Evaluation of the Decontamination of the Reactor Coolant Systems at Maine Yankee and Connecticut Yankee

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Evaluation of the Decontamination of the Reactor Coolant Systems at Maine Yankee and Connecticut Yankee

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REPORT SUMMARY

In 1998, utilities carried out chemical decontamination of the reactor coolant loops at two permanently closed PWR plants. They used EPRI's Decontamination For Decommissioning (DFD) process at Maine Yankee, and Siemens' Chemical Oxidation Reduction Decontamination (CORD) process at Connecticut Yankee. This report describes each application, and presents the results and lessons learned.

Background

In February / March and July / August 1998, full loop decontaminations were performed at Maine Yankee (MY) and Connecticut Yankee (CY), respectively. Both MY and CY are permanently shut down plants undergoing decommissioning. As part of the decommissioning process, management personnel at both plants elected to perform chemical decontamination to reduce radiation exposures. At MY they used EPRI's DFD process, which Consumers Energy used previously at Big Rock Point BWR. CY management selected Siemens' CORD process, modified for decommissioning and renamed CORD D UV. Both processes are designed to remove a small amount of base metal and give the high decontamination factors (DFs) required for decontamination prior to decommissioning. Unfortunately, mechanical problems with the letdown system at CY prevented complete application of the CORD D UV process.

Objective

To evaluate application of the EPRI DFD process at MY, and the Siemens CORD D UV process at CY.

Approach

Operationally, both projects were similar. All major primary side plant systems were included in the decontamination flow path with the exception of the reactor pressure vessel (RPV) and most of the steam generator tubes. Plant personnel installed a flow-through nozzle dam "spider" assembly in the core to interconnect the hot and cold leg nozzles, thus completely bypassing the RPV and at the same time providing a flow path from one coolant loop to another. At Maine Yankee the "spider" had six "legs" while at Connecticut Yankee it had eight. Maine Yankee relied heavily on vendor-supplied equipment for pumps, heaters, and ion exchange columns. They required this self-sufficient operation since the utility could not assure availability of plant equipment. In contrast, Connecticut Yankee used the plant's RHR pumps, demineralizers, and pressurizer heaters. Vendors supplied minimal equipment to the CY site. The report discusses the advantages and disadvantages of these two different approaches.

Results

EPRI's DFD process removed 677 lbs. of metal and 103 Curies of radioactivity at MY, while Siemens' CORD D UV process removed 182 lbs. of metal and 131 Curies at CY. The average contact DF was 31.5 and 15.9 at MY and CY respectively. The large difference in the amounts of metals removed is due to problems with the letdown system at CY that prevented application of the "D" option of the CORD D UV process, the step that removes some base metal. This is also a possible explanation for the lower DF at CY. Plant managers used 625 ft^3 of resin at MY, and 465 ft^3 at CY. The project took 67 days at MY, and 122 days at CY, but the chemical processing times were only 20 and 25 days respectively. Both applications met the plants' goals for radiation exposure reduction during subsequent decommissioning activities.

EPRI Perspective

This report contains a comprehensive list of lessons learned during both applications, and will be extremely valuable to utilities planning similar projects in the future. It is difficult to compare the two applications directly, especially since the "D" step of the Siemens process was not used. However, the results clearly demonstrate the importance of carrying out decontamination projects as soon as possible after final shutdown and the benefits of using high flow pumps during the process. A prior EPRI report, *Review of Experience with the EPRI DFD Process* (TR-109036) presents further details of the EPRI DFD process, including results of the application at Big Rock Point.

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Interest Categories

Decommissioning
Low level radioactive waste management
Radiation field control

Key Words

Low level waste disposal
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ABSTRACT

Maine Yankee and Connecticut Yankee are similar vintage PWRs. Both have been permanently shut down and are to be decommissioned. Prior to any decommissioning work, both were subjected to a full loop decontamination. All major primary side systems, except the pressure vessel and the majority of the steam generator tubes, were included in the decontamination flow path. Maine Yankee elected to use EPRI's DFD process while Connecticut Yankee used Siemens' CORD D UV process. The average contact decontamination factor (DF) obtained at MY was 31.5, while that at CY was 15.9. The average final contact radiation field on system piping and components was 20 mR/hr at MY and 27 mR/hr at CY. The total volume of ion exchange resin used at MY was 625 ft^3 but of this amount 90 ft^3 was used for boron removal leaving a net attributable to DFD of 535 ft^3. At CY the total volume of IX resin was 465 ft^3 but only 115 ft^3 of this total is directly attributable to CORD. The balance is due to the presence of transuranics from fuel failures and the plant's desire to avoid generating a greater than Class C waste form.

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EXECUTIVE SUMMARY

The Maine Yankee (MY) and Connecticut Yankee (CY – also known as Haddem Neck) nuclear power plants are permanently shutdown and are to be decommissioned. To reduce radiation exposure ALARA during decommissioning, management personnel at both plants elected to perform chemical decontaminations of all major systems prior to the start of main decommissioning activities. EPRI's DFD process was selected for MY, while Siemens' CORD D UV process was selected for CY. Both processes are more aggressive than conventional, operational-type decontamination processes such as LOMI, CITROX, and CAN-DEREM™, in that they are designed to remove a few microns of base metal as well as the oxide film. Removal of some base metal is usually required to achieve the high decontamination factors (DFs) typically desired for decontamination for decommissioning. While a DF of 10 is generally considered adequate for operating plants, a target DF of 100 or more is not unreasonable for decontamination prior to decommissioning. To achieve such a high DF, removal of a thin layer of underlying base metal is required in addition to removal of the oxide film. Operational-type decontamination processes are, by design, too "mild". They have been specifically developed not to remove base metal.

Operationally, both projects were similar. All major plant systems were included in the decontamination flow path with the exception of the reactor pressure vessel (RPV) and the majority of the steam generator tubes. A flow-through nozzle dam "spider" assembly was installed in the core to interconnect the hot and cold leg nozzles thus completely bypassing the RPV and at the same time providing a flow path from one coolant loop to another. At Maine Yankee the "spider" had six "legs" while at Connecticut Yankee it had eight. Bypassing the RPV had the advantage of avoiding corrosion of activated material, which could contribute significantly to the amount of radioactivity removed and waste generated, but would have little effect on radiation exposure. Bypassing the SG tubes avoided a large surface area of highly contaminated Inconel, decontamination of which would have contributed very little to exposure reduction since only the outer 10 rows of tubes contribute significantly to external radiation fields. In-situ decontamination of the SG tubes would offer little immediate benefit to the plants.

MY relied heavily on vendor-supplied equipment for pumps, heaters, and ion exchange columns. A self-sufficient operation was required since availability of plant equipment could not be assured. In contrast, CY used the plant's RHR pumps, demineralizers, and pressurizer heaters. Minimal vendor-supplied equipment was brought to site. The advantages and disadvantages of these two different approaches are discussed.

The MY decontamination was performed in two separate applications, each taking about 8 days. Eleven cycles of DFD were applied in the first and 13 cycles in the second. An average contact DF of 31.5 was obtained. The average initial contact radiation field of 460 mR/hr was reduced to 20 mR/hr. On the most radiologically significant points, i.e., those with an initial field of >500 mR/hr, the average DF was 146. The average general area dose rate reduction factor (DRF) was ~4. Total ion exchange resin waste generated was 625 ft^3 of which ~90 ft^3 was consumed in removing boron leaving a net of ~535 ft^3 attributable to DFD. A total of 677 lbs. of metal and 103 Curies of (-emitting radionuclides were removed during this application. A decontamination of parts of the RCS in 1995 removed about 400 Curies of radioactivity. This contributed to the lower-than-expected activity removal (several hundred Curies had been expected), and also to the lower-than-expected DFs (an average contact DF of >100 had been expected).

The CY decontamination was performed in a single application of two cycles of CORD over a 28 day period. However, only about 9 days was attributable to CORD. The balance was due to delays caused by problems with the plant's letdown system. An average contact DF of 15.9 was obtained. The average initial contact radiation field of 234 mR/hr, approximately one-half of that at MY, was reduced to 27 mR/hr. The difference in DF based on initial radiation field, as observed at MY, was not observed at CY. The average DF was essentially the same for points <500 mR/hr as for those >500 mR/hr. The average DRF was ~5. Total waste ion exchange was 465 ft^3 but only 115 ft^3 was directly attributed to CORD. The balance was due to the plant's aim of avoiding a greater than Class C waste as a result of the TRU present in the system from previous fuel failures. A total of 182 lbs. of metal and 131 Curies of activity were removed. The amount of metals removed is much lower than at MY since the "D" option of the CORD D UV process was never implemented during this decontamination. It is this part of the process that removes some base metal. Originally it was planned to apply it in Cycle 3 (and Cycle 4, if necessary), but because of problems with the plant's letdown system and the good decontamination factors obtained after two cycles, the decontamination was terminated after two cycles.

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INTRODUCTION

Full loop decontaminations (FLD)[a] have recently been performed at Maine Yankee (MY) and Connecticut Yankee (CY). Both plants are permanently shutdown and are being decommissioned. As part of the decommissioning process, management personnel at both plants elected to perform FLDs to reduce radiation exposure during decommissioning activities.

Operationally, both projects were similar in that the reactor pressure vessel (RPV) and the majority of the steam generator (SG) tubes were bypassed to avoid decontaminating components that would have little effect on radiation exposure, and to minimize the volume of waste generated. Bypassing the RPV had the additional advantage of avoiding corrosion of activated material that could contribute significantly to the amount of radioactivity as a result of using the more aggressive chemical processes employed for decontamination for decommissioning compared to the milder chemical decontamination processes used for operational-type decontaminations. Bypassing the SG tubes avoided a large surface area of highly contaminated Inconel, decontamination of which would have contributed very little to exposure reduction since only the outer 10 rows of tubes contribute significantly to external radiation fields. Had the tubes been included, it would have also added substantially to the final waste volume due to the large surface areas involved and the relative corrosion sensitivity of Inconel to both decontamination reagents. In-situ decontamination of the SG tubes would thus offer little immediate benefit to the plants for the potentially substantial cost involved.

Chemically the two projects were quite different. MY employed the *EPRI Decontamination For Decommissioning* (DFD) process, while CY used the Siemens *Chemical Oxidation Reduction Decontamination* (CORD) process, especially modified for decontamination for decommissioning and renamed the CORD D UV process. Both processes are described in detail in this report. Both are more aggressive than conventional, operational-type decontamination processes such as LOMI, CITROX, and CAN-DEREM™, in that they are designed to remove a few microns of base metal as well as the oxide film. Removal of some base metal is usually required to achieve the high decontamination factors (DFs) typically associated with decontamination for decommissioning. While a DF of 10 is generally considered adequate for operating plants, a target DF of 100 or more is not unreasonable for decontamination prior to decommissioning. To achieve such a high DF, removal of a thin layer of underlying base metal is required in addition

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[a] This term is used rather than full system decontamination (FSD) since neither the pressure vessel nor the majority of the steam generator tubes were included in the decontamination.

to removal of the oxide film. Operational-type decontamination processes are, by design, too "mild". They have been specifically developed not to remove base metal.

The operational and chemical aspects of both projects are presented. Both the strong and weak points of each process are described. Results obtained (metals and (-emitting radionuclides removed, decontamination factors (DF) and dose rate reduction factors (DRF) achieved, etc.) are presented. Types and quantities of waste generated are discussed. Waste handling techniques at each plant are described. An overall comparison of the key parameters for the two processes is presented based upon data obtained from the actual applications. Lessons learned from each project are presented. There are approximately 20 lessons learned from each application.

For the most part this report focuses on what happened rather than what might have happened under ideal circumstances. The major exception to this is the "D" option of the CORD D UV process at CY. It was always planned to be an integral part of the process, but because of problems and delays caused by the plant's demineralizer system, it was never applied. Statements are included that predict what might have happened had this option been available. These predictions are based on laboratory beaker tests and loop tests with CY artifacts, Siemens' experience, and published references to decontaminations where it was applied. The prognostications are not entirely theoretical.

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APPROACH TO DECONTAMINATION

Maine Yankee

Three of the key primary systems were included in the Maine Yankee full loop decontamination. These were the Reactor Coolant System (RCS) and Pressurizer, the Chemical and Volume Control System (CVCS), and the Residual Heat Removal System (RHRS). These three systems accounted for the vast majority of radiation exposure to plant personnel during operation. Similar exposure results would have been expected for decommissioning activities had these systems not been decontaminated. A brief description of each system and how it was prepared for the decontamination is presented below.

Reactor Coolant System and Pressurizer

The RCS at Maine Yankee consists of:
- a single reactor pressure vessel (RPV)[b],
- three steam generators,
- three reactor coolant pumps (RCPs), and
- associated piping and valves.

The pressurizer, which is part of the Pressurizer and Pressure Relief Systems, consists of:
- one pressurizer vessel,
- a pressurizer surge line, and
- pressurizer spray lines.

The system is arranged in three similar 33½″ diameter piping loops (Loops #1, #2 & #3), each containing one steam generator, one reactor coolant pump, an 8″ bypass line and two loop stop valves. Each loop connects to the reactor pressure vessel at two locations in a symmetrical arrangement around its circumference. During normal operation, flow in each loop passes from

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[b] The RPV itself was not decontaminated, as will be explained in the following sections. The decision to not include it in the flow path was based on four main reasons:
1. There was no significant radiological benefit;
2. The amount of radioactivity removed would be increased through corrosion of activated material;
3. The amount of waste would be increased because of the larger system volume and the additional metal and radioactivity removed;
4. Flow through the vessel was a problem at the proposed decontamination flow rates.

the RPV through hot leg piping to the steam generator. It then flows to the pump and back to the RPV through the cold leg piping. The pressurizer is connected to RCS Loop #1 hot leg by its 12″-diameter surge line.

The primary construction materials of this system are carbon steel, Inconel, and stainless steel. All carbon steel surfaces in the RCS are clad with either stainless steel or Inconel. There is no wetted carbon or low alloy steel surfaces in the RCS other than those that may exist due to cracks in the cladding.

Chemical and Volume Control System

The CVCS at Maine Yankee consists of:

- a regenerative heat exchanger,
- two letdown drag valves,
- a letdown heat exchanger,
- two letdown filters (pre and post),
- a volume control tank (VCT),
- miscellaneous small diameter piping and valves
- four ion exchange vessels[c],
- three charging pumps[c], and
- boric acid makeup, storage and transfer equipment[c]

The CVCS is connected to the RCS via a 2½″ connection on the Loop #1 cold leg. During normal operation, the flow (up to 100 – 120 gpm) is cooled through a vertical, shell and tube, regenerative heat exchanger. System pressure is then reduced through the two drag letdown valves to approximately 300 psig. The fluid is then further cooled to approximately 120°F, filtered, and passed through one of four, 32 cubic foot capacity, ion exchange vessels to the suction of the charging pumps. A 4,000-gallon capacity VCT provides adequate suction head for the charging pumps. Discharge flow from the charging pumps is returned to the RCS via connections on the cold legs of Loops #2 and #3.

The primary construction materials of this system are 300 series stainless steels. There are no wetted portions of this system which contact carbon or low alloy steels.

Residual Heat Removal System

The RHRS at Maine Yankee consists of:

- two centrifugal pumps,
- two RHR heat exchangers[d], and
- associated piping and valves.

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[c] During the decontamination, the ion exchange columns, charging pumps, and boric acid equipment were either bypassed or isolated from the flow path.

[d] The RHR heat exchangers were isolated from the flow path during the decontamination and were only valved in during the latter stages of the final DFD cycle.

The inlet to the RHRS is connected to the RCS on the hot leg of Loop #2. During normal operation, the flow passes directly to the suction header for the two RHR pumps. The pumps discharge through parallel feed trains to the tube side of the heat exchangers and back into the RCS via connections on the cold legs of all three loops.

The primary construction materials of this system are 300 series stainless steels.

Decontamination Preparations

Preparations for the decontamination commenced well in advance of the application. The key activities included the following:

- The fuel, reactor head, vessel internals, and core barrel were all removed from the RPV. The core barrel was placed in the refueling cavity. Because the top of the core barrel extended above the water level in the cavity, additional shielding was required to minimize radiation levels on the refuel floor. Core barrel removal was necessary to permit access to all six of the 33½″ RCS hot and cold leg piping connections at the RPV nozzles.

- The decontamination equipment was shipped to site and setup on the 20′ elevation adjacent to the equipment hatch inside containment. This equipment consisted of a circulating pump, heater, filter, chemical injection equipment, ion exchange columns complete with water shield walls, interconnecting piping and hoses and waste processing equipment. The vendor was required to supply a self-sufficient operation since availability of plant equipment could not be assured.

- A flow-through nozzle dam assembly was installed in the six hot and cold leg nozzles of the RPV to completely bypass the RPV and provide a flow path from one RCS loop to another. A photograph of this assembly is shown in Figure 2.1. Installation was performed from the vessel side using the polar crane. This package of equipment was assembled on the refuel floor, lowered into the vessel, aligned with the nozzles and supported on the vessel flange. The individual dams were then mechanically positioned into each of the six nozzles. Once in place, the double seal package on each dam was pressurized from a remote control panel located on the refuel floor and a leak test completed. Monitoring of seal pressure was performed at regular intervals throughout the decontamination.

- The RCP seals were prepared for the decontamination by increasing the seating pressure between the stationary and rotating faces of the fourth stage vapor seal. In addition, the lower three stages of the four-stage seal package were bypassed and a vent line was run from the fourth stage of each seal to a 55-gallon drum. Provision was also made to bring the other seal stages on-line if significant leakage was experienced. This alternative was not necessary.

- Jumpers were installed between the primary side hot and cold leg manways of each of the three steam generators. This was necessary to bypass the large surface area of tubing in the steam generators. In addition, the jumpers included connections to the system from the decontamination equipment.

- The RCP seal injection lines were isolated from the flow path.

- A temporary, flexible, 3″ jumper was installed across charging pump, P-14A, to permit flow around the charging pumps.

- The media was removed from the letdown pre-filter and the purification post-filter to eliminate them as potential high pressure-drop components during the decontamination.

- The internals were removed from the two letdown drag valves, LD-A 9 and LD- A 10 to reduce overall CVCS pressure drop during the decontamination.

- A telemetric dosimetry system was installed to remotely monitor radiation levels at fourteen different locations within the decontamination flow path.

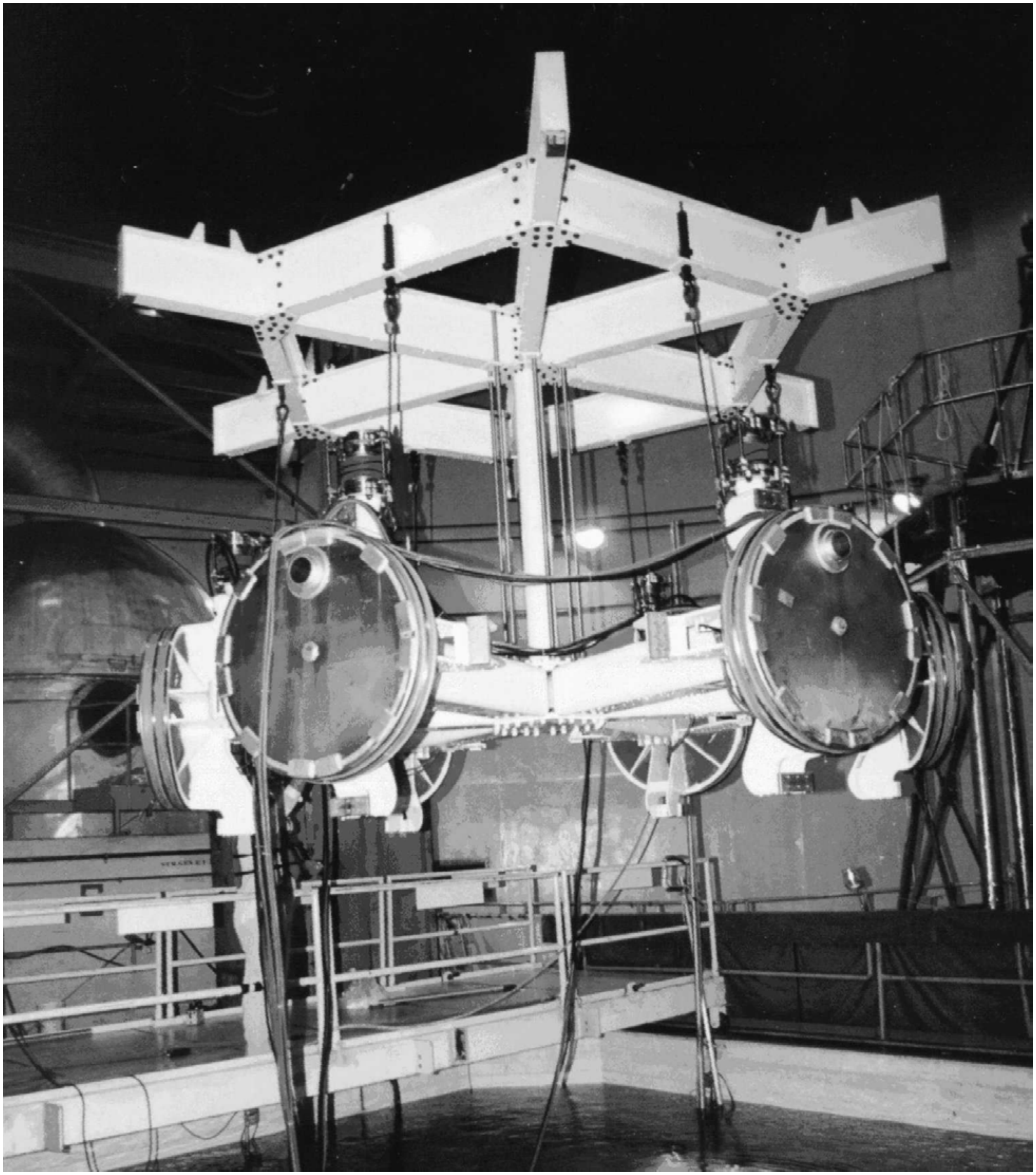

Figure 2.1

Maine Yankee Nozzle Dam Spider Assembly

Connecticut Yankee

Similar to Maine Yankee, the three key primary systems included in the Connecticut Yankee full loop decontamination were the Reactor Coolant System (RCS), the Chemical and Volume Control System (CVCS), and the Residual Heat Removal System (RHRS). In addition, portions of the Purification System and the High Pressure Safety Injection (HPSI) System were also included. A brief description of each system and how it was prepared for the decontamination is presented below.

Reactor Coolant System

The RCS at Connecticut Yankee consists of:

- a single reactor pressure vessel (RPV)[e],
- four steam generators,
- one pressurizer complete with surge line and heaters with 1200 kW of available capacity
- four reactor coolant pumps (RCPs), and
- associated piping and valves.

The system is arranged in four similar 27½″ diameter piping loops (Loops #1, #2 #3 & #4), each containing one steam generator, one reactor coolant pump, a 6″ bypass line and two loop stop valves. Each loop connects to the reactor pressure vessel at two locations in a symmetrical arrangement as shown in Figure 4.3. During normal operation, flow passes from the RPV through hot leg piping to the steam generator. It then flows to the pump and back to the RPV through the cold leg piping. The pressurizer is connected to RCS Loop #4 hot leg by its 10″-diameter surge line. Similar to Maine Yankee, the primary construction materials of this system are carbon steel, Inconel, and stainless steel, with all carbon steel surfaces clad with stainless steel.

Chemical and Volume Control System

The CVCS at Connecticut Yankee consists of:

- a regenerative heat exchanger,
- three letdown orifices,
- a non-regenerative heat exchanger,
- three reactor coolant filters (one ion exchange prefilter and two postfilters),
- a volume control tank (VCT),
- miscellaneous small diameter piping and valves
- four ion exchange columns,
- three charging pumps, and
- boric acid makeup, storage and transfer equipment.

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[e] Similar to Maine Yankee, the RPV was not decontaminated, . The decision to exclude it from the flow path was made for the same reasons. See footnote b on page 2-1.

For the decontamination, the letdown orifices were removed, the charging pumps and prefilter were bypassed, and the boric acid equipment was isolated from the flow path.

The CVCS is connected to the RCS via a 3″ line on the Loop #1 cold leg. During normal operation, the flow (<100 gpm) is cooled through a shell and tube, regenerative heat exchanger. System pressure is then reduced through the three letdown orifices to approximately 250 psig. The fluid is then further cooled to approximately 120°F by the non-regenerative heat exchanger, filtered, and passed through one of the 45 cubic foot capacity, ion exchange vessels to the suction of the charging pumps. A 1,100 gallon capacity VCT provides adequate suction head for the charging pumps. Discharge flow from the charging pumps is returned to the RCS via a connection on the Loop #2 cold leg. The primary construction materials of this system are 300 series stainless steels. There are no wetted portions of this system which contact carbon or low alloy steels.

Residual Heat Removal System

The RHRS at Connecticut Yankee consists of:

- two centrifugal residual heat removal pumps,
- two RHR heat exchangers, and
- associated piping and valves.

The inlet to the RHRS is connected to the RCS on the hot leg of Loop #1. During normal operation, the flow passes directly to the suction header for the two RHR pumps. The pumps discharge through parallel feed trains to the tube side of the heat exchangers and back into the RCS via a connection on the cold leg of Loop #2. The primary construction materials of this system are 300 series stainless steels.

Decontamination Preparations

Preparations for the decontamination commenced well in advance of the application. The key activities included the following:

- A temporary line was added from one of the core deluge connections on the RHR pump discharge to the inlet of the vendor's decontamination equipment. In addition, a special connection was designed and installed at the pressurizer manway to accommodate hoses from the outlet of the vendor's equipment. This modification provided a flow path from the discharge of the operating RHR pump to the Siemens' equipment and from the equipment into the pressurizer. The vendor's decontamination equipment was setup on the 48' 6″, charging floor elevation. This equipment consisted of a 600 kW heater skid, three ultra-violet (UV) burner skids, chemical injection equipment and interconnecting hoses. (Mod. #1)

- Tie-ins were prepared for connections at charging pump 1B in the CVCS for a replacement pump supplied by Siemens. This modification allowed a higher capacity, low pressure pump (letdown return pump) to be installed. The letdown return pump provided the return flow from the outlet of the on-line letdown ion exchange column back into the RCS. It required sufficient capacity to match the inlet flow provided by the Booster pump installed per Mod #3 below. (Mod #2)

- A booster pump was installed inside containment in the letdown line upstream of the regenerative heat exchangers. The pump was located in the section of line that was removed for artifact testing. The purpose of this modification was to supply sufficient flow to the inlet of the on-line letdown ion exchange column to meet the requirements of the decontamination. (Mod #3)

- A line was added from the outlet of the Spent Fuel Pool (SFP) ion exchange column ('C') to the discharge of the 'A' HPSI pump. The purpose of this line was to connect the outlet of the SFP ion exchange column to the HPSI pump discharge, which returned the flow to the RCS. This enabled the SFP IX column to be used during the decontamination and provided the additional IX flow and capacity required. (Mod #4)

- The three flow orifices and their associated block valves were removed from the letdown line and replaced with a section of piping. The purpose of this modification was to reduce the pressure drop in the letdown line in order to maximize flow to the on-line letdown ion exchange column. (Mod #5)

- Temporary power supplies were prepared and connected to the temporary decontamination equipment. Sufficient power was necessary to run the Siemens' pumps, heaters, and ultra-violet lamps. (Mod #6)

- A flow-through nozzle dam jumper assembly was installed in the eight hot and cold leg nozzles of the RPV to completely bypass the RPV and provide a flow path from one RCS loop to another. In order to access these nozzles, the fuel, reactor head, vessel internals and core barrel were all removed from the RPV. The core barrel was placed in the refueling cavity with additional shielding to minimize radiation levels on the refuel floor. Installation was performed from the vessel side using the refuel crane. This package of equipment was assembled on the refuel floor, lowered into the vessel, aligned with the nozzles and supported on the vessel flange. The individual dams were then mechanically positioned into each of the six nozzles. Once in place, the double seal package on each dam was pressurized from a remote control panel located on the refuel floor and a leak test completed. (Mod #7)

3

DECONTAMINATION METHOD

Maine Yankee

As mentioned in Section 1, the EPRI DFD process was applied at MY. DFD is an acronym for Decontamination For Decommissioning. It was developed by Bradtec under contract from EPRI (1 2 3). It is based on the use of dilute fluoroboric acid, HBF_4, combined with changes in the solution redox potential through the use of potassium permanganate, $KMnO_4$, and oxalic acid, $H_2C_2O_4$. It is a more aggressive process than operational-type decontamination processes such as LOMI, CAN-DEREM™ and CITROX with decontamination factors (DFs) of >1,000 being reported. (2) It achieves these very high DFs through a combination of oxide film dissolution and removal (corrosion) of the top few microns of base metal. The process is described in detail below.

The use of fluoroboric acid for decontamination of reactor materials was developed by Jozef Hanulik in Switzerland in the 1980s under the name DECOHA and originally patented in that country in 1985. (4 5) A US patent was granted in 1989. (6) These patents cover the use of fluoroboric acid in the concentration range 0.5-50%. In EPRI DFD it is employed at a much lower concentration of ~0.1% which makes the process amenable to cleanup by ion exchange resin, a technology with which decontamination vendors are very familiar.

Fluoroboric acid is prepared from a solution of boric acid by reaction with hydrofluoric acid as shown below. It is available commercially worldwide as a 50% solution containing about 1% boric acid.

Production of Fluoroboric Acid

$$\underset{\text{boric acid}}{H_3BO_3} + \underset{\text{hydrofluoric acid}}{3HF} \rightarrow \underset{\text{fluoroboric acid}}{HBF_4} + \underset{\text{water}}{3H_2O}$$

The chemical reactions involved in deposit dissolution and base metal corrosion are shown below.

Dissolution of Magnetite in Fluoroboric Acid

$$\begin{array}{ccccc} Fe_3O_4 + & HBF_4 & \rightarrow & 2Fe(BF_4)_3 + Fe(BF_4)_2 + & H_2O \\ \text{magnetite} & \text{fluoroboric} & & \text{iron} & \text{water} \\ & \text{acid} & & \text{fluoroborates} & \end{array}$$

Corrosion of Base Metal by Fluoroboric Acid

$$\begin{array}{ccccc} Fe + & 2HBF_4 & \rightarrow & Fe(BF_4)_2 + & H_2 \\ \text{iron} & \text{fluoroboric} & & \text{ferrous} & \text{hydrogen} \\ & \text{acid} & & \text{fluoroborate} & \end{array}$$

Early in the development program of the EPRI DFD process, it was found that high DFs could not be achieved on some materials, particularly 300-series stainless steel (SS). This was attributed to a combination of a high concentration of chromium in the oxide layer, which impeded its dissolution (particularly SS from PWRs), and the presence of a thin, highly protective, chromium-rich film on the stainless steel itself. For operational-type decontaminations, only the chromium in the oxide film is removed; that in the thin, inner protective film is left in place to minimize base metal corrosion. It was determined that exposing the SS surface to a low concentration of potassium permanganate (100-300 ppm) in the presence of fluoroboric acid would oxidize the chromium in both the deposited oxide and the in-situ film thus allowing both oxide dissolution and base metal corrosion to take place. The chemical reaction involved in oxidation of chromium oxide to the soluble bichromate form is shown below.

Oxidation of Chromium Oxide in Potassium Permanganate

$$\begin{array}{cccccc} Cr_2O_3 + & 2KMnO_4 + H_2O & \rightarrow & 2HCrO_4^- + & 2MnO_2 + & 2K^+ \\ \text{chromium} & \text{perman-} & & \text{bichromate} & \text{manganese} & \text{potassium} \\ \text{oxide} & \text{ganate} & & & \text{dioxide} & \end{array}$$

The permanganate that does not react with the chromium oxide eventually decomposes to manganese dioxide as shown in the following reaction.

