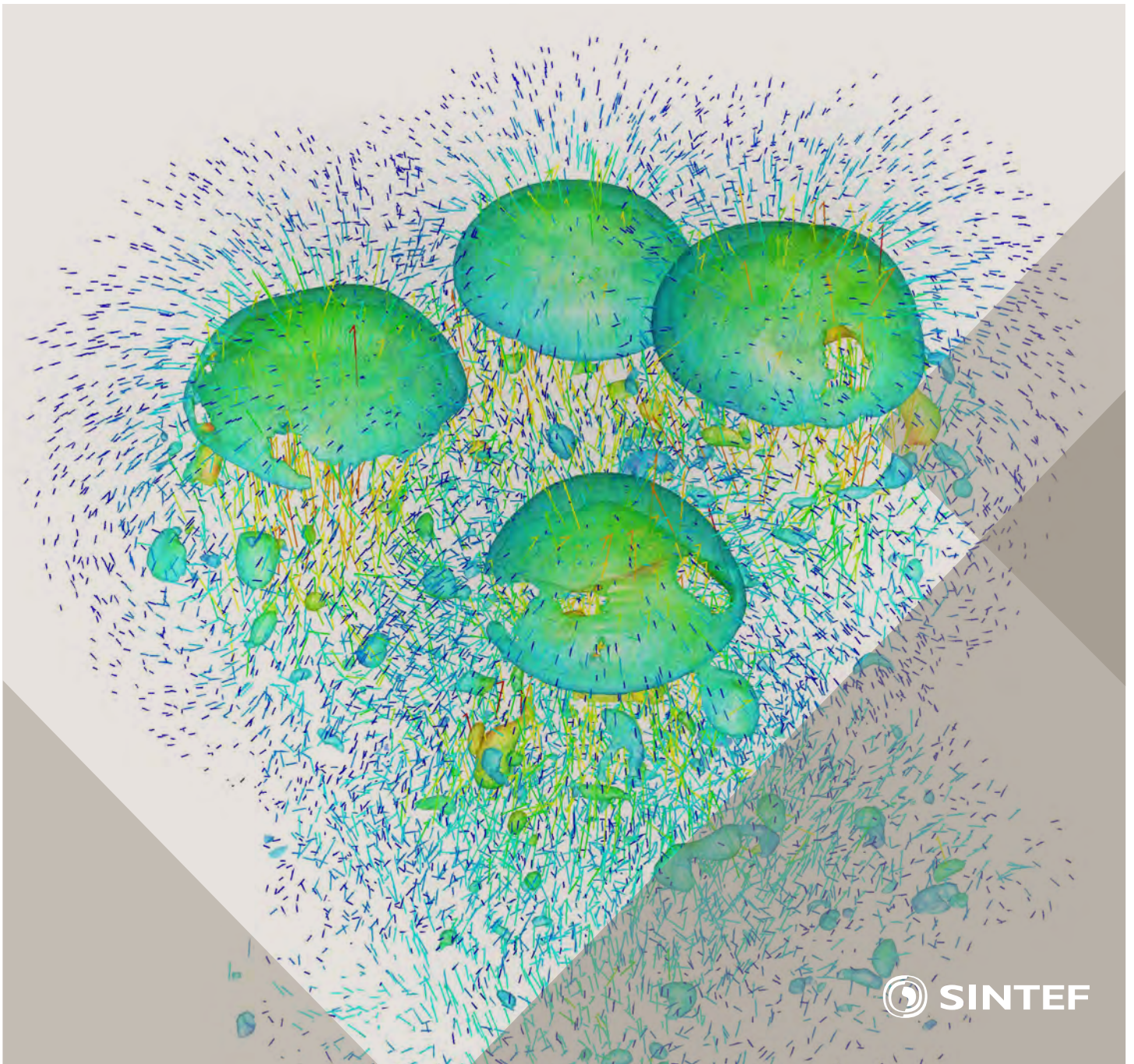


Selected papers from 10<sup>th</sup> International Conference on  
Computational Fluid Dynamics in the Oil & Gas, Metal-  
lurgical and Process Industries

# Progress in Applied CFD



SINTEF Proceedings

Editors:

Jan Erik Olsen and Stein Tore Johansen

## **Progress in Applied CFD**

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Dynamics in the Oil & Gas, Metallurgical and Process Industries

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## PREFACE

This book contains selected papers from the 10<sup>th</sup> International Conference on Computational Fluid Dynamics in the Oil & Gas, Metallurgical and Process Industries. The conference was hosted by SINTEF in Trondheim in June 2014 and is also known as CFD2014 for short. The conference series was initiated by CSIRO and Phil Schwarz in 1997. So far the conference has been alternating between CSIRO in Melbourne and SINTEF in Trondheim. The conferences focus on the application of CFD in the oil and gas industries, metal production, mineral processing, power generation, chemicals and other process industries. The papers in the conference proceedings and this book demonstrate the current progress in applied CFD.

The conference papers undergo a review process involving two experts. Only papers accepted by the reviewers are presented in the conference proceedings. More than 100 papers were presented at the conference. Of these papers, 27 were chosen for this book and reviewed once more before being approved. These are well received papers fitting the scope of the book which has a slightly more focused scope than the conference. As many other good papers were presented at the conference, the interested reader is also encouraged to study the proceedings of the conference.

The organizing committee would like to thank everyone who has helped with paper review, those who promoted the conference and all authors who have submitted scientific contributions. We are also grateful for the support from the conference sponsors: FACE (the multiphase flow assurance centre), Total, ANSYS, CD-Adapco, Ascomp, Statoil and Elkem.

