UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D. C. 20555

SEP 28 1979

Mr. Vincent L. Johnson
Director, Technical Staff
President's Commission
on the Accident at Three Mile Island
2100 M Street, N. W.
Washington, D. C. 20037

Dear Mr. Johnson:

Your letter of July 20, 1979 to Mr. Rehm requesting information on the fate of the iodine isotopes after the Three Mile Island accident has been referred to me for reply.

As you have surmised, we do not have sufficient information to make a complete material balance for the noble gases and iodine isotopes which were released from the damaged fuel. However, from what is known of the Three Mile Island (TMI) accident specifically and the chemical behavior of iodine and noble gases generically, a model can be constructed to explain the large release of noble gases compared to the small release of iodines. In brief, the relatively low amount of iodine released is principally explained by its strong tendencies to remain dissolved in liquids, plateout on surfaces, and become adsorbed on charcoal filters.

As a result of the accident involving TMI, Unit 2, approximately 14 million curies of noble gases (mainly Xe-133, 8 million curies) and 15 curies of iodine-131 were released into the atmosphere in the first month following the accident. Since that time, periodic releases have become relatively insignificant. Although several factors are believed to have influenced this difference in release magnitudes, the extent of that influence has not been precisely quantified for all factors involved at TMI. In order to determine the reasonableness of the xenon and iodine release estimates, it is necessary to consider the following parameters, which are factors influencing releases: pre-accident radionuclide core inventory, fraction of the core inventory released to the coolant, fraction of each radionuclide partitioned into the gaseous phase and the fraction of each radionuclide which is removed by filtration, plateout or other processes.

Prior to the March 28 accident, the core inventory of Xe-133 and I-131 is calculated to have been 1.4×10^8 and 6.5 by 10^7 curies respectively. It has been estimated that 60% of core xenon and 50% of core iodine was released from damaged nuclear fuel into the reactor coolant system (RCS) as a result of the high temperatures observed during the accident.

The radioactive gas releases to the environment resulted from the offgassing and leakage of contaminated liquids which reached the auxiliary building by various sources. Little radioactive gas was released from the reactor building itself.

Essentially all of the xenon-133 that was released is believed to have entered the gaseous phase in the auxiliary building. This assumption is made because noble gases are chemically very insoluble and because the large pressure drop between the RCS and the auxiliary building would tend to drive any entrained noble gases out of the liquid phase. Iodine species, however, are known to be very soluble in water and relatively small fractions are believed to have partitioned into the gas phase during the accident at TMI. As a reference point, the NRC (NUREG-0017) uses partition factors of 0.0075 for iodine and 1.0 for xenon, in developing source term estimates for nuclear plants operating under normal conditions. Although this parameter has not been precisely quantified for the TMI accident an approximation of the iodine partitioning which occurred following the accident at TMI Unit 2 can be obtained based on certain data collected since the accident. The ratio of I-131 activity levels in the containment building air to that in the containment building sump is an estimate of the partition factor of the water that leaked to the sump. In addition the activity levels in the containment building air to that in the reactor coolant system would be an estimate of the partition factor since it is a measure of how much of the activity in reactor coolant system leakage partitioned into the containment building air. Although there is some uncertainty in the containment building, sump and reactor coolant measurements, a review of the data in Tables 1 and 2, indicates general agreement between these two methods of estimating partition factor with an estimated partition factor for iodine of between 5×10^{-6} and 6×10^{-5} probably occurring during the accident. This is somewhat higher than that which is estimated in NUREG-0017 and the actual value may well be somewhere in between the NUREG-0017 estimate and the measurements. Regardless of its precise value, this factor is believed to be the most significant contributor to the difference in observed xenon and iodine release levels.

The iodine removal processes of plate-out and filtration have also affected the TMI release magnitudes. The NRC (Regulatory Guide 1.3 and 1.4) assumes, in its post-accident source term estimates, that 50% of iodines in the gas phase will plate-out on equipment building internals. Conversely, xenon, because it is chemically inert, does not attach itself on these surfaces. No precise data for plate-out at TMI-2 is available although its effect would probably be included in the partition factor measurements of Table 1 and 2.

