

## Article

# Intensification Insights from Chemical Looping Combustion Using Coal–Biomass Mixtures with Fe-Based Oxygen Carrier

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**Abstract:** Chemical looping combustion (CLC) is a modern technology that enables the mitigation of the CO<sub>2</sub> concentration without any expense of energy. Experimental evidence shows that combustion of coal/biomass in CLC technology leads to negative carbon emission by replacing the portion of coal with biomass. In the present work, CLC was simulated using a mixture of coal/biomass in CLC; using their different proportions resulted in enhanced CO and CO<sub>2</sub> fractions in the fuel reactor. The carbon capture and oxide oxygen fraction were also found to increase with the enhancement of the fuel reactor's temperature with different proportions of coal/biomass. Increases in the carbon capture efficiency and oxide oxygen fraction of up to 98.86% and ~98%, respectively, were observed within the experimental temperature range. The simulated results of various parameters were predicted and validated with the published experimental results. The stated parameters were also predicted as a function of the different rates of solid circulation and gasification agents. Higher coal char conversion was confirmed in the fuel reactor with the presence of higher biomass concentrations.

**Keywords:** CLC; carbon capture efficiency; Aspen Plus; coal/biomass



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## 1. Introduction

Combustion of fossil fuels, such as coal and petcoke, produces massive amounts of carbon dioxide (CO<sub>2</sub>), which leads to global warming around the world. Concrete steps are therefore required to mitigate the concentration of CO<sub>2</sub>. Viable technologies, such as oxy-fuel combustion, post-combustion and precombustion, have been suggested. Chemical looping combustion (CLC) is comprehended as one of the key alternative processes for clean combustion technology with inherent CO<sub>2</sub> separation [1,2]. The CLC process is known to be performed in a batch reactor or in the much popular interconnected fluidized bed reactors [3,4]. The oxygen required for combustion is supplied indirectly using carrier materials, such as CuO, Fe<sub>2</sub>O<sub>3</sub>, Mn<sub>2</sub>O<sub>3</sub> and NiO, and has been explored by many researchers. In CLC, the metal oxide passes through a recycle redox reaction in two different reactors (i.e., air reactor and fuel reactor), so that no direct interaction between fuel and air takes place during combustion of the fuel. It should be noted that in CLC, the gasification process of solid fuel is slow and, hence, is considered as a rate limiting process. Furthermore, heat produced due to the combustion of solid fuel in CLC is the same as that of conventional combustion [5].

Chemical looping with oxygen uncoupling (CLOU) is an innovative and improved idea with removal of the rate limiting stage of char gasification in CLC [6,7]. Many experimental studies have highlighted the feasibility of a twin fluidized bed for CLC as well as CLOU using coal, petcoke char particles and biomass as solid fuels [8–12].

This is realized thorough the burning of solid fuels, such as coal, biomass, and petcoke [8–12], in CO<sub>2</sub> and H<sub>2</sub>O, with almost nil or less of an energy penalty compared to the abovementioned carbon capture technologies.

The balance of carbon in the atmosphere in terms of CO<sub>2</sub> produced is maintained through the combustion of biomass in CLC. It should be noted that the produced carbon was absorbed from the atmosphere during its growth in the past [13]. Nevertheless, to consider biomass as a capital source of energy with diverse capacities is yet to be explored, and its effect on the environment is not well known [14]. An experimental study of sawdust biomass CLC with Fe<sub>2</sub>O<sub>3</sub> was investigated in 10 kW<sub>th</sub> setup. The biomass combustion process was carried out continuously for 30 h at a temperature range of 740–920 °C [15]. The impact of the temperature range on the fuel reactor in terms of biomass-to-carbon conversion is also significant when exploiting exhaust CO<sub>2</sub> as a gasification agent. Mendiara et al. [16] carried out an experimental study on the combustion of pine wood in an interconnected fluidized bed using hematite as the carrier material [16]. The performances of CLC and CLOU were not significantly affected by the solid circulation rate parameter. Crushed pine wood was used as CLOU fuel in a 1.5 kW<sub>th</sub> unit with an oxygen carrier based on copper [17]. The rate of char conversion was significantly higher in the CLOU compared to the CLC process at a temperature of 900 °C. The CLC concept was also studied with the torrefaction of biomass [18]. In the present study, the torrefied volatile matter was combusted using an oxygen carrier based on iron. In this process, almost all of the carbon was converted into CO<sub>2</sub>. The utilization of wood as a solid fuel with Cu-based oxygen carrier has also been proposed in CLC. Biomass has been proposed as one of the best substitutes solid fuels for use in the chemical looping process [19].

The choice of oxygen carriers is the main factor for improving the conversion of solid fuels in CLC. Exhaustive surveys on the feasibility, oxygen transport capacities and melting points of different metal oxides were carried out. They suggested that metal oxides based on nickel, iron, cobalt, manganese and copper exhibited good thermodynamic properties and stability during the reduction and oxidation processes in CLC and CLOU of solid fuels [20]. Among all, oxygen carriers based on iron are found to be cost effective, in addition to being naturally accessible metal oxides. Experimentation on the CLC of bituminous coal in a 500 W<sub>th</sub> unit using oxygen carrier based on iron was also studied [21]. The coal char reaction with a mixture of CO<sub>2</sub> and H<sub>2</sub>O as a gasification agent was also reported [22]. Among different oxides of iron, the conversion of hematite (Fe<sub>2</sub>O<sub>3</sub>) to magnetite (Fe<sub>3</sub>O<sub>4</sub>) reported the most favorable conversion from a thermodynamic point of view in CLC configuration for the complete conversion of solid fuels to the desired output [15,23]. Iron-based oxide particles exhibit high reactivity during a long continuous run, with the complete conversion of the volatilized and gasified extent of solid fuels [15,16,20,24–26]. The combustion of a coal/biomass mixture using hematite as an oxygen carrier with a fuel reactor temperature in the range 900–980 °C was used in our work for the current simulation research.

