



USE OF INVASIVE PLANT SPECIES TREE OF HEAVEN (*AILANTHUS ALTISSIMA* MILL.) BIOMASS IN ENERGY PRODUCTION

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ABSTRACT

*Spreading of invasive plant species threatens habitats, ecosystems and indigenous species in areas where they would not come naturally. One of them is a plant species called Tree of heaven (*Ailanthus altissima*, Mill), which is one of the most dangerous invasive woody plants in the world as well as in Croatia. Tree of heaven occupies space to the detriment of other plant species; it produces allelochemicals and shows the tendency of intense spreading and suppression of other domestic species. It reverses the landscape's appearance and does invaluable damage to the ecosystem, by generating enormous quantities of potentially usable biomass. Biomass has a significant potential for further development. The use of biomass which remains after the removal of invasive species has many advantages, both regarding of increasing share of renewable energy production and environmental protection, as well as in reducing greenhouse gas emissions, which adversely affect human health and the environment.*

The research aimed to determine the possibility of using invasive plant species Tree of heaven remainings after its removal from nature as a raw material and determine its energy potential and the possibility of using it for energy purposes, by direct combustion and pyrolysis.

According to the analysis results it can be concluded that the Tree of heaven has a possibility of its utilization for energy purposes. It is justified to assume that this invasive species has potential as raw material for direct combustion, as well as in the process of pyrolysis, i.e., production of bio-oil and biochar.

Keywords: biomass, energy, invasive species

INTRODUCTION

The problem of alien invasive species has become more interesting in recent decades. The control of the introduction and spreading of invasive species, as well as the reduction of their influence on native species and the overall ecosystems, is today one of the most significant challenges of nature protection in Europe. The alien invasive species under The Convention on Biological Diversity, and the Bern Convention as well as in the opinion of numerous authors and institutions, are recognized as the second major threat to biodiversity, right after the direct destruction of natural habitats. In areas where they would never come naturally, alien invasive species endanger habitats, ecosystems and autochthonous species, resulting in numerous negative environmental and economic consequences (Novak and Kravaršćan, 2014) by aggressive spreading. For the last decades, their spread has increased by trade, tourism, and travel (Novak and Kravaršćan, 2014).

The Tree of heaven is an exotic and invasive deciduous tree that can grow in different environmental conditions, regardless of soil condition and location. This deciduous, extremely opportunistic, aggressive and adaptable species, due to its rapid growth, propagation by seeds and by root sprouts, and allelopathic chemical compounds (which suppress autochthonous vegetation) (Kovačić et al., 2008), today is considered to be one of the most invasive tree species in the world and also in Croatia (Novak and Novak, 2017). Tree of heaven was first introduced to Europe in 1740. from central Asia (China). During the 60s of the last century, it was planted in Croatia, as a target species that stabilize the soil and prevent landslides or as an ornamental species (Novak and Novak, 2017). It is known for its use in honey production, herbal medicine, furniture manufacturing, while in China it is used as a construction and tool wood and for the production of cellulose and silk. Most often it could be found at neglected fields, along the road, wild garbage dumps, agricultural areas, and near buildings, houses and other construction objects (Vukojević et al., 2012). According to Novak and Novak (2017), the Tree of heaven is widespread in all counties of Croatia. Its control is challenging and demanding, mainly because of its high regeneration ability, and it is best to use a combination of mechanical and chemical measures of suppression (Novak and Kravaršćan, 2014). Uncontrolled production of large quantities of biomass has aroused testing of its potential for energy production. This study aimed to determine the possibility of using biomass of Tree of heaven, which has remained after its mechanical removal from nature and to identify the differences between locations.

