Micromixing in a Tubular Flow Reactor Under Simulated Microgravity

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Future human development of space will require unit operations with mixing and mass-transfer considerations similar to those used on Earth. Equipment used for these operations will be exposed to gravitational accelerations different from that of Earth (1 g or unit gravity). Thus, gravity is an additional operating variable which must be considered in the design of such equipment (Nyiri, 1976).

Such unit operations may involve multiphase flows or single-phase flows with density gradients. The nature of these flows, as well as any mass-transfer or chemical reaction taking place within them, should be affected strongly by gravity. Multiphase flows, however, are not well characterized even under unit gravity, to the extent that prediction and measurement of gravitational effects would be difficult.

In this work, a simpler system was chosen involving the mixing and chemical reaction of two aqueous reagent streams in a tubular reactor. While theory predicts that gravity would have no influence on such a flow, the system is simple enough that reasonable comparisons could be made among unit gravity, microgravity, and hypergravity data. The results obtained in using this system could provide groundwork for similar experiments involving more complex, multiphase flows.

In addition, the data obtained in this system provide product yield data for a well-defined chemical reaction in a well-defined flow. By contrast, micromixing experiments using this reaction are typically conducted in a stirred-tank reactor. The relatively homogeneous conditions of turbulent flow in a pipe will be used to compare the predictions of a micromixing model with experimental observations.

Fermentation in Space

One unit operation which will be required in space is the cultivation of microorganisms. Several designs of bioreactors for space applications have been proposed (Seshan et al., 1985; Mayeux, 1977; Petersen et al., 1989; Villeneuve and Dunlop, 1991). These bioreactors will be used as part of a carbon-recycling system on long-term manned space missions, where organic waste will be converted to biomass. The biomass can then be used as a food source or food supplement. One advantage that microorganisms offer as a source of food, compared to plant sources, is a relatively quick recovery in the event of system failure.

In addition to the anticipated problems involved in contacting the gas and liquid phases typically involved in a submerged fermentation (Villeneuve and Dunlop, 1991), it is not known if these reactors will have the same mass-transfer performance in space as on the ground. Design and testing of the bioreactors are primarily ground-based, with the implicit assumption that the processes leading to mass transfer and mixing will not be affected by operation in space. Testing this assumption early in the development of a space bioreactor through relatively simple experiments thus seems scientifically important and cost-effective.

Materials and Methods

NASA's KC-135 parabolic flight testbed

The reaction was carried out aboard NASA's KC-135 parabolic flight testbed. This aircraft provides a simulation of microgravity conditions by flying a parabolic flight trajectory. Accelerations equivalent to 10^{-2} g to 2 g occur during the parabola. At the top of each parabola, the downward acceleration of the aircraft matches that of the Earth's gravity, and a net effect of "zero-g" is produced for approximately 20 seconds. While this effect is dynamic, it is approximately constant at 10^{-2} g for 20 s. At the bottom of each parabola, the effect is 2g as the aircraft pulls out into the next parabola. Each flight consists of 40 such parabolas.

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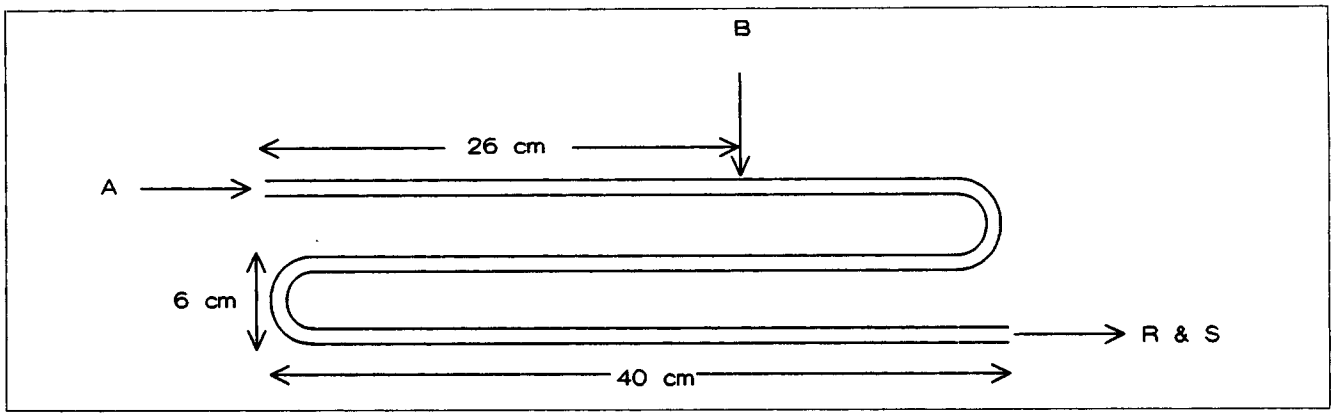

Figure 1a. Experimental reactor.

