

Natural convection mass transfer at horizontal screens

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The free convection mass transfer behaviour of horizontal screens has been investigated experimentally using an electrochemical technique involving the measurement of the limiting currents for the cathodic deposition of copper from acidified copper sulphate solutions. Screen diameter and copper sulphate concentration have been varied to provide a range of $Sc.Gr$ from 22×10^8 to 26×10^{10} . Under these conditions, the data for a single screen are correlated by the equation:

$$Sh = 0.375(Sc.Gr)^{0.305}$$

Results have been compared with previous work on free convection at horizontal solid surfaces where mass transfer coefficients are somewhat lower.

Mass transfer coefficients have been measured also for arrays of closely spaced parallel horizontal screens. The mass transfer coefficient was found to decrease with the number of screens forming the array.

Symbols and units

A	area of mass transfer surface, cm^2 .
C_b	bulk concentration of ionic species, mol cm^{-3} .
D	diffusivity, $\text{cm}^2 \text{s}^{-1}$.
F	Faraday number, 96 494 C g [equiv ⁻¹]
Z	number of electrons involved in the reaction.
I_L	limiting current, A
K	mass transfer coefficient, cm s^{-1} .
Sh	Sherwood number, dK/D
Sc	Schmidt number, $\mu/\rho D$ or ν/D
Gr	Grashof number $gd^3\Delta\rho/\nu^2\rho_s$
μ	solution dynamic viscosity, g cm s^{-1} .
ν	solution kinematic viscosity, $\text{cm}^2 \text{s}^{-1}$.
ρ	solution density, g cm^{-3} .
$\Delta\rho$	density difference between bulk solution and electrode/solution interface, g cm^{-3} .
ρ_s	solution density at electrode/solution interface, g cm^{-3} .
d	screen diameter, cm.
g	gravitational acceleration, cm s^{-2} .

1. Introduction

Horizontal screens are frequently used as a catalytic surface upon which heterogeneous reactions take place. The use of screens as electrodes in electrochemical processes is attractive in view of the high surface area/unit volume. The performance of screens as an electrode or a catalytic surface depends on the mass transfer characteristics of the screen and screen arrangement.

Although some work has been carried out on forced convection mass and heat transfer at horizontal screens, no work on natural convection mass or heat transfer at horizontal screens has been reported apparently. Mass transfer rates to a flowing liquid from a horizontal screen were measured by Vogtlander and Bakker [1] using the electrochemical technique. Maroof and Donald [2] studied forced convection mass transfer at stacked metal screens using the decomposition of hydrogen peroxide vapour; they found that their data agreed well with the heat transfer correlation for a single cylinder. Satterfield and Cortez

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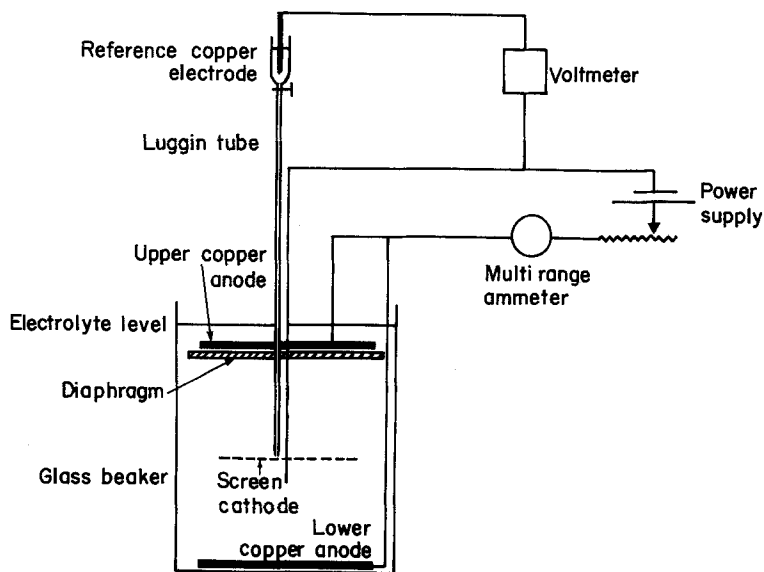


Fig. 1. Cell and electrical circuit.

