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Assessment of Gas Accumulation and Retention - Tank 241-SY-101

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Date Published
March 1993

Prepared for the U.S. Department of Energy
Environmental Restoration and
Waste Management

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P.O. Box 1970
Richland, Washington 99352

Hanford Operations and Engineering Contractor for the
U.S. Department of Energy under Contract DE-AC06-87RL10930

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Title ASSESSMENT OF GAS ACCUMULATION AND RETENTION - TANK 241-SY-101				Unclassified Category UC-	
New or novel (patentable) subject matter? ☒ No ☐ Yes If "Yes", has disclosure been submitted by WHC or other company? ☐ No ☐ Yes Disclosure No(s).				Information received from others in confidence, such as proprietary data, trade secrets, and/or inventions? ☒ No ☐ Yes (Identify)	
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Date(s) of Conference or Meeting		City/State		Will proceedings be published? ☐ Yes ☐ No Will material be handed out? ☐ Yes ☐ No	
Title of Journal					
CHECKLIST FOR SIGNATORIES					
Review Required per WHC-CM-3-4		Yes No		Reviewer - Signature Indicates Approval Name (printed) Signature Date	
Classification/Unclassified Controlled Nuclear Information		☐ Yes ☒ No		} SW BERGIN <i>[Signature]</i> 2/2/93	
Patent - General Counsel		☒ Yes ☐ No			
Legal - General Counsel		☒ Yes ☐ No			
Applied Technology/Export Controlled Information or International Program		☐ Yes ☒ No			
WHC Program/Project		☒ Yes ☐ No		G.D. JOHNSON <i>[Signature]</i> 1-1-93	
Communications		☒ Yes ☐ No		WP Whiting <i>[Signature]</i> 2-4-93	
RL Program/Project		☒ Yes ☐ No		R.E. Christensen <i>[Signature]</i> 2/4/93	
Publication Services		☒ Yes ☐ No		M.K. Oldfield <i>[Signature]</i> 3-24-93	
Other Program/Project		☐ Yes ☒ No			
Information conforms to all applicable requirements. The above information is certified to be correct.					
References Available to Intended Audience ☒ Yes ☐ No		INFORMATION RELEASE ADMINISTRATION APPROVAL STAMP Stamp is required before release. Release is contingent upon resolution of mandatory comments.			
Transmit to DOE-HQ/Office of Scientific and Technical Information ☒ Yes ☐ No
Author/Requestor (Printed/Signature) Date D. A. Reynolds <i>[Signature]</i> 1/28/93					
Intended Audience ☐ Internal ☐ Sponsor ☒ External					
Responsible Manager (Printed/Signature) Date N. W. Kirch <i>[Signature]</i> 1/28/93		Date Cancelled		Date Disapproved	

Document Title:

ASSESSMENT OF GAS ACCUMULATION AND RETENTION -
TANK 241-SY-101

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ASSESSMENT OF GAS ACCUMULATION AND RETENTION - TANK 241-SY-101

1.0 INTRODUCTION

1.1 OVERVIEW

An approximate analysis has been carried out to assess and estimate the maximum quantity of gas that is likely to be accumulated within waste tank 241-SY-101, and the maximum quantity which is likely to be retained after gas release events (GRE).

According to the phenomenological models used for this assessment, based on interpretation of current and recent operational data, the estimated gas generation rate in the tank is approximately $4 \text{ m}^3/\text{day}$ ($147 \text{ ft}^3/\text{day}$). About half of this gas is released as it is generated, which is (essentially) continuously. The remainder is accumulated within the slurry layer of settled solids at the bottom of the tank, and released episodically in GREs, known as "burps," that are induced by unstable buoyant conditions which develop when sufficient gas accumulates in the slurry.

Calculations based on gas volumes to cause neutral buoyancy in the slurry predict the following:

- The maximum gas accumulation (at 1 atm pressure) that can occur without triggering a GRE is in the range of 606 to $1,039 \text{ m}^3$ ($21,400$ to $36,700 \text{ ft}^3$)
- The maximum gas retention immediately after a GRE is equal to the maximum accumulation minus the gas released in the GRE.

GREs do not necessarily involve all of the slurry. In the largest GREs, which are assumed to involve all of the slurry, the minimum gas release (at 1 atm pressure) is calculated to be in the range of 193 to 328 m^3 ($6,800$ to $11,600 \text{ ft}^3$). The corresponding maximum gas retention would be 413 to 711 m^3 ($14,600$ to $25,100 \text{ ft}^3$).

1.2 BACKGROUND

Waste Tank 101-SY was first put into service in 1977, with the introduction of evaporator-concentrated waste known as "double shell slurry." Volumetric growth of this waste was noted almost immediately (Honeyman and Solbakken 1978). Complexant concentrate waste and additional double shell slurry were subsequently added. The tank reached its fully filled condition in 1980, with "slurry growth" continuing to be observed (Reynolds 1981). The tank settled into a pattern of steady volume increase, noted as surface level rise, and episodic gas release (Babad et al. 1992). The history of the surface level measurement is shown in Figure 1-1 (Burke et al. 1991).

Figure 1-1. Tank 241-SY-101 Surface Level Readings.

Tank 101-SY Surface Level Readings

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Concern for this behavior was heightened in 1989, when it was recognized that the gases generated and released included a mixture of hydrogen (a fuel) and nitrous oxide (an oxidizer) that was flammable and potentially explosive even if not mixed with air. This condition had not been considered in the applicable Safety Analysis Report (Koontz 1986).

The gas mixture, known as "slurry growth gas," consists primarily of hydrogen (H_2), nitrous-oxide (N_2O), and nitrogen (N_2), with smaller quantities of ammonia (NH_3) and water vapor (H_2O), plus traces of other gases. This mixture is produced by radiolysis and radiation-induced chemical reactions involving organic materials and inorganic salts in the wastes (Meisel et al. 1992). The energetics of the reaction, and the total chemical heat generation in the tank, are not well known relative to the ^{137}Cs and ^{90}Sr heat generation, which is estimated to be about 40,000 BTU/h, or about 12 kw (Reynolds 1992).

