Liquid-Solid Mass Transfer in Inverse Fluidized Bed

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Conventional fluidized beds, used widely in practice and studied intensively in the last few decades, have the following common characteristics. The solid particles have higher density than those of the fluidizing liquid and/or gas, and the bed is expanded by the upward flow of the continuous fluid. A new mode of three-phase fluidization proposed has the solid phase with a density lower than that of the downward-flow continuous liquid phase. It is called "three-phase inverse fluidization."

There are only a few publications concerning three-phase inverse fluidization (Werner and Schugerl, 1980; Fan et al., 1982a,b). Only the hydrodynamics of this new process has been studied so far. There is no information available yet concerning mass transfer between the solid and liquid phases in the inverse fluidized beds (Muroyama and Fan, 1985). Some differences in mass transfer between inverse fluidization and classic fluidization are due primarily to the different directions of the gas and liquid and the different inertial effects of the particles. The particles in the inverse fluidized bed have smaller mass and smaller inertia than those used in the upflow bed.

The mass transfer rate as a function of the parameters of the fluidized bed is given usually by the following correlations: Sherwood (*Sh*) and Schmidt (*Sc*), Galileo (*Ga*), density (*Mv*) and Reynolds (*Re*) numbers. A good review on the mass transfer correlation for classical liquid fluidized beds is presented by Tournié et al. (1977). In our view, the most general and precise correlations are those proposed by Calderbank (1967) based on the experimental data of dissolution of solid particles and those derived originally for the description of slurry reactors and applied by Lee et al. (1974) to fluidized beds.

$$Sh = 0.31\ (GaMvSc)^{1/3} \tag{1}$$

and the correlation of Tournié et al. (1977):

$$Sh = 0.24\ Ga^{0.323} Mv^{0.3} Sc^{0.4} \tag{2}$$

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One of the most effective methods for determining the mass transfer between fluidized solid particles and liquid is the electromechanical method (Berger and Ziai, 1983; Nikov and Delmass, 1987). It is based on the diffusion-controlled cathode reduction of ferricyanide ions on a fixed sphere. The mass transfer rate can be determined by the current intensity from the liquid to the electrode. Using the data obtained by this method, the following correlation for the liquid-solid mass transfer was proposed by Nikov and Delmas (1987):

$$Sh = 0.34\ (GaMvSc)^{1/3} \tag{3}$$

The accuracy and applicability of this equation in classical liquid-solid fluidized beds were pointed out recently by Arters and Fan (1990).

Inverse fluidized beds are used as bioreactors when the support particles, on which microorganisms are fixed, have a density lower than that of the fermentation broth (usually around 1 Mg/m^3). The inverse fluidized-bed biofilm reactor is up to 15 times more effective than the airlift apparatus (Nikolov and Karmanev, 1987) and allows for effective control of the biofilm thickness (Karamanev and Nikolov, 1988). Therefore, interest in the inverse fluidization has been growing. However, due to a lack of data on mass transfer in inverse fluidized beds, relations developed for the classic fluidized bed have been used for modeling mass transfer (Chavarie and Karamanev, 1986). This note offers mass transfer data of two-phase inverse fluidized beds and mathematically describes this process.

Materials and Methods

Electrochemical method for liquid-solid mass transfer measurements

The method is based on the electromechanical reduction of ferricyanide ions at the surface of a spherical cathode. The electrolyte (liquid phase) used was a 5.10^{-3} M equimolecular solution of potassium ferri- and ferrocyanide with 0.5 M sodium hydroxide. The cathode was a 3-mm brass sphere covered with a 5-μm-thick gold layer; the anode was a nickel plate; a

