PNNL-13337 Add. 1

Pacific Northwest National Laboratory
Operated by Battelle for the U.S. Department of Energy

Derivation of the Buoyancy Ratio Equation from the Bubble Migration Model

Addendum to PNNL-13337
Preventing Buoyant Displacement Gas Release Events in Hanford Double-Shell Waste Tanks

C.W. Stewart
P.A Meyer

April 2005

Prepared for the U.S. Department of Energy
under Contract DE-AC05-76RL01830

PACIFIC NORTHWEST NATIONAL LABORATORY
operated by
BATTELLE
for the
UNITED STATES DEPARTMENT OF ENERGY
under Contract DE-AC05-76RL01830

Printed in the United States of America

Available to DOE and DOE contractors from the
Office of Scientific and Technical Information,
P.O. Box 62, Oak Ridge, TN 37831-0062;
ph: (865) 576-8401
fax: (865) 576-5728
email: reports@adonis.osti.gov

Available to the public from the National Technical Information Service,
U.S. Department of Commerce, 5285 Port Royal Rd., Springfield, VA 22161
ph: (800) 553-6847
fax: (703) 605-6900
email: orders@ntis.fedworld.gov
online ordering: http://www.ntis.gov/ordering.htm

This document was printed on recycled paper.
(9/2003)

PNNL-13337 Add. 1

Derivation of the Buoyancy Ratio Equation from the Bubble Migration Model

Addendum to PNNL-13337,
Preventing Buoyant Displacement Gas Release Events in Hanford Double-Shell Waste Tanks

CW Stewart
PA Meyer

April 2005

Prepared for the U.S. Department of Energy
under Contract DE-AC05-76RL01830

Pacific Northwest National Laboratory
Richland, WA 99352

Contents

Figures

1.0 Buoyant Displacement Gas Releases

Some Hanford radioactive waste storage tanks with deep layers of sediment and supernatant liquid are subject to a gas release phenomenon that has been called *buoyant displacement*. The theory of buoyant displacement describes the behavior of a sediment permeated with discrete gas bubbles that displace the surrounding solid particles. In a buoyant displacement, bubbles form and grow, imparting their buoyant force to the surrounding sediment. When a portion, or "gob," of the sediment becomes sufficiently buoyant to overcome its weight and the strength of the surrounding material restraining it, the gob breaks away and rises through the liquid layer above it. The trapped gas bubbles expand as the gob rises, failing the surrounding material and allowing on the order of half of the bubbles to escape, depending on the waste properties and waste layer depths. The gob releases gas until it is no longer buoyant and sinks back to the sediment layer (Meyer et al. 1997).[a] This process is illustrated in Figure 1. The gob illustrations are staggered for clarity only; the actual process is believed to occur more or less vertically.

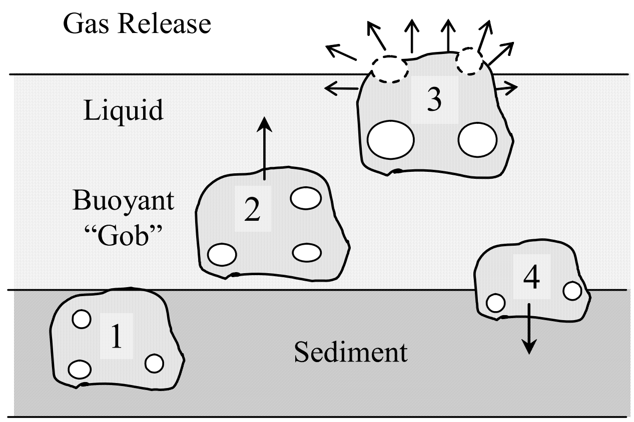

Figure 1. The Buoyant Displacement Process: 1) portion or gob of sediment becomes buoyant; 2) buoyant gob breaks out of sediment layer; 3) rising gob expands, releasing some of its gas; 4) non-buoyant gob sinks back to sediment layer.

