Flow and Mixing in the Liquid between Bubbles

Bruce A. Finlayson, Professor Emeritus of Chemical Engineering
Department of Chemical Engineering, Box 351750, University of Washington, Seattle, WA 98195-1750

Abstract: Mixing is characterized in liquids moving between bubbles when the bubbles are moving down a microfluidic channel. The shape is assumed based on fluid mechanical arguments and experimental observations, and the mixing is characterized for a variety of situations in two- and three-dimensions.

Keywords: Mixing, bubbles, microfluidics

1. Introduction

The type of two-phase flow in a channel, whether the flow occurs in a segmented fashion, as is studied here, or in an annular fashion, or some other way, is usually determined by the mass fluxes of the two phases. For the cases here, the regime is in the segmented domain, as measured by Günther, *et al.*[1] agrees with the predictions from correlations in Perry's Chemical Engineer's Handbook.[2] Under these circumstances, several studies[1,3,4] have shown that the bubbles leave a thin layer of liquid on the boundary so that the mean velocity of the liquid is less than that of the bubbles. If the capillary number, $Ca = \eta U_b / \sigma$, where η is the viscosity, U_b the bubble velocity, and σ the surface tension, is small (0.001) surface tension forces dominate and the bubbles have approximately spherical caps.[1,3,4,5] For a single bubble, G. I. Taylor showed[3] that the ratio of velocities (mean fluid velocity, U, to bubble velocity, U_b)

$$m = \frac{U_b - U}{U_b}$$

is determined by the capillary number. For small numbers ($Ca < 0.1$) the relation is

$$m = 1.0(\eta U / \sigma)^{1/2}$$

Bretherton[4] developed an equation for long bubbles that is similar, although it did not fit the experimental data exactly.

$$m = 1.29(3\eta U / \sigma)^{2/3}$$

Later, Wong, *et al.*[5] developed a theory for the shape of bubbles in a square channel.

More recently in microfluidics, bubbles have been introduced into fluids flowing in micro-channels in order to induce and improve mixing.[1,6,7] While the measured flow profiles indicate that mixing occurs, seldom is the amount of mixing characterized in a quantitative way. Furthermore, it isn't clear how experimental conditions should be chosen to enhance the mixing. The approach taken here is to take a bubble shape that is reasonable based on fluid mechanical studies and solve the Navier-Stokes equation for velocity and the convective diffusion equation for concentration. This is possible because the capillary number is very small so that the shape of the ends of the bubble will be closely approximated by spherical caps. The flow is steady, but the concentration equation is transient. Results show the time approach to an average concentration and variance, for a variety of parameters, shapes, and sizes of bubbles, Peclet number, and leakage rate. Finally the results are compared with those expected from a T-sensor without bubbles. Both 2D and 3D cases are considered.

2. Method

A typical geometry is shown in Figure 1 for a 2D case. In reality the bubbles move with a velocity downstream and most of the liquid follows, with some entering and escaping through the thin liquid layer at the bubble sides. For ease in

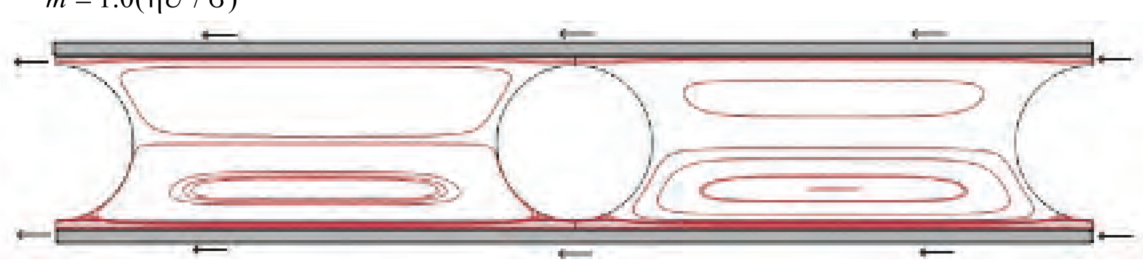

Figure 1. Streamlines between the bubbles (total length 6, height 1)

