Possibilities for obtaining pellets by single-screw extrusion of biomass from black pine (*Pinus nigra* Arn.)

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Abstract. The aim of the present study was to investigate the possibilities for obtaining pellets by single-screw extrusion of black pine (*Pinus nigra* Arn.) biomass. Two fractions of black pine needles – 1) with $d_p \geq 500$ µm and bulk density $\rho_n = 0.243$ g/cm^3 and 2) black pine twigs with $d \geq 500$ µm and $\rho_n = 0.297$ g/cm^3, were obtained. Wheat bran with $d_p \geq 500$ µm and bulk density $\rho_n = 0.258$ g/cm^3 in a ratio of 1:1 was added to the obtained fractions. The experiments showed that getting pellets by single-screw extrusion of biomass from twigs and needles of black pine was possible when mixing them in a 1:1 ratio with wheat bran. The addition of wheat bran improved the smoothness and homogeneity of the produced pellets and increased their density. High-density pellets with a low sectional expansion index were produced during extrusion. The productivity of extruding mixtures of black pine twigs with wheat bran was 30% higher than that made from a mix of black pine needles and wheat bran. The pellets obtained after mixing biomass from black pine and wheat bran had over 13% higher density than those produced entirely from wheat bran. The presence of black pine biomass increased the quality of the final product while at the same time having a positive effect on both productivity and specific energy consumption.

1 Introduction

Renewable natural sources, including waste raw materials from the timber and processing industry, could be used as an alternative energy source [1]. Their utilization was in granular

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form, which created preconditions for disruption of the flow due to adhesion and the appearance of cracks [2-4].

Coniferous species and, in particular, black pine (*Pinus nigra* Arn.) are sources of biomass, not only in Bulgaria but also worldwide. The biomass of plant species was characterized by low energy density and high water content, requiring its pre-treatment (extrusion). Many physical and energy characteristics of black pine biomass, such as density, strength, calorific value, and others, were essential in the bioenergy market. The mechanical properties of the biomass used for extrudates are strongly influenced by the distribution and size of the individual particles in the system [5-7]. There were no standard methodologies in the literature for the properties of extrudates (granules) and values for the coefficients of friction, a slip of black pine biomass with different moisture values.

The density of the obtained granules was an essential parameter for evaluating the mechanical characteristics, as it also determines the pressure used to obtain the pellets [2, 8]. New conditions are created during the increase of the pellet's density to create close interaction between the system's components. This condition lowered the moisture content by decreasing the mass and significantly reducing transport and storage costs [9-11]. The creation of a dense structure of extrudates may also cause a decrease in their mechanical stability due to solid compaction [12].

Granulated biomass absorbed less moisture from the environment [9], but pellet extrudates require higher amounts of energy [13].

Some authors [6, 14] used waste products from soybean processing, sugar cane, eucalyptus wood, wheat straw, algae, willow, and cork in the composition of the extrudates. Their analyzes have identified several energy characteristics of biomass. The determination of mechanical parameters such as hardness assesses the quality of the pellets.

Most of the biomass obtained from processing, and agricultural production comprises lignocellulose, a composite of cellulose, hemicellulose, and lignin. Each of the polysaccharides involved in the biomass composition determined the different binding methods between the particles, which was the main factor determining the heat of combustion. The use of an unsuitable extruder or incorrectly selected extrusion conditions may cause lignin flow and migration of some substances to the surface. All these changes influenced the quality of the obtained extrudates [14].

Coniferous species used for timber, such as black pine, fall into the group of large amounts of biomass. Different energy characteristics were observed between the various anatomical parts of the coniferous species. The higher lipid content and volatile substances in the biomass increased the calorific value of the obtained extrudates used for combustion. For this reason, during drying or extrusion, some representatives of lipids and volatile components were lost, which negatively affected the energy potential of biomass [15].

Because of the increased interest in using bioenergy resources and applying the principles of the green economy in several industries, knowledge of their application is needed. Black pine in Bulgaria is a widely used species in the timber, wood processing, and essential oil industries. The large scale of the waste raw materials obtained during its processing contains materials with a composition that suggests many possibilities for their inclusion in the bioenergy sector. The case of its wide application is influenced by the fact that th black pine species is not a protected species in Bulgaria. To the best of our knowledge, this is the first scientific study evaluating the usage of raw waste products by black pine to produce pellets. Therefore, the aim of this study was to determine the options of obtaining pellets by single-screw extrusion of black pine biomass (*P. nigra*) obtained after extraction of the essential oil.