Many researchers have carried out extensive experiments on coal/biomass mixtures using different carrier particles. Different coal/biomass fractions may improve the plant's efficiency, and such arrangements may provide additional carbon credits [15,26–32] by negative carbon emission. Hence, it has the potential to lower the total cost of carbon capture.

Recently, interconnected fluidized bed reactor configurations (i.e., air and fuel reactors) have been considered the most suitable arrangement for the CLC process. Simulations based on Aspen Plus have been able to effectively simulate the oxidation and reduction processes in the respective reactors [33–37]. The biomass direct chemical looping (BDCL) process was also simulated in Aspen Plus [33]. The predicted results suggest that BDCL is more efficient compared to regular biomass combustion processes. The capability of Aspen Plus was confirmed by simulating the experimental results of coal as CLOU fuel [12,34]. Aspen Plus is also a useful tool to design process models of CLC and CLOU and to represent the relative study of both for material and energy requirements [35]. However, the literature published using mixture of coal/biomass in CLC is limited. Hence, a mixture of coal/biomass was proposed as the immediate solution for the mitigation of CO<sub>2</sub> concentration. It also offers a lower cost of carbon capture than any other CO<sub>2</sub> capture technology [28].

The aim of the present study was to simulate the CLC of coal/biomass mixture as fuel in Aspen Plus. Published experimental data [23] were used to simulate the CLC process using a coal/biomass blend as the solid fuel and hematite ( $\text{Fe}_2\text{O}_3$ ) as the oxidizer carrier material. The effects of the various coal/biomass mass fractions, oxygen carrier circulation rates and gasification agents on carbon capture, oxide oxygen fraction and conversion efficiency are presented.

## 2. Process Simulation Setup

Gu et al. [23] used the experimental setup shown in Figure 1 to conduct coal/biomass blend combustion with an Fe-based oxygen carrier. The setup consisted of two interconnected fluidized bed reactors that coupled with a loop-seal and cyclone separator. High velocity and a spout-fluidized bed were considered as the air reactor and fuel reactor, respectively. The loop-seal connected the spouted fuel reactor with the air reactor at the bottom. The shape of the loop-seal was a rectangular fluidized bed. According to the published literature [23], the loop-seal avoids gas contamination between the abovementioned two reactors by permitting only reduced carrier material. The arrangement of the components and the working procedure are available elsewhere [23]. As the fuels, bituminous coal and sawdust were considered as the coal and biomass, respectively.  $\text{Fe}_2\text{O}_3$  was used as an iron-based oxygen carrier. The properties of both of the solid fuels considered in the present simulation study are shown in Table 1. The fuel particles' sizes were 200–450  $\mu\text{m}$  and that of 100–300  $\mu\text{m}$  for the considered oxygen carrier during the execution of the simulation in Aspen Plus. Flow rates for air at  $0.84 \text{ m}^3 \cdot \text{h}^{-1}$  and for  $\text{N}_2$  at  $0.27 \text{ m}^3 \cdot \text{h}^{-1}$  were considered. The rate of fuel feeding was  $120 \text{ gm} \cdot \text{hr}^{-1}$  using identical shares of the mass portions of sawdust and bituminous coal. The temperature range of 900 to 980  $^\circ\text{C}$  was considered in the fuel reactor.

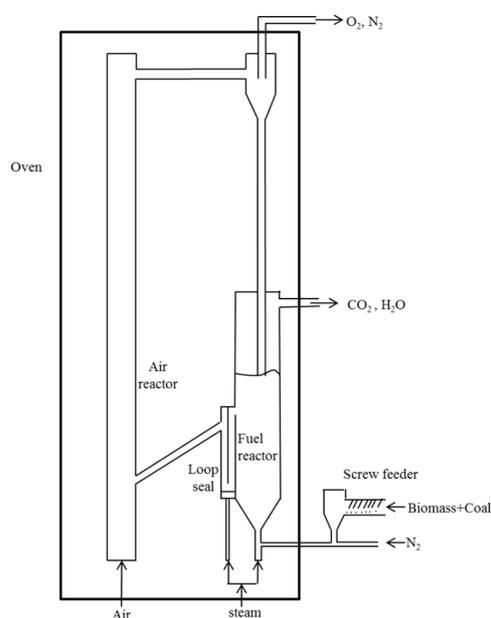


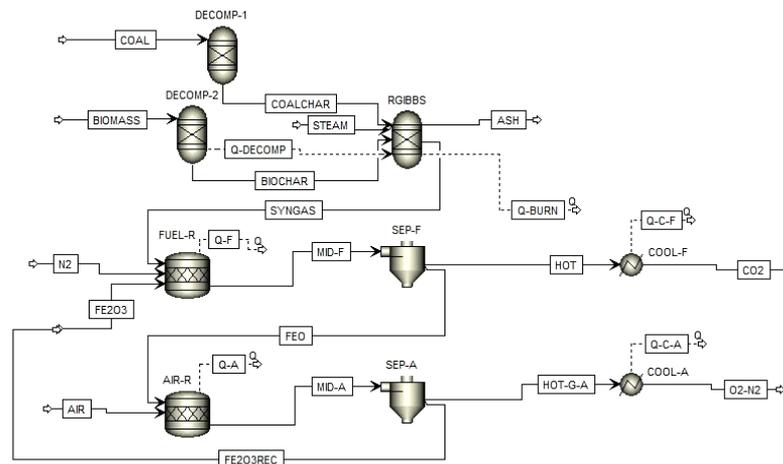
Figure 1. Experimental setup [23].

