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Electro Winning from Dilute Solutions at Enhanced Rates using Three-Phase Flow Reactor

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Abstract: ***Enhancement of mass transfer coefficient is highly desirable for economic design of process equipment. The present study is essentially carried out to know the effect of flow variables such as gas and liquid velocities and geometric parameters of the internal on mass transfer coefficients in a three phase fluidized bed. The mass transfer coefficient data were obtained using a string of cones internal in a three-phase fluidized bed electrochemical reactor. The flow system investigated was nitrogen, a fluid electrolyte and spherical glass beads as gas, liquid and solid phases respectively. Limiting current technique was employed to obtain mass transfer data. The internal comprises of a string of cones arranged concentrically on a central rod which is placed coaxially in a three phase fluidized bed. The presence of internal in three phase fluidized beds augmented the mass transfer coefficient significantly. In the present investigation it was found that the effect of gas velocity, liquid velocity, rod diameter and cone diameter was only marginal. However, the influence of pitch, half apex angle of cone and particle diameter was found to be significant. Correlations were developed based on least squares regression analysis for the prediction of mass transfer coefficient in terms of pertinent variables.***

Keywords: ***mass transfer coefficient, fluidization, limiting current, turbulent promoter, augmentation.***

I. INTRODUCTION

High heat and mass transfer rates can be obtained by employing suitable augmentation technique. 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. In recent times, since three-phase fluidized beds with promoter internals were found to yield maximum mass transfer rates, they found wide application in electrochemical processing units. Some promoters that were employed in three phase fluidized beds were helicoidal tapes[1], string of discs[2], twisted tapes[3], string of angled discs[4], string of spheres[5], string of hemispheres[6] and string of doublecones[7]. It was also observed that string of cones yielded significant enhancement in as-liquid flow systems [8-10].

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Therefore, in the present work, string of cones were employed as turbulent promoters in three-phase fluidized bed electrochemical reactors. It was also found that no study has been reported on mass transfer in three-phase fluidized beds with string of cones as internal [11].

In view of this, investigations have been carried out to study the effect of pertinent dynamic and geometric variables on mass transfer coefficient in a three-phase fluidized bed in the presence of string of cones promoter elements using limiting current technique [12]. Electrochemical system belonging to ferricyanide – ferrocyanide redox reactions has been chosen for the present study. Equimolar Potassium ferricyanide and Potassium ferrocyanide was prepared and used as liquid phase whereas nitrogen is the gas phase and glass beads were employed as solid phase. The electrochemical reactions are

$$Fe(CN)_6^{3-} + e \rightarrow Fe(CN)_6^{4-} \quad \text{(Reduction)} \ldots(1)$$

$$Fe(CN)_6^{4-} \rightarrow Fe(CN)_6^{3-} + e \quad \text{(Oxidation)} \ldots(2)$$

Computation of the mass transfer coefficient is made from measured limiting current using the following equation [12]:

$$k_L = \frac{i_L}{zFAC_0} \quad \ldots (3)$$

The range of variables covered in the present study are presented in Table.1

S.No	Parameters studied	Min.	Max.
1.	Gas flow rate (Q_g x10^5), m^3/s	5	25
2.	Liquid flow rate (Q_Lx10^5), m^3/s	16.7	83.3
3.	Velocity of gas, U_g, [m/s]	0.014	0.074
4.	Velocity of liquid, U_L, [m/s]	0.047	0.234
5.	Promoter rod diameter (d_r) cm	0.6	1.3
6.	Pitch between the cones (p), cm	5.0	10.0
7.	Diameter of the cone (d_c), cm	3.0	5.0
8.	Diameter of the particle (d_p), mm	2.930	5.620
9.	Half apex angle of cone (θ), degrees	30	60
10.	Reynolds number, Re	3725	22723
11.	Particle Reynolds number, Re_p	234	1886

II. EXPERIMENTAL

The schematic of the experimental setup used in the present study is shown in Fig.1. Same setup has been used by the authors in their earlier work.

Hence, in the present paper, the description of setup and experimental procedure is not presented because it is available elsewhere [7].