[3] determined the mass transfer characteristics of horizontal woven screens by measuring the rate of oxidation of hexene and toluene on platinum screen catalyst and concluded that the transport characteristic of a screen catalyst is very similar to that of an assembly of infinite cylinders.

Wragg [4] studied natural convection mass transfer at a vertical mesh electrode using the cathodic reduction of $K_3Fe(CN)_6$ and concluded that the mass transfer behaviour at a vertical mesh electrode is very similar to that at a vertical flat plate of the same total exposed area. In the present investigation, natural convection mass transfer at a single horizontal screen and at arrays of closely spaced horizontal screens was studied using the electrochemical technique. The mass transfer coefficient was determined by measuring the limiting current of the cathodic deposition of copper from $CuSO_4$ solution using H_2SO_4 as a supporting electrolyte to eliminate the transfer of copper ions to the screen surface by electrical migration. Under these conditions, the limiting current is related to the mass transfer coefficient by the equation

$$K = \frac{I_L}{ZFAC_b} \quad (1)$$

2. Experimental

The cell consisted of a 1 l cylindrical glass container, 12.5 cm tall and 10 cm in diameter. The

anode assembly was made of two copper discs each 9 cm diameter, one disc being placed at the bottom of the container and the other at a distance 11 cm above. The circular screen cathode was placed midway between the two anodes thus assuring uniform current distribution on both sides of the screen. To avoid interference between natural convection at the upper anode and that at the screen cathode, a porous diaphragm made of synthetic fibre and supported on a plastic screen was placed below the upper anode 5 cm from the screen cathode. According to Wragg [5], at a distance of 5 cm from the cathode, the presence of a diaphragm has no effect on the mass transfer rate at the horizontal cathode. The upper anode and diaphragm were held in position by mounting them on the vertical Luggin tube used in measuring the cathode potential, the Luggin tube being clamped to a stand. The screen cathode was held in position by a vertical copper wire of 1 mm diameter, electrically insulated except at its contact with the screen cathode.

The electrical circuit, shown in Fig. 1 with the cell, consisted of a 6 V – 7 A d.c. power supply and a multirange ammeter connected in series with the cell. Before each run, the screen cathode was degreased with trichloroethylene, and then washed with alcohol and with distilled water. The cell was filled with 950 c.c. of the electrolyte solution. Current–potential curves were constructed by increasing the applied current stepwise and measuring

