Improvement of energy properties of lignocellulosic waste by thermochemical conversion into biochar

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Abstract

Peach stones, a valuable agro-industrial by-product available in many countries worldwide, comprise a renewable resource, which can be widely applied for multifunctional purposes. Its important advantages such as high-energy value, low ash content, low price and wide abundance, make peach stones an ideal fuel for energy production, but also for new materials synthesis. Although peach stones exhibit adequate combustion properties, allowing their direct use with minimal physical/chemical treatment, they often need further modification in order to improve their thermal properties, where slow pyrolysis is frequently used. This study aims to provide a practical and effective solution to the revalorization of waste biomass originating from the fruit processing industry, through slow pyrolysis in order to convert this waste into carbonaceous material – biochar. The thermo-chemical conversion of raw biomass resulted in a stable material with excellent fuel properties, with higher mass energy density and grinding ability, providing biochar with properties, in energy sense, similar or even better than a coal. Biochar has a higher fixed carbon content and a higher energy potential than biomass itself, and its application as a biofuel might reduce emissions of greenhouse gases, as it reduces the amount of waste landed and increases the share of energy generated from renewable sources.

Keywords: waste biomass, peach stones, renewable sources, pyrolysis, fuel properties.

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1. INTRODUCTION

Continuous growth of energy inputs is the result of intensive technological development and ongoing society industrialization worldwide. Exhaustion of fossil fuels, increasing energy costs (especially nowadays) and environmental pollution associated with fossil fuel application have resulted in intensive efforts to find alternative energy sources. In 2014, the European Commission proposed the EU Energy Security Strategy that has identified several renewable alternatives that can deliver clean and renewable energy (RE) to replace fossil fuels, so to possibly decreasing the EU dependence on fossil fuels, diversify energy supplies and reduce greenhouse gas emissions [1]. It was expected that energy produced by biomass would contribute to more than half of the EU's RE objective (which was set to be 32 % share of RE in gross final energy consumption), identifying biomass as an important RE source to reach the EU objectives [2]. Analysis of the Serbian energy sector reveals that it is primarily based on coal combustion. According to the data contained in the Strategy for the development of the energy sector up to 2025 of the Republic of Serbia, the greatest potential of RE lies in biomass reaching 3.4 tonne of oil equivalent (142.36 GJ), which represents more than half of renewables potential. Although significant, this potential is still underutilized. One of the important biomass sources in Serbia might be food processing industry wastes, such as different fruit stones, nutshells, residues from orchards/vineyards, etc. Most of this biomass has great importance as being a source of energy for domestic purposes

by direct combustion, which is neither economical nor environmental, so these biomass sources have to be converted into forms that are more useful by using different conversion technologies [3]. Having in mind that the production of biomass waste is constantly increasing, the application of suitable biowaste materials for heat/electricity and functional materials production might significantly reduce the amount of landfill waste, slowing the depletion of fossil resources and minimizing negative impacts on the climate change.

Biomass, as one of the main renewable resources, can play a considerable role in a more diverse and sustainable energy mix. It includes all biodegradable organic materials produced as industrial and municipal waste. Although there are multiple methods for utilisation of energy stored in biomass, the most common forms include direct combustion and co-firing, which are often connected with certain risks such as fire-explosions, excessive slagging, and ash formation, as well as chlorine corrosion. Among all the techniques for biomass conversion, the pyrolysis process offers a number of benefits, including lower emissions of greenhouse gases (GHG) and reusing of all by-products. Another benefit that can be obtained by thermochemical conversion of raw biomass lies in better and easier grinding of biochars compared to raw biomass.

Pyrolysis is a thermal decomposition process, which takes place in the absence of oxygen representing one of the fundamental thermochemical conversion processes that can be used to transform biomass into the energy rich fuel materials. The pyrolysis process is mainly characterised by solid fuel thermal degradation, involving the rupture of carbon–carbon bonds and formation of carbon–oxygen bonds, most often in the temperature range from 400 to 550 °C. The yields and product quality depend on operating conditions such as heating rate, residence time and pyrolysis temperature. Biochar, solid carbon-rich material, obtained by slow pyrolysis, has a completely different set of properties in comparison to the respective feedstock, and can bring sustainable changes in securing future supply of green energy and as well turn the bioenergy into carbon-negative industry [4]. Biochar can be used as a product itself or as a component of a blended product, with a wide range of applications such as a soil improvement agent, improved energy source, remediation agent in particular environmental pollution problems, and certainly as an avenue for GHG mitigation.

