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Mass transfer to volumetric electrodes with two-phase flow

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Experimental studies of mass transfer were conducted in stacked screens with a gas–liquid mixture flowing through the bed. Depending on the gas and liquid flow rates and on the geometric characteristics of the screens, different flow regimes are obtained. In the heterogeneous flow regime the gas phase controls mass transfer, meanwhile in the transition and bubbling flow regimes the influence of the liquid flow prevails. Appropriate dimensionless groups correlate the mass transfer coefficients with the pertinent variables for the different regimes.

Nomenclature

A electrode area (cm^2)
A_1 surface area of one screen (cm^2)
c_0 bulk concentration ($mol\,cm^{-3}$)
D diffusivity ($cm^2\,s^{-1}$)
d particle or wire diameter (cm)
F Faraday's constant
i limiting current (A)
k mass transfer coefficient ($cm\,s^{-1}$)
N distance between wires (cm)
Re_g Reynolds number for gas flow, $Re_g = v_g R_h \nu_g^{-1}$
Re'_g Reynolds number for gas flow, $Re'_g = v_g d \nu_g^{-1}$
Re_l Reynolds number for liquid flow, $Re_l = v_l R_h \nu_l^{-1}$
Re'_l Reynolds number for liquid flow $Re'_l = v_l d \nu_l^{-1}$
R_h hydraulic radius of screen bed (cm)
Sc Schmidt number, $Sc = \nu_l D^{-1}$
Sh Sherwood number, $Sh = kdD^{-1}$
Sh_0 Sherwood number without gas, $Sh_0 = kdD^{-1}$
v_g superficial gas velocity ($cm\,s^{-1}$)
v_l superficial liquid velocity ($cm\,s^{-1}$)

δ screen thickness (cm)
ε porosity
ν kinematic viscosity ($cm^2\,s^{-1}$)
ϕ specific area (cm^{-1})

1. Introduction

There are many electrochemical processes of industrial importance which involve a multiphase system, particularly a gas–liquid system. Processes either with gas evolution at the electrodes or with gas introduction from the outside are present, for example, in water electrolysis, chlor-alkali electrolysis or in the electrochemical removal of chlorine and sulphur dioxide from effluent gases. In these processes the effect of gas stirring is very important, since the presence of gas bubbles in the liquid phase produces a substantial improvement of the mass transfer rate.

The study of mass transport to electrodes with gas evolution [1–8] or mass transport to wall electrodes with gas sparging [9–13] has been the subject of numerous investigations. However, little work has been done to study the effect of gas–liquid two-phase flow through three-dimensional electrodes. Delaunay *et al.* [14] and Barthole *et al.* [15], for instance, carried out electrochemical studies of liquid–solid mass transfer in packed beds of spheres with upward gas–liquid flow. Both related the mass transfer coefficients to the rate of energy dissipation in the liquid. Sedahmed [16] measured mass transfer rates at a H_2 evolving electrode, consisting of a packed bed of spheres. He found that the mass transfer coefficient depended on the discharge rate of hydrogen and to a lesser degree on the bed height and electrolyte concentration.

There are also several studies on mass transfer in fixed beds under two-phase flow which employed the electrochemical method. Mochizuki and Matsui

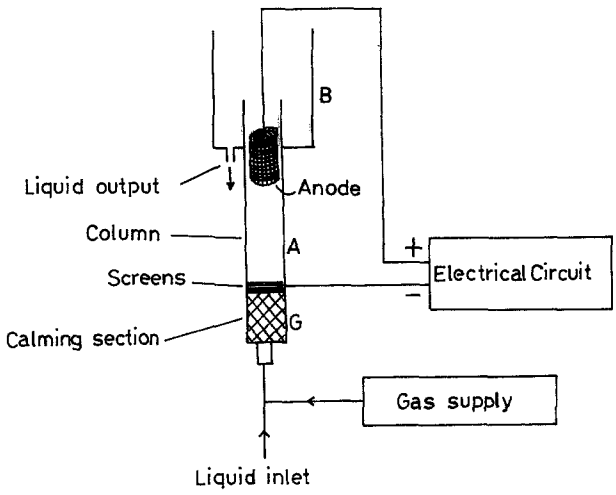

Fig. 1. Experimental set-up.

