

Effect of drag-reducing additives on the rate of mass transfer in a parallel-plate electrochemical flow reactor

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The effect of polyox and CMC drag-reducing polymers on the rate of mass transfer in a parallel-plate flow cell was studied by measuring the limiting current for the cathodic reduction of potassium ferricyanide in alkaline medium. Reynolds number and polymer concentration were varied over the range 3500–21 000 and 10–200 ppm respectively. Under these conditions it was found that polyox and CMC reduce the rate of mass transfer by a maximum of 42% and 35% respectively.

Nomenclature

| | |
|--------|-----------------------------------------------------------------------------|
| a | a constant |
| C | concentration of ferricyanide ion (g mol cm ⁻³) |
| D | diffusivity of ferricyanide ion (cm ² s ⁻¹) |
| d_e | equivalent diameter of the cell (4 × cross-sectional area/wetted perimeter) |
| F | Faraday's constant (96 487 C mol ⁻¹) |
| I | limiting current density (A cm ⁻²) |
| K | mass transfer coefficient (cm s ⁻¹) |
| L | electrode height (cm) |
| (Re) | Reynolds number ($\rho v d_e / \mu$) |
| (Sc) | Schmidt number ($u / \rho D$) |
| (Sh) | Sherwood number ($K d_e / D$) |
| u | solution viscosity (poise) |
| v | flow rate of the solution (cm s ⁻¹) |
| Z | number of electrons involved in the reaction |
| ρ | solution density (g cm ⁻³) |

1. Introduction

Previous studies of the effect of drag-reducing polymers on electrochemical mass transfer have dealt with the rotating-cylinder electrode [1–5], the rotating-disc electrode [6] and the tubular

electrode [7–9]. Despite the importance of the parallel-plate cell in practice, no studies have been reported of the effect of drag-reducing polymers on the rate of mass transfer in such cells. Previous studies [10] of the effect of drag-reducing polymers on momentum transfer have shown that the geometry of the solid surface has a profound effect on the performance of the polymer. The object of the present work is to study the effect of polyox and CMC drag-reducing polymers on the rate of mass transfer in parallel-plate flow cells under turbulent flow conditions. The study will assist in assessing the feasibility of applying the phenomenon of drag reduction to electrochemical processes to cut down the pumping power requirement.

2. Experimental technique

The apparatus used (Fig. 1) consisted of a 24 dm³ plexiglass storage tank, a stainless steel centrifugal pump and a vertical parallel-plate rectangular cell. The cell consisted of three sections, the inlet section, the working section and the outlet section. The inlet section had a height of 250 cm (63 d_e), this being sufficient to give fully developed

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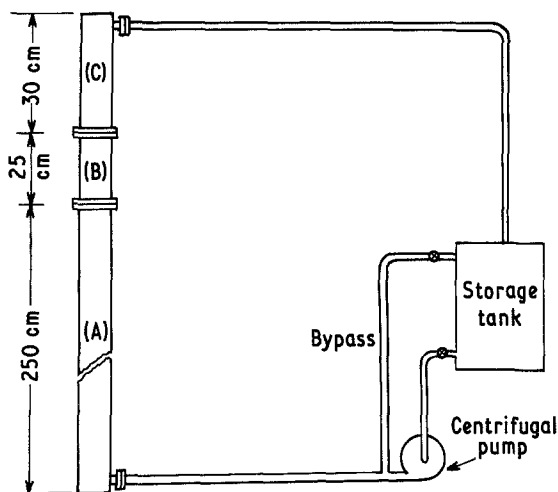


Fig. 1. Experimental apparatus. (A) Entrance section, (B) working section, (C) outlet section.

flow in the working section [11]. The outlet section had a height of 30 cm. The working section (electrolysis cell) had a height of 25 cm and consisted of two flat nickel-plated copper electrodes: each electrode was fixed in position through grooves cut in the walls of the working section and the surface of each was flush with the walls of the inlet and outlet sections. The electrodes were 25 cm in height and 6 cm wide; electrode separation was 3 cm. The electrical circuit consisted of a 6 V d.c. power supply with a voltage regulator, and a multi-range ammeter connected in series with the cell.

