THE ORGANIC WASTE FRACTIONS RATIO OPTIMIZATION IN THE ANAEROBIC CO-DIGESTION PROCESS FOR THE INCREASE OF BIOGAS YIELD

by

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Biogas obtained by anaerobic digestion process from various organic fractions of waste is increasingly used as a renewable energy sources for the generation of electricity and heat. The quantity of biogas produced by anaerobic digestion depends on many factors: types and characteristics of organic waste, elemental composition of waste, C/N ratio, pH value, inhibitors, retention time, content of nutrients, etc. In addition to the selection of parameters that influence the process of anaerobic digestion, biogas yield can also be influenced by choosing the optimal combination and ratio of organic fractions of waste. In this paper, an analysis of the influential parameters in the process of anaerobic digestion was performed on biogas yields and an overview of the essential characteristics of waste (elementary composition, C/N ratio, lignin content, etc.) for different fractions of organic waste (organic municipal waste, various types of waste of animal origin, as well as agricultural waste). In order to choose the optimal mixing ratio of different fractions of organic waste for maximum biogas yield, a mathematical model has been developed using the multi-criteria optimization method. The boundary conditions set for the multi-criteria optimization was the C/N ratio in the range of 20 to 30 and the minimum content of the lignin in the substrate. The application of the developed model was carried out on the case study of the city of Nis, and the optimal mix of different types of organic waste was determined, as well as the optimal amount of each waste fraction and biogas yield.

Key words: *anaerobic digestion, biogas yield, multi-criteria optimization, organic waste fraction*

Introduction

The anaerobic digestion (AD) process is a mechanism for biogas production whereby the organic matter, *i. e.* feedstock, is decomposed under the influence of bacteria into a gas mixture, whose highest percentage is made up of CH_4 and CO_2 in the conditions without oxygen. The AD can be used to treat the organic fraction of solid municipal waste (OFMSW), as well as other types of organic waste (OW) with high moisture content. The advantage of this waste treatment is the production of biogas that can be used to obtain energy, as well as compost that can be used as an organic fertilizer. When the elemental composition of waste treated in AD is not favorable for obtaining a sufficient amount of biogas, the anaerobic co-

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digestion is applied, whereby the mixture of several different types of OW is used to achieve the optimal composition of the substrate. Anaerobic co-digestion of agricultural waste with manure is best way to dispose waste considering especially the process stability, secondary contaminant generation (organic fertilizer), level of safety risk for workers, and communities and noise levels near installation during operation [1]. The AD is a complex multiphase process with many influential parameters that define its direction and flow: environmental (*e. g.*, temperature, pH, *C/N* ratio, retention time), process by-products (*e. g.*, volatile fatty acid and ammonia nitrogen), and physical and chemical properties of the feedstock, *e. g.*, volatile solids (VS), nutrient content, complex chemical structure [2]. Some of them can be controlled and thus managed by biogas production. Factors that have significant influence on AD are elemental composition pointed through *C/N* ratio, pH-value, organic loading rate and hydraulic retention time.

Elemental composition of the substrate in the digester should contain an appropriate level of carbon, oxygen, hydrogen, nitrogen, potassium, calcium, and magnesium. All OW consists of three basic types of organic compounds: proteins, lipids and carbohydrates. When these organic compounds are submitted to the AD process, each of them dissolves differently. Decomposition of organic compounds by AD process is shown in fig. 1. Carbohydrates dissolve more easily than proteins or lipids, therefore larger amounts are desirable in substrate. In first phase of AD process (hydrolysis) they are dissolved onto simple building components: sugars and alcohols. In the acid phase (acidogenesis and acetogenesis) sugars and alcohols are decomposed into volatile fatty acids, and then into acetic acid. In the last phase of AD process (methanogenesis) acetic acid is decomposed producing CH_4 and CO_2. Proteins breakdown very similar to carbohydrates. Only difference is that proteins in hydrolysis dissolve onto amino acids, followed by their decomposition into volatile fatty acids in acid phase. Lipids decompose differently from other organic compounds: in hydrolysis they dissolve into glycerin and long chain fatty acids. Glycerin is than decomposed in the same way as proteins and carbohydrates, while long chain fatty acids are difficult to dissolve by acid bacteria due to their complex chemical structure. There are some compounds as lignin (mostly found in cellulose rich plants) which is difficult to be decomposed by anaerobic bacteria [3]. Lignin can be found in substrate of plant origin such as: straw, silage, yard waste, *etc.* Two studies have compared the CH_4 yields of various unprocessed lignocellulose substrates on the basis of composition. These studies found that lignin rich substrates yielded less biogas than substrates

