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EFFECT OF SPRAY FLOW RATE
ON WASHOUT OF GASES
AND PARTICLES IN THE
CONTAINMENT SYSTEMS EXPERIMENT

July 1971

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EFFECT OF SPRAY FLOW RATE
ON WASHOUT OF GASES AND PARTICLES
IN THE CONTAINMENT SYSTEMS EXPERIMENT

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July 1971

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ABSTRACT

The results of a large-scale containment spray experiment, Run A9, are reported in detail. This experiment was performed in the 26,500 cubic foot Containment Systems Experiment (CSE) vessel in a steam-air environment. The spray flux was 0.3 gal/min per ft² which is higher than that of current PWR containment system designs. All of the fission product simulant materials were washed out very rapidly with the exception of methyl iodide. Correlation of these results with those of earlier experiments showed that the initial removal rates of elemental iodine vapor and cesium and uranium particles were nearly linear with spray flow rate over the range of conditions investigated.

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1.0 INTRODUCTION

This report presents the results of large-scale experiments on the removal of fission products from containment atmospheres by water sprays. Engineered safety systems for many power reactors include containment spray systems to remove fission products which could become airborne in the containment vessel as a result of accidents. The rapid removal of airborne fission products reduces the source term for potential radiation exposure to the public due to an accident.

Iodine-131 is usually the critical fission product in analyses of radiological consequences of fission product release accidents involving water reactors; hence its rate of absorption by spray systems enters importantly into the site safety analysis. Because of the importance of iodine absorption by sprays, the USAEC Division of Reactor Development and Technology has sponsored an extensive program to develop experimentally verified mathematical methods for predicting the iodine absorption rate under post accident conditions. Work reported in this document is part of that program.

The Containment Systems Experiment (CSE) program conducted by Battelle-Northwest for the Commission provides a large-scale model containment vessel in which the removal of airborne fission products by natural processes and by engineered safety systems may be studied. The CSE containment vessel has many features of an actual power reactor containment building. Its 30,000 ft³ shell is painted carbon steel and has several smaller compartments which can affect the transport of aerosols and gases. The size of the vessel permits spray experiments

with most important parameters appearing in mathematical models to be either at full scale or within a factor of three of full scale.

Previous reports^(1,2) of results obtained in the CSE system have presented the results of 7 of the 8 spray tests performed. The present report presents the results of the remaining test (A9) which used a higher spray rate (0.3 gal/min ft^2) than was used in other tests.

Spray flow rate is one of the important parameters in spray system design. Simple calculations which neglect drop interactions lead to the conclusions that fission product washout for a single species would be linearly related to the flow rate. More refined analyses, which consider coalescence of drops, predict that the effectiveness of the spray per unit liquid volume would be less, due to larger drop sizes. The washout rate still increases with increased flow rate, however.

The CSE spray tests have employed spray rates from 0.025 to 0.3 gpm/ft^2 , which covers the range applicable to large power reactor systems. In this report the results of the high spray flow rate experiment, Run A9, are reported and the effect of flow rate on relative effectiveness for fission product absorption observed in the CSE spray tests is compared with theoretical predictions.

2.0 SUMMARY AND CONCLUSIONS

The results of a single spray experiment in the CSE vessel, Run A9, are reported in detail. This experiment differed from earlier CSE spray experiments^(1,2) in that the flow rate was increased to about 150 gal/min, making the equivalent spray flux of $0.3 \text{ gal/min per ft}^2$ higher than current PWR spray system designs. The results, when correlated with those of earlier experiments, provide an experimental evaluation of the effect of spray flow rate on washout of iodine and particles.

After simulated postaccident atmospheric conditions (250°F, 44 psia) were established, the fission product simulant materials were injected rapidly and sampling begun. Then a cold, caustic-borate spray, pH 9.4, was operated in four separate periods. Over 1000 gas and liquid samples were taken to determine the washout characteristics for each fission product simulant material.

The theoretical aspects of the influence of spray density on washout of iodine were examined briefly. Together with the experimental measurements, they support the following conclusions:

- The experimentally determined initial washout rate for elemental iodine was essentially linear with spray flow rate up to 0.3 gal/min per ft², the maximum investigated. This agrees with the conclusion reached from theoretical considerations of coalescence that in the CSE vessel coalescence would reduce absorption by less than 5%. Therefore, similar considerations suggest that for typical PWR spray fluxes and drop fall heights, the elemental iodine washout rate should be within 10% of being linear with flow rate.
- Cesium and uranium particles were washed out at initial rates which were essentially linear with spray flow rate.
- The two-hour iodine dose reduction factor (DRF) is not directly proportional to spray flow rate because of the presence of non-elemental iodine forms and the possibility of iodine sources such as desorption from paint or liquid pools. For example, if 10% of the total iodine released to containment is as methyl iodide, increasing the spray flow rate from 1500 to 3000 gal/min in a large PWR would raise the 2-hr DRF only from 8 to 9.

- The rate of absorption of elemental iodine by the wall film is not influenced strongly by flow rate until waves form. However, even with waves, the absorption by the wall film is small compared to that by the drops for typical PWR containment systems using reactive sprays.
- The effect of coalescence up to 0.3 gal/min ft² spray flux is negligible compared to other parameters such as the gas-liquid partition coefficient, drop size and fraction methyl iodide.

3.0 THEORETICAL PREDICTION OF THE INFLUENCE OF SPRAY FLOW RATE ON FISSION PRODUCT ABSORPTION

3.1 MATERIAL BALANCE EQUATIONS

A material balance written for an airborne species in the main volume of a containment system may be expressed as

$$G + (C_{g2} - C_g) B_{12} = \sum_j [k_g (C_g - C_{gi}) A]_j = R_G + V \frac{dC_g}{dt} \quad (1)$$

in which

G = generation rate of component within main containment volume,

C_{g2} = gas phase concentration in connected volume,

C_g = gas phase concentration in main containment volume,

B_{12} = exchange coefficient for interroom transport,

k_g = instantaneous surface average mass transfer coefficient,

C_{gi} = gas concentration of solute at a deposition surface

A = exposed surface area for deposition surface,

R_G = rate of gas phase reaction of species in main volume,

V = volume of main containment gas space,

t = time.

The first term on the left side of Equation (1) accounts for generation or addition of a species to the main containment volume. The second term of the left hand member accounts for species transport between the main volume and connected rooms. The first term of the right hand member, shown as a summation over index j , represents deposition of a species onto j surfaces within the main room. The second term of the right member of Equation (1) accounts for disappearance of a species by a homogeneous gas phase reaction. The final term is the accumulation rate which results from the other input and output terms.

Equation (1) is only one of the set of simultaneous material balance equations which are needed to describe the gas phase concentrations. Equations similar to Equation (1) need to be written for each of the connected volumes. A second set of equations for the liquid volumes is also needed to account for phase equilibria.

