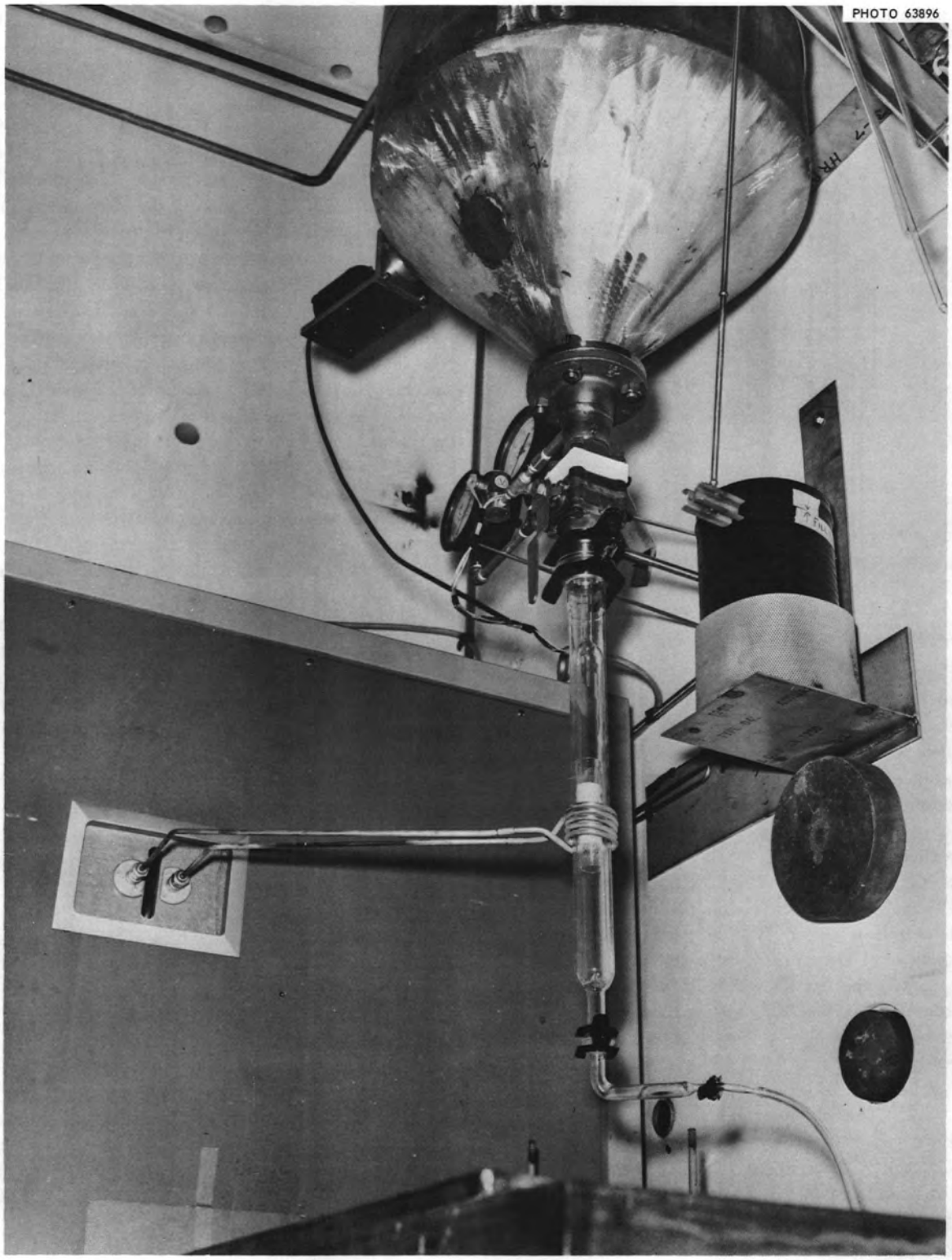

Fig. 3.5. Arrangement of the CMF Furnace Tube, Zirconia Crucible, and Induction Heating Coil.

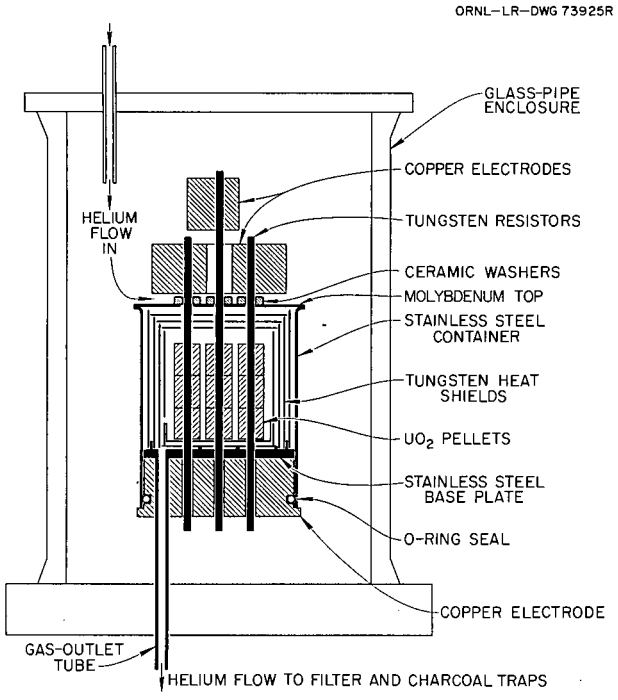

Fig. 3.6. Tungsten Resistor Furnace with Clustered Elements; Test No. 2-6-63.

3.2.1.5 Nuclear Safety Pilot Plant. – The Nuclear Safety Pilot Plant (NSPP) facility consists of a plasma-heating furnace, a model containment vessel, a gas-sampling and cleanup system, and a decontamination system (*20, 38*). The equipment is in a cell in the former Homogeneous Reactor Experiment-2 building. The cell is accessible from the top and is lined with stainless steel. The existing 4-ft-thick concrete shielding walls will permit a mixed fission-product inventory in excess of 1000 curies, although it is expected that most experiments will deal with much smaller sources. The facility can be dismantled and decontaminated remotely but will be assembled by direct procedures.

A schematic diagram of the facility is shown in Fig. 3.7. The plasma-torch heat source generates an inert gas at a temperature of more than 30,000°F which can be directed on the specimen. The melting is performed in a furnace in which a specimen, supported by a water-cooled boat and a ceramic grog (granular ZrO_2 or UO_2), is passed under the flame. Pyrometry and TV viewing are performed through sight ports. The vaporized material is swept from the surface of the melt and transferred to the model containment vessel by means of a short transfer tube. The released material can be either transferred into the containment vessel or directed to a gas-sampling train, depending on the particular experiment.

The model containment vessel is a vertical, cylindrical stainless steel tank 10 ft in diameter and 15 ft long. The vessel is instrumented to determine the disposition of the fission products and associated debris by use of paper-tape air samplers, May Packs, metal coupons, and fallout trays. Also, means are available for testing countermeasures, such as recycle filtration of the containment vessel atmosphere, water spray, and combined steam and water spray removal of small particles by using their properties as condensation nuclei. A decontamination system is built into the vessel for cleanup and total inventory sampling. Liquid wastes are transferred to an existing concrete decontamination pit for sampling and are disposed of through existing facilities at the site. The interior of the vessel is accessible through a 36-in. port at the top and an 18-in. manhole in the side. In addition, two 8-in. ports and two 8-in. purge flow ports are available in various combinations for added flexibility. Equipment for sampling the purge gas is being provided. The vessel is designed to withstand 15 psig external pressure and approximately 50 psig internal pressure.

Secondary containment for the plant is effected by maintaining the cell at subatmospheric pressure using the stack-house blowers. Removal of high-activity-level samples from the containment vessel prior to decontamination will be done with long-handled tools through small openings. Sufficient air flow will be maintained into the openings by the stack blowers, through ducting to the containment vessel, to prevent back diffusion of harmful material. This flow will be passed through particulate filters and iodine-removal beds prior to exhausting it up the stack. The rate of flow through this purge system will be controlled by the allowable concentration of radioactive nuclides in the stack exhaust. The model containment vessel is purged at rates to provide 100 fpm inward air flow through the openings used. A purge flow of 1000 scfm is deemed sufficient to do this.

Plasma Heating Furnace. – An AVCO plasma-gun system (*31*) with a PS 350 series 42-kw power source and control console was adapted to generate plasma capable of melting UO_2.

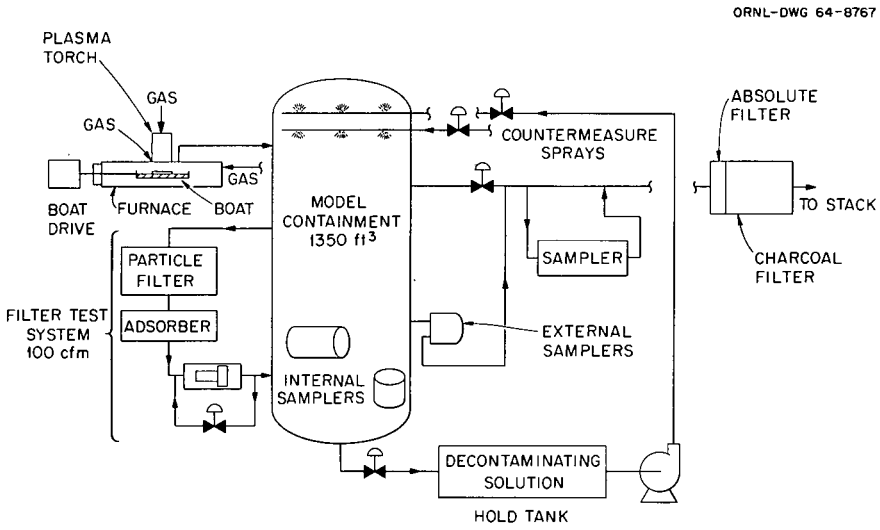

Fig. 3.7. Nuclear Safety Pilot Plant.

Tests were run to check adaptability, feasibility, and reliability of such systems for use in studies of fission-product release from fuel element configurations.

Design information showed that UO_2 pellets could be melted with the carrier moving under the torch at a speed of 1 in./min when the top of the specimen was mounted $2\frac{1}{2}$ in. below the torch and the current setting was 700 amp. Uranium dioxide temperatures can be maintained at lower levels by reducing the current.

It has been demonstrated that the melting can be observed by viewing the specimen through high-density optical filters of the type used by welders. It has also been demonstrated that a Leeds and Northrup matching-wire optical pyrometer can be used to measure the temperature of molten UO_2 in the presence of the plasma. Surface readings between 2700 and 2800°C were measured when the UO_2 began to melt while being heated with the plasma gun.

The final design of a furnace for the NSPP was based on the prototype experimental test results. The furnace was fabricated from stainless steel, employing controlled containment welding techniques. Design provisions incorporated complete in-cell maintenance capability, including in-place charge loading and unloading, and in-place decontamination. The furnace shown in Fig. 3.8 is built in two sections. The trough bottom section has internal tracks for the carrier, or "boat," and a stuffing-box drive-end connector. The rectangular lid section has an attached tantalum heat shield, the torch, an off-gas line to the containment vessel pipe connectors, a pyrometry port, and a visual observation port. Both furnace parts and the "boat" are water jacketed. The furnace halves bolt together on a joint sealed with two rubber gaskets; the space between the gaskets is monitored by a leak detector. All components of the furnace, including a specially built torch and torch disconnect joint, are built to be remotely maintainable from an overhead maintenance shield. The charge carrier boat, capable of handling fuel elements up to 2 in. in diameter and 6 in. long, includes a cover attachment to permit contained transfer of charged boats from the loading cell to the furnace, and of the melted residue from the furnace to the analysis hot cell. A separate boat drive is coupled to the furnace to permit replacement of the furnace or the drive mechanism when necessary.

3.2.1.6 In-Pile Melting of Reactor Fuels in the ORR. – A series of in-pile experiments (*33, 39–41*) is being conducted in which miniature fuel elements are melted by fission and gamma heat in the Oak Ridge Research Reactor (ORR). The fuel specimens are heated internally as a result of neutron and gamma fluxes; these experiments simulate reactor accident conditions, in this respect, more realistically than do out-of-pile experiments.

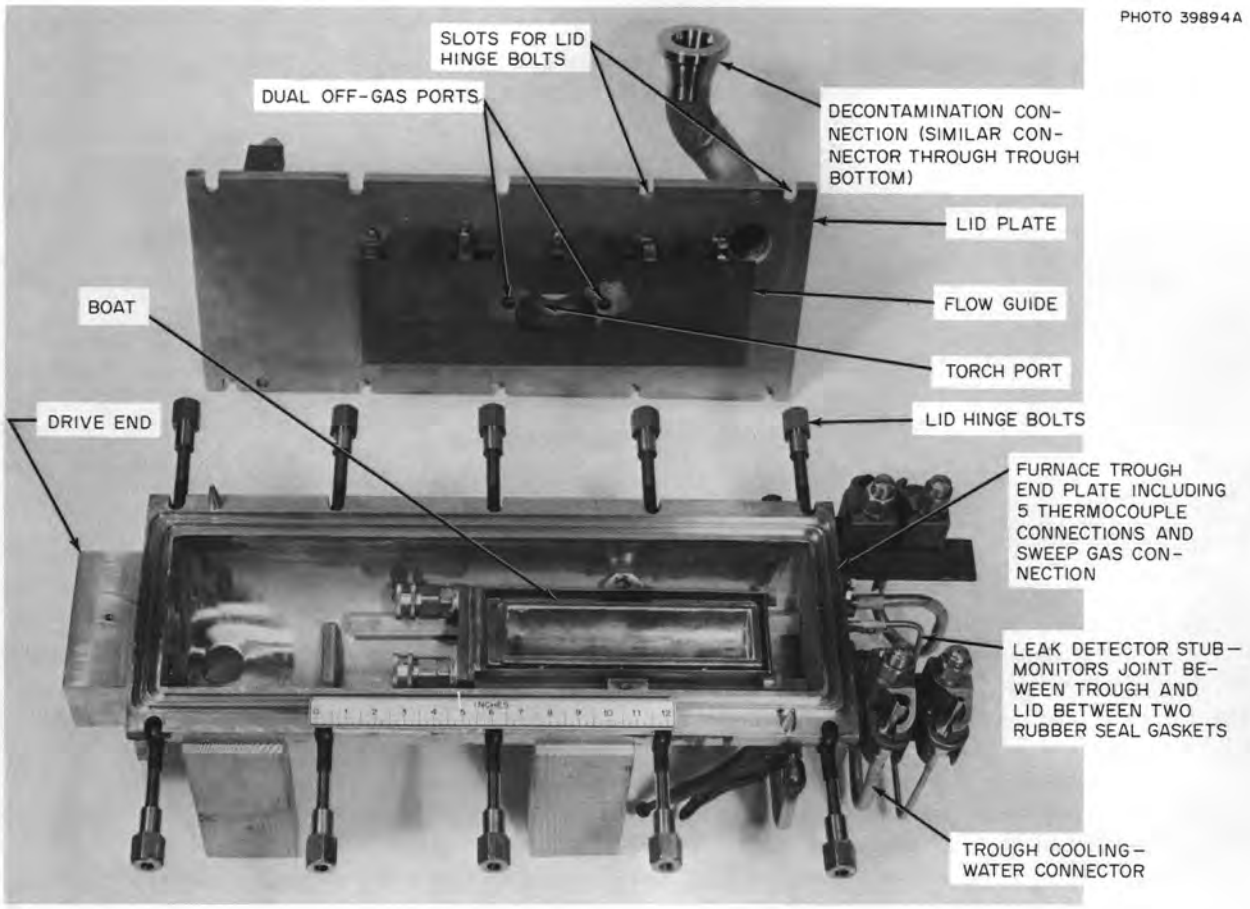

Fig. 3.8. Plasma-Heating Furnace Used in the Nuclear Safety Pilot Plant.

In a typical in-pile meltdown experiment, a fuel specimen is placed in a reactor furnace (Fig. 3.9) and moved to a position close to the top of the lattice of the ORR to build up traces of fission products. After an irradiation period the specimen is lowered to a position in the lattice where fission and gamma heat causes it to melt. Helium or some other gas sweeping over the specimen carries gas-borne fission products to suitable filters and traps, where they are separated and collected. After the specimen is withdrawn from the flux, the furnace, filters, traps, and associated tubing are removed from the reactor. These parts are then cut up in a hot cell, examined, and, finally, analyzed for fission products.

The details of the reactor furnace are shown in Fig. 3.9. Specimens are stainless-steel-clad UO_2 cylinders, 0.210 in. in diameter by 1 in. long. The porous thoria holder serves as a container for the molten UO_2, and both the thoria and a zirconia base serve as heat insulators. These parts are all contained in a primary stainless steel container, which is enclosed in a secondary stainless steel container. Both the top and the bottom of the primary container have aluminum-filled caps to transfer heat from the ends of the primary container to the side of the secondary container. The furnace is cooled by reactor-tank water flowing past the secondary container. The integrity of the system is monitored by stagnant helium pressurized above reactor-tank pressure. Leaks in either containment system are detected by a drop in the helium pressure.

During the time an experiment is in the reactor, a flow of helium or other carrier gas (see Fig. 3.10) passes through the primary system and carries

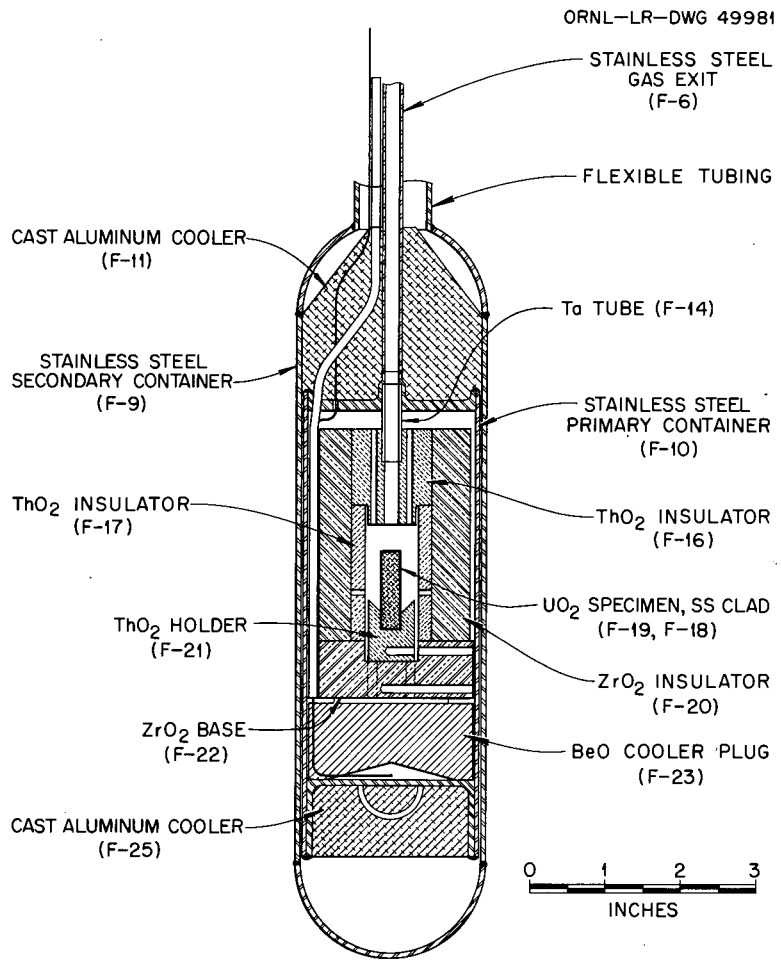

Fig. 3.9. Reactor Furnace for In-Pile Meltdown Experiments.

particulate matter and fission products from the furnace through a stainless steel tube to filters and a charcoal trap. The filters retain particulate matter, and the charcoal, maintained at reactor-tank water temperature (120°F), removes iodine and possibly other fission products from the gas stream. The sweep gas then passes through a liquid-nitrogen-cooled charcoal trap located outside the reactor pool. This charcoal retains all the rare gases released from the irradiated fuel specimen. Radioactive gases in the inlet and exit lines to this trap are monitored by ion chambers connected to recording electrometers in the experiment control room. From the cooled-charcoal trap the helium sweep gas flows into the reactor off-gas system.

Experiments are installed in a permanently mounted facility tube (Fig. 3.11) which enters the ORR top plate through the V-5 access flange and is bent to pass into the F-9 lattice position. The lower end of the facility tube fits into a lattice insert which is machined to fuel element dimensions. A hydraulic positioning mechanism is mounted on the top of the facility tube for the purpose of lowering the furnace into the reactor lattice or raising it to a position above the lattice while the reactor is operating. This mechanism moves the experiment up or down over a 36-in. range. Solenoid valves operated from the control panel direct the flow of process water which operates the mechanism.

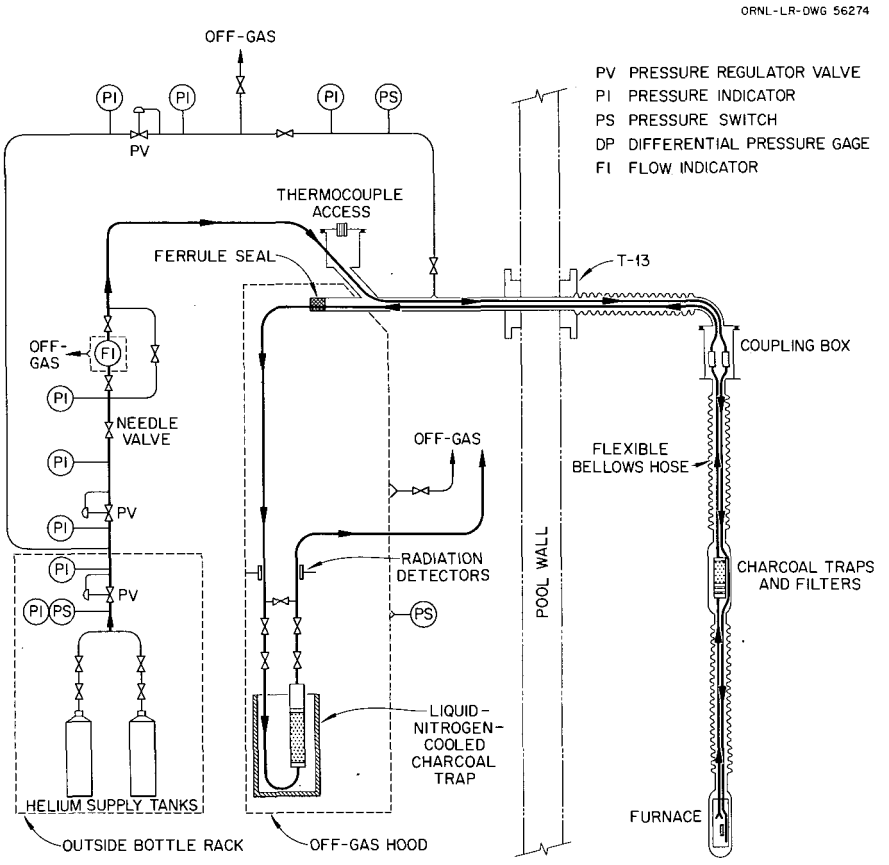

Fig. 3.10. Flow Diagram of the ORR In-Pile Meltdown Facility.

A pipe which penetrates the top of the reactor tank through the flange supporting the facility tube permits tank water to be directed by a pair of mechanically linked valves either into the facility tube, to provide a cooling medium for the experiment, or alternatively through a pipe which runs to the bottom of the facility tube, to operate a hydraulic plunger which closes a foot valve in the cooling water outlet. When the mechanically linked valves are in the latter position, the facility tube is isolated from the reactor tank, and the tube may be opened either to insert or to remove an experiment while the reactor is operating.

The reactor furnace and sample collection apparatus have recently been modified as shown in Fig. 3.12, to permit melting of specimens in a steam atmosphere.

3.2.1.7 In-Pile Melting of Reactor Fuels Under Transient Reactor Conditions. – The purpose of the ORNL fission-product-release experiments (*34*) in the ANL TREAT facility is 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. A major interest in the study is the comparison of fission-product release from transient melting with that from slower, in-pile melting experiments that simulate loss-of-coolant accidents.

A schematic diagram of the experimental assembly is shown in Fig. 3.13. The apparatus is composed of three interconnected parts: a fuel autoclave, a diffusion system that includes a flow regulator, and a second autoclave, initially evacuated, to collect filtered gas from the fuel autoclave. The fuel autoclave contains an electric heater to preheat the fuel specimen in an argon atmosphere. The flow regulator allows gas from the fuel autoclave to flow slowly, after the reactor transient, into the gas-collection autoclave until pressure equilibrium is attained.

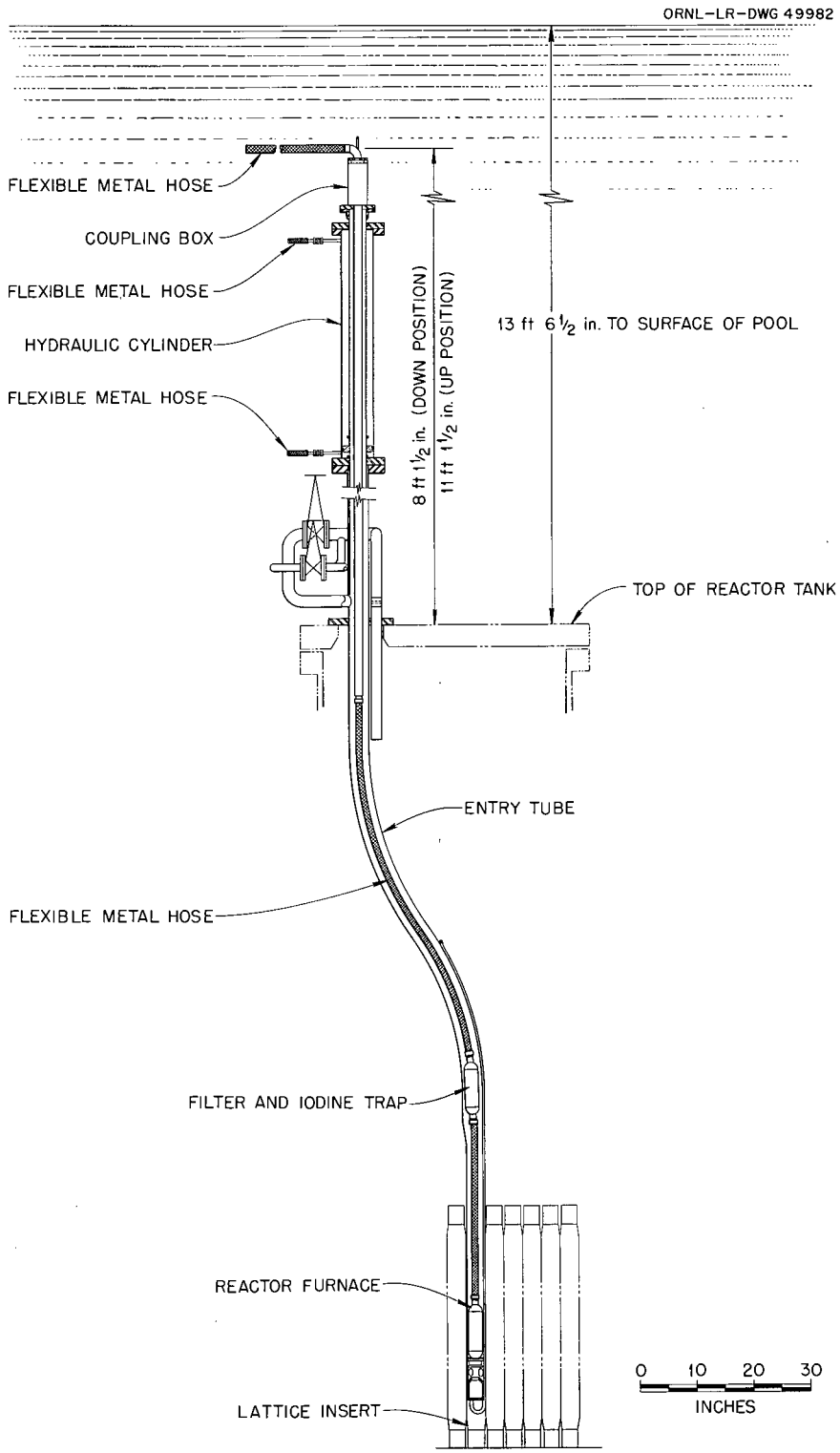

Fig. 3.11. Fuel Meltdown Experiment Assembly in Reactor.

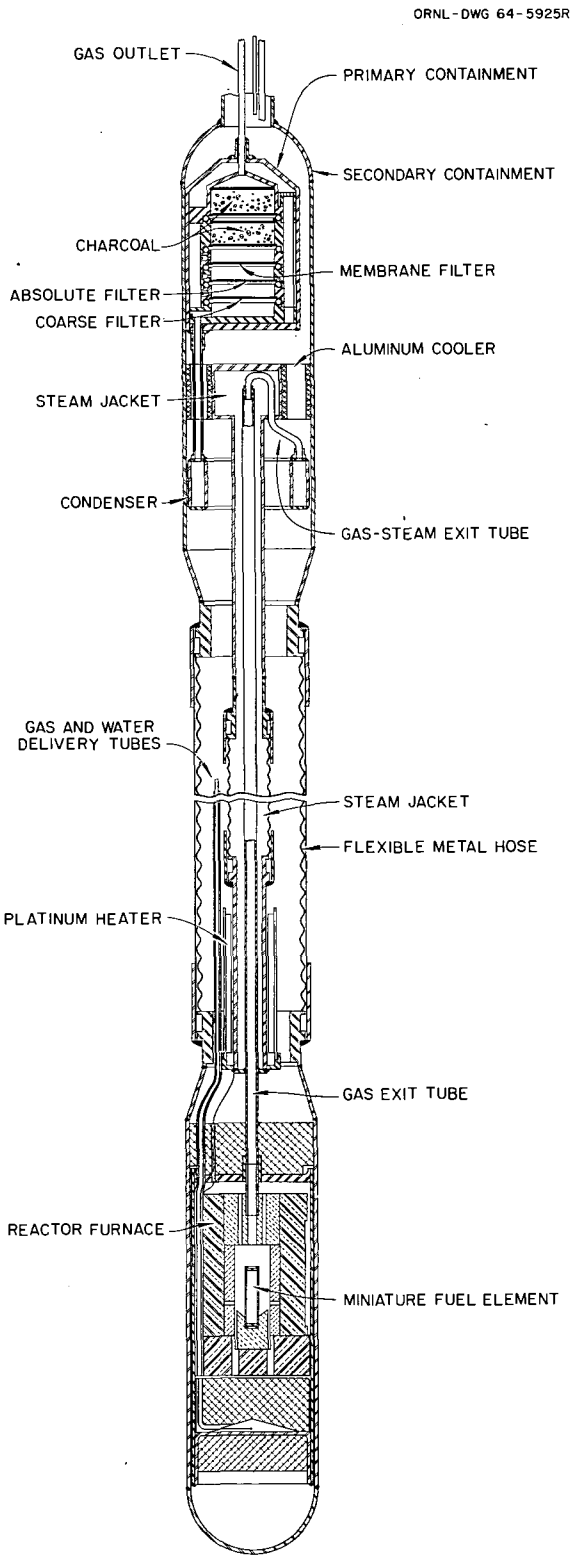

Fig. 3.12. In-Pile Meltdown Apparatus Modified for Use of a Steam Atmosphere.

A drawing of the fuel autoclave is shown in Fig. 3.14. The stainless-steel-clad UO_2 fuel is supported by a $\frac{7}{8}$-in.-long rod on the lower end of the specimen that rests on the bottom of the alumina furnace insulation. The specimen is also supported at the top, by spot-welding to the stainless steel basket. The basket is in turn spot-welded to rods screwed into the autoclave top. Spiraled bright-finish tungsten sheet provided reflective thermal insulation for the heater in experiments conducted with a helium atmosphere. Evacuated stainless steel flasks were used for this purpose in experiments employing oxidizing atmospheres. A Pt–10% Rh heater and a Pt–10% Rh thermocouple are encased in the cast alumina insulator. The 10-mil-diam wires of the Pt–10% Rh thermocouples are individually spot-welded to the cladding to provide accurate and rapid temperature response, and a Chromel-Alumel thermocouple is attached to the fuel autoclave exterior.

The diffusion system consists of a 6-ft-long diffusion tube, 0.195 in. in inside diameter, and a flow controller which regulates the flow to 47 cm^3/min at the temperature and at the pressure range prevailing in these experiments. Valves at the inlet and outlet of the diffusion system isolate the diffusion system from the fuel autoclave and the gas-collection autoclave until after the reactor transient, when they are opened by means of an electrically initiated explosion. The diffusion tube is employed to permit determination of particles in the 30- to 100-A range.

A series of filters at the entrance of the gas collection autoclave provides a means of sampling particles.

