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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).

Keywords: Lumped kinetic model; Tar decomposition; Steam gasification; Volatiles char interaction; CFD modeling

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

1 heat required for the process.

2 In a recent study, we demonstrated the potential of the downer for decomposing
3 tar in the TBCFB reactor system [15]. In that study, a drop-tube reactor (DTR) was
4 used to investigate tar reforming over the char surface under reaction conditions
5 similar to those expected for a downer reactor, wherein coal, char particles, and gas
6 flow co-currently. It was demonstrated that nascent tar from the pyrolysis of Loy Yang
7 (LY) brown coal was significantly decomposed by the volatiles-char interaction.
8 However, that study concentrated on the laboratory-based experimental investigation
9 without considering the establishment of a mathematic model based on the
10 experimental results for coal conversion in the DTR.

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

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

30 Therefore, in this study, we have attempted to develop a CFD model coupled with
31 a lumping kinetic model for coal conversion that accounts for the volatiles-char
32 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/N₂ 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.

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, CO₂, and light C₁-C₄ 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, steam reformation, and char gasification. The lumping model approach is widely employed traditionally to describe such complex mixtures by lumping large numbers of chemical compounds into smaller groups of pseudo-components in order to estimate kinetic parameters for the numerous and complex reactions, for example, the catalytic cracking of petroleum fractions [25,26].

Previous studies [32,33] found that the evolution of tar, in terms of primary pyrolysis, is completed during heating before the coal particle temperature reaches at 600 °C, even at a heating rate of 1000 °C/s. Moreover, an important purpose of the DTR is to provide a very high heating rate to the sample. So, under our conditions, it is assumed that the coal sample has already been converted into tar, carbon gases, char, and soot by primary pyrolysis at the top of the DTR, where the temperature is 500–900 °C, before entering the isothermal zone, where the temperature is 900 °C. Therefore, in our model the feedstock is lumped into tar, carbon gases, char, and soot derived from the primary pyrolysis. Carbon oxides gases and hydrocarbon gases were lumped into one pseudo-compound as carbon gases for simplification of the kinetic model, although the carbon oxides gases and hydrocarbon gases have different roles in reactions. A reaction network among lumps in the DTR is proposed as shown in Fig. 1. The possible main reactions in the DTR were considered to be thermal cracking/steam reforming of volatiles, gasification, and the volatiles-char interaction.

The results obtained from the TS-TR experiment were used to estimate the initial amount of tar, carbon gases, char, and soot at the DTR inlet. It should be noted that there is a possibility that the product yields from TS-TR are different from the ones in DTR as the solid holdup (feeding rate) can affect heating rate of particles and the local volatiles concentrations around particles, and thus induce some changes in primary pyrolysis characteristics. The amounts of these species were derived from the primary pyrolysis of LY combined with the amount of co-fed GLYC normalized on a carbon-mole basis. These normalized amounts of tar, carbon gases, char, and soot were defined as the initial amount of each lump fed into the isothermal zone of the DTR. The initial amounts of tar, carbon gases, and char were 7.4, 3.9, and 88.7

mol-C/100-mol-C-(LY+GLYC), respectively. The yield of the products will be indicated in the unit of mol-C per 100-mol-C of the sample (or mol-C/100-mol-C-(LY+GLYC)).

3.2. Reaction network

A mathematic model for the proposed lumped model is established and expressed as;

$$\frac{dC_1}{dt} = -(k_{12} + k_{13} + k_{14})C_1 + k_{21}C_2 \quad (1)$$

$$\frac{dC_2}{dt} = -k_{12}C_2 + k_{12}C_1 + k_{32}C_3 + k_{42}C_4 \quad (2)$$

$$\frac{dC_3}{dt} = -k_{32}C_3 + k_{13}C_1 \quad (3)$$

$$\frac{dC_4}{dt} = -k_{42}C_4 + k_{14}C_1 \quad (4)$$

where C_i represents the concentration of each lump ($i = 1-4$); $k_{i,j}$ refers to the apparent rate constant for the reaction of lump i to lump j ; and t is the residence time. The numerical solutions for these differential equations (1)–(4) were obtained using a fourth-order Runge–Kutta method. The set of apparent rate constants $k_{i,j}$ in the proposed lump model was estimated so that the sum of squared residuals between the predicted values and the corresponding experimental values were minimized.

