



Modeling coal pyrolysis in a cocurrent downer reactor



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ABSTRACT

In this paper, a model for fast coal pyrolysis in a cocurrent downer reactor is developed, in which both hydrodynamics and coal pyrolysis kinetics are simultaneously considered. The results of simulations based on this model display reasonable agreement with experimental data obtained using Huolinhe coal as the feedstock, and this model is therefore suitable for predicting the fast pyrolysis of specific coal types. A series of simulations of fast coal pyrolysis in a cocurrent downer demonstrated that coal devolatilization is almost complete in the inlet region within a time span of 0.4 s, and that higher temperatures improve the pyrolysis efficiency. However, the yield of liquid products is decreased with increasing pyrolysis temperatures, especially above 670 °C, because of additional cracking of the liquids.

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Introduction

Coal pyrolysis has been considered an attractive means of using coal, especially low-rank coal (He, 2004) and, beginning with the worldwide oil crisis of the 1970s, researchers have focused significant effort on studying coal pyrolysis. In China, a number of studies have been performed in this area since the 1980s, many new technologies have been proposed, such as the MRF process developed by the Beijing Research Institute of Coal Chemistry (Du, Dai, & Yu, 1995; He, Liu, Dai, Du, & Shi, 1994), the DG process developed by Dalian University of Technology (Han, Guo, Luo, & Zhang, 1992), and the polygeneration technologies coupling pyrolysis with combustion developed by Zhejiang University (Cen et al., 1995), the Institute of Coal Chemistry (Wang, Liang, Dong, & Bi, 2005), the Institute of Engineering Thermophysics (Lv, Liu, Na, Zhao, & He, 2009) and the Institute of Process Engineering (IPE) (Kwauk, 1998; Kwauk, Yao, Lin, Li, & Wang, 2001; Yao & Kwauk, 1995; Yao, Wang, Lin, Li, & Kwauk, 2001). Research into the MRF process was suspended because of low yield of tar and high energy consumption, while the DG process also showed similar problems due to low heating rates. The process proposed by Kwauk et al. (2001) of IPE aims to develop a polygeneration process combining gases and light liquid product extracts, as well as electricity generation with heat

production to realize the efficient utilization of the chemical and thermal energy of the coal. This process has the potential to offer higher yields of liquid products and better performance. The core of this technology is the so-called *coal topping process*, which uses a fast mixing downer reactor as a pyrolyzer and coal ash as the heat carrier, thus eliminating the need for sand or other solid carriers. Many studies (Cui, Lin, & Yao, 2006; Dong et al., 2012; Wang, Lu, Yao, Lin, & Cui, 2005) have focused on the coal topping process on the laboratory or pilot plant scale, and the results showed that the coal topping process was economically viable and improved yields of high value-added products. To date, however, there have been few publications in the open literature concerning simulations of the coal topping process, especially in a downer pyrolyzer. Liang, Wang, and Bi (2008) simulated coal pyrolysis with a solid heat carrier in a moving-bed pyrolyzer, and found that their model was effective in predicting the product yields, but this model did not take into account the hydrodynamic phenomena inside the reactor.

In this paper we report a model for coal pyrolysis in a downer reactor, taking into account both the hydrodynamic characteristics and the kinetics of coal pyrolysis. This model aims to predict the yields of pyrolysis products under different conditions as well as the hydrodynamic characteristics inside the reactor, such as the axial velocities of gases and solids, solids holdup, and residence times of gases and solids within the downer. The model may therefore provide valuable information with regard to designing and operating such reactors.

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Table 1
Proximate and ultimate analysis results for Hlh coal.

Coal sample	Proximate analysis (wt%, dry)			Ultimate analysis (wt%, dry)				
	Volatile matter	Ash	Fixed carbon	Carbon	Hydrogen	Nitrogen	Sulfur	Oxygen
Hlh coal	36.05	25.56	38.39	54.42	3.50	0.91	0.56	15.05

Modeling of coal pyrolysis in cocurrent downer reactor

Kinetics of coal pyrolysis

Several models have been developed to study coal pyrolysis. For example, the coal pyrolysis process was considered as a first-order reaction by [Badzioch and Hawksley \(1970\)](#), while [Kobayashi, Howard, and Sarofim \(1977\)](#) regarded coal pyrolysis as two parallel competing reactions, one dominating at low temperature and the other at high temperature. [Anthony, Howard, Hottel, and Meissner \(1975\)](#) treated pyrolysis as a series of independent parallel reactions and [Suuberg, Peters, and Howard \(1978\)](#) suggested that the generation of each key product of pyrolysis could be modeled using one or a few first-order reactions depending on the observed behavior during the pyrolysis. With the development of modern analytical instruments, several models focusing on coal structure have appeared, such as the functional group–depolymerization–vaporization–cross-linking model ([Solomon, Hamblen, Carangelo, Serio & Deshpande, 1988](#)), FLASHCHAIN model ([Niksa & Kerstein, 1991](#)), and chemical percolation model for devolatilization ([Grant, Pugmire, Fletcher, & Kerstein, 1989](#)). However, these models require large amounts of experimental data, such as those obtained from field ionization mass spectrometry ([Retcofsky, Thompson, Hough, Friedel, & Larsen, 1978](#)), thermogravimetric analyzer coupled with Fourier transform infrared analysis ([Carangelo, Solomon, & Gerson, 1987](#); [Solomon, Hamblen, & Schlosberg, 1985](#)), and nuclear magnetic resonance ([Friebolin, 1991](#)), to provide the necessary input information for the model, which limits their applications.

