Hydrogen Rich Syngas Production from Oxy-Steam Gasification of a Lignite Coal – A Design and Optimization Study

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ABSTRACT

This study describes an experimental and computational fluid dynamics (CFD) effort towards optimizing: hydrogen rich syngas production and cold gas efficiencies during the thermochemical conversion of a lignite coal using oxygen and steam as gasifying agents. A bubbling bed gasifier was systematically designed, constructed and commissioned to achieve these objectives. The bed temperature was maintained at 1023 K during the gasification testing of the highly reactive lignite coal.

The hydrogen levels in the syngas were examined as a function of oxygen, coal and steam flow rates. A maximum hydrogen concentration of 50% (dry-basis) was achieved at low oxygen to carbon ratios and the cold gas efficiencies were in the range 80 – 90%. The observed experimental trends in syngas compositions and cold gas efficiencies were reasonably well represented by the CFD simulations and compared favorably with data obtained from a transport reactor integrated gasification system. Simulations predicted that the major product gases at the reactor outlet were close to their equilibrium levels. The decrease in hydrogen concentrations in the syngas at high oxygen and steam flow rates resulted from changes in the oxidation reaction rates, hydrodynamics and steam levels. The levels of steam were attaining saturation conditions.

Keywords: Lignite; Gasification; Bubbling bed; CFD

1. Introduction

Coal utilization in an environmentally friendly manner can be accomplished through gasification technologies where coal is partially oxidized and reacted with steam to produce syngas consisting of hydrogen and carbon monoxide. Lignite coal resources in the US are abundant. In order to take advantage of this abundant resource, technologies must consider its high moisture content (which reduces its calorific value), high reactivity, abundance of oxygen, and high levels of organically associated alkali (sodium) and alkaline earth (calcium) elements. Lignite is a highly reactive coal in reducing environments due to the abundance of free radicals that are formed during thermal transformations of its oxygen functional groups [1]. Similarly in gasification systems employed for power generation, the high moisture content can increase the flow rates to the turbines and promote water-gas shift (WGS) reaction.

There are three types of commercially available gasifiers, namely: entrained flow gasifiers, fixed-bed gasifier, and fluidized bed gasifiers. In order for lignites to be used in entrained flow gasifiers they must be dried and either fired as a finely ground coal particles ($\sim 100\text{ }\mu\text{m}$) or as a slurry. The entrained flow gasifiers operate at high temperatures to achieve high carbon conversion, produce a tar-free syngas, while ensuring that all of the ash is melted into slag. Besides drying the lignite prior to use in the entrained flow gasifier, another drawback of this gasifier is their high oxidant requirement for achieving good conversion which increases their operating cost. The low particle volume fractions resulting from the finely ground fuel employed in these gasifiers make them amenable to simulation employing an Euler-Lagrangian approach [2 - 4]. In a moving bed gasifier, large particles are employed in a bed that passes through different reaction zones (drying, carbonization, gasification and combustion) countercurrent to the flowing gases. These gasifiers can operate in dry ash modes or slagging modes where their temperatures are controlled by the amount of steam going into the gasifier. A key advantage of this type of gasifier, is their lower oxidant requirement compared to entrained flow gasifiers and the lower grinding requirement for feedstock preparation since larger sized fuel feed can be employed. However, the requirement of the removal of fine coal particles from coal feed and tar production are some disadvantages associated with this gasifier type. North Dakota (ND) lignite has been utilized to produce methane on a commercial scale

by Dakota Gasification Company's Great Plains Synfuel Plant that employs a moving bed gasifier [5]. This is also the first plant to utilize the produced energy from coal to separate and sequester CO₂ emissions by sending oxygen and steam in the moving bed gasifier that produces primarily CO and H₂ which is then employed to produce methane. The combustion zone within the gasifier attains a temperature of around 1450 K [5]. Fragmentation, ash melting, and agglomeration in the oxygen/steam blown moving bed gasifier employing ND lignite coal was recently studied by Mangena et al. [6]. While mathematical models for the moving bed gasifier have been reported in the literature [7], the authors are not aware of any CFD studies carried out with this type of gasifier.

