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A TRANSIENT RESPONSE METHOD FOR A SIMPLE EVALUATION OF MASS TRANSFER IN LIQUIDS AND DISPERSIONS

EXPERIMENTAL TEST OF A MATHEMATICAL MODEL FOR AN ENSEMBLE OF BUBBLES

B. GAL-OR[†], **J. P. HAUCK**[‡] and **H. E. HOELSCHER**[†]
The Johns Hopkins University, Baltimore, Maryland

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Abstract—A transient response method has been developed for simple and fast physical measurement of concentration and volumetric mass-transfer coefficients in gas–liquid dispersions. The method involves the use of a transient response in the form of a step change in the composition of the feed gas. The resulting change in the composition of the liquid phase of the dispersion is measured by means of a Clark Electrode [26] which permits rapid and accurate analysis of oxygen and carbon dioxide concentrations in gas, blood or any liquid mixture. Experimental volumetric mass-transfer coefficients are calculated from

$$k_L a = \frac{\mathrm{d}\xi/\mathrm{d}\theta\,(\theta)}{\xi_{\mathrm{sat}} - \xi(\theta)}$$

where ξ is the electrode output (or recorder reading in any arbitrary units) and ξ_{sat} is the final reading of the electrode at saturation.

The method was applied to test a simple mathematical model that permits the estimation of $k_L a$ values for an ensemble of bubbles. Simultaneous measurements of gas holdup fractions were also made since knowledge of average bubble residence time and gas holdup is necessary to predict $k_L a$ values with the aid of this model. The mathematical model predicts lower $k_L a$ values than the observed ones but in general gives the lower bound for the behavior of the dispersion as a function of the operating variables.

NOMENCLATURE

a,	surface area of the bubbles per unit volume of clear liquid;
a^*,	surface area of the bubbles per unit volume of dispersion;
a_v,	mean volume radius, equation (11);
A,	total bubble surface area in the dispersion;
c,	concentration of dissolved gas in liquid;
c_0,	average concentration in the liquid bulk;
c_i,	concentration at interface;
C_I,	concentration in entering liquid phase;
d,	bubble diameter;
D,	diffusivity;
D_s,	surface diffusion coefficient of surfactants;
F,	volumetric liquid flow rate;
h_L,	liquid height without gas;
$\bar{h}_d$,	average dispersion height;
k_L,	mass-transfer coefficient, liquid film;
$k_L a$,	volumetric mass-transfer coefficient;
$\overline{N}_T$,	total average mass-transfer rate in the disperser;
Q,	volumetric flow rate of dispersed phase;
Q^*,	flux of absorbed surfactants;
P^*,	flux of desorbed surfactants;
R,	gas constant;
T,	absolute temperature;
U,	magnitude of bubble velocity relative to the liquid;

[†] University of Pittsburgh, Pittsburgh, Pennsylvania.
[‡] 401 Douglas Street, Clifton Forge, Virginia.

V,	liquid volume in the vessel.

Greek symbols

γ,	retardation coefficient of surfactants;
Γ,	surface concentration of surfactants;
Γ_0,	equilibrium concentration of surfactants on the interface;
$\bar{\delta}$,	average thickness of diffusion boundary layer;
θ,	transient response time;
$\bar{\theta}$,	average residence time of the bubbles in the dispersion;
μ,	viscosity;
ρ,	density;
σ,	surface tension;
Φ,	fractional gas hold-up;
ξ,	electrode output or recorder reading.

Subscripts

c,	refers to the continuous phase;
d,	refers to the dispersed phase.

INTRODUCTION

EXPERIMENTAL evaluations of volumetric mass-transfer coefficients and efficiency of contacting bubbles with liquids in various dispersers are usually determined by measuring absorption rates of a pure gas or of a component of a gas mixture as a function of the operating variables involved. The basic requirement of these methods is that the rate step for the physical absorption should be controlling and not, for example, the chemical reaction rate. The experimental method which has gained the widest acceptance involves the oxidation of sodium sulfite [4–13, 30]. It is usually regarded as the standard method for testing absorption rates of oxygen in gas–liquid dispersions and in fermentation systems [7, 30]. In some of the more recent studies, measurement of the absorption rate of carbon dioxide in aqueous solutions of sodium hydroxide or monoethanolamine solutions has been used extensively [14–25]. Disadvantages of these methods are their complexity, uncertainties [5, 6, 18, 23, 24, 30] and time consuming chemical analyses.

In the present study a simple and fast physical method that directly gives concentrations and k_La values in gas–liquid dispersions is developed. The method involves the use of a transient response due to a step change in the composition of the feed gas. The resulting change in the composition of the liquid phase of the dispersion is measured by means of a Clark Electrode [26–28]. This electrode permits rapid and accurate analysis of oxygen [26] and carbon dioxide [27, 28] concentrations in gas, blood or any liquid mixture [27].

