

See discussions, stats, and author profiles for this publication at: <https://www.researchgate.net/publication/231272827>

Simulation Study of an 800 MWe Oxy-combustion Pulverized-Coal-Fired Power Plant

ARTICLE *in* ENERGY & FUELS · APRIL 2011

Impact Factor: 2.79 · DOI: 10.1021/ef200023k

CITATIONS

20

READS

252

4 AUTHORS, INCLUDING:



Haibo Zhao

Huazhong University of Science and Technol...

115 PUBLICATIONS 898 CITATIONS

SEE PROFILE

Simulation Study of an 800 MW_e Oxy-combustion Pulverized-Coal-Fired Power Plant

Jie Xiong, Haibo Zhao,* Meng Chen, and Chuguang Zheng

State Key Laboratory of Coal Combustion, Huazhong University of Science and Technology, Wuhan, 430074 Hubei, People's Republic of China

ABSTRACT: Oxy-combustion technology has been considered as a feasible choice to reduce the CO₂ emissions from the coal-fired power plants through adding a cryogenic air separation unit (ASU) process and a flue gas treatment unit (FGU) process to a conventional combustion process. This technology is capable of enriching the CO₂ concentration and then allowing CO₂ sequestration in an efficient and energy-saving way. To study the operation characteristics of the oxy-combustion process, an 800 MW_e oxy-combustion pulverized-coal-fired power plant was simulated and analyzed using the commercial flowsheet software Aspen Plus. The base simulation result indicates that Aspen Plus is a proper tool to research the oxy-combustion system. The net efficiency of the oxy-combustion system is 10.36% [lower heating value (LHV)] lower because of the ASU and FGU processes; the unit power consumption for the oxygen production in the ASU is 0.247 kW h (kg of O₂)⁻¹. Many important parameters and different cases were also studied in this paper. The results show that a recycle ratio range of 0.705–0.726 is proposed for cold recycle cases, whereas 0.617 is recommended for the hot recycle case. The CO₂ concentrations of flue gases from the oxy-combustion boiler range from 84 to 92 mol % for cold recycle cases and from 57 to 58 mol % for hot recycle cases because of the high H₂O content, respectively. However, the CO₂ purity from the FGU for each case can reach 99 mol %. In addition, in comparison to the air-combustion case, oxy-combustion cases have higher SO_x fractions but a similar unit of the SO_x production rate (0.78 mol/kg of coal) and much lower NO_x fractions in the flue gas. Moreover, a sensitivity analysis about the concentration of the oxygen product from the ASU and an optimization of the distillation column in the ASU were also performed. The optimal value of the total stage number, the reflux ratio, and the number of feed stage for the distillation column in the 95 mol % oxygen case are 28, 1.04, and 15th, respectively. All of these results will be helpful to the oxy-combustion system design and operation.

1. INTRODUCTION

In 2008, the emission of CO₂ in China had reached about 6.55 gigatons (22.3% of the world's CO₂ emissions).¹ The largest CO₂ emission source in China is coal combustion power plants, which supply over 60% of the total energy. Oxy-combustion (or oxy-fuel) technology, combining a conventional pulverized-coal-fired power plant (PC) with a cryogenic air separation unit (ASU) and a flue gas treatment unit (FGU) (as shown in Figure 1), is one approach to produce high-purity CO₂ gas stream and finally control the CO₂ emissions. In the oxy-combustion technique, an oxygen-rich stream (greater than 95% by volume²) and recycled flue gases (about 70–80% of the flue gas³) are mixed and then fed to combustors, keeping the combustion temperature inside the furnace within the conventional range by appropriately adjusting the temperature and mass flow of two feed gases. Because the fuels are burned in a N₂-lean and CO₂- and H₂O-rich environment, the resulting flue gases consist primarily of CO₂ and water vapor. The remaining flue gas, which may contain more than 95% CO₂ in some bench-scale experiments,⁴ is first cooled to remove water, then compressed, followed by separation of noncondensable gases (Ar, O₂, and N₂) from CO₂, and finally boosted to pipeline pressure.⁵ Obviously, CO₂ capture from the highly CO₂-enriched flue gas is less energy-demanding than from conventional air-fired combustion flue gases with a high N₂ fraction and relatively low CO₂ fraction (13–15% by volume).^{4,6}

Research on oxy-combustion includes (1) combustion characteristics,^{7–10} (2) pollutant (SO_x, NO_x, CO₂, etc.) production

properties^{4,11,12} and impurity purification processes,^{13,14} (3) technical economic studies,^{2,3,15,16} etc. As an example of a technical economic study, Chalmers University has evaluated a retrofitted 865 MW_e lignite-fired power plant in Germany.² In the study, a cryogenic ASU was integrated into the power plant to produce pure O₂ required for combustion and the optimal concentration of product O₂ was proposed to be 95%. In comparison to 42.6% in the reference plant, the net efficiency becomes 33.5% in the oxy-combustion plant, and the CO₂ avoidance cost was calculated to be \$26/ton. Besides some lab- or pilot-scale experiments, there are also many demos¹⁷ or commercial-scale testing works.² One demo-size study¹⁷ is a 30 MW_{th} size plant built by Vattenfall. In the study, the CO₂ avoidance cost was 20–25 €/ton. The CO₂ emission control efficiencies were reported to be more than 90%.^{2,17} On the other hand, to study the holistic performance of an oxy-combustion system, a process simulation tool is necessary. Some commercial flowsheet software, such as Aspen Plus, Thermoflex, and Hysys, are usually used.

Thermoflex focuses on the thermodynamic process,¹⁸ and Hysys is suitable for the petroleum production and chemical process.¹⁹ Aspen Plus is a process simulation software suitable for a variety of steady-state modeling applications. It has the

