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Effect of Surface Roughness on the Rate of Mass Transfer at a Vertical Cylinder Under Swirl Flow in Relation to Electrochemical and Catalytic Reactor Design

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Abstract

In order to enhance the rate of production of catalytic and electrochemical annular reactors used to conduct diffusion controlled reactions the effect of surface roughness on the rate of mass transfer at the inner cylinder of the annulus was studied under swirl flow. The effect of the following parameters on the rate of mass transfer was studied by the electrochemical technique: solution velocity, degree of roughness, physical properties of the solution, height of the inner cylinder and effect of drag reducing polymers. Roughness was made by cutting longitudinal V grooves in the inner cylinder transverse to swirl flow. The mass transfer data at the rough cylinder were correlated by the dimensionless equation:

$$Sh=0.435\ Sc^{0.328}\ Re^{0.86}\ (L/de)^{-0.53}\ (e/de)^{0.45}$$

Drag reducing polymers were found to decrease the rate of mass transfer by an amount ranging from 5 to 23%. Importance of the present results for the design and operation of high space-time yield electrochemical reactors and biochemical reactors employing immobilized enzymes was pointed out. Also, the importance of the present results in the design and operation of membrane processes employing annular apparatus with a corrugated membrane was highlighted. By virtue of the analogy between heat and mass transfer the present results can be used to design more efficient double tube heat exchangers.

Keywords: Electrochemical reactors; Mass transfer; Catalytic reactors; Heat transfer; Swirl flow; Surface roughness; Diffusion controlled reactions

Introduction

Annular flow reactors are used frequently in industry to conduct liquid-solid diffusion controlled catalytic and electrochemical reactions in view of their advantages which include: (i) Rapid removal of heat generated in case of highly exothermic reactions by passing cold water through the inner side of the inner tube and the cooling jacket surrounding the outer tube. The built in heat transfer facility makes it possible to use the reactor to conduct exothermic reactions involving heat sensitive catalysts or heat sensitive products such as immobilized enzyme catalyzed biochemical reactions. (ii) Uniform current and potential distribution, this property is highly desirable especially in case of electro organic synthesis where a high degree of selectivity is needed. (iii) The reactor occupies a low floor space and can be extended vertically thus saving floor space and capital costs.

Despite these merits the annular flow reactor suffers from the limited surface area which limits the reactor rate of production. To make up for the limited reactor area previous studies tried to increase the rate of mass transfer by a suitable means such as gas sparging [1], vibration and pulsation [2], inner tube rotation [3], gas generation [4], surface roughness [5,6], and swirl flow [7-11].

The aim of the present work is to enhance the rate of mass and heat transfer in the annular reactor by the combined swirl flow and surface roughness. Although some work has been done on the mass transfer behavior of smooth annular reactor under swirl flow [7-8] no studies have been reported on the effect of combined surface roughness and swirl flow on the rate of heat or mass transfer in an annular reactors. The effect of surface roughness on the rate of mass transfer has been studied for annular reactors only in axial flow [5,6], surface roughness would not only increase the rate of heat and mass transfer by promoting turbulence but would also increase the effective area of the reactor with a consequent increase in the rate of production. Roughnesses used in the present work are in the form of longitudinal parallel grooves transverse to the direction of swirl flow. The rate of mass transfer was determined by an electrochemical technique which involves measuring the limiting current of the catholic reduction of $K_3Fe(CN)_6$ in a large excess of NaOH as supporting electrolyte [12]. The present mass transfer study is important for the rational design and operation of relatively high space-time yield annular reactor suitable for conducting liquid-solid diffusion controlled reactions such as catalytic reactions which include photo catalytic reactions, immobilized enzyme catalyzed biochemical reactions and electrochemical reactions involved in waste water treatment and electro organic synthesis. The area of membrane technology can benefit from the present work in designing high efficiency annular dialyzers by using swirl flow and corrugated cylindrical membranes. Drag reducing polymers are used in practice to decrease the pumping power requirement of turbulent flow equipments in view of the ability of polymer molecules to damp the small scale high frequency energy dissipating eddies which prevail in the buffer layer of the hydro dynamic boundary

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layer [14]. Damping of these eddies leads also to decreasing the rate of turbulent flow heat and mass transfer, hence the second aim of the present work is to find the extent to which rates of mass transfer are reduced under swirl flow in view of fact that no previous studies have been reported on the effect of drag reducing polymers on the rate of mass transfer under swirl flow. Polyethylene oxide drag reducing polymer (POLOX WSR-301) a product of Union Carbide was used in the present study.

Experimental Techniques

The Figures 1a and 1b shows the apparatus used in the present study and the swirl inlet nozzle dimensions respectively. The apparatus consisted of Plexiglass cylindrical reactor of length 40 cm and a diameter of 5.0 cm lined with a cylindrical stainless steel sheet which acted as anode and an inner cylinder of nickel plated copper tube of diameter 2.5 cm which acted as cathode. Smooth and rough cylinders were used, peak to valley height of the roughness elements were: 0,1, 2, 3, 4 mm. Roughness was made by cutting V-threads (angle 45°) in the inner cylinder. The annulus was fed with the solution by a tangential feed nozzle of 1.2 cm diameter fitted at the lower end of the reactor to generate swirl flow. A plastic centrifugal pump of 0.27 horse power was used to circulate the solution between a glass storage tank of dimensions 30 × 30 × 30 cm and the reactor. The top of the reactor was fitted with an overflow weir from which the solution return to the storage tank. A bypass with plastic valves was used to regulate the solution velocity in the reactor.

