

# Mass transfer characteristics of gas-sparged fixed-bed electrodes composed of stacks of vertical screens

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Received 12 March 1985; revised 1 June 1985

Mass transfer rates at a gas-sparged fixed-bed electrode made of stacks of vertical screens were studied by measuring the limiting current for the cathodic reduction of potassium ferricyanide. Variables studied were air flow rate, physical properties of the solution and bed thickness. The mass transfer coefficient was found to increase with increasing air flow rate up to a certain point and then remain almost constant with further increase in air flow rate. Increasing bed thickness was found to decrease the mass transfer coefficient. Mass transfer data were correlated by the equation

$$J = 0.2(ReFr)^{-0.28}(L/d)^{-0.28}$$

For a single vertical screen electrode the data were correlated by the equation

$$J = 0.187(ReFr)^{-0.26}$$

## Nomenclature

$a$  constant  
 $C$  concentration of ferricyanide ion (mol cm<sup>-3</sup>)  
 $d$  screen wire diameter (cm)  
 $D$  diffusivity of ferricyanide ion (cm<sup>2</sup>s<sup>-1</sup>)  
 $F$  Faraday's constant  
 $Fr$  Froude number ( $V^2/dg$ )  
 $g$  acceleration due to gravity (cm<sup>2</sup>s<sup>-1</sup>)  
 $I_L$  limiting current density (A cm<sup>-2</sup>)  
 $J$  mass transfer  $J$  factor ( $StSc^{0.66}$ )

$K$  mass transfer coefficient (cm s<sup>-1</sup>)  
 $L$  bed thickness (cm)  
 $Re$  Reynolds number ( $\rho Vd/\mu$ )  
 $Sc$  Schmidt number ( $\nu/D$ )  
 $St$  Stanton number ( $K/V$ )  
 $V$  superficial air velocity (cm s<sup>-1</sup>)  
 $Z$  number of electrons involved in the reaction  
 $\rho$  density of solution (g cm<sup>-3</sup>)  
 $\nu$  kinematic viscosity (cm<sup>2</sup>s<sup>-1</sup>)  
 $\mu$  solution viscosity (g cm<sup>-1</sup>s<sup>-1</sup>)

## 1. Introduction

The study of forced convection mass transfer behaviour of stacked screen systems has recently received considerable interest in view of their possible use in building electrochemical reactors of high specific area and high mass transfer rate [1-6]. These studies have been conducted using single-phase flow. The object of the present work is to test the possibility of using gas sparging (with no electrolyte flow) as a means of enhancing the rate of mass transfer at a fixed bed of closely packed vertical screens. Ibl *et al.* [7] and

Ettel *et al.* [8] who studied the effect of gas sparging on the rate of mass transfer in a parallel plate reactor found that gas sparging was an effective and economic way of stirring. The high efficiency of gas sparging in increasing the rate of mass transfer compared to single phase flow was attributed by Kast [9] to the ability of gas bubbles to induce radial momentum transfer. Some studies have been carried out on mass transfer at fixed beds of spheres using two-phase flow, leading to the conclusion that at relatively low liquid flow rates gas stirring predominates in the enhancement of the rate of mass transfer

while at high liquid flow rates the role of gas sparging becomes insignificant. The implication of this conclusion is that gas sparging combined with low solution flow rates can be used in the operation of continuous electrochemical reactors in order to achieve a high degree of conversion per pass and a high mass transfer coefficient. In the present work electrodes were oriented in such a way that electric current flow was perpendicular to gas flow. The advantage of this configuration over the configuration where current flow and gas flow are parallel is that current distribution is more uniform [4].

## 2. Experimental technique

Fig. 1 shows the cell and electrical circuit used in the present work. The cell consists of a cylindrical plastic container of 10 cm diameter and 30 cm height fitted with a 3G sintered glass distributor at its bottom. The anode was made of a cylindrical stainless steel screen of 9.5 cm diameter and 30 cm height. The cathode was made of a number of rectangular nickel-plated copper screens measuring  $4 \times 5$  cm, each screen with a mesh number of 28 per  $\text{cm}^2$ , a wire diameter of 0.0623 cm, a porosity of 0.75 and a specific area of  $25 \text{ cm}^2 \text{ cm}^{-3}$ . The screens were held tightly together using a 1.5 mm diameter nickel-plated copper wire which penetrated the screen bed at its centre. This wire which acted as a holder and current feeder was isolated with Epoxy resin except at the contact with the bed. The number

of screens forming the cathode ranged from one to seven.

