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SPRUCE AND BARLEY ELEMENTAL AND STOCHIOMETRIC ANALYSIS AFFECTED BY THE IMPACT OF PELLET PRODUCTION AND TORREFACTION

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Biomass is a potential biofuel which may help fighting high carbon dioxide emissions and negative impacts of global warming. Analysis of Norway spruce (*Picea abies*) and barley (*Hordeum vulgare*) were performed at the laboratory of Czech University of Life Sciences. Material was torrefied in an inert nitrogen atmosphere at the temperatures of 250 °C and 280 °C for 45 minutes. Elementary and stoichiometric parameters were monitored and impact of torrefaction and pellet production on carbon footprint was determined. Torrefied and pelleted material showed better fuel properties in comparison to the original material. Calorific value of the torrefied spruce wood chip increased by 12.27% when torrefied at the temperature of 250 °C, and by 25.41% when torrefied at the temperature of 280 °C.

Keywords: biomass; biofuel; carbon footprint; greenhouse gas; calorific value

The Regulation 2018/1999 of the European Parliament and Council on the Governance of the Energy Union and Climate Action sets a Europe-wide target of 43% reduction of greenhouse gas emissions compared to 2005 levels. Czech Republic's National plan has a goal to reduce greenhouse gas emissions by 30% by 2030 (compared to 2005).

Following the statistics of Global Carbon Project organization, Czech Republic was on the 27th position in global ranking out of all 221 countries, with average consumption 10.4 Mt of CO₂/capita in 2015, which is a big warning and motivator for a behavioural change.

Help may be found in the increased usage of biomass for energy purposes, production of which brings many environmental benefits that contribute to climate change mitigation. Biomass is a renewable material that is considered carbon neutral and thus helping to counteract the greenhouse gas emissions (García et al., 2019). Utilization of biomass made of materials from agricultural production and waste biomass, the potential of which cannot be neglected, comes to the forefront and is very topical (Kažimírová et al., 2020). Importance of renewable energy sources becomes one of the major factors for permanently sustainable development not only in agriculture, but also in society as a whole (Gürdil et al., 2009).

Due to photosynthesis, forest and agricultural biomass is considered to have neutral carbon footprint and low content of sulphur and nitrogen, which leads to low emissions of NO_x and SO₂ during its combustion. Biomass combustion is an important part of future energy concepts (Stelte et al., 2011).

Despite the advantages of biomass as a suitable heating medium, there are issues such as high moisture reducing combustion power, difficult comminution to small particles

and relatively low bulk density, as well as increasing storage, transport, and handling requirements. Pellet production and torrefaction of these materials directly solves the aforementioned disadvantages (Kaliyan and Vance Morey, 2009).

The torrefaction, such as low-temperature pyrolysis, is a controlled carbonization process, in which biomass is heated in the temperature range of 230 °C to 340 °C in an oxygen-free environment or in the presence of only small amount of oxygen (Tamelová et al., 2021). This process leads to a reduction in the biomass moisture content and biomass conversion into a product with similar properties to coal. The torrefied material is further processed into pellets or briquettes intended for energy use (Malaťák et al., 2020a; Gendek et al., 2018).

Torrefied biomass has three main advantages over traditional biomass: higher calorific value (amount of energy per unit of mass), higher energy density (amount of energy per unit of volume) and improved range of physical properties, such as dimensional stability, homogeneity, and hydrophobic behaviour. Furthermore, especially in terms of plant biomass, the issues related to combustion of large and small sources, e.g., fouling, sintering, chlorine corrosion, are eliminated (Malaťák et al., 2020b; Skanderová et al., 2015). On the other hand, with biomass torrefaction, there are losses of approx. 10% of the energy accumulated in the volatile combustible, which is released when heated to temperatures above 300 °C. However, this disadvantage is richly compensated by the advantages described above (Stupavský, 2012). The least resistant to torrefaction are wood polyoses – hemicelluloses, which decompose at the temperature range of 170–240 °C. The most durable component of wood is lignin. The active decomposition of

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lignin takes place at temperatures of 300–400 °C (Kučerová et al., 2011).

