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REACTOR TECHNOLOGY MEMORANDUM -- FISSION PRODUCTS - IODINE
FORMATION AND REMOVAL IN POWER REACTOR FACILITIES

The enclosed RTM sets forth proposed DRL evaluation guidelines for iodine formation and removal in power reactor facilities. I recommend that these guidelines be factored into our safety evaluations. Further, I would appreciate receiving any comments you care to make as a result of reviewing the RTM by November 4, 1968, in order that necessary revisions can be made promptly.

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Enclosure:
RTM - Fission Products

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REACTOR TECHNOLOGY MEMORANDUM

FISSION PRODUCTS - IODINE

FORMATION AND REMOVAL IN

POWER REACTOR FACILITIES

I. INTRODUCTION

The formation of radioactive iodine and its release characteristics under potential accident conditions as well as the engineered capability for its removal from the containment atmosphere by mechanical and chemical means are under continuing study by the Regulatory staff. The evaluation guidelines presented in this memorandum are recommended for use by DRL in the evaluation of power reactor facilities until such time as changes are made on the basis of further results from continuing research and development. These guidelines are generally consistent with past practice.

II. EVALUATION GUIDELINES

(1) Total Available Iodine in Containment

The percentage of iodine available for release from the containment atmosphere to the environs should be assumed as 25% of the entire core inventory at end of core life, in accordance with assumptions in TID-14844.

(2) Organic Iodide in Containment Atmosphere

The average percentage of all iodine isotopes present as methyl iodide (or higher homologues) within the containment atmosphere for both the two hour period and thirty day period subsequent to the design basis loss of coolant accident should be assumed to be 10% of the iodine available for leakage from the containment atmosphere of a single barrier system and 8% of iodine available for leakage from the containment atmosphere of a pressure suppression system.

(3) Particulate-Associated Iodine in Containment Atmosphere

The average fraction of all iodine isotopes within the primary containment atmosphere, subsequent to the design basis loss of coolant accident, that is associated with airborne particulate matter should be assumed to be 5% of the iodine available for leakage from the containment atmosphere.

(4) Iodine Removal - Single-pass Filter System

Credit given in terms of an iodine reduction factor for an acceptably designed single-pass filter system for removal of

iodine isotopes (other than those associated with particulate matter and those in the methyl form) from a stream of air taken from a secondary containment source and passed through the filter system should not exceed a factor of ten, except that a factor of up to twenty may be allowed for systems designed to rigid standards supported by a program for extensive testing and surveillance.

(5) Iodine Removal - Containment Recirculation Filter System

Credit given in terms of an iodine reduction factor for an acceptably designed containment recirculation filter system for removal of iodine isotopes (other than those associated with particulate matter and those in the methyl form) from the containment atmosphere shall be calculated by the method given in Appendix A, Part IV. Claims exceeding a factor of four should be referred to RT for evaluation.

(6) Iodine Removal - Containment Spray System

Credit given in terms of an iodine reduction factor for an acceptably designed containment spray system for removal of iodine isotopes (other than those associated with particulate matter and those in the methyl form) from the containment atmosphere shall be calculated by the method given in Appendix A, Part V. Claims exceeding a factor of four should be referred to RT for evaluation.

(7) Iodine Removal - Combination of Containment Spray and Single-pass Filter

Credit given in terms of an iodine reduction factor for a combination of acceptably designed containment spray and single-pass filter systems for removal of iodine isotopes (other than those associated with particulate matter and those in the methyl form) from a stream of air taken from a secondary containment source and passed through the filter system should not exceed a factor of twenty.

(8) Iodine Removal - Combination of Containment Recirculation Filter & Containment Spray System

Credit given in terms of an iodine reduction factor for a combination of an acceptably designed containment recirculation filter system and containment spray system for removal of iodine isotopes from the containment atmosphere should not exceed a factor of six. All such systems should be referred to RT for evaluation.

III. APPENDIX

Appendix A provides a discussion of the bases for the evaluation guidelines established in this memorandum.

APPENDIX A

BASES FOR EVALUATION GUIDELINES

I. ORGANIC IODIDE FRACTION IN CONTAINMENT ATMOSPHERE

A. THEORETICAL ANALYSIS

A theoretical study performed by the Division of Reactor Licensing of the various organic iodide formation mechanisms [1-5] (in terms of percentage of iodine available for leakage from the containment building and based on TID-14844 release and plateout assumptions [6]) indicates that less than 0.5% would be derived from the fuel, a maximum of about 5% could be formed in the pressure vessel by gas phase reaction, and an additional 1-5% formed by a time dependent surface reaction in the containment building. Therefore, on the basis of these calculations, the maximum percentage of methyl iodide which could be produced under optimum conditions would not be expected to exceed 10% of the total iodine in the containment building. Details of calculations will be published separately.

The contributing formation mechanisms and applicable values are:

1. Formation in Fuel and Release at Meltdown.

The calculation is based on the joint probability of combination of the volume fractions of the core inventory fission product iodine and 300 ppm carbon. This yields 0.12% of the core inventory iodine or, assuming a subsequent four fold depletion of the inorganic fraction, about 0.5% of the iodine ultimately available for leakage from the containment.

2. Gas Phase Reaction Near Core.

The calculation is based on γ -induced formation of methane from an assumed two percent carbon dioxide, radiolytic and thermal formation of methyl radicals and a non-equilibrium quasi-kinetic treatment of reactions with available molecules to produce both methyl iodide and methane according to published rate constants. An estimated 2% of the released iodine may be converted to methyl iodide by this mechanism or, assuming a subsequent two fold depletion of the inorganic fraction, about 4% of the iodine ultimately available for leakage from the containment.

