

Oxygen Diffusion in Wet Air Oxidation Processes

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Operations Research Branch
Report 72-4

Mathematics and Information Sciences Division

August 25, 1972



NAVAL RESEARCH LABORATORY
Washington, D.C.

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Abstract: The diffusion of dissolved oxygen is an essential step in wet air oxidation processes. This report is an investigation of the conditions under which this diffusion step becomes a limiting factor for such processes which use an air bubble column reactor for sewage treatment. The results show that the importance of oxygen diffusion as a rate-limiting step depends mainly on reaction temperature and pressure, chemical oxygen demand, bubble diameter, air supply rate, and reactor height. The implications of these results for shipboard waste treatment processes currently being considered by the Navy are examined.

PURPOSE AND SCOPE

A wet air oxidation process is being considered by the Navy for shipboard sewage treatment. The objective of this treatment is to reduce the chemical oxygen demand (COD) of sewage sludge generated aboard to an acceptable level. The wet air oxidation process accomplishes this reduction by combining the organic molecules of the sewage with oxygen dissolved in the sludge. This liquid phase oxidation occurs in a reactor at high temperature and pressure. Dissolved oxygen is supplied to the reaction by bubbling air through the sludge.

The movement of oxygen molecules by diffusion from inside the air bubbles to the organic molecules in the sludge is an essential step in the wet air oxidation process. It is the purpose of this report to find the conditions under which this diffusion becomes the rate-determining step, or limiting factor, in the process. This information is important for the proper design of a sewage treatment unit based on this process and for estimating the degree to which the efficiency of such a unit can be increased by an increased air supply rate or by agitation of the reactor contents.

The results obtained here show that the importance of oxygen diffusion as a rate-determining step depends primarily on reaction temperature and pressure, COD, air bubble diameter, air supply rate, and reactor height. These results are also consistent with empirical findings reported in a patent (1) related to this process. As was assumed in earlier analyses of their performance (2,3), oxygen diffusion does not appear to be an important constraint on the efficiency of the type of treatment unit contemplated for shipboard use, so long as air is supplied at an adequate rate. In some cases, however, it is probable that mechanical agitation or countercurrent sludge flow could significantly reduce the aeration rate required to maintain this efficiency.

THE DIFFUSION PROCESS

The sewage oxidation in this process occurs in a bubble column reactor, where air supplied by a compressor is bubbled through a sludge which is about 99% water. As a bubble rises through the sludge, oxygen diffuses from the bubble through the sludge. At the same time, dissolved oxygen is removed from the sludge by the wet air oxidation reaction.

This analysis proceeds by examining the rates of both oxygen consumption and diffusion. The effects of each air bubble are treated independently in this analysis because Yoshida and Akita (4) have shown

NRL Problem B01-14; Project SF35-432-012. This is an interim report; work is continuing on the problem. Manuscript submitted May 30, 1972.

experimentally that little error is introduced by this procedure if the ratio of gas volume to liquid volume in a bubble column is less than 1/10. This condition holds in most of the cases of interest here.

OXYGEN CONSUMPTION

Empirical patent data (1) indicate that the wet air oxidation process behaves approximately as a first-order reaction in COD, provided that dissolved oxygen is present in adequate concentration. That is, in a perfectly mixed-batch reactor,

$$\frac{dA}{dt} = -kA$$

where

A = COD value

k = reaction rate constant .

The value of k as a function of reaction temperature was estimated in a previous report (2), and is shown here in Fig. 1.