One of the operational challenges associated with employing the high sodium (Na) ND lignite coal at higher temperatures is that, sodium can vaporize and/or react with other ash species to produce low melting point phases that can contribute to agglomeration and ash accumulation of gasifier and downstream surfaces. For example, in high gasifiers equipped with syngas coolers, Na can vaporize and condense downstream in the syngas cooler and hot gas clean up systems and create operational challenges. Therefore, for coals that are prone to ash deposition, there exists an operation window where the temperature can be high enough to achieve good carbon conversion by taking advantage of the enhanced reactivity of lignite coals but low enough to prevent ash deposition. Therefore, there is considerable interest in understanding the operational characteristics of high Na coals such as ND lignite in low temperature gasification systems.

Fluidized-bed gasifiers can take advantage of lignites high reactivity while managing the behavior of Na-rich ash through careful bed materials that do not cause agglomeration [1]. Fluidized beds promote efficient mixing, and as a result, nearly uniform temperatures are sustained within the reactor bed. In fluidized reactor regimes, the operating temperature should be high enough to achieve good carbon conversion as well as decompose the tars and other liquid products produced during pyrolysis and devolatilization. At the same time, the temperature should be lower than melting point ash materials produced. Due to this temperature constraint highly reactive coals, such as lignite are often utilized so that a good carbon conversion can be achieved at the lower operating temperatures. Due to the high granular

phase volume fractions encountered within the reactor, CFD simulations of the fluidized bed gasifier are carried out in an Euler-Euler modeling framework [8 - 11].

This study, describes the optimization of hydrogen rich syngas and cold gas efficiencies during oxy-steam gasification of a ND lignite coal in a bubbling bed gasifier whose bed temperature was maintained at 1023 K to minimize the shortcomings and challenges associated with the utilization of the high Na fuel. The gasifier was recently designed, constructed and commissioned to achieve these objectives [3].

Initially, during the commissioning of our gasification system, experiments were performed at 1073 K. During these experiments, agglomerates were formed between the sodium and the silica sand bed to the point that the bed was no longer fluidized. These agglomerates resulted from the physical and chemical transformations associated with the alkali and alkaline rich silicate glass liquid phases in lignite. These resulted in: changes to the viscosity, surface tension and sintering characteristics of the agglomerates, a lower quality syngas, as well as damage to the system. As a result of this, the operating temperature in this study was lowered to 1023 K to prevent system damage as well as to ensure that quality experiments could be performed. Experimental measurements are complimented by CFD simulations to obtain insights into the bed fluidization characteristics and reaction rates in order to explain the variability in the syngas compositions to the changes in the operating conditions.

2. Methods

2.1 Experimental Setup

Figure 1a shows the bubbling/fluidized bed reactor system that was systematically designed, constructed and commissioned to achieve the objectives of this study. The fluidized bed reactor was constructed from a 304 stainless steel pipe and had an internal diameter of 0.0762 m and was 1.651m tall. 2.7 kg of sand (corresponding to a bed height of 30.5 cm) acts as the fluidizing medium inside the reactor. Steam and oxygen, which are both pre heated to 600 K enter from the bottom of the reactor and fluidize the sand bed. Coal enters the side of the reactor, above the fluidized sand bed. The densities of sand and coal particles were measured to be 2650 kg/m³ and 833 kg/m³ respectively. Uniform particle sizes of 500

microns and 400 microns were assumed for the sand and coal particles respectively. The proximate, ultimate and ash analysis of the coal is shown in Table 1.

