JOURNAL OF APPLIED ELECTROCHEMISTRY 11 (1981) 727–733

Mass transfer in agitated vessels: energetic aspects

A. STORCK, J. F. BRODBERGER, D. HUTIN, G. VALENTIN

Laboratoire des Sciences du Génie Chemique, CNRS-ENSIC, 1 rue Grandville, 54042 Nancy Cedex, France

Received 2 February 1981

This paper deals with the experimental determination of the mass transfer rates between the liquid of an agitated vessel and a spherical particle immersed in a reactor. The spatial distribution of the mass transfer coefficients is obtained using an electrochemical method and the influence of the most pertinent hydrodynamic parameters (impeller speed and fluid residence time inside the vessel) is deduced from experimental results. The study considers the two limiting cases of mechanical agitation alone and agitation induced by the liquid jets generated by the feed nozzles. It is shown that knowledge of the specific power dissipated per unit mass of fluid can be useful for the theoretical prediction of the mass transfer rates.

Nomenclature

D	molecular diffusion coefficient
d_p	particle diameter
D_i	impeller diameter
D_v	vessel diameter
E	hydrodynamic parameter defined in Equation 8
H	vessel height
k	mass transfer coefficient
l	characteristic length in Equation 2
M	mass of liquid in the vessel
N	rotational speed of agitator
N_p	specific power number
P	specific power delivered
Q_v	volumetric flow rate of the feed fluid
S	area for fluid injection
(Sc)	$= \nu/D$ = liquid Schmidt number
(Sh)	$= kd_p/D$ or kl/D = Sherwood number
U	local fluid velocity
V	vessel volume
x	vertical distance between the bottom of vessel and the measuring point
α	coefficient in Equation 2
ν	kinematic viscosity
ρ	fluid density
τ	$= V/Q_v$ = residence time in vessel

1. Introduction

In chemical processing, agitated vessels are often used to carry out steps in which interphase mass transfer controls the overall rate (e.g., in dissolution, crystallization, solid–liquid extraction and, more recently, fermentation). A survey of scientific literature dealing with this general problem shows that extensive studies have been carried out in the fields of mass transfer from suspended solids to the liquid of the vessel (see for example [1–4]) and mass transfer at the walls of the vessel [5–8], but only a few studies are related [9–11] to the case of a fixed particle immersed in an agitated vessel.

The local study of mass transfer from fixed solute surfaces in a fully 'baffled' tank was first proposed by Miller [9] who measured the dissolution rates of benzoic acid particles. The experimental results were used to verify a theoretical model of velocity distribution inside the reactor. It seems, however, important to note the scatter (± 40%) observed in the results. From experimental results in the literature, Calderbank and Moo Young [12] have deduced a general energetic correlation applying to heat and mass transfer in mixing vessels where the solid phase is in the form of a single, fixed, submerged body. They predicted an increase in the mass transfer coefficients as the specific power dissipation level rises.

More recently, Le Lan *et al.* [10, 11] have applied an electrochemical measuring method to the study of local mass transfer to a fixed particle

0021–891X/81/060727–07$02.70/0

in three similar cylindrical vessels with or without baffles. The authors have established that the stirring regime induced by a turbine is virtually independent of the dimensions of the vessel and that the peripheral speed of the blade is a good criterium for extrapolation. For a baffled tank [11], it was also shown that the fluid velocity at a given position in the vessel was proportional to the blade speed. This result was verified by other authors [13–15] from experimental measurements of velocity distributions.

For processes operated in continuous, stirred-tank reactors, the agitation level inside the vessel is determined by two contributions: (a) that due to the impeller rotation, (b) the agitation induced by the kinetic energy of the liquid feeding the reactor. In processing, where the fluid residence time inside the vessel has to be small, contribution (b) can be significant and can considerably modify the results obtained in a classical tank. To our knowledge this is the first work to consider this configuration from a mass transfer point of view.

