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UC-80 - Reactor Technology

REMOVAL OF ELEMENTAL IODINE FROM REACTOR
CONTAINMENT SYSTEMS BY STRIPPING

OAK RIDGE NATIONAL LABORATORY

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REMOVAL OF ELEMENTAL IODINE FROM REACTOR
CONTAINMENT ATMOSPHERES BY SPRAYING

L. F. Parsly

SEPTEMBER 1971

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OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
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for the
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FOREWORD

The Spray and Pool Absorption Technology Program is coordinated by Oak Ridge National Laboratory for the AEC. The program involves research on all aspects of containment spray systems proposed for use as engineered safety features in pressurized-water reactor containment buildings and investigation of certain aspects of the pool pressure-suppression containment concept as applied to boiling-water reactors.

This report summarizes work on elemental iodine removal by sprays under simulated loss-of-coolant-accident conditions. The removal of organic iodides and particulates are the subjects of other reports by L. F. Parsly. The material presented in these reports was previously described in a series of documents prepared to specifically report program work on spray systems. The general heading of this report series is "Design Considerations of Reactor Containment Spray Systems"; these reports, listed below, all carry the USAEC Report number ORNL-TM-2412 and a part number:

T. H. Row, L. F. Parsly, and H. E. Zittel, Design Considerations of Reactor Containment Spray Systems - Part I, USAEC Report ORNL-TM-2412, April 1969.

C. Stuart Patterson and William T. Humphries, Design Considerations of Reactor Containment Spray Systems - Part II. Removal of Iodine and Methyl Iodide from Air by Liquid Solutions, USAEC Report ORNL-TM-2412, Part II, August 1969.

J. C. Griess and A. L. Bacarella, Design Considerations of Reactor Containment Spray Systems - Part III. The Corrosion of Materials in Spray Solutions, USAEC Report ORNL-TM-2412, Part III, December 1969.

L. F. Parsly, Design Considerations of Reactor Containment Systems - Part IV. Calculations of Iodine-Water Partition Coefficients, USAEC Report ORNL-TM-2412, Part IV, January 1970.

J. C. Griess, T. H. Row, and C. D. Watson, Design Considerations of Reactor Containment Spray Systems - Part V. Protective Coatings Tests, USAEC Report ORNL-TM-2412, Part V, October 1970.

L. F. Parsly, Design Considerations of Reactor Containment Spray Systems - Part VI. The Heating of Spray Drops in Air-Steam Atmospheres, USAEC Report ORNL-TM-2412, Part VI, January 1970.

L. F. Parsly, Design Considerations of Reactor Containment Spray Systems - Part VII. A Method for Calculating Iodine Removal by Sprays, USAEC Report ORNL-TM-2412, Part VII, February 1970.

H. E. Zittel, Design Considerations of Reactor Containment Spray Systems, Part VIII. Boiling Water Reactor Accident Radiolysis Studies, USAEC Report ORNL-TM-2412, Part VIII, October 1970.

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B. A. Soldano and W. T. Ward, Design Considerations of Reactor Containment Spray Systems, Part IX. The Utility of Ice Cubes as an Absorbent for Gaseous Fission Products, USAEC Report ORNL-TM-2412, Part IX (to be issued).

T. L. Hebble, Design Considerations of Reactor Containment Spray Systems, Part XII. Variability of the Dose Reduction Factor for the Removal of Elemental Iodine, USAEC Report ORNL-TM-2412, Part XII, May 1971.

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REMOVAL OF ELEMENTAL IODINE FROM REACTOR CONTAINMENT ATMOSPHERES BY SPRAYING

L. F. Parsly

Abstract

Containment sprays are frequently used as an engineered safety system for power reactors. One of their principal functions is to remove radioiodine from the containment atmosphere and thus to reduce the possibility of its leaking to the environment. An analytical model that describes the removal of elemental iodine from containment atmospheres by sprays is presented. Elemental iodine is usually the predominant form released. Experimental results from experiments reported by others, as well as results from 28 iodine-removal experiments, are presented and discussed. The results show that present models for iodine removal by sprays are adequate and that sprays are an effective way to remove iodine.

1. INTRODUCTION

A determination must be made when the construction of a large power reactor is proposed that operation of the reactor at its proposed site will not result in undue risk to the health and safety of the public. As part of the safety analysis, it is customary to postulate a highly improbable, very severe, design-basis accident (DBA) and to show that the radiation exposure of the public resulting from the DBA would not exceed prescribed limits.¹ For water-cooled reactors, the postulated DBA is almost always rupture of a large coolant pipe and sudden expulsion of the primary coolant into the containment building. Analysis of the consequences of such an accident generally shows that the prescribed safety limits will be exceeded unless some action is taken, and most power reactors are provided with engineered safety systems to reduce the potential radiation exposure to less than the prescribed limits.

Among the engineered safety systems commonly provided are emergency core-cooling systems and containment sprays. Emergency core-cooling systems are provided to remove the fission-product decay heat so that the fuel will not overheat and melt. Containment sprays are provided to

remove heat from the containment atmosphere, and thus limit the pressure buildup, and also to remove fission products from the containment atmosphere.

Although melting of the fuel can thus be prevented, it is expected that the thermal and mechanical shocks resulting from the loss of coolant would cause extensive damage to the fuel cladding and that substantial release of the more volatile fission products from the fuel could occur. For purposes of analyzing the consequences of the DBA, it is assumed that 100% of the noble gases, 50% of the halogens, and 1% of the other fission products will be released into the containment building.² Of the material thus presumed to be released, the radioiodine is of greatest importance, because it is both volatile and biologically active.

Even though the radioactivity associated with it is very high, the concentrations of iodine that must be dealt with are very small. Data developed by the Chemical Technology Division of the Oak Ridge National Laboratory indicate that the core of a 1000-MW(e) reactor would contain 121 g-at. of iodine, principally ^{129}I .³ Based on 50% release to the containment building, half of which is airborne in the containment atmosphere, this means that the maximum quantity airborne is 15.1 g-moles of I_2 . A typical PWR containment building might have a volume of $2 \times 10^6 \text{ ft}^3$ or 5.6×10^7 liters. Thus the maximum concentration of the gas in the atmosphere might be 2.7×10^{-7} moles of I_2 /liter. This is slightly less than 7 ppm.

The released iodine will be principally in the form of either I_2 or HI , with small amounts in the form of organic iodides (conservatively estimated as 5% of the amount released into the containment building) and an unidentified compound or compounds (some evidence⁴ indicates HOI) in amounts of the order of 1% of the iodine reaching the containment building. Since iodine is extremely reactive and plates out rapidly on surfaces, it is also assumed here that only half the iodine reaching the containment atmosphere is available to leak to the environment. The principal way in which sprays can reduce the radiation dose resulting from the DBA is to remove the HI or I_2 from the containment atmosphere so that radioiodine no longer can leak to the environment. Since HI is much more

soluble in water than I_2 , any system that will remove I_2 will remove HI at least as efficiently.

The typical spray installation in a PWR containment vessel consists of some 200 pressure nozzles located in the upper part of the vessel and arranged to cover the interior of the vessel as completely and uniformly as possible. Solution is supplied to the nozzles through a suitable header system by two or more solution-circulating pumps. The spray solution is made by mixing additive concentrate with borated water taken from the refueling-water storage tank. The water in this tank is also used initially as the water supply source for the emergency core-cooling system. The water in the storage tank is sufficient for cooling 30 to 45 min. When it is used up, the pump suction is switched to the containment building sump. Usually there is some provision for cooling the solution once recycling starts. The containment spray system is activated when the instruments detect pressure inside the containment building in excess of a set point.

Containment spray systems are emergency systems that may never be used, since only high-severity accidents will require their operation, and yet when they are needed they must work. In any accident that required operation of containment sprays, emergency core cooling would also be needed, and once recycling of the solution started, the same pool of solution would act as the source for containment spray solution and emergency core coolant.

The constraints peculiar to the emergency coolant include the following:

1. Any additive used to enhance the iodine collection and retention ability of the solution must be stable on very long-term storage.
2. The solution must include chemical shim to maintain the core subcritical. Generally this means that the mixture must contain boric acid.
3. The final solution must not corrode the core.
4. The solution must not corrode anything in the containment building that could deposit on fuel element surfaces in the core and thus lead to ineffective cooling.
5. There must be no degradation products of the solution that could foul fuel element surfaces.

6. The solution should be nonflammable and nontoxic and have high resistance to degradation by radiation.

Since spray systems are being relied on to assure the safety of plants now being designed and built, the Division of Reactor Development and Technology of the U.S. Atomic Energy Commission has sponsored a research program to evaluate the effectiveness of sprays.⁵ A major part of the program has consisted of experiments to measure the rate and extent of elemental iodine removal by sprays and the development of an analytical model to explain the results. This report summarizes the experimental results of other investigators, as well as the results of the experimental program at the Oak Ridge National Laboratory.

2. THEORY OF IODINE ABSORPTION BY SPRAYS

It is necessary to consider several factors to understand absorption of iodine by sprays. First, there is the question of what a spray is like as determined by the process of forming it and its subsequent history; second, what are the aerodynamic and hydrodynamic behaviors of the drops, including possible alteration of the drop-size distribution generated by the spray nozzles due to collisions between drops that result in their coalescence; third, what is the nature of the chemical reaction between iodine and the spray solution and what, if any, equilibria exist (included in this is the equilibrium distribution between the solution and the atmosphere); fourth, what is the mass transfer process to the drop and in the drop; and fifth, what is the change in the temperature of the containment atmosphere due to the effect of the sprays? These factors are examined in this chapter and then synthesized into a model describing the performance of the spray.

2.1 The Nature of a Spray

Sprays can be produced in a variety of ways, depending on the degree of dispersion required and other factors. Most containment spray systems use some type of relatively simple pressure nozzle. A common type is the swirl-chamber nozzle in which the liquid enters a cylindrical swirl

chamber tangentially and exits through an orifice centered on the axis of the chamber. The liquid exits from the nozzle in the form of a thin conical sheet. As the sheet spreads from the nozzle it becomes thinner and surface waves develop. Eventually the sheet breaks into rings and these into drops. The drops thus produced cover a range of sizes, and the size spectrum can be approximated by a log-normal size distribution (this means that the log of the size is normally distributed). The log-normal distribution is described by two parameters: a mean diameter, which can be defined in terms of several of the drop properties, each giving a different value for the mean, and a standard deviation, which is the same for all the means. Also, if one mean and the standard deviation are known, any other mean can be calculated.

For each of the large main spectrum drops, the process of sheet break-up produces several very fine drops (of the order of $1\ \mu$ in diameter). These settle very slowly and thus must be considered an aerosol capable of leaking from the containment atmosphere. However, it can fairly readily be shown that the finer aerosol represents a negligible fraction of the spray.

In modeling such a system, there are two possible approaches. One is to try to find a suitable definition of mean diameter; that is, one which will let the calculation yield the same result as is observed. The second is to approximate the spray by a number of groups of uniform-sized drops. This approach has the advantage that it is not necessary to know the best choice for the mean diameter. Also it is possible to deal with such problems as the effect of collisions between drops on the performance of a spray. However, a computer is required to carry out the many calculations involved. A reasonable solution is to use the second approach until the mean diameter that should be used in the simplified calculations is known.

2.2 Chemical Interaction of Sprays with Iodine

Iodine is a highly reactive substance, and it undergoes instantaneous reactions with many solutions. The reactions are generally of one of two types: hydrolysis and oxidation-reduction.

The hydrolysis reactions are readily reversible. Eggleton has presented an excellent discussion of the process and uses the following set of equations to describe it:⁶

The first of these reactions is the gas-liquid equilibrium relationship for iodine molecules. Reaction (2) shows complex ion formation. It can be an important factor in iodine dissolution when dissolved iodide ion is present. Reactions (3) and (4) are the hydrolysis reactions, with reaction (3) being the most important. The equation for reaction (2) is written the way it is to emphasize that HI is almost completely ionized, while HOI is almost completely un-ionized.

Eggleton also makes the point that in hydrolysis there can be no net oxidation or reduction. This leads to

From the definitions of K_1 through K_4 in Eqs. (1) through (4) and from Eq. (5) it is possible to solve for the total iodine concentration in the solution when the concentration of the I_2 species in either gas or liquid is known. The partition coefficient is then defined as

$$H = \frac{\text{Total iodine concentration in the liquid}}{\text{Iodine in gas}} \quad .$$

There are also cases when it would be useful to be able to calculate the partition coefficient given the total concentration in the liquid phase (for instance, when spray solution containing dissolved iodine is recycled to a containment building).⁷ In this case, solution of Eqs. (1)

through (6) would require trial and error. Fortunately, in practical cases requiring calculation of the partition coefficient from the total dissolved I, the total concentration is relatively low and the contributions of Eqs. (2) and (4) become negligible. Under these circumstances, the concentrations of I^- and HOI become equal, and a direct solution for the partition coefficient is possible.

