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Wall-to-bed mass transfer in a three-phase fluidized bed with coaxially placed string of spheres internal

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Abstract

Turbulent promoters are generally used to realize enhancement in heat and mass transfer rates. With a view to exploit this advantage for the favor of obtaining magnitudes of improvements in mass transfer rates in electrochemical processes, in the present study experiments were conducted in three phase fluidized bed electrochemical cells in the presence of coaxially placed string of spheres. Diffusion controlled electrode reactions were employed to obtain limiting current densities, and hence, mass transfer coefficients. An equimolar potassium ferrocyanide and potassium ferricyanide solution in the presence of sodium hydroxide supporting electrolyte was used as the liquid phase. Glass spheres of different sizes were employed as fluidizing solids and nitrogen was used as the inert gas phase. In three phase fluidized bed, mass transfer coefficient increased with increasing sphere diameter and particle diameter. The parametric effect of gas and liquid velocities on mass transfer coefficient was found to be marginal. The mass transfer coefficient was found to decrease with increase in pitch. A correlation to predict mass transfer coefficient has been developed by using least squares regression analysis.

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Keywords: Fluidization; mass transfer coefficient; augmentation; turbulent promoter

1. Introduction

Three-phase fluidized beds provide adequate contact between the phases. The principal advantages are high macro mixing, good axial dispersion, high reactant conversions, mixed flow patterns, temperature uniformity,

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Nomenclature

A	area of the reacting surface, [m^2]
C_0	concentration of reacting ion, [kmol/m^3]
D_c	diameter of the column, [m]
d_p	particle diameter, [m]
d_b	sphere diameter, [m]
d_r	rod diameter, [m]
F	Faraday constant, [C/mol of electrons]
g	acceleration due to gravity, [m/s^2]
i_L	limiting current, [A]
k_L	mass transfer coefficient, [m/s]
p	pitch, [m]
U_g	superficial gas velocity, [m/s]
U_L	superficial liquid velocity, [m/s]
X	longitudinal distance, [m]
n	number of electrons released or consumed per ion during the reaction, [-]

Greek Symbols

ε_g	gas holdup, [-]
ε_L	liquid holdup, [-]
ε	bed porosity, [-]
μ_L	liquid viscosity, [kg/m.s]
ρ_L	liquid density, [kg/m^3]

Dimensionless groups

Fr_g	Froude number based on gas velocity = $\frac{U_g^2}{gd_p}$
j_D	Coulburn j-factor = $\frac{k_L}{U_L}Sc^{2/3}$
Re_p	Reynolds number based on particle diameter = $\frac{\rho_L d_p U_L}{\mu_L}$
Sc	Schmidt number = $\frac{\mu_L}{\rho_L D_L}$

elimination of hot regions, significant enhancement in heat transfer, low intra particle diffusion resistance, low external liquid-solid mass transfer resistance, ease in catalyst replacement, good controllability of catalyst activity etc. Accounting all the advantages mentioned above, the three-phase fluidized beds find large number of applications in the process industry [1]. Application of three-phase fluidization to electrochemical processes gained attention in recent times. The presence of internals in process equipment often becomes inevitable, either due to arrangement of baffles, immersion heaters, draft tubes, geometrical irregularities etc., or due to the placing of turbulent promoters with a purpose of enhancing heat and mass transfer rates. As a consequence the presence of internals increases the complexity of bed hydrodynamics and transport phenomena.

Literature survey revealed that a few studies were taken up earlier in this direction [2-4]. Internal elements such as cross-flow elements [5], helicoidal tape turbulent promoter elements [6], disc promoters [7] and twisted tapes [8] were used previously in three-phase fluidized beds. Among several turbulent promoters, a string of spheres on a rod, which was arranged coaxially in the electrochemical cell, appeared to yield good enhancements in mass transfer coefficient in homogeneous flow and liquid fluidized beds [9]. This promoter was chosen for the present study because the fabrication of the promoter assembly is simple and easy and the enhancements in mass transfer were very high in homogeneous flow and liquid-fluidized beds. In view of these observations, in the present study investigations were carried out using string of spheres as a promoter element for obtaining magnitudes of improvements in wall-liquid mass transfer in three-phase fluidized beds. The flow system selected for the present study comprises of a fluid electrolyte as liquid phase, glass spheres as solid phase and nitrogen as gas phase. Limiting current technique is used to obtain mass transfer coefficients. The electrochemical system chosen for the present investigation belongs to ferricyanide-ferrocyanide redox system. The electrode reactions involved are:

