

APPENDIX A

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FISSION PRODUCT REMOVAL EFFECTIVENESS OF CHEMICAL  
ADDITIVES IN PWR CONTAINMENT SPRAYS

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## ABSTRACT

The presence of gaseous iodine in severe accident situations is based upon a regulatory source term prescription whose basis predates the accident at Three Mile Island-Unit 2 and the source term research that TMI-2 stimulated. This report reviews the current best-estimate of source term characteristics, and the experimental bases that establish the effectiveness of spray additives. Based on this review, several current operating practices, vis-a-vis the addition of additive(s), may warrant regulatory reevaluation.

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## 1. INTRODUCTION

Commercial pressurized water reactors (PWR) are equipped with Containment Spray Systems (CSS) to limit the peak pressure in containment below the design pressure in the event of a blow down associated with a design basis accident. The CSS is composed of spray pumps, spray rings, nozzles, and necessary pipes and valves. Coolant is supplied by the Refueling Water Storage Tank. Many CSS are also equipped with a spray additive tank and an associated pump. These additives are intended to increase the capacity of the coolant to absorb gaseous iodine in the event of fuel clad failure or core melt. Hence, an important secondary function of the CSS is the attenuation of fission products released to the containment. Section 6.2.2 of the Standard Review Plan (SRP)<sup>1</sup> describes the performance objectives of the CSS as a heat removal system, while SRP 6.5.2 addresses the function of the CSS as a fission product cleanup system.

The performance objective of CSS as a fission product cleanup system, given the source term assumptions given in Regulatory Guide 1.4,<sup>2</sup> coupled with the containment leakage rate is to aid in meeting the design basis accident (DBA) dose guideline of 10CFR100.<sup>3</sup> The basis of source term prescription given in Regulatory Guide 1.4 is given in Reference 3. The source term consists of 100% of the noble gases (Xe, Kr), 25% of the iodine and 1% of other solids. Iodine is assumed to be primarily gaseous based on the observed release from the Windscale accident. (Regulatory Guide 1.4 further prescribes the following iodine chemical composition: 91% elemental, 5% particulate, and 4% organically bound). The Atomic Energy Commission adopted this source term to establish criteria for licensing of plant/site combinations and assessing the potential hazard to the public. The use of this hypothetical source term, which would be associated with substantial core damage to assess the consequences of a DBA, which should not result in any substantial core damage since the Engineered Safety Features (ESFs) should terminate the accident prior to core damage, was viewed as intentionally conservative. Application of this approach led to the conclusion that gaseous iodine dominated the off-site radiation doses. This in turn led to increased efforts to scrub iodine from the containment atmosphere, including the use of a chemical additive to increase the effectiveness of the sprays to absorb and retain gaseous iodine.

On March 28, 1979, Three Mile Island Unit 2 experienced a partially mitigated loss of coolant accident. Substantial core damage occurred and significant amounts of radionuclides were released from the fuel. Environmental monitoring of the accidental releases indicated 0.02 to 0.08 of the noble gas inventory and only  $3 \times 10^{-7}$  of the iodine inventory were released to the environment. No metallic radionuclides are known to have been released. The difference in the fractional releases of noble gases and iodine were attributed to the following:

1. Noble gases are inert, volatile, and only slightly to fairly soluble in water.
2. The chemically reducing environment in the reactor vessel promotes the stability of cesium iodide which is nonvolatile (in the containment atmosphere) and water soluble.

3. Injection of sodium hydroxide into the CSS would have enhanced the absorption of gaseous iodine if it was released during the accident.
4. Filters effectively trapped iodine in the auxiliary fuel handling building from which environmental releases occurred.

The inference that the majority of iodine released from the TMI-2 reactor vessel was CsI and not molecular iodine focused attention upon the TID-14844<sup>4</sup> source term assumptions and the measures taken in response to these assumptions, e.g., the design of the engineered safety features.

This report focuses on the technical data base that is available to support the use of chemical additives in the CSS. Computer searches of several literature data bases were also carried out to identify relevant materials. These searches are documented in Appendix A:

## 2. PAST AND CURRENT SOURCE TERM CHARACTERISTICS

Regulatory Guide 1.4 prescribes that the following source term be considered, for example, in assessing the DBA doses guidelines values set forth in 10CFR100:

1. 100% of the noble gas inventory,
2. 25% of the iodine inventory; Composition - 91% molecular, 5% particulate, 4% organic.
3. Release to the containment assumed to be instantaneous and well mixed in the containment atmosphere.

NUREG-0772 addressed the impact of source term assumptions on regulation.<sup>5</sup> Specifically, the impact of the observation that particulate CsI and not gaseous I<sub>2</sub> was the predominant chemical form of iodine released to containment was assessed. Based upon the analyses presented in NUREG-0772, it would appear that except for those accidents in which the fission products are released through water, the amount of iodine calculated to be released would not be substantially reduced by the chemical form (I<sub>2</sub> or CsI).

As a result of the observations of radionuclides released at TMI, substantial research efforts were initiated. An effort sponsored by the NRC has resulted in a set of computer codes, the Source Term Code Package (STCP), which simulates the progression of severe nuclear reactor accidents and estimates the release of materials from the fuel, through the reactor coolant system and to the environment. The function and status of these codes are described in NUREG-0956.<sup>6</sup> Some typical results of the code package and preliminary observations regarding accident source terms are also presented. In addition, the basis of STCP methodology has been reviewed by a study group of the American Physical Society.<sup>7</sup> While this review understandably noted many areas of uncertainty and identified several phenomena not fully analyzed, it generally concluded that considerable progress has been made since the publication of the Reactor Safety Study (RSS).<sup>7</sup> (The RSS, published in 1975, also predicted substantial releases of gaseous iodine to the containment, and was more or less consistent with the Regulatory Guide 1.4 Source Term Prescription.)

In comparison to the Regulatory Guide 1.4 source terms, several substantial differences exist in regard to the characteristics of the fission product (FP) release predicted by current, state-of-the art methods of source term estimation. These characteristics have been described elsewhere, e.g., see References 7 and 8. A brief summary of the characteristics is given below.

The total release of FP material to the containment can be divided into two distinct phases: the initial, in-vessel phase where material is released from the damaged or melted fuel and the ex-vessel phase where material is released from the core/concrete interaction. In the in-vessel phase, the release is dominated by noble gases (-100% release), cesium (-100%), iodine (-100%), and tellurium (-30-70%), which are rather volatile at the temperature excursions predicted during core degradation. Very much smaller amounts of the refractory groups, Ba, Sr, Ru, La, and Ce, are predicted to be released in-vessel. As the volatile materials, with the exception of the

noble gases, migrate away from the core to cooler regions in the reactor coolant system (RCS), they are assumed to condense on surfaces or onto aerosols. Based upon observations made at TMI-2, and subsequent thermochemical analyses,<sup>5</sup> iodine is assumed in the STCP to be present as CsI, and any iodine release from the RCS is modeled as an emission of CsI in aerosol form. The behavior of aerosols released from the RCS can be modeled in several alternate fashions which depend upon the particular type of reactor and sequence being considered. For BWR sequences, where the RCS blowdown is vented through a suppression pool,<sup>9</sup> aerosol decontamination factors (DF) are calculated by SPARC, a computer code within the STCP. Similarly, for PWR's<sup>4</sup> equipped with ice condensers, the ICEDF code within STCP is executed to estimate aerosol DF's associated with this ESF. Ultimately, the behavior of aerosols in the containment atmosphere is simulated by the code NAUA-4. This code models several natural processes, e.g. aerosol agglomeration and settling, that can deposit airborne aerosols onto reactor surfaces and, hence, result in a decrease in the airborne activity. The duration of the in-vessel FP release is limited to the period from the start of core degradation to bottom head failure. The onset of core damage is plant and sequence dependent. Typical estimates of the times for the start of core melt are 25 minutes, Surry AB sequence, and 135 minutes, Surry S<sub>2</sub>D,<sup>8</sup> from the time of scram. In the AB sequence, the CSS is assumed to fail. In the S<sub>2</sub>D sequence, the CSS is operational, and sprays initiate 20 minutes after scram. It is interesting to note that by the time the release of FP's has begun, the CSS has already entered the recirculation mode. It should be noted that current regulatory guidance effectively requires immediate injection of additives into the sprays, once the CSS is initiated.

The ex-vessel FP releases result from the core/concrete interaction where gases generated from the decomposition of concrete sparge through the molten core debris. The large volumes of gases which pass through the melt and the increased surface area associated with these gases accelerate the vaporization of melt constituents, which subsequently condense into aerosols after leaving the melt. Another mode of aerosol generation is also modeled. This is the formation of mechanical aerosols which are a result of the gas bubbles breaking through the upper melt surface. Hence, all FP release from the core/concrete interaction is modeled as being in aerosol form. The ex-vessel release is dominated by the Ba, Sr, Ru, La, and Ce groups. In general, a small fraction of the core inventory of iodine is predicted to be retained in the core debris at the time of bottom head failure and is passed to VANESA, a computer code in the STCP which estimates ex-vessel releases, for release during the ex-vessel phase. VANESA assumes this iodine release to be in the form of CsI aerosol. The duration of the ex-vessel release starts shortly after bottom head failure and is typically calculated ten hours beyond initiation, although the majority (~90%) of ex-vessel release generally occurs within three hours of the initiation of the core/concrete interaction.

In summary, results of severe accident simulation with the state-of-the-art methodology incorporated into the STCP indicate two phases for fission product release. The in-vessel phase is associated with core degradation and releases are dominated by noble gases, cesium, iodine, and tellurium. Iodine is assumed to be in the chemical form CsI. With the exception of the noble gases, all releases from the RCS are in aerosol form. The ex-vessel release phase results from the interaction of the molten core and the concrete



basemat. The ex-vessel release is dominated by the Ba, Sr, Ru, La, and Ce groups. Small amounts of volatiles are also released ex-vessel, notably iodine in the form of  $\text{CsI}$ . All releases are in the form of aerosols. When appropriate intermediate codes, SPARC AND ICFDF, estimate FP aerosol retention in ESFs. The removal of airborne aerosols, generated either in-vessel or ex-vessel, by natural deposition processes is estimated in NAUA.

The STCP, as currently implemented, does not model any gaseous iodine release, nor is there any explicit modeling of gaseous iodine behavior, or the effect of any chemical additive to the CSS. The physical washout of aerosol by sprays is modeled and results in substantial removal of aerosols from the containment atmosphere. Figure 1 shows a typical comparison of the accumulated masses of material leaked to the environment for a sequence, Zion  $\text{S}_2\text{D}-\epsilon$ , where the CSS operates and another sequence, Zion TMLB'- $\epsilon$ , where the CSS is assumed to fail.<sup>6</sup> A similar comparison for the Surry plant is shown in Figure 2.<sup>6</sup> The reduction of  $\text{CsI}$  aerosol leaked to the environment for these reactor-sequence combinations is approximately 50 to  $10^5$ , respectively. Hence, given the current best estimate of the fission product, release characteristics and the processes that can act on the FP releases, the physical action of the CSS and natural aerosol removal processes which proceed with or without the CSS can substantially reduce airborne concentration of FP's. This includes iodine as it is modeled to appear in aerosol form. It is noteworthy to mention at this point that the CSS chemical additive(s) effect only gaseous forms of iodine,  $\text{I}_2$ ,  $\text{HI}$ , and, depending upon the specific additive, organic iodine, and do not contribute to the physical removal of aerosols. Aerosol removal by CSS washout is modeled as a purely physical process. The most common additive (namely,  $\text{NaOH}$ ), however, may play a secondary but important role in ~~raising~~ the sump pH and mitigating radiolysis assisted evolution of iodine in gaseous form.

