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Kinetics of the Desorption of Ammonia from Water by Diffused Aeration

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Abstract

Ammonia desorption by diffused aeration was investigated in a lab-scale, batch aeration column, and the theoretical basis for the operation was developed. A formula was derived for the calculation of the fraction of ammonia present in the solution in undissociated form (susceptible for desorption) for any given temperature and pH. The experimental work shows that desorption is equilibrium controlled, which is at variance with some of the results reported in the literature. Lower limits of the ammonia mass transfer coefficient for bubbles rising through the water column were estimated. The impact of the surface effect on the rate of desorption was taken into account in the calculations.

INTRODUCTION

Conventional wastewater treatment plants have only limited capacity for nitrogen removal. Plant effluents thus contain large concentrations of nitrogen, with ammonia nitrogen being the main form. Ammonia nitrogen concentrations in biologically treated wastewater are generally in the range 12–35 mg/L (as N) (1). Several technically and economically feasible methods are available to reduce this concentration. They are characterized in Table 1.

The relative costs of the treatment processes listed in Table 1 are controversial, and some authors claim that ammonia desorption is by far the

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TABLE I
Comparison of Nitrogen Removal Processes

Process	Advantages	Disadvantages	Efficiency [2] (%)	Relative cost [1]
Biological nitrification-denitrification	N_2 end product All forms of nitrogen removed	Long detention time Sensitive to inhibitors changing flow, and temperature	70-95	1.00
Breakpoint chlorination	Complete removal can be obtained Temperature variation and inhibition proof	Chemicals added increase TDS greatly Dechlorination is often necessary Continuous control equipment required	90-100	—
Ammonia stripping	Simplicity of operation Ease of control No by-products	Scale formation Inefficient at low temperatures	50-95	1.32
Selective ion exchange on clinoptilolite	Temperature change and inhibition proof	By-product formation (brine)	90-97	1.54

least expensive nitrogen removal process (3). Low cost and other advantages of this process (Table 1) make it an attractive alternative, particularly where atmospheric conditions are favorable and when it is used in conjunction with high-lime phosphorus removal.

Desorption of ammonia from wastewater occurs when liquid of high pH is brought into contact with large amounts of air. There are several possible designs for providing air-water contact in order to desorb ammonia. These include:

1. Countercurrent or crosscurrent stripping towers with stationary bed
2. Stripping ponds (with and without agitation)
3. Spray towers
4. Diffused aeration
5. Three-phase fluidized bed

Considerable laboratory and pilot-scale data on the first three methods are available in the literature (2-8). The operation of stripping towers is particularly well documented, and these have found application in several full-scale water reclamation plants (9-11). The available literature data on ammonia desorption by diffused aeration are very limited. Some authors have stated that, due to the high solubility of ammonia, air bubbles leaving the water should be saturated with ammonia and that therefore the process is equilibrium controlled (12, 13). These observations were experimentally confirmed by Bennenworth and Morris (8). They were unable to detect any decrease in the rate of desorption with decreasing water levels for depths as little as 1.5 cm. However, their results were not conclusive (for discussion, see Ref. 14). Srinath and Loehr (15) investigated the effects of pH, temperature, viscosity, air flow rate, and initial ammonia concentration on the rate of ammonia removal by diffused aeration. In conclusion the authors stated that for design purposes the mass transfer coefficient for diffused aeration should be determined experimentally for a particular system or estimated from semiempirical relationships provided in the paper. Mass transfer limitations of the desorption process were also reported by Shpirt (16). He found that the mass transfer coefficient was a function of depth of submersion of the diffuser, turbulence, and size of the air bubbles. Specifically, the author found fine bubble aeration to be twice as efficient as coarse bubble aeration.

From the review presented it is apparent that opinions as to whether the process is equilibrium or mass transfer controlled are contradictory. The objective of the present study is to give insight into this problem.

