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Interfacial Area and Mass Transfer Coefficients in Liquid-Gas Ejectors[1]

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Abstract—Measurements and correlations are reported for the interfacial area and mass transfer coefficients as a function of energy dissipation in a liquid-gas ejector. The correlations for interfacial area and mass transfer coefficients have been developed using Kolmogorov's theory and Levich's hydrodynamic derivations. The present developed correlations are validated using experimental results.

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INTRODUCTION

The production/absorption rate of gas-liquid systems is often controlled by gas-liquid interfacial mass transfer. Particularly for the removal of acid gases from natural gas, packed columns have been used traditionally. Since acid gases undergo chemical absorption with amine solutions (absorbents), large number of stages is no more required for attaining equilibrium. Simultaneous aspiration and dispersion of the entrained fluid takes place in the cocurrent gas-liquid ejector systems, which causes continuous formation of fresh interface and hence the generation of large interfacial area [1]. Very small bubbles/droplets of the dispersed phase were produced in the liquid-gas ejectors, thereby increasing the contact between the phases, which in turn enhances the mass transfer rates. The higher efficiency of ejectors for gas dispersion is due to the micro turbulence which causes the bubble break up [2]. It was also reported that the mass transfer coefficient in high intensity gas-liquid mixers is two orders of magnitude higher than in conventional gas-liquid contactors. Higher values of interfacial area and mass transfer coefficients lead to a substantial reduction in the size of the contacting equipment, thereby reducing the capital investment cost of the equipment. The important design and the scale-up parameters for such type of contactors are the interfacial area for mass transfer and the true mass transfer coefficients apart from the phase holdup. Even though there have been a number of publications [3–11] on liquid jet gas ejectors, none of them provides detailed information, that is required for the design and scale-up. A detailed literature review pertaining to the hydrodynamics of the liquid-gas ejector is presented in our earlier paper [12]. Hence in the present work, an attempt is made to develop a theoretical expression from the first principles, to represent the interfacial area and mass transfer coefficients data for liquid-gas ejectors.

EXPERIMENTS

The schematic diagram of the present experimental setup is shown in Fig. 1 and the details of the experimental procedures are explained in our earlier work [12]. The experimental assembly consists of the ejectors E, which expends into the air-water separator SE. Instrumentation have been provided for metering all the flows and for the measurement of the pressure distribution along the ejector. Liquid, the motive fluid, is pumped from the tank T, while carbon dioxide, the secondary fluid, is supplied by the compressor C. An intense mixing zone occurs within the ejector, where CO_2 disperses into the alkaline solution as fine bubbles. Then the mixture moves upward through the column and the unreacted CO_2 is separated at the top of the column. The gas phase holdup was measured by the liquid displacement method [12]. The interfacial area was determined by the chemical reaction method by the absorption of CO_2 in sodium hydroxide [13]. The pressure points were modified to obtain continuous gas samples for infrared analysis. The range of experimental conditions used for the present study was as follows: liquid flow rate 30–150 ml/s; CO_2 flow rate 40–300 ml/s; area ratio of nozzles 3.16–28.44; inlet concentration of CO_2 5%; inlet concentration of NaOH 0.7 N. Special care was taken to ensure the conditions so that the absorption took place in the pseudo first order, fast reaction regime [14]. True liquid side mass transfer coefficient was determined by using two techniques, the pseudo first order - interme-

