Gas-Solid Mass Transfer in a Jetloop Reactor

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Recycle reactors have been used as laboratory tools for catalyst testing and kinetic studies for years (Berty, 1984). Weekman (1974) discussed laboratory reactors and their limitations. Berty (1984) indicated that recycle reactors are superior for studying catalyst kinetics. Reactors such as those of the spinning basket type (Tajbal et al., 1966; Brisk et al., 1968) have been shown to have relatively poorer mass-transfer rates (Caldwell, 1982) than those of the Berty-type (Berty, 1974; Caldwell, 1983). Nevertheless, with careful design the performance of spinning basket reactors can be significantly improved (Hidajat et al., 1987; Carberry and Martinez, 1995). For kinetic studies in such reactors, the following assumptions are usually made: (1) the reactor is well mixed; (2) conversion across the catalyst bed is differential and the rate can be based on the exit gas concentration; and (3) heat- and mass-transfer rates are high enough to be neglected.

The first assumption can be verified by tracer studies. Conversion over the catalyst bed can usually be estimated if the flow pattern is known (recycle ratio) and this would generally follow from the tracer studies. The measurement of heat- and mass-transfer rates are not easily achieved and in many cases cannot be neglected. In many reactors the influence of mass transfer can be determined by varying the stirrer speed, but this however does not guarantee that the mass-transfer rate is sufficiently high. Many criteria have been developed to predict operating conditions which will not be limited by heat and mass transfer rates (Luft and Herbertz, 1969).

This article presents results of a study of the mass-transfer rates in a new internal recycle reactor, the jetloop reactor (JLR) as proposed by Luft (Shermuly and Luft, 1977). This reactor has been used, among other things, to study selective oxidation on palladium alumina catalysts (Haas and Gaube, 1989). The residence time studies (Möller et al., 1995) in this reactor have shown that it behaves as a well-mixed reactor, with recycle ratios easily exceeding the minimum value of 20 required for CSTR behavior (Gillespie and Carberry, 1966). In this system, however, the influence of mass-transfer rate on the reaction rate cannot be determined by varying the impeller speed, as the recycle ratio is dependent on the flow rate and must therefore be measured independently. The objective of this work was to symmetrically investigate the effect of particle size and flow rate on the mass-transfer rates to catalyst particles with a view to using this reactor for kinetic studies.

Experimental Studies

The JLR in Figure 1 is discussed in detail by Möller et al. (1995). The JLR operates on the principle that momentum transferred to the bulk fluid by a high-pressure, high velocity jet causes the gas to recycle because of its novel construction. Recycle ratios up to 70 have been measured (Möller et al., 1995). For the mass-transfer studies three types of packing were used: (1) 3.8 mL of 6-mm glass balls (approximately 20 balls); (2) 5.6 mL of 4-mm glass balls (approximately 100 balls); and (3) 6 mL of 2-mm glass balls.

The recycle ratio was measured independently for each of these systems, using methods outlined previously (Möller et al., 1995). For the mass-transfer measurements, a naphthalene particle was placed randomly among the glass beads. The naphthalene particle used had an equivalent spherical diame-

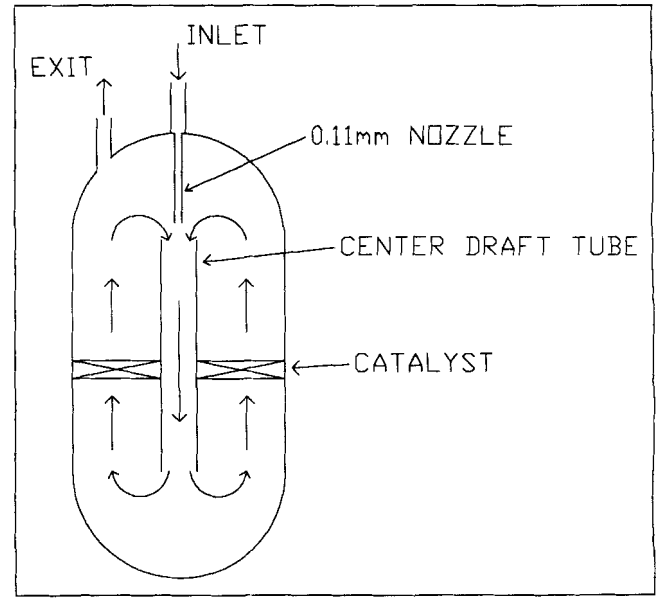

Figure 1. Jetloop reactor.

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ter of 5.8 mm based on equivalent surface area (as determined from micrometer measurements). The naphthalene concentration was continuously monitored as a function of the flow rate using a flame ionization detector. The flow rate leaving the reactor was measured using a soap bubble meter. The naphthalene concentration was less than 0.1% in the exit gas stream and would thus not significantly effect the total flow rate or the properties of the N_2 carrier gas, nor cause excessive mass loss of the naphthalene particle for the duration of an experiment.