Decomposition of Permanganate in Acid Solution

$$\begin{array}{cccccc} 4H^+ + & 4MnO_4^- & \rightarrow & 4MnO_2 + & 3O_2 + & 2H_2O \\ \text{acid} & \text{perman-} & & \text{manganese} & \text{oxygen} & \text{water} \\ & \text{ganate} & & \text{dioxide} & & \end{array}$$

The manganese dioxide is dissolved with a stoichiometric amount of oxalic acid as shown in the following reaction. An excess of oxalic acid may be added to assist in oxide film dissolution and removal of a small amount of base metal

Dissolution of Deposited MnO_2 With Oxalic Acid

$$\underset{\substack{\text{manganese}\\\text{dioxide}}}{MnO_2} + \underset{\substack{\text{oxalic}\\\text{acid}}}{H_2C_2O_4} + 2H^+ \rightarrow \underset{\substack{\text{manganous}\\\text{ion (soluble)}}}{Mn^{2+}} + \underset{\substack{\text{carbon}\\\text{dioxide}}}{2CO_2} + \underset{\text{water}}{2H_2O}$$

The oxalic acid also converts the bichromate, $HCrO_4^-$, to Cr^{3+} as shown in the following reaction. This allows the chromium to be removed as a cation on ion exchange resin, as explained below.

Reduction of Bichromate With Oxalic Acid

$$8H^+ + \underset{\substack{\text{oxalic}\\\text{acid}}}{3H_2C_2O_4} + \underset{\text{bichromate}}{2HCrO_4^-} \rightarrow \underset{\text{chromium}}{2Cr^{3+}} + \underset{\substack{\text{carbon}\\\text{dioxide}}}{6CO_2} + \underset{\text{water}}{8H_2O}$$

A complicating factor with Cr^{3+} is that it tends to form chromium oxalate, $Cr(C_2O_4)_3^{3-}$, from the excess oxalic acid. This compound is not broken down by cation exchange resin and thus chromium tends to build up in solution during the course of a DFD application. It is removed very efficiently at the end of the application when anion exchange and mixed bed resins are placed on line.

Following addition of the oxalic acid, the solution is then passed through a cation exchange column to remove all cations (e.g., K^+, Mn^{2+}, Fe^{2+}, Ni^{2+}, Co^{2+}, Cr^{3+}, etc.) leaving only fluoroboric acid in solution. The reaction involved in removal of cations on ion exchange is shown below.

Removal of Cations by Ion Exchange Resin

$$\underset{\text{iron}}{Fe^{2+}} + \underset{\substack{\text{cation}\\\text{resin}}}{2H{:}R} \rightarrow \underset{\substack{\text{captured}\\\text{metal}}}{Fe{:}R_2} + \underset{\substack{\text{hydrogen}\\\text{ions}}}{2H^+}$$

If excess oxalic acid was added, a stoichiometric amount of $KMnO_4$ is added to convert it to CO_2 and water. The removal of cations, particularly Mn^{2+}, must be completed before the start of the next cycle. This is called the “transition phase” of the cycle.

Once all cations have been removed, the ion exchange columns are valved out and additional potassium permanganate is then added to start the second cycle. As many cycles as are necessary are then performed depending upon the thickness of the film and the final DF required.

In summary, the DFD process consists of the cyclic application of three chemical phases that are summarized below. The solvent base is 10 millimolar (0.088%) fluoroboric acid which results in a pH of ~2, a value that is maintained throughout the process. It is added initially once the operating temperature is reached. For 300 series stainless steel and Inconel systems, 200ºF is the temperature typically used. A crud burst and some metal dissolution may occur following its addition. The water in the system is circulated through a filter and cation resin until the dissolved metals are <10 ppm and most particulate material has been removed. The cation exchange columns are then valved out in preparation for phase 1.

(i) Oxidation phase. Addition of ~200 ppm $KMnO_4$. It is circulated at 200ºF until it has all been converted to MnO_2 as described above. Ion exchange resin is not used in this phase, which typically takes between 2 and 4 hours.

(ii) Reduction phase. Addition of a stoichiometric amount of oxalic acid to dissolve the MnO_2 and assist in oxide film dissolution. It also reduces the $HCrO_4^-$ to Cr^{3+} as explained above. The cation exchange columns are placed online following addition of the oxalic acid. The flow rate should be sufficient to give a purification half-life[f] of <1 hour, with ~30 minutes being preferred. Once manganese dioxide dissolution and cation cleanup are complete, the IX columns are valved out in preparation for phase (iii). Phase (ii) typically takes 4 to 6 hours, but may be as long as 12 hours.

(iii) Transition phase. The third phase returns the solvent to the fluoroboric acid-only condition.[g] This phase is initiated by taking the IX columns off line and adding a stoichiometric amount of $KMnO_4$ to oxidize any remaining oxalate from phase (ii) to carbon dioxide and water. Cation exchange resin is then employed as in phase (ii) to remove any cations that are generated. Once cation cleanup is complete, the IX columns are valved out in preparation for the second cycle commencing with phase (i). Phase (iii) typically takes 2 to 4 hours.

Completion of the transition phase completes one cycle of DFD. As many additional cycles are applied as necessary to achieve the desired DF. Typically between 6 and 12 cycles are required.

After the last cycle, the temperature is lowered to ~160ºF and anion exchange columns are placed online to remove the fluoroboric acid and any other anions that may have accumulated during the application (e.g. SbO_3^-). The reaction of fluoroborate with anion resin is shown below. The anion resin, combined with cation resin to remove the last traces of cations, results in only pure water being left in the system.

Removal of Fluoroboric Acid by Anion Resin

H^+	$+ BF_4^- +$	$R{:}OH$	$\rightarrow R_2{:}BF_4 +$	H_2O
acid	fluoro- borate anion	resin	captured fluoroborate	water

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[f] Purification half-life is defined as the time required to reduce the concentration of any species by a factor of 2, assuming it is being removed with 100% efficiency by the ion exchange resin. It is calculated as follows:

$$t_{½} = \text{System Volume} \times \ln(2) \div \text{IX Flow Rate}$$

[g] In theory, this is what should happen. In practice, chromium builds up in solution and is only removed during the final cleanup phase when anion exchange resin is placed on line.

The EPRI DFD concept is summarized in Table 3.1. The only three chemicals a vendor brings to site for a decontamination are fluoroboric acid (typically a 50 wt % aqueous solution), potassium permanganate and oxalic acid dihydrate.

Table3.1
The EPRI DFD Concept

Feature	Advantages
Uses fluoroboric acid at dilute concentration (10 mM) with redox potential control by potassium permanganate and oxalic acid	Reagent dilute and chemical costs low Suitable for cleanup on IX resin
Three blended steps: (i) Permanganate oxidation (ii) Manganese dioxide dissolution and oxide film dissolution with oxalic acid (iii) Oxalate destruction	No reagent clean-up between steps Fluoroboric acid continuously recycled
Continuous clean-up by cation exchange resin	System is “rinsed” with progressively cleaner solution, a feature essential for high DFs
Process chemistry progressively dissolves base metal	Essential for high DFs
Process not excessively corrosive to reactor materials	Suitable for use in fuel-out full system decontamination of a permanently shut-down plant
Oxalic acid used in the process is converted to CO_2	No chelants in waste
Fluoroboric acid removed by anion exchange resin	Ion exchange resin waste only
Under ideal conditions, decontamination factors typically range from 1,000 – 500,000, depending on number of cycles	Metal can be cleaned to free release limits

Connecticut Yankee

As mentioned in Section 1, the Siemens CORD D UV process was applied at CY. CORD is an acronym for Chemical Oxidation Reduction Decontamination. It evolved from the OZOX process (from OZone/OXalic acid) in which the original concept was to employ ozone as the oxidizing agent, but because of technical difficulties alkaline permanganate was used for all field applications. (7) In the mid-1980s permanganic acid was substituted for alkaline permanganate and the name was changed to CORD. In the early 1990s the UV option was added to decompose the residual oxalic acid and thus reduce the volume of ion exchange resin required. (8) This modification is discussed further below. In the mid-1990s, the D (for decommissioning) option was added. This involves exposing the decontamination solution to intense UV light. (12) This reduces the solution redox potential and makes it much more aggressive to base metal. Removal of some base metal is required for the high DFs associated with decontamination for decommissioning. This option is also discussed further below.

CORD is a regenerative, DCD process that consists of four blended steps:

(i) Oxidation step. Addition of permanganic acid ($HMnO_4$ or more simply HP for analogy with AP and NP) at a concentration of 50-300 ppm and a temperature of 190°F. This step typically takes 3-6 hours. The permanganic acid can be produced in-situ by cation exchange of a solution of potassium permanganate, or it can be produced in a concentrated form in a separate ion exchange module prior to the start of the decontamination. Production of permanganic acid from potassium permanganate by ion exchange is shown in the reaction below. Permanganic acid only exists in solution.

Production of Permanganic Acid from Potassium Permanganate by Cation Resin

$$\underset{\text{potassium permanganate}}{KMnO_4} + \underset{\text{cation resin}}{H{:}R} \rightarrow \underset{\text{captured potassium}}{K{:}R} + \underset{\text{permanganic acid}}{HMnO_4}$$

Oxidation of the chromium in the oxide film by permanganic acid is shown in the reaction below. The chromium is converted from the insoluble Cr_2O_3 form to the highly soluble $HCrO_4^-$ form.

Oxidation of Chromium Oxide in Permanganic Acid

$$\underset{\text{chromium oxide}}{Cr_2O_3} + \underset{\text{permanganate}}{2MnO_4^-} + H_2O \rightarrow \underset{\text{bichromate}}{2HCrO_4^-} + \underset{\text{manganese dioxide}}{2MnO_2}$$

Some decomposition of the permanganate also occurs as shown in the following reaction. The manganese dioxide that is formed by these reactions is insoluble and deposits on system surfaces.

Decomposition of Permanganate in Acid Solution

$$\underset{\text{acid}}{4H^+} + \underset{\text{permanganate}}{4MnO_4^-} \rightarrow \underset{\text{manganese dioxide}}{4MnO_2} + \underset{\text{oxygen}}{3O_2} + \underset{\text{water}}{2H_2O}$$

(ii) Reduction step. Addition of a stoichiometric amount of oxalic acid to reduce the remaining $HMnO_4$ to Mn^{2+}. The chemical reaction involved is shown below. The oxalic acid also dissolves deposited MnO_2 as shown in the second reaction below. If 300 ppm of $HMnO_4$ was used, approximately 550 ppm of oxalic acid would be required. There is no cleanup of the water at this point. Step (iii) follows directly.

Dissolution of Residual $HMnO_4$ With Oxalic Acid

$$\underset{\substack{\text{perman-}\\\text{ganate}}}{2MnO_4^-} + \underset{\substack{\text{oxalic}\\\text{acid}}}{5H_2C_2O_4} + 6H^+ \rightarrow \underset{\substack{\text{manganous}\\\text{ion (soluble)}}}{2Mn^{2+}} + \underset{\substack{\text{carbon}\\\text{dioxide}}}{10CO_2} + \underset{\text{water}}{8H_2O}$$

Dissolution of Deposited MnO_2 With Oxalic Acid

$$\underset{\substack{\text{manganese}\\\text{dioxide}}}{MnO_2} + \underset{\substack{\text{oxalic}\\\text{acid}}}{H_2C_2O_4} + 2H^+ \rightarrow \underset{\substack{\text{manganous}\\\text{ion (soluble)}}}{Mn^{2+}} + \underset{\substack{\text{carbon}\\\text{dioxide}}}{2CO_2} + \underset{\text{water}}{2H_2O}$$

(iii) Decontamination step. Addition of an excess of oxalic acid to perform the actual decontamination step. The chemical reaction for dissolution of magnetite in oxalic acid is shown below. Typically about 1,500 ppm is used. In practice, steps (ii) and (iii) are performed as one by the addition of 2,000 ppm of oxalic acid.

Dissolution of Magnetite by Oxalic Acid

$$\underset{\text{magnetite}}{Fe_3O_4} + \underset{\substack{\text{oxalic}\\\text{acid}}}{4H_2C_2O_4} \rightarrow \underset{\substack{\text{ferrous}\\\text{oxalate}}}{3FeC_2O_4} + \underset{\text{water}}{4H_2O} + \underset{\substack{\text{carbon}\\\text{dioxide}}}{2CO_2}$$

After all MnO_4^- has been converted to Mn^{2+} by the oxalic acid, a cation exchange column is placed on line. This serves two functions. Firstly, it removes the Mn^{2+} ions produced by reduction of the $HMnO_4$. Secondly, it removes cationic metal species such as Fe^{2+}, Ni^{2+}, Co^{2+}, etc., which are formed as the oxalic acid dissolves the deposit. These chelate complexes breakdown when passed through strong acid cation resin (H:R). The resin breaks the chemical bond between the metal ion and the chelating anion, captures and retains the metal ion, and releases the chelating agent (oxalate) and a hydrogen ion, which are returned to the system to dissolve more deposits. The oxalic acid is regenerated as shown below.

Regeneration of Ferrous Oxalate by Cation Resin

$$\underset{\substack{\text{ferrous}\\\text{oxalate}}}{FeC_2O_4} + \underset{\substack{\text{cation}\\\text{resin}}}{2H{:}R} \rightarrow \underset{\substack{\text{captured}\\\text{metal}}}{Fe{:}R_2} + \underset{\substack{\text{oxalic}\\\text{acid}}}{H_2C_2O_4}$$

(iv) Decomposition/Cleanup step. Removal of the oxalic acid on anion exchange resin, or decomposition of the oxalic acid by hydrogen peroxide and UV light. The original CORD process used ion exchange resins for the cleanup step, but a recent innovation by KWU/Siemens is the use of hydrogen peroxide and intense UV light to decompose the oxalic acid to carbon dioxide and water. (8) The chemical reactions involved for both methods are shown below. The carbon dioxide generated in the second reaction is vented off leaving only the metal cations to be removed by IX resin. This greatly reduces the amount of resin required for reagent removal. A small amount of mixed bed resin is required for final polishing. For a 10,000 gallon system using 1,500 ppm of oxalic acid, this technology results in the saving 25 - 60 ft^3 of anion resin depending on whether high capacity weak base resin or lower capacity strong base resin is used. For a FSD of an 80,000 gallon system involving multiple CORD cycles, it can result in saving approximately 700 ft^3 of ion exchange resin. Not only is significantly less waste IX resin generated, but also the problem of chelates in the waste is avoided.

Removal of Oxalic Acid by Anion Resin

$$\underset{\text{acid}}{2H^{+}} + \underset{\substack{\text{oxalate}\\ \text{anion}}}{C_2O_4^{2-}} + \underset{\text{resin}}{2R{:}OH} \rightarrow \underset{\substack{\text{captured}\\ \text{oxalate}}}{R_2{:}C_2O_4} + \underset{\text{water}}{2H_2O}$$

Decomposition of Oxalic Acid by Hydrogen Peroxide and UV Light

$$\underset{\substack{\text{hydrogen}\\ \text{peroxide}}}{H_2O_2} + \underset{\substack{\text{oxalic}\\ \text{acid}}}{H_2C_2O_4} \xrightarrow[\text{UV}]{} \underset{\substack{\text{carbon}\\ \text{dioxide}}}{2CO_2} + \underset{\text{water}}{2H_2O}$$

The process described above is a single CORD cycle. Most applications employ three to five such cycles. A typical CORD cycle takes 4-16 hours if IX cleanup is employed, and 10-36 hours if H_2O_2/UV decomposition is used. For the first CORD cycle, the oxidizing step with permanganic acid may be omitted if there is known to be an outer layer of deposit that contains very little chromium. This is typically the case in BWRs.

The CORD D (for Decommissioning) UV (for exposure to ultraviolet light) is similar to the basic CORD process described above, except in Step (iii), the Decontamination Step. After addition of the oxalic acid, the solution is exposed to an intense UV light source for the duration of step (iii). This results in a reduction of the solution redox potential from ~+100 mV (versus Ag/AgCl) to ~–275 mV (versus Ag/AgCl) and significantly increases the corrosivity of the solution. Base metal attack occurs as a result. Up to 10 µm of base metal dissolution may occur, depending upon the number of cycles applied, resulting in very high DFs, possibly as high as ~1,000 which may make the material suitable for free release. The mechanism by which UV light brings about such a significant reduction in redox potential remains proprietary to Siemens

For the CY application, two cycles of CORD were proposed to remove the bulk of the contaminated oxide film, followed by one or two cycles of CORD D UV to remove some base metal giving the very high DFs desired by the plant. The same scenario was tested in the laboratory on a CY artifact and gave very good results, i.e., DFs of 30-75 after 3 cycles, and 300-500 after 4 cycles. (10 11)

The “D” option described above was never implemented during this decontamination. Originally it was planned to apply it in Cycle 3 (and Cycle 4, if necessary), but because of problems with the plant demineralizers (see Sections 4 and 5) and the good decontamination factors obtained after two cycles (see Section 8), the decontamination was terminated after two cycles.

The CORD D UV concept is summarized in Table 3.2 below. A description is also available in the literature. (12) The only three chemicals the vendor brings to site for a decontamination are permanganic acid (2-4 wt% aqueous solution), oxalic acid dihydrate (solid), and hydrogen peroxide (30-50 wt% aqueous solution). For some applications solid potassium permanganate is brought to site from which permanganic acid is produced in a separate IX module prior to addition to the system, or in-situ using the system IX columns to remove the K^+ ions thus converting potassium permanganate to permanganic acid.

Table3.2
The CORD D UV Concept

Feature	Advantages
Four blended steps: Oxidation (of Cr by $HMnO_4$) Reduction (of $HMnO_4$ by oxalic acid) Decontamination (with oxalic acid) Decomposition (of oxalic acid by H_2O_2/UV)	No need to cleanup or drain system between steps Simplifies operation Shorter time Less waste Oxidation step can be omitted for first cycle
Oxidation step uses permanganic acid	Faster and more efficient than AP and NP Less waste since lower concentration, no K^+, and no additives such as NaOH or HNO_3
Reduction step performed in-situ with oxalic acid, the same chemical used for the decontamination step	Avoids contact of IX resin with permanganate ion Stoichiometric calculation unnecessary since large excess added so that decontamination step begins as soon as reduction of $HMnO_4$ is complete
Decontamination step is regenerative using continuous flow through cationic IX resin UV light reduces system redox potential and causes base metal corrosion resulting in much higher DFs	Mn^{2+} ions from reduction step are removed Continuous removal of dissolved contaminants so radiation fields remain low around equipment Reagent is, in effect, reused Effective concentration much higher than low initial concentration suggests UV light can be left off for first 2 cycles to minimize base metal corrosion and dissolve only the oxide film
For full system application, reactor does not necessarily have to be defueled	Simplifies operation Fuel is decontaminated Reduces rate of recontamination
Decomposition of oxalic acid by H_2O_2 and UV light	Decomposition products are innocuous (CO_2 and H_2O) No anion exchange resin required for reagent removal No chelates in waste
Dissolves most metal oxides found in water-cooled reactors	Can be used in all types of water-cooled reactors Separate pretreatment step not required - it is integral in the process
Each CORD cycle is independent	Number of cycles can be adjusted according to final dose rate or DF required
Decontamination factors range from 10 to >1,000, depending on number of cycles and whether basic CORD or CORD D UV is applied	Metal can be cleaned to free release limits, if required.

4

DECONTAMINATION OPERATIONS

Maine Yankee

The decontamination was performed in two separate applications using temporary equipment external to the station systems. No station equipment (e.g., pumps, heaters, demineralizers, etc.) was operated to support the decontamination. The flow path for Application 1 included the CVCS, portions of the RCS (primarily Loop #2 piping and the hot leg of Loop #3), and the pressurizer along with its spray lines. The flow path for Application 2 included the RHRS and the RCS in total. Some portions of the RCS system were common to both applications to facilitate flow through the RHRS and back to the decontamination equipment. The flow paths for both applications are shown in Figures 4.1, 4.2A, and 4.2B and are described briefly below.

An overall schedule for key decontamination tasks on site is presented in Table 4.1.

Table 4.1
Maine Yankee
Site Decontamination Schedule

Description of Project Step	Start Date	Finish Date	Step Duration (days)	Project Duration (days)
Receipt of Vendor Equipment On Site	1/26/98	2/2/98	8.0	8.0
Setup & Test Vendor Equipment	1/27/98	2/10/98	15.0	16.0
Install and Test Nozzle Dams	2/3/98	2/7/98	4.0	16.0
Prepare & Heatup Plant Systems	2/10/98	2/14/98	4.0	20.0
Perform Application 1	2/14/98	2/23/98	8.5	28.5
Prepare Systems for Application 2	2/23/98	2/26/98	3.5	32.0
Perform Application 2	2/27/98	3/6/98	7.5	39.5
Final Cleanup and MnO_2 Removal	3/6/98	4/3/98	27.5	67.0

Application 1

With the cavity filled, and the nozzle dams in place and pressurized, as much of the liquid in the CVCS, RCS and RHRS as possible was drained. This step was necessary to minimize boron levels prior to the start of the decontamination. Station valve alignments were then setup for refilling the systems and approximately 38,000 gallons of demineralized water was added to them. Leak and flow checks of the RCS, CVCS, and RHRS were then successfully performed and final valve alignments for the Application 1 flow path completed. The circulating volume for the Application 1 flow path was determined to be approximately 18,000 gallons. The flow path is shown schematically in Figure 4.1.

To commence Application 1, a flow rate of approximately 300 gpm was initiated at the circulating pump in the temporary equipment. Heatup to operating temperature (195°F ± 10°F) was then performed over the next 15 hours and a final pre-decontamination walkdown performed. During this period the boron concentration was reduced to approximately 15 ppm using the mixed bed resin columns. Eleven cycles using the DFD process were then performed on the CVCS and portions of the RCS over a period of about nine days.

The flow was directed into the station systems via two flow paths. The first passed from the decontamination equipment through interconnecting lines to the manway jumper on the steam generator in Loop #3. With the Loop #3 cold leg loop stop valve shut, this flow was directed through the hot leg of Loop #3 into the spider assembly at the RPV. From the spider, it proceeded into the cold leg of Loop #1. With the cold leg, loop stop valve of Loop #1 also shut, flow passed into the pressurizer spray and letdown lines. To protect the nozzle dams, pressure as measured at the temporary connections on the Loop #3 steam generator manways was limited to less than 75 psig.

Letdown flow was directed through the CVCS system piping and components and returned to the RCS on the cold leg sides of Loops #2 and #3. Due to the relatively high pressure drop through this system and the limiting nozzle dam pressure, the maximum CVCS flow achievable was approximately 50 gpm. This fluid was then passed to the suction of the decontamination pump through the RCS piping on Loops #2 and #3, the vessel spider assembly and the connection on the Loop #2 steam generator manway. During the first six phases of Application 1, the 4,000-gallon volume control tank (VCT) remained full. At the end of Cycle 6, the VCT was emptied. A portion of this liquid (approximately 2,400 gallons) was drained through ion exchange to the station aerated drain tank (ADT). The remainder of the VCT liquid was transferred to the pressurizer. The purpose of this exercise was to minimize fluid heat loss from extended residence time in the VCT and lower the dose rate from a hot-spot at the bottom of the tank.

The pressurizer spray flow was also split. A portion passed into the pressurizer vessel and returned to the hot leg of Loop #1. The remainder flowed back to the RCS into the cold leg of Loop #2. The pressurizer spray fluid also returned to the decontamination equipment in the same manner as the CVCS flow.

The second flow path from the decontamination equipment connected to the RCS loop drain header at valve DR-12. Fluid was directed back to the RCS and the decontamination equipment through the RCS drain and fill lines for Loop #2, as well as the high pressure drain cooler and the seal water heat exchanger. The cooler and heat exchanger flow was routed through the charging piping where it joined the CVCS flow at the outlet of the VCT. During Cycle 9, the flow through the drain cooler and seal water heat exchanger was stopped and all of the flow directed through the Loop #2 drain/fill lines. This change was made to improve the flow through the bypass line in RCS Loop #2.

Gas generation occurred in the system during each cycle from both the destruction of oxalic acid and the corrosion of base metal. The bulk of this gas was trapped in the tubes of the steam generators and resulted in a gradual movement of liquid from the steam generators in Loops #2 and #3 to the pressurizer. Because the steam generator in Loop #2 was at a lower operating pressure than that in Loop #3, more of the gas collected in the Loop #2 generator. When the liquid in the steam generator of Loop #2 dropped to the level of the bottom of the tubesheet, further fluid transfer was minimized by venting the gas from the system through temporary connections at the generators. During the period of the fluid transfer, the level in the pressurizer rose from a reading of approximately 10% to about 30%. During Cycle 5, approximately 1,800 gallons of fluid were drained from the system through ion exchange to reduce the level in the pressurizer.

Application 2

In preparation for Application 2, station valves were realigned, ion exchange resin was sluiced to high integrity containers (HICs), the filter cartridges were changed, the boron concentration was reduced from 422 ppm to 35 ppm, and the system was heated to an operating temperature of 195°F ± 10°F. These steps were performed over a period of approximately 3½ days. The circulating volume for the Application 2 flow path was determined to be approximately 22,000 gallons.

To commence Application 2, a flow rate of approximately 650 gpm was initiated at the circulating pump in the temporary equipment. This was higher than that for Application 1 because the system pressure drop was much lower. Thirteen cycles using the DFD process were then performed on the RHR pumps and piping and the remaining portions of the RCS over a period of about eight days.

The Application 2 flow path included the RHRS as well as the remainder of the RCS piping not exposed during Application 1. Flow was initiated at the circulating pump in the temporary equipment and directed into the station systems via two flow paths. This is shown schematically in Figure 4.2A. In the first, flow passed through interconnecting lines to the manway jumper on the steam generator in Loop #3. With the Loop #3, hot leg, loop-stop valve shut, flow was directed through the cold leg and bypass lines of Loop #3. This fluid was then passed through the spider assembly at the RPV and into the RHRS inlet through the hot leg of Loop #2.

RHRS flow was directed through the pumps and piping of the system bypassing the heat exchangers and was then returned to the RCS through the normal connections on the cold leg sides of Loops #1 and #2. This flow was then passed back to the suction of the decontamination pump through Loop #1 (in the reverse direction to normal flow), the vessel spider assembly, the cold leg piping of Loop #2 and the Loop #2 steam generator manway connection.

The second flow path directed the remaining fluid from the discharge of the decontamination pump, through a temporary interconnecting line to the inlet of the RCS drain header at valve DR-12. The liquid was then passed into the Loop #1 and #3 drain/fill lines where it joined the first flow path and returned to the decontamination equipment.

These flow paths were maintained for the first five cycles of Application 2. At the start of Cycle 6, flow through the RHRS was reduced by closing valve LSI-M-11, which had been directing flow back to the Loop #1 cold leg. To compensate for this reduction, valve RC-M-31 on the hot leg of Loop #3 was cracked open. This redirected a portion of the flow from the Loop #3 hot leg and bypass piping into Loop #1 through the spider assembly.

During Cycle 8, the valve adjustments performed for Cycles 6 and 7 were returned to their Cycle 1 positions.

At the end of Cycle 8, flow through the RHRS was terminated by closing valve RH-M-1, the inlet to the RHR on the hot leg of Loop #2. In addition, the cold leg Loop #2 stop valve, RC-M-21, was cracked open just enough to provide a flow path back to the decontamination equipment through the hot leg of Loop #2.

During Cycles 9 through 12, periodic flow reversals were performed in the RCS piping to improve solvent access to system surfaces. This flow path is shown schematically in Figure 4.2B.

For Cycle 13, the RHR system was returned to the flow path. Near the end of this cycle the heat exchangers were placed on line to flush residual chemicals from potential deadlegs.

During the cleanup phase of Application 2, a number of flow path variations were introduced to access as many deadlegs as possible and maximize flow through each individual section of the in-line piping and equipment.