Spray flow rate would influence the spray absorption primarily by affecting the mass transfer coefficients and surface areas for transfer. Although the interroom exchange coefficient could be influenced by the spray flow rate, this factor is expected to be of little practical influence. The influence of spray flow on the exchange coefficient would depend on the design of the openings between rooms and the extent of forced circulation provided.

The deposition terms may be listed as

$$\sum_j (k_g A)_j = (k_g A)_{\text{drops}} + (k_g A)_{\text{wall film}} + (k_g A)_{\text{dry surfaces}} \quad (2)$$

For reactive gases, uptake by dry surfaces will be negligible compared to that by wetted wall surfaces and falling drops. Hence, sorption by dry surfaces may be neglected, giving

$$\sum_j (k_g A)_j = (k_g A)_{\text{drop}} + (k_g A)_{\text{wall film}} \quad (3)$$

Since these are the dominant terms for spray absorption, only they will be considered in the following discussions.

3.2 EFFECT OF FLOW RATE ON ABSORPTION BY SPRAY DROPS

The influence of spray flow rate on drop absorption may be investigated by considering absorption by individual spray drops. For a single drop of constant size, the increase in solute concentration during its contact time is

$$q = \frac{6}{d} \int_0^{t_e} k_g (C_g - C_{gi}) dt \quad (4)$$

where

t_e = contact time,

q = amount absorbed per unit liquid volume,

d = drop diameter

Equation (4) may be solved for a number of cases of practical interest. For cases of practical interest in containment systems, C_g may be taken as constant during the drop fall time, and the various models revolve around the question of how C_{gi} varies with time.

For a well-mixed drop, and for a solute which obeys Henry's law,

$$q = C_g H \left(1 - e^{-\frac{6 k_g t_e}{H_o d}} \right) \quad (5)$$

where

H_o = effective gas-liquid equilibrium coefficient

For large values of t_e/H_0 the drop becomes saturated during its fall and q approaches a maximum value.

Equation (5) is based on the assumption that gas phase mass transfer resistance entirely controls the absorption rate. This assumption is not met in all cases and other models need also to be considered. A second limiting case is for negligible gas phase mass transfer resistance. For a rigid drop, Dankwerts⁽³⁾ equation applies:

$$Q = 8\pi a D_\ell C^* \sum_{n=1}^{\infty} \frac{ka^2 t_e + \frac{D_\ell n^2 \pi^2}{k + \frac{D_\ell n^2 \pi^2}{a^2}} \left(1 - \exp \left(-t_e \left[k + \frac{D_\ell n^2 \pi^2}{a^2} \right] \right) \right)}{ka^2 + D_\ell n^2 \pi^2} \quad (6)$$

where

Q = amount of solute absorbed by drop,

C^* = concentration of solute at surface of the drop,

k = first order reaction rate constant,

t_e = exposure time,

a = drop radius,

D_ℓ = diffusivity in liquid.

If both gas and liquid phase resistance are important, a more complex equation by Dankwerts⁽³⁾ may be used.

$$Q = 8\pi m^2 C^* D_\ell a^2 \sum_{n=1}^{\infty} \frac{kt_e (k + D_\ell \alpha_n^2) - D_\ell \alpha_n^2 \left(\exp \left(-t_e [k + D_\ell \alpha_n^2] \right) - 1 \right)}{(k + D_\ell \alpha_n^2)^2 (a \alpha_n^2 + m[am-1])} \quad (7)$$

where

$$m = \frac{k_g}{H_o D_\ell}$$

$$C^* = H_o C_g$$

$$\alpha_n = \text{nth root of } (a\alpha)\cot(a\alpha) + am - 1 = 0,$$

Equations (5), (6) and (7) may be generalized by writing for a single drop:

$$\left(\frac{C_\ell}{C_g} \right)_t = g(\alpha, \beta, Sh) \quad (8)$$

where

$$\alpha = kt_e$$

$$\beta = \frac{D_\ell t_e}{a^2}$$

$$Sh = \frac{k_g A}{H_o D_\ell}$$

The spray washout rate divided by the gas phase concentration may be written as

$$\frac{k_g (C_g - C_{gi}) A}{C_g} = F g(\alpha, \beta, Sh) \quad (9)$$

where

F = volumetric spray flow rate,

A = total surface area of all drops.

From Equation (9) the spray washout rate is equal to the spray flow rate multiplied by an effectiveness factor, g . Any change in the effectiveness factor with flow rate will cause

the spray washout to vary non-linearly with spray flow rate. Examination of the parameters α , β and Sh show that only t_e , a and k_g could be influenced by the spray flow rate. The numerical values of k , D_ℓ and H_0 vary with temperature and concentrations, but not directly with the spray flow rate. The primary influence of increased flow rate on the effectiveness factor is thought to be a decrease in k_g and t_e as a result of larger drops being formed by coalescence.

Parsly⁽⁵⁾ has presented calculations of the influence of drop coagulation based on a simple, gravity agglomeration model. Typical results are presented in Figure 1, where the decrease in effectiveness is shown as a function of spray flux. The predicted reduction in spray effectiveness is of little consequence for reactor containment systems. Figure 1 predicts that effectiveness would be reduced by only 5% for the CSE vessel at the highest spray rate used (Run A9), and <10% for typical PWR containment systems. Pasedag and Gallagher⁽⁶⁾ also treated the problem of coalescence with the conclusion that coalescence would reduce the total drop surface area by about 10% for typical reactor spray systems. Thus it appears that coalescence effects are negligible compared to uncertainty in the initial drop size distribution and other spray parameters. Experimental data from the CSE spray tests are probably no more precise than $\pm 20\%$, so coalescence effects in the CSE would not be expected to be observable. All drop coalescence models currently available use many simplifying assumptions. Indeed, rigorous calculations of drop coalescence are probably not feasible. However, the models used should predict coalescence with sufficient accuracy for typical PWR spray systems.

The other main factor influenced by spray rate is temperature. The gas phase temperature will decrease much faster as

FIGURE 1. Predicted Effect of Spray Flux on Washout of Elemental Iodine

the spray flow rate increases. If chemical reaction rates are not controlling, then the transfer coefficient would be expected to increase with decreasing temperature. This effect is shown in Figure 2 where the drop enrichment for 1000- μ drops is shown as a function of temperature. The drop enrichment is directly proportional to the spray washout rate. For the perfect sink model, the drop enrichment is nearly constant over the range 50 to 120°F. Above 120°F, the drop enrichment decreases with increasing temperature. Figure 2 shows that the drop enrichment increases about 25% as the temperature cools from 250°F to 120°F. In effect, then, one should expect that higher flow rate sprays would operate at lower average temperatures and thus be slightly more effective on a real time basis than predicted by a linear extrapolation of flow rate. However, since the rate of temperature decrease is approximately proportional to flow rate, the washout rate should be independent of the dimensionless spray liquid volume, Ft/V .

