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Characterising gas behaviour during gas–liquid co-current up-flow in packed beds using magnetic resonance imaging

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HIGHLIGHTS

- Liquid and gas hold-up are measured during co-current gas–liquid up-flow in packed beds.
- Dynamic and static contributions to gas hold-up are identified.
- Bubble size and rise velocities determined as a function of gas and liquid flow rate and packing size.

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ABSTRACT

Magnetic resonance (MR) imaging techniques have been used to study gas phase dynamics during co-current up-flow in a column of inner diameter 43 mm, packed with spherical non-porous elements of diameters of 1.8, 3 and 5 mm. MR measurements of gas hold-up, bubble-size distribution, and bubble-rise velocities were made as a function of flow rate and packing size. Gas and liquid flow rates were studied in the range of 20–250 $cm^3\ s^{-1}$ and 0–200 $cm^3\ min^{-1}$, respectively. The gas hold-up within the beds was found to increase with gas flow rate, while decreasing with increasing packing size and to a lesser extent with increasing liquid flow rate. The gas hold-up can be separated into a dynamic gas hold-up, only weakly dependent on packing size and associated with bubbles rising up the bed, and a 'static' hold-up which refers to locations within the bed associated with temporally-invariant gas hold-up, over the measurement times of 512 s, associated either with gas trapped within the void structure of the bed or with gas channels within the bed. This 'static' gas hold-up is strongly dependent on packing size, showing an increase with decreasing packing size. The dynamic gas hold-up is comprised of small bubbles – of order of the packing size – which have rise velocities of 10–40 mm s^{-1} and which move between the packing elements within the bed, along with much larger bubbles, or agglomerates of bubbles, which move with higher rise velocities (100–300 mm s^{-1}). These 'larger' bubbles, which may exist as streams of smaller bubbles or 'amoeboid' bubbles, behave as a single large bubble in terms of the observed high rise velocity. Elongation of the bubbles in the direction of flow was observed for all packings.

1. Introduction

Fixed-bed three-phase reactors in which gas and liquid phases are contacted with a solid packing which may have catalytic properties are commonly used in chemical and bio-chemical processes; however, their hydrodynamics are still not fully understood and their modelling still raises a number of issues (Attou and Ferschneider, 1999; Bordas et al., 2006). Within the broad subject of gas–liquid flow in fixed or packed beds, studies of co-current down-flow and counter-current flow in packed beds are more widely reported than consideration of the co-current up-flow mode of operation. Raghavendra Rao et al. (2010) have noted this in particular with reference to measurements of gas–liquid interfacial area. Co-current up-flow through fixed beds is most commonly used industrially for gas absorption accompanied by chemical reaction (Hofmann, 1983). As discussed by Hofmann (1983), up-flow operation with continuous liquid and dispersed gas phase is preferred when the liquid has to be treated with a small amount of gas or when a large liquid residence time is

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required. For example, if the overall reaction rate is low, at low liquid flow rates the contacting efficiency of the solid, as well as the gas–liquid mass transfer and hence the apparent reaction rate will be higher in up-flow than in down-flow operation. Examples of applications of the co-current up-flow mode of operation include amination of alcohols, selective hydrogenation of acetylenes and oxidative treatment of waste liquids. Recent interest in co-current up-flow has been reported with respect to studies of flow through structured packings (Kishore Kumar et al., 2012) and hydrodynamics of inclined gas–liquid concurrent up-flow in packed beds (Bouteldja et al., 2013). The present work focuses on the up-flow mode of operation and, in particular, on the behaviour of the gas within the bed. Magnetic resonance (MR) techniques are used to identify regions of the bed which remain gas-filled at all observation times, while discrete bubbles are also identified and their rise velocities determined.

Previous work on packed beds operated in co-current up-flow mode (Khan et al., 1997; Murugesan and Sivakumar, 2002; Moreira and Freire, 2003; Iliuta and Thyrion, 1997) has shown that three main hydrodynamic regimes exist: bubbling, pulsing and spray flow. The transitions between these regimes are controlled by both the liquid and gas flow rates. The bubbling regime is characterised by bubbles of gas passing through a continuous liquid phase, while the pulsing regime is characterised by alternating liquid-rich and gas-rich 'pulses' through the bed. Iliuta and Thyrion (1997) suggest that, for a column of inner diameter 45 mm, the transition from bubbling to pulsing flow occurs at a gas flow rate of $\sim$7 L min^{-1} (70 mm s^{-1}), and is largely independent of liquid flow rate. Specchia et al. (1974) predict a higher transition flow rate of $\sim$13 L min^{-1} (130 mm s^{-1}). The spray regime occurs when the gas flow rate is sufficiently high to entrain the majority of liquid as individual droplets.