Figure 2 depicts the CLC process simulation that used two interconnected fluidized bed reactors developed in Aspen Plus. In the current simulation study, bituminous coal and sawdust were considered as the two solid fuels for combustion in the presence of  $\text{Fe}_2\text{O}_3$ . Figure 2 presents the arrangement of various Aspen Plus library models to signify the system level processes. RYIELD reactors represent the pyrolysis of the coal and biomass fuel separately [13]. The RGIBBS reactor collected the outcomes of both RYIELD reactors and also performed char gasification in the presence of steam. The combustion of the generated flue gases performed in RSTOIC (fuel reactor) in the presence of an oxygen

carrier ( $\text{Fe}_2\text{O}_3$ ) to produce  $\text{CO}_2$ ,  $\text{H}_2\text{O}$  and  $\text{Fe}_3\text{O}_4$  in the fuel reactor can be presented as shown in Figure 2.

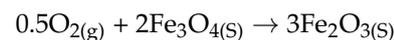
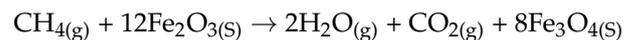
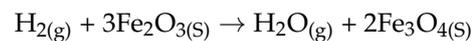
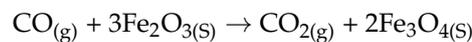
**Table 1.** Properties of the bituminous coal and sawdust [23].

	Sawdust	Bituminous Coal
Proximate analysis (wt %)		
Ash	1.01	4.76
Fixed carbon	10.1	54.13
Volatile matter	74.61	35.1
Moisture	14.28	6.01
Ultimate analysis (wt %)		
Oxygen	40.55	13.81
Nitrogen	1.02	1.03
Hydrogen	5.61	4.3
Carbon	37.43	69.57
Sulphur	0.1	0.52
LHV ( $\text{MJ}\cdot\text{kg}^{-1}$ )	14.5	27.1



**Figure 2.** Component arrangement for CLC of coal/biomass mixtures.

Two RYIELD reactors along with RGIBBS and RSTOIC reactors together performed the process carried out in the experimental fuel reactor. This was due to the fact that not all of the mentioned processes, such as the pyrolysis of the fuel particles, gasification of the char particles and combustion of the same, cannot be modeled explicitly as a single Aspen Plus component.



The equation used to calculate the mass flow rate to RSTOIC (fuel reactor) [16]:

$$\Phi = \frac{F_{OC}R_{OC}}{\Omega_{SF}\dot{m}_{fuel}} \quad (1)$$

The mass flow of the oxygen carrier is indicated by  $F_{OC}$ , and the  $\text{O}_2$  carrying capacity of the oxygen carrier particle is expressed by  $R_{OC}$ . The mass flow of fuel into the system is indicated by  $\dot{m}_{fuel}$ . The air reactor supplies the amount of oxidizer that is needed for complete conversion of the solid fuel, indicated by the  $\Phi$  parameter. The parameter  $\Omega_{SF}$

shows the necessary stoichiometric moles of  $O_2$  that are essential to fully combust one kilogram of supplied fuel (coal/biomass blend) in the system into  $CO_2$  and  $H_2O$ . The value of  $\Omega_{SF}$  can be calculated from the properties of the fuel used [38]:

$$\Omega_{SF} = \frac{f_c}{M_c} + 0.25 \frac{f_H}{M_H} + \frac{f_S}{M_S} - 0.5 \frac{f_O}{M_O} \quad (2)$$

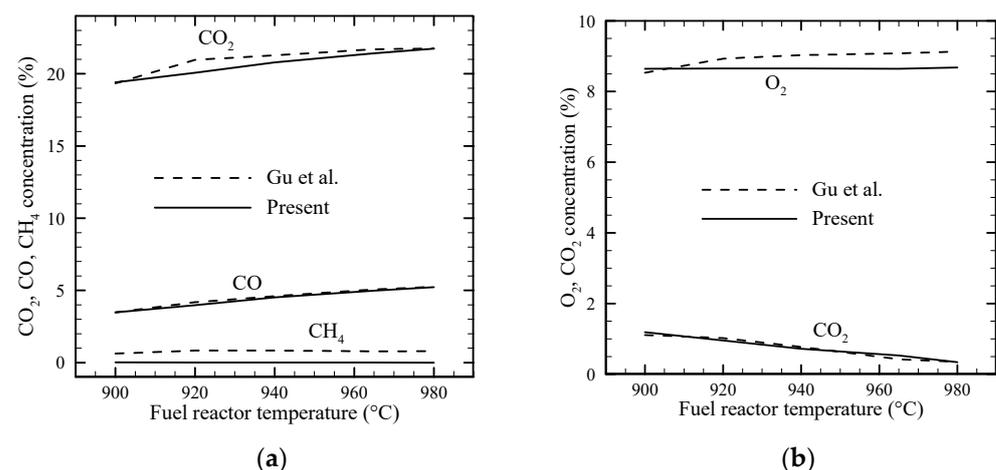
where  $x$ th is the component's fraction denoted by  $f_x$ , and its molar mass is indicated by  $M_x$ .

