



Numbered-up gas–liquid micro/milli channels reactor with modular flow distributor

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H I G H L I G H T S

- ▶ Numbering-up gas–liquid flow in microreactor suitable for kg/h production capacity.
- ▶ Flow non-uniformity is studied using barrier-channels concept.
- ▶ Taylor flow with uniformity larger than 90% achieved in the parallel channels.
- ▶ Six fluids are studied with different viscosities, surface tensions and flow rates.
- ▶ Three channels type are studied square and circular in steel and square in glass.

A R T I C L E I N F O

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A B S T R A C T

Gas–liquid processing in microreactors remains mostly restricted to the laboratory scale due to the complexity and expenditure needed for an adequate numbering-up with a uniform flow distribution. Here, the numbering-up is presented for multi-phase (gas–liquid) flow in microreactor suitable for a production capacity of kg/h. Based on the barrier channels concept, the barrier-based micro/milli reactor (BMMR) is designed and fabricated to deliver flow non-uniformity of less than 10%. The BMMR consists of eight parallel channels all operated in the Taylor flow regime and with a liquid flow rate up to 150 mL/min. The quality of the flow distribution is reported by studying two aspects. The first aspect is the influence of different viscosities, surface tensions and flow rates. The second aspect is the influence of modularity by testing three different reaction channels type: (1) square channels fabricated in a stainless steel plate, (2) square channels fabricated in a glass plate, and (3) circular channels (capillaries) made of stainless steel. Additionally, the BMMR is compared to that of a single channel regard the slug and bubble lengths and bubble generation frequency. The results pave the ground for bringing multi-phase flow in microreactor one step closer for large scale production via numbering-up.

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1. Introduction

The high rates of mass and heat transfer, minimum axial dispersion and the high interfacial area allow micro/milli channel reactors to run highly exothermic, toxic or even explosive reactions safely, permitting greener routes for processing [1–5]. Microreactors are very attractive devices for many different applications [6–9]. Different from the traditional scale-up, micro/milli channel reactors reach bulk chemicals productions via so called numbering up, placing multiple channels in parallel [10–13]. Because the dimensions of the microchannel where mixing, heating and

reaction remains the same as those of the laboratory scale, industrial production starts directly from the lab [14–16].

The simplest scheme for scale-up via numbering-up is shown in Fig. 1. In the laboratory, scale-up of a single channel is investigated while “smartly” keeping the excellent properties of the micro/milli channels reactor [17,10]. The second scale-up step is to number-up the single channel in one single device – the modular unit. The last step is to arrange all these modular units together in what Hasebe [12] named the plant lay-out.

The main block for the numbering-up is the modular unit. The modular unit can be defined as a device which contains different functional elements such as: distributor, mixer, reaction channels, heat exchanger and separator, and being fed by one single feeding unit for each phase. The modular unit should maintain equal flow conditions in the parallel channels, all of the functional elements

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Nomenclature

Ca_B	capillary number based on the liquid properties and the bubble velocity (-)
d	channel nominal hydraulic diameter (m)
H	channel height (m)
W	channel width (m)
N	number of channels (-)
P	pressure (Pa)
q	flow rate (m ³)
Re_B	Reynolds number based on the liquid properties and the bubble velocity (-)
L	channel length (m)
U	superficial velocity (m/s)
u_B	bubble velocity (m/s)

Greek symbols

γ	surface tension (N m ⁻¹)
μ	viscosity (Pa s)
ρ	density (kg/m ³)

Subscripts

M	manifold channel (-)
B	barrier channels (-)
T	T-mixer (-)
C	gas-liquid flow channel (-)
G	gas (-)
L	liquid (-)

should be integrated in one device, and the fabrication method should be suitable for bulk production of the reactor.

For single phase flow, many modular units are already available in the market for industrial production [18,19,10,17]. For multi-phase flow, development of modular units is still in a preliminary stage [20–22]. This is mainly due to the difficulty in managing the flow distribution for multi-phase flow [23,24,20,25]. Improper flow distribution, specially for gas-liquid flow, can result in a deformation of the flow pattern or in gas-liquid channeling [26,27], some channels filled only with liquid while others are filled with gas.

