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STUDIES ON IONIC MASS TRANSFER IN AN ELECTROLYTIC CELL IN THE PRESENCE OF SEMICIRCULAR TEETHED PROMOTERS

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ABSTRACT

Limiting current densities were measured at copper microelectrodes fixed flush with the surface of a rectangular Perspex electrode support firmly fixed in an electrolytic cell. Semicircular teethed promoters were inserted at the bottom of the cell. The electrolyte was equimolal potassium ferricyanide, potassium ferrocyanide and excess sodium hydroxide. The mass transfer data were obtained using diffusion controlled reaction (reduction of ferricyanide ion). The mass transfer coefficient increased with increased flow rate of the electrolyte and height of the promoter and decreased with increase in spacing between the promoters. And the correlation obtained is

$$\mathbf{J_D = 60.1\ R_e^{-0.66}\ (h/s)^{0.13}}$$

KEYWORDS: Limiting current, promoters, and mass transfer coefficient.

INTRODUCTION

Research will always with the aim of the maximum output with reduced equipment size so as to minimize the unit product cost. This is particularly true in the design of electrolytic cells where the mass transfer limiting conditions exist. Use of turbulence promoter is one such technique that increases the mass transfer coefficients by several folds over the smooth flow. The study was conducted to know the performance of the system in augmenting mass transfer coefficient. The process of improving the performance of the system is termed as augmentation or intensification. Different augmentation techniques have been used for improving the ionic mass transfer rates in electrolytic cells. Among the various techniques available to obtain the mass transfer rates in liquids, the electrochemical method is more reliable and accurate.Reports of several authors are as, Venkateswarlu et al.,[1] studied the effect of turbulence promoters on mass transfer rates using the right angle triangular/semicircular cylindrical promoters which were inserted at the bottom of a rectangular electrolytic cell. Their correlation is as: $J_d = 29.6Re^{-0.64}(S/H)^{-0.06}$**.** Vikranth Prithvi [2] studied the influence of geometric variables of promoter along with flow rate of electrolyte on mass transfer coefficients. Square teethed promoters were arranged at the bottom of the electrolytic cell. The correlation proposed was: $J_d = 0.07Re^{-0.7324}(w/s)^{0.207}(h/D_e)^{0.1028}$**.** Bhupesh Mitra [3] conducted experiments in rectangular electrolytic cell to study the enhancement of ionic mass transfer coefficient with square teethed baffles and observed that mass transfer coefficients increases with increase in baffle width. The correlation proposed was: $J_D = 0.0019\ Re^{-0.662}\ (w/s)^{0.175}(h/D_e)^{0.137}$. Sarika [4] studied the mass transfer rates using semicircular teethed baffles as turbulence promoters of different widths and spacing's. The correlation equation as follows: $J_D = 48.7\ Re^{-0.65}\ (w/s)^{0.15}$. Subba Rao and Venkateshwarlu [5] carried out ionic mass transfer studies in an open cell in the presence of circular cylindrical promoters. The data were well correlated using the following equation: $j_d = 16.6Re^{-0.55}(d/s)^{0.045}$. Pickett and Stanmore [6] done experimental investigations in a parallel plate electrochemical cell under both laminar and turbulent flow. The influence of decreased mass transfer at the edges of the electrodes due to changes in the velocity profile was found to be small. Venkata Rao and Venkateswarlu [7] conducted study on vertical disc electrode vibrated in a rectangular electrolytic cell. The mass transfer data were obtained was correlated with the following equation: $J_d = 4.45Re_V^{-0.56}(d_d/d_e)^{0.28}$**.** Rajendra Prasad et al., [8] conducted experiments for model development for coaxially placed entry region coil-disc as turbulence promoter in circular conduit. Experimental mass transfer function in terms of geometric parameters have been developed and presented as below:

$$\bar{g} = 5.76[Re_m^+]^{-0.003}\left(\frac{P_c}{d}\right)^{0.003}\left(\frac{L_c}{d}\right)^{0.001}\left(\frac{d_d}{d}\right)^{0.616}\left(\frac{H_d}{d}\right)^{-0.00001}$$

Mallika Rani [9], Ramya Krishna [10], Padma Priya [11] were studied the mass transfer in the presence of 6-blade flat turbine, Triangle teethed baffles and 4-blade flat turbine in an electrolytic cell respectively.

Yamuna Priya[12] studied mass transfer in an electrolytic cell. The average mass transfer coefficient was derived from experimental data and correlated to obtain the following equation:

