Analyses of Pellets Produced from Spruce Sawdust, Spruce Bark, and Pine Cones in Different Proportions

Nikola Čajová Kantová [1] , Michal Holubčík [2,*] , Alexander Čaja [2] , Juraj Trnka [1] and Jozef Jandačka [2]

1   Research Centre, University of Žilina, Univerzitna 1, 010 26 Žilina, Slovakia;
    nikola.cajovakantova@uniza.sk (N.Č.K.); juraj.trnka@uniza.sk (J.T.)
2   Department of Power Engineering, Faculty of Mechanical Engineering, University of Žilina, Univerzitna 1,
    010 26 Žilina, Slovakia; alexander.caja@fstroj.uniza.sk (A.Č.); jozef.jandacka@fstroj.uniza.sk (J.J.)
*   Correspondence: michal.holubcik@fstroj.uniza.sk

Abstract: A lot of residual biomass has energy value and can be used for further applications through suitable treatments, such as pelletization. This treatment can improve properties, mainly energy density, but can also lead to problems due to their low ash melting temperatures, high ash content, and the formation of harmful compounds during combustion. This article deals with the energy potential of pellets produced from spruce sawdust, spruce bark, and pine cones in different proportions. The impact of cone and bark contents on pellet properties was also observed. The energy properties of the produced pellets were measured, such as the contents of carbon, hydrogen, nitrogen, moisture, volatile, fixed carbon, and ash, as well as calorific values and ash melting temperatures. Based on the results, it can be concluded that the addition of pine cones and spruce bark to spruce sawdust mainly affected the contents of nitrogen and ash and melting temperatures. Despite this, all produced pellets met the standard EN ISO 17225-2 for the content of nitrogen, ash, and also lower calorific value at least B quality. However, only three pellet samples of five met this standard for A2 and B quality for ash melting behavior. Therefore, they present an alternative fuel with interesting energy potential.

Keywords: biofuels; pellets; cones; energy properties; calorific value; ash; melting temperatures

1. Introduction

Residual biomass is a useable energy source. Without further usage, it often rots in landfills. However, raw residual biomass has a high moisture content, low mass, and low energy density [1,2]. There are also problems with the handling, transport, and storage of raw biomass. Therefore, it is important to use a compressed form, briquettes or pellets. Compression can cause a higher energy density, lower moisture, and better fuel homogenization [3,4]. It also reduces the prices of transport and storage.

However, agricultural pellets have some disadvantages, such as low ash melting temperatures, higher ash content, and the formation of harmful compounds during combustion. Due to this, ash slags and sinters are created in combustion chambers, heat exchangers, flue gas paths and using other heat sources. This leads to incorrect combustion processes, reducing combustion efficiency, increasing emissions production and increasing the risk of damage to heat sources. Therefore, it is important to optimize the heat source or use an appropriate burner, and also allow co-combustion with other wood materials [5,6].

Residual biomasses such as tree cones belong to forest waste after seed harvesting. Despite this, cones are a suitable material for the production of compressed fuels. In the work of Aniszewska et al., the physical properties of conifer cones were investiagated. The results of these authors confirmed that the conifer cones have good potential as an energy source [7]. Gendek even dealt with the calorific values of the pine sawdust mixed with pine cones. The obtained lower calorific values were between 17.98 MJ/kg for pure pine sawdust and 18.32 MJ/kg for crushed pine cones [8]. Palacka et al. investigated the energy potential of different parts of the spruce tree such as roots, bark, pure wood,

branches, needles, and also cones. They concluded that the aforementioned parts have potential for further application and processing [9]. Garcia et al. blended pine sawdust with alternative residual biomass for pellet production. The following materials were used as residual biomass: almond shells, olive stones, pine cone leafs, coffee dregs, coffee husks, grape pomace, hazelnut shells, miscanthus, pine kernel shells, switchgrass, and cocoa shells. It was determined that the nitrogen and ash contents were the most restrictive parameters when deciding on the proportion between biomass and added blends. To meet the industrial quality, the suitable proportion for pine cone leafs was less than 30%. Despite this, the results still support using the residual biomass blended with wood material [10]. In this article, we used spruce sawdust mixed with spruce bark and pine cones with the aim of pellet production, and we also investigated their energy properties.

