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Memorandum

To: Engineering File – General's Tanks Atmosphere Control and Preliminary Characterization Project, 30% Design

Re: General's Tanks Radio-Chemical Environment and Associated Hazards

This memorandum is intended to document work performed to define the radionuclide and chemical content of the General's Tanks at Material Disposal Area A at Technical Area 21 of the Los Alamos National Laboratory. Further, this memorandum addresses potential combustion or deflagration of the theoretical maximum content of hydrogen gas that may have been radiolytically generated in the tanks as a result of decay of radioactive materials in the presence of water. The potential effects of a deflagration in one of the tanks have also been modeled and described relative to existing overburden soils and relative to nearby structures.

The attachment to this memorandum, written as a White Paper, contains a summary of the calculations and analyses that have been performed in support of the modeling of the tank contents and the effects of inadvertent ignition of tank contents.

Todd R. Clark

6-9-06

Date

c: J. D. Ritchey
P. G. Torpy

Correspondence file number: 7958-033

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WHITE PAPER

General's Tanks Radio-Chemical Environment and Associated Hazards

Background

The General's Tanks are two buried steel tanks located within the Nuclear Environmental Site (NES) at Material Disposal Area (MDA) A in Technical Area (TA) 21 at the Los Alamos National Laboratory (LANL). The tanks each measure 62 feet and 10 inches in length and 12 feet in diameter, with approximate internal capacities of 50,000 gallons each based on original design drawings (LASL 1945, 24448 and LASL 1945, 24450). These original drawings coupled with descriptions of later surface soil additions represent the most useful descriptions of the tank design and placement.

The tanks were constructed in 1945 and filled with liquids containing trace quantities of plutonium between 1946 and 1949 (Purtyman 1969, 00520). The liquids are thought to have been held for potential future recovery pending anticipated improvements in chemical separation efficiency.

A liquids retrieval project was initiated in 1974 (LANL 1996, LA-UR-96-1283). Both tanks were accessed in 1974 by excavating through overburden soils, cutting through the 8-inch reinforced concrete slab and removing additional loose soils to get to the central portion of each tank shell and by cutting through each tank's shell (Tucker 2003, 092454). By the time the tanks were accessed, only about 0.7% of the original activity of radionuclides was still in solution in the tank liquids (Voelz 1973, 00483). Relatively high pH in each tank supported precipitation of the radionuclide metals.

After completing the liquids retrieval, the tanks were sealed and excavation holes were refilled in 1985 (LANL 2005, 88052.65). Samples of the sludge left in each tank after retrieval of tank liquids were collected in 1981 (Bucholz 1981, 00459). The sample results and records from interviewing current LANL employees who worked on the liquids retrieval project represent the most recent data located for describing the tank contents.

Objective

A reliable estimate of the tank contents is desired, in support of determining the risks associated with accessing the tanks. Using the information collected from drawings, samples, and interviews, a conceptual model of the tank contents and the associated hazards can be described or bounded. The objective of this white paper is to utilize existing information combined with calculations and evaluation approaches to define or bound the tank contents and provide an understanding of the hazards associated with accessing and working with the tanks.

Radio-Chemical Constituents

Radionuclide Inventory

As early as 1971 there was interest in determining the contents of the General's Tanks based on the concern that the tanks would need to be removed at some point (Meyer 1971, 00515). An estimate for the combined tanks contents based on records of input was reported as 344 grams total alpha equivalent (Gibson 1971, 00513). The writer speculated that the majority of the activity should be expected from ^{241}Am in-growth from ^{241}Pu decay. However, in 1981 when samples were taken of the sludges left in each tank after retrieval of the tank liquids, the activity associated with ^{241}Am was far exceeded by that of the ^{239}Pu in the sediments (Bucholz 1981, 00459).

