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Modeling Carbon-to-Nitrogen Ratio Influence on Biogas Production by the 4th-order Runge–Kutta Method

Latif Fagbemi,[†] David Adamon,[‡] and Evrard Karol Ekouedjen*[,†]

[†]Ecole Polytechnique d'Abomey-Calavi, Université d'Abomey-Calavi, Cotonou 01BP2009, Benin
[‡]Université Nationale des Sciences Technologies Ingénierie et Mathématiques, Abomey 01BP2009, Benin

ABSTRACT: Several parameters influence anaerobic digestion, e.g., hydraulic retention time, temperature, pH, pressure, C/N ratio, etc. This work presents the influence of the C/N ratio of the substrate on biogas production modeling. To do this, a mathematical model is developed for predicting the molar composition of biogas as a function of the C/N ratio. Inspired by the AM2HN model, this model is a system of nine partial differential equations that have been solved numerically in SCILAB software by the 4th-order Runge–Kutta method. Obtained results show an increase in production of methane with the C/N ratio. Thus, optimal production of methane is observed for a C/N value between 20 and 30. The proposed model is then tested with two other models; these tests show that the results of the proposed model match well with numerical results of those two models.

1. INTRODUCTION

Anaerobic digestion is the process whereby organic matter is degraded to give a gas mixture consisting mainly of methane and carbon dioxide called biogas. Several parameters such as temperature, pH, substrate carbon-to-nitrogen ratio, hydraulic retention time, etc. influence biogas production. One of the important parameters influencing the anaerobic digestion process is the carbon-to-nitrogen ratio of the substrate (C/N). Several previous experimental studies reported in refs 1–5 indicate that for optimal biomethane production, this ratio should be in the range between 20 and 35. However, few studies dealt with modeling of the influence of this parameter on biogas production, without experimental studies conducted.[6,3] Only experimental studies were carried out, and this implies time squandering. It is better to find numerically optimal conditions before experimentation. In this context, several models of description of anaerobic digestion have been developed. These include the model of Simeonov et al.[4] developed in 1996 to describe the dynamic behavior of anaerobic digestion of animal dung under mesophilic conditions. The first comprehensive model developed to describe anaerobic digestion as a whole is the anaerobic digestion model (ADM1). A first version of this model was proposed by Batstone et al. in 2002[5] under the aegis of the International Water Association Task group. It is the most complete phenomenological model that takes into account all stages of anaerobic digestion. It is a complex model of 32 differential equations modeling reactions and exchanges between different liquid and gaseous species. To control and optimize anaerobic digesters, a much more appropriate 2-step (AM2) model is used. It is a model with two fundamental stages: acidogenesis and methanogenesis. AM2 includes six differential equations, which were developed in the framework of the European project AMOCO by Bernard et al. in 2002.[6] The aim was to reduce or simplify the ADM1 model and to find a model dedicated to the control of anaerobic digesters. There were many other models inspired by the two previous ones, in particular the AM2B model proposed in 2013 by Benyahia et al.,[7] which is a model adapted to membrane bioreactors; it is inspired by AM2. The goal was to model anaerobic digestion in membrane bioreactors. However, when dealing with complex substrates, the hydrolysis step is no longer negligible. Thus, the AM2HN model of Hassam et al. was launched in 2015.[8] This model adds the hydrolysis step to the AM2 model and takes into account the ammonia nitrogen. It comprises three stages, which are as follows: hydrolysis, acidogenesis, and methanogenesis. More recently, in 2017, an anaerobic membrane bioreactor (AnMBR) model was developed[9] based on the combination of a simple fouling model and the anaerobic model 2b (AM2b) to describe biological and membrane dynamic responses in an AnMBR. In 2018, a study of Khedim et al.[10] showed that the control parameters can greatly affect biogas yield and, thus, process performance.

The aim of this work is to find a numerical model that could allow us to predict the best substrate C/N ratio that leads to optimize biogas production (best methane yield). More precisely, it involves developing a mathematical model to predict the molar composition of biogas as a function of the C/N ratio of the substrate, by using the AM2HN model.

