

FLUID DYNAMIC CHARACTERIZATION OF A 50 kW UNIT FOR CLC OF COAL WITH IMPROVED CONFIGURATION

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Abstract –Chemical Looping Combustion (CLC) is a promising technology to perform the CO₂ capture at a low economic and energetic cost because CO₂ separation is inherent to the process. In CLC, oxygen from air is transferred to fuel by means of the use of a solid oxygen carrier which is continuously circulating between two reactors, namely fuel and air reactors. At ICB-CSIC, a 50 kW_{th} CLC unit for solid fuels, e.g. coal, has designed and constructed, which includes two interconnected circulating fluidized bed reactors with the oxygen carrier continuously circulating between them. In this work, the effect of main operating conditions on the fluid dynamics of the 50 kW_{th} CLC unit is evaluated. Thus, the effect of the gas flow in air or fuel reactors and the solids inventory in the CLC unit on both the solids circulation rate and the inventory of solids in the fuel reactor is investigated. As a result, it is found that the solids flux is mainly influenced by the amount of solids in the CLC unit, which highly affects both to the pressure drop in the fuel reactor and the global solids circulation flow. Also, the solids circulation rate between reactors increases with the gas flow in air or fuel reactors. In addition, the solids circulating flow rate can be controlled by acting on a dedicated double loop seal.

INTRODUCTION

Anthropogenic CO₂ emissions to the atmosphere are empowering the naturally occurring greenhouse effect, which results in a rise of the average temperature in the Earth's surface. There is a general scientific consensus on the global warming will have great consequences to the environment, the biological mechanism of the animals and plants, and our life-style and economy (Field et al., 2014). To prevent these severe effects, the Paris Agreement urges to decarbonize the world energy systems in the near future in order to limit the increase in the average world temperature to 2 °C above pre-industrial levels (UNFCCC, 2015). In order to reach this goal, CO₂ capture and storage from fossil fuel combustion can represent a relevant mid-term solution considering the future trend in the use of this kind of fuels (IPCC, 2005). In this sense, Chemical Looping Combustion (CLC) has been identified as a promising technology to perform CO₂ capture at a low economic and energetic cost because the CO₂ separation is inherent to the process regardless of the type of fuel used, either gaseous, liquid or solid (Adanez et al., 2012).

In CLC with solid fuels, the fuel is physically mixed with the oxygen carrier. A scheme of the CLC with solid fuels is shown in Fig. 1. The oxygen needed is supplied by an oxygen carrier, normally a metal oxide, which circulates between the so-called fuel and air reactors. In the fuel reactor, the fuel is oxidized to CO₂ and H₂O, which facilitates CO₂ capture once the water has been condensed. The reduced oxygen carrier is then transported to the air reactor where it is re-oxidized before starting a new cycle. The net chemical reaction and combustion enthalpy is the same as in a conventional combustion and CO₂ capture is inherent to this process, as the air is not mixed with the fuel. Significant advances have been made in the development of CLC using solid fuels in recent years (Adanez et al., 2012), by means of two processes: *in-situ* gasification CLC (*i*G-CLC) and Chemical Looping with Oxygen Uncoupling (CLOU).

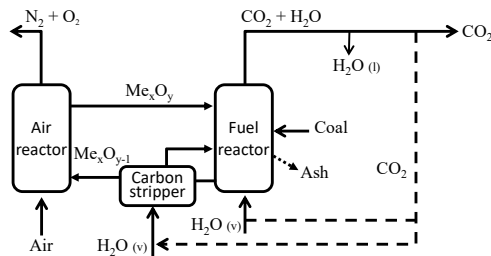


Fig. 1. Reactor scheme of CLC process with solid fuels.

