Gas-phase influence on the mixing in a fluidized bed bio-reactor

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Summary. The liquid and solids mixing in fluidized bed bio-reactors containing particles with a density only slightly higher than water (1100 kg/m³) is generally consistent with the results found in previous studies for reactors with particles of higher density. The liquid mixing can be described by an axial dispersion model for a large variety of conditions while the solids follow the streamlines of the liquid. In the presence of a gas phase the degree of mixing of both the liquid and the solid phase increased. This effect became larger with increasing reactor diameter. In the extrapolation of laboratory data of three phase

fluidized bed bio-reactors to pilot plant systems this effect should be taken into account. The liquid and solids mixing may have a substantial effect on overall conversion rates and on possible microbial stratification in the reactor.

Nomenclature:

Bo,	Bodenstein number $v L/D$ (—)
D_r ,	diameter of the fluidized bed reactor (m)
D_l ,	Dispersion coefficient of the liquid phase (m ² /s)
D_g ,	dispersion coefficient of the solid phase (m ² /s)
$E(\text{in})$,	normalized dye concentration function entering the ideally mixed tank reactor (—)
$E(t)$,	normalized dye concentration function as measured (—)
L ,	length of the axial dispersed reactor (m)
t ,	time after dye injection (s)
t_m ,	time constant for microbial selection (s)
t_s ,	solid mixing time constant (s)
Δt ,	time interval in which a particle migrates within the bed (s)
v_l ,	superficial gas velocity (m/s)
v_u ,	superficial liquid velocity (m/s)
Δz ,	migration distance of a particle in the bed (m)
μ_1 ,	in situ growth rate of a dominant organism (s ⁻¹)
μ_2 ,	in situ growth rate of a recessive organism (s ⁻¹)
τ ,	average residence time in the axial dispersed reactor (s)
τ_i ,	average residence time in the ideally mixed tank reactor (s)

Introduction

For the quantitative interpretation of microbial activities and overall mass balances in fluidized bed bio-reactors it is essential to have an accurate knowledge of the liquid and solids mixing, since these factors influence the concentrations as experienced by the individual particles in the reactor. Thus the question might be raised whether the liquid and solids mixing may influence overall kinetics of the substrate removal and also the possible selection of microorganisms in the microbial communities of a biofilm. The latter is of particular interest in the case of undefined mixed culture systems such as in waste water treatment plants.

In the course of a feasibility study of microbial oxidation of sulphide and denitrification for waste water treatment we used a laboratory model of a fluidized bed reactor filled with biomass covered sand particles. The reduction of nitrate to nitrogen-gas in the biofilm leads to the evolution of gas bubbles, and therefore the influence of this gas phase on the liquid and solids mixing must be taken into account. To avoid complications caused by biologically active particles the effect of the gas phase was studied with inert particles. Since it is very difficult to simulate gas evolution from these particles we have chosen to study two extremes: a two phase fluidized bed reactor (without gas) and a three phase system in which gas is introduced artificially at the bottom.

The density of the particles in bio-reactors is usually only slightly higher than that of water (cells or enzymes immobilized in alginate, about 1050 kg/m^3 ; biomass covered sandparticles, about 1070 kg/m^3), while the particles often are several mm's in diameter. Since earlier studies on liquid mixing in fluidized bed reactors (Shah et al. 1978; Kim and Kim 1981; Deckwer and Schumpe 1984; Joshi 1980; Krishnawamy et al. 1978; Wild et al. 1982) involved considerably smaller particles and/or particles with a much higher specific density the results are not directly applicable to our case. Two recent studies (Ching and Ho 1984; Kikuchi et al. 1984) investigating two phase (L, S) fluidized bed reactors used particles that were representative for biological systems. The influence of gas evolution on the flow dynamics was, however, not taken into account.

For fluidized bed reactors with heavier particles (2500 kg/m^3) such as studied by Joshi (1980) and Kim and Kim (1981) a significant influence of the gas phase on the liquid mixing was reported. In his review of the work of Ostergaard, El-Tentamy and Muromuya, Epstein (1981) concluded that for fluidized bed reactors with three phases the degree of liquid mixing sharply increased with increasing reactor diameter. Similarly, the equations describing the liquid mixing in the three phase system of vertical sparged bubble columns as compiled by El-Tentamy et al. (1985) all take the superficial gas velocity and the reactor diameter into account. In contrast to the behaviour of the liquid very little is known about the solids mixing in three phase fluidized bed reactors, except for the fact that the solids will most likely follow the streamlines of the liquid as long as the particles are neither too large nor too heavy, as was also reported by Epstein (1981) and Joshi (1980). The aim of this study was to establish to what extent the trends found for the liquid and solids mixing in fluidized bed reactors with heavy particles also apply to reactors with a solid phase of considerable lower density, with special focus on the influence of the gas phase on the operation of the systems. These phenomena have been investigated as a function of the diameter of the reactor.

