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The investigation of NO_x formation and reduction during O_2/CO_2 combustion of raw coal and coal char

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Abstract

This study investigated the behavior of NO_x emissions during combustion of pulverized coal char and pulverized raw coal in O_2/CO_2 environments under nearly isothermal condition in a drop tube furnace. A representative Chinese coal Ling Nancang (LNC) and LNC CO_2-char (made in CO_2 condition) were studied with four different excess oxygen ratios (α=0.6, 0.8, 1.2, and 1.4). Combustion experiments were performed with an elevated concentration (850 ppm) of NO. When α increased from 0.6 to 1.4, the total amount of NOx reduction was increased in both pulverized coal and char combustion. To investigate the effects of coal and char on NO_x reburn reactions, LNC coal char and the raw coal combustion experiments were also performed under three high background NO concentrations (400, 850, and 1200 ppm) in O_2/CO_2 atmosphere. During experiments with elevated NO concentrations, there was an increase in net NO reduction; however, there was also a decrease in the calculated NO reduction ratio.

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Keywords: coal combustion; O_2/CO_2; NO_x reduction

Introduction

Coal burning power plants are the largest source of CO_2 emissions in the world and their contribution to global warming has been widely recognized. However, in the long run, coal combustion power generation still represents the main source of power, particularly in countries such as China, which uses coal to generate electricity. O_2/CO_2 coal combustion is one of the most promising technologies that mitigate the rise of CO_2 in the atmosphere [1-6]. In this method, oxygen is separated from air and then burned with hydrocarbons to produce exhaust with a high concentration of CO_2 for storage. From a cost point of view, O_2/CO_2 combustion has an advantage as the air separation hardware can be added to the existing technology [3].

O_2/CO_2 combustion is promising not only because it is an easy way to recover CO_2, but also because of its low NO_x emissions [3, 5]. A fundamental study was conducted in single-pass combustors to examine coal combustion at high CO_2 concentrations with NO_x injection at the furnace entrance [7]. As discussed by Sun

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et al. [8], the reduction of recycled NO by gas–phase reaction was enhanced by a simulated volatile-N in fuel rich conditions. Hao et al. [9] concluded that O_2/CO_2 coal combustion with heat recirculation could further reduce NO emissions through a decrease in the volume of exhausted flue gas and through the low oxygen-fuel stoichiometric ratio. Ping et al. [10] studied the NO heterogeneous reduction with coal and reported that, for their experimental conditions, the char-N contribution to the total NO_x reduction reached more than 40%. A detailed understanding of the interaction effects of the different formation and reduction routes of NO_x during O_2/CO_2 combustion is lacking [7]. Although there have been a number of studies on NO_x formation and destruction in pulverized coal oxyfuel combustion, few studies have investigated the effects of CO_2-char to NOx formation and reduction. The present study experimentally investigated the effects of initial NO_x concentration and excess oxygen ratio on NO_x reduction during combustion of CO_2-char and coal under O_2/CO_2 atmosphere.

2. Experimental test facility and methods

Experiments were conducted in a drop tube furnace (DTF) reactor system. The reactor is a 2.2 m height corundum tube with an internal diameter of 55 mm. This was electrically heated. The effective heating length of the reactor was 1.8 m and fuel particles were entrained by mixture gases into the furnace by a top mounted scraper coal feeder (MFEV-10, Sankyo Piotech, Japan,). The residence time of coal particles in the reactor was estimated to be approximately 1 s in all cases as, in present study, the total gas flow through the DTF was maintained at 5.0 L/min. Gas flow rates were measured using calibrated mass flow controllers and a gas mixture was formed by mixing the two gases before being fed into the furnace. The fuel feeder rate changed with α, the operation temperature was maintained at 1200℃, and the oxygen concentration was kept at 20 vol. %.

The coal used in all tests was a candidate China coal (LNC). The coal char were prepared in the DTF at 1200 ℃ under a CO2 atmosphere. The particle sizes of the coal samples were in the range of 75-106 μm and the samples were dried under a vacuum for approximately 24 hours at room temperature prior to the experiment. The analysis of the coal and chars is given in Table 1. The NOx concentration in the flue gases was measured by an online multifunctional gas analyzer (Ecom-J2KN, RBR, Germany), with an accuracy of ± 5 ppm. Gases were cooled and dried before they entered into the analyzer.

