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EVALUATING DETONATION POSSIBILITIES IN A HANFORD RADIOACTIVE WASTE TANK

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SUMMARY

Since the early 1940s, radioactive wastes generated from the defense operations at the Hanford Site have been stored in underground waste storage tanks. During the intervening years, the waste products in some of these tanks have transformed into a potentially hazardous mixture of gases and solids as a result of radiolytic and thermal chemical reactions. One tank in particular, Tank 101-SY, has been periodically releasing high concentrations of a hydrogen/nitrous oxide/nitrogen/ ammonia gas mixture into the tank dome vapor space. There are concerns that under certain conditions a detonation of the flammable gas mixture may occur.

There are two ways that a detonation can occur during a release of waste gases into the dome vapor space: (1) direct initiation of detonation by a powerful ignition source, and (2) deflagration to detonation transition (DDT). The first case involves a strong ignition source of high energy, high power, or of large size (roughly 1 g of high explosive (4.6 kJ) for a stoichiometric hydrogen-air mixure[1]) to directly initiate a detonation by "shock" initiation. This strong ignition is thought to be incredible for in-tank ignition sources. The second process involves igniting the released waste gases, which results in a subsonic flame (deflagration) propagating into the unburned combustible gas. The flame accelerates to velocities that cause compression waves to form in front of the deflagration combustion wave. Shock waves may form, and the combustion process may transition to a detonation wave.

Flame acceleration depends on the "sensitivity" of the mixture and on geometric factors such as obstacles, scale, and amount of expansion volume near the accelerating combustion wave.[2] For dry air-hydrogen mixtures, Sherman and Berman[3] define mixture class in terms of equivalence ratio, while for air-hydrogen-steam mixtures, Sherman and Berman propose the detonation cell size as a measure of detonation sensitivity and define mixture class in terms of this parameter. Photo-

graphic evidence shows that the detonation zone consists of a complex, three-dimensional cellular structure.[4] The detonation cell width characterizes the size of the cells, and it is an experimentally measured parameter. In the absence of a full range of relevant experimental data, we have extended this methodology with calculations carried out using STANJAN,[5] an equilibrium code to solve the properties of a wave, and ZND,[6] a code that approximates the Shepherd interpretation of the Zel'dovich, von Neumann, and Döring model of the detonation reaction zone thickness,[7] to predict the mixture's sensitivity to detonation. Experiments conducted by Akbar and Shepherd[8] in mixtures composed of hydrogen, nitrous oxide, nitrogen, and oxygen were used to relate the measured detonation cell size to the calculated reaction zone thickness determined by ZND.

On the basis of the detonation cell size, mixtures are divided into five sensitivity classes: (1) mixtures that are extremely detonable near stoichiometric; (2) mixtures that are less likely to detonate; (3) mixtures that have been observed to undergo detonations in geometries that favor flame acceleration; (4) mixtures that have been observed to propagate a detonation, but a DDT has not been observed; and (5) mixtures that are unlikely to undergo DDT. The flame acceleration potential of a given volume is classified into one of five geometric classes, beginning with geometric class 1 being the most conducive to flame acceleration, to geometric class 5 being the least conducive.

For a selected waste gas composition released into the vapor dome space, we calculate the detonation cell size as a function of time and space for the waste tank dome vapor space and the attached ventilation system. We observe that the most sensitive mixture class occurs in small volumes of the dome vapor space for nearly the duration of the gas release period. However, the geometry class is not inclined to promote DDT, and the overall judgment is that DDT is possible but unlikely. In the ventilation system, a geometry judged very favorable for DDT, the volume fraction of hydrogen in the gaseous mixture is lower than that observed for DDT. Thus, DDT in the ventilation system is thought to be highly unlikely. Therefore, a detonation in the overall dome-space/ventilation system configuration during a release of waste gases is judged to be possible to highly unlikely.

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ABSTRACT

Since the early 1940s, radioactive wastes generated from the defense operations at the Hanford Site have been stored in underground waste storage tanks. During the intervening years, the waste products in some of these tanks have transformed into a potentially hazardous mixture of gases and solids as a result of radiolytic and thermal chemical reactions. One tank in particular, Tank 101-SY, has been periodically releasing high concentrations of a hydrogen/nitrous oxide/nitrogen/ ammonia gas mixture into the tank dome vapor space. There are concerns that under certain conditions a detonation of the flammable gas mixture may occur.

