

I. INTRODUCTION

In evaluating the acceptability of proposed sites for nuclear power reactors, and the design bases for engineered safety features, the DRL staff considers the potential radiological consequences of loss-of-coolant accidents. These evaluations are based on the suggested calculational model presented in TID-14844 regarding the fraction of the fission products contained in the reactor core which is assumed to be released into the containment atmosphere and available for leakage to the environment. In TID-14844 it is stated that

"In accidents of the 'maximum credible' type, it is usually assumed that the radioactive materials . . . would be dispersed in the coolant through melting or rupture of fuel elements and then find passage to the outer containment barrier through breaches in the coolant system At the same time, a certain amount of airborne fission products would be removed by such phenomena as absorption, deposition, plate-out and steam condensation within the reactor building or containment structure."

Specifically, for the model suggested in TID-14844 it is assumed that (1) 100% of the noble gases, 50% of the halogens, and 1% of the solids in the core fission product inventory are released into the containment, and (2) 50% of the iodines that are released into the containment are adsorbed onto internal surfaces of the reactor building or adhere to internal components, and are thus not available for leakage from the containment.

For plants in which the containment is equipped with a chemical additive spray system, the staff also calculates the amount of the halogens that would be removed from the containment atmosphere by the spray, based on the

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specific physical characteristics of the proposed system and containment. The assumption suggested in TID-14844 that 50% of the iodines released from the core are not available for leakage because of adsorption and adherence to surfaces (referred to herein as the "plateout factor") is an arbitrary assumption and not based on a detailed analysis of each plant, whereas the staff calculates the magnitude of the spray removal factor specifically for each plant.

In subsequent sections of this paper we discuss the general properties of iodine removal mechanisms, the specific model used by the staff in calculating the spray removal factor for iodines, the technical validity of the assumed plateout factor of two, and the technical basis for continued use of the assumed plateout factor of two in plants which also utilize chemical additive sprays as engineered safety features for removal of iodine from the containment atmosphere.

We conclude that the use of a plateout factor of two for iodine is conservative even for plants using chemical additive spray removal systems, and that the method used by the staff to calculate the spray removal factor is sufficiently conservative that the performance of actual systems will exceed that calculated.

II. IODINE REMOVAL (GENERALIZED MODEL)

The rate of depletion of the airborne iodine concentration by one or more iodine removal processes (such as sprays, filters, or plateout) is directly proportional to the residual airborne (gas phase) iodine concentration, thus

$$\frac{dc}{dt} = -\lambda c \quad [1]$$

where c = gas phase iodine concentration at time t
 t = time
 λ = iodine removal constant (proportionality constant)

The fraction of iodine remaining airborne at any time is obtained by integration of equation [1] over the time of operation. This gives

$$c = c_0 e^{-\lambda \Delta t} \quad [2]$$

where c = airborne iodine concentration at time t
 c_0 = initial airborne iodine concentration
 Δt = duration of operation of iodine removal process(es)

The reciprocal of the fraction remaining at any time, or the ratio of the initial to final airborne iodine concentrations, is designated as the decontamination factor (DF)

$$DF = \frac{C_0}{c} \quad [3]$$

For the purpose of calculating doses, the time-averaged mass of iodine available for release from the containment building with the engineered safety systems operative is used. This is obtained by integration of equation [2] over the appropriate time limits.

$$\begin{aligned}
 M &= L c_0 \int_0^t e^{-\lambda t} dt \\
 &= \frac{L c_0}{\lambda} [1 - e^{-\lambda t}] \quad [4]
 \end{aligned}$$

M = total mass of iodine available for leakage

where L = containment leakage fraction per unit time. The mass of iodine which would be available for leakage in the absence of a removal system is

$$\begin{aligned}
 M' &= L c_0 \int_0^t dt \\
 &= L c_0 t \quad [5]
 \end{aligned}$$

The dose reduction factor (DRF) for iodine given by the engineered safety systems is the ratio of the mass which would be released in the absence of a removal system to the mass which would be released with the system operative.

$$\begin{aligned}
 \text{DRF} &= \frac{M'}{M} \quad [6] \\
 &= \frac{\lambda t}{1 - e^{-\lambda t}}
 \end{aligned}$$

The above equations for the overall decontamination factor and for the time-averaged dose reduction factors are both based on the conservative assumption of a puff release; that/ the mass of iodine released is present initially at its maximum concentration and decreased by the time-dependent removal mechanisms. The effect of radiological decay is treated separately in the dose calculation.

