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Modeling Air Stripping of Ammonia in an Agitated Vessel

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ABSTRACT
A model has been developed to predict the rate of removal of ammonia (NH₃) from solution in a sparged agitated vessel. The model is first-order with respect to liquid-phase concentration of NH₃. The rate constant for the first-order equation is a function of parameters related to the vessel/impeller characteristics, the air/liquid properties as well as the process conditions. However, the vessel/impeller characteristics, the air/liquid properties, and the process conditions reduce the rate constant dependence to only three parameters, namely, the air sparge rate, the liquid volume or batch size, and the Henry’s law constant of NH₃ for the liquid or solution. Thus, the rate of removal is not mass-transfer limited. High air sparge rates, high temperatures, and low liquid volumes or batch sizes increase the rate of removal of NH₃ from solution. The Henry’s law constant effect is somewhat reflected in the temperature since Henry’s law constant increases with increasing temperature. Data obtained from actual air stripping operation agree fairly well with the model predictions.

IMPLICATIONS
A model that relies solely on semi-empirical correlations and literature data as inputs rather than actual measurements to quickly predict rate of ammonia (NH₃) removal from solution in sparged agitated vessels has been developed. The model could be a significant tool in operations planning for determining sampling times, optimum process conditions, key controlling process parameters, and time to reach target NH₃ removal levels. It also could be a beneficial tool in the design of batch stripping units. Even though the model is based on NH₃, it could be used for any contaminant in solution that is amenable to air stripping.
INTRODUCTION
Ammonium salts deposit on process vessel vent (PVV) filters during plutonium and uranium extraction (PUREX)\(^1\)\(^2\) operations at United States Department of Energy’s (DOE) Savannah River Site (SRS) in Aiken, South Carolina. To remove the deposits, the PVV filters are flushed with 0.7 M nitric acid solution. This is followed by evaporating the ensuing solution to ~4 M nitric acid, and neutralizing with excess 50 wt % sodium hydroxide (NaOH) solution. The excess NaOH makes the solution alkaline with a pH of ~14. The bulk of the solution typically contains 1 to 4 M sodium nitrate (NaNO\(_3\)) and 1 M NaOH.

The alkaline PVV filter flush solution is sent to a holding vessel for further processing through transfer lines and pump pits. The flammability of ammonia (NH\(_3\)) is well known due to its use in industrial processes\(^3\) and as a refrigerant (R-717).\(^4\) Ignitable concentrations are hard to produce in well ventilated areas. However, in confined areas enough gas can accumulate to produce a combustive explosion.\(^3\) Safety Analysis requires flammability controls be placed on the intermediate collection points. To minimize the NH\(_3\) contribution to the flammability calculations, the NH\(_3\) in the solution is removed by air stripping in an agitated vessel. The estimated target final concentration of NH\(_3\) in solution required to be in compliance in terms of flammability concerns is 0.056 wt %.\(^5\) To aid in operations planning regarding determining sampling times, optimum process conditions, key controlling process parameters, and most importantly when the target NH\(_3\) removal level will be reached; a model was developed to predict the rate of removal of NH\(_3\) from the solution during the stripping process.

The model uses mostly correlations from literature as inputs and experimental data from similar systems instead of actual measured parameters. The model predictions are compared to actual stripping operation data.

MODEL DEVELOPMENT
The vessel being modeled is essentially a bubble column with stagnant liquid and mechanical agitation. In this vessel, air is introduced or sparged into the bottom of the vessel via an orifice and exits at the top of the vessel. In other words, the NH\(_3\) is transferred from the liquid to the air bubbles, and these bubbles are removed as they enter the vessel’s vapor space. The agitation of the liquid ensures uniform distribution of the air throughout the vessel. The mechanical agitation
is accomplished with two flat-blade turbine impellers. Figure 1 is a schematic diagram of the vessel under consideration.