The fluidized bed was designed to operate in a bubbling mode, at atmospheric pressure over the range of fluid and coal flow rates investigated in this study [12]. The reactor is externally heated with three different sets of ceramic heaters, capable of reaching the target operating temperature (1023 K) for fluidized bed combustion and gasification. The reactor temperature is controlled by thermocouples on each zone located on the outside wall of the reactor. In addition, three thermocouples at each zone located inside the reactor monitor the actual temperature of the reactor. The fluidized bed also contains a differential pressure gauge to monitor the fluidization characteristics within the reactor.

The syngas from the gasification travelled up out of the reactor, and through a cyclone. The cyclone removed any solid particulate matter, such as ash or soot and was kept insulated to keep the gas hot. Next, the syngas passed through two condensers where any remaining organic material, such as tar or aqueous material was removed. Once the syngas was clean and cooled a sample of it was analyzed in a Laser Gas Analyzer, which measured its composition. The gas analyzer was able to detect hydrogen, oxygen, carbon monoxide, carbon dioxide, hydrogen sulfide, methane and water vapor. The rest of the syngas was sent via an inductor system and out into the atmosphere. The average duration for each experimental condition examined in this study was 20 - 30 minutes while ensuring that steady state syngas compositions were being achieved.

2.2 Simulation Methodology

The Multiphase Eulerian model in the commercial code ANSYS FLUENT® [13] was employed to carry out the CFD simulations. The multiphase Euler-Euler model assumes the gas and solid phases as continuous and fully interpenetrating within each control volume and the momentum, continuity, and energy equations are solved for each phase (gas-phase, sand and coal) in an Eulerian reference frame with a single pressure field being shared by all 3 phases. The momentum exchange between the phases was modeled through interaction terms between the phases [13]. By assuming that the turbulence transfer among the phases would play a strong role in determining the bed dynamics, the k- ϵ per-phase turbulence

model was employed in the simulations where equations for the turbulence kinetic energy and turbulence dissipation rate were solved for each phase. The different models and settings employed in the simulation process are summarized in Table 2.

Since the coal was being injected from the side of the reactor above the bed height, a truly accurate representation of the reactor configuration for modeling purposes would require three-dimensional (3D) simulations. However, in order to allow fast and efficient transient simulations of the reactor conditions, simulations were carried out in a 2D axisymmetric domain employing 12,430 computational cells with the coal particles injected along with the oxygen and steam at the bottom of the reactor.

The different homogeneous and heterogeneous reactions modeled in this study are summarized in Table 3. It is important to note that due to the considerably variability in the composition of solid fuels, there are no unique models to accurately describe the species evolved during the devolatilization process in gasification systems. In this study, the correlations (as a function of the volatile matter content) proposed by Weimer and Clough [14] were employed to determine the species distribution in the coal volatile matter. Due to the high reactivity of lignite char and its open pore structure as described previously, the heterogeneous char oxidation reactions were assumed to proceed at a rate determined purely from Arrhenius kinetics assuming no diffusion resistance encountered by the oxidant when reaching the particle surface. The simulations were run to approximately 20 seconds of real time and ensuring that the outlet syngas composition converged to their steady state values. An overview of CFD modeling in fluidized bed combustion and gasification has been provided by Singh et al. [11].

3. Results and Discussion

The optimization of hydrogen rich syngas production and cold gas efficiencies was carried out in three stages where the mass flow rates of oxygen, coal and steam were changed individually while maintaining the other two flow mass flow rates constant. For the CFD study, a select number of experimental conditions were then identified to ensure that the observed experimental trends were being adequately replicated. Table 4 summarizes the different flow rates investigated in the CFD study.

