Combustion experimental analysis into effects of interaction between blended coals on the NO emission

Yuanjing Chen1, Sheng Su1, Song Hu1, Jun Xu1, Kai Xu1, Yi Wang1*, Jun Xiang1
1 State Key Laboratory of Coal Combustion, School of Energy and Power Engineering, Huazhong University of Science and Technology, Wuhan 430074, China
E-mail address: alenwang@hust.edu.cn

Abstract. In studies of blended coal combustion, blending methods have an effect on NO emissions, but few experimental studies investigate the coal combustion in different height of furnace. In order to compare the effects of different blending methods on the NO emission during combustion process, bituminous coal, anthracite and their blended coal were burned in various modes in a two-layer and layer distance adjustable reactor. The results showed that NO emission of blended coal decreased nonlinearly with the blending ratio of bituminous coal (BBR) increasing. It indicated that interactions between component coals affected NO formation, leading to different NO emission in these three blending methods (two in-furnace blending methods and one out-furnace blending method). Compared with other methods, the in-furnace blending method (bituminous coal on the upper layer and anthracite on the lower layer) showed a great potential to reduce NO emission. Particularly, when the layer distance increased to 200 mm and the BBR is 70%, the NO emission reached the lowest.

1. Introduction
It is well known that NOx can cause serious environmental problems such as acid rain and photochemical fog. NOx emissions of coal-fired power plants take a great part of NOx in the environment[1, 2]. Conceivably, the comprehensive understanding of conversion mechanism of nitrogen species during coal combustion would help control NOx emission. Many investigators found that there is a relationship between NOx and coal quality[3-5]. However, coal is a complex substance with different properties related to ranks, maceral composition and impurities. Although many contributions have been taken, the effects of blended coal during combustion on NOx emissions, cannot be formulate a perfect approach predicted from those of individual coals and blending ratio due to non-additive factors[6-7].

Extensive studies have been performed to the combustion characteristics, emissions and ash behavior of blended coals[8-10]. However, only a few studies investigated the influence of the blending method on pollutant emission. Regarding blending methods, a power plant can use two different coal blending methods. In the out-furnace blending method, two types of coal are simultaneously injected into the furnace after being mixed together. In the in-furnace blending method, each coal is injected by a separate burner without prior blending. They are then mixed in the furnace. Baek et al. [9] carried out the computational and experimental works to investigate the influence of the blending method on NOx emissions. The simulation results showed the blending method has a little effect on NOx emission, but the field tests showed the in-furnace blending method substantially reduced in NOx, compared with the out-furnace blending method, also reported in other studies[10-12]. Unfortunately, the present study studies have not focused on the effects of the interaction between different coals on NO emission with...
different blending method although it was vital for controlling the NO emission. Furthermore, no more in-depth studies were available to fully understand the mechanism of NO emission in different blending method. In view of the significant influences of coal blending methods on NO emission, as well as its unclear interaction mechanism between different coals, it is necessary to conduct a further study and provide a fundamental insight into the NO emission during co-combustion with different coal blending methods.

As part of our ongoing efforts to understanding interaction mechanism between blended coals during combustion, this study aims to investigate the effects of interaction on NO emission. Coals are burned in separate quartz sieve plates of a modified multilayer reactor to simulate that coals are injected into the furnace by separate burners of a real power plant. The mixing ratio of two coal and the layer distance are also varied in this study.

2. Experimental

2.1. Coal samples
Two Chinese coal of different rank are used in this study: an anthracite and a high-volatile bituminous coal. They have different combustion characteristics but the similar nitrogen content. The coals are pulverized and sieved to obtain a particle size fraction of 70-100 μm. Before all the experiments, the coal samples are dried at 105°C for 24 h. The proximate and ultimate analyses of the coal are shown in Table 1.

| Samples    | Proximate analysis(%) | Ultimate analysis(%) |
|------------|-----------------------|----------------------|
|            | Volatile | Ash  | Fixed carbon | C  | H  | O  | N  | S  |
| Bitumite   | 29.08    | 7.99 | 62.93        | 73.55 | 4.79 | 12.12 | 0.77 | 0.78 |
| Anthracite | 5.89     | 28.48| 65.63        | 66.35 | 2.02 | 1.85  | 0.78 | 0.52 |

* By difference

2.2. Experimental process
The experiments of combustion are performed in an experimental modified multilayer reactor. The schematic presentation of the experimental apparatus is described in our previous study[13]. The reactor is made of two concentric quartz tubes with three quartz sieve plates. 1 L/min of mixed oxygen and nitrogen (79:21) is introduced to the reactor and then the reactor put in furnace at the moment when the furnace held the experimental temperature (1000°C) for 10 min. 200 ± 5 mg Coal samples of different blending method are introduced into the first and second quartz sieve plates prior to an experiment. The blending ratios of bituminous coal on the basis of total weight percentage (BBR) are 0%, 30%, 50%, 70%, 100%, where 0% refers to feed of anthracite only and 100% refers to feed of bituminous coal only. The produced gas is dried and filtered, then detected by the gas analyzer (MRU OPTIMA7, Germany) whose data is recorded by a computer.