3. Gas Phase Reaction in Containment.

These calculations are based on the simultaneous equilibrium thermal formation of methyl iodide in a steam-air atmosphere and on the

equilibrium radiation formation for an absorbed dose of 10^6 rads for iodine in the gas phase in the containment. Both mechanisms would probably convert less than 0.5% of the iodine in the containment to the organic form. Rapid depletion of the airborne iodine probably makes these mechanisms important only for about a three hour period.

4. Surface Formation.

A mechanism involving deposition of molecular iodine on surfaces, conversion to the organic form, and subsequent rapid desorption has been postulated. A deposition rate constant of 1×10^{-1} cm/sec has been used, representing an approximate upper limit for common reactor construction materials. A slow removal constant of 1 hr^{-1} was assumed/or the depletion of the airborne iodine by all other mechanisms. The average rate of methyl iodide formation was calculated for bare stainless steel in a steam-air atmosphere and a maximum iodine surface concentration, using a rate constant of $1.5 \times 10^{-3} \text{ } \mu\text{g}/\text{m}^2 \cdot \text{min}$. The quantity of organic iodide formed by this mechanism is cumulative, rising to a maximum of 0.5% of the containment iodide concentration within three hours after release and dropping gradually to about 0.1% after 12 hours. In the following days the cumulative effect would continue. It is estimated that the maximum quantity might rise to as much as 10% from all sources during the 30 day post accident period.

A summary of the cumulative percentage of methyl iodide as a function of total iodine concentration in the containment building and based on the above theoretical model is given in Table 1.

B. EXPERIMENTAL RESULTS

Laboratory experiments on release from irradiated UO_2 , or with simulants, have produced organic iodide fractions ranging from less than 1% to approximately 4% under conditions representative of those which prevail following a loss-of-coolant accident. Higher values have been obtained under unusual conditions not considered typical of post-accident conditions. These include:

1. High H_2 and low I_2 concentration
2. Unrealistically high methane concentration
3. Very high air temperatures (above 1000°C)
4. CO_2 gas cooled reactor atmospheres.

TABLE 1

ORGANIC IODIDE FORMATION

Hourly Formation of Organic Iodide as Fraction of Total Iodine in Containment

Time (hrs)	Formation Mechanism				Total % CH ₃ I
	Fuel	Gas Phase		Surface*	
		Reactor	Containment		
0	0.5	4			4.50
1			0.2		4.70
2			0.2	0.3	5.20
3			0.1	0.5	5.80
4				0.5	6.30
5				0.4	6.70
6				0.3	7.00
7				0.3	7.30
8				0.3	7.60
9				0.3	7.90
10				0.2	8.10
11				0.2	8.30
12				0.1	8.40
12 - 720				0.1 - 0.0	10.00 max

*All values are given for typical PWR. Reduce surface formation values to 0.7 of the above values for typical BWR.

TABLE 2

SELECTED REFERENCES ON ORGANIC IODIDE FORMATION

<u>Reference</u>	<u>Experimental Conditions</u>	<u>Percent Organic Iodide</u>
ORNL-3776 Semiannual Report December 1964	CMF: simulatant, I-131 tracer + I-127 ($2\text{mg}/\text{m}^3$) 1. Air 30 psia 4 hr 2. 50% steam-air 40 psia 4 hr 3. 50% steam-air 42 psia 18 hr 4. 50% steam-air 40 psia 5 hr organic contamination 5. 50% steam-air 44 psia 5 hr 0.25% CH_4 added 6. 50% steam-air 44 psia 18 hr 0.25% CH_4 added 7. 50% steam-air 44 psia 9 hr 0.25% CH_4 added	0.6% 1.0% 2.4% 3.2% 5.0% 1.0% 42.0%
ORNL-3843 Semiannual Report June 1965	CMF: 1. UO_2 7000 Mw/T melted in air SS clad 50% steam-air 40 psia 3.5 hr 2. simulatant 7.2 $\text{mgI}_2/100\text{g}$ UO_2 melted in air SS clad 50% steam-air in tank 27 psia 5.5 hr 3. simulatant - UO_2 mixture melted in air Zr clad 50% steam-air in tank 27 psia 5 hr 4. simulatant - UO_2 mixture melted in steam-He Zr clad 50% steam-air in tank 27 psia 5 hr TREAT: Zr clad UO_2 melting by transient 1000 psia steam	0.6% 2.6% 1.4% 1.6% 2.5%
ORNL-3915 Semiannual Report December 1965	CMF: UO_2 7000 Mw/T melted in steam-air SS clad 50% steam-air 29 psia 3 hr Flow Exp: I_2 1 mg/m^3 2 atm 100° C RH=90%	1.2% 1.7%

TABLE 2 (continued)

<u>Reference</u>	<u>Experimental Conditions</u>	<u>Percent Organic Iodide</u>	
ORNL-4071 Annual Report December 1966	CMF:		
	UO ₂ 1000 Mwd/T melted in steam-air Zr clad		
	50% steam-air 29 psia 3 hr	1.2%	
	TREAT:		
	underwater melting of Zr clad UO ₂ by transient	0.005%	
	CRI:		
	I ₂ release under ambient conditions 30 psig		
	1200 ft ³ SS tank 22 hr	0.7%	
ORNL-4228 Annual Report December 1967	ORR:		
	UO ₂ melted airflow for 20 min	initial	final
	5 hr aging at 35° C	2.6%	4.0%
	at 103° C	3.0%	3.6%
	NSPP:		
	800 mc I-131 + 400 mg I-127		
	1300 ft ³ 70 hr continuous agitation	2.0%	
	CRI:		
	I-130 SS liner 22 hr	3.5% (max)	
	HI-130 SS liner 22 hr	3.3% (max)	
BNL-11329 April 1967	UO ₂ into steam 1000°-1300° C	10.1%	
	UO ₂ into steam + hydrogen 1000°-1300° C	16.0%	
AEC-CONF-650407 Int. Symp. on Fiss. Prod. Release and Transport under Accident Conditions April 1965	Zr clad UO ₂ (0.5-100 Mwd/T) heated to can melting, some fuel melting, released to steam-air atm	0.4-3%	
ORNL-TM-1864 Nucl. Safety Report March-April 1967	In-pile, Zr clad UO ₂ (melt)	3.59-4.19%	