There is considerable evidence that the hydrolysis model described above is not valid at the low concentrations of interest in reactor accident analysis.⁸⁻¹³ Watson, Bancroft, and Hoelke reported a very high partition coefficient for elemental iodine between water and air.⁸ Several workers have measured the distribution of iodine between water and organic solvents by using very low iodine concentrations (e.g., carrier-free ^{131}I) and have found the distribution coefficient (water-to-organic ratio) to be higher than at high concentration.⁹⁻¹⁴ Generally, the explanation advanced is the existence of some unidentified water-soluble iodine compound.

Eggleton showed that the partition coefficient would be affected by the redox potential of the solution. He stated that the redox potential is given by

$$E = -0.5345 - \frac{0.05914}{2} \log \frac{[I_2 (aq)]}{[I^-]^2} \quad (6)$$

He also defined

$$K_2 = \frac{[I_2 (aq)]}{[I^-]^2},$$

and from these definitions it follows that

$$K_2 = \exp \left(\frac{E - 0.5345}{0.01284} \right) \quad (7)$$

Then the partition coefficient can be calculated provided either $[I_2 (g)]$ or $[I_2 (aq)]$ and K_1 , K_2 , K_4 , and E are known.

Eggleton pointed out that the partition coefficient is a minimum when equal amounts of I^- and HOI are formed. The redox potentials are given below as a function of the pH at which the partition coefficient is

minimum:

pH	E (V)
3	-0.81
5	-0.75
7	-0.69
9	-0.63
11	-0.57

If the redox potential were controlled by some external factor, this could explain the high partition coefficient that apparently exists in dilute neutral-to-acid solutions.

The difficulty is that it is not really known why the partition coefficients are higher than predicted by the hydrolysis model. Therefore it is not possible to guarantee that high partition coefficients will be found under accident conditions. Even though it probably is too conservative, it is considered necessary to continue to accept the hydrolysis model.

The other type of reaction utilized to fix iodine in solution is oxidation-reduction. Here we are talking about reactions in which the iodine is reduced from the I^0 oxidation state to the I^- state, while something else, often a sulfur atom, is oxidized. The principal reagent proposed for carrying out this type of reaction is sodium thiosulfate. The first-stage reaction is

Further oxidation can occur in basic solution and lead ultimately to formation of sulfate. Failure to consider this leads only to underestimating the capacity of the solution and is conservative.

Another oxidation reduction reaction which might be used is that with sulfite:

With reactions of this type, if the concentration of reagent is high enough, the iodine reacts at the drop surface and the partition coefficient becomes

infinite. The concentrations of thiosulfate proposed for containment spray systems are high enough so that the reaction occurs at the drop surface.

2.3 Mass Transfer to Spray Drops

The process of transferring iodine from a gas into a drop comprises transfer across a stagnant film from the body of the gas to the surface of the drop, establishment of equilibrium at the drop surface, and transport from the surface into the body of the drop. In the case of thiosulfate, as discussed above, all iodine reaching the drop surface is instantaneously reduced to iodide, and there is no iodine present in the drop. In this case, the resistance in the drop is zero.

2.3.1 The Mass Transfer Model

In the case of the borax solutions there is always a finite concentration of the I_2 species in the liquid, and thus there can be liquid-phase resistance. A mathematically simple model of a drop absorbing iodine from a gas is that of a rigid sphere with surface resistance. In this model, transport of the iodine from the surface into the body of the drop is presumed to occur by molecular diffusion along radii. The model is inherently conservative, since any internal circulation results in faster mass transfer than occurs by molecular diffusion.

The model is described by the diffusion equation in spherical coordinates:^{*}

$$D \left(\frac{\partial^2 C}{\partial r^2} + \frac{2}{r} \frac{\partial C}{\partial r} \right) = \frac{\partial C}{\partial t} . \quad (10)$$

This equation has the following initial conditions:

$$C = C_0, \quad 0 \leq r \leq r_0, \quad t = 0 ,$$

which say that the concentration in the drop has some uniform initial value when the drop starts to fall. The boundary conditions are

^{*} See Nomenclature, p. 36, for definition of terms.

$$D \frac{\partial C}{\partial r} = k_c \left(C_G - \frac{C}{H} \right), \quad r = r_0, t > 0,$$

which state that the current of iodine through the gas equals the current into the drop while the drop is in contact with the gas.

The most general solutions to equations of this sort are obtained if the variables are converted to dimensionless form. Approximate dimensionless variables for the present problem are

$$\begin{aligned} \chi & \text{ (dimensionless concentration)} = \frac{HC_G - C}{HC_G - C_0}, \\ \rho & \text{ (dimensionless radius)} = \frac{r}{r_0}, \\ \theta & \text{ (dimensionless time)} = \frac{D_L t}{r_0^2}, \\ \text{Sh} & \text{ (Sherwood number)} = \frac{k_c r_0}{HD_L}. \end{aligned}$$

With these substitutions, the equations become

$$\frac{\partial^2 \chi}{\partial \rho^2} + \frac{2}{\rho} \frac{\partial \chi}{\partial \rho} = \frac{\partial \chi}{\partial \theta} \quad \chi = 1, 0 \leq \rho \leq 1, \theta = 0, \quad (11)$$

and

$$\frac{\partial \chi}{\partial \rho} = \text{Sh} \chi \quad \rho = 1, \theta > 0. \quad (12)$$

The solution of this equation is well known, and Carslaw and Jaeger have presented an excellent discussion of it.¹⁶

The solution for the average concentration in the drop is

$$\bar{\chi} = \sum_{n=1}^{\infty} \frac{6\text{Sh}^2 \exp(-\alpha_n^2 \theta)}{\alpha_n^2 [\alpha_n^2 + \text{Sh}(\text{Sh} - 1)]}. \quad (13)$$

The α_n are the roots of the equation

$$\alpha_n \cot \alpha_n + \text{Sh} - 1 = 0. \quad (14)$$

Equations (13) and (14) can be readily evaluated on a digital computer. This has been done,¹⁷ and typical results are shown in Fig. 1. In examining the results it became evident that large values of Sh are associated with predominantly liquid-phase mass-transfer resistance and small values with gas-film-controlled systems. By trial, it was found that if $Sh = 30$, at least 90% of the mass-transfer resistance is in the liquid, and when it is less than 0.4, over 90% of the mass-transfer resistance is in the gas film.

From the definition of $\bar{x}$ given above, it follows that $1 - \bar{x}$ is the fraction of the total possible concentration change that is attained by the drop in time θ . Thus Eq. (13) permits the absorption by a drop to be calculated when there is significant liquid-film resistance. The analysis also identifies a dimensionless parameter (Sh) whose magnitude serves to determine whether liquid-phase resistance will be significant.

Thus a valid approach seems to be to treat absorption by the drops as gas-film controlled for $Na_2S_2O_3$ solutions under all conditions and for borax solutions when Sh is less than a suitable cutoff value. When Sh is less than 0.1, the liquid-phase resistance is a small enough fraction of the total so that it can safely be neglected.

2.3.2 Calculation of the Gas-Film Mass-Transfer Coefficient

Griffiths suggested the use of the following equation,¹⁸ attributed to Ranz and Marshall,¹⁹ for calculating the gas-film mass-transfer coefficient, k_c :

$$\frac{k_c d}{D_V} = 2.0 + 0.6 N_{Re}^{\frac{1}{2}} N_{Sc}^{\frac{1}{3}} \quad (15)$$

In a review of the analogous problem of heat transfer between immiscible liquids, Sideman cites 16 equations proposed by various workers.²⁰ Eight of these are variations of Eq. (15); that is, the equation is of the same form, but there are minor differences in the values of the constants. Thus the choice of Eq. (15) seems adequately justified.

Fig. 1. Typical Results of Calculation of Absorption by a Liquid Drop with Surface Resistance.

2.3.3 Conservatism of the Mass-Transfer Model

It is generally accepted that safety calculations must be inherently conservative. Therefore we wish to emphasize the conservatism of the model we propose. First, we consider the spray droplet as a rigid body in which the only mass-transfer mechanism is molecular diffusion. The severe changes in shape associated with the birth of a drop are known to cause oscillation in newly formed drops, although this apparently damps out as the drop ages. Frictional drag may set up circulation inside the drops. Sideman presents an excellent and well-documented discussion of drop circulation.²⁰

Plit has published a number of papers on the subject of absorption by drops.²¹⁻²⁶ He makes a distinction between "small" and "large" drops on the basis of mass-transfer performance. Drops are considered small if enhanced mass transfer due to internal circulation does not occur. From Ref. 27 it appears that Plit considers that the transition from small to large behavior occurs at a diameter of about 3 mm (3000 μ) for water at room temperature. Because of the effect of temperature on the viscosity of water, the transition diameter should probably decrease with increasing temperature.

The essential point, though, is that the rigid-drop model takes the most conservative view possible of mass transfer inside the drop. If there is any circulation, the mass-transfer rate can only increase. In addition, Sideman cites extensive data to show that when there is circulation in the drop, the outside mass-transfer coefficient is increased as well.²⁰

An additional element of conservatism is introduced by our method of calculating the partition coefficients. We calculate the partition coefficient at the drop surface on two bases: (1) an assumption that the concentration in the liquid at the drop surface is in equilibrium with the bulk gas and (2) the concentration in the drop, if the drop contains any iodine when it enters. Then we choose the lower of these two values. The first procedure generally underestimates the partition coefficient because it is based on the assumption that the concentration in the gas at the drop surface is the same as in the body of the gas. When there is significant gas-film resistance, the concentration at the drop surface is smaller than

in the body of the gas. Since the partition coefficient varies inversely as the square root of the gas-phase concentration,⁷ this procedure underestimates the partition coefficient and overestimates the liquid-phase resistance.

If the partition coefficient calculated on the basis of the dissolved iodine is lower than that calculated on the basis of iodine in the gas, this means that stripping of iodine from the spray solution will occur. This may be expected to happen when recycling starts, because the first solution will be relatively rich in iodine, and mixing of used spray solution prior to recycling will necessarily be imperfect. The concentration peaks resulting from desorption will be appreciably lower than the original concentration of iodine in the gas phase, and the iodine will be reabsorbed after a very short time as less concentrated solution is recycled.

2.4 Drop Mechanics

As discussed above, spray drops are born as a result of the breakup of a conical sheet of liquid or by some other process in which significant mechanical energy is dissipated by creating a large amount of new liquid surface. For modeling purposes, it is convenient to assume that all the drops have a velocity whose magnitude is the linear velocity necessary to get the liquid through the flow area in the nozzle (nozzle area less area occupied by air core, if any), and whose direction is determined by the spray cone angle. Also the nozzles are assumed to be aimed vertically downward. Generally the vertical velocity of the drops at the nozzle is appreciably greater than their terminal velocity, so the drops slow down as they fall.

Dallavalle proposed a procedure for calculating drop trajectories that is simple to use and reasonably accurate.²⁹ The forces acting on the drop are taken as friction drag operating along the velocity vector and opposed to the velocity and gravity acting vertically downward. The drag coefficient is represented by three different equations, depending

the magnitude of the Reynolds number. For $N_{Re} \leq 2$, $C_D = 24/N_{Re}$; for $2 < N_{Re} \leq 500$, $C_D = 18.5/N_{Re}^{0.6}$; and for $N_{Re} > 500$, $C_D = 0.44$.

In applying Dallavalle's procedure we found that it worked best if the vertical velocity were allowed to decrease 1% in each time step. The time steps are of variable length, and the calculation is assumed to have converged when the vertical acceleration becomes less than 10 cm/sec^2 . Thus the calculation yields not a true terminal velocity but, rather, a close approximation to it.

The calculation is carried out to determine the terminal velocity and the residence time of drops of a particular size. It includes provisions to deal with the possibility that the drops may hit either the walls or the floor of a containment vessel before they have slowed down to terminal velocity. This happens in pilot-plant equipment but is improbable in reactor containment buildings.

A spray is made up of drops of various sizes and can be characterized by a suitable mean diameter and measure of dispersion. Such a distribution is conveniently approximated by a number of groups, each of which is assumed to contain all drops of the same size. We started with 61 size groups, each covering 0.1σ of the size-group range (σ is the standard deviation of the size distribution). In recent development of the computer program, the number of calculations per size group has been increased severalfold. Thus it became expedient to reduce the number of size groups, if possible. We found that a model comprising 11 size groups, each covering 0.5σ of the range, gave results quite close to those obtained from the 61-group calculation, and since the half-lives calculated from the 11-group model were slightly longer than those from the 61-group model, the error was in the conservative direction.

Mason suggested the following model for growth of a drop by capture of smaller droplets:²⁰

$$\frac{dm}{dt} = E_1 E_2 \pi R^2 W (V - v) . \quad (16)$$

In this equation, dm/dt is the growth rate of the large drop; E_1 is the probability that if two drops are supposed to occupy the same space at the same instant they will in fact collide; E_2 is the probability that a collision will result in coalescence of the drops; R is the radius of the large drop; v is the velocity of the small drop; V is the velocity of the large drop; and W is the mass concentration of the small drops.