In the fuel reactor, after ensuring that the coal/biomass blend is completely combusted, the  $Fe_3O_4$  particles are transported to the fast fluidized air reactor (AR) for oxidation. To begin a new cycle, oxidized carrier particles ( $Fe_2O_3$ ) are transferred from the RSTOIC (air reactor) to the RSTOIC (fuel reactor). To simulate the same approach in Aspen Plus, to realize the recirculation of oxygen carrier particles, the fuel reactor inlet stream ( $Fe_2O_3$ ) and air reactor outlet stream (FE2O3REC) are believed to be the same, as shown in Figure 2. The drawback of an interconnected fluidized system is that a number of char particles that remained unconverted in the fuel reactor would move through the air reactor along with the oxidized carrier particles. They would then react and again be oxidized with ambient air. The exit flue gas stream of a fast fluidized bed (AR) thus comprises unreacted  $O_2$ ,  $N_2$  and  $CO_2$ . In a fuel reactor, almost all char particles are converted at higher temperatures and, hence, a fewer number of char particles move towards the air reactor, reducing the  $CO_2$  extent of the air reactor.

### 3. Results and Discussions

#### 3.1. Reactor Gas Concentration

The temperature in a fuel reactor plays a crucial role in CLC's performance. Using solid fuel, Figure 3a compares the experimental and numerically computed fuel reactor extent when equal coal/biomass blends (1:1) were used as fuel. As per the experimental analysis by Gu et al. [23], temperature variations in a fuel reactor range from 900 to 980 °C. The concentrations of CO and  $CO_2$  in the fuel reactor were observed to rise as the temperature of the fuel reactor increased. The concentration of  $CH_4$  almost remained constant in the considered temperature range in the spouted reactor (FR).



**Figure 3.** Variations in the (a) concentrations of  $CO_2$ , CO and  $CH_4$  in the fuel reactor and (b) in the concentrations of  $O_2$  and  $CO_2$  in the air reactor at the temperature of the fuel reactor.

The coal/biomass blend is combusted in the fuel reactor, where interactions between them during combustion may be present [32]. Devolatilization of the biomass and coal can easily take place at lower fuel reactor temperatures. Moreover, for biomass, char particle conversion is faster due to the fact of high volatility, and they are reacted faster compared to bituminous coal. This may accelerate syngas (i.e., CO and  $H_2$ ) production in the freeboard of the air reactor. Fe-based carrier particles have a lower oxygen transport capacity, and due

to the fact of this, lead to a reduction in the time available for oxidation of the generated syngas. This increases the CO concentration. Char gasification using steam is a rather slow process in solid fuel CLC. According to an experimental study by Gu et al. [23], the existence of alkaline metals in biomass act as a catalyst for decreasing the activation energy for coal. Though, due to the endothermic nature of gasification, we can expect that the coal gasification is slower compared to biomass gasification and, hence, can interact more actively with the oxygen carriers in the nearby bed area. This process causes better oxidation and, hence, the CO<sub>2</sub> concentration increases in the fuel reactor. The variance in the gas concentration in the air reactor as a function of the fuel reactor temperature is shown in Figure 3b.

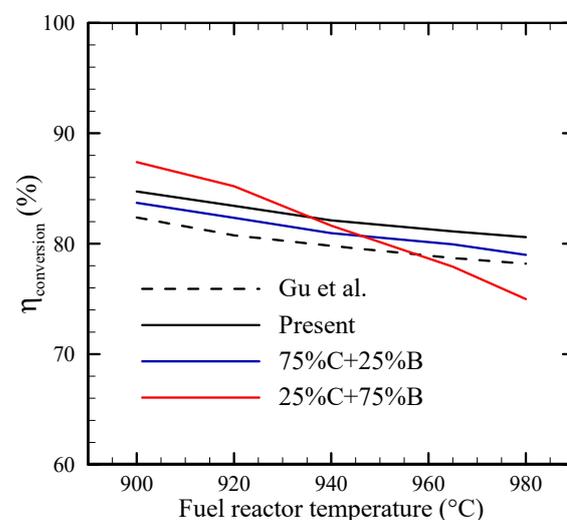
With an increase in the temperature of the fuel reactor, the O<sub>2</sub> concentration remained almost constant and decreased the CO<sub>2</sub> concentration. This was due to the fact that at higher fuel reactor temperatures, most of the coal/biomass char was converted in the fuel reactor, result in in lesser or no char that moved to the air reactor, leading to a decrease in the CO<sub>2</sub> concentration.

