REAL TIME WASTE CHARACTERIZATION DURING A FULL SYSTEM DECONTAMINATION

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ABSTRACT
In December of 1996, the joint owners of the Connecticut Yankee (CY) - Haddam Neck Plant decided to permanently shut down the plant. Due to the design of the plant and its operating history, the personnel exposure estimated to decommission the plant made it appropriate to perform a full system decontamination prior to commencing dismantlement activities. The topic of this paper is the difficult challenges presented in handling the waste to be generated during this decontamination and how a monitoring system was developed to characterize the waste on essentially a real time basis.

INTRODUCTION
In December of 1996, the joint owners of the Connecticut Yankee Atomic Power Company, Haddam Neck Plant (CY) decided to permanently shutdown and immediately decommission the plant after 28 years of successful operation. The plant is a 582 Mwe Westinghouse Pressurized Water Reactor. During the plant's operation it achieved a lifetime capacity factor of 71% and twice set the world PWR record for longest run. Due to the design and operating history, occupational radiation exposure to decommission the plant was estimated to be nearly 2000 person rem. This factor made it appropriate and proactive to perform a full system chemical decontamination of the plant prior to decommissioning.

Occupational radiation exposure comes primarily from the Co-60 tightly held in the corrosion films on plant systems that have contacted the reactor coolant. During a chemical decontamination, dilute acids are injected into the plant systems and dissolve the oxide film thereby fluidizing the trapped radionuclides. The nuclides are carried by the decontamination fluid to ion exchange vessels where they are removed by organic resins. The decontamination chemicals are regenerated by the resins and are returned for additional oxide removal. The handling and disposal of these resins presented a difficult challenge.

Scope of the CY Chemical Decontamination
The systems to be included in the CY chemical decontamination were determined by their contribution to the total decommissioning radiation exposure. The chosen systems were the Reactor Coolant System and auxiliary systems such as the Chemical and Volume Control System and Residual Heat Removal System.
IDENTIFICATION OF THE PROBLEM

During the initial planning for the CY Chemical Decontamination, waste generation information from other plants that had performed chemical decontamination’s was gathered. At one of those plants, the resins that were generated were very near to Class C limits (maximum specific activity allowed for disposal at Barnwell, South Carolina disposal site). This was despite the fact that the decontamination that was performed at this other plant was of a lesser scope when compared to that planned for CY. Upon further investigation it was determined that the transuranic isotopes contained on the resins at that plant were the nuclides that challenged the disposal limits. The level of transuranic isotopes allowed in buried waste is very low due to their very long half lives making them hazardous for an extended period of time. The data in Table I provides the ratios of the Co-60 to transuranics for the operating resins and the resins generated during the chemical decontamination at the Referenced Plant. It can be seen in Table I that the ratios of transuranics to gamma emitters (primarily Co-60) were much higher (1.5 to 10.6 times) then had been observed during the plant’s operation.

<table>
<thead>
<tr>
<th>Isotope Transuranic</th>
<th>Operational Resin Ratio Co-60/Transuranics</th>
<th>Decon Resin Ratio Co-60/Transuranics</th>
<th>Increase in Co-60/Transuranics</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu-238</td>
<td>1754</td>
<td>558</td>
<td>3.1 times</td>
</tr>
<tr>
<td>Pu-241</td>
<td>8.8</td>
<td>5.9</td>
<td>1.5 times</td>
</tr>
<tr>
<td>Am-241</td>
<td>2923</td>
<td>279</td>
<td>10.6 times</td>
</tr>
<tr>
<td>Cm-243/244</td>
<td>1760</td>
<td>245</td>
<td>7.3 times</td>
</tr>
</tbody>
</table>

The pathway of the transuranic activity onto the plant system piping is as follows: Transuranic isotopes are released into the primary coolant when a breach of the fuel cladding occurs (termed failed fuel). As the transuranic isotopes are circulated in the Reactor Coolant System, the radionuclides are incorporated into the oxide film on the inside of the piping. This occurs in the same manner as corrosion products, such as activated cobalt (Co-60), are circulated and incorporated into the corrosion film. These isotopes remain in the film until removed by the chemical decontamination as earlier described. Operating without failed fuel after earlier fuel failures would result in less transuranic activity in the reactor coolant then in the oxide film. This would explain the difference in the transuranic content between the operating and decontamination resins.
This information caused concern for CY due to the following factors:

- CY had experienced failed fuel and has seen higher transuranic ratios in plant surveys.
- CY was considering a decontamination that would contact much more surface area than that affected during the Referenced Plant’s Chemical Decontamination.
- CY was considering using a process that generated considerably less resin per unit surface area than prior processes used in the United States (CY planned to use the Siemens Cord Process). This reduction is accomplished by decomposing the decontamination chemicals using UV light rather than removing them on ion exchange resins. This combined with the above factors made the generation of Greater Than Class C (GTCC) waste a definite possibility.

