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Mixing performance assessment of a multi-channel mini heat exchanger reactor with arborescent distributor and collector

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HIGHLIGHTS

- ▶ A design of mini-scale, multi-channel heat exchanger reactor is proposed.
- ▶ Rapid mixing is observed by using visualization and competitive reactions.
- ▶ Comparable mixing performance is obtained with that of several micro-structured designs.
- ▶ Arborescent structure provides uniform distribution for the numbering-up of channels.

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ABSTRACT

A mini reactor with 16 channels and with channel diameter of 1 mm or 2 mm is designed, fabricated and studied experimentally. Two fluids are divided into 16 channels through distributor, then contacted at T-mixers, and finally collected. Both distributor and collector are designed following the arborescent shape. Visualization with pH indicator bromothymol blue (BTB) is used to qualitatively observe the global mixing; while for the micromixing performance assessment, we used competitive iodide/iodate (Villermaux/Dushman) reactions. Segregation index and micromixing time, with respect to energy dissipation rate are used to characterize micromixing performance.

Visualization with pH indicator shows that complete global mixing at the collector by liquid impingement is obtained under our tested conditions. Iodide/iodate reaction shows a rapid mixing at molecular level as the flowrate increases and impingement being enhanced. Micromixing time estimated by IEM (Interaction by Exchange with the Mean) model is compared with other micro-structured mixers in the literature. Comparable mixing performance is obtained using our millimetric reactor with that of micro-structured designs.

The design of multi-channel reactor with arborescent distributor and collector makes possible the numbering-up of multiple channels. Process intensification is highlighted by high throughput and compact size, thus may be attractive for industrial application.

1. Introduction

Process intensification, particularly the design of high yield processes or compact equipments in the field of process engineering, has become one of the pressing industrial needs in recent years. Paths leading to intensification include the miniaturization of process equipment, system integration of multiple devices or processes, etc. [1–4]. Similarly, *continuous processing* is preferable for designing future processes. It ranked on the first of the Top 10 green engineering research areas in pharmaceutical industry summarized by American Chemical Society Green Chemistry Institute in 2007 [5]. Together with inline analysis, continuous processing provides high safety and quality control in many areas of chemical and process engineering.

One of the routes to reach both process intensification and continuous processing is the use of mini/microscale equipments. Reactors with mini/microchannels enhance heat and mass transfer characteristics mainly due to their high surface/volume ratio. One important issue for industrializing their application is the numbering-up problem. Currently most researches about mini/microchannel reactor end up with single channel study. Several studies have used multi-channels to gain throughput, however only a few researches take into account of the flow maldistribution problem [6–12]. Nonuniform flow among channels is usually a main cause of performance deterioration or malfunction of these

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Nomenclature

C	concentration (mol/L)	V_m	mixing volume in reactor (m^3)
ϵ	energy dissipation rate (W/kg)	X_S	segregation index
ϵ_{353}	extinction coefficient at 353 nm for tri-iodide (L/(mol cm))	Y	selectivity of acid consumption
e	fitting exponent	Z	ionic charge
I	ionic strength (mol/L)		
k	kinetic constant	*Indices*	
l	length of light path (cm)	0	initial
n	molar quantity (mol)	1	reactor with channel diameter 1 mm, M1
OD	optical density, spectrophotometric absorption (a.u.)	2	reactor with channel diameter 2 mm, M2
ΔP	pressure loss (bar)	diss	energy dissipation
Q	volume flowrate (mL/min)	m	mean, mix
t	time (s)	macro	macro-observation
V	internal volume of reactor (m^3)	micro	micromixing
		ST	total segregation

devices. As a result, how to propose novel designs of multi-channel mini/micromixer-reactors with uniform flow distribution between channels receives more and more attention in recent years.

