JOURNAL OF APPLIED ELECTROCHEMISTRY 9 (1979) 361–367

Local study of wall to liquid mass transfer in fluidized and packed beds. II. Mass transfer in packed beds

D. HUTIN, A. STORCK, F. COEURET*

Laboratoire des Sciences du Génie Chimique, CNRS–ENSIC, 1, rue Grandville, 54 042 Nancy, France

Received 26 May 1978

Mass and momentum transfer at a wall in liquid–particle systems are studied with a two-dimensional model which consists of fixed spherical turbulence promoters arranged in a simple cubic lattice in a rectangular channel. Local values of the mass transfer coefficient and shear stress at a wall of the channel have been measured at identical locations. The results show that there are large differences between the local values but their distribution along the transfer surface is reproduced identically. The dependence of these local values on each other allows one to obtain a general relationship between overall mass and momentum transfer as well as a correlation of mass transfer results for exchange between a wall and a flowing liquid in a fixed bed of particles.

Nomenclature

a_g	particle specific area
a	coefficient in expression $s = a\beta^q$ $(q > 0)$
a', b	coefficients in expression $j_M = a'(Re)'^{-b}$
d_p	particle diameter
d	microelectrode diameter
D	molecular diffusion coefficient
h_K, h_B	constants in Ergun equation
$j_M = (\bar{k}/u/\epsilon)(Sc)^{2/3}$	Colburn j-factor
k	local mass transfer coefficient
k'	local mass transfer coefficient in inert wall
$\bar{k}$	overall mass transfer coefficient
L	length of the transfer surface
q	exponent in expression $s = a\beta^q$
$(Re)' = (u\,d_p)/[\nu(1-\epsilon)]$	modified Reynolds particle number
$(Sc) = \nu/D$	Schmidt number
$s, \bar{s}$	velocity gradients at the wall
u	superficial liquid velocity
α	coefficient in Equation 1
β	characteristic length
ϵ	bed porosity
ρ_F	fluid density
η	dynamic viscosity
ν	kinematic viscosity
$\tau, \bar{\tau} = \eta s$	shear stress at the wall
$\Delta P/L$	fluid pressure gradient

1. Introduction

It is well known that overall mass transfer rates between a flowing liquid and a wall can be increased by the presence of fixed turbulence promoters such as, for example, spherical particles or porous plastic cloths placed near the transferring surface (membranes in electrodialysis or electrodes in electrolytic cells).

A few investigations [1–4] of wall to liquid mass transfer in packed beds of inert particles have been made and the overall effect of different hydrodynamic and geometrical parameters can be summarized as follows:

(a) the length L of the transferring wall has no measurable influence on the mass transfer rates,

(b) the overall mass transfer coefficient $\bar{k}$ increases with decreasing particle diameter d_p,

(c) $\bar{k}$ increases with the superficial liquid velocity u,

* Present address: L.E.A.T., Laboratoire d'Etudes Aérodynamiques et Thermiques, 40, Avenue du Récteur Pineau, 86022 Poitiers, France.

 © 1979 *Chapman and Hall Ltd.*

(d) mass transfer rates in a packed bed are always higher than in a fluidized bed, the other parameters being unchanged.

Most of these investigations were concerned with overall phenomena (i.e. average coefficients). But recently mass and momentum transfer have been measured locally [5] on an assembly using cylindrical turbulence promoters located near a transferring wall. The corresponding parametric study has shown that large deviations exist between the local mass transfer rates, which are maximal just below the cylinders where the local fluid acceleration is the largest.

The advantage of fixed compared to fluidized promoters appeared when it was shown that the mass transfer coefficients were higher in packed beds [4] and some practical processes using fixed promoters (plastic cloth in the 'Swiss Roll Cell' [6] or in electrodialysis cells [7]) are now already available.

The present paper reports an investigation of the local effect of spherical obstacles on the mass transfer coefficient and the fluid shear stress at a wall, both being measured at the same point. The study is carried out on a two-dimensional system and one of its aims is to allow an interpretation of mass transfer between a flowing liquid and a wall immersed in a fixed bed of grains.