Results and Discussion

The basic mass-transfer equation

$$N = FC = K_x a(C_s - C) \tag{1}$$

can be manipulated, by noting that the detector response R for a gas concentration C (mol/mL) is given by

$$\frac{R_s - R_0}{R - R_0} = \frac{C_s}{C} \tag{2}$$

and making use of

$$\tau = \frac{V}{F} \tag{3}$$

to yield

$$\frac{1}{R - R_0} = \left(\frac{V}{K_x a(R_s - R_0)}\right)\frac{1}{\tau} + \frac{1}{R_s - R_0} \tag{4}$$

Equation 2 is normalized with respect to the concentration on the naphthalene surface C_s, i.e., saturation pressure. Mass transfer in packed beds can be represented by the correlation of Wakao and Funazkri (1978)

$$Sh = 2.0 + 1.1\, Sc^{1/3} Re_p^{0.6} \tag{5}$$

Since the jetloop system operates at constant temperature and pressure, K_x (cm/s) is only a function of velocity. The velocity (Eq. 6) is proportional to the reciprocal residence time if it can be assumed that the recycle ratio is constant

$$u = \frac{(1 + RR)V}{\tau S} \tag{6}$$

Grouping all the constant terms in Eq. 5 and rearranging, the mass-transfer coefficient can then be represented by

$$K_x = A + B\left(\frac{1}{\tau}\right)^{0.6} \tag{7}$$

where

$$A = \frac{2D_m}{d_p} \tag{8}$$

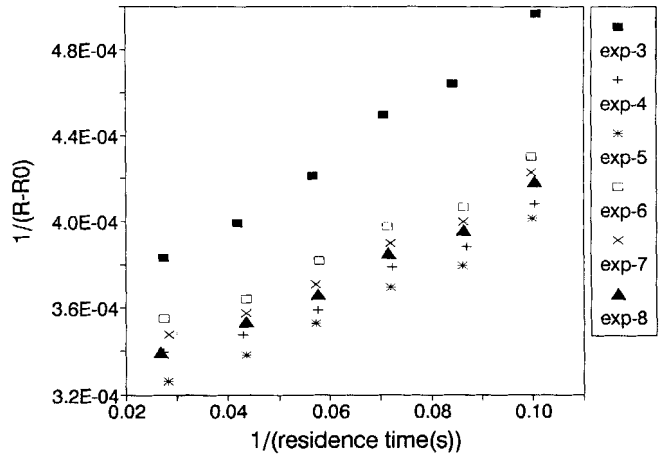

Figure 2. Experimental detector response as a function of reciprocal reactor residence time for 2-mm-dia. glass beads.

$$B = 1.1\frac{(1 + RR)V}{S}\frac{D_m}{d_p} Sc^{1/3}\left[\frac{\rho d_p}{\mu}\right]^{0.6} \tag{9}$$

Note that B is not necessarily constant as the recycle ratio will vary with flow rate at large residence times. This expression may be inserted into Eq. 4 to yield a general expression for the mass-transfer rate of naphthalene

$$\frac{1}{R - R_0} = \frac{1}{R_s - R_0}\left[\left(\frac{V}{a\left(A + B\left[\frac{1}{\tau}\right]^{0.6}\right)}\right)\left[\frac{1}{\tau}\right] + 1\right] \tag{10}$$

By using nonlinear regression techniques, the three parameters of Eq. 10, A, B, and R_s may be determined. Thus it is possible to determine the mass-transfer coefficients without having to determine the saturation vapor pressure of naphthalene, as shown previously (Caldwell, 1983). In the case of zero flow, saturation should be achieved, and thus extrapolation to infinite residence time should yield R_s.

Figure 2 shows the results obtained for six experiments done using the 2 mm glass balls. The differences in magnitude are due to variations in the attenuation of the FID. These data were used to determine the constants of Eq. 10. In all cases B was found to be zero, indicating that data was linear and that the mass-transfer coefficient was independent of flow rate or residence time. Figure 3 shows the typical linear fit of the experimental data. Table 1 shows the mass-transfer coefficients obtained.

