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Catalysis Today 105 (2005) 667–672
CATALYSIS TODAY
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The pressure drop experiment to determine slug lengths in multiphase monoliths

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Available online 11 July 2005

Abstract

The length of the liquid slugs, that separate the elongated bubbles in Taylor flow, is an important parameter for mass transfer, flow stability and pressure drop in capillary microchannels. In this work, pressure drop measurements are used to determine the length of slug in Taylor flow in downflow monoliths. The method is sensitive if the slugs are relatively short, less than 10 times the channel diameter. The pressure drop measurements are a cheap and fast alternative to tomographic or electric methods. Experiments using different distributors indicate that the slug length varies significantly with changes in the hydrodynamics in the feed section of the monoliths. Slug length correlations that are based on parameters inside the channels can therefore not safely be used for a different setup. As a result, the slug length should be measured in each experimental setup, which makes a inexpensive and robust method to do so very welcome.

Keywords: Pressure drop; Taylor flow; Monoliths; Multiphase reactors; Fluid mechanics

1. Introduction

The segmented flow pattern of elongated bubbles and slugs in microchannels, Taylor flow, has many features that are advantageous for chemical processing. In particular, when a catalyst is applied on the channel walls, very high mass transfer rates [1,2], fast liquid mixing [3], absence of internal diffusion limitations [4], plug flow [5–8] and low pressure drop [9,10] can be combined.

For commercial applications, the microchannels must be scaled up to accommodate the flowrates required in industry. The microreactor community, which attempts to bring the cheap microscale mass-production of the electronics industry to chemical processing, has heralded scaling out or *numbering up* as the method of choice. Most of the solutions currently under investigation involve cascades of T-junctions and similar manifolds, combined with sections of very small channels to ensure equal flowrate due to pressure drop [23]. For small volume processes, several microchannels may indeed be enough; on the other hand, reliable feed sections that can economically and reliably feed many ($\gg 10^3$) multiphase channels by flow splitting have yet to be developed.

The multiphase monolith reactor is a different technological option to scale out microchannels. In monoliths, a gas/liquid distributor must feed all channels – within design limits – the same amount of gas and liquid, and the reactor must be constructed in such a way that the channels behave more or less the same. In contrast to microreactors, channels are not fed individually, and the proper choice of distributor is vital. Early attempts at using monoliths in multiphase systems exhibited significant maldistribution [5]. In particular, the design of upflow distributors was found to be very problematic if not impossible [11]. For this reason, downflow is preferred in industry [12]. In a recent paper [6],

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0920-5861/$ – see front matter
doi:10.1016/j.cattod.2005.06.007

Nomenclature

a	interfacial area (m^2/m^3)
d	diameter (m)
g	gravitational constant (m/s^2)
L	length (m)
P	pressure (Pa)
u	velocity (m/s)
U	sum of gas and liquid superficial velocity (m/s)

Greek letters

γ	surface tension (N/m)
ϵ	holdup
μ	viscosity (Pa s)
ρ	density (kg/m^3)

Dimensionless groups

Ca	Capillary number ($=\mu\ U/\gamma$)
f	friction factor
Re	Reynolds number ($=\rho\ Ud/\mu$)

Subscripts

B	bubble
G	gas
L	liquid
s	superficial
TP	two-phase
UC	unit-cell, i.e. a bubble and a slug

we have demonstrated with a slug length-dependent pressure drop model that downflow operation can be stable and that upflow is prone to hydrodynamic instability.

The channels in monoliths are so regular, and the Taylor flow pattern is so well defined, that design equations of high accuracy are quite possible. There is one big ''if'': the length of the bubbles and slugs is important, and must be known for design purposes. Horvath et al. [13] demonstrated that the pressure drop depends on the slug length, and now the underlying reason for the slug length dependence, Laplace pressure terms, are well understood [10,14–16]. The slug length dependence of the pressure drop in a single channel has profound implications on a reactor scale, where the analysis of stability and residence time distribution begins with realising that in all channels the pressure drop is the same. Horvath et al. also demonstrated experimentally that the liquid–solid mass transfer depends on slug length. Berčič and Pintar [17] demonstrated in a single channel the slug length dependence for gas–liquid mass transfer. Recent work by van Baten and Krishna [2] has improved the analysis, and their model also requires the bubble length to be known.

