JUN 5 1968

UNCLASSIFIED

BNWL-SA--668

DE92 003510

FISSION PRODUCT AEROSOL BEHAVIOR IN THE PRTR
FUEL ROD FAILURE OF SEPTEMBER 29, 1965

MASTER

By

R. W. Perkins

C. W. Thomas

W. B. Silker

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Radiological Chemistry
Chemical Research
CHEMISTRY DEPARTMENT

November 22, 1965

Battelle Memorial Institute
Pacific Northwest Laboratory
Richland, Washington

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:
A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or
B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.
As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

UNCLASSIFIED

MASTER

DISTRIBUTION RESTRICTED TO U.S. ONLY
REPRODUCTION OF THIS DOCUMENT IS UNLIMITED

UNCLASSIFIED

BNWL-SA-668

ABSTRACT

On September 29, 1965, at 0510 a purposely defected, partially molten fuel rod in the rupture loop test facility of the PRTR reactor failed in an unexpected manner. The rupture which resulted in a loss of about 39% of the $\text{UO}_2\text{-PuO}_2$ fuel from the rod was accompanied by the formation of a hole of approximately 1/2 inch diameter in the surrounding process tube. The subsequent flashing of the highly contaminated superheated water, released a fission product aerosol to the containment vessel atmosphere. This release resulted in about half of the noble gases reaching the containment vessel atmosphere. About 1 percent of the radioiodine and a somewhat smaller fraction of the solid fission products entered the containment vessel atmosphere. A subsequent radiochemical study provided detailed information on the behavior of some 15 fission products and of plutonium in their movement, following the rod failure, through the various liquid and gaseous systems of the reactor and its containment vessel. The ratios of the fission products (other than the noble gases) which were released to the containment vessel atmosphere were not far different from those in the fuel rod showing that relatively little fractionation occurred in the processes of dissolution and aerosol formation. Only a small fraction of the ^{113}I and other fission products, which were released to the containment vessel atmosphere, were deposited on the containment vessel surfaces. About 99% of the fission product aerosol was removed in the condensate from air coolers which processed about 6 percent of the containment vessel's atmosphere per minute. The ratios of the solid fission products which were deposited on surfaces in the containment vessel were quite similar to those in the fuel rod. Ce^{141} and $\text{Zr}^{95}\text{-Nb}^{95}$ were often exceptions, running several fold lower on the surfaces. The ratio of radioiodine to other fission products was about ten-fold higher on the walls and ceilings than on floor surfaces on the first two days following the incident but after six days the ratios were comparable. The deposition per unit of surface area of the radioactive aerosol was about 4 to 5 times greater on the floors than on wall or ceiling surfaces. The observed deposition per unit area on metal surfaces was much higher than on painted surfaces. Much of the behavior of the radionuclides in this system was similar to what might be expected in the melt-down of any water cooled reactor and the information gained from the study should be applicable to the design of safeguards for such events.

-
- (a) This paper is based on work performed under United States Atomic Energy Commission Contract AT(45-1)-1830. Permission to publish is gratefully acknowledged.

UNCLASSIFIED

1

BNWL-SA-668 ~~CONFIDENTIAL~~

FISSION PRODUCT AEROSOL BEHAVIOR IN THE PRTR FUEL ROD FAILURE OF SEPTEMBER 29, 1965^(a)

R. W. Perkins,^(b) C. W. Thomas,^(b) and W. B. Silker^(b)

INTRODUCTION

On September 29, 1965, at 0510 a purposely defected, partially molten fuel rod in the rupture loop test facility of the PRTR reactor failed in an unexpected manner. The failure which resulted in a loss of about 39% of the fuel from the rod was accompanied by a hole in the surrounding process tube. This hole allowed the superheated loop cooling water to flash, and resulted in a fission product aerosol reaching the containment vessel atmosphere. Automatic ventilation containment was initiated and maintained for about 17 hours to avoid loss of the contaminated vessel atmosphere to the environment. After analysis of the vessel atmosphere it was discharged through a double set of absolute filters and charcoal beds.

The urgent need for defining the amount and behavior of the radioactivity released to the vessel atmosphere and to process waters, together with the research interest in radionuclide behavior in containment systems prompted a very extensive radiochemical study of the incident. The study has provided a good insight into the processes which occur on release of partially molten fuel into its superheated coolant water and the subsequent release of contaminated steam to the containment vessel. Much of the behavior of the radionuclides in this system was similar to what might be expected in the melt-down of any water cooled reactor

(a) This paper is based on work performed under United States Atomic Energy Commission Contract AT(45-1)-1830. Permission to publish is gratefully acknowledged.

(b) Senior Research Scientists, Radiological Chemistry, Chemistry Department, Pacific Northwest Laboratory, operated by Battelle Memorial Institute, for the United States Atomic Energy Commission, Richland, Washington.

and the information gained from this study should be useful in the design of safeguards for such an event.

SUMMARY AND CONCLUSIONS

The behavior has been determined for some fifteen fission produced radionuclides and for plutonium in their escape following the failure of a partially molten fuel rod. The failure of both the fuel rod and its surrounding process tube allowed the superheated process water to flash into the low pressure helium region of the reactor core from which it found its way to the containment vessel atmosphere. The hole in the process tube allowed water to leak from the tube (at about 27 gallons per minute after the initial burst) carrying with it about one-third of the total I^{133} in the fuel element plus large fractions of the other fission products to the disposal tanks. About 2.5% of the plutonium arrived at the disposal tanks with the radioiodine. The leak from the process tube through the helium system resulted in about half of the noble gases from the fuel rod entering the containment vessel atmosphere. In addition, as a result of the flashing of the contaminated process water, about 1.0 percent of the radioiodine and a somewhat smaller percent of the solid fission products was released as an aerosol to the containment vessel atmosphere. The ratios of the fission products (other than the noble gases) which were released to the containment vessel atmosphere were not far different from those in the fuel rod showing that relatively little fractionation occurred in the processes of dissolution and aerosol formation. Approximately 7 curies of I^{131} , in addition to other fission products, were released to the containment vessel atmosphere and only a small fraction of these were deposited on the containment vessel surfaces. About 99% of the fission product aerosol and about 70% of the radioiodine were removed in the condensate from 8 air coolers which circulated air at a rate of about 25,000 cfm

(or ~ 6% of the containment vessel's atmosphere per minute). Had the air coolers not been in operation, essentially all of the radioactive aerosol would presumably have deposited on surfaces in the containment vessel and the general contamination level would have been 100 fold higher. The fact that air coolers served so efficiently in decontaminating the air was surprising and was one of the more significant findings of this study. The condensate from 3 of the 8 air coolers representing about 2/3 of the total air volume was collected in a 50 gallon tank which pumped automatically, when the float valve tripped, to a process sewer line. This resulted in activity bursts which were detected by automatic monitoring equipment in a manhole through which this and other normally clean waste water was discharged to the Columbia River. Although the activity level of the water was well below that which would initiate automatic liquid containment in the reactor vessel, its continued disposal to the Columbia River was undesirable. Therefore, the flow to the river was diverted to a ditch crib until the source of the activity could be determined and isolated. The radioiodine in the containment vessel air several hours after the incident was mainly in the form of organic iodides. Measurements of the airborne radioiodine subsequent to flushing the containment vessel showed 60 to 80% organic and the remainder in the form of inorganic vapor or particulate.

Flushing of the containment vessel air (through particulate and charcoal bed filters) for two days at 10,000 cfm resulted in a 10,000-fold decrease in airborne radioiodine and a 25,000-fold decrease in the radioxenon content of the air.

The total deposition of I^{131} on surfaces in the containment vessel was about 0.05 curies. The deposition of other short-lived fission products ranged up to a few tenths of a curie. The ratios of the solid fission products deposited on surfaces in the containment vessel were quite similar to those in the fuel rod. Ce^{141} and Zr^{95} , Nb^{95} were often exceptions, running several

fold lower on the surface. The ratio of radioiodine to other fission products was about 10-fold higher on the walls and ceilings than on floor surfaces on the first two days following the incident but after six days the ratios were comparable. The deposition per unit of surface area of the radioactive aerosol was about 4 to 5 times greater on the floors than on wall or ceiling surfaces. The observed deposition per unit area on metal surfaces was at least 2 to 6 times higher than on nearby painted surfaces.

Description of the Incident

A detailed description of the incident including all of the reactor operational data will appear elsewhere.⁽¹⁾ An abbreviated description is presented here which includes the information pertinent to understanding the radionuclide release and behavior. The PRTR reactor is a heavy water moderated - heavy water cooled test reactor. The fuel elements, which are either 88 or 58 inches long, are centered in process tubes and are cooled with recirculating heavy water (primary coolant). Surrounding the process tube is a shroud tube which separates it from the heavy water moderator which is contained in the reactor calandria. Reactor gas (helium) flows in the space between the process tube and shroud tube in a low pressure dry gas system which contains rupture discs to allow venting to the containment vessel atmosphere in case of overpressurization. The center process tube of the reactor is used as a fuel element rupture test facility and is cooled with light water supplied through an independent system. This system is designed to operate at a high outlet temperature (500 °F) but its entire stream is cooled after each pass and is routed through an ion exchange system for fission product removal during fuel element rupture tests.

On September 29, 1965, the PRTR reactor was operating with a 4% plutonium (uranium oxide-plutonium oxide) fuel element in its rupture loop test facility. One rod of this standard 19 rod

fuel element had been purposely defected by drilling a 1/16 inch hole through the zirconium cladding to the surface of the fuel. A study was being made with the fuel element to determine what behavior could be expected from operating a defective, partially molten fuel element of this construction. The defective rod, which was an outside member of the cluster, had been exposed in the reactor for about 14 days during a two-month interval. Several hours prior to the incident the outlet temperature of the rupture loop coolant had been reduced from its nominal 500 °F to 300 °F then slowly raised, over several hours, back to 500 °F. Shortly after reaching 500 °F at 0510, the reactor scrammed due to a rupture loop Hi-Lo pressure trip followed immediately by a low flow annunciator. To assure cooling of the entire element, emergency cooling was initiated. At 0511 the reactor went into automatic ventilation containment due to high radiation levels in the exhaust air and all personnel that could be spared were evacuated to the staging area in accordance with emergency procedures. The radiation level in the control room was 35 mR/hr and fresh air masks were worn.