There are numerous possibilities for energy utilization of biomass. The most commonly used is a direct combustion process where biomass, without previous conversion into other forms, serves as a fuel (Trkmić and Janješ, 2012) for obtaining electrical and thermal energy. Apart from combustion, for further energy production or biomass conversion into various types of solid, liquid or gaseous fuels and products, other biochemical and thermochemical processes are used. Pyrolysis is a process which involves heating of organic materials (biomass) to temperatures from 300 to 1.400 °C in the absence of oxygen. At such high temperatures, organic materials thermally decompose releasing a vapor phase and a residual solid phase (biochar). On cooling the pyrolysis vapor, polar and high molecular-weight compounds condense out as a liquid (bio-oil) while low-molecular-weight volatile compounds remain in the gas phase (synthetic gas) (Laird et al., 2009). By this processes, instead of just burning biomass, valued products can be obtained. Energy characteristics and the possibility of using the Tree of heaven biomass, through the direct combustion and

pyrolysis for energy purposes were examined, as well as the production of bio-oil as an energy source and biochar as a value-added product.

MATERIALS AND METHODS

The research was conducted at the Department of Agricultural Technology, Storage and Transport at University of Zagreb Faculty of Agriculture. Samples of the invasive plant species biomass were collected in five different locations in the area of Split-Dalmatia County (Klis, 43° 33'34.6 "N 16° 31'18.8" E, Lokvičići, 43° 27'56.3 "N 17° 05'25.4 "E, Gornji Muć, 43° 40'46.5" N 16° 29'38.9 "E, Gornji Muć, 43° 40'31.7" N 16° 29'41.9 "E, Žrnovnica, 43° 31'09.4" N 16 ° 32'54.9 "E) in April 2017. The analyzes were preceded by grounding of samples in the laboratory mill (IKA, Germany), followed by standard methods for determining the water content (HRN EN 18134-2:2015), ash (HRN EN ISO 18122:2015), coke (CEN / TS 15148: 2009), fixed carbon and volatile matter (CEN / TS 15148: 2009), and in the calorimeter IKA C200 (IKA, Germany) the higher heating values in the samples were determined (HRN EN 14918: 2010). Determination of cellulose, hemicellulose and lignin content of raw materials was conducted by the modified standard method ISO 5351-1: 2002. After the analysis of raw materials, i.e., the biomass of Tree of heaven, pyrolysis of samples was conducted in laboratory conditions at a temperature of about 400 °C. The proportion of obtained biochar and bio-oil was calculated, and the biochar was analyzed by the above-mentioned standard methods. Analysis of variance (ANOVA) was performed of results obtained by biomass and biochar analyzes, while the differences between the mean values were tested by t-test.

RESULTS AND DISCUSSION

Suitability of the raw material for the production of energy by direct combustion can be assessed by determination of its physicochemical, structural and energy properties. Therefore, Table 1 shows the mean value of the water content in the fresh sample, ash content, coke, volatile matter, fixed carbon as well as the higher and lower heating value of dry matter of the Tree of heaven biomass.

Table 1 The characteristics of the Tree of heaven biomass samples (dry matter)

	Location				
	L1	L2	L3	L4	L5
*Moisture (%)	25,88 ^{BC}	40,67 ^A	28,89 ^B	19,30 ^C	34,81 ^B
Ash (%)	5,43 ^B	6,60 ^A	5,37 ^B	5,83 ^B	5,65 ^B
Coke (%)	15,59 ^A	14,35 ^A	15,99 ^A	13,97 ^A	16,47 ^A
Volatile matter (%)	81,11 ^B	82,36 ^A	79,38 ^C	81,39 ^B	78,88 ^C
Fixed carbon (%)	6,63 ^C	11,60 ^B	14,14 ^A	12,12 ^B	14,62 ^A
**LHV (MJ kg ⁻¹)	14,68 ^C	15,71 ^A	15,01 ^B	14,48 ^{BC}	15,98 ^A
***HHV (MJ kg ⁻¹)	15,96 ^C	17,03 ^A	16,35 ^B	16,15 ^C	17,29 ^A

*Moisture content analysis is done on raw biomass; **LHV = lower heating value, ***HHV = higher heating value