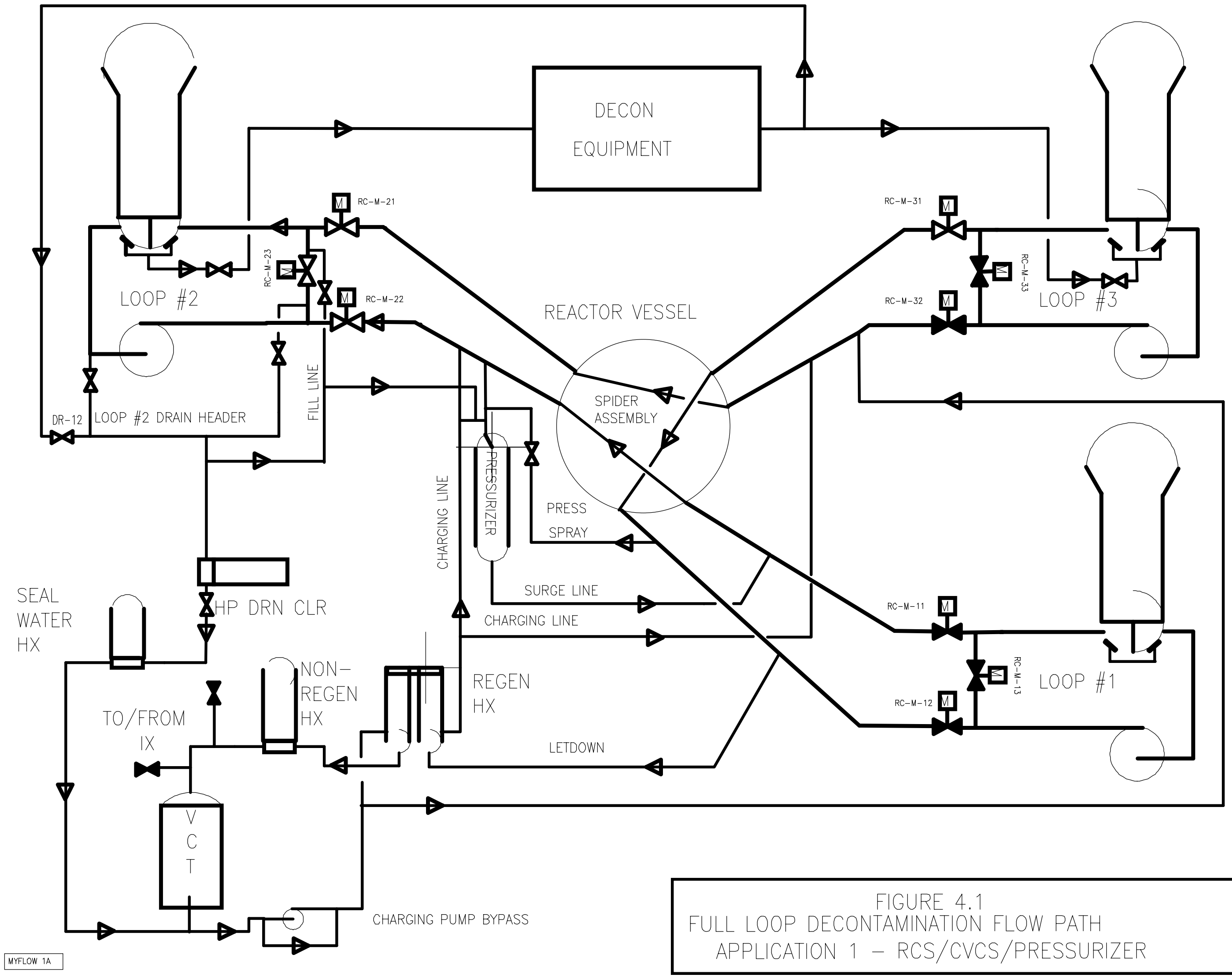

FIGURE 4.1
FULL LOOP DECONTAMINATION FLOW PATH
APPLICATION 1 – RCS/CVCS/PRESSURIZER

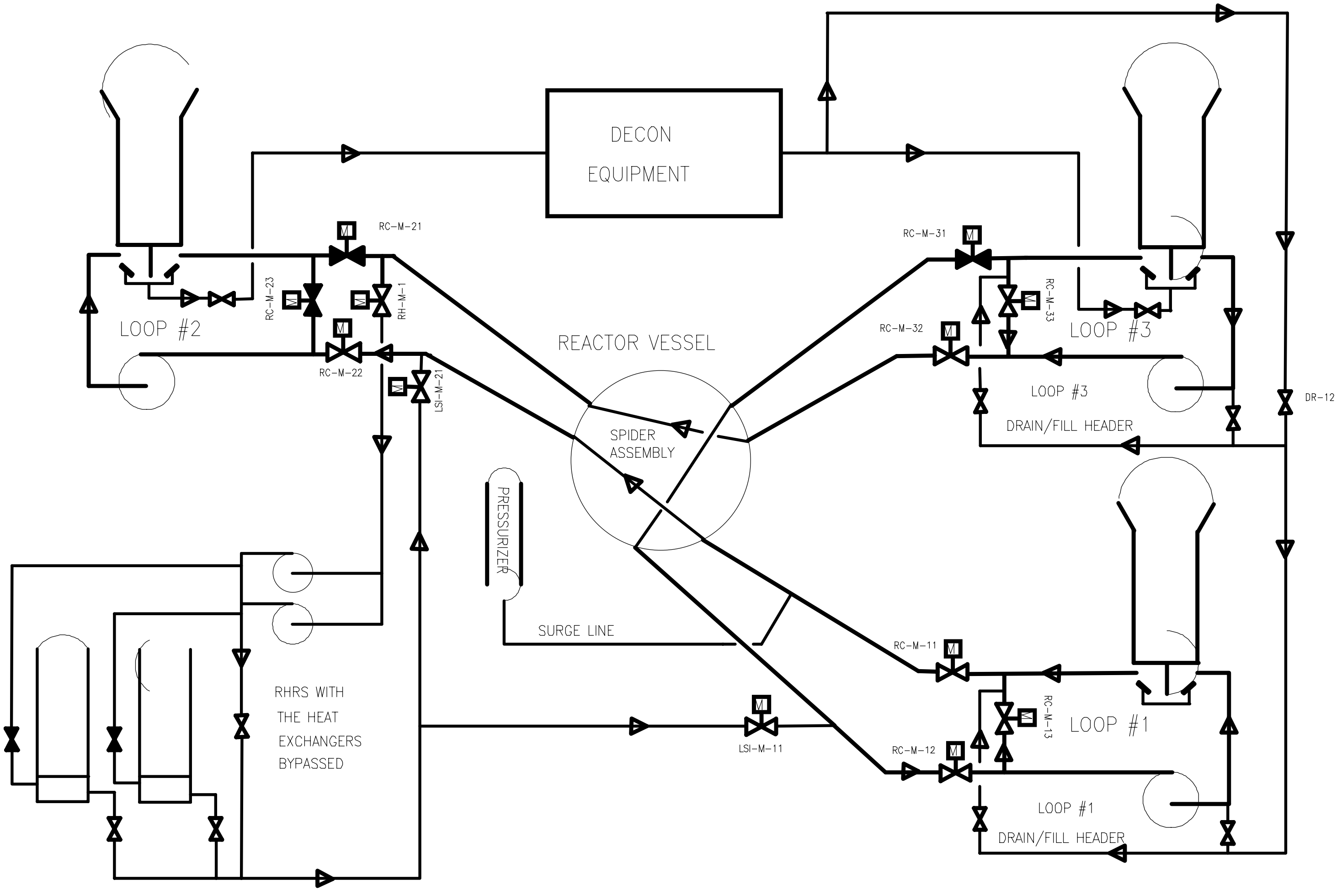

FIGURE 4.2 A
FULL LOOP DECONTAMINATION FLOW PATH
APPLICATION 2A – RCS/RHRS

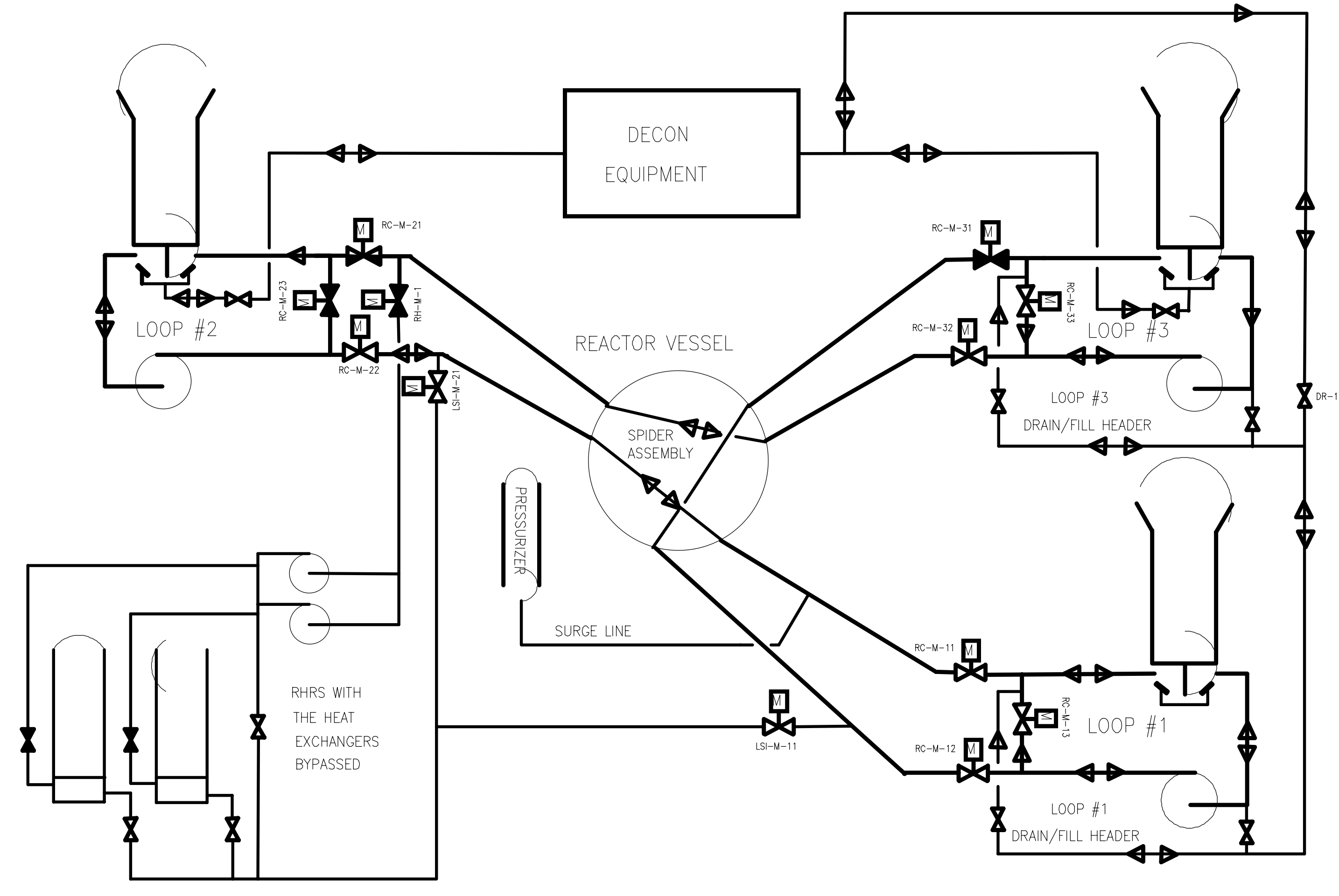

FIGURE 4.2 B
FULL LOOP DECONTAMINATION FLOW PATH
APPLICATION 2B – RCS/RHRS (RHRS ISOLATED)

Connecticut Yankee

Unlike Maine Yankee, the decontamination was performed in a single application using a combination of existing plant equipment as well as temporary equipment external to the station systems. An RHR pump, the pressurizer heaters, and the plant demineralizers were used to support decontamination operations. The temporary vendor-supplied equipment was added between the discharge of the RHR pump and the line added (Mod #1) to the pressurizer. The flow path for this application is shown in Figure 4.3. The operations required to complete the decontamination are described briefly below. An overall schedule for key decontamination tasks on site is presented in Table 4.2.

Table 4.2
Connecticut Yankee
Site Decontamination Schedule

Description of Project Step	Start Date	Finish Date	Step Duration (days)	Project Duration (days)
Receive, Install and Test Nozzle Dams	5/29/98	6/25/98	27	27
Receipt of Vendor Equipment On Site	6/17/98	6/30/98	13	32
Setup & Test Vendor Equipment	6/19/98	7/6/98	17	36
Prepare & Heatup Plant Systems	7/7/98	7/25/98	18	57
Perform Cycle 1	7/26/98	8/8/98	13	71
Prepare for Cycle 2	8/8/98	8/10/98	2	73
Perform Cycle 2	8/10/98	8/21/98	11	84
Deadleg Flushing & Final Cleanup	8/21/98	9/28/98	38	122
Dismantle & Package Equipment	8/24/98	9/3/98	10	
Decon Equipment Off Site	9/22/98	9/23/98	2	
Nozzle Dams Removed	10/20/98	10/23/98	3	

System operations commenced at 04:20 on July 25 after the system was filled, vented and aligned for the startup of the 'A' RHR pump. During initial operations, a leak-check was performed and RHR flow raised to approximately 2,000 gpm. The Siemens' equipment was then valved into the system and a second leak check was performed. Flows through the letdown and spent fuel pool purification loops were initiated and heatup of the system to 200°F was commenced. During this period, some delays occurred due to screen plugging, faulty valves and filter/resin trap differential pressures. When corrected, Step 1 (Oxidation) of Cycle 1 commenced at 22:00 hours on July 26, with the addition of 210 gallons of permanganic acid solution. This step was terminated after about 12 hours and Step 2 (Reduction) commenced with the addition of 882 lbs. of oxalic acid. The system was recirculated for approximately one hour in this configuration. At this point, the letdown ion exchange column was being placed into service to initiate Step 3 (Decontamination) when problems were experienced in attempting to get flow through the ion exchange column. After system leakage in the vicinity of the columns was identified, the letdown system was isolated. It was determined that about 1,000 gallons of decontamination fluid had leaked from the system. A detailed root cause investigation was performed in parallel with decontamination cleanup operations. The cause of the leak was determined to be a blockage in the system likely caused by a valve out of position.

The decontamination was resumed approximately 6 hours after the leak (17:30 on July 27), when flow was initiated through the ‘C’ ion exchange column at a rate of ~63 gpm. It continued in this configuration until 20:00 on July 29. At this time, the spent fuel ion exchanger was isolated based upon the quantity of transuranic isotopes removed on the ‘C’ ion exchange column. This step placed the decontamination on hold because the necessary repairs and testing to place the letdown ion exchange back into service were not yet complete. As a result, circulation at 200°F continued without ion exchange until 22:20 on August 6 when the letdown (‘A’ column) cation exchange was placed on line at ~150 gpm. This continued for about one day (~26 hours) when the ‘A’ column was isolated and the ‘D’ mixed bed column put on line at the same flow rate. While the cation (‘C’ and ‘A’) ion exchange columns were on line, 30% hydrogen peroxide (~60 gallons) was added to the system (Step 4) and the UV lamps operated to decompose the oxalic acid remaining in the system. At 15:25 hours on August 8, Cycle 1 was completed and the ‘D’ ion exchange column was isolated from the system.

To commence Step 1 of Cycle 2, RHR flow was adjusted to approximately 1800 gpm. At 03:15 on August 10, approximately 240 gallons of permanganic acid solution were then added to the system. Step 1 was terminated 6 hours later, after chromium levels had stabilized. Step 2 commenced at 10:00 on August 10 with the addition of 882 lbs. of oxalic acid to the system. Regeneration through ion exchange started between 12:00 and 12:30 to commence Step 3 when a flow of ~63 gpm was initiated through the ‘C’ ion exchange column and ~145 gpm through the ‘A’ column. Flow was then raised to 90 gpm through the ‘C’ bed at 14:00 hours. Decomposition of the oxalic acid commenced at 00:50 on August 11. At 14:00 hours the ‘C’ ion exchange column was taken off line due to high radiation levels in the outlet line from the column. These elevated levels were caused by resin that had passed through both the internal screen and the post IX filter/resin trap and into the column’s outlet line. The cause of this resin release was eventually traced to an existing flaw in the internal screen and dissolution of the resin-retaining screen in the resin trap.[h] The resin trap had been manufactured prior to the start of Cycle 1 due to a problem with high differential pressure. It was determined that ~25-30 ft^3 of resin (out of a total of 45 ft^3) was released from the column. During this period, flow to the ‘A’ ion exchange column was maintained at ~145 gpm.

Flow to the ‘A’ ion exchange column stopped at 12:35 on August 12, when the booster pump tripped due to electrical problems. An attempt to restart the pump was made at 16:05 on August 13 but leaks developed in the discharge line from the booster pump and in the letdown piping downstream of the IX columns. After repairs had been completed, flow to the ‘B’ ion exchange column was initiated at 14:45 on August 18. This continued for 16 hours to remove the remaining concentrations of iron and cobalt. Final cleanup was initiated at 20:00 on August 19 when column ‘D’ was placed on line. It was replaced with fresh cleanup resin in column 'B' at 00:00 on August 21 (see Table 7.2 for details). The target final cleanup parameter of <0.5 ppm

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[h] Connecticut Yankee Root Cause Investigation ACR 98-0698 RCS Chemical Decontamination Cycle 2 Spent Fuel Pool Ion Exchanger SFP IX-1-1C Transient

chromium in the system was met by 10:00 on August 21 and from the vendor’s perspective the decontamination was complete at this time. Flushing of deadlegs, the 'A' IX column and the outlet line of IX column 'C' commenced shortly thereafter. Periodic cleanup with mixed bed resin was required after these operations to bring the Cr level back to <0.5 ppm. It tended to increase to a “few ppm” each time a line or IX column was flushed. These operations were performed by plant personnel on a non-critical path basis over a period of several weeks. The RHR pump was turned off at 12:10 on September 28 after all flushing operations had been completed and the final equilibrium dissolved Cr concentration was <0.5 ppm. This marked the official completion of the decontamination from the plant’s perspective. The nozzle dams were not removed immediately. They were left in place for another four weeks to keep the RPV isolated from the loops while the lines to the pressurizer were cut and capped. This operation was completed by October 20 and the nozzle dams were removed at that time, as shown in Table 4.2.

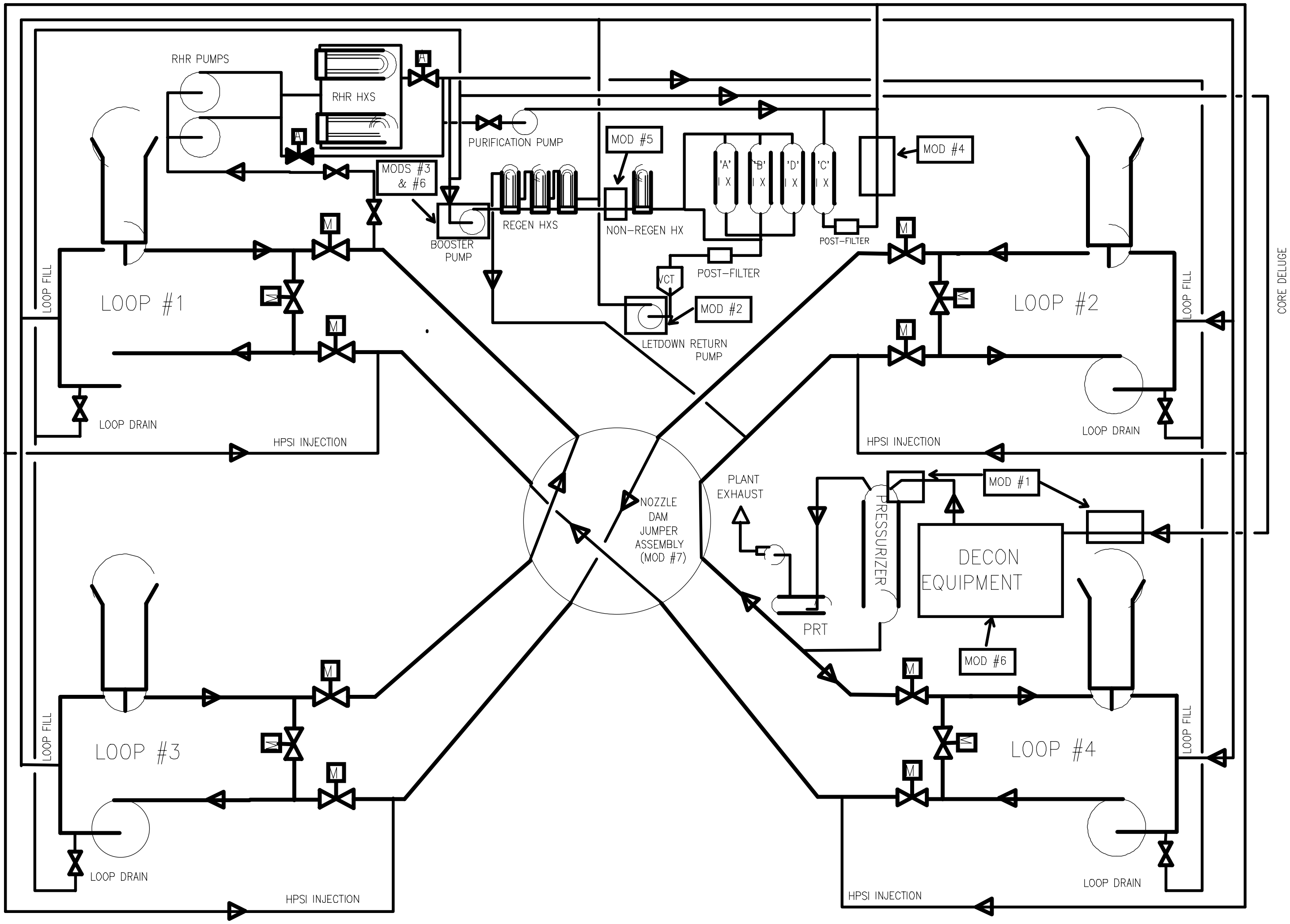

FIGURE 4.3 – CONNECTICUT YANKEE
RCS FULL LOOP DECONTAMINATION FLOW PATH

5

DECONTAMINATION CHEMISTRY

Maine Yankee

Application 1

A pre-decon sample taken on February 13, 1998 indicated a boron concentration of 114 ppm. This was higher than the recommended limit of 40 ppm, so the anion IX columns were placed on line to reduce it. It is estimated that 20 ft^3 of resin capacity was consumed in reducing the boron level to 14 ppm.

Chemical operations for Application 1 started at 20:15 on February 14, 1998 when 69 L of fluoroboric acid solution (48 wt%) was added to the system. This quantity was based on an assumed system volume of 15,000 gallons. However, subsequent pH measurements and boron concentration data indicated that the system volume was approximately 17,900 gallons, thus an additional 20 L was added on February 15. This resulted in a (calculated) fluoroboric acid concentration of 9.7 mM and a pH of ~2.2 during the fluoroboric acid-only phase. Both of these parameters were within specification.

Over the next nine days, 11 cycles of DFD were applied as described in Section 3 of this report. A summary of the metals and (-emitting radionuclides removed during the application is presented in Table 5.1. The cumulative data is shown graphically in Figure 5.1. A total of 57.5 curies of radioactivity was removed in the application. The vast majority (97%) was Co-60. The "others" category included small amounts of Mn-54, Co-58, Zr-95, Nb-95 and Sb-125. Sb-125 was the largest contributor to this category. In solution it exists as the antimonate ion (SbO_3^-) and is only removed during the cleanup phase when anion exchange resin is placed online. This is the reason for the disproportionately high amount in the "others" column during the cleanup phase.

Metals removed during the application are also given in Table 5.1. A total of 309.8 pounds of metal was dissolved. This includes metals from both oxide film dissolution and base metal corrosion. Since film dissolution and base metal corrosion occur concurrently, there is no easy method to differentiate between the two sources of metals. However, as will be shown in Section 6, if it is assumed that 75 lbs. of the metal was from oxide film dissolution and the balance from base metal corrosion, a reasonable correlation between measured and calculated corrosion is obtained. This amount of metal was converted to oxide film by dividing by 0.73, the approximate fraction of metal in a typical oxide film. Table 5.2 gives a summary of total

(-emitting radionuclides and total oxide film removed during each DFD application. The calculated specific activity of the oxide film is also given.

Figure 5.2 shows the specific activity of the material removed in each cycle. This is calculated from the total (-emitting radionuclides removed in the cycle divided by the total amount of metals (Fe + Ni + Cr). A small amount of low specific activity "sludge" was removed during initial circulation of the HBF_4. Given the small amount of activity removed relative to the amount of metals, it is unlikely that this represents dissolution of the adherent oxide film. During Cycles 1 and 2, the specific activity of the removed material was much higher indicating removal of the contaminated oxide film. The abrupt drop in specific activity at Cycle 3 suggests that by this time the majority of the oxide film had been removed and the major process now occurring was base metal corrosion.

The specific activity slowly dropped between Cycles 3 and 11 as decreasing amounts of radioactivity were removed, while the amount of metal removed remained fairly constant.

Table 5.1
Summary of Metals and Radionuclides Removed During Application 1 of the Maine Yankee DFD

Step	Length of Step (hours)	Metals Removed (pounds)				Radionuclides Removed (curies)		
		Fe	Ni	Cr	Total	Co-60	Others	Total
Initial HBF_4	7.5	2.4	3.7	1.1	7.2	0.8	0.02	0.9
Cycle 1	12.6	9.4	8.7	3.7	21.9	14.1	0.01	14.1
Cycle 2	10.4	7.9	5.6	3.1	16.6	8.6	0.14	8.7
Cycle 3	21.8	16.0	10.8	6.1	32.8	7.4	0.01	7.4
Cycle 4	24.6	19.2	17.9	7.6	44.6	7.9	0.13	8.0
Cycle 5	19.3	15.0	12.7	4.6	32.3	4.7	0.23	4.9
Cycle 6	18.4	14.3	13.2	7.4	34.9	3.7	0.33	4.0
Cycle 7	16.5	11.5	10.9	9.5	32.0	2.9	0.38	3.3
Cycle 8	18.0	11.6	8.8	4.8	25.3	1.9	0.01	1.9
Cycle 9	14.5	7.7	5.5	4.3	17.5	1.2	0.14	1.2
Cycle 10	13.5	8.1	6.5	5.1	19.8	1.2	0.05	1.3
Cycle 11	13.7	6.3	5.3	3.5	15.1	1.0	0.10	1.1
Cleanup	14.3	1.6	3.6	4.7	9.8	0.3	0.20	0.5
Totals	205.1	131	113.2	65.5	309.8	55.7	1.8	57.5

Table 5.2
Summary of Radionuclides and Oxide Film Removed During Maine Yankee Decontamination

Application	Total Activity Removed (Curies)	Total Metals Removed (lbs.)	Estimated Metals From Corrosion (lbs.)	Oxide Film Removed (lbs.)	Specific Activity of Oxide Film (mCi/g)
1	57.5	309.8	234.8	102.7	1.23
2	45.2	367.5	278.4	121.9	0.82
Total	102.7	677.3	513.2	224.6	1.01

Additional chemical data is shown in graphical form in Appendix A. These graphs include:

Figure A-1	Dissolved Co-60 Concentration During Application 1 (IX in and IX Out)
Figure A-2	Dissolved Iron Concentration During Application 1 (IX in and IX Out)
Figure A-3	Dissolved Nickel Concentration During Application 1 (IX in and IX Out)
Figure A-4	Dissolved Chromium Concentration During Application 1 (IX in and IX Out)

These graphs show that Co-60 was removed with good efficiency (85-95%) throughout the application. Iron started to break through the IX columns at hour 120. This was traced to column F (see Table 7.1 in Section 7) which was receiving preferential flow. It was taken off line and the iron concentration at the IX outlet returned to its normal low value of ~2 ppm. At hour 130 breakthrough of iron was again observed. The remaining three cation columns (A, B, and C) had broken through. Calculations indicated that the beds were essentially 100% spent at this point so they were isolated. The balance of Application 1 was completed using beds D and E. Nickel was removed with excellent efficiency (95-99%) throughout the application. It did not breakthrough when the iron did indicating that it is held more strongly by the resin than is iron. Chromium was removed with relatively poor efficiency (15-20%) throughout the application. This is because in the reduced form, Cr^{3+} forms complexes with fluoride and oxalate. These compound are quite stable and are only partially broken down as they pass through cation resin. As a result the chromium concentration tends to build up in solution as shown in Figure A-4. Only when the anion columns are placed online during the cleanup phase is chromium removed completely. This is obvious from the sharp downturn in chromium concentration at the end of the application.

Chemical usage during the decontamination is summarized in Table 5.3. The totals for Application 1 include 23.5 gallons of concentrated fluoroboric acid solution, 436 pounds of potassium permanganate and 374.5 pounds of oxalic acid dihydrate.

Application 2

A pre-decon sample taken on February 24 indicated a boron concentration of ~420 ppm. This was above the maximum qualified value of 40 ppm, so the mixed bed columns were placed on line to reduce it. However, they became saturated after reducing it to only 250 ppm.[i] This meant that these beds had to be sluiced and refilled. Once replaced, deboration continued until a value of 35 ppm was obtained. It is estimated that about 70 ft^3 of anion resin was used in removing the boron.

Chemical operations for Application 2 started at 22:45 on February 26, 1998 when 105 L of fluoroboric acid solution (48 wt%) was added to the system. This quantity was based on a calculated system volume of 22,000 gallons. This resulted in a pH of 2.11 during the fluoroboric acid-only phase which was identical to that of an 11.8 millimolar solution prepared for lab analysis and which is within specification.

Over the next 8 days, 13 cycles of DFD were applied as described in Section 3 of this report. A summary of the metals and (-emitting radionuclides removed during the application is presented in Table 5.4. The cumulative data is shown graphically in Figure 5.3. A total of 45 curies of radioactivity was removed in the application. The vast majority (97.5%) was Co-60. Small amounts of Mn-54, Co-58, Zr-95, Nb-95, and Sb-125 were also removed. Sb-125 was the largest contributor to this category and was removed during the cleanup phase when the anion exchange resin was placed online.

Table 5.3
Chemical Usage During Maine Yankee DFD

Step	Fluoroboric Acid (48% Solution) (gallons)		Potassium Permanganate (pounds)		Oxalic Acid Dihydrate (pounds)	
	App. 1	App. 2	App. 1	App. 2	App. 1	App. 2
Initial HBF_4	18.2	27.7				2.2
Cycle 1			29.2	36.7	21.0	29.3
Cycle 2	5.3		26.4	38.9	22.0	29.3
Cycle 3			30.4	52.6	26.4	46.4
Cycle 4			42.4	36.7	40.2	29.3
Cycle 5			47.4	43.3	44.9	41.4
Cycle 6			53.0	63.8	41.8	56.1
Cycle 7			41.4	36.7	38.3	29.3
Cycle 8			41.8	56.3	32.1	51.7
Cycle 9			44.0	54.6	36.5	64.2
Cycle 10			40.0	48.2	34.8	44.2
Cycle 11			40.0	39.6	36.5	39.8
Cycle 12			-	50.6	-	47.3
Cycle 13			-	44.0	-	60.5
Subtotals	23.5	27.7	436.0	602.0	374.5	571.0
Grand Totals	51		1,038		946	

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[i] They had previously been used to reduce the boron concentration prior to Application 1 and also for the final cleanup of the Application 1 solvent.

Metals removed during Application 2 are also given in Table 5.4. A total of 367.5 pounds of metal was dissolved. This includes metals from both oxide film dissolution and base metal corrosion. A detailed corrosion analysis was not done for Application 2, so an estimate of the distribution of metal removed by corrosion and oxide film dissolution was not obtained in this manner. However, by assuming the same fraction of the total metal dissolved as in Application 1 was from oxide film dissolution, an estimate of the distribution was obtained. The results are given in Table 5.2.

Figure 5.4 shows the specific activity of the material removed in each cycle. This is calculated from the total (-emitting radionuclides removed in the cycle divided by the total amount of metals (Fe + Ni + Cr). As in Application1, a small amount of low specific activity "sludge" was removed during initial circulation of the HBF_4. Given the small amount of activity removed relative to the quantity of metals, it is unlikely that this represents dissolution of the adherent oxide film. During Cycle 1, the specific activity of the removed material was much higher indicating removal of the contaminated oxide film. The abrupt drop in specific activity by a factor of about 2 at Cycle 2 suggests that by this time the majority of the oxide film had been removed and the major process now occurring was base metal corrosion. Due to the many variations in system flow path over the 13-cycle application, particularly between Cycles 6 and 13, it is very difficult to correlate the changes in specific activity from one cycle to the next.

Additional chemical data from Application 2 is shown in graphical form in Appendix B. These graphs include:

Figure B-1	Dissolved Co-60 Concentration During Application 2 (IX in and IX Out)
Figure B-2	Dissolved Iron Concentration During Application 2 (IX in and IX Out)
Figure B-3	Dissolved Nickel Concentration During Application 2 (IX in and IX Out)
Figure B-4	Dissolved Chromium Concentration During Application 2 (IX in and IX Out)

These graphs show that Co-60 was removed with good efficiency (85-95%) throughout the application. Iron started to break through the IX columns at hour 75 during Cycle 8. Columns A, B, and C were switched over to D, E and F and the IX out iron concentration returned to its normal low value. At hour 140 breakthrough of iron was again observed. Columns D, E, and F were essentially 100% spent at this point so they were isolated and replaced with A, B, and C. These columns had been sluiced and refilled with fresh resin. The balance of Application 2 was completed using these columns. Similar to Application 1, nickel was removed with excellent efficiency (95-99%) throughout Application 2. Chromium was again removed with relatively poor efficiency (10-30%) on cation resin but was completely removed during the cleanup phase.

Chemical usage during Application 2 is summarized in Table 5.3. In total, 27.7 gallons of concentrated fluoroboric acid solution, 602 pounds of potassium permanganate and 571 pounds of oxalic acid dihydrate were used.