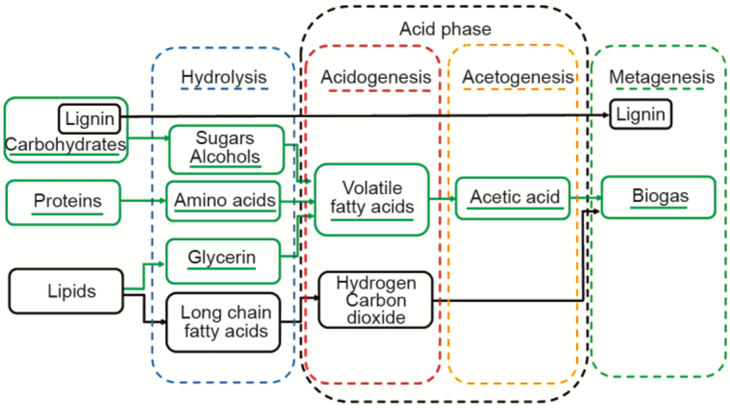

Figure 1. Mechanism of organic compounds decomposition in AD process

containing lower concentrations of lignin [4, 5]. Carbon and nitrogen content is of essential significance for the AD process, carbon is crucial for securing energy for the life and growth of microorganisms, and the presence of nitrogen is important because of the construction of proteins that are building blocks of microorganisms. Experimental investigations have shown that AD conditions can be followed by a *C/N* ratio, with the best conditions being achieved when this ratio is within the range from 20/1 to 30/1. If this ratio is higher – there is a decrease in biogas production, and if lower, there is a rise in ammonia in the digester, which has a bad effect on methanogenic bacteria [6]. Substrate made as a mixture of various fractions of organic waste could improve the *C/N* ratio and enhance the activity of methanogens [7].

The *pH-value* of the AD process depends on the phase of AD: hydrolysis, acid phase, and methanogenesis. Bacterial cultures in the first two phases of the process are more resistant to changes in pH than methanogenic bacteria. The acid synthesis in the acid phase (acidogenesis and acetogenesis) reduces the pH during the process. Methanogenic bacteria exist in pH neutral or slightly alkaline environments. Once the process has stabilized, it is the best to maintain the pH between values 7 and 8 [8].

Organic loading rate is the quantity of organic material added per unit volume of the AD reactor in a day. The mass of organic material can be expressed as volatile solids [9]. Methane yield dependents on the organic loading rate. For maximum biogas yield, the organic loading rate should be kept at maximum, but at the greater concentration, may inhibit process of AD because of the accumulation of volatile fatty acids that may cause a decrease of pH within the digester [9].

Hydraulic retention time is an average time of the substrate remained inside the digester. Decrease in the hydraulic retention time, can cause washout of the active bacterial population. On the contrary, the increase in the hydraulic retention time increases the capital cost of the reactor [10].

