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U.S. NUCLEAR REGULATORY COMMISSION

June 2, 1981

U. S. Nuclear Regulatory Commission
Division of Licensing
Washington, DC 20555

Attn: Chief, Operating Reactors Branch 4

Ref: Docket 50-54

References: a. Letter from N.R.C. to Union Carbide Corp.
Dated Dept. 13, 1973
Subj: Change No. 11, License R-81

b. Amendment No. 14 to License R-81
Dated May 17, 1979: Technical Specifications

c. Letter Union Carbide Corp. to N.R.C.
Dated May 23, 1980
Subj: Renewal, Operating License R-81

Gentlemen:

It is requested that item 3.5.2 c (5) in the Technical Specifications for the Union Carbide Nuclear Reactor, as contained in Refs. b. and c. above, be changed to read as follows:

(5) The iodine inventory of a single capsule shall be limited to 500 Curies I-131 dose-equivalent.

A Safety Analysis supporting this request is enclosed.

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The requested change will enable more efficient use to be made of a scarce national energy resource material (uranium-235) in the production of radioisotopes for nuclear medicine applications. The present wording of the Technical Specifications item requires double encapsulation for any quantity over 70 Curies. This results in substantially less effective utilization of the U-235 target material. It also greatly increases the temperature attained by the inner capsule, which is undesirable from those safety considerations principally relating to mechanical strength and to fission product diffusion out of the UO_2 into the gas phase. The volume of irradiated waste is also increased, which is undesirable for disposal reasons.

For the record, in the past 12 years a total of approximately 120 singly and 6700 doubly-encapsulated capsules have been irradiated in the reactor with no instance of capsule failure. We believe this is evidence of good experiment design and quality control.

We propose this requested change as a Class III Amendment and enclose our check for $2000.

Very truly yours,

K. D. George
Kenneth D. George
Senior Development Scientist

KDG:js

3 Enclosures: 1. Safety Analysis
2. Appendix A
3. Appendix B

(13 copies enc.)

STATE OF NEW YORK)
SS
COUNTY OF ORANGE)

On this 11 day of June 1981, before me personally came Kenneth D. George to me known and known to me to be the individual described in and who executed the foregoing instrument and acknowledged that the executed the same.

ANNE R. WOLYNIES
Notary Public, State of New York
No. 4332700
Qualified in Orange County
Certificate Filed in Orange County
Term Expires March 30, 19 83

Anne R. Wolynies
Notary Public

SAFETY ANALYSIS

Single Encapsulation of Capsules Containing Enriched Uranium

NOTE: References are listed at the end of this Analysis. Pertinent extracts from these are included for convenience in an Appendix B.

1. Abstract

The need to limit single ecapsulation to a maximum capsule content of 70 Curies I-131 dose-equivalent (Ref. 1) is re-examined. Published data on release-limiting physical processes are shown to result in an iodine decontamination factor that is about 500 times greater than that used in the N.R.C. Staff analysis. If a margin of 10 for uncertainties is then applied, the capsule inventory can be safely increased by a factor of 50 without exceeding the acceptable doses calculated by Staff for a 70-Curie (I-131 dose-equivalent) capsule. Other present restrictions in the Technical Specifications, such as maximum fission power, limit the content of a capsule to 500 Curies I-131 dose-equivalent so the practical increase in inventory for a singly-encapsulated capsule would be only a factor of a little over 7.

2. N.R.C. Staff's Safety Evaluation:

The source term bases used by the Staff (Ref. 1) for evaluation of possible radiation doses to personnel both within the containment building (hereafter called "onsite") and offsite at the plant boundary were as follows:

100% of the noble gas and iodine inventory is released from a capsule.

100% of the noble gases appear in the containment building air.

10% of the iodines appear in the containment building air.

Mixing takes place with only 50% of the building air.

Doses to personnel onsite and offsite for a 500-Curie capsule were then calculated for two different conditions of the engineered safeguards, as follows:

a. Engineered Safeguards Fail.

   Thyroid doses both within containment and offsite were shown to be unacceptably high (i.e. 204 and 84 rem respectively), a not unexpected result considering the above source term assumptions.

b. Engineered Safeguards Function Correctly.

   Doses within containment (onsite) were not affected.

   Doses offsite were reduced to acceptable levels.

Based on the above evaluation, Staff requested that capsules containing more than 70 Curies I-131 equivalent be doubly encapsulated to reduce the probability of occurrence of capsule failure. The failure of capsules containing less than 70 Curies I-131 equivalent of fission products will result in onsite personnel exposures that do not exceed those permitted in 10 CRF Part 20. This limitation was made a Technical Specification condition.

It was further requested that the engineered safeguards be evaluated for reliability and modified to provide assured performance. This licensee made the evaluations and completed the resulting modifications on a schedule agreed to by Staff. These changes give assurance that the engineered safeguards will operate properly and, in case of capsule failure and release of all contents, result in acceptable offsite doses for capsule contents up to 500 Curies I-131 dose equivalent.

3. Re-evaluation of Iodine Release:

It will be shown below that the source term bases of the Staff's 1973 evaluation, as listed above, are excessively conservative for iodines.

Release of fission products to containment atmosphere can be broken down as follows:

3.1 Capsule Damage Mechanisms

a. Mechanical damage, e.g. crushing

b. Material defects, e.g. bad welds, and cracks

c. Improper seals, i.e. loose entry port caps

d. Capsule melting, e.g. flow blockage, excessive heat flux

3.2 Type of Fission Product Release

a. Release due to the first three mechanisms above (3.1 a,b,c,) is that of the gas-phase contents of a capsule. This will therefore be similar to the "gap activity" release resulting from cladding failure in power reactor fuel. The inventory fractions so released are small, typically of the order of .001-1%. For example the method of (proposed) ANSI/ANS Standard 5.4 (see Appendix A) gives release fractions of 0.1% for Xe-133 and 0.16% for I-131 from the UO_2 in a capsule. These calculated results are roughly confirmed by the measurements of 2.5% and 0.1% respectively made by this licensee in 1973. In any event the source term is below that assumed by Staff by a factor of 40 or more for noble gases, and a factor of 500 or more for iodines.

b. Release with the fourth mechanism (above, 3.1 d) should be similar to that from UO_2 at high temperature, in the presence of molten metal (stainless steel or zirconium) and of steam. The inventory fraction released from the fuel itself can approach 100%, as in the Staff's evaluation. In the case of iodines only a small portion of this is, however, available for transport through the pool water to the containment air, as indicated below.

3.3 Release-Limiting Mechanisms:

a. The first type of release above (gas-phase fraction) will be subject to absorption in the 24-ft-thick layer of reactor pool water, for capsules in the core. For capsules being transported outside the core, the water depth may be less (but at least 10 ft. because of shielding requirements).

b. The second type of release (capsule melting) will in the case of iodines be reduced by interaction with molten capsule material, by chemical combination with other fission-product elements and by scavenging with condensing steam. Any gaseous iodine escaping those natural consequence-limiting mechanisms will be subject to absorption and dilution in the 24-foot thick layer of reactor pool water before release to the building atmosphere.

3.4 Magnitude of Source-Term Reduction Factors

a. In the case of the "gap-activity" or gas phase type of release (3.2 a above), the fraction of the inventory available for release is assumed to be the larger of the values cited above, namely 2.5% for gases, 0.2% for iodines. Additional evidence is two experiments (Ref. 2, Table 1.5) in which Zircaloy-clad UO_2 fuel was heated enough to rupture the cladding, but not to melt either the fuel or the clad, that gave iodine release fractions of 0.189% and 0.115% of the contained inventory. Iodines will be further reduced by pool water absorption. Staff assumed a pool decontamination factor of only 10. Using the latter number and a release fraction of 0.2% the iodine reduction factor for this case is about 5000. Using the more realistic decontamination (or trapping) factors discussed below, the iodine reduction factor would be much greater than 5000.

b. For the capsule-melting type of release (3.2 b above), in which it is conservatively assumed that 100% of the iodines (and gases) are released from the UO_2, the following considerations apply:

(1) Scavenging By Steam and Molten Metal

Research programs conducted at Oak Ridge National Lab. (and elsewhere) around 1965 on fission product release from UO_2 fuel subjected to high temperature damage has produced data that are applicable to the capsule melting situation under discussion here. The fact that the UO_2 configuration differs is not relevant because we are conservatively assuming 100% release of gas and iodines from the fuel matrix. In these research programs UO_2 fuel clad with stainless steel or with Zircaloy was heated, electrically or with a reactor transient, sufficiently to melt the cladding and in some cases the fuel as well. The environment around the fuel element was either steam or water. It was found that both the molten cladding metal and the condensing steam are highly effective in absorbing and retaining iodine. Experiments in which SS-clad UO_2 was melted in moist air (#3,4, in Table 1 below) resulted in only about 1% of the I-131 inventory being transported from the autoclave by the sweep gas. Two experiments in high-pressure (1000 psi) steam, in which the clad, but not the fuel, melted (#5, 6Z below) showed fairly small I-131 release from the fuel (1-10%) and a small release from the autoclave (#5). A series of four experiments (#7, 8Z, 9, 10Z below), was conducted in which UO_2 was melted under water with low-pressure steam conditions that should approximate those in an open pool reactor. Release of I-131 from the fuel itself was large (up to about 75%) but the majority of this deposited on adjacent surfaces of the autoclave. The fraction of I-131 released from the autoclave through venting of the steam was less than 20% in all cases. The important point, however, is that in all cases practically all of this vented I-131 condensed and was retained in water traps. Only insignificant fractions were left for absorption in membrane filters or charcoal traps. The results of experiments 3 through 10 are summarized in the following table:

TABLE 1

DISTRIBUTION OF I-131, PERCENT OF INVENTORY

Experiment #	Fuel + Clad	Autoclave	Released From Autoclave	Condensed In Water Traps	Trapped In Filters	Reference[d]
3,4	60	38.1	1.1	~ 0.9[b]	.17	3, Table 2
5	99	0.55	~0.45[b]	0.15	.0038	3, Table 5
6Z[a]	90	10	0[c]			3, Table 6
7	18	77	5	5	.002	4, Table 1.2
8Z	25	68	~7.4	7.4	.007	4, Table 1.3
9	15	66	19	18.5	.004	5, Table 1.3
10Z	39	49	11.8	11.8	.0056	5, Table 1.3

NOTES:
- a. Z denotes Zircaloy cladding
- b. Estimated by difference
- c. No steam release due to blockage
- d. References are listed in Sect. 4

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Some conclusions from the results of the last four experiments were (Ref. 5, pp. 19-20):

(a) There was no significant effect of cladding material (stainless steel vs Zircaloy).

(b) Fission product release was similar in all four experiments, the twice larger I-131 transport in #9 and 10Z being ascribed to faster steam release.

(c) Rapid formation of non-volatile water-soluble iodine compounds was indicated by the results of #9, 10Z.

(d) Although 60-80% of the I-131 was released from the melted fuel, only .0006 - .005% was found on charcoal traps or collected as gas.

(e) Condensing steam is effective in trapping particulates.

As further evidence of the efficacy of condensing steam for removal of iodine, laboratory experiments are cited (Ref. 6, Table 4) in which fission products were released from heated uranium. It was found that 97% of the released I-131 was retained by the condensing steam, leaving only 3% for transport elsewhere.

