Article

Oxy-Fuel Combustion Characteristics of Pulverized Coal under O₂/Recirculated Flue Gas Atmospheres

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Abstract: Oxy-fuel combustion is an effective technology for carbon capture and storage (CCS). Oxy-combustion for coal-fired power stations is a promising technology by which to diminish CO₂ emissions. Unfortunately, little attention has been paid to the oxy-combustion characteristics affected by the combustion atmosphere. This paper is aimed at investigating the oxy-fuel combustion characteristics of Australian coal in a 0.3 MW_{th} furnace. In particular, the influences of various oxygen flow rates and recirculated flue gas (RFG) on heating performance and pollutant emissions are examined in O₂/RFG environments. The results show that with increases in the secondary RFG flow rate, the temperatures in the radiative and convective sections decrease and increase, respectively. At a lower oxygen flow rate, burning Australian coal emits lower residual oxygen and NO concentrations. In the flue gas, a high CO₂ concentration of up to 94.8% can be achieved. Compared to air combustion, NO emissions are dramatically reduced up to 74% for Australian coal under oxy-combustion. Note that the high CO₂ concentrations in the flue gas under oxy-coal combustions suggest great potential for reducing CO₂ emissions through carbon capture and storage.

Keywords: oxy-fuel combustion; CO₂ emissions; NO emissions; recirculated flue gas; CCS

1. Introduction

Electricity is a crucial part of most industries and livelihoods. Although there has been rapid development of alternative energy, the major power supply is still mainly produced from combusting fossil fuels like coal and natural gas. Taking this into consideration for coal-fired power plants, it is necessary to develop novel technologies to mitigate their impact on the environment from pollutant emissions and to improve efficiency. In 1982, Abraham et al. [1] proposed the concept of oxy-fuel combustion to improve enhanced oil recovery by producing CO₂-rich flue gas. Prior to combustion, an air separation unit is utilized to produce the oxygen required for oxy-fuel combustion. Using this method, fuel only combusts in an oxygen-diluted environment with recirculated flue gas (RFG), and CO₂ and H₂O are mainly produced in the flue gas (FG). Therefore, a high concentration CO₂ stream occurs, because CO₂ can be easily separated. It is worth noting that oxy-fuel combustion can reduce the amount of flue gas treated, can produce high levels of CO₂, and is feasible for retrofitting current power plants.

Oxy-coal combustion with recycled flue gas was first carried out by the International Flame Research Foundation (IFRF) at a pilot scale in the 1990s [2]. Subsequently, air separation technology has made considerable progress, promoting oxy-fuel combustion as it becomes a viable option for carbon

dioxide capture in the industry [3]. Oxy-fuel combustion provides a promising option for achieving higher CO₂ removal and lower pollutant emissions. Under oxy-fuel combustion, without N₂ being replaced by RFG, there will be a high concentration of CO₂ in the flue gas. The water vapor in the flue gas can be easily removed by condensation [4]. Using this method, without subsequent separation, the flue gas can be compressed and sequestered. Nevertheless, the combustion characteristics (such as flame stability [5], flammability limits [6], ignition delay [7,8], and pollutant emissions [9]) and heat transfer characteristics [10] of oxy-fuel combustion are quite different from those of air-firing, because there are significant differences in the physical and chemical properties of CO₂ and N₂, including heat capacity [11], density [7], emissivity [7], and the diffusion coefficient [12].

Recently, several studies were done to investigate oxy-fuel combustion characteristics, with a focus on pressurized oxy-combustion [13], sulfur emission from pulverized lignite fuel [14], and a combination of oxy-combustion with biomass [15]. Under oxy-combustion, considering the mixture of oxygen and recirculated flue gas (RFG), the higher emissivity and higher specific heat capacity of RFG will contribute to radiation heat transfer and lower flame temperature, respectively. Furthermore, compared to the O₂/N₂ atmosphere, ignition delay slightly increases in the O₂/CO₂ environment, due to the combined effect of the high heat capacity of CO₂ [16] and the properties of combustion gases [17]. The flame speed will slow down, and flame stability will also decrease under oxy-combustion [18]. Therefore, for better heating performance and improved flammability, it is crucial to optimize the ratio of O₂ and RFG, which is a significant parameter in oxy-fuel combustion.