In this paper, we consider experimental techniques to determine the lengths of slugs in multiphase monolith reactors. In a previous work [18], we have reported slug's lengths for one type of distributor in one type of setup. These experiments were performed using electrodes and required considerable experimental effort. More recently, Gladden and co-workers used MRI tomography [19–21] and reported slug and bubble lengths in many channels simultaneously.

The first aim of this work is to present how pressure drop can be used to accurately estimate the slug lengths in monolith reactors. Various attempts have been made to find agreement for slug length data obtained in different setups [22] by formulating correlations based on the hydrodynamics inside the channels, such as bubble velocity and channel diameter. The failure of such attempts indicates that the hydrodynamics in the feed section, which vary widely from setup to setup, in fact determine the length of bubbles and slugs in the channels. The second aim of this paper is to report the significant impact of distributor type on slug lengths, and we demonstrate the importance of proper distributor design for a successful scale-up of microchannels.

2. Theory

If the design equations for most of the phenomena in microchannels depend on slug or bubble length, one should be able to use these 'design' equations to calculate the slug length from experimental data. Preferably, this inverse problem is (1) well-defined and (2) experimentally simple and cheap to perform. The current state of modelling gas–liquid mass transfer still leaves some doubt to the effect of slug length on k_La, and because of the high mass transfer rates it requires very accurate dissolved-gas sensors. Liquid–solid mass transfer experiments require treatment of the channel with a coating like benzoic acid that is to be dissolved in experiments, which is cumbersome experimentally. Liquid-to-catalyst or gas-to-catalyst mass transfer under reacting conditions requires coatings of very active catalysts and such experiments require significant experimental effort.

Pressure drop measurements have the benefit of being both simple to perform and very sensitive to slug length. In Taylor flow, surface tension effects dominate over viscous effects ($Ca \ll 1$), and differences in curvature between the front and the rear of the bubble give rise to a Laplace pressure difference that is significant with respect to the viscous losses in the slug. Kreutzer et al. [10] used different liquids to independently vary Re and Ca in a single channel setup that allowed the independent variation of bubble and slug length. Pressure drop measurements ($\Delta p/L$) were correlated, after correcting for the static head $\rho g\epsilon_L$, as an additional term in the friction factor for the slugs

$$fRe = \frac{(\Delta p/L - \rho g\epsilon_L)d^2}{2\mu U\epsilon_L} = 16\left[1 + \frac{0.17d}{L_{slug}}\left(\frac{Re}{Ca}\right)^{1/3}\right]. \quad (1)$$

where Ca and Re are based on the sum of gas and liquid velocity $U = u_{Ls} + u_{Gs}$. Note that $Re/Ca = \rho d\gamma/\mu^2$ is independent of velocity and constant for a given liquid in a channel. The additional term is inversely proportional to L_{slug}/d, which means that for long slugs, the inverse calculation of slug's lengths from pressure drop data becomes inaccurate. For instance, for water in a 2 mm channel (fRe) doubles from 16, the limit of infinitely long slugs, to 32 at (L/d) = 10, to 48 at (L/d) = 5, and to very high values as the slug length decreases further. The additional term is related to surface tension, so for organic liquids the values are lower than for water, but even for a viscous organic liquid the sensitivity is high enough for slugs shorter than seven times the channel diameter.

Apart from the slug length, all variables in Eq. (1) are either known at the start of an experiment (d, fluid properties) or easily measured outside the monolith (u_{Ls} and u_{Gs} can be easily obtained from the feed flowrate, open frontal area and column diameter).

3. Experimental

The 200 cells per square inch (cpsi) monolith (d = 1.56 mm) blocks of 10 cm diameter were mounted in a column. The monolith column was filled with 2 blocks of 0.13 m on top and below these two blocks a 0.24 m block was mounted. The blocks were loosely stacked on top of one another. The blocks were sealed inside the column using Teflon tape, which prevented by-pass of gas or liquid. Water and air were distributed on top of the column with one of four different distributors. Two FullJet nozzle-type distributors from Spraying Systems were used, one with a 10 mm opening (large nozzle) and one with a 8 mm opening (small nozzle). Two shower-head distributors of 8 cm diameter were used. The first shower-head had approximately 100 holes of 1.5 mm diameter (coarse shower-head), the other had approximately 200 holes of 1.0 mm diameter (fine shower-head). All distributors were positioned at a height above the column, such that the expanding cone (nozzles) or the outer liquid jets (shower) just hit the column wall.