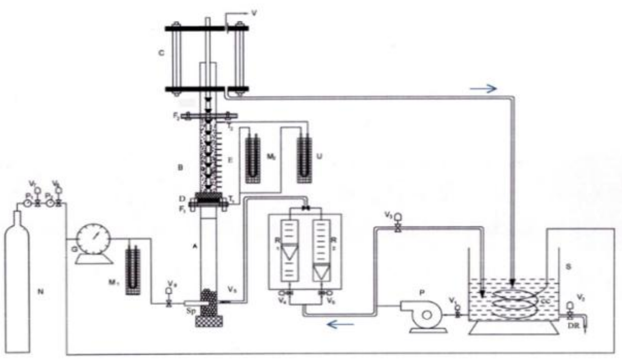
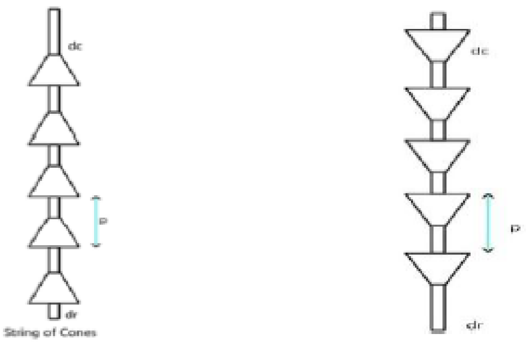


Fig.1. Schematic of the experimental unit

A - entrance calming section; B- test section; C- exit section; D-distributer wire mesh; DR- drain; E- electrodes; F_1, F_2- flanges; N- nitrogen cylinder; P-pump, R – Regulator, R_1 and R_2 – rotameters; S – storage tank; T_1, T_2- pressure tapings-manometer; V- vent; V_1 to V_7- valves.

String of cones placed concentrically on a rod essentially acted as promoter internals in the present experimental studies. The schematic of the string of cones promoter assembly is shown in Fig.2. Two types of arrangements are considered: Regular cone and Inverse cone. Regular cone arrangement means the base of the cone acts as leading edge and the apex of the cone acts as trailing edge in the direction of the flow whereas the inverse cone is vice-versa. The promoter elements of different geometric characteristics viz., diameter of rod (d_r), pitch (p), diameter of the base of the cone(d_c) and half of the apex angle (θ) were fabricated and used.



Regular cone Inverse cone

Fig.2. Configuration of cone promoter {d_r = rod diameter; d_c= cone diameter; p = pitch}

III. RESULTS AND DISCUSSION

Fig. 3 shows the $k_{L,avg}$ data obtained in the present investigation plotted against liquid velocity U_L for four cases: (i) homogeneous liquid flow in conduit of circular cross-section (Plot A), (ii) three-phase fluidized bed without any promoter element (Plot B), (iii) three-phase fluidized bed with regular cone promoter element (Plot C) and (iv) three-phase fluidized bed with inverse cone promoter element (Plot D). Plots B to D represent the data corresponding to three-phase fluidized beds. The data were obtained when the bed material employed was glass balls of 4.243 mm diameter and the superficial gas velocity was maintained at 2.34 cm/s. The magnitudes of improvements over homogeneous liquid flow are shown through plots B, C and D. Plot A is the data predicted from Lin et al[12] for the case of pipe flow and plot B for the three-phase fluidized bed without any promoter internal. The data of plot B was found to be in accordance with Ramesh et al [1]. The plots C and D represent the present experimental mass transfer coefficient data in three-phase fluidized beds in the presence of conical promoters viz., regular cone and inverse cone respectively. Plots A and B show that the improvements in the mass transfer coefficients obtained in three-phase fluidized bed were upto 10 fold when compared with homogeneous flow of electrolyte in pipe flow (plots B and A). Plot C gives the magnitudes of augmentation in $k_{L,avg}$ in three-phase fluidized bed due to the introduction of regular cone promoter in which the enhancements were upto80 percent over three-phase fluidized bed without any promoter (Plots C and B) and the augmentation in mass transfer coefficients due to the introduction of inverse cone promoter is 120 percent (Plots D and B). The plots thus reveal that significant enhancement in mass transfer rates could be realized with string of cones promoter. Depending on the geometry of the conical element the augmentation varied upto a maximum of 120 percent. One can reason that the conical promoter elements yield reckonable augmentation of mass transfer coefficients. Since highest mass transfer coefficient values were realized for inverse cone promoter, for the present investigation inverse cone promoter was chosen.

