Empirical Formula to Predict the NOx Emissions from Coal Power Plant using Lab-Scale and Real-Scale Operating Data

Gyeong-Min Kim 1,†, Jong-Won Jeong 1,†, Jae-Seong Jeong 1, Dong-Yeop Kim 2, Seung-Mo Kim 3 and Chung-Hwan Jeon 1,3,*

1 School of Mechanical Engineering, Pusan National University, Busan 46241, Korea
2 Combustion Technology Center, Hadong Thermal Power Site Division, Korea Southern Power Co., Ltd., Busan 52353, Korea
3 Pusan Clean Coal Center, Pusan National University, Busan 46241, Korea
* Correspondence: chjeon@pusan.ac.kr; Tel.: +82-51-510-3051; Fax: +82-51-510-5236
† These authors contributed equally to this work. Jong-Won Jeong is the co-first author and has the same contribution to this paper.

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Abstract: The use of fossil fuels has drastically increased throughout the world as the demand for energy increases. Accordingly, it has become critical that we reduce the oxides of nitrogen (NOx) and oxides of sulfur pollutants. Therefore, studies related to these activities have increased. This study was aimed at helping take pre-emptive action on NOx emissions by developing a formula that would predict NOx generation using factors related to the combustion characteristics and basic material properties of coal. In this study, the experiments were conducted using a drop tube furnace, and the correlation between coal’s major characteristics and NOx generation was analyzed and measured. Our results showed that the major factors affecting NOx generation are moisture, fixed carbon, and fuel ratio. Moisture tended to decrease NOx generation by delaying the ignition of coal and fixed carbon exhibited a tendency to be directly proportional to NOx generation. The $R^2$ value for NOx of moisture and fixed carbon were derived as 0.7659 and 0.7063, respectively. Our results also showed that the fuel ratio had an exponential relation with the conversion of fuel-N to NOx. Based on the results of our analyses, we used moisture, fixed carbon, and fuel ratio as the major factors for creating an experimental formula. Through these results, we confirmed that the prediction formula reflects the actual amount of NOx emitted from the powerplants.

Keywords: NOx concentration; drop tube furnace (DTF); pulverized coal combustion; empirical formula

1. Introduction

Energy consumption has been increasing explosively throughout the world in both developed and developing countries. Energy production from fossil fuel makes up 80% of the world’s energy and all researchers are concerned with the environmental problems caused by the use of fossil fuels. Oxides of sulfur (SOx) and nitrogen (NOx) are emitted from powerplants that use the fine dust of pulverized coal since a power source must be reduced. Therefore, regulations on emissions must be more restrictive, not only those from powerplants but also from cars, ships, and other fossil-fuel vehicles [1–4].

In countries such as China and India, where there is a rapidly increasing industrial demand for power, the increase in the demand brings increases in the price of coal, which causes several countries, such as Korea, to import low-quality or other types of coal to use as fuel [5]. These pulverized-coal powerplants have several problems related to combustion, particularly exhaust emissions from the fuel combustion process. In general, the main environmental pollutants are greenhouse gases (GHGs), fine dust particles, and precursors of Sox and NOx. Since the fine dust–related social issues are
becoming more important, a reduction in SOx and NOx generation is one of the most important issues. Therefore, accurate predictions and measurements of combustion emission gases are very important for establishing better environmental effects from boiler operations and fuel use. The reaction mechanism in SOx production is well known and the number of SOx generated is predictable. Therefore, powerplants already use this information as a basis for estimating fossil fuel shift time using SOx [6]. Conversely, it is difficult to predict the amount of NOx generated, and most efforts to predict the amounts from the complicated mechanisms of action through which this pollutant is generated are formulas based on experiments and experience and cannot be precisely explained [7]. However, guidelines are needed for predictive values that are easy to apply to the emission of exhaust under established operational conditions and powerplant and research and development (R&D) infrastructure facilities continue to create indices to use as experimental factors in a prediction formula. Therefore, R&D on identifying the factors for predicting NOx generation based on experimental and empirical formulas is becoming more important in setting up operational conditions in the backend facilities to decrease NOx and operate boilers in thermal powerplants that use various types of fuel.