THEORETICAL BASIS OF AMMONIA DESORPTION

In order to develop a mathematical formula for the rate of ammonia removal from a batch reactor, let us first consider the mass balance in one bubble rising through the liquid layer. It may be assumed that in the short time required for the bubble to rise through the water, the concentration of ammonia remains constant in the water and that it is uniform throughout the column. Change in the mass of ammonia in the bubble is described by

$$dm/d\tau' = F\alpha(c^* - c_g) = F\alpha(cKP - c_g) \quad (1)$$

where m = mass of ammonia in the bubble

τ' = time

F = surface area of the bubble

α = mass transfer coefficient (based on gas phase)

c_g = ammonia concentration in the bubble at time τ'

c^* = equilibrium concentration of ammonia in the bubble in contact with solution having ammonia concentration c

c = concentration of ammonia in the liquid ($\text{NH}_4^+ + \text{NH}_3$)

P = fraction of the ammonia present in the solution as NH_3

$K = c^*/cP$ = equilibrium constant

For a bubble of diameter d ,

$$dm = dc^* \frac{\pi d^3}{6}, \quad F = \pi d^2 \quad (2)$$

Substitution into Eq. (1) yields

$$\frac{dc_g}{cKP - c_g} = -\frac{6\alpha}{d} d\tau' \quad (3)$$

Integration from time 0 to τ and rearrangement gives

$$c'_g = cKP[1 - \exp(-a\tau)] \quad (4)$$

where c'_g = final concentration in the bubble as it leaves the water

τ = residence time of the bubble in the column

$a = 6\alpha/d$

The change of the ammonia concentration in the water is governed by the expression

$$-V \frac{dc}{dt} = c'_g Q \quad (5)$$

where V = reactor volume

Q = air flow rate

In examining the ammonia mass balance in the solution, ammonia desorbed from the free water surface should be taken into account. The change in ammonia concentration in the solution due to the ammonia desorption from the water surface can be described by

$$-V \frac{dc}{dt} = A \alpha' (c_g^* - c_g'') = c K P s (1 - f) \quad (6)$$

where c_g'' = ammonia concentration in the gas entering the space above the water surface

α' = mass transfer coefficient for the free water surface

A = surface of the water-air interface

$s = \alpha'^* A$

f = degree of saturation of the air entering the space over the water surface with respect to the bulk liquid ammonia concentration
($f = c_g''/c_g^*$)

The change in the mass of ammonia present in the solution due to the mass transfer from the bubble surface and from the free water surface is additive. Therefore, on combining Eqs. (4), (5), and (6) we obtain

$$-V \frac{dc}{dt} c K P [1 - \exp(-\alpha \tau)] Q + c K P s (1 - f) \quad (7)$$

Integration of Eq. (7) yields Eq. (8) for the rate of ammonia removal:

$$\ln \left(\frac{c_0}{c} \right) = \frac{Q t}{V} K P [1 - \exp(\alpha \tau) + s/Q(1 - f)] \quad (8)$$

where c_0 = initial ammonia concentration in the solution.

For the air in a bubble reaching the water surface, $c_g'' = c_g'$, and therefore from Eq. (4) we obtain

$$f = 1 - \exp(-\alpha \tau) \quad (9)$$

Substitution into Eq. (8) gives

$$\ln \left(\frac{c_0}{c} \right) = \frac{Qt}{V} KP [1 - (1 - s/Q) \exp(-\alpha\tau)] \quad (10)$$

In a given experiment Q , V , K , P , d , α , s and the water level (and thus τ) can be assumed to be constant. Therefore, a change in ammonia concentration expressed in the form $\ln (c_0/c)$ should be directly proportional to the air-to-water volume ratio, with a slope equal to

$$KP[1 - (1 - s/Q) \exp(-\alpha\tau)]$$

The value of the equilibrium constant K at 25°C is equal to 7.00×10^{-4} (17). Correlation of the data on partial pressure of ammonia above its solution in the temperature range 21–32°C [17] has given

$$p/p_0 = \exp[0.04473(t' - 25)] \quad (11)$$

where p = partial pressure of ammonia at temperature t' (°C)

p_0 = partial pressure of ammonia at reference temperature (25°C)

This formula was used in processing the experimental data to correct for the small temperature fluctuations around the designated operational temperature which occurred during our runs.

The fraction P of ammonia present as NH_3 molecules, and therefore susceptible to desorption, can be calculated from

$$P = \frac{[\text{NH}_3]}{[\text{NH}_4^+] + [\text{NH}_3]} = \frac{1}{\frac{K_l}{K_w} \times 10^{-\text{pH}} + 1} \quad (12)$$

where K_l is the dissociation constant of ammonia and K_w is the ion product for water. Inclusion of the temperature dependence of the constants K_l (18) and K_w (19) yields

$$P = \frac{1}{1 + 10^{10.06 - \text{pH} - 0.0327t'}} \quad (13)$$

where t' = temperature (°C).

