Characterising Gas Behaviour During Gas-Liquid Co-Current Up-Flow in Packed Beds Using Magnetic Resonance Imaging

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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.

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 & 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 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 & Sivakumar, 2002; Moreira & Freire, 2003; Iliuta & 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 & 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 ~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 & Shah, 1977; Lamine et al., 1992; Lara-Marquez et al., 1992; Cassanello et al., 1998), the liquid level within the column (Goto & Gaspillo, 1992; Molga & Westerterp, 1997a,b; Sivakumar et al., 1999), and the electrical conductivity (Achwal & 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 & Stepanek (1976) also showed that high liquid and gas flow rates was 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. (2004 a,b), Bordas et al. (2006), Mena et al. (2008), Jo & 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 ~100 mm s^{-1}. Individual spherical bubbles with diameters of less than 2 mm were found to rise more slowly at ~10 – 40 mm s^{-1}. Bordas et al. (2006) performed experiments on a refractive index matched packed bed of square cross-section (30 mm × 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,b) 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 & Westerterp (1997a,b), Murugesan & 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 & Sivakumar (2005) and Raghavendra Rao et al. (2010), both of whom studied the flow of air through sodium sulphate solutions. Molga & Wedsterterp (1997a,b) reported two studies of the CO_2 and diethanolamine system which highlight the potential influence of operating pressure on the interfacial area. For a reactor operating at near atmospheric pressure, they found that the interfacial area was significantly affected by gas superficial velocities of less than ~ 50 mm s^{-1} (~ 5 L min^{-1} volumetric flow of gas in the 45 mm i.d. bed being studied), consistent with the previous works discussed; while at higher gas pressures (2 – 65 atm) the gas flow rate had only a marginal effect on interfacial area. Further, Molga and Westerterp (1997a,b), Murugesan & Sivakumar (2005) and Raghavendra Rao et al. (2010) have all reported studies of the effect of packing size and shape on the interfacial area and found that interfacial area generally increases with packing size (based on the equivalent sphere diameter). In general, the results show interfacial areas per unit reactor volume of between 200 – 2000 m^2/m^3 over the wide range of experimental parameters considered.

In summary, previous work has investigated several aspects of the hydrodynamics in packed beds operated under conditions of co-current up-flow. Bubble sizes and rise velocities have been measured in packed beds during up-flow, but only at gas hold-ups low enough such that optical techniques can be used, which require substantially lower gas flow rates than used in operating packed beds. Bordas et al. (2006) and Pokusaev et al. (2004 a,b) both report bubbles becoming trapped within the void structure, however the fraction of trapped bubbles in an operating packed bed was not quantified.

Nuclear Magnetic Resonance (NMR) and Magnetic Resonance Imaging (MRI) techniques are commonly used to study single- and multi-phase flows (Fukushima, 1999; Mantle & Sederman, 2003). It is the purpose of this paper to demonstrate the use of a combination of MR techniques to study gas hydrodynamics during co-current up-flow within a packed bed. In particular, we demonstrate the use of MR to discriminate between discrete bubbles rising up

the bed and gas either trapped within regions of the void space or existing as gas channels within the bed. Measurements of gas bubble size, their shape and rise velocity are also presented. These data are obtained at gas and liquid flow rates reflecting realistic operating conditions. MR techniques are well established in application to mapping gas-liquid distribution in co-current down-flow in packed beds (e.g., Sederman and Gladden, 2001a), the velocity fields of both gas and liquid during trickle flow (Sankey et al., 2009), and the mechanism of the trickle-to-pulse transition during co-current down-flow in packed beds Anadon et al. (2006). MR is also used to characterise aspects of gas and liquid dynamics in gas-solid fluidised beds (e.g., Savelsberg et al., 2002; Müller et al., 2006). In Müller et al. (2006), ultra-fast one-dimensional MR profiling techniques were used to track bubble-rise velocity within a gas-liquid fluidised bed. The present work brings together MR techniques previously used to study gas-liquid flow in co-current down-flow in packed beds along with methods used to track bubble rise in gas-solid fluidised beds to explore the extent to which MR can give insight into gas behaviour during gas-liquid co-current up-flow in packed beds.

Experimental

A polytetrafluoroethylene (PTFE) column of length 80 cm and inner diameter 43 mm was used. The column was fitted with a gas distributor which produced bubbles of approximately 1 – 2 mm in diameter; the distributor was 2.5 cm high and 1 cm in diameter, and made of bonded irregular glass granules with a pore size of ~0.5 mm. The gas inlet was placed at the centre of the column, with two liquid inlets, spaced symmetrically around the centre of the bed, at a distance of 1 cm from the column wall. Non-return valves were placed onto both the liquid and gas lines. A single outlet at the top of the bed, open to the atmosphere, was used to return the liquid to a reservoir. The bottom of the column was filled with irregular 1 – 2 mm diameter non-porous ceramic cylindrical packing to a height of approximately 5 cm, above which the bed was packed with non-porous soda-lime glass spheres of diameter 1.8, 3 or 5 mm; sphere diameters are quoted ±10%. The beds were packed by pouring ~20 ml volumes of the spheres of interest into the bed after which the bed was consolidated by tapping. The bed was fully flooded following complete loading. This packing procedure resulted in a bed voidage of 0.37 ±0.01 for the 3 and 5 mm spheres, and 0.39±0.01 for the 1.8 mm spheres, as determined by NMR, consistent with previously reported data (Benenati & Brosilow, 1962; Thadani & Peebles, 1966). The fluids used in these experiments were air and water. Liquid was pumped into the bed using a Bronkhurst rotary pump (AB70 Electric Rotary), and the

flow rate was controlled using a mass flow controller (Bronkhurst Hi-Tec Cori-Flow M42). The gas flow rate was monitored and controlled manually using a rotameter. Liquid flow rates in the range 0-200 cm^3 min^{-1} and gas flow rates in the range 20-250 cm^3 min^{-1} were used; flow rates were measured to an accuracy of ±5%. Under these operating conditions the bed will be operating well within the bubble-phase flow regime (Iliuta & Thyrion, 1997; Turpin & Huntington, 1967; Khan et al., 1997; Murugesan & Sivakumar, 2002; Moreira & Freire, 2003). The liquid phase used was water doped with copper (II) sulphate at a concentration of 0.75 g/L. The T_1 of the resulting solution was 0.1 s. MRI data were acquired at a ^{1}H frequency of 199.7 MHz on a Bruker Spectrospin DMX 200, 4.7 T magnet with a birdcage coil of inner diameter 6.3 cm, with shielded gradient coils providing a maximum gradient strength of 13.5 G cm^{-1}. The imaging region was centred on a point ~50 cm above the inlet of the bed, extending ~4 cm above and below this point (for a total imaging field-of-view in the z-direction of 8 cm). The distance of the imaging volume from the distributor exceeds the 30 particle diameters suggested by Bordas et al. (2006) as the minimum distance required for the bubble size to stabilise within the bed. Five different MR experiments were performed on each of the 3 fixed beds. Note that in all the MR pulse sequences used, we acquire signal from the liquid phase only – no signal is obtained from the gas phase or the non-porous packing.