The higher calorific value of torrefied material is due to the decreasing oxygen content. In the original state, the oxygen to carbon (O/C) ratio in wood biomass is more than 1. The torrefaction process brings this ratio below 1, and with increasing torrefaction temperature, O/C ratio of less than 0.4 can be achieved. As a result, the calorific value increases from 17 MJ·kg⁻¹ for raw wood biomass to 27 MJ·kg⁻¹ for torrefied biomass (Wilk et al., 2016). As the oxygen content of torrefied material decreases, the amount of hydrogen decreases at the same time. Due to the decrease in oxygen and hydrogen contents in material – more precisely due to the decrease in hydroxyl groups in the material, which occurs during thermal treatment – the resulting material shows hydrophobic behaviour (Bates and Ghoniem, 2012). Promising published results suggest that the potential of torrefaction technology is significant (Poudel et al., 2015). It is possible to improve the mechanical and burning qualities of final biofuels by adding the wooden biomass to the herbage-based agro-pellets production (Souček et al., 2019).

Aim of this paper is to evaluate and confirm the positive impacts of torrefaction and pellet production on elemental and stoichiometric properties of biomass as a potential biofuel in households and industries.

Material and methods

The samples of wood chips of Norway spruce (*Picea abies*) were collected from the University Forest Establishment in Kostelec nad Černými lesy.

The samples of barley (*Hordeum vulgare*), delivered for the Suchdolský Jeník brewery, did not fulfil its original purpose due to the lower beer demand caused by Covid restrictions. Original various size samples of spruce wood chips and barley were grinded to a maximum particle size of 1 mm and analysed.

Torrefaction

Torrefaction is a process in which the fuel properties of lignocellulosic

biomass are improved. This method converts non-uniform biomass into a highly homogeneous bioenergetic material (Adams et al., 2015).

The material analysed was torrefied in electric cylindrical furnace (torrefaction unit). The torrefaction process took place at two different average temperatures of 250 °C and 280 °C with a total time of 1 hour in the furnace (Figure 1 – A). Due to the furnace proportions, the actual time in it at a given temperature was approx. 45 minutes. To follow the spontaneous combustion, inert nitrogen from the pressure bottle was released into the furnace.

The material entered the torrefaction unit through a hopper (Figure 1 – B) and was transported into the furnace by a self-propelled screw conveyor (Figure 1 – C), which was connected to a hopper via dispenser. The furnace was heated by resistance electric heaters and insulated with aluminium foil. The furnace shell includes temperature wells for the operating temperature management.

Pellet production

Biomass pellet production simplifies handling and reduces transport and storage costs. It is mainly used to increase the energy density of biomass and to create a uniform shape that will facilitate the use of biomass in domestic and industrial energy-related applications, including combustion and gasification. Additionally, biomass fuel pellets have standardized and uniform properties (Monedero et al., 2015).

A small pellet press with a disk matrix was selected for pelletizing the samples. The material was moistened to 15% before the actual pellet production. This moisture was maintained throughout the entire process of pellet production by the Ohaus Moisture Analyzer MB23 device. Final pellets had dimensions of approx. 7 mm in diameter and 21 mm in length.

Analysed and pelletized mixtures:

- Spruce wood chips (66%) + Malting barley (33%) – SAMPLE A.

Fig. 1 Torrefaction unit

- Spruce wood chips torrefied at 250 °C (66%) + Malting barley (33%) – SAMPLE B.
- Spruce wood chips torrefied at 280 °C (66%) + Malting barley (33%) – SAMPLE C.

Elemental analysis

The analysis of the samples took place at the laboratories of the Department of Technological Equipment of Buildings, Faculty of Engineering, Czech University of Life Sciences in Prague. The analysis focused on the contents of water and ash, combustion heat and elemental composition. Analytical samples were formed from the primary biomass, dried biomass, biochar, and pelletized mixtures. Original samples were ground by Retsch SM100 grinder to a maximum particle size of 1 mm.

The moisture content was determined using a LECO TGA701 thermogravimetric analyser. According to the ISO EN 18134-3, 1 gram of each sample was dried in a weighing oven at 105 °C until the level of constant weight.

The ash content in samples was also determined in a weighing furnace by annealing the same sample at 550 °C until the level of constant weight at elevated oxygen concentrations in accordance with the ISO EN ISO 18122.

Combustion heat was measured in an Loper AC600 isoperibolic calorimeter by combustion at a reference temperature of 28 °C and a charge pressure in a bomb of 3 MPa. Stainless steel cups were used for the measurement and cotton thread was used for ignition. The device was calibrated with benzoic acid. The conversions of combustion heat and calorific value were based on the ISO EN ISO 1928.

The representation of elements was measured with the LECO CHN628 + S analyser using the LECO instrumental combustion method for biomass. To determine the C, H and N values, a 0.1 g sample was burned in oxygen at 950 °C. This apparatus was calibrated with ethylenediaminetetraacetic acid, rice and rye flour.

