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NEUTRON AND GAMMA PROBE APPLICATION TO HANFORD TANK

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ABSTRACT

A neutron (moisture-sensitive) and gamma (in-situ radiation) probe technique has been utilized at a number of Hanford radioactive waste tanks for many years. This technology has been adapted for use in tank 241-SY-101's two Multifunction Instrument Trees (MITs), which have a hollow dry-well center opening two inches (5.1 cm) in diameter. These probes provide scans starting within a few inches of the tank bottom and traversing up through the top of the tank, revealing a variety of waste features as a function of tank elevation. These features have been correlated with void fraction data obtained independently from two other devices, the Retained Gas Sampler (RGS) and the Void Fraction Instrument (VFI). The MIT probes offer the advantage of nearly continuous count-rate versus elevation scans, and they can be operated significantly more often and at lower cost than temperature probes or the RGS or VFI devices, while providing better depth resolution.

The waste level in tank 241-SY-101 had been rising at higher rates than expected during 1998 and early 1999, indicating an increasing amount of trapped gas in the waste. The use of the MIT probes has assisted in evaluating changes in crust thickness and level, and also in estimating relative changes in gas stored in the crust. This information is important in assuring that the tank remains in a safe configuration, and will support safe waste transfer when those operations take place.

Based on the MIT probe data starting in February 1999, the 241-SY-101 crust surface elevation and thickness increased during the intervening months into May 1999. During this period, the bottom-of-the-crust elevation decreased to the point that there was concern that it might reach the mixer pump inlet, potentially reducing the pump efficiency. However, subsequent to the operation of the Mechanical Mitigation Arm (MMA) in May 1999, the crust surface level decreased slightly, while the crust bottom elevation increased (and crust thickness decreased) steadily until about mid-July, and then stabilized. The MMA had introduced crust penetrations that provided for a controlled release of some of the stored gas, as intended.

INTRODUCTION

There are a number of radioactive waste storage tanks at Hanford in which potentially flammable gases are produced. These tanks continue to undergo close scrutiny, evaluation and remediation (as required) to assure that they remain in a safe configuration. Perhaps the best known of these tanks has been the double shell tank 241-SY-101 (SY-101). For years prior to 1993, hydrogen gas generated in the waste periodically built up at lower tank levels to the point that increased buoyancy caused a 'roll-over' event in which the bottom waste exchanged position with the waste at the top, releasing a relatively large quantity of gas. Often after such a gas release event (GRE) in SY-101, the hydrogen flammability limit was exceeded in the tank headspace for a short time, until exhaust fans exchanged enough air to reduce the hydrogen concentration. Although the tank was engineered to eliminate ignition sources, an additional margin of safety was achieved in July 1993 when a high power mixer pump was installed in the tank. This mixer pump was intended to prevent the build up of trapped gases in the waste by providing for continuous gas release at headspace concentrations well below the flammability limit.

Prior to the installation of the SY-101 mixer pump, GREs were occurring roughly every 100 days; there have been no GREs in this tank since the installation/operation of the pump. However, the crust surface level of SY-101 started to rise in late 1997, even though no additional material had been added to the tank. This rise was assumed to indicate an increase in gas retention within the waste, causing an increase in waste volume. Thus, the rising waste level might be an indication of a decreasing margin of safety for the SY-101 tank.

In order to mitigate potential problems that might arise from the increasing crust level, an aggressive program was developed to transfer a portion of the radioactive waste in SY-101 to Tank 241-SY-102 (1). As part of this program, a mechanical mitigation arm (MMA) was developed and deployed in SY-101 to produce relatively small holes in the waste crust. This step released some of the gas trapped in the waste, providing temporary relief from crust growth, and has allowed more time to complete the waste transfer operations. When the transfer operations are started, waste in both tanks will be diluted with water to reduce viscosity, which is expected to reduce the quantity of gases trapped in the waste.

Neutron and gamma probe hardware in use at Hanford has been adapted for use in SY-101 MITs, starting in February 1999. These instruments have provided data that has been useful in evaluating and tracking gas void concentrations within the SY-101 waste, as will be discussed. These MIT probes offer the advantage of nearly continuous count-rate versus elevation scans, and they can be operated significantly more often and at lower cost than temperature probes or the RGS or VFI devices relied on previously for this type of data. The exact depth of major waste features is also much more accurately determined using the neutron and gamma probes.

It is expected that use of these MIT probes at SY-101 will continue after the waste transfer operations have been completed in order to monitor the new conditions in this tank.

EQUIPMENT

Liquid Observation Well Probes

Neutron probes have been used at Hanford for a variety of purposes for more than 20 years. Currently, both neutron and gamma probes are being used in liquid observation wells (LOWs) installed in more than 60 radioactive waste storage tanks. The LOWs are typically fiberglass closed-end tubes, of nominally 3.5-inch (8.9-cm) outside diameter, generally 40 ft to 60 ft (12 m to 18 m) long, inserted vertically into the waste of a given tank with the closed end near the tank bottom. A neutron (or gamma) probe is lowered to the bottom of the LOW, then raised slowly while providing count-rate versus elevation data. Thus, a 'scan' of waste properties can be obtained without contaminating the test probe. A custom control file is provided for each well that allows the LOW van computer to adjust scanning speed to provide 'high-resolution' data through zones of particular interest.

