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RESEARCH NOTES

Assessment of the Effects of High-Pressure Operation on the Liquid−Solid Mass-Transfer Coefficient in Trickle-Bed Reactors

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Trickle-bed reactors are used widely in industry and are usually operated at high pressure. All the studies on liquid−solid mass transfer in such reactors were performed under atmospheric pressure and, hence, the empirical correlations for liquid−solid mass-transfer coefficients (k_{ls} and $k_{ls}a$) were developed based on atmospheric data. However, these correlations incorporate one or more of the parameters affected by pressure (e.g., liquid holdup, catalyst wetting efficiency, pressure drop, gas density). In this work the effects of high pressure and high gas flow rates on the predicted coefficients using some of these correlations are evaluated. It is shown that there are discrepancies in the prediction of these correlations, and the use of them at high operating pressure is unjustified. This work is an attempt to bring to the attention of the industrial practitioners the fact that the atmospheric liquid−solid mass-transfer correlations do not exhibit the same trends with increased pressure and some of them do not capture the physics of the system. Thus, experimental investigations to quantify the effect of reactor pressure on k_{ls} and $k_{ls}a$ and a new correlation for a wide range of operating pressures are needed.

Introduction

Trickle-bed reactors are fixed beds of solid catalyst particles contacted by cocurrent downflow of gas and liquid. They are used widely in industrial processes and are usually operated at high pressure (up to about 30 MPa). The rate of mass transfer of the reactants and products from liquid to catalyst particle surface is very important and needs to be accounted for in evaluating trickle-bed reactor performance. Hence, the liquid−solid mass-transfer coefficient (k_{ls}) and the volumetric liquid−solid mass-transfer coefficient ($k_{ls}a$) are important design, scale-up, and operating parameters. Unfortunately, they are difficult to accurately predict *a priori*. Many empirical correlations have been reported in the literature for predicting the liquid−solid mass-transfer coefficient; however, all of the studies were performed at atmospheric pressure and did not account for the high pressure at which these reactors are operated in industry (Lakota and Levec, 1990; Bartelmus, 1989; Latifi *et al.*, 1988; Rao and Drinkenburg, 1985; Delaunay *et al.*, 1982; Tan and Smith, 1982; Yoshikawa *et al.*, 1981; Chou *et al.*, 1979; Satterfield *et al.*, 1978; Specchia *et al.*, 1978; Hirose *et al.*, 1976; Sylvester and Pitayagulsarn, 1975; Lemay *et al.*, 1975; Morita and Smith, 1978; Goto and Smith, 1975, etc.). Recently, it has been found that at high pressure and high gas flow rates, due to an increase in the gas density, the liquid holdup decreases, the pressure drop increases, the catalyst wetting efficiency and the gas−liquid interfacial area increase due to improved liquid spreading across the bed, and the flow regime transition between the trickle and pulsing flow regimes shifts toward higher liquid throughput due to the decrease in liquid holdup (Al-Dahhan and Duduković, 1994, 1995, 1996; Larachi *et al.*, 1991a,b,c; Wammes *et al.*, 1990a,b, 1991a,b). These hydrodynamic parameters influence the overall rate of mass transfer and its coefficients. For instance, liquid holdup and pressure drop affect the intrinsic gas and liquid flow rates through the reactor which, in turn, affect the mass-transfer coefficients, improving catalyst wetting efficiency, increasing the liquid−catalyst contacting area, and hence utilizing more catalyst for liquid-limited reaction systems which improves the reactor performance, etc. However, the quantitative contribution of k_{ls} or $k_{ls}a$ to the reactor performance and the overall conversion depend on the reaction kinetics (liquid- or gas-limited), reaction system type (volatile or nonvolatile), hydrodynamics, bed structure, specifications, etc.

It is noteworthy that, in the developed correlations, the liquid−solid mass-transfer coefficient is correlated in terms of parameters mentioned earlier that are affected by elevated pressures such as dynamic or total liquid holdup (Lakota and Levec, 1990; Rao and Drinkenburg, 1985; Lemay *et al.*, 1975; Ruether *et al.*, 1980; Specchia *et al.*, 1978; Sato *et al.*, 1972; Hirose *et al.*, 1976); power dissipation by the liquid phase per unit mass of liquid holdup (Lemay *et al.*, 1975; Ruether *et al.*, 1980); gas density via gas mass flow (Sylvester and Pitayagulsarn, 1975; Bartelmus, 1989; Chou *et al.*, 1979; Goto and Smith, 1975); and catalyst wetting efficiency (Goto *et al.*, 1975; Delaunay *et al.*, 1982; Chou *et al.*, 1979; Rao and Drinkenburg, 1985; Specchia *et al.*, 1978; Tan and Smith, 1982; Satterfield *et al.*, 1978). Therefore, the liquid−solid mass-transfer coefficients predicted by the aforementioned correlations should be affected by increased reactor pressure. The question is, however, whether the effect of pressure is properly captured since the correlations were developed based on an atmospheric pressure database only.