On the other hand, pollutant emissions also play an important part in oxy-combustion. A significant reduction in the formation of nitrogen oxides (NO_x) can be achieved for oxy-fuel combustion, due to the absence of N₂ in an oxidant stream [19]. Woycenko et al. [20] reported that the NO_x emission rate (mg/MJ) in oxy-coal combustion was significantly reduced, at about 60% lower than that in air-firing mode. Normann et al. [21] reviewed some methods to control NO_x emissions during oxy-combustion, including reburning, staging, a low-NO_x burner, RFG, and high temperature NO_x reduction. Each method has its own pros and cons. However, RFG is more promising because it reduces NO_x by about 65% and is a necessary component of oxy-combustion [7]. Stadler et al. [22] carried out experiments on the NO_x emissions of oxy-coal combustion in a pilot plant. They found that compared to CO₂/O₂, for a swirl flame, O₂-enriched recirculated flue gas (O₂/RFG) can reduce NO_x emissions by up to 50%, while in flameless combustion, O₂/RFG can reduce NO_x emissions by approximately 40%. Subsequently, Ikeda et al. [23] also found that NO_x emissions in the O₂/RFG mode were greatly reduced, by about 50%, compared to air-firing mode. This is because the NO_x contained in the RFG is fed back to the flame through a secondary stream. Li et al. [24] investigated NO_x and SO₂ emissions during oxy-coal combustion at a high oxygen concentration (50%) in a 0.1 MW_{th} circulation, fluidized bed combustor. It was found that a low excess oxygen ratio contributes to a reduction in fuel N conversion, and in this situation, the CO₂ concentration in the flue gas was between 84.5% and 92.6%. De las Obras-Loscertales et al. [25] studied the effect of coal rank on NO and N₂O emissions in a bubbling fluidized bed combustor in the oxy-fuel combustion mode. They found that higher ranked coals lead to a larger fuel N conversion to NO and N₂O, and that an increase in moisture content results in a slight reduction in NO emissions. Hu et al. [26] investigated the NO emission behavior for three types of Chinese coal in both air and oxy-coal combustion modes. In both air and oxy-coal atmospheres, it was observed that NO emissions increase with decreases in the coal rank, and decrease with decreases in the O₂ fraction. Ndibe et al. [27] studied NO and NO₂ formation in an oxy-combustion pulverized fuel (PF) system. The study indicated that wet recirculated flue gas has better reduction efficiency on NO. Muto et al. [28] applied a large-eddy simulation to study the effect of O₂ concentration on NO_x formation in a pulverized coal-fired furnace.

SO₂ is another key component in pollutant emissions. Croiset and Thambimuthu's study [29] showed that the conversion of sulfur into SO₂ is significantly influenced by the type of combustion environment. The conversion rates were, respectively, 91%, 75%, and 64% for combustion in air, an O₂/CO₂ mixture, and an O₂/RFG mixture. During oxy-fuel combustion, the level of SO₂ emissions

is proportional to the fuel S , which is similar to combustion in air. It has been reported that the in-furnace SO_2 concentration significantly increases in an O_2/RFG atmosphere. Nevertheless, in terms of the total sulfur mass output, SO_2 emissions are lower during oxy-fuel combustion than during air-combustion [2]. Recently, Duan et al. [13] found that when the combustion pressure was increased, the SO_2 emissions were significantly reduced, due to the enhanced self-desulfurization of ash under high pressure.

This paper is aimed at investigating the oxy-combustion characteristics of Australian coal in a $0.3 \text{ MW}_{\text{th}}$ furnace. The influence of various oxygen flow rates and recirculated flue gas (RFG) on heating performance and pollutant emissions are examined in O_2/RFG environments.

2. Experimental

2.1. The $0.3 \text{ MW}_{\text{th}}$ Multi-Fuel Combustion Test Facility

Figure 1 schematically shows the major experimental apparatus, which is called the $0.3 \text{ MW}_{\text{th}}$ multi-fuel combustion test facility [30]. It contains a vertical furnace with a multi-fuel burner, a fuel and oxidant (air or oxygen/RFG) supply system, a post-combustion treatment unit (including a bag filter and heat exchanger (HEX)), and a measuring apparatus. More details are provided as follows.

Figure 1. Configuration of the $0.3 \text{ MW}_{\text{th}}$ multi-fuel combustion test facility. HEX: heat exchanger; IDF: induced draft fan.

2.1.1. Combustion Furnace with a Multi-Fuel Burner

Figure 2a depicts the combustion furnace, which is composed of three sections: the radiative, convective, and flue gas sections. The adjustable swirl burner is set at the top of the radiative section, and is available to burn both liquid and solid fuel, as shown in Figure 2b. The swirl strength varies with changes in the angle of the swirl generator. The radiative section is a down-fire combustion chamber (with a maximal heat release rate of $0.3 \text{ MW}_{\text{th}}$), which has a 56 cm inner diameter and is 305 cm in height. It is composed of five sections of steel-made shell, with three layers of firebricks inside. Each section has a window available to observe flame characteristics and stability. In addition, each section is equipped with an R-type thermocouple, ($T_{w1}-T_{w5}$), as illustrated in Figure 2a. The convective section is composed of six sections of steel-made shell, and is equipped with five K-type thermocouples ($T_{g1}-T_{g5}$). The upstream parts of the convective section near the radiative section are cooled with a water-cooling system because of high-temperature flue gas, and then the downstream parts are cooled by an air-cooling system. The third section is the flue gas section, which includes a gas sampling port, a pulse-jet cleaning bag house, a heat exchanger (shell-and-tube type), and a chimney. The operating pressure is maintained at +100 Pa in order to prevent air leakage [31].

