WM'00 Conference, February 27 – March 2, 2000, Tucson, AZ

UNDERSTANDING GAS RELEASE EVENTS IN HANFORD DOUBLE SHELL TANKS

Perry Meyer & Beric Wells
Pacific Northwest National Laboratory

ABSTRACT

Large episodic gas release events (GREs) in Hanford Double Shell Waste Tanks have been considered a potentially serious safety hazard since the phenomenon began to be studied in 1990. Prior to 1993, one tank occasionally released a sufficient volume of gas to make the tank headspace flammable. The GREs are believed to be caused by the mechanism of *buoyant displacement*. Questions persist as to why some tanks exhibit GREs and others do not. This paper attempts to answer to these questions by considering the physics of buoyant displacements. An overview of the simple yet insightful methods used to predict them are presented. The models give criteria that must be satisfied in order for buoyant displacements to occur. These models are derived from fluid stability theory, mechanics, energy arguments, and bubble transport theory. The models are validated by laboratory scale experiments and by comparison to data from observed gas release events from the waste tanks. Application of the models to some current and planned waste tank activities is presented. These include the current effort to remediate the unexpected level rise in Tank SY-101 by transfer and back dilutions. Additionally, the initial retrieval activities for Tank AN-105 will be discussed in light of buoyant displacement GRE potential.

INTRODUCTION

The flammable gas hazard in Hanford waste tanks first gained notoriety in the behavior of double-shell tank (DST) 241-SY-101 (SY-101). The waste level in the tank exhibited periodic rising and sudden drops that were attributed to gas release. Installation of gas monitoring equipment revealed that the gas release events (GREs) in SY-101 were in fact hazardous. On at least two occasions, the entire tank headspace concentration of hydrogen exceeded the lower flammability limit (LFL). Flammable Gas Watch List (FGWL) tanks were identified as having a serious potential for the release of high level waste due to uncontrolled increases in temperature or pressure from a flammable gas burn. In addition to SY-101, five other tanks on the FGWL (AN-103, AN-104, AN-105, AW-101, SY-103) were subsequently identified to exhibit the same type of episodic gas release as SY-101 but of much smaller magnitudes.

The GREs are believed to be caused by a buoyant displacement (BD) mechanism. The buoyant displacement gas release process is shown in Fig. 1. In a buoyant displacement, a portion or "gob," of the settled solid/liquid sludge-like matrix at tank bottom (referred to as the nonconvective layer, or NCL) accumulates gas until it becomes sufficiently buoyant to overcome its weight and the strength of the surrounding material restraining it. At that point it breaks away and rises through the supernatant liquid layer (referred to as the convective layer, or CL). The stored gas bubbles expand as the gob rises, failing the surrounding matrix so a portion of the gas can escape from the gob into the headspace. After releasing a portion of its gas, the gob is no longer buoyant and sinks back to the bottom of the tank. This gas release process has been identified as a buoyant displacement gas release event, or BD GRE. It is important to note that BD GREs can only occur in tanks with a NCL, where gas retention can occur.

The ability to explain and predict BD GREs was of extreme importance for the Hanford site. Simple physical models have been developed that give criterion which must be satisfied in order for BDs to occur. These models are derived from fluid stability theory, mechanics, energy arguments, and bubble transport theory. To the extent available, the models are validated by laboratory scale experiments and comparison to data on observed BD GREs. Applications of the models to current and planned waste tank activities will also be presented. These include the current effort to remediate the unexpected level rise in Tank SY-101 by transfer and back dilutions and the initial retrieval activities for Tank AN-105.

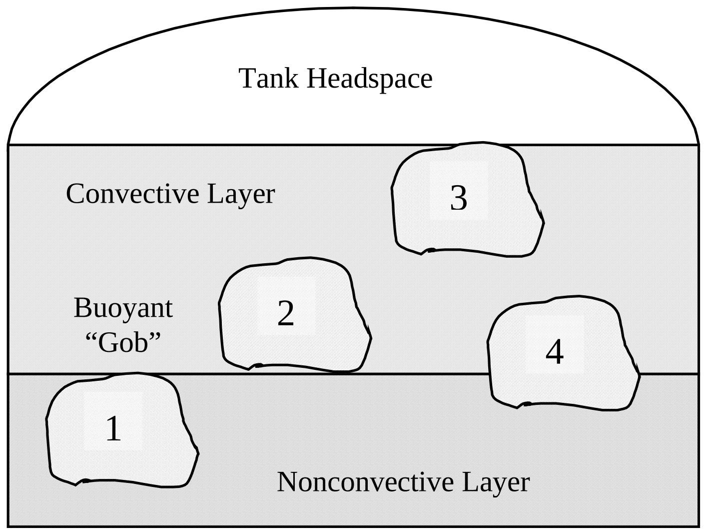

Figure 1. The buoyant displacement gas release process in Hanford Double Shell Waste Tanks: (1) Portion of NCL becomes buoyant. (2) Buoyant gob breaks free from NCL. (3) Rising gob expands, releasing some of its gas into tank headspace. (4) Partially degassed gob sinks back to bottom of tank.