This formula is valid for the temperature range encountered in wastewater treatment practice (5–25°C) and for any pH. The error of the formula is

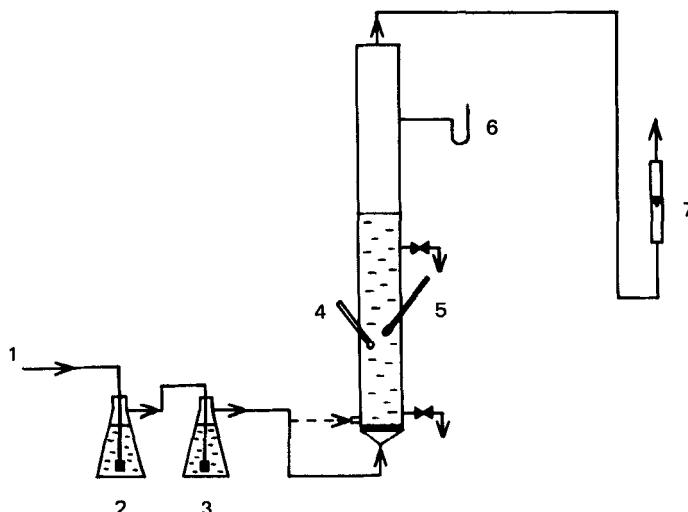


FIG. 1. Schematic of the experimental set-up: (1) house air, (2) scrubber with H_2SO_4 , (3) scrubber with NaOH , (4) pH electrode, (5) thermometer, (6) U-manometer, (7) rotameter.

<1.5% (of the value of P), which is accurate enough for all practical purposes. The precision of the calculations is limited by the accuracy of the pH measurements. For example, at $t' = 15^\circ\text{C}$ and pH 10.0 and 10.1, P equals 0.720 and 0.772, respectively. It should be emphasized that activities, rather than concentrations, should be used in these formulas, and the final formula is thus exact for dilute solutions with low ionic strengths.

EXPERIMENTAL

Experiments were carried out in a laboratory scale, batch desorption column at 25°C . The experimental apparatus is illustrated in Fig. 1. Test solutions for each run were prepared by adding NH_4Cl to deionized water and then increasing the pH to $11.65 + 0.05$ with 0.5 N NaOH solution. Air supplied to the column bottom was purified in two scrubbers which contained H_2SO_4 and NaOH solutions. During each experiment gas flow rate, ammonia concentration, temperature, pH, water level, and difference in pressure between column and atmosphere (resulting from head loss in tubing connecting the column and rotameter) were monitored and recorded. Experiments were stopped after a decrease in ammonia concentration to about one-third its initial value was obtained.

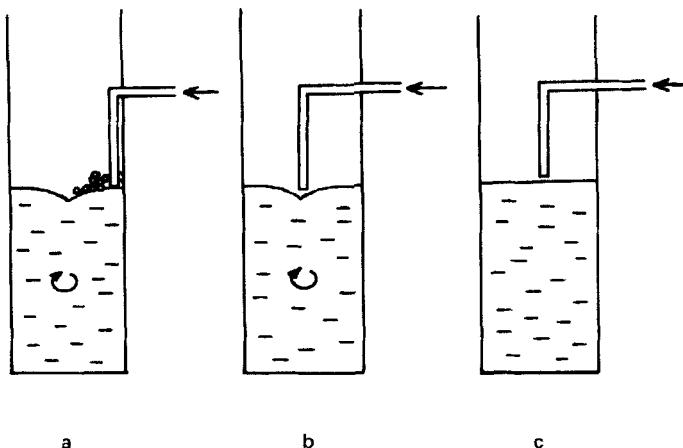


FIG. 2. Means of air-water contact in the experiments with surface aeration.

The effects of air flow rate, bubble size, and depth of submersion of the diffuser on rate of ammonia removal were investigated. Fine, medium, and coarse bubbles were applied with average diameters 0.32, 4.2, and 6.1 mm, respectively. Depth of diffuser submersion ranged from 1.6 to 47 cm, and air flow rate ranged from 0.27 to 1.1 L/min.

To determine the average bubble contact time, the level of water in the column was measured with and without air flow for different water levels and sizes of bubbles. From the results the average bubble contact time was calculated for each water level and bubble size. In the above described experiments the height of the water levels included the surface bubble layer and thus the calculated contact times include the time when the bubble was sitting on the water surface before breaking.