In the *iG*-CLC concept, coal is fed directly to the fuel reactor and mixed with the oxygen carrier. Steam or recycle CO₂ are supplied to the fuel reactor as fluidizing agents. The *in-situ* gasification of coal happens, generating volatile matter and gasification products, which are oxidized by reactions with the oxygen carrier. In this process, gasification is the limiting step of this process for coal conversion and some unconverted char in the oxygen carrier stream can reach the air reactor, where it would be burnt. This loss of carbon in the air reactor decreases significantly the CO₂ capture efficiency of the process and therefore it should be avoided. To increase the residence time of char particles in the fuel reactor, without excessively increasing the reactor size, a carbon separation unit after the fuel reactor is included.

In the CLOU concept (Mattisson et al., 2009), the used oxygen carrier can release gaseous oxygen by taking advantage of the thermochemical property of some metal oxides, e.g. CuO and Mn₂O₃, to decompose at high temperature. Thus, complete combustion and CO₂ capture rates close to 100% has been found in a 1 kW_{th} CLOU unit due to the fast combustion of coal with gaseous oxygen generated *in-situ* in the fuel reactor (Abad et al., 2012).

Several design concepts have been developed for *iG*-CLC ranging from 0.5 kW_{th} to 4 MW_{th} units, but no operation at scale higher than 1 kW_{th} is found for CLOU (Abad et al., 2017). Recently, a 50 kW_{th} CLC unit for solid fuels has been designed to be operated both in *iG*-CLC and CLOU modes (Abad et al., 2015a). This unit was designed to allow an independent control of solids circulation flow rate and solids inventory in the fuel reactor. Thus, direct relation between operational conditions and performance of the CLC unit could be easily evaluated (Pérez-Vega et al., 2016). In this unit, the performance of the *iG*-CLC and CLOU processes burning different fuels (lignite, bituminous coal and biomass) will be evaluated using different materials (Fe-based minerals or copper-based oxygen carrier) in order to improve the development of the technology of CLC with solid fuels. In CLC, the main operating conditions, i.e. solids circulation rate and solids inventory, are closely related to the oxygen carrier characteristics, i.e. oxygen transport capacity and reactivity (Abad et al., 2007). In this work, a fluid dynamic study is performed to determine the flexibility of the 50 kW_{th} CLC unit regarding the solids circulation flow rate and the solids inventory in the fuel reactor. Thus, the capability of this unit to process solid fuels with different oxygen carrier materials may be assessed. Instead of the common task of extrapolating results from a cold flow model, the fluid dynamic study is done under realistic conditions, i.e. high temperature and using an oxygen carrier as particulate material. Focus is done on the effect of “external” factors, such as the gas flow fed to the fuel and air reactors or the total solids inventory in the CLC unit, on the “internal” operating conditions, such as solids circulation rate and solids inventory in the fuel reactor.

EXPERIMENTAL

A scheme of the CLC unit for solids fuels is shown in Fig. 2. The nominal thermal power is 20 kW_{th} for *iG*-CLC mode and 50 kW_{th} for CLOU mode. The difference of nominal power is because the better performance of CLOU with respect to *iG*-CLC on the basis of CO₂ capture and combustion efficiency (Adánez et al., 2013; Mendiara et al., 2016). The CLC unit is based on two interconnected circulating fluidized bed reactors, the air and fuel reactor, and a carbon stripper, being a bubbling fluidized bed. The solid fuel feeding point was just above the distributor plate in the fuel reactor. It is intended to convert the fuel to CO₂ and H₂O, minimizing the unburnt compounds in both the gaseous stream, which reduces the combustion efficiency, and the oxygen carrier stream, which reduces the CO₂ capture. Both fuel reactor and carbon stripper can be fluidized by steam and/or CO₂, while air is used for the air reactor. The use of steam is required in *iG*-CLC mode due to the more efficient gasification with this gas (Cuadrat et al., 2012), but CO₂ is desired for CLOU in order to avoid losses in the energetic efficiency due to steam generation for the process. The fuel reactor temperature for design conditions was considered to be 1000 °C in *iG*-CLC and 950 °C in CLOU. The CLC unit is not expected to be auto-thermal due to heat losses associated to the unit size. Then, the fuel reactor, air reactor, carbon stripper, and loop seals are heated by separate furnaces.

