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# Equilibrium Hydrogen Concentrations of the 800 Series Tanks in Deactivation Mode

By

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**Abstract**

The objective of this analysis is to evaluate the steady state equilibrium hydrogen concentration in the 800 Series tanks of the F-Area of Savannah River Site in their deactivation mode. These tanks (Tank 804, Tank 808 and Tank 809) are underground tanks and each of them is housed in a cell. Currently, these tanks still contain sludge heels with plutonium and other radionucludes that generate hydrogen by radiolysis. In the deactivation mode, the ventilation system will be completely shut down. In order to vent the hydrogen to the surrounding, the manway hole at the top of the tank and the plug hole on the top of the cell must be kept open. To prevent the introduction of foreign object into the cell, a filter panel will be used to cover the plug hole. The present analysis uses a diffusion model to perform a series of parametric studies to evaluate the steady state hydrogen concentration in the cell and the tank as a function of size and geometry of the filter panel. The results of the analysis serves as a basis for the design of the filter by enabling the determination of the appropriate size of the filter to ensure the hydrogen concentration in the tank stay below 25% of the Lower Flammability Limit (LFL) with no active ventilation.

## Introduction

The 800 Series Tanks (Tanks 804, 808, and 809) are underground tanks in the F-Area of the Savannah River Site. Each of these tanks is housed in a separate cell. Currently, these tanks still contain sludge heels with plutonium and other radionuclides that generate hydrogen gas by radiolysis. In the deactivation mode, the ventilation system will be completely shut down. In order to vent the hydrogen to the surrounding, the manway hole at the top of the tank and the plug hole on the top the cell must be kept open. To prevent the inadvertent introduction of objects into the cell, a filter panel is used to cover the plug hole. The purpose of the present analysis is to perform an evaluation of the steady state equilibrium hydrogen concentrations in the 800 Series Tanks during the deactivation mode.

The physical dimensions of the tanks and the cells are given in Table 1 and their nuclide inventories and the hydrogen generation rates are given in Table 2.

**Table 1 Tank and Cell Dimensions[1]**

| | Tank 804 | Tanks 808/809 |
|---|---|---|
| Tank orientation | Vertical | Horizontal |
| Tank height | 11 ft | 36 ft |
| Tank diameter | 10 ft | 9 ft |
| Manway hole diameter | 20 inches (min) | 20 inches |
| Manway hole height | 6 inches | 6 7/8 inches |
| Cell length | 21 ft | 44 ft |
| Cell width | 13 ft | 12 ft |
| Cell height | 32 ft 3 inches | 32 ft |
| Plug length | 1 ft 11 ½ inches (min) | 1 ft 11 ½ inches |
| Plug width | 2 ft 5 ½ inches (min) | 2 ft 5 ½ inches |
| Plug height | 2 ft | 2 ft |

Note: Cell plug is tapered. The length and width of the plug used here are their minimum values.

**Table 2 Nuclide Inventory and Hydrogen Generation Rates in Tanks**

| | Nuclide Inventory in Heel[1] | H2 Generation Rate[1] |
|---|---|---|
| Tank 804 | 1,530 g Pu | $6.15 \times 10^{-6}$ ft$^3$/s |
| Tank 808 | 317 g Pu | $1.10 \times 10^{-6}$ ft$^3$/s |
| Tank 809 | 92.4 g Pu | $4.13 \times 10^{-7}$ ft$^3$/s |

**Analytical Model Used for Analysis**

In the absence of any forced ventilation flow in the deactivation mode, the transport of hydrogen from the tank to the ambient is controlled by the diffusion process, which can be described by Fick's Law[2]. At equilibrium, the molar rate of hydrogen diffused through the manway hole and its filter are:

$$J = J_1 = D\frac{C_1 - C^*}{L_1 / A_1} \tag{1}$$

$$J = J_2 = D_{f1}\frac{C^* - C_2}{L_{f1} / A_{f1}} \tag{2}$$

where

$J$ = hydrogen diffusion rate (mole/s)

$C_1$ = hydrogen concentration in tank (mole/m$^3$)

$C^*$ = hydrogen concentration at the filter inlet (mole/m$^3$)

$C_2$ = hydrogen concentration in cell (mole/m$^3$)