2. MATERIALS AND METHODS

There are several numerical methods for solving first-order differential equation systems such as the Euler method, modified Euler method, Runge–Kutta method, etc. Each of them has advantages and limitations. The simplest one is Euler's method but presents inaccurate results, and Runge–Kutta method appears to be more precise but takes a greater calculation time taking place under mesophilic conditions (25–35 °C) to take into account the climatic realities of the West African region.[11−13] Taking into account Table 1 information, we chose the 4th-order Runge–Kutta method because of

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Table 1. Comparison of Numerical Methods

methods	advantages	disadvantages
Euler	simple and direct	less accurate and numerically unstable
		requires larger computation time
		applicable to explicit differential equations
	improve Euler Method	less accurate
modified Euler	simple	
	easy to implement	requires large computer time
	numerically stable	error estimation is not easy to be done
Runge–Kutta	high precision	

its high precision and numerical stability. The proposed model has three stages, that is, hydrolysis, acidogenesis, and methanogenesis. It is based on the AM2HN model for reactions. The following equations are derived from the AM2HN model.

Hydrolysis

$$S_0 + H_2O \overset{\mu_0}{\rightarrow} k_0S_1 \tag{1}$$

Acidogenesis

$$k_1S_1 \overset{\mu_1}{\rightarrow} X_1 + k_2S_2 + k_4CO_2 \tag{2}$$

Methanogenesis

$$k_3S_2 \overset{\mu_2}{\rightarrow} X_2 + k_5CO_2k_6CH_4 \tag{3}$$

Microbial growth is given by Monod kinetics at the acidogenesis stage and by Haldane kinetics for methanogenesis

$$\mu_1 = \mu_{\max}\frac{S}{K_y + S}\text{Monod} \tag{4}$$

$$\mu_2 = \mu_{\max}\frac{S}{K_y + S + \frac{S^2}{K_I}}\text{Haldane} \tag{5}$$

For $[C/N]S = w$ and $[C/N]S = w$, we can deduce that $[C]S = [N]{*}w$ (a). Taking into account the assumption of constant return, relation 4 becomes 6. Thus, by rewriting relation 4 by introducing the carbon concentration, one obtains

$$\mu = \mu_{\max}\frac{A_C}{K_y + A_C} \tag{6}$$

$$\mu = \mu_{\max}\frac{[C]_S}{K_y + [C]_S} \tag{7}$$

$[C]_S$ in function according to the ratio $[C/N]$ represented by w in relation (a) and introducing it into relation 7. In relation 8, the nitrogen content of the substrate $[N]S$ is assumed to be constant because only carbon is involved in the various reactions of anaerobic digestion. Therefore, in this new relation, w is the variable parameter.

The mathematical model that we propose takes into account three (03) basic steps as indicated above

$$\mu = \mu_{\max}\frac{[N]_S{*}w}{K_y + [N]_S{*}\,w} \tag{8}$$

In this study, hydrolysis is described as first-order kinetics. r_0 is the reaction rate, $r_0 = k_{\text{hyd}}w_0$; k_{hyd} is the hydrolysis constant; and w_0 corresponds to the C/N ratio of the initial complex substrate. The speed of the reaction is $r_1 = \mu_1([N]_Sw_1)$. X_1, such that $\mu_1([N]_Sw_1)$ is the specific growth rate of X_1 on w_1. This specific growth rate is given by Monod kinetics. The mathematical equations of the model mass balance are as follows