The relationship between overall mass transfer coefficients and the liquid pressure gradient in a packed bed is also investigated and deduced from local experimental results.

2. Experimental

The equipment has been described previously [5]. The parallelepipedic cell (dimensions 100 x 50 x 10 mm) encloses 50 spherical particles (diameter d_p = 10 mm = channel thickness) arranged in a simple cubic lattice. The liquid flow in such a channel simulates the flow near a wall immersed in a packed bed of grains.

The principle of electrical measurements is described in part I of this work. Given the symmetry axis of the porous system, the study of the experimental variations of k and k' can be restricted to an elementary rectangular pattern with 5 x 10 mm dimensions.

The cover of the cell includes (Fig. 1): a rectangular 30 x 40 mm platinum plated copper electrode, and three rows (noted a, b and c) of six microelectrodes distributed regularly in the 5 x 10 mm elementary pattern A.

It includes also a second pattern B of 18 microelectrodes in an inert wall.

The viscosities of potassium ferricyanide and potassium ferrocyanide solutions in 0·5 M sodium hydroxide were varied by adding solid commercial polyethyleneglycol (Emkapol 6000).

3. Experimental results

3.1. *Existence of an elementary pattern*

For a given fluid velocity, Fig. 2 presents the experimental variation of k' in the two elementary patterns A and B located 30 mm from each other (i.e. three particle·diameters).

Within experimental precision due to the difficulty of determining the exact position of microelectrodes with respect to spheres, agreement between the values of k' in the two patterns is good.

This result allows the existence of an elementary pattern in the momentum transfer mechanism to be deduced, this pattern being reproduced nearly identically along the transfer surface. Therefore, the measurements of k and k' will be restricted to the pattern A.

3.2. *Variations of* k *and* k' *with the position between particles*

Figs. 3 and 4 are given as examples of the variations of k and k' with the position between consecutive particles.

From these figures and other results not presented here, the following remarks can be made. Large deviations exist between the local values of the mass transfer coefficients depending on the position with respect to the spheres. For example, the mass transfer rates are very small just under a spherical grain and much higher in the vicinity of the contact points between grains and wall.

In the same row (a, b or c) the variations of k and k' correspond to curves with a maximum or minimum. (a) k and k' present maximum values between two consecutive contact points of the particles with the wall (row a). (b) For all other cases (rows b and c), the experimental curves present minimum values.

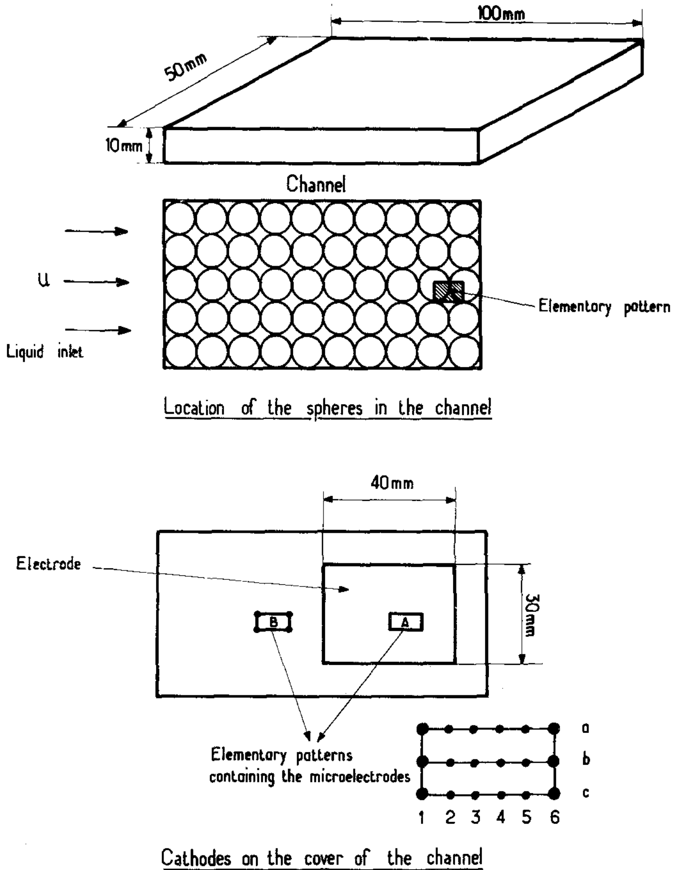

Fig. 1. Schematic view of the cathodes and spheres in the channel.

