Waste Tank Deflagration Source Generation Mechanisms (U)

by

D. F. Paddleford
Westinghouse Savannah River Company
Savannah River Site
Aiken, South Carolina 29808

J. K. Thomas

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WASTE TANK DEFLAGRATION
SOURCE GENERATION MECHANISMS (U)

D.F. Paddleford and J.K. Thomas
Westinghouse Savannah River Company
Centennial Corp. Center 992-1W
Aiken, SC, 29801

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ABSTRACT
This paper describes the models employed and analyses performed to estimate the source terms which would result from deflagrations in waste storage and processing tanks at the Savannah River Site (SRS). It should be noted that SRS waste tank deflagrations are very low probability events. The three main source generation mechanisms associated with waste tank deflagrations are: (1) entrainment from the liquid surface, (2) vaporization from the liquid surface, and (3) solids fire ignited as a result of the deflagration. The results of this work show that entrainment would be the dominant source term generation mode for cases where the liquid waste solution level is near the vent path. Vaporization would be the dominant mode for cases where the liquid level is lower. The maximum vaporization source term is approximately an order of magnitude less than that maximum entrainment source term. The source term associated with a solids fire would be insignificant in comparison.
1. INTRODUCTION

Prior safety analyses dealing with waste storage tank deflagrations at the Savannah River Site (SRS) employed source terms based on the maximum aerosol density which could be established in the tank vapor space at some time following the event. However, this approach does not consider the higher aerosol densities which could be established during or immediately following a deflagration, or the loss of aerosol from the vapor space due to venting. Furthermore, this approach does not explicitly treat potential aerosol generation modes. The guidance given in Mishima (1993) for gas-phase explosion source terms is based upon shock wave aerosolization experiments, and is therefore not applicable to deflagrations. The work described in this paper was therefore carried out to provide a set of simplified models which could be used to provide realistic source term estimates for waste tank deflagrations. The approaches adopted in this work retain some degree of conservatism since they are intended for use in safety analyses.

The three main source generation mechanisms associated with waste tank deflagrations are: (1) entrainment from the liquid surface, (2) vaporization from the liquid surface, and (3) solids fire ignited as a result of the deflagration. In the following sections, the models and data employed are presented and typical results are given for each source generation mechanism. Three pieces of information are required in order to calculate a source term (Mishima 1993): (1) the amount of material impacted by the deflagration (e.g. amount of liquid entrained, amount of liquid vaporized, amount of solid combusted), (2) the airborne release fraction (ARF), which is this fraction of the radionuclides present in this material which can be suspended in air and made available for transport, and (3) the respirable fraction (RF), which is the fraction of the airborne radionuclides which can be transported and inhaled into the human respiratory system [generally taken to be particles with a mean aerodynamic equivalent diameters less than 10 μm (Mishima 1993)]. The source term is calculated as the product of these three parameters.

The waste tanks of interest (e.g. those with the greatest potential to develop combustible gas mixtures) are of the Type IIIA design class. Figure 1 is a cross section of a Type IIIA waste tank. The primary liner has an inside diameter of 85’ and a height of 33’; the central column has an outer diameter of 6’ 9.5”. The Type III tanks have an internal volume of approximately 5.2x10^6 liters. The waste tanks have a number of penetrations through the top slab (risers) which are normally closed with concrete plugs. In the event of a deflagration, these riser plugs would be ejected at internal tank pressures ranging from a few psig up to 13 psig (depending on the riser and riser plug geometries) and would provide anywhere from 25 to 60 ft^2 (depending on the tank). The risers have minimum diameters ranging from 1 to 3 feet.
II. ENTRAINMENT

The pressure rise associated with a waste tank deflagration would cause ejection of one or more riser plugs, creating vent paths which would limit further increases in pressure. The flow of gas through the vents, which could be choked flow if the pressure is sufficiently high, would create a radial flow of gas toward the region immediately below the vent opening. The magnitude of the gas velocity would be determined by the ratio of vent radius to freeboard height, and if sufficiently large would result in entrainment from the liquid surface. This gas flow induced entrainment was estimated by two different approaches: (a) a semi-mechanistic approach based on a simple radial flow approximation to the gas flow field in conjunction with approximate relationships for predicting the onset and magnitude of entrainment, and (b) an empirical correlation based on small scale test data.

A. Radial Flow Model

Critical gas velocities for atomization of flowing films of varying thickness have been measured in wind tunnel experiments (van Rossum 1959), pressure drop and entrainment have been measured for annular two-phase flow in horizontal pipe experiments [Wicks and Dukler (1960) and Wallis (1968)], and reviews of experimental and analytical work regarding inception of liquid entrainment have been provided by Zuber (1962), Kutateladze (1972), and Ishii and Grolmes (1975). Along with the turbulent jet entrainment model of Taylor (Morton et al. 1956), as modified for vastly different jet and ambient densities (Ricou and Spalding 1961), these provide the basis for the first method of estimating entrainment.

Epstein (1993) proposed a model for the entrainment of liquid out a hole in the top of a partially filled tank, based on high velocity horizontal gas flow shearing off wave tips. With exception of the use of a simplified radial flow assumption in the vicinity of the vent in place of a more detailed potential flow solution, the model described below is that proposed by Epstein.

