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Experimental investigation on the generic effects of gas permeation through flat vertical membranes

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ABSTRACT

This work reports the effects of gas extraction through flat vertical membranes on bubble dynamics in a fluidized bed. Bubble properties such as size, number, velocity and shape play a key role in the hydrodynamics and consequently heat and mass transfer characteristics of fluidized bed (membrane) reactors. Thus the main focus of this work is to understand the bubble behaviour over different fluidization velocities, particle sizes, gas extraction rates and gas extraction locations. A pseudo 2D experimental setup with flat vertical porous plates placed at the back of the column was used for simulating gas extraction through a flat vertical membrane in a fluidized bed reactor. A Digital Image Analysis (DIA) experimental technique was applied in order to extract the bubble properties.

Experimental results showed that the variation of gas extraction fraction has a minor effect on the bubble dynamics, with significant effects only present for high extraction rates and small particle sizes. Shifting the location of gas extraction more towards the centre of the bed had a larger influence on bubble dynamics. Deactivation of the two outmost membranes created a more uniform lateral bubble distribution profile which would be beneficial for reactor performance. However, deactivation of additional membranes caused the formation of central densified zones which obstructed the rising gas from reaching the central membranes. These effects could be clearly observed for small particles (196 µm), while larger particles (500 µm) showed little or no sensitivity to changes in gas extraction rate or location.

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1. Introduction

The use of fluidized bed membrane reactors (FBMRs) has gained a high degree of attention in the last few years, due to a number of industrially important applications and several significant advantages over conventional reactors. FBMRs offer the advantages of excellent separation properties of membranes (for example selective product removal), which circumvent the thermodynamic equilibrium limitations, and excellent heat and mass transfer characteristics of fluidized beds. These combined advantages have led to the development of highly energy efficient FBMR concepts. A typical example of a process using FBMR technology is selective removal of hydrogen from steam methane reforming using palladium-based membranes [1–10]. It was concluded that fluidized bed membrane reactors provide a better overall reactor performance compared to fluidized bed reactors without membranes and other conventional reactors.

Despite the proven advantages, FBMRs are still a relatively young field and substantial improvement in their performance can be achieved with better understanding of the effect of membrane insertion on the overall bed dynamic behaviour. In other words, detailed fundamental research in understanding the effect of the presence of membranes and the associated gas extraction is of high importance for exploiting the full potential of this technology. Moreover, it is well known that solid mixing, heat and mass transfer phenomena, and separation performance of gas-solid fluidized bed membrane reactors are highly dependent on the bubble properties and dynamics. The spatial distribution of bubbles, their shapes, sizes, numbers, and velocity play a key role in the hydrodynamics and thereby in the overall performance of the fluidized bed membrane reactors. Thus, a deeper understanding of bubble dynamics and prediction of their properties in such system is of great practical significance for process design and scale-up.

1.1. Studies on membrane fluidized bed hydrodynamics

A number of recent studies have paid more attention to the fundamental aspect of FBMR hydrodynamics. De Jong et al. [11] demonstrated

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Fig. 1. Process flow diagram of the experimental setup (all dimensions in mm) (the area coverage of active membranes decreases towards to the centre — from Via_7 to Via_1).

experimentally the effect of gas permeation through horizontal membranes on both solid and bubble phase properties. Results showed that the presence of horizontal membranes enhanced bubble breakage and resulted in a decrease in the average equivalent bubble size and increase in the number of bubbles, which provides an improvement in mass transfer between the bubble and emulsion phases. It was also reported that the largest effect was caused merely by the presence of the membranes tubes: both the solids flux and the average bubble size were decreased by a factor of three compared to a fluidized bed without inserts. The permeation rate was found to have a minor effect on the extent of solids circulation in the bed. Asegehegn et al. [12] and Julian et al. [13] also confirmed that only the presence of horizontally immersed tube banks in fluidized beds, without gas extraction, influences the bubble properties and behaviour. On the other hand, Medrano et al. [14] studied the hydrodynamics in the vicinities of horizontally inserted membranes in fluidized beds, and reported the formation of gas-pockets surrounding the membranes which might be detrimental for the performance of a fluidized bed membrane reactor. Deshmukh et al. [15–17] performed both numerical and experimental works on the effects of gas permeation via membranes and also found a reduction in both bubble sizes and solids motion. Tan et al. [18–20] also studied the effect of gas permeation through vertical membranes for small scale fluidized bed reactors and reported that the influence of gas permeation decreases with decreasing permeation rate or increasing membrane area.