The results show a qualitative analogy between the variations of k and k' representative to mass and momentum transfer respectively. The question is now whether it is possible to find a relation between these two coefficients.

3.3. *Relationship between* k *and* k'

In a previous study relative to mass transfer in the presence of cylindrical promoters [5], it has been shown that the local relationship between k and the fluid shear stress τ is expressed as

$$k = C\frac{D^{2/3}}{\eta^{1/3}}\left(\frac{\tau}{\beta}\right)^{1/3} = CD^{2/3}\left(\frac{s}{\beta}\right)^{1/3} \qquad (1)$$

where β is a characteristic length depending on the free cross-sectional area for liquid flow between the cylinders and the wall, and C a constant.

For a given position between the grains and two different values of the liquid Schmidt number, Fig. 5 presents the separate variations of k and k' with the liquid superficial velocity u. k and k' increase with increasing u and vary similarly in the range of flow rates considered.

These results and others not presented here can be used in the following manner for investigating the relation between k and τ.

For a given microelectrode, the values of k and k' corresponding to each liquid velocity are presented graphically (Fig. 6).

The following two conclusions can be drawn: k and k' are proportional to one another for any position between the spheres, and for a given microelectrode, the relationship between k and k' does not depend on the Schmidt number.

A simple mathematical development analogous to the one presented in [5] allows Equation 1 to be obtained and confirms its validity.

For cylindrical promoters, β is the distance between the microelectrode and the imaginary leading edge of the diffusional boundary layer. For spherical particles, the geometrical complexity

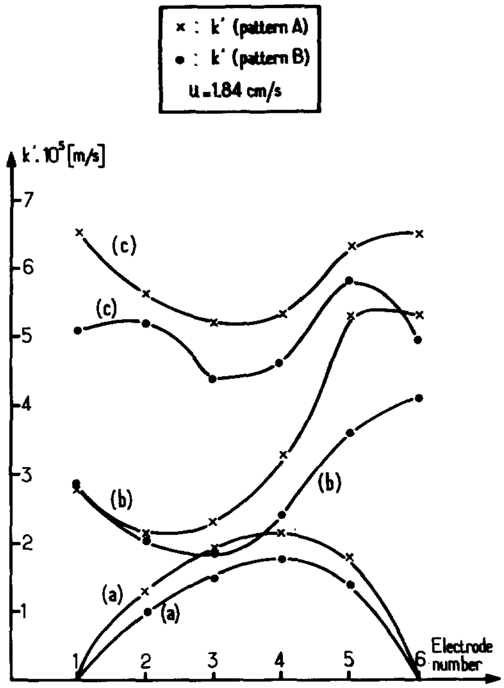

Fig. 2. Comparison of coefficients k' in the two patterns A and B for $u = 1{\cdot}84\ \mathrm{cm\ s^{-1}}$.