All gaseous releases resulting from the March 28 accident were passed through charcoal filters located in the auxiliary building. Two parallel filter trains, each rated for 30,000 cfm flow, filtered ventilation air from the auxiliary building while another set of parallel trains filtered flow from the fuel handling building. Laboratory tests performed on charcoal samples, which were in-place during the period of significant release, indicate that these filters removed approximately 85% of the I-131 in the flow. Since noble gases are not filtered in these systems, the iodine-131 to xenon-133 release ratio can be assumed to have decreased.

In addition to the above discussion, Attachment 1 provides a discussion of various measurements taken at TMI Unit 2 which support the thesis that iodine tends to remain in solution in the various water systems in the plant.

Although all factors which influenced the xenon to iodine ratio of about one million which was experienced in the atmospheric releases have not been quantified, there is a good basis for expecting it. Those factors contributing to the high xenon to iodine ratio are discussed in the enclosure. The NRC is continuing to update this analysis as further information becomes available and these results should provide better characterization of source terms used in our accident analyses.

Sincerely,

Harold R. Denton, Director
Office of Nuclear Reactor Regulation

Enclosures:
1. Table 1
2. Table 2
3. Fission Gas and Iodine Releases from TMI-2

TABLE 1

ESTIMATED IODINE PARTITION FACTOR

BASED ON MEASURED TMI 2 CONTAINMENT BUILDING

AND SUMP ACTIVITY LEVELS

Date of Measurement	Activity Level In Containment Building (μci/cc)	Activity Level In Containment Sump (μci/cc)*	Estimated Partition Factor**
3/31	0.063	4.8×10^{3}	1.3×10^{-5}
5/4	0.0028	2.6×10^{2}	1.1×10^{-5}
5/14	0.0013	1.1×10^{2}	1.2×10^{-5}
5/27	0.0014	3.6×10^{1}	3.9×10^{-5}
5/30	0.0017	2.7×10^{1}	6×10^{-5}
5/31	0.001	2.5×10^{1}	4×10^{-5}
6/21	1.2×10^{-4}	4.1	3×10^{-5}
6/26	1.2×10^{-4}	2.7	4.4×10^{-5}
7/14	3×10^{-5}	0.57	5.2×10^{-5}

* Based on containment sump measurement and corrected back for decay to corresponding containment building air measurements.

** Partition Factor = $\frac{\text{Activity Level in Containment Building (μci/cc)}}{\text{Activity Level in Containment Sump (μci/cc)}}$

TABLE 2

ESTIMATED IODINE PARTITION FACTOR

BASED ON MEASURED TMI 2 CONTAINMENT BUILDING

AND REACTOR COOLANT SYSTEM ACTIVITY LEVELS

Date of Measurement	Activity Level In Containment Building (μci/cc)	Activity Level In Reactor Coolant (μci/cc)	Estimated Partition Factor*
3/31	0.063	1.2×10^{4}	5.2×10^{-6}
5/4	0.0028	6.0×10^{2}	4.7×10^{-6}
5/14	0.0013	2.8×10^{2}	4.6×10^{-6}

* Partition Factor = $\frac{\text{Activity Level in Containment Building (μci/cc)}}{\text{Activity Level in Reactor Coolant System (μci/cc)}}$

Fission Gas and Iodine Releases from TMI-2

Introduction

The fission products volatilized from zircaloy-clad UO_2 fuel elements when they fail from overheating are the xenons, the kryptons, the iodines, and the cesiums. Much lower amounts of other fission products are released. The cesium is thought to volatilize as hydroxide, oxide, metal vapor, or cesium iodide. Under reducing conditions in contact with reactor coolant, the likely chemical species in the coolant would be Cs^+, I^-, Xe°, and Kr°. Laboratory tests have shown that low percentages of these nuclides are released from irradiated UO at temperatures below 2400°F. In the TMI-2 accident, the high percentages of fission gases, iodine and cesium found in the reactor coolant samples or the containment building air samples (see below) indicate that temperatures well above 2400°F were reached in approximately one third of the core. This is consistent with the estimate from the amount of hydrogen produced during the accident that about 40% of the zircaloy reacted with steam to form ZrO_2. At these high temperatures, similar fractions (about 30 to 60%) of the core inventories of fission gases, iodine and cesium should be released from the fuel.