$J_D = 51.53Re^{-0.644} (h/s)^{0.22}$. Shrikanth [13] studied combined effect of radial flow 2-blade flat turbine and electrolyte flow on mass transfer performance in an electrolytic cell. The data was correlated by the following equation: $Sh_f/Sc^{1/3} = 0.019\ Re_r^{0.12}\ Re_f^{-0.1}\ (w/d_e)^{0.125}$. Swethasri [14] conducted experiments in the prersence of double stage 2-blade flat turbine in a rectangular electrolytic cell. Proposed correlation was: $J_D = 210.5\ Re_r^{-0.906}\ (d/d_e)^{0.0902}$. Subbaiah et al., [15] obtained mass transfer data by electrolyte circulation, Electrolyte used is cupric sulphate and sulphuric acid. Mass transfer coefficients increased with increase in impeller speed, directly proportional to the power when turbulence is developed though circulation. The data is related as $K_L = 6.81\text{x}10^{-6}\ N^{0.65}$. Ravi et al., [16] obtained mass transfer data on the walls of the fluidized beds with potassium ferri-ferrocyanide couple. Correlation based on regression analysis was: $J_D = 0.6(\frac{Rep}{1-\varepsilon})^{-0.3}(\frac{dp}{dc})^{0.12}\ (\frac{s}{dc})^{-0.135}\ (\frac{dk}{dc})^{0.13}$. Gopala Krishna et al., [17] investigated ionic mass transfer in a circular conduit in the presence of a ring promoter by limiting current technique. The data is correlated by the following dimensionless equation: $J_d = 1.8\ Re^{-0.47}\ (d_R/(d_c\text{-}d_o))^{0.3}(s/(d_c\text{-}d_o))^{-0.15}$. Coeuret and Legrand [18] observed that the local mass transfer coefficient was increased when the annular Reynolds number was increased above 300 on the surface of a rotating inner cylindrical electrode. Satyanarayana et al.,[19] studied the augmentation of mass transfer coefficients in homogeneous flow by inserting spiral tape wound on rod in a circular conduit. $R(h^+) = 2049(Re_m^+)^{0.412}(P/D)^{0.014}(H/D)^{-0.053}(W/D)^{0.128}$ is the developed correlation. Ramesh et al., [20] investigated the wall to bulk mass transfer in two phase gas-liquid up flow bubble columns in the presence of a helicoidal tape promoter. The effects of pertinent geometric and dynamic variables were studied. Correlation proposed in j_D-Re format. $J_D = 110.3(Re)^{-0.8424}(Fr_g)^{0.089}$. Grau and Bisang [21] studied mass transfer at a rotating 35 mm diameter cylinder electrode of woven-wire meshes for the reduction of ferri-cyanide in batch reactor. The data were correlated by the equation: $Sh_d/Sc^{1/3} = 0.967\ (Re_d \times r_2/\bar{r})^{0.58}\ (H/\bar{r})^{0.47}$. EI-Shazly et al.[22] studied the diffusion controlled corrosion of the base of agitated vessels with flat and conical bottom by using diffusion controlled dissolution of copper in acidified dichromate technique. From the experimental data, the correlations obtained were:

$Sh = 0.586\ Sc^{0.33}Re^{0.658}$ (for flat bottom)
$Sh = 0.204\ Sc^{0.33}Re^{0.7}\ (L/r)^{-0.33}$ (for conical bottom)

Afshar et al.,[23] correlated the mass transfer data in turbulent flow for upright rotating cone electrode as $Sh_L = 0.04\ Re^{0.95}$. Deslouis et al.,[24] studied mass transfer in transition and turbulent flows closed to a rotating disk. They correlated the mass transfer data as: $Sh_{av} = 0.00767Re^{0.91}$. Subramaniyan et al.,[25] studied mass transfer rates at spheres, discs and downward facing cones of different sizes rotating about their axis of symmetry in a closed electrolytic cell by employing diffusion-controlled electrode reaction. The data were correlated by the following equations:

$(K_{Lr}/K_{Lo}) = 0.169(\omega sin\alpha/\nu)^{0.5}$ for cones
$(K_{Lr}/K_{Lo}) = 0.169(\omega/\nu)^{0.5}$ for discs and spheres

Asashi Kitamoto and Yoichi Takashima [26] studied the rate of ionic mass transfer at the limiting current density which was measured in two-dimensional flows both laminar and turbulent, through a channel between a pair of ion-exchange membranes. For laminar flow, the experimental results showed good agreement with those of numerical analysis, in which the rate was assumed to be controlled by the boundary layers produced near the membrane surfaces. For turbulent flow, the analytical results were also brought into good agreement with the experimental results, by introducing a new concept of the "eddy migration coefficient.

Lin et al., [27] obtained design information for processes involving mass transfer, including many electrochemical processes. Transfer rates from a flowing electrolyte to the surface of an electrode were measured. The transfer rates for the cathodic reduction of ferricyanide ion, quinone, and oxygen, and for the anodic oxidation of ferrocyanide at various temperatures and flow rates. In a stagnant solution, the steady rate of discharge for a certain species of ion can be expressed as follows:

$$\frac{i}{nFA} = -D_L\frac{\partial C}{\partial x} - Cu\frac{\partial \Phi}{\partial x}$$

Eisenberg et al.,[28] studied ionic mass transfer at nickel electrode rotating about their own axes in the Centre of the stationary electrode using ferri-ferro cyanide couple in alkaline solution. They proposed the following correlation:

$$J_D = (K_L/V_r)Sc^{2/3} = 0.0791Re_r^{-0.3} \qquad (1000<Re_r<100000)$$

EXPERIMENTATION

The limiting current data are measured at microelectrodes (point electrodes) fixed flush with the surface of the electrode support for the reduction of ferricyanide ion using electrochemical method. The following electrode reactions are involved for the cathodic reduction of ferricyanide ion and anodic oxidation of ferrocyanide ion.

$$[Fe(CN)_6]^{3-} + e^- \longrightarrow [Fe(CN)_6]^{4-}$$
$$[Fe(CN)_6]^{4-} \longrightarrow [Fe(CN)_6]^{3-} + e^-$$

