ELSEVIER

Chemical Engineering Journal 66 (1997) 57–63

CHEMICAL
ENGINEERING
JOURNAL

Short communication

Mass transfer at solid dissolution

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Received 13 November 1995; accepted 15 March 1996

Abstract

An experimental method is presented for the determination of the individual mass transfer coefficient at solid dissolution and of the solid dissolution rate. This method assumes that dissolution takes place at the particle level, the particle falling in a stationary liquid. The method is applied to the dissolution of urea particles in water and in aqueous urea solutions.

Keywords: Solid dissolution; Mass transfer coefficient; Urea solution; Urea particles

1. Introduction

Solid dissolution is a fully studied process, the technical literature today comprising a large number of papers dealing with the theoretical and experimental study of mass transfer in solid dissolution. Mass transfer is analyzed at both the particle and the bulk level (fixed bed, fluidized bed or suspension) [1–14].

As a result of the research concerning mass transfer at the particle level, mathematical models are proposed for the individual mass transfer coefficient, the dissolution rate and the complete dissolution time. Based on the Levich and Frosling equations in [8], three mathematical models are established for the dissolution time of a spherical particle falling freely in a liquid. The mathematical model was verified experimentally using urea particles and water.

In [9,10] the authors propose a physical model based on which a mathematical model is established for the computation of the individual mass transfer coefficient of a cubic dissolving particle falling freely in a liquid. The mathematical model is verified experimentally using cubic KCl particles and aqueous $(NH_4)SO_4$ solution.

After a detailed presentation of the criterial equations for mass transfer between a spherical particle and a forced convection liquid, the authors of [12] establish two criterial equations for the dissolution process using an electrochemical technique.

In the present paper a rapid method for the experimental determination of the individual mass transfer coefficient of dissolving solids and of their dissolution rate is proposed. The method implies experimental determinations for individual spherical particles falling freely in a liquid in which they dissolve. According to this method the falling time for three different sections is determined for each particle. This method does not require a knowledge of the particle's initial diameter. The method is tested using urea particles dissolving in water or aqueous urea solutions of several concentrations.

2. Method for the determination of the individual mass transfer coefficient k_1

In the case of a particle formed of only one component A which moves by free falling in a solvent, the following mass balance equation can be written:

$$-\mathrm{d}m_A = k_1 S(C_A^e - C_A^0)\,\mathrm{d}t \tag{1}$$

where m_A is the particle mass (kg) at moment t, k_1 the individual mass transfer coefficient (m s^{-1}), S the exterior area (m^2) of the particle at time t, C_A^e the mass concentration of solute at equilibrium (kg m^{-3}), and t the duration of the process (s).

Since the dissolution rate for component A may be expressed by the relation:

$$v_D = -\frac{\mathrm{d}m_A}{S\mathrm{d}t} \tag{2}$$

the mass balance Eq. (1) becomes:

$$v_D = k_1(C_A^e - C_A^0) \tag{3}$$

If the particle is spherical then:

$$S = \pi D^2 \tag{4}$$

$$\mathrm{d}m_A = \rho_s \mathrm{d}V = \frac{\pi\rho_s}{2}D^2\mathrm{d}D \tag{5}$$

PII S1385-8947(96)03133-8

and the dissolution rate defined by Eq. (2) may be written as:

$$v_D = -\frac{\rho_s}{2}\frac{dD}{dt} \quad (6)$$

where D is the particle diameter (m) at time t (s), ρ_s the particle density (kg m^{-3}), and V the particle volume (m^3) at moment t.

During the free fall particle dissolution occurs. As a result the diameter decreases continuously. For short intervals of time the process may be regarded as a sedimentation process and the particle diameter may be determined using the expression for the sedimentation rate [15]:

$$v_0 = \left(\frac{4D(\rho_s - \rho)g}{3\rho\zeta}\right)^{1/2} \quad (7)$$

where ζ is the friction factor (dimensionless), ρ the liquid density (kg m^{-3}), and g the gravitational constant (m s^{-2}).