The main aims of this paper are:

(a) to determine experimentally the distribution of mass transfer coefficients inside a vessel agitated simultaneously by an impeller and by the kinetic energy of the feed fluids;

(b) to determine the quantitative influence of the hydrodynamic parameters (impeller speed and fluid residence time);

(c) to predict the mass transfer rates from a knowledge of the specific power dissipated in the reactor and to deduce energetic correlations.

2. Experimental

The design characteristics of the 0.78 dm^3 tank used in this study are shown in Fig. 1 and correspond closely to the standard configuration proposed by Holland and Chapman [16]. The tank was provided with a six-blade, flat-paddle agitator and the agitator speed range in the tank was 0–2000 r.p.m. The liquid is continuously fed to the reactor from two injection nozzles located at the bottom of the tank, as indicated in Fig. 1. The fluid residence time τ (defined as the ratio of the reactor volume to the volumetric flowrate Q_v) was varied between 5.7 s and infinity (corresponding to $Q_v = 0$). The mass transfer rates are measured on a spherical nickel probe (diameter 4 mm)

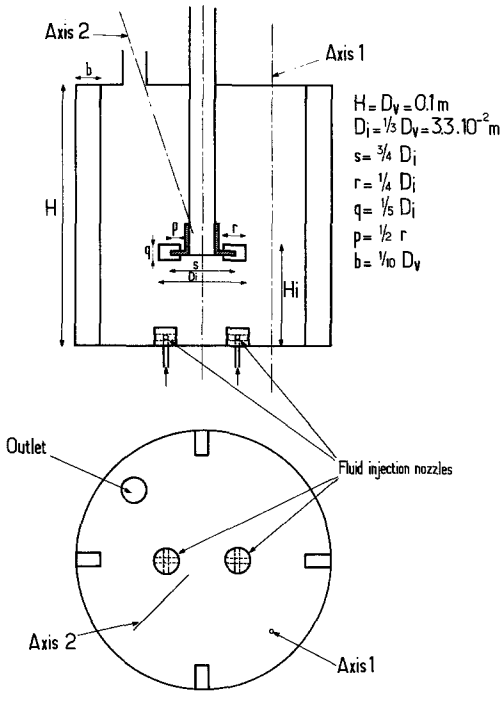

Fig. 1. Schematic view of the agitated vessel.

located at the end of a small-diameter glass rod in order to prevent any hydrodynamic perturbation by the probe.

The spatial distribution of mass transfer rates was determined in two directions, corresponding to axes 1 or 2 in Fig. 1. As in previous studies [17–19], the overall mass transfer coefficients between the spherical particle and the liquid were obtained electrochemically by means of the cathodic reduction of ferricyanide ions. The liquid (electrolyte) is a mixture of potassium ferricyanide (10^{-3} M) and ferrocyanide (5×10^{-2} M) in a supporting electrolyte of NaOH (0.5 N). Experimental values obtained at a constant temperature (30° C) for the electrolyte kinematic viscosity ν, the ferricyanide diffusion coefficient D and the Schmidt number (Sc) are respectively 0.914×10^{-6} m^2 s^{-1}, 8.8×10^{-10} m^2 s^{-1} and 1040.

3. Experimental results

As an example, Fig. 2 presents the experimental distribution of k for a given impeller speed N and fluid residence time τ. This figure clearly shows the existence of three zones:

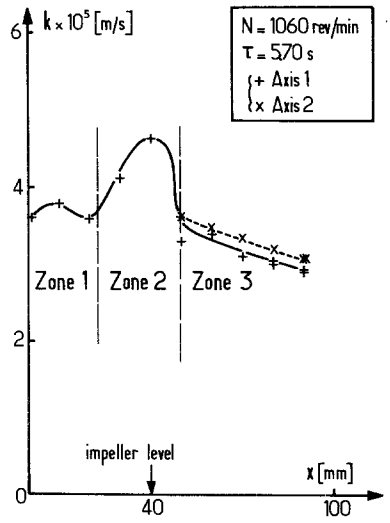

Fig. 2. Experimental distribution of the mass transfer coefficient inside the vessel ($N = 1060$ min^{-1} and $\tau = 5.7$ s).

