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THE INFLUENCE OF CARBON STRIPPER EFFICIENCY ON CO₂-CAPTURE RATE IN A CHEMICAL-LOOPING COMBUSTION PROCESS FOR SOLID FUELS

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ABSTRACT

In the present work a Chemical-Looping Combustion process for solid fuels is simulated on the 100 MW_{th} scale. The coal is gasified inside the fuel reactor by recirculated CO₂ and H₂O. A carbon stripper downstream of the fuel reactor is used to reduce the carryover of char from the fuel to the air reactor. The influence of the carbon stripper on the CO₂ capture rate is investigated. The results demonstrate the significance of the carbon stripper in this process.

INTRODUCTION

Chemical-Looping Combustion (CLC) is an interesting variant for the inherent separation of carbon dioxide inside a power generation process. It has recently attracted much attention by numerous research groups (e.g. 1-4). Most research focuses on the realization of CLC in two interconnected fluidized bed reactors. In between these two reactors solids are circulated which transport chemically bound oxygen taken up from the air inside the air reactor to the so called fuel reactor. The oxygen carrier (OC) particles provide the fossil fuel (e.g. natural gas or coal) with oxygen and will themselves be reduced. Reduced oxygen carrier particles are then cycled back towards the air reactor for re-oxidation. CLC has the advantage that the carbon dioxide of the off-gas will not be diluted by nitrogen. After condensation of water, almost pure carbon dioxide can be obtained and transported to its designated storage location.

If solid fuels are to be used in CLC, the complexity increases compared to gaseous fuels. A scheme for a solid fuels CLC process is shown in Figure 1. The direct reaction between the solid carbon and the oxygen bound to the solid oxygen carrier particles will not proceed at a significant rate. Thus the solid carbon has to be gasified. In CLC it is self-evident to recycle the fuel reactor off-gases consisting of H₂O and CO₂ to use them as gasifying agents. During carbon gasification H₂ and CO will be produced. The OC particles will oxidize these intermediate products further towards H₂O and CO₂. Gasification is a rather slow process compared to the reaction of CO and H₂ with the OC (5) and thus sufficient residence time of the coal particles in the fuel reactor must be

provided. On the other hand to yield a sufficient flow of oxygen for fuel oxidation a rather large circulation flow of OC particles is needed. The solids flow leaving the fuel reactor consists of a mixture of unreacted char particles, ash and oxygen carrier particles. The char particles must not enter the air reactor since they would combust with the air-oxygen present. The formed CO_2 would not be captured and thus decrease the CO_2 capture rate of the plant. To reduce the amount of carbon slip towards the air reactor a carbon stripper can be used. The OC particles and the char particles can be separated according to their terminal settling velocity.

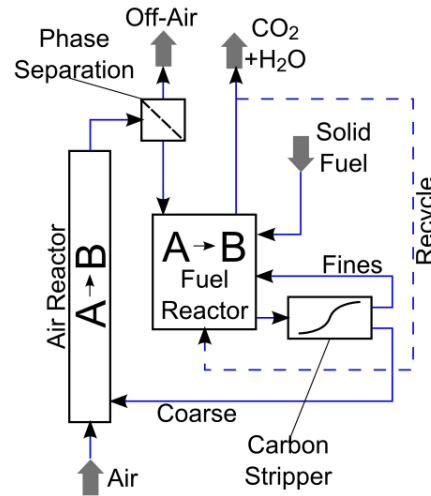


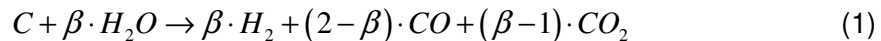
Figure 1: Scheme of a CLC process for solid fuels

The aim of this investigation is to investigate how the CO_2 capture rate is influenced by the carbon stripper.

THEORY

Char Gasification

In CLC both CO_2 and H_2O can be used for gasification of the coal char. It is repeatedly reported in literature that steam gasification proceeds at a higher rate than carbon dioxide gasification (e.g. [6](#)). The net gasification reactions are shown in the following equations:



The factor β in equation (1) was introduced by Matsui et al. ([7](#)) and summarizes the following two reactions:



For this investigation β is set to 1.2 according to ([7](#)). A set of kinetic equations was chosen that describes the gasification of coal char by CO_2 and H_2O ([7,8](#)).