Containment Air Analyses

As the gas bubble in the reactor coolant was vented to the reactor containment atmosphere, the gas would be expected to carry much of the released fission products which volatilized from water solution. An additional amount of volatile fission products would be released from reactor coolant which spilled out to the containment building floor. The analyses of the first three samples of containment air are given in Table 1.

For the three measurements, core inventory fractions of noble gases released to the containment tended to vary.

However, the values of 33% of inventory for Kr-85, 37% for Xe-135, and the values of 29% to 62% for Xe-133 are considered by the staff to be the more reliable values, since later analyses for these isotopes were consistent with releases of 30% or more of the core inventory of the volatile fission products. An additional 10% of inventory of Xe-133 was estimated to be released to the environment outside the plant. The staff considers, then, that at least 30% of the Xe-133 inventory was released from the core to the containment air and/or to the environment. Radiologically, Xe-133 is the most important noble gas fission product.

On the other hand, for these three measurements only 0.007% of the core inventory of I-131 appeared in the containment air. The ratio of fractions of containment inventory of xenon to iodine is thus about 30% ÷ 0.007%, or more than 4000.

The absence of I-131 in the containment air may also be in part because all known iodine compounds plate out more readily from the gas phase on solid surfaces than do noble gases. In addition, the tendency for iodine to remain in solution would also be increased by the NaOH added to the reactor coolant and the containment building with the water sprays during the accident.

It is, therefore, consistent with the known chemistry of iodine that, under the conditions of the TMI-2 accident, iodine should have a strong tendency to remain dissolved in reactor coolant water. On the other hand, the known solution properties of noble gases indicate that their solubility in water is low, particularly at elevated temperatures.

Reactor Coolant Analyses

Significant information regarding the fate of iodine isotopes after the TMI-2 accident may be adduced from the analyses of reactor coolant samples shortly after the accident. Table II presents the analysis of a sample of reactor coolant taken 3/29/79 and Table III represents the analysis of a sample of the reactor coolant sampled on 4/10/79.

It is seen in Table II that the inventory percentages of iodine and cesium in the coolant are approximately equal for the reactor coolant sample taken on 3/29/79. The same is approximately true for the reactor coolant sample taken on 4/10/79. These results are consistent with the Oak Ridge test results that at high temperatures equal fractions of iodine and cesium fuel inventories would be released from UO_2 fuel.

Also to be noted in Table III is that the fractions of inventory of cesium and iodine released to the coolant are of the order of 13% to 16% of core inventory.

Further, in agreement with the predictions of laboratory studies, the fractions of inventory released of I-131, Cs-137 and Cs-136 are similar. (The fraction of Cs-134 inventory released is unaccountably about 8% lower). This correspondence is useful in that the behavior of Cs-137 and Cs-136 isotopes may be taken to be representative of the liquid phase behavior of the iodine isotopes. In particular, the thesis is confirmed that the iodine isotopes, under the TMI-2 accident conditions, remain largely in the reactor coolant solution as do the cesium isotopes.

Auxiliary Building Tank Analyses

Leaks from the reactor coolant system, including the reactor and volume control system (CVCS), introduced radioactive materials into the seven tanks in the auxiliary building. Samples from each tank were analyzed and corrected for decay from 3/28/79. For each isotope, the corrected activity was divided by the total core inventory on 3/28/79, as given by the ORIGEN code. Table IV presents these values as percentages.

Table I

Percent of Core Inventory in Containment Air

Isotope	Core Inventory, Ci[b]	Percent of Core Inventory[a]		
		Sample 1	Sample 2	Sample 3
Kr-85	1.09E5	-	-	33
Xe-131M	4.32E5	-	-	3.7
Xe-133m	2.12E7	6.8	-	-
Xe-133	1.42E8	29.	62	46.
Xe-135	3.31E7	37.	-	-
I-131	6.55E7	0.0070	-	0.0065

a
Sample 1 was counted at 0700 hr, 3/31/79, Sample 2 at 1030 hr, 4/2/79, and Sample 3 at 2130 hr, 5/5/79. The activities were corrected for decay.

b
The core inventory at 0400 hr, 3/28/9 was calculated by the ORIGEN computer code.

c
1.09E5 represents 1.09×10^5.