Hydrolysis

$$[N]_Sw_0 + H_2O \overset{\mu_0}{\rightarrow} k_0[N]_Sw_1 \tag{9}$$

Acidogenesis

$$k_1[N]_Sw_1 \overset{\mu_1}{\rightarrow} X_1 + k_2[N]_Sw_2 + k_4CO_2 \tag{10}$$

$$\mu_1(w_1) = \mu_{1\max}\frac{[N]_S{*}w_1}{K_{y1} + [N]_S{*}w_1} \tag{11}$$

Methanogenesis

$$k_3[N]_Sw_2 \overset{\mu_2}{\rightarrow} X_2 + k_5CO_2 + k_6CH_4 \tag{12}$$

$$\mu_2(w_2) = \mu_{2\max}\frac{[N]_S{*}w_2}{K_{y2} + [N]_S{*}w_2 + \frac{[[N]_S{*}w_2]^2}{K_I}} \tag{13}$$

$r_2 = \mu_2([N]_Sw_2)$, X_2 is the speed of the reaction,

$\mu_2([N]_Sw_2)$ is the specific growth rate of X_2 on w_2. This specific growth rate is given by Haldane kinetics.

K_{y2} is the half-saturation constant associated with w_2,

$\mu_{2\max}$ is the maximum specific growth rate of X_2 on w_2, and

K_I is the inhibition constant associated with w_2. This constant represents the inhibition of bacterial growth due to the accumulation of volatile fatty acids in the reactor.

K_{y1} is the half-saturation constant associated with w_1. $\mu_{1\max}$ is the maximum specific growth rate of X_1 on w_1.

In this work, we mainly measure the production of biogas as a function of the C/N ratio. This measurement of methane production is done in batch digesters that have been preferred to continuous ones because of available data, which are obtained with batch digesters. In this type of digester, the substrate is loaded once at the beginning of digestion and also at the end of the reaction when the digester only contains the digestate. This implies a lack of continuous and periodic recharging of the reactor; it is deduced that there is no volume flow and therefore the dilution ratio D is equal to 0 ($D = 0$). We obtain systems of eqs 14 and 15

$$\begin{cases} \dfrac{dX_1}{dt} = \mu_1(w_1)X_1 \\ \dfrac{dX_2}{dt} = \mu_2(w_2)X_2 \\ \dfrac{dw_0}{dt} = -k_{\text{hyd}}w_0 \\ \dfrac{dw_1}{dt} = -k_1\mu_1(w_1)X_1 + k_{\text{hyd}}w_0 \\ \dfrac{dw_2}{dt} = k_2\mu_1(w_1)X_1 - k_3\mu_2(w_2)X_2 \\ \dfrac{dZ}{dt} = k_1(N_{w_1} - N_{\text{bac}})\mu_1(w_1)X_1 - N_{\text{bac}}\mu_2(w_2)X_2 + k_{d,1}N_{\text{bac}}X_1 \\ \qquad + k_{d,2}N_{\text{bac}}X_2 \\ \dfrac{dC}{dt} = -q_{CO_2} + k_4\mu_1(w_1)X_1 + k_5\mu_2(w_2)X_2 \\ \dfrac{dCH_4}{dt} = k_6\,\mu_2(w_2)X_2 \\ \dfrac{dCO_2}{dt} = k_5\,\mu_2(w_2)X_2 + k_4\mu_1(w_1)X_1 \end{cases} \tag{14}$$

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Table 2. Kinetic Parameters

parameters	proposed model values	Guérin-Rechdaoui et al. values	Mottelet et al. values	units
$\mu_{1\max}$	0.33	3.4	3.63	(d^{-1})
$\mu_{2\max}$	0.13	1.5	2.67	(d^{-1})
K_{y1}	0.40	41.32	1.02	$(\mathrm{gCOD\ L^{-1}})$
K_{y2}	2.93	3.14	3.45	$(\mathrm{mmol\ L^{-1}})$
K_I	207	207	1.44	$(\mathrm{mmol\ L^{-1}})$
k_{hyd}	05	0.189	0.189	(d^{-1})
k_1	20	2.9	2.9	$(\mathrm{gCOD\ g\ VS^{-1}})$
k_2	464	1.9	0.42	$(\mathrm{mmol\ g\ VS^{-1}})$
k_3	514	1	1	$(\mathrm{mmol\ g\ VS^{-1}})$
k_4	310	45.35	45.35	$(\mathrm{mmol\ g\ VS^{-1}})$
k_5	600	600	600	$(\mathrm{mmol\ g\ VS^{-1}})$
k_6	253	0.88	0.88	$(\mathrm{mmol\ g\ VS^{-1}})$

