A Two Mixture Fraction Approach for Modeling Turbulent Combustion of Coal Volatiles and Char Oxidation Products

Daniel V. Flores and Thomas H. Fletcher
Chemical Engineering Department
Brigham Young University
Provo, Utah 84602

Paper 95S-120
Spring Meeting of the Central States/Western States/ Mexican National Sections of the Combustion Institute and the American Flame Research Committee. April 23-26, 1995

ABSTRACT

Previous coal combustion models using assumed-shape PDF's to treat turbulence-chemistry interactions have used only one progress variable to treat products from coal reactions. This assumes that the products of all coal reactions have the same composition. However, the composition of the combustion products of coal particles is known to vary with burnout, especially between devolatilization and char oxidation. In this work, two progress variables were implemented which distinguish between the products of devolatilization and those of char oxidation. In addition, this new approach neglects turbulent fluctuations on char combustion products. Required input parameters are the elemental composition of char and volatiles, together with the volatile yield. Calculations are presented for an axisymmetric (hypothetical) case using both the previous and new models. Significant differences are shown in predicted values of gas temperature, CO_2 and NO near the burner.

1. INTRODUCTION

In many comprehensive coal combustion codes, one progress variable is used to treat coal combustion products. Gas phase reactions between coal combustion products and inlet oxidizers (e.g., air) are assumed to be mixing-limited, and treated using an assumed-shape PDF. The use of one progress variable for the coal combustion products means that the composition of the coal off-gas is the same as that of the parent coal and is constant throughout both devolatilization and char oxidation. Therefore, to characterize the chemistry in turbulent environments, one mixture fraction is used for the oxidizer streams (primary versus secondary air) and another for the coal off-gas. However, it is well known that coal volatiles are released from the coal particle at a faster rate than char oxidation products. Since the coal volatiles are generally enriched in hydrogen and oxygen, compared to the coal char, the use of distinct mixture fraction variables for volatiles and char off-gases is warranted, in which case, a total of three mixture fractions are needed.

1.2 Previous research

Two studies directly related to the present work are found in the literature. Brewster, et al. [1] developed a generalized theory that includes several mixture fractions to describe the gas phase turbulence/chemistry interactions. Brewster and coworkers used a version of PCGC-2 (Pulverized Coal Gasification and Combustion, 2-Dimensional) that was coded for only two mixture fractions [2], although they presented the generalized theory for multiple progress variables. By setting the oxidizer in the primary and secondary inlets to the same composition and temperature, they were able to use two mixture fractions to model the volatiles, the char off-gases and the oxidizer. Furthermore, they assumed the residual char to be pure carbon and a fixed volatile yield. These calculations showed a drastic difference in the gas temperature predictions from the calculations using one mixture fraction for coal combustion products. Eddings, et al. [3] used different mixture fractions to account for devolatilization and char oxidation products separately. In addition, they assumed the volatiles nitrogen to appear as HCN in the gas phase while the char nitrogen was assumed to appear as NO and N_2 in a fixed proportion. However, the difference in elemental composition of both volatiles and char-off gases was not accounted for. Calculations with their new method predicted trends of NO_x emissions as a function of coal type for a laboratory scale furnace, whereas the one coal mixture fraction approach was unsuccessful.

1.3 Present Contribution

In the present work, one mixture fraction is used to distinguish between primary and secondary air streams, and two progress variables are used to treat mixing between oxidizers and pulverized coal combustion products. The source terms of char off-gases and volatiles to the gas phase are obtained from Lagrangian treatments of particle trajectories and reactions. The Chemical Percolation Devolatilization (CPD) model [4] is used for devolatilization rates, and char oxidation rates are taken from Hurt and Mitchell [5]. The total devolatilization yield is as estimated by the CPD model. The elemental composition of the char is taken from elemental composition data for fully-devolatilized chars in a flat flame burner [6]. The elemental composition of the volatiles is calculated by mass balance from the volatiles yield and the elemental composition of the char.

