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PHYSICAL MECHANISMS CONTRIBUTING TO THE EPISODIC GAS RELEASE FROM HANFORD TANK 241-SY-101

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ABSTRACT

Volume growth of contents in a waste storage tank at Hanford is accompanied by episodic releases of gas and a rise in the level of tank contents. A theory is presented to describe how the gas is retained in the waste and how it is released. The theory postulates that somewhat cohesive gobs of sludge rise from the lower regions of the tank and buoyancy overcomes the cohesive strength of the slurry; this quantitatively explains several of the measured phenomena and qualitatively explains other observations.

INTRODUCTION

Tank 241-SY-101 is a 3.8×10^6 liter (1 million gallon), double wall, waste storage tank (see Figure 1) that has been used to store concentrated radioactive defense waste since 1977.

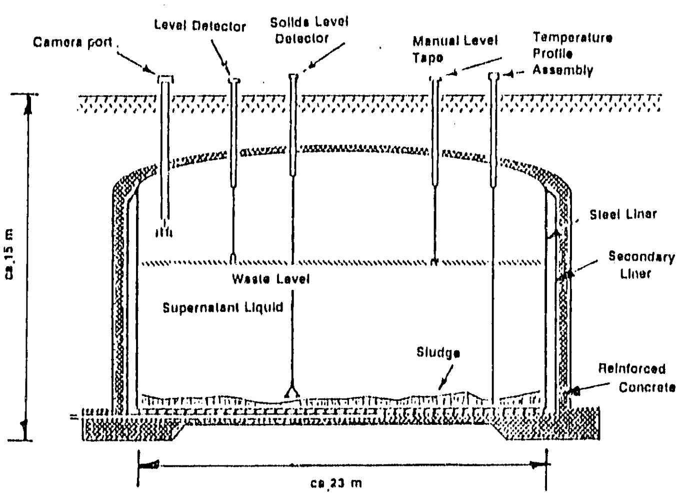

Fig. 1. Configuration of Hanford Double Shell Waste Tank, 241-SY-101

Several different batches of waste were added to the tank from 1977 until 1980. Monitoring of the tank liquid level showed that the level of fluid was rising in the tank; this phenomenon was termed "level growth". The general rise in level was interrupted periodically by level drops, which were quite rapid, occurring in less than 1 day. The level drops were accompanied by the sudden release of gas into the tank head space and through the filtered vent system. These releases, termed "burps", were found to contain non-radioactive gases, but hydrogen and nitrous oxide were present in potentially flammable ratios and concentrations in some large releases. This fact categorized the tank as a safety concern. Research investigating the causes of gas generation and the large periodic releases was initiated to find ways to mitigate the problem.

This safety problem would be eliminated if the gas production were stopped or if the gas was released in smaller batches, or steadily, rather than in large "burps". The purpose of this research was to explore physical mechanisms involved in the "burp" phenomenon to provide insights into what physical mitigation techniques might be feasible, particularly if other schemes that stop the generation of flammable gases were not feasible. This paper discusses some analyses performed, that were fundamentally based on data obtained by Westinghouse Hanford Company, operators of the tank farm. The analyses suggest a mechanism by which the gases are retained and periodically released in large amounts. The suggested mechanism also agrees with other data obtained from analyses of tank 241-SY-101.

TANK 241-SY-101 DATA

Level

Figure 2 shows an example of the long-term history of the waste level in the tank. The sudden large drops in level are accompanied by a release of considerable volumes of gas. The level drops have occurred in a range of sizes and a size spectrum shows that small drops are statistically more frequent than large level drops. Figure 3 is a magnified view on log-log scale of the level for a time after a release. In this figure the level has the small basal rate of level growth subtracted (to show the effect of the gas being released) and appears to be following a power-law function of time until between 6 and 10 days have passed.

