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Combustion characteristics and kinetic analysis of heavy tar from continuous pyrolysis of camellia shell

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ABSTRACT

The objective of this research was to study the combustion kinetics of heavy tar from continuous pyrolysis of camellia shell. TG-DSC was used for combustion study with heating rates of 10, 20, and 30 °C/min, temperatures from 26 to 800 °C, and air flowrate of 100 mL/min. Results showed that the combustion process, with three stages, includes low-boiling components volatilization, light components decomposition and combustion, and heavy components and char combustion. The correlation among heating rate, maximum mass loss rate and mass loss ratio and the combustion behaviors at each stage with different heating rates were analyzed. The activation energy and frequency factor of each stage were obtained by Friedman and Ozawa methods. The reaction mechanisms are the random nucleation and growth, and the kinetic parameters were obtained by fitting experiment data with theoretical models. The mechanisms and kinetic parameters obtained could be used to describe the heavy tar combustion.

1. Introduction

Biomass could be converted into three phases of products, including bio-char, bio-oil (heavy tar and light liquid), and non-condensable fuel gas by pyrolysis process, and the yields and quality of the products vary with reaction conditions. Comparing with biomass, the bio-oil is about 4-fold higher in energy density, and is convenient for transportation, storage, further refining, and direct combustion; thus bio-oil is commonly considered as the potential substitute for liquid fossil fuel [5,16]. The comparison of experimental study on combustion of bio-oil from slow pyrolysis and diesel showed that the bio-oil flames were shorter, wider, and brighter than that of diesel when combusted in a circular jet spray at atmospheric pressure [21].

However, from the standpoint of clean fuel application, the combustion of bio-oil is not easy due to its undesired properties such as high water content, high viscosity, and high corrosiveness [24]. In addition, bio-oil is a mixture of oxygen rich organics with varied molecular weights such as organic acids, ethers, esters, aldehydes, ketones, phenols, and alcohols [22]. These organics have different combustion characteristics; especially the heavy tar, which contains ring compounds such as phenol, furan, naphthalene, and some trace contents of char particles, which are the main obstacles for clean combustion [16]. In order to improve and optimize the combustion conditions and efficiency, it is necessary to understand the mechanism and kinetics of bio-oil combustion. TG (thermogravimetry), TG-DTA (thermogravimetry and differential thermal analysis), or TG-DSC (thermogravimetry and Differential Scanning Calorimetry) are the common tools used for combustion thermal analysis and methods such as Kissinger method, Friedman method, and Ozawa method are effective approaches to obtain the kinetic parameters for reaction kinetic models [14,15,18,19].

Due to the complexity of bio-oil combustion, the combustion process has been divided into three or more stages in general. The combustion characteristics of tar from pine wood pyrolyzed at 500 °C in fluidized bed has been studied [26], and three reaction stages of aqueous and light component volatilization, heavy component carbonization and char contained component combustion were found, and the heavy tar was comparable to fuel oil in combustion characteristics. The perovskite, dolomite, and mayenite catalyzed combustion kinetic behaviors of tar from gasification of fowl dropping have been studied with TG and DTA and the three-stage model consisting of volatilization, low-temperature oxidation, and high-temperature combustion was developed [12,13]. The result also indicates the TG data fitted the first-order combustion model. The TG- and DTA-based study on combustion characteristics of bio-oil from hydrothermal pyrolysis of swine manure

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also showed the combustion process consisted of three stages including evaporation of water and lighter compound, ignition and burning of heavier compound, and carbonate compound decomposition, respectively, and the TG data fitted with the diffusion model [23]. The combustion characteristics of HZSM-5 catalyzed upgraded bio-tar (also referred to as bio-oil) from camellia shell pyrolysis showed four stages including light component volatilization, volatiles combustion, decomposition of combustion of non-volatile component, and residue combustion [6].

Bio-oil is a mixture of heavy tar and light liquid, and heavy tar has a higher energy content than that of bio-oil. Bio-oil can be used as energy source for combustion or can be separated to combust heavy tar alone. To our knowledge, there is very limited information about the combustion properties of heavy tar. The objective of this research was to study the combustion kinetics of heavy tar from continuous pyrolysis of camellia shell.

Camellia is one of the most important economic forest resources in China [8], while camellia shell is the residue of oil production from camellia, and most of the residues are not utilized adequately resulting in environmental pollution. In this study, the combustion characteristics of heavy tar from continuous pyrolysis of camellia shell were studied using TG-DSC analyzer. The Friedman method and Ozawa method were employed to calculate E and A during heavy tar combustion process using TG data. The reaction models were obtained by fitting the experiment data with the common kinetic mechanism models. This research aims at exploring the combustion behaviors of heavy tar, obtaining fundamental parameters of combustion of heavy tar, simulating the combustion processes, and providing reference for design of high efficient combustor for heavy tar.

