Validation of Coal Combustion Model by Using Experimental Data of Utility Boilers*  

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Applicability of a coal combustion model was validated by comparing its predictions with experimental data of utility boilers. The coal combustion model had gasification and NOx reaction submodels and it was developed by using a small drop-tube-furnace (coal feed rate 0.6 kg/h). A turbulence combustion simulation program was developed by introducing the coal combustion model. The program was validated by comparing its predictions with 23 sets of experimental results which contained different plant, load and coal data. The temperature difference between simulated and experimental results was within 30°C at the furnace exit. The decreasing characteristic of coal burnout with increasing load was well predicted. The NOx emission difference between simulated and experimental results was less than 15%. The coal combustion model was judged applicable to utility boilers.

Key Words: Coal Combustion, Gasification, Numerical Simulation, Pulverized Coal, Utility Boiler, NOx, Furnace Exit Gas Temperature, Burnout, Furnace

1. Introduction

Decrease of NOx emission is required for pulverized coal combustion boilers. Studying combustion mechanisms in fuel-rich regions, where NOx is reduced to N2, is important to develop boilers with lower NOx emission. Also, combustion process modeling and development of a numerical simulation program that predicts NOx concentration, coal burnout, and temperature distribution are required to develop new boilers in a short time.

There were a few pulverized coal combustion simulation programs which predict carbon dioxide and oxygen concentrations in furnaces(1),(2). However, there are none which can predict concentration distributions of carbon monoxide and cyan, and NOx emissions. The gasification reactions, which are the reactions between coal char and carbon dioxide or steam, are the main reaction in the fuel-rich region. The reaction rate parameters of the gasification reactions have been measured in nitrogen atmosphere. The gasification reaction models that have been developed based on the experimental data were sometimes not introduced into the simulation programs, because these models did not contribute to better prediction accuracy.

Yamamoto et al.(4) developed an accurate gasification reaction model based on experimental data measured by using a drop-tube-furnace (coal feed rate was 0.6 kg/h). The prediction error of coal burnout was less than 3% in the SR (stoichiometric ratio) range of 0.4 to 1.5. They claimed that the gasification reaction rates measured under the coal-air combustion condition were accelerated. Furthermore, about 10% of coal char was found to be consumed by the gasification reactions at SR 0.8 which is a normal operating condition for utility boilers. Therefore, the gasification reactions must be introduced into the simulation program to predict utility boiler performances.

Taniguchi et al.(5) developed a NOx reaction submodel based on experimental data measured by using the drop-tube-furnace. The most important reaction to predict NOx concentration was found to be the reaction between NOx and hydrocarbon radicals formed in the gasification reaction region. Yamamoto et al.(4) developed a turbulent combustion simulation program which adopted the gasification reaction submodel and the NOx reaction submodel.
The program was validated by comparing its predictions with the experimental data measured by using a laboratory scale furnace with a coal feed rate of 25 kg/h. The simulation program predicted not only concentration distributions of major gas species but also those of carbon monoxide and cyan, which are mainly formed in the fuel-rich region. But the applicability of the program to the simulation of utility boilers was not examined. Also, the prediction accuracy of gas temperature, which is important to predict NOx emission and coal burnout, was not validated.

The purposes of this paper are (1) to apply the program to the simulation of utility boilers and (2) to validate the prediction accuracy of NOx emission, gas temperature, and coal burnout.

2. Numerical Simulation Models

2.1 Governing equations

In this paper, flow, combustion and heat transfer equations were solved to predict the boiler performances. All transport processes except radiation were represented by a general convective transport equation which could be written for Cartesian coordinates as

\[ \frac{\partial \phi}{\partial t} + \frac{\partial}{\partial x_i} \left( \rho u_i \phi + \Gamma_\phi \frac{\partial \phi}{\partial x_i} \right) = S_\phi \quad (1) \]

\[ \phi = \{1, u, v, w, h, Y_j, k, \epsilon, C_{char}, C_{vol}, C_{ash} \} \quad (2) \]

\[ \Gamma_\phi = \mu + \mu_T \quad (3) \]