Mass balances, fluid dynamics considerations and the performance of the fuel reactor predicted by a theoretical model previously developed (Abad et al., 2013), were taken into account for the design of the CLC unit. Design data are shown in Table 1, and Fig. 3 was developed for this purpose (Abad et al., 2015b). The cross sectional area corresponded to 0.39 m²/MW_{th} in *iG*-CLC and 0.16 m²/MW_{th} in CLOU. For design conditions, the inlet gas velocity was just above the terminal velocity of oxygen carrier particles; see Fig. 3. When the gases from coal conversion and gas stream from the carbon stripper were considered, the gas velocity was estimated to be 4.7 m/s in *iG*-CLC and 5.7 m/s in CLOU. The gas velocity in the upper part of the reactors was high enough to entrain solids from air and fuel reactor. The solids inventory in the fuel reactor was fixed to 1000 kg/MW_{th} for *iG*-CLC and 400 kg/MW_{th} for CLOU, corresponding to a pressure drop of 25 kPa, and it was considered suitable values from previous works (Abad et al., 2012; García-Labiano et al., 2013). The air reactor was over-sized in order to assure a complete oxidation of the oxygen

carrier. In addition, the gas velocity in the air reactor riser was high enough to be able to entrain all the solids coming from the carbon stripper, i.e. the air reactor neither control nor limit the solids circulation flow rate in the CLC unit.

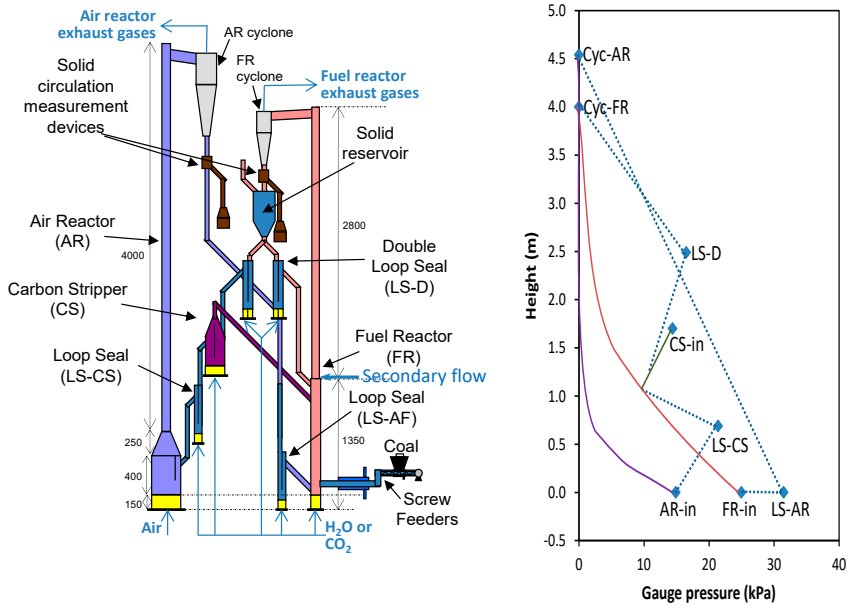


Fig. 2. Left: Schematic layout of the 50 kW facility at ICB-CSIC (dimensions in mm). Right: pressure profile in the CLC unit. Dotted lines are elements connected for different loop seals. Continuous lines: pressure evolution with height in each element of the CLC unit.