Total chemical usage for both Application 1 and 2 was as follows:

Fluoroboric Acid:	51 gal
Potassium Permanganate:	1,038 pounds
Oxalic Acid Dihydrate:	946 pounds

Table 5.4
Summary of Metals and Radionuclides Removed During Application 2 of the Maine Yankee DFD

Step	Length of Step (hours)	Metals Removed (pounds)				Radionuclides Removed (curies)		
		Fe	Ni	Cr	Total	Co-60	Others	Total
Initial HBF_4	3.5	2.1	1.5	0.7	4.3	0.4	0.0	0.4
Cycle 1	8.0	9.6	8.9	3.1	25.8	7.8	0.16	8.0
Cycle 2	7.0	6.8	6.5	2.7	16.0	3.1	0.0	3.1
Cycle 3	15.5	15.2	14.4	5.7	35.3	5.4	0.1	5.5
Cycle 4	5.5	5.1	4.4	2.7	12.2	2.0	0.02	2.0
Cycle 5	12.5	11.3	11.3	5.7	28.3	4.2	0.01	4.2
Cycle 6	12.0	10.8	11.9	3.6	26.3	3.9	0.08	4.0
Cycle 7	7.0	5.8	6.1	3.7	15.6	1.1	0.03	1.1
Cycle 8	16.5	10.3	12.7	5.0	28.0	1.7	0.04	1.7
Cycle 9	23.5	19.2	21.1	7.6	47.9	4.6	0.0	4.6
Cycle 10	14.25	12.6	14.0	4.4	31.0	2.1	0.03	2.2
Cycle 11	12.75	10.7	11.5	4.6	26.8	2.1	0.00	2.1
Cycle 12	18.5	10.2	11.2	5.9	27.3	1.8	0.9	2.7
Cycle 13	26.0	14.1	12.3	5.4	31.8	2.7	0.1	2.8
Cleanup	18.5	3.4	1.2	6.3	10.9	0.5	0.3	0.8
Totals	201	147.2	149	67.1	367.5	43.4	1.8	45.2

Figure 5.1
Maine Yankee - DFD Application 1
Metals and Activity Removal

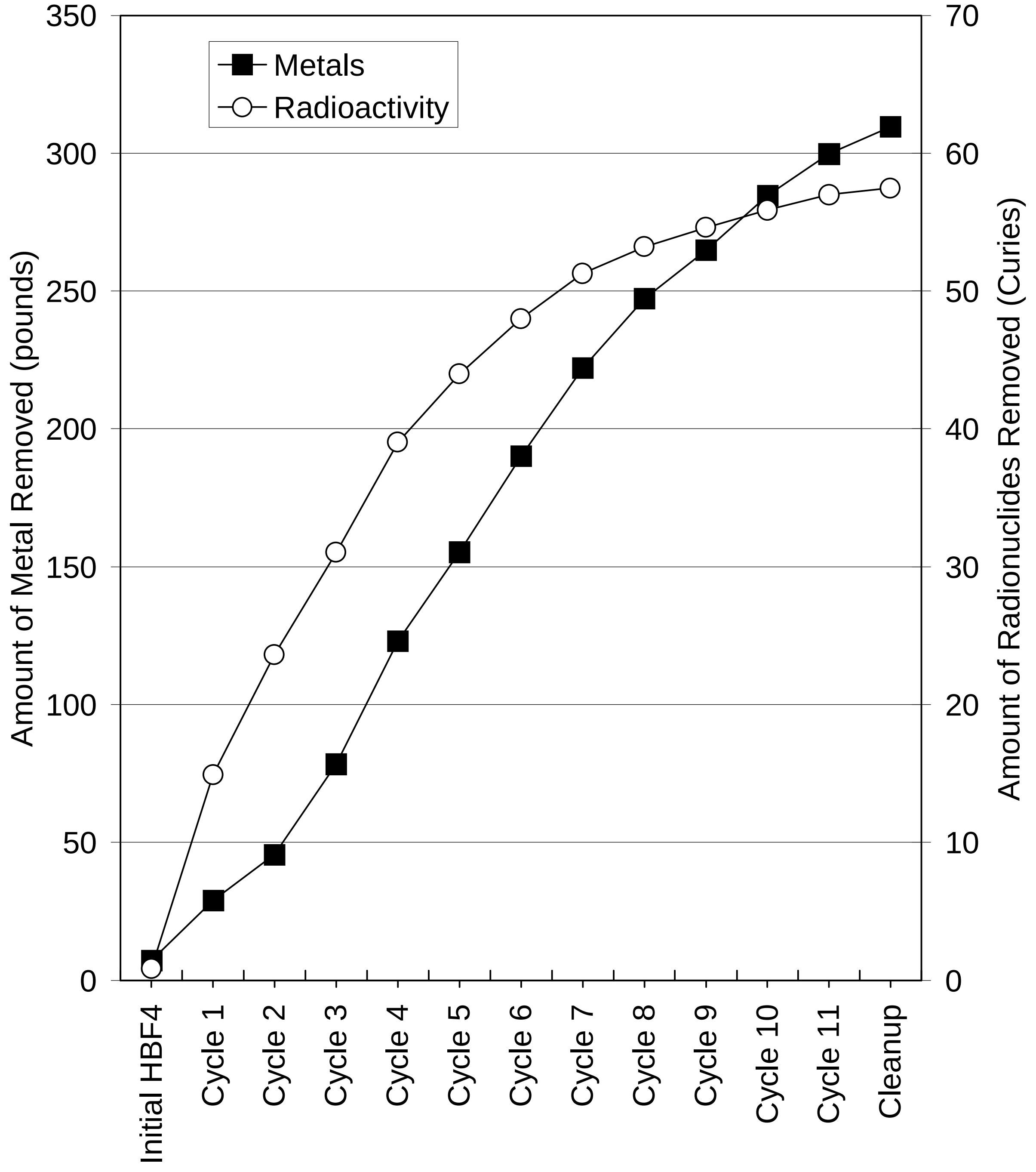


Figure 5.2
Maine Yankee - DFD Application 1
Specific Activity of Material Removed in Each Cycle

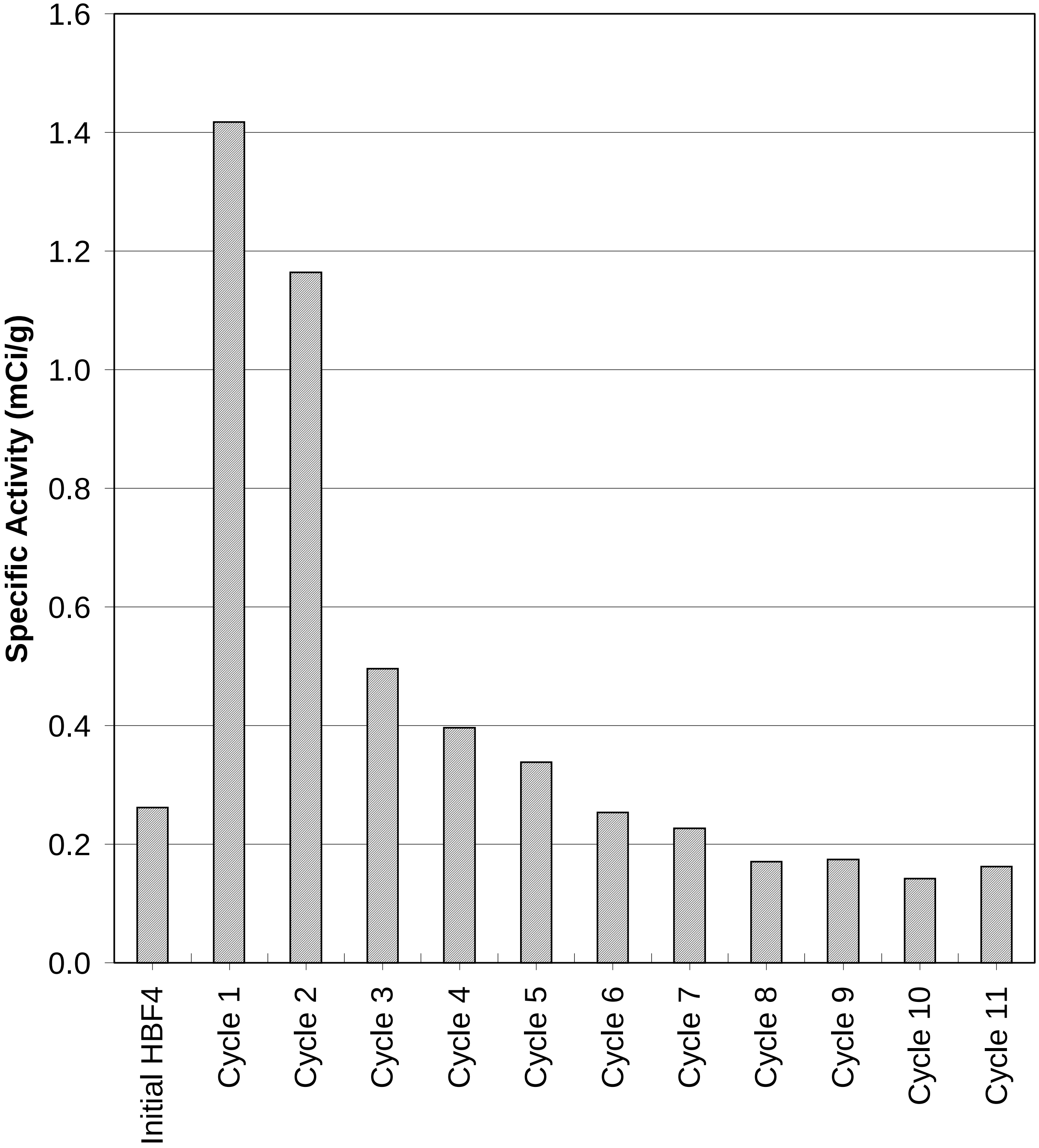


Figure 5.3
Maine Yankee - DFD Application 2
Metals and Activity Removal

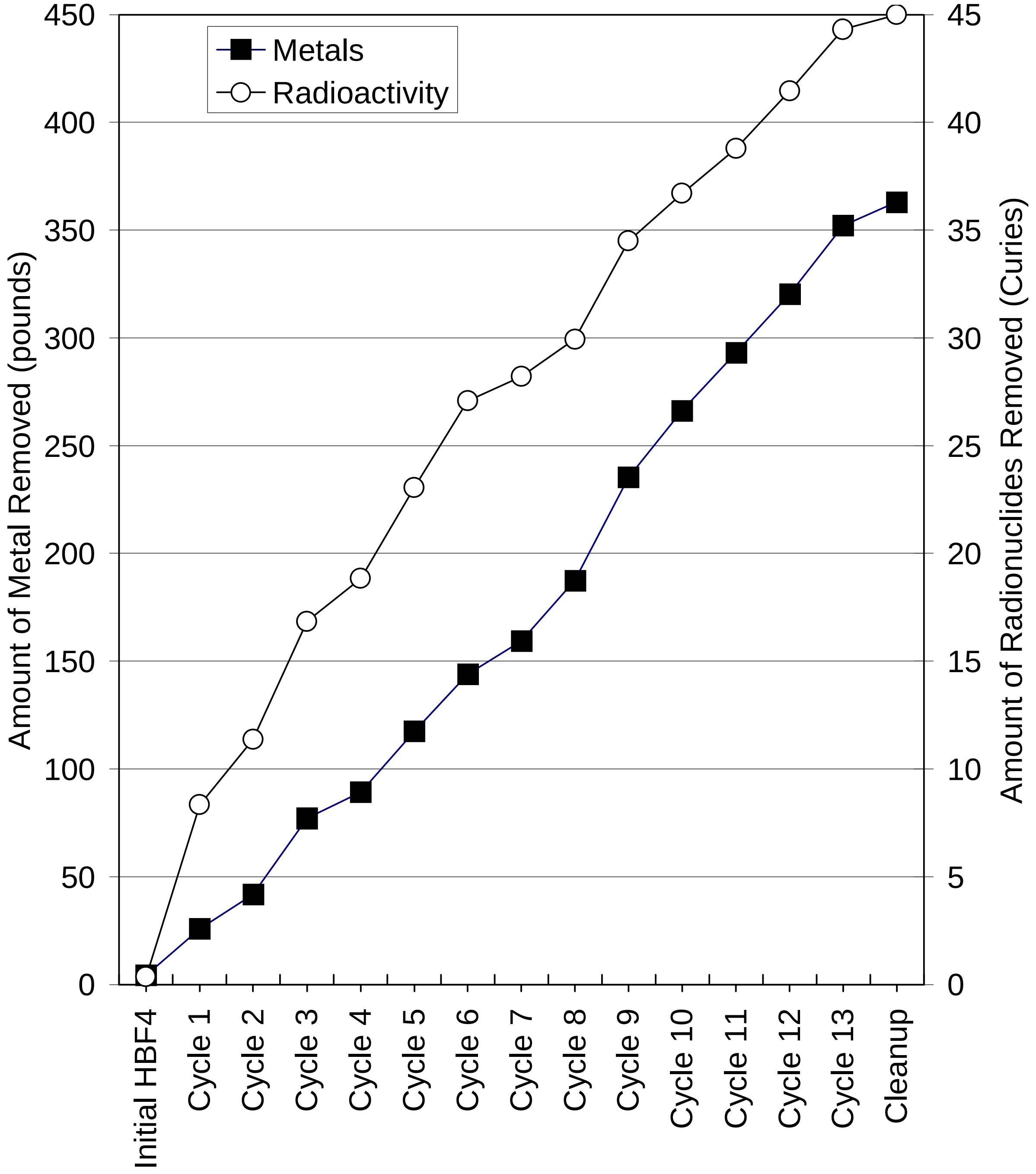
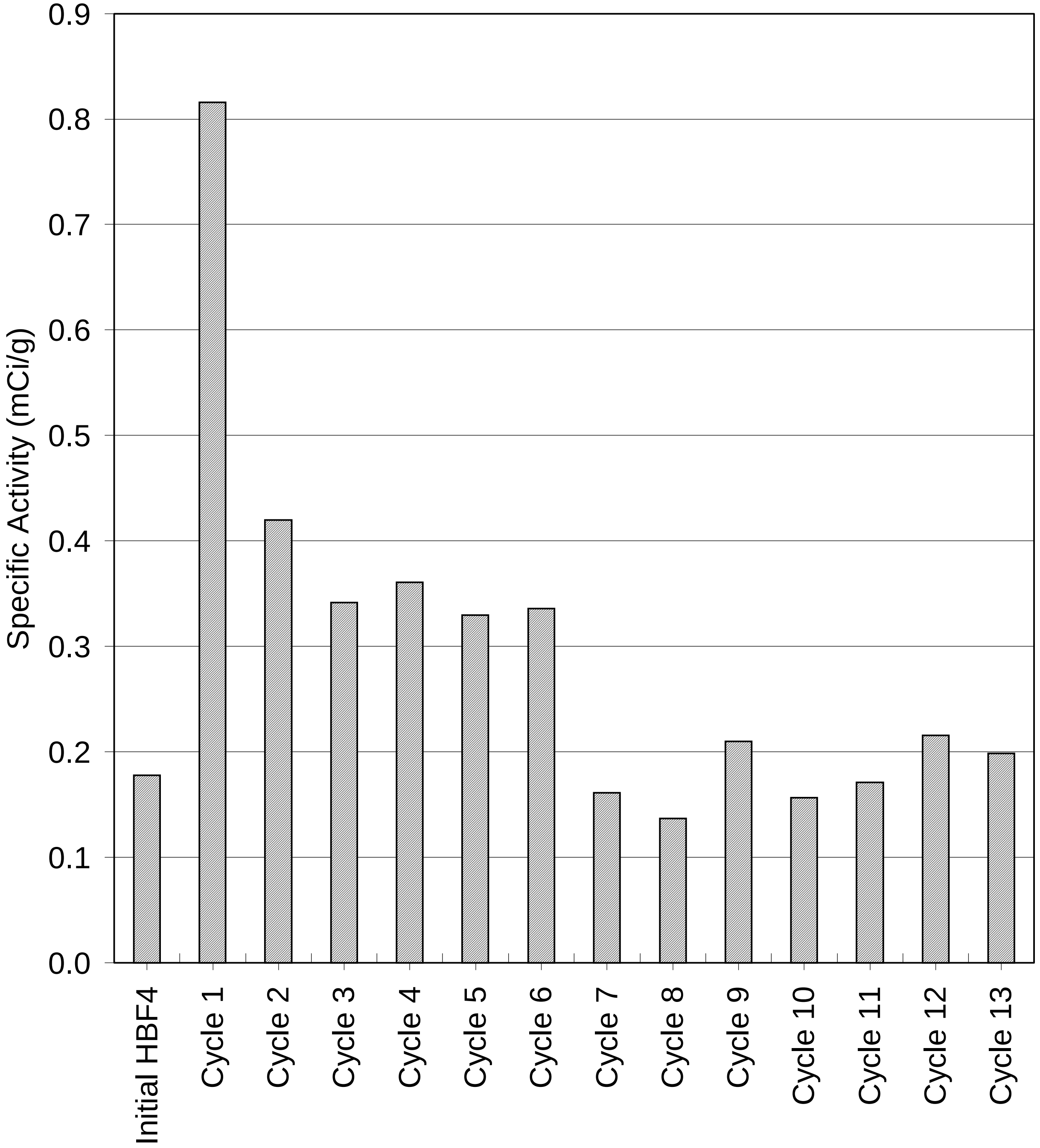


Figure 5.4
Maine Yankee - DFD Application 2
Specific Activity of Material Removed in Each Cycle



Connecticut Yankee

Cycle 1

Chemical operations for Cycle 1 started at 22:00 hours on July 26,[j] with the addition of 210 gallons of 3.0% permanganic acid solution. This resulted in a maximum concentration of permanganic acid of 220 ppm indicating that the system volume was ~30,000 gallons (114 m^3). Over the next 10½ hours its concentration slowly dropped to ~150 ppm and the dissolved chromium concentration slowly increased to ~6.5 ppm. It was steady at this value over the last 3 hours of the oxidation step indicating that chromium removal was essential complete after 7½ hours.

At approximately 10:00 on July 27 this step was terminated and Step 2 (Reduction) commenced. Over the next 1½ hours 882 pounds of oxalic acid dihydrate dissolved in 400 gallons of water was added. Reduction of the residual permanganic acid was complete within about 30 minutes. At 11:30 an attempt was made to bring on line the cation exchange column in the letdown system to initiate Step 3 (Decontamination). Despite several attempts, flow could not be established.[k] At 13:00 a major leak developed when a drain line connection failed. Approximately 1,000 gallons of solvent was lost to the waste holdup tank before the letdown system could be isolated.

Approximately 6 hours later at 17:30 on July 27, Step 3 continued with flow of ~63 gpm using the spent fuel system 'C' ion exchanger only. It operated satisfactorily for the next 8 hours but was then taken out of service because of the amount of Am-241 being removed. There were concerns that when it was sluiced to the spent resin holding tank, the final resin mixture would exceed the Class C limits for transuranic radionuclides (TRU). However, early on July 28, the official analysis of the contents of the spent resin tank were received. These results indicated less TRU than assumed, so column 'C' was placed back in service at 10:30. It was finally taken out of service at 20:00 on July 29, again because of TRU limits. The column was ~45% spent at this time.

Addition of hydrogen peroxide commenced at 01:00 on July 28 to aid in decomposition of the oxalic acid (Step 4) added during Step 2. By 20:00 on July 29, when the spent fuel ion exchanger was isolated, the total oxalate concentration had been reduced to ~250 ppm (free oxalate ~150 ppm). Since the repair and testing required to place the letdown ion exchange into service had not yet been completed, the system continued to circulate at 200°F until 22:20 on August 6. At that time, the 'A' ion exchange column in the letdown train was placed in service at a flow rate of 150 gpm. Step 4, oxalate destruction, continued. After approximately 26 hours of operation, the free oxalate concentrate had been reduced to <20 ppm. The 'A' column was then isolated and the 'D' mixed bed column put on line at the same flow rate. Cycle 1 was terminated at 15:25 hours on August 8 by isolating 'D' ion exchange column when the chromium concentration had been reduced to <5 ppm. System temperature was maintained at 200°F in preparation for the commencement of Cycle 2.

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[j] The boron concentration at this time was 440 ppm. No attempts were made to reduce it since the process had been qualified for up to 2,000 ppm boron. (10 11)

[k] The problem was eventually traced to a faulty valve. It appeared to be open when in fact it was not.

Cycle 2

Chemical operations for Cycle 2 started at 01:35 hours on August 10, with the addition of 210 gallons of 3.0% permanganic acid solution. Approximately 1½ hours later, a further 30 gallons was added when it became obvious that the minimum required concentration of 150 ppm would not be achieved, possibly because of reaction with residual oxalic acid in deadlegs and low flow areas which had diffused into the main system in the one and a half day pause between the end of Cycle 1 and the beginning of Cycle 2. The dissolved chromium jumped from 2.6 ppm, the pre-decon value, to 5.0 ppm within 1½ hours. There was no further increase over the next 5 hours despite the addition of the extra permanganic acid.

At 10:30 on August 10 this step was terminated and Step 2 (Reduction) commenced. Over the next 1½ hours 882 pounds of oxalic acid dihydrate dissolved in 400 gallons of water was added. Reduction of the residual permanganic acid was complete within about 30 minutes, the same time as observed in Cycle 1. Ion exchange flow was brought on line between 12:00 and 12:30 to commence Step 3. Flow was raised to 63 gpm through the 'C' column and approximately 145 gpm through the 'A' column for total regeneration flow of 208 gpm. At 20:50, the flow was raised to 90 gpm through the 'C' bed for a total flow of 235 gpm. Oxalic acid decomposition (Step 4) commenced at 00:50 on August 11. At 14:00 hours on August 11 the 'C' ion exchange column was taken off line due to radiation level anomalies in the system. These problems were eventually traced to failure of the internal IX column screens and the post IX filter. This combined failure allowed IX resin beads to escape from the column. Through measurement of the amount of resin remaining in column 'C', it was determined that ~20-25 ft^3 of resin was released to the piping downstream of the column. This ion exchange column was not used again during the decontamination. It remained isolated. The resin was removed from the piping by flushing with water to a HIC after completion of the decontamination.

At 12:35 on August 12, flow to the 'A' bed was lost when the booster pump tripped due to electrical problems. It was briefly restarted at 16:05 on August 13 but leaks developed in the discharge line from the booster pump and in the letdown piping downstream of the IX columns. By this time the total oxalate had been reduced to ~500 ppm and the free oxalate to ~250 ppm. Flow was restored at 14:45 on August 18 after repairs had been completed. Operation on column 'B' (cation resin) continued for 16 hours while the final stages of iron and cobalt removal and oxalate decomposition were performed. Column 'D' (mixed bed) was placed on line at 20:00 on August 19 for final cleanup. By 04:00 on August 21, the interim target cleanup parameters of <0.5 ppm chromium and <20 ppm oxalate had been met and deadleg flushing commenced shortly thereafter. Deadleg flushing was done to remove liquid of potentially high chromium content and thus reduce the load on the rad waste system IX columns. No MnO_2 was expected during these operations and none was observed. Some deadlegs were flushed by forcing clean water into the system while others were flushed by draining. Any liquid collected was returned to the system. The cation columns that would eventually be sluiced to the spent resin holding tank were also "rinsed". This included column 'C' and the downstream piping. This system was rinsed at a very low flow rate (5-10 gpm) so as to avoid disturbing the resin that was in the pipes. Cleanup with mixed bed resin continued throughout these operations. By 12:10 on September 28, all flushing had been completed and the dissolved chromium concentration was <0.5 ppm. The RHR pumped was secured and the decontamination was declared officially complete.

System Volume

During Cycle 1, the volume of the decontamination system was calculated to be ~30,000 gallons (~114 m^3) with neither IX system on line. The spent fuel IX system added about 1,000 gallons while the letdown system added about 2,000 gallons. Thus the total system volume with both IX systems in operation was ~33,000 gallons (~125 m^3). These values were calculated from system chemical concentrations after addition of a known amount of the chemical, e.g., permanganic acid or oxalic acid. Engineering calculations (14) had indicated a total system volume of ~36,000 gallons (136 m^3). The difference between the two methods is 3,000 gallons (11 m^3) or 9%, which is certainly reasonable for these types of calculations.

Metals and Radionuclides Removed

A summary of the metals (Fe, Ni, and Cr), (-emitting radionuclides (Co-60, Mn-54, Am-241, and Ag-110m), and residual chemicals (oxalate and Mn from permanganic acid) removed during each CORD cycle is given in Table 5.5. The amount of manganese in Table 5.4 agrees to within 1% of the amount that was added with the permanganic acid. This provides some assurance that the analytical results and methodology used in the calculations to obtain the data in Table 5.5 are reasonably accurate, and that negligible manganese was left behind as MnO_2. The oxalate was removed in the form $Cr(C_2O_4)_2^-$. There was negligible free oxalate. The amount of oxalate removed in Cycle 2 is higher than in Cycle 1 since the decomposition step was cut short. By this time the decision had been made not to proceed with Cycles 3 and 4, so in the interest of saving time it was decided to cleanup with the mixed bed resin rather than extend the decomposition step.

Table 5.5
Summary of All Chemical Species Removed on IX Resins During Connecticut Yankee Decontamination

Cycle	Duration[1] (hrs)	Amount Removed								
		Metals (lbs.)				Radionuclides (Ci)				Oxalate (lbs.)
		Fe	Ni	Cr	Mn	Co-60	Mn-54	Am-241	Ag-110m	
1	305 (110)	44	14	13	24	87.6	0.99	0.22	0.19	28
2	335 (190)	62	29	20	28	41.0	0.37	0.10	0.05	69
Total	640 (300)	106	43	33	52	128.6	1.36	0.32	0.24	97

Table 5.6 gives a summary of total (-emitting radionuclides and total oxide film removed during each CORD cycle. Oxide film removal was estimated from the amounts of Fe, Ni and Cr dissolved by assuming that the Fe existed as Fe_3O_4, the Ni as NiO, and the Cr as Cr_2O_3, after correction for the amounts dissolved by base metal corrosion. Base metal corrosion is discussed at greater length in Section 6. Based on a surface area of 21,500 ft^2 of stainless steel and 10,000

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[1] The first number is the total number of hours from initial permanganic injection until the end of cleanup. The second number is actual chemical process time, i.e., the first number less the time caused by plant delays.

ft^2 of Inconel[m], the 222 pounds of corrosion products removed is equivalent to an average film thickness of 7 µm (0.27 mils). This value is consistent with the value of 5-7 µm[n] obtained during the 1986 steam generator decontamination, (9) but is considerably higher than predicted from artifact test programs. The Siemens artifact test program indicated a film thickness of 1.5 µm, (10 11) while testing by London Nuclear in 1986 prior to the steam generator decontamination indicated a film thickness of 0.5-1.0 µm. (13) The reason for these large discrepancies between laboratory and field data are unexplained as of this writing. The most likely explanation is that there were regions of heavy crud deposits that were dissolved during the field applications but did not show up on the artifacts, e.g., in heat exchangers.

The specific activity of the oxide film is also given in Table 5.6. The value of 1.30 mCi/g is similar to that obtained at MY (1.01 mCi/g – see Table 5.2).

Table 5.6
Summary of Radionuclides and Oxide Film Removed During Connecticut Yankee Decontamination

Cycle	Total Activity Removed (Curies)	Total Metals Removed (lbs.)	Estimated Metals From Corrosion (lbs.)	Oxide Film Removed (lbs.)	Specific Activity of Oxide Film (mCi/g)
1	89.0	70.8	2.6	93.7	2.09
2	41.5	110.6	17.6	127.4	0.72
Total	130.5	181.4	20.2	221.1	1.30

Note that more oxide film was removed in Cycle 2 than in Cycle 1. This is because the letdown system was not in-line during Cycle 1. The heat exchangers in this system are thought to be a source of a large amount of low specific activity crud.

Figure 5.5 shows the cumulative metals and radioactivity removed during each cycle. Figure 5.6 shows the specific activity of the removed material. It is shown based on total metals removed (for ease of comparison with the MY data shown in Figures 5.2 and 5.4), and also on the basis of oxide film removed.

Chemical Usage

Table 5.7 summarizes the total quantity of chemicals used during the Connecticut Yankee RCS decontamination.

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[m] The original estimate of wetted Inconel surface area performed by plant technical staff was 19,400 ft^2. (14) However, this was based on a RCS flow rate of ~3,000 gpm. The actual flow rate was only ~1,500 gpm. This fact, in conjunction with corrosion data presented in Section 6, suggests that the actual surface area of Inconel exposed was about ½ the original estimate, or ~10, 000 ft^2.

[n] The reason for the range of 5-7 µm is due to uncertainty over the height of the SG tubes actually decontaminated.

Table 5.7
Chemical Usage During the Connecticut Yankee RCS Decontamination

Cycle	Permanganic Acid (3% Solution) (gallons)	Oxalic Acid Dihydrate (pounds)	Hydrogen Peroxide (30% Solution) (gallons)
1	210	882	60
2	240	882	68
Total	450	1,764	128

Additional Chemical Data

Additional chemical data is shown in graphical form in Appendices C and D. These graphs include:

Figure C-1	Dissolved Metals During Cycle 1
Figure C-2	Dissolved Radionuclides During Cycle 1
Figure C-3	Oxalic Acid Concentration During Cycle 1
Figure C-4	Activity Removal During Cycle 1
Figure D-1	Dissolved Metals During Cycle 2
Figure D-2	Dissolved Radionuclides During Cycle 2
Figure D-3	Oxalic Acid Concentration During Cycle 2
Figure D-4	Activity Removal During Cycle 2

These graphs show that very little base metal corrosion occurred during Cycle 1 (Ni and Fe concentrations essentially constant during period of no IX flow – see Figure C-1), but some corrosion did occur during Cycle 2 (Ni and Fe concentrations increased during period of no IX flow – see Figure D-1). Corrosion is discussed at greater length in Section 6.

Am-241 concentration was monitored by a special low energy gamma detector. It was removed with high efficiency (>98%) by cation resin.

There were two incidents of iron release from cation resin – at hours 10 and 30 on Figure D-1. The first was a result of the column having been used in the final stages of Cycle 1. During the oxalate destruction phase with H_2O_2, iron is converted to Fe^{3+} and is removed on the resin in that form in the absence of significant free oxalate. During the early stages of Cycle 2 when there was a large excess of oxalate, the Fe^{3+} on the column was complexed by the oxalate and removed from the resin since the Fe^{3+}–oxlate complex is too strong to be broken down by the resin. It was subsequently reduced to Fe^{2+} and reabsorbed by the resin. A small amount of chromium on the resin was similarly released. The second release occurred during the initial stages of oxalate destruction. Too much hydrogen peroxide was added over a short period of time. It was added faster than it could react with the oxalate so a small amount found its way to the IX resin where it oxidized the iron to Fe^{3+}. Since there was still excess free oxalate at this time, the Fe^{3+}–oxlate complex was formed as above and was released from the resin. Here again, it was subsequently reabsorbed by the resin. Neither Ni nor Co-60 was released since neither exists in the +3 valence state. It is only when metals are in the +3 valence state that they form very strong complexes with oxalate which cannot be broken down by cation resin. The complexes with metals in the +2 valence state are orders of magnitude weaker and are easily broken down by the resin.

Note that the oxalate concentration was significantly higher during the delay phase of Cycle 2 than during Cycle 1. Compare Figures C-3 and D-3. This had implications for base metal corrosion as will be discussed in Section 6.

Figure 5.5
Con Yankee - CORD Application
Cumulative Metals & Activity Removed

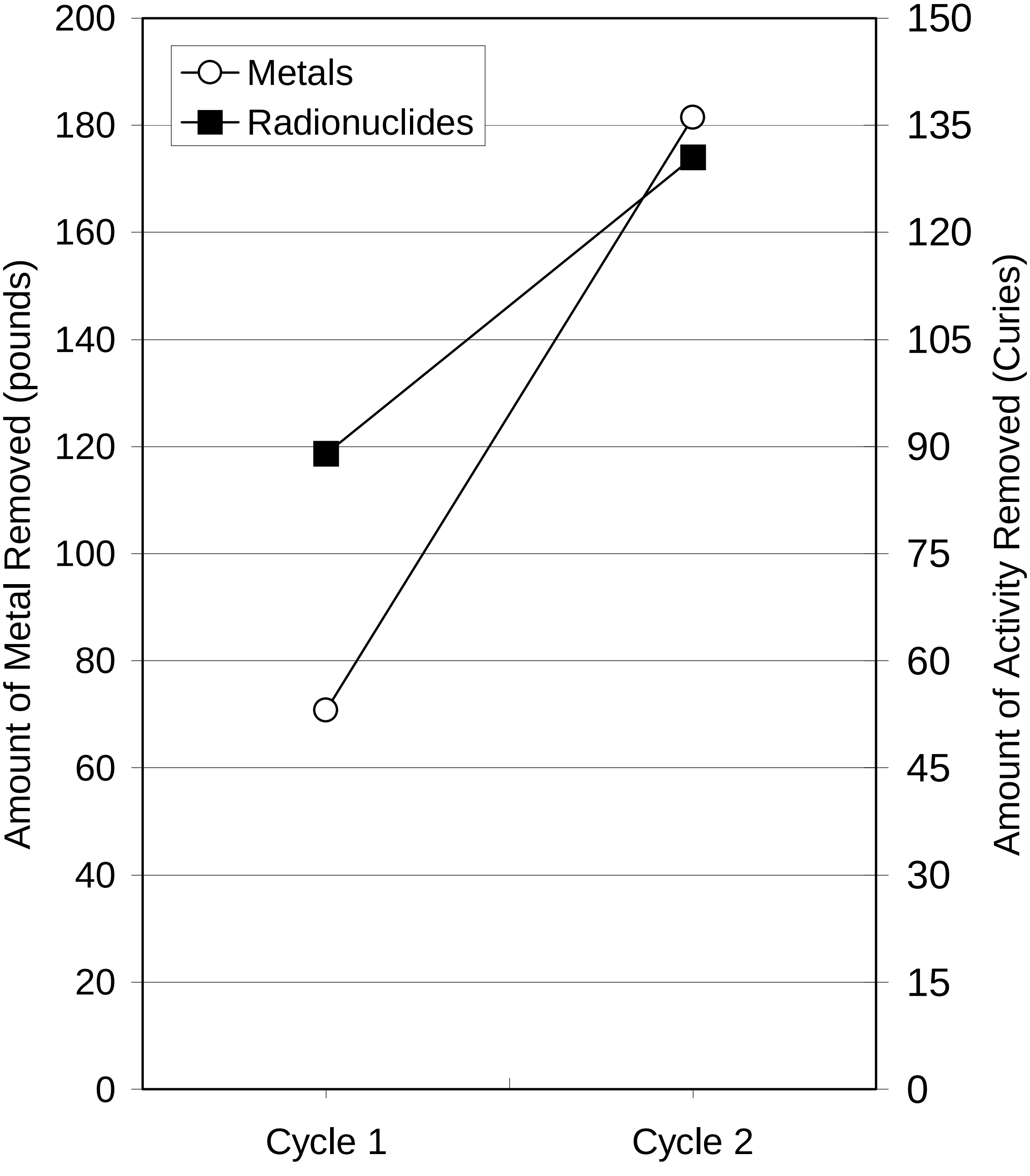
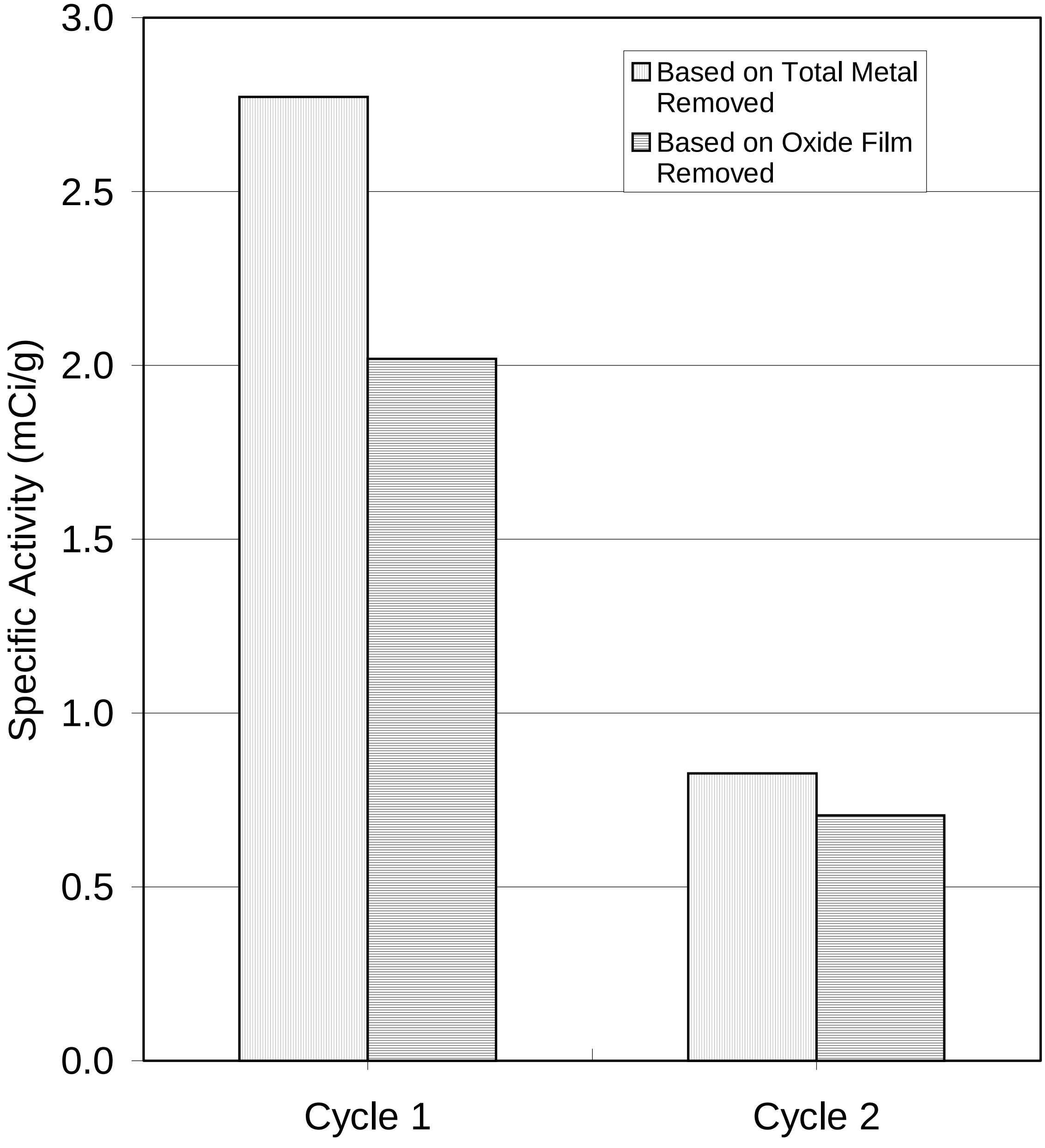


Figure 5.6
Con Yankee - CORD Application Specific Activity of Material Removed in Each Cycle



6

DECONTAMINATION CORROSION

Maine Yankee

Corrosion information for the DFD application at Maine Yankee was available from three sources. The first was weight-loss measurements taken from corrosion coupons that were installed in the process flow path at the temporary decontamination equipment during Application 1. The results of these measurements were based upon the average of two coupons of each material and are shown in Table 6.1 below.

