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Mathematical Model of a Thermophilic Anaerobic Digestion for Methane Production of Wheat Straw

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Abstract: This paper presents a newly created mathematical model of thermophilic anaerobic digestion of wheat straw carried out in a 2 dm³ bioreactor for methane production. Two batch processes, with 30 mL/dm³ and 35 mL/dm³ organic load, are carried out—one set for parameter identification and one set for model verification. The identification of model parameter values is based on dynamical experiments. It is fulfilled using two different techniques: deterministic sequential quadratic programming algorithm and metaheuristic genetic algorithm. Verification of the developed mathematical models is conducted based on the different data sets of the process. Both models predict the set of the experimental data for all considered process variables well. Genetic algorithm visually fits the data with a higher degree of accuracy, as confirmed by the numerical results for the objective function value.

Keywords: thermophilic anaerobic digestion; lignocellulose; mathematical model; parameters identification; metaheuristic algorithm; verification



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1. Introduction

Fossil fuels consumption is increasing and has become a major contributor to the increasing concentration of carbon dioxide in the atmosphere, leading to global warming [1]. Attention currently is paid to diverse renewable energy sources; although the development and use of renewable energy is not high yet, it is expected to increase as fossil fuel supplies decline. Various technologies are projected to provide renewable energy production, renewable biomass utilisation being one of them [2]. The development and utilisation of biomass as an energy source is still very small compared to its potential. However, lignocellulosic biomass is a promising alternative for meeting the global sustainability criteria [3]. Biogas is a renewable gas, mainly consisting of methane and carbon dioxide that can be used to produce heat and electricity. Biogas can also be utilised directly as a vehicle's fuel after an upgrade to biomethane. Increased production of biogas in the world will favour energy supply needs [4]. Utilising agricultural wastes adds another advantage to their use for the production of biofuels [5].

Wheat straw is considered a favourable substrate for biogas production. Lignocellulosic biomass is one of the most abundant among various renewable alternatives. However, due to its complex and rigid structure, its biodegradability during anaerobic digestion is usually low. Challenges associated with the substrate pretreatment are very important for the overall process performance because this is an expensive step that affects the efficiency of all subsequent steps of sustainable valorisation of the biomass components [6]. Temperature and substrate composition are among the main factors affecting the realisation and stability of anaerobic digestion processes. Pretreatments are mainly carried out at high temperatures and appear effective in realising lignin removal. Cellulose and hemicellulose could be hydrolysed into monomeric sugars such as glucose, xylose, and arabinose, which

could then be converted to biofuels such as methane [7]. The sought results are in the direction of the appearance of structural changes and improved enzymatic hydrolysis [8]. Liquid hot water pretreatment was studied to explore the feasibility of improving the methane yield of wheat straw in anaerobic digestion, and the results showed that the crosslinking structure of wheat straw had been broken [9].

Improved biomethane yield was also obtained after thermal pretreatment [10]. Conveying the process of anaerobic digestion of wheat straw under thermophilic conditions could strongly affect the performance of biogas digesters, leading to increased hydrolysis rates. Temperature accelerates biochemical processes, enabling higher degradation rates and biogas yields from a wide variety of substrates compared to mesophilic anaerobic digestion processes [11].

The operating physicochemical parameters of anaerobic digesters and the methods of acceleration and optimisation used to improve biogas yields remain of utmost importance [12].

The complicated model structure of anaerobic digestion processes is typically described using nonlinear differential equations. The mathematical modelling of such processes is a complex problem, and the selection of a proper method for optimisation is fundamental for the accurate estimation of model parameters.

There are a few techniques that are appropriate in the considered case problem. Among them are conventional optimisation methods, such as sequential quadratic programming (SQP) [13,14], and metaheuristic techniques, such as genetic algorithms (GA) [15,16]. As a good opportunity for conventional optimisation methods, metaheuristic techniques have been recognised. They are capable of finding a suitable solution for acceptable computational time [17–19]. Some of the most powerful metaheuristic algorithms inspired by nature—such as genetic algorithms, artificial bee colony algorithm, Cuckoo search algorithm, etc.—have been proposed and validated for many optimisation problems and have been endorsed as prosperous for the estimation of bioprocess models parameters [20–22].