In three additional runs air was supplied to the column above the surface rather than from the submerged diffuser. In the first one air was released from a tube which touched the water surface, creating a bubble layer on the top of the liquid (Fig. 2a). In the second one the end of the tube was not in contact with water but the contents of the column were continuously mixed (Fig. 2b). In the third one air was released over a quiescent water surface (Fig. 2c).

By analogy with Eq. (10), experimental data for each run were correlated in the following form:

$$\ln \left(\frac{c_0}{c} \right) = b \left(\frac{Q}{V} t \right) \quad (14)$$

where b is constant for a given run.

The actual formula used for calculation of the air-to-water volume ratio Qt/V for each sampling interval i had the form

$$\frac{Q_i(t_i - t_{i-1})}{V_{i-1} - \Delta V} \frac{760 + \Delta p}{760} \exp [0.04473(t'_i - 25)] \quad (15)$$

which includes corrections due to

1. Changing volume of water in the column due to sampling, evaporation, and leaks (ΔV)
2. Differences in pressure between the top of the column and the atmosphere (Δp)
3. Small fluctuations of the water temperature t'_i during the course of the experiment

Air-to-water volume ratios for each time interval were calculated from Eq. (15) and subsequently added to give a cumulative air-to-water volume ratio Qt_i/V corresponding to decreasing ammonia concentration expressed as $\ln(c_0/c_i)$. Then the quantities Qt_i/V and $\ln(c_0/c_i)$ were correlated; values of the slope b obtained in this way for all the runs are shown in Figs. 3-6.

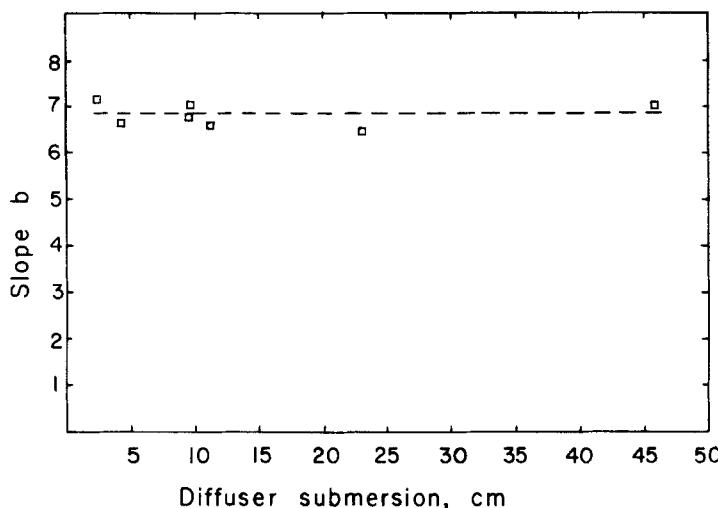


FIG. 3. Dependence of the slope of the correlation equation on the diffuser submersion. Fine bubbles, flow = 0.6 L/min.

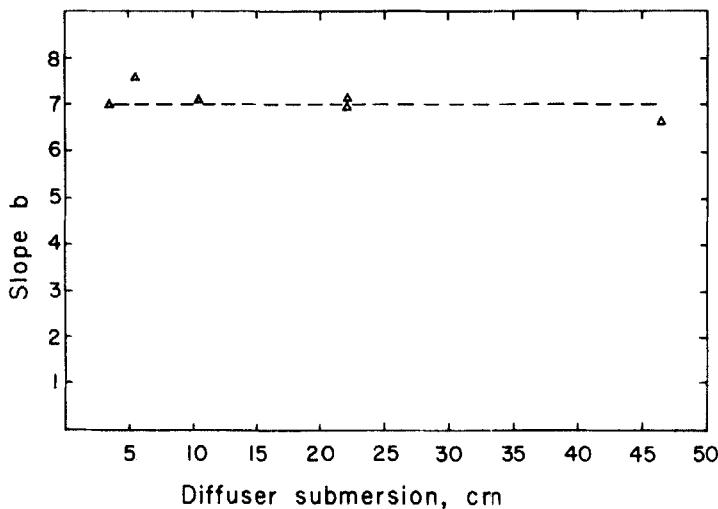


FIG. 4. Dependence of the slope of the correlation equation on the diffuser submersion. Medium bubbles, flow = 1.1 L/min.