(i) A simple 'pulse-acquire' pulse sequence, with no spatial encoding, is used to measure the amount of liquid present in the bed, and hence determine the liquid hold-up. 128 scans with a recycle time of 1 s were acquired giving a total data acquisition time of 128 s. Measurements were made of both a fully water-saturated bed and the bed operating under conditions of co-current gas-liquid up-flow, from which the liquid hold-up and hence gas hold-up within the imaging region are determined.

(ii) Quantitative 1D profiles of the liquid content of the bed were acquired using a spatially-resolved, velocity-compensated spin-echo sequence (Pope & Yao, 1993). The velocity-compensated spin-echo pulse sequence measures the spatial distribution of liquid along the axial (z) direction of the bed. Signal is integrated in the x-y plane, and therefore the MR signal provides a measure of the total liquid hold-up at each axial location in the bed. The 1D profiles were acquired with a field-of-view of 10 cm, with 512 data points, thereby giving a spatial resolution of 195 μm in the z-direction. 1024 profiles, each of acquisition time 2.5 ms, were acquired with a recycle time of 0.5 s. A reference profile of a liquid-saturated bed was also

acquired under the same acquisition conditions. The T_1 relaxation time of 0.1 s achieved by doping the water with copper (II) sulphate enables the velocity-compensated profiles of water distribution along the length of the packed bed to be acquired (using the data acquisition parameters given) without influence of any T_1-contrast, in addition to the removal of velocity contrast in the data achieved by using the velocity-compensated pulse sequence. These data are used to determine the total gas hold-up at each of the 512 locations along the length of the bed.

(iii) To record 1D profiles of liquid content along the axial direction of the bed as fast as possible, such that bubbles rising with the bed can be tracked, a 1D gradient-echo FLASH (Fast Low Angle SHot) sequence was used (Haase et al., 1986). 8192 profiles, each with an acquisition time of 2.5 ms, were acquired in immediate succession; a recycle time of 20 ms and an excitation pulse of 20° were used. A vertical field-of-view of 10 cm with 512 data points, giving a spatial resolution of 195 μm, was used. A reference profile of a liquid-saturated bed was also acquired under the same acquisition conditions. While the FLASH pulse sequence allows faster acquisition rates (i.e., it requires a delay, referred to as the recycle time, of only 20 ms compared to the time period of 0.5 s for the velocity-compensated pulse sequence used in (ii)), the signal intensity of the acquired data will now include signal contrast due to the velocity of the flowing liquid. However, the purpose of this measurement is to track in real time the progress of bubbles within the bed and therefore all that is needed is discrimination between gas- and liquid-phase species. Time series of these profiles allow the rise velocity of the gas bubbles and the dimension of the bubbles along the z-axis to be calculated (Müller *et al*., 2006). The accuracy of this measurement increases with increase in bubble size since it is easier to identify the gas-liquid interface for larger bubbles.

(iv) 2D images of the bed were acquired using a rapid acquisition and relaxation enhancement (RARE) sequence (Hennig et al., 1986). Vertical (x-z) slice images through the centre of the bed were acquired using a single slice selective excitation pulse. The imaging slice thickness was nominally 2 mm. A Gaussian-shaped slice selective pulse was used with a bandwidth of 3300 Hz. The images were acquired in a 64×64 data array, with a recycle time of 0.5 s. The effective echo time for the images was 9.8 ms. A train of 64 echoes was acquired for each excitation. The x-z images had a field-of-view of 50 mm × 70 mm, with an in-plane resolution of 780 μm × 1.1 mm. A sequence of 512 images was acquired with a total acquisition time of 5

min. A reference image of a liquid-saturated bed was also acquired. Comparison of the images of the bed recorded during operation with the reference image allows the gas-filled regions of the bed to be identified. Morphological analysis of these regions (Baldwin et al., 1996) allowed estimates of bubble size in the vertical (z) and horizontal (x) directions to be determined. For completeness, 2D images in the x-y plane were also recorded. The x-y images had a field-of-view of 50 mm × 50 mm, with an in-plane resolution of 780 μm × 780 μm. As with the x-z images, a sequence of 512 images was acquired with a total acquisition time of 5 min. The x-y images were acquired to confirm that the values of bubble dimension in the x-direction determined for the x-z images are indeed consistent with the equivalent dimension determined from the x-y images. The bubble shape characteristics reported are taken only from the x-z images.

(v) Full 3D structural images of the liquid-saturated packed beds were acquired using a 3D spin-echo sequence (Sederman et al., 2001a). Data were acquired over an imaging region of 50 mm × 50 mm × 100 mm containing 256 × 256 × 512 voxels, thereby giving an isotropic voxel size of 195 μm. A recycle time of 100 ms was used with 8 averages to give a total acquisition time of ~14 h. Morphological analysis (Baldwin et al., 1996) of the 3D image data was used to provide information on the structure of the void spaces between packing elements (Sederman et al., 2001b).

Gravimetric measurements were used to provide an independent measure of gas hold-up for comparison with the MR measurements. The column was suspended, via a collar attached 5 cm below the outlet, from an electronic scale (Salter SA3N253), which had an accuracy of ±5 g. The inlet and outlet pipes were supported separately from the column, such that their contribution to the measured weight is reduced. The weight of the liquid within the packed bed saturated with liquid was recorded, W_0, as well as the weight during the operation of the bed, W. The total gas hold-up in the bed is then given by 1- (W/W_0). The average error in the gravimetric measurement of gas hold-up was calculated to be ±0.035. Agreement, to within experimental error, of the total gas hold-up determined from the gravimetric measurements with the values obtained from the 'pulse-acquire' measurements (i) and the spatially-resolved, velocity compensated profiles, integrated along the length of the column (ii), was obtained.

Data Analysis

The procedures implemented for analysis of the various MR datasets acquired are now summarised.

