Impact of oxygen transport on char conversion in dual fluidized bed systems

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Abstract-

In the context of thermochemical conversion of solid fuels in dual fluidized bed (DFB) reactors, oxygen carrier bed materials have been found to enhance the rate of char gasification [1, 2]. Char gasification is typically the rate limiting step in the conversion process [2, 3], which plays a role on the performance of the overall process and constrains the design of the reactor. The quantity of oxygen carried by the bed material in relation to the fuel feeding rate is likely to influence char conversion. This parameter is commonly characterized in literature by the oxygen-carrier-to-fuel ratio (ϕ), and it can be adjusted by either varying the fuel feed and/or the circulation rate of bed material in the DFB. The residence time of the fuel is typically altered by these changes, which results in an unclear role of the oxygen-carrier-to fuel ratio on the final char conversion. Mendiara et al reported that char conversion was not significantly affected by changes to fuel flow for ϕ = 1.1-2.4, and the bed circulation rate showed some influence only at ϕ <1 [3, 4].

The present work investigates the impact of oxygen transport on char conversion in the 2-4 MW_{th} DFB gasifier at Chalmers University of Technology. An iron ore is used as oxygen carrier and steam as fluidizing agent. A parametric study is conducted, in which a wide range of oxygen-carrier-to-fuel ratio (ϕ =3-12) are achieved by varying fuel flow and bed material circulation. Char conversion is calculated on the basis of gas analysis and mass balance calculations. The results confirm that oxygen transport enhances char conversion, in line with previous research in smaller units. Both fuel feed and circulation of bed material had an impact on the final char conversion under the conditions tested, which differs from the results by [3, 4]. For a given residence time of the fuel particles in the reactor, char conversion was found proportional to the oxygen-carrier-to-fuel ratio.

1. Introduction

Dual fluidized bed (DFB) systems are a type of reactor applied to a number of thermo-chemical processes, such as Chemical Looping Combustion (CLC), Chemical Looping Reforming (CLR) and Indirect Gasification (IG). In DFB systems two fluidized beds are interconnected and a bed material circulates between them transporting heat and, in some cases, chemical species. The bed material is exposed to different conditions in the two vessels. Typically, one reactor is fluidized by air resulting in an oxidizing atmosphere, and the second reactor has a more reducing environment created by fuel. Depending on the process at hand, the two vessels fulfil different functions. When an oxygen carrier (OC) is used as bed material, a phenomena known as oxygen transport occurs. Figure 1 summarizes the flows of oxygen carrier in a DFB reactor, where the material oxidizes in the combustor or air reactor, and it transports oxygen to the fuel reactor in the form of oxidized material. Oxygen transport is the basis of CLC, which aims at oxidizing the fuel with the oxygen supplied by the oxygen carrier particles producing a flue gas stream free of N₂.

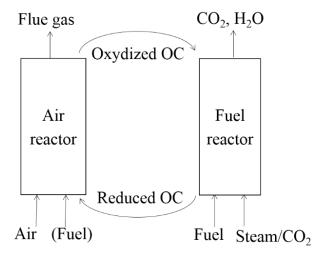


Figure 1. Basic scheme of a dual fluidized bed system with an oxygen carrying bed material.

Chemical Looping Combustion was initially demonstrated for gaseous fuels [5], and more recently for coal [6] and biomass [4]. The latter fuel becomes an interesting option as it would lead to negative net CO₂ emission. Chemical Looping Combustion of solids fuels is typically referred to as *in situ* gasification CLC (iG-CLC) owing to the conversion steps taking place in the fuel reactor. As the fuel heats up in the fuel reactor it dries and releases the volatile matter (Reaction 1), which is then combusted in contact with the oxygen carrier in a heterogeneous gas-solid reaction (Reaction 2). The remaining char is gasified with the gasification agent according to Reaction 3, and the resulting gasification products are oxidized by a gas-solid reaction with the oxygen carrier (Reaction 4) [7]. Char gasification has been identified as the rate limiting step of the overall conversion process [2, 3], therefore it becomes a key step that determines the performance of the unit.