Pellet quality is also influenced by bark content, mainly ash content, as well as ash melting temperature and the produced emissions [11]. Then, it is also important to improve the maintenance of heat sources due to deposit formation, slagging, or sintering [12]. The effect of pine bark on pellet quality was investigated by Filbakk et al. They produced pellets with 0%, 5%, 10%, 30%, and 100% pine bark blended with pinewood. The results confirmed the increased ash content with the increasing amount of bark in pellets (up to 2.5%). Additionally, there were no problems with sintering for 5% or 10% bark contents [13].

Based on this knowledge, it can be surmised that residual biomasses such as tree cones present interesting potential energy sources. Waste materials are currently used, and thus their abundance is decreasing. This article focuses on the energy potential of pine cones, which are blended with spruce sawdust and bark. The impact of cones and bark content on pellet properties was observed. The energy properties of the produced pellets were measured and analyzed, such as the contents of carbon, hydrogen, nitrogen, moisture, volatile, fixed carbon, and ash, and also calorific values and ash melting temperatures.

2. Materials and Methods

This section describes methods of elemental, thermogravimetric (TGA), calorific, and ash melting temperature measurements. The first part of this paper focuses on pure materials that are useable during pelletization such as spruce sawdust, beech sawdust, pine bark, spruce bark, beech bark, and pine cones. These materials were tested on elemental and TGA analyzers. Then, spruce sawdust, spruce bark, and pine cones were added during pelletization in different proportions. These materials were chosen due to the available amount of their waste. Spruce sawdust is the most common waste from sawmills, but pines cones were abundant in our surroundings during this study. The second part deals with created pellets and analyses of elemental, TGA, calorific values, and ash melting temperatures.

2.1. Elemental Analysis

Our elemental analysis was realized on an elemental analyzer CHN628 (Leco Corporation, St. Joseph, MI, USA). This elemental analyzer can determine the contents of carbon, hydrogen, and nitrogen through the combustion of pre-weighed individual samples with an approximate value of 0.1 g. The combustion is realized in the absence of atmospheric gases from the transfer process. In the furnace is pure oxygen, ensuring the complete combustion and recovery of the elements. Operate temperature of this device starts at 950 °C. This device features a software operated through an external to control the system operation and data management.

2.2. TGA Analysis

TGA analysis was realized on a thermogravimetric analyzer TGA 701 (Leco Corporation, St. Joseph, MI, USA). A TGA analyzer can determine the content of moisture, volatile matter, fixed carbon, and ash. During the measurement, samples lose weight and the temperature increases. This device features a software operated externally to control the system operation and data management. Pre-weighed individual samples with an approximate value of 1.2 g were firstly heated at 107 °C in the air atmosphere, then at 900 °C in the nitrogen atmosphere,

and finally decreased at 550 °C in the air atmosphere. Moisture, volatile matter, fixed carbon, and ash content were detected in these individual steps.

2.3. Calorific Values Analysis

This analysis was realized on a calorimeter Leco AC 500 (Leco Corporation, St. Joseph, MI, USA). The calorimeter can determine the higher calorific values (HCV) of pre-weighted individual samples with an approximate value of 1 g. This device also features a software operated externally to control the system operation and data management. Then, lower calorific values (LCV) can be calculated from the determined HCV based on Equation (1).

$$\text{LCV} = \text{HCV} - r_{H2O}\left(W_p + 8.94x_H\right) \tag{1}$$

where r_{H2O} represents the water heat of vaporization (MJ/kg); W_p represents the water content (kg/kg); 8.94 is the hydrogen to water conversion coefficient; and x_H was obtained from Equation (2) [5].

$$x_H = H_h \cdot B_p \tag{2}$$

where H_h represents the hydrogen content (kg/kg), and B_p represents the volatile contents (kg/kg) of the tested samples.