The computer code used for these calculations was PCGC-3 (Pulverized Coal Gasification and Combustion, 3-Dimensional) [7].

2. THEORY

2.1 Major Gas Species

The mixture fractions to be used are defined as follows:

$$f_1 = \frac{m_s}{m_p + m_s} \qquad (1)$$

$$f_2 = \frac{m_v}{m_p + m_s + m_v} \qquad (2)$$

$$f_3 = \frac{m_c}{m_p + m_s + m_v + m_c} \qquad (3)$$

where m_p, m_s, m_v and m_c represent the mass of gas originating from the primary gas stream, secondary gas stream, coal volatiles, and char oxidation products, respectively. Each mixture fraction may vary from zero to unity. On the other hand the enthalpy h is partitioned into the adiabatic enthalpy of the mixture $h(f_1, f_2, f_3)$ and an average residual energy ($\tilde{h}_r$) as follows:

$$h = h(f_1, f_2, f_3) + \tilde{h}_r \qquad (4)$$

Thus, the scalar properties β of the gas phase, such as temperature and density, are only a function of the mixture fractions and $\tilde{h}_r$. The Favre-average values of any property β are then calculated by convolution of instantaneous equilibrium properties over a clipped Gaussian distribution based on f_1, f_2, f_3 as in the following equation:

$$\tilde{\beta}\left(f_1, f_2, f_3, \tilde{h}_r\right) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \beta\left(f_1, f_2, f_3, \tilde{h}_r\right)\tilde{P}\left(f_1, f_2, f_3\right) df_1 df_2 df_3 \qquad (5)$$

In this research, a simplification is made to the model proposed by Brewster, et al. [1] in order to reduce the computational time requirement. The fluctuations due to turbulence in the char off-gases are neglected (i.e., $g_{f_3} = 0$). This simplification is justified as follows:

1. The char off-gas is defined to be the mass of char that ends up in the gas phase. For example, the carbon in the CO or CO_2 produced from char oxidation is accounted for in f_3, but the oxygen is not included in f_3 because it probably comes from the primary or the secondary air streams. The oxidizer and C from char combustion are therefore mixed as soon as reaction occurs, and fluctuations are not needed.
2. The char reactions are thought to be slow enough to be independent of temperature and concentration fluctuations.
3. Experience in calculations has shown that the turbulent fluctuations are only significant (>5% of the mean) in the near-burner regions, where devolatilization occurs.

With this simplification, the gas phase properties are still calculated by convolution over the clipped Gaussian distribution, as follows:

$$\tilde{\beta}\left(f_1, f_2, \tilde{f}_3, \tilde{h}_r\right) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \beta\left(f_1, f_2, \tilde{f}_3, \tilde{h}_r\right)\tilde{P}\left(f_1, f_2\right) df_1 df_2 \qquad (6)$$

and the joint probability distributions are separated, as in previous calculations. In order to model the chemistry/turbulence interaction, transport equations for the Favre-averaged mixture fractions and their variances need to be solved. The equations for the mixture fractions are:

$$\nabla\cdot\left(\begin{array}{l}\bar{\rho}\tilde{u}\left[\tilde{f}_1\left(1-\tilde{f}_2\right)\left(1-\tilde{f}_3\right)\right]\\ -D_f^t\,\nabla\left[\tilde{f}_1\left(1-\tilde{f}_2\right)\left(1-\tilde{f}_3\right)\right]\end{array}\right)=0 \qquad (7)$$

$$\nabla\cdot\left(\bar{\rho}\tilde{u}\left[\tilde{f}_2\left(1-\tilde{f}_3\right)\right]-D_f^t\,\nabla\left[\tilde{f}_2\left(1-\tilde{f}_3\right)\right]\right)=\bar{S}_p^{\tilde{f}_2} \qquad (8)$$

$$\nabla\cdot\left(\bar{\rho}\tilde{u}\tilde{f}_3-D_f^t\,\nabla\tilde{f}_3\right)=\bar{S}_p^{\tilde{f}_3} \qquad (9)$$

where $\bar{S}_p^{\tilde{f}_2}$ and $\bar{S}_p^{\tilde{f}_3}$ are the rates of production of the volatiles and char off-gases, respectively. Descriptions of the transport equations for the variances g_{f_1} and g_{f_2} may be found elsewhere [8].