3.1 Optimizing oxygen flow rates

The first set of experiments aimed to find the optimum oxygen flow rate, while maintaining constant flow rates of coal and steam. Measurements of the steady state syngas composition were made at six flow rates with the oxygen to carbon ratio (O/C) varying from 0.8 to 1.6. Simulations of three of those conditions were carried out. The steady state compositions of the major and minor gases in the syngas from the six conditions are summarized in Table 5. Also shown are the results from the Transport reactor integrated gasification (TRIG) system that have been under development for the past several years through programs conducted at the Energy and Environment Research Center (EERC, Grand Forks, ND) and Southern Company. Data from the EERC TRIG gasification system were used to scale up the technology at the power systems development facility (PSDF) near Wilsonville, Alabama. Lignites have the potential to be excellent feedstocks for coal gasification systems such as the TRIG system which is a fast-moving circulating fluidized-bed system that is operated at lower temperature with higher conversion rates [1]. Table 5 shows that the syngas compositions in the bubbling bed gasifier compare very favorably against the TRIG system. Dakota Gasification Company's Great Plains Synfuel Plant is the first plant to utilize the produced energy from coal to separate and sequester CO₂ emissions. The CO₂ is separated and transported in a pipeline to an oil field in Canada 205 miles away for enhanced oil recovery. Oil recovery rates have more than doubled, and millions of tons of CO₂ have been sequestered [1]. Therefore, the product gas produced in this study may be employed to produce natural gas and by-products including CO₂ (used for enhanced oil recovery), fertilizers, gases, and chemicals.

Figure 2a represents these results graphically and highlight the fact that as the oxygen to carbon ratio increases the amount of hydrogen decreases. Next, cold gas efficiency (η_{CG}) was defined as a function of the low heating values (LHV), volumetric flow rates (Q) and mass flow rates (m):

$$\eta_{CG} = \frac{LHV_{syngas} \times Q_{syngas}}{LHV_{coal} \times m_{coal}} \times 100 \quad (1)$$

Figure 2b shows the variation in the cold gas efficiencies determined experimentally as a function of O/C ratio. Cold gas efficiencies close to 90% were obtained in our experiments, which compares well against an efficiency of 79% that was obtained with the TRIG system. η_{CG} stays relatively constant across all O/C

ratios due to the fact that the decrease in H_2 concentrations with increase in O/C ratio is offset by increases in CO concentrations and volumetric flow rates of the syngas. These results are in contrast with those obtained by Karimipour et al. [19] who found that the cold gas efficiency goes down with a decrease in O/C ratio as a result of decreasing the carbon conversion and the resulting decrease in the volumetric flow rate of combustible gases resulting from the lower carbon conversions. Due to the use of a highly reactive ND lignite coal in conjunction with the oxygen blown gasification conditions in this study, the carbon conversion was likely not impacted over the range of O/C ratios investigated here.

Figure 3 compares the measurements and predictions of the major syngas species at different O/C ratios. While the experimental trends are well replicated by the simulations in general, there is a significant over-prediction of CO_2 concentrations at an O/C ratio of 1.6. Figure 4 shows contours of bed (sand) volume fraction at different O/C ratios. Due to the increase in inlet gas flow rates with O/C ratios, the transition from a bubbling regime towards a slug flow regime is observed. The inlet gas velocity at O/C of 1.6 was 0.14 m/s whereas the minimum slugging velocity at these operating conditions was determined to be 0.17 m/s [12].

Contours of O_2 mole fractions along with CO and H_2 oxidation rates are shown in Figure 5. Since the oxidation rates of these species are proportional to the O_2 concentrations (Table 3), an increase in their oxidation rates and rapid consumption of O_2 near the inlet is observed at higher O/C ratios. This oxidation process results in an increase in the local gas temperature as well as a reduction in CO concentration. This favors the reverse WGS reaction leading to lower H_2 concentrations at higher O/C ratios. The observed trends in H_2 , CO and CO_2 ratios can therefore be attributed to these oxidation reactions. Under the actual experimental condition, since the coal was injected through a side inlet over the bed and not through the gas inlet as represented in the simulations, the drastic increase in CO oxidation rates and O_2 consumption rates near the inlet with the O/C ratios as shown in Figure 5 would likely not occur in reality. The significant over-prediction of the CO_2 concentrations at higher O/C ratios in Figure 3 can therefore be attributed to the representation of the actual 3D experimental conditions in a 2D axisymmetric domain.

