THE DECOMMISSIONING OF THE FISSION PRODUCT DEVELOPMENT LABORATORY AT HOLIFIELD NATIONAL LABORATORY

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ABSTRACT

The decontamination of the Fission Product Development Laboratory at Holifield National Laboratory was initiated in FY 1975 after 17 years of processing fission product waste streams to produce commercial quantities of $^{90}$Sr, $^{137}$Cs, $^{144}$Ce, and $^{147}$Pm. The objective of the decommissioning program is the removal of all radiation and contamination areas in the facility to a level which will be compatible with the environment in the foreseeable future.

*Operated by Union Carbide Corporation for the Energy Research and Development Administration.
INTRODUCTION

The fission product research and development work at Holifield National Laboratory (HNL) leading to the construction of the Fission Products Development Laboratory (FPDL) began in 1948 with the investigation of a number of methods for removing long-lived fission products from waste solutions. Fission products were first separated in curie quantities in laboratory glassware using ion-exchange processes. Later, hundred curie quantities were separated in small stainless steel vessels. Finally in 1958, the FPDL facility (Fig. 1) went on stream to separate kilocurie amounts of $^{137}$Cs, $^{90}$Sr, $^{144}$Ce, and $^{147}$Pm from Redox and Purex type waste streams. This program was successfully demonstrated during the first two years of operation, and kilocurie quantities were made available for worldwide distribution.

With the advent of the AEC's Systems for Nuclear Auxiliary Power (SNAP) and increased demand for $^{137}$Cs gamma sources, the requirements for $^{137}$Cs, $^{144}$Ce, and $^{90}$Sr were increased to a level of megacuries per year. To meet this demand, concentrates of $^{137}$Cs, $^{90}$Sr, and $^{144}$Ce were shipped from the Hanford Atomic Products Operation (HAPO) to the FPDL for preparation of the fuel compounds used in the SNAP program. The FPDL facility was modified in 1963 to allow the continuous production of three product streams as follows:
Fig. 1. Fission Product Development Laboratory Holifield National Laboratory
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1. A semi-pure fraction of $^{137}$Cs from HAPO was chemically purified at the FPDL and converted to $^{137}$CsCl powder. This powder was cold pressed into pellets which were doubly encapsulated in stainless steel; the capsules were tested for leaks, and then shipped to the customer.

2. Purified $^{90}$Sr was received from HAPO and converted to $^{90}$Sr titanate. This material was then compacted by vacuum hot press techniques to form a high density pellet. The pellets were encapsulated, usually in Hastelloy "C", and the capsules were tested for leaks and decontaminated before being loaded into a thermoelectric generator for shipment to the customer.

3. A mixed rare earth fraction was received from HAPO; the $^{144}$Ce was separated by solvent extraction, and converted to $^{144}$Ce oxide. The powder was cold pressed and sintered and then sealed in compatible containers for shipment to the customer.

A total of 10.0 megacuries of fission product material was successfully processed at the FPDL during the past decade. During this period, in-cell maintenance requirements were at a minimum and only six cells were decontaminated for the installation of new equipment or repairs to existing equipment. Thus, the decommissioning of 25 individual cells (Fig. 2) highly contaminated with three potent long-lived fission products presented a formidable task for the operating personnel.
Fig. 2. Isometric View Fission Product Development Laboratory
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DISCUSSION

The decontamination program at the FFDL was divided into four distinct phases.

I. Removal and encapsulation of $^{90}\text{Sr}$ products.

II. Decontamination of the manipulator cells used for powder preparation.

III. Decontamination of the process cells which contained the vessels and associated piping used in the purification processes.

IV. Decontamination of the tank farm cells and cell ventilation system.

The original FFDL decontamination proposal was to accomplish this effort in two years based on a 3-shift operating crew of 25 chemical operators. Due to a limited budget in FY 1975, the crew was reduced to ten chemical operators. This reduction in personnel drastically reduced the flexibility of rotating personnel on work in radiation zones while maintaining the minimum practical individual radiation exposure. The crew was placed on a day shift schedule and all work assignments were made on the basis of >75% remote operations to limited radiation exposure. The initial goals for each cell were to reduce the radiation and contamination level to <5 R/hr to allow direct access to the cell for maintenance personnel to remove contaminated equipment to the HNL Solid Waste Storage Area.
REMVAL OF STRONTIUM-90 PRODUCTS

Approximately 500,000 curies of $^{90}$Sr titanate powder was stored in the FPDL facility at the beginning of FY 1975. A physical inventory of the powder was made by weighing each batch and determining the $^{90}$Sr content calorimetrically on a representative sample from individual batches. The powder was then loaded into stainless steel cans (25,000 Ci/can) and sealed with a press fit cap. The inner container was cleaned to a smear level of <10 mr/hr and placed into an outer stainless steel container. The outer containers were TIG welded, leak tested, and cleaned to a smear tolerance of <10,000 dis/min. The sealed containers were placed in AEC-DOT approved shipping containers and are being held for future customer orders.