Two mass balances are needed to develop the model: an overall NH$_3$ balance on the vessel, and an NH$_3$ balance on a differential element of the vessel for the air stream. An overall NH$_3$ mass balance on the vessel is given by eq 1.

$$ V_L \frac{dC_L}{dt} + \varepsilon_G V_L \frac{dC_G}{dt} = Q_G C_{Go} - Q_G C_{Gf} $$

where $V_L$ is the liquid volume (m$^3$), $C_L$ is the liquid-phase NH$_3$ concentration (kmol/m$^3$), $t$ is time (sec), $\varepsilon_G$ is the gas (air) holdup or the volume fraction of the air bubbles entrained in the liquid (dimensionless), $C_G$ is the NH$_3$ concentration in the air bubbles (kmol/m$^3$), $Q_G$ is the air sparge rate or air volumetric flow rate (m$^3$/sec), and $C_{Go}$ and $C_{Gf}$ are the NH$_3$ concentration in the air bubbles entering and leaving the vessel respectively (kmol/m$^3$).

For dilute solutions, the accumulation of NH$_3$ in the air is negligible. Hence, the second term in eq 1 is zero. Note also that $C_{Go} = 0$. This yields eq 2.

$$ V_L \frac{dC_L}{dt} = -Q_G C_{Gf} $$

The concentration of NH$_3$ in the air bubbles leaving the vessel is needed to solve eq 2. It is obtained by performing a mass balance on a differential element of the vessel for the air stream. The mass balance is described by eq 3.

$$ \varepsilon_G dz A \frac{dC_G}{dt} = Q_G C_G - Q_G (C_G + dC_G) + dz A k_L a (C_L - C_L^*) $$

where $dz$ is the height of the differential element (m), $A$ is the vessel cross-sectional area (m$^2$), $k_L$ is the liquid-phase mass transfer coefficient (m/s), $a$ is the specific interfacial area of bubbles per unit volume of air-liquid mixture (m$^2$/m$^3$), and $C_L^*$ is the NH$_3$ concentration in the liquid in equilibrium with the air bubbles (kmol/m$^3$). The last term in the equation is the rate of mass transfer across the air-liquid interface. For this air-liquid system, the rate of mass transfer is
liquid-phase controlled.\textsuperscript{6-9} Again, the air accumulation term (i.e., first term) in eq 3 is zero as mentioned earlier.

The equilibrium relationship between the air and liquid is given by the Henry’s law equation.

\[ C_G = K_H C_L \]  

(4)

where \( K_H \) is the Henry’s law constant (kmol/m\(^3\)/kmol/m\(^3\)). Combining eqs 3 and 4, setting the first term in eq 3 to zero followed by rearranging, and integrating over the effective height of the air-liquid mixture with the condition that \( C_G = C_{G_0} \) when \( z = 0 \), and \( C_G = C_{G_f} \) when \( z = H_{Le} \) give

\[
\int_{C_{G_0}}^{C_{G_f}} \frac{dC_G}{C_L - \frac{C_G}{K_H}} = \frac{k_L a A}{Q_G} \int_0^{H_{Le}} dz
\]

(5)

where \( H_{Le} \) is the effective height of the air-liquid mixture or liquid that is entrained with air bubbles (m). The effective height of the liquid that is entrained with bubbles is given by

\[
H_{Le} = H_L \left(1 + \varepsilon_G\right)
\]

(6)

where \( H_L \) is the height of the liquid with no entrained bubbles (m). It must be noted that the NH\(_3\) concentration in the liquid is approximately the same everywhere in the vessel because of the mechanical agitation. In other words, \( C_L \) does not change along the height of the vessel. It changes only with time. Equation 5 agrees with the air stripping equation given by Gonen and Gvirtzman.\textsuperscript{7}

Integrating eq 5 while setting \( C_{G_0} = 0 \) and rearranging give eq 7.

\[
C_{G_f} = K_H C_L \left[1 - \exp\left(-\frac{k_L a A H_{Le}}{Q_G K_H}\right)\right]
\]

(7)
Substituting eq 7 into eq 2, rearranging and integrating with the condition that $C_L = C_{L0}$ when $t = 0$, and $C_L = C_L$ when $t = t$ lead to

\[
\int_{C_{L0}}^{C_L} \frac{dC_L}{C_L} = -\frac{Q_G K_H}{V_L} \left\{ 1 - \exp \left( -\frac{-k_L a AH_{Le}}{Q_G K_H} \right) \right\} \int_0^t dt \tag{8}
\]