When the pressure exceeds the critical pressure, defined as:

\[ p > p_{\text{crit}} = p_{\text{atm}} \left( \frac{\gamma + 1}{2} \right)^{\frac{\gamma}{\gamma - 1}} \]  

then the discharge rate is limited by the critical flow rate (choked flow), which can be written as:
where: \( p \) = Gas pressure in tank, 
\( P_{\text{crit}} \) = Critical pressure, 
\( P_{\text{atm}} \) = Atmospheric back pressure, 
\( \gamma \) = Specific heat ratio (= \( C_p/C_v = 1.4 \) for air), 
\( \dot{m}_g \) = Gas mass flow rate, 
\( G_c \) = Critical gas mass flux, 
\( A \) = Vent flow area, 
\( C_D \) = Vent discharge coefficient, and 
\( \rho_g \) = Gas density (at tank conditions).

At lower pressure the discharge rate is,

\[
\dot{m}_g = C_D A \left( \frac{\gamma p \rho_g}{\gamma + 1} \left( \left( \frac{P_{\text{atm}}}{p} \right)^\gamma - \left( \frac{P_{\text{atm}}}{p} \right)^{\gamma+1} \right) \right)^{0.5}
\]  

(2)

It is convenient to define a pseudo discharge gas velocity (\( V \)) as:

\[
V = \frac{\dot{m}_g}{\rho_g A}
\]

(4)

Note that \( V \) is not the actual discharge velocity at the vent exit since it is based on the density of the gas inside the tank. The pseudo discharge gas velocity is appropriate for estimating the gas velocity distribution over the liquid surface inside the tank when using a constant volumetric flow rate assumption.

The minimum gas velocity required for entrainment is (Kutateladze 1972):

\[
V_{\text{ent}} = 3.1 \left( \frac{\sigma g \rho_f}{\rho_g^2} \right)^{1/4}
\]

(5)

where: \( V_{\text{ent}} \) = Minimum entrainment velocity, 
\( \rho_f \) = Fluid (liquid) density, 
\( \sigma \) = Surface tension, and 
\( g \) = Gravitational constant.

The entrainment rate is calculated by integrating the local entrainment rate over the affected liquid surface area (e.g. where the gas velocity exceeds the minimum entrainment velocity). Using the model of Ricou and Spalding (1961), the entrainment rate can then be expressed as:
\[
\dot{m}_e = \int E_0 \left( \rho_r \rho_f \right)^2 \frac{1}{v(r)} 2\pi r dr
\]

where: 
- \( \dot{m}_e \) = Entrainment rate (entrained liquid mass flow rate),
- \( E_0 \) = Entrainment coefficient (taken as 0.1),
- \( v(r) \) = Gas velocity distribution, and
- \( r \) = Radial distance from centerline of vent.

The gas velocity distribution in the vicinity of the vent can be approximated as by a radial flow distribution if the vent opening is sufficiently far from walls or other obstructions. This assumption allows the gas velocity distribution to be written as:

\[
v(r) = \left( \frac{1}{2} VR_o^2 / H \right)^{1 \over r}
\]

where: 
- \( H \) = Freeboard (vertical distance between vent plane and liquid interface), and
- \( R_o \) = Vent radius.

Substituting this expression into eqn.(6) and integrating over the region where \( v(r) \) exceeds \( V_{ent} \) from \( r=R_o \) to \( r=VR_o^2/(2HV_{ent}) \) allows the entrainment rate to be expressed as:

\[
\dot{m}_e = E_0 \left( \rho_r \rho_f \right)^2 V \left( \pi R_o^2 \right) \left( \frac{R_o}{H} \right) \left[ \frac{1}{2} V \left( \frac{R_o}{H} \right) - 1 \right]
\]

In solving these equations, the gas discharge and entrainment rates were held constant over a small time step. The gas density was updated at the end of each time step to account for the gas discharge rate [eqn. (2) or (3)] along with the following relationship:

\[
\frac{d\rho_g}{dt} = -\frac{1}{Volume} \dot{m}_e
\]

The tank pressure was updated assuming isentropic expansion, using the expression given below, until atmospheric pressure was reached:

\[
\frac{p}{p_0} = \left( \frac{\rho_g}{\rho_{g,0}} \right)^{\gamma}
\]

The 'o' subscript denotes values at the beginning of the time step. This approach implicitly assumes that available vent area is sufficient to rapidly relieve the internal tank pressure (e.g. remainder of the deflagration occurs at a significantly reduced pressure). It would be necessary to couple the equation set developed above with a deflagration analysis for cases where this assumption is not valid. Alternatively, a conservative approximation suitable for some safety
analysis applications is to employ the ratio of the entrained liquid to discharged gas mass flow rates, as shown below:

\[
\frac{\dot{m}_e}{\dot{m}_g} = E_0 \left( \frac{\rho_l}{\rho_g} \right)^{\frac{1}{2}} \left( \frac{R_o}{H} \right) \left[ \frac{1}{2} \left( \frac{v}{V_{\text{end}}} \right) - \left( \frac{R_o}{H} \right) - 1 \right]
\]  

(11)

The total entrainment can be estimated by using the initial value of this ratio along with the total gas mass which must be vented. The mass of gas discharged assuming an isentropic blowdown to atmospheric pressure would be:

\[
m_g = m_{\text{initial}} \left( 1 - \left( \frac{14.7}{p} \right)^{\frac{1}{2}} \right)
\]

(12)

Applying the initial \( \dot{m}_e/\dot{m}_g \) value throughout the blowdown gives an entrained liquid mass of:

\[
m_e = m_g \left( \frac{\dot{m}_e}{\dot{m}_g} \right)_{\text{initial}}
\]

(13)

B. LBL Correlation

During a nuclear plant small loss of cooling accident (LOCA), stratified flow could occur in horizontal piping and, if the break were above the horizontal interface, liquid could be entrained due to vapor acceleration in the vicinity of the break (Zuber 1981). A number of studies have been carried out to develop modeling capability for this type of accident, and many of these studies have applications to the more general problem of vessel blowdown.

