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An investigation into a carbon burnout kinetic model for oxy–coal combustion

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ABSTRACT

This paper investigates coal char burnout kinetics in the oxygen-enriched environments of oxy–coal combustion. The Carbon Burnout Kinetic model (CBK) developed by Hurt et al. was used as the basic model for air–coal combustion and a modified version for oxy–coal combustion. In order to assess the performance of the original CBK model published experimental data from standard char combustion studies at near-atmosphere and elevated pressures were used. The results of the evaluation suggested that the modified model can be utilized for oxy–coal applications. The modifications include the integration of Langmuir–Hinshelwood kinetic rate mechanisms for C–CO_2 and C–H_2O reactions and the effects of CO and H_2 concentration on these reactions. The modified code is validated using published experimental data. It is shown that using this modified code, the burnout prediction in both N_2 and CO_2 environments has improved significantly. Computed results of Loss on Ignition (LOI) in a 1 MW_{th} pilot scale combustion test facility are compared with experimental data.

1. Introduction

Oxy–coal combustion in recycled flue gas (RFG) and oxygen is a carbon abatement technology that can be used for the capture of carbon dioxide (CO_2) from coal-fired power plants [1,2]. Successful implementation of oxy–coal with RFG depends on fully understanding the difficulties resulting from replacing nitrogen with CO_2 in the oxidizer stream. The change from inert nitrogen to reactive CO_2, can change coal devolatilisation and lead to the formation of a char with a different structure and an ash with a different level of unburned carbon [3–5].

Generally, the amount of unburnt carbon or the Loss on Ignition (LOI) depends on the particle residence time in the furnace, the temperature and the availability of oxygen along the particle path. Extensive studies of char burnout in air–coal combustion have been reported [1–5]. The char particle temperature is balanced by the burning rate and heat flux to the surrounding environment. Therefore, the correct prediction of burning rate is essential to calculate accurately the particle temperature and the heat transfer profile. The main challenge regarding modeling char burnout in oxy–coal combustion is that the approach and physical sub-models used in the numerical calculations are based on air–coal combustion conditions where the assumption is that oxygen is the only oxidizing gas. In oxy–coal combustion with RFG, the CO_2 content is significantly higher in the combustion zone and therefore neglecting its effect on combustion could lead to false predictions. In a more complicated situation, it has been suggested that the steam reaction with char, may inhibit the C–CO_2 gasification [6–8]. The endothermic nature of the gasification reactions affects the particle temperature and therefore the gas temperature in the combustion zone.

In this study, carbon burnout kinetic models have been assessed for their capabilities of prediction of LOI at oxy–coal combustion conditions. Based on the results, the Langmuir–Hinshelwood (LH) kinetic rate expressions have been selected to describe these gasification reactions for their implementation in these models. Validation of the modified code by comparison to the published experimental data is presented. An application of the burnout model for modeling of the air- and oxy–coal combustion in a pilot scale test facility is discussed.

2. Char combustion models

Several studies have been carried out in order to obtain data for a fundamental interpretation of the oxy–coal combustion process with a focus on the char combustion and prediction of the burnout [1–5,9,10]. For instance, in a study by Wall et al. [2], it was suggested that in the low temperature region (zone I), combustion rates are similar in both air–coal and oxy–coal environments but that with increasing temperature (transition to zone II), char reactivity increases in oxy–coal combustion as gasification reactions become important. However, it should be noted that the C–CO_2 and C–H_2O reactions are much slower than that of char combustion [11]. In the diffusion-controlled region (zone III), lower combustion rates are observed since O_2 diffusion in CO_2 is lower than its diffusion in N_2 and the influence of this zone is extended to lower temperatures. Transition between the combustion zones is important since it dictates the variation of the diameter and density of the particle during combustion process.

Detailed coal/char kinetic data is required to predict combustion performance and the extent of the LOI. Structural properties of the

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coal such as total surface area and porosity are important parameters that affect the char combustion process significantly. Gaseous species such as oxygen react with the carbon on the particle surface and inside the pores. The surface area of the char particle is a variable function of time–temperature history of the initial coal particle, coal rank and petrography of the coal. The total surface area depends on the heating rate, rate of devolatilisation and the oxidation and fragmentation stages. With the growth of pores during devolatilisation, total surface area increases and this area decreases as the oxidation process progresses and influence of ash is increased. However, assuming a constant value for total surface area based on the coal rank is a common practice in char combustion modeling which might contribute to considerable errors in prediction of burnout rate.

There have been different approaches for modeling char combustion, whether the reaction is assumed to occur only on the particle's external surface or on the total available surfaces of the particle. Most of the standard/classic char combustion models provide adequate results if only an approximate prediction is required. In most of these models, the carbon oxidation reaction mechanism is described in terms of the elementary processes of absorption, complex formation and desorption. However, these models are not expected to work as well under oxy–coal conditions as they do for conventional combustion. A comprehensive review of the common char combustion models is given by Edge et al. [1], Wall et al. [2] and Williams et al. [12].

The intrinsic model is an advanced char combustion model introduced by Smith [13]. In this model, pores and their effects on diffusion, which have great influence on char burnout, are considered along with the reaction on the particle external surface. Intrinsic reactivity is defined in an Arrhenius form which allows for a detailed analysis of the combustion of the carbon. In the intrinsic model of Smith [13], a burning mode parameter, alpha, is used to model the transition between the combustion zones. Burning mode is a measure of the degree of oxygen diffusion onto the external surface of the coal particle. A value of 0.25 for alpha that is selected for this study indicates a combination of diffusion and kinetic control.

