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INLINE MONITORS FOR THE SRS SMALL COLUMN ION EXCHANGE PROCESS

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ABSTRACT

A Small Column Ion Exchange (SCIX) system, designed by the Oak Ridge and Savannah River National Laboratories (ORNL and SRNL), is a potential way to reduce Cs-137 concentrations in high-level radioactive waste at the Savannah River Site (SRS). SRNL has developed gamma-ray monitors for six locations within the SCIX system to verify the proper operation of the ion exchange system, detect cesium-137 and barium-137m breakthrough, and confirm the presence of cesium before and after used resin is transferred to a grinder module. Two sodium iodide breakthrough monitors, one Geiger-Mueller (GM) breakthrough monitor, and three Geiger-Mueller transfer monitors were used. The present work provides a means of measuring the Cs-137 and Ba-137m breakthrough by taking multiple measurements in a process flow diversion and isolation loop. A lead shield was used for the NaI detectors, and the aperture of the collimator tube in this shield was designed using Monte Carlo analyses to provide the desired count rate for the gamma rays of interest. A computer program was written to collect data from the process monitors, provide alarm notification, and plot the data for ease of operation.

INTRODUCTION

In order to reduce Cs-137 concentrations in high-level radioactive waste at the Savannah River Site, a Small Column Ion Exchange (SCIX) system has been designed by the Oak Ridge and Savannah River National Laboratories (ORNL and SRNL). SRNL was asked to develop gamma-ray monitors at six locations within the SCIX system. Gamma-ray monitors are required to verify the proper operation of the ion exchange system, detect cesium breakthrough, and confirm presence of cesium before and after used resin is transferred to a grinder module.

The only observable gamma ray in the decay of Cs-137 is from its short-lived Ba-137m daughter. Chemical processes, such as the SCIX, may disrupt the secular equilibrium between this parent-daughter pair; meaning that measurement of Ba-137m will not necessarily yield information about Cs-137 content. While this is a complicating factor that can not be ignored, it is controllable by either: allowing sufficient time for equilibrium to be reestablished (about 20 minutes), or by making multiple measurements with sufficient statistical precision to determine the extent of disequilibrium. The present work provides a means of measuring the Cs-137 and Ba-137m by taking multiple measurements in a process isolation loop that contains the process solution of interest.

SYSTEM DESCRIPTION

General Description
Gamma-ray monitors have been provided at six SCIX locations for the purposes of measuring Cs-137 breakthrough and for monitoring normal transfers of radioactive materials as shown in Figure 1. Two sodium iodide breakthrough monitors, one Geiger-Mueller (GM) breakthrough monitor, and three Geiger-Mueller transfer monitors are shown. GM_2 is a breakthrough monitor for the ion exchange column, while GM_1 monitors the solution radioactivity before the column, and GM_3 and GM_4 monitor the spent-column slurry when it is being transferred through the grinder.

Detectors/Electronics

Two thallium-doped sodium iodide, NaI (Tl), gamma-ray spectrometers provide redundant, quantitative Cs-137 concentration data. Each uses a flow diversion and isolation loop. Once the sample is isolated, data acquisition protocols provide a rapid succession of short duration counts, and these are analyzed to determine the current Cs-137 / Ba-137m equilibrium. In the event that one system fails, the other is able to provide equivalent data. While reaching secular equilibrium of the Cs-137 / Ba-137m is not a necessary condition, final analytical results will be determined more precisely if measurements are made at that time.

