

Predicting Flammable Gas Mixtures in Hanford Double-Contained Receiver Tanks

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PREDICTING FLAMMABLE GAS MIXTURES IN HANFORD DOUBLE-CONTAINED RECEIVER TANKS

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ABSTRACT

This study presents a methodology to estimate the maximum concentrations of flammable gases (ammonia, hydrogen, and methane) which could exist in the vapor space of a double-contained receiver tank (DCRT). DCRTs are temporary storage tanks which receive highly radioactive liquid wastes from salt well pumping of Hanford single-shell tanks (SST). The methodology of this study could be used in other applications involving the storage and transfer of radioactive liquid wastes which generate or contain various dissolved flammable gases.

I. INTRODUCTION

The methodology presented here is used to determine the maximum levels of ammonia, hydrogen, and methane which could be expected in the dome space of a DCRT during and after the DCRT is filled to 80% of its total volume with liquid waste from an individual SST. Henry's Law constants, which are calculated based on several empirical models for the solubility of gases in liquid salt mixtures, are used to predict the amount of flammable gases which may be transported from a Hanford Site SST to a DCRT and released to a DCRT dome vapor space. In addition, hydrogen generation within the radioactive liquid waste in the DCRT is considered. The sources of hydrogen generation include radiolysis, thermal degradation of organic in the waste, and DCRT corrosion.

II. MODEL DESCRIPTION

The equilibrium concentrations of gases which exist between the vapor phase and aqueous solutions can be represented by the Henry's Law expression:

$$X = K_H P \quad (1)$$

where P is the partial pressure of a particular gas expressed in atmospheres, K_H is the Henry's Law constant for that gas, and X is the concentration of that gas in the aqueous solution. The Henry's Law constant for a gas depends on several variables, including temperature and the concentrations of the various ions in solution.

Of the models reviewed by Shekarriz et al.¹, the Schumpe model² gave the best agreement with experimental values from simulated waste. The Schumpe model is given by

$$\log(c_{G,0}/c_G) = \log \left(\frac{K_{H,G}(\text{water})}{K_{H,G}(\text{solution})} \right) = \sum_i (h_i + h_G) c_i \quad (2)$$

where $c_{G,0}$ and c_G denote the gas solubility of gas G in pure water and in a salt solution, respectively; $K_{H,G}(\text{water})$ and $K_{H,G}(\text{solution})$ are Henry's Law constants for soluble gas G in pure water and salt solutions, h_i and h_G are the ion and

gas-specific coefficients, and c_i is the concentration of ion "i" in the salt solution.

The gas-specific constant, h_G , is assumed to be a linear function of temperature:

$$h_G = h_{G,0} + h_T(T - 298.15^\circ\text{K}) \quad (3)$$

where $h_{G,0}$ is the reference value, and h_T is the temperature-specific coefficient.

Values of $h_{G,0}$ and h_T for ammonia, hydrogen, and methane are given in Table 1.

Table 1. Gas Specific Constants.³

| Gas | h_T | $h_{G,0}$ |
|----------|-----------|-----------|
| Ammonia | 0* | -0.0481 |
| Hydrogen | -2.99 E-4 | -0.0218 |
| Methane | -5.24 E-4 | 0.0022 |

* h_T for ammonia is set equal to zero since an experimentally determined value is not available.¹ For temperatures greater than 25 °C, if h_T were also negative, lower vapor pressures would be predicted. Therefore, setting h_T equal to zero is more conservative since higher ammonia vapor pressures would result.

The terms $\sum c_i$ and $\sum h_i c_i$ are calculated using ion concentrations obtained from a variety of sources and assumptions based on similar waste types. The values for the ion-specific constants h_i are presented in Table 2.