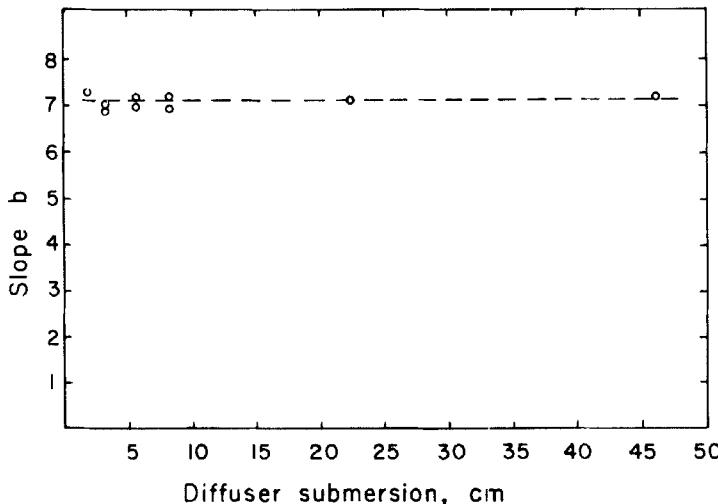


FIG. 5. Dependence of the slope of the correlation equation on the diffuser submersion. Coarse bubbles, flow = 1.1 L/min.

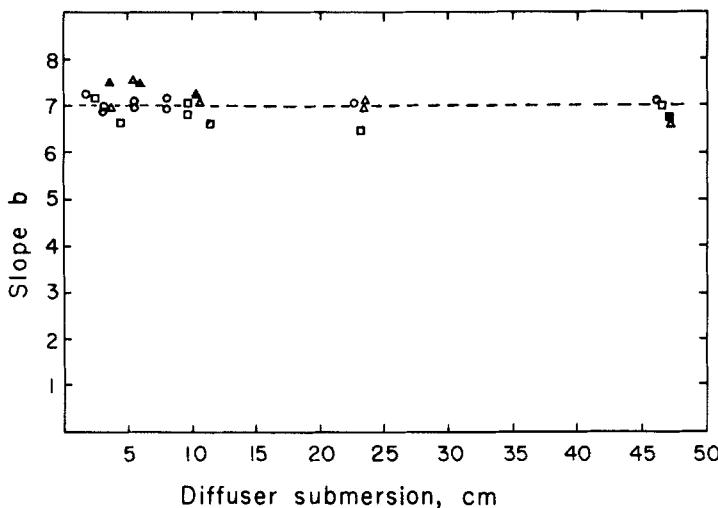


FIG. 6. Dependence of the slope of the correlation equation on the diffuser submersion. All runs: (□) fine bubbles, air flow = 0.60 L/min; (■) fine bubbles, air flow = 0.27 L/min; (△) medium bubbles, air flow = 1.1 L/min; (▲) coarse bubbles, air flow = 1.1 L/min; (○) coarse bubbles, air flow = 0.60 L/min.

DISCUSSION

Surface Effect

For the experiments with surface aeration only (without bubbling) $\tau = 0$ and $f = 0$. Therefore, from Eqs. (10) and (14) we get

$$s/Q = b/KP \quad (16)$$

Values of the term s/Q calculated for three different mixing conditions, as shown in Figs. 2a, b, and c, are 0.94, 0.77, and 0.62, respectively. This indicates, as expected, that s/Q is a function of the hydrodynamic conditions at the water-air interface. Comparison of these numbers with Eq. (10) indicates that, even for $\exp(-a\tau)$ approaching 1, this surface effect provides significant saturation of the air with ammonia (with fraction of saturation equal to s/Q).

Effect of Bubble Diameter and Contact Time on Rate of Ammonia Removal

According to Eqs. (10) and (14),

$$b = KP \left[1 - (1 - s/Q) \exp \left(-\frac{6\alpha}{d} \tau \right) \right] \quad (17)$$

The term in brackets represents the effect of the mass transfer rate on the rate of ammonia removal. For a given bubble diameter d , the mass transfer coefficient α can be assumed to be constant. For constant Q , hydrodynamic conditions over the water surface are similar and therefore s/Q can be assumed to be constant. Thus an increase in contact time τ should result in an increase of the slope b with its value reaching KP for sufficiently high values of τ .