The paper strives to propose a mathematical model of thermophilic anaerobic digestion of wheat straw carried out in a 2 dm³ bioreactor for methane production. For the purposes of modelling, two batch processes with 30 mL/dm³ and 35 mL/dm³ organic load, are carried out—one set for parameter identification and one set for model verification.

The numerous successful GA applications in the field of modelling bioprocesses have inspired the authors to adopt and employ GA for the first time for solving the challenging problem of parameter identification of nonlinear model of the considered anaerobic digestion process. The obtained numerical estimates of model parameters are compared to the results from application of SQP—one of the prominent techniques for solving constrained nonlinear optimisation problems. Both obtained mathematical models are verified with an independent data set.

The paper is organised as follows. After the Introduction, experimental setup and mathematical modelling problem are provided in Section 2. The experimental results are presented and discussed in Section 3. Concluding comments and future research aspects are summarised in Section 4.

2. Materials and Methods

2.1. Experimental Setup

In this study, as substrate for the anaerobic digestion, mechanically pretreated wheat straw was used. For this purpose, native wheat straw was ground to a final particle size of 1–2 mm using a hammer mill, followed by additional milling in a knife mill until a satisfactory particle size was reached.

The experiments were conducted in a bioreactor with a working volume of 2 dm³ for methane production. Initial experiments, as well as inoculum maintenance, were conducted in a bioreactor of 1 dm³. In all experiments, a temperature suitable for growing thermophilic microorganisms (55 °C) was maintained automatically, and continuous stirring was also

applied. All experiments were carried out in batch mode of operation. The daily released biogas was kept and measured using a gas holder based on the water displacement method.

Cellulose content was determined by a spectrophotometric method [23]. Cellulose-containing materials were cleared of impurities such as lignin, hemicellulose, xylosans, and other low molecular weight compounds by extraction with an acetate-nitrite mixture. Purified cellulose was dissolved in 67% H₂SO₄, followed by a colour reaction using an anthrone reagent. Cellulose concentration was determined after measuring the absorbance at 620 nm. Glucose was analysed using the Miller method [24], which is based on the redox reaction between reducing sugars and sodium dinitrosalicylate, resulting in a reddish-brown derivative. Absorption was measured at a wavelength of 530 nm. The concentration of acetate was determined by a Thermo Scientific gas chromatograph (Focus GC model) equipped with a Split/Splitless injector, column: TG-WAXMS A (length 30 m, diameter 0.25 mm, film thickness 0.25 µm), and flame ionisation detector (FID). Prior to injection, the pH of the sample taken from the bioreactor was adjusted to pH 2.0 with 37% H₃PO₄. After one hour, the sample was centrifuged at 15,000 × g min⁻¹ for 10 min, and an aliquot of the supernatant was mixed with an equal volume of 1% 2,2-dimethyl-butylric acid (as internal standard).

The content of the biogas produced from the bioreactor (containing methane) was estimated with a “Dräger, X-am7000” model device (Germany), equipped with infrared sensors for measuring the relative content of CH₄ and CO₂ in percent by volume and a catalytic sensor for H₂S (in ppm).

2.2. Mathematical Modelling Problem

In order to find a set of design parameters, $x = \{x_1, x_2, \dots, x_n\}$, optimisation techniques are used. The resulting outcomes can be defined as optimal ones. Usually, a minimisation or maximisation of a predefined system characteristic, dependent on x , is considered. In a more complex formulation, the objective function $f(x)$, which should be minimised (maximised), might be subject to constraints. The problem can be described as follows:

$$\min_x J(x) \quad (1)$$

where x is the vector of design parameters, $J(x)$ is the objective function.

The discrepancy between the model and real data is configured as an optimisation criterion:

$$J = \sum_{i=1}^k \sum_{j=1}^m (Y_{ij} - \hat{Y}_{ij})^2 \quad (2)$$

where k is the number of process variables, m is the number of data points, Y_{ij} represents the observed experimental data values, and \hat{Y}_{ij} represents model predicted values.

The obtained solution depends mainly on the problem complexity, i.e., the number of design variables and constraints. Solution efficiency also depends on constraints characteristics and the objective function.

2.3. Sequential Quadratic Programming

Quadratic programming (QP) considers the minimisation (maximisation) of a quadratic objective function with linear constraints. Trustworthy solution techniques are readily accessible. Constrained minimisation is the problem of finding a vector x that is a local minimum to a scalar function $J(x)$ subject to constraints on the allowable x . The method mimics Newton’s method for constrained optimisation, just as is carried out for unconstrained optimisation. A quasi-Newton updating method is used at each iteration in order to approximate the Hessian of the Lagrangian function. A QP subproblem solution, needed to form a search direction, is then generated. The background of the basic SQP algorithm applied here is described in [25].

