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Emission characteristics of coal combustion in different O_2/N_2, O_2/CO_2 and O_2/RFG atmosphere

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Abstract

This study investigates the emission characteristics of CO_2, SO_2 and NOx in the flue gas of coal combustion by varying the compositions and concentrations of feed gas ($O_2/CO_2/N_2$) and the ratios of recycled flue gas. The differences between O_2/recycled flue gas (O_2/RFG) combustion and general air combustion are also discussed. Experimental results indicate that the maximum concentration of CO_2 in O_2/CO_2 combustion system is 95% as the feed gas is 30% O_2/70% CO_2. The average concentration of CO_2 in the flue gas of O_2/CO_2 coal combustion system is higher than 90% and much higher than that of O_2/N_2 coal combustion system. This high concentration of CO_2 is beneficial for the separation of CO_2 from the flue gas by adsorption or absorption technologies. The maximum concentration of CO_2 in O_2/N_2 combustion system is only 34% at the feed gas 50% O_2/50% N_2, the concentration of CO_2 is increased with the concentration of O_2 in feed gas.

By O_2/CO_2 combustion technology, higher concentration of SO_2 is produced as the feed gas is 30% O_2/70% CO_2 or 40% O_2/60% CO_2, while higher concentration of NOx is produced as the feed gas is 20% O_2/80% CO_2 or 50% O_2/50% CO_2. The mass flow rates of CO_2, SO_2 and NOx in the flue gas are all increased with the ratio of recycled flue gas except for the feed gas 20% O_2/80% CO_2. The enhanced mass flow rates of air pollutants in such O_2/RFG combustion system are also beneficial for improving the control efficiencies of air pollution control devices. By O_2/N_2 combustion technology, higher concentrations of SO_2 and NOx are produced as the feed gas is 21% O_2/79% N_2. The results also indicate that the formation of NOx in general air combustion system is higher than that in O_2/RFG or O_2/CO_2 combustion system.

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Keywords: Coal combustion; O_2/CO_2; O_2/RFG; SO_2; NOx

1. Introduction

As the global warming and climate change due to greenhouse effects are continuing serious, mitigating the emission of greenhouse gas CO_2 becomes an international and imperative issue. Since only 21% oxygen is contained in the auxiliary air, the concentration of CO_2 formed in general air combustion system is usually lower than 15%. The other 79% nitrogen in the auxiliary air is released as exhaust gas and consumes a lot of heat energy. The low concentration of CO_2 in the exhaust gas is the major reason that leads the removal efficiency of CO_2 by general end-of-pipe separation technologies ineffective. The major strategies for CO_2 control include physical adsorption, underground or seabed storage, chemical absorption, and organism fixation, etc. [1–3]; and the technologies for separating CO_2 from exhaust gas include physical adsorption, physical absorption, chemical absorption, cryogenic distillation, and membrane separation [4–8]. Practically, the removal efficiency of CO_2 by these technologies is usually depressed due to the deteriorations and poisons of sorbents in the presences of other air pollutants such as NOx, SOx, HCl, fly ash, and O_2. For improving the removal efficiency of CO_2, a lot of extra energy has been consumed [9]. Therefore, many researchers are devoted to develop the "Clean Coal Combustion Technology" or "Clean Combustion Technology". O_2/RFG or O_2/CO_2 combustion technology is one of the clean combustion technologies, and has been studied and applied at coal-fired power plants in Japan and Europe. Moreover, some novel coal combustion technologies such as MATIANT cycle and ZEPP (near zero emission power plants) derived from O_2/RFG or O_2/CO_2 are also developed [10–12].

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O_2/RFG combustion technology is originated in 1992. The technology separates the nitrogen from air and the remained pure oxygen is mixed with the recycled flue gas (the major gas is CO_2) to serve as the feed gas. The concentration of CO_2 in exhaust gas could be significantly increased and the recovery of CO_2 becomes feasible. As reported in the references, the concentration of CO_2 in the flue gas can be increased to higher than 95% by such O_2/RFG combustion technology. This high concentration of CO_2 is beneficial for the recovery and control [13]. The CO_2 recycled coal-combustion system and the CO_2 recycled IGCC (integrated gasification combined cycle) system which also apply this O_2/RFG combustion technology are demonstrated to be the most economic and effective combustion system for power generation [14]. Yamada et al. [15] addressed that the conversion of NOx in air combustion was 30% higher than that in O_2/CO_2 combustion. The emission of NOx and SO_2 could be reduced in O_2/CO_2 combustion. Croiset and Thambimuthu [16] also indicated that the emission of NOx in different combustion atmospheres followed the sequence of air > O_2/CO_2 without recycled flue gas > O_2/CO_2 with recycled flue gas, and the emission of SO_2 also followed the sequence of air > O_2/CO_2 without recycled flue gas > O_2/CO_2 with recycled flue gas.