Figure 2. (a) Locations of the thermocouple measurement and gas sampling in the combustion test furnace. (b) The multi-fuel burner.

2.1.2. Fuel and Oxidant/Recirculated Flue Gas (RFG) Supply System

The fuel supply system is comprised of three parts: gaseous fuel (liquefied petroleum gas, or LPG), liquid fuel (fuel oil or diesel), and solid fuel (pulverized coal or biomass), as illustrated in Figure 2b. The LPG, fed from a high-pressure cylinder, serves as a pilot flame for the combustion furnace. The liquid fuel (diesel) is stored in a daily tank, transported by a pump, and is used for preheating the furnace. The solid fuel, fed by a coal feeder, is transported by primary air to the burner. The feed rates of the gaseous and liquid fuels are controlled by adjusting the valve opening, while the supply rate of coal depends on the speed of the rotary feeder.

An air supply system and an oxygen supply system are combined into an oxidant/RFG supply system. The air supply system includes primary air, used to fluidize solid fuel, and secondary air, used as a source of oxidants. Both of their supply rates are controlled by a valve opening intended to meet the oxidant demand. In addition, both primary air and secondary air can be replaced with RFG, which herein is called either 1st RFG or 2nd RFG, respectively, in the oxy-fuel combustion case. Oxygen (purity 99.9%), fed by a commercial high pressure oxygen cylinder, is introduced and mixed with the 2nd RFG to form an O_2 /RFG mixture, as shown in Figures 1 and 2b.

Two kinds of flue gas are defined, wet flue gas and dry flue gas, depending on whether or not the flue gas flows through the shell and tube heat exchanger. Primary air and secondary air in air-firing

mode can be replaced with wet RFG or dry RFG in oxy-fuel combustion mode to meet the experimental needs, as shown in Figures 1 and 2b. In this study, only dry RFG is used. The dry RFG can be used to transport coal particles from coal feeders to furnaces and other uses, while the wet RFG can be used to control the combustion temperature, due to its much higher water vapor content than dry RFG. Although wet RFG is thermodynamically advantageous over dry RFG, the dry RFG will be needed if the water vapor or SO₂ content in the RFG is limited [21]. In addition, to focus on the effect of CO₂, the dry RFG is applied in the combustion experiments in this study. As a fuel carrier gas, the volume flow rate of the 1st RFG is maintained at 70 Nm³/h to ensure a sufficient gas velocity to transport pulverized coal from the coal feeder into the furnace. When used as a feed oxidizer gas stream, oxygen is mixed with the secondary RFG to form a CO₂-rich mixture of 2nd RFG and O₂, and the pulverized coal is then combusted with oxygen/RFG in the combustion chamber.

2.1.3. Flue Gas Treatment Unit

The post-combustion treatment (flue gas treatment) unit is utilized to remove suspended particles and to cool down the flue gas, for the purpose of reducing pollutant emissions and testing different operating conditions with various RFG formulations. The post-combustion treatment unit is composed of three types of equipment: a pulse-jet cleaning bag house, a heat exchanger, and a flue gas recirculation system. The pulse-jet cleaning bag house has a capacity of 400 Nm³/h and a dust load of 2000 mg/m³, with a pressure pulse of 150 mmAq and a maximum temperature resistance of 400 °C.

2.1.4. Measurement Devices

The control room of the 0.3 MW_{th} furnace is equipped with both a monitoring and control system, which are used to monitor the real-time status of the entire test furnace, e.g., primary and secondary air supply rate, coal feed rate, recirculated flue gas flow rate, stack pressure, oxygen flow rate, and temperature distributions (in the radiative section and convective section).

The sample ports for measuring temperatures using R- and K-type thermocouples are demonstrated in Figure 2a. The measured ranges are from 0 °C to 1450 °C for R-type thermocouples and from 0 °C to 1250 °C for K-type thermocouples, with accuracies of ±1.5 °C and ±2.2 °C, respectively. The measured temperature fluctuated, but about 95% of the recorded temperature fell within ±10 °C of the mean temperature. By assuming those errors are independent of each other, the overall errors are $\pm[(10)^2 + (1.5)^2]^{0.5} = 10.11$ °C and $\pm[(10)^2 + (2.2)^2]^{0.5} = 10.24$ °C, respectively, by the root-sum-square method [32].

Fuji Electric infrared gas analyzers were employed to analyze the composition of the emission flue gas. The model and measurement ranges of the infrared gas analyzers were O₂: ZRJ, 0%–100%, ±0.1%; CO₂: ZRJ, 0%–20%/0%–100%, ±0.1%; CO: ZRE, 0–500 ppm/0–5000 ppm, ±1 ppm; NO: ZRE, 0–1000 ppm/0–3000 ppm, ±1 ppm; and SO₂: ZRE, 0–500 ppm/0–5000 ppm, ±1 ppm. The sample position is shown in Figure 2a. Before the sample gas reaches the analyzers, the sample gas was filtered, cooled, and dried. Hence, emissions (volume concentrations) of the exhaust gas are expressed on a dry gas basis in the present study.