(2) Trapping In Pool Water

In connection with suppression systems for power plants, work has been done in which iodine was injected into water-filled pools and the relative concentrations in water and supernatant air measured. The resulting concentration ratio (or partition coefficient) is dependent on pool water pH and initial concentration. Experiments (Ref. 7) with pool water at 100°C (an extreme case), pH = 6, and initial iodine concentration 6.5×10^{-5} g/l (5×10^{-7}M), gave a partition coefficient of 3×10^{2}. Other experiments (Ref. 8, Fig. 11) in which iodine-gas mixtures were bubbled into a water pool at 25°C with pH = 6 to 7 and iodine concentration 10^{-6} g/l gave partition coefficients of $\geq 10^4$. The initial iodine concentrations in our pool are $< 10^{-8}$ g/l even if all fission-produced iodine is assumed dissolved therein, so our partition coefficient is expected to be more than 10^4.

(3) Water-to-Air Partition:

While this topic is treated above in (2), the emphasis there is on the further trapping of the small amount of iodine that is released from the vicinity of the (assumed) melted capsule. One avenue of iodine release to containment building air that could be postulated, but was not specifically analyzed by Staff, is to assume that 100% of the iodine inventory becomes dissolved in the pool (50% of pool volume to be conservative), and to estimate the resulting air concentration above the pool surface. This requires values of partition coefficients from water-to-air. Reference 9, Tables 9 and 10, gives partition coefficients, calculated from measured concentrations, for water at 50°C and pH in 5-7 range, of $3 \times 10^3 - 3 \times 10^5$ for pool iodine concentrations of $< 4 \times 10^{-9}$ g/l. Reference 10 (Fig. 9) has plots of measured results at 25-80°C, pH 6-7. At an iodine liquid concentration of 10^{-4} g/l, equivalent to 10^{-5} mg/l in gas phase, the partition coefficient is shown as 2×10^3. This work was not extended to lower aqueous concentrations where partition coefficients would be expected to be larger still. In Reference 11 (Fig. 1), partition coefficients calculated from measured parameters are given. In water 25°C and a concentration of 10^{-7} g/l (the smallest used), the partition coefficients for pH = 5 and 7 are 10^4 and $> 10^5$ respectively.

For our pool water conditions (pH 5-7, total iodine concentration $\leq 4 \times 10^{-9}$ g/l) it is reasonable to assume a partition coefficient of at least 10^4. If all radioactive iodine (500 Ci) is dissolved in half the pool water volume of 120,000 gallons the concentration in the water is 1.1×10^{-6} Ci/ml. Applying a partition coefficient of 10^4 gives an air concentration (near the pool surface) of 1.1×10^{-10} Ci/ml. The building air concentration calculated by Staff was 50 Curies in half the building volume of 7700 m^3, or 1.3×10^{-8} Ci/ml, which is larger by a factor of more than 100.

3.5 Summary of Release-Limiting Factors for Iodine (The most conservative values are chosen below)

a. Capsule break (gas-Phase contents): 0.2% of inventory (Sect. 3.4 a).

b. Capsule melting:

(1) Release from UO_2 fuel: up to 84% of inventory (Table 1)

(2) Release to pool at core region: up to 20% of inventory (Table 1). (Includes effect of plateout on metal capsule and housing.)

(3) Release after steam condensation or water trapping in core vicinity: 0.17% (Table 1)

c. Decontamination through 24-ft-thick water layer: 10^4 (Sect. 3.4 b (2)

d. Air concentration from complete dissolution in pool 1.1×10^{-10} Ci/ml (i.e. 100% iodines in half pool volume). (Sect. 3.4 b (3).

3.6 Conclusions:

Even without invoking the large iodine decontamination due to passage through the 24-ft-deep pool, the amount of iodine available for mixing with building air (0.2%) is a factor of 50 below the quantity assumed by Staff (10%). A further reduction by a factor of 10^4 is supportable by the evidence described previously. This leaves 3.5 d above as the avenue more likely to give the greater air concentration in the case of complete fuel melting. As stated previously in 3.4 b (3), 100% dissolution gives a building air concentration that is 1/100 of that used by N.R.C. Staff. From the result cited in Sect. 3.5 b (2) above, a further reduction by a factor of 5 (20% release to pool water) can be justified.

Overall it is reasonable to expect that the containment building air-concentration will, under accident conditions, be lower than that estimated by Staff by a factor of 500. Allowing an order of magnitude margin for uncertainties leaves a safe reduction factor of 50.

While the two reactor systems differ considerably in size and design, there is some corroboration of the above from the results of the TMI-2 power reactor accident. It has been reported (Ref. 12) that 60% of the core I-131 inventory was released to the reactor cooling system, 17% was in the containment building water, and only .007% in the containment building air. These last two figures would indicate a partition coefficient of 2400. Similarly, the corresponding figures for the auxiliary building are 6% and 2×10^{-4}%, for a partition coefficient of 3×10^4.

Even closer confirmation of the correctness of these conclusions comes from tests performed in a system similar to the Union Carbide Nuclear Reactor (UCNR). These are experiments (Ref. 13) done in an open pool reactor (CABRI) in France in which several fuel plates were deliberately melted in order to measure the release of iodine into the water and its transmission through the pool water layer into the room air. The release of iodine into the water was 2-10% of that present in the damaged fuel. It is stated that the fuel, the water, and the materials constituting the interior of the core retain 99.995% of the iodine present in the damaged portion of the core. For a pool depth above the core of 2.5 - 3.3 m, the transmission factor through the water (ratio of activity in air to that in water) was found to be between 5×10^{-6} and 5×10^{-5}. This means that only .0005 - .005% of the iodine released into the water reached the room air, which implies a partition between water and air activity of $2 \times 10^5 - 2 \times 10^4$.

4. References

1: Letter N.R.C. to U.C.C., dated Sept. 13, 1973; Docket 50-54; Subj: Change No. 11, License R-81.

2: Nuclear Safety Program. Annual Report for Period Ending 30 December 1970. ORNL-4647 (May 1971).

3: International Symposium on Fission Product Release and Transport Under Accident Conditions, Oak Ridge TN., April 1965. CONF-650407, pp. 299-310.

4: Nuclear Safety Program Semiannual Progess Report for Period Ending December 31, 1965. ORNL-3915 (March 1966).

5: Nuclear Safety Program Annual Progress Report for Period Ending December 1966. ORNL-4071 (March 1967).

6: Hilliard, R. K., etal, Health Physics 7, 1-10 (1961)

7: Devell L., etal, Nuclear Technology 10, 466-471 (1971).

8: Diffey, R., etal, ibid, CONF-650407, (AERE-R4882), pp. 776-804.

9: Parsly, L. F., Calculation of Iodine-Water Partition Coefficients, ORNL-TM-2412, Par. IV, (Jan. 1970).

10: Nishizawa, etal, Nuclear Technology 10, 491 (1971).

11: Eggleton, A.E.J., etal, Atomic Energy Research Establishment, Harwell, AERE-R4887, (Feb. 1967).

12: Hull, Andrew P., Nuclear News, April 1981, pp. 61-67.

13: Dadillon, J., Bull Inform. Sci. Tech. (Paris), No. 112, 13-18, (Feb. 1967).

APPENDIX A

Volatile Fission Product Release From Oxide Fuel

Proposed American National Standard ANSI/ANS-5.4 (Nov. 1979), as revised to Nov. 1980, recommends methods for calculating oxide-fuel plenum-gas activity. The methods consider high-temperature and low-temperature releases and distinguish between short half-life (less than one year) and long half-life nuclides. The standard requires that releases be calculated with both high and low temperature methods, the larger result being selected.

This standard is applied to calculate releases of iodine and gaseous radionuclides in the analogous case of release into the interior of an intact capsule of the type under consideration here. The principal difference is that the UO_2 fuel in the case of a capsule is in the form of a thin layer.

All the nuclides of interest are short-lived. Comparison of the high and low temperature methods shows releases calculated by the latter to be 10^4-10^5 times greater than by the former. Thus only the low-temperature results are given below.

From Sec. 3.2.2 of the standard, the release fraction F is:

$$F = (10^{-7} \lambda^{.5} + 1.6 \times 10^{-12} P) / \lambda,$$

where: P = specific power = MW/t

λ = decay constant (s^{-1})

Results of the calculation are given in the following table.

TABLE 2

Nuclide	Half-Life	λ (sec.$^{-1}$)	F
Xe-133m	2.19 d	3.66×10^{-6}	4.6×10^{-4}
Xe-133	5.245 d	1.53×10^{-6}	*1.24×10^{-3}
Kr-85m	4.48 h	4.30×10^{-5}	5.0×10^{-5}
Kr-87	76.3 m	1.51×10^{-4}	1.8×10^{-5}
Kr-88	2.84 h	6.78×10^{-5}	3.4×10^{-5}
Xe-135m	15.29 m	7.56×10^{-4}	5.6×10^{-6}
Xe-135	9.09 h	2.12×10^{-5}	9.2×10^{-5}
Xe-137	3.85 m	3.00×10^{-3}	2.3×10^{-6}
Xe-138	14.17 m	8.15×10^{-4}	5.3×10^{-6}
I-131	8.04 d	9.98×10^{-7}	1.6×10^{-3}
I-132	2.30 d	8.37×10^{-5}	2.9×10^{-5}
I-133	20.8 h	9.26×10^{-6}	1.9×10^{-4}
I-134	52.6 m	2.20×10^{-4}	1.4×10^{-5}
I-135	6.61 h	2.91×10^{-5}	*1.6×10^{-4}

*Corrected for precusors per Section 3.3 of Standard.

APPENDIX B

For convenience, pertinent extracts from References cited in the foregoing Safety Analysis are appended.

(These extracts are keyed to the corresponding number in the list of References, Section 4.)

Table I.5. Fission Products Released in TREAT Fuel Rod Failure Experiments FRF-1 and FRF-2

Location	Material Found in Each Location (% of total in center irradiated rod)[a]									
	^{131}I		^{137}Cs		^{129}Te		Fission Products with Low Volatility[b]		U	
	FRF-1	FRF-2	FRF-1	FRF-2	FRF-1	FRF-2	FRF-1	FRF-2	FRF-1	FRF-2
Primary vessel[c]	0.054	0.066	0.046	0.193						
Filter pack										
Deposition by diffusion	0.076	0.037	0.0004	0.005	0.0001	$\sim 1.5 \times 10^{-6}$	0	0	0	0
Deposited with particles	0.046	0.010	0.010	0.090	0.015	$<1.5 \times 10^{-6}$	0.0009	1.4×10^{-6}	0.00024	16×10^{-6}
Condensate										
Unit 1	46×10^{-6}	$<60 \times 10^{-6}$								
Unit 2	43×10^{-6}	$<60 \times 10^{-6}$								
Heated charcoal										
Unit 1	0.0087	0.0002								
Unit 2	0.0038	0.0026								
Total release[d]	0.189	0.115	0.056	0.288						

[a] FRF-1 center rod was irradiated 13.1 full-power days to 645 MWd/MT peak burnup at 15.2 kW/ft peak linear power, peak-to-average flux ratio, 1.15; total fission, 9.8×10^{20}. FRF-2 center rod was irradiated 62.7 full-power days to 2800 MWd/MT peak burnup at 14 kW/ft peak linear power; peak-to-average flux ratio, 1.15; total fission, 4.3×10^{21}

[b] Median of ^{89}Sr, ^{95}Zr, ^{10} Ru, and ^{141}Ce.

[c] Primary vessel leached with 0.5 *N* NH_4OH; only soluble materials reported.