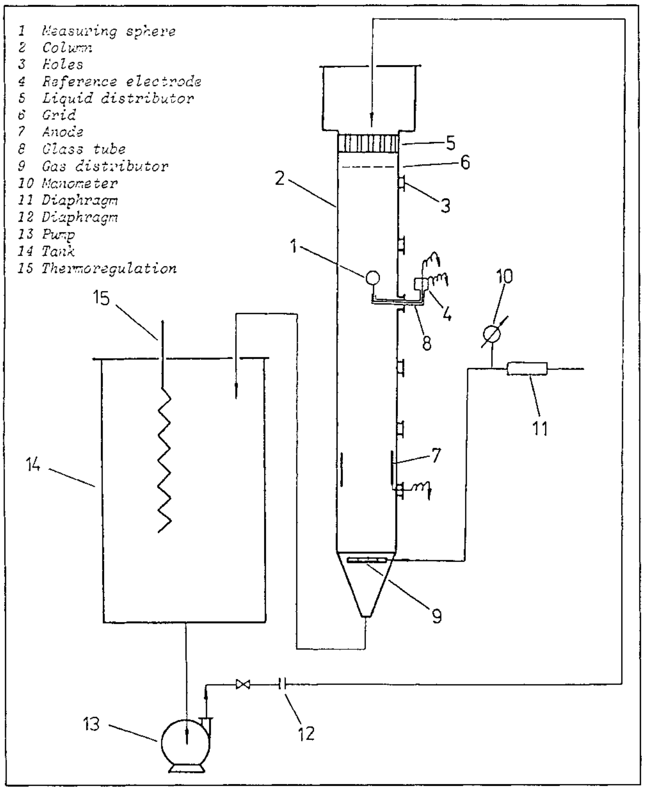

Figure 1. Experimental setup.

saturated calomel electrode was used as a reference. The mass transfer rate between the cathode and the liquid is the same as that between the particles of the bed and the liquid, and is related to the limiting current intensity by the following equation:

$$K = I/(n_e F S C_A) \tag{4}$$

The output signal was connected to a computing microprocessor-controlled multimeter SOLATRON. The signal was integrated (50-200 measurements for each experimental point with a time step of 1 s), and the standard deviation and variation of the signal were computed. The average standard deviation was found to be about 1.5% of the measured value.

Equipment for the inverse fluidization

The inverse fluidized bed consisted of a glass column with an internal diameter of 100 mm (Figure 1). The liquid was delivered by a glass pump to the distributor which was placed at the top part of the column. The bed was kept submerged in the liquid by a grid with 2-mm holes, mounted below the liquid distributor. The cathode was held at the axis of the column by a glass tube. To avoid interferences of the tube on the hydrodynamics around the electrode, the latter was held 30 mm apart from the end of the tube. It was fixed by an isolated wire, 250-μm-thick, which was also used as an electrical conductor. The reference electrode was mounted in the outside reservoir of the cell, that was filled with electrolyte, thus forming an electrolytic bridge. The anode—hollow cylinder of nickel—was placed at the bottom part of the column. The temperature of the liquid was kept at 25 ± 0.2°C by pumping cooling water through a coil placed in the tank.

Table 1. Properties of the Liquids Used as Fluidizing Agents

Conc. of PEG 10000 wt. %	ρ_L kg/m^3	μ_L mPa·s	$D\cdot10^{10}$ m^2/s
0	1,025	1.00	10.4
1.9	1,030	1.55	6.90

Physical properties of the inverse fluidized bed

The physical properties of the liquid phase, density, viscosity, diffusion coefficient, and the concentration of the electrolyte were measured. The density was determined by a pycnometer. The viscosity of the liquid phase was measured by a rotational viscometer, RHEOTEST-2. The viscosity of the electrolyte used was varied by addition of polyethylene glycol (PEG) 10000. All the liquids used were found to be Newtonian.

The effective diffusion coefficients were determined by an original electrochemical method using a rotating Geiss electrode (Nikov et al., 1987). The same method was used to determine the concentration of the potassium ferricyanide. The characteristics of the liquids, used in the experiments, are summarized in Table 1.

The solid phase of the inverse fluidized bed was granulated polymeric materials. Two different types of polymers were used. The expanded polystyrene (styrofoam) has a spherical shape. It is very convenient for inverse fluidized-bed experiments because its density can be changed easily between 50 and 1,000 kg/m^3 by heat expansion. The sizes available are between 2 and 10 mm. This material is used in the inverse fluidized-bed biofilm reactor. The styrofoam is inert for most biological environments and the electrolytic solutions used in this study. It has closed pores, and therefore its density during a long time stay in liquid remains constant. The second solid material was polyethylene. It has a constant density. Its shape is close to the spherical one. The characteristics of the particles used in this study are given in Table 2.

Experimental Results

The mass transfer coefficients from the liquid to the solid phases were studied as a function of the superficial liquid velocity for different types of liquid and solid phases. Liquids with different viscosities and effective diffusion coefficients were used. The dependence of the mass transfer coefficient on the superficial liquid velocity for liquids with different viscosities is shown in Figure 2. The effect of the superficial velocity is negligible, while an increase in viscosity causes a significant decrease in the mass transfer rate. The solid particles used in this experiment had a diameter of 3.6 mm and a density of 677 kg/m^3 ($\rho_L - \rho_s$ = 348 kg/m^3). Figure 2 also presents

Table 2. Properties of the Fluidized Particles

Material	Density kg/m^3	Size mm
Styrofoam	80	4.1
"	90	7.1
"	360	4.5
"	550	2.2
"	677	3.6
Polyethylene	930	3.5

