Detecting densified zone formation in membrane-assisted fluidized bed reactors through pressure measurements

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Highlights
- Effects of gas extraction on the extent of densified zones are presented in detail.
- Extent of densified zones increases with increasing gas extraction rate.
- Maximum variance in pressure drop shifted to lower velocity as gas extraction rises.
- Smaller particles showed major effect to changes in extraction rate.

Abstract
This work reports the results of an experimental investigation on densified zone formation in a membrane fluidized bed reactor using combined pressure fluctuation and PIV (Particle Image Velocimetry) measurements. A pseudo 2D experimental setup was used, where porous plates on the back plate of column mimicked gas extraction through flat vertically inserted membranes. The maximum in the standard deviation of pressure fluctuations, commonly employed to indicate the transition to turbulent fluidization, shifted to lower fluidization velocities with an increase in the fraction of fluidizing gas being extracted. Flow visualization showed that this result is connected to the onset of stable densified zone formation, which occurred at progressively lower fluidization velocities as the gas extraction fraction was increased. It has also been found that the extent of densified zones quantified using instantaneous particle velocity maps collected by PIV increases with increasing gas extraction rates. This effect became larger for smaller particles. Results have therefore shown that the peak in pressure fluctuations in fluidized beds with gas extraction through flat vertical membranes indicates the onset of densified zones formation rather than turbulent fluidization. Such densified zones can have substantial detrimental effects (such as induced mass transfer limitations, gas bypass etc.) on the reactor performance and can be identified via pressure measurements, as illustrated in this work.

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1. Introduction

Membrane Assisted Fluidized Bed Reactors (MAFBRs) have recently emerged as one of the most promising technologies for ultrapure hydrogen production with integrated carbon dioxide
capture [1]. These reactors integrate the catalytic reactions, mostly reforming and water-gas shift reactions for hydrogen generation, and separation through membranes in a single process unit [2–9]. This combination of process units brings a high degree of process intensification with additional benefits in terms of increased process efficiencies. However, the performance of MAFBRs is strongly influenced by the operating conditions of the fluidized bed reactor [10], which can range from bubbling to turbulent to fast fluidization. For instance, turbulent fluidization is often the preferred regime, both for commercial catalytic reactors [10] and MAFBRs [11].

On the other hand, it is very important to prevent the formation of defluidized regions in the bed, which can affect the quality of fluidization and reduce the permeation rates through the membranes. Previous studies [12–14] have shown that defluidized regions close to membrane plates can be induced due to gas extraction via membranes in MAFBRs. These defluidized regions could substantially reduce heat and mass transfer characteristics, and consequently hamper the overall reactor performance.

A typical example of a process using a membrane assisted fluidized bed technology is hydrogen production through steam methane reforming [15]. Clearly, defluidized zone formation will negatively affect the process performance as the hydrogen permeation rates through the membranes and bubble-to-emulsion mass transfer limitations were described as the two main limiting factors for hydrogen production [3,5,6,16]. It is therefore of high importance to detect and determine the onset of densified zone formation in membrane assisted fluidized bed reactors, which is the main focus of this work.

1.1. Regime transition in dense fluidized beds

It is widely accepted that dense fluidized beds operate under bubbling and turbulent fluidization regimes. The transition between these two regimes occurs at a superficial gas velocity which corresponds to the maximum of the standard deviation of the pressure signal measured in the bed. Yerushalmi et al. [17] was the first to propose this technique to quantitatively determine the transition from bubbling to turbulent fluidization. Other measurement techniques were also reported in several studies published over the last three decades, such as visual observations, capacitance signals, optical fibre probes, local and overall bed expansion (reviewed in reference [18]).

In the bubbling regime, bubble coalescence/break-up dominates. The amplitude of pressure fluctuations increases with the bubble size as the superficial velocity increases [19–22]. The transition of fluidization regime occurs when bubbles reach their maximum stable size and start breaking to smaller bubbles as the fluidization velocity is further increased. This well-known behaviour of dense fluidized beds is likely to be affected by gas extraction through membranes which was shown to cause formation of densified zones and alter the bubble dynamics in the bed [14].