Chemical test reaction

The chemical test reaction used to make the micromixing measurements was the reaction of 1-naphthol with diazotized sulfanilic acid (Bourne et al., 1981, 1985; Bourne et al., 1990). The use of this reaction as a micromixing test reaction is well documented in these references. The reaction can be adequately described by the competitive, consecutive scheme in Eq. 1 (Wenger et al., 1992):

$$A+B\rightarrow R$$
$$R+B\rightarrow S \qquad (1)$$

Solution A consisted of 0.0578 mol/m^3 of 1-naphthol, and 12.1 mol/m^3 each of Na_2CO_3 and $NaHCO_3$ dissolved in double distilled water. Solution B consisted of 0.550 mol/m^3 of diazotized sulfanilic acid, synthesized from sulfanilic acid the morning prior to the flight. The product distribution, defined by Eq. 2, was analyzed spectrophotometrically using the method described by Wenger et al. (1992).

$$X_S=\frac{2C_S}{2C_S+C_R} \qquad (2)$$

In addition to being sufficiently fast for this type of study (B is completely consumed in less than 0.5 s), the reaction has several features which make it convenient for use aboard the KC-135. First, the chemicals used are in dilute aqueous solution, which makes their use both nonhazardous and inexpensive. The former is especially important in the closed environment of the KC-135. Additionally, the products are stable enough that samples taken on board the aircraft could be analyzed on the ground. This minimized the amount of analytical and data-collecting equipment which had to be integrated into the experimental apparatus.

Experimental apparatus

The experimental reactor is shown in Figure 1. The reactor was constructed of glass with an inner diameter of 0.01 m. Two turbulence triggers in Figure 1a were placed upstream of the injection point to ensure equilibrium turbulence. Each turbulence trigger consisted of two perpendicular glass rods, 3.0 mm in diameter, placed in the flow. The buffered 1-naphthol solution (A) initially flowing in the pipe encountered the B solution which was injected isokinetically through a 0.0032-m-ID injection pipe. The length of the reactor following the injection point was approximately 1 m. The pipe contained two 180° bends to fit this length of reactor in the space available. At the experimental flow rates, the reaction was complete in the residence time of the glass reactor.

Samples of the product solution were taken from a sampling port located downstream of the glass reactor. The samples were drawn by the experimental operator into initially evacuated test tubes through a septum-covered needle (Vacutainer). The tubes were then kept for ground analysis, which took place the same day as the flight.

Reagent solutions A and B were initially stripped of oxygen using nitrogen and were stored in collapsible polyethylene bags during the flight. These bags were evacuated with a vacuum pump before filling to minimize any gas phase in the bag. This

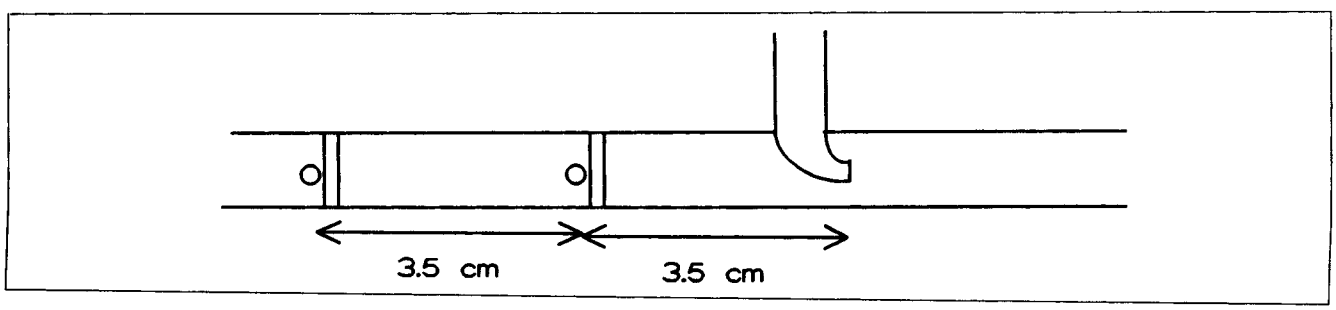

Figure 1b. Expanded view of the injection location showing turbulence triggers.