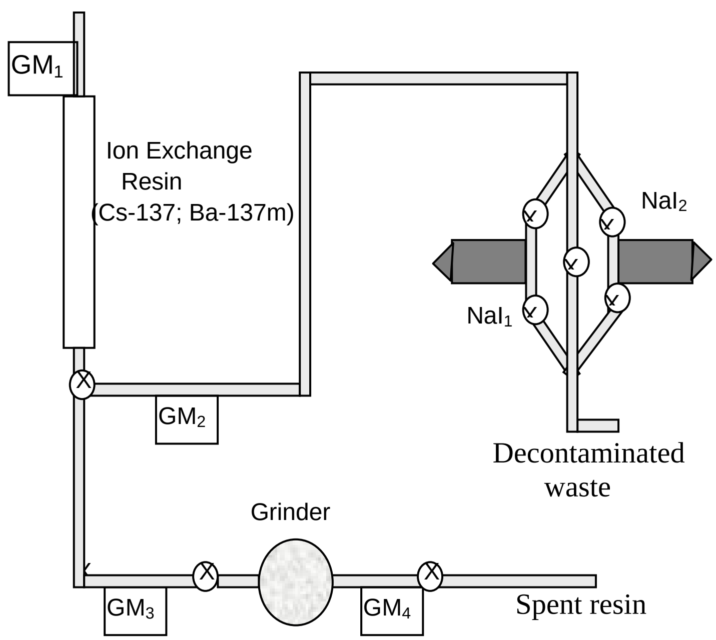

Figure 1. Simplified process monitoring schematic.

Either of two NaI detectors is used to track the buildup or decay of Cs-137. While one is analyzing a sample the other is off line. Samples of the ion exchange column eluant are taken into an isolated pipe and counted repeatedly. Cs-137 beta decays to Ba-137m which decays to the Ba-137 ground state with the emission of a 661.7 KeV gamma ray. The rate of buildup or decay of the 661.7 keV gamma ray from the Ba-137m is used to determine the condition of the ion exchange medium.

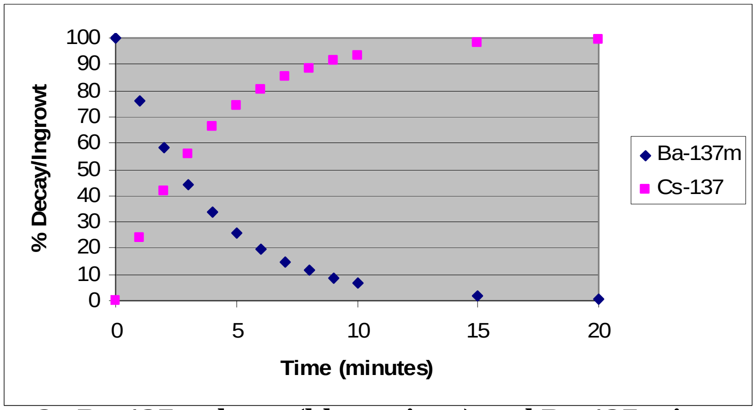

Figure 2. Ba-137m decay (blue points) and Ba-137m ingrowth for Cs-137 (pink points).

As shown in Figure 2, if there is only Ba-137m breakthrough, it will decay in a manner represented by the blue points in this graph. However, if there is only Cs-137 breakthrough, the Ba-137m ingrowth is shown by the pink points. If both Ba-137m and Cs-137 breakthrough occurs, the graph will be a summation of the two curves.

As stated above, four Geiger-Mueller (GM) detectors are also included. GM_1 is placed adjacent to the input line into the ion exchange column. Its purpose is to indicate that material is flowing into the column. When material containing Cs-137 is flowing into the column or isolated above the column, it will read high. The reading will decrease if the material in the piping before the column does not contain as much Cs-137. GM_2 is placed near the output of the column, before the sampling line to the NaI detector. It will read high if an unexpected catastrophic failure in the column sends untreated Cs-137 bearing liquids out of the column. The GM_3 detector is placed adjacent to the inlet to the grinder. It will only read high if the contents of the column are being dumped to the grinder. The GM_4 detector is placed adjacent to the outlet of the grinder. It will only read high when material is flowing from the grinder into the storage tank.

A diagram of the process monitoring electronics is shown in Figure 3. The GM detectors are connected to Aware Electronics PMI-30 modules that convert the non-standard pulses into standard pulses. These PMI-30 modules have been mounted in nuclear instrument modules (NIMs) for convenience and to provide power to the PMI-30. Two PMI-30s are in each of two NIMs. The outputs of the GM detectors go to the PMI-30s in the NIM. Outputs from the PMI-30s go from the NIMs to a National Instruments BNC-2121 that converts the pulses to transistor-transistor logic (TTL) pulses. The BNC-2121 is interfaced to a National Instruments 6602 card mounted in a slot in the desktop PC. The control software obtains count rates from the GM detectors via the BNC-2121 and the