Since the O_2/RFG combustion technology is still in development and many researches focus on its application in power plant and CO_2 recovery, the emission characteristics of other pollutants such as NOx and SO_2 are seldom investigated. The relative studies are never present in Taiwan. Therefore, this study completely investigates the emission characteristics of CO_2, NOx and SO_2 at different combustion atmospheres. The effects of different operating conditions are also discussed. These results will provide useful references for the applications of O_2/RFG combustion technology in the future.

2. Experimental

2.1. Experimental apparatus

Fig. 1 shows the schematic diagram of experimental apparatus. The feed gas with different gas composition and concentrations was prepared by using gas cylinders, air compressor, mass flow meters and gas mixer. For a better control of combustion temperature, a fluidized bed incinerator was used as the experimental reactor. The fluidized bed incinerator is consisted of a feedstock feeder, a main combustion chamber and a secondary combustion chamber of 50 cm height and 5 cm inside diameter. All the chambers are made of 3 mm thick stainless steel (AISI 310). Five thermocouples were inserted into the axis to determine the temperatures in first and secondary combustion chambers and flue gas pipe. Silica sand was used as bed material and its particle size distribution was 300 μm 49.62%, 214 μm 17.35%, 470 μm 15.05%, and <163 μm 12%. Because fluidized bed incinerator has better heat capacity and mixing efficiency, the temperature distribution in the combustion chamber is uniform and stable. The average deviation of incineration temperature in our experiments was controlled within 20 °C. The granular coal was fed into the incinerator by a rotary feeder on the top of fluidized bed incinerator. The exhaust gas was treated by a baghouse, a wet scrubber and finally discharged into the atmosphere. The granular coal used in this work was the bituminous coal produced from Indonesia and it was also used at one coal-fired power plant in Taiwan. The chemical compositions of the coal are 65.53% C, 4.78% H, 13.24% O, 2.6% N and 1.12% S. The coal was pretreated by drying, grinding, and screening. The heating value of dry coal is 6145 cal g^{-1} and the particle size of coal was controlled in the range of 833–1651 μm.

2.2. Experimental procedures

Before performing the experiment, the first and secondary combustion chambers were preheated by electric heaters to 800 and 1000 °C, respectively. The prepared feed gas with desired gas composition and concentration was introduced into the combustion system to verify the airtight of pipeline and the concentration of feed gas. After the temperature in combustion chambers reached steady state, the experiment started and the granular coal was continuously fed into the incinerator at a feed rate of 4 g min^{-1}. The flow rate of feed gas in each run was controlled at 20 L min^{-1}. The detailed operating conditions for each run are listed in Table 1.

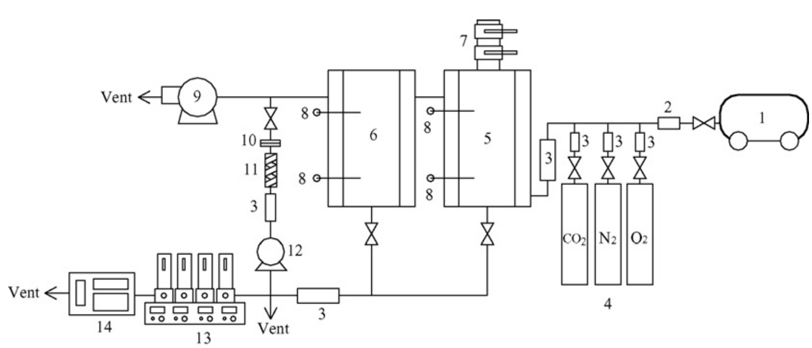

Fig. 1. Schematic diagram of experimental apparatus.

