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## The effect of gas addition on bubble dynamics in a fluidized bed with flat vertical membranes



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#### HIGHLIGHTS

- Gas addition through vertical flat membranes into a dense fluidized bed.
- · Reduced bubble size and velocity, combined with increased generation of small bubbles.
- Enhanced gas-solids contact through reduced gas channelling through the centre.
- Improvements up to 44% in the bubble-to-emulsion gas transfer coefficient.

### ARTICLE INFO

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### ABSTRACT

This study investigates the hydrodynamics of a fluidized bed with flat vertical membranes for selective species dosing. A custom designed pseudo-2D column, equipped with a multi-chamber area on the back for gas permeation, was used to emulate a membrane assisted fluidized bed (MA-FB). The effect of global and local gas additions on bubble properties, solids hold up, solids circulation patterns and regime transition was investigated using Digital Image Analysis, Particle Image Velocimetry and pressure fluctuation measurements. Gas addition throughout the entire permeable area substantially reduced bubble size and velocity, while increasing the number of bubbles. In addition, a significant reduction in gas channelling through the centre of the column was observed. Concentrating gas addition towards the sides of the column further enhances bubble distribution over the lateral direction and totally removes gas channelling. Gas addition also caused an earlier transition to turbulent fluidization. The estimated bubble-to-emulsion mass transfer rate could be doubled when 30% gas addition was applied in the turbulent regime. These benefits maximise the potential of process intensification for fluidized bed reactor concepts using selective dosing through flat vertical membranes.

### 1. Introduction

Membrane assisted fluidized beds (MA-FBs) are attracting increasing research attention for application to a variety of industrial catalytic processes. They combine a high level of process integration and excellent heat transfer characteristics to result in highly intensified chemical processes [1]. Furthermore, fluidized beds eliminate issues associated to species concentration polarization [2] and heat management encountered in fixed bed reactors [1], while membranes overcome reaction equilibrium limitation in catalytic processes to maximize their performance [3].

Dehydrogenation processes are typical examples that use membranes for selective species recovery [3]. These processes involve endothermic catalytic reactions that require high operating temperatures

to maximize reactant conversion, but such high temperatures also favour side reactions that reduce product selectivity [1]. In this category, hydrogen production from natural gas reforming has received the most attention [3] after it was proposed and patented by Adris et al. [4,5]. Insertion of  $\rm H_2$  perm-selective membranes into a fluidized bed results in methane conversion well above equilibrium by integrating the reforming, water-gas-shift and hydrogen separation steps into one single process [3,6,7]. Substantial research efforts have been dedicated to this process combining modelling [8–11] and experimental demonstration activities at scales ranging from lab [12] to pilot [7,13] scales. The expected benefits have been successfully demonstrated. This concept can also be combined with chemical looping reforming [14] or gas switching reforming [15] to produce pure hydrogen with integrated  $\rm CO_2$  capture.

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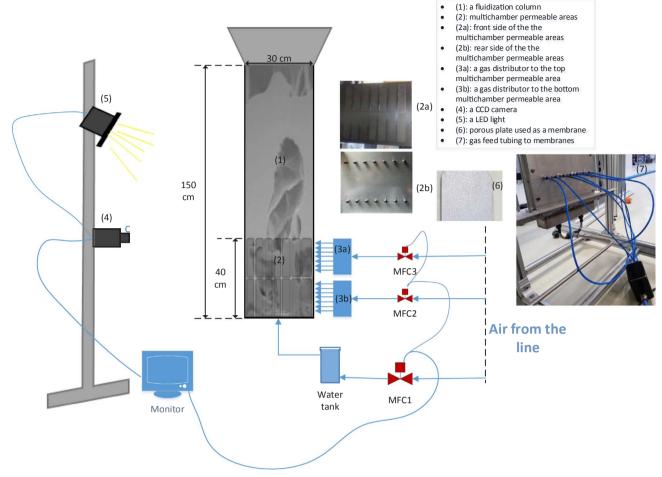


Fig. 1. A simplified scheme of the experimental setup.

Selective species dosing in catalytic processes presents another promising application of MA-FB technology. Particularly, membranes for oxygen addition were applied to a wide range of chemical processes, mainly in oxidative reforming of hydrocarbons. This includes partial oxidation of methane [16], oxidative dehydrogenation of ethane [17], oxidative coupling of methane [18], and conversion of methanol to formaldehyde [19]. Further applications are found in Fischer-Tropsch GTL technology using membranes for oxygen/hydrogen addition [20,21], and the conversion of butane to maleic anhydride [22]. The most important benefits of using MA-FBs with species dosing include: higher conversion and productivity, a broader favourable operating range with respect to the oxygen-hydrocarbon ratio, expansion of the range of reactant feed compositions beyond those normally allowed by safety constraints, enhanced selectivity to desired products, and autothermal operation through heat and process integration [1,23–25].

Successful design and operation of MA-FB reactors requires a proper understanding of the hydrodynamics in these systems. The hydrodynamics affect gas-solid contact and bubble properties, which have a direct impact on species transport and heat transfer within the bed. Additionally, a good understanding of the hydrodynamics of these systems can reduce capital costs by optimizing membrane positioning in the bed to maximize overall process performance at minimal membrane surface area. For example, when a gas species is to be recovered from the reactor, membranes should be concentrated in the regions where most of gas bubbles rise.

In that respect, previous studies in the literature were dedicated to understanding the hydrodynamic effect of, not only the physical presence of membranes in the bed [26,27], but also gas permeation through the membranes [28–31]. Horizontally immersed obstructions

with or without gas extraction were found to alter the solids flux and reduce the bubble size by enhancing bubble breakage [32]. However, gas pockets could form and attach to the horizontally immersed membranes, thus creating an additional mass transfer resistance that could negatively affect reactor performance [33]. On the other hand, gas extraction through vertically inserted flat membranes was found to result in densified zone formation and central gas channelling, which could negatively affect reactor behaviour in a similar manner [29,31]. These densified zones were detected by the early transition on the pressure fluctuation amplitude when gas extraction through membranes was applied [31].

Despite the growing interest in using MA-FB reactors for selective species dosing applications, less attention has been paid to investigating the effect of gas addition on the bed behaviour. Similarly to gas extraction, the existence of gas pockets in the vicinity of the membrane was also reported for the case of gas addition through horizontally inserted cylindrical membranes [32]. This seems to be linked more to the existence of obstructions rather than gas permeation through them [33]. Gas addition through flat membranes placed on the sides of a pseudo-2D fluidized bed was found to result in significantly smaller bubbles compared to a standard dense fluidized bed [29]. This was attributed to the improved gas distribution through the membranes along the height of the bed, but also to inversion of solids circulation patterns [29].

This study aims at giving further insights about the effect of gas addition through flat membranes. In contrast to the study of De Jong et al. [29] that looked at the effect of gas addition through flat membranes placed on the sides of a pseudo 2D setup, this study uses flat membranes covering the entire back area of the bed and allows for

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