Mass transfer at the confining wall of an electrochemical cell with homogeneous flow in the presence of hourglass promoter

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Mass transfer at the confining wall of an electrochemical cell with homogeneous flow in the presence of hourglass promoter

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ABSTRACT

Experiments were conducted to investigate the effect of various dynamic and geometric parameters on wall-liquid mass transfer coefficient and friction factor in the case of flow of an electrolyte through a modified annulus. Hourglass promoter assembly has been employed as the turbulent promoter. To measure the mass transfer coefficient the limiting current technique was chosen. The electrolyte employed was an equimolar 0.01 N potassium ferrocyanide and potassium ferricyanide solution in the presence of an inert sodium hydroxide of 0.5 N. Measurement of limiting current was made at the point electrodes fixed flush with the inner surface of the tube wall. Pressure drop measurements revealed that the friction factor decreased with increasing pitch and increased with increasing characteristic length of the hourglass. Similar trends were also observed for mass transfer coefficient. The mass transfer coefficient data were correlated as Coulburn j-factor expressed as a function of Reynolds number and the geometrical parameters of the promoter. The friction factor data were correlated as a function of Reynolds number and geometrical parameters of the promoter element. © 2013 Trade Science Inc. - INDIA

KEYWORDS

Mass transfer coefficient;
Ionic mass transfer;
Turbulent promoter;
Electrochemical cell;
Homogeneous flow.

INTRODUCTION

The design engineer is always in search of new devices and methodologies that yield higher heat and mass transfer rates. Effective operation, miniaturization of equipment and control are the main objectives that challenge the design engineer's capability. Realization of these tasks can be done by employing a suitable augmentation technique. Various augmentation techniques in use were comprehensively reviewed by Bergles[1]. He classified the augmentation techniques available into two broad categories: active and passive. The active methods require the application of external energy. In passive methods, augmentation is attained by modifying the flow passage for the advantage of increased transfer rates. As revealed by Bergles[1] there is a huge scope and ample opportunities for research in this field. However it is obvious that the emphasis is in the direction of finding cost effective techniques.

Majority of investigations aimed at augmentation of heat and mass transfer rates focused mainly on passive augmentative techniques. These techniques basically modify the flow path thus intensify the turbulence which renders the resistance film thin leading to increased heat and mass transfer rates. Use of treated surfaces, rough surfaces, extended surfaces, displaced enhancement devices, swirl flow devices and coil tubes were the various techniques employed generally for enhancement of heat and mass transfer rates.

Dewan et al[2] reviewed all the passive augmentation techniques. Compound augmentation is also often used by combining the passive and the active methods.

In homogeneous flow of electrolyte, augmentation of mass transfer rates has been investigated experimentally by employing displaced promoters such as cross-flow elements[3], coiled wires[4], string of spheres[5], string of cones[6], string of discs[7], helical tape on a rod[8], twisted tapes[9] etc. All these investigators[3-9] employed limiting current technique for obtaining the mass transfer coefficient between the column wall and the flowing liquid electrolyte.

The limiting current technique has the following advantages for the measurement of mass transfer coefficient: (i) the chemical polarization involved is negligible (ii) the reacting surface remains smooth and unaffected unlike in the cases of solids dissolution or sublimation processes and (iii) the measurements are relatively fast, accurate and reproducible. The fluid electrolyte consists of 0.01 N potassium ferrocyanide and potassium ferricyanide with 0.5 N sodium hydroxide as inert electrolyte. Limiting current is measured for the reduction of ferricyanide ion represented by the following reaction equation:

$$[\mathrm{Fe\,(CN)_6}]^{3-} + e \rightarrow [\mathrm{Fe\,(CN)_6}]^{4-} \quad (1)$$

Computation of wall-liquid of mass transfer coefficient is made by measured limiting current at point electrodes fixed flush with the inner surface of the prespex tube using the following formula[10].

$$k_L = \frac{i_2}{nAFC_o} \quad (2)$$

Although there are a large number of studies carried out in heat transfer a close look at the literature reveals that not too many investigations are aimed towards mass transfer. Very few works were reported in which the studies were directed towards the investigation of hydrodynamics and wall-liquid mass transfer using bluff bodies. Further, investigations employing an hourglass promoter element for enhancement of mass transfer rates were found to be scarce[11].

In view of this, the present study has been attempted to investigate the effect of liquid velocity and the geometric variables of the promoter element on mass transfer coefficient and friction factor in the presence of hourglass promoter. The range of variables covered in the present study has been compiled in TABLE 1.

