



Effects of wet torrefaction on pyrolysis of woody biomass fuels



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ABSTRACT

The pyrolysis of Norway spruce and birch woods under nitrogen atmosphere was studied by means of a thermogravimetric analyzer operated in the non-isothermal mode, followed by a kinetic analysis employing a three-pseudo-component model with n th-order reactions. Raw woods and the woods treated via wet torrefaction in the conditions of various temperatures (175, 200, 225 °C) and holding times (10, 30, 60 min) were included in this work. The study showed that wet torrefaction resulted in higher pyrolysis peaks for the woods, but less mass of volatiles was released during pyrolysis. The effects of wet torrefaction on pyrolysis of the lignocellulosic components are different. The activation energy of hemicellulose was significantly reduced by wet torrefaction. However, those for cellulose and lignin were slightly increased by wet torrefaction.

In addition, a kinetic evaluation with assumption of common parameters was performed. The results confirmed that some kinetic parameters can be assumed to be common for pyrolysis kinetic modeling of different biomasses without substantial reductions in the fit quality. Wet torrefaction converts different biomasses into more homogeneous solid products, of which the pyrolysis kinetics could be modeled by assumption of common parameters.

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1. Introduction

WT (Wet torrefaction) is a promising method for biomass pre-treatment and production of advanced solid biofuels. It may be defined as processing of lignocellulosic materials in HM (hydrothermal media) or HCW (hot compressed water) at temperatures between 180 and 260 °C [1–6]. The main product of the WT is “hydrochar” (hydrothermal biochar), a hydrophobic solid fuel with much better grindability, lower moisture content and higher calorific value compared with the untreated biomass [1–6]. In addition to hydrochar, WT also produces various water-soluble byproducts including acetic acid, formic acid, lactic acid, glycolic acid, levulinic acid, phenol, furfural, HMF (hydroxymethylfurfural), and sugars, which account altogether for approximately 10 wt% of the dry feedstock [4–6]. Separation and utilization of these organic fractions may contribute to an economic improvement of the WT process at industrial scales.

In comparison to DT (dry torrefaction), which can be defined as mild pyrolysis of biomass within the temperature range of

200–300 °C [7–9], WT is able to produce solid fuels with greater heating values, higher energy yields, and better hydrophobicity at significantly lower temperatures and shorter holding times [1,2]. In addition, WT is capable of removing parts of the ash components and hence produces cleaner solid fuels, with respect to inorganic elements [1]. Moreover, WT employs water in sub-critical conditions as reaction media, thus it is capable to work with wet biomass resources such as agricultural residues, forest residues, aquatic residues and other biomass wastes. A major benefit of the WT process is the reduced drying costs for wet biomass, as the solid product can be energy efficiently mechanically dried to about 35% moisture [10], where after other conventional drying methods can be used to further dry the product when needed. Hence, heat of evaporation needs for the moisture content exceeding the mechanical drying moisture content limit can be replaced by energy effective mechanical drying, saving considerable costs related to drying of the product, thus making it possible to produce valuable solid fuels from a variety of initially very wet biomass feedstocks. However, there are some engineering challenges remaining for the WT technology: reactor corrosion, precipitation and deposition of released inorganic salts, as well as handling of aqueous products. These issues may increase the investment cost for the WT process. Moreover, WT does not need pre-drying energy but it requires

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energy to pressurize and heat up water. This amount of heat should be recovered in order to achieve high energy efficiency.

At present, combustion is the most important energy application of solid biomass fuel, considering its contribution to more than 90% of the global bioenergy deployment [11–15]. Pyrolysis (devolatilization) is the first step, after drying, in the combustion process of solid biomass fuel. In addition, pyrolysis can be used as a standalone process for further treatments of biomass fuels [16]. Therefore, it is important to understand the thermal behavior and kinetics of biomass pyrolysis for the design, modification or optimization of the thermal conversion units. For these reasons, past research in the field was very active. For example, Broström et al. [17] employed a multi pseudo-component kinetic model to study effect of DT on the pyrolysis of Norway spruce. The model described closely the actual pyrolysis, assuming first order reactions. Later, Tapasvi et al. [18] established an even more detailed and more complex model, mainly based on the DAEM (distributed activation energy model), to describe better the pyrolysis and combustion kinetics of biomass. On the other hand, Manyà et al. [19] and Conesa and Domene [20] found that the pseudo-component kinetic model with n th order described the biomass pyrolysis kinetics better than the model with first order reactions. More recently, a comparative kinetic evaluation on pyrolysis of dry-torrefied stump biomass has been reported [21]. The results showed the differences in fit quality of the three-pseudo-component model with first order, the three-pseudo-component model with n th order, and DAEM were insignificant. In addition, the three-pseudo-component model with n th order was recommended for a pyrolysis kinetic study of solid biomass fuel.