3.2.2 Preparation and Purity of Radioactive Iodine Sources

Ackley and Browning (*42*) used a source prepared from a palladium iodide precipitate which was formed by addition of an acidified solution of $PdCl_2$ to a basic solution of sodium sulfite containing ^{131}I plus ^{127}I carrier. The precipitate was washed, dried, and decomposed under vacuum at an elevated temperature. The liberated I_2 was collected in a glass U-tube cooled by liquid nitrogen. It was observed that, when gas was passed through the U-tube held at a temperature of –70°C, there was a rapid initial transport of ^{131}I activity followed by a slower, reasonably constant rate for

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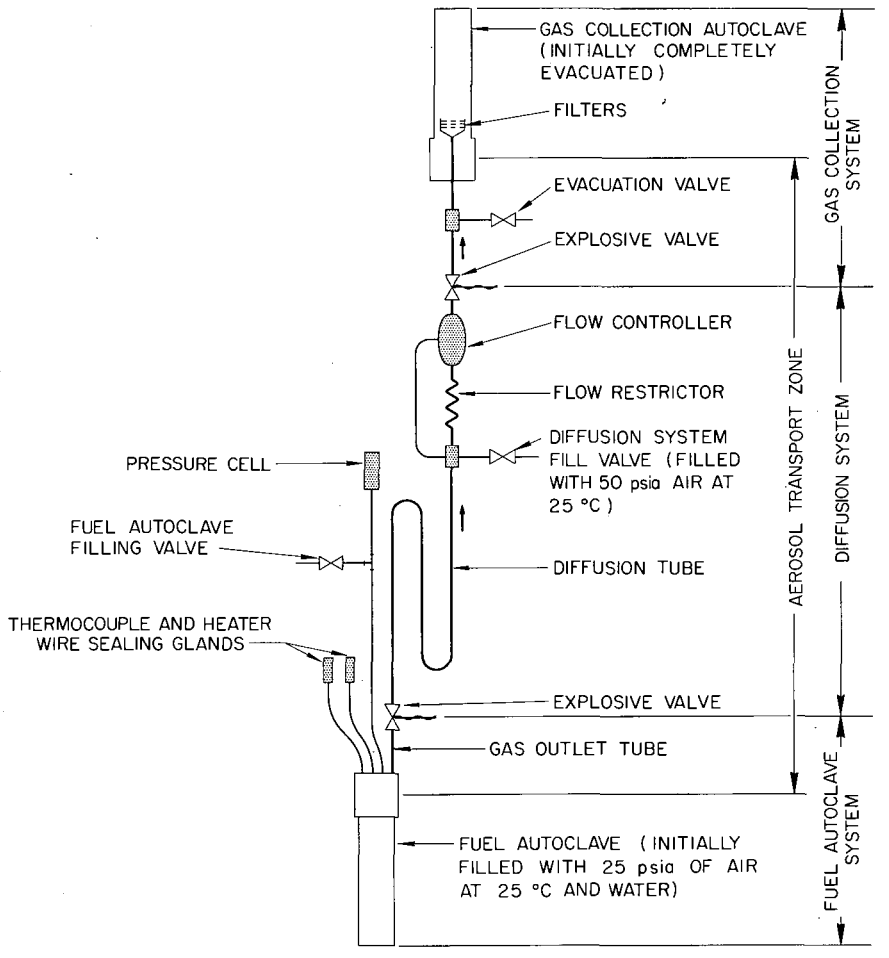

Fig. 3.13. Schematic Diagram of Apparatus for Fission-Product Release Experiments Under Transient Reactor Conditions.

transport of iodine. The initial, rapid release of ^{131}I activity was suggestive of an iodine compound more volatile than elemental iodine. This volatile fraction did not plate out on silver or rubber surfaces, but deposited on a charcoal-lined diffusion tube, with a diffusion coefficient of approximately 0.10 cm^2/sec. The less volatile fraction deposited primarily on silver, with a diffusion coefficient of 0.08 cm^2/sec, which is characteristic of molecular iodine.

Browning and Davis (*43*) reported results of fractionation studies conducted with similar iodine sources. The sources were held at several temperatures ranging from that of dry ice (–78°C) to 0°C with a vacuum of about 10 μ. The volatile material was collected in a liquid-nitrogen-cooled trap and submitted for mass spectrographic analysis. Methyl iodide was a major component of the material volatilized at the lowest temperature, and the amount found in one sample corresponded to 0.09% of the iodine in the original source. The relative amount of CH_3I decreased with increasing temperature and the concentration of I_2 increased, as expected. Trace amounts of HI were observed in all samples, and ICl was found in one source. Since the PdI_2 precipitate was warmed under a vacuum to remove water prior to its decomposition, the CH_3I must have been formed by reaction of I_2

vapor with hydrocarbon present in the vacuum system, possibly from the vacuum pump oil.

Ackley and Browning (*42*) also discussed the purity of iodine sources prepared by the dichromate method. They found a lower purity (98.8% I_2) when air was used as a sweep gas than when helium was employed (99.6% I_2). This method of iodine generation has also been employed in more recent studies (*43*) of iodine deposition in composite diffusion tubes. Evaporation of a solution containing $Na^{131}I$, to which $K^{127}I$ carrier was sometimes added, left a residue which was covered with a 0.5-in.-deep bed of $K_2Cr_2O_7$ crystals. The iodine generated by this method was collected in U-tubes. The temperature or the amount of iodine in the U-tube and the air flow rates were controlled to give an iodine concentration in the air stream of 10^{-3} to 20 mg/m^3.

French investigators (*44*) employed the technique of thermal oxidation of a thin film of NaI in air or oxygen at temperatures of 800 to 900°C to release iodine at a slow, uniform rate over a long period of time. Eggleton and Atkins (*45*) reported that a small fraction of iodine produced by this method consists of two types of compounds, presumably organic iodides, which are discussed in more detail in Sect. 3.6.

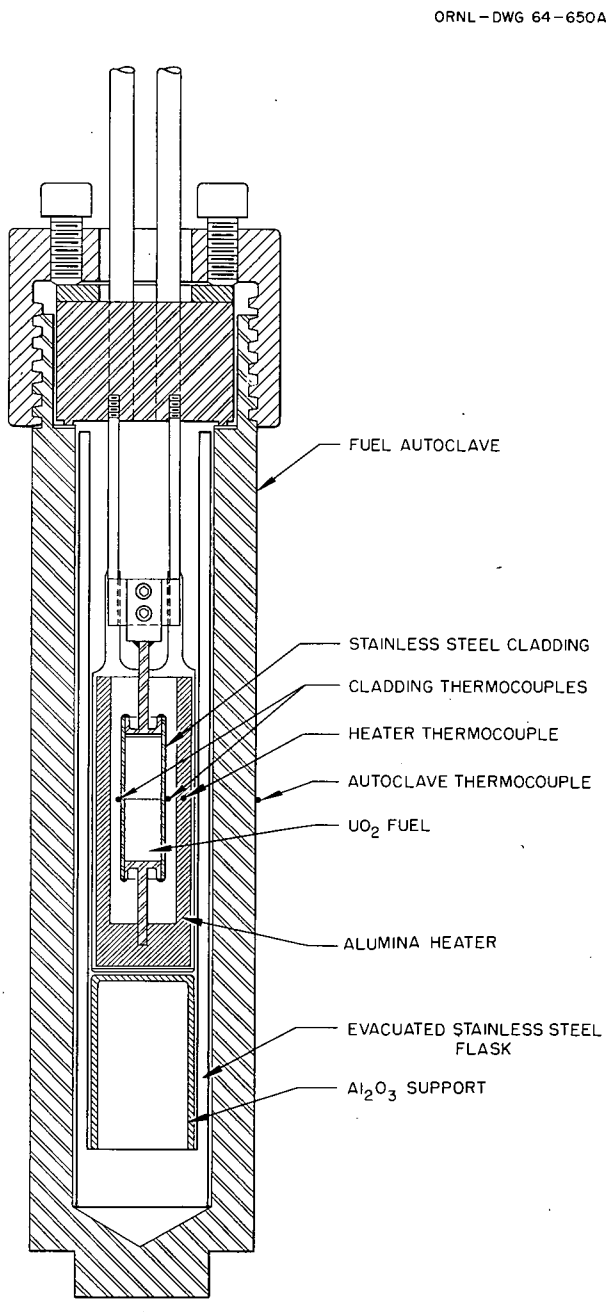

Fig. 3.14. TREAT Fuel Autoclave Assembly.

3.2.3 Simulation of Fission-Product Iodine

Experimental work discussed elsewhere in this report has demonstrated that when small amounts of iodine are released, for example, when trace-level irradiated fuel is heated, its behavior may be different from that observed when it is present in more realistic concentrations. Methods of supplying such concentrations of iodine without the high radiation levels associated with high-burnup fuel are discussed in this section.

Iodine, particularly when it is released in the molecular or atomic form, can apparently associate with airborne particles. It is desirable, therefore, that iodine be released in the presence of particles from overheated fuel or cladding materials in order to simulate accident conditions. Different approaches to this problem have been used or are available for use.

In one method that has been employed (*46*), molecular iodine containing a suitable amount of ^{131}I, along with sufficient ^{127}I carrier to give a concentration of airborne iodine corresponding to that from 7000-Mwd/ton-burnup fuel, was sealed in a small quartz vial. This was separated from the stainless-steel-clad UO_2 specimen (unirradiated) by granular ZrO_2. It was expected that the quartz vial would not rupture until the cladding in the inductively heated specimen melted and ran down through the ZrO_2. However, in the one experiment

of this type that was reported, the capsule ruptured before the cladding and UO_2 melted. The behavior of iodine in this experiment was, nevertheless, quite similar to that exhibited in a similar experiment performed with high-burnup fuel.

A different approach has been employed in tests performed in the Nuclear Safety Pilot Plant (*47*). Tube-shaped UO_2 fuel specimens were employed, and the hollow centers were packed with powdered UO_2 mixed with $Na^{127}I$ and about 10 mc of $Na^{131}I$. The amount of $Na^{127}I$ employed was varied to study the concentration effect when the aerosol, resulting from heating the fuel with a plasma torch, aged in a stainless steel tank (see Sect. 3.2.1.5). Under the NSPP test conditions employed to date, most of the iodine was apparently in the molecular state when the aerosol was initially introduced into the model containment vessel.

The iodine simulation method employed in tests by Collins *et al.* at Windscale (*48*) was similar to that described in the previous paragraph except that molecular iodine apparently was deposited in stainless steel fuel cans which were sealed after the addition of unirradiated UO_2 pellets. Iodine was then released when the cans melted.

The relative merits of these and other possible methods of simulating fission-product iodine, as well as the degree of realism of the simulation, remain to be established.

3.3 PARTICLE SIZE AND NUMBER DETERMINATIONS

Radioactive particles generated by overheated reactor fuel in reactor accidents may vary in size from near-molecular dimensions (~20 A in diameter) to sizes large enough to fall rapidly. The latter group will not travel far enough to constitute a hazard outside of the building that houses the reactor. The small particles, however, are essentially unaffected by gravity and may remain airborne for hours or days before they diffuse to a surface and remain there. Such small particles are not easily collected by conventional filtration equipment, and the amounts and types of radioactive fission products associated with them are consequently a matter of concern to hazards analysts. Much effort has, therefore, been devoted to development of methods for characterizing such particles and to collection of information about their behavior in simulated reactor accidents. Techniques for determining sizes and numbers of particles are discussed in this section; collected data are summarized in Sect. 3.4.

3.3.1 Diffusion Channel Technique

Diffusion is an important mechanism in aerosol deposition, and, since the diameter of a particle is related to its diffusion coefficient, it is possible to measure the size of particles and to identify molecular vapors by means of their diffusion coefficients. A method of determining diffusion coefficients of fine particles and of radioactive vapors such as iodine by measuring the distribution of radioactivity on the walls of cylindrical and rectangular channels previously exposed to gas carrying radioactive materials and flowing under laminar conditions was described (*49–51*). Since cylindrical tubes are currently being employed exclusively in this work, an equation corresponding to deposition of a given species in a cylindrical diffusion channel, obtained from the equation of Gormley and Kennedy (*52*), was presented (*50*):

$$n_s = \frac{N_0 D}{Q}\left(9.4106e^{-11.489DZ/Q} + 6.8309e^{-70.06DZ/Q} + 5.8198e^{-179.07DZ/Q}\right) .$$

In this equation, n_s = number of particles deposited per unit length, N_0 = number entering the channel, D = their diffusion coefficient, Q = volumetric flow rate of carrier gas, and Z = distance from channel entrance. For the case of multiplicity of species, the deposition would be given by a summation of expressions of the form of this equation. The diffusion channel method was tested with a narrow (5-mil) rectangular channel and a 26-ml/min flow rate using radioiodine vapor, ^{131}I-labeled 0.004-μ aluminum oxide particles, and ^{131}I-labeled 0.25-μ tobacco-smoke particles; the particle sizes and/or diffusion coefficients of these three materials were obtainable by independent means. In all three cases, good agreement between theory and experiment was observed, indicating that this method is applicable over the range of interest. The practical upper size limit under conditions currently employed ($\frac{3}{8}$-in. tubes, 100-ml/min flow rate) is approximately 100 A (0.01 μ).

Examination of the above equation shows that it describes a straight line only under conditions that permit the second and third terms to be neglected. Calculations (*53*) show that under the conditions listed above, these terms drop out a few centimeters from the entrance of the diffusion tube for particles smaller than 50 A, but for 100-A particles tube lengths greater than 100 cm are required to give a linear deposition rate and different conditions should probably be employed. One fundamental assumption of the diffusion channel method is that the walls are a "perfect sink" for the particles. There seem to be adequate indications in the literature that this assumption is valid for the particle sizes of greatest interest, smaller than 0.1 μ. Wall materials for iodine collection are considered in Sect. 3.5.

3.3.2 Filter Analysis Techniques

Silverman and Browning (*54*) have developed a method for characterizing radioactive aerosols by determining their distribution as a function of depth in fibrous filters under carefully controlled conditions. The method distinguishes the contributions of three major processes of filtration: diffusion, interception, and inertial impaction. For experiments with air velocities in the range where diffusion was the controlling mechanism, particle sizes in the range of 40 to 300 A thus determined compared favorably with those obtained from concentration and flow data and from electron photomicrographs. This technique has not been used to date in fission-product release experiments. Browning *et al.* reported (*40*) that the sweep gas from in-pile fuel melting and burning experiments is passed through a series of filters to obtain information about the particulate material which is formed. Figure 3.15 shows the filter assembly which was designed for this purpose. It includes a roughing filter, an HV-70 absolute filter, and a 10-mμ membrane filter. The sizes of particles collected on the various filters, their states of aggregation, and certain other information about them can be determined by electron microscopy. The gross

Fig. 3.15. Filter Assembly and Charcoal Trap Used in In-Pile Fuel Destruction Experiments.

amounts of fission products collected are determined by radiochemical analysis.

3.3.3 Deposition Techniques

Both thermal and electrostatic precipitators have been employed to collect small particles generated in simulated reactor accidents. The electrostatic method requires use of a high-voltage device in order to impart a charge to the particles and to permit efficient collection of the charged particles. A device of this type (*55–56*) was employed in the Containment Mockup Facility (*25*) but its efficiency was not evaluated. Two types of thermal precipitators have been described, the hot-wire (*57*) and the hot-plate (*58*) type. Qualitative tests (*57*) indicated a high efficiency for the hot-wire precipitator, and it appears to offer fewer operating problems than the electrostatic precipitator and may be the preferred type for that reason. Samples can be collected in a form suitable for size determinations with an electron microscope. The Andersen Air Sampler (*59*) is useful for collecting particles larger than about 0.6 μ, and it has the advantage that particles in different size ranges are separated and can be submitted for radiochemical analyses or for size evaluations by electron microscope techniques. Other types of inertial samplers have been described (*60*). One that makes use of centrifugal forces up to 26,000*g* has been described by Goetz and Preining (*61*). These investigators state that the aerosol spectrometer, which is commercially available, separates quantitatively airborne particles in the diameter range 3 μ to 0.03 μ from the atmosphere in a continuous band-shaped deposit. They applied the instrument to the analysis of natural and artificial aerosols in the submicron range. Craig (*62*) also employed this type of spectrometer to study the interaction of the $Na^{131}I$ vapor with labeled iron oxide particles.

3.3.4 Counting Techniques

The number concentration of particles (population) in aerosols resulting from the overheating of reactor fuels is of interest, and several techniques have been used or are available for use for particle counting. These range from a simple, relatively inexpensive instrument (*63*) for manual counting of the total number of condensation nuclei to more elaborate and more expensive instruments (*64*) that provide counts of the number of particles of various sizes ranging from 0.3 to 16 μ in diameter. The condensation-type instrument has a maximum scale reading of 10^7 nuclei/cm^3, and it permits size discrimination to a limited degree. The latter instruments use the light-scattering principle, and they have counting rates up to 30,000 particles/min.

3.4 SIZE AND ACTIVITY DISTRIBUTION OF PARTICLES FROM OVERHEATED FUELS

Information on the size and, to a lesser extent, the shape and density of particles evolved from overheated fuels in expected reactor environments is needed in order to estimate the hazard of reactor accidents. Aerosols having a high concentration of small particles are highly unstable, and rapid agglomeration occurs until the number concentration is reduced to such a degree that collisions of particles become infrequent. Both theoretical and experimental studies of particle agglomeration have been made, but the effect of radioactivity has not been determined to date at the high specific activity levels that would result from nuclear accidents. Furthermore, the agglomeration process results in large particles with irregular shapes and, in many cases, particles having unknown amounts of void space within them. Methods of relating microscopic observations of particle agglomerates to probable deposition rates apparently remain to be developed. Experimentally measured particle sizes are reported in this section, along with some information on amounts of iodine and tellurium associated with particles of different sizes. Tellurium is of interest to this study because of the ^{132}I daughter of 77-hr ^{132}Te.

3.4.1 Oxidation of Uranium in Air

The temperature at which uranium metal oxidizes in air was found to have a pronounced effect on the size of particles transported (*65*). Less than 4% of the oxide formed at temperatures ranging from 400 to 1200°C was in the size range below 10 μ. At air velocities of 8.3 cm/sec and lower, the quantity of oxide particles, primarily agglomerates of particles 0.015 to 0.5 μ in diameter, collected 20 in. from the sample, was greatest at 1200°C;

at 400 to 800°C, the average diameter of particles collected under these conditions was 2 to 5 μ. Assuming that the smaller particles, found at both 1000 and 1200°C, arise from the condensation of sublimed UO_3, one would predict a large decrease in their concentration with decreasing temperature, as observed. As air velocities are increased, aerodynamic entrainment of larger particles overshadows release by direct volatilization. Rapid cooling of oxidized specimens resulted in projection of microscopic and macroscopic particles at high velocities. Qualitatively, samples with higher burnup were found to exhibit an accelerated particle release (*66*). Results of preliminary studies (*67*) showed that ^{132}Te was carried on particles with diameters greater than 200 A, while the majority of ^{131}I was carried on particles having a bimodal size distribution with peaks at 15 and 60 A (*67*). In other studies (*68*), also with metallic uranium heated in air, less than 2% of the Te, Cs, Zr, and Ba was found to be associated with particles larger than 1 μ in mean diameter, while 6% of the I deposited with particles larger than 1 μ (*68*). It was reported (*69–70*) that agglomerates of 1- to 2-μ particles were formed when uranium was oxidized at a temperature of 1800°C or higher.

3.4.2 Particles Formed from Vaporization of UO_2

Significant fractions (up to 25%) of small UO_2 specimens vaporized when melted in helium, air, and CO_2, although most of the vaporized material deposited in hot regions (*71*). Most of the particles formed were in the 0.01- to 0.1-μ size range, as shown in Fig. 3.16. Iodine, Te, Cs, and Ru were the major fission products transported out of the heated regions by air. Stainless steel cladding altered the particle size distribution (*37*), as shown in Fig. 3.17. When Zircaloy-clad UO_2 was melted under the same conditions, the amount of uranium collected on filters was reduced by a factor of 800 as compared with the stainless-steel-clad fuel. Eighty-seven percent of the particles transported from unclad UO_2 in out-of-pile tests were less than 0.5 μ in diameter, but when stainless-steel-clad UO_2 was heated in-pile with a helium atmosphere, two size groups were found: one centered about 22 A, the other about 30 A (*39*). Thirty-nine percent of the iodine carried out of the furnace was attached to the 22-A particles. Some cesium, strontium, tellurium, and barium were also found on these particles. When a relatively large quantity (35 g) of unirradiated UO_2 was maintained above its melting point for 5 min in a helium atmosphere, 10% of the UO_2 vaporized but approximately 9%

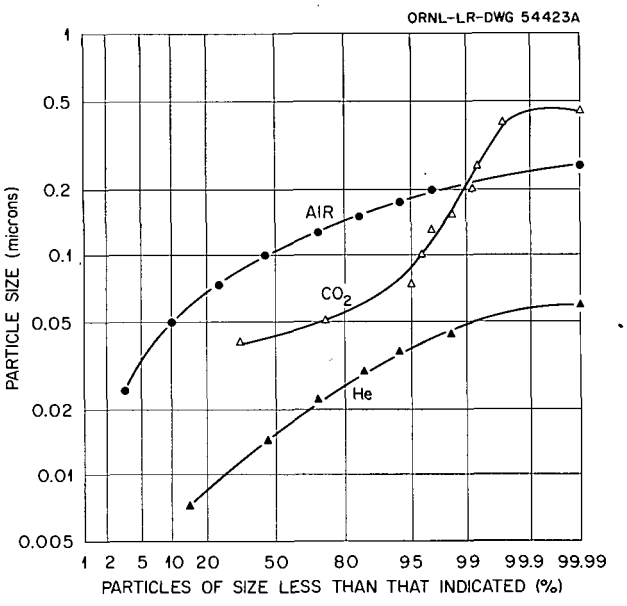

Fig. 3.16. Particle-Size Evaluation of Oxides Vaporized from Melted UO_2 (Arc-Image Furnace).

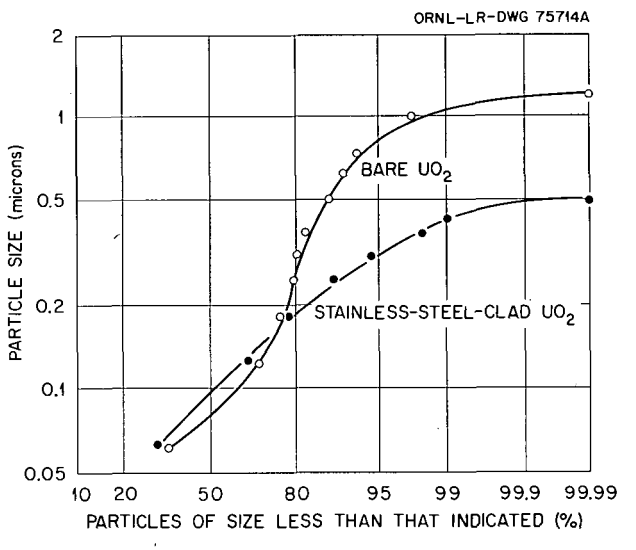

Fig. 3.17. Comparison of Size Distribution of Particles from Bare UO_2 and Stainless-Steel-Clad UO_2.

deposited in the furnace tube and most of the remaining 1% stopped in a low-velocity region. Only about 0.005% reached the filters.

The distribution of released fission products and uranium deposited from a gas flowing laminarly through a diffusion tube was analyzed to yield information about vapors, gas-borne particles in the range of 10 to 100 A in diameter, and the amounts of released material associated with these forms (*41*). In most of the experiments in which stainless-steel-clad UO_2 specimens were heated in-pile, the data indicate that particles having an average diameter of about 35 A were released. Considerable amounts of released ^{131}I, ^{132}Te, and ^{137}Cs are always carried by these fine particles. Particle size distribution data obtained from filters in two of these experiments are shown in Fig. 3.18, along with some data on particles from uranium carbide burning experiments discussed in the following section. Similar data obtained in experiments (*72*) to determine fission-product release from stainless-steel-clad UO_2 under transient reactor conditions are illustrated by Fig. 3.19. The data indicate that an oxidizing atmosphere resulted in a larger number of small particles than were produced (*73*) in helium (experiments 1 and 2).

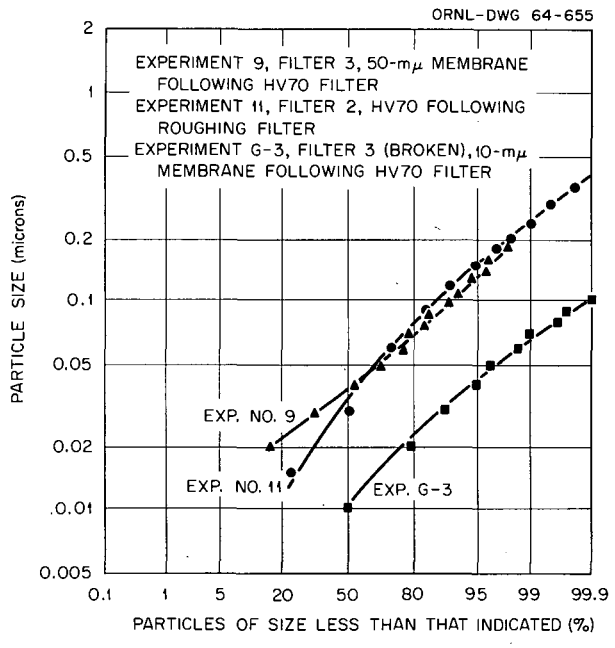

Fig. 3.18. Size Distribution of Particles Retained on Filters in In-Pile Fuel Destruction Experiments.

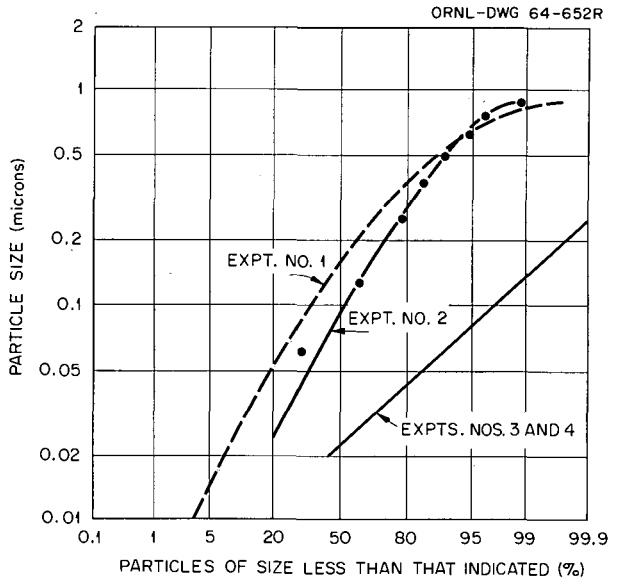

Fig. 3.19. Sizes of Particles Collected on Filters During In-Pile Melting of Stainless-Steel-Clad UO_2 Under Transient Reactor Conditions.

3.4.3 Particles Formed by Oxidation of Uranium Carbide

Little information seems to be available on particle distribution resulting from exposure of uranium carbide to oxidizing atmospheres. Two experiments have been reported (*40*) in which fuel specimens composed of pyrolytic-carbon-coated uranium carbide particles in a graphite matrix were partially destroyed by exposure to air in the ORR at temperatures ranging from about 890 to 1400°C. Varying amounts of iodine (13 or 58%) and of tellurium (8 or 32%) were found to be associated with particles in the 15- to 30-A size range. The size distribution of particles collected on two filters in series during one of these experiments is shown in Fig. 3.20. The median particle diameter of the particles collected on filter No. 2 (type E glass) was 1.2 μ, while that on filter No. 3 (HV-70) was 5 μ. In another experiment of this type, particles collected on a 10-mμ membrane filter after passing through an HV-70 filter showed a median diameter of 0.01 μ, but this filter was broken, possibly during assembly of the filter holder, so there is doubt as to the validity of the data. Size distribution data obtained from this filter are included in Fig. 3.18.

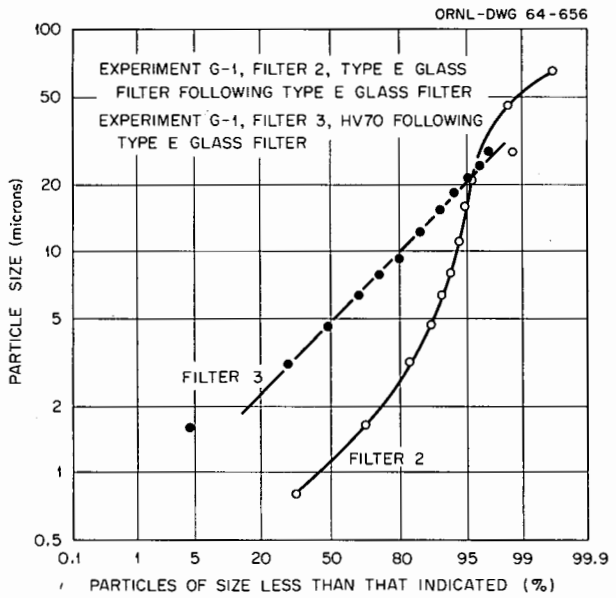

Fig. 3.20. Size Distribution of Particles Retained on Filters in Fueled Graphite Burning Experiments.

3.4.4 Particles Produced on Melting Irradiated Aluminum-Uranium Alloys

Some observations were made (*25*) of the particles produced when highly irradiated (23.6% burnup) aluminum-uranium alloy specimens were melted in a steam-air mixture at about 750°C. Particles remaining airborne in the CMF tank for 1 to 2 hr were collected by an electrostatic precipitator and examined by electron microscopy. The smallest particles noted were approximately 150 A in diameter; the largest had an average diameter of about 1500 A. A portion of the aerosol was simultaneously passed through a 10-ft length of silver-coated diffusion tube, and the remaining particles collected on a membrane filter were found to range in diameter from about 0.05 to 3.0 μ. The large particles were agglomerates of small particles.

3.5 SAMPLING AND MEASUREMENT OF DIFFERENT FORMS OF IODINE

The various forms in which iodine released in reactor accidents may appear have been discussed in previous sections. In this section, methods of separating, identifying, and determining the amounts of these various forms are discussed. Techniques for collecting and measuring the size of particles were considered in Sect. 3.3 and are considered here only in relation to the problem of determining the fraction of iodine present in the particulate form. Airborne iodine can be divided, for sampling purposes, into three classes of materials: gases, particles large enough to have easily measurable deposition rates, and particles small enough (submicron size) so that diffusion processes rather than gravity control their behavior. Considerable progress has been made in recent years both in the development of techniques for characterizing airborne iodine and in understanding its behavior under some possible accident conditions; however, there is a great deal of activity in this field at present, and this discussion should only be considered a progress report that summarizes published information from other installations and some as yet unpublished ORNL developments.

3.5.1 Deposition Samplers

The deposition rate of iodine, particularly when it is in the molecular form, is strongly affected by the physical and chemical nature of the surface on which it deposits. Thus, the adsorption and desorption behavior of iodine varies with the type of surface used for deposition measurements. It is necessary, therefore, to employ structural materials as the sample collection media in experiments where efforts are being made to observe the behavior of iodine. Horizontal samplers placed near the bottom of the containment shell will collect mainly the larger particles. Horizontal samplers at other locations, as well as vertical samplers, may be used to give an indication of the movement of particles within a containment vessel.

The simplest form of deposition sampler is a tray or even a flat or curved surface of known area. Unless a means is provided for removing the sampler during the course of the experiment, or for protecting it from further exposure to the contaminated atmosphere after a controlled period of time, such samplers will indicate only the cumulative deposition occurring during the experiment. While this is useful information, especially in large simulated containment systems where the distribution is not likely to be uniform, deposition rates are of greater interest. Consequently, several arrangements have been developed for obtaining

a series of deposition samplers at a single point.

One device of this type is shown in Fig. 3.21 (*46*). It consists of small cylindrical specimens $^3/_8$ in. in diameter and 0.8 in. long connected by $^1/_2$-in.-diam rods which permit pressure-tight seals with Teflon and Neoprene O-ring seals. Different types of sample materials can be used to collect samples. Initially all segments are exposed to the containment tank atmosphere; they are withdrawn from exposure one segment at a time at the desired time interval. This type of sequence deposition sampler is especially useful where it is not possible or desirable to have large access holes in the containment tank. It can be operated remotely by use of a long handle.

An unassembled view of another recently described sequence deposition sampler that permits taking 11 samples on a predetermined schedule is shown in Fig. 3.22 (*74*). The enclosed vibratory motor shown at the left of the photograph is mounted below the sample holder plate, which it rotates. An O-ring seal limits exposure of the contaminated atmosphere to individual sample collectors placed in the depressions of the plate. It also protects the sampler during the time that the containment tank is being purged and while the samplers are being removed from the tank. The top and the sample holder plate are normally removed together, while the remainder of the device need only be removed for maintenance work or decontamination.