Measurement of gas hold-up

The fraction of the void space of the bed occupied by gas, H_{gT}, is calculated from the MR 'pulse-acquire' data which provides a measure of the total amount of liquid in the imaging volume. Hence:

$$H_{gT} = 1 - I\!\!\Big/\!\!I_0 \; ,$$

(1)

where I and I_0 are the signal intensity recorded for the bed during operation and the water-saturated bed, respectively. MR is an intrinsically quantitative technique, in which the signal I is directly proportional to the number of ^1H nuclear spins, and hence number of water molecules, present.

Identification of gas-filled regions which remain constant over the measurement time

In this analysis the velocity-compensated spin-echo measurements are used. The profiles give a quantitative measure of liquid hold-up, spatially resolved along the length of the bed:

$$H_l(z) = I(z)/I_0(z).$$

(2)

Hence, the gas hold-up is given by:

$$H_g(z) = 1 - H_l(z).$$

(3)

Successive 1D profiles are then plotted to produce a 2D (position-time) data array $H_l(z,t)$. An example of a dataset acquired is shown in Fig. 1a; high intensity (light) corresponds to regions of high liquid content, and low intensity (dark) corresponds to regions of high gas content.

Two general features are seen in these datasets. First, near-vertical dark lines are observed. These features are associated with moving gas bubbles rising within the bed; i.e. the dynamic gas hold-up. The ultra-fast 1D profile recorded with the FLASH pulse sequence to be described shortly, and described in Fig. 2, records the vertically-rising bubbles with higher temporal resolution and it is from these profiles that the bubble rise velocities are calculated. Second, at positions along the height of the bed, bands of lower signal intensity (grey to black) are seen, indicating that they are filled with lower amounts of liquid at all the times over which the experiment was performed. It is noted that within a band of high gas hold-up (a horizontal band of lower intensity), the gas could be stationary or that volume of the bed might be filled with a continuous stream of gas. We refer to the 'horizontal' bands of low signal intensity as regions containing 'static' gas hold-up – in contrast to the clearly identified dynamic gas hold-up associated with rising bubbles. Estimates of dynamic and static gas hold-up are obtained from datasets such as those shown in Fig. 1a as follows.

The principle of the analysis used to estimate the static gas hold-up is described with reference to Fig. 1b which is a schematic representation of the experimental dataset shown in Fig. 1a. Considering Fig. 1b, 3 low intensity (dark) horizontal bands are shown, indicating the presence of some gas at all measurement times at each of those 3 positions along the length of the column. Multiple near-vertical lines are also seen which are associated with dynamic gas hold-up (i.e., rising bubbles). Figures 1 c,d show plots of the mean gas hold-up as a function of axial position along the length of the bed. The mean gas hold-up profile of the bed, $\overline{H_g(z)}$, is calculated as follows:

$$\overline{H_g(z)} = 1 - \frac{\sum_{t=1}^{T} H_l(z,t)}{T} . \tag{4}$$

Figure 1d explains how the estimates of static and dynamic gas hold-up are assigned using the schematic data array $H_l(z,t)$ shown in Fig. 1b. Assuming that the dynamic gas (i.e., the rising gas bubbles) will pass through all heights of the bed (over the imaging height of 8 cm), and that by averaging over a long time, all positions will see, on average, a similar volume of gas passing through them, the form of $\overline{H_g(z)}$ will take that shown in Fig. 1d. The dynamic gas hold-up, $H_{gD}(z)$, is seen as the baseline upon which the 'peaks' in gas hold-up sit – these

'peaks' are characteristic of the static gas hold-up in the bed. Making the above assumption, $H_{gD}(z)$ is constant along the length of the bed. Figure 1c shows $\overline{H_g(z)}$ for the experimental dataset. In this case, identification of $H_{gD}(z)$, and hence the static and dynamic gas hold-up is less clear but making the same assumption that the time averaged $H_{gD}(z)$ is constant along the length of the bed and that some of the bed has no static gas holdup, we can take the minimum value of $\overline{H_g(z)}$ to be the dynamic gas holdup $H_{gD}(z)$. In practice, we also wish to avoid variations due to noise so the threshold is set at this minimum value plus the noise level of the signal. The appropriate gating level, G, is therefore defined as:

$$G = \min\left(\overline{H_g(z)}\right) + \frac{\sqrt{\dfrac{1}{Z}\displaystyle\sum_{z=1}^{Z}\left(I_0(z) - \overline{I_0}\right)^2}}{\overline{I_0}} \tag{5}$$

where the second term represents the noise in the data as calculated from the standard deviation of the signal intensity at each axial position in the reference dataset (i.e. for the fully water-saturated bed). This gating level then accounts for the variability in the recorded signal which is not due to the presence of additional static gas within the bed. When $H_g(z) \leq G$ there is assumed to be only dynamic hold-up, such that $\overline{H_g(z)} = H_{gD}(z)$. When $H_g(z) > G$, there is both dynamic and static gas hold-up such that $H_g(z) = H_{gD}(z) + H_{gS}(z)$. These regions are highlighted on Fig. 1c,d by the shaded areas. In practice the inclusion of the second term to the threshold value has only a small change on the calculated values, and does not affect the overall trend in the data. The average dynamic hold-up is then equal to

$$\overline{H_{gD}} = \frac{1}{T}\sum_{z=1}^{Z}\left(\begin{array}{cc} \overline{H_g(z)} & H_g(z) \leq G \\ G & H_g(z) > G \end{array}\right), \tag{6}$$

and the static hold-up equal to

$$\overline{H_{gS}} = \frac{1}{T}\sum_{z=1}^{Z}\left(\begin{array}{cc} 0 & H_g(z) \leq G \\ H_g(z) - G & H_g(z) > G \end{array}\right). \tag{7}$$

A further term, the fraction of static gas hold-up relative to total gas hold-up is given by, F_{gS}:

$$F_{gS} = \frac{\overline{H_{gS}}}{\overline{H_{gT}}} \qquad , \qquad (8)$$

which describes the contribution of the static gas to the overall gas hold-up in the bed.