TABLE : 1 Range of variables covered in the present study

S.No	Parameters studied	Minimum	Maximum
1	Superficial velocity of liquid V in m/s	0.04	0.26
2	Diameters of the rod, d_i in cm	1.0	---
3	Pitch, p in cm	5.0	10.0
4	Characteristic length of the hourglass element, d_b in cm	3.0	5.0

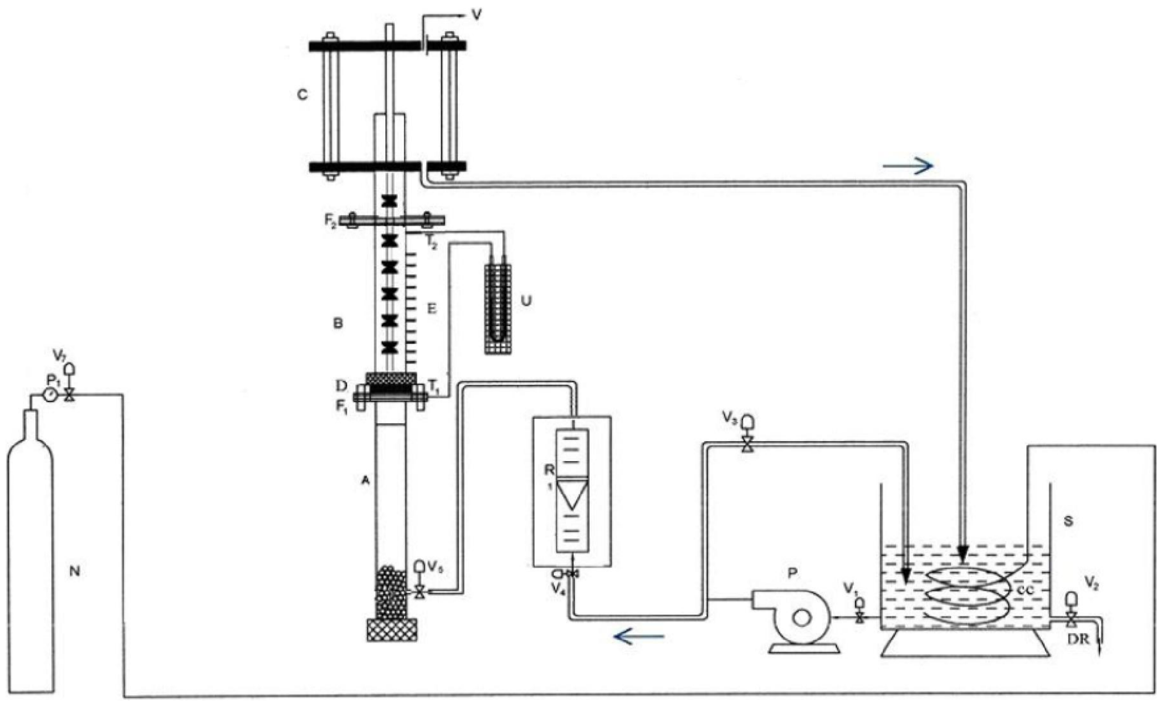
EXPERIMENTAL

The equipment was designed and fabricated to carryout studies on liquid-wall mass transfer at the inner surface of the outer column of an annular electrochemical cell. A string of hour glass elements fixed on a rod with different pitches was employed as a promoterassembly, which was placed concentrically in the electrochemical cell.

The schematic diagram of the experimental set-up used in the present investigation was shown in figure 1. The equipment and apparatus consisted of a sintex cylindrical storage tank (S), centrifugal pump (P) for circulating the electrolyte, a rotameters (R_1) for measuring the flow rate of the electrolyte and a nitrogen cylinder (N) for supply of nitrogen gas along with regulator (R). A U-tube differential manometer (U) was provided to measure the pressure difference across the test section (B). Valves V_1 to V_5 were used to control the flow rates of liquid while through the experimental column.

The storage tank was of 100 liters capacity, completely covered with a sintex sheet to eliminate continuous contact with the surrounding air. The bottom side of the tank was connected to the suction side of the pump (P) through a globe valve (V_1). Another globe valve (V_2) was provided at the bottom of the storage tank to facilitate the periodic cleaning. A spiral coil (CC) with perforations was placed in the storage tank for deaeration of the electrolyte with nitrogen gas. The pump (P) of Kirloskar make was made of stainless steel that has a capacity of 1 hp. The discharge end of the pump was divided into two lines, one directly connected to rotameter (R) and the other to a by-pass line. The by-pass line was provided with a globe valve V_3 to control the flow through rotameter. The rotameter (R) was of Indus make has range of 0 to 60 liters per minute. One more valve V_4 was provided at the entry point of rotameter. Valve V_5 provided at the discharge end of the rotameter facilitated the fluid electrolyte to flow through the experimental column.