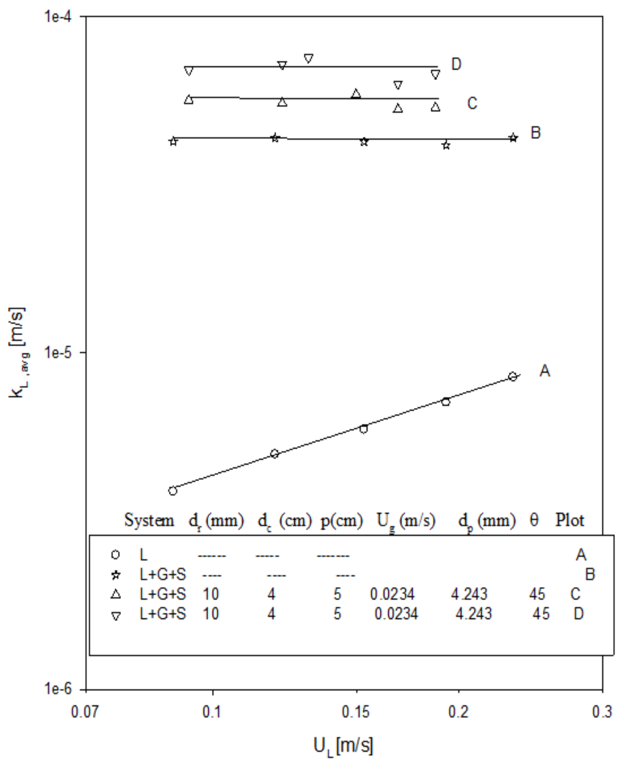


Fig.3. Augmentation of mass transfer coefficient

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3.1. Longitudinal variation of mass transfer coefficient

Vijay et al [10] reported that with inverse cone promoter element in gas liquid upflow columns, the fluctuation in limiting current values in axial direction of fluid flow fell within ±21% about the mean.

Similar fluctuating behaviour was observed with inverse cone promoter in case of gas-liquid fluidized beds also. From the plot of the Fig.4 it could be noticed that, with inverse cone promoter element, the fluctuations fell within ±30%. The reason for the fluctuating behavior can be attributed to the series of contractions and expansions that appear along the entire length of test section in flow direction.

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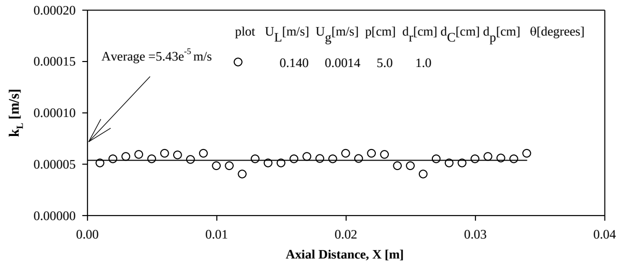

Fig.4. Longitudinal variation of mass transfer coefficient in three-phase fluidized beds in the presence of inverted cone promoter

3.2. Effect of liquid and gas velocities

In the present study mass transfer coefficient data were obtained in a three-phase fluidized bed in the presence of inverse cone promoter {p = 5 cm; d_r = 1 cm; d_c = 4 cm; θ = 45°} at three different gas velocities and shown in Fig.5. The superficial gas velocities employed were 0.014, 0.0234 and 0.0374 m/s respectively. It is clearly seen from the plots of this figure that both the liquid and gas velocities have not exhibited any noticeable effect on the mass transfer coefficient in three-phase fluidized beds in the presence of inverse cone promoter internal. The reason for this can be attributed to the overall turbulence generated by the gas phase, liquid phase, fluidizing particles and promoter internal was very large so that the turbulence added out of the variations in gas and liquid velocities was nearly insignificant. This observation was also conspicuous from figure that was drawn between mass transfer coefficient and gas velocity for three different liquid velocities viz., 0.0935, 0.1217 and 0.1498 m/s.