Table 1
Experimental conditions

Run	Feed gas composition (vol.%)			Recycled flue gas (vol.%)
	O_2	CO_2	N_2	
1–5	20	80	–	0, 30, 40, 50, 60 (1)[a]
6–14	30	70	–	0, 30, 40, 50, 60 (1, 2)[a]
15–23	40	60	–	0, 30, 40, 50, 60 (1, 2)[a]
24–28	50	50	–	0, 30, 40, 50, 60 (1)[a]
29–33	21	–	79	0, 30, 40, 50, 60 (1)[a]
34–42	30	–	70	0, 30, 40, 50, 60 (1, 2)[a]
42–51	40	–	60	0, 30, 40, 50, 60 (1, 2)[a]
52–56	50	–	50	0, 30, 40, 50, 60 (1)[a]

[a] 1: recycling flue gas to first combustion chamber; 2: recycling flue gas to second combustion chamber

The temperature profiles of combustion chambers and the concentrations of CO_2, NOx and SO_2 in the exhaust gas were monitored and recorded during the experiment according to the standard analysis procedures [17–19]. The coal feed rate and feed gas flow rate were checked and controlled for stability during the experiment. The flue gas was passed through a glass-fiber filter to remove fly ash, dewatered by a vapor condenser, and then analyzed by a flue gas analyzer (HORIBA, PG-250). This flue gas analyzer uses non-dispersive IR detection for CO and CO_2, and a galvanic cell or an optional zirconium oxide sensor for O_2 measurements. The standard detection ranges for CO_2, SO_2 and NOx are 0–20%, 0–3000 ppm, and 0–2500 ppm, respectively. In some cases, the flue gas was recycled and introduced into the gas inlet of first and second combustion chambers, respectively.

Because the concentrations of CO_2 and other pollutants emitted from O_2/CO_2 and O_2/RFG coal combustion system are much higher than the detection limits of flue gas analyzer, the dilution of flue gas is required before analysis. For minimizing the analysis errors resulted from gas dilution, highly precise gas dilution system was used and the dilution ratio was controlled as low as possible.

3. Results and discussions

3.1. Emission characteristics of O_2/RFG coal combustion with different feed gas compositions

Fig. 2 shows the variations of CO_2, SO_2 and NOx in the flue gas of coal combustion with different feed gas compositions (without recycling flue gas). At the same O_2 concentration in the feed gas, the concentration of CO_2 emitted from O_2/CO_2 coal combustion is obviously higher than that from O_2/N_2 combustion. Especially for the feed gas composition of 21% O_2/79% N_2, which is same with general air, only 13% CO_2 is produced in the coal combustion, while near 98% CO_2 is produced in the O_2/CO_2 coal combustion at the same feed gas concentration. Moreover, the concentrations of CO_2 produced from O_2/N_2 coal combustion are increased with the concentration of O_2 in the feed gas. The highest CO_2 concentration produced from O_2/N_2 coal combustion is 34% at the feed gas composition 50% O_2/50% N_2. The concentration of SO_2 generated from O_2/N_2 combustion is higher than O_2/CO_2 combustion, and is increased with the O_2

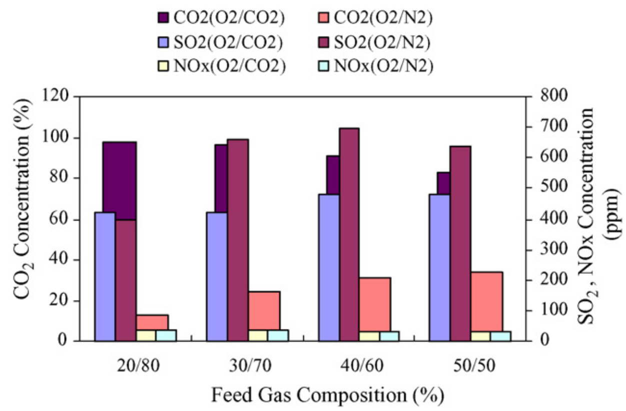

Fig. 2. Concentrations of CO_2, SO_2 and NOx at different feed gas compositions (without recycled flue gas).

concentration in feed gas. The formation of NOx do not have significant variation in O_2/CO_2 or O_2/N_2 combustion. From the viewpoint of CO_2 separation and recovery in a combustion process, O_2/CO_2 is better than O_2/N_2 because high-concentration CO_2 is emitted as the carry gas is CO_2 rather than N_2. Moreover, the specific heat of CO_2 is higher than N_2 and thermal stability of coal combustion in O_2/CO_2 is higher than that in O_2/N_2.

