Heavy metal contaminated energy crops as a local energy carrier – fixed bed gasification process of the pretreated feedstock

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ABSTRACT

In the work experimental analysis of the fixed bed gasification (FBG) of the heavy metal contaminated (HMC) samples of energy crops *Miscanthus* x *giganteus, Sida hermaphrodita* and *Spartina pectinata* was presented. The samples were taken from HMC arable land located in Bytom (southern part of Poland, Silesian Voivodship). The land was a subject of the remediation process. During the plots harvesting, the following options were analysed: (1) control samples plots harvested from soil without additives, (2) samples taken from the soil with N, P and K fertilizer addition and (3) samples taken from land with the microbial inoculum addition to stimulate biomass growth and minimize the negative effect of pathogens. The gasification experiment was carried out using a fixed bed facility. In this study a number of gasification experiments were done by varying the air ratio from 0.12 to 0.27.
The influence of the type of the feedstock pre-treatment method (control/NPK addition/inoculum addition) and the type of the feedstock on the gasification gas parameters (the Lower Heating Value, LHV) and temperature distribution in gasifier was analysed. Results show that LHV was found to be low and it starts to rise until the optimum air ratio of 0.18 and later drops for higher air ratio. The best effect of the LHV increment is visible for NPK addition effect for, *Sida hermaphrodita* sample.

KEYWORDS: remediation; inoculum and fertilizer application; energy crops; heavy metals; gasification; fixed bed installation.

INTRODUCTION

Remediation of contaminated soils has become a long-term challenge as it addresses both scientific and technical aspects as well as social issues (rehabilitation of former industrial sites in ecodistricts, restoration of ecosystem services, etc.) and economic issues (markets of soil rehabilitation; production of plant biomass for feedstock on contaminated soils integrated in the biobased-knowledge for bioeconomy) [1]. In spite of the importance of management options for sustainable and safe use of heavy metal contaminated (HMC) soils, little has been investigated on combining the production of energy crops on the contaminated areas with phytoremediation of these sites. Whereas HMC soils are unsuitable for food production, energy crops can allow the commercial exploitation of these soils by establishing biofuel feedstock production systems. In addition, the cultivation of plants offers opportunities for site stabilization and phytoremediation of contaminated soils [2]. There is a number of typical energy crop species available on the market which have also been tested with success for phytoremediation effect on HMC arable land. They, however, need further tests for different heavy metals to prove their robustness for large scale applications.

Until now species used in Poland as well as in other European Union (EU) countries as energy crops are different clones of willow and poplar [3], Miscanthus [4, 5], Switchgrass [4, 5, 6] and Virginia mallow [7]. All these species are normally grown on non-contaminated agricultural land. Among above listed plant species only willow [8, 9], Switchgrass [10] and Miscanthus [2, 11] were also used for phytoremediation of heavy metal contaminated sites. A well known energy crop *Miscanthus* x *giganteus* was indicated as a plant which could be successfully cultivated on heavy metal contaminated soil as a safe energy crop as it does not accumulate heavy metals [12, 13]. Previous results show that, the amount of metals observed in crops suggests that Miscanthus has a promising potential for Pb and Zn phytoremediation [14, 15] and thus shows also phytoextraction properties. These proven features of *Miscanthus* x *giganteus* open a new potential for the use of this species as both energy crop and for phytoremediation purposes. At the same time there is a number of other plant species (e.g. *Sida hermaphrodita*, *Spartina pectinata*) used as energy crops, but only a few authors tested them for heavy metal phytoremediation purposes [16, 17].

A field experiment has been initiated on heavy metal contaminated sites located in Poland (arable land). It involves testing of 3 preselected plant species: Miscanthus (*Miscanthus* x *giganteus*), Virginia mallow (*Sida hermaphrodita*) and Cordgrass (*Spartina pectinata).* After remediation process, produced fuel is characterized by high heavy metal content. The safe and effective utilization of such fuel can be difficult. Nevertheless, thermal methods (combustion, gasification and pyrolysis) seem to be a good solution for that purpose. Moreover, previous results shows [18] that gasification has several advantages over a traditional combustion process, such as a higher energy recovery and lower-cost atmospheric emission control. As a consequence of the reducing atmosphere, gasification prevents emissions of sulphur and nitrogen oxides, heavy metals and the potential production of chlorinated dibenzodioxins and dibenzofurans. Heavy metals from HMC biomass may be volatilized to the gas phase at high temperature while other elements may be retained in the solid residue, trapping some of sulphur, nitrogen and chloride introduced by the feedstock [19].