In this paper, raw waste biomass generated in the Serbian food-processing sector and derived carbonaceous material, biochar, was investigated regarding some of the main physico-chemical properties with the focus on potentials of these materials for use as solid fuels.

2. MATERIALS AND METHODS

Waste biomass of peach stones (*Prunus persica* L.) was obtained from the Juice Factory Vino Župa Aleksandrovac, Serbia. After washing and drying, stones were grinded by using a vibrating disk mill (Siebtechnik GmbH, Germany), and sieved into different particle sizes, where the class between 0.1 to 0.5 mm was used for further investigations. Part of the grounded peach samples (PS) were pyrolysed at 500 °C under oxygen-limited conditions in a Nabertherm 1300 muffle furnace (Nabertherm, Germany) in order to obtain biochar (PS-B). For that purpose, argon (Ar) gas with gas flow of 100 cm^3 min^{-1} was used, where the heating rate was 10 °C min^{-1} and pyrolysis time was 1 h.

Bulk density of both samples was determined by using a filling and tapping procedure [5]. The average bulk density was determined in triplicates.

Analyses of moisture, volatile matter (VM) and ash were performed according to the ASTM D1762-84 (2007) standard. Fixed carbon (FC) was calculated by subtracting the ash, moisture and VM contents from 100 wt.%. Elemental analysis (C, H, N, and S) of PS and PS-B was conducted by using a Vario EL III CHNS Elementar Analyzer (Hanau, Germany). The content of oxygen was calculated by subtracting the C, H, N and S and ash contents from 100 wt.%. Inorganic analysis (K, Mg, Ca, Na, Fe, Si, Pb, Cu, and Ni) in ash samples (dissolved by the nitric-perchloric acid digestion method) was carried out directly from the solution by using atomic absorption spectroscopy, AAS (Perkin Elmer, AAS Analyst 300).

The mass yield of PS-B (MY, wt.%) was calculated according to equation (1) [6]:

$$MY = \frac{m_{\text{pyrolised sample}}}{m_{\text{raw sample}}} 100 \qquad\qquad (1)$$

where $m_{\text{pyrolised sample}}$ represents mass of biochar and $m_{\text{raw sample}}$ is the mass of raw peach stone sample.

Higher heating value (HHV) was calculated according to the equation (2) given by Channiwala and Parikh [7], while the energy yields (EY) and energy densifications (ED) were calculated by equation (3) proposed by Nachenius RW *et al.* [8] and equation (4), proposed by Kim D *et al.* [9].

$$HHV = 0.3491C + 1.1783H + 0.1005S - 0.1034O - 0.015N - 0.0221Ash \tag{2}$$

$$EY = MY \frac{HHV_{biochar}}{HHV_{raw\ feedstock}} \tag{3}$$

$$ED = \frac{HHV_{biochar}}{HHV_{raw\ feedstock}} \tag{4}$$

Fuel ratio (FR) was calculated as the weight ratio between contant of fixed carbon (FC) and content of volatile matter (VM), as presented ba equation (6) [6]:

$$FR = \frac{FC}{VM} \tag{5}$$

In the present study, the sample weight loss (thermogravimetric) and the rate of weight loss (derivative thermogravimetric) of both samples were performed by Simultaneous Thermal Analyzer, STA 449 F5 Jupiter (Netzsch, Germany), which operated at air flow of 100 cm^3 min^{-1}, while heating from 25 to 900 °C at a rate of 10 °C min^{-1} at atmospheric pressure.

3. RESULTS AND DISCUSSION

Main physic-chemical properties of raw peach stones and its carbonaceous solid obtained by slow pyrolysis are presented in Table 1. The elemental analysis of the investigated materials has shown very low contents of sulphur (less than 0.2 %) and nitrogen (less than 0.3 %), which might contribute to reduction of emissions of gaseous NO_x and SO_2 pollutants if these samples are used for energy purposes.