3.3 EFFECT OF FLOW RATE ON ABSORPTION BY WALL FILMS

The walls of the CSE containment vessel expose about 6 times as much surface to the iodine-contaminated atmosphere as the falling spray drops at a flux of 0.1 gal/min-ft². At the same spray flux, the walls of a large PWR containment vessel have a surface area approximately equal to the drops, as shown by Table 1. The effect of the large amount of interfacial surface provided by the walls on the rate of transfer from the gas phase should be considered.

The gas phase mass transfer coefficient for spray drops is about 100 times greater than for the walls. Therefore, for gaseous solutes whose mass transfer rate is governed by gas phase resistance (e.g., elemental iodine), wall absorption is

FIGURE 2. Effect of Temperature on Elemental Iodine Absorption by a Spray Drop (Perfect Sink Model)

relatively unimportant. For slightly soluble gases which do not react in solution, both drops and wall film become saturated during a single pass through the containment vessel. The wash-out for this case is dependent on the flow rate rather than on the surface area exposed. Absorption by spray drops would dominate because only a small fraction of the spray, usually <5%, would flow down the wall. The wall can be a dominant factor, however, for materials which dissolve and react relatively slowly with the solvent. Such is the case for methyl iodide dissolution in reactive solutions containing chemicals such as sodium thiosulfate. For such cases, the absorption rate depends mostly on the exposed surface area. Estimates of the absorption rate can be made for each of these three cases.

TABLE 1. Comparison of Spray Drop and Wall Film Surface Areas

	<u>CSE (a)</u>	<u>Typical PWR (b)</u>
Spray flow, gal/min	50	1,500
Spray flux, gpm/ft ²	0.1	0.1
Mean drop size, μ	1,000	1,000
Drop fall height, ft	38	100
Total drop surface, ft ²	770	60,000
Wall film surface, ft ²	4,800	60,000
Ratio, wall film to drop surface area	6.2	1.0

a. CSE Runs A4, A6 and A7.

b. Degraded spray system, containment diameter 140 ft.

3.3.1 Transfer Controlled by Gas Phase Resistance

An example of this case is elemental iodine absorption during the fresh spray period. The amount of solute absorbed per unit volume of liquid flowing down the wall at deck level is

$$Q_w = k_w A_w C_g \quad (10)$$

where

Q_w = mass absorption rate for entire wall film,

k_w = gas phase mass transfer coefficient for transfer to wall film,

A_w = total film surface exposed to containment atmosphere,

C_g = mass concentration of solute in the bulk gas phase.

Equation (10) indicates that the absorption rate is independent of flow rate. Knudsen and Hilliard⁽⁷⁾ suggested a method for estimating the gas phase mass transfer coefficient for transfer to vertical wall films. They adapted empirical correlations for heat transfer by natural convection, allowing for mass transfer effects. Their model has been shown to apply in the absence of sprays,^(7,8) but is difficult to apply when containment sprays are operated. Since no better model is known to the authors, it is recommended for use during sprays with the suggestion that the values of the parameters be for the conditions existing in the containment vessel in the absence of sprays. This is believed to be a conservative approach because the presence of sprays would certainly induce considerable turbulence beyond the natural convection case. The recommended equation is:

$$k_w = D/L (Gr' \cdot Sc)^{1/3} \quad (11)$$

where

D = solute diffusivity in gas phase,
 L = height of containment vessel,
 Gr' = Grashof number modified for mass transfer effects,
 Sc = Schmidt number.

Equations (10) and (11) do not contain the liquid flow rate, so on this basis the absorption rate would be independent of spray rate. This prediction is not unequivocal, however. If the wall film becomes wavy, the gas phase mass transfer coefficient would increase, though to what extent is uncertain.

3.3.2 Transfer of Unreactive Gases to the Wall Film

If the solute gas is slightly soluble but does not react with the wall film liquid, the following equation applies.

$$q = \frac{Q_w}{F_w} = H' C_g \quad (12)$$

where

H' = the Henry's Law coefficient,
 F_w = flow rate of wall film.

Examples of fission product materials which fall in this category are the noble gases and, in the absence of reactive chemical additives in the spray liquid, methyl iodide.

Equation (12) applies only to the fresh spray period. During recirculation from the sump, no further absorption would occur since the solution would become saturated during its initial pass through the containment space. If very slow reactions occur or the dissolved solute is adsorbed on paint or other structural materials, there will be a continued slow removal from the gas space during recirculation.

3.3.3 Absorption by Wall Film with Simultaneous Chemical Reaction

Several models may be used here. The minimum absorption rate is given by the assumption of a smooth laminar film and no transfer from the liquid film to the paint or other substrata. For this case,

$$\frac{Q_w}{A_w} = C_{\ell i} \sqrt{k D_\ell} \tanh \left(\delta \sqrt{k / D_\ell} \right) \quad (13)$$

where

$C_{\ell i}$ = mass concentration in liquid at the gas-liquid interface,

k = pseudo first order reaction rate constant,

D_ℓ = liquid phase diffusivity,

δ = liquid film thickness.

For extremely low film flow rates the argument of the hyperbolic tangent is small and the value of the tanh function approaches the value of the argument. Therefore, the absorption rate per unit area varies linearly with the film thickness. The film thickness varies with the 0.33 power of film flow rate, hence the absorption rate should increase with the 0.33 power of the wall film flow rate for low flow rates.

At higher film flow rates, the laminar film becomes turbulent and a large increase in absorption will occur. Ripples would begin to form at a film Reynold's number in the range 5 to 25, and at a Reynold's number of 1000 to 2000 the flow becomes fully turbulent.⁽⁹⁾ Calculations made for conditions applicable to a large PWR vessel indicate that the Reynolds number would be about 1000. Thus the flow would likely be turbulent and Equation (13) would be quite conservative.

Other perturbations of the simple film model would occur during spray operation during a LOCA; e.g., spray drop impingement on the film, steam condensation on the film, uneven distribution of liquid on the walls and transfer of the solute to paint at the liquid-solid interface. All of these effects should increase absorption appreciably above that predicted by Equation (13).

For a well-mixed film, the absorption rate by the entire wall film would be

$$Q_w = k V_w C_{li} + F_w C_{li} \quad (14)$$

where

V_w = volume of liquid holdup on walls.

According to Equation (14), the absorption rate per unit area would increase less rapidly than the film flow rate because the volume of liquid held on the wall would increase less rapidly than the first power of the flow rate.

3.4 EFFECT OF FLOW RATE ON TOTAL ABSORPTION WHERE TWO COMPONENTS ARE PRESENT

Present calculations of dose reduction factors due to operation of sprays are made under the assumption that the major portion of the fission product iodine is released in the elemental form, with a small fraction (usually 0.01 to 0.1) present in a form not readily removed by sprays. This approximates the situation of a mixture of elemental iodine and methyl iodide being absorbed by caustic-borate or boric acid sprays as was done in the CSE experiments. The total concentration is

$$C_{gT} = C_{gI} + C_{gM} \quad (15)$$

where

C_{gT} = total iodine concentration in the gas space,

C_{gI} = concentration of elemental iodine in the gas space,

C_{gM} = concentration of methyl iodide in the gas space.