Volatile matter +
$$MeO \rightarrow CO_2 + H_2O + Me$$
 {2}

$$Char + Gasification \ agent \ (H_2O, CO_2) \rightarrow Gasification \ products + ash$$
 {3}

Gasification products +
$$MeO \rightarrow CO_2 + H_2O + Me$$
 {4}

Oxygen carrying materials have shown to enhance the rate of char gasification at laboratory [1] and pilot [2] scales. It has been suggested that this is the result of the lower concentration of inhibiting species around the char particle in the presence of the oxygen carrier compared to an homologous case with inert bed material [7, 8]. Hydrogen [8-10], and more generally volatile species [11] have been proven to inhibit the steam gasification reaction of both coal and biomass derived char. A recent work concerning woody biomass showed that tar compounds are also inhibiting species, which was investigated by using levoglucosan vapour as model tar [12]. The concentration of inhibiting species in the fuel reactor depends on a number of factors such as the content of oxygen carrier in the bed material [13], type of oxygen carrier [14], and the operating conditions [15].

A set of operating conditions results in an oxygen-carrier-to-fuel ratio (ϕ), which defines the oxygen transported by the bed material as a fraction of the stoichiometric oxygen for full combustion of the fuel. When oxygen transport is undesired (e.g. indirect gasifiers and CLR units) the oxygen-carrier-to-fuel ratio is kept below unity. Contrarily, CLC reactors operate above stoichiometric conditions (ϕ >1) to ensure high degree of fuel combustion. Given the bed material and fuel type, the oxygen-carrier-to-fuel ratio can be adjusted by either varying the fuel feeding rate or the circulation rate of bed material. The impact of these two variables (i.e. fuel and bed material flow) on the performance of the process becomes additionally complex when solid fuels are used, since both oxygen-carrier-to-fuel ratio (ϕ) and the residence time of the fuel particles (τ_{fuel}) are altered. Figure 2 exemplifies the relation between τ_{fuel} and ϕ as bed material circulation rate increases for a perfect mix case and negligible concentration of fuel particles compared to the bed material.

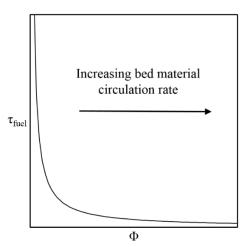


Figure 2. Qualitative relation between bed material circulation rate, residence time of the fuel particles and oxygen-carrier-to-fuel ratio, assuming perfect mix and negligible concentration of fuel particles compared to bed material particles.

The effect of fuel feeding rate and circulation rate on CLC of solid fuels was investigated by Mendiara et al. in a $500W_{th}$ unit using coal as fuel and bauxite waste as oxygen carrier material. Increasing fuel flow corresponding to ϕ between 1.1 and 2.4 did not have a significant impact on char conversion, which was attributed to the small changes in residence time of the char within the range of operation tested. Increasing bed material circulation rate, on the other hand, resulted in lower char conversion in line with the changes induced in the fuel residence time. The trend was attenuated above stoichiometric conditions [3]. The weak correlation between bed material flow and char conversion above stoichiometric conditions (ϕ >1) was also observed with biomass fuel and an iron ore as bed material in the same unit [4].

The purpose of the present work is to investigate experimentally the impact of oxygen transport on char conversion under conditions relevant to industrial applications. The experimental work is conducted in the 2-4 MWth DFB gasifier at Chalmers University of Technology, using a natural titanium iron ore as bed material and steam is the fluidizing agent. The reactor has onbed fuel feeding and commercial wood pellets are used as fuel. Different levels of oxygen-carrier-to-fuel ratio (ϕ =3-12) are induced in the gasification reactor by varying fuel flow and bed material circulation rate, alternatively. Char conversion is calculated based on gas analysis and mass balance calculations.