3.2. Emission characteristics of O_2/RFG coal combustion with different feed gas concentrations

Figs. 3 and 4 show the concentrations of CO_2, SO_2 and NOx in the flue gas of coal combustion at different feed gas concentrations of O_2/CO_2 as the ratio of recycled flue gas are 40% and 50%, respectively. The maximum concentration of CO_2 is higher than 95% at 30% O_2/70% CO_2 feed gas and 40% or 50% recycled flue gas, the next is occurred at 40% O_2/60% CO_2. The formation of SO_2 is similar to CO_2. The higher concentration is occurred at 30% O_2/70% CO_2 or 40% O_2/60% CO_2. However, the formation of NOx is quite different from that of CO_2 and SO_2, and more NOx are formed at the feed gas concentrations of 20% O_2/80% CO_2 and 50% O_2/50% CO_2.

Figs. 5 and 6 show the concentrations of CO_2, SO_2 and NOx in the flue gas of coal combustion at different feed gas concentrations of O_2/N_2 as the ratio of recycled flue gas are 40% and 50%,

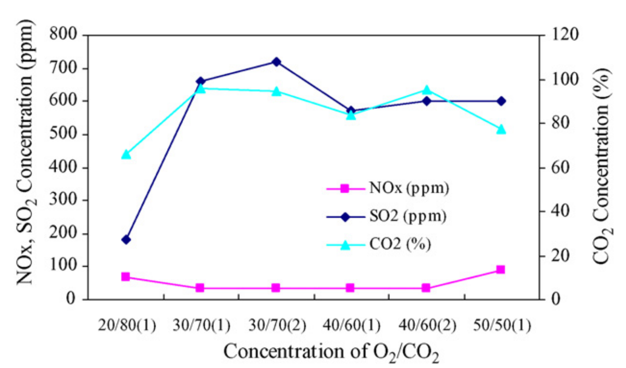

Fig. 3. Concentrations of CO_2, SO_2 and NOx at different feed gas concentrations of O_2/CO_2 with 40% recycled flue gas.

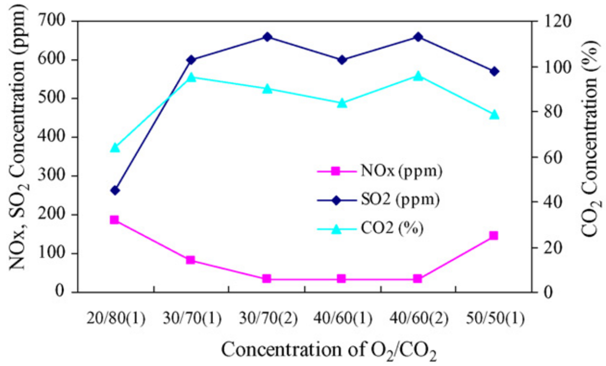

Fig. 4. Concentrations of CO_2, SO_2 and NOx at different feed gas concentrations of O_2/CO_2 with 50% recycled flue gas.

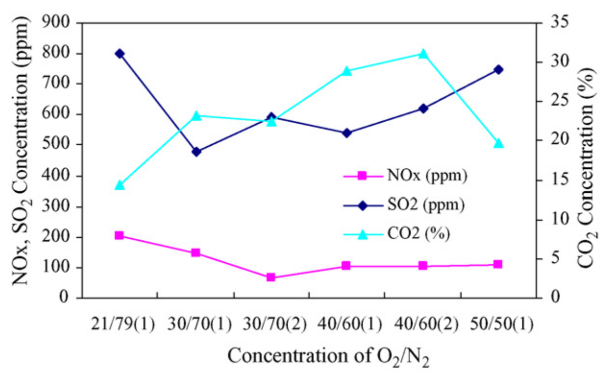

Fig. 5. Concentrations of CO_2, SO_2 and NOx at different feed gas concentrations of O_2/N_2 with 40% recycled flue gas.

respectively. The maximum concentration of CO_2 is occurred at 40% O_2/60% N_2 with 40% or 50% recycled flue gas. The formations of SO_2 and NOx are both higher at 21% O_2/79% N_2 or 50% O_2/50% N_2. These results also demonstrate that the formation of NOx is significant in general air and oxygen-rich combustions. With 30% and 60% recycled flue gas, the maximum concentration of CO_2 is occurred at 50% O_2/50% N_2. Comparing the results of coal combustion with O_2/CO_2 and O_2/N_2 feed gas atmosphere (Figs. 3–6), the concentration of O_2 in the feed gas has different effects on the combustion characteristics and the formations of CO_2, SO_2 and NOx.