3.3. Effect of pitch

Fig.7 shows the mass transfer coefficient data obtained for the case of an inverse cone promoter element {d_r = 1 cm; d_c = 4 cm; θ = 45°} against liquid velocity for three different pitch values viz., 5, 7 and 10 cm at a constant superficial gas velocity of 0.0234 m/s. A close examination of the plots of this figure reveals that higher value of mass transfer coefficient was realized for lower pitch value. As the pitch value was increased from 5 cm to 7 cm the mass transfer coefficient decreased. The reason for this can be explained in the following way. At lower pitch value the flowing fluids are subjected to more number of contractions and expansions and hence the total turbulence in the bed would be larger and high values of mass transfer coefficient would be realized. As the pitch value is increased the number of repeating elements present in the test section becomes less and hence the number of contractions and expansions decreases and hence the total turbulence would decrease which resulted in decreased value of mass transfer coefficient. As the pitch value is decreased further from 7 cm to 10 cm, no further reduction in mass transfer coefficient can be obtained probably due to no further reduction in total turbulence in the bed. Similar trends are also seen from Fig.8 that is drawn against gas velocity.

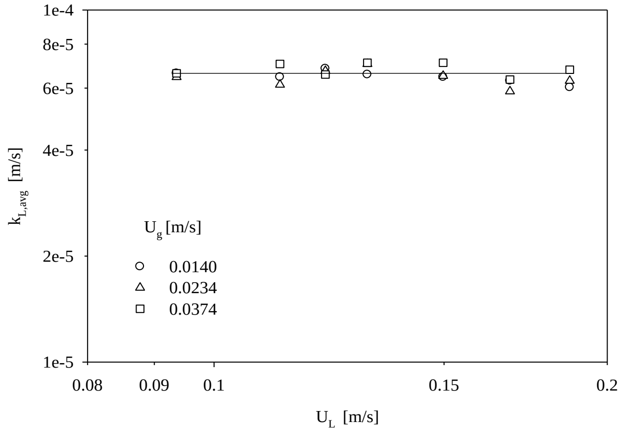

Fig.5. Effect of gas velocity on kL,avg

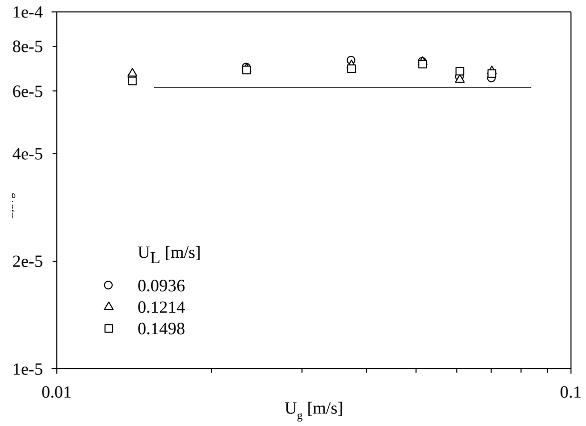

Fig.6. Effect of liquid velocity on kL,avg

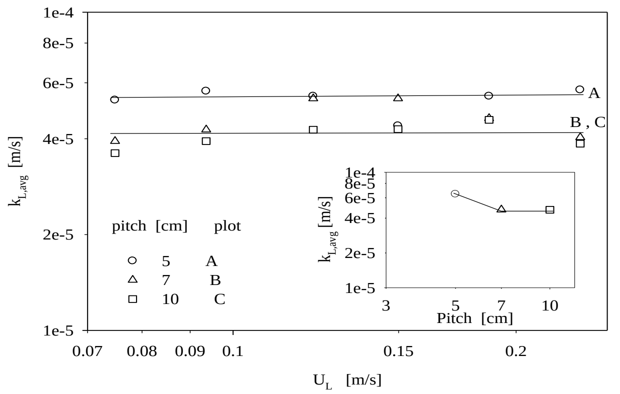

Fig.7. Effect of pitch

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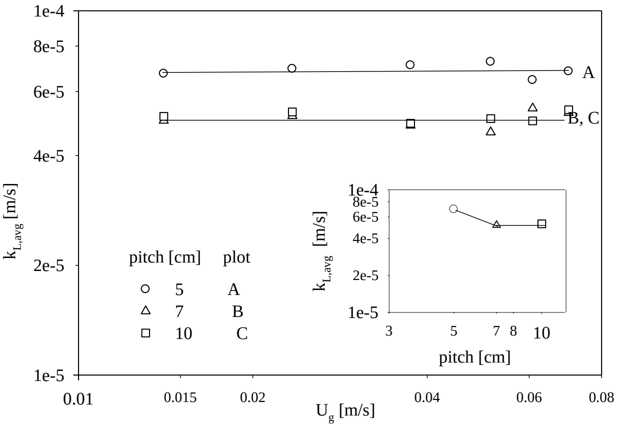

Fig.8. Effect of pitch

3.4. Effect of cone diameter