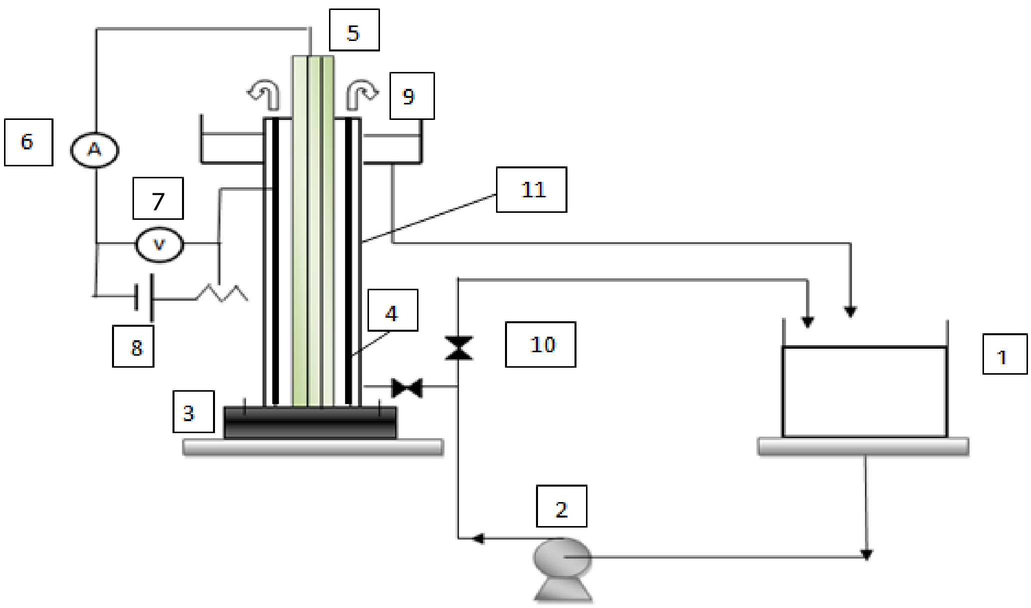
The electrical circuit consisted of 10 volt DC power source with a voltage regulator and a multi-range ammeter connected in series with the reactor. A voltmeter was connected in parallel with the reactor to measure its voltage. Before each run 25 L of the solution were placed in the plastic storage tank, the solution was circulated between the storage tank and the reactor by the centrifugal pump. Solution velocity in the reactor was controlled by the by-pass and was measured volumetrically using a graduated cylinder and a stop watch. The mass transfer behavior of reactor was studied by measuring the limiting current of the catholic reduction of $K_3Fe(CN)_6$ from a solution containing $K_4Fe(CN)_6$ and a large excess of NaOH as supporting electrolyte [12]. Polarization curves from which the limiting current was obtained were plotted by increasing the current stepwise and measuring the corresponding cathode potential against a reference electrode. In view of high anode area compared to the cathode area, the anode was taken as a reference electrode [13].

The following reactions take place during current measurement:

Cathode: $Fe(CN)_6^{-3} + e \rightarrow Fe(CN)_6^{-4}$

Anode: $Fe(CN)_6^{-4} \rightarrow Fe(CN)_6^{-3} + e^-$

All solutions used in the present work were prepared using A.R grade chemical and distilled water, temperature was 25 ± 1°C solution composition and their physical properties are shown in Table 1. Solution density and viscosity needed for determined by a density bottle and an Ostwald viscometer [14], while diffusivity of the $Fe(CN)_6^{-3}$ was obtained from the literature [12]. Each experiment was repeated twice under the same conditions, the reproducibility ranged from 3-5%.



Experimental setup

1-Storage tank
2-Plastic Centrifugal pump
3-Plastic base
4-Stainless steel cylindrical anode
5-Nickel Plated Copper Cylinder cathode
6-Digital ammeter
7-Digital voltmeter
8-DC power supply
9-Overflow weir
10-Plastic valve
11-Plexiglass cylinder lined with cylindrical stainless steel sheet anode

Figure 1a: Apparatus.

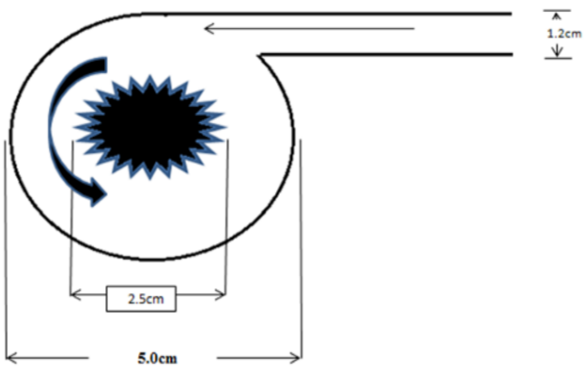


Figure 1b: Swirl inlet nozzle.