The premise for the quality production of biochar is its higher contribution in pyrolysis process (Jurišić et al., 2017). Also, in this study, analyzes of biochar composition were made by standard methods for solid biofuels (Table 2). The water (moisture) in the fuel is found to be a non-combustible ingredient which has a negative impact on the biomass heating values. Part of the energy released during the combustion process is spent on evaporation of water (Francescato et al., 2008). Expected values for water content in biomass vary by about 10% for dried biomass, while for raw biomass they reach up to 50% (Yao et al., 2005). Water content is one of the most important parameters when it comes to fuels properties of biomass because it depends on it how will the biomass be transferred to energy (Ross et al., 2008). Biomass with the low and moderate water content of less than 50% is efficiently used in combustion processes, while alternatively it can be used for thermochemical conversion by gasification and pyrolysis (Permchart and Koupryanov, 2004). Freshly collected biomass typically has a water content of 40 to 65%, while residues of agricultural crops (maize, straw, etc.) that have been exposed to air-drying have about 15% or less (Ross et al., 2008). The water content in the analyzed samples ranged from the lowest value of 19.30% (location 4) up to 40.67% (location 2). A significant difference between the water content of the samples between different locations was determined, as shown in Table 1. The water content in the analyzed biomass samples from most locations is within the expected values. Deviations are only visible at location 2, with slightly elevated values compared to other results (wetland).

Ash is an incombustible mineral residue after the biomass was combusted. The variability of the ash content is affected by the different composition and availability of nutrients in the soil, climatic conditions, type of plant species, part of the plant itself, soil quality as well as fertilization (Vassilev et al., 2010). Vassilev et al. (2010) state that ash content, depending on the type and parts of biomass crops, usually ranges from 0.5% to 3% although they can range from 0.1% to 46%. Francescato et al. (2008) stated that the ash content in agricultural biomass ranges from 2 to 25%. It is desirable that ash content in the raw biomass is not high, given that the ash content and the content of combustible matter are inversely proportional, and by increasing of ash, the content of combustible matter reduces. The ash content in the analyzed samples is significantly different at some locations and differences were noted among the locations, as shown in Table 1. The minimum ash content was 5.37% at location 3, while the highest values were found at location 2 reaching the amount of 6.60%. The results can be compared with the study of grain straw (Grubor et al., 2015) where ash values are ranged from 2.54 to 9.04%. The average amount of ash in biomass of major energy crops in Croatia was 3.14% (Jurišić et al., 2017). Biochar is very heterogeneous and consists of stable and reactive components (Jurišić et al., 2016). Variations in the ash content in biochar depend on the same conditions as in case of biomass. The average value of ash content in biochar samples from all five locations is 22.14%, which is higher than the ash content of biomass. Table 2 shows statistically significant differences in the ash content of biochar between locations. Jurišić et al. (2017) indicate that the average content of ash in the biochar of important energy crops in Croatia was 5.60%, while in the biochar of grain straw; Grubor et al. (2015) indicate ash content values of 9.15 to 16.53%, which are closer to the results obtained in this research. Considering that larger content of ash, cause soot formation and corrosion in biomass combustion systems (Grubor et al., 2015); this biochar does not match the quality of other types of biochar when viewed from the aspect of ash content.

Higher content of coke is a desirable feature of biomass (Boboulos, 2010) and increases the quality of fuel. Coke remains because of the combustion process of flammable or volatile

substances at very high temperatures (Voća et al., 2018). The average coke content obtained in analyzed samples of biomass ranges from a minimum of 13.97% at location 4 to 16.47% at location 5, and there are no statistically significant differences in the content of coke between the results. Grubor et al. (2015) obtained similar results for coke content from 16.20 to 34.76% in grain straw biomass. Jurišić et al. (2017) obtained the average content of coke in biomass of major energy crops in Croatia around 14.9%. From the aspect of coke content concerning other types of biomass, it can be concluded that it is a suitable material for energy production. According to the results shown in Table 2, it can be concluded that there are no significant differences between the locations in the coke content of biochar. Jurišić et al. (2017) stated the average content of coke in the biochar of important energy crops in Croatia at about 60.10%, while Grubor et al. (2015) did obtain values from 49.69 to 62.89% in grain straw biomass. Considering that the higher value of coke is preferable in biochar, it can be concluded that the Tree of heaven biomass is an excellent raw material for the pyrolysis process.

