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Search for Alternative Raw Materials for Pellet Production – a Preliminary Study

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***Abstract* – A preliminary study for a selection of local alternative raw materials and their possible applicability in pellet production was done. 13 alternative biomass sources available in Riga, Latvia were tested: pine mix pellets (PMP); pellets of a mix of wood shavings, synthetic and textile waste (SSK); hogweed (*Heracleum sosnowsky*, HW); the full plant of *Solidago canadensis* L. (SC); the full plant of *Solidago canadensis* L. outgrown (SCO); mowed grass from the municipality (MGM); mowed grass from the garden (MGG); chamomile full plant without stems (*Matricaria chamomilla*, CF); hemp without roots (*Cannabis sativa* L., HWR); hemp stems (*Cannabis sativa* L., HS); hemp leaves with seeds (*Cannabis sativa* L., HLS); spent grain (SG); damaged spent grain (DSG). The determined quality parameters were the moisture content, the ash content, the calorific value and ash melting temperature. A comparison was made with the quality parameters defined in ENplus certification for B-class wood pellets in requirements for producers. Additional determination of chemical composition using XRD *(*X-ray diffraction*)* and SEM-EDX *(*scanning electron microscopy – energy dispersive X-ray spectroscopy*)* for the ash of two selected materials – SC and HLS – was performed to determine possible issues. The main issues highlighted by the results is the high ash content of alternative materials, up to 11.27 wt% while it is below 2.00 wt% for B-class wood pellets, and the low ash deformation temperature, below 1100 °C for 5 of the studied materials. Considering this, a calculation was made to determine the optimal proportion of alternative material to be used in a raw material mix for pellets to achieve a quality corresponding to B-class wood pellets. The aim of the study is to evaluate a set of locally available biomass materials, determining the values of their characteristic parameters, thus identifying alternative resources to wood for the production of pellets. This kind of promoting the diversification of energy resources, increasing energy independence and strengthening energy security.**

***Keywords* – Alternative biofuels; ash content; ash melting; biomass pellets; calorific value; chemical composition; energy; moisture content.**

Nomenclature		
A_d	the ash content of the sample on a dry basis	wt%
DT	the ash deformation temperature	°C
FT	the ash flow temperature	°C

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HHV	the gross calorific value of the sample, the highest heating value	MJ/kg
LHV	the net calorific value of the sample, the lowest heating value	MJ/kg
M	the moisture content used for LHV calculation and comparison	wt%
M_{ad}	the moisture content in the general analysis sample	wt%
M_{ar}	the total moisture content of the material	wt%
MMT	Million Metric Tons	–
MT	Million Tons	–

1. INTRODUCTION

The consumption of fuel pellets in European Union (EU) had experienced significant growth, rising from 15.49 MMT (million metric tons) in 2015 to 24.8 MMT in 2022. This increase can be primarily attributed to the escalating prices of fossil fuels and technology development, leading to a rapid increase in pellet use in the residential sector. Further growth was expected [1]. The newest "Bioenergy Europe Statistical Report" shows that wood pellets remain a resilient fuel for use in the EU, reaching production of 20.7 MT and consumption of 21.9 MT in 2023. However, the overall pellet production and consumption trends show reductions in the industrial wood pellet market, breaking the trend for the first time in 20 years. Additional impact factors are higher input prices and record-breaking warm weather conditions in winter. Despite these challenges, pellet use for residential and commercial heating remains high in the EU reaching 59 % [2].

Over the past years (data available from 2008 to 2023), there has also been a constant growth in fuel pellet consumption in Latvia. In 2008, 16 thousand tons of wood fuel pellets were utilized. However, by 2022, consumption reached a new peak – 194 thousand tons. Available data reveals that the most rapid increase in fuel pellet consumption started in 2012 with 136 thousand tons when consumption had grown more than 4 times compared to the previous year. For the following years until now, the amount of fuel pellet consumption has fluctuated but has consistently remained above 100 thousand tons, signifying a sustained demand for this energy source in Latvia. The pellet consumption decreased in 2023, reaching 164 thousand tons, which aligns with the global trend [3].

Over the last 15 years, Latvia has witnessed rapid growth in wood pellet production as well, becoming the second-largest producer in Europe in 2021 with around 2264 MT, trailing only behind Germany [1], [3]. Latvia has constantly exported around 90 % of the produced and imported wood pellets [3] to other European countries like the United Kingdom, Denmark, Sweden, Estonia, and Finland [4]. In 2021, less than 10 % of the produced and imported fuel pellets were consumed in the domestic market. However, in 2022, there was a significant increase in pellet consumption, with nearly 30 % of produced and imported products consumed in the domestic market [3].

From 2006 to 2021, the fuel pellet price for final consumers in Latvia fluctuated, but on average it was 133 €/t. However, in 2022, the fuel pellet price increased significantly to 184 €/t compared to the previous year's price of 136 €/t. An even bigger price was observed in 2023, reaching 227 €/t. [5] Due to the lack of reliable sources or statistical data on pellet price for 2024/2025 winter season, it is currently impossible to predict whether the pellet price will increase or remain stable. This aligns with consumption reduction and also highlights the fact that new, cheaper alternatives are needed to stabilize the price and consumption of wood pellets in the long run.

Most of the households in Latvia use firewood and wood residues as the main source of energy for heating. A minority of households use densified solid biofuels – briquettes and pellets – for heat generation [6]. The number of boiler houses using wood pellets in Latvia has gradually increased in the last years. In 2007 there were only 8 boiler houses in Latvia that used fuel pellets for heat generation. However, in 2023, the number had increased more than 12 times, to 116 [7].