Different researches were done in order to determine the basic relation of mixing different organic fractions and optimize the process parameters. An experimental determination of the optimal ratio of mixing organic fractions was made in India when poultry dropping and lignocellulose-rich OW were used as a co-substrate. Different mixing ratios were tested for biogas yield and *C/N* ratio to determine the optimum mixing ratio. When poultry droppings were mixed with lignocellulose rich substrates, it was concluded that optimum mixture is 70% of poultry droppings and 30% wheat straw for the unhindered AD process [11]. Also, four experiments were done in order to choosing the best ratio between OW of MSW and animal manure and comparing the results in either case: the OW of MSW with poultry waste, the OW of MSW with cow dung, the OW of MSW with cow dung and poultry waste, and only OFMSW [12]. Further, optimization of process parameters affecting biogas production from OFMSW *via* AD was done in order to obtain the optimal conditions for biogas production. The parameters studied were initial pH, substrate concentration and total organic carbon (TOC) [13]. The experimental results showed the substrate concentration and TOC had significant individual effect on biogas yield as high as was 53.4 l/kg VS at optimum pH, substrate concentration and TOC of 6.5, 99 gTS/L, and 20.32 g/L [13]. Recent study was conducted on a full-scale model, focused on the prediction and optimization of biogas yield from a mixture of cow manure and maize straw. For the development of that model an adaptive neuro-fuzzy interference system was used, and as constraints of the model total solid content, *C/N* ratio and stirring intensity were used. As the result of this research, biogas production increased about 8% under the optimal conditions suggested by the proposed model [14]. Further research conducted in Nigeria [15] focused on achieving maximum biogas yield using as main criteria digester geometry and the co-substrates (poultry droppings, cow and pig manure, and different crop residuals). The optimization of the following problem was done by artificial neural network integrated with a genetic algorithm. The results showed that a cylindrical-shaped digester was the best for biogas production, while cow manure gave the highest biogas yield. The study demonstrated that maximum biogas production could be obtained using a combination of animal wastes supplemented with plantain peels [8]. Similar research was done in 2017 [16] in Central Chinese Economic District with an aim to optimize ratio of agricultural waste and duck manure in AcoD process and determine the optimum temperature for AcoD. Results of this study indicated that the best performance of AcoD is at 30 °C, and the best ratio to mix duck manure with straw is 2,8 to 1 based on their dry mass [16].

In this paper, a mathematical model has been developed in order to choose the optimal mixing ratio of different fractions of OW for maximum biogas yield, using the multi-criteria optimization method. The boundary conditions set for multi-criteria optimization were the *C/N* ratio in the range of 20 to 30 and the minimum content of the lignin in the substrate. The application of the developed model was carried out on the case study of the city of Nis.

Model for optimization of OW fractions ratio for the maximum biogas yield

For the development of the model for optimization of OW fractions ratio for the maximum biogas yield, the simplex method of multi-criteria optimization was used. Elemental waste composition, specifically *C* and *N* content, *C/N* ratio and lignin (L) content were used as input parameters when developing the model. Main problem, when this model was developed, was a decision making and planning problem which involves multiple conflicting objectives that should be considered simultaneously. Since the increase in *C* in the elemental composition of the organic fractions increases and the content of lignin these two constrains are colliding. The method of global criterion is applied, where the distance between some desirable reference point in the objective space and the feasible objective region is minimized [17, 18]. In the developed model, the problem was reduced to three functions of *n* variables, eqs. (1)-(3):

$$C(x)=\sum_{i=1}^{n} C_i x_i \tag{1}$$

$$N(x)=\sum_{i=1}^{n} N_i x_i \tag{2}$$

$$L(x)=\sum_{i=1}^{n} L_i x_i \tag{3}$$

where C [%] is the carbon content in specific OW fraction, N [%] – the nitrogen content in specific OW fraction, L [%] – the lignin content in specific OW fraction, $x = (x_1, x_2, \dots x_n)$ – the OW fractions combination, and n – the number of OW fractions.

The constraints are $x_1 + x_2 + \dots + x_n = 1$, and $x_i \geq 0$, $\forall i$.

For normal activity and reproduction of CH_4 bacteria in AD process, a necessary condition is the favorable *C/N* ratio in digesting matter. It is considered a favorable *C/N* ratio from 20 to 30. The best deal is 30:1. This relationship is needed because CH_4 bacteria are consuming about 30 times more carbon than nitrogen [6]. The additional condition

$20 \le C(x)/N(x)$ can be written in the form $20N(x) \le C(x) \le 30N(x)$, wherefrom two constraints could be recognized, eqs. (4) and (5):

$$20N(x) - C(x) \ge 0 \tag{4}$$

$$30N(x) - C(x) \ge 0 \tag{5}$$

From previously mentioned constraints, the feasible region could be defined.

