Hydrogen Gas Generation Model for Fuel-Based Remote-Handled Transuranic Waste Stored at the INEEL

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HYDROGEN GAS GENERATION MODEL FOR FUEL-BASED REMOTE-HANDLED TRANSURANIC WASTE STORED AT THE INEEL

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ABSTRACT

The Idaho National Environmental and Engineering Laboratory (INEEL) initiated efforts to calculate the hydrogen gas generation in remote-handled transuranic (RH-TRU) containers in order to evaluate continued storage of unvented RH-TRU containers in vaults and to identify any potential problems during retrieval and aboveground storage. A computer code is developed to calculate the hydrogen concentration in the stored RH-TRU waste drums for known configuration, waste matrix, and radionuclide inventories as a function of time.

INTRODUCTION

From 1976 through 1995, Argonne National Laboratory-East (ANL-E) shipped 617 30-gallon drums of RH-TRU waste for storage in the Intermediate Level Transuranic Storage Facility vaults at the INEEL's Radioactive Waste Management Complex. The waste was generated as a result of destructive examinations of irradiated fuel elements at the ANL-E Alpha-Gamma Hot Cell Facility. The majority of fuel elements were irradiated in Experimental Breeder Reactor II at Argonne National Laboratory-West (ANL-W). Out of 617 drums, about 430 drums were shipped without any venting mechanism and are currently stored in underground vaults. The Department of Energy order issued in 1999 requires that vents or other mechanisms to prevent pressurization of containers must be installed on currently stored transuranic waste as soon as practical, unless analysis demonstrates that the waste can otherwise be stored safely.

To address the DOE Order 435.1 requirements, the INEEL developed a gas-generation computer model to calculate decay heat, hydrogen gas generation, and hydrogen concentration in different layers of the ANL-E RH-TRU waste drums as a function of time. Several sites have developed hydrogen generation models or performed analysis to suit their need and waste streams. However, those analyses or models cannot be applied directly to the INEEL ANL-E waste stream. Computer code RadCalc 3.0 (1), which is used in transportation analysis for transport of waste in a CNS-10-160B cask, estimates hydrogen generation in a single layer without any leakage. This paper discusses the development of a computer code to estimate time-dependent hydrogen concentration in a multi-layer packaging system. The computer code is generic, such that with minor modifications it can be applied to other waste streams.

WASTE PACKAGING CONFIGURATION

Following is a brief description of the typical ANL-E waste packaging configuration (2), see Figure 1:

- Waste is placed into two 7.5-gallon waste cans, without a gasket in the can lid.

- The pails are placed into a 90-mil lidless fiberboard liner.

- The liner and pails are placed inside a 20-mil polyvinylchloride (PVC) bag that is heat-sealed closed.

- This package is placed into a 100-mil lidless polyethylene (PE) liner.

- The PE liner and contents are placed inside a 20-mil PVC bag that is heat-sealed closed.

- This package is placed into a 30-gallon drum (DOT 7A, Type A). The drum lid-gasket is 3/8-in. OD x 7/32-in. ID styrene-butadiene rubber.

About 187 of the 617 30-gallon drums (DOT 7A, Type A) are vented with an NF-013 carbon composite drum filter in the drum lid. Of these 187 drums, about 121 contained PVC bags vented with an NF-030 carbon composite filter and 66 contained heat-sealed bags, with no filters. The remaining 430 drums contained no filters in the bag or in the drum lid.
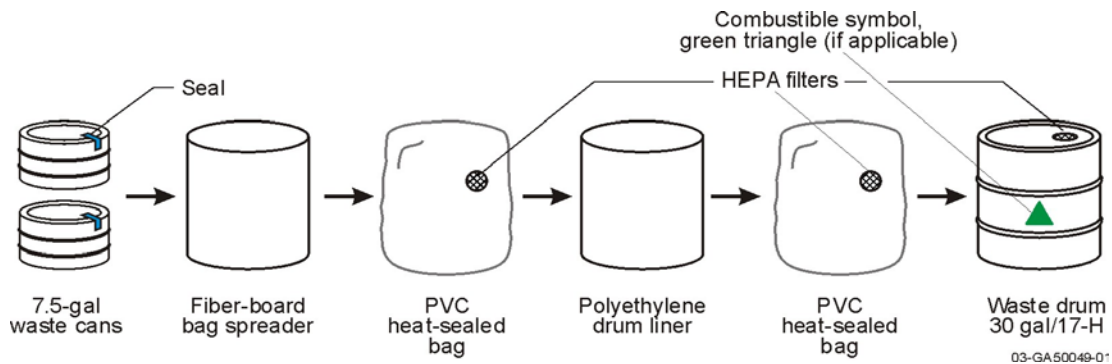

Figure 1. Schematic of 30-gallon drum packaging.