Over the past decade decline in the utilization of natural gas in boiler houses is evident, while the use of pellets and wood chips is gradually increasing, indicating movement towards more sustainable and environmentally friendly alternatives [7]. In 2022, nearly 36 % of all boiler houses used natural gas for centralized district heating systems [7], about 29 % used wood chips and 16 % fuel pellets. The price of natural gas increased rapidly from 334 €/1000 m^3 in 2021 to 1027 €/1000 m^3 in 2022, decreasing to 837 €/1000 m^3 in 2023 [5]. The general increase in energy consumption and fossil fuel prices, as well as growing pollution, are the main reasons for the rapid transition to alternative energy sources and the increase of biomass utilization in household and industrial heating systems. Due to rapidly growing demand, existing resources are depleted, and their prices are rising. It is necessary to find high-quality and low-cost alternatives to cover this increasing demand. Alternative fuels can reduce the use of fossil fuels, promote energy independence, as well as reduce price fluctuations of fuel resources.

1.1. The Properties of Biomass Materials

Numerous laboratory-scale studies have been conducted for the production of non-wood biomass fuel pellets. These studies involve various agricultural materials such as willow, reed canary grass, straw, cereal screenings, rape-seed meal, rape cake, distiller's waste, invasive plant species (hogweed and giant knotweed), hemp, garden residues and others [8]–[11]. Also, their utilization in the residential sector is not sufficiently tested. At the same time, there is an interest in the industrial application of torrefied biomass pellets as a replacement for fossil fuels, but also more studies need to be done [12].

Nilsson *et al.* [10] conducted a review on parameters on different agricultural biomass, revealing variations in the gross caloric value from 18.9 MJ/kg for straw to 26.0 MJ/kg for rape cake. The ash content also varies, with the lowest values found in hemp and willow at 2.3 wt% and 2.9 wt%, respectively, and the highest in cereal screening at 9.8 wt%. [10] In research by Stulpinaite *et al.* [8] different proportions of hemp, lignin, and wood sawdust mixtures were tested. The gross calorific value of pure hemp was 16.80 MJ/kg. By adding 25 % lignin and 25 % sawdust to the mixture, the gross calorific value increased to 21.35 MJ/kg [8]. Gross calorific value for different garden waste material is from 14.85 to 16.15 MJ/kg [13], [14].

The moisture content of pellets plays a crucial role in determining both the net calorific value and mechanical durability. For example, if moisture content increases from about 3 wt% to 15 wt% or higher than that, the durability is ≤ 20 % or less for wood pellets [15]. However, such an extreme increase in moisture content is only possible if fuel pellets are exposed to changing humidity, rainfall, and temperature. Comparing the net calorific value between pellets with moisture content of 3.2 wt% and 9.2 wt%, the change in net calorific value was from 17.38 MJ/kg to 19.25 MJ/kg [15].

A different situation is for pellets from straw. In research by Serrano *et al.* [16], the effect of moisture content on barley straw pellets was analysed. They concluded that mechanical durability increases with straw moisture content, but pellet density decreases with moisture content in this specific case. The optimal moisture content range for each material pelletizing differs. The net calorific value for barley straw pellets made in was 16.42 MJ/kg, ash content

– 10.51 wt%. Barley was grown and the straw was collected and treated in the central part of Spain, Albacete [16].

In research by Theerarattananoon *et al*. [17], four types of biomass pellets were studied - corn stover, sorghum stalk, wheat straw, and big bluestem. The highest bulk density was for wheat straw – 495.8 kg/m^3, but the lowest was for sorghum stalk with value of 365.2 kg/m^3. Increased moisture content decreased mechanical durability for corn stover pellets, wheat straw and big bluestem. For these materials, the maximum durability value was 96.8 % at the moisture range from 9–14 wt%. However, for sorghum stalk pellets, the durability value increased with increased moisture content and reached a maximum of 89.5 % at a moisture value between 14 wt% and 16 wt% [17].

The study by Jasinskas *et al*. [18] looked at the use of maize, hemp and field beans in a combined mixture of biomass for pellet production. The quality and combustion analyses were performed for all the prepared samples. The determined parameters were moisture and ash content, calorific value, ash melting temperatures and density, combustion characteristics and gaseous emissions. All the testing was carried out by methodology based on European and Lithuanian standards with 5 repetitions. The main influence of raw materials comes from selected plant chemical composition, species and physical characteristics. The obtained results varied with moisture content below 10 wt% in all cases, high density (up to 1161 kg/m^3), a net calorific value higher than 16.8 MJ/kg, but a relatively high ash content – from 4.49 wt% to 6.78 wt%. The deformation temperature of pellet ash was between 976 °C and 1007 °C. The gaseous emissions were within the limits of the standards, with a reduction of CO_2 emissions when the biomass of field beans and hemp were mixed with maize [18].

There are different methods for the determination of the chemical composition of materials. XRF (X-ray fluorescence spectroscopy) and ICP-OES (inductively coupled plasma optical emission spectrometry) have been used for biomass ash analysis according to EN ISO 16967 and EN ISO 16968 standards. The content of Al, Ca, Mg, K, Si, S, Fe, Mn, F, Na, Ti, Sr, Zn, Rb oxides and Cl was determined in the study [19] of wood pellet ash obtained in an 11 to 18 kW combustion pilot plant [19].

Regarding the chemical composition of biofuels, major elements (Al, Si, K, Na, Ca, Mg, Fe, P, Ti) are significant in processes regarding ash and corrosion of equipment, but minor elements (As, Ba, Cd, Co, Cu, Cr, Hg, Mo, Mn, Ni, Pb, Sb, Tl, V and Zn) are relevant regarding particulate matter formation and impact on the environment caused by ash. [20] Plants of agricultural origin have higher concentrations of Cl, K, Si, N and S than biomass that is traditionally used in pellet production. [21] High K content impacts ash melting, reducing the melting temperature and enhancing its' deposition in the boiler system where biomass is used. Additional disadvantages of high K content in fuel are increased corrosion risks and reduced heat transfer [22].

