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March 7, 2011

Washington TRU Solutions LLC
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Attention: Mr. Rick Chavez

Subject: Gas Generation from WIPP Waste during the Preclosure Period

Dear Mr. Chavez:

This letter documents an analysis of the gas generation rate in a waste-filled panel during the 30-year preclosure period, when the WIPP repository is operational. The analysis is based on the gas generation model for the 2004 Performance Assessment Baseline Calculation (PABC-2004) (Leigh et al., 2005). The PABC-2004 is an appropriate starting point for this analysis because it was the basis for the 300-year performance demonstration in the Renewal Application for the WIPP's RCRA Permit. This Renewal Application was subsequently approved by the New Mexico Environment Department (NMED) on November 30, 2010 (NMED 2010), so the PABC-2004 represents the baseline for the Permit.

As used in this letter, closure refers to the permanent closure of the WIPP repository, after waste emplacement is complete and permanent closures are installed in the waste-filled panels. During the operational period, waste-filled panels are temporarily "sealed" with steel bulkheads to prevent personnel access and to restrict ventilation air flow through waste-filled panels. The steel bulkheads are not an air-tight seal because the bulkheads may deform as the access drifts creep closed and because leakage may occur through gaps between the steel bulkheads and the adjacent rock walls.

1.0 Conditions during the Preclosure Period

The gas generation model for the PABC-2004 is designed to predict long-term gas generation during 10,000 years of postclosure performance. The long-term environment after permanent closure of the repository may be different than the environment in a sealed panel during the 30-year operational period. During the operational period, the mine ventilation system is continually circulating fresh air that is likely to prevent oxygen from being completely depleted in the sealed panels. While ventilation air cannot circulate directly into a sealed panel, oxygen can diffuse across the steel bulkheads that seal Panels 3 and 4 from the ventilation air flow. Oxygen diffusion is also expected across the brattice cloth barriers that seal the individual rooms in a waste-filled panel. On the other hand, oxygen may be completely depleted in Panels 1 and 2, which are temporarily sealed with a 12-foot-thick wall of mortared concrete blocks. Since Panels 5 through 8 will be sealed with steel bulkheads and brattice cloth barriers, oxygen is unlikely to be completely depleted in most of the sealed panels during the preclosure period. Under these conditions, oxic corrosion and aerobic microbial degradation may be active processes during a portion of the preclosure period.

This situation is opposite to that after permanent closure, when the chemical environment in the repository after closure is expected to be reducing. Any gaseous or dissolved oxygen present in the repository will be consumed quickly, either by aerobic microbes or by oxic corrosion (DOE 1996, Section 6.4.3.4). After oxygen is depleted, anoxic corrosion and anaerobic microbial degradation are predicted to the dominant gas generation mechanisms in the repository.

A second preclosure/postclosure difference is that sealed panels are likely to be humid, with minimal amounts of standing brine, during the preclosure period. The WIPP underground facility is observed to be dry: seeps of brine into the facility are small and isolated, and these seeps evaporate into the ventilation air circulating through the facility. In effect, the disturbed rock zone (DRZ) in the back (roof) and sides of the waste-filled rooms and access drifts is dewatered by circulation of ventilation air until a panel is sealed. In addition, the intact host rock (i.e., the bedded halite) surrounding the DRZ has extremely low permeability, which limits brine inflow during the 30-year preclosure period. Under these conditions, the rooms in a sealed panel are expected to be humid, with little free standing brine on the floor of a room or in the waste.

By way of contrast, the PABC-2004 conservatively assumes that the DRZ surrounding the waste emplacement areas is fully saturated and drains immediately after repository closure. The analysis in this letter is based on humid conditions, with negligible standing water, because this is consistent with observations in the underground facility.

2.0 Gas Generation Model in the PABC-2004

The WIPP gas generation model considers three potential sources of gas in the repository after closure:

$$q = q_{rgc} + q_{rgm} + q_{rgr}, \tag{1}$$

where q_{rgc}, q_{rgm}, and q_{rgr} [kg/m^3/s] are the rates of gas production from anoxic corrosion of iron-based materials, from anaerobic microbial degradation of cellulosic, plastics, and rubber (CPR) materials, and from radiolysis, respectively. The potential contributions from each of these processes to gas production during the preclosure period are explained in the next subsections.