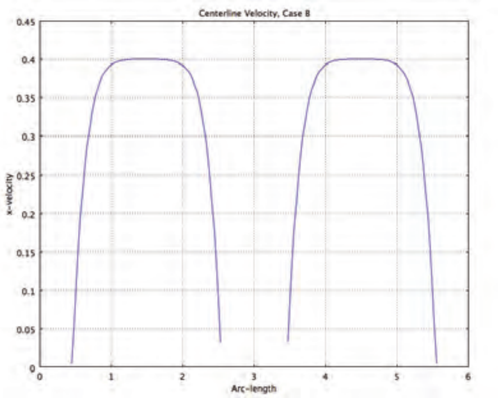

Figure 2. Flow in x-direction along mid-plane

computation, the problem is solved in a coordinate system moving with the speed of the bubbles. The equations are made non-dimensional using the following standards: velocity – bubble velocity, U_b, distance – width of the channel, time – width of the channel divided by the bubble velocity. The non-dimensional Navier-Stokes equations then are solved in Comsol Multiphysics using eta = 1, rho = 1, which corresponds to Re = 1. The shape remains the same and does not move, since the bubbles remain stationary in the moving coordinate system. The boundary conditions for the fluid velocity on the walls are no slip, which requires tangential velocity = –1. Slip is assumed on the bubble surfaces, as an approximation due to the fact that the viscous stress supported by the gas bubble is very small when the capillary number is small. At the exits, where some liquid escapes through the narrow channel, the pressure is set to zero. At the inlet gaps, the velocity must satisfy the following conditions: –1 on the solid boundary, zero slope on the bubble boundary; in addition, one can specify the average flow rate through the gap, which will be a small number since only a small part of the fluid is 'left behind'. Thus, the velocity at the inlet is taken as a quadratic function of the vertical dimension satisfying these conditions. The gap thickness on each side is taken as 5% of the total thickness, unless otherwise indicated.

Figure 1 shows streamlines for such a flow with the total flow rate through the gaps of –0.06667 (compared to 1 for the bubble velocity). What happens is that two recirculation zones are established. In the bottom one, flow goes to the left near the bottom, comes to the last bubble and most of the flow goes up and moves then to the right. A similar thing happens in the top. The reason the recirculation occurs is that only part of the fluid can exit the gap; most of the fluid is carried along with the bubble. The velocity along the mid-plane is shown in Figure 2; it is zero at the edge of the bubble but mostly a constant in between. Conditions for the 3D cases are described below.

Once the flow is determined, it is used in the transient convective diffusion equation. The boundary conditions are zero flux on all walls and bubble surfaces, convective flux out on the left, and a fixed concentration at the inlets. Two cases are considered; in the first, representing mixing described by McMullen and Jensen,[7] the concentration on the bottom inlet is 1 and on the top it is zero. Also, the initial condition is concentration = 1 in small region at the bottom, of the same thickness as the gap, and zero elsewhere. The other case, for comparison with an T-sensor, sets concentration = 1 in the bottom half and zero in the top half. Most cases are solved using a Peclet number of 1000, but some are done with values up to 10,000. Once the problem is solved using Comsol Multiphysics, the average concentration and variance are plotted versus time for both the left and right sections. The average concentration is taken over the fluid region

$$cavg = \int_{A_f} cdA / A_f$$

and the variance is

$$\text{variance}=\int_{A_f} \left((c - cavg)^2 \right) dA / A_f$$

3. Two-dimensional results

Consider first the case illustrated in Figure 1. The bubbles are a distance of 3 channel widths apart, center to center. The average concentration and variance are shown as a function of time in Figure 3. It takes approximately 200 to 300 dimensionless time units to reach steady state. Thus, the actual time is $300 x_s / u_s$. In that time, the bubble travels 300 dimensionless distance units, or 300 times the channel width. Thus, it travels about 100 times the distance between the bubbles before steady state is achieved. Note also that although