The fact that K_x was invariant with respect to flow rate is an anomalous result. Recycle ratios estimated from residence time distribution studies on the equivalent glass beads enable the calculation of particle Reynolds numbers (Re_p) ($\rho u d_p/\mu$). These were found to lie between 10 and 250, and thus the jetloop operates in a regime between laminar and turbulent flow. Using Eq. 5, mass-transfer coefficients may also be estimated based on glass bead particle diameters. The results for a typical case are shown in Table 2. Clearly the mass-transfer

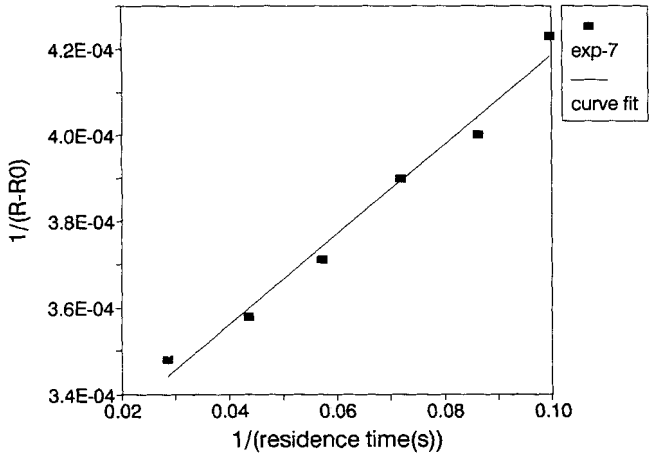

Figure 3. Curve fit of the experimental detector response as a function of reciprocal reactor residence time for 2-mm-dia. glass beads.

coefficients estimated from Eq. 5 show variation with flow rate, but underestimate K_x by a factor of 2 and more. This suggests that mass transfer is enhanced in the JLR, especially at low flow rates. There may be two reasons for this: first, the reactor will be highly turbulent due to the induced turbulence of the jet and the inherent mixing which occurs in the system improving mass transfer and possibly eliminating influence that flow rate might have on K_x; secondly, Eq. 5 strictly does not apply to packed beds in which the bed length is only a few bead diameters.

Table 1 also shows that there is an increase in the mass-transfer coefficient with a reduction in glass bead size. Smaller glass beads would induce better mixing around the larger particle and thus increase mass-transfer rates. When the mass-transfer coefficient is independent of flow rate, Eq. 5 predicts that K_x should be proportional to the inverse particle diameter. This is in fact observed in Figure 4 where K_x is plotted as a function of $1/d_p$ (particle diameter, cm). This confirms that the mass-transfer coefficient in the jetloop reactor are independent of flow rate and dependent on particle size for the conditions studied here.

Comparison with the literature is difficult, because most recycle reactors (or mixed reactors) have impellers which allow variation of the recycle ratio (velocity of the gas through the bed) independently of the residence time; thus data in the literature is usually given as a function of impeller speed. In the JLR the recycle ratio can only be varied by varying the

Table 1. Mass-Transfer Coefficients: 20°C, 121 kPa (Abs.)

Exp. No.	Particle Size mm	K_x cm/s
1	6	6.8
2	4	7.4
3	2	8.8
4	2	9.9
5	2	9.4
6	2	9.1
7	2	10.2
8	2	8.9
Avg. (2 mm)	2	9.4 ± 0.6

Table 2. Mass-Transfer Coefficients for the 2-mm Glass Beads Estimated from Eq. 5

Residence Time s	K_x cm/s
35.1	2.4
22.9	3.3
17.4	3.8
13.9	4.4
11.6	4.8
10.0	5.2
$K_x = 9.4$ cm/s obtained from experiment	

residence time. Table 3 summarizes the present results and those from other studies. The mass-transfer coefficient in the JLR compares very well with that obtained in a Berty-type reactor and both are clearly superior to spinning basket type reactor tested by Caldwell (1982). This is expected as the former two reactors are internal recirculation reactors with high gas velocities over the catalyst. It has been shown by Carberry and Martinez (1995) that a well-designed spinning basket reactor ($K_x = 7$ cm/s at 2,000 rpm) performs as well as the JLR and Berty-type reactor. Superficial gas velocities calculated from the estimated recycle ratios varied between 10 and 70 cm/s.

The mass-transfer coefficient in the spinning basket and Berty reactors are independent of flow rate above minimum impeller speeds, which is also observed in the JLR. This reactor thus offers similar performance to a Berty-type reactor without the need for any moving parts. Disadvantages of the JLR are that high jet head pressures complicate the reactor feed system, and that flow rates can only be varied over a limited range without having to change the jet diameter.

Conclusions

The mass-transfer coefficient in a jetloop reactor has been shown to be similar to that of a Berty-type reactor. This reactor is well suited for the measurement of reaction rates. The mass-transfer coefficient was found to be independent of flow rate and was a linear function of the reciprocal particle diameter.

