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Full Length Research Paper

Evaluating the Energy Contributions of Key Biomass Residues in Croatia

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This paper examines the total quantities of combustible (carbon, hydrogen, and sulphur) and non-combustible matters (oxygen, nitrogen, moisture, and ash) contained in biomass of all major forest crops (oak, beech-tree, maple, yoke elm and fir), woody horticultural crops (apple, grapevine, sour-cherry, plum, and hazelnut) and arable crops (wheat, soybean, rapeseed, and sunflower) in the territory of Croatia. Also, in addition to briquette production, pelleting is the most often used technology in the biomass treatment. For this purpose, pellets were produced from these sorts of biomass and analyzed according to DINplus standard requirements (durability, bulk density, diameter, length, ash content, nitrogen content, sulphur content, chlorine content, volatiles content, coke content, fixed carbon content and lower energy value).

Key words: Renewable energy sources, agricultural biomass, forest biomass, pellets

INTRODUCTION

A persistent increase in prices of fossil fuels and negative impact of these fuels on global climate as well as raising awareness about need to improve the environment management and to achieve secure and stable supply of energy will lead to increasing utilisation of biomass in the world. Considering the existing energy scenarios (lowering stocks of oil and gas as primary non-sustain-able energy sources) and on the environment related goals of both the world-wide policies and the EU policies, it is expected that during the next decade, the production and consumption of biomass will be growing on a global scale (van Dam et al., 2008; Voca et al., 2007). Therefore, renewable energy sources should both substitute fossil fuels generated energy and become a by-pass to the utilisation of some other fuel of the future (Tomic et al., 2008).

Main sources of biomass are the residuals from agricultural production and forestry, organic waste, food processing waste, energy crops and lately, algae, fungi and yeasts. Further growth of the biomass production (in addition to algae, fungi and yeast) should be related to a more intensive use of relatively recent energy crops, such as fast-growing varieties of trees (such as aspen and osier) arable crops and grasses (such as Miscanthus and Sudan grass). The aim of introducing new plants is to find adequate crops which will be acceptable in terms of environment and economy, particularly in those climatic regions where it is not possible to achieve acceptable biomass yields with production of conventional crops (van Dam et al., 2007; Demirbas et al., 2009).

Biomass can be converted into useful energy forms via several processes. The choice of the conversion process depends on type, properties and quantity of available biomass, on desired final energy form, environmental standards and economic conditions. Biomass can be converted into three main products: energy for heating, transport fuel, and chemical raw materials (Saxena et al., 2009). Biofuels derived from biomass are considered as the most pro-mising alternative fuel sources because they are renewable and environmentallly friendly (Hossain et al., 2010).

Combustion is the most important technology which is used for producing heat and energy from biomass. It is generally a process that gives a number of economic and environmental benefits (Erol et al., 2010). Given the low density of biomass due to its bulkiness, the European Union set out the obligation to treat biomass by various

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technologies, the most often used ones being briquetting and pelleting (Wolf et al., 2006).

Pelleting is a 2hermos-plastic process of shaping pellets by compression, where the particles of the raw material form compact pellets. The most important feature of biomass pelleting, when it comes to the energy production, is obtaining the quality product of higher density, higher heating value per volume, which leads to lower transport and storage costs (Holtz, 2006). Pellets produced in this way have standard shape, with diameter of 6 to 8 mm and length of 12 to 15 mm (Mani et al., 2006). Other important features of good quality pellet are its density and resistance to wear and tear, that is, good durability.

The pelleting process and quality of pellets very much depend on physical-chemical diversities of the pelleted biomass, which is especially evident when residuals from agricultural crops, which are exceptionally non-homogeneous, are used for pellet production. If pelleting conditions are defined properly, then the pellet burning does not produce dust, which is commonly a cause of many technical problems during the charging raw material into furnaces and during the combustion (Holt et al., 2006).