Stein Tore Johansen & Jan Erik Olsen



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## HYDRODYNAMIC INVESTIGATION INTO A NOVEL IC-CLC REACTOR CONCEPT FOR POWER PRODUCTION WITH INTEGRATED CO<sub>2</sub> CAPTURE

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### ABSTRACT

This paper presents an investigation of the hydrodynamics of a new Chemical Looping Combustion (CLC) reactor concept for power generation with integrated CO<sub>2</sub> capture. This concept consists of an internal circulating fluidized bed (IC-CLC) where a single reactor is divided into two separate sections in a way that oxidation and reduction of the oxygen carrier take place separately. Such a reactor configuration would significantly reduce the costs and solids handling challenges currently hampering the scale-up of CLC technology.

Fundamental multiphase flow models (based on the Kinetic Theory of Granular Flow or KTGF) have been used to investigate the hydrodynamics in different reactor designs in order to identify the most optimized one which maintains minimized leakage between the two reactor sections. The performance of each design has been evaluated through a quantified parameter which is the volumetric gas/solid leakage ratio between the two reactor sections. Reactor designs with narrow connecting ports proved to be the most reliable in minimizing the gas leakage and therefore would maintain high CO<sub>2</sub> purity and CO<sub>2</sub> capture efficiency.

The most optimized reactor design has then been selected for further investigation through a statistical method (central composite design) where the reactor performance response to the change in the bed loading and the air fluidization velocity has been evaluated. Values close to 1 were found for the fuel/solid leakage ratio parameter implying that high CO<sub>2</sub> capture efficiency can be achieved through the chosen design in a wide range of bed loadings and fluidization velocities. In general, the reactor proved to perform best under conditions with high static bed heights and/or high fluidization velocities.

**Keywords:** Fluidized bed; Chemical Looping Combustion; Simulation-Based Reactor Design.

### INTRODUCTION

Carbon capture and storage (CCS) is increasingly seen as a potential environmentally sustainable way for fulfilling world energy needs while minimizing the environmental impact of fossil fuel combustion (Butt, Giddings et al. 2012; IEA 2012). Chemical-looping combustion (CLC) has arisen as a promising technology to carry out CO<sub>2</sub> capture at low energy penalty compared to CO<sub>2</sub> capture technologies, such as pre- and post-combustion technologies (Ekström, Schwendig et al. 2009). CLC is a combustion process which integrates power generation and CO<sub>2</sub> capture. It consists of two interconnected reactors, an air and a fuel reactor. A solid oxygen carrier circulating between them, thereby playing the role of oxygen transporter which transfers

the oxygen required for fuel combustion from the air to the fuel. Consequently, any contact between the fuel and the air is inherently avoided in this process, and therefore a pure CO<sub>2</sub> stream, ready for compression and sequestration is produced (Ishida, Zheng et al. 1987).

In addition to development and selection of suitable oxygen carriers materials (Abad, Adanez et al. 2007; Hossain and de Lasa 2008; Adanez, Abad et al. 2012), research on CLC has so far been focused predominantly on the use of dual Circulating Fluidized Beds (CFBs) configuration for the recirculation of the oxygen carrier between the air and the fuel reactors (Abad, Garcia-Labiano et al. 2007; Ekström, Schwendig et al. 2009). Although this configuration has been demonstrated experimentally at lab and pilot scales (Kronberger, Johansson et al. 2004; Linderholm, Abad et al. 2008; Ding, Wang et al. 2012), the dual CFB-based CLC process is still facing many technical and operational complexities which arise mainly from the interconnected reactors configuration. Aside from the design and operational complexity created by the need to manage the solids exchange so that mass and heat balances within the closed loop are fulfilled, the exchange of solids itself brings additional costs and complexity. This circulation between the two reactors requires efficient and costly particles separation system such as a cyclone. This particle separation is particularly difficult due to the extremely harsh reactive and high temperature conditions.

Attempts have followed in recent years to address these issues where reactor concepts with no external solid circulation have been proposed (Noorman, van Sint Annaland et al. 2007; Hamers, Gallucci et al. 2013; Zaabout, Cloete et al. 2013). Following this philosophy, this paper will present a new CLC reactor concept with no external solids circulation. The proposed IC-CLC concept consists of a single reactor divided into two sections, the air and fuel sections, with an oxygen carrier circulating internally between them. This concept is expected to bring large benefits in terms of process simplification through avoiding external solids circulation and process intensification by direct heat integration and ability of operation under pressurized conditions. A large flexibility in the solids circulation rate is also granted due to the ability to operate the air section in a wide range of gas velocities between the bubbling and fast fluidization regime. The fuel section is operated under bubbling regime conditions.

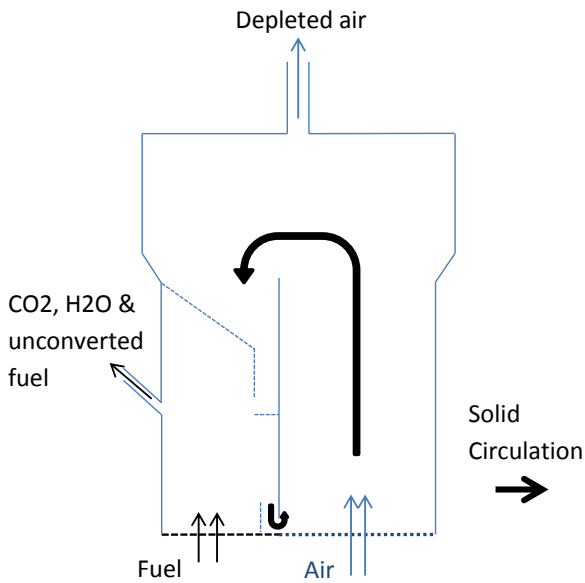
All of these advantages make the IC-CLC concept the most practical solution by which the most important advantages of the interconnected configuration can be



maintained, while alleviating the most important challenges encountered in this configuration.

