Chemical Engineering Journal 69 (1998) 39–45

Chemical Engineering Journal

Mass transfer in fixed bed solid dissolution

Stelian Petrescu *, Ioana Fechete, Claudiu Ciubotariu

Department of Chemical Engineering, Technical University of Iaşi B-ul Copou 22, 6600 Iaşi, Romania

Received 19 September 1996; revised 29 July 1997; accepted 13 October 1997

Abstract

A method for the experimental determination of the dissolution rate and of the individual mass transfer coefficient during the dissolution of a fixed bed formed of spherical particles is proposed in this paper. Using this method, the mean dissolution rate and the mean individual mass transfer coefficient are determined experimentally for a fixed bed of urea particles dissolving in water. The results obtained agree with the data given in literature. The influence of the solvent flowrate and of temperature on the mean dissolution rate are also studied.

Keywords: Solid dissolution; Dissolution rate; Mass transfer; Individual mass transfer coefficient

1. Introduction

Solid dissolution is frequently encountered in industrial processes. The solid phase involved in dissolution is granular in most cases. Dissolution of a solid in a liquid may be realized in a fixed bed, a fluidized bed or in suspension in agitated vessels.

Technical literature reports a number of papers [1–14] in which mass transfer is studied using one of the following techniques: physical dissolution and electrochemical processes.

The studies concerning mass transfer in solid–liquid systems reported in literature [1,8–14], lead to mathematical models and equations which enable the calculation of the individual mass transfer coefficient.

Evans and Gerald [9] and Upadhyay and Tripathi [10] studied the dissolution of a fixed layer of benzoic acid particles in water.

Wakao and Fumazkri [12] show that for Reynolds numbers greater than 3, axial dispersion is negligible. They also establish an equation for mass transfer in systems with spherical particles using the dissolution technique.

In most papers the dissolution rate is determined using the mass balance for the liquid phase and the experimental determination of the solute concentration.

This paper presents a method for the experimental determination of the dissolution rate and of the individual mass transfer coefficient for the dissolution of a spherical particle fixed bed. The method was applied for the study of mass transfer during dissolution of a urea fixed bed in water at atmospheric pressure and for several temperatures. The influence of temperature, of the solvent flowrate and of the particle size upon the mean dissolution rate was also studied.

2. Method of determination of the individual mass transfer coefficient

In order to determine the individual mass transfer coefficient for the dissolution of a fixed bed of particles in a solvent, the mass transfer equation is used:

$$\bar{v}_{\mathrm{D}} = k_1 (C_{\mathrm{A}}^* - C_{\mathrm{A}})_{\mathrm{mean}} \tag{1}$$

The mean driving force is calculated with the relation $((C_{\mathrm{A}}^* - C_{\mathrm{A,in}}) - (C_{\mathrm{A}}^* - C_{\mathrm{A,out}}))/\ln((C_{\mathrm{A}}^* - C_{\mathrm{A,in}})/(C_{\mathrm{A}}^* - C_{\mathrm{A,out}}))$. When the driving force from the top and that from the bottom of the fixed bed are relatively close, the mean driving force may be calculated with $(C_{\mathrm{A}}^* - C_{\mathrm{A}})_{\mathrm{mean}} = ((C_{\mathrm{A}}^* - C_{\mathrm{A,in}}) + (C_{\mathrm{A}}^* - C_{\mathrm{A,out}}))/2$.

The fixed bed particle dissolution rate may be expressed by the equation:

$$v_{\mathrm{D}} = -\frac{\mathrm{d}m}{S \cdot \mathrm{d}t} \tag{2}$$

We assume that all particles are identical and during the process each particle has the same dissolution rate. As a result Eq. (2) becomes:

---

* Corresponding author.

