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FISSION PRODUCT BEHAVIOR UNDER SIMULATED
LOSS-OF-COOLANT ACCIDENT CONDITIONS IN
THE CONTAMINATION-DECONTAMINATION EXPERIMENT

IDAHO NUCLEAR CORPORATION
NATIONAL REACTOR TESTING STATION
IDAHO FALLS, IDAHO

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ABSTRACT

A pilot-plant-scale facility, the Contamination-Decontamination Experiment, was built and operated to produce mixtures of high-level, short-cooled (6- to 10-hr) fission products under conditions simulating an unperturbed loss-of-coolant reactor accident. The mixtures were created by melting irradiated, zirconium-clad UO_2 fuel pins in steam. The fission products released from the fuel were transported by steam to an 86 ft³ vessel containing a saturated air-steam atmosphere. Extensive data on fission product behavior were obtained during five runs in the facility and are reported. The measured release fractions of fission products from the grossly melted fuel were comparable to the release fractions suggested in TID-14844, except for those of tellurium, cesium, and molybdenum, which were a factor of approximately 10 higher. Of the fission products released from the fuel, except the noble gases, most of the cesium and zirconium, little of the molybdenum and ruthenium, and 1/10 to 1/3 of the other fission products were deposited in the transport line. In the containment vessel, where the natural behavior of the fission products was observed as the steam-air atmosphere was allowed to cool, the concentrations of noble gases and organic iodides remained essentially constant. The radioiodine and particulate activities in the vessel atmosphere dropped rapidly (half time of approximately 8 min) for 30 min after injection of fission products into the vessel. The depletion rate of the radioiodine activity then slowed considerably (to a half time of approximately 7 hr). After a 24-hr cooling period, approximately 3% of the radioiodine and approximately 0.2% of the particulate activities injected into the vessel remained in the atmosphere. Tin, volatilized from the cladding, made up most of the mass of particulate matter released to the containment vessel. About one-half the activities removed from the atmosphere was found in the condensate; the other one-half deposited on the vessel surfaces. Condensation was observed to enhance removal of fission products from the atmosphere; cold surfaces in the vessel received more deposition than warm surfaces, and increasing the steam injected into the atmosphere increased the removal of fission products with the condensate. "Maypack" type samplers indicated that 1 to 2% of the radioiodine inventory in the fuel was present in the containment vessel as organic iodides. Subsequent laboratory and tracer runs show that much of this may instead be another iodine species, hypoiodous acid.

SUMMARY

The Contamination-Decontamination Experiment (CDE) is a pilot-plant facility for producing and sampling mixtures of high-level, 6- to 10-hr cooled fission products under conditions simulating an unperturbed loss-of-coolant accident for a water-cooled reactor. The fission product mixtures -- produced by induction melting of irradiated, zirconium-clad UO_2 fuel pins -- are transported by air-free steam to an 86-ft³ stainless steel containment vessel in which the environmental conditions of a loss-of-coolant accident are simulated.

Successful completion of five high-level runs and numerous trace-level and nonradioactive runs has demonstrated the safe operability of the CDE. Although the CDE is primarily a test facility to evaluate decontamination problems and to test samplers and analysis methods for the Loss-of-Fluid Test (LOFT), considerable information on fission product behavior was obtained during its operation.

The behavior of fission products in the integrated-accident-sequence, high-level CDE runs generally confirms the pattern expected from the results of low-level, separate-effects experiments conducted elsewhere. The fractional releases of fission products decreased in the order: krypton and xenon, iodine, tellurium, cesium, molybdenum, ruthenium, barium, strontium, cerium, and zirconium -- as expected. Approximately 100% of the xenon and krypton, 50% of the iodine, 10% of the tellurium, cesium, and molybdenum, and less than 1% of the less volatile fission products present in the fuel were released in the CDE runs, in which the melting of the UO_2 represents the most severe accident conditions for a reactor. Of the fission products released from the fuel (excluding xenon and krypton), most of the cerium and zirconium, little of the molybdenum and ruthenium, and 1/3 to 1/10 of the other fission products deposited in the transport line, which was maintained at approximately 800°F.

The distribution of the fission products between the condensate and wall in the containment vessel depended on the amount of condensate formed (or steam injected) and the solubilities of the fission products in the condensate. Generally, about one-half of the total fission products (other than noble gases) reaching the containment vessel was removed in the condensate, and the other half was deposited on containment vessel surfaces. However, increasing the total amount of condensate increased the fraction of fission products removed in the condensate, and the soluble fission products appeared preferentially in the condensate. The particulates in the containment atmosphere were small (0.001 to 0.7 μ), as would be expected from a volatilization-condensation mechanism, and showed evidence of agglomeration, which is usual for a particulate release into steam. Fission products other than krypton, xenon, and iodine behaved similarly in the containment vessel, indicating that they were mainly present as particulates. The concentrations of xenon and krypton remained essentially constant in the atmosphere, but the airborne iodine concentration decreased with an initial half time of approximately 8 min.

The deposition of fission products on surfaces depends on surface orientation, environmental conditions, and material. Because of particulate fallout, surfaces facing up collected more deposited activity than surfaces in other

orientations. Iodine and particulate deposition on the materials tested appeared to increase in the following order: glass, plastics, metals, and paints. Deposition of iodine on painted surfaces increased markedly at temperatures above the softening point of the paint.

Results first noted in the CDE runs were (a) tin, a volatile component of Zircaloy, constituted the major mass of the particulates, (b) condensation promoted fission product movement toward the wall, resulting in a higher deposition on condensing surfaces than on noncondensing surfaces, and (c) a large fraction of the iodine activity remaining in the atmosphere after environmental conditions had stabilized appeared to be hypoiodous acid (HOI), rather than organic iodides.

Experience in the CDE has revealed a number of sampling and analysis difficulties that may be encountered in the LOFT and has shown that the samplers used in the LOFT must be proof-tested under accident conditions to assure satisfactory results. Also, care must be exercised in interpreting sample analysis results because of problems such as settling or plateout of fission products in condensate samples, loss of volatile iodine species from samples, and adsorption of iodine by sample bottle seals.

Certain improvements in the CDE would increase the amount of "spinoff" information obtained on fission product behavior in future runs. Specifically, measurement of the fuel temperature during the runs, modification of the containment atmosphere samplers to provide positive discrimination between the fission product (especially iodine) species, increased frequency of atmosphere sampling during the early washout period, and correction of sampling and analysis problems would furnish a more quantitative understanding of the accident consequences.

Improvement in the overall water-reactor safety program on fission product behavior is needed. Closer coordination of the numerous experiments being conducted on fission product behavior and centralization of the efforts to combine the various separate-effects results into one overall model would increase the value and timeliness of the information being obtained. In addition, further work is recommended in the following areas:

- (1) Release of fission products as a function of temperature and anticipated accident atmosphere composition over the fuel, particularly at high burnups
- (2) Extent of fission product deposition in the reactor internals, both with and without emergency core cooling systems
- (3) Positive identification of all significant fission product species and their formation and disassociation mechanisms
- (4) Development of methods for scale-up of data from pilot to commercial units.

Increased emphasis in these areas would reduce uncertainties in describing an unperturbed loss-of-coolant accident in a water-cooled reactor.

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I. INTRODUCTION

The Idaho Nuclear Corporation (INC) is conducting several experimental studies in support of the Loss-of-Fluid Test (LOFT)[1,2]. This test is part of the USAEC's reactor safety program and is being conducted by the Atomic Energy Division of Phillips Petroleum Company at the National Reactor Testing Station (NRTS). The LOFT is an engineering-scale test demonstrating the consequences of accidental loss-of-coolant (LOC) in a water-cooled reactor. In its original concept, the LOFT was to simulate a major LOC accident in which none of the engineered safeguards, except the containment building, were to be used to mitigate the severity of the fission product release. In mid-1967, however, the LOFT program was redirected toward the testing of core cooling and other engineered safeguard systems[4]. This document summarizes the information obtained from the INC support studies conducted under conditions simulating those anticipated in the original LOFT concept, ie, extensive cladding failure, followed by gross melting of the fuel and a major release of fission products uncontrolled by the use of engineered safeguard systems.

In the originally planned LOFT, the reactor core coolant (at a pressure of approximately 2500 psig) would be released deliberately by opening the primary coolant loop of an operating 50 MW(t) pressurized water cooled reactor. After the coolant loop was opened, the core coolant water would be rapidly expelled into the containment building, allowing the reactor core to heat from the fission product decay energy. As the core temperature increased, the fuel pins would fail through buildup of internal pressure, oxidation of the zirconium cladding, fracturing of the cladding, and gross melting of the fuel. Gaseous fission products collected in the plenum of the fuel pins during irradiation would be partially released immediately upon cladding failure. Further release of fission products from the fuel-cladding gap and from the bulk of the fuel would occur as the fuel temperature continued to increase. Eventually, some of the fuel pins in the hottest section of the core would crumble or slump and fall to the bottom of the reactor vessel.

After release from the fuel, the fission products would be carried through the reactor interior and into the containment building by steam vaporized from residual water in the bottom of the reactor vessel. Of the fission products carried from the hot fuel zone, a portion would deposit on the interior surfaces of the reactor and primary coolant piping. The fission products remaining in the gas stream would be released to the containment building atmosphere, which is composed of a saturated steam-air mixture at approximately 235°F and 24 psig. Since no engineered safeguard systems (except the containment building) were to be used in the originally planned LOFT, the fission products would be removed from the containment atmosphere by the natural processes of deposition on surfaces, washout by condensing steam, and settling of the particulates.

In 1964, several support studies for the LOFT were assigned to the Nuclear and Chemical Technology Division of Phillips Petroleum Company. Subsequently, when a change in NRTS operating contractors was instituted in 1966, this Division became part of the Idaho Nuclear Corporation. These LOFT support studies included (a) the evaluation of surface coatings and decontamination methods for the interior of the LOFT containment building, (b) testing of the LOFT fission product sampling equipment, and (c) the development of analysis methods for the radioactive samples generated in the LOFT.

The Contamination-Decontamination Experiment (CDE) was initiated as a testing system for the LOFT support studies to produce mixtures of high-level fission products under conditions simulating the LOFT conditions. The fission product mixture is generated by melting an irradiated fuel pin in the presence of steam, using a radio frequency induction furnace. The fission products released from the fuel are transported by steam to a stainless steel containment vessel where the environmental conditions are controlled to match those anticipated in LOFT. Numerous samplers used in the support studies are located in the containment vessel. The average CDE run conditions and those anticipated for the originally scheduled LOFT are compared in Table I.

Installation of the CDE in the Hot Pilot Plant (HPP) at the Idaho Chemical Processing Plant (ICPP) was completed in December, 1965. Five runs have been conducted in the CDE to date, using fuel pins cooled 6- to 10-hr and containing 7,000 to 35,000 curies of fission products per run. Numerous nonradioactive meltdowns and four tracer-level runs using xenon and iodine have also been made. The Reactor Safety Studies Section of the Chemical Technology Branch is responsible for the installation and operation of the CDE.

This document describes the CDE and summarizes the information obtained on fission product behavior during the experiments. Since the primary objective of the CDE is to provide fission product mixtures for the specific LOFT support studies, no intensive effort was undertaken for studying the behavior of fission products under LOC accident conditions. Rather, the information on fission product behavior was gained as "spinoff" from the specific support studies. The information on fission product behavior obtained in these experiments has been summarized in this report because the CDE represents one of the few experiments in the AEC's safety program in which irradiated, zirconium-clad UO_2 fuel pins have been melted under LOC accident conditions. Results of the specific LOFT support studies in which the CDE was utilized are documented in other reports[5-9].

TABLE I
COMPARISON OF CDE AND LOFT OPERATING VARIABLES

Operating Variable		CDE ^[a]	LOFT ^[b]
1.	Fuel Heating Method	Induction Generator	Fission Product Decay Heat
2.	Containment Volume, ft ³	86	302,000
3.	Containment Surface Area, ft ²	115	23,600
4.	Containment Material	SS/Paint	SS/Paint
5.	Containment Volume/Surface, ft	0.75	12.8
6.	Containment Atmosphere	Steam-Air	Steam-Air
7.	Maximum Containment Temperature, °F	235	217 to 235 ^[c]
8.	Maximum Containment Pressure, psig	22	18.7 to 26.1 ^[c]
9.	Fuel Type	UO ₂	UO ₂
10.	Fuel Cladding	Zircaloy-2	Zircaloy-4
11.	Fuel Enrichment, %	8	2.8 to 5.7
12.	Fuel Burnup, MWd/tonne	700	940
13.	Fuel Cooling Time at Meltdown, min	600	5 to 60
14.	Fuel Inventory at Meltdown, <u>Curies in Fuel</u> Cu. ft of Containment Volume		
	Kr-85	0.065	1.75
	Xe-133	1.16	9.87
	I-131	0.474	4.11
	Cs-137	0.002	0.019
	Te-132	0.713	6.83
	Sr-89	0.283	2.56
	Total Fission Products	93	394

[a] Approximate values for average CDE test.

[b] Approximate values anticipated for the original LOFT.

[c] Lower temperature and pressure for primary loop break only; higher temperature and pressure for combined primary and secondary loop break.

II. DESCRIPTION OF FACILITY AND EQUIPMENT

The primary CDE systems and equipment are located in Cell 2 of the ICPP Hot Pilot Plant, a complex of five hot cells adjacent to the fuel processing building. The general orientation of Cell 2 in the HPP is shown in Figure 1. Fuel pins, irradiated in the MTR and transferred to the ICPP in a cask, are lowered onto the dolly and then moved into the hot cell. The meltdown and all

Fig. 1 General orientation of CDE facility.

other operations in Cell 2 are accomplished remotely from the operating corridor located to the north of Cell 2. The cell immediately to the south is utilized for sampling operations and sample handling, which can be performed manually but require ventilation. A sodium iodide (NaI) scintillation crystal is located in a cave in the south corridor, and is used for counting those samples which are too radioactive for handling in the nearby analytical facilities. Cell 5, to the east of Cell 2, is used for storage of radioactive samples.

Cell 2 is 10- by 22- by 20-ft high and is painted with acid-resistant paint, except for the floor and lower 5 ft of the walls which are lined with stainless steel. The cell is designed to handle 100,000 curies of any source material emitting gamma rays of 1 MeV. There are three viewing windows and five manipulators -- four master-slave and one heavy-duty electro-mechanical manipulator -- for remotely performing operations in the cell. A three-ton traveling crane is also available for lifting heavy objects inside the cell. Ventilation air from all the cells is exhausted through both charcoal and particulate filters located in the outlet piping from the CDE cell to the 250-ft-high stack. Piping is also installed for discharging the highly radioactive off-gas from the CDE furnace and containment vessel to an off-gas system in the fuel reprocessing plant which contains two 7-ft³ charcoal beds for retention of radioiodines.

Three mixing tanks for preparing decontamination solutions for the CDE cell and equipment are located in the chemical makeup area above the cell. These solutions can be pumped either to a 15-gal tank in the hot cell, from which the solutions are recirculated through the containment vessel, or to a spray header located around the periphery of the cell 10 ft above the floor. Waste liquids from the equipment and cell drain into one of three 500-gal tanks installed in the basement of the building, which are used for nonradioactive, intermediate-activity, and high-activity wastes. The contents of these tanks can be measured and sampled prior to transfer to the ICPP liquid waste storage area.

A schematic flowsheet of the CDE is shown in Figure 2, and a view of the interior of the hot cell is shown in Figure 3. The major elements of the test system are the furnace, the reactor vessel, and the containment vessel. Details of these elements, the sampling systems, and the auxiliary equipment are given in the following sections.

