

PRELIMINARY RESEARCH OF WASTE BIOMASS AND PLASTIC PYROLYSIS PROCESS

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Abstract: Most plastic materials are non-biodegradable. Thus, disposing of such materials in landfills is undesirable, not only because of environmental concerns but also because of relevant EU policies requiring a certain degree of recycling and reuse of plastic materials. Furthermore, significant quantities of biomass waste, particularly waste sawdust as a consequence of intensive wood processing, represents another problem. Co-pyrolysis techniques have received much attention in recent years because they provide an alternative way to dispose of and convert waste plastic and biomass into high value feedstock and fuels. Recent investigations have shown that biomass and plastic co-pyrolysis achieve a synergistic effect, in the form of increased yield of liquid products, and the improvement of the overall process efficiency. This paper presents the results of technical analysis of waste plastics, waste biomass and mixtures biomass/plastic in the ratio 1:1; 3:1 and 1:3. The most common types of plastic waste in municipal waste: high density polyethylene, polypropylene and polystyrene, as well as two distinctive types of biomass, sawdust beech and spruce sawdust were selected for this investigation. The following parameters were determined: moisture, ash, coke residue, bonded carbon, volatile matter and combustible materials. During the test, the conditions of pyrolysis were simulated, in order to observe the changes of volatile substances in a mixture of biomass/plastic in comparison to theoretically expected values. The results of conducted measurements show that there is an increase in volatile matter, in all the mixtures and their ratios. The largest deviation of volatile matter in relation to the expected theoretical values was observed in the mixtures of sawdust beech/ polystyrene in the ratio 1:1.

Keywords: pyrolysis, biomass, plastic, waste.

1. INTRODUCTION

There is a constant increase in production of plastic waste, as a result of the continuing increase in production and consumption of plastic materials. In the fifties of the last century, the annual production of plastic amounted to about 5 million tons [1], while in 2007 the production was 260 million tons [2]. Majority of plastic materials are non-biodegradable and with a low density, which makes them undesirable for disposal in landfills [3]. Also, disposal in landfills implies an irreversible loss of valuable raw materials and energy. Realizing the importance of this issue, the EU has adopted a series of Directives that require a certain level of recycling and reuse of such materials, and reducing the amount of waste that ends up in a landfill [4,5,6,7]. Also the Republic of Srpska Government issued The

Regulation on the Management of Packaging and Packaging Waste. This Regulation, in line with the current EU Directive, treats the problem of waste plastics, and sets a goal of 22.5% of recycling and generally promotes reuse, recycling and energy recovery of waste plastic [8].

There are a number of technologies for recycling of waste plastic today, among which the technologies of thermochemical (raw material) recycling i.e. pyrolysis processes are of special interest. These processes have advantages over conventional mechanical recycling processes because of the possibility of using mixtures of different types of waste plastic, which does not need to be pre-cleaned of contaminants. Unlike the incineration process, much less waste gas is emitted and there is a possibility of getting valuable product bio-oil, which can be used as fuel or as raw material in industry [9].

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Despite significant progress in the field of recycling in Western Europe, 61% of plastic waste is still disposed of in landfills. The remaining 39% is recycled, while only 2% (0.35 million tons) undergoes chemical recycling processes [9]. Official national statistics provide an insight only in the amount of plastic waste that is produced on an annual basis, while the data on possible quantities of waste that is subject to the recycling process are not available. However, it can be assumed that these quantities are far below the Western Europe's average, and that most of this waste ends up unused and dumped in the official waste disposal sites, in the best case. Concerning the waste biomass, there is also a lack of official data, but according to the available literature it can be stated that there is a significant potential, as a result of intensive primary and secondary wood processing industry. It is estimated that in these industries, about $1.14 \times 10^6 \text{ m}^3$ of wood waste is generated annually at the state level [10].