The removal half-life for a specific mechanism is defined simply as that period of time required to reduce the airborne iodine concentration to

one-half its initial value; that is, the time required for

$$\frac{c}{c_0} = 0.5 \quad [7]$$

The removal half-life and the iodine removal constant can be related by use of equation [2], and

$$\frac{c}{c_0} = 0.5 = e^{-\lambda t_{1/2}} \quad [8]$$

and

$$t_{1/2} = 0.693 / \lambda$$

where

$$t_{1/2} = \text{removal half-life}$$

III. CHEMICAL ADDITIVE SPRAY SYSTEMS

The model for iodine depletion in the containment considered in consideration of TID-14844 does not include/chemical additive sprays, charcoal adsorbers, or similar engineered safety features. Most current reactors, however, do employ iodine reduction systems of this type and the staff has developed conservative models for evaluating these.

The equation used to calculate the iodine removal constant for chemical additive spray systems, developed by Griffiths*, is

$$\lambda_s = \frac{6 v_D f t}{V_c d} \quad [9]$$

Where

v_D = overall iodine deposition velocity into spray drop

f = flow rate of spray

t = $\frac{h}{u}$ = $\frac{\text{fall height}}{\text{average drop velocity}}$ = drop residence time

V_c = effective containment volume

d = drop diameter

* V. Griffiths: The Removal of Iodine from the Atmosphere by Sprays
U. K. Atomic Energy Estab., AHSB(S) R45 (1963)

For the Indian Point 3 reactor, both the AEC staff and the applicant have based their calculations of spray effectiveness on the Griffiths model. The staff calculates a spray iodine removal constant of 4.9 hr^{-1} . The applicant calculates a value of 32 hr^{-1} . The differences in the calculated removal constants arise because the staff uses more conservative values for several of the parameters in the above equation to allow for possible system degradation and uncertainties in the parameters used.

The following is a brief discussion of the specific numerical differences between the values for each term in the Griffiths equation which the regulatory staff believes appropriate for the Indian Point 3 reactor and those used by the applicant.

a. Deposition Velocity

The different values used by the applicant and by the staff for this term are the result of differences in the assumed magnitude of the iodine partition factor between the liquid and gaseous phases, with a consequent difference in the uptake velocity into the drop. The overall deposition velocity is given by

$$\frac{1}{v_D} = \frac{1}{v_G} + \frac{1}{k_L H} \quad [10]$$

where

v_D = overall deposition velocity

v_G = gas film deposition velocity

k_L = liquid film transfer coefficient

H = iodine partition factor

For very large values of the partition factor the second term on the right hand side of the equation becomes negligible and the overall deposition

velocity is approximately equal to the gas film deposition velocity. As the numerical value of the partition factor decreases (lower solubility) the overall deposition velocity is decreased and the uptake velocity into the drop becomes the controlling factor (liquid film resistance).

The applicant has made the assumption that the deposition velocity (transfer velocity of iodine from the atmosphere into the liquid) is controlled solely by the relatively rapid exchange across the stagnant gas film surrounding the spray drops. ^{This assumption is} valid only if the iodine partition factor is very large.

For the specific case of a spray solution using sodium hydroxide alone as an additive, the staff has adopted a value for the partition factor of 3×10^3 , calculated from the theoretical work of Eggleton* and based on a total iodine concentration equal to a release of 25% of the core iodine inventory and a spray solution pH of 8.0 at a temperature of 100°C (212°F).

