A Methodology for Mixing Different Waste Types in an RH TRU Waste Shipment

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ABSTRACT

Each container in the payload of transuranic-(TRU) waste shipping cask must be shown to comply with the 5% (by volume) limit on hydrogen that is converted into an allowable hydrogen gas generation rate limit. The primary mechanism for gas generation during TRU waste transportation is by radiolysis of the waste materials. The G-value defines the hydrogen gas generating potential of a material by radiolysis. Based on a bounding hydrogen G-value the decay heat of a payload container can be converted into a hydrogen gas generation rate for comparison to the limit. Payload containers of different contents, packaging, and different bounding hydrogen G values may be assembled together as a payload, provided that all containers comply with the worst-case (i.e., lowest) calculated hydrogen gas generation rate limit. This is currently the only approved mixing option available for transport of remote-handled (RH) TRU wastes. For contact-handled (CH-) TRU waste, however, a methodology has been developed and approved by the U.S. Nuclear Regulatory Commission to allow mixing of containers of different contents within a single payload. For CH TRU payload mixing, container-specific limits are derived based on the characteristics of each container in the payload.

An added complexity in RH TRU wastes is the presence of significant gamma (γ) activity. The γ-radiation emitted from one drum may interact with the waste materials in other drums and cause radiolytic gas generation in adjacent drums of the payload. This paper presents a methodology for mixing RH TRU containers of different contents in a single payload and accounts for potential gamma ray interactions between containers. The container hydrogen gas generation rate may be calculated through a bounding analysis for comparison to the limit. Alternatively, the container rate may be derived through detailed analyses that account for shielding materials, payload geometry, particle transport, radiation absorption, and interaction effects, and actual percentages of radiolytic gas generating materials present. The paper presents example calculations based on both bounding analyses and more detailed and realistic analyses.

INTRODUCTION

There are numerous requirements for the control of transuranic (TRU) wastes as payload in U.S. Nuclear Regulatory Commission (NRC) certified shipping casks to ensure safe transport of the TRU wastes. Some of the payload parameters that are controlled include restrictions on: the

physical and chemical form; payload materials to ensure chemical compatibility among all constituents; the maximum pressure in the package during the transport period; the amount of hydrogen gas that might be generated during the transport period; the fissile material content; and the weight for the loaded shipping package.

This paper analyzes the restriction on hydrogen gas that may be generated during shipment. Specifically, the hydrogen gas concentration shall not exceed five percent by volume in all void volumes within the shipping package during the shipping period [1]. While the focus of this paper is on remote-handled (RH) TRU waste that is to be shipped in the CNS 10-160B cask, the proposed methodology is applicable to all other shipping packages that may be used to transport RH TRU wastes. The primary mechanism for gas generation during TRU waste transportation is radiolysis of the waste materials. The CNS 10-160B Safety Analysis Report (SAR) [1] has demonstrated that gas generation from other mechanisms such as chemical, thermal, or biological activity is insignificant for the TRU waste payload during a maximum 60-day normal shipping period or a 10-day controlled shipment. The 5% limit on hydrogen concentration is converted into an allowable hydrogen gas generation rate limit. Based on the bounding waste material's potential for hydrogen gas generation by radiolysis, characterized by a content-specific hydrogen G-value, the decay heat of the container can be converted into a container hydrogen gas generation rate that can then be compared to the limit.

TRU waste is classified into content codes, which give a description of the TRU waste material in terms of processes generating the waste, the packaging methods used in the waste container(s), and the generating site. To facilitate container load management operations (e.g., minimize the number of shipments) it is desirable to be able to mix TRU containers of different content codes within a single shipping package payload. Currently, payload containers of different content codes with different bounding G values and resistances to the release of hydrogen may be assembled together as a payload, provided the hydrogen gas generation rate limit for all payload containers within the payload is conservatively assumed to be the same as that of the payload container with the lowest hydrogen gas generation rate limit. For contact-handled (CH) TRU waste, however, a methodology has been developed and approved by the NRC to allow mixing of containers of different contents within a single payload. For CH TRU payload mixing, container-specific limits are derived based on the characteristics of each container in the payload.