Fewer studies have looked at the hydrodynamic behaviour in vertically inserted membrane fluidized bed reactors. De Jong et al. [21] and Dang et al. [22] have experimentally demonstrated that gas extraction through flat membranes (placed on the wall-sides of the bed) induces a change in both solid circulation patterns and average bubble size. It was also found that gas extraction through membranes resulted in a relatively larger average bubble diameter. This results from the fact that gas extraction causes densified zones formation near the membranes, which forces moving bubbles towards the center of the fluidized bed, thereby inducing bubble coalescence. This therefore results in bubbles that are vertically stretched and larger than in the case of no-gas extraction [21].

The present authors [23] studied the effect of vertical flat membranes on the bubble dynamics in a fluidized bed reactor both experimentally and numerically. Uniform gas extraction through the entire back of the pseudo-2D fluidized bed was found to cause densified zone formations on the sides, which force the gas to rise through the centre in the form of a channel. This phenomenon becomes more pronounced as the gas extraction rate is increased. On the other hand, variation of gas extraction location (area) showed a substantially larger impact on the bubble dynamics. For example, gas extraction through the centre was found to alter the bubble behaviour to rise in two channels towards the sides of the bed. Cold flow simulations completed using the Two Fluid Model approach (TFM) closed by the kinetic theory of granular flow successfully reproduced the experimental findings, including the effect of gas

Table 1

An overview of the experimental conditions investigated in this work where Via_i represents the area coverage of active membranes (decreasing towards the center, see Fig. 1).

Experimental campaign	Mean particle sizes [μm]	Extraction percentage [%]	U/U_{mf} [—]	Number of active membrane plates [—]	Number of double frame images
For the effects of gas extraction rate	196 and 500	0	3 and 6	0	2000
		10	3 and 6	14	2000
		20	3 and 6	14	2000
		30	3 and 6	14	2000
For the effects of gas extraction location	196 and 500	0	3 and 6	0	2000
		30	3 and 6	14 (Via_7)	2000
		30	3 and 6	10 (Via_5)	2000
		30	3 and 6	6 (Via_3)	2000
		30	3 and 6	2 (Via_1)	2000

Table 2

An overview of gas extraction velocities for different gas extraction rate through membranes.

Fluidization velocity	Mean particle size (μm)	Extr. percentage (%)	Extr. velocity (m/s)	Extr. percentage (%)	Extr. velocity (m/s)	Extr. percentage (%)	Extr. velocity (m/s)
$3U_{mf}$	196	10	0.0008	20	0.0016	30	0.0024
	500	10	0.0025	20	0.0050	30	0.0074
$6U_{mf}$	196	10	0.0016	20	0.0032	30	0.0048
	500	10	0.0050	20	0.0100	30	0.0145

extraction rate and gas extraction location on the bed dynamics and the formation of densified zones. Similar trends were observed in the simulation of reactive cases, simulating hydrogen production and extraction through membranes in a steam methane reforming process, but the effects of membrane extraction were marginally weaker. This result was attributed to the fact that in the reactive case, membrane permeability was proportional to the local hydrogen concentration and not constant at all membranes like in the cold flow studies.

This paper extends the previous work by the authors [23] on the effects of gas extraction rate and location through flat vertical membranes on bubble dynamics in a fluidized bed. Investigations were carried out over two different fluidization velocities; $3U_{mf}$ and $6U_{mf}$, as well as two particle size distributions; 180–212 μm and 400–600 μm . In the next sections a detailed description of the **Experimental setup** and methodology is presented followed by **Results and discussion**. Finally, a **Summary and conclusions** of the study will be presented.