6602. The signal processing electronics are primarily NIM standard modules, mounted in a NIM bin.

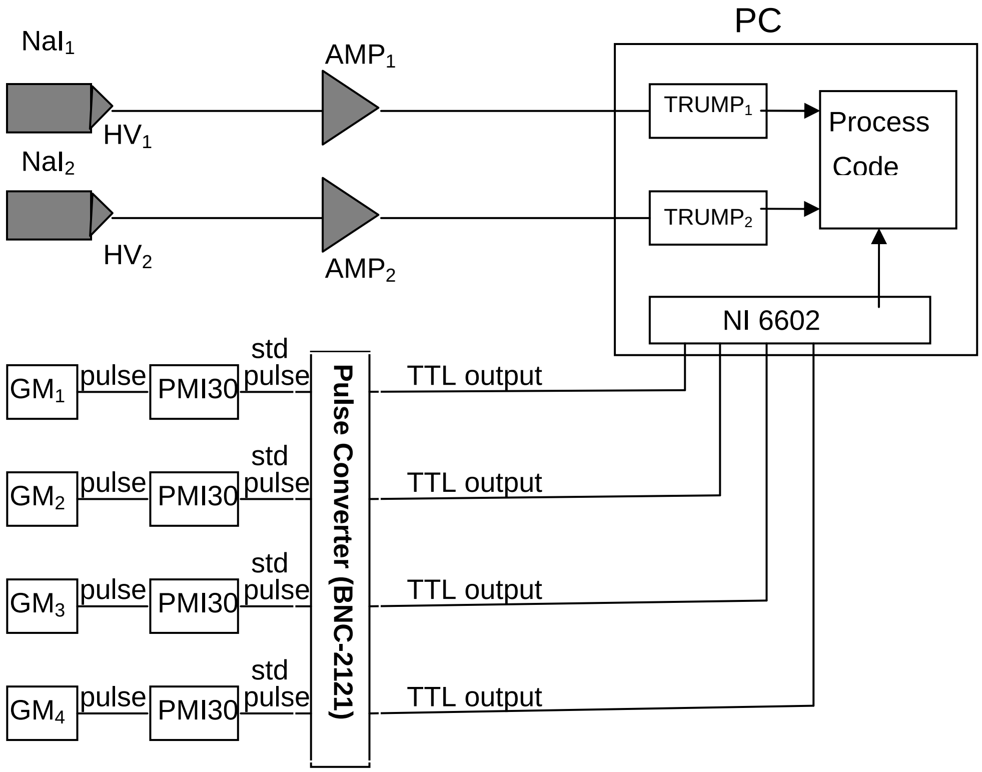

Figure 3. Process monitoring electronics diagram.

For the production system, the following modules are acceptable (equivalent modules can also be used): 1 NIM Bin/power supply (Ortec 4001A/4002A); 2 Amplifiers (Canberra 2015A); 2 High Voltage Power Supplies (Ortec 478); 2 Multichannel Analyzer cards (Ortec TRUMP-PCI); 2 NaI detectors (Scionix 25B25/2-E2-X hermetically sealed); 4 GM detectors (Ludlum 133-6,waterproof with 100 ft cable), 4 PMI-30, mounted in NIM module (Aware Electronics PMI-30); 1 National Instruments BNC-2121; 1 National Instruments 6602; and a Desktop PC

After amplification and pulse shaping, all the detector signals are supplied to boards installed in a desktop PC, where the software analyzes the signals and displays the resulting data on menu driven screens. The two Ortec TRUMP cards receive the input data from the two Scionix NaI detectors. The TRUMP cards are full featured multichannel analyzer (MCA) cards that permit the PC to function as a MCA. In the initial setup of the system, each TRUMP card is run as a standalone MCA. A region of interest (ROI) is established around the Cs-137 (662 keV) peak so that the total number of counts in this peak may be obtained by the operating software whenever required. In this way the total number of counts in the Cs-137 peak is acquired by the software for analysis.

Detector Shielding

The shielding design for the NaI detectors is shown in Figure 4. It is 8.625 inches in outside diameter including the steel outer case. Monte Carlo analyses were done to evaluate the response of the NaI detector to 0.1 Ci/gallon Cs-137 in the sampling pipe, and in a process pipe running behind the detector/shield assembly.