The aim of the work is the investigation of the influence of the properties of the pretreated HMC energy crops used for remediation purposes on the properties of the gas obtained in the gasification process.

2. MATERIALS AND METHODS

Site description

The test site is located in the Upper Silesian Industrial Region, on the outskirts of Bytom - an industrial city about 15 km from Katowice, in the proximity of a shut down large lead/zinc/cadmium works consisting of the ore mining, enriching and smelting facilities. This metallurgical complex was in operation for more than 100 years and contributed significantly to the contamination of the local soils. During the last 30 years the area was used for agricultural purposes. Recently the land has been used for grain crops farming, especially for wheat production. Soil contamination with lead, cadmium and zinc in this area exceeds permissible limits for agricultural soil in Poland.

Site experiment design

Three plants were selected for field trials: Miscanthus (*Miscanthus* x *giganteus*), Virginia mallow (*Sida hermaphrodita*) and Cordgrass (*Spartina pectinata*). Experimental plots (16m^2 each) were established in spring 2014 for each test site (Figure 1). Between the plots a 4 m buffer zone was left to avoid interconnection between experimental options. *Miscanthus* x *gigantheus* was planted from rhizomes, *Sida hermaphrodita* and *Spartina pectinata* from root seedlings. Two weeks before planting ammonium sulphate and Polifoska

(Grupa Azoty Zakłady Chemiczne "Police" S.A., Poland) were applied to the NPK fertilized subplots. Soil at the subplots dedicated to treatment with EmFarma Plus (ProBiotics Polska, Poland) was sprayed with the inoculum solution as well as the roots of the seedlings were soaked.

Figure 1. Planting at the Bytom site

General views of experimental plots at Bytom site at the end of the second growing season (2015) is presented in Figure 2.