[d] Total release of ^{85}Kr was 0.094% in experiment FRF-1 and 0.48% in experiment FRF-2; total release of ^{133}Xe was 0.14% in experiment FRF-1 but was not determined in experiment FRF-2 because of long decay time.

TABLE 1. SUMMARY OF TREAT FISSION PRODUCT RELEASE EXPERIMENTS

EXPERIMENT	RELEASE OF VOLATILE F. P. FROM UO_2 AND CLAD	FUEL MELTED	FUEL AND CLADDING FRAGMENTED	TOTAL FISSION HEAT AND PREHEAT TO SPECIMEN, Cal/g UO_2	REACTOR PERIOD, msec.	INITIAL PRESSURE, psia	ATMOSPHERE	CLADDING	ELECTRICAL PREHEAT OF SPECIMEN, °C	UO_2 BATCH NO.	BURNUP, MWD/T
2	50%	65%		315	87	40	ARGON	S.S.	800	1	0
3	30%	75%		346	74	43	3% Steam- 97% Air	S.S.	800	1	0.2
4	30%	75%		335	77	53	30% Steam- 70% Air	S.S.	800	1	0.2
1	20%	0		309	108	39	ARGON	S.S.	800	1	0
6	10%	0	X	327	78	1000	STEAM	Zr-2	350	2	13
5	0.5%	0		330	77	1000	STEAM	S.S.	310	2	[illegible]

TABLE 2. DISTRIBUTION OF MATERIAL IN TREAT EXPERIMENTS 3 AND 4:
LOW PRESSURE STEAM-AIR, TRACE PREIRRADIATION, 75% MELTED

LOCATION	PERCENTAGE IN EACH LOCATION					
	^{131}I	^{129}Te	^{137}Cs	^{106}Ru	^{89}Sr ^{140}Ba	^{95}Zr ^{144}Ce UO_2
UO_2 FUEL + S.S. CLADDING	60	79	66	95	98	98.3
(S.S. CLADDING)	(3)	(9)	(0.8)	(0.2)	(0.25)	(0.24)
ALUMINA HEATER (1000°C)	25	13	28	2.3	0.4	0.43
AUTOCLAVE LINER	6.8	5.6	5.1	1.5	1.1	0.8
FUEL AUTOCLAVE WALLS (150°C)	6.3	0.6	0.7	0.7	0	0.46
GAS TRANSPORT ZONE	1.1	0.15	0.5	0.001	0.5	0.001
FILTERS (MEMBRANE)	0.02	0.01	0	0.01	0	0.0001
GAS-COLLECTION AUTOCLAVE	0.15	0.001	0.2	0.001	0.05	0.0000

TABLE 5. DISTRIBUTION OF MATERIAL IN TREAT EXPERIMENT 5: 1000 psia STEAM (RELEASED), TRACE PREIRRADIATION, NOT MELTED

LOCATION	PERCENTAGE IN EACH LOCATION					
	^{131}I	^{129}Te	^{137}Cs	^{89}Sr	^{95}Zr	UO_2
				^{140}Ba	^{144}Ce	Ru
UO_2 FUEL + S.S. CLADDING	99	99.6	99.9	99.99	99.998	
(S.S. CLADDING)	(5.3)	(1.1)	(2.1)	(0.12)	(0.1)	
ALUMINA HEATER (400°C)	0.5	0.3	0.06	0.0014	0.0006	
FUEL AUTOCLAVE WALLS (300°C)	0.046	0.004	0.006	0.002	0.0004	
MAIN WATER TRAP WALLS	0.02	0.09	0.0005	0.0009	0.00004	
WATER IN WATER TRAP	0.12	0.0007	0.01	0.0009	0	
SECOND WATER TRAP	0.008	0.002	0.0008	0.0005	0.00002	
MEMBRANE FILTERS	0.0001	0.0001	0.0001	0.0001	0.00005	
CHARCOAL FILTERS	0.0036	-	-	-	-	
GAS-COLLECTION AUTOCLAVE	0.0001	0.0001	0.0002	0.0003	0.00001	

TABLE 6. DISTRIBUTION OF MATERIAL IN TREAT EXPERIMENT 6: 1000 psia STEAM (RETAINED), TRACE PREIRRADIATION, FUEL AND CLADDING FRAGMENTED

LOCATION	PERCENTAGE IN EACH LOCATION						
	^{131}I	^{129}Te	^{137}Cs	^{103}Ru	^{89}Sr	^{95}Zr	UO_2
				^{106}Ru	^{140}Ba	^{144}Ce	
UO_2 + Zr-2 CLADDING	90	95	77	97.7		98.9	
(Zr-2 CLADDING)	(2.5)	(11)	(5)	(10)		(7)	
ALUMINA HEATER (400°C)	4.4	2.6	11	2.1		1.7	
METAL HEAT REFLECTORS	4.8	0.2	0.5	0.11		0.06	
AUTOCLAVE WALLS AND SPACER (300°C)	0.7	0.2	0.5	0.07		0.05	
WATER IN AUTOCLAVE	0.2	0.004	0.07	0.01		0.0002	

Table 1.2. Distribution of Material in TREAT Experiment 7 in Which Stainless-Steel-Clad UO_2 Was Melted Under Water by Transient Fission Heat Input of 504 cal per Gram of UO_2

Location	Material Found in Each Location (%)								
	^{131}I	^{129}Te	^{137}Cs	^{103}Ru	^{89}Sr	^{140}Ba	^{95}Zr	Ce^{a}	UO_2
Material remaining in fuel autoclave	95	97	95	99.8	99.6	99.1	99.96	99.90	99.92
Melted fuel clinker	18	36	18	55	43	41	43	48	45
Autoclave components including scattered fuel	77	61	77	45	57	58	57	52	55
Material released from fuel autoclave	5	3	5	0.2	0.4	0.9	0.04	0.1	0.08
Condensate in water traps	2	0.2	3	0.06	0.08	0.1	5×10^{-4}	0.01	0.01
Walls of water traps and condenser	3	3	2	0.1	0.2	0.5	0.04	0.09	0.07
Membrane filter papers	0.002	0.001	0.005	9×10^{-5}[b]	0.02	0.2	1×10^{-6}[b]	0.005	7×10^{-6}
Charcoal-loaded papers	6×10^{-4}		0.01	2×10^{-5}		0.02		0.001	
Gas-collection tank	5×10^{-5}	3×10^{-4}	0.03	3×10^{-6}	0.06	0.08	5×10^{-7}[b]	2×10^{-4}	7×10^{-7}

[a] Average of ^{141}Ce and ^{144}Ce.

[b] Plus or minus 100%.

Table 1.3. Distribution of Material in TREAT Fxperiment 8 in Which Zircaloy-2-Clad UO_2 Was Melted Under Water by Transient Fission Heat Input of 511 cal per Gram of UO_2

Location	Material Found in Each Location (%)								
	^{131}I	^{129}Te	^{137}Cs	^{103}Ru	^{89}Sr	^{140}Ba	^{95}Zr	Ce[a]	UO_2
Material remaining in fuel autoclave	93	98	94	99.9	99.5	98.7	99.96	99.9	99.9
Melted fuel clinker	25	43	29	56	58	53	54	55	52
Autoclave components including scattered fuel	68	55	65	44	42	46	46	45	48
Material released from fuel autoclave	7	2	6	0.1	0.5	1.3	0.04	0.1	0.1
Condensate in water traps	0.4	0.3	5.5	0.05	0.2	0.4	3×10^{-4}	0.006	0.07
Walls of water traps and condenser	7	2	0.4	0.07	0.2	0.5	0.04	0.09	0.07
Membrane filter papers	0.005	6×10^{-4}	0.01	4×10^{-5}	0.02	0.2	6×10^{-7}[b]	0.003	1×10^{-5}
Charcoal-loaded papers	0.002		0.003	2×10^{-5}		0.01		4×10^{-4}	
Gas-collection tank	1×10^{-4}[b]	3×10^{-5}[b]	0.06	4×10^{-6}[b]	0.1	0.2	2×10^{-5}	3×10^{-4}	3×10^{-6}

[a] Average of ^{141}Ce and ^{144}Ce.

[b] Plus or minus 100%.

Table 1.3. Distribution of Material in TREAT Experiments 9 and 10Z in Which Stainless Steel-Clad (Experiment 9) and Zircaloy-2-Clad (Experiment 10Z) UO_2 Was Melted Underwater by Transient Fission Heat Input of 520 and 540 cal per Gram of UO_2

Location	Experiment No.	Material Found in Each Location (%)								
		^{131}I	^{137}Cs	^{129}Te	^{140}Ba	^{89}Sr	^{103}Ru	UO_2	^{144}Ce	^{95}Zr
Material remaining in fuel autoclave										
Fuel pieces	9	15	27	42	81	79	76	80	81	84
	10Z	39	34	70	84	85	85	87	87	86
Sample holder and crucible	9	18	15	30	17	20	23	19	19	16
	10Z	7.6	14	17	14	13	14	13	13	13
Water rinse of fuel autoclave walls (filtered)	9	44	44	0.17	0.34	0.073	0.00[illegible]	0.00051	0.00020	0.00024
	10Z	34	33	0.0056	0.16	0.16	0.0014	0.00011	0.00009	0.00002
Fuel autoclave walls	9	4.4	1.8	15	0.91	0.45	1.2	0.93	0.46	0.37
	10Z	6.9	1.6	6.5	1.2	1.3	1.2	0.64	0.58	0.55
Total	9	81	88	87	99.6	99.8	99.5	99.95	99.97	99.99
	10Z	88	83	94	98.9	99.2	99.5	99.94	99.95	99.99
Material released from fuel autoclave										
Condensate in water traps (filtered)	9	16.0	11.7	0.22	0.026	0.047	0.003	0.00081	2.5×10^{-6}	29×10^{-6}
	10Z	10.4	15.0	0.01	0.19	0.085	0.002	0.00057	0.0010	56×10^{-6}
Walls of water traps and condenser	9	2.5	0.62	12.3	0.11	0.041	0.53	0.044	0.026	0.0034
	10Z	1.4	1.7	5.9	0.59	0.68	0.50	0.062	0.054	0.0065
Membrane filter papers[a]	9	0.0001[b]	0.0038	0.0006[b]	0.11	0.0069	18×10^{-6}	18×10^{-6}	35×10^{-6}	1×10^{-6}[b]
	10Z	0.0006	0.0034	0.00004[b]	0.08	0.0037	4×10^{-6}	7×10^{-6}	28×10^{-6}	2×10^{-6}[b]
Charcoal-loaded papers and gas-collection tank[a]	9	0.004	0.049	0.00007[b]	0.14	0.11	5×10^{-6}	41×10^{-6}	76×10^{-6}	5×10^{-6}[b]
	10Z	0.005	0.033	0.00011	0.13	0.067	140×10^{-6}	42×10^{-6}	73×10^{-6}	3×10^{-6}[b]
Total	9	19	12.4	12.6	0.39	0.2[illegible]	0.53	0.045	0.026	0.0035
	10Z	11.8	16.7	5.9	1.06	0.83	0.50	0.063	0.055	0.0065

[a] The relatively large amounts of Cs, Ba, Sr, and Ce in these locations are a result of rare gas precursors formed during the transient and spread throughout the experimental apparatus before decaying into the isotopes analyzed.

[b] Plus or minus 100%.

fuel specimen to catch melted fuel and cladding. Small holes allow access for water and steam. The safety-relief rupture disk directly above the primary vessel contains eight radial ports to absorb recoil. The low-pressure rupture housing on top of the assembly is connected to the primary vessel by small-diameter tubing.