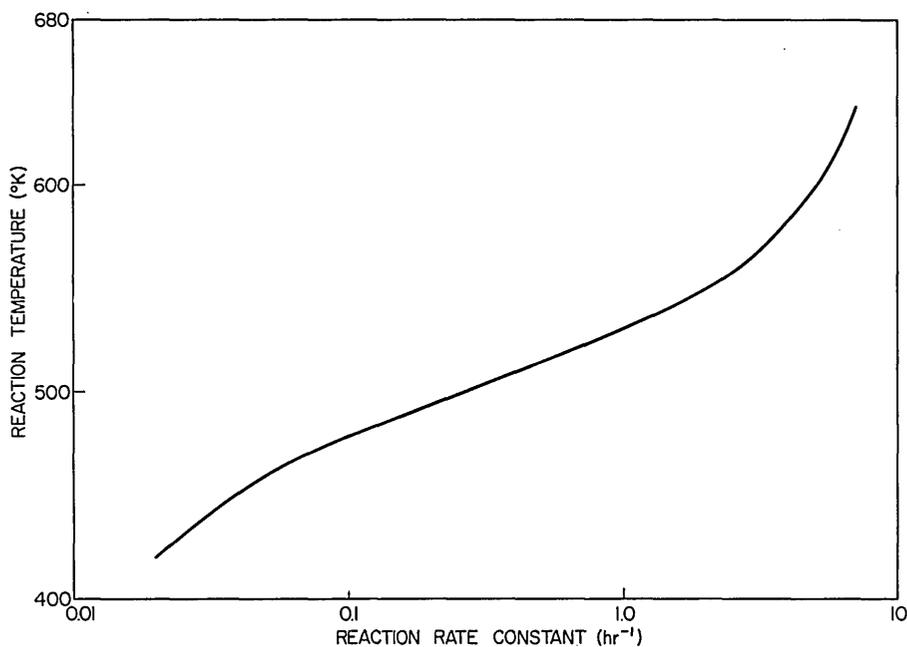


Fig. 1—Reaction rate

This behavior agrees with that predicted on the basis of the actual reaction mechanism by Emanuel, Denisov, and Maizus (5). They show that such first-order reaction kinetics should occur whenever

$$\frac{[O_2]}{A} > L(\theta),$$

where

$[O_2]$ = dissolved oxygen concentration

θ = reaction temperature,

and that the reaction rate should be significantly reduced by lowering the oxygen concentration only if this ratio is less than L . The function $L(\theta)$ is not known for the wet air oxidation reaction at hand, but can be estimated from patent data (1). These data indicate that, if $150^\circ\text{C} < \theta < 250^\circ\text{C}$ and $A = 100 \text{ g/l}$, an oxygen partial pressure of about 10 psia is sufficient to saturate the reaction with oxygen. From experimental solubility data of Himmelblau (6), this partial pressure implies that $L \approx 0.01$ in this temperature range. Since L is actually the ratio of two reaction step rate constants, and since many such rate constants exhibit similar temperature dependence, it will be assumed that $L \approx 0.01$ throughout the temperature range of interest here ($200^\circ\text{C} < \theta < 350^\circ\text{C}$). Thus, it is sufficient for the purpose of this report to find the conditions under which

$$L \geq 0.01 \quad (1)$$

throughout the entire reactor.

Since the overall rate of dissolved oxygen consumption W is the same as the overall rate of COD reduction, it follows that

$$W = - \int_{V_L} \frac{\partial A}{\partial t} dv, \quad (2)$$

where the integration is performed over the liquid portion of the reactor volume. If there is sufficient dissolved oxygen throughout the reactor for a first-order reaction, the integrand is given by the equation

$$\frac{\partial A}{\partial t} = D_s \nabla^2 A - \mathbf{u} \cdot \nabla A - kA, \quad (3)$$

where

D_s = diffusion coefficient for sludge molecules

\mathbf{u} = liquid flow field (vector).

This equation is derived in Kay (7). From Eq. (2), some vector calculus, and the fact that water is incompressible, it follows that

$$W = k \int_{V_L} A dv - D_s \int_{S_L} \nabla A \cdot d\mathbf{s} + \int_{S_L} A \mathbf{u} \cdot d\mathbf{s}, \quad (4)$$

where the last two terms are surface integrals over the liquid surface. These two terms represent the net efflux of COD from the reactor by diffusion and convection, respectively. Each is zero for a batch reactor and negative for a flow-through reactor, since COD is reduced in the reactor, not increased. Thus, the batch reaction is the most demanding of dissolved oxygen, and will be the case considered here for determining aeration requirements. For the batch reactor, the formula for oxygen consumption reduces to

$$W = k \bar{A} V_L, \quad (5)$$

where

\bar{A} = average COD in reactor

V_L = liquid volume in reactor.

The density of water is significantly reduced at the reaction conditions contemplated here. Therefore,

$$\bar{A} = \bar{A}_0 \frac{\rho}{\rho_0}, \quad (6)$$

where

\bar{A}_0 = COD at ordinary temperature, pressure

ρ_0 = ordinary water density (1 g/cm³)

ρ = water density at reactor conditions.