2. Theory

The oxygen carrying capacity (R_o) of the bed material is defined as the maximum oxygen exchange that the oxygen carrier can experience during a redox cycles. It is a property of the oxygen carrier and it is defined as:

$$R_o = \frac{m_{ox} - m_{red}}{m_{ox}} \tag{1}$$

where m_{ox} and m_{red} refers to the mass of the fully oxidized and reduced material respectively. Common titanium iron ores used in CLC research have a theoretical oxygen carrying capacity of 5%, which corresponds to the reduction from Fe₂TiO₅+ TiO₂ to FeTiO₃ of a material with Fe:Ti ratio of 1:1 [16]. In practice, the oxygen carrying capacity depends on the number of redox cycles and pre-treatment of the material and, consequently, a range of values have been reported in literature typically from 2.1 to 4% [17, 18]. Once R_o and the bed material circulation rate is set (\dot{m}_{bed}), the oxygen carrier-to-fuel ratio (ϕ) can be calculated as:

$$\varphi = \frac{\dot{m}_{bed} \cdot R_o}{\dot{m}_F \cdot \Omega_F} \tag{2}$$

The denominator corresponds to the stoichiometric oxygen for complete combustion of the fuel, where \dot{m}_F is the mass flow of fuel and Ω_F is a stoichiometric coefficient for the combustion reaction.

The residence time of the fuel in the reactor (τ_{fuel}) is calculated by means of the dispersion coefficient (D) according to Eq. 3, where L is a characteristic length of the reactor. This definition is considered suitable for the reactor configuration used. In particular, the relatively large size of the reactor together with on-bed fuel feeding system and large fuel particles (i.e.

commercial wood pellets), makes the assumption of perfect mixing of the fuel and the bed material questionable.

$$\tau_{fuel} = \frac{L^2}{2 \cdot D} \tag{3}$$

The dispersion coefficient was derived experimentally in a separate work for the pair wood pellet/iron ore in a wide range of operating conditions covering the ones used in the present investigation [19]. Theoretically, the dispersion coefficient is not dependent on the fuel flow when the concentration of fuel particles is low, as it is the case in the present work (i.e. <4% fuel particles in the inventory). This leads to one single dispersion coefficient for all cases with the same bed material circulation rate regardless the fuel feeding rate.

The degree of char conversion (X_{char}) in the fuel side of the DFB can be derived from the carbon balance throughout the reactor. Char conversion is defined in Eq. 4, where Y_{char}^F , Y_{c}^F , and $\dot{m}_{C,gas}$ refer to the char content of the fuel, carbon content in the fuel and the total carbon measured in the gas, respectively. For simplification, char is assumed to contain only carbon.

$$X_{char} = \frac{\dot{m}_F \cdot Y_{char}^F - (\dot{m}_F \cdot Y_C^F - \dot{m}_{C,gas})}{\dot{m}_F \cdot Y_{char}^F}$$
(4)

Note that this definition is only applicable if the fuel is fully devolatilized, i.e. it will yield negative char conversion if the total carbon measured at the exit of the fuel reactor $(\dot{m}_{C,gas})$ is not enough to justify full devolatilization of the fuel.

3. Experimental

The experiments were conducted in the $2\text{-}4\text{MW}_{th}$ dual fluidized bed gasifier at Chalmers University of Technology. The unit consists of two interconnected vessels: the gasifier section as bubbling bed, and the combustor as circulating fluidized bed. The bed material is oxidized in the combustion side in contact with air, thereafter, it flows into the gasification section via a loop seal (LS1). The system boundary for the carbon balance calculations are set around the gasification unit, which is schematized in Fig. 3. The fuel is fed on the top of the bed and the produced gas leaves the reactor via the raw gas line. The bed material together with the unconverted fuel returns to the combustor via LS2. The gasifier and the loop seals are fluidized with steam. Further details about the gasification and combustion section can be found in [20] and [21], respectively.