Agricultural biomass chemical composition and suitability for use in biofuel production were analysed in a study by Čepauskienė *et al*. [23]. the chemical composition was determined using ICP-AES (inductively coupled plasma atomic emission – spectroscopy) and XRD (X-ray diffraction) methods. The impact of chemical composition on ash melting of reed canary grass, hemp, orchard grass, four types of straw and two woody biomass types was obtained. The highest ash content was observed for buckwheat straw and canola straw – up to 6.5 wt%. During ash melting temperature determination the main focus was on shrinkage starting temperature (SST) because it describes the start of ash melting. The highest SST was for both grass types and hemp ashes – between 946 °C and 1079 °C, the lowest for straw – between 698 °C and 783 °C. The chemical composition analysis shows that Si reduces ash melting temperatures, Na and K impacts initial stage of ash melting, including SST, but Ca final stage.

The most common compounds in biomass are calcium carbonate ($CaCO_3$), potassium sulfate (K_2SO_4) and potassium chloride (KCl) [23].

XRF analysis has also been used for analysis of pine wood and sewage sludge ash. The dominating compounds in wood ash were CaO, MgO, SO_3 and P_2O_5, in the sewage sludge ash the main compounds were SiO_2, Al_2O_3 and P_2O_5 [24]. XRF and XRD have also been used for the analysis of cotton stalk ash. Additional analysis of particulate matter composition was carried out using ESEM-EDX (environmental scanning electron microscopy with energy dispersive X-ray). Elements that were determined in the samples in the highest concentrations were: K, Cl, S, Ca, Mg, P, and Si [25].

Another use of XRF was for chemical composition analysis of cabbage waste pellet ash. For the morphology of the ash TEM (transmission electron microscope) and EDX. To determine the chemical composition of the sample before combustion the XRD method was used. The main elements determined in the fresh sample were Si, Ca, K, Na, and Cl. [26]

XRF method has been used for the analysis of hemp and hemp-wood mixture pellets before combustion, their ash and particulate matter from combustion. Particulate matter with a dominating fraction up to 1 μm mainly contains P, Cl, S, Si, Ca and Zn. The main elements identified in pellets before combustion were: K, Si, S, Ca, Zn, Ti, Cl, Al, P, Fe, and Cr. The elements with the highest concentrations in ash were Ca, K, Si and Mg, in lower concentrations – P, Fe, Al, Na, Mn, S, Cl and F, in small concentrations – Cu, Cr, Zn, Ni, V, As, etc. [21].

The ash of rice husks, corn stalks and cotton stalks and their mixtures were analysed using the XRF method. The main elements identified were K, Ca and Si in the form of oxides. Variable concentrations of Mg, Al, P, S, Cl, and Fe oxides were determined. XRD was used for the analysis of ash crystal structure. ESEM together with EDX was used for morphology and chemical composition analysis. ICP-MS (inductively coupled plasma-mass spectrometer) was used for K and Si content determination for biomass before combustion [27].

The studies reviewed in the paper mostly focus on a specific fuel or combustion parameter giving little information on the possible use of a certain material in energy production. There is a lack of an overall analysis of the chemical composition and physical parameters of fuels and that can lead to misconceptions of the possible biomass characteristics during combustion. Studies show that a fuel of a certain biomass type was made and some fuel parameters were determined or some techno-economic calculation were done, but there is no comparison offered with typical fuel types that could be used as reference values, limiting the ability to evaluate the sustainability of studied fuels.

An evaluation of the suitability of a certain material for use in fuel production is important to reduce the possible risks from the use of it. However, there is a lack of evaluation methods for this kind of task. Here, we propose a quantitative evaluation of a group of alternative raw materials that could be used as replacements for wood in pellet production. The tested materials and their quality parameters were compared to the parameters of the wood pellets according to B-class of the ENplus [31] certification system to find the optimal material proportions to obtain the minimum quality requirements. The aim of the study is to evaluate a set of locally available biomass materials, determining the values of their characteristic parameters, thus identifying alternative resources to wood for the production of pellets. This kind of promoting the diversification of energy resources, increasing energy independence and strengthening energy security.

2. METHODOLOGY

2.1. Laboratory Tests

The methodology of the study is shown in Fig. 1. It summarizes the main stages of the study and the ISO standards that were used as a basis for the laboratory testing. The selection of materials was based on the availability of resources at the local level at the time of the study. The studied materials were:

- pine mix pellets (PMP);
- pellets of wood shavings, synthetic and textile waste mix (SSK);
- hogweed (HW);
- Canadian goldenrod (CG);
- Canadian goldenrod outgrown (CGO);
- mowed grass from the municipality (MGM);
- mowed grass from the garden (MGG);
- chamomile full plant without stems (CF);
- hemp without roots (HWR);
- hemp stems (HS);
- hemp leaves with seeds (HLS);
- spent grain (SG);
- damaged spent grain (DSG).

The laboratory personnel collected materials on-site or obtained them from industry after communication between the personnel and industry representatives. The collection procedure differed depending on the material, but it was not based on any standard. PMP and SSK were received from producers and considered ready for testing without additional pre-treatment. HW, the full plant of CG and the full plant of CGO were collected in Riga by cutting or pulling them out from fields during the full bloom season (CG) and at the end of the bloom season (CGO). Hemp was collected from the producer and divided into parts at the laboratory. SG was obtained from industry, right after use in the beer brewing process. DSG was obtained by accident when SG was damaged during the initial drying process in the laboratory. MGM was collected from the municipality maintenance efforts, but after a heavy rain. A colleague supplied MGG from their yard. CF was taken from dried up decorations, as leftovers.