Platinum microprobes were mounted on opposite sides in several channels, and an alternating current was applied and the voltage difference was measured with a frequency of 10,000 Hz, such that the passing of a slug could be measured by decrease in voltage. One pair of electrodes was located in a channel 2 cm from the column wall and another pair was mounted in a channel in the middle of the column. The results from the two probes were identical within experimental error, and here we only report the results from the probe in the center. From the time-series of the voltage signal, the amount of time for a bubble or slug to pass the electrode could be recorded (see Heiszwolf et al. [18] for an example time-series). The velocity of the bubbles and slugs was estimated assuming zero slip as:

$$U = u_{Ls} + u_{Gs}. \tag{2}$$

From the time t_{slug} in which a bubble or a slug passed the electrode, the slug length was calculated using U by:

$$L_{slug} = \frac{U}{t_{slug}}. \tag{3}$$

The pressure difference between the headspace above the distributor and the bottom of the column was measured in several experiments simultaneously with the electrode measurements. The pressure drop over the monolith is very low compared to standard packings, and pressure losses due to the distributor section was substracted from the measured pressure drop to obtain the pressure drop over the monolith blocks themselves. The pressure losses due to the distributor were, based on Bernouilli's equation, proportional to the square of the liquid superficial velocity [9]:

$$\Delta p_{dist} = C_{dist} \frac{A_{jet}}{A_{col}} \frac{1}{2} \rho u_{Ls}^2 \tag{4}$$

The proportionality constant C_{dist} was different for each distributor. The validity of this approach was checked by checking that the pressure drop per meter length was the same for a single monolith block and three monolith blocks (Fig. 1).

4. Results

Slug lengths were measured for 0.02 m/s $< u_{Ls} <$ 0.2 m/s and 0.02 m/s $< u_{Gs} <$ 0.3 m/s during a period of about 5–10 s. The values reported here are averaged over this period. There was considerable variation in the bubble and slug length, standard deviations of 30% of the full value

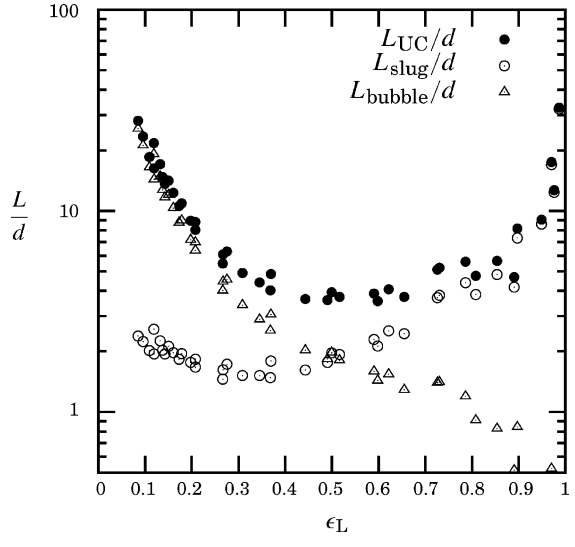

Fig. 1. Unit-cell length, bubble length and slug length as a function of holdup. Data from electrode measurements using the coarse distributor.

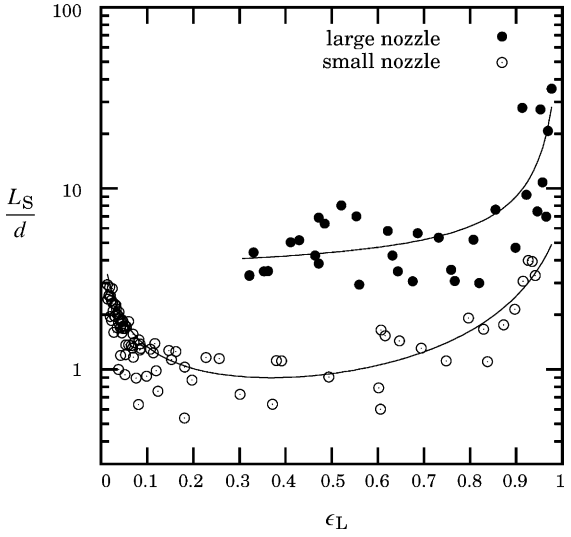

Fig. 2. Slug length vs. liquid holdup for the nozzle distributors. The slug lengths were determined using the electrodes.