Theory

Liquid mixing

Completely mixed and plug flow behaviour are two extreme cases of ideal reactor flow dynamics.

Real reactors, however, seldomly behave as one of these extremes. For the description of non-ideal flow dynamics in tubular reactors the axial dispersion model is widely accepted (Levenspiel 1972; Ching and Ho 1984; Joshi 1980). In this model the residence time distribution (RTD) of an added compound (dye) can be predicted by the following equation (with open-open boundary conditions, analogous to Levenspiel 1972):

$$E(t) = 0.5(\pi \cdot \tau \cdot t / \text{Bo})^{-0.5} \cdot \exp(\text{Bo}(t - \tau)^2 / (4 \cdot \tau \cdot t)) \quad (1)$$

$E(t)$: normalized dye concentration as measured (—)

τ : average residence time in the axial dispersed reactor (s)

t : time after dye injection (s)

Bo : Bodenstein number $v_1 \cdot L / D_1$ (—)

For an accurate description of the experimental set-up (see Experimental) we used the response curve for a plug flow reactor with axial dispersion in series with that of an ideally mixed tank reactor. The residence time distribution in an ideally mixed tank reactor is described by:

$$dE/dt = (E(\text{in}) - E(t)) / \tau_t \quad (2)$$

$E(\text{in})$: normalized dye concentration entering the tank reactor

τ_t : average residence time in the tank reactor (s)

Substituting Eq. (1) as $E(\text{in})$ in Eq. (2) yields the following differential equation:

$$dE/dt = (0.5(\pi \cdot \tau \cdot t / \text{Bo})^{-0.5} \cdot \exp(\text{Bo}(t - \tau)^2 / (4 \cdot \tau \cdot t)) - E) / \tau_t \quad (3)$$

This differential equation was solved numerically on a Hewlett Packard 1000 mini-computer. Parameter estimation and optimization programmes (non linear least squares curve fitting) were used to determine the Bodenstein number from the experimental RTD-data. In graphical representations the theoretical RTD-curves were compared with the measured data.

Solids mixing

The degree of mixing of the solid phase can be represented by an axial dispersion coefficient D . For this we used the modified Einstein-Smol-

chowski equation, according to Van der Meer (1985):

$$D_s = 1/6 \Delta z^2 / \Delta t \quad (4)$$

With this equation it is possible to calculate this dispersion coefficient from so called random-walk data.

Experimental

Liquid mixing

The experimental set-up is shown in Fig. 1. The diameters of the plexiglass reactors used in this study were 2, 4, 8, and 12 cm. The three phases used in this study consisted of tapwater (*L*), air (*G*) and alginate beads (*S*). The alginate beads were prepared as described by Schoutens et al. (1986) and were loaded with yeast cells (*Saccharomyces cerevisiae*) giving a specific weight of 1050 kg/m³ and particle sizes between 2–3 mm. Thus representative particles for biologically ac-

tive systems were obtained. No special care was taken to make the beads exactly spherical.

The residence time distribution of the liquid in these reactors was determined by pulse response experiments. The injection of a suitable dye at the base of the reactor in a very short time interval (0.5–1 s) was assumed to give an ideal pulse. Blue dextran with a concentration of 30 kg/m³ was selected as a dye because of its high specific absorbance and very large molecular weight. The latter was of importance to avoid diffusion of the dye into the alginate beads. Overnight incubation of the alginate beads in the concentrated solution of the dye showed that no significant absorption or diffusion into the particles occurred. The response curves were measured with a Vitatron UV detector by monitoring the change in the absorbance of the liquid phase at 340 nm, after separation of the liquid and the gas phase in a small vessel. Though small, this vessel did have some influence on the response curves measured. Therefore it was necessary to use a model of an ideally mixed tank (the separator) in series with the actual axial dispersion model of the reactor (see Theory).

Solids mixing

In order to determine the migration of particles through the bed, some of the alginate particles were dyed by overnight incubation in an Eriochrome black T solution. The beads were evenly coloured and still had the same physical properties as the uncoloured beads, as was confirmed by the random behaviour of these coloured particles in the reactors. Their migration in the fluidized beds was followed during three intervals of 100 seconds. Data were taken into account only of the particles that had been visible during the complete experiment, and that did not reach the top and/or bottom of the bed during the experiment.