The carbon stripper separates the unconverted char particles from the oxygen carrier according to their different fluid dynamic properties. Char particles are lighter and smaller than oxygen carrier particles. So, char particles can be elutriated from the carbon stripper and recirculated to the fuel reactor using an appropriate fluidization flow, whereas the oxygen carrier particles are left to pass to the air reactor. The gas velocity in this unit must be higher than terminal velocity of char particles ($u_{t, char} = 0.1$ m/s), but lower than terminal velocity of oxygen carrier particles ($u_{t, OC} = 1$ m/s); see Fig. 3. A gas velocity in the carbon stripper of 0.35 m/s seems to be suitable to reach an effective separation of char particles from oxygen carrier.

Loop seals avoid the mixing of gases between different elements in the CLC unit. Also, loop seals must balance the pressure between the elements it connects. Thus, the length of the low pressure hand side of each loop seal is given by the pressure balance in the system, as it is shown in Fig. 2. Estimated pressure drops in each element of the CLC unit for the design condition can be calculated following continuous lines in this figure.

The solids circulation flow rate can be controlled by acting on the double loop seal (LS-D). The LS-D system is designed in order to divide the solids stream from the fuel reactor cyclone in two: (1) solids to the carbon stripper, and then to the air reactor, in order to fulfill the required solids circulation flow rate between air and fuel reactors; and (2) excess of solids was recirculated to the fuel reactor. The use of a double loop seal was considered to be a relevant improvement with respect to existing CLC units for solid fuels. Another important point is the possibility to have a direct measurement of the solids flux exiting from both the fuel and air reactor by means of the respective solid circulation measurement devices.

In this work, the flexibility of the CLC unit has been investigated. Thus, the effect of “external” factors, such as the gas velocity in the fuel reactor and the amount of solids in the whole unit, on “internal” operating conditions, such as solids circulation flow rate and solids inventory in the fuel reactor, is determined. For this purpose, the main and secondary flows in the fuel reactor, the air flow in the air reactor, and the fluidizing conditions in the double loop seal LS-D were modified. Also, the total inventory of solids in the facility was varied. The solids flow entrained from both the fuel and air reactors was measured using respective devices located in the downcomer below cyclones; see Fig. 2. The solids flow measured in the device below the fuel

reactor cyclone defines the internal circulation rate in the fuel reactor, i.e. solids entrained from the fuel reactor. These solids can be diverted in the LS-D in such a way that the global circulation rate in the CLC unit may be lower than the internal circulation rate in the fuel reactor. The global circulation rate defines the flow of solids being circulating from the fuel and air reactors, and it is a basic parameter to know the oxygen carrier to fuel ratio during fuel combustion in a CLC unit (Abad et al., 2007).

Experiments were carried out under similar conditions existing during coal combustion. Thus, ilmenite ($d_p=170 \mu\text{m}$; $\rho_p=3710 \text{ kg/m}^3$) was used as particulate material in the CLC unit. Ilmenite can be considered as a reference material for *i*G-CLC due to the reliability it has shown during coal combustion (Adanez et al. 2012). The gas velocity in CS was 0.35 m/s. In addition, temperature in the fuel reactor and carbon stripper was around 950 °C, while a temperature of 800 °C was in the air reactor.

Table.1: Main design parameters of the CLC unit.

Power (kW)	<i>i</i> G-CLC			CLOU		
	FR	AR	CS	FR	AR	CS
$u_{g,in}$ (m/s)	1.0	0.4	0.35	0.6	0.9	0.35
$u_{g,out}$ (m/s)	4.7	4.0	0.35	5.7	9.0	0.35
S ($\times 10^4 \text{ m}^2$) ⁽¹⁾	78/50	720/72	180	78/50	720/72	180
m_{OC} (kg)	20	55	8	20	55	8
m^*_{OC} (kg/MW _{th})	1000	2750	400	400	1100	160
ΔP (kPa)	25	15	4.5	25	15	4.5

(¹) Cross section in fuel reactor and air reactor is higher in the bottom than in the upper; thus bottom/upper sections are included