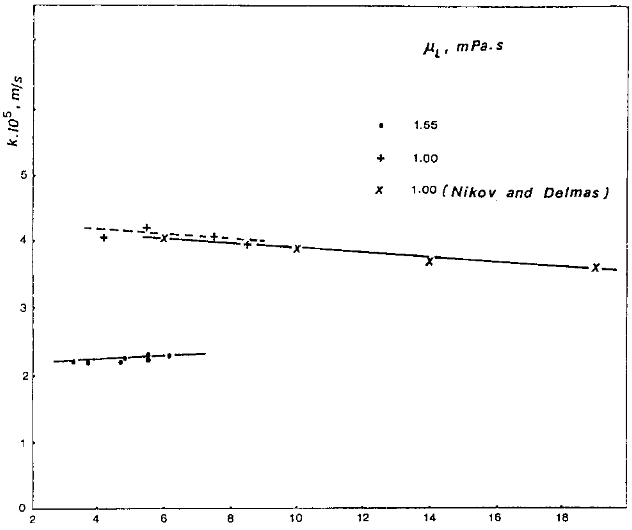

Figure 2. Effects of viscosity and superficial liquid velocity on mass transfer coefficient.

the data of Nikov and Delmas (1987) on mass transfer in conventional liquid-solid fluidized bed with a similar difference of densities between the liquid and solid phases ($\Delta\rho$ = 315 kg/kg/m^3). The particle diameter was 10 mm; as already known, however, this parameter has no effect on the mass transfer rate in classical fluidized beds (Nikov and Delmas, 1987; Arters and Fan, 1990). Mass transfer was determined by the same method as in this work.

The effect of the superficial liquid velocity and the particle density on the mass transfer coefficient is shown in Figure 3. While the mass transfer remains almost unaffected by the velocity, an increase in the particle density (decrease of $\Delta\rho$) decreases the mass transfer rate. The particle sizes with different densities were similar.

Dependency of the liquid velocity and the particle size at a constant density on the mass transfer coefficient was studied (Figure 4). There is a small effect of the particle diameter, which is within 11%. Additional experiments should be performed for a more detailed examination of this effect.

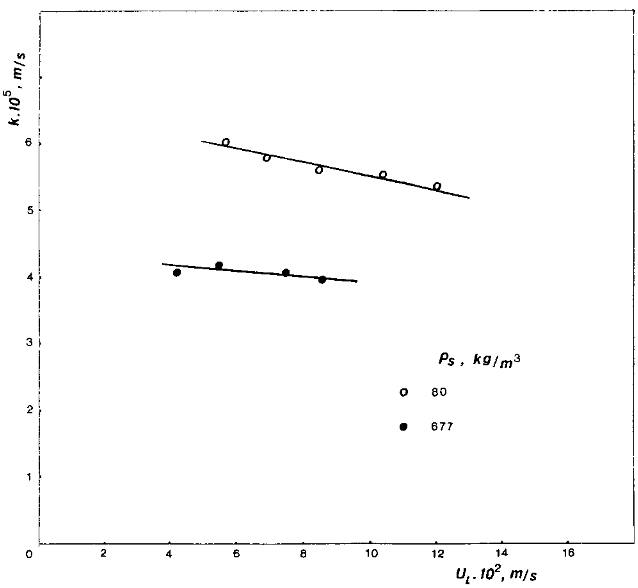

Figure 3. Effects of particle density and superficial liquid velocity on mass transfer coefficient.

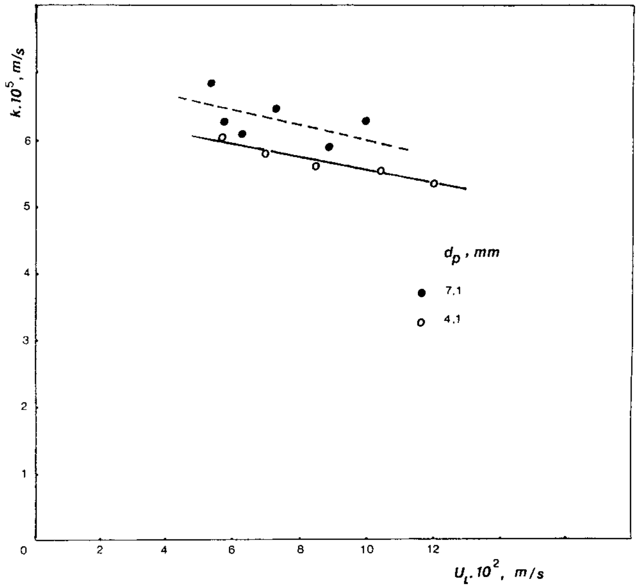

Figure 4. Dependence of particle size and superficial liquid velocity on mass transfer coefficient.