From the measured limiting current, mass transfer coefficient is calculated using the equation

$$k_L = \frac{i_L}{nAFC_0} \qquad \dots (1)$$

2. Experimental

The experimental setup employed in the present investigation is similar to the unit employed by Subramanyam et al [8]. Due to page number restriction in the present journal, this reference is provided for detailed description of the setup. The turbulent promoter in the present cse comprises of an array of spheres of uniform diameter fixed at a given pitch on a stainless steel rod. The circuit employed for the measurement of limiting current essentially consisted of a six volt D.C source, an ammeter, a voltmeter, a rheostat and a commutator.

Freshly prepared very pure distilled water was used to make about 70 liters of equimolar solution of potassium ferrocyanide and potassium ferricyanide each of 0.01 N in the presence of 0.5 N sodium hydroxide from analytical grade reagents. The solution was deaerated by nitrogen. The electrolyte and the nitrogen gas were metered and sent through the experimental column. During each run estimation of the concentrations of the species is made by volumetric analysis [10].

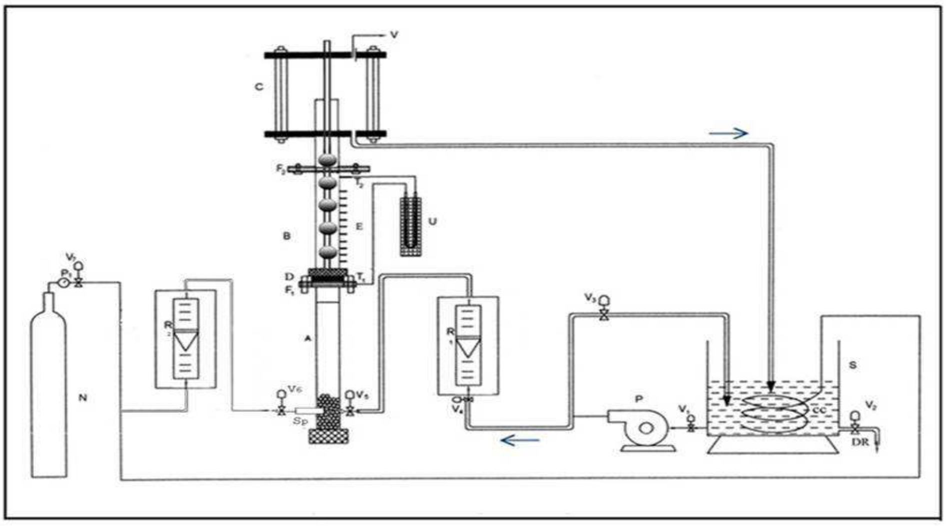

Fig.1: Schematic diagram of experimental setup

A – Entrance calming section, B – Test section, C – Exit section, CC – Copper coil, D – Fluid distributor, D_R – Drain, F_1 and F_2 – Flanges, N – Nitrogen gas cylinder, P – Pump, P1 – Pressure gauge, R_1 and R_2 - Rotameters, S – Storage tank, Sp – Sparger, T_1 and T_2 – Pressure taps, U – U tube manometer, V – Vent in the exit section, V_1 to V_7 – Valves.

3. Results and discussion

Fig. 2 gives the data of the present study plotted as mass transfer coefficient k_L against liquid velocity U_L for five cases of (i) homogeneous liquid flow (Plot A), (ii) gas-liquid upflow bubble column (Plot B), (iii) gas-liquid up flow bubble column in the presence of coaxial string of spheres as promoter assembly (Plot C), (iv) gas-liquid fluidized bed without any promoter internal (Plot D) and (v) gas-liquid fluidized bed in the presence of coaxial string of spheres as promoter assembly.