Direct sampling of the 101-SY contents has confirmed indirect evidence, from the vertical temperature profile in the tank, that the waste in 101-SY, between GREs, is located in several physical layers (Reynolds 1992), as illustrated in Figure 1-2. These layers are as follows:

- An upper layer, known as the "crust," which contains a large fraction of solids, but is obviously of low enough density to float, and which has thermal insulating properties relative to the liquid layer beneath
- A convective liquid layer, of nearly uniform temperature
- A thick, non-convective, layer of liquid and settled solid particles, known as the "slurry" layer. This layer may contain a sub-layer of denser and more viscous material, known as "sludge," at the bottom of the tank.

The episodic gas release behavior is consistent with the following phenomenology:

- a. Gases generated in the slurry are retained and accumulate in the slurry, causing a tank content volume increase and a reduction in the density of the slurry layer. The volume increase is apparent as a rising surface level of the tank contents.
- b. The density of the slurry, or parts of it, becomes less than the density of the overlying liquid.
- c. The unstable density relationship causes a "roll-over" of all (or a part) of the tank contents. The initial instability, triggering upward displacement of the slurry at some point, is augmented, once started, because of gas bubble expansion with the decreasing hydrostatic pressure at increasing elevations.

Figure 1-2. Distribution of Material in Tank 241-SY-101.

- d. The roll-over hydraulics carry liquid, gas, and solids from the slurry layer into the liquid layer and into or through the "crust." Bursts of slurry gas are released as slurry upwellings reach the surface.
- e. After the initial upwellings, gases continue to evolve by "effervescence" through the liquid and crust.
- f. Over a period of time, the heavier components of the slurry settle through the liquid, and the slurry layer is reformed. It appears that a large fraction of the slurry gas is retained in the slurry at this time.

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2.0 ANALYTICAL CONCEPTS/TANK 241-SY-101 CONTENTS

2.1 ANALYTICAL CONCEPT

In general, the accumulation (or storage) rate of gas in tank 241-SY-101 must equal the difference between the gas generation rate and the GRE as follows:

$$\dot{Q}(\vec{r}, t) = \dot{G}(\vec{r}, t) - \dot{R}(\vec{r}, t)$$

where

$\dot{Q}$ = gas quantity accumulation rate per unit volume

$\dot{G}$ = gas generation rate per unit volume

$\dot{R}$ = gas release rate, per unit volume

The parameters may, in concept, be variable in both space and time. The internal gas transport term has been omitted for simplicity.

For this analysis, the gas generation rate, $\dot{G}$, was assumed to be a constant in both space and time. The release rates in the crust and the convective liquid region were also assumed constant and equal to the generation rate, and the net transport between regions was assumed to be zero (that is, these regions were assumed to be at equilibrium). The release rate from the slurry layer was assumed to be zero, between GREs, and to be a complex function of hydraulic rollover phenomenology when GREs occur.

The volume integrated release rate for the tank is the sum of the release term integrals for the three regions, and the integrated gas accumulation is the sum of the accumulation term integrals for the three regions. For this analysis, no attempt is made to perform the space-time dependent gas release analysis. This analysis requires a three component, three phase, three dimensional numerical analysis. The present assessment is limited to evaluating equilibrium, or limiting conditions, in order to estimate the range of potential maximum gas accumulation before and after GREs.

2.2 TANK 241-SY-101 CONTENTS

The tank surface level and corresponding waste volume for this analysis were taken from conditions which existed shortly before the October 1990 gas release event (Barker et al. 1991). The non-convective slurry thickness at that time was estimated from the thermocouple (TC) data to be about 493 to 508 cm (194 to 200 in.). A similar estimate of 196 in. was made from December 1991 TC data (Erhart 1992). Accordingly, a slurry layer thickness of 196 in. was used to represent the thickness just before a "maximum" GRE. It was assumed that the slurry was at the point of neutral buoyancy at that time, and the hydrostatic pressure in the slurry was calculated on that basis.

The crust thickness was estimated, very approximately, on the basis of thermal data, video observations, and sampling information, in addition to the primary observation that "it floats" (Reynolds 1992). Temperature data show a consistent temperature drop between the convective liquid temperature and the tank dome air temperature of some 1.1 to 10 °C (30 to 50 °F). The TCs are four feet apart, placing an upper limit on the thickness. Thermal

conductivity estimates are consistent with a layer thickness of "several feet." Core sampling experience indicates a layer thickness of 61 to 102 cm (24 to 40 in.). Video observations indicate that the layer is discontinuous and highly fluid in behavior. A layer thickness of 0.76 m (2.5 ft), and an average density (specific gravity) of 1.35 were selected. The density corresponds to about 17 percent gas volume, assuming that the volume fractions of liquid and centrifuged solids equivalent are equal. This density value and the gas volume value are uncertain, but seem consistent with available data and observations.

The convective liquid layer thickness, 472 cm (186 in.), is the remainder. The liquid density used in the analysis is the "best" value, based on Window C measurements of supernatant liquid (Reynolds 1992).

Table 2-1 summarizes parameters relevant to component volumes and pressures in the tank.

Vertical temperature profiles before, during, and immediately after the October 1990 and December 1991 GREs are printed in Appendix A. Physical properties data obtained from "Window C" core samples of tank 101-SY are provided in Appendix B.

In this report specific gravity is generally used as density, without denoting the units. Unless noted otherwise, density, with no units given, is the specific gravity and is understood to be in the units of gm/mL, or kg/L.

Table 2-1. Tank 241-SY-101 Contents Data (Pre-GRE).

Layer	Thickness	Specific gravity	ΔP (atm)	P-ave. (atm)	Volume
"Crust"	76 cm (30 in.)	1.35	0.10	1.05	312 m ³ (11,000 ft ³)
Convective liquid	472 cm (186 in.)	1.46	0.67	1.44	1,926 m ³ (68,000 ft ³)
Non-convective slurry	498 cm (196 in.)	1.46*	0.70	2.12	2,039 m ³ (72,000 ft ³)
Total	1,046 cm (412 in.)	N/A	1.47	N/A	4,304 m ³ (152,000 ft ³)

*Assumed to be at the neutrally buoyant condition.
GRE = Gas release events.

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3.0 ANALYSIS

3.1 GAS GENERATION

The gas generation rate is inferred from tank 241-SY-101 operational data. Laboratory and theoretical data indicate that radiolysis, plus chemical reactions, can account for the gas quantities which appear to be produced (Meisel et al. 1992). However, at this time, these data do not account for the relative quantities of the gas constituents, and they are not sufficient to indicate quantitative differences between the various waste layers in the tank. Laboratory data show temperature sensitivity of relevant chemical reactions, as well as radiation sensitivity, but this laboratory data are not at this time correlated to operational data.