Conclusions

In experiments 7, 8Z, 9, and 10Z at ambient pressure, no significant effect of cladding (stainless steel or Zircaloy-2 material) was evident. In all experiments the fuel and cladding residues appeared to have moved about in the fluid state with very little fragmentation or shattering. The residue was foamy or porous when stainless steel cladding was used, but a nonporous solid was formed when Zircaloy-2 cladding was melted.

For fission heat input of 520 cal per gram of UO_2, 16 to 24% of the stainless steel cladding and 41 to 49% of the Zircaloy-2 cladding reacted with water to form hydrogen. These values agree generally with the amount of reaction found in the ANL metal-water reaction studies in TREAT.

Fission-product release and transport was similar in all four experiments, except that about twice as much of the volatile group (tellurium, cesium, and iodine) was carried out of the fuel autoclave in experiments 9 and 10Z (6 to 19%), probably because of faster steam release. Release of UO_2, ^{144}Ce, and ^{95}Zr from the fuel autoclave was less than 0.1%.

In experiments 9 and 10Z, 33 to 44% of the ^{131}I and ^{137}Cs were found in distilled water rinses of the scraped fuel autoclave walls, even though the autoclaves were boiled to dryness (220 to 325°C) after the transients. This indicates rapid formation of nonvolatile water-soluble iodine and cesium compounds. These elements were also generally dissolved in water collected in the first water trap.

Although 60 to 80% of the ^{131}I was released from the melted fuel and cladding, only 0.0006 to 0.005% was found on the charcoal-loaded papers and in the gas-collection tank. All the iodine on the charcoal papers and in the gas-collection tank was nonreactive or penetrating, based on its sorbability on charcoal. The small amount of nonreactive iodine found

Appendix B - (Ref. 6)

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properties of the collected material. Both condensed vapors and particulates were probably retained on the filter. In steam atmospheres, the filter was maintained at 140°C to assure that steam condensation did not occur on the filter surface.

In the tests with steam atmospheres, the off-gas steam was condensed after leaving the furnace tube. It was interesting to note that a very large percentage of the released fission products was contained in the condensate catch flask. Table 4 shows that the retention of steam-borne fission products by condensation of the fission product laden steam is nearly as effective in removing the fission products as an "absolute" filter.

Table 4. Comparative effectiveness of steam condensation vs. filter in removing released fission products

Method of removal	Per cent fission product removed					
	I^{131}	Te^{132}	Sr^{89-90}	$Cs^{136-137}$	Ru^{103}	Ba^{140}
By "Absolute" filter*	98	99	80	55	92	74
By steam condensation†	97	77	40	80	50	50

* Filter temperature 140°C.

† Average of six experiments, uranium heated at 1215°C in steam.

Summarizing the behavior of the released fission products, it can be stated that for the conditions employed in these tests, deposition on cool tube walls, retention on filters and condensation of steam are effective mechanisms for retaining the evolved radioactive materials within the reactor shell or building.

SUMMARY

Release of strontium, barium and zirconium from heated uranium was lower than most previous theoretical estimates, being <0.5 per cent under all conditions tested.

Cesium and ruthenium behaved as "semi-volatile" elements being released from <1 per cent to about 15 per cent. Their release was dependent on time, temperature and atmosphere.

Xenon, iodine and tellurium were volatile, their release from heated uranium being dependent on time, temperature and atmosphere.

The release of the three volatile elements was strongly dependent on extent of oxidation of the uranium.

Release of the volatile elements was greatest in air, next greatest in steam and least in helium atmospheres for similar conditions of time and temperature.

All fission-product elements except rare gases escaping the heated uranium tended to deposit heavily on tube walls. From 10 to 98 per cent of the released material deposited in the furnace tube used in the present study. When heating occurred in steam atmospheres, condensing the steam caused 40–97 per cent of the evolved fission products to remain in the condensate.

"Absolute" filters removed 50–98 per cent of the evolved fission products, the efficiency depending on the isotope and the type of gas used.

The test parameters used in these experiments tend to maximize the release of fission products. One exception to this is the possibility that at higher irradiation levels, release could possibly be greater due to high concentrations of fission products and radiation damage to the uranium.

This laboratory investigation shows that definite trends and correlations are obtainable for application to the fission-product release question. However, the many complicating factors involved necessitate a conservative attitude when applying the data to hazards evaluations.

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POOR ORIGINAL

STUDIES ON IODINE TRAPPING BY WATER SYSTEMS AT STUDSVIK

[illegible]

KEYWORDS: *fission products, iodine, gases, water, chemical reactions, acidity, performance, containment, spray*

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From large-scale experiments on the iodine removal efficiency of a water pool at 100°C as well as from supporting laboratory runs within the concentration range 10^{-8} to 10^{-5} M, it was observed that permanent trapping was achieved to a great extent even at neutral pH conditions. The efficiency was considerably higher at lower iodine concentration. The fraction permanently trapped approximately corresponded to the theoretically evaluated equilibrium amounts of hypoiodous acid and iodide.

A mathematical model designed for the washout of molecular iodine in reactor containment atmosphere by sprays gives iodine concentration in containment versus spray time. The model takes the degree of hydrolysis of iodine at different pH values and concentrations into consideration. Initial results from spray experiments performed in a 2.5 m^3 tank at pH = 6 to 7.5 and gas phase concentrations around 4×10^{-4}, 5×10^{-6}, and 1.5×10^{-6} kg/m^3 gave half-times due to spraying of about 45, 5, and 2 min. respectively. They confirm the expected strong influence of iodine concentration on washout half-time.

INTRODUCTION

Parts of the program at Studsvik for the investigation of fission product release, transport, and removal processes have been devoted to studies concerning trapping of iodine in water system. The initial plans of the general program were presented five years ago by Linder.[1] In 1966, large-scale and laboratory runs were performed to investigate the trapping efficiency of a boiling water system to remove molecular iodine from steam. These studies were run for the Marviken nuclear power plant project. The results were reported[2] at an IAEA conference in Vienna 1967. Based on the findings from those studies and the theoretical examination of the iodine-water partition coefficient made by Eggleton, a mathematical model of the washout of iodine by sprays was developed[3] during 1967. The model was tested against the initial spray results reported from Japan[4] and a fair agreement was achieved. The model gives an explanation of the change in washout rate with decreasing iodine concentration. The model has been used in safety analysis of two BWR plants in Sweden. Our work up to now has been directed to water sprays without very reactive additives. The use of such sprays instead of very reactive sprays offers obvious advantages with respect to simplicity, chemical stability, and low corrosivity. However, the pH value must be sufficiently high to reach the necessary spray efficiency.

During 1969 a spray facility was designed and built to test our model and to serve as an experimental facility for studies of different removal processes for airborne contaminants. A few initial deposition and spray runs have been carried out up to June 1970. The purpose of this paper is to summarize those of our results and techniques that may be of interest for the analysis of the efficiency of different water systems to retain gaseous molecular iodine.

TRAPPING OF IODINE BY WATER POOLS

From work in the U.K.,[5] it has been demonstrated that water pools can act as efficient traps

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for molecular iodine. It was also reported that the iodine was permanently trapped in the water. To investigate the removal efficiency of a specific but less favorable system for trapping, namely an emergency condensor containing boiling pure water, we performed large-scale experiments in a pool containing 0.5 to 2 m^3 of water. Molecular iodine was injected into the 100°C pool water together with superheated (175 to 320°C) steam. Iodine was sampled in the pool water and in the exhausted steam.

The decontamination factor at pH about 6 increased from 3 to 300 when the iodine concentration was decreased from 10^{-5} to $5 \times 10^{-7} M$ in the water pool. Addition of sodium hydroxide to the pool in runs at high iodine concentration increased the removal efficiency. However, at low iodine concentration the addition of sodium hydroxide gave only a small increase of the decontamination factor in accordance with the initial higher degree of hydrolysis at this condition. The fraction trapped was not released despite additional steam supply or boiling. These results, together with observations from laboratory runs, show that the reversal of the hydrolysis was masked in these studies. This is not in agreement with simple theory of hydrolysis.

Figure 1 shows the typical behavior of molecular iodine in our laboratory boiling water system. The molecular iodine is released together with the produced steam, but the products of hydrolysis are retained despite continued boiling. The amount of iodine permanently trapped approximately corresponded to the amount initially hydrolyzed. This observation constitutes one main premise in our spray model. Other experiments at 100°C and pH = 3 to 8 in a closed system without boiling have indicated a decrease during the first hour of the fraction of iodine that was in molecular form and a simultaneous increase of the fraction hydrolyzed. The reduction at low and high concentration (4×10^{-8} and $4 \times 10^{-6} M$, respectively) was moderate, but for the medium concentration ($4 \times 10^{-7} M$), a tenfold decrease was observed.

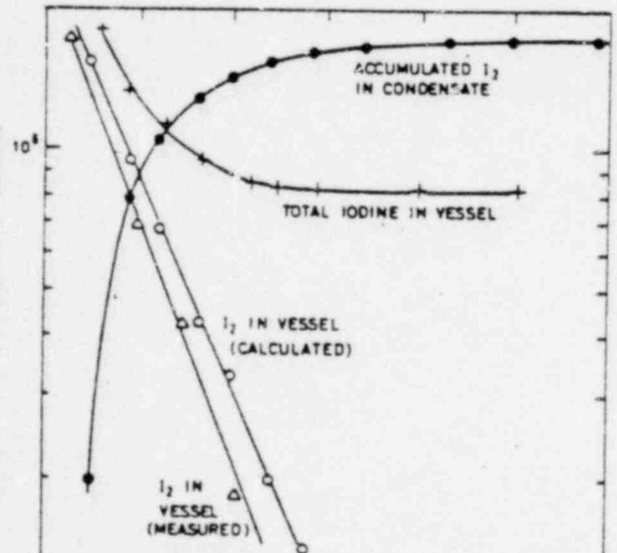


Fig. 1. Iodine release vs boiling time for a water system at pH = 1.3 and iodine concentration = 2×10^{-3} g/liter.

