Evaluation of Urban Tree Leaf Biomass-Potential, Physico-Mechanical and Chemical Parameters of Raw Material and Solid Biofuel

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Abstract: The paper presents the results of research aimed at evaluating the possibility of using selected tree leaf species to produce solid biofuels. The possibility of production of qualitative solid biofuels from urban tree leaves meets the expectations of the municipal sector. Collection of tree leaves in urban areas is very often necessary for road safety reasons, the need to collect biomass rich in dust and pollution as well as biomass infested with pests. The production of solid biofuels from tree leaves allows for effective management of this raw material with energy recovery. The performed research indicates such a possibility, and the obtained ash is used as a soil improver. The conducted research showed that the biomass of leaves of five tree species used in the experiment can be a source of raw materials for production of qualitative biofuels. The obtained pellets were characterized by properties comparable to those of classical wood pellets. The lower heating value of the obtained pellets ranged from 14.5 to 15.5 MJ·kg⁻¹. Physical properties of the obtained pellets described by bulk density (BD 600–660 kg·m⁻³), mechanical durability (DU 90–96%), moisture (Mₘ₉ 10–12.5%) indicate that these products can be used in existing combustion equipment. Preliminary analysis of the obtained ashes (determination of ash melting point, bulk density) indicates that they should not cause difficulties in ash removal systems from the combustion chamber.

Keywords: tree leaves; pellets; bulk density; mechanical durability; self-ignition temperature; ash melting; compaction degree; biodegradable waste

1. Introduction

The share of trees in urban greenery is very important for its qualitative evaluation. Therefore, large trees in cities are extremely valuable. Tree crowns play a very important role in regulating the microclimate of urban zones [1,2]. They perform “physical” functions as barriers against solar radiation and wind for the surface of building elements, such as roads and buildings. Large trees in streets, parks and playgrounds increase the active area of solar radiation absorption above urban traffic. Exposed to sunlight, the crown of the tree becomes an active surface that absorbs energy, thus protecting the surface of the sidewalk and people on it, as well as the walls of neighboring buildings. The mass of leaves, boughs and branches effectively limits heat transfer, and the intensive transpiration of green mass reduces the temperature of the upper layers of the tree crown [3–5]. Thanks to the low conductivity of individual parts of the tree, the heat is not transferred to the roots. Such a climatic effect is not achieved with the use of low greenery (e.g., shrubs).

The trees are distinguished by much more intensive CO₂ sequestration and removal of pollutants from the air per unit of occupied area than low vegetation. The most efficient are the street trees growing in the most polluted environment, absorbing up to three or
four times more pollutants than in places with cleaner atmospheric air [6–9]. As numerous research [3,10,11] show, tree leaves, like bark, are able to accumulate particulate matter (PM) in amounts of about 9–75 µg·cm⁻². Urban trees are an important element of intercepting and delaying rainwater runoff, which significantly reduces flooding and erosion and pollution of rivers and water bodies [12–14].

The above mentioned functions of trees performed in urban agglomerations cause that the leaves are characterized by an increased content of mineral fractions. By cyclic collection of falling leaves from trees there is a possibility to reduce dusts in urban space. Additionally, tree leaves are very often infested by fungi, therefore their removal from green areas is often very important [15–17]. In some cases, it is advisable to harvest and destroy the leaves (mostly by burning) to eliminate overwintering pests, e.g., horse chestnut leaf-miner (Cameraria ohridella Deschka & Dimić) [18,19].

In addition to classic composting, tree leaves can be subjected to combustion processes [20,21]. In this way we effectively reduce the microbiological activity of the leaves with energy recovery. The tree leaves can also be subjected to thermal processes (the torrefaction) aimed at improving the energy quality. Torrefaction of tree leaves makes it possible to obtain a higher calorific value of the product (17.70–22.42 MJ·kg⁻¹) with a weight drop of about 20–30% [22,23]. In addition, the process temperature in the range of 200–300 °C effectively disinfects the obtained product which can be used not only for fuel purposes but also as fertilizer [24,25].