2.2. Experimental Procedures

The Australian coal tested in this research is sub-bituminous coal. The particle size of pulverized coal is an important factor influencing flame stability and combustion characteristics. A pulverizing mill (Raymond mill) was used here to pulverize the Australian coal. Then, the coal particles were sieved and selected to ensure a suitable size of 75 µm (200 mesh), based on the industry standard [33]. Finally, the pulverized fuels were introduced into the combustion chamber by primary air/RFG and combusted with secondary air (or a mixture of O₂ and RFG). Table 1 provides the proximate and ultimate analysis of Australian coal.

Table 1. Properties of Australian coal (AD: air-dried; DB: dry basis).

Ultimate Analysis (wt %)	
Nitrogen (% , DB)	1.65
Carbon (% , DB)	73.10
Hydrogen (% , DB)	4.30
Sulfur (% , DB)	0.48
Ash (% , DB)	15.20
Oxygen (% , DB)	5.27
Proximate Analysis (wt %)	
Ash (% , AD)	14.78
Moisture (% , AD)	2.78
Volatile matter (% , AD)	29.68
Fixed carbon (% , AD)	52.76
Heating value (MJ/kg, AD)	27.52

All of the experiments presented in this study adhere to the following three procedures [31,34]. The first step is furnace preheating. The goal of furnace preheating is to minimize the effect of furnace temperature fluctuations on combustion characteristics and to provide a steady, high-temperature atmosphere for test fuel combustion. The second procedure is combustion tuning. The aim of combustion tuning is to reduce either the air or flue gas flow rate as low as possible, in order to obtain the minimum excess oxygen for complete combustion. The third procedure is long-term, steady combustion. With appropriate air/oxygen or RFG flow rates, test fuel combustions require 4 h to investigate long-term combustion characteristics.

The coal feed rate depends on the experimental conditions, and a detailed coal feed rate for each experiment is provided in Table 2, which also shows the specific parameters of the coal in the oxy-combustion experiments. The parameters include the coal feed rate (heat release rate), the primary RFG flow rate, the secondary RFG flow rate, and the oxygen feed rate. In this study, the heat release rate was fixed at 165 kW_{th}, and the primary RFG was maintained at a constant value (70 Nm³/h), in order to investigate the effects of varied oxygen and secondary RFG feed rates on oxy-combustion characteristics.

Table 2. Parameters of the Australian coal used in the oxy-combustion experiments (RFG: recirculated flue gas).

Australian Coal		
Feed rate (kg/h)	21.54	
Heat release rate (kW)	165	
Primary RFG (1st RFG) (Nm ³ /h)	70	
Oxygen supply (Nm ³ /h)	41.5	38.5
Secondary RFG (2nd RFG) (Nm ³ /h)	70–220	50–170

2.2.1. Furnace Preheating Process

The purpose of furnace preheating is to provide a steady, high-temperature environment for the subsequent experiment, and to mitigate the influence of furnace temperature variations that occur during the experiment on the combustion characteristics. The preheating process can be divided into three phases. First, diesel with a calorific value of 36.14 MJ/L is combusted with air. Initially, the diesel feed rate is maintained at 12 L/h with a secondary air supply rate of 130 Nm³/h for 3 h. Then, the diesel flow rate increases to 16 L/h with a secondary air supply rate of 210 Nm³/h for the next 3 h. The process ends with the diesel flow rate rising incrementally to 20 L/h at a secondary air flow rate of 260 Nm³/h for long-term (about 16 h) preheating, to ensure that the walls of the furnace reach a

quasi-steady condition. That is, so the temperature distributions do not vary dramatically with time, as shown in Figure 3.

Figure 3. Temperature variations during the preheating process.

2.2.2. The Oxy-Fuel Combustion Process

Oxy-fuel combustion is different from air combustion. The oxidant is supplied from pure oxygen instead of air. As a result, the total feed rate of the O_2 /recirculated flue gas (RFG) mixture decreases significantly, and so does the flue gas heat loss. In this regard, the flame temperature increases significantly. Thus, it is necessary to utilize RFG to lower the temperature. Figure 4 illustrates the configuration of the oxygen and RFG supply system. Primary recirculated flue gas (1st RFG) and secondary recirculated flue gas (2nd RFG) flow through the primary and secondary air supply systems, respectively. That is, air is replaced with RFG. Thus, oxygen mixes with secondary RFG to form an O_2 /RFG mixture.

Figure 4. Configuration of oxygen supply and flue gas recirculation.