TABLE 2 (continued)

<u>Reference</u>	<u>Experimental Conditions</u>	<u>Percent Organic Iodide</u>
ORNL-TM-1986 Nucl. Safety Report July-August 1967	In-pile, Zr clad UO_2 (melt) NSPP: run 19; I_2 + dry air (stirred)	3.2% (max) 2.0%
BNWL-581 March 1968	ADF: UO_2 (0.35 Mwd/T) and simulants heated to melting, steam or steam-air sweep gas	0.1-2%
TRG Report 956 (W) UKAEA May 1965	UO_2 (150 Mwd/T), 3-47% I_2 release CO_2/CO + 1000 ppm CH_4 CO_2/CO CO_2/CO sat H_2O	20.0% 10.0% 4.0%

Key to Experimental Facilities

- CMF* (Containment Mockup Facility): 180 l SS vessel,
up to 45 psia steam-air atmosphere
- NSPP (Nuclear Safety Pilot Plant): 1350 ft³ SS vessel
designed to model fission product transport
- CRI (Containment Research Installation): 30 gal primary
simulator tank, 1000 gal containment vessel
- ADF (Aerosol Development Facility): 1.54 m³ coated steel
vessel and 0.9 m³ SS vessel

*The data reported out of this facility have been adjusted (here)
by a factor of two (2) to conform with the TID-14844 assumption
regarding plateout.

Ten percent, therefore, represents a conservative upper limit for the organic fraction of the iodine airborne in the containment under conditions considered typical for a loss-of-coolant accident in a water reactor and includes a margin of conservatism to account for the maximum formation rates by several different mechanisms. Table 2 summarizes the pertinent references.

II. PARTICULATE-ASSOCIATED FRACTION IN CONTAINMENT ATMOSPHERE

An estimate has been made of the maximum weight of particulate-associated iodine which can remain airborne (7-10). The conditions for homogeneous nucleation of iodine particles from the vapor phase have been shown to be unfavorable in the plenum (11) because critical saturation limits are not exceeded. Therefore, it is assumed that all particulate-associated iodine is adsorbed on surfaces of other materials as a monomolecular film with an accommodation coefficient of unity. An average particle radius of 0.2μ is assumed, because this represents the approximate size of minimum settling velocity. A theoretical maximum of 5% of the iodine available for leakage from the containment could be adsorbed on particulate surfaces. This is in good agreement with several experimental results.

A. THEORY

Assumptions:

1. Total of 10^7 particles/cm³ aerosol with $r_{\text{mean}} = 0.2 \mu$
2. Each iodine molecule occupies 50 \AA^2 surface
3. Only iodine is adsorbed on surfaces

Calculations:

1. Surface/particle

$$S = 4\pi r_m^2 = 12.57 r_m^2 = 12.57 (2 \times 10^{-5})^2 \\ = 5 \times 10^{-9} \text{ cm}^2$$

2. Total surface available/cm³

$$S_v = S(\text{particles/cm}^3) \\ = 5 \times 10^{-9} \text{ cm}^2 \cdot 10^7 \text{ cm}^{-3} \\ = 5 \times 10^{-2} \text{ cm}^2/\text{cm}^3$$

3. Total particle surface in containment

$$\begin{aligned} S_t &= S_v \cdot V \quad (V = 4 \times 10^{10} \text{ cm}^3 \text{ for typical PWR}) \\ &= (5 \times 10^{-2} \text{ cm}^2/\text{cm}^3)(4 \times 10^{10} \text{ cm}^3) \\ &= 2 \times 10^9 \text{ cm}^2 \end{aligned}$$

4. Maximum possible number of iodine molecules adsorbed

$$\begin{aligned} N &= \frac{S_t}{A} \\ &= 2 \times 10^9 \text{ cm}^2 / 50 \times 10^{-16} \text{ cm}^2/\text{molecule} \\ &= 4 \times 10^{23} \text{ molecules} \\ &= 0.65 \text{ gm. moles } I_2 \text{ adsorbed} \end{aligned}$$

5. Maximum possible weight of adsorbed iodine

$$\begin{aligned} W &= (\text{no. gm. moles})(\text{molecular weight}) \\ &= (0.65)(254 \text{ g}) \\ &= 165 \text{ g iodine} \end{aligned}$$

6. Total weight of iodine available in containment

$$\begin{aligned} W_T &= \text{core inventory}/4 \\ &= 3500 \text{ g} \end{aligned}$$

7. Maximum percent iodine associated with particulates

$$\begin{aligned} \text{Percent} &= \frac{165}{3500} \times 100 = 5\% \end{aligned}$$

III. SINGLE PASS FILTER SYSTEM

A single pass filter system is located outside the containment building and is not required to handle the bulk of the fission product release. It only filters the out-leakage from the containment building. Therefore, a time dependent reduction factor is not applicable for this case and only the effluent cleanup factor is evaluated.

The overall iodine reduction factor (IRF) for a single pass filter system is taken as the efficiency for the fraction of removable iodine, reduced by the sum of the fractional bypass flow and fraction of non-removable iodine.