From subsequent observations it was learned that overheating of the process tube at the rupture location produced a small round hole of about 1/2 inch diameter in the process tube. A total of about 700 grams of fuel material or about 39% of the total fuel in the rod was lost.⁽²⁾ Approximately 100 grams of this material were found in the bottom of the shroud tube when it was removed from the reactor for examination. Superheated water flashed through the hole in the process tube into the low pressure helium system and from there escaped through rupture discs to the reactor containment vessel. This process evidently provided a mechanism for release of fuel material to the containment vessel atmosphere. Some of the fuel element material which was lost in the rupture was dissolved or suspended in the

superheated water; and, on flashing of this water into the helium system, an aerosol of fission products and fuel element material was formed. On entering the containment vessel particulate material began to deposit slowly on the vessel surfaces while the noble gases and some of the radioiodine remained in the atmosphere. In addition to deposition and sorption of these materials on surfaces in the containment vessel another important mechanism was operative in their removal from the atmosphere. Eight large air coolers were pulling air through at a combined rate of about 25,000 cfm (or about 6% of the containment vessel's air volume per minute!!). It was estimated that, with the high humidity air resulting from steam release in the containment vessel, about 3 gallons of water per minute was condensed on the cooler coils for a considerable period following the incident.⁽³⁾ Two-thirds of this water, which on condensation had scavenged much of the airborne radioactivity, was fed to a 50 gallon storage tank which pumped automatically to a process sewer when its float valve tripped. The remainder went directly to waste disposal tanks. The process sewer collects other normally clean process waters from several locations in the reactor vessel and drains through a manhole (containing automatic radiation monitors) to the Columbia River. The possibility of a relatively large amount of the airborne radioactive contamination leaving the containment vessel by this route (air cooler condensate) had not been considered likely and several automatic pumpings of the cooler condensate to the process sewer occurred before the source of the high radioactivity in the sewer could be determined.

A 500 ml sample of reactor vessel atmosphere was taken at 0715 through an existing 50 ft long sampling line and had a radiation reading of 500 mR per hr at contact. Most of the radioactivity was due to noble gases with smaller amounts being due to radioiodine and other fission products. The containment

vessel was held in ventilation containment until 2214, at which time it had been determined from containment vessel air analyses that the air could be safely discharged. At this time a pressure of 22.6" of water had built up and the radiation level in the center of the vessel was 4 R per hr. At 2214 the pressure in the vessel was relieved through a bypass valve, which discharged the air through two filtration systems, each consisting of an absolute filter for particle removal plus a charcoal bed for radioiodine removal. When the containment vessel pressure had equalized with atmospheric pressure (at 0034, 9-30-65), the ventilation containment valves were opened and air flow was established at 6700 cfm. This flow was increased to 10,000 cfm at 0210 and by 0330 the radiation level in the center of the vessel had dropped from 4 R to 80 mR per hr.

The leak from the process tube allowed the cooling water from the rupture loop to flow into the helium system at the rate of about 27 gallons per minute, and this water flowed from the helium system into the moderator tank diluting the heavy water with highly contaminated rupture loop water. This water filled the moderator storage tank then backed up to partially fill the calandria before a rupture disc on the system failed and the water level dropped to the weir level of the calandria. This water drained to the basement level of the containment vessel and was pumped from there to storage tanks and subsequently trucked to disposal cribs.

In subsequent sections, which discuss the observed radioactivity levels in the containment vessel, references are made to the reactor hall, the - 11 ft level of C cell, and the - 21 ft level of the C cell. The reactor hall is the main area of the vessel extending from ground level to the top of the dome. The - 11 ft and - 21 ft levels are the first and second floors below the reactor hall. There is no physical barrier barring circulation between levels, and free circulation of the air throughout the vessel took place both before and after the incident.

Radiochemical Analytical Methods

The major portion of analytical radiochemistry was performed by instrumental analysis. By use of multidimensional gamma ray spectrometry⁽⁴⁾ it was possible to study the behavior of a much larger group of radionuclides than would otherwise have been practical. The behavior and the physical and chemical form of radioiodine in the containment vessel atmosphere was determined by use of air sampling packs which separated the radioiodine into particulate, inorganic, and organic forms.⁽⁵⁾ The alpha emitters Pu²³⁹ and Cm²⁴² were measured by direct counting (alpha energy analysis) of the residue from evaporated solutions where possible. However, in many cases chemical separations were necessary for the measurement of Pu²³⁹.

RESULTS AND DISCUSSION

This section is divided into six subdivisions to cover the radionuclide behavior in the process water, in the air, in the condensation from the air as cooler condensate, in deposition from the air on the containment vessel surfaces, in fuel particles remaining in the reactor, and other radioactivity in process water. The behavior of the fission products on release in the reactor system is perhaps best illustrated by comparing the absolute and relative concentrations of radionuclides at different points with that in the fuel element. Table 1 presents the plutonium isotope concentrations and the calculated amounts of the fission products in the fuel rod based on its irradiation history at the time of its failure.⁽⁶⁾ In a separate column these values have been normalized to 1 for Mo⁹⁹. These relative values have been used throughout subsequent portions of this section to compare the observed ratios of radionuclides at various locations with that present in the fuel element. Such a comparison gives a direct measure of the fractionation which occurred during transport of the radionuclides.

Radionuclide Composition in the Rupture Loop and in Leakage from the Loop

Prior to the failure of the test fuel rod, fission products were being released in relatively small amounts from the test hole in the fuel rod's jacket. After the incident, sample collection from the rupture loop was not possible for about 15 hours. Table 2 presents analyses of the rupture loop water collected the day prior to the incident, 15 hours after, and on the second and fourth days following the incident. Shortly after the incident the ion exchange system was valved off and the remaining radionuclide removal from the (500 gallon) rupture loop was by leakage at about 27 gallons per minute through the rupture in the process tube. From Table 2 it is evident that the radioactivity in the loop prior to the incident was small compared with that after the incident. Since the system was at steady state following the incident -- addition to and loss from the recirculating system was 27 gpm -- the erosion rate of material from the fuel rod plus that from fuel particles deposited throughout the loop, could be estimated from the radionuclide concentration of the water. For example, the sample collected 10-1-65 at 1600 showed a Mo^{99} concentration of 120,000 dpm per ml, or 2×10^{-4} curies per gallon. Assuming that this rate had been constant for the two days following the incident, and the data in Table 2 indicate that this is likely, then this would have resulted in the erosion of 8 curies of Mo^{99} per day or 0.3% of that originally present in the test fuel rod per day. The ratios of the radionuclides in the rupture loop water to those in the fuel have been normalized to 1 for Mo^{99} and are presented in Table 3. These values show the relative degree of fractionation which occurred on dissolution or erosion of the fuel element material. They indicate that prior to the incident the rupture loop water was high in I^{131} , Ru^{106} , Ba^{140} , and $\text{Zr}^{95}\text{-Nb}^{95}$ relative to the Mo^{99} but following the incident the I^{131} , Ru^{106} , $\text{Zr}^{95}\text{-Nb}^{95}$, and many others dropped considerably relative to Mo^{99} . The

physical form of these radionuclides was determined by filtering a sample of the rupture loop water through 0.3 μ pore size membrane filter. The percent of the various radionuclides present as particulate material is summarized in Table 4. While the Mo^{99} and radioiodine were almost completely in solution, the other radionuclides were present in varying degrees in particulate form. The physical forms (or solubility) of the radionuclides help explain their behavior both in erosion from the fuel rod and at other locations in the system.

The water leaking from the rupture loop process tube ran into and overflowed the moderator storage tank, and this contaminated water was continuously pumped to "340 tanks" for disposal. Table 5 summarizes measurements of Pu^{239} , Cm^{242} , and radioiodine in waste tanks which were filled with this contaminated water and provides an approximate inventory of the radioactivity arriving at this location. The I^{131} value of 205 curies can be compared with the 773 total in the fuel element. Similarly, the 1404 curies of I^{133} can be compared with the 4350 curies in the fuel element. These values indicate that between 1/4 and 1/3 of the radioiodine in the test fuel rod was released and dissolved (see Table 4) as a result of the rod failure. The Pu^{239} measurements suggest that a much smaller fraction of this material arrived at the disposal tanks. A total of 0.24 curies of alpha activity reached the disposal tanks. This was about half $\text{Pu}^{239-240}$ and half Cm^{242} (see Table 5). This $\text{Pu}^{239-240}$ value of ~ 0.12 curies can be compared with the 5.03 curies of $\text{Pu}^{239-240}$ which was initially present in the fuel rod. This would represent only about 2.5% of the Pu in the failed rod. It is entirely possible that the samples of this waste water were not representative of the particulate radioactivity. Although the storage tanks were stirred continuously during pumping to the tank trucks (when the samples were being taken), the

Pu, which was mainly in particulate form, may not have been sampled proportionately. Also, because of the particulate nature of the Pu, some of it was undoubtedly lost in transit from the rupture loop to the disposal tanks.

In addition to the fuel which was subsequently found in the shroud tube and the rupture loop, particles of fuel were found in several dead legs in the bottom of the reactor calandria and dry gas system, indicating that particles were dropping from suspension as water flowed from the damaged process tube to the waste tanks.

The radionuclide Cm^{242} is produced in the fuel element as a result of a neutron capture by Am^{241} (a daughter of Pu^{241}). Its estimated disintegration rate in the fuel element is comparable to the $\text{Pu}^{239-240}$ concentration and since its concentration in the 340 tanks was also comparable to plutonium, its behavior appears to have been similar to that of Pu. In Table 6 the observed to theoretical ratios of several of the fission products and $\text{Pu}^{239-240}$, which were in the 340 waste tank samples, are compared. These ratios which have been normalized to 1 for Mo^{99} indicate the degree of fractionation that occurred on movement of radionuclides from the fuel rod to the disposal tanks. These data show that radioiodine and Mo^{99} were from about 2 to 20-fold higher than the other fission products. Also the $\text{Pu}^{239-240}$ ratio was very low compared to Mo^{99} and the other radionuclides.