Table 3. Initial Conditions

parameters	w_0	X_1	w_1	X_2	w_2	Z	C	CH_4	CO_2
proposed model	25	1.42	0	1.12	0	10	50	0	0
Guérin-Rechdaoui et al.	9	1.42	0	1.12	0	10	50	0	0
Mottelet et al.	8	2.5	0	5.1	0	10	50	0	0
units		$\mathrm{kg\ VS\ m^{-3}}$		$\mathrm{kg\ VS\ m^{-3}}$		$\mathrm{mmol\ L^{-1}}$	$\mathrm{mmol\ L^{-1}}$	liter	liter

$$\begin{cases} \dfrac{dX_1}{dt} = f(w_1, X_1, t) \\ \dfrac{dX_2}{dt} = f(w_2, X_2, t) \\ \dfrac{d\omega_0}{dt} = f(w_0, t) \\ \dfrac{d\omega_1}{dt} = f(w_1, w_0, X_1, t) \\ \dfrac{d\omega_2}{dt} = f(w_2, w_1, X_1, X_2, t) \\ \dfrac{dZ}{dt} = f(Z, w_2, w_1, X_1, X_2, t) \\ \dfrac{dC}{dt} = f(C, q_{CO_2}, w_2, w_1, X_1, X_2, t) \\ \dfrac{dCH_4}{dt} = f(w_2, X_2, t) \\ \dfrac{dCO_2}{dt} = f(w_2, w_1, X_1, X_2, t) \end{cases} \tag{15}$$

We thus obtain a system of partial differential equations of first order. We can write the matrix form of this system as follows

$$Y' = f(Y, t)(\text{formula 15})$$

Let us remember that w_i is the [C/N] ratio of substrate i, t is the time, Z is alkalinity (inorganic carbon concentration), q_{CH_4} is the volume flow of methane, q_{CO_2} is the volume flow of carbon dioxide, $k_{d,1}$ and $k_{d,2}$ are, respectively, mortality rates of X_1 and X_2 microorganisms. These rates were arbitrarily set at 10% of the maximum rates of bacterial growth, $\mu_{1\max}$ and $\mu_{2\max}$, in the AM2HN model. The matrix form obtained is numerically solved with the 4th-order Runge−Kutta method. The proposed model has been implemented to simulate anaerobic digestion to see the influence of the C/N ratio on the production of methane. The kinetic parameters used in the model are those of the AM2HN model from which it is inspired. These parameters are presented in Table 2, while the initial conditions are set in Table 3.

3. RESULTS AND DISCUSSION

The model implementation allows us, knowing the initial C/N ratio of the substrate, to predict the production of CH_4 as well as that of CO_2. The analysis of the different results allows us to say that the cumulative production of methane increases as time increases (Figure 1); the same is true for the production of carbon dioxide, with the difference that the latter starts after 2 days of reaction, whereas in the first case, already after a few hours, methane production is observed (Figure 1). Production start does not mean that optimal production is obtained. Cumulative methane production is estimated at 8.84 liters against approximately 0.3 liter of carbon dioxide, i.e. the proportions of 95% of CH_4 and 5% of CO_2 unlike several studies[13,14] that found the CH_4 rate in the range 50−75. This result, which is not typical, may be due to modeling parameters and kinetic parameters. Figure 1 below gives an overview of the cumulative production of methane and carbon dioxide as a function of time for a value of the C/N ratio equal 25.