Inspection of Figs. 3-5 indicates that there is no decrease in b with decreasing contact time (water level), even for bubbles with the largest diameter (Fig. 5), where mass transfer limitations should be most easily seen. Figure 6 indicates that bubble diameters have no impact on the rate of ammonia removal. Values of the slope b for all bubble sizes, diffuser submersion, and air flow rates are in the range 6.42×10^{-4} to 7.54×10^{-4} . Comparison of these numbers with the value of the equilibrium constant (7.00×10^{-4}) indicates that no significant mass-transfer effect can be noted. (The value of P at 25°C and $\text{pH} = 11.65$ is 0.996.) Differences in the values of b appear to be due to experimental error only. In order to verify this, a statistical analysis was performed.

According to Eq. (17), the dependence between b and α can be expected to be in the following linearized form:

$$\ln \left(\frac{KP}{KP - b} \right) = \frac{6\alpha}{d} \tau - (1 - s/Q) \quad (18)$$

If a correlation existed between mean bubble contact time τ and the term $\ln KP/(KP - b)$, the slope a ($a = -6\alpha/d$) of the correlation line would be statistically different from zero. For our calculations the value for KP has been taken as 7.3×10^{-4} , a value close to the upper limit of the experimental values. The results of the t -test indicate that the slope a is not significantly different from zero at the 90% confidence level. This means that changes in the contact time in the range investigated during the experiments have no discernible effect on the term $[1 - (1 - s/Q) \exp(-a\tau)]$, given the experimental precision achieved. So, even for coarse bubbles, after a very short

contact time corresponding to a water column height of only a few millimeters, the term $(1 - s/Q) \exp(-a\tau)$ is almost zero, and a further increase in contact time does not have a measurable impact on the slope b . The surface term $(1 - s/Q)$ has a damping effect on this result, diminishing it still further. From the results of the experiments on surface aeration, the value of s/Q can be assumed to be about 0.77 (continuous mixing with no bubble formation). Note that the value $s/Q = 0.94$ was obtained with bubble formation on the water surface, and therefore 0.94 is equal to the term $[1 - (1 - s/Q) \exp(-a\tau)]$, rather than to s/Q alone. With this assumption the results of the statistical analysis indicate that α is relatively large, so that even for the smallest contact times used, the term $(1 - s/Q) \exp(-(6\alpha/d)\tau)$ is already close to zero.

It should be emphasized that the results of the above statistical analysis indicate only that b is essentially independent of τ . Rejection of the hypothesis that b is a function of τ makes the assumption that $KP = 7.3 \times 10^{-4}$ erroneous since KP is then equal to the mean experimental value of b , rather than to the upper bound for it. It follows that Eq. (18) in the form presented cannot be the basis for any numerical estimation of α . Thus the statement that the slope a ($a = 6\alpha/d$) is not significantly different from zero is only in apparent contradiction to the conclusion that α is too big to be measured experimentally; this point is clearer if one looks at Eq. (17).

A practical conclusion is that for even coarse bubbles (6.2 mm) and minimal diffuser submersion, the rate of ammonia stripping is for all practical purposes entirely equilibrium and not mass transfer controlled. Air leaving the solution is saturated with ammonia. Therefore, in order to predict the rate of ammonia removal in a diffused aeration system (or to design it), it is enough to know the value of the equilibrium constant K and the fraction P of ammonia-nitrogen present as NH_3 (which is a function of pH and temperature).

This conclusion is at variance with some results reported in the literature (as discussed previously). In particular, findings by Shpirt (16) that a decrease in bubble diameter and an increase in diffuser submersion depth improve the process efficiency are not in agreement with our findings. Also, suggestions that for design purposes the mass transfer coefficient should be determined experimentally or estimated from semiempirical correlations (15) are at variance with the findings of the present study.

From the results presented in this study it is apparent that changes in the ammonia-nitrogen concentration ($\text{NH}_4^+ + \text{NH}_3$) in a diffused aeration system can be calculated from

$$c = c_0 \exp \left(-KP \frac{Qt}{V} \right) \quad (19)$$

for batch operation or

$$c = c_0 \exp \left(-KP \frac{Q}{Q_L} \right) \quad (20)$$

for continuous operation of a stirred tank system, where Q_L is the water flow rate. Values for the equilibrium constant K are available in many sources, while P can be readily calculated from Eq. (13).

Estimation of the Mass Transfer Coefficient

The lower limit of the mass transfer coefficient τ for a bubble rising through the water column can be estimated from the experimental data in this study.

