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Mass transfer behaviour of a flow-by fixed bed electrochemical reactor composed of a vertical stack of screens under single and upward two phase flow

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Abstract

Rates of mass transfer were studied at a vertical array of closely packed screens under single and two phase (gas–liquid) flow by measuring the limiting current for the cathodic reduction of ferricyanide ions. Variables studied were screen characteristics (mesh number and wire diameter), physical properties of the solution, solution flow rate, gas flow rate and the effect of surface active agents. The single phase data were correlated by the equation:

$$J = 0.52\,Re_{\mathrm{L}}^{-0.55}$$

while the two phase data were correlated by the equations:

$$Sh = 0.87\,Sc^{0.33}Re_{\mathrm{L}}^{0.35}Re_{\mathrm{g}}^{0.12}$$

for the conditions $10 < Re < 125$ and $1.4 < Re_{\mathrm{g}} < 77$; and

$$Sh = 0.62\,Sc^{0.33}Re_{\mathrm{L}}^{0.11}Re_{\mathrm{g}}^{0.25}$$

for the conditions $1.1 < Re_{\mathrm{L}} < 22$ and $1.4 < Re_{\mathrm{g}} < 77$. The presence of surfactant was found to reduce the rate of mass transfer in both single phase and two phase flow, the percentage reduction being higher in the case of single phase flow.

List of symbols

A	area of the screen array
a	specific area of the screen
C	ferricyanide concentration
D	diffusivity
d_{w}	wire diameter
F	faradaic number
I	limiting current
k	mass transfer coefficient
N	screen mesh number
V	superficial solution velocity
V_{g}	superficial gas velocity
Z	number of electrons involved in the reaction
J	mass transfer J factor ($St \times Sc^{0.66}$)
Re	solution Reynolds number ($\rho V d_{\mathrm{w}}/\mu$)
Re_{g}	gas Reynolds number ($\rho V_{\mathrm{g}} d_{\mathrm{w}}/\mu$)
Sc	Schmidt number (ν/D)
St	Stanton number (k/V)
Sh, Sh_0	Sherwood number for two phase flow and single phase flow, respectively (kd_{w}/D)

Greek symbols

ν	kinematic viscosity
μ	solution viscosity
ρ	solution density

1. Introduction

The use of screens and expanded metals in building catalytic and three-dimensional electrodes offers many advantages over other packing materials such as high specific area, high turbulence promoting ability, high porosity and relatively low pressure drop, ease of coating with catalyst and ready availability at modest cost. Mass transfer at horizontal screen catalyst and porous flow through electrodes composed of horizontal arrays of woven metallic screens has been the subject of several studies [1–9]. The aim is high space time yield catalytic and electrochemical reactors suitable for processing dilute solutions such as those encountered in electroorganic synthesis and waste water treatment. Unfortunately the flow through porous electrode has met with a limited success on the commercial scale in view of the nonuniformity of current and potential distribution, poor selectivity and low conversion per pass [10]. To avoid these shortcomings attention has been directed to the flow-by electrode [11–15] which has other advantages such as the possibility of using it in the form of a divided or undivided filter press type cell.

The aim of the present work is to study the mass transfer behaviour of a vertical array of closely packed screens under single and two phase flow using a wide range of screen characteristics. In previous work Storck et al. [11] studied the mass transfer behaviour of a vertical stack of screens under single phase flow using fine screens of mesh number 60. No previous studies have been done on the mass transfer behaviour of vertical stacks of screens in two phase flow. Coppola et al. [9] studied the mass transfer behaviour of a flow through electrode composed of a horizontal stack of screens. The study of two phase flow mass transfer behaviour of a vertical screen array would assist in the design and operation of flow-by electrodes used to conduct diffusion controlled or partially diffusion controlled gas–liquid–solid electrochemical reactions such as H_2O_2 synthesis [16] and electrochemical flue gas desulphurization, where SO_2 is absorbed and anodically oxidized to H_2SO_4 [17]. Also two phase flow with an inert gas can be used with advantage to increase the percentage conversion per pass of flow-by electrodes by operating them under low solution flow rate and high superficial gas velocity, where the residence time and the rate of mass transfer are high. Since the present reactor could be used for processing solution containing surfactants, for example, removal of heavy metals from electroplating waste solutions and electroorganic synthesis from solutions containing quaternary ammonium salts [18]. It is also relevant to study the effect of surface active agents on the rate of mass transfer.