Equation (16) can easily be transformed to give the number of small drops being captured per unit time. It is applied in that form to yield the number of groups of each size group being captured by every larger size group; then those values are summed to get the overall disappearance rate of each group. Generally it turns out that the first trial calculation indicates some of the smaller drop sizes being removed faster than they are generated, and an iterative procedure is included to adjust the average population of the several sizes until the capture rate no longer exceeds the birth rate for any group. Then the modified size distribution is used for spray performance calculations.

When this procedure was applied to calculate the effect of coalescence on the performance of the spray system in a typical PWR containment building, the calculations showed that doubling the spray flow increased the removal coefficient by a factor of 1.8, so the effect of coalescence was to degrade the performance by about 10%. Also it is worth noting that the mass-mean drop size in the containment atmosphere is considerably smaller than the mass-mean size generated by the nozzle. This comes about because big drops have shorter fall times than little ones. It is the size in the containment atmosphere that enters into the removal rate calculation.

2.5 Cooling by Sprays

Many of the chemical and physical properties affecting spray performance vary appreciably with temperature. Therefore the model must account for the temperature changes that take place while sprays are operating.

The calculation of the cooling effect of the spray is straightforward. Calculations show that the drops will come to thermal equilibrium with the containment atmosphere.³¹ Thus it is simply a matter of writing a heat

balance involving the afterheat of the fuel as a source, the removal of heat by the spray, and the change in containment atmosphere temperature resulting from the net input or removal of heat.

3. ANALYTICAL MODEL OF SPRAY PERFORMANCE

The factors discussed in the preceding section — drop mechanics, chemical effects, mass transfer rates, and temperature changes — were combined in a computer program to predict the airborne concentration of elemental iodine in a containment atmosphere as a function of time. A detailed discussion and listing of the program are presented in the Appendix. In the program, the continuous drop size spectrum is approximated by an array of discrete size groups. It is assumed that the true size distribution of the spray is log-normal, with a σ_G of 1.5.

The computer program attempts to deal with the following:

1. slowing down of drops as they fall,
2. variation in physical properties of the system with temperature,
3. variation of the equilibrium distribution of iodine between air and spray solution as a function of temperature and concentration,
4. coalescence of the drops,
5. effect of decaying afterheat.

At the start of the computation, it is assumed that steam has been released into the containment atmosphere to achieve a prescribed temperature and pressure, that all the iodine is airborne in the containment atmosphere, and if the behavior of a reactor is to be simulated, that some arbitrary time has elapsed since shutdown. The reactor is presumed to have operated at power for a long time prior to shutdown.

To start the calculation, physical properties, drop trajectories, and coalescence at the initial temperature are calculated. Then, for a suitable time step, the amount of iodine collected by drops of each size group is calculated, the change in airborne iodine concentration is determined by material balance, and the change in containment atmosphere temperature is obtained from a heat balance. In calculating the drop performance, the Sherwood number ($k_c d / 2HD_L$) for each drop size group is

calculated. If this exceeds 0.1 the exact solution to the diffusion equation with surface resistance is used to calculate iodine takeup by the drops; otherwise the equation for gas-film-controlled absorption is used.

The partition coefficient (H) is calculated on the basis of the concentration in the gas. When, later on, solution recycle starts, H is also calculated on the basis of concentration in the liquid, and the smaller of the two values is used. This approach is conservative, since in gas-film-controlled absorption the concentration in the gas phase at the drop surface is less than that in the bulk gas.

The calculation is continued for iodine-free spray solution until the solution inventory is used up. Then it is assumed that recycling of the solution starts. The solution being recycled is assumed to have the average concentration of the solution that left the containment vessel over several time steps. In order to keep the liquid concentration and time relationships right, we assume that part of the solution is sprayed into the core and that the core spray does not pick up any iodine.

The physical property, trajectory, and coalescence calculations are repeated when the temperature has dropped by 10°C since a previous calculation. The density of steam is needed in the heat balance and is recalculated for every time step.

4. EXPERIMENTAL STUDIES OF IODINE REMOVAL BY SPRAYS

Much work has been done on the removal of iodine from various atmospheres by sprays, both at ORNL and elsewhere. The results of other investigators are summarized here, and then the iodine-removal experiments at the Nuclear Safety Pilot Plant are reviewed and analyzed.

4.1 Results of Other Investigators

4.1.1 Argonne National Laboratory Experiment

An early experiment was reported by Liimatainen and Mecham in which the concentration of iodine in flowing air was reduced from 114 to 4.8 ppm in a three-stage horizontal spray column.³² The contact time of the air with the spray was reported as 2.38 sec, and the overall mass-transfer coefficient as $12.3 \text{ lb-mole hr}^{-1} \text{ ft}^3 \text{ atm}^{-1}$.

4.1.2 Canadian Investigations

Watson, Bancroft, and Hoelke conducted a series of experiments to demonstrate the effectiveness of a "dousing" procedure in limiting iodine release from the Canadian NPD-II Reactor.⁸ Part of the containment-pressure-limiting procedure for this reactor consisted of spraying water from a storage tank into the boiler room to cool its atmosphere and thus limit the pressure in it. This procedure was called dousing. Some 43 dousing experiments are reported that indicated significant reduction in the airborne iodine concentration. Six equilibrium experiments are also reported. In these experiments it was found that the partition coefficient ranged from 83 for a relatively concentrated solution (4×10^{-3} mole/liter) of iodine in water to 6.7×10^5 for a solution containing 2.3×10^{-6} mole/liter. No effect of pH was found, except in one run, where a factor of 1000 difference was found between pH 3.9 and 9.6. This insensitivity to pH, which is contrary to theory, has not been explained in a satisfactory way.

4.1.3 Japanese Experiments

Workers at the Engineering Development Division of the Mitsubishi Atomic Power Industry, Inc., have reported a number of experiments on iodine removal by sprays.³⁵⁻³⁶ Their work includes measurements of both equilibrium distribution and absorption rates. The equilibrium data reported are in fair agreement with theory. However, the temperature effect at the lowest concentration is the reverse of that predicted by hydrolysis theory. The absorption experiments were carried out with much higher concentrations than predicted to exist in containment buildings under accident conditions. This was done as a practical measure, since the iodine concentration was being determined by chemical analysis rather than by using gamma counting of ^{131}I tracer. A consequence of using the high concentrations is that liquid-phase resistance in the experiments should be more important than it would be under postulated accident conditions. The experiments show that iodine is removed several times as fast when using reactive solutions as when water is used. Calculations we have made of the expected gas-film-controlled mass-transfer rate predict much lower rates. We believe the reason for this difference is that their tank is

relatively small and much of the spray hits the tank walls. In small vessels, the wall film area is usually several times the area of the air-borne drops.

Workers at the Tokyo Institute of Technology have measured absorption of iodine by single falling drops of water and of KI solution.³⁷ Iodine concentrations were high (0.23 to 2.5 mg I/liter gas). Drop sizes used were 0.6 to 4.6 mm. For 3-mm water drops, it was found that the gas-film controlled for about the first 0.01 sec of drop lifetime and the liquid-film resistance (calculated by Higbie's form of penetration theory³⁸) controlled for times longer than about 0.2 sec. Gas-film coefficients measured by absorbing I₂ in KI solution closely checked predictions by Frössling's equation.³⁹ The results of this investigation generally seem to be in excellent agreement with gas-absorption theory.

4.1.4 Containment Systems Experiment

Ten spray experiments have been carried out at the Containment Systems Experiment (CSE) facility at the AEC's Pacific Northwest Laboratory.⁴⁰ A brief summary of the results is presented in Table 1. The typical behavior observed when the iodine was introduced before the sprays were started was removal by natural effects with a half-life of 10 to 20 min for elevated temperature, 60 min at ambient temperature; accelerated removal of iodine when the sprays were turned on that lasted until the concentration dropped to somewhat under 1% of the value it would have had if there had been no plateout; and then much slower removal by sprays or by natural effects. In most of these experiments the sprays were operated intermittently with relatively long waits between periods of spraying. In the elevated-temperature runs, steam was added continuously to maintain constant temperature.

These experiments thus represent the combined effects of spraying and natural removal processes in the CSE vessel. As indicated in Table 1, the overall decontamination factors for elemental iodine (silver-seeking species in the May packs) generally do not agree with the expectations based on hydrolysis equilibrium in the spray solution. The observed decontamination factors (DF's) are much higher in runs A-3, A-4, and A-7 than expected and are lower in runs A-8 and A-9. However, comparison of the A-3 and A-4

Table 1. Results of CSE Experiments

Run	A-2	A-3	A-4	A-5	A-6	A-7	A-8	A-9	A-10	A-12
Spray solution	None	Borax ^a	Borax	None	Borax	Boric acid	Borax	Borax	Borax	Thiosulfate
pH of solution		9.5	9.5		9.5	4.8	9.5	9.4	9.4	9.2
Flow, gpm	0	12.8	48.8	0	49	49	50.5	145	50	50
Mass-mean diameter of drops, μ		1210	1210		1210	1210	770	1220	1210	1210
Temperature, °F		76	78		250	250	250	250	250	250
Initial half-life, min ^b		5.4	1.64		1.90	2.0	0.63	0.57	0.39 ^c	0.108 ^c
Iodine concentration, $\mu\text{g}/\text{m}^3$										
Initial	10^5	10^5	9.5×10^4	10^5	10^5	10^5	10^5	1.6×10^5	1.6×10^5	1.6×10^5
Final	1.2×10^3	5×10^0	1.5×10^1	4.4×10^1	5×10^1	7.7×10^1	3.1×10^3	2.8×10^3	10^3	6×10^{-1}
Decontamination factor										
Overall	8.3×10^3	10^4	6.3×10^3	2.3×10^3	2×10^3	1.3×10^3	3.2×10^3	5.7×10^3	1.6×10^3	2.7×10^3
Expected, based on solution equilibrium		20	31		2.24×10^3	1.03	2.5×10^3	1.1×10^3	1.1×10^3	=

^a3000 ppm B + 1.5 g/liter NaOH.

^bAverage for first spraying period. In runs A-4 and A-6, samples taken during the first spraying period indicate significantly shorter half-life.

^cSpraying was started before iodine was introduced. The initial half-life was calculated from the maximum concentration obtained.

run results with run A-2 results and of run A-7 results with those of run A-5 indicates that natural effects could have accounted for the observed DF's. In the case of run A-8, thiosulfate solution was used for a final hour of spraying, and if this were included, the overall DF would be 4000.

In runs A-10 and A-12 the sprays were turned on before the iodine was introduced, and iodine was fed at a presumably uniform rate for 10 min. For this case the rate equation has the form

$$\frac{dC}{dt} = \frac{F}{V} - \lambda C .$$

From this equation,

$$C_{\max} = \frac{F}{\lambda V}$$

and

$$C = C_{\max} (1 - e^{-\lambda t}) = C_{\max} (1 - e^{-0.693/t^{1/2}}) ,$$

which was used to estimate the initial removal half-lives in these two runs. It is concluded that the maximum concentration should have been attained in 10 min and that the observed half-life is shorter than would have been expected from earlier experiments.

4.2 Nuclear Safety Pilot Plant Iodine-Removal Experiments

Thirty spray experiments for iodine removal have been carried out at the Nuclear Safety Pilot Plant (NSPP). The variables investigated have included solution composition, nozzle type, flow, temperature, and iodine preloading. These can be divided into two major groups: in about half the experiments spraying was done only for a very short time, while in the other group spraying was continued for several hours.

The pattern seen in the NSPP data is similar to that observed in the CSE data. Much of the iodine transferred to the containment vessel is deposited by natural effects while the iodine is being transferred into the containment vessel, mixed, and sampled. During the first minute or so of spraying, a sharp reduction in airborne iodine concentration normally occurs. After this, removal of iodine proceeds slowly for the rest of the spraying time when the spray is $\text{Na}_2\text{S}_2\text{O}_3$. With borax spray solutions, equilibrium apparently is achieved after about 1 hr of spraying (in 1 hr the solution is sprayed through the containment atmosphere eight to ten times).

The source of the very marked slowdown in iodine removal after the first minute or so of spraying is not known definitely. The CSE staff has suggested that desorption from surfaces may occur as the supply of airborne iodine is depleted by the sprays. Since 70 to 90% of the iodine delivered to the containment vessel apparently plates out before the sprays are turned on, this seems plausible.