Fig. 2 Schematic flowsheet of the CDE (reactor vessel was removed following Run 1).

Fig. 3 The CDE installed in Cell 2 of the ICPP Hot Pilot Plant (reactor vessel not installed).

1. FUEL MELTDOWN EQUIPMENT

The furnace consists of a 1-7/8-in.-diam by 12-in.-long quartz tube placed inside an induction coil of copper tubing. Power is supplied to the induction coil by a Westinghouse 50 kW, Type 50K64, radio-frequency induction generator. As shown in Figure 4, the major part of the generator is located in the operating corridor outside Cell 2. To provide maximum power at the work coil, a tank coil for operation at 2 to 7 Mc is located inside the cell, approximately 3 ft from the work coil. Power is supplied from the generator through the concrete shielding wall to the tank coil with a coaxial cable encased in an 8- by 8-in. aluminum duct. The work coil is cooled with water to prevent overheating.

In CDE Runs 1, 2, and 3, the fuel pin was placed on a bed of approximately 100 g of powdered UO_2 placed earlier in the bottom of the quartz tube. In subsequent runs, the fuel pin was placed on a 20 g bed of powdered UO_2 in a quartz boat which was then inserted into the quartz tube.

The quartz furnace tube is encased in a Vycor glass jacket through which hot glycol is circulated. The glycol temperature is controlled to maintain the quartz tube below its melting point, but above the condensation temperature of the steam flowing through the tube. Except for the controls, the entire glycol system, including surge tank, steam heater, and pump, is located in the hot cell, as shown in Figure 3.

After the fuel pin is inserted and aligned inside the tube, the transport gas line is remotely connected to the tube inlet. Sealing at both ends of the quartz tube is effected with the large couplings (shown in Figure 5) which compress a silicon O-ring against the quartz tube. The piping at both ends of the tube is fixed rigidly to prevent movement of the tube when the test system is pressurized. The mechanical strength of the quartz tube limits the operating pressure of the system; the tube has been tested to 30 psig, and the CDE runs are made routinely at 22 psig.

Piping connections in the operating area enable various transport gases to be passed over the fuel pin as it melts. However, only steam (from the ICPP steam supply system) has been used to date; this supply is measured with an orifice type flowmeter and controlled manually with a needle valve located in the operating corridor. An argon purge, used to eliminate condensation in the flowmeter taps, gives an argon concentration of about 3% in the steam to the furnace.

Fig. 4 View of CDE operating corridor and induction generator.

Fig. 5 Fuel meltdown furnace assembly prior to Run 5.

2. REACTOR VESSEL

The simulated reactor is a 6-in.-diam by 19-in.-high stainless steel vessel constructed with insulation and electric heaters to control the temperature of the walls at any point between ambient and 1500°F. Ring-type deposition coupons can be placed in the vessel for deposition tests by slipping them on a bayonet heating element inserted in the top of the vessel. Removal of the coupons is done with the remote handling equipment. In addition to the inlet and outlet piping, connections are installed for flushing and draining the vessel. The reactor vessel was used in the initial CDE run, and then removed to increase the transport of activity attributable to the less volatile nuclides, eg, Ce-141, Zr/Nb-95, Ba/La-140, and Sr-90, to the containment vessel in order to better evaluate decontamination procedures and sampling devices.

3. CONTAINMENT VESSEL

The simulated containment vessel is a 5-ft-diam by 5-ft-high stainless steel tank with elliptical heads and a volume of approximately 86 ft³. A copper coil, through which either steam or water can be circulated, is wound around the exterior of the vessel. The outside surface of the tank and the copper coil are covered with fiberglass insulation approximately 1 in. thick; a 1/16-in. thick stainless steel jacket protects the insulation from decontamination solutions. Maximum design pressure of the tank is 50 psig at 300°F. Operating experience has shown that the tank can be heated to 275°F with the coils, or by introducing steam directly into the vessel.

Numerous nozzles are provided in the vessel, as shown in Figure 6, for introducing air, steam, decontamination solutions, as well as for inserting the sampling devices and draining the vessel. Three large nozzles (15-, 8-, and 4-in. diam) are provided on the top of the vessel for installing long or heavy sampling devices (eg, LOFT sample capsule rack, deposition tube), and four 4-in. diam nozzles have been provided in the side for inserting small sampling devices, such as racks of coupons. All nozzles are fitted with couplings that are operated remotely so the sampling devices can be removed using the available remote-handling equipment.

Decontamination solutions are supplied to the tank through a circular spray header fitted with four full-cone, multiple-head nozzles. The decontamination equipment in the cell includes a 15-gal tank and a 50-gpm pump for circulating solutions through the vessel. Decontaminating solutions are blended in the chemical makeup area above the cell and then pumped to the 15-gal tank, from where they are circulated through the vessel.

Fig. 6 Cutaway view of containment vessel showing orientation of sampling devices.

4. SAMPLING DEVICES

Numerous sampling devices have been employed in the CDE runs. Most of these were prototype LOFT samplers, or were installed to obtain data on a specific LOFT problem, eg, evaluation of coatings. Descriptions of the samplers are given in the following paragraphs.

4.1 Gas Samplers

Three types of devices were used to sample the atmosphere of the containment vessel -- Maypacks, gas bombs, and the Continuous Sampler-Monitor (CSM). The Maypacks were used as part of the evaluation program on the LOFT gas-particulate sample capsules, shown in Figure 7. The capsule contains a cartridge, referred to as a Maypack, in which the gas sample is drawn successively through particulate filters, silver screens, and charcoal beds to collect particulate fission products and volatile iodine species[10]. The capsule also contains a void space in which a portion of the gas passing through the cartridge is trapped. Analysis of this gas provided data on xenon and krypton. Because of the limited quantity of data that could be obtained with two capsules on the support rack in each run, four additional cartridges were mounted on the 15-in. flange of the containment vessel (Figure 8) for testing in Runs 3 and 4. As the activity levels of the capsules and Maypack cartridges after sampling were generally high ($> 25R$), removal of the samplers from the containment vessel and disassembly of the cartridges into separate components for packaging was performed in the hot cell, using the available remote-handling equipment (Figure 9).

Numerous gas samples were taken in gas bombs which were connected to the containment vessel by a short length of heated tubing. The inlet and outlet valves on the bomb were opened, allowing the containment atmosphere to flow through the bomb to the process off-gas system, until the bomb was flushed with containment atmosphere. Quick-connects in the connecting lines permitted the bombs to be removed from the hot cell immediately for transfer to the counting area.

Fig. 7 LOFT gas-particulate and plateout capsules mounted on support rack for installation in the containment vessel.

Fig. 8 Maypack cartridges mounted on 15-in. flange for installation in the containment vessel.

Fig. 9 Equipment for remote disassembly of samplers.

A Continuous Sampler-Monitor (CSM) is being developed for use in sampling the containment building atmosphere to determine the fission product concentrations during the Loss-of-Fluid Test. The CSM, used in CDE Runs 3 through 5, is shown schematically in Figure 10. This CSM uses a scrubber (Figure 11) located in the containment vessel to sample continuously the atmosphere and separate the fission products into two streams, which flow separately through the containment wall to radiation monitors and samplers located in the adjacent hot cell. The scrubber, located in the containment atmosphere to avoid problems with fission product deposition on long sampling lines, washes nearly all of the condensable fission products from the atmosphere sample into a liquid stream, leaving mostly noble gas fission products in the gas stream. Scrubbing of the atmosphere sample allows the condensable and ingestible fission products -- primarily iodines -- to be monitored separately from the more active but less hazardous noble gases. A model of the CSM scrubber was installed in the CDE containment vessel for testing in each CDE run, starting with Run 3. The liquid and gas streams were sampled at a panel located in an adjacent hot cell (see Figure 12).

Fig. 10 Continuous Sampler-Monitor schematic flowsheet.

4.2 Condensate Samplers

Condensate from the CDE containment vessel was collected, measured, and sampled for analysis in each CDE run. However, obtaining satisfactory samples proved to be difficult for two reasons -- the need for taking samples rapidly during the high rate of condensate formation early in each run, and a tendency for particulate fission products to settle out in the collection tank. Because of these problems, the condensate collecting and sampling system was altered several times. A flow sheet of the system used in CDE Run 5 is shown in Figure 13.

4.3 Deposition Samplers

Several samplers were utilized to obtain fission product deposition data in the CDE. As part of a program to evaluate protective coatings and decontamination methods for the LOFT containment building, coupons varying in size from 3/4 by 1-1/2 in. to 12 by 48 in. and representing various surface

materials, were contaminated in the CDE. Typical installations of the coupons are shown in Figures 14, 15, and 16. Two spherical LOFT plateout capsules were tested in Runs 3, 4, and 5. These are shown in Figure 7, located on the rack in which they were mounted in the CDE containment vessel. Each of these capsules contains two plateout coupons coated with materials being considered for use on the LOFT containment surfaces[10]. A cooled 12-in.-wide by 48-in.-long panel (Figure 17) to which several LOFT coupons were attached, was contaminated in the I-131 tracer runs to compare deposition on a coupon with that on a nearby wall.

Three types of samplers were used to study the effect of condensation on fission product deposition. In Runs 1, 2, and 3, two samplers (Figure 16) were used, which could control the coupon temperature by passing water or steam through the rectangular support to which the coupons were fastened. In Runs 3, 4, and 5, another deposition sampler was used, consisting of one tube cooled with water and one noncooled tube. This sampler (Figure 18) hung vertically inside the vessel and was fabricated so that 1/2-in.-long sections of the tubes could be removed for analysis.

The third type of sampler is shown in Figure 19. This unit consisted of an assembly for holding five LOFT deposition coupons flush with the containment wall, and five at a distance of 6 in. from the wall where the LOFT capsule sampler would normally be located. Two samplers were used; one was fitted with a cowl to prevent condensate flow from the wall over the coupons, and a second, without a cowl.

4.4 Decontamination Solution Samplers

Data on fission product activity removed during decontamination of the CDE containment vessel were obtained by sampling the liquids collected in the 15-gal circulation tank located in the cell. The samples are taken in 15-ml sample bottles, using a standard jet-type sampler used for ICPP process solutions, and then carried in a cask to the counting area. After the decontamination solutions are measured and sampled, they are drained to one of the three tanks in the basement of the facility. These tanks are equipped with similar samplers and level indicators. Samples of the tank contents were generally taken to verify the samples of the individual decontamination flushes.

Fig. 11 CSMScrubber.

Fig. 12 Sampling station for Continuous Sampler-Monitor.

Fig. 13 Flowsheet of condensate sampling system.

Fig. 14 Deposition coupons on racks ready for installation in the containment vessel.

Fig. 15 Deposition panels (12 x 12 in.) in the support rack being lowered into the containment vessel.

Fig. 16 Controlled-temperature samplers.

Fig. 17 Details of 12 x 48 in. deposition panel.

Fig. 18 Deposition tube sampler.

Fig. 19 Wall plateout samplers.

5. SAMPLE HANDLING AND ANALYSIS

Because of the numerous and varied samples (material, geometry, and activity levels) taken in each CDE Run and the high activity associated with many of them, the sampling operations proved to be a substantial effort. The sampling devices were removed from the containment vessel with the remote handling equipment available in the hot cell. The large sampling devices (eg, the LOFT capsule samplers) were generally disassembled in the cell, while the small sampling devices (deposition coupons) were disassembled in a shielded glove-box. In the initial run, it was discovered that iodine desorbed slowly from the samples. Therefore, to avoid biased results as well as the contamination of working areas, all samples were sealed (Figure 20) in bags of a Mylar-[a] polyethylene laminate. Four to five hundred separate samples were packaged and analyzed in each run.

Most of the samples were analyzed for fission products by gamma-ray spectrometry, supplemented by chemical separations especially for the less-

[a] Polyethylene terephthalate resin, product of E. I. duPont deNemours and Co.

volatile, low-activity fission products. For very high activity samples (5 to 50 R/hr), a special counting cave was erected in the corridor to the south of the CDE hot cell. This facility (Figure 21), permitted the sample to be counted at distances up to 40 ft from the NaI scintillation crystal. The counting equipment, a 512-channel gamma-ray spectrometer, was located in the analytical area, approximately 300 ft away. Because of the large number of samples accumulated in each run, a shielded and ventilated storage area (Figure 22) was constructed in Cell 5 to the east of the CDE hot cell.

Fig. 20 Sample bagging equipment.

Fig. 21 High-level counting cave and counting equipment.

Fig. 22 Sample storage barricade.

III. RUN PROCEDURES AND CONDITIONS

The CDE run conditions were chosen to simulate the conditions anticipated for the originally planned unperturbed LOFT (LOFT-U). All CDE runs were conducted as "natural effects" tests, ie, the fission product release, transport and containment behavior were unperturbed by the use of any engineered safeguard systems (with the exception of the containment vessel). The general procedure and the conditions for the CDE runs are summarized in the following sections.

1. RUN PROCEDURE

In general, the procedures followed in each of the five high-level runs were similar. A miniature (9/16-in. diam by 3.5-in. long) fuel pin, encased in an aluminum capsule (Figure 23), was irradiated in the Materials Testing Reactor (MTR). The welded aluminum capsule provided secondary containment for the pin in the reactor and also prevented boiling on the capsule surface during irradiation. The capsule was removed from the reactor at the end of an MTR operating cycle and transferred to the ICPP Hot Pilot Plant (HPP) in a fuel transfer cask. At the HPP, the cask was moved into the CDE cell where the capsule was removed and placed in a bucket of caustic to dissolve the outer aluminum capsule. After the capsule was dissolved, the fuel pin was rinsed, allowed to dry, and placed remotely on a bed of UO_2 powder in the quartz furnace tube. The furnace tube was then sealed remotely and tested for leaks with dry air at 30 psig. Except for Run 1, in which difficulties were experienced in sealing the furnace, the elapsed time from shutdown of the reactor to the start of fuel pin melting varied between 7 and 10.5 hr.

Fig. 23 Fuel pin and capsule.

Prior to transferring the fuel pin to the furnace, the transport gas line, the furnace, and the containment vessel were heated to the desired test temperatures and allowed to equilibrate. After the furnace was tested and found to be leak tight, it was vented to the containment vessel, and the containment system was pressurized to 22 psig with a saturated mixture of air and steam. The furnace was purged with steam, and a controlled flow of steam containing

a small amount of argon (from an instrument purge) was started across the fuel pin which was melted with the induction coil. The fission products released from the fuel were transferred to the containment vessel by the flowing steam. The temperatures of most of the samplers in the containment vessel were noncondensing with respect to the containment atmosphere at the time of meltdown. The time elapsing between the start of heating and the melting of the fuel (which is indicated by a drop in the power consumption of the induction generator) varied from 4 to 34 min. The pins were maintained in a molten condition for periods of 20 to 44 min. Steam was added to the containment vessel during the meltdown period to maintain the pressure at 22 psig.

After each melting period was completed, the induction generator was turned off, and the steam flow over the fuel pin was stopped. The furnace was then purged with argon to the containment for 1 to 2 min to remove residual steam. Following this, the furnace and containment vessel were isolated, and the furnace was depressurized slowly through a vent line containing a charcoal filter to collect any iodine released during the depressurization. The steam flow through the furnace to the containment vessel was discontinued simultaneously with the isolation of the furnace. The temperature of the containment vessel atmosphere was then controlled by cooling and heating coils over a 24-hr period to match the expected temperature decay for the LOFT containment building. During the 24-hr period, the containment atmosphere and condensate were sampled, and the condensate rate in the vessel was measured. Approximately 24 hr after isolation, the containment vessel was vented, and the sampling devices were removed remotely for disassembly and packaging of the samples. At that time, the containment temperature and pressure had decayed to near ambient levels. A small mixing fan located in the containment vessel was started prior to the fuel melting, and operated throughout the 24-hr isolation period.