DEVELOPMENT OF BD GRE MODELS

Models have been developed (1,2) that predict BD GREs based on tank waste conditions and configuration. These models provide a meaningful overall picture of BD GREs in DSTs. They demonstrate BD GRE behavior can be characterized by macroscopic waste properties and waste layer configurations. These include both the CL and NCL densities and depths, retained gas volume fraction (void fraction), NCL yield stress in shear, and gas generation rate. The basic theory of the BD GREs is built around the model of a gob of the NCL retaining enough gas to become buoyant, rising to the waste surface, breaking up, and releasing a fraction of its stored gas. The *buoyancy model* predicts whether buoyant conditions can occur, the *energy release model* predicts whether a buoyant displacement will be energetic enough to break up the gob and release its gas, and the gas *release model* predicts the volume of gas released during a BD.

Buoyancy Model

A simplified model has been developed from bubble transport theory that can predict how much gas can build up in the NCL in order to determine if the layer can become buoyant, thereby initiating a BD GRE (2). A void fraction profile develops within the NCL, resulting from a balance between internal gas generation and slow gas release at the top. Eventually a steady state is achieved. If the integrated average of the void fraction profile is less than the neutral buoyant void fraction (the void fraction required in the NCL making the average density equal to the density of the supernatant liquid), a BD cannot occur. On the other hand, if the integrated average of the void fraction profile becomes greater than the neutral buoyant void fraction, it is implied that a BD would have occurred at some point during the transient. Modeling the transient behavior of the void fraction profile within the NCL is complex. However, the steady state results are sufficient to indicate if a buoyant displacement is possible for a given waste configuration and properties.

The purpose of the model is to find the steady state void profile in the NCL, $\alpha(z)$, where z is the vertical coordinate. The model is described by gas mass and bubble number continuity along with equations for the bubble rise velocity and gas thermodynamic state. The gas mass continuity equation is

$$\frac{d(mu)}{dz} = G(z) \qquad \text{Eq. (1)}$$

where m is the number of moles of gas per unit volume (mol/m^3), u is the bubble velocity (m/day), and G is the volumetric gas generation rate (mol/m^3-day). The bubble number continuity equation is

$$\frac{d(nu)}{dz} = N(z) \qquad \text{Eq. (2)}$$

where n is the number of bubbles per unit volume (#/m^3) and N is the volumetric bubble nucleation rate (#/m^3-day). The ideal gas equation of state is expressed as

$$(m/n) = P(z)V_B(z)/RT(z) \qquad \text{Eq. (3)}$$

where P is the pressure (Pa), V_B is the average bubble volume, R is the gas constant (8314 J/kg-K), and T is the temperature (K). Since the release is assumed to occur by the very slow migration of bubbles, the bubble velocity, u, is computed by a modified version of Stokes law as follows:

$$u(z) = C\frac{\rho_{NCL} V_B^{2/3}}{\mu(z)} \qquad \text{Eq. (4)}$$

where ρ_{NCL} is the NCL density, μ is the viscosity, and C is an undetermined constant.

A solution can be found for Eqs. (1) - (4) under the assumptions of uniform nucleation rate and gas generation rates, and by neglecting spatial variation of pressure and temperature. Additionally, it is assumed that the waste viscosity (determining bubble rise velocity) increases linearly with waste depth from zero at the top of the NCL, and the rate of increase is the same for all tanks. This is a reasonable first approximation since compaction increases the yield strength of the NCL linearly (1). Under these assumptions, the void fraction profile in the NCL is

$$\alpha(\eta) = \frac{C}{\rho_{NCL}}\left(\frac{RT}{P_S}\right)^{\frac{1}{3}} h_{NCL}{}^2(1-\eta)\left[G\eta + \frac{\alpha_0\rho_{NCL}}{Ch_{NCL}{}^2}\left(\frac{P_S}{RT}\right)\right]^{\frac{1}{3}}\left[V_0 N\eta + \frac{\alpha_0\rho_{NCL}}{Ch_{NCL}{}^2}\right]^{\frac{2}{3}} \qquad \text{Eq. (5)}$$

where P_S is the average pressure, $\eta = z/h_{NCL}$, and V_0 is the initial bubble volume at z=0

There are two important limiting cases of Eq. (5). The first assumes a uniform nucleation rate and a zero initial void fraction. This simplification results in the following expression for the void profile:

$$\alpha(\eta) = \frac{C}{\rho_{NCL}}\left(\frac{GT}{P_S}\right)^{1/3} h_{NCL}^2\,\eta(1-\eta) \qquad \text{Eq. (6)}$$

where the nucleation rate is contained in the leading constant. The other limiting case is formed by setting the nucleation rate to zero, assuming all bubbles to enter at the lower boundary and grow due to gas generation as they rise. This case is expressed by

$$\alpha(\eta) = C\left(\frac{GT}{\rho_{NCL} P_S}\right)^{1/3} h_{NCL}^{2/3}\,\eta^{1/3}(1-\eta) \qquad \text{Eq. (7)}$$

where the initial void fraction is contained in the leading coefficient.