An added complexity for RH TRU wastes is the presence of significant gamma (γ) activity. The γ-radiation emitted from one drum may interact with the waste materials in adjacent drums and cause additional radiolytic gas generation. This paper presents a methodology for mixing RH TRU containers of different content codes in a single payload and accounts for potential gamma ray interactions among containers. The allowable hydrogen gas generation rate limit per container is calculated by accounting for the properties of each of the other payload containers in the assembly, which may include dunnage (i.e., empty) containers similar to the CH TRU payload mixing methodology.

The container hydrogen gas generation rate may be calculated from the container decay heat based on bounding assumptions or by using various software codes to arrive at a more realistic rate for comparison to the limit. For the bounding analysis, the hydrogen gas generation rate will be derived assuming no shielding contributions, no radioactive decay, 100% gamma radiation absorption within the waste, and that each container contains 100% of the bounding worst case material for hydrogen radiolysis. This paper presents example limits calculations derived both through bounding assumptions and through more detailed and realistic analyses.

The CNS 10-160B is a cylindrical carbon steel- and lead-shielded cask (with a wall thickness of 12.7 centimeters, of which 4.8 centimeters is lead) designed to transport radioactive waste. It is transported in the upright position and equipped with steel-encased, rigid polyurethane foam impact limiters on the top and bottom (Figure 1). The Type B cask can accommodate ten 208-liter (55-gallon) drums arranged in two 5-packs (Figure 2) stacked one on another.

Fig. 1. CNS 10-160B transportation cask

Fig. 2. Arrangement of a 5-pack of drums for placement in the CNS 10-160B cask.

For this paper, the example proposed payload is comprised of ten RH TRU drums as follows:

- Six drums are Content Code SQ 321A (solid organic waste generated from decontamination and decommissioning activities)
- Two drums are Content Code SQ 321B (solid organic waste of pool filters and resins)
- One drum is Content Code SQ 314A (cemented inorganic process solids from research and development (R&D) operations)
- One drum is Content Code is SQ 322A (solid inorganic waste from R&D operations).

MIXING METHODOLOGY FOR ALLOWABLE HYDROGEN GAS GENERATION RATES

At steady state, the flow of hydrogen gas across each of the confinement layers is equal to the hydrogen gas generation rate. The maximum hydrogen gas concentration in a payload container with filter vents is reached at steady-state. That is, a filtered container with a hydrogen gas generation source has increasing concentrations of hydrogen with time until steady-state conditions are reached. For the purpose of the methodology, it has been conservatively assumed that all payload containers are at steady-state at the start of transport.

Once payload containers are sealed inside the cask, concentrations of hydrogen in the different layers increase due to the accumulation of hydrogen gas in the cask cavity. Some of the hydrogen gas generated during the transport period would accumulate in the container, with the remainder being released into the cavity. The maximum cask cavity mole fraction of hydrogen gas is obtained by assuming that all of the hydrogen gas generated in each payload container is instantaneously released into the cask cavity. The maximum hydrogen concentration in the innermost layer is then limited to 5% (by volume) or 0.05 mole fraction.

The maximum number of moles of hydrogen gas which can accumulate in the cask cavity is:

$$N_{gen} = \sum_{i=1}^{ng} CG_i \; t \qquad \text{(Eq. 1)}$$

where,

N_{gen}	=	Total moles of hydrogen gas generated (mole)
CG_i	=	Hydrogen gas generation rate per gas generator (i.e., container) "i" (mole/sec)
t	=	Shipping period duration (sec)
ng	=	Number of generators in the payload.

The maximum hydrogen gas mole fraction in the cask cavity is then equal to:

$$X_{H2} = \frac{N_{gen}}{N_T} = \sum_{i=1}^{ng} \frac{CG_i \, t}{N_T} \qquad \text{(Eq. 2)}$$

where,

X_{H2} = Maximum mole fraction of hydrogen gas in the cask cavity
N_T = Total moles of gas inside the cask cavity that are equal to:

$$N_T = \sum_{i=1}^{ng} CG_i\, t + \frac{PV_{void}}{RT} \quad \text{(Eq. 3)}$$

where,

P = Pressure inside the cask, conservatively assumed to be constant at 1 atm, because the amount of gas generated is much less than the total amount of air originally in the cavity
V_{void} = Void volume inside the cask cavity which includes the void volume of dunnage (i.e. empty) payload containers
R = Gas constant (0.082057 atm liter/mole-K)
T = Absolute temperature of air originally in the cask cavity (294 K).