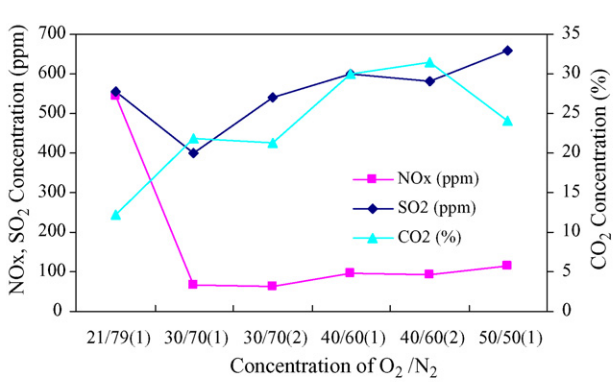

Fig. 6. Concentrations of CO_2, SO_2 and NOx at different feed gas concentrations of O_2/N_2 with 50% recycled flue gas.

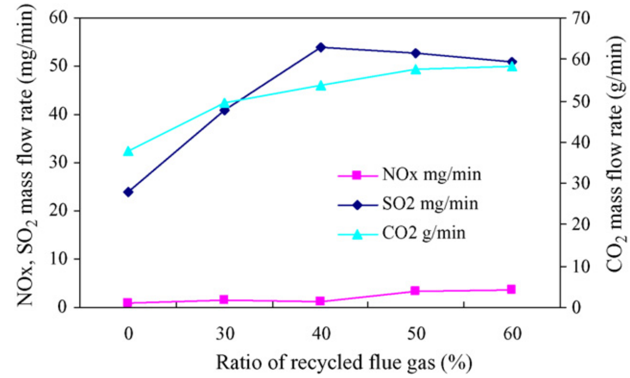

Fig. 7. Mass flow rate of CO_2, SO_2 and NOx at 30% O_2/70% CO_2 feed gas with different recycled flue gas ratios.

3.3. Emission characteristics of O_2/RFG coal combustion with different injection sites of recycled flue gas

For studying the effects of different injection sites of recycled flue gas on the performance of coal combustion, the recycled flue gas is injected into first combustion chamber and second combustion chamber, respectively (as shown in Fig. 1). The number in the parentheses of graduations on the horizontal axis of Figs. 3–6 indicates the injection site of recycled flue gas. From the results of Figs. 3–6, the concentration of CO_2 produced in the case of injecting recycled flue gas into the second combustion chamber is lower than that into first combustion chamber at the same ratio of flue gas recycled and the same feed gas composition of 30%/70% O_2/N_2 or O_2/CO_2. However, the concentration of CO_2 produced from injecting recycled flue gas into the second combustion chamber becomes higher than that into first combustion chamber as the feed gas concentration gas is 40%/60%. This phenomenon is attributed to the different reaction intensities, combustion temperatures and retention time in these two combustion chambers as the flue gas is recycled into the first and second combustion chambers.

3.4. Emission characteristics of O_2/RFG coal combustion with different ratios of recycled flue gas

With different ratios of recycled flue gas, the mass flow rates of CO_2, SO_2 and NOx in the flue gas are shown in Figs. 7–10. The mass flow rates of CO_2 are increased with the ratios of recycled flue gas except for the feed gas composition 20% O_2/80% CO_2. Recycling flue gas, which also recycles the pollutants, may enhance the concentrations of pollutants in the exhausted gas. This increased concentration is beneficial for the control, separation and recovery of CO_2 and other pollutants. The mass flow rates of SO_2 and NOx in the flue gas also have similar trends, only some exceptions are observed at the feed gas composition O_2/N_2. Above results indicate that coal combustion with recycled flue gas not only recovers the heat in the flue gas but also increases the mass flow rate of air pollutants because the volume of N_2 in feed gas is reduced and the combustion efficiency is improved due to the increase of average retention time of

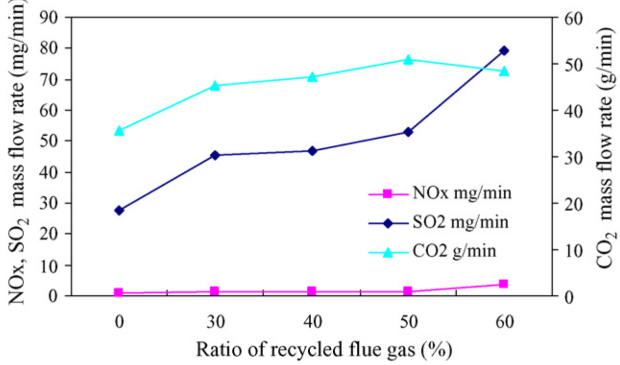

Fig. 8. Mass flow rate of CO_2, SO_2 and NOx at 40% O_2/60% CO_2 feed gas with different recycled flue gas ratios.