A. AVERAGE CASE

For an average system, a 95% removal efficiency for inorganic iodides is assumed. This represents the lower limit reported in charcoal adsorption tests at 100% relative humidity (12). A 3% bypass flow and 10% organic iodide are assumed. The iodine reduction factor is defined as the ratio of the mass fission products vented to the environment without any cleanup system versus the mass released with the specified filter system.

$$\begin{aligned}
 \text{IRF} &= \frac{\text{total release}}{[1-(\text{efficiency} \times \text{fraction throughput} + \text{bypass})] + \text{non-removable}} \\
 &= \frac{1}{[1-(0.95 \times 0.97 + 0.03)] + 0.10} \\
 &= \frac{1}{[1-0.90] + 0.10} = \frac{1}{0.20} \\
 &= 5
 \end{aligned}$$

B. SUPERIOR CASE

A superior system is one built and maintained according to rigid specifications and standards and tested at frequent intervals. For such a system, a 98% inorganic iodide removal efficiency and reduced 1.5% bypass flow may be granted. Then

$$\begin{aligned}
 \text{IRF} &= \frac{1}{[1-(0.98 \times 0.985 + 0.015)] + 0.10} \\
 &= \frac{1}{(1-0.95) + 0.10} = \frac{1}{0.15} \\
 &= 6.7
 \end{aligned}$$

IV. INTERNAL RECIRCULATION FILTER SYSTEM

Reduction of airborne fission products by a continuous recycle filter system is considered here in terms of their time rate of addition to the system. The limiting cases are (1) an instantaneous release of the total quantity at time zero (puff release) and (2) a constant and continuous release over the entire time period under consideration (continuous release). The mathematics for the intermediate case of release over a limited portion of the operational period is also outlined (13-14). The discussion is general and applicable to all filter materials. Specific recommendations are given in Section F.

A. PUFF RELEASE

The rate of change of airborne fission product concentration is

$$\frac{dc}{dt} = - \frac{QEmc}{V} \quad (1)$$

where c = airborne concentration at time t
 t = total time of operation
 Q = flow rate through filter(s) per unit time
 E = removal efficiency
 V = total containment volume
 m = atmospheric mixing factor

A separate equation of this type must be written for each component of interest. Integration of the above equation gives the fraction of the initial concentration remaining airborne at time t

$$\frac{c}{c_0} = \exp -\{[(QEm/V)t]\} \quad (2)$$

where c_0 = airborne concentration at time zero

For a specific reactor system

$$\frac{Q}{V} = n \quad (3)$$

where n = number of air change cycles per unit time

Then

$$\begin{aligned} \frac{c}{c_0} &= \exp -\{[(nEm) \cdot t]\} \\ &= \exp \{- (\alpha_x t)\} \end{aligned} \quad (4)$$

where $\alpha_x = nEm$

= removal constant for component x in units of reciprocal time

The total mass of a single component available for leakage with an operative removal system, over any time period starting from $t = 0$, is obtained by integration of the above equation and multiplying by the leakage fraction. Then

$$\begin{aligned} M_L &= Lc_0V \int_0^t \exp[-(\alpha_x t)] dt \\ &= \frac{Lc_0V}{\alpha_x} [1 - \exp[-(\alpha_x t)]] \end{aligned} \quad (5)$$

where L = containment leakage fraction per unit time
 V = containment volume

The mass available for leakage in the absence of a removal system is

$$M'_L = Lc_0 V \int_0^t dt \quad (6)$$

$$= Lc_0 Vt$$

The iodine reduction factor (IRF) for a filter system is the reciprocal of the ratio of these two quantities

$$IRF = (M'_L/M_L)^{-1} = M'_L/M_L \quad (7)$$

$$= \frac{\alpha_x t}{[1 - \exp \{- (\alpha_x t)\}]}$$

For a number of components derivative from a single fission product isotope, each component is weighted by its mole fraction average over the entire containment volume. Specifically, for fission product iodine the components may be classed as inorganic (elemental and hydrogen iodide), organic (methyl iodide and higher homologues), and that associated with particulates. The overall iodine reduction factor (IRF) for a puff release is then

$$IRF = \frac{1}{\frac{F_i}{\alpha_i t} [1 - \exp \{- (\alpha_i t)\}] + \frac{F_o}{\alpha_o t} [1 - \exp \{- (\alpha_o t)\}] + \frac{F_p}{\alpha_p t} [1 - \exp \{- (\alpha_p t)\}]} \quad (8)$$

where F_i = mole fraction of inorganic iodine
 α_i = removal constant for inorganic iodine
 F_o = mole fraction of organic iodine
 α_o = removal constant for organic iodine
 F_p = mole fraction of particulate iodine
 α_p = removal constant for particulate iodine

B. CONTINUOUS RELEASE

For a continuous and constant release of fission products over the entire period, the rate of decrease in the airborne concentration is

$$\frac{dc}{dt} = \frac{R}{V} - \frac{\lambda E mc}{V} \quad (9)$$

where R = input rate (mass per unit time)

Solution of this equation over the limits $t = 0$ to $t = t$ gives the instantaneous airborne concentration at time t

$$c = \frac{R}{QEm} \{1 - \exp [-(QEmt/V)]\} \quad (10)$$

The ratio of mass of material airborne to total mass added, at time t , is

$$\begin{aligned} \frac{M}{M_T} &= \frac{cV}{Rt} = \frac{V}{QEmt} \{1 - \exp [-(QEmt/V)]\} \\ &= \frac{1}{\alpha_x t} \{1 - \exp [-(\alpha_x t)]\} \end{aligned} \quad (11)$$