Table 6.1
Total Corrosion Coupon Penetration
During Application 1 at Maine Yankee

Material	Average Penetration (µm)
304 ss	2.75
Sensitized 304 ss	76
Inconel	12.6

The second source of information was laboratory tests performed during the development of DFD. (3) This data indicated that ~1 µm of SS-304, 7 µm of sensitized SS-304, and ~4 µm of Inconel 600 was lost per DFD cycle. On this basis, the corrosion during Application 1 should have been 11 µm for SS-304, 77 µm for sensitized SS-304, and 44 µm for Inconel 600. Although these values are significantly higher than those shown in Table 6.1, the ratio between SS-304 and I-600 corrosion, the key system materials, is approximately the same, i.e. 1:4.

The third source of corrosion information was that calculated from the metals removed during Application 1. This was performed by making the following assumptions:

- 5,000 ft^2 of stainless steel and approximately 1,500 ft^2 of Inconel were exposed to the solvent during the application.

- 5% of the stainless steel (250 ft^2) was sensitized
- The corrosion rate of Inconel was 4 times that of stainless steel (based on laboratory testing (3) and coupon data in Table 6.1).
- The corrosion rate of sensitized stainless steel was 7 times that of SS-304 (based on laboratory testing (3)). The coupon data in Table 6.1, which indicated an enhancement by a factor of 28, was not used since these coupons were heavily furnace sensitized and probably not representative of weld sensitization.
- The composition of the stainless steel surfaces is: 74% Fe; 8% Ni; 18% Cr.
- The composition of the Inconel surfaces is: 7% Fe; 73% Ni; 15% Cr.
- Corrosion was uniform over all of the exposed surfaces.
- The amount of metal removed from oxide dissolution is small relative to the metal removed from corrosion. It was assumed to be 75 pounds, equally distributed among Fe, Ni, and Cr. These amounts were subtracted from the totals in Table 5.1 to arrive at the amount of metals removed by corrosion. Since the amount of metals from deposit dissolution is <25% of the total metals, changing the distribution among Fe, Ni and Cr has only a very small effect on the final corrosion results.
- The densities of Inconel and stainless steel are approximately 8 gm/cm^3.

Three separate calculations were performed to determine the amount of corrosion penetration based upon the quantities of Fe, Ni, and Cr removed from the system. The results are presented in Table 6.2.

Table 6.2
Calculated Corrosion During DFD Application 1 at Maine Yankee

Calculation Basis	Penetration of Stainless Steel 304 (μm)	Penetration of Sensitized Stainless Steel (μm)	Penetration of Inconel 600 (μm)
Fe	12.4	87	50
Ni	11.0	77	44
Cr	11.9	84	48
Average	11.6	82	47

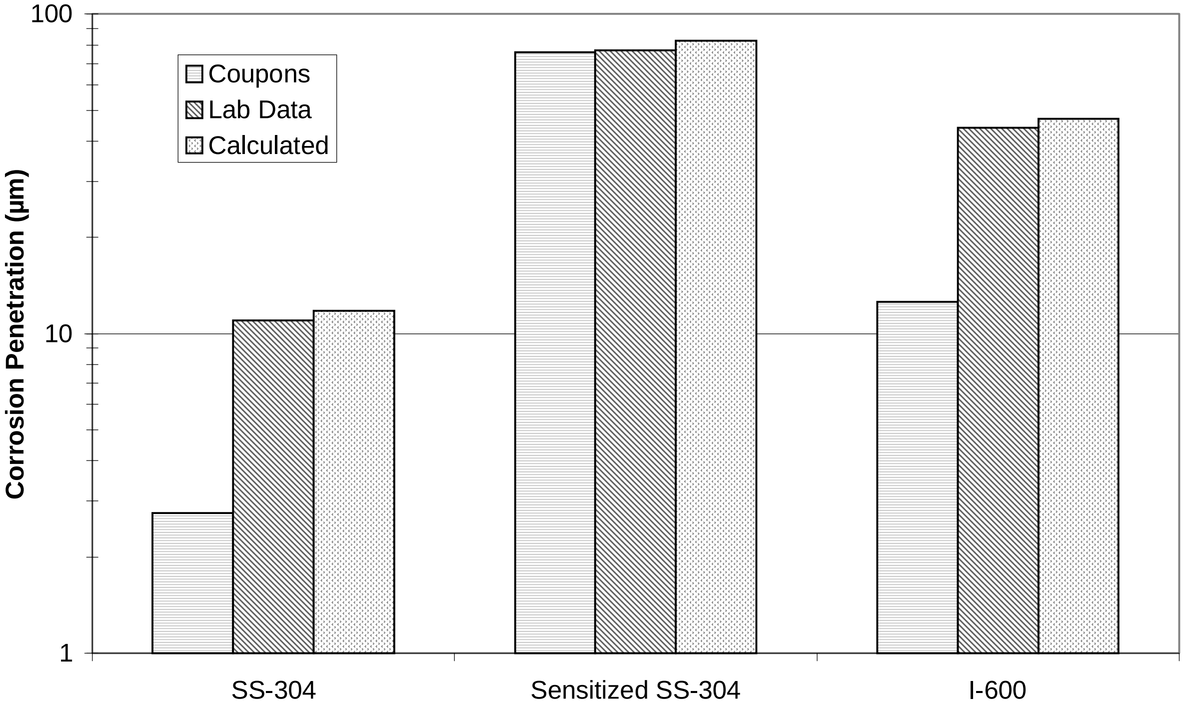
A comparison of the results of Tables 6.1 and 6.2 indicates a difference in the corrosion rates of the key system materials (SS-304 and I-600) by a factor of ~5 between measured and calculated values. In actual fact, the differences may be smaller than those identified in the above tables. The results in Table 6.2 are based upon a calculation that required a number of simplifying assumptions in order to generate a result. These assumptions may have tended to increase the calculated result. For example:

- the surfaces areas may be larger than assumed (the area of the steam generator tubes exposed to the solvent is an educated guess)
- a greater fraction of the metals removed may have come from deposits

In addition, the surface finish of the corrosion coupons may not have duplicated that of the plant piping. The coupons were highly polished whereas the insides of pipes would be much rougher. A rough surface would have an effective surface area much higher than indicated from simple geometry. For sensitized materials, the degree of sensitization is also a factor that will affect the amount of corrosion.

Figure 6.1 is a graphical comparison of the corrosion rates of all three materials as determined by for all three methods. There is excellent agreement between the calculated and laboratory values for the key system materials which suggests that the coupons may not have been exposed to representative process conditions.

Figure 6.1
Summary of Corrosion During DFD Application 1 at Maine Yankee



In summary, it appears that SS-304 underwent ~12 µm (~0.5 mils) of corrosion, or approximately 1 µm per DFD cycle. Inconel 600 underwent about 4 times that amount (~50 µm or ~2 mils), and sensitized stainless steel about 7 times (~80 µm or ~3 mils). The corrosion of sensitized SS-304 (~7 µm per cycle) is not unreasonable. Sensitization causes the steel to lose some of its austenitic nature and it is known from laboratory tests that low alloy carbon steels and 400-series stainless steels can experience corrosion rates in the range 80-150 µm (3-6 mils) per cycle. (3) Corrosion of sensitized SS-304 of 22 times that of unsensitized material has also been reported. (3) This is approximately the same difference as observed with the coupons. Presumably, the material in this test was more heavily sensitized and similar to that of the coupons. The more heavily sensitized the material, the more of its austenitic character it loses and the more susceptible it becomes to corrosion by the DFD process.

Connecticut Yankee

Cycle 1

Corrosion coupons were not installed during this decontamination so the only corrosion information available is from artifact testing and that which can be gleaned from the process application itself.

Artifact testing indicated that corrosion of SS-304 was in the range 0-0.02 µm per CORD cycle. (10 11) However, each artifact test cycle lasted for about 6 hours, not the 300 hours experienced during the actual decontamination. A direct extrapolation from 6 hours to 300 hours is not possible since the majority of the 300 hours was at a very much reduced oxalate concentration (250 ppm versus 2,500 ppm).

During the delay period of Cycle 1 (hours 90[o] to 250 – see Figure C-4), the dissolved Ni only increased by 2 ppm (see Figure C-1), which corresponds to 0.5 lbs. Assuming this all came from base metal corrosion, and assuming a surface area of SS-304 of 21,500 ft^2 and a surface area of I-600 of 10,000 ft^2, and further assuming that corrosion of I-600 is twice that of SS-304,[p] it can be calculated that this corresponds to a corrosion penetration of SS-304 of 0.019 µm and of I-600 of 0.037 µm. To be ultra conservative, 0.02 µm can be added for corrosion of SS-304 that occurred during the first few hours of high oxalate (and 0.04 µm for I-600), bringing the maximum corrosion estimates for Cycle 1 to 0.04 µm for SS-304 and 0.08 µm for I-600. A similar result is obtained if the increase in dissolved Fe is used as the basis for the calculation.

This data is summarized in Table 6.3. It must be emphasized that these are maximum estimates. If some or all of the increase in Ni between hours 90 and 250 was due to slow deposit dissolution rather than corrosion,[q] then corrosion may have been essentially zero.

Cycle 2

Corrosion during Cycle 2 was estimated in a similar manner to that in Cycle 1, i.e., the increase in dissolved Ni and Fe during the delay period from hours 60 to 205. The Ni concentration increased from 3.4 to 25 ppm while the Fe concentration increased from 11.8 to 33 ppm. These increases represent 6.3 lbs. of Ni and 6.9 lbs. of Fe. Applying the same methodology as above, it can be calculated that corrosion of SS-304 was 0.24 µm while that of I-600 was 0.48 µm. Adjusting for corrosion during the initial phases of the cycle, these values become 0.26 and 0.52 µm, respectively.

It is interesting to note that the original estimate of the surface areas of SS-304 and I-600 were 21,500 ft^2 and 19,400 ft^2, respectively. (14) Using these areas, it was impossible to reconcile the

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[o] The delay actually started at hour 70 but examination of the data indicates that deposit dissolution was still occurring between hours 70 and 90. There is an abrupt change in the slope of the Ni, Fe and Co-60 concentration graphs at hour 90. See Figures C-1 and C-2.

[p] This information was supplied by Siemens based on their experience with CORD. It could not be confirmed based on the data available from the artifact testing or the actual decontamination.

[q] The Co-60 concentration increased from 4.8E-2 to 6.4E-2 µCi/mL during the delay period from 90 to 250 hours indicating that some deposit dissolution was occurring. See Figure C-2.

amounts of Ni and Fe dissolved. Only when the surface area for I-600 was reduced to 10,000 ft^2, could a reasonable correlation be obtained. This reduction in effective surface area is considered reasonable since the original calculation was based on a RCS flow rate of ~3,000 gpm. In practice, only ~1,800 gpm was obtained. Therefore, to a first approximation, there should be a corresponding reduction in the surface area of the steam generator tubes that was exposed to the solvent.

The corrosion values given above are approximately an order of magnitude higher than observed in Cycle 1. This is attributed to two factors:

1. The concentration of oxalate was a factor of 2 higher during the delay period of Cycle 2 as compared to Cycle 1 (500 ppm versus 250 ppm). See Figures C-3 and D-3.
2. The majority of the oxide film had been removed by this time so the base metal was exposed and more susceptible to corrosion.

Corrosion information during both CORD cycles is summarized in Table 6.3. Again it must be emphasized that these are maximum estimates. For comparison, the amount of base metal loss expected during a CORD cycle that employs the "D" option is estimated at 0.5-4 µm for SS-304, and 1-8 µm for I-600. These values are based on Siemens experience and the artifact testing program. (10 11) A range is given since the actual value depends upon how long the "D" option is applied, and how "aggressively" it is applied.

Table 6.3
Estimated Maximum Corrosion During CORD Cycles 1 and 2 at Connecticut Yankee

Cycle	Penetration of Stainless Steel 304 (µm)	Penetration of Inconel 600 (µm)
1	0.04	0.08
2	0.26	0.52
Total	0.30	0.60
Typical "D" Cycle[r]	0.5 – 4	1 – 8

The maximum estimated corrosion values correspond to 20 lbs. of base metal dissolved. This value is broken down by cycle and used in Table 5.6 to determine net oxide film removal.

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[r] Shown for comparison only. The "D" option was not employed at CY.

7

DECONTAMINATION WASTES

Maine Yankee

Use of ion exchange resin during the decontamination is summarized in Table 7.1. In total, 625 ft^3 of resin was used. However, as explained in Section 5, approximately 90 ft^3 was used for boron removal. Thus, the net volume attributed to DFD is 535 ft^3.

A breakdown by resin type is also given in Table 7.1. Cation columns A, D, C, and F were employed during most of Application 1. Column F received preferential flow and became spent during Cycle 7, as explained in Section 5. Breakthrough was detected by an increasing iron concentration on the IX outlet. See Figure A-2. It was valved out and Cycle 7 was completed using the remaining three cation beds at a reduced flow of 200 gpm. It had been 300 gpm when all four columns were available. Towards the end of Cycle 8, columns A, B, and C also broke through, as explained in Section 5. They were taken off line and the balance of Application 1 was completed with columns D and E. For cleanup of Application 1, the three strong base anion columns G, H, and J were placed on line in parallel after the two cation columns D and E. Cleanup was continued until conductivity of the system water was <25 µmho/cm.

Resin sluicing and refill operations began immediately upon completion of Application 1.

While preparing for Application 2, it was discovered that the residual boron concentration was much higher than anticipated (~422 ppm). Columns G, H, and J were expended in reducing the boron concentration to approximately 250 ppm. This required that all of the columns except Column K (the mixed bed polishing column) be sluiced and refilled prior to the start of Application 2. To reduce the boron to acceptable levels (<40 ppm) the fresh resin in columns A, B, and C were placed on line in series with columns G, H, and J for a short period of time.

Prior to the start of Application 2, 2½ high integrity containers (approximate capacity of 105 ft^3 each) were filled with the spent resin from Application 1 and boron removal operations.

Table 7.1
Ion Exchange Resin Usage During the Maine Yankee DFD

Step During Decontamination Operations	**Resin Type**									
	Cation (ft^3)						**Anion (ft^3)**			**Mixed Bed (ft^3)**
	Column Designation									
	A	**B**	**C**	**D**	**E**	**F**	**G**	**H**	**J**	**K**
Initial Loading	30	30	30	25	25	30	30	30	30	30
Boron Removal							P	P	P	
Application 1	X	X	X	X	X	X	P	P	P	
Boron Removal							X	X	X	P
Reload	30	30	30	30	30	30	30	30	30	
Boron Removal (Cont.)	P	P	P				P	P	P	
Application 2	X	X	X	X	X	X	P	P	P	
Reload	25	20	20							
Application 2 (cont.)	X	X	X							
Final Cleanup							X	X	X	X
Totals (ft^3)	415						180			30
Grand Total (ft^3)	625									

Notes re Table 7.1:

P = Column is partially spent
X = Column is fully spent

During Application 2, cation columns A, B, and C were placed on line as needed during Cycles 1 through 7. During Cycle 8, it was confirmed that these columns were spent and they were replaced by columns D, E, and F. Sluicing of columns A, B, and C was completed during Cycle 9 and they were refilled with 25 ft^3, 20 ft^3, and 20 ft^3 of cation resin, respectively. During Cycle 12, columns D, E, and F were then replaced with columns A, B, and C, which remained on line until the end of Application 2.

For cleanup and final polishing of the system fluid, and to remove residual chemicals and the anionic radioactivity remaining in solution, anion columns G, H, and J, and mixed bed column K, were placed on line in series with cation columns A, B, and C. Once final water quality specifications had been reached, the columns were taken off line and sluiced to the existing half-full HIC and three additional empty ones for a total of six, 105 ft^3 capacity, waste containers.

Connecticut Yankee

The only waste form produced by the decontamination was ion exchange (IX) resin. All metals from deposit dissolution and base metal corrosion, all radionuclides released from the deposit and the base metal, and all residual chemicals were captured on IX resin. The water in the system was left in a sufficiently clean state (Cr <0.5 ppm and <20 ppm oxalic acid) that it could be drained to either the plant's primary drain tank (PDT) or aerated drain tank (ADT). A summary of the types and quantities of waste IX produced by the decontamination is given in Table 7.2. Note that the recharge of Column C after Cycle 1 was due to its transuranic radionuclide (TRU) loading. This was based upon measuring and extrapolating from the Am-241 loaded on Column C during Cycle 1. Column C was changed out to ensure that the waste decon resin would not exceed the Class C limits for TRU. Chemically, the resin was only about 45% spent. Similarly, a second mixed bed cleanup column was required during Cycle 2 to avoid generating greater than Class C waste on Column D.

Table 7.2
Summary of Ion Exchange Resin Wastes Produced by the Connecticut Yankee Decontamination

Charge	**Column Loading (ft^3)**					**Total Resin Waste (ft^3)**
	A	**B**	**C**	**D**	**Boron Recycle**	
Predecon flush & water cleanup	45	45	45	45	40	220
Initial charge	45 (cation)[s]	45 (cation)	45 (cation)	18 (cation) 22 (anion)[t]		175
Recharge after Cycle 1			45 (cation)			45
Recharge during Cycle 2 cleanup		11 (cation) 14 (anion)[u]				25
Subtotals	90	115	135	85	40	465

The official volume of waste ion exchange resin generated by this project is 465 ft^3. However, much of this is due to the high TRU content of the oxide film. To avoid generating a waste form that was greater than Class C, several beds were sluiced to the spent resin holding tank when they were less than 100% spent chemically. It has been calculated that the volume of resin

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[s] Dowex HCRW-2-H strong acid cation resin in H+ form.
[t] Ionac A-365 weak base anion resin in OH^- form.
[u] Ionac A-642-OH strong base anion resin in the OH^- form.

actually required by CORD was only ~115 ft^3. This consisted of 75 ft^3 of cation resin and 40 ft^3 of mixed bed resin. Even these numbers are conservative since they have been corrected for column utilization efficiency of 90%, i.e., the volume of resin actually consumed in removing ionic species was divided by 0.9.

At the end of the decontamination, 155 ft^3 of resin remained in the four IX columns and 315 ft^3 was in the spent resin storage tank. All resin will be subjected to a full analysis to ensure it meets all shipping and burial regulations. It will then be transferred to HICs and shipped to a licensed disposal site for burial.

8

DECONTAMINATION FACTORS and RADIATION EXPOSURE

Maine Yankee

Reductions in radiation fields during the course of the decontamination were followed by three methods:

1. Contact dose rates on pipes, valves, components etc. A complete survey of 48 points was performed prior to the decontamination, at the end of Application 1, and at the end of Application 2. These points were jointly agreed upon by the utility and the decon vendor. The average of these 48 points would constitute the "official" DF for the project. Interim surveys were taken on a few of these points at various times throughout the applications so that the effectiveness of the decontamination could be monitored.

2. Teledosimetry measurements. Fourteen detectors with remote readout were installed at various critical locations. These detectors were not in direct contact with the pipe. In some cases they were external of shielding while in others they simply monitored area dose rates. Thus, their outputs were useful in determining general area dose rate reduction factors (DRF). Although the DRF values tended to be lower than the contact DF values, they were still extremely useful since they could be read several times a day without any dose being expended.

3. Area dose rates. A survey at 43 locations was performed with a standard survey meter prior to the decon, at the end of Application 1, and at the end of Application 2. A few of the points were read periodically through the decontamination to follow its progress. The locations were selected based on where work was to be performed in subsequent decommissioning activities. Thus the average DRF measured at these locations would provide a good indication of the dose reduction decommissioning personnel would receive as a result of the decontamination.

<u>DF Results</u>

1. Contact dose rates. The average DF of all 48 contact survey locations was 31.5. The average DF on radiologically significant sources (initial contact reading ≥100 mR/hr) was 89.3. The highest individual DF was 300. Average DFs by system are shown in Table

8.1. Average DFs by radiological significance are shown in Table 8.2. Intermediate readings were taken on a few of the points throughout Application 1. Results are shown in Figures A-8 through A-10. This method of DF measurement was the most useful in determining the overall effectiveness of the application. A summary of the DF on all 48 measurement points is shown in Figure 8.1. Note that most of the DFs (~75%) were <10 and it is only the presence of a few very high values (~300) that resulted in the arithmetic average being 31.5.

Table 8.1
Average Contact DFs by System at Maine Yankee

System	DF
RCS (all)	8.7
RCS 33" coolant piping	7.3
RCS 8" bypass piping	13.3
RCP volute	3.6
Letdown piping	91.2
Charging piping	4.2
Pressurizer spray	203
Safety Injection	2.5
Seal water return	3.4
Residual heat removal	4.6
Loop fill and drain	4.6
Average (all 48 points)	31.5
Average (15 points ≥100 mR/hr initial)	89.3

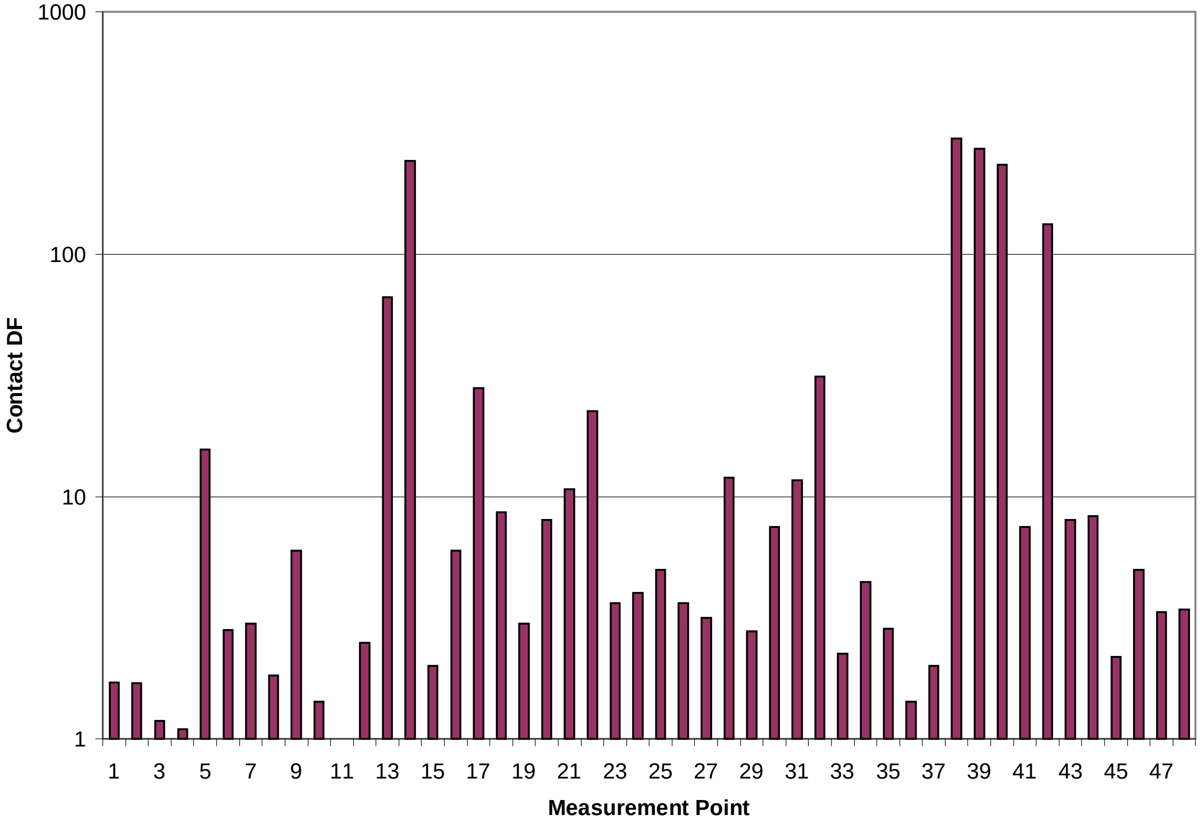
Table 8.2
Average Contact DFs by Radiological Significance at Maine Yankee

Initial Contact Dose Rate	DF
≥1,000 mR/hr	107.1
500 to 1,000 mR/hr	169.5
100 to 500 mR/hr	24.5
<100 mR/hr	5.2

2. The average DF based on 14 teledosimetry readings was 11.7. This is considerably lower than the value reported for contact readings for two reasons. First, the detectors were not in direct contact with the pipe. They were placed as close as practical to the pipe or component being monitored, but in practice they ended up giving results that were midway between contact DFs and general area DRFs. Typical examples of the response of the teledosimetry units are shown in Figures A-5 through A-7 for Application 1 and in Figures B-5 and B-6 for Application 2. Second, only ~25% of the contact DF readings were >10. See Figure 8.1. If the teledosimetry locations did not happen to correspond

with the high DF regions, then a lower average value would be expected. Despite these shortcomings, this method of DF measurement had the advantage of real-time, instantaneous measurements. It was very useful in following the individual process steps and in determining how many cycles of DFD were required.

Figure 8.1
Decontamination Factors at Maine Yankee



3. The average DRF number throughout the areas of the plant directly affected by the decontamination was 4. Average DRFs by location are shown in Table 8.3. This method of DF measurement was the most realistic when it came to determining the reduction in radiation exposure to plant workers as a result of the decontamination. Very high DRFs were achieved in several high flow areas such as the regenerative heat exchanger and pressurizer cubicles (37 and 10.5, respectively). These were a remarkable achievement and of enormous value to the station in subsequent decommissioning activities.

4. In 1995, Maine Yankee performed a loop-stop decontamination on its three RCS loops. Approximately 400 Ci of gamma emitting isotopes were removed. This resulted in a much lower inventory of activity available for removal in this decontamination. This undoubtedly resulted in somewhat lower initial radiation levels immediately prior to the 1998 decontamination, which affected the final DF result. It is generally recognized, all other things being equal, that higher initial starting fields usually result in higher DFs.

Table 8.3
Average Area DRFs by Location at Maine Yankee

Location	DF
Regen heat exchanger cubicle	37.1
Loop 1	3.9
Loop 2	2.6
Loop 3	2.4
Pressurizer cubicle	10.5
Letdown area (PAB)	1.4
Average (43 locations - all areas)	4.0

Radiation Exposure

A goal of 25 Rem had been established for the performance of the decontamination at Maine Yankee. Each of the major tasks associated with receiving radiation exposure during the project had been identified and an ALARA goal established for their performance. With careful monitoring and appropriate controls, the actual exposure to project personnel was limited to a total of approximately 11 Rem. Table 8.4 summarizes the radiation exposure received for these project tasks.

Table 8.4
Maine Yankee Decontamination Radiation Exposure

Project Activity	Exposure Goal (Rem)	Actual Exposure (Rem)
Setup of Decon Equipment & Making System Connections	5.58	2.06
Steam Generator Temporary Manway Installation & Removal	1.44	0.48
Nozzle Dam Setup, Installation & Removal	1.59	0.63
Shielding of Seal Area	0.24	0.12
Preparation of Reactor Coolant Pump Seals	0.64	0.04
Preparation of Drag Valves LD-A 9 & LD-A 10	0.28	0.03
Reactor Head Preparations & Core Barrel Removal	1.44	1.17
Decon Waste Handling (Resin & Filters) and Processing	2.39	0.74
Filter Changeouts During Application 1 & 2	1.59	0.46
Decon System Operations, Testing, Walkdowns and Sampling	3.19	1.63
HP Coverage, Process Evaluation Surveys & Operations Support	3.43	2.05
Decontamination & Demobilization of Decon Equipment	3.19	1.43
Deadleg Flushing		0.50
Totals	25.00	11.34

Connecticut Yankee

DF Results

A total of 36 locations (measuring points) were agreed to by Siemens and CY personnel for the purpose of determining the "official" decontamination factor (DF) for the project. The radiation field at each point was measured on contact using an E-530 meter and a shielded probe, and at 12" (30 cm) using a standard teletector. The former is a measure of the effectiveness of the process while the latter, called a general area dose rate reduction factor (DRF), gives a more realistic estimate of the reduction in exposure likely to be experienced by plant workers. Table 8.5 gives a summary of the DF and DRF readings obtained after each cycle, as well as the final cumulative values.

Table 8.5
Summary of DF and DRF Measurements at Connecticut Yankee

Cycle	Contact DF	General Area DRF
1	5.2	~3
2 (net)	3.1	~1.6
2 (cumulative)	15.9	~5

The 36 measuring points were divided into two groups – those with an initial field of ∃100 mR/hr and those with an initial field of <100 mR/hr. This was done on the basis that it is more difficult to obtain a high DF with a low initial field due to the greater influence of background radiation. Results of DF measurements from the two groups of points are given in Table 8.6.