The experimental column shown in figure 3.1 mainly



A- Entrance calming section; B- test section; C- exit section; CC – copper coil; D- distributer wire mesh; DR- drain; E- electrodes; F_1, F_2- flanges; N- nitrogen cylinder; P-pump, R – rotameter; S – storage tank; T_1, T_2- pressure tapings, U- manometer; V- vent; V_1 to V_7- valves.

Figure 1 : Schematic diagram of the experimental unit

consisted of the following three sections: An entrance calming section (A), a test section (B) and an exit section (C). The entrance calming section (A) made of a copper tube of 6.73 cm inner diameter and 1.07 m long, was filled with marble stones of random size, to eliminate the effects due to tangential entry of the fluid electrolyte and to minimize the flow fluctuations was connected to the main test section (B) by means of a flange (F_1). The inlet provided at the bottom of the entrance calming section facilitated the flow of the metered fluid electrolyte.

The test section (B), which served as the electrochemical cell, was made of smooth Perspex tube of 6.73 cm inner diameter and 0.6 m height. The inner wall of the test section was provided with copper point electrodes of diameter 3.42 mm. The point electrodes 34 in number were machined to the size out of 4 mm diameter copper rod. One end of these electrodes was fixed flush with the surface of the inner wall of the test section while the other end projected outward served as terminal for connecting the electrodes to the external circuit. Two pressure taps, (T_1) at the bottom flange and (T_2) at the top flange have been provided across the test section for pressure drop measurements. The Taps (T_1 and T_2) were connected to the limbs of the U-tube manometer to measure the pressure drop. Carbon tetrachloride was used as manometric fluid. A stainless steel wire mesh (D) was placed at the bottom of the test section served as a supporter to hold the promoter element. The wire mesh also allowed the distribution of liquid into the experimental test section without appreciable pressure drop.

Entrance calming section; B- test section; C- exit section; CC – copper coil; D- distributer wire mesh; DR- drain; E- electrodes; F_1, F_2- flanges; N- nitrogen cylinder; P-pump, R – rotameter; S – storage tank; $T_{1,}$

T_2- pressure tapings, U- manometer; V- vent; V_1 to V_7- valves.

Two hemispheres of same diameter d_b were joined together in the hour glass type arrangement and this arrangement was used as the repeating element in the promoter assembly. The diameter of the hemispheres is considered as the characteristics length of the hourglass assembly. Hourglass elements of different sizes (d_b) were placed concentrically on a stainless steel rod of 1.0 cm diameter with varied pitch p acted as the turbulent promoter shown in figure 2. Promoter elements of different geometrical characteristics (viz., pitch p and characteristic length d_b) were fabricated and the details of the promoter geometries used in the present study are compiled in TABLE 2.

The exit section (C) was also of the same diameter

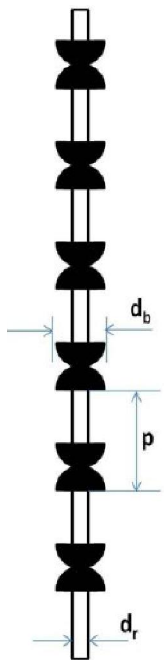


Figure 2 : Details of hourglass promoter

TABLE 2 : Details of promoter geometry

S.No.	Diameter of the rod d_r(cm)	Pitch, P(cm)	hemisphere diameter, d_b (cm)
1	1.0	5.0	4.0
2	1.0	7.0	3.0
3	1.0	7.0	4.0.
4	1.0	7.0	5.0
5	1.0	10.0	4.0

as A and B with its open end into the separator, was connected to the test section (B) by means of a flange F_2. The fluid electrolyte from the test section was drawn from the bottom of the separator (D).

Prior to the assembly of the test section, the surfaces of the point electrodes were polished with a three zero emery paper and cleaned thoroughly. About 70 liters of equimolar solution of potassium ferrocyanide and potassium ferricyanide of about 0.01 N and 0.5 N of sodium hydroxide were prepared from analar grade reagents using distilled water of 5 mmho specification. This solution was used as the electrolyte. The rotameters were calibrated using this fluid electrolyte. The solution was deaerated using nitrogen prior to recirculation through the test section. The test electrodes used were point electrodes of diameter 0.00342 m.

The electrolyte from the storage tank was metered and circulated through the test section. The rotameter R was used to measure the flow rate of electrolyte and the flow rate was adjusted by using the control and by pass valves. Then limiting current was measured by applying an electric potential in small increments between the test electrode (T) and the wall electrode (W). Initially blank runs were conducted with indifferent electrolyte alone. Since no appreciable currents were detected the measured limiting currents obtained during red-ox process were essentially due to the depolarizing agent.