The most important standards regarding wood pellets are: DIN-standard 51731, DIN-Plus and Ö-standard M 7135. Introduction of these standards laid down the fundaments of functional and economical use of pellets (Holtz, 2006). The use of renewable energy sources becomes increasingly important since they do not contribute to higher levels of carbon dioxide (CO_2) which, among other, leads to global warming. Compared to combustion of fossil fuels, biomass combustion produces much lower amounts of nitric oxide (NO) and sulphur dioxide (SO_2) (Cuiping et al., 2004).

Namely, complete biofuel combustion produces only insignificant amounts of carbon dioxide (CO_2) and water (H_2O) which are not harmful for the human health and the environment, while non-complete combustion releases the harmful pollutants and greenhouse gases (GHG), such as carbon monoxide (CO), nitrous oxide (N_2O), methane (CH_4) and polycyclic aromatic hydro carbonates (PAHs) (Bhattacharya and Salam, 2002). Biomass as a renewable energy source is almost CO_2 neutral, and average heating value of bioenergy crops is comparable to that of brown coal. In general, by substituting coal with biomass, it is possible to achieve a 93% decrease of net CO_2 emission per unit of heating value and 84% decrease of this emission by using CHP process where natural gas would be replaced with biomass (Eldabbagh et al., 2005).

Compared to brown coal, biomass has low sulphur-and ash contents, which gives low Sox emissions as well as lower amount of particles during the combustion process. In some instances, the nitrogen content in biomass fuel is high which may result in rather high Nox emissions (Klason and Bai, 2007; Van den Broek, 2000).

In order to compare energy values of investigated biomass, it is necessary to investigate combustible and non-combustible matters contained in it. In biomass, the combustible matters are carbon, hydrogen, and chlorine, whereas oxygen, nitrogen, moisture, and ash are non-combustible ones. Carbon is the most important combustible component and heating value of biomass increases with its CO_2 content. Hydrogen is the second most important element and the part of hydrogen which is bonded to carbon, so called free hydrogen, is active in formation of biomass, creating water and releasing heat, and thus increasing the heat value of the fuel. Sulphur and chlorine are undesirable elements, and in biomass they are found in traces. Sulphur is highly harmful to the environment if bonded to an organic matter (Obernberger and Thek, 2004).

Oxygen is also an undesirable element in the biomass because it bonds carbon and thus lowers the heating value of the fuel. Nitrogen does not develop heat nor it participates in the combustion process and, like oxygen, lowers the heating value of the fuel (Vassilev et al., 2010).

Ash consists of non-combustible mineral particulates, and with the higher ash content the quality of fuel becomes poorer. In terms of fuel quality, it is also necessary to investigate the level of fixed carbon, volatile matters and heating value (Wiinikka et al., 2007). Fixed carbon is one of the most important parts of fuel and it represents firmly bonded carbon. The fuels with higher volatile matter content have lower energy value, that is, they need higher activation energy than the fuels containing less volatile matters (Holtz, 2006). All these components influence the energy value of fuel (MJ/kg), and convert into amount of heat obtained during combustion (Obernberger and Thek, 2004). Therefore, the aim of this investigation was to determine the quantities of combustible and non-combustible matters, as well as to determine and compare the quality of pellets produced from biomass of major forest, arable and woody horticultural crops residues in the Republic of Croatia.

MATERIALS AND METHODS

The crops used in this investigation are the most widespread forest crops, arable crops and woody horticultural crops in Croatia. By type, the analyzed crops were: Forest crops: oak, beech, maple, yoke elm and fir; arable crops: maize, wheat, soy bean, rapeseed and sunflower; woody horticultural crops: apple, grapevine, sour cherry, plum and hazelnut.

The samples of the continental crops were chosen randomly in the forests, orchards and arable fields in eastern Croatia, while grapevine samples come from the south of Croatia. After sampling, all investigated biomass was ground in an “IKA MF 10” grinder at <0.5 mm of average diameter. The ground samples were spread in a thin layer and naturally dried for several days. The investigated biomass samples were analyzed for lower heating value (adiabatic calorimeter C IKA 200), moisture content (NREL/TP-510-42621), volatile matter (CEN / TS 15148:2005), fixed carbon and ash contents (NREL/TP-510-442). Contents of carbon, hydrogen, nitrogen and sulphur were determined by use of CHNS analyzer

Table 1. Chemical analysis of the investigated pruned biomass for average values (in dry matter).