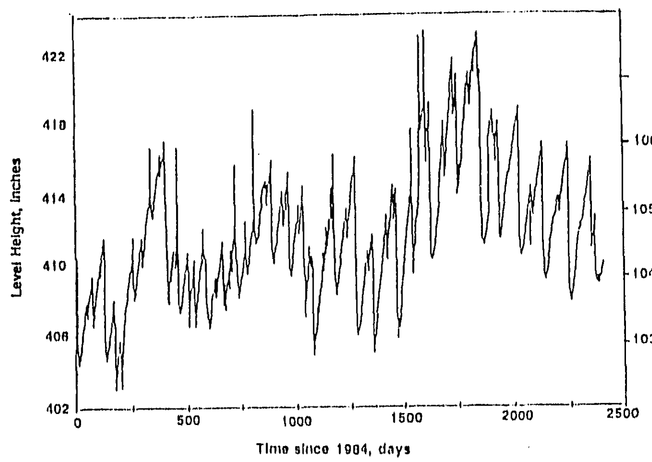

Fig. 2. Level History In Tank 241-SY-101[2]

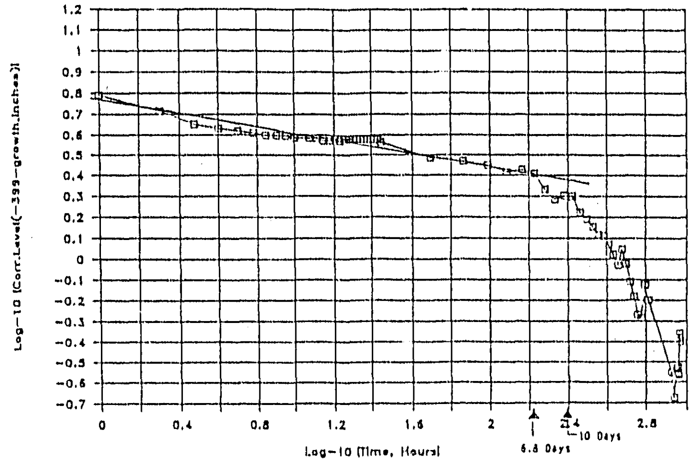

Fig. 3. Level Data for Several Days after the Oct. 24, 1990 Gas Release

Temperature

Figure 4 shows two temperature profiles (before and after release) taken with the single thermocouple stalk during a release in 1990. Before the release, the temperature shows a warm bulge (100 to 500 cm in height) in the lower part of the tank; a constant temperature layer in the mid-section of the tank (500 to 900 cm); and above that (1000 to 1200 cm), a rapid temperature drop to the head space, which is ventilated. The temperature bulge was surmised to be caused by the retention of radioactive decay heat in a nonconvective layer near the bottom of the tank. Above that layer is a convective layer (500 to 900 cm in height) in which the temperature profile is flat because of convection. The temperature drop in the head space was thought to be caused by crust about 1.3-m thick at the top of the waste bulk. During the release event, the sequence of temperature profiles showed that the temperature bulge in the lower part was removed in about 1 minute, while a warm temperature bulge appeared in the upper part of the tank. Figure 5 shows the thermal profile for several days after the release event. One can see that after a