THE PROPOSED METHOD

The volumetric mass-transfer coefficient with respect to unit volume of clear liquid, may be calculated from

$$k_La = \frac{[\mathrm{d}c/\mathrm{d}\theta]\,(\theta)}{C_{\mathrm{sat}} - C(\theta)} \tag{1}$$

where k_L is the liquid-mass-transfer coefficient, a, the surface area of the bubbles per unit volume of clear liquid, $[\mathrm{d}c/\mathrm{d}\theta]\,(\theta)$ the instantaneous slope of the concentration response curve, C_{sat} the final saturated concentration for a given step change in the composition of the feed gas and $C(\theta)$ is the instantaneous bulk concentration of the liquid phase.

Here it is assumed that the final saturated concentration for a given step change, C_{sat}, is also the interfacial concentration (which is practically constant for sparingly soluble gases [31] such as O_2 and CO_2). Hence $C_{\mathrm{sat}} - C(\theta)$ is the driving force for diffusion. This driving force as well as the rate $(\mathrm{d}c/\mathrm{d}\theta)$ are functions of θ—the time since the transient response has been introduced. If the change in composition of the gas is made without changing the gas flow rate and agitation intensity of the dispersion, A and k_L should remain time invariants.

It is assumed here that a relatively small change in the composition of the feed gas will produce a negligible change in the physical properties of the gas–liquid interface, however the time lags of the system and the measuring devices must be accounted for.

Hence at any

$$\theta > (\theta_L^s + \theta_L^M) \qquad (2)$$

where θ_L^s and θ_L^M are the system and measuring devices time lags respectively, the experimental values of k_La should be approximately time invariant if these assumptions are valid. It should be noted that θ_L^s includes the time lag necessary to replace all "old" bubbles in dispersion with "fresh" ones. Since full establishment of this condition may take considerable response time, we expect the time invariant condition for k_La values to be approached asymptotically at

$$\theta \gg (\theta_L^s + \theta_L^M). \qquad (2a)$$

Therefore, any measuring device (with small θ_L^s value) which records a response curve proportional in magnitude to the actual concentration response curve may be employed to evaluate k_La values from (1). Thus, if

$$\xi \propto C \qquad (3)$$

where ξ is the device output or recorder reading in any arbitrary units, then substituting (3) in (1) gives

$$k_La = \frac{\mathrm{d}\xi/\mathrm{d}\theta\,(\theta)}{\xi_{\mathrm{sat}} - \xi(\theta)} \qquad (4)$$

since any conversion factor is cancelled out (i.e. calibration becomes an unnecessary procedure).

When the volumetric mass-transfer coefficient is expressed in terms of interfacial area per unit volume of dispersion $-a^*$, the results can be calculated from

$$k_La^* = \frac{\mathrm{d}\xi/\mathrm{d}\theta\,(\theta)}{\xi_{\mathrm{sat}} - \xi(\theta)}(1 - \Phi). \qquad (4a)$$

For a semi-batch system, however, it is preferable to express the results in terms of k_La since the liquid volume rather than the dispersion volume is the quantity known for design purposes.

THE CLARK ELECTRODE

The Clark Oxygen Electrode [27], is a polarographic cell, consisting of a platinum electrode and a silver anode which are electrically connected by a medium containing chloride ions. When a potential of 0·6 V is applied to the cell, the silver anode reacts to form silver chloride and free electrons which are then used in the reduction of oxygen at the platinum cathode, with the current produced being *proportional* to the oxygen concentration in the surrounding medium [32, 27].

The electrode output is sensitive to several factors: (a) each membrane has a slightly different permeability to oxygen and gives a slightly different reading depending upon the thickness, composition, and age of the membrane (usually polyethylene); (b) the liquid flow past the electrode tip must be sufficiently high to prevent transport of O_2 to the membrane becoming a diffusion controlled (i.e. the electrode is sensitive to the local intensity of turbulence); (c) the electrode is somewhat sensitive to the hydrostatic pressure on the membrane; (d) the electrode reading is proportional to the liquid temperature. Linear temperature calibration curves may be constructed [32].

THEORY

A domain of fine dispersions with high dispersed holdup values in the presence of surfactants

Experiments with fluids having clean interfaces are relatively rare. Many unsuspected impurities in a system can be surface active agents and even the equipment itself can supply enough impurities to make a considerable change in the experimental results. The transfer coefficient, k_L, may be reduced by 150–220 per cent [34–36] in the presence of minute amounts of surfactants. Adventitious surfactants also have a profound effect on the mechanism and time delays of coalescence [44]. Davies *et al.* [35] found a limit to the reduction of k_L with increased concentration of surfactants. This is in agreement with the results of Lindland and Terjesen [45] who found that after a small concentration of surfactant has been used, further additions caused little change in terminal velocity of bubbles.