1385-8947/98/$19.00
PII S1385-8947(97)00107-1

$$v_D = -\frac{dm_p}{S_p \cdot dt} \tag{3}$$

For spherical particles Eq. (3) becomes:

$$v_D = -\frac{\rho_s}{2} \cdot \frac{dd_p}{dt} \tag{4}$$

Eq. (4) shows that the determination of the particle dissolution rate implies establishing the particle diameter at a given time. To do this we use the definition of the fixed bed porosity. For spherical particles, which maintain their shape during dissolution Eq. (5) gives:

$$d_p = d_p^0 \cdot \left[\frac{H(1-\varepsilon)}{H_0(1-\varepsilon^0)}\right]^{1/3} \tag{5}$$

If $\varepsilon \cong \varepsilon^0$,

$$d_p = d_p^0 \cdot \left[\frac{H}{H_0}\right]^{1/3} \tag{6}$$

3. Experimental

The experiments were conducted on the laboratory installation presented in Fig. 1. The installation is composed of a dissolver (1), the installation support (2), a distilled water reservoir (3), a recipient for solution resulting during dissolution (4), a rotameter for the distilled water flowrate (5), a thermometer for measuring the temperature inside the dissolver (6), clamps (7), valves V1 and V2 for distilled water flowrate adjustment and solution sample-drawing.

The dissolver (1) is fixed with a clamp (7) to the installation prop (2). The dissolver is a cylindrical column with a water jacket and a dismountable lid, made of glass. The dissolver inner diameter is 0.0225 m and the height measured from the upper layer surface (1b) to the lid is 0.08 m. A stainless steel sieve (1a) inside the dissolver bears a fixed bed made of glass balls (1b) with a height of 0.02 m, the ball diameter being 0.00225 m. On top of the ball layer is the particle layer (1c) which is to be dissolved. An inlet for solvent (distilled water) admission and a thermometer (6) are fixed on the lid.

The solvent flows from the reservoir (3) in the rotameter and then to the dissolver flowing downward over the fixed particle bed and the glass balls. The necessary water flowrate is adjusted through the rotameter (5) with the valve V1. The temperature inside the dissolver is maintained constant by a thermostat which recycles water through the dissolver mantle. During particle dissolution, aqueous solution flows from the bottom of the dissolver to the lower reservoir (4). The valve V2 is used to draw solution samples at the dissolver outlet.

The experiments were carried out using distilled water as a solvent and urea particles at atmospheric pressure and at the following temperatures: 288 K, 298 K and 303 K. The particles were sifted, using for the experiments the following mean diameters: 0.00205 m and 0.00282 m. The distilled water flowrate was 5.55×10^{-7}, 1.11×10^{-6} and 1.66×10^{-6} $m^3\ s^{-1}$, respectively. The same initial volume of particles

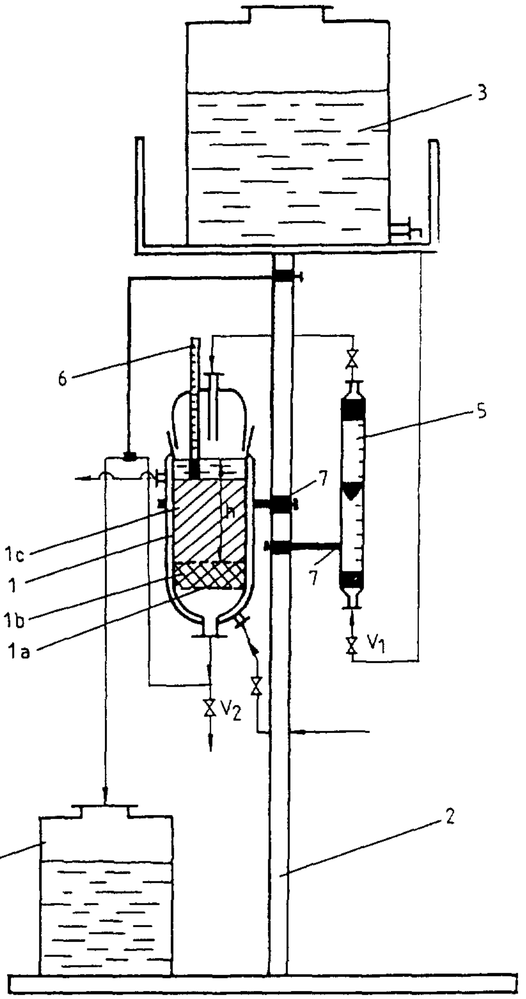

Fig. 1. Experimental installation: (1) dissolver; (2) prop; (3) distilled water recipient; (4) solution recipient; (5) rotameter; (6) thermometer; (7) clamp.