An estimate of the resin to be generated during the CY Chemical Decontamination was made using:

- The Referenced Plant’s multiplication factor for decontamination resins versus operating resins.
- The surface area to be contacted during the CY Chemical Decontamination.
- The estimated Co-60 activity per surface area which had been determined for a portion of reactor coolant pipe removed in the early 1980s.

The results of this calculation was that CY could expect to generate 2000 to 3000 ft³ (with an associated disposal cost of $4 to 6 Million) of resin with activity close to Class C limits if all of the primary system surfaces (including Steam Generator tubes and Reactor Vessel) were decontaminated. ALARA cost benefit evaluations were performed and determined that deleting the decontamination of the Steam Generators Tubes and Reactor Vessel would greatly reduce the surface area to be decontaminated (and proportionally reduce the volume of waste resin generated). The four Steam Generators and the Reactor Vessel are each to be removed in one piece. Due to this factor the dose savings resulting from chemically decontaminating these components are not great. Deleting these components from the project scope further reduced project costs as less ion exchange vessels and resin handling equipment were required.

Working with the decontamination vendor, flow rate through the Steam Generators were minimized. Jumpers across the inlet and outlet nozzles were used to bypass the Reactor Vessel. These two modifications to the chemical decontamination scope succeeded in minimizing waste costs while still achieving the personnel exposure reduction required.

**THE SOLUTION**

CY realized that a better characterization of our isotopic mixture was needed and that a means of monitoring during the decontamination was required. This was due to estimates that the transuranic content of the resins would determine when they needed to be changed rather than chemical exhaustion.
Characterization

A portion of letdown system piping (called an artifact) was removed and sent with an older S/G tube sample for decontamination testing and isotopic characterization. As can be seen from Table II, the characterization confirmed higher percentages of transuranic activity in the oxide film than were present in CY operating resin (2.8 to 7.2 times higher). This data confirmed that transuranic content would need to be monitored on the decontamination resins to insure that they would not exceed Class C limits. If Class C limits were exceeded, the resin would have to be stored indefinitely. This is due to the current lack of an established method of disposal for this type of waste.

TABLE II

CONNECTICUT YANKEE PLANT
Comparison of the Transuranic Content of CY Operational Resin and RCS Artifact

<table>
<thead>
<tr>
<th>Isotope Transuranic</th>
<th>Operational Resin Ratio Co-60/Transuranics</th>
<th>RCS Artifact Ratio Co-60/Transuranics</th>
<th>Increase in Transuranics Relative to Co-60</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu-238</td>
<td>3953</td>
<td>550</td>
<td>7.2 times</td>
</tr>
<tr>
<td>Pu-241</td>
<td>105</td>
<td>218</td>
<td>3.3 times</td>
</tr>
<tr>
<td>Am-241</td>
<td>2170</td>
<td>502</td>
<td>4.3 times</td>
</tr>
<tr>
<td>Cm-243/244</td>
<td>3063</td>
<td>1093</td>
<td>2.8 times</td>
</tr>
</tbody>
</table>

Development of Monitoring Method

The measuring of specific transuranic isotopic content of a material is somewhat difficult. It generally requires separation techniques followed by alpha spectroscopy. These techniques normally are very lengthy and do not lend themselves to process monitoring. One transuranic isotope, Am-241, emits a low energy gamma (59 keV) in addition to it’s characteristic alpha. Normal gamma spectroscopy using a Coaxial Germanium (Coaxial Ge) detector has low efficiency below 122 keV. It cannot detect Am-241 in the presence of other gamma isotopes normally present in commercial power plant waste such as Co-60 and Cs-137. The relatively new Reverse Electrode Germanium (REGe) Detector has as much as 4 times the efficiency for Am-241 then the normally used Coaxial Ge detector.