3.5.2 Paper Tape Samplers

Paper tape air samplers have been used for many purposes, and it was natural to apply them in iodine studies. It is obvious that variations in the amount of particulate iodine present in an aerosol can be followed by collecting the particles on a paper tape, but it would be very desirable to obtain further information. It was suggested (*75*) that two tapes could be used together, the first that the air sample passes through being an "absolute" filter to collect particles and the second a charcoal-impregnated filter. Apparatus of this type has been employed to sample the atmosphere in the containment vessel in the Nuclear Safety Pilot Plant (*76*). Different arrangements have been

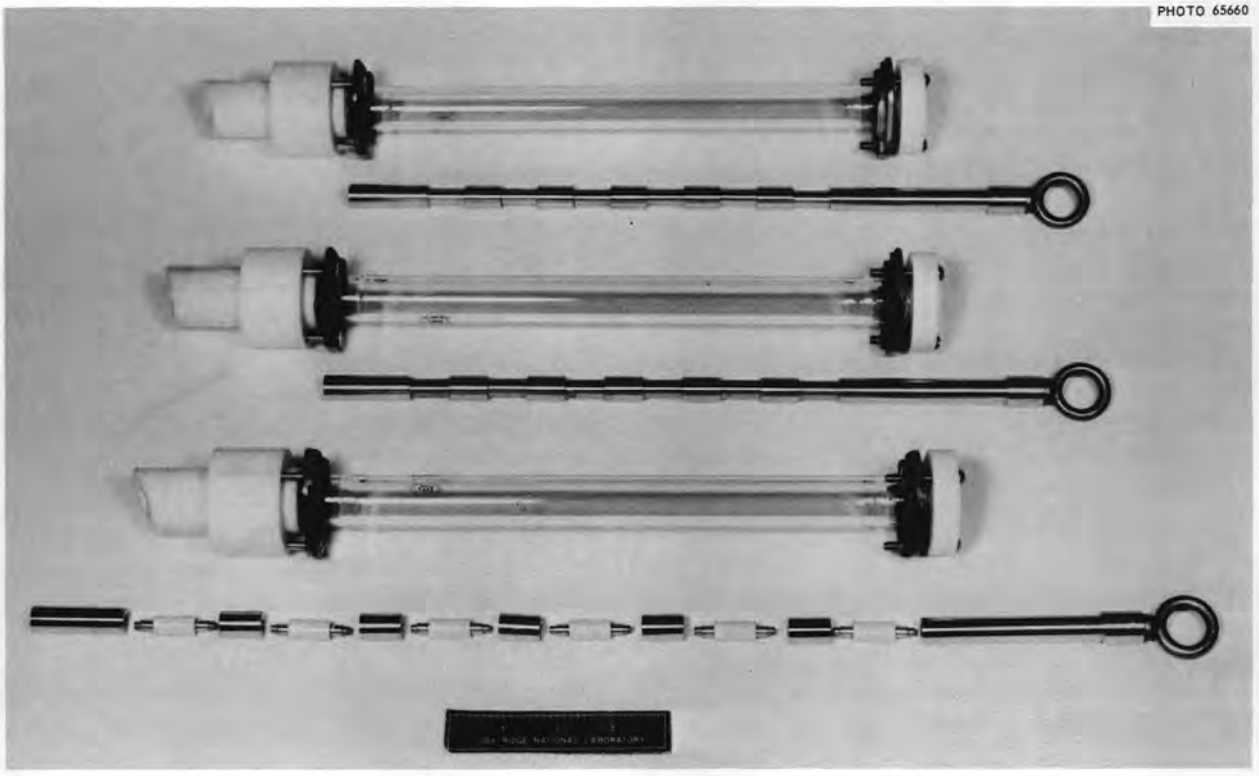

Fig. 3.21. Segmented Deposition Samples Employed in the CMF.

Fig. 3.22. Remotely Operated Deposition Sampler (Unassembled).

tested, and some difficulty was experienced with breakage of ordinary Millipore paper. The apparatus currently in use, which seems to operate satisfactorily, employs a modified Gelman-tape sampler with a 1-in.-wide Microweb Millipore tape (0.45-μ pore size) and type ACG/B charcoal-impregnated filter tape of similar width. As with most tape samplers, sampling is performed at intervals set by interval timers. During the early part of experiments, when rapid changes in the iodine content of the aerosol are expected to occur, sampling is performed for 1 min at a rate of 1 liter/min at 4-min intervals; 5-min samples are collected at intervals of 2 hr toward the end of the 24-hr aerosol aging period. Insufficient data are available at the present time to completely evaluate the usefulness of this apparatus, but recent experience indicates that it may provide a useful supplement to information obtained with May Pack samplers described in the following section.

3.5.3 May Packs

Various arrangements of iodine-retaining materials that permit separation and collection of various forms of airborne iodine are commonly referred to as May Packs, because F. G. May (AERE-Harwell) was the first to use such an arrangement. The version described by Collins (*77*), shown in Fig. 3.23, is fairly typical of current usage. It consists of the following components, through which the gas passes in the order listed:

1. Four layers of fine copper gauze, especially cleaned before use and presumably effective for collecting elemental iodine vapor.
2. Type PH Millipore filter, which is highly effective for collecting small particles and consequently gives a measurement of particulate iodine. However, some molecular iodine may be adsorbed on or react with this paper.
3. Two carbon-impregnated glass fiber papers (Whatman type ACG/B). Activated carbon is the best known adsorber for ill-defined compounds of iodine that can penetrate elemental iodine adsorbers (such as copper) and particulate filters. By using these papers, the equivalent of only a very shallow bed is exposed to the gas stream and only the more reactive compounds are presumed to be retained.
4. Two successive 2-in.-deep beds of SCII charcoal (−18 +52 BSS mesh size). These beds retain the less reactive iodine compounds, such as methyl iodide.

The components of the pack are separated by PVC gaskets and the whole assembly is welded inside a PVC envelope to prevent leakage. Holes to permit gas passage are cut in the envelope just before use. The pack is designed to take a gas flow of 600 cm^3/min. The separate components are analyzed by gamma spectrometry after use in the experiment.

The ORNL version (*46*) of the May Pack shown in Fig. 3.24 differs from that described above.

Since this apparatus is used to study the behavior of iodine released from irradiated fuel, it was considered advisable to have filters in front of the screens in order to remove particles carrying various fission products that might adhere to the screens or be collected on the charcoal-impregnated filter paper and interfere with the determination of iodine behavior. The copper screens have been

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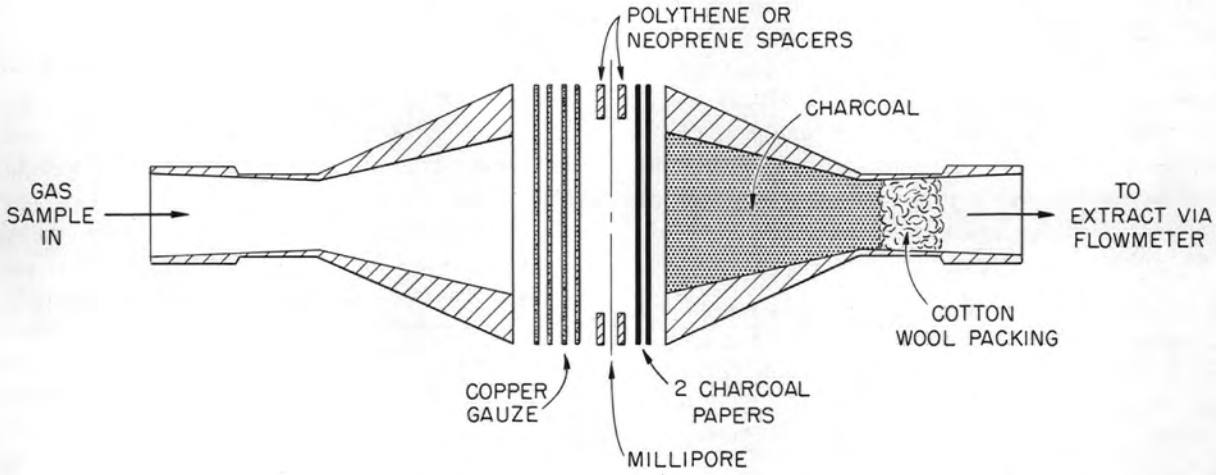

Fig. 3.23. Diagram of May Pack Used for Measuring Distribution of Iodine Forms in Gas Streams.

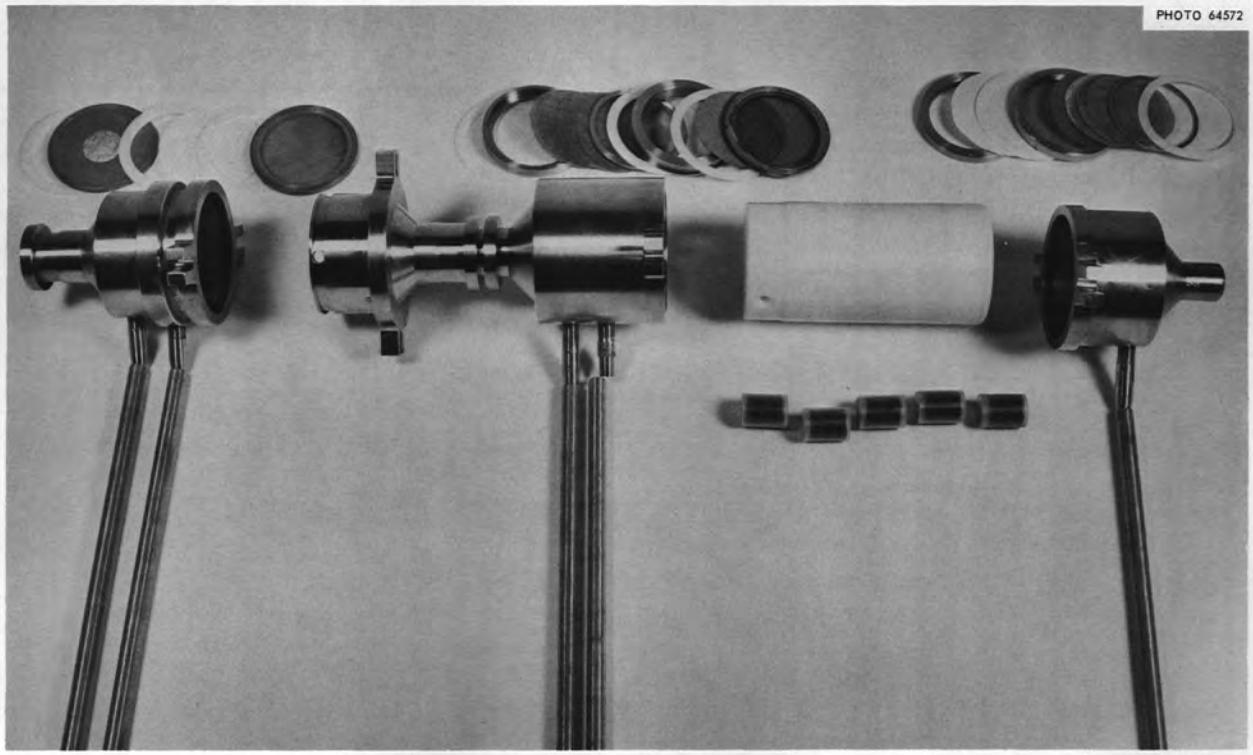

Fig. 3.24. ORNL Version of May Pack.

replaced with silver-coated copper screens because of difficulty experienced in obtaining reliable removal of molecular iodine by copper screens, and eight of these are presently being used. The principal innovation of this version, however, is the use of diffusion tubes to aid interpretation of data obtained with the May Pack. Side streams withdrawn at a rate of 100 cm^3/min and passed through silver-coated $\frac{3}{8}$-in. copper tubing 5 to 10 ft in length before and after individual components of the pack permit evaluation of the efficiency of each component in removing iodine vapor or small particles. The exit stream from the diffusion tubes passes through a filter and charcoal sampler to collect particles and vapors that do not deposit in the tubes.

Use of a series of charcoal cartridges instead of a single bed permits a determination of the distribution of poorly retained iodine compounds, presumably organic iodide vapors, discussed in a later section.

A "May Pack Cluster" designed to procure a series of 12 samples is shown in Fig. 3.25. Iodine-collecting materials are mounted in metal cylinders in the order indicated above, as used by Collins, but not necessarily in the same number of each. Silver-plated screens are used for the reason previously cited. Vacuum can be applied to each of the May Packs in turn by means of a sequential valve. The 12 May Packs are enclosed by a stainless steel shell to protect them from exposure to the atmosphere of the containment shell. In order to operate the cluster at temperatures expected in steam runs, a valve suitable for operation at 300°F will be substituted for the presently used valve, which is limited to a maximum of about 100°F.

3.6 ORGANIC IODINE COMPOUNDS

Radioiodine combined in a form now believed to be organic iodine compounds has been observed in the aerosol formed by heating both lightly irradiated and highly irradiated uranium and in that produced by use of a radioisotope source under a variety of conditions (*45*). The fraction of radioiodine of this form observed in various experiments has varied from a few tenths of a percent to ~30%. Elemental iodine is comparatively easily removed from gas streams, but the organic iodine compounds, being less reactive, are more difficult to remove and activated charcoal is the only material presently known to be capable of trapping them with a reasonable degree of efficiency. A knowledge of the identity of such compounds and of their behavior would be of considerable value in devising better iodine trapping systems and in predicting the probability of escape of iodine in that form in reactor accidents. It should be recognized that the biological half-life of these compounds has not been determined but it seems likely to be much shorter than that of elemental iodine. Establishment of a half-life value for iodine in this form may materially reduce requirements for their retention by reactor containment systems. Methods employed (*45*) for the separation and identification of the organic iodine compounds are discussed in the following sections, along with some information on their chemical behavior in aqueous systems and in a May Pack.

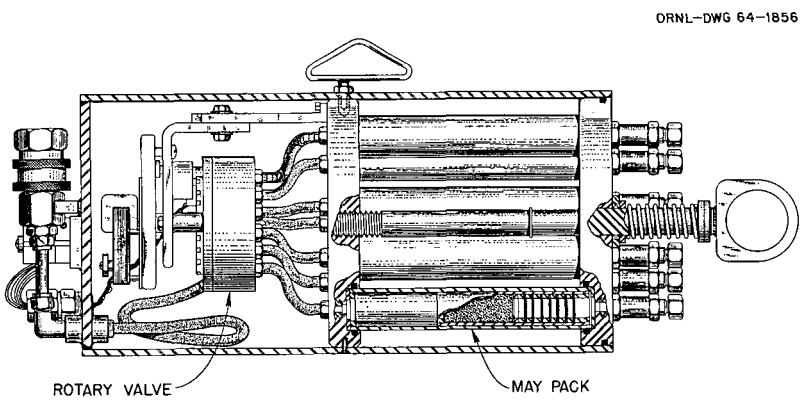

Fig. 3.25. May-Pack Cluster.

3.6.1 Solution Studies

Atkins and Eggleton (*45*) found evidence of two distinct types of iodine compounds (designated fractions A and B) present in an aerosol formed by oxidation of carrier-free $Na^{131}I$ at 400°C in air purified by passage through a charcoal trap at –78°C. Elemental iodine and particulate matter were first removed from the effluent air stream, and it was then bubbled through dilute caustic or sulfuric acid solutions. About 95% of the iodine activity was removed from the air stream by either reagent, mostly in the first bubbler in the train.

When the bubbler solutions were equilibrated with benzene, fraction A extracted into the organic phase with a high partition coefficient and was not back-extracted with NaOH solution. Fraction B, which was apparently more reactive than fraction A, extracted into benzene with a partition coefficient between 1 and 5 and reacted irreversibly with NaOH to give a form insoluble in benzene. Approximately equal quantities of fractions A and B were found in the bubblers.

Oxidation of the mixed fractions with ceric sulfate resulted in the loss of benzene solubility. Sulfur dioxide reduction, with added iodide carrier, followed by oxidation of the carrier to iodine, gave a form of the compound insoluble in benzene. Coprecipitation studies showed that approximately 5% of the mixed activities was carried on the precipitate in the stepwise precipitation of silver iodate. The mixed activities were carried quantitatively on silver iodide following reduction with sulfur dioxide.

3.6.2 Gas-Liquid Chromatography

A gas chromatography apparatus incorporating both a katharometer (apparatus for measuring thermal conductivity of gases) and a radioactivity detector (gamma-scintillation counter) was used to identify the compounds in fraction A. Columns 2 m long and of 6 mm internal diameter containing silicone oil or dinonyl phthalate on Celite as the stationary phase were used with helium as carrier gas. Flow rates of approximately 50 cm^3/min were maintained with column temperatures of 40 ± 1°C. Samples were collected by passing the effluent air stream from the iodine generator through a liquid-oxygen-cooled trap after removal of molecular iodine, particulate material, and fraction B, the last by passing the air through a magnesium perchlorate drying tube. Fraction B has apparently not been studied.

Examination of fraction A showed it to consist of several compounds. The radioactivity detector gave evidence for seven compounds, with the radioactivity in the first (peak 1) comprising about 90% of the total. Since the retention volume for stable methyl iodide obtained using the katharometer was found to be the same as that of peak 1, the identity of this compound was confirmed as methyl iodide.

Table 3.3 contains the measured retention volumes for peaks 1–7, together with those obtained for the series of alkyl iodides arranged in order of their boiling points. The good agreement obtained should not be regarded as final confirmation of the identity of peaks 2–7, but taken in conjunction with the known identity of peak 1, the evidence

Table 3.3. Retention Volume (Uncorrected for System Dead Space) for Simple Alkyl Iodides and for Peaks 1–7 of Fraction A. Silicone oil (15 wt %) on Celite. Temperature 40 ± 1°C. Flow rate 48 cm^3/min (*45*)

Material	Boiling Point (°C)	Retention Volume, V_R	Peak from Organic Fraction	V_R
Methyl iodide	42.5	214	1	214
Ethyl iodide	72.2	424	2	422
Isopropyl iodide	89.5	690	3	680
n-Propyl iodide	102.4	941	4	897
tert-Butyl iodide	100 (d)	962	5	979
sec-Butyl iodide	117.5	1614	6	1750
n-Butyl iodide	131	2370	7	2430

strongly suggests that these substances are the higher alkyl iodides shown in the table.

Oxidation of lightly irradiated uranium gave rise to a compound having the same retention volume on a silicone oil column as methyl iodide. A more accurate comparison of the retention volume of this compound with that for methyl iodide was made by a new method, as follows. A mixture of the radioactive species and an inactive sample of methyl iodide was passed onto the chromatography column, and the peak as indicated by the katharometer was split into two approximately equal parts, each of which was collected in a liquid-nitrogen-cooled trap. The masses of the approximately equal parts were determined from the peak areas obtained on passing each part separately through the column again. The peaks on emerging from the column were absorbed in small charcoal-loaded traps, and their activities were measured. From these figures the specific activity of each part was calculated. These were found to agree within 1%. The column had an efficiency of approximately 1000 theoretical plates for methyl iodide. Calculation shows that for a column of this efficiency, a difference in the retention volumes of the active and inactive species of only 0.1% would lead to a difference of more than 5% in the specific activities of the two halves. Consequently, the identity of this compound as methyl iodide was confirmed.

3.6.3 Molecular Weight Determination

The molecular weight of the major constituent of fraction A was determined by a modification of the classical effusion method. For the gas effusing through a hole whose diameter is small compared with the mean free path of the gas, the rate is proportional to the pressure and inversely proportional to the square root of the molecular weight. In the apparatus constructed the gas was allowed to effuse through a small hole in a platinum diaphragm mounted in the stopcock of a flask into a high vacuum. The flask was placed on the crystal of a gamma-scintillation counter, and the fraction of the radioactivity remaining after various effusion times was measured. The amount remaining decayed exponentially with time, the half-life being proportional to the square root of the molecular weight. A calibration curve was obtained for the apparatus with ^{85}Kr and ^{133}Xe. Using this calibration curve, the measured value of the molecular weight of peak 1 in fraction A was found to be 152. Pure methyl iodide was labeled with ^{131}I by an exchange technique, and the measured value of its molecular weight was found to be 150 (theoretical value 146), thus confirming the identity of peak 1 as methyl iodide.

3.6.4 Retention of Methyl Iodide by May Pack Components

Following the identification of methyl iodide as the major constituent of fraction A, Atkins and Eggleton (*45*) studied the behavior of pure methyl iodide labeled with ^{131}I in a May Pack. The results, summarized in Table 3.4, show that, with a flow rate of 10 liters/min, all the methyl iodide was retained by the pack, with 92% of it being collected by the charcoal bed.

Table 3.4. Retention of Methyl Iodide on May Pack Components (*45*)

May Pack Component (in order of assembly)	% Retention of Methyl Iodide
1. Copper gauze 1	1.5
2. Copper gauze 2	1.7
3. Wiggins Teape charcoal paper	1.2
4. Whatman ACG/B charcoal paper	3.6
5. Charcoal pack (20 g)	92.0

3.7 WET CHEMICAL METHODS OF IODINE FRACTIONATION

The distribution of iodine between water and organic solvents and other wet chemical tests have been used to determine valence states of iodine. This technique has not been widely applied in fission-product-release experiments. Its limitations are discussed below, in Sect. 3.7.3. Three investigations of this type are included here.

3.7.1 The Chemical State of Iodine Released into Steam (*78*)

The chemical states of iodine released from metallic uranium specimens irradiated over the range 2.0×10^{14} to 5.4×10^{19} *nvt* and from UO_2 powder irradiated up to 2.0×10^{16} *nvt* were studied with fuel temperatures in the range 1000 to 1350°C, using the apparatus shown in Fig. 3.26.

In this apparatus, steam passes over the inductively heated fuel sample and carries released fission products into the condenser. At the completion of an experiment, the boiler and condenser aqueous solution and the bubbler solutions are individually analyzed using wet chemical procedures. The distribution of iodine in the various traps depends on the boiling and reflux rates within the heated cell; however, in general, more than 90% of the released iodine is collected along with the condensed steam in the condenser flask.

Typical results for the release of iodine from metallic uranium are listed in the second column of Table 3.5. The majority of the iodine (av 84%) was found in the reduced state, while the remainder was primarily elemental; less than 1% was in the iodate and periodate states. Essentially no difference in the iodine chemical states was found by varying the pH of the water initially charged to the cell between 5.9 and 6.8. Successive filtrations through 1.2-μ, 0.45-μ, 0.3-μ, 100-mμ, and 10-mμ Millipore filters showed that none of the iodine was associated with particles larger than 10 mμ.

Table 3.5. Average Percentages of Chemical States of Iodine

State	Fraction in Each State (%)	
	U-H_2O System	UO_2-H_2O System
I^0	15	80
I^-	84	10
$I^{5+,\ 7+}$	<1	<0.5
Other	1	10

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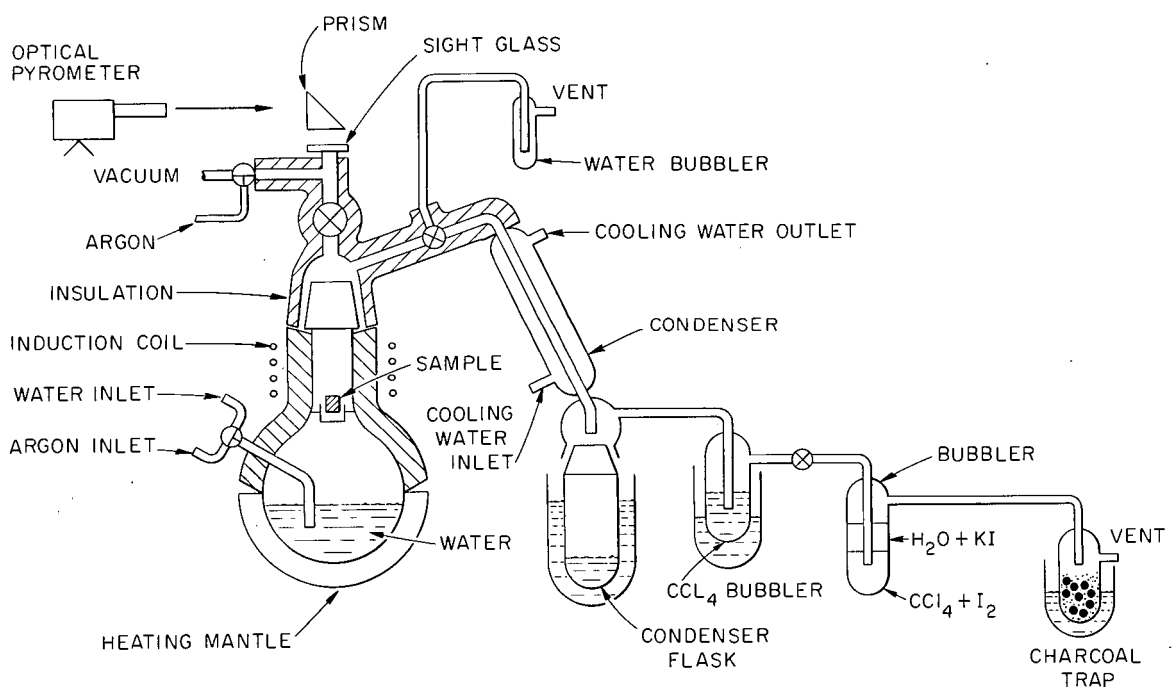

Fig. 3.26. Apparatus for Determining the Chemical Form of Iodine Released from Irradiated Fuel in a Steam Atmosphere.

In contrast to the metallic uranium experiments, an average of 80% of the iodine released from UO_2 into steam was found to be elemental. These results are shown in the third column of Table 3.5. Filtration experiments similar to those mentioned above also showed that none of this iodine was associated with particles larger than 10 mμ, but dialysis membrane retention studies indicated that up to 8% of the iodine may have been attached to smaller particles.

During the course of an experiment the heated uranium metal reacts with the steam to form UO_2 and evolve H_2 and fission products. The initial reaction rate is rapid, and a plot of rate vs time is a parabolic curve. This is followed by a linear reaction rate, during which time an oxide layer of constant thickness is maintained on the surface. The release from the UO_2 powder samples is primarily by diffusion, perhaps enhanced by the lattice expansion resulting from the conversion to $UO_{2.2}$, the thermodynamically stable oxide under the prevailing conditions. Only a small quantity of H_2 is formed by this oxidation process; this is added to the small quantity resulting from the dissociation of steam at the fuel temperatures. Basically, then, the iodine is released along with other fission products from a UO_2 surface in both the metallic uranium and UO_2 studies, the major difference being the quantity of H_2 in the atmosphere surrounding the fuel. Most of the iodine is dissociated to atomic iodine (I) at the temperatures used (1000 to 1350°C). Although atomic I is quite reactive because of its unpaired electron, highly exothermic gas phase reactions with atoms of other fission products to form simple molecules are precluded, since the energy of reaction goes to a vibrational mode and the molecules dissociate. Third-body collisions are required to stabilize the products. Reactions with H_2 should proceed, however, since one hydrogen atom removes the energy of reaction in the form of the translational energy.

The importance of reactions between H_2 (resulting from a metal-water reaction) and iodine was confirmed by introducing H_2 into the release cell during a UO_2 release experiment. The final iodine chemical states were similar to those observed in experiments with metallic uranium, and again most of the iodine was in the reduced state. The important reactions are shown below.

$$2H_2O + U(F.P) \rightarrow UO_2 + 2H_2 + F.P. , \qquad (1)$$

$$H_2 + 2I \rightleftharpoons 2HI , \qquad (2)$$

$$2I \rightleftharpoons I_2 , \qquad (3)$$

$$2I + H_2O \rightleftharpoons HIO + H^+ + I^- , \qquad (4)$$

$$3IO^- \overset{\Delta}{\rightleftharpoons} IO_3^- + 2I^- . \qquad (5)$$

The fission products are released by reaction (1). The released iodine reacts with the H_2 to form HI by (2). The equilibrium expressed in reaction (3) is far to the left, so that little molecular iodine is present within the cell. A small quantity of iodine reacts according to equation (4), but the hydrolysis is very small (see Sect. 2.2.1). Some disproportionation of IO^- takes place at elevated temperatures in solution, giving rise to iodates and periodates via reaction (5). These latter species account for less than 1% of the iodine. Some of the iodine (about 1% from uranium and up to 10% from UO_2) is not in any of the above-mentioned states and is probably in the form of organic iodides. The origin of these species is under study.

The equilibrium between H_2, I, and HI was calculated for the iodine concentration range 10^{-8} to 10^{-14} mole/liter, H_2 partial pressures ranging from 1 to 10^{-5} atm, and temperatures between 298 and 2000°K. The results of these calculations are shown in Fig. 3.27. In the experiments reported here, the bulk average cell temperature was between 500 and 1000°K and the H_2 partial pressure was approximately 10^{-3} atm. Referring to Fig. 3.27, from 75 to 95% of the iodine should be converted to HI under these conditions. All of the metallic uranium experimental results to date fall within this range.

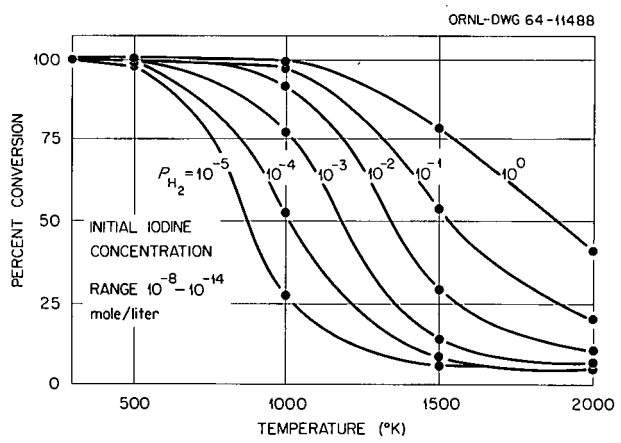

Fig. 3.27. Hydrogen Iodide Equilibrium.

3.7.2 Solubility Behavior of Volatile Iodine Compounds (*79*)

It has been demonstrated by diffusion tube analysis that iodine sources nominally in the elemental form can have a small fraction of the iodine existing in compound forms which are rather difficult to remove from air under certain conditions. At least three vapor species have been distinguished, and workers in England (*45*) have identified one of the forms of gas-borne radioiodine as methyl iodide. Figure 3.28 illustrates the apparatus used to perform an experiment to determine whether methyl iodide is contained in the A fraction of iodine sources prepared by the palladium iodide method and also to investigate the applicability of differential solubility as a tool to characterize the volatile iodine compounds. Three experiments were performed. In the first two experiments the A fractions were separated from iodine sources by use of different techniques, were placed in independent containers, and were introduced into the experiments. In the third, pure methyl iodide vapor, tagged with radioactive methyl iodide (tagged methyl iodide obtained from Volk Radiochemical Co., Skokie, Ill.; produced by the exchange of CH_3Br and $Na^{131}I$ in acteone), was introduced into the experiment. A gas sweep, containing the volatile iodide compound, was passed through a silver-lined tube and a rubber tube prior to entering the separatory funnel containing nonradioactive methyl iodide and water. In the first two experiments, the distribution coefficient between the two phases was about 55 in favor of methyl iodide. This corresponds approximately to the expected distribution considering the solubility of methyl iodide in water. The distribution coefficient in the third experiment using tagged methyl iodide was comparable. The results, presented in Table 3.6, indicate that the mixture of iodine vapor species emanating from the iodine vapor source includes methyl iodide or an iodine compound of similar solubility.

Table 3.6. Sorption and Solubility Behavior of Volatile Iodine Compounds

Sample Identity (see Fig. 3.28)	Relative Amount of Activity[a]		
	Exp. 1[b]	Exp. 2[b]	Exp. 3[c]
A – rubber tube	0.39	0.019	0.008
B – silver tube	0.92	0.023	0.001
C – rubber tube	0.69	0.32	0.92
D – methyl iodide	1.00	1.00	1.00
E – water	0.02	0.015	0.017
F – rubber tube	0.001	0.014	0.011
G – charcoal	0.008	0.053	0.073
H – charcoal	0.007	0.003	0.000
Distribution Coefficient (CH_3I/H_2O)	49.6	61.9	46.2

[a]Data in each column normalized to methyl iodide sample = 1.00.

[b]Experiments using A fraction consisting of iodine compounds volatilized at −70°C.

[c]Experiment using ^{131}I-labeled CH_3I.