Smoglie et al. (1987) at KfK have studied entrainment through upward, horizontal, and downward oriented branches for stratified flow. For an upward branch, Smoglie et al. measured the height of vapor space below which entrainment begins (\( h_b \)) and the magnitude of entrainment as a function of the vapor space height (\( h/h_b > 0.6 \)) over the high quality (95% to 100%) region.

Similar experiments have been performed by Schrock et al. (1986) at LBL extending to much lower incipient entrainment heights (\( h/h_b \) down to 0.1) with qualities down to 2% for upward branches. Schrock et al. (1986) summarize small scale experiments in which the liquid entrainment was measured for critical discharge from small scale breaks (\( \leq 10 \text{ mm dia.} \)) in 10 cm piping containing stratified air and water layers. Two key correlations were developed by LBL as fits to their data.
The first correlation given by Schrock et al. allows the freeboard height below which liquid entrainment begins to be calculated, and is given as:

\[ Fr_g \left( \frac{\rho_g}{\Delta \rho} \right)^{0.5} = 0.395 \left( \frac{h_b}{d} \right)^{2.5} \]  

(14)

where: \( Fr_g = \) Froude Number \((= \sqrt{V/gd})\),  
\( d = \) Vent diameter,  
\( \Delta \rho = \rho_{liq} - \rho_g \),  
\( h_b = \) Freeboard height at which entrainment begins.

The second correlation given by Schrock et al. relates the quality of the vent flow to the reduced freeboard height \((h/h_b)\), and is given as:

\[ x = \left( \frac{h}{h_b} \right)^{3.25(1-h/h_b)} \]  

(15)

where: \( x = \) Quality, and  
\( h = \) Freeboard height.

The ratio of the entrained liquid to gas mass flow rates can be expressed in terms of quality, as shown below:

\[ \frac{\dot{m}_l}{\dot{m}_g} = \frac{1}{x} - 1 \]  

(16)

C. Source Term

The gas velocities through the vent throat are very large \((\approx \text{sonic})\) and have the potential to subdivide liquid droplets to respirable particles, assuming a critical Weber number on the order of 12 applies. Lane (1951) conducted experiments in which droplets were subjected to air blasts from a high pressure blast gun and reports that the 50% mass diameter of the droplet spray was greater than 15 microns for several atmospheres of pressure, even though his relationship indicated only drops of 5 microns should remain intact for air moving at sonic velocity relative to the droplets. Even with a 50% mass diameter of 15 microns, a substantial fraction of droplets in the respirable range cannot be ruled out. Furthermore, the entrained droplets would be subject to evaporation prior to exiting the tank. For the purposes of safety analyses at SRS, the respirable fraction (RF) is conservatively assumed to be unity, although it is recognized that the true value could be significantly lower.
D. Results
As an example illustrating the application of these models, consider the blowdown of a waste tank from a pressure of 25 psia through a 3 foot diameter vent. This pressure is in the range of the maximum values calculated for deflagrations in SRS waste storage and processing tanks. The 3 foot vent diameter is the largest available on the tanks of interest. The liquid and gas densities were taken to be 62 and 0.07 lbs/ft³, respectively.

The results obtained with the radial flow model for the post-deflagration blowdown transient at a freeboard of 3' (R₀/H = 0.5) are shown in Figure 2. This freeboard corresponds to the maximum liquid level for the waste tanks of interest; a freeboard of 2' corresponds to the overflow condition in these tanks. As shown in the figure, the total entrained liquid mass is calculated to be 770 pounds (350 kg). This is a relatively small fraction of the total tank contents and corresponds to a liquid layer thickness of approximately 0.03" (0.8 mm).

Figure 3 shows the ratio of the entrained liquid to discharged gas mass flow rates as a function of R₀/H (pressure of 25 psia and vent diameter of 3 feet) calculated with the radial flow model and LBL correlations [eqns. (11) and (16)]. The entrained liquid masses obtained by the approximate steady-state approach [eqns. (12) and (13)] at a vent radius to freeboard height ratio of 0.5 are 1150 lbs. (522 kg) with the radial flow model and 250 lbs. (110 kg) with the LBL correlation. For the radial flow model this approach overestimates the source term by 50%, which is expected given the decrease in the entrainment ratio with time shown in Figure 2.