2. Experimental technique

The apparatus (Figure 1) consisted of a recirculating fixed bed flow-by reactor and electrical circuit. The reactor consisted of a plexiglass rectangular duct of 8 cm × 8 cm cross section and 60 cm height divided into three sections: inlet, working and outlet sections. The inlet section which was packed with glass spheres of 1.6 cm diameter had a height of 25 cm. The bottom of the inlet section was a perforated Plexiglass plate distributor containing 1800 holes, of 1 mm diameter. The working section consisted of a vertical array of seven closely packed screen cathodes placed between two arrays of closely packed vertical anode screens. The number of screens per anode ranged from 18 to 65 depending on the mesh number of the screen. A vertical plastic screen was inserted between the cathode and each of the two anodes to prevent direct contact between the electrodes. The screens forming the cathode were nickel plated stainless screens while those forming the two anodes were of stainless steel. The cathode and anode screens had the same geometric characteristics (mesh number and wire diameter). The cathode and the two anodes filled the whole cross section of the duct. The high anode area compared to the cathode area allowed the use of the anode as a reference electrode. The cathode and the two anodes were fed with electrical current through vertical insulated nickel plated copper wires. The outlet section was 25 cm in height and was packed with 1.6 cm diameter glass spheres.

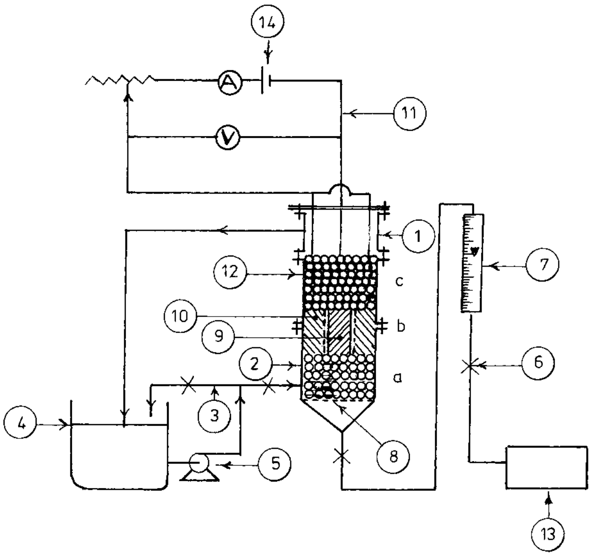

Fig. 1. Apparatus arrangement: (1) rectangular duct (*a* inlet section, *b* working section, *c* outlet section); (2) solution inlet; (3) solution bypass; (4) storage tank; (5) plastic centrigugal pump; (6) control valves; (7) rotameter; (8) plastic gas distributor; (9) cathode; (10) anode; (11) electrical circuit; (12) plastic screen; (13) air compressor; (14) 12 V d.c power supply.

The electrical circuit consisted of a 12 V d.c power supply, a multirange ammeter connected in series with the cell and a high impedance voltmeter connected in parallel with the cell to measure its voltage.

Solution entered the reactor through a 25 mm (1 inch) diameter inlet tube placed above the perforated bottom. A similar outlet tube was placed at the top of the reactor. Solution flow rate was controlled by a bypass and was measured by means of timed collection. In case of the two phase flow, air entered through the perforated plastic distributor. Rates of mass transfer were determined by measuring the limiting current for the cathodic reduction of potassium ferricyanide using a solution containing equimolar amounts of potassium ferricyanide and potassium ferrocyanide and a large excess of sodium hydroxide as a supporting electrolyte. Three NaOH concentrations were used 1, 2 and 4 M.

All chemicals were prepared using AR grade chemicals and distilled water. The concentration of ferricyanide and ferrocyanide were checked by iodometry and permanganate titration, respectively [21]. Before and during measurement of the limiting current under single phase flow, N_2 gas was bubbled in the storage tank to remove dissolved oxygen. Current–voltage experiments were carried out twice at 25 °C. The mass transfer coefficient was calculated from the limiting current using the equation:

$$K = \frac{I}{ZFAC} \tag{1}$$

Screen area A was used to calculate the mass transfer coefficient was calculated in terms of the wire diameter (d_w) and mesh number (N) using the method of Armour and Cannon [22] as follows.

The screen surface area per unit volume of the screen (a) in cm^2/cm^3 is given by

$$a = \pi L N^2 \tag{2}$$

where

$$L = \left[\frac{1}{N^2} + d_w^2\right]^{0.5} \tag{3}$$

In calculating the volume of the screen, the screen thickness was taken as twice the wire diameter. Table 1 shows characteristic of the screen used in the present study.

To test the effect of surfactants on the rate of mass transfer laboratory grade Triton X–100, a nonionic surfactant (octyle phenoxy polyethoxy ethanol) having the formula $C_{34}H_{62}O_{11}$ (MW = 646) was used. Physical properties of the solutions (ρ, μ, and D) used in data correlation (Table 2) were taken from the literature [21, 22].