The prediction of the NOx emissions of coal has been attempted for a long time. Every combustion process of the boilers depends on their own individual combustion technique and operation conditions. Tremendous complicated influence factors affect NOx emissions. Several researchers introduced have experimentally established the NOx prediction formula and used the system indicators (coefficients) to express the operating conditions and boiler types. Researchers, including Shimizu, developed empirical formulas for NOx generation using the characteristics of coal. Based on nine types of coal, researchers conducted the experiments at reactor sites with an electric heating furnace and created an empirical formula for NOx conversion from the amount of carbon monoxide (CO) and CO\textsubscript{2} generated [8]. Other researchers, such as Rozendaal, studied the phenomenon of NOx generation based on different types of coal [9]. A linear regression analysis on NOx emissions and the characteristics of coal showed a dispersity between NOx emissions from the coal’s volatilized amount and the fuel ratio (FR) and created a coefficient of regression and coefficient of determination. However, the results also showed that, while applying the actual results to the data, they did not correspond to the experimental characteristics. Ehrhardt studied NOx reduction using a reburning process under pilot-scale conditions, and Fiveland, through numerical modeling, suggested a low-NOx burner that could be attached to a boiler [10,11].

As mentioned, studies on NOx generation from coal have been conducted. However, it is rare that a prediction corresponds well to all conditions. Although there is a statistical relation between fundamental analysis and heuristic algorithms, predicting the amount of NOx generated remains difficult because coal combustion is nonhomogeneous. Nevertheless, according to researchers Kokkinoes and Eddings, progress on determining the correlations between NOx emissions and coal’s FR is ongoing [12]. Furthermore, several researchers have continued to conduct studies on boilers and burners that continue to operate. A model to predict NOx generation has shown substantial credibility and a tendency to actually model the emissions [2,13–16].

In this study, we created an empirical formula by considering previous research and several factors related to the material properties of coal and their generation of NOx using a drop tube furnace (DTF) and lab-scale equipment. We compared this empirical formula with NOx generation from the operating powerplants. We assumed that the NOx empirical formula from this study will provide the necessary information for taking preemptive actions when setting up conditions of the operating facilities, including the backend facilities and boilers, using the material properties of coal and will also help with cost management. This formula, unlike previous research studies, signifies that the NOx derivation factor is only based on the basic properties of coal. Thus, it can be easily applied to many coals.

2. Materials and Methods

2.1. Coal and Biomass Samples

Twenty different coals were chosen ranging from subbituminous to bituminous coal. These were from coals that are used in actual powerplant operations at the Hadong Thermal Power Site Division
of Korea Southern Power Co., Ltd. The coal samples used in this study were pulverized after drying using a vibratory disc mill (RS 200, Retech GmbH, Haan, Germany) and were curdled using a sieve shaker (AS 200, Retech GmbH, Haan, Germany) to produce a particle size of 75 to 90 μm. Proximate analysis has been conducted on a 5-g sample using a thermogravimetric analyzer (TGA 701, LECO Co., St. Joseph, MI, USA). The final analysis using a commercial element analyzer was conducted (Leco-TruSpec Micro CHNS, LECO Co., St. Joseph, MI, USA).