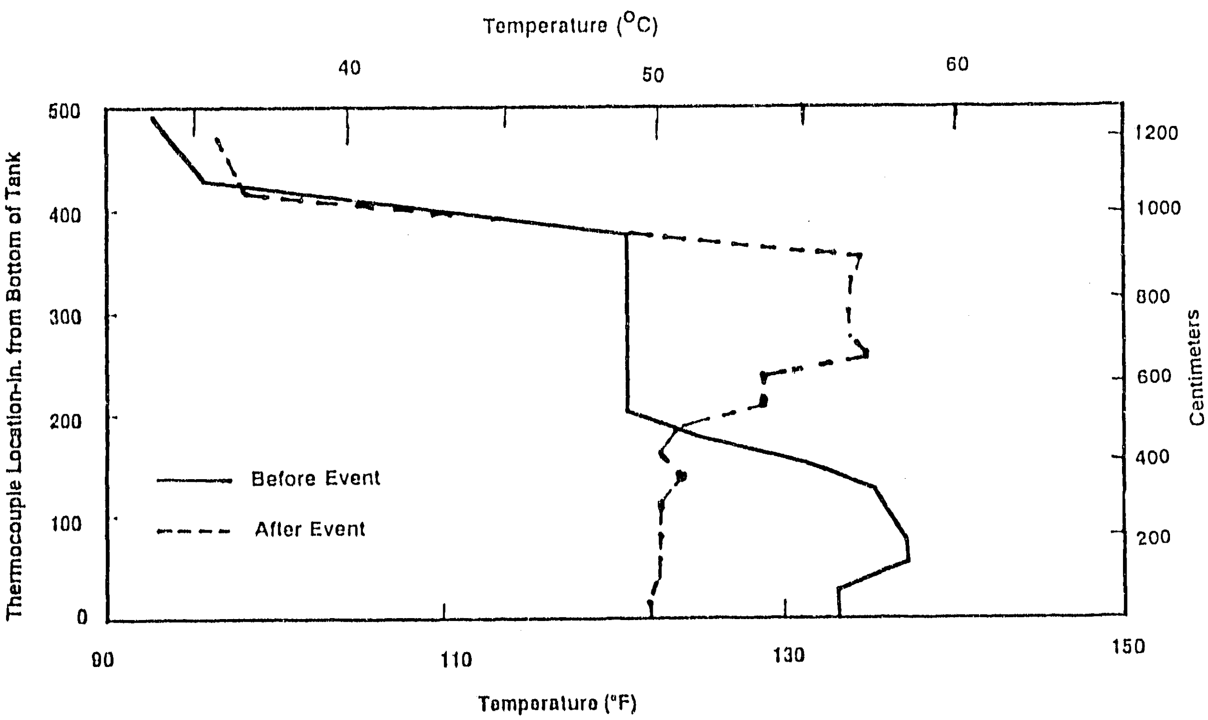

Fig. 4. Temperature Profiles In Tank 241-SY-101 Before and After October 24, 1990 Gas Release

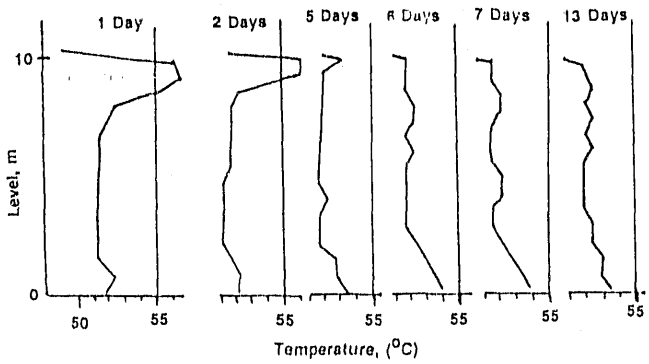

Fig. 5. Temperature Profiles Several Days After Oct. 24, 1990 Event

few days the temperature at the bottom begins to rise again.

Waste Composition and Properties

Samples of the waste extracted by the tank operators have been analyzed. The waste appears to be a sludge or a slurry and is a highly basic, saturated mixture. The waste consists of a mixture of solution (supernatant liquid) and solids containing mostly sodium, nitrate, nitrite, hydroxyl, aluminum, organic chelating agents, and other constituents. The specific gravity of the liquid fraction is 1.3 to 1.5 and of the solid-containing-sludge is 1.6 to 1.7. The sludge shows a significant shear strength and behaves as a thixotropic non-Newtonian power-law fluid when measured for viscosity.

Video and Photographs

Still photos of the upper surface of the waste in the tank showed a rolled surface of white and dark areas. The dark areas seemed to indicate liquid presence. In 1991, black and white videos have been taken of two releases. These show upwellings of liquid and gas pushing flotsam (such as tank measurement equipment) aside. The upwellings are associated with pressure pulses in the vent line. The series of upwellings (each of a few seconds duration) last for minutes to hours, and with each upwelling there is a subsequent "fizzing" of smaller bubbles reaching the surface and bursting.