Solution Composition	ρ (g/cm³)	μ × 10² (Poise)	D × 10⁶ (cm²/s)	Sc
0.02M $K_3Fe(CN)_6$+0.2M $K_4Fe(CN)_6$+1N NaOH	1.083	1.228	6.693	1694
0.02M $K_3Fe(CN)_6$+0.2M $K_4Fe(CN)_6$+2N NaOH	1.134	1.326	5.508	2122
0.02M $K_3Fe(CN)_6$+0.2M $K_4Fe(CN)_6$+3N NaOH	1.14	1.599	4.335	3072

Table 1: Physical properties of ferri/ferrocyanide system used at 25°C.

Results and Discussion

Swirl flow mass transfer at smooth cylinder cathode

Polarization curves with a well-defined limiting current plateau were obtained under different conditions as shown in Figure 2, the mass transfer coefficient was calculated from the limiting current using the equation [14]:

$$I_L/ZF = KAC \tag{1}$$

Dimensional analysis which was used to correlate the present data leads to the following functional equation for smooth surface

$$K = f(\rho, \mu, D, V, de, L) \tag{2}$$

Writing the above equation in a dimensionless form gives

$$\frac{Kde}{D} = \alpha\left(\frac{\mu}{\rho D}\right)^{\alpha}\left(\frac{\rho Vde}{\mu}\right)^{\beta}(L/de)^{\gamma} \tag{3}$$

i.e.,

$$Sh = a\, Sc^{\alpha} Re^{\beta} (L/de)^{\gamma} \tag{4}$$

The equivalent diameter of the annulus de was calculated from equation

$$de = d_{outer} - d_{inner} \tag{5}$$

The present experimental data will be used to determine a, α, β and γ. The Figure 3 Shows the effect of Re on Sh for different Sc, the data fit the equation

$$Sh = \bar{a}\, Re^{0.96} \tag{6}$$

The Re exponent 0.96 from equation (6) reveals mass transfer by highly turbulent flow mechanism which characterize swirl flow. The Figure 4 shows the effect of the dimensionless active cathode height on Sh, the data fit the equation:

$$Sh = a_1\,(L/de)^{-0.432} \tag{7}$$

The decrease in Sh with cylinder height is attributed to the decay of swirl flow in favor of the less effective axial flow as the solution progresses through the annulus. The Figure 5 shows the effect of Sc on Sh, the data fit the equation:

$$Sh = a_2 Sc^{0.328} \tag{8}$$

The exponent 0.328 agrees with the value 0.33 predicted by the hydrodynamic boundary layer theory [15]. The Figure 6 shows that the mass transfer data for the conditions:

$1694<Sc<3072$, $1130<Re<6328$ and $2.73<L/de<8.2$ fit the eqn:

$$Sh = 0.109\, Sc^{0.328} Re^{0.96} (L/de)^{-0.432} \tag{9}$$

With an average deviation of 10.5%.

The Figure 7 shows a comparison between the present swirl flow data and the axial flow data obtained [6] for mass transfer under developing laminar and turbulent flow in an annulus. The present swirl flow data lie above the axial flow data of Mobarak in the laminar and the turbulent flow region. The Table 2 shows that the mass transfer enhancement ratio Sh_{swirl}/Sh_{axial} ranges from 1.4 to 1.74 depending on the operating conditions.

The higher rates of swirl flow mass transfer compared to axial flow at smooth cylinder shows that for a given set of conditions turbulence generated in case of swirl flow is more intense than that generated in case of axial flow. Figure 8 shows that the present swirl flow data at smooth cylinder agree fairly with the data of legentihomme and Legrand [9] and the data of de Sa et al. [7].

Mass transfer at rough cylinders under swirl flow

Following previous studies in heat and mass transfer at rough surface [16-20] geometrical area rather than true area was used in calculating the mass transfer coefficient at rough surfaces. The Figures 8 and 9 shows that the effect of Re on the mass transfer coefficient at rough cylinders fit the eqn:

$$Sh = a_3\, Re^{0.862} \tag{10}$$

The Figure 10 shows that the effect of rough cylinder dimensionless height (L/de) on the mass transfer coefficient, the data fit the eqn:

$$Sh = a_4\,(L/de)^{-0.53} \tag{11}$$

The Figure 11 shows the effect of the ratio between the dimensionless peak to valley height (e) of the roughness element and the equivalent diameter (de) on the mass transfer coefficient, the data fit the eqn:

$$Sh = a_5\,(e/de)^{0.4} \tag{12}$$

The Figure 12 shows that the swirl flow data at rough cylinders for the conditions:

$1694<Sc<3072$, $1130<Re<6328$, $2.7<L/de<8.2$, $0.04<e/de<0.16$ fit the eqn:

$$Sh = 0.435\, Sc^{0.328} Re^{0.86} (L/de)^{-0.53} (e/de)^{0.45} \tag{13}$$

In obtaining the above equation the exponent of Sc was fixed at 0.328 according to equation) of present work. To appreciate the extent to which surface roughness increases the rate of mass transfer compared to smooth cylinders under swirl flow, the enhancement factor Sh_{rough}/Sh_{smooth} was calculated under different conditions. The Table 3 shows that for a given set of conditions surface roughness increases the rate of mass transfer by a factor ranging from 1.02 to 1.94 depending on the operating conditions.