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Table 1. Selected Correlations for Liquid–Solid Mass-Transfer Coefficients, Developed Based on Data at Atmospheric Pressure[a]

author	correlation	method used to determine k_{ls} and parameters affected by reactor pressure
Lakota and Levec (1990)	$\frac{Sh'}{Sc^{1/3}} = 0.487(Re_l^*)^{0.495}$ flow regimes: trickle, pulse particles: cylinder, 6.1 × 4.7 mm; 5.9 × 4.5 mm bed: i.d. = 17.2 cm; $Z = 32–122.5$ cm; $\epsilon_B = 0.31$	$k_{ls} \propto \frac{1}{h_d}$ method: dissolution of naphthalene in water system: air–water
Rao and Drinkenburg (1985)	$\frac{\eta_{CE} Sh''}{Sc^{1/3}} = 0.24(Re'_l)^{0.75}$ flow regimes: trickle, pulse particles: cylinder, 3 × 3 mm; 6 × 6 mm bed: i.d. = 5 cm; $Z = 240$ cm; $\epsilon_B = 0.35–0.36$	$k_{ls} \propto \frac{1}{h_t \eta_{CE}}$ method: electrochemical system: air–water
Delaunay *et al.* (1982)	$\frac{\eta_{CE} Sh}{Sc^{1/3}} = 1.84 Re_l^{0.48}$ flow regimes: trickle, pulse particles: sphere, 4 mm bed: i.d. = 4.5 cm; $Z = 150$ cm	$k_{ls} \propto \frac{1}{\eta_{CE}}$ method: electrochemical system: air–water
Chou *et al.* (1979)	$\frac{\epsilon_B \eta_{CE} Sh}{Sc^{1/3}} = 0.72 Re_l^{0.54}\, Re_G^{0.16}$ flow regimes: trickle, pulse particles: sphere, 7.8 mm bed: i.d. = 15.2 cm; $Z = 135$ cm; $\epsilon_B = 0.36–0.4$	$k_{ls} \propto \frac{\rho_g}{\eta_{CE}}$ method: electrochemical system: air–water
Lemay *et al.* (1975)	$k_{ls} Sc^{2/3} = 0.20\left(\frac{E_L' \mu_L}{\rho_L}\right)^{0.25}$ flow regimes: pulse particles: sphere, 6.25 mm bed: i.d. = 7.6 cm; $Z = 76.0$ cm	$k_{ls} \propto \frac{\Delta P/Z}{h_t}$ method: dissolution of benzoic acid in water, mixed with dye (rhodamine B) system: air–water

[a] Catalyst wetting efficiency, η_{CE}, is evaluated by Al-Dahhan and Duduković's (1995) correlation for low- to high-pressure operation

$$\eta_{CE} = 1.104 Re''^{1/3}_l \left(\frac{1 + \frac{\Delta P/Z}{\rho_l g}}{Ga_l}\right)^{1/9}$$

In this work, we want to bring to the attention of the industrial practitioners the fact that various liquid–solid mass-transfer correlations may not exhibit the same trends with increased pressure and, hence, clearly some of them do not capture the physics of the system. We will compare the predictions for the liquid–solid mass-transfer coefficients (k_{ls} and $k_{ls}a$) at high-pressure operation by using high-pressure data for the parameters incorporated in these correlations, such as liquid holdup, pressure drop, catalyst wetting efficiency, and gas density. By doing this, we gain insight into this issue and motivate the researchers to investigate experimentally the liquid–solid mass-transfer coefficient at high-pressure operation.