### 3.2. Conversion Efficiency

The following is the measurement at a specific fuel reactor temperature of the number of carbonaceous gases converted to CO<sub>2</sub> in a fuel reactor and is described as [23]:

$$\eta_{conversion} = \frac{W_{CO_2,FR}}{W_{CO_2,FR} + W_{CO,FR} + W_{CH_4,FR}} \quad (3)$$

where  $W_{i,FR}$  represent the volume percentage of fuel reactor extent in terms of different gases ( $i = CO_2, CO$  and  $CH_4$ ). This definition considered CO<sub>2</sub>, CO and CH<sub>4</sub> as the only carbonaceous gases in the fuel reactor. Figure 4 shows the variations in the conversion efficiency with the fuel reactor temperature. At higher fuel reactor temperatures, due to the rate limiting char gasification process, fuel reactor carbonaceous gases increased. The carbonaceous gases did not fully convert to CO<sub>2</sub> due to the Fe-based oxygen carrier's restricted oxidizer transport ability; when supplied in a larger quantity of oxygen carrier (Equations (1) and (2)), it will still not lead to complete combustion due to the insufficient contact time, resulting in a decreased conversion efficiency. The CLC of the coal/biomass blends, both with an equal (1:1) and a higher mass concentration of coal in the mixtures, behaved almost like the CLC of pure coal. The effect of various mass concentrations of the coal/biomass blends on the conversion efficiency is also shown in Figure 4.



**Figure 4.** Variations in the Conversion Efficiency with the Fuel Reactor Temperature.

The experimental results corresponding to pure coal as a CLC fuel was well matched with the numerical results corresponding to a higher mass fraction of coal in the coal/biomass blend [5]. Similarly, CLC of coal/biomass blends with a higher mass concentration of biomass behaved similar to CLC of pure biomass [38]. The numerically computed conversion efficiency trend corresponded to a higher mass fraction of biomass in the mixture. It can be seen in Figure 4 that with a higher fraction of biomass in the mixture, the gasification rate increased due the large fraction of highly volatile fuel and thereby increased the syngas concentration. As discussed earlier, the limited oxidizer transport capacity of Fe-based oxygen carrier had a sufficient reaction time to convert the higher concentration of fuel reactor syngas, which resulted in a sharp fall in the conversion efficiency as compared to other configurations. This agrees well with the experimental results in [15,23], corresponding to the pure biomass used as CLC fuel.

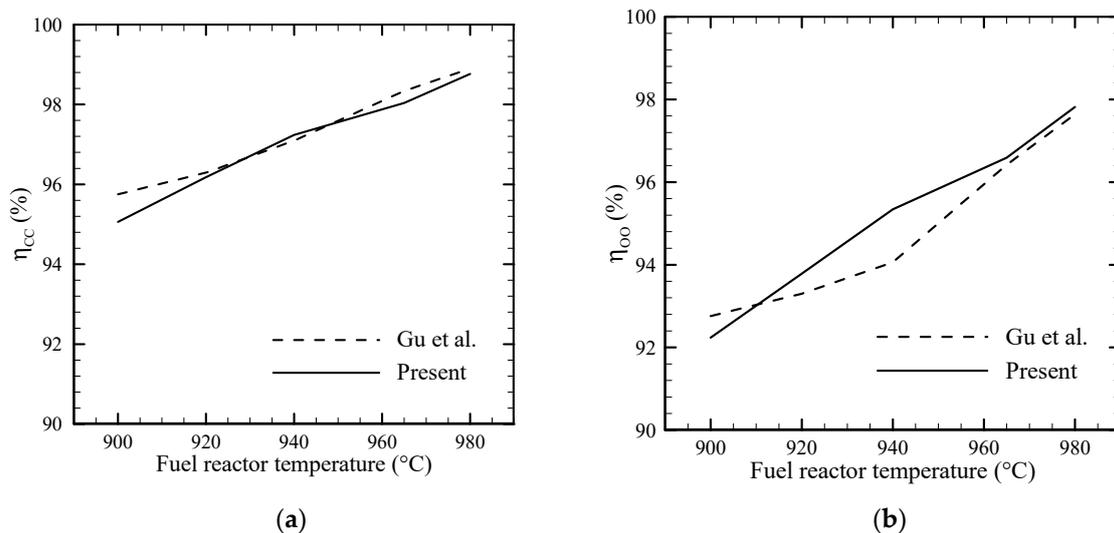
### 3.3. Carbon Capture Efficiency and Oxide Oxygen Fraction

Conversion of char particles takes place in the CLC fuel reactor at particular temperatures, defined as char conversion [16]:

$$X_{char} = \frac{[F_{CO_2,FR} + F_{CO,FR} + F_{CH_4,FR} - F_{C,vol}]_{out}}{[F_{CO_2,FR} + F_{CO,FR} + F_{CH_4,FR} + F_{CO_2,AR} - F_{C,vol}]_{out}} \quad (4)$$

The carbon capture performance relies on the conversion of solid fuel chars. Variations in the carbon capture efficiency with respect to fuel reactor temperature are shown in Figure 5a. As explained earlier, in the CLC fuel reactor, higher temperatures tended to increase the conversion of char and, therefore, less char will occur with a reduced carrier content in the air reactor. Thus, falls in the air reactor CO<sub>2</sub> concentration would result in an increase in the carbon capture efficiency, and it is given as [16]:

$$\eta_{CC} = \frac{[F_{CO_2,FR} + F_{CO,FR} + F_{CH_4,FR}]_{out}}{[F_{CO_2,FR} + F_{CO,FR} + F_{CH_4,FR} + F_{CO_2,AR}]_{out}} \quad (5)$$



**Figure 5.** (a) Efficiency of carbon capture and (b) oxide oxygen fraction as a function of the temperature of the fuel reactor.