Mixing is one of the most important steps to conduct a high yield reaction. Especially for some fast reactions, rapid mixing ($t_{mixing} < t_{reaction}$) is crucial to obtain high chemical selectivity. Mixing process can be categorized into macro-, meso- and micro-mixing according to their mechanisms and dimensional scale [13,14]. Macromixing by fluid turbulence is the first step, which happens usually in the scale of whole equipment; the second step, mesoscale mixing, is driven by fluid viscosity or shear stress between fluids or fluid-wall surfaces; and the final step is micro-scale mixing driven by diffusion of molecules. The whole mixing process happens with three mixing in a cascade manner (macro to meso to micro). Particularly the micro-scale mixing is essential for chemical processes because it relates directly with molecular exchange.

Different methods have been developed by researchers to assess mixing performance. In general mixing characterization methods could be sorted by either direct flow visualization or chemical reaction method. Particles with PIV (Particle Image Velocimetry) system, fluorescence with microscopy or LIF (Laser Induced Fluorescence) method, etc., have been used as tools to observe flow field in transparent prototypes [13,15–18]. Chemical reaction methods mainly involve several competitive reactions whose selectivity is sensitive to mixing. Typical chemical schemes are iodide/iodate reaction [19,20], diazo coupling reaction [21–23], Walker scheme [24–26], etc. The last two reaction schemes are related with thermal treatment because reactions are either exothermic or endothermic. Among the methods above, only chemical reactions can reflect mixing down to microlevel. A detailed review of mixing assessment methods was given by Aubin et al. [27] recently.

Among various micromixing characterization methods, iodide/iodate reaction scheme [28] (or Villermaux/Dushman reaction) is well adapted and widely used. Kinetic data of reactions are well-known so that it is easy to analyze the results quantitatively. A simple model to characterize micromixing process is IEM (Interaction by Exchange with the Mean) model, developed by Villermaux and Falk [29]. Using this model micromixing time can be related with experimental results from iodide/iodate reaction. Recent literature reviews on micromixing performance assessment are presented by Falk and Commenge [30,31] by employing this model on different experimental results.

In this paper, we propose an integrated design of a mini-scale, multi-channel mixer-reactor. Arborescent style distributor is used to achieve uniform fluid distribution among channels. Two fluids are contacted at 16 T-junctions and complete the mixing within channels and in arborescent collector. Heat exchanger, mixer and reactor are integrated together in this continuous processing equipment.

Firstly pH sensitive dye BTB (bromothymol blue) is used as color indicator to visualize the mixing and flow behaviors inside our reactor. Then competitive iodide/iodate reaction with IEM model are implemented to quantitatively explore the micromixing performance of the proposed reactor. Finally a performance comparison between our designs and other micro-structured mixer/reactors is made, in terms of micromixing time and energy dissipation.

2. Heat exchanger reactor design and geometry

The application of multi-scale, nature-inspired structures for fluid distribution and collection has been studied extensively [32–38] during the last decade. Scaling relations were established analytically by Luo and Tondeur [32,33] for the design of multi-scale distributor/collectors, considering energy dissipation and fluid distribution uniformity. Multi-scale components with arborescent internal structures, i.e. distributor/collector, heat exchanger, mixer and their integrations have been designed and investigated [35,36,38].

Based on the experiences above, we developed a multi-functional heat exchanger reactor, integrating heat exchanger, mixer and reactor. Shown in Fig. 1a is the whole geometry, including arborescent structures used for connecting single inlet/outlet and multiple parallel channels. In general we have two inlets of different fluids to be mixed and one outlet for product. Two fluids of each inlet are injected and then distributed into 16 channels. The two fluids then contact through T-junctions where mixing happens with counter-current impingement. After, the mixture goes along the channels to gain complete mixing. The final mixture will then be collected to the outlet. Symmetry design of this nature-inspired distributing/collecting structure makes the flow path of fluid in every channel being identical. Non-uniformity of flow distribution between channels is minimized.

For clarity we define number 0–4 to represent different channel scales in arborescent structure, i.e., 0 representing the lowest scale (each of 16 branches) and 4 representing the highest (main outlet for the case of collector, as indicated later in Fig. 6a).

Heat exchange chamber is located outside the vertical parallel channels. Inlet and outlet are arranged in such a way that utility fluid flows reversely with inside fluid. Counter-current flow is essential for an efficient heat transfer for this shell-and-tube heat