2.4. Genetic Algorithms

GA is a metaheuristic technique inspired by genetics and Darwin's theory of evolution [16].

GA operates on a set of individuals (chromosomes), $Pop(t) = x_1^t, \dots, x_n^t$, for a certain generation t . Each chromosome symbolises a potential problem–solution. Each solution is encoded as a finite vector of variables of a certain alphabet, usually a binary one. The individuals of the offspring are modified by the genetic operators' crossover and mutation [26].

The workflow of a common GA can be roughly presented as follows. The initial set of solutions can be randomly generated. To evaluate the capability of a chromosome compared to the whole population, the fitness function is used. Chromosomes with high values of the fitness function are more likely to be chosen for reproduction. The operators (crossover and mutation) are used in order to reproduce a new offspring, which becomes generation $t + 1$. The offspring replaces chromosomes existing in the population, and the generation is reiterated until the stopping conditions are fulfilled.

3. Results and Discussion

3.1. Experimental Studies

Experimental studies of batch anaerobic digestion of wheat straw at thermophilic conditions are performed. The mechanically chopped wheat straw with the corresponding quantity (30 mL/dm³ or 35 mL/dm³) is suspended in distilled water, and inoculum from a working biomethane generating bioreactor 30% (vol) is also introduced in the batch process, which continues for 15 days. The inoculum quantity is previously determined as it ensures good buffer capacity at the beginning of the process. Stirring is maintained at 100 min⁻¹. The measurements of cellulose, glucose, acetate, and methane content in the biogas in the bioreactor are available for 30 and 35 mL/dm³ initial substrate organic load. The results are presented in Table 1 for 30 mL/dm³ and in Table 2 for 35 mL/dm³. All concentrations given are mass concentrations, not molar concentrations.

Table 1. Batch experiments with 30 mL/dm³ organic load.

Duration, day	Cellulose, mL/dm ³	Glucose, mL/dm ³	Acetate, mL/dm ³	Methane, mL/dm ³
0	10.73	0.013	0.13	0.0000
1	9.69	0.053	1.36	0.0099
2	8.84	0.082	1.69	0.0771
3	8.16	0.101	1.00	0.1224
4	7.62	0.113	0.11	0.1313
5	7.20	0.119	0.14	0.0301
6	6.90	0.122	0.13	0.0122
7	6.68	0.122	0.11	0.0136
8	6.54	0.122	0.09	0.0151
9	6.45	0.122	0.09	0.0130
10	6.39	0.124	0.09	0.0120
11	6.35	0.126	0.09	0.0110
12	6.30	0.126	0.08	0.0100
13	6.23	0.124	0.08	0.0090
14	6.13	0.122	0.08	0.0090
15	5.96	0.120	0.08	0.0090

Table 2. Batch experiments with 35 mL/dm³ organic load.

Duration, day	Cellulose, mL/dm ³	Glucose, mL/dm ³	Acetate, mL/dm ³	Methane, mL/dm ³
0	11.04	0.019	0.15	0.000
1	9.83	0.060	1.40	0.010
2	8.85	0.092	1.70	0.012
3	8.07	0.116	1.27	0.140
4	7.48	0.132	0.96	0.150
5	7.04	0.143	0.43	0.100
6	6.73	0.149	0.28	0.047
7	6.54	0.152	0.12	0.030
8	6.44	0.153	0.08	0.030
9	6.41	0.152	0.08	0.030
10	6.42	0.152	0.08	0.030
11	6.45	0.153	0.08	0.030
12	6.48	0.152	0.08	0.030
13	6.49	0.150	0.08	0.020
14	6.45	0.150	0.08	0.020
15	6.34	0.150	0.08	0.020