Fuel Reactor Model

The fuel reactor is a bubbling fluidized bed, where the solid phase is assumed to be ideally mixed. It was stated above that the fuel reactor should be fluidized by its own re-circulated off-gas. Therefore the gas composition at steady-state is the same at the inlet and the outlet of the fuel reactor. Accordingly the fuel reactor can be described reasonably well by continuous stirred tank reactor characteristics. A corresponding model has been set up that is able to handle multiple reactions and considers changes in volume flow due to reaction. The release of volatiles is assumed to occur

instantaneously upon fuel introduction in the reactor. The composition of the volatiles is calculated according to the model of Jensen (9). The model has been slightly modified in order to neglect the formation of nitrous oxides and the sulfur content of the fuel. All fuel nitrogen is therefore released as gaseous nitrogen. It is assumed that the particle size distribution (PSD) of the char does not change during the initial devolatilization process. During gasification a size reduction of the char particles is considered. It is assumed that shrinkage is a process of external surface reaction:

$$\frac{dR}{dt} = -k_s \left[\frac{m}{s} \right] \quad (5)$$

The PSD of the char at the exit of the fuel reactor is then calculated according to Levenspiel (10). The reaction of volatiles and the gasification products with oxygen carrier particles is usually much faster than the gasification of char. Accordingly the concentrations of the aforementioned gases in the fuel reactor are set to zero due to their reaction with oxygen carrier particles.

Carbon Stripper Model

The carbon stripper is simulated as a classifier. The grade efficiency is described by the Rogers expression (11), which has been adapted for usage with settling velocities instead of particle diameters and to neglect bypass of fines:

$$G(u_{t,i}) = \frac{1}{1 + \left(\frac{u_{t,50}}{u_{t,i}} \right) \cdot e^{\alpha \cdot \left(1 - \left(\frac{u_{t,i}}{u_{t,50}} \right)^3 \right)}}, \quad (6)$$

with the grade efficiency $G(u_{t,i})$, defined as the ratio of mass fraction of the particle settling velocity interval i in the coarse product and the mass fraction of the same interval in the feed. The parameter $u_{t,50}$ designates the cut terminal velocity (50 %-value of the grade efficiency curve) and $u_{t,i}$ is the terminal settling velocity of the particles in size interval i . The sharpness of the separation is defined by α , which can vary between 0.3 (diffuse separation) and 6.6 (analysis-sharp separation). A more common description of the separation sharpness is (12):

$$\chi = \frac{u_{t,25}}{u_{t,75}}, \quad (7)$$

where $u_{t,25}$ and $u_{t,75}$ are the terminal velocities that belong to the values of the grade efficiency curve at $G(u_{t,i}) = 0.25$ and 0.75 , respectively. An ideal separation would have a value of $\chi = 1$ (usual technical sharpness: $0.3 < \chi < 0.6$, technically sharp: $0.6 < \chi < 0.8$, analysis sharp: $0.8 < \chi < 0.9$ according to 12). Reactions are not considered in the carbon stripper.

Simulation Environment

The simulations have been carried out in SolidSim (13), a steady-state flowsheet simulation environment for solids processes.

RESULTS AND DISCUSSION

Test Case

The flowsheet of the test case is shown in Figure 2. To prevent unburnt char from entering the air reactor a carbon stripper (sifter) is located downstream of the fuel reactor. This device separates the mixture of OC and char by differences in settling velocity. It is desired to facilitate this separation as much as possible. Coal particles will naturally decrease in size during combustion and therefore it is chosen to grind the coal fine and have larger oxygen carrier particles in comparison. This strategy is reflected by the flowsheet. The stream of fine particles leaving the carbon stripper is reintroduced to the fuel reactor while the OC rich stream of coarse particles is transported to the air reactor. The feed denoted by OC-refill is necessary to ensure that the target circulation flow of OC particles can be reached during the iterative solution procedure.

