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SUPPRESSION POOL SCRUBBING FACTORS FOR
POSTULATED BOILING WATER REACTOR
ACCIDENT CONDITIONS

INFORMATION REPORT

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ABSTRACT

The incorporation of a suppression pool in the Mark I, II and III containments with a Boiling Water Reactor provides a mechanism that will be highly effective in absorbing fission products should they be released from the reactor vessel following a degraded core accident. The Mark I, II and III containments are configured such that for most accident sequences fission products released from the reactor vessel will enter the drywell and be transported to the suppression pool where they will be absorbed or scrubbed. This report focuses on the scrubbing efficiency of the suppression pool for iodines and particulates.

Iodine and particulate retention factors are presented which account for the effects of suppression pool scrubbing during postulated BWR accident scenarios. The expected behavior of iodine is based on: observations during reactor accidents and destructive tests; normal BWR operating experience; and small and large scale scrubbing tests in water pools. A brief review of iodine chemistry is presented to explain the chemical behavior of iodine and the expected chemical form of iodine released from LWR fuel. Based on the data presented and the expected BWR transport conditions, suppression pool scrubbing factors greater than 10^2 for elemental iodine and particulates, and factors much greater than 10^3 for cesium iodide are currently justifiable for subcooled pools. For saturated pools, scrubbing factors of at least 30 for elemental iodine, and 10^2 for particulates and cesium iodide are currently justifiable. Scrubbing factors several orders of magnitude larger may be demonstrated after more experimental testing. Use of these attenuation factors for the analysis of postulated BWR accidents will provide conservatively realistic estimates of the consequences of severely degraded accidents in performing probabilistic risk assessments.

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1.0 INTRODUCTION

In degraded core accident scenarios, the presence of water in the fission product transport pathways provides an important mitigator to minimize the quantity of airborne fission products. The General Electric Boiling Water Reactor (BWR) uses the pressure suppression primary containment system to provide a water barrier to fission product migration. Thus, significant retention in the pool of radioiodines and other fission products, except noble gases, is expected and must be accounted for in any realistic risk assessment.

In the BWR primary containment configuration, fission products released in a degraded core accident would be transported by steam from the reactor vessel to the suppression pool. The routing would be via the drywell and the horizontal vents from the drywell to the suppression pool in the case of a large break accident. During a transient, the fission product transport with steam from the reactor vessel would be directly to the suppression pool via the safety/relief valve discharge lines and quenchers in the pool. During a combination of a transient and a small break accident, transport would be via both pathways.

The historical licensing basis, as exemplified by Regulatory Guide 1.3 for a loss of coolant accident, has never recognized fission product retention in the suppression pool. The Guide specifies that in a design basis accident, it shall be assumed that 25% of the radioiodines in the core are instantaneously transported to the primary containment air space. This quantity is specified as being 91% elemental iodine, 5% particulates, and 4% methyl iodide. Current information indicates that iodine released from overheated fuel is in the form of inorganic iodides, and that the assumptions in Regulatory Guide 1.3 of the quantity of elemental iodine and methyl iodide are far in excess of any likely expectation. Experimental test data indicate that chemical forms similar to the inorganic iodides and particulates that would be expected to be released from degraded cores would be retained in pools of water like the suppression pool and would not escape into the primary containment air space.

A review of reactor accidents and experimental tests indicates that the presence of water plays an important role in limiting the quantity of iodine and particulate fission products available for leakage to the environment. In addition, natural processes, which are not dependent on operator action, also limit the transport of most fission products during the various accident scenarios. Such processes include plateout, fallout, washout, and retention in leak paths. The overall decontamination factors for postulated BWR scenarios result from the combined effects of pool scrubbing and the other removal processes. The primary focus of the present review is on suppression pool scrubbing.

In this study relevant data from the literature on iodine and particulate transport through pools of water are presented and documented. These data are used as a basis for establishing iodine and particulate pool scrubbing factors for use in probabilistic risk assessments. For the purpose of this report, particulates are defined as all fission products other than noble gases or halogens, except that inorganic iodides are evaluated either as a vapor or as a particulate.

This study focuses on the following areas: (1) dominant BWR fission product transport and removal mechanisms; (2) iodine chemistry and iodine behavior in water; (3) the expected chemical form of iodine released from LWR fuel; (4) behavior of iodine as observed during actual reactor accidents and experimental destructive tests; (5) behavior of iodine in operating BWRs; (6) iodine and particulate decontamination factors from experimental tests related to suppression pool scrubbing; and (7) postulated BWR accident scenarios and expected decontamination factors.

This study demonstrates that the pressure suppression pool in the BWR containment is an important barrier which significantly limits the quantity of iodine and particulates available for release to the environment during postulated reactor accidents. Properly accounting for this barrier will sharply reduce the off-site consequences calculated in probabilistic risk assessments.

2.0 SUMMARY OF RESULTS

A literature search was conducted to identify and document the chemical form of iodine in fuel; the behavior of iodine during reactor accidents, experimental destructive tests, and normal BWR operation; and the removal of iodine and particles by pools of water. The available data base was evaluated. A summary is given below with a detailed evaluation presented in subsequent sections of this report.

2.1 Chemical Form of Iodine in LWR Fuel

The chemical form of iodine released from LWR fuel has been recommended by three independent investigators to be CsI . During BWR accident conditions, the conversion from CsI to I_2 is not expected because of the predominantly reducing environment in the reactor vessel and drywell.

2.2 Behavior of Iodine During Reactor Accidents, Experimental Destructive Tests, and BWR Normal Operation

Observations from three light water reactor (LWR) accidents shows that iodine released from the plant was always much less than the available iodine released from the fuel. The release of iodine was never instantaneous but occurred over a period of several days. The presence of water in the plant systems during the course of the accidents limited the amount of iodine and particulates released. Significant amounts of iodine were released when a wet-reducing environment was not present.

The predominant chemical forms of iodine in the reactor water during normal plant operation are I^- , HIO , and IO_3^- . Iodine carryover from reactor water to steam, as measured by relative concentration in condensate to that in reactor water, varies from 0.2 to 2.5%. Iodine (HIO) scrubbing factors in the main turbine condenser during normal plant operation are on the order of 10^3 . Suppression pool scrubbing factors for HIO have been measured to be greater than ten thousand.

2.3 Removal of Iodine and Particles by Pools of Water

About one hundred technical papers were reviewed from which eleven applicable experimental tests were identified which investigated the effects of fission product retention by pressure suppression pools. The majority of the tests used elemental iodine (I_2). There also were some data on the scrubbing of CH_3I , HI , HIO , and small insoluble particles. There were no reported data on the transport of CsI through a pool of water, however the behavior of CsI as a vapor or as a particle may be inferred from test results using similar chemical forms. Table 7.3.1 presents a summary of the experimental test conditions and measured decontamination factors¹ for each test.

2.4 Evaluation of Data Base

Review of the experimental data base indicates that it can be used to conservatively bound BWR transport and retention conditions existing as a consequence of degraded core accidents. While several tests were reasonably representative of the transport phenomena expected during a postulated accident, the experimental conditions (e.g., shallow pool, small particles, volatile I_2) led to lower pool decontamination factors (DFs) than would be expected in BWR containment system conditions. Dominant BWR fission product transport mechanisms during accident scenarios were identified and the experimental data were assessed for applicability to expected conditions. Minimum pool scrubbing DFs which the current data base can support, and the potentially attainable DFs which could be supported by further testing, are presented for each dominant transport sequence.

The application of pool decontamination factors in risk assessments is presented in Section 9.

Footnote 1: The decontamination factor or "scrubbing factor" is defined as the ratio of fission product mass transported into the pool to the mass which is released from the pool surface.

3.0 CONCLUSIONS

Suppression pool decontamination factors appropriate for use in PWR risk assessments are presented in Table 3.1. Based on the data presented and the expected BWR transport conditions, suppression pool decontamination factors of at least 10^2 for elemental iodine and particulates, and 10^3 for cesium iodide are justifiable for subcooled pools. For saturated pools, decontamination factors of at least 30 for elemental iodine and 10^2 for particulates and cesium iodide are currently justifiable. It is felt that these minimum values can be increased several orders of magnitude by further testing for conditions more representative of the suppression pool during post accident conditions.

Natural processes such as the agglomeration of solids, plateout, deposition, washout, etc., also play an important role in limiting the quantity of fission products available for leakage to the environment. The overall attenuation factor applicable to BWR degraded core accident scenarios includes both the effects of pool scrubbing and of such natural removal processes that will occur in the various volumes of the BWR process system and its multiple containment system.

TABLE 3.1 MINIMUM SUPPORTABLE AND POTENTIALLY ATTAINABLE
SUPPRESSION POOL DECONTAMINATION FACTORS FOR
IODINE AND PARTICULATES

Transport Pathway and Associated Event(s)	Minimum Supportable DFs		Potentially Attainable DFs (3)
	Subcooled Pool (1)	Saturated Pool (2)	
Reactor pressure vessel to pool via safety relief valve and quencher (Transients)	10^3 CsI, I^- , HI 10^2 particulates 10^2 I_2	10^2 particulates (4) 30 I_2	10^5-10^6 CsI, I^- , HI 10^3-10^6 particulates 10^2-10^3 I_2
Reactor pressure vessel to pool via vents (Transients following RPV depressurization, or LOCA post blowdown period)	10^3 CsI, I^- , HI 10^2 particulates 10^2 I_2	10^2 particulates (4) 30 I_2	10^4-10^6 CsI, I^- , HI 10^3-10^6 particulates 10^2-10^3 I_2
Aerosol Transport to Pool Via Vents (Core-Concrete Vaporization Release)	10^2 particulates 10^2 I_2	10^2 particulates (4) 30 I_2	10^3-10^6 particulates 10^2-10^3 I_2

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- (1) During these conditions, complete condensation is expected when the pool is subcooled.
- (2) A subcooled pool is at a temperature below the saturation temperature corresponding to the pressure in the containment, while in a saturated pool steady state boiling "steaming" is occurring.
- (3) Potentially attainable by further testing (saturated-subcooled pools).
- (4) Includes CsI

4.0 MAJOR BWR FISSION PRODUCT TRANSPORT AND REMOVAL MECHANISMS

In this section, a description of the major BWR design features is presented. Postulated BWR accident scenarios, fission product transport and removal mechanisms, and expected transport parameters are presented in order to assess the applicability of the pool scrubbing experiments presented in Section 7.

4.1 BWR Description

General Electric Boiling Water Reactors use a multiple containment system featuring a pressure suppression primary containment of the Mark I, II, or III geometry. A schematic of the current design GE-BWR/6 Mark III system is shown in Figure 4.1. The direct cycle begins with feedwater entering the reactor vessel. Recirculation pumps and jet pumps circulate reactor water through the core region where boiling occurs. A saturated steam/water mixture at about 1050 psia and 550°F passes through a series of steam separators and dryers. The dried steam flows out of the reactor vessel via the main steam lines to the turbine. Steam from the turbine is exhausted to the main condenser. The resulting condensate is pumped through a full flow condensate treatment and feedwater heater system where it is purified and preheated prior to re-entering the reactor vessel.

In the Mark III design, the reactor pressure vessel is enclosed by a drywell structure. A pressure suppression pool containing about a million gallons of water together with the drywell, forms an additional barrier between the reactor vessel and the primary containment air space. Both the drywell and the pool significantly retain fission products released during postulated accidents. The primary containment and the secondary containment shield building fully enclose the drywell, the suppression pool, and the reactor system.

In the BWR containment system, fission products released in a degraded core accident would be transported by steam from the reactor vessel to the suppression pool. In the case of a loss-of-coolant accident, the routing would be via the drywell and the horizontal vents from the drywell to the suppression pool. The five-foot thick drywell wall is penetrated by submerged horizontal vents (27.5' in diameter) which have center lines located at three levels below the surface of the pool, with a minimum depth of seven feet. There are 40 circular vents on each of the three levels.

During a transient or small break event, fission product transport with steam from the reactor vessel would be directly to the suppression pool via some of the 18 to 20 safety/relief valve discharge lines. The steam is released into the suppression pool via quenchers as illustrated in Figures 4.1, 4.2, 4.3, and 4.4. Each quencher ensures rapid condensation of steam in a subcooled suppression pool by discharging the steam through more than a thousand small holes which are approximately 1 cm in diameter.

4.2 Fission Product Transport Mechanisms During Degraded Core Accidents

Degraded core accident scenarios address events where the reactor core may be damaged significantly releasing substantial quantities of fission products from the core. Degraded core accidents are extremely improbable. If an event is initiated by a transient or a primary system break, extensive or total failure of safety and emergency core cooling systems must also occur. This section presents major BWR accident scenarios considered in probabilistic risk assessments and describes the fission product transport mechanisms expected for each scenario. These scenarios are of two major types. Class 1 and 3 considers those where core damage initiates loss of primary containment integrity by overpressure

and cracking. Class 2 and 4 considers those where loss of primary containment integrity by cracking initiates core damage. In Class 1 and 3 scenarios, the suppression pool is subcooled while in Class 2 and 4 the pool is in a saturated condition. At the time of core damage in Class 2 and 4 scenarios, the pool is not expected to be turbulently boiling, but rather in a "steaming" condition.

Within each of these two major accident types, BWR accidents can be further grouped into two categories: (a) discharge through the horizontal vents, and (b) discharge through the S RV discharge lines. Figure 4.2 shows the expected fission product pathway for each of these categories.

During a postulated design basis LOCA a double-ended break of a recirculation line is assumed, resulting in the rapid depressurization and blowdown of the reactor vessel to the drywell and then to the suppression pool by way of the horizontal vents. The resulting high drywell pressure signal will initiate the upper pool dump to the suppression pool.¹ For degraded core conditions to occur, the emergency core cooling system (ECCS) must be assumed to fail resulting in reactor fuel heatup and subsequent damage and melting. Prior to clad melting, hydrogen may be produced from the (Zr-steam) metal-water reaction. If degraded ECCS performance is assumed to continue, melting of the core may continue to propagate, and the melted portion may slump into the bottom head and result in additional steam generation from the water remaining in the lower portion of the reactor vessel. As the cladding melts, fission products released from the fuel may be transported (with the steam and hydrogen) to the drywell by way of the break in the recirculation piping during the post-blowdown period. Fission product transport in a predominantly steam environment can occur from the drywell to the primary containment air space through the suppression pool. The fission products would have to pass through approximately

Footnote 1: The upper pool dump is initiated by any one of three signals: high drywell pressure, low reactor water level, or low suppression pool water level.

thirteen and one-half feet of water¹ before reaching the suppression pool surface. Flow only through the first level vents is expected as steam flow at this time late in the scenario is only about five pounds per second per vent. Approximately 10% to 20% of the vent cross-sectional area on the drywell side is expected to uncover, initially producing bubbles of various sizes. Nearly all of the volatile fission products in the fuel are expected to be released (in a steam environment) during this stage of the accident sequence.

For postulated transients where ECCS makeup water is assumed to be not available, fission products released from the fuel to the reactor vessel may be transported directly to the suppression pool in a predominantly steam environment via the safety-relief valve discharge line and quencher system. In these events a low reactor water level or a high drywell pressure signal will initiate the upper pool dump to the suppression pool. In these events, the fission products must pass through approximately nineteen feet of water before reaching the primary containment air space. Nearly all of the volatile fission products in the fuel are expected to be released (in a saturated steam or superheated steam environment) during this stage of the accident sequence. In the case of small or intermediate size break accidents, both transport pathways occur.

Following either a LOCA or a transient event, the molten core and other material may penetrate the thimbles on the reactor vessel bottom head and drop to the floor of the drywell in the restricted area directly under the vessel. Under LOCA sequences this is not a violent event since the reactor vessel is already depressurized. However, for the transient cases where no large LOCA break exists and when the operator has failed to depressurize the vessel in

Footnote 1: In sequences where ECCS operates as designed, the expected depth of submergence is about 8½ feet.

order to make the low pressure ECCS available¹, there may be additional transport from the vessel to the drywell and suppression pool via the horizontal vents. Vessel depressurization is not expected to occur instantaneously and violently, but instead the depressurization should occur rather slowly as the core material slowly penetrates through the control rod and instrument thimbles and perhaps eventually through the bottom head itself.

Following the vessel penetration sequence, a molten core-concrete interaction may occur on the portion of the drywell floor directly under the vessel releasing aerosols and noncondensable gases such as CO_2 . Figure 4.5 illustrates where this interaction may occur. Note that the very substantial reinforced concrete structure around this central portion of the drywell floor area prevents any direct access of this molten core-concrete mixture to the region of the suppression pool in the BWR Mark III containment system. In addition, features assuring the structural integrity of the suppression pool include the five-foot thick drywell wall, the ten to fifteen foot thick floor under the pool and drywell, and the eight foot thick wall around the pool outer radius. Ground elevation is above the pool water level, as indicated in Figure 4.5.

The noncondensable gases generated will slowly pressurize the drywell and eventually vent through the horizontal vents (top row only) to the suppression pool. Only a small fraction (about 1%) of the vent cross-sectional area on the drywell side is expected to be uncovered producing relatively small bubbles. During this stage, fission products (vaporization release component) attached to aerosol particles may be carried by a CO_2 -steam mixture through the vents and depending upon the accident scenario released under $8\frac{1}{2}$ to $13\frac{1}{2}$ feet of water.

Footnote ¹: The Emergency Procedure Guidelines instruct the operator to depressurize the vessel if the high pressure emergency core cooling systems are not maintaining an adequate vessel water level.

For those fission products which are not absorbed by the water prior to reaching the suppression pool surface, transport may occur in the primary containment building. In the event that the containment pressure continues to increase due to loss of decay heat removal or non-condensable gas generation, General Electric analyses indicate that catastrophic failure of the containment is not expected. The most probable cause of loss of primary containment integrity for the Mark III Standard Containment design would be cracking near the top of the containment structure. Buckling and cracking would be expected in the area where the transition from the vertical wall to the dome is located (refer to Figure 4.5). Therefore, fission products (gases or particulates) released from the pool surface must travel for approximately 130 feet from the pool surface through a tortuous path of stairway gratings and compartments with significant plateout surfaces before reaching the location of the cracks in the primary containment steel. After leakage through the cracks, additional travel through the shield building will occur, with exhaust through the standby gas treatment system and/or directly outside through cracks or other leakage points. Significant areas for plateout and deposition are available in the shield building and the other volumes within the secondary containment zone.

A summary of the dominant BWR fission product transport modes and significant transport parameters is presented in Table 4.2.1. The transport and subsequent removal of fission products by the suppression pool is an expected and highly probable occurrence during most postulated degraded BWR accident sequences. The pool is a large passive system and its capacity to retain fission products should not be underestimated or neglected in any risk assessment.

In addition to removal by pool scrubbing, natural processes in all BWR compartments within the reactor system, the drywell, and the primary and secondary containment air spaces will limit the quantity of fission products available for release to the environment. These mechanisms include: agglomeration in the reactor vessel,

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plateout/deposition, aerosol agglomeration, washout, and retention in leak paths. The overall fission product attenuation factor for a BWR will include the effects from both pool scrubbing and all such natural removal and retention processes.

DWR FISSION PRODUCT TRANSPORT MODES AND
TRANSPORT PARAMETERS DURING DEGRADED CORE ACCIDENT SCENARIOS

TRANSPORT/REMOVAL MODE	REACTOR CONDITIONS PSIG/TEMP °F	FISSION PRODUCT RELEASE MODE FORM OF FISSION PRODUCTS	TRANSPORT RATE	FLOW RATE	FLOW QUALITY	ORIFICE SIZE/ BUBBLE SIZE	DEPTH OF POOL FOR SCRUBBING	POOL PH MIN-MAX	POOL TEMP °F MIN-MAX
1. <u>TRANSIENTS</u> RELEASE FROM RPV DIRECTLY TO POOL VIA SRV LINE AND QUENCHERS	1050/550	Core Melt Release CsI, HI, I-(a) Particles (b) Xe, Kr gas CH ₃ I	Slow and Steady, or Intermittent	100-200 lb/sec. 20-40 lb/sec ft ²	steam/H ₂ steam, H ₂	<1 cm Small Bubbles <1 cm	18.8'	7-10	100-220
2. <u>LOCAS</u> RELEASE FROM RPV TO DRYWELL AND POOL VIA VENTS		↓	Slow and Steady After Blow Down	100-200 lb/sec 0.6-1.2 lb/sec ft ²	steam- H ₂ -air	27.5" vent ~10-20% uncovered small bubbles	8.4-13.5'	7-10	100-220
3. <u>RPV PENETRATION</u> and Blowdown to drywell and pool via vents	1050/550 - 200/380	↓	Slow and Steady	- -	steam- air	↓	8.4-13.5'	~10	150-220
4. <u>CORE-CONCRETE REACTION</u> Transport to drywell and pool via vents	N/A	Vaporization release aerosol particles (c)	Slow and steady	40 lb/s CO ₂ 0.24 lb/s f ₂	CO ₂ -air- steam	27.5" vent ~1% uncovered small bubbles	8.4-13.5'	~10	150-220
5. <u>NATURAL REMOVAL PROCESSES</u> Plateout, deposition	N/A	All F.P. except Xe, Kr CH ₃ I	Depends on holdup time (0.5-8 hrs)	N/A	N/A		N/A	N/A	N/A

(a) in steam

(c) as ~2 μm particles

(b) Cs, Rb, Te, Sr, Ba, Ru, La

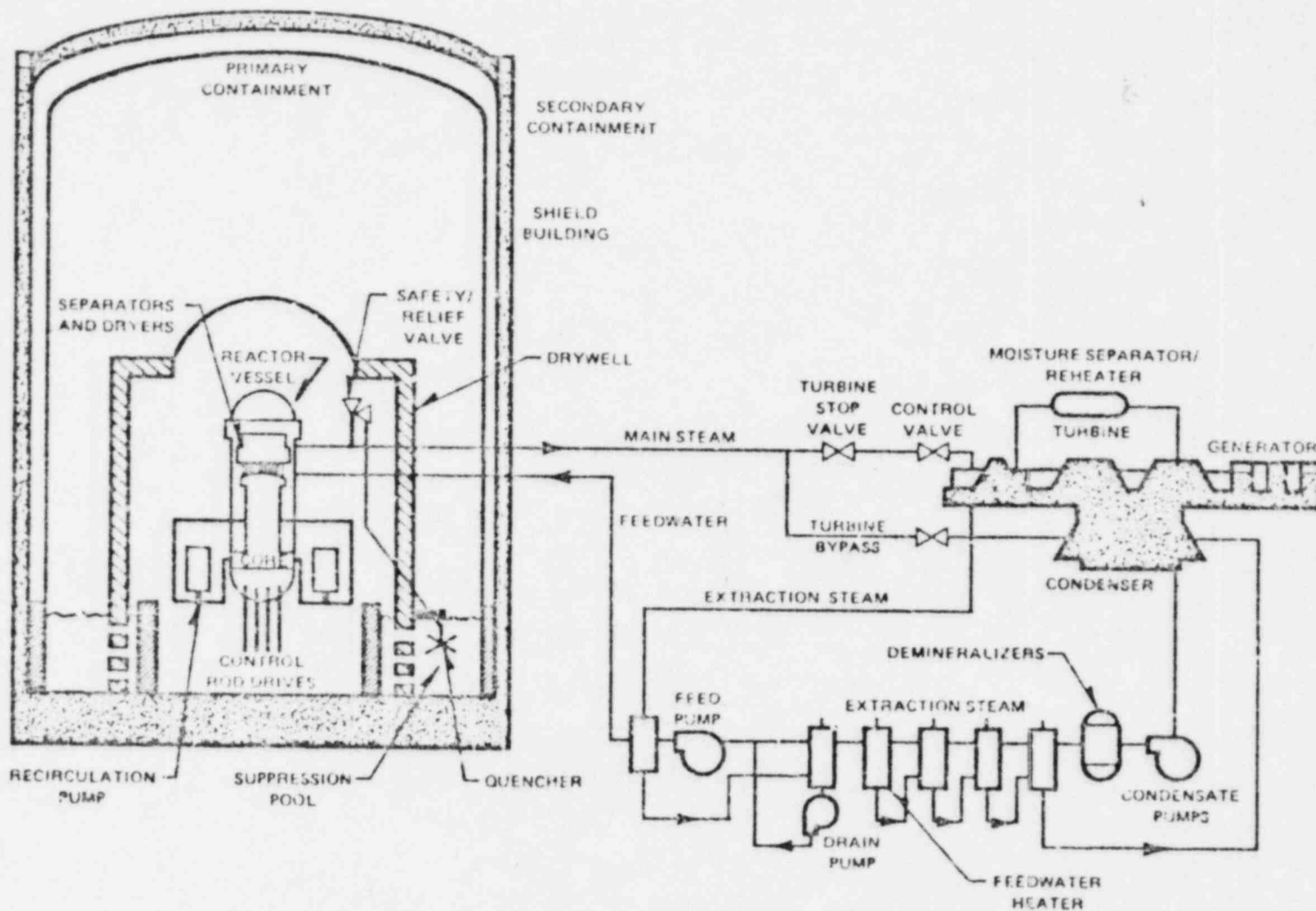
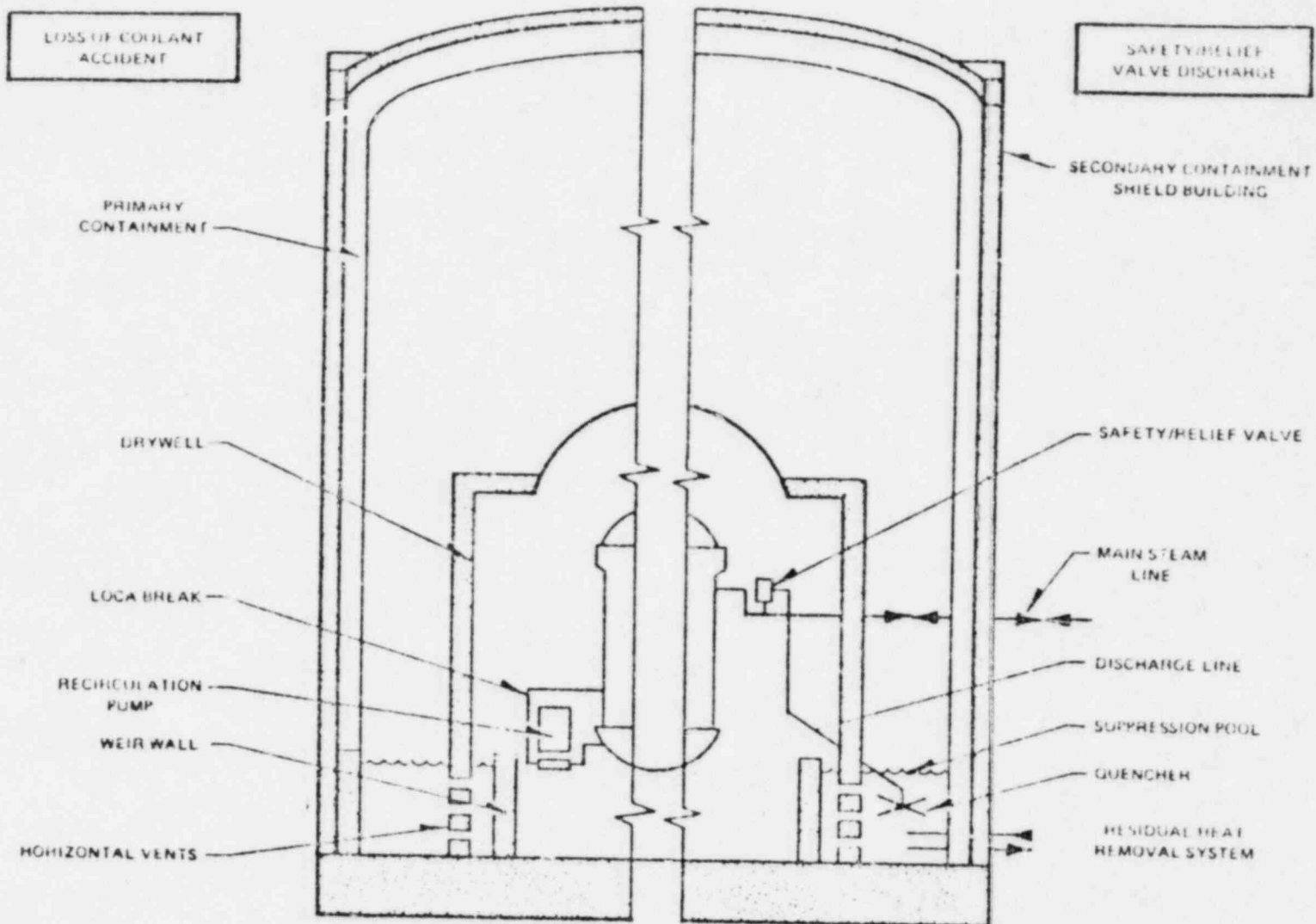