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Table 1.

Screen	d (*cm*)	A_1 (cm^2)	δ (*cm*)	N (*cm*)	R_h (*cm*)	ε	ϕ (cm^{-1})
A	0.0715	34.82	0.1628	0.246	0.08	0.82	10.07
C	0.080	26.57	0.1668	0.338	0.11	0.85	7.5
F	0.0725	32.77	0.1472	0.236	0.08	0.82	10.48
G	0.0996	20.01	0.1963	0.522	0.18	0.88	4.8
H	0.0898	24.07	0.1816	0.398	0.14	0.86	6.24

[17] and Colquhoun-Lee and Stepanek [18] measured mass transfer coefficients under the condition of co-current gas–liquid upward flow through the packed bed while others [19, 20, 21] worked with co-current downflow. All of these investigators used a single active particle placed in the packed bed of inert cylinders or spheres.

The use of volumetric electrodes combined with gas introduction from the outside or with gas evolution is of great interest since both the turbulence-promoting nature of the electrode and the bubble agitation enhance the mass transfer to the electrodes. The present work deals with the problem of mass transfer to a packed bed of screens with two-phase flow.

2. Experimental details

Mass transfer coefficients were determined from the limiting current for the electrochemical reduction of ferricyanide ions in alkaline solutions by applying the equation:

$$k = i/Fc_0A \qquad (1)$$

where A is the transferring surface of the packed bed, which can be calculated from the specific area of the screens and the volume of the packed bed.

The experimental set up is shown in Fig. 1. The test section was constructed from a lucite tube of 5.2 cm in internal diameter and 43 cm length. A calming section was located below the screen bed; this was a 10 cm high bed of 0.4 cm diameter glass spheres.

The solution was circulated by means of a centrifugal pump and was maintained at a temperature of 25° C by means of a cooling coil of stainless steel placed in the fluid reservoir.

The anode consisted of a roll of stainless steel screen mounted at the top of the cell; beds of six nickel-plated bronze screens were used as the working electrode. The parameters characterizing the geometry of the screens were calculated applying the method outlined by Blass [22]. These are summarized in Table 1 for all the electrodes tested.

The electrolyte solution was 10^{-3} M equimolar in potassium ferro and ferricyanide; a buffer 0.4 M equimolar $Na_2CO_3/NaCO_3H$ was used as indifferent electrolyte. At 25° C the density of the solution was 1.060 g cm^{-3}, its viscosity 1.167 centipoise and the diffusion coefficient of the ferricyanide ion was 5.59×10^{-6} $cm^2 s^{-1}$. Liquid velocities were varied between 0.1 and 5 cm s^{-1}.

Nitrogen was used as sparging gas; it was introduced through the calming section at velocities ranging between 0 and 6.5 cm s^{-1}. The gas was humidified before entering the column to avoid the evaporation of the electrolyte, principally at high gas flow rates.

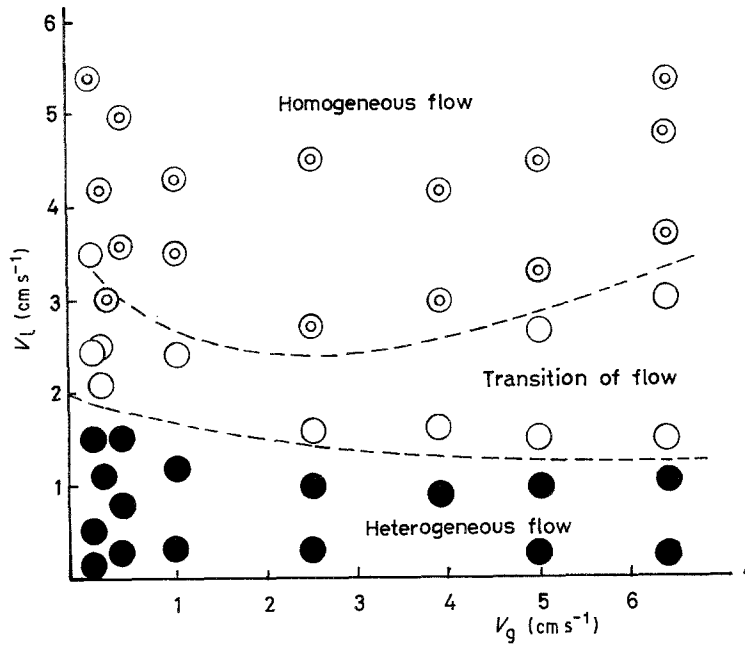

Fig. 2. Flow map for two-phase flow through screen beds.