2.4. Ash Melting Temperature Analysis

This analysis was realized on a temperature analyzer, Leco AF 700 (Leco Corporation, St. Joseph, MI, USA). The temperature analyzer can determine four standardized ash melting temperatures from individual prepared sample ash: (1) the shrinkage temperature (ST); (2) the deformation temperature (DT); (3) the hemisphere temperature (HT); and (4) the flow temperature (FT). The ash was obtained through the combustion of samples in a muffle furnace and then prepared in the shape of a pyramid for analysis. The sample in a muffle furnace was first heated at 250 °C, and then at 550 °C. The temperature analyzer operates a maximum of up to 1500 °C. This device features a software operated externally to control the system operation and data management.

2.5. Pelletization

The pellets were produced with the aim of obtaining fuel with a higher energy density. The compressed fuel is easier to transport because it takes up a significantly smaller volume. First, it was necessary to crush the samples with a hammer mill, and then compress them by a small pellet press with a power of 7.5 kW. The input materials, spruce sawdust, spruce bark, and pine cones, were used, as shown in Figure 1. The moisture of the input material was in the range of 15% to 20%. The particle size was mainly in the range of 0.01 mm to 3 mm for spruce sawdust and bark. It was unable to crush the pine cones into these small elements.
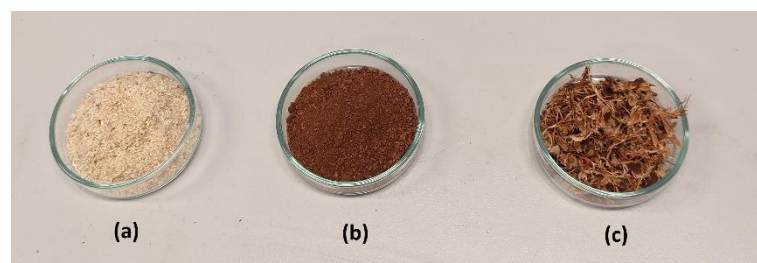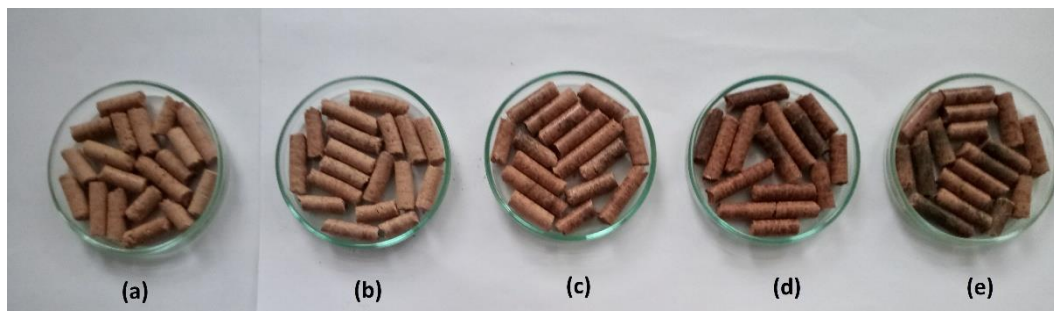

Figure 1. The input materials: (**a**) spruce sawdust; (**b**) spruce bark; (**c**) pine cones.

The pellet press operated approximately under the same conditions. There were produced pellets approximately with the same diameter of 6 mm. Then, they were stored for one week at the temperature of 20 °C and relative humidity in the range from 40% to 50%. Produced pellets were created in different proportions, which are shown in Table 1 and Figure 2.

Table 1. Proportions of created pellets.

Sample	Weight Proportion of Spruce Sawdust [%]	Weight Proportion of Spruce Bark [%]	Weight Proportion of Pine Cones [%]
80/10/10	80	10	10
70/10/20	70	10	20
60/20/20	60	20	20
50/20/30	50	20	30
40/30/30	40	30	30

Figure 2. Individual pellet samples: (**a**) 80/10/10; (**b**) 70/10/20; (**c**) 60/20/20; (**d**) 50/20/30; (**e**) 40/30/30.

2.6. Standard Deviation

The measurements were realized with repetitions three to five times. The results were calculated as mean measurement values with their sample standard deviations (SD) determined in the Microsoft Excel program.