THEORY OF PARTIAL ROLLOVER GAS RELEASE

The inversion of the temperature profile (Figure 4) was termed "rollover". An initial calculation of an unstable temperature overturn in a cylindrical vessel was performed by Eyler[a] using the hydrodynamic code, TEMPEST. This two-dimensional simulation showed the bottom layer rising to the top in several places almost simultaneously. This suggested that complete rollover might not be the preferred mode, and that the sludge might rise in gobs of material; only part of the sludge would rise to the top and release gases during a given release event. The times for the temperature overturn and some features of the temperature profile were reproduced by the numerical simulation, but the simulation was not detailed enough to handle the separation of the gas phase or to predict the level in the tank subsequent to the main release.

The Gob Theory

A proposed theory of partial rollover that can explain many of the quantitative and qualitative observations is called the "gob theory". In this theory a portion of the bottom sludge in the tank retains enough of the gas being produced by the waste, that this portion (gob) breaks loose from the sludge, mass or layer buoyancy overcomes the cohesive forces and viscosity. The gob then rises rapidly to the top of the bulk waste and almost instantly releases the largest bubbles of gas that have already coalesced (or releases gas rapidly because the gas has expanded and coalesced because the motion has reduced the strength of the sludge matrix). The gas still remaining in the gob in smaller size bubbles, percolates ("fizzes") out at a slower rate until the amount of gas can no longer hold the cohesive solid fraction afloat. Then the gob material begins to sink, and the remaining gas is recompressed by hydrostatic pressure.

Results of Calculation

Figure 6 shows the buoyancy stress and the yield strength of the in-tank sludge containing a uniform mass fraction of gas bubbles. The strength of the sludge is

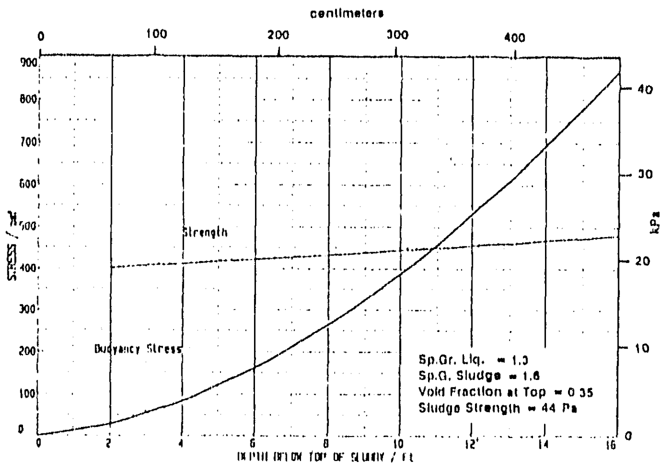

Fig. 6. Calculated Buoyancy Stress and Yield Strength of Nonconvective Layer (Sludge) in Waste Tank

a R.T. Allemann, L.L. Eyler, et al. Status Report on Conceptual Models for Waste Tank Mechanistic Analysis, January 1991, Pacific Northwest Laboratory, Richland, Washington (to be published).

a function of the void fraction, which varies with depth because of hydrostatic head. The buoyancy force of overlying material contributes to a tensile stress at all depths below. This stress has been obtained by integrating the Bousinesq[3] equation of stress contributions of each layer of sludge. Measured values of yield strength and specific gravities were used in the calculation. In the Figure 5 example, the crossover point of the two curves is the depth above which the material should pull away. This depth is a function of the properties assumed and the void fraction. For example, a void fraction of 0.3 would allow the material to rise from a depth of 4.5 m, which would be at the bottom of the waste tank. The actual shape of the parting line for this material is not yet calculated. If the departure from the power-law relation of the level history shown in Figure 3 is caused by the compression of gas remaining in the gob (the gas compresses, so the waste level falls), the volume of that gas can be estimated by assuming a pressure change. From the volume of gas released and the associated level drop it has been calculated that the gas released from the tank was retained at a pressure of about 2 atmospheres, which is about midway in the nonconvective layer. The level rate change beginning at the 6 to 10 day time period implies that 22 m^3 of gas were compressed. On the basis of liquid and slurry specific gravities of 1.3 and 1.6, respectively, the volume that can be supported by this gas is 213 m^3. If the gob were a hemisphere, initially it would have had a radius of 4.6 m, which is about equal to the sludge depth, and would therefore have parted at the bottom of the tank. This would explain the fact that the temperature at even the lowest thermocouple changed during the event.