Determination of bubble size in the vertical direction and bubble rise velocity

The data used in this analysis are the 1D gradient-echo FLASH profiles. An estimate of $H_g(z,t)$ is obtained using the method described by eqns. (2) and (3). The profiles are only an estimate because the FLASH profiles obtained in this acquisition will include influences on the acquired signal intensity (referred to as 'weightings') from the flow of the water in the system as well as T_1 relaxation time contrast. The method used to determine the rise velocity of bubbles has been used previously by Müller et al. (2006) to track rising gas bubbles and bubble size in a gas-solid fluidised bed. Figure 2 shows a schematic identifying how the velocity of a rising bubble is obtained from these data. A gas bubble rising up a column is shown at 4 time points; T0, T1, T2 and T3. At T0, the bubble has not yet entered the column, and hence the region of the column in the field-of-view is completely filled with liquid and therefore associated with uniform signal intensity in Fig. 2b. The bubble then enters the column and the signal intensity at the axial positions at which the bubble exists shows a decrease in signal intensity resulting from the lower liquid content in that region (Fig. 2b, profiles T1, T2, T3). The resulting signal intensity in the 2D map is shown in Fig. 2c. The position of a gas bubble within the bed is therefore identified as a contrast difference between light (liquid-containing) and dark (gas-containing) regions. It follows that, if such a boundary is seen to move upwards in successive profiles recorded as a function of time, the position of the boundary allows us to track the bubble rising up the bed. The gradient of the line formed by the points defining the boundary of the gas and liquid in each profile (for any given bubble) gives us a direct measure of the bubble rise velocity in the bed. Figure 3a shows how these 'tracks' of bubble rise velocity are identified in real experimental data.

The bubble size in the direction of flow can be obtained from the profiles in two ways. As shown in Fig. 2b,c it is possible to estimate this bubble dimension from the vertical dimension of the dark band characterising the presence of the bubble at a given point in time. This approach has the disadvantage that for the case of some larger bubbles (or bubble trains), their length is not fully captured within a single 2D map. The alternative approach, and the one adopted here, is shown in Fig. 3b. The time for the bubble to pass through a given height z in

the bed is found and the vertical height of the bubble is then calculated by multiplying the rise velocity by the passage time, Δt.

Obtaining Bubble Sizes from 2D Images

Figure 4 illustrates the data analysis procedure used to determine bubble sizes from the 2D RARE images. Figure 4a shows an x-z image acquired during up-flow. Figure 4b shows that image following a binary-gating procedure used to discriminate between liquid-containing pixels (white) and those containing gas or solid (black). No signal intensity is associated with gas or solid phases. Figure 4c shows the binary-gated reference image of the liquid-saturated bed; this binary-gated map identifies the location of void space (white) and solid packing elements (black) within the image. Finally, the result of subtracting Fig. 4b from Fig. 4c is shown in Fig. 4d); high intensity white pixels correspond to the location of gas in the original image. Single-pixel bubbles have been discounted in this study, as they have a high likelihood of being associated with noise. Figure 4d provides strong evidence in support of discrete gas bubbles existing within the bed, in agreement with the reports of earlier workers (Iliuta & Thyrion, 1997; Turpin & Huntington, 1967; Khan et al., 1997; Murugesan & Sivakumar 2002, 2005; Moreira & Freire, 2003). From the x-z images alone, it is not possible to demonstrate that each of the gas-filled pixels are not connected in 3D space. However, comparison of the x-z images with the x-y images acquired under the same conditions suggests that this is not the case.

The number and size of these regions are then calculated using the morphological analysis of Baldwin et al. (1996). The analysis identifies contiguous regions, and calculates their area, and from the first and second moments of the regions, the radius of the region (calculated as the radius of gyration of the bubble about its mean position). The minimum bubble radius detectable in these images is ~ 0.6 mm. The aspect ratio e is also calculated from the x-z images as defined by eqn. 9:

$$e = r_z / r_x \, , \tag{9}$$

where r_z and r_x are the radial dimensions in the x- and y-directions, respectively. In cases where the z-dimension of the bubbles or bubble trains do extend beyond the field-of-view of the imaging coil, then e will be an underestimate of the true value.

3D Characterisation of the Void Space of the Bed

Morphological analysis was applied to the high resolution 3D images to characterise the void space within the packed bed and, in particular, to yield a value of the mean void radius. A detailed discussed of the method of analysis is given elsewhere (Baldwin et al., 1996; Gladden & Alexander, 1996).

Results and Discussion

Figures 5a,b show plots of H_{gT} as a function of gas and liquid flow rates (Q_g and Q_l respectively) for glass sphere packings of 1.8, 3 and 5 mm diameter. The error bars are calculated from the compound error of the variability in the signal intensity measured during the flowing experiments, and that recorded during the reference measurement of the liquid-filled bed. The NMR measurements of H_{gT} are in agreement with gravimetric measurements (not shown) within the bounds of experimental error. Considering first the effect of gas flow rate, Fig. 5a) shows an increase in the gas hold-up in the bed with increasing gas flow rate, as expected. For a given flow rate, a significant increase in gas hold-up with decreasing packing size is observed. With reference to Fig. 5b, the gas hold-up shows a small decrease with increasing liquid flow rate. It is noted that the gas hold-up in the co-current up-flow orientation, typically <0.3, is significantly lower than that observed in co-current down-flow experiments (typically ~0.6) for the same gas and liquid flow rates (Sederman & Gladden, 2001a; Anadon et al., 2006). Hold-ups measured here are similar to those predicted using correlations such as those presented by Ellman et al. (1990).

As well as examining the hold-up in terms of the absolute liquid and gas flow-rates, the variation in terms of the mean gas flow rate ratio, β, is examined. This is defined as the ratio of superficial gas velocity, to the total superficial velocity of both fluids within the bed (Bordas et al., 2006):

$$\beta = \frac{Q_g}{Q_g + Q_l} . \tag{10}$$

The ratio β represents the uncompressed volume fraction of gas as a total volume of fluid entering the bed. The ratio H_{gT}/β thus represents the occupation of the bed by the gas relative to the inlet flow rates; a value of 1 representing equal gas and liquid average velocities within the bed. Values of less than 1 represent the gas flowing at a higher velocity than the liquid through the bed, assuming gas compression and expansion in the bed is not significant. Figure 6 shows H_{gT}/β ratio as a function of β. As β increases, the value of H_{gT}/β falls, confirming that relative gas velocity in the bed increases as the quantity of gas flowing into the bed increases. However, for $\beta > 0.5$, H_{gT}/β remains relatively constant in the range 0.2 - 0.4, suggesting that the gas hold-up in the bed is then proportional to the fraction of gas entering the bed, and has a mean velocity significantly higher than that of the liquid phase.