HYDROGEN GAS GENERATION

Hydrogen is generated primarily by radiolysis of hydrogenous materials. In the RH-TRU 72-B Waste Shipping Package Safety Analysis report (72-B Shipping Package SAR) (3), an equation defines the rate of hydrogen generation as

$$n_{H_2,gen} = C \sum_i [F_i G_i (DH)_i] \tag{1}$$

where

C	=	conversion constant = 1.04×10^{-7} (g-mol) (100 eV)/(molecule)(W-s)
F_i	=	fraction of emitted energy from type i radiation absorbed in waste material
G_i	=	number of molecules of hydrogen produced per 100 eV of energy absorbed from type i radiation

DH$_i$ = decay heat (W) of type i radiation.

The methodologies for determining the value of each parameter in Equation (1) are described below.

DECAY HEAT AND DOSE RATE BY RADIATION TYPE

Ten waste drums, 728 through 737, generated during August 27, 1990 through July 17,1991, were selected on which to demonstrate the estimation methodology. The radioisotopic concentrations in the ten drums containing RH-TRU were estimated based on all fuel elements destructively examined during the waste campaign August 28, 1990 through July 17, 1991 (2,4). The radioactive waste is characteristically swarf from irradiated fuel, with an average 7% (atom%) burnup, and an average decay period of 9–12 months after end of irradiation but before being packed into a drum. The ORIGEN2 computer code (5) was used to estimate decay of the inventory and decay heat rates of the waste as a function of time. The total activity inventory as of November 1991 in the ten drums was estimated to be 37.5 Ci, or 3.75 Ci per drum (4).

Based on the average heat generation rates obtained from the ORIGEN2 analyses for each type of radiation, the following equations were developed to estimate the average decay heat rates by radiation type as a function of time, t, per curie of waste at the time of waste drum closure:

$$(DH)_\alpha = 5.169*10^{-4} + 8.53*10^{-6}*t - 1.21*10^{-7}*t^2 \tag{2a}$$

$$(DH)_\beta = 1.725*10^{-3}*e^{-0.8*t} + 4.15*10^{-4}*e^{-0.0237*t} \tag{2b}$$

$$(DH)_\gamma = 3.8*10^{-4}*e^{-0.69*t} + 2.111*10^{-4}*e^{-0.0225*t} \; . \tag{2c}$$

In the case of G-values that are a function of the total dose, the total dose is defined in terms of only the radiation with a very short attenuation range (alpha, beta), which will be completely absorbed by the waste material. The total heat generation or dose (W-yr) for alpha and beta radiation per curie (Ci(0)) of waste is defined by the following equations:

$$D_\alpha(t) = 5.169*10^{-4}*t + 4.2652*10^{-6}*t^2 - 4.033*10^{-8}*t^3 \tag{3a}$$

$$D_\beta(t) = 1.967*10^{-2} - 2.15*10^{-3}*e^{-0.8*t} - 1.751*10^{-2}*e^{-0.0237*t} \tag{3b}$$

FRACTION OF EMITTED ENERGY ABSORBED IN WASTE MATERIAL, F

The fraction of emitted energy from type i radiation absorbed in waste material, F_i, is a function of the fraction of emitted energy that escapes the radioactive particles and the fraction of energy absorbed by the waste material:

$$F_i = f_i \phi_i \tag{4}$$

where

f_i = fraction of emitted energy from type i radiation that escapes the radioactive particles

ϕ_i = fraction of escaped emitted energy from type i radiation absorbed in the waste material.

Self-shielding prevents a fraction of the alpha radiation from being emitted. It has been estimated that for a given particle size distribution of radioactive particles, 82% of the alpha decay energy from particulate contamination is available to interact with waste materials (3). Thus, in this analysis, f_α is assumed to be 0.82. Beta radiation is not subject to self-shielding. The fraction of

escaped emitted alpha and beta energy absorbed in the waste material is assumed to be unity due to the short attenuation range of these types of radiation. As a result, F_α equals 0.82, and F_β equals unity.