Received: January 5, 2011

Revised: April 8, 2011

Published: April 12, 2011

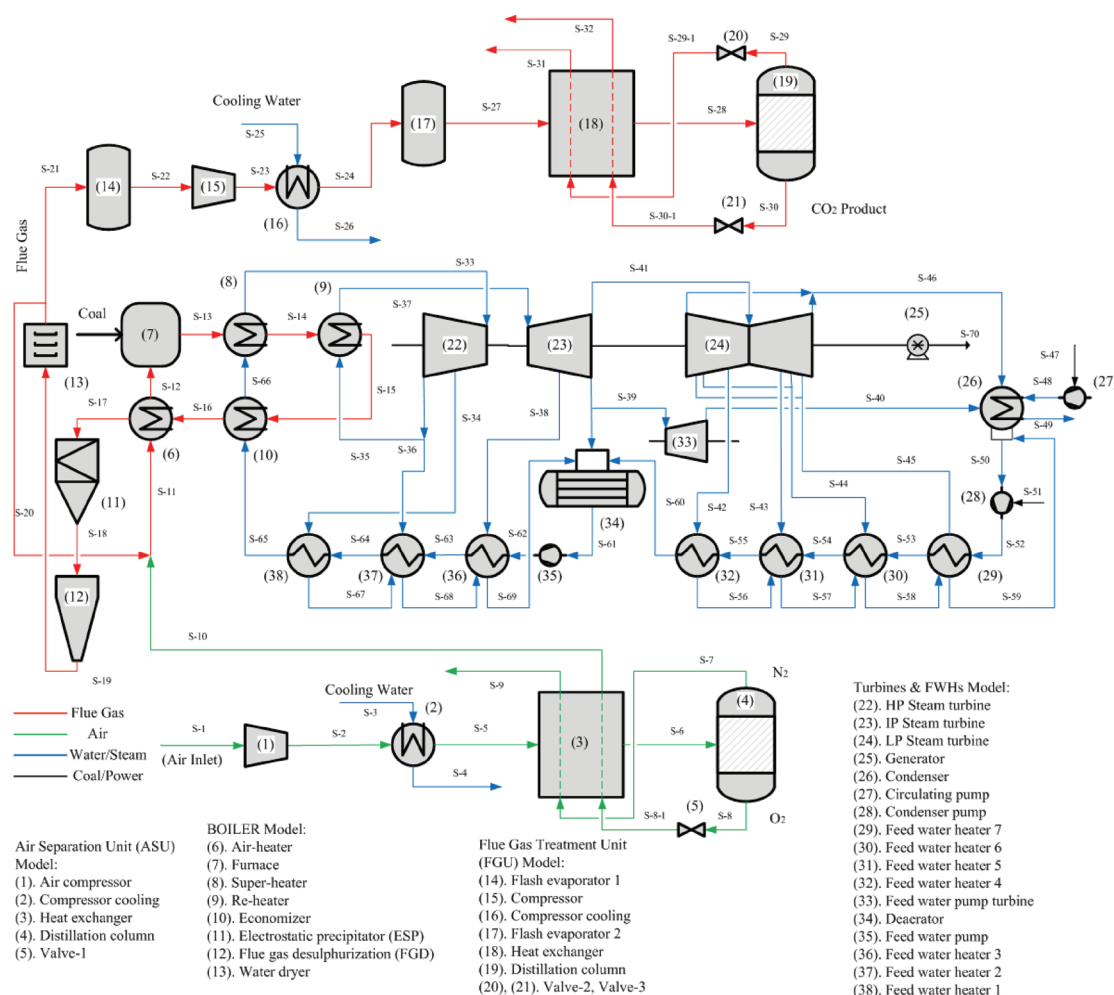


Figure 1. Schematic diagram of the oxy-combustion PC system.

advantage of comprising large data on chemical substances and models that are required for the calculation of low-temperature gas behavior in expansion turbines, determination of combustion products, electrolytic effects in the condensers, and phase equilibrium in the phase separators and rectification processes.²⁰ Many processes, such as CO₂ capture by amine scrubbing,^{21–24} integrated coal gasification combined cycle (IGCC),^{25–27} c;omined cycle co-generation plant,²⁸ and conventional steam power plant,²⁹ have been simulated and analyzed in Aspen Plus. However, the process simulation study of a complete oxy-combustion PC using Aspen Plus is still limited, because it is not specifically designed for energy system process simulation and its block-wise calculation may lead to a rather poor convergence performance.²⁰ One work³⁰ about the oxy-combustion PC employed both Aspen Plus and Thermoflex as the simulators. While the former is used to model the ASU and FGU, the latter focuses more on the modeling of steam generation and the power island, and a connection between the two simulators is thus needed. Another work²⁰ simulated a complete oxy-combustion PC, which is a retrofitted case based on an existing conventional PC, using Aspen Plus only. As a result, no interface to other systems is needed, and the closed simulation of all components in one model delivers a coherent solution with a significantly reduced number of assumptions. In the paper, thermodynamic and economic properties of the system are studied but sufficient

details for the combustion and heat transfer in the oxy-fuel steam generator are limited.

In this paper, Aspen Plus, version 7.1, was used to simulate an oxy-combustion PC introduced by a DOE/NETL report (DOE/NETL-2007/1291).³¹ On the basis of the simulation results of a base case (shown in section 3), sensitivity analysis (shown in section 4) was performed with regard to the O₂ concentration from ASU, recycle ratio of flue gas, recycle position, air ingress, and removal of conventional gaseous pollutants. Furthermore, the distillation columns in ASU and FGU are optimized for better thermodynamic and economic performances. Consequently, the feasibility of simulation by Aspen Plus for such a system combining chemical and thermodynamic processes was explored.

2. SYSTEM SIMULATION DESCRIPTION

The oxy-combustion PC studied in this paper is an 800 MW_e supercritical plant. It includes four sections: boiler, turbines and feed-water heaters (FWHs), ASU, and FGU, which are all shown in Figure 1. There are 38 components and 73 mass/energy streams in the system. The model of the oxy-combustion PC constructed by Aspen Plus is plotted in Figure 2. For the steam/water cycle, there are two high-pressure (HP) turbine stages, two intermediate-pressure (IP) turbine stages, and five low-pressure (LP) turbine stages. The deaerator is

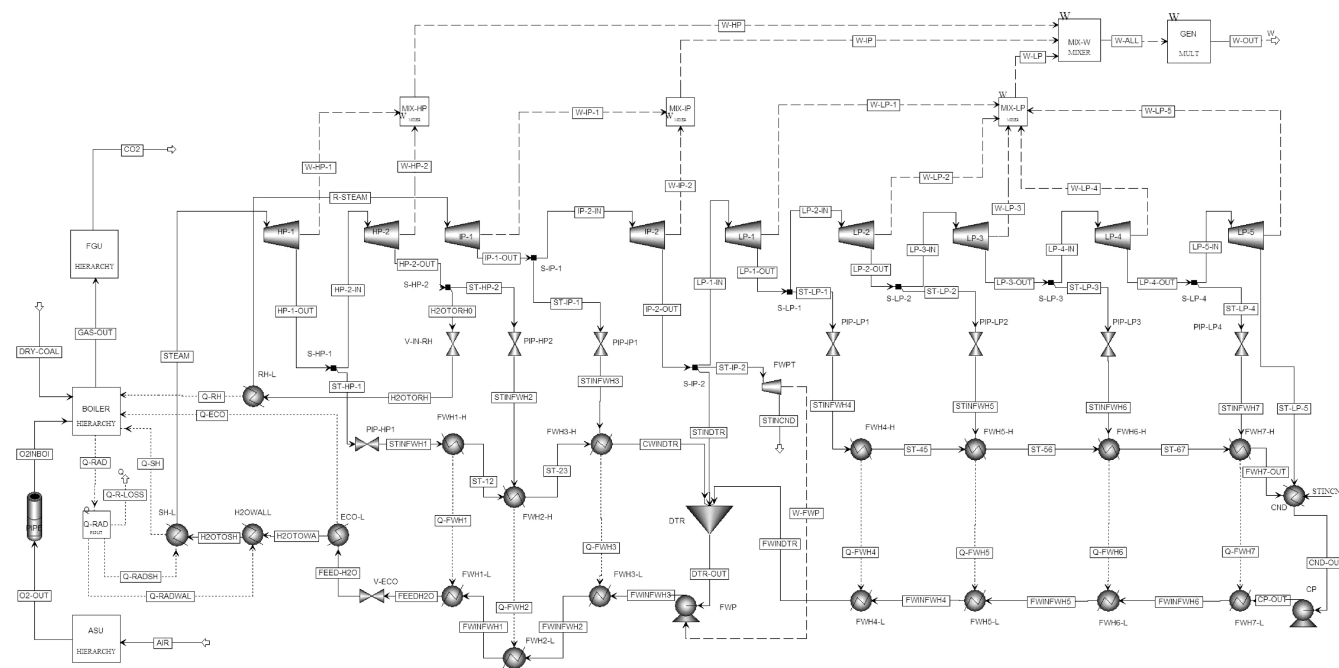


Figure 2. Simulation flowsheet of the oxy-combustion PC system.