On the basis of the detonation cell size, mixtures are divided into five sensitivity classes: (1) mixtures that are extremely detonable near stoichiometric; (2) mixtures that are less likely to detonate; (3) mixtures that have been observed to undergo detonations in geometries that favor flame acceleration; (4) mixtures that have been observed to propagate a detonation, but a deflagration to detonation transition (DDT) has not been observed; and (5) mixtures that are unlikely to undergo DDT. The flame acceleration potential of a given volume is classified into one of five geometric classes, beginning with geometric class 1 being the most conducive to flame acceleration, to geometric class 5 being the least conducive.

For a selected waste gas composition released into the vapor dome space, we calculate the detonation cell size as a function of time and space for the waste tank dome vapor space and the attached ventilation system. We observe that the most sensitive mixture class occurs in small volumes of the dome vapor space for nearly the duration of the gas release period. However, the geometry class is not prone to promote DDT, and the overall judgment is that DDT is possible but un-

likely. In the ventilation system, a geometry judged very favorable for DDT, the volume fraction of hydrogen in the gaseous mixture is lower than that observed for DDT. Thus, DDT in the ventilation system is thought to be highly unlikely. Therefore, a detonation in the overall dome-space/ventilation system configuration during a release of waste gases is judged to be possible to highly unlikely.

INTRODUCTION

There are two ways that a detonation can occur during a release of waste gases into the dome vapor space: (1) direct initiation of detonation by a powerful ignition source, and (2) deflagration to detonation transition (DDT). The first case involves a strong ignition source of high energy, high power, or large size [roughly 1 g of high explosive (4.6 kJ) for a stoichiometric hydrogen-air mixture][1] to initiate directly a detonation by "shock" initiation. This strong ignition source is enough to be incredible for in-tank ignition sources. The second process involves igniting the released waste gases, which results in a subsonic flame (deflagration) propagating into the unburned combustible gas. The flame accelerates to velocities that cause compression waves to form in front of the deflagration combustion wave. Shock waves may form, and the combustion process may transition to a detonation wave. Flame acceleration is dependent on the "sensitivity" of the mixture and on geometric factors such as obstacles, scale, and amount of expansion volume near the accelerating wave front.[2]

The sensitivity of the mixture is normally described as the detonation cell width, which is usually investigated for hydrogen-air systems as a function of the equivalence ratio. This ratio is defined as the ratio of the hydrogen to oxygen mole fractions divided by the same ratio for stoichiometric conditions. Therefore, for stoichiometric conditions, the equivalence ratio is unity, for fuel lean mixtures it is less than 1, and for fuel rich mixtures it is greater than 1. In fact, the equivalence ratio is an exact measure of the amount of hydrogen that exists in the hydrogen-air system.

The detonation cell width is a length scale associated with detonation waves, which are characterized by an unsteady cellular combustion front. It is well known that detonation waves consist of transverse, reflected, and Mach-stem shock waves all meeting at Mach triple points.[3] These loci of triple points actually produce a traceable diamond-shaped pattern that can be measured. The transverse width of this pattern is called the detonation cell width, which has been experimentally related to detonation limits in various geometries.[4]

METHODOLOGY

There is no theory that can predict flame acceleration to detonation for all mixture conditions and geometries. The work at Sandia National Laboratories (SNL),[4] however, is an effort to quantify a methodology to examine and give guidelines for predicting DDT. We will summarize the methodology in the following paragraphs. The methodology is based on extensive experimental data and years of research experience, so for the validity and justification of the methodology, the reader is referred to the original paper.[4]

The methodology is based on the following assumptions:

1. The likelihood of DDT can be expressed as a function of two variables, one based on the sensitivity of the mixture and the other based on the flame acceleration potential of the geometry through which the deflagration propagates.

2. The sensitivity of the mixture is based on the detonation cell width or equivalence ratio for a hydrogen-air system.

3. The flame acceleration potential in a given geometry can be estimated from such characteristics as obstacles and size by reference to simple guidelines.

Based on the detonation cell width, λ, and equivalence ratio, Φ, mixtures are divided into five sensitivity classes:

1. mixtures that are extremely detonable near stoichiometric;

2. mixtures that are less likely to detonate;

3. mixtures that have been observed to undergo detonations in geometries that favor flame acceleration;

4. mixtures that have been observed to propagate a detonation, but a DDT has not been observed; and

5. mixtures that are unlikely to undergo DDT.