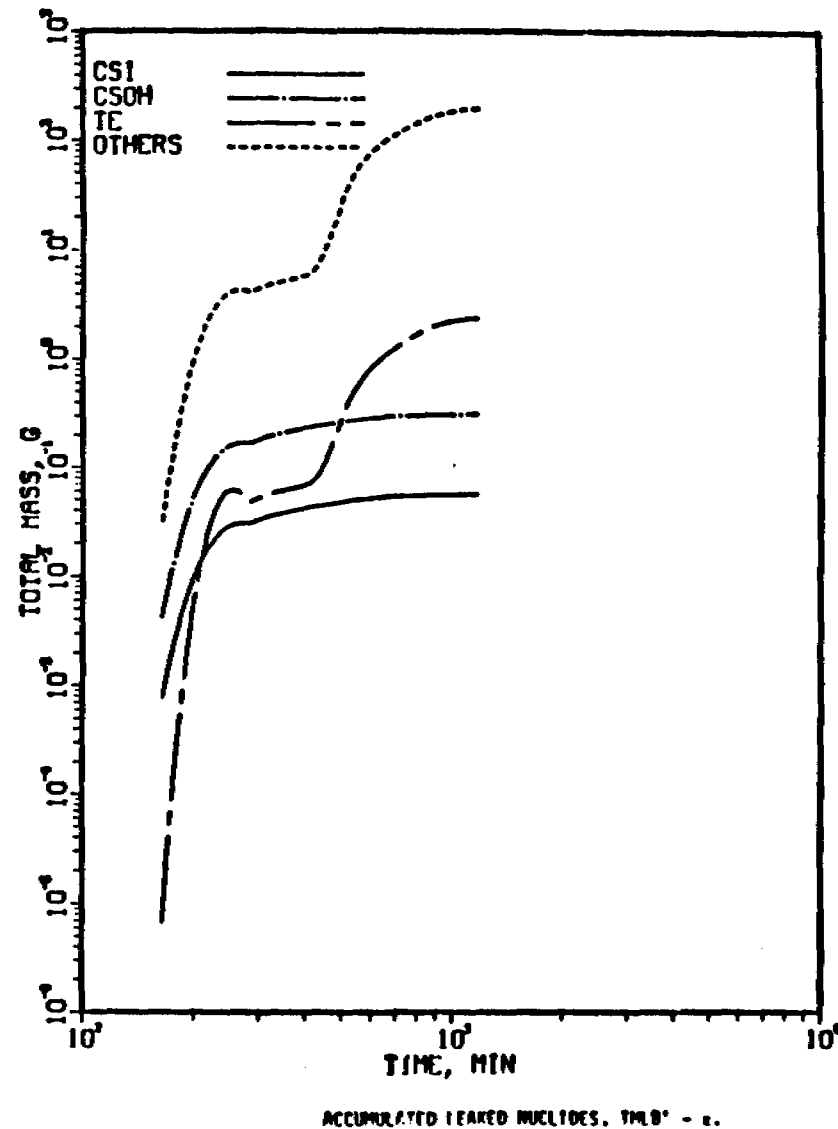
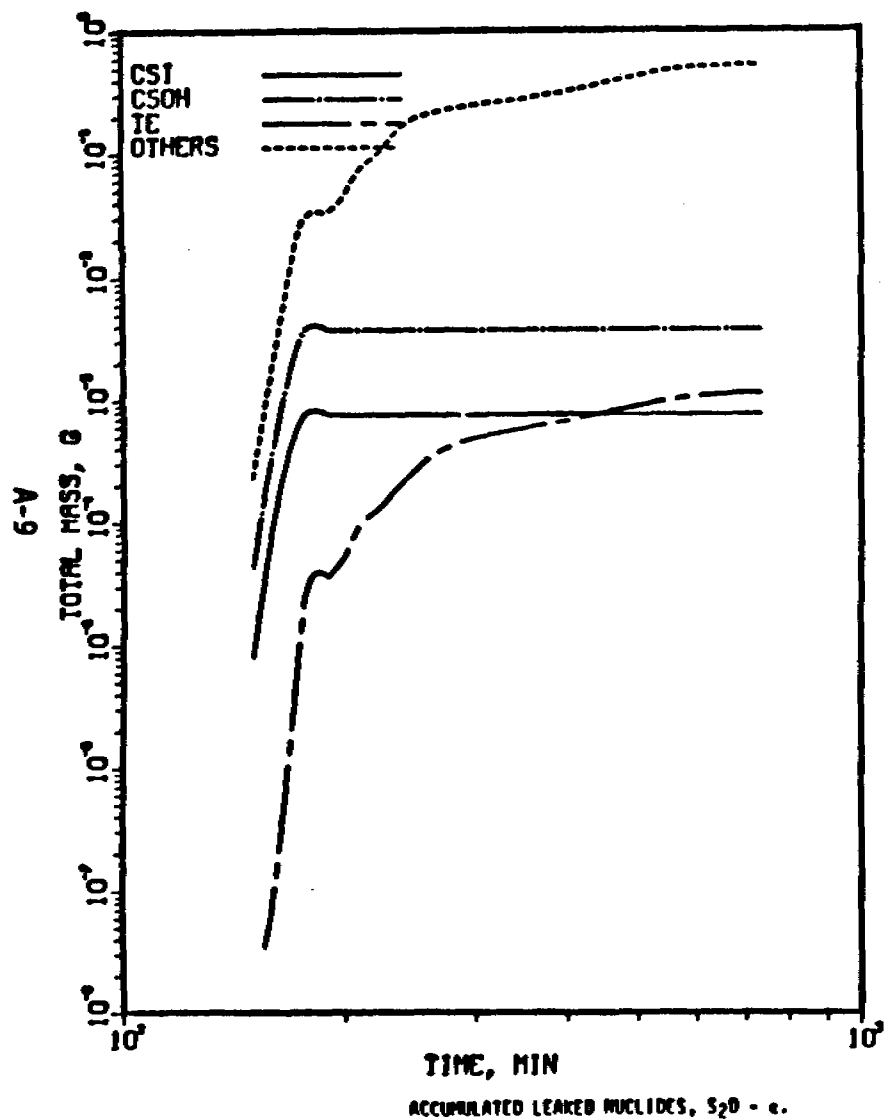
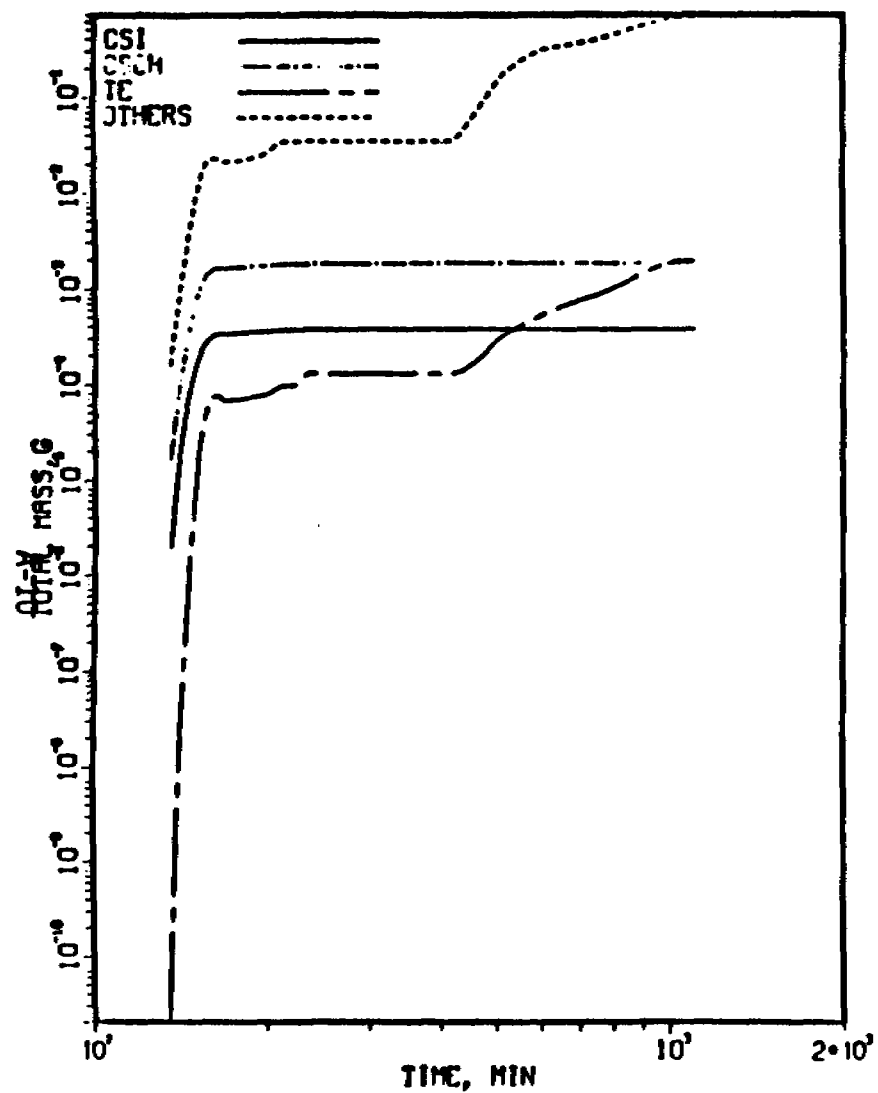
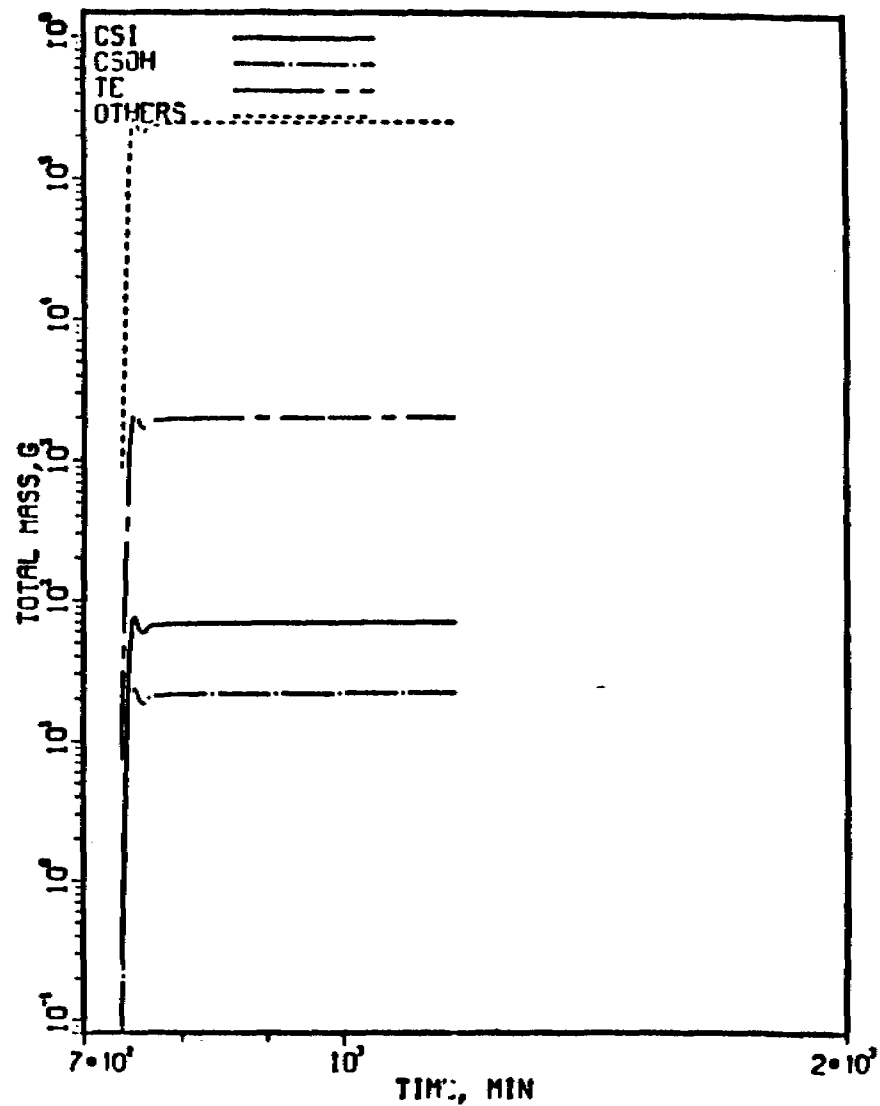


Figure 1 Comparison, for Zion, of the airborne aerosol mass suspended in containment with and without containment sprays.