This paper will use fundamental flow modelling approach for a virtual proof of concept. Fundamental CFD modelling is ideal for this kind of virtual prototyping application because it can be used to quickly and economically evaluate a range of new process ideas and reactor configurations before any expensive and time-consuming physical experiments are conducted. This paper presents a numerical investigation of the hydrodynamics in different reactor designs of the IC-CLC concept. The most optimized design is selected for further hydrodynamic investigation with a dedicated central composite design.

## NEW CONCEPT



**Figure 1: Internal circulating CLC concept IC-CLC**

The proposed IC-CLC concept consists of a single reactor with internal physical partitions creating two reactor sections as shown in Figure 1. Air is injected in one section of the reactor (the fast region) for oxidation of the oxygen carrier while a gaseous fuel is injected to the second reactor section (the slow region) where oxygen carrier reduction takes place. An expanding freeboard region is added where the depleted air transporting oxygen carriers slows down before it leaves the reactor from the outlet at the top while the oxygen carriers fall down into the fuel region. To complete the cycle the oxygen carriers flow back to the air region through the opening at the bottom of the reactor. Gaseous reduction products leave the reactor from the fuel section outlet on the side of the reactor (Figure 1).

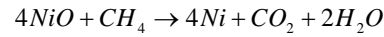
The two reactor sections are connected to each other without any physical separation or seals. This will certainly allow for gas leakage between the two sections in both sides. The viability of this concept will therefore be proved only when the gas leakage between the two sections can be minimized to tolerable quantities.

In order to calculate the amount of gas leakage that will occur, it can be assumed that the gas will leak from one reactor section to the other at a rate proportional to the

solids circulation rate. Hence, a new variable for the volumetric ratio of gas to solids transferred from one reactor section to the next can be defined: the gas/solids leakage ratio. At a fixed gas/solids leakage ratio, the gas leakage will therefore be proportional to the solids circulation rate required to react with a given gas stream. If the rate of solids recirculation is very small in relation to the gas feed streams and the gas/solids leakage ratio is kept small by good reactor design, it is possible that gas leakages can become very small.

As an example, the gas/solids leakage ratio can be calculated for a CLC system using a NiO oxygen carrier and methane as fuel. The reaction occurring in the fuel reactor section is as follows (assuming full oxidation of methane):

**Equation 1 :**



When assuming a solids circulation rate ( $\dot{m}_{solids}$ ) of 1 kg/s, a desired degree of solids conversion ( $c_{NiO}$ ) of 50% and an active material content in the oxygen carrier ( $x_{NiO}$ ) of 40%, the molar flowrate of NiO ( $\dot{n}_{NiO}$ ) to be reacted coming into the fuel reactor can be calculated as:

**Equation 2:**

$$\dot{n}_{NiO} = \frac{\dot{m}_{solids} c_{NiO} x_{NiO}}{M_{NiO}} = \frac{1 \cdot 0.5 \cdot 0.4}{0.0747} = 2.677 \text{ mol/s}$$

According to the stoichiometric ratio of reactants, 0.669 mol/s of methane would be required to achieve 50% conversion of the solids being fed at 1 kg/s. Assuming 99.9% fuel conversion and no side-reactions, the outlet stream of the fuel reactor section would consist of 0.669 mol/s CO<sub>2</sub>, 0.00067 mol/s of unreacted CH<sub>4</sub>, 1.337 mol/s of H<sub>2</sub>O (which will be easily condensed out before compression, transport and storage) and a certain amount of depleted air which leaked from the air reactor.

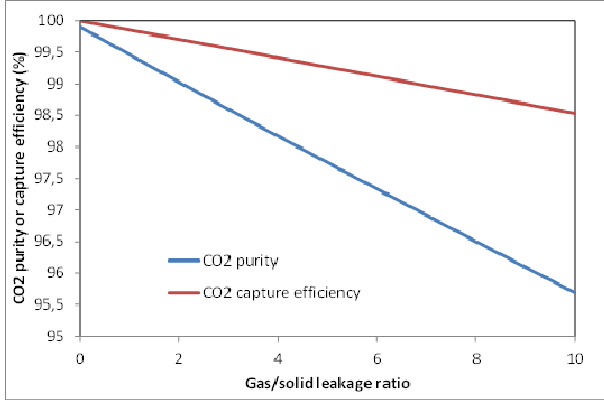
For an oxygen carrier material density ( $\rho_{solids}$ ) of 3400 kg/m<sup>3</sup>, a reactor temperature ( $T$ ) of 1200 K and atmospheric pressure ( $P$ ), the molar flowrate of gas leakage between reactor sections ( $\dot{n}_{leak}$ ) can be calculated as a function of the gas/solids leakage ratio ( $X$ ).

**Equation 3:**

$$\dot{n}_{leak} = \frac{\dot{V}_{leak} P}{RT} = \frac{X \frac{\dot{m}_{solids}}{\rho_{solids}} P}{RT} = X \frac{1}{8.314 \cdot 1200} 101325 = 0.00299 \cdot X \text{ mol/s}$$

Assuming that the gas leaking from the fuel reactor to the air reactor is fully converted (consisting only of CO<sub>2</sub> and H<sub>2</sub>O in a 1:2 molar ratio), the rate of CO<sub>2</sub> leakage to the air reactor will be one third of that calculated in Eq. 3. Using this information, the purity of the CO<sub>2</sub> (after

water condensation) from the fuel reactor and the CO<sub>2</sub> capture efficiency (determined by the CO<sub>2</sub> which leaked to the air reactor) can be plotted as a function of the gas/solids leakage ratio. For this particular example, this plot is given in Figure 2.



