THE EFFECT OF SURFACE ACTIVE AGENTS ON THE MASS TRANSFER COEFFICIENT IN VERTICAL FILM FLOW OF LIQUID OVER THE SURFACE OF EXPANDED METAL SHEET PACKING

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Experimental data have been used on absorption of poorly soluble gases, helium, carbon dioxide, and propane, to study the mechanism of interfacial mass transfer in vertical gravitational film flow down the surface of the expanded metal sheet. It has been found that upon addition of surface active agents into water, the original almost freely moving liquid–gas interfacial surface behaves similarly as a liquid–solid surface. Two adjustable hydrodynamic parameters have been evaluated on the basis of the film penetration theory with the physical meaning of the dimensionless thickness of an unmixed region near the interface $\vartheta^+ = 0{\cdot}4$ and the characteristic length scale of the disturbance $\lambda^+ = 26{\cdot}4$.

The presence of the surface active agents on the gas–liquid surface may cause, under certain hydrodynamic conditions, a significant decrease of the mass transfer rate during absorption[1]. This is the case of the wavy laminar or turbulent gravitational flow of film over a smooth or roughened surface. On the other hand, the effect of surfacants may be neglected during the laminar waveless flow to which solution of the unsteady diffusion into a film of liquid applies. In these cases the diffusivity, and eventually the reaction rate constant, in case of chemical absorption, may be determined from the rate of absorption of poorly soluble gases in the laboratory in *e.g.* a wetted wall column[2]. Present state of knowledge of mass transfer during the film flow of liquid on a wetted wall has been rewieved in a paper by Kulov and coworkers[3].

Apart from the theoretical significance for the understanding of the hydrodynamics and interfacial mass transfer, the study of the effect of surfactants has also some practical importance. The obtained knowledge can be used in modelling and design of absorption equipment. A decrease of the mass transfer coefficient, k_l, in a random packing, brought about by the presence of surfactants, may be compensated or even offset by increased interfacial surface thanks to the better wetting of the packing surface by liquid exhibiting lower surface tension. In real equipment it is often difficult to study both phenomena separately.

In this contribution we shall present results of measurements of the effect of surfactants on the liquid side mass transfer coefficient in vertical film flow of liquid on a single strip of expanded metal sheet under the conditions of known extent of interfacial surface. The expanded metal sheet packing (Fig. 1) has found recently application in mass transfer equipment and can be used for manufacturing of a packing with regular roughness. The expanded metal sheet turns out to be also a suitable material for hydrodynamic studies. This work represents a continuation of previous studies dealing with the effect of diffusivity of gas in liquid and viscosity on the liquid side mass transfer coefficient[4,5]. The experimental results are interpreted by means of the film penetration model proposed in the preceding paper[5].

THEORETICAL

The film penetration model[5] for the liquid side mass transfer coefficient k_l as

$$k_l = u^*/(\vartheta^+ Sc + (\sqrt{\pi}/2)\,\lambda^+ Sc^{1/2}) \qquad (1)$$

has been based on the assumption that the interface behaves as a semirigid surface and that the layer of liquid adjacent to the interface of thickness ϑ does not mix with the liquid from the bulk of the film and that the transfer follows the steady state diffusion. To the boundary of this region arrive periodic disturbances from the bulk of the liquid film of length scale λ and the time scale $\tau = \lambda^2/\nu$, following from the assumption that the Reynolds number of the disturbance equals unity[6]. In this penetration region unsteady mass transfer takes place. Assuming that the thickness of both regions, ϑ and λ, depend on the friction velocity, u^*, and the kinematic viscosity, ν, similarly as in the single phase flow in a duct, these functions can be