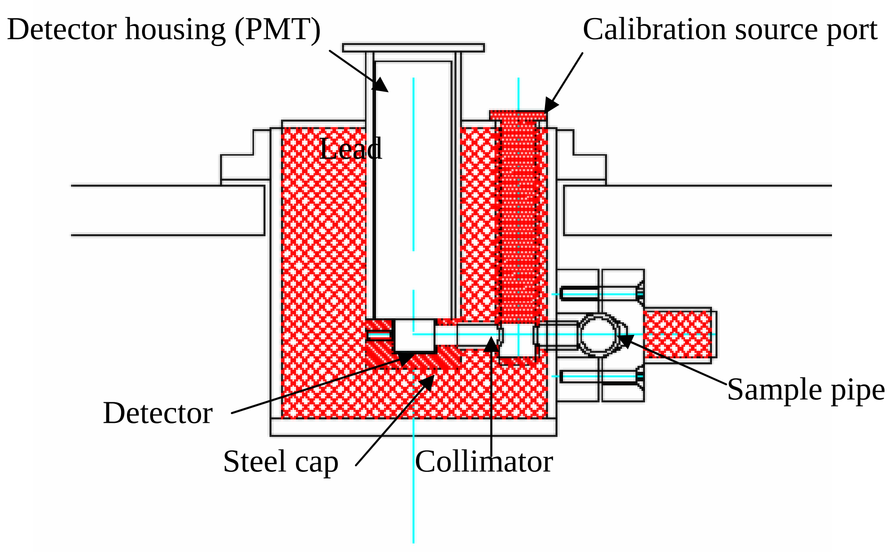

Figure 4. Shield design.

Figure 5 is a drawing showing gamma ray tracks from the process pipe to the NaI detector. This is the result of following 100,000,000 gamma rays emitted from the entire length of the process pipe. It is apparent that only that portion of the process pipe adjacent to the shield contributes to the background gamma rays at the NaI detector. This analysis also showed that the contributions from the adjacent piping were negligible.

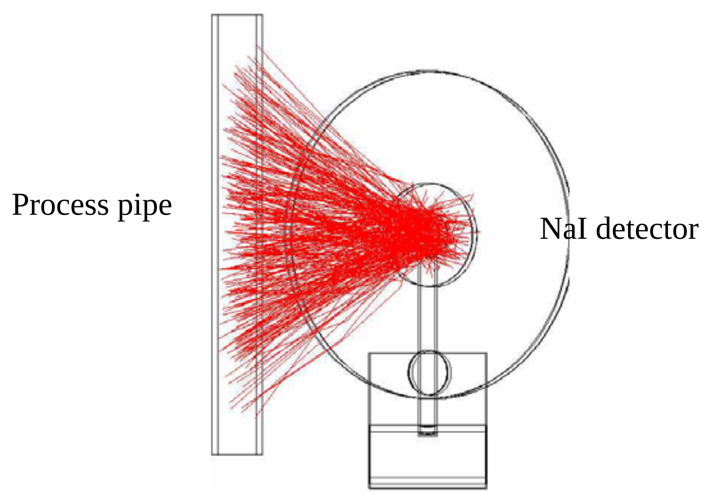

Figure 5. Line drawing showing gamma ray tracks from the process pipe to the NaI detector.

The NaI detector is used to measure the buildup or decay of the gamma rays from the Ba-137m daughter of Cs-137. The TRUMP card in the PC is used as a multichannel analyzer (MCA) and a region of interest (ROI) is set around the 661.7 gamma-ray peak from the Cs daughter, Ba-137m, and the counts in this peak are summed.
The collimator tube from the sample pipe to the NaI detector is crucial in the response of the instrument. Too large an aperture in the collimator will give a high count rate and excessive dead time. MCNP analyses were performed to size the collimator hole. Either a collimator inner diameter of 0.375 or 0.250 inches will provide adequate collimation. The 0.250 inch collimator will provide a lower count rate with reduced dead time. Therefore, the 0.25 inch collimator is recommended since it will provide a greater margin of error if the Cs-137 in the stream increases above the design of 0.1 Ci/gallon.

Detector Calibration

NaI detectors were calibrated using a standard purchased from Analytics Inc., Atlanta, GA (Standard 68709-147). It was counted for 5 minutes with the sodium iodide detector using lead shielding with a 0.25" lead collimator. A detector efficiency of 2.6% was obtained, and the analyzer dead time was 20% for the 0.5 mCi standard that was representative of the design concentration of 0.1 Ci/gallon.