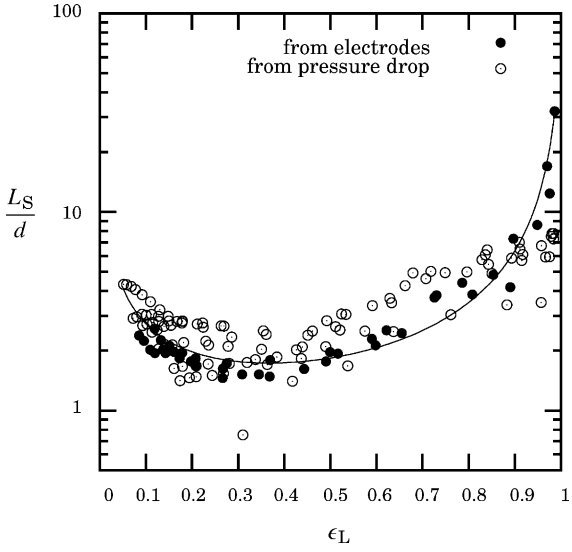

Fig. 4. Slug length vs. liquid holdup for the coarse shower-head distributors, determined directly using the electrodes and indirectly from the pressure drop.

were not uncommon. The holdup was calculated from the superficial gas and liquid velocity by assuming that the liquid holdup in the film separating the bubble from the wall is minimal:

$$\epsilon_L \approx \frac{L_{slug}}{L_{slug} + L_{bubble}} \approx \frac{u_{Ls}}{u_{Ls} + u_{Gs}}. \quad (5)$$

In Figs. 2 and 3, the length of the slugs (made dimensionless by dividing by the channel diameter) is plotted versus the liquid holdup in the channel.

Fig. 4 shows the dimensionless slug length, obtained by the different methods.

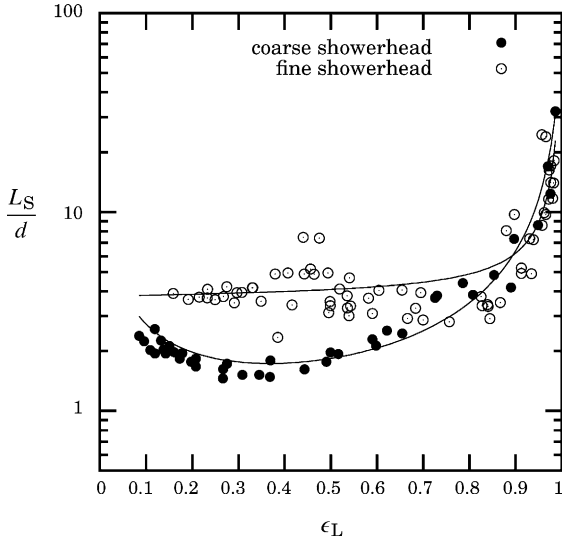

Fig. 3. Slug length vs. liquid holdup for the shower-head distributors. The slug lengths were determined using the electrodes.

5. Discussion

All measurements indicate that for $\epsilon_L \to 1$, the slug length increases. At very high liquid holdup, the gas feed is simply very low; the liquid droplets flood the top of the monolith and coalesce to large drops before entering the monolith column. Fig. 1 shows that for the bubble length similar observations can be made: at low liquid holdup the length of gas bubbles that form in the time between two droplets entering the channels becomes large. At very low holdup, it seems that the slug length increases slightly as well.

5.1. Pressure drop versus conductivity

The agreement of the slug length obtained from pressure drop and the slug length obtained using the electrodes is very good. Fig. 4 was calculated using the single channel correlation Eq. (1), except that the long-slugs limit valid for circular slugs, 16, was replaced with the limit value of 14.2, which holds channels of square cross-section.

The observed values of the slug length found in monoliths is clearly in the range where the pressure drop is sensitive to slug length variations. If the slugs are shorter than seven times the channel diameter, the slug length can be determined from the pressure drop. Along the same line of argumentation, it is not surprising that the very long slugs at $\epsilon_L \to 1$ could not be resolved: for such long slugs the value of *fRe* becomes close to the limit value for infinitely long slugs, and the method loses its sensitivity.