is necessary as the location of the gas phase relative to the liquid phase is undetermined in reduced-gravity conditions and could be at the outlet of the bag. Waste product solution was similarly stored in collapsible bags.

Solution A was pumped by pump A, an external gear pump with Teflon gears (Micropump). Solution B was pumped by a peristaltic pump (Masterflex). Pump A was located at the same height as the reagent bags to minimize changes in the inlet head during the flight. The speed of pump A was monitored with a tachometer and remained constant throughout the flight.

The reaction was carried out in continuous mode with $F_A/F_B = 10$. Two flight experiments were conducted, at Reynolds numbers of 5,300 (flight 1) and 8,600 (flight 2). The mass of the fluids involved was sufficient to maintain the temperature within 1°C of 24 C for flight 1 and within 0.5°C of 26°C for flight 2. Although 40 parabolas were available for each flight, problems were encountered in managing the large amounts of reagents needed for continuous operation over such a time period. Consequently, the data for flight 1 are based on six parabolas, while the data for flight 2 are based on three parabolas.

Kinetic analysis of the test reaction

One consideration that had to be made in the design of the reactor was to ensure that the reaction was complete during the residence time of the reactor. This is especially important since the flow rates to be used were high and the product distribution was to be analyzed off-line. Using the second-order rate constants measured by Bourne et al. (1985), the reactor was modeled as a series of CSTRs. Conversion vs. length was thus predicted in the kinetic regime. This analysis, Figure 2, shows that the reaction is 99.9% complete well before the end of the 1-m reactor even at flow rates substantially higher than those used here. Although such an analysis necessarily assumes instantaneous micromixing, it was assumed that the rate of micromixing did not change the conversion vs. length profile substantially. Visual observations of the reactor in operation confirmed that the reaction was complete, as any visible color changes with length occurred within the first half of the reactor.

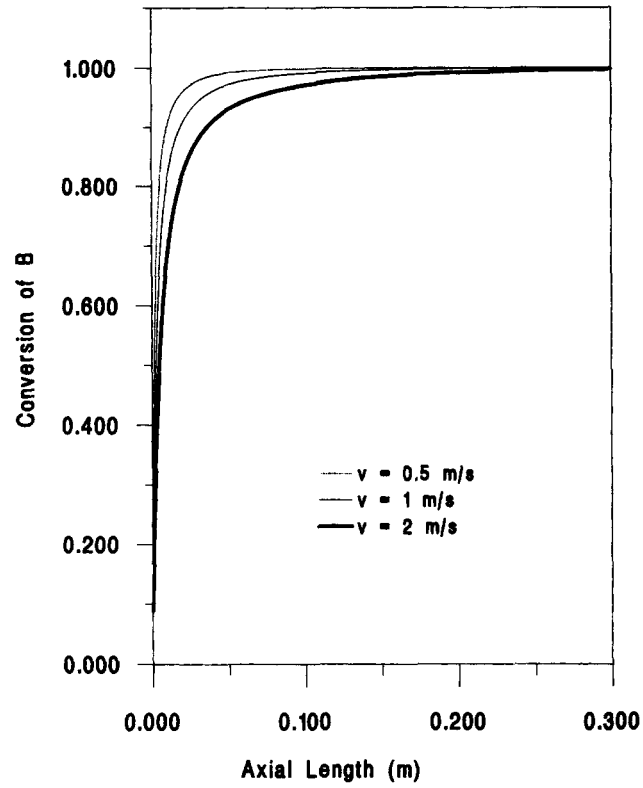

Figure 2. Simulated conversion-length profile for the tubular flow reactor at velocities of 0.5, 1 and 2 m/s.

Reactor was modeled as a series of CSTRs with the reaction operating in the kinetic regime.