3.2. Mathematical Modelling

3.2.1. Structural Identification

A deterministic nonlinear dynamic system of differential equations was used to describe the thermophilic anaerobic digestion process for methane production of wheat straw as follows:

$$\frac{dS_0}{dt} = -\beta X_1 S_0 \quad (3)$$

$$\frac{dX_1}{dt} = \mu_1 X_1 \quad (4)$$

$$\frac{dS_1}{dt} = \beta X_1 S_0 - \frac{1}{Y_1} \mu_1 X_1 \quad (5)$$

$$\frac{dX_2}{dt} = \mu_2 X_2 \quad (6)$$

$$\frac{dS_2}{dt} = Y_b \mu_1 X_1 - \frac{1}{Y_2} \mu_2 X_2 \quad (7)$$

$$Q_{CH_4} = Y_{CH_4} \mu_2 X_2 \quad (8)$$

$$\mu_1 = \frac{\mu_{1max} S_1}{k_{1s} + S_1} - b_1 \quad (9)$$

$$\mu_2 = \frac{\mu_{2max} S_2}{k_{2s} + S_2} - b_2 \quad (10)$$

Equations (3) and (5) present the balance of the effluent substrate (S_0)—cellulose and its transformation to glucose (S_1) after hydrolysis. Equations (4) and (6) describe the dynamics of the biomass concentrations (X_1 and X_2). Equation (7) presents the dynamics of the intermediate product—acetate (A_{C1}), and the algebraic Equation (8)—the flow rate of the methane in the gas phase in the bioreactor. For the specific growth rates of the biomasses, a Monod type function was adopted—Equations (9) and (10).

3.2.2. Parameter Identification

The identification of model parameters values is based on dynamical experiments of thermophilic anaerobic digestion of wheat straw carried out in a 2 dm³ bioreactor with 30 mL/dm³ organic load. It is fulfilled using two different techniques: deterministic SQP algorithm and metaheuristics—GA.

The numerical computations are carried out on a PC/Intel Core i5-2320 CPU@2.67 GHz, 4 GB Memory (RAM), Windows 7 (64 bit) operating system. Modelling and numerical experiments are performed using *Matlab R2013a*.

To locate solution point algorithms such as SQP, usually involves a few iterations. Such local search methods may fall into local minima. It is known that SQP estimates are very sensitive to the initial solutions. At the same time, GA does not have such a dependence, as the initial solutions (initial population) are generated randomly within the predetermined lower and upper limits of each model parameter.

According to the problems considered here, the GA parameters such as population number, crossover and mutation probability, etc., are tuned previously. A series of tests are conducted in order to reach the best solution quality. The optimal GA parameters are presented in Table 3.

Table 3. GA parameters.

Parameter	GA
Population number (N_{pop})	200
Generation gap ($ggap$)	0.97
Maximal number of iterations ($iter_max$)	100
Crossover probability (p_c)	0.75
Mutation probability (p_m)	0.01

When comparing stochastic algorithms, such as GA, the general approach is to operate with the obtained average results. The GA discussed here has been executed 30 times under the same conditions, e.g., algorithms' parameters and functions, presented in Table 3. The obtained results are summarised in Table 4. To better distinguish the results from the algorithms applied for the considered problem, in addition to the obtained values of the objective function, the values of Mean Absolute Error (MAE) [27] are calculated.

Table 4. Results from model parameter identification based on SQP and GA.

Parameter	Deterministic Algorithm	Metaheuristic Algorithm	Deterministic Algorithm
	SQP ₁	GA	SQP ₂
	Value	Best	Value
J	13.91	9.87	10.22
MAE	2.55	0.62	0.64
μ_{1max}	0.29	0.74	0.98
k_{1s}	1.67	19.16	10.96
b_1	0.10	0.0003	0.004
β	0.34	0.51	0.52
Y_1	0.04	0.001	0.001
Y_b	2.03	1998	2476
μ_{2max}	0.5	3.69	4.85
k_{2s}	0.31	1.005	1.83
Y_2	0.18	7.22	5.74
Y_{CH_4}	3.65	0.005	0.01
b_2	0.1	0.046	0.029

Primarily, adjustment of the coefficients was carried out with the SQL algorithm, setting wider search limits. The so-found model does not describe experimental data well (SQP₁ in Table 4). The observed objective value of the model during the identification is $J = 13.91$ and $MAE = 2.55$. For this reason, initially, GA is run for a few iterations, e.g., 100, to give some closure to the optima model parameters estimates. The new lower and upper limits for SQP are defined using these results. Based on the new observed results, a

mathematical model with a much higher accuracy is obtained (SQP₂ in Table 4)— $J = 10.22$ and MAE = 0.64.