The most important merit of the pressure drop method is that it is very fast and cheap to perform. The careful glueing of the electrodes in the 200 cpsi monoliths was very labour

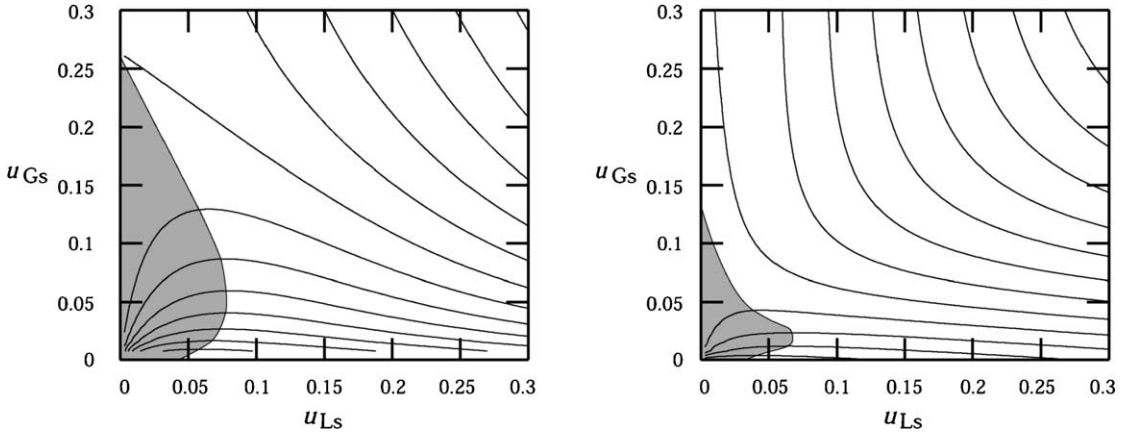

Fig. 5. Impact of the slug length on the (un)stable operating range for the 200 cpsi monoliths used in this study. The lines are contour lines of constant pressure (isobars) on a map of u_{Ls} vs. u_{Gs}. The shaded area is unstable. Left: large nozzle distributor. Right: coarse shower-head distributor.

intensive, whereas the pressure drop measurements could be performed in less than a day.

5.2. Different distributors

Clearly, minimal changes in the distributor can lead to large differences in slug length inside the monoliths. For the nozzle, the result was as expected. The nozzle with the large opening creates a slower jet of larger bubbles than the nozzle with the smaller opening. For most of the experiments, the liquid was the dispersed phase on top of the monolith, so one expects that large droplets in this zone become large slugs in the monoliths.

For the shower-head experiments, we were at first surprised that the coarse shower-head gave smaller slugs. Close visual inspection of the top of the monolith column indicated that the large droplets from the coarse shower-head broke up into smaller droplets upon impact on the monolith structure. This might explain the shorter slugs that were found experimentally.

5.3. Impact of slug length

The results clearly indicate that slug length is not an intrinsic characteristic of the monolith structures themselves, but rather depend on the distributor used. As mentioned in Section 1, many phenomena depend on the slug length. It would go to far to discuss the impact of the difference in slug length on all these parameters, and here we restrict ourselves to a discussion of the stable operating conditions.

In the typical hydrogenation reactions for which monoliths are being considered, the overall mass transfer rate of hydrogen improves with a reduction of the superficial velocities in the channels [1]. The velocities in monoliths, even at zero or negative pressure drop, are high in comparison to trickle beds, and designs for monolith reactors tend to be long if full conversion is required in a single pass. For these two reasons, the optimum for superficial velocities in downflow monoliths is the lowest stable value. Kreutzer et al. [6] recently discussed the unstable region, and found that the boundary of unstable region depends on the free-fall velocity, i.e. the two-phase velocity U of downward Taylor flow when the pressure gradient over the column is zero. At u_{ff}, frictional and interfacial pressure terms cancel hydrostatic terms:

$$u_{ff} = \frac{\rho g d^2}{2\mu(fRe)} \tag{6}$$

in which fRe is slug length-dependent. If the slug length is constant, analytical expressions can easily be formulated for the stable region and for the impact of maldistribution on the RTD. For the slug lengths measured here, we have calculated the unstable region numerically based on the theory presented in [6]. In Fig. 5, the stable regions are indicated for the coarse shower-head, which had the shortest slugs, and the large nozzle, which had large slugs.

Clearly, the unstable region is much larger for systems that have relatively long slugs.

6. Conclusions

1. In this work, a simple method to determine the slug length in monoliths, based on pressure drop measurements, is presented.
2. The method is most sensitive for relatively short slugs.
3. The main advantage of the pressure drop measurement is that it is simple, fast and cheap.
4. Measurements for different distributors indicate that the slug length depends on the hydrodynamics outside the monolith channels.
5. Slug length measurements are essential for proper monolith reactor design, especially if the flowrate through the monolith is low.

Acknowledgement

Corning Inc. is gratefully acknowledged for supplying the monolith samples. This work was financially supported by grant DST 66.4653 of the Dutch Foundation for Technological Science (STW/NWO).

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