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Hydrodynamics of gas–liquid micro-fixed beds – Measurement approaches and technical challenges

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HIGHLIGHTS

- Point electrodes were used in micro-fixed bed fabrication.
- Tracer input point and packing density contribute to the reliability of RTD tests.
- Liquid holdup increases with increasing gas density at iso-G conditions.
- Near-wall RTD determination by microscopic imaging was done as an original work.
- Near-wall RTDs confirm the theoretically predicted maximum velocity in wall region.

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ABSTRACT

Despite many areas that are open to investigation in the hydrodynamic study of micro-fixed bed reactors, conducting research in this field is mostly hampered by a number of experimental challenges that have made many attempts ineffectual. This work provides a summary of the technical challenges, problems and misconstrues one might encounter in performing hydrodynamic experiments on micro-fixed bed reactors. Some of these issues will be pointed out upon comparing classical residence time distribution (RTD) measurements through electrical conductivity probes at micro-fixed bed scale with near-wall RTD obtained via visualizations of dye-tracer elution and monitoring the changes in gray level intensity of the images. Laterally-averaged gray level intensity at both upstream and downstream extremities of the wall regions of interest acted as inlet and outlet curves for the Aris method. The major outcome of this work was experimental confirmation of theoretically predicted maximum liquid velocity in the high porosity zone close to the wall. Finally, the experimental results on pressure drop and liquid holdup obtained upon following the right experimental protocols are presented.

1. Introduction

The emergence of microreactors was the outcome of a world-wide demand for smaller, cleaner, safer and more energy efficient process units. High surface-to-volume ratios promote heat and mass transfer in these miniaturized units making them suitable for highly exothermic [1–3] and mass transfer limited reactions [4]. Due to small testing volumes and short residence times, they can also be employed safely for fast catalyst screening purposes to transfer laboratory results into production more rapidly [5]. The *numbering up* possibility for micro-structured devices is another feature which, although still in its infancy, has the potential to be applied in industry [6,7]. Today, on the other hand, chemical industries highly rely on multiphase catalytic reactions in which gas and liquid phases flow concurrently through a fixed bed of solids. This flow pattern provides efficient contact between the phases and thus has a wide variety of applications [8]. Having combined the pool of benefits for both microreactors and fixed bed reactors, micro-fixed beds (also known as micro-packed beds) are promising candidates to take the industry to the next level. This, in effect, led to a number of researches mainly focused on reaction engineering [9–11] and mass transfer [4,12] in microreactors each confirming their enhanced performance as compared to macro-scale units.

Even in micro-scale reactors, despite the advantages cited, there exist offsets from what is ideally expected in terms of conversion, mass transfer and so forth. These are attributed to deviations from the ideal fluid flow patterns and contacting schemes the extent of which could be addressed by hydrodynamic studies. Results of such analyses might bring modifications to the existing designs for boosted performance and efficiency. Micro-fixed bed reactors are no exception in this regard and thus, hydrodynamic

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experiments such as those performed on their macro-scale counterpart (trickle bed reactors) are required to measure (or determine) the hydrodynamic parameters including: pressure drop (ΔP), liquid holdup (ε_L), residence time distribution (RTD). Also, microscopic visualizations can be utilized for flow regime studies and provide valuable information on the different contacting patterns between the phases. As for the hydrodynamic studies in this field, the attempt to perform RTD measurements for gas–liquid flow in micro-packed beds has been done by van Herk et al. [13] and later by Márquez et al. who performed RTD tests to determine liquid holdup in micro-packed beds using non-volatile [14] and volatile [15] tracers. Transient behavior of micro-packed beds in several startup procedures was also investigated by Márquez et al. [16] and minor deviations in the achieved steady state were observed. In spite of the valuable insights that could be obtained by microscopic visualizations, studies of this kind such as flow regime investigations are quite scarce [17].

It can be perceived from above that there is a huge research potential in this field and yet a dearth of experimental works and publications. This mainly stems from several challenges and problems that arise upon hydrodynamic study of micro-fixed bed reactors during experimentation phase precluding accurate and reliable experiments as compared to hydrodynamic tests on other types of microreactors such as micro-channels. There are a number of significant details in hydrodynamic studies of micro-fixed beds that have been either overlooked and caused successive failures or kept as the know-hows to the previous experimenters. Therefore, the authors' objective in this work is to reveal the bag of tricks gained through experience and following a cause and effect learning logic in this subject so that the prospective experiments on micro-fixed bed reactors could be performed as immaculately as possible. In what follows, the experimental setups and methodologies for both precise RTD studies and visualizations are described first along with useful and detailed guidelines for implementation. Afterwards, the results will discuss the major challenges during experimentation by illustrations drawn from problematic and/or modified experiments, wherever necessary. Finally, a number of experimental results on pressure drop and liquid holdup obtained by committing to the correct experimental procedures are presented.