Figure 4.1. Direct Cycle 2WR Nuclear System



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Figure 4.2 Suppression Pool Scrubbing Pathways

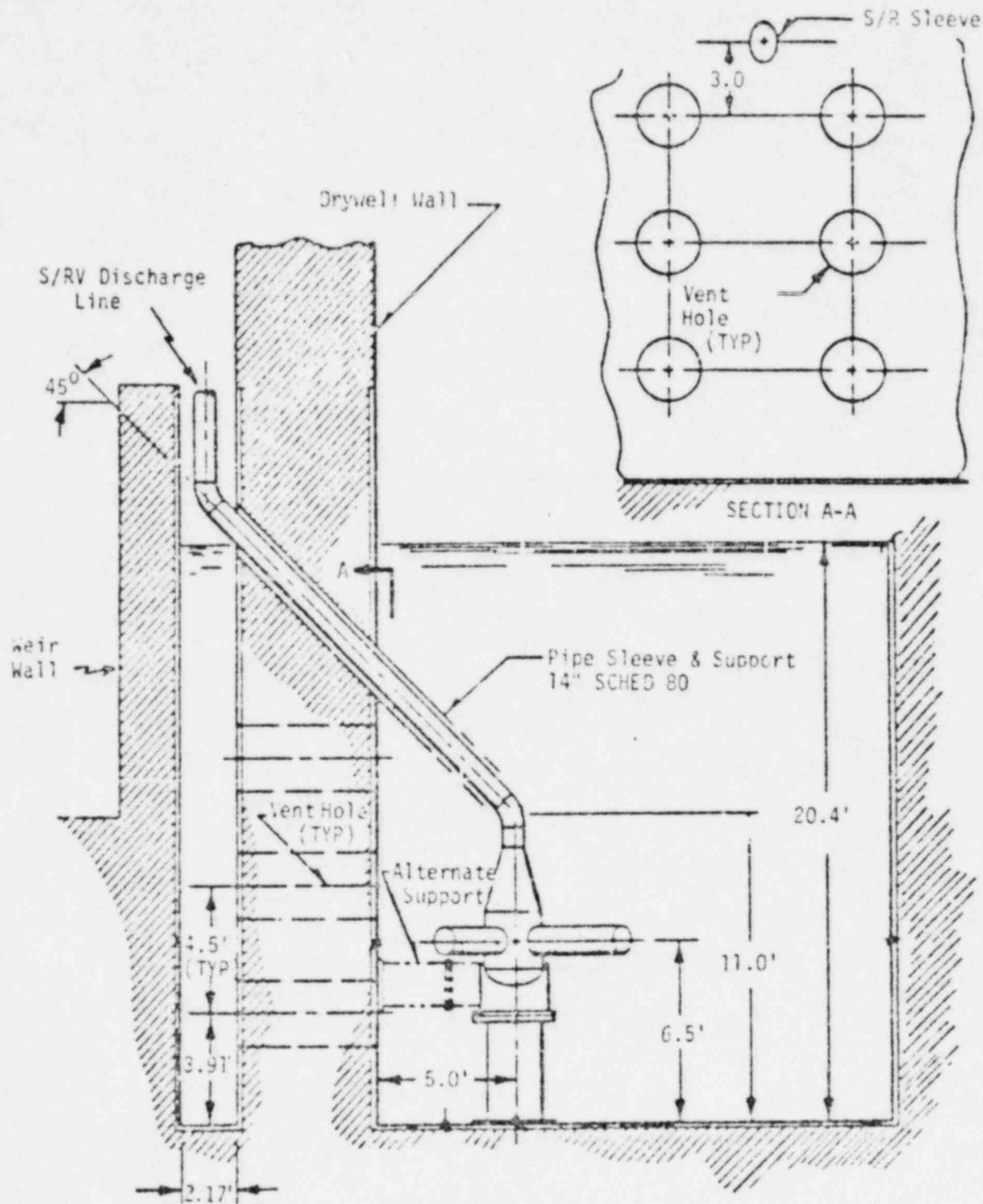


Figure 4.3 Standard Mark III 238 Quencher Arrangement Elevation

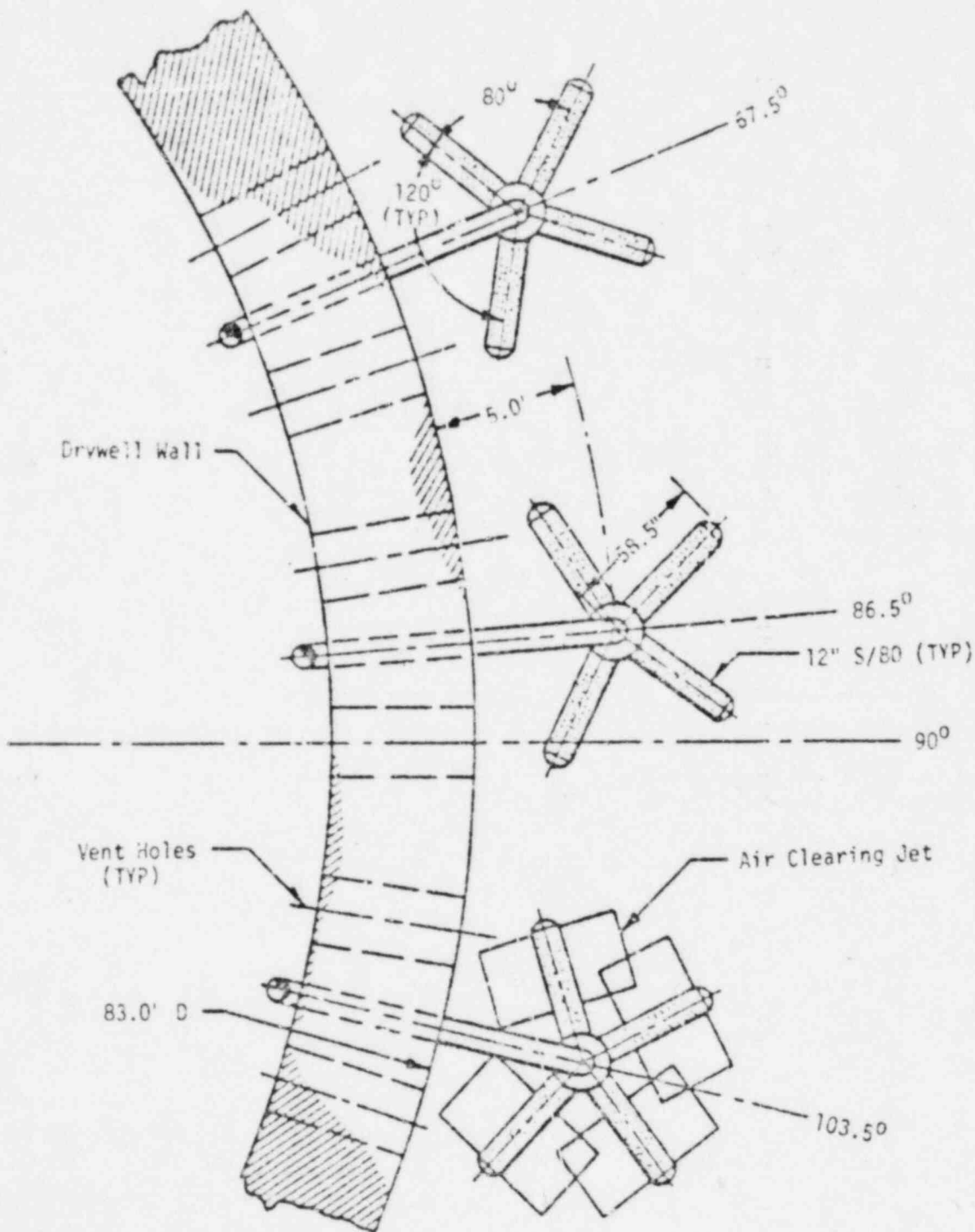


Figure 4.4 Mark III Quencher Plan View (TYP) 238 Plant Arrangement Shown

Figure 1

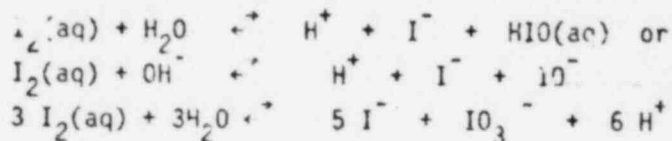
4-13/4-14

5.0 Iodine Chemistry and the Chemical Forms of Iodine in LWR Fuel

5.1 Review of Iodine Chemistry

The behavior of iodine as observed in reactor accidents, normal BWR operation, and during experimental tests involving suppression pool scrubbing, can be better understood by a brief overview of the chemistry of iodine. A complete review of iodine chemistry is beyond the scope of this report. However, a summary of iodine chemical forms and their expected reactions, solubility and volatility, is useful in assessing appropriate decontamination factors to be used in degraded core accident analyses. In such analyses the chemical forms of iodine of interest include elemental iodine (I_2), organic iodides (e.g. CH_3I), cesium iodide (CsI), hydrogen iodide (HI) and hypoiodous acid (HIO).

Elemental iodine (I_2) is volatile at room temperatures, dissolves slightly in water, and may be rapidly hydrolyzed in aqueous solutions by the following reactions:



The degree of hydrolysis depends on I_2 concentration, pH and temperature.

Dissolved iodide (I^-) might be oxidized to molecular iodine in acidic solutions by oxygen in the containment atmosphere, by oxygen dissolved in the water or by radiation effects in the water. Radiation induced chemical reactions in aqueous iodine solutions have been reported (5.1).¹ At concentrations $\leq 10^{-5}M$ (3 ppm), I^- ions can be nearly quantitatively oxidized to IO_3^- with hypoiodous acid, HIO , as a possible intermediate, in solutions with pH below 9 under the influence of high intensity ($5 \times 10^6 R/hr$) gamma radiation. (This

Footnote 1: The notation (5.1) refers to Reference 5.1

mechanism is believed to play a dominant role in producing volatile I_2O during normal BWR operation as will be shown in Section 6.3.) Under degraded core conditions if all the iodine is conservatively assumed to be released from the fuel and reactor system, the maximum iodine concentration in a BWR suppression pool would be approximately 3 ppm or $2 \times 10^{-5} M$ (5.2). The pH of the suppression pool is expected to be ~ 10.5 under such an assumption because of the large quantities of Cs (elemental Cs, Cs oxides, or CsI) also released from the fuel (5.2, 5.3). Also, the pool would not be subject to a high intensity radiation field. Thus, air oxidation or radiation induced oxidation of I^- to I_2 is not very likely to occur in a suppression pool.

The formation of organic compounds of iodine has been studied by several investigators (5.4). Elemental iodine (I_2) was the predominant chemical form of iodine investigated in these studies. The conversion rate of I_2 to organic iodide would be expected to be much larger than the rate for I^- to organic iodide because of differences in volatilities. This occurs because high airborne concentrations of I_2 promote the conversion to organic iodide while low concentrations limit the conversion. Organic iodide (CH_3I) was reported to readily hydrolyze in basic solutions of water at high temperatures ($T=100^\circ C$) (5.5, 5.6).

As cesium iodide is non-volatile and highly soluble in water, it should be effectively retained in a suppression pool. Under degraded core conditions, cesium iodide vapor would be expected to condense into a particulate form at temperatures less than $500^\circ C$. This condensation may occur on the inside surfaces of the reactor pressure vessel or onto other particulate surfaces airborne in the reactor vessel. The transport and retention of a particulate by a suppression pool then becomes important when considering the removal processes of cesium iodide.

Hydrogen iodide (HI) is volatile and is rapidly hydrolyzed in water with a partition coefficient near infinity. Thus, it should be effectively retained in a suppression pool.

Hypoiodous acid (HIO) has never been chemically identified. It is a volatile and highly soluble compound which is rapidly hydrolyzed in aqueous solutions. Thus, it should be effectively retained in a suppression pool.

5.2 Chemical Forms of Iodine in LWR Fuel

The chemical form and behavior of fission products in LWR fuel has been assessed by Adamson (5.7). The fission products Cs, Rb, I, Br, Te, and Se are classed as "volatile-reactive" and their behavior resembles that of the gaseous fission products Xe and Kr at temperatures above 1400°C. The "volatile-reactive" fission products tend to migrate down temperature gradients in the fuel through cracks, etc., and condense or react in the core fuel regions (<1000°C). Since Cs and I are both aggressive chemicals, they may combine to form CsI. The fission yield of Cs is 10 times greater than that of iodine, therefore, the probability that all the iodine could combine with Cs is relatively high.

The boiling point of iodine (I_2) is 184°C, while the boiling point of cesium iodide (CsI) is 1230°C. Since the average clad temperature of LWR fuel is 300°C, I_2 , if present inside the gap, should behave like a fission gas but CsI should not.

In gap purge experiments at ORNL (5.8) fission products were purged from irradiated commercial fuel rod segments at temperatures up to 1200°C. The conclusions from the experiments were that the dominant chemical form of iodine in the fuel is a metal iodide, probably CsI. Crystals of CsI have been identified on the internal cladding surface of an irradiated commercial fuel rod (5.9) by X-ray fluorescence. Other papers also support the conclusion that the chemical form of iodine in LWR fuel rods is cesium iodide (5.10, 5.11, 5.12, 5.13).

An iodine "spiking" release phenomenon from cladding defects has been observed (5.14) when reactor power or pressure is suddenly changed. The spiking of fission noble gases is distinctly different from that of iodine, indicating that the chemical form of iodine released from the fuel is not gaseous elemental iodine.

Organic iodides exist in extremely small amounts under accident conditions. In the TMI-2 accident they amounted to about 0.003-0.01% of the core fission product inventory (5.15, 5.16, 5.17). It is the dominant form of iodine in the gas phase not because there is a large amount present, but because the species that are present in large amounts (the iodides) distribute overwhelmingly into the water phase, leaving only trace quantities in the gas phase. Radiation effect in water may contribute to the formation of organic iodides but the reaction rate is relatively slow. Measurements of iodine in operating BWRs show that very little if any organic compounds of iodine are released from LWR fuel (6.9).

Based on the available data in the literature, experience from reactor accidents (Section 5.0), operating BWRs, and the expected moist-reducing environment in the primary system and drywell, it is concluded that the chemical form of iodine released from LWR fuel is cesium iodide and this species should be used in assessing suppression pool iodine scrubbing factors during BWR degraded core accident scenarios.

6.0 Behavior of Iodine During Reactor Accidents, Experimental Destructive Tests, and BWR Normal Operation

In this section observations are presented on the behavior and transport of iodine and particulates during reactor accidents and small experimental reactor destructive tests. The behavior of iodine in operating BWRs is also presented to illustrate two significant removal processes in a BWR: removal of iodine by condensation processes in the condenser; and scrubbing of iodine by the suppression pool.

6.1 Fission Product Releases From Damaged Reactor Fuel

6.1.1 The Accident at TMI-2

TMI-2 is a pressurized water reactor (PWR) and its containment system does not have the pressure suppression pool which is an integral part of the BWR design. The loss-of-coolant accident at TMI-2 resulted in the release of large amounts of fission products to the reactor coolant and the reactor building containment. A stuck open pressurizer relief valve resulted in the transport of primary system water directly to the containment where it subsequently flashed (see Figure 6.1). It is postulated that extensive core damage occurred when the water level dropped below the top of the active fuel allowing fission products released from the fuel to be transported in the saturated water-steam mixture directly to the containment. Although the magnitude of the radioactive material released during this accident was significantly higher than any previous accident in the commercial nuclear power industry, only modest amounts of iodine escaped from the primary containment.

The President's Commission on TMI-2 (6.1) reported that the total release of iodine 131 (I-131) to the environment from March 28 through April 27 was ~17 curies. This release was insignificant compared to the 7.5 million curies of I-131 which was discharged

from the RPV. As noted in Table 6.1 only 2000 Ci of I-131 became airborne in the reactor building. While measurements were not available immediately after the accident, measurements made several months later showed that the only iodine airborne was organic iodine which is relatively nonreactive. It is evident that the activity remaining airborne was a very small percentage of that released demonstrating that iodine released into a humid environment is readily removed by natural phenomena. Also the majority of the iodine-131 activity remained in the water.

In view of these findings, the President's Commission recommended that "a study should be made of the chemical behavior and extensive retention of radioiodine in water, which resulted in very low releases of radioiodine to the atmosphere."

6.1.2 The SL-1 Accident (5.2)

The SL-1 was a small natural circulation reactor (3 Mwt). The fuel elements were constructed of highly enriched uranium-aluminum alloy sandwiched by aluminum alloy cladding. Compared to a present day commercial power reactor few engineered safety systems existed in this prototype portable military reactor. A sudden manual removal of a central control rod by maintenance personnel caused a rapid reactivity insertion which lead to a power excursion and extensive core melting. A very large fraction of the core was melted and most of the water was ejected from the reactor vessel, yet less than 0.5% of the I-131 core inventory was released to the outside atmosphere. While SL-1 was contained in a "Butler" type structure, which has minor containment capabilities compared to containment structures associated with commercial power reactors, only 10 Ci of I-131 were released to the atmosphere during the first 16 hours and 70 Ci over the next 30 days. The expelled water was highly contaminated with fission products, including radioiodine.

6.1.3 Crystal River-3 Event (6.3)

Crystal River Unit 3, a PWR, experienced an event on February 26, 1980, in which 40,000 gallons of primary coolant water was discharged onto the floor of the closed containment building. An electrical equipment failure resulted in a stuck open power operated relief valve. With the reactor scrammed and the power operated relief valve open, the reactor vessel pressure dropped as the reactor coolant discharged into the reactor coolant drain tank and then flashed into the reactor containment building after the drain tank rupture disk actuated.

The reactor water contained both I-131 and Xe-133. The Xe-133 was immediately released into the containment atmosphere, but the I-131 was observed to be retained to a large extent in the water. The I-131 in the containment atmosphere peaked in about four days as shown in Figure 6.2.

Following the event, the containment air and the water in the containment sump was analyzed for I-131. Approximately 1.8 Ci of I-131 was airborne on March 2, 1980 while approximately 85 Ci of I-131 was in the sump water (6.4) as shown in Figure 6.3. Thus the decontamination factor was approximately 55 days after the incident. A higher DF may have existed on the day of the incident but this could not be evaluated due to lack of data.

The primary phenomenon in this event and in the TMI-2 accident appears to be the flashing of primary reactor water with the subsequent release of fission products to the containment. In spite of this flashing phenomenon, decontamination factors on the order of 50 were observed for iodine.

6.1.4 Windscale-1

The accident at Windscale-1 (a once-through, air cooled, uranium-graphite reactor) occurred when the uranium metal fuel overheated, caught fire, and released fission products to the air stream flowing up a tall stack (6.5). The stack filter was estimated to have removed 2.5×10^4 Ci of particulate iodine, however, 7.0×10^4 Ci of gaseous iodine was released from the stack to the environment. The released iodine represented 12% of the available iodine inventory in the core. Water was not present at Windscale since it was an air cooled reactor, thus accounting for the large fraction of iodine released.

6.2 Experimental Reactors Tested to Destruction

Three experimental reactors have been deliberately tested to destruction to verify that large reactivity excursions were self-limiting and would automatically terminate the nuclear reaction. The tests were designed to violently disassemble the core and melt or vaporize part of the reactor fuel. Dispersion of the radioactive material was monitored to provide information to assess the dispersal areas. Three tests of this nature were the BORAX-1 test (6.6), the SPERT-1 test (6.7), and the SNAPTRAN tests (6.8).

The BORAX-1 experimental reactor was located in a low pressure water tank partially sunk into the ground. There was no building over the reactor. Motion pictures taken during the tests showed that the water tank holding the experiment burst and most of its contents were ejected into the air. All the fuel fragments could be accounted for within 350 feet of the reactor. A wind speed of 8 mph was present during the test yet the phenomenological mechanisms limiting dispersal operated to attenuate the release of radioactive fission products.

The SPERT-1 destructive experiment was also conducted in an open tank facility. It was covered in a light structure not intended for containment purposes. In the test approximately 35% of the core was melted with nearly all the fuel elements in the core experiencing melting to some degree. Metallic debris approximately 100 μm in size was recovered in the water. Only noble gases escaped to the atmosphere from the reactor tank. The radioactivity released to the atmosphere was estimated to be 2.4×10^5 Ci; which represented less than 1% of the fission product inventory in the core. Iodine was detected only in the reactor water.

The SNAPTRAN series of destructive tests were conducted to evaluate the risk of launching the SNAP-10A reactor (a U-Zr hydride, NaK cooled reactor, designed for space applications) over water or land. In the SNAPTRAN-2 test, no water was present and 70% of the iodine was released to the atmosphere. In contrast, when the reactor was placed in an open tank of water (the SNAPTRAN-3 test), there was no detectable iodine released to the atmosphere. Nearly all remained in the water, in the fuel or plated out.

6.2.1 Summary of Observations on Iodine Behavior

In the reactor accidents and destructive tests involving an initial water barrier very little iodine released from the fuel was released to the atmosphere because of retention in the water which was present. Iodine tended to remain soluble in water rather than escape as a volatile gas. This behavior is consistent with that observed from suppression pool scrubbing experiments where iodine (in steam) is passed through a pool of water. Results from these tests will be presented in Sections 7.0 and 8.0.

6.3 Behavior of Iodine in Operating BWRs

The behavior of iodine in operating BWRs is presented in this section and includes: the chemical form of iodine in BWR systems;

removal of iodine by condensation processes in the condenser; and removal of iodine by suppression pool scrubbing during High Pressure Coolant Injection Surveillance tests. Decontamination factors for each of these processes are presented to illustrate how the separation of iodine (HI) from steam favors the water phase rather than the gaseous phase. This behavior would also be expected to occur during BWR accident conditions when steam carrying iodine (HI or CsI) would be rapidly condensed and scrubbed as it passes through the suppression pool.

6.3.1 Chemical Behavior of Radioiodine in BWR Systems (6.9, 6.10)

An examination of the chemical forms of iodine at operating BWRs during normal plant operation and during shutdown conditions indicates that I^- , IO_3^- , and perhaps HI are the predominant species in reactor water. Only traces of I_2 , and organic iodine (CH_3I) were detected and are believed to be from contamination in the analytical procedure. Results of analyses of reactor water for iodine are shown in Table 6.2. During reactor shutdown with no steam distillation, most of the iodine was found in the iodate form in the reactor water. This is probably a result of radiation induced oxidation of I^- to IO_3^- or IO_4^- , since it is well known that I^- can be oxidized by IO_3^- by the influence of radiation in aqueous solutions (5.1).

The amount of radioiodine carried over by the steam has also been investigated. The term "carryover" is defined as the ratio of iodine concentration in the condensed steam to the concentration of the same isotopic species in the reactor water. The moisture content in the steam is very small as a result of the action of the moisture separators and steam dryers in the BWR design. Based on the distribution of nonvolatile ionic species, such as Na^{24} , between the condensate and reactor water, the iodine carryover by mechanical processes, e.g., entrainment, is estimated to be $\leq 0.05\%$. The observed iodine carryover, however, ranges between 0.2 to 2.5% (6.11), therefore, mechanical processes alone cannot account for all the iodine carried by the steam. This suggests that some volatile iodine species must be involved in the carryover process.

Volatile HIO is believed to be the major iodine species distilled from the primary coolant during normal operation and is believed to be produced by the radiation induced oxidation of I^- found in the primary coolant (6.9). Essentially all the iodine species which are carried by the steam from the reactor water, through the turbine, and to the condenser are condensed with the steam in the hotwell. Only trace noncondensable iodine species are found in the gas phase which consists mainly of radiolytic gases. Since HIO is expected to be the major volatile species in steam carryover, it should be expected in the steam condensate. This, indeed, has been shown by examining the chemical forms of iodine in the condensate of operating BWRs (refer to Table 6.3). Very little I_2 , IO_3^- , or organic iodide were found in the condensate samples, and essentially all the iodine was I^- or HIO . Unfortunately, the chemical identity cannot be distinguished between the I^- and HIO forms.

The iodine species in the offgas have been sampled and results are shown in Table 6-4. The majority of the airborne radioiodine is in the organic form. The source of organic iodine and the mechanism of its formation in the offgas has not been precisely identified. The offgas iodines, however, are usually "older" than those found in the reactor water and condensate. This is shown in Table 6-5 where the isotopic distribution of the offgas isotopes are compared with the distribution in the reactor water. This apparent aging is probably due to the delay incurred between the iodine deposition on material surfaces, such as carbon steel, and the subsequent formation and release of organic iodine.

Organic iodide formation may be related to the production of CH_4 , as a result of steel corrosion and carbide/hydrogen interaction (6.12). The conversion of organic iodide from I_2 in 80% humidity air in contact with galvanized steel at ambient temperatures has been recently reported (6.13).

6.3.2 Separation of Iodine in the Condenser

As was shown in Table 6.3, the majority of the volatile radioiodine carried by the steam distributes overwhelmingly into the water phase in the condenser leaving only trace quantities of organic iodide in the gas phase. The iodine separation factor (S.F.) in the condenser is defined as:

$$S.F. = \frac{\text{transport rate of iodine into condensate } (\mu\text{Ci/sec})}{\text{release rate of iodine in non-condensables gas phase } (\mu\text{Ci/sec})}$$

The separation factor has been calculated (6.14) from field measurements at operating BWRs (6.15). Table 6.6 shows that separation factors for removal by condensation range from 140 to 5580 for inorganic iodine (HIO) and from 80 to 2460 for total iodine (I_2 , HIO, and organic iodine).

The removal of iodine in the condenser by condensation processes is expected to be very similar to the process which occurs when steam carrying radioiodine is rapidly condensed by the suppression pool. Suppression pool scrubbing, however, is expected to be more effective in removing iodine because condensation in the pool will be very rapid thereby minimizing the contact time between steam and water. Once the steam is condensed iodine must travel vertically through $8\frac{1}{2}$ to 19 feet of water before it can escape from the pool. In addition, separation of iodine in the condenser occurs under a vacuum (1-2 psia) which will favor the transport to the gas phase. The separation factors or DFs for suppression pool scrubbing thus are expected to be larger than those for condensation processes in the condenser. This indeed is true as will be seen in the next section.

6.3.3 Torus Scrubbing of Iodine (6.16)

Radiological measurements were made in a Mark I suppression pool during a 20 minute High Pressure Coolant Injection System (HPCI)

surveillance test while the plant was at 71% of rated power. During the HPCI test, steam from the reactor drives a turbine and is then discharged to the suppression pool as shown in Figure 6.4. Measurements of torus activities in the water and in the gas phase were made both before and after the surveillance test; torus water phase and gas phase iodine concentration are summarized in Table 6.7. Before the test, the primary iodine species in the torus air was organic iodide (97.8% with only 2.2% as HI). Iodine activities in the water before the test were found to contain 85% as I^- and 15% as IO_3^- . Elemental iodine and organic fractions were not detectable in the torus water. Following the test the gas phase contained 30-45% HI . Partition factors defined as the ratio of iodine concentration in the gas to that in the water are also presented in Table 6.7.

Decontamination factors for pool scrubbing can be calculated from:

$$\text{DF} = \frac{\text{I-131 activity transported to pool}}{\text{change in I-131 activity in the gas phase}}$$

where the change in gas phase activity is taken as the maximum observed change in organic I-131 activity. This will be conservative, i.e., predict a smaller DF than if HI concentration changes¹ were used.

¹ Relatively high background levels coupled with low iodine activity in the steam resulting from minimal fuel cladding defects produced HI concentration changes which may have been on the same order as the error in the analytical procedure. Thus the use of the maximum change in organic iodide activity will provide a conservative estimate of the DF.

The I-131 activity transported to the pool is calculated from the reactor water I-131 concentration, the measured I-131 carryover, and total mass of steam transported to the pool.

From data in (Ref. 6.16) the amount of I-131 transported to the pool is:

$$(5.6 \mu\text{Ci/kg})(0.004)^1 1.44 \times 10^6 \text{ kg} = 3.2 \times 10^4 \mu\text{Ci I-131}$$

The maximum change in organic I-131 concentration from Table 6.7 is:

$$5.2 \times 10^{-10} - 3.7 \times 10^{-10} = 1.5 \times 10^{-10} \mu\text{Ci/cc}$$

The volume of the Monticello torus air space is 102,565 ft³ from Ref. (6.1.), then the activity in the gas phase is

$$\begin{aligned} (1.5 \times 10^{-10} \mu\text{Ci/cc})(102,565 \text{ ft}^3)(28,314 \frac{\text{cc}}{\text{ft}^3}) \\ = 0.44 \mu\text{Ci I-131} \end{aligned}$$

$$\text{and the DF} = \frac{3.2 \times 10^4 \mu\text{Ci}}{0.44 \mu\text{Ci}} = 7.3 \times 10^4$$

Based on this large DF, the suppression pool is expected to effectively scrub radioiodine (I¹³¹) carried by the steam. Data from small scale pool scrubbing tests (presented in Section 1.0) will provide additional information on the expected magnitude of the decontamination factor for different chemical forms of iodine, i.e., I₂, CH₃I, HI and NaI.