Despite the advantages of WT over DT and the importance of understanding the thermal behavior and kinetics of biomass pyrolysis, only few studies on WT [1–6] can be found in the literature, of which the focus was on the effects of process parameters on the yield and fuel properties of the solid product. To our knowledge, there is no open literature available for pyrolysis of hydrochar, except for the one reported by Yan et al. [22]. In that study, the pyrolysis of solid obtained from HTC (hydrothermal carbonization) of loblolly pine was thermogravimetrically analyzed and compared with that of the untreated pine. Two simple kinetic models, Kissinger's and Ozawa's methods, were employed for a kinetic study of the pyrolysis. However, the kinetic information obtained by these methods is limited and does not closely represent the reality, considering the complexity in the chemical composition of lignocellulosic biomass materials. It is therefore crucial for WT technology to carry out more relevant studies on thermal behavior and kinetics of hydrochar pyrolysis.

In addition, Trninić et al. [23] indicated that, in kinetic modeling for thermal decomposition of biomass, it is possible to have common kinetic parameters for various types of lignocellulosic biomass fuels, considering the similarities and differences among the fuels. Also, Tapasvi et al. [18] suggested that if some of the kinetic parameters are assumed to be common, the following benefits can be achieved: (1) the common parameters indicate the similarities in the kinetic behavior of different samples; (2) a given parameter value is based on more experimental data, it is therefore less dependent on the experiment uncertainties. For this purpose, a first approximation of similarities for lignocellulosic biomass materials can be made based on the fact that their main organic components are hemicellulose, cellulose, and lignin [18,23]. If the approximation is good enough, then only the contribution factors for the pseudo-components need to be varied from biomass to biomass.

The present work aims at investigating the effects of WT conditions (temperature and holding time) on the thermal decomposition behavior in nitrogen and pyrolysis kinetics of woody

biomass. Norway spruce and birch woods were first pretreated via WT in different condition [1]. Pyrolysis of the hydrochar obtained from the pretreatment were then studied in a thermogravimetric analyzer and compared with the untreated materials in this work. The three-pseudo-component model with n th order was adopted for the kinetic analysis, including a kinetic evaluation for different model variants by assuming common parameters.

2. Materials and methods

2.1. Materials

The raw and torrefied woods used in this work were obtained from our previous work [1]. A brief description of the materials preparation is given here. Norway spruce and birch wood cubes with sides of 1 cm were used as feedstock. The dry feedstock and distilled water (with a ratio of 1:5 by weight) were placed in a 250 ml Parr reactor series 4650 (Parr Instrument, USA) for WT at a constant pressure of 70 bar but different temperatures (175, 200, 225 °C) and holding times (10, 30, 60 min). After WT, the wet solid products were dried in an oven at 105 °C for 48 h and then stored in a desiccator for further analyses. The proximate and ultimate analyses of the raw and wet torrefied samples used for the TGA in this work are listed in Table 1. The HHVs (higher heating values) were calculated on dry and ash free basis, according to Channiwalla and Parikh [24].

2.2. Thermogravimetric experiments

TGA (thermogravimetric analysis) is a proven method for studying the pyrolysis of biomass [25–28]. In the present work, a Mettler Toledo TGA/SDTA851e was employed and operated in a non-isothermal mode. The fuel samples were first ground by an IKA MF 10 cutting mill, and particles less than 125 μm (sieved by a Fritsch Analysette 3 Pro vibrator) were collected for the pyrolysis/devolatilization study to ensure that the experiments were in the kinetic control regime [29–31]. A sample of 2 mg was put in a 150 μl alumina pan for the TGA in a Mettler Toledo TGA/SDTA851e reactor. It should be noted that the buoyancy effect plays a significant role for such a small sample weight. Therefore, running a blank curve is mandatory, and the weight change of the blank experiment was subtracted from the experimental curves automatically. The experiment started from room temperature, the fuel sample was heated 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 per minute. A nitrogen flow rate of 100 ml/min was applied for all experiments. In addition, all experiments were triplicated and the average values as well as standard deviations are reported.

2.3. Kinetic modeling

In this study, a global kinetic model with three parallel reactions adopted from Branca et al. [29] was employed for modeling pyrolysis kinetics. One advantage of this model is that it does not require testing the fuel at different heating rates. It is because the heating rate within the range from 3 to 108 °C/min does not affect the activation energy, pre-exponential factor and reaction order. Moreover, the model can well describe the separate decompositions of the three main components of lignocellulosic biomass including hemicellulose, cellulose, and lignin [17–20,29,32,33]. The three independent parallel reactions used in this work are shown below:

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