Correlation of Nonverticality and Entrance Effects in Bubble Columns

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The prediction of mixing in bubble columns, especially that of axial dispersion coefficient, has received considerable attention in recent years, and it is clear that wide disagreement exists. A recent compilation of many results (Rice and Littlefield, 1987) showed order of magnitude differences among several research groups.

Generally, mixing in unstirred vessels has been correlated with respect to two key parameters: column diameter and superficial gas velocity. However, two recent papers (Tinge and Drinkenburg, 1986; Rice and Littlefield, 1987) have shown that slight departure from true verticality (<2 degrees) had a much stronger effect than diameter and velocity. However, the former used very small diameter columns, while the latter presented sparse results. Predating these mixing studies, Valdes-Kreig et al. (1975) observed that vertical misalignment can strongly affect surfactant removal in a foam fractionation column.

In this study, we present new visual VCR experimental evidence, using the acid-base technique described by Rice and Littlefield (1987). This technique allowed the following aspects to be studied:

- Entrance length for fully developed bubbly flow.
- Measurement of dispersion coefficient within the bubbly flow region, thereby mitigating inflation of values caused by the entrance region.
- Testing the "summation of resistance" hypothesis for two-zone bubble columns.
- Measurement of enhanced mixing times and effective dispersion coefficient for controlled column tilt.

Measurement Theory

The basis for the batch acid-base technique is to set up two separated regions driven by the concentration gradient of Fickian-like character. The measurement of the movement of the boundary can then be related to the effective dispersion coefficient.

Baird et al. (1976) were perhaps the first to use the acid-base technique for dispersion measurement, although their solution of the Fickian equations differs slightly from that of Rice and Littlefield (1987) who gave the following result:

$$C_{Ao} = \left(\frac{N_B L}{(1-\epsilon)D_e}\right)\left[-\frac{z_n}{L} + \frac{1}{3} + \frac{D_e t_n}{L^2} + \frac{z_n^2}{2L^2} - \frac{2}{\pi^2}\sum_{m=1}^{\infty}\frac{\cos(m\pi z_n/L)}{m^2}\exp(-m^2\pi^2 D_e t_n/L^2)\right] \quad (1)$$

where the pair $[z_n, t_n]$ portrays the position-time movement of the LON (line of neutralization). For long times, because $D_e t_n/L^2 > 0.3$ the series terms can be ignored, so that

$$D_e \simeq \frac{N_B L}{(1-\epsilon)}\left[\frac{1}{3} - \frac{z_n}{L} + \frac{z_n^2}{2L^2}\right] \Bigg/ \left[C_{Ao} - \frac{N_B t_n}{(1-\epsilon)L}\right] \quad (2)$$

A "one-shot" experiment can be performed without using a VCR by measuring the time to neutralize the total column contents, denoted as t_n^*, so when $z_n = L$, then we have

$$D_e \simeq \frac{C_{Bo} q L^2}{6} \Bigg/ [C_{Bo} q t_n^* - n_{Ao}] \quad (3)$$

where we have replaced $N_B = C_{Bo}q/A$. Thus, Eq. 3 allows, for most cases, a simple stopwatch to be used to estimate dispersion-coefficient. To subtract or otherwise account for the entrance zone, a complete plot of z_n vs. t_n is required, as we show presently.

Theory for Entrance Region

The region in the vicinity of the gas sparger, where interfaces are torn and formed, is highly chaotic and generally well-mixed,

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hence we model this as equivalent to a constant volume CSTR, which must be neutralized before upward movement can commence. By subtracting the two material balances, an elementary relationship for the transient difference $\Delta = C_B - C_A$ results:

$$\Delta(t) = C_{Bo} - (C_{Bo} + C_{Ao}) \exp\left[\frac{-qt}{(1-\epsilon)V}\right] \tag{4}$$

The entrance zone (CSTR) is completely neutralized when $\Delta = 0$, which allows the corresponding contact time t_n^c to be determined. This finite zone gives way eventually to the narrow LON, which begins moving up the column as excess alkali is injected (Barbe, 1989). The initial movement is somewhat erratic and difficult to pinpoint with the eye, but gradually it gives way to a clearly visible LON, which moves upward smoothly, completing a sigmoidal shape (see Figure 1; also, refer to the interesting photos of LON published by Rice and Littlefield, 1987).

Equipment and Sparger Details

The three bubble columns tested were constructed of acrylic pipe with diameters 0.1524 m (6 in.), 0.203 m (8 in.) and 0.254 m (10 in.). All columns had an open length of 1.93 m. The tops were fitted with special circular flanges designed to fit within a yoke arrangement (Figure 2) for adjustment of verticality using a micrometer-spring arrangement.

Each of the columns in turn was surrounded by a square water box, 0.305 m wide and 1.53 m tall, as shown in Figure 2. The square box, when filled with water, minimized refractive index discontinuities and promoted clear viewing.