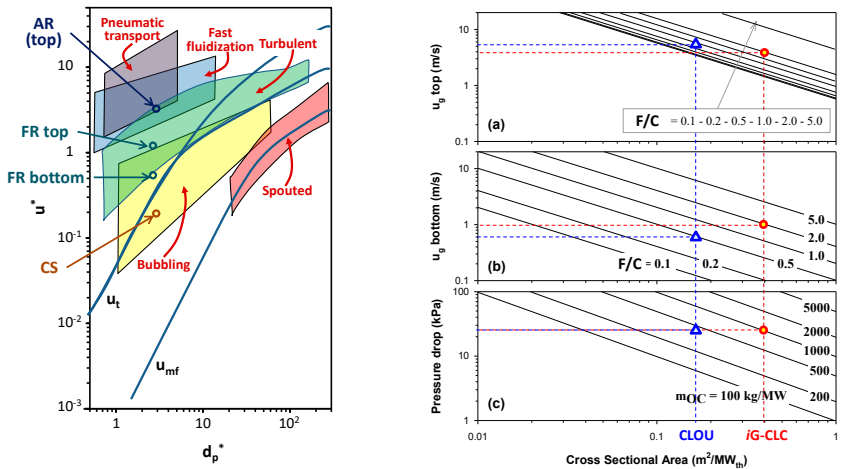


Fig. 3. Left: Fluidization regime in the flow map adapted from Bi and Grace (1995). $u^* = \text{Re}/\text{Ar}^{1/3}$; $d_p^* = \text{Ar}^{1/3}$. Right: Relation between operating conditions and design parameters as a function of the cross section in fuel reactor: (a) fluidizing gas to carbon molar ratio vs. top gas velocity; (b) fluidizing gas to carbon molar ratio vs. bottom gas velocity; (c) solids inventory vs. pressure drop. Symbols show design conditions for *i*G-CLC (●) and CLOU (▲).

RESULTS

The behavior of the 50 kW_{th} CLC unit under several operating conditions is determined with the ultimate purpose of knowing the flexibility of this unit to burn solid fuels under *i*G-CLC or CLOU units with different oxygen carriers, which could differ in its oxygen transport capacity. Most of experiments here presented are performed with no fluidization in the right-side of the LS-D, i.e. all solids recovered by the fuel reactor cyclone are sent to the carbon stripper. At this condition, the global circulation rate is similar to the internal circulation rate in the fuel reactor. Here, the global circulation flux, $G_{s,global}$, has been normalized with the section of the air reactor riser, while the internal circulation flux in the fuel reactor, $G_{s,FR}$, is calculated by

using the section of the fuel reactor riser. In addition, in general secondary gas into the fuel reactor is not used, i.e. $u_{g,sec} = 0$. Later, dedicated experiments are performed to evaluate the effect of the existence of a secondary flow or the effect of controlling the solids circulation rate by acting on the LS-D.

In order to assess the effect of the main flow in the fuel reactor on the flow and inventory of solids, the gas velocity at the fuel reactor bottom was modified in the interval $u_{g,bottom} = 0.7-1.6$ m/s. Fig. 4 shows that the global circulation flow rate increases with the gas velocity, but the pressure drop in the fuel reactor, ΔP_{FR} , decreases. Thus, an increase of the gas velocity facilitates the entrained solids from the fuel reactor. However, this method can not be used to modify the global circulation rate without changing the solids inventory in the fuel reactor, which is directly related to ΔP_{FR} . Also, as expected, changes in ΔP_{FR} have the opposite but smoother effect on the pressure drop in the air reactor, ΔP_{AR} , because of the different size of the reactors.