3. Results and discussion

3.1. *Single phase flow*

Figure 2 shows that for single phase flow Sh increases with increasing Re_L to the exponent 0.45 which agrees fairly well with the value 0.493 obtained, by Storck et al. [14] who studied the mass transfer behaviour of a vertical stack of screens under single phase flow. Figure 3 shows that the single phase data for the conditions $1250 < Sc < 8270$; $0.6 < Re_L < 124$; $0.28 < d_w < 1.2$ mm fit the equation:

Table 1. Screen parameters

Mesh number wire/inch	6	10	14	20	30
Wire diameter/cm	0.12	0.071	0.05	0.04	0.028
Distance between wires/cm	0.303	0.183	0.131	0.087	0.0567
Specific area/cm^{-1}	7.5	12.5	17.5	25.3	37.6
Screen porosity	0.775	0.778	0.782	0.747	0.737

Table 2. Physical properties of the solutions at 25 °C

Solution composition	ρ /g cm^{-3}	$\mu \times 10^2$ /poise	$D \times 10^6$ /cm^2 s^{-1}	Sc
0.01 M $K_3Fe(CN)_6$ + 0.01 M $K_4Fe(CN)_6$ + 1 M NaOH	1.0461	1.1042	6.693	1577
0.01 M $K_3Fe(CN)_6$ + 0.01 M $K_4Fe(CN)_6$ + 2 M NaOH	1.085	1.3822	5.508	2314
0.01 M $K_3Fe(CN)_6$ + 0.01 M $K_4Fe(CN)_6$ + 4 M NaOH	1.1675	2.2743	3.291	5919

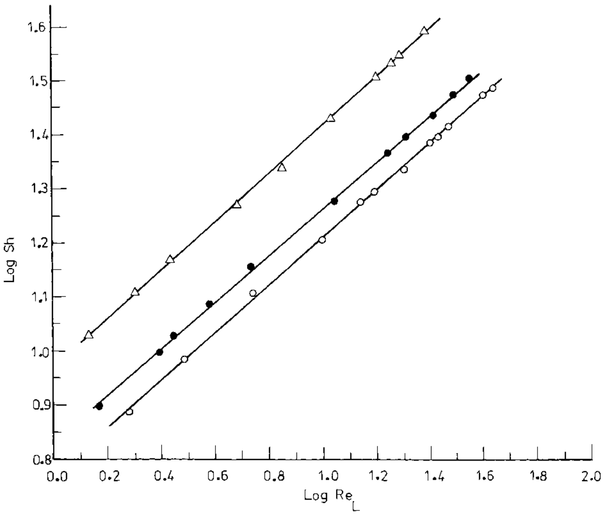

Fig. 2. Log Sh vs log Re_L for single phase flow. Key for Sc: (○) 1720; (●) 2630; (△) 6180.

$$J = 0.52\, Re_L^{-0.55} \tag{4}$$

with an average deviation of ±6%. Wire diameter (d_w) was used as the characteristic length in calculating Re_L.

Figure 4 shows a comparison between the present data and the data of other authors who used horizontal [1–5] and vertical screen arrays [11]. The present data lie below the horizontal screen data and above the data of Storck et al. The higher rates of mass transfer at horizontal screens may be attributed to the local increase in solution velocity (jetting effect) as the solution passes through the openings of the horizontal screens. This effect is absent in the case of vertical screens which are parallel to the flow. The finding that the present data are higher than those of Storck et al. may be explained by the fact that Storck et al. used screens of high mesh number, namely 60 compared to that in the present work which ranged from 6 to 30 wire/in. (i.e., per 25 mm). According to Zaki et al. [23] who studied the effect of mesh number on the turbulence promoting ability of screens there is an optimum mesh number which gives the highest mass transfer enhancement ratio. Zaki et al. studied the effect of mesh number 10, 14, 20, 30 and 40 wire/in. on the rate of mass transfer at a vertical cylinder coated with the screen. The authors found that mesh number 20 gave the highest enhancement ratio followed by 10 and 14, respectively, while screens of high mesh number such as 30 and 40 gave the least enhancement ratio. The presence of an optimum distance between turbulence promoting elements which produces maximum enhancement was also revealed by Fischl et al. [24], who studied the effect of attached

