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Flow distribution characteristics of a gas–liquid monolith reactor

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Abstract

Flow distribution characteristics in a 0.05 m diameter monolith reactor, operated co-current downward in the Taylor flow regime, was investigated and quantified using gamma ray computed tomography (CT). The results indicate that the flow distribution is a strong function of liquid distributor design, and gas and liquid superficial velocities. Additionally, within the range of gas and liquid superficial velocities investigated, a window of operating condition (gas and liquid velocities) was identified which resulted in a close to uniform phase distribution.

Keywords: Monolith; Flow distribution; Liquid saturation; Computed tomography

1. Introduction

Fixed bed catalytic reactors are widely used in many industrial processes. Recently, structured packing elements (e.g., monoliths, Katapak, Mellapak etc.) have been suggested for various chemical processes [6,7,11]. Due to the complex nature of the interaction between gas, liquid and solid phases, one of the major challenges in the design and operation of these reactors is the prevention of liquid flow maldistribution, which could cause portions of the bed to be incompletely wetted. As a result, several problems could be encountered including hot spot formation, reactor runaway for exothermic reactions, decreased selectivity to desired products, and underutilization of the catalyst bed.

To overcome these problems, flow distribution in these reactors over a desired packing structure and at a desirable range of operating conditions need to be quantitatively studied and understood. Among the non-invasive techniques used to study the flow distribution in multiphase reactors, the advanced non-invasive computed tomographic scanner (CT) with gamma ray source at the Chemical Reaction Engineering Laboratory (CREL)-Washington University [13] has been effectively used to characterize flow distribution of in both non-structured and structured packing [2,1,12].

Other examples of the use of tomographic techniques to determine gas/liquid distribution in monolith packing is limited in the literature. Mewes et al. [9] studied flow distribution in monoliths using capacitance tomography technique. A spatial resolution of about 5–10% of the diameter of the measurement plane was in general possible using capacitance tomography. Water was used as the liquid phase and was distributed over a 0.12 m diameter monolith using large number of injection nozzles. With increasing liquid velocity, the liquid distribution improved. More recently, Gladden et al. [4] investigated the flow through monoliths at limited flow conditions. The authors reported a distribution of gas and liquid slug velocities as well as flow maldistribution in the monolith cross-section. Heibel et al. [5] have studied flow distribution in a monolith operated in the film flow regime using magnetic resonance imaging (MRI) technique. Three different nozzle distributor designs were used to cover a narrow range of liquid velocities. For an optimum combination of nozzle distributor and liquid velocity, the authors demonstrated uniform liquid distribution in the monolith cross-section.

The work presented in this article investigates the flow distribution in a monolith packed bed using gamma ray computed tomography. The monolith reactor was operated co-current downflow in the Taylor flow regime. The work evaluates the effect of gas and liquid superficial velocities on the degree of phase distribution on a monolith cross-section. This information is important for proper modeling and

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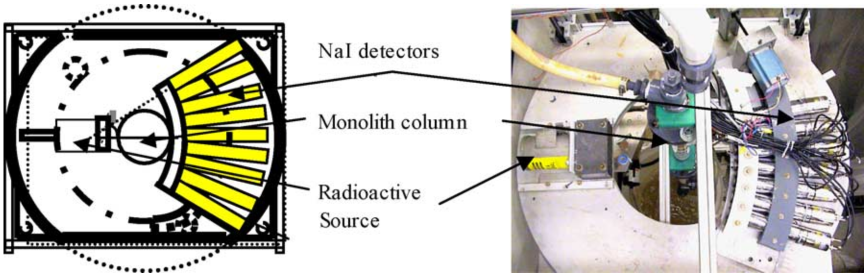


Fig. 1. The top view of the gamma ray computed tomograph set-up.

scaleup of a catalytic monolith reactor for solid catalyzed gas–liquid reactions.

