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Simulation of pyrolysis in low rank coal particle by using DAEM kinetics model: Reaction behavior and heat transfer

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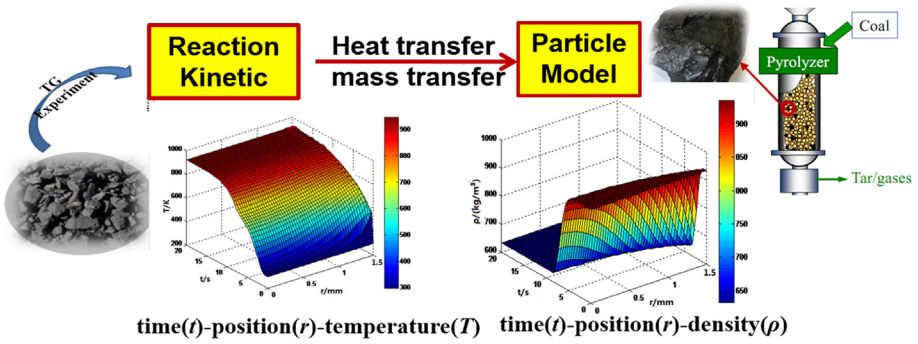
HIGHLIGHTS

- Kinetic parameters of coal pyrolysis were obtained by DAEM via TG/DTG experiments.
- E_0 = 186.5 kJ/mol, $k_0 = 3.96 \times 10^{10}$ s^{-1} and σ = 39.5 kJ/mol were obtained.
- A coal particle pyrolysis model coupling with reaction and heat transfer was proposed.
- Mass fraction and temperature profiles inside the coal particle was well predicted.
- $\Delta T > 300$ K from surface to core of a 3 mm particle was predicted at 900 °C pyrolysis.

GRAPHICAL ABSTRACT



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ABSTRACT

A comprehensive and systematic study on the fundamental pyrolysis behaviors of a single coal particle was performed in this study. The pyrolysis characteristics of coal was investigated by non-isothermal thermo gravimetric analysis whereas the reaction kinetic parameters were obtained by using the distribute activation energy model (DAEM). As three heating rate profiles were applied (10, 20 and 30 °C/min) in TG/DTG experiments with a final pyrolysis temperature of 900 °C, the obtained kinetic parameters, i.e., activation energy (E_0), pre-exponential factor (k_0) and standard deviation (σ) were 186.5 kJ/mol, 3.96×10^{10} s^{-1} and 39.5 kJ/mol, respectively. When these calculated kinetic parameters were used to predict devolatilization curves, the simulation results were in well agreement with the experimental data. As such, a one-dimensional, time-dependent particle pyrolysis model was proposed to characterize the detailed chemical and physical phenomena occurred within a pyrolyzing coal particle. It is found that this model successfully predicted the mass fraction residue and temperature profiles inside the coal particle. In addition, the effect of particle size on pyrolysis performance was also investigated through simulation. It is expected that such a model can be integrated with CFD simulation to provide useful insight for the design of a practical coal pyrolysis reactor.

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Nomenclature

D	particle diameter, mm
E	the activation energy, kJ mol^{-1}
E_0	the mean activation energies, kJ mol^{-1}
f(E)	the activation energy distribution
h_c	the convective heat-transfer coefficient, W m^{-2} K^{-1}
k_0	the pre-exponential factor, s^{-1}
R	the universal gas constant, J mol^{-1} K^{-1}
R_v	the instantaneous volatile evolution rates, kg m^{-3} s^{-1}
T	temperature, °C
T_0	initial temperature of coal particle, °C
T_s	temperature at particle surface, °C
T_f	reactor temperature, °C
V	volatile loss
V^*	the final fractional volatile loss

Greek letters

ΔH	specific reaction heat of pyrolysis, kJ kg^{-1}
α	the extent of conversion
β	the heating rate, K s^{-1}
ε_r	emissivity
λ_s	effective thermal conductivity of coal, W m^{-1} K^{-1}
ρ	coal bulk density, kg m^{-3}
ρ_0	initial coal bulk density, kg m^{-3}
ρC_p	the volumetric specific heat capacity, J m^{-3} K^{-1}
σ	Stefan-Boltzman constant, W m^2 K^{-4}
σ_E	the standard deviations, kJ mol^{-1}

1. Introduction

Coal is the mainstay of energy in China and this situation will remain unchanged in foreseeable future. As we all known, pyrolysis is one of the most promising technologies for clean and effective utilization of low-rank coal, and it is also the first step in most of the existing multi-generation technologies [1–3]. However, most of coal pyrolysis processes need pulverized coal, which suffers from many disadvantages in their industrialized application, such as complication and high-cost of coal pretreatment, difficult separation of tar from pulverized coal, and easy blockage of the pipe by the mixture of tar and ash. Hence, using medium-sized coal particles in the industrial reactor is quite necessary.