Tables 2, 3, and 4 present the conditions and results of the NSPP experiments in summary form. Table 2 gives the information from short-term experiments in which sodium thiosulfate solution was used. Table 3 presents the results from short-term experiments in which borax solution was used, and Table 4 gives the results of the long-term experiments. One notable point in the results is that the overall decontamination factor in the short-term experiments is generally higher than in the long-term experiments. At first glance this seems surprising, but careful attention to the processes going on and the results of model calculations indicate that this is exactly what should happen. In the borax runs it is possible to have an equilibrium between the I_2 in the atmosphere and I_2 plus its hydrolysis products in the liquid. The equilibrium concentration is higher⁷ at elevated temperature than at room temperature. In the long-term borax runs, repeated recycling of the solution is necessary (the stored solution is only adequate for 6 to 10 min of spraying), and the solution and atmosphere should be very nearly in equilibrium with each other. In addition, in these runs a strong effort was made to cool the vessel, and the temperature was reduced from 130°C to ambient during the experiments. In the short-term experiments, all the solution was iodine-free. Calculations

Table 2. Summary Results of Group 1 Nuclear Safety Pilot Plant Iodine-Removal Experiments with Borax Solutions and Short Spraying Time

Run	27	28	30	31	32
Spray solution conditions					
pH	7 ^a	9.4	9.1	9.1	9.0
Temperature, °C	30	120	30	30	30
Nozzle identification	1713 ^b	1713	1713	1713	1713
Mass-median diameter, μ	1800	1750	1080	1100	1630
Flow, gpm	9.9	10.1	15.5	15.3	10.3
Initial containment vessel conditions					
Temperature, °C	130	130	130	30	130
Pressure, psig	45	45	45	3	45
Experimental results					
Initial iodine half-life from gamma intensity data, sec	48	48	24	38	41
Overall decontamination factor	1620	1240	2470	3950	3320
Half-life calculated from overall DF, sec	34	35	32	30	31

^aSolution for this experiment was process water without additives.

^bSpray Engineering Co. Type 1713 nozzle; one nozzle used.

Table 3. Summary Results of Group 2 Nuclear Safety Pilot Plant Iodine-Removal Experiments
With Sodium Thiosulfate Solutions and Short Spraying Period

Run	21	22	26	33	38	42	45	50	51
Spray solution conditions									
pH	7	9.3	9.1	9.1	9.3	9.3	9.3	9.3	9.2
Temperature, °C	30	30	30	30	100	30	30	120	120
Nozzle identification	J-140D ^a	J-140D	1713 ^b	1713	7G3 ^c	7G3	7G3	7G3 ^d	N5-303 ^e
Mass-median diameter, μ	200	200	1660	1550	1290	1290	1290	1290	
Flow, gpm	0.57	0.52	10.3	10.6	11	11	11	14.7	15.9
Initial containment vessel conditions									
Temperature, °C	30	30	130	130	70	130	30	130	130
Pressure, psig	3	3	45	45	10	45	5	45	45
Experimental results									
Initial iodine half-life from gamma intensity data, sec	37	38	31	21	11	12	14	14	35
Overall decontamination factor		6,500	2,730	2,040	416	10,000	3,600	28,000	39,000
Half-life calculated from overall DF, sec		38	32	33	60	27	30	26	22

^aSpray Engineering Co. J-140D misting nozzles; 12 nozzles used.

^bSpray Engineering Co. Type 1713 hollow cone nozzle; one nozzle used.

^cSpraying Systems Co. 7G3 (clusters of seven G-3 full cone nozzles); three used.

^dFour clusters used.

^eBete Fog Nozzle Co. Type N5-303 nozzle; one used.

Table 4. Summary Results of Group 3 Nuclear Safety Plant
Iodine-Removal Experiments with Long Spray Time

Run	37	36	57	72	73	74	75	76
Spray solution conditions								
Solution	Thiosulfate	Borax-I ^a (a)	Borax-I ^a	Borax	Thiosulfate ^b	Borax	Borax	Borax
pH	9.2	9.4	9.4	9.2	8.7	9.2	9.3	9.3
Temperature, °C	120	120	120	Cold	Cold	Cold	Cold	Cold
Nozzle identification	1713	7C3	7C3	1713	1713	1713	1713	1713
Flow, gpm	12	11	11	15	15	15	15	15
Containment vessel conditions								
Initial temperature, °C	130	130	130	130	130	130	130	130
Initial pressure, psig	45	45	45	45	45	45	45	45
Spraying time, min	184.5	480	480	240	240	240	240	240
Iodine distribution, %								
Spray solution	85.75	87.44	76.89	85.13	70.82	74.42	71.21	77.01
Vessel surfaces	3.78	7.25	4.32	9.64	21.36	17.90	8.22	15.72
I ₂ vaporizer	10.18	4.94	18.22	4.60	7.53	6.63	19.91	6.42
Purge system, on silver	0.014	0.05	0.012	0.31	0.038	0.87	0.31	0.61
Purge system, on charcoal	0.076	0.33	0.56	0.32	0.25	0.18	0.35	0.24
I ₂ removal performance								
Initial half-life from gamma data, sec	20	30	20					
Overall decontamination factor	6000	1877	6450	215	2440	107	257	153
Initial half-life from gas-sampling data, sec	59				30	40		25
Overall DF predicted by hydrolysis model				283		240	292	326

^a 0.28 M H₂BO₃; 0.17 M NaOH; 4.4×10^{-4} M I₂.

^b Contained 0.013 M Na₂S₂O₃; 0.06 M NaOH and 0.2 M H₂BO₃.

(continued)

Table 4 (continued)

Run	77	78	79	80	81	82
Spray solution conditions						
Solution	Borax	Borax	Boric acid ^c	Boric acid ^c	Borax	Boric acid ^d
pH	9.2	9.2	7.5	7.3	8.0	7.2
Temperature	Cold	Cold	Cold	Cold	Cold	Cold
Nozzle identification	1713	Special ^e	1713	1713	1713	1713
Flow, gpm	15	15	15	15	15	15
Containment vessel conditions						
Initial temperature, °C	130	130	130	25	130	25
Initial pressure, psig	45	45	45	5	45	5
Spraying time, min	240	345	240	240	240	240
Iodine distribution, %						
Spray solution	83.14	64.15	72.87	68.15	70.25	83.93
Vessel surfaces	10.13	17.67	12.7	7.37	21.55	6.45
I ₂ vaporizer	5.88	16.53	6.41	22.46	6.12	8.04
Purge system, on silver	0.58	1.35	2.05	1.69	1.95	1.55
Purge system, on charcoal	0.26	0.08	0.50	0.32	0.13	0.02
I₂ removal performance						
Initial half-life from gamma data, sec						
Overall decontamination factor	161	62	45	46	48	59
Initial half-life from gas-sampling data, sec	33	65	36	141	24	378
Overall DF predicted by hydrolysis model	222	299	2.2	2.2	37	2.2

^cProcess water used for solution makeup.

^dDistilled water used for solution makeup.

^eSpecial header to deliver all solution onto wall.

from the model show that when a batch of fresh solution is sprayed through a vessel atmosphere, practically all the iodine is picked up by the first part of the batch, and the final concentration in the gas phase can be orders of magnitude below the equilibrium value.

In the case of the thiosulfate solution we do not believe any volatile iodine compounds can be present, but some portion (from the results, less than 0.1%) of the spray solution will be suspended as an aerosol. When this aerosol contains iodine, there will be airborne iodine. The borax solution almost certainly makes a similar aerosol, but the concentration of volatile iodine compounds is much higher.

With one exception, the short-term iodine runs show elemental iodine decontamination factors in excess of 1000. In the exceptional case, the solution was preheated to a higher temperature than the vessel atmosphere. In this case, the outward flux of water vapor would have interfered with diffusion of iodine to the gas-liquid interface.

The long-term runs for which data are presented in Table 4 included two thiosulfate runs, two with borax preloaded with iodine, six with borax spray, three with boric acid spray, and one in which all the spray solution was delivered onto the vessel wall. The two thiosulfate runs gave decontamination factors of well over 1000, even though in one run only 20% of the amount of reagent normally used was provided. The borax runs gave decontamination factors within a factor of about 2 of the values predicted by the hydrolysis model; the observed decontamination factors were always less than calculated.

In runs 56 and 57, the borax solution was preloaded with stable iodine to give a concentration ten times that expected as a result of a postulated loss-of-coolant accident. This was done because of the concentration effect on the partition of iodine; that is, the suspicion that the iodine in solution would interfere with removal of radioiodine by the spray. What happened was quite the contrary and was what should have been expected. The preloaded solution worked better than the iodine-free solution. The reason is that isotopic dilution is superimposed on the distribution of the iodine between the gas and liquid so that while there is almost certainly more iodine in the gas, it contains less radioiodine.

The overall decontamination factors measured in the experiments were lower than predicted on the assumptions that isotopic equilibrium had been obtained and no spray remained in the containment atmosphere as an aerosol. The overall decontamination factors were comparable with those from the thiosulfate experiments.

In run 78 a special header was used to place all the spray solution on the containment vessel wall near the top. The intent was to measure the effect of the wall film. However, from the appearance of the film, it seems that some momentum transfer from the film to the vessel atmosphere may have occurred; there probably was induced circulation in the atmosphere.

In runs 79, 80, and 82, the solution was 0.028 *M* boric acid. The pH of the solutions after use was slightly above 7, probably due to NaOH remaining from prior decontamination. These runs showed overall decontamination factors of approximately 50, in contrast to the expected values of about 2 predicted by hydrolysis theory. As discussed above, the evidence is that hydrolysis theory fails for dilute neutral-to-slightly-acid iodine solutions.

In run 81, neutralization of the boric acid was carried only to a pH of 8. The overall decontamination factor was again approximately 50. However, the initial half-life was shorter than for boric acid.

5. VERIFICATION OF THE ANALYTICAL MODEL

A major goal of our spray program has been to develop the analytical model described earlier in this report. We believe the proper approach to take is to develop a model based on sound theory, use pilot-plant experiments to prove the model, and then use the model to calculate performance in large containment vessels.

In the model we undertook to account for several factors: equilibrium distribution of iodine between the spray solution and the containment atmosphere, rate of mass transfer of iodine from containment atmosphere to spray, and cooling by the spray solution. Experiments may deal with one or more of these factors.

In the long-duration experiments, one important observation was the ultimate equilibrium distribution of the iodine. The spray solution hold tank at the NSPP has a capacity of approximately 100 gal, and flows of 10 to 15 gpm are ordinarily used; therefore if spraying is continued for longer than 6 to 10 min, the solution must be recycled. In many of the experiments, spraying was continued for 3 hr and the solution was recycled some 20 to 30 times. Under these circumstances, equilibrium distribution of the iodine should have been achieved.

If it is assumed that all the iodine was initially in the gas and decontamination factor is defined as the ratio of initial to final gas-phase concentration, a material balance gives

$$DF = 1 + (V_L/V_G)H .$$

This equation, which deals with the ultimate equilibrium only and has nothing to say about the rate process, was used to calculate the predicted decontamination factors given on the last lines of Tables 1 and 4. The results indicate that the calculation predicts the observed DF within a factor of 3 or better in a basic solution (pH = 9.5). In neutral or acid solutions, the observed DF's have consistently been much higher than those calculated.

In Table 1, the overall decontamination factors in runs A-6, A-9, and A-10 are within a factor of 2 of the predicted values, those for A-3, A-4, and A-7 are much higher than predicted, and the DF from run A-8 is much lower. The deviation in run A-3 can be attributed to the use of fresh solution; that in run A-7 is the low pH effect. The differences observed in runs A-4 and A-8 are not readily explained.

In Table 4, agreement within a factor of 3 or better is found in runs 72, 74, 75, 76, 77, and 81. The observed decontamination factor was lower than predicted in run 78 when all the solution was delivered to the containment vessel wall. This may simply reflect less efficient contacting of the spray solution and the iodine in the vessel atmosphere. In runs 79, 80, and 82, where boric acid was sprayed, the observed decontamination factors were over 20 times those predicted.

The data thus appear to support the view that the simple hydrolysis model is valid for pH 9 to 9.5 and conservative for lower pH.

A second aspect of the model to be examined is the absorption rate process. The basic model is gas-film controlled absorption as described by Eq. (15). Tables 5 and 6 present a comparison of the predictions of the model with experimental results from the CSE and the NSPP. Table 5 shows that the calculated values for the CSE tend to be lower than the experimental data. In the NSPP, the observed half-lives are consistently lower than those predicted (Table 6) and are rather insensitive to solution flow. A possible explanation is that much of the absorption occurs in the film of spray solution or condensate or both on the containment vessel wall. The model takes no credit for absorption by the wall film.

The ultimate goal in modeling is to describe the absorption process, including the influence of transient effects. The current version of the model undertakes to do this. Figures 2 and 3 illustrate the application

Table 5. Comparison of Calculated and Observed Iodine-Removal Half-Lives in the CSE

Run	Half-Life (sec)	
	Calculated	Observed
A-3	254	282-318
A-4	66	98 ^a
A-6	75	114 ^b
A-7	75	120
A-8	29	38
A-9	28	34

^aBased on samples taken midway through the first spraying period, the half-life could have been 49 sec.

^bBased on samples taken midway through the spraying period, the half-life could have been 57 sec.

of the model to an NSPP experiment. The calculation covers the first 20 min. In Fig. 2, a comparison of the observed and calculated temperatures is made. Over most of the range, the observed temperatures were lower than calculated. We suspect that the difference can be attributed

Table 6. Comparison of Calculated and Observed Initial Iodine-Removal Half-Lives in NSPP at 120°C

Spray Nozzle Type	Flow (gpm)	Half-Life (sec)	
		Calculated	Observed
1713 ^a	10	214	48
	15	45	32
7G3 ^b	11	62	27
	14.7	46	26

^a Spray Engineering Company, Burlington, Mass.