3. Results and Discussion

This section describes the results from elemental, TGA, calorific values, and ash melting temperature measurements.

3.1. Elemental Results

The results from the elemental analysis of various uncompressed materials, such as spruce sawdust, beech sawdust, pine bark, spruce bark, beech bark, and pine cones, are shown in Table 2. The highest carbon content was measured for pine bark. Almost no nitrogen content was measured for spruce and beech sawdust. The resulting values of the research of Palacka et al. for wood spruce, spruce bark, and spruce cones from the elemental analysis are shown in Table 2 for comparison. The measured values of carbon content in this work are slightly lower than the results of Palacka et al. However, the carbon content of spruce-pin-fir sawdust was 46.2% in the work Zhang et al. [14]. The resulting values in this work are comparable with the mentioned works.

The results from the elemental analysis of individual produced pellets are shown in Figure 3. The measured results for carbon content are in the range of 46.25–47.26%; hydrogen content, 6.26–6.36%; and nitrogen content, 0.24–0.32%. With an increasing amount of bark and cones, nitrogen content increases. The graded wood pellets for commercial and residential applications have to meet various quality requirements, such as standard ISO 17225-2 [15]. To meet the A1 quality, the graded wood pellets should have a nitrogen content lower than 0.3%. When the nitrogen content is lower than 0.5%, these pellets met the A2 quality, and when it is lower than 1.0%, they met the B quality. All the tested pellet samples except one (40/30/30) met the A1 quality for nitrogen content.

Table 2. Results from elemental analysis of various uncompressed materials.

Sample	The Content of Carbon (%)	SD (%)	The Content of Hydrogen (%)	SD (%)	The Content of Nitrogen (%)	SD (%)
spruce sawdust	46.14	0.20	6.29	0.10	0.05	0.00
spruce wood [9]	48.56	-	6.43	-	0.04	-
spruce-pin-fir sawdust [14]	46.20	-	6.40	-	<0.10	-
beech sawdust	44.66	0.11	6.23	0.02	0.00	0.00
pine bark	49.43	0.08	5.88	0.03	0.23	0.00
spruce bark	45.83	0.33	5.88	0.06	0.35	0.08
spruce bark [9]	49.52	-	6.15	-	0.7	-
beech bark	43.44	0.20	5.91	0.08	0.67	0.03
pine cones	44.59	0.31	6.22	0.17	0.45	0.02
spruce cones [9]	49.60	-	6.10	-	0.45	-
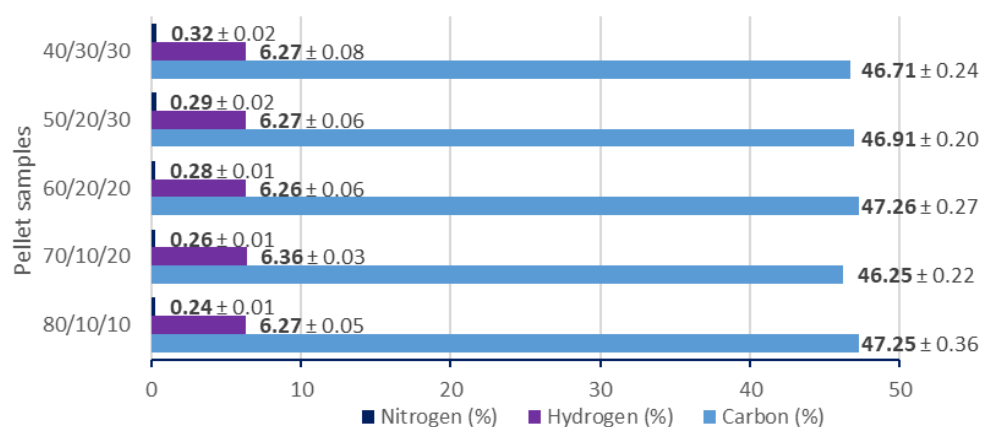
Figure 3. Results from elemental analysis of produced pellets.