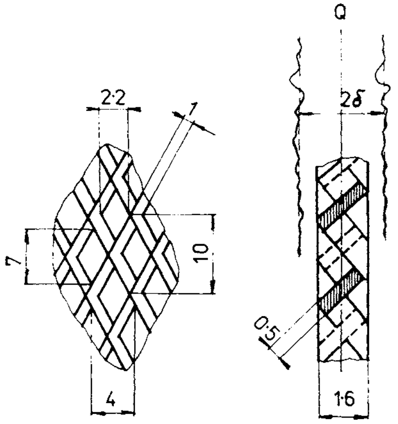

FIG. 1
Sketch of the expanded metal sheet

expressed by means of the dimensionless values ϑ^+ and λ^+ as

$$\vartheta = \vartheta^+ \nu/u^* \quad \text{and} \quad \lambda = \lambda^+ \nu/u^* \,. \qquad (2a,b)$$

The friction velocity, u^*, in this expression is defined in the usual manner

$$u^* = (\tau_w/\varrho)^{1/2} = (g\delta\, 2Hl/A_w)^{1/2} \qquad (3)$$

while the shear stress, τ_w, on the wall of the expanded metal sheet may be determined from the mean thickness of the liquid film, δ, the density, ϱ, gravitational acceleration, g, the length of the film, H, the width of the strip of the expanded metal sheet, l, and the surface of the metal on the sheet, A_w, by the following relation

$$\tau_w A_w = 2\varrho g \delta H l \,. \qquad (4)$$

For the evaluation of the dimensionless coefficients ϑ^+ and λ^+ from the experimental data appears advantageous transformation of Eq. (*1*) into the form

$$u^*/(k_l Sc) = \vartheta^+ + \lambda^+ (\sqrt{\pi}/2)\, Sc^{-1/2} \,. \qquad (5)$$

Values of the dimensionless parameters estimated in the previous paper[5] amounted to $\vartheta^+ = 0{\cdot}04$ and $\lambda^+ = 10{\cdot}6$ for three sizes of the expanded metal sheet, characterized by the following dimensions of the vertical diagonal 10, 16 and 28 mm. The above values were obtained from the experimental data on the rate of absorption of carbon dioxide into nine liquids ranging in kinematic viscosity between 0·6 to 15·1 $\mu \text{m}^2 \text{s}^{-1}$ and in the Schmidt number between 200 and 47 190. The course of the dimensionless mass transfer coefficient, k_l/u^*, as a function of the Schmidt number is shown by curve *2* in Fig. 6.

In the framework of the following study we purposefully chose addition of a surfactant into water as a means of affecting significantly the interfacial surface, while our main concern was to find out what would be the effect on the dimensionless coefficients of the hydrodynamic nature ϑ^+ and λ^+.

For this purpose it is assumed that: *1*) Addition of surfactants will not affect viscosity nor density of water or the solubility and molecular difusivity of gas in water, *i.e.* both in the bulk and at the interface. *2*) The film of surfactants is not affected by the dissolving gas from the side of liquid or gas and *3*) that eventual resistance of the interfacial monomolecular film of surfactants on mass transfer may be neglected.

It has been well established that addition of surfactants in water causes decrease of the surface tension. In order to distinguish between the addition of the surfactant decreasing the surface tension of water from the effect of low surface tension of a pure liquid, yet another experiment was selected: We measured the rate of absorption of carbon dioxide into approximately 88 weight % solution of ethanol in water. In the absence of surfactants the surface tension of this solution was $\sigma =$ $= 0{\cdot}0236\ \mathrm{Nm}^{-1}$. The aim of this experiment was to evaluate the effect of low surface tension on the liquid side mass transfer coefficient, k_1, and to determine the true effect of addition of surfactants on the dimensionless parameters ϑ^+ and λ^+. The results shall be discussed in relation to the film penetration model[5] and the penetration models[1,7-9].