In the case of gamma radiation, no radiation is attenuated by the radioactive particles, and therefore, the fraction of emitted energy that escapes the radioactive particles, f_γ, is unity. Thus, F_γ equals ϕ_γ. A previously developed model calculates the fraction of gamma radiation absorbed in waste material, ϕ_γ, as a function of the gamma radiation energy (E), the characteristic packaging dimension (L), and the waste density (ρ) (6). For this paper, we developed an algebraic expression [Equation (5)] as a convenient means of calculating the fraction of absorbed gamma radiation in terms of these parameters (7).

$$\phi_\gamma = 1 + [A_1 E + A_2 E^2 + A_3 E^3][\frac{A_4}{L} + \frac{A_5}{L^2} + \frac{A_6}{L^3}][\frac{A_7}{\rho} + \frac{A_8}{\rho^2} + \frac{A_9}{\rho^3}] \ . \tag{5}$$

Isotopic distributions obtained from the ORIGEN 2 analysis indicate that at the time of drum loading about 10% of gamma decay heat is from low-energy isotopes (<0.1 MeV) and 90% from higher-energy isotopes (>0.1 MeV). High-energy gamma radiation is primarily emitted from two isotopes (Rh-106 and Ba-137m), each with greater than 0.5 MeV in energy. Equation (5) is used to estimate the fraction of absorbed gamma radiation from the higher-energy isotopes. It was also observed from ORIGEN2 calculations that gamma radiation emitted by low-energy isotopes decreases as a function of time, t, until, after 5 years, most radiation is emitted by higher-energy isotopes. The total fraction of gamma radiation absorbed as a function of time, $\phi_{\gamma,t}$, is calculated by the following equations:

$$\phi_{\gamma,t} = [0.9 + (\frac{t}{5})0.1]\phi_{0.5MeV} + 0.1[1 - (\frac{t}{5})]\phi_{0.1MeV} \ ; \ t < 5 \ \text{yr} \tag{6a}$$

$$\phi_{\gamma,t} = \phi_{0.5MeV} \ ; \ t \geq 5 \ \text{yr} \ . \tag{6b}$$

The fraction of gamma radiation emitted by low-energy radioisotopes absorbed by the waste, $\phi_{0.1MeV}$, is conservatively assumed to equal unity.

HYDROGEN GAS GENERATION POTENTIAL, G

In the case of a drum containing M types of hydrogenous materials, an effective G-value is defined in terms of the waste-specific G-values (3):

$$G_{eff,i} = \frac{M}{\underset{m}{}}(\bar{f}_m G_m)_i \tag{7}$$

where $\bar{f}_m$ is the fraction of energy absorbed by material m, and the subscript i represents radiation type.

Many models simulating hydrogen generation conservatively assume constant G-values. Radiolysis experimental results indicate that the hydrogen-generation potential of many organic materials decreases as a function of total dose due to waste matrix depletion (8). Two different approaches were evaluated to estimate matrix depleted G-values: step function and continuous depletion.

4

Step Function

The G−values of various hydrogenous materials have been tabulated in the 72-B Shipping Package Safety Analysis Report (3). For example, maximum G-values (independent of radiation type) for polyethylene and dry cellulose are 4.1 and 3.2 moles/100 eV, respectively, up to the total dose of 0.012 W-yr. After the total dose 0.012 W-yr, the G-values for polyethylene and dry cellulose are 0.64 and 0.59 moles/100 eV, respectively (8). The highest G-value associated with matrix-depleted organic material is 1.09 moles/100 eV (wet cellulose).

Continuous Depletion

Waste matrix depletion describes the decrease in hydrogen gas generation potential of material as hydrogen in the hydrogenous material is depleted by radiolytic reactions. An expression for G-value (moles/100 eV) in the case of continuous matrix depletion was developed.

$$G = k_1 . e^{-k_2 D(t)} \tag{8}$$

where k_1 (moles/100 eV) and k_2 (W-yr)$^{-1}$ are matrix-dependent constants, and $D(t)$ is total dose in W-yr. For polyethylene, the constants k_1, and k_2 are derived from INEEL matrix depletion study (8) to be 2.34 and 1.45 and for dry cellulose 0.934 and 143.1; respectively.