The concentration of elemental iodine, C_{gI} , can be estimated from Equations (1), (4), and (10). Assuming an instantaneous release and no interroom transport or vapor phase reactions, these equations combine to give

$$C_{gI} = C_{gI_0} \exp \left[- \left(\frac{F_s H_o E}{V} + \frac{k_w A_w}{V} \right) t \right] \quad (16)$$

where

C_{gI_0} = concentration at time zero,

F_s = flow rate to vessel as spray drops,

H_o = effective gas-liquid equilibrium coefficient,

E = average fractional saturation attained by drops.

Assuming that absorption of methyl iodide is negligible during the fresh spray period,

$$C_{gM} = C_{gM_0} = f C_{gT_0} \quad (17)$$

where

C_{gM_0} = concentration of methyl iodide at time zero,

f = fraction of total iodine released as methyl iodide

C_{gT_0} = concentration of total iodine forms in the gas phase at time zero.

Equation (15) now can be expressed as

$$C_{gT} = C_{gT_0} \left[(1 - f) e^{-\left(\frac{F_s H_o E}{V} + \frac{k_w A_w}{V}\right) t} + f \right] \quad (18)$$

Equation (18) shows that the total iodine concentration in the gas space should decrease exponentially immediately after spraying is started, but should approach asymptotically a value corresponding to the methyl iodide concentration. Since absorption by spray drops usually greatly exceeds absorption by the wall film, the initial washout rate constant is approximated by

$$(\lambda_s)_{\text{early}} = \frac{F_s H_o E}{V} \quad (19)$$

H_o , E and V are independent of flow rate, therefore, the initial washout rate constant is approximately linear with flow rate.

An examination of Equation (18) shows that F_s , V and A_w are specified by the system design, and the value of f is usually arbitrarily assumed. This leaves H_o , E and k_w to be estimated from theoretical considerations. Equation (11) can be used to estimate the magnitude of the wall film mass transfer coefficient, k_w . Two expressions have been derived for estimating the drop fractional saturation, E . For well-mixed drops,

$$E_{wm} = 1 - \exp - \left(\frac{6k_g h}{H_o u_t d} \right) \quad (20)$$

where

E_{wm} = fractional saturation for well-mixed drops,

k_g = gas phase mass transfer coefficient based on mean drop diameter,

h = drop fall distance,
 u_t = terminal drop settling velocity,
 d = effective mean drop diameter.

It can be pointed out that for large values of H_o ,

$$E_{wm} \approx \frac{6 k_g h}{H_o u_t d} \quad (21)$$

and

$$\lambda_s = \frac{6 k_g F_s h}{V u_t d} \quad (22)$$

which applies for cases where the drops act as perfect sinks for iodine during their entire fall time.⁽¹⁰⁾

A more general expression for E which includes the effect of liquid phase resistance can be derived from Dankwert's equations⁽³⁾ which assume stagnant, spherical drops.

$$E_{st} = 1 - \sum_{n=1}^{\infty} \frac{6 Sh^2 \exp(-\beta \alpha_n^2)}{\alpha_n^2 [\alpha_n^2 + Sh(Sh-1)]} \quad (23)$$

where

E_{st} = fractional saturation for stagnant drops,

α_n = nth root of equation $\alpha \cot \alpha + Sh = 1$,

$\beta = D_\ell t_e / a^2$

$Sh = k_g a / H_o D_\ell$

Parsly⁽¹¹⁾ has published a computer code which solves Equation (23), using the theoretical values of Eggleton as input for H_o .^(12,13) The two models (stagnant and well mixed drops)

have been evaluated for a range of conditions of practical interest. The results are presented in Figure 3 in terms of the ratio of absorption of stagnant to well-mixed drops. For drop fall heights applicable to CSE and large PWR's, the maximum difference is about 30%. Thus, the potential error associated with internal circulation within drops is not expected to be highly significant in applying theory to containment spray systems. For smaller experimental facilities (drop heights <20 ft) and for solutions with low values of H_0 , the stagnant drop model is preferred.

The proper value for H_0 is probably the least accurately known of all the spray parameters. In some cases serious errors can result from inadequate information about the partition coefficient. Eggleton's⁽¹³⁾ theoretical values have become widely accepted, but as Eggleton pointed out, experimental verification is desirable. Nishizawa⁽¹⁴⁾ measured the partition coefficient in water, sodium hydroxide and boric acid solutions for the temperature range 25 to 80 °C. Coleman^(10,15) made some measurements with pH 9.5 caustic-borate solutions at 120 °C. The two are in agreement at low iodine concentrations, but Nishizawa showed the partition coefficient to decrease to values of about 100 at 10 to 20 mg/l iodine liquid concentration (at 80 °C) while Coleman showed the partition coefficient to remain approximately constant ($H_0 > 5000$) with iodine concentration over the range of 0.01 to 25 mg/l in the liquid (at 120 °C). Further experimental evaluation of the partition coefficient is warranted.

3.5 EFFECT OF FLOW RATE ON DOSE REDUCTION FACTORS

A measure of the merit of a spray system is the reduction in iodine leakage from the containment vessel attributable to

FIGURE 3. Comparison of Drop Absorption for Stagnant and Well-Mixed Drops

spray operation. This can be expressed as the "dose reduction factor" (DRF) which is the ratio of the time integrated concentrations in the containment gas space without and with spray operation.

$$(\text{DRF}) = \frac{\int_0^t C_{gT_o} dt}{\int_0^t C_{gT} dt} \quad (24)$$

C_{gT_o} is a constant and C_{gT} is given by Equation (18). Integration gives

$$(\text{DRF})_t = \frac{\lambda_s t}{(1 - f)(1 - e^{-\lambda_s t}) + f \lambda_s t} \quad (25)$$

where

$$\lambda_s = \frac{F_s H_o E}{V} + \frac{k_w A_w}{V},$$

f = fraction of total release as methyl iodide.

Equation (25) applies for a two-component mixture where one of the components (methyl iodide) is not removed by the spray. If methyl iodide is removed with an exponential rate constant λ_M , Equation (24) becomes

$$(\text{DRF})_t = \frac{\lambda_s \lambda_M t}{\lambda_M (1 - f)(1 - e^{-\lambda_s t}) + \lambda_s f(1 - e^{-\lambda_M t})} \quad (26)$$

or,

$$(\text{DRF})_t = \frac{1}{\frac{(1-f)}{(\text{DRF})_E} + \frac{f}{(\text{DRF})_M}} \quad (27)$$

where $(DRF)_E$ and $(DRF)_M$ refer to the dose reduction factor for elemental iodine and methyl iodide, respectively.