1 The measured carryover was 0.4% (Ref. 6.16)

6.3.4 Summary of Observations on Iodine Behavior Operating BWRs

The behavior of iodine in an operating BWR is well defined by measurements taken during plant tests. Radiochemical measurements at operating plants indicate that gaseous iodine concentrations (HIO) are significantly attenuated by condensation processes such as those observed in the main condenser and during steam condensation by the suppression pool. In view of these observations and the respective solubilities and volatilities of H_2O and CsI , the removal factor for CsI by the suppression pool during a postulated accident is expected to be much greater than the removal factor for HIO.

Table 6.1

¹³¹I MEASUREMENT RESULTS, TMI - 2 (Ref. 5.16)

LOCATION	¹³¹ I (DECAY CORRECTED TO 3/28) (Ci)	FRACTION OF CORE INVENTORY
(a) R.B. Sump	570,000 gal. at 0.012 μ Ci/ml on 8/28 = 1.4×10^7 Ci	20%
Aux. Building Liquids	Mainly RCBT's A, B & C 0.21×10^7 Ci	3%
Letdown Demineralizer	112 gpm from 0730 to 1900. 3/28 at 1.7×10^4 Ci/g = 0.5×10^7 Ci	7%
R.B. Atmosphere	2.2×10^6 ft ³ at .025 μ Ci/cc on 3/31 = 2×10^3 Ci	0.003%
(b) RCS	80,000 gal at .025 μ Ci/g on 8/28 = 0.4×10^7 Ci	6%
R.B. Surfaces	1.31×10^8 cm ² at 5.8 μ Ci/cm ² on 6/1 = 2×10^5 Ci 1.3×10^8 cm ² at 0.0041 μ Ci/cm ² on 8/30 = 3×10^5 Ci	0.4%
Auxiliary and Fuel Handling Buildings	Charcoal Filters - 124 Ci Building Surfaces - 130 Ci Environmental Release (3/28-4/30) - 38 Ci	$\sim 0.0004\%$
(c) OTSG B Liquid	23,000 gals - 440 Ci	$\sim 0.0006\%$
	TOTAL 25×10^6 Ci	37%

(a) R.B. = Reactor Building

(b) R.C.S. = Reactor Coolant System

(c) OTSG = Once Through Steam Generator

TABLE 6.2

CHEMICAL FORMS OF RADIOIODINE IN BWR PRIMARY COOLANT (%)

REACTOR (DATE)	I_2	I^{-*}	IO_3^{-}	ORGANIC	$CH_3I^{\dagger\dagger}$
DURING NORMAL OPERATION					
BWR No. 13 (7/1972)	<1	80	20	<0.1	-†
BWR No. 2 (11/1972)	8	79	10	4	0.03
BWR No. 9 (12/1972)	1	65	34	<0.1	<0.02
BWR No. 9 (1/1973)	<0.1	52	48	<0.1	-
BWR No. 9 (3/1974)	3	79	18	<0.1	-
BWR No. 16 (7/1974)	<0.1	77	23	<0.1	-
BWR No. 16 (9/1975)	4	64	12	<0.1	-
BWR No. 34 (5/1977)	<0.1	67	33	<0.1	-
DURING SHUTDOWN:					
BWR No. 9 (4/1973)	<0.1	2	98	<0.1	-
BWR No. 9 (3/1974)	<0.1	4	96	<0.1	-
BWR No. 16 (9/1975)	3	67	30	<0.1	-
BWR No. 16 (2/1976)**	-	18	82	-	-

* May include HI_0

** During hot standby

† Not measured

†† Determined by distillation, the contribution from the isotopic exchange between I^- and carrier CH_3I was estimated to be ~0.02% of the total iodine activity.

(Ref. 6.9)

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TABLE 6.3

CHEMICAL FORMS OF RADIOIODINE IN THE CONDENSATE (%)

REACTOR (DATE)	I_2	I^{-*}	IO_3^{-}	ORGANIC	$CH_3I^{\dagger\dagger}$
BWR No. 13 (7/1972)	6	82	10	2	-†
BWR No. 2 (11/1972)	2	85	9	4	-
BWR No. 9 (12/1972)	8	88	3	1	0.06
BWR No. 15 (7/1974)	3	93	4	1	-

* May include $HI0$

† Not measured

†† Determined by distillation, the contribution from the isotopic exchange between I^{-} and carrier CH_3I was estimated to be ~0.02% of the total iodine activity.

(Ref. 6.9)

TABLE 6.4
RADIOIODINE SPECIES IN OFF-GAS AT THE SJAE* (%)

REACTOR (DATE)	I ₂	"HIO"	ORGANIC
BWR No. 10	1.5	16.9	81.6
BWR No. 13	30.2	34.6	35.0
BWR No. 16	52.3	18.3	29.4
BWR No. 11	4.5	20.2	75.3
BWR No. 12	6.7	22.6	70.7
BWR No. 13 (7/1972)	30	-†	70 †
BWR No. 2 (11/1972)	2	-†	98 †
BWR No. 9 (12/1972)	12	-†	87 †
BWR No. 9 (4/1973)	12	6	82
BWR No. 34 (5/1977)	10	48	42

* SJAE = Steam Jet Air Ejector

† HIO not determined; the organic fraction may include HIO.

(Ref. 6.9)

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TABLE 6.5

COMPARISON OF IODINE ACTIVITY CONCENTRATIONS IN BWR NO. 13 REACTOR WATER AND OFF-GAS*

ISOTOPE	REACTOR WATER		OFF-GAS	
	$\mu\text{Ci/l}^{**}$	CONCENTRATION RELATIVE TO I-131	$\mu\text{Ci/l}^{**}$	CONCENTRATION RELATIVE TO I-131
I-131 (8.04 D)	3.35	1	0.0068	1
I-133 (20.8 H)	9.55	2.8	0.0139	2
I-135 (6.59 H)	20.4	6.1	0.0251	3.7
I-132 (2.29 H)	25.7	7.5	0.0233	3.4
I-134 (52.6 M)	53.8	16	0.0499	7.3

* Date of analysis: 7-31-72

** Volume measured at ambient temperature

(Ref. 6.9)

TABLE 6.6

IODINE-131 SEPARATION FACTOR IN BWR HOT-WELL
(Reference 6.14)

Reactor (date)	Gas Phase Radioiodine Chemical Composition, % ^a			Separation Factor ^b	
	I ₂	HI0	Organic	Inorganic	Total
BWR No. 9 (12/1972)	--- ^d	---	---	---	1296
BWR No. 10 (1973) ^e	1.5	16.9	81.6	690	111
BWR No. 11 (1973) ^e	4.5	20.2	75.3	2418	498
BWR No. 12 (1973) ^e	6.7	22.6	70.7	1940	407
BWR No. 13 (1973) ³	30.2	34.6	35.0	141	78
BWR No. 13 (7/1972)	---	---	---	---	---
BWR No. 16 (1973) ^e	52.3	18.3	29.4	5584	2462
BWR No. 34 (5/1977)	10.0	48.0	42.0	477	243

a. Measured at steam jet air ejector sample point.

b. See text for definition.

c. Including I₂ and HI0.

d. Chemical form not determined.

e. Data obtained from Ref (6.14).

TABLE 6.7

Torus Airborne Iodine Activities and Their Gas/Water Partition

Sample Location and Time	Water phase, $\mu\text{Ci/Kg}$		Gas Phase, $\mu\text{Ci/ml}^*$				Concentration in gas Particles, Concentration in water			
	I-131	I-133	I-131		I-133		I-131		I-133	
			Organic	H ₂ O	Organic	H ₂ O	Total	Inorganic	Total	Inorganic
Torus Sample ** Station,										
2/27, 9:30 - 2/28, 9:50	$2.5 \times 10^{-3} \dagger$	-	3.68×10^{-10} (97.8%)	8.16×10^{-12} (2.2%)	4.42×10^{-12} (~100%)	-	1.5×10^{-4}	3.3×10^{-6}	-	-
C-2 3/3, 1825-1955	1.8×10^{-3}	-	1.6×10^{-10} (66.1%)	8.71×10^{-11} (33.9%)	-	-	1.34×10^{-4}	4.56×10^{-5}	-	-
C-2 3/3, 2026-2058	$2.46 \times 10^{-3} \dagger$	-	2.76×10^{-10} (55.%)	2.26×10^{-10} (45%)	-	-	2.04×10^{-4}	9.2×10^{-5}	-	-
C-2 3/3, 2155-0015	$1.87 \times 10^{-3} \dagger \ddagger$	5.84×10^{-3}	2.5×10^{-10} (67.5%)	1.14×10^{-10} (32.5%)	3.45×10^{-11} (52%)	3.19×10^{-11} (48%)	1.9×10^{-4}	6.1×10^{-5}	1.3×10^{-5}	5.5×10^{-6}
C-2 3/4, 1124-1421	1.87×10^{-3}	5.12×10^{-3}	1.18×10^{-10} (69.9%)	4.82×10^{-11} (30.1%)	8.51×10^{-11} (69.8%)	3.69×10^{-11} (30.2%)	8.9×10^{-5}	2.65×10^{-5}	2.4×10^{-5}	7.2×10^{-6}
C-1 3/3, 2026-2058	-	-	3.74×10^{-10} (total)	-	-	-	-	-	-	-
C-3 3/3, 2026-2058	$2.36 \times 10^{-3} \dagger \ddagger$	-	4.83×10^{-10} (total)	-	-	-	2.03×10^{-4}	-	-	-
C-4 3/3, 2026-2058	$2.36 \times 10^{-3} \dagger \ddagger$	-	5.2×10^{-10} (total)	-	-	-	2.2×10^{-4}	-	-	-
C-5 3/3, 2026-2058	$2.82 \times 10^{-3} \dagger \ddagger$	-	2.4×10^{-10} (total)	-	-	-	8.5×10^{-5}	-	-	-

*No iodine activities in I₂ or particulate forms were found.

†The I-131 activity in water was found to be ~85% as I⁻, and ~15% as IO₃⁻, and both I₂ and organic fractions were not detectable.

‡Avg. of three samples taken during the sampling period.

**HPCI Test Occurred on 3/3.

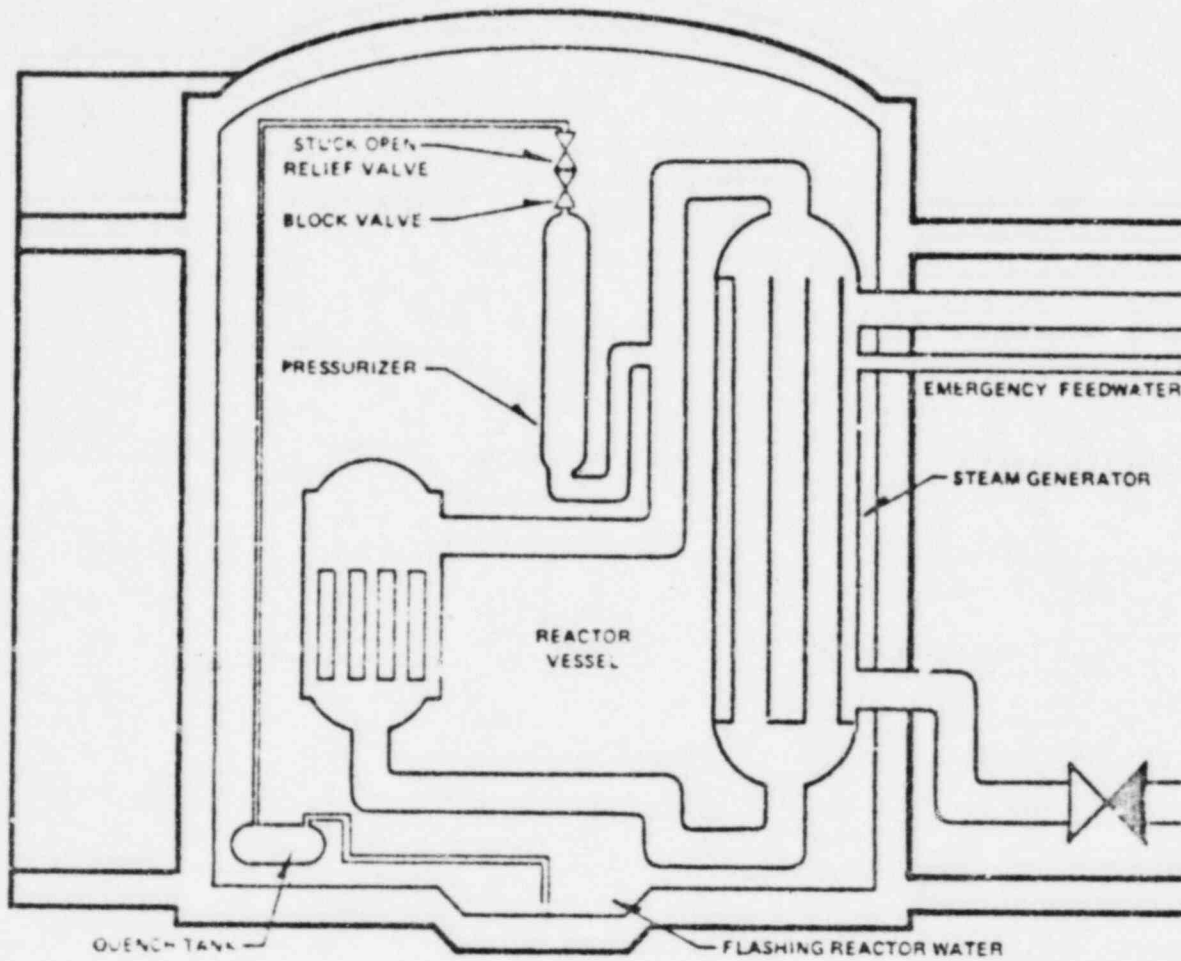


Figure 6.1 Fission Product Transport Pathway at TMI-2

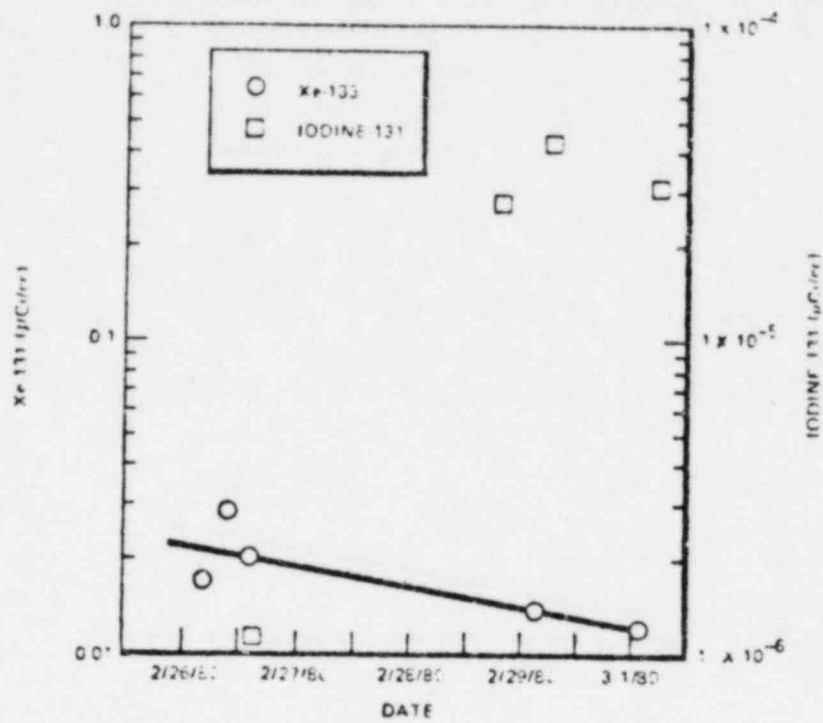


Figure 6.2. Xe-133 and Iodine-131 Levels in Containment Atmosphere at Crystal River

(Ref. 6.3)

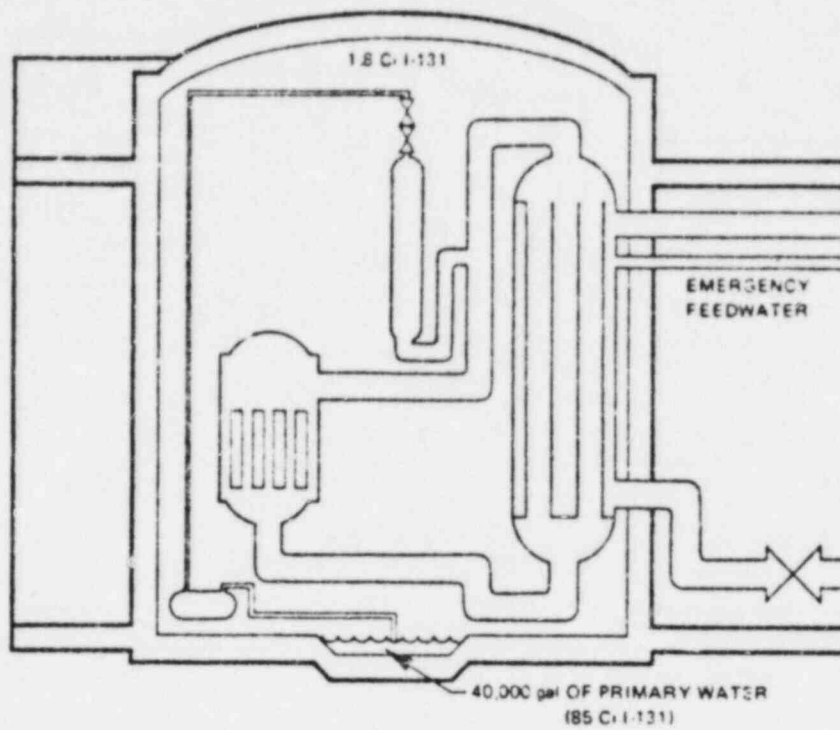


Figure 6.3. I-131 Distribution at Crystal River Unit 3 on 3/2/80

(Ref. 6.4)

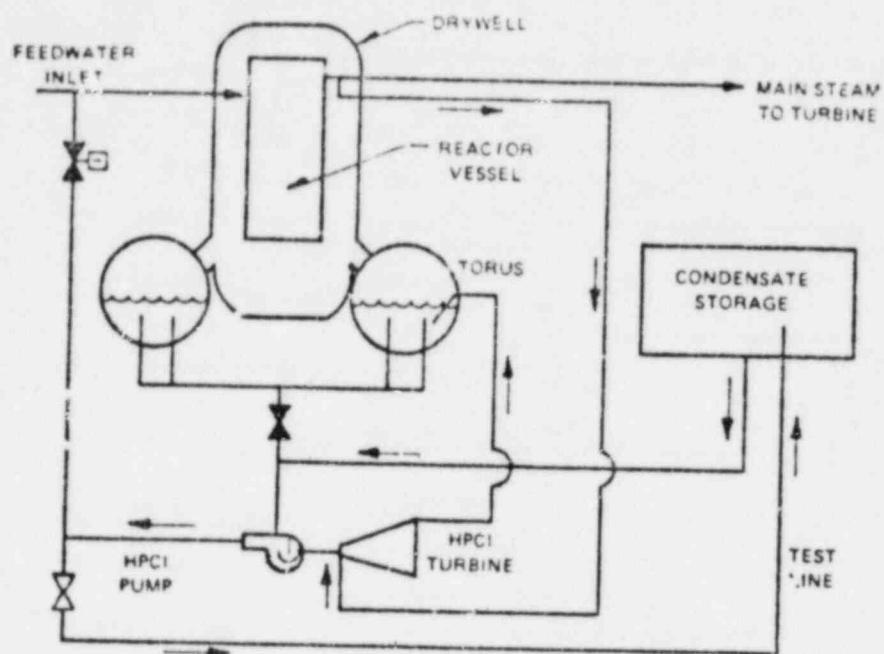


Figure 6.4. HPCI System Simplified P&ID - Test Configuration

(Ref. 6.16)

7.0 Iodine and Particulate Scrubbing By Pools of Water

An extensive review of the literature has been made to identify and document observed iodine attenuation factor, from pool scrubbing experiments. This search identified several papers that are applicable to BWR accident mechanisms.

The majority of the tests used iodine in the elemental form I_2 . However, several investigators used HI, CH_3I , and small diameter insoluble particles. Attenuation of particles is of importance because fission products released during an accident may be in the form of small particles or in aerosol form. The assumption that an aerosol exists has been supported by Sandia tests (7.1). If a core melt occurs, the subsequent high temperatures will result in vaporization of certain metals (structural components, trace metal in the fuel, etc.). As these materials are transported from the core region, they will become cooler and condense into an aerosol. This is the basis for examining aerosol transport in suppression systems.

Summaries from each of the relevant papers are presented in the following sections. The papers are grouped into two categories: scrubbing by subcooled pools and scrubbing by boiling pools.

7.1 Scrubbing By Subcooled Pools

7.1.1 Fission Product Entrainment in Pressure Suppression Systems (7.2)

In 1959 General Electric conducted an exploratory test to evaluate the effectiveness of a pressure suppression system to act as a barrier to the release of fission products following a reactor accident. The tests were run with noble gases (xenon and krypton), halogen (iodine), a soluble salt (sodium iodide), and insoluble particulate matter (2 μ m florescent zinc sulfide particles).

As shown in Figure 7.1.1, the test facility consisted of three interconnected pressure vessels which simulated the reactor pressure vessel, drywell and the suppression pool. Water in the reactor vessel is heated to saturation conditions at 1000 psig and then discharged through an orifice plate into the drywell by breaking a rupture disc. Part of the water, after it flows through the orifice, flashes to steam which in turn discharges from the drywell through vent pipes into the water pool where the steam is condensed. A more detailed description of the test facility can be found in Reference 7.2.

Fission product tracers were placed inside the reactor vessel which was filled with a charge of water. The water was then electrically heated to saturated conditions, at which time it was discharged to the drywell and suppression chamber. Concentrations in the wetwell air space were then determined by taking grab samples. Test results presented in Table 7.1.1 show that the DFs ranged from 10^5 - 10^7 for iodine and insoluble particulates.

The test results show that the suppression pool effectively retained all but a very small amount of the iodine, NaI, and 2 μ m particles, while a significant portion of the noble gases were released from the pool.

Although the tests were exploratory in nature, the test results showed that a suppression pool is an extremely effective barrier to the release of fission products following a reactor accident.

7.1.2 Iodine Removal By a Scale Model of the S.G.H. W. Reactor Vented Steam Suppression System (7.3)

Reference 7.3 reported results from a series of tests in which molecular iodine and small nickel-chromium particles (0.06 μ m) were released into steam, air, and steam-air mixtures. These mixtures

were then allowed to pass through a pool of water and pool decontamination factors were measured. The experimental apparatus, shown in Figure 7.1.2 was a one-third scale model of a section of the SGHW discharge pipes. It was designed to reproduce primary system failure conditions by properly scaling discharge pipe, pool surface area and steam mass flow. A correctly scaled amount of heat passed to the suppression pool, but gas velocities in the discharge pipe and depth of discharge pipe immersion in the pool were full scale. The mass of I_2 used was equivalent to the release of a few percent of the content from several reactor fuel assemblies.

Tables 7.1.2 and 7.1.3 summarize test results with iodine. Decontamination factors (DF) were determined by taking the ratio of iodine in the suppression pool water to that found from sampled air above the pool. The values obtained varied depending on the air to steam ratio. Decontamination factors for I_2 ranged from 20 to 320. Iodine release time, pool temperature, and depth of discharge pipe immersion were found to have little effect on the DF.

Table 7.1.4 presents results from the $0.06 \mu m$ particle tests. The DFs for particles were found to vary only slightly with air/steam ratios and were highest at the larger ratios. Lower DFs were obtained at the high steam flow rates than at the lower ones (compare tests 21 and 22 with 17 and 18). However, at the high steam flow rates a much greater proportion of the total activity was found on the demister, so the smaller DFs may have been due to spray carried up under more vigorous conditions. The very large DFs found with 75 wt % air are much greater than expected (refer to Table 7.1.3). A possible explanation of these results is that when cold air is mixed with saturated steam the air may become highly supersaturated. Under these conditions condensation might occur on the submicron particles which would grow into droplets much larger and more easily removed than the aerosol itself (7.3). In this study the air was cold and the steam was saturated at the entry of the mixing

chamber. In other tests, when the air was not preheated, significantly greater DFs were obtained (7.3). This mechanism may also explain the large proportion of the activity found on the demister, since it would remove droplets although it would not remove the aerosol itself. Decontamination factors were reported between 15 and 1680.

7.1.3 Iodine Clean-up in a Steam Suppression System (7.4)

Experimental work presented in this Canadian paper was similar in scope to that of Hillary et.al. (7.3). The objective was to assess the effectiveness of a steam suppression pool in removing fission products released during a reactor accident. Elemental iodine (I_2), methyl iodide (CH_3I), and hydrogen iodide (HI) were used in the experiments to simulate the behavior of gaseous forms of iodine likely to be present following a reactor accident. In addition, aerosol particles ($0.06 \mu m$ diameter), simulating the behavior of particulate material to which iodine might be attached, were also tested.

Tests were run using both small and large scale equipment. The test equipment is shown diagrammatically in Figures 7.1.3 and 7.1.4. The small scale tests used 3 mm discharge pipes immersed in 6 cm of water. Air containing either an iodine species or a particulate aerosol was injected into the steam flow and the mixture passed through the discharge pipe and into the water pool. Vapor velocities in the discharge pipe were about 34 m/sec. Air was drawn from above the pool surface, collected and analyzed.

The large scale test equipment consisted of a stainless steel tank 4 1/2 feet deep, 3 feet long and 1 1/2 feet wide containing the water pool with single and multiple discharge pipe arrangements. The discharge pipes were 50 mm in diameter and 50 cm below the pool surface. Air containing either iodine species or particulates mixed with steam was passed through the discharge pipes and air samples above the pool were sampled and analyzed. The pool temperature was $50^\circ C$ ($122^\circ F$) and the pH was 7.

Results, in the form of a decontamination factor, are shown in Figures 7.1.5 and 7.1.6 as a function of discharge pipe diameter, air/steam rates, and final concentration. DFs ranged between 10 and 2000 for elemental iodine. DFs ranged between 10-1000 for HI, between 50-500 for 0.6 mm particles and between 1-5 for CH_3I , and were dependent on pool depth, velocity, and % air/steam mixture.

7.1.4 Diffusion of Iodine in Water. The PIREE Experiment (7.5)

The purpose of the PIREE Experiment sponsored by the French CF' ... to investigate the diffusion of radioactive iodine in pure water entrained in a flow of hot pressurized carbon dioxide. The experimental apparatus shown in Figure 7.1.7 consisted of a 42 m^3 (11,160 gal) stainless steel tank (6 meters x 3m dia) filled with demineralized water. A loop consisting of a heater, heat exchanger, compressor, and CO_2 cylinders was connected to the tank so that contaminated CO_2 could be injected into the water at a pressure of 20 kg/cm^2 (280 psi) and 400°C .

The CO_2 carrier gas was contaminated with iodine-131 (I_{131}) produced by the oxidation of active sodium iodide. Altogether, fourteen experiments were performed with the following variations: duration of the injection varied from 20 to 80 seconds; concentration of iodine in the carbon dioxide at the point of injection ranged from 10^{-8} - 10^{-6} g/g; temperature of the circuit ranged from 100 to 300°C ; total I-131 in the water was varied from 10^{-5} Ci to 15 Ci and flow rates ranged from 2 to 20 g/s. The air space above the tank of water was analyzed and the transfer factor of iodine through the water was calculated as follows:

$$K = \frac{A_a}{A_e + A_a}$$

where A_a is the radioactivity of the iodine which passed through the water, and A_e is the radioactivity of the iodine trapped in the water, measured as soon as possible after injection.