In recent years, studies of biomass and plastics pyrolysis process have become increasingly important, providing an alternative way of disposal and conversion of these basically waste materials, into valuable raw materials and fuels. Several authors state in their research that it is possible to jointly pyrolyse (co-pyrolyse) biomass and plastics to achieve a synergistic effect in terms of increasing the yield of liquid products of the process [11, 12, 13, 14, 15, 16]. However, while several other authors state that their research failed to notice, or at least not to a significant level, a synergistic effect of co-pyrolysis of plastics and biomass [17,18,19], this has opened up the way for further research and characterisation of process under different operating conditions. What is characteristic for the above mentioned studies was that they were conducted both in different experimental conditions, often insufficiently explained, and in different types of reactors. Also, these studies treated different types of plastic materials, different types of biomass, different relative shares of plastics and biomass and the different granulation of raw materials particles. All this makes the comparison of these processes quite complex. In most researches, clean samples (freshly synthesized) of individual polymers or their mixtures were used with fixed shares of chosen polymers, while in some researches the variation of these shares were conducted, in order to investigate the optimal share of the individual components. All this further complicates the analysis and comparison of different systems and experimental conditions.

In this work, considering the above facts, preliminary research of possibilities of the pyrolysis and co-pyrolysis of waste plastic and biomass (specific

for our region) have been conducted. Within the possibilities of the technical analysis of these wastes, inert conditions were simulated to determine the total content of volatile matter. Tests have been conducted of individual samples of waste plastic and biomass, as well as of their characteristic blends. Volatile substances that are released during the thermal decomposition of organic material while determining coke residue in the technical analysis, also arise in the process of pyrolysis where, after a series of reactions, primarily in the gas phase, some of them condense in the appropriate system of fast cooling giving a product bio-oil.

The aim of this study was to determine a possible increase in the yield of volatile matter in investigated blends of waste plastic and biomass, compared to theoretically expected values. Theoretical values are obtained by simple computation involving relative proportions of components of the mixture and its yield in the conditions where the components of the mixture do not interact.

The studies were carried out within the realization of the project of research of waste plastic and biomass co-pyrolysis, co-financed by the Ministry of Science and Technology of the Republic Srpska Government, and were used to further the development of facilities for the pyrolysis of waste materials.

2. MATERIALS AND METHODS

The common kinds of waste plastics in municipal solid waste, polypropylene (PP), High Density Polyethylene (HDPE), polystyrene (PS), and two specific types of biomass in the region, sawdust beech and spruce sawdust, were used as materials for the study. Samples of waste plastics were prepared using packaging waste, while sawdust, taken at the sawmill in Mrkonjic Grad, was used as waste biomass.

Samples were first crushed, homogenised and stored in bottles. All samples were dried to constant moisture in order to avoid the effects of humidity variations in raw material. Technical analysis is carried out on individual samples of spruce, beech, PP, HDPE and PS, and then on a mixture of spruce and beech with certain types of plastics in weight ratios of 1:3, 1:1 and 3:1. The following parameters were observed: moisture content, ash, coke residue, combustible matter, volatile matter, and fixed carbon. The tests were conducted according to the regulations governing the technical analysis of solid fuels [20].

In determining the total volatile matter, the samples were heated and annealed at 900°C in an inert atmosphere, i.e. conditions of pyrolysis were simulated. All tests were conducted at the Laboratory of Environmental Engineering, Faculty of Technology of Banja Luka University.

3. RESULTS AND DISCUSSION

The results of the technical analysis of biomass samples are given in Table 1. It can be seen that there is an approximately equal content of volatile matter in spruce and beech, 86.17% and 84.26%, respectively. The content of coke residue in spruce is 13.83%, and 15.74% in the beech, which corresponds to approximately the same content of cellulose and lignin in spruce and beech, whereas the thermal decomposition of lignin provides 1.5 times more than the coke residue of cellulose [21].

Table 1. Proximate analysis of sawdust of spruce and beech

Parameter	Spruce	Beech
Moisture, %	20,04	18,26
Ash, %	3,19*	2,37*
Coke, %	13,83*	15,74*
Fixed Carbon, %	10,64*	13,37*
Combustible matter, %	96,81*	97,63*
Volatile matter, %	86,17*	84,26*

*Dry samples.

Results of technical analysis of plastics materials are given in Table 2. The existence of some, although minimal, ash content in all tested samples is evident, i.e. 0.80% for PP, HDPE 0.54% and 0.23% for PS. This is a common characteristic of waste plastics materials, as "clean" plastics do not contain minerals and combust completely. Since the tested samples of plastic are in origin of products intended for domestic use, various additives are introduced to the plastic during the manufacturing process and given also a possibility of contamination during the use of packaging materials, the presence of some ash in tested samples is quite expectable. Therefore, the content of volatiles is not 100%, but 99.20% of HDPE, PP at 99.46% and 99.77% in PS. Moisture and fixed carbon were not detected.