Radionuclide Composition of the Air

From measurements of the radioactivity in the air cooler condensate and of that deposited on surfaces in the containment vessel it is known that an aerosol of fission products and fuel was released to the containment vessel atmosphere; however, only limited remote sampling of the atmosphere was possible during the first 15 hours following the incident. Much of the behavior

of the aerosol must therefore be implied from other observations.

The first air sample was taken two hours after the incident (at 0715) from the reactor hall through a 50 ft sampling line. This was a stainless steel line, and a considerable loss of inorganic iodine and other fission products undoubtedly occurred. The observed concentrations of noble gases and of I^{131} and Ru^{103} in this sample along with their estimated total amounts in the vessel based on their concentrations are summarized in Table 7. A comparison of the total amounts of the noble gases in the vessel with those calculated to be present in the fuel element (see Table 1) indicates that about half of the noble gases from the fuel rod entered the reactor containment vessel atmosphere. The observed to the theoretical ratios of the noble gases (see Table 7) should all be 1 since they are all normalized to 1 for Xe^{135} . The reason for the observed variations from unity are not known but may be due in part to analytical errors in the measurement of some of these relatively uncommon radionuclides. Some of the variation may also be the result of errors in the computed theoretical values which are used as the basis for this comparison. The low apparent concentration of I^{131} and Ru^{103} reflects losses in the sampling line but serves to place a lower limit on the concentrations of the radionuclides in the air at the time of sampling.

Eight and one-half hours after the incident (at 1345), an air sample was pulled from the remote sampling line through a special radioiodine sampling pack. The sampling pack consisted of a filter to remove particulate radioactivity followed by a silver screen to remove inorganic iodine (I_2 and HI) and finally of a charcoal filter and a 2 inch thick by 2 inch diameter charcoal bed to remove organic iodides. The radioiodine, xenons and other fission products which were collected on the various sections of this pack, are summarized in Table 8. The glass filter contained mainly solid fission products and very little

radioiodine. Greater than 99% of the radioiodine was on the charcoal filter and charcoal bed indicating that the radioiodine reaching the sampling pack was essentially all organic. Radioxenon is collected with a poor efficiency by the charcoal bed but the observed radioxenon on the bed can serve as a basis for estimating the xenon content of the air. The xenon values in Table 8 were obtained by assuming a collection efficiency of 20% (and this appears to be maximum possible collection efficiency) for the flow conditions used (see Table 13). Using this factor, xenon concentrations comparable to those listed in Table 7 are obtained (as is shown in Table 13).

The first entry into the containment vessel was made at 1700 and at this time an air sample was collected through an asbestos particle filter followed by a charcoal impregnated paper. One additional sample was taken at 1900 in the same manner, before the containment vessel was vented, and several were taken after its venting. The air concentrations of radioiodines and other airborne fission products in the sample taken at 1900 are summarized in Table 9. Also included are their physical form, their total amounts in the containment vessel, and their observed to theoretical ratios normalized to $\frac{1}{1}$ for Mo^{99} . At this time better than 99% of the radioiodine and some of the ruthenium appeared to be in the gaseous form. The presence of Ba^{140} on the charcoal filter has not been explained but its presence and part or all of the Ru^{106} may have been due to a leak around the particle filter. The observed to theoretical ratios show that the airborne activity (other than noble gases) was greatly fractionated in favor of radioiodine at this point in time. The combined effects of deposition from the air and condensation on the cooler coils had removed most of the solid fission products from the air.

On the second day following the incident some special I^{131} measurements of the air were begun. A 2-inch inside diameter

sampling pack⁽⁵⁾ consisting of a membrane filter followed by a 4-inch long silver mesh bed, a charcoal filter, and a 2-inch long charcoal bed was operated in the containment vessel. The distribution of radioiodine, from the containment vessel air, on such a pack (but with a longer charcoal bed) is shown in Table 10. These data showed that greater than 98% of the radioiodine would be held on the sampling packs in the routine measurements which employed a 2-inch long charcoal bed.

A summary of the radioiodine measurements of the containment vessel air is presented in Table 11. The samples collected at 0715 and 1345 were pulled through a 50 foot stainless tube, and some loss certainly occurred; however, the air concentrations are of the same magnitude as those observed on entry to the containment vessel. These and subsequent measurements suggest that the inorganic iodine released to the vessel atmosphere was depleted, rapidly leaving the less reactive organic iodides in the vessel atmosphere. An estimate of the total radioiodine entering the containment vessel air is made in a later section based on its deposition on surfaces and its removal in air cooler condensate.

Drawing fresh air through the containment vessel at 10,000 cfm (and exhausting this air through particle and charcoal filters) reduced the radioiodine level of the vessel air by about 10,000 in 24 hours (see Table 11). The concentration of radioiodine in the air dropped very slowly, by only 2 or 3 fold, during the next 9 days. Table 12 shows the chemical form -- particulate, inorganic, or organic -- during the eleven day period following the incident. These measurements show that the major portion of the radioiodine remaining in the vessel some 12 hours after the incident was present as organic iodides. The relatively large fractions showing up as particulates in some samples appear to have resulted from the stirring up of dust by workmen. The

samples containing high particulate radioiodine also contained relatively large amounts of other solid fission products.

Table 13 summarizes the measurements of Xe^{133} (5.2 d) in containment vessel atmosphere following the incident. The purging of contaminated air from the vessel reduced the Xe^{133} content by a factor of 2.5×10^4 in the containment vessel air, and it remained at approximately this level for a 9 day period following the incident. The radioxenon measurements were made on samples collected on charcoal beds (along with the radioiodine) at room temperature. The collection efficiencies of these beds for xenon were determined by comparing their radioxenon collection with that of an absolute xenon trap operating next to them.⁽⁷⁾ For flow conditions other than those for which such calibrations were made the collection efficiency was estimated. This was done by comparing the radioxenon thus collected with that collected just before or after under the calibrated flow conditions. The estimated or measured collection efficiencies for all of the radioxenon measurements are included in Table 13. On 10-5-65, the sixth day after the incident, measurements were made to determine the homogeneity of the vessel atmosphere in respect to its radioxenon. Samples taken in the reactor hall, at two locations on the - 11 ft level of C cell and at one location on the - 21 ft level of the C cell showed little variation in the radioxenon levels (see Table 13).

Radionuclide Removal from the Vessel Air in Cooler Condensate

Following the incident, bursts of radioactivity were observed on a periodic basis by monitoring equipment at "manhole 2." This manhole serves as a monitoring point for the process sewer line which drains heat exchanger water and other process waste water from the containment vessel. The only significant source of radioactivity reaching this point was found to be from air cooler condensate from the containment vessel. An evaluation of both the radiochemical analyses of waste water at manhole 2 and of the automatic radiation monitor charts,

which recorded the radiation levels at this manhole, placed the I^{131} loss via this route at about 3 curies. Table 14 summarizes the manhole 2 samples on which detailed radionuclide measurements were made. These samples were taken both during and between bursts. The activity levels between bursts were at least partially due to residual activity from the preceding burst. This is true since the manhole contains a reservoir of about 1000 gallons and flow rates through this manhole were on the order of a few hundred gallons per minute. In the bottom section of Table 14 the ratios of 11 other radionuclides relative to I^{131} are tabulated. The losses of these radionuclides to the river via manhole 2, relative to that of I^{131} , is given directly by these ratios.

In Table 15 the ratios of the observed to the theoretical concentrations of radionuclides from manhole 2 samples are shown, normalized to 1 for Mo^{99} . It is very significant that the ratios of Mo^{99} , Cs^{136} , Ba^{140} , and the radioiodines are not far different in this water than they were in the fuel element. This indicates that the processes which placed them there; namely, release from the fuel, dissolution by the cooling water, aerosol formation by flash boiling of the process water, and condensation along with air moisture on the refrigeration coils of the air cooler did not discriminate to a great extent against any of these. The reason for the lower relative concentrations of Zr^{95} - Nb^{95} , $Ru^{103-106}$, Ce^{141} , and Pu^{239} may be due to a lower release rate from the uranium oxide-plutonium oxide fuel, or perhaps to deposition in the reactor's gas system prior to reaching the containment vessel atmosphere.

The condensate water from 3 air coolers drains to a 50 gallon storage tank which pumps periodically to the process sewer line. The pump on the tank was turned off on 10-1-65, and a sample of the water was taken later (on 10-8-65), after first

stirring the tank, and was used for the radionuclide analysis. Table 16 shows the radionuclide concentrations in the tank, their physical forms, and the total amount of each radionuclide present in the 20 gallons of water which was in the tank at the time of sampling. Also included are the observed to theoretical ratios of the radionuclides. Although these data do not permit a good estimate of the radioactive material which passed through the tank they do indicate its magnitude. The tank had been pumped numerous times (perhaps 15 to 20), but it still contained about 0.1 curie of Mo^{99} and about 0.01 curie of I^{131} . The observed to theoretical ratios of these radionuclides included in Table 16 show that many of these ratios are again close to unity. This provides further evidence that only limited fractionation of the radionuclides relative to their distribution in the fuel rod occurred.

The 3 air coolers each had coarse rubberized fiber filters over their intake and one of these filters was obtained for radionuclide analyses. Table 17 shows the total amounts of the radionuclides present on this filter. Although the filter would be expected to have a very poor collection efficiency, it did collect millicurie amounts of some radionuclides.

Radionuclide Deposition Behavior on Surfaces in the Reactor Containment Vessel

A picture of the deposition behavior of the radioactive aerosol and the radioiodine components was obtained from the analysis of the radioactivity deposited on various surfaces in the containment vessel. Due to high initial radiation levels in the containment vessel, the first samples were obtained simply by wiping surfaces with dry tissue. In subsequent measurements the deposited radioactivity was nearly or completely removed from specific areas for its measurement. This was accomplished by first cleaning with various materials and finally peeling off the paint.