Sample group	Biomass sample	Moisture (%)	Ash (%)	C_{fix} (%)	Volatile matter (%)	Lower heating value (MJ kg^{-1})
Forest crops	Oak	3.06^{l}	0.49^{ji}	18.14^{ef}	79.68^{a}	18.166^{d}
	Beech	7.89^{d}	0.38^{ji}	17.05^{i}	67.50^{i}	19.218^{b}
	Maple	3.79^{k}	0.45^{j}	14.61^{k}	74.73^{d}	18.752^{c}
	Yoke elm	9.46^{a}	0.59^{i}	15.46^{j}	75.27^{c}	17.233^{gf}
	Fir	7.61^{e}	0.51^{i}	18.05^{ef}	66.91^{j}	19.976^{a}
Arable crops	Maize	8.21^{c}	5.65^{c}	18.81^{c}	60.73^{k}	16.469^{i}
	Wheat	4.64^{i}	5.85^{b}	17.81^{gf}	75.44^{c}	16.443^{i}
	Soy bean	7.88^{d}	8.76^{a}	18.58^{cd}	59.90^{l}	15.746^{j}
	Rapeseed	4.09^{j}	3.35^{e}	18.35^{ed}	78.25^{b}	14.617^{k}
	Sunflower	5.41^{h}	4.74^{d}	17.28^{ih}	73.43^{e}	17.776^{e}
Woody horticultural crops	Apple	6.55^{f}	2.56^{g}	17.58^{gh}	73.52^{e}	16.883^{h}
	Sour cherry	6.56^{f}	2.98^{f}	18.77^{c}	72.66^{f}	17.106^{g}
	Plum	7.74^{de}	3.51^{e}	21.19^{a}	69.72^{g}	16.718^{h}
	Hazelnut	5.94^{g}	2.20^{h}	17.28^{ih}	73.43^{e}	17.368^{f}
	Grapevine	8.61^{b}	2.94^{f}	19.24^{b}	69.10^{h}	16.441^{i}
		$\overline{X}$ = 6.49*** LSD = 0.27	$\overline{X}$ = 2.99*** LSD = 0.17	$\overline{X}$ = 17.88*** LSD = 0.35	$\overline{X}$ = 71.35*** LSD = 0.46	$\overline{X}$ = 17.26*** LSD = 0.21

Data are averages ± SD of three determinations. Different letters within a column indicate significant differences at the 5% level by Duncan test; n.s. = nonsignificant. *Significant with P ≤ 0.05, **Significant with P < 0.01, ***Significant with P < 0.001.

(CEN / TS 15104:2005; CEN / TS 15289:2006).

The investigated samples were pelleted by use of a lab pelleting device (Pellet Press 14-175; Amandus Kahl), and the pellets were analyzed according to DINplus standard requirements: mechanical durability (CEN/TS 15210-1:2005), bulk density (CEN/TS 15103:2005), diameter, length, ash content (NREL/TP-510-42622), nitrogen content (CEN/TS 15104:2005), sulphur content (CEN/TS 15289:2006), chlorine content (CEN/TS 15289:2006), volatile matters (CEN/TS 15148:2005), coke content (NREL/TP-510-42622), fixed carbon content (NREL/TP-510-42622) and lower heat value (CEN/TS 14918:2005).

All analyses were performed in three replications, and average values were calculated for each individual analysis.

RESULTS

In order to compare between different types of biomass, the analyses were performed for combustible and non-combustible elements which are obtained by chemical analysis of the investigated crops (Tables 1 and 2). The obtained results shown in Table 1 showed that the investigated samples contained moisture in a range from 3.06 to 9.64%. The highest moisture level was in the samples of walnut (9.64%) and grapevine (8.61%), while the lowest moisture was found in the samples of oak (3.06%) and maple (3.79%).