It is assumed that the moles of hydrogen gas generated is much less than the moles of gas in the cask cavity at the time of package sealing:

$$\sum_{i=1}^{ng} CG_i\, t << \frac{PV_{void}}{RT} \quad \text{(Eq. 4)}$$

Therefore, from Equation (3):

$$N_T \approx \frac{PV_{void}}{RT} \quad \text{(Eq. 5)}$$

At steady-state the rate of hydrogen gas generation equals the rate of transport due to a hydrogen concentration gradient. Thus, the hydrogen gas generation rate per container, which will yield a maximum hydrogen gas concentration 0.05 mole fraction within the container is then calculated for each payload container "i" as the following:

$$CG_i = \frac{0.05 - X_{H2}}{r_{eff,i}} \quad \text{(Eq. 6)}$$

where,

$r_{eff,i}$ = The effective resistance to the release of hydrogen of payload container "i" (sec/mole)

The effective resistance is computed by summing the individual confinement layer resistances. The resistance of a layer is equal to the reciprocal of the release rate from that layer.

Substituting X_{H2} from Equation (2) into the previous equation and rearranging terms yields the following system of equations:

$$\left(r_{eff,1}+\frac{t}{N_T}\right)CG_1+\frac{t}{N_T}CG_2+\ldots+\frac{t}{N_T}CG_{ng}=0.05$$

$$\frac{t}{N_T}CG_1+\left(r_{eff,2}+\frac{t}{N_T}\right)CG_2+\ldots+\frac{t}{N_T}CG_{ng}=0.05$$

$$\begin{matrix}\bullet & \bullet & \bullet\\ \bullet & \bullet & \bullet\\ \bullet & \bullet & \bullet\end{matrix}$$

$$\frac{t}{N_T}CG_1+\frac{t}{N_T}CG_2+\ldots+\left(r_{eff,ng}+\frac{t}{N_T}\right)CG_{ng}=0.05 \qquad \text{(Eq. 7)}$$

The systems of equations represented by Equations (7) may be written in matrix form as:

$$\mathbf{A \bullet CG = 0.05} \qquad \text{(Eq. 8)}$$

where,

A	=	Matrix of gas generation rate coefficients (i.e., $r_{eff,i}$ and t/N_T terms)
CG	=	Column vector of allowable gas generation rates per container
0.05	=	Column vector of maximum hydrogen mole fractions within the innermost confinement layers.

The solution for the unknown allowable flammable gas generation rate for each generator is given as:

$$\mathbf{CG = A^{-1} \bullet 0.05} \qquad \text{(Eq. 9)}$$

where,

$\mathbf{A^{-1}}$	=	Inverse of matrix **A**.

Dunnage payload containers and containers that do not generate hydrogen are excluded from the system of linear equations.

For the example, all drums in the proposed shipment have the same packaging configuration (i.e., two layers of confinement namely a filtered drum liner inside a filtered 55-gallon drum). Thus, the effective resistance to the release of hydrogen is the same for all ten drums. In this case, the system of equations represented by Equations (7) may be simplified to a single equation for the allowable hydrogen gas generation rate per drum:

$$CG = \frac{0.05}{r_{eff} + \frac{10t}{N_T}} \quad \text{(Eq. 10)}$$

Compliance with the allowable hydrogen gas generation rate must be demonstrated over the entire temperature operating range of the CNS 10-160B cask. However, for purposes of this paper the analysis will focus on the maximum operating temperature of 348.6 K. At this temperature, the maximum effective resistance to the release of hydrogen is calculated as:

r_{eff} = 1/(4.98x10^{-6} mole/sec/mole fraction) + 1/(4.986x10^{-6} mole/sec/mole fraction)
r_{eff} = 401,606 sec/mole

From the ideal gas law, the total moles of gas in the cask cavity at the time the package is sealed for transport, N_T, is 80.34 mole.

Thus, from Equation (10), the allowable hydrogen gas generation rate per drum for a 10-day controlled shipment is:

CG = 0.05/{401,606 sec/mole + (10 days)(86400 sec/day)(10 drums)/80.34 mole
CG = 9.82x10^{-8} mole/sec/drum.