Table 2 The characteristics of the Tree of heaven biochar samples (dry matter)

	Location				
	L1	L2	L3	L4	L5
Ash (%)	17,91 ^D	24,98 ^A	19,30 ^C	23,91 ^B	24,61 ^{AB}
Coke (%)	63,57 ^A	63,10 ^A	63,34 ^A	67,64 ^A	68,21 ^A
Volatile matter (%)	33,14 ^A	33,61 ^A	32,01 ^B	27,71 ^C	27,14 ^C
Fixed carbon (%)	54,60 ^D	60,34 ^C	61,49 ^B	65,79 ^A	66,37 ^A
**LHV (MJ kg ⁻¹)	23,51 ^B	22,12 ^D	22,58 ^C	23,86 ^A	23,82 ^A
***HHV. (MJ kg ⁻¹)	24,26 ^A	22,79 ^B	23,12 ^B	24,40 ^A	24,49 ^A

Fixed carbon content (C_{fix}) represents a solid residue after combustion, i.e., after releasing volatile substances without ash (Garcia et al., 2012). Increase of fixed carbon content the increases heating value, thus improving the quality of biomass as fuel. According to Jarihul et al. (2012), the higher content of fixed carbon in biomass contributes to obtaining a higher amount of biochar regarding bio-oil and synthetic gas. Results of the fixed carbon content obtained from biomass range from 6.63% (location 1) to 14.62% (location 5). There were statistically significant differences between samples from different locations as shown in Table 1. The values of fixed carbon in grain straw biomass are in the range from about 5% (oat straw) to 13% (other straw) (Grubor et al., 2015). Jurišić et al. (2017) indicate that average values of fixed carbon biomass of major energy crops in Croatia are around 11.40%. Tree of heaven biomass has acceptable fixed carbon values, and obtained results were expected since values of the fixed carbon in the biomass of different cultures are similar. According to the results shown in Table 2, it can be concluded that there are significant differences between the locations in the content of fixed carbon in the Tree of heaven biochar. The content of fixed carbon in biochar samples ranges from 54.60 to 66.37%. In the case of grain straw biochar, fixed carbon values range from 33.15 to 50.25% (Grubor et al., 2015). Jurišić et al. (2017) obtained most similar results in the biochar, which also indicate that Tree of heaven is a suitable material for pyrolysis processes, with the 54.50% of fixed carbon in major energy crops biochar in Croatia.

During the combustion process, biomass decomposes on volatile gases and the solid residue. The term volatiles refer to the components such as light hydrocarbons, carbon monoxide, carbon dioxide, hydrogen, moisture, and tars released when the fuel is heated at a high temperature. Biomass generally has a very high content of volatile matter, with values that typically range around 75%, but can even increase up to 90%, which of course depends on the type of sample (Khan et al., 2009). In case of high concentrations of volatile matter, biomass is extremely flammable at lower temperatures, unlike fossil fuels. This is an undesirable feature due to the sudden release of energy at lower temperatures, causing that such fuels have lower energy value (Garcia et al., 2012, Quaak et al. al., 1999). Table 1 shows the significant differences between locations of volatile matter analysis of the analyzed biomass. Tree of heaven biomass analyzes shows the results for volatile matter from a minimum value of 78.88% (location 5) to a maximum value of 82.36% (location 2), and while the biochar values were ranged from 27.14 to 33.06%. According to Grubor et al. (2015), volatile matter in grain straw biomass were ranged from 34.62 to 79.66%, while in the case of biochar from 37.11 to 50.31%. Jurišić et al. (2017) stated average values of volatile matter of major energy crops biomass in Croatia were around 77.58%. Comparison of results shown that biomass, as well as biochar, have similar, even lower percent of volatiles compared to other biomass or biochars, which is positive for Tree of heaven as a material with energy potential.