Equation (25) was solved for a typical PWR spray system and the results are shown in Figure 4. The 2-hr DRF is plotted as a function of spray flow rate for three assumed values of f , the fraction of iodine released to containment as methyl iodide. Figure 4 shows that if there is no methyl iodide, the DRF is linear with flow rate. When methyl iodide is present, the DRF is increased with larger flow rates but the increase is less than linear.

Equations (25), (26) and (27) are not expected to apply for long containment times because small deviations from the simplifying assumptions, not significant at short times, may become significant at longer times. Thus, equilibrium between gas and liquid phases would be approached, and small source terms would become important after the gas phase concentration was reduced to a few percent of the initial value. Numerical integration of Equation (24) should be used for times longer than about 2 hours.

4.0 EXPERIMENTAL MEASUREMENTS

Eight large-scale spray experiments were performed as a part of the CSE program. Each of the experiments entailed several separate spray periods which yielded information of different natures. For example, elemental iodine washout was measured accurately by a short, 3-minute spray period but methyl iodide removal was too slow to detect during the same period. Therefore, long spray periods were included to gain information on methyl iodide washout, but these were of little value for elemental iodine washout measurements because this substance was rapidly removed to low levels which were difficult to distinguish from background. Altogether, 28 individual spray periods were used during the eight spray experiments.

FIGURE 4. Calculated Effect of Spray Flow Rate on Total Iodine Dose Reduction Factor

4.1 DESCRIPTION OF THE CSE SYSTEM

The experimental facility has been described in considerable detail^(1,2,16) which will not be repeated here. However, in order to discuss the experimental evidence of the effect of spray rate on washout rates, a brief description is provided.

All eight experiments were made in the main CSE containment vessel, which measures 25 ft in diameter by 66.7 ft in overall height. Figure 5 is a schematic drawing showing most of the important experimental features. Fresh, room-temperature spray solution was made up in an exterior, stainless steel storage tank. About 2000 gallons were used in most of the experiments, which is about 1.3% of the gas volume in the main room. The spray manifold near the top dome was arranged for either three or twelve spray nozzles at a uniform spacing. About 80% of the gas space in the main room was washed by spray in all tests except Run A9, in which only 50% of the space was covered.

Other features shown schematically by Figure 5 are the fission product iodine and aerosol simulant generation system,⁽¹⁷⁾ the boiler house steam line used for establishing the desired post accident atmosphere, a 360° wall trough near the deck for measuring wall liquid run-off rate and fission product concentrations in the wall film, funnels to catch falling drops, the liquid pool sampling systems, a viewing window, and the gas sampling locations. The gas was sampled at 14 different locations by Maypack clusters.⁽¹⁸⁾ Figure 6 is a schematic diagram of a single Maypack showing the trapping components in the sequence used. Twelve Maypacks were installed in each cluster, each with its own solenoid valve.

FIGURE 5. Schematic of Containment Arrangement for CSE Spray Tests

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 Teflon Gasket	9 Stop Ring	14 End Cap
5 Two Particle Filters	10 Charcoal Bed	15 Outlet

FIGURE 6. Schematic Diagram of a CSE Maypack

4.2 EXPERIMENTAL PROCEDURE

Briefly, the procedure was to establish the desired atmospheric temperature and pressure in the containment vessel by using boiler house steam. (In two tests the atmosphere was room-temperature air at ambient pressure). When the atmosphere reached the desired temperature, the steam feed rate was reduced to a point where thermal equilibrium was maintained, and left at this rate for the duration of the experiment. The condensate produced by warming the steel vessel was discarded. Then the fission product simulant was injected in a 10-minute period. Time was referenced to the start of aerosol injection. Samples were taken during a short waiting period to determine the removal rate by natural effects. Then the fresh spray was started. In six of the experiments the sprays were operated intermittently; in two tests they operated continuously, with recirculation from the sump after all the fresh solution had entered the vessel.

Gas samples were taken at all 14 locations simultaneously by operating electrical solenoid valves from an adjoining laboratory. The gas sample rate was maintained at $0.5 \pm 0.05 \text{ ft}^3/\text{min}$ (STP) for 3.0 ± 0.07 minutes for all samples. Liquid samples were taken more frequently early in the test when concentrations changed rapidly.

Exact timing of the spray periods was accomplished by priming the spray manifold before the test and by the use of an electric ball valve for on-off control. Visual observations confirmed that sprays started and stopped within 3 seconds of the specified times. The spraying rate was controlled to that which gave the specified pressure drop across the spray nozzles.

4.3 EXPERIMENTAL CONDITIONS

The experimental conditions and numerical values of test parameters are listed in Tables 2 through 7. Table 2 gives the physical conditions common to all experiments. Table 3 gives the values of parameters which were varied from test to test. The black lines surrounding certain parameters in Table 3 show the differences in otherwise duplicate experiments. Table 4 lists the total spray flow rates and chemical composition for each spray period. Table 5 lists the start and stop times for the six tests in which sprays operated intermittently. In the tests with a steam-air atmosphere, the cold fresh spray rapidly reduced the temperature and pressure within the vessel. Table 6 lists the atmospheric conditions immediately before and after each spray period.

The masses of iodine, cesium and uranium released into the containment vessel varied slightly between experiments, but the nominal initial concentrations are listed in Table 7. The initial mass concentration of iodine was about the maximum expected with a 50% release of the iodine inventory in the core.

Two experiments (Runs A10 and A12) were made where the sprays were started shortly before start of fission product simulant release and continued without interruption until all the fresh solution had been sprayed into the containment vessel. After a 10-minute period to arrange valves, recirculation from the sump was started and continued for about 24 hours. The results of these two experiments are reported by Postma and Coleman⁽²⁾ and will not be discussed further here.

The results of Runs A3, A4, A6, A7 and A8 were reported by Hilliard et al.⁽¹⁾ The results of Run A9 have not been reported completely in previous CSE reports and so are covered in the present report.