The overall list of analytes covered by the 1981 samples and the late date at which they were taken make them the most useful of all of the available sample data for forecasting the current contents of the General's Tanks. In order to calculate radionuclide inventory in the two General's Tanks in 2007, the year the tanks would be expected to be accessed under the General's Tanks Atmosphere Control and Preliminary Characterization Project, the following assumptions have been made:

1) Sediment depth in each tank is 6 inches, based on a visual observation recorded in an interview with David Salazar conducted by Keith Tucker, April 9, 2003 (Tucker 2003, 092454), resulting in 101.26 cubic feet or 2,867,364 cubic centimeters of sediment in each tank (based on the 12-foot diameter of the tanks and the 62.foot 10 inch length of the tanks)(LASL 1945, 24448). Overall volume of each tank is 6,684.03 cubic feet or 1.89E+08 cubic centimeters.

2) Wet specific gravity of sludge in each tank is 1.37 (85.488 pound per cubic foot or 1.37 grams per cubic centimeter) (8,656.52 pounds or 3,928,289 grams). This assumption is based on specific gravity determined for similar tank sediments at the Oak Ridge National Laboratory.

3) The 1981 analytical data (Bucholz 1981, 00459) is representative of the tank sediments and tank liquids.

1981 General's Tanks Analytical Sample Radiological Data

Analysis	West Tank (KOH) (d/m/g)[1]	East Tank (NH_4OH) (d/m/g)[1]
Alpha	3.60E+07	1.11E+07
^{238}Pu	2.2E+05	6.5E+04
^{239}Pu	3.9E+07	1.0E+07
^{241}Am	9.2E+05	2.4E+05
Total U	2.2E+04	6E+03

[1] Dry basis

4) The date of fill for the tanks is assumed to be 1947. No addition of materials to the tanks after 1947 is considered. RadCalc 4.0 was used to perform aging/decay calculations for tank radionuclides. Annual dates refer to the first day of year. This assumption is based on various recounts, including a memo from George L. Voelz (Voelz 1973, 00483).

5) No ^{240}Pu or ^{241}Pu activities were reported in the 1981 analyses. If all of the ^{241}Am identified in the 1981 samples was a result of decay of original ^{241}Pu content, the original 1947 (assuming all of the tank contents date to 1947) ^{241}Pu content would have been about 66 curies (Ci) based on an iterative back-calculation approach. ^{241}Am content from original reactor products may also have been present in the 1947 tank liquids, since americium was generally not removed well by the bismuth phosphate separation process in use at Hanford in the 1940s, but there is no practical method to assume an initial ^{241}Am value. The calculated 1947 ^{241}Pu value would be overestimated as a result. DOE defines weapons-grade plutonium as plutonium with less than 7% ^{240}Pu. The DOE HDBK-1145-2001 cites weapons-grade plutonium values of 0.4% ^{241}Pu, 6.0% ^{240}Pu, and 93.6% ^{239}Pu by weight (or a weight ratio for ^{241}Pu to ^{239}Pu of 0.0043 to 1. However, the 1981 ^{241}Am content in the west tank would indicate that initial ^{241}Pu levels were only about 0.643 grams (or an initial weight ratio of ~0.00054:1 – about 1/10th the ratio expected). The maximum initial value for ^{241}Pu

in the west tank would have been 0.643g or 6.62E+01Ci). For the east tank, the initial value of 0.166g would apply. These calculated values for ^{241}Pu are used in the table.

To explain the lower than expected ^{241}Am content of the tanks, a "super" weapons-grade plutonium may have been involved. The earliest plutonium production runs were reported to have been of this type, resulting in very limited production of ^{240}Pu and ^{241}Pu versus ^{239}Pu, due to the short reactor cycles. The relationship between ^{240}Pu and ^{241}Pu is considered to maintain a relative constancy, so initial ^{240}Pu is assumed to have existed at approximately an order of magnitude greater concentration than ^{241}Pu. The west tank would have contained 6.4 grams and the east tank would have contained 1.7 grams. Initial ^{238}Pu would have been 5.55E-01 Ci in the west tank and 1.65E-01 Ci in the east tank by an iterative process similar to that discussed for ^{241}Pu. The ^{240}Pu abundance assumption is based on typical relationships between ^{240}Pu and ^{241}Pu content (Carson 1993).

