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EXPERIMENTAL STUDY ON THE EFFECTS OF GAS PERMEATION THROUGH FLAT MEMBRANES ON THE HYDRODYNAMICS IN FLUIDIZED BEDS

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ABSTRACT

In this work, the effects of gas permeation through flat membranes on the hydrodynamics in a pseudo-2D membrane-assisted gas-solid fluidized bed have been investigated experimentally. A combination of the non-invasive Particle Image Velocimetry (PIV) and Digital Image Analysis (DIA) was employed to simultaneously investigate emulsion phase and bubble phase properties in great detail. Counter-intuitively, addition of secondary gas via the membranes, that constituted the confining walls of a gas-solid suspension at conditions close to incipient fluidization, did not result in a larger, but in a smaller bubble diameter, while gas extraction on the other hand, resulted in a larger equivalent bubble diameter, although in this case the effect was less pronounced. This could be explained by changes in the larger scale particle circulation patterns due to gas extraction and addition via the membranes: gas extraction leads to densely packed zones near the membranes, forcing bubbles through the center of the bed, where they become elongated and increase in size. Gas addition, on the other hand, totally inverts the particle circulation compared to a fluidized bed without membranes, splitting up bubbles in the center and forcing them towards the membranes, thus decreasing the bubble size.

INTRODUCTION

Fluidized bed membrane reactors combine the excellent separation properties of membranes with the advantages of fluidized beds. Moreover, the utilization of membranes enables to overcome reaction equilibrium limitations, thus resulting in higher reactant conversions and product yields. These clear advantages have led to an increasing number of applications of fluidized bed membrane reactors being proposed, for both product removal (e.g. hydrogen with palladium membranes) and reactant dosing (mostly oxygen) via membranes (Adris et al. (1); Mleczko et al. (2); Grace et al. (3); Gallucci et al. (4)). Despite the current developments, however, detailed understanding of the effect of the presence and permeation of gas through membranes immersed in a fluidized bed is lacking. The majority of current research relies on experimentally acquired data in experimental setups designed to provide a proof-of-concept and on phenomenological models, which often make use of ad-hoc empirical correlations that neglect the influence of internals.
Al-Sherehy et al. (5) investigated distributed feed and concluded that oxygen distribution is beneficial in expanding the range of reactant feed compositions beyond those normally allowed by safety constraints, while the selectivity was increased. Deshmukh et al. (6) confirmed these findings and, moreover, made great advances with respect to the effect of the presence of – and permeation through – the membranes on the extent of gas back mixing and the bubble-to-emulsion phase mass transfer rate. With ultrasound gas tracer experiments, they showed that due to the presence of membranes, but particularly due to gas permeation through the membranes, the macro-scale solids circulation was strongly reduced, resulting in a near plug-flow behavior for the gas phase. They also found smaller average bubble diameters for higher permeation ratios relative to the total gas flow. Christensen et al. (7) confirmed that such systems indeed lead to a decrease in bubble size and bubble hold-up, and therefore to an increase in the total number of bubbles.

This paper aims to advance the fundamental understanding by investigating experimentally the effect of a change in gas flow rate inside a fluidized bed membrane reactor, where gas is added or extracted through the side-walls of the fluidized bed. Therefore, we focus specifically on bubble formation/annihilation close to the membranes, bubble size distribution and particle mixing as a function of the gas permeation ratio, i.e. the ratio of gas added/extracted relative to the total feed. After a description of the experimental setup and the procedures used for data post-processing, we discuss and compare the PIV/DIA results for cases of gas extraction and gas addition.

