AGRICULTURAL RESIDUES GASIFICATION, DEPENDENCY OF MAIN OPERATIONAL PARAMETERS OF THE PROCESS ON FEEDSTOCK CHARACTERISTICS
/
ГАЗИФІКАЦІЯ СІЛЬСЬКОГОСПОДАРСЬКИХ ВІДХОДІВ, ЗАЛЕЖНІСТЬ ОСНОВНИХ ЕКСПЛУАТАЦІЙНИХ ПАРАМЕТРІВ ПРОЦЕСУ ВІД ХАРАКТЕРИСТИК СИРОВИНИ

Assoc.prof. Ph.D.Eng. Zolotovs'ka O.V. [1)], Prof.Ph.D. Kharytonov M. [1)], Ph.D. Stud. Onyshchenko O.[1)],
[1)] State Agrarian and Economic University, Center of Ecological Agriculture, Dnipro / Ukraine
Tel: 0973456227; E-mail: envteam@ukr.net

Keywords: *agricultural residues, pyrolysis unit, heat generation.*

ABSTRACT

Pilot-scale trial data on combined energy cycle – adjustment for boiler system for pyrolysis gas use had demonstrated technical feasibility of implementation of combined energy cycles for facilities where secondary agricultural residues are available in sufficient quantities.
The results of the energetic analysis of technologies of gas production from solid biomass through the pyrolysis are considered. Examples of the introduction of the technology of direct gasification of secondary agricultural residues and wood (buckwheat husk, rice husk, sunflower husk, sawdust) are shown. It was determined that optimal technological conditions for low-temperature pyrolysis processes depend on character of the feedstock and moister content in the feedstock.
Temperature and pressure pulsing were found to be most important parameters that influence gas quality and yield during pyrolysis process and optimized through developing of mathematical equations describing the process.

РЕЗЮМЕ

Результати експериментального випробування комбінованого енергетичного циклу – коригування бойлерної системи для отримання піролізного газу показали технічну доцільність впровадження комбінованих енергетичних циклів на об'єктах, де вторинні відходи сільського господарства є в достатній кількості.
Зроблений енергетичний аналіз даних технологій виробництва газу з біомаси шляхом піролізу. Наведені приклади впровадження технології прямої газифікації вторинних відходів сільського господарства і деревини (лушпиння гречки, рисового лушпиння, лушпиння соняшника, тирси). Було встановлено, що оптимальні технологічні умови для низькотемпературного піролізу процеси залежать від характеру сировини і вмісту вологи у вихідній сировині.
Зміни у температурі та тиску виявилися найбільш важливими параметрами, які впливають на якість і вихід газу та оптимізовані за допомогою математичних рівнянь, що описують процесс піролізу.

INTRODUCTION

Ukraine is among the countries which have stocks of all kinds of fuel and energy resources (oil, natural gas, coal, peat, uranium, etc.), but the coverage, production and the use are not equally distributed and they do not create the necessary energy safety level, especially in light of existing political situation. The agri-food complex is one of the most important sectors of Ukrainian economy *(Velychko O., 2015*). Agricultural waste and woody biomass are key components of renewable energy potential in Ukraine. Energy crops currently represent a "virtual" part of the potential, except of several experimental plantations (*Bielski S., 2015*). There are two major sources of the feedstock in agricultural forestry sector, which are primary, and secondary agricultural residues. Primary agricultural residues are those materials which remain in fields as by-products after the primary product of crops has been harvested. These include different materials like cereal grain straws, of wheat, barley, rice etc., corn stover (stalk and leaves) etc. Secondary agricultural residues are specific type of residues and include quite wide variety of biomass by-products of processing of agricultural products for food or feed production *(Czernik and Bridgwater, 2004*). Bagasse, sunflower husks, rice husks, nut shells, cocoa bean shells, kidney bean shells and other biomass of such kind is generated and collected at the enterprises which process agricultural crops for food/feed production. Food processing by-products represent a huge amount of waste resources that could be valorized for recovery of compounds for fuels and energy via thermos-chemical, biological and microbial methods. The biomass pyrolysis is attractive because solid biomass and wastes can be readily converted into liquid products. Although the primary agricultural residues represent the largest share of the technical potential (83 %), distribution of secondary agricultural residues are more equal and there are more options to process them using infrastructure of the facilities where agricultural feedstock is processed (*Geletukha et al, 2010*). Ukraine has quite a big potential of agricultural residues which mainly consists of straw from cereals and production residues from sunflower and maize from grain. At present, less than 1 % of the primary agricultural residues potential is used for energy purposes (combustion in boilers, production of pellets and briquettes), mainly because of undeveloped infrastructure and logistics system for the feedstock supply. The situation with secondary agricultural residues is much better though their technical potential is lower than for primary. Over 77 % of sunflower husks biomass, for example, is directly burned in boiler systems, another 20 % is used for pellets production. Almost all sunflower processing facilities have biomass boilers for utilization of sunflower husks. Agricultural residues from the waste streams of commercial processes have typically been considered to have very little inherent value, mainly constituting a disposal problem in the past. Most of the waste generated by sunflower and crop processing for bioenergy facilities are also confronted with the costs associated with collection and transportation in addition to the supply uncertainties in particular case.

