

Effect of drag-reducing polymer on the rate of mass transfer in fixed-bed reactors

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Rates of mass transfer were measured for the cementation of copper from dilute copper sulphate solutions containing polyethylene oxide drag-reducing polymer on a fixed bed of zinc pellets. Starting from a Reynolds number (Re) of 550, the rate of mass transfer was found to decrease by an amount ranging from 7.5 to 51% depending on Re and polymer concentration. The percentage decrease in the rate of mass transfer increased with increasing Re , passed through a maximum at $Re = 1400$ and then decreased rapidly with further increase in Re . The possibility of using drag-reducing polymers to reduce power consumption in fixed-bed operation was discussed in the light of the present and previous results.

Nomenclature

A cross-section of reactor (m^2)
 a specific area of bed (m^2)
 C copper sulphate concentration at time t ($mol\ l^{-1}$)
 C_0 initial copper sulphate concentration ($mol\ l^{-1}$)
 D diffusivity of copper sulphate ($m^2\ s^{-1}$)
 d_p particle diameter (m)
 J_d mass transfer J-factor ($StSc^{2/3}$)
 K mass transfer coefficient ($m\ s^{-1}$)

L bed height (m)
 Q volumetric flow rate ($m^3\ s^{-1}$)
 Re Reynolds number ($\rho V_i d_p / \mu$)
 Sc Schmidt number ($\mu / \rho D$)
 St Stanton number (K / V_i)
 V volume of copper sulphate solution (m^3)
 V_i interstitial velocity (V_s / ϵ), ($m\ s^{-1}$)
 V_s superficial velocity ($m\ s^{-1}$)
 ϵ bed porosity
 μ solution viscosity ($kg\ m\ s^{-1}$)
 ρ solution density ($kg\ m^{-3}$)
 τ storage tank residence time (s)

1. Introduction

Because of their high specific area and high mass transfer coefficient, fixed-bed reactors are gaining an increasing importance in conducting diffusion-controlled reactions involving dilute solutions, such as in removing toxic or valuable metals from industrial waste solutions by either cementation [1–5] or electrolysis [6]. However, fixed beds suffer from high pressure drop especially when operated at high flow rates. According to

Hanna *et al.* [7] the problem of high pressure drop could be alleviated by using drag-reducing polymers. These polymers do not pose any environmental threat as they are non-toxic and biodegradable [8]. Since drag-reducing polymers function by damping small-scale, high-frequency eddies which prevail in the turbulent hydrodynamic boundary layer, it is inevitable that drag reduction inside the bed by the polymer molecules will be accompanied by a parallel decrease in the rate of mass transfer when the

bed is used for conducting diffusion-controlled reactions. The object of the present work is to investigate the extent to which drag-reducing polymers reduce the rate of mass transfer in order to assist in assessing the feasibility of using such polymers in fixed-bed operation. The present study was conducted using the cementation of copper on a fixed bed of zinc pellets as the mass transfer process. The reaction is known to be diffusion-controlled [9–12] and is used in industry to purify $ZnSO_4$ leach liquor from more noble impurities in the electro-winning of zinc. A literature review of the effect of drag-reducing polymers on the pressure drop across fixed beds reveals a striking contradiction among different authors. Laufer *et al.* [13] found that drag-reducing polymers increase the pressure drop across the bed by a factor ranging from 2 to 9 over the blank value. On the other hand, using the same drag-reducing polymers, Hanna *et al.* reported a decrease in pressure drop across the bed which ranged from 10 to 40% depending on particle size, polymer concentration and *Re*. Hanna *et al.* used a particle size range of 0.2373 to 0.3975 cm and a *Re* range of 1 to 1000. Laufer *et al.* [13] used a particle size of 0.1 cm and a *Re* range of 10 to 300. It is hoped that the present work may shed some light on this controversy.

2. Experimental technique

The apparatus (Fig. 1) consisted mainly of a 7-litre storage tank made of plexiglass, a centrifugal pump and the fixed-bed reactor. The reactor was made of a plexiglass tube with an internal diameter of 2.82 cm and a height of 8 cm. The tube was fitted with a porous plastic distributor at its base which contained 44 holes, each 1 mm in diameter. The reactor was closely packed with zinc pellets. Bed height was limited to 1.6 cm from the porous distributor, preliminary experiments having shown that beyond this height the mass transfer coefficient decreases significantly with bed height, especially at high flow rates. The pellets were of 99.9% purity (Merk), each pellet having a diameter of 0.318 cm. The bed porosity was 0.464.