Overall gas hold-up within these beds has been studied, most commonly, by calculating it from the liquid hold-up. Various approaches of determining liquid hold-up have been reported, including using the moments of tracer residence time distribution curves (e.g., Stiegel and Shah, 1977; Lamine et al., 1992; Lara-Marquez et al., 1992; Cassanello et al., 1998), the liquid level within the column (Goto and Gaspillo, 1992; Molga and Westerterp, 1997a, 1997b; Sivakumar et al., 1999), and the electrical conductivity (Achwal and Stepanek, 1976). All of these works report an increase in gas hold-up with increasing gas flow rate through the bed. However, the gas hold-up does not increase linearly with the gas flow-rate; the hold-up shows a greater change with flow rate at the lower gas flow rates. Lamine et al. (1992), Cassanello et al. (1998) and Sivakumar et al. (1999) all found that increased liquid flow rates were associated with decreased gas hold-up in the bed. Sivakumar et al. (1999) suggest this is due to an increase in bubble rise velocities within the bed. Achwal and Stepanek (1976) also showed that high liquid and gas flow rates were associated with a greater variation of gas hold-up as a function of bed height; with a higher gas hold-up at the exit of the bed than at its base. Lamine et al. (1992) and Cassanello et al. (1998) also reported that smaller packing sizes are associated with a higher gas hold-up. Thanos et al. (1996) measured the liquid hold-up in packed beds during co-current up-flow operation, and observed liquid hold-ups of above 0.7 at superficial gas velocities of less than 100 mm s^{-1} (equivalent to a gas flow rate of 1 L min^{-1} in the column of inner diameter 14 mm used). The liquid hold-up was observed to decrease with increasing gas flow rate, but no trend was observed with varying liquid flow rate. Similar results were observed by Lamine et al. (1992).

Several researchers have focused on characterising gas behaviour within packed beds, and two main approaches have been used. First, optical techniques have been used to measure bubble-rise velocities. In these approaches the optical properties of the liquid are modified so as to match the refractive index of the solid particles with that of the liquid; hence enabling identification of the gas present. Second, dissolution rates of gases have been used to calculate the interfacial area of the gas–liquid interface in the bed. This latter approach does not provide direct information on the bubble sizes within the bed; however, the interfacial area is, of course, related to the bubble size, bubble shape, and gas hold-up. Each of these approaches will now be considered briefly.

There are numerous examples of the use of optical techniques for tracking of gas bubbles; for example, Benkrid et al. (2002), Pokusaev et al. (2004a, 2004b), Bordas et al. (2006), Mena et al. (2008), Jo and Revankar (2009, 2010). In particular, Pokusaev et al. (2004b) released small numbers of bubbles of 0.4–2 mm in diameter into the centre of a packed bed of 3 mm diameter glass spheres, with n-decane as the liquid phase. It was found that the rise velocity of the bubbles increases significantly with bubble size. Further, substantial changes in the shape of the moving bubbles were observed. In some cases, a given bubble was described as moving 'as a train, slipping between closely packed particles of the granular bed, and, at times, it moves as an amoeboid that temporarily absorbs separate particles of the packing'. Larger gas 'bubbles' (which might take the form of coalesced amoeboid and train bubbles) with equivalent diameters of 3-4 mm were found to rise at velocities of order $\sim$100 mm s^{-1}. Individual spherical bubbles with diameters of less than 2 mm were found to rise more slowly at $\sim$10–40 mm s^{-1}. Bordas et al. (2006) performed experiments on a refractive index matched packed bed of square cross-section (30 mm $\times$ 30 mm), operating in up-flow and packed with uniform spheres in the size range 2–6 mm. Gas was injected into the bed using a needle valve. It was found that the bubble size was similar to the void space between the packing elements, and was largely unaffected by liquid velocity and viscosity. The bubbles also showed significant shape distortions, especially bubbles larger than the void size, which showed eccentricities (defined by Bordas et al. (2006) as the ratio of longest to shortest cord length measured within the bubble) of between 1 and 2.5. It is noted that the gas fraction in these beds was very low (i.e., gas flow rate fraction, β, $<2\%$), as required by the optical technique used. It is therefore likely that significant bubble coalescence or gas channelling might occur at higher values of β but would not be observed at the low values of β used in this earlier work. Bordas et al. (2006) and Pokusaev et al. (2004a, 2004b) also noted that bubbles could become trapped in the porous medium as they rise through the bed. These trapped gas regions bubbles will be 'static' (albeit not necessarily permanently) within the bed.

Use of the dissolution rates of gases to calculate the interfacial area of the gas–liquid interface in the bed has been reported by Specchia et al. (1974), Lara-Marquez et al. (1992), Molga and Westerterp (1997a, 1997b), Murugesan and Sivakumar, (2005), and Raghavendra Rao et al. (2010), amongst others. Interfacial area measurements do not directly measure the bubble sizes present in the packed beds, but they are influenced by bubble size and gas hold-up. Lower interfacial areas will be associated with one or all of: larger bubbles for a given volume of bubbles, lower gas hold-ups and bubbles with a higher sphericity. Specchia et al. (1974) studied the reaction of carbon dioxide with a solution of sodium hydroxide, from which the interfacial area was inferred. They observed that interfacial area increases with both gas and liquid flow rate within the bed. Similar findings were reported by Lara-Marquez et al. (1992) who studied air flow through a sodium sulphate solution, and CO_2 flow through diethanolamine, and Murugesan and Sivakumar (2005) and Raghavendra Rao et al. (2010), both of whom studied the flow of air through sodium sulphate solutions. Molga and Wedsterterp (1997a, 1997b) reported two studies of the CO_2 and diethanolamine system which highlight the potential influence of operating pressure on the interfacial area. For a