Before and during each run, dissolved oxygen was removed from the storage tank by bubbling nitrogen gas through it. Polarization curves were constructed under different conditions by increasing the applied current stepwise and measuring the corresponding steady-state cathode potential against a reference electrode made of a nickel wire dipped into the cup of a Luggin tube (not shown in Fig. 1). The Luggin tube penetrates the top of the outlet section through a tight rubber stopper; the capillary end of the Luggin is positioned 0.5–1 mm from the cathode surface. Limiting currents were determined from the polarization curves and used to calculate the mass transfer coefficients from the equation:

$$I/ZF = KC. \quad (1)$$

Solutions used were composed of a blank solution

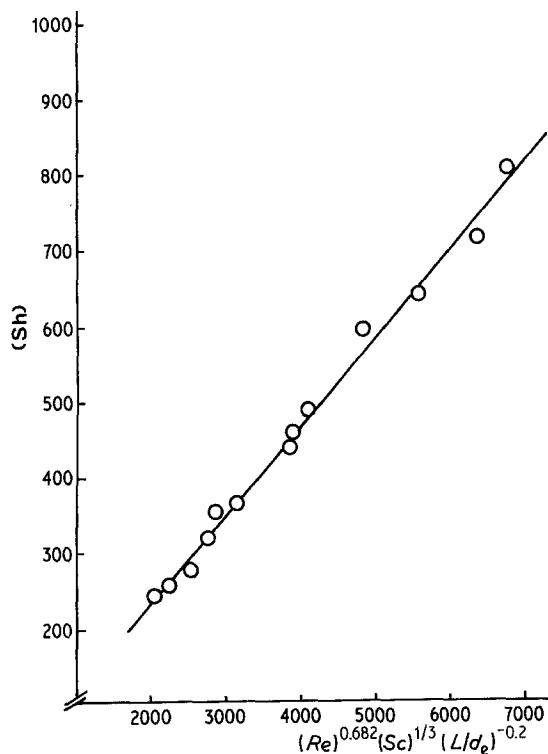


Fig. 2. Overall mass transfer correlation for the cathodic reduction of potassium ferricyanide in solution free of polymer (blank).

and a few ppm of polyethylene oxide (polyox WSR 301, a product of Union Carbide) or carboxymethyl cellulose (CMC-7H, a produce of Hercules Chemical Company). The blank solution consisted of 0.025 M $K_3Fe(CN)_6$ + 0.025 M $K_4Fe(CN)_6$ + 1 M NaOH. All solutions were prepared from AR-grade chemicals and distilled water. Polymer solutions were prepared by digesting the required weight of the polymer in water for 12 hours. After polymer swelling the polymer solution was mixed with a solution containing $K_3Fe(CN)_6$, $K_4Fe(CN)_6$ and NaOH to give the required concentration. Polymer concentration ranged from 10 to 200 ppm. All solutions were found by rheological measurements to be Newtonian. Within the concentration range used here, polymer addition was found to have a negligible effect on the physical properties of the solution. Diffusivity of ferricyanide ion was calculated from the formula of Gordon *et al.* [12].

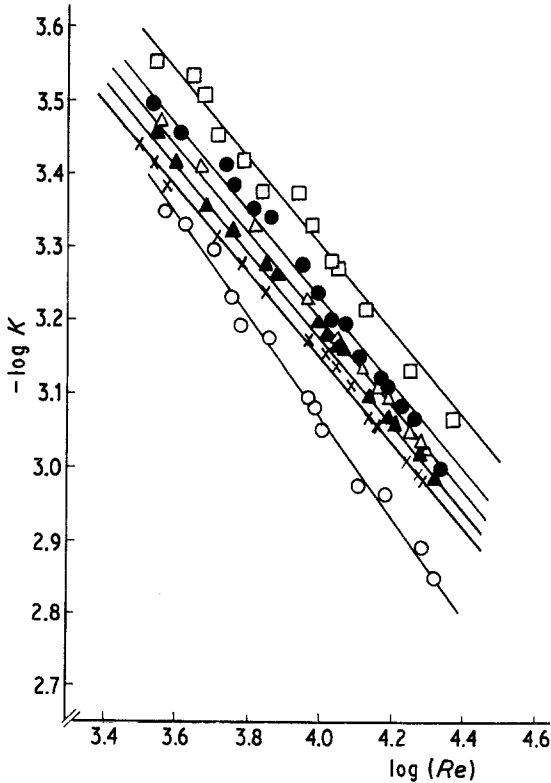


Fig. 3. $\log K$ versus $\log (Re)$ for the cathodic reduction of potassium ferricyanide in solution containing polyox. Concentration of polyox (in ppm): \circ , blank; \times , 10; \blacktriangle , 20; \triangle , 50; \bullet , 100; \square , 200.