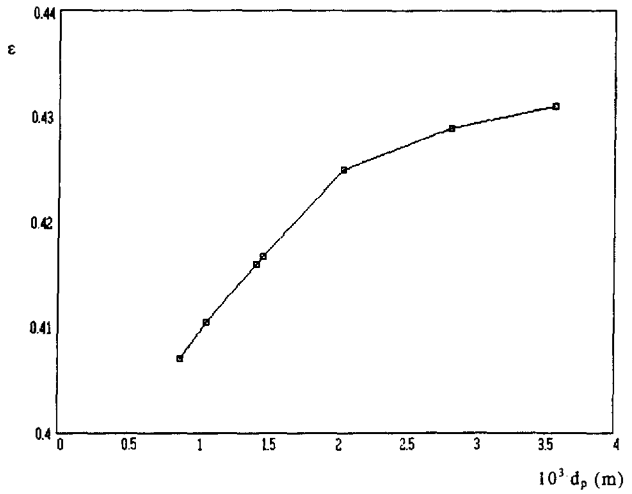

Fig. 2. Variation of the fixed bed porosity vs. the urea particle diameter.

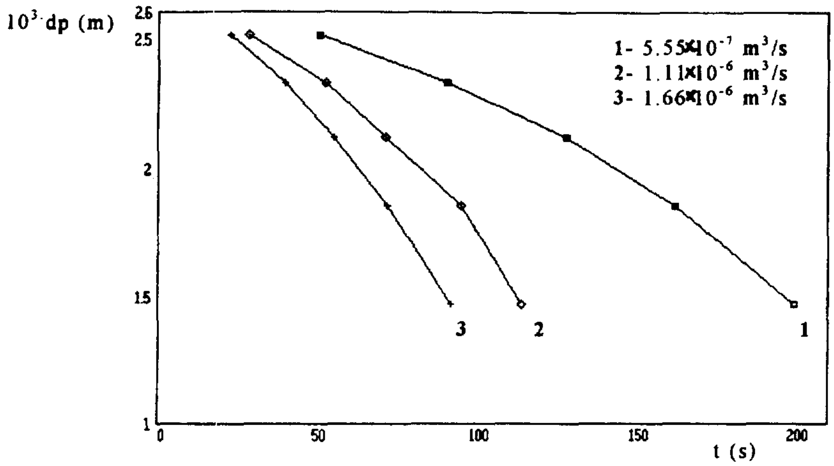

Fig. 3. Dependence $d_p=f(t)$, $d_p^0=0.002825$ m, $T=288$ K.

was used in all experiments and consequently the same initial height of the fixed bed $H_0=0.0713$ m.

4. Results and discussions

In the first stage we studied the influence of temperature, solvent flowrate and urea particle size on the dissolution rate and the time dependence of the dissolution rate. For this purpose the urea fixed bed height was determined and monitored as a function of time in each experiment. The temperature was maintained constant at the prescribed value during the experiment.

Experimental determination of the individual mass transfer coefficient requires knowledge of the solute concentration in the bulk of the liquid phase so that the time dependence of the solute concentration at the dissolver outlet was determined experimentally.

The urea fixed bed porosity was also determined experimentally for several particle diameters; the dependence $\varepsilon=f(d_p)$ is presented in Fig. 2. The fixed bed porosity was determined using a graduated cylinder having the same diameter as the dissolver in the installation. The porosity was determined before the dissolution using spherical urea particles. In order to establish the dependence of porosity on the urea particle diameter, the particles used in the experiments were previously sorted by their diameter. As may be seen the bed porosity is only slightly influenced by the particle diameter. For a diameter between 0.001 m and 0.0032 m the bed porosity lies between 0.41 and 0.43. Consequently Eq. (6) may be used to compute the particle diameter at a given time.

Using Eq. (6) the urea particle diameters corresponding to the fixed bed height determined experimentally were computed. The results are presented in Figs. 3–8. As may be seen the urea particle diameter decreases with time during dissolution for all temperatures, initial sizes and solvent flowrates investigated. The particle diameter decrease is more pronounced for larger values of the dissolution time.

Using Eq. (4), the data shown in Figs. 3–8 and the data reported in Table 1, the particle dissolution rate was computed.