Using the above method, the staff has calculated an overall iodine deposition velocity of 4 cm/sec for the Indian Point 2 case, compared with a value of 7 cm/sec used by the applicant.

b. Flow Rate

The evaluations by both the applicant and the staff are based on the rated flow rate through only one of the two subsystems. (It is assumed that the other subsystem fails, in accordance with the single failure criterion.) The applicant has used the full flow rate while the staff

* A.E.J. Eggleton: A Theoretical Examination of Iodine - Water Partition Coefficients. U. K. Atomic Energy Agency, AERE-R-4887 (1967)

has arbitrarily reduced this value by 10% to allow for local fluid density and viscosity variations, for possible system damage, and for possible system design and/or construction faults.

c. Fall Height

The applicant has used the minimum distance from the spray headers to the operating deck floor as the average free fall height for all spray drops. The staff has reduced this value by 15% to compensate for the smaller fall height over a portion of the area caused by intrusion of the pressurizers and other equipment into this volume as well as to include consideration of those drops which strike either walls or interior surfaces.

d. Containment Volume

The uncertainties^{associated} with this parameter are primarily those concerned with uniformity of distribution and of mixing in the gas phase. The applicant has used the entire free volume of the containment, neglecting these effects. The staff has used a value for containment volume which is 30% less than the numerical value used by the applicant, based on: results reported for the compartmented CSE installation (30,000 cu. ft.) at Battelle Northwest, where a standard deviation of 24% from the mean concentration was reported for a well-mixed atmosphere; on the results of theoretical modeling; and on consideration of the effect of local gas phase depletion due to "channel effects" by successive drops.

e. Drop Diameter

There is a large uncertainty associated with the selection of the appropriate diameter of the spray drops because:

- (1) No measurements of drop diameters and drop size spectra have been made for the installed nozzles under simulated post-accident conditions.
- (2) Drop collision and coalescence are predicted for nearly 20% of all drops on the basis of cloud physics models, yielding both a larger effective diameter for the resultant drops and a skewed drop size distribution.
- (3) An increase in drop diameters may occur as a result of steam condensation. The effect is an additional increase of the diameter of the larger drops by from 5 - 10%.

The applicant has used a surface mean drop diameter of 1000 microns, based on an experimental size determination with water at ambient air temperature and pressure and with a minimum pressure drop across the system of 30 lbs/sq. inch. The staff used a surface mean drop diameter of 1350 microns as released from the nozzle, based on data from both Oak Ridge and Battelle Northwest/indicates ^{which} ^{that} this value is more appropriate for consideration of the nozzles used, and further modified by/the above uncertainties in drop coalescence and steam condensation. The combined uncertainties yield a maximum expected surface mean drop diameter of 2000 microns, or twice the value stated by the applicant.

IV. IODINE REMOVAL BY PLATEOUT

The quantity of fission products available for leakage from a reactor containment following a loss-of-coolant accident is reduced by the action of removal mechanisms on the fission products which have been released from the core. One of these possible removal mechanisms is irreversible deposition on internal surfaces ("plateout"). Molecular iodine passes from the gaseous phase directly into the solid phase without a liquid transition phase at a relatively low temperature and is therefore deposited readily on a variety of surfaces in the containment. The plateout mechanism is a major inherent removal process for iodine in the containment. The staff has reviewed the experimental and theoretical information available on the mechanism of iodine plateout, and has evaluated the factors affecting the magnitude of the reduction by plateout of the iodine released from the fuel following a loss-of-coolant accident (LOCA).

In the calculational method suggested in TID-14844, instantaneous plateout of 50% of the halogens released is assumed, so that this fraction never becomes available for leakage. The staff has attempted to assess the validity of this assumption, and the degree of conservatism associated with it, by use of a time-dependent plateout model.

Iodine plateout, or transport to reactor surfaces with subsequent retention or washdown, can occur in several regions of the facility. First, iodine removal may occur in the core region, either by deposition on fuel cladding or core internals or by direct steam transport to surfaces. In this case the removal may

be considered to occur instantaneously, since the fraction of iodine removed does not reach the containment. Although there is considerable evidence that some iodine retention would occur by these mechanisms,

because of uncertainties as to the magnitude of these effects, the staff conservatively assumes for purposes of this discussion that no plateout or deposition occurs in the core region. Next, iodine deposition may occur during transport from the core to the primary containment. This is very likely, since progressively cooler surfaces are encountered. However, again because of uncertainties as to the magnitude of this effect, the staff conservatively assumes here that no plateout occurs during this phase. Finally, plateout may occur on the various surfaces in the primary containment by a time-dependent mechanism, in direct competition with iodine removal by engineered safety systems. It is only this final stage of iodine plateout which is considered here.

