

WSRC-TR-2007-00200

Keywords: Radiolysis,  
Hydrogen

# Evaluation of Radiolysis-Induced Hydrogen Generation in DOT 6M Drums from INTEC

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Materials Science and Technology

Publication Date: June 2007

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**Aiken, SC 29808**



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This document was prepared in connection with work done under Contract No. DE-AC09-96SR18500 with the U. S. Department of Energy

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**DOCUMENT:**      **WSRC-TR-2007-00200**

**TITLE:**            **Evaluation of Radiolysis-Induced Hydrogen Generation in DOT 6M Drums  
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## 1.0 EXECUTIVE SUMMARY

Three DOT 6M 30-gallon drums are scheduled to be shipped from the Idaho Nuclear Technology Engineering Center (INTEC) at the Idaho National Laboratory (INL) to L-Area at the Savannah River Site (SRS). These three drums contain radioactive materials that resulted from the material recovery effort following a small explosion that had occurred in the Idaho Chemical Processing Plant (ICPP) hot chemistry laboratory (HCL). In support of the shipment and subsequent storage of the three DOT 6M drums, an evaluation of the potential for molecular hydrogen production in the drums has been completed and documented herein. The potential sources of hydrogen evaluated in the current report include radiolytic decomposition of polymeric materials in the DOT 6M drums #3031 and #3598 and the radiolytic decomposition of water in drum #20102. No other potential sources have been identified based upon reported drum contents and packaging configuration.<sup>1-2</sup>

A parametric approach was used to evaluate the maximum quantity of molecular hydrogen that can be expected to evolve in two DOT 6M 30-gallon drums in support of receipt and subsequent interim storage prior to canyon processing. These drums are two of three drums scheduled for shipment from INTEC to SRS as part of the decommissioning effort of the INTEC facility. The three DOT 6M drums will be received at L-Area in SRS and stored for up to 13-years prior to final disposition at HB-Line in 2020. Results of the current analysis do not include parametric analysis of drum #20102 containing 114/133 SAL (salvage) which contains UO<sub>3</sub> powder. This drum has not been identified as containing polymeric materials and a conservative calculation indicates that the maximum gross molecular hydrogen production due to the radiolysis of adsorbed moisture would yield a production rate of 5.1-cm<sup>3</sup>/yr, driven primarily by the large surface area to volume ratio of the oxide powder. The remaining two drums, #3031 and #3598 contain polymer bags and/or bottles that will be subject to radiolytically induced hydrogen gas generation due to decomposition of the polymers. Conservative values for hydrogen gas generation rates and rates of pressure increase within the drums have been determined based upon a number of inputs and assumptions. The results are that hydrogen will be produced at a rate of 1.93-cm<sup>3</sup>/yr and 1.50-cm<sup>3</sup>/yr, respectively for drums #3031 and #3598. Projected molecular hydrogen concentrations at 2020 have been calculated to remain below the lower flammability limit of 4% molecular hydrogen by volume in air.

## 2.0 INTRODUCTION

Three DOT 6M 30-gallon drums are planned to be shipped from the Idaho Nuclear Technology Engineering Center (INTEC) at the Idaho National Laboratory (INL) to L-Area at the Savannah River Site (SRS). These three drums contain radioactive materials that resulted from the material recovery effort following a small explosion that had occurred in the Idaho Chemical Processing Plant (ICPP) hot chemistry laboratory (HCL). The three DOT 6M drums would be received at L-Area in SRS and stored for up to 13-years prior to final disposition at HB-Line by 2020. In support of the shipment and subsequent storage of the three DOT 6M drums, an evaluation of the potential for molecular hydrogen production in the drums due to the radiolytic decomposition of the polymer bottles in drums #3031 and #3598 and due to radiolytic decomposition of water reported in drum #202102 has been completed and documented herein.

## 3.0 MATERIALS DESCRIPTION

Three DOT 6M 30-gallon drums are planned to be shipped from the Idaho Nuclear Technology Engineering Center to the Savannah River Site. The debris in each drum has been carefully characterized by personnel at the ICPP HCL.<sup>3</sup> Two of the three DOT 6M 30-gallon drums from INTEC, #3031 and #3598, contain a variety of materials that are characterized as quarter circles, pins, pellets, foils, wires, chips, turnings, and small filings/specks. The two largest pieces that are roughly shaped as quarter circles with a 5-cm radius and that are two centimeters thick are contained in drum #3598. They are

characterized as 4.5 wt % uranium fissium alloy which are contaminated with  $^{239}\text{Pu}$  (200-400 ppm).<sup>2</sup> These two pieces are located in areas 7 and 14 of Figure 1. Figure 2 provides a graphical depiction of loading of the two 30 gallon DOT 6M drums. In drum #3598, the two quarter circles are each within a low-density polyethylene bag and the two bags within a 500-ml high-density polyethylene (HDPE) bottle. The HDPE bottle is closed with a screw cap and placed within a crimp-sealed tin can, HCL-2, that is placed inside the sealed 2R container within the DOT 6M 30-gallon drum. The can contains 1,190 grams of total uranium of which 617 grams are  $^{235}\text{U}$ .