It was found that the dissolution rate of urea particles in a fixed bed increases with time.

In Figs. 9 and 10 the mean dissolution rate is plotted vs. the solvent flowrate for several temperatures. The mean dissolution rate was calculated as a mean value in time.

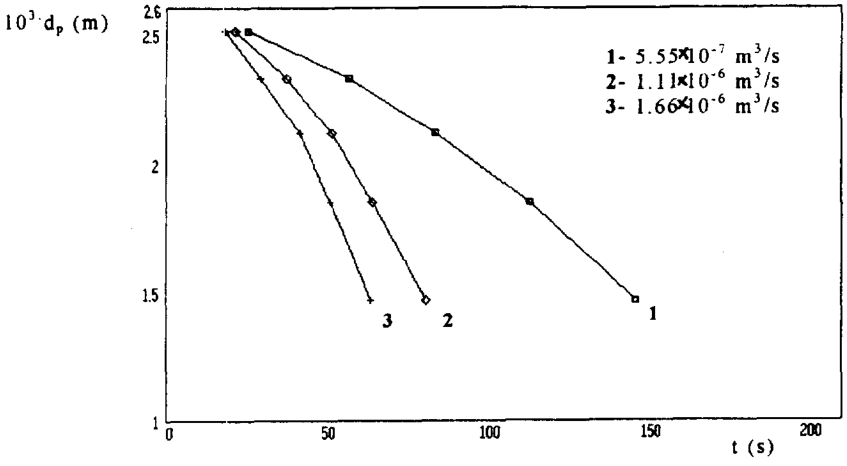

Fig. 4. Dependence $d_p=f(t)$, $d_p^0=0.002825$ m, $T=298$ K.

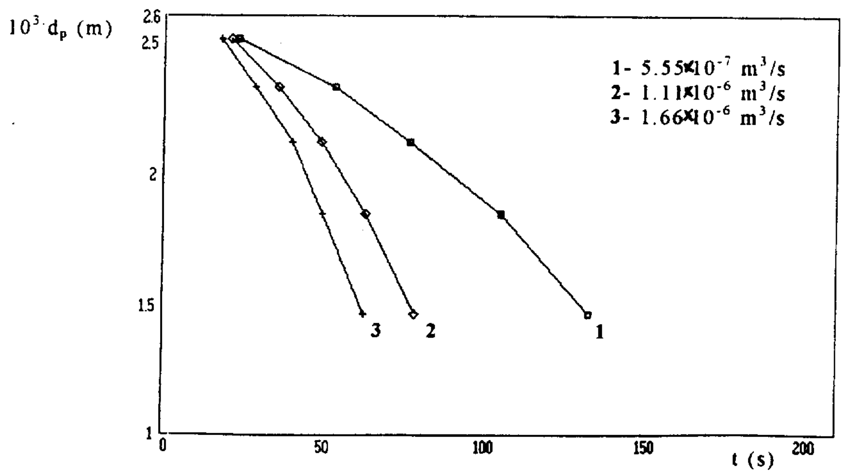

Fig. 5. Dependence $d_p = f(t)$, $d_p^0 = 0.002825$ m, $T = 303$ K.

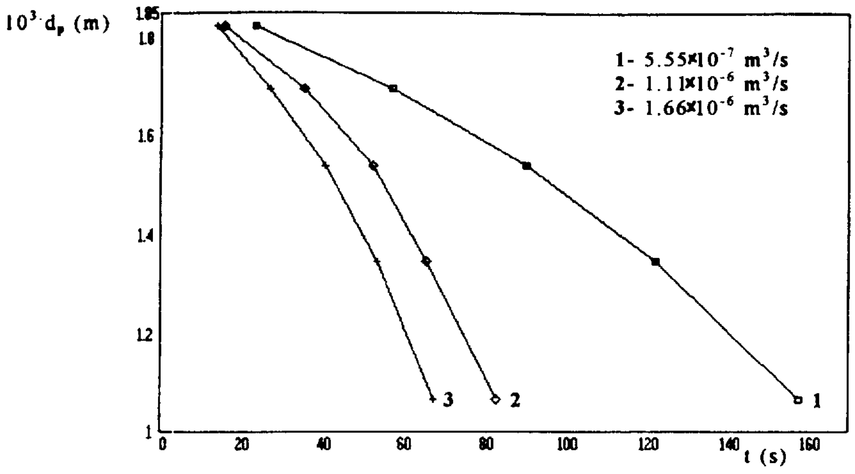

Fig. 6. Variation of the urea particle diameter in time, $d_p^0 = 0.00205$ m, $T = 288$ K.

