



A role of hydrocarbon reaction for NO_x formation and reduction in fuel-rich pulverized coal combustion

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ABSTRACT

We have investigated an index for modeling a NO_x reaction mechanism of pulverized coal combustion. The reaction mechanism of coal nitrogen was examined by drop-tube furnace experiments under various burning conditions. We proposed the gas phase stoichiometric ratio (SR_{gas}) as a key index to evaluate NO_x concentration in fuel-rich flames. The SR_{gas} was defined as:

SR_{gas} ≡ amount of fuel required for stoichiometry combustion/amount of gasified fuel

where, the amount of gasified fuel was defined as the amount of fuel which had been released to the gas phase by pyrolysis, oxidation and gasification reactions. When SR_{gas} < 1.0, NO_x concentration was strongly influenced by the value of SR_{gas}. In this condition, the NO_x concentration was hardly influenced by coal type, particle diameter, or reaction time. We developed a model to analyze NO_x and XN(HCN, NH₃) concentrations for pulverized coal/air combustion and coal/CO₂/O₂ combustion, based on the index. NO_x and XN concentrations did not reproduce the experimental results without considering reactions between hydrocarbons and NO_x. The hydrocarbon reaction was important for both NO_x and XN, especially for air combustion. In the present model, an empirical formula was used to estimate the total concentration of hydrocarbons in coal flame. The reaction of heavy hydrocarbons which had plural aromatic rings was very important to analyze the reaction mechanism of hydrocarbons for coal combustion in detail. When burning temperature and SR_{gas} were the same, total hydrocarbon concentration in a coal flame was larger than that of a light gaseous hydrocarbon flame. Total hydrocarbon concentration in oxy-fuel combustion was lower than that in air combustion. We verified the proposed model by experimental results obtained for a drop-tube furnace and a laboratory-scale furnace that had an installed low-NO_x burner.

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1. Introduction

As computers become even more sophisticated, their use is increasing for the numerical analyses that are needed in designing pulverized coal firing boilers [3,5,14,19,21–23]. Numerical analyses were first applied to heat designs such as predictions of the steam temperature [19]. Recently, they have been applied to such environmental performance factors as NO_x emission [3,6,21–23] and to control furnace wall corrosion [14]. The reduction of the computation load is important for engineering design of the actual boilers. Our purpose in this study was to propose a reaction model that can predict NO_x emission concentrations reliably.

Several NO_x reduction technologies have been proposed, such as staged combustion [8], re-burning [4,8], and in-flame NO_x reduc-

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tion by using a low-NO_x burner [1,18]. A point which is common to all these technologies is that the NO_x reduction region was formed in the flame where the oxygen concentration was very low. To predict NO_x emission well, it is important to predict NO_x concentration reliably in the NO_x reduction region. However, for combustion of coal, NO_x performance changes easily with the burning conditions, such as coal properties and coal particle diameter. The NO_x reduction effect seems to depend more on the coal properties. However, boiler design and development cannot be done efficiently if it is necessary to change the method of NO_x reduction for each coal property. In the present study, we proposed a key index to estimate NO_x reduction performance. This index has hardly any influence from the burning conditions. We also proposed a reaction model which uses the proposed index.

Several studies have been made to investigate the reaction of volatile matter which focused on the first half of the coal combustion phenomenon [5,9,12] or the second half of the coal combustion, the char combustion region [2,13,17]. In [17], we proposed a reduced NO_x reaction model which focused on the gas phase reaction in the char combustion region. A key reaction in the model

Nomenclature

[CO]	mole fraction of carbon monoxide (-)	[OH _{eq}]	equilibrium OH mole fraction (-)
[CH ₄]	mole fraction of methane (-)	O _x (OH)	oxidative radicals, such as OH
[THC]	mole fraction of total hydrocarbon as CH ₄ (-)	SR _{in}	inlet stoichiometric ratio (-)
[H ₂]	mole fraction of hydrogen (-)	SR _{gas}	gas phase stoichiometric ratio (-)
L	distance between the burner nozzle and sampling port (mm)	[TR]	mole fraction of tracer (-)
[O ₂]	mole fraction of oxygen (-)	[TR] ₀	mole fraction of tracer in combustion supporting gas (-)
[O ₂] ₀	mole fraction of oxygen in combustion supporting gas (-)	X	axial location (mm)
		XN (HCN, NH ₃)	nitrogenous species

is reduction of NO_x by hydrocarbons. The model can predict NO_x concentration well in the fuel-rich condition if the hydrocarbon radicals are formed not only by volatile combustion, but also by char combustion.