The gas sparger was of unique design and comprised 3 mm thick latex sheet stretched over and clamped to a circular drumhead. The circular sparger to column diameter was set at 0.67, so from the smallest to largest column, these diameters were 0.102, 0.136 and 0.171 m, respectively. The sparger assembly was threaded directly into the square sheet facing the column base, hence the sparger was neatly flush-mounted on the base so that "dead zones" for mixing purposes would be minimized.

The drilled holes in the 3 mm latex sheet were arranged on a triangular pitch (to maximize number per unit area) with a spacing of 7.5 mm. The holes were drilled with a 0.378 mm steel bit at around 1,200 RPM. The numbers of drilled holes, from smallest to largest sparger, were 110, 196 and 400, respectively.

Another key feature, in addition to the property of uniform bubbling at each hole on the face of elastic spargers, is to eliminate "jetting" phenomenon, since as gas pressure increases, the sparger sheet (and hole sizes) expand in proportion. Thus, the hole size is self-regulating.

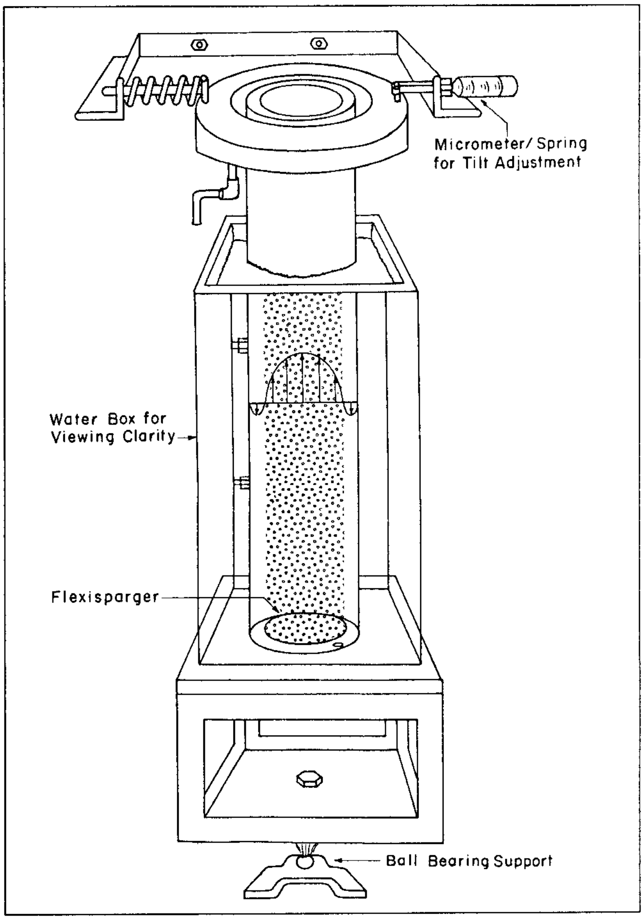

Figure 2. Water box and column tilt adjustment.

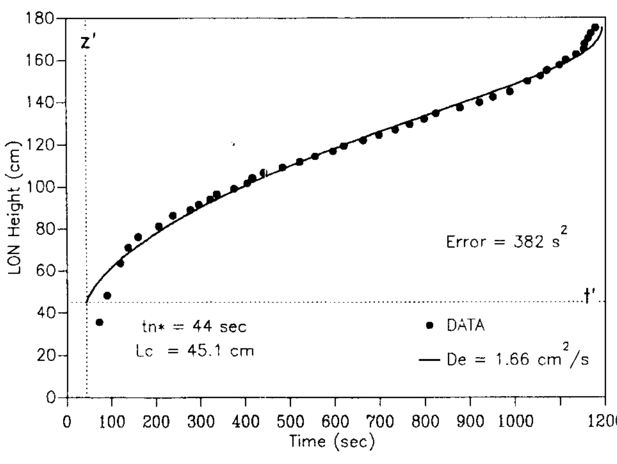

Figure 1. Comparison of theory and experiment for continuous (explicit) parameter estimation routine.
$u_{og} = 1.48$ cm/s, $d_c = 0.152$ m

Parameter estimation

Two parameters are needed to fit the Fickian diffusion model represented by Eq. 1 to actual LON data (z_n vs. t_n): D_e and L_c, *the effective dispersion coefficient and height of entrance zone.*

We tested two parameter estimation routines, both predicated on finding the proper coordinate system (z', t') to represent the beginning of Fickian-like behavior, somewhere up from the sparger. To find this "initiation" position, which is defined as $z' = z - L_c$, Rice and Littlefield (1987) allowed only discrete data be used to estimate L_c.