Operational data, as well, provide no basis on which to differentiate gas generation in the various waste layers, nor to distinguish time or temperature dependence. Accordingly, the analytical model used for the gas generation term is as follows:

$$\dot{G} = \text{constant, in both space and time}$$

The use of a constant value for the gas generation rate, without functional dependence on temperature, radiation level, or waste constituents or physical states, does not mean that such dependencies do not exist. Current operational, laboratory, and theoretical data do not establish values for such dependencies; however, if future operations or mitigation programs cause large changes in waste parameters, significant changes in gas generation rates could occur. For example, an increased temperature could cause an increased gas generation rate.

The reference value for $\dot{G}$ is calculated from recent operational data, (i.e., the time period 1989-1992). The calculation is dependent on assumptions regarding the gas retention term, which will be discussed later. The assumptions are as follows:

- Gas release from the convective liquid and crust layers equals gas generation, in the time period between GREs
- Gas release from the slurry is zero in these time periods.

The measured surface level record since January 1989 is shown in Figure 3-1. The average level growth rate measured by the FIC during the periods of consistent level increase is found graphically to be approximately 0.23 cm/day (0.09 in./day). Figure 3-2 shows an integration (by Reynolds) of the positive surface level growth, in which level drops of over 1.3 cm (0.5 in.) have been deleted.

Figure 3-3 shows a weekly average surface growth rate for nearly the entire 241-SY-101 operating period. Again, sudden surface level reductions were deleted. Neglecting the time period of about 2,300 to 3,000 days, influenced by "air lancing events," the growth is seen to have been about 0.25 to 0.28 cm/day (0.10 to 0.11 in./day) for most of the prior history, but appears to have declined to about 0.23 cm/day (0.09 in./day) currently.

Figure 3-1. Tank 241-SY-101 Surface Levels.

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Figure 3-2. Tank 241-SY-101 Surface Level Rise Rate.

Figure 3-3. Tank 241-SY-101 Growth Rate Since June 27, 1981.

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A surface level growth rate of 0.23 cm/day (0.09 in./day) equates to a waste volume increase rate of 0.94 m³/day (33.1 ft³/day). If all of this growth is due to gas accumulation in the slurry layer, at an average pressure of 2.12 atm, this corresponds to a gas accumulation rate, which equals the generation rate, of 2 m³ (70 ft³) (gas at 1 atm pressure).

NOTE: The temperature correction is neglected*.

The corresponding volumetric gas generation rate is as follows:

$$G = \frac{70}{72,000} \frac{\text{m}^3 \text{ (gas at 1 atm) / day}}{\text{m}^3 \text{ (waste)}}$$

$$= 0.97 \times 10^{-3} \frac{\text{m}^3 \text{ (gas @ 1 atm) / day}}{\text{m}^3 \text{ (waste)}}$$

The calculated total gas generation rate, then, is as follows:

$$(0.97 \times 10^{-3}) \times 4,300 \text{ m}^3 = 4.16 \text{ m}^3 \text{ (gas @ 1 atm) / day}$$

According to our model then:

- 2 m³/day (70 ft³/day) x 365 d = 725 m³ (25,600 ft³) = annual gas generation in the slurry released during GREs.
- 2.2 m³/day (77 ft³/day) = continuous release from the waste surface. (This gas would appear at a concentration of 89 ppm in a ventilation flow of 17 m³/min (600 ft³/min); if 36 percent of the gas is H₂, a H₂ concentration of 32 ppm is calculated.)

Reported estimates of GRE gas releases from April 1990 through August 1991 are a minimum of 765 m³ (27,000 ft³) to a maximum of 1,019 m³ (36,000 ft³) (Babad et al. 1992). Adding 255 m³ to 340 m³ (9,000 ft³ to 12,000 ft³) for the December 1991 event gives a 2-year total of 1,019 m³ to 1,359 m³ (36,000 ft³ to 48,000 ft³), or an annual GRE release of 510 m³ to 680 m³ (18,000 ft³ to 24,000 ft³), in reasonable agreement with the model estimate.

Recent H₂ concentrations in the 101-SY ventilation, measured by new "MOS" instrumentation, are typically in the range 25-35 ppm.

*Gas generated and accumulated in the slurry layer is at an average temperature of 49 to 54 °C (120 to 130 °F). Correction to the tank dome air space temperature would be a volume reduction of about 5 percent; correction to Standard Temperature and Pressure would be a reduction of about 15 percent.

3.2 GAS ACCUMULATION

Because the gas release rate in the crust and in the convective liquid is assumed to equal the gas generation rate, it is of interest to estimate the probable maximum equilibrium accumulation of gas in these regions.

This analysis also uses a model to estimate gas release from the slurry on the basis of postulating limiting conditions of gas accumulation; therefore, a gas accumulation estimate precedes a gas release estimate.

3.2.1 Gas Accumulation in the Crust

Observations regarding crust characteristics are described in Section 2.0. A nominal crust density of 1.35 ("it floats"), together with a rather arbitrary assignment of equal parts "centrifuged solids" equivalent ($\rho = 1.8$) and supernatant liquid ($\rho = 1.46$), results in a gas void volume fraction of 0.17. This gas volume is approximately 57 m³ (2,000 ft³), in situ and at 1 atm; the corresponding surface level increase is about 13 cm (5 in.).

3.2.2 Gas Accumulation in the Convective Liquid Layer

A typical method for calculating the gas inventory in a liquid with internal gas generation would be based on gas bubble size and bubble vertical rise time. Bubble sizes are not calculated, nor are bubble size measurements. However, the retained gas volume is estimated to be negligible, based on the rise time of an arbitrary small bubble. For example, the bubble terminal velocity is calculated from the Stokes equation:

$$V_{\infty} = \frac{1}{18} \cdot \frac{[D^2 \cdot g \cdot (\rho_f - \rho_g)]}{\mu} \quad (\text{Wallace 1969})$$

where

g = gravity
 D = bubble diameter
 ρ_f = liquid density
 ρ_g = gas density
 μ = liquid viscosity

For a 0.1 mm bubble, $D = 3 \times 10^{-4}$ ft. Using $\rho_f = 90$ lb/ft³,

$$\rho_g = 0, \text{ and } \mu = 30 \text{ cP} = 6 \times 10^{-4} \frac{\text{lb-sec}}{\text{ft}^2},$$

the velocity is $V_{\infty} = 8 \times 10^{-4}$ ft/sec = 65 ft/day (20 m/day)

The average travel distance through the convective layer is 236 cm = 2.4 m (93 in. = 7.75 ft). The average travel time is 0.12 days, and the corresponding gas hold-up is about 0.23 m³ (8 ft³) (which is negligible).

