Invited Paper

A Simple Mathematical Model of the Anaerobic Digestion of Wasted Fruits and Vegetables in Mesophilic Conditions

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Abstract: Anaerobic digestion is an effective biotechnological process for treatment of different agricultural, municipal and industrial wastes. Use of mathematical models is a powerful tool for investigations and optimisation of the anaerobic digestion. In this paper a simple mathematical model of the anaerobic digestion of wasted fruits and vegetables was developed and verified experimentally and by computer simulations using Simulink. A three-step mass-balance model was considered including the gas phase. The parameter identification was based on a set of 150 days of dynamical experiments in a laboratory bioreactor. Two step identification procedure to estimate 4 model parameters is presented. The results of 15 days of experiment in a pilot-scale bioreactor were then used to validate the model.

Keywords: Anaerobic digestion, Wasted fruits and vegetables, Laboratory experiments, Pilot-scale experiment, Mathematical model, Parameters identification, Verification.

Introduction

Anaerobic digestion (AD) is an effective biotechnological process for the treatment of diffrent agricultural, municipal and industrial wastes [1, 6]. It combines environmental depollution (ecological aspect) with production of renewable energy – biogas, the main component of which is methane (energetical aspect). However, AD is a very unstable process in regard to biogas reactors operation due to the complicated interactions between different microbial species, as well as to the complex transformations of organic matter affected by a variety of environmental factors [9]. In this context, the use of mathematical models is a powerful tool for investigations and optimisation of the AD [2, 7, 12, 17]. ADM1 [2] is the most complex and powerful AD model. However, such types of models are very complex for practical use. That is why for particulate substrates more simple models are developed [7, 12, 17].

Wasted fruits and vegetables (WFV) are produced in large quantities at markets in many large cities [5, 8, 11] and it is inadequately treated by land application. AD reduces the need for waste disposal and leads to the formation of biogas (energy) and digestate (potential manure). Our previous studies demonstrated good performances of AD of WFV at mesophilic temperature [15, 16]. However, until now very few studies were carried out concerning mathematical modeling of the AD of WFV [4].

The aim of this study is to develop and verify experimentally a simple mathematical model of the anaerobic digestion of wasted fruits and vegetables in mesophilic conditions.

Materials and methods

Materials

Specificity of the WFV as substrates for AD

In this study, as substrate for AD, a mixture of WFV, at the ratio of 40% wasted potatoes (WP), 20% wasted tomatoes (WT), 20% wasted cucumbers (WC) and 20% wasted apples (WA) was used. The WFV were collected from markets in Sofia. The material was homogenized in an electric blender.

The following parameters were obtained using analytical methods: total solids (TS), volatile solids (VS), pH and concentration of volatile fatty acids (VFA).

Experimental setup (laboratory scale bioreactor)

The experiments in mesophilic temperature $(34\pm0.5 \text{ °C})$ were carried out in a 5-L bioreactor (BR) with a working volume of 3 L. The BR was operated in semi-continuous mode. The scheme of the laboratory scale is presented on Fig. 1.



Fig. 1 Scheme of a laboratory scale bioreactor

In Fig. 1 Controller 1 is for temperature regulation (using sensor Pt 100), Controller 2 - for regulation of the speed of the stirrer (50 rpm usually), Influent and Effluent flows have been realized using peristaltic pumps.

Studies were performed for different values of the dilution rate (*D*) and for constant concentration of the total solids in the influent ($TS = 70 \text{ g/dm}^3$) [16]. The values of *D* and of the corresponding hydraulic retention times (HRT = 1/*D*) for both bioreactors during experiments are summarized in Table 1.

Samples for pH measurements and biochemical analyses were taken from the effluent of the bioreactor. The biogas flow rate and the contents of CH_4 and CO_2 in the biogas were measured once a day. Corrections of pH were done (if necessary) with additions of 2 N *NaOH* in the influent.