Fig.9 shows the mass transfer coefficient data obtained for the case of an inverse cone promoter element {p = 7 cm; d_r = 1 cm; θ = 45°} plotted against gas velocity for three different cone diameter values viz., 3, 4 and 5 cm at a constant superficial liquid velocity of 0.121 m/s. A close inspection of the plots of this figure indicates that these plots obeyed no specific trend.

The reason may be attributed to the following explanation. One can argue that the turbulence generated in the test section is due to gas velocity, liquid velocity fluidizing solids and promoter geometry. The combined effect is exhibited on the reduction of the thickness of the resistance film that appears on the electrode surface. It can be pointed out that, in these circumstances, maximum turbulence is generated in the presence of small diameter cone. By increasing the cone diameter there is no further addition of turbulence. Hence there is no change in the mass transfer coefficient value with the increase in cone diameter.

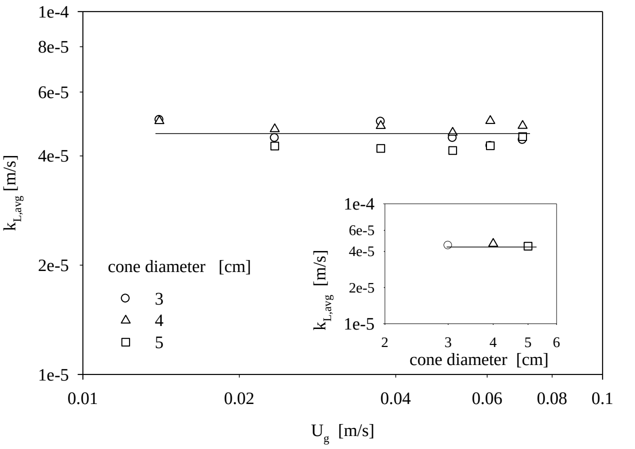

Fig.9. Effect of cone diameter

3.5. Effect of half apex angle of cone

Fig.10 shows the mass transfer coefficient data obtained for the case of an inverse cone promoter element {p = 7 cm; d_r = 1 cm; d_c = 4 cm} against liquid velocity for three different half cone apex values viz., 30, 45 and 60° at a constant superficial gas velocity of 0.0234 m/s. A close examination of the plots of this figure indicates that the mass transfer coefficient values increased with increase in cone angle. One can understand that as the cone angle is increased the streamlined nature of the cone decreases and simultaneously the bluff body nature increases. This resulted in an increase in mass transfer coefficient.