We have revised this method to account for a continuously varying entrance length by exploiting Eq. 4. Thus, coordinates for the Fickian region are taken to be

$$z'_n = z_n - L_c, \quad t'_n = t_n - t_n^c \tag{5}$$

where t_n^c is related to entrance length L_c by way of Eq. 4, since $V = AL_c$. Now, the parameter estimation problem can be posed to have a continuously varying L_c so that a global best fit for L_c and D_e can be found, by minimizing the criterion function:

$$F = \sum_{i=1}^{N} (t'_{ni} - \hat{t}'_{ni})^2/N \tag{6}$$

Entrance length

The acid-base method has the added feature of yielding information regarding estimates of entrance length, which occurs in the vicinity of the sparger where CSTR behavior was observed. Figure 3 shows the entrance length correlated with column diameter, so $L_c/d_c \simeq 2.77$. Dispersion coefficients in the Fickian zone were practically independent of gas velocity, taking averaged values of 2, 8 and 27 cm²/s, respectively, for smallest to largest columns.

Scaling law

In a recent published letter, Rice (1989) suggested the entrance and smooth zones could be treated as a series of resistances, so that

$$\frac{L_t}{D_o} = \frac{L_c}{D_{ce}} + \frac{L}{D_e} \tag{7}$$

The mixing coefficients are D_o (overall), D_{ce} (entrance zone) and D_e (Fickian zone) respectively. Also, L_t is the total length, L_c is the entrance length, and the length for smooth Fickian movement is $L = L_t - L_c$. Now, if D_{ce} obeyed a large-scale mixing law such as the Baird-Rice relation, it is clear that $D_{ce} \gg D_e$, so that approximately

$$\frac{L_t}{D_o} \simeq \frac{L}{D_e} \tag{8}$$

Relative to the previous experiments, D_o would correspond to application of Eq. 3, using total time to neutralization (with no corrections for entrance length). This computation should yield results similar to a tracer injection method of analysis. We test the predictions according to Eq. 8 in Figure 4, where a slope of 0.7 replaces unity.

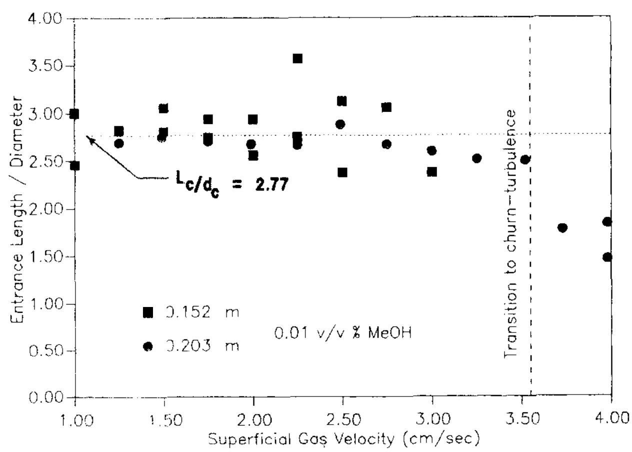

Figure 3. Correlation of entrance length for bubbly flow.

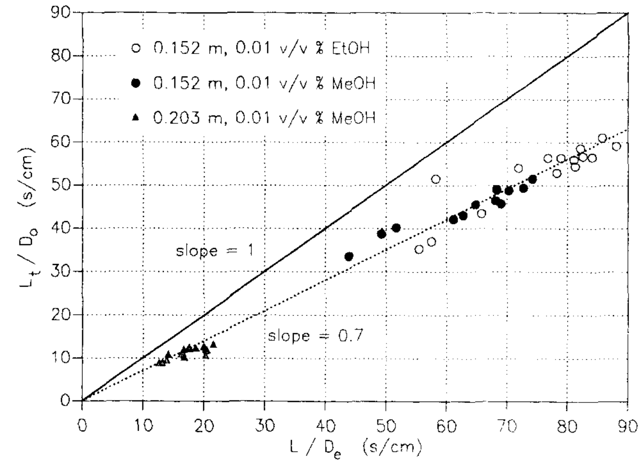

Figure 4. Correlation of overall dispersion coefficient relative to values in the developed bubbly (Fickian) zone.

Effect of nonverticality

Because of the difficulty in pinpointing the entrance zone for tilted (hence skewed circulatory, see Rice and Littlefield, 1987) columns, we shall use only total time to neutralization (t_n^*) applied to Eq. 1 (if $D_e t_n / L^2 < 0.3$) or otherwise to Eq. 3; in either case, we place $z_n/L = 1$. We denote mixing coefficients based on total time as D_o, i.e., overall coefficient.