DECONTAMINATION OF MANIPULATOR CELLS

After the removal of the $^{90}$Sr products it was possible to remove all solid waste from nine of the manipulator cells and transfer it to the HNL Solid Waste Storage Area. A typical manipulator cell and its associated equipment are shown in Fig. 3. The solid waste consisted of glassware, tygon tubing, calorimeters, balances, blenders, graphite die bodies, manipulator-operated tools, ceramic filters, and all equipment that could be disassembled with the manipulators. This material was thoroughly washed in the cell and placed in a plastic-lined 55-gallon drum. The drum was
Fig. 3. Typical FPDL Manipulator Cell
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sealed with a standard ring using special tools designed for manipulator operation. The drums were decontaminated in the manipulator cell using high pressure soap spray equipment and then wiped to a smear tolerance of <1 mR/hr. A total volume of 200 ft$^3$ of solid waste was transferred in a shielded drum waste carrier to the HNL Solid Waste Storage Area and lowered into holes for storage.

Large pieces of equipment, such as a vacuum hot press (Fig. 4), were dismantled in the cell using specially designed tools that could be operated with manipulators. The highly contaminated parts were transferred to the HNL Solid Waste Storage Area. The stainless steel vacuum chambers were cleaned in the cell with high pressure soap solutions and chemicals to remove the insoluble particles of $^{90}$SrTiO$_3$. The cells were then cleaned using the same techniques by spraying through a 12-in.-diam opening in the top of the cell.

When the radiation level in the cell was reduced to <10 R/hr, the top blocks were removed and a 3-in. armor plate shield was placed in the cell block opening. This shield was equipped with a 2-in.-thick plexiglass shield approximately 3.5 ft in diameter. Since the remaining contamination in the cell was $^{90}$Sr ($^{137}$CsCl dissolves readily in hot water), the plexiglass was adequate shielding for the soft beta radiation emitting from the cell opening. The shield had various sized holes with removable plugs that would permit the access of high pressure spray equipment to
Fig. 4. Vacuum Hot Press
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reach a large portion of the cell surfaces. The large diameter of the plexiglass allowed excellent line of sight to all areas of the cell.

When the radiation level in the cell was reduced to <5 R/hr and the contamination level <500 mr/hr, the shield was removed and large pieces of equipment were removed from the cell and decontaminated by hand to a level acceptable for reuse. Permanent equipment (Fig. 5) such as a vessel mounted on the cell wall and its associated piping were decontaminated during the initial cell clean up work. This was accomplished by flushing the system repeatedly with hot solutions of 1 N HNO₃, 1 N NaOH, versene, Turco 4501, oxalic acid, and water flushes between each chemical addition. Analysis of the decontamination solutions would generally indicate a few millicuries of beta activity after the third cycle of the chemical treatment. If the vessel and piping were reading <5 R/hr, maintenance personnel would hot rod the piping and the equipment brackets with a carbon graphite-copper clad cutting electrode on a long rod from the top of the cell. The small pieces were removed from the cell using the manipulators to transfer the material to the drum waste station. Large vessels (~100 gal) were lifted through the top of the cell into plywood containers and transferred to the HNL Solid Waste Storage Area for underground storage.

After the removal of piping, conduits, and instrument lines, additional hot spots were found that would require decontamination or lead shielding to reduce the cell background to <1 R/hr for direct access to the cell.
Fig. 5. Manipulator Cell Process Equipment
As of June 30, 1975, four manipulator cells were decontaminated to a background of 1 R/hr, and direct access for maintenance personnel was approved. The remaining five manipulator cells have a radiation background ranging from 10 R/hr to 100 R/hr and are expected to require approximately ten manyears of effort to make them accessible for direct maintenance. Exposure to operating personnel during this period was maintained below the ERDA limits of 5 Rem per year of penetrating dose ($D_c$) and 15 Rem per year of skin dose ($D_s$). The skin dose became the controlling factor due to the beta component of $^{90}$Sr. The ratio of $D_s:D_c$ ranged from 5:1 to a high of 30:1 and limited our ability to decontaminate with open access to the cell. Special shields, tools, and spray systems were required to limit the $D_s$ dose to the operator on top of the cell bank.

