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Comparative study on the thermal degradation of dry- and wet-torrefied woods

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HIGHLIGHTS

- Dry and wet torrefaction affect differently on the pyrolysis and combustion kinetics of Norway spruce stem wood.
- Degradation peaks of dry-torrefied wood are located at lower temperatures than those for wet-torrefied woods.
- Dry-torrefied wood shows a significantly higher combustion peak than wet-torrefied woods.
- Dry torrefaction does not influence the activation energy of wood cellulose and lignin, but wet torrefaction does.
- Dry and wet torrefaction have different effects on the kinetic parameters of char.

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ABSTRACT

In this study, effects of dry and wet torrefaction on the pyrolysis and combustion kinetics of Norway spruce stem wood were directly compared. The dry- and wet-torrefied woods were produced at appropriate conditions on the basis of a similar mass yield. Thermal reactivity of the woods was thermogravimetrically studied in nitrogen and air to simulate pyrolysis and combustion conditions, followed by a kinetic evaluation employing multi-pseudo-component models. The results show that the woods devolatilize more actively in air than in nitrogen. Moreover, the devolatilization and combustion peaks of the dry-torrefied wood are always located at lower temperatures than those for the wet-torrefied woods. Also, the dry-torrefied wood shows a significantly higher combustion peak than the wet-torrefied woods, but it ends at a lower temperature. Dry torrefaction removes more hemicellulose from the wood than wet torrefaction, at comparable conditions. It appears that dry torrefaction has unpronounced effects on the activation energy of cellulose and lignin in the subsequent thermal conversion processes, while these figures increase after wet torrefaction. In addition, wet and dry torrefaction show opposite trends in the char combustion: while dry torrefaction increases both the activation energy and pre-exponential factor of char, wet torrefaction decreases these kinetic parameters. Nevertheless, the mass fraction of char for the wet-torrefied woods is slightly higher than that for the dry-torrefied wood.

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1. Introduction

Nowadays, torrefaction is recognized as a promising pretreatment method to overcome the inherent drawbacks of biomass fuel, which include low bulk density, high moisture content, poor heating value, and low grindability. Two alternative torrefaction technologies have been introduced based on the treatment media:

dry and wet torrefaction. Dry torrefaction (DT) is defined as biomass pretreatment in the absence of oxygen and within the temperature range of 200–300 °C [1–4]. On the other hand, wet torrefaction (WT) is a hydrothermal pretreatment of biomass in hot compressed water at temperatures within 180–260 °C [5–7]. Although the biomass is pretreated in two different ways, both torrefaction processes are capable of producing a hydrophobic solid fuel with much better grindability, lower moisture content and improved heating value than the raw biomass. However, each torrefaction method has its own pros and cons. For example, moisture in the feedstock is critical for DT, but it is not problematic for WT [8]. As employing hot compressed water as reaction media, WT is

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more suitable for upgrading wet biomass and biomass residues such as forest residues, agricultural wastes, sewage sludge and aquatic biomass wastes, without a need of an energy intensive pre-drying step. Working with these wet feedstocks is challenging for DT. In addition, WT is capable of reducing the ash content in the solid product [9], while DT is not. However, up-scaling a DT process is less complicated than for WT because DT works at atmospheric pressure and with inert gases. WT requires a more advanced reactor capable of withstanding corrosive media at high pressures. Therefore, an industrial WT process may require higher investment cost than for DT.

A quantitative comparison of the solid products from WT and DT was carried out and reported in our previous work [7]. The comparison was performed based on a same solid yield, and looked at various criteria including the proximate and ultimate compositions, heating value, energy yield, hydrophobicity and grindability of the solid fuels produced by WT and DT. However, an investigation of the reactivity of these fuels in subsequent conversion processes (e.g. pyrolysis and combustion) was not included. Due to the differences in the process conditions between dry and wet torrefaction, the composition and structure of the torrefied biomasses are anticipated to vary. For example, the mass loss in DT is mainly caused by the release of water and volatiles from the thermal degradation of hemicellulose in an inert environment [10]. On the other hand, hydrolysis of hemicellulose is the main mechanism in WT, which contributes to the reduction of solid product yield [11,12]. Moreover, with a same mass yield of about 74%, it was found that the dry-torrefied wood had higher fixed carbon and lower volatile matter contents than the wet-torrefied woods. However, it has slightly lower HHV than the wet-torrefied woods, due to higher oxygen and lower hydrogen contents. These differences consequently affect the thermal behaviors of the torrefied woods. Because these fuels are feedstocks for further conversion processes, it is essential to understand the reactivity and kinetics of torrefied biomass fuels during these processes. Nevertheless, the number of kinetic studies on pyrolysis and combustion of torrefied biomass is still limited. Moreover, recent published works focused on the effects of a single torrefaction method, either WT [13–16] or DT [17–20]. No paper reporting a direct comparison between the two can be found in the literature so far. This motivates the need of this work.