6) Uranium in the tanks is enriched to 0.95% ^{235}U. This is reasonable based on Hanford operations history. Reactor depletion of ^{235}U is unknown and has been ignored as not contributing significantly to the final calculated radionuclide values. The 1981 uranium analytical data is partitioned to the major uranium isotopes based on Portsmouth Gaseous Diffusion Plant typical data (USEC 1999).

7) Conversion of 2007 radionuclide inventory to ^{239}Pu fissile gram equivalents will be performed using alternate techniques to permit a comparison between the resulting values. The RadCalc 4.0 WIPP calculation is per TRAMPAC (WTS 2003). A second method utilizes the Nevada Test Site Waste Acceptance Criteria (NTSWAC) Revision 6, Appendix B conversion factors. The preferred method utilizes the dose conversion factors from EPA Federal Guidance Report 11 (EPA-520/1-88-020, September 1988). The most restrictive clearance class is utilized with 1 micron particle size assumption.

8) No cesium or strontium carry-over into the tanks is expected, since tank contents represent materials resulting from multiple separation processes. Cs and Sr are effectively removed by initial separation processes in the plutonium process stream.

RadCalc 4.0 was used in combination with the assumptions to prepare a radionuclide inventory for the tanks applicable to 1981. Radionuclides not included in the 1981 samples were estimated based on assumptions described. Then the radionuclide inventory was aged to 2007 to represent the project access date.

2007 Estimated Radionuclide Inventory

Radionuclide	West Tank (g)	East Tank (g)
^{234}U	3.46E+00	1.06E+00
^{235}U	6.57E+02	2.02E+02
^{238}U	6.84E+04	2.10E+04
^{238}Pu	2.01E-02	6.01E-03
^{239}Pu	1.19E+03	3.12E+02
^{240}Pu	6.35E+00	1.69E+00
^{241}Pu	3.55E-02	9.22E-03
^{241}Am	5.63E-01	1.46E-01
^{239}Pu Equiv[1]	1279	335

[1] Based on EPA Dose Conversion

Criticality Potential

The tanks have been evaluated for criticality potential several times in the past several years. The consistent result is a determination that the form and distribution of the fissile materials in the tanks contribute to ensure that a criticality is non-credible for the tanks (McLaughlin 2003, 91859).

Dose Potential

Based on data available, the tank radionuclide contents are primarily alpha emitters, representing an inhalation and/or ingestion hazard rather than an external dose hazard.

Hydrogen Evolution

The General's Tanks contain enough nuclear material to consider the possibility of radiolytic processes in the tanks. Additional assumptions associated with radiolysis include:

1) The water content in the tanks is sufficient to allow maximum potential hydrogen evolution from radiolytic processes. Contaminants in the water, such as nitrates, are known to significantly reduce hydrogen evolution rates, but reductions are not considered in the calculations.

2) G-values, in this case the yield of radiolytic hydrogen from pure water for alpha, beta, and gamma radiation from the tanks contents, are 1.7, 0.45, and 0.45 molecules H2 produced per one hundred electron volts (eV) absorbed, respectively (Ausloos 1968)(Choppin 1989)

3) The tanks were sealed in 1985, based on records from LANL operations (DOE 1987, 08664).

4) As a result of the high altitude at the Los Alamos site (~6700 feet above sea level at MDA A), the mass of air in the tank headspace is reduced to 80% of STP values, from 8322 gram-moles to 6707 gram-moles, ultimately increasing the relative concentration of hydrogen to air mass.

Hydrogen content in the tanks was calculated using the dedicated subsystem within RadCalc 4.0 for radiolytic hydrogen evolution, using the G-values for pure water and the radiation associated with the decay of the calculated radionuclide inventory between 1985 and 2007, as adjusted for Los Alamos atmospheric pressure.

The effects of high levels of nitrates have not been incorporated into the calculations. Nitrates are known to retard the production of free hydrogen by scavenging hydrogen and releasing oxygen in the process. Radiolysis of nitrates and associated chemical reactions potentially create additional hazards in the tanks, particularly from the generation of nitrous oxides and ammonia, both of which would contribute to combustion issues, but as a trade-off to hydrogen production. Nitrate-based reactions are also known to produce nitrites and nitrides. Nitrate content would also lead to oxidation of any organic species present in the tanks.