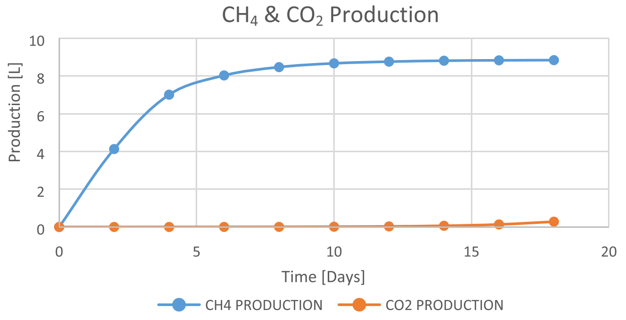

Figure 1. Cumulative production of methane and carbon dioxide given by the proposed model.

There is no experimental study conducted here; this is a numerical approach. To validate the model, two different simulations were performed using data and kinetic parameters of two other models whose inputs and initial and kinetic parameters are summarized in Tables 2−4 by respectively exploiting the data from Guérin-Rechdaoui et al.[15] and those of

Table 4. Experimental Conditions[17]

	proposed model	Guérin-Rechdaoui et al.	Mottelet et al.
substrate type	sludge from the sewer treatment plant (step) of the seine downstream	sludge from the sewer treatment plant (step) of the seine downstream	sludge from the sewer treatment plant (step) of the seine downstream
type of digester	batch	batch	batch
volume of the digester	500 mL	500 mL	500 mL
C/N ratio of the substrate	9	8	8
temperature	mesophilic conditions (35 °C)	mesophilic conditions (35 ± 0.2 °C)	mesophilic conditions (35 ± 0.2 °C)
stages considered	hydrolysis, acidogenesis, and methanogenesis	hydrolysis, acidogenesis, and methanogenesis	hydrolysis, acidogenesis, and methanogenesis

Mottelet et al.[16] The computation of the proposed model gives us a cumulative production of methane of about 1.29 L per g VS biomass, while the experimental value originally obtained is 1200 mL per g VS biomass and that of the model of Guérin-Rechdaoui et al. is 1100 mL per g VS biomass. Table 5 gives us a comparison between cumulative production obtained with the proposed model and cumulative production of Guérin-Rechdaoui et al. The proposed model gives results close to the simulated case, which remains a benchmark in terms of model comparison. However, Guérin-Rechdaoui et al. obtained a gap of 4% between their model and experimental results. This is justified because the parameters of Guérin-Rechdaoui et al. were adjusted by calibration on the experimental results obtained (88 different experiments resulting from the triplicate replication of 30 different substrate/inoculum mixtures). This difference can also be explained by the difficulties related to the experimentation of the theoretical conditions of the model. Figure 2 presents the different cumulative productions of methane as a function of time obtained by the proposed model and by the simulation of Guérin-Rechdaoui et al.

Table 5. Comparison between Cumulative Production Obtained with Model and Cumulative Productions of Guérin-Rechdaoui et al.

	cumulative production of methane mL per g VS
proposed model	1298
Guérin-Rechdaoui et al. experimental result	1200
Guérin-Rechdaoui et al. model's result	1100

The analysis of Figure 2 reveals that the production of methane in the case of the proposed model starts after a few hours (about 3 h) and stabilizes from the 4th day of digestion, while in the Guérin-Rechdaoui model, the proper digestion begins after 4 h and stabilizes from the 4th day. These results, when analyzing Figure 2, are substantially identical to those obtained by Guérin-Rechdaoui et al. and can be explained by many reasons. It is not typical that methane production starts so early, but it may happen depending on the substrate nature and also microbial activities or other parameters (temperature, hydraulic retention time, humidity, pH, pressure, etc.). Here, we used sludge from waste water treatment. It is also important to notice that no experimental studies have been carried and all of the parameters used are taken from previous numerical studies.[17,15,16] In our paper, we just present results as obtained with the model. Otherwise, the proposed model fits well with the numerical results proposed by Guérin-Rechdaoui et al.