From above equation, it is clear that there are increments in the carbon capture efficiency with a higher char conversion in the fuel reactor.

Figure 5b presents variations in the oxide oxygen fraction with respect to the fuel reactor temperature, and it is defined as [23]:

$$\eta_{OO} = \frac{0.21 - W_{O_2,AR} - W_{CO_2,AR}}{0.21 - W_{O_2,AR} - (0.21 \times W_{CO_2,AR})} \quad (6)$$

where  $W_{i,AR}$  denotes the volume percentage of gases ( $i = CO_2, CO$ ) at the exit of the air reactor.

Both the oxide oxygen fraction and carbon capture efficiency depend on the air reactor extent of  $CO_2$ . Within a defined temperature range, the  $O_2$  concentration was almost independent of temperature, while the  $CO_2$  concentration declined in the air reactor. This was primarily due to the fact that almost all char particles in the fuel reactor were converted, while a smaller number of unconverted char particles reached the air reactor. The numerically computed values of  $\eta_{CC}$  and  $\eta_{OO}$  agreed well with the experimental results. Differences in the respective values of  $\eta_{CC}$  and  $\eta_{OO}$  were found, as Aspen Plus does not include the hydrodynamic losses that occur in fluidized bed combustion [34,36].

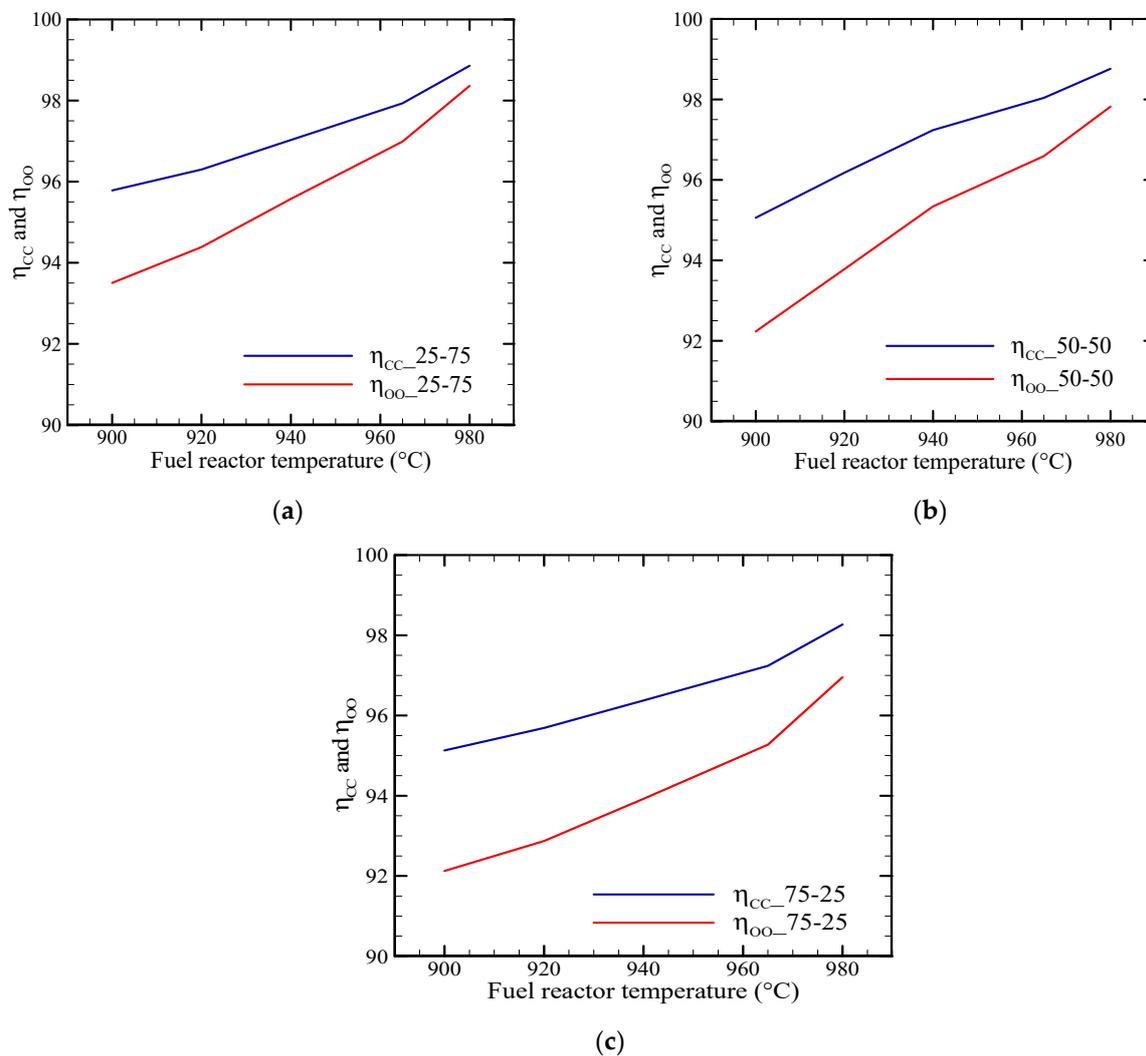
To use biomass as fuel over coal has the important advantages of the presence of highly reactive alkali and alkaline metal in higher concentrations. Hence, coal conversion accelerates due to the reduction in activation energy and gasification temperature of coal in the presence of biomass earth metals [32]. As a result, biomass' higher alkali and alkaline earth metal content functions as a low-cost catalyst, ensuring that the coal/biomass mix burns almost entirely. This understanding indicates that biomass functions, at the same time, as both a fuel and a catalyst. An experimental study by Gu et al. [23] concluded that the co-combustion coal/biomass mixtures may decrease the problem of sintering and agglomeration of the carrier particles. This is a critical issue in the CLC of pure biomass. Experimental study of CLC using various coal/biomass mass fractions in the mixtures may affect the performance parameters such as oxide oxygen fraction and carbon capture efficiency.