**Figure 2:** CO<sub>2</sub> purity and CO<sub>2</sub> capture efficiency as a function of gas/solid leakage ratio.

It can be seen that both the CO<sub>2</sub> purity and CO<sub>2</sub> capture efficiency remain high even at high gas/solid leakage ratios. This is a result of the very small volumetric flowrate of solids that is required to react with a given volumetric flowrate of gas and shows that the IC-CLC concept holds great promise for this particular process. The potential for practically controlling the gas/solids leakage ratio will be further investigated in this work through CFD simulations. This paper will explore options of reactor and connection ports design and operating conditions which would result in minimized gas leakage between the two reactor sections. It is important that the gas leakage is minimized in a way to result in high CO<sub>2</sub> capture efficiency and CO<sub>2</sub> purity. The effect of reducing the contact areas between the two reactor sections by adding physical separation walls (as show the dashed lines in figure 1) close to the connecting opening at the top and the bottom will be investigated using CFD simulations.

### SIMULATIONS

Simulations were carried out using the well-established two fluid model (TFM) closed by the kinetic theory of granular flows (KTGF).

#### Model equations

Conservation equations are solved for each of the two phases present in the simulation. The continuity equations for the gas and solids phases phase are given below:

#### Equation 4:

$$\frac{\partial}{\partial t}(\alpha_g \rho_g) + \nabla \cdot (\alpha_g \rho_g \vec{v}_g) = 0$$

$$\frac{\partial}{\partial t}(\alpha_s \rho_s) + \nabla \cdot (\alpha_s \rho_s \vec{v}_s) = 0$$

Momentum conservation for the gas phase is written as

#### Equation 5:

$$\frac{\partial}{\partial t}(\alpha_g \rho_g \vec{v}_g) + \nabla \cdot (\alpha_g \rho_g \vec{v}_g \vec{v}_g) = -\alpha_g \nabla p + \nabla \cdot \bar{\bar{\tau}}_g + \alpha_g \rho_g \vec{g} + K_{sg}(\vec{v}_s - \vec{v}_g)$$

And for the solids as

$$\frac{\partial}{\partial t}(\alpha_s \rho_s \vec{v}_s) + \nabla \cdot (\alpha_s \rho_s \vec{v}_s \vec{v}_s) = -\alpha_s \nabla p - \nabla p_s + \nabla \cdot \bar{\bar{\tau}}_s + \alpha_s \rho_s \vec{g} + K_{gs}(\vec{v}_g - \vec{v}_s)$$

The inter-phase momentum exchange coefficient ( $K_{gs} = K_{sg}$ ) was modelled according to the formulation of Syamlal and O'Brian (Syamlal, Rogers et al. 1993).

Solids phase stresses were determined according to the KTGF analogy. The conservation equation for granular temperature is given below:

#### Equation 6:

$$\frac{3}{2} \left[ \frac{\partial}{\partial t}(\alpha_s \rho_s \Theta_s) + \nabla \cdot (\alpha_s \rho_s \vec{v}_s \Theta_s) \right] = (-p_s \bar{\bar{I}} + \bar{\bar{\tau}}_s) : \nabla \vec{v}_s + \nabla \cdot (k_{\Theta_s} \nabla \Theta_s) - \gamma_{\Theta_s} + \phi_{gs}$$

This partial differential equation was simplified to an algebraic equation by neglecting the convection and diffusion terms (Van Wachem, Schouten et al. 2001). The two final terms in Eq. 6 are the collisional dissipation of energy (Lun, Savage et al. 1984) and the interphase exchange between the particle fluctuations and the gas phase (Gidaspow, Bezburuah et al. 1992). Solids stresses are calculated according to shear and bulk (Lun, Savage et al. 1984) viscosities. The shear viscosity consists of three parts: collisional (Gidaspow, Bezburuah et al. 1992; Syamlal, Rogers et al. 1993), kinetic (Syamlal, Rogers et al. 1993) and frictional (Schaeffer 1987). The solids pressure formulation by Lun et al. (Lun, Savage et al. 1984) was enhanced by the frictional pressure formulation by Johnson and Jackson (Johnson and Jackson 1987). The radial distribution function of Ogawa and Oshima (Ogawa, Unemura et al. 1980) was employed.

#### Boundary conditions

A simple no-slip wall boundary condition was set for the gas phase. The Johnson and Jackson (Johnson and Jackson 1987) boundary condition was used for the granular phase with a specular coefficient of 0.25.

#### Equation 7:

$$\bar{\bar{\tau}}_s = -\frac{\pi}{6} \sqrt{3} \zeta \frac{\alpha_s}{\alpha_{s,max}} \rho_s g_{0,ss} \sqrt{\Theta_s} \vec{U}_{s||}$$

The inlet condition was specified as a velocity inlet injecting air at a flow rate of 0.6 m/s. CO<sub>2</sub> was injected through a velocity inlet on the back wall to mimic the experiment. In the 2D simulation, CO<sub>2</sub> was injected via a source term. The outlet was designated as a pressure outlet at atmospheric pressure.