Table 1. Chemical and fuel properties of raw peach stone, biochar and lignite samples

Parameter		PS	PS-B	Lignite [12]
Bulk density (kg m^{-3})		675	540	-
Content, wt.% (air dried)	Moisture	7.70	2.18	6.31
	VM	82.42	25.20	34.23
	Ash	0.34	0.73	44.53
	FC	9.54	71.89	14.93
Fuel properties	O/C weight ratio	0.97	0.40	-
	H/C weight ratio	0.13	0.04	-
	HHV, MJ kg^{-1}	18.89	24.58	13.12
	EY, %	-	36.62	-
	ED	1	1.3	-
	FR , %	0.12	2.85	0.44

Mass yield of peach stone biochar was found to be 28.1 %. This value is close to those presented by Weber *et al.* [11] who obtained 29.5 and 27.6 % for the mass yield of forest residue and walnut shells, respectively. The results of elemental composition analysis show a decrease in oxygen and hydrogen contents and an increase in the carbon content in pyrolysed sample as compared to the raw material. PS is composed of 47 % of carbon, 6 % of hydrogen and 46% of oxygen, while biochar contains 69 % of carbon, 3 % of hydrogen and 27 % of oxygen [12]. During the pyrolysis process, a series of chemical reactions occurs, resulting in the increase in the condensation and aromatization degrees of the material, which translates into the change in molar ratios O/C (carbonization degree) and H/C (aromatization degree) [13]. From the combustion point of view, fuels with lower O/C and H/C weight percent ratios are favourable because their application reduces energy loss, and production of smoke and water vapour [14]. As it can be seen, the O/C and H/C weight percent ratios for the PS fuel are 0.97 and 0.13. When PS was subjected to the pyrolysis process at 500 °C these ratios are reduced to 0.4 and 0.04, respectively (Table 1). These results suggested that pyrolysis changed the elemental proportions of the biomass bringing them closer to those of coal [1].

As can be also seen from Table 1, the volatile matter content of the PS material is high (about 82 wt.%), while its fixed carbon content is low (about 9.5 wt.%). For the carbonaceous materials, PS-B, the content of volatile matter is reduced, and the fixed carbon content is increased (25 and 72 wt.%, respectively). This increase in the fixed carbon content of biochar is due to the relative increase of this fraction (which is thermally stable and constant on the absolute basis) as the fraction of thermally unstable volatile matter is lost.

The ash content also increased from 0.13 wt.% for PS to 0.73 wt.% for PS-B. The mineral components in biomass do not contain aluminosilicates, but consist of simple inorganic salts of potassium, calcium, magnesium, and sodium in the form of phosphates, sulphates, and chlorides. Analysis of major components of the mineral matter in ashes of PS and PS-B revealed K, Ca and Mg as major components. The content of K was 25.4 and 19.9 wt.%, for PS and PS-B, respectively. Magnesium content ranged between 7 and 9 wt.%, while the Ca content was close to 20 wt.% for both samples. The amount of Fe was approximately 2 wt.%, while the amount of other components analysed was close to 0.1 wt.% in both samples. A characteristic feature of the biomass ash is the high content of phosphorus and potassium compounds, where the portion of phosphorous in the PS ash sample was found to be close to 27 wt.%. Considerable contents of calcium, magnesium, phosphorus, and potassium were noted in the ashes produced by combustion of both samples. Ca and Mg compounds usually increase the ash melting temperature, while the effect of K and Na is opposite [15]. These processes are highly important, given the risk of fouling and ash slagging on the walls of furnaces or heat-exchangers surfaces. Having in mind low quantities of ashes after combustion, it is assumed that this ash content cannot have significant effects on combustion behaviour of both samples.

Bulk density represents one of the factors that have to be considered for estimation of the space requirements for transport and storage. The bulk density value of PS is higher than the value for PS-B (Table1). In general, pyrolysis decreases the bulk density of biochars, which should be further subjected to a densification process in order to improve its mechanical stability, and transport and storage-related properties. This can be achieved by pelletisation, resulting in a secondary fuel with higher energy parameters suitable for use in combustion/co-firing processes [15].

Energy yield calculation is based on the mass yield and is always lower than 100 %. It is observed that the energy yield was around 36% (Table 1), which is a consequence of lower mass yield at 500 °C. The fuel ratio (FC/VM) increased from 0.12 for PS to 2.85 for PS-B (Table 1), due to the FC increase and VM decrease during thermochemical conversion and loss of volatiles in the gaseous form. Pyrolysis reactions lead to a significant increase in the mass energy density of biochars, which can be explained by the higher energy contained in carbon−carbon bonds than in carbon−oxygen and carbon−hydrogen bonds [14].