Variations in count-rate usually correspond to variations in moisture content in the vicinity of the LOW for both the neutron and gamma probes. The neutron probe contains a 'fast' neutron source (AmBe) and a 'slow' neutron detector (BF_3); water is very effective in slowing the source neutrons into the energy range of the detector (due to hydrogen atom collisions with the fast neutrons). Thus, high neutron probe count-rates are associated with relatively high moisture content of the waste surrounding the LOW at a given elevation. The gamma probe uses a standard Geiger-Muller (GM) tube, which includes sensitivity to radioactive isotopes, primarily cesium in this application. Due to its relatively high solubility, the cesium tends to follow the moisture in the waste, so that the gamma probe is also sensitive to tank moisture. These two separate instruments used together provide independent verification of moisture features observed in the tank waste.

These probes are operated from a mobile LOW van that provides power, computerized counting instrumentation, and vertical positioning capability. The van computer controls the probe vertical position via a highly accurate cabling system. The probe can be raised or lowered throughout the LOW's nominal 50 ft (15.24 m) length while the probe's elevation is known accurately to within 0.25 inches per 100 ft (0.208 mm per meter) of LOW length, by design specification. In practice, the actual elevation accuracy is often better than 0.10 inches (2.54 mm) at the typical 40 – 60 ft (12 – 18 m) survey depth.

The LOW probes are used primarily to track the interstitial liquid level (ILL) in these waste tanks. A change in the ILL, particularly a decrease in elevation, can indicate a leak in the tank.

MIT Probes

Use of neutron and gamma probes in SY-101 can assist in understanding the rising crust level, and would have the potential of identifying and tracking elevations of gas buildup in the tank waste. (Increasing gas void in the waste effectively decreases moisture content, causing a reduction in probe count-rate at that particular elevation.) However, an LOW does not exist in SY-101, and installing one was known to be prohibitively expensive.

A viable alternative was to modify the LOW probe design to reduce the probe diameter from 2.75 inches (6.99 cm) to nominally 1.85 inches (4.70 cm); the modified probe could then be lowered down the 2 inch (5.08 cm) validation tube of either of two MITs already installed in SY-101. An MIT is a vertical assembly extending from above the tank to within a few inches of the tank bottom, and containing an array of thermocouples to measure the waste temperature as a function of elevation. The validation tube is a hollow dry-well central opening that runs nearly the length of the MIT; it is used to calibrate MIT thermocouples (TCs) by allowing a calibrated temperature device to be lowered to matching TC elevations for comparison of their readings.

MITs are made of 300 series stainless steel (rather than fiberglass), and for the length of interest (portion embedded in the headspace and waste), essentially consist of a tube within a tube, separated by bulkheads and sealed at the bottom. The outer tube has an outside diameter of nominally 3.5 inches (8.89 cm), while the inner validation tube has an inside diameter of 2 inches (5.08 cm); TCs pass through the bulkheads and are attached at various elevations between the tubes. For the probes, the effective outside diameter of the MIT of 3.5 inches (8.89 cm) matches the LOW diameter, while the difference between stainless steel and fiberglass has only a minor effect on the operation of the probes. The MIT steel is actually more ‘invisible’ to the neutrons than the LOW fiberglass, but is slightly more shielding for the gamma probe.

The new MIT probes have a reduced design diameter of 1.85 inches (4.70 cm), and an approximate length of 28 inches (71.1 cm). The reduction in probe diameter required procurement of new detectors for both types of probes, consistent with the new probe geometry. The MIT probes are fully compatible with the existing LOW van hardware and instrumentation, which resulted in a significant cost savings in extending service to SY-101, since no new hardware or procedures were required (other than the probes).

Operation

An illustration of the operation of the MIT probes is given in Figure 1, in which actual neutron probe data obtained on February 18, 1999 is attached as an example. The LOW van is driven to the location of the appropriate tank riser, and the probe is set up over a pulley assembly specially attached to the MIT valve cover flange. After setting the ‘zero’ elevation point, the probe is lowered by the computer-controlled winch to near the tank bottom, and count-rate data collection is initiated. During data acquisition, the probe is raised slowly to the top of the riser at varying rates to allow increased data resolution at the elevations of special interest.

The LOW van computer tracks elevation as measured from the bottom of the probe; however, during later analysis, a probe offset is added to this raw data elevation to compensate for the ‘effective’ position of the detectors within the probe. For the MIT gamma probe, an offset of 1.8 inches (4.57 cm) is determined from the geometry of a radial slot cut in a tungsten shielding block positioned around the detector. The MIT neutron probe offset of 5.83 inches (14.81 cm) was determined from a series of water submersion tests in which the count-rate is tracked versus elevation as the probe is lowered through the surface of a water bath. This count-rate data was compared to the known elevation of the air/water interface to determine the probe offset.