Logistic management of tree leaves is very difficult due to the low bulk density. Therefore, it is recommended to process the leaves by grinding or pelletizing in order to reduce transport or storage costs. This reduction can be described by compaction degree ID defined as relation of granules density to the bulk density of raw material [26]. In the case of different biomass ID is about: 4.01 for onion husk [27], 2.15 for tobacco waste [28] or 2.1 for reed canary grass [29] and 2.6–4.1 for different wood species [30]. Probably the similar or higher values can be expected for leaf biomass.

Research conducted by Pradhan et al. [31] on the processing of garden waste biomass (leaves, branches) showed similar dependencies as in the processing of typical wood biomass. They showed, among other things, that with the increase in the degree of biomass fragmentation the yield and quality of obtained pellets increases. The highest quality of pellets (bulk density BD = 609 kg·m⁻³, mechanical durability DU = 95.0%, specific density DE = 1310 kg·m⁻³) was obtained from raw materials comminuted on the smallest sieve with a mesh size of 6.35mm. These studies indicate that the processes of biomass densification (pelletization, briquetting) can significantly help in the management of waste from greenery maintenance, especially from a logistics point of view [32]. Many biomass types, if they have proper parameters, are considered for biofuel production [33–36]. Leaves can also be a valuable raw material for this purpose [37].

The processes of thermal processing of herbaceous biomass generate significant amounts of waste in the form of ashes. Biomass ash has a suitable chemical composition for further use. The amount of ash generated during combustion depends on the origin of the fuel and ranges from about 1% (wood biomass) to over 10% (agro biomass) [38]. Ash rich in silica can be used as an additive, so it can be successfully used in the ceramic and cement industry [39,40]. It can also be used as a raw material or component of granulated fertilizers [41].

In order to be able to properly design the processing of tree leaves it is necessary to know the basic physicochemical properties. Knowing the physical characteristics associated with the raw material, the final product and changes in the most important parameters during the application of technological processes is essential to apply optimal technical solutions [42–46]. Determining the basic physical properties of raw materials and final products is crucial for the proper design of further processing processes such as transport, storage or combustion [47,48]. Therefore, this research aimed to initially assess the usefulness of selected tree leaf species for processing into solid biofuels. The results of these
studies should indicate further directions of research on effective treatment of residues from the municipal management.

2. Materials and Methods

Research conducted to date on the energetic use of residues from landscape maintenance were focused on mixtures of various leaves and branches. There is a lack of information on the variability of key properties of raw materials and solid biofuels produced from leaves of different tree species. Therefore, the presented research includes analyses of properties relevant to leaf pellet production and combustion processes.

The research was conducted using the leaves of 5 tree species very common in urban planting. The material was obtained in November 2019 on the campus of the University of Agriculture in Cracow (50.07907, 19.86413). Tree leaves from the grassy area were raked with hand tools and placed in containers (containers with mesh walls, capacity 1 m³) using snow shovels. The weather in the period of 1 week immediately before harvesting leaves was rainless, sunny/moderately cloudy. In the research the leaves of the following tree species were used:

- Weeping golden willow (*Salix × sepulcralis* ‘Chrysocoma’) — code: Salix,
- Norway maple (*Acer platanoides* L.) — code: Acer,
- Common oak (*Quercus robur* L.) — code: Quercus,
- London plane (*Platanus acerifolia* (Aiton) Willd.) — code: Platanus,
- American sweetgum (*Liquidambar styraciflua* L.) — code: Liquidambar.

Three containers of leaves were collected from each tree species and transported to the heated laboratory hall for natural drying. After a period of about 4 weeks, the material was subjected to scheduled analyses.

The research plan included the performance of the following tests and analyses:

- determination of the chemical composition of the raw materials,
- determination of moisture content of the raw materials and the produced pellets,
- determination of characteristic densities of raw materials and pellets,
- pelletization tests (including raw material preparation),
- determination of ash content in the obtained pellets,
- determination of characteristic ash melting temperatures,
- determination of self-ignition temperature of the obtained pellets,
- determination of mechanical durability of the pellets,
- determination of calorific value of the obtained pellets.

2.1. Determination of the Chemical Composition of Raw Materials

Laboratory samples were formed by reducing the collective sample weight. Laboratory samples of leaves were dried in a temperature of 65 °C, homogenized and subjected to wet mineralization in a closed system by means of microwave energy. A microwave system made by Anton Paar Multivawe 3000 (Anton Paar GmbH, Graz, Austria) was used for mineralization. Leave samples were mineralized in a mixture of nitric acid solution (V) and dihydrogen dioxide at a volume ratio of 1:3. Weight of the analytical test portion was max. 0.5 g of dry weight.