The void profiles given by Eqs. (6) and (7) can be integrated to find the average void fraction in the NCL. The ratio of the average void fraction to the neutral buoyancy void fraction will determine if a BD is possible. This ratio is referred to as the buoyancy ratio. The neutral buoyancy void fraction is given by

$$\alpha_{NB} = 1 - \frac{\rho_{CL}}{\rho_{NCL}}$$

The buoyancy ratios are then found to be

$$\frac{C}{\rho_{NCL} - \rho_{CL}} \left(\frac{GT}{P_S} \right)^{1/3} h_{NCL}^{2} \qquad \text{Eq. (9)}$$

$$\frac{C\rho_{NCL}^{2/3}}{\rho_{NCL} - \rho_{CL}} \left(\frac{GT}{P_S} \right)^{1/3} h_{NCL}^{2/3} \qquad \text{Eq. (10)}$$

The leading coefficients, C, in Eqs. (9) and (10) must be determined from tank data. Once they are specified, Eqs. (9) and (10) may be applied to any DST. If the ratio is less than unity, then the model suggests that a BD is not possible even in steady state. However, if the ratio exceeds unity, the model suggests a BD will have occurred at some point during the transient void build up in the NCL. The two limiting results given by Eqs. (9) and (10) represent two bounding limits of probable behavior in gas generating NCLs. The second case which assumes the nucleation rate is zero is probably more physical than the former because nucleation is suppressed by the presence of existing bubbles. Since it also provides somewhat more conservative predictions, Eq. (10) is chosen for application to the DSTs.

Energy Release Model

The gas release model predicts whether the NCL can become buoyant, initiating a BD. However, even if a BD occurs, it is not a given that the gas within the buoyant gob will be released into the tank dome space. There must be sufficient energy released during the process to fail the gas-bearing solid/liquid matrix. Basic energy conservation principles can be applied to the process of gas release during a BD to determine the conditions required for it to occur. The model is based upon the argument that the total amount of energy stored in a buoyant gob must exceed the energy required to yield the gob.

The initial gob volume possesses potential energy due to its buoyant state and the distance it can rise before reaching the waste surface. Not all of this energy will go into yielding the buoyant gob, as some will be dissipated in viscous motion. The stored buoyant energy can be calculated from the work done in raising the gob a distance h, given by

$$E_b = \int_0^h F(z)dz \qquad \text{Eq. (11)}$$

where F(z) is the net buoyant force on the volume and z is the vertical location of the centroid of the gob in the tank.

The energy required to yield the buoyant gob is estimated by assuming that the material has a finite yield stress that must be overcome to release gas bubbles. Regardless of material characteristics, the energy required to produce a specified strain in a material is equal to the work done on the volume by an externally applied stress to create that strain in the entire volume. It is postulated that, at a sufficiently high strain, the material fails or begins to act as a fluid such that it can no longer restrain gas bubbles from escaping. This strain energy is expressed mathematically by

$$E_y = \int_0^{\varepsilon_y} \tau d\varepsilon \qquad \text{Eq. (12)}$$

where τ is the stress applied to the volume, ε is the strain, and ε_y is the strain at failure.

The ratio between the buoyant energy given by Eq. (11) and the energy required to yield the gas-bearing gob participating in the BD given by Eq. (12) is

$$\frac{E_b}{E_y} = \frac{\alpha_o \rho_{CL} gh}{(1-\alpha_o)\varepsilon_y \tau_y}\left[\left(1+\frac{1}{\gamma}\right)\ln(1+\gamma) - k\right] \qquad \text{Eq. (13)}$$

where α_o is the initial void fraction of the waste, g is the acceleration due to gravity, and h is the distance from the center of the participating gob to the top of the liquid layer. The initial void fraction may be taken to be the critical void fraction defined by

$$\alpha_C = \alpha_{NB} + \frac{\beta \tau_y}{\rho_{NCL} g h_{NCL}} \qquad \text{Eq. (14)}$$

Eq. (14) expresses that an increase in gob void fraction is required in order to overcome the strength of the material which resists a BD. The second term in Eq. (14) is generally small, and is the appropriate first order correction to account for the effects of NCL strength on gob buoyancy. The waste material may yield in a combination of tension and shear. The von Mises yield condition implies that the yield stress in tension is $\sqrt{3}$ times the yield stress in pure shear. Therefore, the stress at yielding may be given by $\beta\tau_y$ where $1 \le \beta \le \sqrt{3}$. $\beta = 1$ will be used in this study as since this provides good correlation between measured void data and the calculated values for the DSTs (1). The parameter γ is determined from

$$\gamma = \frac{\rho_{CL} gh}{P_A} \qquad \text{Eq. (15)}$$

and the parameter k from

$$k = \frac{\alpha_{NB}(1-\alpha_o)}{\alpha_o(1-\alpha_{NB})} \qquad \text{Eq. (16)}$$

At tank scale, the parameter k is generally very close to 1. If the energy ratio given by Eq. (13) is large compared to 1, then a BD would be expected to release gas. On the other hand, if the ratio is much less than 1, little gas release would be expected.

Gas Release Model

The previous models, while describing whether a BD GRE can occur, give no information on the size of a BD GRE. The average size (and subsequent gas content) of an unstable gob undergoing buoyant displacement is believed to be controlled by two competing physical phenomena. The first relates to static forces and buoyancy. Because the NCL is a viscoplastic material, the concept of buoyancy is modified somewhat from the familiar meaning it has in Newtonian fluids. The buoyant forces resulting from normal stress distribution around a region of low density viscoplastic material must overcome both the weight of the region and the material strength on the boundaries of the region to achieve instability. The gas volume in the region required to generate sufficient buoyant force will depend on the size and shape of the gob as well as the yield stress of the material. The second phenomenon relates to dynamic instabilities. A viscoplastic material behaves like a fluid once the yield stress is surpassed. It is known that when a heavier fluid lies on top of a lighter fluid layer, regions of the heavier fluid will fall and similar volumes of the lighter fluid will rise according to the Rayleigh Taylor instability theory. The wavelength, or characteristic size of the moving regions, depends on the densities and viscosities of the two layers. If the NCL is viewed as a viscous fluid, the size of an unstable gob will depend on density (gas accumulation) as well as viscosity.