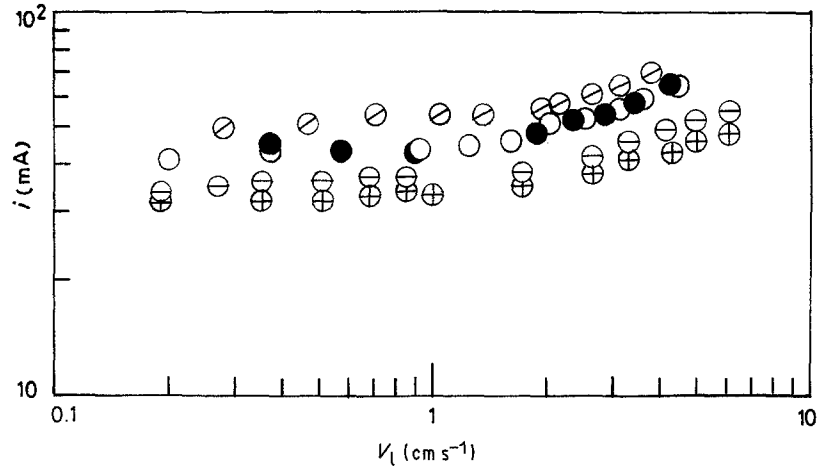

Fig. 3. Influence of liquid velocity on limiting current for different screen beds. V_g = 0.9 cm s^{-1}. ⊘, Screen A; ○, screen C; ●, screen F; ⊕, screen G; ⊖, screen H.

3. Results and discussion

Before considering the mass transfer process with bubble flow through the packed bed, the flow characteristics were analyzed by changing the liquid and/or gas velocities and the geometry of the bed. A visual observation was possible only in the region over the screens. In this zone three flow regimes could be distinguished. At low liquid velocities a heterogeneous flow regime was produced over the whole range of gas velocities; bubbles of variable size and very large bubbles could be observed. With increasing liquid velocity the large bubbles disappeared, the size of bubbles becoming more uniform. A transition of flow regime clearly occurred. Finally, at high liquid flow rates, a homogeneous flow regime was present with small bubbles of uniform size. Figure 2 presents the flow map for the system; it shows the range of liquid and gas velocities where each regime prevails.

Figure 3 illustrates the behavior encountered with the different screen beds when the liquid flow rate was varied, the gas velocity being held constant. As can be seen, the measured limiting current is not affected by an increasing liquid flow velocity until a value of $v_l \cong 1\,\mathrm{cm\,s^{-1}}$ is reached. The same behavior was observed for other gas flow rates.

Typical experimental data of mass transfer coefficients are shown in Fig. 4 as a function of the liquid velocity, the gas velocity being the parameter. It can be seen that for all gas velocities a change of slope occurs at liquid velocities between 1 and 2 cm s^{-1}, corresponding to the transition of flow regime mentioned before.

In order to relate the mass transfer coefficients to the liquid and gas velocities and to the geometric characteristics of the screens in a dimensionless form, Sh, Re_l and Re_g were defined. For the Reynolds numbers the hydraulic radius of the packed bed was taken as characteristic length, meanwhile for the Sherwood number the wire diameter was introduced.

Figure 5 shows some results obtained at low liquid flow rates (heterogeneous flow regime). When comparing with the Sherwood number obtained in the absence of gas bubbling, it can be seen that mass transfer enhancement due to gas stirring is produced at Re_g greater than 0.1.

In the heterogeneous regime of two-phase flow Sherwood numbers more than three times those for single-phase flow can be obtained owing to the presence of gas bubbles.