2.1 Radiolysis

Hydrogen gas production from radiolysis been analyzed by WTS for the waste emplaced in Panel 3 of the WIPP repository (Devarakonda 2006). The analysis considered hydrogen production from the radiolysis of water and from radiolysis of plastics and other organic materials in the waste. The total hydrogen generation rate was estimated as 4.497×10^{-5} moles/s for a waste-filled panel (Devarakonda 2006, Table 1). Converting this rate for a 55-gallon drum on a yearly basis:

$$(4.497\times10^{-5}\text{ moles/s/panel})(3.156\times10^{7}\text{ s/year})(1\text{ panel/81,000 drums}) = 0.0175\text{ moles/drum/yr}$$

The total hydrogen generation rate is based on "*G*" values for the various waste materials, where *G* defines the number of molecules of hydrogen produced per 100 eV of energy absorbed. The *G* values are based on head-space hydrogen data from actual containers, when available, or on mean values for transportation analysis. These *G* values overestimate the gas generation rate because, as the waste matrix is depleted, the hydrogen generation rate is expected to decrease asymptotically to very low values (Devarakonda 2006, Figure 1). The value of 0.0175 moles hydrogen per drum per year is therefore considered an upper bound, as confirmed by the screening analyses discussed next.

The potential for postclosure radiolytic gas generation is also considered in screening analyses for features, events and processes (FEPs) that are included in or excluded from performance assessment (PA). Radiolytic gas generation has been eliminated from PA models on the basis of low consequence to the performance of the disposal system (DOE 2004, Attachment SCR, Section SCR-6.5.1.6 (FEP W52) and Section SCR-6.5.1.7 (FEP W53)). The relevant results for postclosure gas generation are as follows:

- Molecke (1979) compared experimental data on gas production rates caused by radiolysis of cellulose and other waste materials with gas generation rates by other processes, including bacterial (microbial) waste degradation. The most probable range of gas generation caused by radiolysis of cellulosic material is 0.005 to 0.011 moles per year per 55-gallon drum (Molecke 1979, p. 4) (DOE 2009, Attachment SCR, Section SCR-6.5.1.7.3).
- Reed et al. (1993) performed experiments for radiolytic gas generation from brine (DOE 2004, Attachment SCR, Section SCR-6.5.1.6.3). These experiments involved WIPP-relevant brines that were spiked with $^{239}Pu(VI)$ at concentrations between 6.9×10^{-9} and 3.4×10^{-4} molal. Based on the experimental results, the radiolytic gas generation was estimated by assuming that the total excavated volume of the repository, 436,000 m^3, was completely filled with brine. This is clearly an excessively conservative assumption during the preclosure period because brine is not observed to be flowing out of any sealed panels, as would occur if such a large volume of water was present in these panels. Assuming instead that each panel has 100 m^3 (26,400 gallons) of brine, the radiolytic gas production from brine is calculated as:

$$(0.6\ \text{mol/drum/year})(100\ \text{m}^3\text{/panel})(10\ \text{panels})/(436{,}000\ \text{m}^3) = 0.0014\ \text{mol/drum/yr}$$

The general conclusion from these analyses is that radiolytic gas generation will not produce significant amounts of gas during the preclosure period.

2.2 Corrosion

Gas production from corrosion of iron-based materials is expected to be insignificant during the preclosure period. Conditions in a sealed panel are expected to be humid, with insignificant amounts of standing brine during the preclosure period. The gas production rate from corrosion of iron-based materials in a humid, anoxic environment is zero in long-term performance assessment because significant corrosion in a humid environment has never been observed under WIPP-relevant conditions. More specifically, steel specimens exposed at 30°C to the vapor phase of Brine A with an N_2 overpressure of 10 atm showed no discernable corrosion reaction

(Telander and Westerman, 1997, Section 7.1). Similar testing with 10 atm of CO_2 or 5-atm of H_2S also showed insignificant corrosion (Telander and Westerman, 1997, Sections 7.2 and 7.3).

During the preclosure period, oxygen may not be depleted from all panels. When oxygen is present, oxic corrosion could occur if the water vapor in humid air condenses on exposed iron-based surfaces (DOE 1995, Section 4.1.1.1). If oxic corrosion does occur, it would consume atmospheric oxygen, resulting in a decrease in the panel pressure. This decrease in pressure is ignored in this analysis because conditions during the preclosure period make oxic corrosion unlikely for two reasons. First, the paint on 55-gallon drums and other waste containers prevents corrosion of the outside surfaces by protecting these surfaces from direct exposure to air and water (Atkins 1998, Section 29.7(b)). Second, the drums and other waste containers will remain sealed until lids fail or containers crush from creep closure of the rock walls or from roof fall. A sealed container prevents corrosion of the inner surfaces and of iron-based materials in the waste because air and water cannot access the interior spaces.