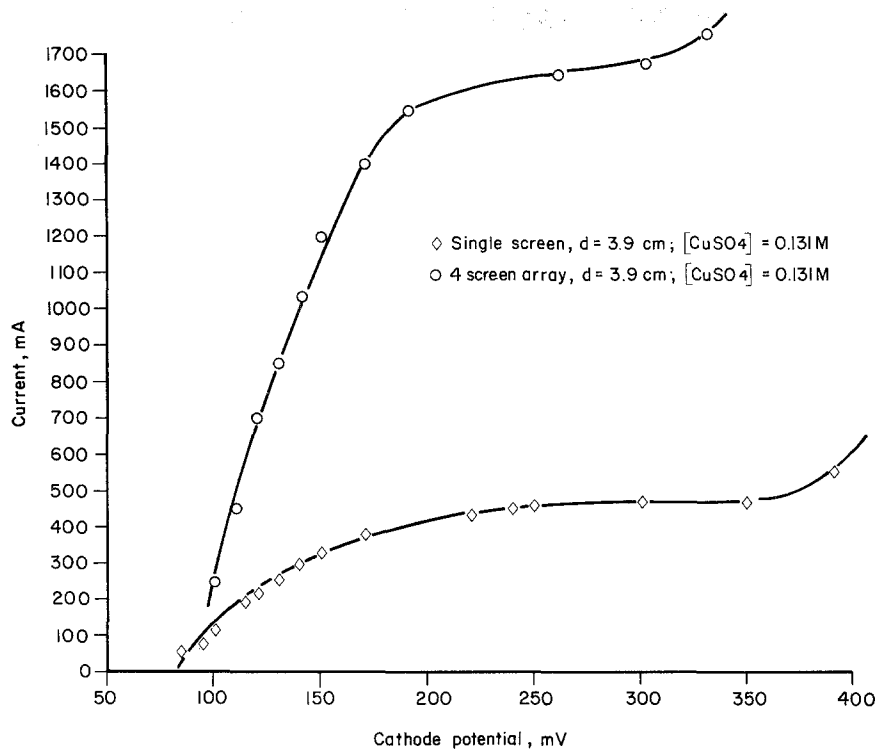


Fig. 2. Typical current-potential curves for a single screen and for an array of screens.

the corresponding steady-state cathode potential. The latter was measured with a voltmeter against a reference copper electrode placed in the cup of the Luggin tube. The Luggin tube had its capillary tip positioned 0.5–1 mm from the screen surface; the tube being filled with the same solution as in the cell. Each experiment was repeated 2 to 3 times. Temperature was kept constant at $20 \pm 0.5^\circ \text{C}$.

Five screen cathodes of diameters 2.88, 3.91, 5.13, 6.26 and 7.50 cm were used; all were made of stainless steel screen with a count of 20 square apertures per lineal inch, wire diameter of 0.375 mm and porosity of 0.75. Solutions of CuSO_4 of four different concentrations were used: 0.05, 0.131, 0.25 and 0.46 M; in all cases 1.5 M H_2SO_4 being employed as a supporting electrolyte.

In the case of the screen arrays, sets of screens, from 2 to 7 in number, all of the one diameter, 3.9 cm, were prepared. The screens were closely packed (touching each other, hence with zero spacing) and were held together and fixed in position by a vertical 1 mm copper wire passing the array centre line. The copper wire was insulated except at the point of contact with the array.

The Luggin tip was positioned 0.5–1.0 mm from the top screen of the array at the centre line.

3. Results and discussion

Typical current-potential results are shown in Fig. 2 for a single screen cathode and for an array of 4 screens. The plateau corresponding to the limiting current shrinks with an increase in the number of screens per array. This number was limited to a maximum of 7 in the present work, since beyond this number, the plateau of the limiting current became ill-defined. From the current-potential curves, the limiting currents were determined and the mass transfer coefficients calculated using Equation 1. The true surface area of the screen was calculated from the number of apertures/lineal cm and the wire diameter using the method of Armour and Cannon [6].

Fig. 3 shows the relationship of screen electrode diameters to the mass transfer coefficients for the 4 different solution concentrations investigated. The coefficients increase directly with concentration despite Equation 1 which implies an in-

Table 1. Mass transfer at a single screen electrode

		$C_{H_2SO_4} = 1.5\text{ M}$		$T = 20^\circ\text{ C}$			
$CuSO_4$ Conc. (M)	Screen diameter (cm)	Limiting current (mA)	$K \times 10^5$ ($cm\ s^{-1}$)	Sh	Sc	$Gr. \times 10^{-4}$	$Sc.Gr \times 10^{-6}$
0.050	2.88	74	52.4	282.6	2343	94.7	2 220
0.050	3.91	132	50.7	371.2	2343	229	5 367
0.050	5.13	225	50.0	480.0	2343	535	12 550
0.050	6.26	330	49.4	579.0	2343	973	22 790
0.050	7.50	460	47.9	672.7	2343	1 673	39 200
0.131	2.88	250	67.6	343.4	2148	250	5 370
0.131	3.91	465	67.4	464.6	2148	626	13 440
0.131	5.13	780	66.4	600.8	2148	1 413	30 350
0.131	6.26	1125	64.3	709.9	2148	2 567	55 140
0.131	7.50	1500	60.0	794.0	2148	4 415	94 840
0.250	2.88	510	72.3	395.8	2449	380	9 312
0.250	3.91	925	71.0	527.7	2449	952	23 300
0.250	5.13	1550	69.0	672.9	2449	2 149	52 630
0.250	6.26	2250	67.0	800.0	2449	3 905	95 620
0.250	7.50	3100	65.0	926.8	2449	6 715	164 500
0.460	2.88	1100	84.7	487.9	2691	704	18 940
0.460	3.91	2000	83.4	652.2	2691	1 761	47 380
0.460	5.13	3400	82.0	841.3	2691	3 977	107 000
0.460	6.26	5000	81.3	1076.0	2691	7 226	194 400
0.460	7.50	7000	79.0	1185.0	2691	12 430	267 200

