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High Level Waste Tank Gamma Profiling

Frank S. Moore*, D.M.C. Odell*, Larry.J. Harpring*, Phillip J. French*, John R. Sessions** *Savannah River National Laboratory ***Westinghouse Savannah River Co, PO Box 616 Aiken SC 29802*

Abstract–*Cleanup at the Savannah River Site brings with it the need to clean out and close down the radioactive waste tanks constructed in support of the fuel rod dissolution process. An innovative technique for assaying waste tanks has been developed at the Savannah River Site. The technique uses a gamma detector in the annular space between the inner and outer walls of double walled tanks. Unique shielding, counting electronics, and deployment techniques were developed to facilitate mapping interstitial liquid levels, sludge layers and other structures in the waste tank located near the tank walls. The techniques used, results, and lessons learned will be discussed.*

I. INTRODUCTION: As part of the cleanup of the Savannah River Site, the radioactive waste tanks constructed to support the fuel rod dissolution process need to be emptied, cleaned and closed. Some of these tanks have lain dormant for many years, while others have had extensive waste transfers both into and out of the tanks. As a result, while fill histories exist, detailed knowledge of the contents of the tanks is incomplete. Since many of the tanks are double walled, the annular space between the two walls offers a location to introduce gamma-ray probes to assay the distribution of the radioactivity within the tanks. In some of these tanks, the liquid level is below the level of the salt cake in the tank. In these tanks one has a "dry" salt cake above the liquid level and saturated salt cake below this level.

The initial task was to locate the interstitial liquid level in one tank. The fundamental assumptions were fourfold. First, Cs-137 would be the principal gamma emitter, and that because of its solubility, almost all of the Cs would be in the liquid. Second, as seen in terrestrial hydrology, there would be three distinct zones in the tank, the saturated zone, a capillary fringe, and dry salt cake. Third, the dry portion of the salt cake would be very well drained, and fourth, the salt cake would be homogeneous. Information about the capillary zone in particular would assist in the ongoing effort to model the drainage characteristics of the salt cake, hence the required information was the location of the liquid level, and the thickness of the capillary zone. The only knowns were the height of the salt cake in the tank, and the depth of the liquid in a pump well which had been bored through the salt some distance from the tank wall. The "expected" line in figure 3 represents what was anticipated.

II. DETECTOR DEVELOPMENT: Detectors, shields, and deployment mechanisms were interdependent and depended on the tank type and the Curie content of the interstitial liquid within the salt cake. Curie per liter values ranged from about 0.5 to more than 6. Two tank types were mapped, the Type I which has 12.7 cm diameter riser ports in the annular space, located from 30cm to 45cm from the inner wall, and the Type III which has 20cm riser ports approximately 38cm from the inner tank wall. To accommodate both types, two different detector/shield assemblies and deployment mechanisms were developed. The first was a single, magnetic wheeled crawler called the "skate" and the second was a rail-guided cylindrical shield referred to as the "boat anchor". In both cases, it was decided that total count rate would be measured, and hence Gieger-Muller (GM) detectors manufactured by Ludlum Measurements were employed. The series 133 detectors were chosen because of their small size (2.5 cm diameter by 10cn long) and availability in ranges up to 1000R/hr.

II.A. Skate: For the type III tanks, the riser ports were only approximately 38 laterally from the inner tank wall. This meant that a detector/shield assembly could be lowered into the annular space between the two tank walls and swung over to reach the inner wall. A magnetic wheel on the detector housing (skate) attached the system to the inner wall and permitted raising and lowering the skate while attached to the tank wall. A 5-sided box of machinable Tungsten 1.27 cm thick surrounded the GM tube to ensure that it was 90 percent forward looking into the tank (figure 1). The location of the shield below the wheel and inside the wheel's axis ensures that the housing remains vertical to the tank wall, and the single wheel ensures that it tracks vertically. Two cables are used to pull the detector assembly up the side of the tank. The light weight of the assembly (approximately 3.6 kg) allows it be raised and lowered manually.