In [17], our emphasis was on NO_x formation in fuel-rich flames, but, we did not verify the hydrocarbon concentration that remained in the flame. In the present study, we measured hydrocarbon concentration, and we studied a model for hydrocarbons. The hydrocarbon that remained in the flame influenced concentrations of XN (NH₃ and HCN). Based on experimental hydrocarbon concentration data, we studied a reaction model to explain XN (NH₃ and HCN) concentrations both for air combustion and CO₂/O₂ combustion. Finally, we examined whether NO_x prediction was reliable, if we used the developed reaction model for hydrocarbons and XN.

2. Experimental

We used the same drop-tube furnace as previously [17]. Fig. 1 shows its structure. Combustion supporting gases were air or a mixture of N₂, O₂, CO₂, and H₂O. Air, N₂, O₂, CO₂ were supplied from gas cylinders. Liquid H₂O was supplied from a tank, and then vaporized with an electric heater before mixing with other gases. Pulverized coal was pre-mixed with all of the combustion supporting gases, and then injected through a nozzle. We defined the stoichiometric ratio calculated by supplying all the coal and all the combustion supporting gas as an inlet stoichiometric ratio, SR_{in}. Temperature in the nozzle was kept around 473 K to prevent pyrolysis of coal particles before injection. The nozzle was covered

with firebrick and an SiC tube to prevent too much cooling of the injected gas. Flow rate of combustion supporting gas was 0.96 Nm³/h. Coal feed rate was varied for each experiment, from 0.02–0.5 kg/h. The reaction zone (maximum length, 1200 mm) was an alumina tube which had an inner diameter of 50 mm. Four sets of electric heaters were arranged around this tube. Temperature of each heater could be controlled independently in order to keep the temperature distribution of the tube wall constant. The axial temperature distribution of the heated gas was measured along the center axis of the tube. The heating rate was around 15,000 K/s. The burning temperature (measured as the wall temperature) was 1373–1773 K. The length (*L*) from the nozzle to the sampling probe was changed between 100 and 1000 mm. The reaction time was changed between approximately 0.1–1.4 s. The axial sampling position was varied for the experimental conditions. Injecting water into the probe quenched the reaction.

Char was collected by filtration through a 7 μm pore paper filter. C, H, N and ash contents in the char were analyzed. Coal burnout was obtained by assuming that the amount of ash remained constant during combustion. Concentrations of HCN and NH₃ were obtained from the concentrations of NH₄⁺ and CN⁻ in the water in a trap and total gas flow rate. NO_x, O₂, CO and CO₂ were measured with continuous analyzers. O₂, CO, CO₂, N₂, H₂ and hydrocarbons (CH₄, C₆H₆ and other hydrocarbon species) were measured by gas chromatography.

Analyses of six coal samples are listed in Table 1. Three types were tested: subbituminous, hv bituminous, and lv bituminous coals. An example of the particle diameter distribution of coal B

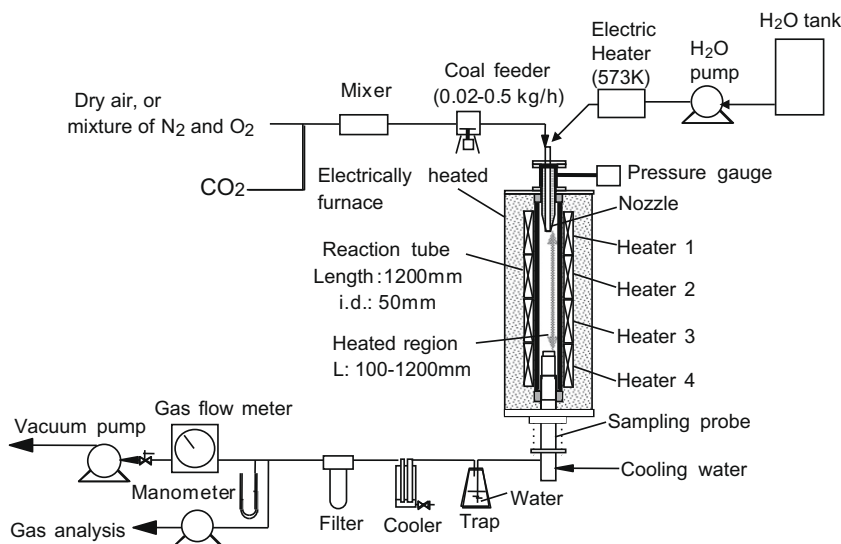


Fig. 1. Structure of the drop-tube furnace.

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