It should be noted that, as shown in Figure 3, \( \dot{m}_l/\dot{m}_g \) goes to zero for R₀/H less than 0.17 (radial flow model) or 0.27 (LBL correlation); this occurs because the calculated gas velocity is below the value required for entrainment. Thus, these models predict no entrainment from vent flow when the freeboard height is greater than two or three vent diameters.
III. VAPORIZATION

Vaporization would occur at the surface of the liquid waste due to heat transfer from the hot product gas. For the conditions applicable to a waste tank deflagration, the radiative heat transfer rate to the liquid surface would be much larger than that due to convection until the deflagration was complete and the gas temperature and pressure decreased significantly. Only radiative heat transfer was considered in this evaluation.

A. Radiative Heat Flux

The radiative heat flux incident of the liquid waste solution surface from the product gas region was calculated using the Deflagration Pressure Analysis Code (DPAC), which is described in Hensel and Thomas (1994). The radiative heat flux is calculated assuming that the temperature of the surface is low compared to that of the gas [e.g. \( (T_g/T_s)^4 \gg 1 \)] and that the product gas can be treated as a gray body. The radiative heat flux can then be calculated as (Hottel and Sarofim 1967, Edwards and Matavosian 1984):

\[
q_r = \sigma_s \varepsilon_{\text{eff}} T_g^4
\]

\[
\varepsilon_{\text{eff}} = \frac{1}{1/\varepsilon_g + 1/\varepsilon_s - 1}
\]

where:  
\( q_r \) = Net radiative heat flux,
\( \sigma_s \) = Stefan-Boltzman constant \((5.67 \times 10^{-8} \text{ W/m}^2/\text{K}^4)\),
\( \varepsilon_{\text{eff}} \) = Effective emittance,
\( T_g \) = Gas temperature,
\( \varepsilon_g \) = Gas emittance, and
\( \varepsilon_s \) = Surface emittance.

Hydrogen (H\(_2\)) and benzene (C\(_6\)H\(_6\)) are the main combustible gases which can be generated in SRS waste tanks [benzene is only generated in the tanks employed with the In-Tank Precipitation Process (ITP)]. Of the gases which would be present in a hydrogen-benzene-air deflagration, only H\(_2\)O, CO\(_2\), and C\(_6\)H\(_6\) would participate in the radiative heat transfer process. The other components are diatomic (O\(_2\), N\(_2\), and H\(_2\)), do not emit thermal radiation, and are essentially transparent to incident thermal radiation (Incropera and DeWitt 1981, Siegel and Howell 1981). Benzene would only be present in the product gas if the reactants were hyperstoichiometric, which is not the case for any condition of interest. The total gas emittance for a gas containing H\(_2\)O and CO\(_2\) can be written as:

\[
\varepsilon_g = \varepsilon_w C_w + \varepsilon_c C_c - \Delta \varepsilon
\]
where: \( \hat{\varepsilon}_j \) = Emittance of jth gas component at a total gas pressure of 1 atmosphere, 
\( C_j \) = Correction for actual total gas pressure for jth component, and 
\( \Delta \varepsilon \) = Correction for interference from multiple emitting species.

The 'w' and 'c' subscripts denote water vapor and carbon dioxide gas, respectively. If H\(_2\) is the only combustible species in the reactant gas mixture (e.g. tanks other than those employed with the ITP process), the 2nd and 3rd terms in this expression would be zero. Correlations for the terms given in this expression were constructed using the data given by Hottel (1954); the correlations employ temperature, pressure, and mean beam length as the independent parameters. The product gas temperature and pressure are calculated by DPAC, and vary throughout the deflagration. Based on the data given in (Siegel and Howell 1981), the mean beam length (\( L_e \)) for an arbitrary enclosure can be calculated as:

\[
L_e = 3.5 \frac{V_g}{A_s}
\]

where: \( V_g \) = Gas volume, and 
\( A_s \) = Solid surface area.

The method described above was benchmarked against experimental radiative heat flux data measured in the NTS hydrogen dewar with a 9.9% H\(_2\)-air mixture deflagration (Ratzel 1985). The spherical dewar has an interior volume of 2.1x10\(^6\) liters, which is 40% of that for an empty waste tank. The calculated peak radiative heat flux is approximately 20% lower than the experimental value. The result is expected since the data employed for the water vapor emittance (Hottel 1954) is low relative to later evaluations (Boynton and Ludwig 1971, Siegel and Howell 1981). However, it is expected that the calculated radiative heat fluxes would be conservative for deflagrations involving higher combustible gas concentrations since DPAC assumes complete combustion without consideration of the actual product gas equilibrium concentration and hence overpredicts product gas temperatures.

B. Liquid Vaporization

To compute the amount of liquid vaporized, it is conservatively assumed that no heat is conducted away from the liquid surface (e.g. all energy is deposited and remains in a thin liquid layer near the surface). The mass of liquid vaporized can therefore be expressed as:

\[
M_l = \frac{A_l q_r t_b}{h_{fg}}
\]

where: \( M_l \) = Mass of liquid vaporized, 
\( q_r \) = Average radiative heat flux over total burn time, 
\( A_l \) = Liquid surface area over which \( q_r \) is incident,
\( t_b \) = Burn time (total time required to complete deflagration), and
\( h_{fg} \) = Heat of vaporization for liquid.