Table 2. Ion Specific Constants.³

| Ion | h_i |
|-------------------------------|--------|
| Na ⁺ | 0.1143 |
| Al ⁺ | 0.2174 |
| Fe ⁺ | 0.1161 |
| Cr ⁺ | 0.0648 |
| Ni ⁺ | 0.1654 |
| K ⁺ | 0.0922 |
| OH ⁻ | 0.0839 |
| NO ₃ ⁻ | 0.0128 |
| NO ₂ ⁻ | 0.0795 |
| CO ₃ ⁻² | 0.1423 |
| PO ₄ ⁻³ | 0.2119 |
| SO ₄ ⁻² | 0.1117 |
| F ⁻ | 0.0920 |
| Cl ⁻ | 0.0318 |
| Li ⁺ | .0754 |
| Br ⁻ | 0.0269 |

The various Henry's Law constants, $K_{H,G}(\text{solution})$, for each tank waste solution are obtained by dividing the appropriate temperature dependent value of $K_{H,G}(\text{water})$ calculated with Equation 5, 6, or 7 by its corresponding Schumpe ratio of $K_{H,G}(\text{water})/K_{H,G}(\text{solution})$.

The Henry's Law constants for the major flammable gas constituents ammonia, methane, and hydrogen are calculated for the waste in each tank considered.

The Henry's Law constant obtained through the above procedure must be converted from a molal basis, moles of solute per kg of solvent (water) in solution, to a basis of moles per volume of solution. The conversion is accomplished by calculating

$$K_H, L \text{ liquid waste basis} = (K_H, \text{kg water basis}) \rho_L \omega_L \quad (4)$$

where ω_L is the weight fraction of water in the solution and ρ_L is the solution density.

Equations for Henry's Law constants are reported for various gases including ammonia, hydrogen, and methane in water.⁴ The equation and its source for each gas at 1 atmosphere in equilibrium with water are given below. The equation for methane, which is incorrectly given in one reference,⁵ is correctly reported in another reference¹.

Ammonia⁵

$$K_{H,NH_3} [\text{mole/kg water-atm}] = \exp[-8.0964 + 3917.50/T - 0.00314 * T], T = ^\circ\text{K} \quad (5)$$

Hydrogen⁶

$$K_{H,H_2} [\text{mole/kg water-atm}] = \exp[-121.922 + 5528.45 / T + 16.8893 * \ln T], T = ^\circ\text{K} \quad (6)$$

Methane⁷

$$K_{H,CH_4} [\text{mole/kg water-atm}] = \exp[-412.1421 + 15557.56 / T + 65.2553 * \ln T - 0.06167 * T], T = ^\circ\text{K} \quad (7)$$

A. Calculation of Maximum Concentrations of Methane, Ammonia, and Hydrogen in a DCRT

Liquid from various SSTs is salt well pumped to their respective DCRTs. The following methodology is used to calculate the maximum concentrations of methane, ammonia, and hydrogen that can be expected in the vapor space of a DCRT during and after a tank is filled from 0% to a maximum of 80% of its volume.

Methane

The time to maximum methane concentration is the fill time of a DCRT from empty to 80% full. The % lower flammability limit (LFL) contribution from methane is obtained by dividing the calculated vapor space percent methane by 5.0% (100% LFL for methane⁸) and multiplying by 100%.

The percent methane (CH₄) for an equivalent volumetric flow rate of gaseous methane, (G_g), into a DCRT, a ventilation rate, (V_v), and a constant vapor space volume in the DCRT, (Volume), at time, (t), to fill the DCRT is given by Estey.⁹

$$\% \text{ CH}_4 = \frac{\text{Gr} * (1 - e^{-(\text{Gr} + \text{Vv}) * t / \text{Volume}})}{\text{Gr} + \text{Vv}} * 100\% \quad (8)$$

Ammonia

The mole % or volume % of ammonia in the dome space of a DCRT is calculated assuming that the partial pressure or concentration in the vapor space is always in equilibrium with the concentration in the liquid waste phase. This is obtained by dividing the concentration of ammonia in the liquid waste by the appropriate Henry's Law constant for ammonia. The % LFL contribution from ammonia is obtained by dividing the calculated vapor space percent ammonia by 15.5% (100% LFL for ammonia⁸) and multiplying by 100%.