Figure 1 below depicts the relative efficiencies of the Coaxial Ge and REGe detectors. Testing at CY showed that Am-241 could be consistently detected in the presence of Co-60 concentrations over 500 times that of Am-241 using the REGe detector.

Note: Although not a problem during the Chemical Decontamination due to the absence of Cs-137 in the oxide film, the REGe is less effective in detecting Am-241 in the presence of low energy gamma emitters (such as Cs-137 normally present in operational wastes). This is due to a higher number of backscatter counts at the Am-241 energy due to these isotopes.
Final Preparations

The Reactor Coolant System artifact results showed that the Co-60 to Am-241 ratio in the Connecticut Yankee oxide film was consistently in the range of 500:1. As the Co-60 ratios to other transuranic isotopes were also consistent, the final resin volume from the chemical decontamination was estimated at 500 to 800 ft³. This shows the effect of being able to confidently monitor the transuranic content of the resin and the reduction in the surface area contacted by the decon chemicals. The corresponding reduction in waste disposal cost was estimated at $3 to 4.4 Million (a 75% savings).

To facilitate the tracking of Am-241 during the chemical decontamination, a spreadsheet and plotting program was set up. This program first determined the difference between the resin bed influent and effluent sample concentrations of Am-241. This result was next multiplied by the bed flow rate and time period between samples. The product of those values is the quantity of activity removed during a time step. The cumulative quantity of
activity removed was automatically graphed versus time. The graphs included predetermined alert levels and limits as to transuranic activity on a resin bed.

Figure 2 below shows an actual tracking plot from the CY Chemical Decontamination Resin:

**Figure 2**

*Graph of Am-241 Loading versus time for Demineralizer “D”*

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**RESULTS**

As is exemplified on Figure 2, the monitoring program allowed the Chemical Decontamination Resin beds to be loaded to a relatively high percentage of the Class C limits. This assured regulatory compliance while minimizing waste volumes and cost.

**Table III**

<table>
<thead>
<tr>
<th>CY Chem Decon Performance Factors Expected Vrs Results</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Performance Factor</strong></td>
</tr>
<tr>
<td>Avg Dose Rate Pre/Post (DF)</td>
</tr>
<tr>
<td>Area Dose Reduction</td>
</tr>
<tr>
<td>Co-60 Removed</td>
</tr>
<tr>
<td>Total Activity Removed</td>
</tr>
<tr>
<td>Total Resin Generated</td>
</tr>
<tr>
<td>“Process” Resin Generated</td>
</tr>
<tr>
<td>Resin Activity as % of Class C Limits</td>
</tr>
</tbody>
</table>
Table III above shows the overall performance results of the CY Full System Chemical Decontamination. It should be noted that the dose reduction goals were achieved despite less Co-60 being present on the decontaminated surfaces than anticipated. As predicted, the relatively high level of transuranics in the oxide film resulted in the generation of 465 ft³ of resin whereas the chemical “process” required only 115 ft³ of resin to be effective. This substantiates CY’s focus on reducing resin volumes. A complete isotopic analysis of the Decon resins determined that the actual % of Class C limits was within 8 % of that predicted by the monitoring program. This is very good agreement considering the numerous measurement errors that could have existed in the monitoring program.

The reduction in plant area dose rates was the primary goal of the CY Chemical Decontamination. To monitor of the effectiveness of the process, CY developed a monitoring and tracking process similar to the resin tracking program. Thirty-nine (39) remote reading radiation detectors were mounted at representative locations on the systems being decontaminated. The data was downloaded at least once a shift and feed to a plotting program for effectiveness tracking. Figure 3 illustrates the dramatic dose reduction achieved through the Chemical Decontamination for some of the monitoring points.

CONCLUSIONS

In conclusion, the following was learned in the process of preparing for and implementing a Full System Chemical Decontamination and the CY Haddam Neck Plant:

- If a nuclear power plant has had a history of failed fuel, the transuranic content of the oxide film on the inside of plant piping can be a major concern during a chemical decontamination.
• Should the situation described in the first conclusion be true, careful monitoring will be required to assure that Greater Than Class C resin is not generated during the performance of the chemical decontamination.

• With careful monitoring, the waste minimization qualities of current chemical decontamination technology can be used to minimize waste disposal costs.