

Mass Transfer and Bed Expansion of Solid-Slurry Fluidized Beds

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In recent years biotechnological applications calling for development of new fluidized bed processes have often added new complexities to traditional liquid-solid fluidized bed designs. Multisolid fluidized beds, in which two or more distinct sizes of particles reside in a batch mode in the fluidized bed, or slurry-solid fluidized bed systems, in which a fine particle slurry in the transport mode flows through a fluidized bed of larger and/or denser particles, are general examples. A specific example of the latter is the so-called multisolid fluidized bed bioreactor (slurry-solid fluidized bed bioreactor by the present notation) developed by Battelle Memorial Institute (Allen, 1986), in which a slurry of fine particle product adsorbent flows continuously through a fluidized bed bioreactor containing particles with immobilized cells. In the present work, water, benzoic acid pellets, and various fine particles are employed to study the effect of liquid flow rate, fluidized bed particle size, slurry particle size and density, and slurry particle loading on solid-liquid mass transfer in a slurry-solid fluidized bed system. Comparison with conventional liquid-solid fluidized bed mass transfer is performed. Data are also presented regarding the effect of the slurry flow on the bed expansion characteristics of particles of various shapes, sizes, and densities and are analyzed with respect to the conventional Richardson-Zaki correlation (Richardson and Zaki, 1954).

Experimental Method

The experimental procedure for determining mass transfer via dissolution in liquid-solid fluidized beds is well documented (McCune and Wilhelm, 1949; Couderc et al., 1972). The experimental set-up and procedure were essentially those used by Arters and Fan (1986) for liquid-solid fluidized beds, with some modifications. The column itself was of 10.2 cm ID and 137 cm height. The liquid distributor consisted of a vacant calming sec-

tion 20 cm in height and a 0.075 cm mesh retaining screen; the feed tank included a stirrer and screen-covered overflow. Procedural modifications included continuous sieving of the column outlet flow to recover the slurry particles, which were washed and reintroduced into the feed tank on a periodic basis every 30 s. Slurry concentration samples were taken from the feed tank during runs. The cylindrical benzoic acid particles recovered at the conclusion of each run were dried, remeasured with respect to diameter, height, weight, and density, and reused as described by Arters and Fan (1986). The density of the particles was found to remain constant ($1.290 \pm 0.009 \text{ g/cm}^3$) regardless of particle size. Benzoic acid sample concentrations were obtained using spectrophotometric analysis at 228 nm.

Bed expansion experiments run with inert fluidized bed particles were simpler in that the slurry could be recycled directly back into the feed tank. In all cases, the bed height was measured visually.

Fluidized bed particles consisted of cylinders of benzoic acid (4BZ, 3BZ, 2BZ); 0.4, 0.2, and 0.1 cm glass beads (4GB, 2GB, 1GB, respectively); 0.25 cm nylon balls (NYL); and 0.4 cm PVC cylinders (PVC). Slurry particles used were 330 μm glass beads (330 gb), 460 μm glass beads (460 gb), and 315 μm Good-year Pliolite plastic particles (315 pl). Slurry particle size was analyzed using a Nikon optical microscope and Optomax Image Analyzer. Slurry and fluidized particle properties are listed in Table 1. Tap water was used as the liquid. Table 2 summarizes the range of the experimental conditions.

Results and Discussion

Bed expansion and phase holdup

Slurry-solid bed expansion was found to be greater than the corresponding liquid-solid expansion under the same fluid flow conditions, even when the space taken up by the slurry particles

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Table 1. Particle Properties

Type	Abbrev.	Material	d_p cm	ρ_p or ρ_s g/cm ³
Fluidized bed	4GB	Glass spheres	0.40	2.47
	2GB	Glass spheres	0.21	2.47
	1GB	Glass spheres	0.10	2.87
	NYL	Nylon Spheres	0.25	1.15
	PVC	PVC cylinders	0.37	1.47
	4BZ	Benzoic acid cylinders	0.45, 0.44, 0.43, 0.42, 0.41	1.29
	3BZ	Benzoic acid cylinders	0.32, 0.30	1.29
Slurry	3BZ	Benzoic acid cylinders	0.28, 0.26, 0.25	1.29
	460gb	Glass bead	0.046	2.50
	330gb	Glass bead	0.033	2.50
	plio	Pliolite granules	0.031	1.02

is taken into account. Both slurry particle type and concentration were found to affect the fluidized bed expansion. Larger terminal velocity slurry particles, i.e., denser and/or larger, had a greater effect on the bed expansion. Likewise, higher concentrations of slurry particles were found to have a larger effect than lower concentrations. The effect of the slurry was found to be greater for small and/or light, low terminal velocity fluidized particles than for denser, higher terminal velocity particles. This trend of lowered effect with increasing fluidized bed particle terminal velocity was followed consistently to the case of the highest terminal velocity particles tested, 0.4 cm glass beads (4GB), in which none of the slurry particles used had a measurable