Although direct burning of secondary agricultural residue was widely introduced into de practice during last 10 years, when wood or other solid biomass are directly combusted and coupled to a steam turbine, it is not possible to achieve high rate of efficiency. Only combined energy cogeneration cycles allow versatile and high effective use of biomass residues but in this case using of biomass in combustion/boiler system requires primary production of power from biomass (*Wang et al, 2014*).

As far as solid biomass cannot be fed into a gas turbine or diesel engine – a liquid or gaseous fuel is required to operate an advanced cycle, which means direct liquefaction or gasification of biomass is required.

An economic analysis was conducted for biomass gasification and pyrolysis and electricity generated to meet local market demand, including the higher-value peaking power. Biomass-based gasification eliminates the need for waste disposal and reduces electricity consumption from the grid, making it a valid investment (*Lau et al, 2002*).

Although pyrolysis technologies are more developed and available at the present day, they are preferable to others. Pyrolysis is one of thermos-chemical processes, which convert the solid biomass in to liquid (bio-oil), gas, and char. Pyrolysis has been practiced for centuries for production of charcoal. More recently, studies into the mechanisms of pyrolysis have suggested ways of substantially changing the proportions of the gas, liquid and solid products by changing the rate of heating, temperature and residence time (*Tilmann D., 2000; Wang et al, 2002*). This requires relatively slow reaction at very low temperatures to maximize solid yield (*Suri and Horio, 2010*). The diverse range of biochar applications depends on its physicochemical properties, which are governed by the pyrolysis conditions (heating temperature and duration) and the original feedstock (*Jindo et al, 2014; Lei and Zhang, 2013*).Thus, detailed information on the complete production process is a key factor in defining the most suitable application of biochars.

Biomass pyrolysis converts essentially 80–95% of the feed material into gases and bio-oil. The pyrolysis process is to maximize the production of gaseous fraction (*Williams and Besler, 2006*).

There are few fundamental experimental and theoretical studies, dealing with the biomass combustion and emission characteristics and physical and chemical properties of various biomass feed stocks (*Gaskin et al, 2008; Lei and Zhang, 2013; Mimmo et al, 2014*). There are several studies made on development of reliable kinetic and thermos-transport models for investigation of biomass thermal conversion process (*Brewer et al, 2009; Mc Beath et al, 2013*) Although some certain adjustment to every type of feedstock and to every type of combined energy cycle is required..

In light of existing situation in the alternative energy sector of Ukraine, it is important to obtain a better understanding of this technology and their potential for implementation and existing markets.

MATERIAL AND METHOD

The aim of the present study is to perform a technical and economic assessment of the pyrolysis operation as a secondary agricultural residues utilization process. The study included pilot test at the facility that might be suitable for implementation of biomass utilization combined cycle for evaluation of operating costs and revenue potential for a generic gasification process, and a cost sensitivity study. To perform general evaluation of the technological process of oxidative pyrolysis, laboratory pyrolysis unit was constructed. The general scheme of the pyrolysis unit is shown in figure 1.