The conclusion regarding insignificant gas production from corrosion is confirmed by monitoring for explosive gases in waste-filled rooms and panels of the repository. The monitoring program has provided long-term data on the generation of hydrogen and methane gases in sealed panels and rooms. Only very low levels of hydrogen have been detected by long-term monitoring, implying that significant corrosion of iron-based materials is inhibited by the dry environment underground and by the surface coatings on the waste containers. It follows that gas production from oxic or anoxic corrosion of iron-based materials should not be a major source of gas during the preclosure period.

2.3 Microbial Degradation

Since radiolytic gas production and corrosion of iron-based materials will not produce significant quantities of gas during the preclosure period, the potential for microbial gas generation is examined in detail. The rate of gas production from microbial degradation, q_{rgm} [kg/m^3/s], is defined as (DOE 2004, Appendix PA, Section PA-4.2.5 and Nemer and Stein 2005, Section 5.5.3)

$$q_{rgm} = \left(R_{mi} S_{b,eff} + R_{mh} S_g^*\right) D_C\, y(gas \mid C) M_{gas} B_{fc}, \qquad (2)$$

where R_{mi} is the rate of cellulose biodegradation under inundated conditions [mol C/ kg $C_6H_{10}O_5$/s], where $C_6H_{10}O_5$ is the assumed molecular composition of cellulose;

R_{mh} is the rate of cellulose biodegradation under humid conditions [mol C/ kg $C_6H_{10}O_5$/s];

$S_{b,eff}$ is the effective brine saturation due to capillary action in the waste materials [-];

S_g^* is (1 - $S_{b,eff}$) if $S_{b,eff} > 0$ or is 0 if $S_{b,eff} = 0$;

D_C is the average density of cellulosic materials in the waste [kg biodegradable material/m^3 of disposal volume];

$y(gas|C)$ is the average stoichiometric factor for microbial degradation, i.e., the moles of gas generated per mole of carbon consumed by microbial action [mol gas/mol C];

M_{gas} is the molecular weight of the gas [kg gas/mol gas]; and

B_{fc} represents the uncertainty in whether or not microbial gas generation could be realized in the WIPP at the experimentally measured rates in the laboratory [-].

3.0 Expected Microbial Gas Generation Rate During Preclosure

Microbial consumption of biodegradable materials will be the dominant gas generation process during the preclosure period, as explained in Section 2. Microbial consumption of biodegradable materials will produce carbon dioxide, nitrogen, and hydrogen sulfide, based on the reaction pathways for denitrification and sulfate reduction (Nemer and Stein 2005, Section 5.6). The rate of gas production, Q_{rmg} [moles of gas/m^3/s], is defined as:

$$Q_{rmg} = \frac{q_{rgm}}{M_{gas}} = \left(R_{mi} S_{b,eff} + R_{mh} S_g^{*}\right) D_C \, y(gas \mid C) B_{fc}, \tag{3}$$

The mean or expected values of the individual terms on the right-hand side of Equation (3) are evaluated in the following subsections.

3.1 Rate of Cellulose Biodegradation

The rates of cellulose biodegradation under inundated and humid conditions are defined by two uniform distributions (Nemer and Stein 2005, Section 5.5 and Table 5-5):

$$R_{mi} = Uniform[0.00097, 0.0176] \text{ mol C/kg } C_6H_{10}O_5\text{/year}, \tag{4}$$

$$R_{mh}^{*} = Uniform[0.0, 0.0324] \text{ mol C/kg } C_6H_{10}O_5\text{/year}, \tag{5}$$

where the molecular formula for cellulose is assumed to be $C_6H_{10}O_5$. The experimental data from gas generation experiments indicate that the maximum humid microbial gas generation rate in Equation (5) is greater than the maximum inundated gas generation rate in Equation (4). This is physically unrealistic because, given the lack of water under humid experimental conditions, the humid rate is expected to be much less than the rate for microbes inundated with brine. The humid rate is therefore constrained by the sampled value of the inundated rate:

$$R_{mh} = Min(R_{mi}, R_{mh}^{*}). \tag{6}$$

In other words, $R_{mh} \leq R_{mi}$. If $S_{b,eff}$ is greater than zero, the first term in Equation (3) can then be bounded as:

$$\begin{aligned} R_{mi} S_{b,eff} + R_{mh} S_g^{*} &= R_{mi} S_{b,eff} + R_{mh}(1 - S_{b,eff}), \\ &\leq R_{mi} S_{b,eff} + R_{mi}(1 - S_{b,eff}), \\ &\leq R_{mi}. \end{aligned} \tag{7}$$