^b Spraying Systems Company, Bellwood, Ill.

Fig. 2. Calculated and Experimental Temperatures in NSPP Run 77.

Fig. 3. Calculated and Experimental Iodine Concentration in NSPP Run 72.

in part to the way the model treats the heat stored in the vessel shell. It is assumed that the stored heat is released instantaneously as the vessel atmosphere cools. In reality, there may be a delay.

Figure 3 shows the comparison of observed and predicted airborne iodine concentration. The model gives a reasonably good representation of the concentration-time behavior.

6. APPLICATION TO DESIGN

Figure 4 illustrates the application of the model to predicting the performance of the spray system in a typical PWR containment building. It shows the airborne iodine concentration as a function of time over the first 2 hr. The figure indicates the almost negligible effect of liquid-phase resistance. If there were no liquid-phase resistance, the left-hand line would be straight, but there is a barely perceptible curvature in the upper part of the line. The results of this calculation justify neglecting the liquid-phase resistance.

The form of the curve after recycling starts is determined by the assumptions made as to how well the spray solution becomes homogenized in

Fig. 4. Calculated Iodine Removal for 1000-MW(e) PWR.

the containment vessel sump. If it were assumed to be homogenized completely, the curve would level off and rise slowly due to the decrease of H with temperature. The present curve was calculated by assuming partial homogenization.

Another point illustrated in Fig. 4 is the extent to which the airborne concentration is reduced below its equilibrium value before recycling of the solution begins. This confirms that larger decontamination factors may be expected in short-term spray experiments where only fresh solution is used than in longer duration experiments where the solution is recycled.

The calculation produces an average airborne I_2 concentration over the 2-hr period, as well as the concentration versus time data, so the dose-reduction factor may be conveniently calculated.

7. CONCLUSIONS

Sprays can effectively remove elemental iodine from containment building atmospheres under accident conditions. An adequate theory exists

for predicting the performance of both basic sodium thiosulfate and borax solutions. For thiosulfate, the extent of iodine removal becomes limited by formation of a fairly stable aerosol of spray solution, but the limit is less than 0.001 of the total iodine in the containment vessel. In the case of borax solution, the extent of removal is determined by the equilibrium between the solution and the vessel atmosphere, in accordance with hydrolysis theory. Boric acid solutions remove much more iodine than hydrolysis theory predicts they should. The partition coefficient for iodine between water and air in such solutions is much higher than predicted by the hydrolysis theory, but other data on dilute iodine solutions indicate that the high partition coefficients should be expected.

Both experimental results and calculations with an analytical model indicate that liquid-phase mass-transfer resistance is negligibly small for borax solutions under accident conditions. This is also true for basic sodium thiosulfate solutions. In the case of boric acid solutions, the initial half-lives are longer than for the other solutions, and it appears probable that the liquid-phase resistance is significant.

Additional research and development on spray systems would be valuable in the following areas:

1. An effort to upgrade the present hydrolysis model should be made so that the distribution of iodine between water and air as a function of temperature, concentration, and pH at concentrations of 10^{-5} moles I_2 /liter of water and lower can be predicted. This will require some precise fundamental chemical research to identify all the iodine compounds present in the gas and liquid phases and to measure their concentrations.
2. Scoping tests are needed to determine what, if any, effect radiation has on the performance of spray systems.
3. Studies to determine the sensitivity of spray systems to uniformity of liquid distribution are needed. Sprays undoubtedly entrain much gas and generate turbulence in building atmospheres. Therefore, it is likely that significant maldistribution can exist without serious deterioration of spray performance. However, no data available today substantiate this reasoning.

There seems to be plenty of evidence that containment sprays will collect iodine and make it unavailable for leakage, at least to the extent needed and probably better. The critical factors appear to be mechanical — will pumps run and valves operate? A high degree of confidence in the reliability of the mechanical elements of the spray system must be established and maintained.

NOMENCLATURE

$\bar{C}$	concentration, g-mole/cm ³
C_D	drag coefficient, dimensionless
C_G	concentration in gas phase, g-mole/cm ³
C_{max}	maximum concentration in gas phase, g-mole/cm ³
C_0	concentration in spray solution entering containment vessel, g-mole/cm ³
d	drop diameter, cm
D_L	diffusivity in liquid, cm ² /sec
D_V	diffusivity in gas, cm ² /sec
E_1	probability that a collision of two drops actually will occur, dimensionless
E_2	probability that a collision between two drops will result in their coalescence, dimensionless
F	rate (assumed constant) at which iodine is being fed to containment vessel
H	partition coefficient (ratio of concentration in liquid to concentration in gas at equilibrium), dimensionless
K_1	equilibrium constant of reaction (1), dimensionless
K_2	equilibrium constant of reaction (2), cm ³ /g-mole
K_3	equilibrium constant of reaction (3), g-mole/cm ³
K_4	equilibrium constant of reaction (4), g-mole/cm ³
k_c	mass-transfer coefficient, cm/sec
m	mass, g
r	radius, cm
R	radius of large drop, cm
t	time, sec
$t_{\frac{1}{2}}$	half-life, sec

v	velocity of smaller drop, cm/sec
V	velocity of larger drop, cm/sec
V_c	containment volume, cm ³
W	concentration of small drops in gas volume, g/cm ³

Greek Letters

α_n	n th root of characteristic equation, dimensionless
θ	dimensionless time ($4Dt/d^2$)
λ	rate coefficient, sec ⁻¹
μ	viscosity, poises
ρ	dimensionless radius, $2r/d$
ρ_G	density of gas, g/cm ³
ρ_L	density of liquid, g/cm ³
σ_g	standard deviation of log-normal distribution (84.17% size/50% size)
χ	dimensionless concentration, $(HC_G - C)/(HC_G - C_0)$
$\bar{\chi}$	average dimensionless concentration

Dimensionless Groups

N_{Re}	Reynolds number, $dV\rho/\mu$
N_{Sc}	Schmidt number, $\mu/\rho_G D_V$
Sh	Sherwood number, $k_c d/2\pi D_L$

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Appendix

COMPUTER PROGRAM FOR EVALUATING SPRAY PERFORMANCE

Main Program

The program starts by reading in data from a set of three cards. The arrangement of data on the input cards and the significance of the variables are presented in FORMAT statement 1 and the following COMMENT cards. Then the initial values of a number of the variables are assigned. Subroutine PTCDF is called to calculate the initial value of the partition coefficient; the diameters of the drops in each of the 11 size groups are calculated; and subroutine DIST is called to calculate the weight fraction of each size group at the spray nozzles. Subroutine ENTH is called to calculate the enthalpy of the containment atmosphere at time zero.

Then a stepwise calculation of the time-dependent behavior is started. Memory space is provided for 720 time steps (enough to cover 2 hr using 10-sec time steps). On the first time step, subroutine PRPP is called to calculate the physical properties of the solution and the containment atmosphere, and subroutine STEP to calculate the drop trajectory, fall time, and mass-transfer coefficient. These calculations are repeated only when the containment atmosphere temperature has decreased by 10°C or more from the last value at which a calculation was made.

Coalescence of the drops and a size distribution corrected for coalescence are then calculated from the data provided by STEP. The coalescence calculation also is repeated only when new trajectory data have been calculated.

Then PTCDF is called to calculate the partition coefficient based on concentration in the gas. Next a calculation is made of the iodine concentration in the spray solution entering the containment vessel. The calculation is designed on the basis that the inlet concentration is zero until all the stored spray solution is used up. The initialization procedure includes a calculation of the number of time steps needed to use up the stored solution. After the supply of fresh solution is used up, solution previously sprayed through either the containment atmosphere or

the core must be recycled. It was thought to be unrealistic to assume either that complete mixing of the solution would occur or that no mixing would occur. As a compromise it was assumed that the concentration of the recycled solution was the average of the concentration of the solution leaving the vessel over some period of time. The number of time steps to be used for the averaging process is specified in the input for a case. It is thought that averaging the exit concentration over 5 to 10 min is probably sufficient.

After the inlet concentration is calculated, subroutine PTCF is used to calculate a partition coefficient based on the concentration in the drop. If the partition coefficient based on the liquid concentration is less than that based on the gas concentration, it is substituted for the latter. Then the Sherwood number is calculated. If the Sherwood number is found to be less than 0.1, the absorption or desorption is assumed to be gas-film controlled, and the rate is calculated on that basis. If Sh exceeds 0.1, the rate is calculated by the exact solution of the diffusion equation. The rate coefficients so calculated for each size group are summed to obtain a rate coefficient for the spray. Then subroutine LAPP is called to evaluate the change in gas concentration over the time step. By material balance, the new exit liquid concentration is calculated.

Next, a heat balance on the building is made to determine the temperature change during the time step. Subroutine HTBAL is used. The source terms include afterheat from the fuel (computed by the Way-Wigner equation from data on the reactor power and time after shutdown) and stored heat in the walls and internals. The solution is assumed to come to temperature equilibrium with the atmosphere and to enter the vessel at its initial temperature until recycling starts. Then a fixed fractional approach to the cooling-water temperature in an external heat exchanger is assumed.

The program is fixed so that the user can specify the frequency of printout, since printout of every time step is usually not necessary.

Subroutines

Subroutine DIST

The DIST subroutine contains a calculation of the fraction of the drops in each of the 11 size groups. It evaluates the error integral over the size range spanned by each group.

Subroutine PROP

The subroutine PROP contains the calculation of a number of physical properties needed in the program. The sources of the equations used are identified by comments in the program.

Subroutine STEP

The STEP subroutine contains a calculation of the drop trajectories and the gas-film coefficient. Drops are assumed to enter at an initial velocity determined by the flow, liquid flow area, and cone angle for the nozzle. The drag coefficient is calculated and then the vertical acceleration and the time necessary to reduce the vertical velocity by 1% (or to increase it in the rare case that the initial vertical velocity is less than terminal velocity). The horizontal acceleration and horizontal velocity are calculated for the time step. Horizontal and vertical displacements are calculated by using arithmetic-average velocities for the time step. The gas-film coefficient at the beginning of the time step is calculated by the Ranz-Marshall equation, and the product $k_c \Delta t$ is stored. This procedure is repeated in successive time steps until the vertical acceleration is greater than -10 cm/sec^2 . At this point, the velocity is assumed to be terminal, and the remainder of the fall is done in a single time step. The program contains provisions to deal with drops striking either the vessel wall or floor before attaining terminal velocity. Also there is a provision to deal with failure to converge. Drop fall time and the time-averaged gas-film coefficient are returned to the main program.

Subroutine PTCOF

In subroutine PTCOF, partition coefficients based on known concentrations in the gas are calculated. The subroutine includes a correction to account for the effect of temperature on pH. Empirical equations fitted to tabulated data are used to evaluate equilibrium constants at temperature. The calculation is that described in Ref. 1.

Subroutine PTCF

Subroutine PTCF contains a calculation of the partition coefficient based on knowing the concentration in the solution. It is assumed that under conditions in which this equation is applied, only one hydrolysis reaction is significant:

With this assumption, an explicit calculation of the partition coefficient is possible based on knowledge of the liquid concentration, temperature, and pH.

Subroutine LOOP

Subroutine LOOP evaluates the gas-phase concentration change during a time step. In the subroutine, each time step is broken up into 100 substeps to evaluate equations

$$\Delta C_G = \lambda C_G \Delta t$$

and

$$C_G = C_G - \Delta C_G .$$

Subroutine HTBAL

The function of subroutine HTBAL is to calculate a heat balance on the system and to determine the temperature change during a time step. A trial-and-error procedure is used, and the balance is considered acceptable if it is within 1%. To prevent waste of computer time, an abnormal termination is taken if 100 trials are made without achieving an acceptable heat balance.

Subroutine ENTH

Subroutine ENTH calculates the enthalpy of steam. It contains an empirical equation fitted to tabulated data for the enthalpy of saturated steam over the range 0 to 150°C. The values calculated by the equation deviate from the tabulated values by, at most, 1 in the fourth significant figure.

