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BEHAVIOR OF FISSION PRODUCTS RELEASED INTO A STEAM-AIR
ATMOSPHERE FROM OVERHEATED UO₂ PREVIOUSLY
IRRADIATED TO 20,000 MWD/T

(Nuclear Safety Pilot Plant Run No. 14, Part I)

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ABSTRACT

This report summarizes the results of the first run made in the Nuclear Safety Pilot Plant using Zircaloy-2 clad UO₂ irradiated to 20,000 MWD/T and reirradiated shortly before the run to replenish the inventory of short-lived fission products. The run was made by melting the fuel in a plasma under reducing conditions (steam and free hydrogen present in the furnace) with a steam-air atmosphere in the model containment vessel. Only limited melting of the fuel was achieved, and except for a few isotopes, the release was low. An exceptional case was ¹⁴⁰Ba, where approximately 64% release was found. We observed 10% release of ¹³¹I, 6% of ¹³⁷Cs, 2% of ¹²⁵Sb, and less than 1% of other isotopes studied. In general, 1/4 to 1/3% of the amount released reached the containment vessel; however, only 2% of the released Ba reached the containment vessel. In the model containment vessel, 80-90% of the activity apparently deposited during the meltdown phase when a draft tube was being used to mix the containment vessel contents. Removal was by convection rather than sedimentation. A significant part of the remaining material apparently acted as nuclei for a water fog, which then settled. At the end of the run, circulation of the vessel atmosphere through a test filter system for 4 hr reduced the airborne concentration by a factor of approximately 60. Prototypes of remotely actuated samplers for the LOFT experiment were tested successfully.

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INTRODUCTION

The Nuclear Safety Pilot Plant (NSPP) is a facility for studying the behavior of fission products in a pilot plant size (1350 ft³) model of a reactor containment vessel. The facility has been described in a previous report.¹ The principal features are the model containment vessel (MCV), a water-jacketed furnace using a plasma jet heat source to melt clad-UO₂ fuel-pins, a sidestream loop which enables the MCV atmosphere to be circulated through filters and adsorbers to test a simulated air cleaning system, and a variety of remotely operable sampling devices. The equipment is installed in a shielded underground cell which enables experiments using fairly large amounts of radioactivity to be carried out. All sampling devices (except static deposition coupons) are remotely operable, and all samplers located inside the MCV, plus several located outside the MCV, are so mounted that they may be disconnected and removed with long-handled tools without entering the cell.

Prior to the present experiment, 12 runs had been made in the NSPP.^{2,3,4} These comprised two runs with UO₂ alone, five with iodine, two with trace-irradiated UO₂, and three with UO₂ containing mixtures of stable and radioactive isotopes of fission product elements simulating the chemical composition of UO₂ irradiated to high burn-up.

The purpose of the present run was to obtain data on the release, transport, and behavior in a containment vessel of fission products released from UO₂ irradiated to high burnup. By comparing these results with those obtained in the runs using simulated high burn-up fuel, we can evaluate the simulation method.

The principal sampling devices installed in and on the MCV are listed and their locations are given in Table 1. Complete details of the sample installation are given in Part II (Ref. 5) of this report.

Table 1. Principal Sampling Devices Installed in MCV for Run 14

May Pack Clusters - 12 May Packs Each

No. 1	North half, 5 ft above center, radius = 56 in.
No. 2	North half, at center, radius = 30 in.
No. 3	South half, 5 ft above center, radius = 49 in.
No. 4	South half, 5 ft above center, radius = 31 in.
No. 5	South half, 5 ft above center, radius = 15 in.
No. 6	South half, 5 ft below center, radius = 31 in.

LOFT Samplers

Gas-particulate
Plateout

Single May Packs

No. 1	South 5 ft above center, radius = 32 in. (inlet within 3 in. of No. 3 inlet on May Pack Cluster No. 4)
No. 2	South at center, radius = 30 in.

Carousel Fallout Samplers

"B"	At center line, radius = 33 in.
"C"	5 ft below center, radius = 33 in.

Rainout Collector

5 ft below center line, radius = 31 in.

Wall Runoff Collector

SE wall 5 ft below center line, collects runoff in 1 in. wide gutter. Liquid drained into 6 bombs of which first is filled during heat-up and remaining 5 during run.

Deposition Coupons

32 sets of 6 cylindrical coupons located as follows:

4 sets	5 ft above center, at radius 20 in.
4 sets	5 ft above center, at radius 40 in.
4 sets	at center - at radius 20 in.
4 sets	at center - at radius 40 in.
4 sets	at center - at radius 59 in.
4 sets	5 ft below center at radius 20 in.
4 sets	5 ft below center at radius 40 in.
4 sets	5 ft below center at radius 59 in.

There is one set at each compass point in each of the groups indicated.
One hanger containing 48 disc coupons on the NE wall 5 ft below center.
One hanger containing 40 coupons at various angles located at center elevation.
One hanger 5 ft below center.

Table 1 (continued)

Connecting Line from Furnace

One 3-May Pack Cluster at inlet from furnace
 One 3-May Pack Cluster at center of line

Filter Loop

One 6-May Pack Cluster at inlet
 One 6-May Pack Cluster downstream of demister
 One 6-May Pack Cluster downstream of absolute
 filter
 One 6-May Pack Cluster downstream of charcoal
 bed

Purge Line

One 6-May Pack Cluster
 Soil column (for Health Physics)

Observations made in this run included:

1. Collection of air samples
 - a. From two points on the line connecting the furnace and model containment vessel during the period of furnace operation
 - b. From six points in the interior of the containment vessel at eleven different times during the run.
 - c. From four points on the by-pass filter loop during the period of filter operation
 - d. From the vent line as the vessel was purged after the run
2. Exposure of deposition coupons
 - a. 32 sets of 6 hangers in the interior of the MCV
 - b. 40 coupons on the MCV wall
3. Collection of a sample of the condensate running down the MCV wall in 5 time increments
4. Collection of fallout at 2 locations in 11 time increments
5. Test of the effect of a recirculating filter system on MCV atmosphere cleanup.
6. Evaluation tests of sampling devices
 - a. LOFT-Gas particulate sampler
 - b. LOFT Deposition sampler
 - c. Fibrous filter analyzer

The purpose of this report is to collect and disseminate the results obtained in the run. Since a very large amount of information is involved, we have decided to issue two reports covering the run. This report contains a discussion of the results documented with graphs and summary tables. It is being given the standard distribution for ORNL's Nuclear Safety Program reports. The companion report⁵ contains detailed documentation (e.g., tables showing concentrations of all isotopes determined in all of the gas samples). It is being given limited distribution, but copies will be made available on request to workers who may wish to examine the data in more detail.