Copper sulphate solution was circulated from the storage tank through the reactor by means

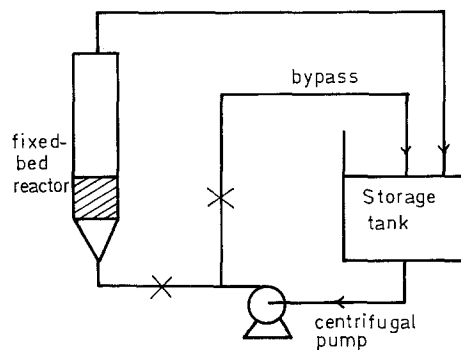


Fig. 1. Experimental apparatus.

of the pump. Flow rate was regulated using a bypass and was measured using a graduated cylinder and a stop-watch. The rate of the reaction was followed by withdrawing a sample of 5 cm³ of solution from the storage tank every 10 min and analysing it by iodometry [14]. The time of each experiment was limited to 90 min. Copper sulphate concentration ranged from 1 to 10 g l⁻¹ and polymer concentration ranged from 10 to 300 p.p.m. The polymer used was polyethylene oxide (Polyox WSR-301; Union Carbide). For each run a fresh solution and fresh zinc pellets were used. All experiments were carried out at pH 5.5 and a temperature of 30 ± 1°C. All polymer solutions were Newtonian. The diffusion coefficient of copper sulphate required to correlate the data was taken from the literature [15].

3. Results and discussion

Fig. 2 shows typical concentration–time curves for copper deposition at different flow rates. It is seen that depletion of Cu^{2+} becomes more rapid with higher circulation rates. For diffusion-controlled reactions taking place in a batch recirculating plug flow reactor, Walker and Wragg [16] derived the following approximate relation between concentration and time.

$$C = C_0 \exp \left\{ \frac{-t}{\tau} \left[1 - \exp \left(\frac{-KAaL}{Q} \right) \right] \right\} \quad (1)$$

The above equation was used to calculate the mass transfer coefficient in the present work. An

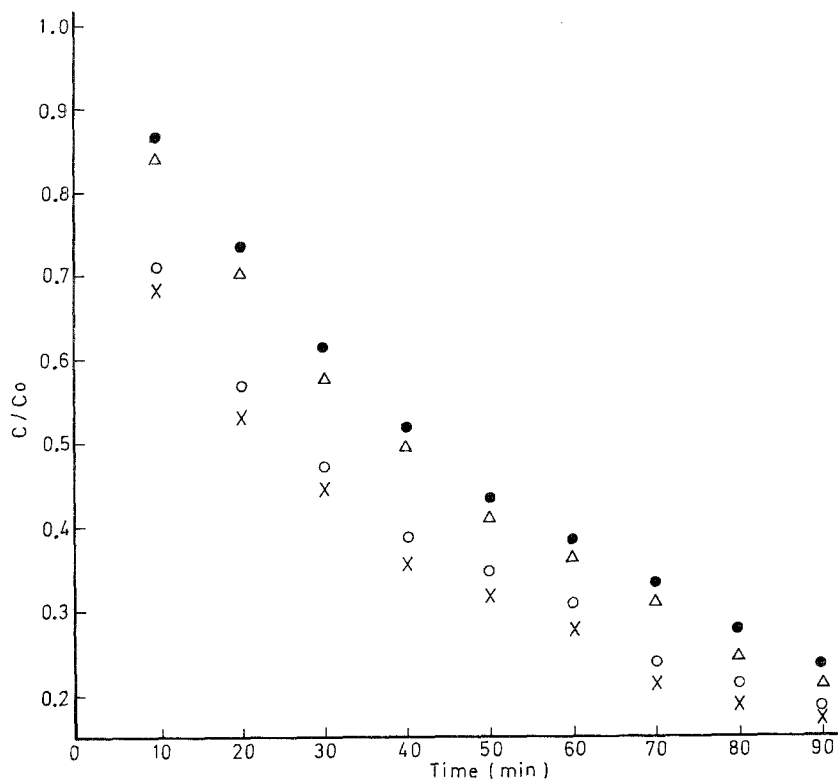


Fig. 2. Concentration-time plot for different solution flow rates: \times , 12.8 cm s^{-1} ; \circ , 11.2 cm s^{-1} ; Δ , 4.86 cm s^{-1} ; \bullet , 4.0 . Initial CuSO_4 concentration, 2.5 g l^{-1} .