The retained gas volume required for buoyancy is determined by the ratio of the bulk gasless densities of the liquid and sediment layers. The balance of weight and buoyant force at neutral buoyancy for a volume of gasless sediment, V_S, containing gas volume, V_G, can be expressed by

$$g\rho_S V_S = g\rho_L \left(V_S + V_G\right) \quad (1)$$

(a) The buoyant displacement mechanism was defined through study of the large gas releases from Tank 241-SY-101 from the mid-1980s through early 1990s. Leila Sasaki, Nick Kirch, and Dan Reynolds of Westinghouse Hanford Company, Rudy Allemann of PNNL, and Steve Eisenhawer of Los Alamos National Laboratory, among others, contributed to its development. The "gob theory" was first documented in a paper by Rudy Allemann for the April 15, 1992, Waste Management Conference in Las Vegas entitled, *Physical Mechanisms Contributing to the Episodic Gas Release from Hanford Tank 241-SY-101.*

where g is the acceleration of gravity (m/s^2), and ρ_L and ρ_S are the density of the liquid and the bulk gasless density of the sediment layer, respectively (kg/m^3). The gas volume fraction in the sediment layer is defined as

$$\alpha_S = \frac{V_G}{V_G + V_S} \tag{2}$$

Substituting Eq. (2) into Eq. (1) and solving for the gas volume fraction provides the neutral buoyancy volume fraction, α_{NB}:

$$\alpha_{NB} = 1 - \frac{\rho_L}{\rho_S} \tag{3}$$

In systems where the solids are soluble in the liquid, the densities of the two layers tend to be similar and Eq. (3) gives a relatively small gas volume required for buoyancy. Neglecting the strength of the sediment layer, when the average gas volume fraction in the sediment exceeds the neutral buoyancy gas fraction, a buoyant displacement will occur.

Gas generation and accumulation in the sediment layer is not uniform. Individual portions or "gobs" of sediment are believed to accumulate gas at varying rates and undergo buoyant displacements with sufficient independence that eruption timing becomes almost random. The effect is similar to popcorn with each grain heating up and popping independent of the rest. This effect is consistent with observations in waste tanks. In-tank video observations during the largest gas releases showed a series of local eruptions from different areas of the tank. Waste temperature profiles, measured by thermocouples spaced along a pipe hanging vertically in the waste, seldom change during smaller eruptions. This indicates that the gobs have a length scale smaller than the tank diameter such that a single eruption rarely disturbs the sediment around a thermocouple tree.

Typical gas release volume estimates indicate that only 10- 15% of the sediment layer achieves buoyancy in any one event, implying the presence of 6-10 gobs. The observed frequency of gas releases can only be explained as the result of a number of independently developing gobs. The typical interval between gas releases is less than one year. Based on the observed gas retention rate, a gob should require six to ten years to gain buoyancy. If there are eight to ten randomly evolving gobs, the release frequency is consistent with the retention rate.

Gobs are not absolutely independent, however. A gob disintegrates to a large extent as it releases its gas. The disintegrating gobs liberate a cloud of sediment particles that effectively increase the density of the liquid layer and reduce the neutral buoyancy gas fraction via Eq. (3). Other gobs that were not buoyant with respect to clear liquid may erupt in the heavier suspension of settling sediment particles from the first one. This effect is believed to have produced the large gas releases observed in Tank SY-101 as a series of several sequential eruptions a few minutes apart. Multiple eruptions have also been identified in the other tanks (Wells et al. 2002).

2.0 Theory of Gas Accumulation

Most Hanford waste tanks do not have buoyant displacements, even those with deep submerged sediment layers and moderate gas generation rates. What is unique about the few that do? Most tanks apparently achieve a steady state where gas generation is balanced by a steady background release so they never attain the neutral buoyancy gas fraction given by Eq. (3). In a few tanks, however, gas generation exceeds the background release, which allows gas to accumulate to the point of buoyancy.

The mechanism for this background release is unknown, but it must exist or all tanks would have buoyant displacements. We postulate that the background release is a slow migration of bubbles through the sediment, as will be discussed in Section 3.[a] In the meantime, to show the overall view and explain the phenomenon, we assume a flux of gas occurs from the waste surface and apply the ideal gas law to mass conservation on the gas in the sediment layer assuming a constant density. The result is

$$V_S \frac{d\alpha}{dt} = V_S G \frac{RT}{p} - A_S J_\alpha \tag{4}$$

where G is the average gas generation rate (moles/m^3-s), p and T are the average sediment pressure (Pa) and temperature (K), respectively, R is the gas constant (8314 J/mole-K), and J_α is the average gas volume flux across the sediment layer surface (m^3/m^2-s).

The average pressure, p, exerted on retained gas bubbles in the sediment layer is the hydrostatic pressure computed at the layer midpoint. Assuming that the solid particles are self-supporting and bubbles displace only liquid, this can be expressed as follows:

$$p = p_A + \rho_L g\left(H_T - \frac{H_S}{2}\right) \tag{4x}$$

where H_S is the height of the sediment layer (m), p_A is the ambient tank headspace pressure (Pa), and H_T is the total waste depth (m).