The magnitudes of improvements over homogeneous liquid flow were shown through plots B, C, D and E. Plot A is the mass transfer coefficient data predicted from Lin et al [11] for the case of homogeneous liquid flow (empty conduit flow), while plot B is the present experimental mass transfer coefficient data obtained in gas-liquid upflow bubble column without any promoter internal. The present data on two-phase bubble column with the string of spheres is shown as Plot C. Plot D shows the data in three-phase fluidized bed without at internal whereas Plot E represents the mass transfer coefficient data in the presence of coaxial string of spheres as promoter internal

All the above data in bubble columns and three-phase fluidized beds were obtained at a constant superficial gas velocity $U_g = 0.0234$ m/s. Plots A and B show that the improvements in the mass transfer coefficient due the introduction of gas phase into homogeneous flow were up to a maximum of 8.7 fold. The improvements due to the addition of promoter in a gas-liquid bubble column were up to 10 percent over that without using a promoter (Plots C and B). The addition of fluidizing solids to the system represented by Plot B which is otherwise gas-liquid upflow bubble column resulted in the augmentation in mass transfer coefficient by 14 percent (Plots B and D). Introduction of the coaxial string of spheres into the three-phase fluidized enhanced the mass transfer coefficient by 20 percent (Plots D and E). The addition of fluidizing solids to the system represented by Plot C resulted in the augmentation in mass transfer coefficient by 22 percent (Plots C and E). The plots revealed that highest mass transfer coefficient could be realized in a three-phase fluidized bed in the presence of coaxial string of spheres as turbulent promoter.

The coaxial string of spheres on a rod when used as the turbulent promoter in a three-phase fluidized bed was found to show an augmentation in the mass transfer coefficient up to a maximum of 12 fold in the lower velocity range over that of the empty conduit flow (Plots E and A).

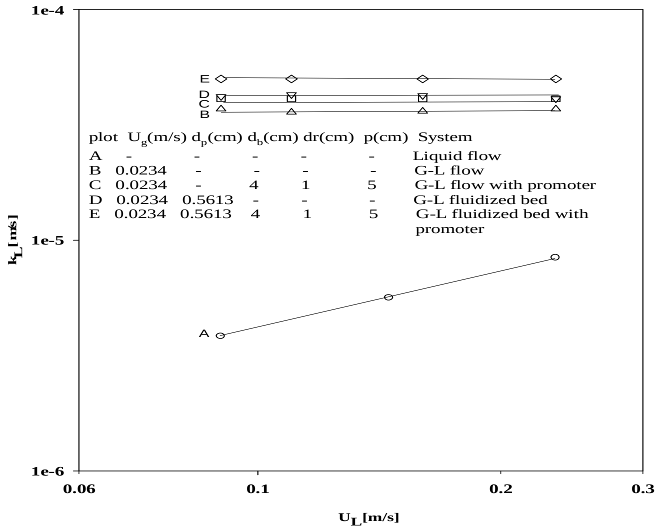

Fig.2: Comparison of k_L values in different flow systems

These observations indicate that the simultaneous introduction of gas phase, fluidizing solids and turbulent promoter into empty conduit flow is definitely advantageous as they enhance turbulence resulting in increased mass transfer coefficient. Moreover, a reduced hydraulic mean diameter will result when a string of spheres on a rod is inserted coaxially in the test section. The reduced hydraulic mean diameter causes an increase in the flow velocities of the fluids in addition to imparting radial component to the flowing fluids. This radial component superimposed on the axial flow moves towards the confining wall resulting in a tractive shear along the wall over which the electrochemical redox reactions are taking place.

3.1. Longitudinal variation of mass transfer coefficient

Initially in the three phase fluidized beds with string of spheres promoter, mass transfer coefficient data were obtained at each of the 34 electrodes. These data have been shown in Fig.3 for one specific promoter. The promoter was arranged in such a way that, the maximum diameter normal to the flow direction is aligned exactly in line with an electrode. So that the variation in mass transfer coefficient from this electrode to next such electrode, the distance between them is equal to pitches, has been measured and shown in Fig.3 for the pitch value of 15cm.