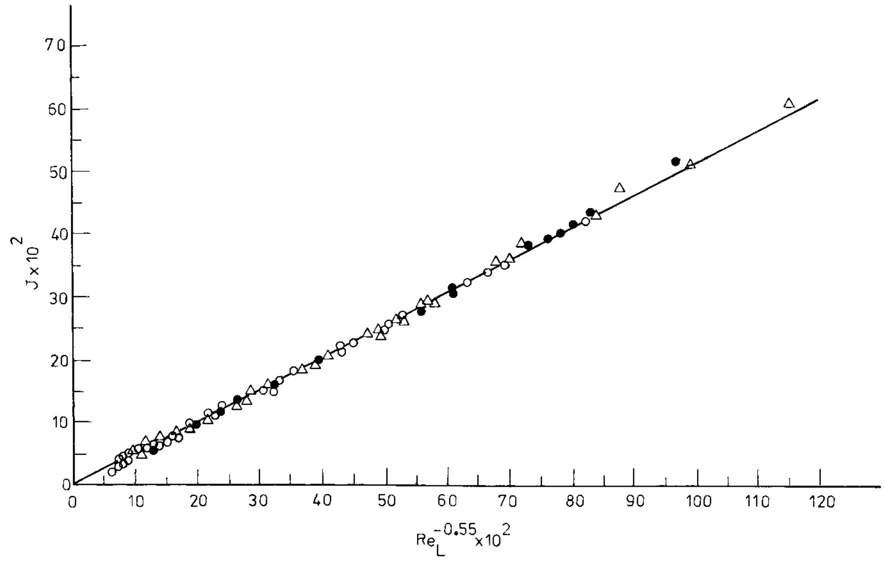

Fig. 3. Overall mass transfer correlation for single phase flow. Key for *Sc*: (○) 1720; (●) 2630; (△) 6180.

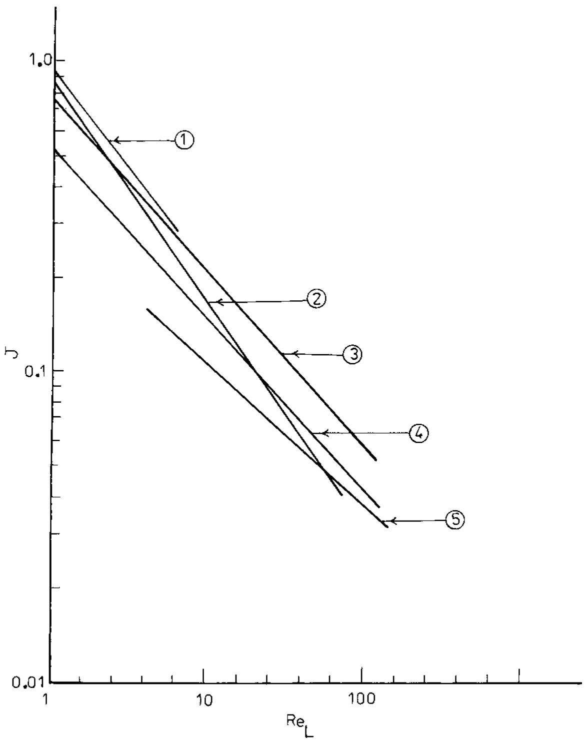

Fig. 4. Comparison of the present single phase mass transfer data with previous data at horizontal and vertical screen arrays. Key: (1) horizontal array, Sioda [2]; (2) horizontal array, Gay and Maugham [3]; (3) horizontal array, Shah and Roberts [5]. (4) vertical array, present work; (5) vertical array, Storck et al. [11].

turbulence promotors normal to the flow on the rate of mass transfer at a vertical plate.

3.2. *Two phase flow*

Figure 5 shows the effect of *Re* on *Sh* at different superficial air velocities: two regions can be distinguished. At low solution velocity ($V < 2.94$ cm s^{-1}) visual observation revealed churn-turbulent flow, under these conditions the rate of mass transfer is determined mainly by air superficial velocity. With increasing solution velocity ($V > 3.8$ cm s^{-1}) the flow becomes almost bubbly. Under these conditions the role of the liquid velocity becomes dominant while the role of gas velocity becomes modest. This behaviour is consistent with the results obtained by different authors who studied the effect of two phase flow on the rate of mass transfer in fixed beds packed with spheres and cylinders [25–27].

The high rate of mass transfer at low solution flow rates is caused by the ability of the rising bubbles to generate turbulence in their wakes and induce radial momentum [28], as well as collision with the mass transfer surface. By increasing Re_L the rise velocity of the bubbles increases, with a consequent decrease in the gas holdup, that is, the concentration of the turbulence promoting bubbles decreases with a consequent decrease in their influence on the rate of mass transfer. Mainstream turbulence and turbulence induced by the screen wires become responsible for enhancing the rate of mass transfer. However, Figure 5 shows that even at high solution flow rates the higher the superficial gas velocity the higher the mass transfer coefficient. This may be attributed to the decrease in the cross-sectional area available for solution flow and the subsequent increase in the interstitial solution velocity as a result of the increase in gas holdup with increasing gas velocity.