Drum #3031 contains the remaining metallic pieces shown in the photograph of Figure 1. These pieces are counted as 45 pins, 19 pellets, 7 foils, 4 wires, 3 chips, 131 turnings, and 1083 small filings/specks. These pieces are contained within a 500-ml HDPE bottle that is contained in a crimp-sealed tin can, HCL-1 that is sealed in the 2R container within the DOT 6M 30-gallon drum (see Figure 2). All of the pellets and chips and a majority of pins, foils, and small filing/specks are described as uranium fissium alloy low in zirconium. The uranium fissium alloy is 3.5 or 4.5 wt % fissium. The remaining material in this can consists of a small number of pins, foils, and small filing/specks that consist of a uranium alloy high in zirconium. Also stored in the can as foils, wires, turning, wire, and small filings/specks is a small quantity (gram amounts) of metal: Inconel, stainless steel (304), thorium, and nickel alloy. The material is contaminated with  $^{239}\text{Pu}$  (200-400 ppm). The can, HCL-1, contains 290 grams of total uranium of which 255 grams are  $^{235}\text{U}$ .

The third DOT 6M 30-gallon drum is labeled #20102 and contains material described as soil samples. The soil sample material is characterized as  $\text{UO}_3$  granular powder, a high-grade uranium oxide product obtained from reprocessing fuel and scrap material at INTEC.<sup>1</sup> The material is comprised of 29 grams of total uranium, of which 26 grams are  $^{235}\text{U}$ . In addition, this drum is reported to contain 0.45-g of water due to the hygroscopic nature of the uranium-oxide. This material is contained within the crimp sealed tin can 114/133 SAL shown in Figure 4 that is within the 2R inside the DOT 6M 30-gallon drum as depicted in Figure 3.



**Figure 1** Photograph of the recovered metallic pieces recovered from ICPP HCL.

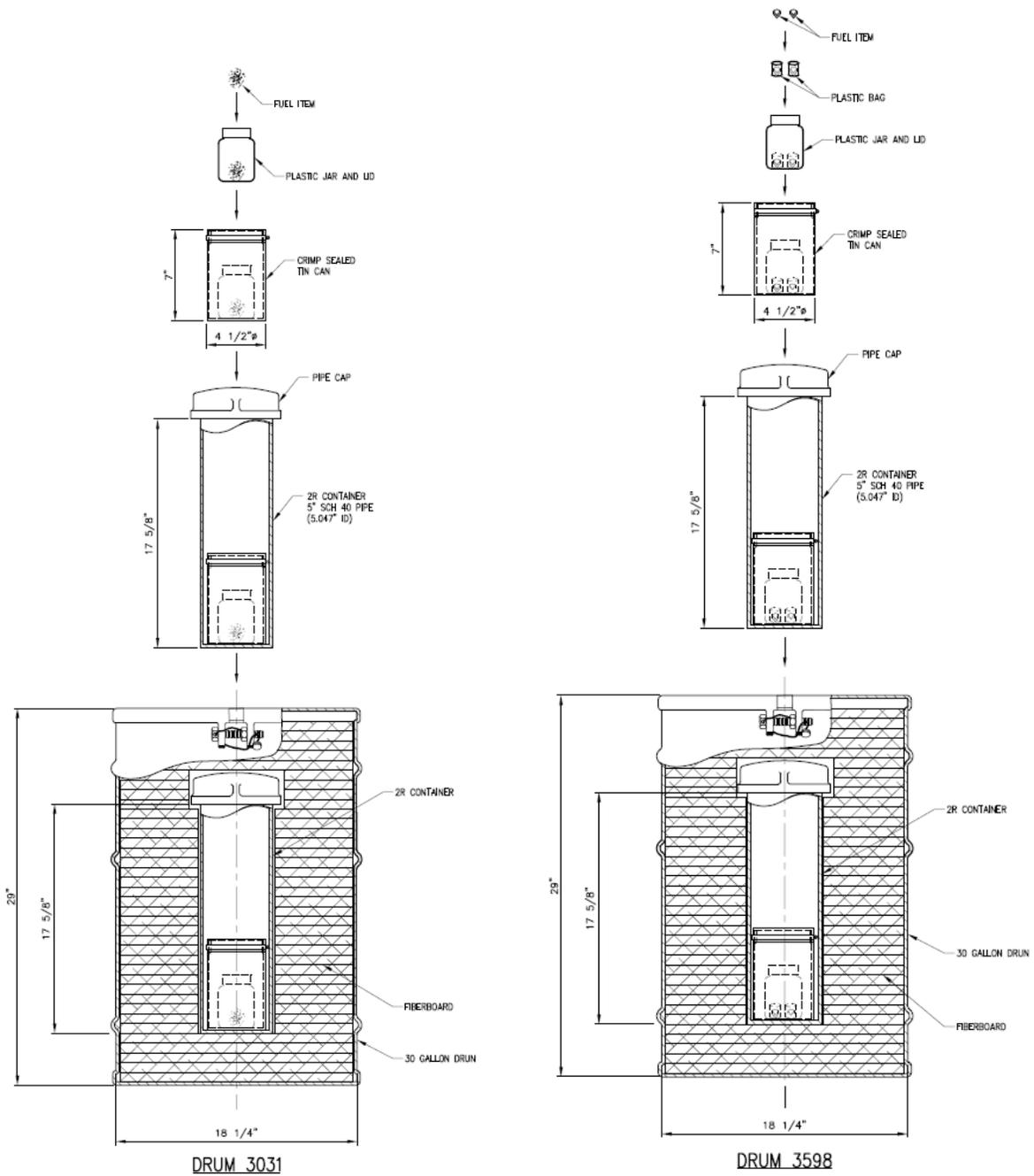


Figure 2 Schematic representation of the packaging configuration for DOT 6M 30-gallon drums #3031 (Left) and #3598 (Right).