In Table 18, the analysis of two dry tissue smears of a painted floor surface and a stainless steel barrel are tabulated. The smears were taken the day following the incident, and the values shown are the ratios of the observed to theoretical concentrations of the radionuclides normalized to 1 for Mo^{99} . The fact that many of these are near unity (in similar ratios to that in the fuel rod) suggests that the radionuclides have behaved as a group in moving from the fuel to the atmosphere and finally depositing on surfaces. The ratios are similar to those observed in samples from manhole 2 (which contained the radionuclides removed from the air as cooler condensate). The relatively high radioiodine ratios on the barrel (the smear was taken of the sides and top) are believed to be due to preferential adsorption of radioiodine (which was in the gaseous form) on vertical surfaces compared with the uptake of solid fission products. This is substantiated in subsequent observations. On the second day after the incident, painted areas of the floor, wall, and ceiling were cleaned with dry tissue for further study of radionuclide ratios. These data clearly show (see Table 19) fractionation of the radioiodine. Its ratio to the other fission products is much higher on the walls and ceiling than on the floor. On the sixth and seventh days after the incident, various cleaning techniques were employed to determine how well the deposited radioactivity could be removed from surfaces and to measure its deposition per unit area. A standard cleaning method was adopted which employed three consecutive cleaning processes. A measured area of 4 ft^2 was cleaned by rubbing for two minutes with (1) dry tissues, (2) tissues moistened with 0.5 N HNO_3 , and (3) tissues moistened with ethyl alcohol. This was followed in some cases by removal of the paint. Tables 20 and 21 contain the observed radionuclide activity per unit area of surface and the percent of each radionuclide removed by each cleaning process. The areas selected for

these measurements were in the reactor hall and at the -11 ft level in C cell. The measurements show that the activity is not easily nor completely removed by any of the cleaning process; however, greater than 80% of all the radionuclides except I^{131} were removed from the painted surfaces by the three consecutive processes. The activity levels per unit area presented here and in subsequent tables provide information for estimating the total deposition of airborne radioactivity in the containment vessel. The Co^{60} contamination observed in these smears occurred prior to the present incident but is included here and in other tables since it is a part of the radioactive contamination in the vessel.

In Table 22 the observed to theoretical ratios of the radionuclides on the walls from Tables 20 and 21 are compared. These ratios further indicate the relatively small fractionation which had occurred between the fuel element and the deposited aerosol for many fission products.

Table 23 compares the radionuclide activity levels found on adjacent painted areas of the floor, wall, and ceiling at the - 11 ft level of C cell. The activities shown are the total removed in the three consecutive cleaning processes and should therefore represent better than 80% of solid fission products and 60 to 65% of the radioiodine (see Tables 20 and 21). Also included in Table 23 are the relative abundances of each radionuclide on the floor, wall, and ceiling. As indicated in the table the radionuclide activity per unit surface area was about 1/4 as high on the walls as on the floor, and about 1/5 as high on the ceiling as on the floor. The ratios of the various radionuclides on floor, wall, and ceiling are quite similar.

Ratios similar to these were also observed at two locations in the reactor hall (see Tables 25 and 27).

In Table 24 the observed to theoretical radionuclide ratios normalized to 1 for Mo⁹⁹ are tabulated for surfaces listed in Table 23. These data further verify the observed relatively small radionuclide fractionation, for many radionuclides, between the fuel rod and the containment vessel deposition surfaces. Also, the small variation in these ratios for radionuclide deposition on floor, wall, and ceiling is indicated. In contrast to the fission products the relatively low observed to calculated ratio for plutonium indicates that it was not free to move, as were the fission products, through the steps required for the aerosol formation.

Tables 25 through 27 summarize additional measurements of radionuclide deposition on various surfaces, and compare the activity levels and the radionuclide removal by the three consecutive cleaning methods. Tables 25 and 26 present the measurements for adjacent painted areas of floor, wall, ceiling, and of a vertical aluminum surface in the reactor hall and of a floor, wall, and vertical aluminum surface in - 11 ft level of C cell. Table 27 compares just a floor and wall sample from a separate area in the reactor hall. As previously indicated, these tables show that the activity levels on the floor are several times higher for all of the radionuclides than on the walls or ceiling. Also, from the relative amounts of each radionuclide removed by each cleaning process, it is evident that in general the first two cleaning processes removed more than 75% of the total which is removed in the cleaning processes. On comparing the radionuclides removed per unit of surface area from the wall surfaces with that removed from the vertical aluminum surfaces (see Tables 25 and 26) it is evident that a much higher deposition occurs on aluminum metal than on a painted

surface (with this type of paint^(a)). Since the amount remaining on the aluminum after cleaning has not been determined, a precise comparison of total deposition is not possible; however, since the radioisotopes are mainly removed from the painted surfaces by the cleaning processes, a minimum factor for the higher deposition on aluminum can be calculated. Such values have been calculated for the painted wall surfaces and aluminum surfaces in Tables 25 and 26 and are summarized in Table 28. The factors show that two to six times more of most radionuclides were removed from the aluminum than from the painted surface. Thus, the total deposition on these surfaces was at least two to six times greater for the various radionuclides.

The average deposition per unit area of surface can be used to provide an estimate of the airborne radioactivity which deposited. For this purpose an estimated total surface area inside the containment vessel of one-half million square feet and an I^{131} contamination level of $0.1 \mu\text{c}/\text{ft}^2$ was used. This average contamination level was estimated from the measured values in the preceding tables. These values would place the total I^{131} deposition on vessel surfaces at 0.05 curies. This value can be compared with the value of about 7 curies of I^{131} , which was estimated to have been released to the reactor atmosphere based on that condensed on cooling coils 2/3 of which was discharged through manhole No. 2 and that remaining in the containment vessel atmosphere. The amounts of the other radionuclides deposited on surfaces are estimated to be in proportion to I^{131} and similar to that in deposition surface samples discussed in

(a) Phenoline 300 on floors and Phenoline 305 on walls, Carboline Co., 32 Hanley Court, St. Louis, Missouri.

this section. The very significant implication of these observations is that the condensation of steam by the air cooler coils actually reduced the airborne activity and thus surface deposition of all the radionuclides (except the noble gases by about 100-fold). The beneficial effect of the air cleaning is extremely obvious.

Radionuclide Composition of a Fuel Particle

Since all of the samples of both gaseous and liquid origin from the rupture loop were rich in fission products and poor in plutonium relative to the fuel rod, it would be expected that an analysis of the fuel residue would show the opposite. A single particle of the fuel, which was obtained in a dead leg in the rupture loop, was analyzed and did show this expected relationship. Table 29 presents the results of this analysis by comparing the observed to theoretical ratios of the radionuclides normalized to 1 for $\text{Pu}^{239-240}$. As expected the $\text{Pu}^{239-240}$ ratio is high compared to most of the fission products.

Other Radioactivity in Process Water

A source of radioactivity entering manhole No. 2 which appeared significant at the beginning was the light water from the primary D_2O heat exchanger. Significant concentrations of the short-lived fission products were observed in samples of this light water which were available the day following the incident (when the heat exchanger flow was discontinued), and subsequent measurements confirmed their presence. The analyses of two samples taken the day after the incident and a third sample taken 8 days later are included along with ratio calculations in Table 30. Although the short-lived fission product spectrum was similar to that in the manhole samples, the radionuclide concentrations were shown to be several hundred-fold too low to have produced the activity bursts observed in the manhole No.

2 samples.⁽³⁾ The source of contamination of this secondary coolant was evidently backflow and is receiving further study.

ACKNOWLEDGMENT

The authors are indebted to J. H. Reeves, D. R. Edwards, B. G. Christensen, R. M. Campbell, and J. G. Pratt for much of the careful work in sample collection, counting, and calculations. Several other individuals from PRTR, Radiological Analysis and Radiation Protection organizations provided help and information which is gratefully acknowledged.

REFERENCES

1. Investigating Committee Report of the PRTR Incident.
To be published.
2. Anonymous. Nucleonics Washington Daily, November 18, 1965.
3. R. H. Nelson and R. A. Harris, Battelle Northwest Reactor Engineers, Personal communication, November, 1965.
4. R. W. Perkins. An Anticoincidence Shielded-Multidimensional Gamma Ray Spectrometer, Nuclear Instruments and Methods, vol. 33, pp. 71-76. 1965.
5. R. W. Perkins. "Physical and Chemical Form of I^{131} in Fallout," Health Physics, vol. 9, pp. 1113-1122. 1963.
6. J. L. Maryott. "Calculated Fission Product Inventory of the Failed UO_2 - 4.0 PuO_2 Rod."
7. J. D. Ludwick. "Xenon-133 as an Atmospheric Tracer," Presented at the National Meeting of the American Chemical Society, Atlantic City, New Jersey, September 12-16, 1965.

TABLE 1
CALCULATED RADIONUCLIDE CONTENT OF THE FUEL ELEMENT ROD
AND RELATIVE ABUNDANCE FOR THE IRRADIATION HISTORY^(b)

Radionuclide	Half-life, Days	Thousands of Curies/Rod	Relative Abundance
Mo ⁹⁹	2.75	2.76	1.00
Kr ^{85m}	0.183	0.514	0.186
Kr ⁸⁷	0.0542	1.11	0.402
Kr ⁸⁸	0.117	1.65	0.60
Zr ⁹⁵ -Nb ⁹⁵	65	1.48	0.536
Ru ¹⁰³	40	1.06	0.384
Ru ¹⁰⁶	365	0.164	0.0594
I ¹³¹	8.05	0.773	0.280
I ¹³²	0.0958	1.99	0.721
I ¹³³	0.875	4.35	1.58
I ¹³⁴	0.0368	7.67	2.78
I ¹³⁵	0.279	6.61	2.39
Te ¹³² -I ¹³²	3.25	4.16	1.51
Xe ¹³³	5.27	1.33 ^a	0.482
Xe ¹³⁵	0.383	1.83 ^a	0.663
Cs ¹³⁶	13	0.0216	0.00783
Ba ¹⁴⁰	12.8	1.18	0.428
La ¹⁴⁰	1.675	0.954	0.346
		1.04 ^b	0.377
Ce ¹⁴¹	32.5	1.17	0.424
Pu ²³⁹	8.897 x 10 ⁶	0.00503	0.00182
Pu ²⁴⁰	2.469 x 10 ⁶		

^a Daughter of moderate lived parent

^b Curies at maximum build up (after 2.8 days)

TABLE 2

RADIONUCLIDE CONCENTRATIONS IN THE TEST LOOP WATER BEFORE
AND AT VARIOUS TIMES AFTER THE TUBE FAILURE

(dpm per ml)