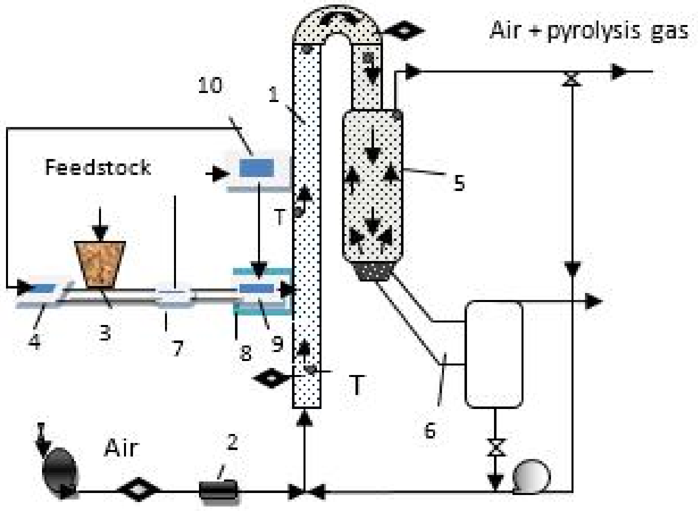

Fig.1 - The general scheme of the pyrolysis unit

C1–C6 – plan sifter compartments; Break 1–5 – break rolls; MG1, MG2 – semolina machines; M1A, M1B, M2–M6 – reduction rolls; F, F1, F2 – flour

The unit consists of elbow shaped chamber (2) with internal diameter of 100 mm with total length of 5700 mm, which allows conducting the pyrolysis on fluidized bed. In the lower part of the chamber, the air primary heated up in radiator is blown (1). Before entering the chamber, air goes through the numerous ceramic rings to average out air velocity profiles along the tube section. The feedstock is loaded into the camera by the screw dispenser. The design of the dispenser is allows to control feed volume and impermeable inlet joint. In the pyrolysis chamber inlet the automatic moister control sensor (7) was installed. Thus in the feedstock before entering the chamber and passing though the moister the moister is automatically measured. The data from sensor is automatically transformed to converter (10) which controls the feed (4).

In the pyrolysis camera satellite-lifting motion of particles is supported when satellite motion of particles velocity is 1.5 – 2 times lower than the air movement. As particles have almost similar size and mass, the layer of particles can be considered as uniform substance with average thermos-physical parameters. Drying, heating and party pyrolysis occur during the movement of the particles suspended in air. The cyclone is installed on the camera outlet where the separation the gaseous and solid phases is taking place. Solid particles fall into cyclone bunker where further devolatilization is taking place up to full decomposition. The air mixture of gaseous products of pyrolysis is sucked out through the smokestack and 10 % of the mixture is returned back each time for the gas enrichment which increases its calorific value.

To avoid the condensation of resins, which gaseous products of pyrolysis contain, the cyclone, pyrolysis chamber, bunker and connecting pipes were insulated. The experimental study on evaluation of the process of biomass thermal conversion was performed in the following stages. First stage consists in heating up the system and determining initial moister content in the feedstock. Second stage – the setting of the parameters of the process (biomass and air feed rate), which define the overall process parameters. Third stage consists in achieving stability of the process (up to 15 minutes) and determination of its general efficiency indicators - the ration of certain pyrolysis products in the mixture, temperature along the pyrolysis camera. Fourth stage consists in finishing the process and evaluating the mass balance. Comprehensive experimental study was conducted to analyze new configuration for combined cycle heating system based on pyrolysis gas generation process for most common types of secondary agricultural residues.

RESULTS

At the beginning stage, biomass heating up process results in moister evacuation (strongly marked endothermic process), where moister content, accordingly, is one of the main process indicators. It was shown the condition of the plant material which is used as a feedstock for fuel production process. This process affects certain parameters, such as heating up time, particles movement velocity, bulk yield of the volatile products, gas permeability of the waste layer and its hydraulic resistance, biomass and air feed rate, initial temperature of air heating, actual biomass feed rate. Energy potential indexes of four types of secondary agricultural residues are shown in the table 1.

Table 1

Energy potential indexes of most common types of secondary agricultural residues

Type of biomass	Moister content, %	Energy capacity MJ/kg	KW hours/ kg
Sawdust	20	14.1	3.9
	6	18.2	
Buckwheat husk	12	13.8	3.8
	2	17.9	
Rice husk	12	14.3	3.9
	2	18.5	
Sunflower husk	17	14.2	3.9
	4	18.3	

Initial moister content of processed biomass is very variable. Feedstock with higher moister content requires more energy per batch, which is supported by increasing of temperature during the process of organic matter conversion. In order to adjust moister control sensor, the moister content was measured for biomass samples (sawdust) with moister content 1–40 %. Results of the measurements and optimal diapasons of drying for each type of biomass are depicted on plots “a”, “b”, “c” and ”d” in the figure 2.