EXPERIMENTAL

The experimental equipment and method have been taken over from the previous works[4,5] and only minor alterations were implemented for the purpose of measurement with surfactants. Tap water was rid of mechanical impurities in a fritted glass filter, and a layer of activated charcoal removed possible traces of surfactants. Then the water was thermostated to 25 ± 0·1°C and the main portion of dissolved air was removed in a vacuum stripping column. Under vacuum the water was pumped by a glass pump with a PTFE plunger into an overflow tank and from here to the top of the absorption column. Diluted solutions of surfactants, prepared from concentrated solutions specified in Table I by dilution in the ratio 1 : 1, 1 : 10, 1 : 100, and 1 : 500 were continuously dispensed in the requested ratio by a micropump into the water prior to its metering in a rotameter. Its good mixing was ensured in the pipe leading to the top of the absorption column.

The rate of absorption of poorly soluble gases, helium, carbon dioxide and propane, into the two mentioned systems, characterized in Table II, was measured on a single strip of expanded metal sheet 63 mm wide with the length of the liquid film 570 mm. The vertical diagonal of the mesh of expanded metal sheet measured 10 mm. A laboratory gasometer was used to measure the rate of absorption of carbon dioxide, while a gas burette was used in case of helium and propane. Surface tension of distilled and tap water and of the solutions of surfactants in the concentration range between 0·1 and 1 000 ppm was measured on the Mohr balance. Each measurement was repeated several times and its accuracy was about $\pm 0{\cdot}001\ \mathrm{Nm}^{-1}$. For the determination of the mean thickness of the liquid film on the expanded metal sheet a method of weighing[5] was used to measure the hold-up of liquid as a function of the concentration of surfactants.

TABLE I

Investigated samples of surface active agents

Designation	Chemical formula	Weight %
ETOXAN	$C_nH_{2n+1}O(C_2H_4O)_2C_2H_4OSO_3Na$ (n = 12–14)	40
W-50	$C_{15}H_{31}SO_3Na$	60
JAR	commercial dish washing detergent	25

RESULTS

The solid line in Fig. 2 shows the course of the surface tension of the solution of ETOXAN in distilled water. In region of the concentrations of the surfactants less than 0·1 ppm the employed technique of measuring of small changes of surface tension was not sufficiently sensitive. In the region above 40 to 60 ppm the addition of surfactant had little effect on further decrease of the surface tension. This concentration approximately corresponds to the critical micellar concentration (CMC) and the measured surface tension at this concentration was $\sigma = 0{\cdot}035\ \mathrm{Nm}^{-1}$. A similar course was also found for other investigated samples of surfactants.

The experimental points in the same figure depict the decrease of the mass transfer coefficient due to the addition of ETOXAN into the tap water. The character of the dependence of the mass transfer coefficient, k_l, is similar to the dependence of the surface tension. Full points on the abscissa correspond to k_l in the absence of surfactants. These values were checked always at the beginning and at the end of each experimental run for a given value of the Reynolds number. From Fig. 2 there also follows that at sufficiently high concentration of the surfactants the liquid side mass transfer coefficient reaches a minimum for the given Reynolds number. The concentration of 30 ppm of the surfactant, evaluated for the Reynolds number $Re = 200$, is in good agreement with the critical micellar concentration of 35 ppm, evaluated from the measurement of the surface tension. At higher values of the Reynolds number the liquid film is more intensively mixed and hence higher concentrations of surfactants are needed to supress mixing in the proximity of the

TABLE II

Physico-chemical properties of liquids used and solubilities of gases

Quantity	Water	88% (weight) aqueous solution of ethanol
$\nu \,.\, 10^6\ \mathrm{m^2\,s^{-1}}$	0·896	1·82
$\varrho\ \mathrm{kg\,m^{-3}}$	997	823
$\sigma \,.\, 10^3\ \mathrm{Nm^{-1}}$	72; 35 (PAL)	23·6[16]
$x \,.\, 10^4\ \mathrm{mol\,.\,mol^{-1}}\ (C_3H_8)$	0·2704[13]	—
$x \,.\, 10^4\ \mathrm{mol\,.\,mol^{-1}}\ (CO_2)$	6 . 111[13]	38·3[17]
$x \,.\, 10^4\ \mathrm{mol\,.\,mol^{-1}}\ (He)$	0·07046[13,14]	—
$D \,.\, 10^9\ \mathrm{m^2\,s^{-1}}\ (C_3H_8)$	1·16[10,11]	—
$D \,.\, 10^9\ \mathrm{m^2\,s^{-1}}\ (CO_2)$	1·92[12]	3·3[15]
$D \,.\, 10^9\ \mathrm{m^2\,s^{-1}}\ (He)$	6·28[12]	—