$D$ = hydrogen diffusivity in air (m$^2$/s)
$D_{f1}$ = “material” diffusivity of the filter (m$^2$/s)
$L_1$ = length of the manway hole (m)
$A_1$ = area of the manhole hole (m$^2$)
$L_{f1}$ = length of the filter (m)
$A_{f1}$ = area of the filter (m$^2$)

Equations 1 and 2 can be written as:

$$C_1 - C^* = J\left(\frac{L_1 / A_1}{D}\right) = J\,R_1 \tag{3}$$

$$C^* - C_2 = J\left(\frac{L_{f1} / A_{f1}}{D_{f1}}\right) = J\,R_{f1} \tag{4}$$

where $R$ is the diffusion “resistance”. Subtracting Eq. 4 from Eq. 3 leads to:

$$C_1 - C_2 = J\,(R_1 + R_{f1}) = J\,R_1^* \ . \tag{5}$$

By the same token, it can be shown that the concentration difference between the cell and the ambient is:

$$C_2 - C_0 = J_D\,(R_2 + R_{f2}) = J_D\,R_2^* \ , \tag{6}$$

where $R_1^*$ and $R_2^*$ are the total resistances of the manway hole and the cell plug. If the “breathing” effect is considered, the rate of transfer of hydrogen by the convective purge is given by:

$$J_C = Q_B (C_2 - C_0) \tag{7}$$

where $Q_B$ is the breathing rate, which is equal to 0.5% of the net cell volume per day:

$$Q_B = 0.005 \times (V_{cell} - V_{\tan k}) / (24 \times 3600) \, . \tag{8}$$

Combining Equations 6 and 7 leads to:

$$J = J_D + J_C = (C_2 - C_0)\left[\frac{1}{R_2^*} + Q_B\right] \tag{9}$$

or
$$C_2 - C_0 = J\left[\frac{1}{R_2^*} + Q_B\right]^{-1} \tag{10}$$

Subracting Eq. 10 from Eq. 5 results in:

$$C_1 - C_0 = J\left[R_1^* + \left(\frac{1}{R_2^*} + Q_B\right)^{-1}\right] \tag{11}$$

Since the hydrogen concentration in the ambient is basically zero, Equation 11 can be written as:

$$C_1 = J\left[R_1^* + R_2^*\left(\frac{1}{1 + R_2^* Q_B}\right)\right] , \tag{12}$$

which is the equilibrium hydrogen concentration in the tank. The molar concentration in the cell can be calculated by using Eq. 10 by setting $C_0$ to zero; i.e.,

$$C_2 = \frac{J}{\left(\frac{1}{R_2^*} + Q_B\right)} . \tag{13}$$

The hydrogen concentration can also be expressed in fraction of LFL, i.e.,

$$C' = \frac{C}{C_{LFL}} = \frac{C}{X_{LFL}\left(\frac{P}{\bar{R}T}\right)} , \tag{14}$$

where $X_{LFL}$ is the volume fraction of hydrogen at LFL, which is equal to 4%. Notice that the diffusivity used in this analysis is the effective diffusivity of hydrogen in the filter material (called "material diffusivity" in this analysis). For generality, the formulation includes the filters for both the manway hole and the cell plug. In actual calculations, only the filter of the cell plug is used.

From Reference 2, it is also known that diffusivity in gas has a 1.5 power dependence on temperature (in K). The hydrogen diffusivity in air is based on 0°C (273.15 K) and the evaluation of hydrogen diffusivity in the filters is based on the temperature of 25°C. Since this analysis assumes a temperature of 15°C for the vapor space of the tanks and the cells, all diffusivities are corrected to the assumed ambient temperature accordingly. There is no evidence that the hydrogen diffusivity in the filter media has the similar dependence on the temperature. However, the assumed temperature of 15°C is only slightly lower than 25°C (on the absolute temperature scale) and applying the temperature correction for the hydrogen diffusivity in the filter media is conservative.

This aforementioned methodology is applied to Tanks 804, 808 and 809.