The dimensions of the cell promoters and electrode support are accurately measured. The microelectrodes on the electrode support and cell wall are polished with zero emery paper to obtain a smooth surface. It is degreased with dilute hydrochloric acid followed by through washing with distilled water and acetone. The diameters of the microelectrodes are measured with a traveling microscope. The electrode support is fixed at a distance of 0.28 m from the cell entrance and is situated at 0.04 m height from the cell bottom. The copper anode is also firmly fixed in the cell at a distance of 0.05 m from the discharge end of the cell. 100 liters of the electrolyte, consisting of equimolal 0.01N potassium ferricyanide, 0.01N potassium ferrocyanide and 0.5N sodium hydroxide, is prepared in the storage tank (ST). Excess sodium hydroxide is used as a supporting electrolyte to suppress the migration of the reacting ion and to make the reaction diffusion controlled. Electrolyte is analyzed for ferrocyanide ion concentration by permanganate titration and ferricyanide ion by iodometric titration. The electrolyte is pumped to the electrolytic cell from the storage tank through calibrated rotameters at a desired flow rate through the electrolytic cell by operating the control and by-pass valves (V_2 and V_3). The flow rate of the electrolyte is measured with a rotameter (R_1 or R_2). The electrolyte enters through the inlet pipe and spreads gradually over the entire cross section, the liquid level being built up progressively. The electrolyte is discharged through the outlet pipes at the exit of the cell in to the storage tank and gets recirculated. After the steady flow rate is attained, a potential is applied across the microelectrode on the electrode support and copper electrode in small increments (100 mV) and the corresponding current is measured for each increment. The attainment of limiting current is indicated by the negligible change in current for a considerable increase in the potential. The limiting current data are obtained at different microelectrodes on the electrode support at semicircular teethed promoters for reduction of ferricyanide ion to ferrocyanide ion for varying flow rates, for fixed promoter height and varying promoter spacings. The experiments are repeated for different heights and spacings.

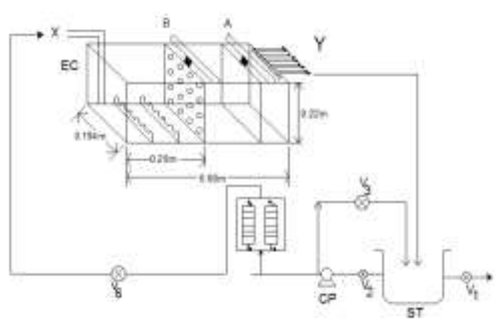

Figure.1: Schematic diagram of the experimental set-up

EC – Electrolytic cell, A- Copper anode, B-Electrode support,
X-Inlet flow of electrolyte, Y- Outlet flow of electrolyte, ST – Storage Tank,
($V_1 – V_6$) – Control Valves

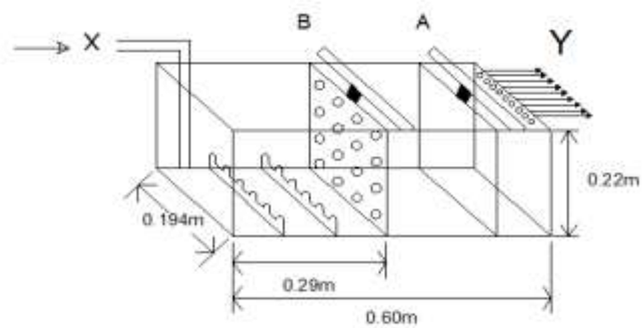

Fig.2: Details of the electrolytic cell

A- Copper anode, B-Electrode support,
X-Inlet flow of electrolyte, Y- Outlet flow of electrolyte

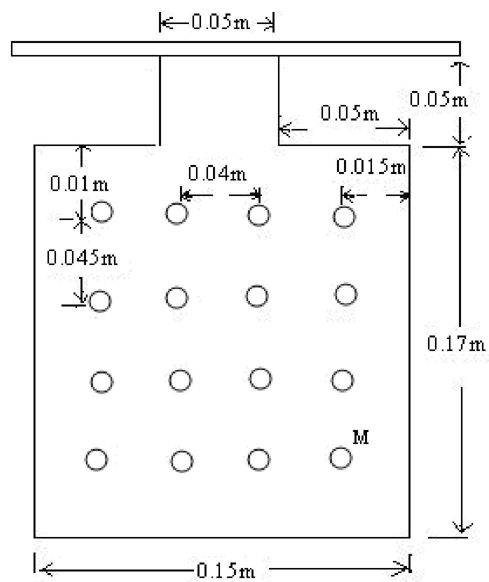

Fig.3: Details of the electrode support

M-Micro electrode

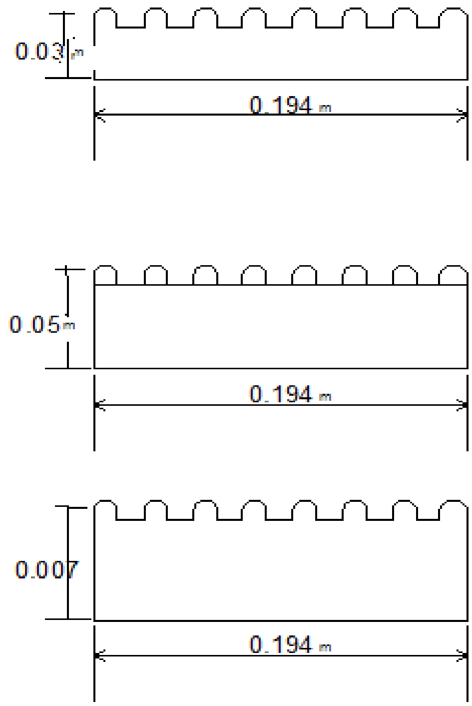

Fig.4: Details of semicircular teethed promoters

RESULTS AND DISCUSSIONS

Experimental investigations have been made on ionic mass transfer in the presence of semicircular teethed promoters in a rectangular electrolytic cell under both laminar and turbulent flow. The semicircular teethed promoters of heights h = 0.03m, 0.05m and 0.07m are fixed rigidly and symmetrically on the bottom of the cell at equal spacing. The electrode support is fixed rigidly in the cell with the point electrodes perpendicular to the electrolytic flow. Limiting current measurements are obtained at sixteen point electrodes fixed on a rigid electrode support, for the reduction of ferricyanide ion.