Concentration of the elements under analysis in the obtained solutions was determined by means of the atomic emission spectroscopy method, using an Optima 7600 instrument (Perkin Elmer, Waltham, MA, USA). The lengths of waves, by means of which the concentration of the tested elements and limits of determination with regard to the methods used were determined, are presented in Table 1. To control the accuracy of elements analyses, a certified reference materials, IAEA-V-10, was used. Table 1 shows the results of reference material analyses and the recovery value, on the basis of the analyses performed in four repetitions.
Table 1. Parameters of the analysis method.

| Parameters | Wavelengths (nm) | Limit Detection (mg dm⁻³) | Content in Cert. Material (mg kg⁻¹) | Measured (mg kg⁻¹) | Recovery (%) |
|------------|-----------------|---------------------------|--------------------------------------|------------------|-------------|
| Ca         | 317.933         | 0.01                      | 21,600                               | 20,896           | 97          |
| Cd         | 228.802         | 0.0027                    | 0.03                                 | 0.0305           | 102         |
| Cr         | 267.707         | 0.0071                    | 6.5                                  | 6.860            | 106         |
| Cu         | 327.393         | 0.0097                    | 9.4                                  | 10.01            | 106         |
| Fe         | 238.204         | 0.0046                    | 185                                  | 168.5            | 91          |
| K          | 766.490         | -                         | 21,000                               | 20,740           | 99          |
| Mg         | 285.208         | 0.0016                    | 1360                                 | 1423             | 105         |
| Mn         | 257.608         | 0.0014                    | 47                                   | 44.83            | 95          |
| Na         | 589.592         | 0.069                     | 500                                  | 472.4            | 94          |
| Ni         | 231.604         | 0.0151                    | 4                                    | 4.359            | 109         |
| P          | 213.617         | 0.076                     | 2300                                 | 2248             | 98          |
| Pb         | 220.353         | 0.0425                    | 1.6                                  | 1.508            | 94          |
| Zn         | 206.200         | 0.0059                    | 24                                   | 25.32            | 106         |

2.2. Determination of Moisture Content

Determination of the moisture content of the tested materials was performed in accordance with the methodology included in the PN-EN ISO 18134-1 standard [49]. Samples of whole leaves, after grinding as well as pellets were dried in the laboratory dryer (SLW 115, Pol-Eko, Wodzisław Śląski, Poland) at 105 °C. The samples were dried to a constant weight. Preliminary studies showed that the drying time of the samples to constant weight was 12 h. The formula was used to determine the moisture content:

\[ M_{ar} = \frac{m_b - m_c}{m_b - m_a} \times 100\% \]  

where: \( M_{ar} \)—the moisture content of the sample (%); \( m_a \)—the mass of the empty crucible (g); \( m_b \)—mass of the crucible with the sample in the analytical state (g); \( m_c \)—mass of the crucible with the sample after drying (g).

2.3. Determination of Bulk and Specific Density

The determination of the bulk density BD of the tested raw materials was performed in accordance with PN-EN ISO 17828 [50]. The following measuring vessels were used to measure the bulk density of individual materials:

- 50 dm³—for whole leaves,
- 5 dm³—for shredded raw materials and pellets,
- 0.1 dm³—for ash.

After stabilizing the pellet samples, their solid density DE was measured using a quasi-fluid pycnometer (GeoPyc 1360, Micromeritics Instrument Corp., Norcross, GA, USA), the details of which are described in previous work [51].

On the basis of the obtained results of the bulk densities, the three types of compaction degree ID—was calculated. Compaction degree after milling IDm, compaction degree after pelletizing IDp in relation to the raw material after milling and absolute compaction degree after pelletizing IDa in relation to the raw material before milling. The general formula of ID is based on the BD of the material after the process to the BD of the raw material before the process:

\[ I_D = \frac{BD_A}{BD_B} \]  

where: \( I_D \)—compaction degree; \( BD_A \)—bulk density of material after the process kg m⁻³; \( BD_B \)—bulk density of raw material before the process kg m⁻³.
2.4. Process of Shredding and Pelletization of Tested Raw Materials

After harvesting, the tree leaves were placed in openwork containers and subjected to the process of seasoning. This process was carried out in a heated laboratory hall, where after a period of about 20 days the moisture content of raw materials was obtained at the level of 12–14%.