TABLE 2. Physical Conditions Common to all Spray Experiments

Volume above deck including Drywell	21,005 ft ³	595 m ³
Surface area above deck including Drywell	6,140 ft ²	569 m ²
Surface area/volume	0.293 ft ⁻¹	0.958 m ⁻¹
Cross section area, main vessel	490 ft ²	45.5 m ²
Cross section area, Drywell	95 ft ²	8.8 m ²
Volume, middle room	2,089 ft ³	59 m ³
Surface area, middle room	1,363 ft ²	127 m ²
Volume, lower room	3,385 ft ³	96 m ³
Surface area, lower room	2,057 ft ²	191 m ²
Total volume of all rooms	26,477 ft ³	751 m ³
Total surface area, all rooms	9,560 ft ²	888 m ²
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 ^(a)	
Thermal Insulation	All exterior surfaces covered with 1 in. fiberglass insulation ^(b)	

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

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

TABLE 3. Experimental Conditions - CSE Spray Tests

	Run A3	Run A4	Run A6	Run A7	Run A8	Run A9
Atmosphere	Air	Air	Steam- Air	Steam- Air	Steam- Air	Steam- Air
Temperature, °F	77	77	250	250	250	250
Pressure, psia	14.6	14.6	44	50	48	44
Nozzle type	(a)	(a)	(a)	(a)	(b)	(c)
Drop MMD, μ ^(d)	1210	1210	1210	1210	770	1220
Geom. Std. Dev., σ	1.53	1.53	1.53	1.53	1.50	1.50
No. of nozzles	3	12	12	12	12	12
Spray rate, gal/min	12.8	48.8	49	49	50.5	145
Total spray volume, gal	510	1950	1960	1960	2020	2300
Spray solution	(e)	(e)	(f)	(g)	(f)	(f)

- a. Spraying Systems Co. 3/4 7G3, full cone.
b. Spraying Systems Co. 3/8 A20, hollow cone.
c. Spraying Systems Co. 3/4 A50, hollow cone.
d. Mass median diameter.
e. 525 ppm boron as H_3BO_3 in NaOH, pH 9.5.
f. 3000 ppm boron as H_3BO_3 in NaOH, pH 9.5.
g. 3000 ppm boron as H_3BO_3 in demineralized water pH 5.

TABLE 4. Spray Flow Rates and Solutions
Used in CSE Experiments

	Run A3	Run A4	Run A6	Run A7	Run A8	Run A9
1st Spray						
Total flow rate, gpm	12.8	49	49	49	50	148
Volume sprayed, gal	128	490	490	490	150	444
Spraying pressure, psid	40	40	40	40	40	51
Solution	(a)	(a)	(b)	(c)	(b)	(b)
2nd Spray						
Total flow rate, gpm	12.8	49	50	48.5	50	151
Volume sprayed, gal	385	1480	1500	1455	1850	453
Spraying pressure, psid	40	40	40	40	40	52
Solution	(a)	(a)	(b)	(c)	(b)	(b)
3rd Spray						
Total flow rate, gpm	12.5	42	16	45.5	47	149
Volume sprayed, gal	735	1890	960	2730	2820	1490
Spraying pressure, psid	40	29	4	36.5	36.5	51
Solution	(d)	(e)	(e)	(e)	(e)	(b)
4th Spray						
Total flow rate, gpm	(g)	(g)	(g)	48.6	50.4	138
Volume sprayed, gal	(g)	(g)	(g)	2428	2520	8280
Spraying pressure, psid	(g)	(g)	(g)	40	40	49
Solution	(g)	(g)	(g)	(f)	(f)	(e)

-
- a. Fresh, room temperature. 525 ppm boron as H_3BO_3 in demineralized water. NaOH added to pH of 9.5.
- b. Fresh, room temperature. 3000 ppm boron as H_3BO_3 in demineralized water. NaOH added to pH of 9.5.
- c. Fresh, room temperature. 3000 ppm boron as H_3BO_3 in demineralized water. No NaOH added. pH5.
- d. Fresh, room temperature demineralized water.
- e. Solution in main vessel sump recirculated. No heat exchanger used.
- f. Fresh, room temperature. 1 w/o $Na_2S_2O_3$, 3000 ppm boron as H_3BO_3 in demineralized water. NaOH added to pH 9.4.
- g. No fourth spray.

TABLE 5. Timing of Spray Periods

		Time After Start of Iodine Release, min					
		Run A3	Run A4	Run A6	Run A7	Run A8	Run A9
First Spray	Start	40	40.5	30	30	30	30
	Stop	50	50.5	40	40	33	33
	Duration	10	10	10	10	3	3
Second Spray	Start	140	140	80	80	80	55
	Stop	170	170	110	110	117	58
	Duration	30	30	30	30	37	3
Third Spray	Start	1473	1205	1565	1323	200	90
	Stop	1533	1250	1625	1383	260	100
	Duration	60	45	60	60	60	10
Fourth Spray	Start	(a)	(a)	(a)	1443	1350	210
	Stop	(a)	(a)	(a)	1493	1400	270
	Duration	(a)	(a)	(a)	50	50	60

a. No fourth spray.

TABLE 6. Atmospheric Conditions in CSE Spray Experiments

	Run A3	Run A4	Run A6	Run A7	Run A8	Run A9
Containment vessel insulated	No	No	Yes	Yes	Yes	Yes
Forced air circulation ^(a)	Yes	Yes	No	No	No	No
Start of 1st Spray						
Vapor temp., °F ^(b)	77	77	255	248.7	250	249
Pressure, psia	14.6	14.6	44.2	50.0	50.7	44.0
Relative humidity, %	70	88	100	100	100	100
End of 1st Spray						
Vapor temp., °F ^(b)	77	77	229	234.5	243	230
Pressure, psia	14.6	14.6	38.6	44.4	48.2	38.5
Start of 2nd Spray						
Vapor temp., °F ^(b)	77	77	237	240	243	235
Pressure, psia	14.6	14.6	40.8	46.0	49.3	39.5
End of 2nd Spray						
Vapor temp., °F ^(b)	77	77	202	203	188	218
Pressure, psia	14.6	14.6	29.5	36	34.1	34.5
Start of 3rd Spray						
Vapor temp., °F ^(b)	77	77	246	248	217	223
Pressure, psia	14.6	14.6	43.9	46.8	41.5	36.0
End of 3rd Spray						
Vapor temp., °F ^(b)	77	77	233	230	218	181
Pressure, psia	14.6	14.6	40.7	41.8	32.2	27.1
Start of 4th Spray						
Vapor temp., °F ^(b)	(c)	(c)	(c)	232	247	208
Pressure, psia	(c)	(c)	(c)	42.4	52.4	29.8
End of 4th Spray						
Vapor Temp., °F ^(b)	(c)	(c)	(c)	192	175	197
Pressure, psia	(c)	(c)	(c)	32.7	32.4	28.0

a. Fan without duct located in bottom of drywell. 2400 ft³/min discharge.

b. Average of 5 thermocouples located at various elevations and radii.

c. No fourth spray.

TABLE 7. Typical Initial Fission Product
Simulant Concentrations in the
Gas Space

	<u>C_{g_o} mg/m³</u>
Elemental Iodine	100
Particulate-Associated Iodine	5
Methyl Iodide	2
Cesium	5
Uranium	5

4.4 RESULTS OF RUN A9

The primary purpose of Run A9 was to demonstrate the removal of iodine and aerosol particles from containment atmospheres at a high spray flow rate. The spray rate chosen for Run A9 of 145 gal/min is believed to have given a higher spray flux than would be attained in current generation PWR's with all spray systems operating.