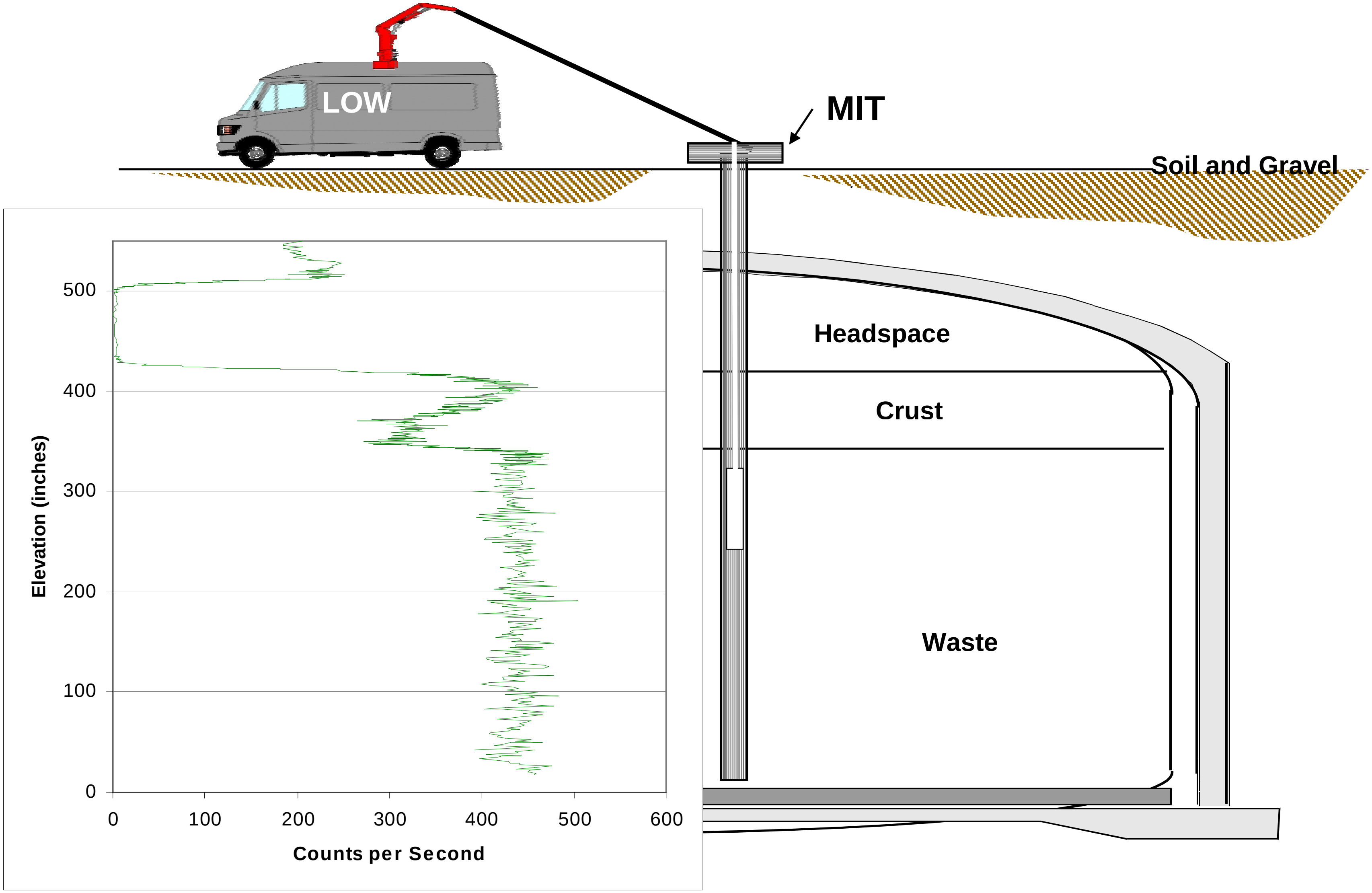

Figure 1. Illustration of MIT Neutron Probe Lowered into MIT 17B on 2-18-99.

DATA ANALYSIS

Data Interpretation

The February 18, 1999 data scans (including offset adjustment) for both MIT neutron and gamma probes results are plotted in Figure 2, along with Retained Gas Sampler (RGS), Void Fraction Instrument (VFI) and temperature probe data. For comparison purposes, the neutron and gamma probe count-rates have been divided by 5.5 and 90, respectively, allowing their presentation on the same scale as the RGS, VFI and temperature data. Note that the temperature probe data (2) was obtained four days after the MIT neutron and gamma probe runs (in the validation tube of the same 17B MIT); the RGS (3) and VFI (4) data was obtained three to five months earlier.

From near the tank bottom to about the 340-inch (864-cm) elevation, all five data sets indicate a nearly homogeneous waste condition. The relatively high neutron probe count-rate is consistent with this region being a low void fraction slurry, a conclusion supported by the RGS and VFI data which indicate that the void fraction is generally less than 3 percent.