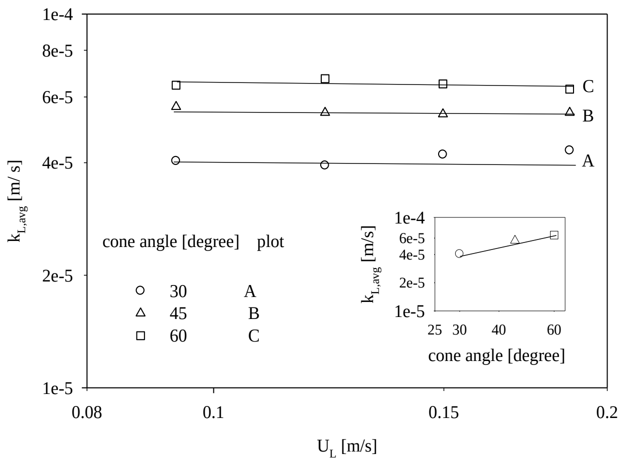

Fig.10. Effect of cone angle

3.6. Effect of rod diameter

The mass transfer coefficient data obtained for the case of an inverse cone promoter element {p = 7 cm; d_c = 4 cm; θ = 45°} were plotted against superficial liquid velocity for three different rod diameters viz., 0.6, 1.0 and 1.3 cm at a constant superficial gas velocity of 0.0234m/s and is shown in Fig.11. A close look at the plots of this graph indicates that the points fell closely indicating that the influence of rod diameter on mass transfer coefficient is relatively insignificant.

The promoter rod is placed coaxially at the centre of the column. When the flow is taking place, presence of inverse cone is likely to lead to the formation of sudden contractions and expansions, resulting in turbulence.

The promoter rod diameter being small in comparison with the column diameter, the change in local fluid velocities due to the presence of the promoter rod is marginal. Hence, the average mass transfer coefficient is not likely to be affected by promoter rod diameter.

3.7. Effect of particle diameter

Fig.12 shows the mass transfer coefficient data obtained for the case of an inverse cone promoter element {p = 7 cm; d_r = 1 cm; d_c = 4 cm; θ = 45°} against gas velocity for three different particle diameters viz., 2.942, 4.243 and 5.6 mm. An examination of the plots of this figure indicates that the mass transfer coefficient increased with increase in particle diameter.

As the particle diameter increases, at a given velocity, the momentum of the particle of higher diameter would be high. Hence it strikes the electrode surface with greater momentum thus contributes to the reduction of the thickness of the resistance film. Hence with increase in particle diameter an increase in mass transfer coefficient is observed.

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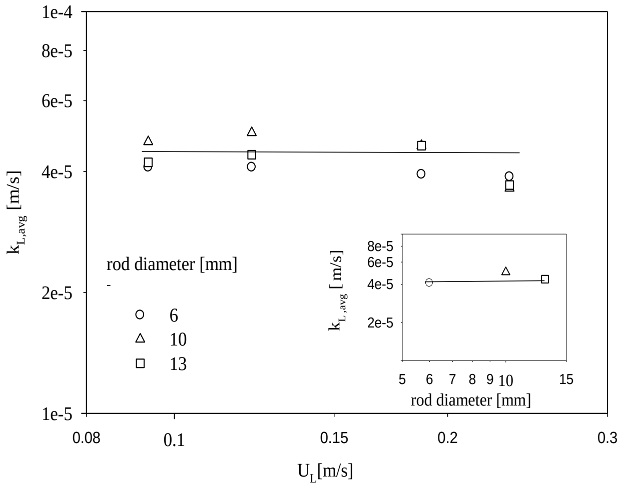

Fig.11. Effect of rod diameter

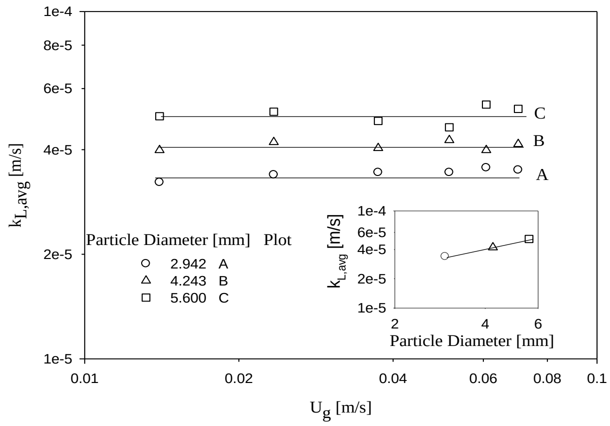

Fig.12 Effect of particle diameter

3.8. Correlation

The entire data on $k_{L,avg}$ obtained in three-phase fluidized bed with inverse cone promoter was correlated with Colburn *j*-factor, particle Reynolds number, Froude number based on superficial gas velocity and the geometrical parameters of the conical element. The following correlation equation is obtained based on least squares regression analysis.