This study will shed light on densified zone formation in membrane assisted fluidized beds through a dedicated experimental campaign combining pressure fluctuation measurements, flow visualization and particle velocity measurements using particle image velocimetry (PIV). The focus is on understanding the mechanism of formation, in addition to quantitatively characterize the extent of densified zone formation and its relation to gas extraction through the membranes. Furthermore, the effect of gas extraction through vertical membranes on the regime transition in a membrane fluidized bed will be investigated and discussed. The well-established pressure fluctuation method will be used to determine the critical regime transition velocity in a membrane fluidized bed. Flow visualization and PIV (for 180–212 μm particle size distribution) will be used to study the change in the fluidization regime towards the densified zone as detected by the standard deviation of pressure fluctuations, thus validating this method.

In the next sections a detailed description of the experimental setup and methods are presented. Then, the effects of gas extraction (gas extraction fraction and membrane area) through vertical membranes on the apparent critical velocity (so called transition velocity from bubbling to turbulent) will be discussed. Subsequently, a criterion to identify the densified zones formation and a method to quantitatively estimate the extent of densified zones using the PIV technique will be developed. An analysis of the extent of densified zones as a function of the gas extraction fraction will follow. Next, an analysis of the recalculated apparent critical velocity after taking into account the densified zones is presented. The paper then concludes with a summary and conclusions.

1.2. Densified zone detection in fluidized bed reactors

A defluidized zone can form due to several reasons, such as a sudden change in feed gas composition [23], a strong decrease in fluidization velocity, an increased pressure caused by downstream problems [24] or gas extraction through membranes [14]. Early detection of this phenomenon in an industrial process is of high importance for improved process performance, high product quality and cost saving by avoiding suboptimal process operation and unplanned shutdowns. Several measurement tools have been employed to monitor the defluidized bed behaviour and to specifically detect this phenomenon. The most straightforward and simplest one is the average pressure drop over parts of the bed [24]. However, this technique only shows a strong change when the bed is completely defluidized. Other measurement techniques such as capacitance probes, optical instruments, heat transfer probes and electrodes for measuring triboelectric currents have also been reported in the literature, but they have the drawbacks of only detecting small local defluidized regions and requiring multiple measurement points [25]. In contrast measurement techniques based on pressure fluctuations, such as chaos analysis of the bed pressure drop fluctuations [26] and pressure fluctuation measurements, offer a larger detection volume, are insensitive to small changes in gas velocity, can handle multiple signals and can detect a change in fluidized bed hydrodynamics [24]. Moreover, the pressure fluctuation measurement technique includes the effect of many different dynamic phenomena happening in the bed (bubble formation and breakage). In this measurement method, one measurement point could be sufficient; however, two or three measurement positions are always recommended (to increase the reliability of the detection and in case of sensor malfunctioning) [27].

2. Methodology

2.1. Experimental setup

The experiments were carried out using a pseudo-2D fluidized bed column with 1.5 m height, 0.3 m width and a depth of 0.015 m. A schematic flow diagram of the setup is shown in Fig. 1. The front wall was made of transparent glass to allow optical access (for image recording), while the back wall was made of black anodized aluminum (to obtain a good contrast between the particles and the gas phase). In order to investigate the effect of gas extraction a multi-chamber area with porous plates (membranes) on the front was constructed and mounted at the bottom of the back plate. The chambers are placed close to each other to maximize membrane surface area per unit bed volume. The porous plates used to mimic vertical membrane insertion were 3 mm in thickness with a mean pore size of 5 μm which is the lowest
available porosity to obtain high pressure drop and circumvent possible gas by-pass. Fourteen individual porous plates were arranged in two rows (each 200 mm in height) and seven columns spanning the width of the bed (details of membrane configuration is shown in Fig. 2). The distributor at the bottom of the bed was made of porous metal with an average pore size of 20 µm and 3 mm thickness. Three different glass beads with particle size distribution of 70–110 µm, 180–212 µm and 400–600 µm (with density of 2500 kg.m⁻³, Geldart B classification) were used. The minimum fluidization velocity of 0.027 m/s, 0.070 m/s, and 0.22 m/s was measured by standard pressure drop measurement method for the three different particle sizes respectively.