A more significant gas retention mechanism in the convective liquid may be the adherence of gas bubbles to solid particles. Observations have been reported where there are solids that do not seem to settle out over long periods. Measured solid fractions from core samples (Window C) indicated an average weight fraction of about 16 percent solids, and about 12 percent near the top of the layer (Reynolds 1992). This upper material, of 12 weight percent solids, is about 10 volume percent solids. Tingey reported 7 volume percent (9 weight percent) solids in the convective liquid. Assuming that the solids do not settle out because of adherent voids, an upper limit of gas accumulation in the convective liquid can be estimated. The volume fraction of gas voids to provide neutral buoyancy for 7-10 volume percent solids (the "solids" density in this case is the density of the "centrifuged solids" portion of the core sample, about 1.8) is about 2 volume percent. This corresponds to somewhat more than 28 m³ (1,000 ft³) at the average pressure of 1.44 atm, and roughly 57 m³ (2,000 ft³) gas at 1 atm. The equivalent surface level increase is about 7.6 cm (3 in.).

3.2.3 Gas Accumulation in the Slurry Layer

The slurry gas release function is physically complex. Detailed multi-component, multi-phase, three dimensional models are required to compute this behavior (Allemann et al. 1991). Uncertainties in physical properties are significant to the calculated results, and sensitivity studies are necessary to assess the importance of key uncertainties.

A simplified model for $\dot{R}_g$ is used in the following analysis, to estimate the maximum quantity of gas that could accumulate in the slurry, and the maximum amount which could be retained following a GRE.

The slurry is represented by its average properties and state conditions. It is assumed that the gas can accumulate in the slurry until the gas concentration is such that the mixture density equals the density of the convective liquid.

Non-uniform gas distribution is actually to be expected as potentially contributing to variations in individual GRE magnitudes (see, for example: Alleman 1992). The total gas quantity required to produce neutral buoyancy of the entire slurry layer is only slightly sensitive to such variations in gas distribution.

This model assumes that any cohesive stress tending to hold the slurry to the bottom of the tank is negligible. If there were a cohesive stress of 0.3 psi (typical of measured shear stress of slurry core samples at 50 °C), an additional positive buoyancy force of 86,638 kg (191,000 lb) would be required for roll-over of 100 percent of the slurry. This would correspond to an additional gas volume of about 57 m³ (2,000 ft³) in situ, about 113 m³ (4,000 ft³) at 1 atm (which is not included in the following assessment of potential gas accumulation).

The slurry is a three-component mixture. The gas concentration in the mixture that is required to produce neutral buoyancy is dependent on the concentrations and densities of the liquid and solid components. Neutral buoyancy is defined as that mixture density that equals the density of the supernatant fluid, (i.e., the convective liquid).

In general

$$\rho_m = \sum_i f_i \rho_i$$

where

$$\begin{aligned}\rho_m &= \text{mixture density} \\ \rho_i &= \text{component density of component "i"} \\ f_i &= \text{volume fraction of component "i"}\end{aligned}$$

Assuming that gas accumulation displaces the other components equally, the solid fraction and liquid fraction may be taken as mutually dependent, and the gas fraction for neutral buoyancy can be calculated as a function of either the solid or liquid fraction.

Define:

$$\begin{aligned}f_{s,o} &= \text{solid fraction at zero gas content} \\ f_{l,o} &= \text{liquid fraction at zero gas content}\end{aligned}$$

then, by definition:

$$f_{s,o} + f_{l,o} = 1$$

Defining:

$$\alpha = f_g = \text{gas (void) fraction}$$

then, the three-component mixture density is as follows:

$$\rho_m = [1 - \alpha][f_{s,o} \rho_s + (1 - f_{s,o}) \rho_l]$$

Figure 3-4 is the graph of α (gas void fraction) vs. $f_{s,o}$, the numerator of the solid/liquid ratio, $f_{s,o}/f_{l,o}$, for the density parameters:

$$\begin{aligned}\rho_m &= 1.46 \\ \rho_s &= 2.18 \\ \rho_l &= 1.46\end{aligned}$$

Figure 3-4. Neutrally Buoyant Void vs. Solid Packing:
Tank 241-SY-101 Slurry Layer.

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The liquid density value of 1.46 is the best value for the supernatant liquid based on "Window C" sample characterization data (Reynolds 1992). The solid density value (2.18) is the average of the densities of NaNO_3 (2.26) and NaNO_2 (2.1).

Substantial characterization has been done on material samples from 101-SY (Herting et al. 1992; Reynolds 1992). No direct measurements exist, however, for several of the key parameters for this analysis, particularly:

- The slurry solid/liquid ratio
- Gas void content
- Density of the solid component.

These parameters then must be assumed, inferred, or deduced from the available experimental data, observations, and the background body of knowledge.

The most direct interpretation of available data has been selected as our base case, and alternate interpretations have been applied for comparison.

3.2.3.1 Gas Accumulation in the Slurry Layer: Base Case. Centrifuging of tank 241-SY-101 core samples compacted the so-called "centrifuged solids" to an average density of 1.8. For the base case, this representative value is taken for the compaction of the slurry in tank 241-SY-101. Any gas content remaining after sample preparation and centrifuging is assumed to be negligible, either because the gas quantity is virtually zero or any retained gas is so tightly held that it would not be subject to episodic release.

Using the selected solids density of 2.18 g/mL, the solid and liquid fraction of the "centrifuged solids" are calculated as follows:

$$(f_{s,o} \times 2.18) + (1-f_{s,o})(1.46) = 1.8$$

$$f_{s,o} = \frac{1.8 - 1.46}{2.18 - 1.46} = \frac{0.34}{0.72} = 0.47$$

then

$$f_{l,o} = 0.53$$

The gas void content to bring this solid/liquid mixture to neutral buoyancy, α_b , is as follows:

$$\alpha_b = \frac{1.8 - 1.46}{1.8} = 0.19$$

The corresponding calculated neutrally buoyant gas accumulation in the slurry layer is

$$(0.19)(72,000) = 388 \text{ m}^3 (13,700 \text{ ft}^3) \text{ in situ}$$

This would produce a 0.94 m (37 in.) surface level increase.