C. Source Term
The radionuclides in the waste solution would be present as dissolved salts, precipitates, and small oxide particles. The salt concentration in the surface layer would increase during vaporization and, as a result, additional precipitates would form. Since the time required to complete the deflagration is relatively short, the increased concentration of the surface layer would not be significantly reduced by diffusion back into the bulk of the liquid. Some of these oxide and salt particles, as well as water droplets created at the surface, would be entrained by the steam lifting off the water surface.

Slow evaporation from a liquid surface would be expected to give a very small ARF. Alternatively, very rapid boiling would be more closely related to a liquid phase explosion than to slow evaporation and would give a much higher ARF. Vaporization during a waste tank deflagration lies between these two cases in terms of the time scale involved. In addition to considering time scale, it should be recognized that the physical mechanisms involved in entrainment by steam created via surface boiling due to radiant energy deposition (e.g. a waste tank deflagration) are not the same as those for entrainment of steam created within a volume of liquid which subsequently rises to the liquid surface (e.g. a boiler). The main mechanism for entrainment with a boiler is the production and dispersion of small liquid drops from steam bubbles rising to and breaking through the surface. In the case of the deflagration, the steam would be created at the surface and hence fewer droplets would be created; conversely, there would be some horizontal gas flow across the surface in a deflagration due to product gas expansion which would tend to promote entrainment of droplets and particles.

SRS safety analyses of non-reactor facilities generally assume an ARF of \( 10^{-4} \) for slow evaporation from a liquid surface, which is generally considered to be conservative. Mishima (1993) recommends bounding and median ARF values of \( 3 \times 10^{-5} \) and \( 6 \times 10^{-7} \) for heating of aqueous solutions in flowing air without generation of bubbles, and ARF values of 0.002 and 0.001 for boiling solutions in flowing air. Godbee (1973) gives data for surface entrainment via steam generation for a wide variety of evaporators; release fractions of approximately 0.1 were reported for large-scale boilers operated with mass fluxes representative of those calculated using the approach discussed above. Mishima (1993) recommends a bounding and median ARF values of 0.1 and 0.02 for venting of superheated liquids with 30 to 100°C of superheat (e.g. flashing upon depressurization); higher ARF values would be applicable to liquids with higher superheats.
Based on the large boiler and superheated liquid data and the considerations discussed above, it is assumed that 10% of the radionuclides in the liquid mass vaporized would exit the tank and contribute to the source term (ARF = 0.1). It is judged that this value is slightly conservative. Since airborne liquid droplets would continue to evaporate prior to exiting the tank and would be subject to large shearing forces in the high velocity gas stream passing through the tank risers, it is assumed that essentially all airborne droplets and particles would be respirable (RF = 1). The source term is therefore taken to be 10% of the liquid mass vaporized (ARF-RF = 0.1).

D. Results
The worst-case with respect to liquid vaporization would be a nearly empty waste tank since this gives the maximum vapor space volume and hence maximizes both the radiative heat flux and the total time over which it acts. Of course, low liquid levels will decrease the combustible gas generation rate and increase the time required to develop a given vapor space combustible gas concentration. For the waste storage tanks, a high fill level is required in order to achieve a flammable mixture in any reasonable amount of time following a loss of ventilation. However, for the ITP waste tanks, flammable H2-C6H6 mixtures could be formed even at low fill levels over time periods relevant for safety analyses. The results discussed below are for Tank 48, which has the smallest vent area of the ITP tanks and hence gives the largest gas pressures and temperatures. A C6H6/H2 ratio of 1.5 was employed, which approximates the nominal condition for this tank during the period of maximum combustible gas production.

Table 1 gives the burn times and average radiative heat fluxes calculated by DPAC for mixtures of combustible gas with air at the lower flammability limit (LFL), at the stoichiometric concentration, and half way between the LFL and stoichiometric concentrations. The mass of liquid vaporized is also given in Table 1. The liquid mass vaporized is essentially the same for all cases; the decrease in burn time and increase in radiative heat flux as the combustible gas concentration increases from the LFL up to the stoichiometric concentration offset one another so that the total amount of energy transferred to the liquid during the deflagration is only weakly dependent upon the gas concentration. The corresponding liquid volume and depth, based on the density for saturated liquid water at a pressure of 2 atmospheres (calculated peak deflagration gas pressure), are approximately 0.4 m³ and 0.8 mm, respectively. Using ARF and RF values of 0.1 and 1.0, the corresponding source term mass for these cases would be approximately 40 kg. Note that this maximum vaporization source term is approximately one order of magnitude less than the maximum entrainment source term.
IV. SOLIDS FIRE
Solid deposits can be formed on internal surfaces near the waste solution - air interface in liquid waste storage and processing tanks. For waste tanks which contain organic materials, as with ITP waste tanks where tetraphenylborate (TPB) is employed to precipitate cesium from liquid waste solutions, these deposits may be combustible.

A. Solid Deposit Description
The solid deposit characterization data relevant to solid combustibility are summarized in Thomas (1993). The deposits formed in the ITP tanks are composed of soluble solids, insoluble solids, and water. The soluble solids are composed primarily of sodium nitrate (NaNO₃), but also include sodium nitrite, hydroxide, and sulfate in lesser amounts. The insoluble solid component is almost entirely potassium tetraphenylborate (KTPB, KBC₂₄H₂₀), but also contains small amounts of cesium and sodium tetraphenylborate salts and sodium titanate. It was assumed that the solid fraction of a deposit would be comprised entirely of NaNO₃, since deposits rich in soluble solids ignite and support combustion most readily and since the thermal properties of the other solid components are not greatly different than those for NaNO₃.

