Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

Disclaimer:

Simulating Gas Release from an Agitated Vessel with MSTAR Lattice-Boltzmann Computational Fluid Dynamics -21161

Michael R. Poirier*, John Thomas, and John M. Pareizs***

***Savannah River National Laboratory**
Aiken, SC 29808

****M-Star Simulations, LLC**
Baltimore, MD

ABSTRACT

The Savannah River Site (SRS) currently manages the risk for retained hydrogen in the Defense Waste Processing Facility (DWPF) vessels by implementing a Retained Hydrogen Program. The current program relies on conservative assumptions concerning gas release and retention to develop allowable vessel Q-times. SRS would like to establish less conservative inputs to the Retained Hydrogen Program. To address this need, the authors conducted a literature search on work investigating the retention and release of gas from DWPF type slurries. The search revealed that it is likely that the retained hydrogen is not released instantaneously upon the start of agitation, and that hydrogen gas could be released under quiescent, non-agitated, conditions. The literature search also identified a Lattice-Boltzmann computational fluid dynamics (CFD) model that could potentially be used to model and predict hydrogen release from DWPF vessels. After developing and benchmarking the model, it was applied to DWPF process vessels to predict release rates of retained hydrogen. This paper describes the development and application of a model to show its ability to describe retained hydrogen release from DWPF vessels and relax the conservativisms in the Retained Hydrogen Program.

The work for this report used the M-Star® Lattice-Boltzmann CFD model to simulate retained hydrogen gas release from vessels during sludge batch qualification testing and DWPF process vessels. The model uses a cylindrical tank with rotating impellers to simulate mixing of a non-Newtonian slurry, and the release of gas bubbles. A volume of gas bubbles is added to the tank, the impeller is started, and the fluid motion and bubble movement in the vessel are calculated as a function of time. When the hydrogen bubbles reach the fluid-gas interface, they are released. This feature of the model may lead to faster hydrogen gas release, which would be conservative for the DWPF vessels. The release rate of hydrogen gas bubbles is tracked, and fit to an empirical model. This empirical model is used as the input to a second simulation, the fluid motion and hydrogen concentration in the vessel head space.

The conclusions from this work follow.

- The model was used to simulate the release of retained hydrogen during Sludge Batch 3 qualification, and was found to agree with the test data.
- Model simulations of the DWPF Sludge Receipt and Adjustment Tank (SRAT) predicted 50% of the retained hydrogen would be removed in 21 – 29 minutes depending on operating conditions.
- Model simulations of the DWPF SRAT showed that the maximum hydrogen concentration in the head space would be more than an order of magnitude below the theoretical maximum corresponding to all retained hydrogen being released instantaneously.
- Model simulations of the DWPF Sludge Pump Tank (SPT) predicted 50% of the retained hydrogen would be removed in ~15 minutes for the conditions simulated.
- Model simulations of the DWPF SPT showed that the maximum hydrogen concentration in the head space would be ~60% of the theoretical maximum corresponding to all retained hydrogen being released instantaneously.
- Investigation of bubble size on the release of hydrogen from slurries shows no significant impact for the range of bubble sizes expected at the DWPF. This result confirmed expectations, because momentum forces generated by the impeller are much larger than the buoyancy forces acting on the bubbles.

- Investigation of the impact of yield stress on the release of hydrogen from the SRAT shows a small effect when it is between 1.5 and 21 Pa. Increasing the yield stress from 1.5 to 21 Pa (a 7X increase), increased the time required to remove 50% of the hydrogen from the SRAT slurry by 27%.
- Investigation of the effect of bubble concentration on the release of hydrogen from slurries shows no significant impact when the concentration is less than 0.1 volume%, and a noticeable effect at a concentration of 1 volume%.
- Investigation of the effect of impeller speed on the release of hydrogen from slurries shows hydrogen bubble release to increase with increasing impeller speed, as expected.

INTRODUCTION

The Savannah River Site (SRS) currently manages the risk for retained hydrogen in the Defense Waste Processing Facility (DWPF) vessels by implementing a Retained Hydrogen Program. The program primarily relies on the timely operation of vessel agitators to ensure generated hydrogen is controlled such that flammable vapor spaces do not exceed safe limits. The program defines a Quiescent time (Q-time) for the Sludge Receipt and Adjustment Tank (SRAT), Slurry Mix Evaporator (SME), Melter Feed Tank (MFT), and Sludge Pump Tank (SPT) to prevent a flammable vapor from forming within the vessels (> 95% composite lower flammability limit [CLFL]). The Q-time is the maximum time that the vessel can remain unagitated without generating and accumulating sufficient hydrogen gas to challenge flammability safety limits. The Retained Hydrogen Program utilizes conservative assumptions regarding the retained hydrogen. These assumptions are all hydrogen generated while the vessel is quiescent is retained, and upon the start of agitation, the retained hydrogen is released instantaneously. SRS would like to establish less conservative inputs to the Retained Hydrogen Program.

The authors identified a Lattice-Boltzmann computational fluid dynamics (CFD) model that could potentially be used to model and predict hydrogen release from DWPF vessels. The authors used data from the qualification of previous DWPF sludge batches to develop and benchmark the model before applying it to DWPF vessels to predict retained hydrogen release rates.

APPROACH

The modeling employs a commercial Lattice-Boltzmann CFD package called M-Star® CFD developed by M-Star Simulations, LLC. This package was selected because it was recommended by experts in the field of fluid mixing and is used by mixing equipment suppliers. Once the software is described, this section will describe how the model was developed and benchmarked to accurately predict H_2 release to address the need to relax the current conservative assumptions in place at the DWPF.

M-Star Model-Fluid Mechanics

M-Star® CFD is a multi-physics modeling package used to simulate fluid flow, heat transfer, species transport, chemical reactions, particle transport, and rigid-body dynamics. M-Star® CFD contains three primary components: M-Star® Build, M-Star® Solve and M-Star® Post. M-Star® Build is a graphical interface used to prepare models and specify simulation parameters. M-Star® Solve uses input files generated from the interface to execute the simulation. M-Star® Post renders and plots the data generated by the solver.

Within M-Star CFD, the incompressible Navier-Stokes equations are solved using the lattice-Boltzmann method (LBM). The Navier-Stokes equations, which model the conservation of momentum of a fluid particle, can be written as:

$$\frac{\partial \vec{V}}{\partial t} + \vec{V} \cdot \nabla \vec{V} = g - \frac{\nabla p}{\rho} + \nabla \cdot \left(\nu \, \nabla \vec{V}\right), \tag{Eq. 1}$$

where $\vec{V}$ is the velocity of the fluid particle at a given location at a given time t, g is the acceleration due to gravity, p is pressure, ρ is the fluid density, and ν is the fluid kinematic viscosity at a given location and time. Note that this expression is inherently transient: the left-hand side represents the acceleration of the fluid particle, while the right-hand side represents the forces due to gravity, pressure gradients, and shear stress.