Following removal of all samples from the cell, the fuel pin residue and quartz furnace tube were placed in separate casks and moved to another hot cell where the fuel residue was dissolved and sampled. The CDE cell was then decontaminated, first by spraying the exterior of the equipment and cell walls, and, finally, by manual cleanup using a portable spray rig.

The only significant deviations from this procedure occurred in Runs 1 and 3. In Run 1, the meltdown was performed at ambient pressure because of a furnace connection which leaked under pressure. Following the meltdown, the containment vessel was pressurized to the desired level with air and steam. In Run 3, the furnace tube broke just prior to isolation of the furnace from the containment. The break scattered fuel residue throughout the cell; however, the isolation was effected so rapidly that little gas was released from the containment vessel.

Operating procedures for the four tracer-level runs were similar to those of the high-level CDE runs, except that the run duration was shorter and the containment vessel was not preheated. The CDE containment vessel was leak tested, pressurized to approximately 1.5 psig with dry air, and steam was injected to increase the containment pressure and temperature to approximately 22 psig and 215°F, respectively. After this pressure was reached, a glass ampule containing a previously measured amount of either I-131 or Xe-133 was broken and swept with argon into the CDE containment vessel over a 3- to 4-min

period. The mixing fan in the containment vessel was operated throughout the runs. During, and for 5 min after the injection, the containment pressure was maintained at approximately 22 psig by injecting steam. Following this period, the temperature of the containment atmosphere was allowed to decay naturally for 5 to 8 hr. The pressures and temperatures of the containment atmosphere during the tracer runs were similar to the early high-level runs.

2. RUN CONDITIONS

The fuel pins used in all the runs, except Run 1, were fabricated at the Battelle Northwest Laboratories (BNWL) from 8% enriched UO_2 pellets and Zircaloy tubing. The pin used in CDE Run 1 was fabricated in the NRTS shops of 1.5% enriched UO_2 pellets and Zircaloy tubing. Details of the pins supplied by BNWL are shown in Figure 24. Each pin was irradiated in the MTR for one or two operating cycles (16 to 35 days) at a flux of approximately 5×10^{13} neutrons/cm²-sec. The centerline temperatures of the fuel pins were below the melting temperature of UO_2 during irradiation, except for Run 2. An estimated 50% of the UO_2 in the fuel pin used in Run 2 was molten during irradiation.

Fig. 24 Details of CDE fuel pin and irradiation capsule.

Operating conditions for the five CDE runs and estimated conditions for a LOFT-U test are compared in Table II. Additional comparisons are also shown in Table I. Pressure and temperature conditions in the CDE containment vessel for a typical run are compared with those anticipated in the LOFT-U in Figures 25 and 26.

TABLE II
COMPARISON OF OPERATING CONDITIONS FOR CDE RUNS 1 THROUGH 5
WITH ANTICIPATED LOFT U-CONDITIONS

Variable	CDE Run 1	CDE Run 2	CDE Run 3	CDE Run 4	CDE Run 5	Anticipated LOFT-U
Fuel Pin:						
Diameter (inches)	0.562	0.562	0.562	0.562	0.562	0.411
Length (inches)	3.45	3.45	3.45	3.45	3.45	36
Cladding	Zircaloy-2	Zircaloy-2	Zircaloy 2	Zircaloy-2	Zircaloy-2	Zircaloy-4
Fuel Type	UO ₂ Pellets	UO ₂ Pellets	UO ₂ Pellets	UO ₂ Pellets	UO ₂ Pellets	UO ₂ Pellets
Total Uranium (grams)	74.6	70.2	70.2	69.9	69.9	545
Enrichment (percent)	1.5	8	8	8	8	2.4 / 5.3
Fuel Burnup (MWD/Tonne)	1200	2000	740	530	460	940
Irradiation Flux Level (n/cm ² -sec)	5.2 x 10 ¹³	3.5 x 10 ¹³	6.8 x 10 ¹²	7.9 x 10 ¹²	8.5 x 10 ¹²	3.5 x 10 ¹³
Irradiation Time (days)	34.4	16.3	30.4	18.5	16.0	16.6
Total Activity in Fuel at Reactor Shutdown (curies)	10,600	35,250	7,100	7,800	8,200	2.10 x 10 ⁸
Iodine-131 Activity in Fuel at Reactor Shutdown (curies)	63.3	171	41.5	39.9	39.5	1.24 x 10 ⁶
Cooling Time until Fuel Meltdown (hours)	30.8	10.5	7	9.75	9.0	0.1 - 1.0
Time after Start of Heating until Initial Fuel Melting Occurred (min)	23	5	4	7	34	5 - 60
Time Fuel was Molten (min)	44	26	20	25	23	---
Composition of Transport Gas over Fuel	Steam and 3% air	Steam and 3% argon	Steam and 3% argon	Steam and 3% argon	Steam and 3% argon	Steam
Velocity of Transport Gas over Fuel (ft/sec)	1.4	1.7	1.8	1.6	1.3	0.11 - 1.4
Furnace and Containment Vessel Pressure During Meltdown (psig)	0	22	22	22	22	19 - 26
Containment Atmosphere Temperature During Meltdown (°F)	212	250	218	224	215	217 - 245
Composition of Containment Atmosphere During Meltdown	100% steam (air and steam added after meltdown to achieve 36% air and 64% steam at 22 psig)	36% air and 64% steam	50% air and 50% steam	50% air and 50% steam	50% air and 50% steam	60% steam and 40% air
Number of Sampling Systems Installed:						
Raw Gas Sample Bombs	1	1	2	2	2	0
Gas-particulate Cartridges	2	4	4	4	0	0
Gas- particulate Capsules	0	2	2	2	4	219
Plateout Capsules	0	2	2	2	0	456
Plateout Coupons (at ambient temperature)	168	114	156	170	138	0
Plateout Coupons (heated)	12	16	16	0	0	0
Plateout Coupons (cooled)	12	16	16	0	0	0
Continuous Sampler-Monitor	0	0	1	1	1	3
Deposition Tube (heated)	0	0	1	1	1	0
Deposition Tube (at ambient temperature)	0	0	1	1	1	0

Fig. 25 Containment atmosphere temperature during Run 2.

Fig. 26 Containment pressure during Run 2.

IV. FISSION PRODUCT BEHAVIOR

The principal objective of the AEC's reactor safety program is to establish that licensing and regulation criteria for commercial power reactors will provide adequate protection of the public. Because of the excellent safety record of the nuclear industry, little accident experience is available, and the licensing criteria are based primarily upon experimental investigations. The difficulties of handling large quantities of fission products have required that most of the experimental work be done on a small scale and at low levels of radioactivity. Also, the experiments are generally conducted to evaluate a specific phase of the accident sequence -- the release of noble gases at the instant of cladding failure or the washout of elemental iodine in the containment, as examples.

The CDE, although of small size compared to a commercial reactor facility, represents an integrated test, ie, an attempt was made to simulate the entire accident sequence, fuel irradiation, fuel melting, transport of the fission products to the containment, and the natural removal of fission products from the containment atmosphere. Thus, operation of the CDE for the LOFT support programs to evaluate samplers, protective coatings, and analytical methods has provided an excellent opportunity to obtain information on unperturbed fission product behavior in an integrated test at high-activity levels and with short-cooled fuel.

Because emphasis was placed on testing of prototype sampling units, limitations on the number and scheduling of samples made it difficult to obtain time-dependent concentrations in the containment vessel. Despite this, the large quantity of data accumulated in the CDE falls into a consistent pattern describing the extent, distribution, and characteristics of a fission product release from a melting fuel element under simulated loss-of-coolant accident conditions. These data are presented and discussed in the following paragraphs.

1. FISSION PRODUCT AND FUEL MATERIAL ACCOUNTABILITIES

One of the most difficult problems encountered in high-level fission product experiments is that of obtaining a satisfactory material balance. The fission product accountability determines to a large extent the credibility of the data from the experiment, particularly with respect to the fission product release. In an effort to improve the quality of the data obtained in the CDE runs, material balances were calculated for the fission products of major interest and the fuel materials, uranium and zirconium. These data are presented in Table III for the five runs (the run conditions are summarized in Table II).

The fission product accountabilities shown in Table III were determined by totaling the fission products found in the fuel residue, meltdown furnace, transport line, containment condensate, decontamination solutions, and the atmosphere at the end of the run, and then dividing these totals by the calculated inventories of fission products available in the fuel. All activity values from

TABLE III

FISSION PRODUCT, FUEL, AND CLADDING ACCOUNTABILITIES

Fission Product or Fuel Material	Percent Recovered				
	CDE-1	CDE-2	CDE-3	CDE-4	CDE-5
Sr-89	27	140	137	112	128
Zr-Nb-95	27	18	90	116	51 ^[e]
Mo-99	11	4 ^[a]	66	101	130
Ru-103	18	14	100	74	44
I-131	31	28	19	57	57
Te-132	7	9	26	22 ^[c]	68
Cs-137	84	23	27	93	85
Ba-140	36	74	89	83	97
Ce-141	55	10	91	47	101
Uranium (Fuel and Bed)	98	101	85 ^[b]	107	98
Zirconium (Cladding)	103	98	[d]	103	[d]

[a] Fuel residue solution analyzed too late to obtain reliable data.

[b] Based on the U-235 recovered from the fuel pin residue.

[c] Not reported in the fuel residue sample.

[d] Not determined.

[e] Zr-95 only.

an individual run were corrected to a common time -- either the time of reactor shutdown or the time of containment vessel isolation. The fission product inventories were calculated using the RSAC Curie Code^[11] and fuel pin burnup. The burnup was determined for each run by totally dissolving the melted fuel pin and then determining the change in uranium isotopic ratio (U-235/U-236) which occurred during irradiation. To check the isotopic ratio method, the burnup of the pin used for Run 3 was determined by (a) analysis of flux wires irradiated with the fuel pin and (b) the uranium isotopic ratio. The burnups obtained by the two independent methods agreed within 5%. The accountabilities of fuel materials were obtained by comparing the fuel materials recovered after a run with the fuel specifications.

As shown in Table III, the fission product accountabilities improved generally with each succeeding run, while the uranium and zirconium accountabilities were excellent for all but Run 3, in which the furnace failed. Based on the latter CDE runs, the accountabilities of the low- and moderate-volatility fission products (Zr-Nb-95, Sr-89, Mo-99, Ru-103, Cs-137, Ba-140, Ce-141) were, in general, 80 to 120% of the calculated inventories, while those of the volatile fission products (I-131 and Te-132) were consistently low at 60 to 70% of the inventories.

A comparison of the excellent uranium and zirconium accountabilities with the poorer accountabilities for the fission products (particularly I-131, Te-132) illustrates the difficulties encountered in sampling and analyzing fission products. Examples of specific problems encountered in the CDE runs include (a) a large proportion of the Te-132 in the condensate was found associated with particulates which settled in sampling lines and sampling bottles, (b) iodine was found concentrated at the top of sample bottles in the rubber sealing gasket, changing the counting geometry from that used for calibration, and (c) recent investigations show that certain iodine species (particularly organic iodides and to a lesser extent hypoiodous acid) are volatile and are easily lost from liquid samples. The difficulties encountered in the CDE tests will be observed in other tests and in the LOFT, unless extensive efforts are expended to characterize all significant fission product species, establish accurate counting geometries for the samples, and pinpoint the causes of the sampling problems.

2. FISSION PRODUCT RELEASE, TRANSPORT, AND DISTRIBUTION

Following the loss-of-coolant in a reactor accident, fuel rod temperatures rise from the heat released through fission product decay and cladding-water reaction. As the temperatures increase (assuming no engineered safeguard systems are operated), the release of fission products proceeds through three stages: (a) the prompt release of gaseous fission products collected in the fuel-cladding gap to the reactor coolant or atmosphere at the instant of cladding failure, (b) the slower release of fission products which diffuse through the fuel and escape through the failed cladding as the fuel temperature continues to increase (significant quantities of particulates, particularly tin oxides from the cladding, are released during this step), and (c) breakup of the fuel rod, creating particulates and, if the temperatures are high enough, slumping and vaporization of the fuel with further release of fission products.

During normal operation of a reactor, fission products are released to the fuel element cladding gap by fission fragment recoil and by diffusion through the fuel, particularly along cracks and grain interfaces, toward the cool, outer surface of the fuel pellets. The noble gases and iodines, being volatile and thus more mobile than the other fission products, make up the bulk of the fission product activity collected in the cladding gap and also the "prompt" release when the cladding is initially breached.

As the fuel cladding temperature approaches 1830°F, the Zircaloy cladding begins to react exothermally with steam to form ZrO_2 ^[12]. The ZrO_2 , being

friable and having low mechanical strength, distorts and fractures. The Zr and ZrO_2 also go through crystalline phase changes^[13] which affect the structural stability and physical properties of the cladding. It is possible that solid solutions of UO_2 and ZrO_2 , and Zr and stainless steel will be formed and will affect the cladding stability at higher temperatures. For example, solid solutions have been observed in melted fuel residues^[14], and Zircaloy can form an eutectic mixture with stainless steel at temperatures as low as 1715°F^[15].

When the cladding temperatures approach melting, significant quantities of particulates are released, primarily as oxides of tin (tin is used as an alloying component of Zircaloy). As the fuel temperature rises above the melting point of UO_2 , molten UO_2 oozes through cracks in the cladding and is exposed directly to steam flowing through the reactor. When sufficient fuel has slumped and collected in the bottom of the reactor, additional melting can take place which will release more fission products through vaporization. The manner in which the CDE fuel pins were melted most closely simulates the last stages of release, ie, that which occurs during total melting of the UO_2 .

The fission products released from the fuel in an unperturbed accident will be carried by steam generated from water remaining in the reactor through the intact portions of the core and out the primary coolant line to the containment building. The fission products will be released both as gases (eg, the noble gases and iodines) and as particulates generated by the vaporization of fuel or by adsorption of fission products on nonradioactive particulates from the fuel cladding (eg, tin oxides). Significant deposition of fission products from the gas stream takes place in the reactor internals and primary piping. The extent of deposition is dependent upon the chemical species and form (gaseous, particulate, etc) of the fission products, the temperature difference between the gas and surface, the gas velocity, the reactor and piping geometry and surface conditions (eg, stress, corrosion film, etc), and the environmental conditions.

Once in the containment vessel, the fission products, excluding the noble gases and the organic iodines, will be removed rapidly from the atmosphere by deposition on surfaces and washout in the condensing steam. Unless extensive rainout (the significance of this removal mechanism has yet to be demonstrated) occurs in the containment building, removal rates and, hence, the distribution of the iodines and particulates will be controlled by their movement from the atmosphere to the building surfaces which act as "sinks" upon which the activity is deposited. Part of the fission products deposited may be removed by condensate washing over the surfaces. The movement of the gaseous iodine to the walls occurs by diffusion due to the concentration gradient, transport with the bulk flow of steam toward the wall upon which condensation is occurring, and transport due to the natural convective air currents (caused by buoyancy effects) within the building. Transport of particulate fission products to the wall is affected by gravitational settling, in addition to those factors mentioned previously. The chemical form of the fission products and the size of the particulates will also influence the removal rates. Elemental iodine, being reactive and soluble in water, is removed quite rapidly from the atmosphere, while the organic iodide forms are removed more slowly. Large particulates move more slowly toward the walls but settle faster than small particulates. The relative efficiencies of the mechanisms determine the overall distribution of fission products in the containment building.