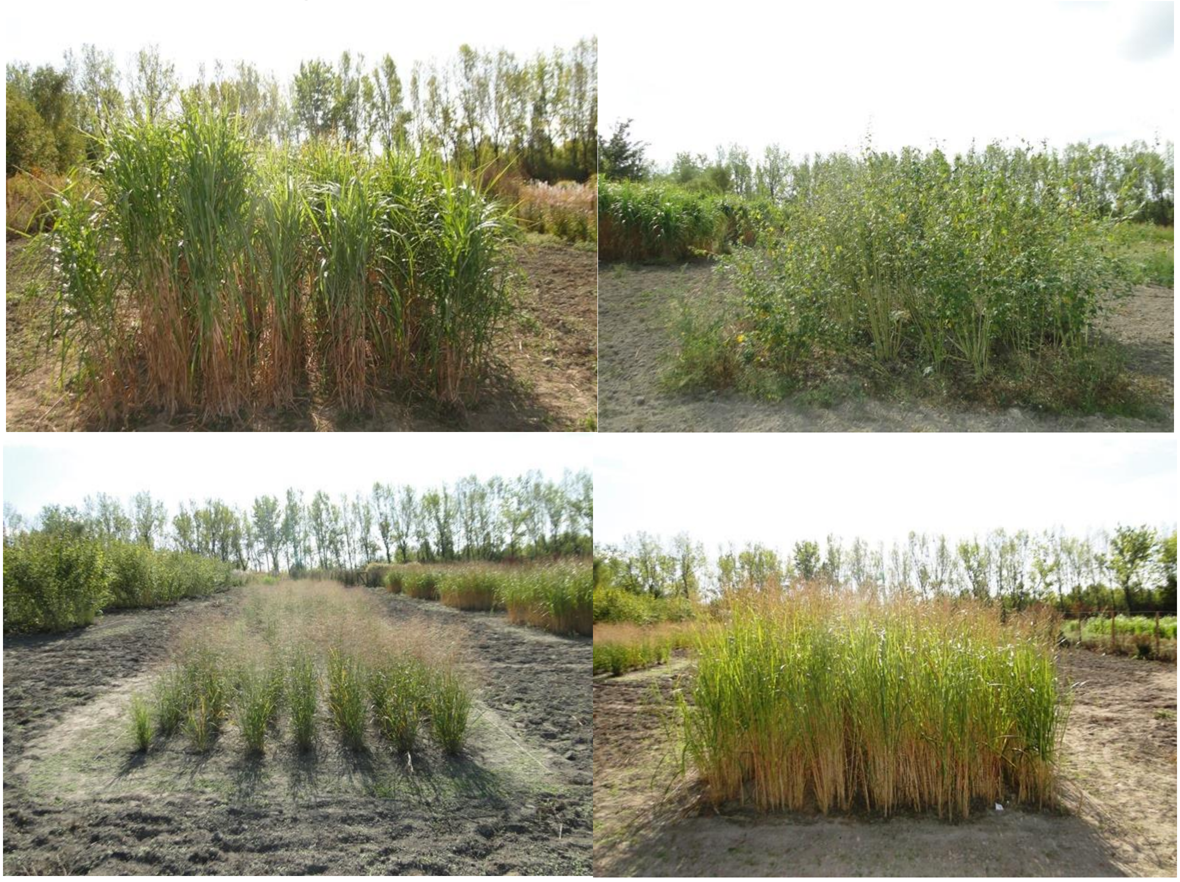

Figure 2. General view of experimental plots growth at the end of growing season

The simplest method of yield improvement is the application of chemical fertilizers to the soil before plant growth. The most common chemical fertilization relies on soil treatment with

nitrogen, phosphorus and potassium (N, P and K) elements, in proportions that are species and field specific. Chemical fertilizers are mainly responsible for enhancing the amount and availability of the most important nutrients in plant growth and development, but fertilizers can also improve soil physico-chemical parameters. Taking into consideration it, the subplots for each species include the following options:

- Control (no additives).
- NPK standard fertilization addition - calculation based on specific plant requirements, applied once before plant establishment.
- Inoculum addition - commercial microbial inoculum Em Farma Plus was applied on rhizomes before planting and on the leaves as aerosol in the middle of each month of the growing season (from May to September 2014).

Gasification experiment

The current study was conducted using a fixed-bed gasification (FBG) facility. A schematic of the facility is shown in Figure 3 [20].

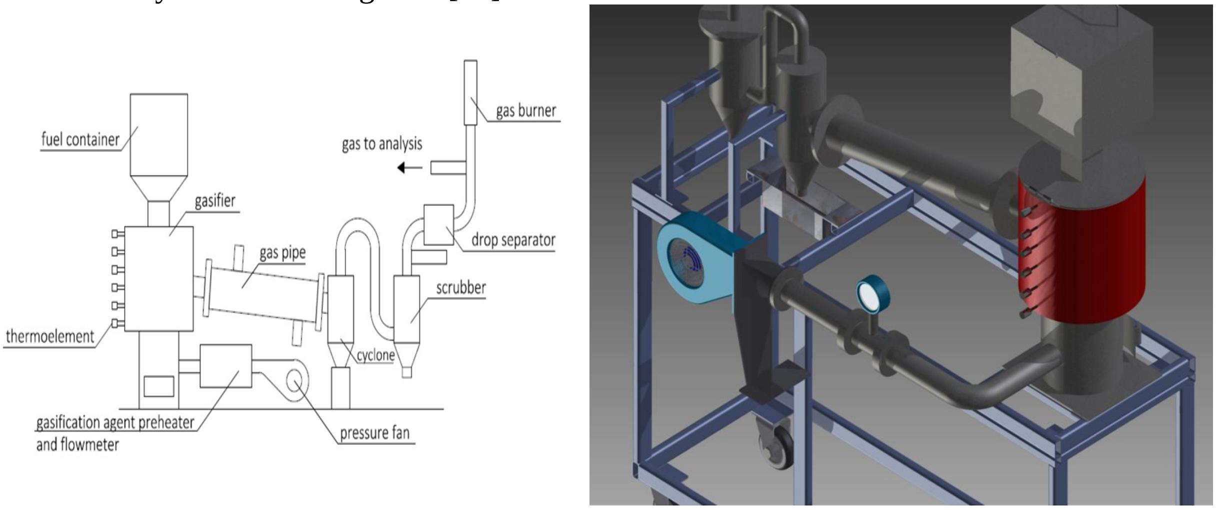

Figure 3. Scheme (left side) and visualization (right side) of the fixed bed gasification installation [20]