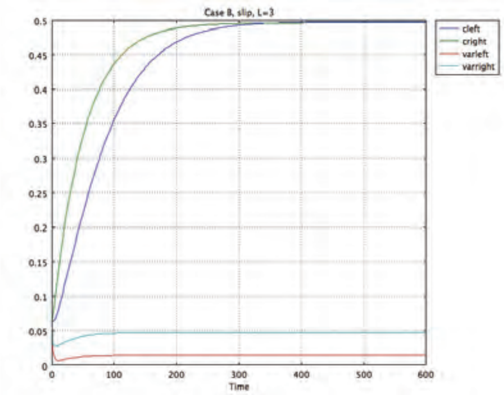

Figure 3. Average concentration and variance on each side of the center bubble

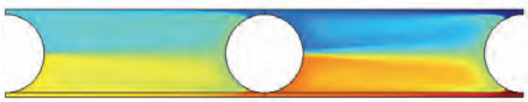

Figure 4. Concentration distribution in fluid

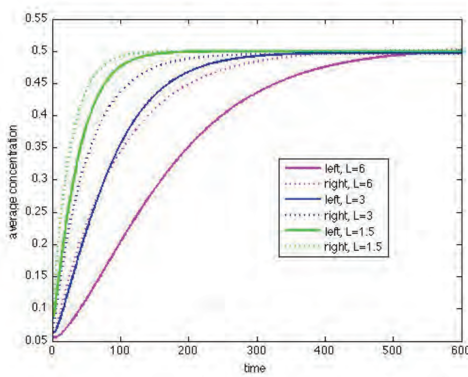

Figure 5. Average concentrations for bubbles separated by lengths 1.5, 3, and 6 (center to center)

the average concentration on both sides approaches 0.5, the variance approaches an asymptote and does not continue to decrease. This behavior is representative of all the cases shown here and is the major insight. A surface plot of the concentration at steady state is shown in Figure 4; this plot does not change even if the time is extended.

Next consider what happens if the bubbles are closer together or further apart. The average concentration is shown in Figure 5 and the asymptotic variance is listed in Table I. It is clear that for bubbles closer together the variance is higher but it doesn't take as long to reach steady state.

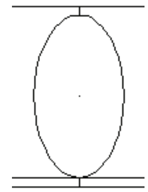

Figure 6. Flattened bubble shape

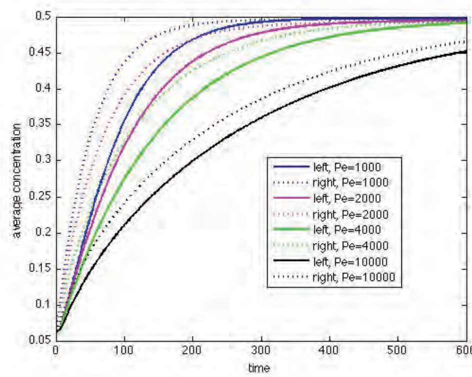

Figure 7. Average concentrations for cases with different Peclet numbers

Next consider what happens if the bubbles are elongated. Here the bubble is taken as a hemisphere on the end joined by a cylinder. Bubbles of total length (from left-most point to the right-most point) of 1, 3, 5, and 9 are considered; the distances between the right-hand side of one bubble and the left-hand side of the next bubble to its right is 5, 4, 3, and 1; with the spherical caps the lengths are 6, 5, 4, and 2. The approach to steady state is about the same for all these cases, although the variance increases as the length of the bubble increases (Table I).

The effect of shape is elucidated by taking a flattened shape as illustrated in Figure 6. This had little effect, which partially justifies the approach taken here of assuming a shape and solving the flow problem and convective diffusion equation.

The effect of Peclet number is shown in Figure 7, and the asymptotic variance is listed in Table I. Clearly as the Peclet number increases, it takes longer for the fluid to become well mixed and the steady state variance is larger. This occurs because the diffusion across the mid-plane is slower.

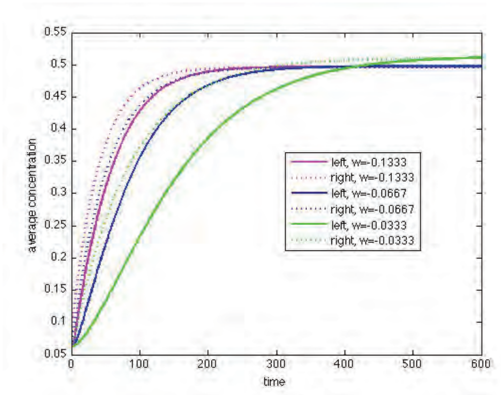

Figure 8. Average concentrations for cases with different leakage rates: half, standard, double