3.2. TGA Results

The results from TGA analysis based on their dry basis of mentioned uncompressed materials are shown in Table 3. The highest volatile content was measured for spruce and beech sawdust. The differences in individual volatile contents could be caused due to the different conditions during the growth of individual trees, different soil properties, and atmospheric conditions. The highest ash content was detected for beech bark, and the lowest ash content for spruce sawdust. The resulting values of the research of Zhang et al. for spruce-pin-fir sawdust from TGA analysis are stated in Table 3 for comparison. Based on the research of Aniszewska et al., the ash content of different types of cones was in the range of 0.33–2.12%, with pine cones being at 0.33% [7]. However, Bukhanko et al. stated the ash content of spruce cones with the value of 0.9% [16], and Palacka et al. with a value of 1.06% [9]. The resulting values in this work are comparable with the mentioned works.

The moisture content determined by the TGA analyzer is stated in Table 4 together with the investigated raw materials and produced pellets. The highest content of moisture from the investigated uncompressed materials was measured for pine bark, but beech bark had the lowest value of moisture content. The highest content of moisture from the produced pellets was measured for pellet sample 60/20/20, but pellet sample 50/20/30 had the lowest value of moisture content.

Table 3. Results from TGA analysis of various uncompressed materials based on their dry basis.

Sample	The Content of Volatile (%)	SD (%)	The Content of Fixed Carbon (%)	SD (%)	The Content of Ash (%)	SD (%)
spruce sawdust	82.13	0.13	17.45	0.13	0.42	0.01
spruce-pin-fir sawdust [14]	84.55	-	15.06	-	0.39	-
beech sawdust	82.09	0.14	17.17	0.10	0.73	0.07
pine bark	66.26	0.34	31.46	0.37	2.28	0.04
spruce bark	68.17	0.08	27.62	0.08	4.22	0.05
beech bark	73.35	0.25	20.14	0.27	6.28	0.08
pine cones	79.26	0.32	20.13	0.29	0.61	0.03

Table 4. The moisture content of investigated various uncompressed materials and produced pellets.

Sample	The Content of Moisture (%)	SD (%)
spruce sawdust	7.08	0.05
beech sawdust	7.44	0.10
pine bark	7.98	0.12
spruce bark	7.45	0.06
beech bark	7.05	0.07
pine cones	7.09	0.20
pellet sample 80/10/10	5.30	0.10
pellet sample 70/10/20	5.07	0.29
pellet sample 60/20/20	6.43	0.05
pellet sample 50/20/30	4.60	0.16
pellet sample 40/30/30	5.11	0.16

The results from TGA analysis based on their dry basis of produced pellets are shown in Figure 4. The measured results of volatile contents are in the range of 77.95–80.60%; fixed carbon contents, 18.64–20.46%; and ash contents, 0.76–1.59%. With the increasing amount of bark and cone in pellets, the contents of ash and fixed carbon increase, but the volatile content decreases. Holubcik et al. found that with the usage of 10% bark in wood pellets, the ash content can increase by 0.04% [11]. According to EN ISO 17225–2 [15], the ash content has to be less than 0.7% for A1 quality, less than 1.2% for A2 quality, and less than 2% for B quality. All tested pellets met the standard for at least B quality.
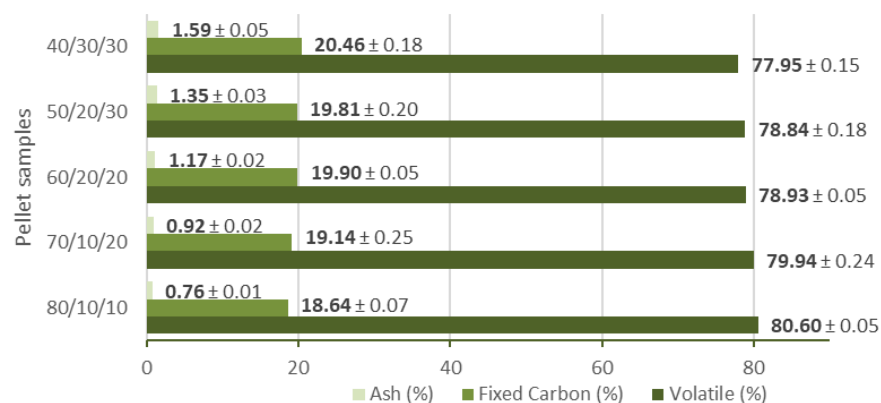
Figure 4. Results from TGA analysis of produced pellets based on their dry basis.

