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Micro-structured fluidized bed membrane reactors: Solids circulation and densified zones distribution

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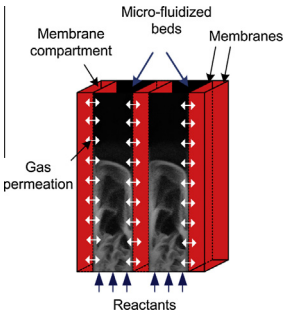
HIGHLIGHTS

- The hydrodynamics of a novel membrane assisted micro-structured fluidized bed (MAmFB) is studied in this work.
- Particle Image Velocimetry (PIV) and Digital Image Analysis (DIA) techniques have been used for non invasive study.
- The turbulent fluidization regime with a relatively low gas extraction velocity reduce the densified zones.
- The micro-structured reactors can be used with state-of-the-art membranes with reduced densified zone formation.

GRAPHICAL ABSTRACT



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ABSTRACT

The present paper reports an experimental investigation on the hydrodynamics of a novel membrane assisted micro-structured fluidized bed (MAmFB) operated in bubbling or turbulent flow regimes. The effects of gas addition and gas extraction through flat porous membranes confining the fluidized bed on the bubble size distribution, solids holdup distribution and solid circulation patterns have been evaluated by the combination of two non-invasive techniques, viz. Particle Image Velocimetry (PIV) and Digital Image Analysis (DIA). The experimental results show that the micro-structured fluidized bed membrane reactor improves the solid circulation compared with bigger size membrane reactors where the extraction of gas result in parts of the bed that are completely defluidized and stagnant at the membrane walls. However, also in case of small reactors great care has to be paid to the gas extraction velocity relative to the fluidization velocity. All the results indicate that the amount of densified zones (zones where solids lumps with a local solids hold-up close to the minimum solids packing have a much lower velocity than the rest of the emulsion phase) can be reduced drastically by working in the turbulent fluidization regime with a relatively low gas extraction velocity. This study indicates that actual state-of-the-art membranes can be used in the turbulent regime without the formation of densified zones, thus avoiding additional mass transfer resistances (concentration polarization).

1. Introduction

Membrane-assisted fluidized bed reactors have recently been proposed and studied and have been shown to be efficient alternatives to more conventional reactor systems such as packed bed membrane reactors for a variety of reaction systems, among which are methane reforming and autothermal reforming and ethanol reforming [1–6]. The different applications of high temperature membrane reactors can typically be classified in two main categories: (i) dehydrogenations where one of the products (hydrogen) is selectively extracted through the membranes, and

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Nomenclature

APF	Absolute Pressure Fluctuation
DIA	Digital Image Analysis
DEM	Discrete Element Model
PIV	Particle Image Velocimetry
MAmFB	Membrane Assisted micro-Fluidized Bed
MAFB	Membrane Assisted Fluidized Bed
A	total area of a bubble (mm^2)
d	averaged-bubble diameter (mm)
F, G	parameters for Eq. (1) fitted to DPM simulation results
Hmbr	membrane length (mm)
ε^{2D}	two dimensional solids fraction (–)
ε^{3D}	three dimensional solids volume fraction (–)
U	superficial gas velocity ($m\ s^{-1}$)
Δt	delay time between two image pair (μs)
$\varepsilon_p(x,t)$	instantaneous local solid hold-up obtained from DIA
2D	two dimensional solids hold-up distribution
3D	volume solid hold-up distribution

(ii) partial oxidations where air (or oxygen) is distributively fed through porous or selective membranes.

The reasons why fluidized bed membrane reactors can outperform packed bed membrane reactors are essentially [7,8]:

- Negligible pressure drop, which allows using small particle sizes avoiding internal mass and heat transfer limitations.
- Strongly reduced bed-to-membrane mass transfer limitations (referred to as concentration polarization) [9].
- (Virtually) Isothermal operation.
- Flexibility in installing membrane and heat transfer surface area and arrangement of the membrane bundles.
- Improved fluidization behavior as a result of:
  - Compartmentalization, i.e. reduced axial gas back-mixing.
  - Reduced average bubble size due to enhanced bubble breakage, resulting in improved bubble-to-emulsion mass transfer.