The material prepared in this way was first subjected to the process of grinding. This process was carried out on the SYNERGIA BIO type MU500 hammer mill (ANT+ Grzegorz Szewczyk, Poznań, Poland) dedicated to the grinding of herbal biomass. The mill equipped with a sieve with a 10 mm hole diameter. Installed engine power was 11 kW, working rotor speed 2800 rpm.

Then the raw materials crushed on the mill were subjected to the process of pelletization. This process was carried out on a semi-industrial pelletizing machine (MGL200, KovoNovak, Czech Republic). For the agglomeration matrices with holes of 6 mm and a compression factor of 4.2 were used. The power of the main engine of the pelletizing system was 7.5 kW. The device is equipped with a system for sieving and cooling the obtained pellets. The final temperature of the pellets did not exceed 35 °C. The obtained pellets, before further quality assessment tests, were stabilized for 48 h.

2.5. Determination of Ash Content and Melting Temperatures

The determination of ash content in the tested materials was performed in accordance with the methodology of EN ISO 18122:2015 [52]. The ashing process was carried out in the muffle furnace (Czylok, Jastrzębie-Zdrój, Poland) at 550 °C.

Ash melting temperatures were also determined as a parameter affecting the correct course of the combustion process. From the ash sample, prepared in accordance with the methodology of ash content determination (EN ISO 18122:2015), melting lozenges were made in accordance with the methodology contained in PN-ISO 540:2001 [53]. The process of determination of characteristic ash melting temperatures was performed in a tube furnace (Czylok, Jastrzębie-Zdrój, Poland) equipped with a special camera for recording thermal processes. Figure 1 shows examples of images subjected to analysis registered during the melting process.

![Figure 1. View of sample photos recorded during the ash melting process.](image-url)
2.6. Relative Self-Ignition Temperature

The measurement of the Relative self-ignition temperature of tested samples (RSIT) of the tested materials was performed in accordance with the guidelines contained in the Commission Regulation (EC) No 440/2008 [54]. This Regulation defines the method together with the apparatus equipment to determine the auto-ignition temperature of solids, i.e., the ambient temperature expressed in °C at which the analyzed sample will be ignited. All measurement experience was carried out on the test stand of Czylok (Jastrzębie-Zdrój, Poland).

In the measuring chamber of the stand, in which the sample container was placed, there are two thermocouples. One records the temperature inside the heating chamber and the other records the temperature in the mass of the analyzed sample. The heating of the sample in the measuring chamber was performed at a speed of 0.5 °C·min\(^{-1}\). The measurement was performed until the temperature of the sample reached 400 °C. The self-ignition temperature was calculated according to the procedure presented in Figure 2. The measurements were taken in three repetitions for each analyzed sample and then the results were presented as an average with standard deviation.

![Figure 2](image_url)  
*Figure 2. Procedure for determining the auto-ignition temperature of a sample based on test data.*

2.7. Mechanical Durability of Pellets

Pellets obtained from the tested raw materials were subjected to the assessment of crushing in a chamber test. The test was conducted in accordance with the PN-EN ISO 17831-1 standard [55]. This test describes the resistance of the tested granules to mechanical destructive factors related to logistics.

According to the procedure of the standard, the test was performed in two repetitions, where the prepared sample of 500 ± 10 g was placed in the rotating chamber of the tester and tumble for 10 minutes (500 revolutions) at 50 ± 2 rpm. At the end of the test, the fine particles were separated on a #3.15 mm sieve and the mass of the remaining pellet was related to the mass of the original sample according to the relationship:

\[
DU = \frac{m_A}{m_E} \cdot 100
\]

where: DU is the mechanical durability (%); \(m_E\) is the mass of the sieved pellets before the tumbling treatment (g); \(m_A\) is the mass of the sieved pellets after the tumbling treatment (g).
2.8. Calorific Value of Pellets

One of the most important energy parameters which is the lower heating value (LHV) \( q_{p,\text{net,ar}} \) (J·kg\(^{-1}\)) was also determined. This parameter allows to determine the amount of energy that can be obtained during the combustion. The measurements were made with an IKA C6000 (IKA-Werke, Staufen, Germany) automatic calorimeter. The calculations were made in accordance with PN-EN-ISO-18125_2017-07E standard [56].