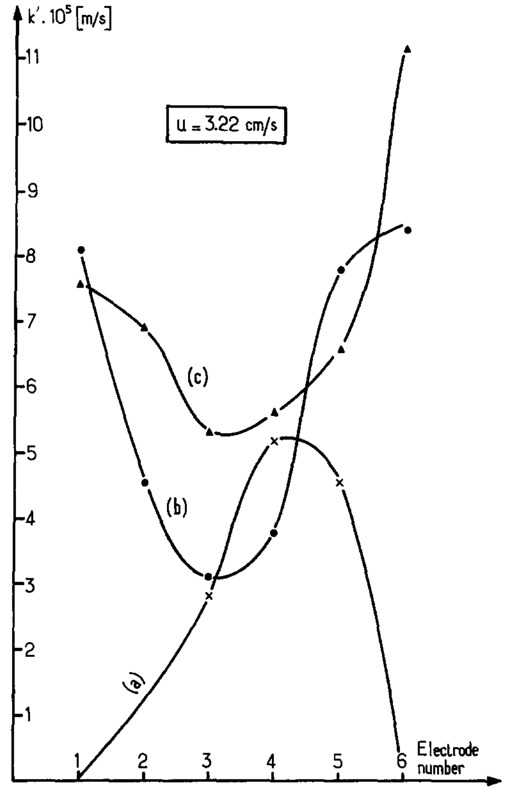

Fig. 4. Experimental variations of k' with the position between particles ($u = 3{\cdot}22\ \mathrm{cm\ s^{-1}}$).

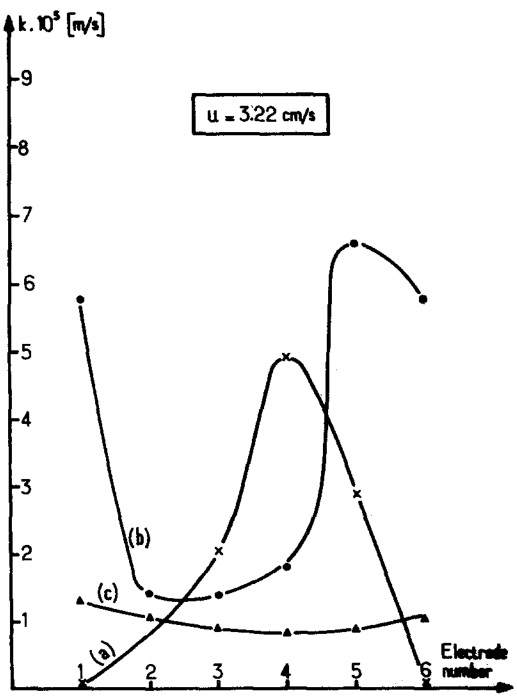

Fig. 3. Experimental variations of k with the position between particles ($u = 3{\cdot}22\ \mathrm{cm\ s^{-1}}$).

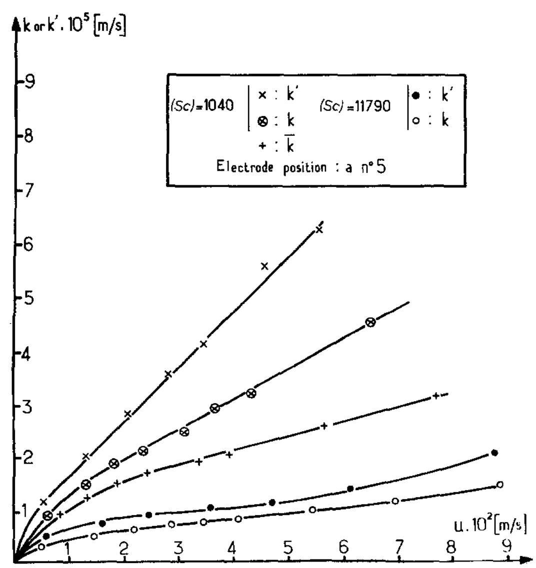

Fig. 5. Variations of k and k' with the liquid velocity u for different positions between particles and fluid Schmidt numbers.