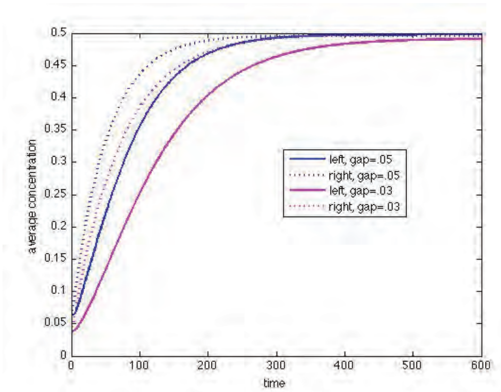

Figure 9. Average concentrations for cases with different gap thicknesses: 0.03, 0.05

The effect of the leakage rate is studied for a case with twice the value and half the value as in the first case. The average concentration is shown in Figure 8, and the asymptotic variance is listed in Table I. As the leakage rate increases, the variance increases and the time to steady state decreases. This effect results because at the higher leakage rate, fluid will leak out before it has had a chance to be circulated between the bubbles. The same thing is true for the narrower gap. The effect of a thinner gap is shown in Figure 9, and the asymptotic variances are listed in Table I for two gap sizes and two leakage rates. As the gap size increases, the variance increases and the time to steady state decreases. The smallest variances are for smaller gaps and small leakage rates. The leakage rate of –0.01 is an average of what is predicted by Taylor[3] (-0.012) and Bretherton[4] (-0.0072) for this Capillary Number (0.00014 for air/water).

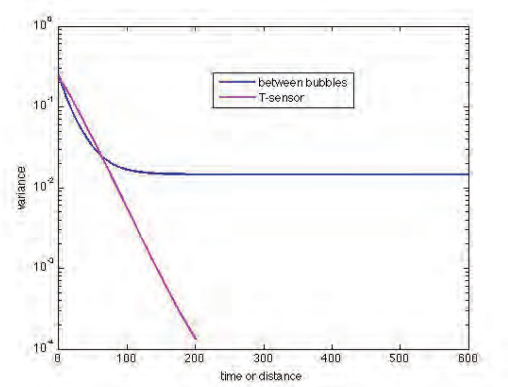

Figure 10. Variance when fluid concentration starts at 1 in the bottom half, 0 in the top half

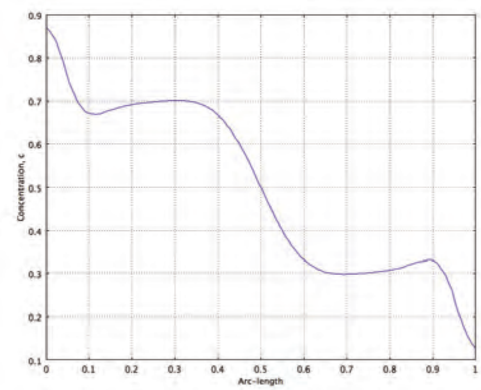

Figure 11. Concentration distribution halfway between the bubbles, $t = 600$

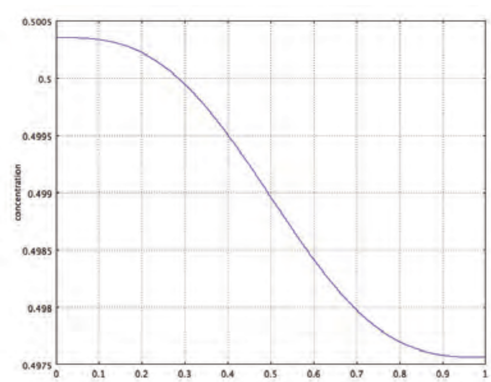

Figure 12. Concentration distribution at the end of the T-sensor, $L = 300$

It is also of interest to compare the mixing between two bubbles with the mixing that would occur in a T-sensor. For the bubble case, the fluid takes an initial concentration of 1.0 in the bottom half and 0.0 in the top half. The average concentration remains at about 0.5 for all time; the variance is shown in Figure 10; and the concentration distribution at the end of the time, halfway between the bubbles is shown in Figure 11. Figure 10 also gives the corresponding results for a T-sensor. The length of the T-sensor was taken as 300, so that the average time spent in the T-sensor would be half that in the bubble case; excellent mixing occurs long before L = 300. Figure 12 shows the concentration distribution near the end of the T-sensor; note the difference in scale between Figures 11 (0.8 for bubbles) and 12 (0.003 for the T-sensor).