Analysis

In order to demonstrate the effect of operating pressure on liquid–solid mass-transfer coefficients, k_{ls} and $k_{ls}a$ using the current atmospheric correlations (which are the only ones available), five correlations that incorporate the parameters affected by pressure mentioned above have been selected as shown in Table 1 (Lakota and Levec, 1990 ($k_{ls} \propto$ 1/dynamic liquid holdup); Rao and Drinkenburg, 1985 ($k_{ls} \propto$ 1/(total liquid holdup and catalyst wetting efficiency)); Delaunay *et al.*, 1982 ($k_{ls} \propto$ 1/catalyst wetting efficiency); Chou *et al.*, 1979 ($k_{ls} \propto$ gas density/catalyst wetting efficiency); Lemay *et al.*, 1975 ($k_{ls} \propto$ (pressure drop/total liquid holdup)). Lemay *et al.* (1975) studied liquid–solid mass transfer in the pulse flow regime, while the others mentioned above covered the trickle flow regime as well. This is only a sampling of the many correlations available in the literature in which k_{ls} is correlated in terms of one or more of the parameters affected by pressure.

In order to evaluate k_{ls}, at high-pressure operation using these atmospheric correlations (Table 1), the values of liquid holdup, pressure drop, and catalyst wetting efficiency (η_{CE}) at high pressure are required. Al-Dahhan (1993) and Al-Dahhan and Duduković (1994, 1995) studied the effect of high pressure and high gas flow rate on liquid holdup, pressure drop, and catalyst wetting efficiency in the trickle flow regime using water–nitrogen/helium and hexane–nitrogen/helium in beds (inside diameter = 2.2 cm) of spherical ($d_p = 1–1.5$ mm) and cylindrical particles (size = 1.57 × 4.3 mm). In the trickle flow regime, the catalyst particles can be either partially wetted ($\eta_{CE} < 1$) at low liquid mass velocities or fully wetted ($\eta_{CE} = 1$) at high liquid mass velocities. Using their operating conditions and bed characteristics along with the related high-pressure data for liquid holdup and pressure drop and catalyst wetting efficiency evaluated by their high-pressure correlation (the evaluated η_{CE} is between 0.7 and 1.0 at 0.31–3.55 MPa), the liquid–solid mass-transfer coefficient, k_{ls}, can be evaluated by the correlations presented in Table 1. The bed characteristics and fluid superficial velocities and physical properties used to develop these were close to Al-Dahhan's (1993) beds and operating conditions. The volumetric liquid–solid mass-transfer coefficient, $k_{ls}a$, can then be estimated by

$$k_{ls}a = k_{ls}a_t\eta_{CE}$$

Figures 1 and 2 demonstrate the effects of pressure and gas velocity on the liquid–solid mass-transfer coefficient, k_{ls}, in the beds of spherical and cylindrical particles, respectively. In the case of the bed of spherical particles, which are smaller than the cylindrical particles, the values for k_{ls} estimated by the correlations of Lakota and Levec (1990), Chou *et al.* (1979), and Lemay *et al.* (1975) increase at constant liquid mass velocity as pressure and gas velocity increase. This is due to the increase in pressure drop, gas density and velocity, and wetting efficiency and the decrease in the liquid holdup at higher pressure and higher gas velocity. However, both the estimated k_{ls} value and its increase with pressure and gas velocity are different for each correlation. Lemay *et al.*'s (1975) correlation, developed based on atmospheric data obtained in the pulse flow regime, gives a larger increase in k_{ls} with pressure and gas velocity because the correlation incorporates both pressure drop and liquid holdup. The value for k_{ls} estimated by Rao and Drinkenburg's (1982) correlation is essentially unchanged because the decrease in holdup is balanced by the increase in the wetting efficiency, due to the way these parameters are incorporated in the correlation. However, the prediction for k_{ls} of Delaunay *et al.* (1982) decreases with pressure because in their correlation k_{ls} is inversely proportional to the wetting efficiency, and this is the only parameter in their correlation that is affected (increased) by operating pressure.

For the bed of cylindrical particles, the results are the same as those in the bed of spherical particles except for k_{ls} evaluated by the correlations of Rao and Drinkenburg (1985) and Chou *et al.* (1979). Rao and Drinkenburg's correlation shows an increase in k_{ls}; this is because the increase in the wetting efficiency at higher pressure and velocity does not compensate for the decrease in the liquid holdup. Chou *et al.*'s correlation shows a decrease in k_{ls} with pressure and gas velocity. This is because the effect of the increase in the wetting efficiency at higher pressure and high gas velocity would be larger than that of the increase in $Re_G^{0.16}$ (i.e., increase in both the gas velocity and density). One should recall that all the effects of pressure and gas velocity on k_{ls} are due to the ways the different parameters are incorporated into each correlation.