Table 2. Range of Experimental Conditions

Particle Abbrev.	U_{sl} cm/s	Slurry	C_s	Symbol (Figs. 1, 2)
4GB	8.5–11.9	460gb	0.032	▼
		460 gb	0.022	▼
		330gb	0.050	▼
2GB	7.6–11.2	None	—	▲
		460gb	0.032	▲
		460 gb	0.022	▲
1GB	6.9–12.0	None	—	◇
		330gb	0.049	◆
		330gb	0.013	◆
		Plio	0.018	◆
NYL	3.9–6.1	None	—	▷
		330gb	0.005–0.010	▷
		Plio	0.018	▷
PVC	6.7–11.8	None	—	□
		460gb	0.032	■
		460gh	0.004–0.017	■
		330gb	0.046	■
		330gb	0.013	■
		Plio	0.014	■
4BZ	5.8–10.9	None	—	○
		460gb	0.001–0.003	○
		330gb	0.001–0.004	○
		330gb	0.006–0.012	○
3BZ	6.1–11.4	Plio	0.001–0.003	○
2BZ	5.5–10.1	460gb	0.0005	○
		330gb	0.002–0.003	○
		Plio	0.002	○

effect. Conversely, under certain conditions of high slurry concentration and high terminal velocity slurry particle, the lower terminal velocity fluidized particles (2BZ, 3BZ, NYL) were transported from the bed at relatively low slurry velocities ($U_{sl}/U_{tp} < 0.8$).

In order to quantify the effect of the slurry, the correlation of Richardson and Zaki (R-Z; 1954), widely used in the calculation of liquid-solid fluidized bed expansion, was employed:

$$U_i/U_t = \epsilon_i^n \quad (1)$$

where, for the range of conditions in the present study,

$$n = 2.39 \quad Re_{tp} > 500 \quad (2a)$$

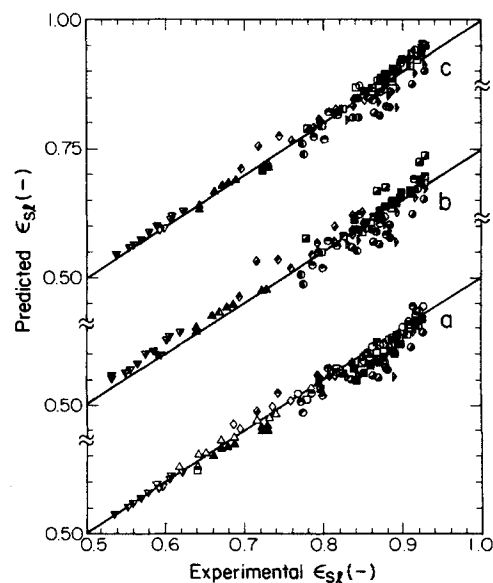
$$n = 4.45 Re_{tp}^{-0.1} \quad 200 < Re_{tp} < 500 \quad (2b)$$

and U_b , the apparent fluidized particle terminal velocity, is correlated by

$$\log(U_i) = \log(U_{tp}) - d_p/D_{col} \quad (3)$$

Values of the terminal velocity of fluidized bed particles as well as slurry particles were calculated through the equations given by Clift et al. (1978). These values compared well with those obtained by dropping individual particles into stagnant liquid in the column and timing their fall through a known distance, although the difference was larger for the cylindrical PVC particles (8% vs. an average of 4% for the spherical particles).

Plot *a* in Figure 1 presents the results of applying the R-Z equation to the slurry fluidized system using liquid properties alone in the calculations. As seen in the figure, the R-Z equation was found to fit data for the case of no slurry particles in the system reasonably well, the average deviation being about 4%

**Figure 1. Experimental vs. predicted slurry phase hold-up.**

a. Richardson-Zaki (1954) equation using liquid properties alone
b. R-Z equation using calculated slurry properties
c. R-Z equation using Eq. 4 to determine U_i
Symbols as in Table 2

for bed voidages below about 92%. (The phenomenon of changing slope in the log-log velocity vs. voidage curve above a certain voidage has been well documented, e.g., Jean and Fan [1987], and these points have been excluded from calculations.) However, data from slurry-solid beds deviate from the predicted values by as much as 30% for beds of low terminal velocity NYL particles in a high-concentration slurry.