The limiting current data have been obtained at point electrodes for the following variables are the flow rate of the electrolyte, height of the semicircular teethed promoter and spacing between the promoters.

The average mass transfer coefficient (k_{Lavg}) is obtained by calculating the simple arithmetic average of local mass transfer coefficients on electrode support. The improvement in limiting current density, and hence mass transfer coefficient, due to the presence of turbulence promoters is assessed. The effects of the flow rate of the electrolyte, height of the promoters and spacing between the promoters were studied.

General Observations:

In the present study, the electrolyte from the inlet pipe impinges on the cell bottom and spreads gradually over the entire cell bottom. The liquid head is developed and the electrolyte is discharged through the outlets provided at the other end of the cell. Presence of semicircular teethed promoters at the cell bottom creates disturbance in the flow path, where the turbulence is created at the bottom of the cell and spread on to the electrode support. The electrode support also blocks the flow of electrolyte and changes the flow patterns around it. The turbulence created by the promoters along with that created on the electrode support lead to higher mass transfer rates.

Measurement of limiting current data:

Sixteen point electrodes on the electrode support were fixed, column wise, in four rows as shown in fig. 3.3. The electrodes 1, 2, 3 and 4 constitute the row-I, rows II, III and IV consist of the electrodes 5 to 8, electrodes 9 to 12 and electrodes 13 to 16 respectively. The limiting current data are taken at all the sixteen electrodes for varying velocity of the electrolyte (v_f), height of the promoter (h) and spacing between the promoters(s) systematically.

The arithmetic averages of the limiting current densities at all point electrodes on the electrode support are computed and the average value is termed as average limiting current density.

Effect of height of the Semicircular teethed promoters:

Variations of average mass transfer coefficient with different values of semicircular teethed promoter's height are shown in fig. 5(a)-5(b). These plots indicate that mass transfer coefficient increases, as the height of the teethed promoters increases. Increase in the height of the promoter causes decrease in the available cross-sectional area for the flow, with an increase in local velocities, resulting in increase of mass transfer coefficient.

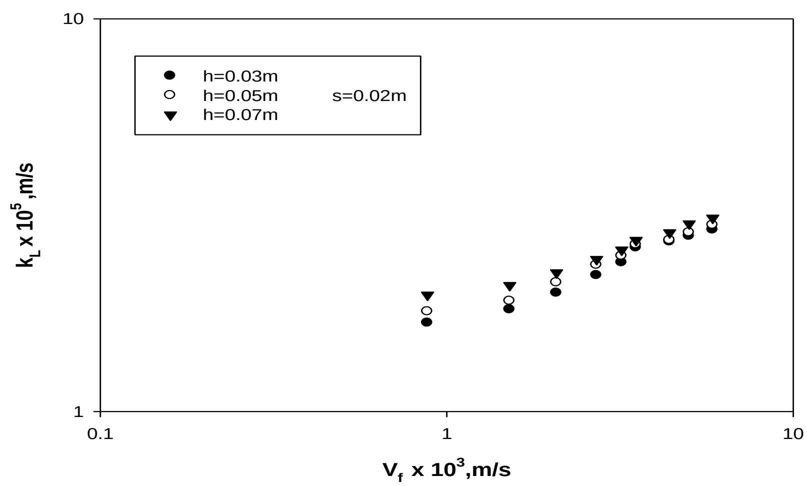

Fig.5(a) : Effect of velocity of electrolyte on average mass transfer coefficient for various heights of semicircular teethed promoters.

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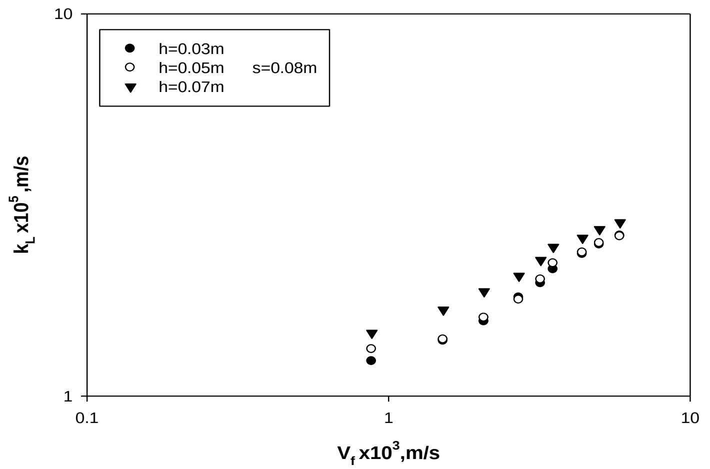

Fig.5(b) : Effect of velocity of electrolyte on average mass transfer coefficient for various heights of semicircular promoters.