4. CFD simulation

4.1. Mathematical model

The presented CFD simulation includes a kinetic model for chemical reactions included in coal conversion in the DTR of which parameters were all estimated empirically based on the experimental results by numerical fitting. The empirically justified kinetic model was then combined with the CFD model to visualize chemically reacting gas-solid flow in the laboratory scale DTR. This approach should have a potential to be extended to predict/estimate the gas-solid two-phase reacting flow of larger scale downer reactor simply by changing the boundary conditions since the theoretical basis developed in this work will be principally valid.

The multiphase Eulerian-Eulerian approach is widely used to predict the fluid dynamics in downers [22,34]. In this study, an Eulerian-Eulerian model was applied

to simulate the fluid dynamics in the DTR. Eulerian-Eulerian model is a powerful model can all handle this type of problem correctly, even in the very low loading of the particles [35]. In this two-fluid model, the particles are treated as a continuum, similar to the gas phase. Thus, there are two interpenetrating phases, *i.e.* gas and solid, where each phase is characterized by its own conservation of motion equations. A summary of the governing equations and constitutive relationships is provided in the Supplementary Information.

For this gas-solid mixed flow in the DTR, a laminar model was employed for the gas phase. Under typical conditions, the Reynolds number $Re = DU_g \rho_g / \mu_g$, where D is the diameter of the DTR (8 mm), U_g is the superficial gas velocity (3 m/s), ρ_g and μ_g are the density and viscosity of the gas (0.22 kg/m³ and 4.40×10⁻⁵ Pa·s at 900 °C, respectively), was about 124, meaning that the flow is laminar.

4.2. Simulation layout

An axial illustration of the isothermal zone of the DTR is provided in the Supplementary Information. The LY and the char were fed into the isothermal zone with steam and N₂. The elemental composition of the char was determined by elemental analysis, and the average molecular mass of the carbon gases was calculated based on the results of the GC measurement. Soot was assumed to be pure carbon whereas element composition of solid char was assumed based on reported literatures [36–38]. In addition, molecular weight of tar was not easy to determine and, in this study, was also assumed from the literature [39]. The compositional formulae of the four lumps were found to be $C_{1.12}H_{1.83}O_{0.86}$ (carbon gases), $C_{38.57}H_{56.80}O_{17.52}$ (tar), $CH_{0.07}O_{0.04}$ (char), and C (soot).

The DTR is an axisymmetric cylinder. In order to save iteration time and improve convergence efficiency, a simplified geometry for the simulation was used, as illustrated in the Supplementary Information. There is a gas phase and a solid phase in our experiments. The gas phase is comprised of carbon gases, tar, steam, H₂ and N₂. The solid phase is comprised of char and soot. Six global chemical reactions were considered in the simulation. Two homogeneous reactions (5–6) and four heterogeneous reactions (7–10) were considered according to the developed lumped kinetic model, as follows:

The boundary conditions for the numerical simulation are shown in the Supplementary Information. In addition, the convergence criteria are set in a transient fashion with a time step of 0.001 s and residuals for continuity less than 10^{-4} , energy less than 10^{-6} , and all species less than 10^{-6} . Typically, 350,000 iterations were required. The ANSYS FLUENT 14.0 (ANSYS Inc.; USA) was used for the CFD computations.

5. Results and Discussion

5.1. Effect of residence time

Fig. 2 shows the variation of product yields with residence time and their calculated results by the four-lump model. The residence times for the experimental values in the Fig. 2 were derived based on some conventional methods [40]. The methods to obtain them are described in the Supplementary Information. We found that the tar yield decreases sharply within very short residence time, while the yields of char, gases, and soot increase significantly. According to the calculated results, the sharp variation for each lump is completed at around 0.3 s, and then the variation levels off.

Initially, within a very short time, extensive tar deposition likely occurs over the char surface due to volatile-char interaction [1], which is induced by coexistence of high concentrations of tar and nascent char providing active sites for tar decomposition. It is believed that the decomposition takes place in micro- and nanopores of char, depositing carbon onto the pore surfaces [1]. After 0.3 s, the decomposition rate of tar slows down presumably owing to the reduction of the amount of active sites that is induced by carbon deposition, though the chemical nature of the active sites is undefined yet. Char gasification becomes dominant in downstream and contributes further increase in the yield of carbon gases.

5.2. Effect of temperature

The product distribution from coal conversion during *in situ* tar reforming in the DTR at different temperatures is shown in Table 1 (runs 1–3). The tar, char, and soot yields decrease with increasing temperature, while the carbon gases yield increases. Similar results and possible reasons were reported in our previous paper [15].

In this work, we found that the reaction rate for *in situ* tar reforming in the DTR is strongly related to reaction temperature. The apparent rate constants of the four-lump model are estimated based on these experimental data and given in Table 2.