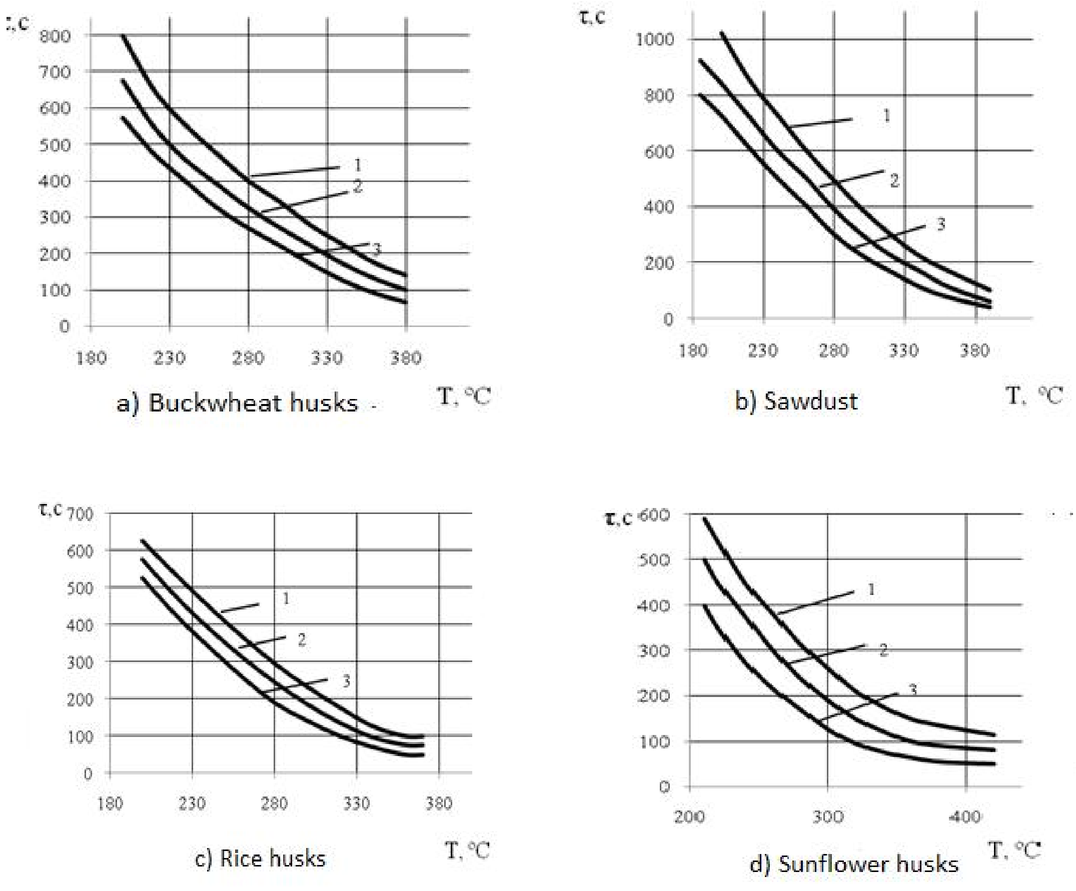

Fig.2 - Dependence between heating temperature and moister content for different types of processed biomass

1 – particle with 40 % moister content; 2 – particle with 20 % moister content; 3 – particle with zero moister content.

After few runs of the pyrolysis unit analysis of the obtained data it has shown that it was necessary to decrease of the feedstock moister content, because overall energy gain of the process depends on the energy capacity of the feedstock entering the pyrolysis chamber. Whereas biomass-drying stage is included into pyrolysis unit scheme, the adjustment of the drying and pyrolysis regimes is required. Such adjustment, as experimental study demonstrated, could be performed by incorporation of online moister control automatic sensor. To support required temperature in the camber, it is crucial to provide certain feedstock and air proportion which can be defined theoretically. Accordingly, there is certain dependency of decomposition process parameters for the feedstock with certain moister content on the temperature regime and amount of air in mixture. The regulation of gas air mixture proportions also can be achieved by regulation of pressure pulsing. To evaluate the rate of impact of thermal treatment regimes on the quality and content of obtained gas mixture, a mathematical model was developed. As main indicator of obtained gas quality the volumetric output of gaseous products from the feedstock was taken as those having impact on the indicators of Y group, such parameters as amount of air (X_1), air temperature (X_2), moister content in the feedstock (X_3) and air pressure drop (X_4). General data obtained during experimentation with pyrolysis unit regimes are given in the table 2.

