



Article

Pelletization of Post-Harvest Tobacco Waste and Investigation of Flue Gas Emissions from Pellet Combustion

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Abstract: The paper presents the results of the pelletization (agglomeration) tests of post-harvest tobacco waste as a feedstock for fuel pellet production. The experiment was conducted on a prototype pelleting–briquetting device with a flat matrix. The influence of the tobacco waste moisture content (17, 21 and 25wt.%) and the rotational speed of the agglomerating rolls (120, 170 and 220 rpm) on the power demand and on the pellet's kinetic durability and density were determined. It was found that the moisture content has a significant impact on the pellet's density but slightly affects its kinetic durability. The obtained pellets were characterized by a high density above 1000 kg·m⁻³ and kinetic durability above 97%. In order to examine the exhaust composition, the obtained pellets were combusted in a 25 kW fixed great boiler. High amounts of CO, SOx, NO and HCl were obtained, which suggests that the selection of a different technology for tobacco waste pellet combustion should be made.

Keywords: pellets; combustion; bio-waste; emission; tobacco waste

1. Introduction

Today, the growing energy demand and the need for a significant reduction of greenhouse gas emissions are some of the main reasons for an intensive search for alternative, clean and cheap energy sources [1–3]. Increasingly, waste biomass is considered as an attractive solution for these requirements [3–5]. Popular research objects in this area are shells and husks [6,7], fruit stones [8,9], stovers [10], straw [11,12], pomace [13,14] etc. According to FAO (Food and Agriculture Organization of the United Nations) [15], in 2018, the worldwide production of tobacco reached over 6 million tonnes, of which 25% is considered as harvest and production waste [16,17]. Tobacco waste disposed in landfills may result in ground and water pollution, caused by several chemicals applied to the plant during its growth [18,19]. Therefore, the possibilities of its use as a substrate for thermal processes are being investigated. Tobacco stalks are one of the types of waste material obtained during the production of tobacco leaves [19]. Czerwińska et al. [20] reported that tobacco stalks are 40% cellulose; moreover, their mass constitutes up to 45% of the total plant dry mass over-ground. Cai et al. [21] investigated the carbonization behavior of tobacco stalks and hydrochar produced from tobacco stalks, by means of thermogravimetry, finding higher values of activation energy for hydrochars (caused by the higher contents of fixed carbons in hydrochars). Pyrolysis experiments with tobacco stalks conducted by Strezov et al. [22] showed that the biochars produced were particularly rich in potassium. According to Yang et al. [23], tobacco waste may contain more ash and nitrogen than other types of lignocellulosic biomass. During high-temperature processes, the nitrogen contained in fuels is emitted

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in the form of harmful NOx oxides [24]; a high ash content may contribute to slag deposition in the boiler, which hinders heat transfer and results in the accelerated wear of the metal elements of the installation due to corrosion [25]. This problem can be solved by using fuel mixtures of tobacco waste and higher-quality biomass or fossil fuels [26]. Cong et al. [27] confirmed that metal salts, which tobacco stalks are rich in, could work as catalysts in the co-combustion processes and improve the process efficiency. It should be emphasised that most of available research papers detail the problem of tobacco stalk thermal utilization with the use of instrumental methods, mainly thermogravimetry (TGA). The obtained results are important due to the valuable data received, which may become the design assumptions for combustion and pyrolysis installations [28]. In order to more precisely determine the suitability of a given fuel for its use in a particular installation, real-scale tests should also be carried out. The problem that arises during the energetic use of tobacco stalks is their low homogeneity, which primarily hinders the stability and automation of the thermal process. A solution enabling the use of agri-food production waste in prosumer and small-scale energetic installations is supplying them in the form of pellets [29]. The pelletization process, as a result of internal and external forces, allows obtaining a product in a solid geometric form. The material restrictions for the process are often a too-high [30] or too-low moisture content [6], high fat content [8] or large particle size [31]. In connection with the above, agri-food waste must be properly prepared before the pelletization process, e.g., by wetting, drying or adding binder materials. The operations preparing the material for pelletization must be optimized to obtain a high-quality product (pellet), with the possible reduction of the process energy consumption.

