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Modeling of gas/particle flow in coal conversion with a drop tube reactor using a lumped kinetic model accounting volatiles–char interaction

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ABSTRACT

Coal conversion including reforming of nascent tar over the char surface in a drop tube reactor (DTR) was studied both experimentally and numerically. Victorian brown coal and char prepared from the same coal were co-fed into an atmospheric DTR. The effects of reaction temperature (973–1173 K), solid hold-up (8.31×10^{-6}–2.50×10^{-4}), residence time (0–4.6 s for gas; 0–0.78 s for solid particles), and steam partial pressure (0–0.05 MPa) on the conversion characteristics were investigated. A 4-lump kinetic model consisting of tar, gases, char, and soot with 6 global reactions was developed based on the experimental results. The lumped kinetic model was integrated with a computational fluid dynamics (CFD) simulation using an Eulerian–Eulerian approach for mixed phase flow to simulate the coal conversion experiments in the DTR. The CFD results for product distribution during coal conversion in the DTR showed reasonable agreement with the experimental results. The CFD approach presented is suitable for use in designing and optimizing a pyrolyzer for a triple-bed combined circulating fluidized-bed coal gasifier, consisting of a downer (pyrolyzer), a bubbling fluidized bed (gasifier), and a riser (combustor).

1. Introduction

Gasification is an efficient and environmentally attractive process for coal utilization [1,2]. A major challenge in coal gasification is dealing with the refractory aromatic compounds that constitute the tar formed during gasification. This problem is especially severe in low temperature coal gasification processes, which are designed to have improved cold gas efficiency (CGE) [3]. Tar is an undesirable product of gasification because of various problems associated with condensation, formation of tar aerosols, and polymerization to form more complex structures, which can impede and damage the process equipment, as well as the engines and turbines used in product gas consumption [4]. Also, other volatile materials generated during coal gasification, such as light hydrocarbon gases and hydrogen may strongly inhibit char gasification [5–7]. Therefore, the development of technologies and methods for the reduction or removal of tar has received substantial attention in recent years [4,8,9]. Current liquid scrubbing techniques and catalytic reforming methods suffer from undesirable overall thermal efficiency loss and/or catalyst deactivation because of the deposition of chlorine/sulfur compounds and carbon [4,8,9]. Thus, it is necessary to develop a tar reforming system with high thermal efficiency and without catalyst poisoning or deactivation.

Recently, a triple-bed combined circulating fluidized-bed reactor system (TBCFB), consisting of a downer (pyrolyzer), a bubbling fluidized bed (BFB, gasifier), and a riser (combustor), was proposed [10–13]. This reactor system was designed to decompose tar through extensive interaction between the volatiles and char in the downer [10–13]. In this system, tar is decomposed inside the downer, which reduces the thermal efficiency loss associated with liquid scrubbing, and char is employed as a catalyst and/or promoter for tar decomposition. If the char is deactivated by interaction with the volatiles [1,14], it can be burned in the combustor to supply the heat required for the process.

In a recent study, we demonstrated the potential of the downer for decomposing tar in the TBCFB reactor system [15]. In that study, a drop-tube reactor (DTR) was used to investigate tar reforming over the char surface under reaction conditions similar to those expected for a downer reactor, wherein coal, char particles, and gas flow co-currently. It was demonstrated that nascent tar from the pyrolysis of Loy Yang (LY) brown coal was significantly decomposed by the volatiles–char interaction. However, that study concentrated on the laboratory-based

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experimental investigation without considering the establishment of a mathematic model based on the experimental results for coal conversion in the DTR.

Coal conversion incorporating the volatiles–char interaction proceeds *via* a number of complex parallel reactions, and produces a wide range of products, such as char, soot, light hydrocarbons, and both single and polycyclic aromatic hydrocarbons. An approach in which complex mixtures with large numbers of compounds are lumped into smaller groups of pseudo components has been widely employed to provide a tractable number of kinetic equations and estimate kinetic parameters [16–18]. However, lumped component models have, to date, been mainly applied to hydrogenation or catalytic cracking of heavy oil and other residues [19–21].

Computational fluid dynamics (CFD) has been employed as a powerful tool for obtaining a deeper understanding of the gas–solid mixed flows employed in downer reactors [22–24]. Ropelato et al. [22] proposed an Eulerian–Eulerian approach for predicting fluid dynamics in the downer reactor with good agreement between experimental data and model predictions. The published works [10,22–24] include numerical simulation of gas–solid flow in downer reactor for coal conversion without considering chemical reactions. Some works [25,26] include numerical simulations of gas–solid flow combined with chemical reactions only in the riser-type fluid catalytic cracking (FCC) reactors. However, little reports are available on the CFD simulation for gas–solid flow combined with chemical kinetic model for coal conversion accounting the volatile–char interaction in the downer reactor.

Therefore, in this study, we have attempted to develop a CFD model coupled with a lumping kinetic model for coal conversion that accounts for the volatiles–char interaction. The effects of residence time, reaction temperature, solid hold-up, and steam partial pressure on the conversion characteristics of coal in the DTR are investigated. The kinetic parameters are estimated under different operating conditions, and the correlation between the kinetic parameters and operating conditions is established based on the experimental results. A practical combined CFD and lumped kinetic model is likely to be an effective tool for facilitating to develop coal gasification in a TBCFB.