Table 2

Main process factors variation levels

№	Factor	Symbol	Variation levels					Δ
			-1.411	-1	0	1	1.414	
1	**Air content in pyrolysis gas, %**	X_1	50	57.5	65	72.5	80	7.5
2	**Air temperature, ° C**	X_2	140	200	260	320	380	60
3	**Particles moister content, %**	X_3	6	10	20	30	36	10
4	**Pressure drop in the chamber, MPa**	X_4	0.06	0.1	0.2	0.3	0.36	0.1

For developing the process model, orthogonal central composition plan of the second order was used. After elimination of the factors and interactions, which coefficients had lesser module meanings of set thresholds of significance for general level of significance $\sim = 0.5$, following dependencies were obtained:

Hydrogen: $Y_1 = 4.124 - 0.3322X_1 + 0.1X_2 - 0.3157X_3 - 0.1415X_4 - 1.0483X_1^2 - 0.7983X_2^2 -$

$$- 0.2985X_3^2 - 0.7983X_4^2 - 0.5563X_1X_2 - 0.98125X_1X_3 - 0.09377X_1X_4 - 0.1071X_2X_3; \quad (1)$$

Methane: $Y_2 = 3.4584 - 0.1319X_1 + 0.0634X_2 - 0.2561X_3 - 0.5382X_4 - 0.764X_1^2 - 0.514X_2^2 -$

$$- 0.639X_3^2 - 0.514X_4^2 + 0.1251X_1X_2 - 0.606X_1X_3 - 0.0987X_2X_3; \quad (2)$$

Carbon oxide:

$$Y_3 = 4.132 - 0.1574X_1 + 0.0713X_2 - 0.2484X_3 - 0.55969X_4 - 0.8307X_1^2 - 0.581X_2^2 - 0.556X_3^2 -$$
$$- 0.556X_4^2 - 0.1068X_1X_2 - 0.759X_1X_3 - 0.185X_1X_4 - 0.085X_2X_4; \quad (3)$$

Carbon dioxide:

$$Y_4 = 3.6176 + 0.3238X_1 - 1.1171X_2 + 0.2484X_3 + 0.5341X_4 + 0.9044X_1^2 + 0.6544X_2^2 +$$
$$0.7794X_3^2 + 0.7669X_4^2 - 0.0844X_1X_2 - 0.0794X_1X_3 - 0.4144X_1X_4 - 0.906X_2X_3 + 0.122X_3X_4 +$$
$$+ 0.356X_3X_4; \quad (4)$$

Nitrogen: $Y_5 = 0.1997 + 0.0184X_1 - 0.087X_2 + 0.0075X_3 - 0.4082X_4 + 0.56X_3^2 + 0.0441X_4^2 +$

$$+ 0.016X_1X_3 - 0.018X_1X_4 - 0.017X_3X_4; \quad (5)$$

Carbohydrates: $Y_6 = 3.2532 + 0.075X_1 + 0.462X_2 - 0.2996X_3 - 0.331X_4 - 0.6834X_1^2 -$

$$0.581X_2^2 - 0.284X_1X_2 - 0.044X_1X_3 + 0.094X_1X_4 - 0.1025X_2X_3 - 0.0775X_3X_4; \quad (6)$$

Hydrogen sulfide: $Y_7 = 0.4488 + 0.0186X_1 - 0.0071X_2 - 0.0708X_4 + 0.0983X_2^2 + 0.129X_4^2 -$

$$- 0.075X_1X_4 - 0.057X_2X_4 - 0.0365X_3X_4 \quad (7)$$

It was proved that proposed models are adequate with confidence level meaning of 0.95 (fig.3).