The Boltzmann transport equation, which models the conservation of transport carrier probability density, can be written as:

$$\frac{\partial f}{\partial t} + \vec{v} \cdot \nabla f = \Omega(f, f), \quad \text{(Eq. 2)}$$

where f represents the probability density function, $\vec{v}$ represents the phase space velocity, and Ω represents a collision operator. The Navier-Stokes equations are solved by linking f to the fluid momentum and $\Omega(f, f)$ to the fluid kinematic viscosity.[1] When solving Eq. 2, we discretized the velocity space using a D3Q19 lattice and relaxed the distribution using the Bhatnagar–Gross–Krook (BGK) operator. Fluids interact with the tank walls via a no-slip boundary condition, as enforced via a zero-velocity bounce back method.[2] Fluids interact with the moving impeller via the immersed boundary method, which enforces a no-slip velocity along the impeller/shaft surfaces as they move through the fluid.

A large-eddy simulation (LES) turbulence closure model was incorporated into the solution of Eq. 1. Within the context of LES, the unresolved (sub-grid) motions at each lattice site j are modeled by computing the local eddy viscosity, $\nu_{r,j}$:

$$\nu_{r,j} = (C_s \Delta x)^2 S_j \quad \text{(Eq. 3)}$$

where C_s is the Smagorinsky coefficient and S_j is the magnitude of the local strain rate tensor. When solving Eq. 1, this local eddy viscosity is calculated and added to the molecular viscosity at each time step. The Smagorinsky coefficient is set to 0.1, a value calculated from direct numerical simulation.[3] The computational models were built and solved using M-Star® CFD, a GPU-based computational physics package tailored for chemical engineering simulations. [4]

M-Star Model-Bubble Dynamics

Bubbles are modeled individually as discrete spheres assumed to be small relative to the fluid cells. Each bubble enters with a user-defined velocity, gas density, and bubble diameters sampled at runtime from a user-set diameter probability density function. Bubble trajectories are governed by Newton's second law, such that:[5]

$$m_i \vec{a}_i = \vec{F}_{i,g} + \vec{F}_{i,a} + \vec{F}_{i,p} + \vec{F}_{i,D} \quad \text{(Eq. 4)}$$

where m_i and $\vec{a}_i$ represent the mass and acceleration vector of bubble i, $\vec{F}_{i,g}$ is the gravity force, $\vec{F}_{i,a}$ is the added mass force, $\vec{F}_{i,p}$, is the instantaneous pressure gradient force, and $\vec{F}_{i,D}$ is the instantaneous drag force on the bubble. The trajectories of the bubbles are advanced in tandem with the fluid algorithm using the velocity Verlet algorithm.[6]

In accordance with Newton's third law, particles are two-way coupled to the fluid, such that a body force at fluid lattice voxel j, $\vec{F}_{f,j}$:

$$\vec{F}_{f,j} = -\sum_i^{i \in j} \vec{F}_{i,a} + \vec{F}_{i,D} \quad \text{(Eq. 5)}$$

is superimposed on the fluid lattice voxel j containing the set of bubbles $i \in j$. Note that this fluid body force does not include the pressure gradient force on the particle, as this force is already incorporated into Eqn. (1).

M-Star Model-Species Transport

Species transport within the fluid is modeled via the advection-diffusion-reaction equation:

$$\frac{dc_j}{dt} = \nabla \cdot \left(D\nabla c_j\right) + \nabla \cdot \left(\vec{v}_j c_j\right) + \frac{\dot{n}_j}{V_j} + R_j \quad \text{(Eq. 6)}$$

where c_j is the species concentration in fluid lattice voxel j, D is the gas diffusion coefficient, $\vec{v}_j$ is the fluid velocity vector, V_j is the volume of the fluid lattice voxel, $\dot{n}_j$ is the species transfer rate between the bubbles and the fluid, and R_j is the local species reaction rate. We use the lattice-Boltzmann particle velocities to solve Eq. 6 at each timestep for each fluid voxel [7]. A van Leer flux limiter is applied to control numerical diffusion [8].

Modeling Set-up

The initial work focused on using the data collected during qualification of Sludge Batch 3 and Sludge Batch 5 to develop and benchmark the model.[9],[10] The model replicated a small scale test apparatus, and used known slurry properties, and operating parameters of the SRAT and SME that were employed during the qualification testing. The model results predicted the hydrogen release rate from the vessel upon start of agitation, which was then compared to the experimental data.

The model developed makes a number of assumptions that must be identified and discussed. These assumptions follow.

- The hydrogen gas bubbles are uniformly distributed in the slurry.
- The slurry yield stress is uniform.
- No bubble coalescence occurs
- The hydrogen gas bubbles release instantaneously when they reach the slurry-air interface
- Surface tension effects that would slow gas bubble release were not included.

The model assumes that the yield stress is uniform throughout the slurry, and the hydrogen gas bubbles are distributed uniformly in the slurry. When the impeller is spinning and the vessel fluid motion has reached “steady-state”, these two assumptions accurately describe the vessel. However, in the DWPF SRAT, when the impellers are stopped, the solid particles will settle to the bottom of the vessel. The insoluble solids concentration will be above the tank average at the bottom of the vessel, and below the tank average at the top of the vessel. Because of the settling, the slurry yield stress will be above the tank average at the bottom of the vessel and below the tank average at the top of the vessel. If the insoluble solids concentration is low enough at the top of the vessel, there will be no yield stress, and no significant hydrogen gas retention in that part of the vessel. When the impellers are stopped, the concentration of hydrogen bubbles is likely to be higher at the bottom of the vessel than at the top.

The model developed for this task did not include bubble breakup from the impeller or bubble coalescence. The M-Star software has the ability to include those phenomena, and they will be added in the future. Previous work demonstrated that momentum forces greatly exceed buoyancy forces under the expected operating conditions. Therefore, the hydogen release rate is not sensitive to hydrogen bubble size over the conditions expected in the testing or the DWPF. The absence of bubble coalescence or breakup should not have a significant effect on the model predictions for this application.

The model assumes that the hydrogen gas bubbles are released when they reach the slurry-gas interface and does not include effects such as bubble surface tension. This assumption will lead to a faster release of hydrogen from the slurry than if these effects are included, resulting in conservative predictions for this application.

The metrics for comparison of the model predictions with the experimental data are the shape of the hydrogen release rate curve, the time after the start of agitation at which the peak hydrogen concentration (hydrogen release rate) is observed, and the time to release 50% of the retained hydrogen.

For the Sludge Batch 3 qualification simulations, the model used 350 lattice points across the diameter of the vessel, a lattice spacing of 0.00031 meters, and a time step of 7 x 10^{-4} seconds. For the DWPF SRAT simulations, the model used 250 lattice points across the vessel diameter, a lattice spacing of 0.015 meters, and a time step of 2.35 x 10^{-4} seconds. For the DWPF SPT simulations, the model used 250 lattice points across the vessel diameter, a lattice spacing of 0.015 meters, and a time step of 3.0 x 10^{-4} seconds.