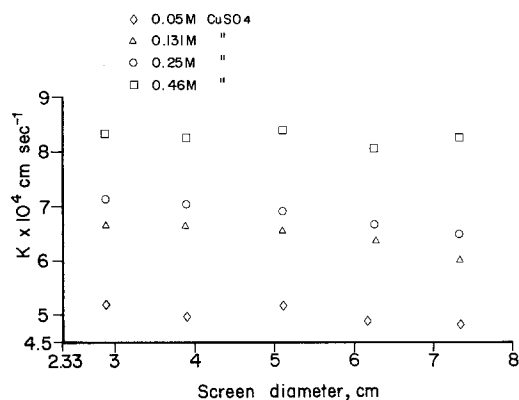


Fig. 3. Effect of screen diameter on the mass transfer coefficient.

verse relationship. The increase in K with solution concentration can be attributed to the change in physical properties of the solution (ρ , μ , D) with increasing concentration. Since $K = D/\delta$, where δ is the Nernst diffusion layer thickness, it appears that the net effect of increasing $CuSO_4$ concentration is to increase the buoyancy force, $(\rho - \rho_s)g$, and decrease δ much more than the decrease in D . Hence the K values increase with greater $CuSO_4$ concentration.

The values of K (Fig. 3) are almost independent of screen diameter, with a tendency towards a slight decrease in value with increasing electrode diameter. Edge effects, decreasing in relative importance with diameter, would probably account for this trend. According to hydrodynamic boundary layer theory, transfer coefficients become independent of electrode dimensions in the case of turbulent natural convection heat or mass transfer. A means for differentiating between laminar and turbulent mechanisms is thus provided, and the results here support the concept that mass transfer at the screen surface takes place by a turbulent flow mechanism. Similar findings were reported by both Wragg [5] and Fenech and Tobias [7] in their studies on mass transfer by free convection at horizontal solid electrodes.

An overall correlation of the data in terms of the dimensionless groups, Sh , Sc and Gr , was envisaged, and accordingly the values for these groups were calculated. Table 1 summarizes the results of the calculations for single screens. Values for the physical properties of the solutions used (ρ , μ and D) were taken from appropriate sources [8–10]. The characteristic dimension for the Sh and Gr numbers was chosen as the screen electrode diameter.

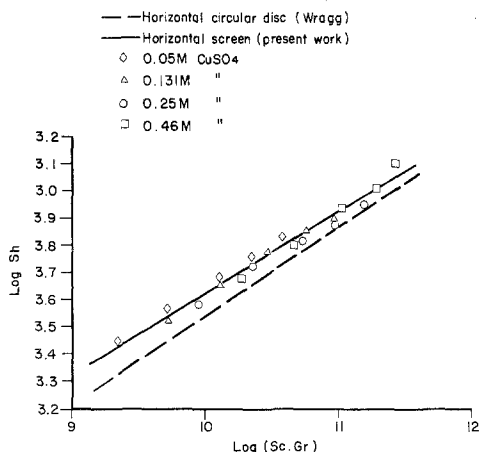


Fig. 4. Log Sh versus $\log (Sc.Gr)$.