In Table I we present the classification of mixture sensitivity to detonation for dry hydrogen-air mixtures at 20°C and 1-atm pressure.

The flame acceleration potential of a given volume is classified into one of five geometric classes, beginning with geometric class 1 being the most conducive to flame acceleration to geometric class 5 being the least conducive.

TABLE I
CLASSIFICATION OF HYDROGEN-AIR MIXTURES AT 20°C AND 1-ATM PRESSURE

Mixture Class	Detonation Cell Width (mm)	Equivalence Ratio
1	20 to 15	$0.75 < \Phi \leq 1.5$
2	40 to 20	$0.63 \leq \Phi < 0.75, 1.5 < \Phi \leq 2.2$
3	320 to 40	$0.42 \leq \Phi < 0.63, 2.2 < \Phi \leq 4.1$
4	1200 to 320	$0.37 \leq \Phi < 0.42, 4.1 < \Phi \leq 5.6$
5	No Data	$\Phi < 0.37, \Phi > 5.6$

A description of these classes follows.

Geometric Class 1. Large geometries with obstacles in the path of the expanding unburned gases. Partial confinement favors gas expansion past obstacles. An example is a large tube with numerous obstacles and with ignition going from a closed end to an open end. Class 1 geometries are the most favorable to large flame acceleration.

Geometric Class 2. Geometries similar to class 1 but with some features that hinder flame acceleration. Examples would be a tube open on both ends or large amounts of transverse volume for expansion to the direction of the flame propagation.

Geometric Class 3. Geometries that yield moderate flame acceleration but are neutral to DDT. Examples are large tubes without obstacles and small tubes (several inches in diameter) with obstacles.

Geometric Class 4. Geometries unfavorable to flame acceleration. Examples are (1) large volumes with few obstacles and large amounts of transverse expansion to the flame path, and (2) small volumes without obstacles. DDT will not usually occur in a class 4 geometry.

Geometric Class 5. Geometries are so unfavorable to flame acceleration that not even large volumes of stoichiometric hydrogen-air mixtures are likely to detonate. Examples are a totally unconfined geometry at large scale or a small spherical geometry without obstacles and central ignition.

Table II gives the result class as a function of mixture and geometric classes. The entries in this table are based subjectively on investigations of highly experienced detonation physics specialists at SNL.[3]

TABLE II
DEPENDENCE OF RESULT CLASS ON MIXTURE AND GEOMETRIC CLASSES

Geometric Classes	Mixture Classes				
	1	2	3	4	5
1	1	1	2	3	4
2	1	2	3	4	5
3	2	3	3	4	5
4	3	4	4	5	5
5	4	5	5	5	5

The entries in Table II can be interpreted as follows:

Result Class 1: DDT is highly likely.
Result Class 2: DDT is likely.
Result Class 3: DDT may occur.
Result Class 4: DDT is possible but unlikely.
Result Class 5: DDT is highly unlikely to impossible.

We intend to use the framework of this methodology to evaluate the potential of detonation occurring in the dome vapor space. A matrix of experiments has been conducted[5] to determine the sensitivity of the major components (hydrogen, nitrous oxide, and nitrogen) of the waste gas diluted in air. The Shepherd interpretation of the Zel'dovich, von Neumann, and Döring (ZND) model[6] was verified for the purpose of interpolation and extrapolation of these experimental data. In these experiments and calculations, we have assumed a given waste gas release volume fraction composition of hydrogen (0.385), nitrous oxide (0.303), and nitrogen (0.312).[6] Using this information and the SNL methodology, we have classified the waste gas diluted with dry air into five sensitivity mixture categories, as summarized in Table III.

Table III includes the addition of ammonia to the waste gas mixture, but not the trace amounts of methane and carbon monoxide.[7] Both methane and carbon monoxide are relatively insensitive gases compared to hydrogen, and trace additions of the two are not expected to change the sensitivity of the waste gas mixtures to detonation.

Even though the detonation cell width was used to determine the mixture class, we observe in Table III that the hydrogen volume fraction or percentage serves the same purpose for defining the sensitivity of the mixture.

