



Characteristics of rice husk tar secondary thermal cracking



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ABSTRACT

A two-stage fixed bed pyrolysis system for rice husk tar high-temperature thermal cracking was designed. Pyrolysis of rice husk and high-temperature thermal cracking of the rice husk tar were separated. High-temperature flue gas was used to promote rice husk pyrolysis, and then high-temperature secondary thermal cracking tar started in the high-temperature environment. The characteristics of secondary thermal cracking of rice husk tar were investigated. Results show that rice husk tar yield is 18 mg/kg of dry rice husk, or 11.7 mg/Nm³ at 1200 °C, and no tar is collected at 1300 °C. At the temperature of 1200 °C and residence time of 0.5 s, the rate of decline of tar reaches the maximum. The oxy-organics content in the tar at 700 °C reaches the maximum 24.5% and becomes zero at 1100 °C. The naphthalene content in the tar reaches the maximum 26.4% at 800 °C. With the increase in temperature, all the tar converts into non-condensable gas and char finally.

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1. Introduction

Biomass tar is a complex mixture of condensable hydrocarbons, which is generated from the process of biomass chemical conversion [1]. It is generally defined as an organic matter with a molecular weight larger than benzene, including oxygen-containing and complex polyaromatic hydrocarbons [2]. The definition on biomass tar is made mostly from condensing temperature of tar and molecular weight, which is mainly confirmed according to the research scope and condition of the investigators, or based on specific requirements [3,4]. However, there is no uniform definition on tar. Any organic matter that is generated by organic materials under heated condition or partial oxidation (gasification) effect can be considered as tar [5]. Also, it is usually believed that the tar is mainly composed of large molecular aromatic hydrocarbons. Currently, more than 100 species have been analyzed while it is still hard to define all of the components. What can be ensured is that the number of the main components is more than 20. Most of the components are derivative of benzene and PAH (polycyclic aromatic hydrocarbon). More specifically, there are seven types of benzene with its component greater than 5%, namely benzene, naphthalene, methylbenzene, dimethylbenzene, styrene, phenols, and indene [6].

Broido proposed the formation and variation model of tar in the process of biomass pyrolysis [7]. According to Broido-Shafizadeh model [7,8], biomass pyrolysis generates gaseous product (pyrolysis gas), liquid product (tar and water) and solid product (semi-char and ash) after pyrolysis. It is defined as the first reaction. The tar generated will continue to produce gas, char, and secondary tar in the pyrolysis reaction under a high-temperature environment. The process is defined as the secondary reaction. Kinetic model [9], product yields and kinetics [10], particle pyrolysis [11] and heat of reactions [12] for the process were studied, but till now, no statement has been widely accepted in the secondary reaction. The secondary pyrolysis of volatile matter involves a series of complicated chemical reactions. Generally, it is a process where chemical bond, which is supported by thermal decomposition and free radical theory, breaks, recombines, and lowers the average molecular weight of reactant. Under the condition of many pyrolytic reactions, experimental data show that light gas was the prominent product generated in the process of secondary pyrolysis. Hence, the simplest secondary reaction path “volatile matter → light gas” has been adopted widely. However, under such conditions, especially high temperature, and long residence time, semi-char will also become an important product of the secondary reaction, and significantly influence the distribution of pyrolysis product. On the other hand, the tar obtained from high-temperature pyrolysis was significantly different from the liquid obtained after cooling the primary volatiles [13]. According to the constituents of products of these two liquids, tar generated from conventional pyrolysis is the secondary product formed in the further disaggregation and

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condensation of volatile matter, namely the so-called secondary tar [3,14].

Tar blocks and contaminates the burning line, filter, engine, and turbine. The tar yield of using circulating fluidized beds for biomass pyrolysis and gasification is 1–30 g/Nm³ [15]. The tar yield of other forms of gasification equipment, such as downdraft gasifiers and updraft gasifiers, is 0.1–150 g/Nm³ [14,16]. In terms of most biomass pyrolysis and gasification application equipment, the required content of tar should be less than 0.05 g/Nm³ [17]. Hence, tar removal has become an urgent problem that must be resolved in the process of biomass pyrolysis and gasification technology. At present, biomass limited oxygen combustion is adopted in most biomass pyrolysis and gasification processes, but the tar content of product gas generated is high.

Tar can be reduced by thermal cracking that requires heating the tar to a high temperature or exposing the tar to catalysts at lower temperatures [3]. It is believed that biomass high-temperature pyrolysis is the most effective method to reduce tar content [18,19]. However, since the addition of oxygen to increase temperature will reduce the energy content of the gas product, most conventional biomass gasification processes operate below 900 °C, even if the reactors can operate at a higher temperature. Moreover, since biomass pyrolysis and high-temperature thermal cracking of tar usually occur in one-stage gasifier, it is difficult to control the temperature and residence time, which are the main factors affecting the tar content in product gas. Another issue is tar sampling for analysis. CST (Cold solvent trapping) and SPME (solid phase microextraction) adsorption are widely used in practice for tar online sampling in pyrolysis gas [20–22]. The CST method is more convenient for sampling tar from the gas with a high content of tar [20], but SPME adsorption is more precise and quicker, simpler and easier to operate [22]. For online tar sampling in the pyrolysis gas, both of the methods have their strengths and weaknesses.