(a) Zone 1: the lower part of the vessel (between the bottom and impeller level), where the spatial variations of k are low;

(b) Zone 2: located in the vicinity of the impeller level, where a sharp increase of the mass transfer rates can be observed due to the corresponding increase of the fluid velocity by the impeller;

(c) Zone 3: the upper part of the vessel where k decreases slowly from the agitator to the outlet. The small differences observed between the two axes 1 and 2 suggest that transfer is homogenous in this zone.

As indicated above, two contributions have to be considered in the overall transfer phenomena and it is important to study each of these contributions separately

3.1. *Influence of the residence time τ (for $N = 0$)*

Fig. 3 reports the variations of k with the parameter $1/\tau$. The position of the probe inside the vessel is different for the three categories of results, which correspond to the three zones mentioned in the last section. Examination of this figure leads to the following conclusions:

(a) The mass transfer rates increase with $1/\tau$, i.e. with increasing volumetric flow rate Q_v. This result is expected on the basis of the theories of mass transfer phenomena.

(b) The relative classification for the values of

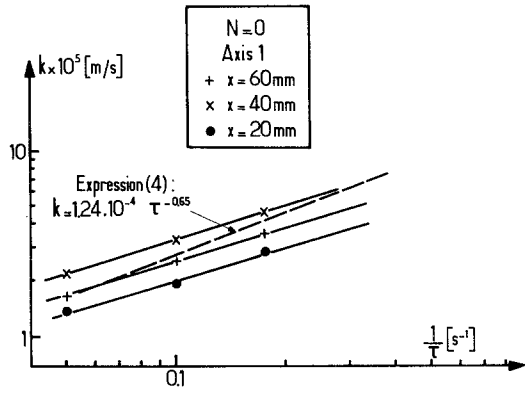

Fig. 3. Influence of the fluid residence time τ in the vessel (for $N = 0$). Comparison with theory.

k in Fig. 3 is in good agreement with the conclusions deduced from Fig. 2, i.e. the rates at the level of the impeller ($x = 40$ mm) are higher than those obtained at $x = 20$ or 60 mm. The differences observed are quite important and may be attributed to the large velocity distribution induced by the feeding fluid inside the reactor.

(c) The logarithmic plot of the results shows that k varies as $\tau^{-0.6}$, whatever the position in the reactor.

3.2. *Influence of the impeller speed N (for $1/\tau = 0$)*

Fig.4 reports the variations of k with impeller speed N for $Q_v = 0$ (no feed). For a given value

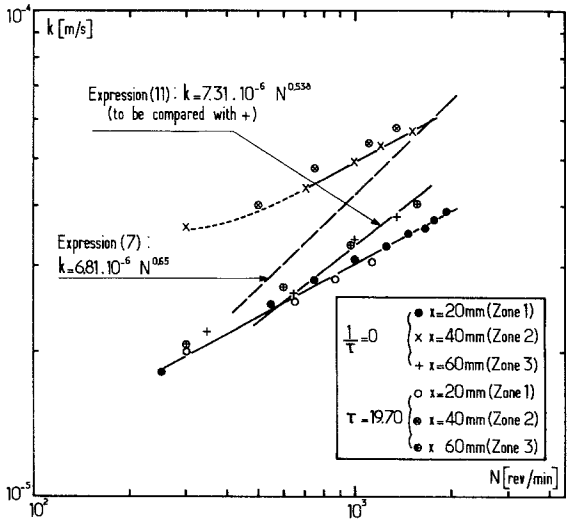

Fig. 4. Influence of the impeller rotation speed N for $1/\tau = 0$ (no feeding fluid) and $\tau = 19.7$ s. Comparison of the results with empirical correlations.

of N, no significative difference exists between the two curves corresponding to $x = 20$ mm (lower part of the vessel) and $x = 60$ mm (upper part). In contrast, at the level of the impeller, higher mass transfer rates occur due to the local increase of the fluid mean velocity in the stream induced by the agitator. The variations of k with N are such that k varies as $N^{0.37}$ for the range of N considered.