2. Material and methods

2.1. Material

Camellia shell from Meizhou, China, was used as the feedstock for this study. The camellia shell was ground into particle size less than 830 μm for pyrolysis. The proximate and ultimate analysis of camellia shell was listed in Table 1. The bio-oil was produced from a continuous pyrolysis reactor (Fig. 1) at the reaction condition of 550 °C for 10 min with a feeding rate of 527 g/h. The bio-char was collected at controlled temperature of 250 °C to prevent pyrolysis vapors from condensation. The liquid products in the volatiles were condensed in the condenser first and then transferred into a separating funnel standing for 24 h for heavy tar precipitation. The heavy tar was separated from the bottom of the separating funnel. The yield of heavy tar was 2.91%/h (wt). The heavy tar pH value of 3.85 was measured at ambient temperature by pH meters (Hanna PH 211, Italy), and the moisture content, density, and lower heating value were 3.18% (wt), 1275 kg/m^3, and 30.22 MJ/kg according to standards of GB T 260–1977, GBT 2013–2010, and GB/T 384–1981, respectively.

A single Quadrupole GC–MS system (Thermo Fisher, TX, USA) was used to analyze the chemical composition of heavy tar. The GC–MS conditions were as follows: the chromatographic column was HP-INNOWAX (30 m × 0.25 mm × 0.25 um); the programmed temperature started at 35 °C and increased to 150 °C at a rate of 10 °C/min, and held for 5 min, then increased to 200 °C and held for 5 min, finally the temperature was increased to 230 °C and held for 10 min. The sample injection temperature was 240 °C; the injection volume for each sample was 1 μL; the split ratio was 10:1; the sampling mass range was 35–450 amu; the carrier gas was high-purity Helium at the flow rate of 1 mL/min. Table 2 shows chemical composition of heavy tar from camellia shell pyrolysis.

2.2. Apparatus and method

The TG-DSC analyzer (NETZSCH STA449C Jupiter®, Germany) was used to study the combustion characteristics of heavy tar from continuous pyrolysis of camellia shell. The TG-DSC analyzer had a TG resolution of 0.1 μg and the DSC resolution was less than 1 μW. For each analysis, 20 mg of the heavy tar sample was used. The reaction temperature ranged from 26 °C to 800 °C with the heating rate of 10, 20, and 30 °C/min, respectively. The flow rates of the purge gas of nitrogen and the reaction air were 30 mL/min and 100 mL/min, respectively. In order to eliminate the influence of crucible thermal behavior on the experiment, all the crucibles were calcinated at 1000 °C before used.

2.3. Kinetic parameters

Eqs. (1) and (2) are integral form and differential form of kinetic equations, which are widely used for determination of the kinetic parameters of combustion, pyrolysis, and other treatments of biomass feedstocks [3,4,10,11,20,25].

$$G(\alpha) = \int_0^\alpha \frac{d\alpha}{f(\alpha)} = \frac{A}{\beta}\int_0^T \exp(-E/RT)dT = \frac{AE}{\beta R}\int_\infty^u \frac{-e^{-u}}{u^2}du = \frac{AE}{\beta R}p(u) \quad (1)$$

$$\frac{d\alpha}{dT} = \frac{A}{\beta}f(\alpha)\exp(-E/RT) \quad (2)$$

where α stands for the fraction of conversion at time t, calculated by Eq. (3) and u is a defined variable calculated by Eq. (4) [9]. A is the frequency factor (1/s), E is the apparent activation energy (J/mol), R is the universal gas constant, 8.314 (J/mol K), T is temperature (K), β is the heating rate (K/min), M_0 is the mass ratio at the beginning of the reaction (%), M_i is the mass ratio at temperature T (%), and M_∞ is the mass ratio at the end of the reaction (%).

$$\alpha = \frac{M_0 - M_i}{M_0 - M_\infty} \quad (3)$$

$$u = \frac{E}{RT} \quad (4)$$

To solve Eq. (1), many approximate calculation methods were developed based on different treatment on $P(u)$ in the equation, such as Ozawa method, Phadnis method, Coat-Redfern method, Segal method, etc. The Ozawa method was used to obtain Eq. (5) [18].

$$\lg\beta = \lg\left(\frac{AE}{RG(\alpha)}\right) - 2.315 - 0.4567\frac{E}{RT} \quad (5)$$

According to Eq. (5), with different β at constant α, $G(\alpha)$ is a constant, thus $\log\beta$ and $1/T$ are in linear relationship. E was obtained from the regression slope of $\log\beta$ and $1/T$. The Ozawa method for solving E does not employ the detailed mechanism function, therefore, it could avoid the error on the E. Meanwhile, the frequency factor A could not

Table 1
Proximate and ultimate analysis of camellia shell.

Proximate analysis (air dry basis, %)				Ultimate analysis (dry and ash free basis, %)					Lower heat value (MJ/kg dry mass)
FC	V	M	A	C	H	O	N	S	Q_L
16.75	64.70	15.66	2.89	42.27	5.11	46.70	0.52	0.40	15.95

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