Figure 7 shows plots of static, H_{gS}, and dynamic, H_{gD}, gas hold-up as a function of the mean gas flow rate ratio, β. Considering first Fig. 7a, which shows a plot of H_{gS} against the gas flow rate ratio β, it is seen that the static gas hold-up increases with decreasing packing size. This observation is consistent with the gas being trapped by capillary forces between the packing elements. Static gas hold-up also increases with increasing β for all packing sizes. Table 1 reports the values of static gas hold-up relative to total gas hold-up as a function of gas flow rate and packing size for two different gas flow rates at a constant liquid flow rate of 50 ml min^{-1}. The data show that the static gas phase represents a significant proportion of the total gas hold-up within the bed for all gas flow rates considered, and that it increases significantly with decreasing packing size. The dynamic hold-up fraction H_{gD} is plotted as a function of β in Fig. 7b and, as expected H_{gD} increases with β. However, considering the error bars of the measurements, the values for H_{gD} are similar for the 1.8 mm and 3 mm, and only slightly lower for the 5 mm packing.

In summary, the MR data clearly support the results of previous workers, in that while some gas remains trapped within the bed due to capillary forces, the remaining gas rises through the bed as discrete bubbles (Bordas et al., 2006, Pokusaev et al., 2004a,b). Combining the results shown in Fig. 7, the data suggest that the differences in gas hold-up between the different packing sizes are largely due to stronger capillary forces between smaller packing elements trapping a larger amount of gas in the bed; 'excess' gas then rises through the bed.

Figure 8 shows the bubble size distribution for the 3 mm glass spheres. The image resolution

gives a minimum detectable bubble radius of 0.6 mm. Bubbles closer together than the pixel resolution of the image (780 μm) will not be individually identified, therefore 'swarms' or 'flocks' of small bubbles travelling together in, for example, bubble trains (Pokusaev et al., 2004b), will be identified as a single long bubble in the present work. As seen in Fig. 8, which shows data recorded for a constant liquid flow rate of 50 ml min⁻¹, there is a general increase in bubble size with increasing gas flow rate, from a mean volumetric radius of 4.6 mm at a gas flow rate of 20 ml min⁻¹, to 6.0 mm at a flow rate of 250 ml min⁻¹. The bubble size distributions show there is a significant fraction of large bubbles, defined here as r >10 mm, which increases with increasing gas flow rate; from a volume fraction of 0.045 at 20 ml min⁻¹ to 0.17 at 250 ml min⁻¹. There is an associated decrease in the relative number of smaller bubbles (r < 5 mm) from 0.62 to 0.44. This observation is most likely explained by an increased rate of bubble coalescence within the bed at higher gas hold-ups.

The data shown in Fig. 8 suggest larger bubble sizes existing within the bed than have been reported by earlier workers, who used visual observations (Bordas et al., 2006, Pokusaev et al., 2004a,b). These workers reported, typically, bubble diameters in the range 0.5 – 5 mm. In making this comparison we note, in particular, the low gas flow rates used in the work of Bordas et al. (2006). An estimate of predicted interfacial gas-liquid surface area based on our bubble-size data give values of $100 - 200$ m²/m³, which are at the lower limit of those reported by Molga & Westerterp (1997a,b), Murugesan & Sivakumar (2005) and Raghavendra Rao et al. (2010), who reported values in the range $200 - 2000$ m²/m³ per unit volume of the reactor. This discrepancy is consistent with the MR technique not being sensitive to the resolution of bubbles of radius <0.6 mm. We note that in the distributions shown in Figs. 8 and 9, the median bubble size is significantly larger than the minimum detectable bubble size. We estimate that <5% of the gas volume was associated with bubbles of radius <0.6 mm.

The effect of the packing size on the bubble-size distribution for a gas and liquid flow rate of 50 ml min⁻¹ is shown in Fig. 9. The distributions show an increase in the smaller bubbles with increasing packing size, with a noticeable peak at 1.4 mm for the 5 mm packing. Table 2 shows the statistics of the bubble-size distributions shown in Figure 9, along with the relevant characteristics of the void space comprising the packed bed. The bubbles in the 1.8 mm and 3 mm packings are significantly larger than the size of the void space between packing elements, and the ratio of bubble size to void size increases with decreasing packing size. In the 5 mm

packing, the bubbles are closer to the size of the packing voids, with the peak observed at 1.4 mm being similar to the modal void size of 1.5 mm. This suggests that in the larger packings the bubbles begin to travel predominantly between the packing elements, rather than encompassing them.

Elongation of the bubbles in the direction of flow was observed for all packings. The aspect ratio, e, was found to be 1.58±0.11, 1.53±0.12 and 1.44±0.14 for the 1.8, 3.0 and 5.0 mm packings, respectively, for gas and liquid flow rates of 50 ml min^{-1} and 250 ml min^{-1}, respectively. The errors are based on the mean error due to the resolution in determining the aspect ratio of the bubbles. There was found to be no significant variation in the aspect ratio as a function of either liquid or gas flow rate. A similar effect, described as 'bubble trains' and amoeboid structures, was reported by Pokusaev et al. (2004b); such gas structures are likely to be interpreted as a single bubble of a high aspect ratio in the present work. Bordas et al. (2006), also noted that single bubbles could be elongated in the direction of flow.

As described earlier, the rise velocity of these bubbles can be calculated from the time sequence of rapidly acquired 1D gradient echo profiles. Figure 10 shows the average bubble rise velocities for bubbles with of r_z = 15 mm; the data shown represent an average of ~20 bubbles of that radius rising through the bed. Figure 10 shows the bubble rise as a function of (a) gas flow rate for a constant liquid flow rate of 50 ml min^{-1}, and (b) liquid flow rate at a constant gas flow rate of 50 ml min^{-1}. Considering first Fig. 10a, the bubble rise velocity within the 1.8 and 3 mm packings are relatively independent of gas flow rate; with the rise velocity being slightly faster in the 3 mm packing than in the 1.8 m packing. In contrast, the rise velocity of the bubble in the 5 mm packing is significantly faster for all gas flow rates and shows an increase with gas flow rate, increasing from 275 mm s^{-1} at a gas flow rate of 20 ml min^{-1} to 303 mm s^{-1} at a gas flow rate of 250 ml min^{-1}. Similar trends are observed as a function of liquid flow rate, Fig. 10b, although there is also a small increase in bubble rise velocity as a function of liquid flow rate of the bubble within the 1.8 and 3 mm packings. A clear trend was observed for large bubbles to rise faster through the bed, with the rise velocities in the range ~100 – 400 mm s^{-1}. A similar trend is observed in data recorded at other flow rates. This is in agreement with what is seen in unpacked columns, where the bubble rise velocity generally increases with increasing bubble size (e.g., Peebles & Garber, 1953). These velocities are slightly lower than those seen for individual bubbles in a near infinite media, which for bubbles of radius 8 – 15 mm are ~ 200 – 450 mm s^{-1} (Kulkarni &

Joshi, 2005). The measurement of the average velocity of smaller bubbles (radius ≤10 mm) gave a typical value of ~10-20 mm s^{-1}. These estimates are in good agreement with the velocities measured for the small bubbles observed by Pokusaev et al. (2004a,b) and Bordas et al. (2006).