Integrating eq 8 and rearranging give eq 9.

\[
\frac{C_L}{C_{L0}} = \exp \left[ -\frac{Q_G K_H}{V_L} \left\{ 1 - \exp \left( -\frac{-k_L a AH_{Le}}{Q_G K_H} \right) \right\} t \right] \tag{9}
\]

Equation 9 is an expression for the relative concentration or the fraction of NH$_3$ remaining in solution at any time $t$. Since $C_{L0}, Q_G, V_L,$ and $A$ are known, the inputs needed to compute the concentration, $C_L$, at any time $t$ are $K_H, k_L, a,$ and $H_{Le}$. Note that eq 9 is essentially a first-order rate equation with the term in the square brackets without the $t$ parameter and the negative sign as the rate constant. Other researchers have found air sparging or stripping from liquid to be first-order with respect to liquid-phase concentration.$^7,^{10,11}$

**CALCULATION OF GASSED IMPELLER POWER**

Two graphs are used in the estimation of gassed impeller power, $P_G$ (W or N-m/sec).$^6$ The first graph relates the impeller power number, $P_n$ (dimensionless), to impeller Reynolds number, $Re_i$ (dimensionless), for several blade turbine impellers in ungassed (or unsparged) baffled vessels. Knowing $Re_i$, $P_n$ is obtained from the graph. The ungassed impeller power, $P_{ng}$(W or N-m/sec), is given as follows.

\[
P_{ng} = \frac{P_n \rho_L N^3 d_i^5}{g_c} \tag{10}
\]

where $Re_i$ is impeller Reynolds number = $d_i^2 N \rho_L / \mu_L$ (dimensionless), $d_i$ is impeller diameter (m), $N$ is impeller speed (revolutions/sec), $\rho_L$ is density of liquid or solution (kg/m$^3$), $\mu_L$ is viscosity of...
liquid or solution (kg/m/sec), and $g_c$ is Newton’s law proportionality factor for gravitational force unit = 1 kg-m/N/sec$^2$.

The second graph relates the ratio of gassed impeller power to ungassed impeller power (i.e., $P_G/P_{nG}$) to $Q_G/Nd_i^3$. The graph comprises two straight lines described by the two equations below.

For $Q_G/Nd_i^3 < 0.037$, \[ \frac{P_G}{P_{nG}} = 1 - 12.2 \left( \frac{Q_G}{Ndi^3} \right) \] (11)

For $Q_G/Nd_i^3 > 0.037$, \[ \frac{P_G}{P_{nG}} = 0.62 - 1.85 \left( \frac{Q_G}{Ndi^3} \right) \] (12)

The ungassed impeller power, $P_{nG}$, calculated using the first graph mentioned earlier is then plugged into either eq 11 or 12 to obtain $P_G$.

**CALCULATION OF MEAN BUBBLE DIAMETER, AIR HOLDUP, AND BUBBLE SPECIFIC INTERFACIAL AREA**

For sparged agitated vessels with flat-blade turbine impellers, and for $Re_i^{0.7} \left( \frac{Nd_i}{U_G} \right)^{0.3} > 30,000$ the following equations apply.\(^5\)

\[ a = \frac{6\varepsilon_G}{d_b} \] (13)

\[ a = \left[ 1.44 \left( \frac{P_G}{V_L} \right)^{0.4} \left( \frac{\rho_L}{g_c \sigma} \right)^{0.2} \left( \frac{U_G}{U_{br}} \right)^{0.5} \right] \left[ 8.33 \times 10^{-5} Re_i^{0.7} \left( \frac{Nd_i}{U_G} \right)^{0.3} - 1.5 \right] \] (14)
\[ d_b = \left[ 2.25 \left( \frac{V_i}{P_G} \right)^{0.4} \left( \frac{g_c \sigma^3}{\rho_L} \right)^{0.2} \left( \frac{\mu_G}{\mu_L} \right)^{0.25} \right]^{0.4} \]  

(15)

where \( U_G \) is superficial velocity of the air (m/sec), \( d_b \) is mean bubble diameter (m), \( \sigma \) is interfacial tension of liquid or solution (N/m), \( U_{br} \) is bubble rise velocity or bubble terminal velocity (m/sec), and \( \mu_G \) is viscosity of air (kg/m/sec). The others parameters have same meanings as defined before.