The results of considerable experimental and theoretical work concerning the plateout of iodine under LOCA conditions have been published. Plateout occurs in the primary containment under conditions which, in general, are amenable to experimental investigation. Reasonably accurate estimates of its magnitude and time dependence are possible. From these

data, extrapolation of the experimental results to conditions not specifically covered by these experiments (for example, to systems where the effects of plateout and spray removal are combined) is possible.

The principal experimental work on iodine plateout and deposition under simulated reactor accident conditions can be divided into three types:

- (1) Small-scale laboratory tests have been performed on a large number of different types of materials and on various surface coatings at the Battelle Memorial Institute and summarized in Reports BMI-1863 (Fission Product Deposition on Primary Surfaces), BMI-1865 (Fission Product Deposition on Containment System Surfaces) and BMI-1874 (Development of Reactive Coatings).
- (2) Applied engineering tests have been conducted in the Containment Research Installation (CRI) at the Oak Ridge National Laboratory and at the Contamination-Decontamination Experiment (CDE) facility at Idaho Nuclear Corporation. The CRI equipment consists of a fission product release and containment facility, the latter with approximately 1000 gal capacity and capable of being fitted with liners of various materials. Results are reported in the publications of the ORNL Nuclear Safety Program. The CDE facility is designed to simulate release from an unperturbed melt accident, with transport and plateout in an 86 ft³ vessel under saturated steam conditions.

Initial results were reported in IN-1172 (Fission Product Behavior Under Simulated Loss-of-Coolant Accident Conditions).

- (3) Relatively large-scale experiments have been performed at the Containment Systems Experiment (CSE) facility at the Battelle Northwest Laboratory. The results are summarized in Report BNWL-943 (Fission Product Transport by Natural Processes in Containment Vessels). The CSE system has a volume of greater than 20,000 ft³. The containment atmosphere, surfaces and convection patterns of a reactor containment can be simulated.

Most experiments which investigated natural deposition from either an air atmosphere or a saturated steam-air atmosphere (all in the absence of sprays) have yielded values of the initial iodine plateout half-life in the range from 2 to 15 minutes. For a variety of release conditions in comparison tests of iodine simulants at the Battelle Northwest Laboratory, plateout half-lives ranging from about 3 to 15 minutes were observed, with the longer times for extremely large iodine concentrations. In meltdown experiments in the CSE an initial/iodine half-life of eight minutes was reported. Extrapolation of these results to a large PWR with different surface area to volume ratio yields anticipated iodine plate out half-lives ranging from 10 to 20 minutes if only the area of the outer walls is considered available for deposition, and much shorter half-lives if all the available surface area is included (e.g., steam generators and other equipment, piping, compartment walls, and floor surfaces).

The actual rate of deposition of iodine on containment surfaces depends on a number of variables, including the containment geometry, the nature of specific containment surfaces, the containment atmosphere from which deposition occurs, the specific driving forces (e.g., temperature and concentration gradients, and steam flux), and the gas phase halogen concentration.

The overall plateout phenomenon can be divided into two successive processes: (1) transport to surfaces and (2) adsorption on surfaces. As stated above, the staff analysis conservatively assumed that plateout only occurs in the primary containment. Here we further assumed that transport to surfaces occurs only by a natural convection process and have neglected the large additional transport driving force which would be provided by the flow of steam to the colder surfaces of the containment building.* This transport by flowing steam would actually be expected to account for a major portion of the iodine transfer to surfaces under actual post-LOCA conditions, adding further conservatism to the staff model.

The adsorption process in the primary containment building has been evaluated as a function of both surface temperatures and type of material. The iodine deposition rates used for the containment surfaces are conservatively chosen as representative values for specific materials,

* The calculational model used by the staff is closely analogous to that used in BNWL-943 "Fission Product Transport by Natural Processes in Containment Vessels."

maximum expected air temperatures, and airborne iodine concentrations typical of those expected in the containment following a LOCA.*