Subroutine DENS

The DENS subroutine calculates the pressure and density of saturated steam from a known temperature. Equations given in Keenan and Keyes are used.²

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Program Listing

```

C THIS PROGRAM HAS BEEN WRITTEN TO CALCULATE THE PERFORMANCE OF A SPRAY SYSTEM
C IN REDUCING THE TEMPERATURE AND AIRBORNE IODINE IN A CONTAINMENT BUILDING
      IMPLICIT REAL*8 (A-H,O-Z)
      DIMENSION D(11),VGAV(11),VTER(11),T1(11),CL(720),W(11),WT(11),
1DROPV(11),DROPS(11),DNDT(11,11),DNDT(11,11),AN(11),SONDT(11),
2THETA(11),E(11),SH(11),WTFR(11),OL(11),AP(11),ALPHA(50),TW(720),
3IDENT(10),AN2(11),W2(11),XPL(720),YPL(720),U(4),Y(4),XTPL(720),
4TEN(720)
      READ(50,1)IDENT,VOL,HT,RAD,VLIO,DMEAN,SIGMAG,ANGLE,DORIF,ITYPE,
1NNOZ,Q,T,PH,NSTEP,TSTEP,CO,N2,POWER,N3,TO,TCW,WALCAP,MTEFF,F
1 FORMAT(10A4,4D10.2,F10.1,F5.1,F5.1,F10.3,11,19,F10.2,F5.1,F5.1,11C
1,F10.1/D10.2,110,D10.3,110,F10.0,F10.1,D10.2,2F5.7)
C THE FOLLOWING DATA ARE SUPPLIED IN THE READ STATEMENT
C IDENT A 40-CHARACTER NAME FOR THE SYSTEM BEING CALCULATED
C VOL SYSTEM VOLUME IN CUBIC CENTIMETERS
C HT FALL HEIGHT FOR SPRAY IN CM
C RAD VESSEL RADIUS IN CM
C VLIO SUPPLY OF RECIRCULATING LIQUID AVAILABLE
C DMEAN MEAN DIAMETER OF DROPS, MICRONS
C SIGMAG STANDARD DEVIATION OF DROP SIZE DISTRIBUTION
C ANGLE INCLUDED ANGLE OF CONE PRODUCED BY SPRAY NOZZLE, DEGREES
C DORIF DIAMETER OF ORIFICE IN SPRAY NOZZLE, INCHES
C ITYPE TYPE NOZZLE (0 = HOLLOW CONE, 1 = FULL CONE)
C NNOZ NUMBER OF NOZZLES IN BUILDING (EACH ORIFICE COUNTS AS 1 NOZZLE)
C Q FLOW TO BUILDING, GPM
C T INITIAL BUILDING TEMPERATURE, DEG C
C PH ROOM-TEMPERATURE PH OF SPRAY SOLUTION
C NSTEP NUMBER OF TIME STEPS TO BE CALCULATED
C TSTEP DURATION OF TIME STEP, SEC
C CO INITIAL CONCENTRATION OF I2 IN CONTAINMENT, MOLES/L
C N2 NUMBER OF TIME STEPS OVER WHICH CONC. IN RECYCLING SOLUTION IS
C TO BE AVERAGED
C POWER THERMAL POWER OF REACTOR, MEGAWATTS
C N3 OUTPUT DATA TO BE PRINTED EVERY N3 TIME STEPS
C TO TIME INTERVAL BETWEEN REACTOR SHUTDOWN AND SPRAY STARTUP, SEC
C TCW TEMPERATURE OF COOLING WATER, DEG C
C WALCAP HEAT CAPACITY OF VESSEL SHELL & INTERNALS, G CAL/DEG C
C MTEFF IS THE FRACTION TEMP. APPROACH IN THE COOLANT HEAT EXCHANGER
C F IS THE RATIO OF CORE + CONTAINMENT SPRAY FLOW TO CONT. SPRAY FLOW
      DO 2 I = 1,11
        VGAV(I) = 0.
        VTER(I) = 0.
      2 T1(I) = 0.
      NDT2 = 0
      WRITE(51,52)IDENT,T,POWER,Q,NNOZ,DORIF,PH,TCW,DMEAN,SIGMAG
52 FORMAT(10A4// 'INITIAL TEMPERATURE = ',F6.1,' DEG C REACTOR PO
POWER = ',F7.1,' MEGAWATTS// ' SPRAY SOLUTION FLOW = ',F10.1,' GPM
3 NUMBER OF NOZZLES = ',15,' ORIFICE DIA = ',F6.3,' INCHES// ' PH
4 OF SOLUTION = ',F5.2,' COOLING WATER TEMPERATURE = ',F5.1,' DE
5G C// ' MEAN DROP DIAMETER = ',F6.1,' MICRONS STD DEVIATION = '
6,F4.1// ' TIME, SEC TEMP DEGREES C GAS CONC MOLES/L RECYCL
7E LIO MOLES/L PARTITION COEFFICIENT SH (MEAN DROP DIA)// '
      DO 3 I = 1,720
        XTPL(I) = 0.
        TEN(I) = 0.
        TW(I) = TCW
        XPL(I) = 0.
        YPL(I) = 0.
      3 CL(I) = 0.
      VI = 1./VOL
      AFTO = POWER*(2.38690**10)/(6.220-2)

```

C 2.388905 CONVERTS MEGAWATTS TO GCAL/SEC, 6.22D-7 IS WAY-WIGNED CONSTANT

```
IX = 1
SUMCG = 0.
QA = 62.08*0
VL = VOL/(F*QA*STEP)
TIM = 0.
NI = 10*INT(VL/Q/(F*QA*STEP))
TEMPK = 273.16 + T
CALL PTCOF(CC, TEMPK, PH, PHT, H, AK1, AK2, H1ONCI
WRITE(51,43) TIM, T, CC, CLIN, H
NPT = 0
DT = 1.
TNEW = TEMPK
DM = 1.0-4*DMEAN
PHI = 0.01745329*(ANGLE/2.)
CG = CC
DO 4 I = 1,11
D(I) = DM*DEXP((1(I) - 1)/2.) - 2.5)*DLOG(SIGMAGI)
```

4 CALL DIST(I,WT(I))

C CALCULATE FLOW CROSS-SECTION IN NOZZLES ASSUMING .20DIA AIR CORE FOR
C HOLLOW CONE OR FULL THROAT FOR SOLID CONE

```
IF(ITYPE.EQ.1) GO TO 5
ACORIF = 0.26*0.7854*((2.54*DOORIF)**2)
GO TO 6
5 ACORIF = 0.7854*((2.54*DOORIF)**2)
6 OLORIF = QA/NONNOZ
AL = QA/(3.141592*RAD*AD)
DO 10 J = 1,11
DROPS(J) = 0.523598*(D(J)**3)
10 DROPS(J) = AL*WT(J)/DROPS(J)
CALL ENTH(T, HOLD)
NP1 = 1
NP2 = 1
IPL1 = 0
IPL2 = 0
THAX = NSTEP*STEP
WRITE(15,80)
80 FORMAT(' L. F. PARSLEY / XYPL ')
DO 7 NS = 1, NSTEP
IF((TEMPK - TNEW).LT.10..AND.NS.GT.1) GO TO 8
CALL PROP(TEMPK, RHOLIO, RHOATG, RHOATM, RHOATH, P, PAIR, PTOTAL, ETA,
1 ETAAIR, ETAMIX, DIAIR, DIWTR, DIMIX, SMITNO, ETASCL, OL17, OLCH31)
DO 9 I = 1,11
9 CALL STEP(I, D(I), PHI, ACORIF, OLORIF, PAC, HT, ETAMIX, RHOATH, SMITNO, DIMIX
1, RHOLIO, VTER(I), VGAV(I), T(I))
DO 11 J = 1,11
11 W(J) = AL*WT(J)*T(I)/HT
DO 12 J = 1,11
DO 12 I = 1,11
DMDT(I,J) = 0.
DMDT(I,J) = 0.
12 AN(J) = W(J)*HT/DROPS(J)
20 DO 13 I = 2,11
IM1 = I - 1
DO 14 J = 1, IM1
DX = D(I) - D(J)
IF (VTER(I).LE.VTER(J)) GO TO 14
DMDT(I,J) = 0.7859*(DX**2)*W(J)*(VTER(I)-VTER(J))
14 DMDT(I,J) = DMDT(I,J)*AN(I)/DROPS(J)
12 CONTINUE
DO 15 J = 1,11
15 SMDT(J) = 0.
DO 16 J = 1,11
DO 16 I = 1,11
16 SMDT(J) = SMDT(J) + DMDT(I,J)
DO 17 I = 1,11
IF (SMDT(I).LE.DROPS(I)) GO TO 17
W(I) = 0.9*W(I)
```



```

17 CONTINUE
  IF(SDNDT(1)/DROPS(1).LE.1.0)GO TO 18
  DO 19 J = 1,11
19 AN(J) = W(J)*HT/DROPV(J)
  GO TO 20
18 TEMPR = TNEW
  DO 60 J = 1,11
  AN2(J) = DROPS(J) - SDNDT(J)
60 W2(J) = AN2(J)*DROPV(J)
  WEIGHT = 0.
  DO 51 I = 1,11
51 WEIGHT = WEIGHT + W2(I)
  DO 52 I = 1,11
  WTF(I) = W2(I)/WEIGHT
52 QL(I) = QA*WTF(I)*TSTEP
  8 CALL PTCF(CG,TNEW,PH,PHT,H,AK1,AK2,HIONC)
  SAM = 0.
  DO 31 I = 1,11
  THETA(I) = (DLI2*TI(I)*6.1/(D(I)*D(I)))
31 E(I) = 0.
  IF(NS.LE.N1)GO TO 22
  K = NS - N1
  KI = K + (N2 - 1)
  IF (N1.LT.(2*N2))KI = K + N1/2
  SUNC = 0.
  JA = 0
  DO 33 I = K,KI
  JA = JA + 1
33 SUNC = SUNC + CL(I)
  CLIN = SUNC/JA
  CALL PTCF(CLIN,TNEW,HIONC,AK1,AK2,H1)
  IF(H1.LT.H) H = H1
  GO TO 34
32 CLIN = 0.
34 DO 35 I = 1,11
  SH(I) = VGAV(I)*D(I)/(2.*H*DLI2)
  IF(SH(I).GT.1)GO TO 36
C AMOUNT COLLECTED BY DROPS IN GAS FILM CONTROLLED ABSORPTION
  AM(I) = .01*QL(I)*VGAV(I)*TI(I)*6.1/(D(I)*H)
  GO TO 35
36 DO 37 J = 1,51
  X = 3.141592654*J
  B = 1.570796327
38 FCFX = X*DCOS(X) + (SH(I) - 1.)*DSIN(X)
  FCFB = (X-B)*DCOS(X-B) + (SH(I) - 1.)*DSIN(X-B)
  IF(FCFX.GT.0..AND.FCFB.LT.0..OR.FCFX.LT.0..AND.FCFB.GT.0.)GO TO 39
  X = X - B
39 B = B/2
  IF (B.GE.1.0-16)GO TO 38
37 ALPHA(J) = X
  SUM1 = 0.
  N = 6.*SH(I)*SH(I)
  G = SH(I)*(SH(I) - 1.)
  DO 40 J = 1,50
  B = ALPHA(J)**2
  C = DEXP(-B*THETA(I))
  TERM = D/C/(B*(B + G))
  SUM1 = SUM1 + TERM
  IF(TERM.LT.1.0-18)GO TO 41
40 CONTINUE
41 E(I) = 1. - SUM1
  AM(I) = .01*QL(I)*E(I)
35 SAM = SAM + AM(I)
  CGX = CG
  CALL LOOP (CG,SAM,H,CLIN,VI,CG2)
  CLINS = CLIN + VL*(CG - CG2)
  CG = CG2
  TIM = TIM + TSTEP

```

```

C      IGRID IS 0 FOR TIC MARKS ALONG AXES, 1 FOR FULL GRID
      IF(ITYPE.EQ.1) GO TO 83
      WRITE(15,84)
      PRINT 84
84  FORMAT(30X,'TIME, SECONDS'/14X,'CONCENTRATION IN GAS, MOLS/L'/22X,
      1'CONCENTRATION IN GAS VS TIME')
      GO TO 91
83  WRITE(15,85)
      PRINT 85
85  FORMAT(22X,'TIME,SECONDS'/4X,'CONCENTRATION IN GAS, MOLS/L'/14X,
      1'CONCENTRATION IN GAS VS TIME')
91  WRITE(15,93)
      PRINT 93
93  FORMAT('YLOG')
      WRITE(15,94) TMAX, CO
      PRINT 94,TMAX, CO
94  FORMAT('      0.00',D10.3, /'      1.0-11',D10.3)
C THIS GIVES RANGE OF ABSCISSA AND ORDINATE
      NPT1 = NPT
      DO 150 NSET = 1,NP1
104  IF(NPT1.LT.96) GO TO 100
      NPT1 = NPT1 - 96
      NPTS = 96
      GO TO 102
100  NPTS = NPT1
      IF(NPTS.EQ.0) GO TO 103
101  WRITE(15,102) NPTS
      PRINT 202, NPTS
202  FORMAT(' 4'/0',I2)
102  FORMAT(' 4'/I2)
      DO 95 NP = 1, NPTS, 4
      DO 96 JJ = 1,4
      U(JJ) = 0.
96  Y(JJ) = 0.
      DO 97 JJ = 1,4
      JJJ = 96*(NSET - 1) + NP + JJ - 1
      IF(JJJ.GT.NPT) GO TO 97
      U(JJ) = XPL(JJJ)
      Y(JJ) = YPL(JJJ)
97  CONTINUE
      WRITE(15,98)(U(JJ),Y(JJ),JJ=1,4)
      WRITE(51,98)(U(JJ),Y(JJ),JJ=1,4)
98  FORMAT(8E10.3)
95  CONTINUE
      IF(NPTS.LT.96) GO TO 103
150  CONTINUE
103  CONTINUE
      NPT2 = NSTEP
      WRITE(15,81) ITYPE, NP2, ISIZE, IGRID
      PRINT 81,ITYPE,NP2,ISIZE,IGRID
      IF(ITYPE.EQ.1) GO TO 105
      WRITE(15,106)
      PRINT 106
106  FORMAT(20X,'TIME, SECONDS'/13X,'TEMPERATURE, EGREES C'/12X,'CONTA
      INMENT VESSEL ATMOSPHERE TEMPERATURE VS TIME')
      GO TO 107
105  WRITE(15,108)
      PRINT 108
108  FORMAT(22X,'TIME, SECONDS'/7X,'TEMPERATURE, DEGREES C '/4X,'CONTAI
      INMENT VESSEL ATMOSPHERE TEMPERATURE VS TIME')
107  WRITE(15,109) XTPL(NSTEP)
      PRINT 109,XTPL(NSTEP)
109  FORMAT('LINEAR'/0.00',E10.2/'      0.00      1.602')
      DO 152 NSET = 1,NP2
      IF(NPT2.LE.96) GO TO 111
      NPT2 = NPT2 - 96
      NPTS = 96
      GO TO 112