2.1 Fuel Pin Meltdown

The physical deterioration of the fuel pin during the CDE meltdowns is difficult to describe quantitatively, since fuel temperatures were not measured; however, the following general sequence of events was observed in each meltdown. Little damage was evident until the cladding became orange-hot ($> 2000^{\circ}\text{F}$). At that point, small cracks appeared in the cladding, and although not detected by the high-range radiation instruments in the cell, small quantities of fission products (the "prompt" release) were probably released. Shortly after the cladding becomes white-hot ($> 2700^{\circ}\text{F}$), general deterioration of the pin occurs. Flakes of cladding spall off, and sparks are created as the cladding is contacted by occasional water droplets. The UO_2 melts through the lower portion of the Zircaloy cladding (the lower half of the CDE fuel pins is protected by a bed of UO_2 powder, and, hence, is not converted to ZrO_2). During this interval (1 to 2 min), the main fission product release ($> 90\%$ of the total release) occurs. Shortly after the major fission product release, gross quantities of particulates (primarily oxides of tin and uranium) are released in the form of a white smoke which is transported to the outlet end of the furnace by the flowing steam. Extensive deposition of greenish-brown particulate materials (primarily uranium oxides), occurs at the outlet end of the furnace. A continuous slow release of fission products occurs during the period the fuel is maintained in the molten state, and is generally accompanied by one or more smaller releases. No further release of fission products takes place as the pin cools following shutdown of the induction generator. A plot of the activity levels in the containment vessel during a fuel pin meltdown is shown in Figure 27, and the fuel residue from a typical meltdown is shown in Figure 28.

Fig. 27 Typical activity release to containment vessel during a CDE fuel pin meltdown.

(a) Typical Fuel Residue in quartz boat

(b) Typical Fuel Residue after removal

Fig. 28 Typical melted fuel residue.

As shown in Figure 28, most of the UO_2 fuel and the protected lower portion of the Zircaloy cladding melt. The upper portion of the cladding is converted to a ZrO_2 crust which does not melt and is stable enough to maintain its shape. The ZrO_2 crust is quite brittle, however, and fractures easily during removal of the pin from the boat (shown in the lower portion of Figure 28). Since the UO_2 fuel melted but the ZrO_2 did not, estimates of the approximate fuel temperature range could be made. These estimates indicate that the fuel center line reaches maximum temperatures of 5160 to 5430°F.

To obtain additional information on the cladding failure mechanisms, one meltdown was conducted with an unirradiated fuel pin, and the residue was examined by X-ray diffraction, optical metallography, and microprobe analyses. The upper one-half of the Zircaloy cladding was completely oxidized to ZrO_2 (melting point of approximately 4890°F), and was fractured but did not melt. The lower, protected portion of the Zircaloy cladding did not oxidize but was melted (melting point of approximately 3360°F). Most of the UO_2 (melting point of approximately 5125°F for low burnup fuel) was melted, and metallic inclusions, identified as a uranium-zirconium alloy, were found in the cladding- UO_2 boundary region. The zirconium content of the matrix surrounding these inclusions varied up to 18 wt%. This matrix is probably a UO_2 - ZrO_2 solid solution.

During Run 5, a mass spectrometer was installed to measure the concentrations of N_2 , He, H_2 , O_2 , A, and CO_2 in the furnace. Figure 29 shows the concentrations of N_2 , O_2 , and H_2 on a "dry-gas" basis as measured in the furnace outlet line during the Run 5 meltdown. The He (from the blanket gas in the fuel pin and from ternary fission) concentration was below the detectable limit, and A (added as a transport-line flowmeter-purge) was present in the concentration expected. The gas analyses indicated a small amount of air was present during meltdown; this is believed to have been due to incomplete purging of the spectrometer sampling system prior to sampling. The results also showed a buildup and subsequent decrease of H_2 during the meltdown and an increase in the N_2 to O_2 ratio, indicating depletion of O_2 . The depletion of O_2 and the buildup of H_2 indicated that a metal-water reaction does occur under the CDE run conditions.

2.2 Release of Fission Products Other Than Krypton and Xenon

The percentages of fission products released from the fuel during Runs 1 through 5 are shown in Table IV. These releases are based on fission product inventories calculated using the RSAC Curie Code^[11] and the analyses of all samples. All activities were corrected to a specific time -- the time of reactor shutdown for Runs 1, 2, and 3 and the time of meltdown for Runs 4 and 5. The fissions occurring in the fuel pin during irradiation, required as input data for the RSAC Curie Code, were computed from pre- and post-irradiation uranium isotopic analyses.

The releases shown in Table IV agree relatively well for the five runs, even though several meltdown parameters, such as the rate of temperature increase during the fuel meltdown and the transport gas velocity over the fuel pin during melting, were varied. The maximum temperature attained by the fuel during the runs also varied but was not measured. Part of the variance noted between the early and later runs is probably due to improvements in the sampling and analysis techniques which evolved throughout the program.

The releases of fission products and fuel materials (arranged in order of descending amount released) obtained in Runs 1 through 5 are compared with certain physical properties of the materials in Table V. This comparison illustrates that, generally, the order of the quantities released is the same as the order of the vaporization temperatures of the species in the fuel. Three significant exceptions -- that of tellurium, molybdenum, and ruthenium -- are apparent. In these cases, the diameters of the molybdenum and ruthenium compounds and the tellurium molecule are smaller than the materials having the next lowest vaporization temperature. The smaller diameter molecules have larger diffusivities and should be more mobile than the larger diameter molecules. Further, the release of molybdenum and ruthenium should be higher than that of other materials of comparable vaporization temperatures (BaO , SrO , and ZrO_2), since molybdenum and ruthenium do not form solid solutions with UO_2 , but form volatile oxides. The partial pressures of molybdenum and ruthenium in the fuel are near the pressures expected for the pure materials, while those of BaO , SrO , and ZrO_2 -- which probably form solid solutions with UO_2 -- are orders of magnitude lower than the pure components. In the CDE runs, the fuel was maintained at a temperature slightly above the melting point of UO_2 (approximately 5125°F) for only about 30 min. Had the fuel been maintained molten for a period of several hr, the release percentages for the high boiling materials BaO , Ru , SrO , UO_2 and ZrO_2) would have been greater.

The only available data on fission product releases obtained under simulated unperturbed LOC accident conditions for a reactor are compared in Table VI. These releases were obtained by the induction melting of Zircaloy-clad UO_2 fuel pins of varying burnups in a steam atmosphere. The I-131 releases reported by Collins and Hillary^[19,20] varied between 13.7 and 15.4 and 8 and 35% for low burnup fuel pins (1 and approximately 100 MWd/te, respectively). These releases are comparable to the 15 to 44% releases obtained in the CDE for fuel pins with burnups of 460 to 2000 MWd/te.

Release fractions of several fission products from a high burnup (7000 MWd/te) fuel pin have been reported by Creek, et al^[21]. The pin used in this test was allowed to decay for several years after the initial irradiation and then was reirradiated briefly just prior to melting to increase the I-131 content to a measurable level. A steam-air mixture was started over the fuel pin and maintained until the fuel temperature reached an estimated 4000°F . At this temperature, the steam flow was discontinued because of steam condensation

Fig. 29 Concentrations of N_2 , O_2 , and H_2 in furnace outlet line during meltdown.

TABLE IV

FISSION PRODUCTS AND URANIUM RELEASED FROM CDE FURNACE

Fission Product or Fuel	Percentage Released ^[a] From Furnace				
	CDE-1	CDE-2	CDE-3	CDE-4	CDE-5
Sr-89	<0.01	0.05	0.02	0.03	---
Zr-Nb-95	Trace	Trace	0.1	0.001	0.0001
Mo-99	2.5	4	8	6.5	Trace
Ru-103	<0.01	0.07	0.7	0.2	0.003
I-131	15	17	17	24	44
Te-132	7	9.5	16	22	12
Cs-137	11	13	21	8	3
Ba-140	[b]	0.04	0.04	0.2	0.05
Uranium	[b]	[b]	0.26	0.05	[b]

[a] Determined by dividing the sum of the fission products found in the containment vessel, simulated reactor vessel, and furnace outlet line by the total amount of each fission product present (determined by RSAC Curie Code and isotopic analyses) in the fuel. Does not include fission products deposited in the quartz furnace.

[b] Insufficient data.

in the furnace tube. Dry air flow was continued for the balance of the heating period (creating a more oxidizing atmosphere than the steam-air mixture). As shown in Table VI, the releases of I-131, Sr-89, and Cs-137 obtained in the ORNL experiment were higher, and the release of Te-132 was lower than those obtained in Runs 1 through 5.

A comparison of the data obtained in the different experiments shows a trend toward an increase in the iodine release with an increase in fuel burnup. This could be due to grain growth or an increase in fuel fracturing with increasing burnup, which would tend to increase the iodine mobility during the heatup of the fuel prior to melting. More likely, however, the fuel temperatures reached or the melting periods differed, resulting in the variations in the reported releases.

TABLE V

COMPARISON OF FISSION PRODUCT RELEASE AND PHYSICAL PROPERTIES^[13,16-18]

Probable Major Species in Fuel	Probable Major Vapor Phase Species	Range of Fission Product Release (% of Inventory)	Radius of Atom or Molecule in Fuel ($\text{\AA}$)	Vaporization Temperature of Major Species in Fuel ($^{\circ}\text{C}$)
Xe	Xe	89	1.90	-107
Kr	Kr	86	1.69	-152
I	I	15 to 44	1.36	184
Te	Te	7 to 22	1.33	1390
Cs	Cs	3 to 21	2.55	670
Mo	MoO ₃	2 to 8	1.36	5560 ^[13]
BaO ^[16]	BaO	0.04 to 0.2	2.67	3000
Ru	RuO ₃ and Ru	0.003 to 0.7	1.33	3900
SrO ^[16]	SrO	0.01 to 0.05	2.45	3500
UO ₂	UO, UO ₂ and UO ₃	≈ 0.05	3.00	3850
ZrO ₂ ^[16]	ZrO ₂ and ZrO	nil	2.12	4100

Based on thermodynamic calculations, the releases of some of the fission products should be affected by the composition of the atmosphere over the fuel pin. For example, an increase in the oxygen content of the atmosphere over the fuel could promote the formation of MoO₃ which would increase the release of molybdenum, while an increase in the H₂ content (which occurs during oxidation of the cladding) would promote the release of zirconium as ZrO. More run data are required to substantiate the thermodynamic calculations.

The quantity of fission products released from the fuel to the containment building following a reactor accident is an important factor in determining the siting criteria for commercial power reactors. The current release fractions suggested by the AEC for modeling a maximum credible LOC accident are documented in TID-14844^[22] as 100, 50, and 1%, respectively, of the total noble gas, iodine, and solid fission product inventories in the core at the time of the accident. Based on the data obtained in the CDE runs, these releases are reasonable on an overall basis for that portion of the core in which the fuel melts. Although the CDE fuel pins were maintained molten for only 20 to 44 min, the releases of tellurium, cesium, and molybdenum exceeded the 1% value quoted in TID-14844. In a reactor accident, a portion of the core would remain molten for several hours, and the releases of tellurium, cesium, and molybdenum from this portion would probably exceed 1%, depending on the composition of the atmosphere over the fuel. However, the total release in a reactor accident should be less than the TID-14844 values because not all of the core will be

TABLE VI

FISSION PRODUCT RELEASE FROM HEATED ZIRCALOY-CLAD UO_2

Experiment	British ^[19]	British ^[20]	CDE	CMF ^[21]
Fuel Burnup (MWd/te)	1 to 2	~100	460 to 2000	7000
Fission Product Release (% of Fuel Inventory):				
Sr-89			0.01 to 0.05	- 0.18
Zr-Nb-95			0.0001 to 0.1	
Mo-99			2.5 to 8.0	
Ru-103			0.003 to 0.7	0.22
I-131	13.7 to 15.4	8 to 35	15 to 44	90.8
Te-132			7 to 22	2.3
Cs-137			3 to 21	30.0
Ba-140			0.04 to 0.2	

molten, and no "credit" was allowed for fission product deposition in the reactor internals and outlet line to the containment (discussed in a subsequent section). Mathematical descriptions^[23] are a more logical method of estimating the release since they consider both time and temperature; unfortunately, the experimental data required as input to the models are not available.

2.3 Release of Krypton and Xenon

To evaluate the performance of the LOFT atmosphere sampling devices on the noble gases, the release of xenon and krypton was determined in Run 4. The fuel residue from the Run 4 meltdown was dissolved, and the xenon and krypton were quantitatively trapped and analyzed by gas chromatography^[25]. The fuel pin was dissolved by contacting it alternately with HNO_3 and HF (the UO_2 fuel dissolves in the HNO_3 , and the Zircaloy cladding dissolves in the HF), and the off-gases from each treatment were collected separately. The xenon and krypton release from the UO_2 was monitored by a NaI(Tl) detector and was continuous over the fuel dissolution period, indicating that the gases remaining in the fuel were bound in the UO_2 lattice. The amounts of gas released during dissolution of the UO_2 were 0.02 std cc of krypton and 0.14 std cc of xenon. The total amount of gas (both xenon and krypton) in the cladding were less than the detectable limit of 0.001 std cc.

The fractions of xenon and krypton released during the Run 4 meltdown are shown in Table VII and were estimated, based on the calculated inventories in the fuel at the time of meltdown and at the time of dissolution (10 days later). The total amount of krypton did not change from the time of meltdown to the time of dissolution, but the quantity of xenon changed, due to the effects of precursors (I and Te) and xenon decay (to Cs-133). To determine the amount of xenon released at the time of meltdown, initial xenon releases of 90% and I₂ and Te releases of 25 and 50% were assumed. Two values were chosen for the I₂ and Te releases, because the observed I₂ release in Run 4 was 25% and the I₂ material balance was only 57%.

TABLE VII
CALCULATED XENON AND KRYPTON RELEASE FRACTIONS

Element	Total Atoms in Fuel at Meltdown	Total Atoms Found in Fuel at Dissolution[a] (Corrected to Meltdown)		Release During Meltdown (%)	
		Case 1 ^[b]	Case 2 ^[c]	Case 1 ^[b]	Case 2 ^[c]
Xe	2.78×10^{19}	2.78×10^{18}	3.17×10^{18}	90	89
Kr	3.83×10^{18}	5.36×10^{17}	5.36×10^{17}	86	86

[a] (Atoms present at time of dissolution) - (Atoms of precursor decaying to Xe) + (Atoms of Xe or Kr decaying to daughter).

[b] Assuming 90% Xe release and 25% release of I and Te.

[c] Assuming 90% Xe release and 50% release of I and Te.

Based on these assumptions, the release fractions for krypton and xenon in Run 4 were 86 and 89 to 90%, respectively (depending on the I₂ and Te release fractions used). The calculations also show that for the Run 4 conditions, the xenon release is not greatly dependent on the amount of iodine and tellurium remaining in the fuel after meltdown.

Visual observations during the meltdown indicated that part of the fuel was not located in the induction coil and, therefore, did not melt. This could account for the <100% release of noble gases during the meltdown and the slow, continuous release of the gases during dissolution of the fuel pin residue.