ACCUMULATED LEAKED NUCLIDES, S<sub>2</sub>D-1



ACCUMULATED LEAKED NUCLIDES, TMLB-1

Figure 2 Comparison for Surry of the airborne aerosol mass suspended in containment with and without containment sprays.

### 3. CSS EFFECTIVENESS AS A FP REMOVAL SYSTEM

As mentioned heretofore, adoption of the TID-14844 source term assumptions to assess the consequences of DBA resulted in the conclusion that molecular iodine dominated off-site dose. This, in turn, fostered consideration of the CSS as a fission product removal system and resulted in the addition of additives, namely, sodium hydroxide for  $I_2$  and HI removal and sodium thiosulfate for the further removal of organic iodine, to increase the effectiveness of the spray solution to absorb and retain iodine.

A primary data base upon which much of the thinking regarding the effectiveness of spray as a fission product removal system was the large scale containment system experiments (CSE).<sup>10</sup> The CSE were carried out in a one-fifth linear scale containment having an internal volume of 751 m<sup>3</sup>. Experiments A-3, A-4, A-6, A-7, and A-8 were performed to determine the effectiveness of CSS to remove airborne FP. The major variables considered included containment temperature and pressure, spray solution composition, and initial or recirculatory phase. Details of the experiment may be found in the original references<sup>10</sup> or the concise summary provided by Albert,<sup>11</sup> which is reproduced here as Appendix B. The results are displayed in the appendix in Figures 46 through 50 (original text numbering) as plots of airborne iodine to the concentration versus time. The absorption iodine was generally interpreted as being governed by a first order process:

$$\frac{dc}{dt} = -\lambda C,$$

where C is the airborne concentration of iodine, t is the time, and  $\lambda$  is the first order removal constant. The results are tabulated in Table 1. Only the first spray period results are given since other processes, such as desorption from wall or the effect of inhomogeneous mixing, complicate the interpretation of subsequent spray periods.

Several observations can be noted. The DF's, ratio of iodine concentration prior to spray initiation divided by the concentration immediately after spray has been stopped, range from 5 to 100. The differences in run A-3 and run A-4, may in part, reflect the change in spray flow rate. The differences in A-4 and A-6 could result from either the increased buffering capacity of the spray solution or the change in initial containment temperature, 25 versus 124°C, respectively. However, a comparison of runs A-5 and A-6 with run A-7, where the solution is unbuffered  $H_2BO_3$ , suggest the latter. In addition, it is interesting to note that runs A-6 and A-7 gave comparable DF's, indicating that in the initial injection phase buffering, pH control, had little observable effect. A superficial comparison of the results given in Table 1 for runs A-7 and A-8 indicate comparable performance on the basis of DF, while showing significant difference in  $\lambda$  or  $t_{1/2}$ . This results from two changes in the experimental procedure: spray nozzles that delivered a smaller mean drop size and a shorter duration of initial spray operation. Given these changes to the experimental protocol, the results of run A-8 would appear consistent with the former runs.

Table 1 Summary of CSE First Spray Results<sup>a</sup>

Run	Solution Composition <sup>b</sup>	$t_{1/2}$ <sup>c</sup> (min)	$\lambda$ (min <sup>-1</sup> )	DF <sup>d</sup>
A3	525 ppm H <sub>3</sub> BO <sub>3</sub> , pH 9.5	5.5	0.13	5
A4	525 ppm H <sub>3</sub> BO <sub>3</sub> , pH 9.5	1.4	0.50	100
A6	3000 ppm H <sub>3</sub> BO <sub>3</sub> , pH 9.5	2.1	0.33	30
A7	3000 ppm H <sub>3</sub> BO <sub>3</sub> , pH 5	2.2	0.32	30
A8	3000 ppm H <sub>3</sub> BO <sub>3</sub> , pH 9.5	0.64	1.1	30

<sup>a</sup>Adapted from Reference 10.

<sup>b</sup>Fresh room temperature solution.

<sup>c</sup>Corrected for other removal mechanisms, e.g., reaction wall; corrections were <10%;  $t_{1/2} = \ln 2/\lambda$ .

<sup>d</sup>Ratio of airborne iodine concentration direct before and after spray operation.

Two additional experimental sources of information on the effectiveness of spray additives were located. These experiments were performed in the PISCO 10 facility<sup>12</sup> and by JAERI.<sup>13</sup>

The PISCO 10 model containment had an internal volume of 95 m<sup>3</sup>. Twelve experimental runs were carried out. Service water and 1% sodium thiosulfate (Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub>) solutions were tested. Service water can certainly be considered additive free. The authors of Reference 12 concluded that the removal rates were similar for both service water and 1% thiosulfate solution. A summary of these results is given in Table 2.

Nishio et al.<sup>13</sup> quoted results obtained at the Japan Atomic Energy Research Institute (JAERI). However, no citation was given and the original manuscript could not be located. Based upon the description given by Nishio, the experiments were carried out in a 708 m<sup>3</sup> cylindrical vessel. Two experiments, BIS-1 and BIS-2, were done under conditions that simulated a BWR LOCA. One experiment (PIS-9) was performed for conditions which simulated a PWR LOCA. Summary results are given in Table 3. The range of values observed in the JAERI tests is in fair agreement with previous studies. However, direct comparison in all cases is not possible since the spray flow rates are atypical of the regime anticipated in domestic PWRs. However, these results are applicable to BWR containments.

Table 2 Summary of PISCO First Spray Results<sup>a</sup>

Run	Solution Composition <sup>b</sup>	$t_{1/2}$ (min)	$\lambda$ (min <sup>-1</sup> )
101	1% Na <sub>2</sub> S <sub>2</sub> O <sub>3</sub> , 62°C	1.3	0.53
106	1% Na <sub>2</sub> S <sub>2</sub> O <sub>3</sub> , 30°C	1.0	0.69
107	1% Na <sub>2</sub> S <sub>2</sub> O <sub>3</sub> , 18°C	0.3	2.3
108	1% Na <sub>2</sub> S <sub>2</sub> O <sub>3</sub> , 14°C	1.5	0.46
103	service water, 86°C	3.0	0.23
102	service water, 80°C	0.3	2.3
104	service water, 16°C	2.0	0.35
109	service water, 26°C	3.0	0.23
110	service water, 34°C	2.0	0.35
111	service water, 35°C	3.5	0.20
112	service water, 33°C	1.2	0.58

<sup>a</sup>Adapted from Reference 12. DF's not calculated, since spray duration varied widely.

<sup>b</sup>Solution composition, temperature of spray.

Table 3 Summary Results of JAERI Tests<sup>a</sup>

Run	Solution Composition <sup>b</sup>	$t_{1/2}$ (min)	$\lambda$ (min <sup>-1</sup> )
BIS-1	pure water, 70°C	2.9	0.24
BIS-2	pure water, 70°C	6.5	0.11
PIS-9	1.4% H <sub>3</sub> BO <sub>3</sub> , pH 9.6, 40°C	0.50	1.4

<sup>a</sup>Adapted from Reference 13. DF not calculated; insufficient data.

<sup>b</sup>Solution composition, temperature of supplied spray.

#### 4. SPRAY MODELS

##### 4.1 Analytical Procedure

The original treatment of the removal of iodine from the containment atmosphere is presented in Reference 10. The summary description given here is based upon the discussion given by Albert.<sup>11</sup>

The removal of iodine from the containment atmosphere is traditionally modeled as a first order process:

$$\frac{dC}{dt} = -\lambda C, \quad (1)$$

where  $C$  is the airborne iodine concentration,  $t$  is the time, and  $\lambda$  is the removal rate constant. Integration of Equation (1) yields

$$C = C_0 e^{-\lambda t} \quad (2)$$

with  $C_0$  is the initial concentration of iodine. The removal coefficient has been defined<sup>10,14</sup> as

$$\lambda = FP\epsilon/V, \quad (3)$$

where  $F$  is the volumetric flow rate,  $P$  is the partition coefficient of iodine between the spray liquid and the gas phase,  $V$  is the sprayed containment volume, and  $\epsilon$  is the removal efficiency. The removal efficiency has been theoretically defined for several mass transport limiting processes. These expressions are for the stagnant drop model

$$\epsilon = 1 - \sum (6 N_{sh}^2 e^{-a_n^2} / a_n^2 [a_n^2 + N_{sh}(N_{sh}-1)]) \quad (4)$$

for the stagnant film model

$$\epsilon = 1 - \exp [(-6k_g t_e) / d(P + k_g/k_1^0)], \quad (5)$$

for the well mixed drop model

$$\epsilon = 1 - \exp [(-6 k_g t_e) / dP], \quad (6)$$



and for the gas phase controlling resistance model

$$\epsilon = 6 k_g h/P v_g d. \quad (7)$$

In these equations,

$k_g$  = gas film mass transfer coefficient,

$k_l^0$  = liquid film mass transfer coefficient (no reaction),

$t_e$  = drop exposure time,

$d$  = drop diameter,

$N_{sh} = k_g d + (2 P D_l)$

$D_l$  = liquid phase diffusion coefficient,

$\phi = 4 D_l t_e/d^2$ ,

$a_n$  = nth root of the equation  $a_n \cot(a_n) + N_{sh} - 1 = 0$

$h$  = drop fall height, and

$v_g$  = terminal drop velocity.

In the initial spray phase, when the solution is fresh and therefore contains no dissolved iodine, it might be expected that the mass transfer to the drop is limited by gas phase transfer. Combining Equations 3 and 7 one obtains

$$\lambda = \frac{6k_g F h}{V v_g d} = f(P). \quad (8)$$

Note that if gas-phase transfer is the rate limiting step, then the first order removal coefficient is predicted to be independent of solution composition. Similar first spray removal rate coefficients for various solutions have been observed (see previous section), although the equilibrium partition coefficient certainly does vary with the composition of the solution.<sup>15</sup> Another variable, which will find use in the next section, is the total drop surface area,  $A$ , created per unit time per unit sprayed containment volume.

$$A = \frac{S_T}{V} = \frac{n S_d}{V} = \frac{1}{V} \left( \frac{6F}{\pi d^3} \right) (\pi d^2) = \frac{6F}{Vd} \quad (9)$$

A is simply the total surface area ( $S_T = nS_d$ ) created by drops of mean diameter,  $d$ , at the flow rate,  $F$ , and divided by  $V$ , the sprayed containment volume. Substituting Equation 9 into 8 yields:

$$\lambda = k_g h A/v_g \quad (10)$$

#### 4.2 Reevaluation of Existing Data

In an earlier section, available data on the iodine removal effectiveness was reviewed. Care was taken to select and display only data for fresh spray solutions. In both the CSE and PISCO experiments, multiple spray periods, including recirculation, were investigated. These latter results are expected to reflect the effects of increased iodine concentration in the drop, as well as the heretofore mentioned complications of wall desorption and inhomogeneous mixing. However, when the spray solution was fresh, all solutions appeared to effectively reduce the airborne iodine concentration, regardless of the presence or absence of an active spray additive. To demonstrate this contention, and account for the major variables causing variation, selected first spray data from the CSE, PISCO, and JAERI tests are displayed in Figure 3. In this figure, the first order removal rate coefficient is plotted against the normalized total new drop surface,  $A$ . Also displayed are the range of flow regimes of several typical PWR's. These were estimated from information obtained from the Surry FSAR<sup>16</sup> and information provided in References 17 and 18. The upper limit of a range represents both spray header systems are operating, while the lower limit represents operation of only one of the two redundant spray systems. Approximately 3/4 of the PISCO data and both JAERI BWR test data are not plotted, as they would lie well beyond the anticipated range of  $A$  for domestic PWR's.