Converting to atmospheric pressure (and neglecting temperature change):

$$\text{Buoyant gas content} = 321 \text{ m}^3 (29,000 \text{ ft}^3) \text{ (gas at 1 atm)}$$

This calculation did not make use of the measured density of bulk samples of the non-convective slurry; the "best" value from these measurements was 1.69 (Reynolds 1992). The difference between 1.8 and 1.69 is interpreted as a difference in solid/liquid ratio. Centrifuging of slurry layer samples compacted the "centrifuged solids" to an average of 75 percent of the sample volume. Since the true solids volume was unchanged, the volume fraction in the bulk sample was 0.75 times the volume fraction in the centrifuged solids:

$$(0.75)(0.47) = 0.35$$

The calculated bulk density with this solid fraction, with negligible void, is

$$\rho_m = (0.35)(2.18) + (0.65)(1.46) = 1.71 \text{ g/ml.}$$

which is close to the measured value, 1.69.

3.2.3.2 Gas Accumulation in the Slurry Layer: Alternate Case 1. The base case assumes negligible gas content in the "centrifuged solids," but there are no experimental data which prove this assumption. An estimate of the potential gas content that could exist if the solids could be more tightly compacted can be made by assuming that the solid/liquid components of the slurry can be compacted to the consistency of clay; the solid volume fraction of clay is reported as 0.65 (refer to Table 3-1).

The density of a void free mixture of 65 percent solids and 35 percent liquid is $(0.65)(2.18) + (0.35)(1.46) = 1.93$.

The gas void fraction to reduce the density to 1.8 would be 0.07. The gas void fraction for neutral buoyancy is 0.24. The corresponding neutral buoyancy gas quantity is $490 \text{ m}^3 (17,300 \text{ ft}^3)$, in situ, and $1,039 \text{ m}^3 (36,700 \text{ ft}^3)$ at 1 atm. This interpretation of the available information is considered to be conservative; that is, to be a probable over-estimate of potential gas accumulation.

3.2.3.3 Gas Accumulation in the Slurry Layer: Alternate Case 2. It may be that the slurry compaction, in situ, is not as great as the centrifuged compaction, and is better represented by the bulk core samples. If the samples are void-free, the solids fraction is about 0.35, (as calculated above). The corresponding buoyant void fraction is 0.14. The corresponding gas volume is $286 \text{ m}^3 (10,100 \text{ ft}^3)$, in situ, and $606 \text{ m}^3 (21,400 \text{ ft}^3)$ at 1 atm.

The base case is dependent on the assumption that gas content in the "centrifuged solids" is negligible, as well as the assumed solid particle density of 2.18. Experimental verification or rejection of these assumptions is desirable.

Table 3-1. Summary of Slurry Layer Gas Accumulation Calculations.

	Base case	Alternate case 1	Alternate case 2
Void-free slurry density g/mL	1.8	1.93	1.7
Solid/liquid volume ratio	.47/.53	.65/.35	.35/.65
Buoyant void fraction	0.19	0.24	0.14
Maximum gas, in situ	388 m ³ (13,700 ft ³)	490 m ³ (17,300 ft ³)	286 m ³ (10,100 ft ³)
Maximum gas, 1 atm	821 m ³ (29,000 ft ³)	1,039 m ³ (36,700 ft ³)	606 m ³ (21,400 ft ³)

NOTE: These values do not include any estimate or allowance for positive buoyancy, beyond neutral buoyancy, due to cohesive shear strength of the slurry.

3.2.4 Comparative Estimate of Gas Accumulation Volume

An alternate estimate of the total gas volume in tank 241-SY-101 can be made on the basis of a tank inventory balance. The measured tank level just prior to the October 1990 GRE, 1,046 cm (412 in.), represents a volume increase of approximately 255 m³ (9,000 ft³) over the initial fill volume equivalent to a level of 983 cm (387 in.). From this, the volume loss associated with water evaporation must be subtracted. The nominal estimate of water evaporation is 45 kg/day (100 lb/day), which gives approximately 255 m³ (9,000 ft³) evaporated in 15 years. However, the actual volume (and level) loss would be slightly less due to precipitation of dissolved solids. The actual volume (level) loss is estimated to be approximately 227 m³ (8,000 ft³) or 53 cm (21 in.). Approximately 7.6 cm (3 in.) of water was added to the tank during water lancing in the late 1980s (approximately 28 m³ [1,000 ft³]). Therefore, the total void volume in the tank just prior to the October gas release event is estimated by this method at 453 m³ (16,000 ft³) or 109 cm (43 in.). Assuming that approximately 85 m³ (3,000 ft³) or 20 cm (8 in.) of this is in the crust and convective liquid layers, approximately 368 m³ (13,000 ft³) or 89 cm (35 in.) are left in the non-convective liquid layer 793 m³ (28,000 ft³) when expanded to 1 atm). This is close to the base case value derived in Section 3.2.1, and is within the range of alternate cases considered. The gas volume calculated by this method is estimated to have an uncertainty of ± 50 percent (primarily due to uncertainty in the quantity of water evaporated).

3.3 GAS RELEASE

The model of steady state gas release from the crust and convective liquid has been described previously. The model for rollover of the tank

contents and release and/or retention of accumulated gas from the slurry is a complex one, requiring sophisticated multi-component, multi-phase, multi-dimensional analysis. The characteristics of rollovers are conceived as being sensitive to material properties and initial gas distributions. The latter, in turn, may be dependent on prior gas release histories. Such analyses are beyond the scope of this assessment.

Just as a simple neutral buoyancy concept is used in this assessment to estimate the maximum potential gas accumulation, a similar concept is used to calculate the minimum quantity of gas which must be released. The model is as follows:

- All or part of the slurry rises under buoyant forces, just when it passes neutral buoyancy, to float on top of the convective liquid layer.
- The gas in this rising slurry expands due to the reduced hydrostatic head.
- At a minimum, the excess gas is released from the slurry, until the slurry at its new location returns to just below neutral buoyancy.
- At this point, the slurry can begin to sink; if cohesive or attractive forces are sufficient to entrain the remaining gas, it would be re-compressed and retained in the re-established slurry. An indication that such a phenomenon may be present has been noted.