3.3. *Simultaneous influence of N and τ*

The experimental variations of k with N for three values of the residence time τ are reported in the three figures (Figs. 5, 6 and 7) corresponding respectively to the different positions on axis 1. For sufficiently high values of N ($N > 500$ r.p.m.) and residence times larger than 10 s, k depends only weakly on τ (a maximum 25% effect) and the corresponding zones have been hatched on the figures. As the overall mass transfer rate is essentially determined by the contributions of the kinetic energy of the feed jets and the agitation power delivered by the impeller, this result would lead to the conclusion that, for $N > 500$ r.p.m. and $\tau > 10$ s, only the second contribution has an appreciable influence on the mass transfer coefficients.

For $N > 500$ r.p.m. and smaller residence times ($\tau < 10$ s) a different behaviour is observed and the above conclusion is no longer valid. In fact

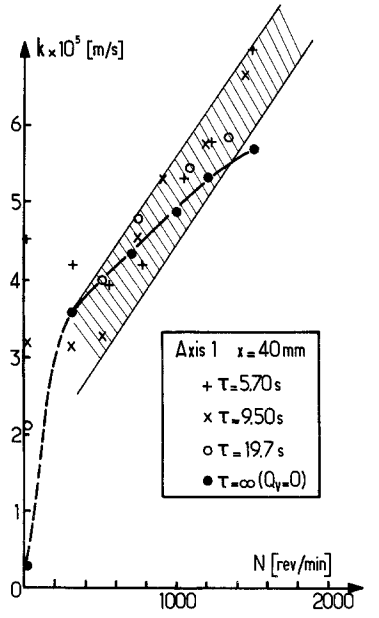

Fig. 6. Simultaneous influence of N and τ on the mass transfer rates in Zone 2.

(and this is particularly true at the bottom of the vessel in Zone 1), the mass transfer coefficients for $5.7 \text{ s} < \tau < 10$ s present higher values compared to those of the hatched zones. As will be shown in the next paragraph, a decrease of τ is synonymous with an increase in the mechanical power delivered by the feeding fluids, which then becomes competitive with the specific power of the impeller. However, in the vicinity of the

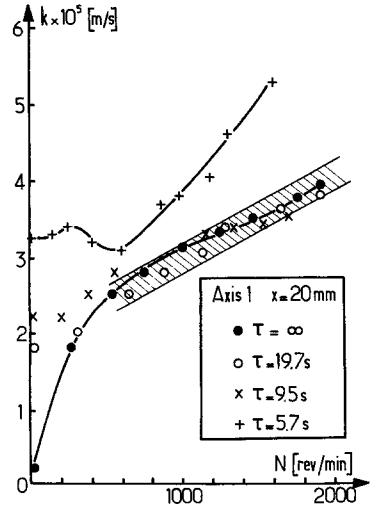

Fig. 5. Simultaneous influence of N and τ on the mass transfer rates in Zone 1.

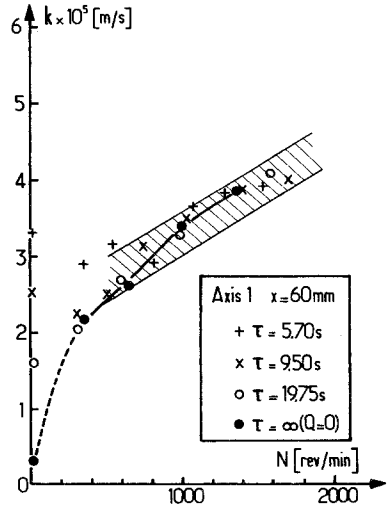

Fig. 7. Simultaneous influence of N and τ on the mass transfer rates in Zone 3.

