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Production and Characterization of Pellets from Carbonized Pinus Patula Sawdust

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Abstract

The low bulk density of wood wastes causes handling, storage, and transportation issues, limiting its large-scale application. Pelletization can solve this inherent problem by converting biomass into dense and compact pellets with a regular shape and size. In order to evaluate the effect of particle size and binding agents on the pellets of carbonized Pinus patula sawdust, the particle sizes of 0.6 mm, 1.18 mm, and 2.3 mm, and binding agents of cow dung, molasses, and wastepaper are used. The experimental results indicate that the addition of cow dung and molasses into the carbonized sawdust results in the increase of calorific value, decrease of the ash content, and increase of the fixed carbon. As a result, the maximum calorific value of 28.47 MJ/kg, ash content of 2.93%, and fixed carbon of 59.32% are obtained by using molasses. The calorific value of 25.8 MJ/kg, ash content of 6.03%, and fixed carbon of 52.77% are obtained by using cow dung, whereas addition of wastepaper into carbonized sawdust results in a lower calorific value of 22.3 MJ/kg, highest ash value of 8.35%, and low fixed carbon of 43.2%. Therefore, the use of cow dung and molasses as a binder can be considered as a sustainable approach to improve the physico-chemical properties of biomass pellets.

Keywords: *Binding agent, Carbonization, Pellet, Pinus patula, Sawdust.*

1. Introduction

As an abundant source of renewable energy, biomass entails significant environmental, social, and economic benefits through utilization of residues and mitigation of environmental impacts of waste disposal [1, 2]. Among wastes, wood residues are a low-cost and plentifully available sources from wood processing industries, construction, and demolition activities [3, 4]. Sawmill residue’s are estimated to be over 25,000 tonnes per year in Ethiopia [5]. However, wood residues in its raw form has a low volumetric energy content, a low bulk density, a high moisture content, and a hydrophilic character [6]. Due to this, it can not be utilized efficiently and have little economic value, thus they are frequently abandoned or stacked up and left to rot due to a lack of alternative applications [7]. The bulk density that ranges 150–200 kg/m^3 for woody biomass can be densified up to 700 kg/m^3 by increasing its compositional homogeneity through pelletization process [8]. Pelletization provides standardized physical properties, and combustion characteristics along with material uniformity, high energy content, high density, low moisture content, and reduced handling, transportation and storage cost [9, 10]. However, the lack of binding capability of cellulosic biomass leads to a low-quality pellets that have a low strength, dusty and still unsuitable for handling and utilization [9]. As a result, various researchers and industry stakeholders have been working to increase the effectiveness of the inter-particle binding in biomass pellets. In this regard, factors like particle size of biomass and its distribution [11], moisture content [12], pre-treatment of biomass [13], type of pelletizing machine [14], and the use of binding agents [15] have been explored by several researchers. Binders increase the pellet strength, quality, and prevent wear on production equipment by acting as a bridge or matrix, allowing the biomass components to create strong inter-particle bonds [16, 17]. However, few categories of binders may result in environmental pollution when used to increase pellet strength [18]. For instance, inorganic and chemically treated binders may cause problems regarding emissions, deposit formation, and corrosion. Since commercial binders are rich in minerals (like bentonite), sulfur (like lignosulfonate) or nitrogen (like protein), emission problems are expected due to the presence of high N, Cl, S, and heavy metals [19]. This negative impact of commercial binders have dictated the researchers to turn into organic, locally available, and environmentally friendly binders. Several authors have investigated the effect of different binding agents on quality of pellets. Bustamante *et al*. [19] have utilized carbohydrates (molasses and fructose), and have revealed that the use of carbohydrates improve the heating value of pellets. In addition, Mišljenović *et al*. [20] have investigated the effects of molasses on wheat straw palletization, and have found that addition of molasses reduce energy consumption and improve the pellet quality. Besides using binding agents, carbonization of biomass has been proven to increase the energy density of pellets [21]. Carbonization increases energy density as char heating value is about 25-30 MJ kg^{-1} compared to 15 MJ kg^{-1} for raw biomass [22]. Carbonization also increases the physical properties such as hydrophobicity and resistance to microbiological attack [21]. Therefore, this work intends to show how pellets are produced from carbonized *Pinus patula* sawdust in a lab-scale pelletizing machine using locally available, organic binders (cow dung, molasses, and waste paper), and investigates the effects of binding agents and particle size on the physico-chemical properties of pellets.