3. Results and Discussion

Laboratory experiments on processing of tree leaf biomass allowed to collect parametric data. The collected data are presented below, together with their evaluation in the context of literature data.

3.1. Chemical Analyses of the Investigated Materials

From an environmental point of view, biomass is one of the best energy sources. However, its use for direct combustion is associated with certain technological difficulties. One of the most frequently defined problems in the combustion of biomass is the high content of alkaline elements. During thermal biomass conversion processes, onerous compounds are formed that limit the efficiency of combustion and heat exchange processes. The parameters that deteriorate the quality of biomass for combustion are the high content of sodium, potassium as well as calcium and magnesium [57,58]. The results obtained in own studies indicate a high variability of the chemical composition of the analyzed materials. The lack of repeatability of biomass intended for fuel combustion is one of the most frequently defined disadvantages of this material [59]. From the point of view of the use for energy purposes, the greatest usefulness was found in the case of Liquidambar, Quercus and Platanus. The potassium contents in these materials were 2713 mg·kg\(^{-1}\), 3967 mg·kg\(^{-1}\) and 3924 mg·kg\(^{-1}\), respectively (Table 2). The content of this element in Salix and Acer leaves was more than twice as high. In the case of sodium, the highest amounts were found in Salix leaves, while the lowest in Liquidambar. In terms of the suitability of the tested materials for combustion processes, the leaves of Liquidambar and Platanus turned out to be the best, while the worst properties were characteristic for Salix leaves [60]. The content of heavy metals in the analyzed materials does not indicate anthropogenic enrichment. Ashes from biomass combustion can be used as fertilizers [61].

3.2. Characteristics of Raw Materials and Obtained Pellets

The results obtained concerning the characteristics of tree leaves as well as the obtained pellets showed that this material is a potential source in the production of biofuels (Table 3). However, it should be noted that it is not a source of raw material for production of high quality pellets. The biggest disadvantage of this material is the increased ash content. The highest ash content values were registered in willow (Salix) leaves and amounted to \( A_{\text{ar}} = 20.12\% \). This significantly increased value could be associated with the possibility of collecting the leaves together with the soil and sand fractions. Small willow leaves are exposed to soil contamination in the autumn period.
Table 2. Average values and standard deviations (±) of the determined elements in the analysed materials (mg kg⁻¹).

| Element | Salix | Acer | Quercus | Platanus | Liquidambar |
|---------|-------|------|---------|----------|-------------|
| Ca      | 15,435.7 | 12,678.5 | 14,407.9 | 11,613.5 | 9720.8     |
| ±       | 1722.5 | 996.5 | 1098.6 | 896.8 | 1102.5 |
| Cd      | 0.9426 | 0.1947 | 0.1254 | 0.1988 | 0.3612   |
| ±       | 0.0723 | 0.0282 | 0.0663 | 0.0234 | 0.0411 |
| Cr      | 20.231 | 10.742 | 8.332 | 13.342 | 5.250     |
| ±       | 1.5226 | 2.492 | 1.729 | 1.509 | 0.729   |
| Cu      | 4.640 | 4.189 | 4.021 | 2.886 | 2.172     |
| ±       | 0.753 | 0.581 | 0.277 | 0.439 | 0.128   |
| Fe      | 854.1 | 395.8 | 426.3 | 446.1 | 330.1     |
| ±       | 125.5 | 28.4 | 58.63 | 27.62 | 38.53   |
| K       | 6574.2 | 6317.1 | 3967.6 | 3924.0 | 2713.9 |
| ±       | 483.5 | 594.7 | 411.9 | 272.9 | 496.7   |
| Mg      | 904.4 | 1021.7 | 1292.1 | 1175.5 | 1473.0 |
| ±       | 192.6 | 138.7 | 100.9 | 168.2 | 211.6 |
| Mn      | 81.47 | 312.57 | 93.36 | 73.49 | 220.03 |
| ±       | 14.77 | 48.23 | 9.16 | 14.91 | 36.8 |
| Na      | 36.235 | 21.330 | 35.614 | 29.918 | 18.488 |
| ±       | 2.893 | 1.728 | 2.83 | 3.628 | 2.466 |
| Ni      | 1.2529 | 0.5845 | 0.6754 | 0.6386 | 1.4686 |
| ±       | 0.096 | 0.0709 | 0.0842 | 0.0859 | 0.1163 |
| P       | 1038.3 | 1479.1 | 1185.12 | 801.9 | 653.6 |
| ±       | 121.8 | 163.4 | 96.85 | 66.82 | 94.11 |
| Pb      | 3.352 | 2.272 | 2.029 | 1.590 | 1.488 |
| ±       | 0.427 | 0.339 | 0.173 | 0.186 | 0.118 |
| Zn      | 120.36 | 87.98 | 38.22 | 31.58 | 59.23 |
| ±       | 9.419 | 11.62 | 6.73 | 4.48 | 9.16 |