2. Gamma ray computed tomography (CT) scanner

Newly installed computed tomography (CT) used in this study was a modified unit of the third generation fan-beam configuration used by Kumar et al. [8] at the Chemical Reaction Engineering Laboratory (CREL). The details of the tomography unit have been published elsewhere [12]. In summary, the CT consists of an array of NaI (TI) detectors (5 cm in diameter) and an encapsulated ~100 mCi Cs^{137} source located opposite to the center of the array of detectors, Fig. 1. The entire source–detector assembly was rotated around the column using two program automated stepping motors to obtain numerous projection data (~12500). The projection data was then used to reconstruct the cross-sectional attenuation density profile using estimation–maximization algorithm [13]. The entire scanning process takes about 4–5 h. Separately, few reference scans were also performed to obtain the attenuation coefficients of column with water, column with dry and wet monolith and column packed with monolith and completely filled with water. The final holdup distribution images were obtained from the attenuation data using the relationships and the procedure developed by Chen et al. [2] for packed bed. The liquid saturation profiles thus obtained represents the liquid present only in the monolith channels.

Since the newly constructed CT unit was used for the first time to study flow distribution in monoliths, several validation scans were performed to evaluate its accuracy for such scans. One such validation is mentioned here.

2.1. CT validation scan

In this validation scan, a concentric cylinder assembly made of plexiglass was used as a phantom object, as shown in Fig. 2. The phantom object was scanned under two different circumstances. In the first instance, the annular

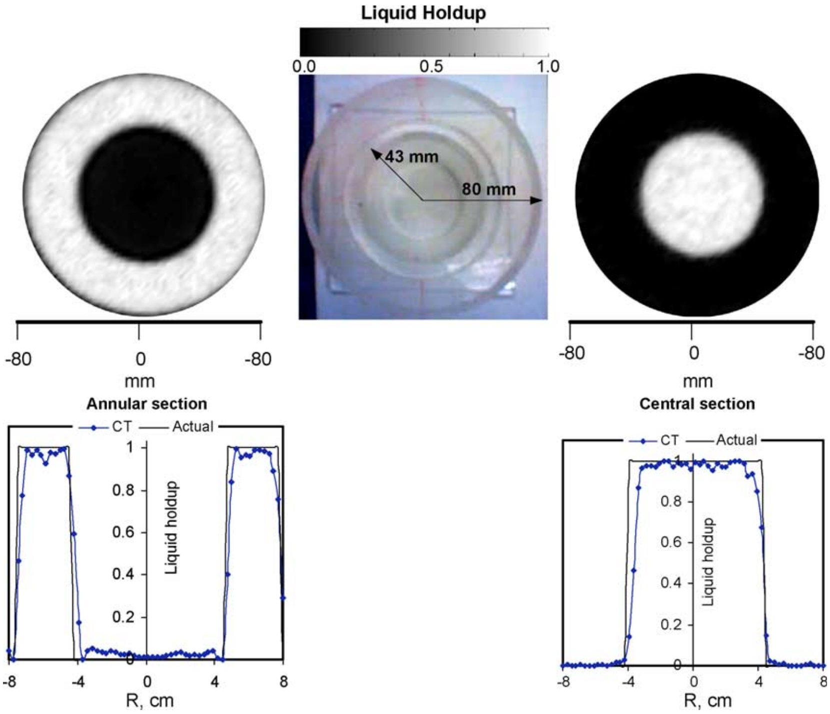


Fig. 2. Examples of tomography scan of a phantom object and comparison of liquid holdup.

section of the object was filled with water and scanned. The resultant scan showed the liquid holdup value close to the actual liquid holdup ($\sim$1.0) in the annular region. The remaining portion had close to zero liquid holdup, which reflects the actual situation. The radial profile corresponding to this cross-sectional profile when compared with the actual liquid holdup showed good agreement.

In the second instances, the central section was filled with water and scanned. Once again, it was seen that the liquid holdup obtained from the CT scan is close to the actual liquid holdup value in the central section. Average difference between the values obtained from tomography and true liquid holdup was found to be $\sim$3.5%.