To assess the extent to which two phase flow increases the rate of mass transfer over the single phase value, the enhancement ratio Sh/Sh_0 was plotted against Re_L at different gas superficial velocities as shown in Figure 6. The enhancement ratio ranges from 1.05 to 3.1 depending on the gas and liquid flow rates. The enhancement ratio increases with increasing superficial gas velocity and decreases with increasing solution flow rate. The two phase mass transfer data were correlated by two overall mass transfer correlations depending on the range of operating conditions.

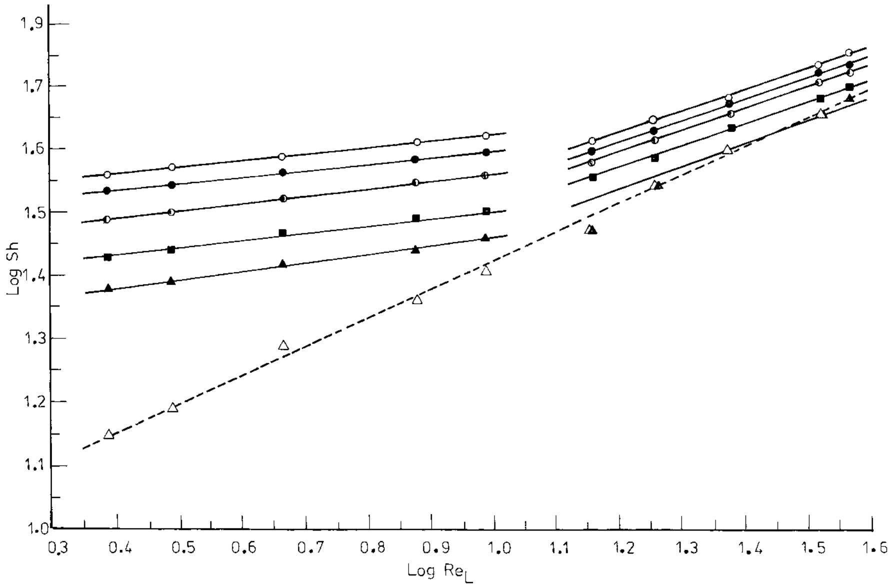

Fig. 5. Log *Sh* vs log Re_L at different superficial gas velocities. Mesh number = 10; Sc = 5630. V_g/cm s^{-1}: (○) 6.64; (●) 5.1; (◐) 3.56; (■) 2.1; (▲) 1.4; (- - - -, △) 0 (single phase flow).

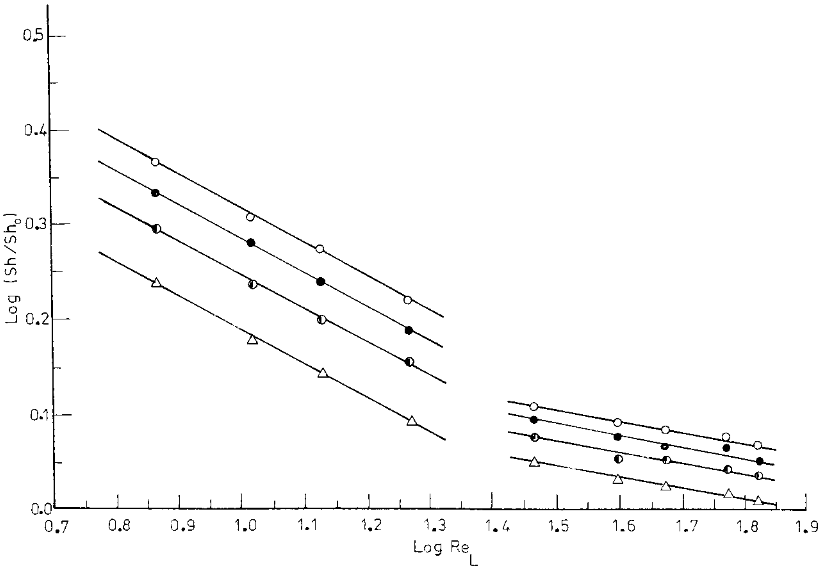

Fig. 6. Log (Sh/Sh_0) vs log Re_L at different superfical velocities. Mesh number = 10; Sc = 1455. V_g/cm s^{-1}: (○) 6.64; (●) 5.1; (◐) 3.56; (△) 2.1.