MODEL FOR CALCULATING WASHOUT OF MOLECULAR IODINE BY SPRAYS

For calculation of the washout of molecular iodine in reactor containment atmospheres by sprays, a mathematical model has been developed. The detailed mathematical treatment will be given elsewhere.[3] The model takes the degree of hydrolysis at different pH values and concentrations into consideration and is based on iodine data collected and analyzed by Eggleton.[6] According to results from calculations, the washout behaves differently in three ranges of iodine concentration in the gas phase (cf., Fig. 2). At very high concentration the washout process is liquid-phase-controlled due to the diffusion of molecular iodine into the water drops. In this first region the half-time can be expressed by

$$T_{1/2} = \frac{0.093W}{KF\alpha} \quad ,$$

where

W = containment volume (m^3)

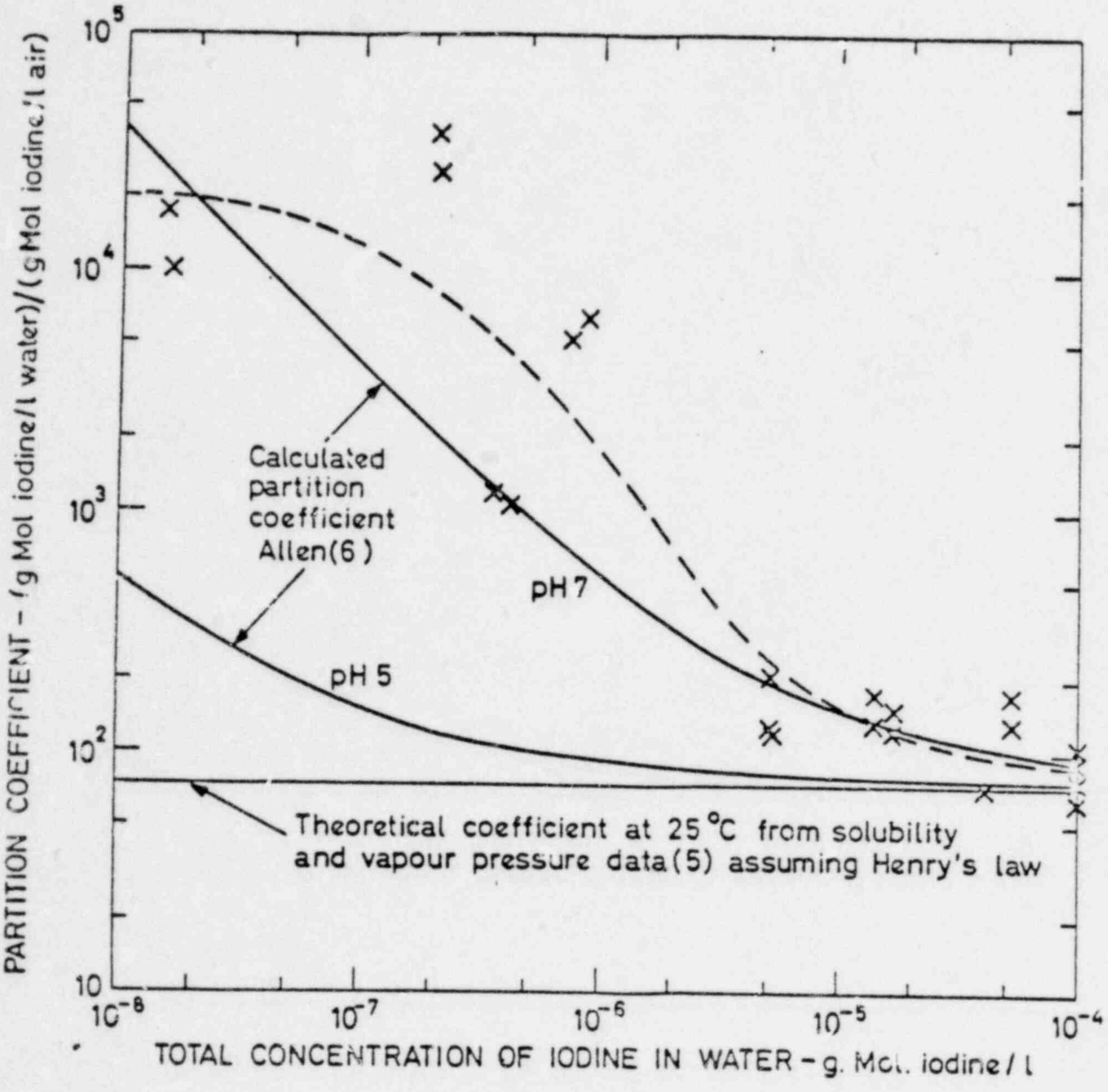
K = partition coefficient for molecular iodine

F = spray water flow rate (m^3/sec)

α = degree of saturation of iodine in the drops.

For this high concentration range the expression reflects the analytical treatment given by Griffiths.[7] In a region of lower concentration the hydrolysis of iodine within the drops becomes more important and increases the washout rate. The rate grows higher for lower concentrations and higher pH values. The mathematical treatment for this second region gave the following final expression for the iodine concentration in the gas phase:

$$C_G = \frac{\beta}{K^2} - \frac{\beta}{K}\,\frac{F\theta}{W}\,\sqrt{\alpha} + \left(\frac{F}{W}\right)^2 \frac{\beta}{4}\,\theta^2\alpha \quad ,$$



EFFECT OF IODINE CONCENTRATION ON THE EQUILIBRIUM PARTITION COEFFICIENT FOR IODINE AT 25 °C.

FIGURE 11. A.E.R.E.-R. 4882.

ORNL-TM-2412
Part IV

Table 9. Results of Calculations, $t = 50^{\circ}C$, pH = 5.0

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RESULTS OF IODINE-WATER PARTITION COEFFICIENT CALCULATIONS
TEMPERATURE =  50.0 DEG C     PH = 5.0
K1 = 2.62E 01 K2 = 4.30E 02 K3 = 4.80E-12 K4 = 2.10E-11
SOLUBILITY OF I2 IN WATER = 2.90E-03 MOL/L

                                            CONCENTRATIONS IN MOLS/L                   ↓                              ↓

CONC I2 IN LIQ     CONC HIO       CONC H2OI+      CONC IODIDE      CONC I3-  TOTAL I2 IN LIQ  LOG TOTAL I      PARTITION COEFF
     2.90E-03      5.62E-05       2.45E-09        2.40E-05        3.22E-05      2.89E-03         -2.539           2.70E 01
     1.40E-03      3.35E-05       1.46E-09        2.01E-05        1.35E-05      1.45E-03         -2.840           2.71E 01
     7.00E-04      2.12E-05       9.25E-10        1.59E-05        5.33E-06      7.27E-04         -3.139           2.72E 01
     3.50E-04      1.40E-05       6.12E-10        1.20E-05        2.02E-06      3.66E-04         -3.436           2.74E 01
     1.75E-04      9.55E-06       4.17E-10        8.81E-06        7.40E-07      1.85E-04         -3.732           2.77E 01
     8.75E-05      6.62E-06       2.89E-10        6.36E-06        2.67E-07      9.44E-05         -4.025           2.83E 01
     4.37E-05      4.64E-06       2.02E-10        4.54E-06        9.53E-08      4.85E-05         -4.314           2.90E 01
     2.19E-05      3.26E-06       1.42E-10        3.23E-06        3.39E-08      2.52E-05         -4.599           3.01E 01
     1.09E-05      2.30E-06       1.00E-10        2.29E-06        1.20E-08      1.32E-05         -4.878           3.17E 01
     5.47E-06      1.62E-06       7.09E-11        1.62E-06        4.25E-09      7.10E-06         -5.149           3.40E 01
     2.73E-06      1.15E-06       5.01E-11        1.15E-06        1.50E-09      3.88E-06         -5.411           3.72E 01
     1.37E-06      8.11E-07       3.54E-11        9.11E-07        5.32E-10      2.18E-06         -5.662           4.18E 01
     6.84E-07      5.73E-07       2.50E-11        5.73E-07        1.98E-10      1.26E-06         -5.901           4.82E 01
     3.42E-07      4.05E-07       1.77E-11        4.05E-07        6.65E-11      7.47E-07         -6.126           5.73E 01
     1.71E-07      2.87E-07       1.25E-11        2.87E-07        2.35E-11      4.58E-07         -6.339           7.02E 01
     8.54E-08      2.03E-07       8.85E-12        2.03E-07        8.31E-12      2.88E-07         -6.540           8.84E 01
     4.27E-09      1.43E-07       6.26E-12        1.43E-07        2.94E-12      1.86E-07         -6.730           1.14E 02
     2.14E-08      1.01E-07       4.43E-12        1.01E-07        1.04E-12      1.23E-07         -6.911           1.51E 02
     1.07E-08      7.17E-08       3.13E-12        7.17E-08        3.67E-13      8.24E-08         -7.084           2.02E 02
     5.34E-09      5.07E-08       2.21E-12        5.07E-08        1.30E-13      5.60E-08         -7.252           2.75E 02
     2.67E-09      3.58E-08       1.56E-12        3.58E-08        4.59E-14      3.85E-08         -7.414           3.78E 02
     1.34E-09      2.53E-08       1.11E-12        2.53E-08        1.62E-14      2.67E-08         -7.574           5.23E 02
     6.68E-10      1.79E-08       7.82E-13        1.79E-08        5.74E-15      1.86E-08         -7.731           7.29E 02
     3.34E-10      1.27E-08       5.53E-13        1.27E-08        2.03E-15      1.30E-08         -7.886           1.02E 03
     1.67E-10      8.96E-09       3.91E-13        9.96E-09        7.17E-16      9.13E-09         -8.040           1.43E 03
     8.34E-11      6.34E-09       2.77E-13        6.34E-09        2.54E-16      6.42E-09         -8.193           2.02E 03
     4.17E-11      4.48E-09       1.96E-13        4.48E-09        8.96E-17      4.52E-09         -8.345           2.84E 03
     2.09E-11      3.17E-09       1.39E-13        3.17E-09        3.17E-17      3.19E-09         -8.496           4.00E 03
     1.04E-11      2.24E-09       9.78E-14        2.24E-09        1.12E-17      2.25E-09         -8.648           5.65E 03
     5.22E-12      1.58E-09       6.91E-14        1.53E-09        3.96E-18      1.59E-09         -8.799           7.98E 03
     2.61E-12      1.12E-09       4.89E-14        1.12E-09        1.40E-18      1.12E-09         -8.950           1.13E 04
     1.30E-12      7.92E-10       3.46E-14        7.92E-10        4.95E-19      7.93E-10         -9.101           1.59E 04
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Table 10. Results of Calculations, $t = 50^\circ$C, pH = 7.0

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RESULTS OF IODINE-WATER PARTITION COEFFICIENT CALCULATIONS
TEMPERATURE =  50.0 DEG C     PH = 7.0
K1 = 2.62E 01 K2 = 4.80E 02 K3 = 4.80E-12 K4 = 2.10E-11
SOLUBILITY OF I2 IN WATER = 2.80E-03 MOL/L