Figures 3 and 4 show the effect of pressure and gas velocity on the volumetric liquid–solid mass-transfer coefficient, $k_{ls}a$, in beds of spherical and cylindrical particles, respectively. The effects of pressure and gas velocity are now greater than those in Figures 1 and 2. This is because both k_{ls} and the liquid–solid contacting area, a, are increasing at higher pressures and high gas velocities. The increase in k_{ls} is due to the reasons given above, while the liquid–solid contacting area increases because of an increase in catalyst wetting efficiency, which is caused by an improvement in liquid spreading at high pressures (Al-Dahhan and Duduković, 1995, 1996). For both beds (spherical particles and cylindrical particles), the correlations of Lakota and Levec (1990), Rao and Drinkenburg (1985), Chou *et al.* (1979), and Lemay *et al.* (1975) show an increase in $k_{ls}a$ with pressure and gas velocity. For the correlations of Rao and Drinkenburg (1985), Delaunay *et al.* (1982), and Chou *et al.* (1979), the effect of η_{CE} on k_{ls} is canceled by the multiplication of the effective interfacial area, a ($a = a_t\eta_{CE}$), when evaluating $k_{ls}a$. This is why Delaunay *et al.*'s correlation shows no dependence on pressure for $k_{ls}a$, while $k_{ls}a$ obtained by Rao and Drinkenburg's and Chou *et al.*'s correlations depend only on liquid holdup and gas Reynolds number, respectively. It is obvious that all correlations predict an increase in k_{ls} and $k_{ls}a$ with liquid mass velocity.

The effect of bed characteristics (particle size, shape, and bed voidage) on k_{ls} can be seen by comparing Figures 1 and 2 with Figures 3 and 4. It is clear that k_{ls} in the bed of spherical particles (smaller diameter) is larger than in the bed of cylindrical particles (larger diameter).

Closing Remarks

It has been shown that there are discrepancies in the evaluations of k_{ls} and $k_{ls}a$ for high-pressure operation using some of the current available correlations. This is also the case for all the other correlations whether they incorporate or not a parameter affected by high-pressure operation. This is because these correlations have been developed empirically based on data at atmospheric pressure. Thus, the use of these correlations for predicting liquid–solid mass-transfer coefficients at high pressure is unjustified. Since the fluid dynamic parameters such as pressure drop, liquid holdup, catalyst wetting efficiency, and gas–liquid interfacial area have been found to be affected by reactor pressure, the liquid–solid mass-transfer coefficient would be affected as well. Accordingly, experimental investigation to determine k_{ls} and $k_{ls}a$ at high-pressure operation is recommended. A new correlation incorporating pressure drop, an easily on-line measurable parameter, rather than the other mentioned parameters such as liquid holdup, catalyst wetting efficiency, etc., is recommended; the phenomenological approach proposed by Al-Dahhan and Dudukovic (1994, 1995) could be useful in this regard.

Acknowledgment

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Notation

A = column cross-sectional area [m^2]
a_t = total external area of particles per unit volume of bed [m^2/m^3], S_pN_p/V_B
a = liquid–solid contacting area, $a = a_t\eta_{CE}$
D = diffusivity of solute in liquid [m^2/s]
d_{eq} = average equivalent particle diameter, defined as 6 times the volume to surface ratio [m]
d_p = particle diameter [m]
d_p' = equivalent particle diameter of a sphere having the same surface area as the particle in question [m]
d_r = reactor diameter [m]
E'_L = liquid power dissipation parameter [W/kg liquid holdup], $(\Delta P/Z)V_L/h_t\rho_L$
g = gravitational acceleration [m^2/s]
G = superficial gas mass velocity [kg/(m^2 s)]
Ga_l = dimensionless liquid Galileo number, $d_p^3\rho^2g\epsilon_B^3/\mu^2(1-\epsilon_B)^3$
h_d = dynamic liquid holdup per unit volume of bed
h_t = total liquid holdup per unit volume of bed
i.d. = reactor inside diameter
k_{ls} = liquid–solid mass-transfer coefficient [m/s]