Only two research groups (Tinge and Drinkenburg, 1986; Rice and Littlefield, 1987) have studied the effect of tilt on mixing in any systematic way. Our new measurements varied several physical quantities, with ranges:

$$0 < \alpha < 6.75 \times 10^{-3} \text{ radians}$$
$$1.5 < u_{og} < 4 \text{ cm/s}$$
$$15 < d_c < 25 \text{ cm}$$
$$120 < L_t < 180 \text{ cm}$$

We looked at each of these variables in turn, and unlike Tinge-Drinkenburg, we found effects arising from other than simple angle and diameter dependences. Barbe (1989) performed multiple regression analysis on more than 80 experiments to yield the correlation shown in Figure 5 as a ratio tilt to untilted dispersion coefficient:

$$D'_o/D_o = 1 + 313 \cdot \alpha^{1.73} Fr^{-0.36} \left(\frac{L_t}{d_c} - 2.77\right)^{1.18} \tag{9}$$

The angle dependence is in remarkably good agreement wtih Tinge and Drinkenburg (1986). However, we discovered a gas velocity dependence they did not report, through the Froude number ($u_{og}^2/d_c g$). This dependence is essentially $Fr^{-1/3}$, which is identical to the dimensionless representation of the Baird-Rice equation which was rearranged by Rice et al. (1981) to show that $Pe \propto Fr^{1/3}$.

The effect of length scale ratio (L_t/d_c) is somewhat weaker than that obtained by Tinge-Drinkenburg. This arises because we have corrected the length scale ratio to account for entrance length. We reasoned that tilt should have no effect when the emulsion height-diameter ratio approached the ratio for the

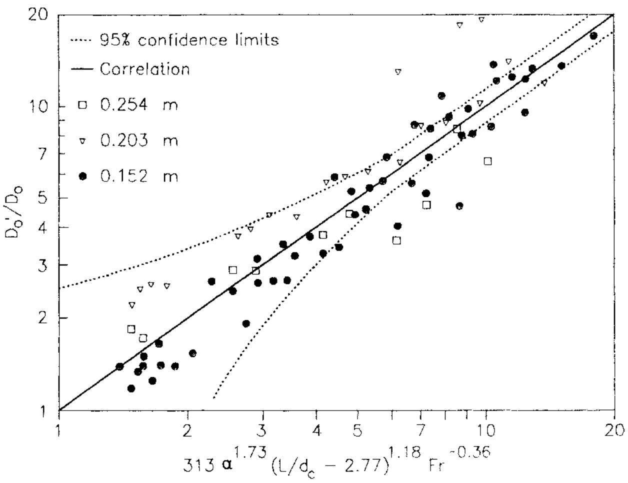

Figure 5. Correlation of dispersion ratio for tilted columns.

entrance zone, thus we expect (for any angle) $D_o' \sim D_o$ when $L_t/d_c \leq 2.8$, i.e., the condition for very short, stubby vessels.

Our results suggest that tilt will not be a significant factor for short columns with high gas injection rates. However, for tall columns operating in the homogeneous bubbly flow regime, tilt is the singularly most critical variable. This circumstance probably explains the wide disparity (two orders of magnitude) in the older literature on bubble columns, as pointed out by Rice and Littlefield (1987).

Acknowledgment

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Notation

- A = column cross-sectional area, m^2
- C_A = acid composition, mol/m^3
- C_B = base composition, mol/m^3
- C_{Ao} = initial acid composition, mol/m^3
- C_{Bo} = injected base composition, mol/m^3
- d_c = column diameter, m
- D_e = Fickian-Zone axial dispersion coefficient, m^2/s
- D_o = overall axial dispersion coefficient, m^2/s
- D_o' = overall coefficient for tilted column, m^2/s
- D_{ce} = entrance zone dispersion coefficent, m^2/s
- Fr = Froude number (U_{og}^2/gd_c)
- g = acceleration due to gravity, m/s
- k = reaction rate constant, m^3 · mol^{-1} · s^{-1}
- L_t = total emulsion height, m
- L = Fickian emulsion height, m
- L_c = entrance emulsion height, m
- L_o = clear solution height, m
- N = total number of data points
- n_{Ao} = $(1 - \epsilon)\ LA\ C_{Ao}$, total moles acid initially
- N_B = flux of alkali to column ($q\ C_{Bo}$)
- Pe = Peclet number ($u_{og}d_c/D_o$)
- q = volume rate of flow for alkali, m^3/s
- t = time, s
- t_n = time corresponding to LON position, s
- t_n' = $t_n - t_n^c$, corrected time
- $\bar{t}_n'$ = theoretical corrected time (computed from Eq. 1)
- t_n^c = time to neutralize entrance zone, s
- t_n^* = time to neutralize column contents, s
- u_{og} = superficial gas velocity, m/s
- V = volume of CSTR region
- z_n = local LON position, m
- z_n' = $z_n - L_c$, corrected LON position, m
- z = axial height coordinate, m

Greek letters

- α = angle of tilt, radians
- ϵ = overall gas voidage

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