In the considered identification problem, the SQP and GA optimisation algorithms find two different solutions for the model parameters values. The estimates for $\mu_{1\max}$, β , $\mu_{2\max}$, and Y_2 are in similar ranges; the model parameters Y_b , Y_{CH_4} , and b_2 are identified with a small difference; while the estimates of k_{1s} , b_1 , Y_1 , and k_{2s} are very different. As no information has been found in the literature on the values of the respective coefficients in such processes, it is not possible to conclude which coefficients should be preferred.

The presented comparison (Figures 1 and 2) of the experimental data and model predictions for cellulose, glucose, acetate, and methane obtained by both algorithms demonstrates the better performance of the proposed GA compared to the SQP algorithm.

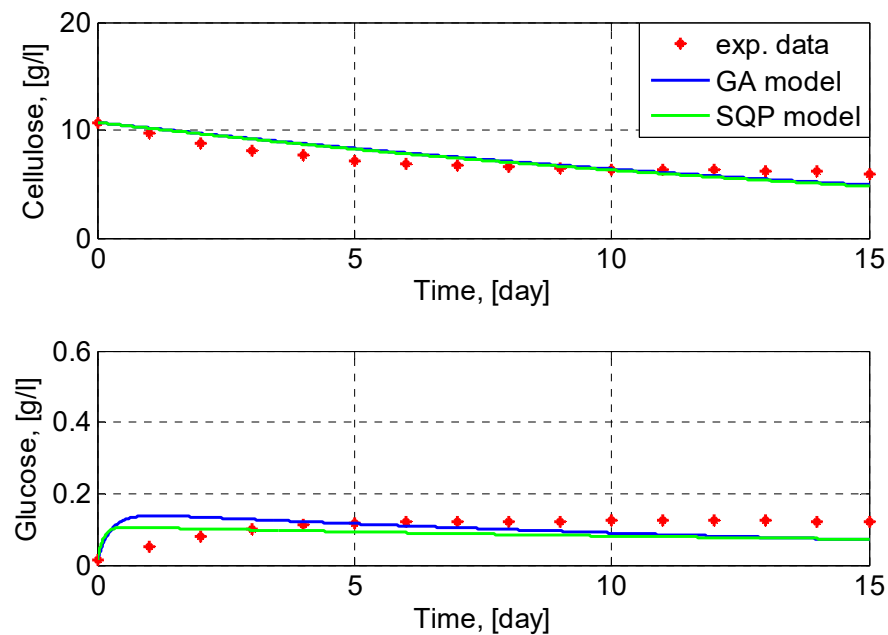


Figure 1. Comparison of the experimental data and model predictions for substrates cellulose and glucose obtained by GA and SQP models—identification results.

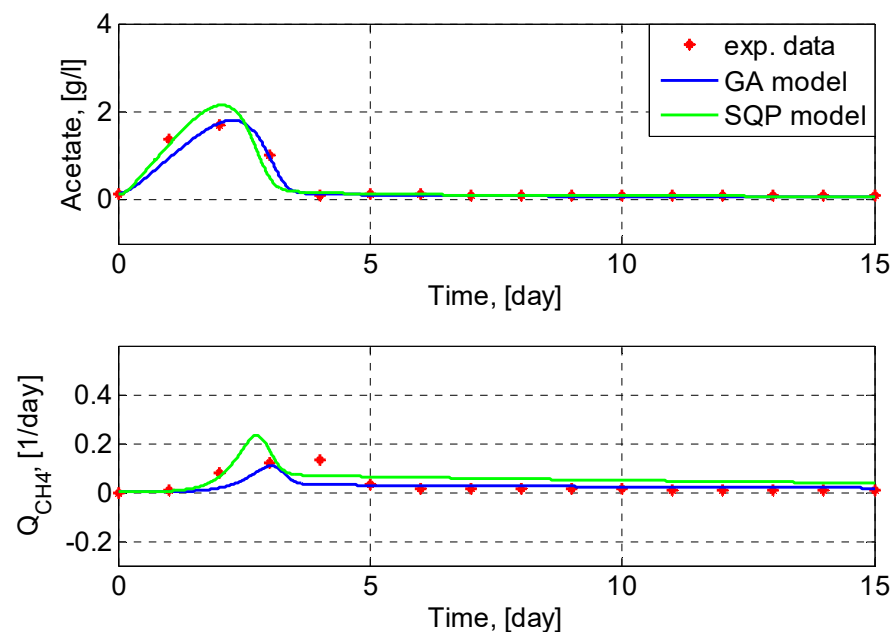


Figure 2. Comparison of the experimental data and model predictions for acetate and methane obtained by GA and SQP models—identification results.