HYDROGEN GAS GENERATION MODEL

Combining Equations (1) and (7), and simplifying the resulting equation, yields an expression describing the hydrogen generation rate:

$$n_{H_2,gen} = C[f_\alpha G_{eff,\alpha}(DH)_\alpha + G_{eff,\beta}(DH)_\beta + \phi_\gamma G_{eff,\gamma}(DH)_\gamma] \tag{9}$$

It is assumed that all hydrogen gas generation occurs within the innermost layer of confinement. The accumulation rate of hydrogen within the innermost layer of confinement is defined by the hydrogen generation rate and gas leakage rate from that volume:

$$\frac{dn_{H_2,1}}{dt} = n_{H_2,gen} - L_{R,1\rightarrow2}\left[\frac{n_{H_2,1}}{n_{T,1}} - \frac{n_{H_2,2}}{n_{T,2}}\right] \tag{10}$$

where

$n_{H_2,j}$ = moles of hydrogen within the jth void volume

$n_{H_2,gen}$ = hydrogen generation rate, mol s^{-1}

$L_{R,1\rightarrow2}$ = hydrogen leak rate across innermost layer of confinement, mol s^{-1} (mol fraction)$^{-1}$

$n_{T,j}$ = moles of gas in jth void volume

t = time, s.

Hydrogen gas accumulation in the outer layer of confinement can be defined by the generic equation,

$$\frac{dn_{H_2,j}}{dt} = L_{R,j-1}\left[\frac{n_{H_2,j-1}}{n_{T,j-1}} - \frac{n_{H_2,j}}{n_{T,j}}\right] - L_{R,j}\left[\frac{n_{H_2,j}}{n_{T,j}} - \frac{n_{H_2,j+1}}{n_{T,j+1}}\right] \tag{11}$$

where $n_{H_2,j}$ and $n_{T,j}$ are the moles of hydrogen and total gas within the jth layer of confinement, respectively. The moles of hydrogen immediately outside the drum is assumed to be zero at all times. The system of differential Equations (10) and (11) are solved using the Runge-Kutta-Fehlberg numerical method (9).

LEAK RATES

For heat-sealed polymer bags, the hydrogen leak rate; $L_{R,P}$ (mole/sec-mole fraction), is a function of hydrogen gas permeability across the polymer, bag thickness, bag surface area, gas concentration, and pressure:

$$L_{R,P} = \frac{\wp A_p P c}{x_p} \tag{12}$$

where

$\wp$	=	hydrogen gas permeability, cm³(STP) cm⁻¹ s⁻¹ (cm Hg)⁻¹
A_p	=	bag permeable area, cm²
P	=	gas pressure, cm Hg
c	=	gas concentration, mol/cm⁻³
x_p	=	bag thickness, cm.

$\wp$ = hydrogen gas permeability, $cm^3(STP)\ cm^{-1}\ s^{-1}\ (cm\ Hg)^{-1}$

A_p = bag permeable area, cm^2

P = gas pressure, cm Hg

c = gas concentration, mol/cm^{-3}

x_p = bag thickness, cm.

HYDROGEN GAS GENERATION IN DRUMS 728–737

The model developed above was used to calculate hydrogen generation in unvented DOT 17-H 30-gallon waste drums, as shown in Figure 1. For the purpose of this exercise, the radionuclide inventory contained in drums 728 –737 was used. The TRU-contaminated items were loaded into ten RH-TRU waste drums (00728 through 00737) on November 1, 1991. Total radioinuclide inventory at the time of drum loading is estimated to be 37.5 Ci in the ten drums, or, on average, 3.75 Ci per drum. The Radioactive Waste Management Information System (RWMIS) data spreadsheets indicate that most batches reported an average of about 2.5 Ci per drum, and a maximum total of about 5 Ci in a few drums at the time of drum loading.

Three scenarios are analyzed: (1) best-estimate, in which 3.75 Ci in the waste drum was used as an initial loading and best-estimate values for other parameters, (2) conservative, in which 3.75 Ci were used in the waste drum as an initial loading and conservative values for other parameters, and (3) bounding analysis, in which the average inventory, 3.75 Ci, was increased by 50% to 5.6 Ci as an initial loading, and conservative values were used for other parameters.