Although the plotted data exhibit some scatter, a generally good correlation is found. Hence, when spray solution is fresh, the removal of iodine from the containment atmosphere is dominated by gas phase mass transport and is effectively independent of the equilibrium iodine partition coefficient of the solution, and primarily controlled by the amount of available surface to which iodine may be transported. At a first level of approximation, the good correlation of  $\lambda$  and  $A$  observed indicates that the combination of terms not explicitly examined in Equation 10 is effectively constant or slowly varying over the range of experimental conditions investigated. It should also be noted that 3/4 of the PISCO and JAERI data not plotted, that data which was taken in a flow regime atypical of domestic PWR's, does not correlate well with  $A$  alone. There are several potential reasons why this occurs, however, these have not been examined since it is felt that the experimental conditions are basically atypical of those conditions associated with domestic commercial PWR's.

Values of  $k_g$ , computed from Equation (10) and for the experimental data displayed in Figure 3, are given in Table 4. For comparison, estimates of  $k_g$  and  $\lambda$ , based upon a well known correlation for heat transfer to a single drop, are also given. Assuming the minimum observed experimental  $k_g$ , 3 m/min, maximum fall height<sup>16,18</sup> and that both spray headers are operating, first order removal coefficients of 0.8 min<sup>-1</sup> and 2.0 min<sup>-1</sup> are estimated from Equation (10) for the Surry and Zion plants, respectively. If it is assumed that structures, e.g., the reactor pressure vessel and steam generators reduce the effective drop fall height by 50% to 60% of the maximum, then estimated  $\lambda$ 's of 0.4 min<sup>-1</sup> and 1.3 min<sup>-1</sup> are obtained for these plants, respectively.

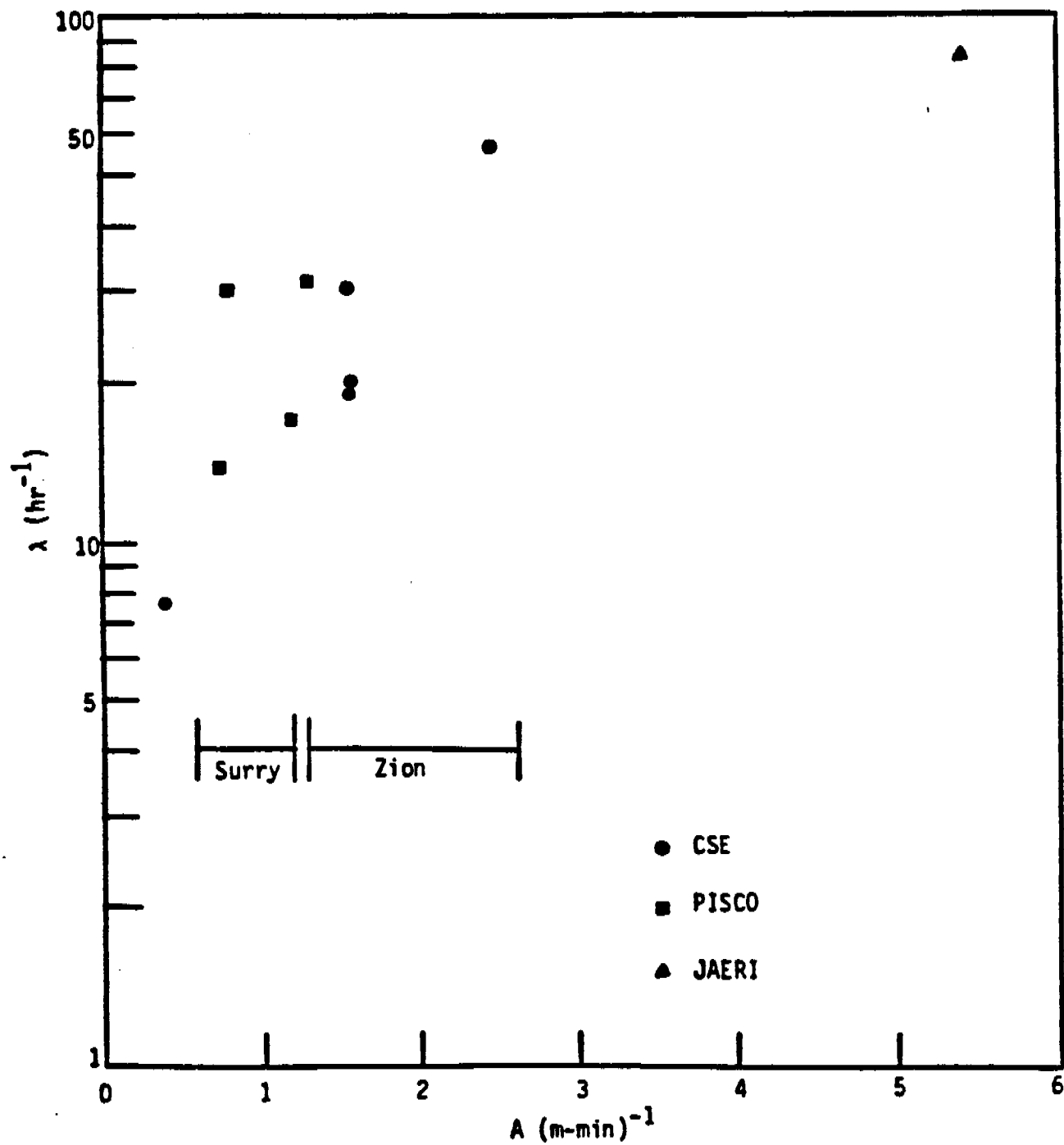


Figure 3 Removal constant versus normalized, new drop surface area; fresh spray data.

## 5. DISCUSSION AND CONCLUSION

In previous sections of this report, the current best estimate description of source term characteristics was summarized and available data on the effectiveness of spray as a fission product removal system were presented, reviewed, and reevaluated. CSS can effectively reduce airborne concentrations of aerosols which current methods of source term estimation predict to overwhelmingly dominate fission product releases. The inert noble gas releases are unaffected by sprays. Aerosol removal by the sprays is a physical process and this process is not altered or aided by the presence of chemical additives. Other natural processes, modeled by NAUA-4, can also reduce airborne aerosol concentration. The relative benefit accrued is closely related with the time available prior to containment failure for these natural processes to act. Data presented in Chapter 2 indicated that the combined effect of these processes can reduce airborne CsI aerosol concentrations by a factor 50 to  $10^5$ , depending upon the specific reactor and accident sequence being examined.

Current NRC sponsored analytical modeling of severe accidents, i.e., the STCP, does not predict the emission of gaseous iodine in the anticipated accident environment associated with commercial LWR's. This is based upon an examination of the TMI-2 accident, and subsequent thermochemical analyses. The actual chemical form of iodine is still subject to a degree of uncertainty. Evolving experimental and analytical evidence<sup>7,19,20</sup> indicates boron may be chemically associated with Cs; hence, boron may be in competition with iodine and potentially liberate iodine in another form, possibly gaseous. Preliminary experimental results<sup>21,22</sup> also suggest reaction of CsOH and CsI with the stainless steel surfaces of the RCS, with the reported emission of gaseous iodine in some cases. The reproducibility of these experiments are currently being investigated. In addition, some experimental evidence<sup>23</sup> has been obtained that indicates the conversion of CsI aerosol to gaseous iodine during hydrogen burns. These observations certainly reinforce the diversity of material interactions and phenomena that can occur and give rise to uncertainty. Additional research is in progress and is required to resolve this uncertainty. However, regardless of the extent of gaseous iodine conversion, the washout by an operational CSS would occur and the data presented in Chapters 3 and 4 clearly suggest that it is effective. Moreover, the fresh spray data suggest efficient iodine removal regardless of the presence of additives during the initial injection phase. This is not to say that NaOH is not ultimately required to increase the absorption capacity of the spray solution and mitigate iodine reevolution from the reactor sump. On the contrary, sufficient evidence exists to warrant pH control in the long term. The regulatory option to be reconsidered is whether or not the presence of NaOH is required during the initial injection phase of CSS operation. Two items affect this decision. One, is the effectiveness of fresh spray solution, and the other is that since the CSS is activated on high containment pressure, it is quite possible that the CSS will have switched from the injection to the recirculation phase prior to the release of any fission product activity.

A potential alternative is to add pH control directly to the reactor sump rather than in the initial injection supply of the CSS. Additionally, it would be attractive to initiate pH control on some feedback directly related to the release of activity rather than on high containment pressure. This

would have the obvious advantage of not introducing the additive(s) until required. A secondary benefit should the reactor incident be terminated without the release of FP activity, would be a simplified cleanup recovery.

Table 4 Comparison of Experimental and Estimated Mass Transfer Constants

Run	$A(m-min)^{-1}$	$t_e(min)$	$\lambda(min^{-1})$	$\lambda_{est}$	$k_g \frac{m}{min}$	$k_g est$
A3	0.40 <sup>a</sup>	0.050 <sup>b</sup>	0.13	0.14 <sup>c</sup>	6.5 <sup>d</sup>	6.7 <sup>e</sup>
A4	1.55	0.050	0.50	0.52	6.5	6.7
A6	1.55	0.050	0.33	0.65	4.3	8.4
A7	1.55	0.050	0.32	0.65	4.2	8.4
A8	2.45	0.083	1.1	1.8	5.4	8.9
PIS-9	5.36	0.092	1.4	3.7	2.9	7.4
109	0.74	0.029	0.23	0.15	11	6.7
110	1.35	0.020	0.35	0.18	13	6.9
111	1.19	0.020	0.20	0.16	8.6	6.9
112	0.79	0.029	0.58	0.16	25	6.9

(a) Calculated from reported data for F, V and d.

(b)  $t_e = h/v_g$ . Velocity calculated from

$$v_g = (4(\rho_a - \rho_g) g d / 3 \rho_g \xi)^{1/2},$$

where  $\xi = 18.5/Re^{0.6}$ , and

$\rho_a$  is the drop density,  $\rho_g$  is the gas density, g is the gravitation constant,  $\xi$  is the drag coefficient and Re is the Reynolds number; Handbook of Multiphase Systems, G. Hetsroni, Hemisphere Pub. Corp., New York, NY, 1982.

(c)  $\lambda_{est} = A t_e k_g est$ .

(d)  $k_g$  back calculated from experimental data.

(e)  $k_g est$  estimated from the correlation of Ranz and Marshall, Chem. Eng. Prog., 48, 1952.

$$k_g = D/d (2 + 0.6(\rho_d v_g / \mu)^{1/2} (\mu / \rho D)^{1/3}),$$

where D is the diffusivity of  $I_2$ . All gas phase variables are for air.

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APPENDIX A

Computerized searches of the literature were performed to obtain pertinent citations on the subject containment spray and spray additives for PWRs. The following databases were queried:

EDB (Energy Data Base, DOE/RECON, 1976-present)

The search resulted in 171 citations; of which 18 were selected and acquired in support of the research project; and

NSA (Nuclear Science Abstracts, DOE/RECON, 1967-1976)

The search resulted in 8 citations.

The search strategy was:

PWR

and

[containment spray systems

or

(containment system) and (atmospheres)

or

((sprays or droplets or particles or iodine removal or particulates or additive(s)) and (containment systems))].

APPENDIX B

The following chapter was reproduced from NUREG/CR-4081. It provides a brief description of the CSE and a concise summary of the CSE results and their precision.

