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Studies on surface to bulk ionic mass transfer in bubble column

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Abstract

Mass-transfer coefficient with immersed surface of diameter 0.016 m placed axially in bubble column (internal diameter = 0.0515 m), was measured using electrochemical technique with ferro-ferricyanide system. Perforated-plate sparger (hole diameter = 0.0015 m) and single-nozzle spargers (hole diameter = 0.001, 0.0015 and 0.002 m) were used. Mass-transfer coefficient was found to be independent of initial bed height. It decreases as axial position of transfer surface from bottom increases at low gas velocities. The effect of type of sparger on mass-transfer coefficient was observed at all axial positions. It increases with increase in hole diameter for single-nozzle sparger. It is higher for perforated-plate distributor than that for single-nozzle sparger with same hole diameter. The present values are in comparison with data of Patil and Sharma [Chem. Eng. Res. Des. 61 (1983) 21]. Following correlation correlated present set of data:

$St_{\mathrm{m}} = 0.057(1-\varepsilon)(Re\,Fr\,Sc^2)^{-0.25}$.

Keywords: Mass transfer; Bubble column; Gas–liquid contactor; Sparging; Electrochemical process; Ferro-ferricyanide system

1. Introduction

Though heat-transfer studies in bubble column have been reviewed by many investigators, only limited studies on surface to bulk mass transfer have been made [1–4]. Such studies have been carried out using electrochemical technique. Patil and Sharma [1] studied surface–liquid mass-transfer coefficient in bubble column by dissolution of copper in acidic solution containing potassium dichromate, in three different bubble columns. Average values of mass-transfer coefficient were found to be higher for column of large diameter than that for column of small diameter. In this study, different spargers were used in different columns. Open area of holes were 0.96, 0.423 and 0.214% for columns of diameter 0.146, 0.380 and 1.00 m, i.e. as the column diameter increased, open area decreased systematically. Therefore, different values of mass-transfer coefficient in various columns may be due to different sparger used. Local values of mass-transfer coefficient were found to be constant up to $U = 0.06\,\mathrm{m\,s^{-1}}$, but significant variation in radial and axial position in bubble column were observed at high gas velocities. Same was observed from mass-transfer studies in sectionalised bubble column [2]. Assuming Chilton–Colburn analogy to hold good, mass-transfer correlation analogous to correlation for heat transfer proposed by Joshi et al. [5] was given as [1]:

$$\frac{kD}{D_{\mathrm{AB}}} = 0.105\left[\frac{D^{1.33}g^{1/3}(U-\varepsilon U_{\mathrm{b}})^{1/3}\rho}{\mu}\right]^{0.8} \times\left(\frac{\mu}{\rho D_{\mathrm{AB}}}\right)^{1/3}\left(\frac{\mu}{\mu_{\mathrm{w}}}\right)^{0.14}. \quad (1)$$

It requires the knowledge of U_{b}, not readily available. Equation analogous to that given by Deckwer et al. [6] in case of heat transfer was also given as [1]:

$$St_{\mathrm{m}} = 0.052(Re\,Fr\,Sc^2)^{-0.25}. \quad (2)$$

Eq. (1) indicates that mass-transfer coefficient is proportional to $D^{0.066}$, which shows weak dependence of k on D. Therefore, in columns of diameter 0.146 and 0.380 m, values of k should be 88 and 94% of values of k in 1.0 m column. The data of Patil and Sharma [1] shows large difference in values of k for different columns. According to Eq. (2), mass-transfer coefficient does not depend on column diameter at all. Therefore, values of k for different column reported by Patil and Sharma [1] may be due to different sparger used.