Dividing both sides of Eq. (4) by the sediment volume, noting that the volume is equal to the product of the height and area, gives

$$\frac{d\alpha}{dt} = G\frac{RT}{p} - \frac{J_\alpha}{H_S} \tag{5}$$

(a) This concept and initial theory were introduced by Dr. Kemal Pasamehmetoglu (Los Alamos National Laboratory) at a meeting of the Westinghouse Hanford Company Flammable Gas Project on February 27, 1997. The bubble migration model was derived from this theory by Perry Meyer (PNNL) over the next two years.

Eq. (5) illustrates the simple balance between source and outflow that determines whether the gas volume fraction in the sediment increases. If the volumetric gas generation exceeds the rate at which gas leaves the sediment layer, gas must accumulate.

The exact mechanism by which gas migrates through the sediment is not known but, since gas is observed in waste samples as bubbles, it can be explained by slow movement of bubbles. With this model, the gas volume flux can be expressed as the product of the number density, size, and velocity of bubbles crossing the upper surface of the layer:

$$J_\alpha = n v_B u_B \tag{6}$$

where n, is the bubble number density (1/m^3), v_B is the average bubble volume (m^3), and u_B is the velocity (m/s) of the bubbles as they exit at the top of the sediment layer. Because the gas volume fraction is defined by the bubble number density and volume, Eq. (6) can also be written as

$$J_\alpha = \alpha u_B \tag{7}$$

Therefore the rate that gas leaves the sediment layer is directly proportional to its volume fraction. Substituting Eq. (7) into Eq. (5) gives

$$\frac{d\alpha}{dt} + \alpha \frac{u_B}{H_S} = G\frac{RT}{p} \tag{8}$$

For the initial condition, $\alpha = 0$ at $t = 0$, the solution to Eq. (8) is

$$\alpha(t) = \frac{RT}{p}\frac{GH_S}{u_B}\left(1 - e^{-\frac{u_B}{H_S}t}\right) \tag{9}$$

Eq. (9) qualitatively describes the gas accumulation trend typically observed in tanks that do not have buoyant displacements. The gas volume fraction asymptotically approaches a constant steady state. If the steady state value is greater than the neutral buoyancy gas fraction, a buoyant displacement will occur before the tank arrives at its steady state. Therefore, if the bubble rise velocity and gas generation rate were known, the potential for a buoyant displacement could be evaluated.

3.0 Bubble Migration Model

The bubble migration velocity should be controlled by a balance between the bubble buoyant force and some retarding force. It is reasonable to express these relationships with the scaling (but not the magnitude) of Stokes law for creeping flow:

$$u_B(z) = S\frac{\rho_S g v_B(z)^{2/3}}{\mu(z)} \tag{10}$$

where S is a proportionality constant[a] and μ is an effective bulk viscosity (kg/m-s) that describes the retarding force on the bubble including the important effects of the unknown bubble migration mechanism. The bubble length scale is expressed as the cube root of the volume.

Stokes law strictly applies only for creeping motion (Re < 1) in Newtonian fluids. We assume that, for vanishingly small shear rates, the non-Newtonian waste with a finite yield stress in a waste tank sediment layer behaves qualitatively as a fluid. However, even accepting this extrapolation, the proper viscosity to apply is unknown because all viscosity measurements are performed at finite shear rates. Therefore, we use the yield stress, τ_y (Pa), as determined from in situ measurements using the ball rheometer (Meyer and Wells 2000) as an proportional indicator for the "correct" viscosity as follows:

$$\mu(z) = \frac{1}{K}\tau_y(z) \tag{11}$$

where K is an unknown constant shear rate (1/s). Based on the viscosity determined from ball rheometer data at a ball speed of 0.1 cm/s, K ≈ 1/9,000 (1/s). Equation (11) can be justified as the low shear rate approximation to apparent viscosity μ for a Hershel-Bulkley or Bingham fluid, or as acknowledging that shear strength and consistency probably vary together for some classes of non-Newtonian fluids, such as Hanford tank wastes.