NOTE: This is not a prediction of this behavior model, although it does appear that the gas release in GREs is much less than 100 percent efficient. This is simply a limiting-case model that is consistent with the observation that the slurry layer does re-establish itself after GREs. If it did not, the tank would have come to a condition with much or all of the solids in a solid/liquid/gaseous foam floating on the convective liquid.

NOTE: This analysis does not account for dissolved gases or water vapor. It has been estimated that the dissolved gases would account for no more than 2-3 percent of the total release, and water vapor would contribute less than 3 percent in situ and less than 5 percent in vented gas.

Consider a GRE involving 100 percent of the slurry, occurring at the time of maximum gas accumulation. Assuming no mixing with the convective liquid, the slurry layer would have a new pressure of 1.10 atm at the top, and $1.10 + 0.70 = 1.80$ at the bottom, for an average of 1.45 atm. The original 388 m^3 ($13,700 \text{ ft}^3$) (base case) gas volume at 2.12 atm expands to 566 m^3 ($20,000 \text{ ft}^3$). However, 388 m^3 ($13,700 \text{ ft}^3$) is all that is required for neutral buoyancy; the excess 178 m^3 ($6,300 \text{ ft}^3$) (at 1.45 atm) must be released for the slurry material to sink back to the bottom of the tank. The in-tank volumes of 178 m^3 ($6,300 \text{ ft}^3$) released and 388 m^3 ($13,700 \text{ ft}^3$) retained correspond to a minimum release of 258 m^3 ($9,100 \text{ ft}^3$) (1 atm) and a maximum retention of 564 m^3 ($19,900 \text{ ft}^3$) (at 1 atm).

The results of this calculation for the three gas accumulation cases of Section 3.2.3 are given in Table 3-2 and 3-3. Similar calculations can be done for partial slurry rollover assumptions. The general conclusions are as follows:

- The minimum gas release (at 1 atm) equals the accumulated gas in the involved slurry (at 1 atm) times

$$\left[1 - \frac{\rho \text{ (floating)}}{\rho \text{ (@ bottom)}} \right];$$

for the average slurry pressure in this tank, this multiplier is

$$1 - \frac{1.45}{2.12} = 0.32$$

- The maximum gas retention after a GRE equals the maximum potential accumulation minus the actual GRE release.

A summary of the over-all assessment of potential gas inventories in tank 241-SY-101 is shown in Table 3-2.

Table 3-2. Minimum Gas Release and Maximum Retention.

Gas volume	Base case m ³ (ft ³)	Alternate case 1 m ³ (ft ³)	Alternate case 2 m ³ (ft ³)
Slurry at bottom			
in situ, m ³ (ft ³)	388 (13,700)	490 (17,300)	286 (10,100)
@ 1 atm m ³ (ft ³)	821 (29,000)	1,039 (36,700)	606 (21,400)
Floating slurry: original gas contents			
in tank m ³ (ft ³)	566 (20,000)	716 (25,300)	419 (14,800)
@ 1 atm	821 (29,000)	1,039 (36,700)	606 (21,400)
Floating slurry: neutrally buoyant			
in tank, m ³ (ft ³)	388 (13,700)	490 (17,300)	286 (10,100)
@ 1 atm	564 (19,900)	711 (25,100)	413 (14,600)
Minimum release			
@ original pressure	122 (4,300)	156 (5,500)	91 (3,200)
@ floating pressure	178 (6,300)	227 (8,000)	133 (4,700)
@ 1 atm	258 (9,100)	329 (1,600)	193 (6,800)
Maximum retention:			
@ original pressure	266 (9,400)	334 (11,800)	195 (6,900)
@ floating pressure	388 (13,700)	490 (17,300)	286 (10,100)
@ 1 atm	564 (19,900)	711 (25,100)	413 (14,600)

NOTE: These values do not include any estimate or allowance for net positive buoyancy to overcome slurry cohesive shear strength.

Table 3-3. Gas Inventory Summary.

Layer	m^3 (ft ³) In place		m^3 (ft ³) @ 1 Atm		Δ Surface cm (in.)	
	pre-GRE	post-GRE	pre-GRE	post-GRE	pre-GRE	post-GRE
"Crust"	57 (2,000)		57 (2,000)		13 (5)	
Convective liquid	28 (1,000)		<57 (<2,000)		8 (3)	
Slurry	286-195 (10,100 - 6,900)		606-413 (21,400 - 14,600)		69-48 (27 - 19)	
	388-266 (13,700 - 9,400)		821-564 (29,000 - 19,900)		94-66 (37 - 26)	
	490-334 (17,300 - 11,800)		1,039-711 (36,700 - 25,100)		119-81 (47 - 32)	
Total					89-69 (35 - 27)	
					114-86 (45 - 34)	
					140-102 (55 - 40)	

GRE = Gas release events

3.4 UNCERTAINTY ASSESSMENT

The range of uncertainty for this assessment is estimated as follows.

- Gas Generation Rate**--The absolute uncertainty is large (± 25 percent). The impact on the maximum gas accumulation estimate is small. The uncertainty is an important consideration relative to predicting the effects of mitigation concepts which involve significant changes in temperature, radiation levels, or chemical or physical characteristics of the waste.
- Gas Release**--This assessment estimates only a minimum gas release, and a corresponding maximum gas retention, for GREs. The uncertainty in the maximum gas release fraction is not bounded or assessed in this study. Detailed modeling and numerical analysis is required to predict gas release dynamics and to perform a systematic sensitivity study.
- Gas Accumulation**--Significant uncertainty in gas accumulation is associated with the density and packing fraction of the solid slurry components (± 25 percent). An uncertainty is associated with yield properties of the material and the corresponding potential for sustaining positive buoyancy forces (estimated as an uncertainty of about + 10 percent, -0). Overall uncertainty in maximum gas accumulation (± 30 percent).

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APPENDIX A

**GAS RELEASE EVENT VERTICAL TEMPERATURE PROFILES:
OCTOBER 1990 AND DECEMBER 1991**

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Figure A-1. Temperature Profiles Associated with the October 24, 1990 Vent Cycle.

29208019.7

Figure A-2. Thermocouple Temperature Profile Three-Dimensional Plot
Looking Forward in Time and Down in Depth; October 1990
Gas Release Event

Tank 101-SY October 24, 1990

79106024.3

Figure A-3. Thermocouple Temperature Comparison Before, During and After Gas Release Event - December 1991.