The measurement of limiting current was made in the lines similar to those reported earlier in the studies on ionic mass transfer[9,10]. During each run the reacting ion concentration was obtained by volumetric analysis.

Ferrocyanide ion concentration was estimated by permanganometric titration method and the ferricyanide concentration was obtained using iodometric titration method[12].

RESULTS AND DISCUSSION

Before commencing the experimentation with the present set up, mass transfer coefficient data were obtained in homogeneous flow with disc promoter. The k_L values thus obtained are found to agree the data of Venkateswarlu et al[7] within 8% deviation.

Mass transfer

When a solid surface and a flowing liquid come into contact, there appears a boundary layer otherwise also known as viscous sublayer. If either heat or mass transfer to occur from the liquid to the solid surface or vice versa, the major resistance for any such transfer process is offered by this laminar sublayer. The more the thickness of this layer, the more the resistance to the transfer process. By increasing the turbulence, the thickness of this layer can be reduced. The increase of turbulence can be done by increasing the fluid velocity and also by creating vigorous churning of the liquid. If the fluid is allowed to pass through a cross section that contains a series of sudden expansions and contractions, then the resulting scouring action leads to intense turbulent fields yielding reduced film thickness. Hourglass promoter element is one such internal element that modify the flow passage such that a series of sudden contractions and expansions results.

Therefore, it can be anticipated that the hourglass promoter has the capacity of yielding high heat/mass transfer coefficients.

Augmentation with hourglass promoter

The experimental data obtained in the present experiment have been analyzed graphically in relation to various dynamic and geometric variables. Figure 3 gives the data of the present study plotted as average mass transfer coefficient $k_{L,avg}$ against liquid velocity V for three cases of (i) homogeneous liquid flow through empty column (Plot A), (ii) homogeneous liquid flow in the presence of an hourglass promoter {d_b = 4 cm; p = 10 cm} (Plot B), (iii) homogeneous liquid flow in the presence of another hourglass promoter {d_b = 5 cm; p = 7 cm} (Plot C). The magnitudes of improvements over empty column were shown in plots B and C. Plot A is the data predicted form Lin et al[10] for the case of homogeneous flow of electrolyte through an empty conduit. The present experimental data obtained in homogeneous liquid flow with two different hourglass promoters were presented as plots B and C. Plots A and B shows the improvements in $k_{L,avg}$ due to the presence of an hourglass promoter {d_b = 4 cm; p = 10 cm} were upto 50% on lower liquid velocity end and upto 140% on higher liquid velocity end. Plots A and C shows the improvements in $k_{L,avg}$ due to the presence of introducing an hourglass promoter {d_b = 5 cm; p = 7 cm} were upto 5.5 fold on lower liquid velocity end and upto 7.5 fold on higher liquid velocity end. These observations indicate that the presence of an hourglass promoter assembly in a homogeneous flow is definitely advantageous as it enhances turbulence resulting in increased mass transfer.

Effect of axial length

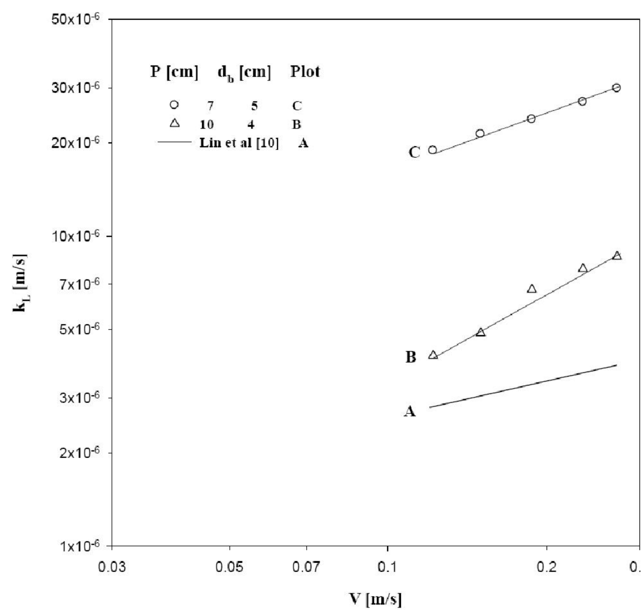


Figure 3 : Augmentation of mass transfer coefficient

When a fluid is flowing in a circular or annular conduit or in any conduit of uniform cross sectional area, and if the test section is taken in such a way that the boundary layer is fully developed, then the velocity profile remains same along the axial length of the test section provided steady flow is maintained. One can then expect that the measured limiting current densities would not be affected by the axial distance. If an internal such as an hourglass promoter, which contains a central rod on which a string of hourglass elements was placed at equispaced distances coaxially in the test section, then the flowing fluid would be subjected to a series of sudden contractions and expansions resulting in regions in which the degree of turbulence intensity varies. Since the thickness of boundary layer is the essential component of resistance offered to transfer process, more the turbulence, less the thickness of the boundary layer. Thus it can be anticipated that the measured local mass transfer coefficient would vary along the length of the test section. Figures.4a, 4b and 4c show the plots of mass transfer coefficient drawn against axial distance for hourglass promoters of three different pitches viz., 5, 7 and 10 cm. In all these cases, the mass transfer coefficient was found to fluctuate with the longitudinal distance.