2.2 NO_X

The NO_X submodel used in PCGC-3 [9] was modified in order to differentiate between the nitrogen evolved with the volatiles and that evolved with the char off-gas. The solution procedure of the new submodel is analogous to that of the former versions and is described here briefly. Because the reaction rates involved in the mechanism of NO_X formation are slower than the turbulent fluctuations, the effects of turbulence in the kinetics cannot be neglected as in the mixing-limited approach (Eq. 6). In contrast, the rates of nitrogen addition to the gas phase from char and volatiles are not directly affected by the turbulent fluctuations. Because the concentrations of NO_X are very low (parts per million), the reactions involved in its formation are assumed not to affect the variables such as temperature, the mixture fractions, and the concentrations of major species. Thus, the NO_X calculations can be decoupled from the solution of the major variables, and operated as a post-processor. Conservation equations are solved for the three main species in the reaction mechanism (NO, HCN and NH_3). The global rate expression for fuel NO is taken from DeSoete [10]; thermal NO calculations are based on parameters reported by Bowman [11] and by Miller and Bowman [12]. The mean values of the gas phase reactions are obtained by convolution over the fluctuating mixture fractions [9]. It is assumed that the local fractional conversion x_i (where i= NO, HCN or NH_3) is not affected by the turbulent fluctuations. Thus, the local instantaneous concentrations (for the PDF solution) can be calculated from the fractional conversion obtained using the Favre-averaged concentrations (from the conservation equations). This is an iterative procedure.

As illustrated in Figure 1, in the new approach the nitrogen evolved with the volatiles appears in the gas phase either as HCN or as a user-specified partition (α)between HCN/NH_3. The nitrogen evolved form the char is assumed to appear as NO and N_2 only; the partition (ζ_{char}) is user-specified. The recommended value for ζ_{char} is taken from Pershing and Wendt [14], who showed that about 15 % of the nitrogen contained in the char is converted to NO.

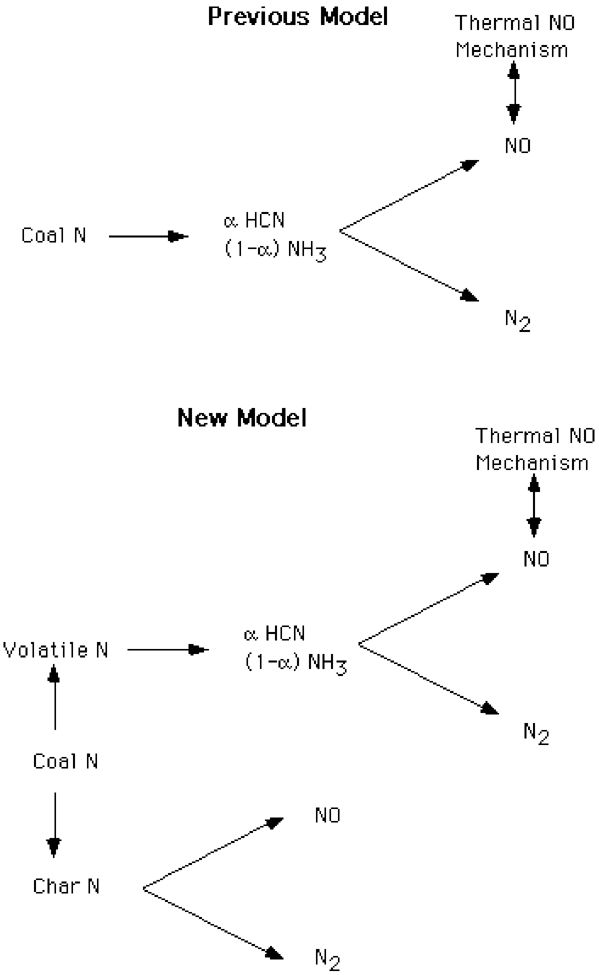

Figure 1. Illustration of the previous and new NO mechanisms.