SUMMARY AND CONCLUSIONS

In this run, the fission product characterization at release was dominated by the effect of the Zircaloy-2 cladding on the fission product chemistry. The Zircaloy acted as a strong reducing agent. The most unexpected effect was a very high release of barium; about 64% of the barium inventory was released from the fuel, but only about 2% of the barium inventory was transported to the MCV.

Because of the relative amounts of the several isotopes under study, this run gave us good data for particles, but only a limited amount of acceptable data for iodine. A little over 10% of the I was released from the fuel and about 75% of this deposited in the furnace.

Most of the activity entering the MCV was collected in the film of condensate on the vessel wall. We estimated 80-90% of the total was collected here. We believe that the deposition in the condensate film occurred while the fuel was being melted, and the draft tube was being operated to mix the MCV atmosphere. The concentrations in the May pack samples of the MCV atmosphere show pronounced maxima, and these maxima occur at different levels depending on the elevation of the May pack cluster in the MCV. We find maxima occurring at 1 1/2 hr 5 ft above the centerline. A possible explanation is that the maxima indicate the formation of a fog cloud on radioactive particles and its settling out. The observed rate corresponds to a particle 3 microns in diameter of unit density.

Our test filter system was operated during the later stages of the run and was effective.

We carried out a reasonably successful field test of prototype LOFT samplers.

Considerable deposition data were collected. As has been the case in all of our steam-air atmosphere runs, we found the effect of material on fission product deposition was relatively small.

The results of this run illustrate the importance of cladding in determining fission product chemistry. For the case of zircaloy-clad cores under reducing conditions it appears that Iodine is released rather slowly from the fuel and that the released iodine is in a form which is readily deposited. On the other hand, the high barium release may be a matter of concern, since it suggests greater release of solid fission products than normally occurs.

We found here, as we did in the simulant runs reported previously,⁴ that both vapor and particulate forms of the released fission products deposited very rapidly in the model containment vessel during the melt-down phase of the experiment. High turbulence resulting from the use of the draft tube to homogenize the MCV atmosphere, plus rapid condensation, apparently cause the rapid deposition. A model proposed by Goldsmith and Stinchcombe⁶ gives a fair fit. This states that for highly turbulent conditions (Reynolds number over 10^4), deposition of particles is a function of condensation rate. If one can posit the required turbulence in a containment building under accident conditions, this model should give a basis for estimating a credit for removal of particles in the course of a loss-of-coolant accident.

DESCRIPTION OF EXPERIMENT

The fuel used in this run consisted of a bundle of eight Zircaloy-2 clad UO_2 pins 5/16 in. diam $\times$ 4 1/2 in. long containing a total of 200 g. of uranium. These had been irradiated in the NRX reactor from February 25, 1960 to April 3, 1961, stored at ORNL and were reirradiated in the ORR from 1415 on February 15, 1966 to 0930 on February 25, 1966 and 1300 on March 15, 1966 to 0300 on March 22, 1966 at an estimated flux of

6.05×10^{10} neutrons-cm⁻² sec⁻¹. Complete details of the fuel history are incorporated in Part II of this report.⁵ Table 2 lists estimated activities of long and short-lived isotopes of interest in the fuel.

Table 2. Estimated Activity
in Fuel Specimen

Activity	Disintegrations per Minute at 0800, 4/1/66
Long-Lived Isotopes	
⁹⁰ Sr	2.87×10^{13}
¹⁰⁶ Ru	1.40×10^{12}
¹²⁵ Sb	3.34×10^{11}
¹³⁴ Cs	4.70×10^{12}
¹³⁷ Cs	2.82×10^{13}
¹⁴⁴ Ce	9.51×10^{12}
Short-Lived Isotopes	
⁹⁵ Zr	1.02×10^{11}
⁹⁹ Mo	7.80×10^{10}
¹⁰³ Ru	8.27×10^{10}
¹³¹ I	1.21×10^{11}
¹³² Te	7.61×10^{10}
¹⁴⁰ Ba	2.46×10^{11}
¹⁴¹ Ce	1.56×10^{11}
²³⁹ Np	3.63×10^{11}

After the equipment was made leak-tight (less than 1% leakage per day indicated by pressure decay and no significant leaks identifiable by probing all mechanical joints with a helium leak detector), the fuel piece was loaded into the furnace, the furnace lid was replaced using remote tools, and leak-tightness of the furnace lid joint was verified by a vacuum leak detector. The cell was then sealed and the high pressure system was brought to 25 psig by injecting open steam into the MCV. Heating required only about 30 minutes, but operating problems forced us to hold the MCV at pressure for about 2 hours prior to torch ignition. This sort of delay permits most of the condensate formed during the heat-up period to drain off the surfaces and to collect in the bottom of the MCV.

At torch ignition time (1225, April 1, 1966), the vessel conditions were 23 psig and 105°C. The torch was operated for 25 minutes, during which time the fuel was passed under the plasma jet once at a speed of 3/4 in./min and a second time at a speed of 1/4 in./min. Only partial melting of the fuel was achieved as is evident from the appearance of the residue shown in Figs. 1 and 2. However, our examination of the residue indicates that the cladding was breached in every pin.