Figure 7 shows that for the conditions $1250 < Sc < 8270$; $10 < Re_L < 125$; $1.4 < Re_g < 77$ (bubbly flow) the data fit the equation

$$Sh = 0.87\,Sc^{0.33}Re_L^{0.35}Re_g^{0.12} \tag{5}$$

with an average deviation of ±5%.

Figure 8 shows that for the conditions $1250 < Sc < 8270$; $1.1 < Re_L < 22$; $1.4 < Re_g < 77$ (churn-turbulent) the data fit the equation

$$Sh = 0.625\,Sc^{0.33}Re_L^{0.11}Re_g^{0.25} \tag{6}$$

with an average deviation of ±3%.

The Re_g exponent 0.25 of Equation 6 agrees with the value obtained by different authors who studied the effect of gas sparging on liquid–solid mass and heat transfer, the exponent also agrees with the prediction of the surface renewal model [28, 29] which is based on the ability of the rising bubbles to induce radial turbulent momentum to the mass transfer surface.

Figure 9 shows a comparison between the present data and the two phase data obtained by Coppola et al. [9] who used a horizontal array of screens; Figure 9 shows that for a given set of conditions the horizontal array produces higher rates of mass transfer. As mentioned earlier the higher rate of mass transfer at the horizontal orientation may be ascribed to the local increase in the solution velocity (jetting effect) when the solution passes through the screen openings.

In the light of the present comparison between the mass transfer data at the vertical and the horizontal

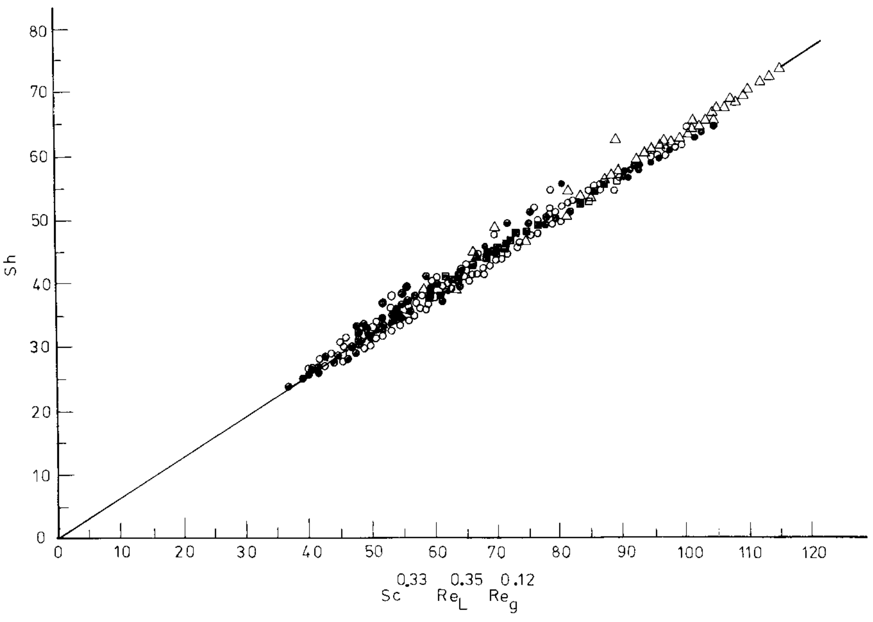

Fig. 7. Overall mass transfer correlation for two phase flow (churn-turbulent). Key for *Sc*: (○) 1610; (●) 2370; (△) 6130.