The amount of material available for leakage with an operational internal recirculation filter system is then

$$\begin{aligned} M_L &= \int_0^t (LM) dt \\ &= \int_0^t \frac{LM_T}{\alpha_x t} \{1 - \exp [-(\alpha_x t)]\} dt \end{aligned} \quad (12)$$

and since $M_T = Rt$

$$M_L = \int_0^t \frac{LR}{\alpha_x} \{1 - \exp [-(\alpha_x t)]\} dt \quad (13)$$

Integrating

$$M_L = \frac{LR}{\alpha_x} t + \frac{LR}{\alpha_x^2} \exp [-(\alpha_x t)] - \frac{LR}{\alpha_x^2} \quad (14)$$

and the fractional leakage is

$$\frac{M_L}{M_T} = \frac{L}{\alpha_x^2 t} [\alpha_x t + \exp [-(\alpha_x t)] - 1] \quad (15)$$

The amount of leakage in the complete absence of a safeguard system would be

$$\begin{aligned} M'_L &= \int_0^t LRt dt \\ &= \frac{LRt^2}{2} \end{aligned} \quad (16)$$

and the fractional leakage is

$$\frac{M'_L}{M_T} = \frac{\frac{LRt^2}{2}}{Rt} = \frac{Lt}{2} \quad (17)$$

The iodine reduction factor (IRF) for the case of continuous fission product input and simultaneous removal is then

$$\begin{aligned} \text{IRF} = \frac{M'_L}{M_T} &= \frac{Lt/2}{(L/\alpha_x^2 t) [\alpha_x t + \exp[-(\alpha_x t)] - 1]} \\ &= \frac{\alpha_x^2 t^2}{2[\alpha_x t + \exp[-(\alpha_x t)] - 1]} \end{aligned} \quad (18)$$

C. RELEASE OVER LIMITED TIME PERIOD

For the case where the fission product release is constant and continuous over the time period $t = 0$ to $t = t$, and operation of the cleanup system continues beyond that time for a period T for cumulative time $t = t + T$, the applicable equations are:

1. the fraction of fission products airborne at any time

$$\frac{M_{t+T}}{M_T} = \frac{1}{\alpha_x t} \{1 - \exp[-(\alpha_x t)]\} \{\exp[-(\alpha_x T)]\} \quad (19)$$

2. the fraction of fission products leaked

$$\frac{M_L}{M_T} = \frac{L}{\alpha_x^2 t} \{\alpha_x t - \exp[-(\alpha_x T)] + \exp[-(\alpha_x(t+T))]\} \quad (20)$$

and

3. the iodine reduction factor

$$\text{IRF} = \frac{\alpha_x^2 t (t+2T)}{2 \{\alpha_x t - \exp[-(\alpha_x T)] + \exp[-(\alpha_x(t+T))]\}} \quad (21)$$

D. FILTER EFFICIENCY

The initial efficiency of the charcoal filter system for elemental iodine and for hydrogen iodide can be assumed as 90%, with a 5% reduction for each successive pass. Therefore

$$\alpha_p = m \sum_p \{ .90 - [.90 (p-1) .05] \} \quad (22)$$

where α_p = cumulative value for α_x

where p = number of passes

For most systems, the average efficiency of the filter system for inorganic halogens during the initial two hour period is then about 0.80, and the mixing factor can be taken as 0.9 for the total containment volume. Over a longer time period, the average filter efficiency is taken as 0.50, and the mixing factor as 0.75 of total volume.

E. NON-REMOVABLE COMPONENT

For the case of a puff release

$$M_L = LC_0 V \left[\epsilon \int_0^t dt + (1-\epsilon) \int_0^t \exp(-\alpha_x t) dt \right] \quad (23)$$

$$= LC_0 V \left(\epsilon t + \frac{[1-\epsilon]}{\alpha_x} [1 - \exp(-\alpha_x t)] \right)$$

and, with no removal system

$$M'_L = LC_0 V t \quad (24)$$

then

$$IRF' = \frac{LC_0 V t}{LC_0 V \left(\epsilon t + \frac{[1-\epsilon]}{\alpha_x} [1 - \exp(-\alpha_x t)] \right)} \quad (25)$$

where IRF' = modified IRF
 ϵ = fraction unremovable iodine

but, for all cases of interest $1 \gg \exp(-\alpha_x t)$

$$\text{and } IRF' = \frac{t}{\epsilon t + \frac{1-\epsilon}{\alpha_x}} \quad (26)$$

$$= \frac{\alpha_x t}{\alpha_x \epsilon t + (1-\epsilon)}$$

$$\text{or } \frac{1}{IRF'} = \epsilon + \frac{1-\epsilon}{IRF}$$

F. RECOMMENDED PROCEDURE

The iodine reduction factor for the two hour release is to be calculated on the basis of a 1/2 hour initial release time of the iodine from the core and on the calculated number of containment air changes per hour. A mixing factor of 0.9 for single containment shells and 0.8 for sectioned compartment shells is recommended.

Credit for effective removal of inorganic iodine is to be given only for representative charcoals which have been tested under post-accident conditions and have shown removal capability of 98+% for representative bed depths at 98% relative humidity. Fractional removal credit for the organic iodide fraction may be granted for certain impregnated charcoals under conditions of low face velocity and incident relative humidity of less than 70%.

In the absence of specific safeguards to prevent the charcoal temperature from exceeding 250°C (480°F), where appreciable fission product desorption begins, no removal credit is to be given for iodine dose reduction by internal recirculation filter systems for post-accident times in excess of two hours. However, the two hour holdup credit is to be applied in all instances. It is recommended that the maximum permitted dose reduction factor for systems with cooling adequate to dissipate fission product decay heat be based on the charcoal iodine retention capability under conditions of reduced flow. This is dependent on the number of filter units, reactor power, carbon weight and surface area and the design flow rate. All cases will be evaluated by RT on an individual basis.