In reference to the above, Serrano et al. [32] assessed the mechanical strength, density, length and moisture of granules from barley straw and barley straw mixed with pine sawdust and reported that with the addition of pine sawdust, the pellet's kinetic durability increased. Mediavilla and colleagues [33], by compacting various mixtures of vine shoots and cork, found that with an increase in the addition of industrial waste cork to vine shoots, the energy consumption of granulation decreased, and the ash content decreased during the combustion of the obtained pellets. Nasrin et al. [34] examined the physical properties of briquettes from oil palm waste and concluded that the fragmentation and mixing of seed nests, fibers and grains improves the quality properties of the obtained briquette and at the same time decreases the briquetting process energy consumption. Stahl and Berghel [35] produced fuel pellets composed of sawdust and turnip waste. The turnip waste was obtained in the production of turnip oil. In the investigation, it was found that with an increase in the content of turnip waste, the pellet's mechanical durability and density decreased. Miranda et al. [36] characterized pellets made from grape pomace and found that up on the addition of Pyrenean oak, the durability and bulk density were improved. In other work, Miranda and colleagues [37] compacted olive pulp waste produced in the production of oil, and they found that the addition of Pyrenean oak waste to the olive pulp ensured a more effective granulating of the mixture and also improved the kinetic durability of the products. Obidziński et al. [38] claimed that tobacco waste most preferably should be pelletized at temperatures above 70 °C to obtain a high-density pellet with the optimal pelletizer's energy consumption. Reducing the power demand of tobacco waste pelletization is also possible with the use of a binder in the form of potato pulp. The starch contained in the pulp, under the high temperature of the process, gelatinizes and thus binds together the particles of the material; moreover, the moisture contained in it, acting as a lubricant, reduces friction in the holes of the pelletizer's matrix, directly affecting (decreasing) the energy consumption of the process.

The aim of the study was to determine the usefulness of post-harvest tobacco waste as a feedstock material for fuel pellet production. The scope of the work involved testing the influence of the material (moisture content) and process (rotational speed of compacting rolls) conditions on the density and kinetic durability of the obtained pellets and on the power demand of the pelletizing system. Moreover, the ultimate analysis, high heating value (HHV) and low heating value (LHV) of the obtained products were investigated. Subsequently, tobacco stalk pellets were combusted in a prosumer-scale installation, and the flue gas composition was determined.

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2. Materials and Methods

The research was performed according to the plan shown in Figure 1.

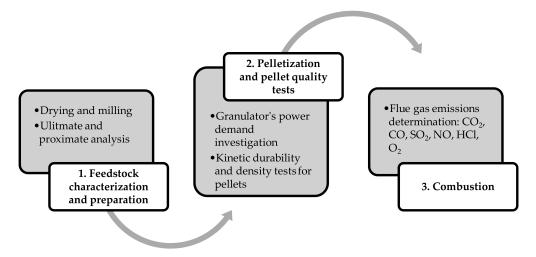


Figure 1. Scheme of the research.

2.1. Feedstock

The post-harvest tobacco waste used for the pelletization tests was obtained from a tobacco plantation placed in Suchowola, Poland (Figure 2a), and contained plant stalks, shoots, flowers and the leaves of leaf blades. Before processing, the material was pre-dried under atmospheric conditions by convection (Figure 2b) and subsequently ground in the POM H115 mill using 5 mm mesh sieves (Figure 2c).



Figure 2. Cont.

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Figure 2. Post-harvest tobacco waste (stalks): (a) after-harvesting tobacco, (b) during pre-drying and (c) milled and pre-dried (own source).

2.2. Methods

2.2.1. Feedstock Property Determination

The granulometric composition of the milled post-harvest tobacco waste was determined in accordance with PN-R-64798:2009 [39], with the use of a LPz-2e laboratory shaker (produced by MultiservMorek) and a set of sieves with mesh diameters of 6, 4, 2, 1, 0.5 and 0.25 mm, according to the methodology presented by Obidziński and Hejft [40]. A sample of 100 g was placed on the upper sieve. After 5 min, the residue on each of the sieves was weighed on a balance with an accuracy of 0.01 g. The analysis was repeated three times; as the final result, the arithmetic average was calculated.

Prior to the granulation process, the moisture content of the post-harvest tobacco waste was determined in accordance with PN-EN ISO 18134:2017 [41]. A WPE 300S moisture balance with an accuracy of 0.01% was used for the test. The analysis was repeated three times; as the final result, the arithmetic average was calculated.

