Biomass and Bioenergy xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Biomass and Bioenergy

journal homepage: www.elsevier.com/locate/biombioe

Research paper

# Effects of the biomass moisture content and pelleting temperature on the pressure-induced agglomeration process

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ARTICLE INFO

*Keywords:*
Cellulose crystallinity index
Lignocellulosic polymers
Moisture content
Pellets
Strength
Thermal treatment

ABSTRACT

The aim of this study was to determine the thermal characteristics and structural changes of pelletized biomass at different moisture contents. Miscanthus, Jerusalem artichoke and *Spartina pectinata* at 10, 20 and 30% moisture contents were used. Irrespective of the type of biomass, heating induced changes in the FTIR bands at 875 and 1420 $cm^{-1}$, characteristic of lignin monomers and amorphous cellulose, respectively. An increase in the crystallinity index was observed, and the most resistant to compressive loads pellets were obtained with a 20% moisture content of *Spartina* and Jerusalem artichoke. Miscanthus and Jerusalem artichoke had the same trends in the distribution of their maximum cellulose melting points temperature peaks according to the moisture content used. For Miscanthus, this temperature was higher by 24 °C than for Jerusalem artichoke, and two-step decomposition of cellulose was observed, while one-step decomposition was observed for Jerusalem artichoke as well as for *Spartina*. The novelty of these studies was the use of modern instrumental methods (DSC and FTIR) to better understanding the changes occurring in biomass with a wide range of moisture content (10–30%) as a result of heating the material during the pelleting process.

## 1. Introduction

Biomass pellets are produced by pressing milled material through a die that is shaped either like a ring or a flat plate. The biomass is exposed to high pressures and heat that arise from outer friction resulting from contact between the surfaces of the channel walls and the particles. It can also arise from the inner friction generated among particles.

There are different binding mechanisms in biomass granules and agglomerates. Agglomerations are formed by creating covalent bonds between adjacent particles by heating lignin above its glass transition temperature; by hydrogen bonds via chemical reactions, by increasing attractive forces, such as van der Waals forces; and by mechanical interlocking between fibres and particles [1,2]. In particular, Kaliyan and Morey [3] found that preheating the feedstock was not necessary, and they concluded that friction among particles and between particles and the die was sufficient to heat the material to the point where the binding agents were activated (i.e., the glass transition temperature of the binder, which is approximately 75 °C).

There are several key mechanisms relevant to the biomass pelleting process. The amount of moisture in the material and the temperature of the glass transition of the lignin have both been shown to have a significant impact on the pelleting process [4]. Additionally, varying the frictional properties of the material within the die ultimately impacts pellet durability and density [5,6]. The presence of free moisture between particles, especially in a wet agglomeration process, increases the cohesive forces between particles. Several studies have shown that the strength and durability of the products increased with increasing moisture content until reaching an optimum level (8–12%, w.b.) [3]. Moisture contents from 12 to 20% (w.b.) may improve the densification process under room temperature. Elevated temperatures promote plastic deformation of thermoplastic particles. The plastic deformation of particles is important for making permanent bonds. The range of optimum conditioning temperatures (65–100 °C) depends on the type of biomass. At higher temperatures, Kaliyan and Morey [1] predict an

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http://dx.doi.org/10.1016/j.biombioe.2017.10.029
Received 26 September 2016; Received in revised form 6 September 2017; Accepted 24 October 2017

increase in interfacial area between adjacent particles, resulting in a larger adhesive strength due to increased van der Waals forces, hydrogen bonding, mechanical interlocking and inter-diffusion of particles.

The mechanical properties of lignocellulosic polymers, i.e., strength and stiffness, are strongly affected by the moisture content and temperature. Hemicellulose, cellulose and lignin are the three main components of biomass, and they generally compose approximately 20–40%, 40–60%, and 10–25 wt%, respectively, of the total lignocellulose biomass [7]. The decomposition of these compounds occurs in the following order: hemicellulose, cellulose and lignin at temperatures of 220–315, 315–400 and 190–900 °C, respectively [8]. Lignin and hemicellulose are essentially thermoplastic polymers, whereas cellulose is partly crystalline with highly ordered crystallites interrupted by amorphous, disordered regions. For an amorphous polymer, the main softening temperature is critically important because many properties, e.g., the elastic modulus, change significantly as the material transitions from a glassy to a rubbery state.

Thermal analysis involves heating the solid sample and observing the physical changes and chemical reactions that occur under the influence of heat. During the heating process, various physicochemical phenomena characteristic of the tested substance can occur, such as a change of state (melting, crystallization or sublimation), phase transformation (creation of a crystal network), polymorphic transitions and chemical reactions, thermal dissociation, or reactions between the substrates in the solid phase.

The most important and commonly used method of thermal analysis is differential scanning calorimetry (DSC), which is a thermoanalytical technique in which the difference in the amount of heat required to increase the temperature of a sample is measured as a function of the temperature. By varying the heat flow, the melting point of substances and temperatures of polymorphic transitions can be determined as a function of the added binders, solvent, pressure and temperature. Structural changes in a substance induced by these factors can be studied by Fourier transform infrared spectroscopy (FTIR). Both the DSC and FTIR methods can be used to assess the ability of materials, such as biomass, to generate energy through the combustion process. These methods also determine the potential of obtaining value-added products through the agglomeration process.