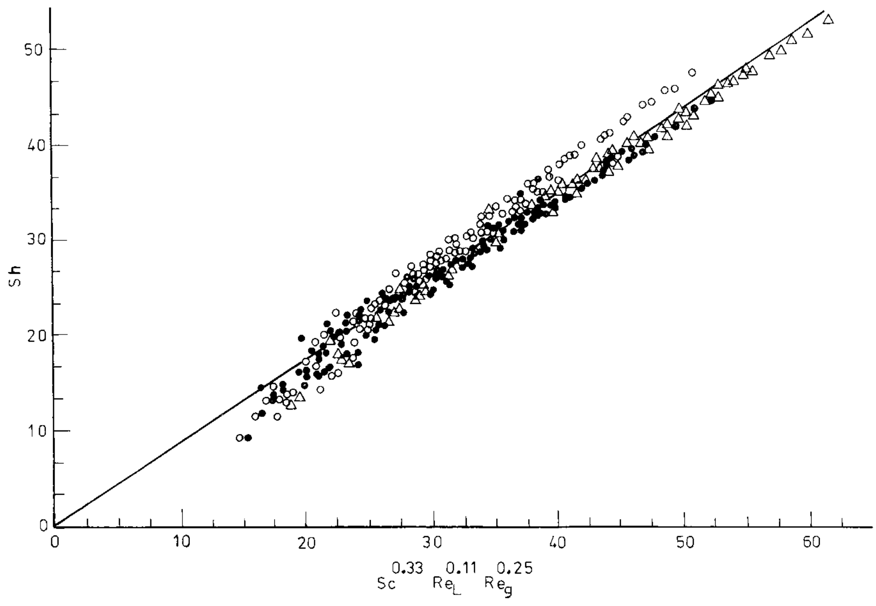

Fig. 8. Overall mass transfer correlation for two phase flow (bubbly regime). Key for *Sc*: (○) 1610; (●) 2370; (△) 6130.

screen array the choice between the horizontal and the vertical orientation in construction of an industrial reactor depends on whether the reactor is catalytic or an electrochemical reactor. For a catalytic chemical reactor intended for conducting diffusion controlled reactions, the horizontal orientation is preferable but for electrochemical reactors the vertical orientation should be used to ensure uniform current and potential distribution.

3.3. *Effect of surfactants on the rate of mass transfer*

Figure 10 shows the effect of Triton surfactant concentration on the single phase mass transfer coefficient, the mass transfer coefficient decreases with increasing Triton concentration up to 0.3% and then remains almost constant with further increase in concentration. The decrease in the rate of mass transfer under single phase flow ranges from 8.2% to 39.7% depending on Triton concentration and solution velocity. The decrease in the rate of mass transfer may be ascribed to the increase in the interfacial viscosity as a result of adsorption of the surfactant molecules on the electrode surface [30]; the increase in interfacial viscosity gives rise to a corresponding decrease in the effective diffusivity of the reacting ion and makes the diffusion layer more resistant to thinning by the shear forces. The fact that Triton ceases to affect the rate of mass transfer beyond a concentration of 0.3% may be attributed to the

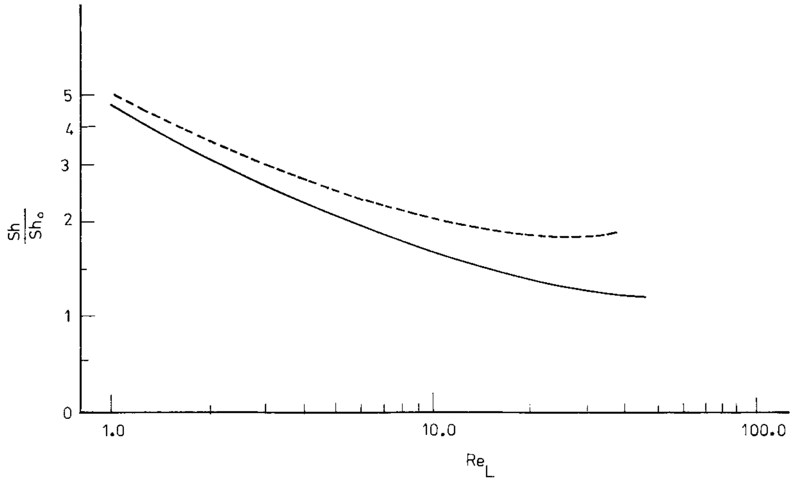

Fig. 9. Comparison between the present two-phase data and the data of Coppola et al. [9] for a horizontal array. $Re_g = 60$. Key: (- - - - -) horizontal array, Coppola et al. [9]; (———) vertical array, present data.