Different coal samples are put on the first and second quartz sieve plates, simulating the actual burners in different positions and their blending time is controlled by varying the distance between the two sieve plates. The char is produced from the same mass of coal in the second quartz sieve. In case A, the anthracite is put on the first sieve plate (lower layer) while the bituminous coal is on the second sieve plate (upper layer). In case B, the position of anthracite and bituminous coal is opposite to case A. In case C, coals are introduced into the reactor with prior blending. Case A and B simulated the in-furnace blending method in actual burners in power plants, while case C simulated the out-furnace blending method.
2.3. Gas analysis

Under our experimental conditions, NO is mainly released by oxidation of fuel nitrogen, so the ratio of nitrogen converted to NO \((X_e)\) is calculated as the following equations,

\[
X_e = \frac{\int_0^t (\omega \times V \times P / RT)}{M(\text{fuel}) \times Y(N, \text{fuel}) / M(N)}
\]  

(R1)

where \(X_e\) is the experimental nitrogen conversion ratio, \(\omega\) is the volume fraction of NO measured by gas analyzer when the moment is \(t\), \(V\) is the volume flow rate of the gas, \(P\) is atmospheric pressure, \(R\) is the ideal gas constant, \(T\) is absolute temperature of the gas, \(M(\text{fuel})\) is the weight of sample in the experiment, \(Y(N, \text{fuel})\) is the nitrogen content of fuel, and \(M(N)\) is relative atomic mass of nitrogen.

To eliminate the linear effect of the mixing ratio variations, theoretical conversion ratio is determined as the sum of the NO conversion ratio of single coal supposing that there is no effect on nitrogen conversion during combustion of two coals. Theoretical conversion ratio of NO \((X_t)\) can be expressed as

\[
X_t = \frac{A \times \omega_1 + B \times \omega_2}{M \times Y_1(N, \text{fuel}) \times \omega_1 + M \times Y_2(N, \text{fuel}) \times \omega_2}
\]  

(R2)

where \(A\) and \(B\) are the nitrogen weight of the released NO during the separate combustion of bituminous coal and anthracite respectively, \(\omega_1\) and \(\omega_2\) are the proportion of bituminous coal and anthracite in the blending coal, \(Y_1(N, \text{fuel})\) and \(Y_2(N, \text{fuel})\) are the nitrogen mass fraction of bituminous coal and anthracite, and \(M\) is the weight of the fuel.

3. Results and discussion

3.1. Effects of blending methods on NO emission

![Figure 1. comparison of NO emission and nitrogen conversation ratio at different conditions](image)

NO emission of coal in different blending method is investigated on the modified multilayer reactor. This study mainly discusses the concentration of NO, due to the lower production of N\(_2\)O whose volume concentration is generally less than 20 ppm when the experimental temperature is 1000°C during pulverized coal combustion\(^5, 6\). Figure 1a shows the comparison of NO emission in different conditions during co-combustion of anthracite and bituminous coal. Compared to bituminous coal, the NO release of anthracite spends a longer time, which is related to its low combustion reactivity. The bituminous coal has high volatile content, which facilitates the release of intermediate nitrogen containing products, such as HCN and NH\(_3\), and then the conversation from fuel-N to NO proceeds more rapidly during subsequent oxidation. Besides, the NO emission peak value of anthracite is higher than that of bituminous coal, although the proportion of N in the two kinds of coal is approximated. Certainly, the \(X_e\) of anthracite is larger than that of bituminous coal as shown in Figure 1b. One reason is that the release of more volatile in combustion of bituminous coal can enlarge the porosity and superficial area of char, and likewise enhance the activity of char, which is beneficial for the reduction
of NO\(^7\), \(^8\). The other reason is that a relative large amounts of HCN and NH\(_3\) for low rank bituminous coal at low oxygen concentration would lead to less net formation of NO through greater NO reduction\(^\[12, 14\]\). Therefore, the total nitrogen conversion ratio of bituminous coal is much lower than that of anthracite\(^8\). It can be seen from Figure 1a that the NO emission varies with different blending method when the blending ratio of bituminous coal (BBR) is 50%. Apparently, the X\(_e\) is not equal to X\(_t\) (determined as half of the sum of the NO conversion ratio of single coal) as shown in Figure 1b. It implies that the interaction of component coals can’t be neglected. The nitrogen conversion ratio of case A is 11.51%, which is the lowest. Because the volatile-N converted to HCN and NH\(_3\) as the intermediate products through pyrolysis, which is partly oxidized to NO, and another part reduced the generated NO to N\(_2\)\(^\[15\]\). The anthracite coal in the lower layer is the first to contact with the oxygen to ignite and combust. It consumes a large amount of oxygen, and also the bituminous coal releases lots of volatile, so that the oxygen concentration in the combustion zone of bituminous coal in upper layer is lower and the reducing atmosphere is stronger. It results in that: firstly, volatile-N converts to HCN and NH\(_3\) as the intermediate products, and the generated NO reacts with intermediate products to generate N\(_2\). Secondly, CH\(_i\) free radicals generated in the combustion process reacts with generated NO to generate HCN, then the generated NO and HCN react with each other to create N\(_2\)\(^\[14, 16\]\). Meanwhile, the NO produced by anthracite coal on the lower layer will also take vision reduction reaction with the bituminous char to generate N\(_2\)\(^7\). As a result, the total NO emission of blending coal decreases.