Air was used as fluidizing gas and the flow rate was controlled by two digital mass flow controllers (Bronkhorst), with a range up to 400 L/min and 250 L/min, depending on the particle size investigated. In order to reduce electrostatic forces between the wall and particles that may be generated during fluidization, the air was humidified to 50–70% relative humidity by passing the feed through a water column which was mounted online with the gas feed. The two rows of porous plates representing the membranes (7 each) mounted at the bottom of the back plate were connected to two low-pressure mass flow controllers (Bronkhorst) and in turn connected to a vacuum pump (with a capacity up to 60 m³/h) for gas extraction. The required flow extraction rate is controlled by the low pressure mass flow controller. Images for solid velocity measurements were recorded by a high resolution CCD camera (Flow sense EO 11 M Camera from Dantec operated with Dynamic-studio software), with a resolution of 4032 x 2688 pixels and a frame rate of 1.6 Hz, which was placed in front of the column. 1000 double frame images (with a time step of 0.75 ms) were recorded and processed to determine the time averaged velocity profile.

2.2. Experimental techniques

2.2.1. Pressure fluctuation

The pressure fluctuation measurement technique is the most common and widely used method to analyse the transition velocity of flow regimes in gas-fluidized beds. Commonly, two main arrangements have been used in pressure fluctuation measurements [24]: (1) a single-point absolute pressure fluctuation (APF) and (2) double-point differential pressure fluctuation (DPF).
absolute pressure fluctuation technique refers to the measurement of pressure fluctuations at a single location across the bed, and the differential pressure fluctuation measures a pressure difference between two nearby points across the bed (axial direction). The absolute pressure fluctuation provides information on both local and global hydrodynamic behaviour, whereas, fluctuation in the pressure difference only reflect the local hydrodynamic behaviour between the two measurement locations [24]. In this work the APF method has been selected as they reflect more global phenomena and are less influenced by the axial location of the pressure transducer [24,28].

In this study, three pressure transducers (S-11 model, with a range of 10 psi) from WIKA were mounted flush with the inner side-wall of the column at 10 cm height intervals from 20 cm above the distributor (Fig. 1) (Since all of them nearly revealed similar results, only the results at the axial location of 0.3 m is presented). The openings of the probes were covered with a filter of 40 μm meshes (lower than half of the smallest particle size used) to prevent the glass-beads entering into the probes and block the probes tip. Additionally, to further minimize the risk of filter blockage a back-flush of air through the filter was applied regularly (after every experiment). The outputs from the transducers were connected with a data acquisition box. The signals of the transducers were then recorded with a sampling frequency of 50 Hz for a period of 180 s.

2.2.2. Particle Image Velocimetry (PIV)

Time averaged particle velocity profile is one of the methods that have been used to illustrate the densified zone formation as a function of gas extraction fractions through vertical membranes. In this work, PIV and Digital Image Analysis (DIA) techniques were applied. PIV is a non-intrusive optical measurement technique based on the comparison of two consecutive images recorded within a very short time step with a high speed CCD camera. In order to obtain the time averaged emulsion phase velocity, each image is divided into interrogation areas, where cross correlation on two consecutive images is applied. The PIV image pairs are post-processed using the commercial software package DynamicStudio by Dantec. A multi-pass algorithm using an interrogation area of 64 × 64 pixels with 50% overlap was applied to reconstruct the corresponding particle velocity vector maps. However, before the images were post-processed on PIV for the time-averaged velocity analysis, the large particle velocities associated with particle-raining through the bubbles were filtered out [29]. The DIA technique was applied for filtering out the particle velocities inside bubbles. DIA is an image post-processing algorithm which uses pixel intensity to discriminate between the bubble and emulsion phases. If the pixel intensity is below a threshold value, the pixel area is assigned to the bubble phase, if not to the emulsion phase. Then the images are again imported to Dynamic-studio and processed to obtain instantaneous and time averaged emulsion velocity maps.