Hydrogen

The mole % or volume % of hydrogen in the dome space of a DCRT is calculated based on the assumption that all of the hydrogen dissolved in the liquid waste entering the DCRT and all of the hydrogen generated in the DCRT are released instantaneously to a variable vapor space, which for the base case, varies from 100% to 20% of the volume of the DCRT as the tank is filled from empty to 80% full.

The calculation of DCRT headspace hydrogen volumes and concentrations are based on a numerical analysis technique for the solution of initial value ordinary (i.e., only one independent variable) differential equations. The independent variable is time. This technique uses a differential equation solver provided in the Professional Edition of Mathcad[®] V6.0 software. The baseline modeled situation is that an empty DCRT begins to fill at 15.14 L/m (4 gpm) until the tank is 80% full. The DCRT is provided with a positive ventilation of 85.0 L/h (3 ch). The waste being added to the DCRT carries with it dissolved hydrogen

which is immediately released on entering the tank. Additionally, the inventory of waste in the DCRT generates hydrogen due to radiolysis, chemical reactions, and corrosion of the wetted DCRT surfaces. The hydrogen generation rates used in the analysis are provided by the models documented by Hu.¹⁰ The dissolved hydrogen concentrations in the waste are calculated using the calculated Henry's Law constants discussed earlier and the Henry's Law expression.

The situation modeled in this analysis does not lend itself to solution via analytical techniques because the rate of hydrogen input to the DCRT headspace and the DCRT headspace volume vary as functions of time. In this case the differential equation is written:

$$d(\text{H}_2 \text{ volume})/dt = f_1(t) - [f_2(t)/f_3(t)] * (\text{H}_2 \text{ volume}) \quad (9)$$

where

$f_1(t)$ = H₂ generation rate

$f_2(t)$ = tank exhaust rate = [tank ventilation rate + liquid volume addition rate + $f_3(t)$]

$f_3(t)$ = tank headspace volume

In this analysis, the units of the independent variable t are hours. The time period of interest is zero hours to the time at which the DCRT is 80% full at the given DCRT fill rate. The solution evaluates the value of $f_1(t)$ at a given time step using the hydrogen generation models and including the dissolved hydrogen release rate associated with the DCRT fill rate. The functions $f_2(t)$ and $f_3(t)$ are calculated as the fill rate continually reduces the volume of the DCRT headspace. The differential equation solver is then used to estimate the volume of H₂ in the DCRT headspace at the time of interest and the corresponding value of $d(\text{H}_2 \text{ volume})/dt$ is calculated by substituting these results into the differential equation.

The % LFL contribution from hydrogen is obtained by dividing the calculated vapor space percent hydrogen by 4% (100% LFL for hydrogen⁸) and multiplying by 100%.

B. Total Percent LFL from Ammonia, Methane, and Hydrogen in DCRT Vapor Space

The total dome space % LFL is obtained by adding the respective % LFL contributions of methane, ammonia, and hydrogen. However, the total dome space % LFL for the steady state case results from considering only the steady state ammonia concentration in the dome space, the hydrogen generation within the DCRT, and the ventilation rate.

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III. ASSUMPTIONS

These calculations of expected maximum concentrations of flammable gases in a DCRT are based on a scenario in which liquid wastes are transferred from an individual SST to an empty DCRT. The base case assumptions are:

1. The liquid waste being transferred into the DCRT contains the dissolved gases hydrogen and methane which are in equilibrium with a 97% hydrogen and 3% methane gaseous mixture. The pressure used in the calculations was the average gas pressure calculated for gas trapped in the waste.¹¹ The conservative value of 97% hydrogen is based on a hydrogen release in tank 241-105-AN that may have exceeded 90%.¹² The 3% methane value results from assuming a gas mixture in the waste of only hydrogen and methane.
2. Because hydrogen and methane are very insoluble, it is assumed that all of the methane and hydrogen are released to the vapor space as soon as they enter the DCRT.
3. The DCRT ventilation rate is set equal to 3 cfm based on the assumption that two of the bubbler tubes used for level measurements provide a total air flow of at least 3 cfm.
4. A liquid waste fill rate of 4 gpm is assumed.
5. Because ammonia is a soluble gas it is not reasonable to assume that 100% of the ammonia is released. Therefore, it is assumed that the DCRT vapor space is to continuously have a concentration of ammonia which is in equilibrium with the ammonia in the liquid waste entering the DCRT.
6. The total % LFL of flammable gases in a DCRT is the sum of the individual % LFL contributions of hydrogen, methane, and ammonia. These are the significant flammable gases in SSTs.
7. The transport of hydrogen via bubbles entrained in liquid being pumped into a DCRT is assumed to be small. Based on the conservative assumption that 100% of the soluble hydrogen in an SST is transported to a DCRT, it is assumed that no hydrogen is transported from the SST to a DCRT via bubbles.
8. One of the alternative assumptions used is a ventilation rate of 5 cfm. This is based on information for tank waste operation that the weight factor bubbler can be operated at maximum flow rates of 2.6 to 2.8 cfm.

IV. RESULTS

The results of the analyses performed are presented in Table 3. They are based on the assumptions made in these analyses. If the assumptions are changed, specific results may change. As shown in Table 3, the predicted maximum concentrations of flammable gases in the dome space of a DCRT are below the 25% LFL limit for all of the tanks considered with the exception of tanks 13, 15, and 22.

However, operational controls can be used to reduce the potential flammable gas concentrations in these tanks further as can be seen in Table 3 as Alternate Cases 1, 2, and 3. These results are given only for tanks that have predicted total (% LFL) values exceeding 20% for their respective base cases. The methodology of this study could be used in other applications involving the storage and transfer of radioactive liquid wastes which generate or contain various dissolved flammable gases. A more detailed presentation of the application of this methodology is given by Hedengren et al.¹³

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Table 3. Concentrations of Hydrogen in Dome Space and Total DCRT Dome Space % LFL for Hydrogen, Methane, and Ammonia.

