Available online at www.sciencedirect.com

SCIENCE DIRECT®

Catalysis Today 105 (2005) 667–672

CATALYSIS TODAY

www.elsevier.com/locate/cattod

The pressure drop experiment to determine slug lengths in multiphase monoliths

Michiel T. Kreutzer [a,*], Menno G. van der Eijnden [a], Freek Kapteijn [a], Jacob A. Moulijn [a], Johan J. Heiszwolf [b]

[a] Reactor and Catalysis Engineering, DelftChemTech, Faculty of Applied Sciences, Delft University of Technology, Julianalaan 136, 2628 BL Delft, The Netherlands

[b] Albemarle Catalyst Company BV, Nieuwendammerkade 1-3, P.O. Box 37650, 1030 BE Amsterdam, The Netherlands

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],

* Corresponding author. Tel.: +31 15 278 90 84; fax: +31 15 278 50 06.
E-mail address: kreutzer@tnw.tudelft.nl (M.T. Kreutzer).

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

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