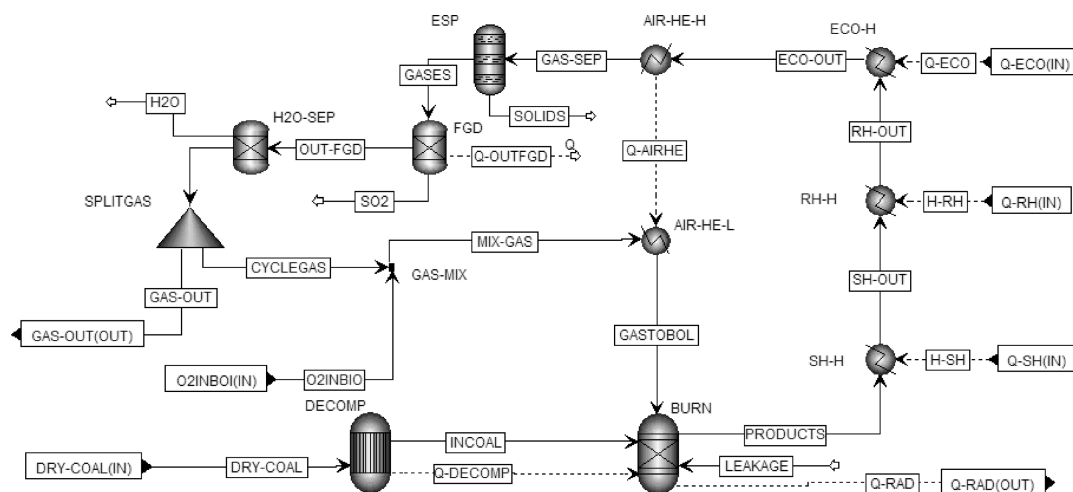


Figure 3. Simulation flowsheet of the boiler hierarchy.

simulated by a mixer block. In Aspen Plus, there is no existing model for FWHs. A pair of heater blocks is used to simulate a FWH. Moreover, to set and control the terminal temperature difference (TTD) in the FWH, tranfer and analyzer blocks and a design spec option should be used. TTD means the difference between the saturated temperature of the steam entering FWH and the temperature of the outlet feedwater in FWH.

For the boiler model (shown in Figure 3), there is no component corresponding to “coal” in the Aspen Plus component library and it is defined as a nonconventional component. The raw coal is decomposed to each element (e.g., H_2O , ASH , C , H_2 , N_2 , Cl_2 , S , and O_2) contained in the coal and then combusted in the furnace (unit 7) with recycled flue gas (S-20) and pure oxygen (S-10) from ASU. The flue gas goes through the superheater (SH, unit 8), reheater (RH, unit 9), economizer (ECO, unit 10), air heater (AH, unit 6), electrostatic precipitator (ESP, unit 11), flue gas desulfurization (FGD, unit 12), and water dryer (unit 13), and

then about 70% flue gas (S-20) is recycled back into the furnace. It is also a pair of heater blocks used to model the SH, RH, ECO, and AH in Aspen Plus. In the base case, SO_x and water are assumed to be removed completely in the FGD and water dryer, respectively, and it is assumed that there is 2% air ingress in the furnace. Some other cases about the boiler model will be discussed later.

There are four blocks in the ASU simulation (shown in Figure 4): multi-stage compressor (MCOM, units 1 and 2), heat exchanger (HEX, unit 3), distillation column (column, unit 4), and expansion valve (unit 5). In the MCOM, a configuration of four stages with intermediate cooling was devised. In addition, the discharge pressure from the final stage is set to be 6.3 bar. The isentropic efficiency and mechanical efficiency of each stage are 0.8 and 0.97, respectively, and the intermediate cooling temperature is 20 °C. In the HEX, the thermodynamic state of the compressed air flowing out is set to be the saturated vapor and the pressure drops of the three pairs of streamflow are all 0.1 bar. The

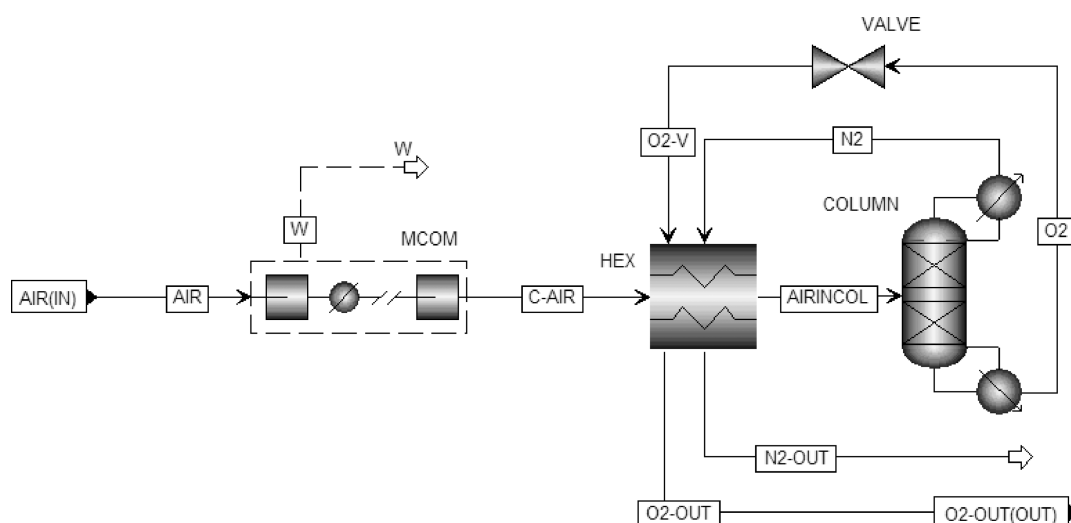


Figure 4. Simulation flowsheet of the ASU hierarchy.

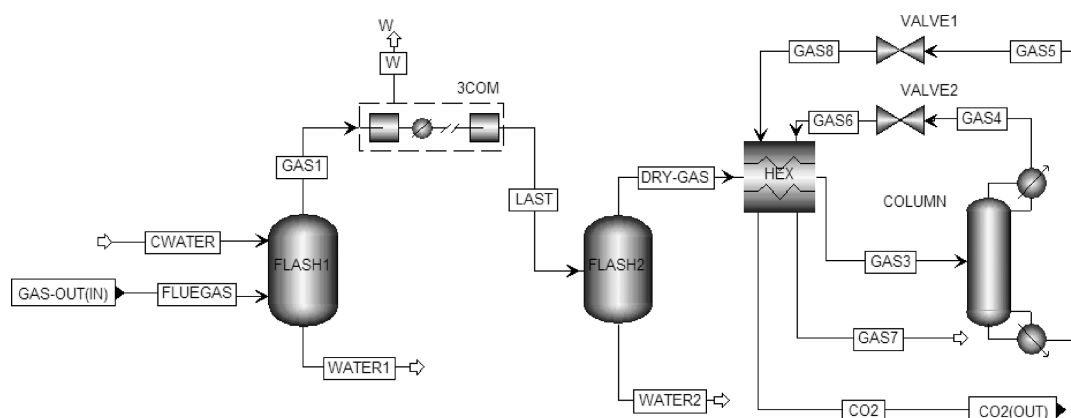


Figure 5. Simulation flowsheet of the FGU hierarchy.

discharge pressure of the expansion valve is set to be 1.5 bar. The distillation column is the most important unit in the ASU model. In the column, the bottom product is liquid O_2 and the top product is vapor N_2 . Obviously, there are some impurities in each product, and the O_2 concentration is designed to be greater than 95 mol %. Sensitivity analysis on the O_2 concentration will be discussed further. The mole ratio of the bottom product to feed stream (B/F) is set to be 0.215 in the simulation. There are three important parameters in the column simulation: feed stage, total number of stages, and reflux ratio. The three parameters should be fixed to satisfy the distillation requirement. Moreover, an optimization work, which will be discussed in section 4.6, can be performed on the column to obtain the optimal values of the three parameters: 15th, 28, and 1.04.

In the FGU process (shown in Figure 5), flue gas is cleaned, compressed, dried, and distilled to obtain the pure CO_2 . As a first step, flue gas is flashed with cooling water in a flash evaporator (unit 14). Then, it is compressed to 3 MPa in a three-stage compressor with intermediate cooling (units 15 and 16). The remaining water is removed by another flash evaporator (unit 17). After the flue gas flows through the HEX (unit 18), it is distilled in the column (unit 19). The CO_2 product (bottom) purity is designed to be 99%. The two product streams from the column are expanded in the two valves (units 20 and 21). There are also some important design parameters in this distillation column, and the value of the feed stage, total number of stages, boil-up ratio, and B/F is 1, 8, 0.38, and 0.83, respectively.