The ball rheometer data also show that the yield stress increases approximately linearly with depth from zero at the top of the sediment layer. The linear variation of the yield stress with elevation can be expressed as

$$\tau_y(z) = m_\tau\left(H_S - z\right) \tag{12}$$

where m_τ is the rate at which the shear stress increases with depth (Pa/m). Combining Eqs. (10), (11), and (12) expresses the bubble velocity as

(a) For true Stokes flow, $S = \frac{2}{9}\left(\frac{3}{4\pi}\right)^{2/3}$.

$$u_B(z) = \frac{SK}{m_\tau}\frac{\rho_S g v_B(z)^{2/3}}{\left(H_S - z\right)} \tag{13}$$

We will later use tank data to establish the proper value for the combined coefficients S, K, and m_τ.

To apply the bubble rise scaling expressed by Eq. (13), we generalize the sediment-average gas accumulation equation of Eq. (8) to allow variation in the vertical coordinate, z. This one-dimensional model uses gas mass and bubble number conservation along with equations for the bubble velocity and the gas equation of state to derive an expression for the gas volume fraction as a function of z.

The gas mass conservation equation is

$$\frac{d(mu)}{dz} = G(z) \tag{14}$$

where m is the number of moles of gas per unit total waste volume (mol/m^3), u is the bubble velocity (m/s), and G is the volumetric gas generation rate (mol/m^3-s). The solution to Eq. (14) is given by

$$[mu](z) = [mu]_0 + \int_0^z G(z)dz \tag{15}$$

where $[mu]_0$ is the gas flux (mol/m^2-s) at the boundary z = 0 at the tank bottom, normally assumed to be zero except possibly for a leaking tank. Assuming the generation rate is uniform, Eq. (15) reduces to

$$[mu](z) = [mu]_0 + Gz \tag{16}$$

The bubble number conservation equation is

$$\frac{d(nu)}{dz} = N(z) \tag{17}$$

where n is the number of bubbles per unit total waste volume (1/m^3), u is the bubble velocity, and N is the volumetric bubble nucleation rate (1/m^3-s). The solution of Eq. (17) is

$$[nu](z) = [nu]_0 + \int_0^z N(z)dz \tag{18}$$

where $[nu]_0$ is the bubble number flux (1/m^2-s) at the tank bottom boundary z = 0, again usually zero. Assuming the nucleation rate is uniform, Eq. (18) reduces to

$$[nu](z) = [nu]_0 + Nz \tag{19}$$

The average bubble volume is given by the ideal gas equation of state as

$$v_B(z) = [m/n](z)\frac{RT}{p} = \frac{[mu](z)}{[nu](z)}\frac{RT}{p} \tag{20}$$

where R is the gas constant (8314 J/kg-mol-K), p is the average pressure (Pa), and T is the average temperature (K) in the sediment. The average pressure and temperature can be used in Eq. (20) because neither varies appreciably across the layer.

The gas volume fraction can be written as the product of the bubble number density and volume, as follows:

$$\alpha(z) = n(z)v_B(z) = \frac{[nu](z)}{u(z)}v_B(z) \tag{21}$$

Substituting Eq. (13) for u(z) and Eq. (20) for $v_B(z)$ in Eq. (21) gives

$$\alpha(z) = \frac{m_\tau}{SK\rho_S g}\left(\frac{RT}{p}\right)^{1/3}(H_S - z)[mu](z)^{1/3}[nu](z)^{2/3} \tag{22}$$

Now, substitute Eq. (16) for [mu](z) and Eq. (19) for [nu](z), letting the bubble velocity at the tank bottom boundary be zero:

$$\alpha(z) = \frac{m_\tau}{SK\rho_S g}\left(\frac{RT}{p}\right)^{1/3}(H_S - z)[Gz]^{1/3}[Nz]^{2/3} \tag{23}$$

Finally, define a nondimensional elevation $\eta = z/H_S$ so that $\eta = 0$ at tank bottom and $\eta = 1$ at the top of the sediment. Applying this definition to Eq. (23) gives the following equation for the gas volume fraction profile in the sediment:

$$\alpha(\eta) = \frac{m_\tau}{SK\rho_S g}\left(\frac{RT}{p}\right)^{1/3}G^{1/3}N^{2/3}H_S^2\eta(1-\eta) \tag{24}$$