The main system component was a stainless reactor (gasifier) with a 150-mm internal diameter and a total height of 300 mm. The reactor was well insulated to prevent major heat loss. In this study, biomass from the fuel container was fed into the top of the gasifier. The gasification air was fed from the bottom by a pressure fan. The HMC biomass were circulated in a counter current direction to the process gases. There were four zones in the reactor. In the drying zone, water was evaporated from fuel. In the second zone (the pyrolysis zone), the biomass was thermally decomposed into volatiles and solid char. In the third zone, carbon was converted into the main combustible components of syngas. In the last zone, the remaining char was combusted. The combustion zone provided a source of energy for the gasification reactions in the upper zones. The gasification reactions are mainly endothermic. The internal reactor temperature was measured by six N-type thermoelements integrated with an Agilent temperature recording system. The thermocouples were located along the vertical axis of the reactor. The temperature of syngas produced in the gasifier was also measured at the outlet of the reactor. The gasification air flow rate and the syngas flow rate were measured by flow meters. The syngas was transported from the gasifier by a pipe. The syngas was cleaned by a cyclone, a scrubber and a drop separator. The volumetric fractions of the main syngas components were measured online using a Fisher Rosemount and ABB integrated set of analyzers. The system was also equipped with a sampling port to collect gas for

chromatographic analysis by an Agilent 6890N gas chromatograph. Table 1 shows the operating conditions for all the experiments.

Table 1

Gasification experiment methodology

Feedstock	Treatment applied	Gasification agent (t=25°C)	Air ratio λ	Parameters
(1) *Miscanthus* x *giganteus* (*MG*) (2) *Sida hermaphrodita* (*SH*) (3) *Spartina pectinata* (*SP*)	(1) No treatment (2) NPK fertilizer application (3) EmFarma Plus application	Atmospheric air	0.18	(1) Temperature distribution in gasifier (2) Gasification gas composition (3) LHV of gasification gas

The air ratio was calculated taking into account the gasification air flow rate and biomass flow ratio $\frac{m_{air}}{m_{fuel}}$. The lower heating value (LHV) in MJ m^{-3}_n of the gasification gas was estimated using the formula given below [21]:

$$LHV=0.126\cdot CO+0.108\cdot H_2+0.358\cdot CH_4 \qquad (1)$$

Feedstock properties

Picture of the samples to fixed bed gasification experiments are presented in Figure 4.

Figure 4. Picture of analyzed samples

In Tables 2 and 3 ultimate and proximate analysis of the energy crops are presented. The main components in the analyzed energy crops were determined using PO-ATI-16 Method with Perkin-Elmer 2400 analyzer. Total organic carbon determination (TOC) is done according to EN 1484:2006 [22]. The moisture was obtained following standard PN-EN 14774-3:2010 [23]. The volatile content was determined according to standard PN-EN 15402:2011 [24]. The ash content was obtained using standard PN-EN 15403:2011 [25].

Table 2

Ultimate analysis of the analysed fuels[a]

Plant	Carbon (%)	Hydrogen (%)	Nitrogen (%)	Sulphur (%)	Oxygen (%)
Miscanthus x *giganteus* –control	46.90	7.32	1.38	<0.20	44.20
Miscanthus x *giganteus* - nutrients NPK	45.50	6.88	1.13	<0.20	46.29
Miscanthus x *giganteus* – Em Farma Plus	46.50	7.13	1.49	<0.20	44.68
Sida hermaphrodita- control	46.20	6.69	0.43	<0.20	46.48
Sida hermaphrodita- nutrients NPK	46.40	7.22	0.38	<0.20	45.80
Sida hermaphrodita- – Em Farma Plus	47.00	7.06	0.30	<0.20	45.44
Spartina pectinata - control	46.70	6.33	0.32	<0.20	46.45
Spartina pectinata -nutrients NPK	46.30	6.77	0.38	<0.20	46.35
Spartina pectinata - commercial inoculum	47.00	7.07	0.59	<0.20	45.14

[a]all values on a dry basis, in mass percent

Table 3

Proximate analysis of the analysed fuels[b]

Plant	Moisture	Volatiles	Total organic carbon	Ash
Miscanthus x *giganteus* –control	8.60	74.90	21218.00	5.50
Miscanthus x *giganteus* - nutrients NPK	8.30	76.50	24442.00	4.20
Miscanthus x *giganteus* – Em Farma Plus	8.20	75.30	24002.00	4.90
Sida hermaphrodita- control	9.80	75.80	19050.00	2.70
Sida hermaphrodita- nutrients NPK	9.10	76.90	17537.00	2.40
Sida hermaphrodita- – Em Farma Plus	9.40	76.60	15151.00	4.80
Spartina pectinata - control	8.30	77.90	22534.00	3.70
Spartina pectinata -nutrients NPK	8.40	77.50	21175.00	3.40
Spartina pectinata - commercial inoculum	9.50	75.70	20521.00	3.40