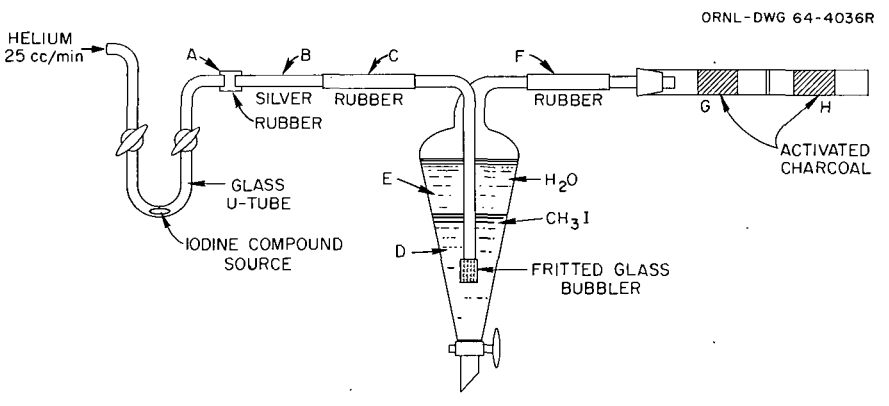

Fig. 3.28. Apparatus for Investigating Solubility of Selected Iodine Vapor Species.

3.7.3 Identification of Chemical Forms of Iodine by a Liquid Contactor Method

One method used (*80*) to determine the chemical state of radioiodine is that which makes use of equilibration of tracer iodine between two solvents such as water and carbon tetrachloride (CCl_4). The accepted value for the distribution of elemental iodine is approximately 80 parts in CCl_4 to 1 in water, while I^-, IO_3^-, and IO_4^- concentrate in water on equilibration. It appears, therefore, that the distribution of iodine between solvents should afford a method for determining whether iodine is present as I_2 or in another valence state. However, this method, as it is currently used, gives results of questionable value in fission-product-release experiments because: (1) organo-iodine compounds may extract into CCl_4, for example, and become identified as I_2; (2) submicron particles carrying iodine may possibly distribute between solvents without reference to the state of oxidation of the radioiodine; (3) a rapid change in oxidation state may occur due to reaction with impurities in the solvents, especially at very low concentrations of iodine; and (4) rapid isotopic exchange of radioiodine in one state with that in another previously added as a carrier to the solvent systems is possible. Information derived without due regard for these various possibilities is likely to be in error.

Studies conducted to date have not quantitatively defined any of the above-mentioned effects, but preliminary tests of the liquid-contactor method were performed in an effort to supplement data obtained by use of the modified May Pack and diffusion tubes.

An experiment was performed in which ambient air carrying a mixture of ^{131}I and ^{127}I was introduced into a stainless steel tank. After the aerosol had aged for 5 hr and the pressure in the tank had been reduced from 15 to 2.5 lb, a gas stream from the aerosol tank was removed and split into two parts. One stream was directed to the CCl_4 system, and the other to the CH_3I system, as shown in the block diagram, Fig. 3.29, which also gives the observed distribution data.

In the CH_3I system, the gas stream passed through eight silver screens to remove I_2 and any HI that might be present. The screens removed about 81% of the airborne iodine. The distribution of iodine among the eight screens suggests that part of the deposited iodine was attached to small particles. There seems to be little doubt that interpretation of distribution data would be simplified by use of filters having a high efficiency for removal of small particles. The gas was then bubbled through CH_3I (maintained at approximately 6°C), which extracted most of the iodine not removed by the screens. This solvent should remove organic iodine compounds, such as CH_3I, but, as mentioned above, part of the 18.8% of the iodine found in this scrubber may have been due to particles. After passing through the CH_3I, the gas flowed through a water scrubber and a charcoal trap.

In the CCl_4 system, the gas was first scrubbed with CCl_4, which was cooled to approximately 6°C. This treatment removed I_2 and, presumably, organic iodine compounds as well. Nearly all

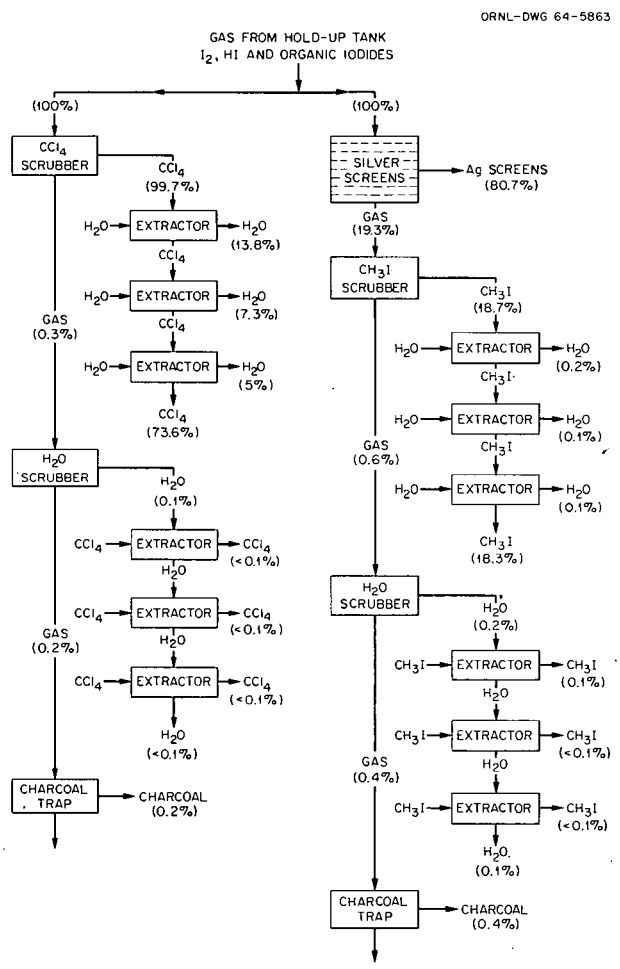

Fig. 3.29. Distribution of Iodine in Water and Organic Solvents.

(99.8%) of the airborne iodine was removed by this scrubber. The CCl_4-scrubbed gas was bubbled through water to remove HI and any other water-soluble iodine compounds present and then passed through a charcoal trap.

A significant amount (~26%) of the total activity in CCl_4 was extracted by three water washes. This indicated the presence of nonelemental iodine, which could be due to a change in state of some of the original elemental iodine in solution or to water-soluble iodine compounds, such as might be carried by particles trapped in the CCl_4. The distribution of iodine activity between the three successive backwashes and the CCl_4 indicated, however, that little, if any, of the iodine in the CCl_4 was present as HI. Very little water-soluble iodine was found in the CH_3I scrubber.

It appears that part of the radioiodine that remained airborne for more than 5 hr and was extracted from air by CCl_4 was later in a form which was water-extractable. This fraction was not found in the CH_3I and, therefore, must have been retained on silver screens. More data will be required to determine the effect of iodine concentration on the fraction of iodine in CCl_4 converted to the extractable form and, in general, the value of distribution data for characterizing the chemical form of gas-borne radioiodine.

3.8 SUMMARY

Several facilities in the U.S. are currently providing information on the release and transport behavior of iodine from irradiated fuels. These range in size from laboratory-scale equipment to the Nuclear Safety Pilot Plant, which is equipped with a 1350-ft^3 containment vessel. A facility with similar capabilities is in operation at Windscale, but conditions employed in experiments there are pertinent to gas-cooled reactor accidents (CO_2-CO coolant) rather than water-cooled or water-moderated reactors, which are currently more important in American reactor technology. The construction of new and larger facilities, including the Loss-of-Fluid Test at the National Reactor Testing Station, reflects the present concern for improving the realism of accident simulation. In addition to in-pile and out-of-pile tests under loss-of-coolant accident conditions, experiments on release and transport of fission products under transient conditions are under way.

A number of methods of furnishing radioactive iodine sources are described in the literature. In nearly all cases, a small but significant fraction of the iodine produced is found to be in the form of organic iodides. It is especially important to provide better information on the fraction of released iodine that is converted to penetrating organic iodides and to gain understanding of the mechanisms responsible for their production. It is also considered essential that iodine releases be made in conjunction with production of particles from overheated fuel and cladding in order to study the transport of iodine and that the atmosphere be representative of that expected in accidents. Steam-air mixtures are currently being emphasized, because of the above-mentioned importance of pressurized- and boiling-water reactors, and investigations of the effect of impurities in the water on iodine behavior remain to be performed.

Considerable progress has been made in the development of techniques and equipment for sampling aerosols containing radioactive iodine and for determining its distribution among the various possible forms. Some of the newer techniques, such as diffusion tubes, fibrous filters, and remotely operable samplers, have not been properly evaluated, but rapid progress in this direction seems to indicate that satisfactory sampling and fractionation methods under ordinary conditions are, or soon will be, available.

Much work remains to be done in order to determine the applicability of sampling and iodine fractionation techniques under conditions such as temperatures higher than 100°C with steam or steam-air atmospheres.

4. TRANSPORT OF FISSION-PRODUCT IODINE AND NATURAL REMOVAL PHENOMENA

Natural phenomena affecting the transport behavior of released fission-product iodine are considered in this section. Induced removal and trapping methods are treated in Sect. 5.

A great deal of effort has been directed to the problem of defining the probable behavior of iodine in reactor accidents. Although some allowance is made (*4–5*) for deposition of iodine in a containment shell, it is generally believed that the deposition factor (0.5) employed in TID-14844 and in many hazards summary reports may be quite conservative and that a much higher attenuation factor could be justified by availability of more complete information on iodine deposition rates in containment systems. The complex chemical behavior of iodine demonstrated in the previous section and the various uncertainties involved in defining accident conditions combine to make prediction of the extent of iodine deposition such a difficult problem that it may be necessary to accept a considerable degree of uncertainty in the iodine deposition factor. Because of this fact, engineered safeguards discussed in Sect. 5 may be required to supplement iodine attenuation in containment systems by natural removal methods. Available information, including some data obtained under conditions that are unlikely to prevail in reactor accidents, is summarized here in an effort to provide a reasonably clear picture of the present state of knowledge in this field rather than final answers to the problems involved.

4.1 TRANSPORT ON PARTICLES

Following its release from the fuel, fission-product iodine will be transported in the primary system and then into the containment volume. Part of the released iodine will deposit in close proximity to the fuel and is of little consequence to the overall hazard. The remainder, which may be in the vapor state or attached to particulate matter, is transported out of the high-temperature region, where it may condense, agglomerate and settle, or, in the case of volatile vapors and fine particles, remain airborne for extended periods of time. The ultimate deposition of the iodine depends not only on its chemical and physical properties but also on the properties of the particles to which it may become attached.

High concentrations of vaporized fuel particles in conjunction with the static gas surrounding the heated fuel element may result in formation of large particles which will settle out in a reactor pressure vessel and carry much of the released iodine with them.

4.1.1 Sorption on Particles

As Chamberlain (*81*) has pointed out, particles may remove iodine from the atmosphere by physical adsorption, a reversible process, or by chemisorption, which is irreversible (at least at normal room temperatures). He adapted Fuchs' equation (*82*) for evaporation from small droplets to give the following equation for the rate of removal of iodine from the atmosphere by the irreversible process:

$$\frac{1}{C}\frac{dC}{dt} = \frac{-4\pi r n D}{(D/rN\alpha) + (r/r + \Delta)}, \qquad (1)$$

where

t = time, sec,

C = concentration of iodine in air,

r = radius of particles, cm,

n = number of particles per cm^3,

D = diffusion coefficient of iodine in air,

R = gas constant,

T = absolute temperature,

M = molecular weight of iodine,

$N = (RT/2\pi M)^{1/2}$ (N has the value 3913 cm/sec at 20°C for I_2),

α = accommodation coefficient or sticking probability of iodine on particles,

Δ = mean free path of iodine molecules in air ($= 4.19 \times 10^{-6}$ cm at 20°C).

For very small particles ($r << \Delta$), this equation reduces to:

$$\frac{1}{C}\frac{dC}{dt} = -4\pi r^2 nN\alpha \ . \qquad (2)$$

For particles of about micron size, $r >> \Delta$ and $r/r + \Delta = 1$. The first and second term in the denominator on the right hand side of the more general equation above may then be dominant, depending on whether $D/rN\alpha$ is greater or less than unity. If $r = 10^{-4}$ cm, $D = 0.080$ cm^2/sec, and $N = 3913$ cm/sec, the condition $D/rN\alpha > 1$ is equivalent to $\alpha < 0.2$. Thus, if the accommodation coefficient is less than 0.2, its magnitude will determine the rate of adsorption of iodine. For 10-μ particles α must be less than about 0.02 for the same to be true, and in general the larger the dimensions of the surface the less important is α in determining the rate of uptake.

In Table 4.1 is shown (*81*) the theoretical rate of uptake of iodine for various aerosols according to Eq. (1), with values of the half-life in the gas phase calculated for $\alpha = 1$ and for $\alpha = 10^{-4}$.

In practice, the value of α is likely to depend on the nature of the surface of the particle, and in particular on the amount of iodine adsorbed there. Also shown in Table 4.1 is the amount of iodine in μg per m^3 of air which would give a monolayer coverage on the particulate aerosols.

Megaw and May (*83*) estimated accommodation coefficients of 4.7×10^{-3} and 1.2×10^{-3} for iodine adsorption on Aitken nuclei in two tests performed in the Pluto reactor shell. They state that in small-particle studies accommodation coefficients of unity are generally assumed, although Chamberlain states (*81*) that chemisorption on such particles has not been observed. Consequently, they interpreted the low values obtained in these experiments, together with the observation that radioiodine adsorbed on small particles did not appear to interchange with inactive iodine, as indicating that irreversible chemical adsorption of iodine occurred with only a small fraction of the nuclei present in the gas. This theory is also supported by data obtained in the same experiments showing that the fraction of iodine attached to particles dropped off more rapidly than did the number of Aitken nuclei. The authors state that the data available did not permit a decision as to whether the particulate fraction of the iodine is the result of attachment of iodine atoms to a relatively small proportion of chemically suitable nuclei or the result of attachment to an even smaller number of much larger particles. They favored the first alternative.

Table 4.1. Theoretical Rate of Uptake of Iodine on Particles in Air (*81*)

	Aerosol Type and Number				
	Aitken Nuclei		Lead Fume	Water Fog	Raindrops
	I	II	III	IV	V
Particle radius, μ	0.01	0.1	1	10	500
Particle density	3	3	9	1	1
No. per cm^3	10^5	10^4	10^3	10^2	10^{-4}
Terminal velocity, cm/sec	4×10^{-5}	6×10^{-4}	7×10^{-2}	1.2	400
Mass per m^3 of air, mg/m^3	1.2×10^{-4}	1.2×10^{-2}	38	420	52
Surface area per m^3 of air, cm^2/m^3	1.2	12	120	1200	3.1
Mass of I_2 to give monolayer, μg/m^3	0.4	4	40		
Half-life of I_2 in gas phase, min					
$\alpha = 1$	2.4	0.33	0.13	0.12	480[a]
$\alpha = 10^{-4}$	2.4×10^4	2400	240	24	2000[a]

[a]These figures include allowance for the "ventilation factor" which is the increased rate of transfer caused by the fact that large droplets move rapidly through air.

Chamberlain (*81*) reported that, in experiments performed by Eggleton and Cousins, when uranium oxide particles with an effective diameter of 0.3 μ (7 mg/m^3) produced by burning uranium were introduced into a box containing 0.09 μg of iodine vapor per m^3, equilibrium was established in less than 10 min when about half of the iodine was adsorbed on particles. When air was passed through a filter paper on which particles were collected, most of the iodine was removed, indicating reversible adsorption. Experiments with "Magnox" oxide (Magnox contains about 99% Mg and 1% Al) were said (*81*) to yield confusing results, with the fraction of the total iodine adsorbed reversibly varying erratically. A maximum adsorption of 1.5 μg of I_2 per mg of oxide was achieved, corresponding to about 0.1% of a monolayer coverage. A drastic reduction in rate of deposition of iodine on silver and other surfaces after the introduction of MgO particles was noted. In other experiments (*81*), also performed with Magnox oxide, the amount of iodine adsorbed on the oxide particles corresponded to only 1% of a monolayer with iodine concentrations (in air) of 5 μg/m^3 and 13 mg/m^3. The iodine appeared to be irreversibly adsorbed on the oxide at both iodine concentrations. The uptake of iodine on Magnox wire was found to be roughly proportional to the iodine concentration in the gas phase in the range 40 μg/m^3 to 60 mg/m^3, with more than a monolayer amount adsorbed at the latter concentration. The reason for the different behavior of the two materials might be accounted for by the fact that the wire had an MgO surface over a metal substrate, implying that the iodine vapor may penetrate the oxide coating and react with the metal.

Chamberlain also points out (*81*) that normal methods of sampling for iodine do not permit determination of the amount of iodine adsorbed reversibly on particles and that this becomes confused with the radioiodine vapor fraction or the iodine compound fraction, depending on the type of sampler used. He also states that both reversible and irreversible adsorption of iodine on particles will result in a much lower rate of deposition than for iodine vapor and that it would therefore be unwise to consider the rapid deposition of iodine observed in the Dido-Pluto experiments to be typical of all types of reactor accidents.

Craig (*62*) has summarized available information on interaction of particles and airborne radioactive materials, and he also reported the results of an investigation of the adsorption of sodium iodide vapor containing ^{131}I on iron oxide particles labeled with ^{59}Fe. He used the Goetz Aerosol Spectrometer to characterize the adsorption of NaI molecules on the iron oxide aerosol as a function of particle size with radionuclide-to-aerosol concentration ratios of 3×10^2 to 3×10^5 molecules per particle and with mean interaction times varying by a factor of 2.5. His conclusions were: (1) considerable quantities of the radionuclide became attached to the aerosol but the adsorption process was not irreversible; (2) variation of the experiment conditions over the above-mentioned range did not significantly affect the results; and (3) the adsorbed activity was proportional to a function $D(I)^Z$, where $D(I)$ is the aerosol diameter and $2 < Z < 3$, over the approximate size range 0.06 to 0.65 μ and possibly up to particle diameters of 1.2 μ.

The first conclusion is rather surprising considering the fact that NaI (reported boiling point 1300°C) undoubtedly has a very low vapor pressure at room temperature. The author apparently failed to examine the possibility that impurities in the stream of argon that was used to transfer the radionuclide vapor from the furnace where NaI was heated into the aerosol chamber might have produced some molecular iodine vapor or other volatile iodine compounds. Therefore, his conclusion concerning the reversible adsorption of NaI molecules on iron oxide particles seems questionable.

4.1.2 Transport on Fuel and/or Cladding Particles

Many experiments on release and transport of fission products from irradiated fuels have been performed, but there seems to be little data in the literature that will permit a valid comparison of transport of iodine liberated from clad and unclad fuel in air or steam-air mixtures. Consequently, no attempt will be made in this section to define the effect of particles of vaporized cladding materials on iodine transport.

Data (*84*) on the amount of iodine liberated by heating Al-U alloy fuel that was carried by particles is given in Table 4.2, as a function of fuel temperature in different atmospheres.

Table 4.2. Transport of Fission-Product Iodine Released from Aluminum-Uranium Alloy (*84*)

Fuel Temperature (°C)	Percent of Iodine Inventory Transported to Filters by Particles		
	Air	Helium	Steam-Air[a]
700	2.8		0.3
800	4.5	1.4	0.1
900	4.2	1.5	0.7
1000	0.9	13.3	4.1
1100	3.9	17.4	3.9
1150	9.2		

[a]80% steam and 20% air, by volume.

The experiments conducted in helium and in a steam-air mixture show some evidence of increased transport of iodine on particles with increased temperature, but the air experiment data show too great a scatter to demonstrate a definite trend. In one experiment, where the fuel specimen was heated to 1100°C and 17% of the iodine was caught on the particulate filter, the particles were found to vary in size from 0.04 to 4 μ. The deposit on the filter contained zinc (an impurity in the fuel) and aluminum. When a helium stream was passed over heated alloy material in a quartz tube, a shiny deposit on the wall of the tube was observed and was identified as zinc. The deposit contained more than 90% of the released iodine and almost half the cesium but very little uranium. Diffusion tube studies of the aerosol produced in the steam-air experiment, after it had aged for 1 to 2 hr, showed that 95% of the iodine present was in the molecular form, while the other 5% was attached to particles with a calculated diameter of 26 A. These figures correspond to 17% and 10% of the original fuel iodine inventory.

The transport of iodine liberated by heating UO_2 fuel, especially stainless-steel-clad specimens, has been examined in both in-pile and out-of-pile experiments. Data (*40*) on the size of small particles produced by heating clad UO_2 in flowing helium with fission heat in the Oak Ridge Research Reactor were obtained by using the stainless steel exit tube from the furnace as a diffusion device. The particles with which iodine (as well as other fission products) was associated were found to have an average diameter of about 20 A, and variable amounts of the iodine (3 to 56%, average 23%) were apparently carried by these small particles. Similar results were obtained when pyrolytic-carbon-coated uranium carbide particles in a graphite matrix were partially destroyed by burning in the ORR in-pile facility.

Out-of-pile studies (*85*) of the aerosol produced by melting trace-irradiated, stainless-steel-clad UO_2 fuel in air after a 2- to 3-hr aging period showed that 80% of the airborne iodine was in the molecular form, about 15% was attached to particles with an apparent diameter of 26 A, and 5% was associated with particles 108 A or larger in diameter. These values correspond approximately to 20%, 4%, and 1%, respectively, of the original iodine inventory of the fuel.

4.1.3 Agglomeration and Settling of Particles

Data indicating that significant quantities of radioactive iodine released in nuclear accidents may be attached to particles are discussed in the previous sections. It becomes of interest to examine the rate at which such particles might disappear from the atmosphere of a reactor containment shell. Stokes' law gives the velocity (in cm/sec) for the fall of small spheres in air as

$$V = \frac{2}{9}\, r^2 \rho g/\eta = 1.2 \times 10^6\, \rho r^2 ,$$

where r is the particle radius in cm, ρ is the particle density, η is the coefficient of viscosity of air, and g is the acceleration of gravity. This equation is said (*86*) to hold within 5% for particles in the 1- to 50-μ range. At the lower end of the scale, the calculated velocity for a particle with a density of 5 (approximate value for iron oxide) is 0.6 mm/sec, while the corresponding velocity for 10-μ particles is 6 cm/sec. When the aerosol is stirred, as by convection currents, the number concentration n of particles at a given time t is represented by the equation (*86*):

$$n = n_0\, e^{-vt/h} ,$$

where v is the velocity of fall of particles in a rectangular box of height h. This equation shows that the number concentration of particles and the rate of settling (in terms of the number of particles per second) decrease exponentially with time.

The differential equation governing the decrease in number of particles due to coagulation is (*87*):

$$\frac{dn}{dt} = Kn^2 .$$

This equation holds only for particles that are large compared with the mean free path l (10^{-5} cm in air at room temperature). For smaller particles, the Cunningham slip correction (*88*) must be applied and the corrected equation is:

$$\frac{dn}{dt} = K(1 + 0.9\ l/r)n^2 .$$

This shows that 0.1-μ particles coagulate 88% faster than 10-μ particles.

Since K has the approximate value 5×10^{-10}, it is obvious that coagulation rates are likely to be low for particle concentrations below $10^6/cm^3$.

Browning and Fontana (*89*) have calculated the theoretical rate of agglomeration of particles in the NS "Savannah" containment system. The authors chose to illustrate their method of calculation by use of the concept of iodine vapor agglomerating to form iodine particles of 0.3-μ diameter and calculated that 235 sec would be required for this to occur if all the iodine produced in the 69-Mw reactor during 600 days of operation were to be uniformly dispersed in the ship's containment system. Although the authors termed the assumption that iodine agglomerates into particles of pure iodine an artifice, the fact that it is utterly unrealistic to assume that iodine particles large enough to be retained by filters will exist might be overlooked by people unfamiliar with iodine behavior.

It should be recognized that the effect of high specific activity levels, such as may be expected in vaporized fuel particles produced in reactor accidents, on coagulation rates remains to be determined. Some efforts in this direction have been made, but the activity levels were too low to give significant results.

4.2 VARIATION OF AIRBORNE IODINE WITH TIME

4.2.1 Experiments in Reactor Shells

Deposition experiments in the Zenith, Dido, and Pluto reactor shells have been reported by Megaw and May (*83*) and by Croft and Iles (*90*). Their data showed changing deposition rates during the aging of an iodine aerosol released initially as molecular vapor. The initial airborne iodine concentrations in dry air were 0.01 to 14 $\mu g/m^3$ in the Dido and Pluto experiments and 0.3 to 0.8 $\mu g/m^3$ in the Zenith shell. The maximum iodine concentrations used were lower than possible accident conditions by a factor of 100 or more and the absence of vaporized fuel particles reduced the degree of accident simulation, but the results obtained are, nevertheless, of considerable interest. Iodine-132 with normal iodine carrier was used in all these experiments. The variation of ^{132}I concentration with time for six Pluto experiments is shown in Fig. 4.1. Experimental conditions for the Dido and Pluto experiments are given in Table 4.3. Detailed distribution data and Aitken nuclei counts are shown in Fig. 4.2 for

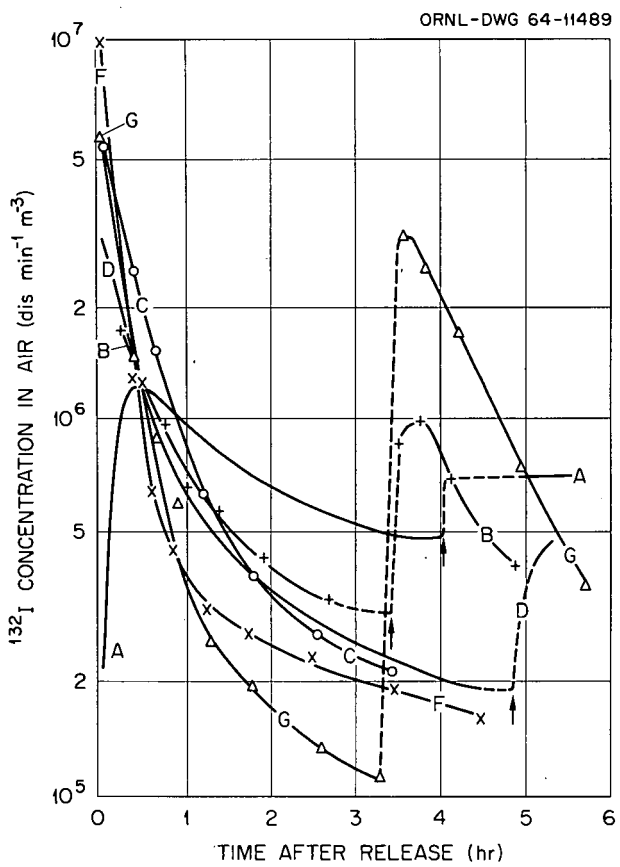

Fig. 4.1. Variation of ^{132}I Concentration with Time (*83*) (Corrected for Radioactive Decay from $t = 0$).

Table 4.3. Details of Experimental Releases (*83*)

Reactor	Run	Quantities of Iodine Released		Calculated Added Concentration		Maximum Measured Concentration (dpm/m^3) at Position 2	Remarks
		^{127}I (mg)	^{131}I (mc)	($\mu g/m^3$)	(dpm/m^3)		
Dido	1	7	26	1.0	8.2×10^6	4.7×10^6	Scrubbers operated
	2	1	31	0.14	9.8×10^6	2.8×10^6	
	3	7	31	1.0	9.8×10^6	2.9×10^6	
Pluto	A	0.09	28.2	0.013	8.9×10^6	1.45×10^6	
	B	0.95	22.9	0.14	7.2×10^6	1.75×10^6	
	C	99	30.6	14.1	9.7×10^6	5.4×10^6	
	D	99	20.1	14.1	6.4×10^6	3.0×10^6	
	E	7	2.7	1.0	8.5×10^5	9.3×10^5	Scrubbers operated
	F	9.9	48.9	1.4	1.5×10^7	1.0×10^7	
	G	10	39	1.4	1.2×10^7	5.5×10^6	

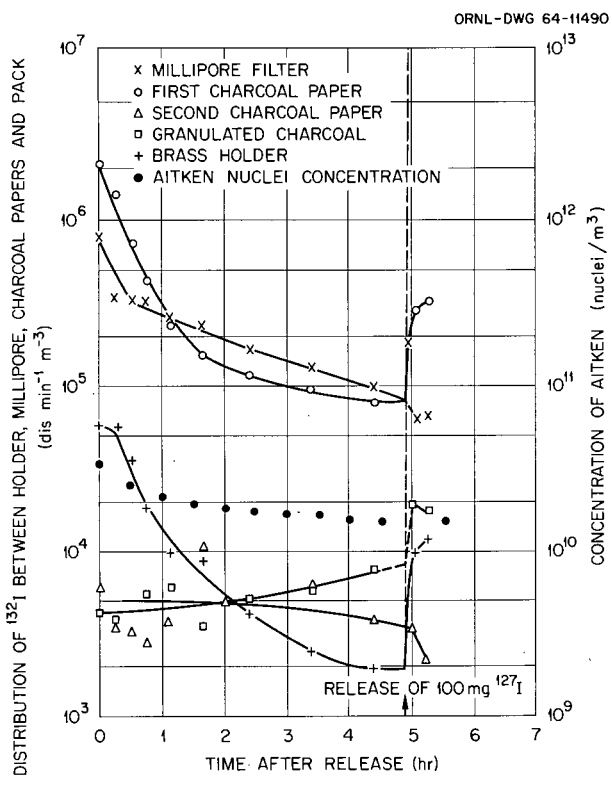

Fig. 4.2. Run D; Variation of ^{132}I Concentration with Time (*83*).

one experiment (run D) where the initial calculated iodine concentration was 14.1 $\mu g/m^3$. As shown in Table 4.3, the maximum measured concentrations were much less than the calculated concentrations (20 to 60%) except in one experiment. This was ascribed to the fact that about 40 min was required for uniform mixing of gases in the reactor shell under the prevailing experimental conditions. Consequently, part of the iodine diffused to the shell wall before mixing was complete.

The sharp breaks in several of the curves shown in Figs. 4.1 and 4.2 are due to liberation of quantities of ^{127}I vapor in the shell, 10 g in run G, and 100 mg in A, B, and D. The ^{127}I vapor obviously interchanged with part of the deposited ^{132}I and caused it to become airborne.

Two charcoal-coated filter papers were used following the Millipore filter in the May Pack in the Pluto experiments; Fig. 4.3 shows the ratio of iodine activity on the first paper to that on the second as a function of time. The ratio decreased rapidly and then became relatively constant at about 20 to 60 after $2\frac{1}{2}$ hr. The authors ascribed this to an unknown vapor compound of iodine, designated "compound X" that has subsequently been identified as organic iodides (see Sect. 3.6). The variation in the fraction of airborne iodine collected on Millipore paper is shown in Fig. 4.4. The data show that this fraction increased

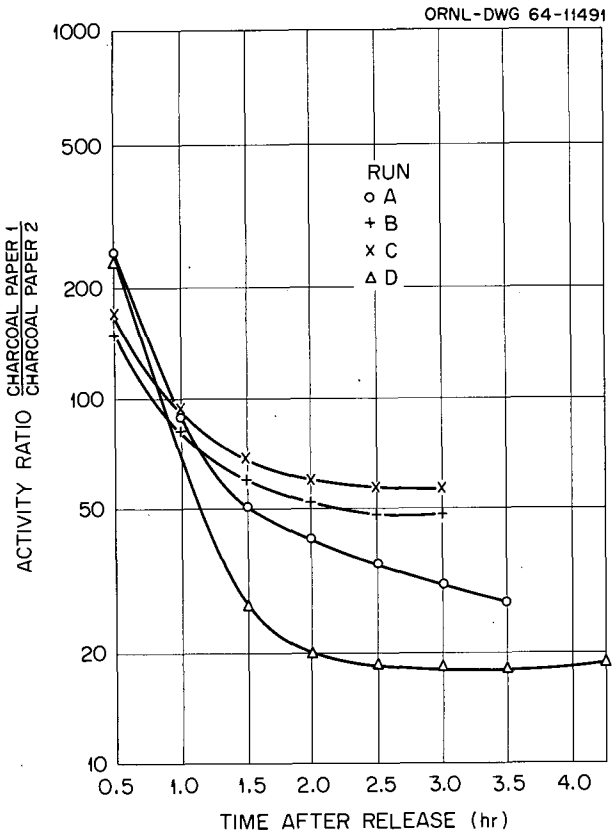

Fig. 4.3. Variation of the Ratio of Iodine Activity on the First Charcoal-Coated Filter Paper to That on the Second with Time (*83*).