Note: x is mole fraction of dissolved gas in liquid at 25°C and partial pressure of 101 325 Pa.

interface. As follows from Fig. 2 the addition of surfactants for practical purposes is disadvantageous as it substantially decreases the liquid side mass transfer coefficient The rate of absorption is significantly decreased if the resistance to mass transfer is concentrated on the liquid side and the increase of the interfacial area due to the presence of the surfactants cannot offset this decrease. For the purposes of our measurement the high concentration of surfactants is advantageous firstly because above the CMC k_l is practically independent of the concentration of surfactants and, secondly, its low value permits also higher precision of the determination of the coefficient k_l from the degree of saturation of liquid.

Fig. 3 plots values of the mass transfer coefficient, k_l, obtained from absorption of carbon dioxide in water, ethanol and water with 63 and 660 ppm of the surfactant JAR *versus* the Reynolds number. From the figure it is apparent that addition of surfactant causes a significant decrease of the mass transfer coefficient. During absorption into pure liquids the expanded metal sheet exhibits by a manifold higher values of the mass transfer coefficient compared to the smooth wetted wall. For the latter see the full line for the first and the second laminar regime and the turbulent regime of the liquid film flow[3]. It is interesting to note that after addition of the surfactant values of the mass transfer coefficient for the expanded metal sheet

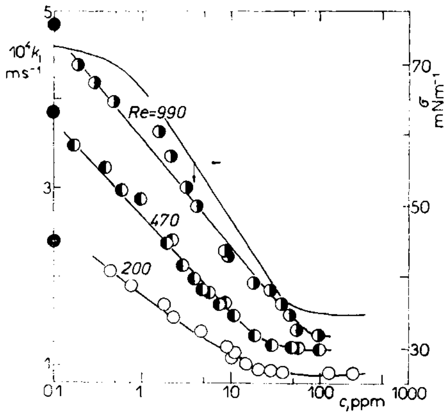

FIG. 2

Surface tension and mass transfer coefficient as functions of the concentration of surfactants; no surfactants ●, with surfactants: ○ *Re* = 200, ◐ *Re* = 470 ◑ *Re* = 990

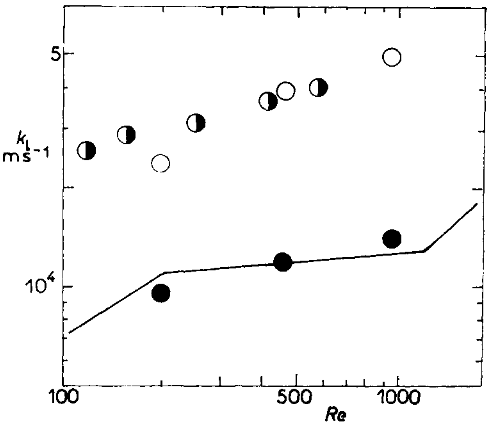

FIG. 3

The mass transfer coefficient as a function of the Reynolds number. Solid lines computed from correlation[3] valid for the first and the second laminar and the turbulent flow on a smooth wetted wall. Experimental data on expanded metal sheet ○ CO_2/water, ◑ CO_2/ethanol, ● CO_2/water + surfactant decrease to a level corresponding to the second wavy laminar regime of the smooth wetted wall. At the same time, however, addition of surfactants suppresses formation of the waves on the smooth wetted wall and also substantially reduces the mass transfer coefficient, k_l.