The total conversions of individual elements at dry sample state without ashes and combustibles were determined according to the ISO EN ISO 16993. Oxygen was determined by the method of calculation up to 100% of the dry sample.

Stoichiometric analysis

Stoichiometric calculations are considered a basis for any thermal calculation. They play an important role in controlling the operation of combustion plants and are necessary for comparison of the theoretical and actual combustion processes. They exploit the results of previous analyses (Malaták et al., 2020b).

These calculations determine:

- Mass combustion:
 - theoretical amount of air for perfect combustion ($\text{kg}\cdot\text{kg}^{-1}$);
 - theoretical mass amount of dry flue gas ($\text{kg}\cdot\text{kg}^{-1}$);
 - theoretical mass concentration of carbon dioxide in dry flue gas (%).
- Volumetric combustion
 - theoretical amount of air for perfect combustion ($\text{m}^3\text{N}\cdot\text{kg}^{-1}$);
 - theoretical volumetric amount of dry flue gas ($\text{m}^3\text{N}\cdot\text{kg}^{-1}$);
 - theoretical volumetric concentration of carbon dioxide in dry flue gas (%).

Results and discussion

Analysis of primary material

Mixture of spruce wood chips and grain was calculated according to the ratio of dry matter (2 : 1), as the mixture itself was prepared in a later step. Table 1 shows the basic analysis of primary material. Spruce wood chip showed a very high moisture content leading to lower content of other elements in comparison with barley. Samples were measured 10 times each, with standard deviation in the interval from 0.01% to 1.89%.

Analysis of dried and torrefied material

Dried and torrefied material was analysed in the same scope as original samples. Table 2 shows a reduction in the material moisture content. Dried and torrefied material consists of increased content of carbon leading to the increase in combustion heat and calorific value of material. Material torrefaction decreased the content of hydrogen and oxygen. As a negative effect of torrefaction, the higher temperature of torrefaction process is accompanied by a higher ash content in the sample. This result is in line with evaluation of Pimchui et al. (2010) and Rudolfsson et al. (2015), who analysed that torrefaction pre-treatment increases the efficiency of biomass combustion processes.

As shown in Table 2, it is possible to observe a significant increase in carbon content and higher calorific value in the torrefied material. The material torrefied at an average temperature of 280 °C shows higher carbon content and calorific value than the material torrefied at an average temperature of 250 °C. Torrefaction at temperatures higher than 280 °C has not been tested, because the degradation

Table 1 Elemental analysis of primary material

Sample	Humidity (%)	Ash (%)	C (%)	H (%)	N (%)	S (%)	O (%)	Combustion heat ($\text{MJ}\cdot\text{kg}^{-1}$)	Calorific value ($\text{MJ}\cdot\text{kg}^{-1}$)
Spruce wood chip – original	41.36 (± 1.89)	0.21 (± 0.02)	30.22 (± 0.13)	3.54 (± 0.02)	0.14 (± 0.01)	0.00	24.53	11.80 (± 0.06)	10.02
Barley – original	14.52 (± 0.76)	1.90 (± 0.03)	38.22 (± 0.14)	5.51 (± 0.02)	1.35 (± 0.02)	0.09	38.41	15.24 (± 0.08)	13.68
SAMPLE A – original	32.41 (± 1.51)	0.86 (± 0.02)	32.81 (± 0.13)	4.26 (± 0.02)	1.14 (± 0.01)	0.00	28.53	12.89 (± 0.07)	11.17

Table 2 Elemental analysis of dried and torrefied material

Sample	Humidity (%)	Ash (%)	C (%)	H (inc. water; %)	H (in combustible part; %)
Spruce – dried	7.19 (±0.09)	0.33 (±0.01)	47.83 (±0.15)	6.41 (±0.02)	5.61 (±0.02)
Spruce – torrefied 250 °C	4.14 (±0.07)	0.92 (±0.01)	53.12 (±0.09)	5.98 (±0.03)	5.61 (±0.03)
Spruce – torrefied 280 °C	3.38 (±0.06)	1.40 (±0.02)	58.83 (±0.12)	5.73 (±0.03)	5.27 (±0.03)
Barley – dried	6.67 (±0.07)	2.08 (±0.02)	41.73 (±0.18)	6.76 (±0.02)	6.01 (±0.02)

Sample	N (%)	S (%)	O (in combustible part; %)	Combustion heat (MJ·kg ⁻¹)	Calorific value (MJ·kg ⁻¹)
Spruce – dried	0.23 (±0.01)	0.00	38.82	18.67 (±0.07)	17.28
Spruce – torrefied 250 °C	0.20 (±0.02)	0.00	36.77	20.70 (±0.12)	19.40
Spruce – torrefied 280 °C	0.23 (±0.01)	0.00	30.13	22.91 (±0.10)	21.67
Barley – dried	1.47 (±0.02)	0.10	41.94	16.64 (±0.08)	15.17