                                         CONCENTRATIONS IN MOLS/L                 ↓                          ↓

CONC I2 IN LIQ     CONC HIO       CONC H2OI+       CONC IODIDE       CONC I3-  TOTAL I2 IN LIQ  LOG TOTAL I      PARTITION COEFF
     2.80E-03       5.62E-04       2.45E-10        2.40E-04        3.22E-04       3.68E-03          -2.434          3.45E 01
     1.40E-03       3.36E-04       1.46E-10        2.01E-04        1.35E-04       1.87E-03          -2.728          3.50E 01
     7.00E-04       2.12E-04       9.26E-11        1.59E-04        5.33E-05       9.65E-04          -3.015          3.61E 01
     3.50E-04       1.40E-04       6.12E-11        1.20E-04        2.02E-05       5.10E-04          -3.292          3.82E 01
     1.75E-04       9.56E-05       4.17E-11        9.82E-05        7.40E-06       2.78E-04          -3.556          4.16E 01
     8.75E-05       6.62E-05       2.89E-11        6.36E-05        2.67E-06       1.56E-04          -3.806          4.68E 01
     4.37E-05       4.64E-05       2.02E-11        4.54E-05        9.53E-07       9.11E-05          -4.041          5.45E 01
     2.19E-05       3.26E-05       1.42E-11        3.23E-05        3.39E-07       5.48E-05          -4.261          6.57E 01
     1.09E-05       2.30E-05       1.00E-11        2.29E-05        1.20E-07       3.41E-05          -4.468          8.16E 01
     5.47E-06       1.62E-05       7.09E-12        1.62E-05        4.25E-08       2.18E-05          -4.662          1.04E 02
     2.73E-06       1.15E-05       5.01E-12        1.15E-05        1.50E-08       1.42E-05          -4.847          1.36E 02
     1.37E-06       8.12E-06       3.54E-12        8.11E-06        5.32E-09       9.49E-06          -5.023          1.82E 02
     6.84E-07       5.74E-06       2.50E-12        5.74E-06        1.89E-09       6.42E-06          -5.192          2.46E 02
     3.42E-07       4.06E-06       1.77E-12        4.06E-06        6.65E-10       4.40E-06          -5.357          3.37E 02
     1.71E-07       2.87E-06       1.25E-12        2.87E-06        2.35E-10       3.04E-06          -5.517          4.66E 02
     8.54E-08       2.03E-06       8.85E-13        2.03E-06        8.31E-11       2.11E-06          -5.675          6.48E 02
     4.27E-08       1.43E-06       6.26E-13        1.43E-06        2.94E-11       1.48E-06          -5.831          9.06E 02
     2.14E-08       1.01E-06       4.42E-13        1.01E-06        1.04E-11       1.04E-06          -5.985          1.27E 03
     1.07E-08       7.17E-07       3.13E-13        7.17E-07        3.67E-12       7.28E-07          -6.138          1.79E 03
     5.34E-09       5.07E-07       2.21E-13        5.07E-07        1.30E-12       5.12E-07          -6.290          2.51E 03
     2.67E-09       3.59E-07       1.56E-13        3.59E-07        4.59E-13       3.61E-07          -6.442          3.54E 03
     1.34E-09       2.54E-07       1.11E-13        2.54E-07        1.62E-13       2.55E-07          -6.594          5.00E 03
     6.68E-10       1.79E-07       7.82E-14        1.79E-07        5.74E-14       1.80E-07          -6.745          7.06E 03
     3.34E-10       1.27E-07       5.53E-14        1.27E-07        2.03E-14       1.27E-07          -6.896          9.98E 03
     1.67E-10       8.96E-08       3.91E-14        8.96E-08        7.17E-15       8.98E-08          -7.047          1.41E 04
     8.34E-11       6.34E-08       2.76E-14        6.34E-08        2.54E-15       6.35E-08          -7.197          1.99E 04
     4.17E-11       4.48E-08       1.96E-14        4.48E-08        8.97E-16       4.49E-08          -7.348          2.82E 04
     2.09E-11       3.17E-08       1.38E-14        3.17E-08        3.17E-16       3.17E-08          -7.499          3.98E 04
     1.04E-11       2.24E-08       9.78E-15        2.24E-08        1.12E-16       2.24E-08          -7.649          5.63E 04
     5.22E-12       1.58E-08       6.91E-15        1.58E-08        3.96E-17       1.59E-08          -7.800          7.96E 04
     2.61E-12       1.12E-08       4.89E-15        1.12E-08        1.40E-17       1.12E-08          -7.951          1.13E 05
     1.30E-12       7.92E-09       3.46E-15        7.92E-09        4.95E-18       7.92E-09          -8.101          1.59E 05
     6.52E-13       5.60E-09       2.44E-15        5.60E-09        1.75E-18       5.60E-09          -8.252          2.25E 05
     3.26E-13       3.96E-09       1.73E-15        3.96E-09        6.19E-19       3.96E-09          -8.402          3.18E 05
     1.63E-13       2.80E-09       1.22E-15        2.80E-09        2.19E-19       2.80E-09          -8.553          4.50E 05
     8.15E-14       1.98E-09       8.64E-16        1.98E-09        7.74E-20       1.98E-09          -8.703          6.37E 05
     4.07E-14       1.40E-09       6.11E-16        1.40E-09        2.74E-20       1.40E-09          -8.854          9.01E 05
     2.04E-14       9.90E-10       4.32E-16        9.90E-10        9.68E-21       9.90E-10          -9.004          1.27E 06
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half-lives defined above. However, for prolonged spray at lower temperature, rate of decrease of iodine concentration has a tendency to saturate, and at higher temperature rate of decrease nearly follows the rule of the first-order reaction. If the partition equilibrium between water phase and gas phase in the vessel is realized, airborne iodine concentration can be represented by the following equation from material balance:

$$c_1 = \frac{c_0}{\frac{(H-1)(200+Ft)}{V} + 1} \quad , \tag{2}$$

where

c_1 = iodine concentration in air, mg/liter

c_0 = initial iodine concentration, mg/liter

H = partition coefficient of iodine, concentration in liquid/concentration in gas

F = spray rate, liter/h

V = volume of vessel, liter

t = spray time, h.

The curve c_1 in Fig. 8 is the relation between c_1 and t, taking 100 liter/h as the spray rate, 100 as the partition coefficient, and 4900 liter as the vessel volume. The curve c_1 will shift to c_1' because 200 liters of water initially charged contribute slowly to the gas-liquid equilibrium.

Another model that can be assumed is that the partition equilibrium is realized at all times between spray drop and gas phase while drop falls, and that iodine transferred to water does not return to the gas phase. Then the rate of decrease of the airborne iodine concentration can be represented as follows:

$$-\frac{dc_2}{dt} = \frac{HF}{V}\, c_2 \quad . \tag{3}$$

We have by integration,

$$c_2 = c_0 \exp\left(-\frac{HF}{V}\, t\right) \quad , \tag{4}$$

where each symbol corresponds to that of Eq. (2). The straight line, c_2 in Fig. 8, shows the relation between c_2 and t when c_0 is unity. Comparing Eqs. (2) and (4) with the experimental curves it appears that the curve for the room temperature can be approximated by Eq. (2), while the curves for the initial temperature of 125 and 150°C do not indicate a tendency to saturate and they can be represented by Eq. (4). The curve for 100°C may show a transient condition between Eqs. (2) and (4).

Gas-liquid partition coefficients for iodine and iodides are determined to make clear the washout effect for iodine. The determination is performed at 25 to 80°C using a glass vessel, distilled water, and ^{131}I; the results are shown in Fig. 9. At any temperature, the partition coefficient H is much larger at the lower concentration than that at the higher concentration. At much higher concentration near saturation, however, H becomes constant probably due to the realization of Henry's law. The iodine concentrations at which H becomes constant would be higher at temperatures of 50 and 80°C than at room temperature because the equilibrium constant for hydrolysis of iodine increases with increasing temperature.[8] Since the relation between the airborne iodine and H may be represented by straight lines as shown in Fig. 9, the following equations are obtained from the equations of the straight lines and Eq. (3):

$$-\frac{dc_2}{dt} = \frac{88F}{V}\, c_2 \;(25°\text{C},\; c_2 > 0.5 \text{ mg/liter}) \tag{5}$$

$$-\frac{dc_2}{dt} = \frac{70F}{V}\, c_2^{0.67} \;(25°\text{C},\; c_2 < 0.5 \text{ mg/liter}) \tag{6}$$

$$-\frac{dc_2}{dt} = \frac{53F}{V}\, c_2^{0.67} \;(50°\text{C}) \tag{7}$$

$$-\frac{dc_2}{dt} = \frac{39F}{V}\, c_2^{0.67} \;(80°\text{C}) \quad . \tag{8}$$

Equations (5) through (8), with 4 mg/liter as the initial iodine concentration and 100 liter/h as the spray rate, are shown in Fig. 10 as three curves.

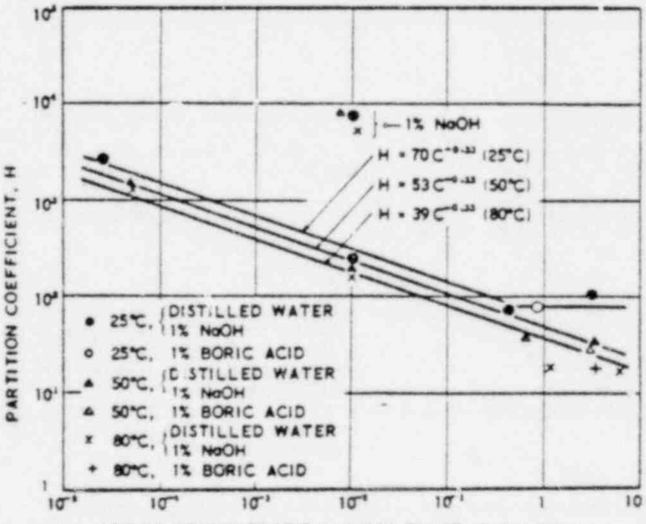

Fig. 9. Partition coefficient vs iodine concentration in gas phase.

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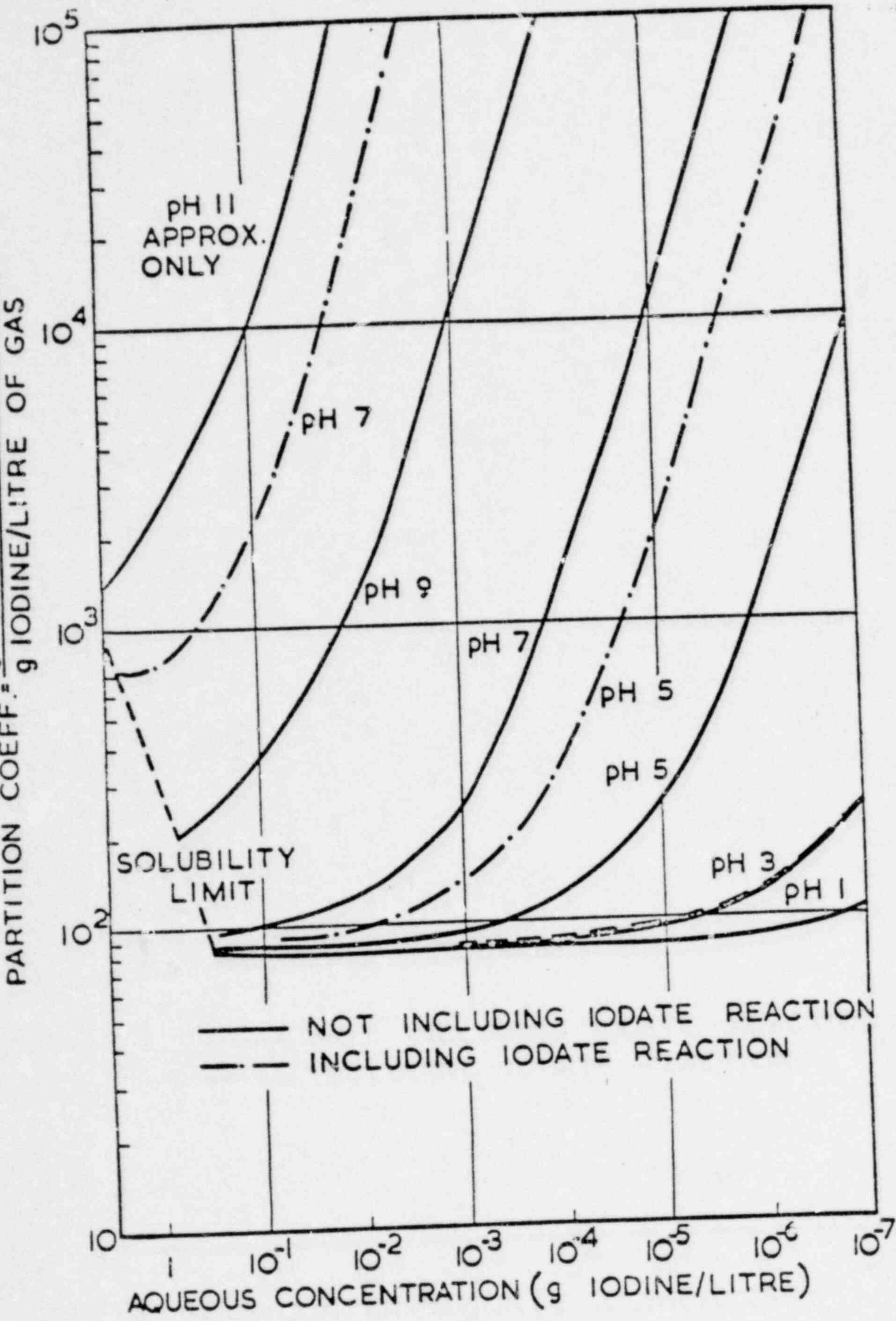
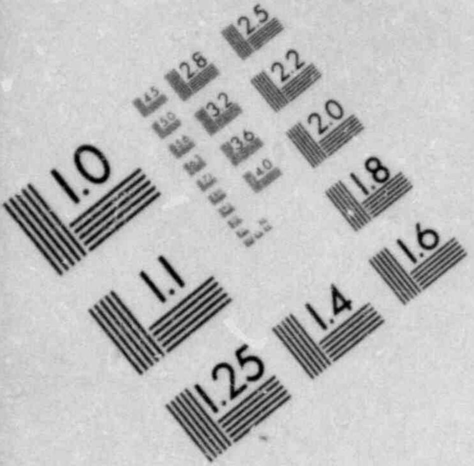


