EFFECT OF SURFACE ACTIVE AGENT ON OXYGEN ABSORPTION IN BUBBLE AERATION I.

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The effect of a surface active agent on mass transfer between phases implies a practical as well as theoretical problem which has caused wide interest among many workers \((1, 2, 3, 4, 5, 6, 7)\). The deterioration of oxygenation capacity of a specific plant in the biological treatment of domestic sewage due to the marked increase of household consumption of surfactants in recent years and the measures to improve the situation are being debated on a world-wide basis. On the other hand, the problem is intimately related to a subject, the discussions on which will eventually lead to a perusal of the current theories on mass transfer.

A considerable number of reports, especially during the past several years, have shown that surfactants adversely affect the rate of oxygen transfer in bubble aeration \((2, 3, 8)\). However, most of the papers reported so far on the adverse effect of surfactants describe the degree of deterioration in terms of liquid film coefficient which incorporates the effect of transient state in bubble formation and the effect of bubbling liquid surface. This sort of evaluation is likely to underestimate the adverse effect of the respective surface active agent on the liquid film coefficient of oxygen transfer. Furthermore, the mechanisms which have been presented by some workers to account for the increase of resistance to mass transfer in the presence of surfactants are considered to deserve further experimental verification and pertinent discussion.

Following the previous reports on oxygen transfer \((9, 10)\), the experimental determination of the effect of a surface active agent on the liquid film coefficient of oxygen transfer, excluding the ambiguous effects originating from bubbling liquid surface as well as from the bubble formation, will be described first. Secondly, by referring to certain model experiments, the mechanism to elucidate the increase of resistance to oxygen transfer will also be tentatively discussed in this paper.

Although the surface active agent employed in this series of experiments was only one type among the various kinds of surfactants now available on the market, the experimental results and discussions presented later will be of help for interpreting the phenomenon which is commonly associated with the use of surfactants in bubble aeration in the fermentation technology and biological treatment of sewage and industrial wastes.
MATERIALS AND METHODS

(a) The aeration column employed in this study is shown in Fig. 1. At the bottom of the column of pyrex glass (inner diameter and height of ca. 30 and 1,000 mm, respectively), a glass nozzle, about 3.5 mm in internal diameter was fixed. Its opening was directed towards the bottom as shown in Fig. 1 to facilitate a constant rate of bubble formation.

The air flow rate measured with an orifice flow meter ranged from 40 to 70 cm$^3$/min depending on the liquid level in each experiment; namely, when a high liquid level was used, a lower value of the aeration rate was employed to keep the ascending pattern of air bubbles in the column as constant (refer to Table 1 later). The pattern was changed appreciably, if aerated otherwise.

The bubbles generated were assumed to be oblate. In order to calculate the surface area and volume of these bubbles, the major and minor axes of each bubble were determined by taking photographs after immersing the aeration column into another rectangular glass vessel filled with water. The equivalent diameter of the bubbles tested in this study was about 8 mm (see also Table 1).

The liquid used was a 1% NaCl aqueous solution. The surface active agent used was sodium lauryl sulfate which was added to the solution at concentrations from 3 to 200 ppm.

The solution, without the surfactant, placed in the aeration column to each predetermined level was gassed with nitrogen gas for a few min to purge dissolved oxygen from the solution. After adding the surfactant solution, the aeration was started and the course of increase of dissolved oxygen concentration in the solution with the time of aeration was determined by sampling the solution intermittently and measuring the dissolved oxygen concentration polarographically.

The potential of dropping mercury was regulated from $-0.45$ V (for the...
case without the surfactant) to $-1.6$ V (for the case with the surfactant).

From the calibration test which was conducted with solutions of known dissolved oxygen concentration, the diffusion current of polarographic circuit was ascertained to be proportional to the concentration of dissolved oxygen in respective cases.

Regarding each concentration of the surface active agent, experiments were repeated several times varying the liquid level from 110 to 850 mm. Although the sampling taps were fixed at both the bottom and side (around one-half of the column height) of the aeration column as shown in Fig. 1, the preliminary experiments confirmed that the effect of the sampling position on the final data of liquid film coefficient was negligible.