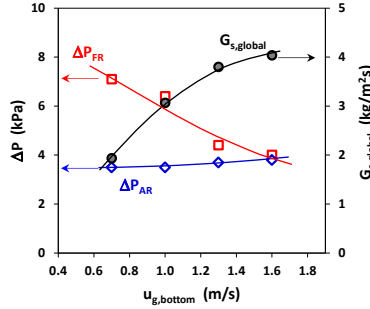


Fig. 4. Influence of the gas velocity at the fuel reactor bottom on the global circulation flux and the pressure drop in the fuel and air reactors ($u_{g,sec}=0$; $u_{g,AR}=0.9$ m/s; $m_f=92$ kg; $F_{LS-D,FR}=0.06$ Nm³/h).

Fig. 5(a) shows the effect of the secondary flow in the fuel reactor on the CLC unit performance. In this case, an increase of the secondary flow slightly affects to the global circulation rate or the pressure drop in fuel and air reactors. The relative effect of sharing a determined gas flow to fuel reactor between the bottom and the secondary flow can be evaluated by calculating a total gas velocity in the fuel reactor $u_{g,FR} = u_{g,bottom} + u_{g,sec}$; see Fig. 5(b). It can be seen that feeding the additional gas flow in the bottom of the reactor (see curve with $u_{g,sec}/u_{g,bottom}=0$) is more effective on the increase of the solids circulation rate than feeding the additional gas by the secondary flow. This fact is related to the solids distribution in the fuel reactor, which is a circulating fluidized bed in the turbulent regime; see Fig. 3. The amount of solids in the fuel reactor is low enough in order to maintain the dense bed below the inlet level of the secondary flow. Thus, the secondary flow has not influence on the entrained solids from the dense bed to the dilute region, and hence it has little influence on the global solids circulation rate. On the contrary, the flow of solids entrained from the dense bed to the dilute region could be easily reached by increasing the gas velocity from the bottom.

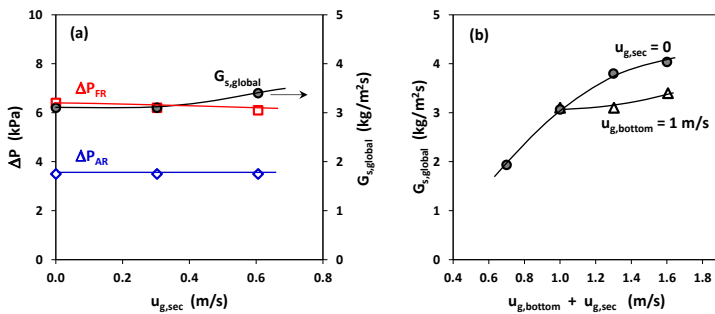


Fig. 5. (a) Effect of the secondary gas flow on the global circulation flux and the pressure drop in the fuel and air reactors ($u_{g,bottom}=1$ m/s; $u_{g,AR}=0.9$ m/s; $m_f=92$ kg; $F_{LS-D,FR}=0.06$ Nm³/h); (b) relative influence of feeding an additional gas flow by either the bottom or the secondary flow in the fuel reactor.

Fig. 6 shows that an increase in the air flow improves the solids circulation rate, but the pressure drop in the air reactor, ΔP_{AR} , is decreased. Associated with it, the pressure drop in the fuel reactor increases. Thus, the

effect of the air reactor flow variation on the air reactor behavior is similar to the changes observed in the fuel reactor when the fuel reactor main flow was modified, shown in Fig. 4. However, the effects are more attenuated than when the flow in the fuel reactor was varied. Considering the design of the air reactor compared to that of the fuel reactor, the decrease of a similar amount of solids would represent a lower decrease in the pressure drop in the wider air reactor. Note that the solids entrained from the air reactor are mainly determined by the gas velocity in the riser. So, the global circulation linearly rises until it reaches a maximum when the gas velocity is high enough to entrain all solids entering to the air reactor. As a consequence, the global circulation rate may not properly controlled by changing the air flow. In addition, the solids circulation rate could not be freely varied without modify the solids inventory in the fuel reactor, as the pressure drop in the fuel reactor is also affected.