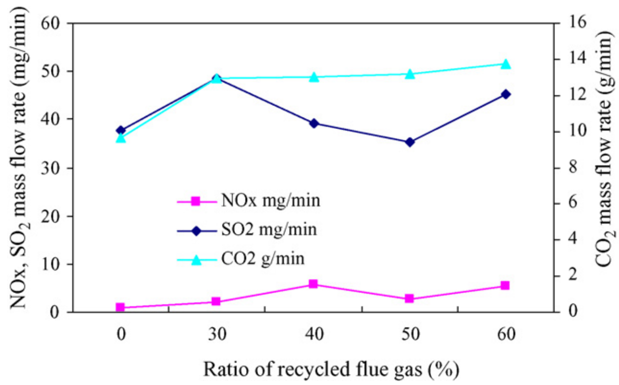

Fig. 9. Mass flow rate of CO_2, SO_2 and NOx at 30% O_2/70% N_2 feed gas with different recycled flue gas ratios.

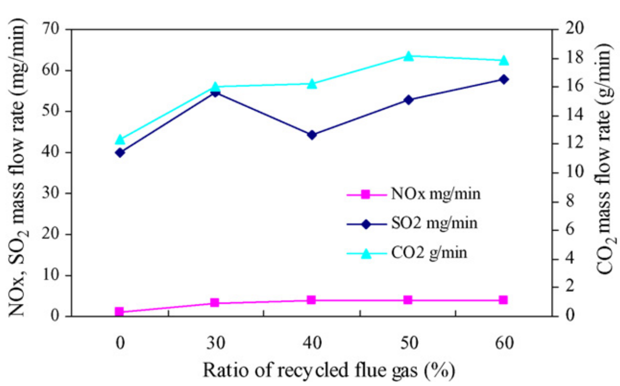

Fig. 10. Mass flow rate of CO_2, SO_2 and NOx at 40% O_2/60% N_2 feed gas with different recycled flue gas ratios.

reactants in combustion system. With suitable gas composition O_2/CO_2 and sufficient O_2 concentration in the feed gas, higher combustion efficiency and effective control of air pollutants during coal combustion can be achieved.

4. Conclusions

This study investigates the emission characteristics of CO_2, SO_2, NOx in the flue gas of O_2/RFG coal combustion with different feed gas compositions ($O_2/CO_2/N_2$) and ratios of recycled flue gas. The concentration of CO_2 generated from O_2/CO_2 combustion is much higher than that from O_2/N_2 combustion; the average concentration of CO_2 is higher than 90% and is beneficial to separate CO_2 from the flue gas by absorption or adsorption. The concentration of CO_2 generated from O_2/N_2 combustion is increased with the O_2 concentration in feed gas and the maximum concentration of CO_2 is near 34%.

In O_2/CO_2 (O_2/RFG) combustion, the maximum concentrations of CO_2 and SO_2 are both generated at the feed gas 30% O_2/70% CO_2, and the next is at 40% O_2/60% CO_2. The higher concentration of NOx is generated at 20% O_2/80% CO_2 or 50% O_2/50% CO_2. Except for the feed gas 20% O_2/80% CO_2, the mass flow rates of CO_2, SO_2, and NOx in the flue gas are all increased with the ratio of recycled flue gas. This result indicates that O_2/RFG combustion technology is really effective in enhancing the mass flow rates of CO_2 and other pollutants. The treatment efficiency of air pollution control devices or the separation and recovery efficiency of CO_2 can be therefore improved. In O_2/N_2 combustion, the maximum concentration of CO_2 is generated at 50% O_2/50% N_2, while the maximum concentration of SO_2 and NOx is generated at 21% O_2/79% N_2 or 50% O_2/50% N_2. The formation of NOx in conventional air combustion or oxygen-rich combustion is significant. Increasing the ratios of recycled flue gas, the mass flow rate of CO_2 in the flue gas is slightly increased and that of SO_2 and NOx are unapparent.

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