Table 8.6
Summary of DF Measurements at Connecticut Yankee Based on Initial Radiation Field

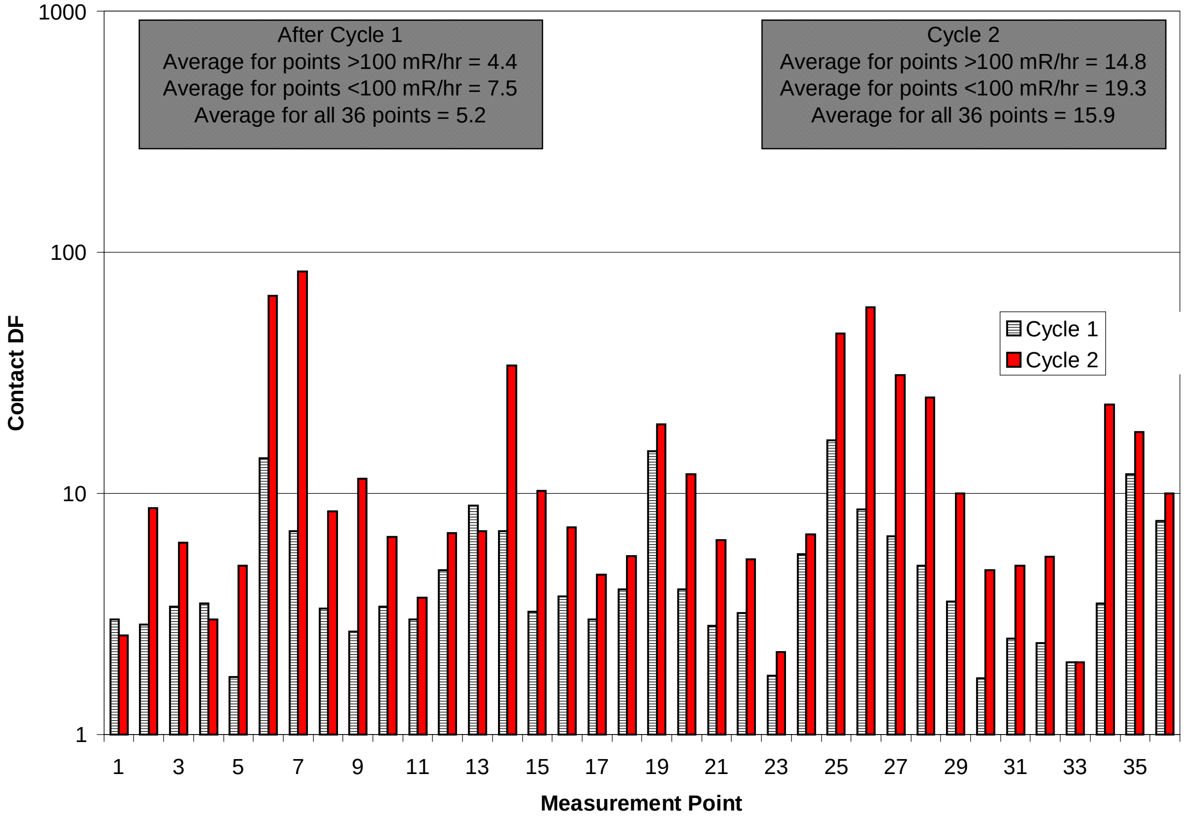
Contact DF (initial field ∃100 mR/hr) (27 points)	Contact DF (initial field <100 mR/hr) (9 points)
14.8	19.3

The effect of lower DFs with lower initial starting values that was observed at MY was not duplicated here. In fact, the opposite was observed. Although the differences are small, the lower initial fields actually resulted in higher DFs.

The results for all 36 measurement points are shown in Figure 8.2. The same scale has been used as in Figure 8.1 for ease of comparison between the two sets of results. Note that the majority of the DFs (~67%) are ≤10. It is only because of a few high DFs (33% are in the range 10-83) that the arithmetic average was >15. This is almost identical to what happened at MY.

The results are similar to those obtained at MY. Compare Figures 8.1 and 8.2. One reason for the difference in average DFs between the two decontaminations is that MY had several very high initial readings (up to 8,000 mR/hr), while the highest initial value at CY was 1,200 mR/hr. It is generally recognized that it is easier to obtain high DFs with higher initial fields. In addition, more base metal was removed at MY than at CY, with a higher resulting overall average DF.

Figure 8.2
Decontamination Factors At Connecticut Yankee



Radiation Exposure

A goal of 39 Rem had been established for the performance of the decontamination at Connecticut Yankee. Despite the long delays in the project and the large amount of repair work that had to be done on the letdown system, only 30 Rem were expended. This was accomplished through detailed ALARA planning for each entry that had to be made to radiation areas. A summary of radiation exposure by major tasks is given in Table 8.7.

Table 8.7
Summary of Decontamination Radiation Exposure at Connecticut Yankee

Activity Description	**Exposure Goal (Rem)**	**Actual Exposure (Rem)**
Initial System Modifications (Mod#1- Mod #7)	18.6	11.6
Decontamination	18.4	16.4
Post-Decon System Restoration	2.0	2.0
Project Total	39.0	30.0

9

DISCUSSION/COMPARISON OF RESULTS

The results from the Maine Yankee and Connecticut Yankee decontaminations have been presented in previous sections. In this section they are briefly summarized and compared.

These applications have a number of key similarities that include:

- Both plants are PWRs that operated for long periods under similar reducing water chemistry conditions. This resulted in similar types of oxide films being laid down on the primary system materials of construction.
- Both have similar (but not identical) materials of construction for the systems exposed to the decontamination chemicals.
- Both chose virtually the same primary systems to be exposed to the decontamination chemicals.
- The plants are similar vintage (MY 1972, 825 MWe, and CY 1968, 583 MWe).
- Although different in detail, both selected regenerative decontamination processes which operate under similar alternating chemistry conditions (oxidizing/reducing cycles which are repeated), using similar decontamination chemicals (both processes use oxalic acid for reducing conditions and permanganate for oxidizing conditions).
- Both decided to bypass the RPV with the installation of a nozzle jumper assembly consisting of nozzle plugs and jumper lines.

There are, as well, a number of important differences between the applications. These include:

- Maine Yankee selected the DFD chemical process for its application while Connecticut Yankee selected the CORD process (see Section 3).
- Maine Yankee chose not to use any station pumping equipment in support of decontamination operations. Connecticut Yankee chose to operate its RHR pumps during the decontamination. This resulted in flow rates that were between 3 and 6 times higher than those at Maine Yankee.
- Maine Yankee opted to bypass their steam generators by installing jumpers between the hot and cold leg manways of each generator.[v] Connecticut Yankee did not install

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[v] Although the SG tubes were bypassed, the bottom 6-12 inches of each tube were filled with fluid and effectively exposed to the decontamination process as the fluid level rose and fell due to changes in system pressure, flow rate, etc. As discussed in Section 6, it was estimated that the total surface area of Inconel exposed was 1,500 ft^2.

jumpers and allowed the flow to pass through a portion (estimated at about 10% -15%) of each steam generator's tubing. This would tend to increase the activity removal results at CY (without a corresponding increase in DF) relative to MY.

- Maine Yankee chose not to use any of its installed ion exchange capacity, instead relying on the decontamination vendor's equipment. Connecticut Yankee decided to use its installed ion exchange columns. The result was that Maine Yankee had much more flexibility and range with chemical regeneration, metal and radioactivity removal, and chemical cleanup.
- The DF objective for Maine Yankee was 25 while that for Connecticut Yankee was 15.
- The decontamination at Maine Yankee was performed in two separate DFD applications with some piping and equipment exposed to both applications. All of the Connecticut Yankee systems were exposed to a single, two-cycle CORD application.
- In 1995, Maine Yankee performed an operational decontamination of the SG channel heads and RCS piping between the loop-stop valves. Approximately 400 Ci of gamma emitting isotopes were removed. This resulted in a much lower inventory of activity available for removal in this decontamination. This undoubtedly resulted in somewhat lower initial radiation levels immediately prior to the 1998 decontamination, which affected the final DF result. It is generally recognized, all other things being equal, that higher initial starting fields usually result in higher DFs (although for reasons that are not understood, this was not observed at CY – see Table 8.6).

In Table 9.1, the major parameters from each decontamination are summarized. Some of the items require explanation.

Total chemical processing time is the overall time from first chemical injection until the completion of final cleanup. It includes all delays and the changeover from Application 1 to 2 (MY) and from Cycle 1 to Cycle 2 (CY). It does not include post-decontamination deadleg flushing, final cleanup, IX resin sluicing, etc. These operations took about 4 weeks at MY and 5 weeks at CY. Overall project schedules are given in Section 4.

Total solvent flow rate is the total flow put out by the pump. Since both decontaminations employed several parallel flow paths, the flow rate in any one path was much less than the value listed. For example, at MY the flow through the CVCS system in Application 1 was about 50 gpm. At CY the flow through the RCS piping was 700-800 gpm.

The average general area dose rate reduction factor (DRF) for MY appears abnormally low given the high contact DF (31.5) and the significant drop in average contact radiation fields (from 460 to 20 mR/hr). There are some regions of the plant where the DRF was <3 (see Table 8.3). It appears that a disproportionate number of the readings were taken in these regions resulting in an average DRF of only 4. If the six general area DRFs in Table 8.3 are averaged, the overall average DRF is 10. This appears more reasonable when compared to the CY results.

DFD is not currently qualified in the presence of boron, so some IX resin (90 ft^3) was required to reduce its concentration to <40 ppm. At CY boron was not a concern, but TRU was. Several of the IX columns had to be valved out and replaced when they reached the Class C limit to avoid generating a waste form that was unacceptable for shipping and burial. They were not totally spent at this point so the total volume of resin used was much larger than it would have been had TRU not been present.

Table 9.1
Summary of Major Parameters from the Decontaminations

Parameter	Plant	
	MY (App. 1/App. 2)	CY
Process	EPRI's DFD	Siemens' CORD
Number of chemical process cycles	11/13	2
Total chemical processing time (hours & days)	480 (20)	610 (25)
Net chemical processing time (hours)[w]	202/193	238
Deadleg flushing, final cleanup, IX sluicing, etc.	4 weeks	5 weeks
Total solvent flow rate (gpm)	300/650	1,800
System volume (gal)	18,000/22,000	33,000
Average IX flow rate (gpm)	250/300	140
IX purification half life (min)	50/50	160
Number of measurement locations for DF	48	36
Overall average contact DF achieved	31.5	15.9
Highest single contact DF	300	83
Average initial contact radiation field (mR/hr)	460	234
Average final contact radiation field (mR/hr)	20	27
Overall average area DRF achieved	4[x]	5
Highest single area DRF (and location)	37 (regen HX cubicle)	20 (loop bypass line area)
Total metals removed (lbs.)	677	182
Base metal (SS-304) penetration (µm)	12/14	0.3
Total (-emitting radionuclides removed (Ci)	103	131
Total IX resin waste generated (ft^3)	625	465
Net IX resin due to decontamination (ft^3)	535[y]	115[z]

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[w] Time due to plant delays, boron removal, changeover between cycles and applications, etc. not included.

[x] It appears that a disproportionate number of the 43 readings were taken in regions where the DRF was <3. An average DRF by area results in 10, a value that seems more reasonable when compared to the CY results.

[y] IX resin used in boron removal not included.

[z] Undepleted IX resin sluiced prematurely to avoid generating a Class C waste not included.

10

LESSONS LEARNED

Maine Yankee

1. Start decommissioning and decontamination planning well in advance of the final plant shutdown. Have the maximum number of plant systems and pumps available and online.

2. The flow rates of 300 gpm in Application 1 and 650 gpm in Application 2 were too low. With higher flows (~2,000 gpm) fewer cycles would have been required, higher DFs would have been achieved in a shorter time, less chemicals would have been required, less MnO_2 would have settled in deadlegs, and less waste would have been generated. It may not be possible to achieve flows in this range with vendor supplied pumps, so consideration should be given to using some of the smaller plant pumps such as those in the RHR system.

3. DFD has a low holding capacity for dissolved metals compared to organic acid processes and LOMI. As a result, more rapid turnover through the IX system is required. Where a cleanup half life of >1 hour may be adequate for other processes, it must be #1 hour for DFD. At MY, for example, it was about 50 minutes. This may require the use of a larger IX system than with other processes, not because of the amount of metals removed, but because of the requirement for a higher flow rate.

4. Fluoroboric acid, HBF_4, is completed dissociated to H^+ and BF_4^-. The BF_4^- partially dissociates to F^- and BF_3 resulting in an aggressive solution. Some of the consequences of this aggressiveness are:
   - Standard neoprene, Buna-N etc. hoses should not be used in the temporary decontamination system. Any hoses in the circulating flow path should be Teflon lined or otherwise protected with an inert lining. Such hose is expensive and currently in short supply, so the temporary decon system tends to have more hard piping than with other processes.
   - The solution is very aggressive to carbon steel making decontamination of mixed metal systems (i.e., carbon steels, stainless steels, Inconel) difficult. Decontamination of mixed metal systems has, however, been successfully accomplished at Big Rock Point. (16)

5. Although the process is aggressive, it is easily controlled. The corrosion rate at any given moment is not that high. On stainless steels it is always <0.1 µm/hr and typically ~0.02 µm/hr. Corrosion rates of Inconel tend to be 3 to 4 times that of stainless steel.

6. Relatively clean water is needed before the first addition of $KMnO_4$. Small amounts of organics can cause decomposition of the $KMnO_4$, which is already at a low concentration (100-200 ppm). In addition, due to the lack of available data, initial boron concentrations had to be reduced to relatively low values before the start of the decontamination (e.g., <40 ppm). Further testing is required in this area to determine if a higher initial boron concentration can be tolerated.

7. $KMnO_4$ as employed in DFD, tends to decompose more rapidly than in Alkaline Permanganate (AP) or Nitric Permanganate (NP). There are several factors that contribute to its decomposition:
   - (i) Base metal comes into solution during the oxidizing step. Thus, there is chromium from both the oxide film and the base metal. Even as the system becomes "cleaner" towards the end of the application, the chromium from base metal dissolution still consumes permanganate.
   - (ii) Base metal dissolution also brings iron into solution. It dissolves in the +2 valence state but in the presence of permanganate is rapidly oxidized to the +3 state. This consumes permanganate.
   - (iv) Chromium tends to build up in solution during a DFD application. It is reduced to the +3 valence state during each reduction phase, but is reoxidized during the oxidation phase of the next cycle.[aa] Thus, as the chromium concentration builds up, the more permanganate that is required to oxidize it.
   - (v) At the end of each transition phase, chromium is thought to remain in solution as an anionic oxalate complex. Thus, in addition to the permanganate that is consumed in oxidizing residual chromium as discussed in (iv), some is consumed through reaction with oxalate.[aa]
   - (vi) Potassium permanganate is quite stable in a solution of pure fluoroboric acid, but dissolved metals catalyze its decomposition.[bb] Since there is typically 10-20 ppm Fe, 5-15 ppm Ni, and 20-30 ppm Cr present in solution during the oxidation phase, there is ample dissolved metal to catalyze the permanganate decomposition reaction.

8. Because of the rapid loss of permanganate during the oxidation phase for the reasons discussed in 7 above, with a long, complex system and low flow rates, the $KMnO_4$ may have decomposed before it reached the end of the system. Oxidation may not have occurred at the farthest locations from the chemical injection point. This phenomenon may have resulted in higher DFs close to the injection point and lower DFs at the most remote locations. Having as a high a system flow rate as possible can minimize this effect.

9. Maintaining a lower level in the VCT would have been beneficial. The uninsulated VCT functioned as a crud trap (due to a standpipe) and caused heat loss due to its significant holdup volume and the relatively long turnover time.

[aa] $Cr(C_2O_4)_3^{3-}$ is very stable and is likely only partially oxidized by permanganate during the oxidation phase.

[bb] Private communication, Dr. D. Bradbury of Bradtec Decon Technologies, 9/19/98.

10. Jumpers around the letdown drag valves in the CVCS would have been beneficial. Although the drag valve internals had been removed, a high pressure drop still remained which limited the flow in the CVCS. Higher flow may have resulted in higher DFs and perhaps fewer DFD cycles.

11. Remote monitoring of radiation fields using teledosimetry worked well in assessing process effectiveness and is highly recommended. However, care must be taken to place the detectors in appropriate locations. Ideal locations are in direct contact with vertical sections of pipe. They should not be located on the outside of shielding or in locations where they are subjected to a background source that is unaffected by the decontamination. Locations on the bottom of pipes or close to deadlegs where MnO_2 may accumulate should also be avoided. MnO_2 is a well-known “getter” for Co-60 and other radionuclides. The true test of their effectiveness is that they can be used for interim dose rate measurements (instead of manual surveys performed by health physics personnel) to determine whether or not additional chemical steps will be required to reach the desired DF/DRF goals.

12. Low flow areas appear to have lower DFs. Adequate motive force should be available to ensure good flow to all locations. Individual DFD cycles are short (8-12 hours) and the change from oxidizing to reducing conditions happens over a few hours. Adequate chemical access and replenishment are essential if the full benefits of the process are to be realized.

13. With adequate flow, it appears that the majority of the oxide film is removed within the first few cycles of DFD. Thereafter, the DF increases mainly through base metal attack. If minimizing the volume of waste IX resin is critical, the number of DFD cycles can be reduced to provide the optimum DF with minimal waste. This may be important on large surface area components such as heat exchangers.

14. For similar decontaminations in the future, it is recommended that some additional flexibility be incorporated in the CVCS flowpath in order to control the temperature in the letdown portion of the system. For example, be able to bypass the charging section of the CVCS so that there is less flow through the charging side of the regenerative heat exchangers.

15. Considerable MnO_2 was formed during the DFD application and settled out in dead legs. It was not completely dissolved during the oxalic acid phase of each cycle. When the “official” cleanup was complete, the plant started to drain the various systems. Copious quantities of MnO_2 were observed in some of the drained liquid. A temporary filtering system was setup by the decon vendor to process the water. Repeated flushing of several system low points was required to achieve clean water. The entire secondary cleanup phase took about four weeks. However this work was done off critical path and had minimal impact on other decommissioning activities. This problem could have been minimized by:
    - During the planning phases, make every effort to minimize deadlegs in the flow path. When establishing the decontamination boundaries, every potential deadleg should be identified and an assessment made at that time as to whether or not there will be a problem with chemical/activity hideout.
    - Having higher flow. A higher linear velocity would have reduced the tendency for MnO_2 to settle in deadlegs.

- Performing a high concentration oxalic acid “flush” in the last DFD cycle, say 500-1,000 ppm versus the 50 ppm typically employed in DFD. In principle, this should dissolve all MnO_2 on system surfaces and in low flow areas, but would have no effect on that which had already settled into deadlegs. In addition, it would increase the waste volume and could potentially create a chelated waste problem where none existed before. The advantages/disadvantages of this recommendation would have to be evaluated on a case-by-case basis.
- Developing a plan to deal with these specific areas at the end of the decontamination. A post-decontamination hot-spot removal procedure could be developed which incorporates such things as high flow draining to prearranged collection tanks, back-flushing into the system during final polishing or flow path modifications near the end of operations.
- Perform a thorough final survey not just for post-decontamination DF measurements but also for unanticipated hot spots. This should be completed before the end of operations so that the decontamination equipment can be used to support whatever adjustments are required.
- Plan for the effort that may be necessary after the decontamination is complete and the equipment disconnected to deal with areas "manually".

16. Field changes to operating procedures will undoubtedly be necessary during the decontamination. A mechanism needs to be in place in advance to respond to required changes quickly and efficiently.

17. Mockup training for potentially high dose tasks was extremely useful. Three such tasks which were successfully mocked up prior to their actual performance were:
    - Steam generator temporary manway installation
    - High Integrity Container handling and transport
    - Filter cartridge replacement

18. PN Services was responsible for providing their own craft labor support. This improved the co-ordination between the vendor and their craft support. It also placed what has been traditionally a time and materials cost to the utility under the vendor's fixed price umbrella.

19. During the planning stages for the decontamination, Maine Yankee project staff were able to declassify all of the systems to be decontaminated as non-nuclear safety (NNS). This proved to be an extremely useful and time-saving tool for the preparation of procedures and work packages.

20. The nozzle dam spider assembly was an effective tool for bypassing the reactor pressure vessel. The only improvement recommended was for more insulation to be installed to reduce heat loss in the jumper lines interconnecting the dams. A significant amount of energy was transferred to the water in the reactor vessel cavity during high temperature operations.

Connecticut Yankee

1. If a decontamination for decommissioning is to be performed after a station has been permanently shutdown from power operation, it should be completed as soon as possible after that shutdown. After prolonged periods (i.e., greater than 6 months), problems may be encountered with the state of readiness of station equipment and the availability of sufficient experienced station personnel to plan for and execute the decontamination. Under ideal circumstances, the decision to enter into such a project should be made while the plant is still running so that the decontamination can be performed as soon as possible after shutdown.

2. Satisfactory performance of a full loop decontamination requires the co-operation and commitment of all station departments in the earliest possible stages of the project. It is very important to elicit support from each when the initial project team is being formed.

3. If foreign nationals are to be hired as decontamination contractors, there will be additional time, effort and cost associated with some of the administrative aspects of having them on site (i.e., badging). In some cases it may not be possible to obtain all the information required by NRC regulations for issuing of a site badge and full time escorts may have to be provided.

4. Careful consideration should be given as to whether or not station equipment such as pumps, ion exchange vessels, and heaters should be used to support decontamination operations. If the decision is made to use such equipment, their maintenance routines should be kept up through completion of the project. In addition, functional testing and detailed inspections of that equipment should be performed in advance of the application to minimize the potential for breakdowns while chemicals are circulating. Calibrations for the appropriate in-line instruments should also be kept up to date. In summary, use of existing installed station equipment offers several advantages over vendor-supplied equipment, provided it can be relied upon to operate satisfactorily throughout the course of the decontamination. The potential advantages include:
    - less vendor equipment so easier and faster equipment mobilization/demobilization
    - higher flow rates since vendor-supplied pumps are invariably smaller than even the smallest station pump
    - possibility of combining all systems to be decontaminated into a single flow path thus allowing for a single application rather than multiple applications
    - no shielding for temporary IX columns since the station's IX columns are already in shielded locations
    - no temporary resin sluice lines since the stations IX columns are directly connected to the spent resin storage tank
    - lower vendor charges

    Potential disadvantages include:
    - generally the station's IX columns are not large enough to accommodate the waste from a full loop RCS decontamination and several resin changeouts would be required. The situation at CY was somewhat unique in that the station's IX capacity was larger than normal and the CORD processes uses significantly less IX resin than other processes

because of its unique UV/H_2O_2 decomposition of oxalic acid.

- if problems develop with station equipment they are much more complicated and time consuming to correct than equivalent problems with the vendor's equipment. This was demonstrated several times during this decontamination.
- less flexibility in making changes to the system such as valving in and out IX columns. The vendor can do this very quickly but station procedures are much more complicated and time consuming. As a result Siemens did not request valving out an ion exchange column for an hour or two when it would have been appropriate to do so. This would have prevented release of some iron from the column.

5. The decision to use the plant's RHR pumps as the main motive force was a good one. They provided far higher flows than could be achieved with portable vendor-supplied pumps and were, in part, responsible for the satisfactory DFs obtained after two cycles.

6. The decision to use the plant's pressurizer heaters as the primary source of heat was a good one. These heaters functioned satisfactorily and reduced the amount of vendor-supplied equipment that had to be brought to site.

7. Another advantage of using vendor-supplied IX beds is flexibility. To apply the CORD process with maximum efficiency, the IX beds need to be valved in and out frequently with short notice. If the vendor is in charge of this operation, it can be performed very quickly. If the plant is in charge, the process is more involved and may take up to several hours. As a result, the beds were not always taken off line when it would have been appropriate to do so.

8. If the plant's IX system is used as the primary cleanup system for the decontamination, a vendor-supplied backup is strongly recommended. Any problems with the plant's IX system can have very adverse consequences on the process schedule and the vendor's ability to satisfactorily complete the decontamination. However, this decision needs to weighed against the added exposure and the need for additional shielding, berming, sluice lines, etc.

9. The CORD process may, at times, leach iron from IX columns, especially if they have not been able to be isolated as discussed in (7) above. Although this did not result in any adverse effects and it was eventually reabsorbed by the IX beds, it did come as a surprise to some non-Siemens chemistry personnel who were not expecting it and thought that it was a sign of IX bed saturation. This was not the case.

10. An independent, detailed review performed by a knowledgeable outside group can be a very useful tool to determine the state of readiness prior to the start of the application. This group can ensure that lessons learned from similar decontaminations have been properly addressed.

11. If permanganic acid will be used to perform oxidizing steps during the decontamination, it should be stored in a cool, dry location. Under hot conditions (>90°F) the sealed containers may overpressurize and start to leak. Some decomposition of the permanganic acid may also occur.

12. A decontamination can mobilize measurable quantities of fine particulate matter which will collect in filters and ion exchange resin around the system. Filter media with appropriate

porosity (i.e., 20–50 µm) should be used to avoid premature plugging and unnecessary additional changeouts that may occur with very low porosity filters (i.e., 1-5 µm).

13. If changes to the original flow path will be required to ensure that all of the desired plant equipment will be exposed to the circulating chemicals, they should be implemented as early in the operating sequence as possible. This will ensure adequate exposure time for all of the piping and equipment which was planned for decontamination.

14. Judiciously placed remote TV cameras were of great value in monitoring for small system leaks. They were responsible for the detection of small leaks in the pipe trench long before they could be detected through volume and pressure changes.

15. The CORD process is relatively "forgiving" and can handle prolonged system upsets without significant adverse consequences. For example, very little corrosion of system materials occurred, despite the very long CORD cycles (305 hours in Cycle 1 and 334 hours in Cycle 2). It has been estimated that 20 pounds of base metal was dissolved as a result of corrosion. The maximum values are equivalent to penetrations of 0.3 µm (0.01 mils) and 0.6 µm (0.02 mils) on stainless steel 304 and Inconel 600, respectively.

16. CORD was able to function with a very much reduced IX cleanup rate. At one stage it was only 63 gpm in a 31,000 gallon system. This corresponds to a purification half-life of 5.7 hours. The step took longer to complete, but nonetheless was completed satisfactorily.

17. Negligible MnO_2 was generated during the oxidizing steps with permanganic acid. The amount that was produced corresponded to the amount of chromium oxidized. There was no catalytic decomposition of the permanganate. Essentially 100% of the manganese that was added was recovered on IX resins during the following step.

18. The CORD process can operate in the presence of up to 400-ppm boron without adverse consequences. In addition, based upon the results of artifact testing, the process appears capable of operating satisfactorily in up to 2,000-ppm boron.

19. The CORD process produces a relatively small amount of waste IX resin. The controlling factor is the amount of metals removed by the decontamination. Of the three decontamination chemicals brought to site, the only one that consumes IX capacity is the manganese in the permanganic acid. The other two are converted to gases or water.

20. Controlled use of the UV option during Step 3, Decontamination, of the CORD process (not CORD "D") was very useful in reducing Fe^{3+} to Fe^{2+}. This ensured its efficient removal by cation exchange resin. The ferric-oxalate complex is very stable and is not readily broken down by cation resin.

21. Although the Siemens procedures and techniques are certainly adequate for controlling and monitoring the CORD process, they are inadequate for determining the total concentration of oxalate present prior to final cleanup. As a result, the amount of oxalate on the mixed bed IX resin could only be estimated. Since a conservative estimate was used, this shortcoming may

have resulted in the resin from the decontamination being unnecessarily designated a chelant-containing resin, i.e. >0.1% by weight oxalic acid.

22. The H_2O_2 decomposition process was able to reduce free oxalate to acceptably low levels (<20 ppm),[cc] but total oxalate values were ~120 ppm due to the relatively stable chromium oxalate complex. This was not low enough to prevent the resin from being considered chelated waste. However, the Siemens analytical technique was only able to provide an estimate of the total amount of oxalate. The technique of direct titration of the sample with permanganate gives a "permanganate demand" value but conversion of this to a total oxalate concentration is complicated by the presence of Cr^{3+} in the sample which also reacts with permanganate. In addition, the exact nature of the chromium-oxalate complex at this stage of the process is not well characterized.

    A recommended approach is to precipitate all available oxalate as the insoluble calcium salt through the addition of $CaCl_2$ or $CaSO_4$ followed by filtration through a glass fiber filter.[dd] Titration of the filtered calcium oxalate with permanganate will give an accurate measure of the **available** oxalate remaining in solution. This is the value that should be used for calculating the chelant loading on the mixed bed resin. A sound technical case can be made for claiming that any oxalate that does not precipitate in the presence of excess calcium is so strongly tied up with chromium that it can no longer be considered a chelant. According to Siemens, this position has been accepted by regulatory authorities in Sweden and Japan but they were unable to supply any supporting documentation.

23. The installation of a remote dosimetry system was useful in reducing the amount of radiation exposure required to monitor the reducing radiation levels during Cycles 1 and 2.

24. If the plant has high transuranic activity in the oxide film, resin loading will need to be monitored very closely to ensure acceptability of the waste form.

[cc] For Cycle 2, this value was amended to <100 ppm to shorten the decomposition/cleanup time.

[dd] This is a well-known, standard analytical technique for the determination of oxalate in solution. It has been used many times in the past with CITROX, CAN-DEREM™ and oxalate rinses following AP or NP.