2.4 Fission Product Transport

The fission products released from the melting fuel pin in the CDE are transported by steam from the furnace to the CDE containment vessel through a 1-in.-diam stainless steel line, approximately 2-ft long. For Runs 4 and 5, the line was installed in three sections which could be separated remotely. The transport line was heated externally to near 800°F, the temperature of each section was monitored, and velocities of 1.3 to 1.9 fps (Reynold's number of 700 to 900) were maintained during the meltdowns.

Following each run, the line was removed and disassembled, and each section was leached separately in 3M HCl followed by 4M HNO₃ to remove the deposited fission products. The quantities of fission products deposited in each section of the pipe, per unit area, are shown in Figure 30 for Run 4. Comparable quantities were found in Run 5. The contamination per unit area was calculated, assuming the deposition was homogenous over the entire inside surface of each pipe section.

The levels and distribution of activity for both Runs 4 and 5 are shown in Table VIII. As shown in Table VIII, approximately 1/10 to 1/3 of the fission products, except for molybdenum, ruthenium, and cesium, released from the fuel were deposited in the transport line. Essentially all of the cesium, but only small quantities of molybdenum and ruthenium were deposited in the line. Because the transport line was heated externally, the temperature gradient from the gas to the wall was the opposite of that which would occur in a reactor accident, both radially and axially. However, the correction (which would increase the deposition) is small[26]. The fractions of fission products deposited in a reactor accident would be much larger than in the CDE because of the large and complex surface area of the reactor internals. The greater deposition density (activity/cm²) in the transport line compared to that observed in the containment vessel (factors of 10 to 100) supports this conclusion.

2.5 Fission Product Distributions

The fission product distributions following a reactor accident are of interest, since they indicate the relative efficiencies of the natural removal processes and the quantity of fission products which could be released from the reactor containment building. For example, the fission products which are released from the fuel but deposit in the reactor internals and coolant line, and

Fig. 30 Deposition of fission products in transport line.

TABLE VIII

DEPOSITION OF FISSION PRODUCTS IN TRANSPORT LINE

Fission Product	Percentage of Release Deposited in Transport Line ^[a,b]		Percentage of Fuel Inventory Deposited in Transport Line ^[b]	
	Run 4	Run 5	Run 4	Run 5
Sr-89	29		0.0085	
Zr-95	37	93	0.00037	0.00011
Mo-99	trace		trace	
Ru-103	2.5	4.3	0.006	0.00013
I-131	20	12.8	4.8	5.8
Te-129		6.7		0.23
Te-132	15		3.3	
Cs-137	8	31.2	0.63	9.9
Ba-140	17		0.019	
Ce-141	100		0.004	

[a] Release considered as sum of nuclide activity in transport line and containment vessel.

[b] Blanks indicate that no data were obtained.

those which are removed in the condensing steam (unless the steam condensate is circulated through the containment building spray system) would not be available for release. The fission product distributions for Run 4, which are typical of those for the other runs, are shown in Table IX. The figures shown for the furnace outlet line, the containment condensate, the containment walls and the painted coupons were calculated, based on the results of sample analyses. The figures for the fuel and furnace were calculated assuming that all fission products which were not accounted for (refer to accountabilities, Table III) remained in the fuel or furnace. All figures were corrected for radioactive decay to the time of meltdown.

These figures show that iodine, tellurium, cesium, and molybdenum are released in significant quantities to the containment vessel. Generally, the fission products, other than iodine and cesium, were distributed approximately equally between the condensate and the containment walls. Larger fractions of the iodine and cesium were found in the condensate than on the walls, possibly because of the greater solubility of these fission products in the condensate. The total surface area of all the painted coupons exposed in Run 4 was approximately 1/30 of the surface area of the stainless steel containment vessel wall; however, the painted coupons absorbed approximately 1/9 as much total iodine as the walls, indicating the high affinity of paint for iodine. Since the interior surfaces of commercial power reactor containment buildings are covered with

TABLE IX

DISTRIBUTION OF FISSION PRODUCTS FOLLOWING TYPICAL CDE MELTDOWN

Fission Product	Total Fuel Inventory (curies)	Percent Activity Deposited ^[a]				
		In Fuel and Furnace	In Transport Line	In Containment Condensate	On Containment Walls	On Painted Coupons
Sr-89	18.0	> 99.9	0.01	0.01	0.01	
Zr/Nb-95	21.9	> 99.9	Trace	Trace	Trace	Trace
Mo-99	90.0	> 93.4	Trace	3.6	2.9	0.05
Ru-103	13.8	> 99.7	0.01	0.09	0.13	Trace
I-131	38.8	75.4	4.8	13.2	4.6	0.5
Te-132	69.0	84.2	3.3	4.3	8.0	0.2
Cs-137	0.1	> 92.2	0.6	5.4	1.7	0.05
Ba-140	65.9	> 99.8	0.02	0.06	0.07	Trace
Ce-141	34.4	> 99.9	Trace			

[a] Activities present in quantities less than 0.01% were considered trace amounts. Blank spaces indicate none detected.

[b] ~1.5% of I-131 remained in containment atmosphere.

paint rather than stainless steel, the distribution of iodine should be more evenly divided between the painted surfaces and the condensate than that indicated in the CDE runs.

3. FISSION PRODUCT BEHAVIOR IN CONTAINMENT VESSEL

The fission products transported to the containment vessel are removed from the atmosphere by fallout, deposition, and washout, and the importance of each removal process depends upon the character of the fission product aerosol. In defining their behavior, fission products can be separated into three groups -- noble gases, halogens, and particulates. The noble gases, typified by Xe-133 and Kr-85, are chemically inactive and are removed, to a significant extent, only through radioactive decay and leakage from the vessel. The halogens, typified by I-131, generally exist in the chemically reactive forms, I₂ and HI,

which are highly soluble in the condensate and adsorb readily on particulates and organic materials. Removal of the chemically active forms of iodine from the containment atmosphere is quite rapid. Recently, interest^[27] has focused on the organic compounds of iodine. The organic compounds are not as reactive chemically as elemental iodine and are not removed as easily by deposition, washout, or by adsorption on charcoal which is used in atmosphere cleanup systems. Except for the noble gases and halogens, fission products reaching the containment vessel are in the form of particulates. These fall out (if large enough), deposit on surfaces, or act as nuclei for condensation. Fission product behavior in the containment vessel is being studied at other sites^[28,29,31], but generally at trace levels of activity or with fission product simulants. The runs in the CDE provided information on fission product behavior at higher activity levels and release conditions which are representative of actual accident conditions.

3.1 Character of the CDE Aerosol

The fission products in the CDE atmosphere were identified by analyses of samples taken with the Maypack samplers and Continuous Sampler-Monitor (described in Section III). Numerous fission products were detected (Table X), but the fission products contributing the major amount of activity in the atmospheric samples were I-131, Mo-Tc-99, Te-I-132, Xe-133, and Xe-135. The activities of the more volatile fission products with short half-lives masked the minor ones during early counting; consequently, the samples were counted twice -- within a week after sampling, and 30 to 60 days after sampling, when the activities of the major short half-life nuclides had decayed to a level low enough to show the peaks of the minor long half-life nuclides in the gamma-ray spectra.

Particulate Concentration, Size, and Composition: To obtain information on the particulates in the CDE aerosol, samples of the containment atmosphere were analyzed to determine particle composition, particle concentration, and particle size during meltdowns of unirradiated fuel pins prior to Runs 3 and 4. The aerosol sample from the containment vessel was passed through 3 ft of heated tubing to a membrane filter. The composition of the material collected on the filter was determined by X-ray fluorescence and emission spectrography. Similar information was obtained during Runs 3 and 4 by analyzing the filter elements from Maypack sampling units. The results are summarized in Table XI. In all cases, tin was the major constituent of the particulate matter.

Several filters used to collect particulates in the two "cold" tests were examined by electron microscopy. Particles on the Run 4 set (Millipore Type AA^[a]) were transferred to electron microscopy grids by dissolving the filter. Stringy material, believed to be filter residue, characterized the pictures. The Test 5 filters (Millipore Type VM^[b]) were photographed at magnifications of 6800 to 165,000 by the Analytical Chemistry Division of ORNL where the particles from randomly selected areas of each filter were transferred to electron microscope grids by a carbon vapor deposition and membrane dissolution technique.

[a] 0.80 μ pore-size membrane filter supplied by Millipore Filter Corp, Bedford, Mass.

[b] 0.05 μ pore-size membrane filter supplied by Millipore Filter Corp, Bedford, Mass.

TABLE X

FISSION PRODUCTS DETECTED IN THE CDE CONTAINMENT VESSEL ATMOSPHERE

Fission Product	Half-Life		Relative Abundance
Zr-95	65	d	minor ^[a]
Nb-95	35	d	minor
Mo-99	67	h	major ^[b]
Tc-99	6	h	major
Ru-103	41	d	minor
Te-129	33	d	minor
I-131	8	d	major
I-132	2.3	h	major
Te-132	77.7	h	major
I-133	20.8	h	major
Xe-133	2.3	d	major
Xe-135	12	d	major
Cs-137	30	y	minor
Ba-140	12.8	d	minor
La-140	40.2	h	minor

[a] < 10% of fuel inventory.

[b] > 10% of fuel inventory.

Typical photomicrographs are shown in Figures 31 through 33. The particle diameters ranged from 0.001 to 7.5 μ . There were few particles larger than 1 μ , and a plot of particle number versus size indicates that the maximum size of the statistically important particles is closer to 0.7 μ . The particles larger than 0.7 μ may be dust or other foreign material originally in the containment atmosphere.

All samples, including those taken immediately after vessel isolation, show evidence of particle agglomeration. Two general types of particles were present--agglomerates of small particles, and larger spherical particles. The latter particles were similar to those reported in characterizations of UO_2 - Na_2O aerosols^[32,33] where the sodium oxide formed large spheres to which chain-like agglomerates of the finer UO_2 particles clung. In the CDE runs, these spheres may be created by vaporized tin, the major component of the aerosol.

TABLE XI

CONCENTRATION AND COMPOSITION OF PARTICULATES IN CDE ATMOSPHERE

	Test ^[a]			
	"Cold" CDE-4	"Hot" CDE-4	"Cold" CDE-5	"Cold" CDE-5
Time Sample Taken ^[b] , min	15	15	5	65
Concentration, $\mu\text{g}/\text{ft}^3$ ^[c] :				
Tin	690	28 ^[d]	80	40
Uranium	570	2.3	<10	<10
Silicon ^[f]	11	[e]	< 0.1	< 0.1
Zirconium	1	[e]	< 0.01	< 0.01

[a] "Cold" refers to nonradioactive meltdown made prior to radioactive or "hot" run.

[b] Time after melting ceases.

[c] Single samples unless noted otherwise.

[d] Average of four samples.

[e] Not determined.

[f] From quartz furnace.

Because it is difficult to establish the size of the agglomerates, no quantitative measure was obtained of the growth of the agglomerates in the 1-hr period following isolation of the containment vessel.

LOFT atmosphere samplers were tested extensively in CDE Runs 3 and 4. In these runs, the atmosphere was sampled 30 min and 24 hr following meltdown. The nearly identical depletion (approximately 99%) of the fission products, other than the iodine and noble gases, over this period indicated that they were present in the same form, probably particulates (see Table XVI). The iodine persisted in one or more gaseous species, as evidenced by the lower depletion ratio for I-131.

Hydrocarbon Content of Containment Atmosphere: Organic iodides, even if a small fraction of the iodine present in a containment vessel, are important because their chemical stability and low solubility in the condensate result in low removal rates from the building atmosphere. To obtain a measure of the

Fig. 31 Photomicrograph of particles in containment atmosphere immediately following vessel isolation (37,500X) - Run 5.

Fig. 32 Photomicrograph of particles in containment atmosphere 1 hr after vessel isolation (37,500X) - Run 5.

Fig. 33 Photomicrograph of particles in containment atmosphere immediately following vessel isolation (165,000X) - Run 5.

hydrocarbons available to form organic iodides in the CDE, the hydrocarbon content of the containment vessel atmosphere was determined in "cold" runs prior to Runs 3 and 4. The instrument used was a Perkin-Elmer Model 213 Hydrocarbon Detector, which operates on the flame ionization principle. Samples were taken through an outlet line on the side of the containment vessel before the fuel melting and after the vessel cooled for about 2 hr. The results, presented in Table XII, show that a significant amount (an increase of approximately 50%) of hydrocarbon was added to the vessel during the fuel melting.

TABLE XII

HYDROCARBON CONTENT OF CONTAINMENT VESSEL ATMOSPHERE

	Cell Atmosphere [a]	Containment Before Meltdown [a]	Containment After Meltdown [a]	Total Hydrocarbon Released During Meltdown, mg
Run 3	3.2	4.8	7.0	3
Run 4	3.8	19.0	29.0	15

[a] In units of ppm, methane equivalent.

Iodine Species: The iodine in a containment building atmosphere following a loss-of-coolant accident is generally classed as one of three forms -- inorganic volatile species (I_2 , HOI, HI), iodine adsorbed on particulates, and organic iodides. Considerable information on the iodine species was obtained in Runs 3 and 4, in which testing the LOFT particulate-iodine capsule sampler was a primary task. Six Maypack-type cartridges were placed in the containment vessel for each run. Four were suspended independently in the vessel, and two were placed in the spherical housing used for the LOFT capsule samplers. The components of the Maypacks and their intended functions are summarized in Table XIII. In the independently housed Maypacks, an aluminum dummy inlet preceded the particulate filter, and an aluminum spacer was placed between the silver membrane and the charcoal beds. The dummy inlet provided an estimate of the plateout of fission products on the inlet to the sampling cartridge, and the aluminum spacer replaced an unused component.

The distribution of iodine between the various species as indicated by the activity distribution in the cartridges is given in Table XIV. The values for particulate iodine were obtained by subtracting from the activity on the first particulate filter, a correction for the vapor phase inorganic iodine adsorbed on the first filter. This correction was estimated from the vapor phase inorganic iodine found on the second and third particulate filters. The values for the inorganic iodine obtained from the silver membranes were corrected by adding to them the iodine found on the dummy inlet, the inlet section, and the calculated vapor phase inorganic iodine on the particulate filters. The activity found on the charcoal beds was assumed to represent the organic iodides.

TABLE XIII.

MAYPACK COMPONENTS AND THEIR FUNCTIONS

Adsorber	Intended Function	Type	No. Units
Particulate Filter	Removal of particulate fission product	Flanders F-700 ^[a] (glass fiber)	3
Silver Membrane	Adsorption of inorganic iodine species (HI, I ₂)	Selas Flotronics ^[b] , 5-μ pore size	3
Charcoal Bed	Adsorption of organic iodides	3/4-in beds of ^[c] Pittsburg PCB activated coconut charcoal impregnated with 2% KI	2
Absolute Filter	Retention of charcoal dust	Flanders F-700 (glass fiber)	1

[a] Product of Flanders Filters, Inc., Riverhead, N. Y.

[b] Product of Selas Corp. of America, Dresher, Pa.

[c] Product of Pittsburg Activated Carbon Co., Pittsburg, Pa.

In Run 3, some of the UO₂ powder (particle diam of 25 to 200 μ) on which the fuel pin rests during melting, was inadvertently blown into the containment vessel prior to the meltdown. This probably explains why a large fraction of particulate iodine was found 30 min after meltdown in Run 3, but not in Run 4. The indicated amounts of organic iodides in the vessel atmosphere represent 1.7 and 2.4% of the total iodine inventory in the fuel for Runs 3 and 4, respectively.