Three kinds of chemical properties were used in the simulation: Peng–Robinson with Boston–Mathias alpha function (PR–BM) for the boiler section, National Bureau of Standards (NBS)/National Research Council of Canada (NRC) steam table equation of state (STEAMNBS) for the turbines and FWHs section, and Peng–Robinson (PENG–ROB) for the ASU and FGU section. The calculation sequence of the simulation is described as follow. First, the thermodynamic parameters of the feedwater, main steam, and RH steam are used to calculate the heat needed, and the coal consumption amount can be obtained using its heating value. Then, the coal consumption amount is used to obtain the oxygen consumption. Finally, a “design spec” option is adopted to obtain the flow rate of the raw air (S-1) flowing into the ASU.

3. SIMULATION RESULTS

The coal used in this study is Illinois No. 6 bituminous coal, the proximate and ultimate analyses of which are all listed in Table 1. In the table, all data are on the as-received (ar) basis. C, H, O, N, S, and Cl mean carbon, hydrogen, oxygen, nitrogen, sulfur, and chlorine in the coal, respectively. HHV (LHV) is the higher (lower) heating value of the raw coal. Moreover, some basic inputs needed in the simulation, such as turbine heat balance, are given in Table 2.

Table 1. Proximate and Ultimate Analyses of Coal (ar Basis)

proximate analysis (wt %)		ultimate analysis (wt %)	
moisture	11.12	C	63.75
volatile matter	34.99	H	4.50
ash	9.70	O	6.88
fixed carbon	44.19	N	1.25
LHV (kJ/kg)	26171	S	2.51
HHV (kJ/kg)	27135	Cl	0.29

Table 2. Basic Inputs Needed in the Simulation

item	value
SH steam	598.89 °C, 242.35 bar, and 625.994 kg/s
RH steam	621.11 °C and 45.09 bar
O ₂ excess factor	1.05 ³⁶
furnace outlet	1100 °C and 1 atm
recycle ratio	0.705
air ingress	15 °C, 1 atm, and 2% of total gas supplied in the boiler
condenser outlet	38.74 °C and 0.368 bar
turbine stage discharge	77.07, 49.02, 21.36, 9.515, 5.013,
pressure (HP1–LP5) (bar)	1.323, 0.5771, 0.2473, and 0.0689
TTD (FWH1–FWH7) (°C)	−1.111, 0, −1.111, 2.778, 2.778, 2.778, and 2.778
generator efficiency (%)	98.58

As mentioned above, the coal feed rate is calculated on the basis of the mass flow rates and enthalpies of the SH and RH steams, as well as the heating value of the coal, which is described as below

$$M_C = (M_{65}(h_{33} - h_{65}) + M_{35}(h_{37} - h_{35})) / (\eta_b \text{LHV}) \quad (1)$$

in which “*M*” means mass flow rate (kg/s), “*h*” means enthalpy (kJ/kg), “ η_b ” means the thermal efficiency of the boiler (0.93 in this paper), subscript “*C*” means coal, and numerical subscripts mean the numbers of the streams shown in Figure 1. Then, the theoretic volume of oxygen needed for the coal burning can be calculated using the relation as shown in eq 2.

$$v_O = (C_{ar}/12 + H_{ar}/4 + S_{ar}/32 - O_{ar}/32) \times 22.4 \quad (2)$$

For the base case, there are three oxygen sources for coal combustion: recycled flue gas, pure oxygen from ASU, and air ingress. Because of the flue gas recycle, an iteration convergence option (design spec) is needed to calculate the oxygen amount from ASU and then the raw air amount flowing into the ASU.

After the system diagram is built, the sections and components are defined, the chemical properties are chosen, and the values needed are provided, the final step is to run the process and obtain results. Tables 3 and 4 show some important simulation results for the base case. In Table 3, only compressors and pumps are considered in calculating the power consumption. The results show that the net efficiency is 10.36% lower because of the ASU and FGU processes.

4. DISCUSSION

4.1. Recycle Ratio. In the base case, 0.705 is chosen for the recycle ratio (illuminated in Table 2). The recycle ratio is defined

Table 3. Simulation Results for the Base Case

power generated (MW)	
gross system	799.23
HP (19)	245.99
IP (20)	234.20
LP (21)	319.03
power consumed (MW)	
ASU (1)	133.70
FGU (14)	58.67
circulating pump (24)	5.68
condenser pump (25)	1.00
total	199.05
coal feed rate (kg/s)	70.95
oxygen stoichiometric amount (kmol/s)	4.47
raw air flowing into the ASU (kmol/s)	21.74
net fuel input (MW)	1856.83 (LHV)
gross efficiency (%)	43.04 (LHV)
net efficiency (%)	32.32 (LHV)
unit power consumption for	
oxygen production [kW h (kg of O ₂) ^{−1}]	0.247
unit power consumption for CO ₂ emission control [kW h (kg of CO ₂) ^{−1}]	0.330
CO ₂ emission control efficiency (%)	96.84

as the mass flow rate ratio of the recycled flue gas to the total flue gas produced, viz., $M_{20}/(M_{20} + M_{21})$. In this section, a sensitivity analysis of the recycle ratio is studied and the change range is set to be 0.7–0.75, 0.005 for step size. Three parameters were observed: O₂ content in S-11 (mixing) and O₂ and CO₂ contents in S-21 (flowing out the boiler). The results (lines “w air in”) are shown in Figure 6. It shows that the O₂ content in S-11 is strongly correlated with the recycle ratio. Moreover, when the recycle ratio is 0.705, the O₂ content in S-11 is about 30%, and this value is suggested by some researchers^{2,32,33} because the resultant combustion temperature in the furnace is closer to that in air-fired combustion. In this case, the O₂ mole fraction in S-21 is 1.56%. In this paper, the recycle ratio relating to a 30 mol % O₂ content in the mixing gases (S-11) is considered to be an optimal value. In addition, as the recycle ratio increases, the power consumptions for ASU and FGU all decrease but the influence is very low.

For the oxy-combustion technology, the recycle ratio affects the combustion characteristic in the furnace and flue gas composition; therefore, it is a very important parameter for the system operation. In addition, the recycle ratio chosen is also related to the coal type fired in the boiler.

4.2. Oxygen Concentration. The effect of the oxygen concentration to the system configuration and simulation results is discussed in this section, and the range of the concentration is set to be 0.95–0.98, 0.005 for step size. It should be emphasized that, in each concentration case, the recycle ratio is also changed to adapt the mole-based O₂ concentration of 30% in the feeding mixture gases into the furnace. Figure 7 shows the optimal recycle ratio in each oxygen concentration case, and Figure 8 shows the results obtained for different oxygen concentration cases with a 0.705 recycle ratio. The results indicate that the optimal recycle ratios range in 0.705–0.715, relating to a 95–98% O₂ concentration. Furthermore, the influence of the oxygen concentration from the ASU on the O₂ contents in S-21

Table 4. Simulation Results for Flue Gas Composition

component	value (mole fraction)							
	oxy-firing (with 0.705 recycle ratio)							
	air firing	S-13			S-21		S-11	
		ingress (LIFAC)	ingress	no ingress	ingress	no ingress	ingress	no ingress
N ₂	0.737	0.085	0.085	0.022	0.097	0.025	0.072	0.022
O ₂	0.018	0.012	0.014	0.015	0.016	0.017	0.301	0.322
NO	1.93×10^{-4}	5.29×10^{-5}	5.68×10^{-5}	2.95×10^{-5}	6.47×10^{-5}	3.39×10^{-5}	4.49×10^{-5}	2.28×10^{-5}
SO ₂	2.20×10^{-3}	1.29×10^{-4}	3.21×10^{-3}	3.43×10^{-3}	0	0	0	0
SO ₃	2.24×10^{-5}	1.06×10^{-6}	2.82×10^{-5}	3.12×10^{-5}	0	0	0	0
HCl	2.43×10^{-4}	1.15×10^{-3}	1.16×10^{-3}	1.24×10^{-3}	1.32×10^{-3}	1.43×10^{-3}	9.17×10^{-4}	9.61×10^{-4}
CO ₂	0.152	0.751	0.745	0.797	0.848	0.916	0.589	0.617

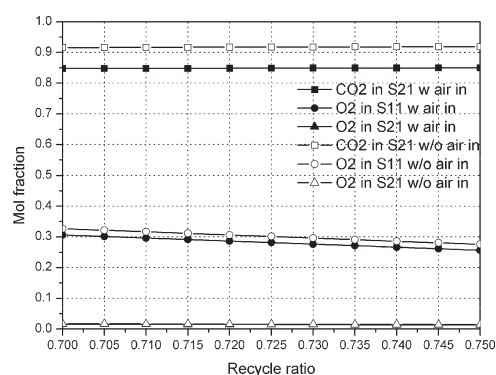


Figure 6. Sensitivity analysis for the recycle ratio.