The bed cathode was positioned at the centre of the cell at 5 cm from its base. The circuit consisted of a multirange ammeter and a 6 V d.c. power supply connected in series with the cell. A high impedance voltmeter was connected in parallel with the cell to measure its voltage. Polarization data were obtained by increasing the current stepwise and measuring the steady state cathode potential against a reference electrode by means of a potentiometer. The reference electrode consisted of a nickel wire immersed in the cup of a Luggin tube filled with the same cell solution; the tip of the Luggin tube was positioned 0.5–1 mm from the cathode surface. Solutions used were made of equimolar amounts of  $\text{K}_3\text{Fe}(\text{CN})_6$  and  $\text{K}_4\text{Fe}(\text{CN})_6$  dissolved in 2 N NaOH. Three concentrations of  $\text{K}_3\text{Fe}(\text{CN})_6$  and  $\text{K}_4\text{Fe}(\text{CN})_6$  were used, namely 0.05, 0.1 and 0.2 M. All chemicals were Analar grade and distilled water was used in preparing the solutions. Before each run the cathode was activated in a manner similar to that used by Eisenberg *et al.* [10]. Air was introduced to the cell at the required rate by means of a variable speed air compressor. The air superficial velocity which ranged from 0.02 to  $0.637 \text{ cm s}^{-1}$  was measured by means of a calibrated rotameter. Each experiment was carried out twice at  $25 \pm 1^\circ \text{C}$  using a fresh solution. The diffusivity of ferricyanide ion in the different solutions was calculated from the formula [11]

$$D\mu/T = 2.5 \times 10^{-10} \text{ cm g s}^{-2} \text{ K}^{-1} \quad (1)$$

## 3. Results and discussion

Fig. 2 shows typical polarization curves with well defined limiting current plateaux obtained at different superficial air velocities using a bed composed of three screens. Such polarization curves could not be obtained for thicker beds with a number of screens higher than three at superficial air velocities higher than  $0.17 \text{ cm s}^{-1}$ . The limiting current was determined from the polarization curves and used to calculate the mass transfer coefficient according to the equation

$$I_L/ZF = KC \quad (2)$$

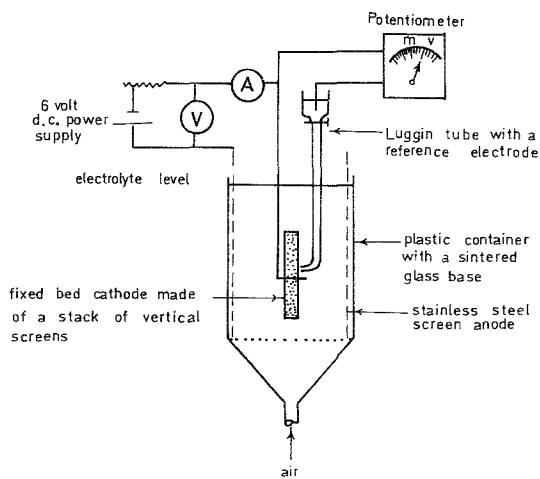


Fig. 1. Cell and electrical circuit.

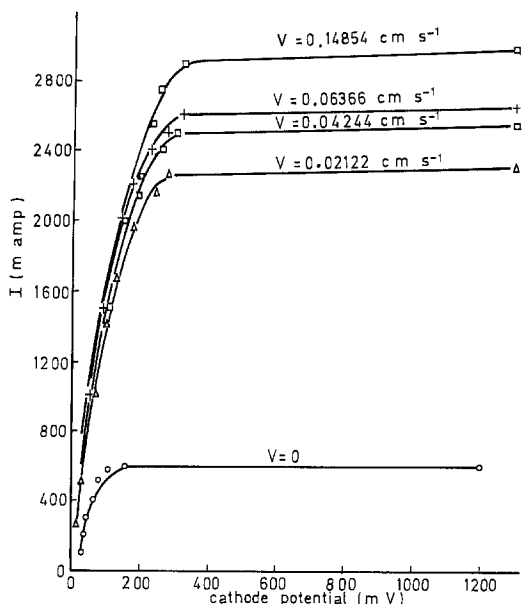


Fig. 2. Polarization curves at a screen stack cathode containing three screens at different air superficial velocities. ( $C = 0.2 \text{ M}$ .)