RESULTS

Comparison with Sludge Batch Qualification Data

Data from qualification testing for Sludge Batch 3 and Sludge Batch 5 showed evidence of retained hydrogen release at the start of agitation.[9],[10] These data were used for benchmarking the hydrogen gas release model.

During Savannah River National Laboratory (SRNL) Sludge Batch 3 Qualification testing, a release of retained hydrogen was measured in the air purge prior to acid addition during the second SRAT Cycle. Using the vessel dimensions, sludge volume, purge rate, and slurry properties, simulations were performed to predict the hydrogen release from the sludge and the hydrogen concentration in the air purge exiting the vessel head space.

The following parameters and operating conditions were used for the Sludge Batch 3 simulations. The sludge/slurry volume was 342 mL, the slurry yield stress was 4.06 Pa, the slurry consistency was 7.37 cP, and the slurry density was 1.27 g/mL.[11],[12] The vessel diameter was 11 cm, and the slurry height was 3.6 cm. The impeller speed was not measured during the testing, but was set so that a small vortex formed (as described in the test report). Simulations were performed with varying impeller speeds to select a speed at which a slight vortex forms, and to assess the sensitivity of hydrogen release rate to impeller speed. The impeller is a 5.08 cm diameter flat blade impeller. The vessel head space has a diameter of 9.87 cm, a height of 21.9 cm, and a volume of 1721 mL. The vessel was purged with air at a rate of 100 mL/min.

A volume of hydrogen gas bubbles was added to the slurry, and the rate at which the bubbles were released was calculated. The volume of bubbles added was selected to approximate the volume of hydrogen release during Sludge Batch 3 qualification testing. A second calculation was performed in which the volume of gas from the released hydrogen bubbles was added to the head space and the rate at which hydrogen gas was removed by the air purge determined. The measured hydrogen concentration in the air purge exiting the vessel in the simulation was compared with the gas chromatograph data collected during Sludge Batch 3 qualification.

Figure 1 shows the geometry of the model used for the simulation of retained hydrogen release during Sludge Batch 3 Qualification testing. The figure on the left shows the vessel containing the sludge, with the impeller. The figure on the right shows the head space with the air inlet and the exhaust, including the rotating shaft from the impeller. While the sludge batch qualification data provide good comparisons to bench-mark the model, effort is being made to find additional data for bench-marking the model at larger scales.

Figure 1. Geometry for Sludge Batch 3 Retained Hydrogen Release Simulation

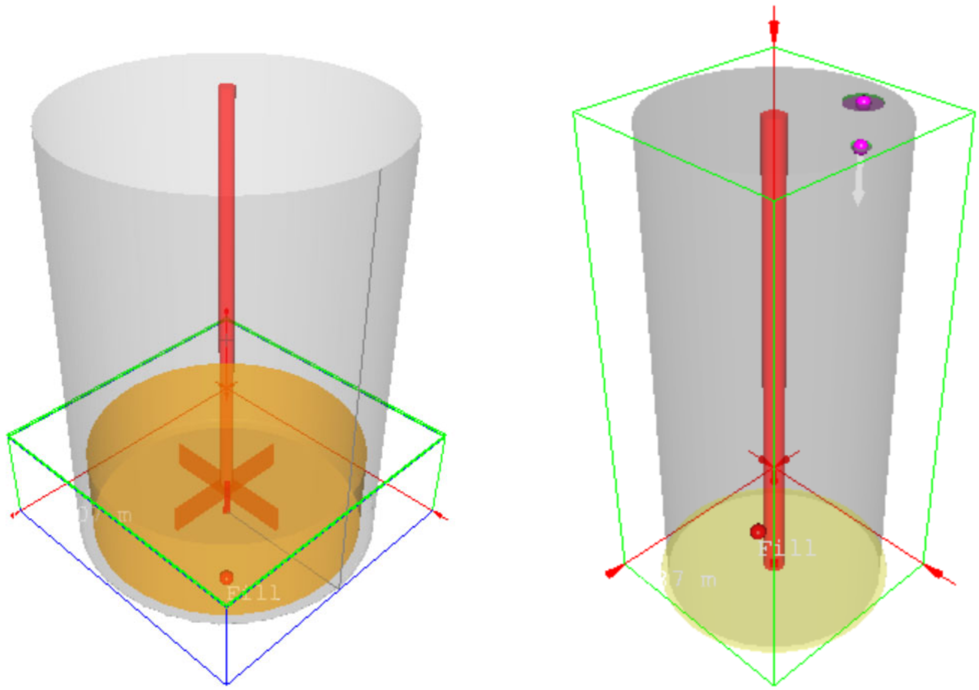


Figure 2 shows the release of retained hydrogen from the slurry during Sludge Batch 3 qualification testing. The graph on the left shows the cummulative release, and the graph on the right shows the release as a function of time. The graphs also show the fit of the data with a model based on the behavior of a non-reactive tracer added to a continuous stirred tank reactor (CSTR). The cumulative amount of tracer (i.e., hydrogen) leaving the CSTR is described by Eq. 7

$$R_c(t) = V_0\,[1 - \exp(-t/\tau)] \qquad \text{(Eq. 7)}$$

where $R_c(t)$ is the cummulative amount of hydrogen released from the slurry, V_0 is the initial amount of hydrogen in the slurry, t is time, and τ is the time constrant for the CSTR. The time constant was determined empirically by fitting the data. The amount of the tracer (i.e. hydrogen) leaving the slurry as a function of time, R(t), is described by Eq. 8.

$$R(t) = V_0 \exp(-t/\tau) \qquad \text{(Eq. 8)}$$

The CSTR model fits the cummulative release data very well, but not the amount of hydrogen leaving the vessel as a function of time. The gas-release as a function of time data differs from the exponential decay function initially, and then demonstrates an exponential decay behavior after ~ 80 seconds. Some plausible explanations for this behavior are the starup of the impeller and the small volume in the vessel. The CSTR model assumes that the tank is at steady-state when the tracer is added. In the simulation (and in the Sludge Batch 3 Qualification test), the impeller was initially at rest, and began spinning when the simulation started. The small volume in the vessel may also have contributed to the initial release not agreeing with CSTR theory. Finally, a significant fraction of the hydrogen gas bubbles were located near the slurry-air interface and may have released during startup of the impeller.