3. Results and discussions

Fig. 2 shows that the mass transfer data in the fully hydrodynamic flow region can be correlated under turbulent flow conditions for the blank solution by the equation:

$$(Sh) = 0.114(Sc)^{1/3}(Re)^{0.682}(L/d_e)^{-0.2} \quad (2)$$

with an average deviation of 2.3%. In obtaining the above equation the exponents of (Sc) and (L/d_e) were assumed to be $\frac{1}{3}$ and -0.2 respectively. The present data for the blank solution agree with the equation obtained by Pickett and Ong [13] under similar conditions:

$$(Sh) = 0.125(Sc)^{1/3}(Re)^{0.66}(L/d_e)^{-0.22} \quad (3)$$

with an average deviation of -6.8% .

Figs. 3 and 4 show the effect of Reynolds number on the mass transfer coefficient in solutions containing different concentrations of polyox and CMC respectively. The data can be represented by the following equations:

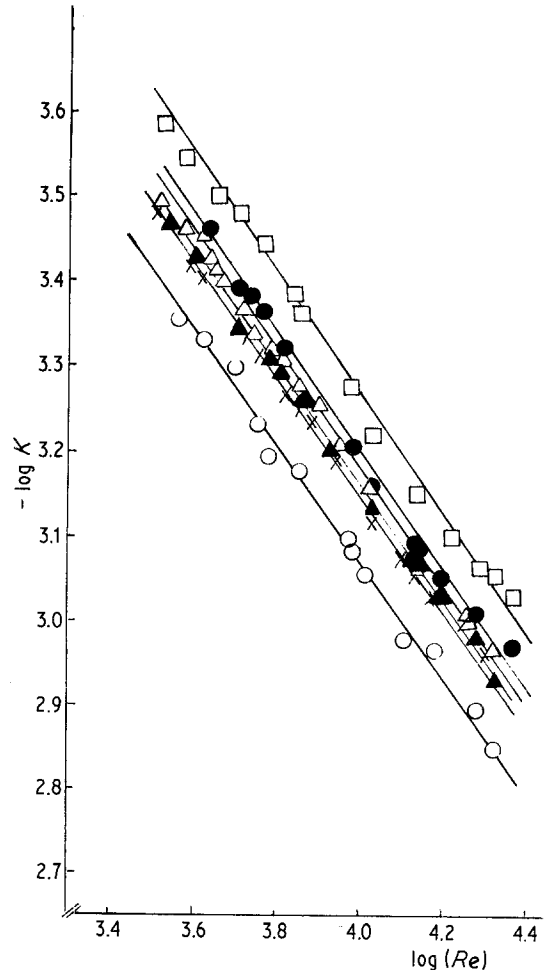


Fig. 4. $\log K$ versus $\log (Re)$ for the cathodic reduction of potassium ferricyanide in solutions containing CMC. Concentration of CMC (in ppm): \circ , blank; \times , 10; \blacktriangle , 20; \triangle , 50; \bullet , 100; \square , 200.

$$K = a(Re)^{0.687} \quad \text{blank} \quad (4)$$

$$K = a(Re)^{0.63} \quad \text{polyox} \quad (5)$$

$$K = a(Re)^{0.687} \quad \text{CMC.} \quad (6)$$

The constant a in the above equations decreases with increasing polymer concentration.

Figs. 5 and 6 show that the percentage reduction in the mass transfer coefficient ranges from 6 to 41 and from 15 to 36 for polyox and CMC respectively. The higher the polymer concentration, the higher the percentage reduction in the mass transfer coefficient. The percentage reduction in the mass transfer coefficient increases with Reynolds number until it reaches a maximum and then starts to decrease with further increase in

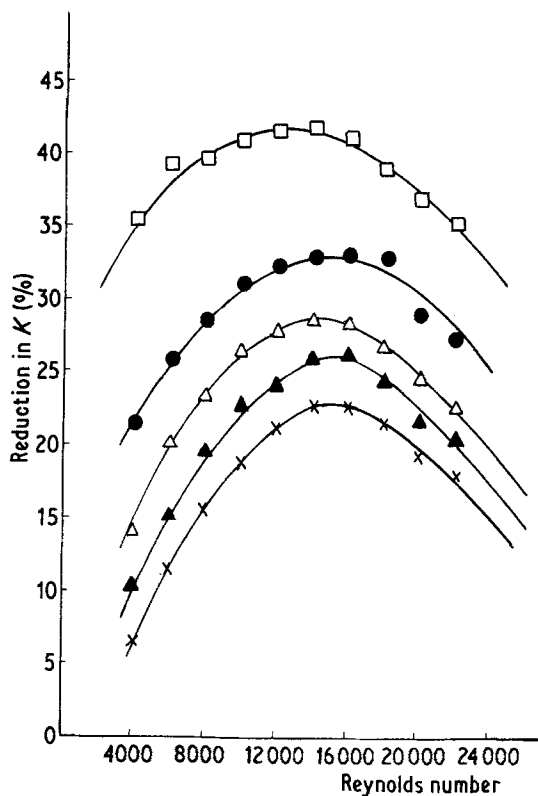


Fig. 5. Effect of Reynolds number and polyox concentration on the percentage reduction in the mass transfer coefficient of the cathodic reduction of potassium ferricyanide. Concentration of polyox (in ppm): \times , 10; \blacktriangle , 20; \triangle , 50; \bullet , 100; \square , 200.