According to a research of the Norwegian University of Science and Technology, the yield of volatile combustible matter depends on the biomass

feedstock pretreatment, but first of all on the plant species being used [22]. Chemically processed biomass, in terms of the resulting product based on cellulose, has less than 1% of lignin itself, while mechanically processed biomass has about 26% of hemicellulose and 28% lignin. Ash content varies from 1% in plants and paper for printing, up to 28% in cardboard paper, while the related carbon ranges from 10% in paper and printing plants up to 5% in the card, which has a considerable impact on the yield of volatiles. Participation of oxygen in the raw material is about 45%, and it dramatically reduces the heating value of pyrolysis-generated products.

Table 2. Proximate analysis of high-density polyethylene (HDPE), polypropylene (PP), polystyrene (PS).

Parameter	PP	HDPE	PS
Moisture, %	0,00	0,00	0,00
Ash, %	0,80*	0,54*	0,23*
Coke, %	0,80*	0,54*	0,23*
Fixed Carbon, %	0,00*	0,00*	0,00*
Combustible matter, %	99,20*	99,46*	99,77*
Volatile matter, %	99,20*	99,46*	99,77*

*Dry samples.

On the other hand, plastic materials are composed exclusively of carbon and hydrogen with calorific values 2-4 times higher than the average calorific value of biomass materials. There are no inorganic substances in plastic materials, or coke, which guarantees a high yield of volatiles. The strength of C-C bond is 347 kJ / mol, and of C-H bond it is 471 kJ / mol, which indicates that under controlled conditions, a weaker C-C bond will always break with formation of shorter hydrocarbon chains. Thermal decomposition is affected by the type of a substituent existing on polyolefin skeletons, so the PS decomposes more easily compared to other plastics materials. Table 3 presents the value of technical analysis of samples of spruce and paper, and samples of HDPE, PP, PS and PVC as a representative of plastic materials, according to the mentioned research [22]. As it can be seen, the given values confirm the results of our study (Table 1, Table 2).

The results of the technical analysis of relevant mixture of spruce, beech, PP, HDPE and PS are presented in Tables 4-9. The purpose of this study was to determine the optimal ratio of biomass and plastic, for better yield of volatile combustible matter as well as to explore possible changes in volatiles compared to theoretically expected values.

Table 3. Proximate analysis of some types of biomass and plastics [22].

Sample	Volatile matter (%)	Fixed Carbon (%)	Ash (%)
Newsprint	88,5	10,5	1,0
Cardboard	84,7	6,5	8,4
Recycled paper	73,6	6,4	20,2
Coated paper	67,3	4,7	28,0
Spruce	89,6	10,2	0,2
HDPE	100,0	0,0	0,0
LDPE	100,0	0,0	0,0
PP	100,0	0,0	0,0
PS	99,8	0,2	0,0
PVC	94,8	4,8	0,4

Table 4. Proximate analysis of mixture of spruce and polypropylene (PP) at ratios of 1:3, 1:1, 3:1.

Parameter	Spruce/PP, (1:3)	Spruce/PP, (1:1)	Spruce/PP, (3:1)
Moisture, %	5,01	10,02	15,03
Ash, %	0,74*	1,70*	2,26*
Coke, %	1,54*	4,99*	10,78*
Fixed Carbon, %	0,80*	3,29*	8,52*
Combustible matter, %	99,26*	98,30*	97,74*
Volatile matter, %	98,46*	95,01*	89,22*

* Dry samples.

Table 5. Proximate analysis of mixture of spruce and high-density polyethylene (HDPE) at ratios of 1:3, 1:1, 3:1.

Parameter	Spruce/HDPE, (1:3)	Spruce/HDPE, (1:1)	Spruce/HDPE, (3:1)
Moisture, %	5,01	10,02	15,03
Ash, %	1,00*	1,35*	3,94*
Coke, %	1,42*	3,82*	8,59*
Fixed Carbon, %	0,42*	2,47*	4,65*
Combustible matter, %	99,00*	98,65*	96,06*
Volatile matter, %	98,58*	96,18*	91,41*

* Dry samples.

Table 6. Proximate analysis of mixture of spruce and polystyrene (PS) at ratios of 1:3, 1:1, 3:1.