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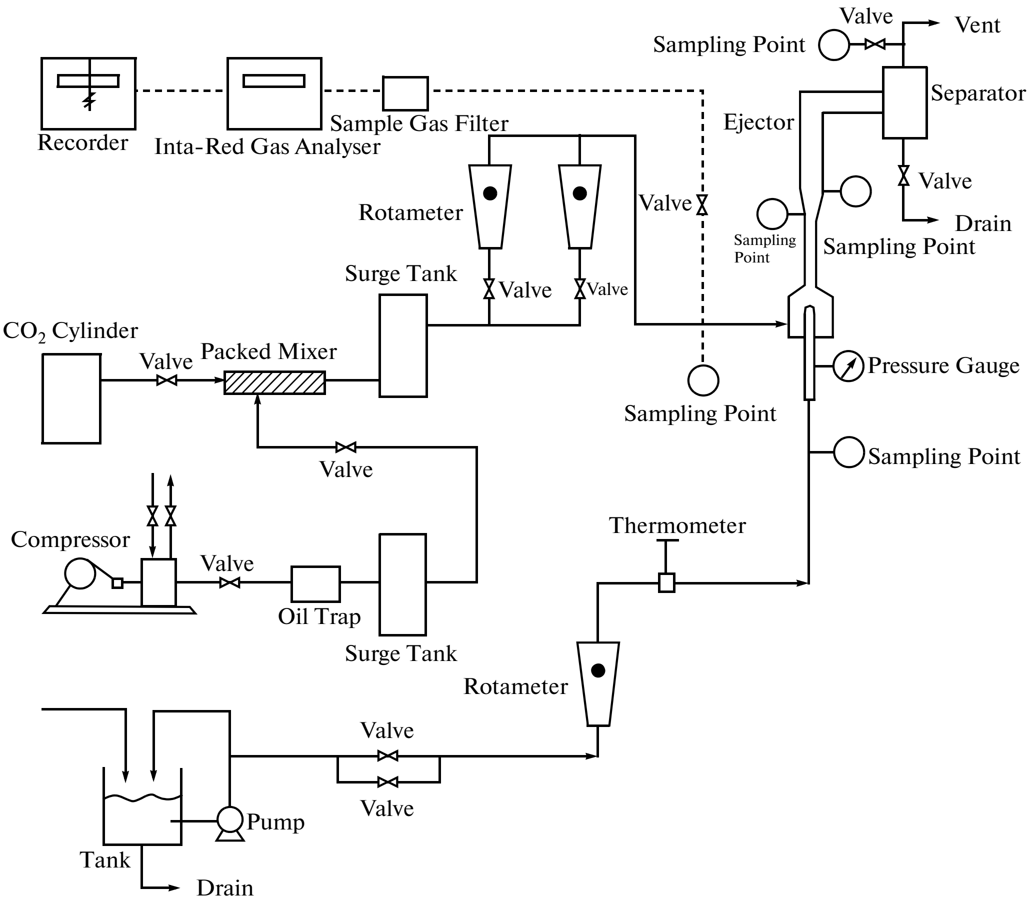

Fig. 1. Experimental setup.

diate rate regime chemical absorption method [14] and physical absorption method [4]. Absorption of CO_2 in carbonate-bicarbonate buffers was used for the first method and for physical absorption CO_2 - water was used. All the required conditions for ensuring pseudo first order intermediate rate regime as recommended by Danckwerts and Sharma [14] were maintained. Few experiments were conducted to test whether the two methods gave similar results under the identical hydrodynamic conditions and a satisfactory agreement was found. In the present experiments, the values for the specific interfacial area a were obtained in the range of $3 \times 10^2 - 44 \times 10^2$ m^{-1}, whereas for the volumetric coefficient k_{la} ranged from 2×10^{-4} to 15×10^{-4} m/s.

DEVELOPMENT OF CORRELATION FOR INTERFACIAL AREA

The variation of the interfacial area with the primary fluid flow rate is shown in Fig. 2. Similar results were obtained with all the other nozzles tested. The interfacial area and mass transfer coefficient are the important parameters for the design of gas-liquid contacting equipment and have been extensively studied for various configurations [15]. It has been generally recognized that the dominant parameter affecting the interfacial area and mass transfer coefficients in gas-liquid systems is the specific energy input to the system. Various authors [3–11] have attempted to use some form of the pressure drop as a measure of the energy dissipation and to correlate the pressure drop with the interfacial area and liquid side mass transfer coefficient. In the liquid-gas ejector systems, the interfacial area and the mass transfer coefficients are intimately connected with the structure of the turbulence. Therefore, it should be possible to relate these parameters with the characteristics of turbulence, viz., its scale and intensity, through Kolmogorov's theory [16] and Levich's hydrodynamic deviations [17]. If a bubble is placed in turbulent fluid, it breaks up under the action of turbulent eddies. This fragmentation process takes place when the dynamic pressure force exceeds the capillary force holding the bubble together. The difference in dynamic pressure between the opposite sides of a gas bubble is given by [17]