G. CALCULATED VALUES

The following data are for Turkey Point Units 3 and 4, Florida Power and Light Company and are illustrative of typical PWR current units (15).

$$Q = 37,500 \text{ cu. ft./min per unit}$$

$$V = 1.55 \times 10^6 \text{ cu. ft.}$$

The number of installed filter units is three. A single failure criterion reduces this to two units operational under post-accident conditions. Then, the number of containment air changes per hour is

$$n = \frac{2 \times 3.75 \times 10^4 \times 60}{1.55 \times 10^6} = 3$$

For the initial two hour period an average filter efficiency of 0.80 and a mixing factor of 0.90 are assumed for inorganic iodine. The fraction of inorganic iodine is taken as 0.90, with the remainder considered unremovable.

1. Puff Release

$$\begin{aligned} \text{IRF} &= 1 / \left[\frac{0.90}{3 \times 0.80 \times 0.9 \times 2} \right] [1 - \exp(-3 \times 0.80 \times 0.9 \times 2)] \\ &= \frac{1/(0.95)}{4.3} (1 - \exp(-4.3)) \\ &= 4.3 / (0.95) (0.986) \\ &= 4.3 \end{aligned}$$

2. Continuous Release

$$\begin{aligned} \text{IRF} &= \frac{(4.3)^2}{2 [4.3 + \exp(-4.3) - 1]} (0.90) \\ &= \frac{18.4}{2 (4.3 + 0.014 - 1)} (0.90) \\ &= \frac{18.4}{6.6} (0.90) \\ &= 2.6 \end{aligned}$$

3. Release Time (t) of 1/2 Hour and Filter Operation Time (t + T) Over a Two Hour Period

$$\begin{aligned} \text{IRF} &= \frac{(2.17) \times 1/2 \times 7/2}{2 [2.17/2 - \exp(-2.17 \times 1.5) + \exp(-4.3)]} (0.90) \\ &= \frac{4.7 \times 7/4}{2 (1.08 - 0.05 + 0.01)} (0.90) \\ &= 3.6 \end{aligned}$$

V. SPRAY SYSTEM

The removal of iodine by sprays is evaluated by applying the following procedure, modeled on the work by Griffith (16-18). For the general case, transfer of iodine into the drop proceeds across a stagnant gas surface layer followed by adsorption into the liquid by either a diffusion mechanism or by chemical reaction.

A. PHYSICAL ADSORPTION (EQUILIBRIUM PARTITION)

Spray systems using only water have a maximum removal capability determined only by the equilibrium iodine partition factor. This partition factor,

defined as the ratio of the weight of iodine per unit volume of water to that in an equal volume of air, is strongly temperature dependent and decreases from about 100-200 at 20°C to 10 at 100°C at a pH of 7. The solubility increases rapidly with increasing pH value. The actual removal capacity of a water spray is therefore dependent on ratio of solution to air volume, temperature, and pH. Only the case for a puff release will be considered here. That for a continuous release is similar if the equilibrium time is rapid and the total release is complete during the time period under consideration.

At equilibrium

$$\frac{C_L}{C_G} = R \quad (1)$$

where C_L = iodine concentration in liquid phase

C_G = iodine concentration in gas phase

R = equilibrium partition factor

Assuming all the iodine is initially in the gas phase, the material balance at equilibrium is

$$V_G (C_{Gi} - C_{Gt}) = V_L C_L \quad (2)$$

but $C_L = R C_{Gt}$

then

$$V_G (C_{Gi} - C_{Gt}) = V_L R C_{Gt} \quad (3)$$

The iodine reduction factor is

$$\begin{aligned} \frac{C_{Gi}}{C_{Gt}} &= \frac{V_G + V_L R}{V_G} \\ &= 1 + \frac{V_L}{V_G} R \end{aligned} \quad (4)$$

B. GENERALIZED GAS ABSORPTION INCLUDING CHEMICAL REACTION.

The parameters required for the calculation of iodine by falling drops are:

1. Gas Film Mass Transfer Coefficient (Ranz and Marshall)

$$k_g = \frac{D M_{I_2}}{d R T} \left[2 + 0.6 (Re)^{1/2} (Sc)^{1/3} \right] \quad (5)$$

where D = diffusivity of I_2 in air-steam ($\text{cm}^2 \text{sec}^{-1}$)

M_{I_2} = molecular weight of iodine

d = drop diameter (cm)

R = gas constant = $82.3 \text{ cm}^3 \text{ atm deg}^{-1}$

T = temperature ($^{\circ}\text{K}$)

Re = Reynolds number = $\frac{\rho v d}{\mu}$

Sc = Schmidt number = $\frac{\mu}{\rho D}$

ρ = density of air-steam

v = terminal velocity of drop

μ = viscosity of air-steam mixture

2. Gas Phase Deposition Velocity

$$V_G = \frac{RT}{M_{I_2}} \cdot k_g \quad (6)$$

For iodine

$$V_G = 94.7 \cdot k_g \text{ cm sec}^{-1} \text{ at } 20^{\circ}\text{C}$$

$$= 121 \cdot k_g \text{ cm sec}^{-1} \text{ at } 100^{\circ}\text{C}$$

3. Liquid Film Mass Transfer Coefficient

$$K_L = \frac{2\pi^2}{3} \frac{D_L}{d} \quad (7)$$

where D_L = diffusivity of iodine in water

= $1.14 \times 10^{-5} \text{ cm}^2 \text{ sec}$ at 20°C

= $5.15 \times 10^{-5} \text{ cm}^2 \text{ sec}$ at 100°C

4. Water-Air Partition Factor for Iodine (pH dependence) (19,20)

$$R = \frac{[I_2] + (1 + 2K_2 [I_2]) (K_3/[H^+] + v_4)^{1/2} (1/[I_2] + K_2)^{-1/2}}{K_1 [I_2]} \quad (8)$$

where $[I_2]$ = iodine concentration in solution

	25°C	100°C
$K_1 = \frac{[I_2]_{aq}}{[I_2]_{gas}}$	83	9.1
$K_2 = \frac{[I_3^-]}{[I_2]_{aq} [I^-]}$	768	305
$K_3 = \frac{[H^+] [I^-] [HIO]}{[I_2]_{aq}}$	5.40×10^{-13}	1.57×10^{-10}
$K_4 = \frac{[H_2 I^+] [I^-]}{[I_2]_{aq}}$	1.2×10^{-11}	3.9×10^{-11}