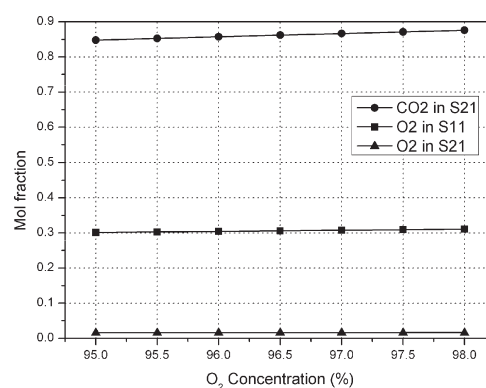
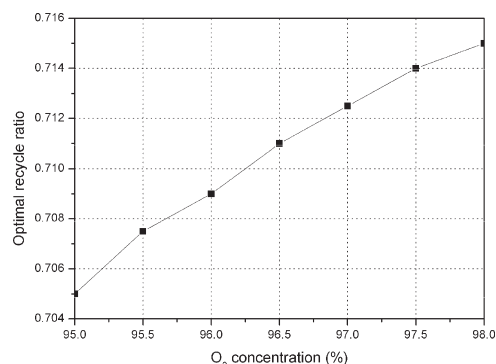
Figure 8. Sensitivity analysis for the O₂ concentration.

Figure 7. Optimal recycle ratios relating to different oxygen concentrations.

and S-11 are very low, whereas for the CO₂ content in flue gas, the influence is bigger.

The 95 mol % O₂ concentration should be high enough for the oxy-combustion system operation. Actually, not the higher, the better. A higher O₂ concentration usually means a higher power consumption and investment, and an optimal O₂ concentration might be obtained when the power consumptions in different units and the investment are considered together. However, this topic is beyond the scope of this paper.

4.3. Air Ingress. In the base case, a 2% air ingress is assumed (negative pressure). In this situation, N₂ dilutes the flue gas a little and the purity of CO₂ in the flue gas is not very high.

To avoid the air ingress, a positive gauge pressure for the boiler operation could be considered. However, it should be illuminated that a poisoning protection should be kept in mind during the positive operation because of the emissions of some poisonous gases, such as CO, SO₂, and H₂S. In this section, a 1.1 atm operation pressure for the boiler is chosen and the air ingress is thus avoided. The simulation results relating to S-13, S-21, and S-11 are shown in Table 4. The results show that O₂ and CO₂ contents in the three streams are all greater without air ingress because of the much lower N₂ content. Moreover, the CO₂ mole fraction in the S-21 can exceed 90%.

Additionally, the recycle ratio is also studied to search for the optimal value in the case without air ingress, and the results (lines "w/o air in") are shown in Figure 6. When the results under the two different cases are compared, each pair of lines has a similar change rate and tendency with the change of the recycle ratio. Without air ingress, the optimal recycle ratio is about 0.726, which is a little greater than that in the air ingress case. Then, the O₂ mole fraction in S-21 is 1.56%, which is the same value as that in the air ingress case. Besides the results in Table 4 and Figure 6, some other results relating to the streams flowing in/out of the furnace are shown in Figure 9.

The air ingress indeed should be avoided for a better operation property in the oxy-combustion system, but in this case, a seal action should be performed during the boiler operation. For the conventional air-fired boiler, no seal action exists. Therefore, this might be a problem for the oxy-combustion boiler design.

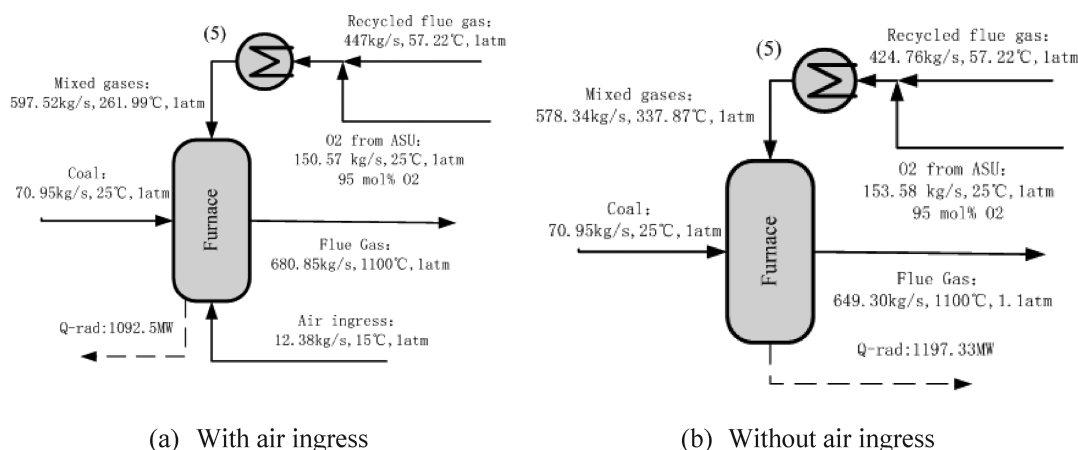


Figure 9. Simulation results relating to the streams flowing in/out of the furnace.

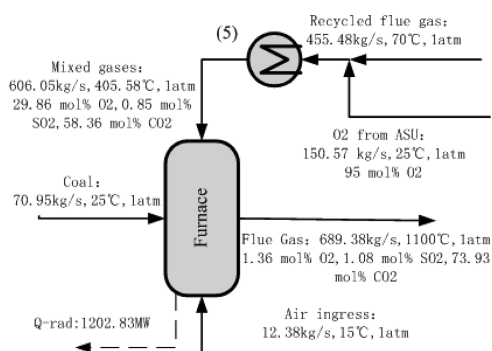


Figure 10. Simulation results about the furnace with SO_x treated by the FGU process.

4.4. SO_x and NO_x . Table 4 shows the content of SO_x and NO_x in the flue gas for different cases. The mole fractions of NO_2 in each stream are less than 10^{-6} ; therefore, the values are not shown in the table. In comparison to the air-firing (with a 1.1 air excess factor) environment, the NO content in flue gas is much lower in oxy-firing cases. In the base case, a wet FGD method is chosen to remove SO_x in the flue gas. In this situation, the concentrations of SO_x increase a little in oxy-firing cases. However, the unit SO_x production rates for air firing and oxy-combustion are nearly equivalent (0.78 mol/kg coal), whereas for the NO_x production rate, it is 1.37×10^{-2} mol/kg coal for the oxy-combustion case compared to 6.79×10^{-2} mol/kg coal for the air-firing case.

Because of the concentration increase of SO_x in the furnace, another SO_x removal option, limestone injection into the furnace and the activation of unreacted calcium (LIFAC),³⁴ could be performed, the simulation result of which is also listed in Table 4. In this option, limestone is injected into the furnace and the Ca/S ratio is set to be 1.05. The results show that SO_x can be removed to be a very low concentration in the flue gas in this case.