3.3. Calorific Value Results

The results from the calorific value analysis of the produced pellets are shown in Figure 5. The measured HCV results are in the range of 18.33–18.69 MJ/kg. The calculated LCV results are in the range of 17.05–17.50 MJ/kg. Based on the research of Aniszewska and Gendek, the average LCV of the cones from selected forest trees species was in the range of 17.81–19.86 MJ/kg; for pure pine cones, this was 18.11 MJ/kg [17]. Another work

of Gendek was focused on the mix of pine sawdust and pine cones, while the results of LCV were in a range between 17.98 MJ/kg for pure pine sawdust and 18.32 MJ/kg for crushed pine cones [4]. The resulting values in this work are in accordance with the mentioned works. The minimum net calorific value (also known as lower calorific value) is 16.5 MJ/kg for meeting the standard EN ISO 17225-2 [15]. All tested pellets met this standard.
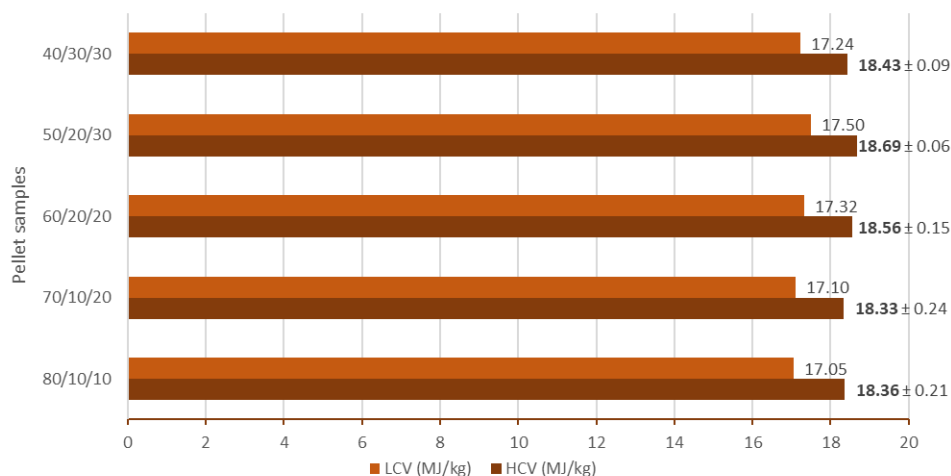

Figure 5. Results from calorific value analysis of produced pellets.