In oxy-fuel combustion with RFG, the flow rates of primary RFG (1st RFG) and oxygen are fixed, and the flow rate of secondary RFG (2nd RFG) is gradually adjusted. The residual oxygen in the flue gas involved in the combustion process is recirculated back to the furnace. Therefore, the oxygen/RFG blend ratio should take the residual oxygen $[O_2]$ into consideration. In this study, the effect of the oxygen/RFG blend ratio (Ψ_{O_2}) on the combustion characteristics of oxy-coal firing is examined. The oxygen/RFG blend ratio (i.e., the oxygen concentration (vol %) used in the RFG for each of the oxy-fuel tests) is defined as [30]

$$\Psi_{O_2} = \frac{\dot{Q}_{O_2} + (\dot{Q}_{1st \text{ RFG}} + \dot{Q}_{2nd \text{ RFG}}) \times [O_2]}{\dot{Q}_{O_2} + \dot{Q}_{1st \text{ RFG}} + \dot{Q}_{2nd \text{ RFG}}} \times 100\% \text{ (vol \%)} \quad (1)$$

where $[O_2]$ is the residual oxygen (vol %) in the flue gas, $\dot{Q}_{O_2}$ is the volume flow rate of pure oxygen, and $\dot{Q}_{1st\ RFG}$ and $\dot{Q}_{2nd\ RFG}$ are the volume flow rates of the primary and secondary RFG, respectively. Therefore, the oxygen concentration (vol %) utilized in the O_2 /RFG mixture for each of the oxy-fuel combustion experiments can be calculated.

It is significant to investigate the influence of flue gas recirculation on the characteristics of oxy-fuel combustion, in order to maintain the similar combustion temperature and heat transfer characteristics in the boiler as in the conventional coal-fired power plant. In this study, the feeding rate of the pulverized coal was controlled at 21.54 kg/h (with heat releasing rate of 165 kW_{th}). The volume flow rate of the primary RFG (1st RFG) was maintained at 70 Nm³/h to ensure that gas velocity was sufficiently large to carry pulverized coal from the coal feeder to the furnace. Meanwhile, the secondary RFG (2nd RFG) is gradually adjusted from low flow rate to high flow rate. Notably, in order to maintain the oxygen/RFG blend ratio Ψ_{O_2} (calculated using Equation (1)) in the range of around 18%–30%, as in [22], and to maintain the turndown ratio of the flow meter, the flow rate of the 2nd RFG must not be less than 50 Nm³/h. Therefore, for different recirculated flue gas ratios (oxygen/RFG blend ratios), it is easier to observe the heat transfer characteristics in the radiative zone in a short time. In addition, if the 2nd RFG is lower than 50 Nm³/h, there will not be enough high-concentration carbon dioxide (recirculated gas) to absorb the high heat generated during the oxy-coal combustion, resulting in burner damage for long-term operation. Therefore, the 2nd RFG was controlled in the range of 50–220 Nm³/h.

With an oxygen supply rate of $\dot{Q}_{O_2} = 41.5$ Nm³/h, five 2nd RFG flow rates ($\dot{Q}_{2nd\ RFG} = 70, 100, 140, 180, \text{ and } 220$ Nm³/h, corresponding to $\Psi_{O_2} = 29.89, 27.64, 24.07, 22.57, \text{ and } 21.51$ vol %, respectively) were utilized to assess the influence of oxygen/RFG blend ratio on oxy-coal combustion characteristics. With an oxygen supply rate of $\dot{Q}_{O_2} = 38.5$ Nm³/h, seven 2nd RFG flow rates ($\dot{Q}_{2nd\ RFG} = 50, 70, 90, 110, 130, 150, \text{ and } 170$ Nm³/h, corresponding to $\Psi_{O_2} = 26.81, 25.05, 22.64, 21.28, 20.31, 18.74, \text{ and } 17.31$ vol %, respectively) were used to assess the influence of the oxygen/RFG blend ratio on oxy-coal combustion characteristics. For each case, the temperatures (in the radiative and convective sections) and the compositions (in the flue gas) were recorded after at least 30 min of stable operation.

3. Results and Discussion

3.1. Temperature Distributions in Oxy-Combustion of Australian Coal

For Australian coal under oxy-fuel combustion, Figure 5 shows the temperature variations in the radiative and convective sections with an oxygen supply of 41.5 Nm³/h, while Figure 6 displays those with an oxygen supply of 38.5 Nm³/h. The results show similar trends in the temperature variations under these two operating conditions. That is, when the coal feed rate (21.54 kg/h) and primary RFG (1st RFG) flow rate (70 Nm³/h) are fixed at a constant oxygen supply (41.5 Nm³/h or 38.5 Nm³/h), with an increase in the flow rate of secondary RFG (2nd RFG), the temperature in the radiative section decreases, while the flue gas temperature in the convective section increases. This is because a larger flow rate of 2nd RFG (a higher flue-gas recirculation rate) leads to a lower flame temperature, and therefore a reduction in the radiative heat transfer [14,15]. On the other hand, carbon dioxide and water vapor have high thermal capacities compared to nitrogen, which will lead to an enhancement in the heat transfer in the convective section of the furnace. Thus, higher flue gas recirculation results in a higher gas temperature in the convective section. In addition, a significant temperature drop between T_{g1} and T_{g2} will be observed because of the cooling system between the radiative and convective sections.