EXPERIMENTAL SETUP

A pseudo-2D setup 30cm in width, 1.5cm in depth and 1m in height has been constructed. For the front of the bed a glass plate is used, for the back an anodized aluminum plate to provide good contrast between emulsion phase and background. The distributor is a porous plate with a mean pore size of 40 μm. At both sides of the fluidized bed, up to a height of 30cm above the distributor, gas can be added to or extracted from the fluidized bed through a 10 μm porous plate. For all experiments, glass beads with a particle size distribution of 400-600 μm and a density of 2500 kg/m³ (Geldart B) have been used. Air has been used as a fluidization agent. The

Figure 1: Process flow diagram of the experimental setup
process flow diagram is given in Figure 1. The minimum fluidization velocity \( U_{mf} \) was determined to be 0.25 m/s by slowly decreasing the fluidization velocity. All experiments reported here have been performed with a total gas feed corresponding to 2.6 \( U/U_{mf} \) (see Table 1). A lower velocity would significantly de-fluidize the bed during gas extraction experiments, while a much higher gas velocity is not possible with the current setup. The number of images has been determined to be sufficient for obtaining reliable time-averaged results; the error in the vector plots presented in this paper is below 6%. The error in the equivalent bubble diameter depends on the number of bubbles detected at a certain height in the bed, and ranges from <1% at a height of 2.5 cm to 3% at a height of 45 cm.

PIV/DIA Procedure

PIV is a non-intrusive optical technique based on the comparison of two images recorded with a very small time delay (here 0.82-1.98 ms) with a high speed CCD camera. It divides every image into interrogation zones (here 32x32 pixels were used), and uses a special cross-correlation on two consecutive images to obtain an average displacement of the particles in that interrogation zone. These PIV image pairs were post-processed using the commercial software package DaVis.

DIA is an image post-processing algorithm, that discriminates bubble and emulsion phase based on the pixel intensity. Prior to the actual bubble detection, the algorithm corrects for the camera lens effect, inhomogeneous lighting and shadow effects near the walls. For every measurement series at least 10 random images were inspected visually, to ensure that the script is functioning correctly. Only by using the combination of PIV and DIA, it is possible to determine the time-averaged emulsion phase velocity profiles from the obtained instantaneous particle velocity profiles and correct for particle raining through the bubbles to avoid under-estimation of the particle fluxes in the centre of the bed (Laverman et al. (8)).

<table>
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<tr>
<th>Measurement name</th>
<th>Background gas flow / velocity</th>
<th>Total membrane flow / velocity</th>
<th>Number of images</th>
</tr>
</thead>
<tbody>
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<td>Total membrane flow / velocity</td>
<td>For DIA [-]</td>
</tr>
<tr>
<td>Reference</td>
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<td>0 [%-] 0 [m/s]</td>
<td>2700</td>
</tr>
<tr>
<td>100% + 20%</td>
<td>100 [%-] 0.65 [m/s]</td>
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<td>2700</td>
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<tr>
<td>100% + 40%</td>
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<tr>
<td>100% - 20%</td>
<td>100 [%-] 0.65 [m/s]</td>
<td>-20 [%-] 0.065 [m/s]</td>
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<tr>
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<td>100 [%-] 0.65 [m/s]</td>
<td>-40 [%-] 0.130 [m/s]</td>
<td>1350</td>
</tr>
</tbody>
</table>

RESULTS & DISCUSSION

The discussion on the experimental results on the effect of gas permeation on the hydrodynamics of the fluidized bed is started by first focusing on the solids circulation patterns, followed by the bubble properties.

Emulsion Phase Circulation Patterns

The time averaged solids circulation pattern and the time-averaged lateral profile of the axial emulsion phase velocity at different heights in the bed is shown in Figure 2. In the reference series without secondary gas extraction or addition, the characteristic pattern for fluidized beds with an upwards directed solids flow through
Figure 2: Time-averaged particle movement and time-averaged lateral profile of the axial emulsion phase velocity for different heights in the fluidized bed with (a) 40% gas extraction, (b) 20% gas extraction, (c) the reference series with no gas addition/extraction, (d) 20% gas addition and (e) 40% gas addition.