Assuming that a gob of this size would spread out at the top of the bulk waste to a disk about 1-m thick, the rate of percolation of the remaining gas after the major release was calculated. In this calculation, a log-normal distribution of bubble size was assumed and the bubbles were assumed to rise at a terminal velocity dependent on a closed form drag coefficient solution of bubble motion through a non-Newtonian power-law fluid.[4] The power-law coefficient of $n = 0.75$, measured for the slurry, was used. The result of this calculation gives a rate of gas percolation release very close to a power law. This type of gas release can also be inferred from the power-law level decrease of Figure 3. With a choice of mean bubble size and geometric standard deviation for the percolating bubbles the time at which the gob would begin to sink could be matched; this analysis gave the rate of gas release shown in Figure 7. The mean bubble size required for the October 1990 event was 2.1 cm. It was interesting to note that the tank videos taken in August 1991 and December 1991 showed the existence of percolation and that the visible bubbles were estimated to be in this size range.

The December 1991 video showed a series of upwellings about 10 seconds apart following the initial major burst. These may be explained as satellite gobs

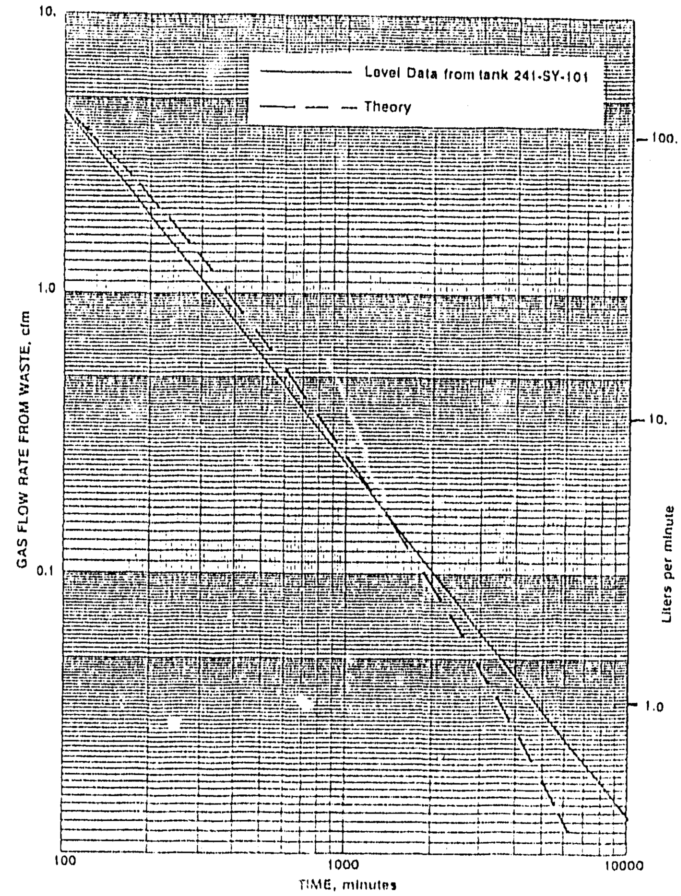

Fig. 7. Rate of Gas Release Subsequent to Main Release of October 24, 1990 Event

that are can peal upward once the cohesive material on one side has broken away with the main gob. The random nature of the gas release (and level drop) size is probably the result of the compounding of void fraction effects remaining from previous releases. Large void fractions can trigger a gob release from shallow depth; these would give the more numerous small level drops.