Figure 2. Release of Hydrogen from Slurry During Sludge Batch 3 Qualification

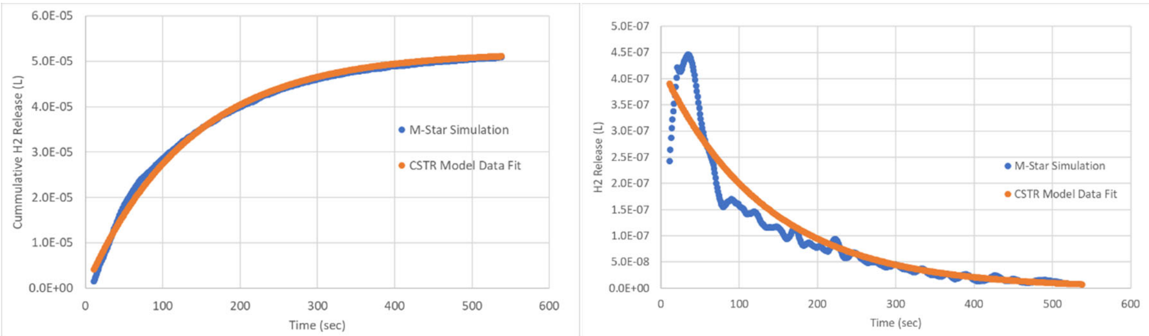
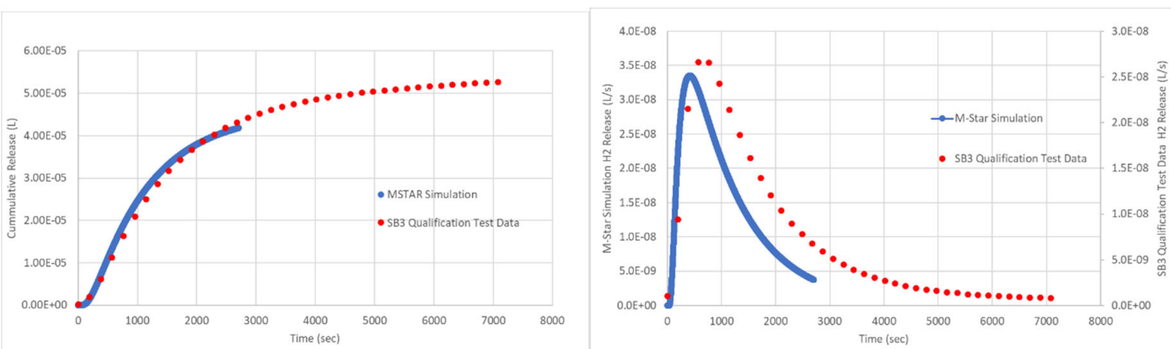


Figure 3 shows the release of retained hydrogen from the head space during Sludge Batch 3 qualification testing. The graph on the left shows the cummulative release, and the graph on the right shows the release as a function of time. The cummulative release data show very good agreement with the experimental data from Sludge Batch 3 Qualification testing. The graph on the right shows simular behavior to the experimental data, but the peak release rate occurs ~ 25% sooner.

Figure 3. Release of Retained Hydrogen from Headspace During Sludge Batch 3 Qualification



In Figure 2, 50% of the retained hydrogen is released from the slurry in 85 seconds, and the peak release occurs at 36 seconds. In Figure 3, the simulations predict 50% of the retained hydrogen to be released from the test vessel in ~18 minutes compared with 50% being released from the test vessel in ~19 minutes during Sludge Batch 3 Qualification testing. In the simulations, the peak release from the test vessel is observed after 417 seconds compared with 573 seconds in the Sludge Batch 3 Qualification testing.

Simulations of Sludge Batch 5 qualification were also performed and agreed well with the experimental data. Those results are not included here for brevity.

The good agreement between the M-Star® simulations with the sludge batch qualification data, provides confidence that the M-Star® software can describe the mixing and hydrogen release in the DWPF vessels as well.

Application of Model to DWPF SRAT

Following the verification of the M-Star model with Sludge Batch Qualification data, the model was used to predict hydrogen release rates in the SRAT in the DWPF. The SRAT is 3.66 m (12 ft) diameter and 4.27 m (14 ft) tall, with a volume of ~45.42 m^3 (12,000 gallons). The maximum working volume of sludge is 34 m^3 (9,000 gallons) with a minimum vapor space of 7.57 m^3 (2,000 gallons). These volumes were adjusted to match the operating conditions corresponding to the DWPF Sludge Batch 8 operating data. The vessels contain a cooling coil assembly that is composed of three sets of coils. The coils surround the agitator and the assemblies have diameters of 1.18, 1.33, and 1.49 m (46.5, 52.5, and 58.5 inches). The coils are constructed of 2 inch schedule 40 pipe. The cooling coil assembly forms a draft tube which is ~1.14 m (45 in) inner diameter and ~1.52 m (60 in) outer diameter. The vessels are mixed with two impellers, a 0.91 m (3 ft) diameter 4 bladed radial flow impeller near the bottom of the cooling coil assembly, and a 0.91 m (3 ft) diameter 3 blade axial flow impeller near the top of the cooling coil assembly. The impeller speed is either 65 rpm or 130 rpm. These dimensions and operating parameters were used to model slurry mixing and hydrogen release in the SRAT, SME, and MFT. The following parameters were used for the simulations: 31.2 m^3 (8,250 gallon) slurry volume, 1.16 g/mL slurry density, 7.2 Pa slurry yield stress, 9.1 cP slurry consistency, and 272 SCFM air purge rate.

Figure 4 shows the model of the SRAT. The figure on the left shows the slurry-containing part of the SRAT, and the figure on the right shows the head space of the SRAT.

Figure 4. Model of DWPF SRAT

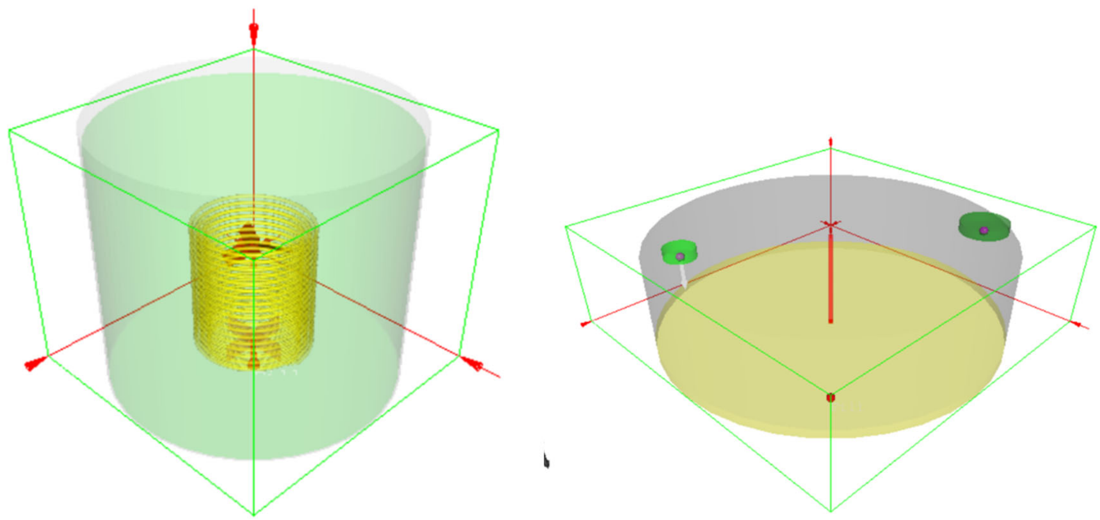


Figure 5 show the rate of release of hydrogen from the slurry in the SRAT. The graph on the left shows the cumulative release rate, along with the fit of the data to a CSTR model. The graph on the right shows the hydrogen release rate from the slurry as a function of time. It is fit with an exponential decay function, which also comes from modeling the vessel as a CSTR. The exponential decay function also shows a good fit to the data, but with some deviation from the equation fitting the data. The data show that 50% of the hydrogen is released from the slurry in ~ 26 minutes. The peak release of hydrogen from the slurry occurred at 208 seconds (~3.5 minutes) after the start of agitation.