Measured transfer factors, i.e., the retention capability of the water, are shown as Table 7.1.5 with the average error calculated on the basis of the uncertainties in the determination of the activities. Based on the results presented it was concluded that the transfer factor is independent of the parameters investigated and thus variations in activity in the water from 10^{-5} to 15 Ci did not have any noticeable effect on the retention of iodine. The transfer factor, on the other hand, decreased with increasing height of the water column, and with decreasing flow of the carrier gas. For a flow of 20 g/s the transfer factor varied from 1×10^{-2} to 6×10^{-4} as the level varied from 2 to 6 meters, while at a flow rate of 2 g/s under the same conditions, the transfer factor varied from 10^{-3} to 10^{-4} .

The transfer factor is related to the decontamination factor DF by the following relationship:

$$DF = 1/K$$

where the DF is defined as the ratio of total activity in the air above the water pool to the total activity in the water pool. Decontamination factors for each of the 14 experiments were calculated and results are shown in Table 7.1.5.

DFs range from 71 to 10^4 and are dependent on the height of water in the tank, and the flow rate of the carbon dioxide carrier gas. The report concludes that "water has a very high scrubbing power" and that iodine releases from a water pool would be only a small fraction of the total iodine released to the pool.

7.1.5 Elemental Iodine Retention by Pressure-Suppression Pools (7.6)

Experiments designed to provide information about the retention of I_2 by pressure suppression pools were conducted at the Oak Ridge National Laboratory. Test conditions were selected to conform as closely as possible to the design basis accident. Elemental iodine was used as the simulated fission product. The experimental apparatus shown in Figure 7.1.8 consisted of a large aluminum tank which contained approximately 260 gallons of water. Saturated steam at 125 psig was supplied to the tank through simulated downcomer pipes. The downcomer pipes were 0.680 inches ID. Elemental iodine was injected into the steam in quantities sufficient to produce concentrations of 0.5-10 ppm in the pool.

The investigators considered the following cases: (1) saturated steam (containing elemental iodine) injected into water initially containing no iodine; (2) saturated steam (containing elemental iodine) injected into water having a known initial concentration of iodine; (3) the injection of saturated steam plus 2 wt % air (this mixture containing elemental iodine) into water initially containing no iodine; (4) the injection of saturated steam plus 2 wt % air (the mixture containing elemental iodine) into water having a known initial concentration of iodine; (5) the same as Case 3 except the pool contained 1000 ppm CO_2 and was at a pH of 9; (6) the same as Case 4 but in addition the pool contained a known concentration of iodine; (7) the same as Cases (3) and (4) but with multiple downcomers.

Cases 1-4 involved final iodine concentrations ranging from 0.5 to 10 ppm in the pool water. The other cases involved iodine concentrations of 5 to 10 ppm in the pool water.

Approximately 200 lbs of steam were discharged to the pool in each test. Steam mass flow rates were from 79,000 to 238,000 lb hr^{-1} ft^{-2} . In the first few experiments, a relatively large number of samples were taken at various elevations in the tank to determine the extent of possible concentration gradients. The data indicated, however, that gradients were negligible. In the subsequent experiments fewer samples were taken and these were primarily at the horizontal midplane of the tank.

Table 7.1.6 lists the quantity of iodine removed normalized to micro-grams of iodine per pound of injected steam for each of the various test conditions. The data are grouped according to the quantity of iodine injected into the system and subgrouped according to the steam flow rates and steam-air mixture ratios. These data were used to calculate decontamination factors for each test by determining the ratio of total iodine injected to the total iodine removed. DFs ranged from 57 to 1445. Under all conditions tested the quantity of iodine released was a small fraction of the total, the largest value (smallest DF = 57) was ~1.8%.

7.1.6 Scale Model Tests of Methyl Iodide Removal in Suppression Pools (7.7)

In a General Electric Company test, a 1/10,000 scale model of a BWR pressure suppression system (Mark I design) was constructed to study the absorption of methyl iodide in suppression pools under loss-of-coolant accident conditions. The major components of the scale model containment (pressure vessel, drywell and wetwell) and associated piping are shown in Figure 7.1.9. Table 7.1.7 lists data relative to the design and actual size of the facility.

Results are presented in Table 7.1.8 as a function of quantity of methyl iodide injected, (C); pH of the pool; depth of submergence of downcomer, (S); time of methyl iodide release, (R); and pool temperature. The effectiveness of the pool in removing methyl iodide is presented as percentage absorption in the pool. Absorption (A), is related to the decontamination factor DF by the relationship:

$$A = 100\% - \frac{100\%}{DF}$$

The absorption varied from 11% to 69% resulting in DFs ranging from 1.1 - 3.0. The results showed that absorption of methyl iodide in the model suppression pool was not affected by increasing the containment inventory of CH_3I from 0.2 to 2 mg. No change in absorption was observed due to changes in the radial position of the downcomer, or changes in the pH of the pool from 7 to 10. Pool absorption for a downcomer submersion depth of 1 foot was about 20% lower than for a 4 foot submersion; the absorption for an initial pool temperature of 150°F was about 28% lower than that for a pool temperature of 90°F. An increase in the delay time between the start of the blowdown and the addition of CH_3I to the drywell initially increased the pool absorption. For delays greater than 2 seconds, the pool absorption decreased with increasing delay until the absorption for a 6 second delay was about 24% less than for a zero delay.

7.1.7 The Marviken Full Scale Containment Experiments (7.8)

A series of 16 blowdown tests was performed in the full scale containment of the Marviken power station. The behavior of iodine in the containment during the blowdown and post blowdown period was examined so that the transport behavior of iodine in a multi-compartment, pressure-suppression type containment could be more fully understood. The main objectives of the tests were to investigate: (1) the removal of methyl iodide (CH_3I) by natural

processes from the containment atmosphere during a limited time; (2) the removal of elemental iodine in Kg quantities, (3) the effects of spray cooling on the removal of iodine from the containment atmosphere; (4) the trapping of iodine in the wetwell pool; and (5) the leakage of iodine from the containment.

The test facility consisted of a pressure vessel and the containment system of the shutdown Marviken nuclear plant. A cross sectional view of the containment is shown in Figure 7.1.10. The containment is divided into two compartments, the drywell which surrounds the pressure vessel, and the wetwell which contains a condensation water pool connected to the drywell by a vent system. The drywell, consisting of several compartments (shown as encircled numbers) in Figure 7.1.11 has a total air volume of 1934 m^3 including the vent system to the normal water level inside the vent pipes. The wetwell lies below the drywell; the normal depth of the wetwell water pool is 4.5 m, giving a pool volume of 560 m^3 (10^5 gallons) and a wetwell air space of 1584 m^3 . The vent system consists of four channels (with a 1.2 m ID) connected to a header located in the wetwell air space. From this header 58 vent pipes (with a 0.3 m ID) lead vertically downward to the wetwell pool. The normal submergence depth is 2.8 m.

The pressure vessel has an inside diameter of 5.22 m and is 25.6 meters high. The net volume of the vessel is 414 m^3 . The vessel is designed for a pressure of 834 psia and a temperature of 272°C .

The simulation of a pipe rupture was possible at three locations: in the upper drywell (Room 124); and in the lower drywell (Room 122) where simulated breaks of the feedwater system and main steam line system were possible. The equipment for injecting iodine was located in Room 121 just behind a door opening to Room 111 (See Figure 7.1.11). During the injection, iodine was carried by air into Room 111. Gas sampling of iodine occurred at five sample points in the drywell and two in the wetwell atmosphere (sample points are shown as a, b, and c in Figure 7.1.11). Water samples from the pool could be obtained from the center of the pool and at the periphery of the pool.

A series of 16 blowdown tests were conducted. A brief description of each test and initial conditions is summarized in Table 7.1.9. The events and experimental conditions for each test, including the quantity of iodine injected and the time of injection, are summarized in Table 7.1.10. A more detailed description of each test can be found in Reference 7.3.

The transport behavior during the blowdown period was investigated first using methyl iodide. CH_3I was injected prior to the blowdown to investigate the transport, gas removal and trapping in the water pool by natural processes. When CH_3I was injected prior to the blowdown, it was completely removed from the drywell atmosphere during the blowdown phase. In the wetwell, CH_3I was partially present in the pool water and in the wetwell atmosphere. Test times for these experiments were 3-4 hours and these experiments were too short for equilibrium conditions to be reached. Following the blowdown, CH_3I was retransferred into the drywell through the wetwell pressure relief valves. Results from two typical tests (Run 4 and Run 8) are presented in Tables 7.1.11 and 7.1.12. From these data decontamination factors can be calculated for various times. These DFs along with theoretical equilibrium DFs are presented in Table 7.1.15. The experimental DFs vary between 1.2 to 4.9 and are larger than the predicted equilibrium values.

The transport behavior of elemental iodine during the post-blowdown period was investigated in Runs 15 and 16. In Run 15 spray cooling of the lower drywell occurred, while spray cooling in the lower drywell did not occur in Run 16. Two kg of elemental iodine was released during the post-blowdown period. Injection, for example, occurred at 1 and 0.7 hours, however, the blowdown period was over at 0.6 and 0.08 hours respectively for runs 15 and 16 (Refer to Table 7.1.10), therefore very little steam was present to transport the I_2 through the pool. In both runs the concentration levels in the upper drywell and wetwell gas spaces were extremely low and at times the concentrations were below the detection limit. The

concentration levels in the lower drywell were also low and of the same order in both runs. Due to the high rate of deposition of elemental iodine, the advantages of spray cooling were concluded to be only marginal. Results from runs 15 and 16 are presented in Tables 7.1.13, 7.1.14, and 7.1.16. DFs of 225 and 265 were reported for runs 15 and 16 respectively (7.8) after 3 hours.

The results indicate that a DF of at least 200 can be expected for I_2 during the post blowdown period due to natural deposition processes. One would have expected a much larger DF if sufficient steam had been present to transport the I_2 through the pool during the post blowdown period. Unfortunately this case was not investigated.

7.1.8 Simulation of Container Venting Under Sea Water (7.9)

In this Mine Safety Appliances Co. experiment the under water release of radioactive contaminants from a reactor compartment was simulated by injecting contaminated steam and air from an open end 2 inch pipe in 10 feet of sea water. A schematic of the test apparatus is shown in Figure 7.1.12. Two 40 gal. autoclaves were used to bring contaminated coolant to a temperature of 550°F and a pressure of 1065 psig. Piping was arranged so that water from both autoclaves could be discharged simultaneously into an empty 10,000 gallon tank. A 2-inch pipe led from the top of the tank to the sea water container which was constructed from five 55-gallon barrels welded end to end. Radioactive Na_2CO_3 , $RbCl$, I_2 , and Y_2O_3 were added to the reactor solution in concentrations of 2, 7.5, 1, and 10 ppm respectively. The contaminated coolant was discharged to the 10,000 gallon tank. The gases passed through the sea water and were collected and analyzed.

Results from the tests are summarized in Tables 7.1.17 and 7.1.18. The major portion of the contaminants were observed to be retained in the 10,000 gallon tank, thus hindering the evaluation of the sea water column as a decontaminating agent. The area in the 10,000

gallon tank where the unflashed water and condensed water collected was found to contain the highest activity. Decontamination factors for plateout and water removal mechanisms were calculated from the data in Table 7.1.17 and results are summarized in Table 7.1.18. For elemental iodine (I_2) plateout DFs were on the order of 10^4 , and a DF=6 was calculated between the water and the air space above the water. Plateout DFs were on the order of 10^5 for Na^+ and Rb^+ , and were approximately 10 for Y_2O_3 . DFs between the water and the air were 30 and 50 for Y_2O_3 and Rb^+ respectively. Plateout phenomena in the tank occurred before pool scrubbing occurred.

7.1.9 Scrubbing of Iodine in a Column of Water (7.10)

A series of small scale tests was performed by Westinghouse to obtain a quantitative measure of the iodine and carbon dioxide absorption from gas bubbles by a surrounding liquid. The test apparatus was set up to provide release of gas bubbles of controlled diameter at the bottom of a water column. Sampling devices above the column allowed measurement of the iodine or carbon dioxide remaining in the gas bubbles reaching the water column surface. Decontamination factors were computed as the ratio of total trace component injected in the bubbles to that found by the gas space samplers.

The test assembly shown in Figure 7.1.13 consisted of an eight foot glass column having a nominal inside diameter of nine inches. Gaseous iodine (I_2) in helium was released at the bottom of a column of liquid. The iodine vapor carried by helium was injected into the column via a heat traced stainless steel line to prevent iodine deposition in the injection line. The solution in the column was maintained at 120°F. The gas space above the liquid column was purged with helium at a rate of 2 liters/minute to remove the injected helium plus whatever iodine emerged from the column of liquid. The purge flow was initiated before the iodine bubble injection began and continued for ten minutes after the injection had been completed. The results shown in Table 7.1.19

include all the pertinent information on test conditions and iodine inventory. Decontamination factors are calculated from the data in this table as the ratio of iodine injected, to the sum of iodine collected above the test solution. Results for different depths of solution and varying bubble diameters are also presented. DFs are seen to range from 88-1960. For depths of release greater than 7 feet the DFs were measured to be greater than 200.

7.1.10 Scrubbing of Particles by Water (7.11)

Particulate scrubbing by a pool of water was investigated at the Hanford Engineering Development Laboratory. One pound of sodium iodide (NaI) was vaporized at 1700°F and carried by 0.5 CFM N_2 through a foot deep pool of water containing a gravel bed. The resultant scrubber efficiency of the pool was 99% or a DF=100. In other tests a DF=20 was observed for 4μ sodium oxide (Na_2O) particles through 2 feet of water.

7.2 Iodine Scrubbing by Boiling Pools

7.2.1 Trapping of Iodine in Water Pools at 100°C (7.12)

Unlike the earlier references reviewed, this Swedish reference reports data on trapping of I_2 by water pools under saturated conditions, i.e., the pool is boiling. In addition the pool pH was varied between 1 and 12 and pool iodine concentration was varied between (0.0001-1 ppm) to assess the overall effectiveness of the pool to trap I_2 . Two types of experiments were conducted. In the first, a laboratory study evaluated iodine release from boiling water to which elemental iodine was added. In the second, large-scale experiments investigated which conditions were needed for direct breakthrough of iodine without reaction with the pool water. The latter tests are the most relevant to this study.

The large-scale system is shown in Figure 7.2.1. The pool water was preheated by means of a heat exchanger in the pool bottom. An

accident was simulated by inserting a 17 mm diameter tube vertically in the pool. Through this tube, superheated steam at 175-320°C and at flow rates of 0.1-0.4 kg/sec (800-3000 lbs/hr) was injected into the water just above the pool bottom. Elemental iodine, dissolved in water, was injected into the steam, upstream of the pool. Samples were taken from both the pool water and the exhausted steam. Two experiments were performed in the large scale system. In the first experiment iodine was added to the steam for 20 minutes, followed by a period with pure steam injection. The second experiment consisted of simple boiling runs as in the smaller laboratory study, and involved an instantaneous addition of iodine.

The total amount of iodine released in the boiling runs is shown in Figure 7.2.2. These results were consistent with the small scale laboratory study and indicated a strong dependency on the pool pH. Note that higher retention factors were observed under basic conditions.

In the experiment in which iodine at a constant rate of 1.3 mg/sec was injected with steam, the concentration of iodine in the exhausted steam increased with time as shown in Figure 7.2.3. A rapid decrease of iodine concentration in the exhausted steam occurred as soon as the addition of (I_2) was stopped, although the steam continued to be injected into the pool. Differential decontamination factors, defined as the ratio of the concentration of iodine in the injected steam to the concentration of iodine in the exhausted steam, were determined for the first 10 minutes and the direct breakthrough of iodine was "of negligible significance." The increase in iodine in the exhausted steam during the period of addition (refer to Figure 7.2.3) appeared to depend on the decreased degree of hydrolysis as the total concentration in the pool became higher.

Decontamination factors during the period of iodine addition are shown in Figure 7.2.4 as a fraction of iodine concentration in the pool. The DFs ranged from 3 to over 100. Major conclusions from the tests were that no dependence of DF on water height or steam flow rate was observed, however, a rather strong dependence on pool (I_2) concentration and pool pH was observed.

7.2.2 Transfer of Iodine From Aqueous Solutions to Saturated Vapor (7.13)

This Russian test was similar to the previous test in that transport of elemental iodine (I_2) from a saturated liquid was investigated using small scale laboratory equipment. A schematic of the test apparatus is shown in Figure 7.2.5. Steam from a steam generator was fed through a bubbling column. The column was fitted with a thermosteam jacket so that steam condensation on the walls would not occur. Experiments were conducted at steam flow rates of 0.2-1.0 kg/hr. with vapor spaces of greater than 70 cm above the water phase. Very low ($10^{-5}\%$) carryover of moisture drops by steam occurred. Investigations were carried out at three pressures: 27, 57, 142 psia (with boiling points at 118, 143, and 179°C, respectively). The experiments were conducted over a broad range of iodine concentrations (10-100 ppm) and pH values of the solution (5.5-10.5) in the column. Samples of steam were taken for analysis after it was allowed to pass through the iodine solution in the bubble column, and the apparent distribution coefficient, K, defined below was evaluated during these steady state conditions.

$$K = \frac{[HI + I_2] \text{ vapor}}{[HI + HI + I_2] \text{ sol.}}$$

The apparent DF = 1/K

Results for the three different pressure temperature combinations are shown in Figures 7.2.6, 7.2.7, and 7.2.8. The apparent distribution coefficient is seen to vary from 0.01% at pH = 7 to as low as 0.004% for pH=10. The apparent decontamination factor thus varied from 10^4 to 2.5×10^4 .

The abrupt decline in K as the pH of the solution increases is related to the fraction of the unhydrolyzed molecules of elemental iodine in solution. If B is the fraction of hydrolyzed I_2 , then (1-B) is the unhydrolyzed fraction. The dependence of the unhydrolyzed

fraction on the pH is shown in Figure 7.2.9 for I_2 concentrations between 2.5 - 250 ppm. This figure shows that elemental iodine is more hydrated in basic solutions and therefore less would be expected to transfer into the vapor phase.

This relationship between the DF and pH of the pool water is consistent with the hydrolysis of iodine as discussed in Section 5.0, i.e., elemental iodine tends to remain hydrolyzed in a basic solution resulting in a minimal release from the pool.

In BWR core damage accident scenarios the pH of the pool is expected to be approximately 10.5 because of the large quantities of Cs that would be released (as Cs ions, elemental Cs, Cs oxides, or CsI) from the fuel. Under these conditions iodine in the pool (as I^- or as I_2) would be hydrolyzed and would stay in the water even under saturated conditions.

7.3 Summary of Suppression Pool Scrubbing Tests

A summary of the data found in the literature on iodine and particulate scrubbing by a pool of water is presented in Table 7.3.1. Relevant data includes the experimental test conditions, chemical species tested, carrier gas, pool volume, depth of release, orifice size, and transport rate. The magnitude of the attenuation factor (DF) is a strong function of the chemical form of iodine, particle size of particulates, presence of steam, depth of release, and the transport rate.

The applicability of these data to BWR transport mechanisms is addressed in Section 3.0.

TABLE 7.1.1

DECONTAMINATION FACTORS FROM FISSION PRODUCT ENTRAINMENT STUDIES

<u>ELEMENT</u>	<u>INITIAL AMOUNT IN PRESSURE VESSEL</u>	<u>PERCENT RELEASED TO SUPPRESSION CHAMBER AIR SPACE</u>	<u>DECONTAMINATION¹ FACTOR "DF"</u>
Kr	250 cc (std)	25 - 50	4 - 2
Xe	250 cc (std)	< 45	2
I ₂	1 lb.	6×10^{-5} 7×10^{-4} @ 5 hrs.	2×10^6 1×10^5
NaI	110 gm	2×10^{-4}	5×10^5
ZnS ₂ (2 μ m)	110 gm	2×10^{-6}	5×10^7

¹DF is defined as the ratio of mass in wetwell air space to the mass released in the reactor vessel. This is an overall DF and includes the effect of plateout in the drywell.

TABLE 7.1.2

IODINE DECONTAMINATION FACTORS AGAINST STEAM FLOW AND PROPORTION OF AIR

Test No.	Rig steam flow lb/sec.	Rig air flow lb/sec.	Proportion of air wt. %	Iodine release mg	Xe133 recovery % of release	Iodine D.F. = $\frac{I \text{ in pond} + I \text{ in chimney}}{I \text{ in chimney}}$		REMARKS
						Total	Total less large droplets	
1(a)	8.8	0	0	3	-	> 34	> 35	High Xe133 count on charcoal reduced sensitivity Total D.F., 1(a) + 1(b): >28
(b)	-	0.33	100			177	192	
2	8.7	2.4	21.6	8		94	141	
3	7.7	7.3	48.7	7	102	20	28	
4(a)	6.0	0	0	5	-	321	349	Total D.F., 4(a) + 4(b): 99
(b)	-	0.33	100			144	150	
5	6.2	1.5	19.5	5	75	280	336	
6	6.0	7.3	55	5	-	20	52	
7	6.0	7.3	55	5	107	34	40	
8(a)	2.6	0	0	4	-	139	144	Total D.F., 8(a) + 8(b): 85
(b)	-	0.33	100			271	229	
Elution	-	0.7	100		-	146	159	
9	3.1	0.7	19	1	85	70	79	
Elution		0.7	100		-	100	106	
10	3.0	7.3	71	11	-	20	41	

(Ref. 7.3)

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TABLE 7.1.3

IODINE DECONTAMINATION FACTORS AGAINST OTHER VARIABLES

$$\text{Iodine D.F.} = \frac{\text{I in pond} + \text{I in chimney}}{\text{I in chimney}}$$

Test No.	Variable	Rig steam flow, lb/sec.	Rig air flow, lb/sec.	Proportion of air wt. %	Iodine release mg	Xe recovery % of release	Total	Total less large droplets	REMARKS
11	Iodine released over few seconds	3.6	7.3	67	10	95	29	63	Compare Test 10 (Table 7.1.2)
12		7.5	1.75	19	4	-	66	85	Compare Test 2 (Table 7.1.2)
13	Depth of immersion of tubes: 1 ft only	3.3	0.7	18.1	12	81	113	144	Compare Test 9 (Table 7.1.2)
14	Higher initial pond temp. (60°C instead of normal 0-10°C)	5.2	1.7	24.7	6	89	126	249	Compare Test 5 (Table 7.1.2)
Elution			1.0	120			238	284	Elution 3 min. only
15	Steam not present (air only)	0	7.3	100	7	88	14	22	Compare Tests 3, 6, 7 and 10 (Table 7.1.2)

(Ref. 7.3)

TABLE 7.1.5 DECONTAMINATION FACTORS FOR ELEMENTAL IODINE IN A CO₂ CARRIER GAS

H - Level of water in the tank
 Cm - Concentration of iodine in the CO₂ at injection level
 Δti - Duration of the injection
 Δta - Duration of the first exhaust above the water
 Ae - Activity measured in the water
 Aa - Activity measured in the air of the first exhaust (first filter + cocoon film)
 k - Transfer factor for iodine through water, calculated after the first exhaust, from:

$$k = \frac{Aa}{Ae + Aa}$$

$$DF = \frac{1}{k}$$

1) FLOW RATE: 20 g/s

Exp.	H (m)	Cm (μg/s)	Δti (s)	Δta (min)	Ae (μg)	Aa (μg)	k	DF [†]
1	1.7	2.7 10 ⁻⁷	73	60	3.5 10 ⁻⁴	4.8 10 ⁻⁶	1.4 10 ⁻² + 75%	71
2	1.7	2.6 10 ⁻⁸	180	70	7.8 10 ⁻⁵	*4. 10 ⁻⁷	5.1 10 ⁻³ + 30%	196
3	1.7	1.1 10 ⁻⁷	35	60	6.7 10 ⁻⁵	*6.1 10 ⁻⁷	9.0 10 ⁻³ + 30%	111
4	1.7	4.7 10 ⁻²	45	25	11.9	4.8	4.0 10 ⁻³ + 10%	250
5	2.7	3. 10 ⁻⁷	90	10	3.7 10 ⁻⁴	8.5 10 ⁻⁷	2.3 10 ⁻³ + 60%	435
6	2.7	1.5 10 ⁻³	30	100	3.6 10 ⁻⁴	5.7 10 ⁻⁶	1.6 10 ⁻² + 35%	62 ⁵
7	2.7	1.5 10 ⁻²	75	2**	13.6	1.9 10 ⁻²	10 ⁻³ + 10%	1000
8	3.7	4.3 10 ⁻²	105	165	61	1.3 10 ⁻¹	2.2 10 ⁻³ + 10%	455
9	4.7	1.3 10 ⁻¹	20	60	37.5	1.7 10 ⁻²	4.5 10 ⁻⁴ + 30%	2222
10	5.7	1.4 10 ⁻⁵	60	115	1.4 10 ⁻²	6.2 10 ⁻⁵	4.4 10 ⁻³ + 25%	227
11	6.4	8.7 10 ⁻²	30	30	113	7 10 ⁻²	6.2 10 ⁻⁴ + 10%	1613

2) FLOW RATE: 2 g/s

12	1.7	3.7 10 ⁻¹	55	111	19	7.7 10 ⁻²	2. 10 ⁻³ + 10%	500
13	3.7	5. 10 ⁻⁴	100	15	106	6.1 10 ⁻²	5.8 10 ⁻⁴ + 15%	1724
14	5.7	7.5 10 ⁻¹	60	130	90	9.0 10 ⁻³	10 ⁻⁴ + 35%	10 ⁴

* The quantities of iodine deposited on the cocoon film could not be determined because the activities involved were small

** The transfer factor is an underestimate because the duration of the first exhaust from above the water was very short.

† Added for this study.

TABLE 7.1.4

DECONTAMINATION FACTORS FOR THE 0.06 μ m Ni/Cr AEROSOL

Test No.	Rig steam flow lb/sec.	Rig air flow lb/sec.	Proportion of air wt. %	Aerosol D.F. = $\frac{\text{Aerosol in pond} + \text{Aerosol in Chimney}}{\text{Aerosol in chimney}}$	
				Total	Total less droplets
16	2.5	0.097	3.7	123	241
17	2.5	0.375	13.1	120	241
18	2.5	1.8	42	381	> 751
19	2.5	7.5	75	531	> 221
20	2.5	7.5	75	1681	> 841
21	9.0	1.8	16.7	15	49
22	9.0	7.4	45	60	> 181

(Ref. 7.3)

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Table 7.1.6 (Ref. 7.6)

Iodine Removal Data for Various Blowdown Rates, Blowdown Mixtures,
and Initial Iodine Concentrations in the Suppression Pool Water

Run No.	Steam Flow Rate ^a (lb/hr)	Air in Steam (Wt%)	Iodine Injected Into Water (g)	Air Sample Data Iodine Removal $\mu\text{g/lb}$ Steam	DF ^d
16	200	0	0.5	3.3	753
19	200	2	0.5	18.9	132
17	400	0	0.5	3.5	714
20	400	2	0.5	8.9	281
18	600	0	0.5	3.5	714
21	600	2	0.5	8.0	313
4	200	0	5.0	22.8	1096
7	200	2	5.0	126.2	198
3	400	0	5.0	17.3	1445
8	400	2	5.0	75.5	331
6	600	0	5.0	25.5	980
9	600	2	5.0	128.2	195
10	200	0	10.0	217.7	230
13	200	2	10.0	880	57
11	400	0	10.0	160.4	312
14	400	2	10.0	521.0	96
12	600	0	10.0	91.3	548
15	600	2	10.0	499.0	100
22) b	400	2	5.0	22.8	1096
23) b	400	2	10.9	231.6	216
25) c	600	2	5.0	333.6	75
24) c	600	2	10.0	748.0	67

Footnotes:

- a. Normally, each run involved, the injection of 200 lb of steam
- b. Water initially contained 1000 ppm Na_2CrO_4
- c. Double steam downcomers of 0.680 inches I.D. were used
- d. Calculated for this study.