## 7. EXPERIMENTAL DATA

The experimental data used to compare with the results of the spray model are from the Containment System Experiments (CSE).<sup>3</sup> Experimental runs A-3, A-4, A-6, A-7, and A-8 of this series are large scale spray system tests to determine the effectiveness of a spray system for removing airborne fission products. The results of these tests are reported in terms of the gas phase elemental iodine concentration versus time and also in terms of the liquid phase elemental iodine concentration versus time. The parameters for the spray experiments are the spray flux, the drop size, the gas phase temperature, pressure, and humidity, and the liquid spray composition. The physical dimensions of the CSE vessel are listed in Table 3 and are shown in Figure 44. Since

Table 3. Physical conditions common to all spray experiments (Hillard<sup>3</sup>)

Volume above deck including drywell	21,005 ft <sup>3</sup>	595 m <sup>3</sup>
Surface area above deck including drywell	6,140 ft <sup>2</sup>	569 m <sup>2</sup>
Surface area/volume	0.293 ft <sup>-1</sup>	0.958 m <sup>-1</sup>
Cross section area, main vessel	490 ft <sup>2</sup>	45.5 m <sup>2</sup>
Volume, middle room	2,089 ft <sup>3</sup>	59 m <sup>3</sup>
Surface area, middle room	1,363 ft <sup>2</sup>	127 m <sup>2</sup>
Volume, lower room	3,384 ft <sup>3</sup>	96 m <sup>3</sup>
Surface area, lower room	2,057 ft <sup>2</sup>	191 m <sup>2</sup>
Total volume of all rooms	26,477 ft <sup>3</sup>	751 m <sup>3</sup>
Total surface area, all rooms	9,560 ft <sup>2</sup>	888 m <sup>2</sup>
Drop fall height to deck	33.8 ft	10.3 m
Drop fall height to drywell bottom	50.5 ft	15.4 m
Surface coating	All interior surfaces coated with phenolic paint. <sup>a</sup>	
Thermal insulation	All exterior surfaces covered with 1-in. fiberglass insulation. <sup>b</sup>	

<sup>a</sup>Two coats Phenoline 302 over one coat Phenoline 300 primer. The Carboline Co., St. Louis, Missouri.

<sup>b</sup> $k = 0.027 \text{ Btu}/(\text{hr})(\text{ft}^2)(^\circ\text{F}/\text{ft})$  at 200°F, Type PF-615, Owens-Corning Fiberglass Corp.

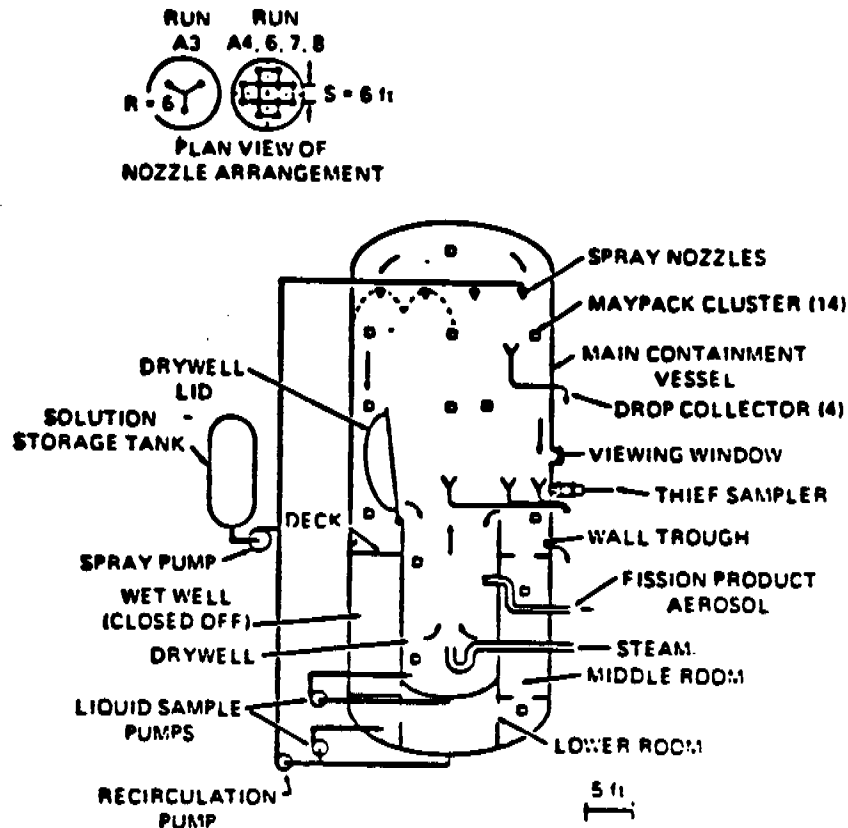


Fig. 44. Schematic diagram of containment arrangement used in CSE spray tests (Hillard<sup>3</sup>).

these tests were made in realistic and not idealized equipment and conditions, the liquid and gas flow patterns are complex and not well characterized. The results from the new spray model will be compared with these results, but no better than approximate agreement can be expected. This data, however, can still provide a means for useful and meaningful evaluation of the spray model.

The CSE vessel is a large scale vessel (see Table 3 and Figure 44). The overall dimensions of the vessel are 20.34 meters high and a diameter of 7.62 meters. The vessel has a drop fall height of 15.4 meters. The overall volume of the vessel is 751 cubic meters.

The tests varied the temperature, pressure, pH of the drop, spray nozzle configuration and drop size. The conditions for run A-3 are a temperature of 298K, 1 atmosphere of pressure, pH of 9.5 and a drop diameter of 1210 microns. For all of the tests, the spray solution temperature was at 25°C, and the solutions were all buffered. For run A-4, the conditions were the same as for A-3 except for a higher spray flow rate and a different spray nozzle configuration. Run A-6 increased the temperature of the gas to 397K and the pressure to 3 atmospheres. Run A-7 changed the pH to 5, lowered the temperature to 394K and raised the

pressure to 3.4 atmospheres. Run A-8 changed the drop diameter to 770 microns. See Figure 44 for spray nozzle arrangements, Table 4 for spray nozzles used, Table 5 for the atmospheric conditions, Table 6 for the spray flow rates and solutions used in the tests and Table 7 for the timing of the spray periods.

The experimental procedure for the molecular iodine spray absorption tests involved first heating the containment vessel with steam until the specified temperature was reached. A flask containing molecular iodine traced with 1 curie of iodine-131 was heated electrically. Air was passed over the flask to release molecular iodine. Samples were taken prior to turning on the sprays to determine how molecular iodine behaves without sprays. After the first spray period

Table 4. Nozzles used in CSE spray experiments  
(Hillard<sup>3</sup>)

<u>Runs A3, 4, 6, 7</u>		
Nozzle type:	Spraying Systems Co. 3/4 — 7G3	
Nozzle characteristics:	Fog type, full cone	
	<u>A3</u>	<u>A4, 6, 7</u>
Number	3	12
Layout	Triangular	Square grille
Spacing	10 ft 5 in. apart	6 ft apart
Pressure	40 psid	40 psid
Rated flow	4 gpm	4 gpm
MMD	1210 $\mu$	1210 $\mu$
$\sigma_g$	1.5	1.5
<u>Run A8</u>		
Nozzle type:	Spray Systems Co. 3/8 A 20	
Nozzle characteristics:	Fine atomization, hollow cone	
Number used	12	
Layout	Square grid	
Spacing	6 ft apart	
Pressure	40 psid	
Rated flow	4 gpm	
MMD	770 $\mu$	
$\sigma_g$	1.5	



Table 5. Atmospheric conditions in CSE spray experiments (Hillard<sup>3</sup>)

	Run A3	Run A4	Run A6	Run A7	Run A8
Containment vessel insulated	No	No	Yes	Yes	Yes
Forced air circulation <sup>a</sup>	Yes	Yes	No	No	No
Start of 1st spray					
Vapor temperature, °F <sup>b</sup>	77	77	255	248.7	250
Pressure, psia	14.6	14.6	44.2	50.0	50.7
Relative humidity, %	70	88	100	100	100
End of 1st spray	--				
Vapor temperature, °F <sup>b</sup>	77	77	229	234.5	243
Pressure, psia	14.6	14.6	38.6	44.4	48.2
Start of 2nd spray					
Vapor temperature, °F <sup>b</sup>	77	77	237	240	243
Pressure, psia	14.6	14.6	40.8	46.0	243
End of 2nd spray					
Vapor temperature, °F <sup>b</sup>	77	77	202	203	188
Pressure, psia	14.6	14.6	29.5	36	34.1
Start of 3rd spray					
Vapor temperature, °F <sup>b</sup>	77	77	233	230	218
Pressure, psia	14.6	14.6	40.7	41.8	32.2
Start of 4th spray					
Vapor temperature, °F <sup>b</sup>	c	c	c	232	247
Pressure, psia	c	c	c	42.4	52.4
End of 4th spray					
Vapor temperature, °F <sup>b</sup>	c	c	c	192	175
Pressure, psia	c	c	c	32.7	32.4

<sup>a</sup>Fan without duct located in bottom of drywell.  
2400 ft<sup>3</sup>/min discharge.

<sup>b</sup>Average of 5 thermocouples located at various elevations and radii.

<sup>c</sup>No fourth spray.

Table 6. Spray flow rates and solutions used in CSE experiments (Hillard<sup>3</sup>)

	Run A3	Run A4	Run A6	Run A7	Run A8
<b>1st spray</b>					
Total flow rate, gpm	12.8	49	49	49	50
Volume sprayed, gal	128	490	490	490	150
Spraying pressure, psid	40	40	40	40	40
Solution	a	a	b	c	b
<b>2nd spray</b>					
Total flow rate, gpm	12.8	49	50	48.5	50
Volume sprayed, gal	385	1480	1500	1455	1850
Spraying pressure, psid	40	40	40	40	40
Solution	a	a	b	c	b
<b>3rd spray</b>					
Total flow rate, gpm	12.5	42	16	45.5	47
Volume sprayed, gal	735	1890	960	2730	2820
Spraying pressure, psid	40	29	4	36.5	36.5
Solution	d	e	e	e	e
<b>4th spray</b>					
Total flow rate, gpm	g	g	g	48.6	50.4
Volume sprayed, gal	g	g	g	2428	2520
Spraying pressure, psid	g	g	g	40	40
Solution	g	g	g	f	f

<sup>a</sup>Fresh, room temperature. 525 ppm boron as H<sub>3</sub>BO<sub>3</sub> in demineralized water. NaOH added to pH of 9.5.

<sup>b</sup>Fresh, room temperature. 3000 ppm boron as H<sub>3</sub>BO<sub>3</sub> in demineralized water. NaOH added to pH of 9.5.

<sup>c</sup>Fresh, room temperature. 3000 ppm boron as H<sub>3</sub>BO<sub>3</sub> in demineralized water. No NaOH added. pH 5.

<sup>d</sup>Fresh, room temperature demineralized water.

<sup>e</sup>Solution in main vessel sump recirculated. No heat exchanger used.

<sup>f</sup>Fresh, room temperature. 1 wt% Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub>, 3000 ppm boron as H<sub>3</sub>BO<sub>3</sub> in demineralized water. NaOH added to pH 9.4.

<sup>g</sup>No fourth spray.



Table 7. Timing of spray periods  
(Hillard<sup>3</sup>)

	Time after start of iodine release, min				
	Run A3	Run A4	Run A6	Run A7	Run A8
<b>First spray</b>					
Start	40	40.5	30	30	30
Stop	50	50.5	40	40	33
Duration	10	10	10	10	3
<b>Second spray</b>					
Start	140	140	80	80	80
Stop	170	170	110	110	117
Duration	30	30	30	30	37
<b>Third spray</b>					
Start	1473	1205	1565	1323	200
Stop	1533	1250	1525	1383	260
Duration	60	45	60	60	60
<b>Fourth spray</b>					
Start	a	a	a	1443	1350
Stop	a	a	a	1493	1400
Duration	a	a	a	50	50

<sup>a</sup>No fourth spray.

of each run was started, many samples were taken from the gas phase, from the liquid in the sump, from the wall trough and from the spray drop collectors. When the first spray period was ended, more samples were taken to determine how molecular iodine acts. A second, third, and sometimes a fourth spray period were used with many samples taken from the gas and liquid phases. The gas phase concentrations were determined by Maypack samplers (see Figure 45), and the liquid phase concentrations were determined by measuring the amount of iodine-131 tracer present. For more information see Reference 3.