A number of assumptions were made in estimating absorbed gamma radiation. Since the characteristic packaging dimension (drum radius) of a 30-gallon drum is less than the values used to define the coefficients in Equation (5), the lower limit (L = 0.95 ft, the same as a 55-gallon drum) is used. Liekhus (10) estimated the waste densities between 0.26 to 0.73 g cm⁻³ using the weight range for 30-gallon drums and drum liner volume containing waste described by INEEL content code IDC 104. The maximum waste density of 0.73 g cm⁻³ is used in all calculations.

Total volume of innermost layer is estimated to be 98,420 cm³ (26 gallons) (11). The innermost volume contains two 7.5-gallon pails of waste material. Total volume occupied by the waste and drum liner is assigned to be 28,390 cm³ (7.5 gallons). Therefore, the void volume in the innermost layer would be 70,030 cm³. The remaining two layers (middle and outer layer) are assigned void

volume of 7571 cm^3 (2 gallons) each. The middle layer is the void volume between the two polymer bags. The outer layer is the void volume between the drum and the polymer bag.

The quantity of waste composition is often unknown; therefore, it is assumed that all organic waste consists of polyethylene. Polyethylene has the highest hydrogen generating potential of any material expected in the waste drum. The G-value is a function of the total dose and radiation type. For the best-estimate case, hydrogen generation rates for alpha and beta radiation is estimated using time-dependent (dose-dependent) Equation (8).

In the conservative case, for alpha and beta radiations the maximum nonmatrix-depleted G-value is assumed to be 4.1 moles/100 eV up to the total dose of 0.012 w-yr (12). After the matrix depletion, the effective G-value of 1.09 moles/100 eV, constant for alpha and beta radiations, is used. This value is the highest G-value associated with matrix-depleted organic material (i.e. wet cellulose). The gamma radiation G-value is assumed to remain constant and to be independent of the total radiation dose. This assumption is based on the large mass of polymeric packaging material, as well as the significant decrease in the fraction of absorbed gamma radiation with increasing gamma radiation energy. For gamma radiation, a G-value of 4.1 moles/100 eV, constant, is assigned. Note that all the hydrogen generated from gamma-radiation is assumed to be in the innermost volume.

The hydrogen leak rate across the heat-sealed polymer bags, $L_{R,P}$, is a function of hydrogen gas permeability across the polymer, bag thickness, bag surface area, and gas concentration, as shown in Equation (12). Hydrogen gas permeability across polyethylene is on the order of 10^{-9}; polymer bag thickness is 5×10^{-2} cm (20 mil) (11). The permeable surface area of a polymer bag is estimated, based on the cardboard liner diameter of 69 cm and height of 69 cm, to be 1.5×10^4 cm^2. Assuming standard conditions (STP = standard temperature and pressure), pressure and gas concentration are 76 cm Hg, and 4.1×10^{-5} moles/cm^3, respectively. Note that it is assumed there is no significant pressure buildup inside the bags; consequently, the gas concentration per volume unit remains unchanged. As a result, the hydrogen leak rate from a heat-sealed polymer bag in a 30-gallon waste drum using Equation (12) is estimated to be about 1×10^{-6} mol/s/mol fraction. In the case of bags taped closed, hydrogen leak rates are on the order of 10^{-6} to 10^{-7} mol/s/mol fraction (12). The leak rate value 1×10^{-6} mol/s/mol fraction was used in the best-estimate scenario. In the case of conservative and bounding scenarios, the leak rate across polymer bags of 1×10^{-7} mol/s/mol fraction was used (13).

Measured hydrogen diffusion characteristics across an unvented 55-gallon drum lid gaskets were on the order of 10^{-6} to 10^{-7} mol/s/mol fraction (14). It is assumed that the leak rate across the lid gasket is proportional to the circumference of the gasket; thus, the hydrogen diffusion characteristic across the gasket of a 30-gallon drum is on the order of 10^{-7} mol/s/mol fraction. This leak rate was used for all three scenarios.