Therefore, to obtain good reproducibility, the design of the experimental work (as well as the establishment of practical theories) should be considered under such a "degree of contamination" where adventitious fluctuations in surfactant impurities will cause negligible fluctuations in k_L values. For example, a protein "degree of contamination" of 1 mg/m^2 of interface or higher is enough to obtain constant k_L values [35] (i.e. tap water is expected to yield less fluctuations than distilled water). Hence, most laboratory and all industrial dispersions should be analyzed theoretically as though their interfaces are not those of pure fluids [3].

To analyze the effect of surfactants on convective transfer we employ Levich's equation [38]

$$U_{\text{single}} = 3U_{\text{Stokes}}(\mu_c + \mu_d + \gamma)/(2\mu_c + 3\mu_d + 3\gamma) \tag{5}$$

$$U_{\text{Stokes}} = \frac{2}{9}\frac{(\rho_d - \rho_c)\,ga^2}{\mu_c} \tag{6}$$

where γ, the retardation coefficient due to the presence of surfactants may be expressed as [38]

$$\gamma = \frac{4\Gamma_0}{3d}\left[\frac{\partial P^*}{\partial \Gamma} - \frac{\partial Q^*}{\partial \Gamma}\right]^{-1}\frac{\partial \sigma}{\partial \Gamma} \tag{7}$$

when the absorption on the surface is the controlling rate, or, as

$$\gamma = 4RT\Gamma_0^2\bar{\delta}/3D_s\,d\,c^0 \tag{8}$$

when diffusion of surfactants from the liquid bulk to the surface is the controlling rate.

Equations (5), (7) and (8) show that as the bubble diameter decreases, the quantity of surfactants necessary to make a bubble or drop behave like a solid particle, becomes smaller. Hence, under these conditions we define a domain where most industrial dispersions (small and medium size particles in liquid–liquid and gas–liquid dispersions) behave essentially as a dispersion of "solid" particles. Under these circumstances, we may estimate the terminal velocity of a swarm of bubbles or drops by using Happel's "free surface model" [37] for sedimentation that predicts

$$U_{\text{ensemble}} = U_{\text{Stokes}} \cdot \frac{3 - \frac{9}{2}\Phi^{\frac{1}{3}} + \frac{9}{2}\Phi^{\frac{5}{3}} - 3\Phi^2}{3 + 2\Phi^{\frac{5}{3}}}. \tag{9}$$

For equal volumes of dispersed and continuous phases for example, ($\Phi = 0{\cdot}5$) equation (9) predicts that the particle's Stokes velocity (which is already very small for relatively fine dispersions) should be reduced further by a factor of thirty-eight due to hindering effects of its neighbor particles in the ensemble. Hence, in the domain of high Φ values and relatively fine dispersions in actual practice where surfactants are present we assume that the particles are completely entrained by the continuous phase resulting in a negligible convective transfer [3].

A simplified mathematical model for an overall mass-transfer rate in dispersions

Several recent mathematical models have been proposed [1–3] for this domain. In the present study we have experimentally tested the use of the simplest of these [1]. This theoretical model predicts total mass-transfer rates to or from an ensemble of bubbles dispersed in a liquid phase as a function of the average bubble residence time $-\bar{\theta}$, volumetric gas fraction $-\Phi$, volumetric gas flow rate Q, volumetric liquid flow rate F, average bubble size $-a_v$, gas diffusivity in the liquid phase $-D$, interfacial concentration $-C_i$ and concentration of dissolved gas in the liquid phase entering a continuous system $-C_I$. Altogether eight parameters of which three, namely, $\bar{\theta}$, Φ, and a_v are interacting parameters of the ensemble. These can be found experimentally as a function of the operating variables. Interaction effects between adjacent bubbles in the dispersion are taken into account in this model as a function of a_v and $-\Phi$. It is logical to assume [3] that in the case of dispersions, the transfer rates depend on quantities characterizing an ensemble of bubbles or drops. For the case of uniformly distributed particles, $\bar{\theta}$, Φ, and a_v are such quantities. It has been shown [2, 40] that if other distribution functions for the residence time and particle size are employed instead of the average $\bar{\theta}$ or a_v, it is usually found that the results are not markedly affected and the error introduced is usually negligible. The expression thus obtained for the total mass-transfer rate from an ensemble of bubbles in a dispersion is (1):

$$N_T^{\text{ensemble}} = \frac{6Q(c_i - c_I)\left[\dfrac{D\bar{\theta}}{2\Phi^{\frac{1}{3}}a_v^2(\Phi^{-\frac{1}{3}} - 1)^2} - \displaystyle\sum_{n=1}^{\infty}\frac{\exp\left[-n^2\pi^2 D\bar{\theta}/a_v^2(\Phi^{-\frac{1}{3}} - 1)^2\right]}{n^2\pi^2} + \frac{1}{6}\right]}{\dfrac{1}{\Phi^{-\frac{1}{3}} - 1} + \dfrac{3Q}{a_v F}\left[\dfrac{a_v(\Phi^{-\frac{1}{3}} + 2)}{6} + 2\displaystyle\sum_{n=1}^{\infty}\frac{\exp\left[-n^2\pi^2 D\bar{\theta}/a_v^2(\Phi^{-\frac{1}{3}} - 1)^2\right]}{n^2\pi^2}(a_v[\Phi^{-\frac{1}{3}}(-1)^n - 1])\right]} \tag{10}$$