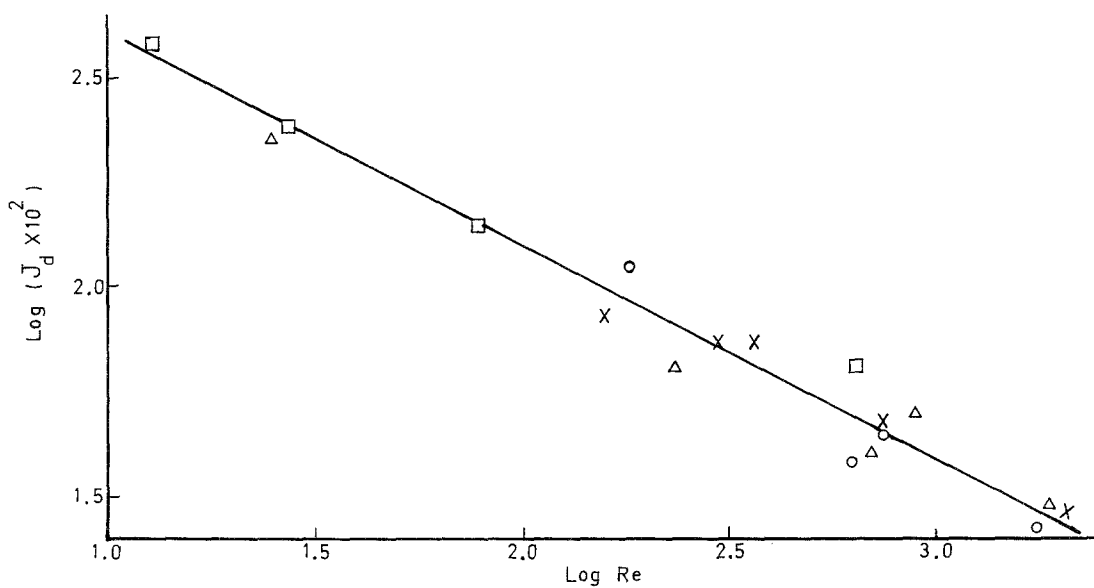


Fig. 3. $\text{Log } J_d$ versus $\text{log } Re$ for different CuSO_4 concentrations: \times , 1.0 g l^{-1} , $Sc = 1265$; \circ , 2.5 g l^{-1} , $Sc = 1346$; Δ , 5.0 g l^{-1} , $Sc = 1357$; \square , 10.0 g l^{-1} , $Sc = 1393$.

overall mass transfer equation was obtained for non-polyox-containing solutions (Fig. 3) using the dimensionless groups J_d and Re . The data can be represented for the conditions $10 < Re < 1970$ and $1265 < Sc < 1393$ by the equation

$$J = 12.55Re^{-0.5} \quad (2)$$

with an average deviation of $\pm 10.7\%$. The interstitial velocity (V_i) was used in calculating J_d and Re . A literature review shows that some studies have been carried out on mass transfer in fixed beds at high Re comparable to that used in the present work. Wilson and Geankoplis [17] covered the Re range of 0.0016 to 1500. For the Re range 0.0016 to 55 and the Sc range of 165 to 10690 they correlated their data by the equation

$$\varepsilon J_d = 1.09Re^{-0.66} \quad (3)$$

and for the Re range of 55 to 1500 by the equation

$$\varepsilon J_d = 0.25Re^{-0.31} \quad (4)$$

Upadhyay *et al.* [18] correlated their data in the range $10 < Re < 150$ and $767 < Sc < 42400$ by the equation

$$\varepsilon J_d = 0.455Re^{-0.4} \quad (5)$$

Granados *et al.* [19] correlated their electrochemical mass transfer data at fixed beds in the range $18 < Re < 325$ and $Sc = 1039$ by the equation

$$J_d = 1.875Re^{-0.55} \quad (6)$$

Wang *et al.* [20] found that the mass transfer coefficient of the deposition of copper on a fixed-bed cathode is related to Re by the equation

$$K \propto Re^{0.501} \quad (7)$$

for $20 < Re < 70$. Bravo de Nahui and Wragg [3], who studied the rate of copper cementation on a fixed bed of iron particles under the conditions $37 < Re < 550$ and $Sc = 1375$, correlated their data by the equation

$$J_d = 2.55Re^{-0.43} \quad (8)$$

Simonsson [6] correlated his mass transfer data for the electrodeposition of copper on a fixed-bed cathode for the conditions $10 < Re < 160$ by the equation

$$J_d = 1.46Re^{-0.28} \quad (9)$$

The present Re exponent (-0.5) agrees well with the value obtained by Wang *et al.* [20] and agrees fairly well with the values obtained by Bravo de Nahui and Wragg [3] and Granados *et al.* [19]. However, the present coefficient (12.55) is much higher than the value reported by other workers who used spherical particles. This may be attributed to the turbulence promoted by the randomly packed pellets and the increase in the interstitial velocity as a result of copper deposition on the pellets in addition to the fact that the true area of the rough pellets is higher than the approximate geometrical area used in calculating the mass transfer coefficient. The high mass transfer coefficients obtained in the present work are also consistent with the findings of Bravo de Nahui and Wragg [3], Kubo *et al.* [4] and Simonsson [6].