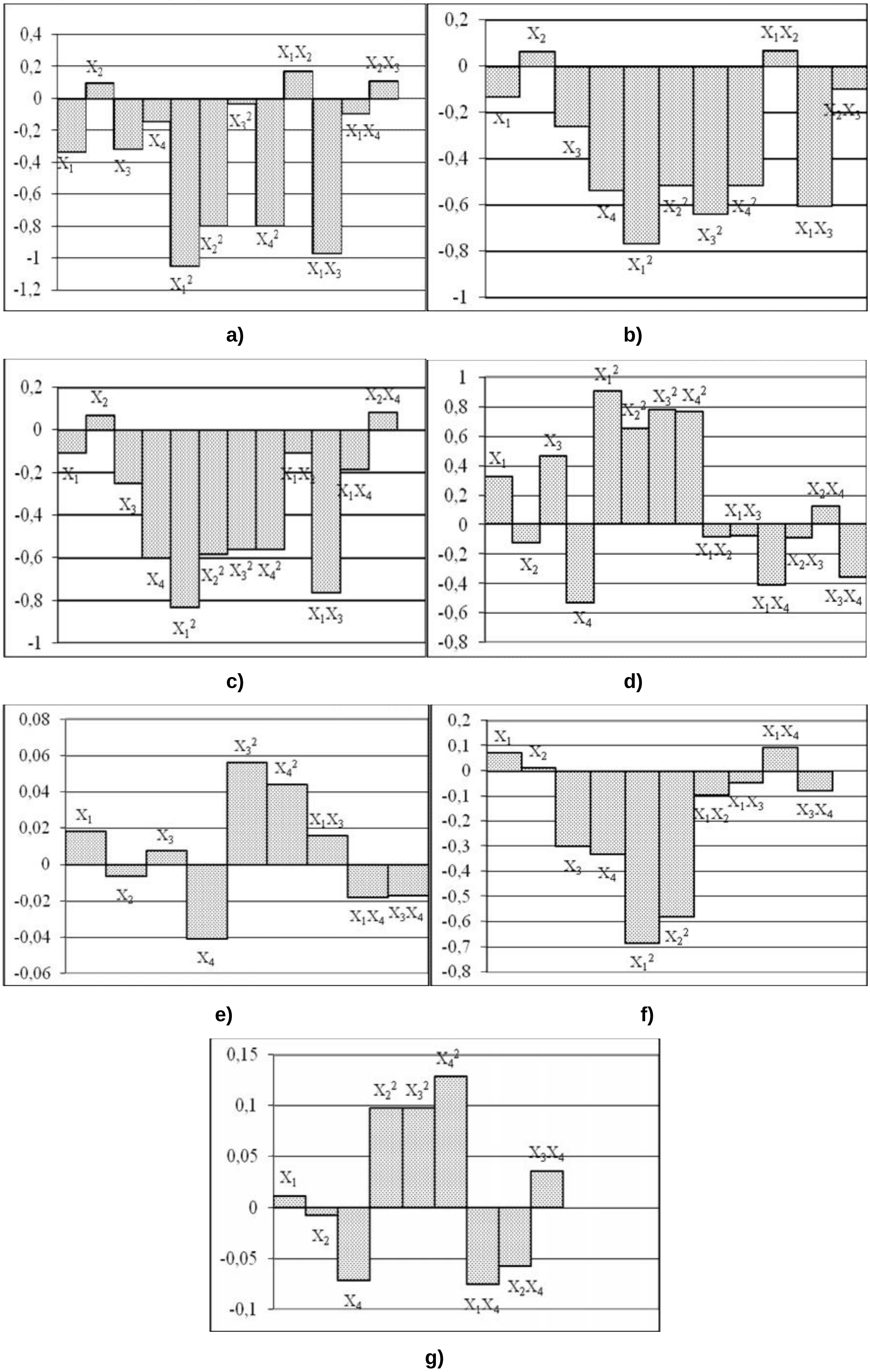

Fig.3 - Diagrams representing levels of significance of model factors

a)hydrogen; b) methan; c) carbon oxide; d) carbon dioxide; e) nitrogen; f) heavy carbohydrates; g) hydrogen sulfide

It can be used for adjustment of the pyrolysis process and output prognosis for pilot large scale processes. When the temperature of pyrogas/air mixture is increased, range of conditions when ignition can occur are wider, thus pressure and temperature interaction effects are more complicated. Pressure increase (X_4) for hydrogen (Y_1) narrows down the range where ignition can occur, although for methane and other components of gas mixture (Y_3, Y_5, Y_6, Y_7) the range becomes wider. Thus, an obtained mathematical model proves result obtained in practice i.e. by regulating pressure pulsing, gas composition and temperature at which ignition occurs can be controlled.

