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INVESTIGATION OF FLAMMABLE GAS RELEASES FROM HIGH LEVEL WASTE TANKS DURING PERIODIC MIXING

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ABSTRACT

The Savannah River Site processes high level radioactive waste through precipitation by the addition of sodium tetraphenylborate in a large (~1.3 million gallon) High Level Waste Tank. Radiolysis of water produces a significant amount of hydrogen gas in this slurry. During quiescent periods the tetraphenylborate slurry retains large amounts of hydrogen as dissolved gas and small bubbles. When mixing pumps start, large amounts of hydrogen release due to agitation of the slurry. Flammability concerns necessitate an understanding of the hydrogen retention mechanism in the slurry and a model of how the hydrogen releases from the slurry during pump operation.

Hydrogen concentration data collected from the slurry tank confirmed this behavior in the full-scale system. These measurements also provide mass transfer results for the hydrogen release during operation. The authors compared these data to an existing literature model for mass transfer in small, agitated reactors and developed factors to scale this existing model to the 1.3 million gallon tanks in use at the Savannah River Site. The information provides guidance for facility operations.

Background Information

The retention and release of flammable gases plays an integral part in the storage of high level radioactive waste. High level waste tanks store a variety of waste forms at the Savannah River Site with a variety of rheological properties. These waste forms include insoluble sludges and crystallized salt cake and tetraphenylborate slurries, all with associated supernate salt solutions. These studies examined the mass transfer coefficients of flammable gases in a waste tank containing non-Newtonian tetraphenylborate slurry. Depending on the density of the salt solution and agitation of the tank, the slurry solids may collect at the bottom of the tank or the surface of the salt solution or may be dispersed throughout the liquid.

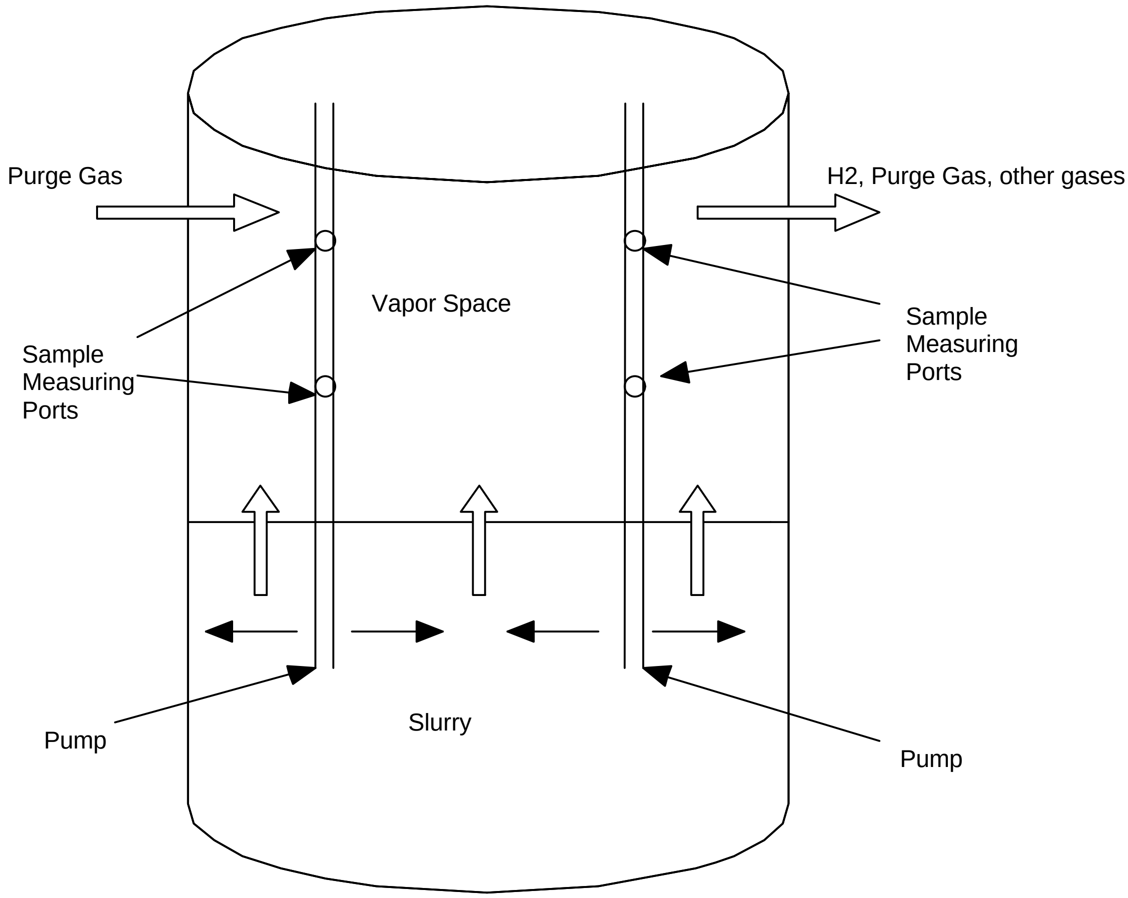
Hydrogen gas forms from radiolysis of water in Savannah River Site High Level Waste Tanks. The release of the hydrogen gas from the high level waste primarily occurs through the operation of mixing pumps in the tank. As the pumps operate, hydrogen gas transfers through the boundary layer and accumulates in the vapor space. This accumulation poses an important safety hazard. Knowledge of the mass transfer coefficient for hydrogen allows definition of operating schedules for the pumps that avoid unsafe conditions. Without this information, it becomes extremely difficult to knowingly operate the pumps long enough to rid the solution of the hydrogen while avoiding an unsafe concentration of hydrogen gas in the vapor space.