Zaki et al. [4] studied surface–liquid mass transfer for vertical Ni-coated stainless screens using ferro-ferricyanide system. Screens produced higher rates of mass transfer

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Nomenclature

C_b	bulk concentration of ferricyanide ion ($kg\,m^{-3}$)
C_p	heat capacity of liquid ($W\,s\,kg^{-1}\,K^{-1}$)
d	diameter of orifice in gas sparger (m)
D	column diameter (m)
D_{AB}	diffusivity of active species ($m^2\,s^{-1}$)
F	Faraday's constant ($=9.64867 \times 10^4\,C\,g\,mol^{-1}$)
Fr	Froude number ($=U^2/gD$)
g	acceleration due to gravity ($m\,s^{-2}$)
h	heat-transfer coefficient ($kW\,m^{-2}\,K^{-1}$)
H	settled bed height (m)
H'	expanded bed height (m)
i	limiting current density ($A\,m^{-2}$)
k	mass-transfer coefficient ($m\,s^{-1}$)
n	number of electrons transferred to and from the electrode during reaction
Re	Reynolds number ($=U\rho D/\mu$)
s	number of electrons participating in transfer of n electrons
Sc	Schmidt number ($=\mu/\rho D$)
St_h	Stantan number for heat transfer ($=h/\rho C_p U$)
St_m	Stantan number for mass transfer ($=k/U$)
U	superficial gas velocity ($m\,s^{-1}$)
U_b	bubble rise velocity of single bubble ($m\,s^{-1}$)
Z	axial position measured from the bottom, i.e. from the distributor plate (m)

Greek symbols

ε	gas hold-up
μ	viscosity of liquid ($kg\,m^{-1}\,s^{-1}$)
μ_w	viscosity of water ($kg\,m^{-1}\,s^{-1}$)
ρ	density of liquid ($kg\,m^{-3}$)

than with other geometries. It may be due to the fact that screen wire acts as turbulent promoter. Screen parameters were found to have little effect on mass-transfer coefficient. Eq. (2) with constant 0.052 replaced by 0.095 correlated their data well.

These limited studies show that sparger design could be an important parameter. Kast [7] used single nozzle, sintered nozzle and sieve plate spargers for heat-transfer studies and found it to be independent of sparger type. However, Eq. (1) for mass-transfer coefficient and analogous equation for heat-transfer coefficient [5] indicate that transfer coefficient depends on gas hold-up. Modification of Eq. (2) by Verma [8] for heat transfer as given below also implies such dependence:

$$St_h = 0.12(1-\varepsilon)(Re\,Fr\,Sc^2)^{-0.25}. \quad (3)$$

It is expected, since liquid-circulation velocity, which depends on gas hold-up, is responsible for convective heat transfer. Sparger design has been found to have effect on gas hold-up, though only in "bubbly flow regime" [9]. Therefore, at low values of superficial gas velocity, effect of sparger design on mass-transfer coefficient may be expected. Since gas hold-up varies in axial direction, axial variation of mass-transfer coefficient is also expected.

Electrochemical technique using ferro-ferricyanide system is suitable for measurement of local and instantaneous values of mass-transfer rate and are being used recently to measure local values of solid–liquid mass-transfer coefficient in bubble columns [4], three-phase fixed and fluidised beds [10–12] and three-phase jet loop reactors [13]. The technique was first used by Lin et al. [14] and has been reviewed by Mizushina [15] and Selman and Tobias [16].

In the present work, effect of sparger design and axial position of transfer surface on bulk surface mass-transfer coefficient was studied using electrochemical technique with ferro-ferricyanide system.

2. Experimental set-up

The schematic diagram of the experimental set-up is presented in Fig. 1. Two Perspex columns with internal diameter = 0.0515 m and height = 1.52 m were used. In the first column, perforated plate having 1, 8, 13, 18 and 19 holes arranged on circles of radius 0.0, 0.008, 0.011, 0.016 and 0.021 m, respectively, was used as gas sparger. It had total 58 holes of diameter 0.0015 m. This perforated plate was placed at a distance of 0.1 m above from bottom plate, which had an air inlet at the centre and a drain at its side. The portion between the bottom plate and distributor served as calming section. In the second column, three different single nozzle with hole diameter = 0.001, 0.0015