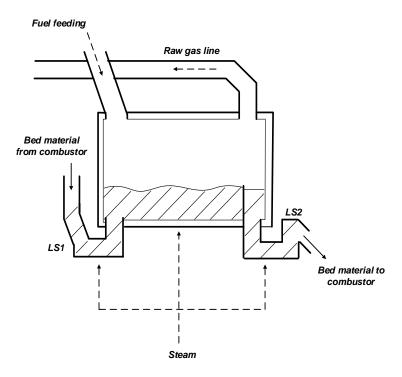


Figure 3. Schematic of the gasification vessel and main flows in and out of the reactor.

The gasifier is fed continuously with pellets of the composition detailed in Table 1. The quantification of total carbon in the gas is accomplished by means of a high temperature reactor (HTR), where a slip stream of produced gas is heated up to 1700° C and all carbon containing species decompose into CO and CO₂. The technique has been described and validated in [22]. The gas at the exit of the HTR is cooled down and measured by a micro gas-cromatograph (micro-GC, Varian Model CP4900) equipped with a Poraplot Q and MS5A columns, using He and Ar as carrier gas respectively.

Table 1. Fuel composition.

Ultimate Analysis		Proximate Analysis		
C (% mass, daf)	50.30	Moisture (% mass, as received)	8.6	
H (%mass, daf)	6.22	Ash (%mass, dry) 0.4		
O (% mass, daf)	43.17	Volatile content (% mass dry) 80.8		
		Char (% mass dry)	18.8	

The bed material is a natural iron ore from Australia with an Fe:Ti ratio of approximately 1:1, and its oxygen carrying capacity was determined experimentally. A bed material sample was collected from LS1 (See Figure 3) and tested in a lab scale reactor at similar operating temperature to that used during the gasifier experiments, i.e. 820°C. The sample (i.e. 15g) was reduced by exposure to a stream of synthetic gas consisting of 50% vol H₂ and 50% vol CO. The reduced material was then exposed to a controlled flow of synthetic air until full oxidation was reached. The composition of the gas stream at the exit of the reactor was measured online, which allows for quantification of the oxygen uptake by the material and, thereby, its oxygen

carrying capacity (Ro). Detailed description of the experimental setup and procedure is provided elsewhere [16]. The experimental $R_{\rm o}$ obtained by this method was 10%, which is significantly higher than the theoretical 5% and it remains to be confirmed. Oxygen carrying capacity above 5% was also reported by Bidwe et al. [23] when testing a similar Australian ore and it was suggested that some of the iron was probably reduced to metallic iron. In the present work, the even higher oxygen carrying capacity than the measured by Bidwe et al. could be related to two factors: full reduction to metallic iron, and also to the history of the particles investigated, which were exposed to biomass ash and oxidizing and reducing atmosphere during several weeks in the DFB unit.

The bed material circulation rate is measured experimentally according to [20], by monitoring the pressure drop in the combustor, while the return of bed material to the boiler (LS2) is temporarily interrupted. Table 2 summarizes the tests conducted and main operational parameters. Several levels of oxygen-to-fuel ratio (ϕ) were reproduced by changing fuel and bed material flow sequentially. The experiments were carried out in two series: high and low solids flow respectively. In each series fluidization velocity remains constant and fuel flow is varied between 200-400 kg/h stepwise. The temperature of the bed was kept as similar as possible to allow comparison between tests.

Solids (ton/h)	circulation	Fuel flow (kg/h)	Average temperature of the bed material (°C)	Steam flow (kg/h)	Case
High	20	200	825	200	Low fuel
	20	300	827	200	Middle fuel
	20	400	828	200	High fuel
Low	10	200	816	200	Low fuel
	10	300	818	200	Middle fuel
	10	400	821	200	High fuel

Table 2. Experimental matrix.