In the case of liquid flow alone through packed beds of screens the following correlation, deduced in a

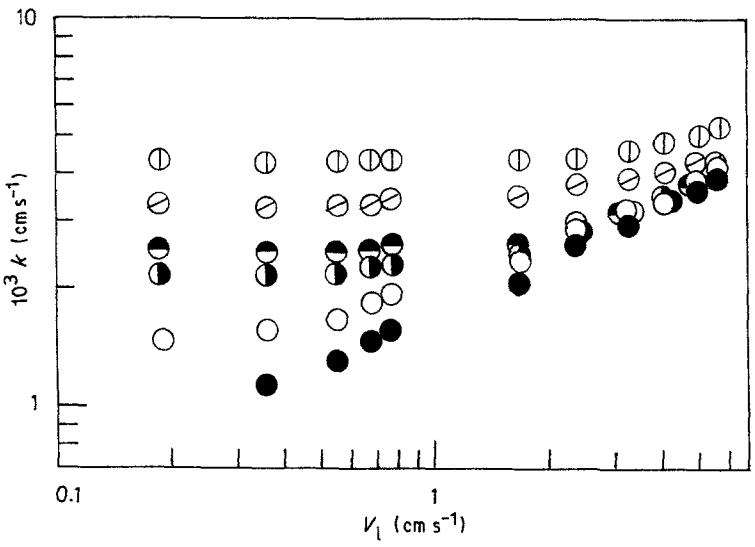

Fig. 4. Mass transfer coefficients for screen G as function of liquid and gas velocities. V_g (cm s^{-1}) = 0, ●; 0.075, ○; 0.329, ◑; 0.973, ◓; 2.410, ⊘; 6.501, ⊙.

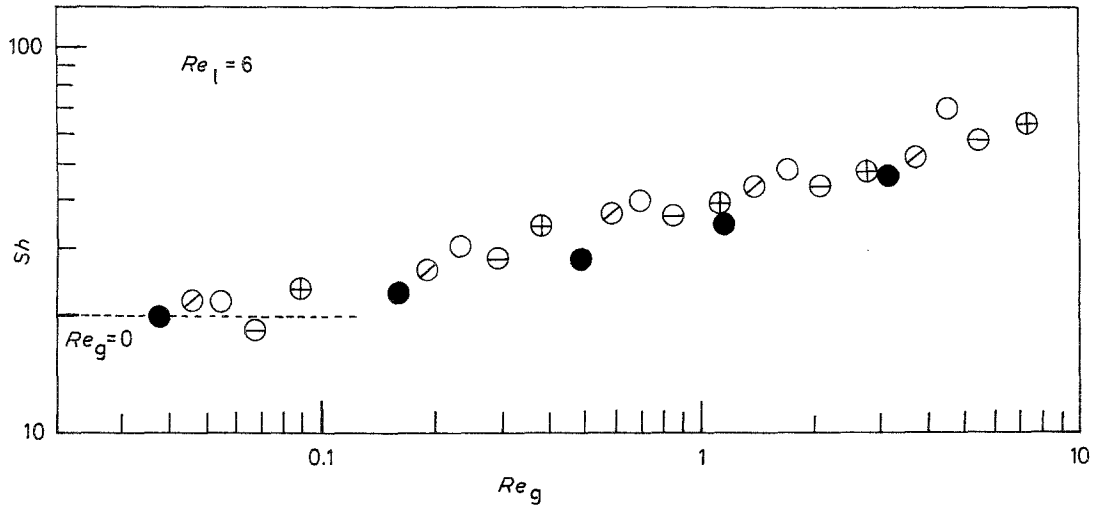

Fig. 5. Sherwood number plotted against gaseous Reynolds number for different screen beds. ⊘, screen A; ○, screen C; ●, screen F; ⊕, screen G; ⊖, screen H.

previous investigation [23], holds:

$$Sh_0 = 0.908 Sc^{0.33} Re_l'^{0.34} \quad (2)$$

where Re_l' is based on the wire diameter as characteristic length.

In the case of gas–liquid flow through the packing, the flow regime has to be taken into account, the transition from one regime to the other lying at a liquid velocity of about 1 cm s^{-1}.