FIG.1. PARTITION OF IODINE BETWEEN GAS PHASE AND WATER AT 25°C



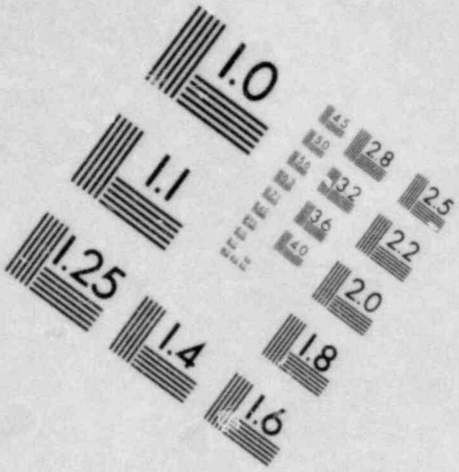
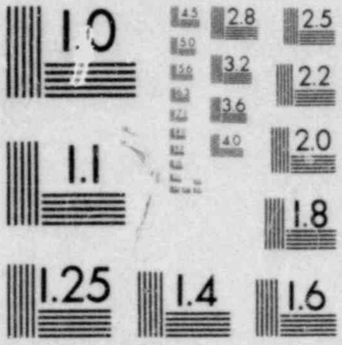
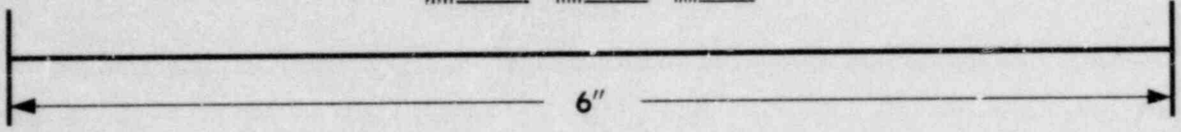
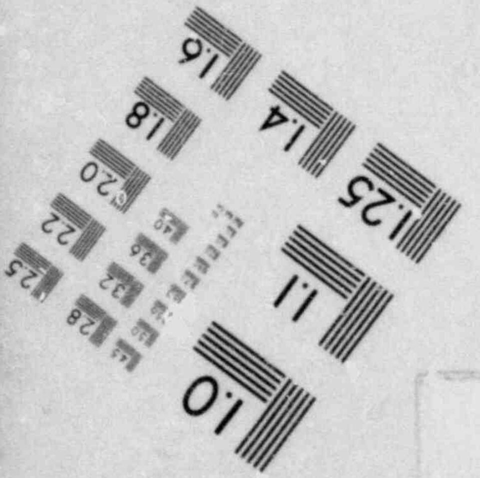
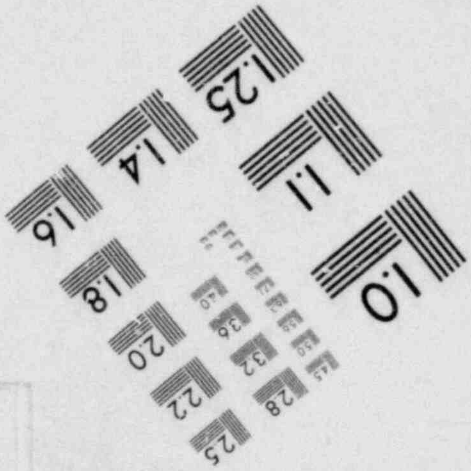


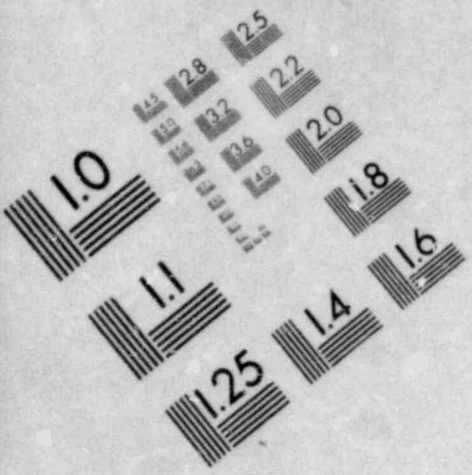
IMAGE EVALUATION TEST TARGET (MT-3)











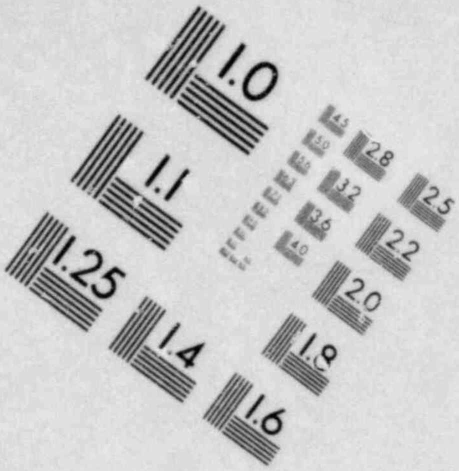
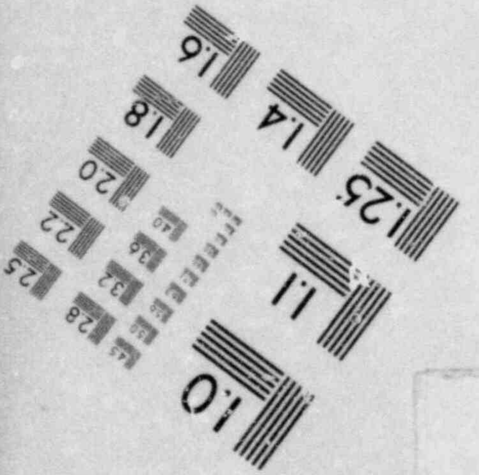
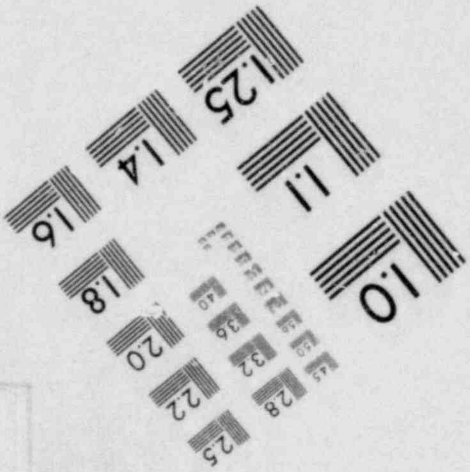
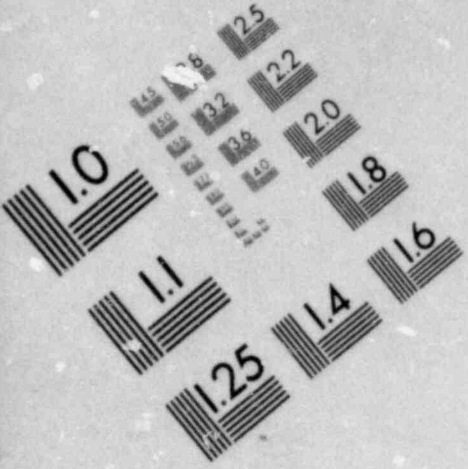


IMAGE EVALUATION TEST TARGET (MT-3)







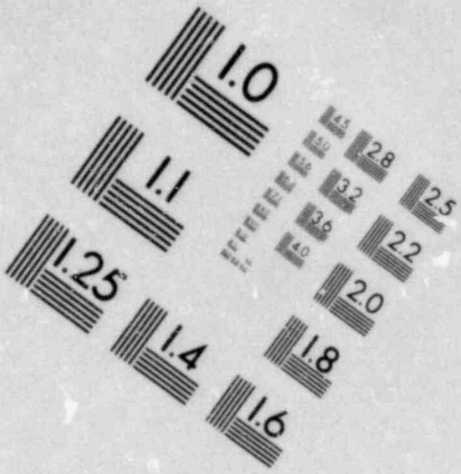
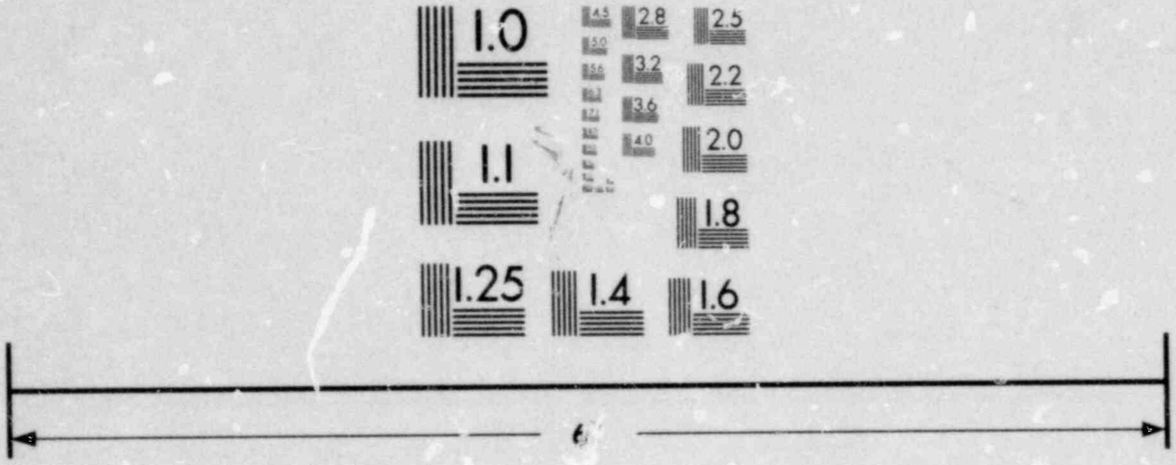
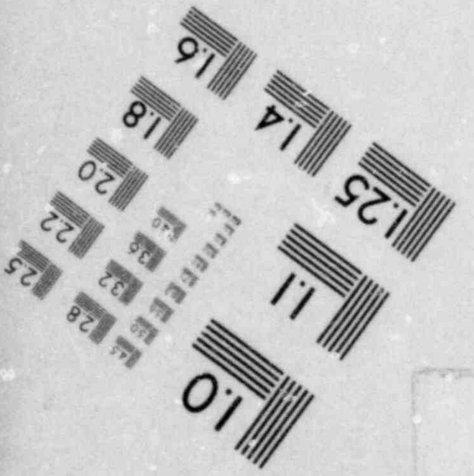
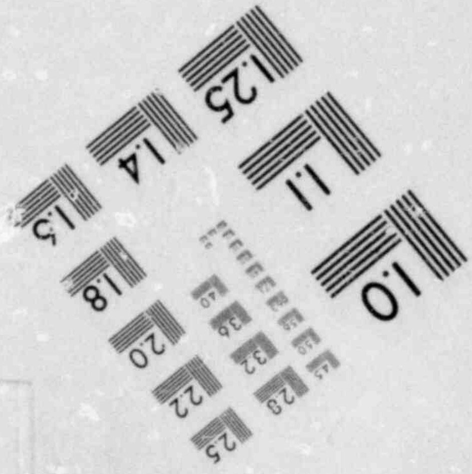


IMAGE EVALUATION
TEST TARGET (MT-3)







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Étude expérimentale du comportement des produits de fission en cas d'accident sur un réacteur piscine

Par J. DADILLON *

RÉSUMÉ :

Un élément essentiel et cependant parfois négligé parce que mal connu, dans l'évaluation des risques nucléaires associés au fonctionnement d'un réacteur, est la connaissance, en cas d'accident, de la nature, de l'importance et du comportement de la contamination réellement libérée, à l'intérieur de l'enceinte.