The equilibrium constant for the WGS reaction can be represented by Eq. 2 [20]:

$$K_{WGS} = \frac{[CO_2][H_2]}{[CO][H_2O]} = \exp(-3.7774 + \frac{4118.6}{T(K)}) \quad (2)$$

The temperature and gas compositions at the reactor outlet in the simulations were then employed to determine K_{WGS} to ascertain if the simulation domain (Figure 1b) was adequate to capture the equilibrium compositions of the major gas species (Note that the gas samples were drawn downstream of the reactor and were likely at equilibrium). Table 6 compares the K_{WGS} predicted by Eq. (2) and compares it against the K_{WGS} predicted by the simulation. It is noteworthy that the major product gases are therefore close to their equilibrium values. At O/C ratios of 1.1 and 1.6 however, further extending the simulation domain would likely drive the reverse WGS reaction towards equilibrium, resulting in decreases in H_2 and CO_2 compositions while increasing H_2O and CO concentrations.

3.2 Optimizing coal flow rates

Once the optimum amount of oxygen needed for hydrogen generation was identified as $5.31e-05$ kg/s, the amount of coal was varied while maintaining constant mass flow rates of oxygen and steam as shown in Table 4. Three different coal feed rates were selected to determine syngas composition as a function of steam to carbon ratio (H_2O/C). Figure 6 compares the measurements and predictions of the major gases in the syngas with the variations in the coal flow rates. Experimentally, the hydrogen production reached its peak value at a steam to carbon ratio close to unity. The trends in H_2 and CO_2 concentrations are also well represented by the simulations. Figure 7a shows an increase in gas temperature inside the reactor with an increase in coal flow rate. This is attributable to an increase in the heterogeneous reaction rates involving the volatile fuel. While this result in an increase in CO , an optimum temperature is reached before CO is oxidized to CO_2 . This is evident in the measurements reported in Figure 6 where the highest CO and lowest CO_2 concentrations are observed at an H_2O/C ratio close to unity.

The CFD simulations were unable to predict the CH_4 concentrations adequately. The measured CH_4 mole fractions in all of the runs carried out in this study varied in general between 2.5 – 4% (for instance see Table 4). However, negligible CH_4 production throughout the domain was predicted in the simulations, which is attributable to the devolatilization model employed (Table 3). Calculations employing the

measured values showed that CH_4 contributed nearly 15 - 20% to the heating value of the outlet gas. Figure 7b, compares the experimentally calculated cold gas efficiencies (accounting for heating values of only CO and H_2) against numerical predictions. Experimentally and numerically, optimum cold gas efficiencies are observed at $\text{H}_2\text{O}/\text{C}$ ratios close to unity. Although higher CO concentrations were predicted numerically, differences in the flow rates of the product gases between the experiments and simulations, cause the measured cold gas efficiencies to be higher than the corresponding numerical predictions.