Results and discussion

Liquid mixing

In Fig. 2 experimental data are shown together with the fitted theoretical RTD-curves. The measured data shown here are from 8 representative experiments out of a total of 52. The theoretical curves were generated with formula (3) and by variation of the Bodenstein numbers a best fit was obtained. The good agreement between the theor-

Fig. 1. Experimental set-up for measuring liquid axial dispersion numbers (a tapwater supply; b air supply; c sintered glass plate; d septum for injection of dye solution; e plexiglass column; f solids retainer; g gas/liquid separator; h U.V. detector + recorder)

Fig. 2. Comparison of theoretical residence time distribution curves with the experimental response data in reactors with increasing diameters (A, 2 cm; B, 4 cm; C, 8 cm; D, 12 cm) in the presence and absence of gas (experimental: Δ two phase, $\blacktriangle$ three phase, $v_g \approx 0.1$ cm/s; theoretical: — two phase, $\cdots$ three phase; for the experimental conditions see Table 1)

etical curves and the experimental data under widely varying conditions indicate that the axial dispersion model gives a good description of the liquid mixing in the reactors studied. It also implies that the delta-pulse assumption of the tracer addition was valid. The validity of the axial dispersion model for three phase systems was also found for fluidized bed reactors with heavier particles (Joshi 1980; Wild et al. 1982). In contrast, Ching and Ho (1984) stated that a more complex model (two parameters) might be necessary to describe the flow dynamics in their experimental set-up. However, the same authors observed that the apparent complex behaviour of their experimental system might have been influenced by diffusion of the NaCl-tracer into the polymer particles. Similarly, Alvarez-Cuenca et al. (1979) also proposed a two parameter model to describe the hydrodynamics in two- and three phase fluidized bed reactors. This was necessary to describe the region where the gas and liquid were distributed. Apparently, in the present study the sintered glass

plates used as distributors created a flow pattern regular enough to allow the direct application of the axial dispersion model.

In our own RTD-experiments in the absence of particles anomalous behaviour occurred. Visual observations indicated that in these cases the dye-solution and the water phase did not mix completely, probably due to minor differences in specific weight and surface tension. The presence of the alginate particles completely nullified this effect.

The liquid dispersion coefficients, D_l , calculated from the Bodenstein numbers as a function of the reactor diameter are shown in Fig. 3. The experimental conditions and the average dispersion coefficients with standard deviations are presented in Table 1. The increase of the dispersion coefficients with the diameter of the reactor, though only very slight in the absence of gas, is in accordance with the observations of Joshi (1980) and Epstein (1981). We did not observe any substantial effects of the superficial liquid velocity

Fig. 3. Axial dispersion coefficients (calculated from the RTD-curves) as a function of the reactor diameter in the presence and absence of gas (— Δ — two phase, $\cdots \blacktriangle \cdots$ three phase, $v_g \approx 0.1$ cm/s)

(0.5–2.0 cm/s) on the degree of axial dispersion in our reactors. The measurements of Kikuchi et al. (1984) with polystyrene beads (1.887 mm, 1050 kg/m³) in a reactor of 3 or 4 cm diameter resulted in dispersion coefficients (0.06–1.0 cm²/s) that were very close to our results with the two phase systems. Particularly interesting was the effect low superficial gas velocities (0.05–0.2 cm/s) had on the axial dispersion of the liquid phase. The influence of the gas increased sharply with increasing reactor diameters. This effect, though more pronounced, agrees well with the trends described for three phase fluidized systems containing particles with a higher density (Joshi 1980; Kim and Kim 1981; Epstein 1981).

It is clear that for the interpretation of laboratory scale studies of three phase fluidized bed bio-reactors the degree of mixing of the liquid phase should be taken into account. The extrapolation of laboratory data to pilot plant experiments is unwarranted, because with this usually increases

Table 2. Migration distances and calculated solid phase axial dispersion coefficients (according to Van der Meer 1985) of coloured beads in two and three phase fluidized bed reactors

Reactor diameter (m)	v_l (cm/s)	v_g (cm/s)	Migration distance (cm)	$D_s \times 10^4$ (m ² /s)
0.02	1.0	—	8 ± 3	0.1
0.02	1.0	0.12	11 ± 2	0.2
0.04	1.1	—	14 ± 3	0.3
0.04	1.1	0.09	19 ± 4	0.6
0.08	1.0	—	32 ± 5	1.7
0.08	1.0	0.11	83 ± 12	11.5

of the reactor diameter from about 2 or 4 cm to above 10 cm are involved. The influence of the mixing behaviour on the performance of a fluidized bed reactor, however, will greatly depend upon kinetics and rates of reaction(s) occurring in the system under study.