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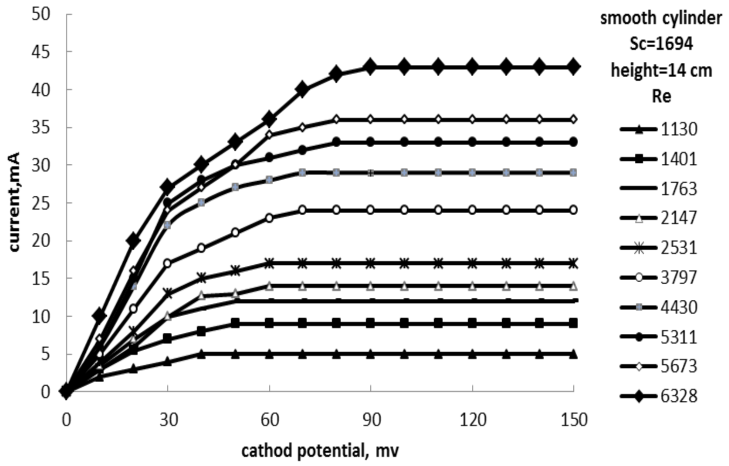

Figure 2: Typical polarization curve for smooth cylinder at different Re.

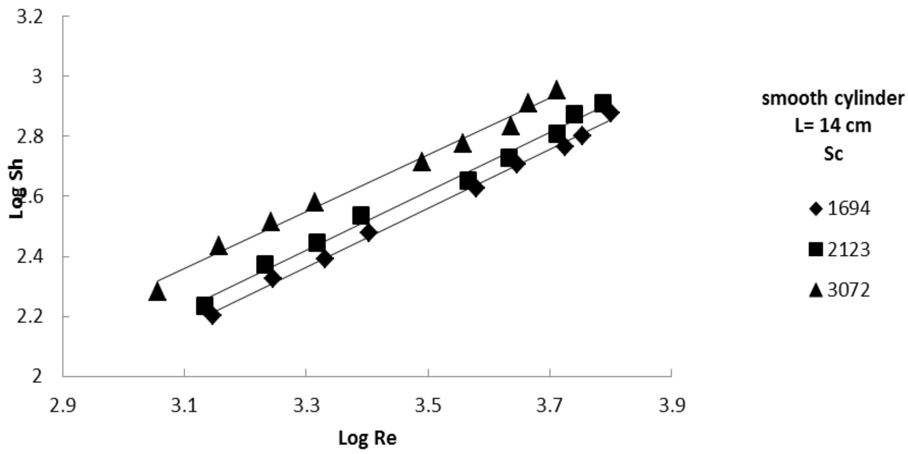

Figure 3: Effect of Re on Sh for a smooth cylinder at different Sc.

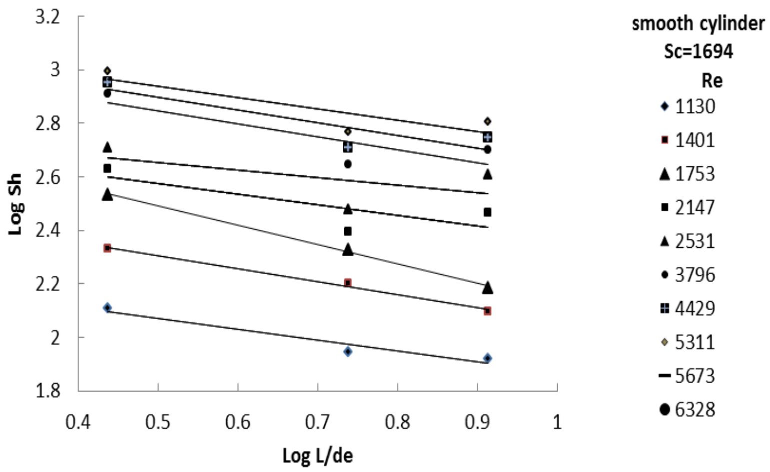

Figure 4: Effect of smooth cylinder height on Sh at different Re.