```

```

111 NPTS = NPT2
    IF(NPTS.EQ.0) GO TO 113
112 WRITE(15,102) NPTS
    PRINT 202, NPTS
    DO 114 NP = 1,NPTS,4
    DO 115 JJ = 1,4
    U(JJ) = 0.
115 Y(JJ) = 0.
    DO 116 JJ=1,4
    JJJ = 96*(NSET - 1) + NP + JJ - 1
    IF(JJJ.GT.NSTEP) GO TO 116
    U(JJ) = XTPL(JJJ)
    Y(JJ) = TEM(JJJ)
116 CONTINUE
    WRITE(15,98)(U(JJ),Y(JJ),JJ=1,4)
    WRITE(51,98)(U(JJ),Y(JJ),JJ=1,4)
114 CONTINUE
152 CONTINUE
113 WRITE(15,95)
    PRINT 99
    99 FORMAT(' -1'/' THE END')
    STOP 101
    END

```

```

SUBROUTINE DIST(I,DISTR)
  IMPLICIT REAL*8(A-H, O-Z)
  X=DFLOAT(I)
  Y = ((X - 1.)/2.) - 2.25
200 IF(I.EQ. 6) GO TO 201
  Z = Y - 0.5
  U = DABS(Y)
  V = DABS(Z)
  UF = DEFF(U/DSORT(2.D0))
  VF = DEFF(V/DSORT(2.D0))
  DISTR = 0.5*DABS(UF - VF)
  GO TO 202
201 DISTR = DEFF(Y/DSORT(2.D0))
202 RETURN
  END

```

```

SUBROUTINE STEP (I,D,PHI,AORIF,QORIF,RAD,HT,ETAMIX,RHOATM,SMITNO,
  IOINIX,RHOLIO,VTER,VGAV,TI)
C IN THIS SUBROUTINE A STEPWISE CALCULATION IS MADE OF THE DROP TRAJECTORY. THE
C DROPS ARE ASSUMED TO ORIGINATE AT THE NOZZLE WITH AN INITIAL VELOCITY WHOSE
C MAGNITUDE IS DETERMINED BY THE VOLUME IS DETERMINED BY THE VOLUME DISCHARGED
C AND THE LIQUID CROSS-SECTION IN THE NOZZLE (THE NOZZLE MAY HAVE AN AIR CORE)
C AND WHOSE DIRECTION IS DETERMINED BY THE INCLUDED ANGLE OF THE SPRAY CONE
C DETERMINED BY THE NOZZLE (THE NOZZLE IS ASSUMED TO POINT DOWN)
  IMPLICIT REAL*8(A-F, O-Z)
C SET INITIAL VALUES OF VARIABLES
  TIME = 0.
  X = 0.
  Y = 0.
  VGIT = 0.
  K = 1
  DELTAT = .01
  VTER = 0.
  V = QORIF/AORIF
  VX = V*DSIN(PHI)
  VY = V*DCOS(PHI)
  DELVX = 0.
  DELVY = 0.
  TEST = 0.

```

```

C RVK = RECIPROCAL KINEMATIC VISCOSITY
RVK = RHOATM/ ETAMIX
TOPCON = (-3.*RHOATM)/(4.*RHOLIO*D)
SNI = 0.6*(SMTNO**0.333)
DID = DIMIX/D
VCI = 0.
300 RENNO = D *V*RVK
C DRAG COEFFICIENTS ARE TAKEN FROM DALLAVALLE MICROMERITICS
IF(RENNO .LE.500.) GO TO 304
CSUBR = 0.44
GO TO 305
304 IF(RENNO .LE.2.) GO TO 306
CSUBR = 18.5/(RENNO **0.6)
GO TO 305
306 CSUBR = 24./RENNO
305 TOP = TOPCON*V*CSUBR
307 DELVY = (980. + TOP*VY)*DELTAT
TEST = DABS(DELVY /VY)
IF(TEST.LT.0.01) GO TO 308
DELTAT = DELTAT/2.
IF(DELTAT.LT.1.E-6) GO TO 397
GO TO 307
308 VY = VY + DELVY
Y = Y + (VY - .5*DELVY )*DELTAT
DELVX = TOP*VX*DELTAT
VX = VX + DELVX
X = X + (VX - .5*DELVX )*DELTAT
SNI = 2.0 + SNI*DSORT(RENNO)
VGI = SNI*DID
VGIT = VGI*DELTAT
TIME = TIME + DELTAT
V = DSORT(VX**2 + VY**2)
IF(DELVY 1400,399,500)
400 NSTEP = 1
DO 401 K = 2,500
402 RENNO = D *V*RVK
IF(RENNO .LE.500.) GO TO 403
CSUBR = 0.44
GO TO 404
403 IF(RENNO .LE.2.) GO TO 405
CSUBR = 18.5/(RENNO **0.6)
GO TO 404
405 CSUBR = 24./RENNO
404 TOP = TOPCON*V*CSUBR
C CONVERGENCE CRITERION
IF((980. + TOP*VY).GT.-10.) GO TO 407
DELVY = -0.01*VY
411 DELTAT = DELVY/(980. + TOP*VY)
414 DELVX = TOP*VX*DELTAT
C SOMETIMES VX TRIES TO OSCILLATE AROUND 0. THIS IS TO FORCE IT TO 0.
IF(DABS(DELVX).GT.VX) GO TO 408
X = X + (VX + 0.5*DELVX)*DELTAT
Y = Y + (VY + 0.5*DELVY)*DELTAT
C IN THE SMALLER SYSTEMS, THE DROPS MAY HIT THE WALL OR THE FLOOR
IF(X.GT.RAD) GO TO 409
IF(Y.GT.HT) GO TO 410
VX = VX + DELVX
IF(VX.LT.-1)VX = 0.
VY = VY + DELVY
412 SNI = 2. + SNI*DSORT(RENNO)
VGI = SNI*DID
VGIT = VGIT + VGI*DELTAT
TIME = TIME + DELTAT
401 V = DSORT (VX**2 + VY**2)
C A NORMAL TERMINATION OF THE DO LOOP INDICATES FAILURE TO CONVERGE
GO TO 397
408 DELVY = DELVY/2.
IF(DELTAT.GE..00001) GO TO 411
X = X + 0.5*VX*DELTAT
VX = 0.

```

```

VY = VY + DELVY
Y = Y + (VY - .5*DELVY)*DELTAT
GO TO 412
409 X = X - (VX + 0.5*DELVX)*DELTAT
Y = Y - (VY + 0.5*DELVY)*DELTAT
DELTAX = PAD - X
DELTAT = DELTAX/VX
X = PAD
TIME = TIME + DELTAT
VGIT = VGIT + VGI*DELTAT
Y = Y + VY*DELTAT
GO TO 399
410 Y = Y - (VY + 0.5*DELVY)*DELTAT
X = X - (VX + 0.5*DELVX)*DELTAT
Z = HT - Y
DELTAT = Z/VY
Y = HT
X = X + (VX + 0.5*DELVX)*DELTAT
TIME = TIME + DELTAT
VGIT = VGIT + VGI*DELTAT
GO TO 399
407 IF(VX.LT.1.) GO TO 413
DELVY = 0.
VTER = VY
DELTAT = 0.01
GO TO 414
413 Z = HT - Y
DELTAT = Z/VY
TIME = TIME + DELTAT
Y = HT
VGIT = VGIT + VGI*DELTAT
VTER = VY
X = X + VX*DELTAT
GO TO 399
500 NSTEP = 1
DO 501 K = 2,500
502 RENNO = 0 *V*RVK
IF(RENNO .LE.500.) GO TO 503
CSUBR = 0.44
GO TO 504
503 IF(RENNO .LE.2.)GO TO 505
CSUBR = 18.5/(RENNO **0.6)
GO TO 504
505 CSUBR = 24./RENNO
504 TOP = TOPCON*V*CSUBR
IF((980. + TOP*VY).LT.10.) GO TO 507
DELVY = 0.01*VY
511 DELTAT = DELVY/(980. + TOP*VY)
514 DELVX = TOP*VX*DELTAT
IF(DABS(DELVX).GT.VX) GO TO 508
X = X + (VX + 0.5*DELVX)*DELTAT
Y = Y + (VY + 0.5*DELVY)*DELTAT
IF(X.GT.PAD) GO TO 509
IF(Y.GT.HT) GO TO 510
VX = VX + DELVX
IF(VX.LT.1)VX = 0.
VY = VY + DELVY
512 SNI = 2. + SMI*DSORT(RENNO)
VGI = SMI*DID
VGIT = VGIT + VGI*DELTAT
TIME = TIME + DELTAT
501 V = DSQRT(VX**2 + VY**2)
GO TO 397
508 DELVY = DELVY/2.
IF( DELTAT.GE .00001) GO TO 511
X = X + 0.5*VX*DELTAT
VX = 0.
VY = VY + DELVY
Y = Y + (VY - 0.5*DELVY)*DELTAT
GO TO 512

```

```

509 X = X - (VX + 0.5*DEL VX)*DELTAT
    Y = Y - (VY + 0.5*DEL VY)*DELTAT
    DELTAX = RAD - X
    DELTAT = DELTAX/VX
    X = RAD
    TIME = TIME + DELTAT
    VGIT = VGIT + VGI*DELTAT
    Y = Y + VY*DELTAT
    GO TO 399
510 Y = Y - (VY + 0.5*DEL VY)*DELTAT
    X = X - (VX + 0.5*DEL VX)*DELTAT
    Z = HT - Y
    DELTAT = Z/VY
    Y = HT
    X = X + (VX + 0.5*DEL VX)*DELTAT
    TIME = TIME + DELTAT
    VGIT = VGIT + VGI*DELTAT
    GO TO 399
507 IF(VX.LT.1.) GO TO 512
    DELVY = 0.
    VTER = VY
    DELTAT = .01
    GO TO 514
513 Z = HT - Y
    DELTAT = Z/VY
    TIME = TIME + DELTAT
    Y = HT
    VGIT = VGIT + VGI*DELTAT
    VTER = VY
    X = X + VX*DELTAT
    GO TO 399
397 WRITE(51,396)
396 FORMAT(' THE CALCULATION FAILED TO CONVERGE TO TERMINAL VELOCITY')
399 VGAV = VGIT/TIME
    TI = TIME
    RETURN
    END

```

```

SUBROUTINE HTBAL(TEMPK,DT,TLIN,TIM,T0,PF,DELT,V0I,QA,TSTEP,WALCAP,
1F)
C DECAY HEAT BY WIGNER-WAY APPROXIMATION, VERY LONG IRRADIATION TIME
IMPLICIT REAL*8 (A-H,O-Z)
TEMPC = TEMPK - 273.16
CALL ENTH(TEMPC,HOLD)
CALL DENS(TEMPK,RHOSTM,PSTM)
OLDHT = HOLD*RHOSTM
W = 1.
DT = 0.
OT = F*QA*TSTEP
TC = TEMPC - TLIN
TIME = TIM + T0
N = 0
IF (TIME .LE.0.) GO TO 1005
QDK = PF*TSTEP/((TIM + T0)**0.2)
GO TO 1004
1005 QDK = PF*TSTEP
C HEAT REMOVED BY COOLING SOLUTION
1004 QSOL = OT*TC
ONET = QSOL - QDK
C QNET IS NET HEAT REMOVED
IF(QNET)1003,1002,1010
1003 DT = -1.
1023 QSOL = OT*(TC - DT)
ONET = QSOL - QDK
TNEW = TEMPK - DT
CALL DENS(TNEW,RHNEW,PSTM)
T = TNEW - 273.16