The initial atmosphere was a saturated steam-air mixture at 250°F, 44 psia. Fresh room temperature water with boric acid and caustic, pH 9.4, was used. Twelve Spraying Systems Co.^(a) spray nozzles No. 3/4 A50 (hollow cone) were used at 50 psi differential pressure. Information provided by the manufacturer suggests that the mass median diameter was 1220 μ with a geometric standard deviation of about 1.5. The iodine, methyl iodide, cesium, and uranium oxide were released into the vessel and allowed to mix for 30 min. The spray system was operated for 3 min. A second 3 min. spray and then a 10 min. spray was performed for a total of 2220 gal of fresh spray solution. Finally, the spray liquid in the sump was recirculated for one hour.

a. Spraying Systems Co., Bellwood, Illinois.

The overall material balance for iodine and cesium is presented in Table 8. It shows that 15% of the iodine and 18% of the cesium remained on the vessel surface after the experiment was completed and the vessel decontaminated. This is comparable to results in earlier tests.⁽¹⁾

Figures 7 through 13 show the time history of concentrations in the main room gas space for elemental iodine, particulate iodine, iodine associated with charcoal paper, methyl iodide, total iodine, cesium, and uranium, respectively. The concentration half lives were determined by the best visual fit of the data. These observed half lives were corrected for natural removal processes⁽¹⁾ and the resulting washout constants are listed in Table 9.

Elemental iodine was removed rapidly during the first 3-minute spray period, with very little further reduction in concentration by subsequent sprays. Methyl iodide was not removed to any significant extent during the four spray periods. What reduction is shown in Figure 10 can be accounted for by diffusion into the lower rooms. Cesium and uranium were removed rapidly by the first spray period. Samples were not analyzed for uranium after the first spray period, but cesium removal continued throughout the run until its concentration was reduced to $<10^{-4}$ of its initial value.

The concentrations of iodine and cesium in the main containment vessel (CV) sump and in the drywell (DW) sump are shown in Figure 14. The liquid volumes are also shown. The concentration in the spray drops caught by funnels at various radii and elevations are shown in Figure 15. Also shown in Figure 15 is the concentration of the wall film caught by a 360° trough near the main deck. The flow rate of this wall film during spraying was about 9 gal/min, or about 6% of the total spray flow rate.

TABLE 8. Overall Material Balance - Run A9

<u>Location</u>	<u>Iodine</u>		<u>Cesium</u>	
	<u>grams</u>	<u>% (a)</u>	<u>grams</u>	<u>% (a)</u>
<u>Aerosol Generation</u>				
Starting Material	101.00	100.00	11.53	100.00
Generation Apparatus	6.16	6.10	4.90	42.50
Injection Line	1.47	1.46	1.34	11.62
Injection Line Samplers	0.25	0.25	0.03	0.26
Accounted For	7.88	7.81	6.27	54.38
Delivered to Containment by Difference	93.12	92.19	5.26	45.62
<u>Containment</u>				
	<u>grams</u>	<u>% (b)</u>	<u>grams</u>	<u>% (b)</u>
Delivered to Containment	93.12	100.00	5.26	100.00
In Liquid Pools Prior to Decontamination(c)	75.56	81.14	4.12	78.32
Samples	0.68	0.73	0.022	0.42
Purge to Stack	0.77	0.83	0.00006	0.001
Decontamination	2.05	2.20	0.135	2.56
Accounted For	79.06	84.90	4.277	81.31
On Surface (by Difference)	14.06	15.10	0.983	18.69

a. Percent of starting mass.

b. Percent of delivered mass.

c. Includes spray solution and steam condensate at t = 22 hr.

FIGURE 7. Elemental Iodine Gas Phase Concentration, Run A9

FIGURE 8. Particulate Iodine Gas Phase Concentration, Run A9

FIGURE 9. Gas Phase Concentration of Iodine Associated with Charcoal Paper, Run A9

FIGURE 10. Methyl Iodide Gas Phase Concentration, Run A9

FIGURE 11. Total Iodine Gas Phase Concentration, Run A9

FIGURE 12. Cesium Gas Phase Concentration, Run A9

FIGURE 13. Uranium Gas Phase Concentration, Run A9

TABLE 9. Washout of Gases and Particles in Run A9

	First Spray		Second Spray		Third Spray		Fourth Spray	
	$t_{1/2}$	$\lambda_s^{(a)}$	$t_{1/2}$	$\lambda_s^{(a)}$	$t_{1/2}$	$\lambda_s^{(a)}$	$t_{1/2}$	$\lambda_s^{(a)}$
	min	min ⁻¹	min	min ⁻¹	min	min ⁻¹	min	min ⁻¹
Elemental Iodine	0.58	1.19	42	0.017	34	0.020	180	0.0038
Particulate Iodine	0.59	1.17	(b)	(b)	(b)	(b)	(b)	(b)
Iodine Associated with Charcoal Paper	1.3	0.528	4.7	0.148	37	0.019	(b)	(b)
Methyl Iodide	(b)	(b)	(b)	(b)	(b)	(b)	(b)	(b)
Total Iodine	0.78	0.885	17	0.042	37	0.019	150	0.0047
Cesium	1.08	0.643	2.0	0.34	5.4	0.13	33	0.021
Uranium	2.3	0.31	(c)	(c)	(c)	(c)	(c)	(c)

a. Due to spray only. Corrected for natural process removal.

b. Indeterminate.

c. Not analyzed.

FIGURE 14. Liquid Volumes and Concentrations in Vessel Sumps, Run A9

FIGURE 15. Concentrations in Spray Drops and in Wall Film, Run A9

During the first spray period, approximately 19 grams of iodine was lost from the gas space, as calculated from the concentration reduction shown in Figure 11. From Figure 15, the concentration of iodine in the spray drops is approximately

$$C_{\ell} \text{ drops} \approx 5 \times 10^4 e^{-1.4t} \quad (28)$$

Integration for the first spray period gives approximately 18 grams gained by the spray. The good agreement is considered fortuitous, for the accuracy of this calculation is low, but the exercise serves as a check on evaluations made solely on the basis of gas phase sampling.

The effect of the cold, fresh sprays on containment atmosphere temperature and pressure is shown graphically in Figure 16. The rates of change in temperature and pressure are given in Table 10.

4.5 SUMMARY OF EXPERIMENTAL WASHOUT MEASUREMENTS

The experimental washout coefficients measured during the first spray periods of Runs A3, A4, A6, A7, A8 and A9 are listed in Table 11. The last column gives values of λ_s for "total inorganic iodine," which represents all iodine forms except methyl iodide. Methyl iodide washout was not measurable during the first spray periods and thus is now shown in the table.