The friction factor ζ is given by the relation [16]:

$$\zeta = a(Re)^{-n} = a\left(\frac{\rho v_0 D}{\eta}\right)^{-n} \quad (8)$$

From Eqs. (7) and (8) we obtain:

$$D = C \cdot v_0^{2-n/n+1} \quad (9)$$

$$C = \left(\frac{3a}{4g}\right)^{1/n+1} (\rho_s - \rho)^{-1/n+1} \rho^{1-n/n+1} \eta^{2-n/n+1} \quad (10)$$

In order to compute the dissolution rate Eqs.(6), (9) and (10) are used, where dD/dt is substituted by $\Delta D/\Delta t$:

$$v_D = -\frac{\rho_s}{2}\frac{\Delta D}{\Delta t} \quad (11)$$

The significance of the finite differences ΔD and Δt results from Fig. 1:

$$\Delta D = D_2 - D_1 \quad (12)$$

$$\Delta t = \frac{t_1 + t_2}{2} + t_3 \quad (13)$$

where D_1 and D_2 are the particle diameters when falling from A to B and from C to D respectively, and t_1, t_2, t_3 the falling times corresponding to sections A–B, C–D, B–C respectively.

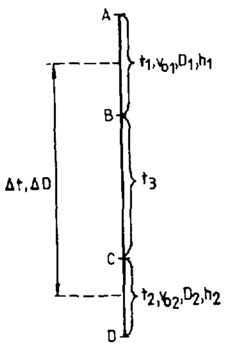

Fig. 1. The significance of some physical variables.

The particle diameters D_1 and D_2 are expressed by the relations:

$$D_1 = C \cdot v_{01}^{2-n/n+1} \quad (14)$$

$$D_2 = C \cdot v_{02}^{2-n/n+1} \quad (15)$$

Knowing the dissolution rate, the individual mass transfer coefficient k_1 is determined using Eq. (3). The sedimentation rate for sections A–B and C–D is determined from the relations:

$$v_{01} = \frac{h_1}{t_1} \quad (16)$$

$$v_{02} = \frac{h_2}{t_2} \quad (17)$$

where t_1 and t_2 are determined experimentally.

Eqs. (10), (14) and (15) for the sedimentation region ($a = 18.5$, $n = 0.6$) [16] become:

$$C = 1.242(\rho_s - \rho)^{-0.625}\rho^{0.25}\eta^{0.375} \quad (18)$$

$$D_1 = C \cdot v_{01}^{0.875} \quad (19)$$

$$D_2 = C \cdot v_{02}^{0.875} \quad (20)$$

3. Experimental

The experimental determination of the dissolution rate and of the individual mass transfer coefficient for granules falling freely in a solvent implies the experimental determination of the falling time of the particle between A–C, B–C and C–D.

For this purpose we used an experimental installation which is presented in Fig. 2. According to Fig. 2 the experimental installation consists of a vertical glass column (1) with an inner diameter 0.55 m and height 1 m. The column has four marks A, B, C and D and is supported by a metal prop (2) with clamps (3). At the upper part of the column there is a funnel (4) where the granules are introduced. The

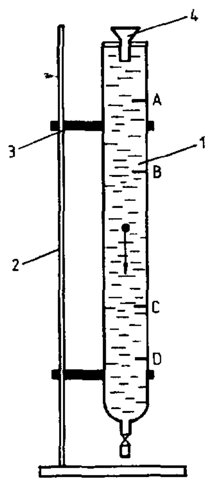

Fig. 2. Experimental installation: (1) column; (2) support; (3) clamps; (4) funnel.

Table 1
Moving duration of the urea particles (s)

No.	$C_A^0 = 0\%$			$C_A^0 = 3.75\%$			$C_A^0 = 7.5\%$			$C_A0 = 15\%$		
	A–B	B–C	C–D	A–B	B–C	C–D	A–B	B–C	C–D	A–B	B–C	C–D
1	2.5	5.5	3.5	3	5	3.7	3.2	7.8	7.4	3	5.1	3.6
2	2.85	5.25	4.5 5	3	7	6.5	2.3	5	3	3.2	5.4	3.5
3	2.85	6.15	4.8	3	6.5	4.7	3.2	6.1	4.6	3.4	6.8	4.7
4	2.9	5.6	4.6	3	4.8	3.5	3	6.8	4.4	2.5	5	3.1
5	2.8	5.8	4.2	2.2	4.1	2.6	2.5	4.7	3.1	2.7	5.7	3.7
6	2.4	4.3	3.2	2.5	4.4	3.3	2.5	5.8	3.7	2.7	4.7	3.3
7	2.5	5.5	3.6	2.7	5.1	3.9	2.4	4.3	3	2.7	5.3	3.3
8	2.1	4.3	2.6	2.2	4.8	3.4	2.4	5.1	3	–	–	–
9	2.6	4.2	4.9	2.4	4.4	3.2	2.3	4.9	3.1	–	–	–
10	2.4	5.5	4.5	2.5	4.9	3.7	–	–	–	–	–	–
11	2.5	5.6	5.5	2.2	5.1	3.5	–	–	–	–	–	–
12	2.2	4.4	3.2	–	–	–	–	–	–	–	–	–

