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Thermogravimetric analysis and kinetic study of bamboo waste treated by *Echinodontium taxodii* using a modified three-parallel-reactions model

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HIGHLIGHTS

- A modified three-parallel-reactions model based on isolated lignin was proposed.
- Fungal pretreatment greatly enhanced the thermal degradation of bamboo.
- Fungal pretreatment decreased the negative effect of extractives on pyrolysis.
- Fungus made difficultly degradable component in lignin easier to thermal degradation.

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ABSTRACT

In this study, the effect of pretreatment with *Echinodontium taxodii* on thermal decomposition characteristics and kinetics of bamboo wastes was investigated by thermogravimetric analysis. The results showed fungal pretreatment can enhance the thermal degradation of bamboo. The negative effect of extractives in bamboo on the thermal decomposition can be decreased by the pretreatment. A modified three-parallel-reactions model based on isolated lignin was firstly proposed to study pyrolysis kinetics of bamboo lignocellulose. Kinetic analysis showed that with increasing pretreatment time fungal delignification was enhanced to transform the lignin component with high activation energy into that with low activation energy and raise the cellulose content in bamboo, making the thermal decomposition easier. These results demonstrated fungal pretreatment provided a potential way to improve thermal conversion efficiency of bamboo.

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1. Introduction

Moso-bamboo (*Phyllostachys pubesescens*), a usual fast-growing tree species for industrial purposes, is used widely as chopsticks, toothpicks, furniture and construction materials (Zhang et al., 2007). Millions of tons of bamboo wastes, including bamboo joint, bark and sawdust, are generated annually during bamboo wood processing. These wastes are usually burned for cooking and heating in the rural households or deposited in the landfill. In order to more effectively utilize bamboo wastes, some recent studies have focused on the pyrolysis of bamboo for biofuels and chemicals production (Jiang et al., 2012; Mui et al., 2008). Efficient thermal conversion of cellulose in bamboo is crucial to developing an efficient pyrolysis process because cellulose fraction is the most abundant and accessible component in pyrolysis of lignocellulosic biomass. However, conversion of cellulose fraction is usually affected by lignin fraction with high thermal resistance because the cellulose microfibril is usually embedded in the hemicellulose covalently linked to lignin (Yu et al., 2013). Thus, various physical, chemical and biological pretreatment methods have been used to deconstruct the lignocellulose matrix so as to decrease the biomass recalcitrance to the following thermal conversion (Das et al., 2004; Misson et al., 2009).

Compared with physical or chemical pretreatments (e.g., milling, alkaline or steam explosion), biological pretreatment has been considered a more energy saving and environmentally friendly technology (Zeng et al., 2011a). Particularly, a series of researches demonstrated that biological pretreatment using white-rot fungi with selective delignification ability can overcome biomass structure barrier and facilitate subsequent pyrolysis reactions, leading to increasing the thermal conversion rate, diminishing the

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temperature demand or decreasing the coke yield (Yang et al., 2010a, 2010b; Zeng et al., 2011b). However, few reports focused on the effect of fungal pretreatment on bamboo pyrolysis. Recently, our studies found that a novel white-rot fungus, *Echinodontium taxodii*, can improve the thermal decomposition of bamboo (Zeng et al., 2012). In general, the effects of fungal pretreatment on pyrolysis behavior of biomass are determined by fungal strain and pretreatment condition (Ma et al., 2013), but the effect of pretreatment condition on pyrolysis was neglected in the previous study. Kinetic study of pyrolysis is important to a better understanding of thermal decomposition behavior of biomass. In fact, thermal decomposition process involves many types of complex thermochemical reactions of heterogenous components in lignocellulosic feedstock (Mohan et al., 2006). However, our previous kinetic study assumed that bamboo pyrolysis is a homogeneous reaction. Thus, it is necessary to model the thermal decomposition and determine the kinetic parameters of overall pyrolysis reaction for elucidating in detail the influence of fungal pretreatment on the thermal behavior of bamboo wastes.

Numerous modeling methods have been proposed for elucidating pyrolysis mechanism (White et al., 2011). One of the most widely used models is independent parallel reactions model, especially three-parallel-reactions model which assumes that each component in lignocellulosic biomass decomposes independently to volatiles and char (Grønli et al., 2002; Sun et al., 2011). The three-parallel-reactions model regards the pyrolysis of lignocellulose as a combination of independent reactions of three pseudo-components which are considered to be lignin, cellulose and hemicellulose, respectively. Total reaction rate of biomass pyrolysis is the sum of the partial contributions of three pseudo-components. Moreover, the contents of three components in lignocellulose can be also determined by kinetic analysis (Sun et al., 2011). This model has a simple form and provides an accurate prediction between calculated values and experimental data. However, the introduced three pseudo-components can't truly represent three fiber components in lignocellulosic biomass and the effect of other non-fiber components (e.g., extractives) on pyrolysis is ignored. Particularly according to this model, the pyrolysis of pseudo-lignin happens after cellulose pyrolysis. Actually, lignin has a higher thermal stability and decomposes in an extensive temperature range (from 150 to 900 °C) (Yang et al., 2007). In addition, some studies demonstrated that the pyrolysis of lignin is not just a simple first-order reaction (Manya et al., 2003). Therefore, it's necessary to modify the three-parallel-reactions model for a better description of pyrolytic behavior of bamboo lignocellulose.