The fraction of airborne iodine consisting of organic iodides, as obtained from the Maypack data, is high, even at 30 min past meltdown. Subsequent laboratory tests^[34] of Maypack components at the ICPP indicated that most of the iodine passing the silver membrane and appearing on the charcoal bed was hypoiodous acid (HOI), an iodine species created by the hydrolysis of iodine at temperatures above 90°C and at pH levels above 8. This was substantiated in the tracer runs following Run 5, in which the species injected was elemental iodine tagged with I-131. During the cooling period following injection of the tagged iodine, samples of the atmosphere were taken with adsorber packs and in gas bombs. Analysis of the gas bomb contents by vapor phase chromatography showed organic iodide concentrations of 0.2 to 1.0% of the injected iodine, which are well below that indicated by the absorber packs, which are similar to the Maypack samplers used in the CDE Runs 3 and 4.

TABLE XIV

DISTRIBUTION OF IODINE SPECIES IN THE CDE ATMOSPHERE

Iodine Form	30-min Past Meltdown		24-hr Past Meltdown	
	$\mu\text{g}/\text{ft}^3$	% at Time of Sampling	$\mu\text{g}/\text{ft}^3$	% at Time of Sampling
<u>Run 3</u>				
Particulate	0.12	48	0.0025	7.5
Inorganic	0.019	7	0.0025	7.5
Organic	0.11	45	0.029	85
Total - Run 3	0.25	100	0.034	100
<u>Run 4</u>				
Particulate	0.0054	3	0.001	3
Inorganic	0.049	26	0.0012	3
Organic	0.135	71	0.036	94
Total - Run 4	0.19	100	0.038	100

The existence of hypoiodous acid as one of the iodine forms following an accident may explain the large percentages (42 to 99.8%) of organic iodides reported[35] to be in the containment building following the PRTR accident at Hanford, September 29, 1965. Sampling for iodine was performed using sampling cartridges with a conventional arrangement of internals -- membrane filter, silver screen, charcoal filter, and charcoal bed. About 30% of the iodine passing the silver screens and believed to be organic iodides was found on the charcoal filter, which ordinarily will not collect much organic iodide. Such a distribution would be expected if iodine passing the silver screen were present as hypoiodous acid.

3.2 Removal of Fission Products from the Containment Atmosphere

Fission products released to a containment building atmosphere are removed by several natural processes -- radioactive decay, washout in the condensate,

deposition on the walls, and fallout as particulates. Information pertaining to these processes was obtained by analysis of the atmosphere samples and by analysis of the condensate removed from the containment vessel. Data obtained in CDE runs on the various types of fission products are summarized in the following sections according to fission product classification.

Removal of Noble Gases: The samples of the containment atmosphere taken after meltdown and analyzed for the krypton and xenon content included sampling cylinders containing raw gas, sampling cylinders of the gas stream from the Continuous Sampler-Monitor, and the void space in the LOFT gas-particulate capsule sampler. Results from the different samplers varied, and, generally, the amounts of krypton and xenon indicated by the analyses represented only a small fraction of the total krypton and xenon which should have been released from the fuel. Therefore, following Run 5, two tracer runs were made with Xe-133 to resolve possible sampling problems. In these runs, the Xe-133 was injected into the CDE containment vessel after it had been pressurized to 22 psig with a steam-air mixture. The temperature and pressure were then permitted to decay, as in the meltdown runs.

Three independent sampling systems -- a raw gas, a filtered gas, and the CSM -- were operated during each tracer run. Samples taken with the filtered gas system passed successively through a heated line, glass fiber filter, silver membrane, and a condenser, before being collected in a sample bomb. The CSM samples were collected by bypassing the gas stream from the scrubber through a sample bomb.

Average Xe-133 concentrations determined with the three independent sampling systems checked closely -- within 4% as shown in Table XV. The concentration (on a dry basis) during the cooling periods following injection of the Xe-133 remained nearly constant showing that condensation or deposition removes very little of the noble gases from the containment vessel. Approximately 0.005% of the Xe-133 appeared in the condensate -- about that indicated by solubility data for xenon in water.

Removal of Particulates: As described in Section 3.1, particle size and composition data were obtained in "cold" runs prior to the CDE Run 5. The concentration of tin, the major constituent in the aerosol, showed a decrease from 80 to 40 $\mu\text{g}/\text{ft}^3$ during the period from 5 to 65 min after isolation of the containment vessel. The extent of particulate removal was also determined from Maypack data in Runs 3 and 4. The concentrations of the fission products present as particulates were determined 30 min and 24 hr following meltdown. These data are presented in Table XVI as ratios of the concentrations at the sampling time to those at time zero. The rate of removal of particulates from the atmosphere is comparable to that of the iodine over the first 1/2 hr. After that, the particulate content dropped more rapidly than the iodine content -- resulting in 99.5+% removal in 24 hr of the particulate activities entering the containment vessel.

Further evidence of the rapid removal of particulates by the condensate early in the run is shown in Figure 34, where the percentage of each fission product removed by the condensate (based on the total containment vessel inventory) is plotted versus time after meltdown. During the first 1/2 hr, 30 to 65% of all the particulates removed were collected in the condensate.

TABLE XV

CONCENTRATION^[a] OF Xe-133 IN CDE CONTAINMENT ATMOSPHERE

CDE TRACER TEST 2

Time After Injection	CMS Gas ^[d]		CSM Liquid		CSM Gas + Liq		Filtered Gas ^[d]		Raw Gas ^[d]	
	VPC	GRS	GRS				VPC	GRS	VPC	GRS
10 min	15, 17 ^[b,e]	9 ^[b]	3	--			61	53	66	56
30 min	[c]	[c]	[c]	[c]			63	56	--	--
40 min	69	50	5	55			64	57	--	--
1 hr	65	52	4	56			64	52	--	--
2 hr	62	49	5	54			--	52	--	--
3 hr	--	50	5	55			63	54	--	--
4 hr	52	50	5	55			--	50	60, 58 ^[e]	52
6 hr	64	55	4	59			61, 63 ^[e]	52	--	--
7 hr	63	50	4	54			66	50	61, 63 ^[e]	53
8 hr	60	48	3	51			58	52	--	--
Average	62	50.5	4.1	55			63	53	62	54
Standard Deviation	5	2.1	0.7	2.2			2.4	2.2	3.5	2.4

[a] μCi of Xe-133 per standard liter of air (calculated concentration at zero time = 79 $\mu\text{Ci/liter}$).

[b] Not included in average.

[c] Bad sample -- flow recorder chart shows that the gas flow was stopped while trying to take this sample.

[d] VPC -- separation by vapor phase chromatography and analysis by gamma-ray spectrometry.

GRS -- direct analysis of sample by gamma-ray spectrometry.

[e] Duplicate analyses of same sample.

TABLE XVI

FISSION PRODUCT CONCENTRATIONS IN CONTAINMENT VESSEL

Nuclide	Concentration C_0 ^[a] at T_0 (mCi/ft ³)	Normalized Concentration C/C_0		% Depletion Between 0.5 and 24 hr
		0.5 hr after T_0	24 hr after T_0	
<u>CDE Run 3</u>				
Mo-99	80	0.480	0.00146	99.7
Ru-130	1.48	0.338	0.00157	99.5
I-131	79	0.236	0.0334	86.4
Te-132	118	0.543	0.00197	99.6
Cs-137	0.402	0.224	0.000874	99.6
Ba-140	0.37	0.316	0.00158	99.5
<u>CDE Run 4</u>				
Mo-99	70	0.174	0.00160	99.1
Ru-103	0.37	0.135	0.00135	99.0
I-131	85	0.164	0.033	80.0
Te-132	146	0.094	0.0007	99.3
Cs-137	0.105	0.189	0.00221	98.8
Ba-140	1.06	0.0434	0.000175	99.7

[a] Based on the total amount of the various nuclides found in the containment vessel.

Fig. 34 Time-dependent removal of fission products by condensate.

Cesium, the most soluble fission product, showed the largest percent removed by the condensate. Similar behavior was observed at BNWL^[36] with simulated fission product cesium.

A summary of the percentages of fission products collected in the condensate and the amount of condensate formed in each run is given in Table XVII. This tabulation shows a definite trend toward the removal of a larger percentage of the fission products with an increasing amount of condensate formed. The total condensate collected for each run also furnishes a rough measure of the condensation rate on the walls of the containment vessel. During the fuel melting periods, wall temperatures were adjusted so the vessel would cool to follow a predetermined temperature decay plot after the vessel was isolated from the furnace. The vessel walls were cooler in those runs in which more condensate was formed (more steam was added to hold a pressure of 22 psig until completion of the melting period).

Removal of Iodine: The removal of various iodine forms (inorganic, iodine sorbed on particulates, and organic) from a steam-air atmosphere was evaluated in a number of high- and tracer-level CDE runs. The iodine forms, depending on their physical characteristics, were removed to varying extents from the atmosphere. Although development of the samplers has not progressed

TABLE XVII

REMOVAL OF FISSION PRODUCTS FROM CONTAINMENT VESSEL BY STEAM CONDENSATE

Run No.	1	2	3	4	5
Total Condensate Collected, cc	2500	5525	9820	20,870	14,390
Fission Product	% in Condensate ^[a]				
Mo-99	16	65	52	54	[b]
I-131	24	24	54	72	67
Te-132	6	8	30	52	35
Cs-137	56	58	65	75	51
Ba-140	[c]	30	4	45	64
Ru-103	[d]	[d]	10	39	57

[a] Determined by dividing amount of fission product in condensate by total found in containment vessel.

[b] Below detectable limits.

[c] None detected.

[d] Not determined.

sufficiently to completely describe the removal processes, some general conclusions can be drawn from the CDE data.

A compilation of data on the time-dependent total airborne concentration of iodine in the saturated steam-air environment of the CDE runs is shown in Figure 35. These data were obtained in both high-level meltdown and tracer-level runs using "Maypack" type samplers and the Continuous Sampler-Monitor. The total iodine behavior in the containment atmosphere was characterized by a rapid removal during the first 1/2 hr after iodine injection (airborne half-time = 8 min), followed by a period of much slower removal (airborne half-time = 7 hr). The initial rate of iodine removal closely followed the depletion of steam from the vessel, indicating that the initial removal process is dependent primarily on the condensation rate or heat flux through the wall, as suggested by other investigators^[3,30].

Fig. 35 Time-dependent I-131 concentration in containment atmosphere.

The total iodine removal rate was comparable to that of the particulate fission products during the early stages of the washout period (see page 53). In the later stages of the washout period, the airborne iodine concentration would be determined by an equilibrium between the iodine in the condensate remaining in the tank, the iodine deposited on the walls, and the iodine remaining in the atmosphere. Over the 24-hr cooling periods of the CDE runs, the decontamination factors achieved by natural washout were approximately 25 times greater for particulates than for the total iodine content.

The indicated concentrations of organic iodides in the containment atmosphere obtained in Runs 3 and 4 and in the I-131 tracer runs following Run 5 are plotted in Figure 36. The concentrations indicated in Runs 3 and 4 are considerably higher than those indicated in the tracer runs, probably as the figures for Runs 3 and 4 include another specie, hypiodous acid, as explained in Section 3.1. The data from the tracer runs were obtained by separating the organic iodides from a gas sample with a vapor-phase chromatograph followed by gamma-ray counting. This technique produced more accurate results than did the Maypack cartridge used in Runs 3 and 4, as it isolates the organic iodides from the hypiodous acid. The concentration of organic iodides, as indicated in the tracer runs, remained constant throughout the cooling period at about 1/4 to 1% of the iodine injected.

Fig. 36 Time-dependent organic iodide concentration in containment atmosphere.

3.3 Fission Product Deposition

Deposition on the surfaces of a containment building is one of the principal mechanisms for removal of fission products, other than noble gases, from the containment atmosphere. The deposition rate and total quantity of fission products deposited under anticipated reactor accident conditions were affected by a number of variables including (a) the concentration gradient from the atmosphere to the surface, (b) the temperature of the surface on which deposition occurred, (c) the rate and type (film or drop-wise) of the condensation which occurred on the surface, (d) the composition (paint, metal, etc) of the surface, and any chemical reactions between the surface and the depositing fission products, (e) the physical conditions of the surface (thickness of the corrosion film, surface roughness, discontinuities, stress, etc), (f) the species of the fission products being deposited, (g) the orientation of the surface, and (h) the presence of surface contaminants (grease, decontamination solutions, etc). Experiments have been conducted and are continuing to determine the relative importance of these variables[5,9,28,29,31,37]. However, a satisfactory model incorporating all the significant variables has not yet been developed[24].

As part of the program of determining fission product behavior during the LOFT, a sampler was developed for obtaining time- and space-dependent data on fission product deposition. One objective of testing in the CDE was to determine if data obtained from LOFT prototype plateout samplers accurately

described the deposition occurring on the containment surfaces. To accomplish this, the effects of numerous variables (fission product species, orientation of surface, material, temperature, condensation) affecting fission product deposition were studied using LOFT prototype samplers and several "custom made" deposition samplers. The results of this testing, as they pertain to fission product deposition, are summarized in the following paragraphs.

Normal Variation of Deposition on Painted Surfaces: To determine the relative effect of variables on fission product deposition, the normal variation in the deposition data resulting from sampling, handling, and analysis must be known. To determine the normal variation for typical deposition samples, the data from 50 painted coupons (1/2-in.-diam cylinders and 3/4-in.-diam disks) were examined. These coupons were contaminated under comparable conditions in the CDE and counted (the entire contaminated surface was counted) using a NaI(Tl) detector. Data from these coupons -- the coupons from the LOFT plateout sampler, the deposition tube sampler, and the wall plateout sampler -- showed mean deviations of ± 12 and $\pm 18\%$ for the I-131 and particulate fission products (Mo-99, Te-132, Cs-137, Te-129, and Ru-103), respectively. The comparable maximum deviations (highest to lowest values) for both I-131 and particulates were about 200%.

An additional measure of the normal variation of deposition on painted surfaces was obtained during Run 5. Two 12- by 12-in.-painted panels, contaminated in the containment vessel, were gamma scanned for I-131 at different locations using a NaI detector and a 1/2-in.-diam collimator hole. The mean deviation of the I-131 analyses obtained in this manner was $\pm 10\%$ -- reasonably close to the variation observed when counting the small plateout coupons from the LOFT capsule.

The effect of scanning still smaller areas was examined in both Runs 4 and 5, using coupons from the deposition tube sampler (Figure 18). For each run, two coupons were taken from a tube with its surface maintained at steam condensing conditions, approximately 70°F below the containment atmosphere temperature; another set of coupons was taken from the uncooled tube which stayed at the temperature of the containment atmosphere. Each sample was gamma-scanned with a Ge(Li) detector at four locations spaced evenly around the circumference of the cylindrical coupon. A lead collimator, with a slit width of 1/16 in. by 1/2 in., was used to isolate small areas of the coupons during the scanning.

Iodine was the principal activity found on all of the samples. The large quantities of I-131 deposited on the atmospheric temperature samples during Run 4, masked the presence of other fission products present on the samples. The samples for Run 5 were allowed to cool for 50 days before scanning, to obtain data on nuclides present in smaller activities; consequently, the data for Mo-99 and Te-132 were not obtained, as their activities had decayed below detection limits. The relative distribution of each nuclide on each of the samples is given in Table XVIII. The maximum variance in the relative distribution of I-131 was about 200%, while the measured particulate activities showed a maximum variation of about 500%. The corresponding mean deviations were ± 13 and $\pm 30\%$. These data indicate that iodine deposition is more uniform than that of the other fission products, which is to be expected, since the iodine is present mainly as a gas in the CDE atmosphere.