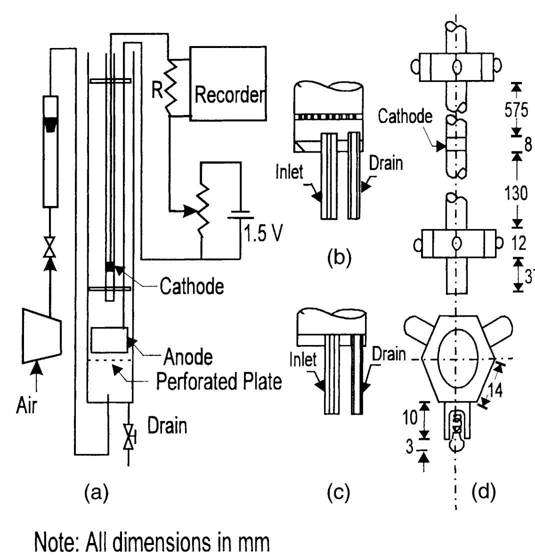

Fig. 1. Schematic diagram of the experimental set-up: (a) bubble column and electrical circuit; (b) perforated-plate sparger; (c) single-nozzle sparger; (d) mass-transfer probe and support.

Table 1
Properties of the electrolytic solution

Concentration of ethylene glycol (wt.%)	ρ ($kg\,m^{-3}$)	μ ($\times 10^3\,kg\,m^{-1}\,s^{-1}$)	σ ($N\,m^{-1}$)	D ($\times 10^{10}\,m^2\,s^{-1}$)	Sc
0.00	1018.8	59	53	13.00	445
0.075	1024.8	72	50	10.63	661
0.150	1035.4	87	40	8.79	956

and 0.002 m were used. A drain was also provided at its side. After completion of all runs with single nozzle, bottom plate was replaced with another having single nozzle of different diameter. Flow rate of air supplied through a compressor was measured by rotameter.

Ni cathode ring of width 0.008 m, fixed at a distance of 0.167 m from the lower end of an acrylic tube of outer diameter 0.016 m. Wire connecting cathode to electrical circuit was taken out through annular space in the tube. The bottom of the tube was closed so that no liquid could enter into it. Ni cathode served as mass-transfer surface. The tube was placed at centre of the column and could be moved in vertical direction so that the transfer surface is positioned at any desirable distance above the gas sparger. To avoid vibrations of the probe due to vigorous bubbling, two three-legged supports at a distance of 0.037 and 0.672 m from the lower end of the probe were provided. One of the legs in each support was telescopic and spring loaded to give better support to the immersed tube. A Ni ring of width 0.026 and diameter 0.051 m and was placed near the gas sparger. This very large anode worked as reference electrode.

The voltage supplied by 1.5 V dry cell (6 I Eveready) was varied using a potentiometer. The current was measured with a multimeter, as potential drop across a standard resistance and recorded with Omniscribe strip chart recorder.

The electrochemical technique with ferro-ferricyanide system with NaOH as inert (0.005 M $K_3Fe(CN)_6$, 0.005 M $K_4Fe(CN)_6$ and 0.5 M NaOH) has been used. Ethylene glycol was added to change Schmidt number. Properties of the electrolytic solution is given in Table 1.

3. Experimental procedure

Gas hold-up (ε) was obtained from expanded and settled bed heights as:

$$\varepsilon = \frac{H' - H}{H'}. \quad (4)$$

The values of H and H' were noted for each run along with mass-transfer values.

Dissolved oxygen can lead to appreciable error in mass-transfer measurement [16]. Therefore, nitrogen was bubbled for 10 min before and after each run to remove it. The solution was kept in dark when not in use to avoid poisoning in presence of sunlight [16] and poisoning of Ni electrodes due to presence of cyanide ions. After each run the cathode and anode was buffed with soft tissue paper and washed with distilled water to remove any cyanide ion. Same cleaning method was followed before each run. The mass-transfer probe was washed by operating the column with distilled water. Even after these precautions the solution could last for 2 days only. The concentration of ferricyanide ion was determined by titration. Diffusivity of the ferricyanide ion was obtained from correlation given by Eisenberg et al. (1956) (referred by Selman and Tobias [16]).