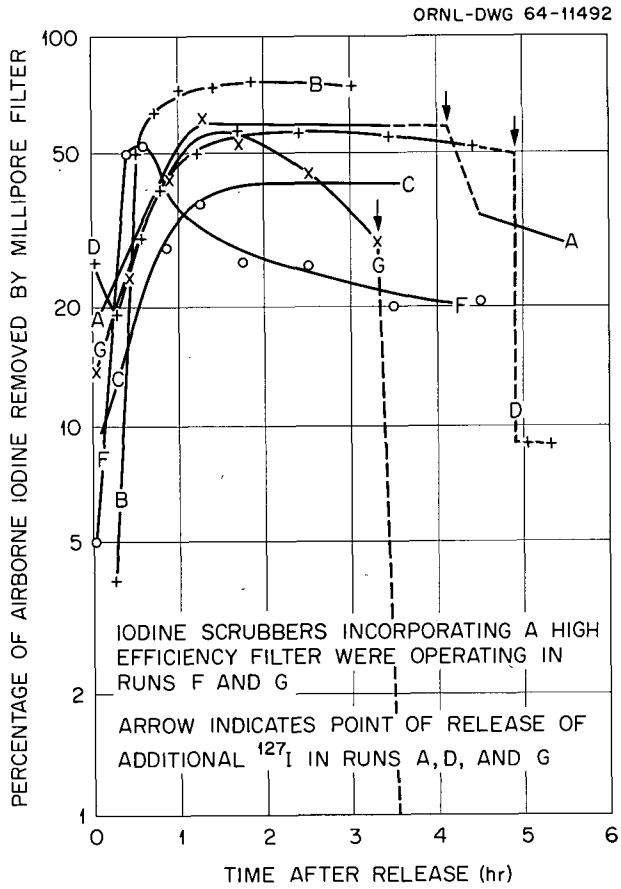

Fig. 4.4. Percentage of Total Iodine Collected by the Millipore Filter Stages of the Sampler (*83*).

rapidly during the first hour when molecular iodine was depositing rapidly and then leveled off, or decreased as in runs F and G where the air was being circulated through a high-efficiency filter.

The experimental conditions that prevailed in the Zenith experiments were similar to the Dido-Pluto conditions except for the narrower concentration range (0.3 to 0.8 $\mu g/m^3$), and the same May Pack was used. It is not surprising, therefore, that comparable results were obtained. Data obtained with samples taken in the containment shell are shown in Fig. 4.5. The authors say that in this experiment (No. 2) the first charcoal-impregnated filter paper (ACG/B) and the brass holder together collected any molecular iodine present, with the brass collecting about 20% of the total reaching the sampler. They concluded that from about 4 hr onward the first charcoal paper was sampling about 50% molecular vapor and 50% some other form not sampled by the May Pack and being deposited in the containment shell more slowly than molecular iodine. They also say that the amount of iodine found on the second charcoal paper could be accounted for by a 95% efficiency for the vapor form "compound X."

4.2.2. Experiments in Smaller Enclosures

Croft, Iles, and Davis (*91*) describe experiments to determine the deposition characteristics of airborne iodine released initially as molecular vapor in quantities appropriate for power-reactor operations if all the iodine generated during long-term operation is assumed to be released. The release was made into a room about 30 m^3 in volume. They measured the deposition characteristics from an aging aerosol in both saturated and unsaturated air at ambient temperatures and with dry and wet surfaces. Figure 4.6 shows the

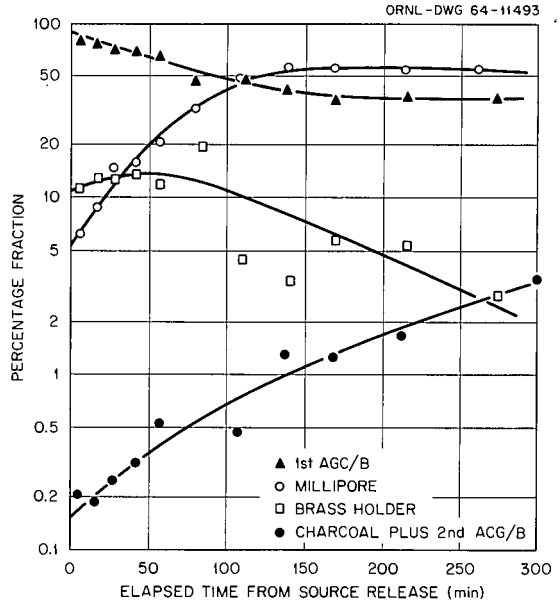

Fig. 4.5. Distribution of Airborne Iodine Expressed as Percentage of the Total (*90*).

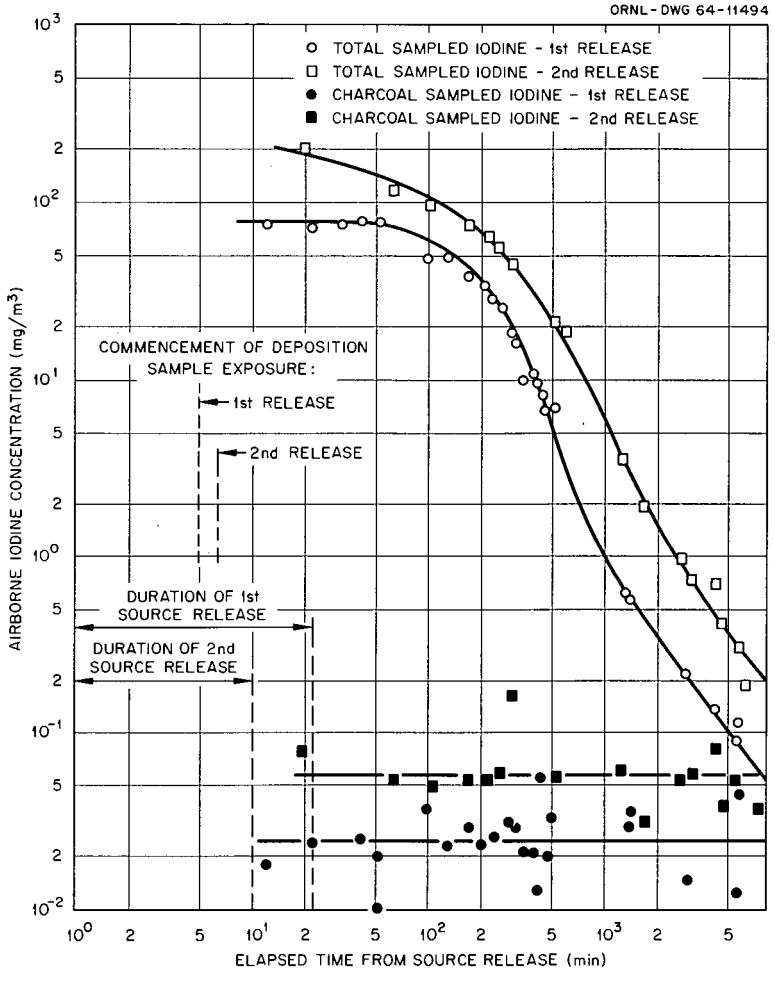

Fig. 4.6. Total and Charcoal-Sampled Airborne Iodine Concentrations in the Two Dry Experiments (*91*).

variation of airborne iodine with time during the dry experiments, while Fig. 4.7 gives results of the wet experiment. The general pattern is seen to be similar in both cases. The airborne iodine concentration, after initial dispersion, falls steadily by deposition to ~1% of the initial value after 1 day, leakage from the containment being negligibly small during this period.

A log/linear plot, as in Fig. 4.8, shows that in each of the three releases the time dependence of iodine concentration can adequately be described by a negative exponential function over an initial period of approximately 8 to 10 hr, whereafter the rate of attenuation decreases. Evaluation of the three exponential curves shown in Fig. 4.8 is given in Table 4.4.

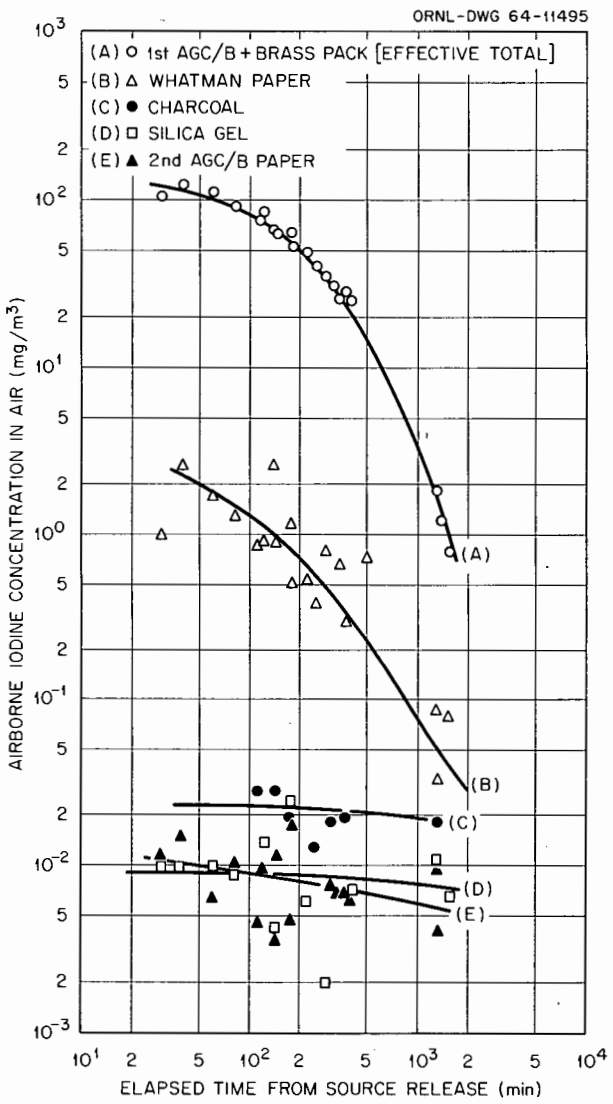

Fig. 4.7. Airborne Iodine Concentrations as Sampled by the Separate Components of the Sampling Pack in the Wet Experiment (*91*).

The measured deposition velocities are very close to those at related stages of the Zenith, Dido, and Pluto containments when the airborne iodine concentrations were several orders of magnitude ($\sim 10^5$) less. This suggests that the main factor limiting sorption is gas-phase diffusion to the surface.

Parker *et al.* (*46*) give data, shown in Table 4.5, on the variation of airborne iodine in a small (6.7-ft^3) stainless steel tank. The aerosol was produced by melting in air a stainless steel UO_2 fuel specimen that had been irradiated to 7000 Mwd/ton. The data show that the iodine concentration in the tank dropped rapidly during the first few minutes after melting, as might be expected for molecular iodine in a vessel having a large surface-to-volume ratio. The concentration then remained relatively constant for almost 2 hr; then a large fraction of the iodine that had deposited on the walls of the container was removed by passage of 1.5 tank volumes of argon through it.

Croft *et al.* (*91*) reported data (Table 4.6) showing that the effective deposition velocity for iodine on different surfaces varied with the time interval over which the measurements were made.

The 0–20-min values of effective deposition velocity in Table 4.6 are those most nearly applicable to molecular iodine vapor, in which form a major part of the source was deposited in the early stages. The decreased deposition velocity during the first 500 min indicates the influence of forms other than molecular iodine after the initial 20-min period. The data indicate that equilibrium was very nearly established during the first 500 min, with little change in rate occurring during the final 1000-min exposure period.

Data on the deposition of iodine, released as molecular iodine, in an air-filled stainless steel tank (1350 ft^3) are shown in Fig. 4.9 (*47*). The air in the tank was not stirred during this test, and it was unfiltered, with normal humidity. Two different deposition rates are distinguishable; the slower rate seemed to be constant over a rather extended period. The first curve extrapolates

to an initial value of 1.25 mg/m^3, about half the value calculated assuming 100% transfer of iodine from the furnace to the containment tank and uniform dispersion in the tank.

Results of a simpler experiment (*92*) in a smaller (6.7-ft^3) stainless steel tank are shown graphically in Fig. 4.10. Molecular iodine in this case was released into the tank filled with a steam-air mixture at 30 psig total pressure. The half-life of deposition varied from an initial value of about 6 min to 2100 min over the last 240 min of the exposure period.

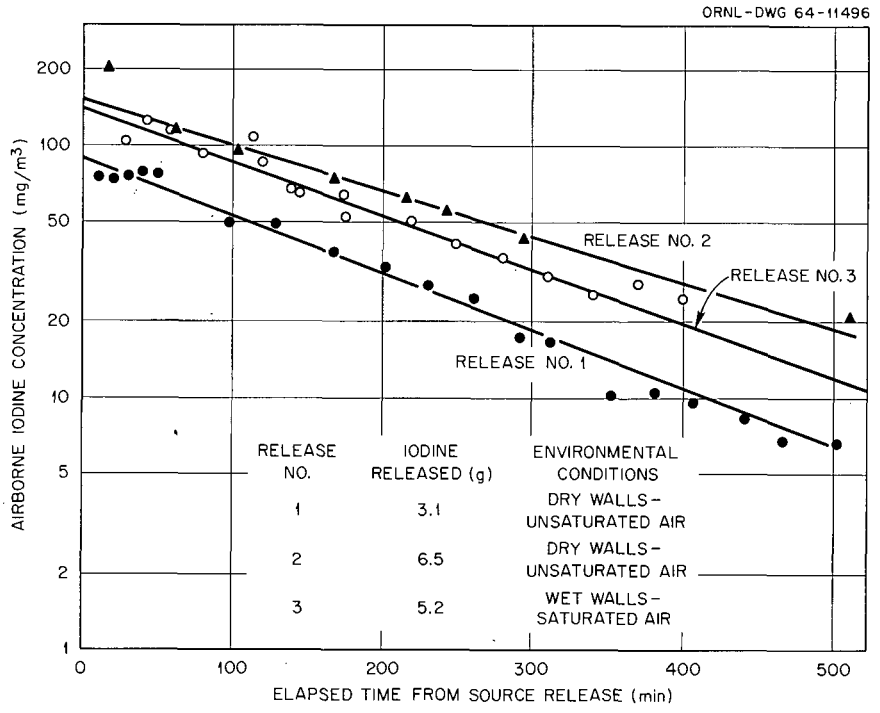

Fig. 4.8. Total Sampled Airborne Iodine Concentration for the First 8 hr Following Each Release (*91*).

Table 4.4. Evaluation of the Exponential Attenuation of Airborne Iodine Concentration (*91*)

Experiment Number	Environmental Conditions	Mass Released (g)	Mass Released / Volume of Containment (mg/m^3)	Exponential Function for Concentration, C (mg/m^3), Against Time, t (min)	Effective Deposition Velocity,[a] V (cm/sec)
1	Dry	3.1	113	$C = 90e^{-0.693t/130}$	4.0×10^{-3}
2	Dry	6.5	237	$C = 150e^{-0.693t/170}$	3.1×10^{-3}
3	Wet	5.2	190	$C = 140e^{-0.693t/140}$	3.7×10^{-3}

[a]The effective deposition velocity, V, is derived from the experimental exponential decay constant, λ, by the relation $\lambda = vA/V$, where A is the effective deposition surface area and V is the free volume of the containment.

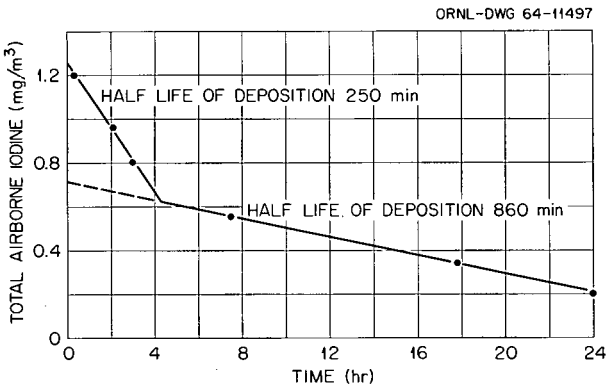

Fig. 4.9. Deposition of Iodine in the Stainless Steel Containment Vessel of the NSPP – Molecular Iodine Released in Unstirred Air.

Table 4.5. Variation of Airborne Iodine Released by Melting Irradiated (7000-Mwd/ton) Stainless Steel UO_2 (*46*)

Time After Melting (min)	Percent of Iodine Inventory Airborne
0	91[a]
10	11
25	21
55	5
115	6
118	11
300	72[b]

[a]Calculated from total amount released to the confinement vessel.

[b]Calculated from total amount collected in the filter-absorber system.

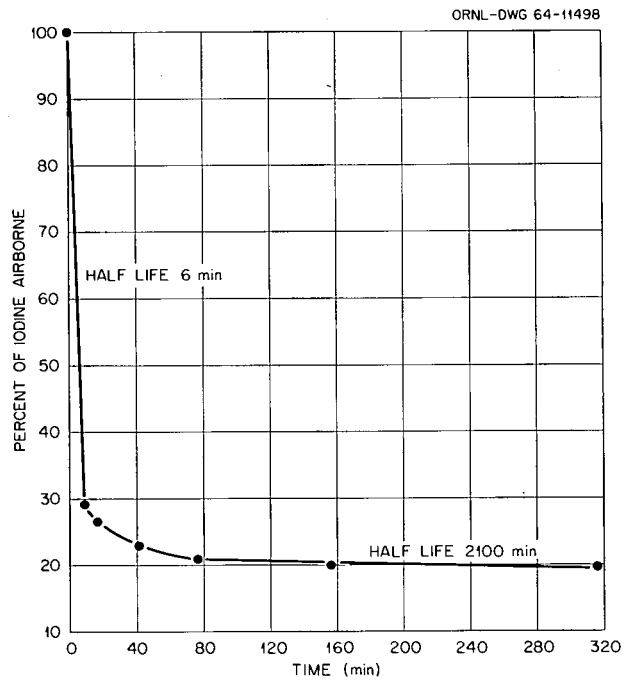

Fig. 4.10. Deposition of Iodine in Steam-Air Filled Stainless Steel Tank (6.7 ft^3).

Table 4.6. Values of the Effective Deposition Velocity for Varying Exposure Times (*91*)

Time from Source Release (min)	Exposure (x) (mg min^{-1} m^{-3})	Effective Deposition Velocity from $t = 0$ for Release No. 1 (y/x) (cm/sec)			
		Mild Steel	Concrete	Paint[a]	PTFE[b]
	$\times 10^3$	$\times 10^{-1}$	$\times 10^{-2}$	$\times 10^{-3}$	$\times 10^{-3}$
0–20	1.5	6.2	6.7	7.2	5.0
0–100	7.7	2.0	6.7	3.7	1.1
0–500	17	1.13	6.7	2.7	0.68
0–1500	19	1.06	6.1	2.6	0.64

[a]Chlorinated-rubber-based paint on a hard asbestos surface.

[b]Polytetrafluoroethylene.

4.3 PARAMETRIC EFFECTS ON DEPOSITION OF IODINE ON SURFACES

A great deal of attention has been devoted to study of factors affecting the rate of deposition of iodine on surfaces. The chemical form of iodine used for the study obviously will determine, in part, deposition rates. However, atmospheric and surface conditions also affect this rate, and these factors are discussed in this section. Interaction of variables undoubtedly occurs, and the contribution of such interactions to observed iodine behavior cannot be readily evaluated in some cases. It should also be recognized that while this section is devoted primarily to deposition studies, desorption occurs simultaneously with sorption except for surfaces that are a perfect sink for all forms of iodine present during the experiment. Although some overlap results from treatment of desorption in a separate section, this procedure can be justified on the basis that conditions existing during many desorption measurements were different from those under which deposition occurred.

4.3.1 Surface Composition

Croft *et al.* (*91*) conducted experiments on deposition of high concentrations of iodine (100–200 mg/m^3) on various surfaces. Deposition was found to be dependent on the nature of the surface, with bare mild steel and concrete surfaces showing higher deposition values than painted surfaces, which in turn were more receptive than the plastic (PTFE and polyethylene) surfaces (Table 4.7). Oxidized or polished mild steel surfaces were similar to each other in regard to deposition behavior. The metal surfaces were rapidly coated with a reddish corrosion layer. They were able to load surfaces to the high iodine levels indicated below (in mg/m^2) without apparent saturation: mild steel, 2000; concrete, 1400; paint, 75; PTFE, 17. However, there was some evidence of a reduced deposition rate at the higher loading levels.

No significant differences were noted in deposition on five sets of dry painted surfaces. This is in direct contrast to the measurements made under wet conditions, which showed faster deposition

Table 4.7. Effective Deposition Velocity on Surfaces (*91*)

Surface	Effective Deposition Velocity for 1500 min (cm/sec)	
	Dry	Wet
Room surfaces	4.0×10^{-3}	3.7×10^{-3}
Mild steel	10.6×10^{-2}	5.8×10^{-2}
Concrete	6.1×10^{-2}	4.0×10^{-2} (est.)
Mean of all painted surfaces	2.6×10^{-3}	
Mild steel with chlorinated-rubber-based paint		8.0×10^{-3}
Concrete with chlorinated-rubber-based paint		3.3×10^{-3}
Hard asbestos with chlorinated-rubber-based paint		5.6×10^{-3}
Mild steel with epoxy-resin-based paint		1.6×10^{-2}
Concretes with epoxy-resin-based paint		1.7×10^{-2}
Polytetrafluoroethylene (PTFE)	6.4×10^{-4}	1.9×10^{-3}
Polyethylene sheet		7×10^{-3}
480 cm^3 of water static in a vitreous enameled experimental dish, surface area 680 cm^2		2.5×10^{-3}

Table 4.8. Sorption of Iodine in Presence of Air at Room Temperature (*93*)

Material	Expt. No.	I_2 Conc. in Air ($\mu g/m^3$)	Rate Coefficient of Sorption in Linear Part of Curve (cm/sec)	Amount of Sorbed Iodine at Plateau of Curve ($\mu g/cm^2$)
Concrete	4	20	0.1	
	5	2500	0.08	70(?) or higher
Granite	4	20	0.020	
	5	2500	0.0032	2
Acrylate	4	20	0.012	
	5	2500	0.003	1.8
Polyvinyl chloride	4	20	0.005	
	5	2500	0.0004	0.2

rates onto epoxy-resin-based paint. This will be discussed further in the section on effects of moisture.

Forberg *et al.* (*93*) exposed small samples of granite, plastic materials, and concrete to elemental iodine vapor. Their results are reported in Table 4.8 (a rearrangement of the published table). The rate of sorption on concrete was independent of iodine loading up to 50 $\mu g/cm^2$. Forberg's results show relationships of the various materials which are similar to those obtained by Croft *et al.* (*91*), Table 4.9, third column, and by Hudswell *et al.* (*94*).

The results of a number of investigations are summarized in Table 4.10 (*81*). The results are arranged in the order of increasing dosage of iodine. The period of exposure of the absorbing surfaces to iodine was 60 min, except in the experiments referred to in columns 8 and 9 of Table 4.10, where it was approximately 200 min.

The amount of iodine sorbed on the copper samples varied by nearly a factor of 10^6 in these experiments, but the rate coefficient of uptake (velocity of deposition) varied by less than a factor of 10.

In the lower part of Table 4.10, the amount of iodine sorbed in the various surfaces is expressed as a percentage of that sorbed on copper.

The sorption on silver was somewhat higher than that on copper, except in the experiments of columns 6 and 7, where it was rather less.

Table 4.9. Velocity of Deposition of Iodine on Surfaces in Containment Vessels (*81*)

Surface	Velocity of Deposition (cm/sec)	
	Pluto[a]	Winfrith[b]
Copper	0.30	
Charcoal paper	0.28	
Mild steel	0.24	0.22
Painted steel	0.18	0.0039
Aluminum	0.14	
Concrete		0.055
Polyethylene	0.028	
PTFE		0.0012

[a]0.35 to 0.36 μg of I_2 per m^3 of air.
[b]7.8×10^4 to 1.6×10^5 μg of I_2 per m^3 of air.

The sorption on mild steel was nearly as high as that on silver and copper in the Pluto experiments, and this was also found in wind tunnel experiments in which the iodine carrier level was low.

At higher iodine concentrations and especially at higher temperatures and in dry air, the sorption on mild steel was only a few percent of that on silver and copper.

Table 4.10. Relative Deposition of Iodine on Various Surfaces (*81*)

	Pluto (7000 m^3) Col 1	Col 2	Laboratory Apparatus Col 3	Laboratory Apparatus Col 4	Col 5	Laboratory Apparatus Col 6	Laboratory Apparatus Col 7	Laboratory Apparatus Col 8	Laboratory Apparatus Col 9	Winfrith 27-m^3 Room Col 10
Period of exposure, min	60	60	60	60	60	60	60	~200	~200	60
I_2 vapor concentration, $\mu g/m^3$	0.35	40–100	100–1000	10^3–10^4	10^4–10^5	700	4×10^4	2×10^4	4×10^4	8–16 $\times 10^4$
Atmosphere	Moist air	Dry air	Dry air	Dry air	Dry air	Dry air	Dry air	Steam + air	Slightly moist air	Moist air
Temperature, °C	Ambient	Ambient	Ambient	Ambient	Ambient	200	200	150	150	Ambient
Absorbing surfaces	2-in. disks	⅛-in. rods	⅛-in. rods	⅛-in. rods	⅛-in. rods	⅛-in. rods	⅛-in. rods	2-in. disks	2-in. disks	2¼-in. disks
I_2 sorbed on copper, $\mu g/cm^2$	3.6×10^{-4}	$\sim 10^{-1}$	~1	~10	~30	4	240	83	150	80[a]
Velocity of deposition on copper, cm/sec	0.29	0.6	0.6	0.4	0.2	1.2	1.5	0.3	0.3	0.22[a]
Amounts sorbed relative to copper = 100										
Copper	100	100	100	100	100	100	100	100	100	
Silver		128	102	105	173	74	88	150	144	
Charcoal paper	110									
Mild steel	83	28	56	32	14	13	2	39	0.4	100[b]
Stainless steel								18	1.7	
Painted steel	52							4.5	4.0	1.8[b]
Concrete										25[b]
Aluminum	62	3.0	2.8	3.4	42	3.6	0.35	1.7	0.15	
Magnesium		1.9	4.4	3.4	6.9	1.7	0.30			
Graphite		3.6	8.2	4.2	3.1	1.4	0.36			
Glass		0.13	0.25	0.78	0.87	1.2	0.06	1.8	Nil	
Polythene	9									
PTFE										0.5[b]

[a]Amount sorbed on mild steel.

[b]Relative to mild steel = 100.

Table 4.11. Iodine Sorption on Steel Surfaces in Filtered Air at 15 psig and Ambient Temperature (*46*)

Exposure Time (min)	Iodine Sorbed ($\mu\mu g/cm^2$)		
	Stainless	Mild[a]	Painted
6	3	20	9
14	4	114	58
34	11	286	174
74	11	529	267
134	19	550	413
214	35	678	535

[a]Preoxidized by exposure to steam at a high temperature to produce a black oxide film on the surface typical of that found in high-temperature water systems.

The sorption on aluminum, magnesium, and graphite surfaces at ambient temperatures was also found to be generally a few percent of that on copper. The rate coefficient for sorption on these materials was of the order of 0.001 to 0.005 cm/sec.

A monolayer coverage of iodine is about 0.3 $\mu g/cm^2$. Copper, silver, charcoal paper, and concrete apparently sorb many monolayers of iodine with little reduction in the rate of deposition. Mild steel at ambient temperature and in moist atmospheres may belong in this group.

Stainless steel, aluminum, magnesium, and graphite adsorb about a monolayer of iodine. Glass, painted surfaces, and plastics adsorb from one to ten monolayers.

The rate of sorption of iodine from air at ambient temperature on different types of steel surfaces was determined as a function of time, and the results are recorded in Table 4.11 (*46*). The uptake of iodine increased over the time period studied. In contrast, similar specimens exposed in a steam-air atmosphere at initial temperatures ranging from 100 to 120°C showed comparatively little change in iodine sorption with exposure times up to 5 hr, and the amounts sorbed on all three types of surfaces were much less than in dry air.

4.3.2 Oxidation Conditions

A comparison of the location of the fission-product iodine released from uranium-aluminum alloys in three atmospheric environments, air, helium, and an 80% steam–20% air mixture, is shown in Table 4.12 (*84*). This type of study is of value as a measure of the transport efficiency and, in the case of the steam atmosphere, of the washout effect resulting from condensation of steam. A comparison of the apparent chemical effect of each atmosphere is also of interest. The data show that up to 88% of the iodine released in air was apparently in the molecular form and was carried through the filters to charcoal absorbers. In helium, on the other hand, the percentage of iodine reaching the charcoal was 0.1% or less with the bulk having deposited in the furnace tube and on the absolute filters, indicating that almost all the iodine was in the reduced form.

Davies *et al.* (*95*) found that stainless steel retained iodine at temperatures up to 300 to 400°C when helium or nitrogen was used as the carrier gas (see also Sect. 2 of this report).

Castleman (*96*) studied the deposition of iodine as a function of surface temperature (discussed in the next section) in different atmospheres. He found that most of the iodine deposited on hot (250–350°C) surfaces in helium but in air less than 3% deposited at temperatures above 100°C. Additional experiments by Castleman (*28*) concerning the chemical and physical behavior of the released fission products iodine, cerium, barium, lanthanum, molybdenum, and tellurium released from metallic uranium in purified helium and in air are described in Sect. 3.4.2 of Chap. 3, "Radioactivity Generation, Release, and Transport," of the *Reactor Containment Handbook*.

Raines *et al.* (*97*) reported results of experimental and theoretical studies of fission-product deposition from helium circulating in a closed stainless steel loop. They state that correlation of iodine deposition data was not attempted because of their complex nature.

Castleman and Salzano (*98*) reported that iodine, released from uranium metal in a helium atmosphere, deposits on gold, stainless steel, and quartz surfaces at 250 to 350°C in a combined form, probably as a uranium compound. The comparative performance of paints may depend on humidity, as well as on surface temperature as discussed in Sect. 4.3.5.

Parker *et al.* (*92*) have compared the sorption of iodine on painted steel at about 120°C with that on preoxidized mild steel and stainless steel

Table 4.12. Distribution of Fission-Product Iodine Released from Uranium-Aluminum Alloys in Various Atmospheres (*84*)

Transport Fraction	Fuel Temp. (°C)	Percent of Total Inventory		
		Air	Helium	Steam-Air
Total release	700	37.8		27.0
	800	78.6	29.8	76.8
	900	91.9	52.8	90.6
	1000	97.3	82.1	95.6
	1100	98.4	82.4	96.8
	1150	94.2		
In hot zone (iodide form)	700	19.9		17.0
	800	30.6	28.4	9.3
	900	11.3	51.3	12.9
	1000	38.2	69.6	13.0
	1100	6.3	65.8	10.3
	1150	17.4		
Transported to filters by particles	700	2.8		0.3
	800	4.5	1.4	0.1
	900	4.2	1.5	0.7
	1000	0.9	13.3	4.1
	1100	3.9	17.4	3.9
	1150	9.2		
Absorbed in charcoal traps (principally molecular iodine)	700	15.0		0.002
	800	44.2	0.08	0.05
	900	76.4	0.03	0.7
	1000	59.0	0.11	8.5
	1100	88.0	0.06	9.6
In the steam condensate	700			9.8
	800			66.9
	900			77.0
	1000			69.8
	1100			73.0

at 200 and 300°C in a steam-air atmosphere. No difference was found in the amount sorbed in the stainless steel surfaces at the two temperatures (3 $\mu\mu g/cm^2$) but the sorption on mild steel was slightly less at the higher temperature (22 $\mu\mu g/cm^2$) and both had less iodine than the painted surface (90 $\mu\mu g/cm^2$).

4.3.3 Temperature

Croft *et al.* (*91*) found that concrete retained a high proportion of iodine at temperatures to 300°C and only lost 12% during a 24-hr test at 200°C. Davies *et al.* (*95*) found that carrier-free iodine was retained on stainless steel at temperatures of 300–400°C when helium or nitrogen was used as a carrier gas (see also Sect. 2). The absence of oxygen appears to be important in this case. Copper and silver adsorbed more iodine at higher temperatures than at room temperature as shown by the data (*81*) in Table 4.13 (a rearrangement of the published table). This effect was also noted by Browning (*99*). At higher iodine concentration levels and especially at high temperatures in dry air, the sorption on mild steel was only a few percent of that on silver or copper.

Table 4.13. Comparison of Sorption of Iodine on Various Surfaces in Air at 20°C and 200°C (Period of Sorption, 1 hr) (*81*)

I_2 Conc. (mg/m^3)	Temp. (°C)	I_2 on Surface ($\mu g/cm^2$)						
		Glass	Mg	Graphite	Al	Mild Steel	Cu	Ag
0.50	20	0.015	0.031	0.075	0.037	0.50	0.90	0.81
0.73	200	0.043	0.21	0.063	0.107	0.36	2.0	1.6
37.2	20	0.40	1.96	1.32	1.15	1.52	17.2	40
46.5	200	0.45	1.83	1.00	1.32	5.7	108	90

4.3.4 Concentration

Croft *et al.* (*91*) state that in their experiments with high concentrations of iodine (100–200 mg/m^3) the measured deposition velocities were very close to those observed at related stages in the Zenith, Dido, and Pluto experiments, where the airborne iodine concentrations were several orders of magnitude (approximately 10^5) less. This suggests that the main rate-determining factor in sorption is the rate of gas-phase diffusion to the surface.