Time of continuous cultivation,	<i>D</i> ,	HRT,
[days]	$[day^{-1}]$	[day]
1 - 21	0.067	15
22 - 54	0.010	100
55 - 62	0.020	50
63 - 82	0.040	25
83 - 132	0.020	50
133 - 189	0.040	25

Table 1. Dilution rates during AD of FVW

Experimental setup (pilot scale bioreactor)

The experiments were carried out in a 100-L pilot scale anaerobic BR with a working volume of 80 L in mesophilic temperature $(34\pm0.5 \text{ °C})$ [14, 18]. The bioreactor was operated in semi-continuous mode. The scheme of the pilot-scale ABR is shown on Fig. 2.



Fig. 2 Scheme of a pilot scale experimental set-up:
1 – vessel for the influent (substrate); 2 – vessel for the effluent (digestate);
3 – heater control; 4 – sensors for Q, CH₄, CO₂; ABR – anaerobic bioreactor;
GH – gas holder; M – AC drive of the stirrer; P – peristaltic or progressive cavity type pump;
t – sensor for the temperature in the bioreactor;
Press – sensor for the pressure in the bioreactor.

The substrate (WFV) was stored in plastic can of 25 L placed in the influent line of the BR. The digestate taken out of it during semi-continuous operation (feeding one to 24 times daily) is stored in plastic can of 50 L in the next-door auxiliary service premises of the biogas plant.

A biogas outlet from the upper bioreactor flange leads off the biogas to a 200 L metal gasholder (GH) operating on the water displacement principle (the inner vessel, placed in vessel with water, is displaced vertically by the biogas).

Methods

Analytical methods

TS and VS. TS and VS have been measured according to standard methods [10].

Biogas yield. Daily biogas production was measured by the water displacement technique (graduated gasholder). The biogas flow rate was measured through transformation of the linear shift of the inlet vessel of the GH into normalized electrical signal (sensor developed by our team).

Biogas composition. The biogas composition has been measured with computerized device of Dräger (type X-am 7000 with infrared sensors) for the laboratory experiments and with infrared sensors of MSR for the pilot scale experiments.

pH in the bioreactors have been measured daily in the effluent with laboratory pH-meter. Samples for pH measurements and biochemical analyses were taken from the effluent of the bioreactor (digestate). Corrections of pH were done (if necessary) with additions of 2 N NaOH in the influent.

Chemicals. All chemicals used have been analytical grade and have been obtained from commercial sources.

Calculations

For comparison of laboratory and pilot scale experimental data the specific biogas flow-rate $(Q_{sp}, [dm^3 \cdot biogas/L \text{ of liquid} \cdot day])$ obtained from 1 L of the working volume of the BR was calculated according to the following formula:

$$Q_{sp} = \frac{Q}{V_{work}},$$

where Q is the daily biogas flow-rate and V_{work} is the working volume of the corresponding bioreactor.

Experimental results

AD of WFV in mesophilic conditions in laboratory bioreactor

The start-up of the anaerobic BR was done with activated sludge (AS) from the Sofia waste waters treatment plant. After a period of co-digestion of WFV and AS in different proportions, the AD process has continued with WFV only. AD of WFV only was studied at mesophilic conditions for a time period of 250 days for different values of D, depicted on Table 1 and for TS = 70 g/L. The biogas flow-rate and of the biogas composition (CO_2 and CH_4 contents) were measured. The specific biogas flow-rates Q_{sp} were calculated according to the above presented formula. The average specific biogas flow-rates Q_{sp} for the corresponding steady-states are presented in Table 2.

$\begin{array}{c} \boldsymbol{D},\\ [\mathrm{day}^{-1}] \end{array}$	Number of days for <i>Q_{sp}</i> calculation	$\underbrace{\boldsymbol{\mathcal{O}}_{sp}}_{[dm^3/L/day]}$
0.01	33	0.64
0.02	49	0.84
0.04	65	1.45
0.06	47	2.15

Table 2. Values of Q_{sp} in study-state for different values of D

It is evident from Table 2 that with the increase of D (up to the critical value $D = 0.1 \text{ day}^{-1}$), Q_{sp} increased 3-4 times. At the same time the methane and the carbon dioxide in the biogas varies in the intervals 50-70% and 27-42%, respectively.