where the mean volume radius $-a_v$ is defined by (2)

$$a_v = \{\sum_{i=1}^{N} n_i a_i^3 / \sum_{i=1}^{N} n_i\}^{\frac{1}{3}}. \tag{11}$$

Equation (10) has been developed for the case of sparingly soluble gases (1). Expressing the volumetric mass-transfer coefficient as

$$k_L a^{\text{ensemble}} = \frac{N_T^{\text{ensemble}}}{V(c_i - c_0)} \tag{12}$$

where c_0 is given by (1)

$$c_0 = c_i - \frac{[Fa_v^2/3Q]\,[c_i - C_I]}{\dfrac{Fa_v^2}{3Q} + a_v(\Phi^{-\frac{1}{3}} - 1)\left[\dfrac{a_v(\Phi^{-\frac{1}{3}} + 2)}{6} + 2\displaystyle\sum_{n=1}^{\infty}\frac{\exp\left[-n^2\pi^2 D\bar{\theta}/a_v^2(\Phi^{-\frac{1}{3}} - 1)^2\right].\,a_v[\Phi^{-\frac{1}{3}}(-1)^n - 1]}{n^2\pi^2}\right]} \tag{13}$$

one realizes that the model enables the prediction of $k_L a$ using a readily available system variables.

In the present experimental study a semi-batch system is operated. Hence, on substituting (10) and (13) in (12), F and C_I are cancelled out resulting in

$$k_L a^{\text{ensemble}} = \frac{6Q(\Phi^{-\frac{1}{3}} - 1)}{V}\left[\frac{D\bar{\theta}}{2\Phi^{\frac{1}{3}}(\Phi^{-\frac{1}{3}} - 1)^2 a_v^2} - \sum_{n=1}^{\infty}\frac{\exp\left[-n^2\pi^2 D\bar{\theta}/a_v^2(\Phi^{-\frac{1}{3}} - 1)^2\right]}{n^2\pi^2} + \frac{1}{6}\right]. \tag{14}$$

Similarly to (1) and (4) the units of (14) are time^{-1}. Therefore, (14) will be employed in the experimental comparison with (4).

EXPERIMENTAL

The Clark electrode

A potential of 0·6 V was supplied across the electrode terminals by an amplifier that converted the electrode current (which is proportional to O_2 concentration at the tip of the electrode) to a voltage suitable for a recorder input. The voltage was recorded on a Bausch and Lomb VOM-6 Recorder (~0·5 s full scale response time). The response time of the electrode for a full scale deflection under the conditions of the experiments, was checked by using a 0·5 ml polyethylene membrane and was found to be less than 3·75 s. To assure that liquid phase concentration was measured, rather than some combination of the gas and liquid concentration, (the electrode tip is introduced into the dispersion) the electrode tip was covered by a screen of 0·15 mm average opening for part of the runs. No change was observed in the response curves for identical changes, indicating that the contact time of the bubbles with the membrane was negligible and that the liquid phase concentration was the quantity measured.

The contactor

The equipment used consists of a one-stage of a recently proposed [41] multistage gas–liquid contactor with a negative pressure drop per stage and a high efficiency of contacting (Fig. 1). The vessel was made from a Plexiglass cylinder 13·4-in I.D. and 24·2-in length. The cylinder is divided into a contacting section 11·5-in length, and a feed and drainage section through which the gas is fed. The feed section is connected to the contacting section by a 0·5-in I.D. pipe located under the center of the impeller and extending 3·56 in into the contacting section. Agitation is provided by a twelve-bladed vaned disc impeller (Fig. 2), 4·06 in. in diameter ($D_{imp}/D_{tank} = 0{\cdot}3$). To provide high shear rates to the dispersion, the contacting section is fitted with a twelve-blade stator (Fig. 3). The clearance between the impeller and stator ring is 0·12 in.

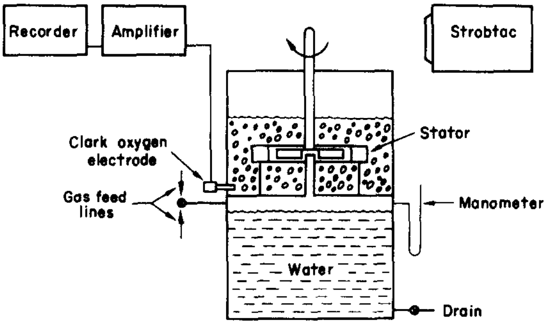

FIG. 1. Schematic diagram of experimental equipment.