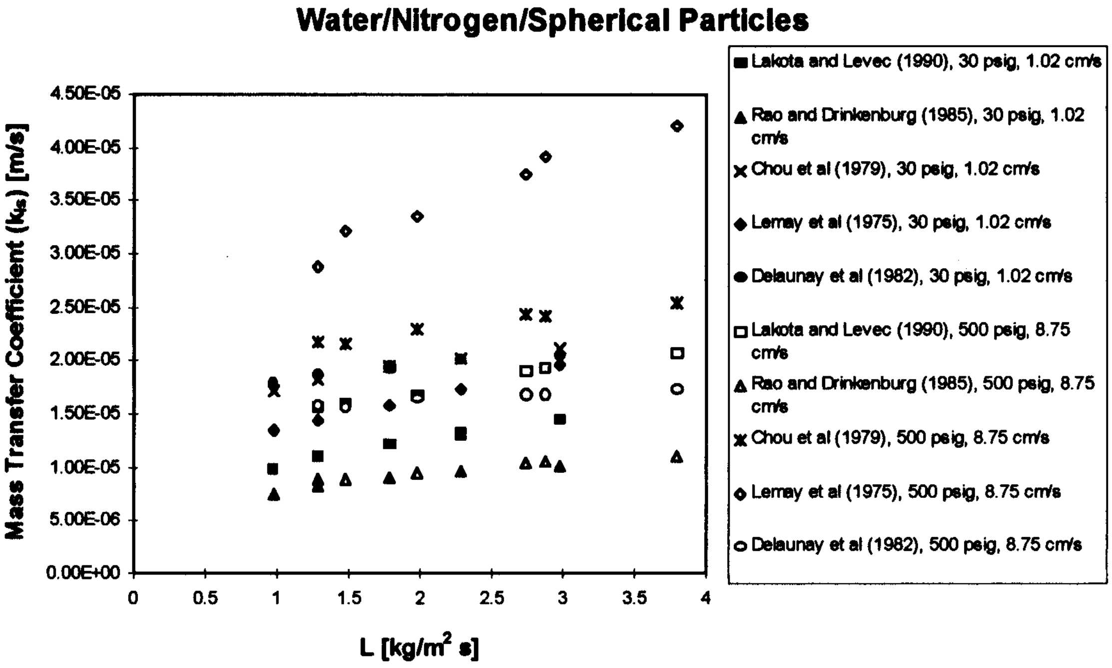

Figure 1. Effect of pressure and gas velocity on the mass-transfer coefficient (k_{ls}) in the bed of spherical particles ($d_p = 1.14$ mm, $\epsilon_B = 0.39$, $d_r = 2.19$ cm). 30 psig = 0.31 MPa; 500 psig = 3.55 MPa.