Table 1. Coal and char analysis

	Proximate analysis (ad, wt. %)				Ultimate analysis (ad, wt. %)				
	Volatile matter	Moisture	Ash	Fixed carbon	Carbon	Hydrogen	Oxygen	Nitrogen	Sulfur
Coal	29.68	2.96	23.96	43.41	57.96	4.18	8.50	1.12	1.33
CO_2-Char	1.44	2.08	56.43	40.06	39.63	0.40	0.44	0.53	1.15

For temperatures less than 1200℃, NO_x is mainly derived from coal-N and the conversion ratio of NO_x can be defined as [7]:

$$N_{conversion\ \ ratio} = \frac{C(NO_X = 0,1) \times V \times P / RT}{M(fuel) \times Y(N, fuel) / M(N)} \times 100\% \quad (1)$$

where $C(NO_x{=}0,1)$ is the concentration of NOx in the flue gas when that in the carrier gas is zero, V is the volumetric flow rate of gases, P is the system pressure, R is the idea gas constant, T is the absolute temperature of the flue gas, M(fuel) is the mass flow rate of fuel, Y(N, fuel) is the mass fraction of nitrogen in the fuel, and M(N) represents the atomic weight of nitrogen.

As the initial concentration of NOx in the carrier gas is not equal to zero, it will be reduced by coal or char, and the reduction ratio of NO_x was defined as:

$$(NOx)_{reduction\ ratio} = \frac{C(NOx \neq 0,0) + C(NOx = 0,1) - C(NOx \neq 0,1)}{C(NOx \neq 0,0)} \times 100\% \quad (2)$$

where C(NO_x≠0, 0) denotes the initial NO_x concentration of the carrier gas, which is not equal to zero and C(NO_x ≠ 0, 1) denotes the NO_x concentration in the exhaust gas when the initial concentration is not equal to zero.

3. Results and discussion

3.1 The effect of excess oxygen ratio (α) on the formation and reduction of NO_x

To approach the reduction of NO_x for coal and char combustion, experiments were conducted under O_2/CO_2 atmosphere, with α ranging from 0.8 to 1.4, and an initial NO_x concentration of 0 and 850 ppm.

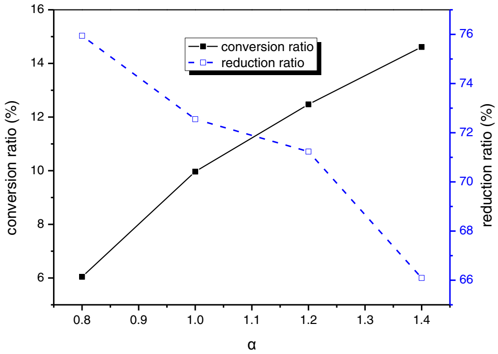

Figure 1. Coal-N transfer ratio and NO_x reduction ratio versus at different α condition

Results for coal-N conversion ratios and NO_x reduction ratios are shown in Figure 1. With an increase in α, the coal-N conversion ratio was gradually increased, but the reduction ratio of NO_x showed the opposite trend. This may be due to the faster formation of NO_x in the oxygen-enriched environment compared to that in the oxygen-deficient environment, which is favorable to the generation of NO_x. The NO_x reduction ratio decreases with an increase in α, particularly in an oxygen-enriched atmosphere. This may result from a larger effect of the increase of oxygen concentration on the newly formed CHi radicals during combustion, which more easily react with oxygen than NO_x. In addition, heterogeneous reduction reactions of coal-N at the char surface weaken in an oxidizing atmosphere along the combustion of coal [11].