Gas feed streams are supplied to the feed section through a nominal 1-in quick opening tee valve open to the atmosphere or connected to prepurified nitrogen supply tanks. Step changes in gas feed inputs are achieved by rapidly switching this valve.

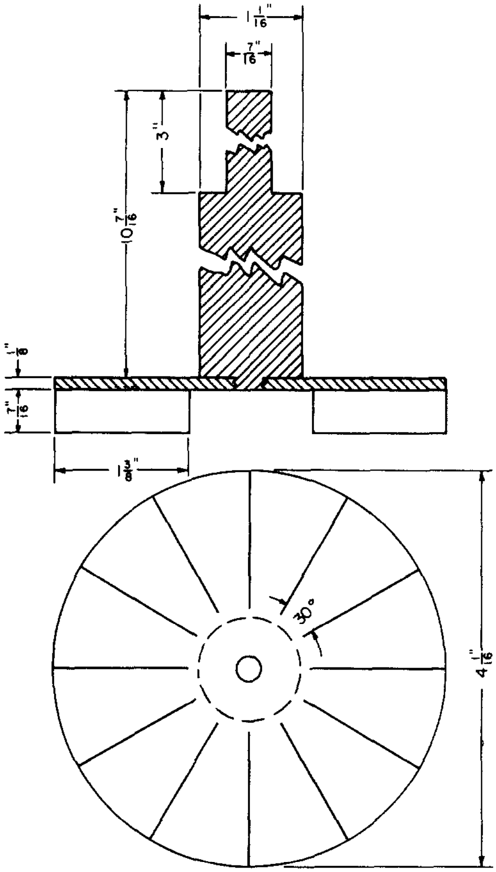

FIG. 2. Data of the twelve-bladed vaned disc impeller.

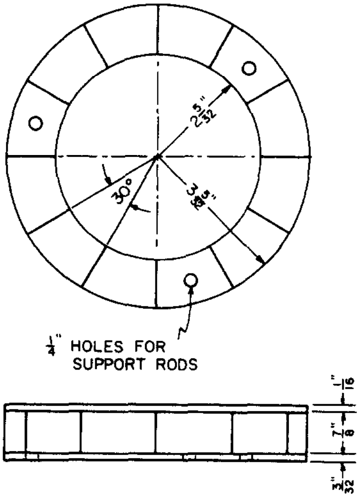

FIG. 3. Data of the twelve-bladed stator.

Auxiliary equipment

Dispersion surface profiles are estimated by taking pictures of the surface profile from a 45° angle above the surface using a series of seven scales 0·25 in. in width spaced (starting from the impeller shaft) 1·5, 1·5, 0·75, 0·75, 0·75 and 0·75 in o.c. along the radius of the tank. The scale heights were adjusted for minimal penetration into the dispersion so that any disturbance to the dispersion motion is minimized. Four pictures were taken for each run with a Sawyers Mark IV twin lens reflex camera equipped with closeup lens. Illumination was provided by a 40-W-s Spiralite electronic flash unit having a flash duration of 1/1000 s.

Procedure

All runs were performed at a temperature of 22·2°C. This temperature was chosen to coincide with the computer solutions of the theoretical model [1]. Tap water was used in all runs to allow the presence of adventitious surfactants as required by the proposed domain [3]. The feed section was filled with water to within 1·25 in of the gas entrance port to minimize the entrance section volume. Using air as a feed, water was added to the contacting section while maintaining a constant prespecified impeller speed until water started to drain to the feed section. Water was then allowed to drain to the feed section under a constant impeller speed until equilibrium was established, i.e. the maximum dispersion height for each particular impeller speed was obtained.

The pressure in the gas feed section was measured after equilibrium had been established and the gas feed was then switched to nitrogen and the flow rate readjusted until the feed section pressure equalled that observed for air feed. The indicated nitrogen flow rate was reported as the volumetric *air* flow rate. This procedure is required, since it is very difficult to measure air flow rates in this system without introducing significant changes in the flow rate due to the pressure drop across the metering device.

The dispersion temperature increased slowly with agitation time due to viscous dissipation. At dispersion temperature of 22·2°C the transient response was initiated by switching the feed gas back to air.

Treatment of data

The volumetric gas holdup fraction, Φ, was estimated from

$$\Phi = \frac{\bar{h}_d - h_L}{\bar{h}_d}. \tag{15}$$

Average dispersion heights, $\bar{h}_d$ were calculated assuming the surface profiles were approximated by straight line segments joining each adjacent scale position. Recalibration of the electrode output is necessary for obtaining concentration data rather than $k_L a$ values. Each set of experimental data contains sufficient information in case recalibration is required since two points are available in each run for this recalibration: The electrode output for nitrogen feed prior to the feed change (assuming zero concentration of oxygen) and the steady-state electrode output with air feed (assuming saturation for the given partial pressure of oxygen). This procedure eliminates the need for any turbulence and pressure correction curves since these are included in each calibration. Liquid phase oxygen concentrations may be thus calculated from Henry's Law with corrections for the partial pressure of water at the given temperature. Typical electrode response is shown in Fig. 4. Volumetric mass-transfer coefficients were calculated from equation (1) or more frequently from the simplified equation (4).