DISSOLVED OXYGEN GENERATION

Dissolved oxygen is supplied to the sludge by mass transport from the air bubbles. This type of mass transfer has received much experimental investigation. For a bubble with a completely mobile surface, Griffith (8) reports that the equation

$$\dot{m} = \pi D d(c - c_s) \left(2 + 1.13 \sqrt{\frac{ud}{D}} \right), \quad (7)$$

where

c = free stream dissolved gas concentration

c_s = saturation concentration of dissolved gas

D = gas diffusivity in liquid

d = bubble diameter

m = mass of gas in bubble

u = bubble velocity through liquid (scalar)

accurately describes gas transport if the dissolved gas does not react chemically, and if

$$0.6 \leq Sc \leq 3200$$

$$1.8 \leq Re \sqrt{Sc} \leq 600,000$$

where

$$Sc = \text{Schmidt number} \left(\frac{\mu}{\rho D g} \right)$$

$$Re = \text{Reynolds number} \left(\frac{d \mu \rho}{\mu} \right)$$

and where

$$\begin{aligned} g &= \text{effective gravity} \\ \mu &= \text{liquid viscosity} \\ \rho &= \text{liquid density.} \end{aligned}$$

Griffith (8) also reports that gas bubbles in water may develop rigid (i.e., noncirculating) surfaces if contaminants are present in the water. This rigidity changes the flow field near the bubble, and also the gas transfer rate. For this case, Treybal (9) recommends the use of the empirical formula

$$\dot{m} = \pi D d(c - c_s) \left[2 + 0.347 \left(ud \sqrt{\frac{\rho}{\eta D}} \right)^{0.62} \right] \quad (8)$$

for mass transfer without chemical reaction. The region of validity is the same as for the preceding formula. Since the effects of the water contaminants in sewage sludge are not known, the cases of both rigid and mobile bubble surfaces were considered in this study. The results for the two cases are fairly similar in this problem, however, so only the more likely case of mobile surfaces is carried on from this point.

If the dissolved gas is consumed by a chemical reaction, as it is here, the concentration field of the dissolved gas is altered. Since this alteration changes the rate of mass transfer from the bubbles, Eqs. (7) and (8) must be modified to account for this change. Johnson, Hamielec, and Houghton (10) have determined the degree of this change for second-order reactions by numerically integrating the Navier-Stokes equation describing the fluid motion near a bubble. If the data in the problem of interest here were from a second-order reaction, this computed change would be negligible for all the reaction conditions of interest. Since this change is even less pronounced in the type of reaction at hand, this effect need not be considered here as an important mechanism affecting dissolved oxygen distribution. Hence, it is also reasonable to assume that the dissolved oxygen concentration is approximately uniform throughout the reactor, except in a negligibly small portion immediately adjacent to the air bubbles, as long as the reactor column height is not too great.

As an air bubble rises through the sludge, it loses oxygen according to Eq. (7). From the experiments of Himmelblau (6), it is known that the saturation concentration of the dissolved oxygen $[O_2]_s$ is given by the equation

$$[O_2]_s = H(\theta)p, \quad (9)$$

where

$$p = \text{oxygen partial pressure}$$

$$H(\theta) = \text{Henry Law constant (shown for oxygen in Fig. 2).}$$

Since oxygen behaves approximately as an ideal gas (3) here, this partial pressure is proportional to the mass of the oxygen remaining in the bubble throughout its immersion in the sludge. Combining these facts with Eq. (7), it follows that

$$\dot{m} = \pi D d \left(2 + 1.13 \sqrt{\frac{ud}{D}} \right) \left([O_2] - \frac{Hp_0}{m_0} m \right), \quad (10)$$