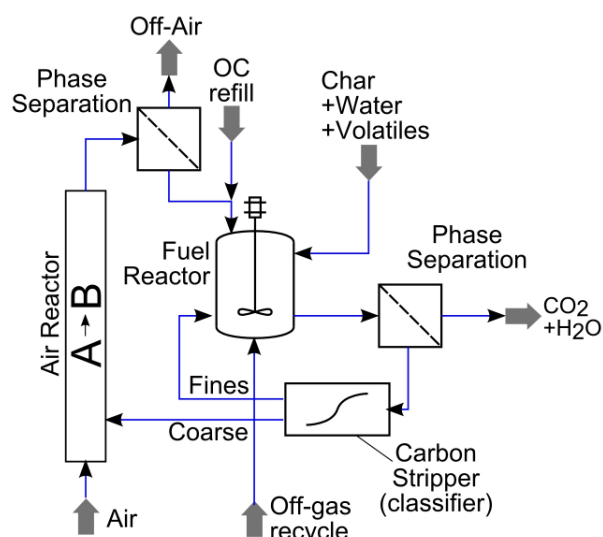


Figure 2: Simulated flowsheet of the CLC process for solid fuels.

As fuel a Columbian anthracite coal from the El Cerrejon mine has been selected. Proximate and ultimate analysis results are shown in Table 1 and Table 2.

Table 1: proximate analysis results of coal ‘El Cerrejon’ (14)

LHV [MJ/kg] (waf)	Water [wt.-%] (raw)	Ash [wt.-%] (wf)	Volatiles [wt.-%] (waf)
31.98	15.39	10.3	41.9

Table 2: ultimate analysis results of coal ‘El Cerrejon’ (14)

C [wt.-%] (waf)	H [wt.-%] (waf)	O [wt.-%] (waf)	N [wt.-%] (waf)	S [wt.-%] (waf)
81.0	6.01	10.70	1.50	0.79

El Cerrejon coal char has an apparent density of 1500 kg/m³ (14). The total flow of coal should represent a fuel input of 100 MW_{th} at complete combustion. Dividing the coal feed into separate flows 1.528 kg/s char, 0.536 kg/s H₂O, 0.780 kg/s CH₄, 0.567 kg/s CO, 0.126 kg/s N₂ and 0.036 kg/s CO₂ are fed to the fuel reactor (sulfur content is neglected). The fate of the ash is not tracked in this investigation. The initial PSD of the coal is the same as a state of the art coal mill for pulverized coal boilers delivers (15).

A copper based OC was selected that consists of 10 wt.-% of active CuO. The inert phase is Puralox NWA155 from Sasol, Germany, which is a porous aluminum oxide of medium size and a possible inert support for impregnation (16,17). Since the particle size distribution of synthetic OC prepared by impregnation is defined by the support particles, the particle size distribution of the Puralox (measured by laser diffraction) was chosen for the OC particles of this investigation. The apparent density is taken to be 1800 kg/m³ for the oxidized state. The particle size distributions of char and OC are shown in Figure 3. It can be observed, that there is a certain overlap of the two distributions. The difference in particle density is not very large, thus the settling velocity distributions of the two solids in Figure 4 shows a similar overlap. Figure 4 shows only a small part of the distribution of OC particles. At 8 m/s the distribution approaches finally the value 1. The settling velocity distributions are calculated for fuel reactor conditions. According to Figure 4 the cut separation velocity should be chosen between 0.2 m/s and 1.0 m/s.

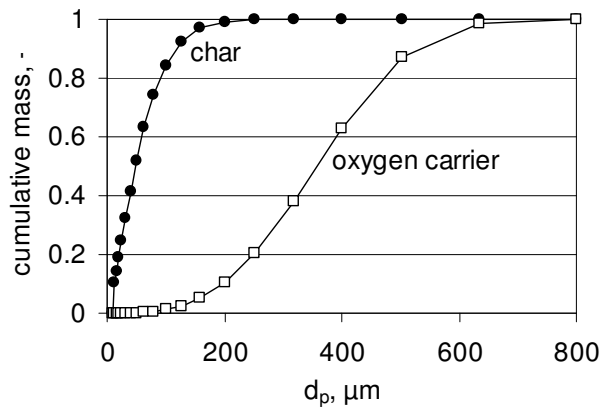


Figure 3: Particle size distributions of char (dots) and oxygen carrier particles (empty squares).