2. Materials and methods

2.1. Sample collection and preparation

Around 100 kg of the *Pinus patula* sawdust sample was collected from the Munessa sawmill industry (Arsi Negelle), which is located 167 km from Addis Ababa. The collected sawdust sample was transported to the laboratory of Forest Products Innovation Research and Training Center, Addis Ababa. The sawdust was spread over a canvas carpet, and was left to air dry for two weeks in the shade until its moisture content was 10-12wt%. The dried sawdust was sieved into three particle sizes (0.6 mm, 1.18 mm, and 2.36 mm) EN16126 (2012) [23]. Molasses were collected from the Wonji Shoa Sugar Factory, which is 110 km from Addis Ababa, whereas the other two binding agents were collected from vicinity to the experiment site, i.e. cow dung was collected from the local dairy manufacturing enterprises and waste paper was collected from the Yekatit Paper Products Factory.

2.2. Carbonization of Pinus patula sawdust

Cylindrical barrel metal kiln with chimney, which was fabricated locally using a drum of about 1.2 m height and 90 cm diameter was used for carbonization of the sample. During carbonization, sawdust of selected particle sizes were spread over the inner-perforated surface of the kiln. Then the kiln was closed with screw immediately after dehydration completed when cloudy smoke became closely to blue so as limited oxygen environment was created, and it was left to carbonize for an average of 1 to 1:30 hours [24]. After that, the chimney was covered by clay mud for cooling in controlled manner. Subsequently, the carbonized sawdust was removed from the kiln, and stored in separate airtight containers to avoid moisture absorption before the experiment.

2.3. Pelletization process

A JS5RK/DL manual pellet press machine with a flat ring die was used for the experiment. According to the manufacturer's specification, the machine had the capacity to pelletize up to 20 kg/h. The rollers were of 12 cm in diameter and weigh 6 kg each. Molasses, cow dung, and wastepaper were used as binders to make the pellets. For pelletization, 3 kg of the carbonized sawdust sample of different particle size was manually mixed with 1 kg of the binder (3:1) [25]. Water was added to facilitate mixing and increase adhesion between the intermolecular particles. The mixture was fed into a pellet mill and pressed to get pellets of uniform surface, shape, and size. The pellets were then manually spread on the floor to air dry, and the dried pellets were then collected and stored in airtight bags at the room temperature to avoid the absorption of moisture from the atmosphere.

2.4. Characterization of pellets

Determination of moisture content: Moisture content (MC) on a dry basis was determined using the standard method ASTM D3173 (2017) [26]. 1 g of pulverized pellet sample was oven-dried at 105 °C for 2 h and weighed until the mass remained constant. Then MC of the sample was calculated using the following formula:

$$\mathbf{MC}(\%) = \frac{\mathbf{W1 - W2}}{\mathbf{W1}} \times \mathbf{100} \tag{1}$$

where W_1 is the initial weight of sample, W_2 is the weight after drying, and MC is the moisture content.

Determination of volatile matter: The volatile matter (VM) was determined using the standard method (ASTM D3175-18, 2018) [27]. 1 g of the pulverized pellet sample was taken and heated at 550 °C for 10 minutes, cooled in desiccator, and weighed until its mass remained constant. Finally, VM was calculated as follows:

$$\mathbf{VM}(\%) = \frac{\mathbf{W1 - W2}}{\mathbf{W1}} \times \mathbf{100} \qquad (2)$$

where VM is the volatile matter, W_1 is the original weight of the sample, and W_2 is the weight of the sample after cooling.

Determination of ash content: The ash content (ASH) was determined using standard of ASTM D3174-12(2018) [28]. In order to determine the weight of the ash, 1 g of the pulverized moisture free sample was heated in the furnace at 550 °C for 4 h, and weighed after cooling in a desiccator.

$$\mathbf{ASH}(\%) = \frac{\mathbf{W2}}{\mathbf{W1}} \times \mathbf{100} \qquad (3)$$

where W_2 is the weight of cooled ash, and W_1 is the original weight of the dry sample.

Determination of fixed carbon: The fixed carbon (FC) was determined by deducting the sum of VM, ASH, and MC from 100, as follows:

$$\mathbf{FC} = \mathbf{100\%} - (\mathbf{ASH} + \mathbf{MC} + \mathbf{VM}) \qquad (4)$$

Determination of calorific value: The calorific value (CV) of the produced pellets was determined using a Parr Oxygen bomb calorimeter following the standard of ASTM D5865-13(2019) [29]. The bomb calorimeter was first calibrated using a standard sample of benzoic acid that had a known calorific value of 26.4 MJ/kg. 2 g of the pulverized pellet sample was placed in the crucible, and further, the bomb calorimeter was operated according to the procedure described in the standard.

Determination of sulfur content: The sulfur content was determined according to the ASTM-D 3177-02 (2018) standard [30]. 1 g of pulverized pellet sample was mixed with 3 g of Eschka mixture in a porcelain crucible. After that, the mixture was covered with 1 g of Eschka mixture. The crucible was then placed in a cold muffle furnace and gradually heated to 800 °C over the course of 60 minutes.

$$\mathbf{Total\ sulphur} = \mathbf{A} - \mathbf{B} * \mathbf{13.738/C} \qquad (5)$$

where A is the mass of barium sulfate from the sample, B is the mass of barium sulfate from the blank, and C is the mass of the sample.