The proximate analysis of the tobacco waste pellets (moisture [41], volatile matter [42], ash content [43] and fixed carbon by difference) was conducted by thermogravimetry using a Libra 209 F1 NETZSCH thermobalance. The ultimate analysis was performed on a CHN628 analyzer. The contents of carbon, hydrogen, nitrogen [44] and sulfur (dry basis (d.b.) %) [45] were determined by IR (infrared radiation) detection through high-temperature combustion; nitrogen (d.b.%) was analyzed by a catarometric method, which was also described in [46]. The chlorine content (d.b.%) was examined using the Mohr method [47], with the use of Eschka's mixture.

The investigation of the HHV (high heating value) and LHV (low heating value) was performed in accordance with PN-ISO 1928:2002 [48], with the use of a Precyzja-Bit KL-12Mn calorimeter. The HHV and LHV of the fuel were calculated in an automatic manner (according to the internal software managing the calorimeter). Having specified values of the fuel HHV and LHV (d.b.%), it was possible to calculate the calorific value of the material at another known moisture content [48]:

$$Q_i = \frac{100 - w}{100} Q_{i(d.b.)} - \frac{\gamma \cdot w}{1000} (\text{MJ·kg}^{-1})$$
 (1)

where the terms are defined as follows: Q_i (d.b.)—the HHV of the fuel (MJ·kg⁻¹), γ —the heat of water vaporization at 20 °C equivalent to a 1% moisture content ($\gamma = 24.55 \text{ kJ·kg}^{-1}$), and w—water vapor content in combustion gases (%).

Basing on the abovementioned abovementioned properties, the so-called fuel value index (*FVI*), which describes the pro-energetic value of biomass, was calculated for the pellets according to the formula [49]:

$$FVI = \frac{Q_i \cdot \rho_b}{1000 \cdot w \cdot A_d} (-) \tag{2}$$

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The range of the *FVI* is given in the results as the effect of the differences in the bulk densities of the pellets obtained: the lowest value stands for the lowest density.

2.2.2. Pelletization Process and Pellets' Quality Determination

In the present study, a prototype PROTECHNIKA (Poland) pelletizing–briquetting device [50,51] was used to perform the agglomeration tests. The pellet mill was provided with a 22 kW motor equipped with a recorder connected to a computer and a universal meter for taking the measurements of the power demand of the pelletizer in continuous mode. At 50 Hz, the rotational speed of the compacting rolls was 220 rpm, and a variable-frequency drive was provided to the device to vary the rotational speed of the rolls. The feedstock material was provided to the pelletizer's chamber by a Hydrapress screw feeder (0.75 kW) continuously.

Studies on the process of post-harvest tobacco waste pelletization were carried out at a constant material feed rate of $Q = 42 \text{ kg} \cdot \text{h}^{-1}$, gap between the matrix and rolls of h = 0.4 mm, die hole diameter of 12 mm and die hole length of 28 mm. Feedstock at moisture contents (w) of 17, 21 and 25% was agglomerated at rotational speeds (n) of 120, 170 and 220 rpm for each.

Quality tests were performed 24 h after pelletization. The particle density of the produced pellets was determined by measuring the height and diameter of 10 randomly selected pellets with a caliper with an accuracy of ± 0.02 mm. Next, their masses were determined by means of a WPS 360 analytic balance with an accuracy of ± 0.001 g. The density was calculated as the ratio of the mass and volume of the obtained pellets. The pellets' kinetic durability was determined in accordance with PN-EN ISO 17831-1:2016-02 [52] using a Holman tester, which cascades them in an air stream, causingthe pellets to collide with each other and the hard, perforated surfaces of the test chamber. The analysis was repeated three times for each sample type.

2.2.3. Combustion Tests and Flue Gas Analysis

The combustion of the produced pellets was carried out at the laboratory of Low Emission Combustion Technologies described in papers [8,53].