Figure 5. Release of Hydrogen from Slurry in the SRAT

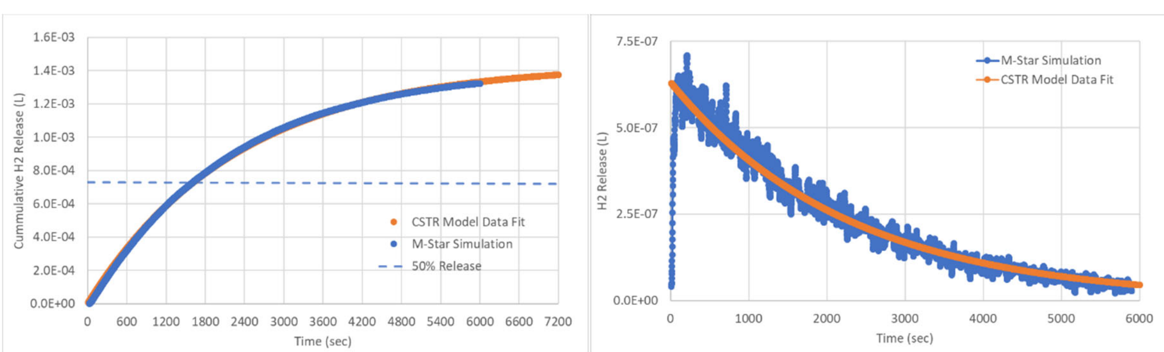
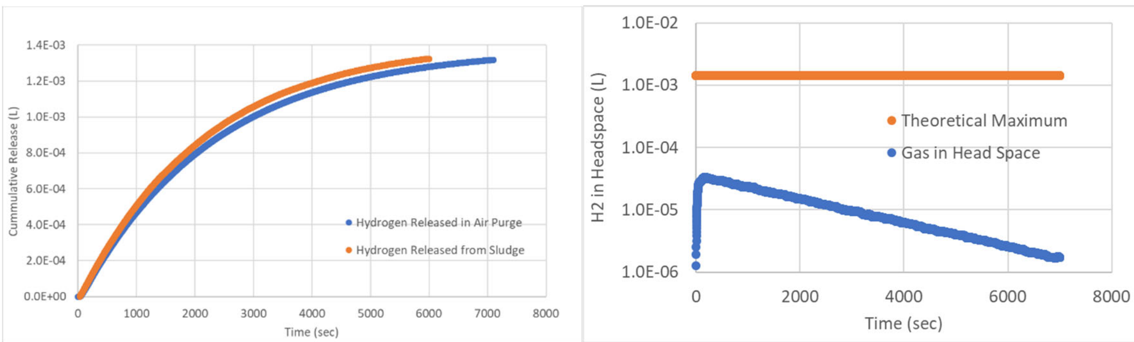


Figure 6 shows the hydrogen release from the SRAT head space and the hydrogen volume in the SRAT head space. The graph on the left shows the cumulative volume of hydrogen released from the head space. The cumulative volume of hydrogen released from the slurry is shown for comparison. The graph shows that the released hydrogen is very rapidly removed from the head space by the air purge. The graph on the right shows the hydrogen volume in the head space as a function of time. This graph allows for comparison of the hydrogen concentration in the head space with the hydrogen volume in the head space if all of the retained hydrogen was release instantaneously upon the start of the agitator. This value is referred to as the theoretical maximum. The maximum hydrogen volume in the head space is more than an order of magnitude less than the theoretical maximum. The data show that 50% of the retained hydrogen is released from the head space in ~29 minutes, and the peak volume of hydrogen in the head space occurs in 3 minutes after the start of agitation.

Figure 6. Hydrogen Release from and Hydrogen Volume in SRAT Head Space



An additional simulation was performed in which 1 volume % hydrogen was added to the slurry under the same operating parameters, and its release simulated. Figure 7 shows the results. The graph on the left shows the cumulative hydrogen release as a function of time. The graph on the right shows the release rate as a function of time. The data show that 50% of the retained hydrogen is released in 21 minutes, and the peak hydrogen release occurs between 2 and 4 minutes after the start of agitation.

Figure 7. Hydrogen Release from SRAT Slurry with 1 Volume% Retained Hydrogen

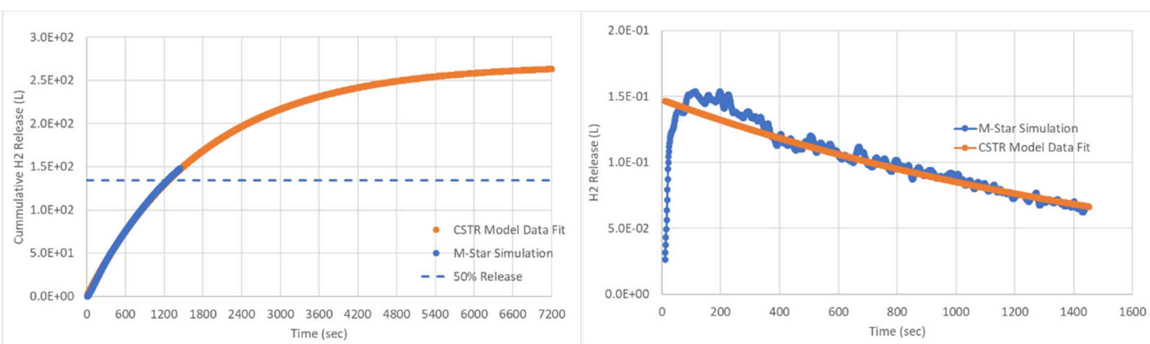
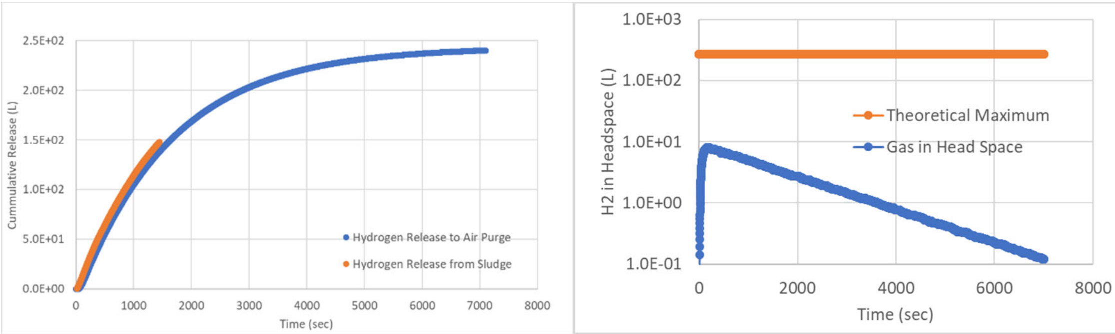