Let us assume that a decrease in the slope b , due to mass transfer limitation, would be experimentally detected if its value was below the 95% confidence interval of the mean experimental value of b . Therefore

$$b_m = \bar{b} - \delta t_{95} \quad (21)$$

where b_m = maximum value of b experimentally detected as different from KP

δ = standard deviation of b

\bar{b} = mean experimental value of b

If we assume that KP is equal to the mean experimental value of b , Eq. (17) can be rearranged into the form

$$\alpha_m = \left[\ln \left(\frac{\bar{b}}{b - b_m} \right) + \ln (1 - s/Q) \right] \frac{d}{6\tau} \quad (22)$$

where α_m is a lower limit of α . For our calculations, s/Q was taken as 0.77 which was the value obtained from the experiment on desorption from the stirred water surface. The minimal values of α calculated from this equation are given in Table 2, Column 6. They should be considered rough estimates, due to the number of assumptions involved in the calculations. Actual values for α must be higher than that calculated if a decrease in slope b is not detected with the experimental techniques applied.

The limiting values of α , calculated in the above described manner, can be compared to the ones predicted theoretically. From the Lewis and Whitman

TABLE 2
Comparison of Theoretically Predicted and Obtained Values of Mass Transfer Coefficients

1	2	3	4	5	6
Bubbles size (mm)	k_g (cm/s)	k_L (cm/s)	Ratio of gas film to liquid film resistance	\propto Theoretically predicted (cm/s)	Minimum value of α obtained from experiments, α_m (cm/s)
Fine, 0.35	38.3	0.135	51	32.0	0.016
Medium, 4.2	3.19	0.039	177	3.17	0.271
Coarse, 6.1	2.20	0.032	210	2.19	0.249

two-film theory (20), the overall mass transfer coefficient α (gas phase based) is equal to

$$\frac{1}{\alpha} = \frac{K}{k_L} + \frac{1}{k_g}$$

where K = equilibrium constant

k_L = liquid phase mass transfer coefficient

k_g = gas phase mass transfer coefficient

Values of k_L and k_g for bubbles rising in the water can be calculated from the semiempirical formulas available in the literature (21, 22), generalized for any water-gas-air system. Detailed calculations can be found elsewhere (14), and the results are shown in Table 2.

Comparison of Columns 5 and 6 in Table 2 indicates that there is no contradiction between the values for the mass transfer coefficient calculated theoretically and the limits of α obtained experimentally.

In Column 4, Table 2, the ratio of the water-phase to the gas-phase resistance is given. This indicates that, even for the smallest bubbles used, virtually all resistance to mass flow is located in the gas phase. This is due to the high solubility of ammonia in water, which means that the mass of dissolved ammonia present in the bubble water boundary layer is adequate to saturate essentially a large volume of air without replenishment from the bulk solution. Therefore, to provide adequate mass transfer conditions, there is no need for an energy-consuming formation of a large air-water interfacial area.

From Eq. (8) it can be calculated that at 25°C approximately 3300 m³ air/m³ water is necessary to remove 90% of the ammonia, assuming no mass transfer limitations.

The above considerations indicate that constructions with very low resistance to air flow are preferred for ammonia desorption from water. Diffused aeration does not satisfy such requirements, and therefore its application to ammonia removal seems to have limited prospects.

CONCLUSIONS

1. Desorption of ammonia from water by diffused aeration is equilibrium controlled for all technically feasible bubble sizes and depths of submersion. The bubbles leaving the water were saturated with ammonia.
2. The depth of submersion of the diffuser and the bubble size have no impact on the efficiency of the process.
3. The rate of ammonia removal can be calculated readily from Eqs. (19) and (20). Thus, experimental investigations of the rate of ammonia removal and the derivation of empirical correlations for design purposes do not seem to be needed.
4. The percent of ammonia present in solution in undissociated form can be calculated from

$$P = \frac{1}{1 + 10^{10.06 - \text{pH} - 0.0327r'}}$$

5. Values for the mass transfer coefficient, as estimated from the experimental results, are not in conflict with the ones theoretically predicted.
6. Due to mass-balance and mass-transfer considerations, diffused aeration is not an economical process for ammonia removal. The large amounts of air required and rapid (gas-phase controlled) mass transfer rate indicate that apparatus with very low resistance to air flow will be more economical.

Acknowledgments

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