The average of mass transfer coefficient computed from arithmetic mean is also shown in the figures. Average mass transfer coefficient data have been considered in all future discussions as the transfer process from wall to liquid or liquid to wall is the main objective of present study.

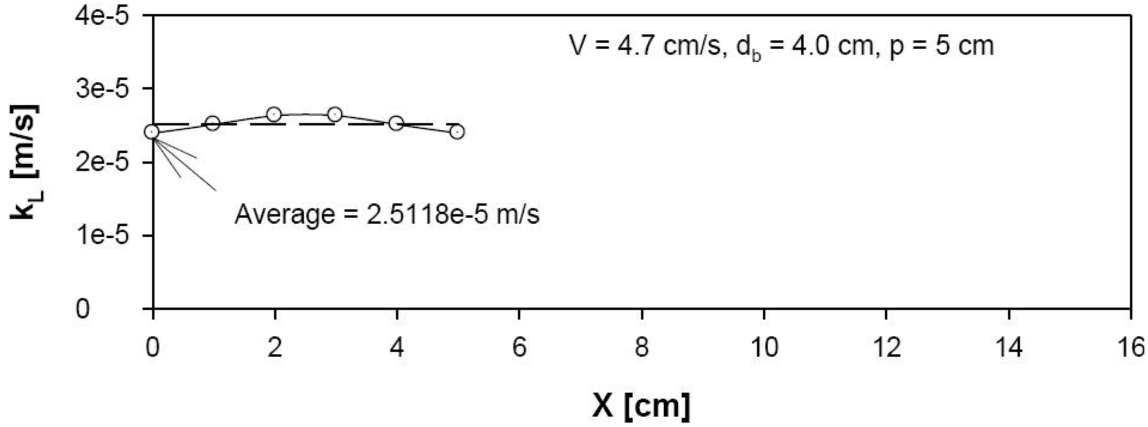


Figure 4a : Variation of kL with longitudinal distance for a pitch of 5 cm

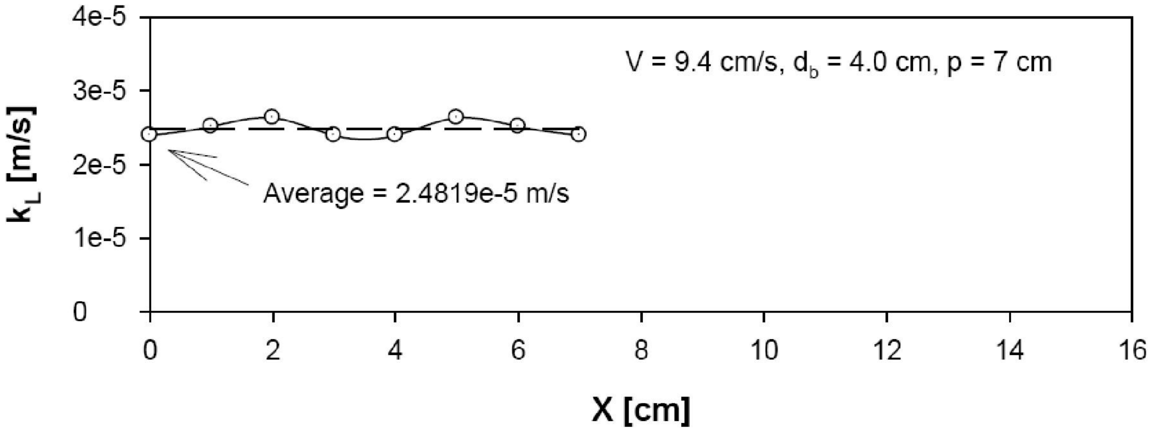


Figure 4b : Variation of kL with longitudinal distance for a pitch of 7 cm

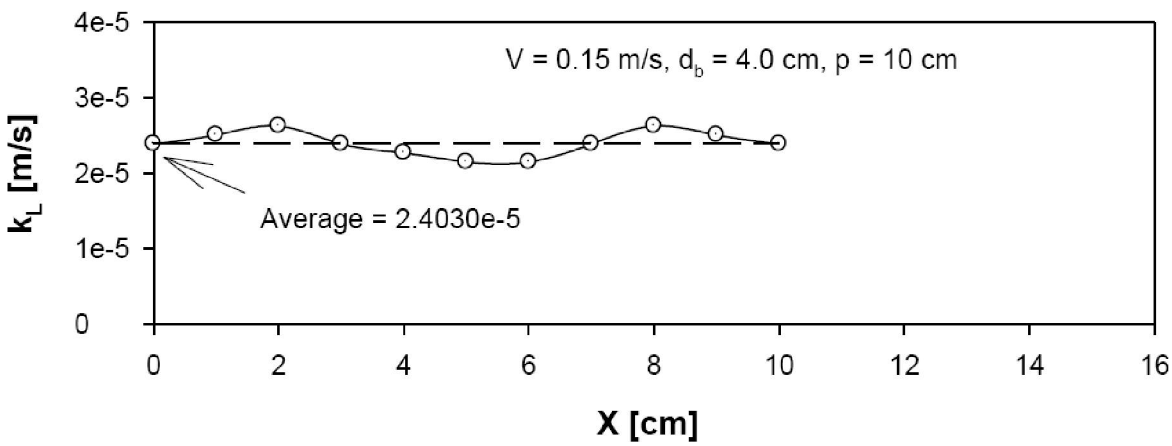


Figure 4c : Variation of kL with longitudinal direction for a pitch of 10 cm

Effect of pitch

Pitch of the hourglass promoter element is an essential parameter that influences the flow pattern at the wall thus significantly affecting the thickness of the resistance film. It can be anticipated that the increase in pitch may cause a decrease in mass transfer coefficient. Figure 5 represents the data on $k_{L,avg}$ plotted against liquid velocity for three different pitch values. Plot A corresponds to a pitch of 5 cm, plot B for 7 cm and plot C for a pitch of 10 cm. It is observed that the higher the pitch the lower is the mass transfer coefficient as expected. As the pitch increases the mass transfer coefficient decreases which is as also revealed by figure 5a, the inset of figure 5.

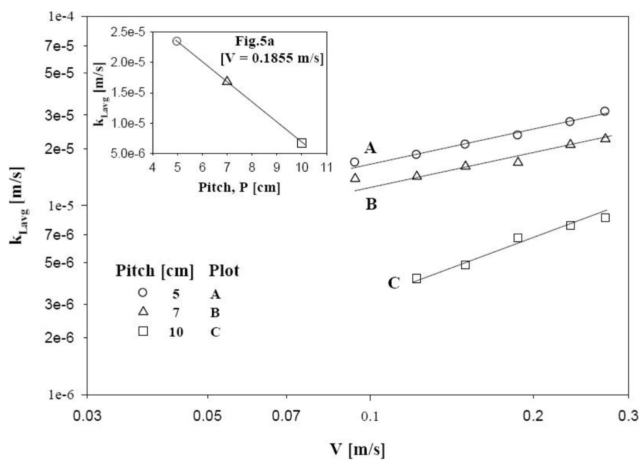


Figure 5 : Effect of pitch on kL for db = 4 cm

Effect of characteristic length