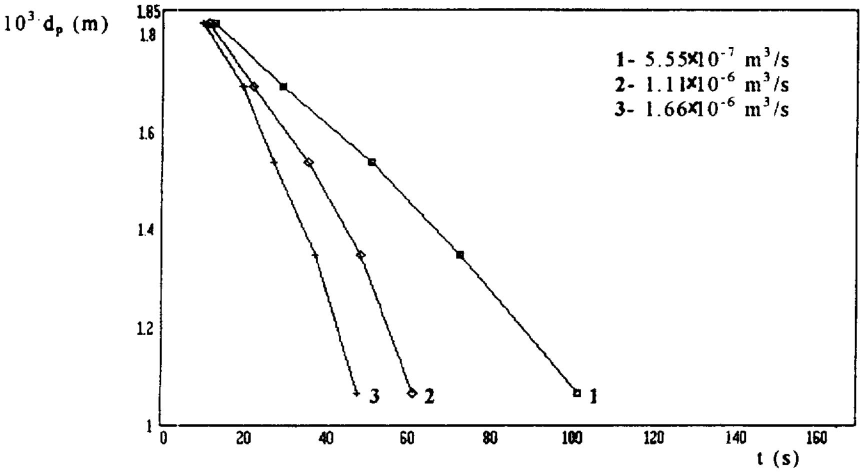

Fig. 7. Variation of the urea particle diameter in time, $d_p^0 = 0.00205$ m, $T = 298$ K.

Temperature exerts a positive influence over the dissolution rate of solid particles in fixed beds. For a small solvent flowrate, temperature has a slight influence, while for a large solvent flowrate the influence of the temperature is somewhat greater.

The solvent flowrate also has a positive influence on the dissolution rate as may be seen in Figs. 9 and 10. An increase in the solvent flowrate increases the solvent velocity in the bed and thus intensifies the mass transfer.

The initial particle diameter has a slight influence on the mean dissolution rate because the porosity is only slightly dependent on the particle diameter.

Based on the measurements of mean dissolution rate, and using Eq. (1) and Table 1, the mean individual mass transfer

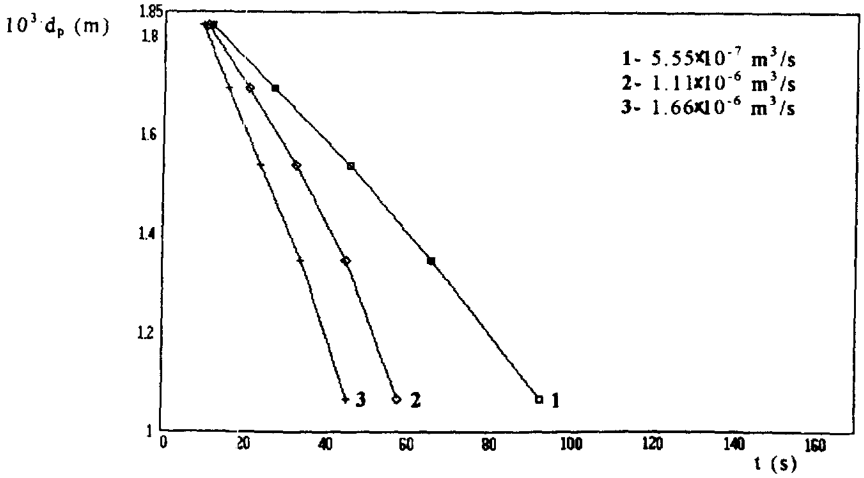

Fig. 8. Variation of the urea particle diameter in time, $d_p^0 = 0.00205$ m, $T = 303$ K.