Dans le cas particulier des réacteurs piscines, le fluide de refroidissement s'avère être une barrière, très efficace vis-à-vis de la contamination.

Trois expériences ont été faites dans le réacteur CABRI au cours desquelles, a été provoquée au sein même du cœur, la fusion de quelques plaques d'éléments combustibles.

SUMMARY:

In the estimation of nuclear risks connected with the running of a reactor an essential factor, sometimes neglected because insufficiently known, is the knowledge of the type, amount and behaviour of the contamination actually released inside the container in the case of an accident.

In the special case of swimming-pool reactors the cooling fluid proves to be a very efficient barrier against contamination. Three experiments were carried out in the reactor CABRI, during which several fuel element plates were melted inside the core itself.

Sans présumer de l'ampleur maximale de la fusion susceptible de se produire dans le cœur du réacteur, il est primordial de connaître, en premier lieu, l'ordre de grandeur du taux d'émission à partir du combustible, des produits de fission les plus dangereux et, en second lieu, le pouvoir de rétention du fluide de refroidissement. Le problème, extrêmement complexe dans son ensemble, se trouve simplifié lorsqu'il s'adresse à un type de réacteur déterminé.

En ce qui concerne les réacteurs piscines, il a été choisi de commencer cette étude dans des conditions très voisines de la réalité, ce qui rend l'utilisation des résultats beaucoup moins délicate. Malheureusement, la cinétique et les conditions

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d'une fusion accidentelle d'une partie du cœur d'un réacteur risquent de ne pas être toujours rigoureusement identiques : une extrapolation peut alors s'avérer nécessaire.

Les trois expériences réalisées dans le réacteur CABRI ont permis de déterminer les taux d'émission de l'iode et du tellure à partir du combustible fondu, ainsi que le pouvoir de rétention de l'eau. Le niveau, toujours très bas du taux d'irradiation des éléments combustibles de la pile (quelques dizaines de MWJ/T) et l'ampleur de la fusion volontairement limitée à quelques plaques (une pour la première expérience, trois pour les suivantes), nous ont contraints à nous limiter aux produits les plus volatils en portant toute notre attention sur l'iode qui est le plus dangereux d'entre eux.

Les deux premières expériences ont été faites au cours de tests du comportement de la pile dans certaines conditions de température et de débit, la troisième a été spécialement préparée pour l'étude de la diffusion de la contamination libérée par la fusion du combustible.

Lors de chaque test, la température moyenne de l'eau était de 30° C et son débit, à travers le cœur de 400 m³/h environ, la circulation se faisant toujours de haut en bas. Le niveau d'eau a varié de 5,30 m à 6,10 m, ce qui correspond à des hauteurs d'eau au-dessus du cœur de 2,50 m à 3,30 m.

De nombreux prélèvements effectués dans l'eau, à différents instants après la fusion, ont montré que la contamination était très homogène.

I. TAUX D'ÉMISSION DES PRODUITS DE FISSION A PARTIR DE COMBUSTIBLE FONDU

Nous rappellerons qu'un chargement complet du cœur de CABRI se compose d'environ 300 plaques d'alliage uranium-aluminium (en poids : 20 % d'uranium - 80 % d'aluminium) représentant au total 4 kg d'uranium enrichi à 93 % en U-235 (90 % à l'époque où s'est déroulée la présente expérimentation).

A l'issue des deux dernières expériences, plusieurs échantillons de combustible ont été prélevés sur les plaques, tant dans les parties intactes que dans les parties fondues. Leur analyse a permis de déterminer le poids d'alliage disparu et les quantités d'iode présentes avant chaque fusion. Par ailleurs, le calcul a fourni des valeurs comparatives concernant l'iode 131 ainsi que les activités des autres iodes et du tellure 132.

La transmission à travers l'eau étant très faible comme on le verra, le taux d'émission d'un produit de fission à partir du combustible, est égal au rapport de son activité dans l'eau à celle qui se trouvait dans les plaques avant leur fusion. Il est bien évident que la partie, non endommagée du cœur, n'entre pas dans ce calcul.

Le taux d'émission de l'iode a varié de 2 à 10 % suivant l'expérience. Celui du tellure de 0,2 à 5 %.

Nous l'avons déjà dit, le taux d'émission des produits de fission, au cours d'une fusion de combustible, est un phénomène beaucoup trop complexe pour qu'il soit possible, actuellement, d'en fixer les valeurs avec certitude, surtout quand il s'agit des produits les plus volatils. Seule une valeur probable peut être avancée : ainsi, dans le cas d'un réacteur à eau, une fusion même très étendue du cœur, mais limitée dans le temps à quelques dizaines de secondes, ne devrait pas provoquer une émission d'iode et de tellure dans l'eau, supérieure à 10 %. Par contre, le taux d'émission des gaz mesuré de façon peu précise à partir de leurs descendants solides, pourrait avoisiner 100 %. Quant aux produits solides ou peu volatils, ils n'ont pas été détectés dans l'eau, à l'issue des présentes expériences. Cependant, leur taux d'émission à partir d'un combustible déterminé est beaucoup moins influencé par les conditions de fusion et demeure, de toute façon, très faible. Les valeurs déterminées au cours d'expériences à petite échelle, effectuées notamment aux États-Unis, peuvent être considérées comme parfaitement acceptables.

L'analyse des échantillons prélevés sur les plaques a fait apparaître une perte d'uranium de l'ordre de 70 %, même en des endroits où la gaine n'avait pas fondu. La présence entre les plaques, de coulées d'alliage, laisse supposer que celui-ci peut se trouver en fusion sans que la gaine, plus refroidie, ne soit fondue et que, de ce fait, il s'écoule par les points où la gaine a réellement fondu.

II. PIÉGEAGE DES PRODUITS DE FISSION DANS L'EAU

Seule la rétention de l'iode dans l'eau a pu être déterminée au cours de ces expériences. En effet, ce facteur étant très important, les activités qui atteignent le hall-pile sont très faibles. En ce qui concerne les gaz, dont la rétention est moins grande, les systèmes de piégeage dont nous pouvions disposer, ne permettaient pas de faire des prélèvements sélectifs suffisamment longs.

Le facteur de transmission de l'iode à travers l'eau s'exprime de la façon suivante :

$$k = \frac{AH}{AE + AH}$$

AH activité de l'iode émis dans le hall-pile.

AE activité de l'iode dans l'eau.

Suivant les expériences, les isotopes et les prélèvements à partir desquels il a été calculé, ce coefficient se situe entre $5\ 10^{-6}$ et $5\ 10^{-5}$. Autrement dit, seule une partie comprise entre 0,0005 % et 0,005 % de l'iode émis dans l'eau pendant la fusion du combustible, atteint le hall-pile lui-même.

En outre, une série d'expériences hors pile (PIREE) à une échelle voisine de la réalité, a montré que, même dans le cas où l'iode était entraîné dans l'eau par un gaz sous une pression de 20 kg/cm², à une température de 400° C et suivant un débit important de 20 g/s pendant une minute, seule une partie égale à 0,1 % traversait une couche d'eau de 3 mètres.

Par conséquent, dans le cas d'un accident de fusion de combustible survenant dans un réacteur piscine, on peut considérer comme valeur probable du coefficient de transmission de l'iode à travers l'eau 0,005 % et comme limite supérieure 0,1 % étant précisé qu'à notre sens, on resterait certainement assez éloigné de cette valeur limite.

III. COMPORTEMENT DE L'IODE DANS LES FILTRES

Deux dispositifs expérimentaux de piégeage ont été utilisés au cours de cette expérimentation :

— des dispositifs simples composés d'un papier Delbag suivi de 5 cm de charbon ;

— des ensembles composites comprenant en nombre variable suivant les cas :

- papiers d'amiante, laine de cuivre activée à l'argent, papiers imprégnés au charbon, couches de 1 cm de charbon.

Les papiers amiante et le charbon étaient du même type que ceux utilisés dans les installations nucléaires industrielles.

Seul, le premier dispositif a été utilisé lors des deux premières expériences ; sa constitution même ne permettait pas d'obtenir des informations sur le comportement de l'iode. Il est seulement possible de dire que le papier Delbag retient des quantités d'iode inférieures à 10 % de celles adsorbées dans le charbon, ce qui signifierait que moins de 10 % de l'iode se trouvent associés à des aérosols.

Pour la troisième expérience, une enceinte de confinement était placée au-dessus de la cuve du réacteur, réduisant le volume libre, au-dessus de l'eau, à 150 m³ seulement au lieu de 2.500 m³ qui représentent, approximativement, le volume total du hall. En outre, des ensembles composites étaient cette fois utilisés.

Une assez forte quantité de vapeur apparaissant au-dessus du réacteur au cours de la montée en puissance, le taux d'humidité relative atteignait 95 % à l'intérieur de l'enceinte de confinement.

Les quantités d'iode fixées sur les couches successives de charbon d'un même ensemble, étaient souvent du même ordre, ce phénomène étant dû, en premier lieu, au taux d'humidité élevé qui a diminué, dans des proportions considérables, l'efficacité des charbons. Il est impossible dans ces conditions de déterminer le pouvoir d'arrêt réel de ces ensembles, mais les diverses couches de charbon ont présenté des efficacités qui, ramenées à la valeur de 5 cm de charbon seraient généralement inférieures à 95 %. Le taux d'humidité n'est probablement pas la seule cause de cette diminution d'efficacité des charbons, la formation de composés moins bien arrêtés que l'iode moléculaire pourrait être une explication très vraisemblable.

IV. CONCLUSION

Il apparaît que les conséquences d'un accident mettant en cause la fusion d'une partie du cœur d'un réacteur piscine, ne seraient pas aussi graves que ce que l'on pouvait penser jusqu'à présent. Ces quelques expériences ont montré que le combustible, l'eau et les matériaux constitutifs à l'intérieur du cœur retenaient 99,9995 % de l'iode présent dans la portion de cœur endommagée. Il n'est pas possible de généraliser encore au cas de fusions extrêmement brutales engendrant de grandes quantités de vapeur. Néanmoins il n'est pas déraisonnable de penser que dans la très grande majorité d'accidents entrainant la détérioration d'une portion du cœur seul 0,1 % des iodes serait émis au-dessus des piscines.

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