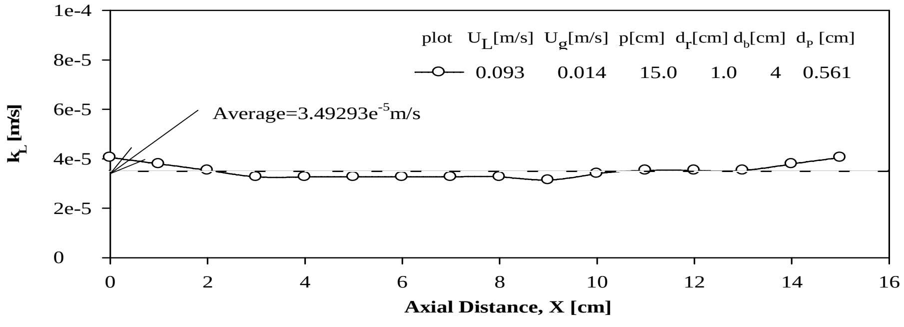

Fig.3. Longitudinal variation of mass transfer coefficient

An inspection of the plot of this graph indicates that the mass transfer coefficient exhibited fluctuations and the average value is also shown by dashed lines. This observation reveals that the mass transfer coefficient varies along the axial direction because of varied turbulent intensity. Similar observations were noticed for other pitch values viz., 5, 7 and 10 cm employed in the present study (graphs not shown). As the fluid electrolyte passes along the axial direction, it is subjected to contraction and expansion of flow area and hence the turbulence intensity varies. This greatly affects resistance film on the electrode surface. Hence a longitudinal variation is noticed. Therefore all the 34 electrodes were joined together and limiting current data at this combined electrode were obtained for all subsequent runs. Average mass transfer coefficient value has been obtained from these limiting current values.

3.2. Effect of gas and liquid velocities

The mass transfer coefficient data in a three-phase fluidized bed in the presence of coaxially placed string of spheres {$p = 5.0$ cm, $d_r = 1.0$ cm, $d_b = 4.0$ cm, $d_p = 5.613$ mm} for three superficial liquid velocities 0.094, 0.122 and 0.150 m/s were plotted against superficial gas velocity and shown in Fig.4. Similarly, in Fig.5 the mass transfer coefficient data were plotted against liquid velocities to three superficial gas velocities 0.014, 0.0234 and 0.0374 m/s. A close examination of the pots of these two figures revealed that the influence of liquid and gas velocities on mass transfer coefficient appears to be insignificant and very marginal. The reason for this can be attributed to the relative insignificant contribution to the turbulence generated by liquid and gas velocities when compared to the total turbulence generated in the bed. Although marginal effect of liquid and gas velocities could be seen, their contribution to the combined effect seems to be negligible. Hence no remarkable influence of gas and liquid velocities on mass transfer coefficient could be observed.

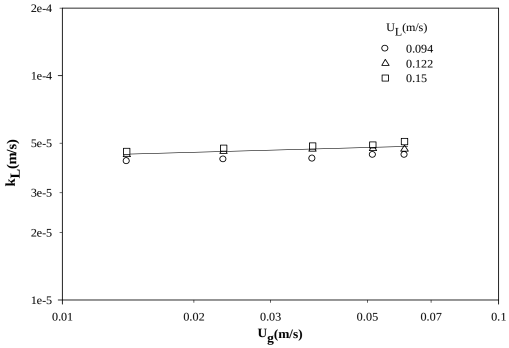

Fig.4: Effect of liquid velocity (U_L) on k_L at constant $d_r = 1$ cm, $d_b = 4$ cm, $p = 5$ cm, $d_p = 5.613$ mm