Another SO_x treatment option in oxy-combustion PC is co-separation with other impurities in the FGU process or co-sequestration with CO_2 . In this section, this option is also considered. SO_x generated in the boiler was not treated until it flows into the FGU process. The simulation results show that the power consumption for the FGU increases 1.3% comparing to that in the base case. It is worth noting that the CO_2 purity from the FGU in this case is 97.61 mol % (1.46% for SO_2) using the

same design parameter values for the distillation column as those in the base case. That is because it is difficult to separate CO_2 and SO_2 . Furthermore, the corrosion protection about the FGU should be considered because of the high acidity of the flue gas. The temperature of the flue gas in the FGU is below 70 °C, and SO_2 is in the liquid phase if its temperature is below 103 °C with a 3 MPa pressure; therefore, the left SO_2 flowing into the distillation column goes with the CO_2 product. Figure 10 shows the simulation results relating to the streams flowing in/out of the furnace in this case.

In addition, the results in Table 4 show that the NO content in any stream of the three is lower in the oxy-firing case without air ingress, because of the lack of N_2 , as compared to the oxy-firing case with 2% air ingress. However, the content of SO_x is a little greater without air ingress.

In the base case, a 100% SO_x removal efficiency in the FGD, mass flow rate ratio of the SO_x removed to the SO_x flowing into the FGD, is assumed. This removal efficiency is defined as

$$\eta_s = (M_{S-18} - M_{S-19})/M_{S-18} \quad (3)$$

in which subscript “S” means SO_x , and this removal efficiency is changed in a range of 80–100% in this section. The relation between the SO_2 content in the S-13 and the removal efficiency with a 0.705 recycle ratio is shown in Figure 11. As the removal efficiency decreases, the SO_2 content in the flue gas flowing out the furnace increases but the increase rate is slight. Furthermore, as the recycle ratio decreases, the SO_2 content also increases, regardless with or without air ingress, which is shown in Figure 12. Without air ingress, the SO_2 content is a little higher than that in the air ingress case. Taking one case for example, with a 90% removal efficiency and a 0.705 recycle ratio, the mole fraction of SO_2 is 3.45×10^{-3} , which is about 1.5 times of that in the air-firing case.

The oxy-combustion technology decreases the NO_x production and increases the SO_x mole fraction in the flue gas. Therefore, the LIFAC method and even the FGU co-separation method could be chosen to remove SO_x in the oxy-combustion system for a better economic property. Moreover, there might be no more treatment conducted to NO_x in the flue gas.

4.5. Recycle Position. The recycle position in the base case is after the FGD and the water dryer; therefore, the recycled flue gas is dry and cold. In this section, a hot (wet) recycle case is introduced, the recycle position of which is after the ECO. Flue

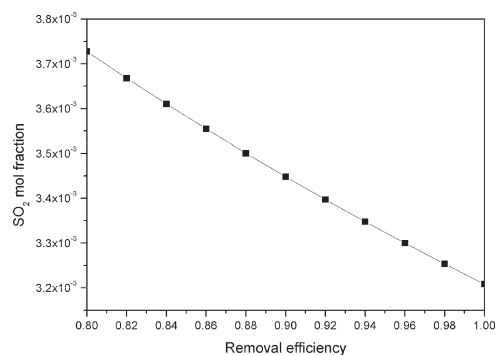


Figure 11. Relation between the SO₂ content in flue gas and the SO₂ removal efficiency.

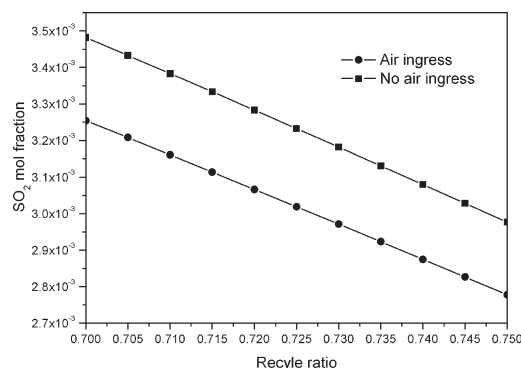


Figure 12. Relation between the SO₂ content in flue gas and the recycle ratio.

gas after the ECO is hot and wet; therefore, the air heater and the water dryer could be removed in the boiler model. The new boiler model diagram is shown in Figure 13, and the values of some thermodynamic parameters of the mass streams in the model (with a 0.705 recycle ratio) are also given. Because the flue gas flowing into the FGU contains much water, the multi-stage compressor in the FGU model should be placed after the flash units. The simulation results of the flue gas composition are shown in Figure 14. The "O₂ in S-11" line shows that a 30 mol % O₂ fraction in the mixing stream flowing into the furnace can be reached with a recycle ratio of 0.62. Additionally, the CO₂ mole fraction in the flue gas flowing out of the furnace is less than 60% because of the high H₂O content. It should be emphasized that the SO₂ content in the hot recycle case is much greater than that in the cold recycle case. Moreover, for the hot recycle case, the recycle ratio almost does not affect the SO₂ content in the flue gas.

Furthermore, the values in Figure 13 show that the temperature of the flue gas in the recycle part is very high, which is higher than the sulfur dew point in this situation (about 130 °C), whereas for the cold recycle case, the flue gas in the recycle part has a much lower temperature, which is below its sulfur dew point. Therefore, the hot recycle case would be a better choice from the viewpoint of corrosion protection.

4.6. ASU Design, Simulation, and Optimization. The configuration and some parameters of the ASU model have been introduced above. In this section, the distillation column is studied in detail and an optimization is conducted. Three parameters were chosen to optimize the column: feed stage, total number of stages, and reflux ratio. The reflux ratio is defined

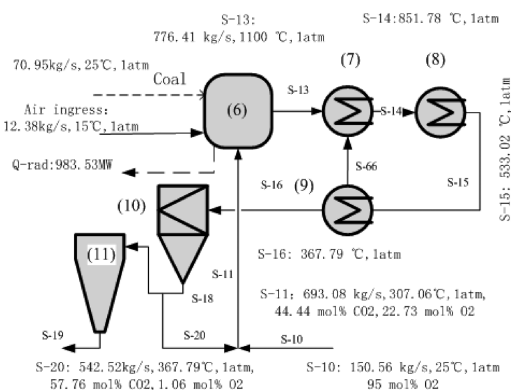


Figure 13. Simulation flowsheet of the boiler model with a hot recycle.

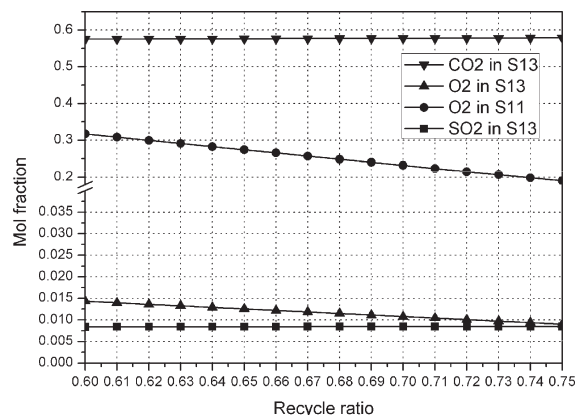


Figure 14. Simulation results relate to different recycle ratios during the hot recycle case.