4.0 Buoyancy Ratio

To determine the conditions for buoyancy, we integrate Eq. (24) from the top down to derive an expression for the average gas volume fraction above dimensionless elevation η_{NB}. If the integral average of the gas fraction profile from the top of the sediment layer becomes equal to the neutral buoyancy gas fraction at some elevation, all the waste down to that elevation is buoyant, as shown in Figure 2. This is expressed mathematically as

$$\alpha_{avg}(\eta_{NB}) = \frac{1}{1-\eta_{NB}} \int_{\eta_{NB}}^{1} \alpha(\eta)d\eta = \alpha_{NB} = \alpha(\eta_{NB}) \tag{25}$$

where η_{NB} is the lowest elevation where $\alpha(\eta) \geq \alpha_{NB}$. Performing this integration on Eq. (24), we find that

$$\alpha_{avg}(\eta_{NB}) = \frac{m_\tau}{SK\rho_S g}\left(\frac{RT}{p}\right)^{1/3} G^{1/3} N^{2/3} H_S^2 \frac{1}{6}(1+2\eta_{NB})(1-\eta_{NB}) \tag{26}$$

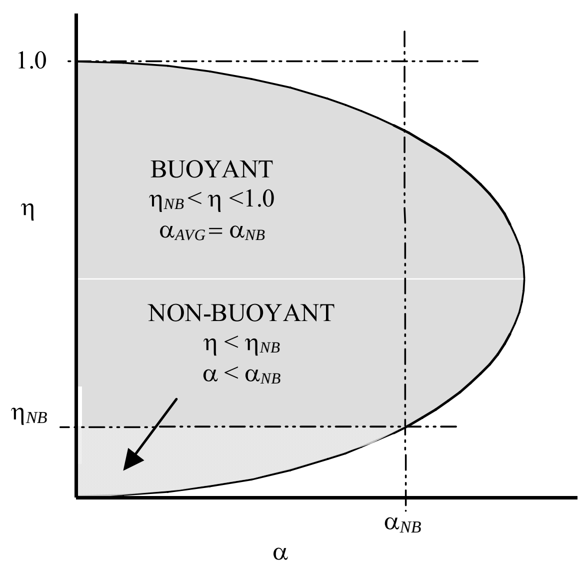

Figure 2. Integration of Gas Fraction Profile

Equating Eq. (26) and (24) evaluated at η_{NB} and solving, we find that $\eta_{NB} = 0.25$. It can also be shown that Eq. (25) is a maximum at $\eta = 0.25$ even when $\alpha_{avg} \neq \alpha_{NB}$. Evaluating Eq. (26) at $\eta_{NB} = 0.25$ gives

$$\alpha_{avg} = \left[\frac{3}{16}\frac{N^{2/3}R^{1/3}m_\tau}{SK\rho_S g}\right]\left(\frac{GT}{p}\right)^{1/3} H_S^2 \tag{27}$$

The ratio of average gas fraction and neutral buoyancy gas fraction, the *buoyancy ratio,* is a measure of the potential for eruptions to occur in sediment layers.

$$B = \alpha_{avg} / \alpha_{NB} \tag{28}$$

If B = 1, the portion of the sediment layer above the dimensionless elevation η_{NB} will become buoyant in the steady state. For B < 1, a layer cannot become buoyant. For B > 1, buoyancy will occur before steady state is achieved. An expression for the buoyancy ratio is derived by dividing Eq. (27) by the neutral buoyancy gas volume fraction, Eq. (3). This results in

$$B = \frac{C}{\rho_S - \rho_L} \left(\frac{GT}{p} \right)^{1/3} H_S^2 \tag{29}$$

where $C = \left[\frac{3}{16} \frac{N^{2/3} R^{1/3} m_\tau}{SKg} \right]$.

The coefficient C in Eq. (29) contains all the constants and unknown parameters that must be determined using tank data. To do this, all the tanks with a liquid-over-sediment waste configuration were evaluated with Eq. (29). Only five of these tanks are observed to exhibit buoyant displacement gas release events. The coefficient C was adjusted so that the minimum buoyancy ratio of these five tanks is exactly one.

5.0 References

Meyer PA, ME Brewster, SA Bryan, G Chen, LR Pederson, CW Stewart, and G Terrones. 1997. *Gas Retention and Release Behavior in Hanford Double Shell Waste Tanks.* PNNL-11536 Rev. 1, Pacific Northwest National Laboratory, Richland, Washington.

Meyer PA and BE Wells. 2000. "Understanding Gas Release Events in Hanford Double-Shell Tanks." *Proceedings of Waste Management 2000*, Tucson, Arizona.

Wells BE, JM Cuta, SA Hartley, LA Mahoney, PA Meyer, and CW Stewart. 2002. *Analysis of Induced Gas Releases During Retrieval of Hanford Double-Shell Tank Waste.* PNNL-13782 Rev.1, Pacific Northwest National Laboratory, Richland, Washington.