TABLE 7.1.7
Methyl Iodide Test Facility Parameters
Comparison of Measurement with Design Specifications

	1/10,000 Prototype	
	<u>Design</u>	<u>Actual</u>
Pressure Vessel		
Total Volume, ft ³	1.58	1.6
Simulated Break Area, ft ²	0.00035	0.00063
Drywell		
Total Volume, ft ³	15.0	14.6
Wetwell		
Total Volume, ft ³	21.6	19.8
Downcomer Area, ft ²	0.0289	0.0295

(Ref. 7.7)

Table 7.1.8

Absorption of Methyl Iodide in the 1/10,000 Scale Model Suppression Pool

Test	Test Conditions						Pool Absorption, %
	C, mg	pH	L	S, ft	R, sec	T, °F	
1	2.01	7	Center	4	0	90	68.6
2	0.20	7.7	↑	4	2.5	90	62.5
3	0.72	7	↑	4	3.5	120	52.3
4	2.07	7.2	↑	4	5.4	120	51.7
5	0.72	7	↑	4	0	150	45.1
6	0.20	7.1	↑	4	6.4	150	35.2
7	2.07	9.9	↑	4	0	90	77.0
8	0.20	10	↑	4	4.6	90	68.6
9	0.72	9.7	↑	4	0	120	50.5
10	2.07	10	↑	4	6.3	120	29.4
11	0.72	10	↓	4	0	150	47.8
12	0.20	10	Center	4	5.7	150	22.9
13	0.72	7.2	Side	4	6.3	90	51.9
14	0.20	7	↑	4	0	116	59.8
15	2.07	7	↑	4	3.2	150	55.6
16	0.72	10	↑	4	6.3	90	57.0
17	0.20	10	↑	4	0	120	58.4
18	2.07	10	Side	4	3.4	150	54.5
19	0.20	7	Center	1	0	90	49.8
20	2.07	7	↑	1	6	93	44.0
21	0.72	7	↑	1	0	124	46.4
22	2.07	7	↑	1	3.5	120	57.6
23	0.20	7.4	↑	1	3.5	150	48.2
24	0.72	7.4	↑	1	6	150	40.2
25	0.20	10	↑	1	0	90	43.6
26	2.07	10	↑	1	6	90	37.2
27	0.72	10	↑	1	0	124	46.9
28	2.07	10	↑	1	3.5	120	50.7
29	0.20	10	↑	1	3.5	150	40.0
30	0.72	10.1	Center	1	6	150	38.6
31	0.72	8.1	Side	1	3.5	95	53.5
32	0.20	7	↑	1	6	120	37.5
33	2.07	7.8	↑	1	0	140	10.8
34	0.72	10	↑	1	3.5	90	58.3
35	0.20	10	↑	1	6	120	35.5
36	2.07	10.2	Side	1	0	152	22.9
37	2.07	8.1	Center	2.5	3.5	92	53.5
38	0.20	7.6	↑	2.5	6	90	47.6
39	2.07	8.1	↑	2.5	0	120	62.1
40	0.72	8.0	↑	2.5	6	120	26.8
41	0.20	7.9	↑	2.5	0	150	40.0
42	0.72	7.8	↑	2.5	3.5	150	51.6
43	2.11	10.1	↑	2.5	3.5	90	63.5
44	0.20	10	↑	2.5	6	90	23.1
45	2.07	10.2	↑	2.5	0	123	62.4
46	0.72	10.3	↑	2.5	6	120	55.4
47	0.20	10.2	↑	2.5	0	150	43.3
48	0.72	10.4	Center	2.5	3.5	150	55.5
49	0.68	7.5	Side	2.5	0	93	61.8
50	0.22	6.7	↑	2.5	3.5	123	43.8
51	2.11	7.2	↑	2.5	6	146	18.0
52	0.68	10	↑	2.5	0	93	61.2
53	0.22	9.8	↑	2.5	3.5	120	28.7
54	2.11	11.2	Side	2.5	6	150	37.4

TABLE 7.1.9

INITIAL CONDITIONS FOR THE MARVIKEN BLOWDOWN TESTS

BREAK CONDITIONS				VESSEL CONDITIONS			CONTAINMENT CONDITIONS					Remarks	Date of performance
Blow-down No.	Type of break	positions of break		orifice diameter	pressure	mass of water	depth of wet-well pool	volume of wetwell pool	vent pipe submergence	temperature of wetwell pool	vent pipe flow area		
		room	pipe	mm	bar	ton	m	m ³ **	m	°C	m ²		
1	steam	124	top*	125	51	110	4.51	561	2.61,2.71,2.81	19	4.03	Orifice area reduced 40% due to damages of measurement equipment	Aug 13, 1972
2	steam	124	top	250	50	110	4.51	561	2.61,2.71,2.81	22	4.03		Sept 1
3	steam	124	top	200	50	120	4.51	561	2.81	19	4.03		Sept 21
4	steam	124	top	200	49	121	4.60	572	2.90	44	4.03		Oct 20
5	steam	124	top	200	50	119	3.38	422	1.68	21	4.03		Oct 26
6	steam	124	top	200	50	119	4.99	618	3.29	18	4.03		Nov 2
7	water	122	feed water	150	50	117	4.50	560	2.80	17	4.03		Nov 21
8	water	122	feed water	150	50	119	4.46	555	2.76	58	4.03	Orifice area reduced 5% by the rupture disc	Nov 30
9	steam	122	steam	330	50	105	4.51	561	2.81	17	4.03		Dec 19
10	water	122	steam	330	51	293	4.50	560	2.80	17	4.03	Orifice area (330 mm) reduced 10% by the rupture disc for 23 seconds Orifice area (330 mm) reduced 22% by the rupture disc	Jan 23, 1973
	water	122	feed water	150									
11	water	122	steam	330	51	293	4.54	561	2.84	18	2.68		
	water	122	feed water	150									
12	water	122	steam	330	52	297	4.50	553	2.80	19	1.84		
	water	122	feed water	150									

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* The special pipe connected to the top-cupola of the pressure vessel is referred to as the top pipe.

** The volume of the wetwell pool includes the water standing in the open vent pipe, but not in the blocked vent pipes.

(Ref. 7.8)

TABLE 7.1.9 (Cont'd)
INITIAL CONDITIONS FOR THE MARVIKEN BLOWDOWN TESTS

Blow-down No.	Type of break	BREAK CONDITIONS		VESSEL CONDITIONS			CONTAINMENT CONDITIONS					Remarks	Date of performance
		positions of break		orifice diameter	pressure	mass of water	depth of wet-well pool	volume of wetwell pool	vent pipe submergence	temperature of wetwell pool	vent pipe flow area		
		room	pipe	mm	bar	ton	m	m ³ **	m	°C	m ²		
13	water	124	top	90	48	308	4.49	551	2.79	17	1.84	Ø 90 only leakage Ø 330-area reduced 32% Ø 150 only leakage	Mar 8, 1973
	water	122	steam	330									
	water	122	feed water	150									
14	water	122	steam	330	48	296	4.52	556	2.82	46	1.84		Mar 22
	water	122	feed water	150									
15	water	124	top	90	50		4.58	563	2.88	18	1.84	Ø 90 opened and closed Ø 330 never opened	Mar 29
	water	122	steam	330		287							
	water	122	feed water	150									
16	water	124	top	90			3.34	412	1.64	14	1.84	Ø 90 only leakage	Apr 5
	water	122	steam	330	50	321							
	water	122	feedwater	150									

* The special pipe connected to the top-cupola of the pressure vessel is referred to as the top pipe.

** The volume of the wetwell water pool includes the water standing in the open vent pipe, but not in the blocked vent pipes.

(Ref. 7.8)

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TABLE 7.1.10
EXPERIMENTAL TEST CONDITIONS FOR THE MARVIKEN BLOWDOWN TESTS

Events/Run No.	3	4	5	6	7	8	9	10	11	12	13	14	15	16
124 top pipe 122 main steam line 122 feed water line														
Blowdown period (secs)		780	780	780	770	835	765	600	480	240	170	184	181	293
Start of iodine injection		-3000	-3600	a)	900	--	-2760	900	a)	600	a)	600	--	2400
Amount of iodine injection		(g)	2	2.8	1.2	2	3.6	2.1	0.8	0.7	--	0.7	--	2000
Amount of iodine -131 injected		mCi	3.2	4.2	2.6		2	4.6	1.8	7.7	--		9.9	13.8
Gas sampling period (secs)														
1 start							-240	300	600	360	-240	540	3350	2100
1 end							-120	420	733	480	-120	660	3480	2220
2 start							480	1500	3000	3000	360	1980	7080	3500
2 end							600	1620	3120	3120	480	2100	7380	3720
3 start							3600	3660	6000	6000	3000	3000	9660	7200
3 end							3720	3780	6120	6120	3120	3120	9840	7330
4 start							7700	7700	10800	10800		6000	12420	9000
4 end							7320	7320	10920	10920		6170	12600	9180
5 start							10800	10800	13800	13800		10800	16620	10800
5 end							10920	10920	13920	13920		10920	16700	10720

a) Injection time indefinite due to a leakage from the injection vessel during the blowdown period.

b) Spray cooling occurred in both upper and lower drywell in runs No. 3 and 4. In later runs spray cooling was only introduced in lower drywell.

c) Spray cooling of lower drywell was repeated at 11400 sec. and continued throughout the run.

NOTE: All times given in the table are referred to the start of blowdown.

TABLE 7.1.10 (CONTINUED)
EXPERIMENTAL TEST CONDITIONS FOR THE MARViken BLOWDOWN TESTS

Events/Run No.	3	4	5	6	7	8	9	10	11	12	13	14	15	16
Water sampling period (secs)														
start			600	3000		400	1620	6000	10800	300	900		7080	3600
end			840	3120		720	1860	6240	11040	3240	1140		7320	3840
start			9600	10200		3600	3600	10800	13800		3000		9660	7200
end			9840	10320		3840	3840	11040	14040		3740		9500	7500
start						7200	7200	13800			6000		12420	9000
end						7440	7440	14040			6240		12660	9200
start						10800	10800				10800		16020	10800
end						11040	11040				11040		16260	11040
Spray cooling period in drywell (secs)														
start		1715	1698			970								
end		2283	2283	None	None	1550	None	None	None	None	3450	650	1100	610 ^{c)}
Spray cooling period in wetwell (secs)														
start		None	None	None	None	None	None	1900	1200	1200	3450	650	None	None
end		None	None	None	None	None	None	2500	16100	12000	13320	17000	None	None
Start of pool water recirculation (cooling) (secs)														
None		None	None	None	None	2000	None	2500	16100	2040	1140	1500	1729	1020
Opening of drain pipe valve (secs)		--	--	--	--	--	--	900	1000	960	650	286	800	560
End of vessel post leakage (secs)		--	--	--	--	--	--	7700	6300	~3500	7080	5600	2200	1660
Opening of wetwell pressure relief valves (secs)		~890	~880	~880	~890	910	~900	720	1100	Not opened	Not opened	1100	1600	1200

a) Injection time indefinite due to a leakage from the injection vessel during the blowdown period.

b) Spray cooling occurred in both upper and lower drywell in runs No. 3 and 4. In later runs spray cooling was only introduced in lower drywell.

c) Spray cooling of lower drywell was repeated in 11400 and continued throughout the run.

NOTE: All times given in the table are referred to the start of blowdown.

(Ref. 7.8)

TABLE 7.1.10 (CONTINUED)
EXPERIMENTAL TEST CONDITIONS FOR THE MARVIKEN BLOWDOWN TESTS

Event/run No.	3	4	5	6	7	8	9	10	11	12	13	14	15	16
End of computer data collecting (secs)	2338	20.0	4500	5262	2742	15160	9410	6942	17060	16960	17100	7432	18889	675
Mass escape from vessel during blowdown (tons)	38	37	35	36	106	107	41	233	184	23	96	156	270	200
Amount of vessel post leakage (tons)	--	--	--	--	--	--	--	small	21	1-5	77	32	small	small

a) Injection time indefinite due to a leak from the injection vessel during the blowdown period.
b) Spray cooling occurred in both upper and lower drywell in runs No. 3 and 4. In later runs spray cooling was only introduced in lower drywell.
c) Spray cooling of lower drywell was repeated at 314.0 sec. and continued throughout the run.

NOTE: All times given in the table are referred to the start of blowdown.

(Ref. 7.8)

TABLE 7.1.11

Spatial distribution of methyl iodide in the containment at various times relative to the blowdown start. Methyl iodide was injected prior to the blowdown. Run No. 4.

Volume concentration of $\text{CH}_3\text{I}-131$ (pCi/l)

Start of sampling secs	In wetwell		In lower drywell			In room #24	
	a	b	a	b	c	a	b
-360	128	84	2047	1896	1952	2261	2298
540	853	865	78	37	0	0	--
1500	692	611	417	375	303	63	45
2760	532	532	541	472	423	198	179

The total and non-volatile iodine contents of the pool water samples.
Run No. 4.

Start of sampling secs	Sampling point	Sample volume (l)	Off-gas activity (pCi)	Sample activity (pCi/l)	Pool iodine contents (pCi/l)	
					Total	Non-volatile
540	A	5	5570	1500	2714	720
	B	6	5520	1840	2760	1130
2760	A	-	-	1290	-	-
	B	-	-	1400	-	-

A: At the center of the pool.

B: At the periphery of the pool.

(Ref. 7.8)

TABLE 7.1.12

The atmospheric distribution of I-131 in the containment at various times relative to the start of the blowdown. Run No. 8.

Start of sampling secs	Volume concentration of $\text{CH}_3\text{I-131}$ (pCi/l)						
	In wetwell		In lower drywell			In room 124	
	a	b	a	b	c	a	b
-240	67	67	1550	2470	2150	516	470
480	1130	1230	22	37	38	168	119
3600	701	--	446	393	379	275	268
7200	552	605	366	344	312	296	270
10800	485	460	378	390	352	270	269

The total and non-volatile I-131 concentrations in the pool water samples. Run No. 8.

Start of sampling secs	Sampling point	Sample volume (l)	Off-gas activity (pCi)	Sample activity (pCi/l)	Pool iodine contents (pCi/l)	
					Total	Non-volatile
480	B	5.76	3730	1040	17.0	247
3600	A	4.25	2330	1140	1688	528
7200	A	3.93	1410	1140	1610	
10800	B	3.20	1240	1170	1558	

A: At the center of the pool.

B: At the periphery of the pool.

(Ref. 7.8)

TABLE 7.1.13

he spatial distribution of elemental iodine in the containment.
Times are referred to the start of the blowdown. Run No. 15.

Start of sampling secs	Volume concentration of I ₂ (pCi/l)						
	In wetwell		In lower drywell			In room 124	
	a	b	a	b	c	a	b
3360	<6	3	94	93	9	<3	1
7080	2	1	287	144	6	1	1
9660	<4	2	71	49	10	2	1
12420	<4	2	30	20	8	<2	<1
16020	4	2	46	44	14	<2	1

The iodine contents of the pool water samples. Times are referred
to the start of the blowdown. Run No. 15.

Start of sampling secs	Sampling point	Sample volume (L)	Off-gas activity (pCi)	Sample activity (pCi/l)	Pool iodine contents (pCi/l)
7080	A	5.2	<28	4920	4920
9660	B	3.3	<18	3200	3200
12420	B	4.2	<29	4370	4370
16020	A	3.4	24	8550	8557

A: At the center of the pool.

B: At the periphery of the pool.

(Ref 7.8)

TABLE 7.1.14

The spatial distribution of elemental iodine in the containment.
Start of blowdown at 216 secs. Run No. 16.

Start of sampling secs	Volume concentration of I ₂ (pCi/l)						
	In wetwell		In lower drywell			In room 124	
	a	b	a	b	c	a	b
2100	<5	8	38	97	33	5	1
3600	<6	3	425	3810	79	7	2
7200	<5	3	27	74	19	<5	2
9000	<4	2	105	142	26	6	3
10800	<6	3	22	79	23	5	10

The total and non-volatile iodine contents of the pool water samples.
Start of blowdown at 216 secs. Run No. 16.

Start of sampling secs	Sampling point	Sample volume (l)	Off-gas activity (pCi)	Sample activity (pCi/l)	Pool iodine contents (pCi/l)	
					Total	Non-volatile
3600	A	4.73	<28	1930	1930	
7200	B	5.22	<24	7280	7280	
9000	B	5.04	68 ^{a)}	9150	8153	8150
10800	A	3.7	<32	7620	7620	

A: At the center of the pool.

B: At the periphery of the pool.

a) Sample was wetted by water.

(Ref. 7.8)

TABLE 7.1.15

DECONTAMINATION FACTORS (DF) OF METHYL IODIDE BETWEEN GAS AND WATER*
DURING THE BLOWDOWN PERIOD

<u>Blowdown No.</u>	<u>DF Experimental</u>	<u>DF Equilibrium</u>	<u>Water Temperature °C</u>
3	1.2	0.53	73
4	2.1	1	57
5	4.9	1.5	46

*transport through pool

Table 7.1.16
DFs For Elemental Iodine During Post Blowdown Period**

<u>Blowdown No.</u>	<u>DF Experimental</u>
15	225
16	265

**DFs due to natural removal processes (not by pool)

TABLE 7.1.17
FISSION PRODUCT REMOVAL BY SEAWATER

Run	Isotope	Initial Charge (mc)	Initial Activity in Water Accountable - %			Activity Distribution Outside Of 10,000 Gal. Tank				Percent Removal By Sea Water Calculated From	
			10,000 Gal. Tank	Autoclave	Total	Activity In Escaping Air		Sea Wtr (B) (μC) ²	Escape Bag (C) (μC) ²	(A-C) n	B (B+C)
						Orig. (μC)	Adj. (%) (μC)				
1	Na-24	50	97.5	-*	97.5	0.172	0.215	nd ³	nd	-*	-
	I-131	6	97.4	-	97.4	0.015	0.105	nd	nd	-	-
	Y-90	36	86.9	-	86.9	2.4	4.5	nd	nd	-	-
2	Rb-86	50	97.0	1.7	98.7	0.317	0.285	0.251	<0.005	>98	>93
	I-131	20	91.2	1.0	92.2	0.748	0.691	0.600	0.094	86	86
	Y-90	30	71.0	10.4	81.4	3.300	2.970	3.000	0.110	97	96
					Totals	4.385	3.946	3.851	0.204	95	95

1. Normalized for air volume and isotope charge. Run 2 used as the basis for normalization.
2. Activity corrected to time out of reactor. Actual activity of Na-24, I-131 and Y-90 measured after approximately 4, 1 and 3 half-lives decay, respectively in Run 1. Actual activity of Rb-86, I-131 and Y-90 measured after approximately 0.2, 1 and 1 half-life respectively in Run 2.
3. Not detected. Minimum detectable limits for the samples analyzed are: $1 \times 10^{-1} \mu\text{C}$ for Na-24; 1×10^{-3} for I-131; 1×10^{-3} for Y-90 and $5 \times 10^{-3} \mu\text{C}$ for Rb-86. The sea water could not be concentrated because of the high salt content, hence the large minimum detectable limit values.
4. Not analyzed.
5. Not applicable. Activity escaping through sea water column below minimum detectable limit.

(Ref. 7.9)

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Table 7.1.18

Decontamination Factors for
Container Venting Under Sea Water

<u>Run</u>	<u>Species</u>	<u>Plateout DF^(b)</u>	<u>Water Scrubbing DF</u>
1	Na	2×10^5	-(a)
	I ₂	6×10^4	-
	Y ₂ O ₃	8×10^3	-
2	RbCl	2×10^5	50
	I ₂	3×10^4	6
	Y ₂ O ₃	1×10^4	27

Overall DFs^(b) = Plateout DF x Water Scrubbing DF

RbCl	1×10^7
I ₂	1.8×10^5
Y ₂ O ₃	2.7×10^5

(a) concentrations below detectable limit

(b) in these tests the plateout mechanisms occurred before the water scrubbing process

TABLE 7.1.19

RESULTS OF SMALL SCALE TESTS WITH IODINE IN A HELIUM CARRIER GAS (a)

Run No.	Gas Injection Time	Injection Pressure (psig)	Injection Vessel Volume (cc)	Solution Iodine Inventory (mg) Initial/Final	Iodine Collected above Solution in Test Column (mg)		Total Iodine Injection (mg)	Solution Depth (ft)	Orifice Diameter (in.)	Bubble Diameter (cm)	Rise Time (sec)	t/d (sec/cm)	Iodine DF
1	4.75	169	540	0	59.25	0.037	0.007	0.269	0.33	3/8	0.854	2.92	178
2	2.25	171	540	0	14.64	0.045	0.012	0.11	0.17	1	2.12	2.38	88
3	1.75	163	540	0	30.25	0.073	0.007	0.17	0.21	1	2.12	2.38	145
4	3.00	171	540	1.27	12.69	0.006	0.005	0.003	0.014	3/8	0.854	8.43	820
5	3.50	170	540	1.27	34.96	0.097	0.004	0.011	0.022	3/8	0.854	8.43	1530
6	4.00	170	590	0	31.26	0.022	0.025	0.024	0.069	3/8	0.854	8.43	1180
7	6.00	173	590	0	81.20	0.020	0.020	0.001	0.041	3/8	0.854	8.43	1990
8	2.00	171	540	1.27	39.37	0.012	0.004	0.013	0.029	1/2	1.00	7.90	1310
9	2.00	170	540	0	46.95	0.001	0.007	0.020	0.028	1/2	1.00	7.90	1310
10	2.30	172	540	1.27	34.90	0.018	0.004	0.01	0.034	1/2	1.00	7.90	930
11	1.75	170	540	1.27	27.92	0.019	0.005	0.007	0.031	1/2	1.00	7.90	360
12	3.20	170	540	0	39.34	0.017	0.005	0.011	0.033	1/2	1.00	7.90	1190
13	0.75	170	590	0	13.96	0.047	0.018	0.001	0.066	1	2.12	6.85	212
14	0.84	171	590	19.70	32.99	0.031	0.032	0.001	0.064	1	2.12	6.85	206
15	1.15	170	590	0	17.77	0.048	0.033	0.001	0.082	1	2.12	6.85	218

(a) All test with iodine vapor is helium carrier gas bubbled through boric and acid solution (pH = 4.3 - 5.0) at 120°F.

Ref. 7.10

Table 7.3.1

SUMMARY OF POOL SCRUBBING TESTS

TEST REF	TEST DESCRIPTION EXPERIMENTAL CONDITIONS	CHEMICAL FORM OF SIMULATED FISSION PRODUCTS (PARTICLE SIZE)	DF	TEST CONDITIONS POOL VOLUME DEPTH OF RELEASE pH TEMP °F	ORIFICE SIZE	CARRIER TO POOL	TRANSPORT RATE
7.2	Simulated LOCA Blowdown to Dry- well & Pool Steam & Sat Water @ 100 psig/500°F	Xe, Kr I ₂ (a) NaI ZnS ₂ (2 μm)	2-4 ^(b) 10 ⁵ -10 ⁶ 5x10 ⁵ 5x10 ⁷	Pool Vol 1000 gal Depth 18" pH 7 Temp -	1"	Flashing Water Air-Steam Mixture	Rapid Blowdown
7.9	Blowdown to a 1300 Ft ³ empty drum connected to a tank of water	I ₂ (a) Y ₂ O ₃ RbCl	2x10 ⁵ 3x10 ⁵ 10 ⁷	Pool Vol 300 gal Depth 10-20' pH - Temp -	2"	Flashing Water Air-Steam Mixture	Rapid Blowdown
7.7	Simulated LOCA 1/10,000 scale model of BWR MK I	CH ₃ I	1.1-3.2	Pool Vol 150 gal Depth 1-4' pH 7-10 Temp 90-150	2"	Flashing Water Air-Steam Mixture	Rapid Blowdown
7.3	Steam-Air mixtures carried simulated fission products through a water tank	I ₂ Ni-Cr (0.06 μm)	14-320 15-1680	Pool Vol 2000 gal Depth 2' pH 6.8 Temp 10-60°C	-	Steam-Air Mixtures Air Wt% 0-100	Steady 25-100 sec Steam 3-9 lb/sec Air 0.1-7. lb/sec
7.8	Simulated LOCA blow- down to drywell and pool	CH ₃ I	1.2-4.9	Pool Vol 10 ⁵ gal Depth 9' pH - Temp. 20-40°C	12"	Flashing Water Air-Steam	Rapid Blowdown
7.13	Steam passed through a pool of sat. water containing I ₂	I ₂ 2.5-250 ppm	10-250	Pool Vol - Depth - pH 5.5-10 Temp. 120-180°C	-	Steam	Steady 0.2-1.0 Kg/hr.

(a) Added to Reactor vessel water

(b) Overall DF includes plateout and pool scrubbing

Table 7.3.1 (Cont.)

SUMMARY OF POOL SCRUBBING TESTS

TEST REF	TEST DESCRIPTION EXPERIMENTAL CONDITIONS	CHEMICAL FORM OF SIMULATED FISSION PRODUCTS	DF	TEST CONDITIONS POOL VOLUME DEPTH OF RELEASE pH TEMP °F	ORIFICE SIZE	CARRIER TO POOL	TRANSPORT RATE
7.4	Steam carrying simulated fission products through a water tank	I ₂ (a1) I ₂ (a2) HI (b) CH ₃ I (c) (d) Ni-Cr (0.06 μm)	10-500 10-500 10-1000 1.5-5 50-100	Pool Vol. 3 gal 150 gal Depth of 2" release 20" pH - Temp 50°C	0.12" 2.0"	Steam-Air Mixtures	0.1 lb/sec system 0.07 lb/sec air 4 lb/sec ft ² steam 3 lb/sec ft ² air
7.10	I ₂ carrying I ₂ through a water pool	I ₂ (20 mg/liter)	88-1500	Pool Vol. 30 gal Depth - 3-8; pH - 4-5 Temp 120°F	3/8-1"	He	Slow 0.05 lb/sec-ft ²
7.6	Saturated Steam at 125 psig carried I ₂ through a pool of water	I ₂ (0.5 - 10 ppm)	70-1100	Pool Vol. 260 gal Depth 4' pH - Temp -	0.68"	Steam-Air Mixtures Air Wt% 0-2	Steady 22-66 lb/sec-ft ²
7.5	CO ₂ carrying I ₂ through a pool of water CO ₂ @ 280 psi/400°C	I ₂ (15 cf max)	70-10 ⁴	Pool Vol. 10,000 gal. Depth 6-20' pH - Temp -	-	CO ₂	0.02-0.04 lb/sec CO ₂
		(a1) 0.6-40 ppm	air/steam Wt% 30-1				
		(a2) 0.01-0.4 ppm I ₂	air/steam Wt% 20-1	(c) (air/steam) Wt%	20-1		
		(b) Air/steam Wt % 30-1		(d) (air/steam) Wt%	50-10		

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TABLE 7.3.1* (Cont.)