5. Overall Mass Transfer Velocity Into Drop

$$\frac{1}{V_D} = \frac{1}{V_G} + \frac{1}{RK_L} \quad (9)$$

6. Total Surface Area of Drops in Containment

$$S = \frac{6fh}{dv} \quad (10)$$

where f = flow rate (cm³/sec)
 h = drop fall height (cm)
 d = drop diameter (cm)
 v = terminal velocity (cm/sec)

7. Removal Time Constant

$$\lambda_s = \frac{V_D S}{V_C} \quad (11)$$

where V_D = overall mass transfer velocity
 V_C = containment volume (cm³)

8. Average Removal Factor

$$\exp - (\lambda_{st})_{ave} = \frac{1 - \exp [-(\lambda_{st})]}{\lambda_{st}} \quad (12)$$

C. DROP SIZE VARIATION

The drop size spectrum can be assumed to approximate a log normal distribution. A complete description includes a mean geometric drop diameter and the standard deviation from this mean geometric drop diameter. The reduced surface area associated with larger drops must be compensated by an empirical reduction factor of the calculated spray removal time constant based on the geometric standard deviation.

<u>σ_{geom}</u>	<u>correction factor</u>
1.0	1.00
1.5	0.50
1.8	0.25
2.0	0.15

D. RECOMMENDED PARAMETERS

The input parameters recommended for calculating the iodine removal capability of alkaline aqueous sprays for elemental iodine are tabulated below:

$$V_G = 6.0 \text{ cm/sec at } 130^\circ\text{C and } 55 \text{ psig}$$

$$K_L = 3.4 \times 10^{-3} \text{ cm/sec based on } 1000 \text{ drop diameter}$$

The water-air iodine partition factor R is calculated by the following procedure.

The total core iodine inventory based on 500 day operation, for a 2500 MWth reactor is approximately 12,000 g. The containment volume is taken as $1.5 \times 10^6 \text{ ft}^3$, and the liquid volume as 1/100 of this. For this, the iodine concentration in solution is calculated as follows:

$$\begin{aligned} 25\% \text{ of } I_2 \text{ released to containment} &= 3 \times 10^3 \text{ g} \\ \text{Air Volume} &= 1.5 \times 10^6 \text{ ft}^3 = 4 \times 10^{10} \text{ cm}^3 = 4 \times 10^7 \text{ l} \\ \text{Liquid Volume} &= 4 \times 10^5 \text{ l} \end{aligned}$$

If the entire amount of iodine is assumed in solution (this introduces only a negligible error in the following calculations), then the aqueous iodine concentration is approximately $1 \times 10^{-2} \text{ g/l}$ or $5 \times 10^{-5} \text{ moles/l}$. This value is then used to derive the water/air partition coefficient. For pH = 9.5, temperature = 25°C , and $[I_2]_{aq} = 10^{-4} \text{ moles/l}$, the value of $R = 9.5 \times 10^2$. Above pH = 9, the partition coefficient is very sensitive to both changes in $[H^+]$ concentration and temperature. The value chosen here is to be considered as representative of average post-accident conditions, with a factor of conservatism introduced by using the minimum temperature.

For solutions which remove elemental iodine from the gas-liquid interface by a rapid and irreversible chemical reaction the liquid film resistance is effectively zero. In this case, the overall mass transfer velocity into the drop equals the gas phase deposition velocity. Experimental results indicate that this model is applicable for several chemical compounds, such as sodium thiosulfate, hydrazine, etc. For solutions where an equilibrium between the elemental iodine and a soluble species exists, the effect of the liquid film resistance may become appreciable. Such a model is applicable for alkaline solutions which are pH controlled by the addition of sodium hydroxide.

In the absence of definitive information on the stability of certain additives under post-accident conditions, it is recommended that all spray systems be analyzed on the basis of an alkaline solution of pH = 9.0 and with the inclusion of a liquid film coefficient as indicated below. The overall iodine deposition (mass transfer) velocity is then

$$\begin{aligned}\frac{1}{V_D} &= \frac{1}{V_G} + \frac{1}{RK_L} \\ &= \frac{1}{6.0} + \frac{1}{(9.5 \times 10^4)(3.4 \times 10^{-3})} \\ &= 0.16 + 0.31 = 0.47 \\ V_D &= 2.1 \text{ cm/sec}\end{aligned}$$

Other input parameters are functions of a specific system and included in the application. Conversion factors required are:

From	To	Factor
gpm	cm ³ /sec	6.31 x 10 ¹
ft	cm	3.05 x 10 ¹
μ	cm	1 x 10 ⁻⁴
cu. ft.	cm ³	2.83 x 10 ⁴
ft/min	cm/sec	5.08 x 10 ⁻¹

E. CALCULATED VALUE

The following example is for Crystal River 3, a typical current DWP spray system. Calculations are based on one of two subsystems operational.