The core and a downward solids flow along the walls of the fluidized bed can be clearly discerned. This well-known solids circulation pattern is also clearly visible in the lateral profiles of the axial emulsion phase velocity at different heights: a broad region in which particles move upwards in the center of the fluidized bed, and near the walls a small region where the particles move downwards. It is interesting to notice the local minimum in the axial emulsion phase velocity in the centre at 10 cm above the bottom distributor plate, corresponding to the well-known average bubble trajectories from the walls towards the centre in the lower sections of the fluidized bed (see also Laverman et al. (8)).

When comparing the cases with gas extraction to the reference case, a striking difference is the stagnant regions near the membranes in case of gas extraction. It is already appearing in the 100%-20% case, but becomes even more pronounced when 40% of the background fluidization gas is extracted. These stagnant zones have two consequences: the first consequence is that the bed height is reduced, implying a smaller number of bubbles or smaller bubbles inside the fluidized bed. Secondly, the velocity plot shows that the peak of upward moving solids has become steeper, while the downward directed ‘peak’ for the downward moving solids has become less pronounced and has shifted somewhat towards the center of the bed. The reason for these phenomena is that the stagnant zones near the...
membranes leave less space for bubbles to rise and for particles to re-circulate to the bottom of the bed, resulting in narrower vortices in the solids circulation.

In contrast to gas extraction, gas addition via the membranes has an even more distinctive effect on the particle circulation pattern: gas addition inverts the circulation pattern. This phenomenon shows that there is a competition between the background gas velocity and the additional gas entering via the membranes to drag the particles along. Already in the 100%+20% series this phenomenon starts to become apparent, but is even more pronounced for the 100%+40% series. Usually particles would move downwards near the walls. However, due to the gas addition, particles near the wall (in the first 30 cm) are dragged upwards instead, causing the particles to move downwards in the center of the bed. This phenomenon is also illustrated by the lowest three lateral profiles of the axial emulsion phase velocity profiles; the upwards directed peak is now near the wall, while the velocity in the center of the bed is slightly negative. Above the membrane (above 30 cm), the particles are pushed towards the center, and continue their way as usual: upwards via the center and downwards via the sides. This division in a part with membrane and a part without membrane results in four vortices inside the fluidized bed, each one rotating differently than its neighbor.

The findings described above can be schematically summarized as depicted in Figure 3. In all cases, the magnitude of the effects depends on the background fluidization velocity and amount of gas extraction or gas addition. It can be expected that the change in particle behavior has a pronounced effect on the bubble properties and bubble size distribution, which is discussed next.

![Figure 3: Illustration of the particle circulation patterns for (a) gas extraction, (b) the reference and (c) gas addition.](image)

**Bubble Properties**

Firstly, the obtained experimental results were validated by comparison with literature; both the equivalent bubble diameter, as well as the bubble rise velocity compared well to the corresponding literature correlations (not shown here). Subsequently, the equivalent bubble diameter as a function of the axial position in the fluidized bed is shown in Figure 4.a. In the lower part of the fluidized bed, the bubbles remain approximately the same size, irrespective of the amount of gas extraction or addition. Only from a height of approximately 20 cm, a difference becomes apparent. However, unlike what would be expected intuitively, extracting gas leads to larger bubbles, while adding gas results in smaller bubbles.
Figure 4: Effect of gas extraction and addition on (a) equivalent bubble diameter as a function of the bed height, (b) average number of bubbles per frame as a function of the bed height, (c) average bubble diameter as a function of the lateral position and (d) bubble hold-up as function of the bed height.

In particular the experimental series in which gas is added via the membranes deviate substantially from the reference case above a height of 30 cm. Note that the largest bubbles for the cases of gas extraction appear at 40 cm height, the ones for the reference case at about 46 cm, and the bubbles for the cases with gas addition appear even at 52 cm height, reflecting the difference in fluidized bed height.