The complexity of the reactions involved does not lend itself to summary in this white paper. The net impact to the combustion issues in the tanks is not believed to be significantly different from the condition presented, i.e., the maximum hydrogen production assumption is bounding for the tank combustion issues.

Estimated Maximum Potential Hydrogen Gas Content in General's Tanks

Result Category	West Tank	East Tank
Gram-Mole Hydrogen Content	296.4	77.6
Sea Level Atmospheric Percent	3.40 %	0.92 %

Adjusted to LANL MDA A Elevation	4.42 %	1.16 %

The lower flammability limit for hydrogen in air is ~4%.

The calculated maximum potential hydrogen concentration in the west tank exceeds the lower explosive limit or lower flammability limit for hydrogen in air. This result does not consider the relatively high rate of diffusion of hydrogen through steel, nor the much higher rate that hydrogen, because of its extremely small molecular size, leaks through otherwise gas-tight seals. There is a very high probability that the tanks were never sealed to the extent that hydrogen could be contained.

Further, hydrogen deflagration in a downward or lateral direction requires additional hydrogen concentration to fully propagate (9% and 6% respectively)(Paillere 2005). The significant diameter and length of the General's Tanks would indicate that a greater concentration of hydrogen would be necessary to sustain a deflagration.

Chemical Constituents

Inorganic Materials

Chemical constituents in the General's Tanks liquids were included in data reported in 1981 by J. Bucholz (Bucholz 1981, 00459). The relationship between the liquids in the tanks and the solids is likely affected by the tank pH. The initial precipitation of the radionuclides would be typical of the higher pH values reported in the tanks in 1981. The potential for additional infiltration of liquids from the surface, such as rainwater as reported in the MDA Summary drafted around the time of the last reported General's Tanks field work (Ahlquist 1986, 00528).

1981 General's Tanks Analytical Sample Chemical Data

Analysis	West Tank (KOH) (mg/L)	East Tank (NH_4OH) (mg/L)
NH_4-nitrogen	24.6	18,000
Na^+	3,800	9,300
Cl^-	3,609	4,998
K^+	36,000	7,300
$SO_4^=$	2,533	1,369
I^-	611	560
Total Dissolved Solids	87,780	233,480
Ca	2.26	9,500
NO_3-nitrogen	6,590	34,500
Al	16	1.1
pH	11.4	8.3

Organic Materials

Although the presence of organic materials in the General's Tanks is possible, based on original chemical processes used to purify plutonium, the expected volume of organic materials in the tanks is expected to be only at trace levels. An interview recorded with one of the site workers

who participated in the liquids retrieval project in the 1970s and 1980s confirmed that no organic materials were encountered or visible in the tanks (Ritchey 2006, 92455).

Potential Impacts from Combustion

Modeling Hydrogen Effects with TNT

In order to determine the potential impacts of a hydrogen combustion/deflagration in the General's Tanks, an equivalent quantity of trinitrotoluene (TNT) was determined and the resulting effects of TNT detonation were evaluated. Hydrogen deflagration-to-detonation (DTD) transition does not match TNT's detonation characteristics for a number of reasons, mainly the slow 2.6 meters per second rate of hydrogen burn-front propagation (Mishima 2002, LA-UR-02-3803) and the lower resulting force created. Secondly, the hydrogen levels in the General's Tanks, although above the lower explosive limit, would still reach the concentrations necessary to detonate or to deflagrate in a downward or lateral direction.

Notwithstanding these differences, hydrogen to TNT equivalence has been estimated because TNT detonations are very thoroughly studied and understood and impacts predicted from the TNT detonation can be considered bounding in this case. The resulting information provides input to engineering design, health and safety, and management level determination. The TNT-equivalence approach will result in greater estimated forces and require greater caution at all distances than the modeled hydrogen event would.

The purpose of this evaluation is to determine 1) the TNT equivalent weight comparable to hydrogen created from radiolytic decomposition of water, 2) the approximate overpressures created in the event of a detonation, 3) the depth-of-burial necessary to completely contain the explosive force, and 4) the effects on the second tank or the natural gas line from a one-tank detonation scenario.