The leaves of other tree species used in the experiment are characterized by a different morphological structure (wide leaves with raised edges) which reduces the risk of significant contamination. The lowest ash content (more than twice as low as that of willow) was determined for the leaves of London plane (Platanus) A亢= 9.76%. The remaining materials were characterized by ash content in the range 10–12%. Similar results were obtained in studies carried out by García-Maraver et al. [62] on olive tree residues. These studies showed that the leaves of these trees contained ash fractions in the range 8.23–14.4%. It was also shown that with the storage period, the concentration of mineral fractions in the material increases and is likely to be mineralized (biodegraded). Similar results were obtained with the analysis of waste biomass from the extraction of wood from forests or from the processing of agricultural raw materials [63–65]. Wood biomass without leaves and bark is characterized by significantly lower ash values of 0.1–2% [66,67].

The tree leaf biomass is also characterized by a very low bulk density. The tree species tested have a leaf density of 12–46 kg·m⁻³. Such a low density is a crucial problem in the management of raw materials. The costs of transport or storage may significantly reduce the profitability of the projects. A certain solution could be the preliminary fragmentation of raw materials at the time of harvesting. The conducted research showed that grinding the leaves on a flail mill allows to increase the bulk density of the raw material even by 500–800%. Studies conducted by Jewiarz et al. [68] on milling processes have shown that milling (beater system) of material through a 6 mm sieve and then through a 2 mm sieve allows to increase the bulk density of Giant miscanthus (Miscanthus × giganteus Greef et Deu) by 191%, while European beech (Fagus sylvatica L.) by 135%. This study indicates that the milling process is a treatment for improving prologistic properties.
The highest bulk density, after preliminary grinding, was characteristic of American sweetgum (Liquidambar) leaves ($BD_{ar} = 144.9$ kg·m$^{-3}$). Other materials were characterized by a density in the range of $93–121$ kg·m$^{-3}$. These values are similar to the density of ground herbaceous biomass used for energy purposes [67,69–71].

The moisture content of collected tree leaves is characterized by low level. The highest moisture content was characterized by willow leaves (Salix) $M_{ar} = 32.8\%$, and the lowest by London plane (Platanus) $M_{ar} = 23.4\%$. This moisture content strongly depends on local weather conditions and the exposure of the area between the trees to sunlight. It should be emphasized that the leaves of trees in urban conditions can be obtained, in appropriate periods of autumn, with relatively low moisture, which significantly facilitates the processing.

The bulk density ($603–665$ kg·m$^{-3}$) and specific density (above $1100$ kg·m$^{-3}$) are similar to the level of typical wood pellets used in the power industry [51,72].

This is probably due to the increased value of mineral fractions. These results confirm only that raw materials based on tree leaves are susceptible to agglomeration processes.

The mechanical durability test of pellets showed that granules made from American sweetgum (Liquidambar) characterized by the highest value of the DU index (96.56%), similar values were obtained for willow (Salix) DU = 90.3%. These values are satisfactory, given that these granules should be used as industrial pellets or for soil applications.