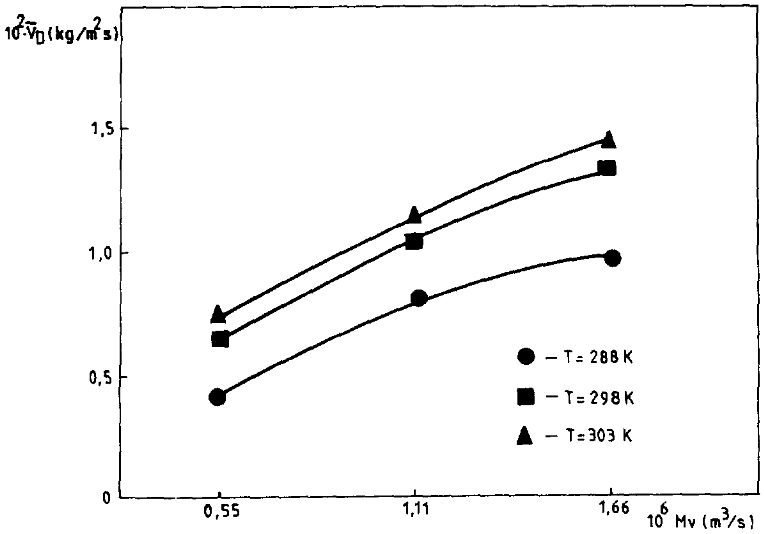

Fig. 9. Mean dissolution rate, $d_p^0 = 0.00205$ m.

Table 1
Physical properties

Properties	Values	References
Density of the particle	1335 kg m^{-3}	[15]
Diffusion coefficient of urea in aqueous urea solution		
288 K	0.995×10^{-9} m^2 s^{-1}	experimental values
298 K	1.098×10^{-9} m^2 s^{-1}	
303 K	1.141×10^{-9} m^2 s^{-1}	
Solubility of urea in water		
288 K	556.7 kg m^{-3}	[15]
298 K	630.0 kg m^{-3}	
303 K	661.8 kg m^{-3}	
Density of the aqueous urea solution		
288 K	1042 kg m^{-3}	[15]
298 K	1038 kg m^{-3}	
303 K	1036 kg m^{-3}	
Viscosity of the aqueous urea solution		
288 K	1.25×10^{-3} N s m^{-2}	[15]
298 K	1.00×10^{-3} N s m^{-2}	
303 K	0.90×10^{-3} N s m^{-2}	

coefficient could be determined. Since the inlet concentration is zero and the outlet concentration is between 208 and 278 kg m^{-3}, the relation used for the calculation of the driving force is:

$$(C_A^* - C_A)_{\text{mean}} = C_A^* - \frac{C_{A,\text{out}}}{2} \qquad (7)$$

The results obtained are presented in Fig. 11 in the form Sh Sc$^{-1/3}$ = f(Re). The data obtained show a small influence of the Schmidt number upon the Sherwood number and the mean individual mass transfer coefficient.

In order to verify the experimental data concerning the individual mass transfer coefficient the Evans–Gerald and Wakao–Furnazkri equations (Eqs. (8) and (9), respectively) were used, written in the form:

$$\text{Sh} = 1.48\text{Re}^{0.48}\text{Sc}^{1/3}, \quad 2 < \text{Re} < 50 \qquad (8)$$

$$\text{Sh} = 2 + 1.1\text{Re}^{0.6}\text{Sc}^{1/3}, \quad \text{Re} > 3 \qquad (9)$$

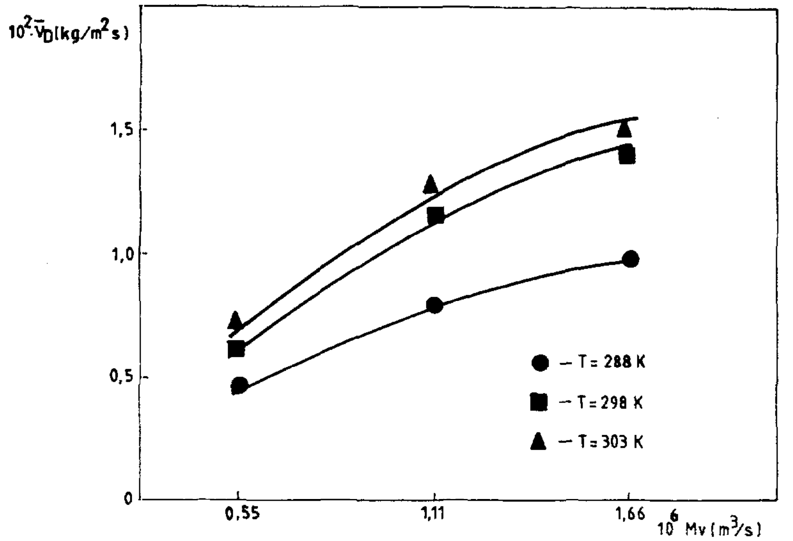

Fig. 10. Mean dissolution rate, $d_p^0 = 0.002825$ m.