In use, the skate is lowered to the bottom of the annular space (a distance of approximately 36 feet from the top of the riser) and swung back and forth until it attaches itself to the side of the tank. It is then raised in known increments for data collection.

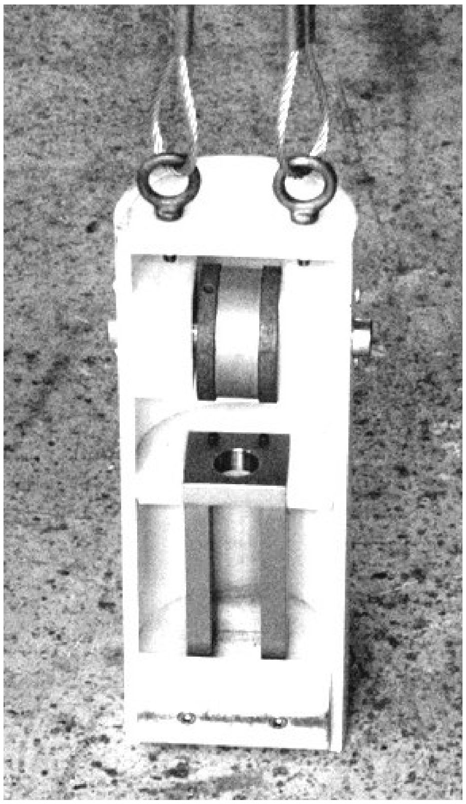

Figure 1, “Skate” Detector Holder

II.B “Boat Anchor”*:*The type I tanks have 12.7 cm diameter risers, located across a range of distances from the tank walls in the annular space. The smaller riser diameter, the variable distance from the inner wall, and the certainty of having to eventually work in a contaminated annular space precluded using the skate. Also, the type I tanks of immediate interest tended to have higher Curie contents than the type III’s. For this reason a new shield design and deployment method were devised. The “boat anchor” shield, named for its weight (approximately 19 kg), was made from 10 cm diameter machinable Tungsten. The hole for the GM detector was offset 2.5 cm to provide more shielding on the tank side, and a 0.5 cm collimator aperture was provided so that the GM tube could see the tank wall (Figure 2). To deploy it, a thin walled aluminum pipe was inserted into the riser port in six foot sections. This pipe has internal ribs which engage a pair of slots cut into the shield to keep it oriented toward the tank wall. A crank mechanism was employed to hoist it up inside this guide pipe while data was acquired at 15 cm intervals.

Figure 2, “Boat Anchor” Shield, Showing 0.200 inch Collimator Aperture

II.C. Counting System: Time constraints in the initial liquid level measurement required the employment of existing counting equipment. An analog Ludlum count rate meter was used for the first data acquisition. This functioned satisfactorily, but there were several difficulties. First, the meter had to be read and the data transcribed. This took time and there were obvious opportunities for transcription errors. Second, scale changes on the meter often meant that one data point was taken with the needle near the high end of the meter, where it could be easily read, while the next point would be acquired with the needle down at the low end of a higher scale where accurate readings were difficult. Further, since meter accuracy is specified as percent of full scale, some of the readings at the low end were subject to significant potential for errors. For future work an improved system was designed.

The digital instrument used subsequently is a Savannah River proprietary design, based on data sampling technology.(1) This permits stable operation over a very wide range of count rates while being small in size and low power. It is totally self contained, including the high voltage supply and can be used with GM tubes for simple counting applications or with spectroscopy detectors to produce energy spectra. In its digital count rate meter mode, it counts for 250 millisecond intervals and feeds its gross counts into a 20-tap digital filter to produce a result every ¼ of a second that represents the linear average of the last five seconds. While simple, this filter has functioned well for measurements spanning a 100:1 count rate range in the field. The instrument is interfaced to a handheld, tablet PC running Windows CE and a Visual Basic touchscreen human interface program which records date, time, tank number, detector elevation in tank inches and the count rate in thousands of counts/minute (Kcpm). The output file produced is in a comma separated variable format commonly used for importing data into Excel spreadsheets. After identifying the tank, riser number and increment between data points, one simply taps the PC’s stylus at one button to acquire the data, and at another button to increment the detector elevation display. In this way, data can be acquired as rapidly as the detector can be relocated to the next data point. Using the skate, four or five riser ports can be mapped in less than half a day. The boat anchor system is slower to deploy because the guide rail pipe system must be disassembled and reassembled in order to be relocated from one riser port to another.