Table 5. Simulation Results for Different Total Stage Numbers

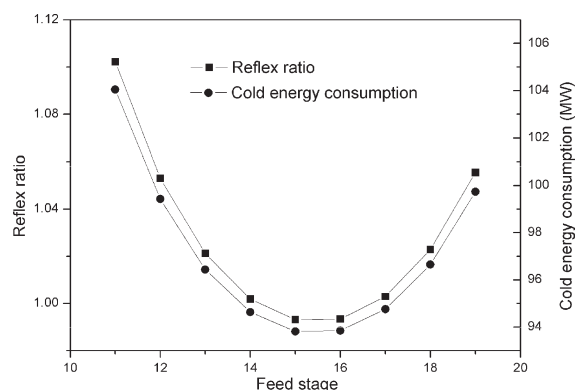
total stage number	30	20	16	14	13
feed stage	15	10	8	7	7
reflux ratio	0.97	1.42	2.58	6.86	>100

as the mole ratio of the reflux liquid to the product flowing out from the condenser in the distillation column. The methods to search the optimal values are presented as follows. It should be emphasized that the optimal values obtained in this section are for the 95 mol % oxygen, and there are different corresponding optimal values for other oxygen concentrations.

4.6.1. Optimization of the Total Number of Stages. For a distillation process, there is a minimal number of total stages to satisfy the distillation requirement. The minimal number can be found by decreasing the number of stages until the required reflux ratio becomes very large³⁵ (assumed to be larger than 100 in this paper). The simulation results are shown in Table 5. The feed stage is assumed to be a fixed ratio of the total number of stages.³⁵ The result shows that the minimal number of total stages is 13. Therefore, the optimal number of total stages is 28 (13 × 2 + 2).³⁵ The product purities and the column height are all related to the number of total stages. As the number of total stages reduces, the product purities would decrease. However, to satisfy the distillation requirement, the reflux ratio increases correspondingly and the increase rate becomes bigger and bigger.

Table 6. Simulation Results for Different Reflux Ratios

total stage number	30	40	50	60	70
feed stage	15	20	25	30	35
reflux ratio	0.97	0.89	0.87	0.87	0.87

**Figure 15.** Cold energy consumptions and reflux ratios with different feed stages.

4.6.2. Optimization of the Reflux Ratio. There is also a minimal reflux ratio, and it can be found by increasing the number of total stages until there is no further reduction in the reflux ratio.³⁵ The simulation results are shown in Table 6. The ratio of the feed stage to total stage is also fixed. The result shows that the minimal reflux ratio is 0.87. Therefore, the optimal reflux ratio is 1.04 (0.87×1.2) according to ref 35. Similarly, the product purities, the column diameter, and the condenser energy consumption are related to the reflux ratio. As the number of total stages increases, a smaller reflux ratio is enough to reach the distillation requirement. However, the reflux ratio decreases slower and slower and finally stops to be the minimal value.

It is obvious that the minimal number of total stages and the minimal reflux ratio obtained should not be chosen for the distillation column design. They are all critical values. The corresponding optimal values should be considered.

4.6.3. Optimization of the Feed Stage. In the distillate column, a reboiler and a condenser are included. There is a heat-exchange process between the reboiler and the condenser in the column. In addition, the column is designed to be the status that there is no more energy needed for the distillation requirement by varying the thermodynamic state of the inlet air (S-6) of the column. A less heat exchange is desired; therefore, the feed stage that minimizes the condenser cold energy is optimal for this distillate column. To find this feed stage, the optimal number of total stages of 28 is chosen in the simulation, B/F is also 0.215, and the required reflux ratio will change with the change of the feed stage. The simulation results are shown in Figure 15. The results indicate that the minimal cold energy consumption occurs at the 15th feed stage; therefore, the optimal feed stage is the 15th stage, and the reflux ratio is then 0.99.

The distillation column is also an important unit in the FGU model, and the optimization process for the FGU model is similar to that for the ASU model.

5. CONCLUSION

An oxy-combustion PC system including an ASU and a FGU was simulated in a Aspen Plus platform. After the base case

simulation work was achieved, a variety of other cases relating to recycle ratio, oxygen concentration, air ingress, SO_x removal option, and recycle position were also discussed.

The simulation results for the cold recycle cases show that the CO_2 concentration in the flue gas from the boiler can be more than 80% and the purity of the CO_2 product from FGU can reach 99%. With a 2% air ingress, the recycle ratios of 0.705–0.715 are proposed for different oxygen concentrations, whereas without air ingress, 0.726 is the most suitable recycle ratio value for the 95% oxygen concentration case. As the oxygen concentration increases, the optimal recycle ratio increases. Without air ingress, the CO_2 concentration in the flue gas from the boiler can exceed 90%.

In oxy-combustion, the NO_x content in the flue gas is much smaller than that in the air combustion case and, although the SO_x fraction in the flue gas is greater than that in the air combustion case, their unit SO_x production rates are equivalent. In this case, a LIFAC method or a FGU co-capture method, instead of the wet FGD method, could be adopted to remove SO_x in the flue gas.

A hot recycle case was also simulated, and in this case, the CO_2 concentration in the flue gas is less than 60% because of the dilution of H_2O but the CO_2 product purity from the FGU can also reach 99%. In comparison to the cold recycle case, the SO_x fraction in the flue gas and the temperature of the recycled flue gas increase much in the hot recycle case. Considering the corrosion protection and the sulfur dew point in the flue gas, the hot recycle method could be a better choice for the oxy-combustion technique.

In addition, optimization work was performed for the distillation columns in the ASU and FGU for better thermodynamic and economic properties. Different optimal parameter values for distillation columns in different oxygen product purity cases were obtained.

AUTHOR INFORMATION

Corresponding Author

*Telephone: +86-27-8754-4779. Fax: +86-27-8754-5526.
E-mail: klinsmannzhb@163.com.

ACKNOWLEDGMENT

The authors were supported by the National Key Basic Research and Development Program (Grant 2011CB707300), National Natural Science Foundation (Grants 50936001 and 50721005), and New Century Excellent Talents in University (Grant NECT-10-0395) for funds. Special thanks are given to Alstom Power Boiler R&D Execution (Windsor, CT) for providing an 1 year internship and the Aspen Plus software. The authors also acknowledge Dr. Shin G. Kang, Dr. Glen D. Jukkola, and Dr. Wei Zhang from Alstom Power Boiler R&D Execution for their valuable suggestions to this paper.

NOMENCLATURE

AH = air heater
ASU = air separation unit
B/F = mole ratio of the bottom product to feed stream
ECO = economizer
ESP = electrostatic precipitator
FGD = flue gas desulfurization
FGU = flue gas treatment unit

FWH = feed water heater
 HEX = heat exchanger
 HHV (LHV) = higher (lower) heating value
 HP, IP, and LP = high pressure, intermediate pressure, and low pressure
 IGCC = integrated coal gasification combined cycle
 LIFAC = limestone injection into the furnace and the activation of unreacted calcium
 MCOM = multi-stage compressor
 PC = pulverized-coal-fired power plant
 RH = reheater
 SH = superheater
 TTD = terminal temperature difference

Scalars

h = enthalpy (kJ/kg)
 M = mass flow rate (kg/s)