Fig. 3 shows the relation between the mass transfer coefficient and the superficial air velocity. The mass transfer coefficient increases with the superficial air velocity up to a certain point according to the equation

$$K = aV^n \quad (3)$$

and then remains almost constant with further increase in superficial velocity. The exponent  $n$  in Equation 3 was found to depend on the bed thickness. Table 1 shows the value of  $n$  obtained for different numbers of screens per bed. The

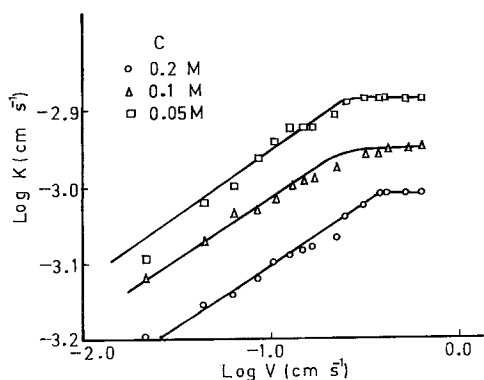


Fig. 3. Effect of superficial air velocity on the mass transfer coefficient at a vertical stack containing three screens.

Table 1. Value of  $n$  obtained for different numbers of screens per bed

Number of screens per bed	Slope of $\log K$ versus $\log V$ ( $n$ )
1	0.200
2	0.180
3	0.159
4	0.150
5	0.133
6	0.121
7	0.080

$\log K$  versus  $\log V$  slope for a single screen (0.2) agrees reasonably well with the values obtained by Louisi (0.19) [12], Steiff & Weinspach (0.22) [13] Lewis *et al.* (0.172) [14] and Fair *et al.* (0.22) [15] who studied heat transfer in bubble columns. The present slope is at variance with the values obtained from the data of Ibl *et al.* (0.36) [7] who studied the effect of gas sparging on the rate of mass transfer in a parallel plate electrochemical reactor. The finding that at relatively high air flow rates the mass transfer coefficient shows little dependence on superficial air velocity is in agreement with results for heat transfer in bubble columns [16, 17]; however, the plateau region was reached earlier in the present work. A possible explanation for the small dependency of the mass transfer coefficient on the superficial air velocity at high velocities may be given in terms of the flow pattern of the displaced solution (Fig. 4). Solution displaced by the air bubbles moves horizontally when it reaches the surface of the solution then moves downward at the container wall to a depth depending on air superficial velocity — the higher the air velocity the deeper the penetration of the displaced solution. The downward moving stream generates large eddies in the bulk of the solution. At high air velocities these eddies prevail in the whole solution and interact with the upward moving gas bubbles. This leads to a complex flow pattern dominated by the large eddies which are less effective in enhancing the rate of mass transfer than the small scale high intensity eddies generated by the movement of air bubbles in the solution [18].

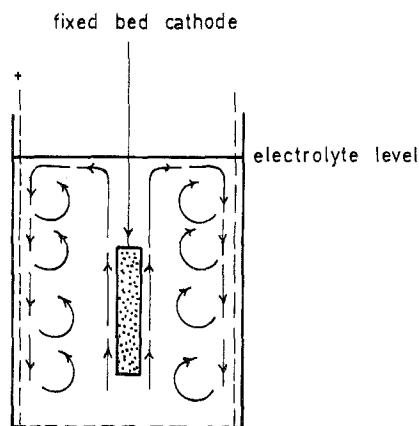


Fig. 4. Approximate flow pattern of the displaced solution.

The finding that the number of screens per bed had a significant effect on the  $\log K$  versus  $\log V$  slope is at variance with the finding of authors who used single phase flow through beds of screens where bed thickness has a negligible effect on the  $\log K$  versus  $\log V$  slope. The discrepancy seems to be caused by the difference in flow conditions. Based on previous studies in heat [9] and momentum transfer [19] in bubble columns it seems that in the present case the radial momentum generated by the gas bubbles in the interelectrode gap contributes a great deal to enhancing the rate of mass transfer along with the axial momentum induced by bubbles which pass along the bed. As the electrode thickness increases by increasing the number of screens forming the bed, the radial momentum induced by air bubbles receives more resistance as it penetrates the electrode. The data shown in Fig. 5 are consistent with the results of Table 1 and show that for a given superficial air velocity the mass transfer coefficient decreases with increasing number of screens per bed. The decrease in the mass transfer coefficient with increase of the number of screens per bed at a given superficial air velocity was also observed in the case of horizontally oriented air stirred fixed beds of screens [20]. An overall mass transfer correlation was envisaged using the dimensionless groups  $J$ ,  $Fr$  and  $Re$  usually used in correlating heat and mass transfer in gas stirred vessels. In view of the present finding that bed thickness affects the mass transfer coefficient significantly, an extra dimensionless term was

