

Deep Sludge Gas Release Event Analytical Evaluation – 14225

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ABSTRACT

The purpose of the Deep Sludge Gas Release Event Analytical Evaluation is to evaluate the postulated hypothesis that a hydrogen gas release event may occur in Hanford tanks containing waste sludge at levels greater than previously experienced. There is a need to understand gas retention and release hazards in sludge beds which are 5.05 – 7.62 m (200 -300 in.) deep. These sludge beds are deeper than historical Hanford sludge waste beds, and are created when waste is retrieved from older single-shell tanks and transferred to newer double-shell tanks. Retrieval of waste from single-shell tanks reduces the risk to the environment from leakage or potential leakage of waste into the ground from these tanks.

However, the possibility of an energetic event (flammable gas accident) in the retrieval receiver double-shelled tank is worse than slow leakage. Lines of inquiry, therefore, are (1) can sludge waste be stored safely in deep beds; (2) can gas release events be prevented by periodically degassing the sludge (e.g., mixer pump); or (3) does the retrieval strategy need to be altered to limit sludge bed height by retrieving into additional double-shelled tanks? The scope of this effort is to provide expert advice on whether or not to move forward with the generation of deep beds of sludge through retrieval of C-Farm tanks. Evaluation of possible mitigation methods (e.g., using mixer pumps to release gas, retrieving into an additional double-shelled tank) are being evaluated by a second team and are not discussed in this report.

While available data and engineering judgment indicated that increased gas retention (retained gas fraction) in double-shelled tank sludge at depths resulting from the completion of single-shelled tank 241-C, Tank Farm retrievals is not expected and, even if gas releases were to occur, they would be small and local; a positive Unreviewed Safety Question was declared [1]. The purpose of this technical report is to (1) present and discuss current understandings of gas retention and release mechanisms for deep sludge in the U.S. Department of Energy complex waste storage tanks; and (2) to identify viable methods/criteria for demonstrating safety relative to deep sludge gas release events in the near term to support the Hanford C-Farm retrieval mission. A secondary purpose is to identify viable methods/criteria for demonstrating safety relative to deep sludge gas release events in the longer term to support the mission of retrieving waste from the Hanford Tank Farms and delivering it to the Waste Treatment and Immobilization Plant.

The potential deep sludge gas release events issue resulted in the declaration of a positive Unreviewed Safety Question [1]. C-Farm retrievals are currently proceeding under a Justification for Continued Operation [2] that only allows tanks 241-AN-101 and 241-AN-106 sludge levels of 4.88 m and 4.95 m (192 in. and 195 in.), respectively. C-

Farm retrievals need deeper sludge levels (approximately 7.87 m (310 in.) in 241-AN-101 and approximately 6.35 m (250 in.) in 241-AN-106) [3]. This effort is to provide analytical data and justification to continue retrievals in a safe and efficient manner.

INTRODUCTION

There is a need to understand gas retention and release hazards in sludge beds which are 5.08 m to 7.62 m (200 in. to 300 in.) deep. These deep sludge beds are built when waste is retrieved from older single-shell tanks (SST) and transferred to newer double-shell tanks (DST). Retrieval of waste from SSTs reduces the risk to the environment from leakage or potential leakage of waste into the ground. However, although a slow leakage is a potential concern, the possibility of an energetic event (flammable gas accident) in a waste tank has the potential for major consequences. Lines of inquiry for this report are (1) can sludge waste be stored safely in deep beds; (2) can gas release events (GRE) be prevented by periodically degassing the sludge (e.g., mixer pump); or (3) does the retrieval strategy need to be altered to limit sludge bed height by retrieving into additional DSTs?

BACKGROUND

The potential deed sludge gas release event (DSGRE) issue resulted in the declaration of a positive unreviewed safety question (USQ) [1]. C-Farm retrieval is currently proceeding under a Justification for Continued Operation (JCO) [2] that only allows tanks 241-AN-101 and 241-AN-106 sludge levels of 4.88 and 4.95 m (192 in. and 195 in.) respectively. C-Farm retrievals need deeper sludge levels [3] (approximately 7.87 m (310 in.) in 241-AN-101 and approximately 6.35 m (250 in.) in 241-AN-106) to meet retrieval milestones and consent decree requirements. The DSGRE USQ [1] is based on a new mechanism for a spontaneous GRE in deep sludge that has been postulated but is not described in the Tank Farms Documented Safety Analysis (DSA) [4]. The DSA controls that prevent potential GRE flammable gas hazards from large spontaneous buoyant displacement gas release events (BDGRE) may not preclude a spontaneous GRE from this new mechanism.