11

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Appendix A

Graphs of Experimental Data Obtained During DFD Application 1 at Maine Yankee

Figure A-1
Dissolved Co-60 Concentration During Application 1

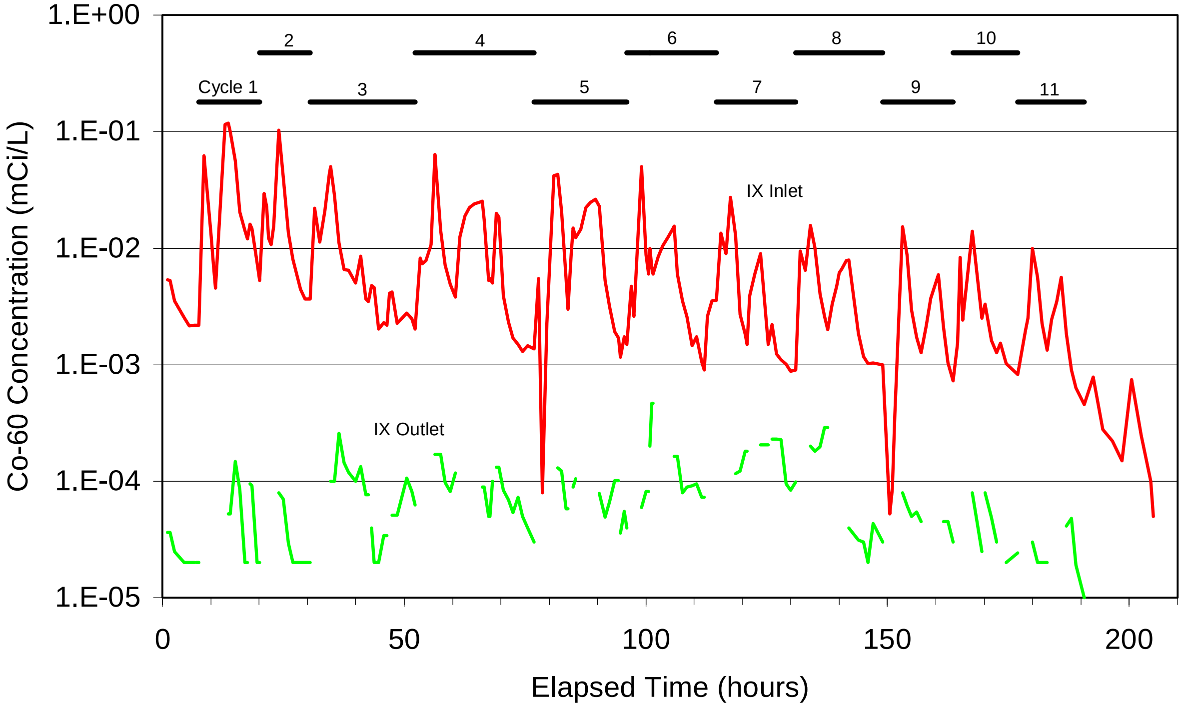


Figure A-2
Dissolved Iron Concentration During Application 1

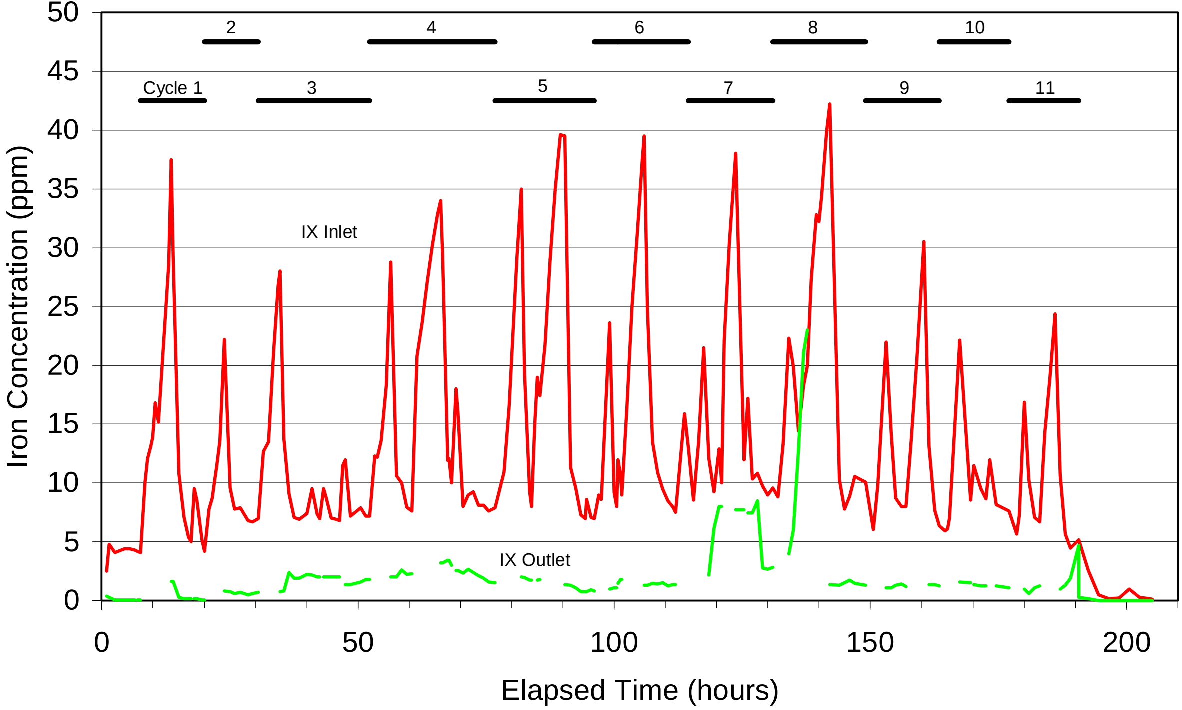


Figure A-3

Dissolved Nickel Concentration During Application 1

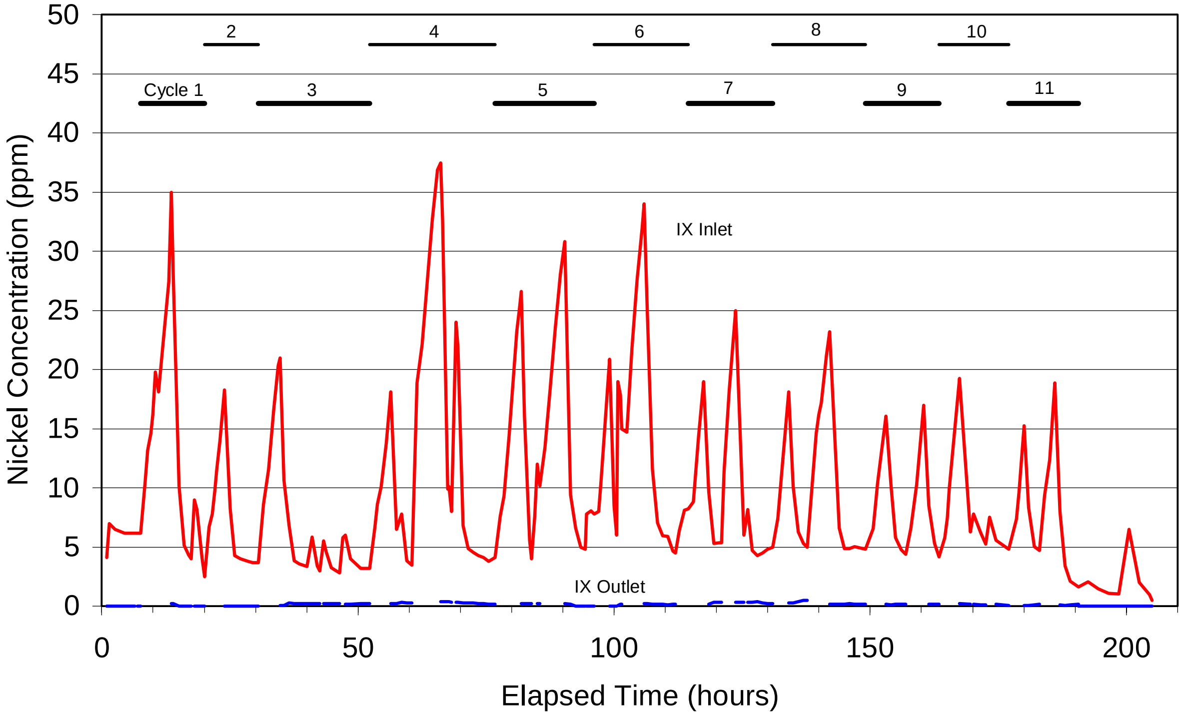


Figure A-4

Dissolved Chromium Concentration In Application 1

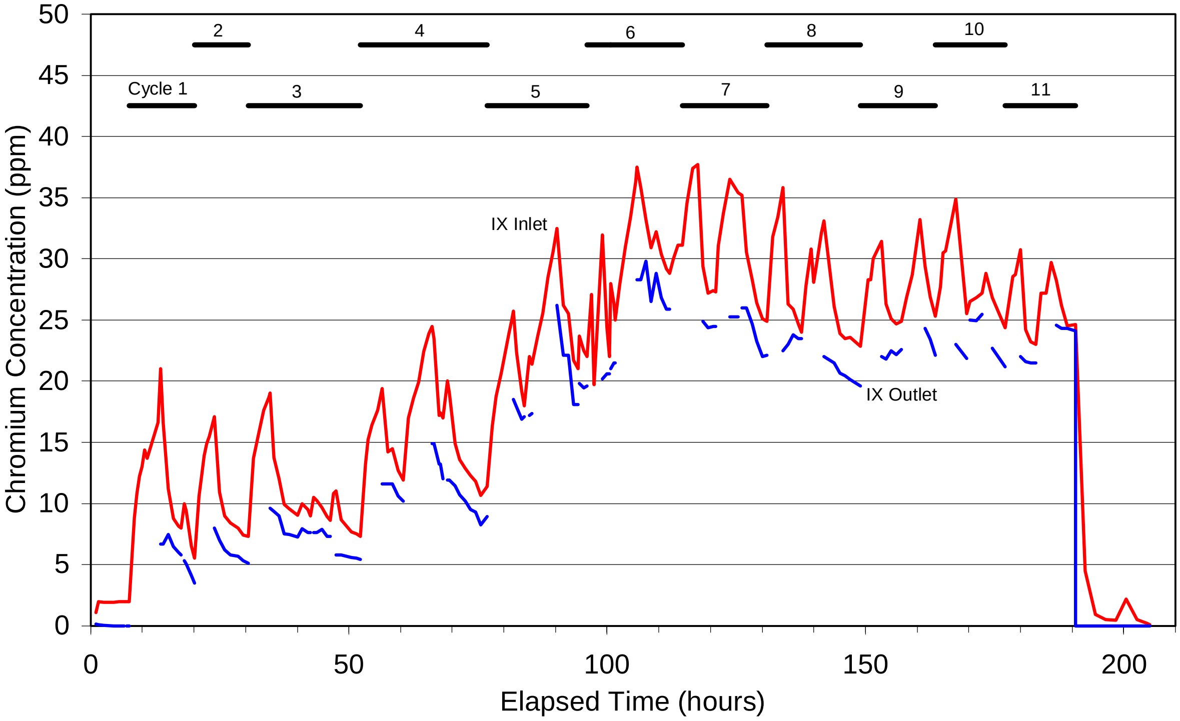


Figure A-5
DRF in Vicinity of Valve LD-1 - Application 1
(Teledosimetry)

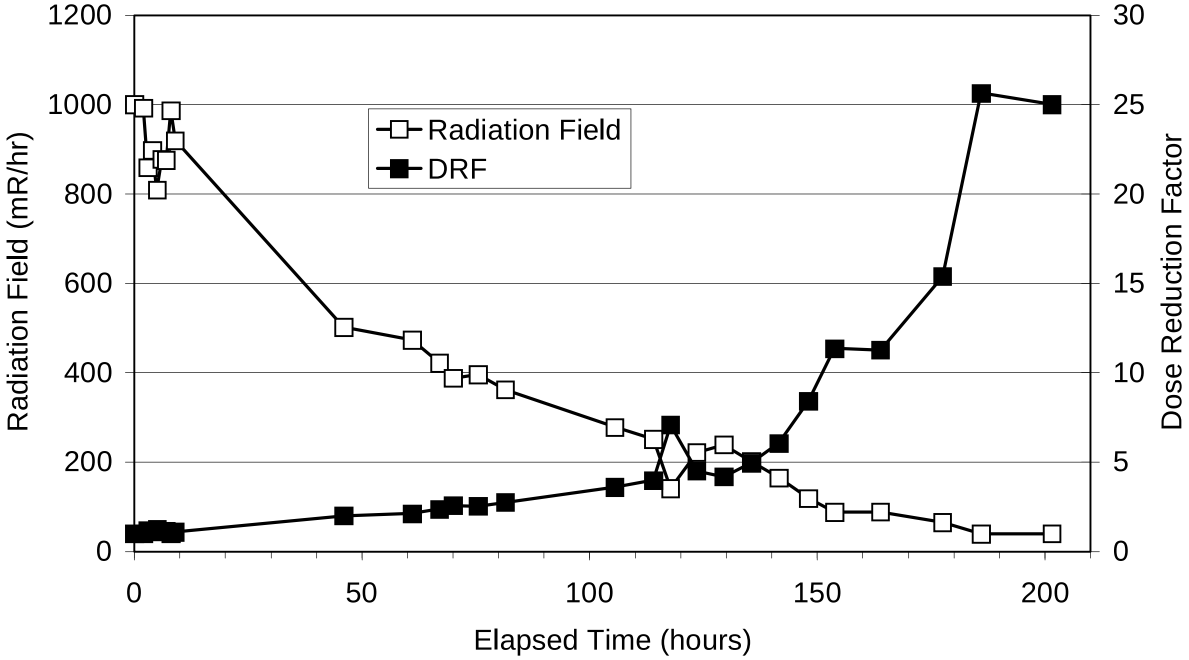


Figure A-6
DRF in Vicinity of Valve CH-M-46 - Application 1
(Teledosimetry)

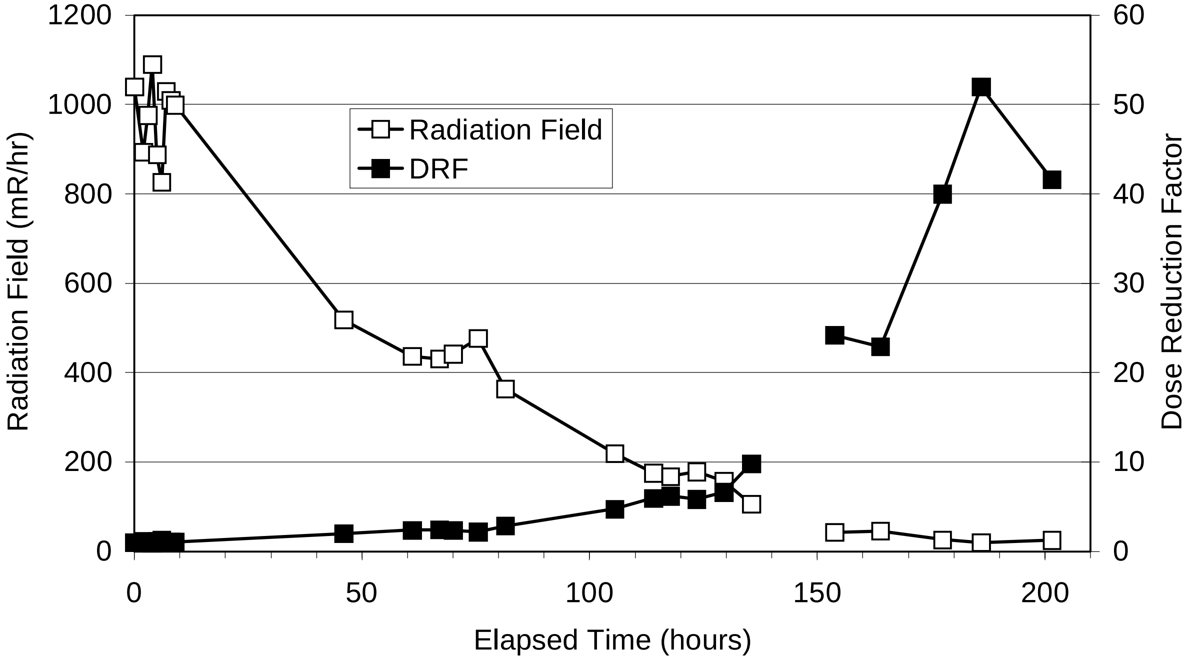


Figure A-7
DRF in Vicinity of Pressurizer Spray Riser Line - Application 1 (Teledosimetry)

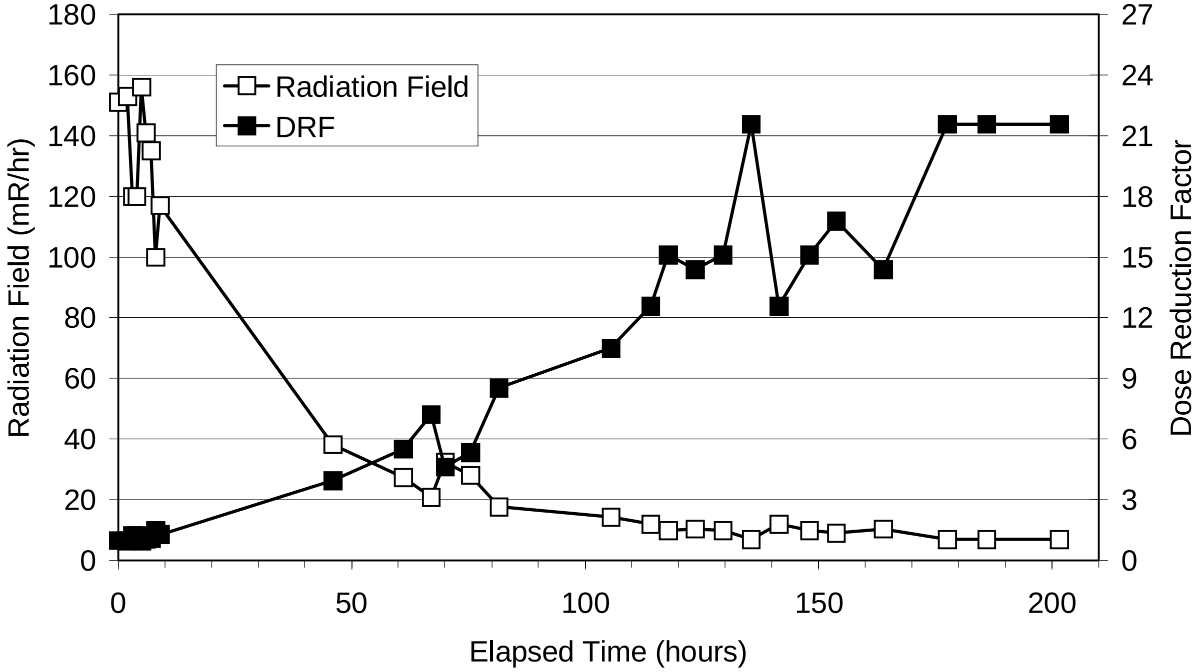


Figure A-8
Contact DF Point 13 - Letdown Line

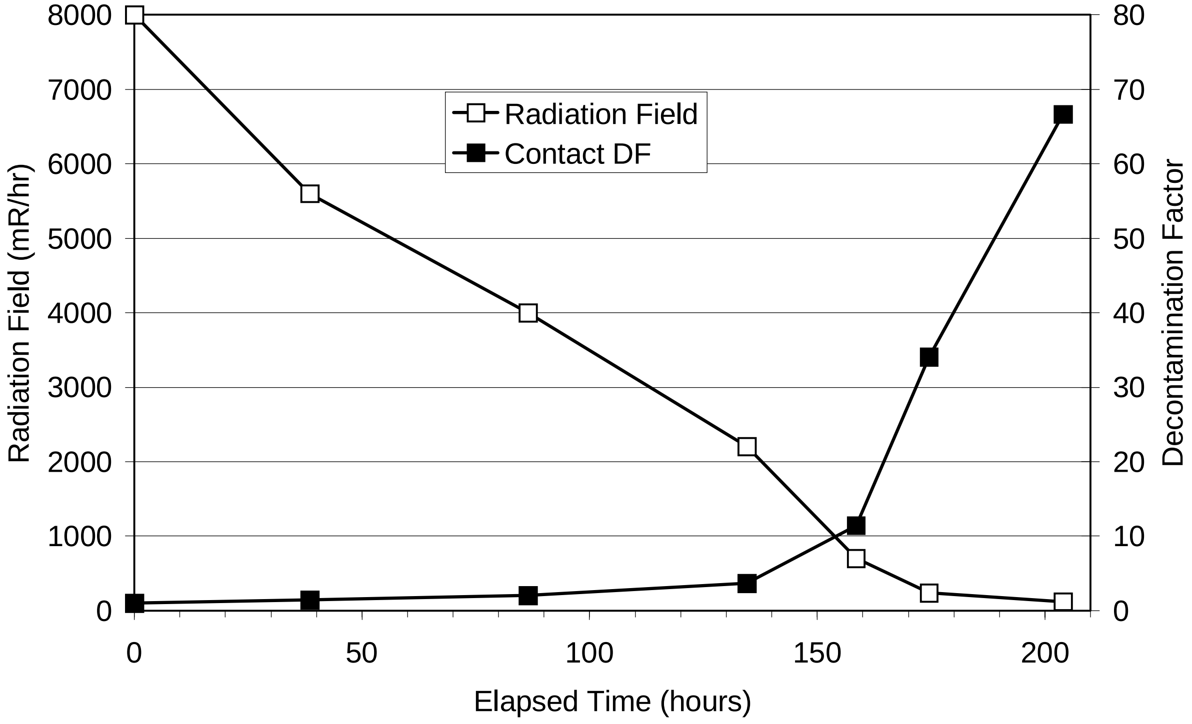


Figure A-9
Contact DF Point 21 - Loop 2 Bypass Line

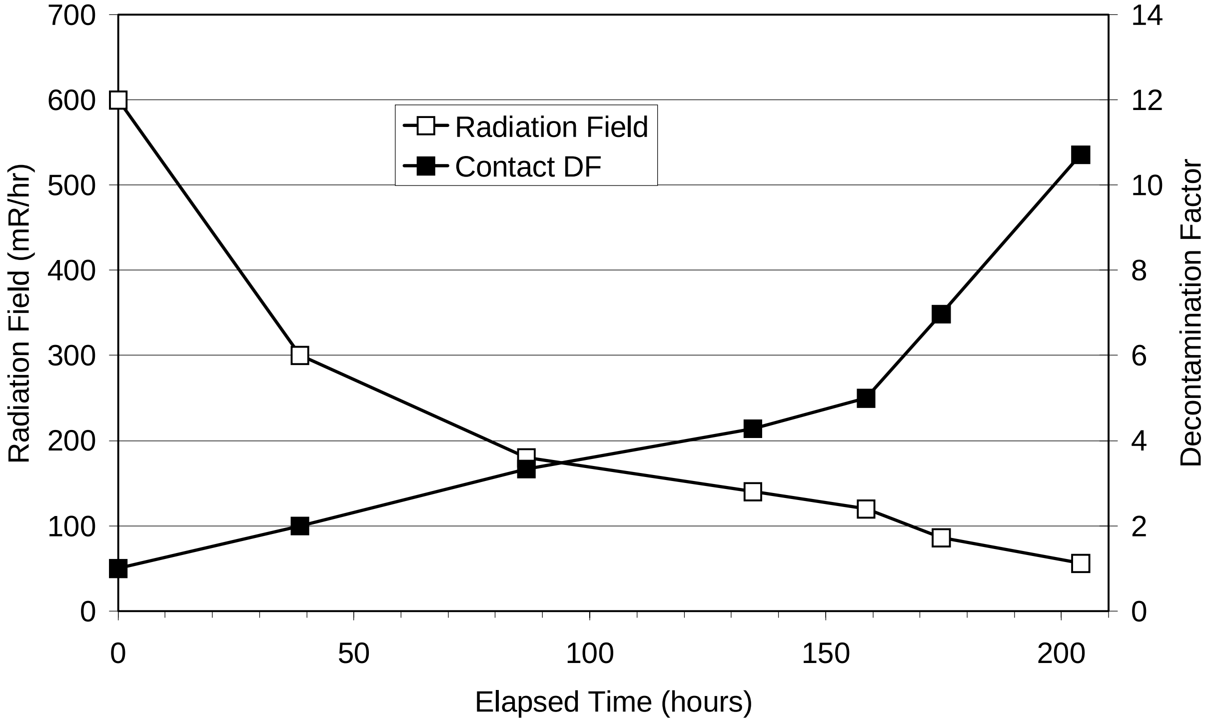
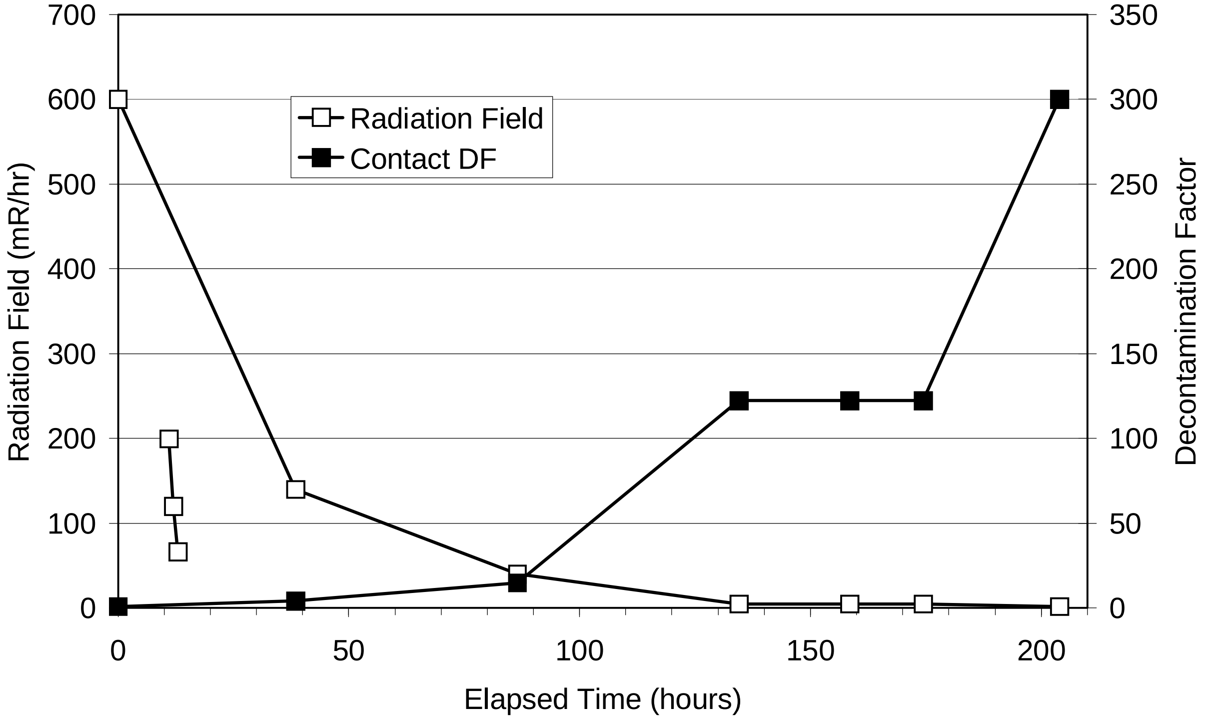


Figure A-10
Contact DF Point 39 - Pressurizer Line



Appendix B

Graphs of Experimental Data Obtained During DFD Application 2 at Maine Yankee

Figure B-1
Dissolved Co-60 Concentration During Application 2

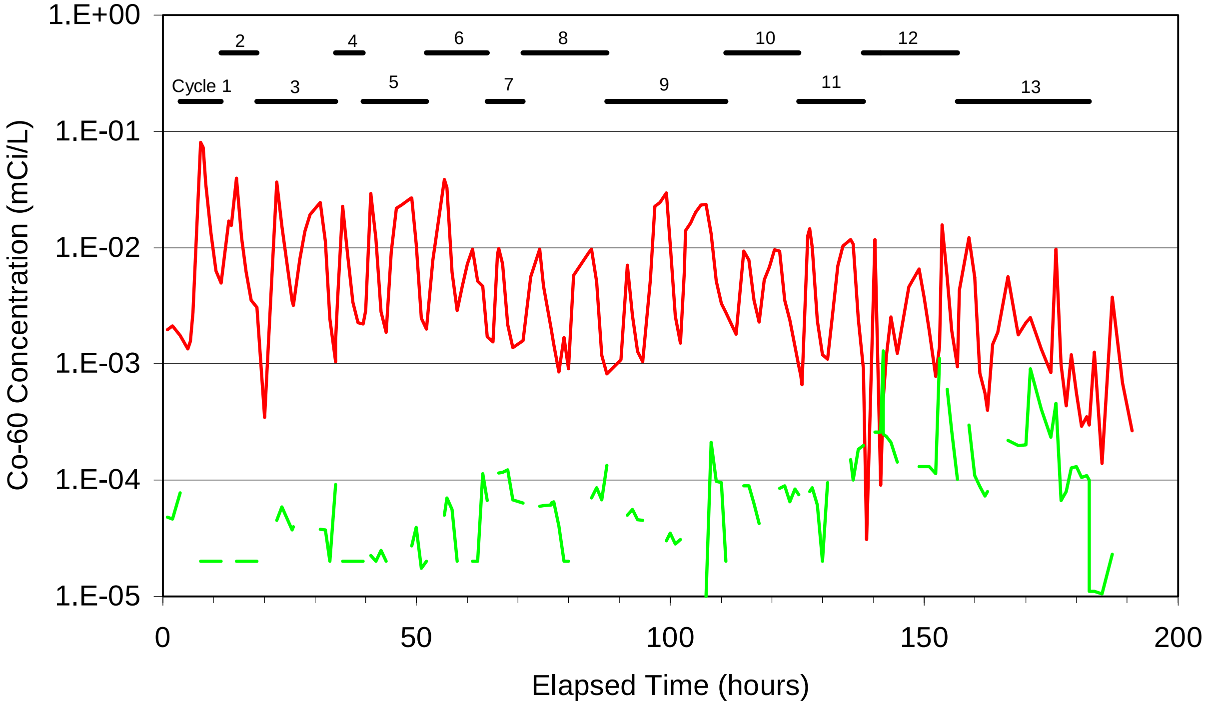


Figure B-2
Dissolved Iron Concentration During Application 2

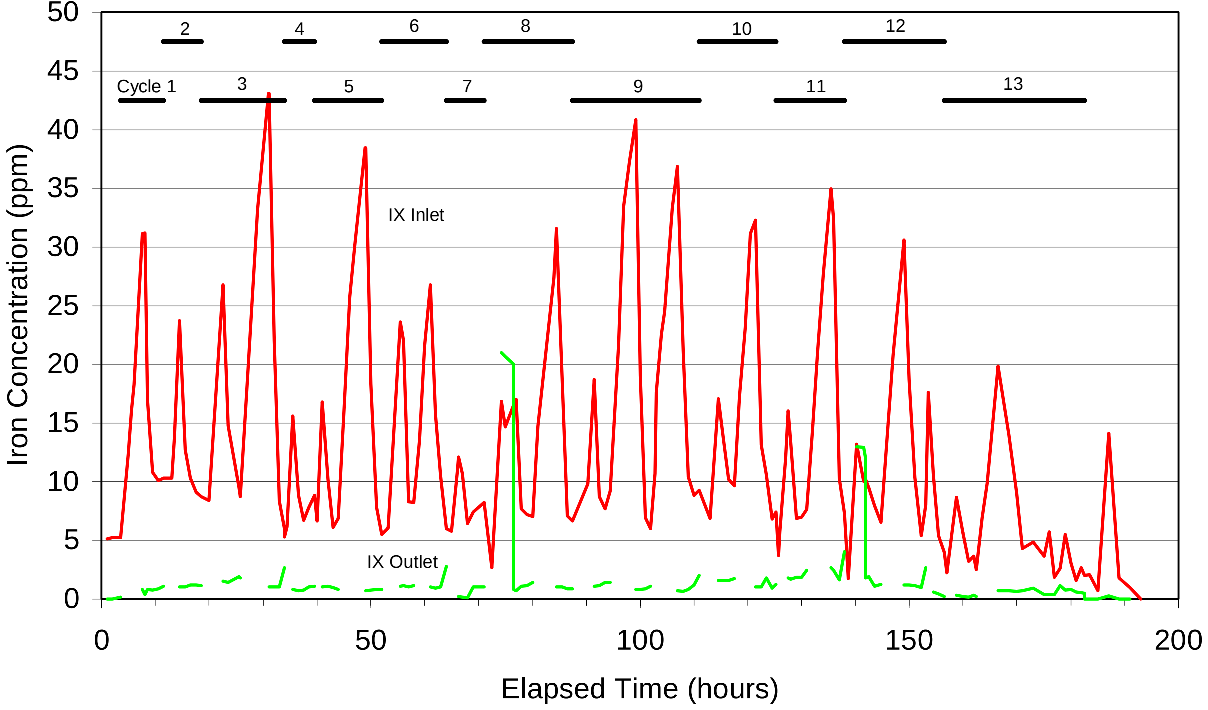


Figure B-3
Dissolved Nickel Concentration During Application 2

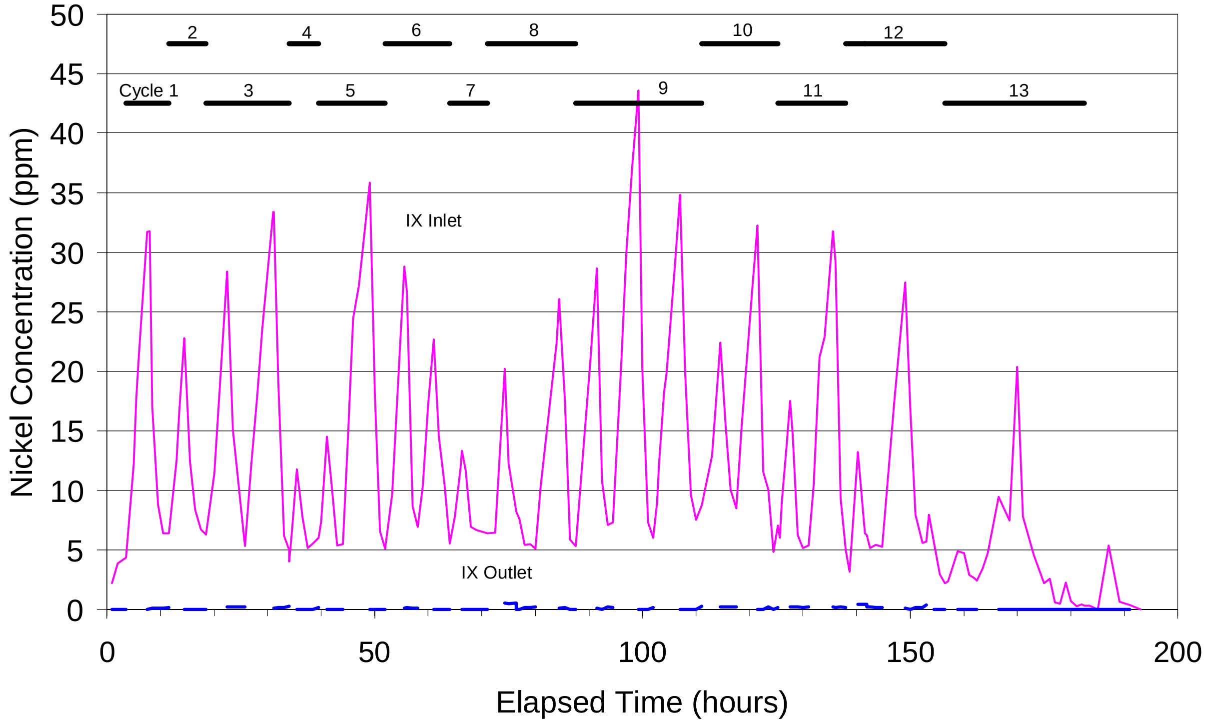


Figure B-4
Dissolved Chromium Concentration In Application 2

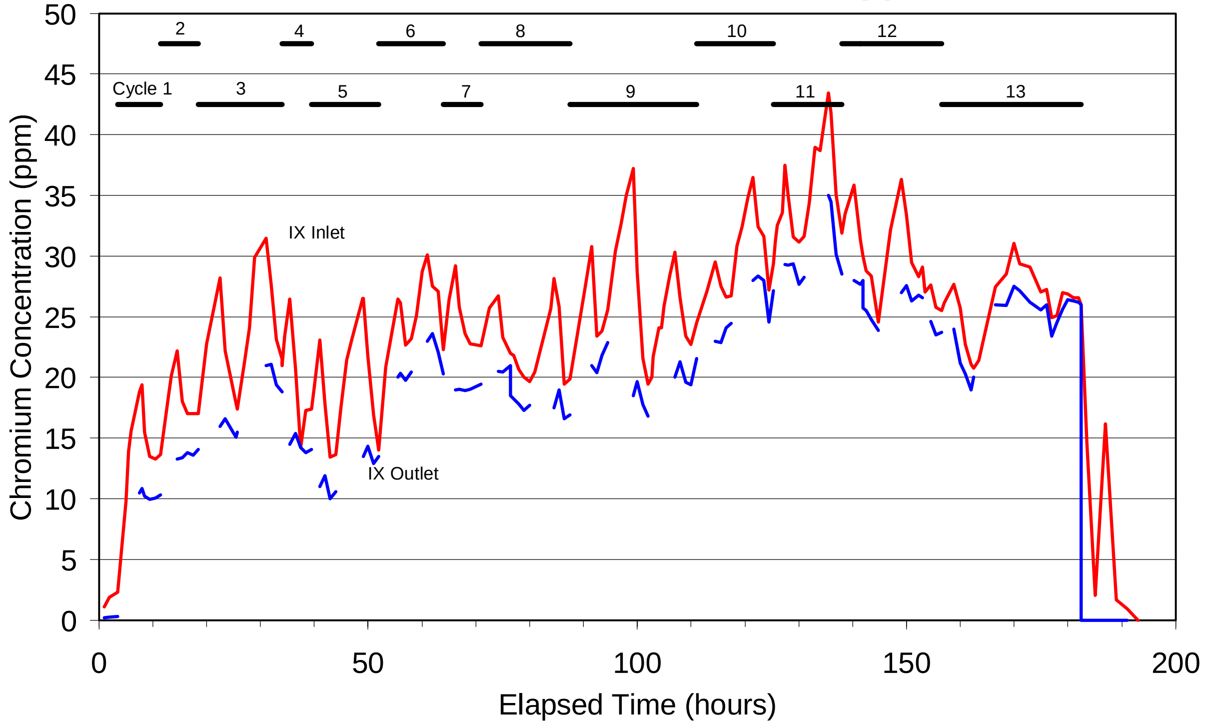


Figure B-5
DRF in Vicinity of 8" Bypass Loop 3 - Application 2
(Teledosimetry)