Plot *b* in Figure 1 shows the fit of the data to the Richardson-Zaki correlation substituting effective slurry properties in the calculations. Slurry viscosity is obtained via the well-known equation of Happel (1957) and density is calculated from a simple volume average. Terminal velocities were calculated using the equations of Clift et al. (1978), using the slurry properties. Voidages calculated using these values are generally more accurate than those using pure liquid properties at low slurry concentrations (<1%), but tend to overpredict the effect of the slurry for higher slurry concentrations and for high terminal velocity fluidized particles.

It was observed that the R-Z index, *n*, remained nearly constant regardless of the presence of slurry in plots of ϵ_{sl} vs U_i , but that U_i varied. A correlation was developed to predict the slurry medium U_i' based on the experimental results. It was determined that $(U_i' - U_i)/U_i$ varied with approximately the square root of the slurry concentration, C_s , under conditions of constant slurry and fluidized bed particle type. A dimensionless terminal velocity number, $T_m = (U_{ip} - U_{is})/U_{is}$, was defined which includes the effects of slurry particle size and density, as well as the interaction between fluidized bed particle and slurry particle. Subject to a least-squares evaluation, the data were correlated by the exponential relation

$$U_i' = U_i(1 - 1.03C_s^{0.55}T_m^{-0.99}) \quad (4)$$

over the range: $61 < Re_p < 530$, $0.0 < T_m < 151$, $0.0 < C_s < 0.050$.

Plot *c* in Figure 1 shows the fit of the R-Z equation utilizing simple liquid properties and U_i' from Eq. 4 in the calculations. Most of the data lie within 6% of the correlation. Those points with a larger error occur for the fluidized bed particles most affected by the slurry (2BZ) and under large slurry concentration conditions. Equation 4 satisfies the boundary conditions of the system: When no slurry is present, $C_s = 0$ and the effect is zero. Likewise, as the slurry particle becomes infinitesimally small and its terminal velocity becomes very small, $T_m^{-0.99}$ approaches zero and the effect again goes to zero. Finally, as the terminal velocity of the slurry particle approaches that of the fluidized bed particle, T_m approaches zero and the effect is undefined, i.e., no fluidized bed can exist for a velocity greater than the slurry particle terminal velocity.

Mass transfer

It was found that the entire range of mass transfer data, regardless of slurry concentration or type, could be accurately fitted by the simple correlation given by Calderbank (1967) with the inclusion of a shape factor, ϕ , as shown in Figure 2:

$$Sh = 0.31\phi^{0.6}Ar^{1/3}Sc^{1/3} \quad (5)$$

The correlation fitted the slurry-solid mass transfer data with an average deviation of 10.9%, vs. 10.3% for the liquid-solid fluidized bed. Both sets of data exhibited a slight bias toward under-

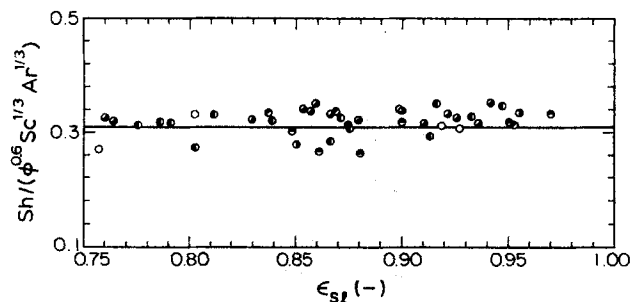


Figure 2. Fit of the equation of Calderbank (1967) to mass transfer data from slurry-solid fluidized bed.

Symbols as in Table 2

prediction by the correlation, probably a result of distributor effects (Damronglerd et al., 1972).

It may be concluded that the presence of a slurry medium, while exhibiting a clear hydrodynamic effect on the fluidized bed, does not alter the factors responsible for determining the mass transfer. Only physical and chemical properties of liquid and solid are required to predict the mass transfer in the liquid-solid fluidized bed. Correlations containing terms of void fraction and liquid velocity (Evans and Gerald, 1953; Couderc et al., 1972), do not represent the fundamental parameters, a point raised by Damronglerd et al. (1975) and Arters and Fan (1986), among others. This is not to say, however, that the mass transfer in a fluidized bed may not be affected by other parameters. Both mechanical agitation (Kobayashi and Saito, 1965; Sanger and Deckwer, 1981) and the addition of gas to the fluidized system (Hassanien et al., 1984; Arters and Fan, 1986) have been found to increase the mass transfer. Increased turbulence in these systems increases particle-liquid relative velocity, a parameter that remains essentially constant (equal to the particle terminal velocity) in an undisturbed liquid-solid fluidized bed.

Another point to be raised from these findings is of significance to future experimental efforts. Couderc et al. (1972) reported that dilution of active particles by inert particles, i.e., beds composed partially of benzoic acid particles and partially of inert particles, affected the mass transfer characteristics of the bed even when the solids had similar geometry and physical properties. This contrasts with the present work's finding that dilution of the fluidized bed particles by slurry particles has no effect on the mass transfer. Couderc et al., however, used a correlation that contained the void fraction of the bed. It is possible, then, that the difference seen in the mass transfer calculation was in fact a difference in the voidage of the bed caused by the use of inert particles of a slightly different hydrodynamic character than the active particle.