Table II

Analysis of First Reactor Coolant Sample (3/29/79)

Nuclide	Core Inventory[a]	Percent of Core Inventory in Reactor Coolant[b]
I-131	6.55E7	9.6
I-133	1.50E8	8.5
Cs-134	3.48E5	6.9
Cs-136	7.38E5	10.
Cs-137	9.50E5	11.
Sr-89	6.68E7	(0.0031)
Sr-90	8.75E5	( )
Ru-106	1.21E7	0.0039
Ba-140	1.33E8	0.0066
Te-132	9.76E7	0.12

[a] Calculated for 0400 hr, 3/28/79 by the ORIGEN code.

[b] Based on a primary coolant volume of 3.8×10^8 ml. If make-up water from the borated water storage tank is included and considerd to be at the same concentration, the fraction of core inventory in the coolant would be about a factor of 3 higher.

Table III

Analysis of Second Reactor Coolant Sample (4/10/79)

Nuclide	Core Inventory[a]	% of Core Inventory in Reactor Coolant[b]			
		SRL[c]	ORNL	Bettis	B&W
I-131	6.55E7	14.	15.5	16.	13.
Cs-134	3.48E5	8.6	9.1	8.3	8.2
Cs-136	7.38E5	12.	12.	12.	10.
Cs-137	9.50E5	13.	13.	13.	11.
Sr-89	6.68E7	( )	0.42	(0.14)	-
Sr-90	8.75E5	(0.60)	2.2	(0.14)	-
Ba-140	1.33E8	0.15	0.18	0.14	0.094
La-140	1.38E8	0.075	0.86	0.075	-
Mo-99	1.36E8	1.2	1.7	-	1.2
Ce-141	1.04E8	0.051	-	-	-
H-3	4.48E3	-	10.	-	-

a Calculated for 0400 hr, 3/28/79 by the ORIGEN code.

b Based on a primary coolant volume of 3.8×10^8 ml. If the make-up water in the borated water storage tank is included and considered to be at the same concentration, the fraction of the core inventory in the coolant would be about a factor of 3 higher.

c This sample was analyzed by the four laboratories indicated.

Table IV

Quantities of Principal Nuclides in TMI-2
Auxiliary Building Tanks

Isotope	Core Inventory, Ci*	% of Core Inventory in Tanks
I-131	6.55 x E7	3.4
Cs-134	3.48 x E7	2.1
Cs-136	7.38 x E7	2.5
Cs-137	9.50 x E7	3.4
Ba-140	1.33 x E7	0.015
H-3	4.48 x E7	5.5

*Inventory at reactor trip at 0400, 3/28/79 as calculated by the ORIGEN code.

It may first be noted that an appreciable fraction of the total core inventory of I-131 (3.4%) was found in these tanks although only 15 Ci of I-131 were vented from these tanks and penetrated beyond the air vent charcoal filter. From analyses of the thermoluminescent dosimeters in the plant vicinity, about 14 million Ci of Xe-133 escaped through the same air vent. The value of 15 Ci of I-131 in air which was vented from these tanks and which passed through the filters was determined from the analyses of charcoal cartridges which nearly continuously monitored the I-131 content of the downstream air.

The 14 million Ci of Xe-133 which left the auxiliary building vent represents about 10% of the original core inventory of Xe-133 (142 million Ci). The 15 Ci of I-131 in the building effluent was less than one-millionth of the core inventory of I-131 (65.5 million Ci) and less than 0.001% of the I-131 in the auxiliary building tanks. These data confirm the conclusions from the containment air analyses that the noble gas fission products tend to volatilize from reactor coolant whereas iodine strongly tends to remain in solution.

Conclusion

In spite of the gaps and uncertainties in our detailed knowledge of the TMI-2 accident, a fairly reliable answer can be formulated to account for the large differences in the plant effluents of Xe-133 and I-131. The data presented here shows I-131 tends more strongly to remain in the coolant solution than Xe-133. Other processes which remove I-131 from auxiliary building air are plate-out on building surfaces and filtration by charcoal filters. The combined operation of these processes results in the observed large difference in the emissions of Xe-133 and I-131 during the TMI-2 accident.