The run was continued for 24 hours, during which time 11 sets of samples of model containment vessel atmosphere were taken on May packs and a variety of other samples were taken. Complete details of sampling are reported in Part II of this report.⁵ Figures 3 and 4 show the variation of temperature and pressure in the model containment vessel with time during this period.

The MCV atmosphere was circulated through a test filter system at a flow rate of 20 cfm (0.89 vessel volumes/hr) during the period 12-16 hr after the start of the run. The test system included a 2-in.-thick style 321 SR York Demister* of Teflon[†] fibers (0.8 mil diam) woven together with stainless steel wires (6 mil diam). Velocities through the Demister are up to 8 ft/sec and the pressure drop is less than 1 in. of H₂O. This was followed by a Flanders style 7C 61R-G circular absolute filter with aluminum separators and an asbestos-fiberglass filter medium containing a binder to provide high wet strength. The last component of the filter train consisted of two Flanders 11-in. square charcoal beds in series packed with Pittsburgh -12 +30 mesh type PCB coconut shell activated carbon. This charcoal was impregnated with potassium iodide at ORNL.

After completing the 24 hr observation period, the MCV was purged with dry air at a flow rate of 30 cfm for 16 hr. To assure maximum mixing of the dilution air with the MCV atmosphere it is introduced as the high pressure motive air in a draft tube mixer. This is calculated to induce a flow of 3000 cfm or over 2 vessel volumes/min. Assuming perfect mixing and no desorption, the reduction factor in airborne concentration during purging is calculated to be 1.6×10^9 .

*Trade-mark - Otto H. York, Co.

†Trade-mark - E. I. DuPont de Nemours and Company, Inc.

NSPP RUN 14—NORTH SIDE

PHOTO 83903

NSPP RUN 14—SOUTH SIDE

The first step in unloading was to open the furnace and take out the boat liner and fuel residue. This was done on Monday, April 4. The MCV was opened on April 5 and the sampling devices were removed on April 5-7. We were fortunate in finding that the radiation level at the top of the MCV was low enough to make the use of a maintenance shield unnecessary; this enabled the unloading to proceed much more rapidly than it would have if the shield had been needed.

Decontamination of the furnace was started as soon as the furnace was closed after removing the fuel. The furnace was decontaminated by circulating 0.5% NaOH at 70°C, followed by 3 water rinses, then by hydrogen-peroxide-inhibited oxalic acid, citric acid, ammonium oxalate (PH = 1.5) at 70°C and three water rinses. The NaOH solution is intended to remove iodine, water-soluble fission products. The oxalate-citrate is a solvent for UO_2 and also appears to be effective for several fission products. This series of decontaminations was completed on April 14.

The MCV was then decontaminated, using the same series of solutions. This work was completed on April 21, and the vessel was opened and surveyed on May 4. The survey showed all horizontal surfaces reading over 20 mr/hr, and we therefore decided more decontamination would be necessary. Based on discussions with several colleagues, we decided to try a commercial automatic dishwashing detergent and also to expose the surfaces to live steam. These were done, and a second survey indicated a significant reduction in contamination, although there still was more than we considered acceptable. Smear samples were analyzed to identify the principal remaining nuclides. Significant amounts of Ru and Ce were found, and further discussion indicated Versene* should be effective. Two batches were applied as well as several water rinses. Another survey indicated that, although the MCV was still contaminated, radiation levels were low enough so that there would be no interference with direct installation of samplers for another run.

Meantime, disassembly of samplers and preparation of samples for the analytical laboratory continued. The samples which were inside the MCV

*Sodium salt of ethylene-diamine-tetracetic acid trade mark, Dow Chemical Company.

during the run were processed first, since we did not want to send our people into the shielded cell to recover the samplers outside the MCV until as much decontamination as possible was done.

Dismantling of the samplers and delivery of samples to the laboratory continued until about June 1. (We could have completed this sooner, but had no incentive to do so since the analytical laboratory could not have processed them any faster because of other commitments.) Over 2100 samples were analyzed.

DISTRIBUTION OF FISSION PRODUCTS

The material balances obtained on the isotopes followed in detail are given in Table 3. The table shows that the amount accounted for ranges from 71.4% for ruthenium-106 to 127% for barium-lanthanum-140.

Table 3. Material Balance in Run 14

Isotope	Percent of Fuel Inventory Found
Strontium-90	89.9
Ruthenium-106	71.4
Antimony-125	112
Iodine-131	76.3
Cesium-137	94.3
Barium-Lanthanum-140	127
Cerium-144	103

On the basis of experience with fission product experiments, these represent excellent behavior. We based the input to the balance on the analysis of a pin which was irradiated simultaneously with those in the fuel specimen, and as shown in Part II (Ref. 5), this gave us far better input data than would have been possible had we calculated the inventories from flux monitor data.

Table 4 presents the distribution of the recovered material. This shows less than 1% release of ^{90}Sr , ^{106}Ru , and ^{144}Ce , moderate release of ^{125}Sb , ^{131}I , and ^{137}Cs and high release of $^{140}\text{Ba-La}$. The latter is

unexpected. The most reasonable explanation seems to be that in the presence of the rather large amount of Zircaloy-2 in the fuel assembly, the Ba was reduced to metal which is much more volatile than BaO. (The fuel pins contained approximately 12.5 g of Zircaloy and 28 g of uranium.)

Table 4. Distribution of Isotopes in Run 14

Location	Isotope						
	^{90}Sr	^{106}Ru	^{125}Sb	^{131}I	^{137}Cs	$^{140}\text{BaLa}$	^{144}Ce
Fuel Residue	99.15	99.64	98.10	89.90	94.33	36.35	99.78
Furnace	0.66	0.34	1.38	7.69	4.93	61.54	0.09
MCV	0.15	0.10	0.46	2.20	0.66	1.87	0.17
Filter System	0.036	0.021	0.066	0.15	0.15	0.32	0.002

BEHAVIOR OF FISSION PRODUCTS IN CONTAINMENT

During the run we collected a good deal of data intended to deal with the question, "What happened to the fission products in the containment vessel?" In this section of the report, we will present our reconstruction of the sequence of events in the MCV and show how this is supported and documented by the various sampling measurements.