Pyrolysis considerably modified the structure of the peach stone sample, resulting in elevated carbon and FC contents, which strongly influence the high heat value. Specifically, HHV increased from 18.89 MJ kg^{-1} for the PS to 25.84 MJ kg^{-1} for the carbonaceous solid PS-B (Table 1), which are higher than the values of the most lignites. Compared with HHVs of lignites widely used in Serbia's thermal plants (which is 13.12 MJ kg^{-1} for coal from Kolubara or 14.00 MJ kg^{-1} for coal from Kostolac) [10], it can be seen that HHV of both raw biomass and biochar are significantly higher. Gross calorific value of peach stone biochar is higher by 30 % than the calorific value of the raw feedstock, which suggests that biochar is a better solid fuel than biomass. In addition, high content of volatiles in raw biomass, might induce higher emissions of CO, CO_2, NO_x, SO_2 *etc.* [16], so the direct combustion of this kind of fuel should be avoided.

TGA represents an effective way to analyse the thermal and combustion behaviour of different kinds of materials such as coal, biomass, and their blends [17]. The thermal profiles of PS and PS-B degradation obtained by thermos-gravimetric analyses are shown in Figure 1.

The curves of the raw material (Fig 1a) revealed several stages of mass loss, presenting a heterogeneous decomposition, as it has been previously described [18]. The TG and DTG curves of thermal degradation of peach stones were associated to decomposition of its main components: hemicellulose, cellulose, and lignin. The greatest mass loss associated with volatile compounds was relatively fast (220 to 380 °C) due to the thermal decomposition of hemicellulose/cellulose components, and reached 58 % of the total mass loss, indicating that a great part of the VM contained in PS has been decomposed. The curves of the raw material (Fig 1a) revealed several stages of mass loss, presenting a heterogeneous decomposition, as it has been previously described [18].
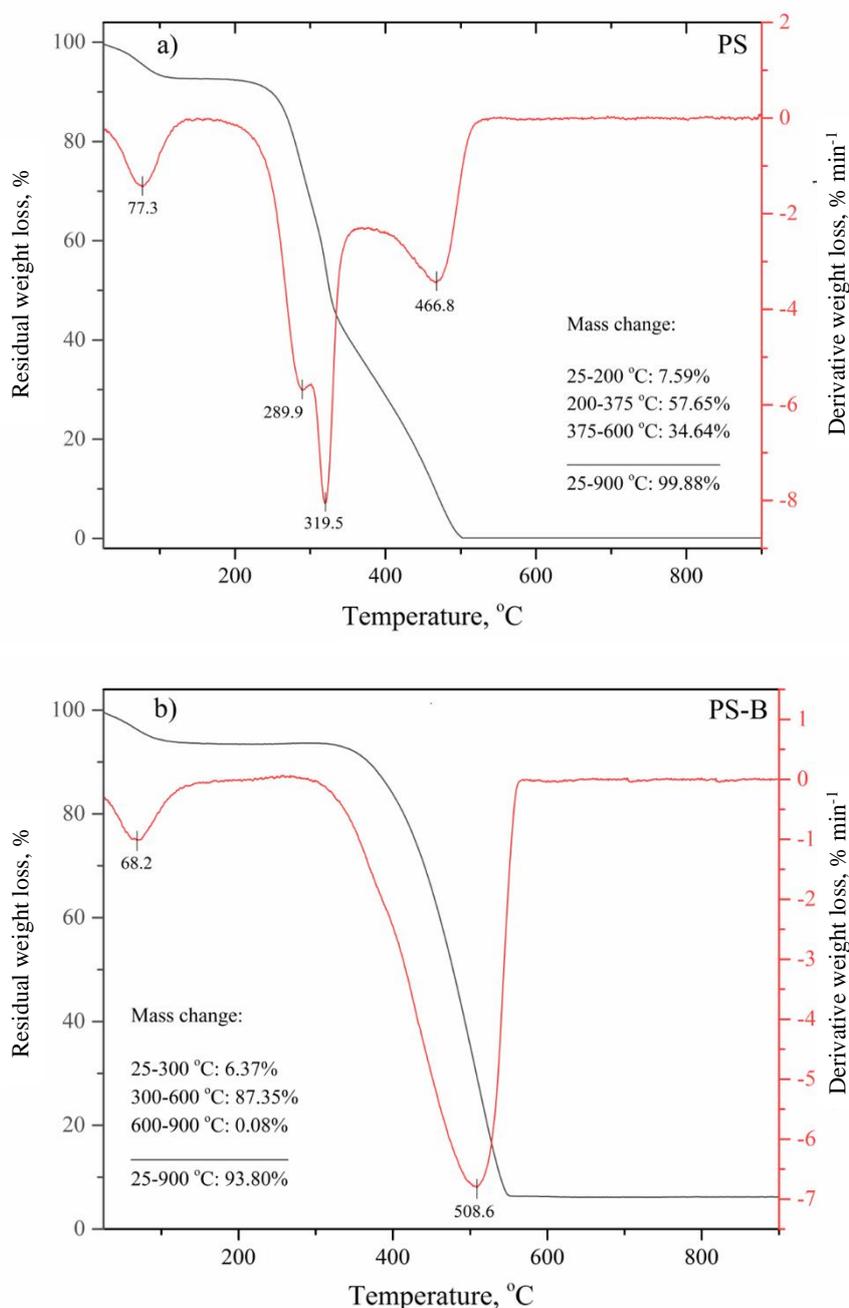
Figure 1. Thermal degradation of (a) PS and (b) PS-B obtained by slow pyrolysis at 500 °C