At about the 340-inch (864-cm) elevation, there is a steep decrease in the neutron probe count-rate, which corresponds with an equally steep increase in the RGS and VFI void fraction (to 43 percent), at least within the resolution of the available data. The gamma probe count-rate and temperature probe data also show a decreasing inflection point at this level that is consistent with increasing void fraction.

The neutron probe data show a roughly 30 inch (76 cm) plateau from approximately 340 to 370 inches (864 to 940 cm) in elevation, which has been interpreted as a bubbly, high void fraction layer at the bottom of the waste crust. Above 370 inches (940 cm), based on the neutron probe data, there is steadily decreasing void fraction until about the 410-inch (1041-cm) elevation, an interpretation that is also supported by the RGS and VFI results where comparable data are available.

Above 410 inches (1041 cm), the neutron probe count-rate decreases steadily to near zero by the 430-inch (1092-cm) elevation mark; this decrease is interpreted as due to the crust drying out at the top, rather than occurring because of increasing void fraction. The gamma probe count-rate also decreases steadily to a clear inflection point at the 430-inch (1092-cm) elevation, which is also interpreted as due to reaching the top of the crust. Independent mechanical instrumentation (ENRAF R1A) measured the surface of the waste at 429.2 inches (1090.17 cm) in elevation on February 18, 1999 (5). Since the crust has a rough surface, varying several inches in height between the measurement locations, this agreement with the neutron and gamma probe values may be partially coincidental.

Above the 430-inch (1092-cm) elevation, the neutron probe count-rate remains low since there is relatively little moisture in the headspace within the limited radial probe range, estimated at 6 to 8 inches (15 to 20 cm) beyond the MIT outside diameter. Once the neutron probe reaches

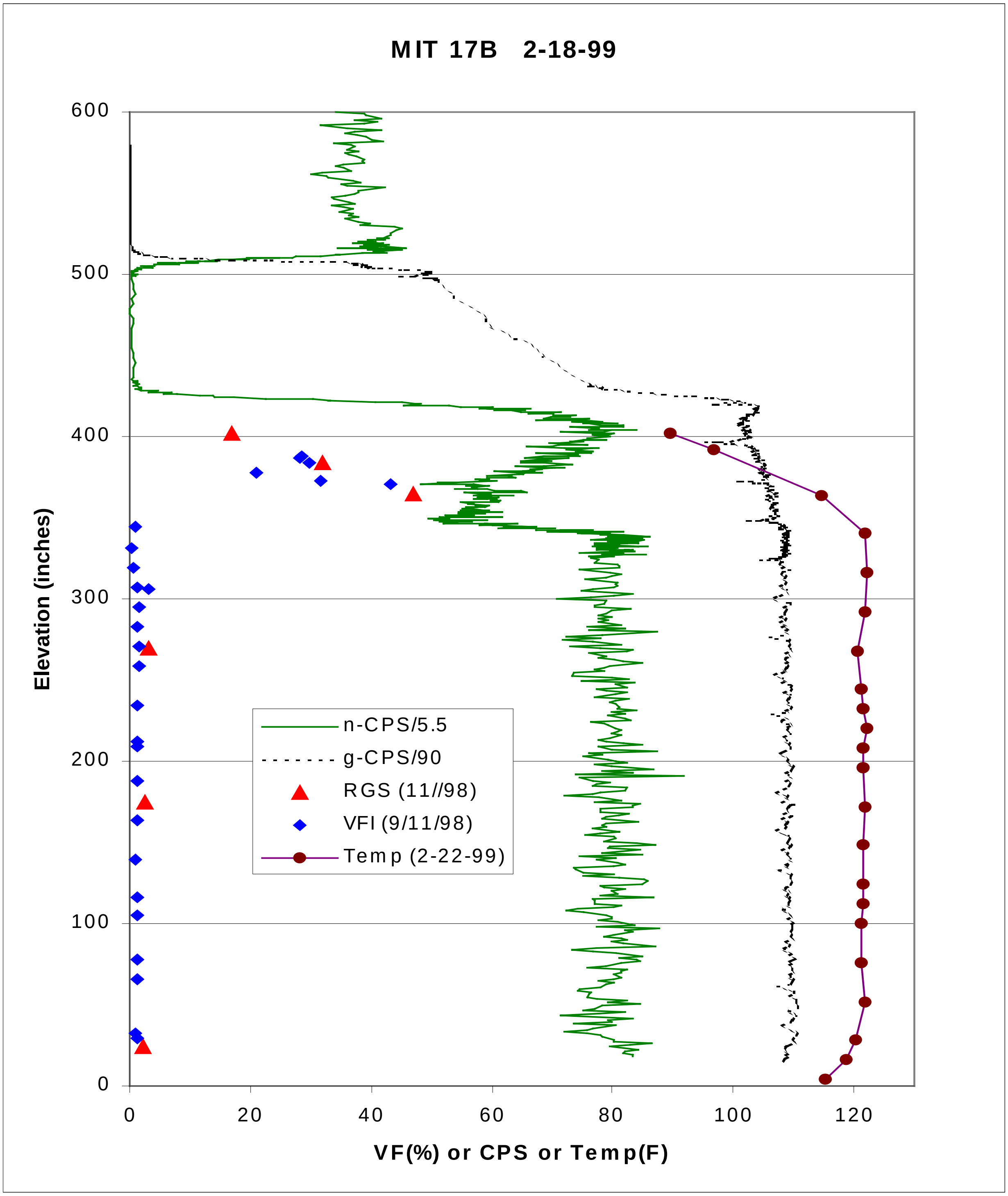

Figure 2. Comparison of MIT Neutron and Gamma Probe Results (2-18-99) with RGS, VFI, and Temperature Probe Data.