A solution for the volume of gas released from a BD has been developed from these principles in (1). The derivation is lengthy, so only the final result is presented here as

$$V_{rel} = C(P_s - 1)\alpha_{NB} h_{NCL} (\tau_y / \rho_{CL}) \qquad \text{Eq. (17)}$$

where C, the leading coefficient, is adjusted so the release volume matches the historical BD GRE data of the tank or group of tanks under consideration, τ_y is the average yield stress in shear, and ρ_{CL} is the CL density. The average pressure ratio of the gas in the NCL is given by

$$P_s = 1 + \frac{\rho_{CL} g}{P_A} \left(h_C + h_{CL} + \frac{h_{NCL}}{2} \right) \qquad \text{Eq. (18)}$$

where P_A is the atmospheric pressure. The thickness of the floating crust layer, convective layer, and nonconvective layer are denoted by h_C, h_{CL}, and h_{NCL} respectively.

To determine whether a gas release will result in a flammable condition in the head space, the lower flammability limit (LFL) fraction may computed from

$$\text{LFLfraction} = \frac{1}{0.04} \left(\frac{V_{rel}[H_2]}{V_h} \right) \qquad \text{Eq. (19)}$$

where $[H_2]$ is the average hydrogen concentration of the gas for a given tank and V_h is the head-space volume of the tank.

VALIDATION OF BD GRE MODELS

The buoyancy model and the gas release model are essentially correlations with leading constants that must be specified. This is done by adjusting the constants to give best comparison of the model results to historical BD GRE data for the Hanford tanks. The input parameters of the six DSTs under consideration are summarized in Table I.

Table I. Waste Properties for the DSTs on the Flammable Gas Watch List.

Property	**AN-103**	**AN-104**	**AN-105**	**AW-101**	**SY-101[(a)]**	**SY-103**
Densities (kg/m^3)						
Convective Layer	1530	1440	1400	1420	1500	1470
Nonconvective Layer	1730	1580	1580	1570	1700	1600
Layer Thickness (cm)						
Waste Level	884	979	1041	1040	1054	691
Convective Layer [(b)]	506	567	591	754	470	366
Nonconvective Layer	378	412	450	286	584	325
Nonconvective Layer Rheology						
Yield Stress (Pa)	130	95	125	150	116	126
Gas Generation Rate ($kmol/m^3$-day)						
Gas Generation Rate	3.4E-6	5.0E-6	2.4E-6	6.2E-6	3.2E-5	3.7E-6
Waste Temperature (K)						
Waste Temperature	314	317	313	414	323	308

(a) SY-101 data represent pre-mixer pump conditions.
(b) Convective layer thickness includes crust layer for the purposes of calculating energy and hydrostatic pressure.

The results of determining the buoyancy ratio via Eq. (10) for the six DSTs are tabulated in Table II. The leading constant is adjusted so that the minimum buoyancy ratio for the six burping DSTs is exactly 1.0. When this is done, Tank SY-101 has the largest buoyancy ratio, consistent with its observed high propensity towards BD GREs. When the same analyses are conducted on all the other Hanford DSTs, the buoyancy ratio is found to be less than 1 in every case. Hence, the buoyancy model accurately separates the tanks known to exhibit BD GRE from those that are known not to. These results give confidence that the model can be used to predict whether BD GREs can occur under new or changing tank conditions.

Table II. Buoyancy ratio calculated for the DSTs on the Flammable Gas Watch List.

	AN-103	**AN-104**	**AN-105**	**AW-101**	**SY-101**	**SY-103**
Buoyancy Ratio	1.0	1.61	1.03	1.23	2.77	1.42

The energy ratio given by Eq. (13) was evaluated for scaled laboratory BD experiments (3) and DST conditions in order to determine what value of the ratio is required for gas release during a BD. The experiments used a bentonite clay simulant with gas produced in situ by the decomposition of hydrogen peroxide. The model inputs and results are shown in Table III. In the first test, a displacement occurred, but very little gas was released because the individual gobs did not break apart and release their gas while rising to the surface. Eq. (13) gives an energy ratio of 4.13 for this case. The conditions of the second case were the same as those of the first, but the depth of the liquid layer was reduced. For this case, no active displacement or gas release was observed. The model predicts an energy ratio of 0.74. In the third and fourth cases, a weaker simulant was used. In case 3, with a predicted energy ratio of 7.93, both an energetic displacement and a gas release were observed. In the final case, the liquid layer was reduced, and again no active displacement was observed. A small amount of gas was released, but the mechanism appeared to be percolation rather than buoyant displacement. The energy ratio for the fourth case is 2.27.

Table III. The energy model applied to scaled buoyant displacement experiments.