The stand includes a UnicaVentoEko boiler fabricated by Moderator and equipped with a retort grate (25 kW) and Dr. Födisch MCA10 flue gas analyzer. In the research, by a controlled automatic screw feeder, a sample of ca. 10 kg was fed into the boiler's combustion chamber. The fuel mass flow and the combustion airflow were selected by the boiler controller in Fuzzy Logic mode. The fuel mass flow was ca. $3.6 \text{ kg} \cdot \text{h}^{-1}$, and the airflow was given as a percentage of the fan performance; therefore, due to highly inaccurate calculations, an exact value is not provided here. The hot exhaust gas was collected from ca. 1.5 m above the grate and continuously analyzed by the MCA10. The contents of CO_2 , CO, NO, SO_2 and HCl and loss of O_2 were determined. The results are expressed in terms of normal conditions (1013.25 hPa, $0 \, ^{\circ}\text{C}$) and normalized to a $10\% \, \text{O}_2$ content according to the formula:

$$Z_{S2} = \frac{21 - O_2'}{21 - O_2''} \cdot Z_{S1} \qquad (\%, mg \cdot Nm^{-3})$$
 (3)

where the terms are defined as follows: Z_{s1} —the component content in the flue gas (%,mg·Nm⁻³), Z_{s2} —the component content in the flue gas for a given oxygen content (%,mg·Nm⁻³), O_2 ′—the set oxygen content in the flue gas (%), and O_2 ″—the obtained oxygen content in the flue gas (%).

In order to investigate the fuel–air conditions in the combustion chamber, the excess air factor λ was calculated as follows:

$$\lambda = \frac{21.5}{21.5 - O_2''} \tag{4}$$

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3. Results and Discussion

3.1. Feedstock Properties

The ultimate analysis of the tobacco waste samples were presented in Table 1. According to Wandrasz and Wandrasz [54], biomass (including waste), compared to coal, is characterized by high contents of oxygen and hydrogen. The high speed of hydrogen oxidation affects the acceleration of combustion and thus the emission of combustion products: increasing the hydrogen content in the combustion chamber results in a decrease in the CO and SOx contents in the exhaust gas and an increase in the combustion temperature [55]. Comparing tobacco waste to wood biomass, it has to be noted that the content of carbon is ca. 10% lower for tobacco [56], which directly influences its calorific value. After milling, the bulk density of the material was 213.5 kg·m⁻³.

Table 1. Physicochemical properties of tobacco waste pellets.

Parameter	Value
Proximate analysis (a.r.) (%wt.)	
Moisture	4.65
Volatile matter	64.54
Ash	14.67
Fixed carbon [†]	16.14
Ultimate analysis (d.b.) (%wt.)	
С	40.54
Н	6.00
N	1.28
S	0.262
Cl	0.087
O [†]	37.16
Metals (d.b.) $(\mu g \cdot g^{-1})$	
Cr	1.17
Mn	80.26
Fe	458.7
Ni	14.97
Cu	31.49
Zn	36.55
Pb	0.76
Al	2033
AAEMs ^{††} (d.b.) (μg·g ⁻¹)	
Ca	8391
Mg	3961
K	24.978
Na	132
Non-metals (d.b.) (μg·g ⁻¹)	
P	2359
HHV ^{†††} (a.r.) (MJ·kg ⁻¹)	15.18
FVI (-)	0.09-0.12

[†] By difference; ^{††} Alkali and alkaline earth metals; ^{†††} Calculated by using the Milne formula [57].

The current standard for the quality of non-wood pellets, ISO 17225-6 [58], does not reflect either recommended quality value for tobacco pellets. However, in the general requirements for so-called agropellets, the analyzed pellets slightly exceed the values for ash, Cu and Ni for the tested characteristics.

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On the other hand, the HHV and LHV (Table 2) of the analyzed waste should allow its successful use in combustion processes for heat or electricity generation. It is interesting, and also observed by other science units [23], that tobacco waste contains high amounts of nitrogen, which in high-temperature conditions (above 800 °C) may decompose to its oxide form.

Moisture Content (%)	HHV (MJ·kg ⁻¹)	LHV(MJ·kg ⁻¹)
0	18.02	16.02
6.71	16.81	14.78
10	16.22	14.17
20	14.42	12.33
30	12.62	10.48

Table 2. HHV and LHV of pellets from tobacco stalks.

The fuel value index, which takes into account the ash content, density, HHV and moisture content, describes the energy properties of a given fuel. In the case of tobacco stalk pellets, the FVI is similar to the FVIs calculated for similar types of agro-waste biomass such as cup plant fanpetals (0.08) or willow leaf sunflowers (0.11) [49]. Woody biomass is characterized by FVIs in the range of 0.6–1.29 [59]. The moisture content has a significant influence on the values of the heat of combustion and the calorific value of the tested pellets from tobacco waste (tobacco stalks). An increase in the moisture content from 0 to 30% results in a reduction of the HHV from 18.02 to 12.62 MJ·kg $^{-1}$ and the LHV from 16.02 to 10.48 MJ·kg $^{-1}$.