The detection systems were calibrated in the B Area Calibration Facility at the Savannah River Site. This gave a calibration in terms of R/hr versus Kcpm. A tank was mocked up in Microshield 5(2) and MCNP(3) to provide a conversion from R/hr to Curies/liter. From this a conversion factor from Kcpm to Ci/liter was obtained. The accuracy of this conversion factor depends on having good knowledge of the material in the tank, and its density. The available information is imprecise because of the number of years of accumulation in the tanks, and thus the accuracy of the Ci/liter value is limited.

III. DEPLOYMENT: The initial application was the determination of the liquid level in Tank 41.Tank 41 is a type III tank with no history of contamination in the annular space, so the skate system was employed. Expectations were that one would see (starting at the top of the tank) a relatively low but constant count rate as the detector came down the tank wall. The count rate would rise in the capillary zone, reaching a higher value in the saturated salt zone, and remain at that value all the way to the bottom of the tank. Five ports were scanned in November, 2002. Figure 3 shows the average of all 5 ports plotted against the distance from the bottom of the tank as well as a graphical representation of the anticipated results.

Several observations can be made from this figure. First, there is no obvious saturated salt zone as had been anticipated. The liquid level in a well drilled to the bottom of the tank was at approximately 150 inches at the time the data was collected, so below this elevation there should be saturated salt, while above it, the salt should be relatively dry. Second, there are indications of non-homogeneity. There is structure in the dry salt zone at approximately 260 inches, and at 200 inches, as well as a sharp spike in the data at 135 inches and a marked rise in activity below approximately 80 inches. Figure 4 shows all five ports individually separated for clarity. In all multiple plots the vertical scale has no significance. Note that the same structures appear in all ports at the same elevations, thus it is unlikely that they are artifacts of data acquisition. The structures are also not symmetric, the spike at 135 being more prominent in ports 4 and 6. The structure at 200 and 260 inches is thinner at some ports than at others. Experience in drilling the well had shown hard layers at 200 and 260 inches. The gross gamma data indicates that these hard layers have higher activity levels which were confirmed by spectroscopy to be Cs-137.