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APPENDIX B

PHYSICAL PROPERTIES OF "WINDOW C" CORE SAMPLES

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Table B-1.* Physical Properties of Samples from Tank 241-SY-101.

Property	32 °C	50 °C	65 °C	80 °C
Segment 19 nonconvective slurry				
Density (g/mL)--bulk	1.68	1.66	1.66	1.74
Density--centrifuged supernate	1.50	1.46	1.44	1.44
Density--centrifuged solids	1.75	1.75	1.73	2.03
Volume % settled solids	100%	100%	100%	100%
Volume % centrifuged solids	78%	72%	65%	62%
Wt % centrifuged solids	79%	76%	67%	72%
Shear strength (dynes/cm ²)	15,000	6,400	3,000	5,100
Wt % water, gravimetric	31.4%			
Segment 8 convective liquid				
Density (g/mL)--bulk	1.53	1.54	1.54	1.48
Density--centrifuged supernate	1.46	1.45	1.47	1.43
Density--centrifuged solids	1.93	1.85	1.82	2.16
Volume % settled solids	44%	48%	54%	56%
Volume % centrifuged solids	22%	23%	20%	8%
Wt % centrifuged solids	28%	27%	24%	10%
Shear strength (dynes/cm ²)	<530			
Wt % water, gravimetric	38.9%			
Segment 22 sludge				
Density (g/mL)--bulk	1.71	1.71	1.71	1.70
Density--centrifuged supernate	1.46	1.51	1.52	1.53
Density--centrifuged solids	1.78	1.82	1.79	1.79
Volume % settled solids	100%	100%	100%	99%
Volume % centrifuged solids	79%	73%	68%	64%
Wt. % centrifuged solids	83%	78%	72%	68%
Shear strength (dynes/cm ²)	116,000	26,400	4,000	14,000
Wt. % water, gravimetric	28.5%			

*Herting, D. L., D. B. Bechtold, B. A. Crawford, T. L. Welsh, and L. Jensen, 1992, *Laboratory Characterization of Samples Taken in May 1991 From Hanford Waste Tank 241-SY-101*, WHC-SD-WM-DTR-024, Rev. 0, Westinghouse Hanford Company, Richland, Washington.

Table B-2. Comparison of Pacific Northwest laboratory Physical Results with Process Control Laboratories Processing Data.

Property	Room temp. (PNL)	Room temp. (Westinghouse Hanford)	65 °C (PNL)	60 °C (Westinghouse Hanford)
Segment 19* and 22* (nonconvective layer)				
Density (g/mL)--bulk	1.70	1.64	1.68	
Density--centrifuged supernate	1.48		1.48	1.48
Density--centrifuged solids	1.76		1.76	1.70
Volume % centrifuged solids	78%		68%	76%
Wt % centrifuged solids	81%		70%	79%
Wt % water**	30.0%	32.5% (33.3)		
Segment 8 (convective layer)				
Density--bulk	1.53	1.55	1.54	
Density--centrifuged supernate	1.46		1.47	1.51
Density--centrifuged solids	1.93		1.83	1.68
Volume % centrifuged solids	22%		20%	18%
Wt % centrifuged solids	28%		24%	25%
Wt % water	38.9	36.3		

*Average of values obtained from composites 3, and 4, processed at Westinghouse Hanford and Segments 19 and 22 analyzed at PNL.

**Value for wt % water determined by Westinghouse Hanford Company was obtained from Segment 19 TGA and Segment 22 TGA determinations. The value shown in parentheses is the average result of two determinations made by Karl Fischer titration.

There is good agreement between the two measurements for most of the values shown. Densities reported for the centrifuged solids, centrifuged liquids and bulk samples are all within 5% for the average results obtained from Segment 19 and 22 and those of the processed composite samples. All density and weight percent water determinations differ by no more than 10%. The volume and weight percent centrifuged solids for Segment 19 and those obtained from composite 3 and 4 averages exhibit larger discrepancies. Generally, the volume and weight percent results are larger for centrifuged solids processed by Westinghouse Hanford. This discrepancy may be due to the temperature difference between methods used or from incomplete separation of liquids from solids in the processed composites.

Table B-3. Physical Properties of Convective Liquid Samples from Tank 241-SY-101.

Segment 4				
Property	32 °C	50 °C	65 °C	80 °C
Density (g/mL)--bulk	1.50	1.51	1.50	1.50
--centrifuged supernate	1.43	1.43	1.41	1.43
--centrifuged solids	2.1	1.9	1.9	1.9
Volume % settled solids	60%	60%	58%	51%
Volume % centrifuged solids	5%	7%	7%	7%
wt % centrifuged solids	10%	9%	9%	9%
Wt % water (gravimetric)	39.4%			
Segment 8				
Density (g/mL)--bulk	1.53	1.54	1.54	1.48
--centrifuged supernate	1.46	1.45	1.47	1.43
--centrifuged solids	1.93	1.85	1.82	2.16
Volume % settled solids	44%	48%	54%	56%
Volume % centrifuged solids	22%	23%	20%	8%
wt % centrifuged solids	28%	27%	24%	10%
Wt % water (gravimetric)	38.9%			
Segment 13				
Density (g/mL)--bulk	1.44	1.45	1.45	1.44
--centrifuged supernate	1.43	1.42	1.41	1.38
--centrifuged solids	2.1	2.1	2.0	2.2
Volume % settled solids	39%	37%	39%	40%
Volume % centrifuged solids	7%	7%	7%	6%
wt % centrifuged solids	11%	10%	9%	8%
Wt % water (gravimetric)	35.0%			

Table B-4. Physical Properties of Non-Convective Samples from Tank 241-SY-101.

Segment 15				
Property	32 °C	50 °C	65 °C	80 °C
Density (g/mL)--bulk	1.67	1.70	1.70	1.66
--centrifuged supernate	1.48	1.58	1.50	1.55
--centrifuged solids	1.74	1.75	1.76	1.79
Volume % settled solids	100%	100%	100%	98%
Volume % centrifuged solids	88%	80%	75%	69%
wt % centrifuged solids	88%	83%	78%	74%
Shear strength (dynes/cm ²)	47,500	7,300	21,000	5,300
Wt % water (gravimetric)	28.4%			
Segment 19				
Density (g/mL)--bulk	1.68	1.66	1.66	1.74
--centrifuged supernate	1.50	1.46	1.44	1.44
--centrifuged solids	1.75	1.75	1.73	1.90
Volume % settled solids	100%	100%	100%	100%
Volume % centrifuged solids	78%	72%	65%	62%
wt % centrifuged solids	79%	76%	67%	68%
Shear strength (dynes/cm ²)	15,000	6,400	3,000	5,100
Wt % water (gravimetric)	31.4%			

Table B-5. Physical Properties of Sludge Samples
from Tank 241-SY-101.