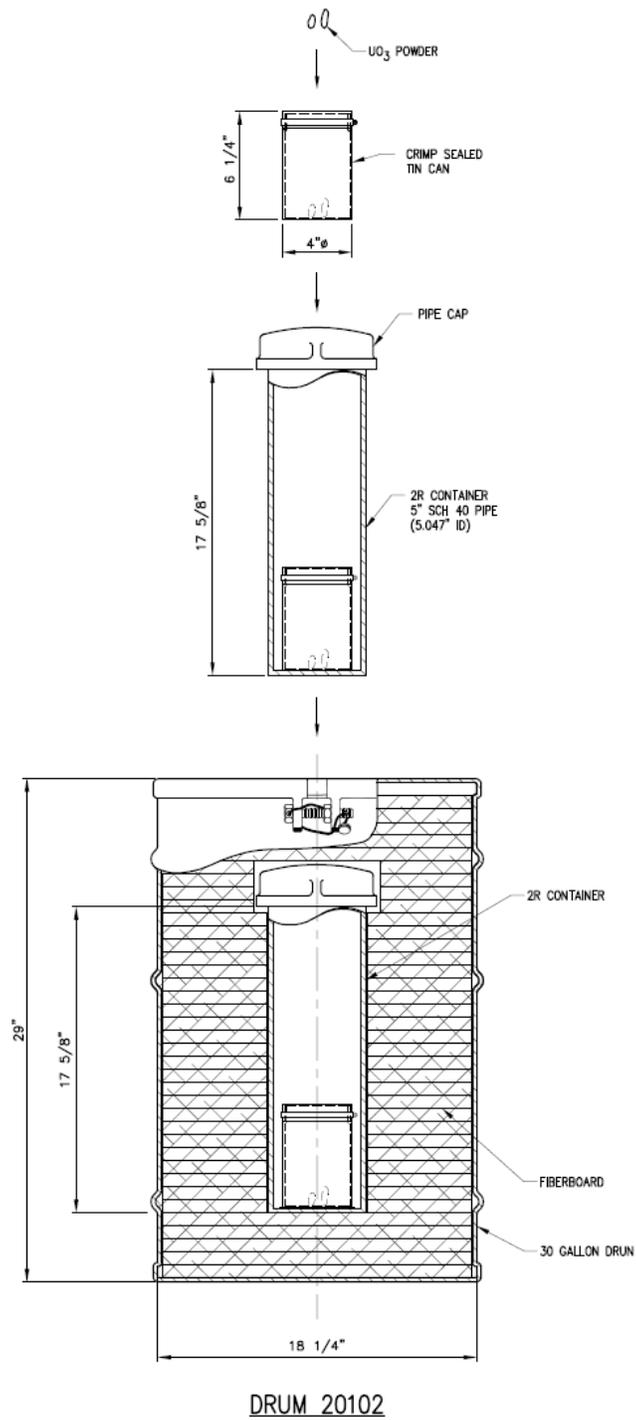


Figure 3 Schematic representation of the packaging configuration for DOT 6M 30-gallon drum #20102.



**Figure 4** Photograph of the crimp-sealed tin can (114/133 SAL) that is stored in a 2R container within the DOT 6M 30-gallon drum #20102.

**Table 1** Summary of the Uranium-Fissium Debris Contained in Three DOT 6M 30-gallon Drums

Can/Drum	Description	Form	Total U (g)	Total Pu (g)
HCL-1 #3031	Uranium-Fissium (3.5-4.5%)	45 pins, 19 pellets, 7 foils, 4 wires, 3 chips, 131 turnings, 1083 filings/specks	290	0.12
HCL-2 #3598	Uranium-Fissium (3.5-4.5%)	2 large quarter round chunks	1,190	0.48
114/133 SAL #20102	High-Grade UO <sub>3</sub> Powder	UO <sub>3</sub> Granular Powder	29	0

#### 4.0 CALCULATIONAL APPROACH AND METHODOLOGY

The potential sources of hydrogen evaluated in the current report include radiolytic decomposition of polymeric materials in the DOT 6M drums #3031 and #3598 and the radiolytic decomposition of water in drum #20102. No other potential sources have been identified based upon reported drum contents and packaging configuration.<sup>1-2</sup>

Ionizing radiation causes radiolytic decomposition of polymers. The decomposition of polymers lead to the production of molecular hydrogen. The general approach used to develop an estimate of the potential for molecular hydrogen generation due to alpha radiolysis of polymers follows the approach used by Croft Associates for the SAFKEG package<sup>4</sup>. The rate of production of H<sub>2</sub> from decomposition of polymeric material in units of cm<sup>3</sup>/sec (V<sub>g</sub>) can be generally expressed as:

$$V_g = \left( D \times G \times k \times v / A_n \right) \quad (\text{Eq. 1})$$

where: D = alpha energy absorbed in the polymer (Watts),  
G = hydrogen gas generation constant (Molecules/MeV)  
k = conversion factor (6.24e12 Mev/J),  
v = volume of 1 mole of gas at STP (2.24e4 cm<sup>3</sup>), and  
A<sub>n</sub> = Avogadro's number (6.022e23 molecules).

For the current work, the alpha energy absorbed in the polymer, D, and the value of the hydrogen gas generation constant, G, are the two unknowns in Eq. 1. For the determination of D, it is necessary to determine the range of alpha particles in the various materials. The energy and material dependant range of an alpha particle in cm (R) is determined by the equation:<sup>5-6</sup>

$$R = 2.3 \times 10^{-4} \left( \frac{\sqrt{M}}{\rho} \right) (1.24 E_\alpha - 2.62) \quad (\text{Eq. 2})$$

where: M = molecular weight of the medium (AMU),  
ρ = density of the medium (g/cm<sup>3</sup>), and  
E<sub>α</sub> = average alpha energy (MeV).