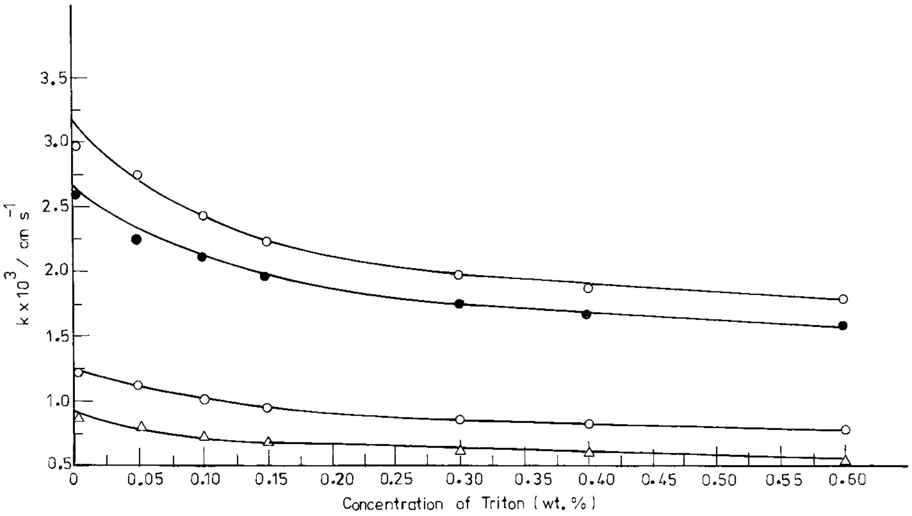

Fig. 10. Effect of Triton concentration on the single phase mass transfer coefficient. Mesh number: 6; $Sc = 1660$. $V/\mathrm{cm\ s^{-1}}$: (○) 11.9; (●) 7.5; (◐) 1.63; (△) 0.72.

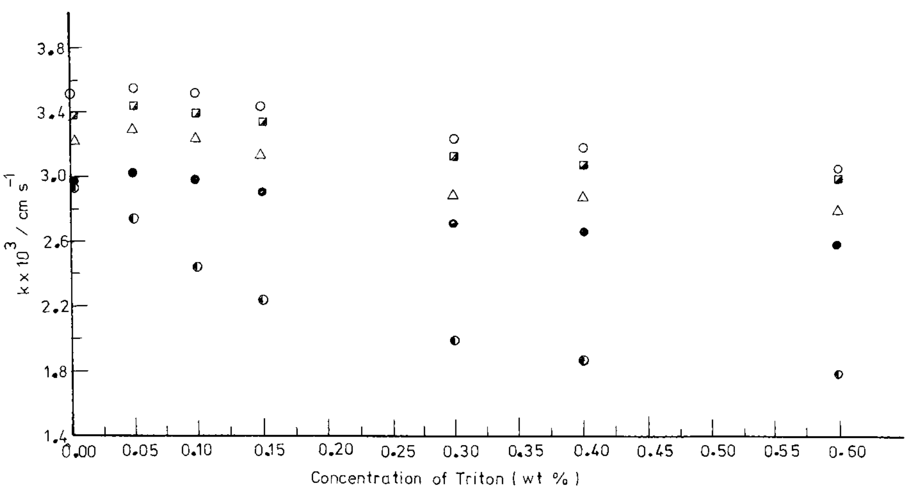

Fig. 11. Effect of Triton concentration on the two phase mass transfer coefficient. Mesh number: 6; $Sc = 1660$; $V = 11.9\ \mathrm{cm\ s^{-1}}$. $V_g/\mathrm{cm\ s^{-1}}$: (○) 5.87; (◪) 4.33; (△) 2.84; (●) 1.4; (◐) 0 (single phase).

attainment of the critical micellar concentration where surfactant molecules no longer adsorp on the electrode surface but associate together to form spherical micelles in the solution [31].

Figure 11 shows the effect of Triton on the two phase mass transfer coefficient. The mass transfer coefficient decreases by an amount ranging from 10 to 29% depending on the superficial liquid and gas velocities. The less negative effect of Triton on the rate of mass transfer in the case of two phase flow may be explained by the fact that the presence of surfactants leads to the production of small-sized noncoalescant bubbles of low rise velocity. As a consequence the gas holdup increases [32] with a subsequent increase in the interstitial solution velocity and the rate of mass transfer which alleviates the decrease in the rate of mass transfer caused by the interfacial increase in viscosity. The finding that surfactants have an enhancing effect on two phase mass transfer agrees with the finding of Parakash et al. [33] who found that the presence of surfactant increases the rate of mass transfer between suspended solid particles and liquid in three phase (solid–liquid–gas) fluidized beds by 100%. In earlier studies [34, 35] surfactants were found to decrease the rate of mass transfer at gas evolving electrodes and gas sparged electrodes by an amount ranging from 7.6% to 81% and 5% to 30%, respectively. The agreement between the effect of surfactants on the rate of mass transfer at gas sparged electrodes and the present two phase results is remarkable.

4. Conclusions

(i) Single and two phase mass transfer correlations were obtained for vertical stacks of closely packed screens using a wide range of geometric screen parameters.

(ii) The rate of two-phase mass transfer is determined at low solution velocities by the superficial gas velocity while at high solution velocities the rate of mass transfer is controlled mainly by the solution velocity.

(iii) The presence of surfactants in the solution reduces the rate of single phase and two phase mass transfer by a maximum of 40% and 29%, respectively.

(iv) For a given set of conditions, the rate of two phase mass transfer at a horizontal screen stack is higher than that at the vertical stack.

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