The main problem is a multi-criteria optimization is presented in eq. (6):

$$\begin{Bmatrix} \max C(x) & \min L(x) \\ x \in D & x \in D \end{Bmatrix} \tag{6}$$

To solve the multi-criteria problem, the global criterion should be considered:

$$G(x) = \left[\frac{C^* - C(x)}{C^*} \right]^2 + \left[\frac{L^* - L(x)}{L^*} \right]^2 \tag{7}$$

By solving the multi-criteria problem presented by eq. (6) using the simplex method, the Pareto optimum point is obtained, that is, the optimal fraction of the OW fractions in order to obtain the maximum yield of biogas in the AD process.

Experimental research

The presented model for the optimization of OW fractions ratio for the maximum biogas yield is applied in the case of the city of Nis. The elemental composition of selected organic fractions is based on literature data [11, 19], while the organic part of solid municipal waste is derived from data of Public Utility Company *Mediana*, Nis [20].

Study area

City of Nis situated in the south-eastern part of the Republic of Serbia occupies an area of 596.71 km^2 [21] and has 260.237 inhabitants, according to data from the last census of 2011 [22].

Municipal waste generated in the city of Nis is collected and transported to the city landfill once a week. Waste is collected from 89452 households from an area of 6770885 m^2. The amount of waste collected as solid municipal waste in the city of Nis in 2014 is 65348 tons per year, from which OFMSW is 27.32%, or 17866 tons per year [20]. In tab. 1 and fig. 2 types and generated amount of organic matter annually in the Nis county are shown. On the territory of the Nis county there are 31283 agricultural farms and 82729 ha of arable land is used. The number of households engaged in cattle breeding is 23951, and the number of livestock unit is 55810. The number of cattle in the territory of Nis county is 24850, while the number of pigs is 92856 [23]. When considering the position of the city, the following selected fractions for mixing with OFMSW can most often be found in its surroundings: pig manure, cow manure, wheat straw, and meadow grass.

The optimization of OW fraction

In selecting the fractions to be used in the process of AD, in the case study, the main criterion for selecting raw materials is the availability in the Nis county. The following four fractions of organic agricultural waste and animal origin, such as pig manure, cow manure, wheat straw, and meadow grass, have been selected on the basis of the previously stated facts.

Table 1. Types and generated amount of organic matter on the territory of the Nis county [14, 20, 22]

Type of organic matter			
Animal excrements	Head of cattle	Amount of animal excrements per head of cattle, [m^3 per year]	Generated amount of excrements, [m^3 per year]
Cows	24.850	20	498.739
Pigs	92.856	1.46	135.569
Biomass	Surface [ha]	Amount of biomass [m^3/ha]	Generated amount of biomass [m^3 per year]
Meadow grass	43.882	10	438.820
Wheat straw	19.615	4-6	78.460-117.960
Municipal waste	Number of residents	Amount of OFMSW [m^3 per resident]	Generated amount of OFMSW [m^3 per year]
OFMSW	260.237	0.06865	17.866

The fifth fraction of OW used in AD is OFMSW. Table 2 shows the elemental composition of five fractions that are covered by the multi-criteria optimization method in order to determine the maximum yield of biogas.

Table 2. Basic characteristic of substrates used in co-digestion [11, 14, 19, 20]

	Pig manure	Cow manure	Wheat straw	Meadow grass	OFMSW
C [%]	39.14	33.61	42.0	41.8	47.9
N [%]	3.92	8.95	0.453	0.733	3.1
C/N	10	3.8	92.72	57.03	15.45
L [%]	1.83	1.7	6.53	5.51	4.59
Biogas yield [$m^3kg^{-1}VS^{-1}$]	0.25-0.50	0.20-030	0.35-0.45	0.56	0.65

Using eqs. (1)-(3), since five fractions of OW have been selected whose composition should be optimized to give the highest biogas yield in the AD process, the problem can be reduced to three functions of five variables:

$$C(x) = 39.14x_1 + 33.61x_2 + 42x_3 + 41.8x_4 + 47.9x_5$$
$$N(x) = 3.9x_1 + 8.95x_2 + 0.435x_3 + 0.733x_4 + 3.1x_5$$
$$L(x) = 1.83x_1 + 1.7x_2 + 6.53x_3 + 5.51x_4 + 4.59x_5$$

where $x = (x_1, x_2, x_3, x_4, x_5)$.