Alternative materials were tested according to the methodology based on ISO standards regarding the testing of solid biofuels. The performed tests were:

- the determination of the total moisture content of the sample,
- the moisture content in the analysis sample,
- the ash content on a dry basis,
- the determination of the calorific value of the material,
- the determination of ash melting temperature using ash obtained in the laboratory environment.

At least two repetitive measurements were performed for each test. The testing was carried out at the accredited *"Environmental monitoring laboratory"* of the Institute of Energy Systems and Environment at Riga Technical University. The accreditation scope of this laboratory is physical and mechanical testing of solid biofuels.

The studied samples were prepared for each test according to LVS EN ISO 14780:2017. The materials were mixed and quarted for the total moisture content determination. The general analysis sample for other tests was obtained using a hammer mill with a sieve pore size of 1 mm. The ashes obtained during the tests were collected and later used for the determination of ash melting temperature [32].

The determination of the total moisture content of the studied samples was carried out according to LVS EN ISO 18134-2:2017. The method describes the amount of sample that needs to be dried at a constant temperature of around 105 °C until constant mass is reached. The moisture content (M_{ar}) is calculated according to the mass change during the drying process as described in the standard and is expressed in weight percentage [33].

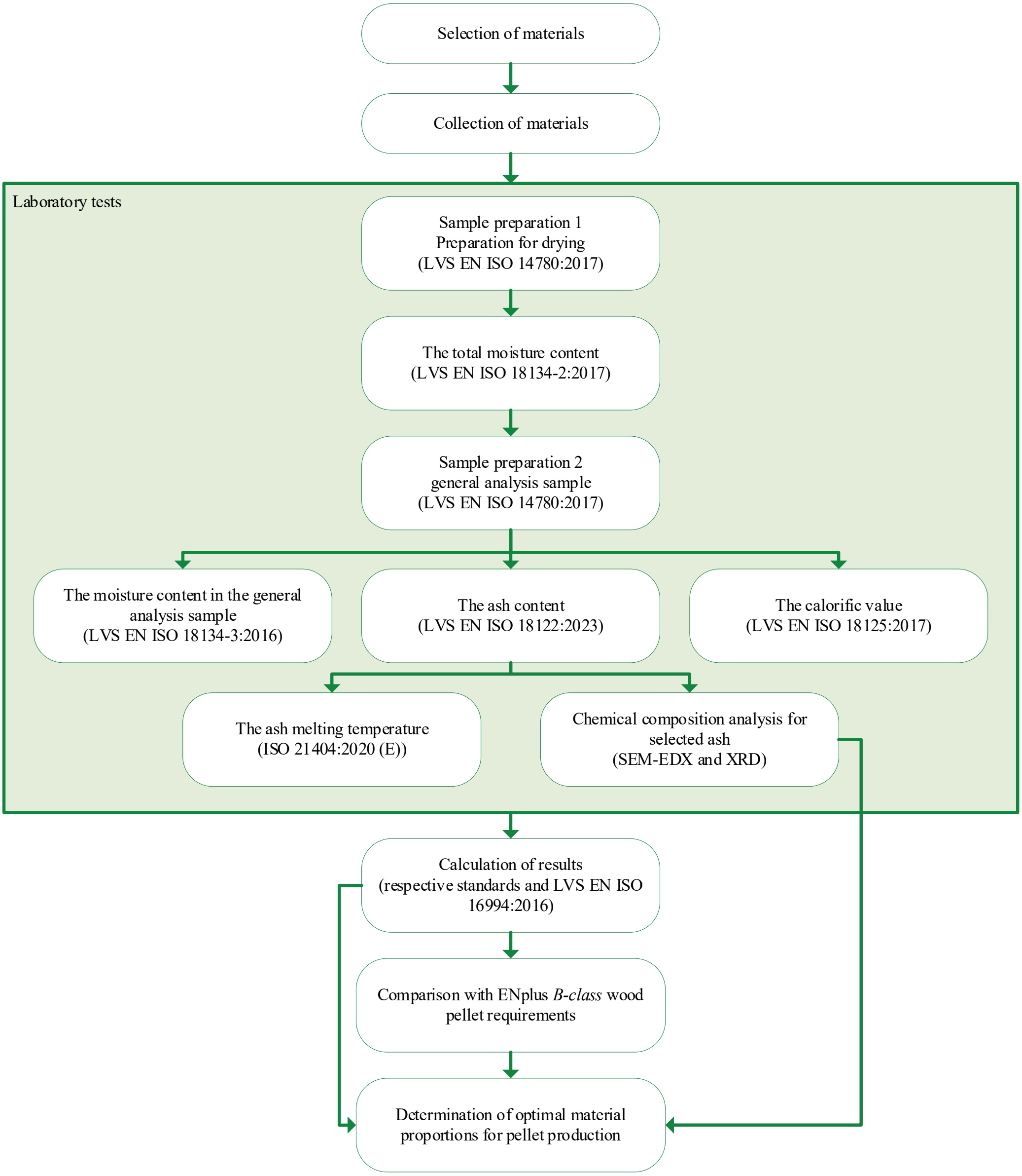

Fig. 1. The methodology of the study.

The moisture content in the general analysis sample (M_{ad}) was determined according to LVS EN ISO 18134-3:2016. The method is similar to the total moisture content determination, using the previously dried and milled sample with a smaller mass and limited air exchange rate in the oven. Drying occurs at the temperature of 105 °C until constant mass is reached. The value is calculated based on mass change and is expressed in weight percentage. This parameter is important for the determination of the ash content and calorific value [34].

The moisture content that was used for the comparison of materials varied depending on how wet the material was before testing. If the material needed to be dried in an oven before preparation, then the moisture content of the general analysis sample was used. If the material was air-dried and needed no additional drying before preparation, then the total moisture content of the sample was used. The net calorific value of samples was calculated based on the selected moisture content (M).