In case B, the gas flows from the bottom of the reactor and the bituminous produces more NO at well-oxygenated condition\(^\[11, 14\]\), the anthracite char will transform N to NO and providing reaction sites for NO conversion of the pre-intermediate species (HCN and NH\(_3\)) formed from the volatile matter of bituminous coal\(^\[14\]\). This condition promotes the generation of NO. Therefore, the nitrogen conversion ratio of case B is 14.68%, which is the highest. In case C, the NO emission is between case A and B due to influence of this two respects. Besides, the X\(_e\) in case A is lower than X\(_t\) demonstrating the blending method of case A can improve the involvement of NO emission.

3.2. Effects of blending ratios on NO emission

Figure 2. comparison of NO emission and nitrogen conversation ratio at different BBRs in case A.

Figure 2a shows the comparison of NO emission as a function of blending ratio of bituminous coal during the combustion of coals in case A. The peak value declines with the BBR increasing as shown in Figure 2a and the X\(_e\) exhibits the same trend in Figure 2b. Two aspects are responsible for this phenomenon. On one side, the anthracite releases higher NO concentration than bituminous coal. The blending coal produces less NO with increasing proportion of bituminous coal. On another side, increase of the bituminous coal leads to improvement of volatile proportion, enhancing the reducing effect on NO. Comparing the experimental and theoretical conversation rate of NO, the X\(_e\) is not equal
to $X_t$. It elucidates the nitrogen conversion ratio is not linear correlated with blending ratio. At initial combustion reaction stage of blending coal, the devolatilization of component coal occurs separately. The nitrogen of high-volatile coal is released firstly and then the low-volatile coal releases fuel-nitrogen as the temperature rises continuously. Obviously the initial emission temperature and release rate of nitrogen are affected by properties of component coal and blending ratio synergistically\cite{16, 17}. When the proportion of bituminous coal is more than anthracite, it’s obvious that $X_e$ is less than $X_t$. It demonstrates that there is inhibition effect between the two coals and the volatiles of bituminous coal control the transformation of fuel-nitrogen to nitric oxide efficiently.

3.3. Effects of layer distance on NO emission

The layer distance also affects the NO emission, as shown in Figure 2a. The NO emission of blended coal exhibits a bimodal character accounting for the asynchronous decomposition of component coal. The first peak is mainly affected by bituminous coal while the second peak is mainly affected by anthracite coal. The two single coal doesn’t release volatile simultaneously, and the release rate of NO is different. The blending time of the gas decomposed by anthracite and the bituminous coal is shortened as the layer distance increases. It leads to that bituminous coal is in oxygen-lean condition for a relatively long time. The generation of NO in high volatile bituminous coal combustion process is restrained. Meanwhile, the anthracite coal combustion degree and oxygen consumption increase with the increase of layer distance, resulting in lower oxygen concentration of bituminous coal combustion area and stronger reducing atmosphere. Thus, the increase of the coal seam distance makes nitrogen conversion ratio decrease, as shown in Figure 2b. In general, when the bituminous coal proportion is relatively high, and the layer distance is large, NO emissions will be greatly reduced due to the homogeneous and heterogeneous reduction mechanism.

4. Conclusion

In summary, the NO emission of bituminous coal blended with anthracite in different blending methods has been systemically investigated by using a modified multilayer reactor. The following conclusions can be drawn.