System Software

A computer program was written in Visual Basic 6.0 to collect data from the process monitors and plot the data for ease of operation. Batch sample counting and waste transfer activities are coordinated between the Distributed Control System (DCS) and the SCIX monitoring software by event signaling. In the batch counting mode, the DCS notifies the SCIX monitor when the sample loop for one of the NaI detectors has been filled. The SCIX monitor performs its counting operation using the appropriate detector and notifies the DCS when the operation has been completed. The SCIX monitor provides alarm notification to the DCS upon detection of such an event. In addition, Routine Ion Exchange and Spent Resin transfer monitor operations are controlled via similar signals from the DCS. The SCIX monitor also provides alarm notification to the DCS upon detection of an alarm event during either transfer operation.

In the first (breakthrough) operating mode, the software monitors one Sodium Iodide (NaI) monitor and one Geiger-Mueller (GM) transfer monitor and one GM breakthrough detectors. The other NaI detector serves as a backup. The four detectors provide a means to verify proper operation of the ion exchange system and to detect Ba-137m/Cs-137 breakthrough from the resin bed. The two GM detectors are operated in a free running mode to serve as a real time monitor for the ion exchange operations.

The second (transfer) operating mode is the Spent Resin Operation. In this mode, two GM detectors are used as transfer monitors to observe the spent resin feed line to the grinder and the ground resin return line to the high-level waste tank. Both detectors operate in free running mode performing continuous 60 second counts for the duration of the transfer.

At startup the Small Column Ion Exchange main screen is displayed. See Figure 6. The system time is displayed in the upper right hand corner of the title bar and updated every second to indicate that the system is active.

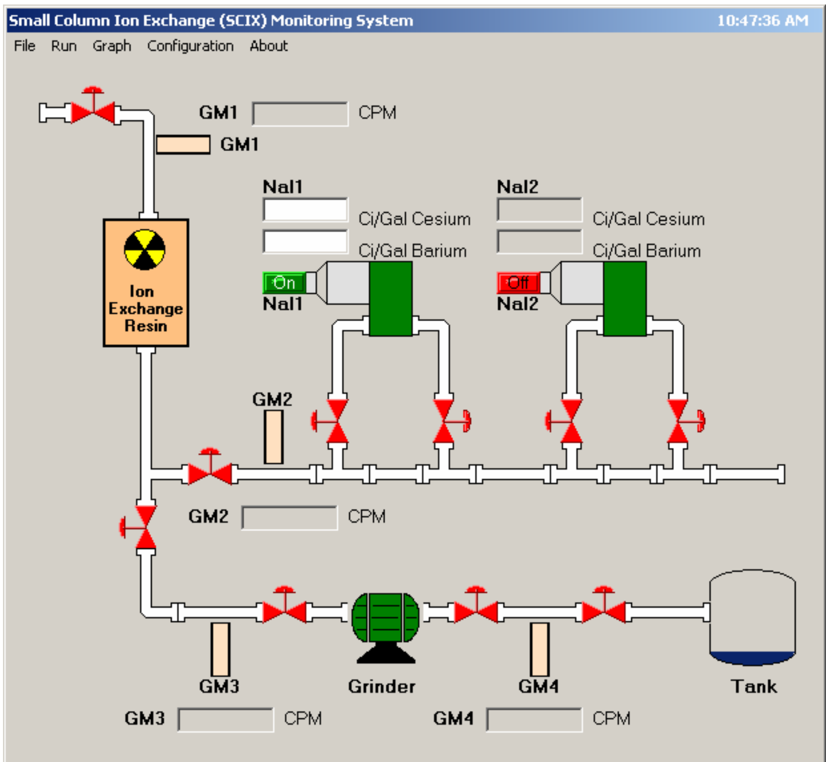

Figure 6 - SCIX Main Screen

From this screen, the user can start either the Routine Ion Exchange monitor or the Spent Resin monitor, change one of the configuration settings, or exit the system. System graphs are not visible until one of the two monitoring operations has been started. NaI detector selection is also performed from this screen by clicking on the appropriate red or green selector button on NaI1 or NaI2. Detector selection is disabled once data collection has started.

