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Research article

# Development of semi-parallel reaction model of devolatilization and heterogeneous reaction for pulverized coal particles

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ABSTRACT

A semi-parallel reaction model for the devolatilization and heterogeneous reaction of coal particles during pulverized coal combustion was developed. The quasi-steady mass transfer around a single coal particle with devolatilization and the oxidation of char were analyzed to investigate the effect of the convective flow generated by devolatilization on the mass transfer of the oxidant to the particle surface at various reaction temperatures and particle diameters. The oxidation rates of char with devolatilization were lower than those without devolatilization. This tendency became pronounced with increasing reaction temperature and particle diameter. This indicated that the convective flow generated by devolatilization inhibits the mass transfer of the oxidant to the particle surface and that the influence of the devolatilization depends on the reaction temperature and particle diameter. In addition, the oxidation rates estimated by the semi-parallel reaction model were compared with those obtained from the conventional sequential reaction model and parallel reaction model. In contrast to the other models, the semi-parallel reaction model more accurately represented the decrease in char oxidation rates with increasing devolatilization rate.



## 1. Introduction

The predominant method of coal resource utilization is combustion. Pulverized coal combustion is used in coal-fired thermal power plants [1] and blast furnace operations [2]. Recently, new energy conversion systems such as gasification [3] and oxy-fuel combustion [4–6] have been developed. In general, immediately after pulverized coal particles are injected into a high temperature atmosphere, they are heated by radiative heat transfer from the furnace walls and convective heat transfer between the gas phase and particle surface. Thermal decomposition of the coal particle starts when its surface temperature reaches 700–900 K; then, a char is produced by coal pyrolysis. The release of volatile matter (VM) during pyrolysis results in an elevation of the particle temperature because of the ignition and combustion of the VM, thereby promoting the oxidation and gasification reactions of the char.

As the reaction processes taking place during pulverized coal combustion involve instantaneous phenomena which are completed within only a few hundred milliseconds, they have been studied through numerical simulations as well as experimentally [7–11]. Computational fluid dynamics (CFD) is a powerful tool for the analysis of a flow field that includes the interactions between chemical reactions and a fluid flow. In CFD analyses of pulverized coal combustion, the gas and particle phases are separated, and information about the coal particle, such as its trajectory and chemical reaction rates, can be calculated. Then, a reaction model for pulverized coal combustion must therefore be based on an actual combustion process, which will have significant impact on the flow field in the CFD analysis. On the basis of previous experiments, various reaction models have been developed and applied to numerical simulations. Hashimoto et al. proposed a tabulated devolatilization process (TDP) model to consider the effect of the heating rates of the coal particles on the devolatilization rates [12]. In this model, a database that included pre-exponential factors and activation energies of devolatilization for various heating rates was prepared and the devolatilization rates were extracted from the database. Huang et al. suggested that there are active sites for $CO_2$ and/or $H_2O$ at the char surface [13]. In response to those results, Umemoto et al. developed a gasification reaction rate equation that considered the active sites with $CO_2$ and $H_2O$ [14].

However, in previous studies, the devolatilization and char oxidation processes have been assumed to be independent. In other words, these processes have been treated as either sequential or parallel reactions in conventional simulations. Fig. 1 shows a schematic diagram of the mass transfer of the oxidant around a single coal particle. Fig. 1 (a, b) illustrates the histories of the devolatilization rate and oxidation rate of the char, assuming these reactions to be sequential or parallel.

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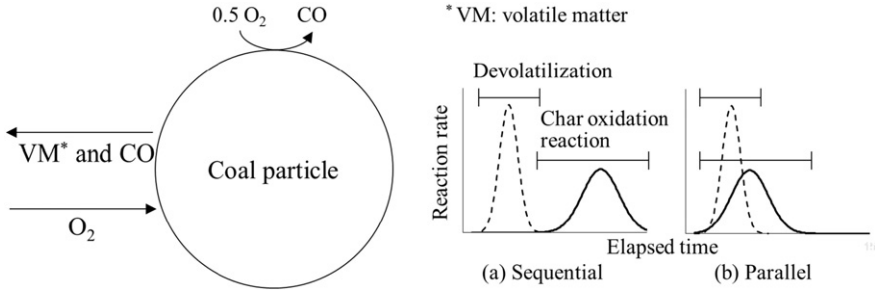


**Fig. 1.** Schematic diagram of the mass transfer of oxidant (left) and the histories of devolatilization rates and oxidation rates of char in the sequential and parallel reaction models (right).