The influence of the tobacco waste moisture content w on the heat of combustion Q_s is described by the following equation:

$$Q_s = -0.18w + 18.017 \tag{5}$$

while the influence of the moisture content w on the calorific value Q_i of the tobacco waste is described as follows:

$$Q_i = -0.1845w + 16.016 \tag{6}$$

The obtained values of the HHV and LHV (Table 2) show that the tobacco stalkswere characterized by high-energy qualities. Similar results for the heating values for tobacco waste briquettes (tobacco stalks) were obtained by Peševski and colleagues [60]. The values of the tobacco waste heat of combustion are similar to those obtained by Obidzinski [61] for buckwheat hulls, Niedziółka and Zuchniarz [62] for barley straw, Maj and Piekut [63] for walnut leaves or Szyszlak-Bargłowicz et al. [64] for *Miscanthus giganteus*.

Plant-derived materials are highly heterogeneous in chemical structure and discontinuous in the distribution of mass concentrated in particles and grains. This, usually, determines the need to modify the material prior to the agglomeration process, i.e., give it an appropriate granulometric composition [34]. Figure 3 shows the results of tests on the granulometric distribution of the milled post-harvest tobacco stalks.

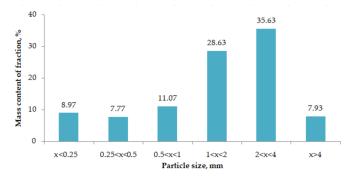


Figure 3. Grain size distribution of shredded post-harvest tobacco waste.

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Based on the granulometric test, it was found that the largest share, i.e., 35.6%, in the milled material was the 2 mm fraction. The next positions were taken by the fractions from the sieves with mesh sizes of 1 and 0.5 mm, which were 28.6 and 11.07%, respectively. The fractions of 0.25 mm and smaller (7.77 and 8.97%) made up a relatively large percentage, which might have had a negative effect on the pelletization process. According to Zawiślak et al. [65], substances with medium and fine grinding, i.e., in the range of 0.4–2 mm, are the most suitable for compacting. The diversity of the tested material fractions means they are highly susceptible to fractional separation. This property is directly affected by the particle size, which depends on the parameters of the milling process. Figure 4 presents a view of the individual fractions obtained during one of the performed granulometric distribution tests.

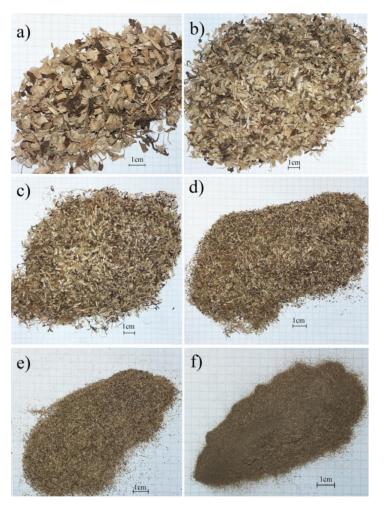
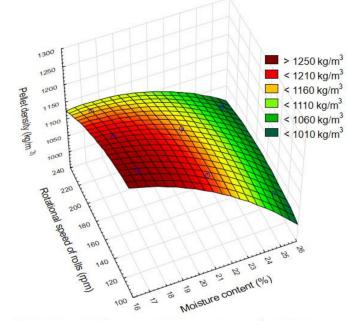


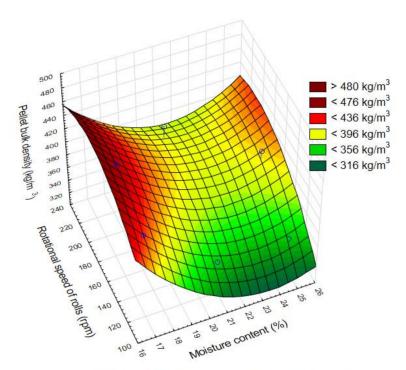
Figure 4. View of individual fractions of shredded post-harvest tobacco waste (own source): (a) 4 mm fraction, (b) 2 mm, (c) 1 mm, (d) 0.5 mm, (e) 0.25 mm and (f) <0.25 mm.