Segment 22				
Property	32 °C	50 °C	65 °C	80 °C
Density (g/mL)--bulk	1.71	1.71	1.71	1.70
--centrifuged supernate	1.46	1.51	1.52	1.53
--centrifuged solids	1.78	1.82	1.79	1.79
Volume % settled solids	100%	100%	100%	99%
Volume % centrifuged solids	79%	73%	68%	64%
wt % centrifuged solids	83%	78%	72%	68%
Shear strength (dynes/cm ²)	116,000	26,400	4,000	14,000
Wt % water (gravimetric)	28.5%			

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APPENDIX C

GAS RELEASE PHENOMENON: TANK 241-SY-101

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Westinghouse
Hanford Company

APPENDIX C

Internal
Memo

From: Environment, Safety, Health, and Quality Assurance
Phone: 6-0644 83-55
Date: December 31, 1991
Subject: 101-SY GAS RELEASE PHENOMENON

To: Rudy Allemann K7-15
cc: G. M. Christensen H4-23
W. L. Knecht H0-34
D. D. Stepnewski H5-32
DES-File/LB

Thanks for sending me the reports of your modelling analysis on the subject. As we discussed a couple of weeks ago, I have looked briefly into a one-dimensional model which assumes that gas is generated at a uniform rate in the liquid phase of the slurry, and is released whenever the slurry achieves a net positive buoyancy for the entire slurry layer above some level.

The model assumes the slurry bed has sufficient mechanical strength to act as a barrier, preventing the rise of lower level buoyant slurry through heavier over-lying layers. The slurry then behaves as follows:

1. Starting with a uniform gas content, the slurry grows lighter, by expulsion of liquid from the settled bed, and first becomes buoyant at the top surface of the slurry.
2. Release of gas from the top layer(s) permits the slurry bed to re-settle on top, and a period of time elapses until gas buildup lower in the bed produces enough buoyancy to balance the heavier slurry above.
3. The slurry layer above the neutral buoyancy point in the bed rises, releasing gas from all layers above the neutral buoyancy level.
4. The layers below the neutral buoyancy point are assumed to be unaffected by the gas release above, except that the releases are followed by resettling of the above solids, increasing the weight of solids which must be overcome by buoyant forces.
5. Eventually, the neutral buoyancy point works its way to the bottom; if the gas release at each slurry lift was assumed to be 100 percent, the process would simply repeat - but if the release is less than 100 percent, the system continues to remember its history, and subsequent releases are affected by the gas which remains in the slurry.

- slurry constituents:
 - 50% volume solids @ 120 lb/ft³ at top of slurry, increasing to 60% at bottom
 - liquid @ 90 lb/ft³
 - gas @ 1 lb/ft³ @ 1 atmosphere(gas accumulation displaces liquid, at no change in slurry volume).
- gas generation rate:
 - 0.002 $\frac{\text{ft}^3}{\text{ft}^3}$ (gas) @ 1 atm pressure
 (liquid)
- gas release on slurry rise event:
 - 50% of gas at all levels above neutral buoyancy
 - zero at lower levels
- model geometry
 - 10 ft. overlying liquid
 - 18 ft. of slurry (9 - 2 ft. layers modelled)

<u>Time</u>	<u>Time Interval</u>
135 days	N/A
197	62
304	107
472	168
533	61
657	124
788	131
866	78
999	133
1081	82

C-4

Rudy Allemann
Page 3
December 31, 1991

Figure 2 shows the average slurry density at and above the plotted layers at the time of each event. The gas release occurs when the equivalent density at any layer goes below 90 (lb/ft³), and 50% of the gas at and above that layer is released at that time. The bed then resettles and awaits enough gas buildup to trigger the next event. (I left the 50% gas at its original layers, rather than mix them up.)

Figure 3 shows the local slurry densities at the onset of each event.

The significant aspects of this model are:

- some mechanical strength of the settled bed is assumed
- viscosity of the fluid is neglected

Since the calculated behavior seems qualitatively consistent with observations, these may be reasonable. If so, there is a simple implication for remediation: fluidizing the slurry bed will eliminate the episodic gas releases. This is consistent with remediation concepts aimed at "mobilizing" the slurry, i.e., keeping the solid particles in suspension by some means of stirring the tank contents.

D. E. Simpson
Consultant

kb

Attachments 3

Figure C-1.

Figure C-2.

Figure C-3

APPENDIX D

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

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APPENDIX D

PNL-SA-20353

Feb. 1992

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 as 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.

Figure 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

A. 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.

Figure 2. Level History in Tank 241-SY-101²

Figure 3. Level Data for Several Days after the October 24, 1990 Gas Release

B. 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 to 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 (shown as "After event" in Figure 4) 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 few days the temperature at the bottom begins to rise again, while the top bulge disappears.

C. 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.

D. 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 (and even 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 Eyler³ 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.

² 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 (draft report).

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

Figure 5. Temperature Profiles Several Days After October 24, 1990 Event

A. 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.

B. 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 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 Boussinesq³ 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 6 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

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

nonconvective layer. The level rate change beginning at the 6 to 10 day time period implies that 22 m³ 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 m³. 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 that can peel 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.

C. 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. Reynolds⁵ 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

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

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 hypothesis has the particles of the sludge being carried up by the bubbles by attachment and by viscous drag. 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.) Also the material would no longer be warm upon reaching the bottom. 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 long-term gas release rate associated liquid level drops in the tank, and 2) percolation bubble size. Qualitatively the theory explains 1) temperature profile overturn and beginning of the profile reconstruction, 2) the temperature balance during a release, 3) the association of a series of upwellings during a release, and 4) the varied release sizes. These results have shown the sensitivity of the projected phenomenon to the physical properties of the waste slurry. Also, 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

This work was conducted by Pacific Northwest Laboratory for the U.S. Department of Energy under Contract DE-AC06-76RLO-2830 to support the High-Level Waste Tank Safety Program activities at Westinghouse Hanford Company. Pacific Northwest Laboratory is operated for the U.S. Department of Energy by Battelle Memorial Institute.

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