SUMMARY OF POOL SCRUBBING TESTS

TEST REF.	TEST DESCRIPTION EXPERIMENTAL CONDITIONS	CHEMICAL FORM OF SIMULATED FISSION PRODUCTS	DF	TEST CONDITIONS POOL VOLUME DEPTH OF RELEASE pH TEMP OF	ORIFICE SIZE	CARRIER TO POOL	TRANSPORT RATE
7.11	Na ₂ O in H ₂ carried through water	Na ₂ O particles 1 μ m	20	Pool Vol - Depth 2' pH - Temp -	-	H ₂	0.5 ft ³ N ₂ /min
7.12	Steam carrying iodine through a saturated pool steam @ (175 psig/ 320°C)	I ₂ (0.1-2 ppm)	2-200	Pool Vol 530 gal Depth 3-12' pH 6-12 Temp 212°F	0.57"	Steam	0.1-0.4 Kg/sec 90-360 lb/sec ft ² (steady-high flow)
BWR OPERATING EXPERIENCE							
6.7	Separation of iodine in BWR condenser	1", H ₂ O	1000	Pool Vol - N/A Depth - N/A pH - 7 TEMP 100°F Pressure 1-2 psia	N/A	Steam Condensing	High Flow-Steady >>100 lb/sec.
6.9	BWR HPCIS TEST Toros Scrubbing Test Data Steam @ 1000 psig 550°F	1", H ₂ O	7x10 ⁴	Pool Vol - 5x10 ⁵ gal Depth 4' Temp -	0.5"	Steam	High Flow-Steady 1300 lb/sec

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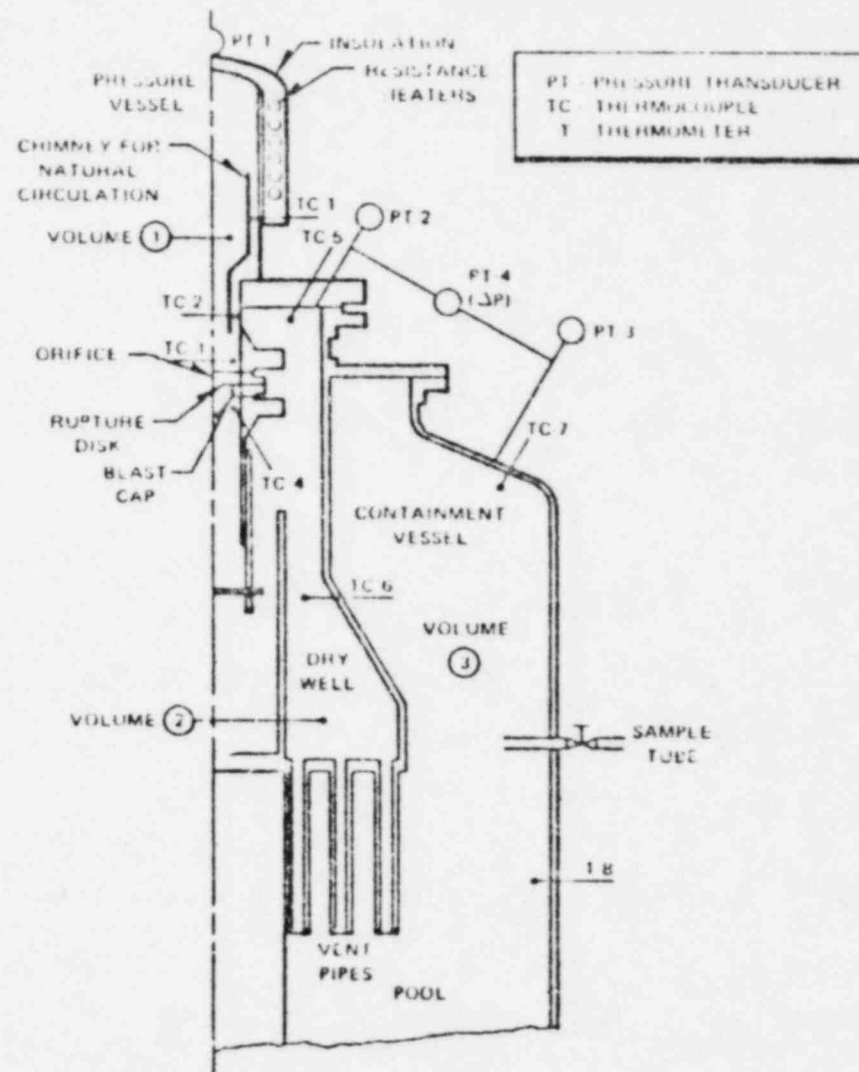
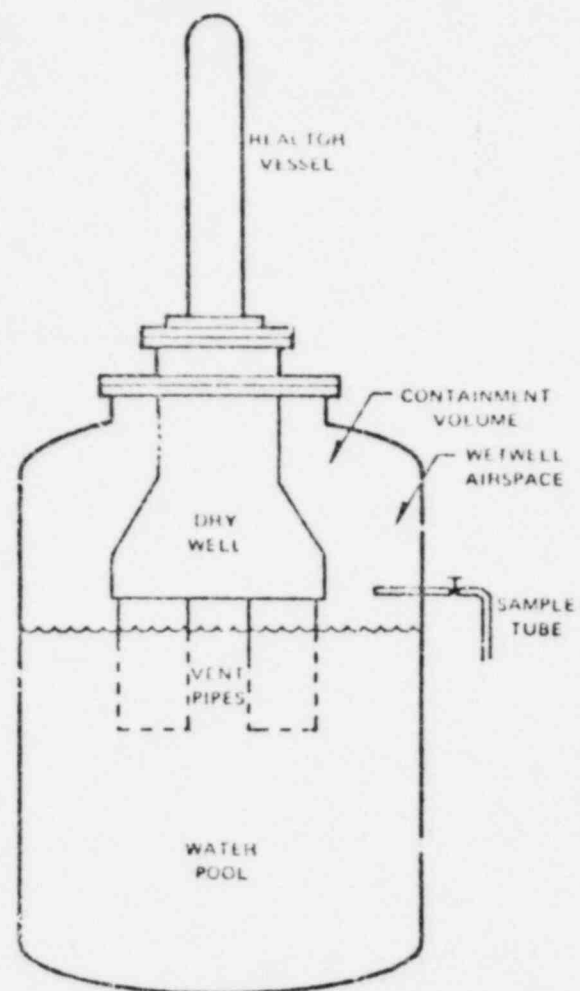
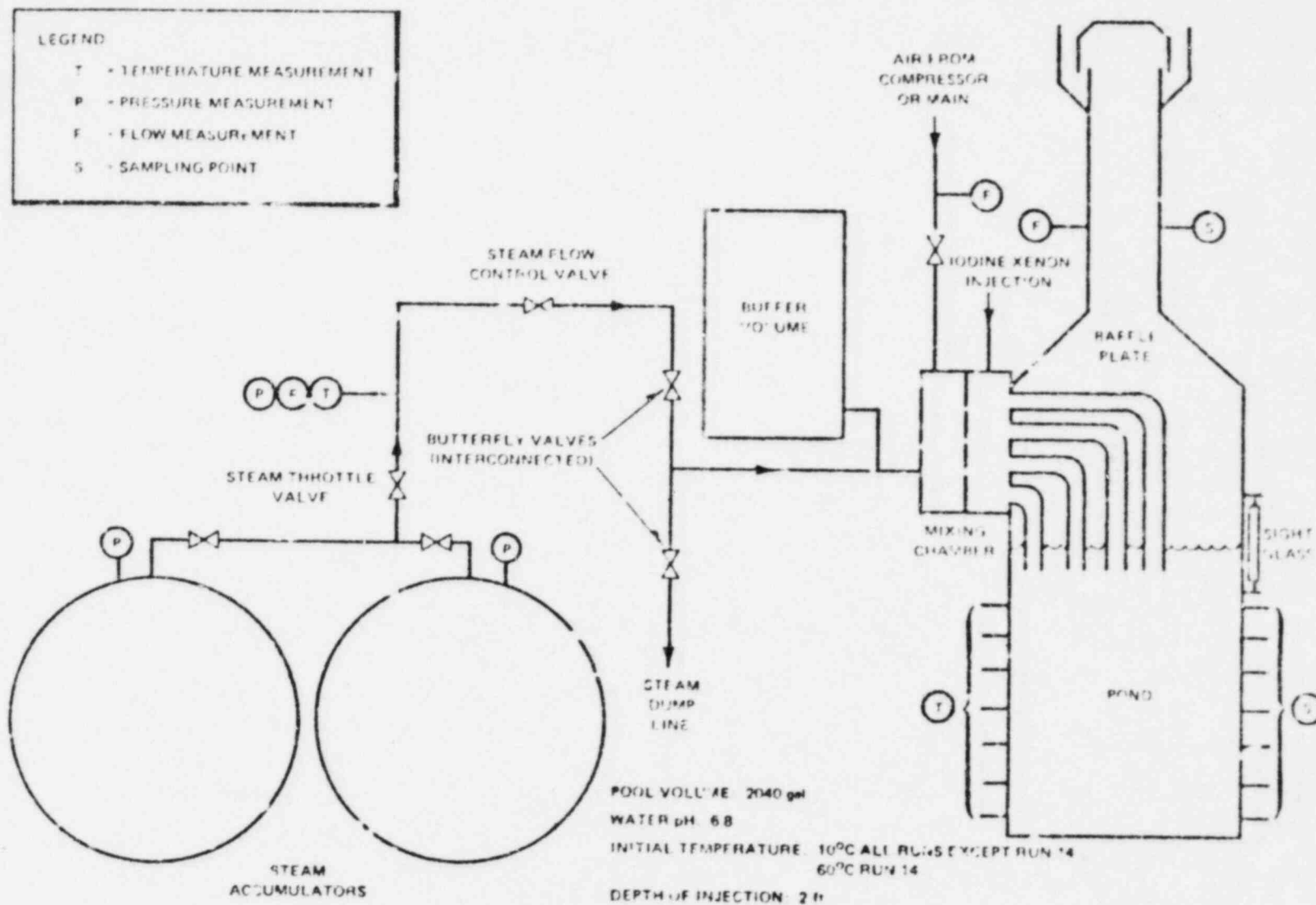


Figure 7.1.1. Transient Pressure Test Facility (Ref. 7.2)

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Figure 7.1.2 Scale Model of the S.G.H.W. Reactor Vented Steam Suppression System (Ref. 7.3)

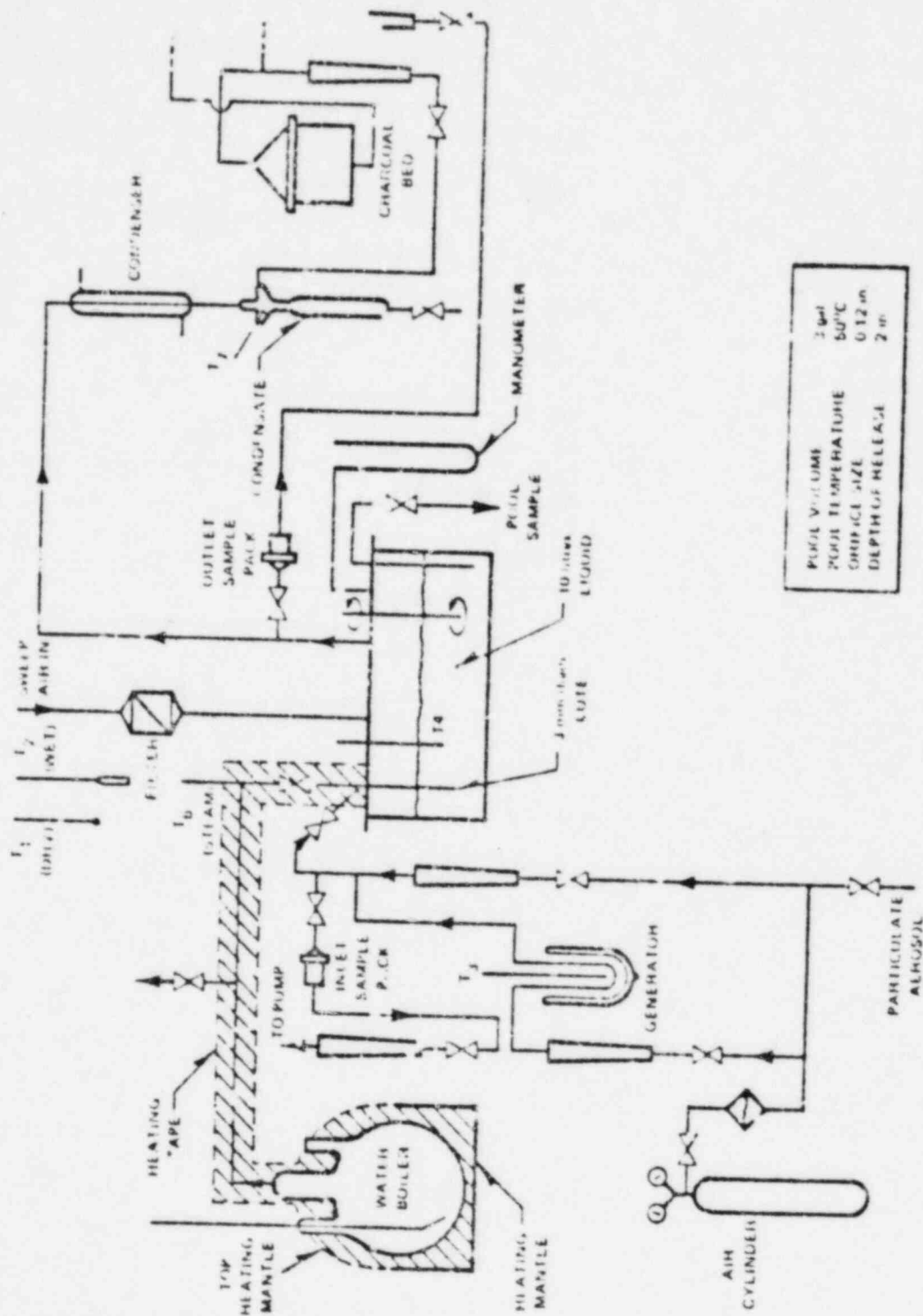
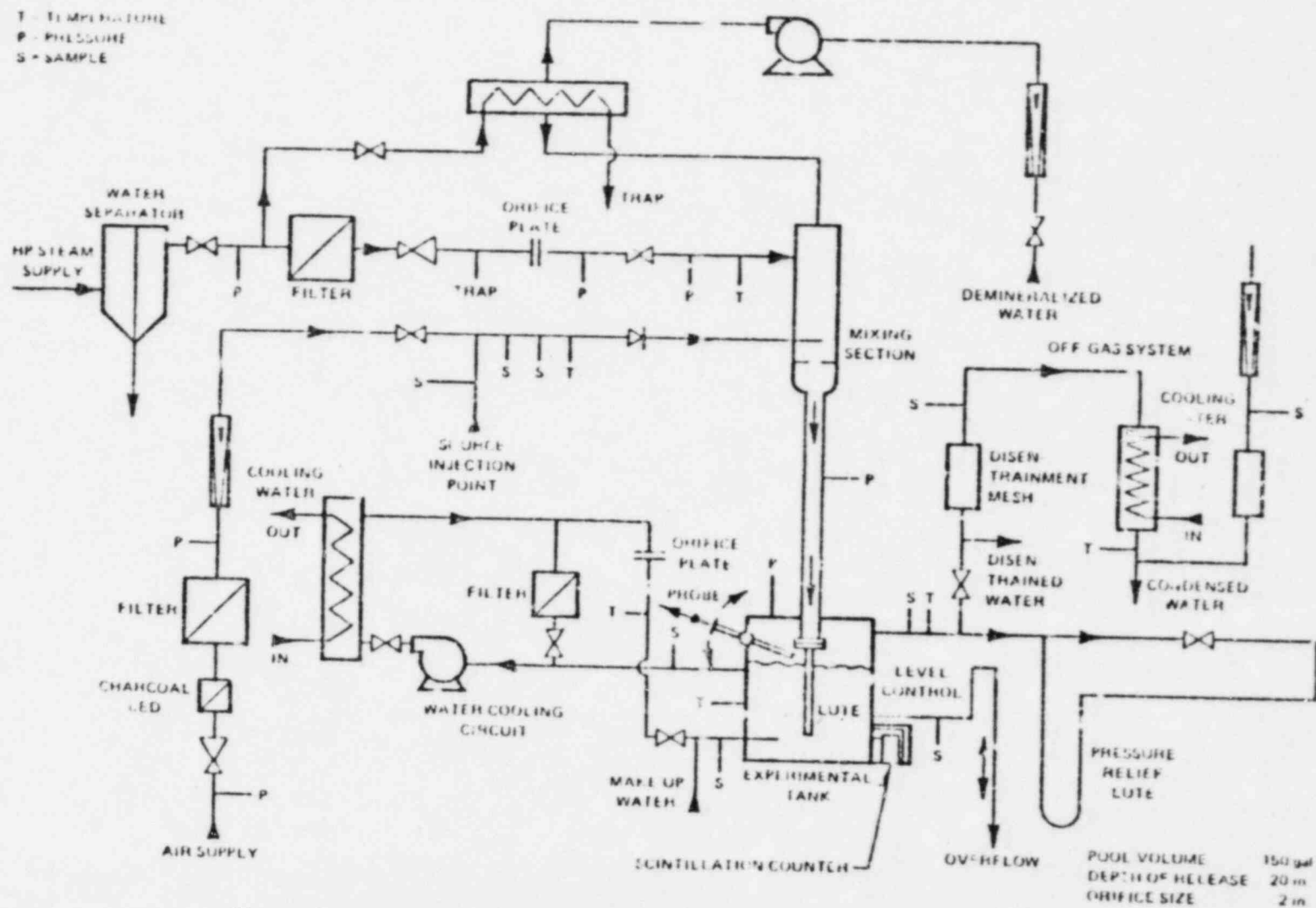


Figure 7.1.3. Small Scale Suppression R_{KK} from D_{0,2} Form (Ref. 7.4)

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Figure 7.1.4. Large Scale Steam Suppression Rig Flow Diagram (Ref. 7.4)

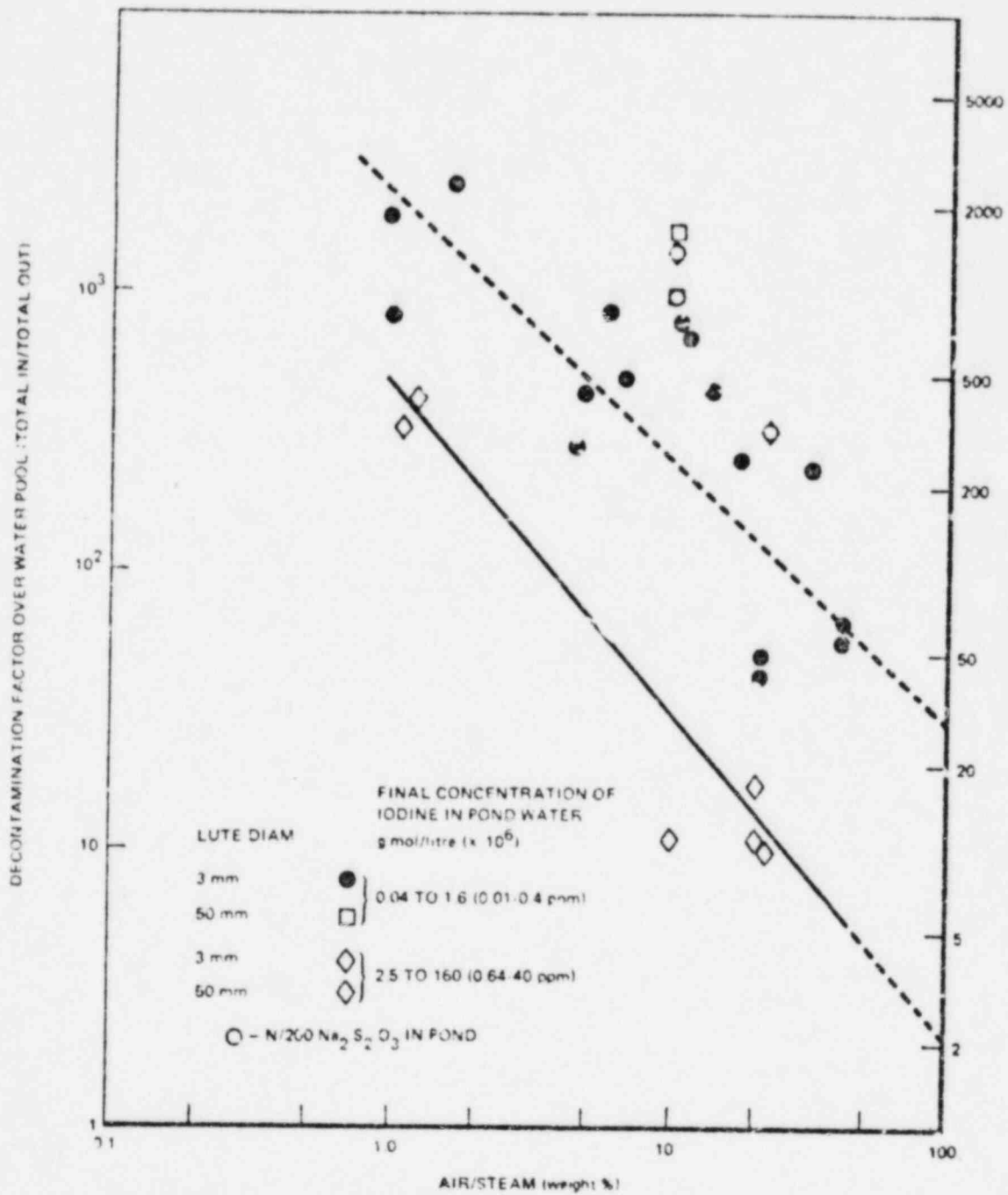


Figure 7.1.5. Removal of Elemental Iodine From a Steam/Air Mixture by Means of a Water Lute (Ref. 7.4)

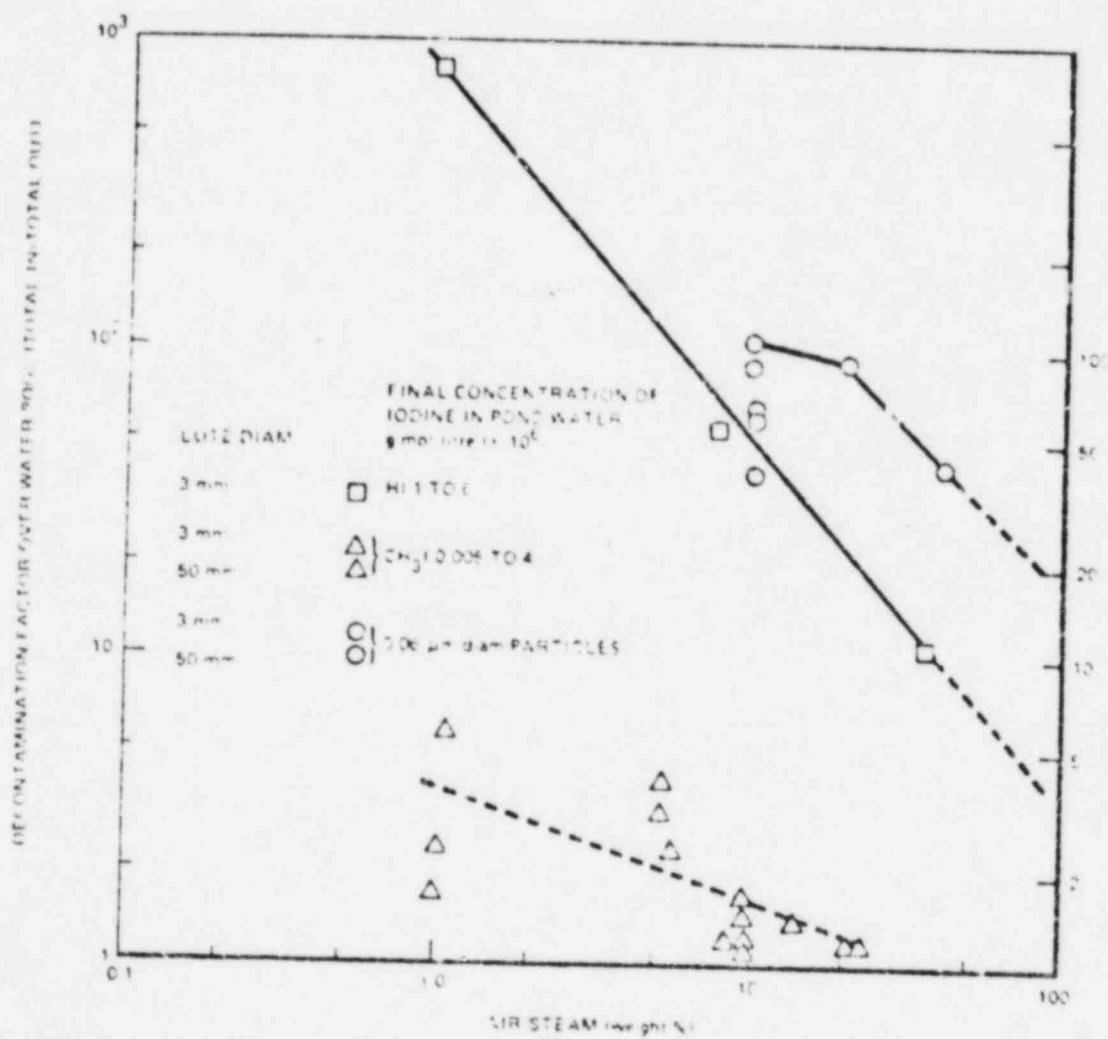


Figure 7.1.6. Removal of Particles and Iodine Compounds From a Steam-Air Mixture by Means of a Water Lute (Ref. 7.4)

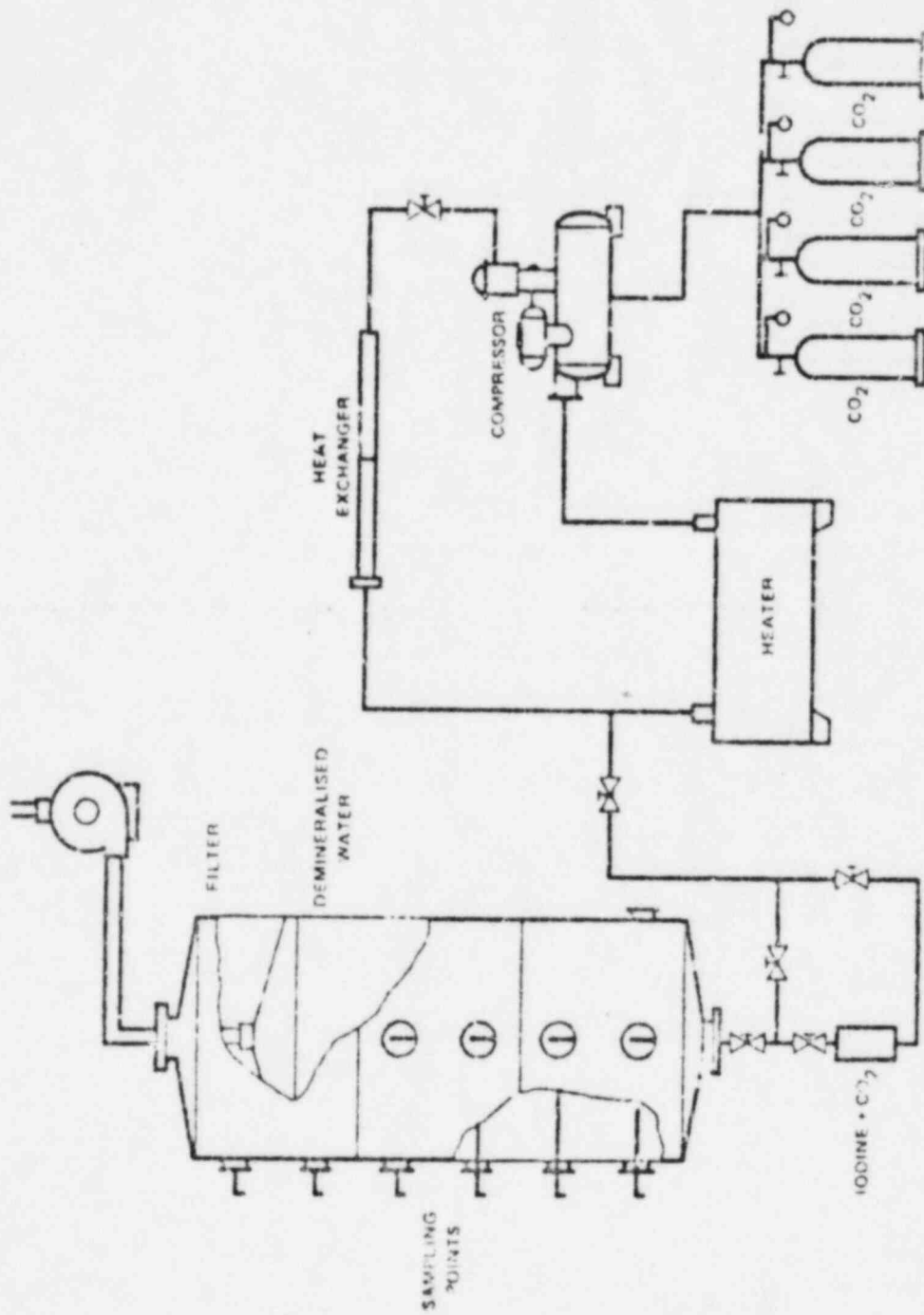
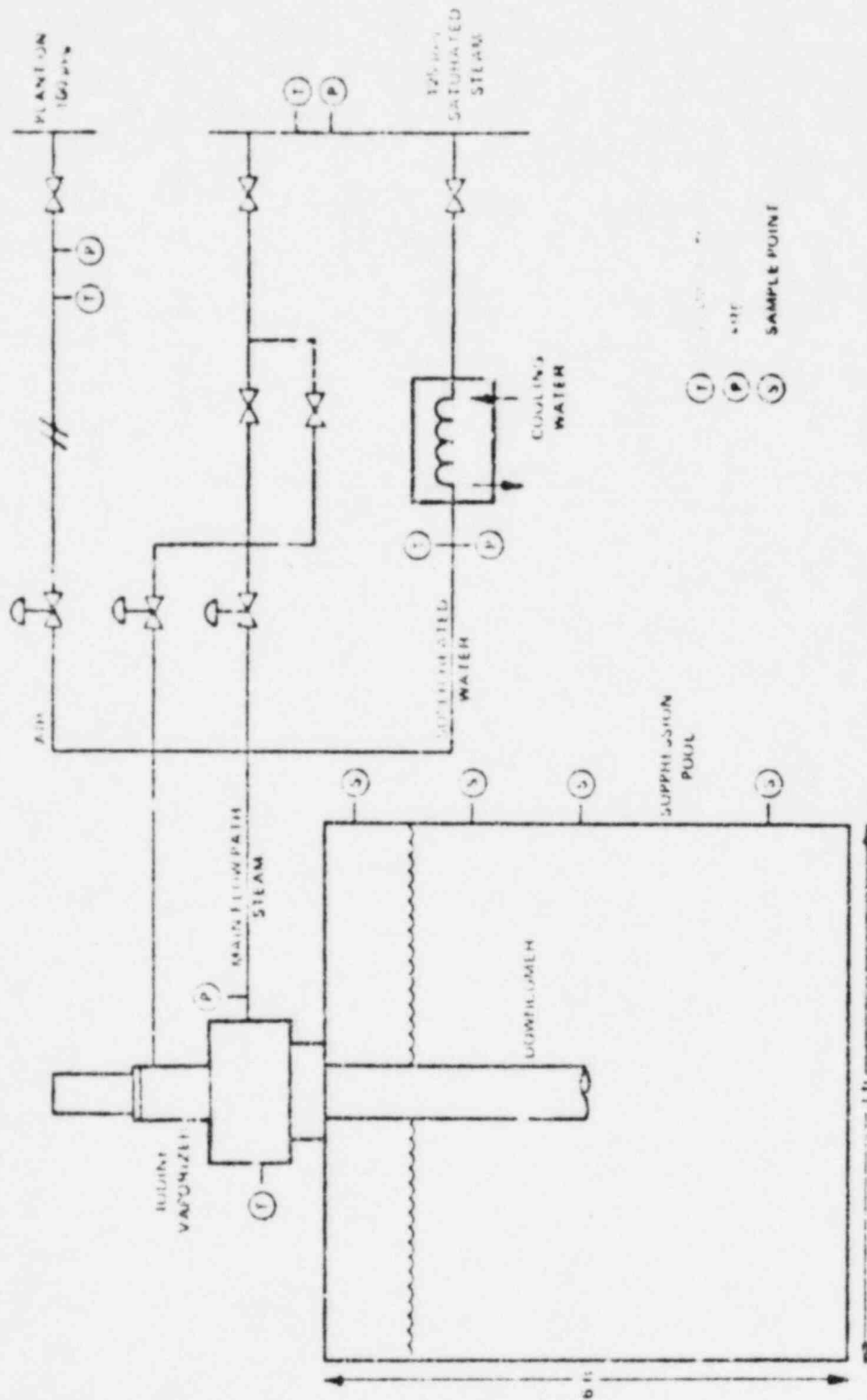


Figure 7.1.7. Removal of Elemental Iodine by a CO₂ carrier Gas (Ref. 7.5)



POOL VOLUME 260 gal
 DEPTH 4 ft
 ORIFICE SIZE 0.68 in.