Applying the previously described model constraints in eqs. (4) and (5), we get the feasible region:

$$D:\begin{cases} x_1 + x_2 + x_3 + x_4 + x_5 = 1 \\ -39.26x_1 - 145.29x_2 + 33x_3 + 27.2x_4 - 14.1x_5 \ge 0 \\ 78.46x_1 + 234.89x_2 - 28.5x_3 - 19.9x_4 + 451.x_5 \ge 0 \end{cases}$$

Using the mentioned optimization method (the simplex method) the solutions of particular problems had been found:

$$\max_{x \in D} C(x) = 46.1338 = C(0,\ 0,\ 0.2994,\ 0,\ 0.7006)$$

$$\min_{x \in D} L(x) = 4.00389 = L(0.4093,\ 0,\ 0,\ 0.5907,\ 0)$$

The solution of particular multi-criteria problem, defined with eq. (7) on the same feasible region is:

$$\min_{x \in D} G(x) = G(0.3245,\ 0.000006,\ 0.000257055,\ 0.539097,\ 0.136374) = 0.01112$$

is the Pareto optimum point.

Finally, the solution of our main problem is the point:

$$x^* = 0.3245, 0.000006\ 0.000257055, 0.539097, 0.136374$$

in which:

$$C^* = C(x^*) = 41.7687, \quad L^* = L(x^*) = 4.1903$$

and the additional condition is satisfied:

$$\frac{C(x)}{N(x)} = 20.0003 \in [20, 30]$$

Results and discussion

When determining the ratio of OW fractions for maximum biogas yield, two separate cases were considered at the outcome: the maximum content of carbon and the minimum content of lignin in the elemental composition of the final substrate which was obtained by mixing five fractions taken into account in the case study of the Nis county. In order for *C/N* to fulfill the condition 20 to 30, it is necessary that the carbon content, as a numerator in this fraction, tends to be the maximum. For this reason, the maximum content of carbon in the elementary composition of the substrate is determined. By maximizing the content of carbon in the elemental composition of the final substrate, the carbon content is 46.13%, while the optimal mixing composition is 0% of pig manure, 0% of cow manure, 29.94% of wheat straw, 0% of meadow grass, and 70.06% of the OFMSW. By minimizing the content of lignin in the elemental composition of the final substrate, the lignin share is 4.00389%, while the optimal mixing composition is 49.03% of pig manure, 0% of cow manure, 0% of wheat straw, 59.07% of meadow grass, and 0% of the OFMSW. In order to achieve maximum carbon content, the optimal mix would only include the OFMSW and wheat straw, while pig manure and meadow grass should be mixed to achieve the minimum lignin content.

It is necessary to determine the optimum ratio of mixing of organic fractions of waste so that both conditions are met, the model previously described is used. The optimal ratio provided by the model is shown in tab. 3. When the OW fractions are mixed in this proportion, the *C/N* ratio is 20,0003, the amount of lignin is 4.1903%, and both constraints are satisfied.

Table 3. Optimal co-substrate ratio in co-digestion

	Pig manure	Cow manure	Wheat straw	Meadow grass	OFMSW
Ratio [%]	32.45	0.0006	0.025	53.90	13.63
Amount [m^3]	42,510	0.786	32.7	70,651	17,866

Given that, the sum of the share of pig manure, meadow grass and OFMSW amounts to 99.98% of the total share in the substrate, cow manure and wheat straw can be excluded when mixing OW fractions. By comparing the elemental composition of pig and cow manure (tab. 1), as well as by comparing the elemental composition of meadow grass and wheat straw, it is noted that it is very similar. It can, therefore, be concluded that cow manure can be used as alternatives to pig manure in the event of a lack thereof, while wheat straw can be used as alternatives to meadow grass.