2. Experimental section

2.1. Microreactor packing procedure

Micro-reactor packing is one of the crucial steps which, if done correctly, ensures reproducibility and reliability of the experimental results. Particles could be easily loaded inside the micro-tube through a handmade tiny metal funnel within multiple steps in each of which a certain portion of the total bed volume is loaded with particles. Frequent tappings are required between the steps to completely densify the bed. Loose packings will adversely affect reproducibility of the experiments (especially in case of pressure drop measurements) and once the gas and liquid are both fed, gaps may start to develop at different locations within the length of packed bed or flow instabilities upstream of the bed might occur and halting the experiment would be inevitable.

2.2. RTD setup fabrication and methodology

High-pressure gradients within the length of micro-fixed bed reactors (in the order of 10^3 kPa/m) necessitate materials for fabrication that could withstand elevated pressures without mechanical failure and subsequent leakages. If microscopic visualizations are not intended (such as in RTD measurements), walls of micro-fixed beds could be made of thick glass or even metals whereas in case of visualizations (e.g., for flow regime studies [17]) the walls should only be made of transparent pristine materials that might be mechanically strengthened by methods such as etching [18]. Installation of electric conductivity probes in the walls of those micro-fixed beds that are utilized for RTD studies was first inspired by the conductivity ring electrodes commonly used for macro-scale packed beds [19]. Fig. 1a shows a schematic of such micro-ring electrodes constructed through inserting perforated metal plates within lateral cuts across the reactor wall and fixed by applying a layer of epoxy glue around that. However, this appeared to be one of the major challenges for the authors as high pressure gradients increase the risk of fluids leakage which led to a series of failures in RTD experiments. As a consequence, the idea was replaced by inserting two point electrodes (instead of rings) via two holes drilled in micro-reactor wall without causing pertur-

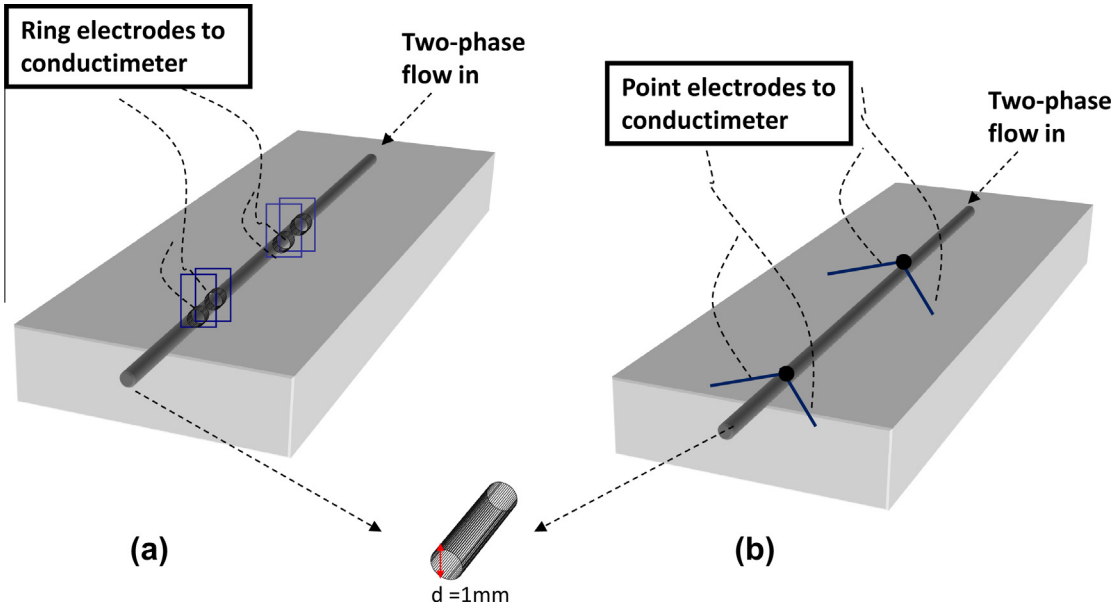

Fig. 1. Two types of electrode configuration proposed for for RTD measurements. (a) Old design of ring electrodes, and (b) new design of point electrodes installed on microreactor.