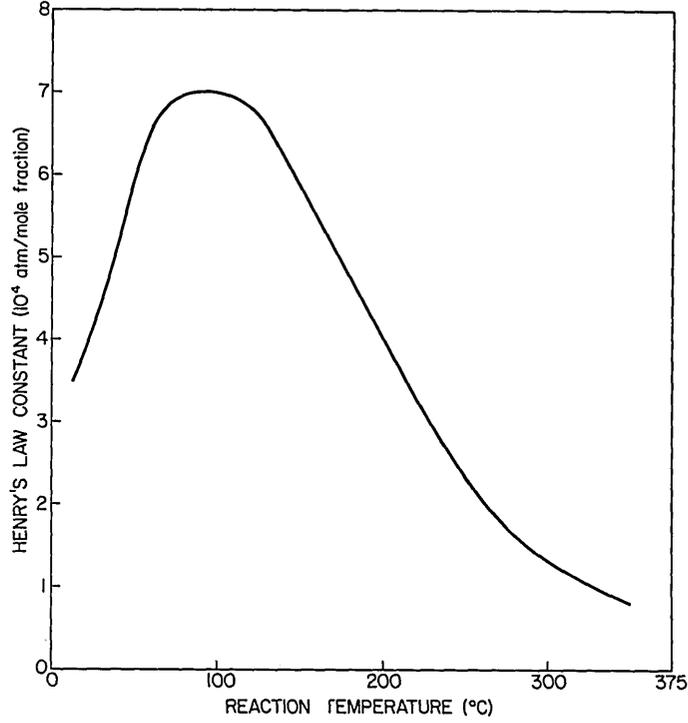


Fig. 2—Henry's Law constant (O_2 in H_2O)

where

m = current mass of oxygen in bubble (varies with time)

m_0 = initial mass of oxygen in bubble

p_0 = initial partial pressure of oxygen in bubble

$[O_2]$ = dissolved oxygen concentration in reactor.

Integrating Eq. (10) over the bubble's entrainment time gives

$$1 - \frac{m_f}{m_0} = \left\{ 1 - \frac{[O_2]}{Hp_0} \right\} \left\{ 1 - \exp \left[-\frac{\pi Hp_0 D d T}{m_0} \left(2 + 1.13 \sqrt{\frac{ud}{D}} \right) \right] \right\} \quad (11)$$

where

m_f = mass of oxygen in bubble when leaving sludge

T = bubble entrainment time.

The total mass rate of oxygen transfer from all the bubbles in the reactor is given by the equation

$$Z = \left(1 - \frac{m_f}{m_0} \right) Q, \quad (12)$$

where

Q = mass rate of oxygen bubbled into reactor

Z = total dissolved oxygen generation mass rate.

It is also easy to compute that

$$T = \frac{h}{u}, \quad (13)$$

and

$$m_0 = \frac{\sigma \pi d^3}{6} \quad (14)$$

where

σ = density of oxygen in incoming bubbles

h = reactor column height.

Combining Eqs. (11 through (14), the dissolved oxygen generation rate Z can be expressed as

$$Z = Q \left(1 - \frac{[O_2]}{Hp_0} \right) \left\{ 1 - \exp \left[\frac{6HhDp_0}{\sigma u d^2} \left(2 + 1.13 \sqrt{\frac{ud}{D}} \right) \right] \right\}. \quad (15)$$

To compute Z for a given set of reactor conditions from Eq. (15), it remains to determine several intermediate parameters.

Since air is approximately 20% oxygen, it can be concluded that

$$p_0 = \frac{P}{5}, \quad (16)$$

where

P = reaction pressure.

The reactor operates near the boiling point of water. Hence the vapor pressure curve for water, shown in Fig. 3, relates P and θ .

The inlet oxygen density can be found approximately from Eq. (16) and the ideal gas law, giving

$$\sigma = 5.3 \frac{P}{\theta} \frac{\text{g}^\circ\text{K}}{\text{l-psi}}. \quad (17)$$

Experimental data are not available for the diffusivity of oxygen in water at the high temperatures and pressures of interest here. However, Treybal (9) recommends a generalized formula for estimating this parameter, which in this case is

$$D = 7.24 \times 10^{-10} \frac{\theta}{\eta} \frac{\text{poise}}{^\circ\text{K}}, \quad (18)$$