Substituting eqs 14 and 15 into eq 13, and rearranging give the air holdup

\[ \varepsilon_G = \left[ 0.54 \left( \frac{\mu_G}{\mu_L} \right)^{0.25} \left( \frac{U_G}{U_{br}} \right)^{0.5} \right]^{1.67} \left[ 8.33 \times 10^{-5} \text{Re}^{0.7} \left( \frac{Nd_i}{U_G} \right)^{0.3} - 1.5 \right]^{1.67} \]  

(16)

Equations 15 and 16 are solved by a trial-and-error procedure. A value for \( U_{br} \) is guessed and substituted into eq 16 along with the values for the other parameters in eq 16 which are known. The value for \( \varepsilon_G \) obtained from eq 16 is then substituted into eq 15 along with the values for the other parameters in eq 15 which are also known to compute \( d_b \).

To check whether the guessed \( U_{br} \) value is satisfactory, the \( d_b \) value obtained from eq 15 is substituted into one of the equations below depending on the value of \( d_b \) to calculate \( U_{br} \).

For \( d_b < 0.7 \) mm, \( U_{br} = \frac{gd_b^2(\rho_L - \rho_G)}{18\mu_L} \)  

(17)

For \( d_b > 1.4 \) mm, \( U_{br} = \frac{2\sigma}{d_b\rho_L} + \frac{gd_b}{2} \)  

(18)

where \( g \) is the gravitational acceleration (m/sec²), and \( \rho_G \) is the density of air (kg/m³). The rest of the parameters have same meanings as previously defined. For the intermediate region (i.e., 0.7 mm < \( d_b < 1.4 \) mm), \( U_{br} \) is estimated by interpolating between the two \( U_{br} \) values at \( d_b = 0.7 \) and 1.4 mm.
The guessed \( U_{br} \) value is satisfactory when the \( U_{br} \) value calculated with either eq 17 or 18 agrees with the guessed value. If not, the whole procedure is repeated till the two \( U_{br} \) values agree. Once a satisfactory \( U_{br} \) value is obtained, eqs 13 and 6 can be used to calculate \( a \), the specific interfacial area of bubble, and \( H_{Leo} \), the effective liquid height respectively.

CALCULATION OF LIQUID-PHASE MASS TRANSFER COEFFICIENT

For sparged agitated vessels where the resistance to mass transfer is solely in the liquid phase, the mass transfer coefficient is given by eq 19.6

\[
\begin{align*}
    k_L &= \frac{D_L}{d_b} \left[ 2.0 + 0.31 \left( \frac{\rho_L - \rho_g}{\rho_L} \right) \frac{d_b^3 g}{D_L \mu_L} \right]^{0.333} \\
    (19)
\end{align*}
\]

where \( D_L \) is the diffusivity of NH3 in the solution (m²/sec).

INPUT PARAMETERS

Table 1 shows the input parameters used in the calculations. The diffusivity of NH3 in water at 25°C was found in the literature.12 The diffusivity at other temperatures was calculated with the Wilke-Chang equation.13 The effect of NaNO3 and NaOH on the diffusivity is not known. Data on the diffusivity of potassium chloride in electrolytes show a change in diffusivity of less than 20% as concentration increased from 0 to 2 M.13 Therefore, the diffusivity values were multiplied by 0.8 for conservatism.