where \( \phi \) is a dependent variable, \( \Gamma_\phi \) is the effective viscosity coefficient, and \( S_\phi \) is a source term. The SIMPLE \(^7\) method was adopted for the calculation of momentum and pressure. The gas compositions \( Y_j \) were solved for \( N_2, O_2, CO_2, CO, H_2O, H_2, CH_4, NO, \) and \( XN. \) \( XN \) represents the sum of HCN and NH, and it is mainly generated in the devolatilization and gasification regions. The standard k-\( \epsilon \) turbulence model was used. The mass concentrations of char (\( C_{char} \)), volatile matter (\( C_{vol} \)), ash (\( C_{ash} \)) were solved in the Eulerian coordinate system \(^6\).

2.2 Gas combustion model

There are two important phenomena in gas combustion. First, if reaction time is sufficiently long, gas concentrations reach the values of chemical equilibrium. Second, total reaction rate depends on chemical reaction rates and a turbulent mixing rate. In particular, the first phenomenon is important to predict the gas concentrations in fuel-rich region, but there are only a few simulation programs that reproduce it \(^8\). In this paper, total reaction rate was calculated only from the turbulent mixing rate because the chemical reaction rate was high \(^9\). Eddy dissipation concept proposed by Magnussen and Hjertager \(^9\) was used to calculate the total reaction rate described as follows,

\[ R_t = \rho (\bar{Y}_f - \bar{Y})/\tau_t \quad (4) \]

where \( \tau_t \) is a characteristic time scale of the turbulent reaction calculated by the their model. When the gas concentration approaches those of chemical equilibrium, the total reaction rate becomes zero. By using this formulation, the two important phenomena are reproduced.

2.3 Coal combustion model

Lagrangian methods are widely used for the calculation of particle transportation in pulverized coal combustion \(^10\). However, it was found that convergence of the solution was not obtained when a few sampling particles were used for the Lagrangian method, so, a simplified Eulerian particle transportation method \(^6\) was used in this study. The transportation equation was solved for three components of the coal (char, volatile, and ash).

The two-competitive reaction model \(^11\) was adopted to predict the coal devolatilization. The reaction rate was

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Nomenclature

- \( Ac \): Frequency factor \( \text{kg/m}^2\text{sPa} \)
- \( C \): Solid concentration in gas \( \text{kg/kg} \)
- \( D \): Diffusion rate \( \text{m}^2/\text{s} \)
- \( Ec \): Activation energy \( \text{J/mol} \)
- \( h \): Specific enthalpy \( \text{J/kg} \)
- \( k \): Turbulence energy \( \text{m}^2/\text{s}^2 \)
- \( k_f \): Surface reaction rate coefficient \( \text{kg/m}^2\text{sPa} \)
- \( k_d \): Diffusion coefficient \( \text{kg/m}^2\text{sPa} \)
- \( M_e \): Molecular weight of carbon \( \text{kg/mol} \)
- \( m_p \): Weight of one particle \( \text{kg} \)
- \( n \): Number density of particles \( \text{1/m}^3 \)
- \( P_f \): Partial pressure \( \text{Pa} \)
- \( R \): Universal gas constant \( (=8.31) \text{ J/mol K} \)
- \( R_j \): Reaction rate \( \text{kg/m}^3\text{s} \)
- \( S_{char}, S_{vol} \): Surface area of a particle \( \text{m}^2 \)
- \( T \): Temperature \( \text{K} \)
- \( u, v, w \): Gas velocity \( \text{m/s} \)

Subscripts

- \( j \): Gas species
- \( m \): Boundary layer value
- \( p \): Particle
- \( r \): Turbulent
- \( 0 \): Raw coal value

Superscripts

- \( * \): Raw coal value
- \( \circ \): Equilibrium value
measured by devolatilization experiments\(^{(12),(13)}\) which used a pulsed LASER to heat up particles.

The char reaction model\(^{(4)}\) used in this study was developed based on the experimental data measured by using a drop-tube-furnace for various conditions of coal-air combustion. Measured and calculated burnouts were compared for wall temperatures of 1 173 to 1 673 K and the stoichiometric ratio of 0.4 to 1.5. The prediction error for the burnout was within 3%.