3.2.3. Model Validation

Verification of the developed mathematical models is carried out based on the different data set of the process, i.e., 35 mL/dm^3 initial substrate organic load. The proposed models based on GA and SQP₂ algorithms, with the parameters listed in Table 4, are simulated with the new initial substrate organic load (35 mL/dm^3 instead of 30 mL/dm^3), as well as with the new initial values of cellulose, glucose, acetate, and methane in the new experimental data set. The obtained results (from the comparison between the model and the new data set) are shown in Figure 3 (cellulose and glucose dynamics) and Figure 4 (acetate and methane dynamics). The observed objective value of the model during the verification is $J_{\text{SQP}} = 18.22$ and $J_{\text{GA}} = 17.91$.

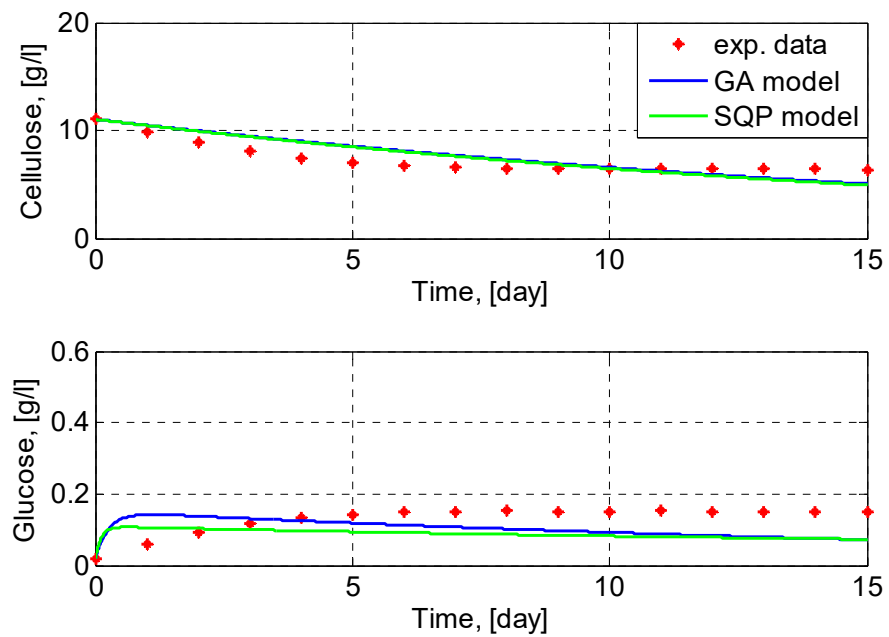


Figure 3. Comparison of the experimental data and model predictions for substrates cellulose and glucose obtained by GA and SQP models—validation results.