Biomass decomposition process with generation of volatile substances, such as hydrogen (Y_1), methane (Y_2) and partly heavy carbohydrates (Y_6) plays the key role in pyrolysis process. As far other factors (X_1, X_3, X_4) besides temperature have influence on yield of volatile substances, it is obvious that all stated factors (X_1, X_2, X_3, X_4) should be considered to control mass transfer which is reflected inbuilt regression equations. Regression dependencies for all given factors appeared to be adequate physical processes, thus can be sued for pyrolysis process control and optimization. The content of gases with highest heating value in the pyrolysis gas was determined as main response function for optimization of the energy yield of the process. The amount of hydrogen Y_1 in the pyrolysis gas was optimized according to obtained optimization model, while numeric limitations were accepted after summarizing of average meanings with variance intervals.

Considering accepted assumptions the function optimization equations were following:

Hydrogen: $$L_1 = Y_1 + \text{a}_1(Y_3 + X_5 + 20.8) + \text{a}_2(Y_4 + X_6 - 19.2) + \text{a}_3(Y_5 + X_7 + 15.54); \quad (8)$$

Methane: $$L_2 = Y_2 + \text{a}_1(Y_3 + X_5 + 20.8) + \text{a}_2(Y_4 + X_6 - 19.2) + \text{a}_3(Y_5 + X_7 + 15.54); \quad (9)$$

Heavy carbohydrates:
$$L_3 = Y_6 + \text{a}_1(Y_3 + X_5 + 20.8) + \text{a}_2(Y_4 + X_6 - 19.2) + \text{a}_3(Y_5 + X_7 + 15.54). \quad (10)$$

To determine optimal $X_к$ meanings, three systems of equations were resolved. After resolution of given equation system, the stationary point was found where meanings were: (X_1=10.44%, X_2=396ºC, X_3=3.35%, X_4=0.153). As it can be observed in given equations, it is hard to define $У_1$ from $У_2$, $У_3$, $У_4$, $У_5$, $У_6$, $У_7$ thus regression was used to evaluate $У_1$ in the same conditions. Multi collinearity was checked in Farrar - Glauber method for all three equation systems (*Farrar and Glauber, 1967*). Proposed approach and found mathematical solutions allow controlling mass transfer during pyrolysis process by variation of meaning for (X_1, X_2, X_3, X_4) which can be done for process of any scale. Obtained results gave all sufficient data for process optimization and further up scaling. During the pilot testing of the pyrolysis unit, different types of feedstock (rice and buckwheat husks, sunflower husks and sawdust) with similar physical and chemical properties were used, which gave similar composition of pyrolysis products with not big differences. The pilot scale study was dedicated to establishing of dependencies of the process parameters on the composition of pyrolysis products and finding of the optimal process intensity to obtain certain gaseous products composition. Pilot unit for thermal conversion of biomass into fuel gas was designed to conduct large scale testing and incorporated into boiler house of municipal enterprise. The proposed technology was based on two stage process. The first stage feedstock undergoes thermal decomposition which results in gas production. This gas is burned on the second stage as depicted on the general process flow scheme (fig.4).

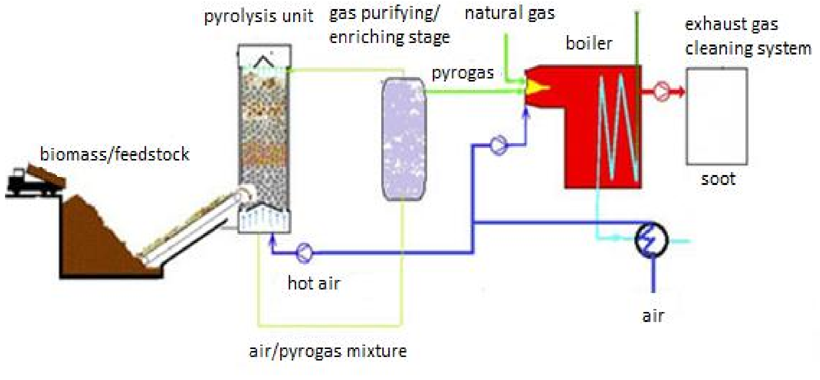

Fig.4 - General process flow diagram

Pyrolysis gas that was obtained in the process used as fuel for water boiling system.