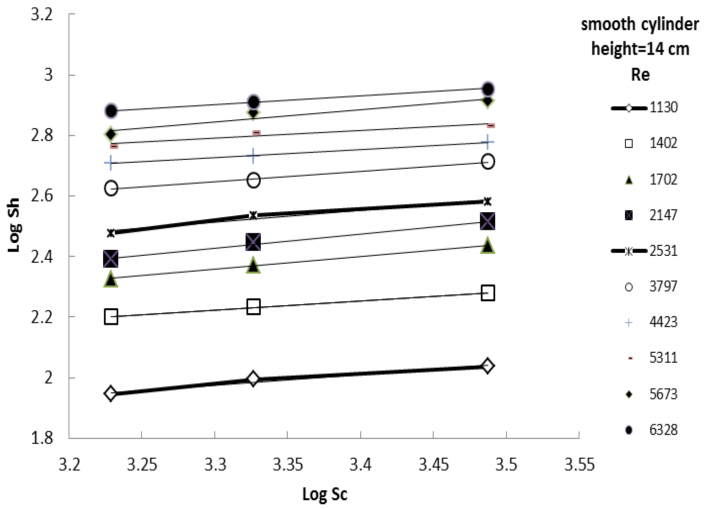

Figure 5: Effect of Sc on Sh at different Re for smooth cylinder.

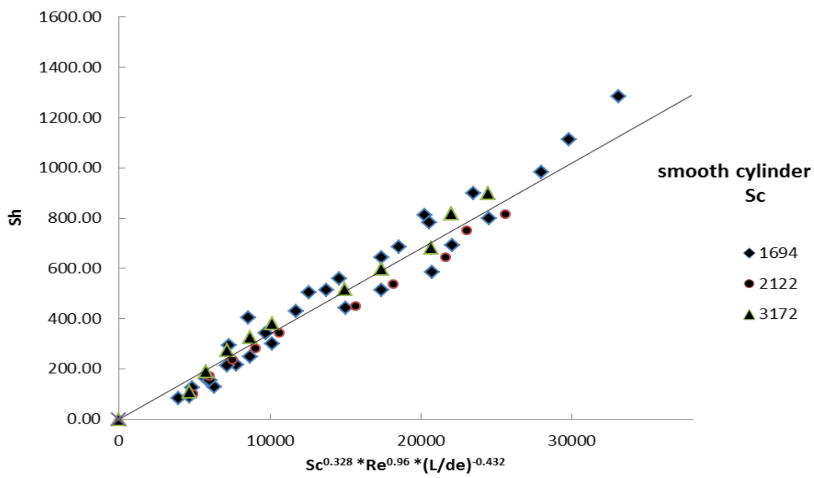

Figure 6: Overall mass transfer correlation at a smooth cylinder under swirl flow.

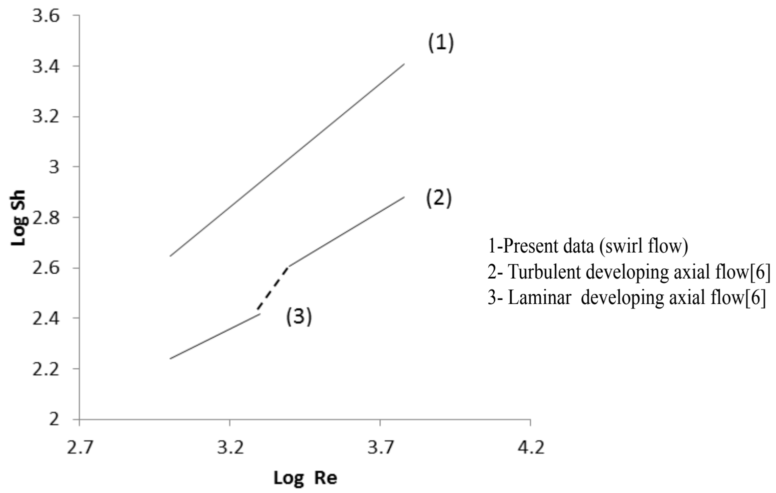

Figure 7: Comparison of the present swirl flow mass transfer coefficient with the value of axial flow in an annulus.

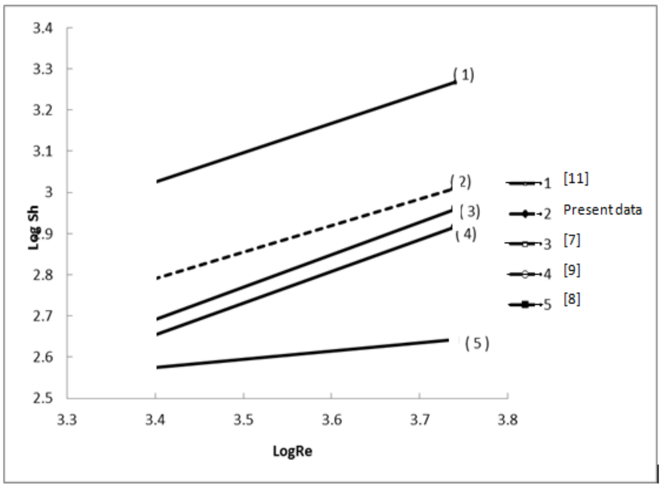

Figure 8: A comparison between the present swirl flow mass transfer data at smooth cylinder with the previous swirl flow studies.