3. Experimental set-up and operating condition

The experimental set-up, schematically shown in Fig. 3, measured gas–liquid flow distribution in a co-current downflow monolith reactor. It consists of a packing section, distributor, and liquid and gas delivery system. The plexiglas column, 0.5 m high and 0.05 m in diameter, holds one monolith element of height 0.15 cm and diameter 0.048 m. The packing element was held in position by a porous padding. The distance between the monolith's top surface and the liquid distributor could be varied. The liquid was recirculated and introduced at the top through a distributor.

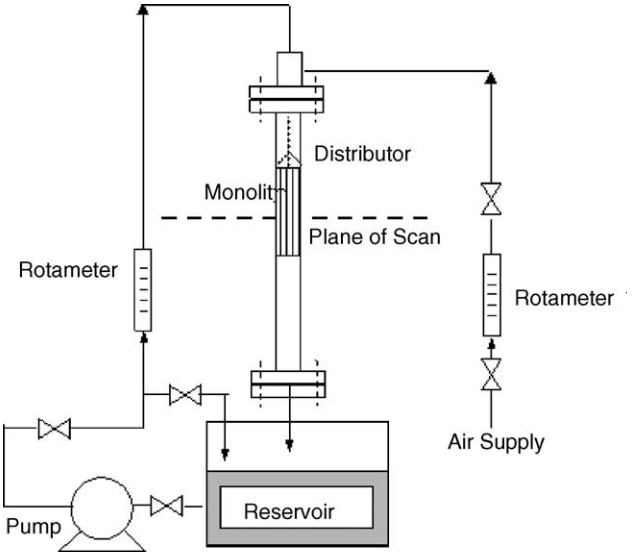


Fig. 3. Experimental set-up to determine gas saturation and phase distribution in a monolith.

The effect of distributor design on the flow distribution was investigated using three different types of distributor, listed in Table 1.

The height of the distributor from the top of the monolith packing is an important factor in achieving uniform phase distribution [5]. In the case of the nozzle distributor, an attempt was made to adjust the nozzle height in such a way that the cone of the nozzle matches with the top of the monolith, as shown in Fig. 4. For the range of superficial liquid velocities used, this height varied from 2.5 to 4 cm.

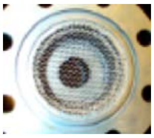
For the showerhead and foam distributors, the distributor height was maintained at 4 cm from the monolith top surface. This was considered adequate as, unlike nozzle distributor, liquid jets do not spread after leaving the distributors.

Water and air were used as the liquid and gas phase, respectively. Flow scans were performed at the middle of the monolith section. The superficial liquid velocity ranged from 0.025 to 0.5 m/s, and the superficial gas velocity ranged from 0.1 to 1.0 m/s. This range of velocities represent Taylor flow regime as depicted by the flow regime map developed by Mishima et al. [10]. Moreover, the flow rates also reflect the industrial flow conditions suitable for industrial applications [3].

4. Results and discussion

Fig. 5a shows a typical time averaged liquid saturation (β_L) distribution in the monolith cross section. By azimuthally integrating the saturation profiles, we get time

Table 1
Distributor characteristics

Distributor type	Characteristics	Image
Nozzle	6 mm orifice diameter	
Showerhead	16 mesh size screen (1.4 mm opening)	
Foam	20 pores per inch and a void fraction of 0.4 made by Porvair	

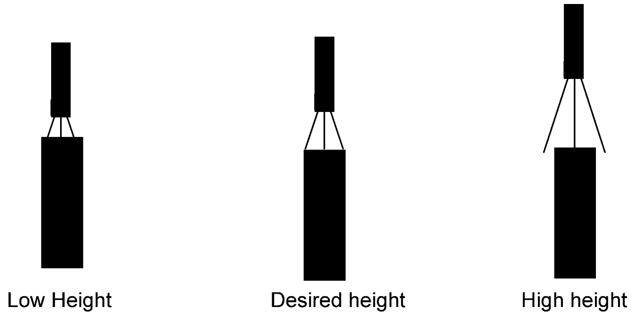


Fig. 4. Schematics of liquid distribution using nozzle at various heights.