3.2. Pelletization Tests

Table 3 and Figure 5 present the results of the tobacco stalk agglomeration tests, where the pellet's density (ρ) , bulk density (ρ_b) and kinetic durability (P_{dx}) were analyzed and the pelletizer's demand for power (N) was investigated.

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 $\rho_b = 1386.07 - 107.46 \cdot n + 2.07 \cdot w + 2.19 \cdot n^2$

(b)

Figure 5. Cont.

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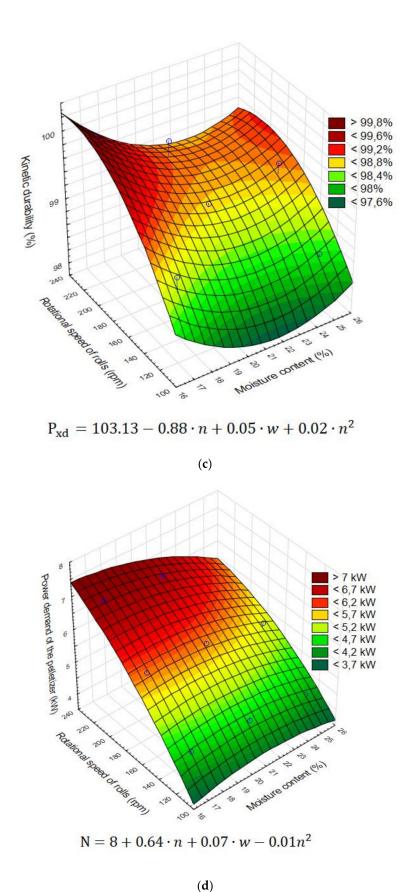


Figure 5. Influence of material and process factors (moisture content of waste and the rotational speed of the compacting rolls) on the (a) pellet density, (b) pellet bulk density (c) pellet kinetic durability and (d) power demand of the granulating system.

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Independent Variables		Dependent Variables				
	$x_1 = w (\%)$	$x_2 = n (rpm)$	$\rho(kg \cdot m^{-3}) \pm SD$	$\rho_b(kg\cdot m^{-3}) \pm SD$	$P_{dx}(\%) \pm SD$	N (kW)
1.	17	120	1260 ± 21.13	430 ± 0.73	98.7 ± 0.2	4.58
2.	21	120	1180 ± 23.56	350 ± 4.28	97.7 ± 1.2	4.52
3.	25	120	1060 ± 25.21	350 ± 9.77	98.2 ± 1.1	4.32
4.	17	170	1220 ± 32.06	460 ± 2.27	99.1 ± 0.2	5.91
5.	21	170	1180 ± 30.46	380 ± 2.66	98.8 ± 0.5	5.84
6.	25	170	1050 ± 34.13	410 ± 3.54	99.0 ± 0.5	5.53
7.	17	220	1160 ± 32.25	440 ± 0.31	99.6 ± 0.3	7.04
8.	21	220	1120 ± 36.21	410 ± 7.07	99.2 ± 0.1	6.91
9.	25	220	1050 ± 38.12	400 ± 5.26	98.8 ± 0.2	5.94

Table 3. Results of densification tests fortobacco waste.

From Table 3 and Figure 5a, it can be seen that as the moisture content of the tobacco waste increased from 17 to 25%, the values of the density decreased, which had an impact on the energy density of the fuel pellets. The highest decrease from 1260 to $1060 \text{ kg} \cdot \text{m}^{-3}$ was observed for the first case of tests, i.e., for the agglomeration speed of 120 rpm. When increasing the agglomeration speed, the observed density decrease was lower by ca. $90 \text{ kg} \cdot \text{m}^{-3}$ for the highest speed of 220 rpm.