In the char reaction model, char was assumed to react with O\(_2\), CO\(_2\) and H\(_2\)O as described in Eqs. (5) to (7). The oxidation reaction was defined as Eq. (5). Gasification reactions were defined as Eqs. (6) and (7). The total char reaction rate depended on gas diffusion rates and an apparent reaction rate (Eqs. (8) – (10)). The apparent reaction rate means the combination reaction rate of a surface reaction and pore diffusion.

The external surface area was calculated by Eq. (11). \(W_c\) and \(W_a\) are the ratios of the remaining weight of char or ash to the weight of an original coal particle, respectively. The initial particle diameter of coal was given by the volume-surface mean diameter \(d_{32}\). The diameter of a particle was assumed to be unchanged on devolatilization and decreased with combustion of char. The surface area was calculated as described Eq. (12) from the mass concentration of char and ash obtained from the transport Eq. (1). The number density of particles was assumed to be unchanged in the combustion processes, and was calculated by Eqs. (13) and (14).

\[
\begin{align*}
C + 1/2 O_2 &\rightarrow CO \\
C + H_2O &\rightarrow CO + H_2 \\
C + CO_2 &\rightarrow 2CO \\
S_{\text{char}} &= \sum_j \frac{P_j S_{\text{ext}j}}{k_{d,j} + \frac{1}{k_{s,j}}} \\
&= C_{\text{char}} + C_{\text{ash}} \\
C_{\text{char}} &= \frac{W_c}{W_c + W_a} \\
C_{\text{ash}} &= \frac{W_a}{W_c + W_a} \\
n &= \rho(C_{\text{char}} + C_{\text{ash}}) / m_p
\end{align*}
\]

\[2.4 \ NO_x \ reaction \ submodel\]

The NO\(_x\) reaction submodel proposed by Taniguchi et al.\(^{(5)}\) was used in this study. The reaction schemes of the submodels are shown in Fig. 1. Figure 1 (a) shows the reaction scheme of nitrogen species. Figure 1 (b) shows the reaction scheme of hydrocarbon radicals. The reaction scheme of volatile N was based on Fenimore’s mechanism\(^{(14)}\). In addition, NO\(_x\) reduction by hydrocarbon radicals (CH\(_i\))\(^{(15)}\), formation of NO\(_x\) from char-N, and the extended Zeldovich mechanism\(^{(16)}\) were introduced. The average difference between calculated and measured NO\(_x\) concentration was less than 17% at the furnace exit.

Here, the reaction route of nitrogenous compounds in low-NO\(_x\) combustion is described briefly. First of all, XN is generated by devolatilization (R1). Since the oxidation reaction rate (R2) of XN is high in the oxygen-rich region, XN changes to NO\(_x\) immediately. Therefore, the increasing point of NO\(_x\) concentration is detected upstream to that of XN concentration\(^{(17)}\).

Next, the low oxygen concentration region is formed in the fuel-rich region, and is generated by devolatilization and char surface reactions. In recent years, with the use of low volatile coal, NO\(_x\) emission has been decreased with a low-NO\(_x\) burner\(^{(18)}\). Therefore, the char surface reactions are important to decrease NO\(_x\) emission. The main reactions of nitrogen species in the fuel-rich region are NO\(_x\) reduction reactions (R3, R6). In the submodels, hydrocarbon radicals were assumed to be generated by the char surface reactions of gasification.

Previously, it has been assumed that hydrocarbon radicals were generated only by devolatilization (R7)\(^{(19)}\). As the char in flames had about one mol of hydrogen to eight mols of carbon\(^{(5)}\), hydrocarbon radicals were assumed to be formed from the hydrogen (R8). The reaction route R6 is particularly important for low volatile coal. When R6 is excluded, XN concentration may be overestimated, and the NO\(_x\) reduction reaction (R3) rate may be underestimated.

Lastly, in the complete combustion region, XN is ox-
idized to NOx by reaction route R2. NOx is formed by the combustion of char (R4) because the char contains nitrogen. Also, the char reduces the NOx simultaneously (R5).