The most important feature of each fuel is the heating value, also called calorific value. The heating value is a parameter that represents the amount of energy that can be obtained by combusting a certain amount of biomass (Garcia et al., 2012). It is an indicator of the chemical-related energy in the fuel, which turns into heat through the combustion process (Krička et al., 2010). The heating value of the biomass can be defined by its higher heating value (HHV), which is the energy content on a dry basis. The lower heating value (LHV) is calculated by subtracting the energy needed to evaporate the moisture content of the fuel (Khan et al., 2009). By analyzing the Tree of heaven biomass and biochar, average results of the HHV and LHV were obtained. Lowest HHV was ranged from the minimum value of 15.96 MJ kg⁻¹ at location 1, to the maximum HHV at location 5 of 17.29 MJ kg⁻¹. The LHV was ranged from the minimum value of 14.68 MJ kg⁻¹ at location 1 to the 15.98 MJ kg⁻¹ at location 5. Table 1 shows the heating values of the analyzed biomass, while Table 2 shows data of the heating values for biochar, with statistically significant differences between the locations. In the grain straw biomass, Grubor et al. (2015) obtained an HHV from 16,41 to 18.24 MJ kg⁻¹, while Jurišić et al. (2017) recorded the average HHV of major energy crops biomass in Croatia of 17.69 MJ kg⁻¹. In case of biochar, obtained HHV of Tree of heaven were ranged from 22.79 to 24.49 MJ kg⁻¹ and LHV from 22.12 to 23.86 MJ kg⁻¹. Grubor et al. (2015) recorded the HHV of biochar from 24.72 to 25.70 MJ kg⁻¹, while Jurišić et al. (2017) recorded an average HHV of biochar 30.67 MJ kg⁻¹. By comparing those values, it is evident that higher heating values of biomasses as well as of the biochar are similar to other results.

Biomass is a complex heterogeneous mixture consisting of key structural components such as cellulose, hemicellulose, and lignin. Voća et al., (2018) stated that most of the agricultural lignocellulose biomass consists of lignin (10-25%), hemicellulose (20-30%), and cellulose (40-50%). Lignin provides structures strength, and biomass with higher lignin content is more suitable for producing electricity and/or heat by direct combustion (Grubor et al., 2015). According to the results shown in Figure 1, it can be concluded that the highest content of cellulose was found in samples of location 5, highest content of lignin was found

in samples of location 3, and highest content of hemicellulose was found in samples from location 1. According to Grubor et al. (2015), the content of cellulose in the biomass of grain straw was ranged from 30.64 to 45.66%, the lignin content was ranged from 22.28 to 29.27%, and the hemicellulose was ranged from 17.56 to 39.67%. By comparing the obtained values with the literature data and the expected values for biomass, it is apparent that the lignin content is higher than expected, and it can be concluded that the biomass is suitable for direct combustion.

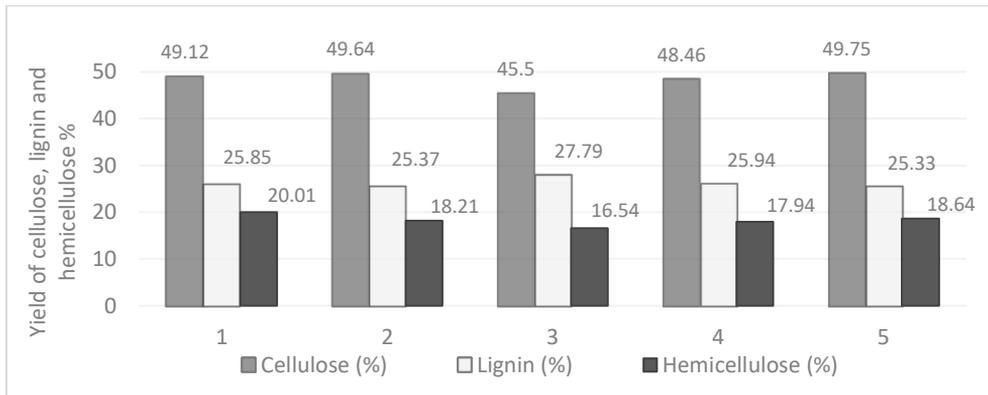


Figure 1 Cellulose, lignin and hemicellulose yield in Tree of heaven biomass

Table 3 Content of carbon, hydrogen, nitrogen, sulfur and oxygen in biomass and biochar (dry matter)