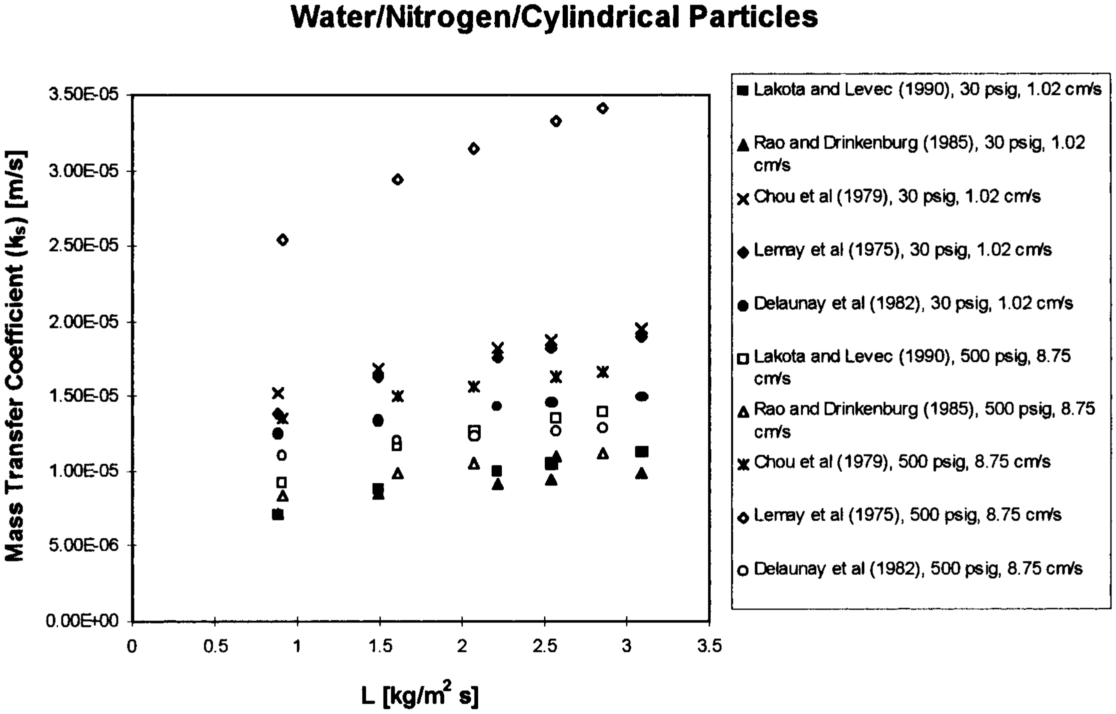

Figure 2. Effect of pressure and gas velocity on the mass-transfer coefficient (k_{ls}) in the bed of cylindrical particles (size = 1.57 × 4.3 cm ($d_{eq} = 1.99$ mm), $\epsilon_B = 0.35$, $d_r = 2.19$ cm). 30 psig = 0.31 MPa; 500 psig = 3.55 MPa.

$k_{ls}a$ = volumetric liquid–solid mass-transfer coefficient [s^{-1}]
L = superficial liquid mass velocity [kg/(m^2 s)]
M_p = total mass of particles
N_p = number of particles, V_t/V_p
$\Delta P/Z$ = pressure drop per unit bed length [N/m^3]
Re_G = gas Reynolds number, Gd_p/μ_G
Re_l = liquid Reynolds number, based on superficial mass velocity, Ld_{eq}/μ_L
Re'_l = modified liquid Reynolds number, $Ld_p'/h_t\mu_L$
Re''_l = modified liquid Reynolds number, $Ld_p/\mu_L(1 - \epsilon_B)$
Re_l^* = modified liquid Reynolds number, based on intrinsic velocity, $[Ld_{eq}/(1 - \epsilon_B)\mu_L]$ (ϵ_B/h_d)
Sc = Schmidt number, $\mu/\rho D$
Sh = Sherwood number, $k_{ls}d_p/D$
Sh' = modified Sherwood number, $(k_s d_p/D)$ $[\epsilon_B/(1 - \epsilon_B)]$
Sh'' = modified Sherwood number, $k_{ls}d_p'/D$
S_p = surface area of one particle [m^2]
V_B = bed volume [m^3]
V_L = superficial liquid velocity [m/s]
V_p = volume of a single particle [m^3]
V_t = total solid volume of particles [m^3], M_p/ρ_p
Z = packed-bed length [m]

Greek Letters

ϵ_B = bed porosity
η_{CE} = external liquid–solid contacting efficiency
μ_G = gas viscosity [kg/(m s)]