the 500-inch (1270-cm) level, the count-rate increases again as it is drawn up through the moisture-laden concrete, soil, and gravel above the top of the tank. Although the gamma probe count-rate continues to decrease with elevation in the headspace, it doesn't drop to zero because the gamma radiation off the waste surface has a much greater effective range (until it becomes shielded when the probe exits the headspace). The 'bump-up' in gamma probe count-rate just prior to reaching the waste surface may be due to concentration of radioactive cesium near the surface due to some sort of a drying out process.

Data Analysis Technique

Although the combined MIT neutron and gamma probe data provides an opportunity to extract more detailed information about the tank crust, due to space limitations for this paper, only crust top and bottom elevations, and crust gas retention trends will be discussed in detail.

For a given MIT neutron or gamma probe scan, the crust top and bottom elevations are determined from inflection points in the scan plot; by definition, these inflection points are the intersection of two lines manually fitted to the scan plot at the elevation corresponding to the feature in question. For example, the average of the neutron probe count-rate from the 100 to 300-inch (254 to 762-cm) elevation is drawn on a plot of the scan, and another line is drawn along the steep count-rate decrease corresponding to the crust bottom. The intersection of these two lines is defined as the elevation for the bottom of the crust. At the top of the crust, two similar lines are graphically fitted to the data at the point where the neutron probe count-rate decreases to essentially zero, with their intersection defining the elevation of the crust top. A similar process is also performed for the gamma probe data. Although more sophisticated techniques for determining the crust top and bottom elevations from the MIT probe data will probably be utilized in the future, the simple methods described here are considered effective for tracking trends. The relative changes in crust feature elevations are determined by subtraction, a process expected to cancel elevation offsets that might vary between techniques.

The following method is also used for calculating a 'crust gas inventory' (CGI) factor, intended to be proportional to the gas retained in the waste crust. Although not quantifying the amount of gas stored in the crust, this factor can be used to indicate relative changes in the stored gas content. The CGI is calculated by determining the area (through numerical integration) produced by the dip in the neutron probe count-rate below the average count-rate observed in the lower tank homogeneous slurry. The boundaries of the numerical integration are set from the bottom of the crust to the mid-point of the maximum count-rate peak before the count-rate falls to near zero at the crust top. For example, the CGI integration bounds would be from about 340 to 406 inches (864 to 1031 cm) in elevation for the Figure 2 neutron probe data.

A major assumption in defining the CGI factor as given above is that the neutron probe count-rate decreases linearly with increasing waste gas quantity. Although this assumption is reasonable to a point, it is known that the single detector BF_3 response to moisture is not linear, and its sensitivity decreases with increasing moisture. In general, the response typically forms a hyperbolic curve, although the exact shape of that curve for this particular detector is not currently known. Also, the upper bound for the integration is defined at a point where two different mechanisms (decreasing void fraction and loss of moisture at the crust top) are affecting the probe count-rate, which may lead to an additional CGI calculation error. Despite these problems, the CGI factor is considered a useful tool for tracking relative changes in crust gas retention.

RESULTS

The first runs of both the reduced diameter neutron and gamma probes in both of the two SY-101 MITs occurred on February 18, 1999. MIT probe runs have continued through the present, and are planned to continue after a portion of the contents of SY-101 have been transferred to another double shelled tank. This will allow tracking of the effect of the waste transfer on the properties of the waste left in SY-101. However, the results presented here will include only the data obtained up to scans performed on November11, 1999, in order to comply with publication deadlines.

Crust elevations and the CGI factors determined from the MIT probe data obtained between February and November of 1999 are given in Table I. The values given in this table for the crust top and bottom elevations represent an average of the neutron and gamma probe results for the particular MIT indicated, unless otherwise noted. Figure 3 presents the trends for these crust elevations, and Figure 4 indicates the CGI trends over the described time period.

As can be observed from the Figure 3 trends, the elevation of the bottom of the crust decreased steadily until about June 1, 1999, while the elevation of the crust top stopped increasing between May 3 and May 12, 1999. The operation of the Mechanical Mitigation Arm (MMA) to puncture the SY-101 crust surface is considered to be a major factor in producing the change observed in the crust growth trends, particularly for the bottom of the crust. The MMA was first used on May 20, 1999, and then again on May 26, 1999. The bottom of the crust elevation stops decreasing on June 1, 1999, which is consistent with the MMA operation reducing (over time) the crust gas inventory (see Figure 4). Although the rise in elevation of top of the crust appears to have slowed before the operation of the MMA, the first significant elevation decrease occurs on May 24, 1999, after the first penetration of the crust.