[b]all values on a as received basis, in mass percent

The value obtained during the biomass experimental tests and presented above represents the average of two determinations. As it can be seen from the ultimate analysis results, the largest share in all the biomass samples is for oxygen and carbon, while nitrogen and sulphur has the smallest share. The percentage of carbon, hydrogen and oxygen are quite similar for all

biomass types. The highest nitrogen content was found for samples of *Miscanthus* x *giganteus* biomass grown on a plot with EmFarma application and NPK treatment. There are no strong differences between feedstock elemental composition so at this stage it can be concluded that treatment applied does not affect on the biomass quality. Analyzing the TOC parameter, it can be concluded that in the case of *Miscanthus* x *giganteus* treatment applied causes that organic component concentration is increasing. Taking into consideration the *Sida hermaphrodita* ash content, treatment applied causes the inreament of this value.

RESULTS AND DISCUSSION

Gasifier temperature profile

The temperature profiles in the reactor were measured by six N-type thermocouples installed at six points along the vertical axis of the reactor. The temperatures were measured at the following distances above the grate, t1: 10 mm, t2: 60 mm, t3: 110 mm, t4: 160 mm, t5: 210 mm and t6: 260 mm. Figure 5 presents the temperature values inside the gasification reactor at six different points located above the gasifier bottom. Analyzing presented results it can be concluded that the temperature at t_3 was always the highest temperature in the reactor; thus, t_3 may have been located in the partial oxidation zone, which would have been the hottest area in the fixed bed gasifier. Correspondingly, the other monitoring sites may have been located as follows: t6 and t5 in the drying zone, t4 in the pyrolysis zone, t2 in the oxidation (combustion) zone and t1 in the ash zone. It can also be concluded that there is no strong influence of the treatment applied on the gasifier temperature.

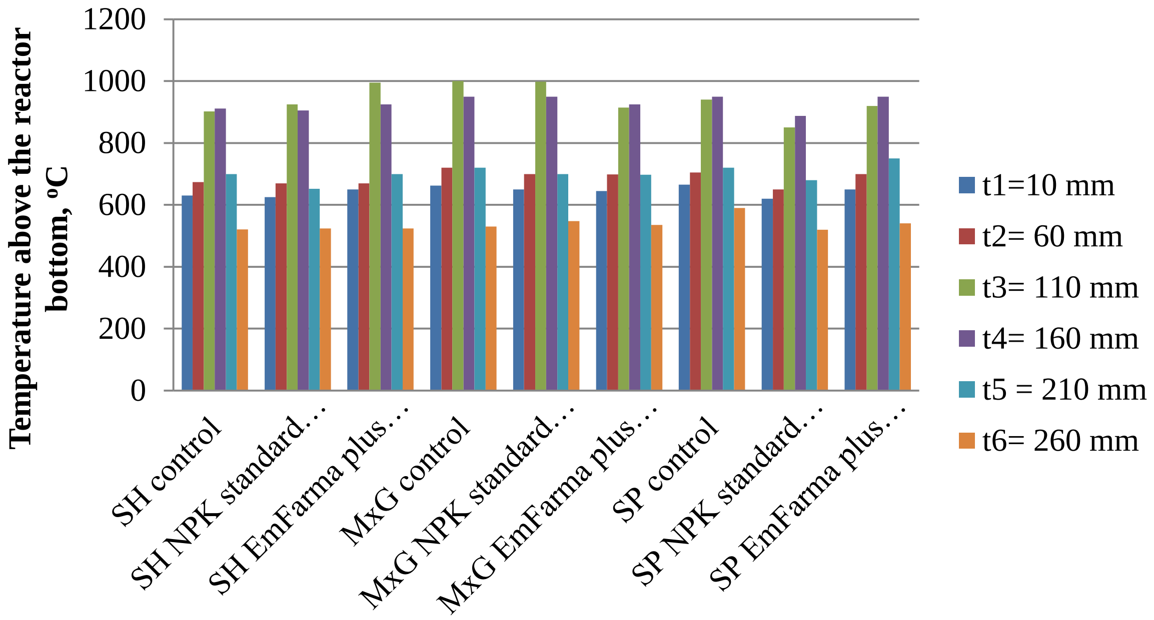

Figure 5. Temperature distribution in the reactor for all analyzed feedstock; air ratio equal to 0.18