Mass-transfer probe was placed in the column at a particular height from the bottom. Limiting current plateau was checked before and after each run. Voltage was applied and current as potential drop across 5 Ω standard resistance was recorded. From fluctuating current, 30 data points at an interval of 0.5 s were used to determine average limiting current (i). The mass-transfer coefficient was determined as:

$$k = \frac{si}{nFC_b}. \quad (5)$$

4. Results and discussion

Gas hold-up values for the electrolytic solution as a function of superficial gas velocity (U) at $H = 0.8$ m and for single-nozzle sparger with $d = 0.001$, 0.0015 and 0.002 m and for perforated-plate distributor at $Sc = 445$, 661 and 996 are shown in Fig. 2. Gas hold-up increases as Sc increases. Gas hold-up for single-nozzle sparger with $d = 0.001$ m at $Sc = 445$ is consistently lower than that for

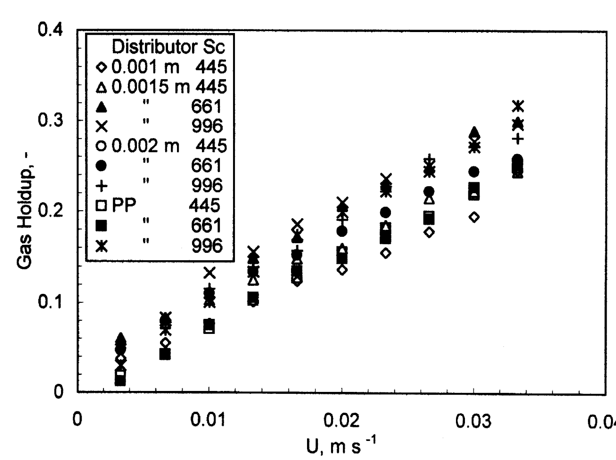

Fig. 2. Effect of concentration of aqueous solution of NaOH on gas hold-up.

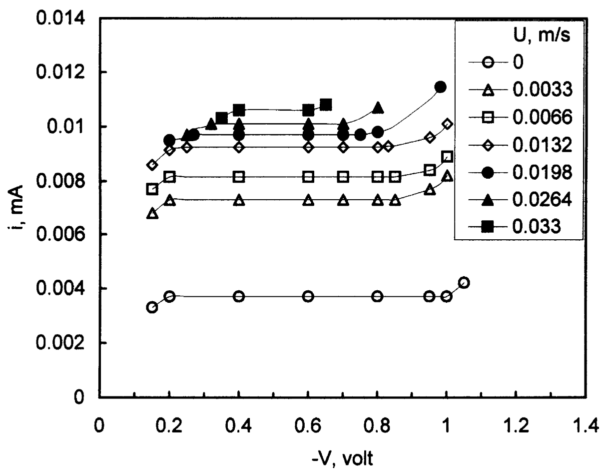

Fig. 3. Limiting current curves.

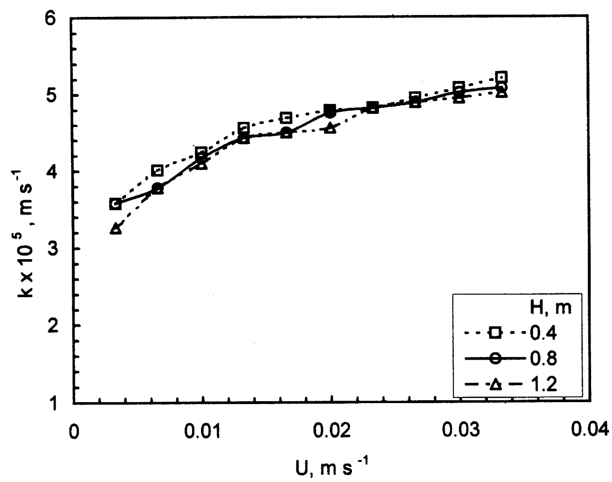

Fig. 4. Variation of k as a function of U for perforated-plate distributor with $d = 0.002$ m at $Z = 0.39$ m and $H = 0.4$, 0.8 and 1.2 m.

other spargers for $U > 0.0167\,\mathrm{m\,s^{-1}}$. Values of gas hold-up for single-nozzle sparger with $d = 0.002$ m are consistently higher than that for other spargers. The trend is in contradiction to the finding that gas hold-up increases as nozzle diameter decreases [17]. Visual observations revealed that large values of gas hold-up are due to accumulation of foam near the top for single-nozzle sparger with $d = 0.002$ m. This foam was composed of small bubbles. The slugs were formed at $Z > 0.4$ m for single-nozzle sparger and $Z > 0.65$ m for perforated-plate sparger. Slug formation took place only for $U > 0.016\,\mathrm{m\,s^{-1}}$.