3. Results and discussion

In order to investigate the effect of gas extraction, two experimental campaigns have been carried out. In the first experimental campaign, the effects of variation of gas extraction through the membranes on the apparent transition velocity and the extent of densified zone formations were studied. Three different extraction fractions from the total inlet flow rate were investigated: 0.1, 0.2 and 0.3 (see Table 1). The standard deviation of the pressure signal was determined and plotted against the equivalent superficial gas velocity for the studied gas extraction fractions (the equivalent superficial velocity is the velocity calculated based on the gas flow rate at the outlet of the fluidized bed). A reference case with no gas extraction was also completed for comparison. Three different particle sizes were investigated: 70–110, 180–212 and 400–600 μm.

In the second experimental campaign, the effect of extraction location (membrane surface area) on the apparent transition velocity was studied. An extraction fraction of 0.2 was selected, while the extraction configuration (the total number of active membrane plates) was varied. The extraction location was varied from the largest area (all 7 membrane columns-A1) to the smallest area (only a single membrane column at the centre A-4) (see Fig. 2).

In both experimental campaigns, the static bed height was kept identical to the total membrane height (40 cm). An example of experimental series is shown in Table 1, presenting gas extraction fractions, the superficial gas flow rate (inflow), and the outlet flow rate (outflow) (as of Eq. (1)). The inflow indicates the amount of gas fed through the bottom distributor for fluidization, while the outflow indicates the amount of gas leaving the fluidized bed (similar to the reference case without gas extraction). The difference between the in-and out flow indicates the amount of gas extracted via membranes. Thus, the amount of gas extraction varies with different gas extraction fractions (i.e. 0.1, 0.2 and 0.3), and outlet superficial gas flow rates. The inlet superficial gas flow rate is adjusted from case to case according to the amount of gas extracted in order to maintain the outlet superficial gas flow rate equal to the reference case.

In flow = \frac{Out\ flow}{1 - a}

(1)

where \( a \) is gas extraction fraction (0 ≤ a ≤ 1).

3.1. Data processing

The absolute pressure fluctuation intensity in the membrane assisted fluidized bed was measured in the domain of time with 50 Hz frequency. The standard deviation of a given pressure fluctuation signal \( p(t) \) is determined mathematically as follows:

\[
\sigma_p = \sqrt{\frac{1}{N-1} \sum_{i=1}^{N} (p(t) - \bar{p})^2}
\]

(2)

where

\[
\bar{p} = \frac{1}{N} \sum_{i=1}^{N} p(t)
\]

(3)

\( p(t) \) is time series of pressure fluctuation signal, \( \bar{p} \) is mean of \( p(t) \), and \( N \) is number of data points.

The standard deviation of pressure fluctuations has been widely accepted as a measure of bubble behaviour in fluidized beds and has been extensively used to demarcate flow regime transitions. In the next sections of this paper, the standard deviation of the pressure signal as a function of superficial gas velocity in a membrane assisted fluidized bed is discussed in detail under different operational conditions.
An example of the pressure signals collected at three different positions in the bed is shown in Fig. 3. Fig. 3a shows the typical spectrum of absolute pressure fluctuation (without gas extraction) as a function of time (180 s) measured at the fluidization velocity of 0.85 m/s which is close to the transition velocity. The similarity between the pressure signals from the different measurement positions is more clearly visible in Fig. 3b in which only shows a time interval of 10 s.

To better quantify the similarity between the results from the three different measurement positions, Fig. 3c and d show the corresponding standard deviation as a function of superficial velocity without and with gas extraction at the three different locations. As can be seen from the figure that the point of the maximum standard deviation in the absolute pressure fluctuation (critical velocity) at different locations from the distributor reveal similar results. Only one pressure measurement position (at a height of 0.3 m) can therefore safely be used for the remainder of the study.