Air ratio is a parameter that quantifies the amount of air/oxygen per unit mass of fuel, as compared to the theoretical amount of air/oxygen needed for complete combustion. Hence the optimum air ratio that favors gasification (incomplete combustion) resulting in combustible gases like CO, rather than the case of complete combustion with an air supply that mainly produces CO_2 need to be determined. In this study a number of gasification experiments were done by varying the air ratio from 0.12 to 0.27. Figure 6 presents the dependence of the Lower Heating Value (LHV) of gasification gas as a function of air ratio forof all analysed feedstock.

Analyzing that figure, it can be concluded that there is an optimal value of air ratio, in which the lower heating value of the obtained gas reaches its maximum. As expected, an increase in the air flow rate caused a decrease in the heating value. A greater amount of oxidizer increases the amounts of noncombustible species and the volumetric fraction of nitrogen, thus decreasing the heating value of the obtained gas.
Rapid growth of LHV observed in the value of the air ratio equal to 0.18 is caused by the dominant role of the primary water gas reaction. The reactions that can occur in the gasifier as a result of the gasification agent flow can be categorized as a the reaction of gasification agent and carbon in the fuel and the reaction of gasification agent and CO in the gas. The reaction of gasification air and carbon is an endothermic reaction that generates mainly CO whereas the reaction of gasification air and CO is an exothermic reaction that generates mainly CO_2 (and H_2). When gasification air is fed with the fuel into the reactor, the endothermic reaction of air and carbon occurs first (e.g. primary water gas reaction $C+H_2O \rightarrow CO+H_2$), and the CO in a gaseous state produced from the fuel reacts with the residuals causing next reactions (e.g. water gas shift $CO+H_2O \leftrightarrow CO_2+H_2$). Thus, the composition of H_2, CO and CO_2 (and simultaneously LHV) in the gasification gas changes according to the amount of the air supplied to the reactor.
Analyzing the LHV values, it should emphasize that nevertheless the analyzed feedstock, gasification process gives the opportunity to obtain valuable gaseous fuel. Such fuel can be effectively utilized in boiler, gas turbines or engines [26].

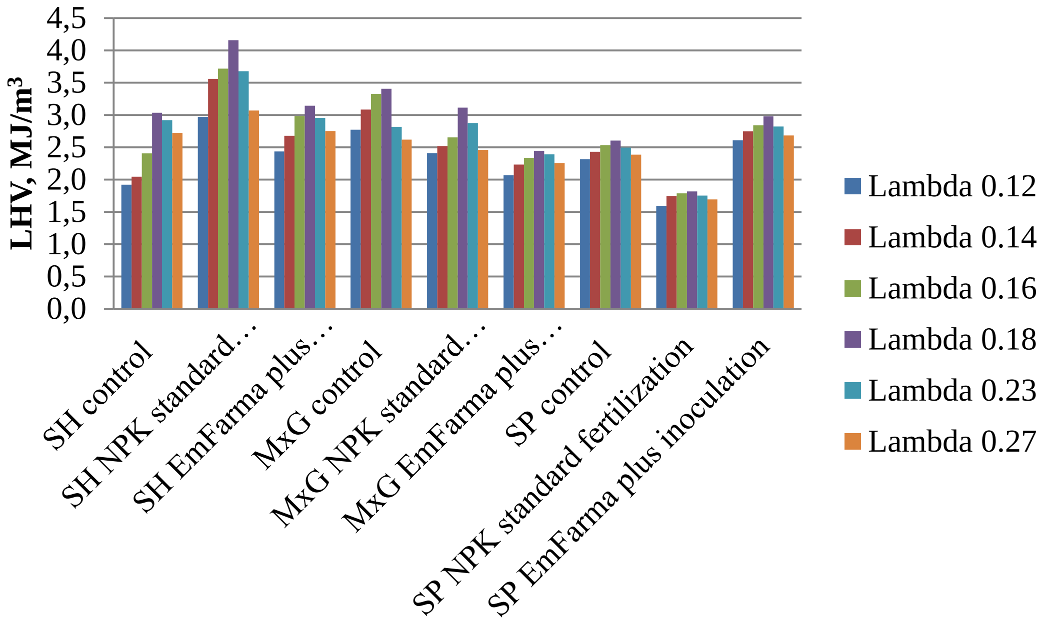

Figure 6. The Lower Heating Value (LHV) of gasification gas as a function of air ratio