On the basis of the performed tests (Figure 5a and Table 3), it was concluded that by increasing the speed of agglomeration, a decrease in the pellet's density is observed, especially for the lowest analyzed moisture content of 17%, where the decrease was from 1260 to 1160 kg·m⁻³. Furthermore, at higher agglomeration speeds and higher moisture contents in the feedstock, the impact on density was slighter. A moisture content above 20% does not significantly increase the granulation rate of the expansion that takes place after exit from the working system. The conducted analysis showed that the density of the tobacco waste pellets depends largely on the material moisture, $R^2 = 0.81$; no correlation was obtained between the pellet density and the pelletization speed. Mediavilla et al. [33] produced pellets from various mixtures of vine shoots and cork in a commercial pelletizer using a 20 mm flat matrix. The feedstock moisture content varied between 15 and 25%. They concluded that the addition of industrial cork residue lowered the process energy demanded, and during combustion, it lowered the ash content. The effects of the feedstock moisture content, its bulk density after a pre-agglomeration process, steam addition and the matrix temperature on the kinetic durability and bulk density of pellets from reed canary grass was reported by Larsson et al. [66]. They found that the moisture content was the factor most affecting the durability and bulk density of pellets. It was also confirmed for the tobacco stalk pellets from this study (Figure 5b and Table 3), where the bulk density was mostly correlated with the moisture content of the feedstock. As a result of the palletization process, the bulk density of the tobacco waste was increased from ca. 213.5 up to 460 kg·m⁻³ for pellets produced with a moisture content of 17% and a roll rotational speed of 170 rpm. However, the level of bulk density described by the norm ISO 17225-6 [57] (i.e., $600 \text{ kg} \cdot \text{m}^{-3}$) was not reached.

Increasing the moisture content from 17 to 25% did not significantly affect the kinetic durability of the produced pellets (Figure 5c and Table 3). The highest values of kinetic durability were observed for the lowest moisture content of 17%. Agglomeration speeds between 120 and 170 rpm did not significantly affect the $P_{\rm dx}$ value. However, at 220 rpm, the kinetic durability of the pellets decreased with an increase in the moisture content. Serrano et al. [32] evaluated the properties of pellets from barley straw: pellets were produced in an annular matrix pelletizer at a semi-industrial scale. They found that during pelletization, the optimum feedstock moisture contents were in the range of 19–23%. A kinetic durability value of 95.5% was obtained under these conditions, and was improved to 97–98% by the addition of 2–12wt.% pine sawdust. The conducted analysis showed that the kinetic durability of tobacco waste pellets has a slight correlation with the pelletization speed, $R^2 = 0.60$.

On the basis of the performed tests (Figure 5d and Table 3), it was found that increasing the rotational speed of the agglomerating roller system results in an increase in the pelletizer's power demand. For example, increasing the speed from 120 to 170 rpm (at 17% humidity) results in an

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increase in power demand by $1.33 \, kW$ (from $4.58 \, to 5.91 \, kW$). Increasing the feedstock moisture content caused a slight decrease in the power consumption, only at a rotational speed of 220 rpm; a noticeable effect of the moisture content was noted—an increase from 17 to 25% resulted in a decrease in the power demand by $1.07 \, kW$ (from $7.04 \, to 5.94 \, kW$). The lowest power demand ($4.32 \, kW$) was recorded at the lowest rotational speed ($120 \, rpm$) and the highest moisture content (25%). For the lowest value of moisture, there was an almost linear dependence of the power requirement on the engine speed. The power demand of the performed process mainly depended on the speed of the pelletizer working system, $R^2 = 0.88$. The feedstock moisture content did not affect the power demand of the device.

Based on the test results presented above, it was concluded that high-quality pellets were obtained in all the tested cases: a density above 1000 kg·m⁻³ and durability above 97%. However, changes in the process and material conditions particularly influenced the pelletizer's power demand, which decreased with an increase in the material's moisture and a decrease in the rotational speed of the rolls. Hence, it was concluded that with a constant process efficiency, the most favorable pelletization conditions were obtained using a roll rotational speed of 120 rpm and a material moisture content of 25%. According to [67], the process conditions for each material and device have to be validated to not reach a specific state where the pelletization efficiency is high but the product is characterized by small-sized particles and low durability values. In the case of tobacco stalks, there is an advantage of the possibility of drying them in the field (Figure 2) until they reachan adequate moisture content, which prevents the additional costs of pellet production [68].

3.3. Combustion Tests

Table 4 presents the averaged results for flue gas composition obtained during the combustion of the tobacco stalk pellets. The results were calculated to a 10% oxygen content in the flue gas, according to the requirements of the Ecodesign Directive [51]. For comparative purposes, Table 4 also shows the results of the flue gas emissions during the combustion of wood pellets at semi-constant heat and flow conditions for the boiler installation.