#### Flow solver and solver settings

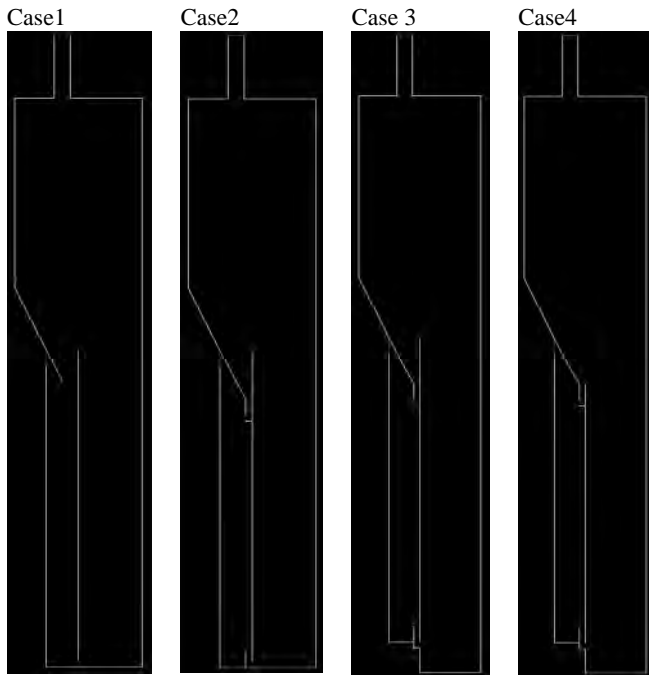
The commercial software package, FLUENT 13.0 was used as the flow solver. The phase coupled SIMPLE scheme (Patankar 1980) was used for pressure-velocity coupling and the higher order QUICK scheme (Leonard and Mokhtari 1990) for the spatial discretization of all remaining equations. First order implicit temporal discretization was used (Cloete, Amini et al. 2011). Unsteady state simulations have been performed with a time step of 0.001s. A restitution coefficient of 0.9 has been used. Further model setting can be found in (Cloete, Zaabout et al. 2013)

## RESULTS AND DISCUSSIONS

The IC-CLC concept allows for large flexibility in terms of designing the partitions and connections

between the fuel and the air zones. The resulting reactor design has a direct impact on the reactor performance. A reliable reactor design should be able to maintain minimized leakage between the two reactor sections to ensure high CO<sub>2</sub> purity and CO<sub>2</sub> capture efficiency. Hence, to reduce the risk, it is highly recommended to use an efficient and quick numerical tool to assess the different reactor design before any expensive and time-consuming experimental demonstration is done. The well-known Two Fluid Model ((TFM) approach closed by the Kinetic Theory of Granular Flows (KTGF)) was used for this purpose as will be shown below.

### Reactor design



**Figure 3: IC-CLC concept: investigated reactor configurations.**

Several reactor configurations have been investigated as shown in Figure 3 where the emphasis was put on changing the extent of the connection areas between the two reactor sections (the high of the unit is 2 m in total; the reactor zone is 1 m height and the freeboard region is 1 m. The fuel section width is 0.1 m and the air section width is 0.2 m). Case 1, for example, exhibits large contact areas between the two reactor sections while, for the three other configurations, the contact areas were narrowed to 2 cm width connecting ports; one at the bottom and the other at top. Parameters such as solids circulation rate, air leakage to the fuel section and fuel leakage to the air section have been quantified for each reactor configuration and the performances of each configuration were compared through the previously discussed gas/solid leakage ratio parameter which describes the ratio between the volumetric gas leakage through the port and the volumetric solids circulation rate. Gas feeds to both reactor sections and the static bed height were kept the same for the four reactor configurations during this comparative study: air was used as a feed gas to the air section with 1 m/s fluidization velocity,  $U$ , and methane was used as a feed gas to the fuel section with a fluidization velocity of 0.3 m/s. The initial static bed height,  $H_s$ , was 0.5 m. Glass

beads of 200  $\mu\text{m}$  mean diameter and 2500  $\text{kg/m}^3$  density were used as a bed to be fluidized (The minimum fluidization velocity is 0.0451 m/s).

As shown in Table 1, results from case 1 show high air and fuel leakages through the two connections with a very low solid circulation rate between the two reactor sections. With large connecting areas between the two reactor sections in case 1, gases in both reactor sections find less resistance to flow freely in both sides, causing therefore a large amount of leakage. The resulting air/solid and fuel/solid ratios are very high, implying that this reactor design fails to deliver high CO<sub>2</sub> purity and CO<sub>2</sub> capture efficiency.

**Table 1: Effect of reactor design: simulations results. Data have been averaged over 30s real process time.**

|   | Case 1  | Case 2   | Case 3  | Case 4   |
|---|---------|----------|---------|----------|
| Air flow rate-bot ( $\text{m}^3/\text{s}$ )     | 0.00189 | 2.02E-05 | 0.00037 | 1.24 E-5 |
| Air flow rate-top ( $\text{m}^3/\text{s}$ )     | 0.00857 | 0.00692  | 0.00604 | 0.00541  |
| Fuel flow rate-bot ( $\text{m}^3/\text{s}$ )    | 0.00837 | 0.00531  | 0.00432 | 0.00781  |
| Fuel flow rate-top ( $\text{m}^3/\text{s}$ )    | 0.01299 | 0.00043  | 0.00092 | 0.00032  |
| Total-solid flow rate ( $\text{m}^3/\text{s}$ ) | 0.00022 | 0.00386  | 0.00337 | 0.00530  |
| Fuel flow rate /solid ratio                     | 93.9225 | 1.4865   | 1.55856 | 1.53449  |
| Air flow rate /solid ratio                      | 46.0169 | 1.795499 | 1.90612 | 1.02368  |

For the three other cases where the connections between the two regions are narrowed, high solids circulation rates were observed. Case 2 and 3 showed comparable values of solids circulation rate although the extension of the air region at the bottom for case 3. Case 4 however showed a 25 % increase in the solids circulation rate compared to case 2 and 3; by shortening the bar separating the two regions in case 4 to the level of the narrow port at the top it becomes easier for more entrained solids to the freeboard to reach the side and fall into the port to circulate to the fuel section. As for the gas leakage between the two reactor sections, the fuel was found to leak mainly through the port at the bottom while the air leaks through the port at the top. It naturally follows the solids circulation path.

