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Design and characterization of bubble-splitting distributor for scaled-out multiphase microreactors

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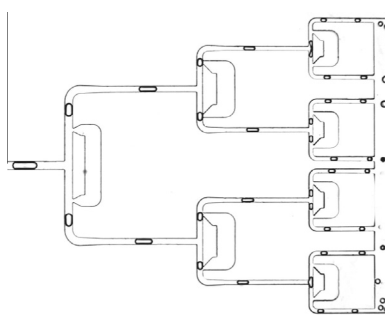
HIGHLIGHTS

- Detailed analysis of the design strategies of a bubble-splitting distributor.
- Identification of the three primary causes of nonuniformities in bubble size.
- Theoretical and experimental analysis of the device's performance.
- Guidelines on how to operate a bubble-splitting distributor.

GRAPHICAL ABSTRACT



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ABSTRACT

This paper reports an analysis of the parallelized production of bubbles in a microreactor based on the repeated break-up of bubbles at T-junctions linked in series. We address the question how to design and operate such a multi-junction device for the even distribution of bubbles over the exit channels. We study the influence of the three primary sources leading to the uneven distribution of bubbles: (1) nonuniformity in the size of bubbles fed to the distributor, (2) lack of bubble break-up, and (3) asymmetric bubble breakup caused by asymmetries in flow due to fabrication tolerances. Based on our theoretical and experimental analysis, we formulate two guidelines to operate the multi-junction bubble distributor. The device should be operated such that: (i) the capillary number exceeds a critical value at all junctions, $Ca > Ca_{crit}$, to ensure that all bubbles break, and (ii) the parameter $(l_s/w) \cdot Ca^{1/3}$ is sufficiently large, with l_s/w the distance between the bubbles normalized by the channel width. More quantitatively, $(l_s/w) \cdot Ca^{1/3} > 2$ for fabrication tolerances below 2%, which are typical for devices made by soft lithography. Furthermore, we address the question whether including a bypass channel around the T-junctions reduces flow asymmetries and corresponding nonuniformities in bubble size. While bubble nonuniformities in devices with and without bypass channels are comparable for fabrication tolerances of a few percent, we find that incorporating a bypass channels does have a beneficial effect for larger fabrication tolerances. The results presented in this paper facilitate the scale-out of bubble-based microreactors.

1. Introduction

Multiphase microreactors have emerged as an attractive class of reactors for the production of fine chemicals and pharmaceuticals [1,2], for the synthesis of micro- and nanoparticles [3–7], and for high-throughput screening applications [8–11]. Besides excellent heat and mass transfer characteristics in microreactors, continuous flow chemistry based on the confinement of reactions in picoliter

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Nomenclature

α_i	design constant for *i*th junction needed in Eq. (6) (–)	l	bubble length (m)
β	design parameter (–)	l_{bp}	bypass length (m)
γ	interfacial tension (N/m)	l_s	slug length (m)
η	fraction of breaking bubbles (–)	$\bar{l}$	average bubble length (m)
μ	dynamic slug viscosity (Pa s)	$\sigma(l)$	standard deviation in bubble length (m)
Ca	capillary number, $\mu v/\gamma$ (–)	m	number of non-breaking bubbles (–)
CV	coefficient of variation (–)	n	number of bubbles (–)
a,b	constants in Leshansky's equation for the critical capillary number	q	volumetric flow rate (m^3/s)
C	constant needed in Eq. (3)	R	hydrodynamic resistance (Pa s/m^3)
$C_1 - C_4$	constants needed in Eq. (4)	$u(x)$	deviation in parameter x
h	channel height of planar device (m)	v_i	average channel velocity (single phase flow) or bubble velocity (two phase flow) in the *i*th generation (m/s)
i	index of channel or junction (–)	w_i	channel width of *i*th generation (m)

to nanoliter bubbles or droplets (a) enhances mixing, (b) reduces axial dispersion, and (c) prevents precipitation at walls and clogging of channels such that higher yields and selectivities are obtained [10,12].