In summary, our results are broadly consistent with the observations of earlier workers. However, in the earlier literature, there appears to be some debate as to whether larger bubbles move faster within the bed. Our results are in agreement with the data of Pokusaev et al. (2004a) who studied the release of single bubbles into a packed bed of 7 mm spheres, and measured rise velocities of between 10 – 40 mm s^{-1}; these workers saw an increase in rise velocity with bubble size. Further experiments (Pokusaev et al., 2004b) on bead packs of 3 mm and 6 mm, again saw similar increases in bubble rise velocity. A sharp increase was seen in the rise velocities of bubbles above effective diameters of 2 mm (referred to as bubble ensembles in their results), which were found to have rise velocities of 100 – 150 mm s^{-1}. These bubbles were noted to travel as elongated trains, slipping between the packing elements, and as amoeboids which temporarily surrounded packing elements. Bordas et al. (2006) also observed bubble rise velocities of 20 - 100 mm s^{-1} for bubbles of similar size to the packing void size. Although they also reported that the bubble rise velocity decreased as the bubble size increased, we note that they only considered bubbles up to 3 times the void size. As stated earlier, this is most likely related to the low total gas hold-ups considered in that work.

Thus, drawing these results together, the bubble population appears to be comprised of small bubbles – of order of the packing size – which move between the packing elements within the bed, along with much larger bubbles, or agglomerates of bubbles, which move with higher rise velocities. Whether these 'larger' bubbles are streams of smaller bubbles, or amoeboid bubbles is, perhaps, less important than the conclusion that they behave as a single large bubble in terms of the observed high rise velocity (i.e. a large bubble moving at 100 – 300 mm s^{-1}) as opposed to a collection of small bubbles all moving with rise velocities of 10 – 40 mm s^{-1}.

Conclusions

The bubble hydrodynamics within a co-current up-flow packed bed have been successfully studied using a range of MR techniques. The overall gas hold-up within the system is in agreement with that obtained using gravimetric measurements. Measurements were conducted

using a range of magnetic resonance techniques to study the gas hold-up, bubble-size distribution, and bubble-rise velocities as a function of flow rate and packing size. The gas hold-up within the beds was found to increase with gas flow rate, while decreasing with packing size and to a lesser extent with liquid flow rate. Some regions of the bed were associated with a constant gas hold-up over the measurement time. This static gas hold-up is strongly dependent on packing size, showing an increase with decreasing packing size. The remainder of the gas passes through the bed as moving bubbles, elongated in the direction of flow. For smaller packings, the mean bubble size was found to be significantly larger than the inter-packing void sizes, suggesting that a single bubble can surround multiple packing elements. Rise velocities of the larger bubbles in the bed were ~100 – 300 mm s^{-1}. The rise velocities of smaller bubbles within the system, were of order ~10-40 mm s^{-1}. Whilst the MR data cannot unambiguously reveal whether the 'large bubbles' are a single large continuous gas bubble surrounding many packing elements, or a stream of small bubbles which cannot be resolved in the image, the dynamics of these gas 'structures' are such that they rise as a single entity.

References

Achwal S.K., and Stepanek J.B., Holdup profiles in packed beds. *Chem. Eng. J.* **12**, 69 – 75 (1976).

Anadon L.D., Sederman A.J. and Gladden L.F.*,* Mechanism of the trickle-to-pulse flow transition in fixed-bed reactors. *AIChE J* **52**, 1523-1532 (2006).

Attou, A. and Ferschneider, G., A simple model for pressure drop and liquid holdup in packed bed bubble reactors. *Chem. Eng. Sci.* **54**, 5139-5144 (1999).

Baldwin, C.A., Sederman, A.J., Mantle, M.D., Alexander, P. and Gladden, L.F., Determination and characterization of the structure of a pore space from 3D volume images. *J. Colloid Interface Sci.* **181**, 79-92 (1996).

Benenati R.F. and Brosilow C.B., Void Fraction Distribution in Beds of Spheres. *AIChE J* **8**, 359 – 361 (1962).

Benkrid K., Rode S., Pons M.N., Pitot P. and Midoux N., Bubble flow mechanisms in trickle beds – an experimental study using image processing. *Chem. Eng. Sci.* **57**, 3347-3358 (2002).

Bordas M.L., Cartellier A., Sechet P., Boyer, Ch., Bubbly flow through fixed beds: microscale experiments in the dilute regime and modelling**.** *AIChE J* **52**, 3722-3743 (2006).

Bouteldja, H., Hamidipour, M. and Larachi, F., Hydrodynamics of an inclined gas-liquid concurrent upflow packed bed. *Chem. Eng. Sci.* **102**, 397-404 (2013).

Cassanello M., Martinez O. and Cukierman, A.L., Liquid hold-up and backmixing in cocurrent upflow three-phase fixed-bed reactors. *Chem. Eng. Sci.* **53**, 1015-1025 (1998).

Ellman, M.J., Midoux, N., Wild, G., Laurent, A. and Charpentier, J.C., A new, improved liquid hold-up correlation for trickle-bed reactors. *Chem. Eng. Sci.* **45**, 1677-1684 (1990).

Fukushima, E., Nuclear magnetic resonance as a tool to study flow. *Ann. Rev. Fluid Mech.* **31**, 95-123 (1999).

Gladden, L.F. and Alexander, P., Applications of nuclear magnetic resonance in process engineering. *Meas. Sci. Technol.* **7** 423-435 (1996).

Goto, S. and Gaspillo, P.D., Multiple hydrodynamic states in gas-liquid upflow and downflow through beds of small packings. *Ind. Eng. Chem. Res.* **31**, 629-632 (1992).

Haase, A., Frahm, J., Matthaei, D., Hanicke, W. and Merboldt, K.D., FLASH imaging, rapid NMR imaging using low flip-angle pulses. *J. Magn. Res.* **67**, 194 - 199 (1986).

Hennig, J., Nauerth, A. and Friedberg, H., RARE Imaging: a fast imaging method for clinical MR. *Magnet. Reson. Med.* **3**, 823-833 (1986).