3.4. Ash Melting Temperature Results

The results from the ash melting temperature analysis of the produced pellets are shown in Figure 6. With the increasing amount of bark and cones in pellets, the ash melting temperatures decrease. Based on the research of Wang et al., spruce wood started to melt at around 1380 °C and was completely fused at around 1480 °C. With the addition of 10% bark, the melting started at 1250 °C, and it was completely melted at 1378 °C [18]. The resulting values in this work are in accordance with the mentioned work. According to EN ISO 17225-2 [15], the deformation temperature (DT) has to be more than 1200 °C for A1 quality, and more than 1100 °C for A2 and B quality. Only three pellet samples of five met this standard for A2 and B quality for ash melting behavior.
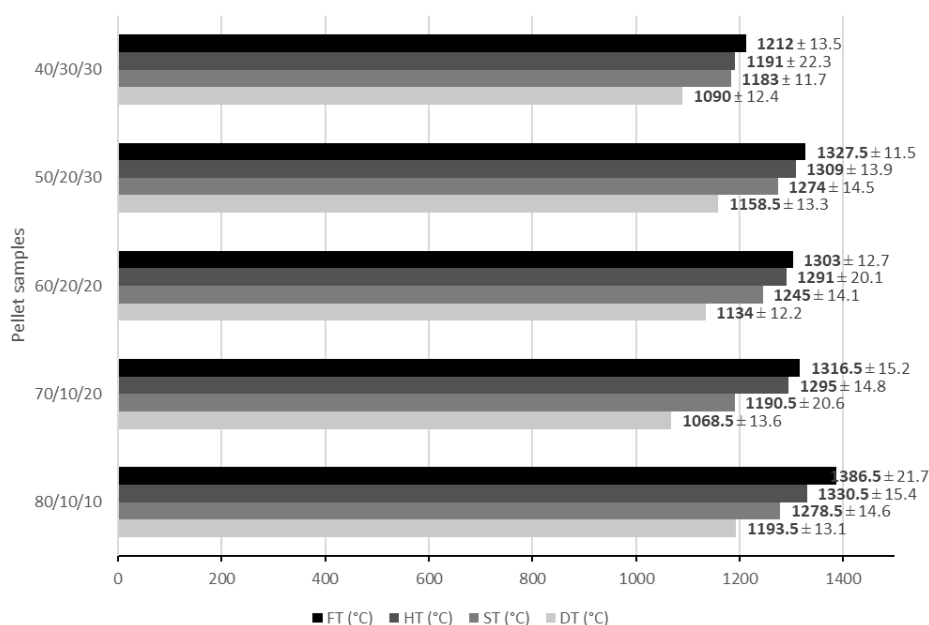

Figure 6. Results from melting temperature analysis of produced pellets.

4. Conclusions

The addition of pine cones and spruce bark to spruce sawdust mainly affected the contents of nitrogen and ash, as well as melting temperatures. The nitrogen content increased from 0.24% (for 80/10/10 pellets) to 0.32% (for 40/30/30 pellets). Despite this, it met the standard EN ISO 17225-2 [15] for graded wood pellets for commercial and residential applications, where the nitrogen content has to be less than 0.5% for A2 quality. All tested samples except 40/30/30 also met the A1 quality (nitrogen content lower than 0.3%). The ash content increased from 0.76% (for pellets 80/10/10) to 1.59% (for pellets 40/30/30). The ash content has to be less than 0.7% according to this standard for A1 quality, less than 1.2% for A2 quality, and less than 2% for B quality. Pellet samples 50/20/30 and 40/30/30 met only B quality for ash content, but the remaining tested samples (60/20/20, 70/10/20, 80/10/10) met A2 quality for ash content. The minimum ash melting temperature was DT with a value of 1068.5 °C for 70/10/20 pellets. The deformation temperature (DT) has to be more than 1200 °C according to EN ISO 17225-2 [15] for A1 quality, and more than 1100 °C for A2 and B quality. Only three pellet samples met the standards for A2 and B quality: 50/20/30, 60/20/20, and 80/10/10. The lower calorific value (also known as net calorific value) met the standard EN ISO 17225-2, where LCV has to be higher than 16.5 MJ/kg. The calculated LCV were in the range from 17.05 MJ/kg to 17.50 MJ/kg and met the mentioned standard. Based on the achieved results, the produced pellets present an alternative fuel with interesting energy potential.

Author Contributions: N.Č.K., investigation, conceptualization, resources, writing—original draft; M.H., writing—review and editing, funding; A.Č., data curation, formal analysis; J.T., data curation and J.J., formal analysis, methodology. All authors have read and agreed to the published version of the manuscript.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Data available in a publicly accessible repository.

Conflicts of Interest: The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results. The authors declare no conflict of interest.