CONCLUSIONS

Agricultural residue/wastes are promising for producing bioenergy, despite the existing considerations, such as spatial distribution, production costs, and an unstable supply. Availability of the feedstock and regional concentration are good preconditions for local bioenergy generation. However, there is a lack of technologies able to support optimal production limits. Considering both positive and negative impacts of various bioenergy technologies and feedstock on social economics and ecological challenges, utilization of existing feedstock sources may be the most effective method to develop sustainable, renewable alternative fuel. Experimental methods used and results obtained in this study would be practical to address the challenges in biomass gasification process optimization and adaptation for large-scale implementation.

ACKNOWLEDGEMENT

The work has been funded by the Ukrainian Ministry of Education and Science.

REFERENCES

[1] Bielski S., (2015), The agricultural production of biomass for energy purposes in Poland. *Agriculture & Forestry*, Vol.61, Issue 1, pp.153-160;

[2] Brewer C. E., Schmidt-Rohr K., Satrio J. A. and Brown R. C., (2009), Characterization of biochar from fast pyrolysis and gasification systems, *Environ. Prog. Sustain. Energy*, 28, pp.386–396;

[3] Czernik S., Bridgwater A.V., (2004), Overview of application of biomass fast pyrolysis oil. *Energy & Fuels,* vol.18, p.590;

[4] Farrar D.E., Glauber R.R., (1967), Multi-disciplinarity in regression analysis: The problems revisited, *The review of Economics and statistics*, Vol.49, No.1, pp.92-107;

[5] Gaskin J.W., Steiner C., Harris K. C., Das C. and Bibens B., (2008), Effect of low-temperature pyrolysis conditions on biochar for agricultural use, *Transactions of the ASABE*, vol.51, pp.2061–2069;

[6] Geletukha G., Zhelyezna T., Lakyda D., Vasylyshyn R., Zibtsev S., Lakyda I., Böttcher, H., (2010). Potential of biomass for energy in Ukraine, Kyiv, p.27, http:// www.eu-bee.info

[7] Jindo K., Mizumoto H., Sawada Y., Sanchez-Monedero M. A., Sonoki T., (2014), Physical and chemical characterization of biochars derived from different agricultural residues, *Biogeosciences,* no.11, pp.6613–6621;

[8] Lau F.S., Zabransky R., Bowen D.A., (2002), Techno-Economic Analysis of Hydrogen Production by Gasification of Biomass Gas, *Hydrogen Program Review Proceeding,* p.12;

[9] Lei O., Zhang R., (2013), Effects of biochars derived from different feedstock and pyrolysis temperatures on soil physical and hydraulic properties, *J. Soil*, Sedim, no.13, pp.1561–1572;

[10] McBeath A.V., Smernik R.J., Krull E.S., Lehmann J., (2013), The influence of feedstock and production temperature on biochar carbon chemistry: A solid-state 13C NMR study, *Biomass Bionenergy*, no.60, pp.121–129;

[11] Mimmo T., Panzacchi P., Baratieri M., Davies C.A., Tonon G., (2014), Effect of pyrolysis temperature on miscanthus (Miscanthus x giganteus) biochar physical, chemical and functional properties, *Biomass Bionenergy*., no.62, pp.149–157;

[12] Suri A., Horio M., (2010), Solid biomass combustion, in: Handbook of Combustion, vol.4, Solid Fuels, edited by: Lackner M., Winter F., Agarwal, A. K., WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim, chapter 3, pp.85–140;

[13] Tilmann D., (2000), The combustion of solid fuels. Academic Press, Boston, p.199;

[14] Velychko O., (2015), Logistical system Fortschrittzahlen in the management of the supply chain of a multi-functional grain cooperative. *Economics and Sociology,* Vol.8, no.1, pp.127-146 Szczecin/ Poland

[15] Wang X., Kersten S.R., Prins W., Van Swaaij W.P.M., (2002), Biomass-syngas from fast pyrolysis vapors of liquids. *Proceedings of the 12th Eurpean Conference on Biomass for Energy, Industry and Climate Protection*, Amsterdam, The Northlands, p.781;

[16] Williams P.T., Besler S. (1996). The influence of temperature and heating rate on the slow pyrolysis of biomass. *Renewable Energy*; 3:233-250.