The ash content of the sample was obtained based on the method described in LVS EN ISO 18122:2023. Ashing was carried out for the general analysis sample with particle size below 1 mm at the temperature of 550 °C. The ash content is expressed on a dry basis (A_d) of the sample as a mass fraction in percentage. M_{ad} is considered when calculating the ash content. Ash content can be calculated and expressed on an "as received" basis using the conversion factors described in LVS EN ISO 16993:2016 [35], [36].

The calorific value was determined according to LVS EN ISO 18125:2017 using an isoperibol bomb calorimeter. Before testing the calorimeter is calibrated by combustion of a certified calibration agent (benzoic acid). The calorific value of the fuel is determined at constant volume and pressure, with the reference temperature of cooling water around 25 °C. The gross and net calorific values of a sample are calculated considering the chemical composition of the sample (C, H, N, S content) and obtained ash, and moisture content values. The C, H, N, S, and O values were assumed from the Annex G of the standard [37] or the Phyllis2 [38] database. If one of the element values was unknown, it was calculated according to the composition of the dry material. The calorific values are expressed in MJ/kg. The gross calorific value is abbreviated as HHV and the net calorific value as LHV [37].

Ash melting temperature was determined using the method described in ISO 21404:2020(E). The ash obtained at 550 °C was used for tests. The determined temperatures are SST – shrinking starting temperature, DT – deformation temperature, HT – hemisphere temperature, and FT – flow temperature. The tests are carried out in an oxidizing atmosphere, temperature results are rounded to the nearest 10 °C. The temperatures that were compared between the samples were DT and FT [39].

Additional testing of chemical composition was carried out at the Institute of Materials and Surface Engineering at Riga Technical University for the ash of two selected samples – the full plant ash of CG and the ash of HLS. The specific samples were chosen due to the different colours and unusual residues visually seen in the ash when compared to other tested samples. The chemical composition was carried out using XRD (Rigaku Ultima+) and SEM-EDX (scanning electron microscopy – energy dispersive X-ray spectroscopy, FEI Nova NanoSEM650 equipped with an EDAX SDD EDX module) operating at 15 kV.

3. Results

3.1. Testing Results of Alternative Materials

The testing results of the 13 analysed samples are displayed in Table 1. The table includes additional data on the limiting parameters of B class ENplus wood pellets (B).

The shaded results show the parameters of the materials that do not reach the requirements of the B-class wood pellets. The results show that the most critical quality parameter for studied materials is the ash content. It ranges from 1.34 wt% for SSK to 16.5 wt% for HLS. The samples containing wood have the lowest ash content, with the only exception being hogweed (1.92 wt%). The other critical parameter is the DT of ash. It is below 1100 °C for 5 of the studied samples. These critical parameters show that the addition of material with low ash content, possibly wood, to most alternatives, should be considered.

The moisture content of the materials varies depending on the collection state, but the one considered in comparison with B-class requirements is the moisture content of the milled material. In this case, there is only one material that exceeds the limit of 10.0 wt% - it is SSK with 11.6 wt%. The LHV of the studied materials in the milled state is satisfying. There are only two samples below the limit of 16.5 MJ/kg, which are SSK and MGG.

TABLE 1. THE PARAMETERS DETERMINED FOR THE ALTERNATIVE MATERIALS

Material	A_d, wt%	M_{ar}, wt%	M, wt%	HHV, MJ/kg	LHV, MJ/kg	DT, °C	FT, °C
PMP	1.99	5.7	5.7	19.9	17.8	1191	1318
SSK	1.34	11.6	11.6	19.7	15.9	1070	1147
HW	1.92	–	8.2	19.5	16.5	1297	1545
CG	6.80	59.2	2.9	19.7	18.3	1133	1545
CGO	6.24	50.6	5.4	19.7	18.2	1028	1183
MGM	9.75	85.7	4.3	19.3	17.9	1119	1165
MGG	7.80	–	7.6	18.8	15.9	958	1074
CF	6.19	–	1.5	18.2	16.6	1160	>1555
HWR	11.27	50.0	8.1	18.6	17.5	1422	1543
HS	3.78	55.0	1.5	18.3	16.9	1350	1498
HLS	16.5	48.8	2.3	18.3	17.2	1300*	1800*
SG	3.85	75.2	1.1	21.2	19.7	925	>1555
DSG	4.12	75.2	7.5	21.6	20.1	944	>1555
B	≤ 2.00	–	≤ 10.0	–	≥ 16.5	≥ 1100	–

* data obtained from [40, p. 2]

3.2. Chemical Composition of Selected Ash Samples

The chemical composition analysis was carried out for CG and HLS sample ash to see whether these materials contain potentially harmful components that could influence their use in combustion processes. Biomass types have different heavy metal accumulation. Herbaceous, fruity and aquatic biomass types have shown the highest levels of Zn and As in combustion tests. These tests did not include hemp or goldenrod, however, these elements should be considered when using herbaceous biomass types for combustion [41]. Canadian goldenrod is an invasive species that can affect the diversity and growth of surrounding plants [42].

The chemical composition of CG ash determined in the laboratory was compared to the data of similar samples available in the literature. The results show that the content of Al, K, Mg, P and S are similar in all the compared studies. Different methods used for the determination of elements also influence the results, but they do show consistency. When considering goldenrod for combustion, attention must be on S, Cl and K contents, as those are the elements that can cause harmful gaseous emissions and promote corrosion and erosion of boilers. The S content of wood and its parts (bark, stems, etc.) used for combustion is below 0.1 wt% [37], which is exceeded in all the studied cases shown in Table 2.