Figure 8 shows the hydrogen release from the SRAT head space. The graph on the left shows the cumulative volume of hydrogen released from the head space. The cumulative volume of hydrogen released from the slurry is shown for comparison. The graph shows that the released hydrogen is very rapidly removed from the head space by the air purge. The graph on the right shows the hydrogen volume in the head space as a function of time. This graph allows for comparison of the hydrogen volume in the head space with the hydrogen concentration in the head space if all of the retained hydrogen was release instantaneously upon the start of the agitator. This value is referred to as the theoretical maximum. The maximum volume in the head space is more than an order of magnitude less than the theoretical maximum. The data show that 50% of the retained hydrogen is released from the head space in ~23 minutes, and the peak volume of hydrogen in the head space occurs in 3 minutes after the start of agitation.

Figure 8. Hydrogen Release from SRAT Head Space with 1 Volume% Retained Hydrogen



Comparing Figures 5 - 8, increasing the hydrogen bubble concentration from 0.00005% to 1% decreased the time for 50% of the retained hydrogen to be released from the slurry from 23 minutes to 21 minutes, and the time for 50% of the retained hydrogen to be released from the head space from 29 minutes to 23 minutes.

Application of the Model to Sludge Pump Tank

To model hydrogen release from the Sludge Pump Tank (SPT), computer aided design (CAD) models were provided to the authors and used to build the geometry of the tank. The properties of the slurry in the SPT were the same as the properties in the SRAT. The volume of the sludge was 21.2 m^3 (5,605 gallons), and the volume of the head space was 5.3 m^3 (1,395 gallons). Figure 9 shows the model of the SPT.

Figure 9. Sludge Pump Tank Model

Figure 10 shows the rate of removal of hydrogen from the sludge pump tank slurry. The graph on the left side of the figure shows the cumulative hydrogen release from the slurry, along with a fit of the data to a CSTR model. The cumulative release shows the same behavior as seen with Sludge Batch 3 qualification and the SRAT. The graph shows that 50% of the hydrogen is released from the slurry in ~15 minutes. The graph on the right shows the hydrogen release as a function of time. The data show a rapid initial release to a peak, and then an exponential decay. The peak release occurs ~3 minutes after the start of agitation.

Figure 10. Hydrogen Release from Sludge Pump Tank Slurry

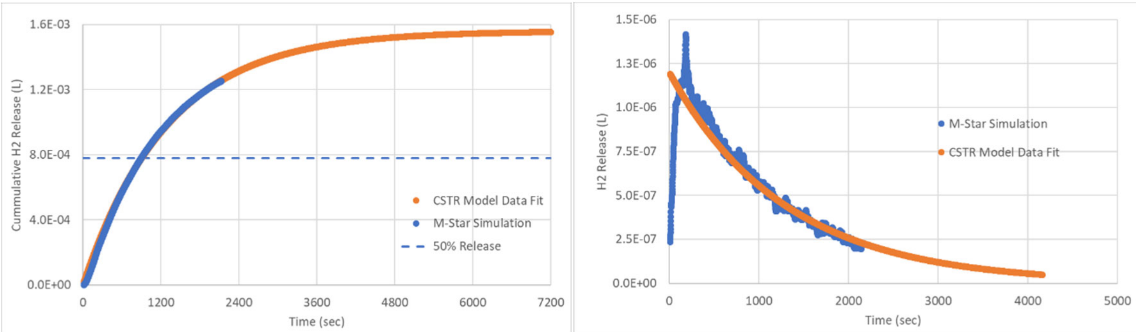
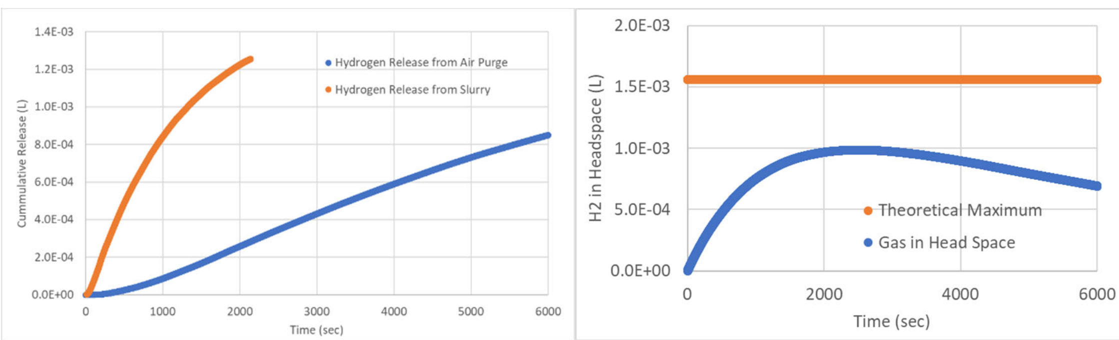


Figure 11 shows the hydrogen release from the SPT head space. The graph on the left shows the cumulative hydrogen release, along with the hydrogen release from the slurry for comparison. The graph shows that a significant time is required to remove the hydrogen released from the slurry from the head space. The time to remove a volume of hydrogen equal to 50% of the hydrogen retained in the sludge is ~ 90 minutes (5,400 seconds). The graph on the right shows the hydrogen volume in the head space as a function of time. For comparison, the theoretical maximum volume that would be in the head space if all of the hydrogen retained in the slurry was instantaneously released is presented. The peak volume of hydrogen in the head space is approximately 60% of the theoretical maximum, and occurs ~41 minutes after the start of agitation.