Figure 7.1.8. Removal of Elemental Iodine by Steam/Air Mixtures (Ref. 7.5)

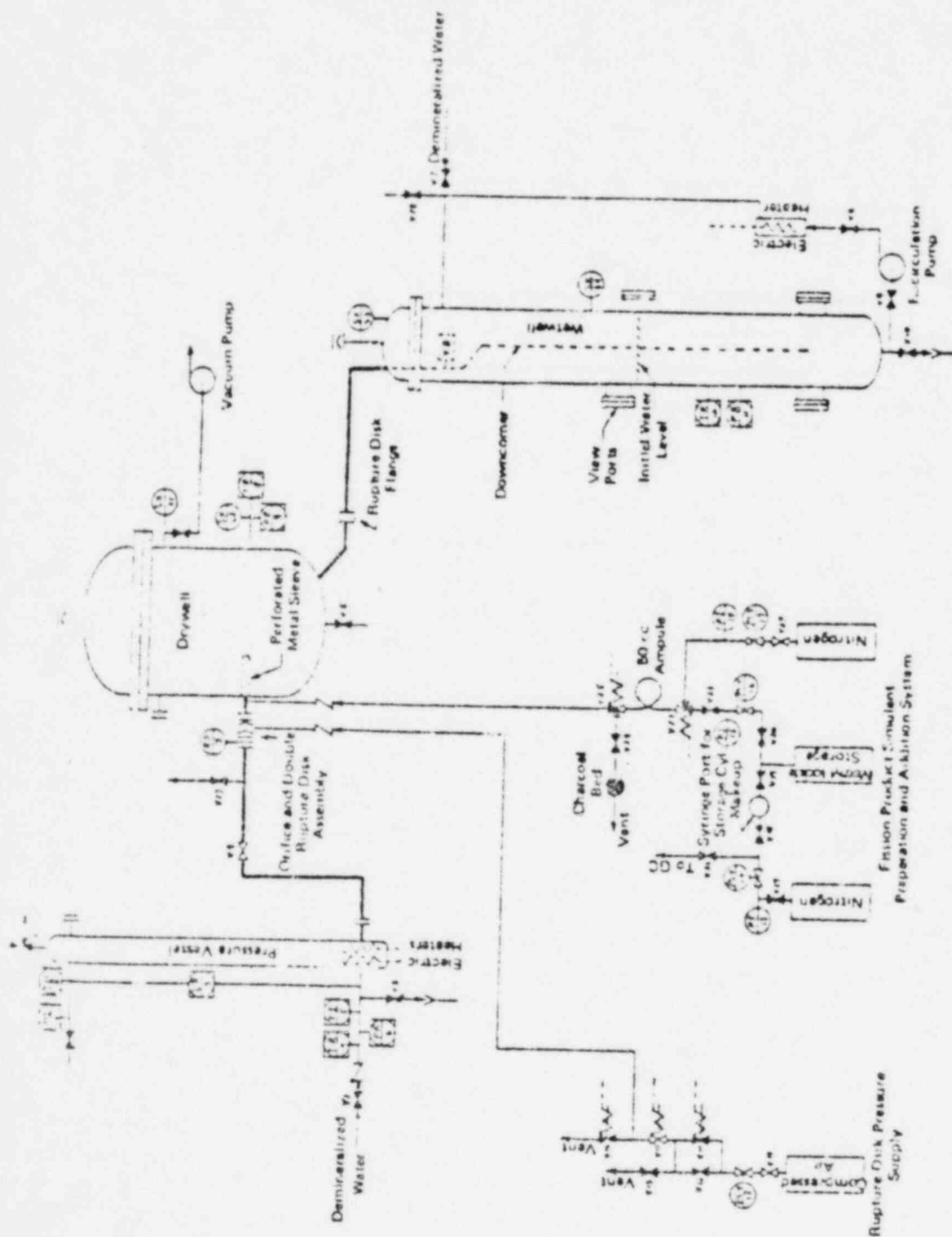


Figure 7.1.9 Fission Product Trapping Facility

(Ref. 7.7)

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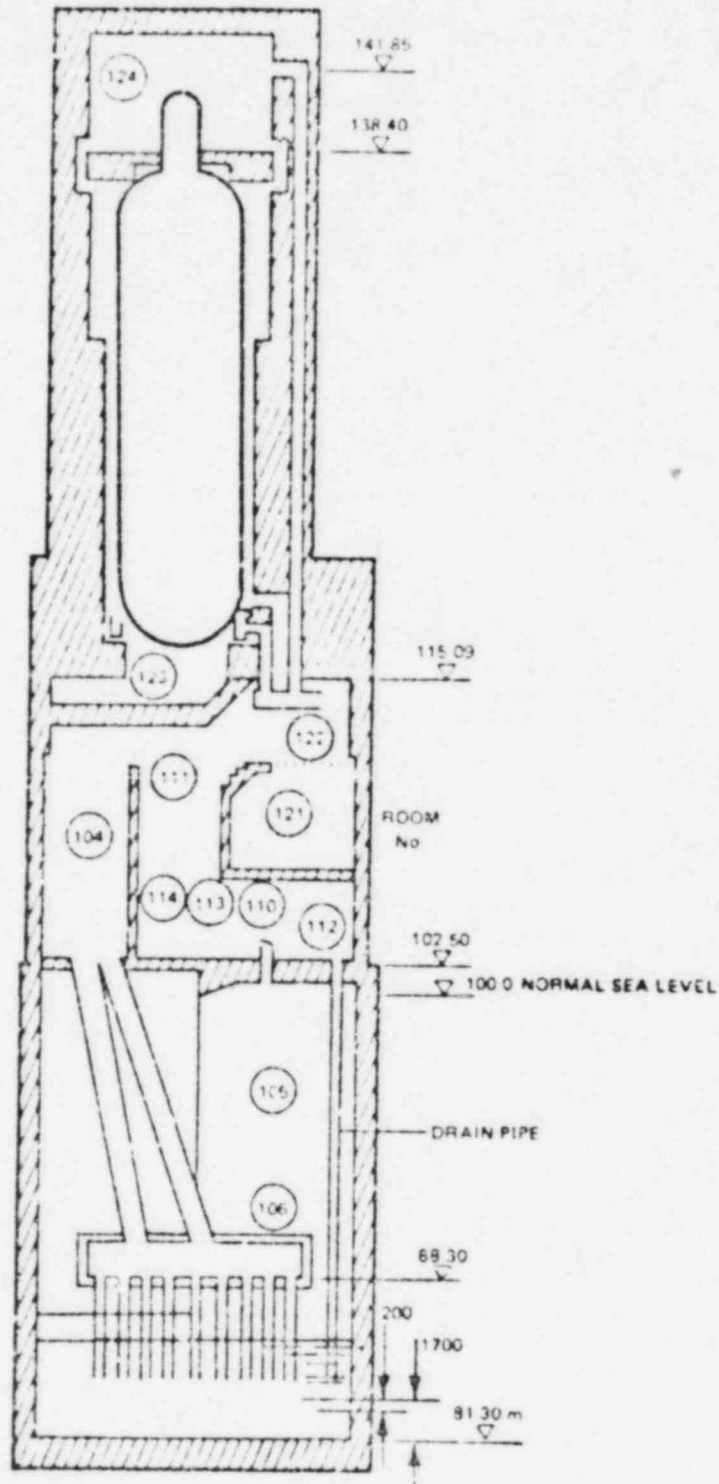


Figure 7.1.10. Cross Sectional View of the Marviken Containment

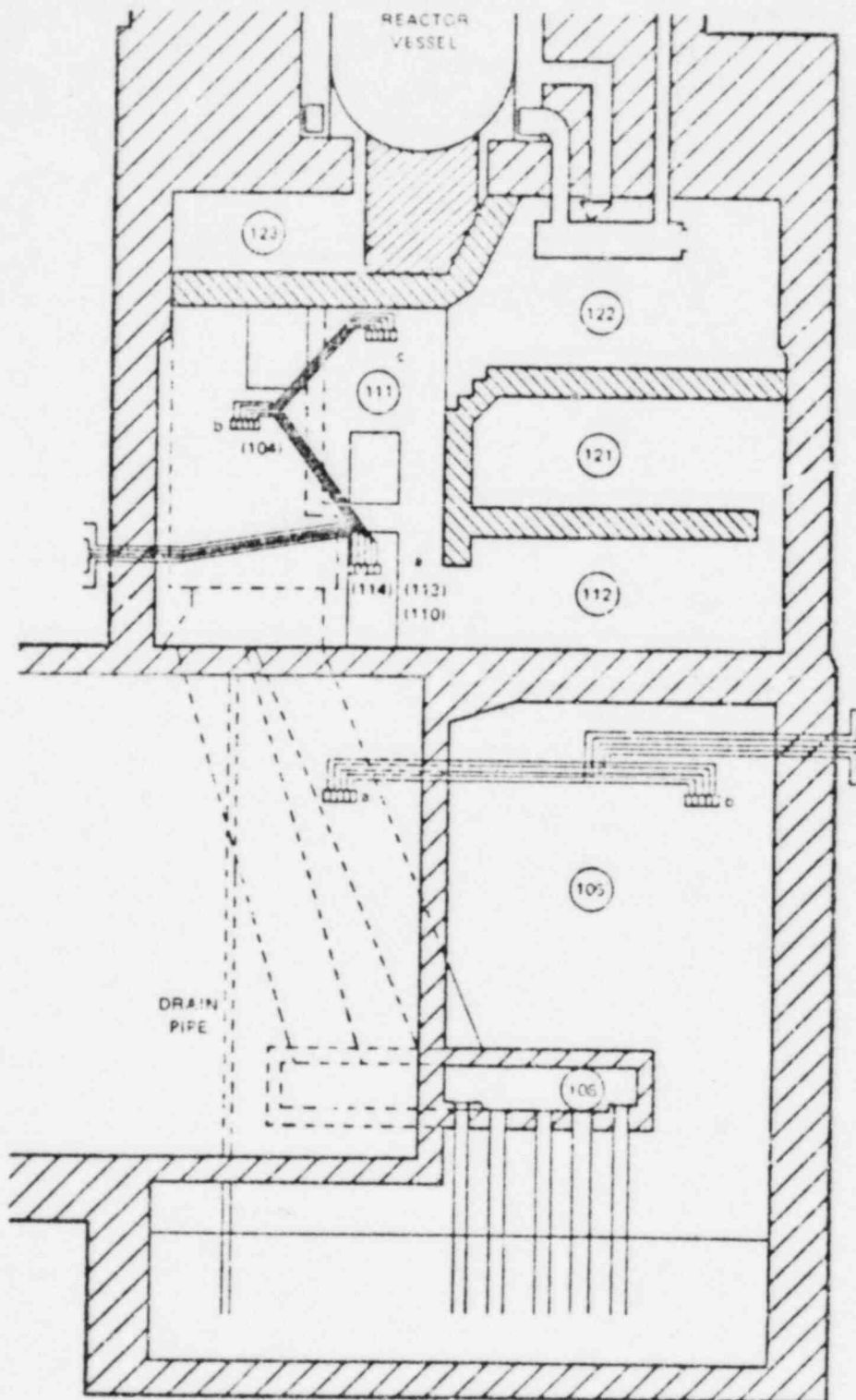


Figure 7.1.11. Marviken Sampling Points and Corresponding Sampling Lines in Wetwell and Lower Drvwell (Ref. 7.8)

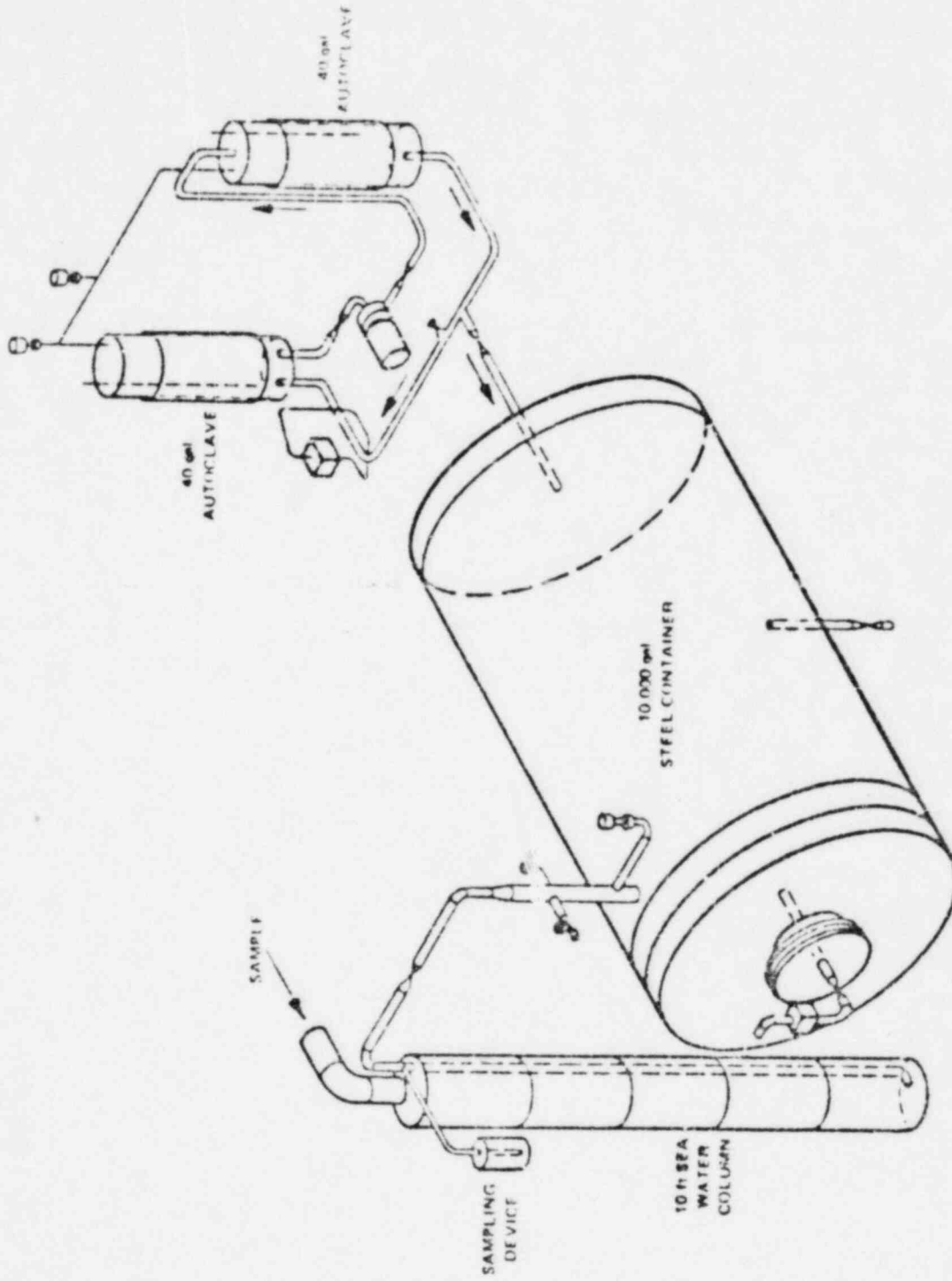


Figure 7.1.12 Flow Schematic for Simulation of Container Venting Under Sea Water

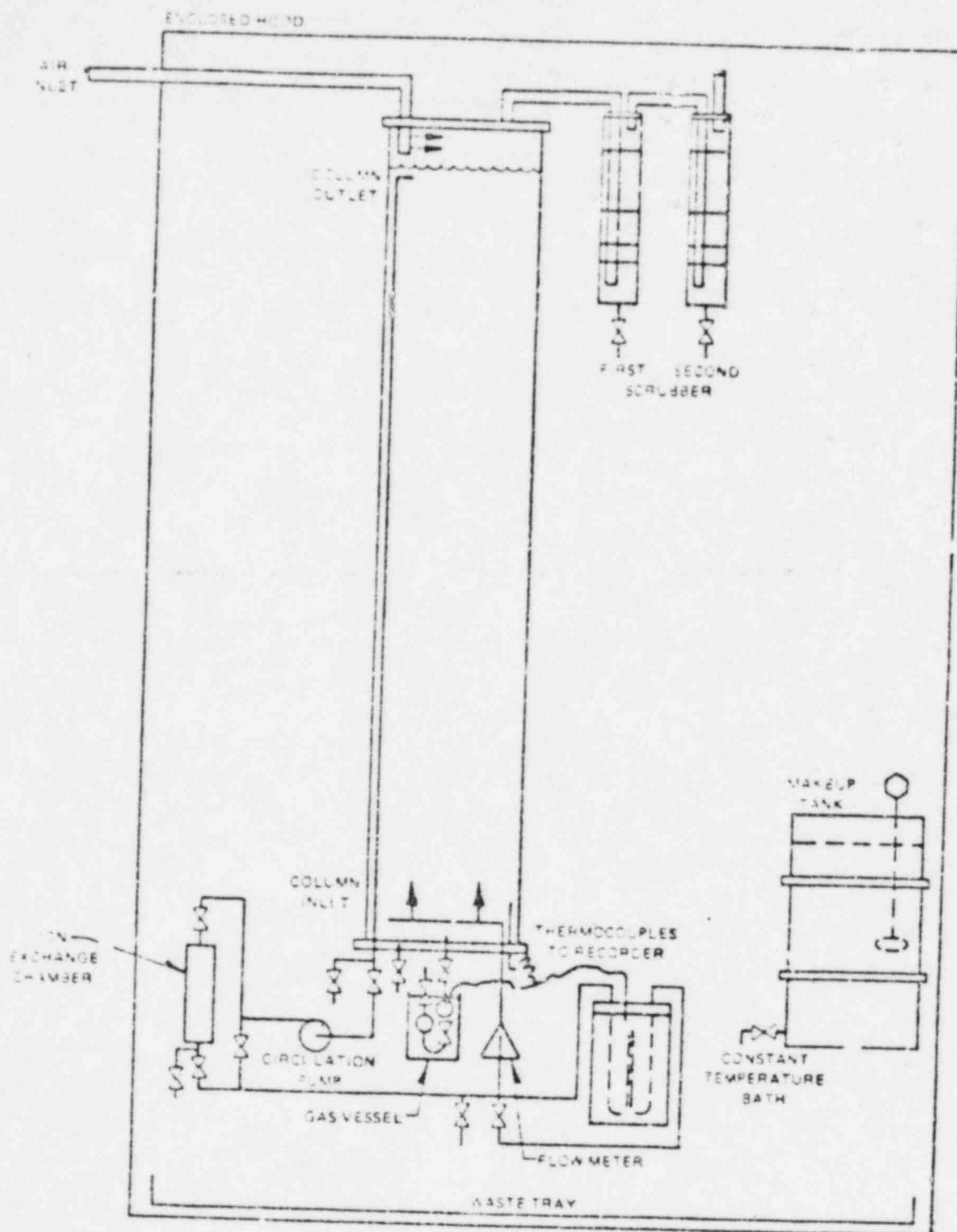


Figure 7.1.13 Small-Scale Test Facility
 I_2 in a Helium Carrier Gas

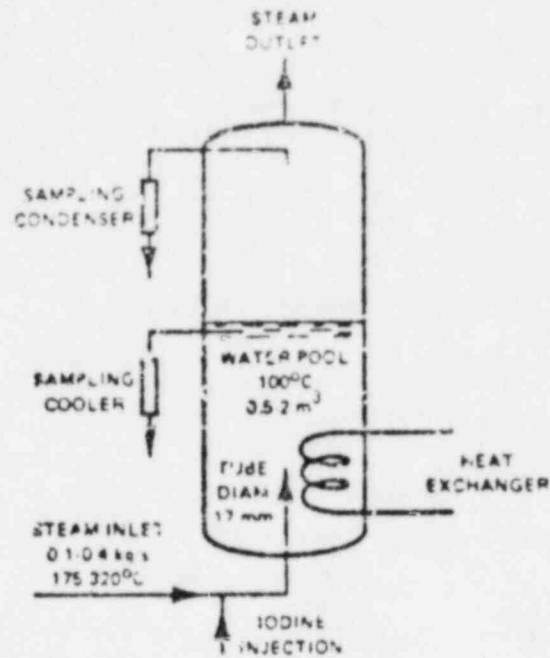


Figure 7.2.1. Principles of Large Scale Experiments on Iodine Decontamination

(Ref. 7.12)

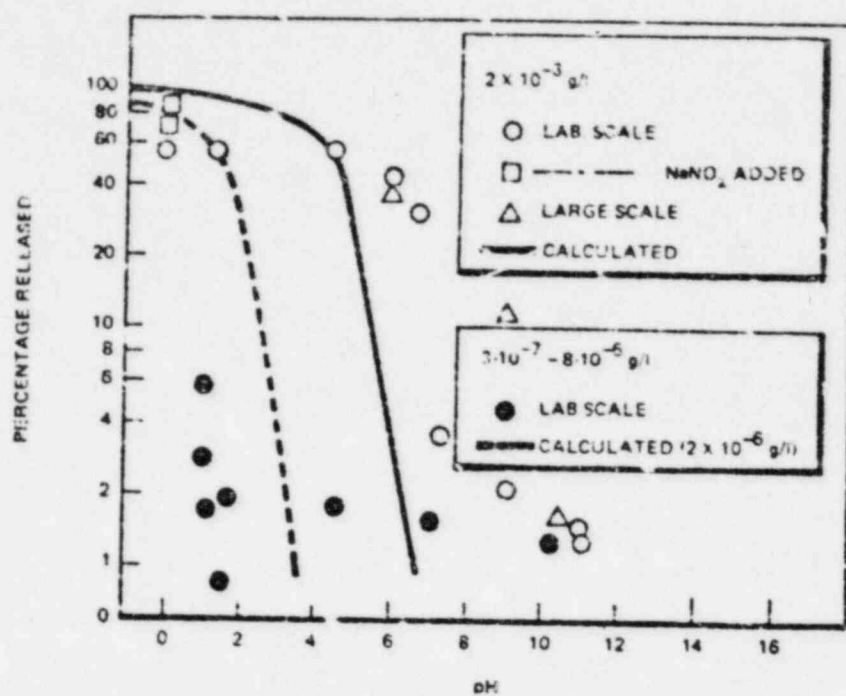


Figure 7.2.2. Release of Iodine From Boiling Water

(Ref. 7.12)

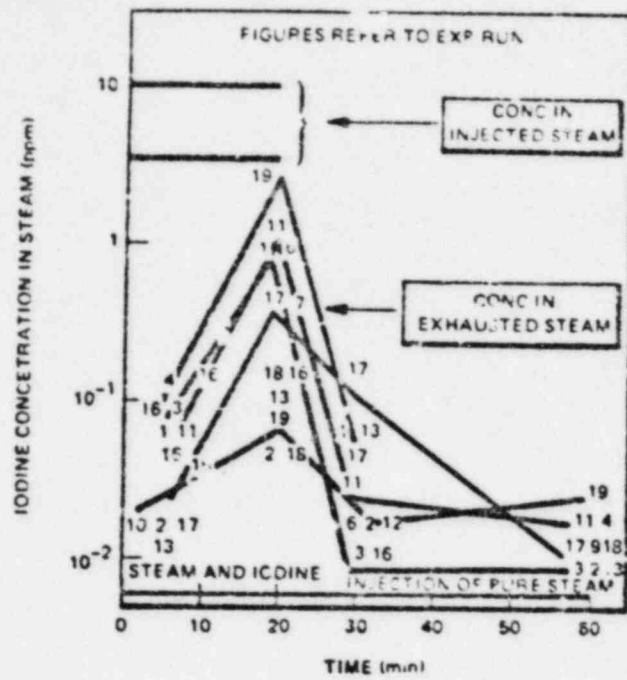


Figure 7.2.3. Iodine Release From a Water Pool at 100°C into Which is Injected an Iodine-steam Mixture (Ref. 7.12)

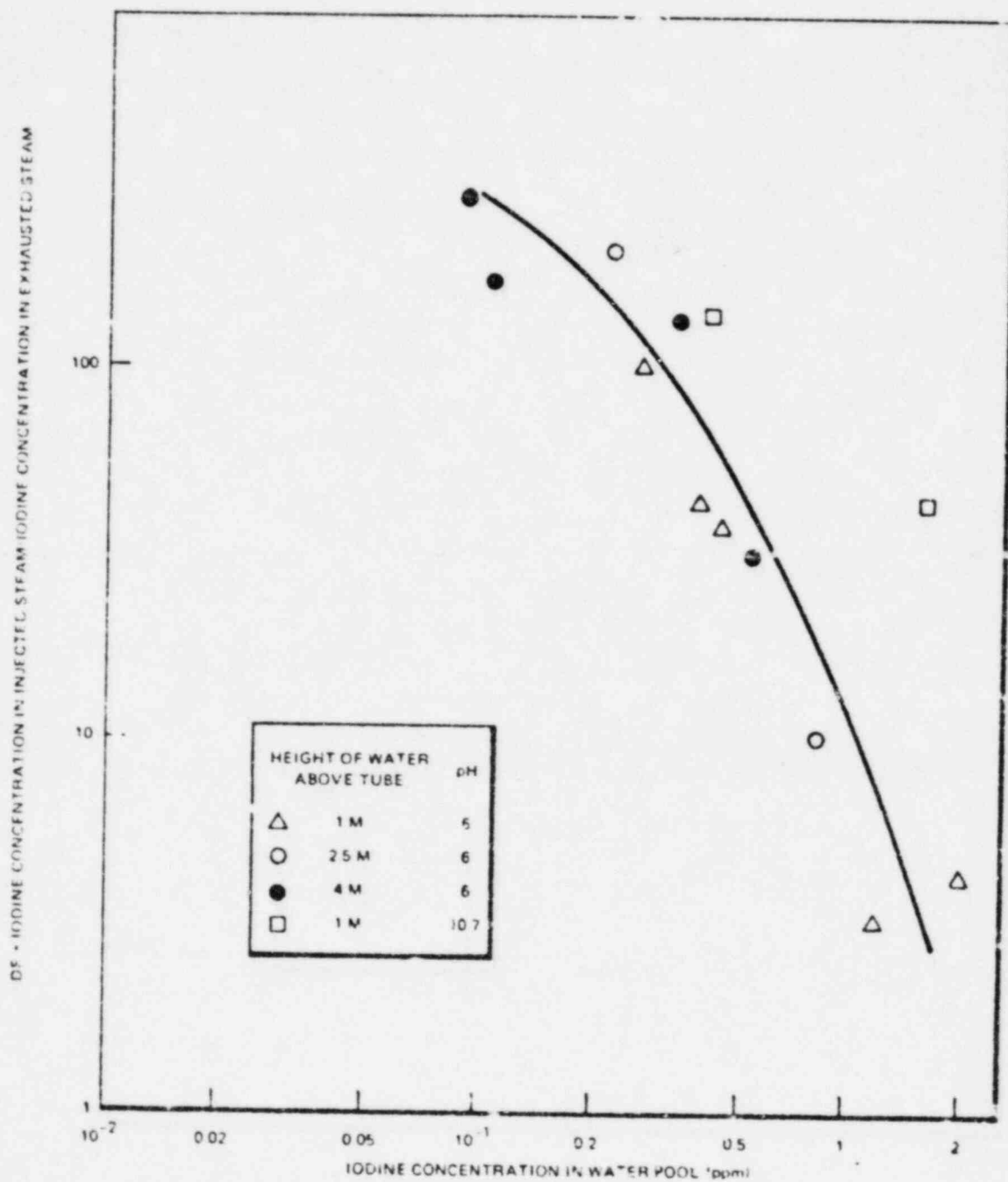


Figure 7.2.4. Decontamination Factors vs Iodine Concentration in a Saturated Pool (100°C)