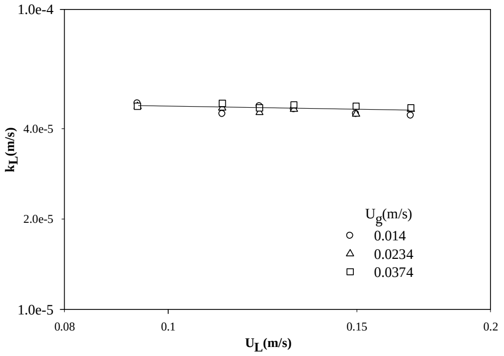

Fig.5 : Effect of gas velocity (U_g) on k_L at constant $d_r= 1$ cm, $d_b= 4$ cm, $p= 5$ cm, $d_p= 5.613$ mm

3.3. Effect of pitch on mass transfer coefficient

Fig.6 shows the mass transfer coefficient data plotted against liquid velocity for the case of coaxially placed string of spheres as an internal {$d_r = 1$ cm, $d_b = 4.0$ cm, $d_p = 5.613$ mm} for the fixed gas velocity of 0.0234 m/s. Three different pitch values were considered. An examination of the plots indicates that the pitch had shown significant influence on the mass transfer coefficient. An increase in pitch value resulted in a decrease in mass transfer coefficient.

The reason for this behavior can be explained as follows. As the liquid which is the continuous phase in the present system, passes over the string of spheres (between trailing edge of the sphere and the leading edge of the next of sphere) in the flow path of the fluid, formation of wakes occur which spread over the entire space between two successive spheres. These wakes move around in the bulk liquid and generate churning action. As the pitch is increased, the number of such wakes decreases and hence the resulting turbulent intensity decreases. Hence, with increase in pitch decrease in mass transfer coefficient would be realized. This trend is also clearly seen from inset Fig. 6a.

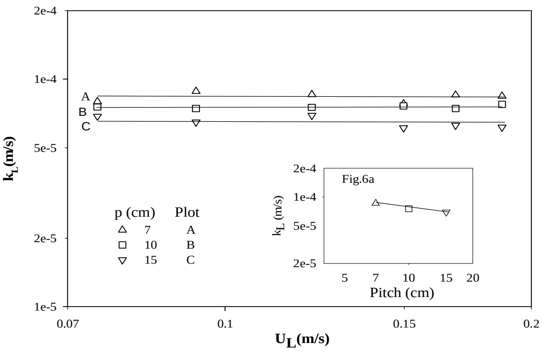

Fig.6 : Effect of pitch (p) on k_L at constant U_g=0.0234 m/s, d_r= 1cm, d_b=4 cm, d_p= 5.613 mm

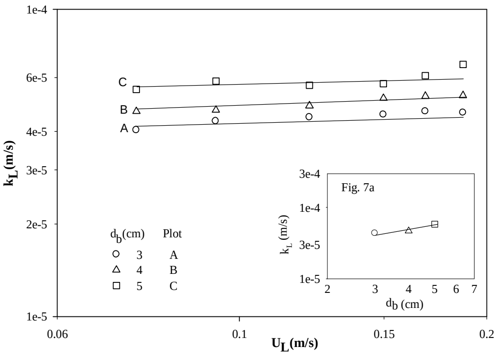

Fig.7 : Effect of sphere diameter (d_b) on k_L at constant U_g= 0.0234 m/s, d_r= 1 cm, p = 5 cm, d_p= 5.613 mm

3.4. Effect of sphere diameter on mass transfer coefficient

The data on mass transfer coefficient plotted against liquid velocity for three different sphere diameters viz., 3.0, 4.0 and 5.0 cm with the experimental conditions {p = 5.0 cm, d_r = 1.0 cm, d_p = 5.613 mm and U_g=0.0234 m/s} and shown in Fig.7. An observation of the plots of this figure reveals that mass transfer coefficient increased with increasing sphere diameter. As the sphere diameter increases, the cross sectional area available for flow of fluid (hydraulic mean diameter) decreases, hence the flow velocities of the fluid increase. It leads to severe agitation resulting in an increase in mass transfer coefficient. This is also evident from the insert Fig.7a.