Table 2
Physical properties at $T = 293$ K

Property		Value	Reference
Density of the particle/kg m^{-3}		1335	[17]
Density of the aqueous urea solution/kg m^{-3}	3.75%	1008.5	[17]
	7.50%	1019	
	15%	1040	
Viscosity of the aqueous urea solution/N s m^{-2}	3.75%	1.03×10^{-3}	[17]
	7.50%	1.06×10^{-3}	
	15%	1.12×10^{-3}	
Solubility of urea in water/%		551.50	[5]
Diffusion coefficient of urea in water/m^2 s^{-1}		1.18×10^{-9}	[5]
Diffusion coefficient of urea in aqueous urea solution/m^2 s^{-1}	3.75%	1.163×10^{-9}	Experimental values
	50%	1.091×10^{-9}	
	15%	1.046×10^{-9}	

distances measured from the upper margin of the column to the marks A, B, C and D are 0.20, 0.40, 0.78 and 0.93 m respectively.

The experiments were conducted at 293 K and atmospheric pressure using urea particles of various sizes and using as a solvent distilled water and urea solution with mass concentrations of 3.75, 7.5 and 15%. The urea particles were selected according to their size and shape, choosing only those which were closest to the spherical form. The granule diameter used for the experiments was in the range $(1.55–2.40) \times 10^{-3}$ m. Each selected particle was carefully introduced into the column so that at the contact with the liquid surface its velocity would be zero.

A large number of particles were used in the experiment, but only those which dissolved without rolling and touching the column walls were used in computation. The falling times corresponding to the sections A–B, B–C and C–D were measured using two chronometers. The data are presented in Table 1.

The values of the physical parameters used in computation are given in Table 2. The value of the diffusion coefficient of urea in aqueous urea solutions with the concentrations of 3.75, 7.5 and 15% was determined experimentally using the capillary method [18].

4. Results and discussions

Using the data in Table 1 and Eqs. (16) and (17) the mean velocity of the particles for the sections A–B and C–D was computed. The results obtained are presented in Table 3. Based on the velocity values, the mean diameters of the particles for sections A–B and C–D were computed. Eqs. (18)–(20) were used in the computations because the particles sediment in the intermediate region, $2 < Re < 500$. The results obtained are presented in Table 3. Using Eqs. (11)–(13) the dissolution rates of the urea particles in water and in urea solution with concentrations 3.75, 7.5 and 15% were computed (Table 3).

The individual mass transfer coefficient for dissolution was computed with Eq. (3). The results are presented in diagrams of $k_1 = f(Re)$. The Reynolds number was calculated with the relation:

Table 3
Dissolution rate of the urea particles

$C_A^0/\%$	$v_{01}/\mathrm{m\ s^{-1}}$	$v_{02}/\mathrm{m\ s^{-1}}$	$10^3\times D_1/\mathrm{m}$	$10^3\times D_2/\mathrm{m}$	$\Delta t/\mathrm{s}$	$10^3\times v_D/\mathrm{kg\ m^{-2}\ s^{-1}}$	$v_{0m}/\mathrm{m\ s^{-1}}$	$10^3\times D_m/\mathrm{m}$	Re_m
0	0.08	0.042	1.672	0.955	8.5	56.1	0.061	1.313	79.97
	0.07	0.032	1.485	0.752	8.95	54.43	0.051	1.118	56.93
	0.0701	0.031	1.485	0.731	9.975	50.22	0.0505	1.108	55.84
	0.068	0.0326	1.452	0.764	9.35	48.89	0.0503	1.108	55.84
	0.071	0.035	1.507	0.814	9.3	49.54	0.053	1.16	61.38
	0.083	0.046	1.727	1.034	7.1	64.9	0.064	1.38	88.16
	0.08	0.04	1.672	0.913	8.55	59.02	0.06	1.292	77.38
	0.09	0.05	1.859	1.111	6.65	74.8	0.07	1.485	103.74
	0.076	0.03	1.463	0.651	7.95	67.9	0.053	1.057	69.39
	0.083	0.033	1.584	0.708	8.95	65.05	0.058	1.146	82.34
	0.08	0.027	1.529	0.594	9.6	64.76	0.0535	1.061	70.34
	0.09	0.04	1.705	0.838	7.1	81.18	0.065	1.271	102.38
3.75	0.066	0.04	1.463	0.946	8.35	41.17	0.053	1.204	62.5
	0.066	0.031	1.463	0.756	10.35	45.36	0.048	1.109	52.16
	0.066	0.042	1.463	0.9867	8.05	41.53	0.054	1.224	64.72
	0.09	0.057	1.914	1.287	6.5	64.14	0.073	1.589	113.6
	0.08	0.045	1.584	0.96	7.3	56.81	0.0625	1.271	96.78
	0.074	0.038	1.485	0.828	8.9	49.06	0.056	1.156	78.65
	0.09	0.044	1.76	0.941	7.6	71.61	0.067	1.351	109.95
	0.083	0.046	1.49	0.979	7.2	60.95	0.064	1.309	101.78
	0.08	0.04	1.584	0.867	8	59.6	0.06	1.225	89.32
	0.09	0.042	1.76	0.904	7.95	71.57	0.066	1.332	106.81
	0.09	0.044	1.76	0.941	7.7	70.67	0.067	1.351	109.95
7.5	0.062	0.02	1.43	0.533	13.1	45.5	0.041	0.981	38.66
	0.086	0.05	1.903	1.188	8.8	54.02	0.068	1.54	100.66
	0.062	0.032	1.43	0.805	10	41.54	0.047	1.111	50.19
	0.076	0.05	1.716	1.188	7.5	46.81	0.063	1.452	87.93
	0.08	0.048	1.639	1.05	7.5	52.17	0.064	1.344	102.88
	0.08	0.04	1.639	0.895	8.9	55.56	0.06	1.267	90.67
	0.083	0.05	1.694	1.089	7	57.47	0.066	1.391	109.51
	0.083	0.05	1.694	1.089	7.8	51.57	0.066	1.391	109.51
	0.086	0.048	1.749	1.094	7.6	61.11	0.067	1.399	111.79
15	0.066	0.041	1.496	1.071	8.4	33.6	0.05	1.283	59.59
	0.062	0.042	1.529	1.094	8.75	33.02	0.052	1.311	63.3
	0.058	0.031	1.32	0.765	10.85	33.97	0.044	1.043	52.81
	0.08	0.048	1.749	1.122	7.8	53.44	0.064	1.435	105.75
	0.07	0.04	1.562	0.957	8.9	45.2	0.055	1.259	79.73
	0.07	0.045	1.562	1.06	7.7	43.32	0.057	1.311	86.03
	0.07	0.045	1.562	1.06	8.3	40.18	0.057	1.311	86.03

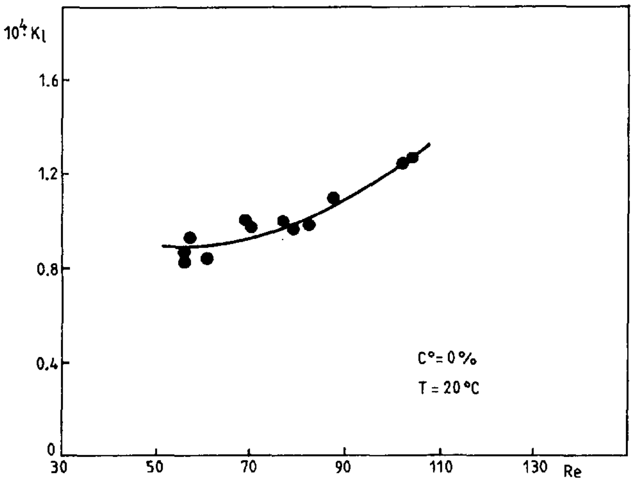

Fig. 3. Individual mass transfer coefficient as a function of the Reynolds number for urea particles dissolving in water.

$$Re=\frac{v_{0m}D_m\rho}{\eta} \tag{21}$$

where v_{0m} is the mean sedimentation rate

$$v_{0m}=\frac{v_{01}+v_{02}}{2}$$

and D_m is the particle mean diameter

$$D_m\frac{D_1+D_2}{2}$$

When computing the diameters D_1 and D_2 we took into account the fact that during free falling the particles change their shape owing to irregular surface dissolution, and so we introduced in the expression for D_1 and D_2 a shape coefficient of 0.77, the value being taken from the literature for a rounded particle [15].