The reported $k_L a$ are those which have become time invariant indicating that the nitrogen bubbles in the disperser had been practically replaced by air bubbles (a constantly decreasing slope of ξ vs. θ is expected for an ideal transient response in which all nitrogen bubbles are replaced instantaneously since here the driving force is monotonically decreased). Thus, the maximum flux does not occur at $\theta = 0$, due to the time lag of the system. The total observed time lag is the sum of the time required for the pulse to reach the contacting section and to displace the nitrogen bubbles in the contacting section, plus the time lags of the analytical equipment. The magnitude of the total time lag indicated by the recorder (before any change in the liquid concentration occurs) is about 3 s, while the time required to reach the maximum instantaneous mass-transfer rate is about 8 s. The total response time required for this system to reach saturation is about 35–45 s.

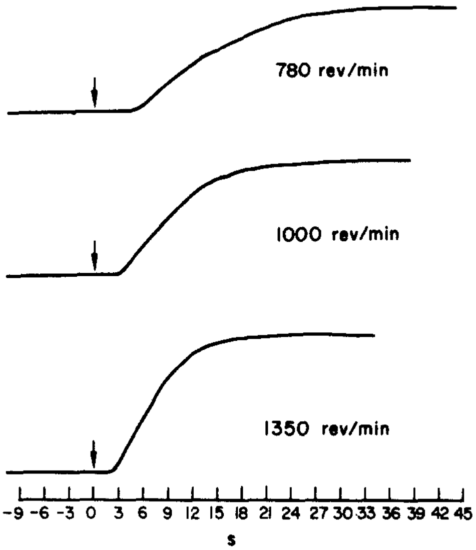

FIG. 4. Typical electrode response curves to a step change in the concentration of oxygen at three impeller speeds.

As expected, k_La values are independent of the transient response time after the maximum rate is reached. A typical example is shown in Fig. 5 which shows that k_La is essentially constant at $\theta \gg 10$ s. This confirms the validity of our assumptions in using equation (4).

RESULTS AND DISCUSSION

Volumetric gas holdup and flow rates

The volumetric gas holdup fraction increases with increasing impeller speeds (Fig. 6). Q, however, increases with impeller speeds up to a maximum at 1300 rev/min (Fig. 7). A decrease in Q is observed above 1300 rev/min which may be attributed to increased recirculation of bubbles that form a secondary gas stream pumped to the impeller zone, thereby decreasing impeller suction of the incoming gas. This is also confirmed by observations of bubble flow patterns [33, 42].

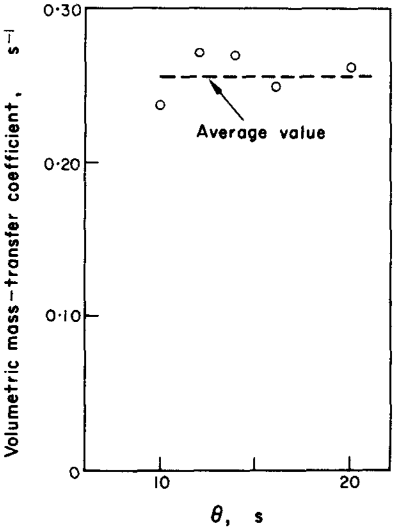

FIG. 5. Typical k_La values as a function of the transient response time. The average values are the reported k_La data for the dispersion.

Average gas residence times

$\bar{\theta}$ values, calculated from

$$\bar{\theta} = \frac{\Phi V}{(1 - \Phi)Q} \tag{16}$$

are shown in Fig. 8, indicating that at impeller speeds less than 1200 rev/min, $\bar{\theta}$ is essentially constant. At higher speeds $\bar{\theta}$ is directly proportional to the impeller speed. This behavior of $\bar{\theta}$ is in agreement with the increasing recirculation of the bubbles that has been discussed above.

Mass-transfer rates

The total interfacial area of the bubbles in a dispersion of sparingly soluble gases can be expressed as [1]

$$A = \frac{3Q\bar{\theta}}{a_v}. \tag{17}$$

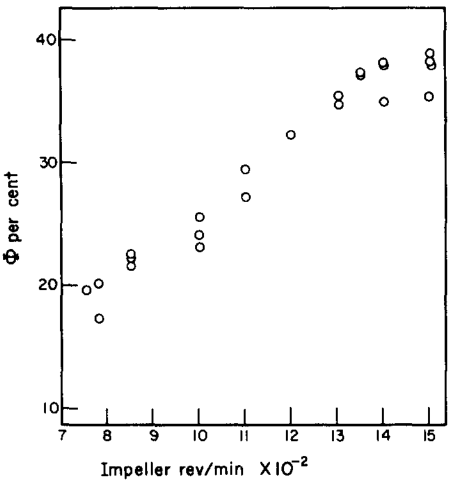

FIG. 6. Volumetric gas holdup fraction vs. impeller speed.