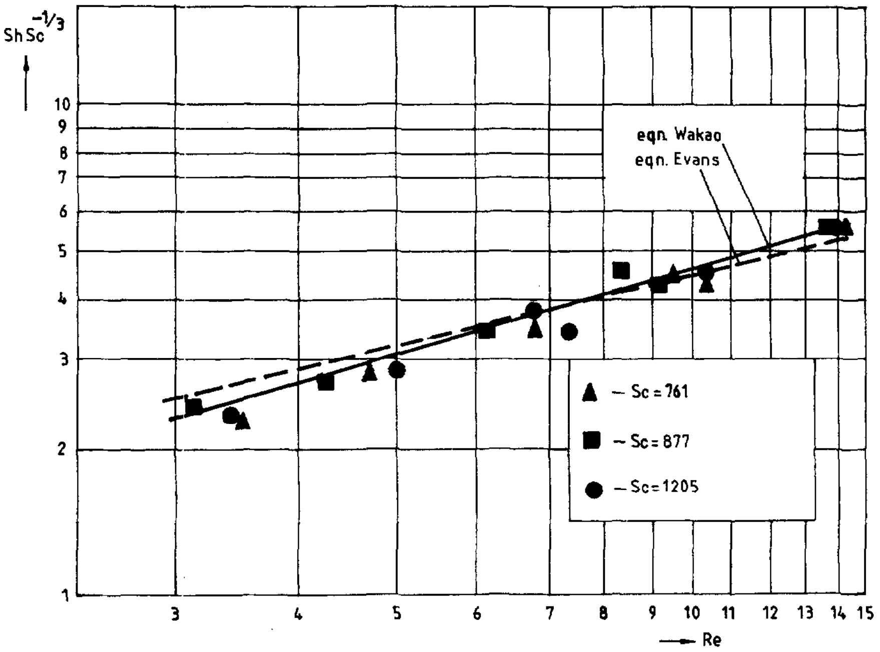

Fig. 11. Dependence of Sherwood number on Reynolds number for dissolving urea particles.

Fig. 11 shows a good agreement between the experimental data and literature correlations for a Reynolds number between 3 and 15.

The experimental values of mass transfer coefficient we obtained are about 5% smaller than the data presented in literature by Wakao and Evans.

The dissolved urea weight, the dissolution rate (v_D'), the individual mass transfer coefficient (k_l') and the Sherwood number (Sh′) were determined using the mass balance of the liquid phase. The values of Sh′ were compared with those of the Sherwood number (Sh) obtained with the proposed method. For this purpose the ratio Sh/Sh′ was calculated at 303 K for several values of the water flowrate. The results are presented in Table 2. The data show that the ratio Sh/Sh′ is close to unity which means that the dissolution rate determined with the proposed method is practically equal to that obtained from the mass balance of the liquid phase.

Consequently we may consider that the method proposed for the determination of the dissolution rate and of the individual mass transfer coefficient is accurate.

5. Conclusions

The paper presents a method for the experimental determination of the individual mass transfer coefficient and of the dissolution rate in a fixed particle bed, using the change in particle diameter with time.

Table 2
The ratio Sh/Sh′ at 303 K

Particle diameter, 10^3 d_p (m)	Sh/Sh′		
	Water flowrate, $10^6 Mv$ ($m^3 s^{-1}$)		
	0.555	1.11	1.66
2.05	1.0393	1.042	1.0316
2.825	0.9833	1.0396	1.0254

This method enables the determination of the individual mass transfer coefficient and of the dissolution rate in a fixed particle bed.