4. Results

The relation between bed material circulation rate, residence time of the fuel (τ_{fuel}), and oxygen-carrier-to-fuel ratio (ϕ) is summarized in Fig. 4 for the operating conditions investigated. As the circulation rate of the bed material increases the fuel residence becomes shorter, resulting in a difference of 2 min in residence time between the cases at high and low circulation rate investigated. Once the circulation rate is fixed, the oxygen-carrier-to-fuel ratio depends on the fuel flow.

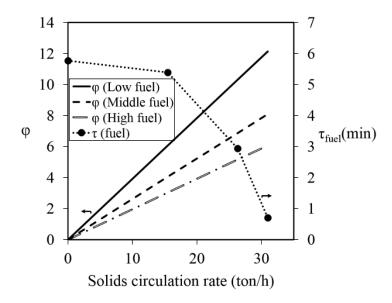


Figure 4. Relation between circulation rate of bed material, oxygen-carrier-to-fuel ratio and residence time of the fuel in the range of operating conditions tested.

The total carbon measured in the gas with the High Temperature Reactor is summarized in Fig. 5 as a function of the estimated oxygen-carrier-to-fuel ratio. The shaded area indicates the carbon conversion that corresponds to devolatilization only. The upper and lower boundary of the area corresponds to the carbon content of volatiles according to the standard proximate analysis and to a pyrolysis experiment of a similar fuel reported in [20], respectively. As shown in Figure 5, all measured points fall above the shaded area indicating that the fuel is fully devolatilized.

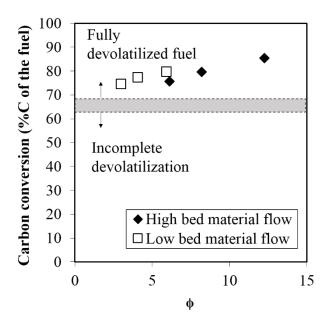


Figure 5. Carbon conversion as percentage of the carbon in the fuel measured in the gas. Experimental data for the two series (i.e. high and low bed material flow) as a function of the estimated oxygen-carrier-to-fuel ratio.

Figure 6 summarizes the results for char conversion as a function of the oxygen-carrier-to-fuel ratio. Both fuel and bed material flow show an impact on char conversion within the range of operating conditions tested. Higher fuel flow results in a decrease of char conversion, while higher bed material flow results in higher char conversion. In both experimental series (i.e. high and low bed circulation), char conversion is found proportional to the oxygen-carrier-to-fuel ratio (ϕ). In the same line, an increase of ϕ induced by a larger solids circulation rate also results in higher degree of char conversion (See dash lines in Fig. 6).

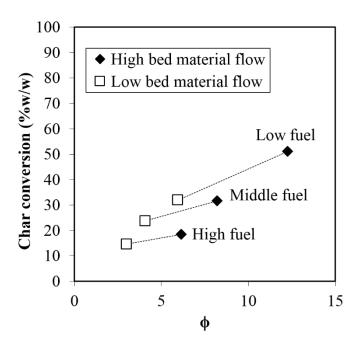


Figure 6. Char conversion as a function of oxygen-carrier-to-fuel ratio.

5. Discussion

The consistent respond of char conversion to changes in ϕ (See Fig. 6) suggests that the rate of char gasification is enhanced by the oxidized oxygen carrier, which is in line with previous findings by [1] and [2]. Accordingly, the higher the oxygen transported by the bed material, the larger the fraction of converted char. In fact, the total carbon conversion in Fig. 5 can be compared to experimental data reported by Israelsson et al., where a common reference material (i.e. quartz sand) was used as bed material in the same unit [24]. The authors closed the carbon balance with the same method used in this work, and found carbon conversion below 70% for similar fuel and operating conditions. Their measurement indicate that the product gas originated from devolatilization products mainly and very limited char was converted, whereas significant degree of char conversion (i.e.14-51%) is achieved in the present work with an oxygen carrier bed material.