In the sequential reaction model, char oxidation does not occur until devolatilization is complete. Assuming that the reaction processes, including devolatilization and char oxidation, are sequential, Richter et al. numerically simulated the detailed chemical reactions around a single coal particle [15]. However, a disadvantage of this reaction model is that char oxidation is completely ignored in the early reaction period, even though the char particle has been heated due to the combustion of the VM. Howard and Essenhigh reported that devolatilization and char oxidation in the initial stage progress in parallel when the particle diameter is small [16]. Asotani et al. performed numerical simulations and predicted ignition behavior assuming that devolatilization and char oxidation are parallel processes [17]. In the parallel reaction model, devolatilization and char oxidation are assumed to occur simultaneously but to be independent of each other. Howard and Essenhigh also indicated that convective flow caused by the release of VM inhibits the mass transfer of the oxidant to the particle surface and that the concentration of oxidant at the particle surface is close to zero when the char particle is over 65 μm in diameter [18].

When the temperature at which devolatilization occurs is relatively high or char oxidation occurs at low temperature, devolatilization will influence the oxidation of the char, as shown in Fig. 2. In other words, the reactions are defined as "semi-parallel" reactions, in which the oxidation rate of the char decreases with the convective flow caused by devolatilization and increases with a decrease in the devolatilization rate. Unfortunately, in almost all the previous CFD studies of pulverized coal combustion, the assumptions about the relationship between the devolatilization process and char oxidation were not described. Specifically, it is unclear whether the assumed relationship between these reactions is sequential or parallel. Of course, the interaction between devolatilization and char oxidation is ignored in both cases. Even in ANSYS Fluent®, one of the most popular commercial CFD software applications, char oxidation begins after the VM is completely evolved [19]. Accordingly, the assumption of the relationship between devolatilization and char oxidation processes in CFD has not yet been established.

To understand basic coal combustion phenomena, a one-dimensional approach employing a single coal particle is useful. Many researchers, using this approach, have considered detailed chemical reactions in the gas phase or at the particle surface [20–24]. However, most detailed simulations of a single coal particle are too difficult and overly complicated to apply to CFD, due to the high computational costs and limits of grid resolution. Therefore, the assumptions should be carefully determined to prevent inconsistencies between the one-dimensional simulation and the CFD for pulverized coal combustion.

In the present study, the relationship between devolatilization and the heterogeneous reaction of char was investigated, and a heterogeneous reaction model capable of considering the effect of devolatilization on the mass transfer of the oxidant is proposed for the CFD analysis. To investigate the effect of the devolatilization process on the mass transfer of the oxidant, the quasi-steady mass transfer around a single coal particle was numerically analyzed under the condition that devolatilization and char oxidation occur in parallel. Then, a parameter study was performed for various reaction temperatures and particle diameters, and the effect of the devolatilization process on the mass transfer of the oxidant to the particle surface was quantified. In addition, a semi-parallel reaction model of the devolatilization and heterogeneous reaction of char was developed by fitting a simple equation to the results of the parameter study. Finally, the difference in the oxidation rates between conventional reaction models and the semi-parallel reaction model was evaluated, and the implementation of this reaction model in the CFD of pulverized coal combustion was discussed.

## 2. Review of the restrictions for calculating coal reactions in CFD

Fig. 3 shows an image of the target system in this study, i.e., the phenomena occurring in the computational grid of the CFD simulation. In the CFD analysis of pulverized coal combustion, development of the semi-parallel reaction model is restricted in two ways:

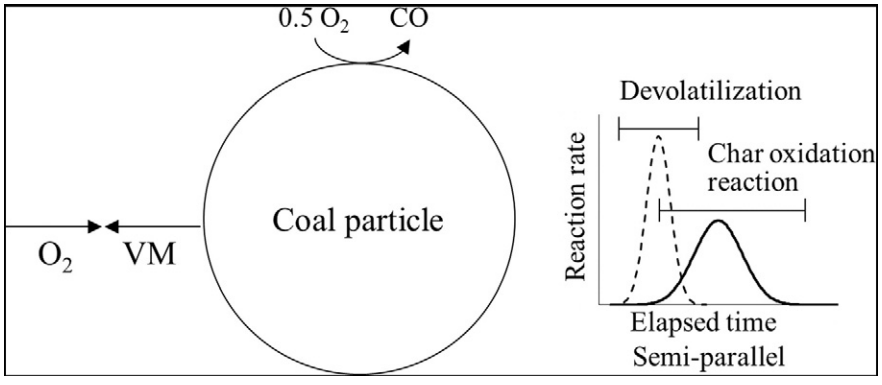


**Fig. 2.** Schematic diagram of the mass transfer of oxidant (left) and the histories of devolatilization rates and oxidation rates of the char in the semi-parallel reaction model (right).