The plasma torch was operated during the first 25 min of the run and the flow of gases into the MCV was continued up to 38.5 min. The draft tube in the MCV was supplied with steam for the first 36 min; this induces a large flow of the MCV atmosphere and is intended to mix the MCV contents to give an initially homogeneous system. From velocity studies we have made, we know that the induced flow decays so that velocities become less than the minimum measurable with high-sensitivity instruments (10 ft/min) within 10-15 sec after the motive fluid is shut off. Therefore, we believe the MCV would have been quiescent after 40 min.

There is good evidence that while the draft tube was running, the MCV acted as a wetted-wall scrubbing column. It appears that 80-90% of the fission products entering the MCV were deposited on the surfaces, and that much of this material was deposited on the vertical walls. We base

this conclusion on the following facts: If we divide the activity collected by the rainout and fallout samples by the fraction of the MCV cross-section intercepted, we obtain numbers which are of the order of 10-20% of the MCV inventory determined by the material balance. When we multiply the activity in the wall condensate samples by the ratio of sampler width to vessel perimeter, we get numbers ranging up to 85% (for ^{137}Cs) of the MCV inventory.

A recently declassified report by Goldsmith and Stinchcombe⁶ appears to explain this. In studying various deposition processes for particles, they found that turbulent diffusion with condensation gave a rapid removal and that, although there was much scatter of the data, the deposition velocity was roughly a linear function of the condensation rate, a condensation rate of $10^{-3} \text{ g cm}^{-2} \text{ min}^{-1}$ giving a deposition velocity of 1 cm/sec. Using their correlation and estimating the condensation rate in the MCV at $1.6 \times 10^{-4} \text{ g (cm}^2\text{)(min)}$ the time-temperature curve we predict a deposition velocity of .16 cm/sec in the MCV or a rate constant of $2.7 \times 10^{-3}/\text{sec}$. The observed rate constant is approximately $1.1 \times 10^{-3}/\text{sec}$. In view of differences in geometry between their case (rectangular duct) and ours, the agreement is good.

However, we also conclude that a region existed in the MCV which was essentially unaffected by the draft tube, because of the high concentration of fission products 5 ft above the vessel centerline (see May Pack results below). We suspect partial plugging of the steam supply cut down the flow enough so that less than normal performance was obtained; we found we could not get all of the purge flow we wanted after the run.

Figures 5 through 8 present the data collected by the six May pack cluster samplers located inside the MCV. A May Pack cluster comprises 12 May Packs installed in an air-tight cylindrical housing. A flow measuring and control system, back-up train and vacuum pump are connected to each May Pack in turn by means of a rotary valve actuated by a stepping motor. Spring-loaded ball check valves are provided at the inlets to the May Packs to isolate them when no sample is being drawn. Our policy is to take samples on 11 of the May Packs and to leave the 12th as a blank. We find the blank useful in two ways. It defines the lower sensitivity limit of our analyses, and it gives us a check point on our sequencing of

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the May Pack tubes. Since we have no feedback, this is of great value in verifying suspected errors. In the present run, for example, on one of the units, sample No. 1 was collected on tube No. 2, etc. The location of the blank sample enabled us to verify that this error had occurred, to make the necessary corrections, and thus to utilize the data from this unit.

The figures present data for ^{137}Cs , ^{144}Ce , and ^{90}Sr for all of the units and for ^{131}I and ^{140}Ba on May Pack cluster No. 4 only. The decision to accept I and Ba data from Cluster No. 4 only was based on the following considerations: This was the first cluster analyzed and the only one in which analyses of the absolute filters and check valves for all isotopes of interest were done by conventional radiochemical methods rather than by gamma ray spectrometry. For several reasons, the taping of the gamma spectra went much more slowly than would have been desirable, and the decay corrections for the fairly short-lived isotopes became excessive. When we examined the iodine data, we observed the type of erratic behavior which we have come to recognize as indicating that the decay correction has become excessive. The best judgment we can make is that I and Ba data of acceptable precision were obtained from Cluster No. 4 only.

All of the data have been normalized to a hypothetical initial concentration, C_0 . This is the concentration which would have existed had all of the material found in the MCV been dispersed uniformly in the vessel atmosphere. The C_0 values are given in Table 5.

Figure 5 presents the Cs data. Since it was the dominant gamma emitter in the mix which entered the MCV, we have better data for this isotope than for any other. The data show maxima indicated at 1.5 hr for the samples located 5 ft above the center line, 3 hr for the samples at the center line and 5 hr for the samples located 5 ft below the center line. If we assume that this displacement of the maxima is indicative of the settling of a cloud of material, we can calculate the settling velocity to be 3.3 ft/hr or 3×10^{-2} cm/sec. Green and Lane⁸ give a terminal velocity for rigid spheres of unit density in air of 1.29×10^{-2} cm/sec for 2 micron and 5.00×10^{-2} cm/sec for 4 micron diam. Thus these

Table 5. Values of the Reference
Concentration (C_0) for Isotopes
of Interest

Nuclide	C_0
	Dis/(Min)(Liter)
Strontium-90	1.26×10^6
Ruthenium-106	8.75×10^4
Antimony-125	5.12×10^4
Iodine-131	5.12×10^4
Cesium-137	6.34×10^6
Barium-140	1.78×10^5
Cerium-144	4.13×10^5

particles could be of unit density and approximately 3 micron diam. It seem reasonable to assume that these particles are fog droplets condensed on fission product aerosol nuclei.