Limiting current curves obtained at different air flow rates is presented in Fig. 3. The plateau was observed at zero air flow rate over a range of overvoltages of −0.2 to −0.9 V. As the air flow rate increases, the range of overvoltages, in which the plateau was observed, is reduced. The plateau is well defined even for the maximum value of air flow rate covered in the present study. Accordingly, 0.15 V was applied during entire study.

4.1. Initial bed height

Mass-transfer coefficient (k) as a function of U with perforated-plate distributor and single-nozzle sparger with $d = 0.002$ m at an axial distance above the distributor plate, $Z = 0.39$ m, and at three different initial bed heights, $H = 0.4$, 0.8 and 1.2 m, is shown in Fig. 4. The values of k increased monotonically with increase in U in all the cases. It was observed even for $U > 0.01\,\mathrm{m\,s^{-1}}$ at which heat-transfer coefficient has been reported to become constant [6]. The values of k at all H are in close agreement indicating that it is independent of initial bed height.

4.2. Sparger design

Values of k as a function of U at $Z = 0.39$ m and with $H = 0.8$ m is shown in Fig. 5. Data for single-nozzle sparger with $d = 0.001$, 0.0015 and 0.002 m and perforated-plate distributor are presented. The values of k with perforated-plate distributor is lower than that with single-nozzle sparger with $d = 0.002$ m by about 10% at $U \leq 0.0233\,\mathrm{m\,s^{-1}}$. At $U > 0.0233\,\mathrm{m\,s^{-1}}$, values of k are in close agreement. It is about 20% higher for perforated-plate distributor than that for single-nozzle sparger for $d = 0.001$, 0.0015 m at $U < 0.0267\,\mathrm{m\,s^{-1}}$. Values of k for single-nozzle sparger with $d = 0.002$ m is about 30% more than that for other single-nozzle spargers up to $U = 0.027\,\mathrm{m\,s^{-1}}$. For $U > 0.027\,\mathrm{m\,s^{-1}}$ difference between values of k for single-nozzle sparger with $d = 0.001$ and 0.002 m, and perforated-plate distributor are in close agreement. It shows that at high gas velocity there is no or little effect of sparger type on mass-transfer coefficient. Such trend is expected, as Reilly et al. [9] did not observe any effect of sparger design on gas hold-up. Heat-transfer coefficient has also been shown to be independent of sparger

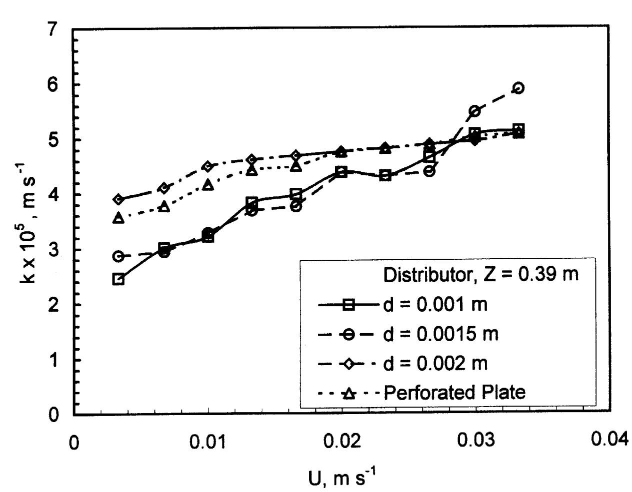

Fig. 5. Values of k as a function of U at $Z = 0.39$ m and $H = 0.8$ m at various sparger types.

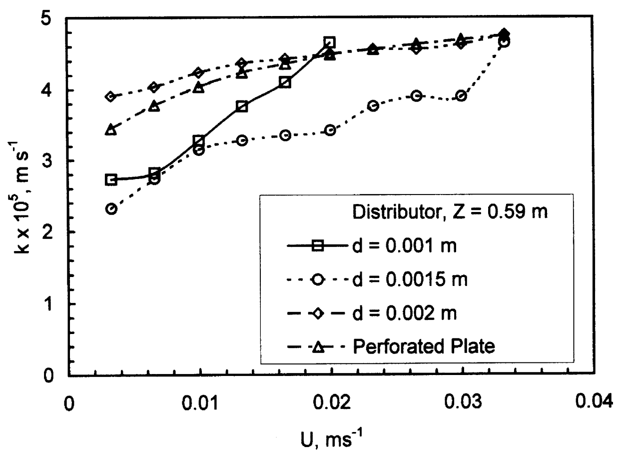

Fig. 6. Values of k as a function of U at $Z = 0.59$ m and $H = 0.8$ m at various sparger types.