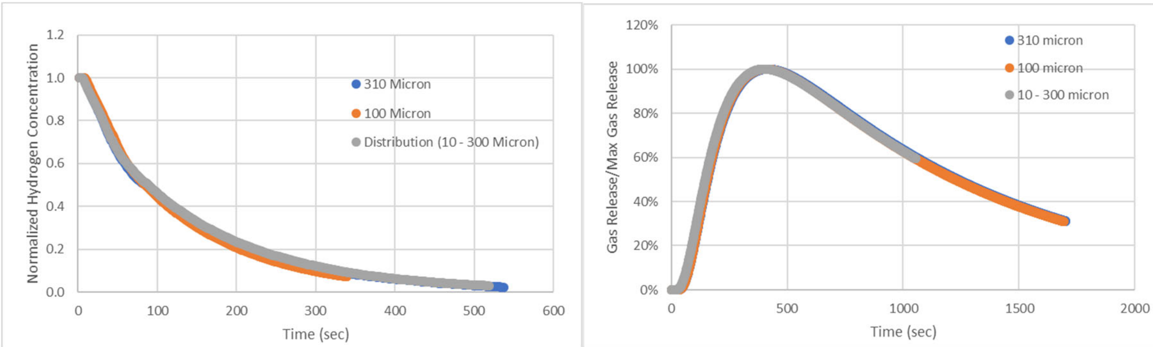
Figure 11. Hydrogen Release from SPT Head Space



Sensitivity of Model Parameters

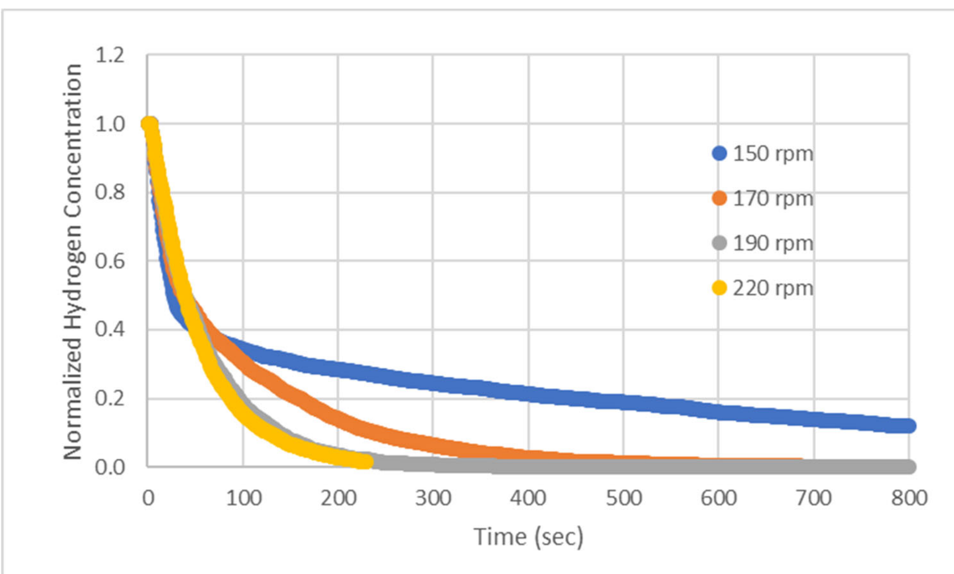
During the simulations of retained hydrogen release during Sludge Batch 3 qualification, the bubble size was varied to examine its effect on the hydrogen release rate. The bubble sizes investigated during these simulations were 310 micron (expected bubble size in the DWPF SRAT), 100 micron (expected bubble size in the Sludge Batch 3 qualification), and a bubble size distribution between 10 micron and 300 micron. Figure 12 shows the results. The graph on the left shows the normalized hydrogen concentration (the hydrogen concentration in the sludge divided by the initial hydrogen concentration in the sludge) in the sludge as a function of time. The graph shows no significant effect of hydrogen bubble size on the rate of release from the sludge. This result is expected. At this range of bubble sizes, the momentum forces from the impeller and fluid motion greatly exceed the buoyancy forces acting on the bubbles. The graph on the right shows the concentration of hydrogen in the head space as a function of bubble size and time. Again, there is no significant effect of bubble size.

Figure 12. Impact of Bubble Size on Hydrogen Release during Sludge Batch 3 Qualification Testing



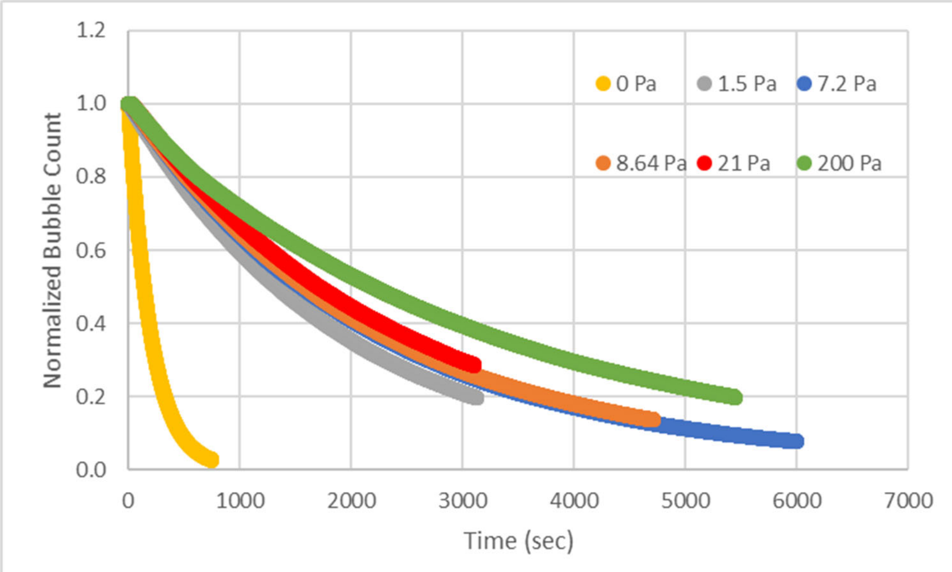
During the simulations of retained hydrogen release during Sludge Batch 3 qualification, the impeller speed was varied to select a speed that best described the mixing conditions during the test and to examine its effect on the hydrogen release rate. Figure 13 shows the normalized hydrogen gas bubble concentration in the slurry as a function of time for impeller speeds of 150, 170, 190, and 220 rpm. The figure shows that hydrogen release rate increases with increasing impeller speed, as expected. However, there is not much difference in the release rate between an impeller speed of 190 rpm and an impeller speed of 220 rpm.