BOUNDING COMPLIANCE EVALUATION METHODOLOGY

The bounding compliance evaluation methodology is presented in the following discussion. The Radcalc code [2] was used to calculate decay heats and hydrogen gas generation rates for each drum. The steps in the methodology are summarized below:

- Quantification of the isotopic composition of each drum in the payload.
- Calculation of the drum decay heat plus error for each drum on the survey date using the Radcalc code. Thus, for the bounding analysis no credit is taken for decay that takes place between the survey date and the proposed shipment date.
- Definition of bounding hydrogen G values for each radiation type (α, β, and γ) and for each content code. The G values assume that waste is comprised of 100% of the worst-case material. The bounding G values by radiation type are summarized in Table I for the four different Content Codes.

Table I. Bounding Hydrogen Gas G Values by α-, β-, and γ- Radiation Type.

Content Code	Bounding Hydrogen Gas G Value (molecules/100 eV)		
	α-radiation	β-radiation	γ-radiation
SQ 321A	4.60	5.61	5.61
SQ 321B	2.51	3.06	3.06
SQ 314A	0.48	0.48	0.48
SQ 322A	0.016	0.016	0.016

- Calculation of the bounding radiolytic hydrogen gas generation rate on the assay date due to α-radiation and β-radiation for each drum. These calculations were performed by executing the Radcalc code (with one input file for each drum) with bounding hydrogen G values for α-radiation and β-radiation.

- Calculation of the total activity of each isotope in the payload by summing the isotope activity present in each of the ten drums. Calculation of the bounding radiolytic hydrogen gas generation rate on the assay date due to γ-radiation for each distinct Content Code-specific γ-radiation hydrogen G value. These calculations were performed by executing the Radcalc code (with one input file for each distinct γ-radiation hydrogen G value). Because there is one distinct bounding γ-radiation hydrogen G value for each of the four Content Codes, four executions of the Radcalc code were required. The resulting rate for each execution is based on the total isotopic activity from the ten drums. This conservatively assumes that all of the γ activity in ten drums is contained in each drum.

- Calculation of the bounding total radiolytic hydrogen gas generation rate for each drum by summing the rates for α-radiation plus β-radiation with the bounding γ-radiation rate. The total hydrogen gas generation rate for each drum was compared to the allowable hydrogen gas generation rate.

EXAMPLE BOUNDING HYDROGEN GAS GENERATION RATE

The calculation of the hydrogen gas generation rate for one of the two drums of Content Code SQ 321B will illustrate the application of the bounding compliance evaluation methodology. Based on calculations using radioassay data, the initial radionuclide compositions of the drum is established on the survey date. The isotopic information and container properties such as type (55-gallon drum), density, volumes, and α-radiation and β-radiation G-values of 2.51 and 3.06 molecules/100 eV, respectively, were entered into the Radcalc code. The hydrogen G-value for γ-radiation was set to zero as this contribution to hydrogen gas generation was analyzed separately as documented later in this paper. No decay period was specified for the bounding analysis. The Radcalc code calculated decay heat plus error for the drum based on the initial isotopic information was 0.005252 watt. The hydrogen gas generation rate from α-radiation and β-radiation was 7.17×10^{-2} cubic centimeters (cc) per hour (hr) at standard temperature and pressure (STP) (defined in Radcalc as 0 $^{\circ}$C and 101.325 kPa or 1 atm).

Because of the nature of gamma-rays, the potential for γ-radiation from one drum to cause radiolytic gas generation in another drum of the payload is accounted for as follows. The total initial activity of each isotope in the payload was calculated by summing the isotope activity present in each of the ten drums. The γ-radiation gas generation rate contribution for each drum was calculated as described in the following.

One Radcalc input file was created for each distinct γ-radiation G value (i.e. 5.61, 3.06, 0.48, and 0.016 molecules/100 eV as listed in Table I). Initial isotopic activities based on the sums of the ten drums were specified as input for each of the four cases. No decay period was specified for the bounding analysis.