All of the experiments were conducted at room temperature (18—23°). The surface tension of each solution with or without the surfactant was measured by the Du Noüy ring method.

(b) A number of glass columns, 10 in all, each 30 and 450 mm in internal diameter and height, respectively were immersed in a water bath (20°). Half of them were filled with 1% NaCl solution, while the other half were filled with sodium lauryl sulfate solution ranging in concentration from 30 to 100 ppm. By purging dissolved oxygen from respective solutions by blowing nitrogen gas prior to the experiment and at a certain time after the start of each run, two glass columns, one containing 1% NaCl solution and the other the surface active agent, were removed from the group of glass columns immersed in the water bath. After mixing the solutions gently to insure uniformity of dissolved oxygen, the concentrations were determined polarographically. After another time interval, two more glass columns were taken out of the remaining group (eight in number) in the water bath to measure the dissolved oxygen concentration. With this procedure, the relationships between the dissolved oxygen concentration and the time were determined.

(c) Since the value of liquid film coefficient of oxygen transfer may largely depend on the dynamic behavior of air-liquid interface, another series of experiments was conducted at room temperature. A number of beakers (each 75 mm i. d. and 75 mm in height) were partly filled with deoxygenated aqueous solution of 1% NaCl (300 cm³, each). The patterns of liquid free surfaces (surface area = 44 cm²), through which oxygen transfers from the ambient atmosphere to each solution were altered by the following experimental techniques.

(i) Standstill

(ii) Beakers were placed on a steel plate, under which a piece of magnet (span: about 40 mm) was rotated at about 3,700 rpm, causing a slight vibration of the liquid free surface.

(iii) A small rod of steel (about 20 mm in length and 4 mm in dia.) was rotated in the magnetic field (about 1,000 rpm) at the bottom of each
beaker. Another rod of glass (sectional area: 2 cm²) was inserted centrally in the beaker to extinguish a vortex which could otherwise be formed at the liquid surface.

The surface area for oxygen transfer was assumed to be (44-2=) 42 cm² in this case.

(iv) Together with the above-mentioned magnetic agitation, the liquid surface was stirred with a sort of circular brush, made of copper screen (about 10 mesh) and attached to an agitator shaft. The brush, about 50 mm in span, was rotated at a rate of 350 rpm.

Under each condition of the surface patterns, the oxygenation rate was measured polarographically by sampling intermittently each solution with a hypodermic syringe and the liquid film coefficient was calculated accordingly. Since the dissolved oxygen concentrations measured with samplings just below and far from the quiet liquid surface, respectively (as for the case (i) referred to above) did not differ appreciably, the dissolved oxygen was considered to have been uniformly distributed throughout the liquid for each condition of the liquid free surface mentioned above.

RESULTS AND DISCUSSIONS

The experimental results obtained are summarized in Table 1.

Before discussing the effect of the surfactant on the liquid film coefficient of oxygen transfer, a brief explanation of some of the columns in Table 1 will be made.

The mean surface area $\bar{S}$ and volume $\bar{V}$ are the arithmetical averages using photographs of bubbles from 6 to about 20 in number. Since the bubbles were assumed to be oblate in shape, identical values of $\bar{V}$ do not necessarily correspond to the same value of $\bar{S}$. Furthermore, the fact that the identical value of $\bar{V}$ does not always result in the same value of $\bar{S}$ in Table 1 is accounted for by a specific distribution of bubble sizes in question.

The term $N$ in the table indicates the number of bubbles present in the aeration column at any instant during the steady state.

When the liquid level was decreased, the average number of $N$ was determined by dividing the liquid level by an average of intra air bubble distance, otherwise an appreciable error in assessing the air bubble just emerging out of the orifice could not have been avoided.