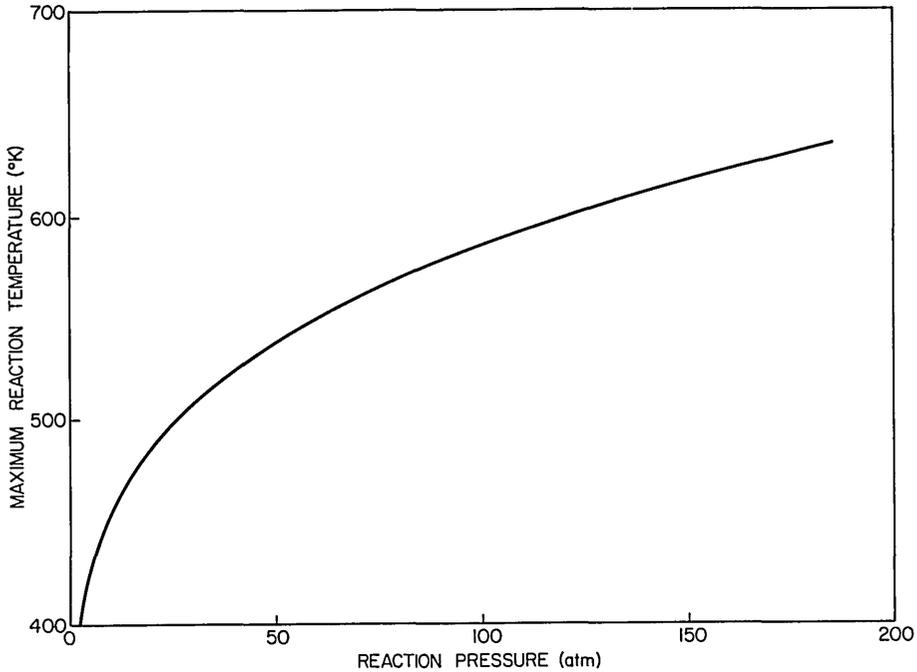


Fig. 3—Water boiling point constraint

where

η = water viscosity.

The viscosity of water was tabulated for these regions by Dorsey (11). At such high temperatures and pressures, it is only about 1/10 the viscosity of water under normal conditions.

For all of the reactor conditions of interest there, the liquid flow around the air bubbles is turbulent. An empirical formula for the velocity of bubble rise u has been developed by Harmathy (12) for turbulent flow. For the case of water, this formula is

$$u = 1.74 \sqrt{gd} f(N_{Eo}), \quad (19)$$

where

g = effective gravitational acceleration acting on bubble

$$N_{Eo} = g\rho d^2/\tau$$

τ = water surface tension

f = empirical function shown in Fig. 4.

The effective gravity is that which would produce the same buoyant force on the bubble if the bubble gas had zero density and the sludge had a density of 1 g/cm³. From hydrostatics and the ideal gas law,

$$g = 981 \left[\rho \left(\frac{\text{cm}^3}{\text{g}} \right) - 0.0266 \frac{P \left(\frac{^\circ\text{K}}{\text{psi}} \right)}{\theta} \right] \frac{\text{cm}}{\text{sec}^2} . \quad (21)$$

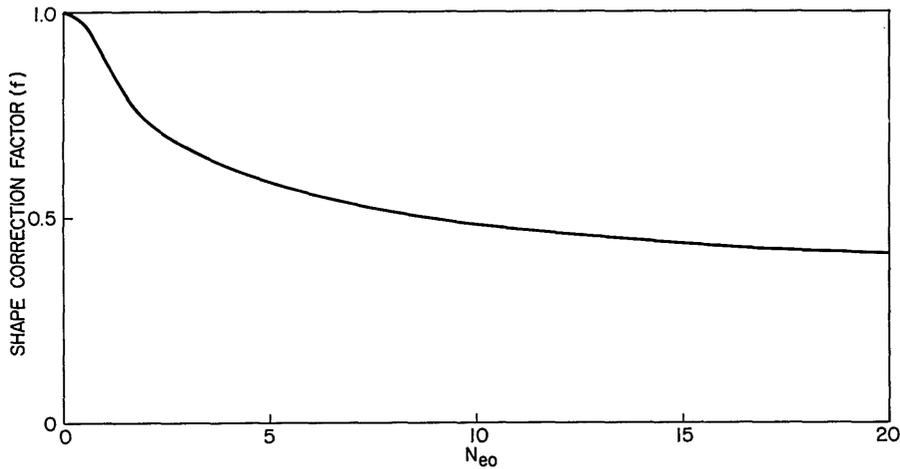


Fig. 4—Bubble velocity correction factor

The density of water ρ varies with temperature and is tabulated in Ref. 11.