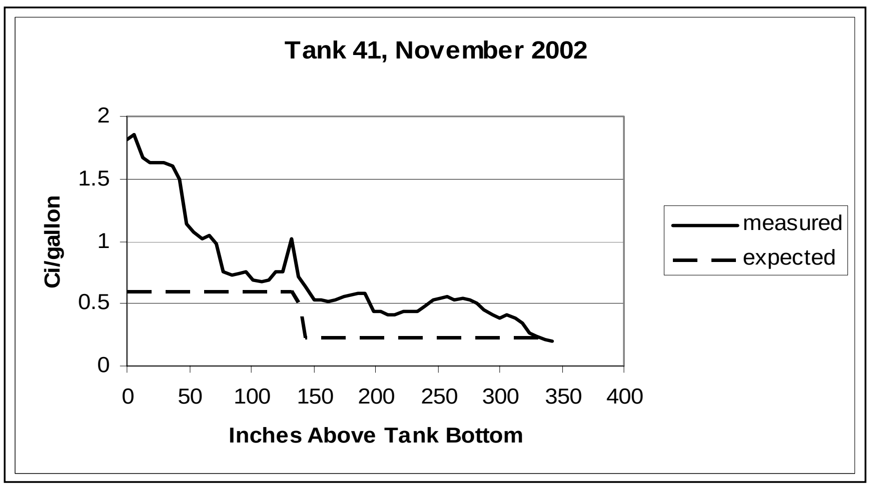

Figure 3, Tank 41, November 2002, Data

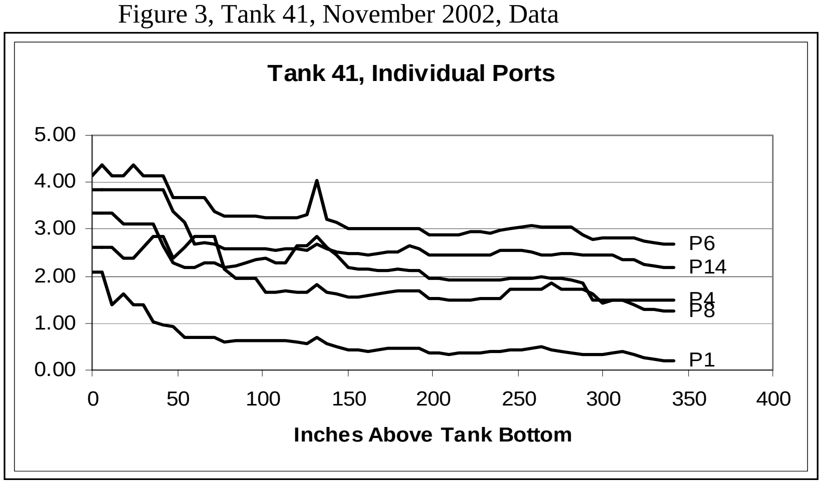

Figure 4, Tank 41, November Individual Port Data

A further scan was performed in March 2003, when the liquid level in the central well was at 170 inches and another in June when the liquid had been drained down to approximately 60 inches. Figure 5 shows average values for these scans plus November, again separated for clarity. Comparing the March, 2003, and November, 2002, scans, it can be seen that the same structure is visible in both sets of data. There is a sharp break in the March data at 170 inches and (except for the spike at 135) the data is flat down to approximately 70 inches. This is the expected behavior of a saturated salt cake zone. The downward transition above 170 is the capillary zone. In retrospect it can be seen that the liquid level transition in the November data is concealed by the spike at 135. The saturated zone was evident in the November data, but insufficient understanding at the time prevented accurate discernment.