Figure 13. Impact of Impeller Speed on Hydrogen Release during Sludge Batch 3 Qualification Testing



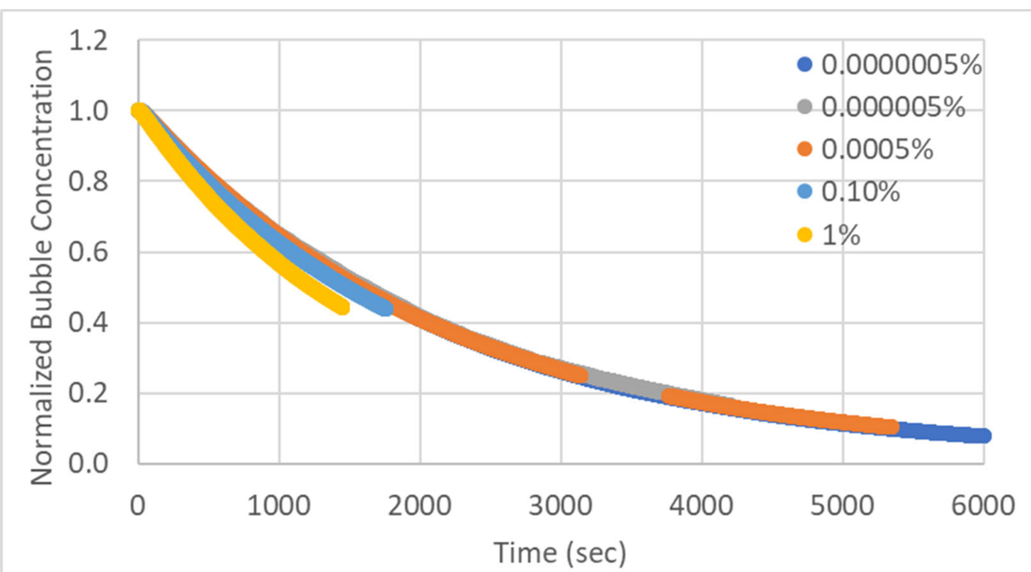
During the simulations of retained hydrogen release in the SRAT, the yield stress was varied to examine its effect on the hydrogen release rate. Figure 14 shows the results. The results show that between a yield stress of 1.5 Pa and 21 Pa, yield stress has a small effect on the release of hydrogen from the slurry in the SRAT (a 27% difference in the time to release 50% of the hydrogen between 1.5 Pa and 21 Pa). At a yield stress of 200 Pa, the time to release 50% of the retained hydrogen is 29% longer than at 21 Pa). With no yield stress, the hydrogen release rate is much faster, but it is not instantaneous.

Figure 14. Impact of slurry Yield Stress on Hydrogen Release from the DWPF SRAT



During the simulations of retained hydrogen release in the SRAT, the hydrogen bubble concentration was varied to examine its effect on the hydrogen release rate. Figure 15 shows the results. At bubble concentrations of 0.0005% and less, no effect of bubble concentration on the hydrogen release rate is observed. At a hydrogen bubble concentration of 1%, an increase in the hydrogen gas release rate is observed. At the concentrations less than or equal to 0.0005 vol%, 50% of the retained hydrogen was released in ~26 minutes. At a concentration of 0.1 vol%, 50% of the retained hydrogen was released in ~25 minutes. At a concentration of 1%, 50% of the retained hydrogen is released in ~21 minutes. At bubbles concentrations of 0.1 vol % and less, the effect of bubble concentration is small. At bubbles concentrations of 1 vol %, the effect becomes noticeable. Additional simulations should be performed at higher bubble concentrations.

Figure 15. Effect of Hydrogen Bubble Concentration on Release Rate from Slurry



CONCLUSIONS

The conclusions from this work follow.

- The model was used to simulate the release of retained hydrogen during Sludge Batch 3 qualification testing, and was found to agree well with the test data.
- Model simulations of the DWPF SRAT predicted 50% of the retained hydrogen would be removed in 21 – 29 minutes depending on operating conditions.
- Model simulations of the DWPF SRAT showed that the maximum hydrogen concentration in the head space would be more than an order of magnitude below the theoretical maximum corresponding to all retained hydrogen being released instantaneously.
- Model simulations of the DWPF SPT predicted 50% of the retained hydrogen would be removed in ~15 minutes based on Sludge Batch 8 properties and planned SPT operating conditions.
- Model simulations of the DWPF SPT showed that the maximum hydrogen concentration in the head space would be ~60% the theoretical maximum corresponding to all retained hydrogen being released instantaneously.
- Investigation of the impact of bubble size on the release of hydrogen from slurries shows no significant impact for the range of bubble sizes expected at the DWPF (10 – 310 micron). This result was expected, because momentum forces generated by the impeller are much larger than the buoyancy forces acting on the bubbles.
- Investigation of the impact of yield stress on the release of hydrogen from the SRAT shows a small effect when the yield stress is between 1.5 and 21 Pa.
- Investigation of the effect of bubble concentration on the release of hydrogen from slurries shows no significant impact when the concentration is less than 0.0005%, and a noticeable effect at a concentration of 1 volume%.
- Investigation of the effect of impeller speed on the release of hydrogen from slurries shows hydrogen bubble release to increase with increasing impeller speed, as expected.

REFERENCES

1. Succi, Sauro. 2001. *The lattice Boltzmann equation: for fluid dynamics and beyond.* Oxford University Press.
2. Krüger, T., Kusumaatmaja, H., Kuzmin, A., Shardt, O., Silva, G., Viggen, E.M. 2017. *The Lattice Boltzmann Method.* Springer International Publishing.
3. Huidan Yu, Sharath S. Girimaji, and Li-Shi Luo. 2005. "DNS and LES of decaying isotropic turbulence with and without frame rotation using lattice Boltzmann method." *Journal of Computational Physics* 209 (2): 599-616.
4. M-Star CFD. 2020. www.mstarcfd.com.
5. Rüde., Christoph Rettinger and Ulrich. 2018. "A coupled lattice Boltzmann method and discrete element method for discrete particle simulations of particulate flows." *Computers & Fluids* 172: 706-719.
6. Tildesley., Michael P Allen and Dominic J. 2017. *Computer simulation of liquids.* 2nd. Oxford University Press.
7. V. E. Küng, F. Osmanlic, M. Markl, and C. Körner. 2020. "Comparison of passive scalar transport models coupled with the Lattice Boltzmann method." *Computers & Mathematics with Applications* 79 (1): 55-65.
8. Leer, Bram Van. 1979. "Towards the ultimate conservative difference scheme. V. A second-order sequel to Godunov's method." *Journal of Computational Physics* 32 (1): 101-136.
9. J. M. Pareizs, D. C. Koopman, D. R. Click, A. D. Cozzi, and N. E. Bibler, "Sludge Batch 3 Qualification in the SRTC Shielded Cells", WSRC-TR-2004-00050, May 2004.
10. J. M. Pareizs, C. J. Bannochie, D. R. Click, D. P. Lambert, M. E. Stone, B. R. Pickenheim, A. L. Billings, and N. E. Bibler, "Sludge Washing and Demonstration of the DWPF Flowsheet in the

SRNL Shielded Cells for Sludge Batch 5 Qualification", SRNS-STI-2008-00111, November 2008.

11. J. M. Pareizs, D. C. Koopman, D. R. Click, A. D. Cozzi, and N. E. Bibler, "Sludge Batch 3 Qualification in the SRTC Shielded Cells", WSRC-TR-2004-00050, May 2004.
12. J. M. Pareizs, T. L. Fellinger, and D. R. Click, "Characterization of the March 2004 Tank 40 (Sludge Batch 3) Dip Samples", WSRC-TR-2004-000208, May 2004.