Figure 6 shows the effect of different fractions of coal/biomass mass in the mixture on the oxide oxygen fraction and the efficiency of carbon capture using the same temperature range that was used by Gu et al. [23]. It was demonstrated that both values (i.e.,  $\eta_{CC}$  and  $\eta_{OO}$ ) increased with a higher temperature of the fuel reactor using various coal/biomass mass fractions in the mixture. An experimental analysis by Gu et al. [23] indicated that this discrepancy ( $\eta_{CC} - \eta_{OO}$ ) indicates the presence unconverted CO and  $CH_4$  carbonaceous gases in the fuel reactor. Equal values of  $\eta_{CC}$  and  $\eta_{OO}$  implies that all combustible gases were converted into  $CO_2$  in the fuel reactor. Greater differences between  $\eta_{CC}$  and  $\eta_{OO}$  were found using equal and higher mass fractions of coal in the coal/biomass blend compared to higher fractions of biomass as depicted in Figure 6a,b. Gu et al. [23] also concluded their experimental study with the same results, using only biomass as CLC fuel. Luo et al. [32] additionally found that a higher mass fraction of biomass in the coal and biomass mixture increased the coal char conversion in the fuel reactor, even at lower fuel reactor temperatures. At a given fuel reactor temperature, this will improve the performance of both carbon capture and oxide oxygen fraction. The deviation between  $\eta_{CC}$  and  $\eta_{OO}$  was thus lower with a higher mass fraction of biomass in the mixture of coal/biomass.

### 3.4. Effect of Gasification Agent

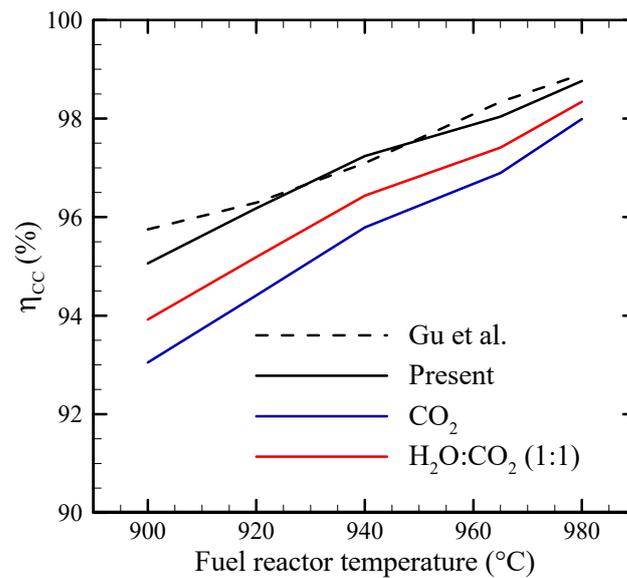
Steam and  $CO_2$  are the most commonly used gasification agents for the CLC of solid fuels in the interconnected fluidized bed reactors [5,21–23,32]. Figure 7 shows the effect of using gasification agents (i.e., steam and  $CO_2$ ) on the efficiency of carbon capture. The simulation study was also carried out using equal fractions of steam and  $CO_2$  (1:1) together as gasification agents to observe the effect on carbon capture efficiency. The char conversion while using steam was high compared to  $CO_2$  as the gasification agent. Leion et al. [21] investigated gasification of coal using steam,  $CO_2$ , and steam and  $CO_2$  together. According

to their results, the char conversion increased as the steam fraction in the gasification agent increased.



**Figure 6.** Variations in the oxide oxygen fraction and carbon capture efficiency depending on the temperature of the fuel reactor considering fractions: (a) 25/75; (b) 50/50; (c) 75/25 (% coal/% biomass).

Roberts and Harris [22] presented data regarding coal char reaction at high pressure in  $\text{CO}_2$  and  $\text{H}_2\text{O}$  mixtures. It was observed that reaction of coal char was not the sum of the reaction rate obtained by using  $\text{CO}_2$  and  $\text{H}_2\text{O}$  as individual gasification agents. However, Mendiara et al. [16] who used steam or  $\text{CO}_2$  as a gasification agent in the CLC of pure biomass showed no major impact on the carbon capture efficiency. In the present simulation, we used a coal/biomass blend as CLC fuel, and it was expected that a mixture with a higher biomass concentration would have no significant impact on the carbon capture efficiency with any gasification agent. Jimin et al. [31] used  $\text{CO}_2$  as a gasification agent in their experimental study with a coal/biomass blend as the CLC fuel. It was found that pyrolysis of coal and biomass occurred earlier as compared to the pyrolysis of pure coal due to the presence of biomass, which may have benefited in the combustion process at lower temperatures.