For the current work, the molecular weight and density of the medium and the average alpha energy are the three unknowns in Eq. 2. Having determined the volume of molecular hydrogen produced using Eq. 1, it is then possible to determine the pressure rise within the package as a function of time (P<sub>r</sub>(t)) by the equation:<sup>4</sup>

$$P_r(t) = \left( P_a \times V_g \times t / V_c \right) \quad (\text{Eq. 3})$$

where: P<sub>a</sub> = ambient pressure (1.0 bar abs),  
t = time (secs), and  
V<sub>c</sub> = free volume of container (cm<sup>3</sup>).

For the current work, the time, t, and free volume of the container, V<sub>c</sub> are the two unknowns in Eq. 3. The next section of the report addresses the assumptions, methodology, and calculations used to develop a conservative estimate of the expected molecular hydrogen production due to the contents of the drums as described in the previous section.

#### 4.1 List of Assumptions

1. The value of the hydrogen gas generation constant (G-value) is assumed equal 3.5 molecules/100 eV or 3.5×10<sup>4</sup> molecules/MeV. This value of G is taken from Reference 7 as the highest reported production rate for all polymers due to radiolytic decomposition by

- incident alpha radiation. This assumption is conservative because no credit is taken for the dose dependent nature of radiation damage in the polymer. Increasing the dose absorbed by a polymer will decrease the effective G-value for hydrogen production due to the depletion of the matrix in the vicinity of the alpha-emitting radioactive source particle. Dose dependent G values are reported as less than 1/5th the value assumed in the current analysis.<sup>8</sup>
2. The value used for the molecular weight of the material in the HDPE bottles is 242-g/mole. This number is consistent with that of plutonium. This number bounds the actual effective molecular weight of the contents of the drums, since the contents include fission elements whose weights are generally less than 100. This assumption is conservative as alpha range increases as the square-root of the molecular weight (see Eq. 2).
  3. The bottle is modeled as a right-circular cylinder with a 4-cm radius and a height of 14.2-cm. This assumption is conservative as the surface area of the modeled ~700-ml bottle is greater than that of the expected 500-ml bottle actually used.
  4. It is assumed that the materials in drums #3031 and #3598 are essentially dry. Unlike the materials in drum #20102, this material is not hygroscopic in nature and water content in these drums would be mostly limited to humidity in the facility. Radiolysis of bulk water in these two drums will not provide significant molecular hydrogen production relative to the production estimated for the production due to polymeric radiolytic decomposition. The short range of the alpha particles, the conservatism introduced by Assumption 1, and the G-value of  $1.7 \times 10^4$  molecules/MeV<sup>9</sup> for H<sub>2</sub> generation from alpha radiolysis of water support this assumption.
  5. The average alpha energy used in Eq. 2 is assumed equal to 5.25-MeV. This value bounds the alpha particles from all isotopes identified as being in the drums per the Appendix A and supporting documentation.
  6. It is assumed that the free volume of the 2R containers and the crimp-sealed tin cans in the DOT 6M 30-gallon drums is equal to 90% of their physical volume. This assumption is conservative as the contents of the tin cans are expected to occupy less than 7% of the tin can and less than 3% of the 2R containers.<sup>3</sup> Increasing the free volume of the containers will further decrease the estimated pressure increase with time by Eq. 3.
  7. It is assumed that only 50% of the alpha particles that are produced within range of the polymer surface contribute to the energy absorbed by the polymer. This assumption accounts for the fact that an isotropic source would generate 50% of its particles in a direction that takes the particle away from the polymer surface.
  8. It is assumed that the methodology employed in the current parametric evaluation of the HDPE bottles whereby the source material is assumed to completely cover the inner surface of bottle with a homogeneous layer of variable density and related alpha particle range dictated by the layer thickness and the total source material available is sound and bounding.
  9. It is assumed that the bags are not present in HCL-2. This is conservative because the surface area of source material in contact with the polymer bottle is significantly increased by ignoring the plastic bags based upon the current model.
  10. It is assumed that there is no polymeric material in DOT 6M 30-gallon drum #20102. There is no indication of the presence of any polymeric bags or bottles as evidenced by the loading schematic shown in Figure 3.
  11. It is assumed that the bottle is not an effective barrier to hydrogen migration for the pressure rise calculations. This is reasonable due to the high permeability of molecular hydrogen through polyethylene.<sup>10</sup> The crimp-sealed tin can is also ignored for pressure rise calculations for the 2R.
  12. The start time for hydrogen accumulation calculations for drums #3031 and 3598 is assumed equal to 1992. This is a conservative assumption based upon the date at which the HCL crimp-sealed tin cans were loaded.<sup>3</sup>

13. The start time for the hydrogen accumulation calculations for drum #20102 is assumed equal to 2006.<sup>11</sup>
14. The 0.45-g of water reported for drum #20102<sup>1</sup> is assumed homogeneously mixed with alpha-producing uranium within the drum. That is to say that no credit is taken for the shielding effects of the UO<sub>3</sub> particles themselves in the current calculation. In addition, no credit is taken for recombination or reverse reactions by which molecular hydrogen is consumed to regenerate water molecules. This is a conservative assumption.