```

```

      CALL ENTH ( T , HNEW)
      QATH = VOL*(OLDHT - HNEW*RHNEW + 0.0002844*DT) + WALCAP*DT
C     0.0002844 = 0.001185*0.24 (IDENS OF AIR)*(SP HT OF AIR)
      Z = DABS(QNET - QATH)
      Y = DABS(.01*QNET)
      IF(Z.LT.Y) GO TO 1002
      X = QNET - QATH
      IF(X)1020,1002,1022
1020 DT = DT - W
      N = N + 1
      IF(N.GT.100) GO TO 1006
      GO TO 1023
1022 W = 0.5*W
      DT = DT + W
      N = N + 1
      IF(N.GE.100) GO TO 1006
      GO TO 1023
1010 DT = 1.
1025 QSQL = DT*(TC - DT)
      QNET = QSQL - QOK
      TNEW = TEMPK - DT
      CALL DENS(TNEW, RHNEW, PSTH)
      T = TNEW - 273.16
      CALL ENTH(T, HNEW)
      QATH = VOL*(OLDHT - HNEW*RHNEW + 0.0002844*DT) + WALCAP*DT
      Z = DABS(QNET - QATH)
      Y = DABS(.01*QNET)
      IF(Z.LT.Y) GO TO 1002
      X = QNET - QATH
      IF(X) 1026,1002,1027
1026 W = 0.5*W
      DT = DT - W
      N = N + 1
      IF(N.GT.100) GO TO 1006
      GO TO 1025
1027 DT = DT + W
      N = N + 1
      IF (N.GT.100) GO TO 1006
      GO TO 1025
1006 WRITE(51,1007)T,DT,PF,QA,TC,QOK,TIM,VOL,DELT,HNEW,HGLD,PSTH,QATH,
      IQNET,QSQL,TIME,TLIN,TNEW,TEPC,TEPPK,TSTEP
1007 FORMAT(1H 1012010.2)
      CALL ERROR
1002 DELT = DT
      RETURN
      END

```

```

      SUBROUTINE DENS (TEMPK, RHOSTH,P)
C THIS CALCULATES THE PRESSURE AND DENSITY OF STEAM
      IMPLICIT REAL*8(A-H,O-Z)
      X = 647.27 - TEMPK
      TOP = 3.2437814 + (5.868260-3)*X + (1.17023790-8)*(X**3)
      DENOM = 1. + (2.18784620-3)*X
      PHI1X = TOP/DENOM
      PHI2X = (2.302585*X*PHI1X)/TEMPK
      PHIP = DEXP(PHI2X)
      P = 218.167/PHIP
      TAU = 1./TEMPK
      TAUSQ = TAU*TAU
      G1TAU = 82.546*TAU - (1.624605)*TAUSQ
      G2TAU = 0.21828 - (1.269705)*TAUSQ
      G3TAU = 3.6350-4 - (6.768064)*(TAU**24)
      B0 = 1.89 - 2641.6*TAU*(10**(80870*TAUSQ))
      B = B0 + (B0**2)*G1TAU*TAU*P + (B0**4)*G2TAU*((TAU*P)**3)+
      1(B0**13)*G3TAU*((TAU*P)**12)
      V = (4.55504*TEMPK/P) + B
      RHOSTH = 1./V
      RHOATH = RHOSTH + 0.001185
      RETURN
      END

```

```

SUBROUTINE ENTH (T,H)
C THIS CALCULATES THE ENTHALPY OF STEAM
IMPLICIT REAL*8 (A-H,O-Z)
HJ = 2.50001102941177D 02 + (1.9388550420168100)*T -
1 (1.98550420168067D-03)*T*T
C ABOVE IS IN JOULES/G, NEXT STATEMENT CONVERTS TO CAL/G
H = 0.23889*HJ
RETURN
END

```

```

SUBROUTINE LOOP(CG,SAM,H,CLIN,VI,CG2)
IMPLICIT REAL*8 (A-H,O-Z)
AX = SAM*VI
CGA = CG
DO 55 IZ = 1,100
DCG = AX*(H*CGA - CLIN)
55 CGA = CGA - DCG
CG2 = CGA
RETURN
END

```

```

SUBROUTINE PYCOF (CG,TNEW,PH,PHT,H,AK1,AK3,HIONC)
C THIS CALCULATES THE PARTITION COEFFICIENT ASSUMING THAT THE CONCENTRATION IN
C THE GAS, THE ROOM TEMPERATURE PH AND THE TEMPERATURE ARE KNOWN
IMPLICIT REAL*8 (A-H,O-Z)
C CALCULATION OF PH AT TEMPERATURE
REFPHT = -2.740962060625957D0 + (1.523327657381936D-02)*TNEW +
1 (2.20147928944370D 03)/TNEW + (1.402521247031774D-17)*TNEW*TNEW
PHT = PH*REFPHT/9.1845
C CALCULATION OF AK1
IF(TNEW.GT.385.5) GO TO 2001
Z = -2.49603475424395D 02 + (2.57 0223743172D 01)*DLOG(TNEW) +
1 (1.50088340394797D 04)/TNEW
GO TO 2002
2001 Z = -2.45241883602302D 01 + 4. 749381276*DLOG(TNEW) +
1 (7.287350258351257D 02)/TNEW
2002 AK1 = DEXP(Z)
C CALCULATION OF AK3
Z = -6.965252198805D-02 - 8.40141724275 03/TNEW
AK3 = DEXP(Z)
AK2 = DEXP(-111.98 + 16.725*DLOG(TNEW) + 6952.3/TNEW)
AK4 = 1.2D-11 + (3.6D-13)*(TNEW - 298.16)
CI2AQ = CG*AK1
HIONC = DEXP(-2.303*PHT)
Y = ((AK3*CI2AQ)/HIONC + AK4*CI2AQ)/(1. + AK2*CI2AQ)
CIMIN = DSORT(Y)
CI3 = AK2*CI2AQ*CIMIN
CHIO = (AK3*CI2AQ)/(HIONC*CIMIN)
CH2OI = AK4*CI2AQ/CIMIN
TOTCI2 = CI2AQ + 0.5*(CHIO + CH2OI + CIMIN + 3*CI3)
H = TOTCI2/CG
RETURN
END

```

```

SUBROUTINE PTCF (CL, T, HIONC, AK1,AK3,H)
C THIS SUBROUTINE CALCULATES THE PARTITION COEFFICIENT WHEN THE TOTAL IODINE
C CONCENTRATION IN THE LIQUID IS KNOWN. IT ASSUMES THAT HIO, I-, AND I2 ARE THE
C ONLY FORMS PRESENT IN THE LIQUID. THIS ASSUMPTION APPEARS TO BE VALID UNDER
C ALL CONDITIONS IF THE CONCENTRATION IN THE LIQUID IS LESS THAN 1.E-4 GMOLS/L
C EXCEPT FOR BRIEF TRANSIENTS, THIS CONDITION IS SATISFIED
IMPLICIT REAL*8 (A-H,O-Z)
H = AK1*AK3/(HIONC*CL)
RETURN
END

```



```

SUBROUTINE PROP (TEMPK,RHOLIQ,RHOAIR,RHOSTM,RHOATM,P,PAIR,PTOTAL,
1ETA,ETAAIR,ETAMIX,DIAIR,DIWTR,DIMIX,SMITNC,ETASOL,DLI2,DLCH3I)
C THIS CONTAINS THE PHYSICAL PROPERTY CALCULATIONS FOR SPRAY-2
C SOURCES FOR THE EQUATIONS USED ARE MAINLY KEENAN & KEYES THERMODYNAMIC PRO-
C PERTIES OF STEAM WILEY, 1936 (K&K) AND THE 3RD & 4TH EDITIONS OF THE CHEMI-
C CAL ENGINEERS' HANDBOOK MCGRAW-HILL 1950,1963 (PERRY3 OR PERRY4)
IMPLICIT REAL*8(A-H, O-Z)
C CALCULATION OF VAPOR PRESSURE OF WATER (K&K EQ (12) P 14)
X = 647.27 - TEMPK
TOP = 3.2437814 + (5.86226E-3)*X + (1.1702379E-8)*(X**3)
DENOM = 1. + (2.1878462E-3)*X
PHI1X = TOP/DENOM
PHI2X = (2.202585*X*PHI1X)/TEMPK
PHIP = DEXP(PHI2X)
P = 218.167/PHIP
C CALCULATE SP VOL OF LIQUID (EMPIRICAL EQ FIT TO DATA IN TABLE 3-46, PERRY4)
VLIQ = 1.0018 + 0.0007615*TEMPK - 293.161 + (3.219E-6)*((TEMPK -
1 293.16)**2)
RHOLIQ = 1./VLIQ
C SINCE THE MASS OF AIR IN THE CONTAINMENT IS FIXED RHOAIR IS INDEPENDENT OF T
RHOAIR = 0.001185
C CALCULATION OF STEAM DENSITY (K&K EQ(13) P 15)
TAU = 1./TEMPK
TAUSQ = TAU*TAU
G1TAU = 82.546*TAU - (1.6246E5)*TAUSQ
G2TAU = 0.21828 - (1.2697E5)*TAUSQ
G3TAU = 3.635E-4 - (6.768E64)*(TAU**24)
BZERO = 1.89 - 2641.6*TAU*(1.0**((80870*TAUSQ)))
B = BZERO + (BZERO**2)*(G1TAU*TAU*P) + (BZERO**4)*G2TAU*((TAU*P)
1**3) + (BZERO**13)*G3TAU*((TAU*P)**12)
V = (4.55504*TEMPK)/P + B
RHOSTM = 1./V
RHOATM = RHOAIR + RHOSTM
C CALCULATE VISCOSITY OF STEAM (K&K EQ(20) P 23)
TOP = (1.851E-5)*DSORT(TEMPK)
DENOM = 1. + 680.1*TAU
ETA0 = TOP/DENOM
F10FP = (P*.E-4)*(0.03103 - (3.65E-5)*P)
ETA = ETA0 + F10FP
C CALCULATE VISCOSITY OF AIR (PERRY3 EQ(2) P370)
ETAIR = (1.709E-4)*((TEMPK/273.16)**0.768)
C CALCULATE MIXTURE VISCOSITY (PERRY4 EQ(3-108) P 3-230)
PAIR = 1.*TEMPK/298.0
PTOTAL = P + PAIR
FRSTM = P/PTOTAL
FRAIR = PAIR/PTOTAL
TOP = FRSTM*ETA*4.242641 + FRAIR*ETAAIR*5.385165
DENOM = FRSTM*4.242641 + FRAIR*5.385165
ETAMIX = TOP/DENOM
C CALCULATE DIFFUSION COEFFICIENT OF IODINE IN AIR (PERRY4 EQ(14-57) P 14-20)
C SUBSCRIPT 1 = IODINE , 2 = AIR , 3 = STEAM
EPS1K = 550.
EPS2K = 97.
EPS3K = 363.
EPS12K = DSORT(EPS1K*EPS2K)
EPS13K = DSORT(EPS1K*EPS3K)

C EMPIRICAL FIT TO DATA IN TABLE 14-45 P 14-21 PERRY4
COLINT = 0.3674 + 0.3478*EPS12K*TAU
RAIR = 3.617
PI = 4.982
RIAIR = 0.5*(RI + PAIR)
FOFM = DSORT((1./254.) + (1./29.00))
B = (10.7 - 2.46*FOFM)*1.E-4
PFUNT = TEMPK**1.5
TOP = B*PFUNT*FOFM
DENOM = PTOTAL*(RIAIR**2)*COLINT
DIAIR = TOP/DENOM

```

C CALCULATE DIFFUSION COEFFICIENT OF IODINE IN STEAM

COLINT = 0.3674 + 0.3478*EPS13K*TAU

RW = 2.655

RIW = 0.5*(RI + RW)

FOFH = DSORT((1./254.) + (1./18.00))

B = (10.7 - 2.46*FOFH)*1.E-4

TC* = B*PFUNT*FOFH

DENOM = PTOTAL*(RIW**2)*COLINT

DIWTR = TOP/DENOM

C CALCULATE DIFFUSIVITY OF I IN AIR-STEAM, USING EQ(6) IN C.R. WILKE, CHEM ENG

C PROGR V46, PP95-104(1950) YA IS ASSUMED TO BE 0

DIMIX = 1./((FRAIR/DIAIR) + (FRSTM/DIWTR))

SMITNO = ETAMIX/(RM*ATM*DIMIX)

T = TEMPK - 273.16

C CALCULATE DIFFUSIVITY OF I2 IN WATER (PERRY4 PP14-21 TO 14-24)

C FOR WATER VISCOSITY CALCULATION SEE PERRY4 TABLE3-267 P 3-201

TFUNC = T - 8.435

ETAINV = 2.1482*(TFUNC + DSORT(8078.4 + TFUNC*TFUNC)) - 120.

ETASOL = 100./ETAINV

DLF = (7.4E-10)*TEMPOK*ETAINV*DSORT(2.6*18.00)

DLI2 = DLF/(71.5**0.6)

C CALCULATE CH3I DIFFUSIVITY IN WATER

DLCH3I = DLF/(62.9**0.6)

RETURN

END