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Pelletization properties of raw and torrefied pine sawdust: Effect of co-pelletization, temperature, moisture content and glycerol addition

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A R T I C L E I N F O

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A B S T R A C T

Biomass shows characteristics that makes it a promising feedstock for complementing traditional fossil fuels as main energy source. It is somewhat limited by its generally poor physical properties, but these can be enhanced by densification processes like torrefaction and pelletization.

The aim of the present work is to evaluate the influence of different solid biomass additives (almond shell, cocoa shell, grape pomace, *Miscanthus*, olive pomace and olive stone), and parameters such as temperature, moisture content and glycerol addition upon pine sawdust, PIN, and its torrefied counterpart, PINT, pelletization performance, paying special attention to their abrasion index, higher heating value and energy density.

It was observed that the addition of small quantities of lignin-rich solid additives, like grape pomace, enhances the natural binding properties of both PIN and PINT during pelletization using a bench-scale device.

It was also found that a 13% overall moisture content and a glycerol addition of between 10 and 20% improve the pelletization properties of PIN and PINT, respectively, and increase their energy density when compared to the raw samples.

1. Introduction

The constant growth in the world's energy demands, which has triggered a 1.9% annual increase in CO_2 emissions during the last three decades [1], could be compensated for by exploiting a diverse range of low carbon energy sources.

Biomass is generally considered a feedstock with promising characteristics for complementing and partially replacing traditional fossil fuels as our main energy source. It will also reduce the energy production impact of the greenhouse effect and global warming due to its CO_2 life-cycle neutrality [2] and its moderate NO_x and SO_2 emissions [3]. In addition to this, the autonomy of the resource will reduce dependence on imported fossil fuels in countries which lack them and in turn encourage self-consumption, stimulate the economy and energize rural area development [4].

The sustainable market price increase in the most common biomass fuels has shown agro-industrial wastes and fast growth energy crops to be a realistic alternative for use in domestic and industrial combustion systems.

It is widely recognized that raw biomass in general and wastes or energy crops in particular have poor physical properties, specially low energy density and a high physical and compositional heterogeneity that restrict their wider use as a general energy source [5]. However, these drawbacks can be overcome by applying certain pre-treatments, the most common of which are torrefaction and pelletization.

Torrefaction can be defined as thermal pre-treatment under mild conditions, i.e., 200–300 °C during 0.5–3 h of reaction time [6], under atmospheric pressure [7] and in an inert or low-oxidizing atmosphere [8]. Under these conditions, hemicellulose is partially decomposed to form volatile gases, while structural lignin and cellulose are barely affected, resulting in a hydrophobic product [9] with enhanced grinding and pelletization properties [10]. Due to a relative increase in the mass of carbon in the sample, torrefied biomass displays increased higher heating values (HHV) [11]. However, the bulk density values are even lower than those of raw biomass due to the loss of light volatiles that increases the porosity of the particles. Because of this, it is necessary to densify the torrefied products by means of pelletization that also enhances the energy density.

Several works are found in the literature on the pelletization of pure and blended, raw and torrefied biomass samples at different scales. Thus, Gil et al. [12] used a bench top press unit to test the pelletization properties of several raw biomass samples (pine, chestnut and *Eucalyptus* sawdust, cellulose residue, coffee husk and grape wastes) blended with two different coals (a high volatile bituminous coal and a semi-anthracite), in order to study their combustion behavior and abrasion indexes.

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Barbanera et al. [13] focused on determining the physical properties, chemical composition and durability of a densified agro-industrial by-product, olive pomace, mixed with olive tree pruning at different mass ratios.

Li et al. [14] used a single pellet press to co-pelletize three types of raw biomass (fir, camphor sawdust and rice husk) with sewage sludge to study the effect of densification variables like pressure, temperature or moisture and the energy consumption balance on the obtained pellets.

Brady et al. [15] developed a laboratory-scale system to manufacture refuse-derived fuel pellets by adding glycerin, a cheap and plentiful by-product of the biodiesel industry, to sawdust and concluded that it enhances the HHV of the raw biomass.

Rudolfsson et al. [16] also used a single pellet press to study the influence of torrefaction degree, pelletization temperature and moisture content on the densification process of Norway spruce.

Cao et al. [17] torrefied cedar and camphorwood at different temperatures and studied the influence of adding castor bean oil cake on their binding properties when co-pelletized using a bench-scale press.

Finally, Shang et al. [18] compared the effect of die temperature, moisture content, additives and torrefaction degree of raw and torrefied wood chips when pelletized in a single pellet press and a pilot pelletizer, respectively, using their strength, durability, density, HHV and grindability as quality criteria.

The aim of this work is to conduct an ample experimental study on the pelletization of raw (PIN) and torrefied pine (PINT) in a bench-scale device, and to evaluate the influence of different parameters such as temperature, moisture and additives (solid biomass wastes and glycerol) on the quality of the product. The obtained results are especially interesting as PIN is the raw material most commonly used in the local pelletization industry. The knowledge acquired from these tests will be used for scaling up to a pelletizing pilot plant level.

The main quality criterion employed in this work was the durability of the pellets obtained. The energy density and combustion properties of the samples with the highest mechanical resistance were also evaluated.