2.5. Data analysis

The data were statistically analyzed using the SAS software (Version 9) and Microsoft Excel (2016). To compare the means that showed significant differences, the Least Significant Difference (LSD) at the P ≤0.001 level was used.

3. Results and discussion

3.1. Variation in proximate analysis, calorific value, and sulfur content on pellets

The proximate analysis, calorific value, and sulfur content of the pellets revealed that the main effect of different binding agents on all test parameters, as well as the interaction effect between the sawdust particle size and the type of binders on MC, FC, ASH, VM, and CV were significantly varied at p = 0.001 (Table 1).

Table 1. Analysis of variance (ANOVA) for proximate analysis, calorific value, and sulfur content of pellets.

Source of variation	DF	Mean square					
		MC	VM	FC	Ash	CV	S
Sieve size	2	5.99***	97.45***	182***	5.62***	747799***	0.023*
Types of binding agents	5	35.56***	191.87***	182***	18.08***	1130489***	0.298***
Interaction effect	10	8.27***	31.54**	114***	5.30***	1351582***	0.041***
CV		1.22	4.04	3.75	8.14	2.28	31.91
R^2		0.99	0.94	0.94	0.93	0.97	0.91

*** = Significant at p <0.001; ** = Significant at p < 0.01 CV = Coefficient of variation, R = Regression factor, DF = Degree of fFreedom

3.2. Effects of binding agents and particle size on proximate analysis, calorific value, and sulfur content of pellets

Moisture content: At fixed binder and sawdust concentration, the increase in particle size resulted in a decrease in the final moisture content of pellets of molasses. This is in a good greement with the study conducted by [16]. The authors [16] reported that the moisture content of pellets made by using molasses decreased from 15% to 9% with increase of particle size. However, moisture contents of pellets of cow dung and wastepaper showed increasing trend with increase in particle size (Table 2). This trend of cow dung pellets is in line with the study conducted by Obidziński *et al*. [31], the authors reported moisture of pellets of sawdust using cow dung as binder increased from 5.7% to 8.3%. These values are also in a good agreement with CEN/TS 14961:2014 [32] standard for Solid biofuels, which states that the moisture of pellets should be in range of 8-10%. However, when the three binding agents compared interms of moisture content, highest value was obtained on pellets of molasses, this can be supported by the study conducted by Wang *et al*. [33], in which the moisture of raw molasses was reported 25.6%, which is quite high. The lower moisture contents were obtained on pellets of cow dung and wastepaper. These results can be supported by the study conducted by Iftikhar *et al*. [8], and Prabhakaran *et al*. [34], in which proximate analysis of cow dung and waste paper were reported 12.8% and 4%.

Volatile matter: From the results, at fixed sawdust to binder concentration (3:1), increasing the particle size resulted in decrease in the volatile matter of pellets of wastepaper and molasses. VM of pellets of molasses decreased with increase in the particle size (Table 2). This is in line with the study of Iftikhar *et al*. [8]. The authors reported the same trend of decreasing VM from 45% to 37%. When the three binding agents were compared transversely, the highest VM value was obtained by using waste paper, and the lowest value VM value was obtained by using cow dung. The highest VM value of waste paper was mainly due to the presence of 62.5% in wastepaper as reported by [34], and the low VM value of cow dung pellets is mainly due to the presence of low VM in cow dung of 33.53% [8]. The high VM values obtained for waste paper is an indication of easy ignition of wastepaper pellets during combustion [35, 36], whereas the low volatile matter of cow dung pellets is indication of incomplete combustion of pellets, which will give rise to the emission of significant amount of smoke and toxic gases as reported by [37].

Fixed carbon: From the results (Table 2), at fixed sawdust to binder concentration, increasing particle size resulted in increase in FC for pellets of waste paper and molasses, whereas the pellets of cow dung showed a different trend with highest value obatained at particle size of 1.18 mm. The increasing trend of FC values of pellets of wastepaper and molasses were in line with the finding of Iftikhar *et al*. [8], they reported that the FC value of pellets increased from 23.86% to 30.28%. When the FC values of pellets of the three binding agents were compared, the pellets made by using cow dung had a higher FC value than the pellets of waste paper and molasses. This could be due to the presence of high fixed carbon of 31.33% in cow dung as reported by [8]. The lowest FC value was recorded on pellet molasses. This low fixed carbon of pellets could be due to low FC of molasses (5.9% to 7.94%) as reported by [33, 38].

Table 2. Proximate analysis, calorific value, and sulfur content of pellets.