$$\Delta P = K_{f1}\rho_1 \frac{u_{1a}^2 - u_{1b}^2}{2}. \quad (1)$$

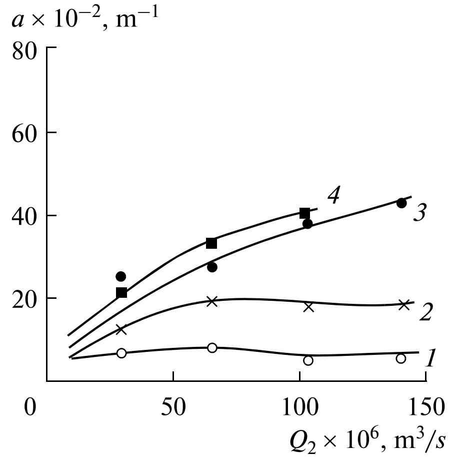

Fig. 2. Variation of the specific interfacial area with liquid and gas flow rates (nozzle 3, $A_R = 10.31$): $Q_1 =$ (*1*) 40 × 10^{-6}, (*2*) 1120 × 10^{-6}, (*3*) 255 × 10^{-6}, and (*4*) 310 × 10^{-6} m^3/s.

For large eddies, the change in velocity over a bubble will be small, the bubble fragmentation is essentially caused by small eddies. According to Kolmogorov's theory of spectral energy transfer [16], energy is extracted from large eddies and cascades down the spectrum to the small eddies till finally lost as heat. Further, it has been postulated that at sufficiently high Reynolds numbers, there is a range of wave numbers where turbulence is statistically in equilibrium. Under such conditions, the only parameters affecting the energy transfer are the density, eddy velocity and eddy scale. By dimensional similarity,

$$\varepsilon = \alpha \frac{\rho_1 v_\lambda^3}{\lambda}, \qquad (2)$$

$$v_\lambda \cong \left(\frac{\varepsilon\lambda}{\rho_1}\right)^{1/3}. \qquad (3)$$

As a first approximation, the eddy velocity v can be taken as the velocity difference over the eddy scale λ, or

$$v^2 = u_{1a}^2 - u_{1b}^2. \qquad (4)$$

Therefore, the dynamic pressure, Eq. (1) becomes

$$P = \frac{K_{f1}\rho_1}{2}\left(\frac{\varepsilon\lambda}{\rho_1}\right)^{2/3}. \qquad (5)$$

According to Levich [17], the drag force acting on a gas bubble sets the gas inside in motion and creates a dynamic pressure proportional to $\rho_2 u_2^2/2$, within the bubble, acting radially outwards. If the dynamic pressure exceeds the capillary forces, the bubble will break. Therefore the condition for the bubble fragmentation is

$$\frac{K_{f2}\rho_2 u_2^2}{2} \geq \frac{\sigma\pi h^2}{V}. \qquad (6)$$

The characteristic bubble dimension in the direction of motion is given by

$$h = \frac{2\sigma}{\rho_1 u_1^2}. \qquad (7)$$

Substituting the value of h in Eq. (6) and with the assumption that the gas velocity u_2 is approximately equal to the interphase velocity u_1, an expression for the maximum bubble volume which can exist in a flow field can be written as

$$V \leq \frac{8\pi\sigma^3}{K_{f2}\rho_1^2\rho_1 u_1^6}, \qquad (8)$$

with $V = \pi d^3/6$,

$$(d)_{\max} = \left(\frac{6}{K_{f2}}\right)^{1/3} \sigma \left(\frac{\rho_1}{\rho_2}\right)^{1/3} \frac{2}{\rho_1 u_1^2}. \qquad (9)$$

In a turbulent field, the breakup of a bubble of diameter greater than d will be caused by turbulent eddies of scale $\lambda = d_B$. Therefore, the eddy velocity of interest v_λ can be obtained from Eq. (3) with $\lambda = d$ or

$$v_\lambda \cong \left(\frac{\varepsilon d}{\rho_1}\right)^{1/3}. \qquad (10)$$

In Eq. (9), the interphase velocity u_1 is the eddy velocity as given in Eq. (10). Substituting this expression in Eq. (9), we get an expression for the maximum bubble size as

$$d_{\max} = \left(\frac{6}{K_{f2}}\right)^{1/3} \sigma \left(\frac{\rho_1}{\rho_2}\right)^{1/3} \left(\frac{2}{\rho_1}\right)\left(\frac{\varepsilon d_B}{\rho_1}\right)^{-2/3}, \qquad (11)$$

which may further be simplified as

$$d_B = C_1 \sigma^{0.6} \varepsilon^{-0.4} \rho_2^{-0.2}. \qquad (12)$$

The proportionality of bubble diameter to $\rho_2^{-0.2}$ is in good agreement with the observations of Cramers et al. [1]. The interfacial area and gas holdup are related to the bubble diameter and the number of bubbles in the system by the equation