The inlet of the SY-101 mixer pump is 236 inches (600 cm) above the tank bottom (6); as the bottom of the crust level decreased, there was concern that it might reach the pump inlet. If the higher void faction waste at the bottom of the crust was drawn into the mixer pump inlet, it is likely that the pump efficiency would have been decreased. Thus, the MIT neutron and gamma probe data has shown that the operation of the MMA has alleviated this potential problem.

Starting in April 1999, the bottom of the crust feature started to broaden as its elevation began to decrease. This suggested that the transition to high void fraction at the crust bottom became more gradual as the total crust gas inventory built up. This gradual transition was retained after the MMA operation reduced the crust gas inventory (see Figure 4). However, in August 1999, the transition region at the bottom of the crust again began to become more and

TABLE I

Summary of MIT Probe Results

MIT Probe Run Date	MIT 17B			MIT 17C			AVERAGE		
	Crust Top in (cm)	Crust Bottom in (cm)	CGI	Crust Top in (cm)	Crust Bottom in (cm)	CGI	Crust Top in (cm)	Crust Bottom in (cm)	CGI
02-18-99	428.2 (1087.6)	339.6 (862.6)	12.15	430.1 (1092.5)	345.8 (878.3)	11.93	429.1 (1089.9)	342.7 (870.5)	12.04
03-11-99	430.0 (1092.2)	337.8 (858.0)	12.06	432.5 (1098.6)	342.0 (868.7)	12.65	431.2 (1095.2)	339.9 (863.3)	12.35
04-16-99	432.3 (1098.0)	323.8 (822.5)	13.04	436.3 (1108.2)	323.0 (820.4)	13.64	434.3 (1103.1)	323.4 (821.4)	13.34
05-03-99	433.6 (1101.3)	306.6 (778.8)	14.93	437.5 (1111.3)	305.0 (774.7)	14.91	435.6 (1106.4)	305.8 (776.7)	14.92
05-12-99	433.5 (1101.1)	295.9 (751.6)	15.12	437.6 (1111.5)	302.9 (769.4)	15.53	435.6 (1106.4)	299.4 (760.5)	15.32
05-17-99	434.2 (1102.9)	297.3 (755.1)	15.68	437.3 (1110.7)	300.9 (764.3)	13.87	435.7 (1106.7)	299.1 (759.7)	14.77
05-24-99	433.9 (1102.1)	294.1 (747.0)	14.89	437.0 (1110.0)	295.4 (750.3)	15.59	435.4 (1105.9)	294.7 (748.5)	15.24
06-01-99	432.5 (1098.6)	294.5 (748.0)	14.11	436.3 (1108.2)	293.2 (744.7)	14.38	434.4 (1103.4)	293.8 (746.3)	14.25
06-14-99	433.6 (1101.3)	303.6 (771.1)	12.95	436.9 (1109.7)	293.8 (746.3)	15.95	435.2 (1105.4)	298.7 (758.7)	14.45
06-24-99	433.6 (1101.3)	307.2 (780.3)	14.08	436.4 (1109.7)	304.9 (774.4)	13.37	435.0 (1105.4)	306.0 (777.2)	13.73
07-06-99	434.2 (1102.9)	310.8 (789.4)	16.25	NA	NA	NA	NA	NA	NA
07-15-99	431.8 (1096.8)	310.7 (789.2)	12.28	434.2 (1102.9)	309.6 (786.4)	14.96	433.0 (1099.8)	310.1 (787.7)	13.62
07-26-99	430.8* (1094.2)	311.5 (791.2)	12.73	433.9* (1102.1)	310.7 (789.2)	13.67	432.4* (1098.3)	311.1 (790.2)	13.20
08-02-99	431.2* (1095.2)	311.2 (790.4)	13.75	433.5* (1101.1)	311.5 (791.2)	13.17	432.4* (1098.3)	311.4 (791.0)	13.46
08-11-99	430.8* (1094.2)	311.2* (790.4)	14.64	433.2* (1100.3)	309.9* (787.1)	15.63	432.0* (1097.3)	310.6* (788.9)	15.14
08-23-99	432.5 (1098.6)	312.7 (794.3)	14.85	435.1 (1105.2)	311.8 (792.0)	15.43	433.8 (1101.9)	312.3 (793.2)	15.14
08-30-99	431.6* (1096.3)	312.4* (793.5)	14.76	433.0* (1099.8)	311.4* (791.0)	15.29	432.3* (1098.0)	311.9* (792.2)	15.02
09-07-99	431.4* (1095.8)	313.3* (795.8)	14.74	433.0* (1099.8)	312.2* (793.0)	15.48	432.2* (1097.8)	312.8* (794.5)	15.11
09-13-99	432.0* (1097.3)	311.9* (792.2)	15.23	433.4* (1100.8)	311.8* (792.0)	15.42	432.7* (1099.1)	311.9* (792.2)	15.33
10-05-99	432.5 (1098.6)	315.3 (800.9)	14.64	436.3 (1108.2)	314.4 (798.6)	15.52	434.4 (1103.4)	314.9 (799.8)	15.07
10-11-99	432.5 (1098.6)	316.2 (803.1)	13.87	435.4 (1105.9)	314.7 (799.3)	14.64	434.0 (1102.4)	315.5 (801.4)	14.26
10-21-99	432.6 (1098.8)	314.9 (799.8)	14.54	435.6 (1106.4)	314.8 (799.6)	15.83	434.1 (1102.6)	314.9 (799.8)	15.19
11-05-99	431.6* (1096.3)	315.6* (801.6)	12.66	433.6* (1101.3)	313.8* (797.1)	15.29	432.6* (1098.8)	314.7* (799.3)	13.98
11-11-99	431.0* (1094.7)	315.8* (802.1)	13.30	433.7* (1101.6)	313.5* (796.3)	14.84	432.4* (1098.3)	314.7* (799.3)	14.07