The flow past an hourglass internal is identical to flow past a bluff body. The flow is transverse to the direction of the hourglass orientation and one can expect vigorous wake formation both at the leading edge and at the trailing edge of the hourglass thus resulting in severe turbulence.

The diameter of the hemisphere has been considered as the characteristic length of the hourglass element. Increasing hemisphere diameter means decreased available flow area, hence subsequent increase in the local velocity of the fluid, thus, augmentation of the average mass transfer coefficient at the wall can be anticipated. This could be established from the trends of the mass transfer coefficient data plotted in figure 6. In this figure the plots were drawn between $k_{L,avg}$ and liquid velocity for three different diameters employed in the present experiment. Plot A shows the mass transfer coefficient data with the characteristic length of 3.0 cm, plot B corresponding to 4.0 cm and plot C represents the data with that of 5.0 cm. A close inspection of the plots of the figure reveals that the mass transfer coefficient increased with increase in characteristic length. This is also evident from the cross-plot presented as inset figure 6a.

Correlation

The data on $k_{L,avg}$ obtained in the present experiment have been correlated in the j_D-Re format by regression analysis and the following equation is obtained.

$$j_D = 0.13\left(\frac{\rho D_c V}{\mu}\right)^{-0.27}\left(\frac{d_b}{D_c}\right)^{0.62}\left(\frac{p}{D_c}\right)^{-2.52} \quad (3)$$

Average deviation = 7.63%
Standard deviation = 9.84%

The correlation plot according to equation (3) has been shown in figure 7.