Effect of spacing between the Semicircular teethed promoters:

The average mass transfer coefficient is plotted against the spacing between the teethed promoters (s). These plots were shown in fig. 5(c)-5(d). These plots indicate that k_{Lavg} decreases as the spacing between the teethed promoters increases. It is found that k_{Lavg} is inversely proportional to spacing. At larger spacing the intensity of turbulence is away from the teethed promoters, and results in low mass transfer coefficient.

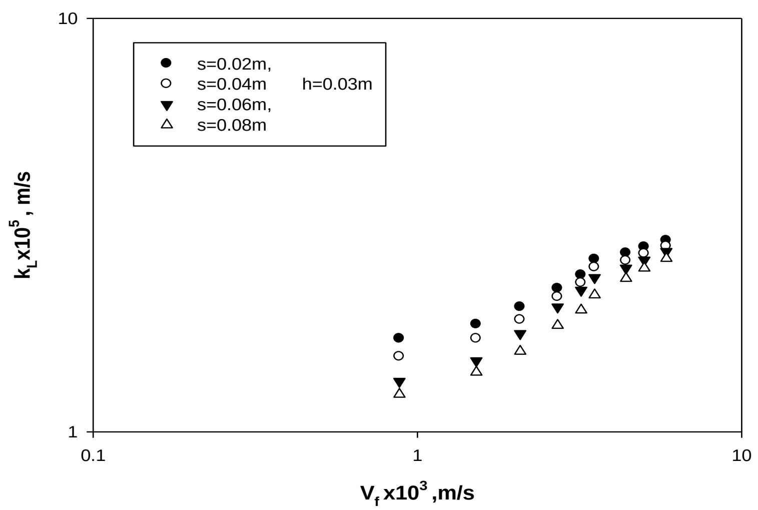

Fig.5(c) : Influence of velocity of electrolyte on average mass transfer coefficient for various spacings of the semicircular teethed promoters.

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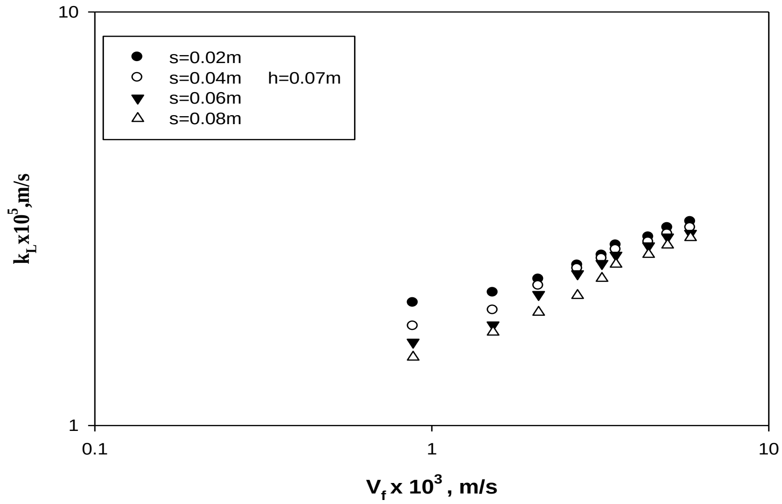

Fig.5(d) : Influence of velocity of electrolyte on average mass transfer coefficient for various spacings of the semicircular teethed promoters.

Augmentation of the mass transfer coefficient:
The mass transfer coefficient obtained in stagnant electrolyte condition (k_{L0}) is $0.3x10^{-5}$ m/s [27]. The enhancement of mass transfer coefficient in the present study is expressed as (k_{Lavg}/ k_{L0})-1. The present data are expressed with [(k_{Lavg}/ k_{L0})-1] as a function of velocity of electrolyte (v_f) for two sets of the data representing maximum mass transfer coefficients (h = 0.07 m, s = 0.02 m) and minimum mass transfer coefficient (h = 0.03 m, s = 0.08 m) as shown in Fig.6. The average mass transfer coefficient is increased over the coefficient in stagnant electrolyte by 3 to 9 folds over the range of variables studied.

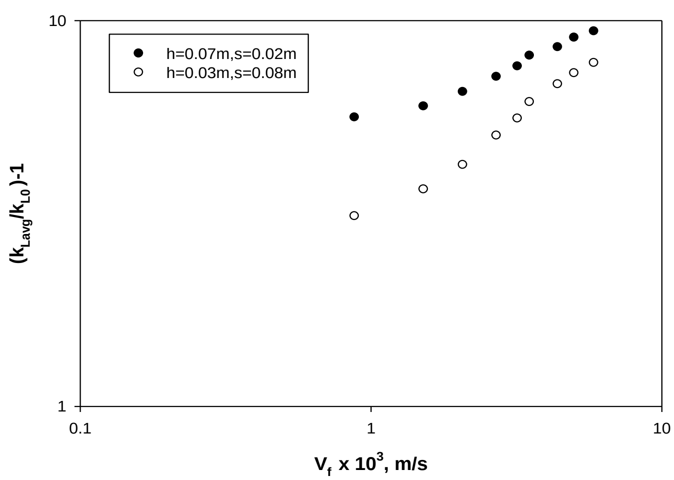

Fig.6: Augmentation of average mass transfer coefficient in the presence of semicircular teethed promoters over the stagnant electrolyte

Development of generalized correlation:
It is observed that the average mass transfer coefficient is proportional to height of the promoter (h) and inversely proportional to spacing between the promoters (s). Therefore, the average mass transfer coefficient is a function of (h/s).