TABLE III
CLASSIFICATION OF CONSERVATIVE GAS RELEASE DILUTED WITH AIR (HYDROGEN, NITROUS OXIDE, AMMONIA, NITROGEN, AND WATER VAPOR)

Mixture Class	Waste Gas Volume Fraction	Air Volume Fraction	Hydrogen Volume Fraction	Detonation Cell Width (mm)
1	>0.45	<0.55	>0.144	<15
2	0.33 to 0.45	0.55 to 0.67	0.105 to 0.144	15 to 100
3	0.26 to 0.33	0.67 to 0.74	0.083 to 0.105	100 to 1000
4	0.22 to 0.26	0.74 to 0.78	0.072 to 0.083	1000 to 10000
5	<0.22	>0.78	< 0.072	>10000

Evaluation of Maximum Expected Release in the Tank Dome

We view the geometric class in the waste tank dome to be highly unfavorable to flame acceleration. The space is a large open volume of at least 40,000 ft^3 with few obstacles, and during a waste gas release, there are large amounts of volume for expansion. Experiments have shown[8] that the expansion of combustion waves inhibits flame acceleration. Therefore, the open dome space volume above the waste surface may be classified between a geometric class 4 and 5.

As the waste gases rise into the dome vapor space, they are quickly diluted with air to mixtures that are less sensitive. In fact, for the maximum expected waste gas release (ventilation system inoperable, riser closed), involving a volume of 297 m^3 (10,480 ft^3) at 1 bar and 300 K, we examine the fraction of the total tank volume (as a function of time), which contains accumulated volume fractions of mixture classes 1, 2, 3, and 4 (Fig. 1). The predicted mixture classes for this case resulted in no mixture classes 1 and 2. The presence of mixture class 3 was negligible. Therefore, in Fig. 1, the accumulated volume fractions of mixture classes are essentially mixture class 4.

Evaluation of Maximum Allowable Release in the Tank Dome

For the maximum allowable waste gas release involving a volume of 354 m^3 (12,500 ft^3) at 1 bar and 300 K, with an inoperable ventilation system and closed riser, we examine the fraction of the total tank volume (as a function of time), which contains accumulated volume fractions of mixture classes 1, 2, and 3 (Fig. 2). The predicted mixture classes for this case resulted in no mixture classes 1 and 2. Therefore, in Fig. 2, the accumulated volume fractions of mixture classes are solely a mixture class 3.

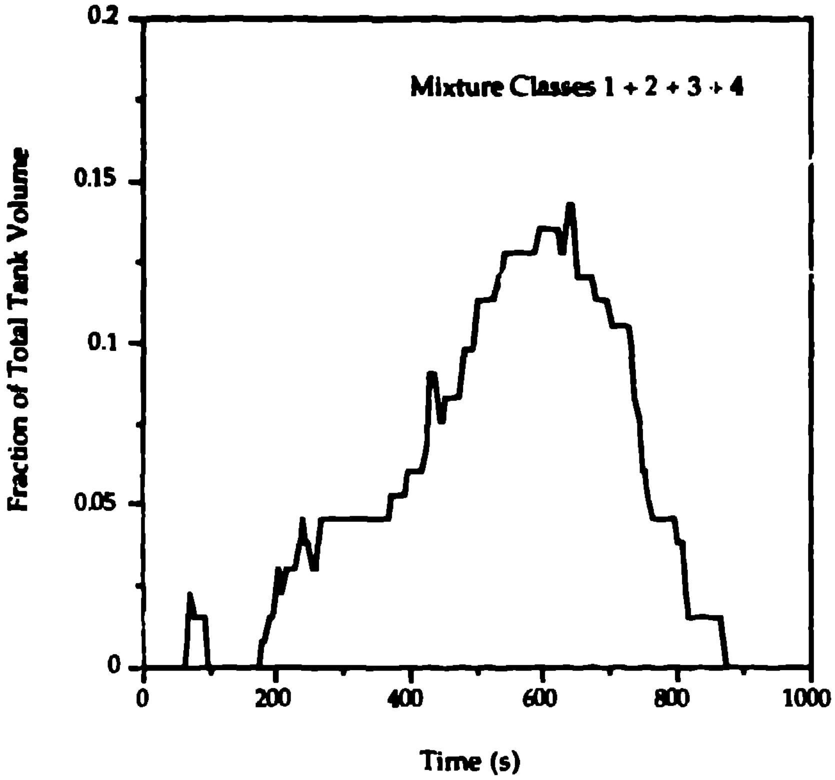

Fig. 1. Fraction of total tank dome space volume containing mixtures 1, 2, 3, and 4 for maximum expected release with inoperable ventilation system and closed riser.