2. Experimental

2.1. Preparation of coal and char samples

Dried Victorian Loy Yang brown coal (LY) with particle sizes ranging from 0.50 to 0.71 mm was used. Gasified LY Char (GLYC) sample was prepared by gasifying LY at 900 °C for 20 min in a steam/N_2 flow using a fluidized-bed reactor. The details of this preparation are reported elsewhere [15]. The elemental compositions of LY, and GLYC are listed in the Supplementary Information. All samples used in this study were prepared by blending the LY and GLYC at a fixed ratio (LY:GLYC = 3:7 on a carbon-mole basis).

2.2. Drop-tube reactor coal conversion experiments

In this study, a quartz tube of 2.550 m length with an isothermal zone of 2.100 m length and an inner diameter (i.d.) of 8 or 15 mm was heated by eight independently controlled electric furnaces. A nitrogen/steam gas mix preheated to 250 °C was supplied to the DTR, and the steam partial pressure was varied over the range of 0–0.05 MPa. The gas flow rate was carefully adjusted to maintain a constant gas residence time in the isothermal zone under the different experimental conditions. Detailed descriptions of the experimental setup and procedure are given elsewhere [15,27,28]. The experimental conditions used to investigate the effects of residence time, reaction temperature, steam partial pressure, and solid hold-up on the conversion characteristics of the coal samples in the DTR are provided in Table 1.

The variation of residence time was achieved by heating only a section of the quartz tube. In the cases of the quarter-length and half-length experiment, hereafter referred to as QL and HL, respectively, only the bottom two and four furnaces were heated to the desired temperature. All other experiments were carried out with the full length of the tube being heated, hereafter denoted as FL. In the FL case, the residence time of gas in the isothermal zone was about 4.6 s, and that of coal particles was around 0.78 s. These values were estimated from the length of the isothermal zone and the terminal velocity of the particles [29]. Consequently, the residence times of the gas and particles were approximately halved in the HL experiments, although the length of the isothermal zone in the HL experiments was not exactly half that used in the FL experiments. The QL experiments were conducted in an analogous fashion.

Table 1
Experimental conditions and product yields on carbon mole basis.

Run no.	1	2	3	4	5	6	7	8	9
Temperature (°C)	700	800	900	900	900	900	900	900	900
Residence time (s)	FL	FL	FL	QL	HL	FL	FL	FL	FL
Feeding rate (g/min)	0.5	0.5	0.5	0.5	0.5	0.176[a]	0.5	1.5	0.5
Steam partial pressure (MPa)	0.05	0.05	0.05	0.05	0.05	0	0	0	0.03
Product yield, mol-C/100-mol-C-(LY + GLYC)									
Tar/lump 1	1.5	1.1	1.1	1.9	1.2	1.4	1.2	0.9	1.0
Gases/lump 2	4.9	6.8	7.9	5.0	7.3	6.7	6.7	6.2	7.7
Char/lump 3	92.3	90.6	90.1	91.8	90.3	88.9	89.7	91.3	90.0
Soot/lump 4	1.3	1.5	1.0	1.3	1.1	3.1	2.5	1.7	1.3

[a] A quartz tube with internal diameter of 15 mm was used.

The solid hold-up, defined as the ratio of total volume of solid particles to that of the isothermal zone of the DTR, was changed by varying the feeding rate and the internal diameter of the quartz tube. In the experiment where the solid was fed at 0.176 g/min, a quartz tube with an i.d. of 15 mm, instead of the 8 mm i.d. tube used in the other experiments, was employed. Feeding rates of 0.176, 0.5, and 1.5 g/min correspond to solid hold-up values of 8.31×10^{-6}, 8.31×10^{-5} and 2.50×10^{-4}, respectively. Thus, the solid hold-up is investigated at 1, 10, and 30 orders of magnitude in these experiments. The solid holdup can be one of the important factors influencing the effective heating rate of the particles and the effective residence time of the volatiles. The former can affect the primary pyrolysis characteristics, whereas the latter can affect the chemistry and kinetics of the subsequent secondary reactions. Therefore there are possibilities of further improvements in the modeling approach by taking other effects given by the solid holdup into account.

2.3. Pyrolysis in two-stage tubular reactor

A rapid pyrolysis of LY at 650 °C, high enough to complete pyrolysis in terms of tar evolution, was performed in a two-stage tubular reactor (TS-TR) to obtain information on the composition of primary pyrolysis products. Details of the TS-TR experiment are presented elsewhere [30,31]. The primary purpose of the TS-TR was to minimize the interaction between char and volatiles, thus preserving the properties of the nascent volatiles. These volatiles, such as CO, CO_2, and light C_1–C_4 hydrocarbons, were analyzed by a gas chromatograph, which was connected in series to the TS-TR. This carbon-containing gas mixture is hereon referred to as 'carbon gases'. The char yield was obtained by weighing the residue recovered from the tube. The tar yield was determined by difference.

3. Kinetic modeling

3.1. Lumping concept

A number of complex reactions occur during *in situ* tar reforming over char in the DTR, and the products consist of a number of compounds resulting from concurrent pyrolysis, gas phase thermal cracking,

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