TABLE 2. THE CHEMICAL COMPOSITION OF GOLDENROD

Element	Content, wt%	Content, wt%	Min, wt%	Max, wt%
Al	0.30	–	0.07	1.13
Ca	29.24	25.07	7.97	22.63
K	14.48	29.11	9.47	25.13
Mg	5.27	5.69	1.21	4.57
Mn	1.75	0.29	–	–
Na	0.37	0.40	–	–
P	9.08	4.55	1.55	6.24
S	2.65	1.76	0.67	1.47
Si	35.87	–	0.95	18.33
Zn	0.18	0.14	1.68	3.49
Cl	–	–	0.60	4.09
As, Cd, Cr, Cu, Mo, Ni, Se, Ba, Fe	0.80	0.07	–	–
Plant part	Goldenrod ash	European goldenrod post-extraction residues	CG ash	
Source	Calculated from [43]	Calculated from [44]	XRD, SEM – EDX	

Industrial hemp is a pollution resistant plant suitable for growing in polluted areas to remove metals and remediate the soil. Metal accumulation in hemp does not influence the quality of its fibres and hurds, allowing for easy use of the material. If hemp is used for pellet production, the underground parts, where metals accumulate, should be excluded to minimize the risk of heavy metal emission during combustion [45]. The ability to absorb different elements from the surrounding soil can also be a disadvantage, depending on element accumulation in hemp parts [46]. The heavy metal accumulation in hemp can be ranged as follows: Cr > Cu > Zn > Pb > Cd, with the largest absorption in plant roots. Cu, Cd, Ni and Pb mostly accumulate in leaves. Se is absorbed in hemp seeds and to a lesser extent in leaves. It does not create a threat to organisms so it can be used in production [45]. The optimal harvesting period for hemp for the production of solid fuels is several weeks from February until April, however, additional drying or combustion in a boiler with a condenser should be considered [47].

The chemical composition of HLS ash determined in the laboratory was compared to the data of similar samples available in the literature. The results show that the content of P, Mg, Si, and Al are similar in all the compared studies. There are similarities for Ca and Cl as well, with higher values for S and Zn. When considering hemp for combustion, similarly as in the case of CG, attention must be on S and Cl contents. The S content of wood pellets is below

0.02 wt%, which is exceeded in all the selected studies. The Cl content in wood materials is below 0.05 wt%, which is exceeded in the case of studied materials as well [49], see Table 3.

TABLE 3. THE CHEMICAL COMPOSITION OF HEMP

Element	**Content, wt%**	**Content, wt%**	**Min, wt%**	**Max, wt%**
K	37.28	32.26	5.05	17.93
Ca	41.06	32.05	8.84	29.94
P	5.53	5.91	3.15	8.16
Mg	8.24	4.19	3.00	6.34
Na	0.44	0.92	–	–
Si	2.93	13.41	1.05	10.03
Al	0.84	0.13	0.11	0.44
Mn	0.43	0.03	–	–
Cl	0.62	10.54	0.39	1.14
S	0.003	0.16	0.97	3.32
F	0.41	0.02	–	–
Zn		0.05	1.09	2.29
B, Ba, Cd, Cr, Cu, Fe, Mo, Ni, Sr, Ti, V	<1.13	0.51	–	–
Plant part	hemp fiber ash	hemp silage (dry)	HLS ash	
Source	Calculated from [21]	Calculated from [48]	XRD, SEM – EDX	

As the samples contain various parts of plants, ash is not homogenous, therefore, the concentration of elements varies in different studied sample sections. The range shows the variance of elements in the materials, highlighted by the pictures from SEM analysis (Fig. 2).

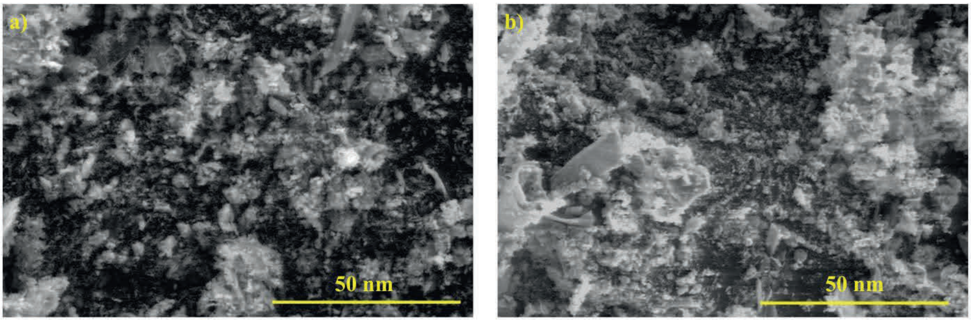

Fig. 2. SEM micrographs of studied ash samples: a) CG ash; b) HLS ash.

The micrographs of both CG and HLS ash samples (Fig. 2a and Fig. 2b) show remnants from the seed shells which have a higher $CaCO_3$ content compared to the adjacent ash. Furthermore, the seed shells did not show any traces of S or Cl. Out of the two, the CG ash sample was more homogeneous compared to the HLS ash sample. According to XRD results, both ash samples mostly consist of carbonates, which is a common occurrence after the combustion process. HLS primarily contains calcite ($CaCO_3$) and quartz (SiO_2). The CG composition is more diverse, consisting of Ca and Mg carbonates as well as K phosphate and Ca hydroxide. EDX shows that HLS contains almost twice the amount of K when compared to CG. As it was not evident in XRD, it means that in HLS K is in an amorphous compound. The amount of S and Cl in both samples is <5 wt%, with HLS containing up to 1.14 wt% Cl while containing more S (up to 3.32 wt%) than CG (Cl up to 4.09 wt% and S up to 1.47 wt%).