LIMITED COMPARISON TO RADCALC 3.0 PREDICTIONS

A limited comparison was made to the Radcalc computer code. The Radcalc computer code was developed by Westinghouse Hanford for the Department of Energy (DOE) (1). Duratek Federal Services (DFSNW) currently maintains it for the DOE's National Transportation Program. Radcalc is a software program that has been used in the transportation and packaging of high- and low-level radioactive waste programs. Radcalc has the capability to calculate the production of hydrogen gas by radiolysis process in the waste matrix of radioactive waste.

The radioactive isotope inventories used to calculate the heat generation rates as a function of time in the INEEL code were used in the Radcalc code. The average inventory case was simulated. Since, the Radcalc does not model hydrogen concentrations in multiple layers (volumes), hydrogen gas generation in a single volume, innermost volume (i.e., 70030 cm^3), was simulated. Also, the Radcalc code does not simulate the leakage or diffusion out of the volume; therefore, in order to compare the two codes, a zero leakage rate was modeled in the INEEL code. The 100% gamma absorption option was used in both cases. An effective G-value was modeled of 4.1 mole/100ev constant for alpha, beta, and gamma radiations. The Radcalc code was executed with decay time equal to zero and with increment of 0.1 to 1.5 year (repeated 15 times). There is an excellent agreement between the INEEL-code-predicted data and the Radcalc predicted values, as shown in Figure 2.
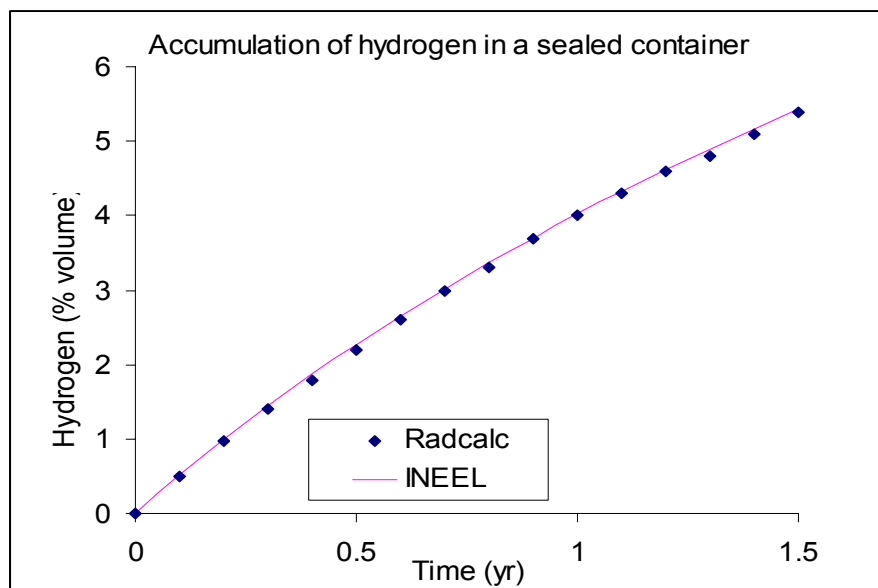

Figure 2. Hydrogen concentration predicted by the INEEL and Radcalc codes.

RESULTS AND CONCLUSIONS

In the best-estimate case, the maximum hydrogen concentration (0.95%) in the innermost volume of a waste drum would occur within the first year after packaging. By comparison, maximum hydrogen concentration (4.22%) in the conservative case would occur in 1.85 years. Most ANL-E drums are more than 15 years old. The hydrogen concentration in 15-year-old drums in the conservative scenario case is expected to be less than 1.51% in the innermost volume and less than 0.2% in the best-estimate case (see Figures 3 and 4).

When the bounding analysis was performed using the conservative leak rates and hydrogen generation rates, under no circumstances was a drum containing the maximum initial loading of 5.6 Ci predicted to exceed 5% by volume within the innermost heat-sealed bag. The hydrogen leak rates from the bags and drum would be sufficient to minimize the accumulation of hydrogen in a drum. The maximum hydrogen concentration of 4.8% was achieved after 1.1 years, and after 15 years the concentration was predicted to be less than 2.28% (see Figure 3). Table 1 below summarizes the results.

Table 1. Predicted hydrogen concentration (vol%)
inside the innermost layer of the 30-gallon waste drums.