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form mainly volatile products whilst lignin mainly forms char due to its stability to thermal degradation. In pyrolysis, hemicellulose decomposes at temperatures of 200 to 260 °C to form more volatiles and less tar and char than cellulose, which degrades at 240 to 350 °C to produce anhydrocellulose and levoglucosan as initial products, which further decompose to volatiles, tars, and char [19]. Lignin decomposes in the temperature range between 280 and 500 °C producing phenols and carbon–carbon bonds, resulting in higher formation of char than cellulose. According to the literature [20], the char containing ash fraction of the biomass obtained after pyrolysis, also might have application as a fuel and furthermore, it may have potential for the improvement of soil quality and might allow carbon sequestration for a long time.

4. CONCLUSION

In this paper, slow pyrolysis of lignocellulosic peach stone waste was studied as a method for preparing solid biofuel-biochar. Main changes on physical/chemical properties of biochar obtained by the thermal conversion were analysed and compared with those of the starting material. Direct application of the waste biomass as a fuel is hindered by the low bulk and energy densities, poor grinding ability and lower fuel ratio. Conversion of the raw biomass into biochar by pyrolysis can result in stable materials with excellent fuel properties, higher mass energy density and better grinding ability, providing properties in energy sense similar or even better than those of coal. The PS and PS-B materials were characterised by higher proportions of potassium compounds and lower proportions of calcium compounds, which might promote formation of low-melting eutectics. Still, the low ash content in biomass reduces the risk of slagging and fouling of heating surfaces. Biochar has higher fixed carbon and higher energy potential than biomass itself, and its application as biofuel might reduce the emissions of greenhouse gases, as it reduces the amount of waste landed and increases the share of energy generated from renewable sources.

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Poboljšanje energetskih svojstava lignoceluloznog otpada termohemijskom konverzijom u biočađ

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(Stručni rad)

Izvod

Koštice breskve, otpadna biomasa poreklom iz industrije prerade voća, dostupna u mnogim zemljama širom sveta, predstavlja obnovljivi resurs koji može imati različitu primenu. Prednosti koje se ogledaju u velikom energetskom potencijalu, malom sadržaju pepela, niskoj ceni i širokoj rasprostranjenosti, čine koštice breskve idealnim resursom za proizvodnju energije, ali i za sintezu novih materijala. Iako koštice breskve imaju svojstva koja im omogućavaju da se koriste uz minimalni fizičko-hemijski tretman, često im je potrebna dodatna modifikacija kako bi se poboljšala njihova svojstva. Jedan on načina poboljšanja energetskih karakteristika otpadne biomase predstavlja termohemijska konverzija materijala u vidu spore pirolize. Ispitivanja prikazana u ovom radu obavljena su sa ciljem obezbeđivanja praktičnog i efikasnog rešenje za revalorizaciju otpadne biomase koja potiče iz industrije prerade voća, putem spore pirolize kojom se vrši konverzija ovog otpada u ugljenični materijal – biočađ. Prikazani rezultati ukazuju na glavna svojstva dobijene biočađi u funkciji njene primene kao čvrstog goriva, istovremeno ih upoređujući sa svojstvima sirove biomase. Termohemijska konverzija polazne lignocelulozne biomase daje stabilni ugljenični material odličnih gorivih karakteristika, veće energetske gustine i boljih mehaničkih svojstava, što rezultuje energetskim svojstvima koja su bolja od uglja. Biočađ poseduje znatno veći energetski potencijal od same biomase, i njegova primena kao biogoriva može redukovati emisiju gasova sa efektom staklene bašte, pri čemu se istovremeno smanjuje količina deponovanog otpada ali i povećava udeo energije generisane iz obnovljivih izvora.

Ključne reči: Otpadna biomasa, koštice breskve, obnovljivi izvori, piroliza, osobine goriva

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