To account for the physical properties of the electrolyte, the average mass transfer coefficient is presented as mass transfer factor J_D and flow velocity is represented as Reynolds number Re. Figs. 7(a) and 7(d) drawn as J_D against Re for different values of height and spacing of the promoter. Considering the variation of mass transfer factor with (h/s), the present experimental data are correlated in the following format of the equation:

$$J_D = C_1 Re^a (h/s)^b$$

The regression analysis yielded the following equation with an average deviation of 5.819 % and Standard deviation of 7.748 %.

$$J_D = 60.1 Re^{-0.66}(h/s)^{0.13}$$

The mass transfer factor data is plotted as a function of Reynolds number in figs. 7(a)-7(d).

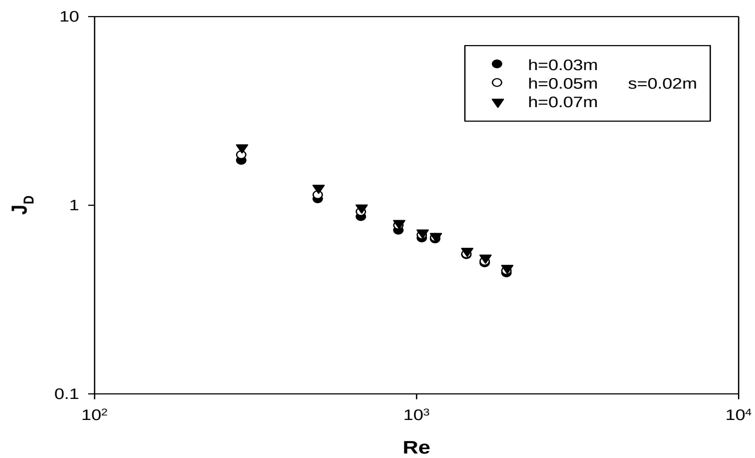

Fig.7(a): Impact of Reynolds number on mass transfer factor -Effect of height of the promoter.

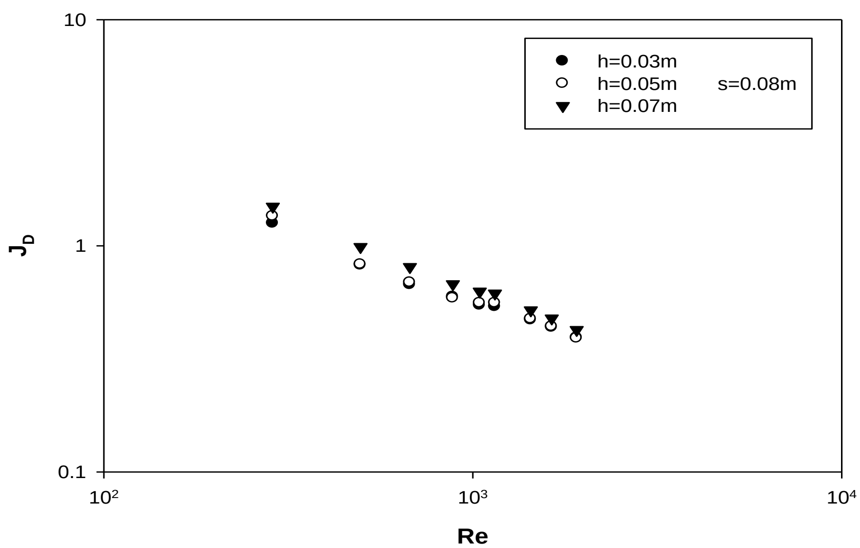

Fig.7(b) : Impact of Reynolds number on mass transfer factor - Effect of height of promoters.

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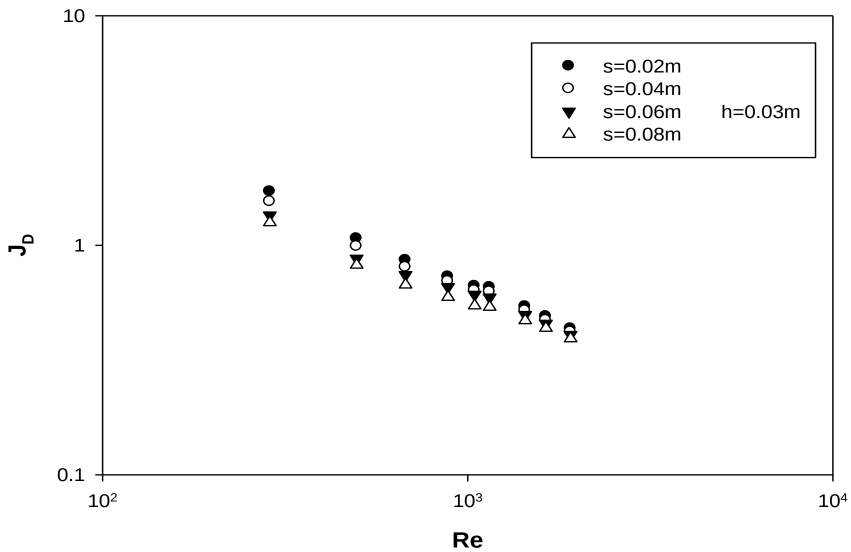

Fig.7(c) : Impact of Reynolds number on mass transfer factor
- Effect of spacing between the promoters.