2.5 Radiation model

A DT (Discrete Transfer) radiation method(20), which is a kind of ray-tracing method, was used to evaluate radiative heat transfer. An energy conservative formulation(21) was adopted to conserve energy. The number of rays was 36 on every radiative heat absorption element. The DT method is an economical and efficient method for the calculation of combustion chambers(21).

A total heat absorption coefficient is the sum of gas and particulate heat absorption coefficients. The heat absorption coefficient of combustion gases, which contain CO2 and H2O, was calculated by the WSGG (Weighted Sum of Gray Gases)(22) method, which is a kind of optical database. On the other hand, heat absorption coefficient of particles (e.g. raw coal, char, and fly ash) was predicted by the Mie scattering theory, which involves the particle diameter and wavelength of thermal radiation. By using this theory, the change of the particle heat absorption coefficient in combustion was calculated.

3. Operating Conditions of Pulverized Coal Combustion Boilers

Boiler performance values such as NOx emission and gas temperature are changed under conditions such as maximum output of boilers, load, and coal type. The numerical simulation program must predict the boiler performance variation according to condition changes. In this study, the simulation program was validated for 23 sets of conditions described in Tables 1 to 3. Three boilers were used for the validation. The maximum electric output of the boilers ranged from 500 to 1000 MW and their basic structure is shown in Fig. 2. Burners were used to burn pulverized coal and air and OAPs were used to burn unburned char or flammable gases such as CO and H2. There were several platen superheaters at the top of each furnace.

The measured values were obtained in steady conditions for which boiler loads are ranged from 35 to 100%. Coal feed rates increased with the load. The properties of the coals are described in Table 3. The fuel ratio (= fixed carbon/volatile matter) of the coals ranged from 1.2 to 2.4 which covers most coals utilized for pulverized coal combustion boilers in Japan. Since the reaction rate of the coals with higher fuel ratio is low, the concentration of NOx reduction species such as hydrocarbon radicals is low. Therefore, the NOx emission is higher if higher fuel ratio coal is used.

4. Results and Discussion

First, the calculated and the measured gas temperatures are compared. The gas temperature in combustion fields is important to predict char and NOx reaction rates. However, it is difficult to measure gas temperatures in pulverized coal combustion boiler furnaces for the following reasons. (1) Inserting a probe into the center of the furnaces is difficult because they are large. (2) measuring temperature continuously is difficult because melting ash sticks to the probe. (3) measuring temperature for a long time is necessary because the combustion fields are unsteady.
FEGT (Furnace Exit Gas Temperature) is measured indirectly from economizer outlet gas temperature and heat absorption of heat exchangers. It is defined as the average temperature at the furnace exit as described in Fig. 2. A flue gas flow rate and gas concentrations are estimated from operating conditions. It is easy to measure the economizer outlet gas temperature because it is low (about 400°C). The specific enthalpy of the flue gases at the economizer outlet is calculated from the measured temperature and the flue gas concentration. The heat absorption of heat exchangers arranged from the furnace exit to the economizer outlet is calculated from the inlet and outlet steam (or water) temperatures. The specific enthalpy of furnace exit flue gases is calculated from that of economizer outlet flue gases and the heat absorption. The measurement error of this method is about ±10°C. The FEGT measured by this method is the space averaged value at the furnace exit.

The differences of measured and calculated FEGTs are shown in Fig. 3 and are within ±30°C. The operating conditions used for this comparison are cases 1 to 19. Furnace heat absorption can be calculated from boiler thermal input and FEGT. The prediction error of the heat absorptions calculated from FEGTs is within ±5%. The heat absorption depends on reaction rates, which vary according to positions of heat sources. Therefore, the highly accurate prediction of heat absorption indicates that the reaction rates are valid.

Figure 4 shows the gas temperature distribution for case 11 on a surface that includes the center of the burners. The gas temperature is high near the burners and it decreases towards the exit of the furnace because of heat absorption by the furnace water walls. The temperature at the center of the furnace on the burner height level is lower than that near burners. This is because the gasification reactions which occur in the area are endothermic reactions.