Radionuclide	Date and Time Sampled			
	9-28-65 @ 1500 ^a	9-29-65 @ 2100 ^b	10-1-65 @ 1600 ^b	10-3-65 @ 1035 ^b
Mo ⁹⁹	590	130,000	120,000	40,000
Zr ⁹⁵ -Nb ⁹⁵	970	8,600	2,200	600
Ru ¹⁰³	290	4,600	--	--
Ru ¹⁰⁶	260	1,900	700	310
I ¹³¹	2,100	11,000	11,000	6,100
Te ¹³² -I ¹³²	350	20,000	5,000	3,800
I ¹³³	--	140,000	91,000	210,000
Ba ¹⁴⁰	1,100	250,000	120,000	70,000
Ce ¹⁴¹	600	1,600	--	--

^a Corrected for decay to sampling time

^b Corrected for decay to time of tube failure (0500, 9-29-65)

TABLE 3

RATIOS OF OBSERVED TO THEORETICAL
ABUNDANCE OF RADIONUCLIDES IN THE TEST
LOOP BEFORE AND AFTER THE INCIDENT

(Ratios Normalized to $\frac{1}{1}$ for Mo^{99})
Observed/Theoretical ^a

Radionuclide	Date and Time Sampled			
	9-28-65 @ 1500	9-29-65 @ 2100	10-1-65 @ 1600	10-3-65 @ 1035
Mo^{99}	1	1	1	1
$\text{Zr}^{95}\text{-Nb}^{95}$	3.1	0.12	0.034	0.028
Ru^{103}	1.3	0.092	--	--
Ru^{106}	7.4	0.25	0.10	0.13
I^{131}	13	0.30	0.33	0.54
$\text{Te}^{132}\text{-I}^{132}$	0.39	0.10	0.028	0.063
I^{133}	--	0.68	0.48	3.3
Ba^{140}	4.4	4.5	2.3	4.1
Ce^{141}	2.4	0.029	--	--

^a Calculated relative abundance in the fuel element based on its irradiation history (see Table 1).

TABLE 4
PER CENT OF THE RADIONUCLIDES IN PARTICULATE FORM IN THE
TEST LOOP COOLING WATER ON 10-3-65

<u>Radionuclide</u>	<u>Per cent Particulate^a</u>
Mo ⁹⁹	~ 0.5
Zr ⁹⁵ -Nb ⁹⁵	59
Ru ¹⁰³	78
Ru ¹⁰⁶	31
I ¹³¹	< 1
I ¹³³	< 1
Te ¹³² -I ¹³²	63
Ba ¹⁴⁰	1.9

^a Per cent of the Radioactivity retained on a 0.3 micron pore size membrane filter.

TABLE 5

INVENTORY OF IODINE, PLUTONIUM AND CURIUM RADIONUCLIDES PUMPED TO THE 340 TANKS FOR DISPOSAL

(Decay Corrected to 0510 on 9-29-65)

Identification		Sampled Date	Volume, (Gal.)	Isotope Concentration			Curies/Tank		uc/Tank	
				I ¹³¹	I ¹³³	μuc/ml Pu ²³⁹⁻²⁴⁰ Cm ²⁴²	I ¹³¹	I ¹³³	Pu ²³⁹⁻²⁴⁰	Cm ²⁴²
46	46	"	1 x 10 ⁴	2.0	14.0	380	78.1	528	14,000	17,000
148	47	"	5 x 10 ³	.23	2.1	-	4.3	30		
99	48	"	1.5 x 10 ⁴	.90	5.1	62	50.0	441	3,500	5,900
58	49	9/30	1 x 10 ⁴	.45	.64		17.0	24		
93	50	"	1 x 10 ⁴	.58	4.2	100	21.9	159	3,800	4,200
100	51	"	5 x 10 ³	.23	5.6		4.3	106		
150	52	"	1 x 10 ⁴	.16	2.2	110	6.1	17	4,200	3,200
98	53	10/1	1.5 x 10 ⁴	.06	.3		3.4	17		
105	54	"	5 x 10 ³	.06	.4	55	1.1	8	1,000	890
106	55	"	5 x 10 ³	.09	.9		1.7	17		
127	56	"	1.5 x 10 ⁴	.09	1.0		5.1	57		
128	57	"	1.5 x 10 ⁴	<						
138	58	10/2	1.5 x 10 ⁴	--						
139	59	"	1.5 x 10 ⁴	<						
146	60	"	1.5 x 10 ⁴	.04			2.3			
154	61	"	1.5 x 10 ⁴	.06			3.4			
157	62	10/3	1.5 x 10 ⁴	.06			3.4			
161	63	"	1.5 x 10 ⁴	.04			2.3			
				Total			205	1404	26,500 ^a	31,200 ^a

^a Total in 5 tanks where both Pu²³⁹⁻²⁴⁰ and Cm²⁴² were measured. The total alpha activity (Pu²³⁹⁻²⁴⁰ plus Cm²⁴²) reaching the disposal tanks was about 0.24 curies.

TABLE 6

RATIOS OF THE OBSERVED TO THEORETICAL^a ABUNDANCE OF RADIONUCLIDES
IN THE WASTE SOLUTIONS PUMPED TO THE 340 TANKS

(Decay Corrected to 0510 on 9-29-65)

(Ratios Normalized to 1 for Mo⁹⁹)

Radionuclide	Date and Time Sampled				
	9-29-65 1220	9-29-65 2005	9-30-65 0420	9-30-65 1230	10-1-65 1300
Mo ⁹⁹	1	1	1	1	1
Zr ⁹⁵ -Nb ⁹⁵	0.19	0.068	0.10	0.24	0.82
Ru ¹⁰³	0.10	0.028	0.069	0.076	0.35
Ru ¹⁰⁶	0.20	0.052	0.17	0.25	0.89
I ¹³¹	2.5	2.1	2.0	1.7	1.1
I ¹³³	3.1	2.0	2.6	~4	1.3
Te ¹³² -I ¹³²	0.26	0.11	0.25	0.12	0.27
Cs ¹³⁶	0.64	0.34	0.72	0.54	0.92
Ba ¹⁴⁰	0.14	0.068	0.13	0.24	0.66
Ce ¹⁴¹	0.084	0.019	0.093	0.11	0.38
Pu ²³⁹⁻²⁴⁰	0.071	0.021	0.052	0.18	0.16

^a Calculated relative abundance in the fuel element based on its irradiation history.

TABLE 7
AIR SAMPLE FROM REACTOR HALL
COLLECTED ABOUT 2 HOURS AFTER THE INCIDENT^a
0715, 9-29-65

(Decay Corrected to 0510, 9-29-65)

<u>Radionuclide</u>	<u>Half-life</u>	<u>Millions of dpm/ft³</u>	<u>Curies in Containment Vessel</u> ^b	<u>Ratio^c Observed/Calculated</u>
Xe ¹³⁵	9.2 H	5840	1050	1.0
Xe ¹³³	5.27 D	1510	272	0.36
Kr ⁸⁸	2.8 H	3640	656	0.69
Kr ⁸⁷	78 M	1690	305	0.47
Kr ^{85m}	4.4 H	1690	305	1.0
I ¹³¹	8.05 D	3.55	0.640	0.0014
Ru ¹⁰³	40 D	0.484	0.087	0.00014

^a Concentrations estimated from gamma-ray spectra by W. Y. Matsumoto. Air from the reactor hall was pulled through a 50 ft. long by 1/4-inch diameter stainless steel line and passed through a 500 ml polyethylene bottle for five minutes. The bottle was then sealed off and used for the above analyses.

^b In determining these values the containment vessel was estimated to have a volume of 4×10^5 ft³ and it was assumed that the radionuclides were uniformly mixed in this volume.

^c Observed to calculated ratios are normalized to 1 for Xe¹³⁵.

TABLE 8

AIR ANALYSIS FROM REACTOR HALL^a

Sampled ~ 1345, 9-29-65
(Decay Corrected to 0510, 9-29-65)
(dpm per ft³ ^a)

<u>Radionuclide</u>	<u>Glass Filter</u>	<u>Silver Screens</u>	<u>Charcoal Filter</u>	<u>Charcoal Bed</u>
La ¹⁴⁰	2,600			
Te ¹³² -I ¹³²	38,000			
Cs ¹³⁶	81			
Mo ⁹⁹	120,000			
Ru ¹⁰³	30,000			
I ¹³¹	~ 3,000	2,300	0.97×10^6	2.4×10^6
I ¹³³	Small	15,000	6.5×10^6	15×10^6 ^b
Xe ¹³³				~ 1500 $\times 10^6$ ^c
Xe ¹³⁵				9000 $\times 10^6$ ^c

^a May-Pac set I-B sample received from R. K. Hilliard.

^b Calculated value based on its observed ratio to I¹³¹ on the charcoal filter.

^c The exact collection efficiency of the charcoal bed for xenon under the collection conditions is unknown but is estimated to be ~ 20%.

TABLE 9

RADIONUCLIDE CONCENTRATIONS, PHYSICAL FORM AND
RELATIVE ABUNDANCE IN THE CONTAINMENT VESSEL ATMOSPHERE
AT 1900 ON 9/29/65. REACTOR HALL

(Decay Corrected to 0510 on 9-29-65)

Radionuclide	Concentrations Thousands of dpm/ft ³	Per cent gaseous ^a	Ratio ^b Observed/Theoretical ^c	Total Activity in Vessel, Curies ^d
Mo ⁹⁹	48	--	1	0.0086
Zr ⁹⁵ -Nb ⁹⁵	0.14	--	0.0054	0.000025
Ru ¹⁰³	6.2	22	0.34	0.0011
Ru ¹⁰⁶	1.8	13	0.63	0.00032
I ¹³¹	5,700	99.7	430	1.0
I ¹³³	27,000	99.7	350	4.9
Te ¹³² -I ¹³²	18	--	0.25	0.0032
Ba ¹⁴⁰	2.7	13	0.13	0.00049

^a Per cent passing through the absolute filter (an H70 asbestos filter), and being collected on the charcoal impregnated filter paper.

^b Ratios normalized to 1 for Mo⁹⁹.

^c Calculated abundance in the fuel element.

^d Assuming an air volume for the containment vessel of 400,000 ft³ and that the radio-nuclides were homogeneous throughout this volume.