The DSA currently concludes that based on the operational history of the DSTs, a spontaneous GRE of sufficient size to cause the tank headspace to reach 100% of the lower-flammability limit (LFL) is not expected. That is, the frequency of a flammable gas accident due to a large spontaneous GRE (i.e., BDGRE) is “beyond extremely unlikely.”

While available data and engineering judgment indicate that increased gas retention in DST sludge at depths at the completion of SST 241-C Tank Farm retrievals is not expected and, even if gas releases were to occur, they would be small and local, a potential inadequacy in the safety analysis (PISA) was declared [1]. A USQ evaluation of the PISA [1] determined that the postulated mechanism for a spontaneous GRE in deep sludge is a positive USQ. The positive USQ is based on the situation where, at this time, there is insufficient information to conclude that the frequency of a flammable gas accident due to a large spontaneous GRE (a DSGRE) in sludge deeper than

historical levels is “beyond extremely unlikely” (i.e., the frequency could be higher than “beyond extremely unlikely”).

APPROACH TO CLOSING THE DSGRE USQ

A positive USQ is closed by updating the DSA to describe the risk (frequency and consequences) of a flammable gas accident resulting from a DSGRE, without controls, and, if credible, specifying any required controls to prevent or mitigate such an accident.

DSGRE Accident Scenario

The postulated flammable gas accident scenario due to a DSGRE involves all of the following.

- Waste retrieval operations result in waste layer density differences (density inversion) in the presence of retained gas.
- Waste density inversions result in instability that causes waste movement and waste movement causes a large spontaneous GRE into the DST headspace.
- The lower flammability limit (LFL) is exceeded in the DST headspace and ignition occurs, resulting in DST failure and release of radioactive and hazardous materials.

Gas Retention and Waste Layer Density Inversions

Information relative to gas accumulation that leads to waste density inversions (a less dense waste layer below a more dense waste layer) was presented and discussed [3]. Salt slurry waste is precipitated soluble sodium salts in thick brine. Salt slurry wastes are relatively weak (100 to 300 Pa). Sludge waste is precipitated aluminum hydroxy salts with a more dilute interstitial liquid. Settled sludge wastes are relatively strong (1,000 to 3,000 Pa). Observed gas retention and release behaviors in salt slurry waste are described by the BDGRE theory which is described as follows.

- A localized region of settled salt slurry waste (a gob) accumulates sufficient gas to become buoyant and break away from settled solids layer
- The buoyant waste gob rises through the waste supernatant layer and the retained gas expands as hydrostatic pressure decreases (buoyant displacement)
- Gas expansion causes the waste gob to break apart releasing retained gas into the tank headspace (gas release event)

Waste transfers are contractually controlled to prevent new BDGRE tanks by either the buoyancy ratio or the energy ratio [3].

Gas Retention in Sludge Waste

Current BDGRE criteria were developed based on observations of salt slurry waste and applied to both salt slurry and sludge. However, sludge waste appears to retain gas differently than salt slurry waste. Examination of material balance discrepancies show

retained gas is released slowly from salt slurry waste but quickly from sludge waste [3]. Sludge waste behavior was not as thoroughly investigated in the 1990s as salt slurry waste was because only the salt slurry waste had issues (e.g., BDGREs, slurry growth, floating crusts). Therefore, data regarding gas retention and waste density profiles for Hanford settled sludge waste is sparse.

A group of Dutch researchers [5] investigated gas retention and release in their contaminated lake bottom sediment sludge. The Dutch observed that channels form in settled sludge as pore water from sludge consolidation and generated gases flow from the sludge to the sludge surface. Experiments in clay waste simulants also showed channel formation and gas release through these channels in relatively strong sludge (480 Pa shear strength). The Dutch theorized, however, that below a certain sludge depth ($d_{\max}$), channels collapse due to lithostatic loading and gas release is restricted; therefore, gas retention increases. However, there is no known experimental or operational experience that has identified $d_{\max}$ (including a 5-m (16.404 ft.) deep sediment experiment conducted by the Dutch group). Preliminary experiments on open channel depths, gas retention, and channel formation, indicate that channel formation and gas release in simulants were many times deeper than the observed open channel depth. Therefore, it is expected that the Dutch $d_{\max}$ equation is not a sound model to apply to Hanford tank waste sludge. In particular, there is no validating experimental data applicable to tank waste sludge.