This study, as a continuation of our previous assessment [7], aims at investigating and comparing the effects of WT and DT on the reactivity and kinetics of woody biomass fuels during pyrolysis and combustion. Dry- and wet-torrefied woods, with a solid yield of approximately 74%, obtained from the previous work, were studied further in a thermogravimetric analyzer in inert and oxidative environments to simulate pyrolysis and combustion conditions. Furthermore, a kinetic evaluation for each conversion process was also carried out employing multi pseudo-component mechanisms. Estimated kinetic parameters for the tested woods from the kinetic modeling are also presented and compared.

2. Materials and methods

2.1. Materials

Stem wood from Norway spruce was used as feedstock in this study because it is the main wood species in Norwegian forests. Wood samples were obtained from a local supplier in Trondheim (Norway), and cut into 1 cm cubes. The full details for WT and DT of these woods can be found in our previous papers [7,21]. In order to have appropriate comparison conditions, dry and wet torrefaction conditions were selected to produce a similar solid yield. The method for generation of these experimental points was also

described in [7]. For a solid yield of approximately 74%, it needs a DT condition of 275 °C and 60 min or less severe WT conditions of 210 °C and 30 min or 222 °C and 5 min. Hereafter, the wood samples are named according to their torrefaction method and temperature, i.e. WT-210, WT-222 and DT-275 for the woods pre-treated respectively by WT at 210 °C, WT at 222 °C and DT at 275 °C; while RS is used for the raw wood. Some fuel characteristics of the raw and torrefied woods used in this study are presented in Table 1.

2.2. Thermogravimetric analysis method and procedure

Prior to thermogravimetric analysis (TGA), all samples were ground by an IKA MF 10 cutting mill. Only particles with size less than 90 μm (sieved by a Fritsch Analysette 3 Pro vibrator) were selected for the kinetic study to ensure experiments in the chemical kinetic regime [22,23]. A thermogravimetric analyzer (Mettler Toledo TGA/SDTA851e) was employed for this study. In each TGA run, about 0.5 mg sample was spread in a 150 μl alumina pan located inside the TGA reactor. The sample was heated from room temperature to 105 °C and held at this temperature for 1 h for drying. Thereafter, the sample was heated up to 700 °C at a constant heating rate of 10 °C/min. Nitrogen and synthetic air (consisting of 21% oxygen and 79% nitrogen in volume) were used in the pyrolysis and combustion studies, respectively. A gas flow rate of 100 ml/min was applied for all experiments. In addition, three repetitions were performed for each tested sample.

2.3. Kinetic modeling

Pyrolysis and combustion kinetic modeling of lignocellulosic biomass fuels have been extensively studied for a couple of decades. In an inert environment (pyrolysis), the decomposition of biomass fuel releases volatiles and produces char. In an oxidative environment (e.g. combustion), the released volatiles and produced char are further oxidized. It is also well-documented that lignocellulosic biomass consists of mainly hemicellulose, cellulose and lignin. These components have different thermal behaviors. Therefore, a model with three parallel reactions is widely used for modeling the devolatilization of biomass fuel due to a high fit quality between the experimental and predicted data [24–27]. Furthermore, the char combustion is described by an additional reaction. In the present study, a general kinetic model is proposed as follows:



where S_i is the pseudo-component which produces volatiles V_i ($i = 1, \dots, 4$). The three first reactions (Eqs. (1)–(3)) are associated with the devolatilization of the three main components of biomass: hemicellulose, cellulose and lignin, respectively. They are used for modeling the pyrolysis of biomass fuel, in an inert environment. In air, the kinetic model includes the fourth reaction, to account for char combustion (Eq. (4)).

The conversion rates of all reactions obey the following Arrhenius expression:

$$\frac{d\alpha_i}{dt} = A_i \exp\left(-\frac{E_i}{RT}\right) (1 - \alpha_i)^{n_i}, \quad i = 1, \dots, 4 \quad (5)$$

where A is the pre-exponential factor, E is the activation energy of the reaction, R is the universal gas constant, T is the absolute temperature, n is the reaction order, and i is for the i th pseudo-

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