TABLE 10

I^{131} DISTRIBUTION IN A SAMPLING PACK FOLLOWING 4.5 HOURS OF OPERATION
C Cell, - 11 ft. Level^a
10-3-65, 0130 to 0600

<u>Sampler Component</u>	<u>Per cent Retained</u>
Membrane filter (0.3 μ)	2
Silver bed (4" long)	13
Charcoal filter	58
Charcoal bed	
0 - 0.6"	15
0.6" - 1.7"	10
1.7" - 2.6"	1
2.6" - 3.4"	~ 0.5
3.4" - 4.3"	~ 0.25

^a Flow rate through the bed was 2 cfm or a linear velocity of 46 cm per second.

AIR CONCENTRATIONS OF THE DIFFERENT FORMS OF I^{131} IN THE
REACTOR BUILDING ATMOSPHERE FOLLOWING THE INCIDENT

(Decay Corrected to 0510, 9-29-65)

Day	Sampled		Sampling Location ^a	Collection		dpm per ft ³			Total
	Time	Date		Time, Min.		Particulate	Inorganic	Organic	
1	0715	9/29	RH						3.55×10^6 c
	1345	9/29	RH	~ 1	~ 3000	2300		3.4×10^6	3.4×10^6 c
	1700	9/29	RH	2					2.2×10^6 c
	1900	9/29	RH	0.25					5.7×10^6 d
2	2030	9/30	RH	5					410
3	1300	10/1	RH	10		67	130	960	1160
	1310	10/1	RH	10		76	33	483	592
	1650	10/1	RH	10		35	112	588	735
	2150	10/1	RH	10		24	80	443	547
4	0100	10/2	RH	10		21	76	386	483
	0300	10/2	RH	10		219	93	397	709
	0907	10/2	RH	10		13	73	364	450
	1300	10/2	RH	240		11	61	255	327
5	0130	10/3	-11	270		14	88	560	662
6	1135	10/4	-11	10		84	154	360	598
7	1353	10/5	-21	10		170	150	490	810
	1400	10/5	-11	10		143	74	351	568
	1410	10/5	-11	10		79	105	321	505
	1422	10/5	RH ^b	10		159	54	1600 ^b	1800
	1435	10/5	-11	490		34	85	286	405
8	2245	10/6	-11	490		46	54	241	341
9	0655	10/7	-11	1600		67	154	395	616
10	0930	10/8	-11	750		25	45	181	251
	2200	10/8	-11	510		13	31	116	160
11	0630	10/9	-11	480		121	38	115	274
	1430	10/9	-11	480		11	29	147	187
	2230	10/9	-11	720		7	22	88	117

^a The symbols RH, -11, and -21 indicate the Reactor Hall, the -11 ft. level of C cell, and the -21 ft. level of C cell, respectively.

^b The top radiation shield covering tube No. 1550 was being removed during the time this sample was collected and may account for the high organic iodide value.

Sample collected through about 50 feet of stainless line. Most of the particulate and inorganic iodine may have been lost in this line.

^d 1.75 ft³ of air was filtered through a charcoal filter. The I^{131} collection efficiency was assumed to be 71% (equal to that collected on the silver bed, and the charcoal filter in Table 10.

TABLE 12

CHEMICAL FORM OF I^{131} IN THE REACTOR HALL
ATMOSPHERE FOLLOWING THE INCIDENT

(Decay Corrected to 0510, 9-29-65)

Day	Sampled		Sampling Location ^a	Collection Time, Min.		Per cent in each Form		
	Time	Date				Particulate	Inorganic	Organic
1	1345	9/29	RH	~	1	~ 0.1	~ 0.1	99.8 ^c
2								
3	1300	10/1	RH		10	6	11	83
	1310	10/1	RH		10	13	6	81
	1650	10/1	RH		10	5	15	80
	2150	10/1	RH		10	4	15	81
4	0100	10/2	RH		10	4	16	80
	0300	10/2	RH		10	31	13	56
	0907	10/2	RH		10	3	16	81
	1300	10/2	RH		240	3	19	78
5	0130	10/3	-11		270	2	13	85
6	1135	10/4	-11		10	14	26	60
7	1353	10/5	-21		10	21	19	60
	1400	10/5	-11		10	25	13	62
	1410	10/5	-11		10	16	21	63
	1422	10/5	RH ^b		10	9	3	88
	1435	10/5	-11		490	8	21	71
8	2245	10/6	-11		490	13	16	71
9	0655	10/7	-11		1585	11	25	64
10	0930	10/8	-11		750	10	18	72
	2200	10/8	-11		510	8	19	73
11	0630	10/9	-11		480	44	14	42
	1430	10/9	-11		480	6	16	78
	2230	10/9	-11		720	6	19	75

^a The symbols RH, -11, and -21 indicate the Reactor Hall, the -11 ft. level of C cell, and the -21 ft. level of C cell, respectively.

^b The top radiation shield covering tube No. 1550 was being removed during the time this sample was collected and may account for the high organic iodide value.

^c This sample was collected through about 50 feet of stainless steel tubing. Most of the particulate and inorganic iodine may have been lost in this line.

XENON-133 CONCENTRATIONS IN THE CONTAINMENT VESSEL
ATMOSPHERE FOLLOWING THE INCIDENT
(Decay Corrected to 0510 on 9-29-65)

Day	Sampled		Sampling Location ^a	Collection Time, Min.	Sample Size ft ³	Xenon Collected per ft ³ of air sampled dpm	Collection Efficiency ^c Per cent	Thousands of dpm ft ³
	Time	Date						
1	0715	9/29	RH					1,510,000
	1345	9/29	RH	~ 1	3/4	3.0×10^8	20 ^d	1,500,000
2								
3	1300	10/1	RH	10	20	1185	1.9	62
	1310	10/1	RH	10	20	1165	1.9	61
	1650	10/1	RH	10	20	1045	1.9	55
	2150	10/1	RH	10	20	643	1.9	34
4	0100	10/2	RH	10	20	579	1.9	30
	0300	10/2	RH	10	20	793	1.9	42
	0907	10/2	RH	10	20	599	1.9	32
	1300	10/2	RH	240	480	41	0.13	32
5	0130	10/3	-11	270	540	52	0.13	40
6	1135	10/4	-11	10	3.16	3870	9.7 ^e	40
7	1353	10/5	-21	10	3.16	5100	10.6 ^e	48
	1400	10/5	-11	10	3.16	4640	10	46
	1410	10/5	-11	10	3.16	6230	10	62
	1422	10/5	RH ^b	10	3.16	7090	10	71
	1435	10/5	-11	490	155	100	0.16	62
8	2245	10/6	-11	490	155	68	0.16	43
9	0655	10/7	-11	1585	501	30	0.048	62
10	0930	10/8	-11	750	237	33	0.11	31
	2200	10/8	-11	510	161	57	0.16	36
11	0630	10/9	-11	480	152	89	0.16	56
	1430	10/9	-11	480	152	143	0.16	90
	2230	10/9	-11	720	228	86	0.11	79

^a The symbols RH, -11, and -21 indicate the Reactor Hall, the -11 ft. level of the C cell, and the -21 ft. level of the C cell.

^b The top radiation shield covering the rupture loop tube was being removed during the time this sample was collected and may account for the relatively high Xe¹³³ value for the reactor hall air.

^c Estimated collection efficiency by comparing xenon collection at different flow rates and collection times with that collected under calibrated conditions.

^d This value may be high. Calibration measurements were not made under comparable conditions.

^e Measured collection efficiency. The xenon collected in the charcoal samplers was compared with that collected in an absolute xenon trap.

TABLE 14

RADIONUCLIDE CONCENTRATIONS IN WATER SAMPLES FROM MANHOLE #2

(Decay Corrected to 0510 on 9-29-65)

Sampled Date	Time	dpm per ml											
		⁹⁹ Mo	²⁴ Na	^{Zr⁹⁵} - ^{Nb⁹⁵}	¹⁰³ Ru	¹⁰⁶ Ru	¹³¹ I	^{Te¹³²} - ^{I¹³²}	¹³³ I	^{Cs¹³⁶}	^{Ba¹⁴⁰}	^{Ce¹⁴¹}	^{Pu²³⁹}
9/29	1110	9,600	120	300	530	94	8,200	3,000	62,000	110	1,500	160	0.7
9/29	1310	70	15	5.3	13	2.2	19	66	130	1.1	39	2.6	
9/29	1605	1,500		40	85	14	820	560	6,400	11	240	25	0.02
9/29	2200	790		14	46	7.7	340	200	2,700	11	93	11	<0.03
9/30	0755	79		8.1	18	2.4	26	100	170	0.65	27	3.8	
9/30	1030	15,000	170	240	300	66	10,000	4,100	84,000	140	1,900	150	<0.2

Ratios of the Various Radionuclides to ¹³¹I

9/29	1100	1.2	0.015	0.037	0.065	0.011	1	0.37	7.6	0.013	0.18	0.020	8.5 x 10 ⁻⁵
9/29	1310	3.7	0.79	0.28	0.68	0.12	1	3.5	6.8	0.058	2.1	0.14	
9/29	1605	1.8		0.049	0.10	0.017	1	0.68	7.8	0.013	0.29	0.030	2.4 x 10 ⁻⁵
9/29	2200	2.3		0.041	0.14	0.023	1	0.59	7.9	0.032	0.27	0.032	<8.8 x 10 ⁻⁵
9/30	0755	3.0		0.31	0.69	0.092	1	3.8	6.5	0.025	1.04	0.15	
9/30	1030	1.5	0.017	0.024	0.030	0.0066	1	0.41	8.4	0.014	0.19	0.015	<2 x 10 ⁻⁵

TABLE 15

RATIOS OF THE OBSERVED TO THEORETICAL ABUNDANCE OF RADIONUCLIDES IN MANHOLE #2 - PRTR
 (Decay Corrected to 0510, 9-29-65)
 (Ratios Normalized to $^1_{99}\text{Mo}$)

Date	Time	$^99_{\text{Mo}}$	Relative Concentration (Observed/Theoretical ^a)									
			$^95_{\text{Zr}} - ^95_{\text{Nb}}$	$^103_{\text{Ru}}$	$^106_{\text{Ru}}$	$^131_{\text{I}}$	$^133_{\text{I}}$	$^{132}_{\text{Tf}} - ^132_{\text{I}}$	$^{136}_{\text{Cs}}$	$^{140}_{\text{Ba}}$	$^{141}_{\text{Ce}}$	$^{239}_{\text{Pu}}$
9/29	1110	1	0.058	0.14	0.16	3.1	4.1	0.21	1.5	0.37	0.039	0.04
9/29	1310	1	0.14	0.48	0.53	0.97	1.2	0.62	2.0	1.3	0.088	
9/29	1605	1	0.050	0.15	0.16	2.0	2.7	0.25	0.94	0.37	0.039	0.009
9/29	2200	1	0.033	0.15	0.16	1.5	2.2	0.17	1.8	0.28	0.033	< 0.02
9/30	0755	1	0.19	0.59	0.51	1.2	1.4	0.84	1.0	0.80	0.11	
9/30	1030	1	0.030	0.052	0.074	2.4	3.5	0.18	1.2	0.30	0.024	< 0.007

^a Calculated abundance in fuel element.