Case	Solid Density (kg/m^3)	α_0	α_{NB}	k	Liquid Depth (m)	Solid Depth (m)	γ	τ_y (Pa)	E_b/E_y	Energetic	Gas Release
1	1087	0.25	0.08	0.261	0.105	0.047	0.011	67	4.13	Yes	No
2	1087	0.25	0.08	0.261	0.012	0.045	0.002	67	0.74	No	No
3	1070	0.15	0.065	0.397	0.101	0.048	0.0102	14	7.93	Yes	Yes
4	1070	0.20	0.065	0.280	0.011	0.048	0.0017	14	2.27	No	No

Table IV shows model results for the DSTs on the FGWL. AN-105 has the highest energy ratio, and SY-103 has the smallest. Since all of the tanks under consideration exhibit periodic GREs, the tank results imply that an energy ratio greater than 6.0 indicates that there is sufficient energy in a BD to release a large fraction of its gas. It may be noted that this value is lower than the energy ratio of case 3 above. Case 3, however, compared well visually with the large, very energetic BDs videotaped in SY-101, and is therefore considered to represent a more severe event.

Table IV. Energy ratio calculated for the DSTs on the Flammable Gas Watch List.

AN-103	AN-104	AN-105	AW-101	SY-101	SY-103
13	22	27	25	20	6

The results from tank data and scaled experiments are insufficient to quantify the relation between gas release and energy ratio precisely. However, they are consistent with the following criteria: No disruptive BD is predicted for $E_b/E_y<1$, BDs with limited gas release might occur for $E_b/E_y>\sim4$, and major gas releases can be expected if $E_b/E_y>\sim6$.

The release volumes determined from Eq. (17) are compared to the historical average release volumes and presented in Table V. The leading coefficient has been adjusted to minimize the result error. The model produces good agreement with the historical data for the six DSTs. The predicted values for GRE volume are within about ± 25% of the historical means except for SY-101, where the volume is under-predicted by about 50%, and AN-103, where the volume is grossly over-predicted. In AN-103, the two small gas releases observed in the tank data since 1995 may not be true BDs. The waste level history also shows no evidence of major gas releases of the size predicted. Therefore, the comparison with historical values may not be meaningful, and AN-103 was not included in the error minimization.

In SY-101, it is clear from in-tank video of several of the last GREs that several gobs of various sizes typically participate in GREs. This is probably due to the extremely high gas generation rate in SY-101, relative to the other five tanks. The gas generation rate in SY-101 can bring most of the waste to neutral buoyancy in several months, compared with several years in the other tanks. Hence, several gobs in the NCL can become buoyant at nearly the same time in SY-101. The comparison in Table V shows that release of two gobs would match historical results closely, but the uncertainty in both historical and predicted release volumes would allow up to six gobs.

To summarize, the models can predict BD GRE behavior closely approximating that seen in the DSTs. They can therefore be applied with some confidence to tanks whose waste configuration is altered as a result of transfers or other activities. The critical limits for the BD GRE models are listed in Table VI. If the buoyancy ration is less than one, a BD GRE is not possible. If the buoyancy ratio is greater than one and the energy ratio is greater than about 4, a BD will occur and it will release its gas. If the previous two limits are surpassed and the LFL fraction exceeds one, then a BD GRE will result in a flammable headspace.

Table V. Release volume model results compared to historical average release volumes.

Standard Volume (m^3)	AN-103	AN-104	AN-105	AW-101	SY-101	SY-103
Model (Eq. (1))	23	13	22	17	68	10
Historical Average	4	11	20	20	144	13

Table VI. Critical limits for BD GRE models.

Model	Critical Limit
BD can occur - Buoyancy Ratio	>1.0
BD will release gas – Energy Ratio	>4.0
BD creates flammable headspace - LFL Fraction	>1.0

APPLICATION OF BD GRE MODELS

To demonstrate the use of the models, they are applied to scenarios in Tank SY-101 and Tank AN-105. In SY-101, the current effort to remediate the unexpected waste level rise by transfer and back dilutions is evaluated. The initial retrieval activities scheduled for Tank AN-105 will also be discussed in light of BD GRE potential.

Tank SY-101 Transfer and Back Dilution

The installation of a mixer pump into SY-101 in 1993 successfully mitigated the problem of gas retention in the NCL in the tank (4,5,6). However, the accelerating waste level growth in 1998 and 1999 has made it necessary to implement new strategies to again mitigate and to remediate the gas retention problem to address flammability and physical limit issues. Plans are proceeding to remove 100,000 to 150,000 gallons of waste in November, 1999 and to subsequently back dilute the tank with water soon after. The intent is to repeat these processes until 350,000 gallons of waste has been removed and 350,000 gallons of water has been added.

Best estimates for the mass of saturated liquid and undissolved (or "free") solids is provided in Table VII. These values are based on volumetric measurements in the tank (1,7) and are necessarily subject to error. Comparison of pre-mixer pump conditions to those currently suggests that these mass estimates may be in error by as much as 7%. Due to the current activity of the mixer pump, the CL has approximately 14% free solids by volume. Waste will be removed from the tank by transferring material from this CL, thus reducing the inventory of liquid and solids.

Table VII. Initial mass of waste in SY-101.

Waste Component	Mass (kg)	Density (kg/m^3)
Liquid	4580000	1485
Free solids	1800000	2200

The subsequent addition of water to the tank will necessarily increase the liquid content of the tank and, due to dissolution, will decrease the amount of free solids in the tank. Experiments on tank waste have provided information on the solubility of SY-101 waste (8). A simplified dilution model has been developed (9) and is used to determine the mass inventory (i.e. mass of free solids and mass of liquid) in the tank after dilution.