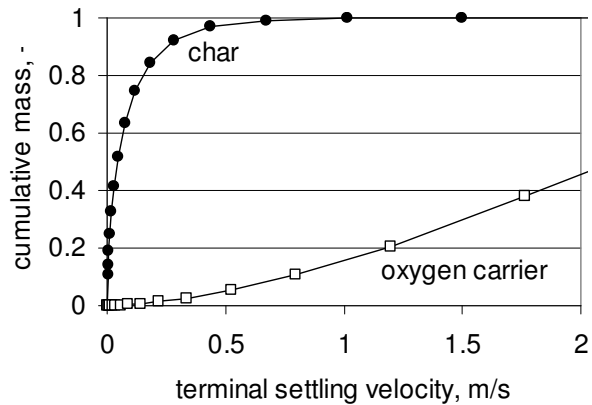


Figure 4: Settling velocity distributions of char (dots) and oxygen carrier particles (empty squares) with marked area of overlap at fuel reactor conditions.

Fuel reactor operation is carried out at 900°C and the solids entering the fuel reactor have a residence time of 240 s (18,19). For fluidization and gasification a mixture of steam and carbon dioxide is fed to the fuel reactor. The CO₂ fraction is assumed as 74 % by weight which would correspond to complete gasification of the char. For the re-oxidation of the OC particles a global excess air ratio of 1.2 is assumed. The circulation mass flow of OC particles is 650 kg/s.

Simulation results

Simulation results are compared on CO₂ capture rate (CCR) basis:

$$CCR = \frac{\text{CO}_2 \text{ flow from fuel reactor}}{\text{total CO}_2 \text{ flow based on fuel input}} \quad (8)$$

The CCR is decreased by CO₂ leaving the process through the air reactor. This is the case, when char is transported to the air reactor where it will combust with air oxygen. In order to minimize the flow of char from the fuel reactor to the air reactor a sifter / carbon stripper was introduced in-between both reactors (Figure 2). The carbon stripper divides the mixed solids flow into a flow of fines and coarse. Because of the aforementioned overlap of the settling velocity distributions the separation of the two types of solids will never return two pure streams of only one species. It is possible though to minimize the flow of char within the stream of coarse particles flowing to the air reactor. This can be achieved by the choice of a high cut velocity. Figure 5 shows the result of a variation of the cut velocity in terms of char flow in the coarse flow for a separation sharpness of $\alpha = 0.84$ which corresponds to $\chi = 0.5$. The flow of char in the coarse flow from the carbon stripper decreases with increasing cut velocity. Increasing the cut velocity does though also increase the flow of OC particles in the flow of fines that is returned to the fuel reactor. If this flow increases, the carbon stripper will have to handle a larger flow of mixed solids. This can generally only be achieved by a larger unit. In Figure 5 the flow entering the carbon stripper unit is shown on the secondary axis. For 3 m/s for instance it can be observed that the flow entering the sifter unit reaches approximately 2500 kg/s which is 3.8 times the flow of OC particles circulating between the reactors.

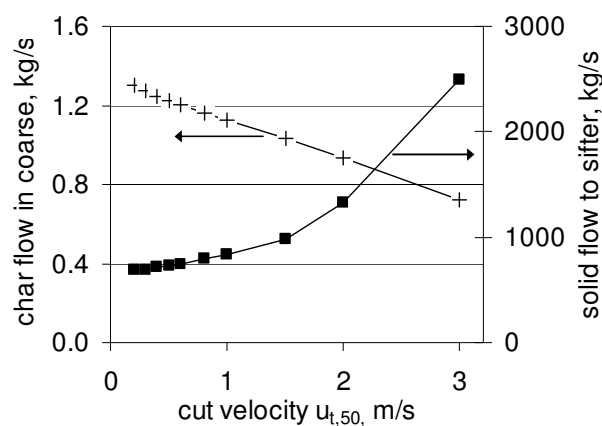


Figure 5: Char flow towards the air reactor in dependence of the cut terminal settling velocity and the corresponding total flow of solids entering the sifter unit ($\chi = 0.5$).