2. Experimental setup and description

2.1. Experimental setup

This experimental study was performed in a pseudo-2D fluidized bed membrane reactor with dimensions of 1500, 300 and 15 mm in height,

width and depth, respectively. A schematic diagram of the setup is shown in Fig. 1. Air was used as a fluidizing medium and was fed to the fluidized bed through a porous plate distributor with a 20 μm mean pore size. The front wall of the reactor is made of transparent glass to allow optical accessibility for recording images. The back wall is made of black anodized aluminum to obtain better contrast between the emulsion phase and the background (bubble phase). A total of 14 membrane plates (porous plates with a mean pore size of 5 μm , 3 mm in thickness and 200 mm in height) arranged in two rows were constructed and mounted on the back of the column. Gas can be extracted in specific locations at the back of the column to investigate the effect of gas permeation through flat vertical membranes. The two rows of membranes were connected to low-pressure mass flow controllers and then to a vacuum pump which enabled specifying the required gas extraction rate. This experimental setup has been described in greater detail elsewhere [23].

Glass beads with particle size distributions of 180–212 μm and 400–600 μm and a density of 2500 kg/m^3 (both Geldart B type) were used as the fluidized medium. The minimum fluidization velocities for these two particle size distributions were determined experimentally using a standard pressure drop method as 0.07 m/s and 0.22 m/s respectively. The experimental results presented in this work have been carried out over two different fluidization velocities: $3U_{mf}$ and $6U_{mf}$. Images of the fluidized bed were recorded by a commercial high resolution CCD

Fig. 2. Equivalent bubble diameter and number of bubbles per frame as a function of axial position at different gas extraction fractions: a) $3U_{mf}$ – 196 μm (left), 500 μm (right), b) $6U_{mf}$ – 196 μm (left), 500 μm (right).

Fig. 3. Comparison of experimental data (without gas extraction) on bubble diameter along bed height with literature correlation for pseudo-2D fluidized beds [30].

camera (FlowSense EO 11 M Camera from Dantec operated with Dynamic-studio software), with a resolution of 4032×2688 pixels and a frame rate of 1.6 Hz (in double frame mode), which was placed in front of the column. 2000 double frame images (with a time step of 1.5 ms) were recorded and processed to determine bubble properties.

In this study, two experimental campaigns have been carried out. In the first campaign, the effects of gas extraction rate on the bubble dynamics were studied, whereas the second campaign investigated the effects of gas extraction location (area) on the bubble properties. An overview of the experiments completed in this study is shown in

Table 1. In both experimental series, the static bed height remained constant at 400 mm, which is the same as the total membrane height.

The selected gas extraction percentage, 10 to 30%, of the fluidizing gas flow rate are in the range of the actual membrane permeation rates that are feasible in hydrogen perm-selective membrane reactors. In the recent review paper by Gallucci et al. [24], a wide range (over almost two orders of magnitude) of permeation gas fluxes through hydrogen perm-selective membranes were reported. The highest membrane fluxes presented resulted in a gas extraction velocity of 0.036 m/s (this flux corresponds to a permeance of $1.5 \times 10^{-2} \text{ mol m}^{-2} \text{ s}^{-1} \text{ pa}^{-0.5}$) at 26 bar and 400 °C. The gas extraction velocities investigated in this work range from 0.0008 to 0.0145 m/s (see Table 2); falling within two orders of magnitude range spanned by membranes presented in the literature.

2.2. Experimental measurement techniques

Several measurement techniques have been applied to experimentally study the hydrodynamics in fluidized beds. These techniques have been commonly classified into two categories (intrusive and non-intrusive) based on the nature and position of the measuring sensors. The intrusive techniques such as resistance, inductance, impedance and thermal probes were extensively used; however, these probes alter the fluidization behaviour due to their intrusiveness [12]. Non-intrusive techniques offer better visual observation without interfering with the fluidization dynamics; these include electric capacity tomography, positron emission particle tracking, magnetic resonance tomography and particle image velocimetry [25–28]. However, these techniques suffer from poor spatial and temporal resolution and often do not provide details on both the bubble and emulsion phases [14].