2.2. Experimental Apparatus

A DTF was used to identify NOx exhaust concentrations during the combustion state of the chosen coals. A schematic of the DTF device is provided in Figure 1. The DTF is divided into three systems—fuel injecting, fuel reaction, and collecting. The fuel injection system is located in the upper part of the device. Solid fuels, such as coal and biomass, are injected into the reactor through a carrier gas. The fuel supply is created by continuously injecting a fixed amount of coal with a nitrogen carrier gas using a syringe pump. A highly pure alumina tube with strong thermal resistance, an inner diameter of 700 mm, and 1200 mm long was used as the fuel-reaction system. Kanthal Super, which can be heated to 1850 °C, was used as the heating element. A cyclone and gas analyzer were used as collectors. The cyclone was located at the lower part of the device and it collected the ash, including unburned carbon emitted after combustion. The gas emitted from the reactor was analyzed/recorded in real time using the gas analyzer (Green Line MK2, Eurotron Instruments, Chemsford, UK). To reduce the experimental margin of error, three tests were conducted for each combustion condition for each sample and NOx generated after combustion was marked according to the 6% oxygen standard. The upper and lower parts of DTF consisted of assemblies of piper and water jacket and were designed to prevent the reaction from occurring near the input of the coolant. A mass flow controller (EX-250S, Kofloc, Kyoto, Japan) was used to control the flows of the carrier gas (N2, 1 L/min) and reaction gas (N2, O2). To create the test environment near the second boiler in the Hadong Thermal Power Site, the temperature of the DTF main furnace, particularly the vertical alumina tube, was fixed at 1400 °C to conform to the combustion environment of the actual coal boiler. In addition, samples with a particle size of 75 to 90 μm were provided with a fixed heat of 5,600 kcal/kg. Therefore, the input heat was calculated to be 1.68 kcal/min considering that the standard fuel supply was 0.3 g/min. By applying a stoichiometric ratio of 1.16, the total gas flow rate was 5 L/min [17].

![Figure 1. Schematic of the drop tube furnace.](image-url)
3. Results and Discussion

3.1. Coal and Biomass Sample Properties

The proximate analysis, calorific value, and ultimate analysis of 20 types of coal (Nos. 1–20) used in this study are provided in Table 1. Among the coal samples, three types were American coals, two were Russian, two were Indonesian, two were Colombian, four were South African, and seven were Australian.

The FR (fixed carbon (FC)/volatile matter (VM)) calculation is provided in Table 1, and the Van Krevelen plot considering the atomic ratio is provided in Figure 2. All samples used were bituminous or subbituminous. The nitrogen content was analyzed to range from 0.99% to 2.36%. The coal’s heating values were between 6200 and 7150 kcal/kg. Three types of American coals exhibited a high moisture and oxygen content but a low sulfur content in the element analysis. Two types of Russian coals show different properties. Glencore had a high moisture and oxygen content whereas, Tugnuisky had high ash content. Indonesian coals exhibited typical characteristics of low-quality coals that have high water and oxygen content. Colombian coals exhibited the highest heat values, according to the higher heating value (HHV) standard, and the Australian coals (except for the NCA coal) exhibited the highest water content. The heating value was also high based on the HHV standard, but the overall ash contents were low. Four types of South Africa coals had very little moisture content compared to the others but had a high ash content overall. However, the fixed carbon content was very high, as was the FR.

Figure 2. Van Krevelen plot of the samples.

Table 1. Proximate analysis, calorific value, and ultimate analysis of coal samples.
Table 1. Cont.

| No. | Sample       | Country     | Proximate Analysis (as-rec basis, wt %) | Fuel Ratio (-) | Calorific Value (kcal/kg) | Ultimate Analysis (daf basis, wt %) | Atomic Ratio (-) |
|-----|--------------|-------------|-----------------------------------------|----------------|---------------------------|-------------------------------------|------------------|
|     |              |             | Mol. VM FC Ash                          |                | HHV LHV                   | C H O N S H/C                      |                  |
| 12  | Moolarben    | Australia   | 1.86 29.03 52.3 16.81                   | 1.8           | 6737 6612                 | 84.71 5.13 7.82 1.74 0.6          | 0.73 0.14        |
| 13  | NCA          | Australia   | 13.68 29.88 47.85 8.59                  | 1.6           | 7036 6673                 | 78.85 5 13.31 2.25 0.59          | 0.76 0.25        |
| 14  | Noble        | South Africa| 3.19 26.39 55.39 15.03                 | 2.1           | 6613 6402                 | 84.65 4.94 8.22 1.49 0.71         | 0.15             |
| 15  | Rio          | Australia   | 3.82 30.59 51.63 13.96                  | 1.69          | 7009 6741                 | 82.58 5.43 9.22 2.15 0.62         | 0.79 0.17        |
| 16  | Trafignura   | South Africa| 3.94 26.83 51.43 17.8                   | 1.92          | 6375 6124                 | 83.1 4.68 9.65 1.5 1.08           | 0.68 0.17        |
| 17  | Trafignura   | Australia   | 6.76 29.64 47.56 16.04                  | 1.6           | 6607 6160                 | 82.73 5.25 9.37 1.68 0.97         | 0.76 0.17        |
| 18  | Trafignura   | Australia   | 4.87 30.52 47.29 17.32                  | 1.55          | 6611 6289                 | 83.65 5.4 7.64 2.36 0.94          | 0.77 0.14        |
| 19  | Tugnansky    | Russia      | 4.14 35.59 45.59 14.68                  | 1.28          | 6802 6520                 | 82.44 5.72 9.16 2.17 0.51         | 0.83 0.17        |
| 20  | Vital        | Colombia    | 9.68 36.5 47.46 6.36                    | 1.3           | 7147 6455                 | 78.53 5.22 14.68 1 0.57           | 0.8 0.28         |