The content of ash differed from one type of investigated samples to another and ranged from 0.38 to 8.76%. The highest amount of ash was present in soy bean (8.76%) and wheat (5.86%), and the lowest ones were found in beech (0.38%) and maple (0.45%). Fixed carbon, in form of firmly bonded carbon, was found to be in a range from 14.61 to 21.19%. The highest amounts were found in plum (21.19%) and grapevine (19.24%), and the lowest ones in maple (14.61%) and yoke elm (15.46%).

Volatile matter varied from 59.90 to 79.69%. The highest content of volatile matter was found in oak (79.68%) and rapeseed (78.25%), while the lowest one was found in soy bean (59.90%) and maize (60.73%). Lower heating value of the investigated pruned biomass was between 14.617 and 19.976 MJ/kg. The highest value was found in fir (19.976 MJ/kg) and beech (19.218%), and the lowest ones in rapeseed (14.617 MJ/kg) and soy bean (15.746 MJ/kg).

Considering the data shown in Table 2, the level of carbon varies from 46.04 to 52.81%, and given the fact that heating value of fuel grows with carbon level, the best fuel properties, or the highest carbon content, were found in fir (52.81%) and beech (54.40%), while the lowest carbon levels were found in rapeseed (45.79%) and wheat (46.04%). The levels of hydrogen were equal in all analyzed samples, ranging from 4.60 to 6.91%. The highest levels were found in wheat (6.91%) and oak (6.90%), and the lowest ones in maize (4.60%) and soy bean (4.83%).

Nitrogen was found in small quantities in the investigated biomass, from 0.15 to 1.01%. It does not develop heat in the fuel, reducing its heating value. Since

Table 2. Mean values of analyses of biomass samples C. H. N. O. S (in dry matter).

Sample group	Biomass sample	Analyses (%)				
		C	H	N	O	S
Forest crops	Oak	49.90^{c}	6.90^{a}	0.17^{ih}	43.01^{k}	0.02^{e}
	Beech	51.40^{b}	6.01^{e}	0.15^{i}	42.42^{l}	0.02^{e}
	Maple	48.70^{h}	6.31^{c}	0.24^{g}	44.72^{g}	0.03^{e}
	Yoke elm	47.72^{l}	6.27^{c}	0.21^{gh}	45.97^{d}	0.03^{e}
	Fir	52.81^{a}	6.11^{d}	0.20^{igh}	40.84^{m}	0.04^{e}
Arable crops	Maize	48.37^{j}	4.60^{j}	0.70^{bc}	46.26^{c}	0.07^{b}
	Wheat	46.04^{m}	6.91^{a}	0.51^{e}	46.31^{c}	0.23^{a}
	Soy bean	48.56^{i}	4.83^{i}	1.01^{a}	45.53^{e}	0.07^{d}
	Rapeseed	45.79^{h}	6.60^{b}	0.32^{f}	47.18^{a}	0.11^{b}
	Sunflower	49.8^{d}	5.81^{f}	0.46^{e}	43.83^{j}	0.10^{bc}
Woody horticultural crops	Apple	46.06^{m}	6.60^{b}	0.74^{b}	46.51^{b}	0.09^{cd}
	Sour cherry	48.92^{g}	5.85^{f}	0.67^{c}	44.46^{h}	0.10^{cd}
	Plum	49.05^{f}	5.31^{h}	0.70^{bc}	44.87^{f}	0.07^{d}
	Hazelnut	49.18^{e}	5.81^{f}	0.73^{b}	44.20^{i}	0.08^{cd}
	Grapevine	48.20^{k}	5.63^{g}	0.60^{d}	45.50^{e}	0.07^{d}
		$\overline{X}$ = 48.70***	$\overline{X}$ = 5.97***	$\overline{X}$ = 0.49***	$\overline{X}$ = 44.77***	$\overline{X}$ = 0.07 ***
		LSD = 0.08	LSD = 0.07	LSD = 0.05	LSD = 0.09	LSD = 0.02