Fig. 4. Typical images of the fluidized bed for different gas extraction fractions: a) no gas extraction, b) 10% gas extraction c) 20% gas extraction, and d) 30% gas extraction ($6U_{mf} = 196 \mu\text{m}$, top and bottom $6U_{mf} = 500 \mu\text{m}$).

Fig. 5. Equivalent bubble diameter and number of bubbles per frame as a function of lateral position at different gas extraction fractions: a) $3U_{mf} - 196 \mu m$ (left), $500 \mu m$ (right), b) $6U_{mf} - 196 \mu m$ (left), $500 \mu m$ (right). The lateral profile was averaged over the top half of the membranes (20–40 cm above the distributor).

In this study, a DIA technique is used to investigate the bubble dynamics. It should be noted that this experimental technique is limited to pseudo-2D fluidized beds due to the required optical accessibility [29]. DIA is an image post processing algorithm which uses the pixel intensity of an image to discriminate between bubble and emulsion phases. Each collected image is processed using an in-house script developed using the Matlab image processing toolbox, to correct inhomogeneous lighting and for removing background noise and shadow effects of the side walls. Subsequently, bubbles are detected based on a prescribed threshold value where all the bubble properties (such as equivalent bubble diameter, number of bubbles, bubble rise velocity and shape factor) in a set of images are determined. The bubble velocity is determined based on

processing two consecutive images with a known time step separating them. Further details about the DIA method are described elsewhere [11,23].

3. Results and discussion

3.1. Effects of permeation rate

The axial and lateral profiles of bubble size and number are shown in Fig. 2 and Fig. 5. The expected patterns are observed in all the plots. Fig. 2 shows a sharp decrease in bubble number and an associated increase in

Fig. 6. Bubble shape factor for the $6U_{mf} - 196 \mu m$ case as a function of axial position (left) and lateral position (right) for different gas extraction fractions. The lateral profile was averaged over the top half of the membranes (20–40 cm above the distributor).

Fig. 7. Average bubble rise velocity as a function of axial position at different gas extraction fractions: a) $3U_{mf}$ – 196 μm (left), 500 μm (right), b) $6U_{mf}$ – 196 μm (left), 500 μm (right).

bubble diameter as bubbles grow due to coalescence along the bed. The data in Fig. 5 show more and larger bubbles rising in the centre of the bed.

As expected, the bubbles developing in the 196 μm particle bed are significantly smaller than those developing in the 500 μm particle bed, at least for the $3U_{mf}$ fluidization velocity. For a fluidization velocity of $6U_{mf}$, however, the bubble sizes are similar. It is expected that bubble size increases both with particle size and fluidization velocity. This was the case when increasing particle diameter and fluidization velocity from the $3U_{mf}$ – 196 μm case, but not when increasing particle diameter and fluidization velocity to the $6U_{mf}$ – 500 μm case.

As shown in Fig. 3, these trends are contrary to the correlation proposed by Shen et al. [30] where an increase in particle diameter and fluidization velocity to the $6U_{mf}$ – 500 μm case caused a large increase in the bubble size. This large difference at large particle sizes and fluidization velocities can be attributed to the fact that Shen et al. [30] used a larger setup (680 mm $\times$ 70 mm, width and depth) than this work (300 mm $\times$ 15 mm), which allows for greater bubble growth along the bed height. In addition, Shen et al. [30] also reported that the proposed correlation over-predicts the average bubble size for larger particles at higher gas velocities and stated that this deviation may be attributed to gas through-flow.