The reactor performance with respect to gas leakage was quantified in the form of fuel/solid and air/solids leakage ratio parameters. The three reactor designs with narrow connection ports show values lower than 2 for both fuel/solid and air/solid leakage ratios implying that the three of them have the potential to maintain high CO<sub>2</sub> capture efficiency and CO<sub>2</sub> purity. The port design is therefore the most influential parameter in the gas leakage between the two reactor sections; narrow connection port creates conditions with solids flowing close to maximum packing which make them play the role of a physical plug that restricts the gas freedom to flow through the port.

In the light of the results presented above, the reactor configuration depicted in Figure 4 was selected for further investigation. The top part of the unit was

expanded in both the right and left sides for allowing for larger air flow rate while preventing solids elutriation. Instantaneous solids holdup in the unit shows denser bed conditions in the fuel section due to low gas feed and dilute bed conditions in the air section due to the high fluidization velocity in that region. Continuous solids circulation between the two reactor sections is established due the difference in the gas feed velocities in each section: solids are first entrained by air to the freeboard then fall into the connecting port at the top, and thereafter flow down to the fuel section. Accumulation of solids in the fuel section leads to hydrostatic pressure build up which drives the solids to flow back to the air section through the connecting port at the bottom.

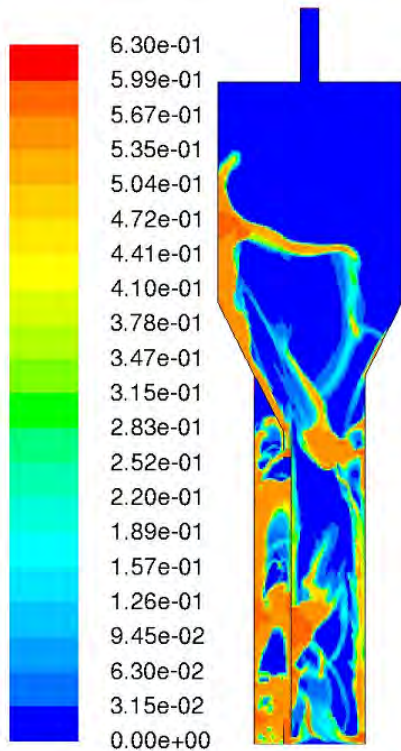


Figure 4: Instantaneous solid holdup in the rig;  $H_s = 0.6$  m and  $U = 1.25$  m/s.

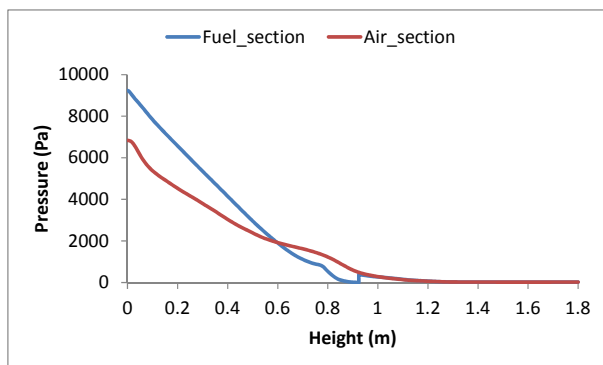


Figure 5: Mean axial pressure plotted along the two reactor sections;  $H_s = 0.6$  m and  $U = 1.25$  m/s.

The axial pressure plotted along the height of the reactor in both the fuel and the air sections (Figure 5) illustrates this solids circulation mechanism where it shows the pressure build up in the fuel section. At the bottom part of the reactor the pressure in the fuel section is higher than in the air section. This forces the

solids to flow to the air region through the port at the bottom. At the top part of the reactor the situation is inverted: the pressure in the air section becomes higher than in the fuel one. This pressure difference drives the solids to flow to the fuel section through the port in the top.

### More detailed investigation of the reactor configuration

In the light of this good gas leakage performance, the chosen reactor configuration was further investigated by means of a central composite design. A central composite design is a statistical method which allows for the fitting of a second order model through a set of data collected from well-defined experiments (simulation experiments in this case). The relative importance of the different effects in relation to each other can be quantified, together with the accuracy by which the model fits the data. In this case, however, only the response surface is required to gain a better understanding of the performance of this concept as two independent variables were varied:

- The first independent variable was the static bed height (solids loading inside the reactor). Since the mass transfer between reactor sections is driven by hydrostatic pressure differences, the height of the beds in the respective sections are expected to have an influence on the solids recirculation rate and the gas leakage. This factor is expressed as the static bed height with which the simulation was initialized (the initial solids volume fraction was 0.6).
- The velocity at which the air bed section was fluidized was the second factor. A greater difference between the fluidization velocities in the two reactor sections is expected to increase the solids recirculation rate. This factor was expressed as the ratio between the fluidization velocity in the fast section and that in the slow section which was kept constant at 0.3 m/s. As can be seen in **Error! Reference source not found.**, the section at the right was taken as the fast section (air section) and the left section as the slow one (fuel section).