## Results of Analysis

The results of calculation for Tank 804 are summarized in Table 3, which tabulates the steady state hydrogen equilibrium concentrations in the tank (in % LFL) for various combinations of filter height (thickness) and filter-to-plug area ratio. For these calculations, a material diffusivity of 0.3 x $10^{-5}$ $m^2/s$ was chosen to represent the metal type filter. The manway hole is assumed to be open and the cell plug is covered with filter(s). In addition, breathing effect is not included. From Table 3, it can be seen that the equilibrium concentration in the tank increases with the filter height (thickness) and decreases with the filter-to-cell plug area ratio. For the extreme case of no filter (i.e., zero inch filter height), the equilibrium hydrogen concentration is 13.9%. If the breathing effect is included for the no filter case, the equilibrium hydrogen concentration would be further reduced to 12.0% (last case of Table 3). With a filter-to-cell plug area ratio of one (i.e., filter area equal to that of the plug hole, or 0.448 $m^2$), the equilibrium hydrogen concentration is below 25% LFL for filter heights up to one inch. However, equilibrium hydrogen concentration increases rapidly as the filter-to-plug area ratio decreases. To keep the hydrogen equilibrium concentration below 25% LFL, the deactivated Tank 804 with metal based filter must be operated in a configuration that is within the unshaded region of Table 3.

Similar calculations have been performed for Tanks 808 and 809. It has been found that the steady state equilibrium hydrogen concentrations for these tanks are significantly lower than that of Tank 804. This is expected because of their very low nuclide inventories in the heels. In fact, the hydrogen concentrations are so low such that only the breathing effect without the plug holes open is sufficient to keep them below the 25% LFL level.

**Table 3 Equilibrium Concentrations for Tank 804 with Metal Filter (in %LFL)**

| **Filter Height (inches)** | **Filter to Cell Plug Area Ratio** | | | | | |
|---|---|---|---|---|---|---|
| | 0.25 | 0.5 | 0.75 | 1 | 2 | 4 |
| 0 | 13.92 | 13.92 | 13.92 | 13.92 | 13.92 | 13.92 |
| 0.1 | 17.38 | 15.65 | 15.07 | 14.78 | 14.35 | 14.13 |
| 0.2 | 20.84 | 17.38 | 16.23 | 15.65 | 14.78 | 14.35 |
| 0.4 | 27.77 | 20.84 | 18.53 | 17.38 | 15.65 | 14.78 |
| 0.6 | 34.70 | 24.31 | 20.84 | 19.11 | 16.51 | 15.22 |
| 0.8 | 41.63 | 27.77 | 23.15 | 20.84 | 17.38 | 15.65 |
| 1.0 | 48.56 | 31.24 | 25.46 | 22.58 | 18.25 | 16.08 |
| | | | | | | |
| No Filter Breathing Only | 12.02 | | | | | |

**Evaluation of Hydrogen Diffusivity for a Panel of Metal Filters**

The term “material diffusivity” for the filter media used in this analysis is what many text books or handbooks call “diffusivity”, which is an intrinsic property of a substance that has no dependence on geometry (i.e., like thermal conductivity). The reason it is called “material diffusivity” in this EC is to distinguish it from the “diffusivity” quoted by the filter vendors (which is called “filter diffusivity” in this analysis).

For the filter vendors, it is reasonable to test their filters and report the filter diffusivities for each individual filter. The “filter” diffusivities implicitly include the geometric effects of a filter.

In the present case, the plug hole (1 ft 11 ½ in by 2 ft 5 ½ in) is much larger than any off-the-shelf filter that can be acquired commercially. Also, fabricating a filter specifically for the 800 Series tanks deactivation may not be exactly economical. Therefore, it is reasonable to use a filter panel that is made up of many individual filters that are commercially available. As a result, the “material” diffusivity can be used to “scale” the diffusivity for a filter panel system of different sizes.

From the data sheet of the carbon based filter NucFil®016 SS HP[3], it is reported that it has a “diffusivity” of 1.65 x $10^{-4}$ mole/s per mol fraction at 25°C. By definition, the rate of hydrogen molecule diffused from a confined space to the ambient through the filter is given by:

$$J_N = \hat{D}\frac{\Delta C_H}{C_A} = \left(\frac{\hat{D}}{C_A}\right)C_H \ , \qquad (15)$$

where $J_N$ = molar diffusion rate (mole/s)

$\hat{D}$ = filter diffusivity = 1.65 x $10^{-4}$ mole/s per mol fraction (of hydrogen)

$C_H$ = hydrogen concentration in the confined space (mole/$m^3$)

$C_A$ = hydrogen/air total molar concentration (mole/$m^3$).