The interfacial area $A_0$ between air bubbles and liquid was obtained by multiplying each value of $\bar{S}$ by respective data of $N$. The relationship between $A_0$ and $H$, thus obtained is plotted in Fig. 2. Although the points are rather scattered as the liquid level $H$ increases primarily due to both different values of $V$ adopted and the presence of the surfactant, a linear relationship which could be a proof of homogeneity of bubble ascent.
Table 1. Experimental results.

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in the aeration column could be assumed, at any rate, between \( A_0 \) and \( H \).

Note that in the same figure the aeration in this series of experiments was conducted in such a way that the bubble size was nearly constant irrespective of the presence of the surfactant.

The term \( KA \), which was determined by the procedure (9) previously reported using data on the course of dissolved oxygen concentration with the time of aeration, is plotted against \( H \) in Fig. 3.

Although a linear relationship between \( KA \) and \( H \) was assumed in the previous report (9) covering a short range of \( H \), it is not possible to assume, per se, the linearity between \( KA \) and \( H \) in the case of Fig. 3. It may be assumed that the term \( KA \) is composed of two other terms of \( k_{LA_0} \) and \( k_{L'A_0'} \), the latter corresponding to the end effect as well as the effect of transient state associated with the bubble formation at the nozzle.

The preliminary experiments confirmed that the end effect, which was estimated by blowing nitrogen gas into the solution within the aeration column, allowing only the ambient oxygen to transfer from the liquid free surface into the solution and measuring the rate of dissolved oxygen balance with time, was of the order of 0.02 cm\(^3\)/sec in terms of \( k_{L'A_0'} \).

Therefore, it may be assumed in Fig. 3 that the term \( k_{L'A_0'} \) which should contribute to a part of the term \( KA \) is composed predominantly of the effect of the transient state of bubble formation.

A schematic representation of the relationship between \( KA \) and \( H \) is shown in Fig. 4. The upper part (I) of the figure assumes that the transient effect is concentrated around \( H=0 \), while the lower part (II) assumes an
“age” of the effect in terms of \( k_L \). The former pattern relating to \( KA \) vs. \( H \) was assumed in the previous report. (9).

However, with reference to the experimental results in Fig. 3 in which a sigmate pattern could partially be observed, it is considered that the latter case (II) in Fig. 4 may also be realized when the liquid level \( H \) is increased to an extent as shown in Fig. 3.

Consequently, in this paper, the liquid film coefficient \( k_L \) was calculated first by assuming approximately the linear relationship between \( KA \) and \( H \) and secondly, by assuming that the sigmate figure relating to \( KA \) vs. \( H \) terminates when \( H \) exceeds 70 cm. In other words, the first procedure for evaluating \( k_L \) corresponds to assuming the upper pattern (I) in Fig. 4, while the second procedure leading to the determination of \( k_L \) corresponds to the lower pattern (II) in the same figure.

The value \( k_L \), plotted against the surfactant concentration \( C_s \) is shown in Fig. 5. The short bars indicate the estimated range of \( k_L \) from the first procedure. The short bars in broken lines refer to \( k_L \) from the second procedure. The ranges of the values of \( k_L \) shown in the figure depend mainly on the scattering of the data points in Fig. 2. It is noted that the first procedure tends to overestimate the transient effect in this case. However, one may argue that both patterns make no significant difference in assessing the magnitude of the transient effect.

The experimental result on the relationship between the surface tension \( \sigma \) of each solution and the surfactant concentration \( C_s \) is also included in Fig. 5. It is interesting to note that the value of \( k_L \) from the figure sharply decreases to a certain value (about one-fourth of that which could be obtained when \( C_s=0 \)) with the addition of the surfactant to a small extent which is less than the so-called critical micell concentration (about 100 ppm in
Fig. 5. Effect of surfactant concentration $C_s$ on liquid film coefficient $k_e$. 

Fig. 4. Schematic representation of $K_A$ vs. $H$. 