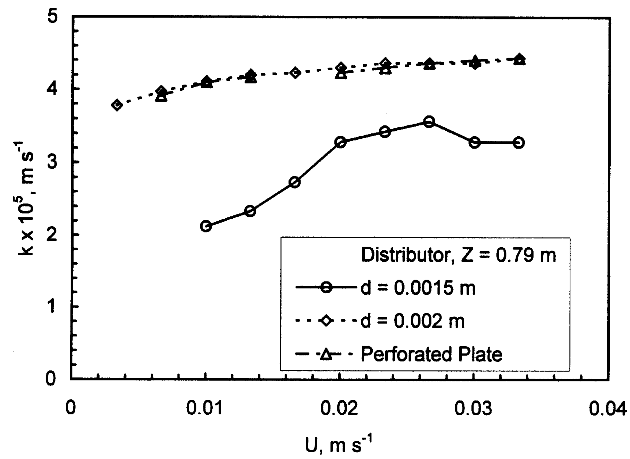

Fig. 7. Values of k as a function of U at $Z = 0.79$ m and $H = 0.8$ m at various sparger types.

design [7,18]. At low gas velocity, no slugs were observed in the lower section of column. Hence, it is not surprising if an effect of sparger design is observed, as the probe was has been placed in lower portion of the column.

Values of k as a function of U at $Z = 0.59$ m and with $H = 0.8$ m with perforated-plate distributor and single-nozzle sparger with $d = 0.001$, 0.0015 and 0.002 m is shown in Fig. 6. Values of k with perforated-plate distributor is about 10% lower than that for single-nozzle sparger with $d = 0.002$ m for $U < 0.01\,\mathrm{m\,s^{-1}}$ and are in close agreement with each other for $U \geq 0.01\,\mathrm{m\,s^{-1}}$. For single-nozzle sparger with $d = 0.0015$ m, the values of k are about 15–20% lower than that with single-nozzle sparger with $d = 0.001$ m. At bed height of 0.39 m, slugs are absent. At bed height of 0.59 m, slugs formation starts. Therefore, effect of sparger design on mass-transfer coefficient at low values of U is expected. At high values of U, mass-transfer coefficient for all spargers are in agreement within experimental error. The formation of more slugs may be responsible for this behaviour.

Values of k as a function of U with perforated-plate distributor and single-nozzle sparger with $d = 0.0015$ and 0.002 m in upper section of the column at $Z = 0.79$ m and $H = 0.8$ m is shown in Fig. 7. The values of k with perforated-plate distributor and single-nozzle sparger with $d = 0.002$ m are in close agreement. However, values of k for single-nozzle sparger with $d = 0.0015$ m are about 45% lower than that with single-nozzle sparger with $d = 0.002$ m at low values of U and the difference decreases at high values of U. Mass-transfer coefficient seems to attain a maximum value for all spargers at $U \geq 0.02\,\mathrm{m\,s^{-1}}$.

Thus, at all axial positions, effect of sparger type on k is observed. It increases with increase in d for single-nozzle sparger. It is also higher for perforated-plate distributor than that with single-nozzle sparger with same d (i.e. $d = 0.0015$ m). The values of $(1 - \varepsilon)$ with single-nozzle sparger decreases with increase in d. This increase is about 10–20%. It could be a possible reason for high values of mass-transfer coefficient with $d = 0.001$ and 0.002 m as reported earlier [8]. Thus, it seems that gas hold-up may be an appropriate variable accounting for the dependence of k on bubble properties.

4.3. Height of transfer surface

The values of k as a function of Z with perforated-plate distributor and $H = 0.8$ m at various U are shown in Fig. 8. It is clear that k decreases systematically with increase in Z for $U > 0.01\,\mathrm{m\,s^{-1}}$. At $U \leq 0.01\,\mathrm{m\,s^{-1}}$ the variation is within experimental error. The decrease is more sharp at high gas velocities. It is expected, as bubble size and number of bubbles varies axially. At high values of Z slugs are formed and hence variation of k with U is not very prominent.