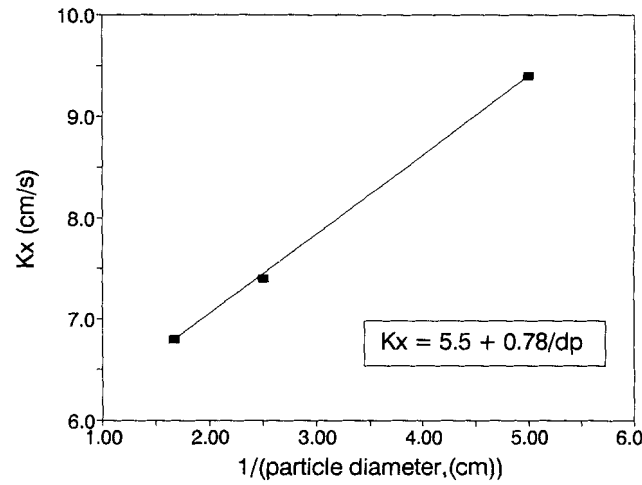

Figure 4. Variation of mass-transfer coefficient with reciprocal particle diameter.

Table 3. Comparison of Mass-Transfer Coefficients with the Literature

Reactor Type	K_x, cm/s	Impeller RPM	Conditions
Spinning basket (Caldwell, 1982)	0.3–1.2	500–2,000	4-mm cylinders, naphthalene/air, 53°C, 1.16 kPa
Berty (Caldwell, 1983)	2.5–9.5	200–2,000	4-mm cylinders, naphthalene/air, 23°C, 0.87 kPa
This work	7.4	—	4-mm spheres, naphthalene/N_2, 20°C, 121.3 kPa

Notation

a = surface area for mass-transfer, cm^2
D_m = molecular diffusion constant, cm^2/s
F = gas flow rate, mL/s
N = mass-transfer rate, mol/s
R_0 = FID zero response
S = cross-sectional area of the annulus, cm^2
Sc = particle Schmidt number, $\mu/\rho D_m$
Sh = particle Sherwood number, $K_x d_p/D_m$
u = superficial gas velocity, cm/s
V = reactor volume, mL

Greek letters

μ = viscosity, g/cm·s
ρ = density, g/cm^3
τ = reactor residence time, s

Literature Cited

Berty, J. M., "Reactor for Vapour Phase Catalytic Studies," *Chem. Eng. Prog.*, **70**(5), 78 (May, 1974).

Berty, J. M., "20 Years of Recycle Reactors in Reaction Engineering," *Plant/Op. Prog.*, **3**(3), 163 (July, 1984).

Brisk, M. L., R. L. Day, M. Jones, and J. B. Warren, "Development of a Stirred Gas-Solid Reactor for the Measurement of Catalyst Kinetics," *Trans. Instn. Chem. Engrs.*, **46**, T3 (1968).

Caldwell, L., "Gas Solid Mass Transfer in a Spinning Catalyst Basket Reactor," *Appl. Catal.*, **4**, 13 (1982).

Caldwell, L., "An Improved Internal Gas Recirculation Reactor for Catalytic Studies," *Appl. Catal.*, **8**, 199 (1983).

Carberry, J. J., and E. Martinez, private communication (1995).

Gillespie, B., and Carberry, J. J., "Influence of Mixing on Isothermal Reactor Yield and Adiabatic Reactor Conversion," *I&EC Fund.*, **5**(2), 165 (May, 1966).

Haas, T., and J. Gaube, "Investigation of the Selective Hydrogenation of 1,3-cyclooctadiene over Pd/Al_2O_3 Catalyst in the Gas Phase," *Chem. Eng. Technol.*, **12**, 45 (1989).

Hidajat, K., F. J. Aracil, J. J. Carberry, and C. N. Kenney, "Laboratory Catalytic Studies: The Role of Transport Phenomena," *J. Catal.*, **105**, 245 (1987).

Luft, G., and H. A. Herbertz, "Zur Auslegung von Kreislaufapparaturen für Reaktionskinetische Messungen," *Chem.-Ing.-Tech.*, **41**(11), 667 (1969).

Möller, K. P., R. J. Becker, J. C. Q. Fletcher, and C. T. O'Connor, "Residence Time Distributions in a Jetloop Reactor," *Chem. Eng. Comm.*, **137**, 111 (1995).

Shermuly, O., and G. Luft, "Untersuchung der Niederdruck-Methanol-Synthese im Triebstrahlreaktor," *Chem.-Eng.-Tech.*, **49**, 907 (1977).

Tajbal, D. G., J. B. Simons, and J. J. Carberry, "Heterogeneous Catalysis in a Continuous Stirred Tank Reactor," *I&EC Fund.*, **5**(2), 171 (1966).

Wakao, N., and T. Funazkri, "Effect of Fluid Dispersion Coefficients on Particle-to-Fluid Mass Transfer Coefficients in Packed Beds," *Chem. Eng. Sci.*, **33**, 1375 (1978).

Weekman, V. W., "Laboratory Reactors and their Limitations," *AIChE J.*, **20**(5), 833 (Sept., 1974).

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