| Tank | DCRT | Base Case 80% Full 4 gpm 3 cfh | | | | Alternative Case 1 80% Full 4 gpm 5 cfh | | | | Alternative Case 2 50% Full 4 gpm 5 cfh | | | | Alternative Case 3 50% Full 4 gpm 3 cfh | | | |
|------|------|---|---------------|------------|---------------|--|---------------|------------|---------------|--|---------------|------------|---------------|--|---------------|------------|---------------|
| | | H2 %LFL | Total %LFL | H2 %LFL | Total %LFL | H2 %LFL | Total %LFL | H2 %LFL | Total %LFL | H2 %LFL | Total %LFL | H2 %LFL | Total %LFL | H2 %LFL | Total %LFL | H2 %LFL | Total %LFL |
| 1 | EX | 3.33 | 10.26 | 4.39 | 11.21 | | | | | | | | | | | | |
| 2 | EX | 9.28 | 14.91 | 4.15 | 9.43 | | | | | | | | | | | | |
| 3 | EX | 13.10 | 22.85 | 6.48 | 15.70 | 12.50 | 22.25 | 3.89 | 13.11 | 5.66 | 15.41 | 2.67 | 11.89 | 5.78 | 15.53 | 4.45 | 13.67 |
| 4 | S | 3.77 | 8.09 | 4.38 | 8.60 | | | | | | | | | | | | |
| 5 | S | 3.17 | 22.32 | 5.32 | 24.39 | 3.02 | 22.17 | 3.20 | 22.27 | 1.34 | 20.49 | 2.16 | 21.23 | 1.37 | 20.52 | 3.59 | 22.66 |
| 6 | S | 1.94 | 10.67 | 5.96 | 14.65 | | | | | | | | | | | | |
| 7 | S | 3.08 | 4.72 | 3.24 | 4.79 | | | | | | | | | | | | |
| 8 | S | 8.72 | 14.01 | 3.19 | 8.16 | | | | | | | | | | | | |
| 9 | S | 19.00 | 20.22 | 3.10 | 3.31 | 18.20 | 19.42 | 1.86 | 2.07 | 8.52 | 9.74 | 1.30 | 1.51 | 8.70 | 9.92 | 2.17 | 2.38 |
| 10 | S | 4.18 | 11.66 | 3.43 | 10.77 | | | | | | | | | | | | |
| 11 | S | 2.96 | 10.48 | 5.52 | 12.96 | | | | | | | | | | | | |
| 12 | S | 5.25 | 7.82 | 11.80 | 14.24 | | | | | | | | | | | | |
| 13 | S | 8.70 | 13.06 | 30.50 | 34.67 | 8.33 | 12.69 | 18.40 | 22.57 | 3.38 | 7.74 | 11.80 | 15.97 | 3.45 | 7.81 | 19.60 | 23.77 |
| 14 | S | 7.09 | 10.70 | 20.90 | 24.55 | 6.78 | 10.59 | 12.60 | 16.05 | 2.83 | 6.44 | 8.11 | 11.56 | 2.89 | 6.50 | 13.50 | 16.95 |
| 15 | S | 9.60 | 14.36 | 36.70 | 41.26 | 9.19 | 13.95 | 22.20 | 26.76 | 3.67 | 8.43 | 14.20 | 18.76 | 3.75 | 8.51 | 23.60 | 28.16 |
| 16 | S | 4.07 | 5.21 | 6.34 | 7.37 | | | | | | | | | | | | |
| 17 | TX | 14.60 | 18.79 | 3.38 | 4.15 | | | | | | | | | | | | |
| 18 | TX | 0.35 | 1.00 | 3.38 | 4.03 | | | | | | | | | | | | |
| 19 | U | 5.20 | 11.77 | 8.17 | 14.55 | | | | | | | | | | | | |
| 20 | U | 3.07 | 14.16 | 12.70 | 23.74 | 2.93 | 14.02 | 7.66 | 18.70 | 1.14 | 12.23 | 5.02 | 16.06 | 1.16 | 12.25 | 8.36 | 19.40 |
| 21 | U | 5.55 | 12.78 | 12.80 | 19.87 | | | | | | | | | | | | |
| 22 | U | 4.32 | 12.29 | 23.00 | 30.90 | 4.14 | 12.11 | 13.80 | 21.70 | 1.51 | 9.48 | 8.91 | 16.81 | 1.54 | 9.51 | 14.80 | 22.70 |
| 23 | U | 5.01 | 10.15 | 6.59 | 11.56 | | | | | | | | | | | | |
| 24 | U | 2.17 | 17.88 | 9.28 | 24.95 | 2.07 | 17.78 | 5.58 | 21.25 | 0.80 | 16.51 | 3.72 | 19.39 | 0.80 | 16.51 | 6.19 | 21.86 |
| 25 | U | 3.95 | 11.55 | 8.99 | 16.47 | | | | | | | | | | | | |
| 26 | U | 2.94 | 11.70 | 9.14 | 17.83 | | | | | | | | | | | | |

Notes:

DCRT: S - 15 ft height, vertical

DCRT: EX, TX, U - 12 ft diameter, 35 ft length, horizontal

BOLD font implies the total % LFL is greater than 25%.

*For tank 17, because it has been recently salt well pumped, it is assumed that for additional salt well pumping, there will be a 4 gpm flow for 25 minutes followed by a 0.5 gpm flow to fill the DCRT to 80% of capacity. It is also assumed that the solution being transferred during the 0.5 gpm period has the dissolved hydrogen concentration reduced by 50% due to low pressure on the solution at this time.

**For tank 18, the liquid salt well pumped is from a waste surface pool which is in equilibrium with the steady state dome space concentrations of hydrogen and methane (<25 ppmv). Therefore, the waste surface pool is essentially free of dissolved hydrogen and methane.

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