	Value			
Parameter	Tobacco Stalk Pellets	Industrial Wood Pellets [43]	Ecodesign Limitations	
CO ₂ (%)	3.52	7.27	-	
CO (mg·Nm ⁻³)	8243.28	329.87	500	
SO ₂ (mg·Nm ⁻³)	153.08	2.87	-	
NO (mg·Nm ⁻³)	224.34	50.85	200	
HCl (mg·Nm ⁻³)	44.79	0.00	-	
The actual oxygen concentration in the exhaust (%)	13.35	9.14	-	
λ (–)	2.63	1.74	-	
Average flue gas temperature in the boiler outlet (°C)	120	170	-	

Table 4. Flue gas composition and conditions of combustion of tobacco pellets.

During combustion, certain attention is given to the carbon monoxide emission, which is treated as a determinant of the presence of furans, dioxins, soot and hydrocarbons in the exhaust [51]. According to the Ecodesign Directive, the maximum CO content in the flue gas for boilers having a capacity below 0.5 MW cannot exceed 500 mg·Nm⁻³ [54]. The high value of CO emissions during the tobacco pellet combustion, reaching ca. 8243 mg·Nm⁻³, indicates problems in the CO to CO₂oxidation and poor stability of the combustion process [62]. The obtained low combustion temperature (Table 4)

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causes high emissions of both nitrogen oxides and hydrogen chloride. Nitrogen oxides are mainly formed from the oxidation reactions of chemical compounds containing nitrogen in the fuel. Naturally, the NOx emissions tend to increase as the nitrogen content in the fuel increases [24,56]. Moreover, the decrease in the combustion temperature causes an increase in NO emissions, which according to researchers is the result of changes in the course of NO synthesis under these conditions [24]. It should be noted that the Ecodesign [51] standard provides a maximum value of NO emissions at a level not exceeding 200 mg·Nm $^{-3}$. Currently, European emission regulations do not limit emissions of HCl and SOx. Certified wood pellets combusted in the same installation generated SOx emissions at the level of ca. 23 mg·m $^{-3}$. Another tested combustion parameter is the excess air factor λ , which describes the contact area of the oxidizing agent and the fuel particle; the lower the value obtained, the closer the process parameters are to stoichiometric combustion (where $\lambda = 1$ stands for stoichiometric combustion). Agri-food waste is often characterized by high λ values due to the higher ash content, which impedes effective combustion. The above-described studies indicate that pellets produced from tobacco stalks require further research for an appropriate method for their thermal utilization.

4. Conclusions

This work outlines the (i) physical and chemical properties of tobacco stalks and tobacco stalk pellets, (ii) pelletization process for post-harvest tobacco waste in the form of stalks, and (ii) combustion properties of the obtained fuel pellets. The ultimate analysis of the post-harvest tobacco waste showed that it is similar to the wood biomass used in industrial pellet production. The differences are clearly notable in the results of proximate analysis, where tobacco stalks were characterized by a high ash content, which during the combustion processes, may decrease its effectiveness. Detailed studies about the calorific value, and the effect of moisture on the HHV and LHV were conducted. It was confirmed that increasing the moisture content from 0 to 30% causes a 30% reduction in the high heating value and the low heating value. During the pelletization process, increasing the moisture content of the raw material caused a decrease in the pellet density. The biggest decrease in density was observed in pellets produced at the lowest analyzed agglomeration speed of 120 rpm. As the roll speed was increased, the differences in density were lower. Moreover, increasing the moisture content and the agglomeration speed slightly affected the kinetic durability of the obtained pellets and decreased the power demand of the pelletizer. The results for tobacco waste pellet combustion, where high emissions of CO and NO were obtained, indicate a need for additional analyses of their suitability as a solid fuel and the selection of other combustion technologies or conditions than those used in this paper.

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Nomenclature

Symbol	Description
LHV	lower heating value (MJ·kg ⁻¹)
HHV	higher heating value (MJ·kg ⁻¹)
w	moisture content (wt.%)
n	pelletizer's matrix rotational speed (rpm)
Z_{s1}	the actual chemical content in the exhaust gas (%,mg·Nm ⁻³)

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Z_{s2}	content of the chemical compounds in the exhaust gas for a given oxygen con	tent (%, mg·Nm ⁻³)

- O_2 ' set value of the oxygen content in the exhaust (%)
- O₂" actual oxygen content in the exhaust gas (%)

FVI fuel value index

 λ excess air factor (-)

ρ pellet's density (kg·m⁻³)

 P_x pellet's kinetic durability (%)

N granulator's power demand (kW)

wt.% weight percent a.r. as received

d.b. dry basis

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