After the main peak passes, the concentration vs time curve becomes nearly horizontal, indicating the presence of an aerosol which is settling very slowly. The time interval over which this behavior was observed is too short for us to obtain very precise settling rate data. Based on the Green and Lane data (op cit) we conclude these particles were smaller than 0.5 micron.

The cloud physics literature suggests an explanation of these observations. Mason⁹ shows that the critical radius for growth of a nucleus consisting of water condensed on a soluble solid particle to form a solution is influenced by the particle mass and we can infer that only solid particles larger than some critical size would have served as nuclei for fog formation. The slow-settling residue comprises the particles which were too small to act as condensation nuclei for fog formation. These particles were readily removed by filtration.

Figure 6 presents our data for ^{144}Ce which generally follow the same pattern. Since substantially less Ce than Cs was present in the samples, there is a good deal of scatter here.

Figure 7 presents data for ^{90}Sr , ^{131}I , ^{137}Cs , ^{140}Ba and ^{144}Ce obtained using May Pack cluster No. 4. With the exception of the Ce, there is a

striking parallelism in the behavior of the various materials. It looks as though we are seeing the same aerosol. In other words, that part of the Cs, Sr, Ba and I which was not rapidly removed appears to form a single aerosol. The different behavior of Ce shown here probably is due to bad data, since Fig. 6 indicates that Ce on the other May Packs behaves as Cs, Sr and Ba do on Cluster No. 4.

Figure 8 presents the data we took on ^{90}Sr . This is limited to the first 14 hr of the run because we did not do radio-chemical analyses unless the absolute filters read at least 1 mr/hr. It conforms to the pattern we have observed for other nuclides.

Another observation of interest from these data is that we are unable to identify an effect of radial position in the vessel. Clusters 3, 4, and 5 were positioned on a single radius to check for such an effect. However, the differences we observe in the concentrations found by these three samplers appear random.

In summary: All of the isotopes of interest found in the MCV appear to have been in particulate form. Most of the entering fission products (80-90% of the total) deposited, primarily on the vessel walls, while the plasma torch was in operation. We suggest that effective contacting of the aerosol-laden air with a water film on the MCV wall as a consequence of the draft tube action caused this rapid deposition. At least 80% of the material remaining in the MCV deposited, principally by gravity settling, during a period of several hours. The settling rate suggests a 3 micron particle diameter. The remaining material appeared to be settling much more slowly, but was efficiently removed by a conventional filter train. The concentration-vs-time data suggest a settling rate consistent with the assumption that fission product aerosol particles exceeding some critical size acted as condensation nuclei for fog formation and that the 3 micron particles were fog droplets. The smaller particles remained and formed the aerosol remaining after the fog had settled out.

FALLOUT RESULTS

Data on cumulative fallout are presented in Fig. 9. Two time-dependent fallout samplers were installed, one at the center ("B") elevation

and one at the 5-ft below center ("C") elevation. The C-level sampler collected approximately twice as much as the B-level one. Significant data for all 11 samples were obtained for ^{137}Cs only. The behavior appears to differ at the two elevations; at the lower level there seems to have been a rapidly settling component amounting to at least 70% of the total; this is not seen at "B" level.

The collection media for the two levels were different. At C level we had small Pyrex cups containing water. At B level we used polyvinyl chloride membrane filters. The 18-hr sample at C-level was spilled during unloading, and we have used an estimated value in our calculations.

CONDENSATE RESULTS

Our efforts to obtain an overall understanding of the role that condensate and condensation play in the fission product removal processes included the following:

1. Observation of the MCV temperature. This probably gives us the best estimate we can get of the water vapor content of the MCV atmosphere. We feel that very little error is possible if we assume the atmosphere is saturated.
2. Collection, measuring, sampling and analysis of condensate accumulated in the MCV.
3. Collection of a series of 5 wall runoff samples during the run proper.

In Fig. 10 we show the fraction of the original condensate remaining in the MCV atmosphere indicated by the MCV temperature and also by the wall runoff data. We believe the significance of these data is that most of the condensation contributing to the wall runoff actually occurred during the heat-up period, before the run began. As an additional check on this point, the wall runoff, if uniform around the vessel perimeter, accounts for 61.1% while the water vapor in the MCV atmosphere at the start of the run calculated from pressure and temperature data is only 26.5%. The difference represents a water film on the vessel wall which was present at the start of the run and drained during the run.

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FRACTION OF ORIGINAL H_2O VAPOR IN MCV ATMOSPHERE

We again found, as we have in the past, that most of the fission products tended to concentrate in the early condensate samples so that after 2 hr, when we had collected 70% of the total runoff, we had 85% of the Cs and Sb, 90% of the Sr and Ba ultimately found in the runoff. We think that this is consistent with the other observations indicating that a high percentage of the fission product inventory in the MCV being deposited early in the run while the draft tube was operating. In fact, we can assume that essentially all fission product deposition on the wall occurred while the draft tube was running and that lower concentrations found in the later samples are the result of dilution by condensate formed later and also, probably, of some loss of fission products from the condensate film to the MCV wall.

Ruthenium and cerium behaved differently from the other isotopes followed in the runoff samples, most of these being found in the sample accumulated between 2 and 6 hr. There was also very much less of these than of any others ($\sim 2 \times 10^4$ dpm compared with 10^6 dpm of I and 4×10^8 dpm of Cs). Therefore, we feel that these either plated out very rapidly or never were picked up by the liquid film.

FILTER SYSTEM TEST

One of the piggy-back experiments associated with this run comprised the testing of a typical air-cleaning filter system. The system contained a York style 321,* Teflon[†] Demister (3 1/2-in. diam $\times$ 2 in.-long), a Flanders No. 7C 61R-G (10 5/8-in. diam $\times$ 6 in.) absolute filter with F-700 fiberglass filter medium, and two 11-in. square $\times$ 1 5/16 in. charcoal beds in series, loaded with iodine-impregnated coconut charcoal. The components are scaled for a flow of 30 cfm.