2 MATERIALS AND METHODS

2.1. Materials

Biomass (needles and twigs with needles) from black pine (*P. nigra*) which was obtained after extraction of the essential oil by water distillation was used in this study. The raw material was collected during the summer season (May - June 2020) in Central Balkan Nature Park, Bulgaria. After distillation, the biomass was dried at a temperature of 20 ± 2 ºC and an air velocity of 0.5 m/s.
The wheat bran (obtained from soft wheat) was purchased from the commercial network (Claremont company).

2.2. Methods

2.2.1. Biomass grinding

The raw material was grounded by a hammer mill [1]. The grinding rate for biomass was $d_p \geq 500$ μm and bulk density was $\rho_n = 0.243$ g/cm^3 (for needles) and $\rho_n = 0.297$ g/cm^3 (for twigs with needles), respectively (Fig.1).

needles twigs with needles

Fig.1. *P. nigra* biomass.

2.2.2. Determination of moisture

The moisture content [5] for needles, twigs with needles and wheat bran biomass were determined as follows 7.10 ± 0.87%, 7.50 ± 0.13%, and 6.12 ± 0.81%.

2.2.3. Ash content

The mineral composition of the samples was 2.86 ± 0.12% (needles), 3.12 ± 0.03% (twigs with needles), and 6.64 ± 0.09% (wheat bran).

2.2.4. Determination of cellulose

The cellulose content of needles (29.20 ± 1.56%), twigs with needles (26.10 ± 2.18%), and wheat bran biomass (8.15 ± 0.12%) was determined according to the method described by Simitchiev et al. [1].

2.2.5. Extrudates obtaining

The experiments were carried out on a single-screw laboratory extruder "Brabender 20 DN" (Germany) with a working screw diameter of 20 mm, equipped with a measuring facility for torque M_n, N.m, the temperature of the material in the die and pressure inside the die. All data were obtained directly from the control unit display of the extruder (Fig. 2).

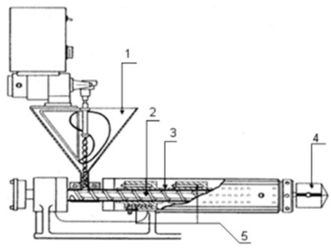

Fig. 2. Single-screw laboratory extruder "Brabender 20 DN".
1. Feeding device. 2. Screw 3. Cylinder 4. Die with nozzle and heater. 5. Heating devices
Cylinder diameter 20.05 mm
Cylinder length 406.5 mm
L/D 20:1
Cooling air, water

The obtained pellets are characterized by the following indicators:

Productivity of the extrusion process (Q_m), kg/h - is determined by calculating the mass of the amount of expired product per unit time according to the following equation:

$$Q_m = \frac{m}{t}, \tag{1}$$

where: m – mass of biomass, kg;
t – time, min.

Sectional expansion index (SEI) – was defined as the ratio of the average diameter of the extrudate from each experiment (D_e) to the diameter of the hole of the matrix nozzle (D_o) by the following equation:

$$SEI = \frac{D_e}{D_o} \tag{2}$$

where: D_e – the average diameter of the extrudate for ten samples, mm;
D_o – diameter of the die nozzle hole, mm.

Specific mechanical energy (SME), (kJ/kg) - determined by the following equation:

$$SME = \frac{\pi . M_n . n}{30 . Q_m} . 3{,}6, \tag{3}$$

where: M_n – torque, N.m. It is recorded from the display of the power supply unit of the laboratory extruder;

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n – the speed of the working screw, min^{-1};

Density of pellets (□), (g/cm^3) - is determined by measuring the mass and the average diameter of the same amount of extrudates (10 pcs.) of the same length. Calculate the volume of each extrudate, assuming that its shape is close to cylindrical. The density is calculated by the following equation:

$$\rho = \frac{M(1-\frac{W}{100})}{V-M\frac{W}{100}}, \tag{4}$$

where: M – the mass of biomass, g,
V – sample volume, cm^3,
W – the moisture of biomass, %.
The moisture of the products was determined by drying for 24 h at 105 °C.

Volumetric flow rate (cm^3/h) - determined by the following equation:

$$Q_v = \frac{Q_m}{\rho} . 10^3 \tag{5}$$

All experiments were performed with an initial humidity W = 10%, determined by drying for 24 h at a temperature of 105°□ (□□□ EN ISO 712:2010) [16].