The Radcalc user is able to select the gamma absorption curve based on the package size and shape analyzed. The gamma absorption curve is a method to estimate the fraction of the decay energy in the form of gamma rays that is absorbed inside the waste package. The gamma rays absorbed within the waste package are assumed to contribute to hydrogen gas production, while gamma rays that leave the package do not. Gamma absorption curves are models that take gamma ray energy and waste density as arguments and return an absorption percentage. The models are package specific, and were derived from numerous MCNP code runs over a range of gamma energies and waste densities. For purposes of bounding the γ-radiation gas generation rate contribution in each drum, the 100% Gamma Absorption Curve was used for all γ-radiation calculations. Thus, attenuation of the gamma energies by adjacent containers, and steel liners is not considered for the bounding analyses. The α-radiation and β-radiation G values were each set to zero as the rate contribution for those radiations were calculated previously. The Radcalc code was executed with each of the four input files. The hydrogen generation rate calculated by the Radcalc code in each case conservatively assumes all the activity for the entire payload is in single drum to arrive at a Content Code-specific γ-radiation radiolytic contribution. For example, with a γ-radiation G value of 3.06 molecules/100 eV, the Radcalc code calculated a hydrogen gas generation rate of 76.33 cc/hr at STP for the payload. This rate is conservatively applied as the rate for each of the two drums of Content Code SQ 321B.

For the example, the total radiolytic hydrogen gas generation rate for the drum of Content Code SQ 321B was calculated by summing the rates for α-radiation plus β-radiation (7.170×10^{-2} cc/hr) with the bounding γ-radiation rate (76.33 cc/hr) for a total hydrogen gas generation rate of 76.40 cc/hr. The total volumetric gas generation rate was converted into a molar gas generation rate of 9.47×10^{-7} mole/sec using the ideal gas law. This total hydrogen gas generation rate for the drum exceeds the rate limit of 9.82×10^{-8} mole/sec/drum by almost an order of magnitude. The drum could not be shipped using this bounding hydrogen gas generation rate methodology. However, the bounding analysis is based on extremely conservative assumptions. A more realistic analysis presented in the remainder of this paper indicates that the actual hydrogen gas generation rate of the drum is approximately 50 times lower than the bounding analysis rate.

REALISTIC ANALYSIS

A more detailed and realistic analysis may be applied to derive container hydrogen gas generation rates if one or more container rates based on a bounding analysis exceed the rate limits. A more detailed analysis accounts for radioactive decay, geometric and gamma absorption effects, shielding effects, and actual percentages of hydrogenous materials in the waste container.

The combined contribution of these effects will result in a much lower calculated hydrogen gas generation rates when compared to the bounding analysis. Each of these factors is discussed in the following.

Radioactive Decay

Containers of TRU waste, once generated, may be stored at a site for several years before shipment. In some cases, containers will be over three decades in age before shipment. For the example drum cited in this paper, the drum would have been surveyed 1,159 days prior to the shipment date. During this elapsed time, the Radcalc code calculates that radionuclide decay and in-growth will reduce the drum decay heat from an initial value of 0.005252 watt to 0.004608 watt. Thus, the hydrogen gas generation rate will be 93% of the initial rate at the time of shipment.

Shielding Effects

The one drum of Content Code SQ 322A has a 1-inch steel shield liner containing the RH waste that is then placed inside a filtered 55-gallon drum. This drum contains over 95% of the total activity of the payload and is the densest drum. It is this drum with nearly 7 watts of decay heat energy that may cause irradiation of the other payload drums. However, because it has the shielding only a fraction of the gamma ray energy will be effective in irradiating the other drums. Based on the isotopes and activities present on the proposed shipment date, the only gamma emitter of significance is Cs-137, which comprises 62% of the initial total activity in the ten drums. Bremsstrahlung radiation from Sr-90, which comprises 34% of the initial total activity in the ten drums, is ignored. The attenuation of the 1-inch thick steel shield for the highest activity drum is calculated using MicroShield [3] with Cs-137 as the source radionuclide. The exposure rate from a 55-gallon drum containing Cs-137 is determined with and without the 1" steel shield at a point 12" from the sidewall of the drum, i.e., at approximately the center of an adjacent drum.

The reduction in the gamma radiation is calculated from [3] as:

$$\mathrm{Rf} = \frac{I}{I_0} \qquad \text{(Eq. 11)}$$

where,

Rf	=	Exposure rate reduction factor
I_0	=	Exposure rate without shielding
I	=	Exposure rate with shielding

$$\frac{I}{I_0} = 0.35 \; or \; 35\% \qquad \text{(Eq. 12)}$$

Thus, the 1-inch shield in the highest activity drum will reduce the γ-irradiation of the other drums by at least 65% without accounting for the attenuation offered by the other steel drums and liners.