Based on the results of tests performed with dried deposits prepared from simulated waste slurries and those taken from vessels employed in pilot scale tests, the liquid and solid volume fractions for conditions relevant to the ITP waste tanks would be approximately 86 and 14%, respectively. Note that the deposit would have a very high porosity if the liquid phase were lost due to vaporization. The corresponding liquid and solid mass fractions are 72 and 28%, respectively.

Differential Thermal Analysis, autoignition temperature, autooxidation temperature tests run in air with dried deposits indicate that the deposit temperature must exceed at least 300°C prior to the onset of any exothermic reactions. On the basis of these data, it is assumed that the surface of a deposit inside the waste tank would have to be heated to at least 300°C before ignition would be possible. It should be noted that ignition tests demonstrated that some deposits will not ignite even under a propane torch flame, so that assuming a 300°C surface temperature would lead to ignition is conservative.

Assuming that the deposit surface can be ignited due to radiant energy deposition, then for the combustion reaction to be self-sustaining the energy liberated by combustion must be equal to or greater than the energy required to heat the underlying deposit to the ignition temperature. The
heat of combustion for a stoichiometric NaNO₃-NaTPB mixture would be 1.2 kcal per gram of solid, or 0.33 kcal per gram of deposit. As discussed above, the minimum temperature required to initiate sustained combustion in a dry deposit is at least 300°C. The energies required to raise the temperature of water from 40 to 100°C and then to boil it are 60 and 539 cal/g (per gram of H₂O) and the energy required to raise the temperature of sodium nitrate from 40 to 300°C is 88 cal/g (per gram of NaNO₃). This gives a total energy requirement of 0.46 kcal per gram of deposit. Since significantly more energy is required to heat the deposit up to the minimum ignition temperature than is released by combustion, the reaction cannot be sustained and only a thin outer layer first dried by radiant heating can combust. The energy deficit would actually be much larger than given above since a maximum energy release and a minimum ignition temperature were employed and since most of the combustion energy would be lost to gas-phase products.

B. Thermal Response

The thermal response of the deposit to the radiative heat flux from the product gas was described using a one-dimensional transient conduction analysis. The deposit can be treated as a semi-infinite solid since the time span over which the heat flux would be imposed is short. The transient temperature profile can therefore be expressed as (Carslaw and Jaeger 1959, Incropera and DeWitt 1981):

\[
\Delta T(y,t) = \frac{2q_r}{k} \sqrt{\frac{\alpha t}{\pi}} \exp\left(-\frac{y^2}{4\alpha t}\right) - \frac{q_r y}{k} \operatorname{erfc}\left(\frac{y}{2\sqrt{\alpha t}}\right)
\]

(22)

where: \(\Delta T\) = Temperature increase,
\(y\) = Distance from surface exposed to the heat flux,
\(t\) = Time,
\(q_r\) = Heat flux,
\(k\) = Thermal conductivity,
\(\alpha\) = Thermal diffusivity \(= k/(\rho C_p)\),
\(\rho\) = Density, and
\(C_p\) = Specific heat.

The temperature increase at the surface (\(x = 0\)) is therefore:

\[
\Delta T_s(t) = \frac{2q_r}{k} \sqrt{\frac{\alpha t}{\pi}}
\]

(23)

It is noted that this approach does account for the energy released by combustion. However, most of this energy would be carried away by product gases so that this approximation is acceptable.
C. Effective Thermal Properties

As discussed above, the solid deposit is assumed to be a NaN03-H2O composite. The deposit effective specific heat and density were taken to be the mass and volume averages, respectively, of the component values. The deposit effective thermal conductivity is dependent upon the geometric arrangement of the phases. For the purposes of this evaluation, the deposit was assumed to be a square grid of NaN03, the pores of which are filled with H2O. The effective thermal conductivity of a unit cell in the square grid, calculated using the electrical resistance analog approach, can be expressed as (Thomas 1993):

\[
\frac{k_{\text{eff}}}{w} = \frac{k_s \left( \frac{2-w}{1-w} \right) + k_l \left( \frac{1-w}{w} \right)}{w \left( \frac{2-w}{1-w} \right) + \frac{k_l}{k_s} \left( 1 - \frac{w}{w} \right) + \frac{1}{w}}
\]  

(24)

where:  
- \( k_{\text{eff}} \) = Effective deposit thermal conductivity,  
- \( k_j \) = Thermal conductivity of the \( j \)th deposit component, and  
- \( w \) = Fractional width of solid ligament between adjacent liquid cells 

(one minus the cube root of the liquid phase volume fraction).

The temperature range of interest is from 40°C (initial vapor space temperature) up to 300°C (minimum deposit ignition temperature). Below 100°C, the effective deposit properties are calculated using those for liquid water. Above 100°C, the water would vaporize and steam properties are employed. Since phase changes were not accounted for in the temperature profile expressions, the contribution from the water heat of vaporization spread over the temperature range of interest was included in the composite specific heat employed for cases where the surface temperature exceeds 100°C. At a given point in the transient, this approach will overpredict temperatures between 100 and 300°C and underpredict temperatures above 300°C. Table 2 gives the resulting property values for temperatures between 40 and 100°C, 100 and 300°C, and the values averaged over the 40 to 300°C temperature range. The table also gives values for the square root of the thermal diffusivity divided by the thermal conductivity, which governs the rate of the surface temperature rise [eqn.(23)]. These values show that the rate at which the surface temperature increases would go up by an order of magnitude once the surface temperature reached 100°C. For most cases, this indicates that if the deposit surface were to be heated to 100°C, then it would reach the ignition temperature (300°C).