3. RESULTS AND DISCUSSION

Calculations were made using PCGC-3 to compare the use of one versus two coal product mixture fractions. Some preliminary results are shown in Figure 2 for a hypothetical non-swirling two-dimensional laboratory furnace similar to the BYU Controlled-Profile Reactor [15]. The simulation was made for 58 kg/hr of pulverized Pittsburgh #8 coal burned in air at atmospheric pressure at an equivalence ratio of 0.96.

Predicted gas temperatures are shown in Figure 2 for the one versus the two coal product mixture fraction approach. The predictions shown here have been radially-averaged (for brevity). At first glance, the calculated temperatures by the two methods seem to be very similar (within 10%). However, major differences were found near the inlet, where the radially-averaged gas temperatures from the two predictions differ by ~200 K. Radial profiles in this near-burner region (not shown) indicate more significant differences.

Radially-averaged predictions of coal burnout and mole fractions CO_2, and NO are shown as a function of axial distance in Figures 3-5. The most significant differences between the two predictions are found near the burner (at axial distances of 0 to 1.5 m). The two predictions seem to become more similar near the exit of the reactor, where products reach chemical equilibrium based on the local enthalpy.

The radially-averaged NO concentrations predicted with two coal mixture fractions were lower and exhibited a smoother profile than those predicted with one coal mixture fraction. This is also seen in radial profiles of NO (not shown here) at different axial distances. The reason for this difference in the predictions is due to (a) the change in the amount and release rate of HCN in the new model, and (b) changes in the local equivalence ratio because the volatiles from the two coal mixture fraction prediction are enriched in hydrogen compared to carbon. In the new model, only ~50% of the nitrogen evolved from the coal passes through the HCN intermediate. The amount of NO produced from the char is totally based on the empirical input efficiency (presently 15%). Because the volatiles are lean in nitrogen compared to the parent coal, lesser amounts of NO-forming species are produced (consistent with the HCN and NH_3 concentration profiles). In addition, only a small fraction of char nitrogen is converted to NO.

4. SUMMARY AND CONCLUSIONS

The differences in predicted temperature, major gas species concentrations, and NO concentration between the one and two coal mixture fraction approaches are significant, especially in the near burner region where low-NO_x burner strategies are generally implemented. The results presented here demonstrate the need to model the composition of the volatiles separately from the char. Detailed comparisons of model predictions versus experimental data are in progress. However, the results shown here indicate that the incorporation of improved physical models (i.e., the two coal gas mixture approach) significantly affects predictions, and should therefore be used instead of the previous approach that assumed uniform coal off-gas composition. There is a significant need for detailed experimental data regarding the yield and composition of volatiles versus char as a function of coal type and operating conditions. A better understanding of the conversion of char nitrogen to NO as a function of coal type and operating conditions is also needed.