Figure 5. Variations in temperature with the 2nd RFG flow rate for Australian coal under oxy-combustion at $\dot{Q}_{O_2} = 41.5 \text{ Nm}^3/\text{h}$, where $\dot{Q}_{O_2}$ is the volume flow rate of pure oxygen.

Figure 6. Variations of temperature with the 2nd RFG flow rate for Australian coal under oxy-combustion at $\dot{Q}_{O_2} = 38.5 \text{ Nm}^3/\text{h}$.

3.2. Emissions in Oxy-Combustion of Australian Coal

For Australian coal with an oxygen supply rate of $\dot{Q}_{O_2} = 41.5 \text{ Nm}^3/\text{h}$, Figure 7a shows variations in the O_2 concentrations and CO_2 emissions with the 2nd RFG flow rate, while Figure 7b shows variations in the CO , SO_2 , and NO emissions with the 2nd RFG flow rate. As shown in Figure 7a, for all operating conditions (with various 2nd RFG flow rates), the residual oxygen concentration in the flue gas ranged from 8% to 10%, while the CO_2 concentration in the flue gas ranged between 87% and 90%. On the other hand, as shown in Figure 7b, CO emissions remained nearly constant, with increases in the 2nd RFG flow rate. With increases in the 2nd RFG flow rate, the NO emission decreased, due to the decrease in the peak flame temperature, which inhibits thermal NO formation

through the absorption of combustion heat by the relatively cooler flue gas, as well as the reduction in the oxygen concentration in the combustion zone, which reduces the availability of oxygen for the formation of fuel NO. Meanwhile, the SO₂ emissions were higher at a higher 2nd RFG flow rate. In these experiments, the flue gas was recirculated without SO₂ removal. Accordingly, there were significant increases in the SO₂ concentrations in the flue gas, due to the accumulated effects of flue gas recirculation and the reduced volume of the flue gas [35].

Figure 7. (a) CO₂ and O₂ emissions; and (b) CO, NO, and SO₂ emissions for Australian coal during oxy-combustion at $\dot{Q}_{O_2} = 41.5\ \text{Nm}^3/\text{h}$.

For Australian coal at $\dot{Q}_{O_2} = 38.5\ \text{Nm}^3/\text{h}$, Figure 8a shows variations in the O₂ concentrations and CO₂ emissions with the 2nd RFG flow rate, while Figure 8b depicts variations in the CO, SO₂, and NO emissions with the 2nd RFG flow rate. As shown in Figure 8a, the residual oxygen concentration in the flue gas ranged between 3% and 5%. Meanwhile, the CO₂ concentration in flue gas remains at about 94% (average value), and reached 94.8% when the 2nd RFG flow rate was 170 Nm³/h. It is noteworthy that the high CO₂ concentration in the flue gas suggests great potential for reducing CO₂ emissions through carbon capture and storage, thus mitigating global climate change.

Figure 8. Variations in (a) CO_2 and O_2 emissions, and (b) CO , NO , and SO_2 emissions with 2nd RFG for Australian coal during oxy-combustion at $\dot{Q}_{O_2} = 38.5\ Nm^3/h$.

Figures 7a and 8a also demonstrate that a higher oxygen supply results in a higher residual O_2 and less CO_2 in the flue gas. The main reason for these results is that the flue gas contains O_2 , CO_2 , and other gaseous molecules, and CO_2 and O_2 are the two main components. Therefore, Figures 7a and 8a indicate that a lower residual O_2 concentration corresponds to a higher CO_2 concentration [36]. The oxygen concentration in the flue gas at $\dot{Q}_{O_2} = 38.5\ Nm^3/h$ is lower than that at $\dot{Q}_{O_2} = 41.5\ Nm^3/h$; therefore, the CO_2 concentration at $\dot{Q}_{O_2} = 38.5\ Nm^3/h$ is larger than that at $\dot{Q}_{O_2} = 41.5\ Nm^3/h$. In air-firing mode, 20% excess air is typically utilized. Oxy-coal combustion requires excess O_2 to reach a residual O_2 concentration in the flue gas similar to that of air combustion, which ranges from 3% to 5% [37]. It is worth noting that the CO_2 concentration in the flue gas remained at a high average value, about 94%, while the residual O_2 concentration in the flue gas was controlled in the range of 3%–5%. This result is in good agreement with the findings of Khare et al. [37]. In the present study, it was also found that in order to achieve the same performance in an existing furnace, the oxygen concentration (about 23%) in oxy-fuel combustion is higher than the oxygen concentration in air, as reported by Wang et al. [38].