According to the film penetration model, Eq. (*1*) ,the decrease of the mass transfer coefficient, following an addition of the surfactant, may be caused by decreased value of the friction velocity, u^*, or increased values of the dimensionless parameters ϑ^+ and λ^+. From Eqs (*3*) and (*1*) there follows that the transfer coefficient is proportional to the square root of the mean film thickness δ. For this reason we have experimentally verified the effect of addition of the surfactants on the mean thickness of the liquid film. Under the conditions of complete coverage of the expanded metal sheet by the film of liquid we found that concentrations of the surfactants in excess of the CMC increase the thickness of the film by 3% at most. We may thus use the original correlation in the form

$$\delta(g/v^2)^{1/3} = aRe^{b} \qquad (6)$$

with empirical constant $a = 0.51$ and $b = 0.46$ even for systems containing surfactants. Since the thickness of the film of liquid with the addition of surfactants slightly increases, the significant decrease of the mass tranfser coefficient cannot be attributed to a change of the friction velocity u^*.

Figure 4 plots the dependence of the dimensionless mass transfer coefficient, k_l/u^*, on the Reynolds number for all three systems used during absorption of carbon dioxide. From the figure it is apparent that, as anticipates Eq. (*1*), the dimensionless mass transfer coefficient is independent of the Reynolds number of liquid. The broken lines show the dependences for absorption of helium, carbon dioxide and propane into water in the absence of surfactants calculated from Eq. (*1*) for $\vartheta^+ = 0.04$ and $\lambda^+ = 10.6$; solid lines pertain to absorption of CO_2 into ethanol.

Fig. 5 plots the experimental data in the variables of Eq. (*5*) permitting values of the dimensionless parameters ϑ^+ and λ^+ to be determined as a segment of the ordinate and the slope of the straight line. In view of the fact that absorption into water containing surfactants was investigated with only three gases (helium, carbon dioxide, propane) the segment of the ordinates, $\vartheta^+ = 0.4$, and the slope of the straight line, $\lambda^+ = 26.4$, were evaluated only graphically. The small effect of the surfactants in the concentration range between 63 and 660 ppm is clearly visible from the graph. The experimental point corresponding to absorption of carbon dioxide into ethanol, *i.e.* liquid of low surface tension, yet free of the surfactants, is in good agreement with the earlier obtained[5] dependence given by the solid line, for which the following values hold $\vartheta^+ = 0.04$ and $\lambda^+ = 10.6$.

DISCUSSION

The liquid side mass transfer coefficient, k_1, decreases with increasing concentration of surfactants and at the critical micellar concentration decreases to about one third of the value in the absence of the surfactants. More accurate values for individual gases, helium, carbon dioxide and propane, computed from Eq. (*1*), are summarized in Table III.

As may be apparent from Table III the relative decrease of the mass transfer coefficient depends on the diffusivity of gas or, in other words, on its Schmidt number. Maximum decrease exhibits propane, which, in comparison with the other gases, has also the greatest part of the resistance to mass transfer concentrated in the unmixed region of thickness ϑ near the interface. This fraction of the total resistance may be expressed from Eq. (*1*) as the ratio $\vartheta^+ Sc/(\vartheta^+ Sc + (\sqrt{\pi}/2)\,\lambda^+ Sc^{1/2})$. In addition, as follows from Table IV, the thickness of the unmixed region ϑ represents only a small portion of the mean thickness of the liquid film on the strip of the expanded metal sheet. To give an example, for the Reynolds number $Re = 1\,000$, the thickness of the unmixed region in case of water and no surfactants equals 0·4 micrometers and for the CMC of surfactants reaches a ten times higher value, *i.e.* 4 micrometers.