The correlation proposed by Shen et al. [30] is given in Eq. (1):

$$D_b = 0.89[(U_0 - U_{mf})(h + 3.0A_0/t)^{2/3}g^{-1/3}] \quad (1)$$

where D_b is bubble diameter, U_0 is fluidization velocity, U_{mf} is minimum fluidization velocity, h is bed height, A_0 is area of distributor per orifice, t is depth of the bed.

The effect of gas extraction on the bubble characteristics proved to be minor. Only the $6U_{mf}$ – 196 μm case showed a clearly observable

effect. This is to be expected since the particle size is small and the gas extraction rate (product of the gas extraction fraction and fluidization rate) is large. The drag force on particles generally increases rapidly with a decrease in particle size, an increase in slip velocity and an increase in local particle volume fraction. For this reason, dense clusters of small particles in the case with the highest extraction rates led to a strong attraction of the particles to the porous plates on the back wall, creating clearly observable densified zones (as shown in Fig. 4). These densified zones altered the bubble dynamics and could have a significant detrimental effect on reactor performance.

Fig. 2 shows that, in the $6U_{mf}$ – 196 μm case, an increase in gas extraction fraction had the expected effect of decreasing bubble size and bubble number towards the top of the bed. The largest effects are expected at the top of the bed since the gas is extracted gradually along the bed height, implying that the specified fraction of gas is only completely removed right at the top of the simulated membrane column. Fig. 5 shows that the bubble diameter became significantly smaller in the centre of the bed for all extraction fractions, but the bubble number only decreased markedly for the case with the highest extraction fraction.

The bubble shape factors ($\frac{\pi \times \text{diameter}}{\text{perimeter}}$) for the $6U_{mf}$ – 196 μm case is reported in Fig. 6. A clear effect can only be observed for the case with the highest extraction rate where clear densified zones were formed in the upper half of the bed as can be seen in Fig. 4d. The larger shape factor in the centre of the bed is created by the densified zones obstructing the rising bubbles in this region of the bed. Otherwise, the trends are as expected: a decrease in shape factor along the height and towards the centre of the bed due to the presence of larger, more elongated bubbles.

Fig. 7 reports the bubble rise velocities in the bed. The expected increase in bubble rise velocity with an increase in particle size and fluidization velocity is observed. It is difficult to discern significant effects of the gas extraction fraction on the bubble rise velocity. The $6U_{mf}$ –

Fig. 8. Equivalent bubble diameter and number of bubbles per frame as a function of axial position at different gas extraction locations: a) $3U_{mf}$ – 196 μm (left), 500 μm (right), b) $6U_{mf}$ – 196 μm (left), 500 μm (right) (30% gas extraction fraction).

196 μm case is the only instance where an increase in gas extraction fraction caused a consistent reduction in the bubble rise velocity. This result links well to the overall decrease in bubble size found for the $6U_{mf}$ – 196 μm case (Figs. 2b, 5b).

It can therefore be concluded that gas permeation rates through flat vertical membranes investigated in this work only has a minor effect on bubble dynamics. However, the combination of highly permeable membranes and small particles could result in the formation of densified zones which significantly impact bed dynamics. For example, implementation of the most permeable membranes developed to date would result in one order of magnitude greater gas extraction velocities than those studied for the 196 μm case (for bed regions where high H_2 concentrations occur). This is likely to lead to significant densified

zone formation and should be avoided in reactor operation by, for example, using larger particle sizes.

One potential method to theoretically predict conditions under which densified zones may become a problem is to calculate the force with which the particles are dragged towards the membrane. This force can be estimated via the classical Ergun pressure drop equation [31].

$$\frac{dP}{dL} = \frac{150\mu}{d_p^2} \frac{(1-\varepsilon)^2}{\varepsilon^3} U + \frac{1.75\rho}{d_p} \frac{(1-\varepsilon)}{\varepsilon^3} U|U| \quad (2)$$

Here, dP/dL is the pressure drop per unit length, μ is the dynamic viscosity, d_p is the particle diameter, ε is the bed voidage, U is the superficial velocity and ρ is the gas density.