The rate of sorption on painted steel surfaces appears to be influenced by the amount of iodine available. The adsorption of copper, silver, charcoal paper, and concrete proceeds with little fall in rate when hundreds of monolayers of iodine have been adsorbed. This is also true for adsorption on mild steel in the presence of moisture. Some high-surface-area systems lose adsorption efficiency at higher iodine loadings in dry air. Glass, painted surfaces, and plastics are limited to a few monolayers of iodine.

4.3.5 Moisture

An accident with a water-cooled and/or -moderated reactor leading to a major release of volatile fission products would result in the simultaneous release of large quantities of steam into the secondary containment and in heavy condensation on containment surfaces. The results of some small-scale experiments by Morris and Nicholls (*100*) on the sorption of iodine on various materials at 150°C in dry air and in an air-steam mixture are summarized in terms of the rate coefficient (deposition velocity) in Table 4.14. There was a much higher deposition velocity of iodine on mild steel in the presence of steam. Thus, unpainted mild steel is an effective getter for molecular iodine in the 150°C temperature range with steam atmospheres (no water condensation).

Table 4.14. Uptake of Iodine by Surfaces During 4 hr at 150°C (*100*)

Material	Deposition Velocity (cm/sec)	
	40% Steam	Dry
Copper	0.31	0.30
Mild steel	0.12	0.0013
Silver	0.47	0.43
Aluminum	0.0060	0.00046
Stainless steel	0.054	0.0050
Painted mild steel	0.014	0.012
Glass	0.0055	0.00001

Parker *et al.* (*84*) reported that steam washout in their experiments accounted for up to 75% of the iodine; even with 20 vol % air present in the saturated steam, only 10% of the iodine remained volatile enough to reach the charcoal filter system. These workers also compared the distribution of fission-product iodine released from irradiated aluminum-uranium alloys in steam and dry atmospheres (*85*), and the data are given in Table 4.15.

Table 4.15. Distribution of Iodine Released from Molten Aluminum-Uranium Alloy in Moist and Dry (Ordinary Humidity) Air (*85*)

Location of Activity	Percent of Released Iodine in Each Location	
	Dry Air	Moist Air
Furnace tube	1.8	3.9
Aerosol tank	82.0	48.1
Steam condensate (30 ml)		6.7
Transite pipe (12 in. long)	1.2	3.1
Filters	6.9	8.9
Charcoal beds	8.0	29.2

It was indicated in Sect. 4.3.1 that there were no systematic differences among the various painted surfaces with dry conditions. Under wet conditions a measurably faster deposition rate onto epoxy-resin-based paints has been observed. This behavior is consistent with measurements by Forberg *et al.* (*93*) on epoxy and chlorinated rubber.

4.3.6 Carrier Gas Velocity

The rates at which carrier gas flows past fuel samples during release of iodine is generally considered to have no effect on the rate of release. Browning *et al.* (*40*) reported data showing that varying the rate at which helium flowed past fuel samples in in-pile experiments from 60 to 350 fpm did not affect the release of iodine from UO_2 specimens or from the high-temperature zone. However, flow conditions, whether laminar or turbulent, do affect the deposition patterns of iodine in off-gas piping or duct systems. Laminar conditions are applicable to the determination of the forms of iodine with diffusion tubes, as previously described in Sect. 3. Higher deposition rates were reported by Raines *et al.* to occur at bends and rough surfaces in loop equipment as a result of increased gas turbulence (*101*). Behavior of iodine on particulates will be influenced by the flow patterns in a reactor system.

Results of experiments (*102*), showing distribution of iodine released into a stainless steel vessel in the form of molecular iodine, are summarized in Table 4.16. Five release experiments (runs) were conducted: filtered air; filtered air and steam with two containment times; and filtered air and steam with two types of organic gases added. In all five runs, the tank contained filtered air at 15 psig. In all runs except 6-11, the tank also contained 10 to 15 psi of steam. In four runs, pressure in the tank was released after 5 hr; in run 8-4 the pressure was held for 18 hr. The results are tabulated below. In clean air a fairly high percentage (80%) of the iodine rapidly plated out on the tank walls. Only 3.0% remained available then for leakage from the pressurized tank. In this case, however, considerable desorption occurred later, bringing the total airborne quantity to 13%. This amount decreased in the presence of steam to 3–6%, while about 50% of the iodine was collected in the condensate formed as the tank cooled over a period of several hours. On the other hand, when organic materials were added, as in run 7-16 where dry ice (CO_2) and acetone were obviously present in the sample and in run 9-10 where a 500-cm^3 sample of freshly prepared mixed hydrocarbons (from the hydrolysis of UC-UC_2) was added to the tank, a decided increase (to 27%) in the amount remaining airborne occurred. The data in Table 4.16 show that factors other than the presence of moisture have a marked effect on the amount of iodine available for leakage from the simulated containment shell after aging of the aerosol.

An attempt to classify the forms of iodine in each test by means of an elaborate filter pack showed no significant trends, except that in each of the two organic runs a large fraction (16%) of the iodine penetrated both filters and the metallic silver or copper gauze, suggesting nonreactive iodine.

Unfortunately, other supporting data are not sufficiently clear to permit unequivocal identification of the nonreactive iodine. The amount of iodine in a form sufficiently nonreactive to penetrate 1.5 in. of charcoal remained low in all cases, although it was noticeably higher in one experiment in the presence of organic gas.

High-velocity deposition distributions are being interpreted in terms of diffusion, interception, and inertial impaction of aerosols in fibrous filters by Browning and Silverman (*103*). See Sect. 5, "Trapping of Fission-Product Iodine and Induced Removal Processes."

Table 4.16. Iodine Deposition and Desorption in the Containment Mockup Facility (Expressed as Percentage of Total Iodine Released into the Tank)

	Air	Steam-Air		Steam-Air and Organics	
	Run 6-11	Run 6-25[a]	Run 8-4	Run 7-16	Run 9-10
Iodine held in containment tank					
Retained on tank walls	79.6	60.8	38.2	19.3	20.9
Collected in steam condensate		34.9	54.0	52.9	47.3
Total retention	79.6	95.7	92.2	72.2	68.2
Iodine removed from tank					
By pressure release	3.0	1.1	2.4	12.9	8.5
By argon displacement	7.4	1.2	2.4	13.3	15.9
By air sweep	2.9	0.3	1.7	0.5	2.6
Total removed, airborne	13.3	2.6	6.5	26.7	27.0
Iodine removed on test samples	6.1	1.7	1.4	0.8	2.2
Distribution of airborne iodine from tank					
Retained on filters	4.1[b]	0.5	1.5	0.1	0.3
Retained on silver/copper screens	5.3	0.8	2.3	6.4	8.5
Retained on charcoal papers	3.0	0.4	0.4	2.0	15.8
Retained in charcoal cartridges	0.3	0.6	2.0	16.4	2.3
Loss through 1.5 in. of charcoal	0.003	0.0002	0.05	0.3	0.03

[a]In 6-25, iodine was released in steam and air under conditions providing a large amount of condensate (uninsulated tank).

[b]One organic membrane filter used in this test probably reacted with and retained some molecular iodine in addition to particulate iodine.

4.3.7 Surface-to-Volume Ratio

When comparing the results of experiments in large reactor vessels with data obtained in smaller-scale laboratory experiments, the surface-to-volume ratios should be considered in the interpretation of the physical and chemical behavior of iodine, since the half-life of iodine in the gas phase will depend to some extent on this ratio. Literature data are sometimes difficult to correlate in this respect because unstated or unmeasured amounts of surfaces other than that of the shell itself were present during the experiments. Ratios calculated for several present or proposed experimental facilities (given in Table 3.1) can be compared with the value of 0.033 ft^{-1} calculated for a typical large reactor shell (Dresden).

A velocity of 0.1 to 0.3 cm/sec for molecular iodine depositing on the metallic and concrete surfaces of a containment vessel has been indicated by Chamberlain (*81*). In the Dido and Pluto reactor containment shells, the surface-to-volume ratios were approximately 0.004 cm^{-1}. The initial velocity of deposition was 0.15 cm, and the initial half-life deposition on the surface of the reactor containment was $0.693/(0.004 \times 0.15) = 1000$ sec. The effective deposition velocity (V) in containment vessels can be derived from the experimental exponential decay constant by the relation $\lambda = VA/V$, where A is the effective surface area and V is the free volume of the containment.

4.4 DESORPTION

It was pointed out earlier in this section that desorption in many cases is occurring to some extent while iodine is depositing. It is important

to know what fraction of the iodine that has deposited on a surface may become airborne again and the conditions affecting this fraction. The factors affecting desorption will be discussed here.

4.4.1 Oxidation of Iodides

Data presented in Sect. 2 show that iodides have much lower vapor pressure than molecular iodine. Consequently, when iodine deposited on surfaces in the reduced state is exposed to atmospheric oxygen, partial desorption frequently occurs. Castleman (*96*) reported that when surfaces on which iodine from irradiated uranium or U-Mo alloys deposited from a helium stream at 250 to 350°C were exposed to air at the same temperature, the deposited iodine was slowly converted to the elemental form. He stated that the iodine probably deposited as UI_2 since CsI is stable in air at these temperatures. In reactor accidents involving pressurized water reactors, reducing conditions may exist in the early phases of the accident due to metal-water reactions. Later, as steam condensation in the shell results in lower pressure, air may enter the shell. The ease with which the reduced iodine will oxidize depends on a number of factors, but one of the more important is the element with which it is combined. There is very little published information on this subject.

4.4.2 Surface Effects and Exchange Reactions

Croft *et al.* (*91*) determined rates of iodine desorption from materials exposed to high concentrations of molecular iodine (see Sect. 4.4.3 for experimental details) and reported the following values for the half-life of desorption in air at ambient temperatures: mild steel, 17 days; concrete, 10 days; painted steel, 4 days; polytetrafluoroethylene (PTFE), 2 days. A more detailed analysis of the desorption measurements showed that the desorption rate decreased with time and that over the 4-day period during which the above values were determined, a fourfold reduction in rate occurred. A correlation between deposition velocity and desorption rate was noted.

Megaw and May (*83*) exposed circular disks with different surfaces in the Pluto experiments to a low concentration of molecular iodine (0.35 $\mu g/m^3$) for 1 hr. After exposure the disks were placed in sealed plastic bags, removed from the reactor shell, and counted as soon as possible. Three disks of each surface type were exposed, and desorption rates were determined in three atmospheric environments: clean air, air containing about 1 μg of stable iodine per m^3, and air containing about 3 g of stable iodine per m^3. Results are shown graphically in Fig. 4.11. It is clear that little desorption of ^{132}I from copper or charcoal surfaces occurred either in the presence or in the absence of airborne ^{127}I. Varying degrees of desorption from the other surfaces were observed, and exchange of surface-bound ^{132}I with airborne ^{127}I increased both the rate and the extent of desorption. These results confirm the desorption results obtained by liberating ^{127}I in the reactor shell, as shown in Figs. 4.1 and 4.2.

Parker and his co-workers (*46*) have found that part of the iodine deposited on the dry stainless steel surface of a simulated containment shell is rather readily removed by passing a stream of argon or air through the vessel. This is shown by data in Table 4.5, discussed in Sect. 4.2.2. In another experiment, molecular iodine (approximate initial concentration, 2 mg/m^3) was allowed to deposit for about 4 hr in the steel vessel filled with air at 15 psig (relative humidity, approximately 60%). Reduction of the air pressure at the end of this period from 15 to 2.5 psi removed about 2.8% of the iodine inventory, while sweeping with $2\frac{3}{4}$ tank volumes of argon removed 7.0% and $2\frac{1}{2}$ tank volumes of air removed an additional 2.9%. Corresponding figures obtained after aging the iodine aerosol for 18 hr in a steam-air mixture were 2.5, 2.4, and 1.7% respectively. Higher removal values were obtained with shorter exposures and where organic impurities were present in the vessel atmosphere. Fairman (*104*) reported losses of 32 to 39% of the ^{131}I during three weeks' exposure to air, from plates prepared from water solutions, using plates on which the iodine was deposited as AgI as a reference for 100% retention. It is obvious that care must be exercised in the handling of deposition coupons and other experimental devices having adsorbed iodine in order to avoid losses.

Collins *et al.* (*48*) reported that comparatively large (several percent of the iodine reaching the charcoal trap) amounts of penetrating iodine,

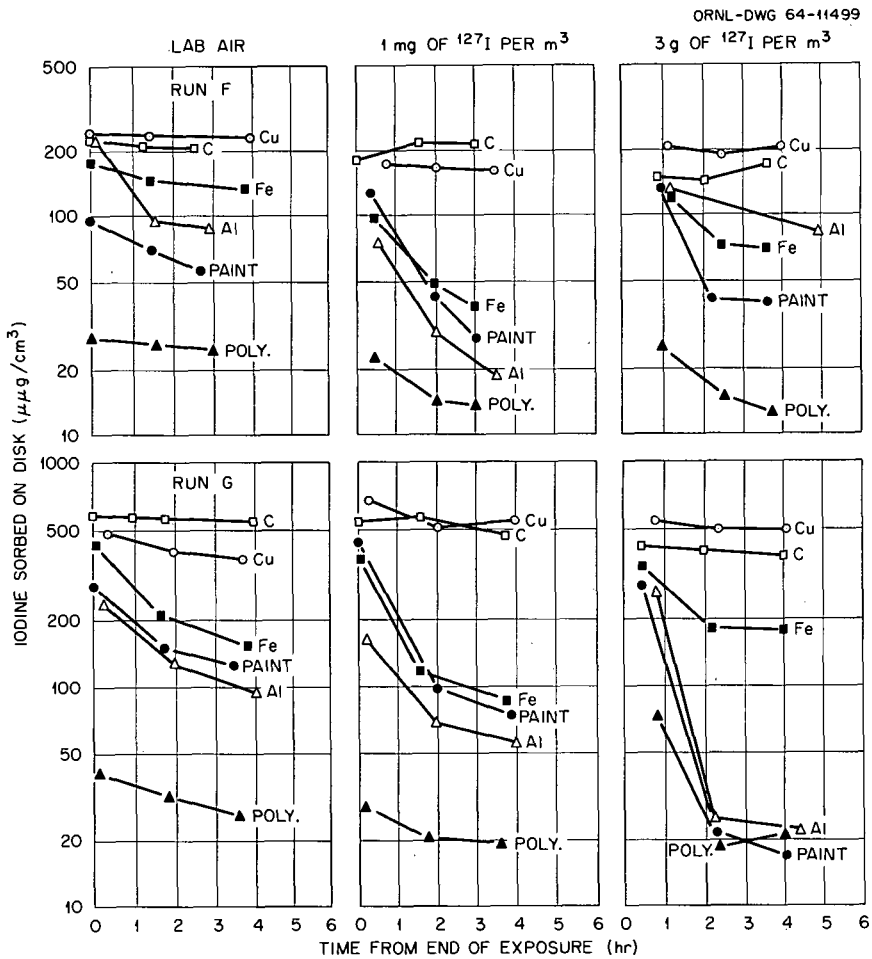

Fig. 4.11. Desorption of Iodine from Disks Exposed in Pluto (*81*).

identified as a mixture of alkyl iodides, were formed in an experiment involving release of iodine from irradiated UO_2 in CO_2 at temperatures in the range 1500 to 1700°C. A large fraction of the iodine released from the fuel deposited on vessel walls and other surfaces at temperatures up to 400°C. The authors state that it has been shown in other experiments (undocumented) that iodine desorbed from surfaces contains an enhanced proportion of alkyl iodides. This observation could have an important effect on the design of trapping systems for post-accident trapping of released iodine.

4.4.3 Desorption in Air at Elevated Temperatures

Two desorption experiments were carried out by Croft and co-workers (*91*). The first sets of deposition samples were heated in a small oven at temperatures of 50, 100, 150, and 200°C, for periods of 2, 4, and 24 hr, and the resultant desorption was measured after cooling. In the second experiment, some steel and concrete samples only were examined at temperatures approaching 300°C. Paint samples were not heated beyond 4 hr at 200°C, as under this condition the surfaces began to exhibit severe discoloration and flaking.

The results of the first desorption experiments are said (*91*) to exhibit certain main features, as follows:

1. The mean value ϕ decreased with the length of heating at a given temperature.

2. The mean value ϕ increased with temperature for a given length of heating;

$$\phi = \frac{1}{S_1} \frac{\Delta S}{\Delta t},$$

where

ΔS = loss in surface iodine concentration during Δt,

$S = S_1$ at $t = 0$.

3. Up to temperatures of ~150°C the relative desorption behavior of the specimens did not change significantly from that under ambient conditions, though the absolute rates increased.

4. PTFE shows the highest desorption factor for the three temperatures to which it was subjected. In contrast, the concrete samples exhibited the least facility for desorption during almost all the tests conducted. This was such that even for 24-hr periods at 200°C the resultant loss from the concrete was still only ~12%. For the painted samples a significant increase in desorption at temperatures approaching 200°C was associated with the observed surface deterioration.

Other steel and concrete samples were subjected to a different heat cycle, involving heating for ~20 min from ambient to a preset temperature, followed by natural cooling in air for ~10 min and subsequent recounting.

The main results of the second desorption experiment were as follows:

1. Up to a maximum cycle temperature approaching 300°C, the concrete samples nowhere showed a rapid increase in resultant desorption. For the metal specimens, however, there was a slow increase in percentage loss up to maximum cycle temperatures of ~225°C, by which time some 5% was lost per cycle, and between 225 and 250°C the onset of a rapid increase occurred, such that at ~270°C some 70% was lost per cycle.

2. Recycling of these metal specimens generally failed to reproduce this rapid increase, the rate remaining virtually independent of temperature after one cycle. One or two samples which had particularly high initial deposition levels showed a second increase at the same temperature, but of reduced proportions. It was also found that other mild steel samples, which had been heated for periods at lower temperatures, failed to exhibit the enhanced desorption at the higher temperature. The onset of the pronounced increase in desorption rate from the mild steel surfaces between 225 and 250°C probably results from a chemical process initiated at this temperature. It is known, for example, that the solid halides of iron sublime at relatively low temperatures, and some such reaction may be occurring here.

4.5 SUMMARY

A great deal of information on the transport behavior of iodine has been accumulated in recent years, but the data obtained serve better to indicate the need of further investigations than to supply definite answers to questions concerning the attenuation factor for iodine deposition in containment shells. One area that needs more attention is the role of submicron particles in iodine transport. It is clear that many factors affect iodine deposition behavior, but few, if any, of the parameters have been adequately evaluated under realistic accident conditions.

Large-scale tests in real or simulated containment systems have been made, in general, with unrealistic sources (molecular iodine liberated in the absence of vaporized fuel or cladding particles). Small-scale experiments with more realistic sources and with atmospheres currently of greatest interest in the U.S. nuclear safety program (steam and steam-air mixtures) have been made, to date, only with stainless steel simulated containment shells. Much useful data are being produced in experiments of this type, but more meaningful deposition rate data can be obtained by use of surfaces more nearly representative of the walls of real reactor shells, such as painted mild steel. Facilities that will permit parametric studies of deposition rates of iodine and other fission products on such surfaces are in operation at Hanford, and similar equipment is presently being designed at ORNL. Information coming from new and from existing installations should markedly reduce the areas of ignorance in iodine transport behavior in containment systems within the next two years. Data obtained under similar conditions with simulated reactor shells of increasing size will aid evaluation of the surface-to-volume ratio parameter and permit extrapolation of results to full-scale reactors. Results of full-scale reactor meltdown experiments in the LOFT facility will, eventually, lend more confidence to the results of the smaller-scale tests.

5. TRAPPING OF FISSION-PRODUCT IODINE AND INDUCED REMOVAL PROCESSES

The consequences of a reactor accident can be significantly reduced by removal of released radioactive fission products from gases, before they escape to the environment. Accordingly, development and testing have been carried out on gas cleaning methods for this application, making use of particulate filters, adsorbers, scrubbers, foam encapsulation, steam condensation, scavenging, diffusion boards, and pressure suppression designs. This general subject has been reviewed in *Nuclear Safety* (*99*), and a series of conferences has been held on the subject of gas cleaning under sponsorship of the USAEC (*105–109*).

The design of a system for removing radioactive fission products from a gas depends upon the physical and chemical forms of the fission products and upon the nature of the gas. They may appear condensed as an aerosol or as gaseous impurities having a variety of chemical forms. Iodine is of special interest because of its importance as a potential contaminant in accidents and because of the previously discussed chemical variability (*25, 40, 67, 110–123*). To maintain complete control over fission products released in an accident, one must provide gas cleaning devices which are effective against each of these particulate and gaseous materials and must know the efficiencies of these devices.

The removal of radioiodine from gases has been the subject of a series of extensive reviews (*124–128*) which covered removal by a variety of materials. Materials used included activated charcoal, beds of silver-plated wire or heated silver nitrate, silver or copper mesh, caustic scrubbers, and dry soda lime. Forms of radioiodine studied included molecular iodine, iodine compounds, and iodine on particulates. Selection, design, testing, and efficiency of removal systems have also been reviewed.

Recent literature (*18, 81, 169*) contains information on the iodine trapping efficiency of these materials, comparing the effect of variations in iodine concentration, gas velocity, temperature, and the concentration of airborne impurities on the efficiencies for iodine removal from air, steam-air mixtures, helium, and carbon dioxide.

5.1 TRAPPING FISSION-PRODUCT IODINE

5.1.1 Particulate Filters

Elemental iodine is only partially removed by filters except when it is adsorbed on large particles. For example, a high-efficiency filter retained >99.99% of the cesium, tellurium, ruthenium, and uranium, as compared with 30% of the iodine released by melting stainless-steel-clad UO_2 in air (*25*). In an in-pile melting experiment in which a stainless-steel-clad UO_2 fuel element was melted in a helium atmosphere, a combination of roughing filter and HV-70 filter removed iodine with 92% efficiency (*40*). Particulate filter efficiencies in general ranged from 50 to 99% for iodine released from metallic uranium fuel elements heated to 1215°C in atmospheres of air, steam, or helium (*30*). In other tests where stainless-steel-clad dispersed-UO_2 fuel elements were melted in air, the released iodine was removed by a CWS-6 filter with 71% efficiency (*112*). In tests in which full-scale metallic fuel elements were melted in air and samples were collected at distances up to 5 miles downwind, iodine was the only fission product which penetrated absolute filters to an extent exceeding 5% (*129*). When aluminum-uranium alloy fuel elements were melted in dry air and in moist air atmospheres, the iodine was removed by the filter with 46% efficiency in dry air and 23% in moist air.

It is beneficial to have particulate filters in the systems for removal of the larger particles containing adsorbed iodine. The remaining iodine is then collected on solid adsorbents described in Sect. 5.1.3. An adaptation of charcoal paper filters has been used in sampling procedures (see Sect. 3.3.1). The activated charcoal filter material has been used primarily for collection of radioiodine near nuclear reactor installations. The paper is a combination of activated charcoal with a cellulose binder. Approximately 60% of the weight of the paper is composed of activated charcoal.

5.1.2 Liquid Scrubbers

J. M. Holmes (*128*) reviewed recent developments in iodine and ruthenium removal, the adsorption of nonradioactive contaminants from off-gas streams, and the design of several new cleanup systems. He emphasized that radioisotopes of iodine which contaminate the gaseous wastes from chemical processing plants are difficult to remove because of the various states in which they may appear in off-gas systems. For example, iodine can be present as iodine vapor; as a particulate solid, such as NaI; or as a solute dissolved in entrained liquid droplets or adsorbed on the surface of aerosol particles. A number of scrubber-type systems are reviewed as follows.

Taylor (*130*) studied the rates of iodine vapor absorption in water and in aqueous solutions of sodium hydroxide, sodium thiosulfate, sodium tetrathionate, and sodium sulfate. With a disk-type laboratory absorption column, the rates were found to be completely gas-phase controlled for the sodium hydroxide and sodium thiosulfate solutions; appreciable liquid-phase resistance to transfer was found with the water, sodium tetrathionate, and sodium sulfate absorbents. The gas-phase absorption rates were quite rapid, and a correlation with ammonia-water absorption data was obtained.

Several recent investigations emphasized the role of iodine adsorption on particles in reducing the efficiency of caustic scrubbers. May and Morris (*131*) conducted a series of iodine-absorption efficiency tests on the British BEPO reactor caustic scrubbers. The system comprised four 3-ft-diam towers, in parallel, containing 6-ft beds of 1-in. Raschig rings. A 5% sodium hydroxide solution was used to scrub 3000 cfm of air containing injected ^{131}I. The predicted decontamination factor was 100 assuming perfect liquid distribution; this factor was 10 to 20 for the expected liquid distribution. The decontamination factors actually obtained for normal operation varied between 29 and 32. Substitution of sodium carbonate solution for the caustic did not change the efficiency. However, when particles were introduced into the inlet air stream, the decontamination factor dropped markedly. In one test, in which the air and iodine vapors were passed over a tungsten wire heated to 900 to 1000°C (a metal fume generator) before injection into the air feed, the decontamination factor dropped to 10.5. In another test the introduction of lead oxide particles reduced the decontamination factor to 1.4. The decrease in efficiency was attributed to the adsorption of iodine on particles that were not removed by the scrubber.

Chamberlain and Wiffen (*132*) demonstrated that iodine in air at a concentration of 10 $\mu g/m^3$ was rapidly and irreversibly adsorbed on lead fume but that the degree of adsorption on 0.1-μ condensation nuclei was small for the same iodine concentration. Experiments performed with the reactor containment shells and air cleaning systems of the British DIDO and PLUTO reactors (*133*) demonstrated the relative iodine removal efficiencies of a sodium carbonate scrubber, a copper knitmesh bed, and a charcoal bed in series with a Vokes filter. Only the activated charcoal bed–Vokes filter combination gave a reasonably high decontamination factor after the initial 1 $\mu g/m^3$ iodine concentration had dropped below 0.03 $\mu g/m^3$ as a result of adsorption on the shell and reactor walls. Since the Vokes filter (efficient down to 0.1-μ particles) did not improve the efficiency of the scrubber or copper bed, the adsorption of iodine on nuclei from the air was not considered to be the cause for their poor performance. The high efficiency of the charcoal bed led to the hypothesis that a gaseous compound of iodine was formed, perhaps from reaction of iodine with trace impurities in the air, and was strongly adsorbed on the charcoal but not removed by the scrubber or the copper bed.

5.1.3 Solid Sorbents

Radioiodine under most circumstances can be removed from gases most efficiently by activated charcoal adsorbers. The large surface area of activated charcoal has a rapid initial retention of elemental iodine. At room temperature the desorption is slight, possibly in the range of 0.001%. Adams and Browning (*114*) passed air through a 0.75-in.-deep bed of ^{131}I-contaminated charcoal at a velocity of 75 fpm for 250 hr. The activity level was monitored periodically, and the decay rate was found to be identical to that of ^{131}I (8.05 days), indicating no loss of iodine at room temperature. At higher temperatures the iodine may become more mobile, particularly in the presence of oxygen, and there is a considerable difference in the performance of various types of charcoal. Chamberlain (*81*) quoted desorption data

at 250°C in air ranging from 0.01% in 4 hr to 22.5% in 2 hr. The coconut-base charcoals have better retention but a lower ignition temperature. Adams and Browning (*134*) found an ignition temperature of 290°C for one type of activated charcoal in flowing oxygen. Chamberlain (*81*) reported ignition temperatures for various charcoals in flowing air of 300–500°C.

Charcoal-filter burning tests in air (*135*) indicate that a water spray activated by a temperature-sensitive device that has been considered for use in the trapping system of the HFIR at ORNL is effective in extinguishing charcoal fires. Carbon dioxide has also been used for this purpose in tests conducted by the Underwriters' Laboratories (*136*). When air flowed through a 20-in. by 20-in. by ½-in.-thick bed at 200 lin ft/min, two 15-lb CO_2 extinguishers were required to cool and extinguish the fire. Approximately 25% of the charcoal was consumed. In general, filter systems are designed with enough natural cooling to prevent temperatures from reaching the combustion point of charcoal.

The presence of alkali metals in charcoal improves its retention, especially at high temperatures. The amount of iodine which a charcoal can retain at high temperature is limited by the number of metallic adsorption sites and also by the tendency of oxygen to replace iodine in metallic iodides at temperatures above 200°C. Possible substitutes for charcoal that would be less susceptible to oxidation effects are discussed in Sect. 5.1.5.

Nonelemental forms of iodine such as the organic iodides are adsorbed by charcoal, although with poor efficiency (see Sect. 5.1.4), but are not removed by silver, copper, or alkaline materials. This is believed to be one reason why copper, silver, and caustic scrubbers have shown poor performance in actual installations. The development of trapping methods for organic iodides is discussed in Sect. 5.1.4. Practical adsorbers have been designed having efficiencies of greater than 99.9% and 99.99% for molecular iodine (*111, 114, 137–141*). High efficiencies persist even after extended operation (*140–142*) and in the presence of steam (*142–144*). Radioiodine has been removed from helium at temperatures up to 700°C by activated charcoal loaded with metal salts (*145–146*). A combination particulate filter and charcoal adsorber collected 99.97% of the iodine released by melting aluminum-uranium alloy fuels in dry or moist air (see Sect. 5.3.4) and 98.6% of that released by melting stainless-steel-clad UO_2 fuel in air (*25*) (Sect. 5.3.5). One iodine vapor species was observed to penetrate activated charcoal under humid conditions in air, although it was effectively removed under dry conditions (*123*). Reduced efficiencies for adsorption of radioiodine by charcoal have been observed, especially at low concentrations, under special circumstances where the iodine had been exposed to impurities from chemical processing plants, metallurgical hot cells, or burning organic materials (*139, 141, 147–149*). Methods for testing iodine adsorbers in place after installation have been developed to ensure their continued effectiveness and are applied routinely to the adsorbers in the reactor compartment ventilation system of the NS "Savannah" (*144, 150–151*) (see Sects. 5.3.1 and 5.3.2). Similar tests were applied to the confinement system of the Oak Ridge Research Reactor (*152*).

Silver and silver compounds (e.g., $AgNO_3$ or AgI used as coatings) are very effective iodine removal agents. Removal efficiencies are as high as 99.9% in dynamic systems (gas flowing past materials such as Fiberfrax rings and metal screens coated with silver or a silver compound). Silver-coated materials have an optimum operating temperature of 190 to 220°C, but they work very well at 260°C. Water vapor has no effect on the removal efficiencies at 220°C for $AgNO_3$-coated materials, but the temperatures must be kept high enough to prevent condensation of water. Silver surfaces are reported (*114, 138–139, 153–154*) to be susceptible to interference by impurities or surface contaminations.

The chemistry of the reaction of iodine with silver compounds indicates that the equation

$$AgNO_3 + \tfrac{1}{2}I_2 \longrightarrow AgI + NO_2 + \tfrac{1}{2}O_2$$

is applicable, but at 190°C both silver iodide and iodate are formed. Silver iodide was stable in air at 350°C, but 78% decomposed at 550°C and 85% decomposed at 700°C in ½ hr. Silver materials are potentially useful when placed ahead of a charcoal adsorber to reduce the generation of decay heat of radioiodine in the charcoal.

A clean reduced copper surface acts as a perfect "sink" for iodine at room temperature. However, it is difficult to maintain an unoxidized surface, and, for this reason, some investigators prefer to

use silver-coated screens in May Packs rather than copper. Copper and copper alloys also effectively remove iodine from solution. Oxidized copper or copper oxide is relatively inefficient for iodine removal; efficiencies of 37.5, 4.3, and 0.5% were obtained at 125, 242, and 295°C respectively (*155*). Copper reacts with iodine below 242°C, but iodine is liberated from copper iodide above this temperature.

Morris *et al.* (*156*) made a thorough study of the adsorption and desorption of iodine in beds of copper; they also give some results of comparative tests with beds containing silver surfaces. Their data show that both materials are effective in removing molecular iodine from air streams.

Glass beads and sea sand are very inefficient for iodine removal. Lead will remove most of the iodine from a solution. Glass, stainless steel, and Lucite will remove a small amount from solution; aluminum appears to remove a slightly greater amount.

Adsorption of iodine on mixtures of silica gel and alumina at 106°C follows a BET theory (*157*) for adsorption on the dual surfaces, with alumina giving a type II and silica gel a type III adsorption. Molecular sieve material showed 99.9% or better (*170–171*) efficiency for iodine retention, with 55% retained in the first $^3/_8$ in. Iodine may be eluted from this material by a continued flow of air, unlike iodine adsorbed on charcoal.

5.1.4 Trapping Methyl Iodide

Collins and Eggleton (*158*) investigated the adsorption of labeled methyl iodide on coconut charcoal at different temperatures over a range of loadings. At 100°C quantities smaller than 100 μg were permanently adsorbed on a 25-g bed, but increasing penetration was observed as the loading was increased above this limit. At room temperature there was a longer delay before breakthrough occurred, due to enhanced physical adsorption, but the amount of methyl iodide permanently retained was considerably less.