Parameter	Spruce/PS, (1:3)	Spruce/PS, (1:1)	Spruce/PS, (3:1)
Moisture, %	5,01	10,02	15,03
Ash, %	0,50*	1,30*	1,74*
Coke, %	0,51*	5,42*	8,00*
Fixed Carbon, %	0,01*	4,12*	6,26*
Combustible matter, %	99,50*	98,70*	98,26*
Volatile matter, %	99,50*	94,58*	92,00*

* Dry samples.

Table 7. Proximate analysis of mixture of beech and polypropylene (PP) at ratios of 1:3, 1:1, 3:1.

Parameter	Beech/PP, (1:3)	Beech/PP, (1:1)	Beech/PP, (3:1)
Moisture, %	4,57	9,13	13,70
Ash, %	1,14*	1,49*	1,97*
Coke, %	1,84*	4,29*	8,86*
Fixed Carbon, %	0,70*	2,80*	6,89*
Combustible matter, %	98,86*	98,51*	98,03*
Volatile matter, %	98,16*	95,71*	91,14*

* Dry samples.

Table 8. Proximate analysis of mixture of beech and high-density polyethylene (HDPE) at ratios of 1:3, 1:1, 3:1.

Parameter	Beech/HDPE, (1:3)	Beech/HDPE, (1:1)	Beech/HDPE, (3:1)
Moisture, %	4,57	9,13	13,70
Ash, %	0,64*	1,27*	1,94*
Coke, %	0,75*	3,17*	6,55*
Fixed Carbon, %	0,11*	1,90*	4,61*
Combustible matter, %	99,36*	98,73*	98,06*
Volatile matter, %	99,25*	96,83*	93,45*

* Dry samples.

The highest content of combustible volatiles matter has been shown in the following mixture: spruce and PS (1:3) 99.5%, beech and PS (1:3) 99.31%, and beech and HDPE (1:3) over 99, 25%. The minimum content of combustible volatiles matter was shown in a mixture of spruce and polypropylene (3:1), with its value of 89.22%. For the sake of easier review, the contents of volatiles of all the

tested material and their mixtures are presented in Figure 1.

Table 9. Proximate analysis of mixture of beech and polystyrene (PS) at ratios of 1:3, 1:1, 3:1.

Parameter	Beech/PS, (1:3)	Beech/PS, (1:1)	Beech/PS, (3:1)
Moisture, %	4,57	9,13	13,70
Ash, %	0,60*	1,20*	2,14*
Coke, %	0,69*	2,29*	7,40*
Fixed Carbon, %	0,09*	1,09*	5,27*
Combustible matter, %	99,40*	98,80*	97,86*
Volatile matter, %	99,31*	97,71*	92,60*

* Dry samples.

It is noticeable that the highest content of volatiles is shown in the mixture of biomass / plastic 1:3, then 1:1, and least values are shown in 1:1 mixture. This is to be expected, since plastic materials studied have a much higher content of volatiles

(over 99% - Table 2), in relation to the content of volatiles of spruce and beech, 86.17% and 84.26%, respectively (Table 1). However, it is particularly interesting to compare the contents of volatiles in the investigated compounds with the expected (theoretical) values. Theoretical values are obtained by simple computation involving relative proportions of components of the mixture and its yield in the condition where the components of the mixture do not interact, according to the following expression:

$$g = \frac{f_B \cdot g_B + f_P \cdot g_P}{m_B + m_P} \quad (1)$$

where: f_B, f_P - are weight fractions of biomass and plastics in the mixture; g_B, g_P - are volatile yields in the experiments with pure samples of biomass and plastic; and m_B, m_P - is the weight of dry samples of biomass and plastics in their mixture.