As fluidized bed membrane reactors circumvent the external mass transfer limitations that adversely affect the performance of packed bed membrane reactors to a large extent, the volumetric production capacity in fluidized bed membrane reactors is limited by the relatively low permeation rate through the membranes (provided that the catalytic activity is sufficiently high).

There are two ways to improve the membrane permeation, namely (i) decreasing the thickness of the membranes and thereby increasing the membrane permeability and (ii) increasing the number of membranes installed per unit volume of the reactor.

Decreasing the thickness of the membranes to increase the permeation flux has a clear limitation. For instance for Pd-based hydrogen perm-selective membranes the lower limit of the thickness for stable membranes has apparently been reached as membranes as thin as 0.1–1 μm are nowadays available on the market [8]. It is not foreseen that thinner membranes can be produced without compromising the membrane perm-selectivity or stability (lifetime). For oxygen selective membranes there is always a compromise between flux and stability. On the other hand, de Jong et al. [10,11] have experimentally demonstrated that also in fluidized bed membrane reactors using membranes with an enormously high permeation flux densified zones close to the membranes will create which would most probably induce strong bed-to-membrane mass transfer limitations.

The second option (installing more membranes per unit of volume) seems to be the only reasonable way to overcome flux limitations. Installing more membrane area (thus more membranes) will drastically reduce the space between membranes where the catalyst is suspended in fluidization. For instance using planar membranes [12] close to each other would result in a small compartment that can be seen as "micro-structured" fluidized bed membrane reactor as theoretically studied by Wang et al. [13]. Their simulation study has also elucidated that in these small confinements the turbulent fluidization regime (with anticipated improved mass transfer characteristics) can be achieved at lower superficial gas velocities.

Although these novel micro-structured fluidized bed membrane reactors seem to answer all the requirements for the perfect intensified reactor, a few important questions still remain unanswered. Is it indeed possible to overcome all the external mass transfer limitations in these small compartments even in case a large amount of gas is extracted through the membranes? Is it possible to operate the small reactors in the turbulent regime in order to circumvent the detrimental formation of densified zones observed previously in large scale reactors [10]? And finally, is it possible to operate these reactors with state-of-the-art membranes without inducing mass transfer limitations (due to densified zones)?

To answer these questions and give guidelines for the design and operation of fluidized bed membrane reactors, in this work a detailed experimental investigation has been carried out on the hydrodynamics of small membrane-assisted fluidized beds operated in both the bubbling and the turbulent fluidization regimes where the gas is added or extracted through two opposed vertical membranes confining the fluidized suspension. First the experimental setup and techniques are shortly described.

2. Experimental methods and setup

In this work the hydrodynamics of a small membrane assisted fluidized bed reactor has been studied with a combination of Particle Image Velocimetry (PIV) and Digital Image Analysis (DIA). This technique has been selected for two main reasons: (i) PIV/DIA is currently the only technique able to give detailed while-field information on both gas (bubble) phase and emulsion phase with high spatial and temporal resolution [14–18] which allows the determination of solid flux profiles and bubble properties simultaneously; (ii) it is a non-intrusive technique; the fluidized bed is very small and only a small amount of gas is fed into/extracted from the fluidized bed, thus any probe inserted in the bed would irremediably disturb the fluid dynamics of the bed. The downside of the selected experimental technique is that visual access is required and thus only pseudo 2D beds can be studied effectively. For comparison, some other available experimental techniques are listed in Table 1. The table is of course not complete as many more techniques are available and each technique has many advantages and disadvantages. An interested reader is referred to the more comprehensive review by Werther [19]. The quantitative extrapolation of the phenomena observed in the 2D beds to real 3D fluidized beds is not trivial. However, the results help to gain a better understanding and to validate numerical models, with which 3D beds can be investigated.

In this work, in order to determine the 3D-particle volume fraction from the 2D (image intensity) pictures, a correlation was developed using artificial images created from Discrete Element