The main duties of the trapping system of an advanced gas-cooled reactor are either to depressurize the pressure circuit after a single channel meltout or to provide continuous treatment of relatively clean coolant after a pressure circuit failure. The trapping system is not therefore required to operate for long periods while retaining a large amount of methyl iodide, and the critical loading at 100°C, which corresponds to 4 g of methyl iodide per ton of charcoal, would not be unduly restrictive.

Twenty or so common aqueous reagents, including strong oxidizing and reducing agents, soluble silver salts, and amines, were brought into contact with a gas mixture containing methyl iodide in a foam column for a period of 10 sec. None of them produced any worthwhile decontamination at room temperature except acid potassium bromate solution, which allowed a penetration of 1 part in 100.

Copper- and silver-impregnated charcoal showed no improvement over the standard coconut charcoal, but a coal-based charcoal (Sutcliffe Speakman and Co., Ltd., type 207B) was found to have a much higher loading limit. At 100°C a 25-g bed loaded with 300 μg of methyl iodide retained all but 2 parts in 10^5 for 12 hr; loaded with 2 mg, the bed retained all but 1 part in 10^6 for 1 hr.

The coal-based charcoal (type 207B) maintained its improved performance down to room temperature. Not only was a penetration as low as 1 part in 10^4 observed in irradiated fuel experiments, but even when a 25-g bed was loaded with 13 mg of methyl iodide, less than 1 part in 10^4 penetrated in 24 hr of elution. This loading, equivalent to 500 g/ton, may be taken as the permissible loading for reactor purposes.

The results obtained by Collins and Eggleton, discussed above, would seem to indicate that methyl iodide is adequately retained by coal-based charcoal either at 100°C or at room temperature. However, data obtained under different conditions by other investigators are less encouraging. May (results quoted by Chamberlain, ref. *81*) found more movement of iodine on a bed of 207B charcoal swept for 3 to 5 days with air, CO_2, and CO at 250°C or with CO_2 at 200°C than occurred in a bed of coconut-based charcoal under similar conditions at 200°C. McCormack (*159*) tested three (unspecified) grades of charcoal with 50- to 100-μg quantities of ^{131}I-tagged methyl iodide, presumably at room temperature, and found that the iodine activity was rather quickly removed when air was passed through the beds. An elution half-time of 6 to 8 hr was found.

More significantly, Adams and Browning (unpublished data) have tested a sample of 207B charcoal under wet and dry conditions and have found a marked difference in retention efficiency.

The iodine source used in these tests was prepared by passing air over molecular iodine (made by the PdI_2 method) at −78°C for 10 min with a flow rate of 30 cm^3/min. The iodine removed under these conditions was shown by diffusion tube measurements to be predominantly in the form of methyl iodide. This material was trapped in an approximately $1\frac{3}{4}$-in.-deep charcoal bed, and the bed was subsequently flushed with room temperature air at a velocity of 30–35 lin ft/min for 5 hr. When the air was dry, the 207B charcoal had an efficiency of 99.99%. However, when the humidity was raised to about 70%, the iodine retention efficiency under otherwise identical conditions dropped to 74%. A coconut-based charcoal tested with moist air under the same conditions (*160*) showed 97.3% efficiency. Collins and Eggleton's tests (*158*) were apparently made with a dry 95% CO_2–5% CO gas mixture. It appears, therefore, that 207B charcoal does not provide a solution to the methyl iodide under most probable accident conditions, where moisture will be present.

Collins *et al.* (*48*) reported that a bed of Hopcalite, a commercial preparation containing MnO_2 and CuO, retained about 90% of the methyl iodide presented to it, with a bed temperature of 100°C, and that most of the iodine penetrating the Hopcalite bed was molecular iodine which was efficiently trapped by a charcoal bed at room temperature. About 99.2% of the methyl iodide was retained by the two beds in series. Reduction of the efficiency of the Hopcalite resulted from the presence of the reducing gas, CO, which discouraged use of this material in CO_2-cooled reactor systems. Tests with this material for the retention of organic iodides in moist or steam-laden air are being planned at ORNL.

Parker *et al.* (*46*) mention use of a heated (620°C) bed of platinized alumina to oxidize organic iodides, backed up by a room-temperature charcoal bed to collect molecular iodine. The efficiency of this device and its optimum operating conditions remain to be determined.

5.1.5 High-Temperature Adsorbers

The efficiency of charcoal for trapping molecular iodine has been well demonstrated, as indicated by the discussion in Sect. 5.1.2 above, but concern has been expressed about the possibility of ignition of charcoal beds by exposure to high-temperature oxidizing gases during a reactor accident. It would be desirable, therefore, to have alternate extraction materials available that would be less susceptible to destruction under such conditions. Preliminary experiments were performed by Barton *et al.* (*161*) to test two different approaches to the problem: the use of activated alumina containing about 0.5% platinum (a hydroforming catalyst), and the use of iodide salts as a chemical exchange medium for gaseous radioiodine.

It was found that when a 3-in.-deep bed of granular KI was exposed to flowing helium carrying molecular iodine tagged with ^{131}I at 500°C, 53% of the trapped iodine remained on the bed after exposure to flowing helium at 500°C for 3 hr and at 600°C for 4 hr. The gas velocity was 10 lin ft/min throughout the experiment. These experiments demonstrated that exchange occurred between the gasborne ^{131}I and ^{127}I in the crystalline KI at 500°C and that potassium iodide beds are reasonably stable in flowing air at temperatures up to 600°C. However, this approach to the iodine trapping problem is not considered promising, because it would probably be necessary to maintain the bed at a temperature of the order of 250–300°C in order to guarantee efficient trapping of iodine by this material.

Tests with beds of platinized alumina showed relatively little movement of iodine in flowing air at 325 or 425°C, but some loss occurred in 1 hr at 525°C and almost 100% elution occurred during a 1-hr exposure at 625°C. In other experiments (*161*) the retention capacity of platinized alumina was compared with that of unplatinized alumina, using fission-product iodine produced by heating irradiated Al-U alloy specimens. It was found that the platinized material had a significantly higher retention capacity but that it lost about 10% of the trapped iodine during a 30-min exposure to air at 300°C flowing at a velocity of 8 lin ft/min. It appears, therefore, that this material would serve only to delay the passage of iodine at temperatures above 300°C and as a heat sink. Cheaper materials could serve effectively for the latter purpose.

Board and Davies (*162*) reported that activated alumina adsorbed 99.76% of the iodine carried by a stream of dry CO_2 with a bed temperature of 18°C and a flow rate of 40 ft/min. Under the same conditions, except for a bed temperature of 200°C, the efficiency was reported to be 99.56%. Data

on the iodine absorption capacity of this material under wet conditions were not reported. It seems likely that saturation by water vapor would markedly reduce its ability to retain iodine efficiently.

5.2 ENGINEERED SAFEGUARDS – DESIGN OF COUNTERMEASURE SYSTEMS FOR FULL-SCALE REACTOR CONTAINMENT

Efforts toward greater reactor safety by the use of mechanical devices or other engineered safeguards are encouraged by the AEC (*5*). However, only limited credit for engineered safeguards is presently allowed in establishing Reactor Site Criteria (*3*). The dependability of such systems under accident conditions must be demonstrated beforehand. Iodine is one of the principal hazards that these countermeasure systems must provide for, and, consequently, these systems are considered here very briefly.

Engineered safeguards, in addition to the containment enclosure, are classified by Piper (*163*) into four general types: (1) emergency cooling to prevent melting of the fuel materials, (2) filter systems for removing fission products from the containment enclosure, (3) methods for reducing the internal pressure, which in turn reduces leakage to the atmosphere, and (4) provision for two or more barriers around the primary system, which will prevent a major leak of fission-product activity.

Typical engineered safeguards systems applicable to the containment of radioiodine and other fission-product elements are described below.

5.2.1 Internal Water Spray and Dousing Systems

Internal water spray and dousing systems can be used to reduce the pressure by lowering the temperature within the containment vessel, provided there is no possibility of a water-metal or water-graphite reaction. Internal sprays may also serve to scrub fission-product iodine out of the containment atmosphere. The efficiency of spray systems for iodine removal under accident conditions has not been clearly demonstrated. Because of the low solubility of iodine in water, addition of chemicals such as NaOH and $Na_2S_2O_3$ to water used in dousing systems has been suggested (see Sects. 2.2 and 5.1.2).

5.2.2 Multiple Containment

Multiple containment represents a container which is contained. The outer provides holdup and dilution volume for fission products that leak from the inner container. Gases are pumped out of this space, filtered, and passed through iodine adsorbers. Molecular iodine and other reactive or readily deposited forms of iodine should be largely retained in such systems, but iodine attached to small particles or in the form of an unreactive gas may still provide a hazard. The NS "Savannah" provides an example of the multiple containment philosophy.

5.2.3 Pressure-Suppression Containment

The compact primary system of a boiling-water reactor is contained within a dry well, and in an accident the primary steam is vented to the containment shell through a pool of water which also provides an emergency water supply for the core spray system. An example is the Humboldt Bay nuclear plant operated by the Pacific Gas and Electric Company. The extent to which iodine would be retained in the suppression pool is not clear at present.

5.2.4 Containment by Building Structures

Nuclear facilities in which the stored energy in the coolant is small are sometimes contained in buildings that are specially designed to have low leakage rates. Cleanup systems are provided for the gases that may be released into the buildings. These systems are designed to trap particulate and molecular iodine. Examples of this type of facility are the Oak Ridge Research Reactor (ORR) and the production reactors at Hanford. The ORR building has a sufficiently low inleakage that the exhausting system can maintain an internal building negative pressure of 0.3 in. of H_2O. This is "dynamic containment," with the exhausted gases passing through a cleaning system.

5.2.5 Underground Containment

European reactor designers have given considerable thought to the possibilities of underground containment. The Swiss installation at

Lucerne, where underground confinement is used for a 30-Mw (thermal) experimental nuclear power plant, includes a D_2O-moderated reactor cooled with CO_2.

5.2.6 Diffusion Board

Silverman (*164*) at the Harvard University Air Cleaning Laboratory has suggested a porous liner for reactor containment systems. The pores of the honeycomb material are filled with a solid adsorbent material for iodine control. Tests with a diffusion board composed of two layers of 1106B filter paper and activated charcoal in the honeycomb pores gave penetrations of less than 0.01% for 0.08-μ uranin particles and less than 0.1% for ^{127}I vapor when operating at a face velocity of 1 fpm. The possibility of replacing the filter paper with a porous ceramic material is to be investigated.

5.2.7 Foam Suppression Containment

"Foam encapsulation," described by Yoder and Silverman (*165*), involves the use of an aqueous high-expansion foam to encapsulate halogens and particles evolved within a containment vessel. Laboratory studies conducted with a uranin aerosol and ^{127}I vapor have demonstrated that encapsulation and retention of up to 95% of an aerosol are possible. Large-scale studies, conducted by Yoder *et al.* (*172*) at an ORNL test cell, using ^{127}I vapor tagged with ^{131}I produced encapsulation and retention efficiencies of up to 90%. The modest improvement in iodine deposition rate, as compared with the rate in the absence of foam, was not very encouraging.

5.2.8 Internal Filter Systems

Filters for the removal of iodine and particulates are provided to clean the containment atmosphere before a substantial amount of radioactivity leaks out or before treated air is released to the environment. Several nuclear power companies have installed filter systems in the newer plants, and some credit will be taken in calculating the effects of the maximum accident. A number of the systems (Table 5.1) have been tested and show >99.9% iodine retention. However, the general policy is to assume only 95% efficiency for an installed filter–charcoal system until detailed behavior of iodine is better established for maximum credible accident conditions.

The filtering system is usually within the containment envelope, where blowers induce air movement through the filter system. Two important considerations are the general reliability of the blowers, filters, filter housings, seals, etc., and the relative vulnerability of the system to damage from particles, missiles, chemical reagents, vapors, etc.

5.3 METHODS FOR DETERMINING TRAPPING EFFICIENCY FOR IODINE

Methods used in distinguishing different forms of iodine were discussed in Sect. 3. This section

Table 5.1. Engineered Safeguards Installed in U.S. Reactors

Facility	Nature of Engineered Safeguard	Assumed Performance
HWCTR	Recirculating filter system for iodine and particulate removal	Iodine removed at 99.9% from 1000 cfm of air
NS "Savannah"	Once-through filter systems for iodine and particulate removal	Iodine removed at 95% efficiency from either of the 1000-cfm systems
Humboldt Bay	Once-through scrubber for iodine and particulate removal	Iodine and particulates removed at 95% efficiency from 134 cfm of air
EGCR	Once-through filter system for iodine removal	Iodine removed at 95% efficiency; particulates at 99%, both from 750 cfm of air

is devoted to descriptions of methods of evaluating the efficiency of trapping systems proposed for use in reactor confinement systems, under expected service conditions, with emphasis on in-place testing. In-place filter testing is generally recognized as the only valid test method for determining the efficiency of installed filters, but this principle has not been widely applied to iodine trapping systems. Tests of the trapping system of the NS "Savannah," described in Sects. 5.3.1 and 5.3.2 below, represent possibly the best published examples of this type of testing. Smaller-scale tests are needed to screen possible trapping materials under the most realistic conditions possible.

5.3.1 Test Method Using Radioactive ^{131}I

Radioactive ^{131}I has been widely used as a tracer mixed with normal iodine, ^{127}I, to simulate fission-product iodine. Chamberlain (*81*) reported the use of this method by May in testing the efficiencies of various charcoals as iodine adsorbers with the equipment shown in Fig. 5.1.

Iodine was released predominantly as elemental iodine vapor by heating the release tube containing sodium iodide carrier and ^{131}I mixed with potassium dichromate until the dichromate melted. Over 90% of the iodine was released in about 2 min, and the flow of air was continued for a further 3 min. Then the first sampler in the sampling train was replaced by a fresh sampler, the release tube was removed to be gamma counted later, and the injection opening in the pressure vessel was sealed with a steel plug. This operation took about 10 min.

The main flow of air was turned on, 60 liters/min at NTP, and sampling recommenced at 4 liters/min at NTP. The temperature of the charcoal bed was raised rapidly, to the working temperature, usually 250°C, by heating both the air passing into the bed and the walls of the surrounding pressure vessel. Thirty minutes was the usual time taken to reach the required temperature, and the bed was then held at this temperature for a further 90 min. At this point the flow of air was replaced by an equal flow of nitrogen while heat was applied locally to the flange at the base of the

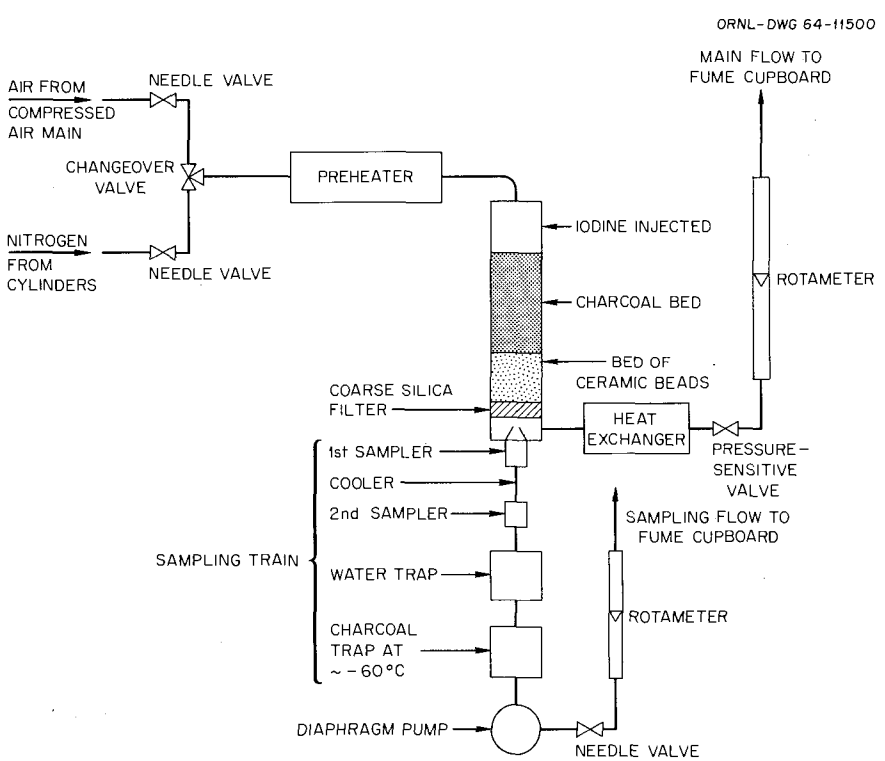

Fig. 5.1. Diagram of Experimental Rig for Determining the Iodine Trapping Efficiency of Charcoal Beds (*81*).

bed to raise the flange temperature to over 350°C. This proved to be necessary to prevent the gaskets from sticking to the flanges, and took about 15 min. All heating of the bed ceased, but the flow of nitrogen was continued for a further 5 min to reduce the temperature and remove all chance of accidental ignition. The flow of gas through the sampling train was stopped at the same time as the main flow. Then the copper cooling tube, behind the first sampler, was replaced by a plug, and the bed was pressurized with nitrogen to cool overnight in an inert atmosphere.

The charcoal bed was removed undisturbed in the inner container, and a scan was made down the bed using a gamma counter with a collimated slit $^1/_{16}$ in. wide.

The scan was made in the following manner. At between 2 and 16 points/in. down the bed, the count rate with the collimated counter was determined in a fixed geometry with a standard deviation of 1% or better. The collimator was then removed and replaced with a solid lead plug, and the scan was repeated. The difference between the count rates was a measure of the number of gamma rays passing down the collimator, and this was plotted against the distance from the top of the bed container.

All components of the sampling train were gamma counted, or washed with a caustic soda solution and the washings gamma counted. In a repeat test carried out on an SC II bed, this method was modified. After the bed had been held at 250°C for 100 min, it was cooled in a flow of nitrogen for 10 min to about 210°C; then the nitrogen flow was stopped and the first sampler in the sampling train was changed. The bed was taken up to

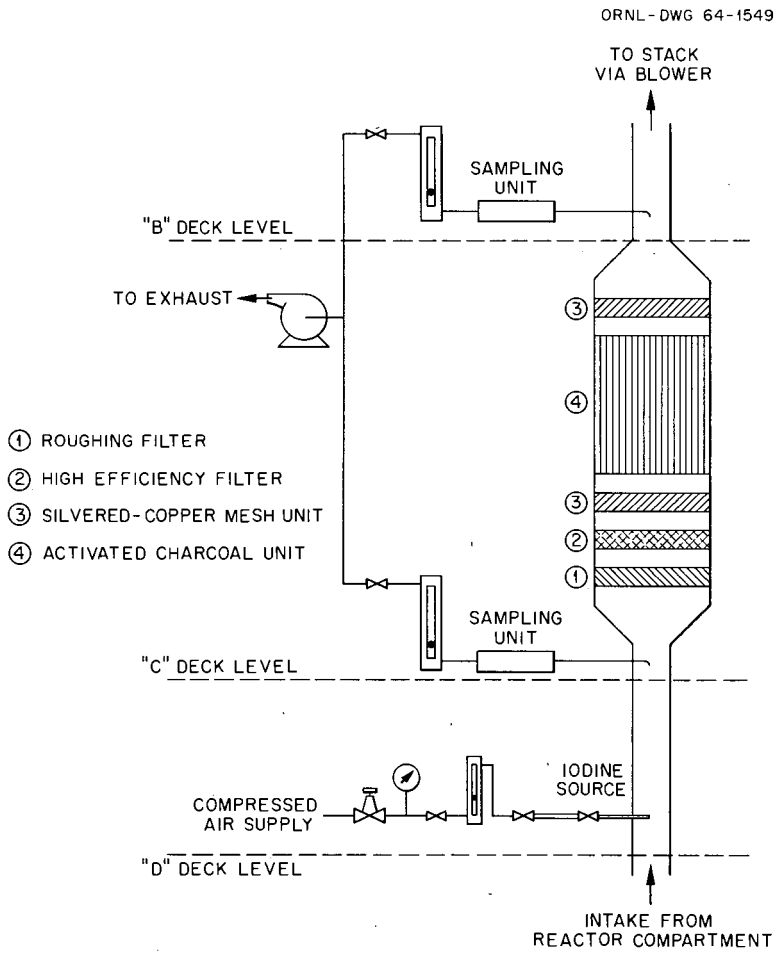

Fig. 5.2. Schematic Diagram of System for Shipboard Testing of Full-Scale Iodine Adsorbers.

working temperature in a flow of air and held at 250°C for a further 2-hr period. The sampler was changed again as before, and a further 2-hr desorption period at 250°C was allowed before the test was completed.

Radioactive ^{131}I has been used by Adams and Browning (*166*) in testing the filter systems on the NS "Savannah." Basically the test was conducted by injecting the iodine vapor (^{127}I tagged with ^{131}I) into the intake of the reactor compartment ventilation system while withdrawing air samples both upstream and downstream of the iodine sorption unit, as shown in Fig. 5.2. These air samples were processed through small sampling units containing activated charcoal to collect any iodine vapor present. The necessary information for computing the efficiency of the iodine sorption unit was obtained by radioassay of the sampling units for ^{131}I.

The success of testing in this manner depends upon penetration of the iodine sorption unit by a sufficient quantity of iodine so that downstream sampling is feasible. To test a highly efficient iodine sorption unit, it is therefore necessary to inject a relatively large quantity of radioactive iodine vapor, so that there is a sufficient amount of radioactivity downstream to measure. On the other hand, a supposedly highly efficient iodine sorption unit could be very inefficient, due to internal damage or faulty installation, and thus allow a major portion of the injected iodine vapor to pass through and be released into the atmosphere. To guard against this possibility while using biologically hazardous ^{131}I, the test procedure was conducted in two steps. The initial test was conducted using a small quantity of radioiodine which could not produce the desired test sensitivity but would demonstrate that a larger quantity of radioiodine could be used safely in the succeeding test. In all cases steps were taken to ensure that the amount of ^{131}I issuing from the ventilation system would not exceed the established limits for the NS "Savannah" operation.

5.3.2 Method Using Nonradioactive ^{127}I and Activation Analysis

Adams and Browning (*166*) have described an activation method for determining the efficiency of an iodine removal system on the NS "Savannah."

Normal iodine is to be preferred over ^{131}I tracer for in-place tests, because of the elimination, by its use, of risk of contamination to the ship. Tests using ^{131}I have had to be performed while the ship is in a relatively unpopulated harbor or at sea, and these tests have sometimes interfered with the operating schedule of the ship. Tests using normal iodine can be performed without risk while the ship is in any harbor or while carrying passengers.

Nonradioactive iodine tests are conducted in much the same manner as the radioactive tests, the main differences being that from 1- to 3-g quantities of elemental iodine vapor (^{127}I) are injected into the duct and that the iodine content of the sampler is determined by neutron activation techniques. While the concentration of iodine used in the test is greater than would occur in a reactor accident, the adsorbers are loaded so far below their capacity that the test is still realistic. The nonradioactive iodine tests are slightly less sensitive than the radioactive tests. Activated charcoal normally contains a small amount of ^{127}I as an impurity, and in most cases the downstream samplers from iodine tests have been found to contain iodine in amounts comparable to the impurity level. To ensure that the charcoal units are not overrated in their iodine efficiency, it is assumed that all the iodine found in the downstream samplers actually came from iodine penetration of the unit under test and was not originally present in the sampler as an impurity. Thus, the iodine efficiencies quoted from the nonradioactive tests represent lower limits and the real efficiencies are in excess of the figures listed.

5.3.3 Testing Charcoal Adsorbers with Freon Gas

The test procedure using ^{131}I, while being very sensitive and producing almost immediate test results, involves the risk of radioactive contamination. The ^{127}I procedure circumvents the contamination problem, but with a resulting loss in sensitivity and with a large increase in the time period from conduction of the test until results are available.

Retention of Freon by activated charcoal was studied on a laboratory scale by Adams and Browning (*166*) as a possible means of frequent nonradioactive testing. A reduction in the time

normally required for a quantity of Freon to penetrate the charcoal unit would be indicative of either the deterioration of the charcoal adsorber or the development of leaks or channels in the charcoal mass. Qualitative information derived from these Freon tests would then be verified periodically by either ^{131}I or ^{127}I tests.

Laboratory tests were conducted under conditions simulating those present during normal operation of the full-scale ventilation system. Freon-12 (CCl_2F_2), -13 ($CClF_3$), -14 (CF_4), and -22 ($CHClF_2$) were used in these investigations. Freon-14 and Freon-22 were dropped from the study because of their very rapid and somewhat erratic penetration of the charcoal test unit. Consequently, Freon-12 and Freon-13 have received the most attention. Experimentally, the length of time (t_{max}) required for a rapidly injected amount of Freon gas (3–5 cm^3, STP) to reach a maximum concentration downstream of a 1.25-in. depth of either PCB or BPL 12/30 mesh charcoal was measured. Air at a superficial velocity of 6.5 fpm was passing through the charcoal mass, which was maintained at 23–24°C. The experimental apparatus used in this study is pictured in Fig. 5.3. The control panel at the right contained provisions for air flow control and Freon injection. The air was humidified by passage through the water vapor generator and then passed into the system. During injection of the Freon, a by-pass arrangement prevented the Freon from contacting the water. Immediately after the injection, the air stream was diverted through the water vapor generator. A General Electric Halogen Leak Detector, set to a sensitivity of 0.1 ppm Freon-12 in air, with a type H-3 probe was used to detect the Freon gas downstream of the charcoal mass under test. The humidity of the system was computed from dew point readings taken with a Weighing and Components Controls, Inc., Dew Point

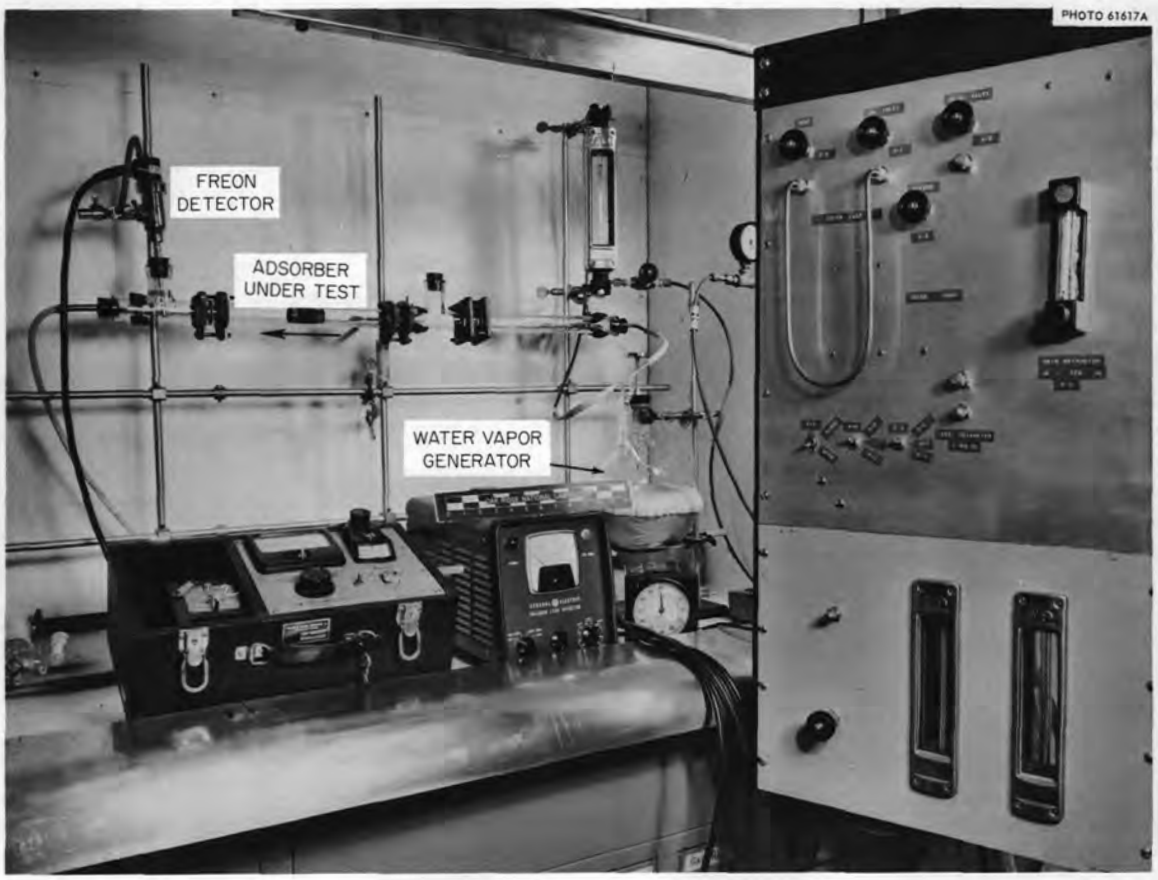

Fig. 5.3. Laboratory Apparatus for Investigation of Freon for Adsorber Testing.

Indicator. With this instrument the dew point is determined by visual observation of the temperature at which moisture condenses on a refrigerated mirror.

Temperature and humidity of the gas-charcoal system were selected as the principal variables for initial study, since it would be difficult to control them aboard ship. Humidity of the gas stream, or more specifically, the water adsorbed on the charcoal surfaces, was found to interfere with the adsorption of the various Freons to a degree larger than anticipated.

Holdup time data obtained on Freon-12 and Freon-13 are displayed in Fig. 5.4. To explain the curves obtained, one must refer to the characteristic adsorption of water vapor by activated charcoal. Unlike desiccant or molecular sieve materials, charcoal physically adsorbs water vapor to an equilibrium level depending upon the humidity of its environment. If the humidity changes, then the quantity of water adsorbed per unit weight of charcoal changes to reflect the new equilibrium relationship. Figure 5.5 depicts this equilibrium adsorption of water vapor by activated charcoals. Note the enhancement of adsorption of Freon in Fig. 5.4 over the humidity range 40 to 60%.

This interference of Freon adsorption by adsorbed water vapor on the charcoal surface is of such magnitude that successful application of this method of testing the charcoal units was considered doubtful. The temperature and humidity of the reactor compartment are subject to wide

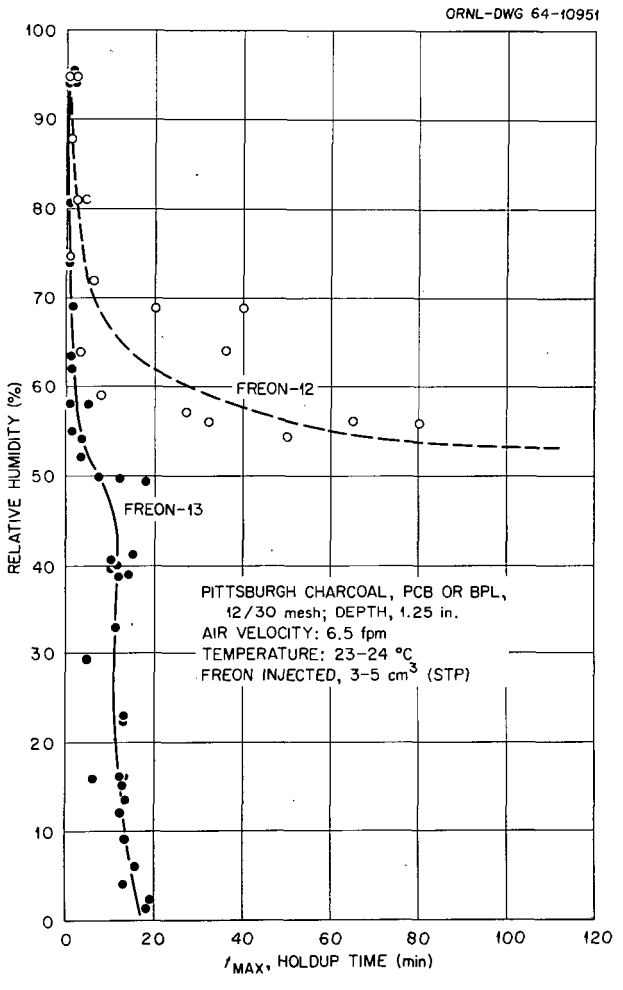

Fig. 5.4. Effect of Humidity on Holdup Time of Freon-12 and -13 by Charcoal.