Results of these experimental tests are shown in Figures 46 through 55 and Table 8. Table 8 shows the material balance of iodine for all of the experimental runs. It should be noted in this table that between 25.65% and 57.58% of the iodine delivered to the containment vessel is unaccounted for and is assumed to be deposited on surfaces. Figures 46 through 50 show the concentration of elemental iodine in the gas phase as a function of time. The data is reported in terms of the half life

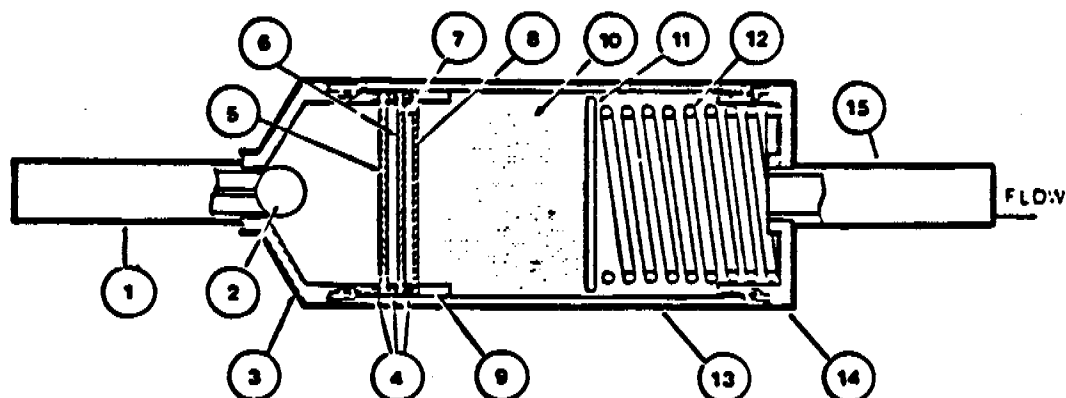
Table B. Iodine material balances  
(Willard<sup>3</sup>)

Location	Run A1		Run A5		Run A6		Run A7		Run A8	
	Grams	% <sup>a</sup>	Grams	% <sup>a</sup>	Grams	% <sup>a</sup>	Grams	% <sup>a</sup>	Grams	% <sup>a</sup>
<u>Aerosol Generation</u>										
Starting material	101.00	100.00	101.50	100.00	101.00	100.00	101.00	100.00	101.00	100.00
Generation apparatus	2.57	3.54	1.13	1.11	0.14	0.14	1.06	1.05	3.45	3.42
Injection line	22.32	22.09	29.32	28.99	1.49	1.47	2.05	2.03	1.62	1.60
Injection line samples	0.36	0.36	0.15	0.15	1.030	1.02	0.32	0.32	0.32	0.32
Accounted for	25.25	25.00	30.59	30.14	2.66	2.63	3.43	3.40	5.19	5.14
Delivered to containment (by difference)	75.75	75.00	70.91	69.86	98.34	97.36	97.57	96.60	95.61	94.68
	Grams	% <sup>b</sup>	Grams	% <sup>b</sup>	Grams	% <sup>b</sup>	Grams	% <sup>b</sup>	Grams	% <sup>b</sup>
<u>Containment</u>										
Delivered to containment	75.75	100.00	70.91	100.00	98.34	100.00	97.57	100.00	95.61	100.00
In liquid pools <sup>c</sup> (Prior to decontamination)	45.32	59.83	37.67	53.11	53.97	54.88	39.28	40.26	53.15	55.59
Samples	0.48	0.63	8.87	12.51	0.556	0.57	0.59	0.60	0.88	0.92
Purge to stack	0.52	0.69	0.73	1.03	0.086	0.09	0.16	0.17	0.11	0.11
Decontamination	5.90	7.78	5.46	7.70	0.820	0.83	1.36	1.39	1.44	1.50
Accounted for	52.22	68.93	52.73	74.35	55.43	56.37	41.39	42.42	55.57	58.12
In surfaces (by difference)	23.52	31.06	18.18	25.65	42.91	43.63	56.18	57.58	40.04	41.88

<sup>a</sup>Percent of starting mass.

<sup>b</sup>Percent of delivered mass.

<sup>c</sup>Includes spray solution and steam condensate.



- |                        |                             |            |
|------------------------|-----------------------------|------------|
| 1 INLET                | 6 SIX SILVER PLATED SCREENS | 11 SCREEN  |
| 2 TEFLON BALL          | 7 SILVER MEMBRANE FILTER    | 12 SPRING  |
| 3 NOSE CONE            | 8 CHARCOAL LOADED FILTER    | 13 BODY    |
| 4 TEFTON GASKET        | 9 STOP RING                 | 14 END CAP |
| 5 TWO PARTICLE FILTERS | 10 CHARCOAL BED             | 15 OUTLET  |

Fig. 45. Maypack Sampler.

of iodine, defined as

$$t_{1/2} = -\ln(1/2)/\lambda, \quad (65)$$

$$= 0.693/\lambda, \quad (66)$$

where  $\lambda$  is the removal rate constant.

The reason the data are in this form is because the old spray models (Equations 1 through 7 of Chapter I, Section 2) are in terms of the removal rate constant. Figures 51 to 55 shows the concentration of iodine in the liquid versus time. As can be seen in these figures, there is a delay in the response of the increase in the concentration of iodine in the liquid phase.

In these tests there are many processes for the removal of molecular iodine from the gas phase. In these large scale realistic tests, there are painted surfaces, non-painted surfaces, insulation, sprays, wet walls, and dry walls. All of these features can contribute to iodine sorption, and heat transfer can also have an effect on the removal rate of molecular iodine from the gas phase. Therefore, one can only hope to develop an approximate model which accounts for the major phenomena involved and considers only the removal by the sprays. If one looks at the drop data, these data are "difficult to interpret, not only because of sampling inadequacies but because the relative fractions of the various iodine forms and particle sizes were changing rapidly with time."<sup>3</sup>

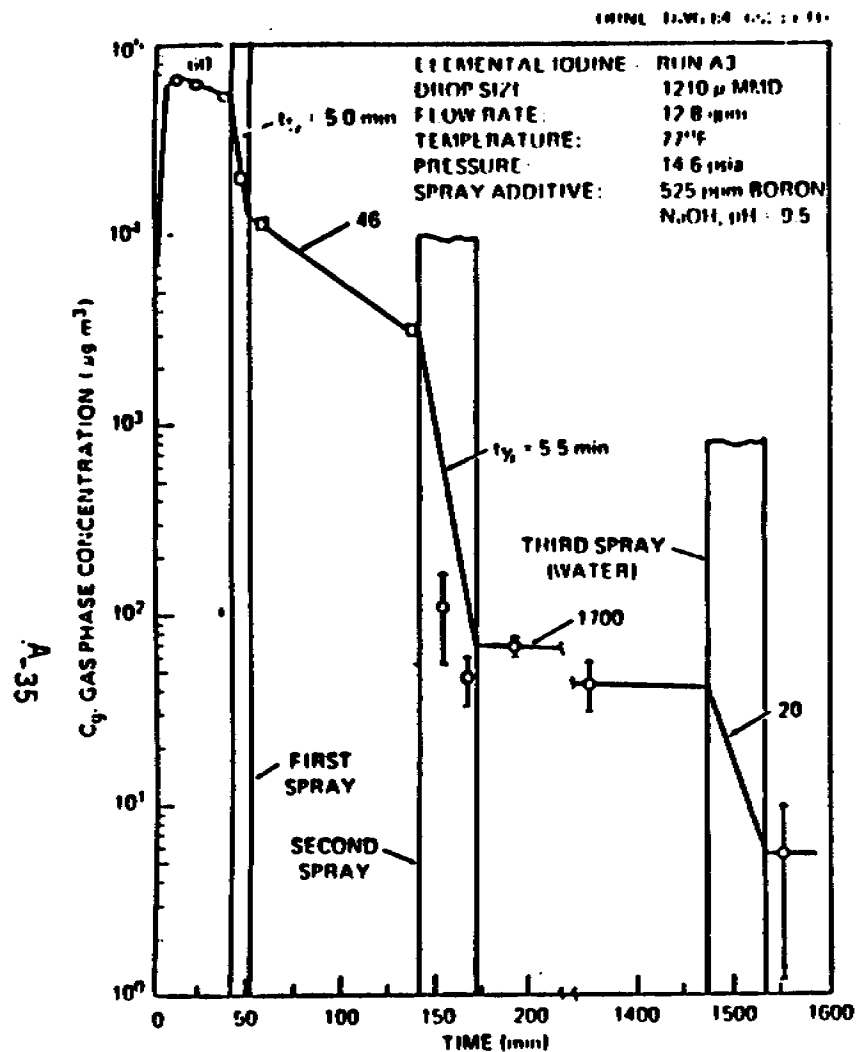


Fig. 46. Concentration of elemental iodine in the main room, run A-3 (Hilliard<sup>3</sup>).

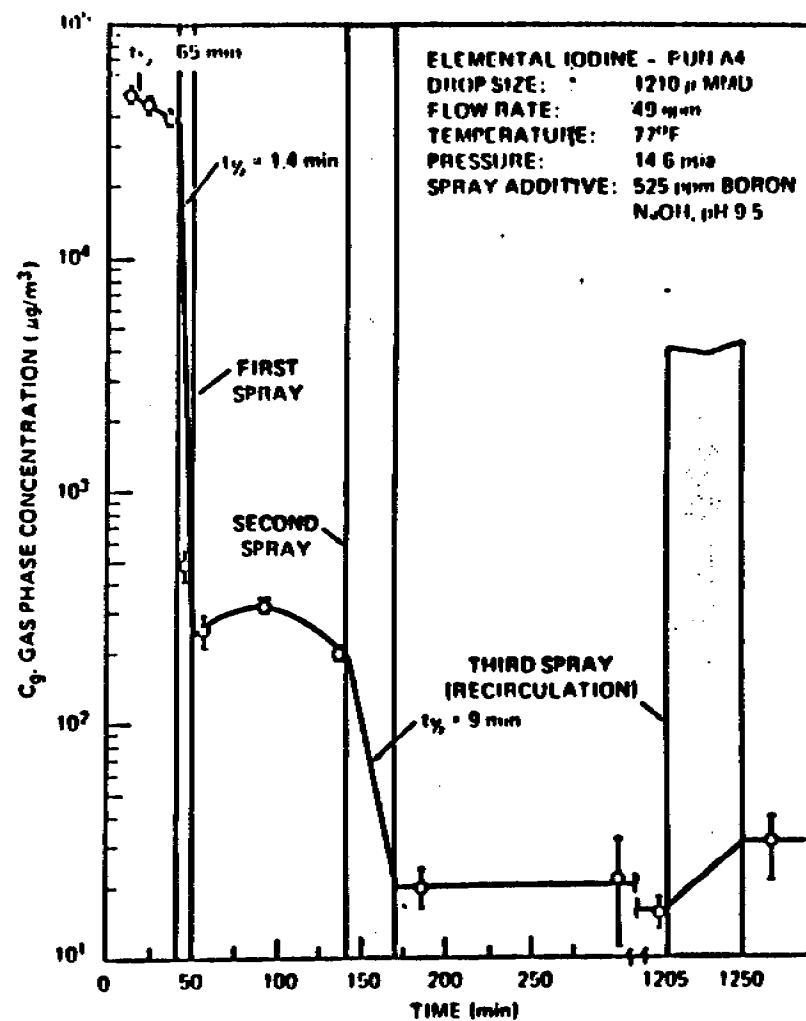


Fig. 47. Concentration of elemental iodine in the main room, run A-4 (Hilliard<sup>3</sup>).