For the heterogeneous regime, that is for $0.1 < v_l < 1$ cm s^{-1}, the following correlation was obtained using a non-linear multiple regression program:

$$Sh = 2.85 Sc^{0.33} Re_l^{0.125} Re_g^{0.238} \quad (3)$$

The standard deviation is 10.8%.

For the homogeneous regime, the regression analysis applied to all the data corresponding to $v_l > 1$ cm s^{-1} yielded

$$Sh = 1.70 Sc^{0.33} Re_l^{0.275} Re_g^{0.123} \quad (4)$$

with a standard deviation of 10.7%.

These two correlations clearly show the influence of each phase on mass transfer. At low liquid flow rates the effect of gas bubbles is important; the bubbles reduce the flow area available for the liquid thus increasing the interstitial liquid velocity, act as turbulence promoters and disturb the diffusion boundary layer. At high liquid velocities the effect of gas bubbles decreases and the turbulence generated in the liquid phase itself affects mass transfer significantly.

When comparing Equations 3 and 4 with Equation 2 it can be easily deduced that within the range of operating variables of this investigation, a greater improvement due to gas sparging is obtained with the heterogeneous flow regime. As mentioned before the increase achieved in this regime may exceed 200%, while with the homogeneous flow regime the improvements at most reach 100%. In Fig. 6 the variation of the ratio Sh/Sh_0 with Re_l' is plotted for $Re_g' = 5$. Also shown are the results of previous studies [14, 15, 17, 18] working with upward flow. The investigations using downflow are not included for comparison since the hydrodynamic flow conditions and operating range of flow velocities are much different. There are no data available to compare for the heterogeneous

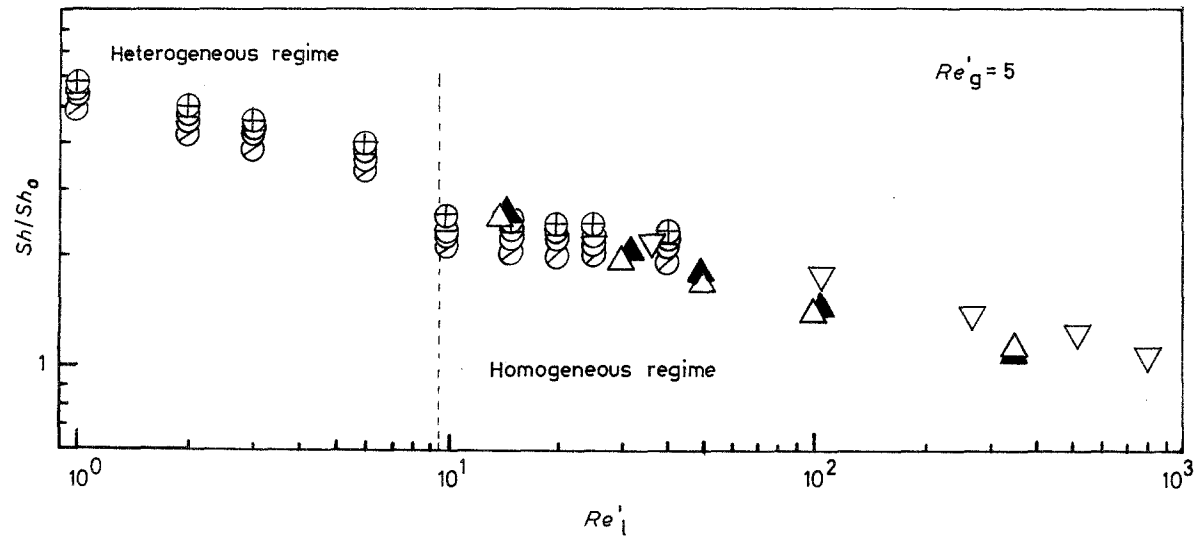

Fig. 6. Mass transfer enhancement by gas bubbling. Comparison with [17], △; [14] and [15], ▲; [18], ▽; this work, ○.

flow regime, but for the homogeneous flow conditions a very satisfactory agreement is found with other work. The enhancement produced by the gas bubbles is seen to be the same for packings of screens, cylinders or spheres.

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