Evaluation of Maximum Allowable Release in the Ventilation System

Combustible mixtures of waste gases are drawn through the ventilation system by the operating fan. To analyze the possibility of a detonation or DDT in the duct work of the ventilation system, we calculate the hydrogen volume fraction at the location where the ventilation system is attached to the dome. In Fig. 3, the hydrogen volume fraction time history is presented and compared to the threshold between mixture classes 4 and 5 (0.072 to 0.083). During the waste gas release phase, the maximum value remains within mixture class 4. Even though the ventilation system duct work (1-ft-diam pipe) is geometric class 2, the result class 4 indicates that detonation or DDT is possible but unlikely.

Summary of Results

Based on work done at SNL and the Explosion Dynamics Laboratory, we have quantified the relative risk of a DDT. The SNL methodology found two parameters to predict the likelihood of DDT. First, the closer a hydrogen-air mixture was to stoichiometric, the more likely the occurrence of a DDT. They listed five categories for mixtures that are related to detonation cell width and equivalence ratios. The second key parameter was the geometry. They found that confined geometries with

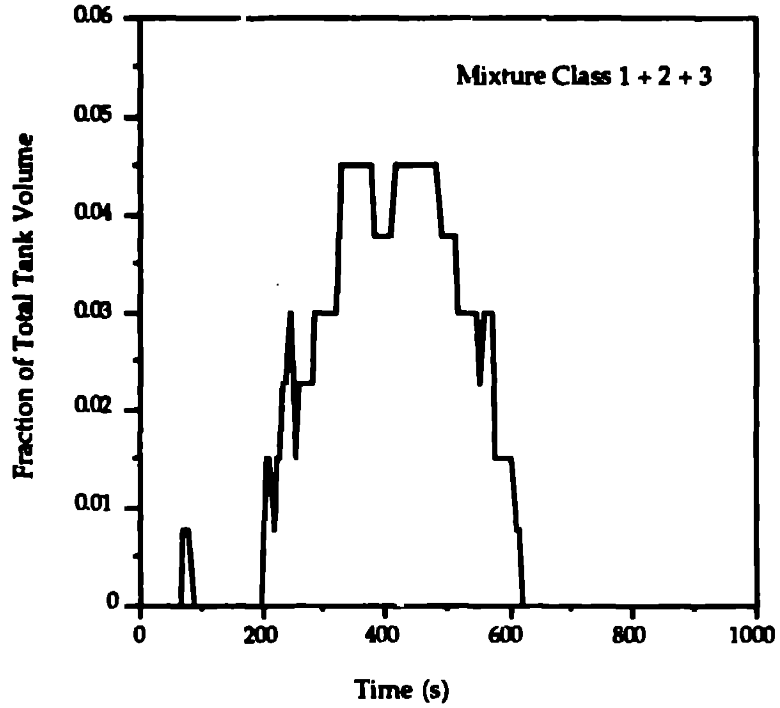

Fig. 2. Fraction of total tank dome space volume containing mixtures 1, 2, and 3 for maximum allowable release with inoperable ventilation system and closed riser.

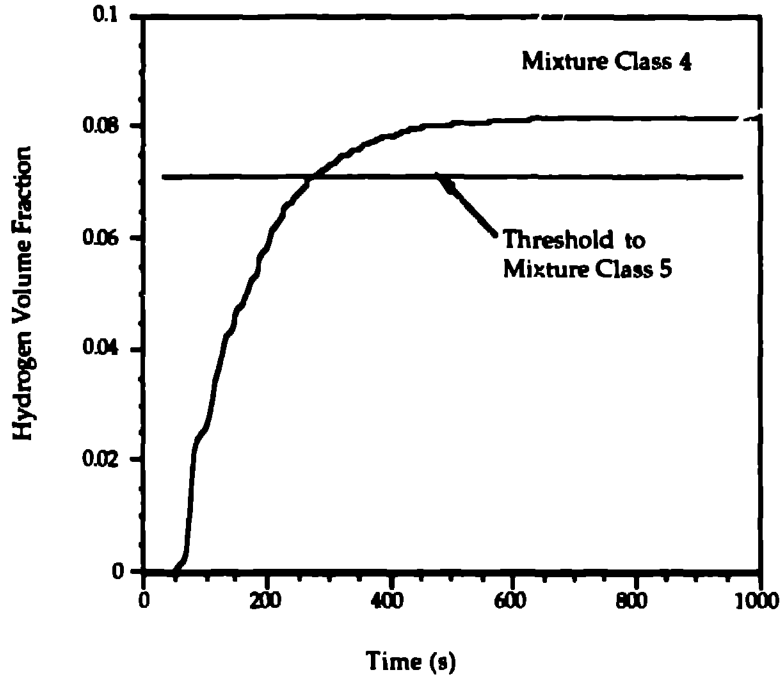

Fig. 3. Ventilation system hydrogen volume fraction for maximum allowable release with operable ventilation system and closed riser.