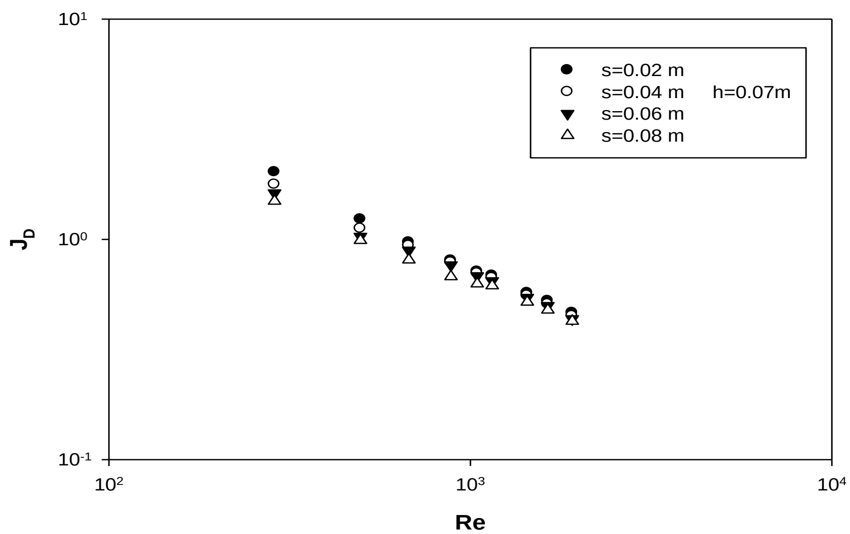

Fig.7(d) : Impact of Reynolds number on mass transfer factor
- Effect of spacing between the promoters.

The data for the above correlation is plotted and shown in fig.8.

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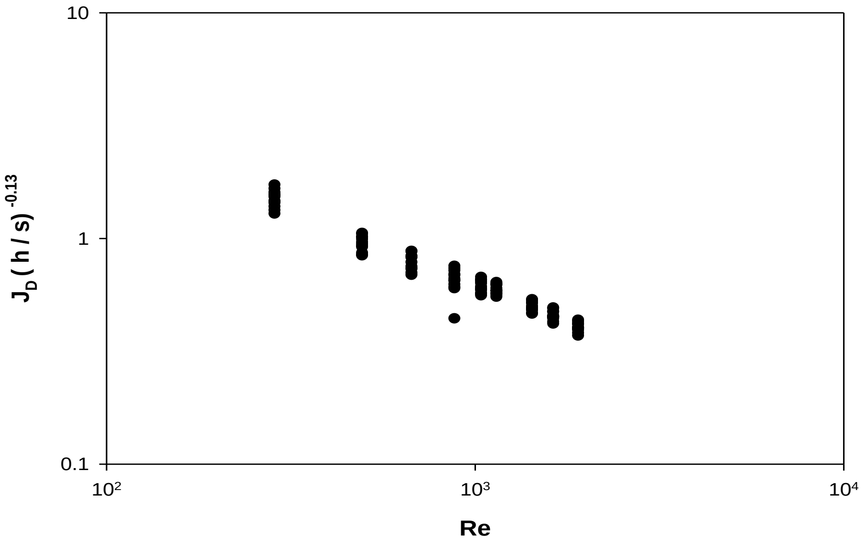

Fig.8: Correlation plot for mass transfer data

Comparison with other investigations:
Fig.9 shows the comparison of the present data with that obtained by Sarika [4] using semicircular teethed baffles. The mass transfer factors obtained in the present study are 82.22 % higher than that obtained by semicircular teethed baffles. This can be attributed to the less turbulence generated by baffles and higher turbulence generated by teethed promoters fixed at the bottom of the cell.

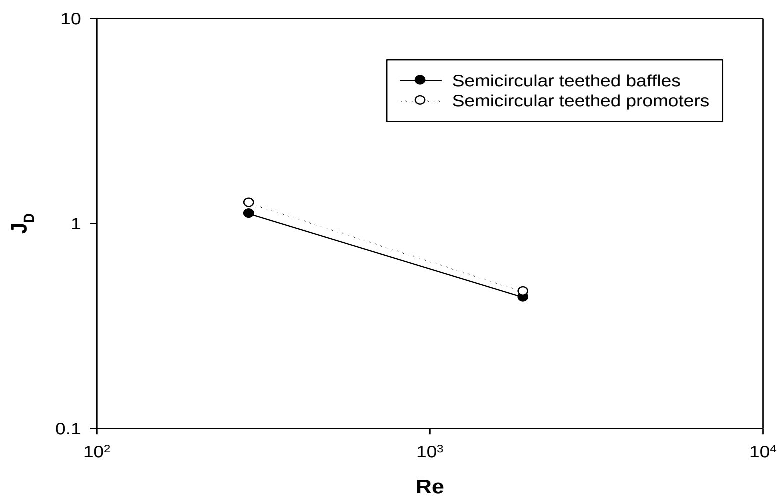

Fig.9: Comparison of data between semicircular teethed promoters and semicircular teethed baffles.

The data obtained in the present study are compared with Vikranth Prudhvi [2] obtained using square teethed promoters fixed at the bottom of the electrolytic cell. The plot is shown in fig.10. It indicates that there is an enhancement in mass transfer coefficient. The mass transfer data for present study are higher than that obtained for square teethed promoters by 85% in the range of variables covered.