The viscosity of the solution at 25 °C was taken as the average of values for 1 to 4 M NaNO3 and 1 M NaOH solutions.14 The viscosity (cP) at other temperatures was calculated with eq 20.

\[
\begin{align*}
    \mu_L^{-0.2661} &= \mu_{L,K}^{-0.2661} + \left( \frac{T - T_K}{233} \right) \\
    (20)
\end{align*}
\]
where $T$ is temperature ($^\circ$C or K) and $\mu_{LK}$ is the known viscosity (cP) at a known temperature, $T_K$. The viscosity of air at 50 and 65 °C was obtained from The Chemical Engineers’ Handbook. The density of the solution was measured at 25 °C. The density at other temperatures was calculated using the average of the temperature dependence trends for 1 to 4 M NaNO₃ and 1 M NaOH solutions. The density of air was calculated using the ideal-gas law equation.

The Henry’s Law constant of NH₃ for the solution at 50 and 65 °C was obtained from the literature for a roughly similar solution (3.7 M NaNO₃, 2.3 M NaOH, and 3.2 M NaNO₂). The interfacial tension of the solution at 20 °C was taken as the average of the values for 1 to 4 M NaNO₃ and 1 M NaOH solutions. The temperature dependence trend for water was used to estimate the values at other temperatures. The interfacial tension values obtained for the solution exhibited very little sensitivity to temperature and was also close to those for water.

RESULTS AND DISCUSSION

Figures 2 to 4 are plots of NH₃ concentration remaining in the liquid versus time for various conditions. The plots were generated using the NH₃ stripping model equation (i.e., eq 9). Figure 2 shows the effect of air sparge rate on the rate of removal of NH₃. The plots indicate that at a given temperature and liquid volume or batch size, the rate of NH₃ removal increases with increasing air sparge rate. The effect of temperature is depicted in Figure 3. High temperatures result in increased rates of removal partly because Henry’s law constant increases as temperature increases. The effect of temperature is more marked at low air sparge rates. Figure 4 shows the effect of liquid volume or batch size. At a given temperature and air sparge rate, the rate of NH₃ removal decreases with increasing liquid batch size. Again, the effect of liquid volume or batch size is more marked at low air sparge rates.

It turned out that the exponential term (i.e., $\exp \left( \frac{-k_i a A H_{le}}{Q_g K_H} \right)$) in eq 9 is approximately zero because the term in parenthesis is large based on the vessel/impeller characteristics, the air/liquid properties, and the process conditions. This implies the rate of removal is not mass-transfer limited. The model equation therefore basically reduces to
The rate of NH$_3$ removal depends essentially on three parameters: $Q_G$, $K_H$ and $V_L$. Enhancement in the rate of removal of NH$_3$ can be effected by varying the air sparge rate ($Q_G$) and/or the liquid volume or batch size ($V_L$). The Henry’s law constant of NH$_3$ ($K_H$) for the liquid or solution is more or less fixed. It is dictated by the concentrations of NaNO$_3$ and NaOH in solution. In practice, the Henry’s law constant of NH$_3$ for the solution can be increased by increasing the concentrations of the NaNO$_3$ and NaOH. This is because increasing concentrations of NaNO$_3$ and NaOH in solution are known to increase the Henry’s law constant of NH$_3$.\textsuperscript{16,17} What is unknown for this solution are the concentration factors that will result in significant increases in Henry’s law constant of NH$_3$.

Figure 5 provides a comparison of the model output with actual stripping operation. The solution was sampled periodically and analyzed for NH$_3$ by ion chromatography. The model prediction is adequate overall.

**CONCLUSIONS**

The model developed for NH$_3$ stripping from PVV filter flush solution follows first-order kinetics with respect to liquid-phase NH$_3$ concentration, a feature that seems to be typical for air sparging of aqueous liquids. Based on the vessel/impeller characteristics, the air/liquid properties, and the process conditions, the rate of removal of NH$_3$ from solution is not mass-transfer dependent. Rather, the rate of removal of NH$_3$ from solution is enhanced by high air sparge rates, high temperatures, and low liquid volumes per batch. The effect of both temperature and liquid volume or batch size is more marked at low air sparge rates. The model agrees reasonably well with actual stripping data.

**ACKNOWLEDGEMENTS**

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flammability calculations to determine the target final concentration of NH₃ in solution required to be in compliance.