* Gamma Probe Data Not Available at this Elevation – Result is Based on Neutron Probe Data Only

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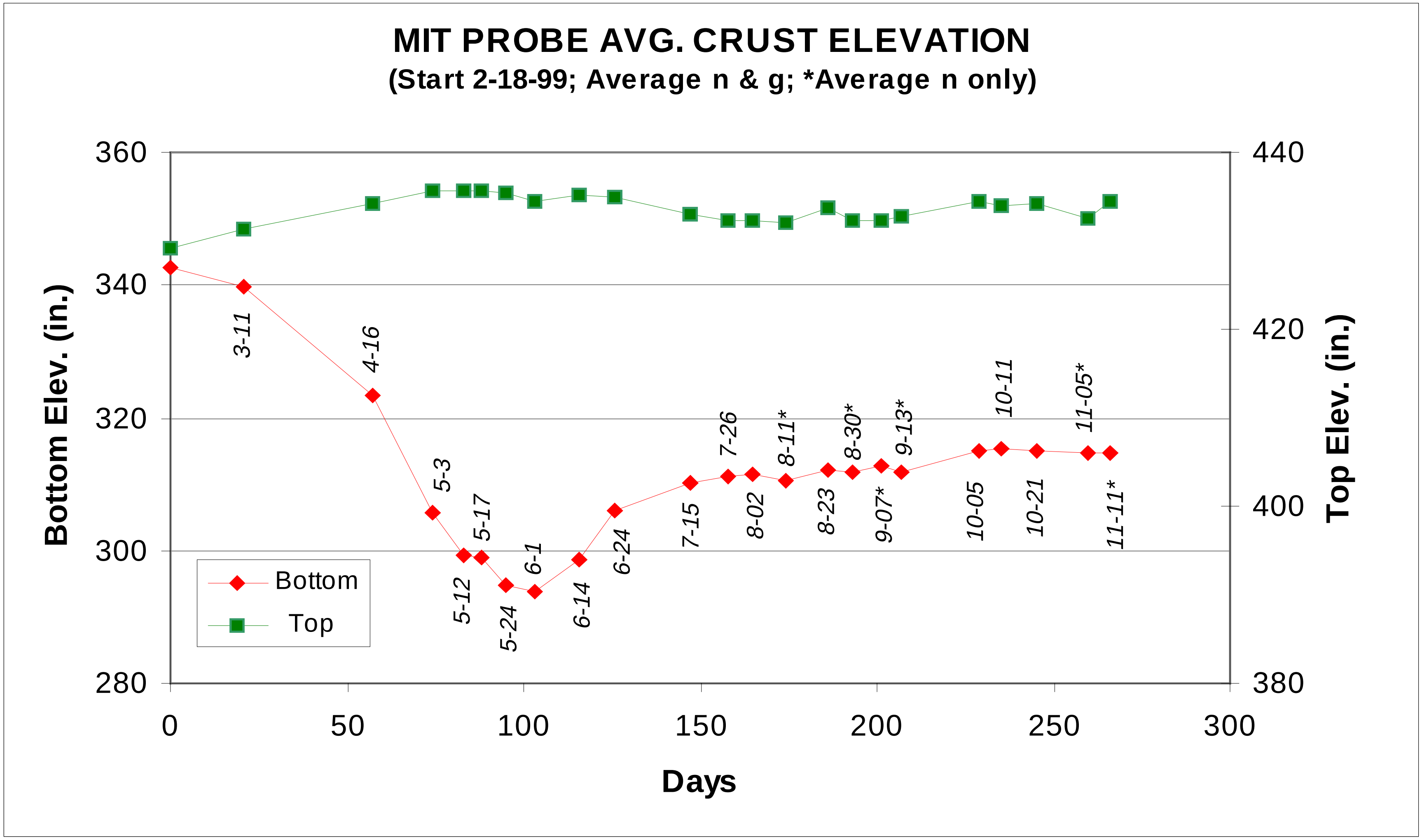

Figure 3. Trending Plot of the Top and Bottom of the Waste Crust Elevation as determined from the MIT Neutron and Gamma Probe Data.