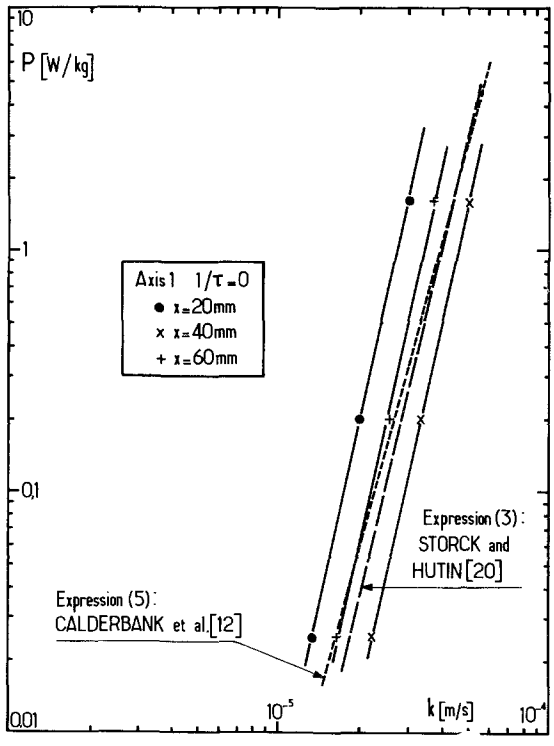

Fig. 8. Experimental and theoretical variations of the mass transfer coefficients with the specific power P delivered by the feeding fluids.

impeller (the results of Fig. 6), the high turbulence generated locally by the blades is not disturbed by the feeding jets, and no favourable effect of τ appears even for small values of this parameter.

For $N < 500$ r.p.m. the power dissipated by the agitator decreases and for very low speeds becomes negligible: it is then difficult to predict the effect of N on the mass transfer rates, because the two contributions to the overall power dissipated locally are probably not additive. From $N = 0$, the variations of k with N present minimum values, which would signify that an antagonistic effect exists between the two types of energy involved inside the reactor.

4. Interpretation of the results and energetic aspects

An extensive study for the interpretation of the mass transfer results would necessitate the measurement of the fluid velocity distribution everywhere in the reactor. This distribution was not determined in the present work and the following interpretation is based on a macroscopic approach to the mass transfer phenomena at the scale of the whole reactor; a tentative determination at the microscopic level will be proposed at the end of the paper.

4.1. *Case 1: Interpretation of the results for $N = 0$*

When the turbulence in the reactor is only generated by the kinetic energy of the feed fluids, it has been considered that the mass transfer rates are entirely determined by the specific power P (per unit mass of fluid) dissipated by the jets. Since the energy efficiency of the jets is 100% and the corresponding power is uniformly distributed over the whole reactor, it can easily be shown that P (in W kg^{-1}) is given by the following expression:

$$P = \frac{1}{2}\frac{V^2}{S^2\tau^3} \tag{1}$$

where S is the total area of injection of the nozzles. Furthermore, in a recent study related to the mass transfer phenomena between walls or grains and flowing liquids, Storck and Hutin [20] have deduced an energetic correlation of the results, which is very useful for a large range of hydrodynamic and geometric configurations:

$$(Sh)(Sc)^{-1/3} = \alpha\left(\frac{Pl^4}{\nu^3}\right)^{0.217}. \tag{2}$$

The Sherwood number is defined with respect to a characteristic length l, whose influence on k can be neglected as a first approximation, given the low value of the exponent in l.