obstacles promoted flame acceleration and DDT. They had five classes for the geometric effects. The equivalence ratio can be quantitatively calculated, but the geometric effects are subjective. Based on these two parameters, SNL developed a matrix of the likelihood of DDT, as shown in Table II. We have updated the SNL methodology for hydrogen in dry air to mixtures of hydrogen, nitrous oxide, and nitrogen diluted with various percentages of air. The Explosion Dynamics Laboratory has obtained experimental data for this more prototypical waste gas composition. This data has been used to calibrate and verify the Shepherd ZND model. Using the Shepherd ZND model, we were able to interpolate and extrapolate the waste gas composition mixtures to determine the cell widths of each detonation and therefore determine the sensitivity of a given mixture. The waste gas mixture compositions, detonation cell size, and mixture classes are summarized in Table III.

For Tank SY-101, we calculated the volume percentage of the dome vapor space that would contain mixtures in the most sensitive classes for the maximum expected and maximum allowable release cases with the ventilation system inoperable and with the riser closed. In addition, we calculated the hydrogen volume fraction that would exist in the ventilation system with the ventilation system operable and the riser closed for a maximum allowable release. For the maximum expected release, we found (Fig. 1) that up to about 15% of the dome volume was mixture class 4, while mixture classes 1 + 2 + 3 were nearly negligible. This small percentage justifies the assumption that the geometry volume containing the sensitive gas mixtures is open and unrestricted. The geometry therefore is judged to be in the class of least likely to detonate. From result classes in II, the possibility of DDT is judged to be highly unlikely to impossible.

For the maximum allowable release, we found (Fig. 2) that none of the dome vapor space contains class 1 mixtures and that the dome volume containing the most sensitive mixtures (classes 1 + 2 + 3) is less than 5%. From these percentages, we conclude that because the geometry is open and the most sensitive mixtures are unconfined, the geometry is the least sensitive, with the lowest potential for DDT. Using the Table G-2 results, we judge the possibility of DDT to be highly unlikely to impossible.

The ventilation system was examined because it is a confined geometry and is judged to be class 2. An analysis of waste gas mixtures in the ventilation system shows that the hydrogen volume fraction, and therefore the detonation cell width, is never above class 4. From Table II, considering geometric class 2 and mixture class 4, the ventilation system has a result class 4, which characterizes the DDT probability as possible, but unlikely.

CONCLUSIONS

In the SNL studies, experimental data were obtained for dry hydrogen-air mixtures. In the waste gas mixture, $H_2/N_2O/N_2/NH_3$ diluted with air, the detonation cell width was measured at the Explosion Dynamics Laboratory. By using this data to calibrate the Shepherd ZND model, we were able to interpolate and extrapolate the relevant data to determine sensitivity mixture classes and relate them to the SNL methodology. Because the SNL geometric classes are directly applicable, we were able to judge the possibility of DDT based on the two relevant parameters of mixture sensitivity and geometry.

In the tank dome during a maximum expected burp or maximum allowable burp in a gas release event, the dome volume at any given time containing the most sensitive gas mixtures is negligible. This volume is considered unconfined, and because there are few obstacles in the dome volume, the volume also is considered to be open. An open unconfined volume of this size is in the least likely geometry class for DDT. When the gas mixtures are considered in this geometry, the result class indicates a highly unlikely to impossible, up to mixture class 3, event of DDT.

The long 1-ft-diam pipes making up the ventilation system are geometric class 2. However, because the sensitivity of the gas mixture during the release phase is never less than class 4, the result class is 4, which indicates DDT is possible, but unlikely.

Based on all these considerations, we conclude that DDT is possible but unlikely for the integrated waste tank dome vapor volume and ventilation system. This assessment of the likelihood of DDT occurring is based on the assumption that ignition has occurred with a probability of 1. New evidence[9] indicates that waste gas mixtures diluted with air are difficult to ignite with mechanically generated sparks. If we consider that the possibility of a mechanical spark ignition is difficult and that mechanical ignition unlikely, then the probability that detonation will occur is possible but very unlikely.

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