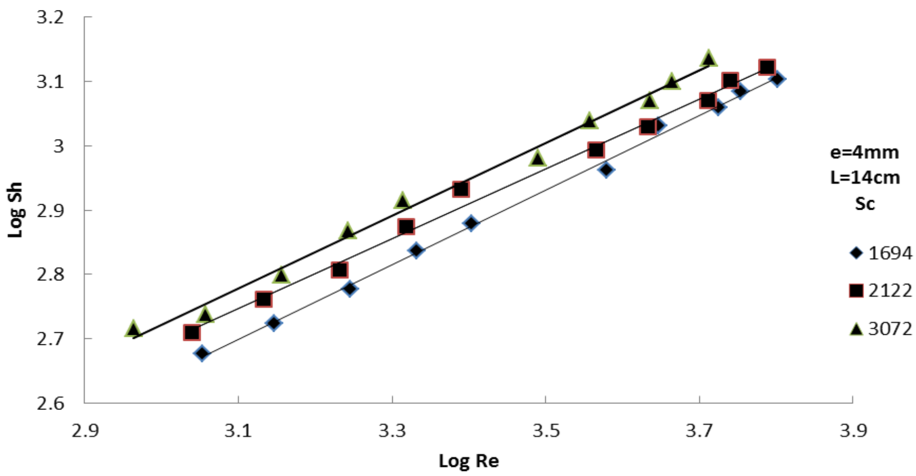

Figure 9: Effect of Re on Sh at rough cylinder (e=4mm) for different Sc.

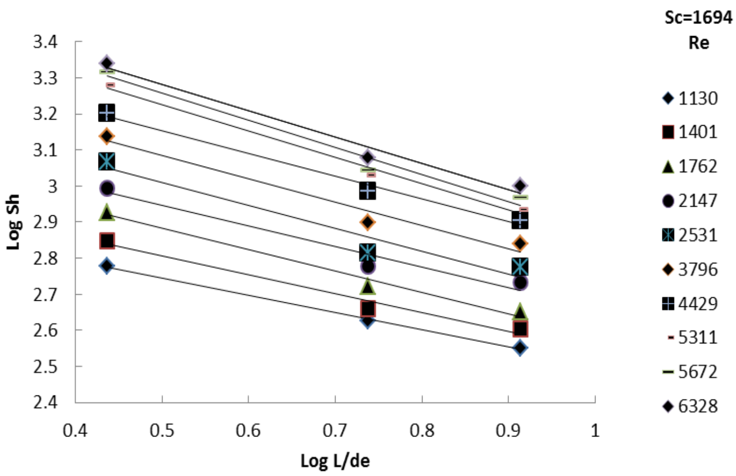

Figure 10: Effect of rough cathode cylinder height L/de on SC at different Re.

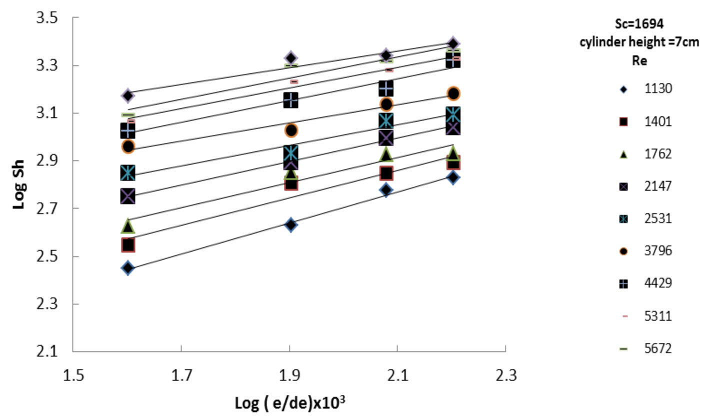

Figure 11: Effect of surface roughness (e/de) on Sh at different Re.

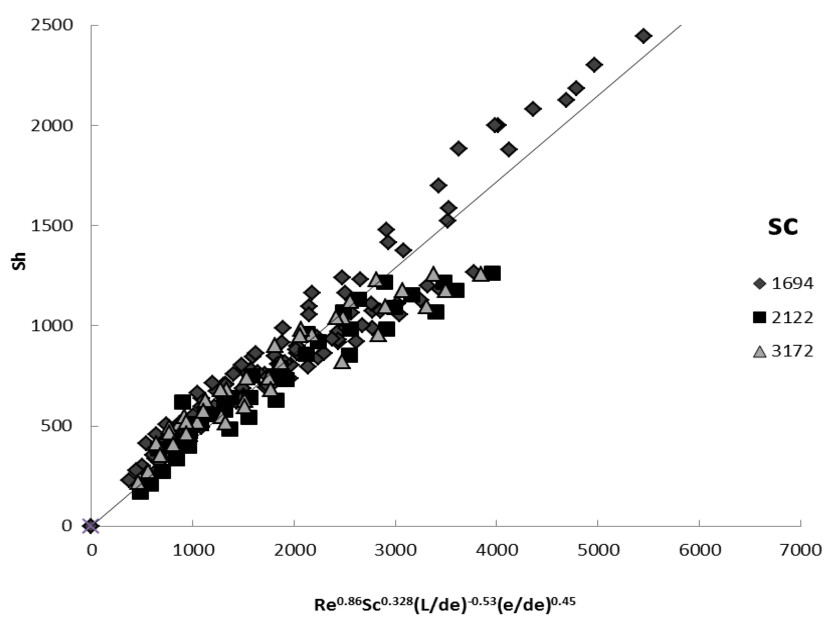

Figure 12: Overall mass transfer correlation at rough cylinders under swirl flow.