Figure 8b shows a similar trend in NO emissions as that shown in Figure 7b. That is, with increases in the 2nd RFG flow rate, NO emissions decrease. For Australian coal during oxy-combustion, Figures 7b and 8b also show a similar trend, where in general, SO₂ emissions increase as the 2nd RFG flow rate rises. However, in the flue gas, the concentration of SO₂ at $\dot{Q}_{O_2} = 38.5 \text{ Nm}^3/\text{h}$ (Figure 8b) is much higher than that at $\dot{Q}_{O_2} = 41.5 \text{ Nm}^3/\text{h}$ (Figure 7b), perhaps because SO₂ converts into SO₃ with greater oxygen supplementation [39]. As shown in Figure 8b, during oxy-combustion with recirculated flue gas, the concentrations of SO₂ (in the range of 850–1180 ppm) are about three to four times larger than those (302 ppm) in air-combustion conditions [40], which agrees well with Tan et al.'s study [35]. In addition, increasing the secondary RFG flow rate corresponds to the enhancement of swirling in the swirl burner, leading to better mixing and more complete combustion; therefore, the concentration of CO in the flue gas is reduced. It was also found that at a fixed secondary RFG flow rate, a lower oxygen supply rate at $\dot{Q}_{O_2} = 38.5 \text{ Nm}^3/\text{h}$ (Figure 8b) results in a higher CO emission than with a higher oxygen supply rate, at $\dot{Q}_{O_2} = 41.5 \text{ Nm}^3/\text{h}$ (Figure 7b), due to the combined effects of increased oxygen concentration in the inlet oxidizer (RFG/oxygen mixture) and strengthened swirling.

The emission of NO decreased with increases in the 2nd RFG flow rate, due to a lower flame temperature. Operating conditions at an oxygen supply rate of $38.5 \text{ Nm}^3/\text{h}$ result in lower NO emissions compared to those with an oxygen supply rate of $41.5 \text{ Nm}^3/\text{h}$. That is, the NO concentration in the flue gas increased with increases in the O₂ concentration in the inlet O₂/RFG mixture. This phenomenon is in agreement with the findings in the literature [41–43]. Okazaki and Ando [41] reported that a decrease in the concentration of NO under oxy-combustion conditions was mainly due to the reburning of the NO in the recirculated flue gas. Hu et al. [42,43] found that an increase in the flow rate of recirculated flue gas led to a decrease in NO emissions. In other words, if the flow rate of the recirculated flue gas decreased, the concentration of oxygen in the burner zone increased, which contributed incrementally to NO emissions. Furthermore, the NO concentration under oxy-combustion conditions was 189.5 ppm (at 6% O₂), with a secondary RFG flow rate of $170 \text{ Nm}^3/\text{h}$, while the NO concentration under air combustion conditions was 739 ppm (at 6% O₂) [40]. This indicates a 74% NO reduction under oxy-firing conditions compared to the air-firing mode. This significant decrease in NO emissions under oxy-combustion conditions coincides with the findings of Nozaki et al. [44] and our previous work [31].

To summarize, the results for Australian coal under oxy-combustion conditions with varied RFG flow rates and different amounts of oxygen supply indicate the same temperature distribution trends in both the radiative and convective sections of the furnace. That is, the furnace wall temperatures in the radiative section decreased while the gas temperatures in the convective section increased with increases in 2nd RFG. In addition, compared to those at $\dot{Q}_{O_2} = 41.5 \text{ Nm}^3/\text{h}$, with a lower oxygen supply ($\dot{Q}_{O_2} = 38.5 \text{ Nm}^3/\text{h}$), the residual oxygen and NO emissions were lower, while SO₂, CO₂, and CO emissions were higher.

3.3. Modified Combustion Efficiency (MCE) in Oxy-Combustion of Australian Coal

Combustion efficiency (CE) is defined as the ratio of carbon released as CO₂ divided by the carbon released in the form of all carbon-containing products of combustion (such as CO₂, CO, non-methane hydrocarbons, and PM_{2.5}) [45,46]. It can be used to determine the completeness of the carbon oxidation that occurs during combustion. Since CE is difficult to measure, the modified combustion efficiency (MCE), which is defined as the ratio of carbon emitted as CO₂ divided by carbon emitted as CO₂ plus CO [47], is typically used as an estimate of CE [48,49]. The modified combustion efficiency (MCE; η) was calculated using the following equation [31,50–52]:

$$\eta = \frac{[\text{CO}_2]}{[\text{CO}_2] + [\text{CO}]} \times 100\%, \quad (2)$$

where $[\text{CO}_2]$ is volume concentration of CO_2 on a dry basis, and $[\text{CO}]$ is volume concentration of CO on a dry basis. Pure flaming combustion usually exhibits an MCE approaching 99% [49]. The MCEs have also been reported to be greater than 99.9%, such as when burning gaseous fuels (Wu et al. [50]), solid fuels (Arvanitoyannis et al. [51]; Bilsback et al. [52]), and liquid fuels (Huang et al. [31]). In this investigation, CO and CO_2 were measured. However, non-methane hydrocarbons and particulate matter content were not measured. Therefore, Equation (2) was used to calculate the modified combustion efficiency (MCE), as reported in our previous study [31] and other literature [50–52]. Based on Equation (2), modified combustion efficiencies above 99.9% were obtained in the present study, which agrees well with Liu and Shao's study [36]. This indicated that excellent combustion efficiency was achieved.