$$\begin{aligned}
 \text{flow } f &= 1500 \text{ gpm} = 9.5 \times 10^4 \text{ cm}^3/\text{sec} \\
 \text{height } h &= 90 \text{ ft} = 2.7 \times 10^3 \text{ cm} \\
 \text{diameter } d &= 1000 \mu = 1.0 \times 10^{-1} \text{ cm} \\
 \text{term. vel. } v &= 400 \text{ cm/sec} = 4.0 \times 10^2 \text{ cm/sec} \\
 \text{cont. volume } V_c &= 2 \times 10^6 \text{ ft}^3 = 5.5 \times 10^{10} \text{ cm}^3
 \end{aligned}$$

Using these values and the above $V_D = 2.1 \text{ cm/sec}$,

$$\begin{aligned}
 \lambda_s &= \frac{6 V_D f h}{V_c d v} \\
 &= \frac{(6)(2.1)(9.5 \times 10^4)(2.7 \times 10^3)}{(5.5 \times 10^{10})(1 \times 10^{-1})(4 \times 10^2)} \\
 &= 1.47 \times 10^{-3} \text{ sec}^{-1}
 \end{aligned}$$

multiplying by $3.6 \times 10^3 \text{ sec/hr}$

$$\lambda_s = 5.3 \text{ hr}^{-1}$$

For an assumed standard deviation of 1.5 from the mean geometric drop diameter the correction factor is $1/2$ and

$$(\lambda_s)_{\text{corr}} = 2.6 \text{ hr}^{-1}$$

The variation of the IRF for the initial two hour period, with different fractions of non-removable iodine, is given by

$$\text{IRF}' = \frac{2}{(2\epsilon - (1 - \epsilon/(\lambda_s)_{\text{corr}}))}$$

ϵ	$1 - \epsilon/\lambda_s$	$2\epsilon - 1 - \epsilon/\lambda_s$	IRF'
0.00	0.38	0.38	5.3
.05	0.36	0.46	4.4
.10	0.34	0.54	3.7
.15	0.32	0.62	3.2
.20	0.30	0.70	2.9

No specific credit has been given for the effect of additives in sprays because neither their compatibility with reactor materials nor their iodine removal effectiveness has yet been conclusively demonstrated. However, the greatly increased partition coefficient due to the hydrolysis of iodine in alkaline solutions has been applied.

The mass transfer coefficient has been calculated with the liquid phase resistance included. This is probably applicable to solutions using sodium hydroxide only, but conservative for solutions which include sodium thiosulphate.

VI. SPRAY & FILTER SYSTEM COMBINATIONS

A. SPRAY AND EXTERNAL FILTER SYSTEM

For a spray plus once - through filter system working out of separate enclosures, where the charcoal adsorbers are not exposed to high temperature and high humidity, these can be considered as successive cleanup systems and the combined iodine reduction factor is the product of that of the separate systems.

B. SPRAY AND INTERNAL RECIRCULATION FILTER SYSTEM

For a spray plus internal recirculating filter system, competing in the same containment, the combined reduction factor must be based on the relative removal rates of the two systems. The spray system can be assumed to be a more rapid removal device and full credit given for its operation. The filter system, working simultaneously, removes a smaller fraction of the entire airborne inventory per pass.

Cleanup is taken as a competitive rate phenomenon of two zero order reactions. Exponential removal factors equal to those for the separate systems are used, with the total factor equal to the product of the exponentials.

The spray removal factor is equal to

$$\lambda_s = V_D S_C \quad (1)$$

where V_D = overall mass transfer velocity

S_C = drop surface area per unit containment volume

The filter removal factor is equal to

$$\lambda_f = nE \quad (2)$$

where n = number of air changes per unit time

E = filter efficiency

However, in order to compensate for differences in removal rates by the spray and filter systems, the rate constant for the filter system is arbitrarily reduced by the ratio of the two constants.

The fraction of fission products remaining at time t is then

$$\frac{c}{c_0} = \exp \{ [-(\lambda_S + \frac{\lambda_F}{\lambda_S} \lambda_F)] t \}$$

$$= \exp [-(\lambda_S + \lambda_F^2 / \lambda_S) t] \quad (3)$$

The overall iodine reduction factor for a puff release is

$$IRF = \frac{\lambda_{SF} t}{[1 - \exp - (\lambda_{SF} t)]}$$

$$\text{where } \lambda_{SF} = \lambda_S + \lambda_F^2 / \lambda_S$$

The dose reduction factor for a combined spray-containment recirculation filter system should be calculated on the basis of the separate constants for each system, with a suitable reduction for the effectiveness of the filter system. A further correction is required to include the effect of a non-removable fraction.

C. IODINE REDUCTION FACTORS GRANTED

The Table below lists the dose reduction factors for spray - filter systems required by approved applicants through February 1968.

DOSE REDUCTION FACTORS

PROJECT	POWER (Mwt)	SITE BOUNDARY (feet)	LOW POPULATION ZONE (Miles)	E.S.G.	REDUCTION FACTOR	
					SITE	LPZ
Indian Pt. 2	2758	1584	0.6	spray-filter	3.0	11.0
Met. Ed.	2568	2000	2.0	spray	5.3	4.0
Diablo	3391	2640	6.0	spray	2.9	0
Rochester	1300	1500	12.0	spray-filter	1.3, 6.0*	0
Point Beach 1	1396	3920	7.0	spray	0**	0
Robinson	2095	1400	5.0	spray	5.0***	0
Conn. Yankee	1473	1740	9.5	spray-filter	0	0

* Factor of 6 required if building wake was not included in meteorological model.

** Reduction factor not required, but stated in P.S.A.R. (Public Safety Analysis) that if dose reduction factors were required of 10 for the site boundary and 20 for the low population zone would be used.

*** Estimated. Not specifically stated in Public Safety Analysis Report.

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