Figure 1 provides a simplified schematic of the tank. The waste tank in question measures approximately 25.9 m in diameter with a total volume of about 4920 m^3. The design includes an approximately 2.1-meter diameter center support column in the tank, along with numerous cooling coils for removing radiolytically generated heat. The tank contains four rotating slurry pumps spaced evenly in the four quadrants of the tank. Two sample measuring ports provide the capability to sample the vapor space at several heights. Gas chromatography determines the concentration of the flammable gases.

The pumps operate by pulling the liquid solution in through the bottom and expelling the slurry in jets. The pumps aid hydrogen release by increasing the agitation in the tank and pushing the hydrogen through the diminishing boundary layer at the liquid-vapor interface.

The authors used operational parameters such as the number of active pumps when comparing the mass transfer coefficient (k) for different operational times. By increasing the amount of agitation in the system, the boundary layer, which opposes transfer, diminishes. Therefore, any alteration in the operational parameters that changes the amount of agitation in the tank will affect the mass transfer coefficient. Since increasing the number of operational pumps increases the agitation in the tank, increases in the k values result.

Figure 1. Schematic of Tank and Pump



Data Regression

The concentration in the vapor phase can be written as

$$Cb_y(t) = C1 * e^{r1*t} + C2 * e^{r2*t} \quad (1)$$

where

$$C1 = \frac{((k * A * Cb_{y,t=0}) - (Cb_{y,t=0} * Q) - (k * A * Cb_{y,t=0} / m) - (V_y * r2 * Cb_{y,t=0}))}{V_y * (r1 - r2)}, \quad (2)$$

$$C2 = CB_{y,t=0} - C1, \quad (3)$$

$$r1 = 0.5 * \left(-b + \sqrt{b^2 - 4c}\right), \tag{4}$$

$$r2 = 0.5 * \left(-b - \sqrt{b^2 - 4c}\right), \tag{5}$$

$$b = \frac{V_y * k * A * m + V_x * Q * m + V_x * k * A}{V_x * V_y * m} \text{ and} \tag{6}$$

$$c = \frac{Q * k * A}{V_x * V_y}. \tag{7}$$

Tank dimensions or monitoring provide all underlying variables for equation 1 except the initial concentration of hydrogen in the liquid phase and the mass transfer coefficient. The authors regressed the measurements obtained from the tank to provide values for the initial concentration of hydrogen in the slurry phase and the mass transfer coefficient for a given set of operating conditions. The regression minimized the difference between the experimentally measured hydrogen concentration in the vapor space and the estimated vapor space concentration.