3 Results and discussion

The main difficulty during extrusion processing was to reach optimal operating modes in order to obtain a product with the best physical and mechanical properties. Therefore, it was always necessary to perform preliminary experiments in which the basic parameters were changed during extrusion [3, 4, 17]. Needles and needle twigs contained resinous substances which melted at temperatures above 40°C. At the same time, the resistance moment and the pressure in the system increased and the molten product was not able to pass through the die. The same problem occured without additional heating in the three zones of the extruder. Due to the friction forces from the rotation of the screw in the cylinder of the extruder, the temperature increased again to the extent that the resin liquefied and the extruder closed [3, 4, 8, 9].

In order to prevent this negative phenomenon, wheat bran was added to the two black pine biomass (Fig. 3) with the following parameters: $d_p \geq 500$ □m, bulk density $□_n = 0.258$ g/cm^3. The biomass ratio of black pine: wheat bran is 1:1.

Fig.3. Wheat bran

New difficulties related to the clogging of the extruder confirmed the binding potential of the bran used [3, 4].

Another problem was the inner diameter of the nozzle through which the pellets come out. Preliminary experiments have shown that if this diameter was less than 8 mm, the product is not able to be extruded. The reason is that the inner hole of the die through which the product passes before reaching the nozzle is 8 mm in diameter. If the nozzle that was mounted to it was smaller, then the product encounters great resistance along the way. This leads to an increase in the resistance moment and the pressure in the extruder, accompanied by an increase in the coefficient of friction between the product and the nozzle. In order to reduce friction, experiments have been performed to increase the humidity of the input raw materials, but the results were unsatisfactory. After numerous experiments, operating modes have been established in which pellets with the best physical and mechanical properties were obtained:

Inner diameter of the nozzle was determined as D_o = 8 mm; the degree of compression of the screw was K = 5:1; the temperatures in the three zones of the extruder was t_1 = 30°C; t_2 = 30°C; t_3 = 40°C; the dosing screw speed was N_f = 100 min^{-1}; and the speed of rotation of the working screw was n = 250 min^{-1}.

The experimental preparation was performed at ambient temperature t_a = 20.5 °C. Five parallel experiments were performed in the modes of operation described above with a mixture of biomass from black pine needles and wheat bran (sample 1), twigs with needles, and wheat bran (sample 2) and only wheat bran (control sample). The results are presented in Table 1.

Table 1. Physical characteristics of the pellets.

	EI	Mass flow Q_m, kg\h	Specific mechanical energy SME, kJ/kg	Volume flow Q_v, cm^3/h	Density, ρ, g/cm^3	Pressure, MPa
Sample 1	1.02 ±0.005	2.187 ±0.016	1077.36 ±13.572	1701.95 ±66.212	1.285 ±0.018	3.6 ±0.112
Sample 2	1.06 ±0.025	3.123 ±0.046	1146.79 ±14.148	2248.38 ±102.014	1.389 ±0.026	4.3 ±0.132
Control sample	1.02 ±0.014	2.012 ±0.021	1311.60 ±16.162	1804.48 ±72.046	1.115 ±0.022	2.8 ±0.094

The data values are expressed as the mean ± SD (standard deviation (n = 5))

The obtained values for the individual parameters of the extrudates were not in agreement with previously reported [3, 4, 9], as the differences in the indicators of the initial biomass - a type of biomass, moisture of the raw material, and type of extruder used could be the reason for the differences. The data obtained in this study showed that the inclusion of wheat bran in the composition of the biomass improved the density, increasing the extruder's productivity.

Figures 4a, 4b, and 4c showed the images of the pellets obtained. They show the good smoothness, homogeneity, and density of sample 1 and sample 2. In contrast, the control sample showed an apparent porosity. There were also microcracks, which indicated poor sealing of the control sample at the exit and from the nozzle.

Fig. 4. Pellets obtained after single-screw extrusion of biomass from black pine and wheat bran: 4a - sample 1; 4b - sample 2; 4c – control sample.