Data are averages ± SD of three determinations. Different letters within a column indicate significant differences at the 5% level by Duncan test; n.s. = nonsignificant; *Significant with $P \leq 0.05$; **Significant with $P < 0.01$; ***Significant with $P < 0.001$.

nitrogen is a negative component in the fuel, the least efficient fuel, among the studied crops, would be that from soy bean (1.01%) and apple (0.74%); and the most efficient ones would be beech (0.15%) and oak (0.17%). Given the fact that oxygen binds a part of combustible matters, it is an undesirable component of the biomass.

In the investigated samples, it level was found to be between 40.84 and 47.13%. The highest oxygen levels were present in rapeseed (47.13%) and in apple (46.42%), and the lowest ones in fir (40.84%) and beech (42.42%).

The level of sulphur is very low in all investigated groups of samples, ranging from 0.02 to 0.11%. Due to very low quantities of sulphur, the studied biomass can be considered as environmentally sound fuel which reduces GHG emissions. Table 3 shows the mean values of the analyzed pellets produced from different types of the investigated biomass. Given the fact that agricultural biomass is not subject to a defined standard, the examined samples were compared to the DINplus commodity standard for woody pellets.

DISCUSSION

Gaur and Reed (1998), Jenkins et al. (1998), Parikha et al. (2005), Telmo et al. (2010), Kricka et al. (2010), Bilandzija et al. (2012) analyzed in their investigations the combustible and non-combustible elements of forest biomass, arable biomass and woody horticultural biomass.

It was established that forest biomass contained 0.40 to 4.32% of ash, 46.2 to 52.3% of carbon, 4.9 to 6.2% of hydrogen, 38.12 to 47.7% of oxygen, 0.00 to 0.57% of nitrogen, 0.0 to 0.07% of sulphur, 74.7 to 87.1 of volatile matter, 12.4 to 22.5% of fixed carbon, 17.60 to 20.70 MJ/kg of lower heat value. Arable biomass contains 0.56 to 8.90% of ash, 43.20 to 53.00% of carbon, 5.00 to 5.90% of hydrogen, 39.14 to 45.46% of oxygen, 0.21 to 0.87% of nitrogen, 0.01 to 0.16% of sulphur, 65.47 to 87.1 of volatile matter, 11.95 to 22.5% of fixed carbon and 14.52 to 18.90 MJ/kg of lower heat value. The content of woody horticultural biomass was: 5.24 to 10.19% moisture, 1.43 to 10.19% ash, 44.73 to 49.72% carbon, 6.06 to 6.85% hydrogen, 38.58 to 42.81% oxygen, 0.56 to 1.35% nitrogen, 0.18 to 0.22% sulphur, 77.84 to 82.13% volatile matters, 17.63 to 22.89% fixed carbon and 17.95 to 20.01 MJ/kg lower heat value.

Kricka et al. (2010), in their investigations, analyzed the amounts of water in different samples of forest and agricultural biomass. Comparing the results of their analyses with the results from this investigation, it can be concluded that the values of the investigated samples were in accordance with the relevant literature and that all investigated groups of samples had equal moisture content.

Gaur and Reed (1998) investigated the fuel quality of biomass which was obtained from various raw materials of the forest and agricultural biomass, and comparing their results of ash analysis, it can be concluded that the forest biomass and woody horticultural biomass, given

Table 3. Mean values of properties of pellets of different groups of the investigated biomass (forest. arable and woody horticultural crops residues).

Sample group	Abrasion (%)	Diameter (mm)	Length (mm)	Moisture (%)	Ash (%)	Density (Kg dm^{-3})	N (%)	S (%)	Cl (%)	Lower heating value (MJ kg^{-1})
Forest crops	2.02	6	13	6.5	0.49	1.18	0.20	0.02	0.02	18.87
Arable crops	2.30	6	13	6.4	5.62	1.12	0.59	0.11	0.54	16.40
Woody horticultural crops	2.22	6	13	7.2	2.79	1.13	0.62	0.08	0.05	17.04

their lower ash content, have better heating properties. These properties are significantly poorer in the arable biomass, which was corroborated by analyzing the samples in this investigation.