Note. Mol.: Moisture, VM: Volatile matter, FC: Fixed carbon, as-rec: as-received, daf: dry and ash-free.

3.2. DTF Experiment Results

3.2.1. NOx Emission Propensity Analyses in DTF Experiments

The concentration of NOx emissions during sample combustion as determined using DTF is provided in Table 2. The concentration of NOx emissions together with the conversion of fuel-N to NOx (CR) are provided in Table 2. In Figure 3, according to the definition of the Pearson correlation coefficient, the factors that had strong correlations and R-squared are displayed. The factors that had weak correlations were excluded. Coal samples were sorted in ascending order based on the results of the DTF test on the concentration of NOx emissions, but, as shown in Figure 3a, there was no correlation with the nitrogen content found considering the ultimate analysis of coal. However, as observed in Figure 3b, in the industrial analysis based on a received standard, the moisture content showed a strong negative correlation ($R^2 = 0.7659$). In general, moisture hinders combustibility and causes a delay in coal ignition. According to Ma et al., the moisture content is believed to be related to NOx emissions and this factor can be used to define minimum NOx emissions from the coal [18]. Figure 3c shows the correlation between NOx concentration and fixed carbon from the industrial analysis. $R^2 = 0.7063$ shows a strong positive correlation.

To consider the amount of the nitrogen component in the coal and biomass needed for NOx emissions, we determined the relation using CR, the conversion factor of fuel-N to NOx proposed by Kurose et al. [19]. CR consists of three types of indices—FR (-), nitrogen amount in fuel-N (FN, kg), and FC (-), which is drawn from the industrial analyses. CR is expressed as follows.

$$\text{CR} = \frac{C_{\text{NOx}}}{2.24 \times 10^{-2} \times \frac{\text{FN}}{\text{V}_{\text{dry}}}} \times 10^2$$

where $C_{\text{NOx}}$ is the NOx concentration at the furnace exit, and $\text{V}_{\text{dry}}$ is the flow rate of dry air per feeding rate of coal (Nm$^3$/kg). The denominator and NOx concentration, in this case, suggest that all fuel-N is converted to NOx. Coal and biomass samples are provided at 0.29–0.34 g/min for a constant heat supply so that $\text{V}_{\text{dry}}$ is calculated according for each sample. Figure 4 shows the relation between the CR and FR/FN, and the dashed line indicates the data from the referenced paper [20]. It is known that bituminous coal with FR < 2.5 exhibits a linear proportional correlation with FR/FN, and it is suggested that CR is proportional to $(1 - \text{VM}) / (\text{VM} \times \text{FN})$. VM, in this case, refers to volatile matter from the industrial analysis. CR in Figure 4 shows an exponential relation to FR/FN, and we can consider the relation with FR by excluding FN, according to the CR definition.
Table 2. Fuel-N to NOx values and DTF NOx concentration at the exit.