3.2. Flow regime transition velocity

Before the detailed discussion on the effect of gas extraction through the membranes on the apparent critical (onset) velocity, a comparison of the flow regime transition velocity of the reference case (without gas extraction) for each particle size with some literature data is presented. Fig. 4a shows the standard deviation of the absolute pressure fluctuations as a function of the fluidization velocity (without gas extraction) for the three different particle sizes as measured by a pressure probe located at the axial position of 0.3 m. The influence of particle sizes has been widely studied by many authors [10,28,30–35] and the experimental result of this work is in line with their prediction showing that bigger particles have a higher transition velocity. The measured critical velocities were then compared with different correlations found in the literature. Fig. 4b shows the relation between the Reynolds number at the critical transition velocity, \(Re_c\), and the Archimedes number. The correlation equations used for this comparison are shown below:

\[
\text{Bi and Grace (APF data) [18]} \quad Re_c = 0.565Ar^{0.461}
\]
\[
\text{Leu and Lan (APF data) [30]} \quad Re_c = 0.568Ar^{0.578}
\]
\[
\text{Yang and Leu (APF data) [31]} \quad Re_c = 0.837Ar^{0.487}
\]

\(Re_c\) obtained from this study (for the three particle sizes used) showed a very good agreement with the correlation of Bi et al. [18], whereas the other correlations, Yang and Leu [36], Leu and Lan [35], over-predicts the critical velocity \(Re_c\) found in this study. Moreover, it is worth mentioning that the reactor size also plays an important role in the transition velocity, as the size may affect the bubble size and rise velocity, which in-turn influences the transition velocity. The influence of the reactor size is more pronounced for smaller bed size but insensitive for larger beds.
The transition velocity decreases with decreasing column width and diameter for smaller beds [11,31].

3.3. Influence of gas extraction

Fig. 5 shows the standard deviation of the absolute pressure fluctuations (APF) at the bed height of 0.3 m with a particle size distribution of 180–212 µm as a function of the superficial gas velocity for (a) different gas extraction fractions and (b) extraction locations through the vertical membranes. It can be seen that, for all the cases, the standard deviation in the pressure fluctuation shows a clear peak. However, the position of this peak shifts to lower superficial gas velocities as the gas extraction fraction is increased. The difference in the measured critical velocities is substantial, varying between 0.88, 0.82, 0.41, and 0.32 m/s for 0, 0.1, 0.2 and 0.3 extraction fractions respectively. The observed shift in the critical transition velocity caused by gas extraction through membranes does not indicate a shift in the transition from the bubbling to the turbulent regime, but rather the establishment of a regime characterized by the formation of densified zones. This transition to the densified zone regime is illustrated in Fig. 6, which shows snapshots presenting typical bed structures in the densified regime ($U_g = 1.5U_c$) for a particle size distribution of 180–212 µm. Increasing densified zone formation is clearly visible as the gas extraction fraction is increased from 0 to 0.3. The densified zone formation can be attributed to the drag force applied by the gas extracted through the membranes on the particles which are consequently dragged towards the membrane wall. The higher the gas extraction, the larger the densified zones formed. When densified zones are established, a large part of the fluidizing gas slips through a relatively stable channel in the centre of the bed, which explains the global decrease in the standard deviation of the absolute pressure fluctuations as shown in Fig. 5.

On the other hand, the effect of gas extraction location on the maximum standard deviation in the absolute pressure fluctuations becomes noticeable only when the extraction takes place from large areas (Fig. 5b). The transition velocity decreased by around 50 % when all 7 or 5 membrane columns were used, whereas extraction from 1 or 3 membrane columns caused a very limited decrease in the transition velocity. It is therefore clear that only extraction from relatively large areas can cause the formation of sufficiently large densified zones to completely alter the bed dynamics.

The sudden change in bed behaviour when changing from extraction through 3 membrane columns to 5 membrane columns is related to the position of the densified zone formation [14]. When extracting through 5 or 7 membrane columns, densified zones naturally form at the walls, while the gas channels through the centre. This is a relatively stable flow situation since the gas naturally tends to rise in the centre of the bed and the densified zones are stabilized by the walls. The result is a fairly stable...
channelling of gas between relatively stable densified zones at the walls which limits the amount of pressure fluctuation in the bed. When extracting through 1 or 3 membrane columns, on the other hand, the densified zones are forced to form in the centre of the column. This is an inherently unstable flow situation where the densified zones are obstructing the natural central channelling of the bed and are not stabilized by the side walls. The result is repetitive formation and destruction of densified zones, which maintains a high degree of pressure fluctuation in the bed.