A parameter called the crystallinity index (CI) has been used to describe the relative amount of crystalline material in cellulose. The CI of cellulose has been measured using several different techniques, including XRD, solid-state $^{13}C$ BMR, infrared (IR) spectroscopy and Raman spectroscopy. The determination of CI using FTIR spectroscopy is the simplest method [9]. This method only gives relative values because the spectrum always contains contributions from both the crystalline and amorphous regions. O'Connor et al. [10] proposed the Lateral Order Index (LOI, $A_{1420}/A_{893}$) to calculate the CI of cellulose. The absorbances at 1420 and 893 $cm^{-1}$ are sensitive to the amount of crystalline versus amorphous structures in the cellulose. In particular, broadening of these bands reflects a more disordered structure [9].

The thermal and mechanical behaviour of plant materials has a great influence on the formation of pellets in the agglomeration process [11,12]. Multiple thermo-analytical methods to investigate polymers were conducted by researchers [13,14].

However, there are only very few studies on changes of cellulose crystallinity in energy plant biomass during the pelletization process. Stelte et al. [2] suggested that it would be interesting to investigate the effects of the moisture content and temperature on the flow properties of the biomass polymers during pelletization, the resulting pellet strength, and inter-particle bonding. In this context, the softening properties of straw lignin and how its properties depend on the moisture content and temperature will be particularly useful.

The aim of the study was to determine the thermal characteristics and structural changes of biomass from energy plants at different moisture contents while forming pellets. The pellets were analysed by using differential scanning calorimetry and Fourier transform infrared spectroscopy.

## 2. Material and methods

### 2.1. General view

For tests, shredded biomass from giant miscanthus (*Miscanthus* × *giganteus*), Jerusalem artichoke (*Helianthus tuberosus*) and *Spartina pectinata* were used as control samples, and pellets were formed while controlling the temperature of the die.

For Miscanthus, *Spartina* and Jerusalem artichoke, the geometric mean values of the particle sizes were 2.60, 1.94 and 1.76 mm, respectively. The dimensionless standard deviation values were 1.86, 1.94 and 2.31, respectively, and the dimensional standard deviation values were 2.29, 2.38 and 2.14 mm, respectively (according to ANSI/ASAE S424.1) [15].

The material moisture contents before agglomeration were 10, 20 and 30% (w.b.). The material samples were stored in a climatically controlled chamber 3001-01 (Ilka, Feutron Greiz, Germany) and were consistently removed for testing.

The plant material moisture content was controlled using a drying-weighting method according to requirements of the ASAE S358.2 standard [16]. Measurements were made for three material samples from each plant with a mass of 20 g. Samples were weighed on laboratory scales WSP 600/C (Radwag, Radom, Poland) within an accuracy of 0.01 g and were then dried in the laboratory dryer (Pol-Eko Aparatura, Wodzisław Śląski, Poland) until a stable mass was observed at a temperature of 105 °C.

Initially, during the pressure agglomeration process of Miscanthus biomass, the temperature of heating the die was 100 °C and the temperature of the material was 85 °C. Further agglomeration of the biomass from Miscanthus, Jerusalem artichoke and *Spartina* was performed at temperature of 140 °C, while the temperature of the material was 93 °C. The die was heated by two thermal bands with a 1.3 kW total power for 1 h to reach the desired temperature. The heating elements were driven by a ESM 3710 controller and a thermocouple type J. The temperature inside the die was measured by melt pressure transmitters with integral thermocouples (OneHalf20 CT6V-15M-TCJ-M14) to an accuracy of 1 °C, screwed into the ring wall at three different die heights (15, 30 and 45 mm from the base) and symmetrically spaced every 120°. The die, a cylinder that was 300 mm in height with a 60 mm external diameter and 8 mm internal diameter, was installed on the bracket of a universal testing machine TIRAtest (Matest Service, Łódź, Poland), which was used to induce agglomeration at a speed of 10 mm $min^{-1}$. The effective length of the die orifice (meaning the height of the pellet in the cylinder) was 60 mm. The portion of single sample of agglomerated biomass was 0.2 g and was weighed to an accuracy of 0.00001 g on WPA 40/160/C/1 analytical scales (Radwag, Radom, Poland). The following parameters of the air in the laboratory room were monitored: temperature 20 ± 2 °C, humidity 65 ± 4% and pressure 1013 ± 11 hPa.

To check the strength of pellets on the compression, three pellets of 22.3 ± 1.5 mm length were made under stable agglomeration conditions. The strength trials were performed on the same strength testing machine (TIRAtest) at a compression speed of 10 mm $min^{-1}$. The maximum force to break pellets placed on their side (the pellets' cylindrical shape oriented horizontally) was measured using a disc shaped metal probe, 50 mm in diameter, attached to 10 kN load cell.

### 2.2. Thermal properties

In the DSC, the sample and reference material were maintained at nearly the same temperature throughout the test. The temperature program was designed such that the temperature of the sample holder increased linearly as a function of time.