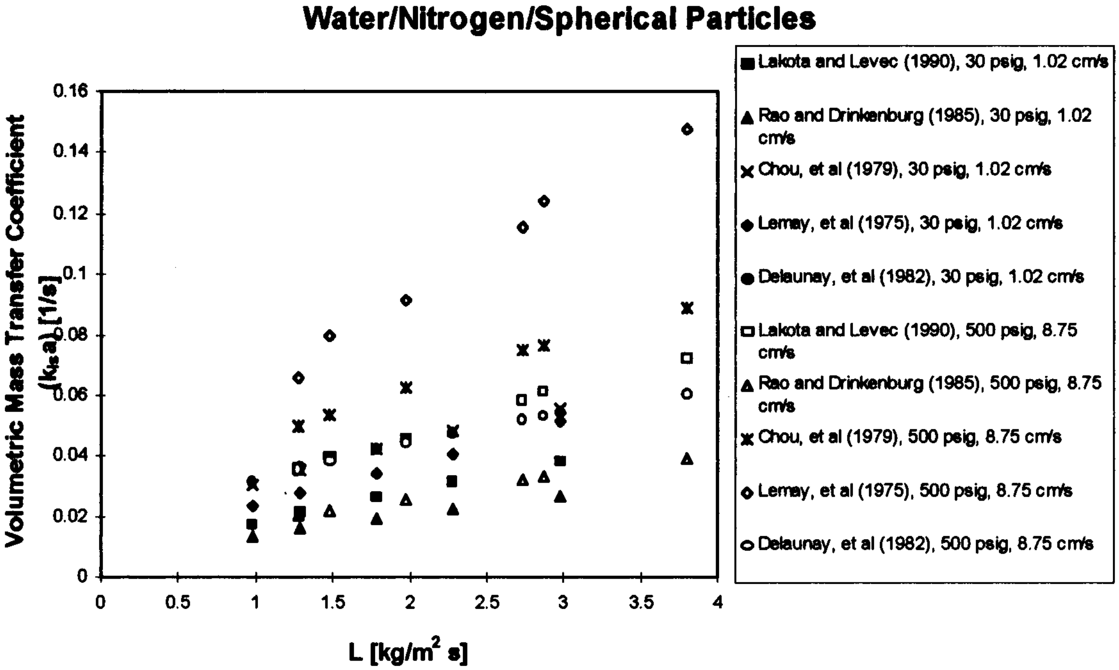

Figure 3. Effect of pressure and gas velocity on the volumetric mass-transfer coefficient ($k_{ls}a$) in the bed of spherical particles ($d_p = 1.14$ mm, $\epsilon_B = 0.39$, $d_r = 2.19$ cm). 30 psig = 0.31 MPa; 500 psig = 3.55 MPa.

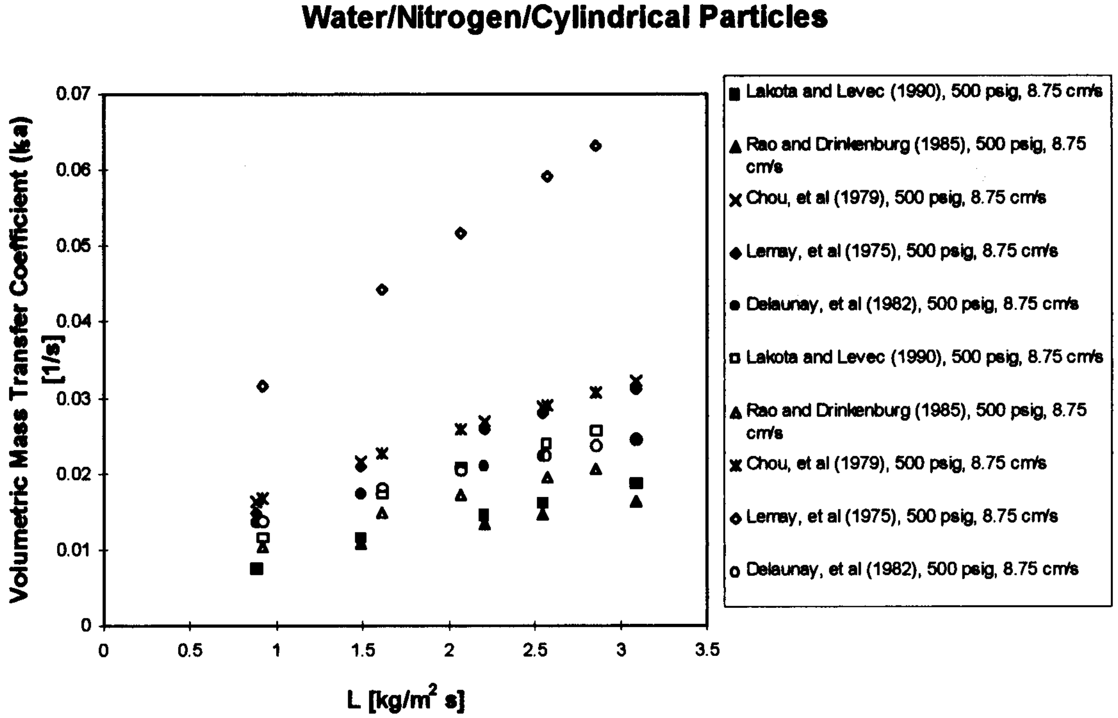

Figure 4. Effect of pressure and gas velocity on the volumetric mass-transfer coefficient ($k_{ls}a$) in the bed of cylindrical particles (size = 1.57 × 4.3 cm (d_{eq} = 1.99 mm), $\epsilon_B = 0.35$, $d_r = 2.19$ cm). 30 psig = 0.31 MPa; 500 psig = 3.55 MPa.

μ_L = liquid viscosity [kg/(m s)]
ρ_L = liquid density [kg/m^3]
ρ_p = particle density [kg/m^3]

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