The magnitude of the shift in the transition velocity caused by gas extraction through the membranes was found to increase sharply with a decrease in particle size (Fig. 7). This effect can be better
visualized by plotting both the ratio of the transition velocity to the minimum fluidization velocity for each particle size and the excess gas velocity \((U_c - U_{mf})\) against the gas extraction fraction. As can be seen in Fig. 7d, the transition velocity is highly sensitive to the gas extraction for the 70–110 \(\mu\)m particles, but almost insensitive to the gas extraction for 400–600 \(\mu\)m particles. Similar trend is observed when the excess gas velocity is plotted as function of gas extraction fraction (see Fig. 8). These results can be explained by noting that the drag force increases with decreasing particle size. Small particles are therefore more easily dragged towards the wall by the extracted gas through membranes, leading to easy formation of densified zones.

### 3.4. Densified zone formation

The effect of gas extraction through membranes on the bubble and solids behaviour has been studied both experimentally and numerically in previous studies [12–14]. It has been shown that both bubble and solids behaviour are altered by gas extraction through membranes, which was found to have a direct impact on the reactor performance [14]. In this study, a further investigation on the formation of densified zones due to gas extraction through membranes has been carried out. The purpose of this study is to validate the pressure fluctuation technique as a useful tool to detect the transition velocity towards densified zone formation using PIV measurements.

A simple example of the detection of densified zones through particle velocity measurements is shown in Fig. 9. The lateral profile of the solids velocity shows the standard pattern with solids rising in the centre and falling at the walls when the fluidization velocity is less than the transition velocity. At the transition velocity and beyond, however, the formation of stable densified zones at the side walls reduces the downwards flow at the wall and causes stable channelling in the centre of the bed (visible in Fig. 6). This more stable flow situation will result in a reduction in the pressure fluctuations measured in the bed.

In addition, a quantitative analysis of the effect of gas extraction through vertical membranes on the extent of densified zones has been performed. The fraction of bed area covered by densified zones as a function of the fluidization velocity for different gas extraction fractions has been determined to quantify the extent of densified zones. A criterion was set on the particle velocity for identifying the densified zones as follows:

\[-\gamma \leq v_i \leq \gamma\]  \hspace{1cm} (7)

where \(v_i\) is the local instantaneous particle velocity and \(\gamma\) is the threshold value (0.08 m/s). The value of the threshold parameter was chosen based on some sample histogram profiles, derived from a sensitivity analysis, in which only positive particle velocities are shown (Fig. 10). The threshold of 0.08 m/s was selected based on the histograms (Fig. 10), especially the ones at \(U_c\) and \(1.5U_c\) where the presence of densified zones was more pronounced; a clear change in the histogram of instantaneous particle velocities can be seen corresponds to a velocity of 0.08 m/s. The same threshold was used for the reference case without gas extraction, which...
showed almost constant extent of densified zone over different fluidization velocities. A Matlab script was developed to process instantaneous particle velocity maps identifying the areas of the bed with low particle velocities meeting this criterion. The identified areas are summed up to calculate the total area, $a_{\text{dens}}$, covered by densified zones in each instantaneous particle velocity map (it should be noted that the velocities of bubble areas were excluded from this procedure). The time averaged ratio of the area of densified zones to the total area of the fluidized bed is then defined as the extent of densified zones.

$$\frac{A_{\text{densified zones}}}{A_{\text{static bed}}} = \frac{\sum a_{\text{dens}}}{N_t \cdot h \cdot d} \times 100\%$$

where $N_t$ is the total number of images, $h$ is the height of the bed under static conditions (from the distributor to end of membrane height), and $d$ is the width of the bed.