Despite the conceptually simple idea of numbering-up as a strategy to increase throughput, parallelization of segmented flows remains a challenge in practice [13]. One basic approach to increase throughput of segmented flow microreactors is to produce droplets or bubbles in each individual channel [14–24]. With a few notable exceptions [25–27], this approach requires that the supply of the fluids to all these channels is identical, as differences in flow lead to corresponding differences in the volume, frequency, and speed of the bubbles or droplets. Integrating resistive channels upstream of the segmented flow channels minimizes cross-talk between the channels and ensures a constant supply of fluids, which is not affected by the dynamic pressure fluctuations in the segmented flow channels [17,28]. de Mas et al. [17] showed that the pressure drop over the resistive channels should be two orders of magnitude larger than the pressure drop over the segmented flow channels. Fulfilling this requirement is particularly challenging for gas–liquid flows, because the low viscosity of gas requires resistive gas channels that are roughly two orders of magnitude smaller in width than the segmented flow channels. These channels should be fabricated with high precision, as small difference in their hydrodynamic resistance lead to differences in the features of the segmented flows running in parallel.

An alternative approach that does not require on-chip integration of resistive feed channels is to feed a segmented flow to the chip, and split the bubbles or droplets at a series of successive junctions [29–33]. To obtain segmented flows with an identical bubble volume and bubble spacing in all channels downstream the bubble distributor, two key questions need to be addressed: (1) how to ensure breakup at all junctions, (2) how to minimize asymmetries in flow. The first question can be addressed based on the understanding of breakup of bubbles or droplets at single T-junctions. Whether a droplet breaks primarily depends on its length relative to the channel width, l/w, and on the capillary number, Ca [34–39]. Of secondary importance is the viscosity contrast between the two phases [40,41]. The second question can be addressed by considering the differences in hydrodynamic resistances of the channels due to fabrication inaccuracies. As well known for single T-junctions, a difference in velocity in the two exiting arms leads to the asymmetric breakup of bubbles [34,42–45]. Consequently, the size of the bubbles and their distance apart is different in the two exiting arms. For a multi-junction device, Adamson et al. [29] identified a second cause for unequal flow distribution: if bubbles enter downstream T-junctions at times that are not precisely coordinated, the backpressure generated when the bubbles split causes an imbalance in the pressure drops across the two exiting arms of the upstream T-junctions. This also leads to asymmetries in segmented flows. They showed that this source of variation is reduced by designing the system such that the magnitude of the pressure pulses is negligible with respect to the total pressure drop over the branches. Another clever trick to reduce the influence of pressure pulses at downstream T-junctions is to reduce the coupling between the successive T-junctions by incorporating a pressure-equalizer at the T-junctions in the form of a bypass-like structure [32]. Although this concept has been demonstrated, no quantative data is available on the influence on this bypass.

Summarizing the work done on multi-junction bubble and droplet distributors, we conclude that – although there are some pointers on how to design and operate these devices – there is no systematic study how key operating conditions influence the performance, and to what extend polydispersity is reduced by incorporating a pressure equalizer.

In this paper, we start with a discussion on the different design strategies and explain why a design that fixes the relative length of the bubbles or droplets is favorable over other types of design. We then identify three primary sources leading to the uneven distribution of bubbles and systematically study their influence on the uniformity of the size of bubbles in the downstream channels of a multi-junction device. Additionally, we quantify to what extend flow asymmetries are reduced with the use of a pressure equalizer. In short, this paper teaches how to design and operate a multi-junction bubble distributor.

2. Theory on the design and operation of a multi-junction bubble distributor

2.1. Design

Non-breaking bubbles are one of the main sources of polydispersity. A straightforward approach to ensure breakup at all successive junctions is to design the network such that l/w and Ca are kept the same at all junctions. Operating the device above the transition line ($Ca_{\text{crit}} = f(l/w)$) at the first junction then ensures breakup at all successive junctions. But, in the planar networks that are commonly used in the field of microfluidics Ca and l/w cannot be fixed at the same time. This is easily seen from the fact that the flow rate entering a junction hw_iv_i equals twice the flow rate in the two exiting channels $hw_{i+1}v_{i+1}$ that lead to the next junctions, with h the channel height, w the channel width, v the bubble velocity, and i the index of the junction. Hence, $v_{i+1} = \frac{1}{2}\frac{w_i}{w_{i+1}}v_i$. Defining the capillary number based on the bubble velocity, the viscosity of the compartments between the bubbles, μ, and the interfacial