If $S_{b,eff}$ equals 0, the gas generation rate is zero and Equation (7) still provides an upper bound. The mean rate of cellulose biodegradation under inundated conditions, $\overline{R}_{mi}$, is defined by the minimum and maximum values of the uniform distribution in Equation (4):

$$\begin{aligned} \overline{R}_{mi} &= 0.5(0.00097 + 0.0176) \text{ mol C/kg } C_6H_{10}O_5\text{/year}, \\ &= 0.0093 \text{ mol C/kg } C_6H_{10}O_5\text{/year}. \end{aligned} \tag{8}$$

The value in Equation (8) also provides an upper bound for the microbial gas generation rate under humid conditions, as noted in Equation (6).

Equation (8) is based on data for anaerobic degradation. During the preclosure period, the sealed panels may maintain oxygen levels that allow aerobic bacteria to provide some microbial degradation of CPR materials. Data from microbial gas generation experiments demonstrates that aerobic degradation rates are lower than anaerobic rates under a range of experimental conditions (Francis et al. 1997, Executive Summary). For example,

- Aerobic inoculated and amended samples produced 0.001 mL total gas/gram cellulose/day and 0.01 μmol CO_2/gram cellulose/day up to 1228 days. The rate was higher in anaerobic inoculated amended samples, which produced 0.004 mL total gas/gram cellulose per day and 0.05 μmol CO_2/gram cellulose/day.
- With excess nitrate, the aerobic inoculated and amended samples produced 0.008 mL total gas per gram cellulose per day, and 0.1 μmol CO_2 per gram cellulose per day. The rate is greater for anaerobic inoculated and amended samples with excess nitrate, which produced 0.01 mL total gas/gram cellulose/day and 0.2 μmol CO_2/gram cellulose/day.

The mean microbial gas generation rate in Equation (8) therefore bounds the gas generation rate in humid or inundated conditions and for aerobic or anaerobic environments.

3.2 Probability-Weighted Average CPR Mass per Drum, $\overline{D}_C V_{drum}$

The equivalent mass of cellulosic materials per drum, $\overline{D}_C V_{drum}$, is evaluated in Table 1, based on the densities of CPR materials in the inventory for the PABC-2004 (Nemer and Stein 2005, Table 5-1), on the volume of a 55-gallon drum, 0.208 m^3 (SNL 1992, Table 3.1-2), and on the carbon conversion factor of 1.7 for plastic versus cellulosic and rubber materials (Wang and Brush 1996, Equation (6)). For the PABC-2004, 100% of the realizations have the potential to degrade cellulosic materials; plastic and rubber materials also degrade in 25% of the realizations (Nemer and Stein 2005, Section 5.4). Using the data in the right-hand column of Table 1, the probability-weighted mass of equivalent cellulosic material per drum is calculated as:

$$\begin{aligned}\overline{D}_C V_{drum} &= 12.48 + 1.93 + (0.25)(2.70 + 1.39 + 15.2 + 2.83 + 6.01 + 1.10), \\ &= 21.7 \,\text{kg}\ C_6H_{10}O_5\,/\text{drum}.\end{aligned} \tag{9}$$

3.3 Average Stoichiometric Factor for Microbial Gas Generation, y(gas|C)

The stoichiometry of gas generation is based on a stoichiometric factor *y*, which is defined by the overall reaction for microbial degradation as:

$$C_6H_{10}O_5 + \mathit{unknown} \rightarrow 6y\ \mathit{gas} + \mathit{unknown} \tag{10}$$

The units of *y* are moles of gas per mole of Carbon degraded. Methanogenesis was removed as a long-term pathway for microbial degradation in the PABC-2004. This is consistent with the expectation of aerobic and dry conditions during preclosure, which preclude methanogenesis. The value of *y* is 0.486 when cellulose alone degrades and 0.495 when all CPR materials degrade (Nemer and Stein 2005, Table 5-6). The upper value of 0.495 is used in this analysis.

