

## Fluidized bed reactor assisted by oxygen transport membranes: numerical simulation and experimental hydrodynamic study.

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### Highlights

- Application of oxygen transport membrane (OTM) technology.
- Hydrodynamic cold model study of a fluidized bed reactor with internals.
- Simulation of methane oxy-combustion and calcium carbonate calcination.

### 1. Introduction

In the near future, hydrogen is bound to become an important energy carrier used for surplus power storage and as a fuel in stationary applications and for sustainable transportation. Hydrogenation of CO<sub>2</sub> is also considered a viable perspective to mitigate the greenhouse effect. Steam-methane reforming (SMR) is a mature technology to produce H<sub>2</sub> in large quantities. New processes include sorption enhanced steam methane reforming (SE - SMR), that is a combination of the SMR process and sorption of CO<sub>2</sub> on a solid sorbent, in one step. Biomass gasification could also play an important role in the production of a H<sub>2</sub>-rich syngas; with the aim of maximizing the H<sub>2</sub> content in the product gas, a CO<sub>2</sub> sorbent can be used in the gasification chamber to capture CO<sub>2</sub> as soon as it is produced. The removal of CO<sub>2</sub> shifts the thermodynamic equilibrium of the water gas shift (WGS) reaction to the hydrogen product. The gas obtained, rich in hydrogen, could be exploited in high efficiency power generation devices like GT (gas turbine) or SOFC (solid oxide fuel cell). One of the most efficient gasification technologies is a dual fluidized bed system, with circulation of bed material between two different reactor chambers, separating gasification and char combustion reactions. When an Ion Transport Membrane (ITM) system is used to supply pure oxygen [1] to the combustor and limestone is added in the bed material, a solid circulation loop is realized that allows to increase the hydrogen content of the gasifier product gas (T=650-750°C) and to obtain an almost pure carbon dioxide stream at the combustor exit (T=800-900°C). This process is studied at pilot scale in the ZECOMIX experimental platform run by ENEA in the Casaccia research centre.

In this work, the interest is focused on the feasibility of the Calcium Looping (CaL) cycle, more specifically on the combustor/calciner fluidized bed reactor, which is of interest for all applications mentioned above. The endothermic calcination reaction occurs utilizing the thermal energy generated from a combustion process between CH<sub>4</sub> and O<sub>2</sub> supplied by a bundle of perovskite membranes housed in the fluidized bed calciner.

### 2. Methods

The work is divided into two parts. First, a hydrodynamic study is presented, carried out with the cold model of a cylindrical bubbling fluidized bed reactor with internals (vertical rods mimicking the OTM system). Cold model studies are widely used to study fluidization quality and to support the design of complex fluidized bed systems, as the calciner unit in ZECOMIX experimental platform. Matching a set of properly chosen dimensionless numbers allows to evaluate the hydrodynamic behavior of the hot pilot plant by means of a fluidized system contained in a Plexiglas column at ambient conditions obeying geometric and dynamic similarity rules.

The knowledge of system behavior acquired in this way will be then combined with the chemical reaction kinetics of methane combustion and calcium carbonate calcination (the reverse process of carbon dioxide capture), as well as the oxygen transport rate into the calciner through the perovskite membrane system, to

develop a two - phase reactor model (bubble and gas-solid emulsion) integrated in MATLAB®. In this way, a reliable investigation of a membrane-assisted fluidized bed reactor is made possible, where simultaneous oxy-methane combustion [2] and calcination reaction [3,4] take place. The flow of the gas is considered as plug flow through the bubble phase and axial dispersion plug flow through the emulsion phase. On the other hand, the particulate solid contained in the emulsion phase (dolomite) is considered perfectly mixed.

### 3. Results and discussion

The vertical tube bundle housed inside the cold fluidized bed, forces bubbles to travel upwards inside the pitch delimited by adjacent rods, reducing their size considerably in relation to a similar bed without internals, as Figure 1 shows clearly. In the operative conditions of the hot calciner ( $u = 17 u_{mf}$ ), the bed expansion is smaller than the configuration without the membrane module inside the experimental apparatus. It is possible to explain this result by assuming that in the presence of the module, bubbles rise faster along the membrane tubes that represent a kind of preferential path to the bed surface. It is obtained that, when the cold model operates with  $u = 17 u_{mf}$ , the bed expands from 153 (static bed height) to 244 mm. Applying the dynamic similarity to the calciner, the bed expands from 500 (static bed height) to 706 mm in this case.

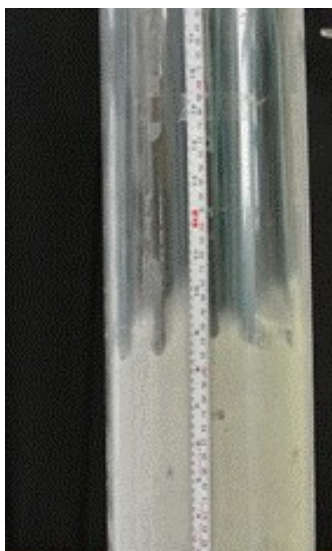


Figure 1. Bed expansion in a cold fluidized bed with internals ( $u = 10 u_{mf}$ ).

### 4. Conclusions

An innovative membrane-assisted fluidized bed for regeneration of a calcium based sorbent in a CaL sorption enhanced cycle is investigated. A hydrodynamic study is performed under ambient conditions by means of a cold model to simulate fluidized bed expansion and to evaluate bubbles behavior in the presence of vertical membrane tubes for oxygen transfer. A numerical model of the hot bubbling fluidized bed calciner with OTM system is developed and integrated in a MATLAB® routine.

### References

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### Keywords

“Membrane fluidized bed, bubbles with internals, calciner modelling, OTM”