3.3 Optimizing steam flow rates

Having optimized the coal and oxygen flow rates for maximum hydrogen production, the mass flow rate of steam was varied to examine its impact on the syngas composition. Figure 8 shows contours of the bed volume fraction and gas temperatures at the three steam flow rates simulated in this study. The inlet gas velocity at $\text{H}_2\text{O}/\text{C}$ ratio of 2.3 was determined to be 0.22 m/s, which is greater than the minimum slugging velocity of 0.17 m/s estimated from theoretical calculations [12]. This results in inadequate mixing and lower temperatures in the lower regions of the reactor. Steam is expected to promote the WGS reaction and increase H_2 production and H_2/CO ratios. The measured and predicted major gas concentrations are compared in Figure 9. While both experiments and simulations correctly predicted a decrease in CO concentrations with an increase in $\text{H}_2\text{O}/\text{C}$ ratios, H_2 production was observed to attain its maximum value at $\text{H}_2\text{O}/\text{C}$ ratio close to unity. As the amount of steam is increased the system becomes saturated with steam and there is too much steam to react with the carbon available thereby decreasing hydrogen production. Further, the WGS reaction produces hydrogen and carbon dioxide at the expense of carbon monoxide, since the system is now saturated with hydrogen the carbon monoxide is consumed and converted to carbon dioxide, which is evident from Figure 9b. This is further highlighted in Figure 10a where the measured major gas concentrations across the $\text{H}_2\text{O}/\text{C}$ ratio investigated in this study are reported. Figure 10b compares the measured and predicted H_2/CO ratios and shows that the WGS reaction is promoted at higher steam flow rates. This strong influence of the $\text{H}_2\text{O}/\text{C}$ ratio on the H_2/CO ratios was also observed by Karimipour et al. [19] and Sandeep and Dasappa [21].

4. Conclusions

An experimental and CFD effort towards optimizing: hydrogen rich syngas production and cold gas efficiencies during the thermochemical conversion of a lignite coal when using oxygen and steam as gasifying agents was described in this study. The overall goal was to operate in a temperature window that takes advantage of the high Na and moisture content in the coal to promote carbon conversion and WGS reaction while minimizing slag formation and ash condensation downstream of the gasifier. The measurements reported in this study are from a bubbling bed gasifier that was systematically designed, constructed and commissioned to achieve the stated objectives. The walls of the gasifier were maintained at 1023 K, and the oxygen, coal, and steam flow rates varied sequentially (while maintaining the other flow rates and conditions constant) to identify the optimum operating conditions. The following conclusions may be drawn from this study:

1. Maximum H_2 concentration of 50% (by volume on a dry basis) was obtained at: 5.31×10^{-5} kg/s of oxygen, 2.04×10^{-4} kg/s of coal, 1.50×10^{-4} kg/s of steam. As the mass flow rates of oxygen or steam increased from these conditions, the H_2 concentrations decreased.
2. An increase in the oxygen flow rate increased the CO and H_2 oxidation rates through their dependence on the oxygen concentration. The resulting higher temperatures from the oxidation process do not favour H_2 formation through the WGS reaction.
3. H_2 formation, H_2/CO ratios, and the cold gas efficiencies increased in general with the steam flow rates by promoting the WGS reaction. However, the gasifier performance was optimized around $\text{H}_2\text{O}/\text{C}$ ratio of about 1.03. At higher steam flow rates, the bed hydrodynamics transitioned from a bubbling bed to a slug flow which impacted the mixing and kinetics, temperature distributions, and consequently the H_2 production.
4. Simulations revealed that the major product gases at the gasifier outlet were already close to their equilibrium levels. While the observed experimental trends in the major product gases (CO , CO_2 and H_2) were well predicted by the simulations they failed to predict the 2-3% CH_4 that was measured at the outlet under these operating conditions. This is likely due to inadequate CH_4 formation during

pyrolysis which could have been better predicted if coal was injected at the right location or through a better model for devolatilization. Therefore, the numerically predicted cold gas efficiencies (when accounting for CO and H₂ only) agreed well with their corresponding experimentally determined values.

5. Experimentally estimated coal gas efficiencies varied between 80 – 90% across the range of operating conditions investigated in this study which compares well with the 79% efficiency that was obtained with the TRIG gasifier.