Hurt et al. [14] developed the CBK package that can be employed for a wide range of coals and the computer code was made generally available for non-commercial use in 1998. This code is a variation of the Smith [13] intrinsic model and it was specifically designed to predict the total extent of carbon burnout and ultimate fly ash carbon content for prescribed oxygen–temperature histories typical of pulverized air–coal combustion. It is particularly suitable for post-processing applications where a comprehensible furnace model provides the particle temperature and vectors [15]. In this zero-dimensional model, defining the time–temperature history of the gas phase for a known coal allows the prediction of particle temperature and diameter along with its burnout ratio. One of the key advantages of CBK is its ability to be used as part of advanced models which include transport properties and geometries such as computational fluid dynamics (CFD) methods. In CBK8 or Extended-CBK/E [16], an ash inhibition sub model and the transition from zone II to I are described explicitly to model the extinction behavior. Backreedy et al. [15] added the effect of coal maceral types to CBK8 and defined the specific surface area as a variable parameter. In this study, CBK8 with maceral effects is optimized and validated for oxy–coal applications.

3. Model development

The CBK8 model was developed by Hurt [14,16,17] using experiments performed in a nitrogen rich environment, such as air. Therefore, this model is not directly applicable to oxy–coal combustion and the first concern is regarding the reaction order. The estimation of reaction order is uncertain in the intrinsic model. It can easily be shown that by changing the reaction order of the Arrhenius equation for intrinsic reactivity that the char reaction rate or its reactivity changes significantly. For instance, char reactivity of Thoresby (a British bituminous coal) has been predicted based on the intrinsic order of 1 and 0.68 and the results are shown in Fig. 1a and b, respectively.

During oxy–coal combustion with RFG, the presence of CO_2 and H_2O in significant quantities contributes towards the overall char oxidation process via gasification reactions and it is important to know the effect of CO_2/H_2O on char reactivity. Distinctive thermophysical and radiative properties of CO_2 compared to other gases in the combustion zone should also be considered. The effect of CO_2 and H_2O specific heat capacity (C_p) on the heat calculations of the gas mixture has to be considered. Although by updating C_p of the mixture one would not expect significant changes in burnout predictions, this modification is necessary for calculating particle temperature.

In addition, CO_2 and H_2O gasify the carbonaceous particle through highly endothermic reactions (ΔH_{C-CO2} ~170 kJ/mol and ΔH_{C-H2O} ~130 kJ/mol) and it is envisaged that the heats of reaction are important parameters required for accurate predictions. After modifying the heat of reaction in the CBK8 code, it was observed that predictions have improved by 10% where as updating C_p and the CO/CO_2 ratio has only contributed to less than 2% improvement in predictions.

CO and CO_2 are products of carbon and hydrocarbon oxidation and their ratio on the particle surface is important for the investigation of reaction of carbon with $O_2/CO_2/H_2O$ and also in determining the energy released during char combustion. In the intrinsic model, this ratio is expressed as an Arrhenius type equation which is validated for a range of coals, particle temperature and oxidant partial pressures. In this study, based on a study reported by Du et al. [18], Eq. (1) is used for the calculation of CO/CO_2 ratio:

$$\frac{[CO]}{[CO_2]} \approx \frac{r_{CO}}{r_{CO_2}} \tag{1}$$

where r_{CO} and r_{CO2} are the rates of CO and CO_2 formation.

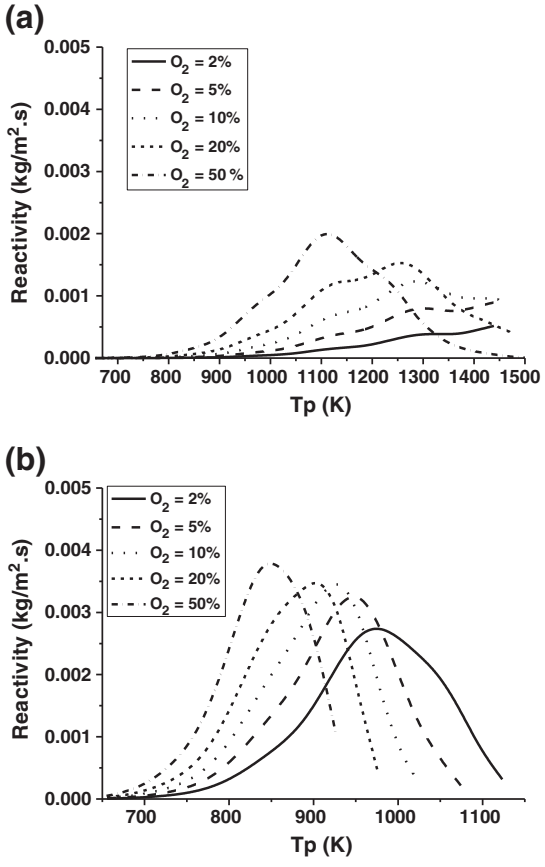

Fig. 1. Char reactivity prediction for the intrinsic reaction order of a) 0.68, and b) 1.0.