2. Materials and methods

2.1. Samples, additives and conditions

A softwood sample (pine sawdust –PIN–) and its torrefied counterpart obtained after 1 h torrefaction at 280 °C (PINT) were selected as reference fuels for this study due to their widespread availability, since this woody biomass is the most commonly used in the Spanish biofuel market. The torrefaction conditions were selected according to the optimum grindability and combustion properties they provide, as described in previous works of this group [19].

Other biomass fuels (almond shell –AS–, cocoa shell –CS–, grape pomace –GP–, *Miscanthus* –MIS–, olive pomace –OP– and olive stone –OS–) were also evaluated as possible additives to PIN and PINT, in the search for a synergetic effect that might enhance the pelletization capability of the base fuels. Most of the selected samples are seasonally available in the Spanish biomass market and cover three of the most important biomass groups: woody, energy-crop herbaceous and agro-waste resources.

The samples were air dried at room temperature to remove external moisture and later ground and sieved between 0.1 and 1 mm; this size range is usually considered as optimum for bench-scale pelletization as particle sizes over 1 mm commonly lead to breakage [12,20]. Fig. 1 shows the cumulative particle size distribution of the samples tested.

The results of proximate and ultimate analyses as well as higher heating value, HHV, of the biomass samples are summarized in Table 1. Fixed carbon and oxygen contents were calculated by difference, as mass percentages on a dry basis by means of Eqs. (1) [21] and (2) [22]:

$$\mathrm{FC}\ (\%) = 100-(\mathrm{VM} + \mathrm{Ash}) \quad (1)$$

$$\mathrm{O}\ (\%) = 100-(\mathrm{Ash} + \mathrm{C}+ \mathrm{N}+ \mathrm{H}+ \mathrm{S}) \quad (2)$$

The typical lignin contents of the studied samples were obtained from different sources [23] and databases [24]. The reference samples (PIN and PINT) were evaluated separately and also mixed to mass percentages of 50%. Blends with all the other biomass samples were also prepared on the basis of 50/50 mass ratio, to yield 15 raw samples: PIN, PINT, PIN-PINT, PIN-AS, PINT-AS, PIN-CS, PINT-CS, PIN-GP, PINT-GP, PIN-MIS, PINT-MIS, PIN-OP, PINT-OP, PIN-OS and PINT-OS.

After mixing, the effects of temperature (room temperature and the drying temperature of 105 °C), moisture content (0% and 13%) and glycerol addition (0, 10, 20 and 30%) were tested for each raw sample, representing a total of 80 different pelletization conditions.

Room temperature was selected as a testing condition to determine the natural pelletization capability of each sample, which is essential to establish any synergetic effect with PIN and PINT. On the other hand, a stove pre-drying temperature of 105 °C was selected to study the pelletization properties of every sample and to simulate the conditions that are to be encountered in the pelletizing pilot plant, where experience suggests that optimum temperatures are slightly below 105 °C, due to friction between the die and the biomass sample. The selected temperature agrees with previous studies [13] and is within the temperature range studied in most works [9,25].

Water is considered a natural binder and lubricant for biomass pelletization [20]. The value of 13% total moisture was selected as the 10–15% range is commonly reported as being the optimum for this purpose [26,27]. This moisture level is obtained by applying a previous 105 °C pre-drying step, followed by spraying and homogenizing the required amount of distilled water on the sample.

Finally glycerol is tested as an additive since it is reported to be a good lubricant, especially for torrefied biomass, enhancing the resulting fuel's higher heating value and reducing NO_x emissions and ash formation during combustion [28]. The HHV value of the batch of glycerol used for these experiments was recorded as 18.0 MJ/kg. The range of 0–30% was chosen on the basis of previous works [29]. To ensure a homogeneous blend of solid biomass and liquid glycerol, the latter was previously mixed with distilled water in a 50% mass ratio and sprayed over the selected fuel. The wet mixture was then dried at 105 °C for several hours in a stove until the external moisture added with the glycerol was completely removed.

In summary, each one of the fifteen raw samples was subjected to six different experimental pelletization conditions: room temperature with no additives, room temperature and 13% moisture, room temperature and X wt.% glycerol addition and pre-drying at 105 °C with X = 0, 10, 20 and 30.

2.2. Pelletization experiments and abrasion index determination

As previously stated durability was considered to be the main quality indicator for the samples prepared under different conditions. The first step in the experimental procedure therefore consisted in making 20 pellets for each sample (6 mm diameter, 3–5 mm height), using a TDP-1.5 single punch tablet press machine, fitted with a 550 W engine that supplied a maximum pressure of 15 kN.

All pellets were subjected to a durability test consisting in 3000 turns at 35 rpm in a specially designed rotating drum described in detail in previous works [30]. The mass of the material remaining after testing was sieved through a 2 mm sieve, so that the abrasion index (A_i), and the durability of the sample (*DUR*) could be calculated by means of Eqs. (3) and (4) [12,31].

$$A_i = \frac{M_0-M_R}{M_0}\cdot 100 \quad (3)$$

$$DUR = 100-A_i \quad (4)$$