In view of the above discussion, we realized that unburned carbon (UC) in fly ash (FA) is also an indicator of combustion efficiency [53]. The high proportion of UC in FA reflects a large amount of energy loss corresponding to low combustion efficiency. In addition, UC is an obstacle to the beneficial use of FA, especially in the cement and concrete industries. In this study, ash analyses were not carried out. However, before starting this work, a loss on ignition (LOI) analysis of air-fuel combustion was performed in various positions, including the ash pot, convective section, and bag house under the following operating conditions: a coal feed rate of 21.54 kg/h and an air supply rate of 210 Nm^3/h . It is worth noting that the LOI level of each position was lower than 6% (by mass). In other words, fly ash can meet the CNS 3036 [54] and ASTM C618 [55] requirements. Liu and Shao [36] reported that oxy-coal combustion is expected to achieve higher combustion efficiency (close to 100%) than air-coal combustion (greater than 99%). The modified combustion efficiency (MCE) of oxy-coal combustion was higher than 99.9%, which supports the argument of Liu and Shao [36].

4. Conclusions

The oxy-combustion characteristics of Australian coal in a 0.3 MW_{th} furnace were investigated. The influence of various oxygen flow rates and recirculated flue gas (RFG) on heating performance and pollutant emissions are examined in O_2/RFG environments. The general conclusions drawn from results of this study are as follows.

With increases in the secondary RFG flow rate, the temperatures ($T_{w1}-T_{w5}$) in the radiative section decreased, while the temperatures ($T_{g1}-T_{g5}$) in the convective section increased. When firing Australian coal in the oxy-combustion mode at an oxygen supply of 38.5 Nm^3/h , with various RFG flow rates, the CO_2 concentration could be stably maintained at about 94%. High CO_2 concentrations in the flue gas of up to 94.8% can be achieved. It is noteworthy that the high CO_2 concentration in the flue gas suggests great potential for reducing CO_2 emissions through carbon capture and storage, thus mitigating global climate change. Furthermore, a higher O_2 supply rate led to higher O_2 and NO concentrations in the flue gas, while it lowered CO , SO_2 , and CO_2 concentrations in the flue gas when firing Australian coal. With increases in the secondary RFG flow rate, the NO emissions decreased while SO_2 emissions increased. Compared to air combustion, a significant reduction in NO emissions of as high as 74% could be achieved under oxy-combustion conditions.

For oxy-coal combustion, an appropriate amount of RFG is required, which not only reduces the flame temperature, but also enhances the mixing of fuel and oxidant in the furnace, resulting in more complete combustion. High CO_2 concentration in the flue gas of up to 94.8% can be achieved when the residual oxygen concentration in the flue gas is controlled in the range of 3–5 vol %. The energy requirement for CO_2 compression is reduced in all CO_2 sequestration options as the purity of the CO_2 increases. It is suggested that 95% should be a reasonable CO_2 purity [56]. Thus, in this study, the high CO_2 concentration in the flue gas around 95% suggests that it is possible and beneficial to achieve easy carbon capture and storage. While this is true, however, in view of the research results, one realizes that using oxy-fuel combustion requires more capex (oxygen plant, flue gas recirculation) and produces a lower power output (oxygen plant and gas compressor power demand) than a conventional coal plant. In addition, CCS requires storage sites for CO_2 , which are not available in many locations. As

such, there are still no commercial oxy-fuel plants in commercial operation, and one of the largest demo plants was the 30 MW_{th} plant at Callide in Australia. The oxy-fuel combustion process is still in the development stage, and a lot of research is still needed to fully clarify the consequences of its implementation in power plants. Furthermore, it would be better to handle economic aspects of operational cost with a comparison with conventional combustion.

In the present study, coal was directly burned under oxy-fuel combustion conditions to achieve high CO₂ concentrations, which is beneficial to carbon capture and storage (CCS). It is recognized that coal gasification is a process that reduces the CO₂ emission and emerges as a clean coal technology. Accordingly, coal can be gasified in a fuel-rich (hypoxic) combustion mode, and then the resulting synthesized gases can be properly burned under oxy-fuel combustion [57]. The coal gasification process is controlled by several operating parameters, including temperature, pressure, reaction time, coal rank, porosity, and choice of catalyst. The reaction temperature is one of the parameters that has the most influence on the gasification reaction. For example, the carbon conversion and gasification rate increased with increases in the reaction temperature. In addition, the mineral matter contained in coal has negative impacts on the environment and equipment, because it produces by-product slag and soot, causes equipment corrosion, and reduces the overall combustion rate. It should be noted that an understanding of the operating parameters influencing coal gasification and the oxy-fuel combustion characteristics of the resulting synthesized gas is important and worth further study.

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