3.3. Evaluation of Materials

The studied materials and obtained quality parameters were compared with ENplus B-class wood pellet quality requirements to assess differences between the materials. This comparison allowed to determine the minimal amount of higher-quality material, such as wood, that should be added to the studied alternative materials to reach the requirements of B-class pellets. The moisture content was not compared with pellet requirements as 11 of the studied materials were not acquired in pellet forms and the moisture content can be controlled more easily through material preparation before pelletizing.

Fig. 3. shows the ash content of studied samples along with the reference value of pellets – the red benchmark line. It can be seen that only 3 of the 13 materials have ash content below the benchmark of 2.0 wt%. It means that most of the alternatives will cause additional maintenance if used alone.

Fig. 4. shows the net calorific value of studied samples along with the reference value of pellets – the red benchmark line. Here, a better situation is observed, as 10 samples have values above the benchmark and one material has a net calorific value equal to the benchmark of 16.5 MJ/kg.

The existing results show the actual moisture content of biomass during tests. It is planned to continue research by ensuring the storage of materials in an open space with known climate conditions. This will offer information on the hydrophobic parameters of materials and their ability to absorb or release moisture in the environment. In long term, it will ensure that the moisture content of all materials is similar. The comparison of gross calorific value is not correct in this case, as these values are for perfectly dry materials, when in reality the material will never be perfectly dry during use. The net calorific value describes the amount of energy that can be obtained from the material during combustion at the time of testing in actual conditions. These results highlight the fact that selected alternative materials have a good potential for use in pellet production and afterwards – heat generation processes.

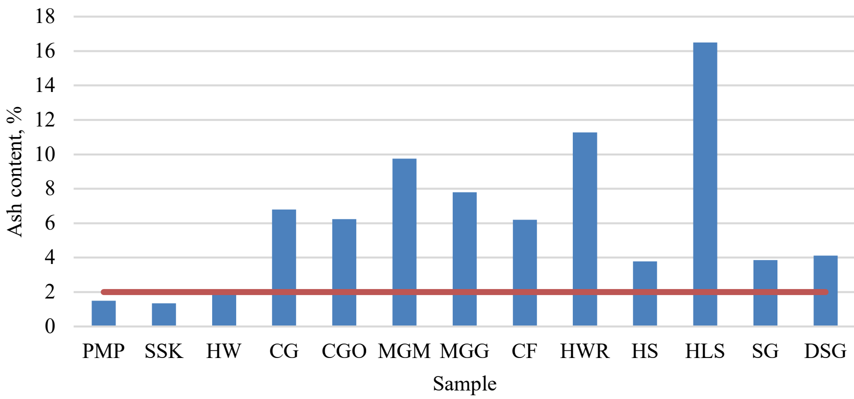

Fig. 3. Comparison of ash content of the studied alternative materials.

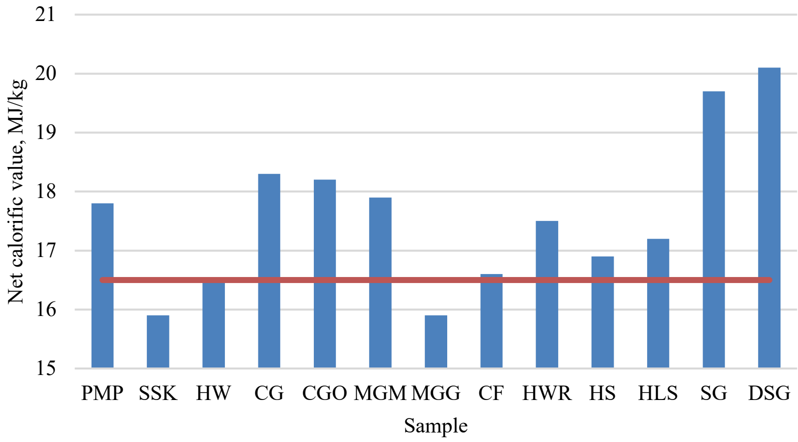

Fig. 4. Comparison of net calorific value of the studied alternative materials.

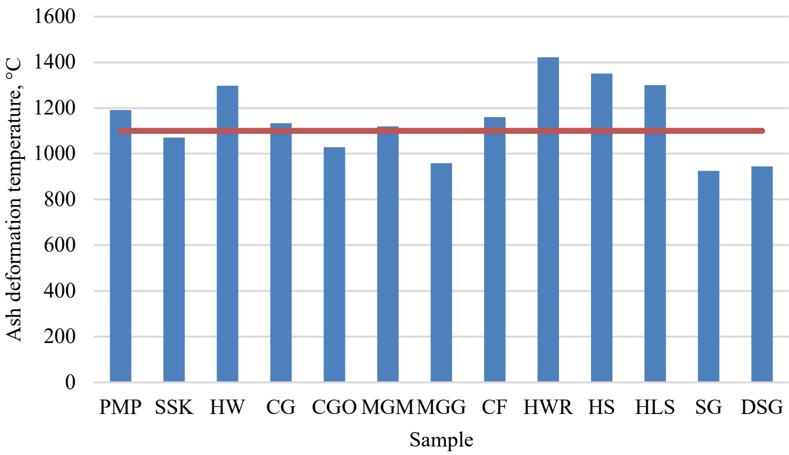

Fig. 5. Comparison of ash DT of the studied alternative materials.

Fig. 5. shows the DT of studied samples along with the reference value of pellets – the red benchmark line. 8 of the samples reach DT above the benchmark of 1100 °C. In this case, the higher the DT results are, the better, as it means reduced slagging risks in the boilers where pellets are used. However, HW is the only material with ash content, net calorific value and ash DT within the limits of wood pellet requirements, suggesting the need for improvement in the quality of pellet composition for other materials by adding wood in a material mix.