The test filter system is sampled at four points: upstream of the Demister, downstream of the Demister, downstream of the absolute filter, and downstream of the entire assembly. The samples are collected on simplified May Packs containing absolute filters, plain charcoal and iodine-impregnated charcoal. The equipment permits a series of six samples to be collected from each point.

*Trade-mark - Otto H. York, Company.

[†]Trade-mark - E. I. duPont de Nemours and Company, Inc.

Detailed tabulations for all nine gamma-emitting isotopes being followed are given in Part II of the run report. Table 6 of this report summarizes the results we obtained. Only the data for Cs and Ce appear to be meaningful. The activities on samples taken downstream of the absolute filter are of the same magnitude as on the blank samples from the MCV May packs and should be considered as upper limits.

The concentration vs time data from the MCV show that the filters were effective in removing airborne activity. We cannot, however, make a precise estimate of filter efficiency from either these data or from the sample data discussed above. What we can say is that certainly the efficiency exceeds 90% and that we have considerable confidence that it was above 99%. We found no evidence of adverse effects on the filters under the conditions of this experiment.

DEPOSITION STUDIES

We exposed 32 sets of 6 static deposition coupons in the MCV interior, 48 flat coupons on the MCV wall, and 68 flat coupons in the samplers designed to check the effect of orientation. The deposition coupons were designed to investigate the following: (1) effect of location in the MCV, (2) effect of material, and (3) effect of orientation.

The materials investigated are as follows:

Metals: 304L Stainless Steel

A-212 Carbon Steel

Copper

Galvanized Steel

Paints: Amercoat 31 on 304L Stainless Steel

Amercoat 33 HN on 304L Stainless Steel

Amercoat 66 on 304L Stainless Steel

Amercoat 66 on Carbon Steel

Amercoat Dimetecote 4 on Carbon Steel

Carboline B-15 on 304L Stainless Steel

Phenoline 302 on 304L Stainless Steel

Phenoline 302 on Carbon Steel

Devoo and Reynolds SK-1661 on 304L Stainless Steel

Table 6. Run 14 Results from Filter Test Loop Samples
Activity in Sample, Dis/Min

		Sampling Time, Hr. from Start of Run					
	Start	12:08	12:40	13:12	13:44	14:15	14:47
	End	12:38	13:10	13:42	14:14	14:45	15:17
<u>Cesium-137</u>							
Inlet		3.14×10^5	3.02×10^6	1.94×10^6	1.32×10^6	1.32×10^6	2.25×10^4
Downstream of Demister		6.65×10^5	6.79×10^3	5.07×10^5	3.04×10^3	1.18×10^6	2.12×10^3
Downstream of Absolute Filter		1.22×10^3	2.09×10^3	1.92×10^3	1.47×10^3	1.04×10^3	1.29×10^3
Downstream of Charcoal		3.92×10^3	1.86×10^3	6.09×10^3	1.13×10^4	6.69×10^3	1.18×10^3
<u>Cerium-144</u>							
Inlet		1.32×10^4	3.89×10^4	6.37×10^3	2.21×10^4	2.26×10^4	6.33×10^2
Downstream of Demister		8.52×10^3	2.14×10^2	1.54×10^3	0	1.88×10^4	1.17×10^2
Downstream of Absolute Filter		2.36×10^2	2.04×10^2	1.24×10^3	0	0	0
Downstream of Charcoal		3.25×10^2	0	0	6.70×10^2	1.17×10^3	0

Devron 600 on 304L Stainless Steel
 Dupont Epoxy 588
 Dupont Epoxy 589
 Turco Fabrifilm on 304L Stainless Steel

Other: Concrete

Pyrex

Each of the interior hangers in the MCV had 2 stainless coupons plus 4 of some other material. The data from the stainless steel coupons were used to evaluate the effect of position in the vessel and to estimate the precision of the data. The stainless also served as a standard to which other materials could be compared.

The stainless data showed us that there was a possible significant effect of elevation in the vessel and no effect of radius.

The overall means and the standard deviation within the pairs of samples on the same hanger are presented in Table 7.

Table 7. Means and Standard Deviations for
 Activity Collected on Stainless Steel
 Coupons in the MCV Interior

Isotope	Mean, Dis/Min (cm ²)	σ , Dis/Min (cm ²)
Krypton-85	4,080	2,500
Zirconium-Niobium-95	8	15
Ruthenium-106	0	0
Antimony-125	408	290
Tellurium-129	171	870
Iodine-131	692	240
Cesium-137	15,200	4,600
Barium-Lanthanum-140	6	22
Cerium-144	121	270

The table shows rather clearly the limitations of data of this type. We believe it shows that useful data were obtained only I and Cs, and even for these, the precision leaves much to be desired. The reasons for the relatively high standard deviations are that we are dealing primarily with particulate matter, which is probably strongly held on wet surfaces and

poorly held on dry surfaces, and except for the cesium, the samples contain so much cesium that it becomes impossible to obtain good data for other nuclides. We are fairly certain that the isotope reported as "Krypton-85" is something else, although our analytical people cannot identify it. The MCV gas sample data show significant amounts of Krypton-85 only in the low temperature charcoal backup traps, as one would expect. The high plateout on the deposition coupons which was reported to be Krypton-85 cannot be real.

The deposition of I and Cs on the several materials tested relative to that on 304L stainless are presented in Table 8.