For the numerical values of the physicochemical parameters considered in this work, the two following expressions can be deduced:

$$k = 3.93 \times 10^{-5}\, P^{0.217} \tag{3}$$

and

$$k = 1.24 \times 10^{-4}\, \tau^{-0.65} \tag{4}$$

(k in m s^{-1}). Comparison of Expression 4 with the experimental results of Fig. 3 shows that there is good agreement, even though the two exponents of τ, 0.6 (for experiment) and 0.65 (for theory), are different. The same comparison is reported in Fig. 8 in the form of the variations of P with k. The correlation proposed by Calderbank and Moo Young [12]

$$k(Sc)^{2/3} = 0.13\,(Pv)^{1/4} \quad (5)$$

is also given and is found to show very good agreement with Expression 2.

It is important to note that this interpretation supposes a uniform distribution of P in the vessel, an hypothesis which is, of course, impossible to verify because of the spatial variations observed in the mass transfer rates. However, it allows the prediction, with a sufficient precision for chemical engineering calculations, of the mass transfer coefficients.

4.2. *Case 2: Interpretation of the results for* $1/\tau = 0$

When only the contribution of the power delivered by the impeller has to be considered (no feeding fluids), an energy correlation analogous to Equation 2 can be tested; the specific power P was determined from electrical and thermal measurements reported in [21]. It was found that the power number $N_p = PM/\rho N^3 D_i^5$ was independent of the Reynolds number in the range of N considered and approximately equal to 6.0. This value is in good agreement with those reported by other authors [22–24] for a six-blade, flat-paddle agitator. For the values of the parameters considered in the study, P (W kg^{-1}) is calculated using the following expression:

$$P = 3.1 \times 10^{-4} N^3 \quad (6)$$

where N is in s^{-1}. Combining Equations 3 and 6 leads to:

$$k = 6.81 \times 10^{-6} N^{0.65} \quad (7)$$

which can be compared to the experimental values of Fig. 4. As can be seen, the slopes which show the variation with N are quite different (slopes of 0.65 and 0.37 for the experimental variations), but the overall correlation (Equation 7) falls graphically between the two categories of results corresponding to $x = 20$ mm and $x = 40$ mm. It seems, however, difficult to explain the low value (0.37) of the experimental slope, particularly at the level of the impeller ($x = 40$ mm). In Zone 2, a possible explanation could be that the probe diameter is too large compared with the impeller thickness (only 6.6 mm).

As mentioned above, a more exhaustive study would necessitate knowing the velocity distribution inside the vessel. For a geometrical configurational analogue to ours, Le Lan and Angelino [11] have determined experimentally the spatial variations of a hydrodynamical parameter E defined by the relation

$$E = 0.6 \left(\frac{U}{\pi N D_i} \right)^{1/2} \quad (8)$$

where U is the local fluid velocity, which is supposed to be proportional to the blade velocity $\pi N D_i$. In the upper part of the vessel (Zone 3) their results showed the existence of a large zone where E remains practically constant and equal to 0.18 (cf. Fig. 5 of [11], p. 911). This result, which gives a quantitative relation between U and N above the impeller level, can be used to determine the mass transfer rates.

The following procedure has been used: first the local fluid velocity U (m s^{-1}) is calculated from Equation 8, given that $D_i = 3.3 \times 10^{-2}$ m,

$$U = 9.33 \times 10^{-3} N. \quad (9)$$

The mass transfer coefficient is then calculated from a general correlation proposed by Gibert *et al.* [25] for mass transfer between a sphere and a flowing liquid:

$$(Sh)(Sc)^{-1/3} = 0.447\,(Re)^{0.538} \quad (Re) > 1250 \quad (10)$$

where (Sh) and (Re) are defined with respect to the particle diameter d_p.

Combining Equations 9 and 10 leads to the two following expressions for k:

$$k = 7.31 \times 10^{-6} N^{0.538} \quad (11)$$

and

$$k = 3.13 \times 10^{-5} P^{0.18}. \quad (12)$$

For a point (axis 1 and $x = 60$ mm) located above the impeller, Fig. 4 compares the experimental values to those calculated from Equation 11. As can be seen the agreement is very satisfactory given the imprecisions in the experiments and the use of a parameter E not measured by the authors of this work. This good agreement would lead to the conclusion that it is possible to relate very simply hydrodynamic parameters to mass transfer phenomena in agitated vessels.