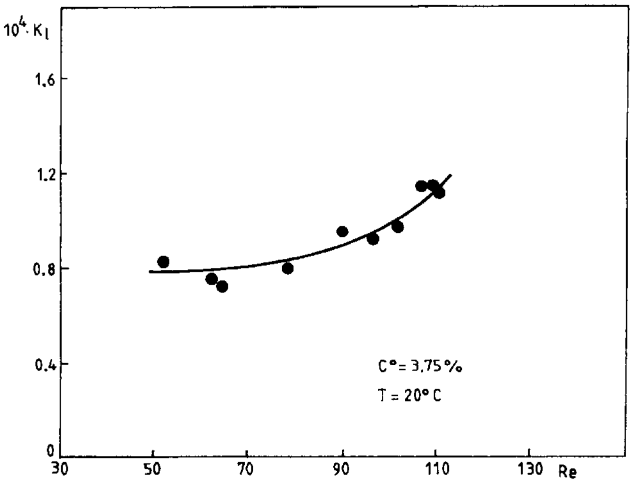

Fig. 4. Individual mass transfer coefficient as a function of the Reynolds number for urea particles dissolving in aqueous urea solution, $C_A^0 = 3.75\%$.

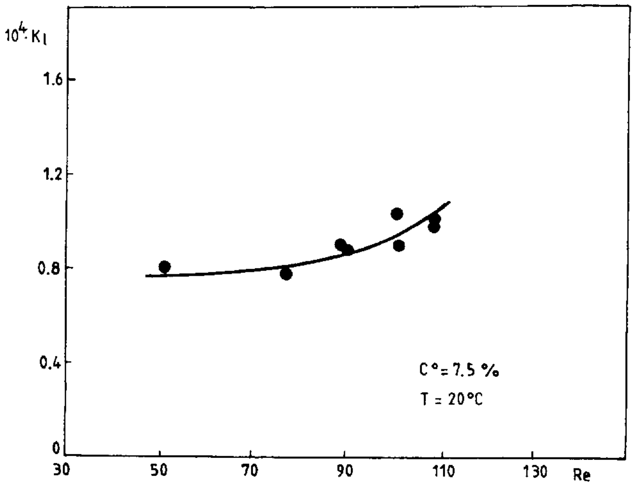

Fig. 5. Individual mass transfer coefficient as a function of the Reynolds number for urea particles dissolving in aqueous urea solution, $C_A^0 = 7.5\%$.

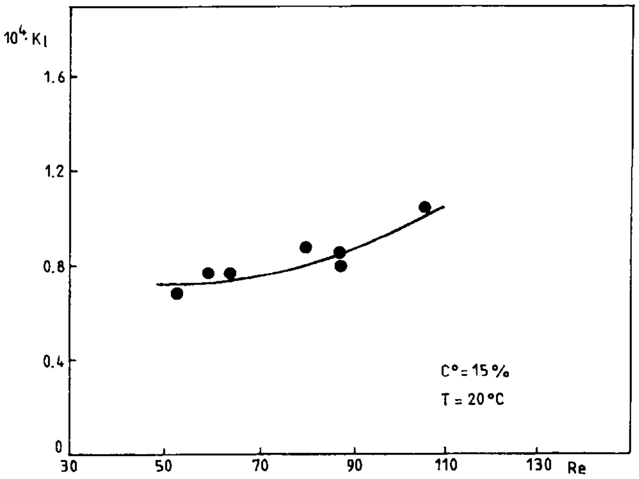

Fig. 6. Individual mass transfer coefficient as a function of the Reynolds number for urea particles dissolving in aqueous urea solution, $C_A^0 = 15\%$.

As may be seen from Table 3, the Reynolds number has a positive influence on the dissolution rate, i.e. for large values of the Reynolds number the dissolution rate has large values.

The urea concentration in the solution has a negative influence on the dissolution rate, an increase in the urea concentration causing a decrease in the dissolution rate.

The diagrams in Figs. 3–6 reveal a positive influence of the Reynolds number over the individual mass transfer coefficient of the urea particles dissolving in water as well as in aqueous urea solution. For large Reynolds numbers the influence is more obvious. This may be explained by the fact that for an increase in the Reynolds number the particle velocity increases and the thickness of the limiting diffusion layer around the particles decreases, at the same time intensifying the turbulent diffusion behind the moving particle. The value of the individual mass transfer coefficient determined by the proposed method represents a mean value on the particle surface.