Table 8. Deposition of I and Cs on test materials
Relative to that on 304L Stainless

Material	No. of Locations	Relative Deposition	
		^{131}I	^{137}Cs
A-212 carbon steel	4	2.9	2.3
Copper	1	0.1	1.4
Galvanized steel	1	2.1	2.3
Concrete	2	1.6	0.2
Pyrex	1	0	1.3
Amercoat 31 on 304L SS	1	2.2	1.5
Amercoat 33HB on 304L SS	4	1.5	1.3
Amercoat 66 on 304L SS	2	1.1	0.8
Amercoat 66 on carbon steel	2	1.3	1.8
Carboline B-15 on 304L SS	2	1.0	1.1
Phenoline 302 on 304L SS	2	1.9	1.9
Phenoline 302 on carbon steel	2	2.8	1.3
Devco & Raynolds SK-1661	1	1.6	1.0
Devron 600	2	1.4	1.7
Dupont Epoxy No. 588	1	1.2	1.4
Dupont Epoxy No. 589	1	2.4	1.8
Turco Fabriform	1	1.9	2.4
Amercoat Dimetecote 4 on carbon steel	2	2.4	1.7

This table shows that there is little difference among the materials tested with respect to their ability to retain I and Cs under the conditions of this experiment. In view of the limited precision of the data illustrated in Table 5, it is questionable whether we should consider these differences significant.

The wall mounted deposition coupons collected 2-4 times the activity we found on the interior coupons. We again found no significant effect of the material. Specific location in the specimen array did turn out to be important. There appeared to be greater deposition on the lower rows than on the upper, and preferential deposition occurred on the coupons in locations 11 and 12.

The higher deposition on the wall coupons possibly reflects the effect of the forced-convection transfer process during the early part of the run. The position variations are perhaps indicative of condensate movement on the coupon hanger. We do not consider that any conclusions with respect to washdown can be drawn from these data.

Two multipositional coupon hangers, intended to explore the effect of coupon orientation with respect to the horizontal were installed in run 14. The results obtained for Cs are shown in Fig. 11. This shows that settling is a more important transport process than diffusion. We can write

$$y = 6 \times 10^4 + 1.38 \times 10^6 \cos \theta ,$$

where

y = activity on coupon, dis/min,

θ = angle with respect to horizontal.

TESTS OF SAMPLING DEVICES

As a part of this experiment, field tests of the following samplers were made:

One LOFT Plateout Sampler

One LOFT Gas-Particulate Sampler

One Fibrous Filter Analyzer

The two LOFT samplers functioned properly in this run. However, the technician who unloaded the plateout sampler (in a glove box) reported that he had difficulty in handling the coupons during disassembly. We understand the LOFT program has already changed their method of mounting coupons.

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The results from the Plateout sampler are presented in Table 9. The coupons in this case were Amercoat 66 on stainless steel. The table shows reasonably good agreement between the two coupons for each isotope which was present in a measurable amount.

Table 9. Activity Collected in LOFT
Deposition Samplers

	Coupons		Holders	
	No. 1	No. 2	No. 1	No. 2
	<u>Disintegrations per Minute</u>			
⁸⁵ Kr	0	0	0	0
⁹⁵ Zr-Nb	0	0	0	0
¹⁰⁶ Ru	0	0	0	0
¹²⁵ Sh	2.70×10^3	0	0	0
¹²⁹ Te	0	0	0	0
¹³¹ I	1.11×10^4	1.12×10^4	1.09×10^4	9.38×10^3
¹³⁷ Cs	5.41×10^4	1.18×10^4	3.37×10^4	1.10×10^5
¹⁴⁰ Ba-La	0	0	0	0
¹⁴⁴ Ce	0	0	4.26×10^3	2.73×10^3
⁸⁹ Mo	8.82×10^4	6.14×10^4	0	0
¹³² Te-I	9.06×10^4	6.40×10^4	0	0
²³⁹ Np	2.70×10^5	1.83×10^5	1.06×10^5	1.42×10^5

The data for the amount found on the coupon surface compare fairly well with the NSPP static deposition coupons for Cs, but show a possibly significant difference for I (compare tables 6 and 8); the I concentration on the LOFT sampler is higher than on the NSPP static coupons.

Table 10 presents a comparison of the results obtained by the LOFT gas particulate samplers and NSPP May Pack samplers at the same elevation in the MCV. The absolute filters from the LOFT sampler and the absolute filter and check valve parts from May Pack Cluster No. 4 were analyzed by radiochemical methods; all other parts were analyzed only by gamma ray spectrometry. The results show fairly good agreement for ¹³⁷Cs and less satisfactory agreement for practically everything else. We attribute this to the difficulty of obtaining good analyses for other isotopes in the presence of very large amounts of ¹³⁷Cs.

Table 10. Comparison of Activity Collected
by LOFT Gas Particulate Sampler with that
on NSPP May Packs at the Same Elevation

Isotope	Activity Found in Sample (Dis/Min ⁻¹ /Liter ⁻¹)				
	1	May Pack Cluster No. 3	4	5	LOFT Sampler
¹⁰⁶ Ru	1.85×10^2	0	6.14×10^4	9.59×10^4	6.51×10^4
¹²⁵ Sb	3.12×10^5	3.09×10^4	3.33×10^4	5.68×10^4	5.70×10^4
¹³¹ I	*	*	4.07×10^3	*	2.26×10^4
¹³⁷ Cs	1.41×10^7	3.70×10^6	8.34×10^6	4.48×10^6	8.97×10^6
¹⁴⁰ Ba-La	*	*	2.09×10^4	*	1.19×10^5
¹⁴⁴ Ce	3.26×10^5	1.14×10^5	5.54×10^3	1.58×10^5	9.71×10^4

*Results were unacceptable because of too-long decay before analysis.

The fibrous filter analyzer¹⁰ has been under development as a device for measuring aerosol particle size. A fibrous filter analyzer, backed up by a single May Pack, was installed in the MCV as a field test.

The analyzer was installed with six 1 in. diam fibrous filters and six 5/8 in. diam fibrous filters backed up by a standard May Pack. It was operated to take a 10-minute sample at 530 cc/min simultaneously with the third set of MCV May Packs. The results for ¹³⁷Cs are given in Table 11. This table shows that approximately 1/3 of the Cs was collected by the fibrous filter elements, about 2/3 by the absolute filter in the May Pack and almost negligible amounts by other components. The behavior in the fibrous filters was anomalous; the count per filter decreased on the first 6 filters, but increased on the last 6. We believe this indicates transitional flow in the second section of the analyzer. Due to these anomalous results, it was decided no attempt should be made to calculate the particle size, and that flow conditions should be changed for later runs.