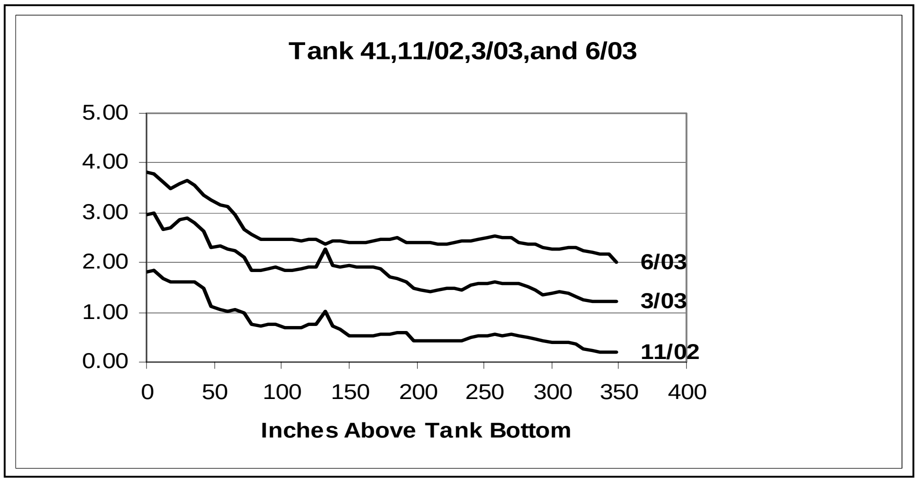

Figure 5, November 2002, March and June 2003, Scans

The June data was taken after the liquid had been drained to less than 36 inches and the level had been allowed to recover to 60 inches over a period of weeks. Except for the structure at approximately 200 and 260 inches, the shape is that expected for drained salt cake. The spike at 135 has disappeared, replaced by a small valley. This has since been interpreted as evidence for a sizable void in the salt cake, previously filled with liquid (though the actual size and distance from the tank wall are not known). The free liquid in the tank was known to be approximately 2 Ci/gallon. This liquid was drained with the remainder of the liquid above 60 inches, leaving the void. It shows up as a spike whenever the void is below the liquid level since wherever the void approaches the tank wall, there is less salt cake to attenuate the Cs-137 gamma rays from the liquid filled void, increasing the count rate, which is interpreted as an increase in measured Ci/gallon.

Figure 6 shows the results of the June scan plus those in July, August, and September, 2003. In July the tank was refilled with water up to the top of the salt cake at approximately 350 inches, and the liquid level has remained constant since that time. The structures at 200 and 260 inches do not appear to be dissolving or leaching Cs-137. The downward spike at 135 inches has disappeared by August, and by September, it has been replaced by a small upward spike. This supports the hypothesis that the spike demonstrates the existence of a void in the salt cake, and as the entrained Cs-137 is washed out of the salt into the liquid, the activity in this refilled volume is increasing. No firm hypothesis has been formed to explain the increase in the activity level near the bottom of Tank 41. Such an increase is not characteristic of all tanks mapped to date.

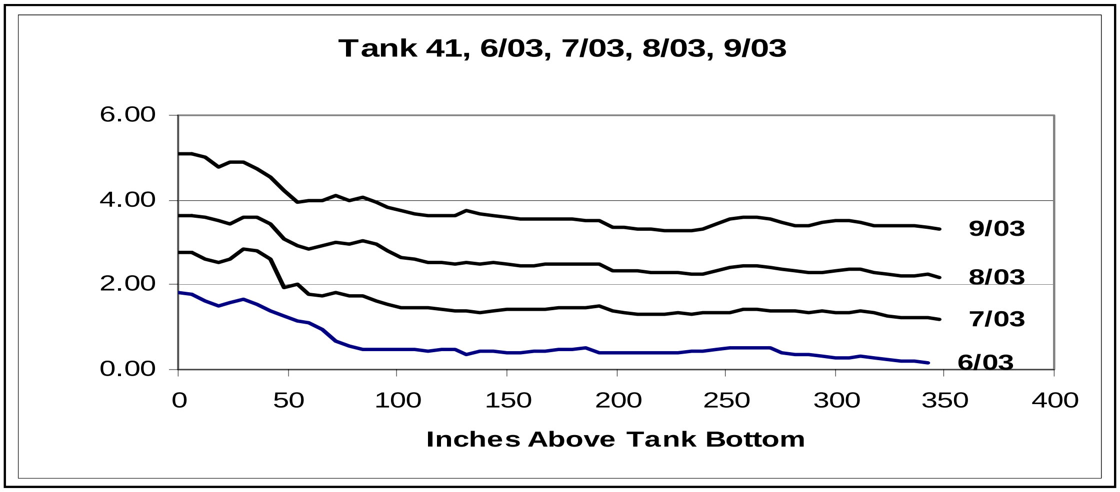

Figure 6, Tank 41, Scans in June, July, August and September 200

.Figure 7 shows a scan of Tank 3, a type I tank. Type I tanks have only 4 risers (as opposed to 14 on Type III's) and they are smaller in diameter and can be farther from the inner wall. Hence in this case the boat anchor shield and aluminum guide rail deployment system were used. Also, as can be seen from the graph, Tank 3 has a higher Curie content than Tank 41. The top of the salt cake in Tank 3 is known to be at approximately 200 inches, which can be seen in the data by observing the major slope changes at approximately 180 and 210 inches. The residual gamma radiation between 200 and 300 inches is partially due to the contribution from the entire top of the salt cake and partially due to salt deposits on exposed cooling coils above the cake. Below 180 inches Tank 3 shows the characteristics of saturated salt cake down to the bottom 100 inches where the activity levels rise above the saturated salt level. This is similar to the Tank 41 lower region and requires further investigation.