To model the possibility of a BD GRE occurring in the tank, the final waste configuration (with the assumption that a steady state condition is reached; i.e. no mixer pump operation or other intrusive activities) is determined based on the final mass inventory. From the current layer volume and density measurements for the tank, the initial free solid and liquid masses are allocated to there respective layers. Waste transfer and back dilution, as discussed above, alter the final mass inventory, from which, based on assumptions of the final waste characteristics, the final waste configuration (layer volumes and densities) can be computed. These parameters are direct inputs into the BD GRE models.

All of the BD models suggest that thick convective and nonconvective layers increase the size of a BD GRE. Water introduced into SY-101 below the crust layer will be well mixed with the slurry of the CL, prior to coming into contact with the crust (10). Therefore, the free solids in the convective and NCLs will initially dissolve at a much greater rate than in the crust. One study concluded that the crust thickness should not be significantly reduced until approximately 150,000 gallons of water are added to the tank (11). Consequently, it assumed that the crust layer will be affected by the water addition only after the soluble solids below the crust layer have been completely dissolved. The insoluble solids of the crust layer are assumed to settle to the bottom of the tank in proportion to the amount of solids dissolved from the crust for a given dilution.

BD GRE model results for the steady state waste configuration after the removal of waste that is 14% free solids by volume followed by back dilution with water equal in volume to the waste removal are presented in Fig. 2. A sensitivity analysis of these results indicates that a 1% change in the model variables may result in a change as high as 8% in the results. Additionally, the possible 7% error in the solid and liquid mass changes the model results by as much as 50%.

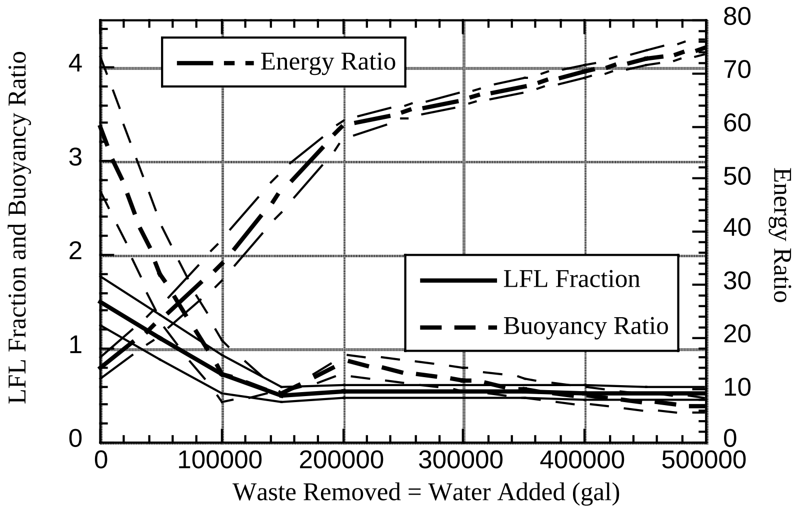

Figure 2. BD GRE model predictions as functions of the volume of waste removed and volume of water added (transfer and dilution volumes are assumed to be equal). The thin lines indicate the bounds for each result from the sensitivity analysis on the mass of the solids in the tank.

The results of the BD gas release event models applied to SY-101 indicate that, for the total anticipated 350,000 gallon transfer and back dilution, it will not be possible for a BD GRE to occur in Tank SY-101. The sensitivity of the BD GRE models to the tank waste characteristics and behavior must be considered when viewing these results.

Tank AN-105 Transfer

AN-105 is scheduled for retrieval for low level waste pretreatment and immobilization. It has been proposed that the retrieval process be initiated by decanting the supernatant liquid. Decanting is an effort to minimize the potential for a BD GRE during retrieval activities, such as the installation of a mixer pump. The possibility exists that the pressure reduction resulting from the decanting of the supernatant liquid will cause a BD GRE.

The gas release model given by Eq. (17) is based on the most probable gob size. In reality, a range of gob sizes is likely to exist. For this analysis, the initial void fraction and gob size are specified in order to examine the potential gas release for a range of initial conditions. The release volume is determined from the specific gob geometry and void fraction. The release volume will be a function solely of P_S for a supernatant removal scenario. The volume of gas released from a specific gob is given by

$$V_{rel}^{*} = f_{rel} V_{gas1} \qquad \text{Eq. (20)}$$

where

$$f_{rel} = 1 - \frac{\alpha_{NB}(1-\alpha_C)}{\alpha_C(1-\alpha_{NB})P_s} \qquad \text{Eq. (21)}$$

and

$$V_{gas1} = P_S h_{NCL} \frac{\pi}{4} D_o^{\,2} \alpha_{gob} \qquad \text{Eq. (22)}$$

where D_o is the in situ gob diameter and α_{gob} is the in situ void fraction of the gob.

In order for a BD GRE to occur, a gob must be able to become buoyant. It is estimated that there are approximately eight gobs with an average diameter of 7 m present in AN-105 at any given time (1). The gas generation rate of the tank is only sufficient to create buoyant conditions in any given gob in approximately 10 years. It is then reasonable to assume, given the historical release behavior of the tank, that there exists a uniform distribution of void fraction over the existing gobs. Depending on a gob's initial void fraction, it may be capable of becoming buoyant at some point during the supernatant liquid removal due to the pressure decrease.