Fig. 9. Typical images of fluidized bed for different gas extraction locations: a) no gas extraction, b) Via_5 (gas extraction through 5 membrane-columns), c) Via_3 (gas extraction through 3 membrane-columns), d) Via_1 (gas extraction through 1 membrane-column) ($6U_{mf}$ – 196 μm).

Fig. 10. Equivalent bubble diameter and number of bubbles per frame as a function of lateral position at different gas extraction locations: a) $3U_{mf}$ – 196 μm (left), 500 μm (right), b) $6U_{mf}$ – 196 μm (left), 500 μm (right) (30% gas extraction fraction). The lateral profile was averaged over the top half of the membranes (20–40 cm above the distributor).

The Ergun equation given above estimates the force per unit volume acting on the particles. When normalized with the gravity force (F_g), it can be calculated that a densified zone with a void fraction of 0.4 will experience a drag force equivalent to $0.128F_g$ for the highest extraction rate with the 196 μm particles and $0.06F_g$ for the highest extraction rate with the 500 μm particles. Since densified zone formation had a significant influence in the 196 μm case and no clear influence on the 500 μm case, this appears to be the range where densified zone formation starts. Densified zone formation was also insignificant for the $3U_{mf}$ – 196 μm cases (maximum force of $0.064F_g$) and for the $6U_{mf}$ – 196 μm case with only 10% extraction ($0.043F_g$), but became visible at 20% extraction ($0.086F_g$) from visual observations as shown in Fig. 4.

This simple estimate suggests that densified zone formation may become a significant issue if membrane permeation rates are high enough to drag particles towards the membranes with a force in the order of $0.1F_g$. It is remarkable that a drag force one order of magnitude smaller than gravity can cause such a significant change in the hydrodynamics of the bed. Even under the assumption that the static friction coefficient between the densified zones and the membranes is equal to unity, the resulting friction force exerted by the membranes on the densified zone is an order of magnitude smaller than gravity.

Densified zone formation can therefore only occur if the densified zones are supported by a consistent upwards force exerted by the rising gas which is similar in magnitude to the downwards gravity force. If such a consistent upwards force was not present, the relatively weak friction force exerted by the membranes would not be able to temporarily hold the densified zones at the sides of the bed immobilized. Further work is required to refine this general guideline for the onset of densified zone formation in beds with gas extraction through flat vertical membranes.

3.2. Effect of gas extraction location

The natural tendency of gas to rise in the centre of the fluidized bed encourages the concentration of expensive membrane surface area only in this central region. For this reason, studies have been carried out to investigate the effect of concentrating gas extraction more towards the centre of the domain.

Axial bubble size and concentration profiles in Fig. 8 once again only shows a clear effect in the $6U_{mf}$ – 196 μm case. As discussed previously, the small particles and high extraction rates in this case enhanced the formation of densified zones due to the large drag force in the direction of the extracted gas acting on dense particle regions.

The effect of gas extraction location can more clearly be observed in the lateral profiles presented in Fig. 10, especially in the 196 μm cases. It is shown that the lateral profile in terms of bubble size and number becomes more uniform when removing the two membranes at the sides of the domain to extract from only 5 membranes. This should have a favourable effect on reactor performance by reducing overall mass transfer limitations and gas back-mixing.

Removing more membranes at the side of the domain, leaving only 3 or 1 active membranes in the centre of the domain, significantly changed the flow situation. In this case, unstable densified zones were formed in the centre of the bed and most of the gas was forced to rise in two channels on either side of this central obstruction (as can be seen in Fig. 9). As can be seen in Fig. 8 (for the $6U_{mf}$ – 196 μm case), this splitting of the flow caused an increase in the bubble number with a corresponding decrease in the bubble size in the upper regions of the bed. Such a situation would also be detrimental to reactor performance as a significant portion of the gas would be forced away from the central regions where the active membrane surface area is located.

Fig. 11. Bubble shape factor for the 196 μm particle size as a function of a) axial position and b) lateral position at different gas extraction locations for fluidization velocities of $3U_{mf}$ (left) and $6U_{mf}$ (right). The lateral profile was averaged over the top half of the membranes (20–40 cm above the distributor).