Fig. 4 shows that polymer addition decreases the mass transfer coefficient at $Re > 500$ by an amount ranging from 7.5 to 51% depending on Re and polymer concentration. The percentage reduction in the mass transfer coefficient increases with increasing Re probably because of the increase in the degree of stretching of the polymer molecules with a consequent increase in their effectiveness in damping small-scale, high-intensity eddies [21]. After passing through a maximum at $Re = 1400$ the percentage decrease in the mass transfer coefficient drops rapidly to a low value, probably because of severe polymer degradation caused by the high shear stress which prevails in the system at high Re . According to Kyle *et al.* [22], turbulent flow in fixed beds starts in the region extending from $Re = 150$ to 250 Hanna *et al.* [7], who studied the effect of polyox on pressure drop across a bed of spheres, found that the onset of drag reduction occurred at a Re between 150 and 250 depending on sphere diameter and polymer concentration. The delay in the onset of polymer effectiveness observed in the present work, as judged by the finding of Kyle *et al.* [22] and Hanna *et al.* [7], may be attributed to the difference in particle geometry which might affect the structure of the turbulence spectrum. The present work used pellets for the bed, while other workers [7, 20] used spherical particles. On comparing the present results with the results of Hanna *et al.* [7] it is found that on the whole the percentage

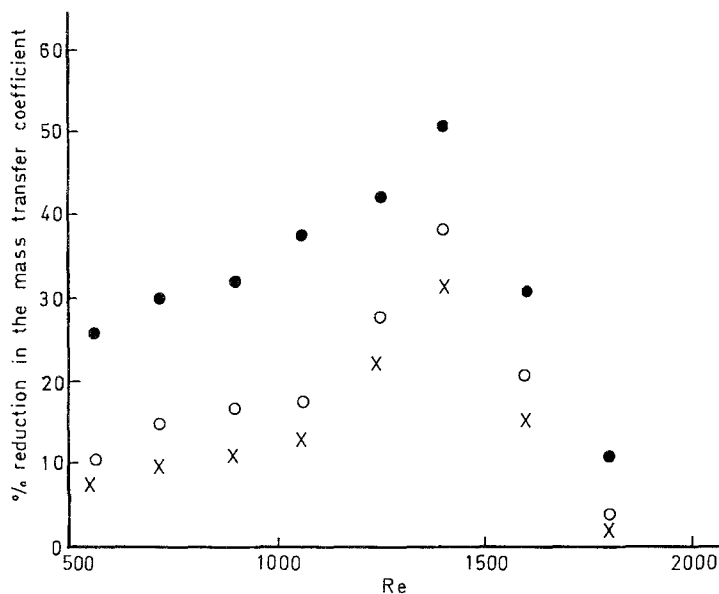


Fig. 4. Effect of Re on the percentage reduction in the mass transfer coefficient at different polyox concentrations (in p.p.m.): \times , 10; \circ , 150; \bullet , 300.

decrease in the rate of mass transfer caused by the drag-reducing polymer is comparable in magnitude to the percentage drag reduction. In view of this result it is difficult to recommend the use of drag-reducing polymers in operating fixed beds used for conducting diffusion-controlled reactions, since the decrease in the rate of production may outweigh the benefit of energy saving. However, drag-reducing polymers can be used with advantage if the reaction taking place in the fixed bed is a chemically controlled reaction. The present finding that the rate of mass transfer decreases by up to 51% in polymer solutions seems to support the previous finding that under turbulent flow conditions dilute polymers act as drag reducers where the pressure drop across the bed decreases by an amount ranging from 10 to 40% [7]. The present result is not consistent with previous findings [13] that in the presence of drag reducing polymers the pressure drop increases by a factor ranging from 2 to 9. The phenomenon observed by Laufer *et al.* [13] was explained on the basis of interaction of polymer elasticity with the porous structure of the bed [23], namely, that the randomly coiled elastic polymer molecules, while flowing in the porous bed, are being pulled into a stretched configuration faster than they can recoil and in so doing they are absorbing energy from the flow. This process accounts for the observed

increase in viscosity and pressure drop inside the bed. Much work remains to be carried out to reach a criterion by which the behaviour of the polymer in the bed can be predicted, i.e. whether it would act as a drag reducer or drag enhancer. A review of the conditions used by different workers shows roughly that polymer molecules tend to increase the pressure drop when the bed is made of small sized particles and Re is relatively low.

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