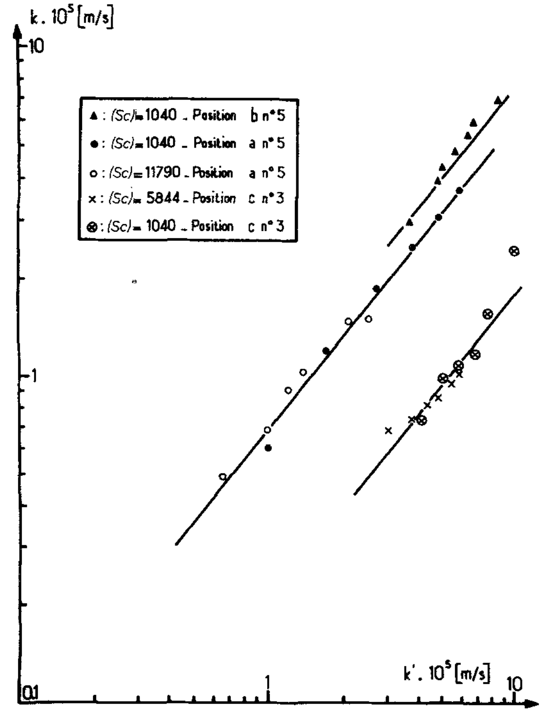

Fig. 6. Relation between k and k' at different positions between the particles.

of the cell packed with spheres does not allow the exact signification of β to be known: β is a characteristic length depending on the free cross-sectional area for liquid flow.

4. Application to overall mass and momentum transfer at a wall in a packed bed

Let us assume that Equation 1 remains still valid for any geometry and disposition of the promoters near a wall and that in a small range of variations, a law of the type

$$s = a\beta^q \quad (q > 0)$$

may be written for the local velocity gradient s. Thus

$$k = CD^{2/3}\left(\frac{s}{\beta}\right)^{1/3} = CD^{2/3}a^{1/3q}s^{(q-1)/3q}$$

From experimental results obtained in this study, we also suppose that for a packed bed of spheres (porosity $\epsilon \simeq 0{\cdot}4$) the values of s are uniformly distributed between two limits 0 and s_1. The probability density function $f(s)$ of s may be therefore expressed as

$$f(s) = \frac{1}{s_1} \quad \text{for } 0 \leqslant s \leqslant s_1$$

$$f(s) = 0 \quad \text{for } s > s_1$$

$g(k)$ denoting the probability density function of $k\,[g(k)\,\mathrm{d}k = f(s)\,\mathrm{d}s]$, the spatial mean value $\bar{k}$ of k may be calculated as follows:

$$\bar{k} = \int_0^{k_1} kg(k)\,\mathrm{d}k$$

$$= \int_0^{s_1} CD^{2/3}a^{1/3q}s^{(q-1)/3q}\,\frac{1}{s_1}\,\mathrm{d}s$$

where k_1 is the k value corresponding to s_1.

Integrating this expression leads to

$$\bar{k} = CD^{2/3}a^{1/3q}\,\frac{3q}{4q-1}\,s_1^{(q-1)/3q}$$

or given that $s_1 = a\beta_1^q$

$$\bar{k} = CD^{2/3}\,\frac{3q}{4q-1}\,\frac{s_1^{1/3}}{\beta_1^{1/3}} \qquad (2)$$

The characteristic length β_1 depends on the particle diameter d_p. As a first approximation β_1 can be expressed as

$$\beta_1 = \alpha d_p \qquad (3)$$

and the mean value $\bar{s}$ of s at the wall is given by

$$\bar{s} = \frac{s_1}{2} \qquad (4)$$

Furthermore, the mean shear stress $\bar{\tau} = \eta\bar{s}$ is related to the liquid pressure gradient $\Delta P/L$ along the wall by the following expression [8]:

$$\bar{\tau} = \mathrm{d}p\,f(\epsilon)\,\Delta P/L \qquad (5)$$

$f(\epsilon)$ being a function of the bed porosity ϵ.

Combining Equations 2–5 leads to the overall mass transfer coefficient $\bar{k}$

$$\bar{k} = C\frac{D^{2/3}}{\eta^{1/3}}\,\frac{3q}{4q-1}\,\frac{1}{\alpha^{1/3}}\,2^{1/3}\,f(\epsilon)^{1/3}\left(\frac{\Delta P}{L}\right)^{1/3}$$

or

$$\bar{k} = Cste\,\frac{D^{2/3}}{\eta^{1/3}}\left(\frac{\Delta P}{L}\right)^{1/3} \qquad (6)$$

An experimental relationship analogous to Equation 6 between $\bar{k}$ and $\Delta P/L$ has been established [9] for mass transfer in a classical fixed bed of inert particles. The constant was found to be 0·2 and independent