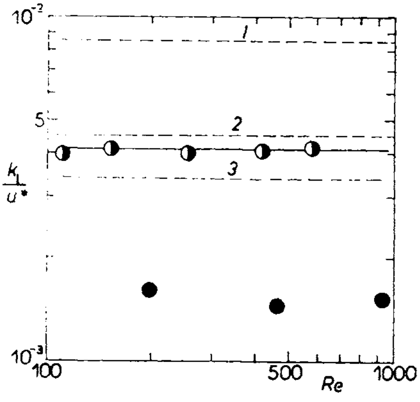

FIG. 4

The dimensionless mass transfer coefficient k_1/u^* as a function of the Reynolds number. ◑ CO_2/ethanol, ● CO_2/water + surfactant. Broken and solid lines computed from[5] Eq. (*1*) for $\vartheta^+ = 0{\cdot}04$ and $\lambda^+ = 10{\cdot}6$. *1* He, *2* CO_2, *3* C_3H_8

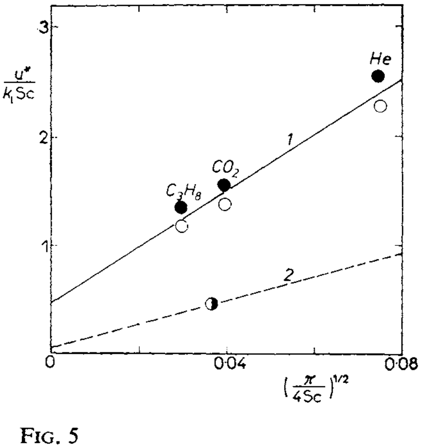

FIG. 5

Evaluation of ϑ^+ and λ^+ from experimental data. Curve *1* $\vartheta^+ = 0{\cdot}4$, $\lambda^+ = 26{\cdot}4$, water + + surfactant ○ 63 ppm, ● 660 ppm. Curve *2* $\vartheta^+ = 0{\cdot}04$, $\lambda^+ = 10{\cdot}6$, no surfactant[5], ◑ CO_2/ethanol

The effect of CMC of surfactants on the decrease of the dimensionless mass transfer coefficient, k_l/u^*, in a broader interval of values of the Schmidt number is apparent from comparison of the solid lines *2* and *3* in Fig. 6. Analysis of experimental data from the region between these two curves was not carried out in detail, even though it could be quite useful for a better understanding of the mechanism of function of the surfactants on the interface. As follows from Fig. 2 this region exhibits a strong dependence of the mass transfer coefficient on the concentration of the surfactants. Accordingly, the employed experimental technique would have to be modified if sufficiently accurate and reproducible data were to be obtained. The high concentrations of the surfactants chosen in this work were advantageous for two reasons: The first is the fact that above the CMC k_l practically does not depend on the concentration of surfactants. The second reason is the fact that the low value of the mass transfer coefficient, k_l, after addition of the surfactant, is advantageous from

TABLE III

The effect of surfactant on the mass transfer coefficient

Quantity	Gas		
	Propane	Carbon dioxide	Helium
Diffusivity $D \cdot 10^9$ ($m^2 s^{-1}$)	1·16	1·92	6·28
Schmidt number	772	467	143
$k_l(CMC)/k_l(0)$	0·304	0·320	0·350
Fractional resistance to mass transfer in unmixed region of thickness ϑ	water 0·110	0·084	0·048
	water + 0·322 + surfactant	0·130	0·060

TABLE IV

The effect of the Reynolds number on the film thickness and the ratio ϑ/δ for water at 25°C with and without surfactants

Thickness of liquid film		Ratio ϑ/δ	
Re	$\delta \cdot 10^3$ (m)	water	water+surfactant
100	0·184	0·0039	0·039
1 000	0·531	0·0008	0·008

the standpoint of acccuracy of its determination. This is particularly significant for absorption of helium, *i.e.* a gas exhibiting highest diffusivity, for which the degree of saturation of liquid approaches unity for already short lengths of the film flowing down the surface structure of the expanded metal sheet. Naturally we considered it worthwhile to increase the number of gases used in experiments in order to expand the range of diffusivity covered.