As may be seen from Figs. 3–6, an increase in the urea concentration in the solvent leads to a decrease of the individual mass transfer coefficient as a result of the increase in urea solution viscosity, which leads to an increase of the limiting diffusion layer on the surface and a decrease of the turbulence behind the particle.

In order to verify the experimental results, the data have been represented in the form of logarithmic plots $Sh \cdot Sc^{-1/3} = f(Re)$, see Figs. 7 and 8. For comparison, a criterial equation [12] valid for the dissolution of the spherical particle surrounded by a forced flowing fluid was chosen.

$$Sh = 0.94 \cdot Re^{1/2} \cdot Sc^{1/3} \quad 2.3 < Re < 225 \tag{22}$$

The diagrams in Figs. 7 and 8 show that for large values of the Reynolds number (50–110) there is good agreement between the experimental values and those computed with Eq. (22). Larger differences are observed for urea dissolution in water. This may be explained by the greater deviation from the spherical shape in the case of particles dissolving in water (owing to the increased urea solubility in water).

Generally, for small values of the Reynolds number the agreement between the experimental results and the results obtained with Eq. (22) are better. The experimental values for urea dissolution in water are approximately 20% larger than the values obtained with Eq. (22), and 14% larger for urea dissolution in aqueous urea solution.

5. Conclusions

In this paper a new method for the experimental determination of the individual mass transfer coefficient and the solid dissolution rate has been presented.

This method implies an experiment conducted individually for several particles which fall freely in a solvent.

The proposed method enables the determination of the individual mass transfer coefficient and of the dissolution rate by measuring the falling time corresponding to three sections, without measuring the initial particle dimensions.

Based on this method the dissolution rate and the individual mass transfer coefficient were determined for urea particles

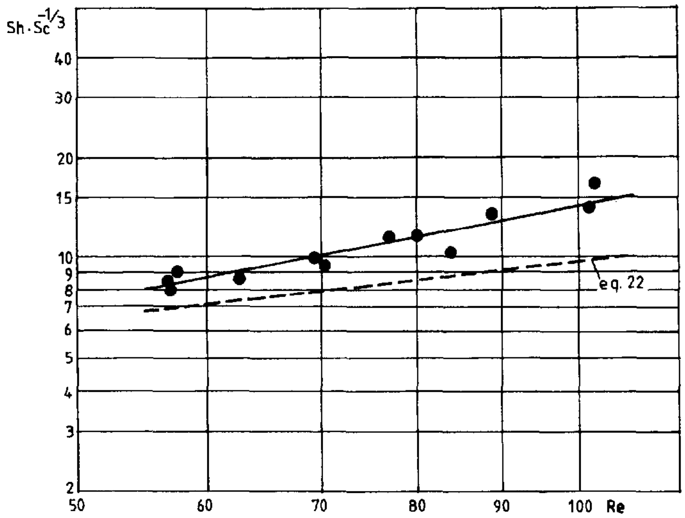

Fig. 7. Dependence of Sherwood number on Reynolds number for urea particles dissolving in water.

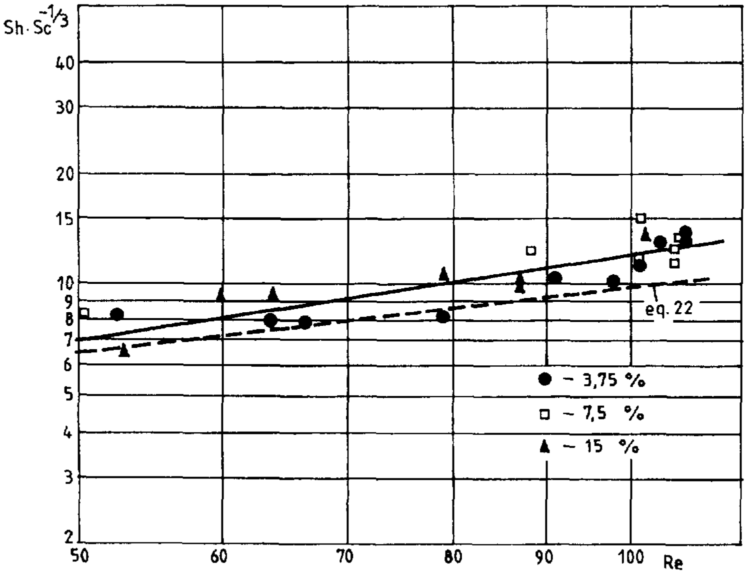

Fig. 8. Dependence of Sherwood number on Reynolds number for urea particles dissolving in aqueous urea solutions.

dissolving in water or in aqueous urea solutions with mass concentrations of 3.75, 7.5 and 15% at 293 K and atmospheric pressure. The influence of the Reynolds number and of the solution concentration on the dissolution rate and the individual mass transfer coefficient was also studied.

