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Characterisation of Pulsing Flow in Trickle-Bed Reactors using Ultra-Fast Magnetic Resonance Imaging

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Abstract. We use ultra-fast Magnetic Resonance Imaging (MRI) to characterise hydrodynamics during pulsing flow in trickle-bed reactors. One-dimensional (1-D) liquid holdup profiles along the flow direction were acquired using Fast Low Angle SHot (FLASH) MRI at spatial and temporal resolutions of 352 μm pixel^{-1} and 3.3 ms, respectively. Liquid pulse properties such as pulse velocity, duration and frequency obtained with MRI are in good agreement with those measured using a well established technique called the conductance method. In addition, MRI gives local measurements and provides additional spatial information to track individual liquid pulses, which cannot be obtained with the conductance method approach.

Keywords

trickle-bed reactor, pulsing, magnetic resonance imaging, hydrodynamics, multiphase flow

1. Introduction

Trickle bed reactors (TBRs) are packed columns with concurrent down-flow of liquid and gas, widely used in chemical reactor design, especially for hydrogenation and oxidation reactions. At high liquid and/or gas flow rates, TBRs are operated in the pulsing flow regime, where the gas/liquid distributions vary and liquid rich pulses move down the bed. The conductance method has been used extensively to characterise these liquid pulses [1] with two or more pairs of conductance rings placed at different heights down the bed. Each pair measures the electrical impedance of the liquid, hence the liquid holdup, in the bed cross section between the ring electrodes at a sampling rate of up to thousands of Hz. This technique is *in situ* and very robust for studying transient behaviour in TBRs. However, an intrinsic drawback is that all measurements made are averaged between pairs of conductance rings and no spatial information is given.

In this paper, we present an alternative technique, ultra fast FLASH MRI [2], to obtain 1-D liquid holdup profiles along the flow direction. Pulse parameters such as velocity, duration and frequency are determined by applying appropriate algorithms to the acquired MRI signals. The results are then compared with those obtained with the conductance method for the same packing and operating conditions. Unlike the conductance method, MRI is capabable of tracking individual liquid pulses, allowing us to study in more detail the dynamics of liquid pulses as they travel down the bed.

2. Experimental

The TBR used was a cylindrical PTFE column of 0.043 m I.D. and 0.7 m in height, packed with 3 mm γ-Al_2O_3 cylindrical pellets. The bed porosity was ~0.4. Air and water were fed from the top of the TBR. The TBR was operated in the pulsing regime ($L = 0.011$ ms^{-1}, $G = 0.36$ ms^{-1} at 3.3 bar) for 1 h to achieve consolidation. Desired gas and liquid flow rates were then set using the gas rotameter and liquid mass flow controller and data were acquired after a minimum time interval of 15 min. Two sets of experimental conditions were used: superficial liquid flow rate $L = 0.011$ ms^{-1} and superficial gas flow rate $G = 0.06$–0.36 ms^{-1}; $G = 0.20$ ms^{-1} and $L = 0.006$–0.015 ms^{-1}.

Two pairs of conductance rings were mounted flush inside the TBR at a separation of 0.2 m. Each pair consisted of two conductance rings at a separation of 0.03 m, generating one conductance signal, which was sampled at 200 Hz. The pulse parameters were determined as follows: (i) the pulse velocity was calculated by cross correlating the two traces, which gives a time-averaged measurement; (ii) the pulse frequency was determined by auto correlating the two traces; (iii) the pulse duration was calculated from the time interval at half height of each liquid pulse.

All MRI data were acquired using a vertical SWB DMX 200 Bruker magnet (4.7 T, ^1H-imaging, 199.7 MHz) using 1-D FLASH MRI in the flow direction. The field-of-view was ~3 cm (352 μm $pixel^{-1}$). A 256 μs Gaussian pulse (10° tip angle), an echo time of TE = 1.5 ms and a repetition time TR = 3.3 ms (sampling rate of 307 Hz) were used. Up to 4096 liquid profiles were recorded consecutively in a single experiment. At least five separate experiments were conducted at each combination of liquid and gas flow rates.

The average stationary liquid content was subtracted from each MRI image to show only the local variation of liquid holdup, from which individual liquid pulses are clearly observed. The pulse velocity was calculated by using a modified cross correlation algorithm, which allows 85 traces to be correlated simultaneously. The pulse frequency was determined by auto correlating the liquid traces. The pulse duration was calculated by identifying individual liquid pulses and measuring the time intervals at half height.

3. Results and Discussion

Figure 1 shows the liquid pulse velocity measured with both MRI and conductance methods as a function of the gas and liquid flow rates. Excellent agreement is obtained between the two methods at all gas flow rates and medium to high liquid flow rates. The results exhibit a similar trend to those shown in the literature. The pulse velocity is highly dependent on the superficial gas velocity and is almost insensitive to the superficial liquid velocity L if $L > 0.01$ ms^{-1}, in agreement with earlier work on packings of glass spheres and Raschig rings [3]. A discrepancy occurs at $L < 0.01$ ms^{-1}. This is because measurements obtained with the conductance method are likely to under-estimate the actual pulse velocity as a result of conductance values being averaged over 20 cm of the column – liquid pulses might still be developing as they flow down through this section of the column. Similar agreement between MRI and conductance measurements of pulse frequency and duration is also obtained.