Fig. 6 shows also the limiting case of the zero thickness of the unmixed region $\vartheta^+ = 0$ as curve *1*. It represents the penetration model with an exponent over diffusivity equalling one half, similarly as is the case of the Higbie model[7]

$$k_l = (4D/(\pi\tau))^{1/2} \tag{7}$$

and the Danckwerts[8] model

$$k_l = (Ds)^{1/2}\,, \tag{8}$$

where τ is the time of penetration and s the rate of surface renewal. Penetration character has also the model of Davies and Levich[9]

$$k_l = 0{\cdot}32 D^{1/2} u^{*3/2} \varrho^{1/2} \sigma^{-1/2} \tag{9}$$

in which the proposed dependence of the mass transfer coefficient on the surface tension of pure liquid with an exponent minus one half was not confirmed by our experiments. Our investigated system carbon dioxide–ethanol, exhibiting surface tension $\sigma = 0{\cdot}035\ \mathrm{Nm}^{-1}$, corresponds to the originally found values of the dimensionless parameters $\vartheta^+ = 0{\cdot}04$ and $\lambda^+ = 10{\cdot}6$ shown in curve 2.

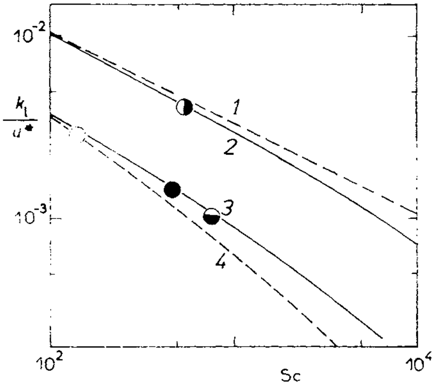

FIG. 6

The dimensionless mass transfer coefficient as a function of the Schmidt number. ○ He, ● CO_2, ◒ C_3H_8/water+surfactant, ◑ CO_2/ethanol

Line	*1*	*2*	*3*	*4*
ϑ^+	0	0·04	0·4	1
λ^+	10·6	10·6	26·4	20

The curve *4* for $\vartheta^+ = 1$ and $\lambda^+ = 20$ was obtained from experimental mass or heat transfer data under the single phase flow in a tube of circular cross section[6,18,19] and thus describes the situation at the solid wall/gas or solid wall/liquid interface. As the values for the dimensionless coefficient, k_l/u^*, and the dimensionless values of the hydrodynamic parameters are very close to the results obtained in the mass transfer experiments with the addition of surfactants, it may be concluded that the originally almost freely moving gas/liquid interface behaves in the presence of surfactants as a solid wall.

LIST OF SYMBOLS

a	empirical constant in Eq. (*6*)
A_w	surface area of metal of expanded sheet, m^2
b	empirical constant in Eq. (*6*)
D	diffusivity of gas in liquid, $m^2\,s^{-1}$
g	acceleration due to gravity, 9·81 $m\,s^{-2}$
H	length of liquid film, m
k_l	liquid side mass transfer coefficient, $m\,s^{-1}$
l	width of strip of expanded metal sheet, m
Q	volume flow rate of liquid, $m^3\,s^{-1}$
$Re = 4\Gamma/\mu$	Reynolds number
s	intensity of surface renewal, s^{-1}
$Sc = \nu/D$	Schmidt number
u^*	friction velocity of liquid defined in Eq. (3), $m\,s^{-1}$
x	mole fraction of dissolved gas in liquid at partial pressure $P = 1{\cdot}01325 \,.\, 10^5$ Pa
$\Gamma = Q\varrho/2l$	linear density of irrigation $kg\,m^{-1}\,s^{-1}$
δ	mean thickness of liquid film on expanded metal sheet (Fig. 1)
ϑ	thickness of unmixed region at the interface, m
ϑ^+	dimensionless thickness of unmixed region at the interface
λ	length scale of disturbance, m
λ^+	dimensionless length scale of disturbance
μ	dynamic viscosity, $kg\,m^{-1}\,s^{-1}$
ν	kinematic viscosity, $m^2\,s^{-1}$
ϱ	density, $kg\,m^{-3}$
σ	surface tension, $N\,m^{-1}$
τ	time scale of disturbance, penetration time, s
τ_w	friction stress on the wall of expanded metal sheet, Nm^{-2}

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