Finally, it is necessary to evaluate the surface tension τ in order to find the value of N_{Eo} to use in Fig. 4. Experimental values are not available for the regions of interest here, but a generalized formula has been developed by Schonhorn (13), which in the case of water gives

$$\tau = 79.1 \exp\left(-\frac{0.00084 \text{ poise}}{\eta}\right).$$

AERATION REQUIREMENTS

At steady state operation, the generation and consumption rates of dissolved oxygen must be the same. Since air is 20% oxygen, it follows from Eqs. (1), (5), (16), and (17) that the required air supply per unit sludge in the reactor is

$$F = \frac{5Q}{V_L} = \frac{5k\bar{A}}{\left[1 - \frac{\bar{A}L}{H\bar{p}_0}\right] \left\{1 - \exp\left[\frac{-6H\bar{d}p_0}{\sigma u d^2} \left(2 + 1.13 \sqrt{\frac{ud}{D}}\right)h\right]\right\}}, \tag{23}$$

where F = required mass rate of air supplied to reactor per unit sludge volume in the reactor.

For a sufficiently high reactor column, this equation reduces to

$$F_\infty = \frac{5k\bar{A}}{1 - \frac{\bar{A}L}{H\bar{p}_0}} \tag{24}$$

where F_∞ denotes the value that F approaches as h becomes very large. Thus, the air supply requirement can be more conveniently summarized as

$$F = \frac{F_\infty(P)}{G(P, d, h)} \tag{25}$$

where

$$G = 1 - \exp \left[- \frac{6Hp_0D}{\sigma u d^2} \left(2 + 1.13 \sqrt{\frac{ud}{D}} \right) h \right]. \quad (26)$$

As indicated by the notation of Eq. (24), F_∞ depends only on reaction pressure, whereas G also depends on air bubble diameter and reactor column height. The factor G is the efficiency of air utilization, varying from 0 for very small reactor heights to 1 for very tall reactors.

To summarize, the air supply rate (per unit sludge volume) required to prevent the diffusion of dissolved oxygen from retarding the wet air oxidation process is computed from Eq. (25), with $L = 0.01$ for the cases of interest here. The quantities F_∞ and G are determined by Eqs. (24) and (26), and the results of the previous section. These results apply only to a single-unit bubble column reactor. If the reactor is actually a series of bubble columns, for example, the effective height for the purposes of this analysis is the sum of the individual column heights, assuming that COD is reduced more rapidly than p .

IMPLICATIONS FOR AGITATION

The results of this analysis have been computed for the cases of major interest here. The quantity F_∞ , the required air supply rate per unit sludge volume in a tall reactor, is displayed in Fig. 5 as a function of reaction temperature. This is the rate required to prevent oxygen diffusion from becoming a process bottleneck. Figure 6 shows a quantity called h^* , which is the reactor height at which the air utilization efficiency is 80%. An expression for this quantity can be derived from Eq. (26):

$$h^* = \frac{\sigma u d^2 \ln(5)}{6Hp_0D \left(2 + 1.13 \sqrt{\frac{ud}{D}} \right)}. \quad (27)$$

The significance of h^* is that less than a 20% increase in air utilization efficiency (a 20% reduction in required air supply rate) is obtained by any further increase in the effective reactor column height. A COD value of 3 g/l was assumed in these computations.

The region of anticipated shipboard operation shown in Fig. 6 applies only to designs using bubble column reactors without any agitation other than that induced by the buoyant motion of the entrained air bubbles. The range of bubble diameters shown is obtained from rough empirical observations of bubble behavior in water. The effective reactor column height range is inferred from the fact that the bubble columns in contemplated shipboard processes are limited to 8 ft in height and from proposals to use as many as four of these columns in series, which increases the effective height as explained earlier.

Additional agitation should not alter the effective reaction rate significantly (given that sufficient dissolved oxygen is present, of course) for the reaction conditions of interest here, since it has been ascertained earlier that the dissolved oxygen concentration is nearly uniform throughout the reactor. Hence the main effect of mechanical agitation or countercurrent sludge flow would be to increase the effective reactor column height. If this effective height already exceeds h^* , however, there is little to be gained from this effect either. If not, the augmented agitation could be used to reduce the rate of compressed air supply needed to maintain the wet air process at full speed.