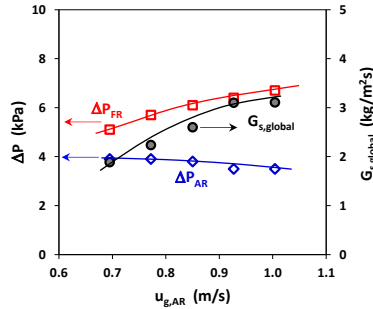


Fig. 6. Effect of the gas flow in the air reactor on the global circulation flux and the pressure drop in the fuel and air reactors ($u_{g,bottom}=1$ m/s; $u_{g,sec}=0$; $m_r=92$ kg; $F_{LS-D-FR}=0.06$ Nm³/h).

The double loop seal, LS-D, has been designed in order to modify the global circulation flow rate without modify the fluid dynamics of the fuel reactor. This device is characterized by two loop seals in parallel below the fuel reactor cyclone. Thus, the solids entrained from the fuel reactor are shared between these loop seals. The left-side loop seal is connected to the carbon stripper (LS-D-CS), and allows the solids to continue its way towards the air reactor. On the contrary, the right-side loop seal is connected to the fuel reactor (LS-D-FR). Thus, solids passing through this loop seal are internally circulated in the fuel reactor and do not count for the global circulation rate. The amount of solids entering in the internal loop can be controlled by modifying the gas velocity in the right-side loop seal, i.e. LS-D-FR. Fig. 7(a) shows that, as expected, the global circulation rate decreases as the gas flow to the LS-D-FR is increased. Virtually, all solids entrained from the fuel reactor contribute to the global circulation rate when the gas flow in LS-D-FR is low. But the fraction of solids passing to the air reactor gradually decreases as the gas flow in LS-D-FR increases; see Fig 7(b). Also is very interesting to observe that the pressure drop in the fuel reactor was barely affected, as well as the internal circulation rate. Therefore, it can be suggested that the global circulation rate can be controlled by modifying the fluidization conditions in the LS-D-FR, but fluid dynamic properties in the fuel reactor will be not affected. This fact is very relevant in order to evaluate the effect of individual operating conditions on the performance of the CLC of solid fuels, e.g. the combustion efficiency or the CO₂ capture.

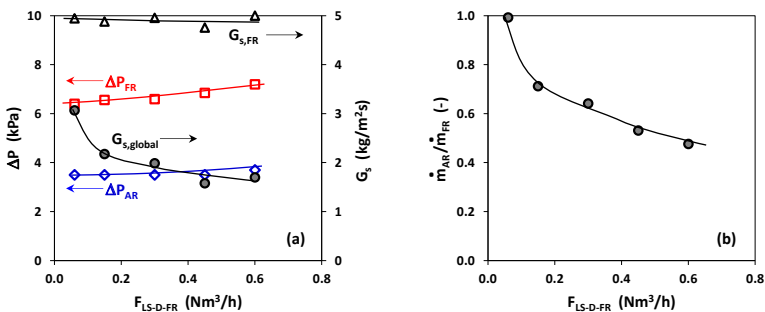


Fig. 7. Effect of the gas flow in right-side of double loop seal connected to the fuel reactor (LS-D-FR) on (a) circulation flux and the pressure drops in fuel and air reactors (note that $G_{s,global}$ and $G_{s,FR}$ are calculated using sections of the risers in the air and fuel reactors, respectively); (b) fraction of solids entrained from the fuel reactor, m_{AR}/m_{FR} , contributing to the global solids circulation rate, m_{AR} ($u_{g,bottom}=1$ m/s; $u_{g,sec}=0$; $u_{g,AR}=0.9$ m/s; $m_r=92$ kg).