1. The blending method of case A (the lower layer is anthracite and the upper layer is bituminous) is suggested to reduce NO emissions. The blending method of case A resulted in a further reduction in the NO emissions.

2. The NO emission nonlinearly decreases with the increase of bituminous coal proportion. The NO emission is apparently affected by interaction between the component coal that occurred in the combustion process of blended coal. The best condition is founded at a BBR of 70% because of the NO reduction effect.

3. In the in-furnace blending method of case A, the enlargement of layer distance can reduce the NO emission because bituminous volatiles in upper layer acts an important role in NO reduction. The reduction is more significant with the BBR increasing.

Acknowledgement

This research was financially supported by National Natural Science Foundation of China (NSFC) (No.51806074, No.51806079). The assistance from Analytical and Testing Center of Huazhong University of Science and Technology is also highly acknowledged. Also, thanks to the facility support of Equipment Center in State Key Laboratory of Coal Combustion, Huazhong University of Science and Technology.

References

[1] Smart J, Nakamura T. (1993) NOx emissions and burnout from a swirl-stabilised burner firing pulverised coal: the effects of firing coal blends. J. Energy Inst., 66(467):99-105.
[2] Ren, Q, Chi H, Gao J, Zhang C, Su S, Leong H, et al. (2020) Experimental study and
mechanism analysis of NO formation during volatile-N model compounds combustion in H2O/CO2 atmosphere. Fuel, 273: 117722.

[3] Sun Z, Su S, Xu J, Xu K, Hu S, Wang Y, et al. (2017) Effects of H2O on NO emission during oxy-coal combustion with wet recycle. Energy Fuels., 31(8).

[4] Liu G, Li Z, Chen Z, Zhu X, Zhu Q. (2012) Effect of the anthracite ratio of blended coals on the combustion and NOx emission characteristics of a retrofitted down-fired 660-MWe utility boiler. Appl Energy, 95(2): 196-201.

[5] Lupiáñez C, Guedea I, Bolea I, Diez LI, Romeo Luis M. (2013) Experimental study of SO2 and NOx emissions in fluidized bed oxy-fuel combustion[J]. Fuel Process. Technol., 106: 587-594.

[6] Saikaew T, Supudomnak P, Mekasut L, Piumsomboon P, Kuchonthara P. (2012) Emission of NOx and N2O from co-combustion of coal and biomasses in CFB combustor. Int. J. Greenh. Gas Control., 10: 26-32.

[7] Yao M, Che D, Liu Y, Liu Y. (2008) Effect of volatile-char interaction on the NO emission from coal combustion. Environ. Sci. Technol., 42(13): 4771-4776.

[8] Liu Y, Che D. (2006) Releases of NO and its precursors from coal combustion in a fixed bed. Fuel Process. Technol., 87(4): 355-362.

[9] Baek SH, Park HY, Ko S H. (2014) The effect of the coal blending method in a coal fired boiler on carbon in ash and NOx emission. Fuel, 128: 62-70.

[10] Ikeda M, Mokino H, Morinage, Higashiyama K, Kozai Y. (2008) Development of reduction technology of both emissions of NOx and unburned carbon by in-furnace blended method. Central Research Institute of Electric Power Industry.

[11] Lee B-H, Eddings EG, Jeon C-H. (2012) Effect of Coal Blending Methods with Different Excess Oxygen on Unburned Carbon and NOx Emissions in an Entrained Flow Reactor. Energy Fuels., 26(11): 6803-6814.

[12] Lee B-H, Kim S-G, Song J-H, Chang Y-J, Jeon C-H. (2011) Influence of Coal Blending Methods on Unburned Carbon and NOx Emissions in a Drop-Tube Furnace. Energy Fuels., 25(11): 5055-5062.

[13] Xu J, Liu J, Ling P, Zhang X, Xu K, He L, et al.(2020) Raman spectroscopy of biochar from the pyrolysis of three typical Chinese biomasses: A novel method for rapidly evaluating the biochar property. Energy, 202:117644.

[14] Li Yonghua. (2001) Study on high-efficiency and low-pollution combustion characteristics of coal blending. North China Electric Power University.

[15] Lans RPVD, Glarborg P, Dam-Johansen K. (1997) Influence of process parameters on nitrogen oxide formation in pulverized coal burners. Prog. Energy Combust. Sci., 23(4): 349-377.

[16] Hill SC, Smoot LD. (2000) Modeling of nitrogen oxides formation and destruction in combustion systems[J]. Prog. Energy Combust. Sci., 26(4): 417-458.

[17] Maier H, Splethoff H, Kicherer A, Fingerle K, Hein KRG. (1994) Effect of coal blending and particle size on NOx emission and burnout. Fuel, 73(9): 1447-1452.