The flow distribution depends on the hydraulic resistance in each of the parallel channels [28–30,18]. In single phase flow, the hydraulic resistance depends on the physical properties of the fluids and on the hydraulic diameter of the channel. For multi-phase flow, the flow distribution depends on the properties of the single phase [31] and in addition on the flow rates, the specific gas-liquid interfacial area, the flow regime [32], and on the way the phases are in contact. The contact between the phases can be continuous like in the falling film microreactor [33] or dispersed like in segmented Taylor flow [34]. Here we only focus on gas-liquid flow in channels operated under the Taylor flow regime [35,36]. Taylor flow is attractive due to its well-defined gas-liquid interface, reduced axial dispersion almost approaching plug flow, and high mass and heat transfer [35,37].

Distributing gas and liquid flows to achieve Taylor flow regime in parallel channels can be achieved via branching, internal distribution (like in the monolith using a douche type), or by using separate gas and liquid feeding for each parallel channel [38]. When hydraulic resistances, so called barrier channels, are placed between the single phase flow distributor and the separate gas and liquid feeding for the parallel micro channels as shown in Fig. 2, (1) gas-liquid channeling is prevented, (2) all flow regimes, viz. Taylor, churn and annular can be successfully realized, and (3) the flow uniformity is substantially improved [20,25].

The barrier-based distributor is an excellent gas-liquid distributor for parallel channels operated in the Taylor flow regime. A major characteristic for this distributor is the hydraulic resistance needed to achieve equal flow distribution. This parameter can be quantified in a generic way as $\Delta\tilde{P}_B$ as given in Eq. (1). It is the average pressure drop over the barrier channels $\overline{\Delta P_B}$ divided by the average pressure drop over the corresponding mixers and micro channels $\overline{\Delta P_C}$. Since $\Delta\tilde{P}_B$ is a ratio of pressure drops, it is dimensionless.

$$\Delta\tilde{P}_B = \frac{\overline{\Delta P_B}}{\overline{\Delta P_C}} \quad (1)$$

De Mas et al. [20] were among the first to demonstrate this type of distributor in micro channel reactors. Their design was successfully run but with barrier channels designed in the range of $\Delta\tilde{P}_B$ larger than 25 and 50 for liquid and gas, respectively. Al-Rawashdeh et al. [39] demonstrated that $\Delta\tilde{P}_B$ can be designed in the range of 4–25 by following a specific design methodology. The design methodology determines the maximum acceptable fabrication tolerance in the barrier channels, mixers and reaction channels.

According to the specific design methodology, the barrier-based micro/milli reactor (BMMR) shown in Fig. 2 was designed and fabricated. The BMMR consists of eight parallel reaction channels all operated in the Taylor flow regime. It is designed to hold pressure up to 20 bar and temperature up to 200 °C, however these two parameters are not examined in this paper. The BMMR is a modular type of reactor with a maximum liquid throughput of 150 mL/min and gas to liquid flow ratio up to 10. The BMMR demonstrates the numbering-up concept for gas-liquid Taylor flow possible for a production capacity reaching kg/h.

In this paper the quality of the flow distribution in the BMMR is reported by studying two aspects. The first aspect is to experimentally examine six different fluids with different viscosities, surface tensions and flow rates. The second aspect is to study the reactor

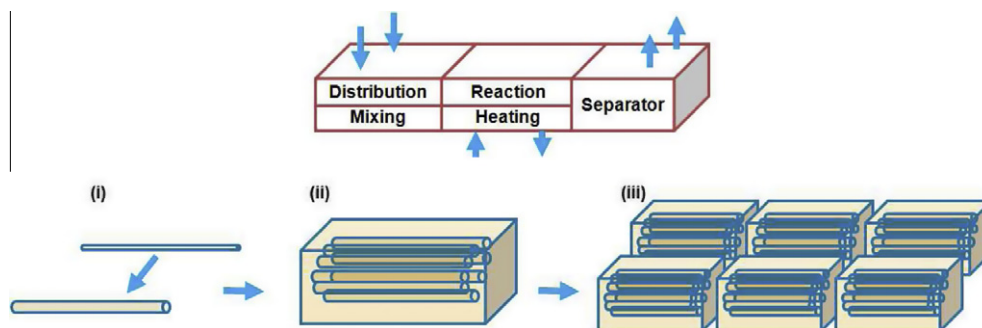


Fig. 1. Scheme for the route of scale-up via numbering-up for micro/milli channel reactors. (i) scale-up of a single channel, (ii) modular unit, and (iii) multi-modular units.

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