Contrasts with Other Theories

Crust Theory. Initial theories of the episodic release were based on the idea that a solid crust was trapping the gas below it and that the crust would periodically break. This theory did not explain the subsequently obtained data of the temperature inversion. Reynolds of Westinghouse Hanford Company suggested the idea of gas being trapped in the sludge below the crust and the concept of a "rollover". Later, the measured temperature inversion gave impetus to the concept of a rollover, that is, that the material at the bottom of the tank had risen to the top of the bulk as the upper region descended. The preferred mode of this rollover would be dependent on a form of Rayleigh number and might be toroidal, or in rolls covering the width of the cylindrical tank. The idea of a total rollover was not supported by the heat balance obtained from the thermal profiles. For a large event, initial heat recovery was 100%; however, in profiles from a few hours later, only a 70% recovery of initial heat was realized. This implied that the turnover was not uniform across the tank, or that only part of the waste was involved.

The numerical fluid dynamic simulations that have been used so far can calculate the general times for the rollover itself, but do not calculate the subsequent level changes and gas releases.

Particle Settling Theory. Another theory has the particles of the sludge being carried up by attachment and by viscosity with the bubbles. The gas release would then be accompanied by the particles settling back down. The reason that the bubbles release suddenly from the particles is not clear. In this case, temperature equalization by heat exchange would be extensive and the temperature profile would be smooth. (It is not.) Therefore, in this theory the rebuilding of the temperature profile at the bottom observed after a few days (Figure 5) would be caused by the slumping of sludge into the region vacated by the risen material. The particle settling theory implies a larger fraction (not measured) of gas release for an event than does the gob theory. The fraction has not been calculated for the particle settling theory.

In the gob theory, the cohesiveness of the sludge that is required to account for the retention of gas near the bottom of the bulk is not totally eliminated by the release of some of the gas. The presence of strength, or lack of strength, is important to the techniques of mixing the tank for mitigation or retrieval.

SUMMARY

The gob theory of episodic gas release from Hanford tank 241-SY-101 explains quantitatively 1) the phenomena of short-term and long-term liquid level drops in the tank, 2) temperature profile overturn and beginning of the profile reconstruction, and 3) percolation bubble size. Qualitatively the theory explains 1) the temperature balance during a release, 2) the association of a series of upwellings during a release, and 3) the varied release sizes. These results have shown the sensitivity of the projected phenomenon to the physical properties of the waste slurry. These results point out the need for specific data concerning the properties of the waste slurry to verify the theory and to show how mitigation methods would operate to interfere with the physical mechanisms of level growth and of episodic gas release. The theory may be extended to explore 1) the dynamics of the gob motion; 2) deformation of the sludge; and 3) the effects of variables such as sludge strength variations, void fraction, and previous history. New data will be obtained as the tank continues to be monitored. These data will serve to help prove, disprove, or refine the mechanistic theory.

Other tanks at Hanford have exhibited some of the level growth phenomenon, but the most prominent action has occurred in 241-SY-101. The instrumentation and observations that have been made on this tank have given insights to the phenomenon and these insights will find application to other tanks as well.

ACKNOWLEDGEMENT

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REFERENCES

1. D. M. STRACHAN, ed. Minutes of the Tank Waste Science Panel Meeting, February 7-8, 1991, PNL-7709, Pacific Northwest Laboratory, Richland, Washington (1991).

2. J. J. BARKER, T. M. BURKE, K. G. CAROTHERS, and D. A. REYNOLDS, Evaluation of October 24, 1990, Tank 241-SY-101 Gas Release Event, WHC-SD-WM-PE-041, Westinghouse Hanford Company, Richland, Washington (1991).

3. B. M. DAS, Advanced Soil Mechanics, Hemisphere Publishing Company, New York (1983).

4. A. ACHARYA, R. A. MASHELKAR, and J. ULBRECHT, "Mechanics of Bubble Motion and Deformation in Non-Newtonian Media," Chemical Engineering Science, 32, 863-872 (1977).

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