Results and Discussion

Effect of gravity on micromixing

Figure 3 shows X_S as a function of Reynolds number for the high and low g portions of the parabola compared with ground-based data. Ground-based data are based on ten different samples taken at the same conditions as the flights. Within the reproducibility of the analysis, it is apparent that gravity had no significant effect on the product distribution. Analysis of variances among the low g, ground, and high g data for the two flights indicated that the differences were not significant at the 5% level. Analysis of variance for the ground data as a function of Reynolds number was significant at the 1% level. As the large amount of scatter for the high g portion of flight 1 was due to two outlying points, it is possible that more data would have revealed a significant effect.

Analysis via micromixing model

One advantage of performing micromixing experiments in a tubular reactor is that the turbulence in such a reactor is relatively homogeneous, compared to a stirred-tank reactor where these experiments are usually performed. In addition, a pipe is a relatively simple geometry for which there is abundant pressure drop (energy dissipation) data available. Thus, the ground-based experiments provide a convenient method of testing the predictions of turbulence parameters by a micromixing model.

The E-Model by Baldyga and Bourne (1989a) treats the rate of vortex formation in the viscous-convective subrange as the controlling rate in the process of micromixing. By this mechanism, two fluid elements of different concentration engulf each other and form a structure in which lamellar stretching, diffusion, and reaction can occur. This model is convenient to use, as the model equations consist of ordinary differential equations and the single parameter E can be related to the rate of turbulent energy dissipation using turbulence theory. For instance, with a given experimental value for X_S, trial and error adjustments of E can be made to give a predicted X_S which matches the experimental data. E can then give an estimate of the rate of energy dissipation ϵ in W/kg, using the equation from Baldyga and Bourne:

$$E = 0.05776\left(\frac{\epsilon}{\nu}\right)^{1/2} \tag{3}$$

Once ϵ is determined, it can be used to calculate the Kolmogoroff microscale using the equation:

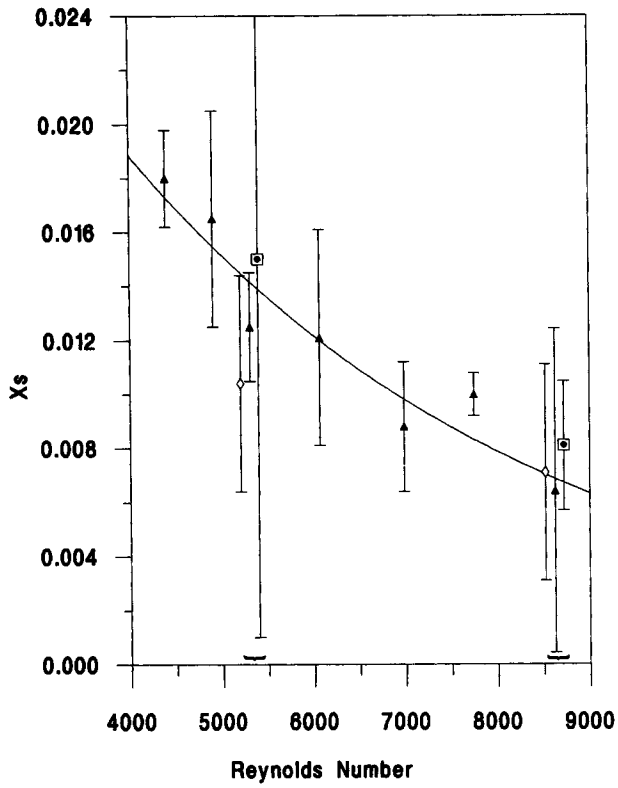

Figure 3. Measured product distribution X_S as a function of Reynolds number in the pipe.

▲, data taken in ground-based experiments; ◇, data taken under low g conditions; ⊡, high g conditions.