Discussion

The study of mass transfer in an inverse fluidized bed, liquid-solid case, raises the following question: What is the difference, if any, between the mass transfer characteristics of this system and those of the classical fluidized bed? The main difference is that the particles in the inverse bed are much lighter. If one compares a particle of an inverse fluidized bed with the same $\Delta\rho$ and size with that of an upflow bed fluidized by identical liquids, the only difference is in the mass of the particles, which becomes greater as $\Delta\rho$ increases. This results in a more intensive movement of the inverse fluidized bed, because of smaller inertia of the light particles. This effect was observed visually and explained earlier (Karamanev, 1988). On the one hand, it could decrease the mass transfer rate because particles move with the turbulent pulsations of the liquid, but on the other, particle-particle collisions increase, intensifying the mass transfer (Valentin et al., 1978).

It has long been established that there is no effect of the liquid superficial velocity on the mass transfer rate (Tournié et al., 1977; Nikov and Delmas, 1987; Nikov, 1987). There is also no effect of the particles size. An increase in $\Delta\rho$ intensifies the mass transfer. The larger the liquid viscosity, the smaller is the mass transfer rate. These dependencies are the same, at least qualitatively, in the inverse fluidized bed. This is why the correlations used for the description of the mass transfer in conventional fluidized beds are used for inverse fluidized beds. Since the Calderbank equation (Eq. 1) is general enough and describes the mass transfer in stirred tanks as well as in classical liquid-fluidized beds with a slight increase in the correlation constant (Nikov and Delmas, 1987), we decided to use the same type of correlation for the inverse fluidization. Values

Table 3. Deviations Between Published Correlations and Experimental Data

Equation	Ref.	Avg. Dev. %	Max. Dev. %	% of Points over Calc.
$Sh = 0.28\ (GaMvSc)^{0.33}$	This Study	9.3	22	37
$Sh = 0.272\ Re^{-0.07}Ga^{0.36}Mv^{0.37}Sc^{0.33}$	*	10	32	50
$Sh = 0.34\ (GaMvSc)^{0.33}$	**	23	29	12
$Sh = 0.245\ Ga^{0.323}Mv^{0.3}Sc^{0.4}$	†	33	80	0
$Sh = 0.253\ Re^{0.004}Ga^{0.319}Mv^{0.299}Sc^{0.4}$	†	33	79	0
$Sh = 0.228\ Ga^{0.323}Mv^{0.3}Sc^{0.4}$	‡	24	67	12

*Riba and Couderc (1980); **Nikov and Delmas (1987); †Tournié et al. (1977); ‡Arters and Fan (1986)

of the constants were determined by a nonlinear regression. The resulting equation is:

$$Sh = 0.28\ (GaMvSc)^{0.33} \qquad (5)$$

The primary conclusion is that the power of this equation is the same as that for the cases of the classical fluidized bed and the slurry reactor. The coefficient (0.28) is close to that for the other two systems, equal to 0.34 (Nikov and Delmas, 1987) and 0.31 (Calderbank, 1967), respectively. Some decrease is noted of the constant in the transition from upflow through zero to downflow liquid superficial velocity.

Table 3 compares the deviations between our experimental data and the predictions of Eq. 5 or using other equations proposed for the determination of the liquid-solid mass transfer in classical fluidized beds. The correlation proposed describes the mass transfer in an inverse fluidized bed better than those for classic fluidized beds. The mass transfer coefficients calculated by the equation of Riba and Couderc (1980) are also close to our experimental data. The constants in their equation have been determined using the experimental data of fluidized beds with low-density particles (1,100–1,300 kg/m^3) and low inertia, while the other correlations have been based on wider ranges of solid density. Their results confirm our findings on the influence of the particle inertia on the mass transfer rate.

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Notation

C_A = electroactive species concentration, kmol/m^3
d_c = column diameter, m
d_p = particle diameter, m
D = coefficient of effective diffusion, m^2/s
F = Faraday constant, A·s/m
g = acceleration of gravity, m/s^2
Ga = Galileo number defined by $d_p^3 g \rho_L^2/\mu L$
I = intensity of diffusion-limiting current, A
K = liquid-solid mass transfer coefficient, m/s
Mv = density number defined by $(\rho_L - \rho_s)/\rho_L^2$
n_e = number of electrons in an electrochemical reaction
S = surface of spherical electrode, m^2
Sc = Schmidt number defined by $\mu_L/(\rho_L D)$
Sh = Sheerwood number defined by Kd_p/D
U_L = superficial light velocity, m/s

Greek letters

μ_L = dynamic viscosity of liquid, Pa·s
ρ_L = density of liquid, kg/m^3
ρ_s = density of solid particles, kg/m^3

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