Since coal is a particularly complicated mixture, how to simulate the complex coal pyrolysis has been studied in the last decades. To date, the empirical models and the network models are mainly applied. Those early empirical models, such as the first-order reaction model [4], the multiple-step reaction model [5], and the two-competing-rate model [6] are too simple to be applied widely. Recently, the physicochemical structure of coal has been well understood so that more fundamental approaches have been used to simulate the coal pyrolysis behavior. For instances, the network models, including the functional group devolatilization vaporization cross-linking model (FG-DVC) [7], the distributed-energy chain statistics (Flash chain) model [8,9], and the chemical percolation devolatilization (CPD) [10,11] were proposed and applied. However, some disadvantages, such as the accuracy of model input parameters in terms of the coal structure and the complexity of the model that limiting their application in combining with the simulation of reactor are still remained. Recently, Bartocci et al. [12] proposed a four-parallel-reaction scheme for a pellet made of 90% sawdust and 10% glycerol. For each pseudo-component (cellulose, hemicellulose, lignin and glycerol), the first order reaction was used, in which the kinetic parameters for each component were derived using a model fitting approach. It is found that the modeling results by using this method well matched over the entire range of experimental data. Pitt [13] had ever proposed a distributed activation energy model (DAEM), in which the pyrolysis of coal was assumed to have a large number of irreversible independent and parallel first-order reactions with different activation energies. This model truly and effectively depicted the main features of chemical behaviors observed in coal pyrolysis. Miura and Maki [14] further developed a simpler and more accurate method to estimate f(E) and k_0(E) in the DAEM, in which only three sets of thermogravimetric analysis (TG)/derivative thermogravimetric analysis (DTG) experimental data obtained at different heating rates were needed. This model was proven to be able to describe the pyrolysis behavior over a wider range of operating conditions and has been successfully applied to study the pyrolysis of complex matters, such as biomass [15,16], coal [17,18], solid waste [19,20] and the synergistic effect of coal and biomass [21]. However, sometimes, the results obtained by isoconversional methods including Miura-Maki method cannot well match with the experimental data over the entire range when compared with those obtained by the model-fit method [22]. In previous work, the definitions of k_0, f(E) and the reaction order in the model-fit method were different. For the f(E) in the DAEM, various distribution functions including Dirac delta distribution (for SRMs), logistic distribution [23] and Gaussian distribution [24] were used. Miura used the Gaussian distributed function for DAEM and found that the assumption was reasonable for 19 type coals [25]. In the optimization schemes, various researchers extracted the kinetic parameters using different methods, such as direct search method [24], simulated annealing method [26], pattern search (PS) method [27], multistart algorithm method [28] and differential evolution algorithm method [29]. Among them, PS method was claimed to be better than others [27]. In the process of fitting, due to the compensation effect [30,31], multiple values of k_0 and E_0 fitted the experimental data well. Thus, the suitable initial values for the model-fit analysis are very important. However, most of studies either pre-fixed the pre-exponential factor (randomly or from the reported values in literature) or maintained it as the optimization parameter based certain assumptions on the distribution of activation energy [32,33]. Therefore, it is necessary to develop a suitable way to choose the appropriate initial values for model-fit method. In addition, the kinetic models mentioned above can be only used when the coal particle is very small and the temperature inside it can be assumed as isothermal. In fact, the pyrolysis behavior of large coal particle is very different from that of the pulverized coal because the intraparticle mass and heat transfer noticeably affect the coal pyrolysis progress which is very important for the design of reactor with better performance. Furthermore, different coals have different structures and compositions, resulting in different pyrolysis characteristics. Besides, even for the same coal, the pyrolysis processes are also different in different reactors due to the variation in heat transfer coefficient. As such, the simulation of kinetics of a given coal particle in a specific reactor is very necessary, which is also the foundation of industrial application of the reactor. However, most of studies on the simulation of reactor used only the intrinsic kinetics, basically used the CFD simulation with the single-step global model [34–36]. In the coal topping process, pyrolysis of millimeter-sized coal particles should be much