The amount of oxygen carrier in the whole plant is a key factor in the performance of the CLC unit, and it affects strongly to the global circulation, as well as the pressure drop in the fuel reactor; see Fig. 8. It seems that the additional material is accumulated inside the fuel reactor, which also influences the solids circulation rate in the same trend. However, the pressure drop in the air reactor is barely affected, which could be an effect of the wider section or that the bottom part of the air reactor being full, thus no additional pressure drop could be allowable. Thus, it will be necessary to determine the required amount of solids to properly operate the CLC unit burning a solid fuel under the desired operating conditions.

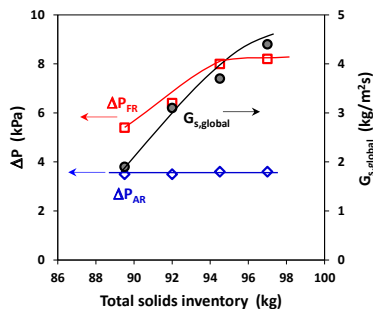


Fig. 8. Effect of the total amount of solids in the CLC unit on the global circulation flux and the pressure drop in the fuel and air reactors ($u_{g,bottom}=1$ m/s; $u_{g,sec}=0$; $u_{g,AR}=0.9$ m/s; $F_{LS-D-FR}=0.06$ Nm³/h).

From results shown above, the gas velocity at the bottom of the fuel reactor and the solids inventory in the unit can be considered the variables with higher influence on both the solids inventory in the fuel reactor and the global circulation rate. Fig. 9 shows a mapping of the solids flux and solids inventory achieved with different amount of solids in the CLC unit and the gas flow fed to the fuel reactor. The individual effects of the gas velocity in the fuel reactor and the total solids inventory was previously described; see Figs. 4 and 8, respectively. But interestingly, from Fig. 9 it can be deduced that the global solids circulation can be varied without modifying the pressure drop in the fuel reactor by selecting the proper conditions, i.e. a suitable combination of the amount of solids in the unit and the gas velocity in the fuel reactor. This strategy represents an alternative to the use of the LS-D to control the solids circulation rate; see Fig. 7. It is also observed that very high solids circulation rate could be obtained by increasing the gas velocity in the fuel reactor, but the amount of solids in the CLC unit should be increased in order to maintain the solids inventory in the fuel reactor.

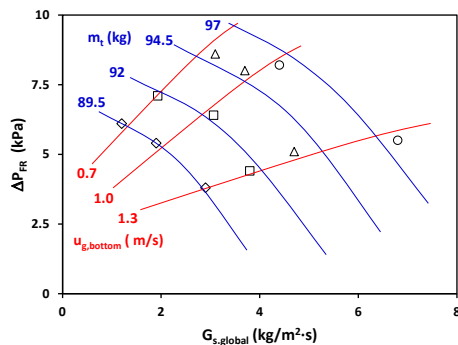


Fig. 9. Mapping of global solids flux and pressure drop in the fuel reactor as a function of the gas flow in the fuel reactor and solids in the whole CLC unit ($u_{g,sec}=0$; $u_{g,AR}=0.9$ m/s; $F_{LS-D-FR}=0.06$ Nm³/h).

CONCLUSIONS

A parametric analysis of the operating conditions on the performance of a 50 kW_{th} CLC unit has been carried out in order to determine the flexibility of this unit to individually modify the solids inventory in the fuel reactor, m_{FR} , and the solids circulation rate. The solids circulation rate, $G_{s,global}$, could be modified in a wide interval by varying the gas velocity at the bottom of the fuel reactor, $u_{g,bottom}$, and the amount of solids in the whole unit, m_t , but in this case m_{FR} was also changed. In order to vary $G_{s,global}$ but maintaining m_{FR} unchanged two different strategies have been identified: (1) the selection of suitable values of both parameters ($u_{g,bottom}$

and m_i), which must be varied at the same time; and (2) the use of the double loop seal included in the CLC unit as a useful device to vary $G_{s,global}$ without modifying the fluid dynamics of the fuel reactor. Following this study, a parametric study on the effect of each individual operating condition on the performance of the CLC of solid fuels can be done.

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