The mean individual mass transfer coefficient and the mean dissolution rate of the fixed bed urea particle dissolving in water were determined using this method. The influence of temperature, solvent flowrate and initial urea particle diameter over the mean dissolution rate were studied.

The results obtained in the experiments were compared to two literature correlations, and for a Reynolds number between 3 and 15 good agreement was found.

The validity of the proposed method was also established using the mass balance of the liquid phase.

Appendix A. Nomenclature

C_A	Current solute concentration ($kg\ m^{-3}$)
D_A	Diffusion coefficient ($m^2\ s^{-1}$)
d_p	Particle diameter at a given time (m)
H	Current height of the fixed bed (m)
k_l	Individual mass transfer coefficient ($m\ s^{-1}$)
k_l'	Individual mass transfer coefficient, $k_l' = \bar{v}_D' / (C_A^* - C_A')_{mean}$, ($m\ s^{-1}$)
m	Mass of all particles in the fixed bed (kg)
m_p	Particle mass at a given time (kg)
n	Number of particles in the bed (dimensionless)
S	Area of the particles exterior surface (m^2)
S_p	Area of the particle surface at a given time (m^2)
t	Time (s)
v	Superficial velocity of the liquid in the fixed bed ($m\ s^{-1}$)
V	Volume (m^3)
v_D	Dissolution rate ($kg\ m^{-2}\ s^{-1}$)
$\bar{v}_D$	Mean dissolution rate ($kg\ m^{-2}\ s^{-1}$)
$\bar{v}_D'$	Mean dissolution rate calculated using mass balance of the liquid phase ($kg\ m^{-2}\ s^{-1}$)

Greek letters

ε	Fixed bed porosity (dimensionless)
η	Dynamic viscosity ($N\ s\ m^{-2}$)
ρ	Density ($kg\ m^{-3}$)

Subscripts

A	Solute component (urea)
FB	Fixed bed
in	Inlet
l	Liquid
out	Outlet
p	Particle
s	Solid
0	Initial conditions

Superscripts

0	Initial conditions
*	Equilibrium conditions

Dimensionless numbers

Re	Reynolds number ($Re = \rho_l \cdot v \cdot d_p / \eta_l$)
Sc	Schmidt number ($Sc = \eta_l / \rho_l \cdot D_A$)
Sh	Sherwood number ($Sh = k_l \cdot d_p / D_A$)
Sh′	Sherwood number ($Sh' = k_l' \cdot d_p / D_A$)

References

[1] T.K. Sherwood, R.L. Pigford, C.R. Wilke, Mass Transfer, McGraw-Hill, New York, 1975, 214 pp.
[2] Z. Sterbacek, P. Tansk, Amestecarea, E.T., Bucuresti, 1969, 168 pp.
[3] D.M. Levins, J.R. Glastonbury, Trans. Inst. Chem. Eng. 50 (1972) 132.
[4] S. Boon-Long, C. Laguerie, J.P. Couderc, Chem. Eng. Sci. 33 (7) (1978) 813.
[5] N. Yutani, N. Ototake, L.T. Fan, Ind. Eng. Chem. Res. 26 (2) (1987) 343.
[6] A. Tamir, O. Falk, Ind. Eng. Chem. Res. 27 (10) (1988) 1930.
[7] A. Lakota, J. Levec, AIChE J. 36 (9) (1990) 1444.
[8] Y. Kawase, J. Ulbrecht, Ind. Eng. Chem. Fundam. 24 (1985) 115.
[9] G.C. Evans, C.A. Gerald, Chem. Eng. Prog. 49 (1953) 135.
[10] S.N. Upadhyay, G. Tripathi, J. Chem. Eng. Data 20 (1975) 20.
[11] P.N. Dwivedi, S.N. Upadhyay, Ind. Eng. Chem. Process Des. Dev. 16 (1977) 157.
[12] N. Wakao, T. Furnazkri, Chem. Eng. Sci. 33 (1978) 1375.
[13] J. Comiti, M. Renaud, Chem. Eng. Sci. 46 (1991) 143.
[14] D. Seguin, A. Montillet, D. Brunjai, J. Comiti, Chem. Eng. J. 63 (1996).
[15] Spravochinik azotchika, tom 2, Izd. Khimia, Leningrad, 1969, 178 pp.