AD of WFV in mesophilic conditions in pilot scale bioreactor

Experimental studies of AD with the same mixture of WFV with step change of the dilution rate from 0.025 to 0.0125 and for constant concentration of the total solids in the influent (TS = 70 g/dm³) were performed with the pilot anaerobic plant at mesophilic temperature (34 °C).

Mathematical modelling of the process

Generally the AD processes can be divided in four main stages:

- hydrolysis of undissolved high-molecular weight compounds (proteins, sugars, fats) to soluble low-molecular weight compounds (monosugars, aminoacids, long-chain fatty acids, glycerol);
- acidogenesis fermentation of low-molecular weight compounds from the previous stage to VFA (propionate, butirate, acetate), hydrogen and carbon dioxide;
- acetogenesis transformation of VFA to acetate, hydrogen and carbon dioxide;
- methanogenesis mediated by acetoclastic methanogens (converting acetate to methane and carbon dioxide) and hydrogenotrophic methanogens (producing methane from hydrogen and carbon dioxide).

Studying the process of AD of WFV we supposed that the stage of acetogenesis is not so important. It this case the three-stage biochemical scheme [17], describing hydrolysis, acidogenesis and methanogenesis, was adopted.

Structure of the model

Liquid phase

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On the basis of the above-presented experimental studies and according to the relatively simple three-stage biochemical scheme of the AD, the following set of five ODE is adopted for the structure of the model in our case:

$$\frac{dS_0}{dt} = -DS_0 - \beta X_1 S_0 + DY_p S_{in}$$
(1)

$$\frac{dX_1}{dt} = (\mu_1 - D)X_1 \tag{2}$$

$$\frac{dS_1}{dt} = -DS_1 + \beta X_1 S_0 - \mu_1 \frac{X_1}{Y_1}$$
(3)

$$\frac{dX_2}{dt} = (\mu_2 - D)X_2$$
(4)

$$\frac{dS_2}{dt} = -DS_2 + Y_b \mu_1 X_1 - \mu_2 \frac{X_2}{Y_2},$$
(5)

in which the bacterial growth is of Monod type:

$$\mu_1 = \frac{\mu_{max1}S_1}{\left(k_{s1} + S_1\right)}, \ \mu_2 = \frac{\mu_{max2}S_2}{\left(k_{s2} + S_2\right)} \tag{6}$$

In this mass-balance model, Eq. (1) describes the hydrolysis in a very simple way, where the first term reflects the effluent flow rate of liquid, the second term – the hydrolysis of the diluted organics by acidogenic bacteria and the third one – the influent flow rate of liquid with

concentration of the diluted organics S_{in} , g/dm^3 . Eq. (2) describes the growth and changes of the acidogenic bacteria (with concentration X_1 , g/dm^3), consuming the appropriate substrate (with concentration S_1 , g/dm^3). The mass balance for this substrate is described by Eq. (3), where the first term reflects the substrate S_1 , g/dm^3 , in the effluent flow rate of liquid, the second term – the substrate S_0 , g/dm^3 , formed as a result of the hydrolysis and the last one – the consumption by the acidogenic bacteria. Eq. (4) describes the growth and changes of the methane producing (methanogenic) bacteria (with concentration X_2 , g/dm^3), consuming acetate (with concentration S_2 , g/dm^3). The mass balance equation for acetate in Eq. (5) has three terms in his right side. The first one reflects the acetate in the effluent liquid, the second one – the acetate formed as a result of the activity of acidogenic bacteria and the third one – the consumption of acetate by the methanogenic bacteria.

The Eqs. (6) present the specific growth rate of the acidogenic bacteria μ_1 , day⁻¹, and the specific growth rate of the methanogenic bacteria μ_2 , day⁻¹, both of Monod type. β , Y_p , k_1 , k_2 , Y_b , Y_1 , Y_2 , μ_{max1} , μ_{max2} , k_{s1} , k_{s2} are coefficients. D, day⁻¹, is the dilution rate – the control input.