%	Biomass					Biochar				
	L1	L2	L3	L4	L5	L1	L2	L3	L4	L5
C	37,73 ^D	44,71 ^B	46,46 ^A	42,9 ^C	43,55 ^{BC}	66,02 ^A	61,19 ^C	66,85 ^A	64,26 ^B	66,31 ^A
H	5,85 ^A	6,03 ^A	6,14 ^A	5,99 ^A	6,01 ^A	3,45 ^A	3,07 ^A	2,49 ^B	2,48 ^B	3,06 ^A
N	2,97 ^A	2,91 ^A	2,56 ^A	1,63 ^B	2,38 ^A	3,99 ^A	2,98 ^{BC}	3,10 ^B	2,62 ^C	3,95 ^A
S	0,38 ^A	0,36 ^A	0,35 ^A	0,30 ^A	0,35 ^A	0,45 ^A	0,35 ^A	0,39 ^A	0,53 ^A	0,52 ^A
O	53,07 ^A	45,99 ^{CD}	44,5 ^D	49,18 ^B	47,72 ^{BC}	26,09 ^D	32,41 ^A	27,18 ^C	30,11 ^B	26,17 ^D

From the obtained values of the Tree of heaven biomass and biochar (Table 3), it can be noted that the highest, most desirable amount of carbon and hydrogen, as well as the lowest amount of sulfur and oxygen content, have locations 2 and 4, although the differences between locations are minimal. The analyzed biomass contains oxygen ranging from 47.20 to 47.71%, carbon in the range of 42.48 to 43.39%, hydrogen ranging from 6.14 to 6.29%, nitrogen ranging from 2.76 - 3.40% and sulfur 0.35-0.40%.

In Figure 2, the mean value of the obtained bio-oil and biochar after the pyrolysis is shown graphically. The pyrolysis process of organic matter is very complex, and this process generates three important products – biochar and volatile and non-volatile gases (bio-oil) (Jurišić et al., 2017). Depending on the conditions of the process (such as the particle size in

the biomass material and the temperature of the process itself), carbon-rich biochar was obtained (yield: 10-35%), volatile gases (yield: 30-70%) and non-volatile gases manifested as bio-oil: 15-35%). Furthermore, the content of biochar after pyrolysis is proportional to the lignin and hemicellulose content in biomass (Jahirul et al., 2012). The highest amount of biochar (39.52%) was obtained by analyzing the samples of location 2 and the lowest amount of biomass (28.30%) by analyzing samples from site 4. It is also evident that the most significant amount of bio-oil (59.60%) was obtained by analyzing samples of location 4, and the smallest amount of bio-oil (40.39%) by analyzing samples of location 2. The biochar yields in grain straw range from about 55 to 75% (Grubor et al., 2015). Jurišić et al. (2017) indicate slightly lower amounts of biochar (23.93%) and bio-oil (33.13%) in major energy crops in Croatia. There is an excellent potential for biochar and bio-oil production compared to the comparative literature approximate values.