FIGURE 16. Containment Atmosphere Response to Fresh Spray, Run A9

BNWL-1591

TABLE 10. Pressure and Temperature
Response to Cold Sprays

	Rate of ^(a) Temperature Change, <u>°F/min</u>	Rate of Pressure Change, <u>psi/min</u>
First Spray	-6.3	-1.8
Second Spray	-5.3	-1.6
Third Spray	-4.2	-1.1

a. Average of 5 thermocouples in the main gas space.

TABLE 11. Summary of Initial Spray Washout Coefficients

Run No.	λ_s Observed, min ⁻¹ (a)			
	Elemental Iodine	Particulate Iodine	Iodine on Charcoal Paper	Total Inorganic ^(b) Iodine
A3	0.126	0.055	0.058	0.125
A4	0.495	0.277	0.063	0.43
A6	0.330	0.32	0.154	0.31
A7	0.315	0.31	0	0.20
A8	1.08	0.99	0.365	0.96
A9	1.19	1.17	0.53	1.14

a. For first spray period, corrected for natural removal on vessel surfaces.

b. Total iodine less methyl iodide.

5.0 INTERPRETATION OF EXPERIMENTAL MEASUREMENTS

The theoretical effect of spray flow rate on washout of iodine was discussed in Section 3. In this section, the experimental measurements made in the CSE are compared with theoretical predictions in an attempt to see if coalescence or other effects at high flow rates caused a deviation from simple theory.

5.1 WASHOUT OF IODINE

The experimental washout coefficients for elemental iodine are plotted as a function of spray flow rate in Figure 17. All the experimental points shown are for tests using the same drop size distribution (1210 μ MMD). The two curves represent the predicted washout coefficient as determined by the simple perfect sink model, Equation (20). Two curves are required because k_g and u_t are temperature dependent. Although the experimental points lie slightly below the curves, the washout coefficients are approximately linear with flow rate.

A more valid evaluation of the relation between washout rate and flow rate can be seen by comparing the ratio of experimental to theoretical washout with flow rate. The question arises as to which predictive model should be used. The coefficients calculated from three models are listed in Table 12. The ratios of the experimental to these predicted values are plotted as a function of spray flux in Figure 18. The curves are fitted to the data points by least squares approximations. If there is no significant coalescence or other unexpected influence at the higher fluxes, the curves should be horizontal lines. The data are too few for good statistical treatment, but the figure shows clearly that there is no downward trend. The conclusion is that the washout rate for elemental iodine is linear with spray flow rate over the range of flow rates investigated. This substantiates the same conclusion arrived at from theoretical considerations in Section 3.

TABLE 12. Comparison of Predicted and Experimental Washout Coefficients for Elemental Iodine

Run Number	Experimental (a)	λ_s , Washout Coefficient, min^{-1}		
		Perfect (b) Sink Model	Well-Mixed (c) Drop Model	Stagnant (d) Drop Model
A3	0.126	0.127	0.163	0.121
A4	0.495	0.405	0.561	0.392
A6	0.330	0.361	0.496	0.382
A7	0.315	0.353	0.485	0.366
A8	1.08	0.842	0.846	0.763
A9	1.19	1.00	1.34	1.16

- a. From first spray periods.
- b. Equation (22), using mass median diameter drop size.
- c. Equations (19) and (20), with $H_o = 5000$ and using surface mean diameter.
- d. Equations (19) and (23), with $H_o = 5000$ and using surface mean diameter.

FIGURE 17. Effect of Spray Flow Rate on Washout Coefficient for Elemental Iodine in the CSE

FIGURE 18. Ratio of Experimental to Predicted Washout of Elemental Iodine

5.2 WASHOUT OF PARTICLES

Figures 19 and 20 show graphs of the washout coefficients as a function of spray flow rate for cesium and uranium particles, respectively. Data points from all six experiments are shown since drop size does not have a large effect on washout of particles.⁽¹⁾ The washout coefficient is dependent on the drop collection efficiency, E , a factor which is usually not known precisely. Therefore, two theoretical curves are shown on each figure for two values of efficiency which span the observed results. Prediction of efficiencies has been discussed in earlier reports.^(1,2,10)

Figures 19 and 20 show that the washout coefficients for both cesium and uranium particles were essentially linear with flow rate. Any deviation from theory can easily be explained by slight errors in our knowledge of aerosol particle size. It is concluded that all particulate materials are removed from the gas space at a rate linearly dependent on spray flow rate.

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FIGURE 19. Effect of Spray Flow Rate on Washout Coefficient for Cesium in the CSE

FIGURE 20. Effect of Spray Flow Rate on Washout Coefficient for Uranium in the CSE

7.0 NOMENCLATURE

a	= radius of drop
A	= total exposed surface area for deposition
A_w	= surface area of wall film
B_{12}	= exchange coefficient for interroom transport
C_g	= gas phase concentration in main gas space
C_{gi}	= gas phase concentration at the phase interface
C_{gI}	= gas phase concentration of elemental iodine
C_{gI_0}	= gas phase concentration of elemental iodine at $t = 0$
C_{gM}	= gas phase concentration of methyl iodide
C_{gM_0}	= gas phase concentration of methyl iodide at $t = 0$
C_{gT}	= gas phase concentration of total iodine
C_{gT_0}	= gas phase concentration of total iodine at $t = 0$
C_{g2}	= gas phase concentration in connected room
C_{li}	= liquid phase concentration at the interface
d	= drop diameter
D	= gas phase diffusivity
D_l	= liquid phase diffusivity
E	= fractional saturation of solute in drop, or drop collection efficiency
E_{wm}	= fractional saturation for well-mixed drops
E_{st}	= fractional saturation for stagnant drops
f	= fraction of total iodine released to containment which is in methyl iodide form
F	= total volumetric spray flow rate

F_s	= volumetric flow rate as drops
F_w	= volumetric flow rate as wall film
g	= effectiveness factor for drop absorption
G	= generation rate of component within containment volume
Gr'	= modified Grashov number
h	= drop fall distance
H_o	= effective gas-liquid equilibrium constant
H'	= Henry's Law constant
k	= pseudo-first order reaction rate constant
k_g	= gas phase mass transfer coefficient in concentration terms, as applied to drops
k_w	= gas phase mass transfer coefficient in concentration terms, as applied to wall film
L	= length of vertical surface
m	= dimensionless ratio = $k_g/H_o D_\ell$
q	= amount of solute absorbed per unit liquid volume
Q_w	= absorption rate for entire wall film
R_G	= gas phase reaction rate in main volume
Sc	= Schmidt number
Sh	= Sherwood number
t	= time
t_e	= drop exposure time
$t_{1/2}$	= concentration half life
U_t	= drop terminal settling velocity
V	= free gas volume in containment vessel
V_w	= volume of liquid wall film holdup

GREEK SYMBOLS

α = dimensionless reaction time term = kt_e

α_n = nth root of equation $\alpha_n \cot \alpha_n + Sh = 1$

β = dimensionless time = $D_{\ell}t_e/a^2$

δ = wall film thickness

λ_s = washout coefficient

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