## 4.2 Determination of the Absorbed Alpha Energy

### 4.2.1 Total Alpha Energy Production

The total production of alpha particles generated in each of the crimp-sealed tin containers is completely dependent upon contents of the tin can. References 3-1 provide a list of the radioactive isotopes and quantities contained in each of the three DOT 6M 30-gallon drums. Tables 2 & 3 provide the alpha decay energy and isotopic content by weight for the three drums. Table 2 shows the calculation to determine the power density for each of the isotopes included in the 6M's. The power density is calculated as the product of the average alpha energy per alpha particle as obtained from the ENDF-VI decay library and the activity density of isotope as obtained from 10CFR71.<sup>12</sup> The total alpha energy generation due to radioactive decay can then be calculated and results are shown in Table 3, where the energy generation by isotope is the product of the power density and the total grams as obtained from the Appendix A documents.<sup>1-2</sup>

**Table 2 Summary Table of Calculations for Power Density Determination<sup>12-13</sup>**

Isotope	E <sub>α</sub> (MeV)	E <sub>α</sub> J/α	Activity Density (Ci/g)	Activity Density (Bq/g)	Power Density (Watts/g)
<sup>234</sup> U	4.8419	7.76×10 <sup>-13</sup>	6.20×10 <sup>-3</sup>	2.29×10 <sup>8</sup>	1.78×10 <sup>-4</sup>
<sup>235</sup> U	4.4709	7.16×10 <sup>-13</sup>	2.20×10 <sup>-6</sup>	8.14×10 <sup>4</sup>	5.83×10 <sup>-8</sup>
<sup>236</sup> U	4.5563	7.30×10 <sup>-13</sup>	6.50×10 <sup>-5</sup>	2.41×10 <sup>6</sup>	1.76×10 <sup>-6</sup>
<sup>238</sup> U	4.2610	6.83×10 <sup>-13</sup>	3.40×10 <sup>-7</sup>	1.26×10 <sup>4</sup>	8.59×10 <sup>-9</sup>
<sup>239</sup> Pu	5.2375	8.39×10 <sup>-13</sup>	6.20×10 <sup>-2</sup>	2.29×10 <sup>9</sup>	1.92×10 <sup>-3</sup>
<sup>240</sup> Pu	5.2429	8.40×10 <sup>-13</sup>	2.30×10 <sup>-1</sup>	8.51×10 <sup>9</sup>	7.15×10 <sup>-3</sup>

**Table 3 Summary Table of Calculations for Maximum Total Energy Generation due to Alpha Decay of Isotopes in HCL-1 and HCL-2 and 114/133 SAL**

Isotope	Composition by Can/Drum <sup>†</sup>			Power Density (Watts/g)	Energy Generation by Can/Drum		
	HCL-1 #3031 (grams)	HCL-2 #3598 (grams)	114/133 SAL #20102 (grams)		HCL-1 #3031 (Watts)	HCL-2 #3598 (Watts)	114/133 SAL #20102 (Watts)
<sup>234</sup> U	2.17	6.25	0.22	1.78×10 <sup>-4</sup>	3.86E-04	1.11E-03	3.91E-05
<sup>235</sup> U	255.00	617.00	26.00	5.83×10 <sup>-8</sup>	1.49E-05	3.60E-05	1.52E-06
<sup>236</sup> U	0.93	1.09	0.1	1.76×10 <sup>-6</sup>	1.63E-06	1.91E-06	1.76E-07
<sup>238</sup> U	31.90	565.66	2.68	8.59×10 <sup>-9</sup>	2.74E-07	4.86E-06	2.30E-08
<b>Total U</b>	290.00	1190.00	29.00		4.03E-04	1.15E-03	4.09E-05
<sup>239</sup> Pu	0.11	0.46	—	1.92×10 <sup>-3</sup>	2.12E-04	8.85E-04	—
<sup>240</sup> Pu	0.01	0.02	—	7.15×10 <sup>-3</sup>	7.15E-05	1.43E-04	—
<b>Total Pu</b>	0.12	0.48	—		2.83E-04	1.03E-03	—
<b>Aggregate Total Energy Generation</b>					<b>6.86E-04</b>	<b>2.18E-03</b>	<b>4.09E-05</b>

<sup>†</sup> Taken from Reference 3.

From Tables 2 & 3, it is determined that the total alpha energy production in HCL-1 and HCL-2 is 6.86×10<sup>-4</sup>-Watts and 2.18×10<sup>-3</sup>-Watts, respectively. The alpha energy generation rate for drum #20102 is used to determine the maximum molecular hydrogen generation rates in this drum due to the radiolytic decomposition of the water contents of this drum as prescribed in Section 4.2.4. This drum is not considered in the parametric calculations used to determine the maximum molecular hydrogen generation rate due to the radiolytic decomposition of the polymers in the two remaining drums. The approach to the determination of the fraction of alpha energy that contributes to the radiolytic decomposition of polymeric material in the two remaining drums, #3031 and #3598 is described in Section 4.2.2.