According to the Arrhenius equation, the apparent rate constant can be formulated as a function of the reaction temperature as below:

$$k = Ae^{-E_a/RT} \quad (12)$$

where A represents the pre-exponential factor of each reaction lump and E_a refers to the apparent activation energy. The pre-exponential factors (A) and the apparent activation energies (E_a) are shown in Table 3.

5.3. Effect of solid hold-up

The effects of the feeding rate on the yield of each lump were investigated (runs 6–8 in Table 1). The change in the solid hold-up greatly influences the product yields. The tar and soot yields are reduced whereas the char yield is significantly increased.

As shown in Fig. 3, the apparent rate constant of the reaction between tar and char (k_{13}) shows significant dependence on the solid hold-up, whereas other apparent rate constants do not. This indicates that high solid hold-up enhances the tar and char interaction. Decomposition of tar occurs on the active sites of pore surface of the char particles. The increase of the local concentration of char particles with increasing solid holdup provides more surface and thus increases the amount of active sites, contributing extensive decomposition of tar. Based on our lumped kinetic model proposed in this study, the obvious effect of the solid holdup on the kinetics is appeared only at the conversion of tar into char (k_{13}) as shown in Fig 3. In addition, it should be noted that the intensive volatiles-char interaction also results in changes in char structure [41] and enhancement of volatilization of inorganic matters [14] in char matrix.

5.4. Effect of steam partial pressure

The effects of steam partial pressure on the yield of each product were

investigated in runs 7, 9 and 3 as shown in Table 1. The results indicate that the steam partial pressure slightly affects the product yield under the conditions used. However, it is apparent that addition of steam suppresses soot formation, which is consistent with results reported previously [42]. A slight increase in gas yield may be attributed to the improvement of gasification under higher steam partial pressure. A considerable amount of H_2O (12 moles per 100 moles carbon in the coal) is produced during the pyrolysis of coal [42,43]. Therefore, it is likely that this amount of H_2O is sufficient to supply the steam required in these reactions under current experimental conditions, and the addition of extra steam has little influence on the *in situ* tar reforming process. As shown in Fig. 4, the apparent rate constants k_{14} (Tar→Soot) and k_{42} (Soot→Gas) vary with the steam partial pressure whereas the other apparent rate constants seem independent of it.

5.5. CFD with the Eulerian-Eulerian approach coupled with lumped kinetic model.

5.5.1. Mass fraction profiles of different lumps in the DTR

A CFD simulation coupled with the developed lumped kinetic model (Reaction equations from (5) to (10)) was conducted. The apparent rate constants such as k_{12} , k_{21} , k_{13} , k_{14} , k_{32} , and k_{42} that were empirically determined through the comparison with the experimental results, were used for reactions (5), (6), (7), (8), (9), and (10), respectively. Fig. 5 illustrates the mass fraction distribution of the four lumps in the DTR (in run 4 in Table 1). A QL drop tube reactor was employed in run 4. As can be seen in the figure, the mass fraction of each lump shows a small change because the gas-solid mixed flow in the DTR is laminar. In the carbon conversion process, reforming and cracking of tar over the char surface occurred, and thus, the mass fraction of tar decreases gradually from inlet to outlet. Char, as a promoter of decomposition of tar during the *in situ* tar reforming process [1], undergoes gasification and coking in the steam atmosphere and its mass fraction also decreases, whereas the mass fraction of carbon gases shows a rapid increase along the drop tube reactor. From the kinetic model, carbon gases can be produced by tar cracking and gasification of char and soot, but is consumed only by pyrolysis to yield tar.

The effect of reaction temperature, solid hold-up, and steam partial pressure on tar consumption along the drop tube reactor is shown in Fig. 6. In Fig. 6b, the value of

tar decomposition is used as vertical axis instead of tar concentration along the drop tube reactor, because the feeding rates of different cases are different and it causes that the initial values of tar concentration at different cases are not the same when compared. As can be seen in the figure, most of the tar (about 85%) is consumed rapidly near the entrance zone of the DTR. The higher the reaction temperature is, the more rapidly the tar is consumed (Fig. 6a), and the higher the solid hold-up is, the stronger the volatiles-char interaction is (Fig. 6b). However, the steam partial pressure has little effect on tar consumption (Fig. 6c).

5.5.2. Comparisons between simulation results and experiments

Fig. 7 shows a comparison of the CFD simulation results and experimental data under various conditions, and confirms that there is good agreement between the two. This means that the proposed CFD model gives acceptable prediction of product yields for carbon conversion during the *in situ* tar reforming process, and that the developed model adequately represents the processes that constitute coal conversion in the DTR.