D. Source Term

The methodology described can be used to estimate the amount of deposit which exceeds the minimum ignition temperature and hence is assumed to combust. The primary radionuclide contained in ITP waste tank deposits is \(^{137}\)Cs. Data from deposit combustion experiments
indicate that no more than 3.35 curies of $^{137}$Cs are released per kilogram of deposit combusted. A RF value of unity was assumed.

E. Results

As with the liquid waste vaporization source term, the largest solid fire source term would result from a deflagration involving a large vapor spaces since this maximizes the radiative heat flux and burn time. The radiative heat flux and burn time values given in Table 1 were also employed in this evaluation; the heat flux values were increased by 10% to represent peak values near the ignition location. The surface temperature was calculated to exceed the assumed 300°C ignition temperature for all three combustible gas compositions. In the worst case (stoichiometric mixture), it was calculated that a deposit depth of 0.4 mm would exceed 300°C prior to the end of the burn.

Assuming a 10 cm wide band of solid deposit were to form over all internal tank surfaces (primary liner, cooling coil tubes, and center column), then the total mass of deposit combusted for a 0.4 mm combustion depth would be 2.74 kg. The corresponding $^{137}$Cs release would be 9.2 Ci. For comparison, the corresponding liquid waste vaporization source term calculated in the preceding section was 42 kg. The minimum liquid waste $^{137}$Cs concentration under the applicable conditions would be approximately 10 Ci/gallon, which gives a $^{137}$Cs source term of 110 Ci. Since this is an order of magnitude greater than the largest source term for a solids fire, it is concluded that the solids fire source term is insignificant in comparison to that from liquid waste vaporization. The conservatisms inherent in the calculation of the vaporization source term more than account for that due to a solids fire.
V. CONCLUSIONS

The three main source generation mechanisms associated with waste tank deflagrations are: (1) entrainment from the liquid surface, (2) vaporization from the liquid surface, and (3) solids fire ignited as a result of the deflagration. The results of this work show that entrainment would be the dominant source term generation mode for cases where the liquid waste solution level is near the vent path. Vaporization would be the dominant mode for cases where the liquid level is lower. The maximum vaporization source term is approximately an order of magnitude less than that maximum entrainment source term. The source term associated with a solids fire would be insignificant in comparison. The source terms calculated in this work are above those based on maximum aerosol densities (prior safety analysis approach) and below those based on shock wave aerosolization data.

The models employed in this work are fairly simplistic and more accurate results could be achieved with more detailed models. Some of the specific areas for improvement which were identified during the course of this work are outlined below.

The radial flow entrainment model invokes a number of assumptions regarding the gas flow field. These assumptions could be avoided to some degree by applying the entrainment equations to the gas flow field calculated by a computational fluid dynamics code. The entrainment data used to develop the models employed in this work were for water-like fluids. It would be useful to perform similar experiments with fluids more representative of those in the waste tanks. Similarly, the entrainment data was taken from small-scale experiments and it would be useful to perform similar experiments on a larger scale. For both the entrainment and vaporization source terms, the ARF and RF values were selected in a conservative manner and additional work in this area would undoubtedly justify the use of lower values.

The vaporization source term model invokes a number of simplistic conservative assumptions, and lower source terms could be achieved by refining this model. In particular, a more accurate calculation of the gas temperature at near-stoichiometric gas concentrations would significantly lower the calculated heat flux. For lower combustible gas concentrations, where the burn time is significant, accounting for the transport of energy away from the liquid surface via conduction would decrease the calculated source term significantly.

These results presented in this paper indicate that no additional work on the solids fire source term would be justified since it is already insignificant in comparison.
VI. ACKNOWLEDGMENTS
The information contained in this paper was developed during the course of work under Contract No. DE-AC09-89SR18035 with the U.S. Department of Energy. The authors gratefully acknowledge the assistance of Michael Epstein (Fauske and Associates) for his deep insight and knowledge in proposing an approach for modeling entrainment from flow fields and identifying available data and correlations. The solid deposit experimental data was provided by Doug Walker (Savannah River Site) and his contributions to this work are gratefully acknowledged. The contributions of Steve Hensel (Savannah River Site) in the DPAC radiative heat flux and solid deposit thermal response analyses are also gratefully acknowledged.
VII. REFERENCES


Schrock, V.E. et al. (1986) *Small Break Critical Discharge-The Role Of Vapor and Liquid Entrainment in a Stratified Two-Phase Region Upstream of the Break*, LBL-22024, UC Lawrence Berkeley Laboratory.