The experimental results were compared to those found in the literature using for this purpose a criterial equation valid for forced convection ($2.3 < Re < 225$).

Good agreement between the experimental results and the results obtained using the criterial equation was observed for $50 < Re < 110$.

The results obtained show that the proposed method is better suited for substances with low solubility.

Appendix A. Nomenclature

a	Parameter in Eq. (8) (dimensionless)
C_{A}^{0}	Concentration of component A (urea) in solution (kg m^{-3})
C_{A}^{s}	Solubility of component A in water (kg m^{-3})
D	Particle diameter at time t (m)
D_1	Particle diameter when falling from A to B (m)
D_2	Particle diameter when falling from C to D (m)
D_{m}	Particle mean diameter (m)
g	Gravitational acceleration (m s^{-2})
h_1, h_2	Distances corresponding to sections A–B, C–D (m)
k_1	Individual mass transfer coefficient (m s^{-1})
m_{A}	Particle mass at moment t (kg)
n	Parameter in Eq. (8) (dimensionless)
Re	Reynolds number (dimensionless)
S	Exterior area of the particle surface at time t (m^2)
t	Time (s)
t_1, t_2, t_3	The falling time corresponding to sections A–B, C–D and B–C (s)
v_0	Sedimentation rate (m s^{-1})
$v_{0\mathrm{m}}$	Mean sedimentation rate (m s^{-1})

v_{01}, v_{02}	Sedimentation rate corresponding to the distances h_1 and h_2 (m s^{-1})
V	Particle volume at time t (m^3)

A.1. Greek letters

η	Viscosity of the aqueous (urea) solution (N s m^{-2})
ρ_s	Density of the particle (kg m^{-3})
ρ	Density of the aqueous (urea) solution (kg m^{-3})
ζ	Friction factor (dimensionless)

References

[1] D.M. Levins and J.R. Glastonbury, *Trans. Inst. Chem. Eng., 50* (1972) 132.

[2] N. Yutani, N. Ototake and L.T. Fan, *Ind. Eng. Chem. Res., 26* (1987) 343.

[3] A. Lakota and J. Levec, *AIChE J., 36* (1990) 1444.

[4] S. Bon-Long, C. Laguerie and I.P. Couderc, *Chem. Eng. Sci., 33* (1978) 813.

[5] A. Tamir and O. Falk, *Ind. Eng. Chem. Res., 27* (1988) 1930.

[6] V.G. Levici, *Fiziko-Himiceskaia Gidrodinamica*, Gosudarstvenoe izdatelstvo fizico-mathematicskoi literaturi, Moskow, 1959.

[7] A. Le Lan, H. Gibert and H. Angelino, *Chem. Eng. Sci., 27* (1972) 1979.

[8] R.G. Rice and P.J. Jones, *Chem. Eng. Sci., 34* (1979) 847.

[9] S. Petrescu and R.Z. Tudose, *Rev. Chim, 39* (1988) 876.

[10] S. Petrescu and R.Z. Tudose, *Rev. Chim, 40* (1989) 237.

[11] S. Petrescu, *Rev. Roum. Chim., 34* (1988) 1799.

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

[13] M. Venkatesk, K.A. Narayan and R.P. Chhabra, *Chem. Eng. Commun., 130* (1994) 181.

[14] G. Le Palec and M. Daguenet, *Int. Chem. Eng., 30* (1990) 683.

[15] R.Z. Tudose et al., *Procese, Operatii, Utilaje in Industria Chimică*, Editura Didactică si Pedagogică, Bucuresti, 1977.

[16] R.B. Bird, W.E. Stewart and N. Lighfoot, *Transport Phenomena*, Wiley, New York, 1960.

[17] *Spravochnik Azotchika*, Vol. 2, Izd. Khimia, Leningrad, 1969.

[18] C.I. Luk, L. Nanis and M. Litt, *Ind. Eng. Chem. Fundam., 14* (1975) 92.