6. Conclusions

Loy Yang brown coal was converted in a DTR under conditions similar to those typically used in a downer reactor, where coal, char particles, and gas flow co-currently. The effects of residence time, reaction temperature, solid hold-up, and steam partial pressure on the conversion characteristics were studied. The temperature showed a significant influence on the rate of every process, and the product yields varied significantly with temperature. The solid hold-up mainly affected the tar-char interaction and tar was decomposed more extensively with increasing solid hold-up. The steam partial pressure only slightly affected the yields of tar, char, and carbon gases, but suppressed soot formation. A kinetic model with four lumps (gases, tar, char, and soot) and six reactions constituting volatiles-char interaction was developed and successfully integrated with a CFD simulation to simulate gas/particle flow in the DTR.

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Table Captions:

- Table 1. Experimental conditions and product yields on carbon mole basis.
- Table 2. Result of estimated apparent rate constants at different temperatures.
- Table 3. The apparent activation energy (E_a) and pre-exponential factor (A) for each apparent rate constant.

Figure Captions:

- Figure 1. Schematic diagram of the four-lump reaction network in the DTR coal conversion process accounting for the volatiles-char interaction.
- Figure 2. Experiment yields and calculated yields vs. residence time.
- Figure 3. The correlation between the estimated apparent rate constant and solid hold-up.
- Figure 4. The correlation between the estimated apparent rate constant and steam partial pressure.
- Figure 5. The contours of different lumps derived from the CFD simulation: (a) carbon gas, (b) tar, (c) char, and (d) soot.
- Figure 6. The effect of reaction conditions (a. Temperature; b. Solid hold-up; c. Steam partial pressure) on tar consumption along the distance from the entrance of the DTR.
- Figure 7. Experimental yields vs. predicted yields for all experimental conditions used in this study.

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.

Table 2 Result of estimated apparent rate constants at different temperatures.

Apparent rate constant (s ⁻¹)	Temperature (°C)		
	700	800	900
K ₁₂ : Tar→Gases	0.30	0.90	2.60
K ₂₁ : Gases→Tar	0.30	0.73	1.50
K ₁₃ : Tar→Char	1.00	2.30	4.60
K ₁₄ : Tar→Soot	0.20	0.90	1.20
K ₃₂ : Char→Gases	0.013	0.040	0.078
K ₄₂ : Soot→Gases	0.10	0.30	0.95

Table 3 The apparent activation energy (E_a) and pre-exponential factor (A) for each apparent rate constant.

Apparent rate constant (s^{-1})	Apparent activation energy (kJ/mol)	Pre-exponential factor (s^{-1})
k_{12} : Tar→Gases	102.2	9.0E+04
k_{21} : Gases→Tar	76.4	3.8E+03
k_{13} : Tar→Char	72.4	7.7E+03
k_{14} : Tar→Soot	86.5	1.0E+04
k_{32} : Char→Gases	86.6	6.0E+02
k_{42} : Soot→Gases	106.4	5.0E+04

Fig. 1. Schematic diagram of the four-lump reaction network in the DTR coal conversion process accounting for the volatiles-char interaction.

Fig. 2. Experiment yields and calculated yields vs. residence time. (at reaction temperature: 900 °C; steam partial pressure: 0.05 MPa; feeding rate: 0.5 g/min); (a) Char, (b) Tar and gases, (c) Soot. Symbols and solid lines represent the experimental results and model calculations, respectively.

Fig. 3. The correlation between the estimated apparent rate constant and solid hold-up (at reaction temperature: 900 °C; residence time: 4.6 s for gas, 0.78 s for solid particle; steam partial pressure: 0 MPa).

Fig. 4. The correlation between the estimated apparent rate constant and steam partial pressure (at reaction temperature: 900 °C; residence time: 4.6 s for gas, 0.78 s for solid particle; feeding rate: 0.5 g/min).

Fig. 5. The contours of different lumps derived from the CFD simulation (in run 4, (a) Carbon gases; (b) Tar; (c) Char; (d) Soot; Sum of mass fractions of gas phases=1; Sum of mass fractions of solid phases=1).

(a)

(b)

(c)

Fig. 6. The effect of reaction conditions (a. Temperature; b. Solid hold-up; c. Steam partial pressure) on tar consumption along the distance from the entrance of the DTR.

Fig. 7. Experimental yields vs. predicted yields for all experimental conditions used in this study.