Discussion of Results

The authors regressed data obtained from one high level waste tank at various times over a four year period. During these time periods, the operational parameters (i.e., number of pumps) and the system parameters (i.e., liquid level, volumetric flow rate of the purge gas, head space temperature and pressure) varied. Personnel calculated a 95% confidence interval, using an F-Test, on each k value. The F-Test consisted of two variables (k and $Cb_{y,t=0}$) with the number of observations ranging from 60 to over 400. These statistical tests indicate the overall accuracy of the regression with the existing data and the precision of the mass transfer coefficient.

Table 1. Data Regression Results

Regression #	Number of Pumps	*k* (*10^5 m/s)	95% confidence interval (*10^5 m/s)	R^2
1	4	2.83	2.72, 2.95	0.937
2	1	1.19	1.15, 1.21	0.932
3	3	3.16	2.84, 3.44	0.955
4	4	3.25	3.09, 3.41	0.979
5	4	2.95	2.90, 3.03	0.987
6	1	3.20	3.08, 3.31	0.924
7	3	3.10	3.03, 3.21	0.994

Figure 2 represents a time period of approximately 700 minutes while four pumps operated. The collected data demonstrates a noticeable curvature, and the regression (solid black line) closely represents this curve. Figure 3 represents a time period of approximately 1600 minutes in November of 1995. During this time period, the data points exhibit more scatter. Although some curvature exists, the breaks in the experimental data diminish the effectiveness of the regression. The regression (solid black line) provides a poorer reproduction of the hydrogen concentration in the vapor space over the 1600 minute duration.

Figure 2. Vapor Phase Hydrogen Concentration During May 1998 Pump Run.

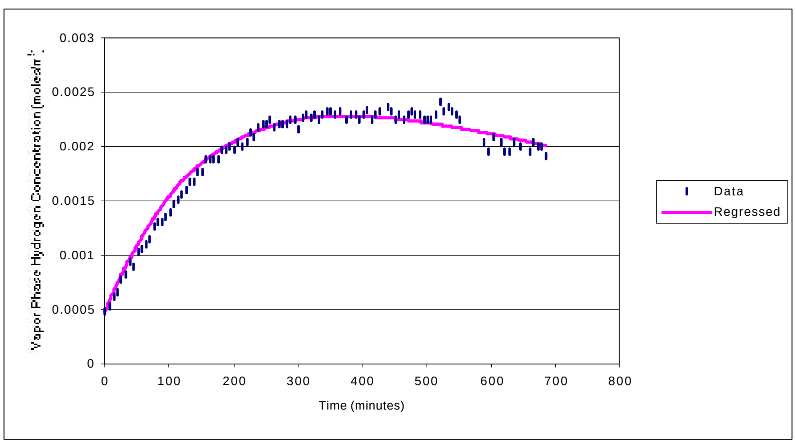
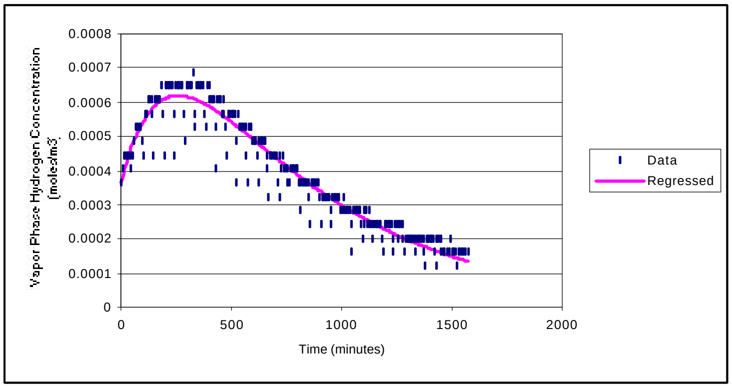


Figure 3. Vapor Phase Hydrogen Concentration During November 1995 Pump Run



The authors used a published model by Y. Kawase and M. Moo-Young(1990). The Kawase and Moo-Young model assumes a periodic transitional sublayer model. This model assumes that the liquid-vapor free surface serves as a rigid boundary. The Kawase and Moo-Young model design considered tanks containing an impeller and differs from other published models concerning the exponential dependencies of the diffusivity, impeller length, viscosity, and power number on the mass transfer coefficient. Also, the model proposed by Kawase and Moo-Young neglects the effect of surface tension on the liquid-phase mass transport coefficient.