The comparison of these results with those obtained by Gaur and Reed (1998), Telmo et al. (2010) makes it evident that, when fixed carbon is concerned, all groups of samples give the fuels of approximately equal quality. However, in terms of quality, the best one would be the fuel obtained from the woody horticultural biomass.

According to the data from Gaur and Reed (1998) and Telmo et al. (2010), it can be concluded that volatile matter in the investigated biomass samples was within the acceptable limits. Also, equal levels of volatile matter were found in all groups of the investigated samples. When comparing the results regarding carbon content obtained by Gaur and Reed (1998) and Bilandzija et al. (2012) with these analyses, we found equal levels of carbon in forest, arable and woody horticultural crops. However, there were no significant differences in hydrogen levels between all investigated samples of pruned biomass, which is in accordance with the literature.

The analyzed data also confirm the literature data, so the highest nitrogen level was found in the forest and arable biomass, while the most efficient fuel, given the lowest level of nitrogen, would be the fuel from forest biomass. The comparison with the data from Gaur and Reed (1998) makes it evident that all analyzed samples were confirmed and, given the percentage content of oxygen, the best performing biomass as fuel was obtained from the group of forest biomass samples.

The results for lower heating value were in accordance with relevant literature which is confirmed by the analyses conducted by Di Blasi et al. (1996) and Cao et al. (2006). Comparing the examined sample groups, it can be determined that given the lower heating value, forest biomass gives significantly more efficient fuel than the woody horticultural and arable crops. Given the prescribed values for diameter (4 to 10 mm), length (12 to 15 mm), water content (< 10%) and density (> 1.12 g/dm^3), it can be concluded that all examined groups of samples fully meet the DINplus standard for these parameters. The approved level of ash (< 0.5%) in pellets was met only by the forest biomass samples, while all other groups of samples did not satisfy the set DIN+ standard. Abrasion resistance requirement (< 2.3%) was met by pellets from all analyzed biomass types. The forest biomass was the only one that meets the maximum approved content of nitrogen (0.30%) and chlorine (0.02%), while the woody horticultural crops (N 0.44%; Cl 0.20%) and arable crops (N 0.55%; Cl 0.54%) diverged from the approved values. The sulphur content below the approved level of 0.04% was found only in the forest biomass samples, while other types of biomass slightly exceeded the allowed sulphur level woody horticultural crops (0.08%) and arable crops (0.09%). The prescribed heating values (> 18.01 MJ/kg) were obtained in the forest biomass, while it was somewhat lower in the woody horticultural (1.91 MJ/kg) and arable crops (17.65 MJ/kg).

Conclusion

Based on our investigations and the comparison of the biomass from various crops, forest crops (oak, beech, maple, walnut, and pine), arable crops (maize, wheat, soy bean, rapeseed, sunflower) and woody horticultural crops (apple, sour cherry, plum, hazelnut, and grapevine), the following conclusions was drawn:

1. Forest and agricultural biomass are an efficient renewable source of energy with good physical and chemical properties. In order to put such biomass in utilisation, it would be necessary to define new standards, because the current commodity standards can be applied only to woody biomass.
2. When considering the individual combustible and non-combustible matters (moisture, ash, C_{fix}, volatile matter, lower heating value, C, H, N, O, S) present in the investigated biomass samples of all groups, it can be concluded that the highest quality biomass comes from the forest material.
3. The pelleting technology gives a new kind of

fuel with high energy value, which is efficient and easy to handle.

The investigations of pellets show that all groups meet the DinPLUS commodity standards for abrasion, diameter, length and water content, while the pellets produced from the forest biomass meet the complete requirements of this standard. Due to acceptable level of sulphur in the investigated samples, the tested pruned biomass can be characterised as an environmentally suitable biofuel, while chlorine and nitrogen contents should be monitored.

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