TABLE XVIII

DISTRIBUTION OF FISSION PRODUCTS ON AN AMERCOAT 66^[d] PAINTED VERTICAL DEPOSITION TUBE

Sample	Position ^[a]	Sample Temperature	Relative Distribution Ratios						
			CDE RUN 4			CDE RUN 5			
			<u>I-131</u>	<u>Mo-99</u>	<u>Te-132</u>	<u>I-131</u>	<u>Cs-137</u>	<u>Te-129</u>	<u>Ru-103</u>
1	A	Atmospheric	1.00 ^[b]			0.97	1.00	[c]	[c]
	B	Atmospheric	0.75			0.93	0.82		
	C	Atmospheric	0.57			1.00	0.92		
	D	Atmospheric	0.71			0.85	1.00		
<hr/>									
2	A	Atmospheric	1.00 ^[b]			0.96	0.67	0.72	[c]
	B	Atmospheric	0.80			0.92	0.34	0.71	
	C	Atmospheric	0.75			1.00	0.75	1.00	
	D	Atmospheric	0.76			0.90	1.00	0.71	
<hr/>									
3	A	Cooled	1.00 ^[b]	1.00	1.00	0.99	0.95	0.70	[c]
	B	Cooled	0.58	0.20	0.23	1.00	0.84	0.81	
	C	Cooled	0.89	0.78	0.61	0.75	0.73	0.62	
	D	Cooled	0.77	0.29	0.61	0.94	1.00	1.00	
<hr/>									
4	A	Cooled	1.00 ^[b]	1.00	1.00	0.69	0.52	0.51	0.97
	B	Cooled	0.93	0.60	0.52	1.00	1.00	1.00	0.75
	C	Cooled	0.83	0.34	0.21	0.70	0.47	0.35	0.58
	D	Cooled	0.96	0.75	0.59	0.82	0.50	0.63	1.00

[a]

[b] Position with highest activity on each sample has been assigned a value of 1.00.

[c] Not reported.

[d] Product of Amercoat Corporation, Brea, Calif.

Effect of Surface Orientation on Fission Product Deposition: To determine the effect of sample surface orientation on fission product deposition, numerous painted coupons were oriented in various directions (facing up, down, or sideways) and attached to three (top, middle, or bottom) levels of the capsule support rack in the CDE containment vessel (Figure 7). The major isotopes found on each coupon contaminated in CDE Run 4, with the corresponding orientation of the coupon, are listed in Table XIX. From these data, it appears that only coupon surfaces facing up show any substantial deviation from the average deposition found on all surfaces. The coupons facing up showed deposition levels of iodine two times higher than the other coupons. For particulates (characteristic of Te-132, Mo-99, Ru-103, Te-129, Cs-137, and Ba-140), the deposition levels for coupons facing up were up to 23 times higher than on coupons oriented differently. The increased deposition on the horizontal surface facing up was probably due to the settling of particles on the surfaces. Similar results were obtained by Parsly and Row during tests in the NSPP^[38] in which stainless-clad UO₂ fuel containing fission product simulants were melted. Parker et al reported similar results for UO₂ fuel meltdowns^[39].

The effect of surface orientation on deposition was also investigated, during Run 3, with coupons on the controlled temperature sampler; however, these coupons were rinsed with water prior to removal from the containment vessel, thus removing most loosely adhering particles. The coupons facing up showed lower deposition levels for iodine than the other coupons. For particulates Te-132, Mo-99, and Cs-137, the deposition levels for coupons facing up were only up to 10 times higher than the other orientations -- indicating that much of the material settling on the surface facing up was not tightly fixed to the surface, and consequently, was easily removed.

Effect of Temperature and Condensation on Fission Product Deposition: Most of the deposition coupons contaminated in the containment vessel for the sampler development and coating evaluation studies were located away from the containment wall where their temperatures were near the atmosphere temperature, and condensation was minimized. To obtain some measure of the effect of lower temperatures and condensation, such as exists at the vessel wall, provisions were made in the first three CDE runs to control the temperatures of two groups of plateout coupons. Sample coupons of 304 stainless steel, carbon steel, and carbon steel coated with a four-coat epoxy paint (Amercoat 66^[a]) were placed on the controlled temperature samplers (Figure 16) and contaminated in the CDE containment vessel. Following contamination, and prior to removal of the samplers from the containment vessel, the coupons were rinsed with water to remove particles loosely adhering to the surfaces. The temperature of one group of coupons was maintained above the containment vessel temperature, and the other group was maintained below the containment temperature. The procedure used to contaminate the coupons was similar for all runs, except that the difference in temperature of the containment atmosphere and the coupons was decreased in succeeding runs.

A comparison of contamination levels on the surfaces having a relatively large temperature difference between the coupon surface and the atmosphere is given by data from Run 2 (Figure 37). The data indicate that the deposition of

[a] Product of Amercoat Corporation, Brea, Calif.

TABLE XIX

EFFECT OF SURFACE ORIENTATION ON FISSION PRODUCT DEPOSITION^[a]

(CDE Run 4)

Coupon Location In Vessel	Orientation	Coupon Activity, dps/cm ² [f]						
		I-131	Te-132	Mo-99	Ru-103	Te-129	Cs-137	Ba-140
Top Level	Facing Up	9.9 x 10 ⁶	4.32 x 10 ⁶	1.95 x 10 ⁶	1.36 x 10 ⁴	4.60 x 10 ⁴	6.57 x 10 ³	1.27 x 10 ⁴
Top Level	Facing Down	4.14 x 10 ⁶	3.27 x 10 ⁵	1.49 x 10 ⁵	1.11 x 10 ³	2.87 x 10 ³	6.04 x 10 ²	2.34 x 10 ³
Top Level	Facing to Side ^[c]	3.90 x 10 ⁶	2.04 x 10 ⁵	1.12 x 10 ⁵	8.23 x 10 ²	2.80 x 10 ³	4.52 x 10 ²	1.97 x 10 ³ ^[d]
Middle Level	Facing Up ^[b]							
Middle Level	Facing Down	4.25 x 10 ⁶	2.18 x 10 ⁵	1.27 x 10 ⁵	8.07 x 10 ²	2.18 x 10 ³	4.43 x 10 ²	[e]
Middle Level	Facing to Side ^[c]	3.40 x 10 ⁶	1.67 x 10 ⁵	1.22 x 10 ⁵	5.64 x 10 ³	1.90 x 10 ³	3.62 x 10 ²	[e]
Bottom Level	Facing Up	9.76 x 10 ⁶	3.69 x 10 ⁶	1.34 x 10 ⁶	1.54 x 10 ⁴	3.60 x 10 ⁴	5.38 x 10 ³	5.06 x 10 ³
Bottom Level	Facing Down	3.35 x 10 ⁶	2.73 x 10 ⁵	1.51 x 10 ⁵	6.70 x 10 ²	1.72 x 10 ³	3.71 x 10 ²	[e]
Bottom Level	Facing to Side ^[c]	4.30 x 10 ⁶	3.79 x 10 ⁵	1.72 x 10 ⁵	1.23 x 10 ³	4.31 x 10 ³	7.54 x 10 ²	4.35 x 10 ³

[a] 3/4-in-diam aluminum coupons painted with Amercoat-66.

[b] Coupon found in downward position after test -- may have slipped on support wire during installation or testing.

[c] Average of four to six coupons located in a vertical position. The coupons were either facing toward or away from the CDE containment vessel wall at distances of 10 and 17 in. from the wall.

[d] Only one value reported.

[e] Amount present was below detectable limits of instrument.

[f] Average of two coupons except where noted -- all activities are corrected for decay to time of meltdown.

Fig. 37 Comparison of contamination levels on stainless steel, carbon steel, and a painted surface with large temperature differences between surface and atmosphere.

fission products (except for the radioiodine on paint) on the colder surfaces with steam condensation is greater (average of 28 times greater) than on warmer surfaces without condensation. A similar, but less pronounced, pattern (average of 2 times greater) was observed at the smaller temperature differences of Run 3 (Figure 38).

The deposition of iodine on paint deviated from this pattern, particularly in Run 2. The iodine deposition on Amercoat 66 was greater on the coupons without condensation (heated) than on coupons with condensation (cooled). The difference in iodine behavior was probably due to an increased penetration of iodine into the organic coating at higher temperatures, since the diffusion coefficient for iodine penetration into an epoxy coating (Amercoat 66) increases rapidly above approximately 194°F [9], and the temperatures of the heated coupons rose above 194°F in Run 2. This explanation is substantiated by the observation that iodine deposition behavior of carbon steel and stainless steel followed the general behavior of the other nuclides.

The results obtained with the controlled temperature sampler (comparable results were obtained with the deposition tube samplers) appear to contradict the observation (discussed in Section 3.2) that greater condensation in the vessel reduces the amount of activity deposited on surfaces. This apparent anomaly can be explained by assuming, logically, that deposition is a function of both time and concentration. A high condensation rate in the containment vessel depletes the atmosphere more rapidly than a low condensation rate, thus the integrated effect of concentration and time exposure of the surface to the fission products is reduced. In the case of small cooled surfaces in the vessel, such as is represented by the deposition samplers, the condensation occurring on the small surface does not grossly alter the rate of fission product depletion in the vessel. The condensation, however, does increase the concentration of fission products in the vicinity of the sampler, probably through bulk flow of steam toward the cold sample surface; thus, the cooled surface will show a higher deposition than the warmer surface. This interpretation implies that, in an accident situation, the total activity deposited on surfaces decreases as the amount of steam condensed in the building increases. However, the colder surfaces in the building will have more deposited activity than the warmer surfaces, assuming the temperatures are below the softening temperatures of the organic protective coatings.

Effect of Condensate Flow on Fission Product Deposition: Autoradiographs of painted panels contaminated in the containment vessel show that condensate flowing over a surface can redistribute the deposited fission products. Several 12- by 12-in.-painted panels were contaminated and then autoradiographed using industrial X-ray film. An autoradiograph taken of one of the painted panels seven days after the run is shown in Figure 39. The numerous hot spots evident on the radiograph are believed to have been left by condensate drops which had dried on the panel.

Because of the interest in the variation of deposition caused by condensate flowing from top to bottom of the LOFT containment building walls, the effect of vertical length was examined in Runs 4 and 5, using the cooled deposition tube sampler (Figure 18) and in two I-131 trace-level runs, using the cooled 12-in.-wide by 48-in.-long cooled panel (Figure 17). The deposition data (Table XX) obtained with these samplers showed considerable variation, but no pattern

CDE RUN 3

Fig. 38 Comparison of contamination levels on stainless steel, carbon steel, and a painted surface with small temperature differences between surface and atmosphere.

Fig. 39 Autoradiograph of fission product deposition on a painted panel.

related to height can be discerned. Apparently, the effects of other variables, eg, slight (5°F) temperature variations along the surfaces, proximity to transport gas inlet, and possible stratification of fission products in the vessel, are sufficient to mask any effect of the condensate flowing over the 4-ft-long surfaces. A deposition surface longer than the 4-ft panel used in the CDE is desirable for examining this variable.

Fission Product Deposition on Various Materials: To find a material of construction with low fission product retention for the Continuous Sampler-Monitor, 18 coupons of uniform dimensions and fabricated of six materials were contaminated in the CDE. The materials tested were Type 304 stainless

TABLE XX

EFFECT OF CONDENSATE WASHING ON DEPOSITION OF FISSION PRODUCTS ON VERTICAL SURFACES

Run	Position of Coupon on Surface	Deposited Activity ^[a,b]							
		I-131	Te-129	Te-132	Mo-99	Ba-140	Ru-103	Cs-137	Zr-95
CDE 4	Top	1.10	[b]	2.00	1.27	1.58	1.17	1.67	1.60
	Middle	1.00		1.23	1.30	1.13	1.00	1.12	1.54
	Bottom	1.36		1.00	1.00	1.00	1.01	1.00	1.00
CDE 5	Top	1.00	1.07	1.06	[b]	[b]	1.09	1.21	[b]
	Middle	1.13	1.17	1.50			1.26	1.23	
	Bottom	1.24	1.00	1.00			1.00	1.00	
Tracer Test 3 (I-131 Only)	Top	1.17							
	Middle	1.00							
	Bottom	1.19							
Tracer Test 4 (I-131 Only)	Top	1.00							
	Middle	1.63							
	Bottom	1.28							

[a] Deposited activities are given relative to lowest activity in each vertical series which has been given an arbitrary value of 1.00.

[b] No analyses requested.

steel, Type 6061 aluminum, Teflon, Kel-F^[a], Kynar^[b], and Pyrex glass. Each sample was analyzed for I-131, Te-132, I-133, and Mo-99 by gamma-ray spectroscopy. The coupons were located adjacent to each other on horizontal rods in the containment vessel. After exposure, and prior to removal from the containment vessel, the coupons were rinsed with water to remove loosely adhering material.

The results of the analyses on the six materials are listed in Table XXI. Of the materials tested, stainless steel showed the highest deposition; the activity on the stainless steel coupons was 2.1 times the average activity on the other materials (excluding Pyrex, which retained the least activity). There were no significant differences in the deposition of these nuclides on Kel-F, Kynar, Teflon, and aluminum. The similar deposition found on aluminum and the organic polymers, Kel-F, Kynar, and Teflon indicates that the deposition mechanism of nuclide retention is predominantly nonchemical. Of the materials tested, Pyrex showed the lowest deposition, which was a factor of 3.7 below that of Kel-F, Kynar, Teflon, and aluminum, and a factor of 7.8 below that of stainless steel.

Uncoated stainless steel and Amercoat 66 coated aluminum coupons located on aluminum holders were contaminated in CDE Run 5 and then analyzed. The results of these analyses are tabulated in Table XXII. Painted coupons received significantly more I-131 deposition by factors of 6.5 and 4.6 than did the uncoated stainless steel coupons. Also, particulates tended to deposit more on the painted surfaces, but the differences were smaller (less than a factor of 3). The greater iodine deposition observed on painted surfaces than on unpainted surfaces is not surprising. It has been demonstrated that at and above the softening point of Amercoat 66 (approximately 194°F), the diffusivity of iodine into Amercoat 66 increases sharply^[9]. The increased particulate deposition on painted surfaces over uncoated surfaces may be the result of a different type of condensation (drop-wise) on the painted surface than that which was on the uncoated surface (film-wise). The different types of condensation for coated and uncoated surfaces were observed in laboratory tests.

[a] Trifluorochloroethylene polymer, product of M. W. Kellogg Co., New York, N. Y.

[b] Vinylidene fluoride resin, product of Pennsalt Chemicals Corp., Philadelphia, Pa.