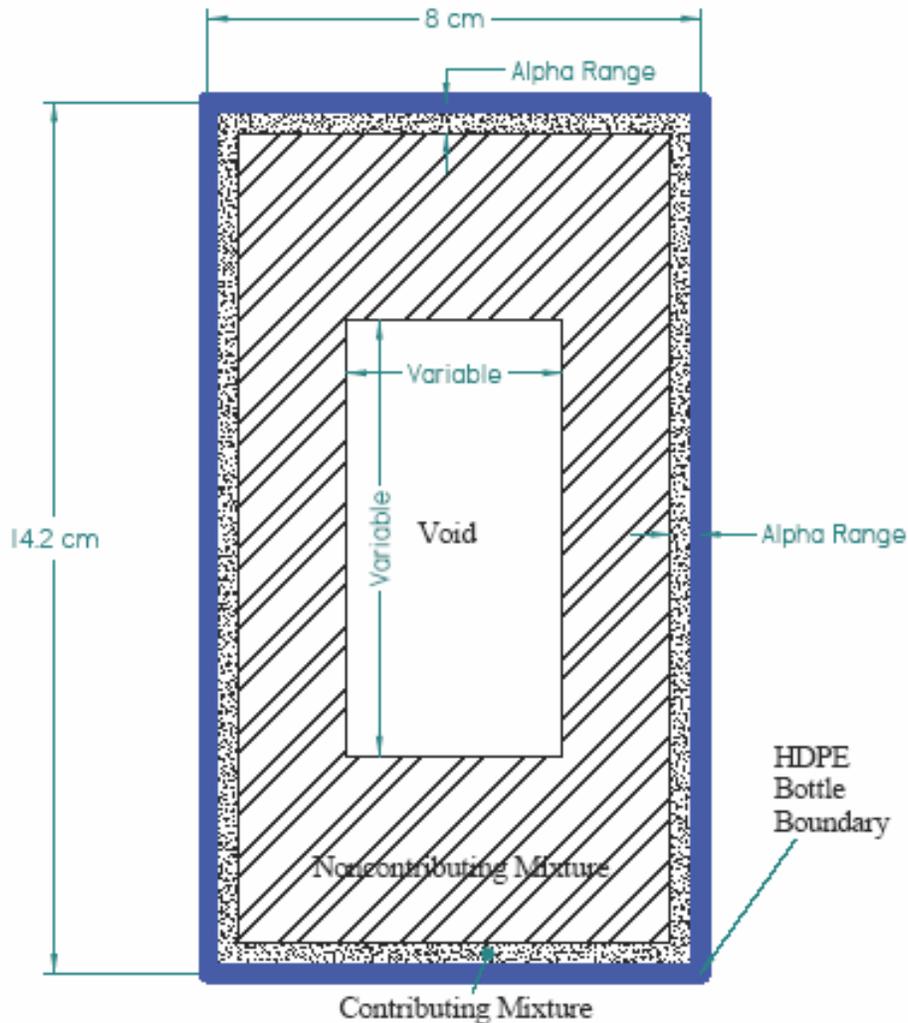
#### 4.2.2 Parametric Evaluation of Fraction of Total Alpha Energy Absorbed by Bottle in Drums #3031 & #3598

A parametric approach was used in the evaluation to determine the maximum fraction of the total alpha particle energy that may be absorbed by the HDPE bottle. Figure 5 provides a schematic of the approach whereby the source material is assumed to occupy a homogeneous volume that is in contact with the inner surface of the HDPE bottle. The variables are the radius and height of the void region in the model (see Figure 5). As the radius and height vary as an increasing fraction of the bottle dimensions, the density of the source region is calculated as:

$$\rho_{source} = \frac{290.12 \text{ or } 1190.48}{V_{bottle} - V_{void}} \quad (\text{Eq. 4})$$

where 290.12 or 1190.48 are used to represent the total source mass in grams in for drum #3031, can HCL-1 and drum #3598, can HCL-2, respectively, V<sub>bottle</sub> is the total bottle volume of 714-cm<sup>3</sup>. For the purpose of this calculation the value of V<sub>void</sub> can vary from ~0-cm<sup>3</sup> to ~714-cm<sup>3</sup>. As the void region increases in volume, the source region volume decreases and the density of the layer on the bottle increases as described in Eq. 4. The values of source region density calculated from Eq. 4 are used in Eq. 2 to determine the alpha range in the homogeneous source material region. The width of the contributing source region is equal to the alpha particle range. The calculated alpha particle range is then used to describe the contributing and noncontributing components of the source regions. The contributing mixture in Figure 5 is the homogeneous source region volume that is within the alpha range of the inner surface of the modeled cylindrical bottle. The total source region volume is the difference between the

bottle volume and the void region volume. The ratio of the contributing mixture volume divided by the total source region volume provides a conservative estimate of the fraction of alpha particle energy that is created within the range of an alpha particle of the surface of the HDPE bottle. This fraction is then applied to the total alpha particle energy in the last row of Table 3 to determine the energy generated by alpha decay in the contributing mixture volume. This total energy is then divided by a factor of 2 to account for the isotropic nature of the source material to result in the total energy that is absorbed in the polymer to generate molecular hydrogen (that is D in Eq. 1).



**Figure 5** Schematic of the model used to determine fraction of alpha energy deposited in HDPE bottles in drum #3031, can HCL-1 and #3598, can HCL-2.

### 4.2.3 Final Estimation of Hydrogen Generation in Drums #3031 & #3598

The parametric approach as described in the previous section results in the consideration of hypothetical scenarios by which the modeled densities range from 0.4-g/cm<sup>3</sup> to more than 5000-g/cm<sup>3</sup>. While these values are unrealistic, irregular material sizes and shapes and uncertainty in material composition and effective packing efficiency have instigated the current parametric methodology that evaluates all theoretically possible values of material and effective packing density. Although such densities are considered in order to bound analytically all possible material densities, it is expected that the actual material density would not achieve a density larger than that of uranium metal or about 18.9-g/cm<sup>3</sup>. The plots of Figures 6 & 7 show that the hydrogen production rates for drum #3031, can HCL-1 and drum #3598, can HCL-2 change only slightly as material density is increased above 18.9-g/cm<sup>3</sup>. From this figure it is apparent that the maximum production rates are achieved as the material density is increased. This indicates that the shielding effect gained by the increased density of the source material, and decreased alpha particle range, as it is moved closer to the inner surface of the HDPE bottle effectively negates closer proximity of the alpha emitting material to the inner surface of the HDPE bottle.