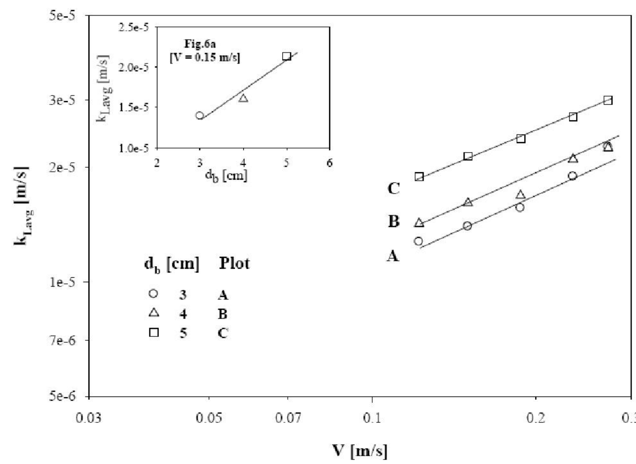


Figure 6 : Effect of characteristic length on kL for p = 7 cm

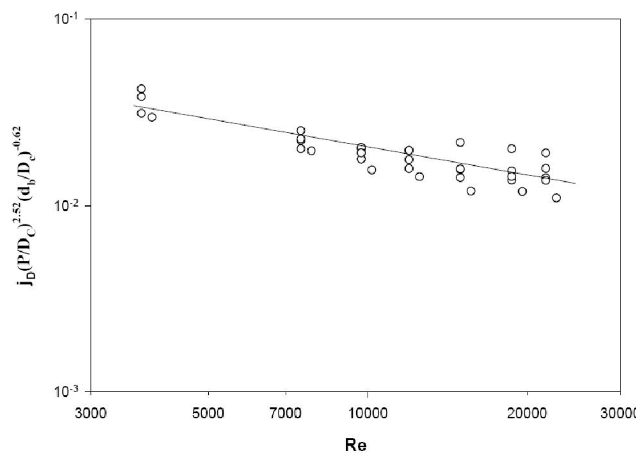


Figure 7 : Correlation plot in accordance with eqn.(3)

Pressure drop studies

Study of pressure drop is very important since it is related to the power consumption in a particular operation. Therefore it is directly affecting the variable costs of the operation and hence the total product cost. In view of this, the pressure drop data were also obtained in the present experiment. The data were analyzed as pressure drop versus velocity.

Effect of pitch

The pressure drop data in the present study have been measured using a U-tube manometer connected between two pressure tapings across the test section. These data were converted into pascals, and that data have been plotted against liquid velocity for various pitches of the hourglass promoter element with the characteristic length being 4 cm. Three pitch values have been considered in the present case viz., 5, 7 and 10 cm. The corresponding data were represented through plots A, B and C respectively in figure 8. It is conspicuous from the plots of the figure that the pressure drop decreased with increase in the pitch value. However, the variation in the pressure drop between pitch values 5 and 7 is very large and the difference between pitch values 7 and 10 is nearly very less. However, for the entire range of pitch under consideration, a decreasing trend has been observed. This trend is also conspicuous from the plot of the figure 8a, the inset figure.

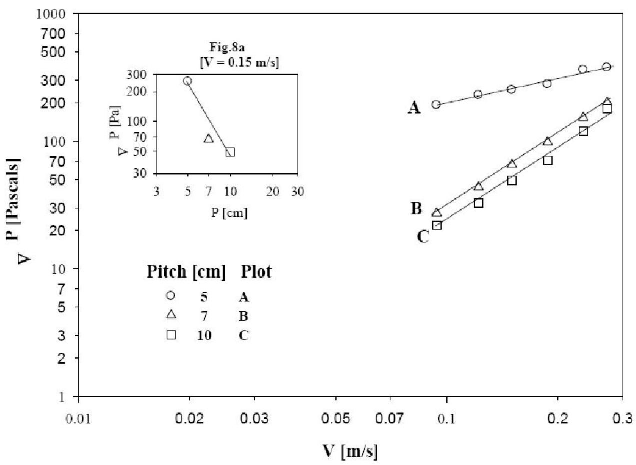


Figure 8 : Effect of pitch on pressure drop for db = 4 cm

Effect of characteristic length

The diameter of the hemisphere d_b has been chosen as the characteristic length of the promoter element i.e., the hourglass element. The data obtained on pressure drop have been plotted against liquid velocity for three different characteristic lengths considered in the present study. It can be expected that an increase in characteristic length reduces the flow area and hence an increase in pressure drop can be anticipated. A close inspection of the plots of figure 9 and its inset figure 9a indicates similar trends.