Hofmann, H., Cocurrent upflow in fixed beds. *In "Handbook of Fluids in Motion" edited by N.P. Cheremisinoff and R. Gupta. Ann Arbor Science Publishers (1983) ISBN-13: 978-0250404582.*

Iliuta, I. and Thyrion F.C., Flow regimes, liquid hold-ups and two phase pressure drop for two-phase cocurrent downflow and upflow through packed beds: air/Newtonian and non-Newtonian liquid systems. *Chem. Eng. Sci.* **52**, 4045-4053 (1997).

Jo, D. and Revankar, S. T., Bubble mechanisms and characteristics at pore scale in a packed-bed reactor. *Chem. Eng. Sci.* **64**, 3179-3187 (2009).

Jo, D. and Revankar, S. T., Effect of coalesence and breakup on bubble size distribtions in a two-dimensional packed bed. *Chem. Eng. Sci.* **65**, 4231-4238 (2010).

Khan, A., Khan, A.A. and Varma, Y.B.G., Flow regime identification and pressure drop in co-current gas-liquid upflow through packed beds. *Bioprocess Eng.* **16**, 355–360 (1997).

Kishore Kumar, R., Ragavendra Rao, A.V., Sankarshana, T. and Khan, A., Liquid holdup in concurrent gas liquid upflow through packed column with random and corrugated structured packing. *Proceedings of the World Congress on Engineering and Computer Science (WCECS) 2012 Vol II*, San Francisco, USA.

Kulkarni, A.A. and Joshi J.B., Bubble formation and bubble rise velocity in gas-liquid systems: A Review. *Ind. Eng. Chem. Res.* **44**, 5873-5931 (2005).

Lamine, A.S., Colli-Serano, M.T. and Wild, G., Hydrodynamics and heat transfer in packed beds with liquid upflow. *Chem. Eng. Sci.* **47**, 3493-3500 (1992).

Lara-Marquez, A., Larachi, F., Wild, G. and Laurent A., Mass transfer characteristics of fixed beds with cocurrent upflow and downflow. A special reference to the effect of pressure. *Chem. Eng. Sci.* **47**, 3485-3492 (1992).

Mantle, M.D. and Sederman, A.J., Dynamic MRI in chemical process and reaction engineering. *Prog. Nucl. Magn. Reson. Spectrosc.* **43**, 3-60 (2003).

Mena, P.C., Rocha, F.A., Teixeira, J.A., Sechet, P., Cartellier, A. Measurement of gas phase characteristics using a monofibre optical prove in a three-phase flow. *Chem. Eng. Sci* **63**, 4100-4115 (2008).

Molga, E.J. and Westerterp, K.R., Gas-liquid interfacial area and holdup in a cocurrent upflow packed bed bubble column reactor at elevated pressures. *Ind. Eng. Chem. Res.* **36**, 622-631 (1997a).

Molga, E.J. and Westerterp, K.R., Experimental study of a cocurrent upflow packed bed bubble column reactor: pressure drop, holdup and interfacial area. *Chem. Eng. Process* **36**, 489-495 (1997b).

Moreira, M.F.P. and Freire, J.T., Influence of gas and liquid flow rates and the size and shape of particles on the regime flow maps obtained in cocurrent gas-liquid downflow and upflow through packed beds. *Ind. Eng. Chem. Res.* **42**, 929-936 (2003).

Müller, C.R., Davidson, J.F., Dennis, J.S., Fennell, P.S., Gladden, L.F., Hayhurst, A.N., Mantle, M.D., Rees, A.C., and Sederman, A.J., Real-time measurement of bubbling phenomena in a three-dimensional gas-fluidized bed using ultra-fast magnetic resonance imaging. *Phys. Rev. Letts.*, **96** 154504 (2006).

Murugesan, T. and Sivakumar, V., Pressure drop and flow regimes in cocurrent gas-liquid upflow through packed beds. *Chem. Eng. J.* **88**, 233–243 (2002).

Murugesan, T. and Sivakumar, V., Liquid holdup and interfacial area in cocurrent gas-liquid upflow through packed beds. *J. Chem. Eng. Jpn* **38**, 229–242 (2005).

Peebles, F.N, and Garber, H.J., The motion of gas bubbles in liquid. *Chem. Eng. Prog.* **49**, 88-97 (1953).

Pokusaev, B.G., Karlov, S.P. and Schreiber, I., Immersion tomography of a gas-liquid medium in a granular bed. *Theor. Found. Chem. Eng.* **38**, 1 – 5 (2004a).

Pokusaev, B.G., Kazenin, D.A. and Karlov, S.P., Immersion tomographic study of the motion of bubbles in a flooded granular bed. *Theor. Found. Chem. Eng.* **38**, 561 – 568 (2004b).

Pope, J.M. and Yao, S., Quantitative NMR imaging of flow. *Concept Magnetic Res.* **5**, 281-302 (1993).

Raghavendra Rao, A. V., Kumar, R.K. and Sankarshana, T. and Khan., A. Interfacial area for concurrent gas-liquid upflow through packed bed. *Can. J. Chem. Eng.* **88,** 929 - 935 (2010).

Sankey, M.H., Holland, D.J., Sederman, A.J and Gladden L.F., Magnetic resonance velocity imaging of liquid and gas two-phase flow in packed beds. *J. Magn. Reson.* **196**, 142-148 (2009).

Savelsberg, R., Demco, D.E., Blümich, B and Stapf, S., Particle motion in gas-solid fluidized granular systems by pulsed field gradient nuclear magnetic resonance. *Phys. Rev. E* **65**, 020301 (2002).

Sederman A.J. and Gladden L.F., Magnetic resonance imaging as a quantitative probe of gas-liquid distribution and wetting efficiency in trickle-bed reactors. *Chem. Eng. Sci*. **56**, 2615-2628 (2001a).

Sederman A.J., Alexander P. and Gladden L.F., Structure of packed beds probed by Magnetic Resonance Imaging. *Powder Tech.* **117**, 255-269 (2001b).

Sivakumar, V., Nirmala, G.S. and Murugesan, T., Gas phase hold-up in gas-liquid cocurrent upflow packed bed reactors at low Reynolds number. *Bioprocess Eng.* **21,** 41-43 (1999).

Specchia, V., Sicardi, S. and Gianetto, A., Absorption in packed towers with concurrent upward flow. *A.I.Ch.E. J.* **20,** 646-653 (1974).