For a Newtonian liquid (n = 1), the Kawase and Moo-Young model takes the following form:

$$k = 0.138 * Sc^{-2/3} * ne \; . \qquad (8)$$

The Schmidt number (*Sc*), a dimensionless term, provides the ratio between the diffusional and kinetic transport for a system.

The Kawase and Moo-Young model provides the relationship between the mass transfer coefficient and the energy dissipation per unit mass (*e*). As the number of active pumps increases, the amount of energy dissipated also increases. Thus, this model indicates that as the number of active pumps increases, the mass transfer coefficient will also increase. In calculating the *k* values with the Kawase and Moo-Young model, the authors assumed the pumps operated at 100 hp and 50% efficiency (Internal Savannah River Site Vendor File 216146, “Bingham 4x18 VRM Pump”, Sheet #31), and the addition of an extra pump essentially increases the tank energy by 50% of 100 hp. Table 2 provides a comparison of the regressed and predicted mass transfer coefficients for the same data sets analyzed in Table 1.

Table 2. Comparison between Data Regression and Kawase and Moo-Young Model

Regression	Number of Operating Pumps	k (*10^5 m/s)	Kawase and Moo-Young k 50% efficiency (*10^5 m/s)	Ratio of Calculated k to Kawase and Moo-Young k at 50% Efficiency
1	4	2.83	1.06	2.67
2	1	1.19	0.749	1.55
3	3	3.16	0.892	3.54
4	4	3.25	0.955	3.40
5	4	2.95	0.958	3.08
6	1	3.20	0.756	4.23
7	3	3.10	0.863	3.59

The Kawase and Moo-Young model gives results that average a factor of 3.15 (with a standard deviation of 0.85) lower than those calculated from operating data. The relatively constant offset allows one to estimate mass transfer coefficients for future pump runs and therefore predict the rate at which hydrogen releases.

One possible cause for the lack of accuracy in this application of the Kawase and Moo-Young model involves the energy dissipation term. As seen in the Kawase and Moo-Young equation, the mass transfer coefficient depends on the energy dissipation per unit mass (e). The Kawase and Moo-Young model derived from data collected in a tank using an impeller, not a pump. In using the Kawase and Moo-Young equation the authors assumed that the power dissipated and the agitation induced by an impeller equals that for operation of a pump. No data exists to determine the validity of this assumption for tetraphenylborate slurries.

Conclusions

Researchers determined the mass transfer coefficients for hydrogen through a tetraphenylborate slurry in a nuclear waste tank. Comparison with mass transfer coefficients calculated from a literature model indicates a consistent offset. Researchers will use this factor to modify the model to predict hydrogen mass transfer coefficients for future tank pump runs.

Future studies will compare the modified model to operational data from other waste tanks -- as it becomes available -- to determine whether accurately predicts the hydrogen mass transfer coefficients for alkaline solutions and for slurries containing metal hydroxide sludge.

Nomenclature

A	surface area (m^2)
Cb_y	concentration in vapor phase ($mole/m^3$)
D	diffusivity of hydrogen in slurry (m^2/s)
K	mass transport coefficient (m/s)
m	Henry's Law Constant (unitless)
n	flow index (unitless)
Q	volumetric purge rate (m^3/s)
Sc	Schmidt number (u/D - unitless)
t	time (s)
V_x	liquid volume (m^3)
V_y	vapor volume (m^3)
e	energy dissipation per unit mass (W/kg)
u	kinematic viscosity (m^2/s)

Reference

Kawase, Y. and Moo-Young, M., 1990, Chemical Engineering Research and Design, 68: 189-194.