Although there may be issues with the $d_{\max}$ model application to tank waste sludge, and the $d_{\max}$ theory may eventually be discounted, there are other mechanisms for creating density inversions in retrieval receiver tanks (241-AN-101 and 241-AN-106). The degassed density of sludge in C-Farm tanks varies from 1.28 to 1.78 g/ml [3]. There are also variations in the gas generation rates for the different sludge [3]. Differences in degassed density and differences in gas generation and retention between waste layers may create density inversions. In addition, the retrieval process may add more dense, degassed sludge, on top of less dense sludge that has already accumulated some gas, thus creating density inversions. Waste density inversions pose the possibility for waste instabilities and thus gas release events.

Waste Instability and Gas Release Events

Several mechanisms for a large spontaneous GRE are presented and discussed below.

BDGREs: The instability is due to a gob of waste accumulating sufficient gas to become buoyant in the tank supernatant liquid and break away from the settled solids layer. The criterion used to predict instability and waste movement is the buoyancy ratio. The gas release then occurs as a result of gas expansion as the gob rises through the supernatant that causes the waste gob to break apart releasing a fraction of its retained gas into the tank headspace. The gas release is predicted/controlled through the energy ratio. The technical team has concluded that the BDGRE model is not the best description of instability and gas release in settled sludge, but would likely bound DSGRE behavior. A model for predicting BDGRE size based on the buoyancy ratio has

been documented [6]. Applying this methodology to 241-AN-101 and 241-AN-106 would support deeper sludge in 241-AN-101 (but short of the 7.87 m (310 in.) required to complete C-Farm retrievals) and deeper sludge in 241-AN-106 (which may support completion of C-Farm retrievals into this tank). This may allow a near-term revision to the deep sludge JCO [2] to support continued retrievals until a technical basis for 7.87 m (310 in.) in 241-AN-101 can be developed.

Rayleigh-Taylor Instability: Waste with density inversions may experience Rayleigh-Taylor (R-T) instabilities. Two scoping experiments with kaolin clay simulants [6] (two clay “sludge” layers, the bottom layer producing gas and the upper layer without gas) developed density inversions and exhibited waste instabilities and waste movement. The buoyant motion of the gas containing slurry layer depended on whether it was more or less dense than the overlying supernatant at the time the settled bed became unstable. The slurry was not buoyant in the first scoping test, but it was in the second. However, there was little gas release associated with the instability events in both experiments. This compares with an older experiment in which gas was generated throughout a single layer of clay simulant which demonstrated a BDGRE and significant gas release.

Limited waste density profile data for salt slurry and salt cake waste tanks were used along with initial proposed R-T stability criteria and compared to observed GRE behavior in these tanks [3]. Tank farm waste density profile data for salt slurry and salt cake wastes demonstrate that there is a potential for motion within waste sediment layers due to bulk density differences based on the proposed R-T instability criterion. However, there was no clear correlation between the potential for R-T instabilities and the observed gas release behavior.

The plastic instability regime is probably more important for tank waste than the elastic regime. The group discussion concluded that R-T instability is a reasonable model for settled sludge wastes with density inversions, and that such instabilities could not be ruled out and should be assumed to occur (i.e., DSGRE prevention based on controlling waste through stability criterion is not a recommended approach).

Gas release from strong (1000 Pa shear strength) sludge was discussed. With high shear strength sludge-like material, the effective viscosity, once yielded, is likely to be high and bubbles (e.g., in gas-filled slits/channels) are expected to be locked into the material (i.e., bubble rise velocity is very slow relative to any bulk motion of the sludge). In other words, gross waste movement may occur but gas release is slow (i.e., the bubbles don’t move through the upwelled material very fast). Settling and consolidation of Hanford sludge is fast compared to gas generation/retention rates, therefore, resulting in a strong sludge before significant gas retention occurs. The material may crack to form channels (gas release through cracks and channels). However, strong materials will exhibit only small gas releases (i.e., large gas releases only occur in weak materials). Some Hanford wastes exhibit shear thinning (and/or rheogram overshoot, which can lead to overestimation of the rheological properties, strength, and effective viscosity) and this needs to be considered when evaluating gas release behavior from

strong sludge upon yielding. The initial conclusion is that gas bubble movement and gas release from a strong, viscous sludge would be difficult resulting in small, slow, episodic gas release, if any, as a result of waste movement from a R-T instability in the sludge layers. There is less confidence that a large and rapid gas release would not occur if the gas retaining layer then rose through a deep supernatant (see BDGRE scenario above). Minimizing the depth of the supernatant is a potential control approach for deep sludge GREs. It should be noted that controlling supernatant depth is a current control associated with preventing the creation of a new BDGRE tank via the energy ratio.