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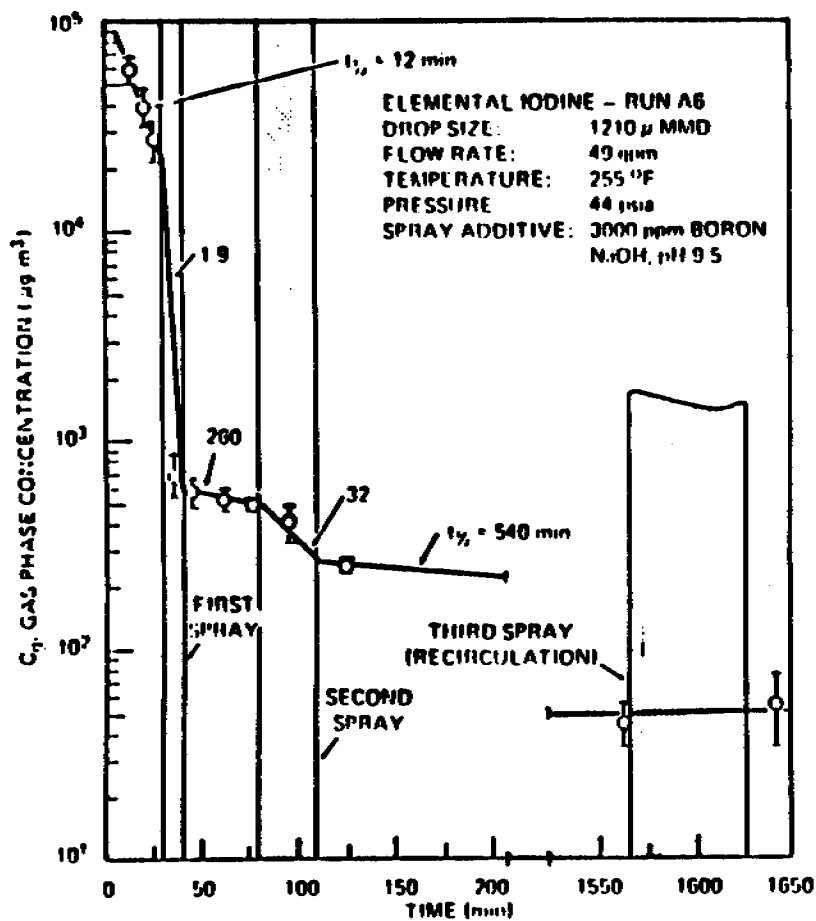


Fig. 48. Concentration of elemental iodine in the main room, run A-6 (Hillard<sup>3</sup>).

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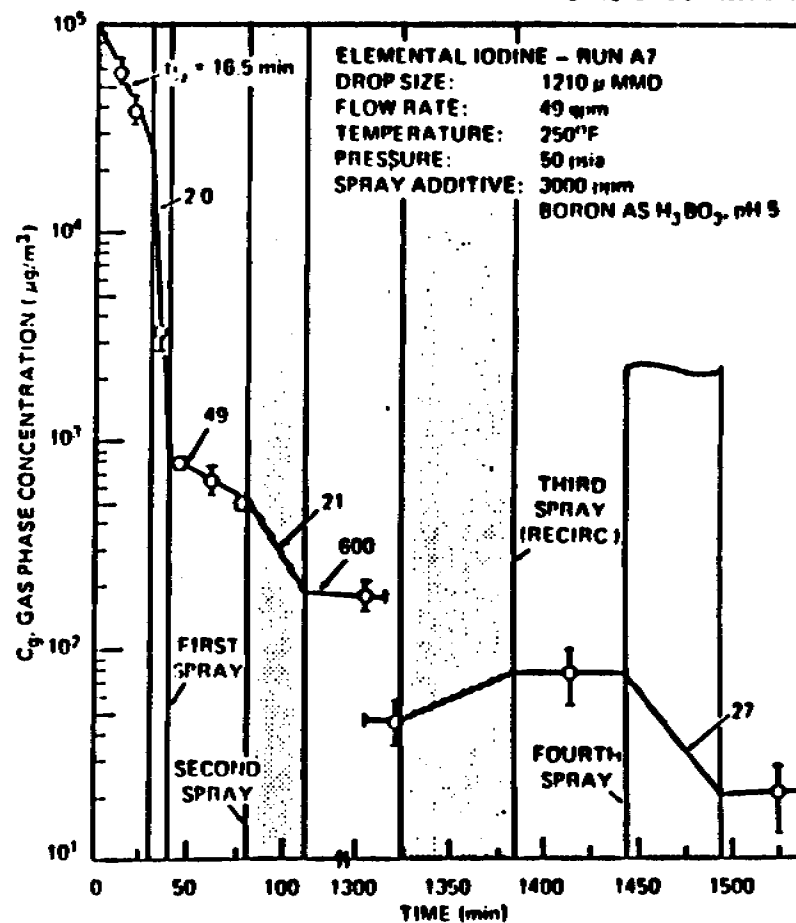


Fig. 49. Concentration of elemental iodine in the main room, run A-7 (Hillard<sup>3</sup>).

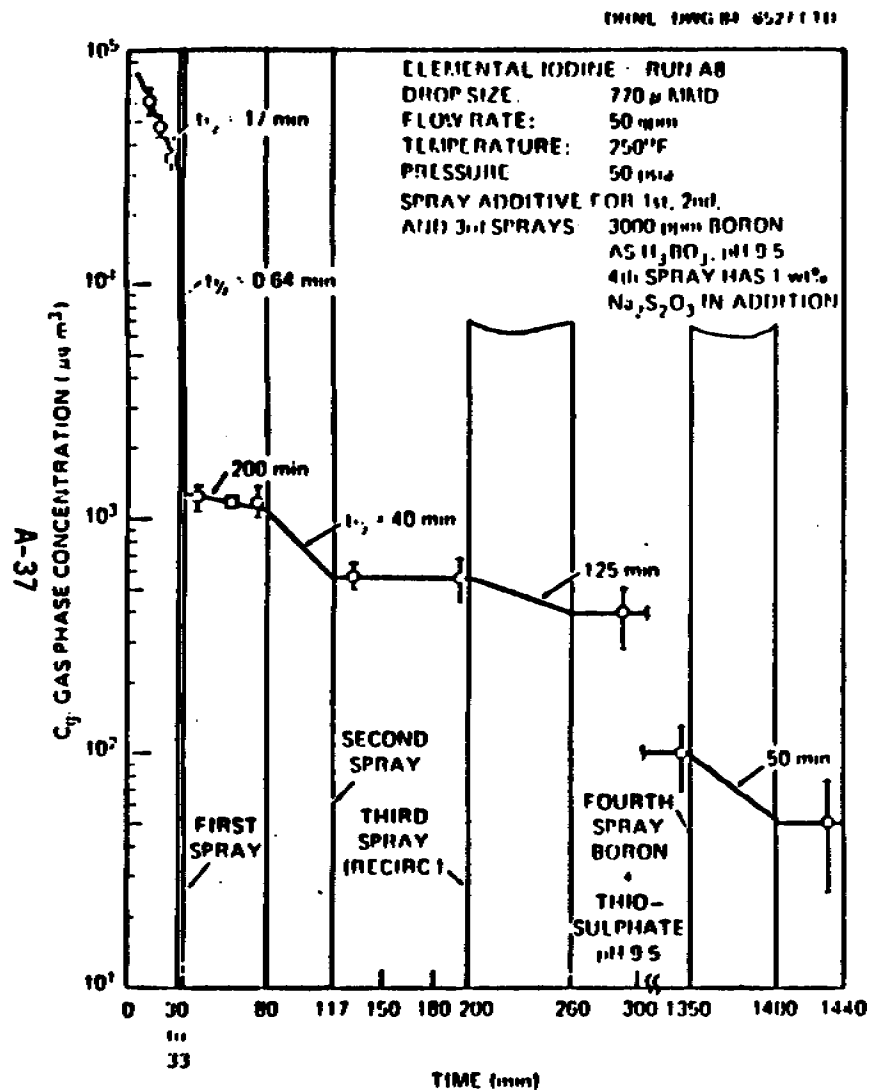


Fig. 50. Concentration of elemental iodine in the main room, run A-8 (Hillard<sup>3</sup>).

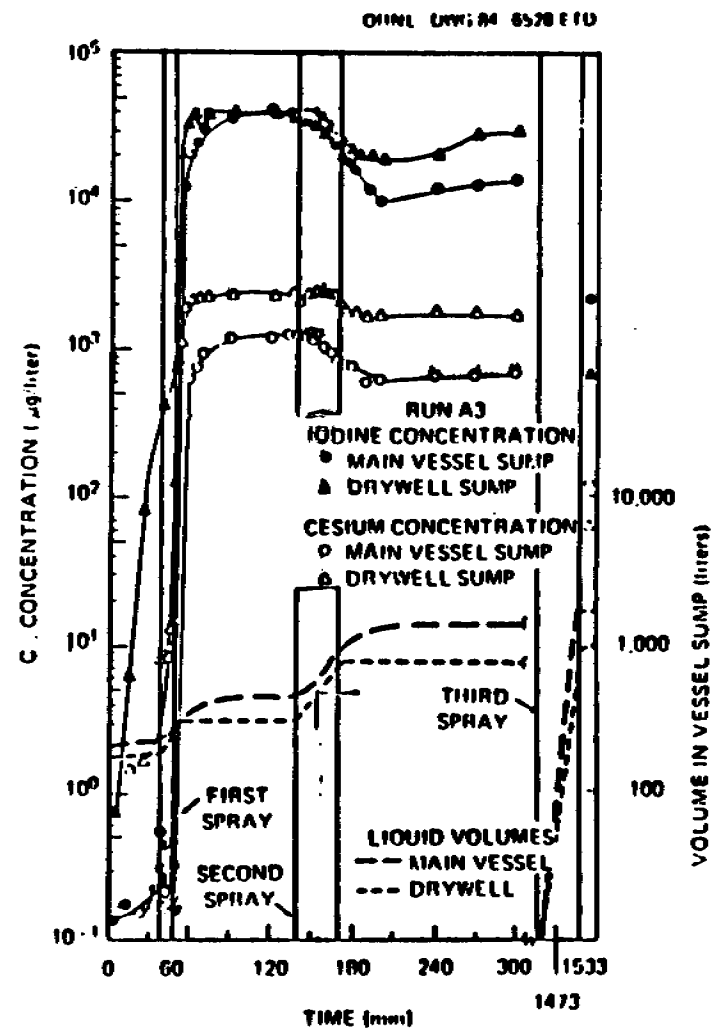


Fig. 51. Liquid volumes and concentration in vessel sumps, run A-3 (Hillard<sup>3</sup>).

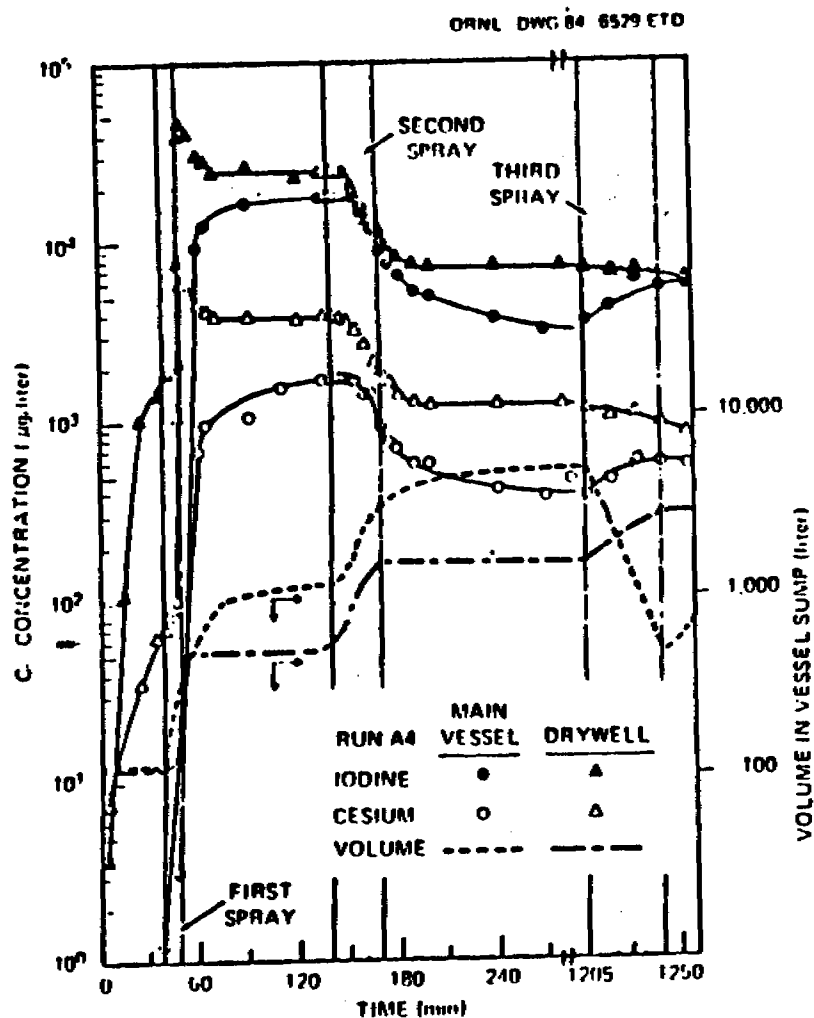


Fig. 52. Liquid volumes and concentration in vessel sumps, run A-4 (Hillard<sup>3</sup>)