| No. | Sample        | Fuel-N to NOx (CR) (-) | NOx Concentration (ppmv) | No. | Sample        | Fuel-N to NOx (CR) (-) | NOx Concentration (ppmv) |
|-----|---------------|------------------------|--------------------------|-----|---------------|------------------------|--------------------------|
| 1   | Anglo         | 0.112                  | 161.65                   | 11  | Mercurai      | 0.105                  | 193.72                   |
| 2   | Clermont      | 0.172                  | 304.59                   | 12  | Moolarben     | 0.151                  | 244.39                   |
| 3   | Cloud peak    | 0.018                  | 40.16                    | 13  | NCA           | 0.067                  | 150.14                   |
| 4   | Flame         | 0.107                  | 225.35                   | 14  | Noble         | 0.172                  | 245.36                   |
| 5   | Glencore      | 0.041                  | 87.28                    | 15  | Rio           | 0.098                  | 202.26                   |
| 6   | Indominco     | 0.054                  | 95.08                    | 16  | Trafigura     | 0.124                  | 178.63                   |
| 7   | Lanna         | 0.097                  | 107.63                   | 17  | Trafigura     | 0.116                  | 186.62                   |
| 8   | Light House   | 0.05                   | 76.8                     | 18  | Trafigura     | 0.09                   | 204.17                   |
| 9   | Light House   | 0.151                  | 187.35                   | 19  | Tugnuisky     | 0.073                  | 147.94                   |
| 10  | Macquarie     | 0.098                  | 147.22                   | 20  | Vitol         | 0.143                  | 132.49                   |

Figure 3. Relation between NOx concentration and fuel-N (a), moisture (b), and fixed carbon (c).

Figure 4. Relation between CR concentration and FR/FN of the samples.
3.2.2. DTF NOx Index

The material properties of coal used to predict NOx emissions are listed in Table 3, using moisture content, according to the industrial analysis, fixed carbon, \( \ln(\frac{\text{FR}}{\text{FN}}) \), and NOx concentration calculated by an empirical formula using these three factors. The calculated and actual NOx concentrations created using the DTF test indicate a relation in Figure 5, which shows a correlation coefficient of 0.9020 and \( R^2 = 0.8137 \), which was calculated using the following equation.

\[
\text{Calculated NOx concentration} = a + b \times \text{Moisture} + c \times \text{Fixed carbon} + d \times \ln(\frac{\text{FR}}{\text{FN}})
\]  

(2)

where \( a, b, c, \) and \( d \) are function system indicators (coefficients), which are values that change according to the type of reactor and the operating conditions. Therefore, using this empirical formula, we compared it to the NOx concentration before entering SCR using the properties of the coals used in the practical operations of boiler #2 in the Hadong Thermal Power Site. At this site, two to three types of coals used for combustion were selected to meet the heat value standard and cofired. Therefore, the factors were determined by arithmetical calculation in accordance with the ratio of used coal from a single type of coal property.

Table 3. Fuel sample properties to the NOx emission prediction.

| No. | Sample  | Moisture Content (as’rec basis, wt %) | Fixed Carbon Content (as’rec basis, wt %) | FR/FN(-) | Calculated NOx (ppmv) |
|-----|---------|--------------------------------------|------------------------------------------|----------|-----------------------|
| 1   | Anglo   | 4.71                                 | 54.75                                    | 141.7    | 218.17                |
| 2   | Clermont | 2.73                                 | 58.84                                    | 115.7    | 246.31                |
| 3   | Cloud peak | 19.73                             | 39.25                                    | 52.7     | 50.42                 |
| 4   | Flame   | 1.98                                 | 48.27                                    | 82       | 205.38                |
| 5   | Glencore | 12.37                                | 45.59                                    | 61.8     | 125.16                |
| 6   | Indominco | 14.29                           | 39.31                                    | 57.3     | 86.46                 |
| 7   | Lanna   | 16.95                                | 38.45                                    | 106.1    | 70.09                 |
| 8   | Light House | 18.35                        | 42.11                                    | 87.9     | 74.7                  |
| 9   | Light House | 6.33                             | 47.33                                    | 116.7    | 175.89                |
| 10  | Macarthur | 9.18                             | 47.62                                    | 86.6     | 156.52                |
| 11  | Moolarben | 4.2                              | 53.67                                    | 109.3    | 215.2                 |
| 12  | NCA     | 13.68                                | 47.85                                    | 71.1     | 126.95                |
| 13  | Noble   | 3.19                                 | 55.39                                    | 140.9    | 230.59                |
| 14  | Rio     | 3.82                                 | 51.63                                    | 78.6     | 206.96                |
| 15  | Trafignura | 3.94                             | 51.43                                    | 128      | 208.82                |
| 16  | Trafignura | 6.76                             | 47.56                                    | 95.2     | 172.61                |
| 17  | Tugnuisky | 4.87                              | 47.29                                    | 65.7     | 181.09                |
| 18  | Vitol   | 9.68                                 | 47.46                                    | 130      | 155.52                |