For the large containment volumes typical of modern FWR plants, we have calculated that the plateout mechanisms would reduce the airborne iodine concentration to half its initial value within 10 to 20 minutes. This value should be considered as an upper limit because of the various factors of conservatism introduced, and a more realistic evaluation model involving steam transport and rapid removal would yield a plateout half-life of two minutes or less. Using the time-dependent plateout model described above, the airborne iodine concentration available for leakage averaged over the initial two-hour period following a LOCA is less than that which is obtained by applying the assumption of an instantaneous plateout factor of two suggested in TID-14844. Our calculation of the time-averaged total reduction in iodine available for leakage achieved by plateout alone, using appropriately conservative parameters, yields a reduction factor which varies from about four to six for typical large containments. This value varies in proportion to the containment volume. The corresponding iodine decontamination factors (defined in Section II) due to a plateout effect of this magnitude range from 10 to 100 for the initial two-hour period following a release. The total reduction of airborne iodine possible by plateout is limited to decontamination factors of about 100 because the available surfaces become saturated with iodine.

* Experimental investigations studying the plateout behavior of iodine on surfaces under laboratory conditions have been completed at both Idaho Nuclear Corporation and at the Battelle Memorial Institute (BMI-1865).

V. COMBINED IODINE REMOVAL PROCESSES

For cases in which two or more iodine removal processes are operating simultaneously, two questions arise: (1) Does the action of any mechanism affect the others so as to alter any of the assumptions used in estimating the magnitude of the individual processes operating independently? (2) What is the overall combined effect on iodine reduction of two or more removal mechanisms operating simultaneously?

The staff has considered the specific case of the interaction of sprays and plateout in terms of overall iodine removal. In terms of the effect of the sprays on plateout, the sprays may decrease the flow of steam to surfaces by increasing condensation but would also increase the turbulent (convective) flow and mixing characteristics within the containment. However, in the evaluation of the conservatism of the plateout assumption (see Section IV), the contribution of steam transport already has been neglected completely and the treatment of convective flow does not include the effect of turbulence induced by the sprays. Therefore, the rate of iodine transport to surfaces calculated by the techniques described in Section IV should always be smaller than the actual value, even during a period when containment sprays are in operation. Iodine removal from the gas phase ^{by plateout} is therefore expected to occur at a rate equal to or greater than the conservatively calculated values given in Section IV.

Adsorption of iodine on surfaces is generally enhanced by addition of a water film. For surfaces wetted by chemical additive sprays, the

reduction of liquid film resistance would further increase the uptake and transport velocity to the surface of the material. Therefore, deposition rates for iodine on surfaces wetted by spray would be expected to be much larger than for the corresponding surface either dry or wetted only by steam condensation.

The removal rate of iodine from the gas phase by chemical additive spray systems would be expected to be independent of the plateout effect, since the spray removal process operates in the bulk phase and is only dependent on the airborne iodine concentration.

Finally, we will consider the combined effect of several iodine removal processes operating simultaneously. If each of these can be considered independent and the rate only a function of the gas phase iodine concentration, then

$$\frac{dc}{dt} = -c \sum_i \lambda_i \quad [41]$$

where λ_i = iodine removal constant for mechanism i

and the overall iodine removal constant is equal to the summation of the several constants. This is applicable only to a well-mixed atmosphere, such as would be expected in the containment following a LOCA.

The fraction of iodine removed from the gas phase by each different mechanism is proportional to its removal (rate) constant, and the total iodine reduction for two or more simultaneous processes is always greater than that obtained by one mechanism alone. Therefore, when the rate constants for both plateout and spray removal are of comparable magnitude,

the plateout process will actually remove half or more of the gas phase iodine. On the other hand, when the spray removal constant becomes very much larger than the plateout deposition constant, nearly all of the gas phase iodine reduction is due to the sprays alone and very little due to plateout. A comparison of the overall effect of two different combined mechanisms is most logically based on the overall iodine reduction factors achieved over a specified time period. On this basis, the staff has compared the effect of

1. A time-dependent plateout mechanism, in conjunction with sprays, operating on the total release fraction specified in TID-14844 (50% halogens) with
2. An instantaneous plateout factor of two, in conjunction with the identical spray system, and operating on the same initial release.

The comparison for the Indian Point 3 reactor, applying the data used by the staff specifically for this unit, is given in Section VI (following).