Reynolds number. The initial increase may be attributed to the increased degree of stretching of the polymer molecules with increasing Reynolds number. The higher the degree of polymer expansion the higher its ability to dampen turbulent eddies [14]. The decrease in the percentage reduction of mass transfer coefficient at relatively high Reynolds number may be attributed to degradation of the polymer molecules by the high shear stress exerted on them at high Reynolds number. The decrease in polymer effectiveness at relatively high Reynolds number was also observed in momentum transfer studies. Levy and Davis [15] measured the drag between a flat plate and a solution containing 15 ppm polyox; they obtained a maximum drag reduction of 65% at a Reynolds number of 7×10^6 and at higher Reynolds number the percentage drag reduction tended to decrease. Hoyt and Fabula [16] measured the drag between

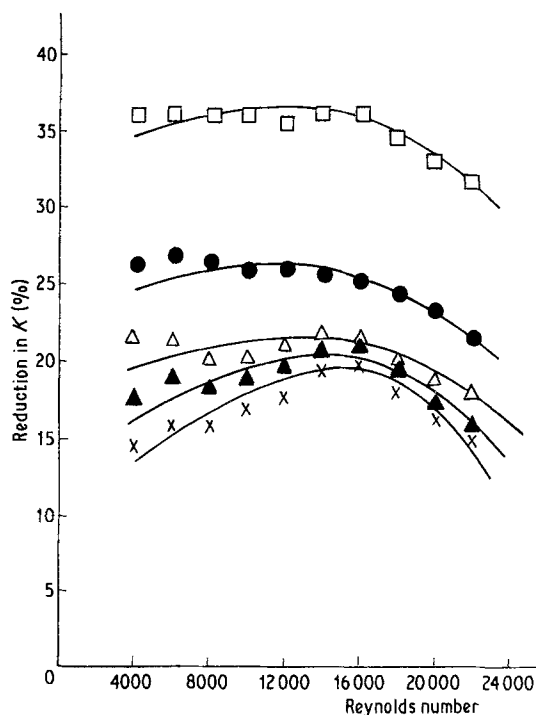


Fig. 6. Effect of Reynolds number and CMC concentration on the percentage reduction in the mass transfer coefficient of the cathodic reduction of potassium ferricyanide. Concentration of CMC (in ppm): \times , 10; \blacktriangle , 20; \triangle , 50; \bullet , 100; \square , 200.

a tube wall and a solution containing polyox; they found that for 30 ppm polyox the percentage drag reduction reaches a maximum of 75% at a Reynolds number of 55 000 and that a further increase of Reynolds number reduced the percentage drag reduction.

In agreement with a previous study [1] using a rotating cylinder, Figs. 5 and 6 show that, in general, polyox is more effective than CMC in reducing the rate of mass transfer. This is also consistent with the fact that polyox is a more effective drag-reducing polymer than CMC because of its higher molecular weight and higher flexibility [10]. A comparison between the mass transfer characteristics of the rotating-cylinder electrode in drag-reducing solutions and the present results indicates that, contrary to the present finding, the percentage reduction in the mass transfer coefficient shows little dependence on Reynolds number and polymer concentration in the case of the rotating cylinder. Since polymer molecules damp only small-scale, high-intensity

eddies prevailing in the buffer layer of the hydrodynamic boundary layer [10], the difference in behaviour between the two geometries may be attributed to the fact that the proportion of such eddies in the turbulence spectrum and its dependence on Reynolds number is different at the two geometries. In the case of the rectangular duct, the hydrodynamic situation is complicated by the presence of a transverse mean flow superimposed upon the axial mean flow [17]. This transverse flow, commonly known as secondary flow, interacts with the axial flow and turbulence structure in a complex manner.