Next, the concentration distributions of CO and NO\textsubscript{x} are examined. The CO distribution for case 11 is shown in Fig. 5(a). The basic mechanism for the low NO\textsubscript{x} burner is improvement of the NO\textsubscript{x} reduction reaction by providing a wide fuel-rich region due to rapid coal ignition\textsuperscript{(25)}. The CO concentration near the burners is high because coal is ignited there by using the low NO\textsubscript{x} burner. The CO concentration increases from the burners to the OAP height because the gasification reactions occur in the fuel-rich region. Downstream from the OAPs, CO concentration de-
creases to almost zero. The behavior that the CO concentration is high near the burners and decreases downstream from the OAPs agrees with experimental behavior\(^{(26)}\). The NO\(_x\) concentration (0% O\(_2\)) on the same surface is shown in Fig. 5 (b). The NO\(_x\) concentration near burners is high. This is because the XN generated by devolatilization (reaction route R1) is oxidized to NO\(_x\) (R2) rapidly. In the fuel-rich region from the burners to the height of the OAPs, the NO\(_x\) concentration is decreased by NO\(_x\) reduction reactions R5 and R6. After the mixing of OAP air, NO\(_x\) concentration is increased because oxidation reactions through the reaction routes R4, R3, and R9 occur in the oxygen rich region. The NO\(_x\) concentration decreases near the furnace exit. This is because NO\(_x\) is decreased by the char reduction reaction (R5). The behavior mentioned above agrees with the measured behavior\(^{(26)}\).

The measured and calculated NO\(_x\) emissions are compared for the cases 8 to 11 and 17 to 23. The results are shown in Fig. 6. The calculated NO\(_x\) emission is the averaged value at the furnace exit. The measured NO\(_x\) emission is obtained at the economizer outlet. The NO\(_x\) reaction is assumed to be quenched at the furnace exit. The difference between measured and calculated NO\(_x\) emissions is less than \(\pm 15\%\). The NO\(_x\) prediction accuracy for the actual utility boilers is almost the same as that for the drop-tube-furnace. The NO\(_x\) model is able to predict NO\(_x\) emissions of utility boilers. The measured NO\(_x\) emission increases with fuel ratio, for example C2 to C3 or C3 to C5. This characteristic agrees with the calculated one. On the other hand, the measured NO\(_x\) emission increases with the boiler load. Since the coal feed rate is increased with the boiler load, the residence time in the fuel-rich region, which is important to reduce NO\(_x\) emission, is decreased. The calculated results agree with the measured ones except for case 8 (boiler B1, coal C5, load 35%). The NO\(_x\) reaction submodel was proper. The combustibility under the operating condition of case 8 is low because of the high fuel ratio coal and low load operation. The NO\(_x\) emission increases due to flame instability which occurs under this condition is not predicted accurately. The char and NO\(_x\) reaction submodel was developed based on the experimental data obtained below 1673 K. The maximum temperature of the calculated results is higher than 2000 K. The submodel is found to be applicable to the high temperature conditions.

Figure 7 shows prediction accuracy of coal burnout for cases 8 to 11. The measured and calculated coal burnouts decrease with increasing boiler load. This is because the combustion time in the furnace is shortened by the increase of coal feed rate. Since the prediction error of coal burnout is less than 0.3%, char reaction model was
found to be applicable to the simulation of utility boilers.

5. Conclusions

Applicability of a coal combustion model was validated by comparing its predictions with 23 sets of experimental results for utility boilers that had different plant load and coal data. The simulation program predicted the NOx emission, furnace exit gas temperature and coal burnout accurately.

(1) The difference in measured and calculated NOx emissions was less than ±15%. The predicted NOx emission increased with coal fuel ratio and also with boiler load. These characteristics of NOx emission agreed with the experimental results.

(2) FEGT (Furnace Exit Gas Temperature) was indirectly measured from economizer outlet temperature and heat absorption of heat exchangers. The difference in predicted and measured FEGTs was within ±30°C. This corresponded to 5% error of the furnace heat absorption.

(3) The measured and calculated coal burnouts decreased with increasing boiler load. The prediction error for the coal burnout was less than 0.3%, and it was the same level for the results of the drop-tube-furnace.

(4) This program predicted the following characteristics: The NOx concentration increased near the burners, decreased in high temperature fuel-rich region, and increased downstream from the over fire air port.

References


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