In the cases using the 500 μm particle size, the effect of membrane location had a small to negligible effect on bubble size and number. In this case, membranes can be safely concentrated towards the centre of the bed without forming the central densified zones observed for the 196 μm particles. This appears to be especially applicable to the $6U_{mf}$ – 500 μm case where the gas is strongly concentrated towards the centre of the domain.

Fig. 11 shows the shape factors for the 196 μm case where significant effects could be observed. In all cases, it can be observed that the cases with extraction in the centre of the domain (3 or 1 active membrane) showed a lower shape factor. This is due to the splitting of the rising bubble stream into two narrow streams on either side of the central densified zone region. The lateral profiles also show a much flatter profile due to more elongated bubbles rising towards the sides of the domain.

Finally, the axial velocity profiles in Fig. 12 only show a clearly discernible effect for the $6U_{mf}$ – 196 μm case. In this case, a change from 7 membranes to 5 membranes improved the lateral distribution of bubbles through the bed (as shown in Fig. 10), thus reducing the average bubble velocity. The significant change in flow dynamics caused by extraction only from 1 or 3 membranes did not have a significant further effect on the axial velocity profile.

4. Summary and conclusions

This work reported the effects of gas extraction rate and location through flat vertical membranes on bubble dynamics in a fluidized bed. Investigations were carried out over two different fluidization velocities; $3U_{mf}$ and $6U_{mf}$, as well as two particle sizes; 196 and 500 μm .

The gas extraction rate, expressed as the fraction of incoming gas extracted from the domain, only had a minor effect on the bubble dynamics in the bed. Significant effects were only observed in the case with the

smallest particle size and largest fluidization velocity (largest gas extraction velocity). In this case, the small particles experienced a large drag force towards the back wall of the domain where the gas was extracted, forming densified zones. Such densified zones formation forces the bubbles to rise towards the centre in a more stretched shape. This may have a significant negative effect on reactor performance, but can be avoided by using larger particle sizes and/or less permeable membranes. As an initial quantitative guideline, it was estimated that densified zone formation can become significant when the drag force towards the membrane exerted by the extracted gas reaches about 10% of the gravity force acting on the densified zone.

Shifting the location of gas extraction more towards the centre of the bed had a more significant influence on bubble dynamics, especially for the 196 μm particles. Removal of the two outmost membranes created a more uniform lateral bubble size distribution profile which would be beneficial for reactor performance. However, when more membranes were deactivated at the sides of the domain to strongly concentrate gas extraction in the centre, unstable densified zones were periodically formed in the centre of the domain, forcing a significant portion of the gas to rise in two narrow channels on either side of these densified zones. Such a situation would be detrimental to reactor performance since more gas would be forced to flow in regions where there is no membrane surface area.

In general, the strong effect of particle size should be further emphasized. The 500 μm particles consistently showed little to no sensitivity to changes in the gas extraction rate or location, whereas significant effects were observed for the 196 μm particles. It can therefore be safely assumed that the effects of extraction rates and location, mostly in the form of densified zone formation, would be substantially greater if particle sizes smaller than 196 μm were employed (the drag force towards the membranes at a given gas extraction rate increases quadratically with a decrease in the particle size). Highly permeable membranes

Fig. 12. Average bubble rise velocity as a function of axial position at different gas extraction locations: a) $3U_{mf}$ – 196 μm (left), 500 μm (right), b) $6U_{mf}$ – 196 μm (left), 500 μm (right).

may therefore require the use of larger than normal catalyst particle sizes in order to avoid densified zones and the associated mass transfer limitations. For perspective, the largest extraction rate investigated for the 500 μm particles corresponded to the most permeable membranes developed to date, whereas the largest extraction rate for the 196 μm particles was three times smaller. This large effect of particle size is therefore likely to become an important MAFBR design consideration as further research improves membrane performance.

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