H_2 Concentration	Inventory[a]	Drum Age after Closure (yr)													
CASE	Ci(0)	1	2	3	4	5	6	7	8	9	10	15	20	25	30
Best Estimate %	3.75	0.95	0.85	0.63	0.46	0.35	0.28	0.23	0.21	0.19	0.18	0.15	0.13	0.12	0.11
Conservative %	3.75	3.11	4.14	3.64	3.19	2.8	2.48	2.24	2.05	1.9	1.79	1.51	1.4	1.33	1.27
Bounding %	5.6	4.6	4.65	4.32	3.95	3.61	3.31	3.07	2.88	2.73	2.61	2.28	2.13	2.02	1.93

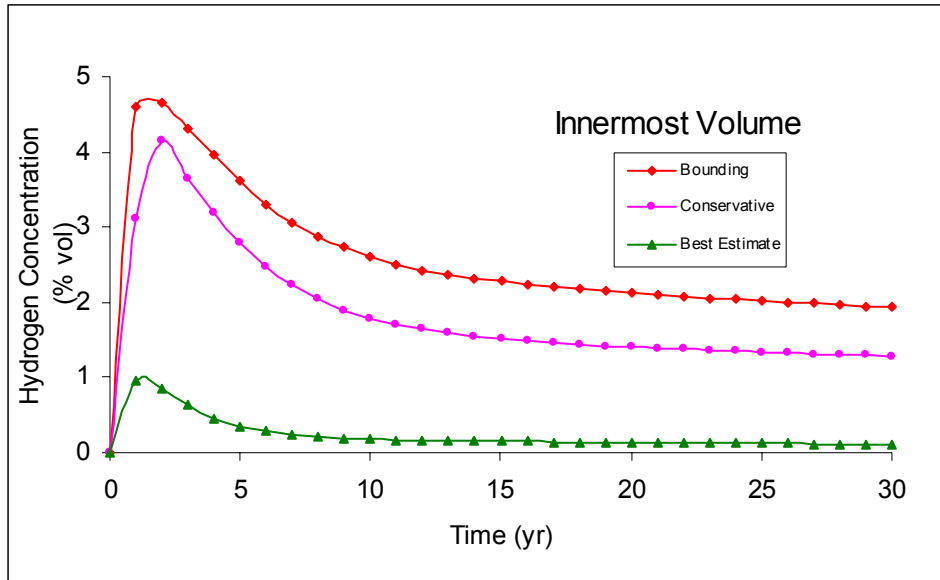a. All inventories are at the time of drum loading.

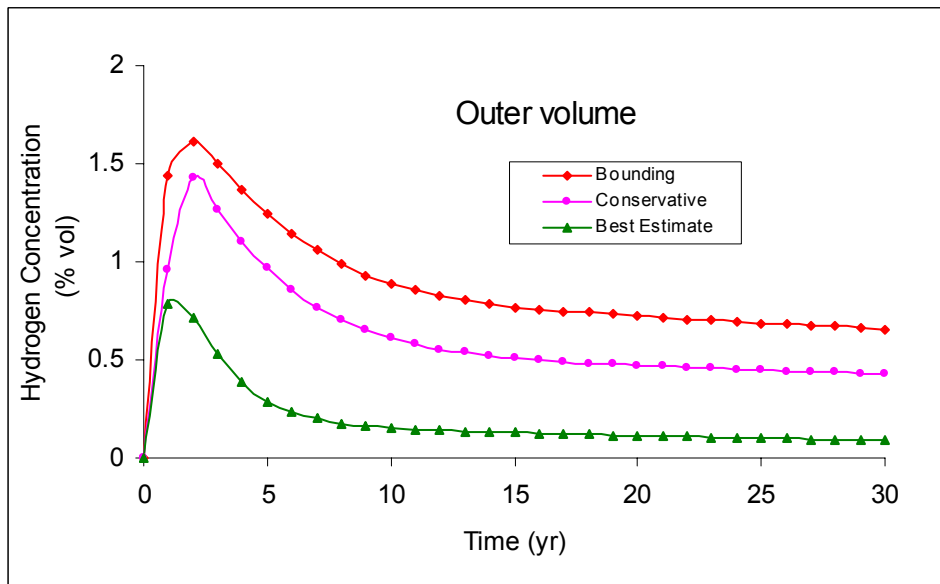Figure 3. Hydrogen concentration in the innermost volume of a drum (vol%).

Figure 4. Hydrogen concentration in the outermost volume of a drum (vol%).

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