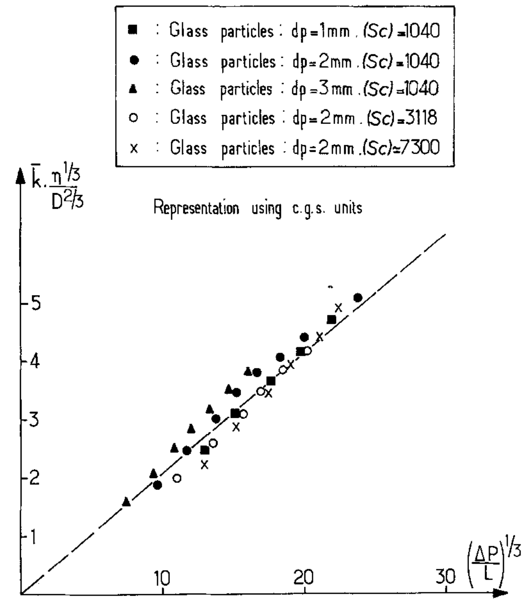

Fig. 7. Experimental variations of $\bar{k}\,\eta^{1/3}/D^{2/3}$ with $(\Delta P/L)^{1/3}$ for fixed beds of spheres.

of particle diameter. The corresponding results are shown in Fig. 7 and confirm the validity of the preceding model.

Moreover, for mass transfer between a flowing liquid and a wall immersed in a packed bed, the overall results have been frequently correlated by empirical relations [4–10] of the form

$$j_M = a'(Re)'^{-b}$$

where j_M is the Chilton–Colburn factor for mass transfer $[j_M = (\bar{k}/u/\epsilon)(Sc)^{2/3}]$ and $(Re)'$ a modified Reynolds number $\{(Re)' = u\,dp/[\nu(1-\epsilon)]\}$.

For packed beds, the liquid pressure gradient $\Delta P/L$ is given by the Ergun equation [11] which may be applied here because $\Delta P/L$ is the same along the wall and across the bed

$$\frac{\Delta P}{L} = h_K \eta a_g^2 \frac{(1-\epsilon)^2}{\epsilon^3} u + h_B \rho_F a_g \frac{(1-\epsilon)}{\epsilon^3} u^2 \quad (7)$$

where h_K and h_B are two constants equal to 4·5 and 0·3 respectively, for spherical particles.

A combination of Equations 6 and 7 allows the two following correlations to be derived:

for the laminar flow regime $[(Re)' < 16]$:

$$j_M = Cste\,(36 h_K)^{1/3}\,(Re)'^{-2/3} = 1{\cdot}09\,(Re)'^{-2/3}$$

for the turbulent flow regime $[(Re)' > 1600]$:

$$j_M = Cste\,(6 h_B)^{1/3}\,(Re)'^{-1/3} = 0{\cdot}24\,(Re)'^{-1/3}$$

Fig. 8 shows the comparison of these two correlations with experimental correlations of mass transfer in packed beds [1–3]. As can be seen, the agreement is satisfactory.

Consequently the investigation of the local effect of spheres combined with the model presented before lead to an interpretation of overall wall to liquid mass transfer in a fixed bed of grains and permit correlations of the transfer results to be calculated. The model includes several unknown parameters (q and α for example) but the agreement with experimental results is satisfactory enough to prove the validity of the relation obtained.

5. Conclusions

This experimental study dealing with the effect of spherical particles disposed in a cubic lattice on local mass transfer rates allows confirmation of the validity of experimental results obtained previously with cylindrical promoters, for example, the existence of an elementary pattern in the transfer mechanism, this pattern being reproduced nearly identically along the wall, and large variations in the local mass transfer coefficients depending on the position between the particles.

The uniformity of the transfer rates is, therefore, not realized but the existence of an elementary pattern results in a pseudo-uniformity of the rates along the wall. The local measurements have shown that the spatial distributions of mass transfer coefficients and shear stresses are similar and that a simple relationship between them could be obtained. This relation, identical to the one valid for cylinders [5], allows the overall mass transfer coefficient in a packed bed from fluid pressure gradient to be obtained using a mathematical model. It also permits correlations of mass transfer results in packed beds to be deduced.