References

1. Picchio, R.; Latterini, F.; Venanzi, R.; Stefanoni, W.; Suardi, A.; Tocci, D.; Pari, L. Pellet production from woody and non-woody feedstocks: A review on biomass quality evaluation. *Energies* **2020**, *13*, 2937. [CrossRef]
2. Werle, S.; Dudziak, M. Gaseous fuels production from dried sewage sludge via air gasification. *Waste Manag. Res.* **2014**, *32*, 601–607. [CrossRef] [PubMed]
3. Sitek, T.; Pospisil, J.; Polacik, J.; Chylek, R. Thermogravimetric analysis of solid biomass fuels and corresponding emission of fine particles. *Energy* **2021**, *237*, 121609. [CrossRef]
4. Gendek, A.; Aniszewska, M.; Malaťák, J.; Velebil, J. Evaluation of selected physical and mechanical properties of briquettes produced from cones of three coniferous tree species. *Biomass Bioenergy* **2018**, *117*, 173–179. [CrossRef]
5. Nosek, R.; Backa, A.; Ďurčanský, P.; Holubčík, M.; Jandačka, J. Effect of paper sludge and dendromass on properties of phytomass pellets. *Appl. Sci.* **2021**, *11*, 65. [CrossRef]
6. Honus, S.; Pospíšilík, V.; Jursová, S.; Šmída, Z.; Milnár, V.; Dovica, M. Verifying the prediction result reliability using k-ε, Eddy dissipation and discrete transfer models applied on methane combustion using a prototype low-pressure burner. *Adv. Sci. Technol.* **2017**, *11*, 252–259. [CrossRef]
7. Aniszewska, M.; Gendek, A.; Zychowicz, W. Analysis of selected physical properties of conifer cones with relevance to energy production efficiency. *Forests* **2018**, *9*, 405. [CrossRef]

8. Gendek, A. Combustion heat and calorifi c value of the mix of sawdust and cones of common pine (*Pinus sylvestris* L.). *Ann. Wars. Univ. Life Sci. SGGW* **2015**, *66*, 137–144.

9. Palacka, M.; Vician, P.; Holubčík, M.; Jandačka, J. The energy characteristics of different parts of the tree. *Procedia Eng.* **2017**, *192*, 654–658. [CrossRef]

10. García, R.; Gil, M.V.; Rubiera, F.; Pevida, C. Pelletization of wood and alternative residual biomass blends for producing industrial quality pellets. *Fuel* **2019**, *251*, 739–753. [CrossRef]

11. Holubcik, M.; Jandacka, J.; Palacka, M.; Kantova, N.; Jachniak, E.; Pavlik, P. The impact of bark content in wood pellets on emission production during combustion in small heat source. *Communications* **2017**, *19*, 94–100. [CrossRef]

12. Matúš, M.; Križan, P.; Šooš, L.; Beniak, J. The effect of papermaking sludge as an additive to biomass pellets on the final quality of the fuel. *Fuel* **2018**, *219*, 196–204. [CrossRef]

13. Filbakk, T.; Jirjis, R.; Nurmi, J.; Høibø, O. The effect of bark content on quality parameters of Scots pine (*Pinus sylvestris* L.) pellets. *Biomass Bioenergy* **2011**, *35*, 3342–3349. [CrossRef]

14. Zhang, B.; Yang, B.; Wu, S.; Guo, W.; Zhang, J.; Wu, Z.; Wang, Z.; Lim, J.C. Effect of torrefaction pretreatment on the fast pyrolysis behavior of biomass: Product distribution and kinetic analysis on spruce-pin-fir sawdust. *J. Anal. Appl. Pyrolysis* **2021**, *158*, 105259. [CrossRef]

15. *ISO 17225*; 2 Solid Biofuels—Fuel Specifications and Classes—Part 2: Graded Wood Pellets. International Organization for Standardization: Geneva, Switzerland, 2021.

16. Bukhanko, N.; Attard, T.; Arshadi, M.; Eriksson, D.; Budarin, V.; Hunt, A.J.; Geladi, P.; Bergsten, U.; Clark, J. Extraction of cones, branches, needles and bark from Norway spruce (Picea abies) by supercritical carbon dioxide and soxhlet extractions techniques. *Ind. Crops Prod.* **2020**, *145*, 112096. [CrossRef]

17. Aniszewska, M.; Gendek, A. Comparison of heat of combustion and calorific value of the cones and wood of selected forest trees species. *Leśne Pr. Badaw.* **2014**, *75*, 231–236. [CrossRef]

18. Wang, L.; Skreiberg, Ø.; Khalil, R.; Li, H. Effect of fuel mixing on melting behavior of spruce wood ash. *Energy Procedia* **2019**, *158*, 1342–1347. [CrossRef]