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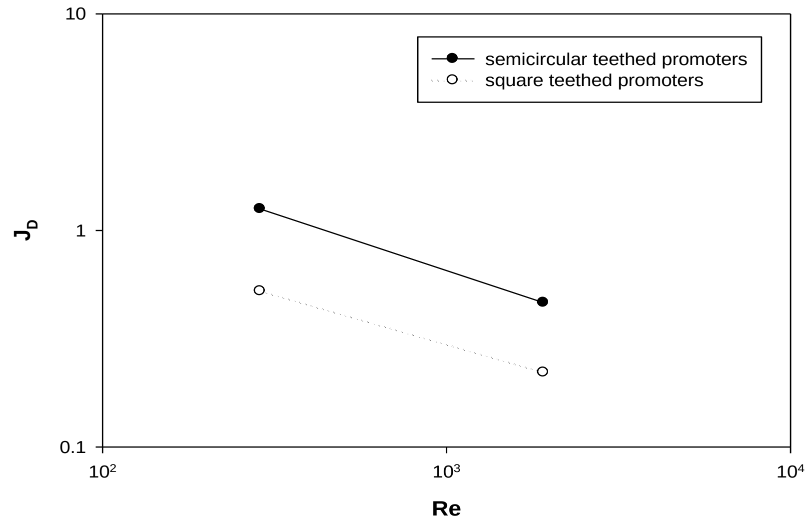

Fig.10 : Comparison of data between semicircular teethed promoters and square teethed promoters.

The present data are plotted in fig.11 against the data obtained for square teethed baffles by Bhupesh [3] and it indicates that the present mass transfer data obtained are 83 % higher than that of square teethed baffles.

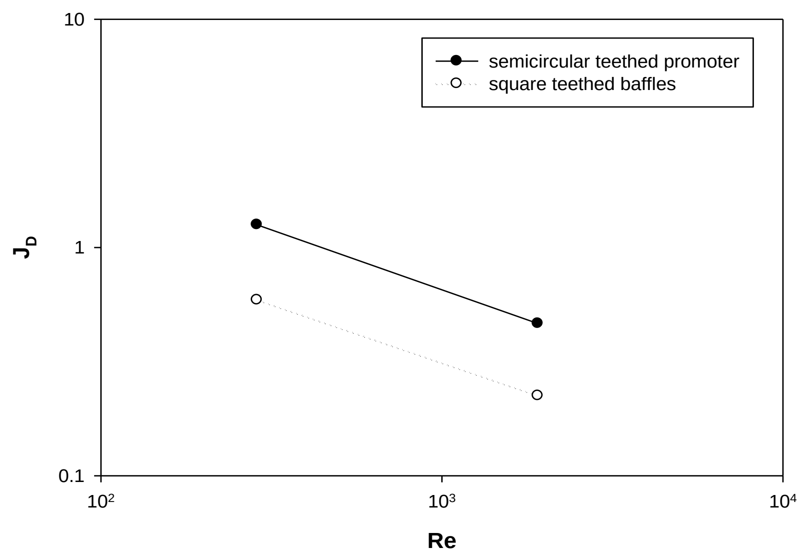

Fig.11: Comparison of data between semicircular teethed promoters and square teethed baffles.

CONCLUSIONS

The mass transfer data are obtained on an electrode support present in an open electrolytic cell when semicircular teethed promoters are fixed on the wall of the cell. The following conclusions were drawn from the analysis of the experimental data:

[1] Higher mass transfer coefficients are obtained with increased flow rate of the electrolyte.
[2] The mass transfer coefficients increased with increase in height of the promoter. However, the coefficients decreased as the spacing between the promoters is increased.
[3] In the presence of promoters, the augmentation in mass transfer coefficient is 3to 9 fold over that of stagnant electrolyte.
[4] The mass transfer data obtained in the presence of semicircular teethed promoters are correlated by the following equation:

$J_D = 60.1\ Re^{-0.66}(h/s)^{0.13}$

Nomenclature

A	:	Cross sectional area of the electrode, m^2
C_o	:	Concentration of the ferrocyanide ion, Kg moles/m^3
C_i	:	Concentration of the ferricyanide ion, kg mole/m^3
D_L	:	Diffusivity, m^2/s
d_e	:	Equivalent diameter of the electrolytic cell, m
F	:	Faraday's constant, coulombs
h	:	Promoter height, m
H	:	Height of the electrolytic cell, m
I	:	Limiting current, A
i_d	:	Limiting current density, A/m^2
i_{dav}	:	Average limiting current density, A/m^2
K_L	:	Local mass transfer coefficient, m/s
K_{Lavg}	:	Average mass transfer coefficient, m/s
L_b	:	Length of the baffle, m
n	:	Number of electrons transferred in the reaction
Q	:	Volumetric flow rate, m^3/s
v_f	:	Velocity of the electrolyte, m/s
s	:	Spacing between the baffle, m
W	:	Width of the electrolytic cell, m
w	:	width of the baffle, m
Y	:	Co-ordinate along channel length, m
Z	:	Dimensionless duct length, m
ρ	:	Solution density, (Kg/m^3)
μ	:	Viscosity of the electrolyte, Kg/m.s
Re	:	Reynolds number, $\rho\, d_e v_f/\ \mu$
J_D	:	Mass transfer factor, $(k_{Lav} / v_r)\ Sc^{2/3}$
J_d	:	Mass transfer factor, $(k_{Lav} / u_f)\ Sc^{2/3}$
Sc	:	Schmidt number, $\mu/\rho D_L$

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