It is also recognized that operational experience with storage of tank sludge at Hanford has shown no large spontaneous GREs. There is no evidence to support the occurrence of large releases via this DSGRE mechanism in Hanford sludge waste to date.

Bubble Cascade. The postulated mechanism is a bubble cascade where, at high bubble concentrations, the disturbance caused by a rising bubble engages other bubbles leading to a spontaneous cascade release in a relatively weak material. There was such a gas release event that occurred in Savannah River Tank 40H in November 2002 [7]. The event occurred in freshly settled slurry, apparently triggered by a small disturbance (an aborted mixer pump run). The release was estimated at 45 m^3 (1589.16 ft^3) of hydrogen, the bulk of which occurred over 5 hours. Savannah River controls gas releases by periodically degassing the sludge with mixer pumps.

It is postulated that bubble cascade events would only occur in relatively weak waste ($\sim 10 - 70 \text{ Pa}$). A scenario was previously postulated for Hanford tanks 241-AN-101 and/or 241-AN-106, where freshly retrieved waste might retain sufficient gas while it is relatively weak (during the settling transient) and experience a bubble cascade event. It was noted that the gas generation rate in the Tank 40H waste was much higher than Hanford retrieved sludge, although gas generated and released from underlying sludge might be retained in the weak, partially-settled sludge layer. The settling rate for the Tank 4-H washed sludge is also likely much slower. It is expected that Hanford sludge could not experience this phenomena due to the relatively high settling rate compared to the gas generation and retention rates. That is, the sludge would settle into relatively strong sludge before sufficient gas could accumulate to cause a bubble cascade event.

Large Trapped Bubble. This postulated mechanism involves gas being trapped below a relatively gas impervious waste layer and then being released if the impervious layer is breached (e.g., cracks and fissures). The size of a "large" bubble necessary to yield even strong sludge (e.g., 1000 to 3000 Pa shear strength) is sufficiently small that any episodic releases would also be small. It is expected that the large trapped bubble mechanism is not applicable to sludge in 241-AN-101 and 241-AN-106. It should be noted that operational experience with storage of tank sludge at Hanford has shown no large spontaneous GREs (i.e., there is no evidence to support the occurrence of large releases via this DSGRE mechanism in Hanford sludge waste to date).

Flammable Gas Deflagration and Consequences

Flammable gas accident phenomena and consequences are described in the Hanford Tank Farms DSA [4]. The consequence of a flammable gas deflagration in the DSA apply to flammable tank headspace conditions, regardless of the mechanism for creating the flammable conditions (i.e., they only relate to the flammable gas concentration, not its origin).

The DSA concludes that a deflagration in the headspace of a DST, without controls, results in an onsite radiological consequence that is < 100 rem, the offsite toxicological consequence is $< \text{Protective Action Criteria-2 (PAC-2)}$, and the onsite toxicological consequence is $< \text{PAC-3}$. However, it is qualitatively determined that without controls a DST headspace deflagration could result in significant facility worker consequences (i.e., grievous injury or death to a facility worker due to overpressure or physical impact from SSC failure [missiles], or from toxicological exposure exceeding PAC-3). Accordingly, safety-significant structures, systems and components (SSCs) and/or technical safety requirements (TSRs) are required to protect the facility worker. (Note: The frequency of a flammable gas accident due to large spontaneous BDGRE is "BEU" and no safety significant SSCs and/or TSRs are required for BDGREs. However, note the DSGRE USQ described above and the required JCO is for DSGREs.)

Approach to Demonstrate Safety Relative to DSGRE Accidents

An initial basic assumption is that DSGREs are bounded by the BDGRE size model. To validate, the path forward is to revise, as necessary, and apply the BDGRE size model [3] to 241-AN-101 and 241-AN-106. Applying this methodology to 241-AN-101 and 241-AN-106 would support deeper sludge in 241-AN-101 (short of the 310 inches required to complete C-Farm retrievals) and 241-AN-106 (which may support completion of C-Farm retrievals into this tank). This may allow a near-term revision to the deep sludge JCO to support continued retrievals until a technical basis for 7.87 m (310 in.) in 241-AN-101 can be developed.

The first postulation is that gas retention and channel formation for gas release is not affected by deep sludge. This is evaluated by the use of the d_{max} theory for waste sludge. There is no known experimental or experience based data to validate that channel formation and gas release in deep sludge is restricted at depth. Data that can be applied to tank waste is needed to determine if the d_{max} theory is valid and applicable to deep waste sludge. Data can be obtained through bubble retention experiments in tall columns to quantify bubble retention above and below estimated values of d_{max} . Experiments will measure gas retention in tall column experiments using waste sludge simulant and quantify if gas retention increases below estimated values of the open channel depth, d_{max} . This task will also measure and interpret open channel depths in representative waste simulants.