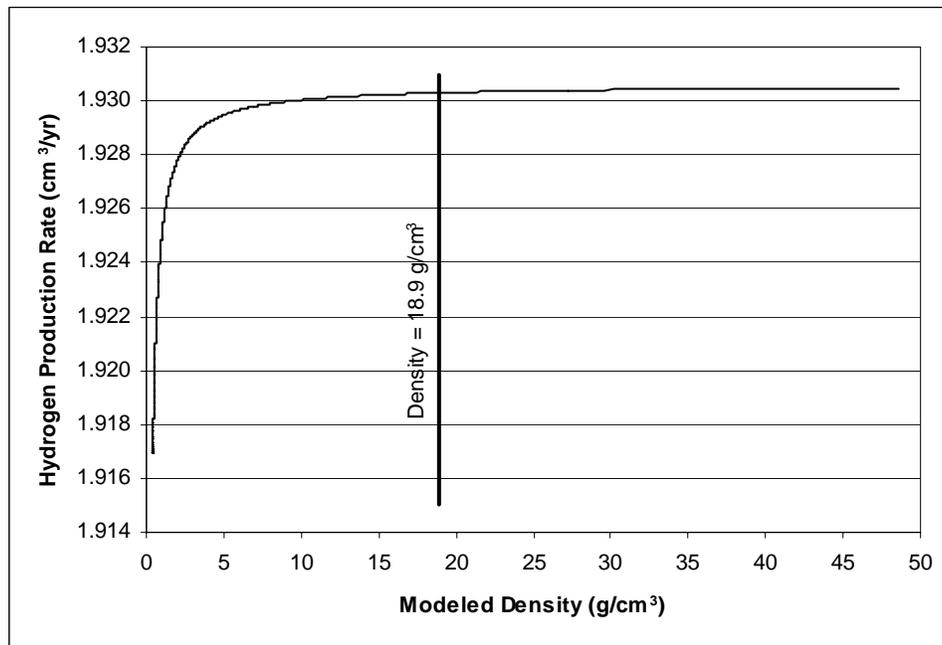
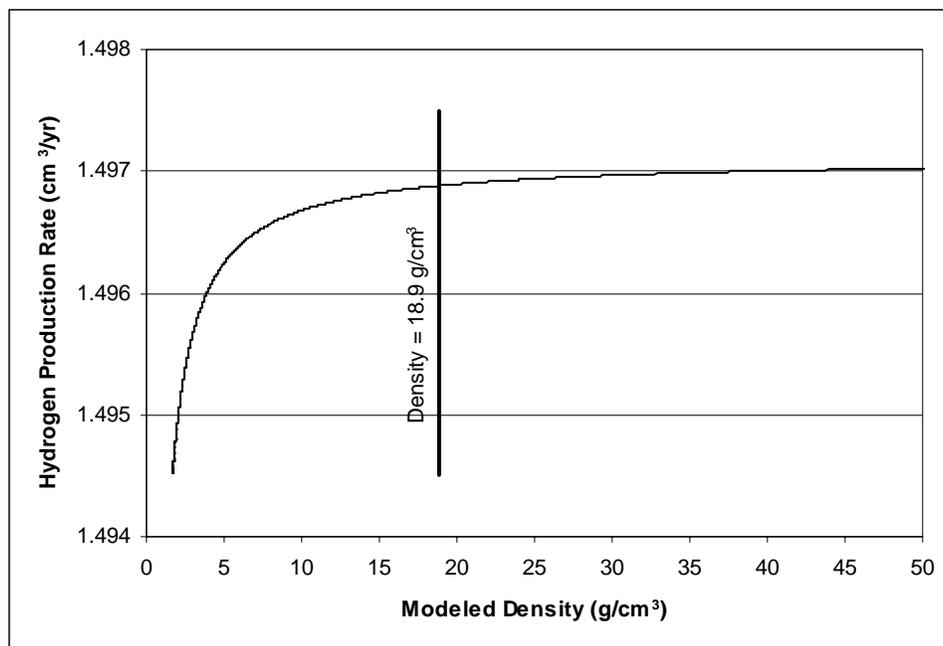


Figure 6 Hydrogen production rate as a function of modeled source density for drum #3031, can HCL-1.



**Figure 7 Hydrogen production rate as a function of modeled source density for drum #3598, can HCL-2.**

The following calculation provides an illustration of the use of the current methodology in the determination of hydrogen production rate in the DOT 6M 30-gallon drums assuming a density of  $18.9\text{-g/cm}^3$ . Based upon assumptions 2 and 5 in Section 4.1 and Eq. 2, the range of a 5.25 MeV alpha particle in source material contained within the HDPE bottle would be calculated as  $7.36\text{-}\mu\text{m}$ . The assumed density of  $18.9\text{-g/cm}^3$  requires that the total mixture volume (that is the sum of the contributing and noncontributing volumes in Figure 5) equal  $15.35\text{-cm}^3$  and  $62.99\text{-cm}^3$ , for drum #3031, can HCL-1 and drum #3598, can HCL-2, respectively. These values are determined as the material mass provided in Tables 1 & 3 divided by  $18.9\text{-g/cm}^3$ . The contributing volume illustrated in Figure 5 is determined with the aid of assumption 3 for HDPE dimensions and the calculated alpha particle range of  $7.36\text{-}\mu\text{m}$ . Based upon the dimensions provided in assumption 3, that is a 4-cm radius and a height of 14.2-cm, the total volume of the bottle is  $713.76\text{-cm}^3$ . The material of density  $18.9\text{-g/cm}^3$  would occupy a space that is outside the cylinder whose radius is  $(4 - 0.000736)\text{-cm}$  and whose height is  $(14.2 - 2(0.000736))\text{-cm}$ . The volume of this bounding cylinder is determined to be  $713.43\text{-cm}^3$ . The difference of the two volumes is the contributing volume of Figure 5 and is calculated to be  $0.33\text{-cm}^3$ . A simple ratio of  $\frac{1}{2}$  of the contributing volume to the total mixture volume determines the fraction of generated alpha particles that are assumed to deposit their energy in the HDPE. By this method, the value of D in Eq. 1 is calculated to be equal to  $7.53 \times 10^{-6}\text{-watts}$  and  $5.84 \times 10^{-6}\text{-watts}$ , respectively for HCL-1 and HCL-2. Eq. 1 then yields  $6.12 \times 10^{-8}\text{-cm}^3/\text{sec}$  or  $1.93\text{-cm}^3/\text{yr}$  for HCL-1 and  $4.74 \times 10^{-8}\text{-cm}^3/\text{sec}$  or  $1.50\text{-cm}^3/\text{yr}$  for HCL-2.