In this section, a model for coal pyrolysis is developed, based on the work of [Suuberg et al. \(1978\)](#), since this represents the most readily constructed model in terms of predicting the product yields from coal pyrolysis compared with other possible models, such as that of [Anthony et al. \(1975\)](#), from which only the yields of char, tar, and the total amount of gases can be obtained.

According to Suuberg model ([Suuberg et al., 1978](#)), coal pyrolysis may be described as a series of first-order reactions, as shown in Eq. (1):



The corresponding reaction rate is described as

$$\frac{dV_i}{dt} = k_i(V_i^* - V_i), \quad (2)$$

where V_i is the amount of resultant i produced up to time t , V_i^* is the amount of resultant i that could potentially be produced (i.e., at $t = \infty$). Here the reaction rate constant, k_i , is given by

$$k_i = k_{i0} \exp\left(\frac{-E_i}{RT}\right). \quad (3)$$

In Eq. (1), the term product_i represents the key products of coal pyrolysis, such as CO_2 , CO, and others, produced at different stages in the reactor. Each stage dominates within a specific range of temperature, so the yield of a given product is equal to the sum of the yields of that product from each stage. As an example, the

production rate of CO_2 is associated with reactions in three stages, and thus can be written as follows:

$$\frac{dV_{\text{CO}_2}}{dt} = \frac{dV_{(\text{CO}_2)_1}}{dt} + \frac{dV_{(\text{CO}_2)_2}}{dt} + \frac{dV_{(\text{CO}_2)_3}}{dt}. \quad (4)$$

The yield of tar is assumed to be controlled by two first-order reactions, one of which takes place at low temperatures and the other at high temperatures. According to the original Suuberg model, the yield of tar increases with increasing temperature within certain extent. However, many experiments ([Cliff, Doolan, Mackie, & Tyler, 1984](#); [Cui et al., 2006](#); [Cui, Yao, Lin, & Zhang, 2003b](#); [Tyler, 1979](#)) showed that tar production will reach a maximum and then decrease with rising temperature. Therefore, the reaction rate of tar is modified to equal the rate of production in the first stage minus the rate in the second stage, as defined below:

$$\frac{dV_{\text{tar}}}{dt} = \frac{dV_{(\text{tar})_1}}{dt} - \frac{dV_{(\text{tar})_2}}{dt}. \quad (5)$$

According to the study by [Jüntgen \(1984\)](#), the activation energy of each product is independent of coal type. However, the frequency factors and yields of products need to be modified to fit the experimental data, which are believed to be related to coal type. In this paper, experimental data obtained from the pyrolysis of Huolinhe (Hlh) coal in a spout-entrainment reactor operating at different temperatures, which had been used to simulate the downer operation ([Cui, Yao, Lin, & Zhang, 2003a](#)), were selected to obtain the proper frequency factors and yields of pyrolysis products. The activation energy constants were obtained from the publication by [Suuberg et al. \(1978\)](#). All the kinetic parameters are listed in [Table 2](#), while the proximate and ultimate analysis data for the Hlh coal are provided in [Table 1](#). The results obtained from simulation of Hlh coal pyrolysis between 200 and 1000 °C are plotted in [Figs. 1 and 2](#), together with the related experimental data.

[Fig. 1](#) summarizes the yields of solids, liquids, and gases at different temperatures, from which it is evident that the yield of solids decreases with increasing pyrolysis temperature because of the onset of additional pyrolysis reactions, which produce more volatiles, including liquid and gas products. With regard to volatiles, the total yield of gases increases continuously as the temperature increases, while the liquid yield (tar and water) increases

Table 2
Kinetic parameters for Hlh coal pyrolysis ([Suuberg, Peters, & Howard, 1978](#)).

Product	Stage	E_i (kcal/mol)	Log k_{i0} (s^{-1})	V_i^* (wt% of coal, dry)
CO_2	1	36.2	11.33	0.58
	2	64.3	16.71	1.02
	3	42.0	6.74	0.32
CO	1	44.4	12.26	1.26
	2	59.5	11.92	7.70
	3	58.4	9.77	2.77
CH_4	1	51.6	14.21	0.95
	2	69.4	14.07	2.01
C_2H_4	1	74.8	21.25	0.17
	2	60.4	12.35	1.56
HC^a	1	70.1	16.23	2.45
	2	37.4	9.99	14.99
Tar	1	75.3	15.56	5.72
H_2O	1	51.4	13.90	6.02
H_2	1	88.8	18.20	0.91

^a All hydrocarbons other than tar, CH_4 and C_2H_4 .

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