Additionally, it is assumed that gas release from strong sludge is small and slow. This is evaluated by observing the gas release behavior for strong sludge during R-T

instabilities. Available theory will be described in a white paper that provides analytical evaluations of bubble motion in strong waste during a R-T instability and the resultant gas release behavior. The white paper will attempt to predict whether or not a high-strength sludge gob generated by the R-T instability will yield as it rises through the sludge or through the overlying supernatant liquid. If it yields, the analysis will include a derivation of an expression for the rate at which yielded high-strength sludge is entrained by and gives up its gas content to surrounding sludge or surrounding supernatant. Based on the assumption that yielded-high-strength sludge possesses a high effective viscosity, a bubble-disengagement argument will be included that evaluates if a large GRE could result in flammable tank headspace. A question to be addressed is if waste is a shear thinning material that may not have a high apparent viscosity once it yields in a R-T instability.

Theory needs to be supported (validated) with data from large scale R-T instability induced gas release experiments. Experiments will collect data to quantify how gas release due to bulk motion caused by an idealized R-T instability changes with increasing test vessel size (e.g. 0.305 m to 1.22 m (1 ft. to 4 ft.) and, 10 Pa to 150 Pa shear strength, geometrically scaled). This data will be used with analytical and numerical models (see above) to estimate gas release in full-scale tanks. Additional R-T experiments will quantify the role of relative layer thickness, gas releases from an upper layer, and multiple layer systems as needed. Waste simulant selection needs to consider potential waste shear thinning behavior.

Additional data is needed, such as waste density profiles in 241-AN-101 and 241-AN-106. Without actual measurements, waste density profiles in 241-AN-101 and 241-AN-106 can only be estimated based on C-Farm sludge densities and retrieval histories with assumptions on sludge settling behavior and gas retention void fractions. Knowing waste density profiles is important to modeling R-T instabilities. Degassed waste density can be obtained from core samples. Bulk density (includes gas voids) would require retained gas core samples. However, core sampling equipment is not currently available.

Another data need is the waste shear strength as a function of sludge depth. This data is needed to relate experiments to tank waste conditions. In-situ shear strength measurements require the use of the cone penetrometer which would take 6 to 12 months to deploy.

Finally, it is assumed that bubble cascade GREs are not credible for 241-AN-101 and/or 241-AN-106. To support this assumption it is necessary to obtain gas generation rate and waste shear strength estimates for the Savannah River Tank 40H bubble cascade event. Then perform an evaluation (contrast) of 241-AN-101 and 241-AN-106 waste properties and operating conditions relative to Savannah River Tank 40H waste and previous PNNL experimental waste conditions that lead to bubble slurry cascade releases.

CONCLUSIONS

The technical team for this activity has concluded that a large GRE cannot be ruled out to occur in tanks 241-AN-101 or 241-AN-106 at planned levels of retrieved sludge of approximately 7.87 m and 6.35 m (310 in. and 250 in.) based on the current level of understanding. However, theoretical model development, validated by experiments and tank waste data, is likely to demonstrate that an accident due to a DSGRE is not credible (may include a control on supernatant depths). However, there may be a possibility of a revised JCO, based on preliminary information, and the application of the BDGRE size model (argued bounding of DSGREs) to allow an interim, incremental addition of sludge via a revised JCO.

RECOMMENDED ACTIONS

The following actions have been identified that must be evaluated in order to assure that a large DSGRE is not viable.

- Apply a revised PNNL-15238 BDGRE size model to 241-AN-101 and 241-AN-106 to support a JCO revision.
- Perform tall column experiments to quantify bubble retention above and below estimated values of $d_{\max}$.
- Generate a technical document that describes gas release behavior for strong sludge during R-T instabilities.
- Perform large scale R-T instability induced gas release experiments.
- Obtain in-situ shear strength measurements as a function of depth to relate experiments to tank waste conditions
- Evaluate (contrast) 241-AN-101 and 241-AN-106 waste properties and operating conditions relative to Savannah River Tank 40H waste and previous PNNL experimental conditions that lead to bubble cascade releases.
- Update the documented safety analysis (DSA) to describe the risk (frequency and consequences) of a flammable gas accident resulting from a DSGRE, without controls, and, if credible, specifying any required controls to prevent or mitigate such an accident.

Once, these actions are completed, it is expected that the results will support continued retrieval of sludge into the Hanford DSTs to depths up to 7.87 m (310 in.).

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