Pioneering researchers have reported by visual observation near the column wall that liquid pulses, as they travelled down the column, continuously acquired more fresh liquid at the front as well as leaving some of the liquid behind [4]. Figure 2 shows binary images which capture individual liquid pulses over a 3 cm length of the column over a period of 1.6 s. For the first time, direct experimental evidence of the dynamics of liquid pulses inside the column is provided: Fig. 2(a) shows the splitting of a liquid pulse into two and Fig. 2(b) shows how two liquid pulses merge into one. We note that the splitting and merging of liquid pulses normally occur at similar positions inside the TBR. As indicated by the yellow circle in Fig. 2(a), there are some liquid pulses which are seen to 'disappear' at a certain time. This occurs primarily when pulses are associated with low signal; when this signal intensity decreases below the gating level they will disappear from the image. This decrease in signal could be due to several factors including an increasing dispersion of the pulse, an increasing pulse velocity or simple image noise. In several cases, including the region highlighted, the pulse is seen to reappear further down the bed as the intensity rises above the gating level again.

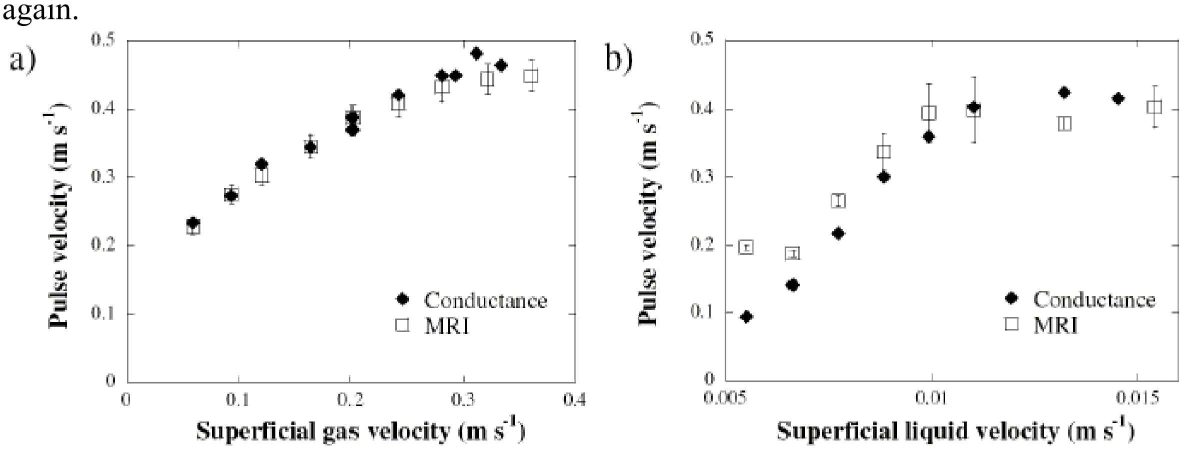

Figure 1: Pulse velocity calculated from both conductance and MRI techniques. (a) As a function of the gas flow rate when $L = 0.011$ ms^{-1}. (b) As a function of the liquid flow rate when $G = 0.20$ ms^{-1} at 3.3 bar. The error bars are the standard deviation of the results obtained in different experiments.

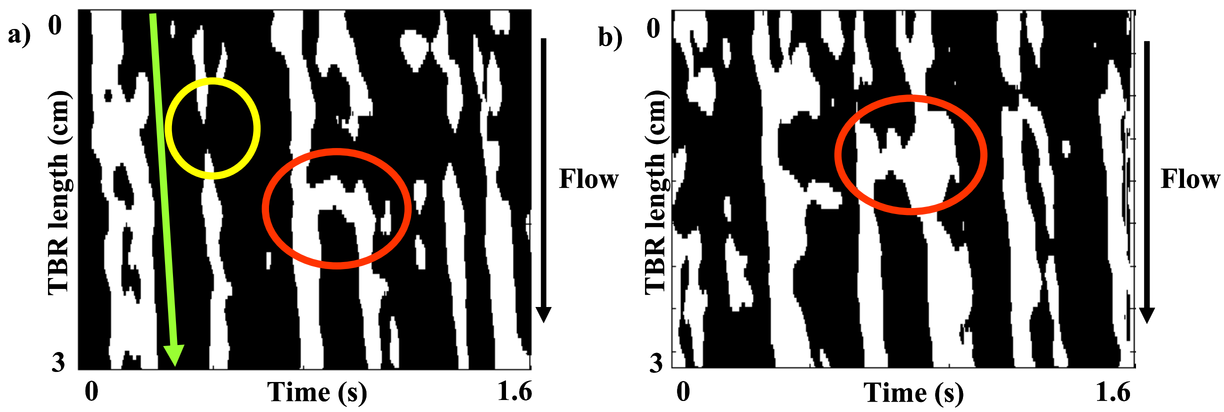

Figure 2: MR images showing the splitting and merging of liquid pulses as they travel down the bed at $L = 0.011$ ms^{-1}, $G = 0.36$ ms^{-1} at 3.3 bar. The liquid pulses appear white and gas pulses black. Indicated in the red circles: (a) splitting of one liquid pulse (b) merging of part of a liquid pulse with another. The gradient of the green line corresponds to the average fluid (gas + liquid) velocity.

4. Conclusions

We have demonstrated the use of MRI as a complementary technique to the conductance method for characterising pulsing flow in TBRs. The pulse velocity, frequency and duration measured with MRI are in good agreement with the results obtained with conductance measurements. Individual pulses are spatially resolved with MRI, thereby enabling pulse splitting and merging to be imaged for the first time.

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