Table 2: Central composite design with varying the fluidization velocity in the air section and the static bed height.

|        | U (m/s) | H (m)  |
|--------|---------|--------|
| 1      | 0.8965  | 0.2586 |
| 2      | 0.8965  | 0.5414 |
| 3      | 1.6036  | 0.2586 |
| 4      | 1.6036  | 0.5414 |
| 5      | 0.7500  | 0.4000 |
| 6      | 1.7500  | 0.4000 |
| 7      | 1.2500  | 0.2000 |
| 8      | 1.2500  | 0.6000 |
| 9 (C)  | 1.2500  | 0.4000 |
| 10 (C) | 1.2500  | 0.4000 |

Ten simulations were completed at different values of these two independent variables as specified in Table 2.

The same particles as in the previous section are used for this central composite design study. The response of the following three dependent variables was recorded:

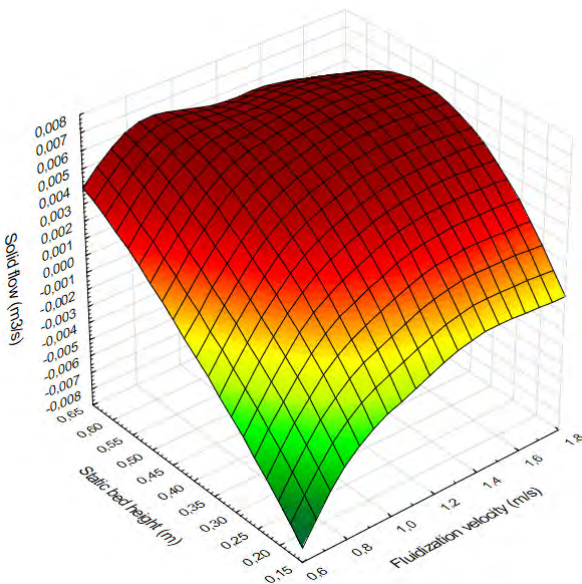
- The solids recirculation rate ( $\text{m}^3/\text{s}$ ) quantified as the average volumetric flowrate of the solids through the two connection ports.
- The gas/solid leakage ratio from the air reactor section into the fuel reactor section.
- The gas/solid leakage ratio from the fuel reactor section into the air reactor section.

### Reactor performance within the central composite design

As expected, both the fluidization velocity in the air section and the static bed height play a major role in driving solids circulation between the two reactor sections. As can be seen in Figure 6, solid flow rate increases with both the initial static bed height and the fluidization velocity in the air section, although it tends to plateau for higher values. Naturally by increasing either the static bed height or the fluidization velocity larger amount of solids is entrained to the freeboard and falls into the fuel region. Greater solids accumulation in the fuel section leads to greater hydrostatic pressure which builds up and thereby forces a greater amount of solids to circulate back to the air region through the port at the bottom.

This is actually true as long as the port in the top is not completely full of solids. The form of the plateau shown by the response surface is due to the restriction imposed by the size of the connections ports on the solids flow; at high solids entrainment the connections ports get completely filled with solids and therefore restrict further increase in the solids circulation rate. At high static bed heights the unit can also face a loss of solids through the outlet in the fuel section. This explains the decrease in the solids circulation rate shown by the response surface at high static bed heights combined with high fluidization velocity in the air section.

Through this kind of surface response it is also easy to identify the operating conditions under which no solids circulation takes place making it therefore clearer to identify where the process would fail.

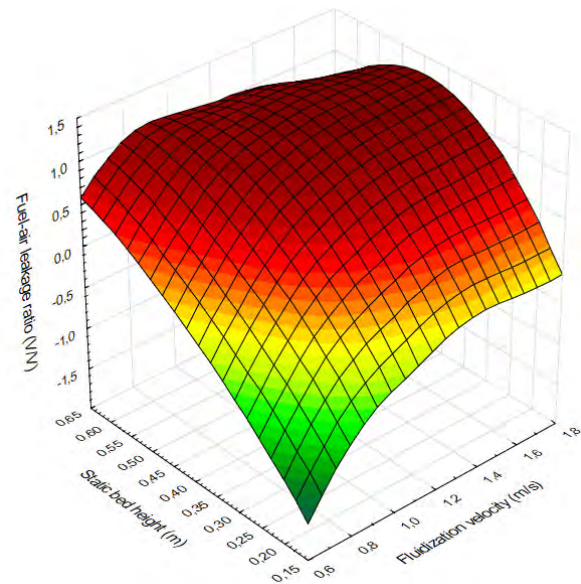


**Figure 6: Volumetric solid flow rate response to the change in the fluidization velocity in the air section and the initial static bed height**

As for the gas leakage it was found that fuel leakage takes place mainly through the port at the bottom while the air leakage takes place through the port at the top.

The fuel/solid leakage ratio response to the change in the fluidization velocity and the static bed height showed similar shape to that found for the solids flow rate (Figure 7); it increases with the increase in the fluidization velocity and static bed height and then plateaus. As explained previously the plateau forms due to the maximum solids packing conditions which take place in the port during high solids entrainment from the air section. In these conditions any further increase in the solids circulation rate and gas leakage is restricted by the port size.