A gob's minimum void fraction may be taken as that which it has immediately after it has risen up, released gas, and settled back to the bottom of the tank (settling is assumed to occur after enough gas has been released to render the gob neutrally buoyant at the waste surface). The minimum gob void is calculated to be 0.058. The maximum gob void fraction is necessarily the critical void fraction; 0.116. A decrease in the pressure at which the gas is retained at will cause expansion of that gas, resulting in an increased void fraction. Therefore, depending on the initial void fraction of the gob, the gob may become buoyant at some point during supernatant liquid removal. The minimum void fraction required for a gob to become buoyant solely due to supernatant liquid removal is approximately 0.078.

As the supernatant liquid is removed, the volume of the waste is decreased, so the head-space volume is increased. Therefore, for a given gob size, the resulting LFL fraction of the lower initial void gobs is decreased, Fig. 3. The energy ratio, when approximately 450 kgal of supernatant liquid has been removed, drops below the critical limit for the gob to release its gas. As a result, an initial gob void of at least 0.082 is required for a gob to become buoyant and still release its gas.

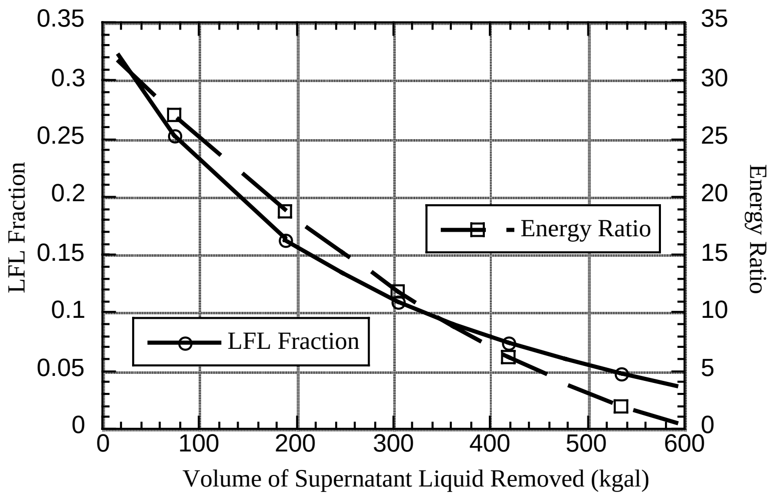

Figure 3: LFL fraction and energy ratio at the time of buoyancy as functions of the volume of supernatant liquid removed.

It follows that a gob with a sufficient initial void below the critical void may reach buoyancy due to the removal of supernatant liquid and then rise and release its gas. The possibility therefore exists that there may be a sequence of BD GREs occurring as the supernatant liquid is being removed. It is of interest then to analyze the head-space concentration of hydrogen as a function of the supernatant removal rate. Consider an even distribution of void fractions between the minimum and maximum gob voids for eight gobs each with a diameter of 7 m. Assuming a supernatant removal rate of 100 gpm, it can be determined, from the start of removal, the time at which a gob with a given initial void fraction will become buoyant. The time-evolving tank headspace hydrogen concentration with zero release rate is found from (12) to be

$$C_{H_2}(t) = C_{H_2}(0)e^{-\left(\frac{Q_V}{Vh}\right)t} \qquad \text{Eq. (23)}$$

where $C_{H2}(0)$ is the initial hydrogen concentration in the head-space and Q_V is the ventilation rate.

For the even distribution of void fraction over the eight gobs at the 100 gpm supernatant removal rate the accumulation of hydrogen in the headspace is not sufficient to reach the LFL. Three of the gobs do not participate due to their initial void being below 0.078. That is, they do not reach buoyancy due to the supernatant removal. For an uneven distribution of void over the gobs, that is if the second gob is at a higher initial void, it is possible that the subsequent release could "add" to the prior release, thereby increasing the LFL fraction. If there are sufficient gobs at a given initial void, say 0.108, a number of gobs at this initial void (five, for the specified void fraction) could in fact drive the hydrogen concentration in the headspace to the LFL. A sufficiently high supernatant removal rate (400 gpm) is capable of "adding" together the separate BD GREs to attain a larger LFL fraction. Conversely, a slow supernatant removal rate, or a sequential removal rate, will alleviate this possible occurrence.

Now consider that, not only is there a distribution of void fraction for the gobs, it is very likely there is also a size distribution for the gobs. The larger the gob, the larger the volume of gas in the gob at the critical void fraction. Therefore, with the smaller head-space volume during the early stages of supernatant transfer, large gobs with high initial void pose a threat to exceed the LFL. It is conservative to consider the situation where the largest gobs in a distribution have the highest initial void. In Figure 4, it is clear that the statements made above hold, and that the effects are exacerbated by the distribution of gob sizes. The initial release in Fig. 4 is representative of the largest historical releases for the tank. Also, the distribution of all BD GREs for the tanks shows the 95% confidence interval is approximately two times the average release, so a release of this magnitude may be expected.

It is expected that decanting supernatant liquid out of Tank AN-105 will cause BD gas release events to occur. While it is unlikely that a single release event will cause the tank head-space hydrogen concentration to exceed the lower flammability limit, the possibility of multiple releases causing an doing so does exist. However, it is evident that, by closely monitoring the gas release behavior in the tank during the supernatant liquid removal, the consequences of the release events can be controlled by slowing or temporarily halting the supernatant removal.