The aforementioned results are shown in Figure 6 in terms of CO₂ capture rate for $\chi = 0.5$ and $\chi = 0.8$ (corresponding to $\alpha = 0.84$ and $\alpha = 3$, respectively). The lower boundary indicated by the dashed line represents the case of the CLC process without a sifter. In this case a CCR of 0.36 is achieved, which corresponds to the reaction of volatiles with the oxygen carrier particles and the

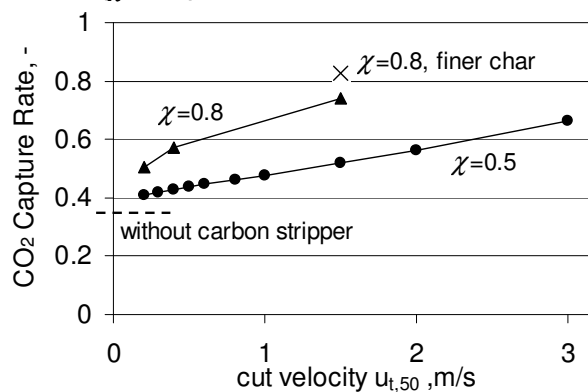


Figure 6: CO₂ capture rate in dependency of the cut terminal settling velocity for $\chi = 0.5$ and $\chi = 0.8$. Additionally the value of CCR is shown for a CLC process without a carbon stripper. The influence of initial char particle size distribution is shown for $\chi = 0.8$ and 1.5 m/s cut velocity.

fraction of char that can be gasified in a single pass through the fuel reactor. Linking the results shown in Figure 6 with those of Figure 5 it can be observed that corresponding to the decreased flow of char towards the air reactor at higher cut velocities the CCR increases at higher cut velocities. For a $\chi = 0.5$ (usual technical sharpness) the CCR is significantly lower than for $\chi = 0.8$, which represents a technically sharp separation. The maximum CCR for $\chi = 0.8$ is 0.74. This value is still rather low but higher CCRs are possible in an optimized process. For instance smaller particle sizes for the coal would facilitate the separation from the oxygen carrier particles in the sifter. In Figure 6 the result of a single simulation at $\chi = 0.8$ and 1.5 m/s cut velocity with finer coal ($d_{p,63} = 50 \mu\text{m}$) is additionally shown. This simulation reaches a CCR of 0.83. This value is still not satisfactory but there is still room for improvements. In general there are two ways to improve the CCR. First the carbon stripper can be improved and second the char conversion in the fuel reactor can be increased. This can be achieved in various ways and the first one was already mentioned above, a further simplification of the separation of char and OC. Other options are:

- Increased residence time of the solids in the fuel reactor
- Higher temperatures in the fuel reactor
- Elevated steam content in the gasification gas
- Usage of special oxygen carriers that release gaseous oxygen in the fuel reactor

Finally, there is a certain spread in the literature concerning char gasification rates with H_2O and CO_2 . The chosen gasification kinetics are a rather conservative choice compared to Ye et al. (6) which differ up to one order of magnitude.

CONCLUSIONS

The influence of the carbon stripper on the CO_2 capture rate of a Chemical-Looping Combustion process for solid fuels was investigated. It can be concluded that the carbon stripper is an important unit operation in a CLC process for solid fuels. Higher cut velocities lead to a decreased slip of char towards the air reactor. This increases the carbon capture rate but on the other hand also increases the load on the carbon stripper. The simulations show rather low CCRs for the chosen process setup and operation. Yet, it is possible to achieve higher CCRs either by improvements regarding the carbon stripper or by increasing the char conversion in the fuel reactor.

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NOTATION

G	Separation grade efficiency,-	β	Split factor for CO / CO ₂ production in H ₂ O gasification, -
k_s	Shrinkage rate, m s ⁻¹	χ	Separation sharpness, -
R	Particle radius, m	CCR	CO ₂ capture rate, -
t	Time, s	CLC	Chemical-Looping Combustion
$u_{t,i}$	Terminal settling velocity of particles in class i , m s ⁻¹	OC	Oxygen carrier
α	Separation sharpness (Rogers),-	PSD	Particle size distribution

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