Geometric and Gamma Absorption and Distribution Effects

The payload is comprised of two 5-packs of drums as shown in Figure 3. Each 5-pack is comprised of drums that are arranged in a circular geometric arrangement (Figure 1). The geometry of the payload configuration and the effects of waste densities, gamma energies, gamma activity distribution within the payload, and container properties may be modeled using particle transport and interaction simulation Monte Carlo codes such as Geant4 [4] and MCNP, Version 5 [5]. Preliminary results indicate that the γ-radiation gas generation contribution is less than 10% of the bounding analysis rate derived from the Radcalc code assuming 100% gamma absorption and all of the γ activity for the entire payload located in each drum.

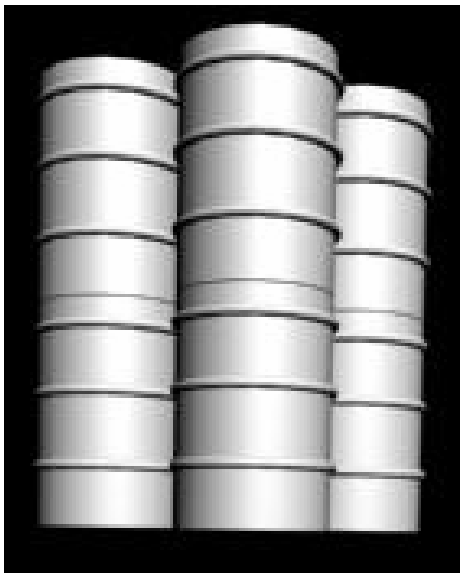

Fig. 3. Arrangement of 10 drums in the CNS 10-160B cask.

Hydrogenous Content of Waste

The bounding analysis assumed that all of the waste in a drum is comprised of only the bounding material for hydrogen radiolysis. In reality, waste records indicate that substantial quantities of inert (i.e., non-hydrogenous) inorganic materials (e.g., glass, metal) are present in drums assigned to solid organic waste content codes. If the fraction of inert materials is 50% in a drum, there would be a 50% reduction in the hydrogen gas generation rate compared to the bounding analysis derived rate.

Realistic Hydrogen Gas Generation Rate

The magnitude of the effects that are not accounted for in the bounding analysis are summarized in Table II. Geometry and gamma absorption effects have the most pronounced effect as exclusion of these from the bounding analysis over predicts the drum hydrogen gas generation rate by a factor of ten. If all the individual effects are accounted for (i.e., by taking the product of the individual factors), the drum hydrogen gas generation rate based on a detailed analysis is approximately 2.0% of the bounding analysis derived rate. For the example drum, the detailed

analysis hydrogen gas generation rate is 0.02 x 9.47×10^{-7} mole/sec or 1.89×10^{-8} mole/sec (20% of the rate limit).

Table II. Magnitude of Effects Not Accounted for in Bounding Analysis

Effect Accounted For in a Detailed Analysis	Percentage Reduction in Hydrogen Rate Compared to Bounding Analysis Derived Rate	Factor to Apply to Bounding Hydrogen Gas Generation Rate
Radioactive Decay	7%	0.93
Shielding	65%	0.35
Geometry and Gamma Absorption	90%	0.10
Hydrogenous Content of Waste (assumed for illustration purposes)	50%	0.50
Combined Effects	98%	0.02

CONCLUSION

Currently for RH TRU waste shipping packages, payload containers of different content codes may be assembled together as a payload, provided the hydrogen gas generation rate limit for all payload containers within the payload is conservatively assumed to be the same as that of the payload container with the lowest hydrogen gas generation rate limit. This paper describes a methodology for mixing RH TRU containers of different content codes in a single payload similar to the CH TRU payload mixing methodology. The container-specific limits are derived based on the characteristics of each container in the payload and accounts for the benefits of dunnage containers. The mixing methodology also accounts for potential gamma ray interactions between containers that are not present in CH TRU wastes. The container hydrogen gas generation rate may be calculated through a bounding analysis for comparison to the limit. However, the bounding analysis will typically over estimate the rate by at least an order of magnitude. Thus, the proposed mixing methodology recommends that a detailed analysis be performed accounting for decay, shielding, payload geometry and gamma absorption fractions, and actual percentages of hydrogenous materials in the waste. The proposed methodology provides an alternate method of demonstrating compliance with the 5% hydrogen gas concentration limit. In summary, through a demonstration using a payload-specific mixing (either bounding or detailed) analysis, the individual payload containers within a payload can be shown to have hydrogen gas generation rates below payload-specific calculated limits.

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