Based on radionuclide inventory in the east and west General's tanks, an equivalency of TNT was derived from the volume of hydrogen evolved since the tanks were sealed in 1985. The equivalency factor (EPRI 1987) states that 1000 standard cubic feet (scf) of hydrogen is equivalent to 27.1 lbs TNT at standard temperature and pressure (stp). The west and east tank's TNT equivalence, therefore, is 6.355 lbs and 1.663 lbs, respectively.

TNT Blast Effects

For TNT detonations on the surface of the ground, the overpressure created/resulting blast forces are thoroughly studied and have been summarized in the table below for the respective equivalent TNT masses calculated for each of the General's Tanks. As points of comparison, eardrums rupture at 2.5 psi, 6 psi causes 99% fatalities to exposed standing persons (from the fall), and 25 psi causes all exposed structures to collapse.

TNT Equivalent Surface Blast Overpressures as a Function of Distance

Distance (feet)	West Tank Blast Overpressure (psi)	East Tank Blast Overpressure (psi)
1	2125	1100
5	100	35
10	21	9
100	0.5	0.29
300	0.15	0.08

However, since the General's Tanks are buried below the ground surface, these forces are better expressed as a depth-of-burial requirement. The depth-of-burial required to completely contain the forces of a detonation is estimated at 11.29 feet for the west tank and 7.11 feet for the east tank. Credit can be taken for the weight of the existing soils over the tanks (taking density and weight into consideration), the structural integrity of the surrounding volcanic tuff bedrock materials, and the weight and structural integrity of the reinforced concrete slab overlying the General's Tanks. The soils overlying the west tank would not provide the 11.29 feet necessary for total containment, but the effect of the reinforced 8-inch thick concrete slab would be very much greater than the corresponding volume of loose soil, as discussed in the following section.

If one tank were to detonate with maximum force (the west tank), the effect on the other tank would be negligible, as it is 20 feet apart, separated by 20 feet of compacted fill. The natural gas line is approximately 34 feet distant, requiring the force to travel through backfill material, undisturbed earth, and is slightly protected due to a lower elevation.

Protection of the gas line and civilian population from conventional blast effects is unnecessary, based on these calculations. Workers in the immediate vicinity of the tanks would require protection during initial tank access activities based on the TNT equivalence analyses alone, but the lower than necessary hydrogen concentration for tank deflagration and the very high potential for hydrogen leakage from the tanks since the 1985 seal date combine to make the deflagration a non-credible event for both tanks.

Evaluation of the Concrete Slab

The ability of the existing concrete slab to withstand the forces imposed on it by an accidental detonation of hydrogen in one of the General's Tanks was evaluated based on mechanical properties of the concrete slab and using the modeled TNT equivalent detonation. For purposes of calculation, the assumption was made that the tank will rupture at a weak point underneath the slab rather than at the former center access point where there has been a previous penetration of both the tanks and the concrete slab.

The slab is situated approximately 1.5 feet above the tank top. The expected force from a detonation at this distance is approximately 2,000 psi. The slab is 8 inches thick and heavily reinforced, but due to uncertain construction techniques and potential degradation since initially placed, the slab was assumed to be 6 inches thick with no reinforcement. Two different sizes of tank ruptures were considered, 3 inches and 24 inches.

The 3-inch rupture would result in shearing force of 14,200 pounds with a concrete resisting force of 19,856 pounds (capacity to demand ratio of 1.4). The 24-inch rupture would result in a shearing force of 75,400 pounds with a concrete resisting force of 136,847 pounds (capacity to demand ratio of 1.80). The shear strength of the concrete was calculated as "two-way" shear in accordance with ACI-318-99 (ACI 1993).

Based on the resulting capacity to demand ratios being greater than 1.0, it was determined that the slab would contain the detonation as long as it did not occur under the existing mid-point opening.

Reinforcement of the central openings in the slab would correspondingly provide assurance that a tank rupture at any of the points under the slab would be contained by the slab integrity.

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