DECONTAMINATION OF THE PROCESS CELLS

The decontamination of the nine FPDL process cells (Fig. 6) during FY 1975 was used as a non-exposure operation to balance the work on the manipulator cells involving exposure to personnel. The process cells contain centrifuges and stainless steel vessels for precipitation, evaporation, crystallization, solvent extraction and storage of fission product cuts. This system was operated from panelboards in non-exposure areas. All vessels and approximately 95% of the transfer lines were operable at the beginning of the decontamination cycle.
Fig. 6. Typical PPDL Process Cell
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A series of hot chemical flushes similar to those used in the manipulator cells were passed through all of the equipment a total of five times. The final decontamination solution contained less than 100 mCi of beta-gamma activity. This was the expected result since the process system had contained fission products in a chemically soluble form.

During the chemical flushes, all vessels were heated to the boiling point of the solution and the vapor allowed to escape to the vessel off-gas system. The vapor flows through the off-gas lines and then is condensed in the vessel off-gas dehumidifier and chilled scrubber system. After the decontamination solution samples indicated a low level of activity in the process system, the main off-gas header was valved-off and chemical solutions were gravity flowed into the off-gas High Efficiency Particulate Air (HEPA) filter chamber. Since the filter chamber is the highest elevation point in the system, the decontamination solutions filled the vessel off-gas scrubbers, dehumidifiers, and lines and overflowed into the process vessels. The off-gas HEPA filter chamber was decontaminated to <1 R/hr and placed in operation with new HEPA filters to contain any residual activity during final phases of decommissioning.

The results of the decontamination of the process system are unknown at this time because removal of the process cell blocks to obtain radiation surveys would involve excessive exposure to personnel. Based on previous decontamination campaigns, the general background in the process cells
should be <10 R/hr with hot spots up to 100 R/hr on lines that could not be
cleaned due to in-cell valve failures. An additional five manyears of
operating effort are expected to be required to reduce the nine process
cells to a radiation level which will permit the direct access of
maintenance personnel to remove the process equipment and piping.

DECONTAMINATION OF TANK FARM CELLS AND CELL VENTILATION SYSTEM

The final phase in the decommissioning program will consist of
decontaminating the process pipe tunnels to the tank farm cells and then
decontamination of the four large tank farm cells. All piping and
tankage will be removed and the cells cleaned to a low smear tolerance
for painting.

The cleaning of the vessels and piping is actually in progress as
all decontamination solutions from the process vessels and cell floor
drains must pass through the tank farm cells on its way to the HNL ILW
system. It is estimated that five manyears operating effort will be
required to complete this work.

The cell ventilation system will be cleaned as each cell is decon-
taminated. Each cell has an individual cell ventilation duct and these
are high pressure sprayed before cell entries are approved. The decon-
tamination solutions flow down the main ducts to the primary cell
ventilation filter pit. This pit contains stainless steel Neva Clog roughing filters plus twelve 1000-cfm HEPA filters.

Decontamination of the cell ventilation filter pit is in progress as this time. The filters have been removed and access is available from the upstream side. Due to erosion of the concrete floor on the downstream side of the filters, we have not been successful in entering this area due to the high radiation readings in the concrete. After decontamination, this area will be reconcreted and painted. New HEPA filters will be installed until the completion of the decommissioning program.

The secondary cell ventilation filter house contains nine HEPA filters which can be replaced in an 8-hour period. Some decontamination and painting will be required to place this system in a surveillance mode of operation.

CONCLUSIONS

The initial phase of the decommissioning of the Fission Product Development Laboratory has been accomplished by reducing the radioactive content in the facility by a factor of $10^3$. The intermediate phase of reducing the curie quantities of fission products by an additional factor of $10^3$ will require considerable personnel exposure and direct contact.
type of operations. The final phase of shielding and sealing to acceptable levels of radiation and contamination can be accomplished with a minimum of personnel exposure. A substantial monetary investment will be required to allow a complete shutdown of the facility.

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REFERENCES


FIGURES

1. Fission Product Development Laboratory, Holifield National Laboratory

2. Isometric View Fission Product Development Laboratory

3. Typical FPDL Manipulator Cell

4. Vacuum Hot Press

5. Manipulator Cell Process Equipment

6. Typical FPDL Process Cell