TABLE 16

RADIONUCLIDES IN THE AIR COOLER CONDENSATE (TKA-5)
COLLECTED ON 10-1-65

(Decay Corrected to 0510 on 9-29-65)

Radionuclide	Concentration dpm/ml	Per cent Particulate	Total Present (millicuries) ^a	Ratio Observed/Theoretical ^b
Mo ⁹⁹	1,300,000	57	44	1
⁹⁵ Zr- ⁹⁵ Nb	97,000	99.5	3.3	0.14
Ru ¹⁰³	190,000	99.5	6.5	0.38
Ru ¹⁰⁶	26,000	98	0.89	0.34
I ¹³¹	290,000	79	9.9	0.80
Te ¹³² -I ¹³²	970,000	96	33	0.47
Cs ¹³⁶	7,700	25	0.26	0.76
Ba ¹⁴⁰	180,000	72	6.1	0.32
Ce ¹⁴¹	52,000	98.5	1.8	0.094
Pu ²³⁹	93	~ 100	0.0032	0.039

^a Total present in the 20 gallons of water in the tank at the time of sampling.

^b Calculated relative abundance in fuel element based on its irradiation history.

TABLE 17

RADIONUCLIDES HELD ON A COARSE RUBBERIZED FIBER FILTER
IN FRONT OF A 5000 CFM AIR COOLER IN THE CONTAINMENT VESSEL

(Decay Corrected to 0510 on 9-29-65)

<u>Radionuclide</u>	<u>Total Radionuclide Content</u>	
	<u>Millions of dpm</u>	<u>μ Curies</u>
Co ⁶⁰	36	16
Zr ⁹⁵ -Nb ⁹⁵	200	90
Mo ⁹⁹	3,660	1,600
Ru ¹⁰³	780	350
Ru ¹⁰⁶	110	50
I ¹³¹	4,000	1,800
Te ¹³² -I ¹³²	1,100	500
Cs ¹³⁶	9.5	4.3
Ba ¹⁴⁰	250	110
Ce ¹⁴¹	123	55

TABLE 18

RATIOS OF THE OBSERVED TO CALCULATED RELATIVE ABUNDANCE OF RADIONUCLIDES
REMOVED FROM A FLOOR SURFACE AND A METAL SURFACE IN THE
REACTOR HALL BY CLEANING WITH DRY TISSUE. 2100, 9-30-65

(Decay Corrected to 0510 on 9-29-65)

(Ratios Normalized to 1 for Mo⁹⁹)

<u>Radionuclide</u>	<u>Observed/Calculated^a</u>	
	<u>Floor</u>	<u>Stainless Steel Barrel Top and Side Surfaces</u>
Mo ⁹⁹	1	1
Zr ⁹⁵ -Nb ⁹⁵	0.21	0.29
Ru ¹⁰³	0.44	0.64
Ru ¹⁰⁶	0.43	0.67
I ¹³¹	0.96	10
I ¹³³	1.1	12
Te ¹³² -I ¹³²	0.11	0.34
Cs ¹³⁶	1.4	0.63
Ce ¹⁴¹	0.14	0.21
Ba ¹⁴⁰	0.43	0.35

^a Calculated relative abundance of radionuclides in the test fuel. All measurements decay corrected to 0510, 9-29-65.

TABLE 19

RATIOS OF THE OBSERVED TO CALCULATED CONCENTRATIONS OF
FISSION PRODUCTS ON SMEARS FROM FLOOR, WALL AND CEILING AREAS ^a

(0945, 10-1-65)

(Ratios Normalized to 1 for Mo⁹⁹)

<u>Radionuclide</u>	<u>Observed/Calculated</u> ^b		
	<u>Floor</u> ^c	<u>Walls</u>	<u>Ceiling</u>
Mo ⁹⁹	1	1	1
Zr ⁹⁵ -Nb ⁹⁵	0.13	0.083	0.69
Ru ¹⁰³	0.35	0.41	1.3
Ru ¹⁰⁶	0.36	0.34	1.2
I ¹³¹	1.2	7.6	9.4
I ¹³³	1.5	9.9	10.5
Te ¹³² -I ¹³²	0.18	0.19	0.12
Ba ¹⁴⁰	0.21	0.25	1.1
Ce ¹⁴¹	0.10	0.37	1.0
Pu ²³⁹	0.081	0.10	

^a All surfaces were painted.

^b Calculated relative abundance of radionuclides in the test fuel element; decay corrected to 0510, 9-29-65.

^c Composite of two dry tissue smears.

TABLE 20

RADIONUCLIDES REMOVED FROM A PAINTED WALL SURFACE
BY CONSECUTIVE CLEANING WITH VARIOUS MATERIALS
Reactor Hall, 10-6-65

(Decay Corrected to 9-29-65 at 0510)

Radionuclide	Total dpm/ft ²	Per cent Removed by Cleaning			Per cent Remaining in Paint
		Dry Tissue	Tissue Moistened with 0.5 N HNO ₃	Tissue Moistened with Ethyl Alcohol	
Mo ⁹⁹	620,000	16	36	37	11
Zr ⁹⁵ -Nb ⁹⁵	290,000	19	43	33	5
Ru ¹⁰³	220,000	23	40	30	7
Ru ¹⁰⁶	26,000	23	39	31	7
I ¹³¹	850,000	8	33	24	35
Te ¹³² -I ¹³²	400,000	11	41	33	15
Cs ¹³⁶	7,100	15	47	29	9
Ba ¹⁴⁰	300,000	16	52	26	6
Ce ¹⁴¹	162,000	21	51	22	6
Co ⁶⁰	3,200	32	45	12	11

TABLE 21

RADIONUCLIDES REMOVED FROM A PAINTED WALL SURFACE
BY CONSECUTIVE CLEANING WITH VARIOUS MATERIALS

C Cell, -11 ft. Level, 10-6-65

(Decay Corrected to 9-29-65 at 0510)

<u>Radionuclide</u>	<u>Total dpm per ft²</u>	<u>Per cent Removed by Cleaning</u>			<u>Per cent Remaining in Paint</u>
		<u>Dry Tissue</u>	<u>Tissue Moistened with 0.5 N HNO₃</u>	<u>Tissue Moistened with Ethyl Alcohol</u>	
Mo ⁹⁹	460,000	31	39	17	13
Zr ⁹⁵ -Nb ⁹⁵	36,000	44	15	28	13
Ru ¹⁰³	80,000	44	21	18	17
Ru ¹⁰⁶	9,700	41	24	20	15
I ¹³¹	190,000	21	24	15	40
Te ¹³² -I ¹³²	420,000	39	31	21	9
Cs ¹³⁶	8,800	38	41	12	9
Ba ¹⁴⁰	130,000	28	50	12	10
Ce ¹⁴¹	15,600	48	20	8	24
Co ⁶⁰	1,134	46	31	8	15

TABLE 22

RATIOS OF THE OBSERVED TO CALCULATED ABUNDANCE OF
RADIONUCLIDES ON TWO WALL SURFACES. PAINT COMPLETELY REMOVED^a

Sampled 10-5&6-65
(Ratios Normalized to 1 for Mo⁹⁹)

Radionuclide	Observed/Calculated ^b	
	Reactor Hall, Zero Level	C Cell -11 ft. Level
Mo ⁹⁹	1	1
Zr ⁹⁵ -Nb ⁹⁵	0.87	0.14
Ru ¹⁰³	0.92	0.46
Ru ¹⁰⁶	0.70	0.36
I ¹³¹	4.9	1.5
Te ¹³² -I ¹³²	0.43	0.61
Cs ¹³⁶	1.5	2.4
Ba ¹⁴⁰	1.1	1.2
Ce ¹⁴¹	0.61	0.08

^a The observed ratios were based on the total removed by both washing and complete removal of the paint.

^b Calculated relative abundance of radionuclides in the fuel rod. All measurements decay corrected to 0510, 9-29-65.

TABLE 23

ABSOLUTE AND RELATIVE ABUNDANCE^a OF RADIONUCLIDES ON
ADJACENT PAINTED AREAS OF THE FLOOR, WALL, AND CEILING

C Cell, -11 ft. Level

Sampled 10-5&6-65

(Decay Corrected To 0510 on 9-29-65)

Radionuclide	Abundance, dpm/ft ²			Relative Abundance ^b		
	Floor	Wall	Ceiling	Floor	Wall	Ceiling
Mo ⁹⁹	1,200,000	400,000	260,000	1	0.34	0.22
Zr ⁹⁵ -Nb ⁹⁵	190,000	31,000	73,000	1	0.16	0.38
Ru ¹⁰³	300,000	67,000	80,000	1	0.22	0.27
Ru ¹⁰⁶	44,000	8,200	10,000	1	0.19	0.23
I ¹³¹	440,000	120,000	90,000	1	0.27	0.20
Te ¹³² -I ¹³²	1,600,000	380,000	330,000	1	0.23	0.20
Cs ¹³⁶	33,000	8,000	2,600	1	0.24	0.08
Ba ¹⁴⁰	520,000	110,000	92,000	1	0.22	0.18
Ce ¹⁴¹	220,000	12,000	45,000	1	0.05	0.20
Pu ²³⁹		16				
Co ⁶⁰	6,400	970	790	1	0.15	0.12

^a Based on the total activity removed in the three consecutive cleaning processes:
dry tissue, 0.5N HNO₃, then ethyl alcohol.