Gas phase modelling

In [3], assuming that the biogas is mainly composed of CO_2 and CH_4 and neglecting the concentration of dissolved methane (due to its very low solubility) the following simplified gases dynamics is presented for the two populations AD model (Eqs. (1-6)):

$$Q = Q_{CH_4} + Q_{CO_2}$$
(7)

with

$$Q_{CH_4} = K_{X_2CH_4} \mu_2 X_2$$
$$Q_{CO_2} = k_L a (CO_2 - K_H P_c)$$

and

$$\frac{dCO_2}{dt} = -DCO_2 - k_L a [CO_2 + S_2 - K_H P_c] + K_{X_1 CO_2} \mu_1 X_1 + K_{X_2 CO_2} \mu_2 X_2$$
(8)

with

$$P_{c} = \frac{\varphi - \sqrt{\varphi^{2} - 4K_{H}P_{T}(CO_{2} + S_{2})}}{2K_{H}},$$

$$\varphi = CO_{2} + S_{2} + K_{H}P_{T} + \frac{K_{X_{2}CH_{4}}}{k_{L}a}\mu_{2}X_{2},$$

where CO_2 is the carbon dioxide concentration in the liquid phase, P_c – the carbon dioxide partial pressure in the gas phase, [atm], P_T – total pressure, [atm], K_H – Henry's constant, [mmol/dm³ per atm], $K_{X_1CO_2}$, $K_{X_2CO_2}$, $K_{X_2CH_4}$ – coefficients, k_La – liquid-gas transfer constant, [day⁻¹].

The algebraic Eq. (7) describes the formation of biogas with flow rate Q, $[dm^3gas/L medium/day]$, including the methane flow rate Q_{CH_4} and the carbon dioxide flow rate Q_{CO_2} .

Eq. (8) describes the mass balance of the carbon dioxide in the liquid phase, where the first term reflects the effluent flow rate of liquid, the second term – the effluent flow rate of carbon dioxide in the gas, the third and the forth terms – the formation of carbon dioxide in the liquid phase by the acidogenic and by the methanogenic bacteria, respectively.

Parameters estimation

We postulated that kinetic and yield coefficients of the mass balance model (Eqs. (1-6)) are known and we adopted their values from [13]: $\beta = 3$; $Y_p = 0.144$; $Y_b = 5$; $Y_1 = 0.15$; $Y_2 = 0.24$; $\mu_{max1} = 0.4$; $k_{s1} = 4.2$; $\mu_{max2} = 0.25$; $k_{s2} = 0.42$.

We supposed that only the 4 coefficients ($K_{X_1CO_2}$, $K_{X_2CO_2}$, $K_{X_2CH_4}$ and k_La) of Eqs. (7) and (8), describing the gas phase, are unknown.

For estimation of these 4 unknown coefficients nonlinear constrained optimization method from the "Optimisation toolbox" for MATLAB with gradient method (Sequential quadratic programming) was used. The procedure finds the minimum of a constrained nonlinear multivariable function (criterion).

The following criteria for minimization were adopted:

$$I_1 = \sum \left(Q_{CH_4}^{\exp} - Q_{CH_4}^{\text{model}} \right)^2 = \varepsilon_1^2 \to \min$$
(9)

$$I_2 = \sum \left(Q_{CO_2}^{\exp} - Q_{CO_2}^{\text{model}} \right)^2 = \varepsilon_2^2 \to \min$$
(10)

Analysing the experimental data and Eq. (7) of the model two steps estimation procedure has been adopted:

- At the first step the unknown methane yield coefficient $K_{X_2CH_4}$ was estimated with experimental data for the specific methane flow-rate $Q_{spCH_4}^{exp}$ calculated from the laboratoryscale data for one step change of the input D (from D = 0.02 to D = 0.04) and minimization of the criterion (9). The estimation was made starting from different initial value of the coefficient in the admissible range (0.01-20.0). Regardless of the initial value of the coefficient $K_{X_2CH_4}$, it retains its value of 17.345 (Table 3).