Based upon the calculated hydrogen production rates at an assumed source material density of  $18.9\text{-g/cm}^3$ , it is calculated that the molecular hydrogen production within HCL-1 will have generated  $54\text{-cm}^3$  of hydrogen in the 28-year period starting in 1992. This is equivalent to approximately 0.033-bar within the crimp sealed tin can and approximately 0.010-bar in the 2R within drum #3031. In the same time period, the production of molecular hydrogen in HCL-2 will have generated  $42\text{-cm}^3$  of hydrogen. This is equivalent to approximately 0.026-bar within the crimp sealed tin can and approximately 0.008-bar in the 2R within drum #3598. Table 4 provides a summary table of these calculations and results.

#### 4.2.4 Hydrogen Production in DOT 6M drum #20102

The UO<sub>3</sub> material in DOT 6M 30-gallon drum #20102 is reported to contain adsorbed water in the amount of 0.45-g. This water is conservatively assumed homogeneously mixed with alpha-producing uranium within the drum. That is to say that no credit is taken for the shielding effects of the UO<sub>3</sub> particles themselves in the current calculation. Based upon this assumption, the total energy deposition rate of  $4.09 \times 10^{-5}$ -Watts is absorbed by the water within the drum. Applying this rate as D in Eq. 1 yields a volumetric hydrogen production rate of  $1.61 \times 10^{-7}$ -cm<sup>3</sup>/sec or 5.09-cm<sup>3</sup>/yr for drum #20102. This production rate would result in a total hydrogen concentration of 4.0% by volume in the crimp-sealed can at 2019 and a hydrogen concentration of 1.4% by volume in the 2R at 2020. The molecular hydrogen concentration within the crimp-sealed can remains below the lower flammability limit for hydrogen in air until 2019.

**Table 4 Summary Table of Hydrogen Generation, Pressure Increase, and Concentration Results**

	<b>HCL-1 #3031</b>	<b>HCL-2 #3598</b>	<b>114/133 SAL #20102</b>
<b>H<sub>2</sub> Production Rate (cm<sup>3</sup>/yr)</b>	1.93	1.50	5.09
<b>Pressure Increase in Can (bar/yr)</b>	1.18E-03	9.12E-04	3.10E-03
<b>Pressure Increase in 2R (bar/yr)</b>	3.71E-04	2.88E-04	9.79E-04
<b>H<sub>2</sub> Concentration in Can (% vol)</b>	3.3 <sup>†</sup>	2.6 <sup>†</sup>	4.0 <sup>‡</sup>
<b>H<sub>2</sub> Concentration in 2R (% vol)</b>	1.0 <sup>†</sup>	0.8 <sup>†</sup>	1.4 <sup>†</sup>

<sup>†</sup> Concentration estimated at 2020.

<sup>‡</sup> Concentration estimated at 2019.

## 5.0 CONCLUSIONS

Three DOT 6m 30-gallon drums will be received in L-Area at SRS and stored for up to 13-years prior to final disposition at HB-Line by 2019/2020. These drums are scheduled for shipment from INTEC to SRS as part of the decommissioning effort of the INTEC facility. Bounding values for hydrogen gas generation rates, rates of pressure increase within the drums, and projected molecular hydrogen concentrations at 2019/2020 have been determined for all three drums based upon the assumptions listed in Section 4.1. Two of these drums, #3031 and #3598 contain polymeric bags and/or bottles that will be subject to radiolysis induced hydrogen gas generation. The maximum quantity of molecular hydrogen that can be expected to evolve in two DOT 6M 30-gallon drums in support of receipt and subsequent interim storage prior to canyon processing has been estimated based on the alpha radiolysis of polymers. An upper bound estimate for alpha energy available for radiolysis of the polymeric materials was determined from the volume fraction occupied by material in the bottle packed within an alpha range of the inner surface of the HDPE bottles. A conservative estimation of the hydrogen generation in DOT 6M drum #20102, can 114/133 SAL that contains UO<sub>3</sub> powder due to the radiolytic decomposition of water has also been completed. The results are summarized in Table 5.

**Table 5 Summary Table of Results**

	<b>HCL-1 #3031</b>	<b>HCL-2 #3598</b>	<b>114/133 SAL #20102</b>
<b>H<sub>2</sub> Production Rate (cm<sup>3</sup>/yr)</b>	1.93	1.50	5.09
<b>Pressure Increase in Can (bar/yr)</b>	1.18E-03	9.12E-04	3.10E-03
<b>Pressure Increase in 2R (bar/yr)</b>	3.71E-04	2.88E-04	9.79E-04
<b>H<sub>2</sub> Concentration in Can (% vol)</b>	3.3 <sup>†</sup>	2.6 <sup>†</sup>	4.0 <sup>‡</sup>
<b>H<sub>2</sub> Concentration in 2R (% vol)</b>	1.0 <sup>†</sup>	0.8 <sup>†</sup>	1.4 <sup>‡</sup>

<sup>†</sup> Concentration estimated at 2020.

<sup>‡</sup> Concentration estimated at 2019.

## 6.0 REFERENCE

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