4. Three-dimensional Results

A three-dimensional case was solved with spherical bubble as shown in Figure 13. The diameter of the sphere was taken as 90% of the channel width. As in the two-dimensional cases, the wall velocity was –1, slip was allowed along the bubble surface, and the outlet conditions were zero pressure. At the inlet, it was desired to have the same leakage rate, -0.01 as in the two-dimensional cases (T and U). To achieve this, a two-dimensional problem was solved as follows:

$$\nabla^2 u = f, u = -1 \text{ on outer walls (square)}$$
$$\partial u / \partial n = 0 \text{ on bubble surface (circle)}$$

The value of *f* was chosen so that the integral of *u* over the domain was –0.01. The velocity is shown in Figure 14. Note in particular that the velocity is much larger in the corner region than in the small gap. In Comsol Multiphysics, an integration coupling variable was used by solving this problem in two dimensions and using it for the inlet velocity in the three dimensional problem. The exit velocity was similar. This problem was, of course, much bigger than the two dimensional case. The flow problem was solved using 33306 elements and 159,833 degrees of freedom, and this showed the same kind of recirculation that occurred in 2D. The initical conditions for concentration are c = 1 in one half and c = 0 in the other half between each set of bubbles. The concentration problem was not solvable for a Peclet number of 500 or

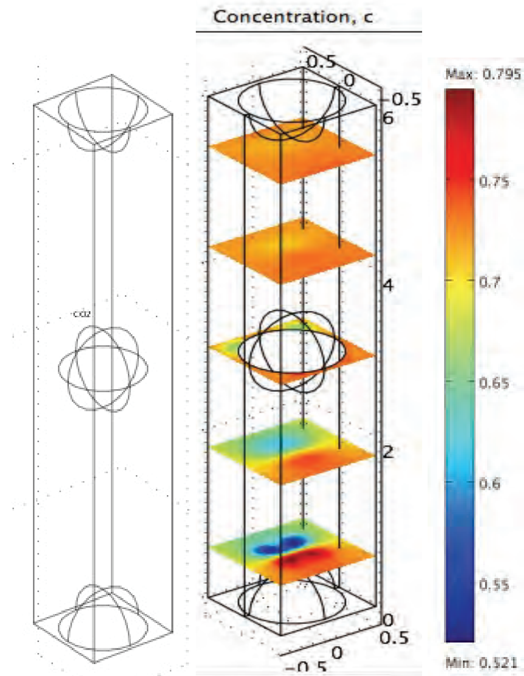

Figure 13/15. 3D geometry/concentration

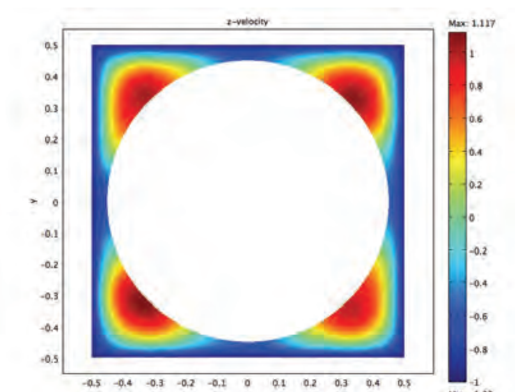

Figure 14. Inlet velocity to 3D channel

1000 even when using 106,603 elements and 156,526 degrees of freedom (for concentration). Thus, solutions are shown for *Pe* = 200.