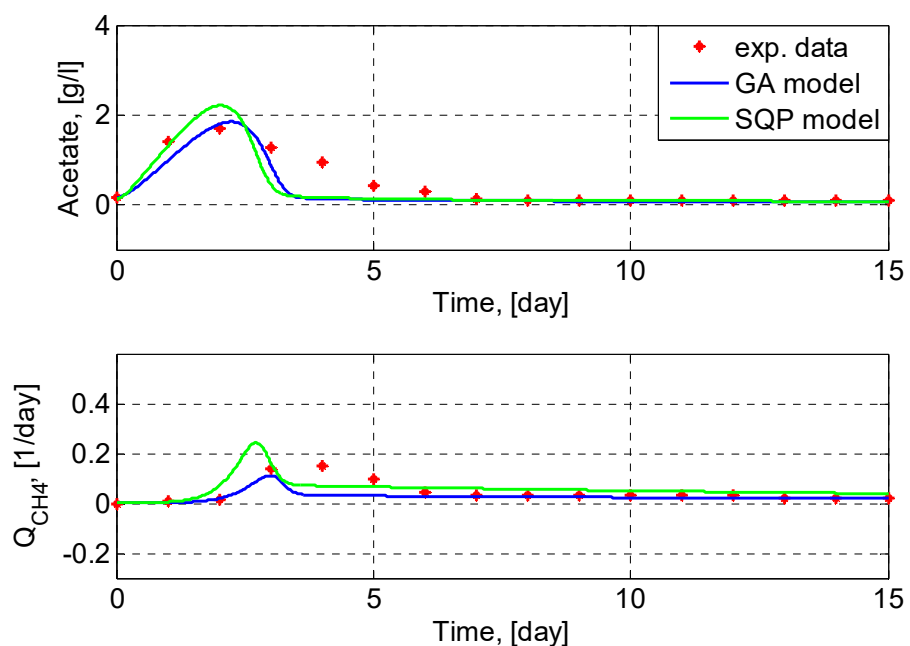


Figure 4. Comparison of the experimental data and model predictions for acetate and methane obtained by GA and SQP models—validation results.

As seen in Figures 3 and 4, both GA and SQP models predict well the new set of the real data for variables cellulose, glucose, acetate, and methane process. However, GA visually fits the experimental data with a higher degree of accuracy, as established by the numerical results for the objective function value J .

Simulation models development for the anaerobic digestion process of municipal solid wastes [28], or animal manure and sewage sludge [29,30], had been carried out with the aim of coping with the harmful effects of such disposals on the health of humans, animals, and the environment. In this study, the effect of elevated temperature was revealed as suitable to be explored with the feasibility of improving the methane yield of wheat straw as agricultural waste in anaerobic digestion, as it increased the rate of the process and made unnecessary harsh pretreatment techniques. The created model presented could serve for monitoring and control of such processes.

4. Conclusions

In this paper, a new model of the anaerobic digestion of wheat straw is presented. The nonlinear model structure depicted is quite simple but with many coefficients and proven for the anaerobic digestion processes. Parameter identification is made using two different methods—deterministic SQP algorithm and metaheuristic algorithm—GA.

SQP separately has not given satisfactory results with the originally defined initial solutions and parameters limits. GA has been applied here as a competitive metaheuristic technique, proven as an effective optimisation method for solving complex, real problems, such as parameters estimation of yeast and bacterial fermentation models. The best results of SQP are obtained using a technique which combines GA and SQP algorithm. The obtained numerical results regarding the computational efficiency of the algorithms unambiguously show that GA have an advantage over SQP.

Results illustrating the model validity are also presented using a different dataset, which showed good performance with the error even smaller compared with the error from the identification. The results show good model validity.

One of the most difficult questions to answer concerns the interpretation of the coefficients of the model. At the moment in the literature, there is no information about the values of the considered coefficients of the processes models proposed in this paper. This is the reason why it is not possible to draw conclusions about which coefficients should be preferred.

In a future plan, the hybrid SQP-GA technic, for model identification could be developed, aiming to take advantage of different optimisation strategies while avoiding the algorithms' weaknesses. Designing an appropriate hybridisation of metaheuristics, an algorithm with better performance can be offered.

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Nomenclature

S_0	Cellulose concentration	[mL/dm ³]
X_1	Acidogenic bacteria concentration	[mL/dm ³]
S_1	Glucose concentration	[mL/dm ³]
X_2	Methanogenic bacteria concentration	[mL/dm ³]
S_2	Acetate concentration	[mL/dm ³]
Q_{CH_4}	Methane flow rate	[mL/dm ³]
μ_1	Specific growth rate of acidogenic bacteria	[h ⁻¹]
μ_2	Specific growth rate of methanogenic bacteria	[h ⁻¹]
μ_{1max}	Maximum specific growth rate of acidogenic bacteria	[h ⁻¹]
k_{1s}	Saturation coefficient for acidogenic bacteria	[mL/dm ³]
b_1	Maintenance rate of acidogenic bacteria	[h ⁻¹]
β	Coefficient of biodegradability	[L/(g·h)]
Y_1	Yield coefficient for acidogenic bacteria	[-]
Y_b	Coefficient	[-]
μ_{2max}	Maximum specific growth rate of methanogenic bacteria	[h ⁻¹]
k_{2s}	Saturation coefficient for methanogenic bacteria	[mL/dm ³]
b_2	Maintenance rate of methanogenic bacteria	[h ⁻¹]
Y_2	Yield coefficient for methanogenic bacteria	[-]
Y_{CH_4}	Yield coefficient for methane	[mL/dm ³]

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