The developed mathematical model was applied to the quantities of organic matter available in the territory of the Nis county, and primarily with the aim of treating the total generated amount of OFMSW annualy. The used mixing ratio is shown in fig. 2. In order to treat 17.866 m^3 of OW generated in the territory of the city of Nis by process of AD, it is necessary to use 70782 m^3 of wheat straw (90.21% of the produced quantity per year), *i. e.* 16.13% of meadow grass produced annually. For optimal mixing of raw materials, it is also necessary to use animal excrements, ie 42600 m^3 per year, which is 31.42% production of pig excrements, or 8.54% production of cow excrements.

By comparing the biogas yield of each substrate and final fubstate used in AD process it can be concluded that the yield of biogas of the final mixture is satisfactory amounts 0.457 m^3/kg VS, as it is shown in fig. 3. On the territory of the Nis county, by using the developted mathematical model, it is possible to generate 4798426 m^3 of biogas annually.

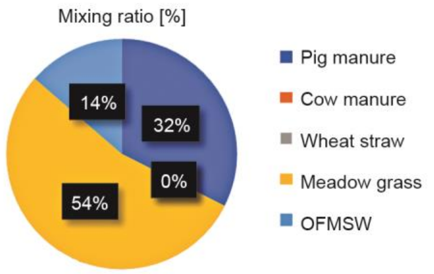

Figure 2. Optimal mixing ratio used in the application of the developed model
(*for color image see journal web site*)

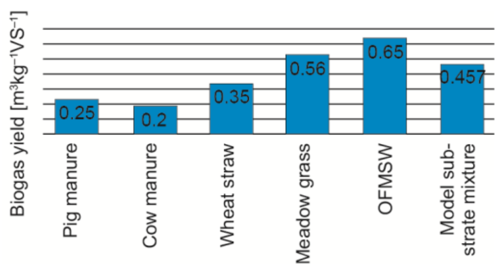

Figure 3. Comparison of biogas yield of individual substrates and optimal mixture provided by the developed model

Conclusions

The biogas yield in the AD process depends on many factors: types and characteristics of waste, *C/N* ratio, pH values, inhibitors, retention times, nutrients content, *etc.* With the optimal mixing ratio of organic fractions of treated waste *C/N* ratio can influenced, and therefore a mathematical model has been developed using the multi-criteria optimization method. The boundary conditions set in the optimization were the *C/N* ratio in the range of 20 to 30 and the minimum content of the lignin in the substrate.

Developed model was applied on the case study of the Nis county. Five organic fractions of waste were taken into account: pig manure, cow manure, meadow grass, wheat straw, and OFMSW collected on the territory of the Nis county. By the multi-criteria optimization method, an optimal mix of these five fractions is obtained to get the highest biogas yield.

With an optimum ratio of 32.45% of pig manure, 0.0006% of cow manure, 0.025% of wheat straw, 53.90% meadow grass, 13.63% of OFMSW, *C/N* ratio is 20.0003, and lignin content 4.1903%. By mixing OW fractions is in this ratio, the *C/N* ratio condition is satisfied in the range between 20 and 30, and the minimum lignin content requirement was achieved. Considering the very small amount of cow manure and wheat straw, it is possible to exclude them from the optimal mixing ratio of organic fractions, as the sum of their shares is 0.02%. In the case of neglecting these two fractions in the optimum mixing ratio of OW fractions, the optimum ratio for obtaining the highest yield of biogas is 32% of pork manure, 54% of meadow grass, 14% of the OFMSW, where initial restraints remain satisfactory. When developed model was applied, with an aim to treat all amount of OFMSW, it was concluded that there is sufficient amount of all OW fraction and that there is satisfactory biogas yield for the selected final mixture.

The obtained results show that the presented model based on the multi-criteria optimization method for determining the optimal mixing ratio of OW fractions for the maximum yield of biogas can be used for mathematical optimization of any OW fractions as well as for different boundary conditions.

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