Fig. 4 is a plot of $\log Sh$ against $\log Sc.Gr$ for single screens over a range of $Sc.Gr$ extending from 22×10^8 to 26×10^{10} . The data fit the equation (the solid line in Fig. 4):

$$Sh = 0.375(Sc.Gr)^{0.305}, \quad (2)$$

with an average deviation of 3.5% and a maximum deviation of 6.5%. Wragg [5] correlated his data for mass transfer at horizontal solid disc electrodes facing upward, within the range of $Sc.Gr$ studied here, by the equation:

$$Sh = 0.16(Sc.Gr)^{0.33}. \quad (3)$$

The present data fit this equation with an average deviation of 15.5% and a maximum deviation of 26% (the broken line in Fig. 4), although all mass transfer coefficients as determined here have greater values than predicted by the equation. The value of the exponent (optimally for these data at 0.305, but a not inappropriate verification of 0.33) points again to a mass-exchange mechanism by turbulent eddies.

For the corresponding case of mass transfer at vertical mesh electrodes compared to vertical flat plates, Wragg [5] found that the mesh electrode data fitted the flat plate correlation ($Sh = 0.45(Sc.Gr)^{0.25}$) with maximum deviations of about 27%. In this case, however, there were both negative and positive deviations, with a preponderance of positive deviations. In this general sense, therefore, both vertical and horizontal screen electrodes give higher mass transfer coefficients than the corresponding flat surfaces. The slightly greater coefficients are to be expected

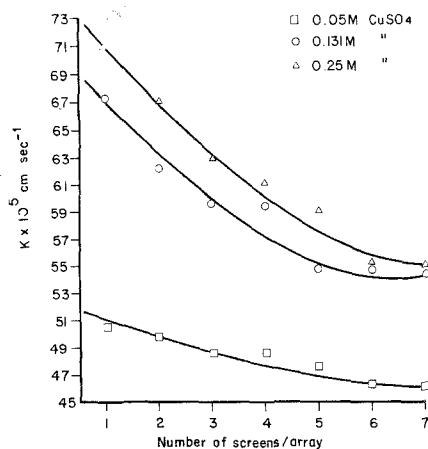


Fig. 5. Effect of number of screens per array on the mass transfer coefficient.

from a consideration of the nature of the screen electrode with parallel anodes on each side of it, and its hydrodynamic characteristics because of the apertures. The cathodically active bounding surfaces of the screen spaces will produce a complex pattern of convective streams merging into main streamlines. The resultant disturbances, no matter how rapidly attenuated, will enhance mass transfer [4]. As Wragg has also indicated [4]: 'Since the surface of the electrolyte is discontinuous, there will be a tendency toward boundary-layer decay and re-establishment, each bar of the mesh acting as a leading edge within a partially developed mass-transfer boundary layer.' While both these tendencies should be somewhat concentration dependent, it is clear from earlier work [4] that the concentration used here are sufficiently high to avoid very significant dependency.

The effect on the mass transfer coefficient of using a number of closely spaced screens to form an electrode array is shown in Fig. 5. The coefficients decrease with an increased number of screens, although the coefficient values appear to be approaching a constant value for 6 and 7 screens (the maximum number for which limiting currents could be determined). The method of forming an array as a single electrode provided comparable conditions for operation and measurement of each array. As a result, the changing nature of the current-voltage curves (gradual disappearance of the plateau) with increased thickness of the array is not considered due to any change in electrochemical mechanism but rather to

different solution concentrations at different parts of the electrode array. The upward flowing solution is progressively depleted in copper ions as it passes through the array. This decrease in average bulk concentration for each successive portion of electrode surface results in the overall decrease in mass transfer coefficient. Nevertheless, the existence of natural convection of substantial magnitude in the case of screen arrays with their high porosity and high surface area/unit volume ratio make such arrays promising electrode systems for electrochemical processes.

Acknowledgement

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