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Table 1: Proximate, Ultimate and Ash Analysis of the ND lignite coal

Mineral Analysis of Ash		Dry basis (wt%)
Silicon Dioxide (SiO ₂)		19.23
Aluminum Oxide (Al ₂ O ₃)		9.2
Titanium Dioxide (TiO ₂)		0.33
Iron Oxide (Fe ₂ O ₃)		7.33
Calcium Oxide (CaO)		19.64
Magnesium Oxide (MgO)		5.07
Potassium Oxide (K ₂ O)		0.79
Sodium Oxide (Na ₂ O)		10.75
SO ₃		23.65
P ₂ O ₅		0.12
Strontium Oxide (SrO)		0.54
Barium Oxide (BaO)		0.47
Manganese (MnO ₂)		0.1
Proximate Analysis	As Received (wt%)	Dry basis (wt%)
Total Moisture	31.2	
Ash	6.38	9.27
Volatile Matter	28.82	41.89
Fixed Carbon	33.6	48.84
Total Sulfur	0.65	0.94
Ultimate Analysis	As Received (wt%)	Dry basis (wt%)
Total Moisture	31.2	
Ash	6.38	9.27
Carbon	47.83	69.52
Hydrogen	6.67	4.62
Nitrogen	0.52	0.76
Total Sulfur	0.65	0.94
Oxygen by Difference	37.95	14.89
Chlorine (µg/g)	31.6	45.9
Heating Value (BTU/lb)	7526	10939

Table 2: A summary of different models and settings employed in the CFD simulations

Physics being modeled	Modeling option [13]
Multiphase hydrodynamics	Euler-Euler
Turbulence	Standard k- ϵ (per phase)
Gas-phase chemistry	Finite rate/Eddy dissipation
Heterogeneous chemistry	Finite rate
Drag law (Gas – Sand)	Wen-Yu
Drag law (Gas – Coal)	Wen-Yu
Drag law (Coal – Sand)	Schiller-Naumann

Table 3: A summary of the reactions and their kinetic parameters modeled in this study

Heterogeneous reactions	Kinetic parameters A, E_a (J/kmol), [Reactant]ⁿ	Reference
Devolatilization: Volatiles $\rightarrow$ 0.13 CO + 0.044 CO ₂ + 0.238 H ₂ O + 0.034 CH ₄ + 0.403 H ₂	492000, 7.4e+07	14
Char combustion: C + O ₂ $\rightarrow$ CO ₂	6.51e+08, 90000, [C] ¹ [O ₂] ¹	10
Steam gasification: C + H ₂ O $\rightarrow$ CO + H ₂	6.2e+08, 110000, [C] ¹ [H ₂ O] ¹	10
Boudouard reaction: C + CO ₂ $\rightarrow$ 2CO	35000, 150000, [C] ¹ [CO ₂] ¹	10
Homogeneous reactions	Kinetic parameters A, E_a (J/kmol), [Reactant]ⁿ	Reference
CO Oxidation: 2CO + O ₂ $\rightarrow$ 2CO ₂	3.16e+12, 1.67e+08, [CO] ^{1.5} [O ₂] ^{0.25}	15
H ₂ Oxidation: H ₂ + 0.5O ₂ $\rightarrow$ H ₂ O	6.8e+15, 1.68e+08, [H ₂] ^{0.5} [O ₂] ^{2.25} [H ₂ O] ⁻¹	16
CH ₄ Oxidation: CH ₄ + 0.5O ₂ $\rightarrow$ CO + 2H ₂	4.4e+11, 1.25e+08, [CH ₄] ^{0.5} [O ₂] ^{1.25}	16
WGS Forward: CO + H ₂ O $\rightarrow$ CO ₂ + H ₂	5e+12, 2.83e+08, [CO] ^{0.5} [H ₂ O] ¹	17
WGS Reverse: CO ₂ + H ₂ $\rightarrow$ CO + H ₂ O	9.5e+10, 2.39e+08, [CO ₂] ¹ [H ₂] ^{0.5}	17
Methane-Shift Forward: CH ₄ + H ₂ O $\rightarrow$ CO + 3H ₂	4.4e+11, 1.68e+08, [CH ₄] ¹ [H ₂ O] ¹	16
Methane-Shift Reverse: CO + 3H ₂ $\rightarrow$ CH ₄ + H ₂ O	5.12e-14, 27300, [CO] ¹ [H ₂] ¹	18