An increase of bed material circulation rate results in a lower residence time of the fuel as shown in Fig. 4, which would be expected to yield a lower degree of char conversion. Yet, the results suggest that the shorter residence time is overcompensated by the higher availability of oxygen in the reactor (i.e. dash lines in Fig. 6). This trend differs from previous observations by Mendiara et al. [3, 4], who found little impact of the solids circulation rate on char conversion

above stoichiometric conditions (ϕ >1). In general, the authors observed a slight decrease of char conversion as the circulation rate increases, which was attributed to the lower residence time of the char particles as the solids flow increases [3]. The lack of agreement between this work and the reported data could be related to differences in the geometry of the two reactors, which in turn determines the possible combinations of residence time and oxygen-carrier-to-fuel ratio that yield a given degree of char conversion.

Note that despite char conversion responds consistently to changes in ϕ , the two cases with the same oxygen-carrier-to-fuel ratio (ϕ =6) yield significantly different degree of char conversion. Such difference is in line with the changes in residence time that follows a variation of the solids circulation rate. Accordingly, the lower residence time in the case of larger bed material flow (i.e. solid symbol at ϕ =6 in Fig. 6) results in char conversion 13% units lower than its counterpart at lower solids flow (i.e. empty symbol at ϕ =6 in Fig. 6).

Finally, the sensitivity of char conversion to changes in bed material circulation rate varies with fuel flow as observed in the slopes of the dash lines in Fig. 6. At lower fuel flows the influence of bed material circulation is significant, whereas at higher fuel flows this effect is attenuated. It is also visible that variations of fuel flow yield larger changes in char conversion at high bed material circulation rate (Compare High and Low bed material flow in Fig. 6). Overall, these trends suggest that as ϕ increases, the response of char conversion to changes in both fuel and bed material flows becomes stronger. This can be related to the decreasing reactivity of the oxygen carrier towards the combustible gases as the oxygen in the material is depleted [23, 25]. Higher ϕ can be interpreted as higher net reactivity of the bed inventory owing to the higher average oxidation state of the oxygen carrier. At higher average oxidation state (i.e. higher ϕ) the reaction rate of the bed material with the gasification products becomes faster [23, 25] ensuring that the gasification products are rapidly removed from the vicinity of the char particle. The char particle is then left readily available for the steam gasification reaction with lower concentration of inhibiting species in the surrounding gas as suggested in previous works related to CLC of solid fuels [1, 7, 8].

6. Conclusions

The impact of oxygen transport on char conversion is investigated experimentally in a 2-4MWth dual fluidized bed gasifier using an oxygen carrier as bed material. Char conversion was quantified under different operation conditions corresponding to oxygen-carrier-to-fuel ratio ranging from 3 to 12, by varying fuel fed and solids circulation rate. The results confirm that the oxygen carrier enhances char gasification rate. In terms of operating conditions, both fuel feed and circulation of bed material showed significant impact on the final char conversion under the conditions tested. The degree of char conversion is the result of the combined effect of residence time and oxygen-carrier-to fuel ratio. For a given residence time, the degree of char conversion was found proportional to the oxygen-carrier-to-fuel ratio.

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Notation

Ro	oxygen carrying capacity [kg O/kg]	$ au_{fuel}$	residence time of the fuel [s]
m_{ox}	mass oxidized material [kg]	L	characteristic length [m]
m_{red}	mass reduced material [kg]	D	dispersion coefficient [m²/s]
φ	oxygen-carrier-to-fuel ratio [kg/kg]	X_{char}	char conversion [kg/kg]
\dot{m}_{bed}	bed material circulation rate [kg/s]	Y_{char}^F	char content of the fuel [kg/kg fuel]
\dot{m}_F	fuel feeding rate [kg/s]	Y_C^F	carbon content of the fuel [kg/kg fuel]
Ω_F	stoichiometric oxygen [kg/kg fuel]	$\dot{m}_{C,gas}$	flow of carbon with the gas [kg/s]

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