From the computations shown in Fig. 6, it appears that there is a possibility in some cases of reducing the rate of compressed air supply by extra agitation of the reactor contents, if the reactor consists of a single bubble column. For a process using four reactor columns in series, however, the effective height would be around 30 ft without agitation; and it appears unlikely that such extra agitation would have any significant effect here.

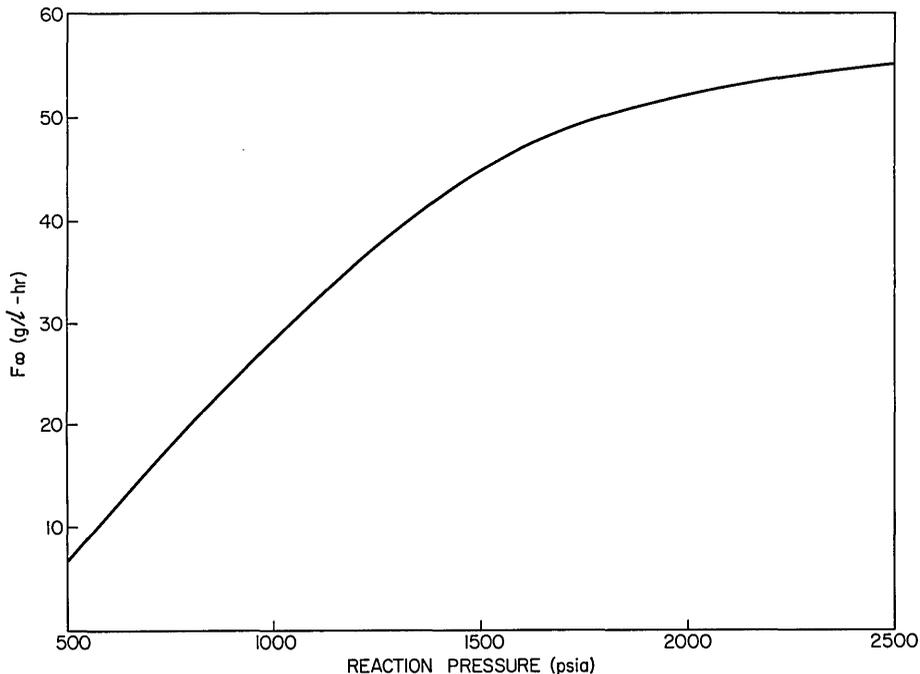


Fig. 5—Required air supply rate per unit sludge volume (tall reactors); initial COD = 3 g/l.

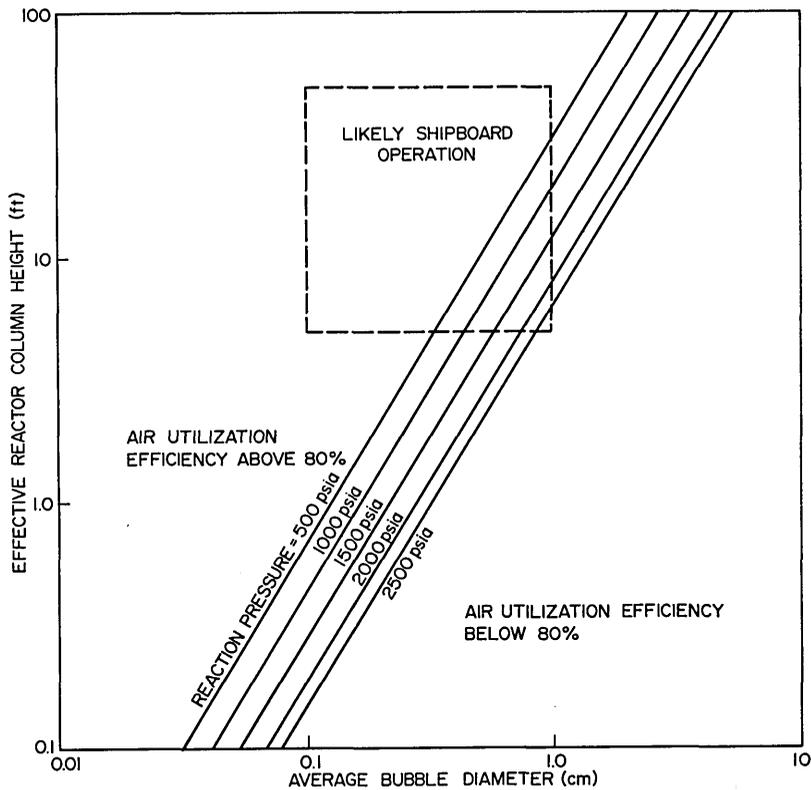


Fig. 6—Contours of 80% oxygen utilization; initial COD = 3 g/l.