When the spray removal constant becomes very large ($\lambda_s > 15 \text{ hrs}^{-1}$), as in the model proposed by the applicant for the Indian Point 3 reactor, the contribution of the plateout effect to overall iodine removal is very small. Therefore, if a very large spray removal constant were to be assumed, then the plateout should be neglected and the total iodine reduction calculated on the basis of spray reduction alone operating on the entire halogen release fraction.

As noted in Section IV, a time-dependent model of plateout in the absence of concurrent iodine removal by chemical additive spray systems has been shown to yield two-hour iodine reduction factors greater than two on a conservative basis. Combined iodine removal by both plateout and sprays operating simultaneously on a time-dependent basis has also been shown to exceed the iodine reduction calculated by the simplified model of an instantaneous plateout factor of two, provided that the spray removal constant is of comparable magnitude to that for plateout. This is the basis for the staff calculations on Indian Point 2. On the other hand, if very large spray removal constants (very short half-lives) are assumed, as in the applicant's calculation, the major portion of the iodine removal is due to sprays, and use of the plateout factor of two would not be warranted.

VI. COMPARISON EXAMPLES

In the following two examples we compare the overall iodine reduction for the Indian Point 3 reactor calculated using the present staff assumptions, including the plateout factor suggested by TID-14844, with that calculated by applying a time-dependent plateout model. In both cases, the spray removal constant used is that calculated by the staff model discussed in Section III above. The two-hour iodine reduction factor calculated by the more realistic time-dependent plateout mechanism slightly exceeds that calculated for the TID-14844 model for instantaneous plateout. Therefore, the currently assumed model of instantaneous plateout of 50% of the airborne.

iodine concentration represents a conservative model, especially in view of the very conservative assumptions used in deriving the removal constant for time-dependent plateout. Both examples assume an instantaneous puff release and operation of the spray system over the entire time period.

A. Current Model Used by Staff for Site Evaluation Purposes for Indian Point 3

Assumptions:

1. 50% of core iodine inventory is released (TID-14844).
2. 50% of the released iodine is removed instantaneously by plateout (TID-14844).
3. 25% of the core iodine inventory is initially available in airborne form in the containment.
4. 10% of this initial airborne iodine concentration is in the organic iodides form of / (nonremovable) (2.5% of core iodine inventory).
5. The chemical additive spray system reduces the inorganic iodine fractions (22.5% of core iodine inventory) with a removal constant $\lambda_s = 4.9 \text{ hr}^{-1}$.

Results:

1. The time-averaged gas phase (airborne) iodine concentration, including both inorganic and nonremovable iodine species, for the initial two-hour period is 4.8% of the entire core inventory.

2. On the basis of a total initial airborne iodine concentration of 25% of the core inventory, the two-hour iodine reduction factor is 5.2.

B. Model Using Finite Plateout Time

Assumptions:

1. 50% of core iodine inventory is released (TID-14844).
2. The airborne iodine is removed by plateout with a half-life of 10 minutes (removal constant $\lambda_p = 4.3 \text{ hrs}^{-1}$) for a duration of 30 minutes.*
3. 5% of the initial airborne iodine concentration is in form of nonremovable species (2.5% of core iodine inventory and same total quantity as in example above).
4. The chemical additive spray system reduces the inorganic fraction simultaneously with plateout, with a removal constant $\lambda_s = 4.9 \text{ hrs}^{-1}$.

Results:

1. The time-averaged gas phase (airborne) iodine concentration, including both inorganic and nonremovable iodine species, for the initial two-hour period is 4.5% of the entire core inventory.

*After 30 minutes, the saturation of available surfaces is assumed to limit further plateout (see p. 15 above).

2. On the basis of a total initial airborne iodine concentration of 50% of the core inventory, the two-hour iodine reduction factor is 11. (For comparison, this is equivalent to an iodine reduction factor of 5.5 based on 25% of the core inventory.)



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	DATE	discussion of the Indian Point 3 spray and plate-out question. Although not polished,

TO (Name and unit) HK Shapar EG Case LD Low	INITIALS	REMARKS we would appreciate review, discussion and
	DATE	<u>early comment.</u>

TO (Name and unit)	INITIALS	REMARKS
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FROM (Name and unit) PAMorris	REMARKS

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