The second simulation was performed using data from the experiment and the model of Mottelet et al.[16] Table 4 lists experimental conditions. Initial parameters and kinetics are given in Table 3. The results obtained are very close to those obtained by Mottelet et al.[16] with a relative error estimated to be 1.14%. Indeed, the cumulative production of methane as a function of time obtained by the proposed model is very close to the results obtained by the latter. It is easy to see that methane production increases with time and has patterns similar to those shown in Figure 3. The results obtained are compared in summary Table 6.

The analysis of Figure 3 reveals that methane production begins after a few hours (about 2 h) and stabilizes after 5 days of digestion, while according to tests conducted by Mottelet et al. (Figure 3), it can be seen that methane production starts almost instantaneously and stabilizes after 5 days. This curve presents a first quasi-linear part, which is also the case with the results of the proposed model. This model shows a cumulative production of methane on the order of 1038 mL per g VS, a value close to that obtained by Mottelet et al.: 1050 mL per g VS biomass. To study the effect of the parameter studied (C/N ratio) on the production of methane, a database was created based on the proposed model. The results are shown in Table 7.

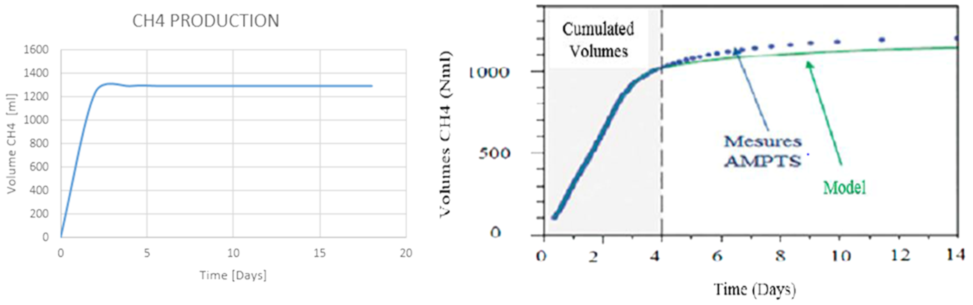

Figure 2. Volume of methane obtained with the model compared to Guérin-Rechdaoui et al.[15] results.

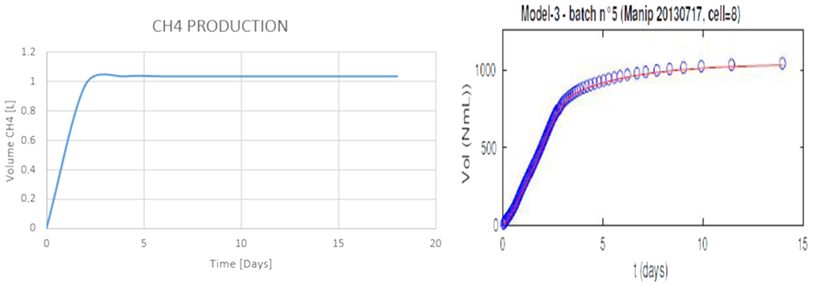

Figure 3. Volume of CH_4 obtained with the model compared to Mottelet et al.[16] results.

Table 6. Comparison between the Results Obtained with the Model and Those Obtained by Mottelet et al.

	cumulative production of methane (mL)	gap (mL)	error (%)
proposed model	1038	12	1.14
Mottelet et al.	1050		