It should be reiterated here that these computations and conclusions depend on reaction kinetics estimates of questionable accuracy. Experimental verification of either the kinetics or the resulting quantitative conclusions would be valuable.

The above computations apply to the case where the initial COD (i.e., \bar{A}_0) is 3 g/l, the intended value for initial shipboard use. Increasing this value by adding galley wastes or bilge to the sludge would increase the required air supply rate by increasing F_∞ . It would not affect the efficiency of air utilization or the value of h^* , however.

Some interesting results which are complementary to those reported here are presented in a doctoral thesis by Ploos van Amstel (14) on wet air sewage oxidation. His results are supported by systematic experimental work. They are limited to agitated autoclave reactors rather than bubble columns for the case of sewage sludge oxidation, however, and thus are not really comparable to the work reported here.

Finally, it is possible that a uniform concentration of dissolved oxygen would not be maintained without augmented agitation in reactors of sufficient height, contrary to the assumption made heretofore. The effect of this nonuniformity would be a reduction in the efficiency of air utilization. The most extreme case of this nature results from a complete lack of vertical motion of dissolved oxygen. A steady state analysis of this case shows that the effect of nonuniform oxygen distribution with respect to the preceding results is simply to multiply the values of h^* by a factor of

$$\frac{4}{\ln(5)} \approx 2.48.$$

The values of F_∞ are unchanged. For the reactor designs envisioned for shipboard sewage treatment, however, it seems likely that the agitation induced by air bubble motion would be sufficient to maintain the distribution of dissolved oxygen nearer the uniform case than this other extreme.

ACKNOWLEDGMENT

The author is grateful to Dr. Bruce Boss of the Chemistry Division for valuable advice concerning the reaction kinetics.

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Security Classification

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY <i>(Corporate author)</i> NAVAL RESEARCH LABORATORY Washington, D.C. 20390		2a. REPORT SECURITY CLASSIFICATION Unclassified	
		2b. GROUP -----	
3. REPORT TITLE OXYGEN DIFFUSION IN WET AIR OXIDATION PROCESSES			
4. DESCRIPTIVE NOTES <i>(Type of report and inclusive dates)</i> An interim report on a continuing problem.			
5. AUTHOR(S) <i>(First name, middle initial, last name)</i> W. W. Willman			
6. REPORT DATE August 25, 1972	7a. TOTAL NO. OF PAGES 15	7b. NO. OF REFS 14	
8a. CONTRACT OR GRANT NO. NRL Problem B01-14	9a. ORIGINATOR'S REPORT NUMBER(S) NRL Report 7443		
b. PROJECT NO. SF35-432-012	9b. OTHER REPORT NO(S) <i>(Any other numbers that may be assigned this report)</i> NRL Operations Research Branch Report 74-4		
c.			
d.			
10. DISTRIBUTION STATEMENT Approved for public release; distribution unlimited.			
11. SUPPLEMENTARY NOTES		12. SPONSORING MILITARY ACTIVITY Department of the Navy Naval Ship Systems Command Washington, D.C. 20360	
13. ABSTRACT The diffusion of dissolved oxygen is an essential step in wet air oxidation processes. This report is an investigation of the conditions under which this diffusion step becomes a limiting factor for such processes which use an air bubble column reactor for sewage treatment. The results show that the importance of oxygen diffusion as a rate-limiting step depends mainly on reaction temperature and pressure, chemical oxygen demand, bubble diameter, air supply rate, and reactor height. The implications of these results for ship-board waste treatment processes currently being considered by the Navy are examined.			

DD FORM 1473 (PAGE 1)
1 NOV 65

S/N 0101-807-6801

14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
<p>Wet air oxidation process</p> <p>Diffusion of dissolved oxygen in sewage</p> <p>Bubble column reactors for sewage treatment</p>						