Table 3 Stoichiometric analysis of dried and torrefied material

	Spruce – dried	Spruce – 250 °C	Spruce – 280 °C	Barley – dried
Mass combustion				
Theoretical amount of air for perfect combustion (kg·kg ⁻¹)	3.64	6.68	7.60	5.07
Theoretical mass amount of dry flue gas (kg·kg ⁻¹)	5.85	9.06	9.99	7.37
Theoretical mass concentration of carbon dioxide in dry flue gas (%)	18.93	22.25	22.53	20.76
Volumetric combustion				
Theoretical amount of air for perfect combustion (m ³ N·kg ⁻¹)	3.64	6.68	7.60	5.07
Theoretical volumetric amount of dry flue gas (m ³ N·kg ⁻¹)	2.75	5.04	5.71	3.83
Theoretical volumetric concentration of carbon dioxide in dry flue gas (%)	20.39	20.24	19.93	20.20

Table 4 Elemental analysis of pelletized material

Sample	Humidity (%)	Ash (%)	C (%)	H (inc. water; %)	H (in combustible part; %)
Sample A – dried	7.01 (±0.05)	0.91 (±0.01)	45.79 (±0.12)	6.53 (±0.04)	5.74 (±0.04)
Sample B – dried	4.98 (±0.04)	1.31 (±0.01)	49.33 (±0.11)	6.24 (±0.05)	5.74 (±0.04)
Sample C – dried	4.47 (±0.05)	1.62 (±0.02)	53.13 (±0.17)	6.07 (±0.04)	5.52 (±0.03)
Sample A – pellets	8.86 (±0.07)	1.16 (±0.01)	44.24 (±0.12)	6.74 (±0.05)	5.75 (±0.05)
Sample B – pellets	7.83 (±0.06)	1.54 (±0.02)	45.63 (±0.13)	6.55 (±0.04)	5.67 (±0.04)
Sample C – pellets	7.67 (±0.06)	1.44 (±0.01)	47.61 (±0.11)	6.48 (±0.04)	5.63 (±0.04)

Sample	N (%)	S (%)	O (in combustible part; %)	Combustion heat (MJ·kg ⁻¹)	Calorific value (MJ·kg ⁻¹)
Sample A – dried	0.64 (±0.01)	0.03	39.86	17.99 (±0.08)	16.57
Sample B – dried	0.62 (±0.01)	0.03	38.50	19.34 (±0.07)	17.99
Sample C – dried	0.65 (±0.01)	0.03	34.07	20.82 (±0.08)	19.50
Sample A – pellets	1.53 (±0.02)	0.00	38.47	17.38 (±0.11)	15.92
Sample B – pellets	1.35 (±0.02)	0.00	37.97	17.90 (±0.10)	16.48
Sample C – pellets	1.48 (±0.02)	0.00	36.18	18.87 (±0.09)	17.46

Table 5 Dry state material theoretical conversion

Sample – dry state material	Ash (%)	C (%)	H (%)	N (%)	S (%)	O (%)	Combustion heat (MJ·kg ⁻¹)	Calorific value (MJ·kg ⁻¹)
Spruce	0.36	51.53	6.04	0.25	0.00	41.83	20.12	18.80
Spruce – 250 °C	0.95	54.98	5.80	0.21	0.00	38.06	21.42	20.16
Spruce – 280 °C	1.46	61.37	5.50	0.24	0.00	31.43	23.91	22.71
Barley	2.23	44.71	6.44	1.58	0.11	44.94	17.83	16.42
Sample A – material	0.98	49.25	6.17	0.69	0.04	42.87	19.36	18.01
Sample B – material	1.38	51.56	6.02	0.66	0.04	40.35	20.23	18.91
Sample C – material	1.71	55.82	5.81	0.69	0.04	35.93	21.88	20.61
Sample A – pellets	1.27	48.54	6.30	1.68	0.00	42.21	19.08	17.70
Sample B – pellets	1.67	49.51	6.16	1.47	0.00	41.20	19.43	18.08
Sample C – pellets	1.56	51.56	6.09	1.60	0.00	39.18	20.44	19.11