Re	L/de=10	Sh_{smooth}/Sh_{axial} L/de=8.2	L/de=5.4	L/de=2.7
1000	1.68	1.66	1.64	1.59
1500	1.61	1.59	1.57	1.53
2000	1.56	1.55	1.52	1.48
2500	1.47	1.46	1.44	1.40
3000	1.52	1.51	1.49	1.45
6000	1.74	1.73	1.70	1.66

Table 2: The enhancement ratio Sh_{smooth}/Sh_{axial} due to swirl flow at a smooth cylinder compared to axial flow [Sc=1500; 2.73<L/de<10].

Re	Sh_{rough}/Sh_{smooth}				
	Smooth	e=1 mm	e=2 mm	e=3 mm	e=4 mm
1000	1	1.02	1.17	1.44	1.68
1500	1	1.06	1.21	1.49	1.73
2000	1	1.08	1.24	1.53	1.77
2500	1	1.10	1.26	1.55	1.80
3000	1	1.12	1.28	1.58	1.83
6000	1	1.18	1.35	1.67	1.94

Table 3: Enhancement ratio Sh_{rough}/Sh_{smooth} at different degrees of roughness (Sc=1500; (L/de)=2.7).

The increase in the rate of mass transfer due to surface roughness may be attributed to:

i) The increased degree of turbulence generated when the tangential flow crosses transversely the vertical roughness elements which protrude outside the laminar sublayer of the hydrodynamic boundary layer, this turbulence penetrate the diffusion layer and enhance the rate of mass transfer [21].

ii) Also surface roughness increases the true surface area over the projected geometrical area with a consequent increase in the rate of mass transfer, the increase in the true area was found to range from 11% to 50% over the projected area. The finding that rough cylinder with peak to valley height of 1 mm behaves almost like smooth cylinder at relatively low Re may be attributed to the fact that a considerable part of these roughness elements (1 mm height) is submerged in the laminar sublayer of the hydrodynamic boundary layer and only a small height (<<1 mm) protrudes outside the laminar sublayer. According to Sclichting [22] for a protrusion in the flowing stream to generate eddies behind it Re should be>100, where R_p is the protrusion Re and is given by $Re_p=\rho ve/\mu$ where e, is the roughness height. The higher the Re the higher the intensity of turbulence and the mass transfer coefficient (Table 3).

Effect of drag reducing polymer on the rate of mass transfer

Drag reducing polymers are used in practice to reduce friction between pipe wall and turbulently moving solution with a consequent decrease in the pumping power required to pump turbulent flow by the ability of polymer molecules to damp the small scale high frequency eddies which prevail in the hydrodynamic boundary layer [13,23]. Despite the beneficial effect of drag reducing polymers on pumping power they may reduce the rate of mass and heat transfer [24,25] taking place at the tube wall in view of damping turbulent eddies which enhance the rate of mass and heat transfer at the tube wall. The aim of this part is to find out the extent to which drag reducing polymers reduces the rate of mass transfer under swirl flow at smooth and rough cylinders.

The Figures 13 and 14 (Tables 4 and 5) show that the presence of polyox drag reducing polymer reduces the rate of mass transfer by a value ranging from 4.9% to 22.86% depending on the operating conditions. The Table 4 shows that the percentage reduction in the rate of mass transfer increases slightly with increasing Re and then decreases with further increase in Re. The initial increase in percentage reduction in k with increasing Re may be attributed to stretching of the polymer molecules under the influence of the high shear stress, stretched polymer molecules are known to be more effective in damping small scale high frequency eddies than coiled polymer molecules [25]. The decline in the percentage reduction in k at high Re may be attributed to mechanical degradation of the polymer molecules under the influence of the high shear stress to ineffective low molecular weight break down products [26].

A comparison between Tables 4 and 5 show that for a given Re rough cylinder produces higher percent reduction in k than smooth cylinder. This is attributed to the ability of rough surfaces to generate a higher proportion of small scale high frequency eddies which are sensitive to the polymer molecules. This result is consistent with the finding of Debrule and Sabersky [24] who studied the effect of drag reducing polymer on the rate of heat transfer in rough tubes, the authors reported higher percent decrease in the rate of heat transfer in rough tubes than in smooth tubes.

Conclusions

The present study has revealed that the combind surface roughness and swirl flow is a powerful tool for enhancing the rate of heat and mass transfer in annular flow reactors. The dimensionless mass transfer correlation obtained in the present work can be used to design and operate annuar electrochemical reactors used to conduct diffusion controlled liquid-solid reactions such as electrochemical removal of heavy metals and organic pollutants from waste water by cathodic deposition and anodic oxidation respectively, and electro organic synthesis. The presence of a builtin heat transfer facility in the annular reactor (outer cooling jacket and the innerside surface of the inner tube) makes it possible to use the annular reactor for conducting exthothermic catalytic diffusion controlled liquid-solid reactions where heat senstive substances are involved such as immoblized enzyme catalyzed biochemical reactions and photochemical reactions. The present results can be used to design and operate more efficient annular membrane dialyzer where a corrugated membrane is used. By virue of the analogy between heat and mass transfer the present results can be used in building more efficient double tube heat exchangers. Drag reducing polymers were found to decrease the rate of mass transfer by a maximum of 22.86%. Accordingly, drag reducing polymers should be used in practice if the benefit of reducing the pumping power outweighs the deleterious effect of reducing the rate of the diffusion controlled process.