3.5. Effect of particle diameter on mass transfer coefficient

The data on mass transfer coefficient obtained in the present case for two different particle diameters were plotted against superficial liquid velocity shown presented in Fig.8. The experimental conditions employed for this study were {p = 7.0 cm, d_r = 1.0 cm, d_b = 4.0 cm, U_L = 0.122 m/s and Ug = 0.0234 m/s} and the diameters of the particles (d_p) used were 0.4248 and 0.561 cm. A close examination of the plots of this figure revealed that the influence of particle size on mass transfer coefficient was found to be significant. This kind of trend can be reasoned as mentioned below.

The contribution to the total turbulence in the present study is from liquid velocity, gas velocity, particle size, promoter geometry etc. By maintaining constant gas velocity, when the superficial liquid velocity is varied for different particle sizes there is reasonable variation in the total turbulence

Hence, one can expect significant variation in the total turbulence and hence the mass transfer coefficient is increases with increasing particle diameter. As the fluid pass through tortuous channels in the three-phase fluidized with string of spheres as internal, the solid particles are subjected to severe agitation. Therefore solid particles strike the reacting surface very strongly. This results in reduced thickness of the resistance film. As the particle diameter is increased, the force with which the particle strike the resistance film also increases and hence the thickness of the resistance film decreases thus yielding high mass transfer coefficient values.

3.6. Correlation

The entire data on reduction of ferricyanide ion and oxidation of ferrocyanide ion, on regression yielded the following correlation equation:

$$j_d \varepsilon_L = 0.3692 \left[\frac{\mathrm{Re}_p\, \varepsilon}{\varepsilon_L (1-\varepsilon)} \right]^{-0.27} (Fr_g)^{0.062} \left[\frac{p}{D_c} \right]^{-0.44} \left[\frac{d_b}{D_C} \right]^{0.42} \qquad (4)$$

Average deviation = 9.81 %, Standard Deviation = 12.28%

correlation plot in accordance with equation (4) has been shown in figure. 9. Here on Y-axis Y_2 has been taken which is defined as

$$Y_2 = j_D \varepsilon_L \left(Fr_g\right)^{-0.062} \left(\frac{p}{D_C}\right)^{0.44} \left(\frac{d_b}{D_C}\right)^{-0.42}.$$

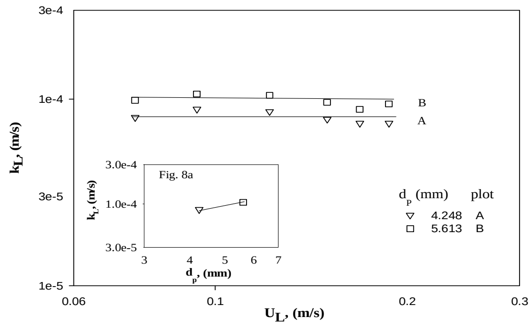

Fig.8 : Effect of solid diameter (d_p) on k_L at constant U_g= 0.0234 m/s, d_r= 1 cm, d_b = 4 cm, p = 7 cm

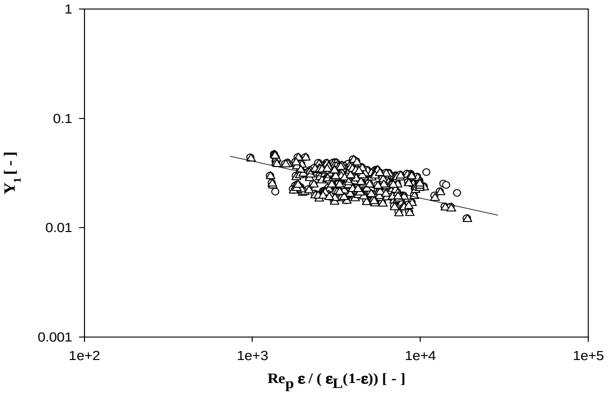

Fig.9. Correlation plot in accordance with eqn. (4)

4. Conclusions

Introduction of the coaxial string of spheres promoter into the three-phase fluidized enhanced the mass transfer coefficient by 20 percent. The parametric effect of gas and liquid velocities on mass transfer coefficient was found to be marginal. The mass transfer coefficient was found to decrease with increase in pitch. An increase in sphere diameter and particle diameter yielded increased mass transfer coefficients. The rod diameter had no significant effect on mass transfer coefficient.

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