Greek Letters

η = efficiency

Subscripts

ar = as-received basis
 b = boiler
 C = coal
 O = oxygen
 S = sulfur oxides

REFERENCES

- (1) International Energy Agency (IEA). *CO₂ Emissions from Fuel Combustion Highlights*; IEA: Paris, France, 2010.
- (2) Andersson, K.; Johnsson, F. Process evaluation of an 865 MW_e lignite fired O₂/CO₂ power plant. *Energy Convers. Manage.* **2006**, *47* (18–19), 3487–3498.
- (3) Buhre, B. J. P.; Elliott, L. K.; Sheng, C. D.; Gupta, R. P.; Wall, T. F. Oxy-fuel combustion technology for coal-fired power generation. *Prog. Energy Combust. Sci.* **2005**, *31* (4), 283–307.
- (4) Hu, Y.; Naito, S.; Kobayashi, N.; Hasatani, M. CO₂, NO_x and SO₂ emissions from the combustion of coal with high oxygen concentration gases. *Fuel* **2000**, *79* (15), 1925–1932.
- (5) Damen, K.; van Troost, M.; Faaij, A.; Turkenburg, W. A comparison of electricity and hydrogen production systems with CO₂ capture and storage. Part A: Review and selection of promising conversion and capture technologies. *Prog. Energy Combust.* **2006**, *32* (2), 215–246.
- (6) International Energy Agency (IEA). *Improvement in Power Generation with Post-combustion Capture of CO₂*; IEA Greenhouse Gas R&D Programme (IEAGHG): Cheltenham, U.K., 2004; Report PH4/33.
- (7) Ditaranto, M.; Hals, J. Combustion instabilities in sudden expansion oxy-fuel flames. *Combust. Flame* **2006**, *146* (3), 493–512.
- (8) Kim, H. K.; Kim, Y. Studies on combustion characteristics and flame length of turbulent oxy-fuel flames. *Energy Fuels* **2007**, *21* (3), 1459–1467.
- (9) Hjartstam, S.; Andersson, K.; Johnsson, F.; Leckner, B. Combustion characteristics of lignite-fired oxy-fuel flames. *Fuel* **2009**, *88* (11), 2216–2224.
- (10) Smart, J. P.; O'Nions, P.; Riley, G. S. Radiation and convective heat transfer, and burnout in oxy-coal combustion. *Fuel* **2010**, *89* (9), 2468–2476.
- (11) Normann, F.; Andersson, K.; Leckner, B.; Johnsson, F. High-temperature reduction of nitrogen oxides in oxy-fuel combustion. *Fuel* **2008**, *87* (17–18), 3579–3585.
- (12) Andersson, K.; Normann, F.; Johnsson, F.; Leckner, B. NO emission during oxy-fuel combustion of lignite. *Ind. Eng. Chem. Res.* **2008**, *47* (6), 1835–1845.
- (13) Sass, B. M.; Farzan, H.; Prabhakar, R.; Gerst, J.; Sminchak, J.; Bhargava, M.; Nestleroth, B.; Figueroa, J. Considerations for treating impurities in oxy-combustion flue gas prior to sequestration. *Energy Procedia* **2009**, *1* (1), 535–542.
- (14) Darde, A.; Prabhakar, R.; Tranier, J. P.; Perrin, N. Air separation and flue gas compression and purification units for oxy-coal combustion systems. *Energy Procedia* **2009**, *1* (1), 527–534.
- (15) Xiong, J.; Zhao, H.; Zheng, C.; Liu, Z.; Zeng, L.; Liu, H.; Qiu, J. An economic feasibility study of O₂/CO₂ recycle combustion technology based on existing coal-fired power plants in China. *Fuel* **2009**, *88* (6), 1135–1142.
- (16) Singh, D.; Croiset, E.; Douglas, P. L.; Douglas, M. A. Techno-economic study of CO₂ capture from an existing coal-fired power plant: MEA scrubbing vs. O₂/CO₂ recycle combustion. *Energy Convers. Manage.* **2003**, *44* (19), 3073–3091.
- (17) Stromberg, L.; Lindgren, G.; Jacoby, J.; Giering, R.; Anheden, M.; Burchhardt, U.; Altmann, H.; Kluger, F.; Stamatiopoulos, G. N. Update on Vattenfall's 30 MW_{th} oxyfuel pilot plant in Schwarze Pumpe. *Energy Procedia* **2009**, *1* (1), 581–589.
- (18) Nord, L. O.; Kothandaraman, A.; Herzog, H.; McRae, G.; Bolland, O. A modeling software linking approach for the analysis of an integrated reforming combined cycle with hot potassium carbonate CO₂ capture. *Energy Procedia* **2009**, *1* (1), 741–748.
- (19) West, A. H.; Posarac, D.; Ellis, N. Assessment of four biodiesel production processes using HYSYS.Plant. *Bioresour. Technol.* **2008**, *99* (14), 6587–6601.
- (20) Rodewald, A.; Kather, A.; Frie, S. Thermodynamic and economic aspects of the hard coal based oxyfuel cycle. *Int. J. Green Energy* **2005**, *2* (2), 181–192.
- (21) Pellegrini, L. A.; Moioli, S.; Gamba, S. Energy saving in a CO₂ capture plant by MEA scrubbing. *Chem. Eng. Res. Des.* **2011**.
- (22) Amann, J. M. G.; Bouallou, C. CO₂ capture from power stations running with natural gas (NGCC) and pulverized coal (PC): Assessment of a new chemical solvent based on aqueous solutions of N-methyldiethanolamine plus triethylene tetramine. *Energy Procedia* **2009**, *1* (1), 909–916.
- (23) Romeo, L. M.; Bolea, I.; Escosa, J. M. Integration of power plant and amine scrubbing to reduce CO₂ capture costs. *Appl. Therm. Eng.* **2008**, *28* (8–9), 1039–1046.
- (24) Lawal, A.; Wang, M.; Stephenson, P.; Yeung, H. Dynamic modelling of CO₂ absorption for post combustion capture in coal-fired power plants. *Fuel* **2009**, *88* (12), 2455–2462.
- (25) Ong'iro, A. O.; Ugursal, V. I.; Al Taweel, A. M.; Blamire, D. K. Simulation of combined cycle power plants using the ASPEN PLUS shell. *Heat Recovery Syst. CHP* **1995**, *15* (2), 105–113.
- (26) Kunze, C.; Spliethoff, H. Modelling of an IGCC plant with carbon capture for 2020. *Fuel Process. Technol.* **2010**, *91* (8), 934–941.
- (27) Perez-Fortes, M.; Bojarski, A. D.; Velo, E.; Nougues, J. M.; Puigjaner, L. Conceptual model and evaluation of generated power and emissions in an IGCC plant. *Energy* **2009**, *34* (10), 1721–1732.
- (28) Zheng, L. G.; Furimsky, E. ASPEN simulation of cogeneration plants. *Energy Convers. Manage.* **2003**, *44* (11), 1845–1851.
- (29) Ong'iro, A.; Ugursal, V. I.; Al Taweel, A. M.; Lajeunesse, G. Thermodynamic simulation and evaluation of a steam CHP plant using ASPEN Plus. *Appl. Therm. Eng.* **1996**, *16* (3), 263–271.
- (30) Hong, J. S.; Chaudhry, G.; Brisson, J. G.; Field, R.; Gazzino, M.; Ghoniem, A. F. Analysis of oxy-fuel combustion power cycle utilizing a pressurized coal combustor. *Energy* **2009**, *34* (9), 1332–1340.
- (31) National Energy Technology Laboratory (NETL), United States Department of Energy (DOE). *Pulverized Coal Oxycombustion Power Plants*; NETL: Washington, D.C., 2007; Report DOE/NETL-2007/1291, Vol. 1: Bituminous coal to electricity.
- (32) Toftegaard, M. B.; Brix, J.; Jensen, P. A.; Glarborg, P.; Jensen, A. D. Oxy-fuel combustion of solid fuels. *Prog. Energy Combust. Sci.* **2010**, *36* (5), 581–625.
- (33) Wall, T.; Liu, Y. H.; Spero, C.; Elliott, L.; Khare, S.; Rathnam, R.; Zeenathal, F.; Moghtaderi, B.; Buhre, B.; Sheng, C. D.; Gupta, R.; Yamada, T.; Makino, K.; Yu, J. L. An overview on oxyfuel coal

combustion—State of the art research and technology development. *Chem. Eng. Res. Des.* **2009**, 87 (8A), 1003–1016.

(34) Anthony, E. J.; Berry, E. E.; Blondin, J.; Bulewicz, E. M.; Burwell, S. LIFAC ash—Strategies for management. *Waste Manage.* **2005**, 25 (3), 265–279.

(35) Luyben, W. L. *Distillation Design and Control Using Aspen Simulation*; John Wiley and Sons, Inc.: New York, 2006.

(36) Wall, T. F. Combustion processes for carbon capture. *Proc. Combust. Inst.* **2007**, 31, 31–47.