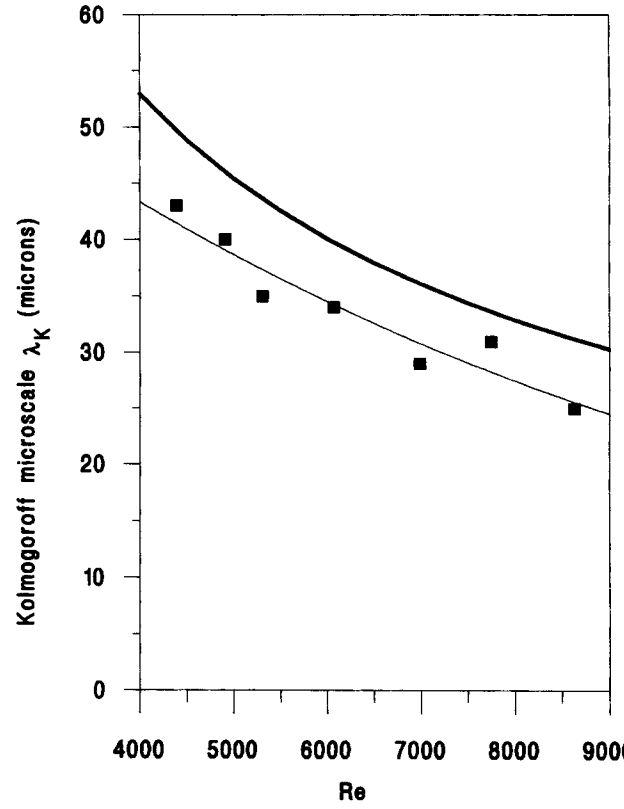

Figure 4. Kolmogoroff microscale as a function of Reynolds number in the 1-cm pipe.

Thick line, microscales predicted from Eq. 6; ■, predictions made by applying the experimental values of X_S to the E Model to determine ϵ in the pipe.

$$\lambda_K = \left(\frac{\nu^3}{\epsilon}\right)^{1/4} \tag{4}$$

Using the E-Model for the case of a continuous reactor as described by Baldyga and Bourne (1989b) (taking self-engulfment of fluid elements into account), λ_K was predicted based on X_S at the experimental Reynolds numbers. These results are shown in Figure 4.

To check the predictions of λ_K as a function of Reynolds number it is possible to calculate the energy dissipation in the pipe based on standard empirical correlations. For turbulent flow ($3{,}000 < Re < 10{,}000$) in a smooth pipe, the Blasius equation applies (Churchill, 1977):

$$f = 0.079(Re)^{-0.25} \tag{5}$$

Thus, at a given flow rate, the energy dissipated per unit mass can be calculated from the pressure drop. For a smooth pipe of arbitrary length, λ_K can be expressed as a function of Reynolds number by Eq. 6:

$$\lambda_K = \left(\frac{D^4}{0.158Re^{2.75}}\right)^{1/4} \tag{6}$$

Predictions of λ_K as a function of Reynolds number using this approach are also given in Figure 4, which shows that the two methods give similar results, with the predictions based on the Blasius equation higher by a nearly constant value of 5-7 micron. One source of energy input neglected in this approach is that caused by flow past the turbulence triggers. It is arguable that additional turbulence caused by these obstructions was being dissipated in the reaction zone, thus decreasing the microscale of turbulence as measured by the reaction.

Conclusions

The results from two flights on the KC-135 indicate that the micromixing intensity in this system was not affected by simulated low and high gravity. This is not surprising, as gravity theoretically should have no effect on a single-phase, constant-density flow. The results obtained confirm that fluid mechanical concepts developed on Earth are applicable to other gravitational environments. This conclusion should also apply to other phenomena in single-phase, constant-density systems, in which fluid turbulence plays a role. In addition, the reaction used in this work would be appropriate for investigating gravitational effects on mixing in multiphase systems.

Application of the micromixing model of Baldyga and Bourne to the experimental situation showed that the model gives reasonable predictions of the Kolmogoroff length scale when compared to empirically based calculations. This is significant because of the relative simplicity of the model and its ability to predict a fundamental turbulence parameter.

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Notation

A = 1-naphthol or its reagent solution
B = Diazotized sulfanilic acid or its reagent solution
C_i = concentration of species i, (mol/m^3)
D = pipe diameter, m
E = engulfment rate, 1/s
f = Fanning friction factor
F = flow rate, m^3/s
R = 4-(1,4-sulfophenylazo) 1-naphthol
S = 2,4 bis(1,4-sulfophenylazo) 1-naphthol
X_S = product distribution

Greek letters

ϵ = turbulent energy dissipation rate, W/kg
ν = kinematic viscosity, m^2/s
λ_K = Kolmogoroff length scale, m

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