The objective of this study is to clarify the effect of fungal pretreatment with *E. taxodii* on thermal decomposition characteristics and pyrolysis kinetics of bamboo wastes using thermogravimetric and kinetic analysis. In order to investigate how fungal pretreatment improves bamboo wastes pyrolysis, the effect of fungal pretreatment time on thermal decomposition behavior of bamboo wastes was described in detail. Furthermore, a modified three-parallel-reactions model was proposed to simulate the overall pyrolysis process of bamboo lignocellulose based on pyrolysis study of natural lignin isolated from bamboo. Via this modified approach, the study provided a better understanding of the effect of fungal pretreatment on the pyrolysis kinetics of lignocellulose and offered new insights into improving the efficient use of bamboo wastes.

2. Methods

2.1. Fungal strain and pretreatment of bamboo

The white-rot fungus *E. taxodii* used for pretreatment of bamboo waste was isolated from Shennongjia Nature Reserve (Hubei, China) (Zhang et al., 2007). The fungal strain was maintained on potato dextrose agar (PDA) slant at 4 °C. Inoculum was grown on PDA plate at 28 °C for 8 days before inoculating. Moso-bamboo waste from Wuhan, Hubei province in China was smashed using a grinding mill, passed through 0.45 mm screen and then was air-dried. Fungal pretreatment was carried out in 250 mL flasks containing 6 g of bamboo waste and 13.5 mL of distilled water. After autoclaving (121 °C, 30 min), a plug cut from the margin of the slant culture was inoculated in the flasks. The flasks were cultured at 28 °C statically for 15, 30, 60, 90 days, respectively and then were dried at 60 °C for 3 days for the following study. Chemical analysis of lignin and cellulose contents in all samples was carried out according to procedures of "determination of structural polysaccharides and lignin in biomass" (Version 2006) from National Renewable Energy Laboratory (Yu et al., 2010).

2.2. Isolation of bamboo lignin

An enzymatic and acidolysis method was used for isolating lignin from bamboo sample (Guerra et al., 2006; Wu and Argyropoulos, 2003). After the sample was hydrolyzed with cellulase (30 FPU/g substrate) at 48 °C for 72 h, the insoluble substrate remaining was collected by centrifugation (2000 r/min), washed three times with deionized water, and then freeze-dried for 72 h. The substrate obtained was further hydrolyzed by an azeotrope of aqueous dioxane (dioxane/water 85:15, v/v, containing 0.01 mol/L HCl) under argon atmosphere. After the hydrolysate was centrifuged (2000 r/min), the supernatant was carefully withdrawn, neutralized with sodium bicarbonate, and finally added dropwise to 1 L of acidified deionized water (pH 2.0). The precipitated lignin was allowed to equilibrate with the aqueous phase overnight, and it was then recovered by centrifugation, washed twice with deionized water, and freeze-dried.

2.3. Thermogravimetric analysis

Thermogravimetric analysis was used to determine pyrolytic characteristics of bamboo samples and the isolated lignin. Thermogravimetric experiments were performed by using a sensitive thermobalance (PerkinElmer, Diamond, China). In order to identify the effect of extractives on pyrolysis, the washed bamboo samples with neutral detergent reagent were also subjected to thermogravimetric analysis (Zeng et al., 2011b). Prior to thermogravimetric experiments, all samples were ground by a grinding mill into powder, and then passed through 0.15 and 0.2 mm screens sequentially to make sure the particle size was between 0.15 and 0.2 mm. Initial sample masses of 5 mg were placed in the pan of the thermogravimetric analyzer microbalance. Nitrogen gas was used as carrier gas. Experiments were carried out using the thermobalance at heating rate of 10 °C/min, with the temperature range from 25 to 900 °C, at a steady nitrogen flow of 100 mL/min.

2.4. Kinetic modeling

The kinetic model of pyrolysis can be established based on the thermogravimetric (TG) or differential thermogravimetric (DTG) curve obtained from thermogravimetric analysis. The determination of kinetic parameters is usually based on the Arrhenius equation. In the gas-solid reaction, the rate of conversion $d\alpha/dt$ can be expressed by Eqs. (1)–(3):

$$\frac{d\alpha}{dt} = A\exp\left(-\frac{E}{RT}\right)(1-\alpha)^n \tag{1}$$

$$\alpha = \frac{m_0 - m_t}{m_0 - m_f} \tag{2}$$

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