To better appreciate the evolution of the cumulative production of methane as a function of the C/N ratio, we created a database (Table 7) by varying this parameter by noting each time the cumulative production of methane. Indeed, these C/N ratios were taken from the literature and each corresponds to a given type of substrate. The close link between the C/N ratio and the substrate makes it impossible to maintain the same kinetic parameters to study its variation. The analysis of the curve obtained in Figure 4 allows us to conclude that the production of methane increases as the C/N ratio increases. It is also observed that the ideal C/N ratio for optimal production is in the range 25−27 for a maximum methane production of about 9 liters. This result fits well with previous studies. We noticed that methane production, after reaching a maximum, decreases when the C/N ratio is greater than 27. This result is consistent with the literature. Indeed, Yen[18] studied in 2006 the co-digestion of seaweed sludge and waste paper; the results obtained suggest that the optimum C/N ratio for co-digestion of algal sludge and paper waste was in the range of 25−30. Further, Li et al. in 2011,[19] in a study on the evaluation and modeling of biogas production from fat, municipal oils, and fats and synthetic kitchen waste in anaerobic co-digestions, showed that a high methane potential occurred when the C/N ratio was between 25 and 30. In 2017, Lu et al.[20] studied the effects of waste sources on anaerobic co-digestion performance by varying the quality of food waste from six sources in the Xi'an region of China, which were co-digested individually with pretreated corn straw and cattle manure. These effects were analyzed in terms of volatile solid (VS) ratios, C/N ratios, and chemical composition of food residues. The results indicated that the C/N ratios between 17 and 24 gave better methane potentials. The trend observed for the evolution of the cumulative production of methane as a function of the C/N ratio is shown in Figure 4. It is important to remark that apart from the C/N ratio other parameters may influence methane yield in biogas composition. Thus, the combined study of their influence may provide more accurate analysis.

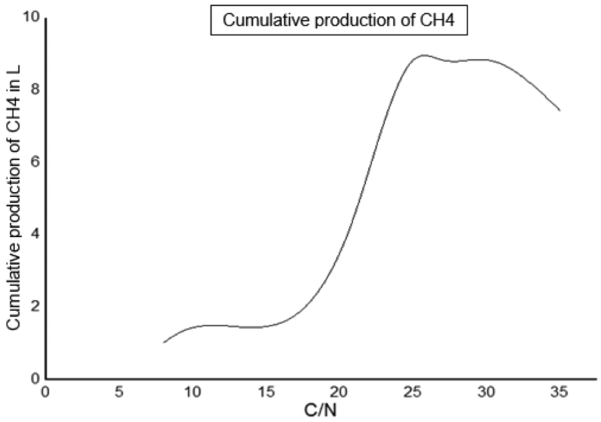

Figure 4. Variation in methane production as a function of the C/N ratio.

4. CONCLUSIONS

The model proposed in this study may allow us to predict the composition of biogas by knowing substrate's C/N ratio. Runge−Kutta's method leads to accurate results. Results showed that an increase of C/N ratio leads to an increase of biogas cumulative production till a C/N ratio of 27 and then to a decrease though the C/N ratio is still increasing. Since experimental studies are more suitable in terms of accuracy, it is important that the coming step of this work deals with experimental studies of the C/N ratio influence on biogas production and a study of the combined effect of several

Table 7. Cumulative Methane Production Values Based on the C/N Ratio

parameters ratio C/N	μ_{1max}	μ_{2max}	K_I	K_{s1}	K_{S2}	k_{hyd}	k_1	k_2	k_3	k_4	k_5	k_6	$[CH_4]$ ($L\ g^{-1}$ VS)	$\%CH_4$	$\%CO_2$
8	3.63	2.67	1.44	1.02	3.45	5	20	0.42	514	310	600	0.88	1.03	70.37	29.63
9	3.4	1.5		41.32	3.14	5	2.9	1.9	1	45.35	600	253	1.29	65	35
20.82	0.4	0.4	0.0025	160	0.82	5	20	464	514	310	600	253	4.35	54	46
25	0.33	0.13	207	0.4	2.93	5.02	20	464	514	310	600	253	8.84	95	05
27	0.33	0.13	207	0.4	2.93	5.02	20	464	514	310	600	253	8.84	95	05
30	0.33	0.13	207	0.4	2.93	5.02	20	464	514	310	600	253	8.85	95.25	04.75
35	3.6	2.5	207	0.4	2.93	5.02	20	464	514	310	600	253	7.45	80.5	19.5

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parameters (temperature, pH, pressure, etc.) on biogas production.

AUTHOR INFORMATION

Corresponding Author

*E-mail: ekouedjen92@gmail.com.

ORCID

Evrard Karol Ekouedjen: 0000-0002-1871-9318

Notes

The authors declare no competing financial interest.

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