Unfortunately no comprehensive study was made of the flow behaviour of drag-reducing fluids in rectangular ducts. Only few data can be obtained from the literature for the purpose of comparison with the present results. Reischman and Tiederman [18], who studied mainly velocity distribution in drag-reducing channel flows, reported 35.3% drag reduction for a solution containing 100 ppm polyox at a Reynolds number of 52 400. The data of Clark and Rodriguez [19] on the flow of drag-reducing fluids in rectangular ducts show that for Reynolds numbers of 11 200 and 14 800 the percentage drag reduction for a solution containing 100 ppm polyox is 52.9 and 56.9 respectively. A comparison of the percentage reduction in the mass transfer coefficient obtained in the present work (Fig. 5) with the percentage drag reduction obtained by Clark and Rodriguez shows that for the same Reynolds number the percentage reduction in mass transfer is less than the percentage reduction in drag. In view of this it can be concluded that in the case of parallel-plate cells, application of the phenomenon of drag reduction to diffusion-controlled electrochemical processes may be economical, i.e. the saving in the pumping power requirement may outweigh the adverse effect of the decrease in the rate of production. For charge-transfer controlled processes conducted under turbulent flow, e.g. electrochemical machining, application of the phenomenon of drag reduction would be of greater economic value. However, two problems remain to be solved if the phenomenon of drag reduction is to be applied to electrochemical processes, namely (a) chemical and mechanical stability of the polymer in the

electrolyte and (b) polymer solubility in the electrolyte. Recent studies have shown that these problems can be bypassed by using solid drag-reducing agents such as dilute suspensions of, for example, asbestos, glass fibres or synthetic fibres [20–27].

References

- [1] G. H. Sedahmed and A. Abdel-Khalik, *J. Appl. Electrochem.* **10** (1980) 245.
- [2] G. H. Sedahmed, B. A. Abdel-Naby and A. Abdel-Khalik, *ibid* **8** (1978) 473.
- [3] *Idem*, *Corros. Sci.* **17** (1977) 865.
- [4] *Idem*, *J. Appl. Electrochem.* **7** (1977) 355
- [5] G. H. Sedahmed, A. Abdel-Khalik, A. M. Abdallah and M. M. Farahat, *ibid* **9** (1979) 567.
- [6] C. Deslouis, I. Epelboin, B. Yribollet and L. Viet, *Electrochim. Acta* **20** (1975) 909.
- [7] G. A. McConaghy and T. J. Hanratty, *AIChE. J.* **23** (1977) 493.
- [8] G. H. Sedahmed and R. G. Griskey, *ibid* **18** (1972) 138.
- [9] G. H. Sedahmed, M. S. Abdo, H. A. Farag and S. G. Tantawy, *Surf. Technol.* **9** (1979) 359.
- [10] J. W. Hoyt and J. Bas, *Engng. Trans. ASME* **84D** (1972) 258.
- [11] J. Knudsen and D. Katz, 'Fluid Dynamics and Heat Transfer', McGraw-Hill, New York (1958).
- [12] S. L. Gordon, J. S. Newman and C. W. Tobias, *Ber. Bunsenges Physik. Chem.* **70** (1966) 414.
- [13] D. J. Pickett and K. L. Ong, *Electrochim. Acta* **19** (1974) 875.
- [14] D. White and R. J. Gordon, *AIChE. J.* **21** (1975) 1027.
- [15] J. Levy and S. Davis, *Int. Shipbuilding Prog.* **14** (1967) 166.
- [16] J. W. Hoyt and A. G. Fabula, *Proc. 5th Symp. on Naval Hydrodynamics, Bergen, Norway*, Office of Naval Research ACR-112 (1964) p. 947.
- [17] F. B. Gessner and J. B. Jones, *J. Fluid Mech.* **23** (1965) 689.
- [18] M. M. Reischman and W. G. Tiederman, *ibid* **70** (1975) 369.
- [19] D. B. Clark and F. Rodriguez, *J. Appl. Polymer Sci.* **20** (1976) 315.
- [20] W. K. Lee, R. C. Vaseleski and A. B. Metzner, *AIChE. J.* **20** (1974) 128.
- [21] P. F. Lee and G. G. Duffy, *ibid* **22** (1976) 750.
- [22] D. D. Kale and A. B. Metzner, *ibid* **20** (1974) 1218.
- [23] R. C. Vaseleski and A. B. Metzner, *ibid* **20** (1974) 301.
- [24] P. Peyser, *J. Appl. Polymer Sci.* **17** (1973) 421.
- [25] A. L. Moyls and R. H. Sabersky, *Int. J. Heat Mass Transfer* **21** (1978) 7.
- [26] I. Radin, J. L. Zakin and G. K. Patterson, *AIChE. J.* **21** (1975) 358.
- [27] G. K. Patterson, J. L. Zakin and J. M. Rodriguez, *Ind. Eng. Chem.* **61** (1969) 20.