Experiments in our laboratory with a denitrifying, sulphide-oxidizing system and those reported by Hamza et al. (1981) showed that nitrogen-gas produced by active denitrifying biomass yielded only very small (1–2 mm) gas bubbles. These bubbles seem to have less influence on the hydrodynamic behaviour than when the gas is added to the reactor. The difference in behaviour could not be explained by the increase of the ionic strength of the waste waters used, because RTD-experiments using tapwater with salt added to a comparable ionic strength did not show any difference with the experiments using tapwater directly (results not shown). The formation of small gas bubbles with little effect on the liquid mixing were also reported for CO₂ production during the butanol fermentation by *Clostridium beyerinkii* (Schoutens et al. 1986). Thus it must be concluded that the results of the experiments with external gas supply to the reactor set the upper limit for modelling of reactors with in situ gas production.

Solids mixing

Table 2 gives the results of the measurements on the particle migration through the bed. The dispersion coefficients for the solids in this table were calculated using equation (4). Again the influence of the gas phase was strongly dependent on the diameter of the reactor. The dispersion coefficients, D_s , for the two phase systems are comparable to those reported by Van der Meer

Table 1. Average liquid dispersion coefficients, D_1 , calculated from the residence time distribution experiments presented in Fig. 2 at the superficial liquid and gas velocities stated. The numbers between brackets indicates the number of experiments

Reactor diameter	v_l (cm/s)	v_g (cm/s)	Without gas $D_1 \times 10^4$ (m ² /s)	With gas $D_1 \times 10^4$ (m ² /s)
0.02	1.3	0.111	1.5 ± 0.5 (4)	1.5 ± 0.5 (17)
0.04	1.2	0.083	1.1 ± 0.4 (4)	6.5 ± 3.6 (17)
0.08	1.0	0.083	1.4 ± 0.5 (2)	21 ± 13 (5)
0.12	1.0	0.114	3.5 ± 1.4 (2)	55 (1)

for ion exchange particles (0.5–0.7 mm, 1300 kg/m³) at comparable liquid velocities. Van der Meer (1985), however, reported no influence of the column diameter on the solids mixing. The results of this study show a slight increase of the solids mixing with increasing diameter for the two phase systems and a much stronger increase for the three phase systems. These results confirm the theory (Epstein 1981; Joshi 1980) that solids in fluidized beds will follow the streamlines of the liquid as long as the particles are neither too large nor too heavy. The relatively light particles used in this study, though large compared to other studies, fulfill this condition.

The consequences of the observed phenomena for the biological performance of fluidized bed bio-reactors are indirect. Under conditions of incomplete mixing the particles present at different heights in the reactor can experience different surroundings in the reactor. In the case of undefined mixed culture systems (such as in waste water treatment) the migration of the particles through the bed will determine whether or not differentiation in the composition of the microbial population on the particles at different heights will occur. In order to assess the possibility of this phenomenon a first approximation can be made by determining the time constants for the two processes (Oosterhuis 1984). For the solids migration the following equation can be used:

$$t_s = L^2/D_s \quad (5)$$

t_s : solids mixing time constant (h)
 L : height/length of the reactor (m)
 D_s : solids dispersion coefficient (m²/h)

For the reactors used in this study the time constant for the solids mixing lies between 0.3–30 h depending on reactor diameter and the presence of a gas phase. The competition between microorganisms can be seen as a difference in growth rate between two competing organisms. The time constant for this process can be calculated with:

$$t_m = 1/(\mu_1 - \mu_2) \quad (6)$$

t_m : time constant for microbial selection (h)
 μ_1 : in situ growth rate of the dominant organism (h⁻¹)
 μ_2 : in situ growth rate of the recessive organism (h⁻¹)

The minimum time constant will be found when the recessive of the two competing organisms

does not grow at all. A generally accepted maximum growth rate in waste water treatment systems is 0.2 h⁻¹ (Heijnen 1984; Strand and McDonnell 1985; Wanner 1985). This would give a minimum time constant in the order of 5 hours. Given a range of time constants for solids mixing of 0.3–30 h the implication is that, depending on the specific reactor, the relative stratification in the fluidized bed indeed may lead to differentiation in the population composition at different heights in the reactor.

In most practical systems (especially with growing particles) the particle size distribution will not be as narrow as in the solids dispersion experiments described here. The influence of stratification will then be stronger as according to the random dispersion behaviour described in this study. In the discussions concerning microbial selection this effect should also be kept in mind.

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