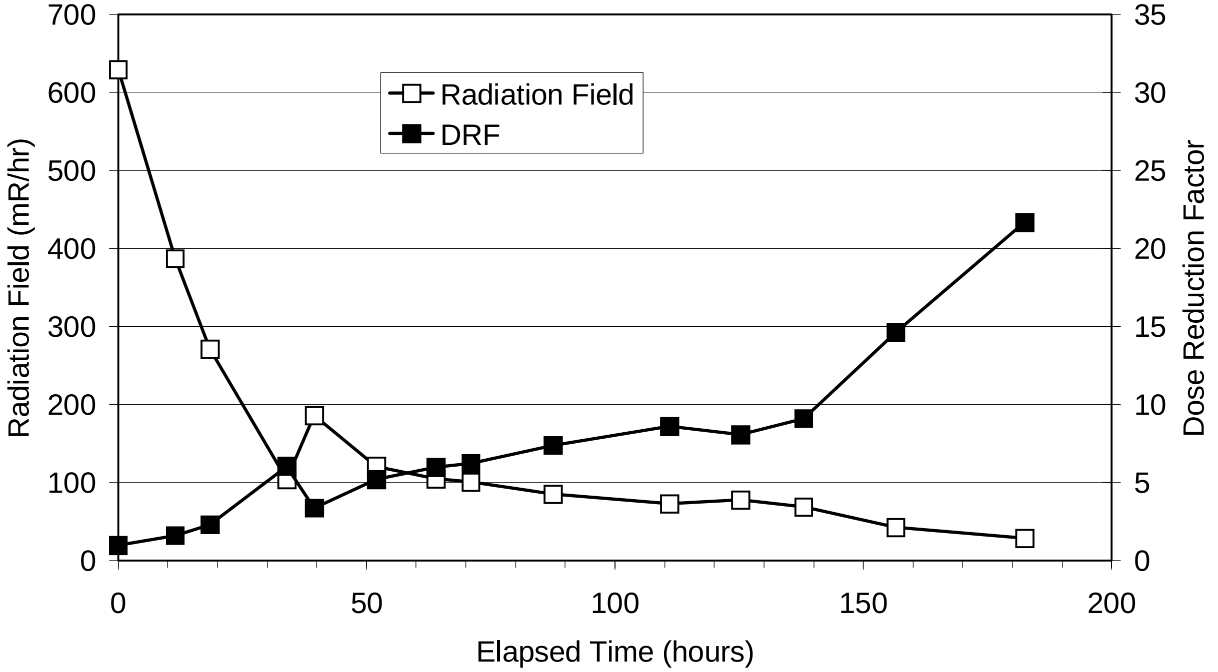
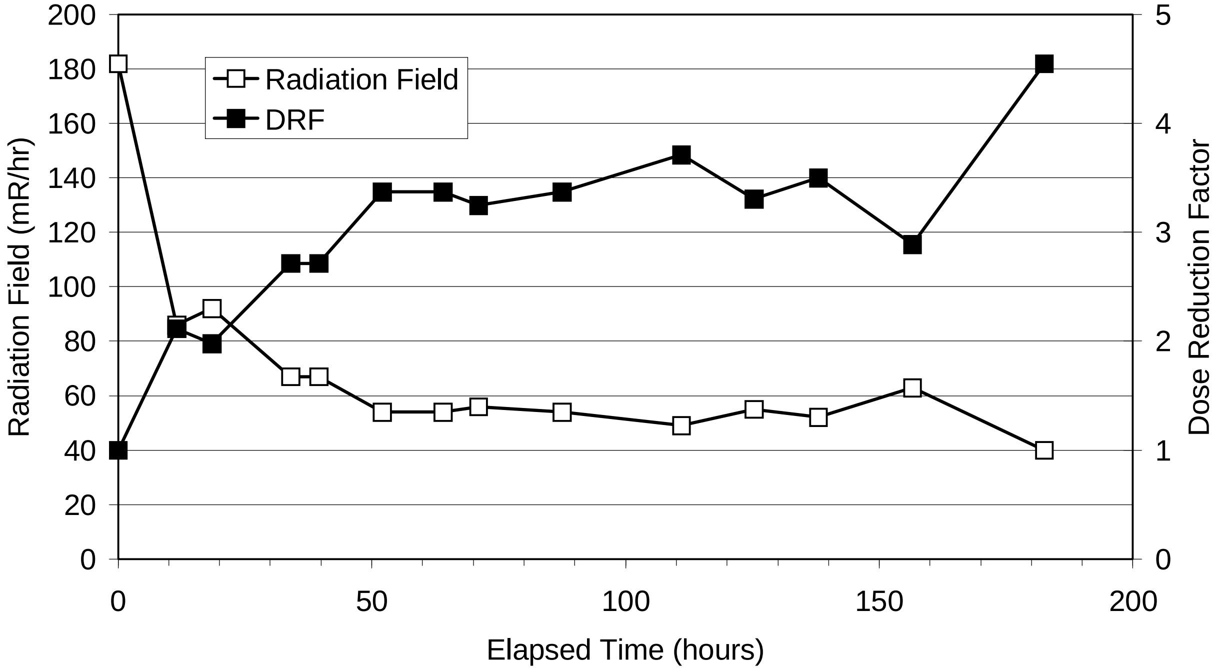


Figure B-6
DRF in Vicinity of Drain Header - Application 2
(Teledosimetry)



Appendix C

Graphs of Experimental Data Obtained During CORD Cycle 1 at Connecticut Yankee

Figure C-1
CORD Cycle 1 - Metal Concentrations

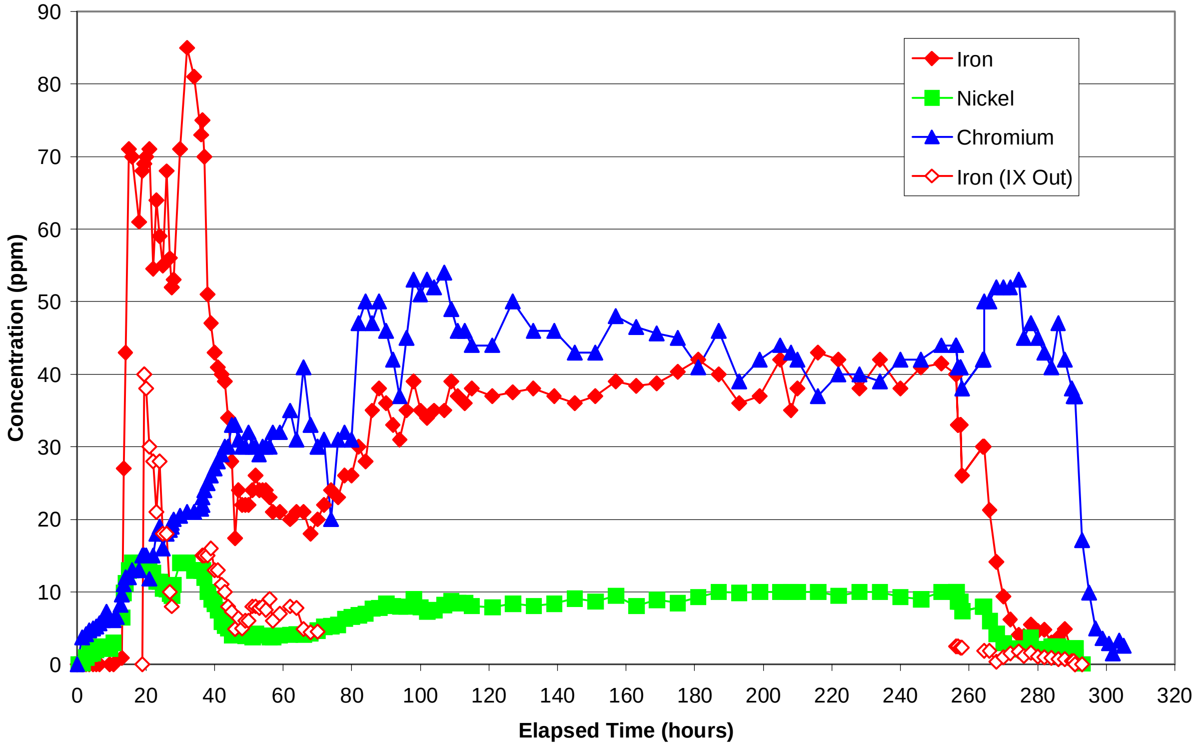


Figure C-2
CORD Cycle 1 - Radionuclide Concentrations

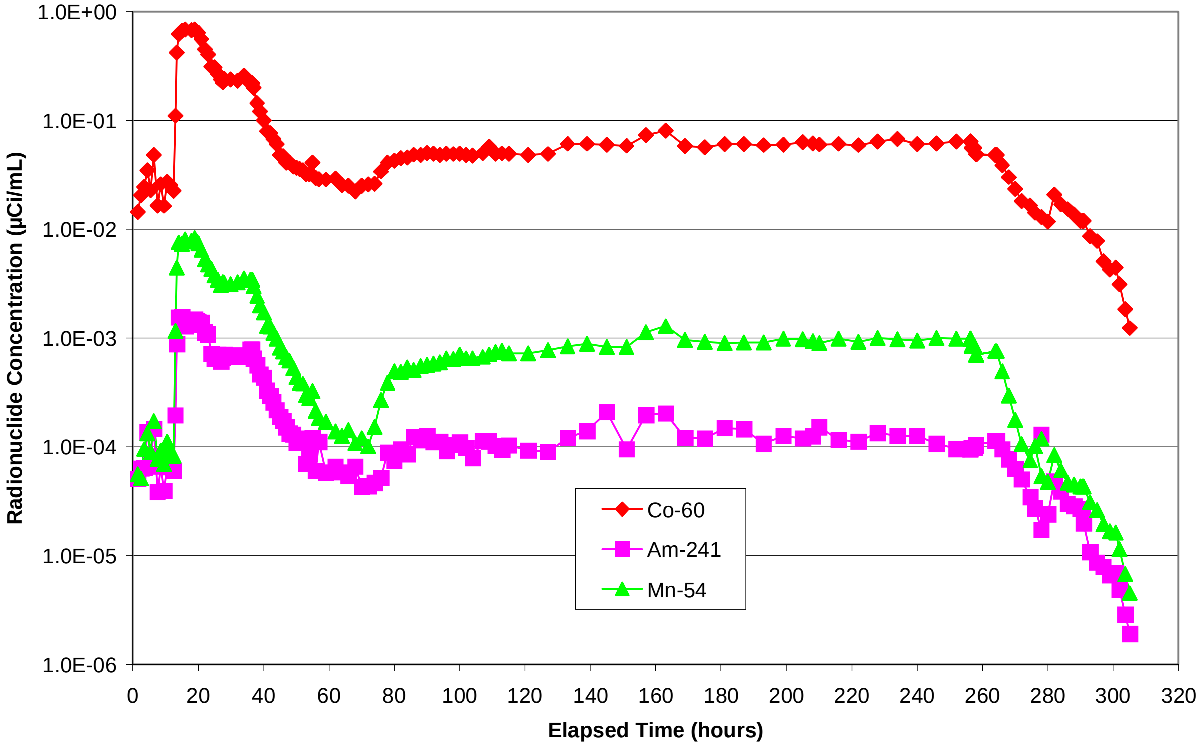


Figure C-3
CORD Cycle 1 - Concentration of Oxalic Acid

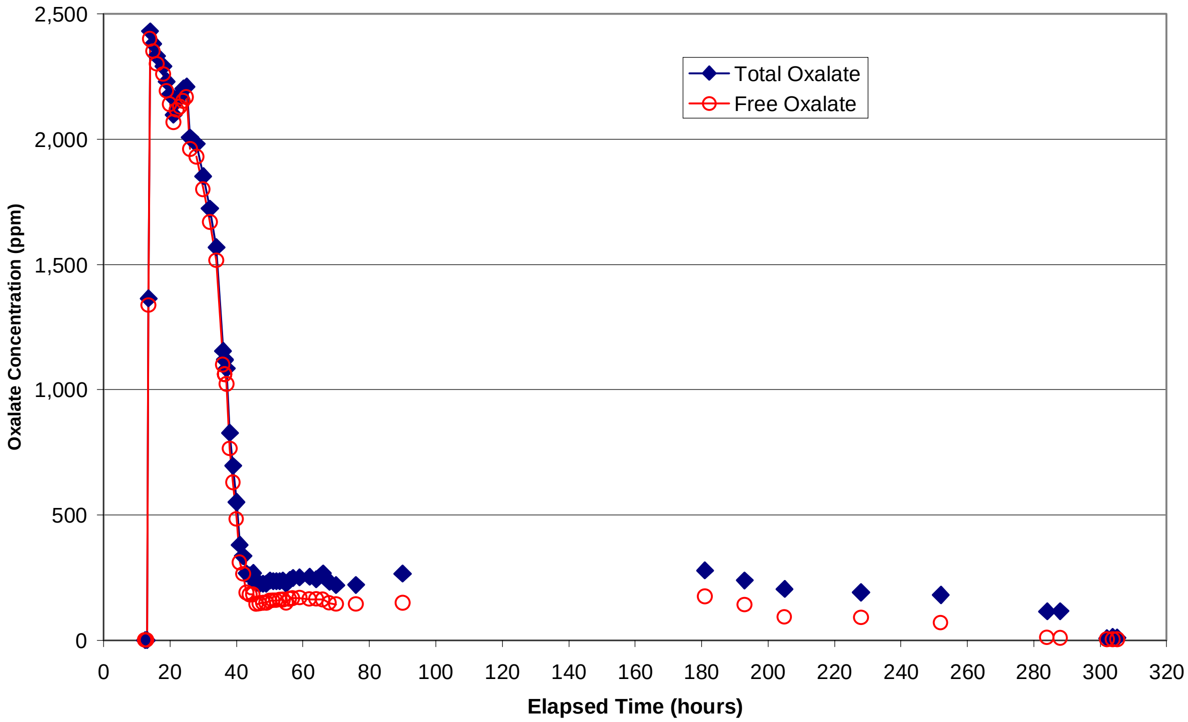
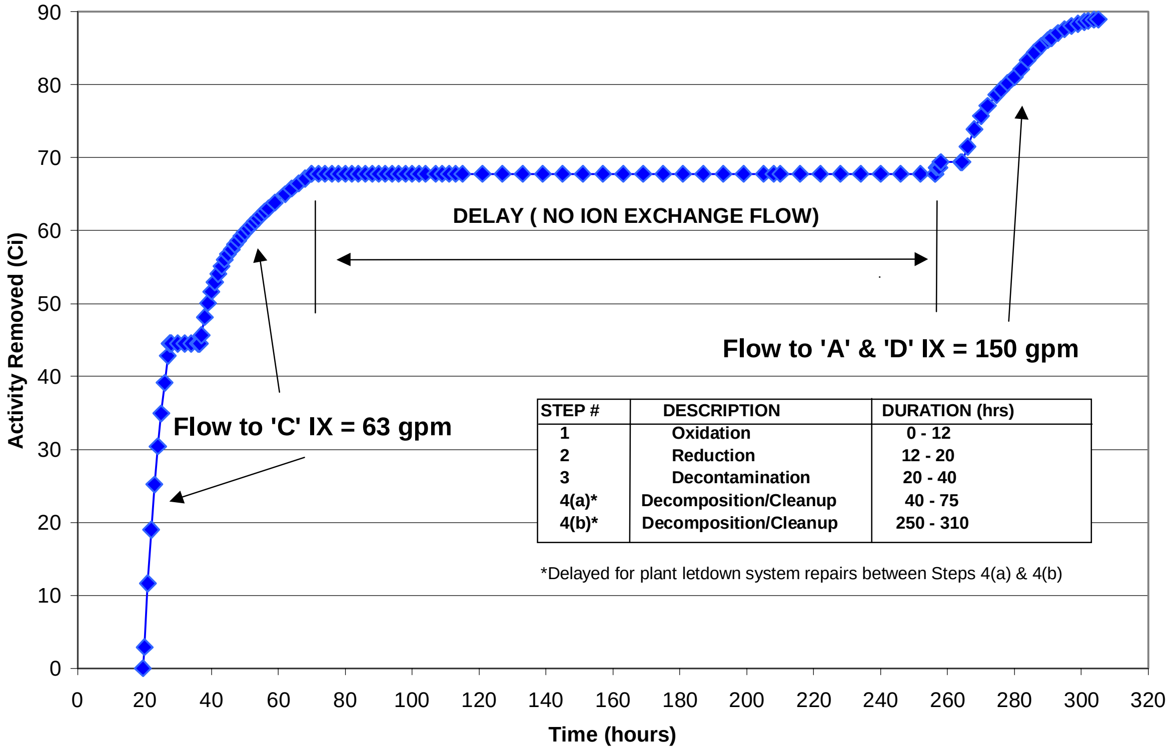


Figure C-4
CORD Cycle 1 - Total Activity Removed on IX Resin



STEP #	DESCRIPTION	DURATION (hrs)
1	Oxidation	0 - 12
2	Reduction	12 - 20
3	Decontamination	20 - 40
4(a)*	Decomposition/Cleanup	40 - 75
4(b)*	Decomposition/Cleanup	250 - 310

*Delayed for plant letdown system repairs between Steps 4(a) & 4(b)

Appendix D

Graphs of Experimental Data Obtained During CORD Cycle 2 at Connecticut Yankee

Figure D-1
CORD Cycle 2 - Metal Concentrations

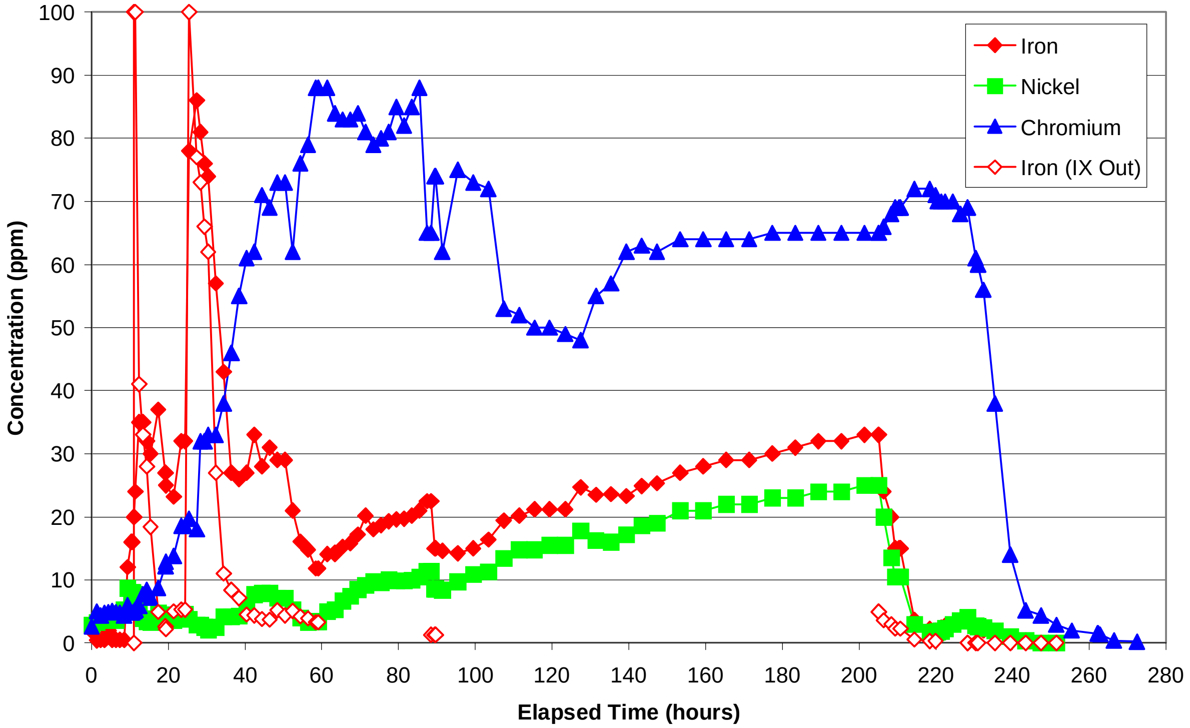


Figure D-2
CORD Cycle 2 - Radionuclide Concentrations

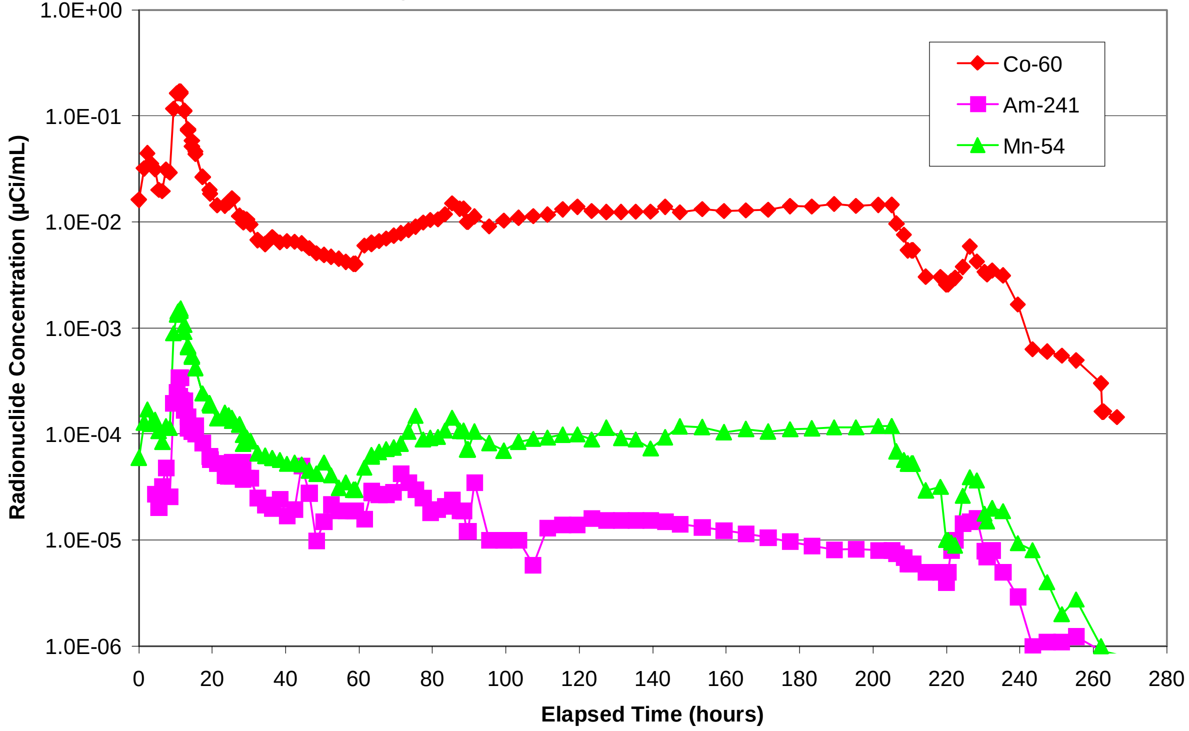


Figure D-3
CORD Cycle 2 - Concentration of Oxalic Acid

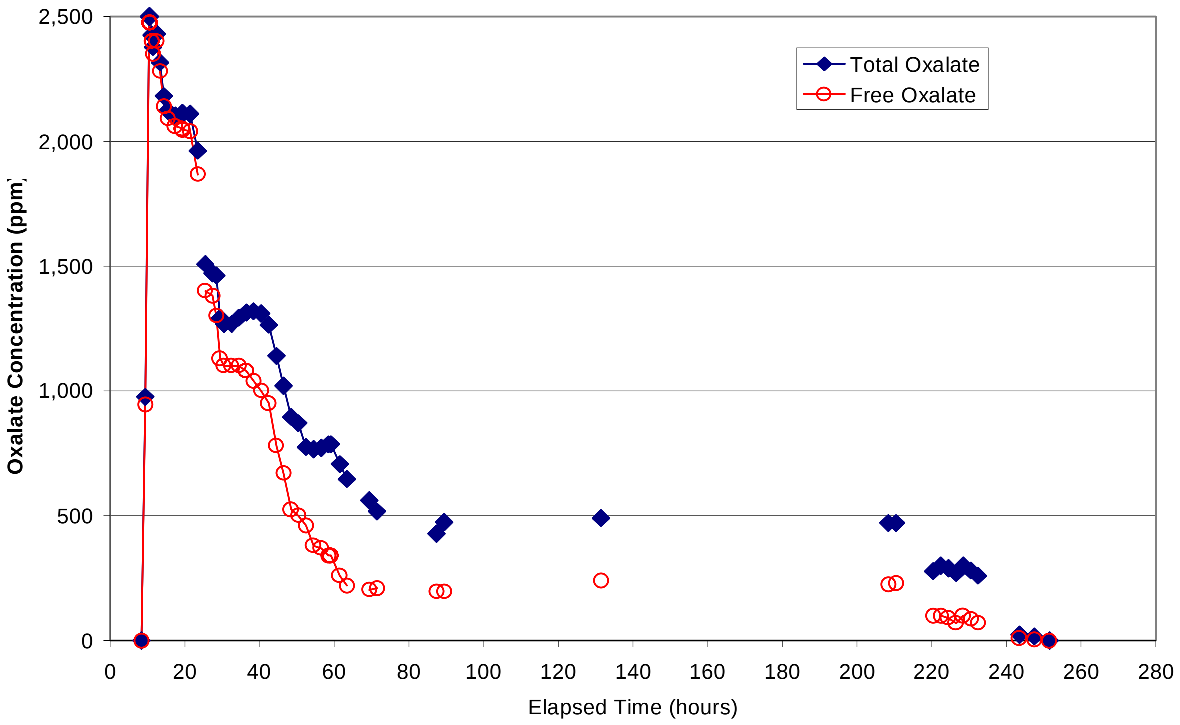
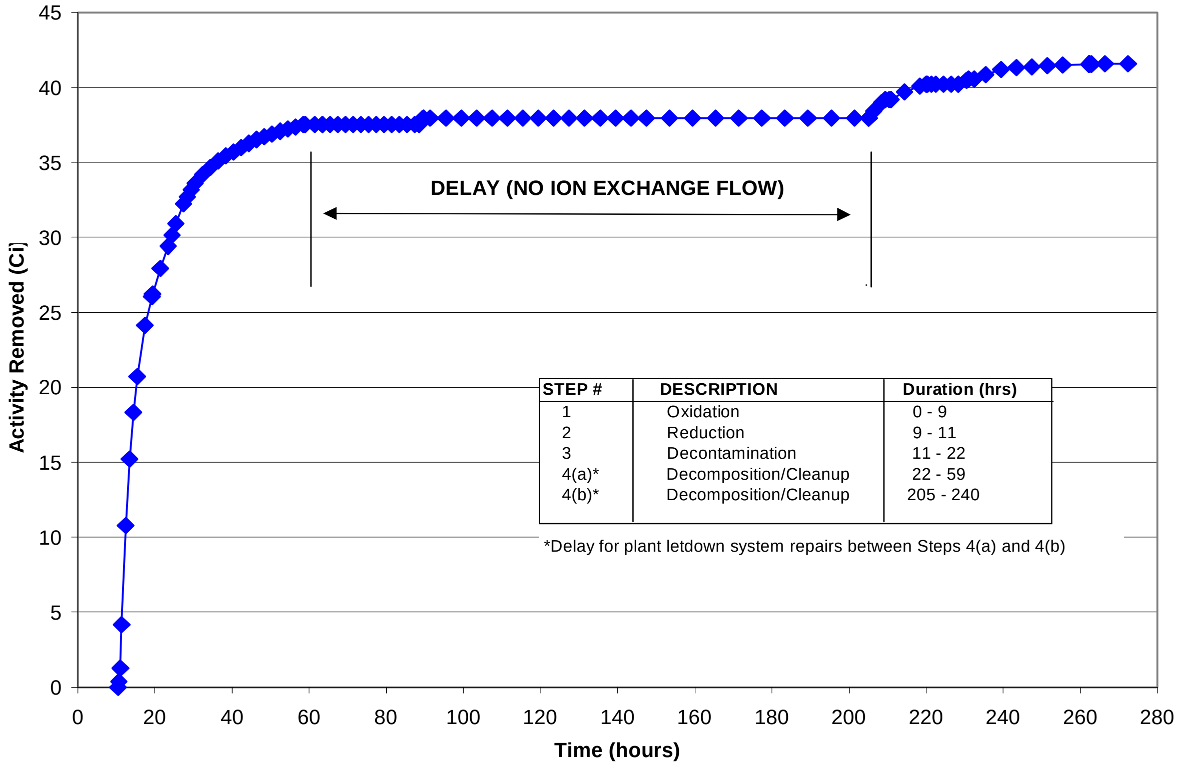


Figure D-4
CORD Cycle 2 - Total Activity Removed on IX Resin



STEP #	DESCRIPTION	Duration (hrs)
1	Oxidation	0 - 9
2	Reduction	9 - 11
3	Decontamination	11 - 22
4(a)*	Decomposition/Cleanup	22 - 59
4(b)*	Decomposition/Cleanup	205 - 240

*Delay for plant letdown system repairs between Steps 4(a) and 4(b)

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