Stiegel G.J., and Shah, Y. T., Backmixing and liquid hold-up in a gas-liquid cocurrent upflow packed column. *Ind. Eng. Chem. Proc. DD* **16**, 37 – 43 (1977).

Thadani M.C. and Peebles F. N., Variation of local void fraction in randomly packed beds of equal spheres. *Ind. Eng. Chem. Proc. DD* **5**, 265-268 **(**1966).

Thanos, A.M., Galtier, P.A., and Papayannakos, N.G., Liquid flow non-idealities and hold-up in a pilot scale packed bed reactor with concurrent gas-liquid upflow. *Chem. Eng. Sci.* **51**, 2709-2714 (1996).

Turpin J.L. and Huntington R.L., Prediction of pressure drop for two-phase two-component concurrent flow in packed beds. *A.I.Ch.E. J.* **13**, 1196-1202 (1967).

Figure Captions

Figure 1: Format of the data obtained and associated analysis. The MR data shown are of the velocity-compensated, spin-echo axial profiles of liquid content with the bed. Successive 1D profiles are plotted to produce a 2D (position-time) data H_l(z,t). A typical experimentally-acquired dataset of liquid hold-up is shown in (a). An idealised 2D map of axially-resolved liquid hold-up is shown in (b). The resulting gas hold-up profiles as a function of bed height, derived from (a) and (b), respectively, are shown in (b) and (d), along with identification of the static and dynamic hold-up. The data in (a) and (c) are shown for the packing of 3 mm spheres for a liquid flow rate of 50 ml min^{-1} and gas flow rate of 250 ml min^{-1}. In (a) and (b), high signal intensity (white) identifies the presence of a liquid-saturated region, while low intensity (solid black) identifies entirely gas-filled locations. The various shades of grey identify the presence of both gas and liquid at any axial location in the bed.

Figure 2: Schematic of the bubble rise velocity measurement. The position of a bubble (black circle) moving upwards in the column is shown at times T0, T1, T2 and T3 in (a). (b) Stacked trace plot of the measured signal intensity at increasing times. The time sequence of 1D profiles of the bed is plotted as a 2D image, shown in (c). In (c), the gradient of the bubble 'track' is equal to its rise velocity in the bed. The *z*-dimension identified by the black arrows is the estimate of the height of the rising bubble.

Figure 3: Time series of profiles recorded using the ultra-fast gradient echo pulse sequence for the packing of 3 mm glass spheres. The gas- and liquid-flow rates were 20 and 50 ml min^{-1}, respectively. The grey scale shows the local gas hold-up; as in Figure 1, darker areas have higher gas content. In (a) the diagonal bubble tracks are used to calculate the bubble rise velocity. The diagonal lines highlight the leading edge of gas bubbles passing through the bed. (b) shows the measurement of the time taken, Δ*t*, for a whole bubble to pass through a given height, z.

Figure 4: Steps in identification of bubble dimension and aspect ratio using 2D RARE images. (a) Single x-z RARE image slice of the bed packed with 3 mm spheres with a gas and liquid flow rate of 50 ml min^{-1}. (b) Binary-gated image of (a); white corresponds to a value of 1 which is associated with the presence of liquid. (c) Binary-gated image of the liquid-saturated bed. (d) Map of gas-filled regions (white pixels) in the bed, obtained from subtraction of (b) from (a). The gated images (b-d) have been interpolated to a higher resolution for clarity.

Figure 5: Total gas hold-up as a function of (a) gas flow rate and (b) liquid flow rate. Data are shown for the fixed flow-rate being 50 ml min^{-1}, and are obtained from the MR pulse-acquire measurements. Data are shown for each of the 3 packing sizes.

Figure 6: Plots of the H_{gT}/β ratio as a function of the mean gas flow rate ratio (β). Data are obtained from the MR pulse-acquire experiments. Data are shown for each of the 3 packing sizes.

Figure 7: (a) Static gas fraction, and (b) dynamic gas fraction within the bed as a function of gas flow-rate ratio β. Data are obtained from the 1D velocity-compensated spin-echo profiles. Data are shown for each of the 3 packing sizes.

Figure 8: Bubble-size distributions for the 3 mm glass spheres. The results are for a liquid flow rate of 50 ml min^{-1}, and plotted for a range of gas flow rates. The distributions are plotted as the volume fraction of bubbles at a given bubble radius. Data are obtained from the 2D MRI experiments.

Figure 9: Bubble-size distributions for the 3 different sphere packings considered. Data are shown for a liquid and gas flow rate of 50 ml min^{-1}. Data are obtained from the 2D MRI experiments.

Figure 10: Plot of the bubble rise velocity for a bubbles of $r_z = 15$ mm, as function of (a) gas flow rate, and (b) liquid flow rate. Data are shown for the fixed flow-rate being 50 ml min^{-1}. Gas bubble rise velocities for 3, 5, and 1.8 mm packings are shown. Data are obtained from the 1D gradient-echo FLASH profiles.

Tables

Table 1: The fraction of static gas hold-up, F_{gS}, as a function of gas flow rate and packing size. The liquid flow rate is 50 ml min^{-1} in both cases.

packing diameter (mm)	F_{gS}	
	gas flow 50 ml min^{-1}	gas flow 250 ml min^{-1}
1.8	0.49±0.13	0.43±0.11
3.0	0.32±0.09	0.28±0.08
5.0	0.17±0.08	0.13±0.09

Table 2: Statistics of bubble-size distributions and how they are related to the void structure of the packed bed. Data are shown for gas and liquid flow rates of 50 ml min^{-1}, for each of the 3 packing sizes. The mean bubble radius ($\bar{r}$), and the volume fraction of bubbles (F) less than 5 mm, and greater than 10 mm in radius are given. The mean void radius characterising the inter-particle space of the bed ($\bar{r}_{void}$), the ratio of the mean bubble radius to the mean void radius ($\bar{r}/\bar{r}_{void}$), and the volume fraction of bubbles of size less than the mean void radius in the bed are also given. S is the volume-weighted interfacial surface area extracted from the 2D image data; i.e. an interfacial perimeter.

d (mm)	1.8	3.0	5.0
$\bar{r}$ (mm)	5.9	5.2	3.6
F(r<5 mm)	0.44	0.57	0.77
F(r>10 mm)	0.11	0.08	0.03
$\bar{r}_{void}$ (mm)	1.0	1.3	1.8
$\bar{r}/\bar{r}_{void}$	5.9	4.0	2.0
F($<\bar{r}_{void}$)	0.06	0.10	0.32
S (mm)	81	26	16