Correlation

A correlation has been obtained between friction factor and Reynolds number by regression analysis presented as eqn.5.2.

$$\mathbf{f} = 12.45\left(\frac{\rho \mathbf{D_c V}}{\mu}\right)^{-0.15}\left(\frac{\mathbf{d_b}}{\mathbf{D_c}}\right)^{4.77}\left(\frac{\mathbf{p}}{\mathbf{D_c}}\right)^{-1.76} \quad \mathbf{(4)}$$

Average deviation = 8.57%

Standard deviation =10.86%

The correlation plot according to equation (4) has been shown in figure 10.

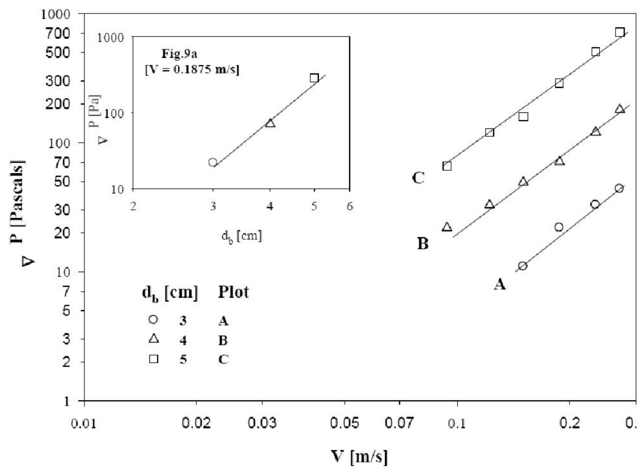


Figure 9 : Effect of characteristic length on Pressure drop for p = 7 cm.

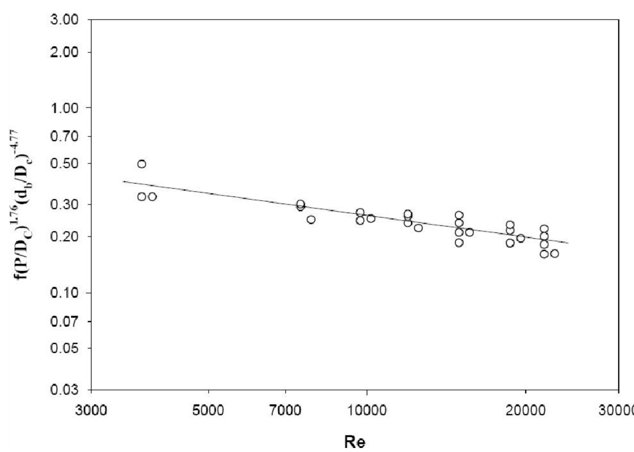


Figure 10 : Correlation plot in accordance with eqn (4)

CONCLUSIONS

The experimental data obtained on mass transfer coefficient have been analyzed from which the following conclusions were drawn:

- About 7.5 fold improvement in mass transfer coefficient has been observed in homogeneous flow due to the presence of hourglass promoter elements in comparison with homogeneous flow without any promoter.

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- The mass transfer coefficient decreased with increasing pitch value
- The mass transfer coefficient increased with increasing characteristic length of the promoter element.
- The mass transfer coefficient is correlated in j_D-Re format of equation.
- The pressure drop decreased with increasing pitch.
- The pressure drop increased with increasing characteristic length of the promoter element.

NOTATION

A	area of the reacting surface	[m^2]
C_0	concentration of reacting ion (Fe^{2+} or Fe^{3+})	[$kmol/m^3$]
d_b	characteristic length of hourglass element	[m]
d_i	rod diameter	[m]
D_C	column diameter	[m]
D_L	diffusivity of reacting ion	[m^2/s]
F	Faraday constant	[C]
I	current	[A]
i_d	limiting current density	[A/m^2]
i_L	limiting current	[A]
k_L	mass transfer coefficient	[m/s]
$k_{L.avg}$	average mass transfer coefficient	[m/s]
p	pitch	[m]
V	superficial liquid velocity	[m/s]
X	longitudinal distance	[m]
n	number of electrons released or consumed during the reaction	[-]

Greek symbols

ΔP	pressure drop across test section	[Pa]
μ	liquid viscosity	[kg/m s]
ρ	liquid density	[kg/m^3]

Dimensionless groups

F	Frication factor, $= \frac{2(\Delta p)\rho}{4LV^2}$
j_D	Coulburn j-factor $= \frac{k_{L\,avg}}{V} Sc^{2/3}$
Re	Reynolds number $= \frac{\rho D_C V}{\mu}$
Sc	Schmidt number $= \frac{\mu}{\rho D_L}$

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