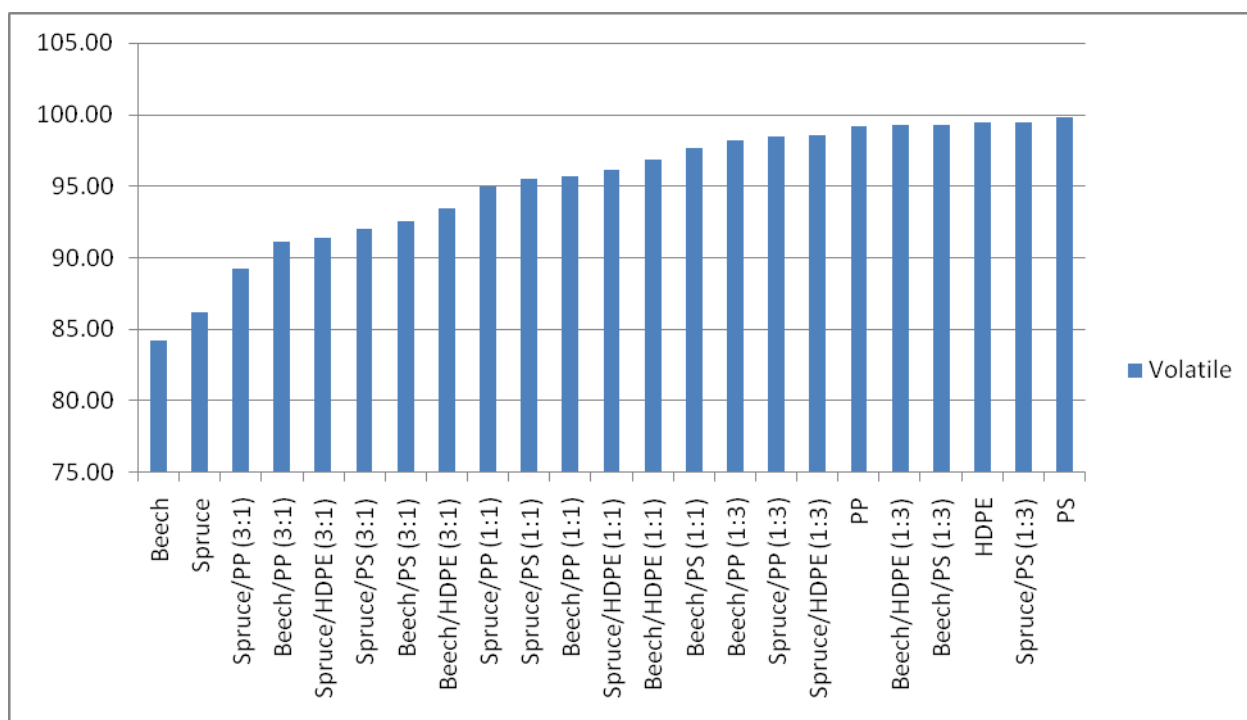


Figure 1. Content of combustible volatile matter in the samples.

In Table 10 theoretical and experimental values of volatile compounds in the tested blends of biomass and plastic are presented. The differences of a few percent are evident, and all tested blends show higher experimental values of volatiles compared to the expected theoretical values. The highest deviations, compared to the expected theoretical values, were observed in 1:1 mixtures of sawdust beech /

polystyrene. Theoretically expected value, for a given mixture of 1:1, is 91.98%, while the experimentally obtained value is 97.71%.

According to the TGA analysis of pyrolysis of commercial (virgin) polymers: low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropene (PP) and particles of sawdust of Chinese pine, particles of sawdust thermally decompose

at lower temperatures (in the range 292-480 °C), compared with the investigated polymers (HDPE 477-521 °C; LDPE 438-509 °C, 447-503 °C) [23]. The same experiment was conducted with the mixtures of sawdust/HDPE - 30:70, sawdust/LDPE - 55:45 and sawdust/PP - 30:70, and it was observed that thermal decomposition began at a lower temperature compared to pure polymers. Furthermore, TGA analysis of the mass loss of these pure samples and mixtures shows a 6-12% increase of the mass loss in the range of 530-650 °C, and the synergistic effect of these compounds is confirmed [23]. This is consistent with our results (Table 10), obtained under other conditions (thermal decomposition at 900 °C), on the other mixture with plastic waste samples. The results of TGA analysis of co-pyrolysis of mixture of plastic samples HDPE/PP/PET (1:1:1) with different ratios of biomass (biomass/plastic - 9:1, 3:1, 1:1, 1:3) [24], also supports the above statement. In these studies, an increase in mass loss around 15-50% at temperatures around 450 °C was observed.

Table 10. Theoretical and experimental values of volatile matter in the samples.