VIII. NOMENCLATURE

\begin{align*}
A &= \text{Vent flow area} \\
A_l &= \text{Liquid surface area over which } q_r \text{ is incident} \\
A_s &= \text{Solid surface area} \\
C_D &= \text{Vent discharge coefficient} \\
C_j &= \text{Correction for actual total gas pressure for } j\text{th component} \\
C_p &= \text{Specific heat at constant pressure} \\
C_v &= \text{Specific heat at constant volume} \\
d &= \text{Vent diameter} \\
E_0 &= \text{Entrainment coefficient (taken as 0.1)} \\
Fr_g &= \text{Froude Number } (= V/\sqrt{gd}) \\
g &= \text{Gravitational constant} \\
G_e &= \text{Critical gas mass flux} \\
h_0 &= \text{Freeboard at which entrainment begins} \\
h_{fg} &= \text{Heat of vaporization for liquid} \\
H &= \text{Freeboard (vertical distance between vent plane and liquid interface)} \\
k &= \text{Thermal conductivity} \\
k_{eff} &= \text{Effective deposit thermal conductivity} \\
k_j &= \text{Thermal conductivity of the } j\text{th deposit component} \\
L_e &= \text{Mean beam length} \\
\dot{m}_e &= \text{Entrainment rate (entrained liquid mass flow rate)} \\
m_e &= \text{Total mass of liquid entrained} \\
\dot{m}_g &= \text{Gas mass flow rate} \\
m_g &= \text{Total mass of gas vented} \\
M_l &= \text{Mass of liquid vaporized} \\
p &= \text{Gas pressure in tank} \\
p_{crit} &= \text{Critical pressure} \\
p_{atm} &= \text{Atmospheric back pressure} \\
q_r &= \text{Net radiative heat flux} \\
r &= \text{Radial distance from centerline of vent} \\
R_o &= \text{Vent radius} \\
t &= \text{Time} \\
t_b &= \text{Burn time (total time required to complete deflagration)} \\
T_g &= \text{Gas temperature} \\
\Delta T &= \text{Temperature increase} \\
\Delta T_s &= \text{Temperature increase at the surface} \\
\nu(r) &= \text{Gas velocity distribution} \\
V &= \text{Pseudo discharge gas velocity} \\
V_{ent} &= \text{Minimum entrainment velocity} \\
V_g &= \text{Gas volume} \\
w &= \text{Fractional width of solid ligament between adjacent liquid cells} \\
x &= \text{Quality} \\
y &= \text{Distance from surface exposed to the heat flux} \\
\alpha &= \text{Thermal diffusivity } (= k/(\rho \cdot C_p)) \\
\varepsilon_{eff} &= \text{Effective emittance} \\
\varepsilon_g &= \text{Gas emittance} \\
\varepsilon_j &= \text{Emittance of } j\text{th gas component at a total gas pressure of 1 atmosphere} \\
\varepsilon_s &= \text{Surface emittance}
\end{align*}
\( \Delta e \) = Correction for interference from multiple emitting species
\( \gamma \) = Specific heat ratio (= \( C_p/C_v \))
\( \rho \) = Density
\( \rho_f \) = Fluid (liquid) density
\( \rho_g \) = Gas density (at tank conditions)
\( \Delta \rho \) = \( \rho_{\text{liq}} - \rho_g \)
\( \sigma \) = Surface tension
\( \sigma_s \) = Stefan-Boltzmann constant \((5.67 \times 10^{-8} \text{ W/m}^2/\text{K}^4)\)
Table 1 Vaporization Source Term.

<table>
<thead>
<tr>
<th>Combustible Gas Concentration [a]</th>
<th>Burn Time (sec) [b]</th>
<th>Radiative Heat Flux (W/cm²) [c]</th>
<th>Mass Vaporized (kg)</th>
<th>Source Mass (kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LFL</td>
<td>38</td>
<td>5.0</td>
<td>409</td>
<td>41</td>
</tr>
<tr>
<td>intermediate</td>
<td>12</td>
<td>15</td>
<td>400</td>
<td>40</td>
</tr>
<tr>
<td>stoichiometric</td>
<td>6.7</td>
<td>29</td>
<td>420</td>
<td>42</td>
</tr>
</tbody>
</table>

Notes: [a] Benzene to hydrogen ratio of 1.5, LFL calculated using Le Chatelier's rule (Zabetakis 1965). [b] Time required to complete deflagration. [c] Averaged over entire burn time.

Table 2 Effective Solid Deposit Properties.

<table>
<thead>
<tr>
<th>Property</th>
<th>Temperature Range (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>40 to 100</td>
</tr>
<tr>
<td>C_p (J/g/K)</td>
<td>3.4</td>
</tr>
<tr>
<td>ρ (g/cm³)</td>
<td>1.2</td>
</tr>
<tr>
<td>k (W/m/K)</td>
<td>0.66</td>
</tr>
<tr>
<td>α (m²/s)</td>
<td>1.6x10⁻⁷</td>
</tr>
<tr>
<td>υ/α /k</td>
<td>6.1x10⁻⁴</td>
</tr>
</tbody>
</table>
Cooled Waste Storage Tank
Stress Relieved Primary Liner  1,300,000 Gallons
Type III

Figure 1 Type IIIA Waste Tank Cross Section.
Figure 2  Radial Flow Model Entrainment Ratio and Integrated Entrainment (p=25 psia, Ro=1.5', H=3').
Figure 3: Steady-State Entrainment Estimates (p=25 psia and Ro=1.5).

Ratio Vent Radius to Freeboard Height