Test parameters	Particle size	Binding agents		
		Waste paper	Cow dung	Molasses
Moisture content (%)	0.6	5.85^{c}	7.47^{c}	12.9^{a}
	1.18	6.23^{b}	7.64^{b}	9.91^{b}
	2.36	6.39^{a}	7.93^{a}	7.48^{c}
Volatile matter (%)	0.6	42.54^{a}	33.73^{a}	42^{a}
	1.18	42.04^{a}	28.96^{b}	30.68^{b}
	2.36	38.47^{b}	34.01^{a}	$30.27b^{c}$
Fixed carbon (%)	0.6	43.2^{c}	52.77^{b}	38.23^{c}
	1.18	43.84^{b}	57.02^{a}	54.72^{b}
	2.36	47.9^{a}	51.79^{c}	59.32^{a}
Ash (%)	0.6	8.35^{a}	6.03^{b}	7.37^{a}
	1.18	7.88^{a}	6.08^{ab}	4.7^{b}
	2.36	7.24^{b}	6.55^{a}	2.93^{c}
Calorific value (MJ/kg)	0.6	22.3^{b}	25.8^{a}	19.78^{c}
	1.18	21.4^{b}	25.6^{a}	24.87^{b}
	2.36	21.7^{a}	24.38^{b}	28.47^{a}
Sulfur (%)	0.6	0.02^{b}	0.17^{a}	0.6^{a}
	1.18	0.02^{b}	0.15^{a}	0.38^{b}
	2.36	0.12^{a}	0.14^{a}	0.24^{c}

Values from top to bottom with the same letters are not significantly different.

Calorific value and ash content: At fixed binder and sawdust concentration, the increase in the particle size resulted in a decrease in the ash content of pellets of wastepaper and molasses, whereas pellets of cow dung showed an increasing trend with increase of particle size (Table 2). Decreasing of ash content of pellets of molasses resulted in increase in the calorific value of pellets. These lower ash values are merely associated with low ash content of 10.6% in molasses, as reported by [33]. The low values of ash content are also attributed to the presence of high carbohydrate content in molasses. The use of carbohydrates helps to lower the mineral content of biomass, which eventually increases the calorific value of biomass pellets [19]. On the other hand, the presence of a higher moisture content could also decrease the calorific value of pellets. When the moisture content of pellets increased, the corresponding heating values of pellets were decreased. A similar trend was reported by Iftikhar *et al*. [8]; the authors reported that the increase of moisture of pellets from 1.25% to 4.44% resulted in the decrease of calorific value from 13.82 MJ/kg to 12.82 MJ/kg. The maximum ash values of 8.35% was obtained on pellets of waste paper. This could be due to the presence of 32% ash in wastepaper, as reported by [34]. The ash values of pellets of cow dung were observed to increase; this resulted in a subsequent decrease in the calorific value from 25.8 MJ/kg to 24.38 MJ/kg (Table 2). This is basically due to the increasing trend of moisture content and high ash content of raw cow dung of 21.41%, as reported by [8].

Sulfur content: The obtained values of sulfur showed the decreasing trend for pellets of both molasses and cow dung with increase in the particle size, whereas the sulfur content of wastepaper pellets showed little increase (Table 2). However, all recorded values of sulfur content for all binding agents were in an internationally acceptable range. The values are in accordance with the German standard for high-quality pellets (DIN EN 15270: 2008-03) [39] and European standard for solid biofuels CTI-R04/5 (2011) [40], which states that the sulfur content for good pellets should be ≤ 0.5%.

4. Conclusion and recommendation

In conclusion, the use of binding agents is a sustainable option to enhance the physico-chemical characteristics of pellets of carbonized *Pinus patula* sawdust. With the use of molasses, the pellets presented good physico-chemical characteristics like 25.8 MJ/kg calorific value, 2.93% ash, and 59.3% fixed carbon, and with the use of cow dung 28.47 MJ/kg calorific value, 6.03% ash, and 52.77% fixed carbon. However, the high ash content of 8.35% and low calorific value of 21.4 MJ/kg was recorded for pellets of wastepaper. The increase in the pellet quality (high calorific value and low ash) of pellets of cow dung and molasses were mainly due to the high proportion of lignin content in raw cow dung and high carbohydrate in molasses. Therefore, from the aforementioned observations, the use of binders from lignocellulosic and starch source can be a sustainable solution to enhance the physico-chemical properties of biomass pellets. Even though the pellets of molasses exhibited a comparable quality with cow dung, few inherited properties like viscous nature, comparative advantage, and cost associated with the transportation of these binder renders common applications in the rural region. Therefore, further studies should investigate the possibility of further pellet quality and cost saving potential for molasses. Cow dung could be considered as one of the most suitable options to be used as binders for biomass pelletization.

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6. Nomenclature

MC	Moisture content
VM	Volatile matter
FC	Fixed carbon
AC	Ash content
CV	Calorific value

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