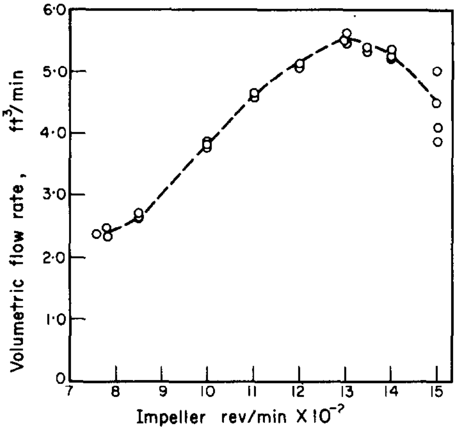

FIG. 7. Volumetric gas flow rate vs. impeller speed.

Experimental evaluation of the particle size distribution in this system [46] shows that $a_v \cong 0{\cdot}07$ cm and is essentially unaffected by the impeller speed. Hence A is proportional to $Q\bar{\theta}$ (i.e. to Φ) and increase of k_La with Φ is due mainly to a proportional increase of the total interfacial area of the bubbles. Hence k_L is essentially unaffected by the turbulence intensity produced by the impeller action.

Computer calculations of equation (14) are shown as a solid line in Fig. 9. Φ, Q and $\bar{\theta}$ values were taken from the present experimental work (Figs. 6–8) with $V = 11{\cdot}8 \times 10^3$ cm^3, $a_v = 0{\cdot}07$ cm [41, 46] and $D = 2{\cdot}3 \times 10^{-5}$ cm^2/s (at 22·2°C) [31]. Equation (14) gives much lower k_La values than the observed ones. The disagreement between the predictions of equation (14) and the experimental data is due mainly to an oversimplified model for the total mass-

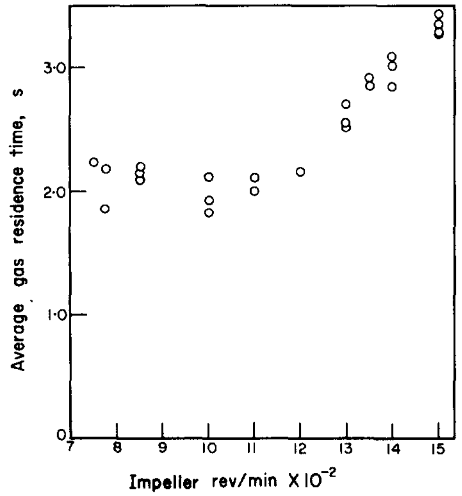

FIG. 8. Average gas residence time vs. impeller speed.

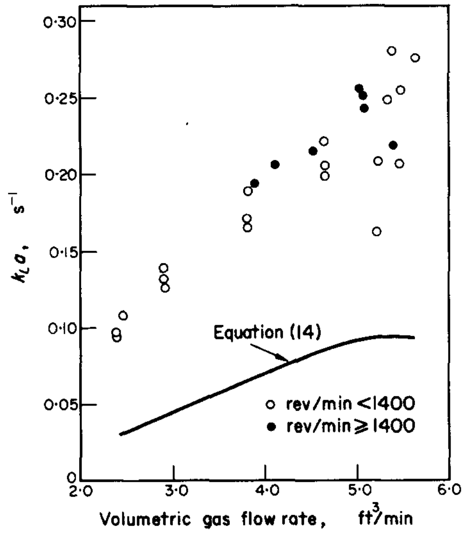

FIG. 9. Volumetric mass-transfer coefficients vs. volumetric gas flow rate.

transfer rate in the dispersion. The main simplifications here include the assumptions of undistorted bubbles, nonexistence of coalescence and breakage of bubbles and a negligible convective transfer. However, a pure convective model for a group of bubbles [3] which has been tested against the present experimental data gives $k_L a$ values few orders of magnitude higher than the observed ones. Hence, experimental values for gas–liquid dispersions will always fall in the region bounded by an upper limit predicted by a pure convective model [3] and a lower limit predicted by equation (14). The bubbles in our system are not completely entrained by the continuous phase eddies (see also [42]) and the convective transfer may contribute about 50 per cent to the mass-transfer flux. Industrial dispersions that contain much more impurities than those present in our tap water–air system will probably give lower $k_L a$ values than the observed ones with a lower bound predicted by equation (14).