In a study concerning the turbulent diffusion on a rough rotating disc, Meklati and Daguenet [12, 13] have shown that the limiting diffusional flux on the rough surface was proportional to the power 2/3 of the angular velocity of the disc. The corresponding mass transfer coefficient k is expressed as

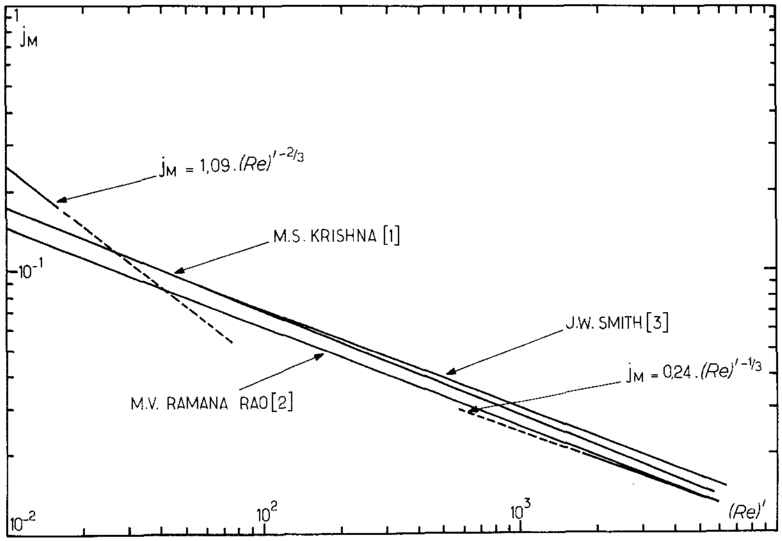

Fig. 8. Variations of the Chilton–Colburn factor j_M with $(Re)'$ for packed beds of spherical particles.

$$k \sim \frac{D^{2/3}}{\eta^{1/3}}\left(\frac{\tau}{l}\right)^{1/3}$$

where l is a characteristic length depending on the roughness dimension. This relation is formally identical to the one (Equation 1) obtained in this work even though the two studies are different. Indeed one deals with turbulence promoters arranged near a smooth transfer surface, whereas the other deals with mass transfer to a rough surface. It seems that such a relation between k and the fluid shear stress τ has a general meaning and can be applied whenever a liquid and a solid boundary exchange mass in a non-established diffusion regime.

References

[1] M. S. Krishna, G. J. V. Jagannadharaju and C. Venkata Rao, *Period. Poly. Chem. Engr. (Budapest)* 11 (1967) 95.

[2] M. V. Ramana Rao and C. Venkata Rao, *Indian J. Technol.* 8 (1970) 44.

[3] J. W. Smith and D. H. King, *Canad. J. Chem. Engng.* 53 (1975) 41.

[4] A. Storck, F. Vergnes and P. Le Goff, *Powder Technol.* 12 (1975) 215.

[5] A. Storck and F. Coeuret, *Electrochim. Acta* 22 (1977) 1155.

[6] P. M. Robertson, F. Schwager and N. Ibl, *J. Electroanalyt. Chem.* 65 (1975) 883.

[7] Y. Winograd, A. Solan and M. Toren, *Desalination* 13 (1973) 171.

[8] A. Storck and F. Coeuret, Communication to the European Congress *Transfer Processes in Particle Systems* Nuremberg (1977).

[9] A. Storck and F. Coeuret, *Canad. J. Chem. Engng.* 55 (1977) 427.

[10] F. Coeuret and P. Le Goff, *Electrochim. Acta* 21 (1976) 195.

[11] S. Ergun, *Chem. Eng. Prog.* 48 (1952) 89.

[12] M. H. Meklati and M. Daguenet, *C.R. Acad. Sci. Paris* 272C (1971) 2027.

[13] M. H. Meklati and M. Daguenet, *J. Chim. Phys.* 7 (1973) 1102.