Table 4: A summary of flow rates investigated in the CFD study

Run identifier	Coal flow rate (Kg/s)	O ₂ flow rate (Kg/s)	Steam flow rate (Kg/s)
Optimizing O ₂ flow rate			
O/C = 0.8	2.04e-04	5.31e-05	1.50e-04
O/C = 1.1	2.04e-04	7.43e-05	1.50e-04
O/C = 1.6	2.04e-04	10.61e-05	1.50e-04
Optimizing coal flow rate			
H ₂ O/C = 0.84	2.49e-04	5.31e-05	1.50e-04
H ₂ O/C = 1.03	2.04e-04	5.31e-05	1.50e-04
H ₂ O/C = 1.3	1.59e-04	5.31e-05	1.50e-04
Optimizing steam flow rate			
H ₂ O/C = 0.625	2.04e-04	5.31e-05	1.10e-04
H ₂ O/C = 1.03	2.04e-04	5.31e-05	1.50e-04
H ₂ O/C = 2.3	2.04e-04	5.31e-05	3.29e-04

Table 5: Measurements of product gas composition as a function of O/C ratio

Gas Constituent	Run 1	Run 2	Run 3	TRIG	Run 4	Run 5	Run 6
H ₂	49.2	42.2	38.5	37.8	37.6	33.7	31.6
CO	12.2	13.6	14.5	26	15.8	16.2	13.2
CO ₂	30.8	31.6	32.7	29.5	36	38.5	34.4
CH ₄	3.2	2.9	2.5	5.2	2.6	2.3	2.3
H ₂ S	0.4	0.6	0.6	0.5	0.7	0.6	0.5
N ₂	0.2	2.6	3.3	0.1	1.5	2.4	13.6
Temperature °C	750	750	750	900	750	750	750
O/C	0.8	0.9	1.1	0.34	1.2	1.4	1.6

Table 6: $K_{eq,WGS}$ predicted at the reactor outlet as a function of O/C ratio

	K_{WGS} (Eq. 2)	K_{WGS} (Prediction)
O/C = 0.8	1.3	1.2
O/C = 1.1	1.3	1.5
O/C = 1.6	1.3	1.4

(a)

(b)

Fig. 1: (a) The constructed fluidized bed reactor system; (b) Simulated geometry of the fluidized bed reactor.

(a)

(b)

Figure 2: Experimental variations as a function of O/C ratio: (a) Syn-gas composition; (b) Cold gas efficiencies.

Fig. 3: Comparing measurements and predictions at different O/C ratios: (a) H₂; (b) CO; (c) CO₂.

Fig. 4: Bed volume fractions at different O/C ratios showing a bubbling regime.

Fig. 5: Contours at different O/C ratios: (a) O₂ mole fractions; (b) CO oxidation rates (kmol-m³/s); (c) H₂ oxidation rates (kmol-m³/s).

(a)

(b)

(c)

Fig. 6: Comparing measurements and predictions at different H₂O/C ratios (coal flow rate optimization): (a) H₂; (b) CO; (c) CO₂.

Fig. 7: (a) Temperature contours (in K) at different H_2O/C ratios; (b) A comparison of cold gas efficiencies ($CO + H_2$ only) determined from measurements and predictions at different H_2O/C ratios.

Fig. 8: Contours at different H_2O/C ratios (steam flow rate optimization) (top) Volume of fraction of sand (bottom) gas temperature (K).

(a)

(b)

(c)

Fig. 9: Comparing measurements and predictions at different H₂O/C ratios (steam flow rate optimization): (a) H₂; (b) CO; (c) CO₂.

(a)

(b)

Fig. 10: Variations as a function of H₂O/C ratios (steam flow rate optimization): (a) Syngas composition (measurements); (b) H₂/CO ratios.