The calorific value of the tested pellets is satisfactory. Considering the relatively high ash content, it can be concluded that they constitute an interesting source of fuel for industrial and municipal sector units. The highest LHV value (15567 J·g$^{-1}$) was characteristic for pellets made from American sweetgum (Liquidambar) leaves, while the lowest value (LHV = 14502 J·g$^{-1}$) was obtained for willow (Salix). These values are comparable with agricultural raw materials obtained from other production/processing residues [35].

The obtained $I_D$ values mean that the milling process, and especially the pelleting process in the case of leaves, will significantly reduce the costs associated with logistics operations.

| Characteristic                     | Salix         | Acer          | Quercus       | Platanus      | Liquidambar  |
|-----------------------------------|---------------|---------------|---------------|---------------|--------------|
| Moisture after harvesting $M_{ar}$ (%) | $32.8 \pm 0.3$ | $25.4 \pm 0.2$ | $23.2 \pm 0.2$ | $23.4 \pm 0.2$ | $26.3 \pm 0.2$ |
| Moisture after grinding $M_{ar}$ (%) | $13.6 \pm 0.2$ | $12.6 \pm 0.2$ | $13.1 \pm 0.2$ | $14.2 \pm 0.2$ | $13.8 \pm 0.2$ |
| Bulk density of whole tree leaves $BD_{ar}$ (kg·m$^{-3}$) | $46.1 \pm 2.6$ | $11.5 \pm 0.6$ | $28.2 \pm 1.4$ | $12.2 \pm 0.8$ | $22.4 \pm 1.3$ |
| Bulk density after grinding $BD_{ar}$ (kg·m$^{-3}$) | $115.7 \pm 7.1$ | $98.4 \pm 5.1$ | $121.1 \pm 7.3$ | $93.4 \pm 5.5$ | $144.9 \pm 8.6$ |
| Ash content (as received) $A_{ar}$ (%) | $2.51 \pm 0.28$ | $8.55 \pm 0.39$ | $4.29 \pm 0.34$ | $7.65 \pm 0.42$ | $6.47 \pm 0.38$ |

Table 3. Characteristics of raw materials and pellets - mean values and standard deviations ($\pm$).

The obtained $I_D$ values mean that the milling process, and especially the pelleting process in the case of leaves, will significantly reduce the costs associated with logistics operations.
3.3. Relative Self-Ignition Temperature of Tested Sample

The research carried out on self-ignition of pellets showed (Table 4) that the temperature at which the initiation of ignition occurs is in the range from 221 to 239 °C. The highest auto-ignition temperature (RSIT = 239 °C) is characterized by pellets from American sweetgum (Liquidambar) leaves and the lowest (RSIT = 221 °C) from willow (Salix). These values are comparable with herbaceous plant biomass and waste fractions from tree harvesting [73,74].

Table 4. Relative self-ignition temperature of tested sample (RSIT).

| Sample  | RSIT (°C) | SD (°C) |
|---------|----------|---------|
| Salix   | 221.0    | 3.6     |
| Acer    | 231.6    | 3.5     |
| Quercus | 232.0    | 5.2     |
| Platanus| 228.3    | 3.5     |
| Liquidambar | 239.0 | 3.0     |

Research conducted by Magalhães et al. [75] concerning self-ignition of waste biomass (almond shells and olive residues) and two types of lignite showed that all fuels had a similar ignition temperature amounted to 227 °C (500 K).

Lower self-ignition temperatures from 150 to 170 °C (depending on its pH) was recorded during peat examination [76]. Studies carried out by Schwarzer et al. [77,78] revealed that an increase in the mineral fraction in organic matter causes a decrease in the auto-ignition temperature. These results confirm the differences observed in self-ignition temperatures for willow (Salix). A significant difference in ash content (almost twice as much as other materials) indicates a lower self-ignition temperature by about 10 °C degrees to maple (Acer), oak (Quercus) and plane (Platanus) and almost 20 °C to American sweetgum (Liquidambar).

Therefore, it can be concluded that automatic pellet firing systems should not have difficulties with the use of this type of fuel. Similarly, storage conditions (self-ignition problems) may be similar to those of classical biofuels made from wood and herbaceous biomass [79,80].