Fig. 11 shows the extent of densified zones calculated for the different superficial gas velocities centred around the critical transition velocity ($\pm 60\%$ of $U_c$ and $U_c + 1.06U_{mf}$) as determined by the standard deviation of pressure fluctuations (six superficial gas velocities were taken as shown in Table 2). The results in Fig. 11a are shown for a case with 0.3 gas extraction fraction and are compared to the reference case without gas extraction. The plots of the extent of densified zones are superimposed onto the plots of standard deviation of the absolute pressure as a function of superficial fluidization velocity. The extent of densified zones remained low and insensitive to the fluidization velocity for the reference case without gas extraction. Bed areas satisfying the criterion in Eq. (7) in this case do not represent stable densified zones, but rather areas which are momentarily at low velocities due to the transient nature of the bed dynamics. When 0.3 gas extraction fraction was applied, the extent of densified zones increased from around 20% at low fluidization velocities (bubbling regime) to 45% for fluidization velocities beyond the transition velocity. A sharp increase in the extent of densified zones is observed at the transition velocity indicating a step change in the fluidization dynamics from bubbling fluidization to stable densified zones with central gas channeling. A step change in the extent of densified zones can also be observed at low fluidization velocities. This may indicate the initial formation of unstable densified zones which dynamically form and break up. These densified zones do not fundamentally alter the bed behaviour.

The data in Fig. 11b depicts that, as the gas extraction rate increases, the extent of densified zones increases while shifting the sharp transition to the densified zone regime towards lower superficial gas velocities. The largest step change in the extent of densified zones always corresponds to the transition velocity identified via pressure measurements. Thus, it is clear that gas extraction through membranes plays a key role in the formation of densified zones and that the transition to a stable densified zone regime occurs suddenly. This sudden regime change can be reliably detected as a peak in the standard deviation of the absolute pressure as highlighted in the previous section.

Fig. 12a gives a clear indication of the increase in the extent of densified zones as a function of the gas extraction fraction at the corresponding transition velocity. This data can be used to recalculate the fluidization velocity by excluding the area covered by the densified zones (assuming that no gas flows through the densified zones); i.e. the total area of identified densified zone was subtracted from the total area of the bed under static conditions to recalculate the apparent gas superficial velocity (from the newly apparent fluidization area). In other words, the stable densified zones forming at the side walls of the domain are considered as impermeable obstructions, causing the gas to accelerate through the narrowed central channel created by these densified zones. This procedure showed that the recalculated transition velocities determined with gas extraction through membranes gets close to the reference case (without gas extraction) when the densified areas are excluded in the analysis of the critical velocity. This indicates that the gas superficial velocity in the stable central channel between densified zones is sufficiently high to achieve turbulent fluidization.
4. Summary and conclusions

This work investigated densified zone formation in flat membrane fluidized bed reactors. A pseudo 2D experimental fluidized bed setup was used, equipped with a multi-chamber area with porous plates on the front mounted at the bottom of the back plate of column, in order to mimic vertically inserted membranes. Gas can be extracted in specific areas through the porous plates thus allowing for investigating the effect of gas extraction rates and location on the fluidization behaviour. The fluidized bed column has a glass plate on the front to allow for visual access and enable application of non-invasive visual techniques to study the bed hydrodynamics.

Gas extraction through membranes was found to shift the maximum standard deviation in the pressure fluctuations (standard indicator of a transition to turbulent fluidization) to lower fluidization velocities. This shift was found to occur when gas is extracted from large membrane areas (from 5 or all 7 available membranes). Extraction from a large membrane area caused the formation of stable densified zones at the walls gas channelling through the centre. This flow situation would have a significant detrimental effect on the reactor performance by inducing significant mass transfer limitations. Use of smaller particle sizes further amplified this effect since smaller particles experience a stronger drag force towards the membrane plates. When gas was extracted from the centre of the bed (1 or 3 central membrane columns), this effect was not observed as stable densified zones could not form in the centre of the bed.

Flow visualization confirmed that the shift in the transition velocity is due to densified zone formation caused by gas extraction through the membranes. The maximum in the pressure fluctuations corresponded to a sudden increase in the extent of densified zone formation quantified via particle image velocimetry, indicating the sudden formation of stable densified zones at the side walls of the bed. This flow situation should be avoided in membrane assisted fluidized bed reactors and can be detected by determining the critical transition velocity and comparing it to the (theoretical) turbulent transition velocity. If the measured transition velocity is substantially lower than the turbulent transition velocity, densified zone formation may occur with consequent detrimental effects on the reactor performance.

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