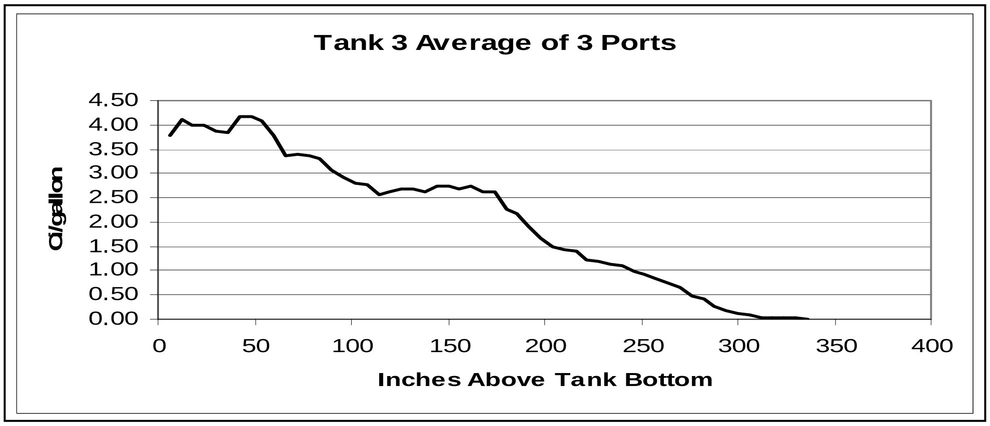

Figure 7, Scan of Tank 3

IV. LESSONS LEARNED: Several important lessons have been learned from the series of scans of Tank 41. First, the tank's contents cannot be assumed to be homogeneous in any axis. There may be layers of insoluble materials in the soluble salt cake which affect both the vertical and radial homogeneity, as indicated by the five individual November port scans which indicate that voids and Cesium concentrating regions may be off center and not uniform in thickness. Second, the interface between saturated and unsaturated salt cake can be determined, so that the top of the interstitial liquid can be located. In practice, this required several scans of Tank 41 and other tanks before a reliable interpretation of the data became possible.

Gamma-ray scanning of tank contents from the annular region has one feature which is both a limitation and an asset. A tank is an infinitely thick gamma source to the detector, but one only sees the outer eighteen to twenty-four inches of the salt cake or liquid within the tank. This means that conclusions cannot be drawn about the tank contents toward the center of the tank. With an overall diameter of 80 to 85 feet, the limitations of sampling only the outer 1 to 2 feet become clear when the degree of inhomogeneity that has been observed is taken into account. It also means that one can be confident that whatever is seen is within two feet of the detector, and one does not have to compensate for the other side of the tank. This has proven especially valuable in simplifying the modeling effort.

Finally, tank scanning has proven to provide information unavailable by traditional sampling methods simply due to practical restrictions of access and taking undisturbed samples at various depths. Core sampling can be conducted only at selected locations with sufficient access, while the gamma scanning can be performed at numerous locations to form a more complete profile. It is also much less expensive and faster than core sampling. Using the skate and tablet PC system, one can typically scan 5 riser ports on a Type III tank in less than half a day at approximately two percent of the cost of sample analysis. While it does not yield the breadth of information about the salt cake that core samples do, it does provide an important input for formulating salt removal strategies and monitoring the effectiveness of salt handling operations.

ACKNOWLEDGEMENTS

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REFERENCES

(1) D.M.C. ODELL, B.S.BUSHART, L.J. HARPRING, F.S. MOORE, T.N. RILEY; Zero Dead Time Spectroscopy Without Full Charge Collection, *Proceedings of the Ninth Symposium on Radiation Measurements and Applications, Nuclear Instruments and Methods in Physics Research A, 422, 363,*

(2)Microshield 5, Grove Engineering, Rockville Md,1996

(3) MCNP4C, Los Alamos National Laboratory, Los Alamos NM, 2000