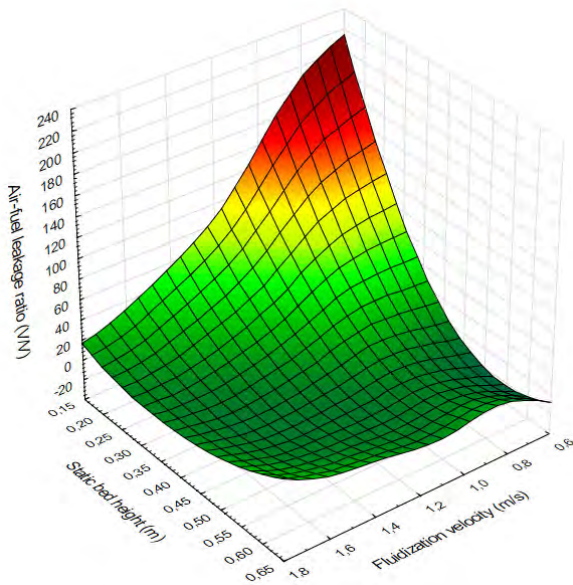
Values close to 1 were found for the fuel/solid leakage ratio for all operating conditions within the central composite design (the negative values at very low static bed heights and fluidization velocities are simply the result of extrapolation). This means that the chosen reactor configuration is able to maintain very high  $\text{CO}_2$  capture efficiency.



**Figure 7: Fuel/solid leakage ratio response to the change in the fluidization velocity in the air section and the initial static bed height**

The air/solid leakage ratio responds differently to the change in the air fluidization velocity and the static bed height. Values close to one were found for large fluidization velocities and static bed heights proving the ability of this reactor configuration to maintain very high  $\text{CO}_2$  purity within these conditions. However, regions of poor performance were identified where operation of the concept should be avoided; combining low air fluidization velocities with low static bed heights results in very high air/solids leakage ratio and thereby very low  $\text{CO}_2$  purity according to the discussion in section 2. The high values of the air/leakage ratio are consequences of the low solids circulation rate between the two reactor sections (Figure 8). As shown earlier, high solids circulation rates are required to create conditions close to maximum solids packing in the port

at the top (from where most of the air leakage takes place), to reduce the freedom of air to leak to the fuel section.



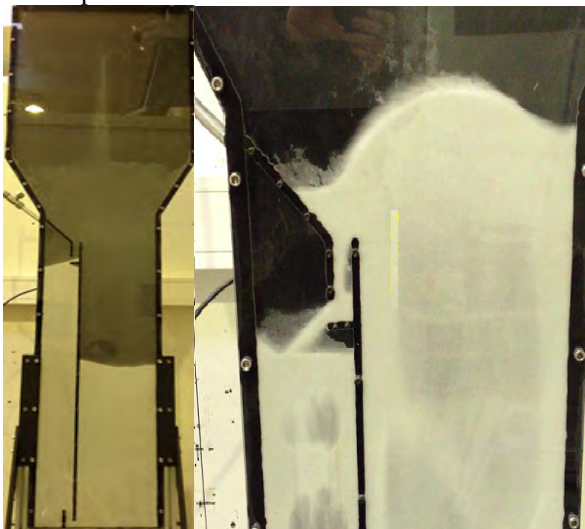
**Figure 8:** Air/solid leakage ratio response to the change in the fluidization velocity in the air section and the initial static bed height

### **Experimental unit**

In the light of the promising results showed by simulations, a pseudo 2D (1.5 cm depth) experimental unit was constructed and operated in order to study the hydrodynamics of the IC-CLC concept. The bottom part of the unit is 1m height and the freeboard is 80 cm. Narrow connections ports of 2 cm width have been made (

Figure 9). First tests of the unit with glass particles of 200  $\mu\text{m}$  showed stable operation with continuous solids circulation between the two unit sections. Conditions where solids flow close to maximum packing in the connection ports were also observed (

Figure 9); no blockage of the ports have been observed to take place in these conditions.



**Figure 9:** Pseudo 2D IC-CLC experimental setup.

Further testing on this unit is still required to prove experimentally the potential of this concept to maintain minimized leakage between the two unit sections. Parameters such as the solids circulation rate and gas leakages between the two unit sections will be quantified within a wide range of operating conditions.

### **CONCLUSION**

A hydrodynamic investigation of an innovative CLC concept based on internal circulating fluidized bed (IC-CLC) has been carried out. The new concept consists of a single reactor with a physical separation inside dividing it into two sections; the fuel and the air sections. Oxygen carriers circulate between the two sections through two connecting ports, one at the bottom and one at the top. These connections allows for gas leakage in both sides which reduces the concept ability to capture CO<sub>2</sub> and generate high purity CO<sub>2</sub>. Good reactor design is therefore the key for the IC-CLC concept to succeed in maintaining high CO<sub>2</sub> purity and high CO<sub>2</sub> capture efficiency.

Fundamental multiphase flow models (based on the Kinetic Theory of Granular Flow KTGF) have then been used in this study to investigate the hydrodynamics in different reactor designs in order to identify the most optimized one before any expensive experimental demonstration is made. Reactor designs with narrow connection ports have resulted in the best leakage performance; small port size creates solids packing conditions which play the role of a physical plug that restricts the gas freedom to flow through the ports. The most optimized design has been further investigated in a framework of a central composite design where the bed loading and gas fluidization velocity in the air section are varied. Values close to 1 have been found for the fuel/solid ratio parameter implying that high CO<sub>2</sub> capture efficiency can be achieved through this design in a wide range of operating conditions. Regions of poor performance were also identified where the design would fail to maintain high CO<sub>2</sub> purity. In particular, low bed loadings and low fluidization velocities in the air section should be avoided as they results in high values of the air/solid ratio.

In the light of the promising results found in simulations, an experimental unit designed with narrow connection ports was constructed. The first tests already showed that low gas/solid leakage ratio can be achieved by this design.

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