List of symbols

A	Geometrical cathode area
$a, \bar{a}, a_1, a_2, a_3, a_4$	Constant
C	Ferricyanide concentration
D	Diffusvity of ferricyanide ion
de	Equivalent diameter of the annulus (do-di)
do	Outer cylinder diameter
di	Inner cylinder diameter
e	Peak to valley height
F	Faraday'sconstant
I	Limiting current
K	Mass transfer coefficient
L	Electrode height
V	Solution velocity
Z	Number of electrons invloved in the reaction
Re	Reynolds number(ρVde/μ)
Sc	Schmidt number(μ/ρD)
Sh	Sherwood number (Kde/D)
μ	Solution viscosity
ρ	Solution denscity
α, β and γ	Constants

Re	% Reduction in K at Different Polymer Concentration		
	100 ppm	200 ppm	300 ppm
1130	11.11	18.52	20.30
1401	10.00	13.33	16.67
1762	7.35	11.76	17.65
2147	5.40	11.01	16.20
2531	11.63	16.28	18.60
3796	7.69	12.30	19.40
4429	11.86	15.25	18.50
5311	6.25	14.06	16.20
5672.	5.97	14.93	15.00
6328	8.33	12.50	14.20

Table 4: Effect of drag reducing polymer on the rate of mass transfer at a rough cylinder under swirl flow [Sc=1694, height=14 cm; rough cylinder (e=4 mm)].

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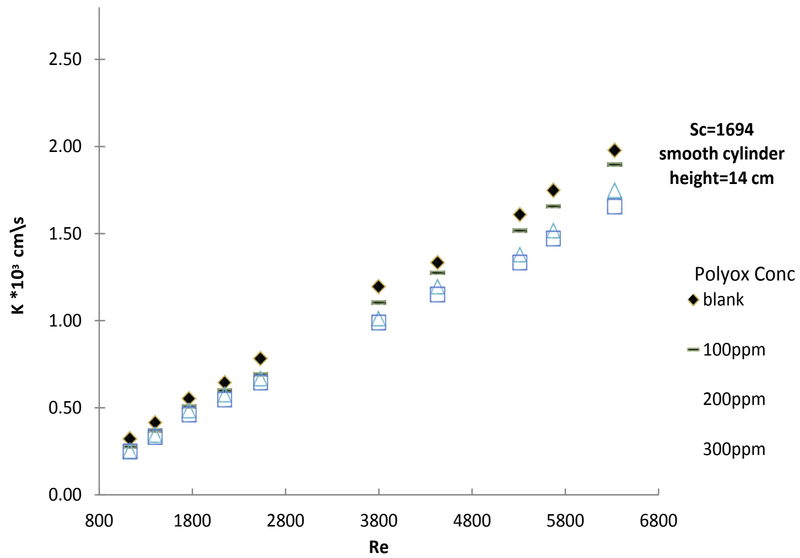

Figure 13: Effect of Re on the mass transfer coefficient at different Polyox concentration (using a smooth cylinder).

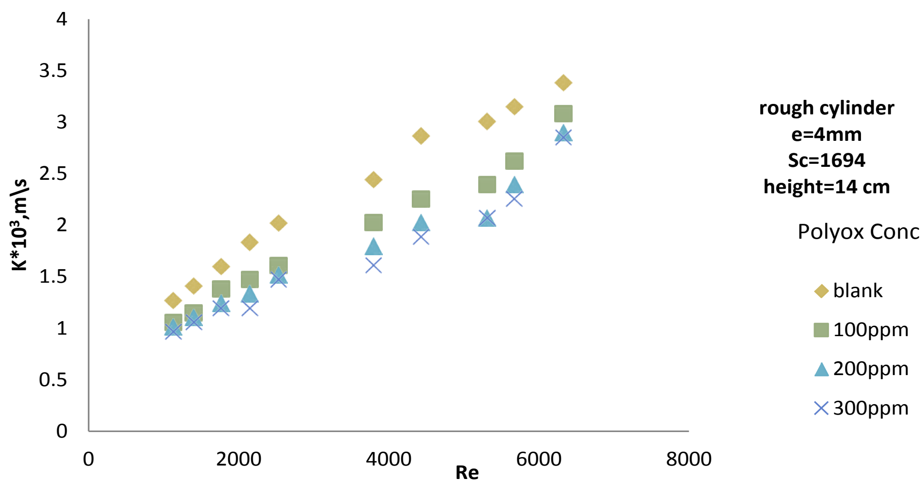

Figure 14: Effect of Re on the mass transfer coefficient at different Polyox concentration using a rough cylinder (e=4mm).

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