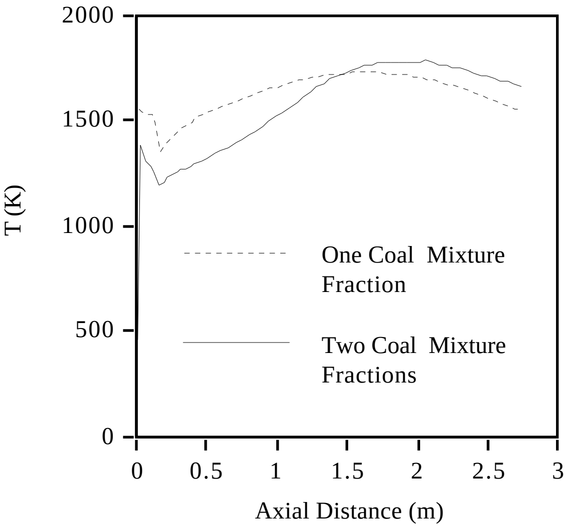

Figure 2. Radially-averaged predictions of the gas temperature as a function of axial distance in a laboratory-scale furnace.

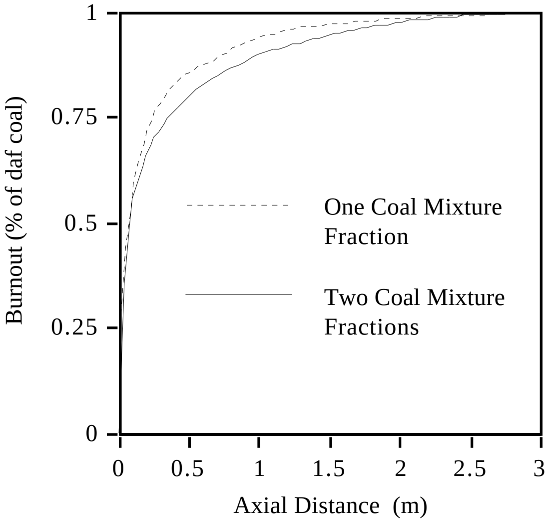

Figure 3. Radially-averaged predictions of coal burnout (% daf) as a function of axial distance in a laboratory-scale furnace.

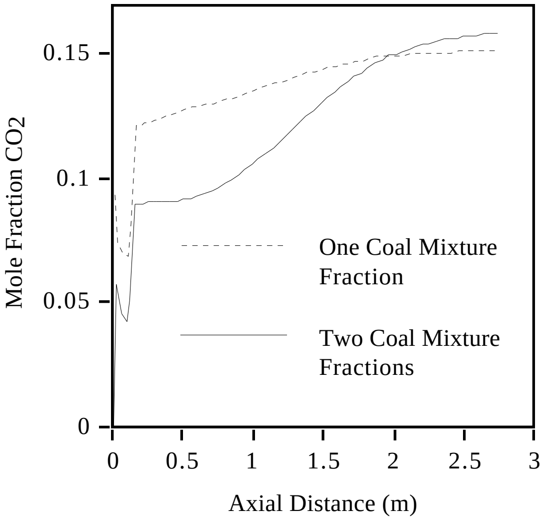

Figure 4. Radially-averaged predictions of the mole fraction of CO_2 as a function of axial distance in a laboratory-scale furnace.

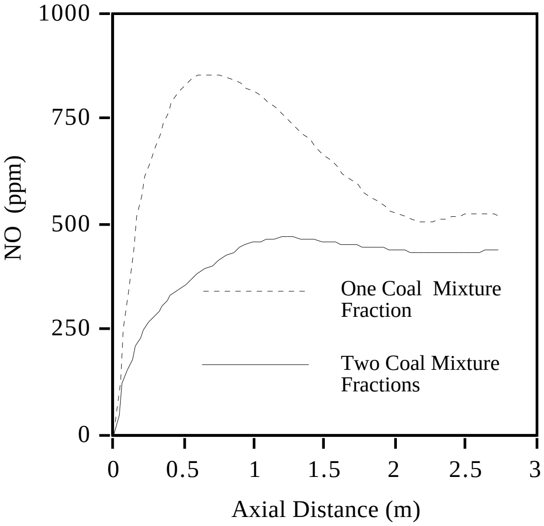

Figure 5. Radially-averaged predictions of NO concentrations as a function of axial distance in a laboratory-scale furnace.

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