Table 11. NSPP Run 14 Fibrous Filter Analyzer
¹³⁷Cs Activity on Component Parts (dis/min)

Stage	Spacer	Inlet Screen	Fibrous Filter Element	Exit Screen	Total
1	9.43×10^4	2.81×10^5	1.62×10^6	1.10×10^5	2.11×10^6
2	7.27×10^4	1.16×10^5	1.51×10^6	8.23×10^4	1.78×10^6
3	2.84×10^4	6.94×10^4	1.10×10^6	5.01×10^4	1.25×10^6
4	1.52×10^4	7.03×10^4	1.18×10^6	6.68×10^4	1.33×10^6
5	1.48×10^4	4.93×10^4	9.64×10^5	3.92×10^4	1.07×10^6
6	1.27×10^4	4.82×10^4	8.81×10^5	5.09×10^3	9.47×10^5
7	1.36×10^4	4.57×10^4	8.99×10^5	4.04×10^4	9.99×10^5
8	1.48×10^4		1.17×10^6	4.43×10^4	1.23×10^6
9	1.37×10^4	3.60×10^4	9.14×10^4	5.16×10^4	1.02×10^6
10	9.23×10^4	5.54×10^4	1.22×10^6	3.60×10^4	1.40×10^6
11	3.10×10^5	1.96×10^4	1.29×10^6	6.45×10^4	1.86×10^6
12	5.65×10^4	4.37×10^4	1.59×10^6	3.27×10^4	1.72×10^6
Subtotal					1.68×10^7
Intermediate Spacers					1.01×10^5
May Pack					
Absolute filter					3.90×10^7
Silver-plated screws					3.13×10^3
Charcoal paper					1.70×10^4
Filter tube					1.48×10^4
Charcoal bed No. 1					4.45×10^3
Charcoal bed No. 2					2.02×10^2
Charcoal bed No. 3					4.82×10^2
Charcoal bed No. 4					1.37×10^3

TRANSFER LINE MAY PACKS

There has been considerable interest in our obtaining samples of the aerosol leaving the furnace and flowing to the MCV. Since we had a lot of trouble in previous steam runs with condensation in the inlet lines and May packs, we rebuilt this part of the sampling system before this run. This new hardware comprises two steam-jacketed May pack clusters, each containing 3 May packs. These are connected to the sample points by 1/8-in.-OD stainless steel tubes, approximately 4 ft long. (This was the shortest run we could use, based on the necessity to provide for remote removal of the sampler and inlet line.) These lines are steam-jacketed also. All three May packs are connected to a manifold at the end of the inlet tube; there are no shutoff valves or check valves on the inlet side. On the exit side of the May packs there is a solenoid valve associated with each. These can be operated from the control room to select the May pack to be used at a given moment. Downstream of the solenoid valves, the lines are manifolded together and tied into a regular NSPP back-up, flow-metering and vacuum system.

One of these samplers picks up a sample at the inlet to the line connecting the furnace to the MCV, and the other gets its sample from the line about midway between the furnace and MCV.

After the run, the inlet lines were removed and sectioned, and analyses were made of the sections. The May packs had collected so much activity that it was considered necessary to remove the absolute filters in a hot cell. (Before the filters were taken out, the radiation level at the end of the May packs was about 10 r/hr.)

The detailed data collected by these samplers are presented in Part II of this report.⁵ We were able to obtain good data for the long-lived particulate activities. We have plotted up the inlet tube data, but these appear too scattered to justify any effort to interpret them as diffusion tube data.

In the May packs, the activity evolved during the first pass of the fuel through the plasma jet was collected on tube 1 and that from the second pass on tube 2. Tube 3 was not used. In both clusters we had some trouble with activity penetrating past the absolute filter. About

36% of the particulate activity was collected downstream of the absolute filter in tube 1 of the furnace and cluster, and about 90% was collected downstream of the absolute filter in tube 2 of the cluster sampling the line center. All of these May packs had been given a DOP test and had demonstrated efficiencies of 99.997 to 99.999% for removing the 0.3 μ DOP aerosol before the run. On disassembly these filters were found to have been torn by the combined action of moisture and differential pressure. Our backup trap data indicate that all of this activity was retained in the May packs.

Table 12 presents a comparison of activity values predicted from the transfer line samples and those found by inventory of the MCV. We used uncertainties in the flow data. The table shows we got good predictions for ^{90}Sr and ^{144}Ce , but not for the other isotopes on which we have comparative data. These results are consistent with our assessment of the analytical difficulties encountered in this run.

Table 12. Comparison of Observed MCV Inventory with that Predicted from Samples of Entering Gas

Isotope	Predicted from Furnace End Sampler Data	Predicted from Line Center Sampler Data	Found by Material Balance
$^{137}\text{Cs}^*$	1.95×10^{11}	1.95×10^{11}	1.95×10^{11}
^{90}Sr	3.12×10^{10}	2.65×10^{10}	2.60×10^{10}
$^{95}\text{Zr-Nb}$	5.43×10^8	2.53×10^8	**
^{106}Ru	9.65×10^8	4.55×10^8	2.22×10^9
^{125}Sb	4.90×10^8	2.06×10^8	1.31×10^9
^{129}Te	1.80×10^{10}	9.68×10^{10}	**
^{131}I	1.70×10^{10}	2.96×10^9	9.57×10^8
$^{140}\text{Ba-La}$	6.23×10^8	1.51×10^8	2.24×10^9
^{144}Ce	5.34×10^9	5.42×10^9	1.01×10^{10}

*Reference.

**Not determined.

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