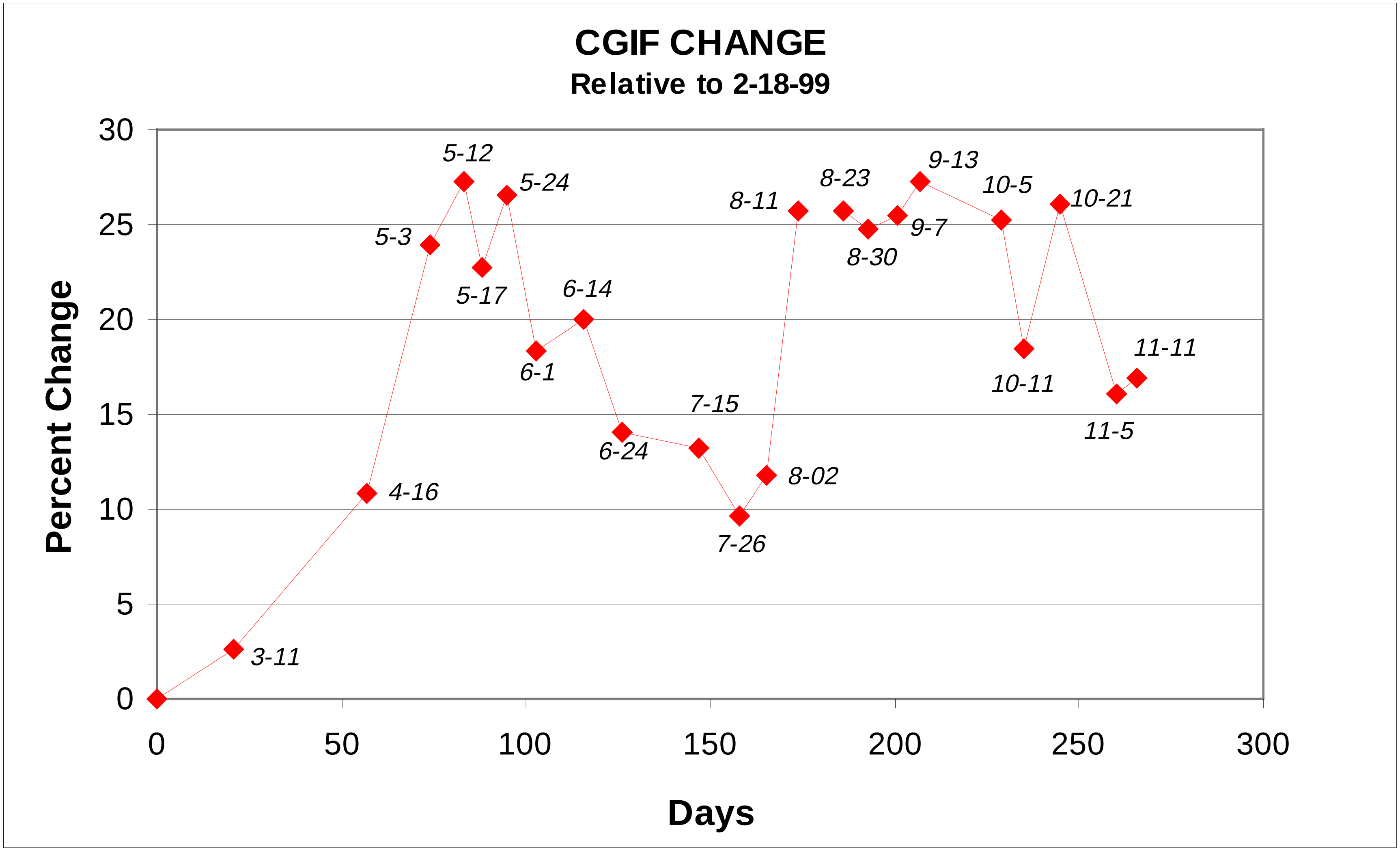

Figure 4. Relative Change (from 2-18-99) of the Stored Gas Inventory of the Crust (as defined by the CGI Factor)

more narrow, an effect that appears to correspond to the increase in total crust gas inventory starting in August, as observed in Figure 4.

It is possible that the increase in crust gas inventory and the increasing transition gradient at the crust bottom may signal that the bottom of the crust may again begin to grow down in elevation toward the pump inlet. Most likely, the partial waste transfer (and back-dilution) from SY-101 will begin before this occurs. Regardless, the MIT neutron and gamma probes will continue to provide a valuable tool to monitor the elevations and trends of various crust features within SY-101.

FUTURE PLANS

Several additional double-shell tanks are on the "Flammable Gas Watch List" at Hanford, indicating that they at least have the potential to retain and possibly release explosive levels of flammable gasses. Since most of these tanks are also equipped with MITs, this same neutron and gamma probe technique will be applied to all appropriate tanks during Fiscal Year 2000. The information obtained should provide important additional understanding about the nature of these retained gasses, and assist in engineering evaluations of the potential for sudden gas release events. The fact that this data can be obtained relatively inexpensively is an added advantage in extending the application of the MIT probes to these tanks.

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