4. Discussion

An improvement of the quality of potential pellets can be achieved by using a mixture of materials, for example, mixing an alternative material with wood to improve the quality of the pellets in general. In this case, the maximum fraction of alternative material in a mixture with wood was determined; see Fig. 6 to reach the benchmark for the critical parameter for each alternative, see Table 4. The critical parameter for most materials is the ash content, with SSK having a net calorific value as the critical parameter. PMP and HW are the only materials that can be considered in pellet production without additions, as they show compliance within the ENplus B-class wood pellet requirements.

In the determination of wood addition to the alternative materials a new benchmark was set – the minimum amount of alternative material in the mixture, with the value of 10 %. This value was selected considering the harvesting and preparation of the material to make it feasible to use. The results are displayed in Fig. 6.

TABLE 4. ALTERNATIVE RAW MATERIALS AND THEIR CRITICAL PARAMETERS

Alternative material	**Critical parameter**
Pine mix pellets (PMP)	None
Pellets of wood shavings, synthetic and textile waste mix (SSK)	Net calorific value
Hogweed (HW)	None
Canadian goldenrod (CG)	Ash content
Canadian goldenrod outgrown (CGO)	Ash content
Mowed grass from the municipality (MGM)	Ash content
Mowed grass from the garden (MGG)	Ash content
Chamomile full plant without stems (CF)	Ash content
Hemp without roots (HWR)	Ash content
Hemp stems (HS)	Ash content
Hemp leaves with seeds (HLS)	Ash content
Spent grain (SG)	Ash content
Damaged spent grain (DSG)	Ash content

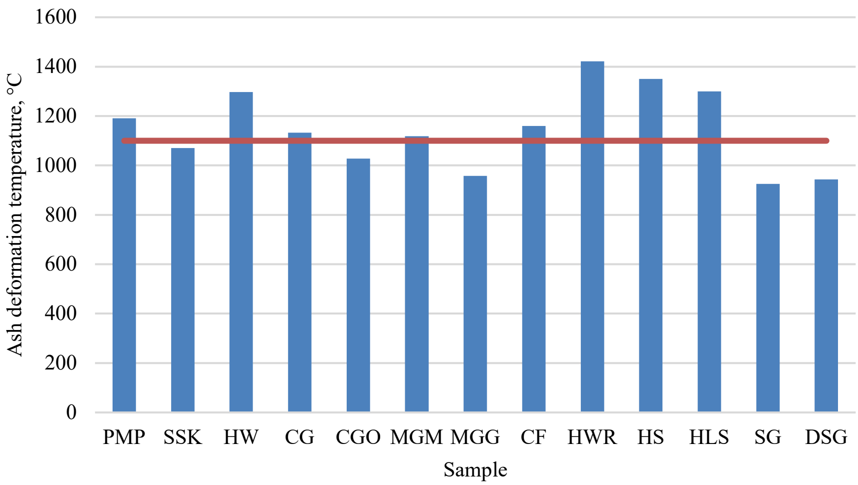

Fig. 6. The maximum alternative material fraction in a mixture with wood to reach quality requirements.

The results show that five of the 13 studied samples are not suitable for pellet production due to low-quality parameters and the necessity to add over 90 % of wood to improve the quality. The materials are CG, MGG, MGM, HWR, and HLS. These results suggest that the use of full plants with their seeds and roots can significantly affect the quality of the product, leading to higher ash content, lower ash melting temperatures and increased slagging risks and maintenance of the combustion equipment. The chemical analysis of CG and HLS enhances this observation as it shows the presence of chemical compounds that increase the ash melting, corrosion and slagging risks when materials are used as fuels. The seeds of plants should be excluded from the materials if possible, as they can significantly impact the quality, as shown by differences between HS and HLS and supported by higher content of carbonates in hemp seeds.

Mowed grass is also unfavourable for use in pellet production as it is a material with more complicated harvesting and preparation. The material collection conditions also significantly influence the quality as rainy, muddy conditions will lead to additional contamination of the grass and the material will require additional drying and treatment to remove the added sand.

PMP and HW are the only materials that can be used without additions. In the case of PMP, it makes sense, as it already is a mixture of woody material. HW shows results that are more surprising. As it is an invasive species in Latvia with wide distribution, the collection of it would be favourable. The tested material mostly consisted of the stem of the plant, so the additional testing including additional plant parts should be considered to support the use of this material further.

All the materials above the benchmark will be considered in pellet production on a laboratory scale to further study their quality in the pellet form and gain additional characteristics regarding their combustion in a boiler and created pollutants, including gaseous emissions and particulate matter concentrations.

5. CONCLUSIONS

Due to growing demand for renewable fuels, the focus is on alternative raw materials for fuel production. In this study, the quality parameters of 13 alternative raw materials are determined and compared to requirements of ENplus B-class wood pellets. Additionally, the fraction of alternative material in a mixture with wood is determined to make alternatives more favourable for use in pellet production.

The results show that out of the 13 studied materials – two - pine wood mix and hogweed - are suitable for pellet production without additions. These are the only materials within the requirements of wood pellets for ash content, net calorific values and ash deformation temperature.

Five of the studied materials have proven to be unfavourable for pellet production – Canadian goldenrod, mowed grass for garden and municipality, hemp with roots and hemp with leaves and seeds. These results suggest that additional impurities, such as sand and some plant parts, like roots and seeds, should be avoided or limited in material collection and preparation. Roots might add on to impurities in the material, while seeds might have completely different chemical composition that affect ash content, formation and melting. This is supported by chemical and SEM analysis of ash from hemp leaves with seeds, as the shells of seeds can be distinguished in the SEM micrographs and seeds also show higher concentration of carbonates, that impacts ash melting.

Additional chemical analysis has to be considered for the raw materials, their ash and particulate matter to fully characterize the material. Further studies will be carried out focusing on the pelletizing of alternative materials in laboratory environment and testing of pellets in a boiler system to gain better insight in the possible expansion of the pellet market.

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