Table 1. Calculation of Equivalent Cellulosic Mass per Drum from Inventory of CPR Materials

Material	Density (kg/m^3)	Drum Volume (m^3/drum)	Conversion Factor (-)	Equivalent Cellulosic Mass (kg)
Average density of cellulosic materials in CH waste	60	0.208	1	12.48
Average density of cellulosic materials in RH waste	9.3	0.208	1	1.93
Average density of rubber materials in CH waste	13	0.208	1	2.70
Average density of rubber materials in RH waste	6.7	0.208	1	1.39
Average density of plastic materials in CH waste	43	0.208	1.7	15.2
Average density of plastic materials in RH waste	8	0.208	1.7	2.83
Bulk density of plastic liners in CH waste	17	0.208	1.7	6.01
Bulk density of plastic liners in RH waste	3.1	0.208	1.7	1.10

Equivalent Cellulosic Mass = Density * Drum Volume * Conversion Factor; Density data from (Nemer and Stein 2005, Table 5-1).

3.4 Average Value of the Uncertainty Factor, $\overline{B}_{fc}$

The conditions inside the WIPP repository may be quite different from the conditions in laboratory experiments that simulated microbial gas generation. The laboratory experiments were designed to promote growth (Nemer and Stein 2005, Section 5.5.3), so microbial action within the WIPP may be reduced from that observed in the experiments. The uncertainty about microbial action under WIPP-relevant conditions includes whether microbes will survive for a significant fraction of the 10,000-year regulatory period, whether sufficient water will be present, and whether sufficient quantities of biodegradable substrates will be present. Due to these uncertainties, an additional sampled parameter, B_{fc}, was added as a multiplicative factor to Equations (2) and (3) for the rate of gas generation. B_{fc} is defined as a uniform distribution between 0 and 1. The expected or mean value of $\overline{B}_{fc}$ is 0.5.

3.5 Calculation of Expected Preclosure Microbial Degradation Rate

Using the values defined in Sections 3.1 through 3.4, the expected or average rate of gas production [moles of gas/year/drum], is defined as:

$$\begin{aligned}\overline{Q}_{rmg} &= \overline{R}_{mi}\overline{D}_{C}V_{drum}y(gas \mid C)\overline{B}_{fc},\\ &= (0.0093\,\text{mol C/kg } C_6H_{10}O_5\text{/year})(21.7\,\text{kg } C_6H_{10}O_5\,/\,\text{drum})(0.495\,\text{mol gas}\,/\,\text{mol C})(0.5),\\ &= 0.0499\,\text{mol gas}\,/\,\text{drum/year},\\ &\approx 0.05\ \text{mol gas}\,/\,\text{drum}\,/\,\text{year}.\end{aligned}$$

4.0 Comparison to Previous Best Estimate of Gas Production

The previous estimate of the expected rate of gas production for panel closure design was 0.1 moles gas/drum/year (DOE 1995, Section 4.1.1.3). This estimate was based an approach that is very similar to the approach in this letter:

- It was considered highly unlikely that significant amounts of hydrogen will be generated by corrosion during the operational period because of the lack of a credible source of moisture required to drive this process (DOE 1995, Section 4.1.1.1)
- The contribution from radiolytic gas generation was not considered in the analysis because it was several orders of magnitude lower than the expected contribution from microbial gas generation (DOE 1995, Section 4.1.1.2).
- The best estimate gas generation rate of 0.1 moles gas/drum/year from microbial degradation is based on humid conditions in the repository. The best estimate value was selected, rather than an upper or lower bound, because of the large number of drums that are placed in a panel (DOE 1995, Section 4.1.1.3).

It is likely that the gas generation rate from (DOE 1995) is greater than the gas generation rate in Section 3.5 of this letter because the microbial gas generation model was changed for the PABC-2004 (Leigh et al. 2005, Sections 2.2 through 2.4). These changes included the probability of cellulose degradation, the reduction in the long-term rate for inundated microbial gas generation, and the removal of methanogenesis from the model. The reduction in the long-term rate for microbial gas generation is likely to be the primary cause of the reduction from 0.1 to 0.05 moles/drum/year; however, no attempt was made to repeat the original calculations with the new gas generation parameters and new inventory of CPR materials.

5.0 Recommendation for Panel Closure Design

I recommend that WTS use a gas generation rate from WIPP waste of 0.1 moles of gas/drum/year for panel closure design calculations. This value is consistent with previous work (DOE 1995), and is conservative relative to the value of 0.05 moles of gas/drum/year derived in Section 3 of this letter. This gas is expected to be a combination of carbon dioxide, nitrogen, hydrogen sulfide and minor amounts of hydrogen, based on the reaction pathways for denitrification and sulfate reduction (Nemer and Stein 2005, Section 5.6) and on the results from long-term monitoring for explosive gases in waste-filled rooms and panels, respectively.

Please feel free to contact me with any questions or issues related to this analysis.

Sincerely,

Michael B. Gross

cc: Tom Klein
Wille Most
Bob Kehrman

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