Table: Surfactant conc. (ppm) 

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<th>Symbol</th>
<th>0 ppm</th>
<th>3 ppm</th>
<th>6 ppm</th>
<th>25 ppm</th>
<th>100 ppm</th>
<th>200 ppm</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.04</td>
<td>0.03</td>
<td>0.02</td>
<td>0.01</td>
<td>0.00</td>
<td>0.00</td>
</tr>
</tbody>
</table>

Graphs showing relationships between variables.
Although many papers pointing out the mechanism of the adverse effect of a specific surfactant on the mass transfer rate have been published (1, 6, 11), a reconsideration of the mechanism from another angle in the case of bubble aeration seemed significant.

The experimental result on the oxygen transfer rate from the liquid surface at standstill, the experimental procedure of which was described in (b) of the previous section is shown in Fig. 6. The ordinate and abscissa are the saturation deficit and the time of exposure, respectively. The slope of a straight line, if drawn through each set of points in the same figure, is proportional to the value of $k_L$. It is apparent from the figure that the increase, if any, of diffusional resistance to oxygen transfer under the influence of the surface active agent is not revealed. Consequently, the degree of decrease in terms of $k_L$ which is manifested in Fig. 5 cannot evidently be accounted for exclusively by the possibility of increased diffusional resistance caused by the adsorption of the surfactant at air-liquid interfaces.

A spectrum of $k_L$ which was obtained by the procedure mentioned earlier is shown in Fig. 7. Clearly the value of $k_L$ largely depends on the dynamic behavior of the surface in question. Since the value of $k_L$ in

\begin{table}[h]
\centering
\begin{tabular}{|c|c|}
\hline
Symbol & Surfactant conc. \\
\hline
\cd & 0 ppm \\
\oa & 30 \\
\da & 100 \\
\hline
\end{tabular}
\end{table}

Fig. 6. Effect of surfactant on oxygen transfer rate relating to liquid surface at standstill.

Fig. 7. Spectrum of $k_L$. 

this example).
bubble aeration without the surfactant was around 0.04 cm/sec in this specific case ($\bar{\theta} \approx 0.8$ cm) and furthermore, since the coefficient decreased to about one-fourth of the former value in the presence of surfactant, we concluded, though qualitatively, that the surfactant mainly exerted a sort of calming action of the complicated motions at air-liquid interface, resulting in the decrease of liquid film coefficient.

It may be inferred that the extent of the calming action could be found, for instance, midway between the magnetic stirring with and without the surface agitation as specified in a previous section (c) of this paper.

**SUMMARY**

Using sodium lauryl sulfate, the effect of a surface active agent on oxygen transfer rate in bubble aeration was studied experimentally. It was found that the value of liquid film coefficient in bubble aeration was reduced by the addition of small amounts of sodium lauryl sulfate, less than 100 ppm, to about one-fourth of that obtained in the case without the surfactant.

This degree of increase of resistance to oxygen transfer was not ascribed directly to an increase of diffusional resistance due to the adsorption of the substance at the air-liquid interface, but principally to a sort of calming action of sodium lauryl sulfate on the oscillation as well as the other complicated motions of bubble surfaces in aeration.

**NOMENCLATURE**

$A_o$: interfacial area between air bubbles and liquid (cm$^2$)
$A_o'$: equivalent interfacial area (cm$^2$)
$C$: oxygen concentration (ppm)
$C_o$: initial oxygen concentration (ppm)
$C_s$: surfactant concentration (ppm)
$C^*$: equilibrium concentration of oxygen at interface (ppm)
$H$: liquid depth (cm)
$KA$: $k_L A_o + k_L' A_o'$ (cm$^3$/sec)
$k_L$: mass transfer coefficient of liquid film (cm/sec)
$k_L'$: equivalent mass transfer coefficient (cm/sec)
$N$: number of bubbles suspended in aeration column
$S$: surface area of one bubble (cm$^2$)
$t$: time of exposure (hr)
$V$: air flow rate (cm$^3$/min)
$V_o$: liquid volume (cm$^3$)
$V_b$: volume of one bubble (cm$^3$)
$\bar{\theta}$: equivalent bubble diameter (cm)
$\sigma$: surface tension (dyne/cm)

———: arithmetical mean
REFERENCES