Table 3				
Experiment	$K_{X_2CH_4}$	Criterion	$K_{X_2CH_4}$ init. cond.	
1	17.345	21.016	0.01	
2	17.345	21.016	10	
3	17.345	21.016	20	

- At the second step we estimate the unknown carbon dioxide yield coefficients $K_{X_1CO_2}$ and $K_{X_2CO_2}$, and the liquid-gas transfer coefficient $k_L a$, with laboratory-scale experimental data for the specific carbon dioxide flow-rate $Q_{spCO_2}^{exp}$ for two step change of the input *D* (from D = 0.02 to D = 0.04 and from D = 0.04 to D = 0.06) and minimization of the criterion (10). The estimation was made starting from different initial value of these coefficients in their admissible ranges (0.01-40.0). The results for estimation of the coefficients $K_{X_1CO_2}$, $K_{X_2CO_2}$ and $k_L a$ are shown in the Table 4.

Experiment	k _L a	$K_{X_1CO_2}$	$K_{X_2CO_2}$	Criterion	k _L a init. cond.	$K_{X_1CO_2}$ init. cond.	$K_{X_2CO_2}$ init. cond.
1	4.005	21.406	0.1	47.765	0.01	0.01	0.01
2	4.006	21.406	0.1	47.765	0.01	0.01	40
3	4.005	21.406	0.1	47.765	0.01	40	0.01
4	4.006	21.406	0.1	47.765	0.01	40	40
5	4.006	21.407	0.1	47.765	40	40	40
6	4.006	21.406	0.1	47.765	40	40	0.01
7	4.006	21.406	0.1	47.765	40	0.01	0.01
8	4.006	21.406	0.1	47.765	40	0.01	40
9	4.006	21.406	0.1	47.765	10	10	10
10	4.005	21.406	0.1	47.765	20	20	20

Тε	ıb	le	4

The conclusion is that all coefficients have unique values which are as follows: $K_{X_2CH_4} = 17.345$, $K_{X_1CO_2} = 21.406$, $K_{X_2CO_2} = 0.1$ and $k_L a = 4.005$.

Experimental data and data from computer simulation of the model with the obtained values of the coefficients are shown on Fig. 3 (for Q_{CH_1}) and Fig. 4 (for Q_{CO_2}).



Fig. 3 Experimental (-----) and model simulation (____) data for Q_{CH_4} for step change of the input D (----) from D = 0.02 to D = 0.04 and from D = 0.04 to D = 0.06



Fig. 4 Experimental (-----) and model simulation (-----) data for Q_{CO_2} for step change of the input D (----) from D = 0.02 to D = 0.04 and from D = 0.04 to D = 0.06

Model validaton

The model validation was made with pilot-scale experimental data for step change of the input *D* from D = 0.025 to D = 0.0125. Some results are shown on Fig. 5 (for Q_{CH_4}) and Fig. 6 (for Q_{CO_2}).



Fig. 5 Experimental (-----) and model simulation (—) data for Q_{CH_4} for step change of the input D (----) from D = 0.025 to D = 0.0125



Fig. 6 Experimental (-----) and model simulation (-----) data for Q_{CO_2} for step change of the input D (----) from D = 0.025 to D = 0.0125

From Fig. 3 to Fig. 6 one may conclude that simulation data for Q_{CH_4} and Q_{CO_2} fit relatively well to the corresponding experimental data.

Conclusion

In this paper a simple mathematical model of the anaerobic digestion of WFV was developed. The following points are important, because they guarantee that our model can be useful to monitor and control the AD process:

- 1. It is based on mass-balance considerations.
- 2. Two-step identification procedure to estimate 4 model parameters was used on the base of the independence of experimental data for dynamics of the specific methane flow-rate $Q_{spCH_4}^{exp}$ and for the specific carbon dioxide flow-rate $Q_{spCO_4}^{exp}$.
- 3. Unique solutions were obtained for all coefficients.
- 4. Experiments in laboratory and pilot scales bioreactors were designed, covering a wide range of experimental conditions in order to develop and validate the model. This diversity was obtained via various organic loading rates (given by various dilution rates).
- 5. The model parameters identification was performed using laboratory experimental data for a broad set of transient conditions.
- 6. The validation of the model was performed using pilot scale experimental data.

The obtained results were used for software sensors design [14].

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