3.4. Characteristics of the Ashes

From the produced pellets, ash was obtained as a result of incineration, which was subjected to determination of bulk density and melting temperatures. The obtained results (Table 5) indicated that the ash from the leaves of trees is characterized by a similar melting temperature FT (range 1310–1430 °C). These temperatures suggests that the combustion process does not cause significant risks related to ash deposition/slagging/fouling. This is very important when introducing fuel into classic combustion systems with limited possibility to control the process. Comparing the analyzed pellets to fuels produced from agro biomass, it can be stated that they are characterized by better parameters [39,81,82]. This is very important in case of an attempt of further use. Ash from biomass combustion is used in the production of cement or soil fertilizers [40,61].

The measurements of the embankment density showed that the ashes from the burning of tree leaves have a low density (216–263 kg·m⁻³) comparable to that of wood sawdust (Table 5). Only in the case of willow (Salix) ash its density was almost twice as high (445 kg·m⁻³), which was probably associated with the higher content of mineral fractions in the pellet (Aar=20.12%).
Table 5. Characteristics of the ashes obtained - mean values and standard deviations (±).

| Characteristic Melting Temperatures of Ash | Salix      | Acer       | Quercus    | Platanus   | Liquidambar |
|-------------------------------------------|------------|------------|------------|------------|-------------|
| DT deformation temp. (°C)                 | 1045 ± 10  | 980 ± 17   | 1045 ± 15  | 1140 ± 13  | 1080 ± 15   |
| ST ball temp. (°C)                        | 1250 ± 12  | 1160 ± 14  | 1250 ± 11  | 1270 ± 15  | 1180 ± 17   |
| HT hemisphere temp. (°C)                  | 1260 ± 9   | 1350 ± 12  | 1260 ± 8   | 1350 ± 7   | 1330 ± 9    |
| FT flow temp. (°C)                        | 1310 ± 11  | 1460 ± 13  | 1330 ± 14  | 1430 ± 11  | 1370 ± 16   |
| Bulk density BD (kg m⁻³)                  | 445.5 ± 10.2| 216.7 ± 8.6| 261.3 ± 15.2| 229.3 ± 2.2| 263.7 ± 11.7|

4. Conclusions

Due to the increasing popularity of urban woodland, there is a growing need to remove tree leaves from green areas. This necessity is dictated by safety (maintenance of pedestrian and car traffic, microbiological safety, etc.) as well as a part of the fight against air pollution. As it was shown in the study, tree leaves are characterized by an increased content of mineral fractions (ash). Probably, a significant part of that fraction comes from air pollution captured by the leaves during the growing season. This should be confirmed by further studies performed in this area. Therefore, it seems to be necessary to indicate the optimal solutions for the management of the leaves of urban trees. Currently, these leaves are composted for the production of soil substrates. Nevertheless, an interesting direction is their combustion with energy recovery. Such solutions can increase the profitability of removing leaves from urban areas. The research works carried out so far indicated that the combustion process as well as the management of residues (ashes) should not cause significant problems. A very important factor in fuel management is its quality, and in particular its repeatability. Therefore, research works are undertaken covering the processing of municipal waste (milling, briquetting, gasification, torrefaction, etc.), including tree and shrub leaves.

In the presented paper, the issue of pelletization of selected tree leaf species in order to describe the obtained quality was addressed. The research carried out allowed to indicate key problems with the raw material in the form of tree leaves. The leaves were characterized by a very low embankment density, which significantly impedes logistic processes. Therefore, milling or pelletization processes effectively increase the bulk density of the raw material. Compaction degree after grinding $I_{Dm}$ is in the range 2.51–8.55, for pelletizing $I_{Dp}$ in the range 4.59–6.55 and absolutely compaction degree $I_{Da}$ in the range 52.47–14.29. Pellets obtained in the performed experiments were characterized by very good quality parameters. The lower heating value (about 15 MJ kg⁻¹) or the bulk density (600–650 kg m⁻³) indicate that it is possible to produce quality biofuels on the basis of leaves. The analysis of chemical composition showed that there are no significant risks associated with the concentration of dangerous elements. Tests concerning fusibility of ash were also carried out in order to indicate possible danger connected with melting of ash on the heat walls of the furnaces. The tests indicated that the risk of ash melting is small and similar to classical biofuels.

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