A SCIX configuration utility allows the user to change system operating parameters for all six detectors (2-NaI and 4-GM). Once changed, the new values are stored in a configuration file for future use. In the NaI setup the user can select the time interval for each count and the number of counts to be performed. The user can also enter values for detector efficiency and background. These values are used in the calculation of the Ba-

137m/Cs-137 concentration (mCi/gal). The user is also able to change the system alarm setting for each detector from this screen.

Results

The SCIX Run Menu allows the user to start either of the two system monitors, the Routine Ion Exchange Monitor or the Spent Resin Operations Monitor. If the Start Routine IX option is selected two graphs will immediately be displayed. The first is the Routine Ion Exchange Data graph which contains plots for GM1 and GM2 data. See Figure 7. By default, the GM data is read from the NI-6602 card every 60 seconds. This value is not accessible to the user. A next-update counter in the lower left hand corner of the screen is updated once a second to show the time remaining until the next GM data pair is available. Figure 7 also shows the second graph displayed when the Start Routine IX option is selected. The PMT data graph is simply a graph of the resin bed Ba-137m/Cs-137 breakthrough concentration (Ci/gal) measured by the selected NaI detector.

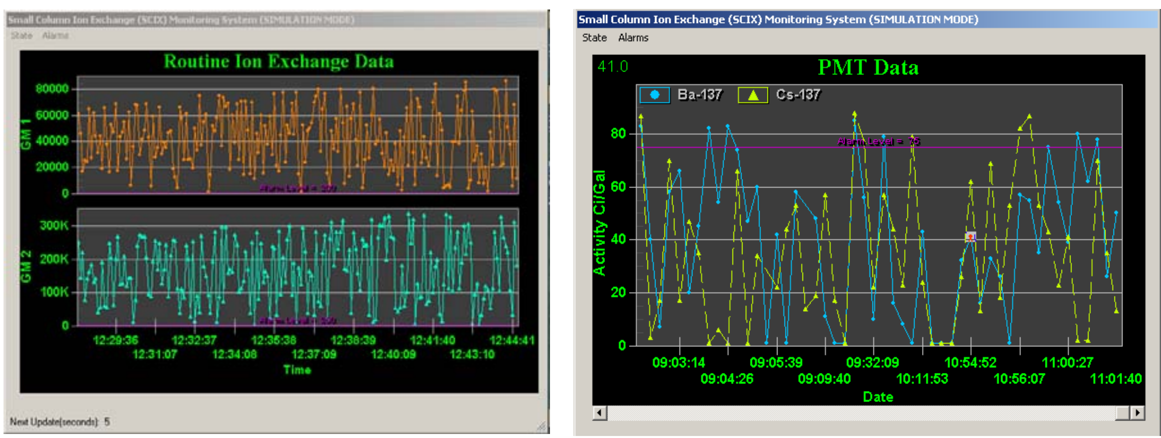

Figure 7 - Routine Ion Exchange Data - GM Graph and PMT Data - NaI Graph

Data for the NaI plot is obtained from a Ba-137m/Cs-137 ingrowth / decay analysis performed by the program. The new data points are also compared to the existing alarm setting and the appropriate sequence is initiated.

Choosing the Start Spent Resin Operation option from the run menu will cause a single graph to be displayed. The graph contains plots for detectors GM3 and GM4, similar to GM1 and GM2 graphs given in Figure 7, for transfer lines to and from the spent resin grinder. Data are also compared to alarm settings and an appropriate response initiated.

CONCLUSIONS

SRNL has developed gamma-ray monitors at six locations within the SCIX system. These monitors verify the proper operation of the ion exchange system, detect cesium breakthrough, and confirm the presence of cesium-137 and barium-137m before and after used resin is transferred to a grinder module. Two sodium iodide breakthrough monitors, one Geiger-Mueller (GM) breakthrough monitor, and three Geiger-Mueller transfer monitors were used. Monte Carlo analyses were made to evaluate the response of the NaI detector to Cs-137 in the sampling pipe, and calibration data confirmed that the collimator design was appropriate. A computer program was written in Visual Basic to collect data from the process monitors and plot the data for ease of operation. This software provides alarm notification for alarm events during SCIX operation.

REFERENCES

1. "Task Technical Plan for Small Column Ion Exchange Gamma-Ray Monitors," WSRC-RP-2003-00158, February 3, 2004.
2. "Small Column Ion Exchange Monitor System Final Report," WSRC-RP-2004-00615, September 30, 2004.