Acknowledgment

This work was supported by National Science Foundation Grant Nos. CBT-8516874 and CBT-8506343.

Notation

- Ar = Archimedes number = $d_p^3\rho(\rho_p - \rho_l)g/\mu_l^2$
- C_s = concentration of slurry particles in liquid, cm^3/cm^3
- D = molecular diffusivity of solid dissolved in liquid, cm^2/s
- D_{col} = column diameter, cm
- d = equivalent particle diameter, cm

g = gravitational acceleration, cm/s^2
 k = solid-liquid mass transfer coefficient, cm/s
 n = Richardson-Zaki index
 Re = superficial particle Reynolds number based on fluid velocity = $U_d \rho_l / \mu_l$
 Sc = Schmidt number = $\mu_l / \rho_l D$
 Sh = Sherwood number = $k d_p / D$
 T_m = Terminal velocity number = $(U_{tp} - U_{ts}) / U_{ts}$
 U = average superficial fluid velocity, cm/s
 U_t = apparent fluidized bed particle terminal velocity, cm/s
 U'_t = apparent terminal velocity of fluidized particle adjusted for presence of slurry, cm/s
 U_{tp} = fluidized particle terminal velocity, cm/s
 U_{ts} = slurry particle terminal velocity, cm/s

Greek letters

ϵ = holdup
 μ = fluid viscosity, $\text{g/cm} \cdot \text{s}$
 ρ = density, g/cm^3
 ϕ = sphericity

Subscripts

l = liquid
 p = fluidized bed particle
 s = slurry particle
 sl = slurry phase
 t = terminal velocity condition
 tp = fluidized bed particle terminal velocity condition

Literature Cited

Allen, B. R., "Multi-Phase Fluidized Bed—An Advanced Bioreactor Concept," Ann. Progress Rept. Battelle Mem. Inst., to Jet Propulsion Lab., under contract 957241 (1986).

Arters, D. C., and L.-S. Fan, "Solid-Liquid Mass Transfer in a Gas-Liquid-Solid Fluidized Bed," *Chem. Eng. Sci.*, **41**, 107 (1986).
 Calderbank, P. H., "Mass Transfer," *Mixing—Theory and Practice*, II V. W. Uhl, J. B. Gray, eds., Academic Press, New York, 66 (1967).
 Clift, R., J. R. Grace, and M. E. Weber, *Bubbles, Drops, and Particles*, Academic Press, New York, 113 (1978).
 Couderc, J. P., H. Gibert, and H. Angelino, "Transfert de Matiere par Diffusion en Fluidisation Liquide," *Chem. Eng. Sci.*, **27**, 11 (1972).
 Damronglerd, S., J. P. Couderc, and H. Angelino, "Mass Transfer in Particulate Fluidization," *Trans. Inst. Chem. Eng.*, **53**, 175 (1972).
 Evans, G. C., and C. F. Gerald, "Mass Transfer from Benzoic Acid Granules to Water in Fixed and Fluidized Beds at Low Reynolds Numbers," *Chem. Eng. Prog.*, **49**, 135 (1953).
 Happel, John, "Viscosity of Suspensions of Uniform Spheres," *J. Appl. Physics*, **28**, 1288 (1957).
 Hassanien, S., H. Delmas, and J. P. Riba, "Hydrodynamique et Transfert de Matiere Solide-Liquide en Fluidisation a Trois Phases," *Entropie*, No. 119, 17 (1984).
 Jean, R. H., and L.-S. Fan, "On the Particle Terminal Velocity in a Gas-Liquid Medium with Liquid as the Continuous Phase," *Can. J. Chem. Eng.*, **65**, 881 (1987).
 Kobayashi, T., and H. Saito, "Mass Transfer from Suspended Solid Particles to Liquid in a Bubble Column," *Kagaku Kogaku*, abridged ed., **3**, 210 (1965).
 McCune, L. K., and R. H. Wilhelm, "Mass and Momentum Transfer in Solid-Liquid System," *Ind. Eng. Chem.*, **41**, 1124 (1949).
 Richardson, J. R., and W. N. Zaki, "Sedimentation and Fluidization. I," *Trans. Inst. Chem. Eng.*, **32**, 35 (1954).
 Sanger, P., and W. D. Deckwer, "Liquid-Solid Mass Transfer in Aerated Suspensions," *Chem. Eng. J.*, **22**, 179 (1981).

Manuscript received Aug. 24, 1987, and revision received Jan. 29, 1988.