Table 6 Stoichiometric analysis of pelletized material

	Sample A – material	Sample B – material	Sample C – material	Sample A – pellets	Sample B – pellets	Sample C – pellets
Mass combustion						
Theoretical amount of air for perfect combustion (kg·kg ⁻¹)	4.11	6.14	6.75	5.41	5.56	5.85
Theoretical mass amount of dry flue gas (kg·kg ⁻¹)	6.36	8.50	9.11	7.72	7.89	8.18
Theoretical mass concentration of carbon dioxide in dry flue gas (%)	19.54	21.76	21.94	21.01	21.22	21.34
Volumetric combustion						
Theoretical amount of air for perfect combustion (m ³ N·kg ⁻¹)	4.11	6.14	6.75	5.41	5.56	5.85
Theoretical volumetric amount of dry flue gas (m ³ N·kg ⁻¹)	3.11	4.64	5.08	4.08	4.20	4.41
Theoretical volumetric concentration of carbon dioxide in dry flue gas (%)	20.33	20.23	20.02	20.09	20.14	20.00

of hemicelluloses and the loss of strength of subsequently produced pellets has already been observed by Rejdak et al. (2019).

The calorific value of torrefied spruce wood chip was increased by 12.27% when torrefied at 250 °C and by 25.41% when torrefied at 280 °C.

The torrefaction shortcoming lies in the higher ash content in fuel. As shown in Table 2, the ash content increased from 0.33% of the pure material weight to 1.4% of the torrefied material weight at 280 °C. Negative effect of torrefaction is also shown in stoichiometric analysis in Table 3 stating the higher demand for air necessary for perfect combustion and higher content of carbon dioxide in dry flue gases.

The analysis result is consistent with Arias et al. (2008) who observed that, thanks to torrefying the fuel material, the biochar showed higher calorific value, low moisture content and it became more brittle than the original biomass.

Analysis of pelletized material

Pellet production has a slightly negative effect on the elemental material composition (Table 4). Produced pellets showed a lower carbon content and calorific value than pure non-pelletized material. The pellets also had a higher ash, water and nitrogen contents.

The final pellet had a slightly higher moisture content (7–8%) in contrast to the input material (4–7%). This was caused by drying of the input material and its subsequent wetting to 15% moisture during pellet production. By increasing the moisture content during pellet production, the carbon content decreased slightly, leading to a decrease in combustion heat and calorific value of the material. Pellet production led to an increase in the nitrogen and ash contents for samples A and B. Sample C showed lower ash content after pellet production. Pellet moisture content was measured 2 weeks after their production, samples were stored in faculty laboratory hall.

Considering the moisture content, the pellets showed a significant decrease in contrast to the moisture content of

the primary raw material of spruce wood chip (41.36%) and barley (14.52%). Furthermore, in comparison to the original material, the combustion heat and calorific value were almost 45% higher.

Pellet burnt better than the original material due to the lower moisture content in pellets and the higher concentration of energy value.

A more precise analysis of fuel composition can be conducted by theoretical conversion of the material to zero moisture (Table 5).

With material torrefaction, an increasing ash content can be observed, which is undesirable in fuel. Ash may block the air supply into the heating chamber and may melt and destroy the stove in combination with high temperatures (Malaťák and Vaculík, 2008).

The carbon content also increases with material torrefaction, which results in an increase in combustion heat and calorific value. Torrefaction has opposite effect on hydrogen and oxygen, contents of which decrease.

As a cereal, barley has a high ash content and a low carbon content, which result in low calorific value and combustion heat.

Pellet production resulted in an increase in undesired ash and decrease in carbon content, combustion heat and calorific value.

On the other hand, pellet production showed a favourable result in terms of stoichiometric analysis (Table 6). It had a positive impact on the amount of air demand for perfect combustion and carbon dioxide content in dry flue gas in the torrefied material. Its negative effects included increased amount of air demand for perfect combustion and increased carbon dioxide content in dry flue gas in non-torrefied original dried material.

Conclusions

Tests conducted at the laboratory of the Faculty of Engineering, Czech University of Life Sciences in Prague proved the positive effect of material torrefaction and pellet production of on fuel properties.

Considering the elemental and stoichiometric analysis results, spruce wood chip in combination with torrefaction showed improved combustion heat and calorific value; in combination with pellet production, improved stoichiometric attributes, making it an appropriate form of biofuel suitable for both domestic and industrial usage.

Despite having higher ash content and lower combustion heat and calorific value in comparison to spruce wood chip, barley still may be used for energetic purposes, if not used otherwise.

Mass utilization of pellets made of torrefied material as biofuel thus may help to meet the goals of greenhouse gas emissions reduction by 30% in 2030 (compared to 2005), as planned in the Czech Republic's National Plan.

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