To overcome these issues mentioned above, this paper designed a two-stage fixed bed pyrolysis system for rice husk tar thermal cracking. High-temperature pyrolysis of rice husk and high-temperature thermal cracking of tar were separated. First, high-temperature flue gas was used to promote rice husk pyrolysis; then high-temperature secondary thermal cracking tar started in the high-temperature environment. Although propane was used as the fuel for combustion in the experiment, it could be replaced by product gas generated from pyrolysis and gasification of the biomass itself for industrial application [23]. This option has more energy cost, but there is less energy loss of tar condensing and post-reduction and no risk of inactivation of catalysts. The objective of the work is focused on the characteristics of rice husk tar secondary thermal cracking. The temperature and residence time for the pyrolysis and secondary thermal cracking were monitored and controlled separately. The tar in the gas was condensed and collected; then the moisture was removed from the tar for GC–MS analysis. The influences of temperature and residence time on compositions of the gaseous product and the tar, and the cracking rate and yield of the tar were investigated. The operating conditions for thermal cracking to reduce rice husk tar will be provided.

2. Materials and methods

2.1. Material for the experiment

Rice husk typically found in Heilongjiang province, China's largest rice-growing area where the rice production was 1.5×10^7 t in 2014, was chosen as the raw material. Rice husk, an agricultural biomass resource, is composed of fiber, lignin, extractives and ash. Moreover, it is recognized that the product gas made from pyrolysis or gasification of the biomass-derived material is carbon neutral. For example, one ton of CO₂ will be mitigated by combustion of 2700 m³ of the gas made from rice husk char, which expresses the benefit of the carbon neutrality [24]. Rice husk is easy to collect since it primarily accumulates in rice processing plants. The rice husks in this paper are from a rice processing plant of Harbin, the capital city of Heilongjiang. The proximate and ultimate analysis of rice husk is shown in Table 1.

2.2. Experimental setup

The two-stage fixed bed pyrolysis system which is illustrated in Fig. 1 mainly consists of a primary burner, a secondary burner, a fixed bed reactor for pyrolysis, a high-temperature cracking reactor for the tar, a condenser, and a mixing chamber. Considering high-temperature flame was generated in the burners, stainless steel mesh was wired on the inner wall, and refractory concrete and construction aggregate were applied inside. Thermal insulation material was covered outside the burners. Propane and air supplied to the burner were tangential. An observation window was set on the end face of each burner for observing the ignition and burning condition. The fixed bed reactor was made of stainless steel with the inner diameter of 148 mm and the length of 700 mm for rice husk pyrolysis. Stainless steel featured by refractory concrete was installed on its inner wall. The primary burner and the secondary burner were the same, but ceramic tubes with small diameter were filled in the tail part of the secondary burner to increase the heat accumulation of fuel gas in the tail region. The high-temperature cracking reactor made of stainless steel with ceramic heat accumulator was for tar secondary thermal cracking into incondensable gas. Several high-temperature cracking reactors were made with the lengths and diameters from 0.5 to 2 m and $\phi 48$ to $\phi 219$ mm respectively.

2.3. Experimental procedures

Dry rice husk in its original size (2–10 mm) with the amount of 1.5–2 kg was fed to the grid plate initially, and valve 10 was open and valve 9 was closed. Propane and air sufficiently combusted in both of the burners. The flue gas analyzer was used to measure the oxygen content of the flue gas and adjust the proportion of propane and air to ensure there was no oxygen in the flue gas (O₂ is less than 0.2%). When the temperatures at the exit of the burners were stable, valve 9 was opened and valve 10 was closed so that the high-temperature flue gas generated from the primary burner entered the fixed bed reactor for the pyrolysis of rice husk. The temperature

Table 1
Proximate and ultimate analysis of rice husk.

Moisture _{ad} ^a (%)	Volatile _{ad} (%)	Ash _{ad} (%)	Fixed carbon _{ad} (%)	C _{daf} ^b (%)	H _{daf} (%)	O _{daf} (%)	N _{daf} (%)	S _{daf} (%)	Q _{net,d} ^c (kJ/kg)
5.08 ± 0.15	63.05 ± 0.60	14.98 ± 0.20	16.89 ^d	46.18 ± 0.50	6.08 ± 0.15	45.02 ^d	2.62 ± 0.08	0.10 ± 0.03	14,556 ± 120

^a ad: air-dry basis.

^b daf: dry, ash free basis.

^c Q_{net,d}: net calorific value, dry basis.

^d By difference.

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