Note that the filter diffusivity has no explicit geometric dependence on the filter and the diffusivity is filter specific implying the geometric effects of the filter have already been implicitly embedded in the filter diffusivity.

The rate of diffusion can also be expressed by the Fick’s Law of Diffusion, which has the following form:

$$J_N = D\,A\frac{\Delta C_H}{L} = \left(\frac{D\,A}{L}\right)C_H \ , \qquad (16)$$

where $D$ = “material” diffusivity ($m^2/s$) of the filter
$A$ = diffusion area ($m^2$)
$L$ = diffusion length (m).

Equating Equations 15 and 16, the relationship between the “filter diffusivity” and the “material diffusivity” is:

$$\frac{\hat{D}}{C_A} = D\frac{A}{L} \quad \text{or} \quad \hat{D} = \left(\frac{D\,A}{L}\right)C_A \ , \qquad (17)$$

where the air-$H_2$ molar concentration $C_A$ can be shown to be 40.87 air-$H_2$ mole/$m^3$. Also, the present analysis is based on a material diffusivity (D) of 0.3 x $10^{-5}$ $m^2/s$ for the metal based filter. The value of the media thickness L can be determined if the thickness of the material used for the filter panel is the same as that of NucFil®016 SS HP (i.e., 0.07 in or 1.778 x $10^{-3}$ m).

From the input, it can be readily shown that the nominal area for the plug hole is 4.82 $ft^2$ (or 0.448 $m^2$). Since the filter panel is made by “bundling” a large number of small filters, the effective diffusion area of the filter media would be somewhat less than the actual physical area of the plug area. For conservatism, this EC recommends the use of at least a 50% area ratio. Substituting these values into Equation 17, the effective diffusivity of the filter panels is:

$$\begin{aligned}\hat{D} &= \frac{(0.3\times10^{-5}\,m^2/s)(0.5)(0.448m^2)}{(1.778\times10^{-3}\,m)}\left(40.87\ \frac{mole\ air-H_2}{m^3}\right) \\ &= 1.55\times10^{-2}\ mole/s\ \ per\ mol\ \ fraction.\end{aligned} \qquad (18)$$

Due to the relatively large anticipated size of the filter panel, it is possible that some form of structural support may be necessary. Therefore, the effective thickness of the filter panel may not be exactly equal to 0.07 inches. However, Table 3 shows that with a filter thickness of 0.1 inches, the equilibrium hydrogen concentration is only 15.7% LFL, which is still significantly less than 25% LFL

Since it is known that the filter diffusivity of NucFil®016 SS HP is 1.65 x $10^{-4}$ mol/s per mol fraction, it can be shown by comparison that diffusivity of the filter panel is about 94 times of that of a single NucFil®016 SS HP filter. In other words, the filter panel requires a minimum of 94 NuclFil®016 SS HP filters to maintain an equilibrium hydrogen concentration significantly below 25% LFL (i.e., 15.7% LFL).

In the present design, the plug hole is covered by a “dog house” made up of four panels, each of which is made up of 30 NuclFil®016 SS HP filters arranged in a 5 x 6 array. With a diameter of 2 inches (5.08 x $10^{-2}$ m) the total diffusion area for 120 filters in parallel would be approximately 0.24 $m^2$. This gives a filter-to-plug hole area ratio of about 53%. With more filters and greater effective filter diffusivity, it is expected that the equilibrium hydrogen concentration of Tank 804 should stay well within the 25% LFL level.

## Conclusions

An analytical model has been developed to parametrically evaluate the steady state equilibrium hydrogen concentrations in the 800 Series Tanks. It has been shown that it is possible to keep the steady state equilibrium hydrogen concentrations below 25% LFL in the tanks and their cells with no active ventilation. The results of the analysis are used as a tool for the design of the filter panel placed on the plug hole of the 800 Series Tanks. With some modifications, this methodology could also be extended to other sites across the DOE-Complex for evaluation of hydrogen concentration in any process equipment or building in the absence of ventilation.

## References

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2. J. P. Holman, "Heat Transfer," McGraw-Hill Company, 1992.
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