In Figure 6, one data dot on the X-axis represents NOx concentration at the SCR entrance generated from used coal combustion during each 8-hour time slot during the daytime shift (D/S), after time shift (A/S), and nighttime shift (N/S). NOx concentrations that were measured in the actual boiler are confidential data so are expressed as an arbitrary unit. NOx concentrations in the actual boiler were measured at two ducts located on the front of the SCR entrance and expressed as average values and actual data. The NOx concentrations measured from the same coals show a 20% to 50% difference. This error in the difference could be from the turbulent flow inside the duct or from an error in the measuring instrument. The green dots that are calculated as evidence of a difference error must show the same NOx values calculated from the same coals used. However, we observed that the NOx concentration from actual combustion shows a major error, in some cases, in the differences in values. It is clear that, because the actual and calculated NOx concentrations do not always show the correct
trends when applying the empirical formula, as some researchers have reported, it appears correct to trust a drift in the generation of NOx concentrations since the combustion products are produced from the actual chemical reactions. Except for abnormal values showing a 25% to 37% difference in NOx concentration to that of a neighboring time slot, it is believed that the overall calculated NOx concentrations accurately represent the actual trend in NOx emissions [21]. The average absolute error containing and excluding an abnormal value were 10.45% and 6.18%, respectively. Therefore, the NOx prediction formula comprising the basic properties of coal is considered to be meaningful, unlike the previous studies, and it will be used as useful data for the preemptive response of power plants.

4. Conclusions

This study analyzed the factors that have a big effect on NOx generation based on a lab-scale DTF test. Based on our results, we developed a formula by which to predict NOx generation, applied it to an actual powerplant model, and compared the predicted and actual amounts of the generated NOx concentrations. Accordingly, we suggest the following.

1. The moisture content in coal has a negative correlation with generated NOx concentration and a positive correlation to fixed carbon. In general, the moisture content and fixed carbon are inversely proportional to each other. It is believed that the reasons for this are the delay in
ignition caused by combustion hindrance from the moisture when the coals are reacted in the DTF heating furnace.

2. CR and NOx conversion factor to fuel-N show an exponential correlation to FR. Therefore, we expressed this using the logarithm function in NOx in the prediction empirical formula to display a linear relation.

3. The suggested formula for NOx concentration was based on three factors—moisture, fixed carbon, and FR correlations. Having compared the calculated amount of NOx generation from the actual powerplant using four system indicator factors (coefficients) while considering the design and operating conditions of each boiler, except for the abnormal values that were much higher than the average NOx concentration values, the trends in the generation of NOx were determined.

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Abbreviations

| Acronym | Description |
|---------|-------------|
| NOx     | Oxides of nitrogen |
| SOx     | Oxides of sulfur |
| DTF     | Drop Tube Furnace |
| GHGs    | Greenhouse Gases |
| CO      | Carbon monoxide |
| FR      | Fuel ratio |
| HHV     | Higher Heating Value |
| LHV     | Lower Heating Value |
| VM      | Volatile Matter |
| FC      | Fixed Carbon |
| FN      | Fuel Nitrogen |
| CR      | Conversion factor of Fuel-N to NOx |
| CNOx    | NOx concentration |
| Vdry    | Flow rate of dry air |

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