and azimuthally averaged liquid saturation as shown in Fig. 5b. The radial profile shown in Fig. 5b indicates two distinct regions. The core section represents the actual phase distribution in the monolith, which in this case (u_l: 0.2 m/s, u_g: 0.1 m/s, nozzle distributor) has a fair degree of uniformity. The clearance between monolith and the reactor wall is packed with Teflon material, and the liquid saturation sharply decreases in this region. In general, computed tomography produces edge artifact during image reconstruction in places where there is a sharp gradient of liquid saturation values, such as wall region. Therefore, in all our calculations, the data from the wall region was discarded and only the monolith section was studied. The characterization of flow distribution in monoliths at various operating conditions was done in a similar manner using the liquid saturation profiles.

Fig. 6 shows the time averaged cross-sectional liquid saturation profiles at four different liquid velocities and at a gas velocity of 50 cm/s. The corresponding radial profiles have also been shown in the figure.

A sense of this distribution can be derived from the radially averaged liquid saturation profiles. A flat radial profile indicates uniform phase distribution, whereas the

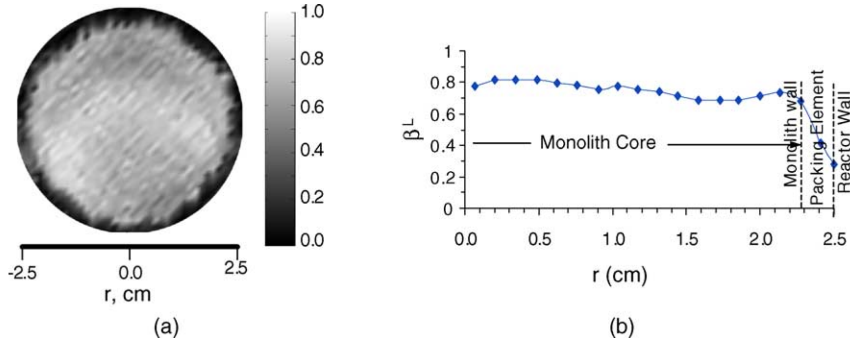


Fig. 5. Liquid saturation β_L for a monolith bed of a 400 cpsi cell density and 75% void fraction. u_l = 0.2 m/s, u_g = 0.1 m/s, nozzle used as the liquid distributor. (a) Time averaged cross-sectional liquid saturation distribution; (b) azimuthally averaged radial liquid saturation profile.