^b Normalized to 1 for the floor values.

TABLE 24

RATIOS OF THE OBSERVED TO CALCULATED ABUNDANCE OF
RADIONUCLIDES^a ON ADJACENT PAINTED AREAS OF THE FLOOR,
WALL, AND CEILING. C CELL, -11 Ft. LEVEL

Sampled 10-5 & 6 -.65

(Ratios Normalized to 1 for Mo⁹⁹)

<u>Radionuclide</u>	<u>Floor</u>	<u>Observed/Calculated^b</u>	
		<u>Wall</u>	<u>Ceiling</u>
Mo ⁹⁹	1	1	1
Zr ⁹⁵ -Nb ⁹⁵	0.30	0.14	0.53
Ru ¹⁰³	0.66	0.44	0.81
Ru ¹⁰⁶	0.62	0.35	0.66
I ¹³¹	1.3	1.1	1.2
Te ¹³² -I ¹³²	0.92	0.64	0.84
Cs ¹³⁶	3.6	2.6	1.3
Ba ¹⁴⁰	1.0	0.67	0.83
Ce ¹⁴¹	0.44	0.070	0.41
Pu ²³⁹		0.022	

^a Based on the total activity removed in the three consecutive cleaning processes: dry tissue, 0.5N HNO₃, then ethyl alcohol.

^b Calculated relative abundance of radionuclides in the test fuel element. All measurements decay corrected to 0510, 9-29-65.

TABLE 25

RADIONUCLIDES REMOVED FROM ADJACENT WALL, FLOOR AND BARE ALUMINUM METAL SURFACES BY
VARIOUS CLEANING MATERIALS. REACTOR HALL, MAIN LEVEL

(Thousands dpm per ft²)

Sampled 10-5-65

Radionuclide	Painted Floor			Painted Wall			Aluminum Surface					
	Total Removed	Relative Amounts		Total Removed	Relative Amounts		Total Removed	Relative Amounts				
		A	B		C	A		B	C	A	B	C
Mo ⁹⁹	580	1	0.72	0.26	94	1	1.2	0.77	490	1	0.52	0.61
Zr ⁹⁵ -Nb ⁹⁵	98	1	0.44	0.19	15	1	0.51	0.46	45	1	0.31	0.47
Ru ¹⁰³	150	1	0.51	0.23	20	1	1.1	0.87	130	1	0.38	0.49
Ru ¹⁰⁶	27	1	0.73	0.32	2.7	1	0.83	0.66	32	1	0.51	0.50
I ¹³¹	400	1	0.81	0.69	170	1	1.7	1.1	290	1	0.77	0.86
Te ¹³² -I ¹³²	490	1	0.29	0.12	82	1	0.58	0.58	360	1	0.62	0.93
Cs ¹³⁶	9.2	1	0.55	0.50	1.5	1	1.3	0.78	~ 1.7	1	~ 0.10	~ 2.8
Ba ¹⁴⁰	160	1	0.59	0.23	27	1	1.2	0.98	100	1	0.68	0.94
Ce ¹⁴¹	58	1	0.50	0.24	8.1	1	0.93	0.66	28	1	0.33	0.47
Co ⁶⁰	21	1	1.4	0.35	1.9	1	0.65	0.62	370	1	5.4	1.6

Cleaning Materials:

- A Dry Tissue
- B Tissue moistened with 0.5 N HNO₃
- C Tissue moistened with ethyl alcohol

TABLE 26

RADIONUCLIDES REMOVED FROM ADJACENT SURFACES OF A PAINTED WALL, PAINTED FLOOR AND BARE ALUMINUM METAL BY VARIOUS CLEANING MATERIALS. C CELL - 11 ft. LEVEL

Sampled 10/5&6/65

(Decay Corrected 9-29-65 at 0510)

(Thousands of dpm per ft²)

Radionuclide	Painted Floor			Painted Wall			Painted Ceiling			Aluminum Surface						
	Relative Amounts			Relative Amt.			Relative Amounts			Vertical Position						
	Total Removed	A	B	Total Removed	A	B	Total Removed	A	B	Total Removed	A	B	C			
														C	C	C
Mo ⁹⁹	1,200	1	0.58	0.23	400	1	1.3	0.55	260	1	0.58	0.41	1,200	1	0.62	0.3
Zr ⁹⁵ -Nb ⁹⁵	190	1	0.41	0.20	31	1	0.33	0.62	73	1	0.41	0.39	120	1	0.60	0.4
Ru ¹⁰³	300	1	0.57	0.18	67	1	0.47	0.39	80	1	0.49	0.46	190	1	0.85	0.5
Ru ¹⁰⁶	44	1	0.50	0.16	8.2	1	0.58	0.48	10	1	0.47	0.36	24	1	0.73	0.5
I ¹³¹	440	1	0.84	0.48	120	1	1.1	0.73	90	1	2.0	0.75	330	1	1.3	1.0
Te ¹³² -I ¹³²	1,600	1	0.49	0.12	380	1	0.76	0.51	330	1	2.4	0.50	800	1	0.88	0.5
Cs ¹³⁶	33	1	0.81	0.19	8.0	1	1.1	0.33	2.6	1	0.53	0.20	~ 14	1	~ 0.33	~ 0.3
Ba ¹⁴⁰	520	1	0.68	0.16	110	1	1.8	0.46	92	1	0.59	0.37	320	1	1.4	0.5
Ce ¹⁴¹	220	1	0.16	0.83	12	1	0.41	0.16	45	1	0.40	0.38	68	1	0.61	0.4
Co ⁶⁰	6.4	1	0.49	0.08	0.97	1	0.67	0.17	0.79	1	0.37	0.71	3.4	1	1.4	0.6

Cleaning Materials

A Dry Tissue

B Tissue moistened with 0.5 N HNO₃

C Tissue moistened with ethyl alcohol

TABLE 27

RADIONUCLIDES REMOVED FROM ADJACENT WALL AND FLOOR SURFACES
BY VARIOUS CLEANING METHODS. REACTOR HALL, MAIN LEVEL

(Thousands of dpm per ft²)

Sampled 10-5-65

Radionuclide	Painted Wall				Painted Floor			
	Total Removed	Relative Amounts			Total Removed	Relative Amounts		
		A	B	C		A	B	C
Mo ⁹⁹	770	1	2.1	0.95	200	1	5.0	1.3
Zr ⁹⁵ -Nb ⁹⁵	100	1	1.0	0.69	9.5	1	1.0	0.85
Ru ¹⁰³	180	1	1.5	0.96	18	1	1.1	0.96
Ru ¹⁰⁶	26	1	1.8	1.1	2.2	1	1.5	0.93
I ¹³¹	1,800	1	6.7	1.1	330	1	4.0	2.9
Te ¹³² -I ¹³²	400	1	1.3	1.0	100	1	1.8	1.4
Cs ¹³⁶	6.3	1	1.5	1.1	1.1	1	1.9	0.65
Ba ¹⁴⁰	130	1	1.4	1.4	23	1	1.6	1.0
Ce ¹⁴¹	34	1	1.6	0	6	1	1.3	0.94
Co ⁶⁰	28	1	3.7	1.4	4.8	1	1.9	1.4

Cleaning Materials

A Dry Tissue

B Tissue moistened with 0.5 N HNO₃

C Tissue moistened with ethyl alcohol

TABLE 28

RATIOS OF THE RADIONUCLIDE DEPOSITION ON ALUMINUM SURFACES
TO THAT ON PAINTED SURFACES ^a

<u>Radionuclide</u>	<u>Radionuclide Ratios: Aluminum Surface/Painted Surface</u>	
	<u>Reactor Hall</u>	<u>- 11 ft. Level of C Cell</u>
Mo ⁹⁹	5.2	3.0
Zr ⁹⁵ -Nb ⁹⁵	3.0	3.9
Ru ¹⁰³	6.5	2.8
Ru ¹⁰⁶	12	2.9
I ¹³¹	1.7	2.8
Te ¹³² -I ¹³²	4.4	2.1
Cs ¹³⁶	1.1	1.8
Ba ¹⁴⁰	3.7	2.9
Ce ¹⁴¹	3.4	5.7
Co ⁶⁰	190	3.5

^a Flat vertical surfaces.

TABLE 29

OBSERVED TO THEORETICAL RATIOS OF RADIONUCLIDES IN FUEL PARTICLE WHICH
HAD BEEN RELEASED FROM THE FAILED FUEL ROD TO THE REACTOR MODERATOR SYSTEM

(Decay Corrected to 0510 on 9-29-65)

<u>Radionuclide</u>	<u>Ratio^a: Observed/Theoretical^b</u>
Pu ²³⁹⁻²⁴⁰	1
Zr ⁹⁵ -Nb ⁹⁵	1.6
Ru ¹⁰³	0.33
Ru ¹⁰⁶	0.30
Ce ¹⁴¹	0.73
Ba ¹⁴⁰	0.53

^a Ratios normalized to 1 for Pu²³⁹.

^b Calculated relative abundance in fuel element based on
its irradiation history.

TABLE 30

SUMMARY OF THE RADIONUCLIDE CONCENTRATIONS AND RATIOS
RELATIVE TO Mo⁹⁹ IN THE HX-1 HEAT EXCHANGER
(SECONDARY COOLANT)
(Decay Corrected To 0510 On 9-29-65)

Radionuclide	Concentration dpm/ml			Ratio Observed/Theoretical ^a		
	9-30-65 1115	9-30-65 2030	10-8-65 1355	9-30-65 1115	9-30-65 2030	10-8-65 1355
Mo ⁹⁹	3700	3500	1500	1	1	1
La ¹⁴⁰	49	45	610	0.042	0.042	0.74
Te ¹³² -I ¹³²	210	780	670	0.077	0.30	0.64
Ru ¹⁰³	150	150	130	0.13	0.13	0.27
I ¹³¹	830	530	350	0.85	0.58	0.92
I ¹³³	5900			1		

^a Calculated relative abundance in the fuel element based on its irradiation history.
Ratios normalized to 1 for Mo⁹⁹.

END

**DATE
FILMED**

01/29/92