Figure 4.b shows a slight difference in the average number of bubbles present in every frame. For gas addition, it can be concluded that there are more bubbles (Figure 4.b) with a smaller diameter (see Figure 4.a). For gas extraction, the number of bubbles is decreased, but they have a larger equivalent diameter. However, the difference in the number of bubbles is less important in comparison with the difference in bubble diameter. The bubble rise velocity as a function of the equivalent bubble diameter (not shown here) is quite similar for all cases. The graphs of the lateral profile of the equivalent bubble diameter and the axial profile of the bubble hold-up (Figure 4.c and 4.d) provide more insight into the bubble behavior. There is a significant difference in the average lateral position of the bubbles. The reference case shows an almost parabolic distribution, as expected, because bubbles are formed over the entire width of the fluidized bed and move towards the center due to bubble coalescence. The 100%-20% and 100%-40% series show a similar distribution, although bubbles are situated more in the center (which is in line with the conclusions drawn from Figure 2). The 100%+20% and 100%+40% series reveal a very different bubble distribution: in these cases the large bubbles are situated much closer to the walls. In the center, a significant decrease in bubble diameter is visible, indicating that the movement of the bubbles
is reversed, i.e. while bubbles are rising and growing, they are moving away from the center and towards the membranes. This is in line with the particle movement seen in Figure 3.

Not only the location, but also the bubble volume is different or these cases. The series with gas extraction show a slightly larger bubble volume, although this difference is very small. However, the series with gas addition reveal that – in particular in the top section of the bed – the bubble volume is much smaller compared to the reference case. Now the question remains why for the 100%+20 and 100%+40 series, both the average bubble diameter as well as the average bubble volume are lower than the reference case. This phenomenon is caused by a combination of particle movement and bubble detection: large gas voids near the walls are likely to be part of the freeboard of the fluidized bed, and are therefore no longer defined as bubbles. This is caused by particles near the freeboard that are – in contrast to the reference case – moving away from the wall toward the center of the fluidized bed, and as a consequence, there are much fewer large bubbles surrounded by emulsion phase. The effect of gas extraction and gas addition on the bubble behavior is schematically depicted in Figure 5.

![Figure 5: Illustration of the bubble size distribution and movement for (a) gas extraction, (b) the reference and (c) gas addition.](image)

**CONCLUSIONS**

A pseudo-2D experimental fluidized bed setup with membranes (porous plates) at both the left and right side has been constructed to investigate the effect of gas extraction or gas addition on the emulsion and bubble phase behavior in detail. A combination of Particle Image Velocimetry (PIV) and Digital Image Analysis (DIA) was employed.

The experimental results revealed that gas addition via the membranes counter-intuitively leads to significantly smaller bubbles, whereas gas extraction slightly increases the bubble size. During gas addition, the bubble size in the top of the bed decreased to 60% of the original bubble size. During gas extraction, a small increase in bubble size was found (an increase in bubble size of 10% and 20% was observed relative to the reference case for 20% and 40% gas extraction respectively). The explanation was found in the lateral bubble distribution and particle circulation patterns. During gas addition, the bubbles are split up and distributed towards both membranes. The particle circulation therefore inverts, and particles move upwards with the bubbles via the sides, and downward through the
center of the bed. During gas extraction, on the other hand, stagnant zones near the membranes emerge. These zones force upwards moving bubbles and particles, as well as downwards moving particles towards the center of the fluidized bed, which results in bubbles that are vertically stretched and therefore slightly larger than in the reference case. The experimental findings have shown a large effect of gas extraction or addition on the fluidized bed hydrodynamics, which should be properly taken into account in the optimization and design of membrane-assisted fluidized bed reactors.

It would be interesting to validate the conclusions from this work in 3D systems, but these systems require different measuring techniques. In the near future, we will compare the obtained results to numerical simulations with a Euler-Euler model. Furthermore, the hydrodynamics in a fluidized bed with a membrane configuration consisting of horizontal membrane tubes instead of vertical membranes will be investigated both numerically as well as experimentally in order to derive improved design rules for future fluidized bed membrane reactors.

ACKNOWLEDGEMENT

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NOTATION

\[ U \] gas velocity [m/s]
\[ U_{mf} \] minimum fluidization velocity [m/s]

REFERENCES