$$A = Nd^2, \qquad (13)$$

$$\alpha_2 V_R = Nd^3. \qquad (14)$$

Therefore,

$$a = \frac{A}{V_R} = \frac{\alpha_2}{d_B}. \qquad (15)$$

Substituting for bubble diameter in Eq. (12), we get

$$a = C_2 \varepsilon^{0.4} \alpha_2. \qquad (16)$$

Equation (16) is well applicable to low gas concentrations where the turbulence is created by the liquid phase alone. Where the gas holdup is high, Calderbank [18] proposed a correction factor to the exponent of the gas holdup to take into account the turbulence created by the gas phase. Incorporating this correction factor m in Eq. (16), we get

$$a = C_3 \varepsilon^{0.4} \alpha_2^m. \qquad (17)$$

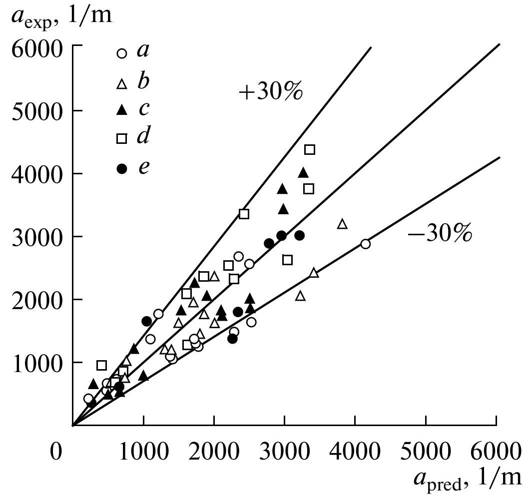

Fig. 3. Comparison of predicted and experimental data on specific interfacial area: (*a*) nozzle 1, $A_R = 3.16$; (*b*) nozzle 2, $A_R = 5.23$; (*c*) nozzle 3, $A_R = 10.31$; (*d*) nozzle 4, $A_R = 16$; and (*e*) nozzle 5, $A_R = 28.44$.

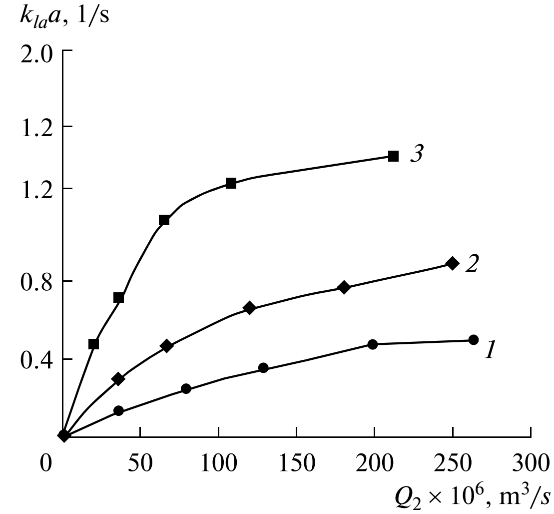

Fig. 4. Variation of the liquid side volumetric mass transfer coefficient $k_{la}a$ with gas and liquid flow rates for the CO_2–water system (nozzle 1, $A_R = 3.16$): $Q_1 =$ (*1*) 30×10^{-6}, (*2*) 67×10^{-6}, and (*3*) 1104×10^{-6} m^3/s.

For the cases where the liquid density is constant, we can replace the energy input per unit mass by the specific total energy input. The total energy dissipated can be expressed as [12]

$$E_t = Q_1 H\left(k_n + k_{th} + k_{di}\right), \tag{18}$$

where H is the jet velocity head given by $\rho_1 v_1/2$. The losses in the nozzle, throat and diffuser were accounted for by three loss coefficients k_n, k_{th}, and k_{di}, respectively [12]. The values for the throat and diffuser loss coefficients were estimated using the available literature correlation [8, 12]. These coefficients represent the fraction of the jet momentum which is lost as irreversible losses. Hence Eq. (17) could be written as

$$a = C_3 E_t^{0.4} \alpha_2^m. \tag{19}$$

A similar approach based on the eddy models for determining k_{la} and a was used by Linek et al. [19] for stirred tank gas-liquid reactors. The measured values of the CO_2 concentration in the inlet and outlet gas streams were used to determine the interfacial area by the method suggested by Voyeur and Miller [20]. The energy dissipation and gas phase holdup corresponding to these conditions were calculated using the correlation developed in the earlier part of this paper [12]. The present data on interfacial area were correlated with the energy input and gas holdup to give the following relationship:

$$a = 7.92 \times 10^3 E_t^{0.36} \alpha_2^{1.53}. \tag{20}$$

The above correlation is similar to theoretical Eq. (19). The lower value for the exponent, 0.36 for the energy against 0.4 in the theoretical equation can be explained from the fact that the theoretical equation is based on the actual energy used for area generation while the correlation uses the total irreversible energy. As predicted by theory, the gas holdup is raised to a power higher than 1. A comparison between the predicted values (Eq. (20)) and the experimental interfacial area data (Fig. 3) and the root-mean-square errors were found to be within ±20%. The range of interfacial area obtained in the present ejector system is higher than that generated in two-phase contactors, two-phase pipe flow, venturi scrubbers, bubble columns and packed columns.

LIQUID SIDE MASS TRANSFER COEFFICIENT

The variation in the liquid side mass transfer coefficients with gas and liquid flow rates is shown in Fig. 4. When a gas bubble is suspended in a turbulent field, the larger eddies entrains the bubble together with the liquid interphase and carries both as a single unit. As we go down the eddy scale, the smaller eddies will not be able to completely entrain the bubble and a relative velocity will exist. Still smaller eddies will flow over the bubble as over a solid surface. Liquid side mass transfer will take place because of this relative motion of the liquid with respect to the gas bubble. The equation of motion of the gas bubble may be written as

$$\rho V \frac{du_2}{dt} = \rho_2 V \frac{du_1}{dt} + F_{drag}, \tag{21}$$

$$F_{drag} = -k_{f3}\rho_1 S\left(u_2 - u_1\right)^2 = k_{f3}\rho_1 S v_r^2. \tag{22}$$

Equation (21) can be written as

$$\rho_1 V \frac{dv_r}{dt} = \left(\rho_1 - \rho_2\right) V \frac{du_1}{dt} - k_{f3}\rho_1 S v_r^2. \tag{23}$$

The rate of change of relative velocity could be approximated by the expression

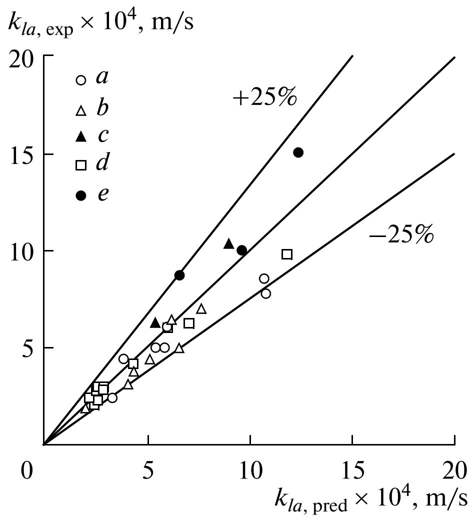

Fig. 5. Comparison of predicted and experimental mass transfer coefficient data: (*a*) nozzle 1, $A_R = 3.16$; (*b*) nozzle 2, $A_R = 5.23$; (*c*) nozzle 3, $A_R = 10.31$; (*d*) nozzle 4, $A_R = 16$; and (*e*) nozzle 5, $A_R = 28.44$.

$$\frac{dv_r}{dt} \cong \frac{v_r}{T_\lambda}, \tag{24}$$

where T_λ is the period of motion of the bubble corresponding to velocity v_r and scale λ. Therefore,

$$T_\lambda = \frac{\lambda}{v_r}, \tag{25}$$

i.e.,

$$\frac{dv_r}{dt} \cong \frac{v_r^2}{\lambda}. \tag{26}$$

Hence the acceleration of eddy can also be approximated by the following expression:

$$\frac{du_1}{dt} \cong \frac{v_\lambda}{T_\lambda} \cong \frac{\lambda}{T_\lambda^2}. \tag{27}$$

Using the dimensional similarity, the velocity and period of an eddy [17] can be represented as

$$v_\lambda = \left(\frac{\varepsilon\lambda}{\rho_1}\right)^{1/3}, \tag{28}$$

$$T_\lambda = \left(\frac{\lambda^2\rho_1}{\varepsilon}\right)^{1/3}. \tag{29}$$

Substituting Eqs. (28) and (29) in Eq. (27), we get

$$\frac{du_1}{dt} = \left(\frac{\varepsilon}{\rho_1}\right)^{2/3}\left(\frac{1}{\lambda^{1/3}}\right). \tag{30}$$