$$J_d = 9.028\,(Re_p)^{-0.9}\left(\frac{P}{D_c}\right)^{-0.26}(1+\sin\theta)^{0.44} \qquad \ldots(4)$$

Average deviation = 14.84 percent

Standard deviation = 19.15 percent

The correlation factor Y is defined as

$$Y_1 = J_d\left(\frac{P}{D_c}\right)^{0.26}(1+\sin\theta)^{-0.44}$$

…(5)

A correlation plot has been drawn by taking Y on Y-axis and Re_p on X-axis. The plot thus obtained is shown presented in Fig.13.

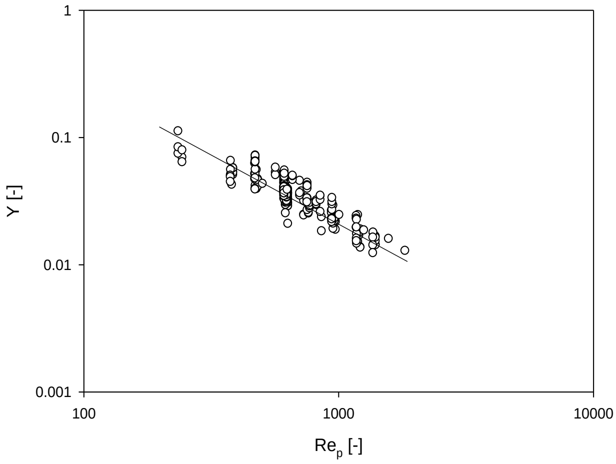

Fig.13. Correlation plot in accordance with eqn.(4)

IV. CONCLUSIONS

The present work is an investigation focused on augmenting mass transfer coefficient in gas-liquid-solid fluidized bed flow reactor in the presence of internal elements. Mass transfer coefficient is obtained for several combinations of dynamic and geometric variables. It was observed that the improvements due to insert promoter in three-phase fluidized beds were up to 20 percent. Axial variation of mass transfer coefficient was noticed and fluctuations were about ±30% for promoter employed in the present study. The mass transfer coefficient was nearly unaffected by the variations in gas and liquid velocities within the range covered in the present study. The mass transfer coefficient decreased when the pitch is varied from 5 to 7 cm and remained constant for further increase in pitch value from 7 to 10 cm. The cone diameter had no effect on mass transfer coefficient. The mass transfer coefficient increased with increase in half-apex angle of cone and particle diameter. Suitable correlation equation has been provided to represent the mass transfer coefficient data obtained in the present study.

NOMENCLATURE

A	= area of the reacting surface	[m^2]
C_0	= concentration of reacting ion	[$kmol/m^3$]
D_c	= diameter of the test section	[m]
D_L	=diffusivity coefficient	[m^2/s]
d_c	= cone diameter	[m]
d_p	= particle diameter	[m]
d_r	= rod diameter	[m]
F	= faraday constant	[C/mol of electrons]
i_L	= limiting current	[A]
k_L	= mass transfer coefficient	[m/s]
p	= pitch	[m]

Q_g =flowrate of gas [m^3/s]
Q_L =flowrate of liquid [m^3/s]
U_g = superficial gas velocity [m/s]
U_L = superficial liquid velocity [m/s]
X = longitudinal distance [m]
z = number of electrons released or consumed per ion during the reaction [-]

Greek Symbols

θ = half-apex angle of cone [degree]
μ_L = Liquid viscosity [kg/m s]
ρ_L = Liquid density [kg/m^3]
ρ_s = Solids density [kg/m^3]

Dimensionless groups

$$j_D = \text{Colburn j-factor} = \frac{k_L}{U_L} Sc^{2/3}$$

$$Sc = \text{Schmidt number} = \frac{\mu_L}{\rho_L D_L}$$

$$Re = \text{Reynolds number} = \frac{\rho_L D c U_L}{\mu_L}$$

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