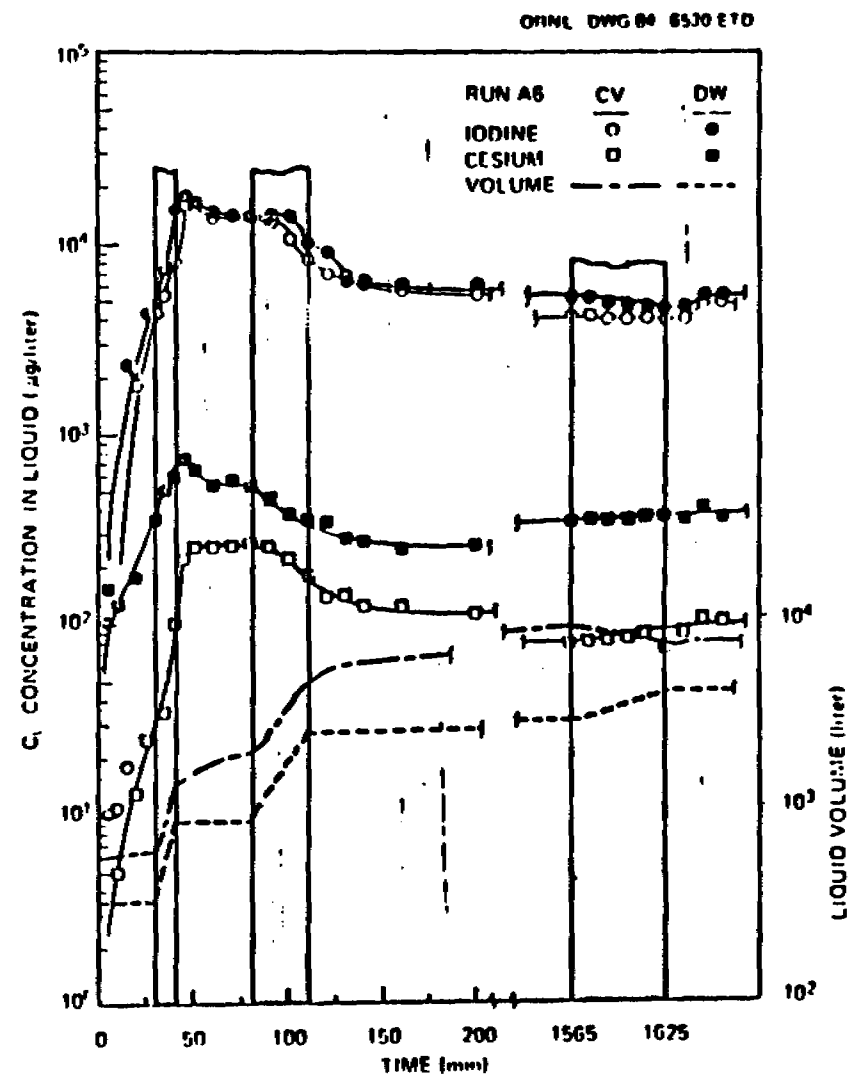


Fig. 53. Liquid volumes and concentration in vessel sumps, run A-6 (Hillard<sup>3</sup>).

(HILLIARD, R4 6531 171)

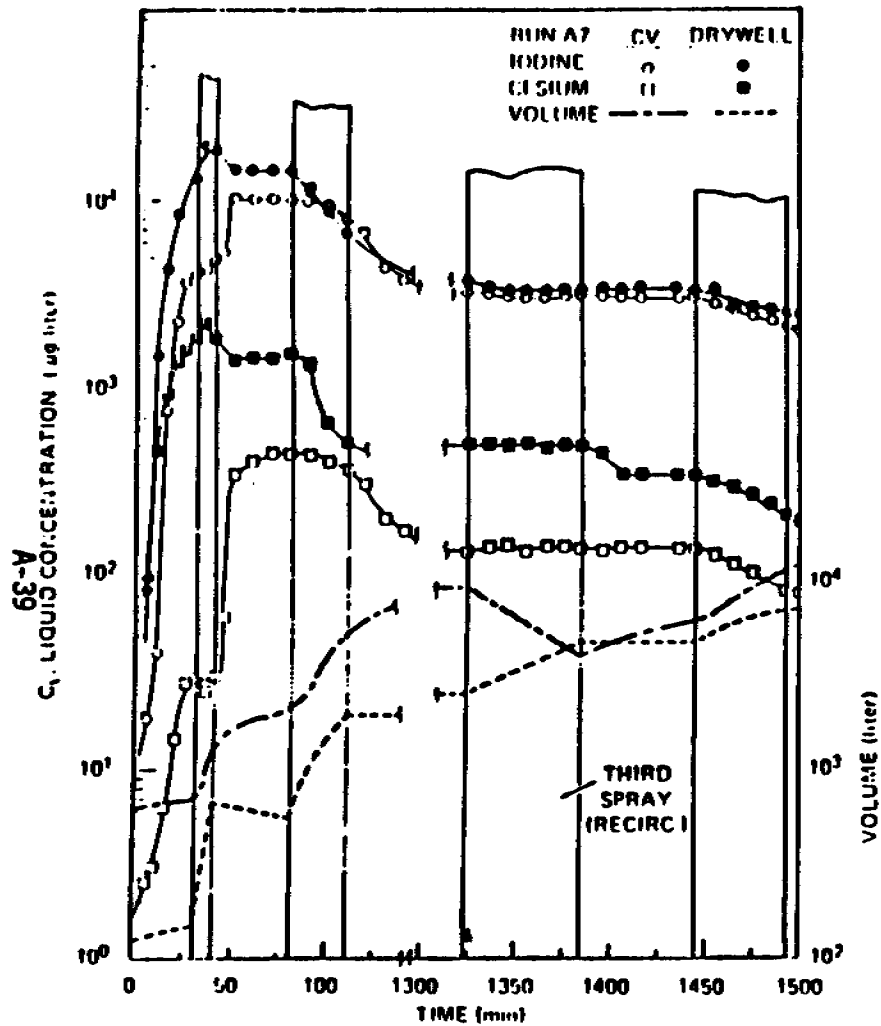


Fig. 54. Liquid volumes and concentration in vessel sumps, run A-7 (Hilliard<sup>3</sup>).

(HILLIARD, R4 6532 171)

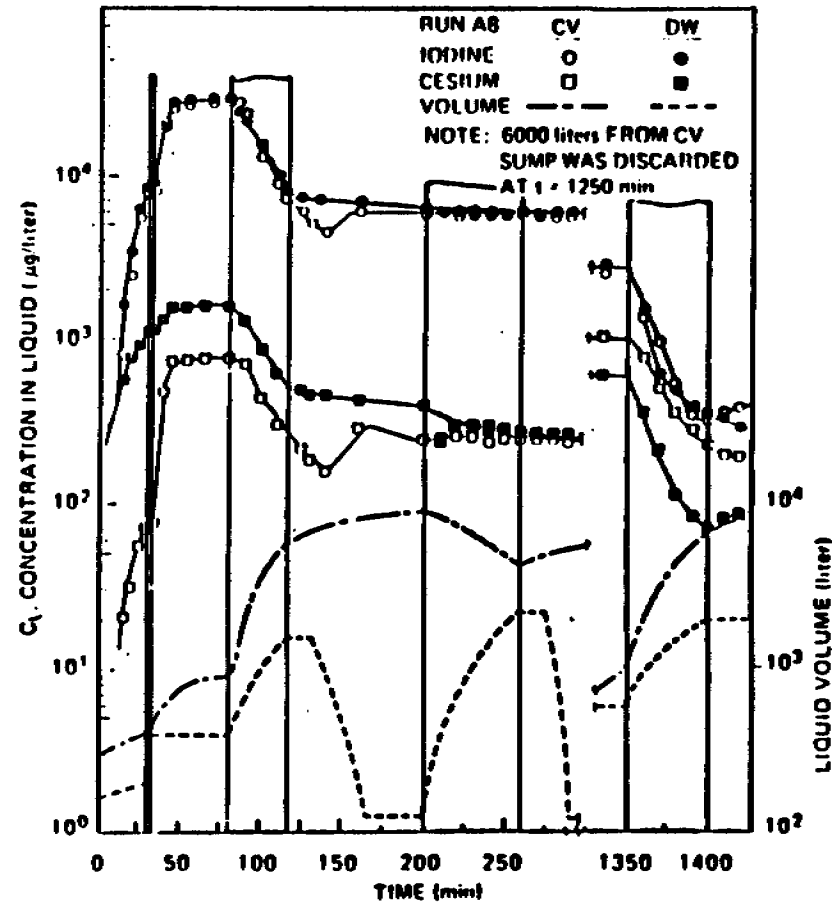


Fig. 55. Liquid volumes and concentration in vessel sumps, run A-8 (Hilliard<sup>3</sup>).



The data for the gas phase are the result of the combined effects of all of the processes for the removal of molecular iodine from the gas phase. But if all of the processes except for the sprays exerted only a small overall effect in the removal of molecular iodine from the gas phase, then these data would be acceptable from the standpoint of usefulness in determining the efficiency of the spray model. The data for liquid in the sumps should eliminate some of the sources of error because these data shows how much molecular iodine is transferred to the liquid. Nevertheless, any iodine that is on the surfaces and is not chemically held to the walls could be washed off in the sumps. Since the sprays were not started at the instant the molecular iodine was released, a significant amount of molecular iodine released into the containment may have deposited on the surfaces, and subsequently been washed off into the sumps or might have been transferred back into the gas phase later when the partial pressure of molecular iodine in the gas phase was smaller than the partial pressure of molecular iodine on the surfaces. The latter effect could result in an underestimate of the removal rate of molecular iodine.

To remove some of the possible sources of error, the comparison of experimental results to the results of the spray model will be limited to the area of the drywell. The drywell had a cross sectional area of 8.8 square meters (which is a diameter of 3.35 meters), a drop fall height of 15.4 meters, and a volume of 135.52 cubic meters. For example, in run A-3, at the start of the first spray period, the initial gas phase concentration was approximately  $5 \times 10^4$  micrograms/cubic meter ( $1.97 \times 10^{-7}$  moles/liter) and the final concentration was approximately  $1.25 \times 10^4$  micrograms/cubic meter ( $4.92 \times 10^{-8}$  moles/liter). The amount of iodine removed from the gas phase during the first spray was 5.082 grams. Also, at the start of the first spray, the concentration of the liquid in the drywell sump was approximately  $8 \times 10^2$  micrograms/liter and the initial volume was approximately 150 liters. At the end of the first spray, the concentration in the drywell sump was  $4 \times 10^4$  micrograms/liter and the volume was approximately 332 liters. The number of grams of iodine transferred to the liquid in the drywell sump was 13.15 grams. The difference between the number of grams of iodine removed from the gas phase and the number of grams of iodine transferred to the liquid phase was -8.1 grams. The resulting relative error based on the gas phase is

$$\text{error} = \left( \frac{\text{grams removed from gas} - \text{grams transferred to liquid}}{\text{grams removed from gas}} \right) \quad (67)$$

$$\times 100\% ,$$

$$\text{error} = \frac{(5.082 - 13.15)}{(5.082)} \times 100\% = -133.4\% . \quad (68)$$

Results of the other runs were similar with more iodine appearing transferred to the liquid than was removed from the gas. In fact, for many cases the error is much greater.