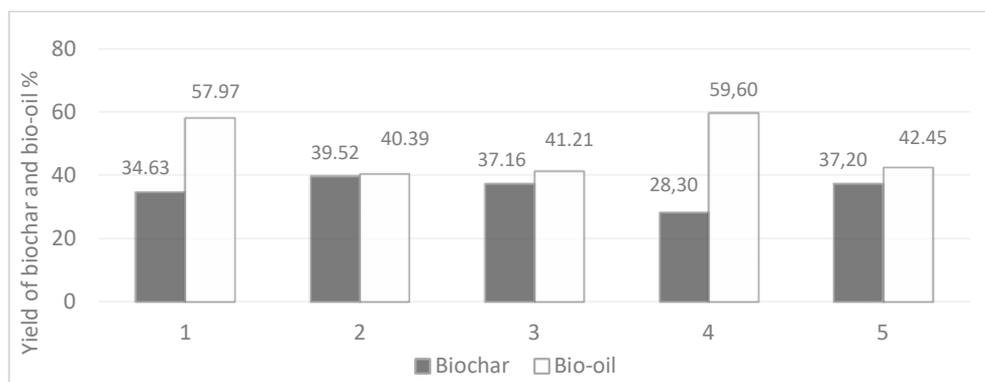


Figure 2 Pyrolysis products in Tree of heaven biomass samples yield

CONCLUSIONS

Based on this research of the invasive species Tree of heaven (*Ailanthus altissima*) biomass, its potential as a raw material for direct combustion, as well as in the process of pyrolysis, i.e., the production of bio-oil as an energy source and biochar as a value-added product was confirmed.

There is a significant difference in Tree of heaven biomass between sites in the content of water, ash, fixed carbon, volatile substances, and heating values. There is a significant difference in the biochar of the Tree of heaven between the sites for ash and heating values.

Tree of heaven biomass contains water ranging from 19.3 to 40.67%, indicating that the results fit into the literature values. Water content from site 2 makes an exception with its elevated value (which is acceptable, given that unlike others, location 2 it is a wetland area).

Analysis of the ash content in biomass (5.37 - 6.60%) and biochar (17.91 - 24.98%) showed higher ash content compared to the literature, which is not a desirable feature of biomass. Expected results of volatile substances stated in the literature, were lower than in biomass what indicates that Tree of heaven is not desirable raw material for direct combustion, but low values of volatile substances in biochar shows that biochar is a desirable raw material

for pyrolysis processes. From the perspective of coke, fixed carbon and heating values in biomass and biochar, biomass represent a potential raw material for energy production.

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KORIŠTENJE BIOMASE INVAZIVNE BILJNE VRSTE PAJASEN U PROIZVODNJI ENERGIJE

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SUMMARY

*Strane invazivne vrste agresivnim širenjem ugrožavaju staništa, ekosustave i autohtone vrste na područjima gdje prirodnim putem ne bi dospjele. Biljna vrsta pajasen (*Ailanthus altissima*), jedna je od najopasnijih invazivnih drvenastih biljaka kako u svijetu, tako i u Hrvatskoj. Preuzima prostor na štetu ostalog bilja, luči alelokemikalije te pokazuje tendenciju intenzivnog širenja i potiskivanja ostalih autohtonih vrsta. Nepovratno mijenja izgled krajolika te nanosi neprocjenjivu štetu na ekosustav stvarajući ogromne količine potencijalno iskoristive biomase. Biomasa ima značajan potencijal za daljnji razvoj. Primjena biomase, preostale nakon uklanjanja invazivnih vrsta ima nebrojne prednosti, kako u povećanju udjela proizvodnje obnovljive energije, tako i u pogledu smanjenja emisije stakleničkih plinova te štetnog utjecaja na zdravlje ljudi i okoliš. Cilj ovog rada bio je istražiti mogućnosti korištenja ostataka invazivne vrste pajasen, nakon njegovog uklanjanja iz prirode te procesom izgaranja i pirolizom biomase ispitati njegov energetske potencijal i mogućnost korištenja u energetske svrhe. Sukladno dobivenim rezultatima analiza, može se zaključiti da pajasen nudi mogućnost iskorištenja u energetske svrhe. Opravdana je pretpostavka da ova invazivna vrsta, ima potencijal kao sirovina za izravno izgaranje, kao i u procesu pirolize, odnosno proizvodnje biouglja i biougljena.*

Ključne riječi: biomasa, energija, invazivne vrste, pajasen