Further work is in progress in studying simultaneous (coupled) absorption in ternary systems (oxygen and carbon dioxide in water or blood), effects of surfactant impurities, physical properties and operating variables, and the results will be reported in due time.

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Résumé—On a exploité une méthode de réponse transitoire pour une mesure physique simple et rapide de la concentration et des coefficients de transport de masse volumique dans des dispersions de gaz et de liquide. La méthode implique l'emploi d'une réponse transitoire sous la forme d'un changement brutal de la composition du gaz injecté. Le changement résultant de la composition de la phase liquide de la dispersion est mesuré au moyen d'une électrode de Clark [26] qui permet une analyse rapide et précise des concentrations d'oxygène et de gaz carbonique dans un gaz, le sang ou n'importe quel mélange liquide. Les coefficients expérimentaux de transport de masse volumique sont calculés à partir de:

$$k_L a = \frac{d\xi/d\theta(\theta)}{\xi_{sat} - \xi(\theta)}$$

où ξ est le signal de sortie de l'électrode (ou la lecture de l'enregistrement avec une unité arbitraire) et ξ_{sat} est la lecture finale de l'électrode lorsqu'on a saturation.

La méthode a été appliquée pour essayer un modèle mathématique simple qui permet d'estimer les valeurs de $k_L a$ pour un ensemble de bulles. Des mesures simultanées des fractions volumiques de gaz ont été également effectuées puisque la connaissance du temps de présence moyen des bulles et la proportion de gaz est nécessaire pour prédire les valeurs de $k_L a$ à l'aide de ce modèle. Le modèle mathématique prédit des valeurs de $k_L a$ plus faibles que celles qui sont observées, mais en général donne la limite inférieure pour le comportement de la dispersion en fonction des variables en cause.

Zusammenfassung—Eine instationäre Messmethode wurde für einfache und schnelle physikalische Messungen der Konzentration und des volumetrischen Massenübergangskoeffizienten bei Gas- und Flüssigkeitsgemischen entwickelt. Die Methode beruht auf der Verwendung eines instationären Signals in Form einer stufenweisen Änderung der Zusammensetzung des zuströmenden Gases. Die sich ergebende Änderung in der Zusammensetzung der flüssigen Phase des Gemisches wird mit Hilfe einer Clark Elektrode [26] gemessen, die eine schnelle und genaue Analyse der Sauerstoff- und Kohlendioxid-Konzentrationen im Gas, Blut oder einer beliebigen flüssigen Mischung erlaubt. Die experimentellen volumetrischen Massenübergangskoeffizienten werden nach

$$k_L a = \frac{\mathrm{d}\xi/\mathrm{d}\theta(\theta)}{\xi_{\mathrm{sat}} - \xi(\theta)}$$

berechnet, wobei ξ die Spannung in der Elektrode (oder der Messwert in beliebigen Einheiten) und ξ_{sat} den Endmesswert bei Sättigung darstellt.

Die Methode wurde zur Prüfung eines einfachen mathematischen Modells, das die Abschätzung von $k_L a$ Werten für eine Ansammlung von Blasen erlaubt, angewendet. Gleichzeitige Messungen von Gasstauanteilen wurden ebenfalls durchgeführt, da die Kenntnis der durchschnittlichen Blasenverweilzeit und des Gasstaues für die Bestimmung der $k_L a$ Werte mit Hilfe dieses Modells notwendig ist. Das mathematische Modell liefert geringere $k_L a$ Werte als der Versuch, aber es gibt die untere Grenze für das Verhalten des Gemisches als Funktion der wirksamen Variablen.

Аннотация—Разработан метод переходных характеристик для простого и быстрого физического определения коэффициентов концентрации и объемного массообмена в газожидкостных смесях. Метод использует переходные характеристики в виде ступенчатого изменения состава подводимого газа. Окончательное изменение состава жидкой фазы смеси измерялось электродом Кларка [26], который позволяет быстро и точно проанализировать концентрацию кислорода и двуокиси углерода в газе, крови или любой другой жидкой смеси. Коэффициенты объемного массообмена экспериментально рассчитывались из

$$k_L a = \frac{\mathrm{d}\xi/\mathrm{d}\theta\,(\theta)}{\xi_{\mathrm{sat}} - \xi(\theta)}$$

где ξ — мощность электрода (или показание прибора в любых произвольных единицах) и ξ_{sat} — конечное показание электрода при насыщении.

Метод использовался для исследования простой математической модели, которая позволяет оценить значения $k_L a$ для группы пузырьков. Также одновременно определялось содержание газоывх фракций, поскольку знание среднего времени существования пузырьков и газовой фазы необходимо для нахождения значений $k_L a$ с помощью этой модели. Математическая модель дает заниженные значения $k_L a$ по сравнению с наблюдаемыми, но в общем дает нижнюю границу изменения $k_L a$ как функции действующих переменных.