Samples	Theoretical values of volatile matter, %	Experimental values of volatile matter, %
Spruce/PP (1:3)	95,95	98,46
Spruce/PP (1:1)	92,83	95,01
Spruce/PP (3:1)	89,48	89,22
Spruce/HDPE (1:3)	96,15	98,58
Spruce/HDPE (1:1)	92,95	96,18
Spruce/HDPE (3:1)	89,72	91,41
Spruce/PS (1:3)	96,47	99,50
Spruce/PS (1:1)	92,89	95,48
Spruce/PS (3:1)	89,79	92,00
Beech/PP (1:3)	95,37	98,16
Beech/PP (1:1)	91,57	95,71
Beech/PP (3:1)	88,35	91,14
Beech/HDPE (1:3)	95,66	99,25
Beech/HDPE (1:1)	91,85	96,83
Beech/HDPE (3:1)	88,14	93,45
Beech/PS (1:3)	95,70	99,31
Beech/PS (1:1)	91,98	97,71
Beech/PS (3:1)	88,51	92,60

Additionally, it is assumed that hydrogen atoms move from the macromolecules of plastic to formed macromolecules of radicals from biomass, and set their stabilization, i.e. there is no further degradation of resulting compounds, the so-called secondary and tertiary reactions, which reduces the yield of bio-oil. Finally, this is reflected as greater

yield of bio-oil produced from biomass compared to the yield of bio-oil from pyrolysis of pure biomass [11,12,13]

4. CONCLUSIONS

Based on conducted research of simulated pyrolysis of samples of biomass (sawdust of spruce and beech) and typical types of waste plastics (PP, HDPE, PS), and theirs mixtures, the following conclusions can be derived:

- Spruce and beech wood, as a biomass and plastic waste (PP, HDPE, PS), due to a high content of volatiles, are a significant feedstock for pyrolysis process;

- Analyzed types of biomass and plastics waste can be mixed with optimal yield of combustible volatile matter;

- The experimentally obtained values of total volatile matter in all analyzed biomass/plastic mixtures are higher than expected theoretical value, and the occurrence of interaction of biomass and plastic particles, i.e. synergy effect in the given mixtures, can be assumed.

- The maximum deviation of volatile matter content compared to the theoretical expected values was observed with the mixture of beech/PS, in 1:1 ratio. The theoretically expected value, for a given mixture of 1:1, is 91.98%, while the experimentally observed value is 97.71%.

- For the successful control of the processes, from the point of view of obtaining the desired products, it is extremely important to know the mechanisms of thermochemical decomposition of the given compounds, as well as the defining performance of processes.

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ПРЕЛИМИНАРНА ИСТРАЖИВАЊА ПРОЦЕСА ПИРОЛИЗЕ ОТПАДНЕ БИОМАСЕ И ПЛАСТИКЕ

Сажетак: Већина пластичних материјала није биоразградљива те је непожељно њихово одлагање на депоније, како са становишта заштите животне средине тако и са становишта важеће политике ЕУ, која захтијева одређен степен рециклаже и поновне употребе поменутих материјала. Такође, посебан проблем представљају и велике количине отпадне биомасе, прије свега пиљевине која се јављаја као посљедица интензивне примарне прераде дрвета. Посљедњих година

технологија пиролизе постаје све значајнија, јер обезбјеђује алтернативни начин збрињавања и конверзије пластичних материјала и биомасе у вриједне сировине и горива. Скорашња истраживања показују да се заједничком пиролизом биомасе и пластике, копиризмом, остварује синергетски ефекат, у виду повећања приноса течних продуката пиролизе те се побољшава укупна ефикасност процеса. У овом раду су представљени резултати техничке анализе отпадне пластике, отпадне биомасе и смјеса биомаса/пластика у омјерима 1:1, 3:1 и 1:3. Изабране су најзаступљеније врсте отпадне пластике у комуналном отпаду: полиетилен високе густине, полипропилен и полистирен, као и двије карактеристичне врсте отпадне биомасе, пиљевина букве и пиљевина смрче. Одређивани су сљедећи параметри: влага, пепео, коксни остатак, везани угљеник, испарљиве материје и сагорљиве материје. Током испитивања симулирани су услови пиролизе, како би се уочила промјена садржаја испарљивих материја у смјеси биомаса/пластика у односу на теоријски очекиване вриједности. Резултати проведених мјерења показују да долази до повећања садржаја испарљивих материја, у свим анализираним смјесама и њиховим омјерима. Највећа одступања садржаја испарљивих материја, у односу на теоријски очекиване вриједности, уочена су код смјесе пиљевина букве/полистирен у омјеру 1:1.

Кључне ријечи: пиролиза, отпад, пластика, биомаса.