TABLE XXI

TOTAL ACTIVITY ON COUPONS TESTED FOR FISSION PRODUCT SORPTION
(dps/coupon)

Material	I-131 (x 10 ⁻⁶)	Te-132 (x 10 ⁻⁶)	I-133 (x 10 ⁻⁶)	Mo-99 (x 10 ⁻⁶)
Stainless Steel 304	41	11	130	4.0
	46	15	150	7.4
	39	13	120	6.3
	Average 42	13	133	5.9
Aluminum 6061	23	5.2	65	2.2
	21	5.5	61	2.7
	18	8.6	49	3.6
	Average 21	6.4	58	2.9
Kynar	20	11	54	4.6
	21	11	54	4.9
	19	7	52	3.2
	Average 20	9	54	4.2
Kel-F	13	13	47	4.9
	13	13	35	5.5
	20	14	33	5.8
	Average 15	13	38	5.4
Teflon	10	3.7	28	1.6
	12	4.3	32	1.8
	14	6.0	37	2.6
	Average 12	4.7	33	2.0
Pyrex	3.5	8.0	5.7	3.4
	3.6	8.0	6.0	3.7
	3.6	7.0	6.0	3.0
	Average 3.6	7.7	5.9	3.4

TABLE XXII

DEPOSITION OF FISSION PRODUCTS ON UNCOATED STAINLESS STEEL

AND ON AMERCOAT 66^[e] COATED ALUMINUM COUPONS^[a]

CDE RUN 5

Nuclide	Activity on Uncoated Coupons (dps/cm ²)		Activity on Painted Coupons (dps/cm ²)	
	Horizontally Oriented ^[b] (Surface Up)	Vertically Oriented	Horizontally Oriented ^[c] (Surface Up)	Vertically Oriented
I-131	2.32 x 10 ⁶	8.23 x 10 ⁵	1.52 x 10 ⁷	3.88 x 10 ⁶
Te-132	2.63 x 10 ⁶	2.62 x 10 ⁵	3.86 x 10 ⁶	5.10 x 10 ⁵
Te-129	2.40 x 10 ⁴	1.76 x 10 ³	3.81 x 10 ⁴	5.03 x 10 ³
Ru-103	3.79 x 10 ²	5.45 x 10 ¹ ^[c]	3.25 x 10 ²	3.94 x 10 ¹
Cs-137	1.83 x 10 ³	1.53 x 10 ² ^[c]	4.54 x 10 ³	5.05 x 10 ²
Ba-140	1.65 x 10 ⁴	[d]	3.07 x 10 ⁴	[d]

[a] 3/4-in. diameter by 1/16-in.-thick coupons. All activities reported are an average of that on four to six coupons except as noted; all activities are corrected for decay to the time of meltdown.

[b] Results are for only one coupon.

[c] Results are for an average of three coupons.

[d] Amount present was below detectable limits of the instrument.

[e] Product of Amercoat Corporation, Brea, Calif.

4. DECONTAMINATION OF THE CDE

Although the CDE is decontaminated primarily to permit reuse of the facility, this experience provides practical comparisons of the relative effectiveness of different decontamination procedures on an engineering scale. The decontamination and waste handling equipment used in the CDE is described in Section III. From the experience gained in the five runs, the following general procedure is considered the most effective for decontamination of the CDE.

- (1) Remove all samples
- (2) Remove the fuel residue and other "hot" items that can be removed remotely
- (3) Spray the containment vessel through the vessel sprays and flush the transport line
- (4) Wash the cell interior with cell sprays
- (5) Enter cell and remove remaining "hot" items, manually
- (6) Enter cell and wash walls and equipment, using a portable decontamination unit (pump, tank, hose, and spray nozzle)
- (7) Enter cell and complete cleanup by mopping, brushing, etc.

Following a test, this procedure, which requires approximately one week to complete, reduces the general radiation level in the cell from approximately 100 R/hr to less than 1 mR/hr.

The effectiveness of the different decontamination treatments on the stainless steel containment vessel for a typical run is illustrated in Figure 40. The procedure shown is the "standard" order of decontamination, ie, the use of alkaline permanganate solution, followed by the oxalate-citrate solution. This order was selected mainly for its performance in the coating and decontamination studies^[7]. Insufficient data have been collected in the runs to clearly define the "best" order of application, but the following general conclusions can be drawn:

- (1) Water washing, alone, is insufficient to effect satisfactory decontamination. The initial water wash removes only 20 to 50% of the gross gamma-beta activity, but is slightly more effective for the soluble fission products, Cs-137 and Sr-89.
- (2) No significant differences were apparent in the decontaminability of the reactor vessel and containment vessel using this procedure, although, the surfaces of the reactor vessel were contaminated at 1500°F.
- (3) Turco Alkaline Rust Remover^[a] was substituted for the Turco 4502^[a] in the decontamination sequence, following one of the

[a] A product of Turco Products, Incorporated, Wilmington, Del.

runs. The Turco Alkaline Rust Remover was not as effective as the Turco 4502 for I-131 and Cs-137; however, the subsequent acid treatment removed the I-131 and Cs-137. These conclusions were based on decontamination of stainless steel surfaces; the decontamination of painted surfaces is described more fully in other documents^[7,8].

Fig. 40 Relative effectiveness of solutions for containment vessel decontamination.

V. CONCLUSIONS AND RECOMMENDATIONS

1. CONCLUSIONS

Successful completion of five runs, in which 6- to 10-hr cooled fuel pins containing 7000 to 35,000 Ci of total fission products were melted in a steam atmosphere (containing no air), and numerous trace-level and nonradioactive runs, have demonstrated the safe operability of the CDE. The testing conducted for the LOFT support studies provided practical experience in sampling, containing, and analyzing both high- and low-level fission product mixtures. The integrated-accident sequence runs, conducted in the CDE at high fission product levels, generally confirm the results of low-level, separate-effects runs obtained elsewhere, and furnish significant new information on fission product behavior. The results of the CDE runs are summarized in the following sections.

Fuel Meltdown: The fuel pins showed no visual evidence of cladding damage until the cladding became orange-hot ($> 2000^{\circ}\text{F}$). The main release of radioactivity (greater than 90% of the total release) occurred over a 1- to 2-min period shortly after the cladding became white-hot ($> 2700^{\circ}\text{F}$), followed by a gross release of particulate material (primarily oxides of tin). A continuous slow release of radioactivity, often accompanied by one or two small "puff" releases, occurred throughout the 30-min period in which the fuel was maintained molten. The cladding, which was exposed to steam during heating, formed a friable shell of ZrO_2 which did not melt; however, much of the UO_2 , and those portions of the cladding shielded from the steam, melted completely. Metallic inclusions of a uranium-zirconium alloy formed at the UO_2 -cladding interface. Analysis of the furnace atmosphere during fuel melting showed a buildup of hydrogen and a depletion of oxygen, indicating the Zircaloy and steam had reacted.

Fission Product Release: The fractional releases of fission products decreased, generally, in the order of decreasing volatility of the species in the fuel. The three exceptions (tellurium, molybdenum, and ruthenium) which were noted can be explained by their physical properties. Approximately 100% of the krypton and xenon, 50% of the iodine, 10% of the tellurium, cesium and molybdenum, and less than 1% of the less volatile fission products present in the molten fuel were released.

The iodine releases in these tests are similar to those reported by British experimenters, but are lower, as are the other fission product releases, than results reported by ORNL workers. There appears to be a trend toward increasing release with increasing fuel burnup. These variations, however, may be due to differences in the maximum fuel temperatures (which were not reported) or heating periods, or to the composition of the atmosphere over the fuel in the various experiments.

Based on data from the CDE tests, the release fractions suggested in TID-14844[22] are reasonable for that portion of the core which undergoes melting, but are high for the portion which does not melt. The releases of tellurium, cesium, and molybdenum in the CDE tests exceeded the 1% value quoted in TID-14844, even though the fuel was molten for only 30 min.

Fission Product Transport: Of the fission products released from the fuel (excluding xenon and krypton), most of the cerium and zirconium, little of the molybdenum and ruthenium, and 1/3 to 1/10 of the other fission products deposited in the transport line (maintained at approximately 800°F) between the furnace and containment vessel. The fractions deposited on internal reactor surfaces in a reactor accident should be even larger because of the large, complex surface area of the reactor internals.

Fission Product Distribution: Of the fission products present in the fuel, only krypton, xenon, iodine, tellurium, cesium, and molybdenum reached the containment vessel in significant quantities. Generally, about 1/2 of the fission product particulates reaching the containment vessel were found in the condensate, and the remaining 1/2 deposited on the stainless steel vessel walls. Iodine and cesium were removed preferentially in the condensate, and essentially all of the xenon and krypton remained airborne in the containment atmosphere. The distribution of fission products was affected by the total amount of condensate (more fission products were removed in the condensate as the total amount of condensate increased), by the composition of the containment surfaces (iodine is sorbed preferentially by paint), and by the solubility of the fission product species in the condensate (soluble fission products appeared preferentially in the condensate).

Composition of Containment Atmosphere: An increase of approximately 50% in the hydrocarbon content of the saturated steam-air containment atmosphere was measured during melting of the fuel, indicating that significant quantities of hydrocarbons were released from the fuel. The particulate aerosol in the containment consisted of small (0.001 to 0.7 μ) particles which showed evidence of agglomeration. The predominant particulate species (> 99% of the total particulates) were oxides of tin (tin is an alloying constituent of Zircaloy) and uranium; very small amounts of zirconium oxide were also present.

Three iodine species -- elemental iodine, organic iodides, and iodine sorbed on particulates -- were identified in the CDE containment atmosphere, and a fourth -- hypoiodous acid -- was postulated. Shortly after fuel meltdown, most of the iodine released to the containment was present as elemental iodine and iodine sorbed on particulates. Several hours after meltdown, the majority of the elemental and particulate iodine had been removed in the condensate or deposited on the containment surfaces, leaving primarily hypoiodous acid and organic iodides in the containment

atmosphere. The discrimination of iodine species using Maypack-type samplers was only partially successful. For example, much of the iodine separated as "organic iodides" by conventional cartridge samplers appears to be hypoiodous acid.

Behavior of Noble Gases in Containment: The krypton and xenon concentrations measured in the high-level runs were erratic, but consistently low, mainly because of sampling problems. Results of Xe-133 tracer-level runs obtained with three independent sampling systems, however, were consistent and showed that airborne Xe-133 concentrations remained constant following release to the containment. Only 0.005% of the Xe-133 was removed with the condensate.

Behavior of Particulates in Containment: The rate of removal of the particulate fission products (zirconium, niobium, molybdenum, ruthenium, tellurium, cesium, and barium) from the containment vessel atmosphere was comparable to that of iodine over the first 30 min after release to the containment. After this period, the particulate content decreased more rapidly than the iodine content -- resulting in > 99.8% removal over 24 hr of the total particulate release to the vessel. Considering the low release of particulate fission products from the fuel and the high removal from the containment atmosphere, the containment source term (1% of particulate fission product inventory) cited in TID-14844[22] is high. Approximately 1/2 the total particulate fission products were removed in the condensate, with the remainder deposited on the containment vessel walls. However, the percentage of fission products removed in the condensate increased as the total quantity of condensate (or steam injected) increased, and the water-soluble fission products (such as cesium) were removed preferentially in the condensate.

Behavior of Iodine in Containment: The total iodine released to the containment vessel in these natural response runs was removed quite rapidly (airborne half-time of approximately 8 min) during the first 30 min after release, and its depletion closely followed the depletion of steam from the containment atmosphere. After the first 30 min, the removal rate was much slower (airborne half-time of approximately 7 hr), and the predominant species were hypoiodous acid and organic iodides (a total of 1 to 2% of the iodine inventory in the fuel). Over the first 24-hr period, the total airborne iodine concentration decreased to 1/30 of the initial concentration, or to about 2% of the total iodine inventory in the fuel.

Deposition of Fission Products: The mean deviations in deposition data from numerous samples were $\pm 12\%$ for iodine and $\pm 18\%$ for the particulate fission products. Deposition of fission products on surfaces depended on surface orientation, environmental conditions during deposition, and the surface material. Due to fallout of particulates, deposition on sample surfaces facing up were two to 23 times higher than that on surfaces facing down or

to the side for iodine and particulate fission products, respectively. The deposition of fission products on carbon steel, stainless steel, and painted surfaces on which condensation had occurred was greater than on surfaces of the same material on which no condensation had occurred. The total amount deposited depended on the temperature differential between the surface and bulk atmosphere; greater temperature differences produced more deposition. Condensate flowing over a surface also redistributed deposited fission products, either by removal in the condensate or by relocation on the surface. Iodine deposition appeared to increase on the various materials in the following order: glass, plastics, metals, and paints. Deposition of iodine on painted surfaces increased markedly at temperatures above the softening point of the paint.

Fission Product and Fuel Accountabilities: The fission product and fuel material accountabilities determine to a large extent the credibility of the data from an experiment. The measured quantities of the individual fission products in the various samples from a CDE experiment averaged 75 to 125% of the calculated fuel inventory. Based on the most recent CDE runs (in which the data are the most reliable), the accountabilities of the low- and moderate-volatility fission products (zirconium/niobium, strontium, molybdenum, ruthenium, cesium, barium, and cerium) were generally 80 to 120% of the calculated fuel inventories, while those of the more volatile fission products (iodine and tellurium) were consistently low at 60 to 70% of the fuel inventories. The low accountabilities of these nuclides reflected the many problems associated with the sampling and analysis of high-level mixtures of fission products. Throughout the runs, the accountabilities of uranium and zirconium (which essentially remained in the furnace) were excellent, averaging 99 to 101% of the inventories, respectively.

Decontamination: The most effective procedure tested for decontaminating the largely stainless steel CDE following the high-level runs consisted of sequential spray applications of Turco 4502, water, Turco 4521, and a final water rinse. An initial water rinse removed 20 to 50% of the gross fission product activity, and slightly more of the fission products which are soluble, such as cesium.

The data presented in this document should be extrapolated to power reactor facilities only after careful comparison of the CDE run conditions with the anticipated reactor accident conditions, and evaluation of any differences in fission product behavior caused by variables such as system size. For example, the fractional release of fission products from fuel depends primarily on the fuel temperature, heating time, and the composition of the atmosphere over the fuel; the total release from a power reactor core would represent the summation of releases from many fuel elements at different temperatures, depending largely on their position in the core. Similarly, the fission product removal rates from the containment atmosphere and deposition on surfaces depend on system size and type of surface covering (eg, painted, stainless steel, etc).

2. RECOMMENDATIONS

Although the CDE runs were conducted primarily for support of the LOFT program, some improvements in the CDE samplers and instrumentation which would increase the "spinoff" information on fission product behavior obtained in the future runs are recommended. Specifically, measurement of the fuel cladding temperature and heating rate would make it possible to determine the fission product release as a function of bulk fuel temperature, and to determine the temperature at the "prompt" release. Improved differentiation of the fission product species and modification of samplers to increase the sampling frequency early in the washout period would provide a better understanding of fission product behavior. Isolation and correction of the sampling and analysis problems encountered in the CDE runs, and "closing" the material balances would increase the data reliability.

In addition to the improvements specific to the CDE, some general suggestions for advancing the overall water-safety program appear in order. Although several high-quality, separate-effects tests are being conducted on fission product behavior, improved coordination of these experiments would increase substantially the value of the data obtained. Currently, most of the experiments are conducted under different conditions, probably because the experimenters lack a common description of a typical loss-of-coolant accident and environment. Efforts to correlate results of the various experiments and to combine the separate-effects models on fission product behavior into one overall model should be intensified.

While extensive experimental studies have been, and are being, conducted to define certain of the fission product behavior mechanisms, insufficient work has been done in the following areas:

- (1) The release of fission products from fuel (particularly at burnups $> 10,000$ MWd/tonne) as a function of bulk fuel temperature and the anticipated composition of the gas over the fuel following an accident
- (2) The deposition of fission products in the reactor internals and coolant lines, both with and without emergency core cooling systems
- (3) The positive identification of fission product species and determination of their formation and disassociation mechanisms
- (4) The development of methods for scale-up of data from pilot to commercial size units.

Increased emphasis in these areas would decrease the uncertainties remaining in describing the consequences of a loss-of-coolant accident.

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