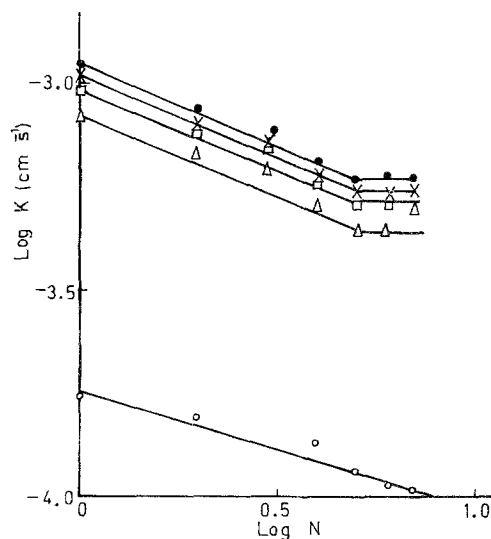


Fig. 5. Effect of number of screens ( $N$ ) per stack on the mass transfer coefficient at different air superficial velocities. ( $C = 0.2 M$ .)  $\circ$ ,  $0.0 V cm s^{-1}$ ;  $\Delta$ ,  $0.02122 V cm s^{-1}$ ;  $\square$ ,  $0.04244 V cm s^{-1}$ ;  $\times$ ,  $0.06366 V cm s^{-1}$ ;  $\bullet$ ,  $0.08488 V cm s^{-1}$ .

used to account for this effect, namely the ratio between bed thickness ( $L$ ) and the screen wire diameter ( $d$ ). Fig. 6 shows that the data for the bed electrode fit the equation

$$J = 0.2(ReFr)^{-0.28}(L/d)^{-0.28} \quad (4)$$

with an arithmetic average deviation of  $\pm 10\%$ . For mass transfer at a single vertical screen electrode, the data were found to fit the equation

$$J = 0.187(ReFr)^{-0.26} \quad (5)$$

with an arithmetic average deviation of  $\pm 16.6\%$ .

Since previous studies in the area of heat and mass transfer have shown that the pore size of the gas distributor has a negligible effect on the rate of heat and mass transfer [7, 18], it follows that the present correlations can be used in practice to predict rates of mass transfer in gas sparged batch and continuous fixed bed reactors (with low solution flow rates) [21–24] regardless of the pore size of the gas distributor. However, Equations 4 and 5 need to be verified using screens of different porosity, wire diameter and mesh number.

Fig. 7 shows the effect of air sparging on the total cell voltage; the observed increase in the cell voltage over the natural convection value may be attributed mainly to the increase in the

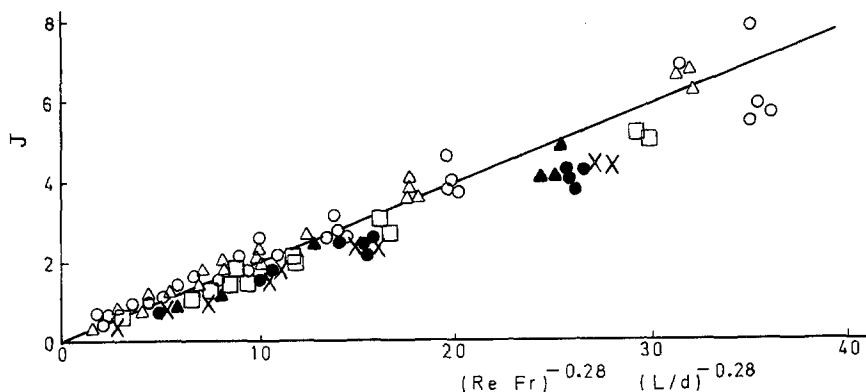


Fig. 6. Overall mass transfer correlation for stacks of vertical screens. Number of screens per stack: O, 2;  $\Delta$ , 3;  $\square$ , 4; x, 5;  $\bullet$ , 6;  $\blacksquare$ , 7.

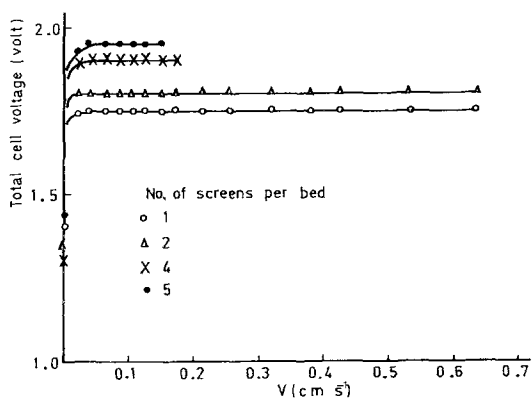


Fig. 7. Effect of superficial air velocity on the total cell voltage at a vertical screen stack of different thickness. ( $C = 0.2M$ .)

$IR$  drop as a result of increasing the limiting current. The constancy of the cell voltage with air superficial velocity after the initial increase may be ascribed to a balance between the decrease in concentration polarization at the counterelectrode (screen anode) and the increase in  $IR$  drop. The anode, which has a larger area than the bed cathode, suffers from concentration polarization because the anodic reaction is taking place below the anodic limiting current.

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