Substituting Eqs. (26) and (27) in Eq. (23) and solving for v_r yields

$$v_r = \frac{[V(\rho_1 - \rho_2)]^{1/2}\,\varepsilon^{1/3}\lambda^{1/3}}{\rho_1^{1/3}[\rho_2 V + K_{f3}\rho_1 S\lambda]^{1/2}}. \tag{31}$$

The scale λ for which the relative velocity is a maximum can be obtained by differentiating Eq. (31):

$$\lambda_{r,\max} = \frac{2\rho_2 V_B}{K_f \rho_1 S}, \tag{32}$$

$$v_{r,\max} = C_1\left(\frac{V\varepsilon}{S}\right)^{1/3}. \tag{33}$$

For a spherical bubble,

$$\frac{V}{S} = \frac{2d}{3}, \tag{34}$$

and hence,

$$v_{r,\max} = C_2(d\varepsilon)^{1/3}. \tag{35}$$

The expression for mass transfer between a spherical bubble and the liquid [17] can be written as

$$J = C_3\left(\frac{D_A v_{r,\max}}{d}\right)^{1/2} d^2 C_{AO}. \tag{36}$$

From which the liquid side mass transfer coefficient can be expressed as

$$k_{la} = \frac{J}{d^2 C_{AO}} = C_3\left(\frac{D_A v_{r,\max}}{d}\right)^{1/2}. \tag{37}$$

Substituting the values of $v_{r,\max}$,max and d_B from Eqs. (33) and (12), respectively, in Eq. (37), we get

$$k_{la} = C_4\varepsilon^{0.3}. \tag{38}$$

Similar to the correlation for interfacial area, by replacing ε with the total irreversible energy input E_t and introducing Calderbank's correction factor for the gas phase turbulence [18], the following expression can be obtained:

$$k_{la} = C_5 E_t^{0.3}\alpha_2^m. \tag{39}$$

The present experimental values of k_{la} are obtained in the range of 2×10^{-4} to 15×10^{-4} m/s, when correlated using the parameters suggested in Eq. (39) with the following constants and indices:

$$k_{la} = 3.38 \times 10^{-4} E_t^{0.385}\alpha_2^{1.38}. \tag{40}$$

The estimated values are compared with that of the experimental data and shown in Fig. 5, and the estimated root-mean-square errors are ±20%.

CONCLUSIONS

Theoretical models based on Kolmogorov's theory of isotropic turbulence [16] and Levich's hydrodynamic derivations [17] were found to correlate the present data on interfacial area and liquid side mass transfer coefficients satisfactorily. These correlations, together with the equations for gas phase holdup α_2 and energy dissipation E_t, can be used for the design of ejectors for gas-liquid contacting. In natural gas processing, the removal of acid gases such as CO_2 is the most important first step to produce commercial natural gas of pipe line or cryogenic quality. Since the viscosity and surface tension ranges studied in the present work are essentially those encountered in amine-carbon dioxide systems, the correlations proposed in the present study should prove useful for the design of cocurrent jet ejectors for the removal of CO_2 from natural gas.

NOTATION

A—cross sectional area, m^2;
A_R—area ratio (A_t/A_n);
a—specific interfacial area, m^2/m^3;
C—constant;
C_{AO}—concentration of solute, mol/m^3;
D—diffusivity, m^2/s;
d—bubble diameter, m;
E_t—specific total energy input, J/m^3;
F_{drag}—drag force, N;
g—acceleration due to gravity, m/s^2;
H—jet velocity head, N/m^2;
h—characteristic bubble dimension, m;
J—flux, mol/(m^2 s);
K—constant;
k—oss coefficient;
k_{la}—mass transfer coefficient, m/s;
m—correction factor in Eqs. (17), (19), and (39);
N—number of bubbles;
P—dynamic pressure, N/m^2;
Q—volumetric flow rate, m^3/s;
r_k—structural parameter;
S—surface area, m^2;
T—period of motion of bubble, s;
u—velocity, m/s;
V—bubble volume, m^3;
V_R—system volume, m^3;
v—eddy velocity, m/s;
α—holdup;
ε—energy per unit mass, J/kg;
λ—eddy scale, m;
μ—viscosity, (N s)/m^2;
p—density, kg/m^3;
σ—surface tension, N/m.

SUBSCRIPTS AND SUPERSCRIPTS

1, 2, k—phases;
B—bubble;
di—diffuser;
exp—experimental value;
f—friction;
m—mixing;
n—nozzle;
pred—predicted value;
t—total;
th—throat.

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