**Figure 7.** Performance of carbon capture as a feature of the gasification agents (i.e., steam and CO<sub>2</sub>).

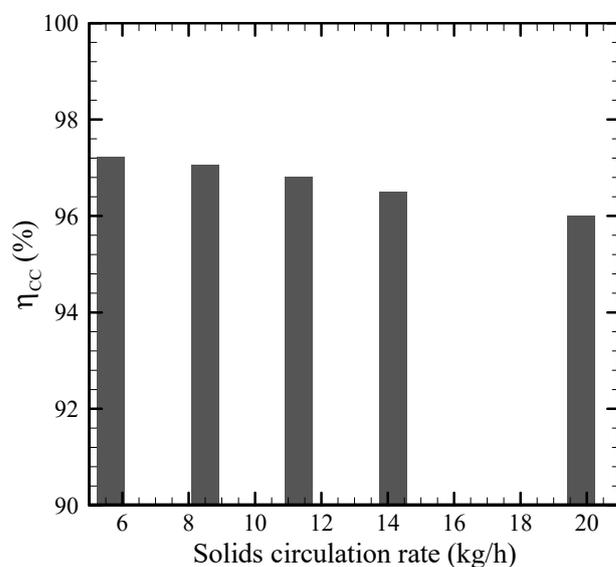
In the present simulation study, there was found a variation of 1.12% in the efficiency of carbon capture at a fuel reactor temperature of 900 °C and of 0.42% at 980 °C while using pure steam and a mixture of equal fractions of H<sub>2</sub>O/CO<sub>2</sub> (1:1) as gasification agents. Compared to pure steam as a gasification agent, no major variations were observed in the efficiency of carbon capture at lower and higher temperatures of the fuel reactor using steam and CO<sub>2</sub> together. Therefore, a fraction of CO<sub>2</sub> along with steam can be used as a gasification agent without compromising the carbon capture efficiency. Moreover, using CO<sub>2</sub> as a gasification agent can reduce the cost of carbon capture, as almost pure CO<sub>2</sub> is readily available in the fuel reactor exit, and a reduction in the cost of steam generation can further reduce the carbon capture cost.

### 3.5. Effect of the Solid Circulation Rate on the Performance Parameters

The sum of oxygen carrier particles circulating in the system per unit of time is known as the rate of solids circulation. In the present simulation study, the oxygen carrier flow rate ranged from 5.67 to 19.85 kg·h<sup>-1</sup>. The ratio of the oxygen carrier/fuel equivalent values was 1 to 3.5. Figure 8 shows the efficiency of carbon capture as a function of the circulation rate of solids at a constant fuel reactor temperature of 940 °C, using the same fraction of the coal/biomass mixture. The improvement in the carbon capture efficiency was negligible, as the solid circulation rate increased. The availability of an oxidizer was adequate for full combustion at higher solid circulation rates, but the residence time of the char particles in the fuel reactor was limited; thus, complete char combustion was not possible due to the reduced residence time. As a result, the carbon capture efficiency would be marginally decreased. Therefore, for the conversion of solid fuel char in the fuel reactor, the availability of sufficient oxygen is required along with the minimum residence time. It was specified that the fuel reactor configuration should be improved for a longer solid fuel residence time in CLC applications [23].

Cuadrat et al. [21] and Mendiara et al. [16] concluded experimentally that the high circulation rate of solids and higher oxygen-carrier-to-fuel ratio had negligible advantages in regard to the conversion of char and, thus, the performance of carbon capture. It is proposed that the fuel reactor configuration should be improved to increase the residence time of the char particles in the fuel reactor. The published literature proposes to supply additional pure oxygen downstream of the fuel reactor (oxygen polishing) to fully convert the carbonaceous gases to CO<sub>2</sub>, thus increasing the carbon capture efficiency [7,8,10,21,25]. The production of pure oxygen, however, represents an energy penalty and additional process expenses and, therefore, high gas conversion in the fuel reactor is needed using the

optimum solid circulation rate. Hence, other options need to be explored to improve the conversion of carbonaceous gases in CLC of solid fuels.



**Figure 8.** Performance of the carbon capture as a result of the circulation rate of solids.

#### 4. Conclusions

In Aspen Plus, combustion of a coal/biomass blend in CLC using  $\text{Fe}_2\text{O}_3$  was simulated. The experimental findings, such as the extent of the fuel and air reactor, conversion efficiency, fraction of oxide oxygen and carbon capture efficiency, were validated depending on the temperature of the fuel reactor. Variations between the carbon capture efficiency ( $\eta_{CC}$ ) and the oxide oxygen fraction ( $\eta_{OO}$ ) confirmed a fuel reactor with the existence of carbonaceous gases that still remain unconverted. The value of ( $\eta_{CC}-\eta_{OO}$ ) was lower at higher fuel reactor temperatures compared to lower fuel reactor temperatures for various coal/biomass blend configurations. This was primarily due to the endothermic nature of the solid fuel gasification response in the fuel reactor. These findings show that higher mass fractions of coal in the mixture of biomass and coal behave in a similar manner to pure coal CLC. A higher mass fraction of steam increased solid fuel gasification, thereby increasing the efficiency of carbon capture. No major increase in carbon capture efficiency was observed using  $\text{CO}_2$  and steam together as a gasification agent in the CLC of coal/biomass mixtures. As a gasification agent,  $\text{CO}_2$  can reduce the cost of carbon capture due to the lower costs associated with steam generation. There were also no notable differences in the performance of carbon capture with a higher oxygen-carrier-to-fuel ratio.

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