The results of the NO_x formation and reduction during the CO_2-char combustion under O_2/CO_2 atmosphere at different values of α, and an initial atmospheric concentration of NO_x of 0.850 ppm are shown in Figure 2. The conversion ratios of char-N at different values of α are similar to but higher than those for coal. The NO_x reduction ratio decreases rapidly as α increase to values less than 1.2, then they increase more gradually for values of α from 1.2 to 1.4. This may be due to the fact that at low values of α, the CO concentration will increase for incomplete combustion of coal char, which promotes the heterogeneous reduction reaction of CO to NO_x. Under the condition of a high value of α, CO will quickly combust with O_2, which is not favorable to the reduction of NO_x. In the case of the oxygen-depleted condition, char would burn slowly and there would be enough time for the coal char to mix with NO_x. By comparing the reduction ratio of pulverized coal and coal char to NO_x, it may be noted that it is beneficial to coal-N and char-N conversion with increasing values of α.

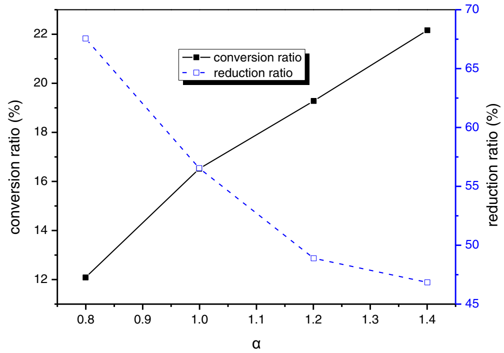

Figure 2. CO_2-char reduction ratio and conversion ratio at different α

3.2 The impact of initial NOx concentration on NO_x reduction

This section examines the reduction of NO_x for pulverized coal and char combustion under different initial NO_x concentrations at an α value of 1.4. The emissions of NO_x during coal and char combustion under different initial concentration of NO_x were tested. Results show that for char, the emission of NO_x in the exhaust gas is almost proportional to the initial NO_x concentration. However, for coal, from 400 to 850 ppm, the increase of NO_x is slower than that from 850 to 1200 ppm. This may be because the mass rate of coal (0.56 g /min) is less than that of char (0.94 g/min) at the same excess oxygen ratio (α).

Figure 3 shows the calculated total amount of NO_x reduction and the reduction ratio of NO_x during the coal and coal char combustion. With an increase in the initial NO_x concentration, the total amount of NO_x

reduction is increased, and the reduction ratio of NO_x is decreased. This shows that the increase in NO_x concentration can increase the total reduction, but the reduction ratio will be decreased. By increasing the concentration of NO_x, it is possible to increase the heterogeneous reduction of strength; however, the concentration of NO_x emissions will increase, and this would not be conducive to NO_x reduction. Under the present experimental conditions, the reduction ratio of NO_x for coal is higher than that for coal char.

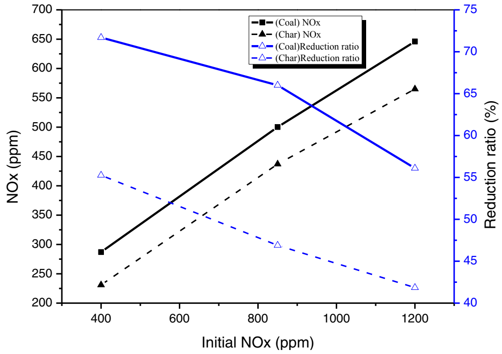

Figure 3. Coal and CO_2-char total NO_x reduction and the reduction ratio at different initial concentrations of NO_x

4. Conclusions

NO_x emission behavior experiments during coal and char in O_2/CO_2 atmosphere were carried out. The effects of varying excess oxygen ratios (α) and initial NO_x concentrations on the reduction ratios of NO_x were studied. Results show:

(1) As the excess oxygen ratio α increases, the reduction ratios of NO_x for both of coal and char decrease. Even for α=1.4, reduction ratios of 66% and 46% in coal and char combustion, respectively, can be attained.

(2) With the increase of initial concentration of NO_x, the reduction ratio of NO_x for coal is higher than that for coal CO_2-char during the combustion. With the increase of initial concentrations of NO_x, there was an increase in net NO reduction; however, there was also a decrease in the calculated NO reduction ratio.

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