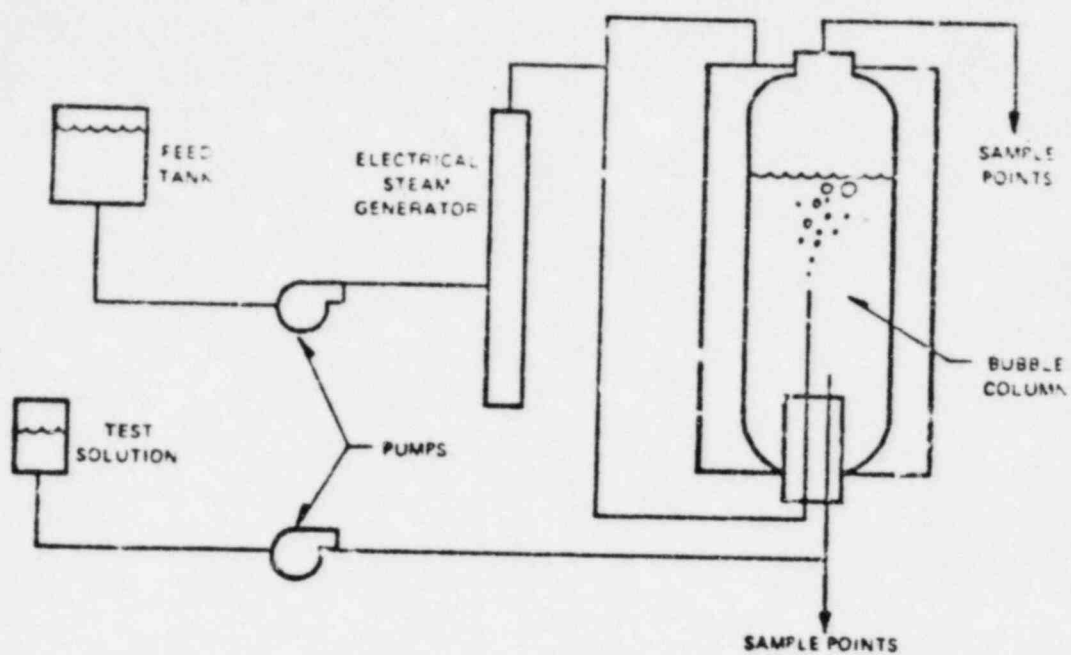


Figure 7.2.5 Test Apparatus to Determine Removal of I_2 from a Boiling Water Column

(Ref. 7.13)

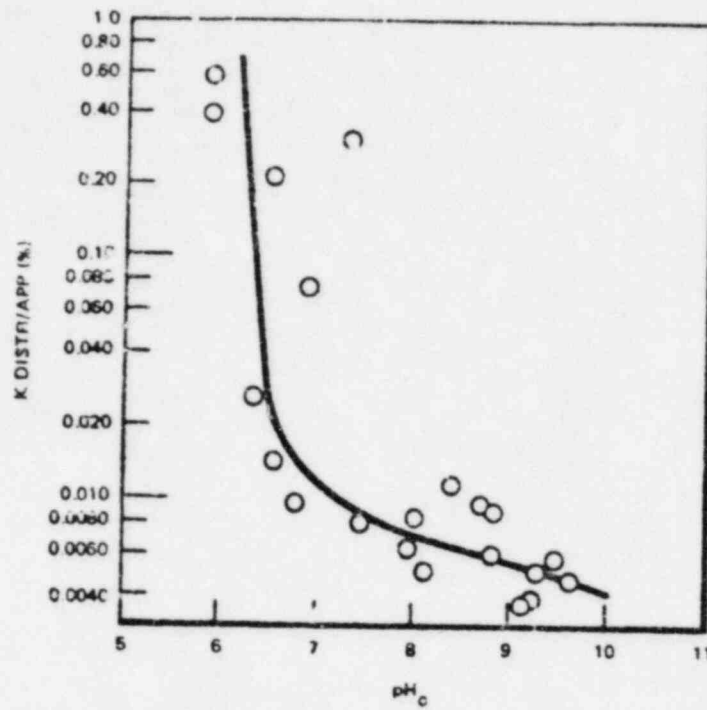


Figure 7.2.6. Dependence of K_{app}^{distr} of Iodine on pH_0 Value at Different Pressures psia: 27

(Ref. 7.13)

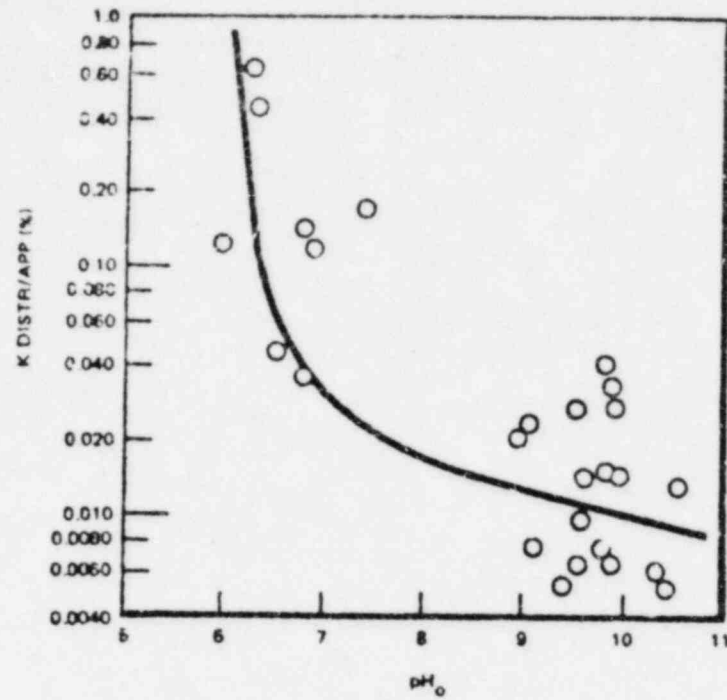


Figure 7.2.7. Dependence of K_{app}^{distr} of Iodine on pH_0 Value at Different Pressures psia: 57

(Ref. 7.13)

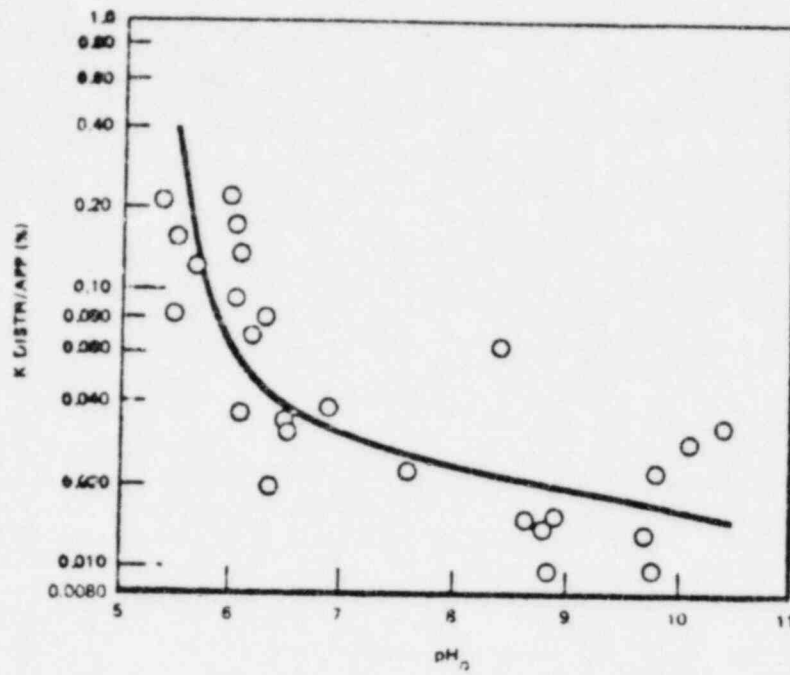


Figure 7.2.8. Dependence of K_{app}^{distr} of Iodine on pH_0 Value at Different Pressures psia: 142

(Ref 7.13)

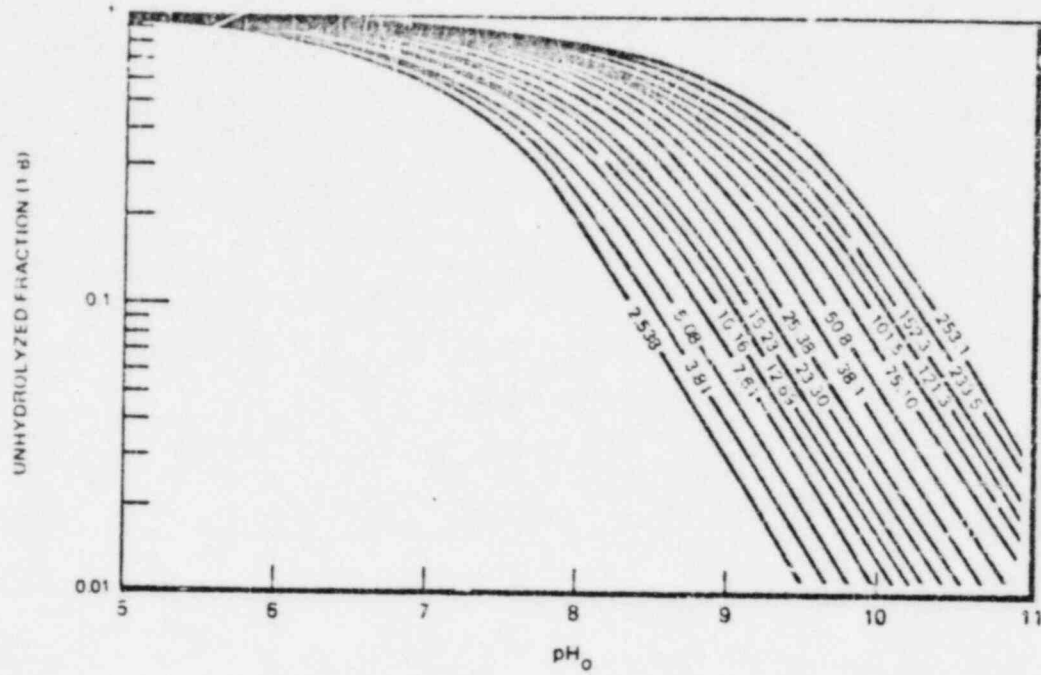


Figure 7.2.9. Dependence of Fraction of Unhydrolyzed Iodine (I-B) on the pH₀ Value of the solution at Various Iodine Concentrations in Solution (mg/liter) (Ref. 7.13)

8.0 Applicability of Pool Scrubbing Test Data to BWR Transport Mechanisms

Based on the experimental test data summarized in Table 7.3.1 and the expected BWR transport mechanisms and transport parameters listed in Table 4.2.1, assessments can be made to determine: whether the experimental data is representative or non-representative of BWR fission product transport conditions and whether the experimental DFs would be conservative or non-conservative when applied to BWR accident scenarios. The results of these assessments are presented in this section along with recommended pool scrubbing factors for each of the dominant BWR transport mechanisms.

In reviewing the experimental data base, it is clear that none of the tests exactly duplicated the expected BWR transport conditions. Several tests, however, are representative of the transport phenomena expected during a postulated accident. For example, the removal processes for small particles and elemental iodine carried by steam/air mixtures through a pool of water are representative of removal processes which occur in a BWR suppression pool. The majority of the tests used volatile iodine (I_2) and thus use of these test results is clearly conservative for cesium iodide in view of the solubility and volatility of CsI.

8.1 Subcooled Pools

Of the BWR transport mechanisms identified in Table 4.2.1., none involve fission product release during rapid depressurization of the reactor in combination with flashing of water and subsequent blowdown to a subcooled suppression pool. Recall that fission product transport to the suppression pool following degraded core conditions is expected to be a relatively slow and steady process during transients following the post-LOCA blowdown period and during the core-concrete vaporization release. In view of these mechanisms, the high flow test data reported in References 7.2, 7.7 and 7.9 are non-representative of expected BWR fission product

transport conditions. However, even during these rather violent depressurization tests through a subcooled pool, the measured overall decontamination factors (plate out and pool scrubbing) were extremely large: 10^5 and 10^7 for I_2 and $2 \mu m$ ZnS_2 particles (Table 7.1.1). While the blowdown tests are non-representative of expected BWR fission product transport conditions, they certainly indicate that in addition to other removal mechanisms a suppression pool can be an additional barrier in retaining fission products, even during rapid and energetic transport conditions.

The smaller scale tests conducted under "steady-state" flow conditions are more representative of the expected BWR transport conditions. Some of the tests reviewed, however, utilized rather shallow pool depths (2 feet) combined with high steam/air velocities. These conditions undoubtedly led to pool scrubbing DFs which were low relative to what would be expected for BWR conditions and this must be considered when assessing the experimental test data.

8.1.1 Retention of Particulates

The retention of particulates by the suppression pool is estimated by two separate methods. The first method is based on the experimental data using $0.06 \mu m$ particles and elemental iodine. Since elemental iodine is expected to be less effectively removed than a particulate (e.g. solids are scrubbed better than gases), using the elemental iodine data will provide a conservative estimate for the retention of particulates. The second method is based on an analytical model which considers retention of particulates in rising bubbles. Both of these methods indicate that the retention factors for particulates are expected to be greater than one hundred.

Several tests (References 7.3 and 7.4) investigated the transport of particles carried by steam/air mixtures through a subcooled pool of water. DFs for $0.06 \mu m$ Ni-Cr particles in steam were approximately 100 or larger. Particulates released from the fuel in a steam environment may grow into large particles due to condensation

effects. WASH-1400 predicted particle sizes on the order of 5-15 μm based on data from the Containment Systems Experiment (8.1) and recommended pool scrubbing $D\text{'s} = 100$ for particulates based on References 7.3 and 7.4. The actual DFs may be much larger under actual BWR conditions, i.e. 8-19 feet of water with small bubbles and complete condensation, larger particles, and potentially soluble fission products attached to particles. In view of these conditions, the available data can support a DF much greater than one hundred ($DF \gg 100$) for particulates in steam.

Particles can also be carried by a CO_2 - steam mixture through a subcooled pool. For these mechanisms, complete bubble condensation may not occur and particle scrubbing will be determined by processes such as settling, absorption, and retention on the interior bubble surface. DFs for these conditions are more difficult to quantify because of the lack of representative data. DFs for particulates, however, can be estimated based on the measured removal factors for elemental iodine. Since particulates would be removed more effectively than a volatile gas, DFs for particulates would be much larger than DFs based on I_2 .

The data that does exist for estimating DFs for particles in a noncondensing gas are based on 0.06 μm Ni-Cr particles in air, 4 μm Na_2O in N_2 gas, and volatile I_2 in air, CO_2 and He gas. Reference 7.4 reported a DF of ~ 50 for 0.06 μm Ni-Cr particles in 30% air and the data in Figure 7.1.6 can be extrapolated to yield a DF of ~ 20 for 100% air. A DF of ~ 10 for I_2 in 20% air was reported and the data in Figure 7.1.5 can be extrapolated to yield a DF of ~ 2 for I_2 in 100% air. Both of these tests were based on a release through 20" of water. In addition Reference 7.11 reported a $DF = 20$ for 4 μm Na_2O particles in a N_2 carrier gas released under 2 feet of water. Thus, DFs for particles are larger than those for volatile I_2 , by approximately an order of magnitude. This is in agreement with what one would intuitively expect. Reference 7.5 reported DFs of ~ 250 for I_2 in CO_2 released below 6 feet of water and DFs of ~ 2000 when the release was below 15 feet of water (refer to

Table 7.1.5). Reference 7.10 reported DFs >200 for I_2 in a helium carrier gas released at a depth of 7 feet below water. Applying the relative relationship between DFs of particles and I_2 in a noncondensable carrier gas, the DFs for particles would be expected to be greater than the values for I_2 at equal release depths.

The retention of particulates can also be estimated by examining particle settling rates and the use of an analytical model. The decontamination factors for large particles in a noncondensable carrier gas ($\geq 5 \mu\text{m}$) will largely depend on the particle settling velocity. The DFs for such particulates can be assessed, therefore, by examining particle settling rates and comparing these rates to the expected bubble rise time through the suppression pool. Particle settling velocities range from 0.25 to 1.0 cm/sec for 5-10 μm particles respectively based on Stokes Law calculations and an assumed particle specific gravity of 4.0. This is realistic for fission product particulates since the specific gravities of cesium iodide, tellerium oxide, and zirconium oxide are 4.5, 5.7, and 5.5 respectively. Based on these expected settling velocities, it would take approximately 0.5 to 2 seconds for 10 to 5 μm particles to settle onto the interior surface of a 1 cm bubble. In addition, turbulence within the bubble may cause the particle to reach the bubble surface even faster. Therefore, particles of this size should have ample time to settle onto bubble surfaces and hence be retained by the water, since bubble rise times in the suppression pool are expected to be on the order of 8-19 seconds depending on the accident scenario.

An analytical model has been developed to predict the removal of particles from single bubbles (8.2). This model was used to estimate pool decontamination factors in 1 cm bubbles having rise times between 8-19 seconds. Results from these calculations indicate that DFs much larger than 100 are expected for 5-10 μm particles having a specific gravity of 4.0. Thus, based on this model particle settling within the bubble should result in substantial removal in the pool.

Therefore, based on 1) conservative application of experimental scrubbing data, or 2) analytical estimates which account for the expected settling rates of particulates in a rising bubble, a DF of 100 or greater is expected for particulates carried through a subcooled pool by a noncondensing gas, and released at a depth of 8 to 19 feet below the water surface.

Vapor release from the molten core/concrete interaction would condense out into an aerosol as it moves to cooler regions away from the limited area of interaction. These aerosols are expected to be approximately $2\text{ }\mu\text{m}$ based on the tests at Sandia (7.1) and should be non-volatile. The aerosol will be carried through the pool in a CO_2 /steam/air mixture and released at a depth of $8\frac{1}{2}$ to $13\frac{1}{2}$ feet depending on the accident scenario. Due to the size and mass of the fission product particulates, pool scrubbing is expected to be more effective than the observed scrubbing of volatile I_2 , or the scrubbing of $0.06\text{ }\mu\text{m}$ Ni-Cr particles. Based on the data presented, a DF of at least 100 or greater is expected for a subcooled pool during core-concrete vaporization releases.

8.1.2 Retention of CsI

Data on iodine scrubbing are limited to I_2 , CH_3I , HI, and HI_0 . There are no reported data on the transport of CsI in steam through a pool of water. However the behavior of CsI as a vapor or as a particle may be inferred from the test results on the behavior of I_2 and small particles. As discussed in Section 5.2, CsI is the expected chemical form of iodine release from the fuel during a core damage accident. CsI would be expected to condense or "plate-out" when it reaches metal surfaces at temperatures at or below $400\text{--}500^\circ\text{C}$, and it would hydrolyze into Cs^+ and I^- ions as soon as water or condensing steam is encountered (8.3). CsI may also be attached to aerosol particles and carried by steam to the suppression pool. For cases where the pool is subcooled, rapid bubble condensation would occur and CsI would rapidly ionize and remain in solution,

since solutions of Cs^+ and I^- are nonvolatile and stable. Experimental data from References 6.13, 6.14, and 7.4 showed DFs of ~ 1000 for HIO in the condenser, DFs of $\sim 10^4$ for HIO transported through 4 feet of water, DFs of $> 10^3$ for HI in steam through 20" of water, and DFs of $> 10^2$ for I_2 in steam through 20" of water. DFs for CsI in steam would be expected to be much higher than 1000 because CsI is much less volatile than HIO , HI , or I_2 . Therefore, a $\text{DF} \gg 1000$ is expected for CsI in steam released at a depth of 8 to 19 feet below the surface of a subcooled pool.

8.2 Saturated Pools

8.2.1 Retention of CsI and Other Particulates

For saturated pools, the behavior of CsI in steam is expected to be similar to the behavior of particles carried by a non-condensable gas through a subcooled pool. Particle settling and retention on the bubble surface is expected to limit the amount of release from the pool. This would be especially true for CsI particles since they would rapidly ionize upon contact with water. Reference 7.12 investigated the effects of I_2 scrubbing by a saturated pool. DFs ranged between 2 and 200 and were a function of I_2 concentration, pH of the water and depth of release (Figure 7.2.4). Extrapolating this data to a concentration of ~ 3 ppm and pH of ~ 10 (maximum expected BWR conditions) a DF of approximately 30 is obtained. This certainly would be a lower bound for volatile I_2 under saturated pool conditions. The scrubbing of CsI particles should be much higher in view of settling processes of particles in the bubble, and ionization of CsI in water. Indeed, experimental data shows that particles are removed more efficiently than volatile I_2 . CsI particles are similar to NaCl particles in that they are both hygroscopic and as such would grow into larger particles in a humid environment and thus would have a greater chance of being removed. Figure 8.1 illustrates how a small NaCl particle can grow into larger droplets in the presence of moist air (8.4). Therefore, DFs of at least 100 or greater for CsI in steam are expected for saturated pools whether the carrier gas is steam or a CO_2 -steam mixture.

8.3 Summary of Minimum Supportable Pool Scrubbing Factors

This assessment has identified the minimum suppression pool scrubbing factors for each dominant BWR transport mechanism for which the data can provide support. Results are summarized in Table 8.1. The actual DFs for scrubbing during a postulated BWR accident are expected to be much higher because the experimental test parameters were conservative (i.e. shallow pool, small particles, and volatile I_2). Potentially attainable DFs which are believed to be supportable by further testing are also presented in Table 8.1.

The application of these scrubbing factors in probabilistic risk evaluations is presented in Section 9.

TABLE 8.1 MINIMUM SUPPORTABLE AND POTENTIALLY ATTAINABLE SUPPRESSION
POOL DECONTAMINATION FACTORS FOR IODINE AND PARTICULATES

Transport Pathway and Associated Event(s)	Minimum Supportable Dfs		Potentially Attainable Dfs (3)
	Subcooled Pool (1)	Saturated Pool (2)	
Reactor pressure vessel to pool via safety relief valve and quencher (Transients)	10^3 CsI, I^- , HI 10^2 particulates 10^2 I_2	10^2 particulates (4) 30 I_2	10^5 - 10^6 CsI, I^- , HI 10^3 - 10^6 particulates 10^2 - 10^3 I_2
Reactor pressure vessel to pool via vents (Transients following RPV depressurization, or LOCA post blowdown period)	10^3 CsI, I^- , HI 10^2 particulates 10^2 I_2	10^2 particulates (4) 30 I_2	10^4 - 10^6 CsI, I^- , HI 10^3 - 10^6 particulates 10^2 - 10^3 I_2
Aerosol Transport to Pool Via Vents (Core-Concrete Vaporization Release)	10^2 particulates 10^2 I_2	10^2 particulates (4) 30 I_2	10^3 - 10^6 particulates 10^2 - 10^3 I_2

- (1) During these conditions, complete condensation is expected when the pool is subcooled.
- (2) A subcooled pool is at a temperature below the saturation temperature corresponding to the pressure in the containment, while in a saturated pool steady state boiling "steaming" is occurring.
- (3) Potentially attainable by further testing (saturated-subcooled pools).
- (4) Includes CsI

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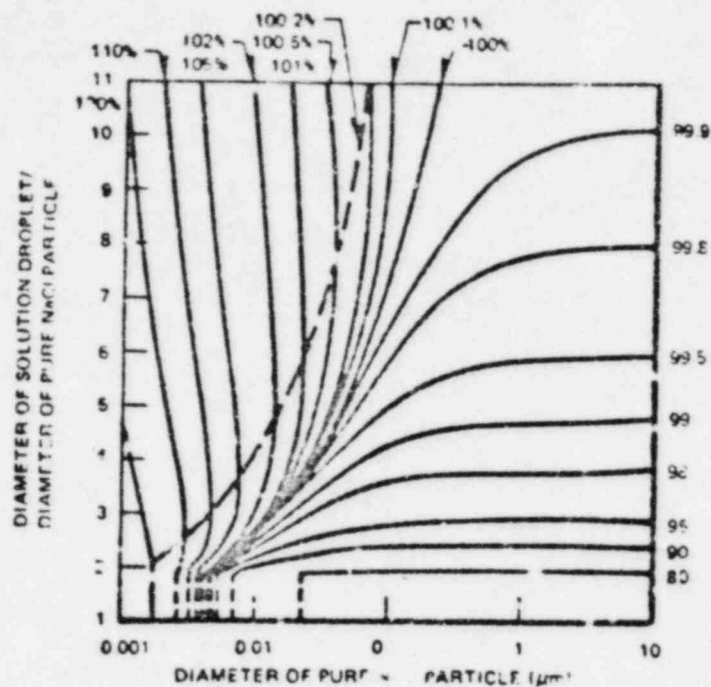


Figure 8.1. The Ratio of the Diameter of NaCl Solution Droplet to the Diameter of the NaCl Particle From Which the Droplet Has Been Formed at Various Levels of Relative Humidities (Ref. 8.4)

9.0 APPLICATION OF POOL SCRUBBING FACTORS IN PROBABILISTIC ASSESSMENTS

In this section the attenuation factors recommended in section 8.0 are applied to the accident scenarios used in BWR Probabilistic Risk Assessments. The accident scenarios are classified in terms of the initiating event and the presence or absence of ECCS flow. Initiating events include anticipated transients, anticipated transients without scram (ATWS) and loss of coolant accidents (LOCA). These events can be represented by four dominant core damage classes which are defined as follows:

- Class 1: A transient, a small break, or an intermediate break LOCA coupled with insufficient makeup water leads to core damage.
- Class 2: Following a transient or any size LOCA, the core is covered with sufficient makeup water. However containment heat removal capability is lost, resulting in primary containment cracking due to overpressure and a possible subsequent loss of makeup water¹ and core damage.
- Class 3: A large break LOCA or an ATWS without sufficient makeup water leads to core damage.
- Class 4: Following an ATWS event the reactor does not immediately become subcritical. However, the core remains covered with sufficient makeup water. Continuous blowdown to the suppression pool results in boiling of the suppression pool, containment pressurization, and primary containment cracking, which for low probability events may also lead to loss of makeup water¹ and core damage.

¹ Loss of makeup water is due to loss of Emergency Core Cooling System NPSH and not the loss of the suppression pool water.

Classes 1 & 3 postulate that a core damage occurs before primary containment cracking, and involves a subcooled pool. Classes 2 & 4 postulate primary containment cracking prior to core damage and involve a saturated pool.

Depending on the postulated initiating event, fission product transport to the suppression pool may occur by way of the SRV/quencher lines, the horizontal vents, or a combination of both pathways. Minimum supportable DFs for each pathway are listed in Table 8.1. In Table 9.1 LFs are applied to each postulated class of accident.

It should be re-emphasized that the values in Table 9.1 are judged to be minimum values supported by the available literature. It is expected that if experimental programs were conducted which include anticipated chemical forms of fission products and representative flow rates, bubble sizes, pool depths, etc., appropriate to the hypothetical accident scenario, that the larger DFs would be proven to exist. However, until such data are available the use of the attenuation factors in Table 9.1 in probabilistic risk assessments is recommended for a realistic estimate of the consequences expected for such accidents.

TABLE 9.1

APPLICATION OF POOL DFs FOR EACH POSTULATED ACCIDENT SCENARIO

SCENARIO	POOL CONDITION	POOL DF
Class 1 and 3	Subcooled	1000 CsI 100 other particulates 100 I ₂
Class 2 and 4	Saturated	100 CsI and other particulates 30 I ₂

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		GOVERNMENT CLASS
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