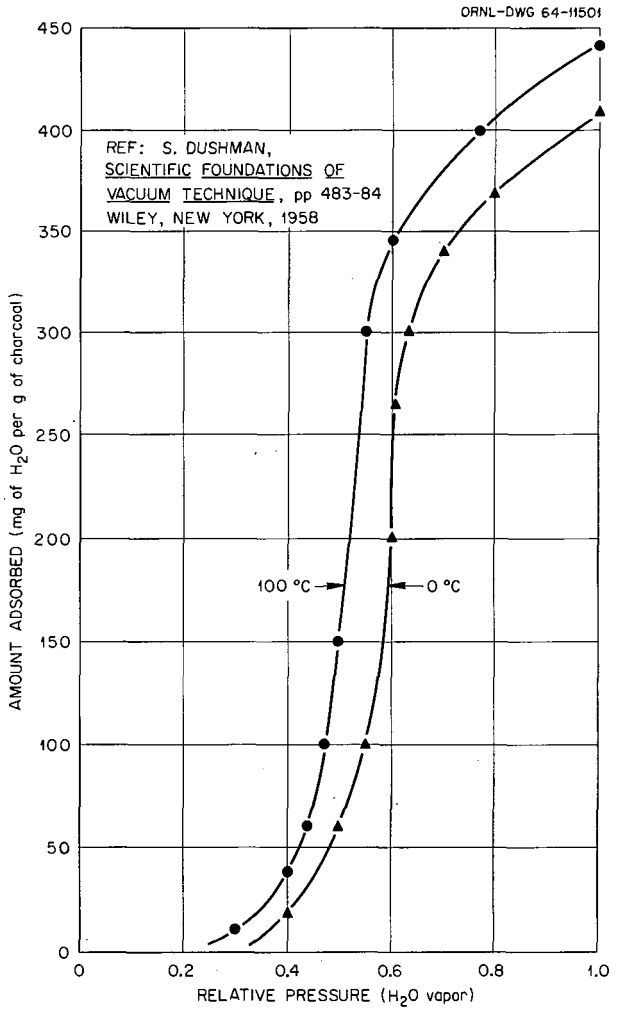

Fig. 5.5. Adsorption of Water Vapor by Charcoal.

variations, depending upon the geographical location of the ship and whether or not the reactor is operating. Because of these changing conditions one would never be certain of the water content of the charcoal in the iodine sorption units. High temperatures and humidities could cause such a reduction in Freon retention that one might suspect damage to the charcoal unit when, actually, none had occurred.

Application of Freon tests under carefully controlled conditions at the Savannah River Laboratory (SRL) has been described by Peters and Muhlbaier (*167*).

Fission products that might be released accidentally into ventilating air of Savannah River Plant (SRP) reactor buildings are removed by passing the air through filters to remove particulates and through carbon beds to remove iodine vapor.

A nondestructive technique was developed by SRL for detecting leak paths in carbon beds. Leak paths, caused by settling or voids in the beds or by leaks around gasket seals, would reduce the halogen adsorption efficiency of the beds. Freon-12 was used as a tracer to detect any leak paths. Freon-12 was readily desorbed from the carbon by normal exposure to the ventilating air stream; however, carbon beds almost saturated with Freon-12 had no loss in efficiency for iodine adsorption.

Freon-12 was injected into the air upstream of the bed to give an inlet concentration of 500 ppm, and the Freon-12 downstream of the bed was measured by an electron-capture-type detector. The usable sensitivity of the detector during routine testing was 0.03 ppm for Freon-12; however, leaks greater than 0.006% of the total air flow were detected readily. The detector is extremely sensitive to halides and practically insensitive to hydrocarbons and other common interferences.

The current technique is said to be suitable for evaluating new activated carbon beds under the following conditions: (1) maximum air velocity, 20 fpm, based on carbon-bed face area; (2) maximum H_2O content in carbon, about 5 wt %; and (3) maximum air temperature, 35°C. The technique was used successfully to evaluate over 800 carbon beds prior to their installation in the containment facilities of the SRP reactor buildings.

Development of a standard test for installed carbon beds has been started at SRL, because of (1) leak paths that can develop in gasket seals during field installation, (2) limitation of water vapor content and air velocity in the current Freon-12 technique, and (3) general lack of a standard for testing carbon beds. The first part of the program, evaluation of other Freon compounds that may reduce current restrictions on the test conditions, is under way.

5.3.4 Tests of the ORR Confinement System with Fission-Product Iodine from U-Al Alloy Fuels

The design of the Oak Ridge Research Reactor (ORR) confinement system is based on the concept that a model confinement system will use filters and charcoal adsorbers in series to trap molecular as well as particulate activity. However, under some conditions extremely small particles, which could penetrate the "absolute" filters presently available, may be formed. The experiments described here were designed to determine the retention efficiency, especially for iodine, of the materials and arrangement proposed for use in the ORR systems.

Since no prior information existed on the size and concentration of particles resulting from melting uranium-aluminum alloy fuels, tests were performed by Parker *et al.* (*161*) to demonstrate the reliability of the confinement filter system by a meltdown of a fuel plate with a fission-product concentration typical of that likely to be encountered in an operating reactor.

Preliminary tests were conducted with bench-size equipment made of quartz and glassware. Two experiments were performed using expected ORR air flow conditions (25 fpm through the roughing filter, 5 fpm through the "absolute" filter, and 50 fpm through two ¾-in.-thick charcoal beds). The results obtained characterized two lots of commercial-grade coconut charcoal, and permitted an evaluation of the iodine retention efficiency of the test system. The results were quite satisfactory, as shown in Table 5.2, although there was some reason to believe that the performance of one of the beds of charcoal, No. 6249, may have been impaired by a slight atmospheric leak around the front cartridge. The results suggest that more than 99% of the iodine was in the molecular state under the conditions employed in these tests. The

Table 5.2. Filter Tests with Radioiodine and Particulates Released by Melting Uranium-Aluminum Alloy (23.6% Burnup) in Air (Sample, ~1 g)

Temperature, 870°C for No. 6249 and 840°C for No. 45
Time at temperature, 60 min
Air flow, 3000 cm^3/min

	Charcoal No. 6249[a]		Charcoal No. 45[b]	
	Iodine (%)	Gross Gamma (%)	Iodine (%)	Gross Gamma (%)
Percent activity on filters[c]				
Roughing (95% aerosolve)	2.0	44.6	1.2	17.6
1st absolute (CWS-6) filter		1.3	0.89	1.7
1st charcoal (¾-in. bed)	97.89	53.8	97.88	80.7
2nd charcoal (¾-in. bed)	0.11	0.1	0.015	0.02
2nd absolute (CWS-6) filter		0.2	0.004	0
Hot charcoal (backup)	0.009	0.009[d]	0.001	0.002[d]
Iodine activity ratio, $\frac{\text{1st charcoal}}{\text{2nd charcoal}}$	867	538	6520	4035
Efficiency of 1st charcoal bed	99.885	99.42	99.98	99.97
Overall efficiency of ORR filter-charcoal system	99.991	99.991	99.999	99.998

[a]4-20 mesh.
[b]4-20 mesh, slightly finer than No. 6249.
[c]Percent of activity reaching filters; gross gamma.
[d]Represents loss through the containment.

overall efficiency of the two charcoal beds in series in both experiments was greater than 99.99%.

Larger-scale tests conducted by Parker and coworkers (*168*) in the Hot-Cell Confinement Mockup Facility showed a 99.97% efficiency for retention of fission-product iodine in both dry and moist air. The aerosol produced by heating highly irradiated alloy specimens in air or a steam-air mixture was allowed to age in a 6.7-ft^3 stainless steel tank for about 45 min before beginning the filter test, which lasted 90 to 100 min. Four charcoal cartridges were provided in the filter pack used in these tests, and the penetration of the first two cartridges could be determined more accurately than in the laboratory tests, where the penetrating iodine was collected in larger traps.

5.3.5 Trapping Tests with Fission-Product Iodine from Irradiated UO_2 Fuel

Collins and Eggleton (*158*) and Collins *et al.* (*48*) have described the design and performance of the trapping system which was used in a high-activity-level test. Steel-clad uranium dioxide fuel (800 g), irradiated to 2000 Mwd/ton and with a total activity of 4000 curies, was placed in an open graphite crucible within a pressure vessel filled with a mixture of CO_2 and CO at 400°C and 25 atm. The use of British gas-cooled reactor conditions and certain other features of the experiment limit the usefulness of the test results. The trapping system consisted basically of a high-efficiency filter for the removal of particulate activity followed by a charcoal bed for the removal

of gaseous iodine. It was necessary to filter the gas before rather than after it passed through the charcoal bed, so that gaseous iodine released by radioactive decay of particulate tellurium aerosols held on the filter could be trapped by the charcoal. The authors state that the gases reaching the trapping system in a reactor can be cooled below 100°C, so this temperature was chosen as being the most stringent condition required for testing the charcoal bed. Since the high-efficiency filter had a relatively low capacity for particulate matter, a coarser pre-filter was also used to prevent clogging. A further high-efficiency filter with associated pre-filter was placed after the charcoal bed to collect any particles of charcoal containing adsorbed iodine dislodged from the bed. The complete arrangement was as follows:

1. A glass fiber pre-filter,
2. A high-efficiency filter of glass fiber paper through which the face velocity of the gas was 5 cm/sec,
3. A bed of coconut charcoal 1.4- to 2.0-mm mesh size, 10 cm deep by 2.5 cm in diameter, operated at 100°C and a face velocity of 70 cm/sec,
4. Another pre-filter,
5. Another high-efficiency filter.

The overall penetration of activity was determined by comparing a sample of the gas stream after the system with one taken before, using the May Pack samplers. As a check, the full flow from the trapping system was passed through a larger charcoal bed, the activity of which was also measured.

This test reproduced certain features of an actual reactor release, such as gas temperature and pressure and the fission-product environment after release. A mild-steel pressure vessel, volume 80 liters, representing the reactor pressure vessel, was connected by suitable valves and pipe work to a scaled trapping system, similar to that described above, containing charcoal at 100°C and thence to mild steel vessels, total volume 1400 liters, representing the reactor containment volume. Gas from the containment volume could be recirculated through the pressure vessel or through the trapping system by means of a pump. All the metal surfaces exposed to the gas were treated with a graphite-loaded paint baked on at 400°C, which is sometimes employed to protect the inner surface of a reactor pressure vessel during construction.

The following sequence of operations, each corresponding to a possible accident condition in a reactor system was carried out.

1. Fission-Product Release from Fuel
   The fuel was heated to 1600–2000°C by an induction heater (this was maintained until the cleanup operation).
2. Initial Blowdown
   After 15 min some gas containing the freshly produced fission products was discharged through the trapping system to the containment volume until the pressure had fallen to 17 atm.
3. Leakage to the Containment
   Further gas was discharged to the containment, this time without passing through the trapping system, until the pressure had fallen to 12 atm.
4. Delayed Blowdown
   After the fission products in the pressure vessel had been allowed to age another hour at approximately 400°C, the remainder of the gas was discharged through the trapping system to the containment volume, reaching a final pressure of 1 atm.
5. Recirculation
   The gas and fission products in the containment volume (previously filled with a mixture of 90% CO_2 and 10% air) were now recirculated for $1\frac{1}{4}$ hr through the trapping system. Periodically they were also circulated through the pressure vessel to examine the effect of heating the fuel in a gas mixture containing air.
6. Cleanup
   The induction heater was switched off, the pressure vessel was isolated, and recirculation through the containment vessel and trapping system was continued for a further 20 hr to represent the condition of a pressure circuit being purged with CO_2 to exclude air from entering a break.

The only activity, apart from rare gases, detected penetrating the trapping system was iodine, the amounts and composition of which are shown in Table 5.3. Little iodine reached the trapping system during the blowdown operations; the majority arrived during the 20-hr cleanup period, when the pressure vessel containing the fuel was

isolated. It must therefore have been adsorbed on the walls of the containment vessel and the pipe work during the blowdown period, and desorbed again later.

From the results of other tests described by the authors, the iodine behavior in the highly active test can be reconstructed. During the initial heating of the fuel in the CO_2-CO mixture, the usual proportions of particulate and penetrating iodine compounds were produced. Since the pressure vessel was hot, most of the iodine compounds decomposed and were adsorbed onto particulate matter and the walls of the pressure vessel. Because of the high graphite temperature,

Table 5.3. Trapping Efficiency of Filter-Charcoal System Tested with Iodine from Highly Irradiated Fuel (*158*)

Operation	Iodine Entering Trapping System (mc)		Total Iodine Leaving Trapping System (mc)
	Particulate Fraction	Gaseous Fraction	
Blowdown	500	50	1
Recirculation	1000	1000	100
Cleanup		8000	900

The fuel contained a total of 20 mg of iodine and 200 curies of ^{131}I.

Table 5.4. Deposition and Transport of Iodine in UO_2 Fuel Meltdown Tests[a]

Location of Iodine	Inventory in Each Location (Percent of Iodine)			
	A	B	C	D
Total release	83.8	63.8	90.9	92.1
Hot zone	6.3	10.6	0.4	0.4
Aerosol tank	47.5	23.8	25.2	34.2
Filter pack				
Roughing filter	15.7	15.1	3.4	0.85
Absolute filter	0.3	0.3	0.5	0.24
Copper screens	4.8[b]	8.1	49.4	2.0
Charcoal paper			4.1	14.7
1st charcoal	5.5	3.0	4.4	39.2
2nd charcoal	2.3	0.2	0.07	0.18
3rd charcoal	0.6	0.02	0.01	0.02
4th charcoal	0.3	0.003	0.002	0.005
5th charcoal			0.0005	0.002
2nd absolute filter	0.00005	0	0.00003	0.00003
2nd copper screens	0.0002	0	0.000005	0.0002
Cold trap backup	0.26	0.04	0.0002	0.0001

[a]Runs A and B were with trace-irradiated UO_2. Run C was performed with 7000-Mwd/ton-irradiated UO_2. Run D was an ^{131}I tracer simulant released by melting unirradiated UO_2.

[b]In run A, bronze screens were used. They were later found to be less efficient than copper screens, which were used in subsequent runs.

a dense particulate fume developed, leading to agglomeration and rapid deposition; consequently little iodine reached the trapping system during blowdown, and that which did was largely particulate. During the recirculation period, the air introduced into the pressure vessel caused desorption of elemental iodine and this then deposited in the cooler pipe work and containment vessel. In the final cleanup period this iodine was slowly desorbed in the form of iodine compounds, which entered the trapping system. The capacity of charcoal to permanently retain methyl iodide is limited, and the mixture of methyl and other organic iodides evidently overloaded the charcoal bed, allowing a significant quantity to penetrate the trapping system.

Parker *et al.* (*46*) have reported results of tests performed with iodine liberated during melting of stainless-steel-clad UO_2 fuel specimens in the CMF (Fig. 3.4). The data given in Table 5.4 show the distribution of iodine in the trapping assembly illustrated by Fig. 3.24. The high retention of iodine by roughing filters observed in experiments with trace-level-irradiated UO_2 is believed to be due to reaction of molecular iodine with filter material, the effect being accentuated by the low concentration of airborne iodine.

Collins *et al.* (*48*) state that testing of the installed trapping plant of the Windscale Advanced Gas-Cooled Reactor had been performed using artificially generated ^{132}I and that further tests were planned. No details were given.

These investigators also examined the occurrence of alkyl iodides both in the laboratory and in large-scale experiments. They say that they had not found a method of producing concentrations of alkyl iodides approaching that which emerged from the pressure vessel in the high-level test described above (several hundred $\mu g/m^3$). They emphasized the role of surface adsorption in the conversion of molecular iodine to charcoal-penetrating organic compounds.

5.4 SUMMARY

The absolute-filter and charcoal-bed combination seems to be generally accepted as the best available trapping system for iodine released from irradiated fuel. The principal shortcomings of this system are the susceptibility of charcoal to ignition and its rather poor retention of alkyl iodides in moist air. The significance of the latter difficulty is not clear at present, partly because the biological effect of organic iodides has not been determined and partly because the fraction of released iodine that may be converted to this form in reactor accidents is not known. In any event, progress in understanding the behavior of alkyl iodides in trapping systems that has been made in the short time since the recognition of their presence in iodine sources and in simulated reactor accidents gives promise of a satisfactory solution to the problem of trapping these materials.

The likelihood of ignition of charcoal beds in reactor accidents is difficult to assess. In situations where a significant probability of this type of failure exists, or where the iodine release that could result from its occurrence would have disastrous results, it would seem prudent to consider the installation of a noncombustible heat sink, such as granular alumina, ahead of the charcoal bed. Beds of this type might also serve to delay the arrival of iodine at the charcoal beds sufficiently to allow at least partial decay of short-lived isotopes and thus alleviate the decay-heat problem. Automatically activated fire extinguishing systems offer an alternate method of minimizing ignition problems.

The effectiveness of some of the proposed engineered safeguards, such as dry-well and wet-well suppression, under realistic accident conditions and, of equal importance, their reliability over long periods of time remain to be determined. Likewise, the behavior of iodine in some of the proposed containment systems will probably not be determined until after the completion of the Containment Systems Experiment at Hanford or other, similar, facilities. Some experimental facilities, such as the NSPP, are equipped to test the effectiveness of filter systems and of sprays for removal of airborne iodine on a fairly large scale.

Final testing of installed iodine trapping systems should be performed with the most realistic iodine sources available, but faults in the trap components or in their installation may be detected by other, less hazardous, means, like the Freon test method. Further study of methods of simulating iodine release from high-burnup fuel in order to improve the realism of such tests with a minimum of hazard seems to be required.

6. SUMMARY, DISCUSSION, AND CONCLUSIONS

6.1 SUMMARY AND DISCUSSION

A significant amount of information on the characterization of released fission-product iodine and on the various processes that affect its transport in containment systems has already been obtained in experimental equipment, and under the specific conditions selected for various tests. On the basis of the information that has been obtained, it appears that the behavior of iodine in a nuclear accident will be very complex. Figure 6.1 summarizes the possible distribution of iodine in accident situations. Its chemical behavior depends on temperature, oxidation conditions, and the types of material that it contacts. The major portion of the iodine is apparently released as molecular iodine under the most probable accident conditions (air or steam-air atmospheres). This active form may combine with or deposit on particulate matter in the reactor atmosphere. Large particles will settle out in the reactor containment shell. The smaller particles, however, will penetrate aqueous scrubber systems and may be only partially retained by

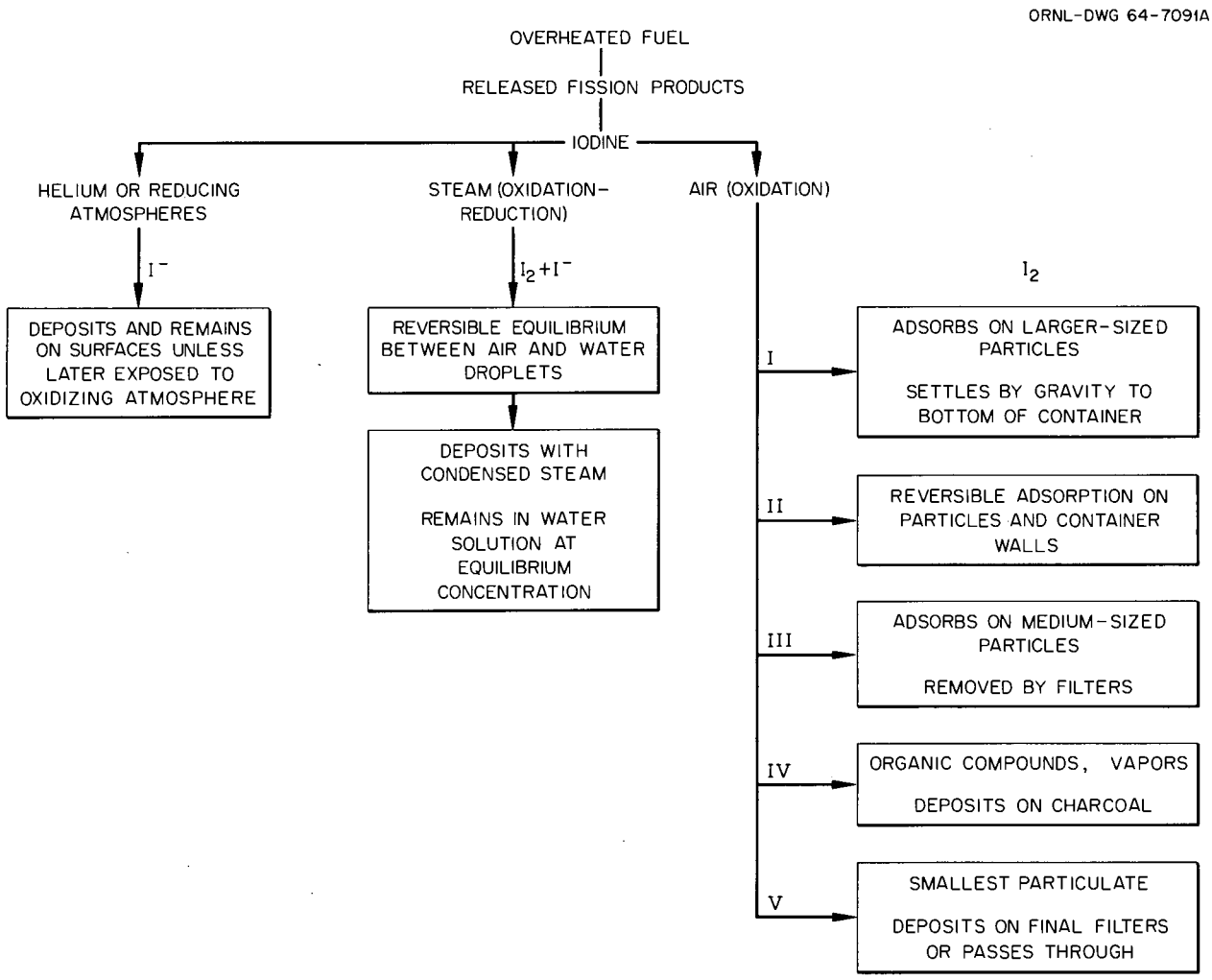

Fig. 6.1. Behavior of Fission-Product Iodine in Reactor Containment Systems.

absolute filter systems. Submicron particles may require secondary filtering or deposition of the diffusion-channel type. There is evidence that some iodine released from melted fuel elements combines with organic materials to form organic iodides. In this state it is relatively inactive and penetrates particulate filters, scrubbers, and, to some extent, charcoal absorption beds.

A large portion of the released molecular iodine apparently will react rapidly with metal surfaces in a reactor containment system. Materials such as plastic coatings and painted surfaces will retain the iodine temporarily. Iodine in this form may be desorbed to some extent by increased temperature and increased oxidation, or by addition of large quantities of carrier iodine. Most of the desorbed iodine can be collected by an absolute filter–charcoal adsorber system, which is currently considered to be the best available fission-product trapping system. Chemisorbed iodine is retained by metals in an inert gas or reducing atmosphere, but may be partially released by later exposure to an oxidizing atmosphere. Airborne iodine that is in the reduced state or is attached to particles larger than $\sim 0.1\ \mu$ is removed by particulate filters. The remaining part, except for that in the form of alkyl iodides or on very small particles, is essentially retained by activated charcoal beds. Catalytic oxidation has been used with some success in changing the alkyl iodides into a chemically active form which is more completely adsorbed by charcoal, but more detailed studies of this technique are needed. Reducing atmospheres such as carbon monoxide interfere with this type of reaction.

The behavior of released fission-product iodine is affected by the surface-to-volume ratio of the containment system. In order to verify extrapolations of the results of laboratory-scale tests to the behavior of released fission-products in actual containment systems, three facilities, each permitting a closer approximation to a full-size reactor meltdown, have been or will be built as indicated in Table 3.1. These facilities are the Nuclear Safety Pilot Plant (NSPP) at ORNL, the Containment Systems Experiment (CSE) at Hanford, and the Loss-of-Fluid Test (LOFT) at NRTS (Phillips Petroleum Company). The NSPP was placed in operation early in 1964. If the fission-product-iodine behavior in these facilities agrees with that predicted by extrapolations of laboratory test data, we will have a much more realistic conception of the consequences of reactor accidents. Thorough evaluations of engineering safeguards will also provide a firm basis for reconsideration of reactor siting criteria. The major fission-product release and transport research programs in the United States, Great Britain, France, and Italy which may contribute useful information in this field are listed in the Appendix.

It is evident, from the studies reported, that elemental iodine vapor can be removed from gas streams by activated charcoal under a variety of conditions with high, or at least adequate, efficiency. It is also apparent at this time that organic iodides present a special problem, since these compounds cannot be removed adequately from gas streams under all conditions. The appearance of these compounds in a variety of iodine sources indicates that this problem is a common one. Laboratory studies are needed of the behavior of methyl iodide (apparently the most prevalent of the alkyl iodides in iodine sources) under conditions simulating those present in reactor off-gas systems following an accident. Activated charcoal is useful under some conditions, but other methods should be studied in order to develop a system that will operate satisfactorily under all conditions. Of interest are modified charcoals, catalysts for the oxidation of the iodides, and reactive chemicals. Determination of the biological half-life of alkyl iodides would aid evaluation of the potential hazard of these materials in nuclear-reactor accidents.

The behavior of radioactive methyl iodide, from a biological standpoint, should be defined and the health physics aspects reviewed, so that the permissible concentration in air can be determined.

Another area of needed study involves the behavior of elemental iodine vapor in atmospheres containing organic vapors, as well as particulate material resulting from vaporization or oxidation of fuel materials. The conversion of elemental iodine into organic iodides while in the atmosphere and adsorption on very small particles can seriously reduce the efficiency of currently used trapping systems. The effect of these factors should be evaluated under carefully simulated reactor accident conditions.

One of the major reasons for limited credit presently being given to engineered safeguards is lack of confidence in the realism of the tests employed to determine their efficiency. Screening tests need to be conducted in small-scale facilities where irradiated fuels can be safely employed to give the maximum degree of accident simulation, and then the results of these experiments should be verified in larger installations that permit closer approximation to conditions in full-scale reactors.

Additional development of devices for characterizing iodine compounds in the field and of methods or devices for periodic testing of installed iodine adsorbers is needed. Nonradioactive simulants for radioiodine would be most useful for these adsorber tests.

Some investigators who are now actively engaged in research on the behavior of iodine have indicated the need for the following studies:

- Reactions of iodine with stainless steel oxide, ceramic material, and other structural materials at high temperatures
- Reactions of iodine in trace quantities with organic compounds
- Loss of iodine from experimental sampling devices by desorption and other effects
- Control of the iodine composition of sources used as test materials for filter systems
- Better materials for diffusion tube experiments in order to improve particle size definition
- Nature of fires in air-ventilated charcoal adsorbers (i.e., full-scale combustion experiments)
- New devices for detection, characterization, and trapping of radioactive iodine

6.2 CONCLUSIONS

A great deal of information is available on the behavior of iodine in various environments, as evidenced by the size of this report and the number of references it contains, but few experiments have been performed that possess all the necessary elements of realism needed for effective simulation of conditions likely to exist in loss-of-coolant accidents in water-cooled reactors. Experiments to be performed in present and projected facilities described in this report should eventually meet the need for this type of data.

The lack of certainty in regard to the chemical form in which iodine will exist in reactor accident environments makes it inadvisable at present to recommend a change in the factor-of-2 credit presently assumed for natural deposition phenomena.

In view of the preceding conclusion, there appears to be a need for greater emphasis on large-scale testing of engineered safeguards under the most realistic accident conditions that can be devised, in order to define their efficiency and to provide more confidence in their reliability. A broad program of laboratory investigations is also needed to aid in designing large-scale tests and interpretation of the results.

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APPENDIX

Table A1. U.S. Fission-Product Release and Transport Research

Contractor	Principal Investigator	Area of Investigation	References
General Electric – ANPD	M. N. Myers T. D. Roberts	Fission-product deposition	*A1–A2*
Battelle Memorial Institute	A. Abriss G. E. Raines	Fission-product deposition	*A3–A5*
Brookhaven National Laboratory	A. W. Castleman, Jr.	Characterization of released fission products	*A6–A8*
Savannah River	A. H. Peters G. H. Prigge	Removal of fission products in containment enclosures	*A9–A11*
Illinois Institute of Technology	J. Rosinski	Removal of fission products in containment enclosures	*A12*
Harvard School of Public Health	L. Silverman	Removal of fission products in containment enclosures	*A13–A15*
Hanford Atomic Products Operation	E. R. Irish R. K. Hilliard	Fission-product release and transport	*A16–A20*
	L. C. Schwendiman	Fission-product transport	*A21–A23*
	H. W. Heacock	Fission-product confinement	*A24–A26*
Arthur D. Little, Inc.		Fission-product removal	*A27*
Mine Safety Appliance Corp.	S. J. Rodgers J. W. Mausteller	Fission-product release	*A28–A29*
Phillips Petroleum Company (NRTS)	T. R. Wilson J. M. Waage	Snaptran	*A30–A32*
	A. A. Wasserman A. H. Spano	Power Burst Facility (PBF)	*A33*
	T. R. Wilson G. B. Matheney	Loss-of-flow Test (LOFT)	*A34–A35*
Sandia Corporation	M. Cowan	Fission-product transport	*A36*
Dow Chemical	P. D. Erickson	Fission-product removal	*A37*
Lovelace Foundation	J. C. Gallimore T. T. Mercer	Fission-product characterization	*A38*
Oak Ridge National Laboratory	W. B. Cottrell G. W. Parker W. E. Browning, Jr. L. F. Parsly R. A. Lorenz	Fission-product release, characterization, transport, deposition, and removal	*A39–A47*

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Table A2. UKAEA Fission-Product Release and Transport Research

Location	Principal Investigator	Position and/or Branch	Area of Investigation
Risley		Authority Health and Safety Branch	
	F. R. Farmer	Head, Safeguards Division	Reactor siting
	W. H. Irvine	Engineering – Head, Containment Group	
	J. R. Beattie	Safeguards Division	Evaluation of reactor accident hazards
	R. O'Neil	Reactor Assessment Group	Safety of pressurized water systems
London	I. G. K. Williams	Administrative Office	
Harwell		Authority Health and Safety Branch	
	W. G. Marley	Radiological Protection Division	
	C. M. Nicholls	Head, Chemical Engineering Division	
	A. C. Chamberlain	Head, Aerosol Group	
	W. J. Megaw	Physics	Physics of very fine particles
	F. G. May		
	J. K. Findlay		
	G. Jackson		
	J. B. Morris		
	R. D. Wiffen		
	P. Goldsmith	Meteorology	Diffusiophoresis
	A. E. J. Eggleton	Physical Chemistry	Iodine compounds
	J. K. Dawson		
	W. H. Denton	Chemical Engineering	Charcoal systems
	K. Stewart	Assistant, Head of Safety Coordination of the Weapons Group from Aldermaston	Plutonium combustion
	B. Cox	Chemistry Division	Metal-water reactions
Culcheth	R. W. Nicholls	Deputy Head, Reactor Materials Laboratory	
	Watkins	In charge of steel and zirconium work	
	N. S. Thumpton	Superintendent of Mechanical Effects Group	
Windscale	Ken Saddington	Head, Reactor Development Laboratory	
	R. D. Collins	Research Manager – Reactor Safety	Charcoal filter removal efficiency and fission-product release
	D. A. Collins		
Winfrith Heath	R. D. Smith	Head, Fast Reactor Physics	Static and kinetic physics work
	J. Codd		Theoretical work on Doppler coefficients
	G. H. Kinchin		Thermal reactor systems
	Ray Cox	Head, Instrumentation and Controls Division	
	G. H. Palmer	Head, Radiological and Safety Division	
	J. F. Croft		Iodine release experiments
	R. S. Iles		Iodine release experiments
Berkeley	Jones	Plant Supervisor	Nuclear power station operation
	Grimshaw		Details of natural U (Magnox type fuel) gas-cooled reactors

Table A3. CEA Fission-Product Release and Transport Research

Location	Principal Investigator	Position and/or Branch	Area of Investigation
Saclay	Jean Bourgeois	President, Subcommittee on Reactor Safety	
	F. de Vathaire	In charge of Reactor Safety Group	Kinetics of gas-cooled reactors, scram devices
	A. Meunier		Heavy water reactors
	Costes	Mechanics Section	Fuel fires, shock effects on fuels, pressure vessel rupture tests, prestressed concrete vessel tests, mechanical effects
	A. Barthous		Fission-product release
	M. Laffaire		Fission-product release
	J. M. Lavie		Fission-product release
	A. Barthous F. Billiard M. Gras G. Chevalier J. Brion J. Miribel	Service de Controle des Radioactiff	Iodine trapping
Grenoble	Paul Ageron		Melusine and Siloe Reactors, safety work
	Elberg		CABRI Reactor
Cadarache	J. Aillous M. Chatoux F. de Vathaire J. P. Millot	In charge of CABRI Program	

Table A4. Ispra and Euratom Fission-Product Release and Transport Research

Organization and Location	Principal Investigator	Position and/or Branch	Area of Investigation
Ispra, Italy	V. Raievski C. Rinaldini	Head, Physics Group, Reactor Safety	TESI Reactor
Euratom, Brussels	Willen Vinck	Euratom Directorate for Industry	Safety evaluation and approval of reactors prior to construction
	M. Fermet	Euratom Directorate General for Industry	Fission-product diffusion studies
	V. Raievski		Purpose and design features of TESI Reactor
	Sebille		Radiation embrittlement to pressure vessel studies, Ispra and ORGEL reactors

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