The concentration profiles are shown in Figure 15. In the 3D case, the average concentration continues to increase past 0.5 (Figure 16), presumably because reagent is coming in faster than it can leave. The variance decreases and then increases slightly in time (Figure 17). The ultimate variance at t = 600 is 0.002, which can be compared with the value for

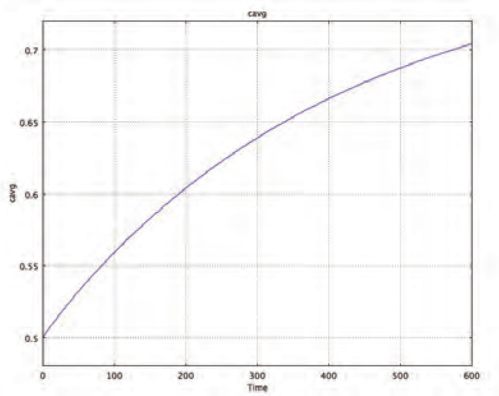

Figure 16. Average concentration in 3D channel, Pe = 200

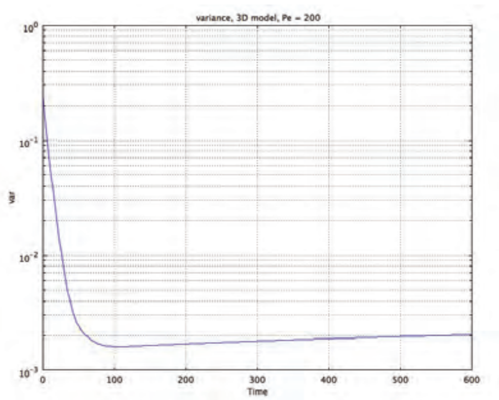

Figure 17. Variance in 3D channel, Pe = 200a similar 2D case (gap = 0.05, leakage rate

the 2D case (gap thickness = 0.05, leakage rate = -0.01, Pe = 200) of 0.0023 on the right and 1.2e-5 on the left. Note that the gap thicknesses are not exactly comparable since they are comparable only at the narrowest part of the gap between bubble and wall in 3D. However, it is interesting that the 2D and 3D results are so similar. If the 3D bubble case is compared with a T-sensor, the variance of a T-sensor is already at 1e-6 for L = 60, which means the fluid in the T-sensor has moved only one-tenth as far as the bubble but has a much smaller variance. Thus, the bubbles, while they do provide mixing, do not provide excellent mixing.

5. Conclusions

The major conclusion is that mixing occurs between the bubbles when they are moving down a channel, but the variance approaches an asymptotic value, which depends upon the parameters. By contrast, in a T-sensor, the variance continues to decrease as the fluid continues down the channel, and the T-sensor provides better mixing for a length equal to the distance the bubbles have moved. In order to get the lowest variance with bubbles, one needs bubbles further apart, spherical (i.e. not elongated), with a low leakage rate in a small gap thickness.

5. References

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Case	L	Re	Pe	gap	leakage	leftvar	rightvar
Effect of length of fluid between bubbles							
A	6	1	1000	0.05	-0.0667	0.0049	0.028
B	3	1	1000	0.05	-0.0667	0.014	0.048
C	1.5	1	1000	0.05	-0.0667	0.055	0.094
Effect of elongated bubbles							
A	6	1	1000	0.05	-0.0667	0.0049	0.028
D	5	1	1000	0.05	-0.0667	0.0073	0.032
E	4	1	1000	0.05	-0.0667	0.012	0.039
F	2	1	1000	0.05	-0.0667	0.043	0.071
Effect of Peclet number							
B	3	1	1000	0.05	-0.0667	0.014	0.048
H	3	1	2000	0.05	-0.0667	0.024	0.058
I	3	1	4000	0.05	-0.0667	0.034	0.063
R	3	10	10000	0.05	-0.0667	0.041	0.063
Effect of leakage rate at two gap thicknesses							
J	3	1	1000	0.05	-0.1333	0.046	0.077
B	3	1	1000	0.05	-0.0667	0.014	0.048
K	3	1	1000	0.05	-0.0333	0.0026	0.021
T	3	1	1000	0.05	-0.0100	4.10E-04	0.010
L	3	1	1000	0.03	-0.0667	0.0044	0.027
U	3	1	1000	0.03	-0.0100	2.80E-04	0.0043
Effect of gap thickness at two leakage rates							
B	3	1	1000	0.05	-0.0667	0.014	0.048
L	3	1	1000	0.03	-0.0667	0.0044	0.027
T	3	1	1000	0.05	-0.0100	4.10E-04	0.010
U	3	1	1000	0.03	-0.0100	2.80E-04	0.0043

Table I. Variances for different cases