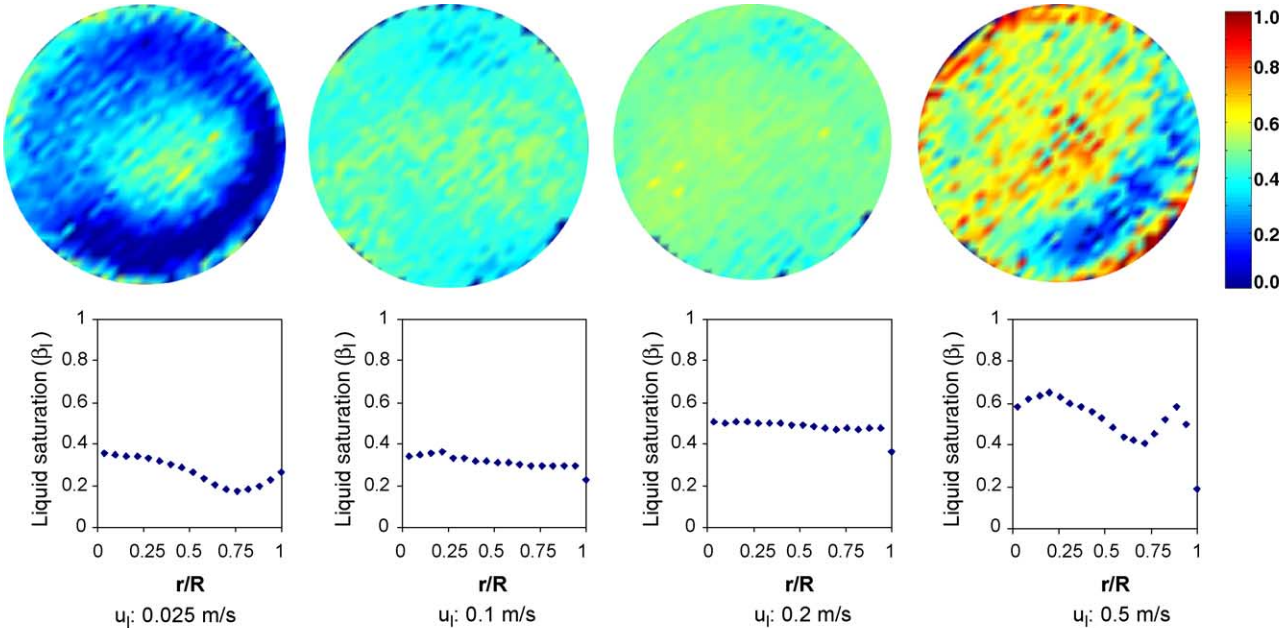


Fig. 6. Time averaged cross-sectional and radial liquid saturation profiles at four different liquid superficial velocities. u_g: 0.5 cm/s, 400 cpsi, 0.75 void fraction monolith, nozzle distributor.

presence of gradients in the radial profile indicates phase maldistribution. Fig. 6 illustrates an interesting phenomenon regarding the flow distribution in monoliths. Even though care was taken to match the cone of liquid flow from the nozzle with the top of the monolith, at low liquid velocity (~0.025 m/s) the spread is not ideal, and most of the liquid was concentrated in the central section. However, some liquid tend to splash and strike the column wall, thus increasing the liquid saturation in some peripheral regions. At intermediate liquid velocities (0.1–0.2 m/s), the spread of liquid flow is nearly constant, and close to uniform flow distribution can be achieved. This is reflected in the nearly flat radially averaged liquid saturation curves. At very high liquid velocity (~0.5 m/s), even though the nozzle performs quite well, as observed visually while performing the experiment, a large amount of turbulence and vortex formation was noticed. This lead to an uneven pressure drop across the inlet of the monolith, resulting in some degree of flow maldistribution.

The degree of non-uniformity was quantified using a newly developed statistical method and is explained elsewhere [12]. In summary, a 100% uniformity factor indicates completely uniform flow, whereas 0% uniformity factor means completely non-uniform flow. It was observed that foam and showerhead liquid distributors produced smaller uniformity factors (<60%, indicating a higher degree of maldistribution, or poor distribution) for all flow conditions studied. Using nozzle distributor, some interesting observations were made. At low liquid velocities (~0.025 m/s) and at high liquid velocities (~0.5 m/s), the uniformity factor was 65% or less. However, at intermediate liquid velocities, the uniformity factor was high, higher than 80%.

Gas velocity had somewhat similar effect on phase uniformity, although not as strong as the liquid velocities. Gas was introduced into the reactor simply by maintaining a constant gas pressure at the inlet of the reactor. Therefore the distribution of gas phase was mostly influenced by the motion of the liquid phase. However, at a very high gas velocity (u_g ~1 m/s), the gas tend to disturb the liquid flow, thereby inducing maldistribution.

5. Summary

A study was performed to investigate the flow distribution characteristics in a monolith bed using computed tomography. Three different distributors were used to evaluate their distribution characteristics. The design of the liquid distributor was found to have a strong bearing on the flow distribution characteristics. Within the gas and liquid flow ranges used, a window of operating conditions was identified in which the flow distribution was close to uniform. Nozzle distributor was found to be most suitable for monolith reactors.

Acknowledgements

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