REFERENCES


**NOMENCLATURE**

The following symbols are used in this paper.

\[
\begin{align*}
A & = \text{vessel cross-sectional area (m}^2) \\
a & = \text{specific interfacial area of bubbles per unit volume of air-liquid mixture (m}^2/\text{m}^3) \\
C_G & = \text{NH}_3 \text{ concentration in the air bubbles (kmol/m}^3) \\
C_{Go} & = \text{NH}_3 \text{ concentration in the air bubbles entering the vessel (kmol/m}^3) \\
C_{Gr} & = \text{NH}_3 \text{ concentration in the air bubbles leaving the vessel (kmol/m}^3) \\
C_L & = \text{liquid-phase NH}_3 \text{ concentration (kmol/m}^3) \\
C_{L0} & = \text{initial liquid-phase NH}_3 \text{ concentration (kmol/m}^3) \\
C_{Lf} & = \text{final liquid-phase NH}_3 \text{ concentration (kmol/m}^3) \\
C_{L*} & = \text{NH}_3 \text{ concentration in the liquid in equilibrium with the air bubbles (kmol/m}^3) \\
d_b & = \text{mean bubble diameter (m)} \\
d_i & = \text{impeller diameter (m)} \\
D_L & = \text{diffusivity of NH}_3 \text{ in the solution (m}^2/\text{sec}) \\
dz & = \text{height of the differential element (m)} \\
g & = \text{gravitational acceleration (9.807 m/sec}^2) \\
g_c & = \text{Newton’s law proportionality factor for gravitational force unit = 1 kg-m/N/sec}^2
\end{align*}
\]
HL = height of the liquid with no entrained bubbles (m)
HLe = effective height of air-liquid mixture or liquid that is entrained with air bubbles (m)
kL = liquid-phase mass transfer coefficient (m/sec)
KH = Henry’s law constant (kmol/m³/kmol/m³)
N = impeller speed (revolutions/sec)
P_G = gassed impeller power (W or N-m/sec)
P_n = impeller power number (dimensionless)
P_nG = no-gas impeller power (W or N-m/sec)
Q_G = air sparge rate or air volumetric flow rate (m³/sec)
Re_i = impeller Reynolds number (dimensionless)
t = time (sec)
T = temperature (°C or K)
T_K = known temperature of a liquid with a known viscosity µ_LK (°C or K)
U_G = superficial velocity of the air (m/sec)
U_br = bubble rise velocity or bubble terminal velocity (m/sec)
V_L = liquid volume (m³)
ε_G = gas (air) holdup or volume fraction of air bubbles entrained in liquid (dimensionless)
ρ_L = density of liquid or solution (kg/m³)
ρ_G = density of air (kg/m³)
σ = interfacial or surface tension of liquid or solution (N/m)
µ_G = viscosity of air (kg/m/sec)
µ_L = viscosity of liquid or solution (kg/m/sec or cP)
µ_LK = viscosity of liquid at a known temperature T_K (cP)

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**Table 1.** Input parameters for the model.

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<th>Parameter</th>
<th>Value</th>
<th>Parameter</th>
<th>Value</th>
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<td>Initial NH$_3$ concentration</td>
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FIGURES CAPTIONS

Figure 1. Schematic diagram of a sparged agitated vessel.

Figure 2. Effect of air sparge rate on the rate of removal of NH$_3$ at 50 °C. Liquid batch size = 9,800 kg, initial NH$_3$ concentration = 0.415 wt %.

Figure 3. Effect of temperature on the rate of removal of NH$_3$. Liquid batch size = 9,800 kg, initial NH$_3$ concentration = 0.415 wt %.

Figure 4. Effect of liquid volume or batch size on the rate of removal of NH$_3$ at 50 °C. Initial NH$_3$ concentration = 0.415 wt %.

Figure 5. Comparison of model predictions with actual stripping operation data at 50 °C. Liquid batch size = 9,800 kg, initial NH$_3$ concentration = 0.415 wt %, air sparge rate = 170 scmhr (100 scfm).
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**Figure 5.** Comparison of model predictions with actual stripping operation data at 50 °C. Liquid batch size = 9,800 kg, initial NH₃ concentration = 0.415 wt %, air sparge rate = 170 scmhr (100 scfm).