5. Conclusions

From this study of mass transfer in agitated vessels, it may be concluded that:

(a) The mass transfer rates present large spatial variations and the maximum values are obtained at the level of the impeller, where the mean fluid velocities are the highest.

(b) When the liquid turbulence is only generated by the feed fluids, an energetical correlation based on the specific power dissipated in the vessel by the jets has been proposed which allows the prediction of the mass transfer rates with sufficient precision.

(c) When the turbulence is generated by the impeller rotation, some deviations exists between the experimental results and the theoretical correlation, due probably to the size of the measuring probe compared with the impeller thickness. However, a microscopic approach using the velocity distribution determined by other authors was found very useful for the prediction of the local mass transfer coefficients from the corresponding local fluid velocities.

(d) For fluid residence times τ larger than 10 s, the coefficients k are entirely determined by the impeller speed $\pi N D_i$ and depend only weakly on τ

References

[1] S. Boon-Long, C. Laguerie and J. P. Couderc, *Chem. Eng. Sci.* **33** (1978) 813.

[2] R. B. Keey and J. B. Glen, *AIChE J.* **12** (1966) 401.

[3] S. Sicardi, R. Conti, G. Baldi and R. Cresta, *Third European Conf. on Mixing, University of York, England* (4–6 April 1979) Paper D2, p. 217.

[4] P. Sykes and A. Gomezplata, *Can. J. Chem. Eng.* **45** (1967) 189.

[5] T. Mizushina, R. Ito, K. Kataoka, S. Yokoyama, Y. Nakajima and A. Fukuda, *Kagaku Kogaku* **32** (1968) 795.

[6] T. Mizushina, R. Ito, S. Hiraoka, A. Ibusuki and I. Sakaguchi, *J. Chem. Eng. Japan* **2** (1969) 89.

[7] M. S. Murthy, M. Balakrishna and J. S. N. Murthy, *J. Ind. Inst. Sci.* **60** (1978) 299.

[8] I. Fort, J. Placek, F. Strek, Z. Jaworski and J. Karcz, *Coll. Czechoslov. Chem. Commun.* **44** (1979) 684.

[9] D. N. Miller, *Chem. Eng. Sci.* **22** (1967) 1617.

[10] A. Le Lan, H. Gibert and H. Angelino, *ibid* **27** (1972) 1979.

[11] A. Le Lan and H. Angelino, *ibid* **29** (1974) 907.

[12] P. H. Calderbank and Moo Young, *ibid* **16** (1961) 39.

[13] S. Aiba, *AIChE J.* **4** (1958) 485.

[14] J. P. Sachs and J. H. Rushton, *Chem. Eng. Prog.* **50** (1954) 597.

[15] W. J. Kim and F. S. Manning, *AIChE J.* **10** (1964) 747.

[16] F. A. Holland and F. S. Chapman, 'Liquid Mixing and Processing in Stirred Tanks', Reinhold Publishing Corporation, New York (1966).

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

[18] D. Hutin and A. Storck, *J. Appl. Electrochem.* **9** (1979) 351.

[19] A. Storck, P. M. Robertson and N. Ibl, *Electrochimica Acta* **24** (1979) 373.

[20] A. Storck and D. Hutin, *Can. J. Chem. Eng.* **58** (1980) 92.

[21] J. F. Brodberger, PhD thesis, INPL, Nancy (1981).

[22] J. H. Rushton, E. W. Costich and H. J. Everett, *Chem. Eng. Prog.* **46** (1950) 467.

[23] P. M. Calderbank, *Trans AIChE* **36** (1958) 443.

[24] A. W. Flynn and R. E. Treybal, *AIChE J.* **1** (1955) 324.

[25] H. Gibert, J. P. Couderc and H. Angelino, *Chem. Eng. Sci.* **27** (1972) 45.