In Figure 7, the influence of the treatment applied on Lower Heating Value (LHV) for air ratio equal to 0.18 is presented. The best result is obtained from the gasification of *Sida hermaphrodita* with standard fertilization, 4.156 MJ/m³. The gasification of the rest of biomass obtaining significantly lower values. More so, in all analysed treatment applied, *Sida* shows higher values of the LHV in comparison to *Spartina.*

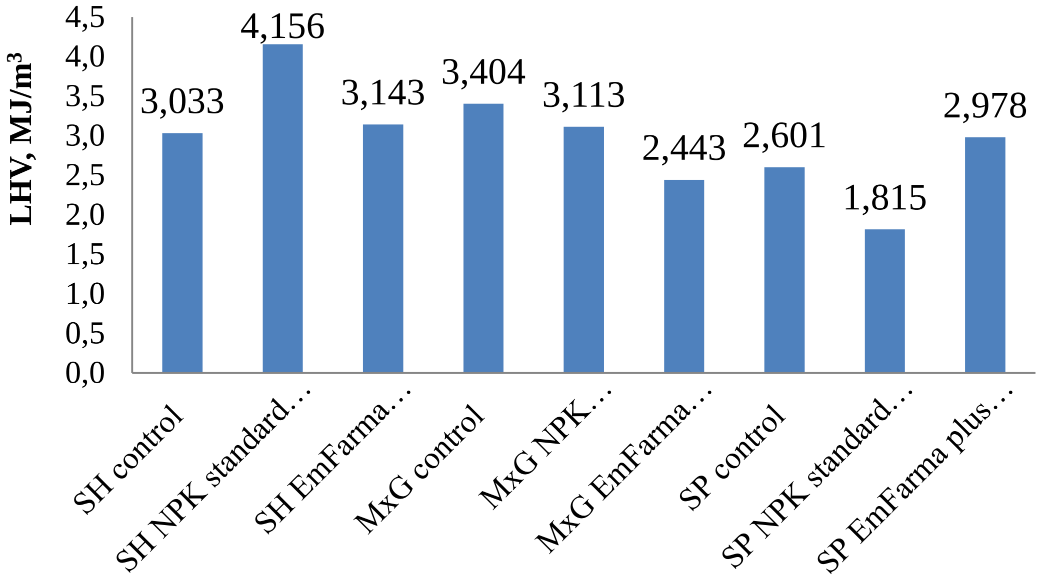

Figure 7. The LHV of gasification gas for air ratio equal to 0.18; all analysed feedstock

CONCLUSION

Phytoremediation is one of the techniques used for remediation of contaminated areas. The group of energy crops has taken into consideration include native and foreign species such as perennial dicotyledonous plants and perennial grass species. The process of heavy metal contaminated biomass in FBG installation has many advantages in comparison to the combustion process. Gasification produces a gaseous fuel, which can be used to produce energy in different types of installations. The LHV of obtaining gas is acceptable taking into consideration the usage of this gas. Gasification of HMC biomass is characterized by a lower emission of gaseous pollutants into the atmosphere in comparison to combustion as well as a majority of the heavy metals from biomass are moved into solid products. Then, the concentration of heavy metals may be reduced after recovery of them from solid products, which will help to protect the environment and increase the financial income of the owner of the gasification installation.

Thanks to the agricultural and microbiological treatment applied, remediation process can be more effective. Nevertheless, the influence of that activity on energy crops used for remediation purpose should be analyzed.

The percentage of carbon, hydrogen and oxygen are quite similar for all biomass types. The highest nitrogen content was found for samples of *Miscanthus* x *giganteus* biomass grown on a plot with EmFarma application and NPK treatment. There are no strong differences between feedstock parameters so at this stage it can be concluded that treatment applied does not affect on the biomass quality.

Presented gasification results show that there is no strong influence of the treatment applied on the gasifier temperature distribution. Additionally, there is no clear influence of the biomass quality on the gasification gas parameters, but the best result is obtained from the gasification of *Sida hermaphrodita* with standard fertilization. The gasification of the rest of biomass, obtaining significantly lower values. More so, in all analysed treatment applied, *Sida* shows higher values of the LHV in comparison to *Spartina.*

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