Present data is compared with that of Patil and Sharma [1] in Fig. 9. Experimental data of gas hold-up for each run was used to calculate values of present set of data. To plot data of Patil and Sharma [1], computed values of gas hold-up reported in their work [1] has been used. Present

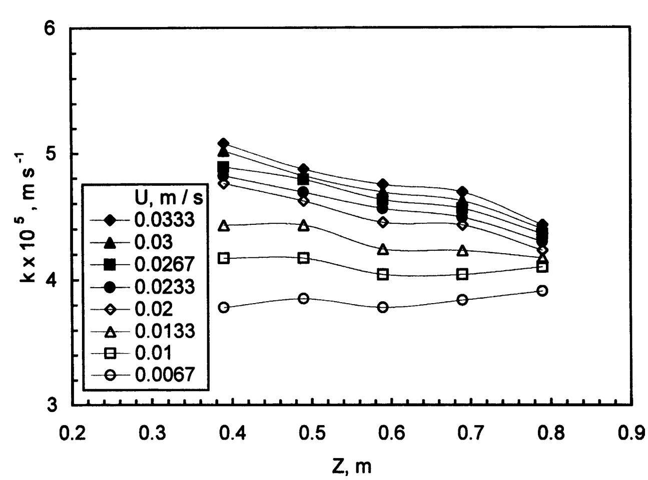

Fig. 8. Values of k as function of Z with perforated-plate distributor and $H = 0.8$ m at various U.

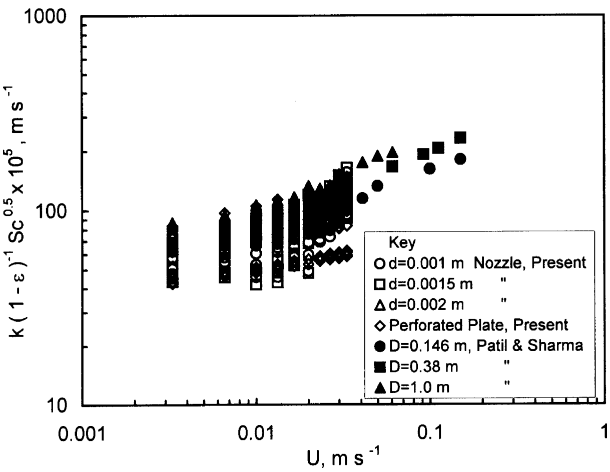

Fig. 9. Comparison of present data with data by Patil and Sharma [1].

data are in comparison with their data as the slopes of the lines and the values of k are in comparison with each other in the overlapping region as well. The scatter in the present data taken on single column are more than that of Patil and Sharma [1] taken with three columns of different size, indicating that sparger design may be more important than column diameter. Patil and Sharma [1] used Eqs. (1) and (2) to correlate mass-transfer coefficient. As discussed earlier, effect of Z on k is observed at large values of U. Also near the top, the effect of U on k is less. It results in large deviations in the values of k. Though the effect of sparger is taken into account by the term $(1 - \varepsilon)$. However, it serves the purpose partially. Use of local gas hold-up may be a more appropriate term. Thus, axial variation of gas hold-up may be important parameter instead of average gas hold-up.

The present set of data are correlated by the following correlation:

$$St_m = 0.057(1 - \varepsilon)(Re\,Fr\,Sc^2)^{-0.25}. \quad (6)$$

The constant 0.057 is within 6.1% for all spargers used. However, scatter is more for perforated-plate distributor. The parameter Z could not be taken into account.

5. Conclusion

Electrochemical technique can be used to measure transient mass-transfer coefficient in bubble columns. Average mass-transfer coefficient increases monotonically with increase in U, even for $U > 0.01\,\mathrm{m\,s^{-1}}$. It is independent of initial bed height and decreases with increase in height above the distributor plate. It is marginally higher for those sparger for which the gas hold-up is high, i.e. high values of k for perforated-plate distributor was observed. For single-nozzle sparger it increased with decrease in hole diameter.

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