The sectional expansion index indicated the increase in the size of the extruded product after it left the nozzle. Index values were greater than or equal to 1 (one) due to the transition from high pressure from the pressing of the product in the cylinder to atmospheric pressure at its exit after the nozzle. It was mainly influenced by the humidity and chemical composition of the extruded product and the temperature regimes, and the rotational speed of the working screw. The obtained results showed that the extruded pellets had low expansion. In this case, the low expansion index was not a negative phenomenon. Instead, it was an indicator of the products' good compaction in the matrix, expressed by the homogeneity and strength of the obtained pellets. Mixtures with twigs with black pine needles (sample 2) had a higher expansion index than those with black pine needles (sample 1) and wheat bran (control). It could be explained by the higher bulk density of sample 2, which allowed for better dosing of the product and led to a more optimal filling of the inter-turn space of the screw with the product and an increase in pressure.
The lower bulk density of the product (sample 1) led to problems with its dosing to the working screw. Therefore, the resistance moment and the pressure in the cylinder acquired lower values than those recorded in the experiments with sample 2. As a result, the productivity and the sectional expansion index of the obtained pellets were affected. A problem with dosing also occurred in the control sample, which affected both the productivity and the density of the pellets.

The density of the pellets is a major indicator on which their hardness and strength depend. It also characterizes the changes in the structure of the product during its movement in the cylinder of the extruder. During the extrusion of pure black pine biomass, the resulting extrudates decomposed when leaving the nozzle and were characterized by high friability and low density. The addition of wheat bran to the two black pine samples positively affected the quality and density of the pellets obtained. A parallel could be made with previous experiments for obtaining pellets from distilled juniper [1, 17]. The pellets obtained in the experiments with a mixture of wheat bran and black pine were characterized by three times higher density than those with a main component of juniper.

On the other hand, the pellets obtained after mixing biomass from black pine and wheat bran had a higher density than those produced with the control sample. Therefore, the presence of black pine biomass increased the quality of the final product while positively affecting both productivity and density. Biomass compaction systems were adapted depending on the moisture of the feedstock, the particle size, and the protein and lignin content, which had a very strong effect on the binding potential between the individual particles. The lignin fraction in the biomass determined the necessity to use technological solutions, such as the inclusion of steam, steam explosion, torrefaction in order to improve the quality of extrudates. The inclusion of natural or synthetic adhesives in the production of low lignin extrudates reduced the specific energy of compaction [17], while improving the index of density and strength, bulk density, moisture and calorific value of extrudates.

Volume productivity is an indicator that represents the ratio between the productivity expressed by mass flow to the density of the pellets. In the present study, the volumetric productivity of the pellets from sample 1 was about 25% higher than that obtained from sample 2. The total volumetric productivity of the product obtained from mixtures of black pine and wheat bran was lower than that obtained in pellets from distilled juniper [1, 3, 4, 16, 17]. It could be explained by the different extrusion modes and the higher density of the mixtures of black pine and wheat bran, due to which the speed of the pellets coming out of the nozzle was lower. Sample 2 had more than 8% higher volumetric productivity than the control sample due to the better filling of the screw inter-turn space with the product [17].

Productivity and energy consumption are essential technical and economic indicators that characterize each production process. In the present experiment, they had a rather indirect influence due to the type of extruder used to produce pellets in this study. Therefore, the calculated productivity was low, with relatively low energy consumption. The results showed a higher energy consumption when working with sample 2. This was mainly due to higher productivity (over 30% higher than sample 1), higher resistance moments, and a higher pressure during extrusion. When using an industrial turbo extruder operating at high pressure, the obtained productivity would be significantly higher, which was a prerequisite for further experiments in this direction. In the control sample, the energy consumption was over 12% higher than that obtained in the other samples. This was due to the lower pressure during the extrusion treatment, which affected the torque and the flow rate of the product from the nozzle of the extruder.

4 Conclusion

The results obtained in this study showed that the production of pellets produced by single-screw extrusion of biomass from needles and twigs with black pine was possible by mixing them with wheat bran in a ratio of 1:1. The addition of wheat bran improved the smoothness and homogeneity of the pellets and increased their density. Extrusion regimes have been established in which high-density pellets with a low sectional expansion index were produced. The productivity of extruding mixtures of twigs with black pine needles with wheat bran (sample 2) was 30% higher than that made from a mix of black pine needles and wheat bran (sample 1).

The pellets obtained after mixing the black pine biomass and wheat bran had over 13% higher density than those produced entirely from wheat bran. This showed that the presence of black pine biomass increased the quality of the final product while at the same time having a positive effect on both productivity and specific energy consumption. The experiments carried out in our study emphasized that the energy performance of the sample increased the quality of the final product. The energy characteristics of black pine biomass will be the subject of future research, because of the potential possibilities for application as an energy source.

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