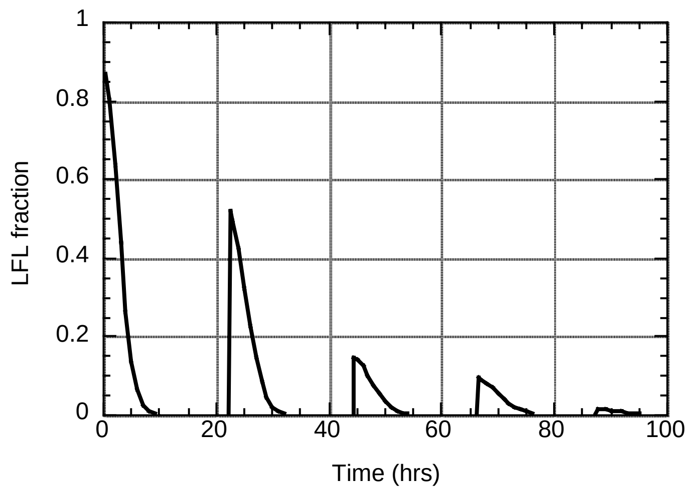

Figure 4: LFL fraction as a function of time (100 gpm supernatant removal rate, varied gob size, largest gobs contain highest initial void).

CONCLUSION

The ability to generally explain and predict buoyant displacement gas release events in the Hanford site double-shell waste tanks has been achieved. Simple models derived from fluid stability theory, mechanics, energy arguments, and transport theory have been developed that give criterion which must be satisfied in order for buoyant displacements to occur. The models have been validated by laboratory scale experiments and comparison with historical tank data on buoyant displacement gas release events. It is determined, through application of the models, that the planned transfer and back dilution scenario in Tank SY-101 will mitigate buoyant displacement gas release events and may be conducted without risk of a flammability hazard. Additionally, the models show that a potential supernatant liquid decant out of Tank AN-105 can be conducted in a safe manner.

REFERENCES

1. P. A. MEYER, M. E. BREWSTER, S. A. BRYAN, G. CHEN, L. R. PETERSON, C. W. STEWART, G. TERRONES, "Gas Retention and Release Behavior in Hanford Double-Shell Waste Tanks", PNNL-11536 Rev.1, Pacific Northwest National Laboratory (1997)

2. L. M. PEURRUNG, L. A. MAHONEY, C. W. STEWART, P. A. GAUGLITZ, L. R. PEDERSON, S. A. BRYAN, and C. L. SHEPARD, "Flammable Gas Issues in Double-Contained Receiver Tanks", PNNL-11836 Rev. 2, , Pacific Northwest National Laboratory (1998)
3. C. W. STEWART, M. E. BREWSTER, P. A. GAUGLITZ, L. A. MAHONEY, P. A. MEYER, K. P. RECKNAGLE, and H. C. REID, "Gas Retention and Release behavior in Hanford Single-Shell Waste Tanks", PNNL-11391, Pacific Northwest National Laboratory (1996)
4. R. T. ALLEMANN, Z. I. ANTONIAK, W. D. CHYALA, L. E. EFFERDING, J. R. FRILEY, W. B. GREGORY, J. D. HUDSON, J. J. IRWIN, N. W. KIRCH, T. E. MICHENER, F. E. PANISKO, C. W. STEWART, and B. M. WISE, "Mitigation of Tank 241-SY-101 by Pump Mixing: Results of Testing Phases A and B", PNL-9423, Pacific Northwest National Laboratory (1994)
5. C. W. STEWART, J. D. HUDSON, J. R. FRILEY, F. E. PANISKO, Z. I. ANTONIAK, J. J. IRWIN, J. G. FADEFF, L. E. EFFERDING, T. E. MICHENER, N. W. KIRCH, and D. A. REYNOLDS, "Mitigation of Tank 241-SY-101 by Pump Mixing: Results of Full-Scale Testing", PNL-9959, Pacific Northwest National Laboratory (1994)
6. M. E. BREWSTER, N. B. GALLAGHER, J. D. HUDSON, and C. W. STEWART, "The Behavior, Quantity, and Location of Undissolved Gas in Tank 241-SY-101", PNL-10681, Pacific Northwest National Laboratory (1995)
7. D. A. REYNOLDS, "Tank 101-SY Window E Core Sample: Interpretation of Results," WHC-EP-0628, Westinghouse Hanford Co. (1993)
8. J. C. PERSON, "Dilution Studies of Tank 241-SY-101 Waste. Preliminary Results", Memo 82100-99-015 to N. W. Kirch, April 22, 1999, Numatec Hanford Corporation, Richland, WA.
9. S. D. RASSAT, C. W. STEWART, B. E. Wells, et. al. "Dynamics of Crust Dissolution and Gas Release in Tank 241-SY-101", PNNL-13112, Pacific Northwest National Laboratory (2000)
10. Z. I. ANTONIAK, and P. A. MEYER, "Potential for Waste Stratification from Back Dilution in Tank 241-SY-101", PNL-13038, Pacific Northwest National Laboratory (1999)
11. W. L. KUBIC, and A. V. BELOOUSOV, "Dilution to Eliminate and Prevent Crust Formation in Tank 241-SY-101", LA-UR-99-3770, Los Alamos National Laboratory (1999)
12. H. L. SULLIVAN, "A Safety Assessment for Proposed Pump Mixing Operations to Mitigate Episodic Gas Releases in Tank 241-SY-101: Hanford Site, Richland, Washington", LA-UR-92-3196 Rev. 14, Los Alamos National Laboratory (1995)