Evaluation of Polynomial Models to Predict Methane Production in Batch Mode Bioreactors *

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Abstract— The modeling and prediction of the methane production with anaerobic digestion processes is a complex task. Usually, first principles nonlinear models are used to fulfill this objective. These model structures are normally quite complex and some of the parameters may not be identifiable depending on the available measurement data. To overcome this problem, it is proposed the use of NAR and NARMA black-box models for modeling and prediction of methane production in batch bioreactors. First, simulations comparing well established nonlinear models with NAR and NARMA are proceeded and the selection of model regressors are based on the identified parameter values and the output relative errors. The domain of validity of the model and its parameters are analyzed for different concentrations of process initial conditions. To complete the validation experimental data are used to show the promising results in use these models for prediction of methane production in anaerobic batch bioreactors.

I. INTRODUCTION

The development and research on monitoring and control of renewable sources of energy has been a key factor for economical and social development due to the constant increase of electrical energy demand [1]. This demand is required for industrial expansion activity and basic infrastructural development. To mitigate the environmental impact of widely used power plants, i.e. large hydro, nuclear and thermal power plants, and at the same time enlarge electrical energy production, alternative electricity sources are needed. One of these alternatives sources is based on integrated biorefining concepts, which may use industrial organic waste, sludge generated from wastewater treatment processes or manure, to name just a few, as a substrate source for renewable electrical energy production.

Typically, the conversion of substrate into renewable energy take place inside a bioreactor. Bioreactors are used to perform biological degradation in several areas as manufacture of beverages, wastewater treatment, food and pharmaceutical industry etc. Due to these numerous possibility of applications, there are different bioreactor configurations (continuous, fed-batch, batch are the most commonly used). In this paper it will be studied a batch bioreactor, which according to [2], only defoamer and acids (or bases) are added to the substrate during the fermentation process in order to control the pH of the mixture. Due to this the volume of substrate mixture during the process remains approximately the same.

In this study the biodegradation process is anaerobic. The reference [3] points out that the anaerobic processes have an increased ability to degrade concentrated and complex substrates. Another advantage of this process is that it produces small quantity of sludge, requiring less power and in addition has the capacity to recover energy by using the combustion of produced methane. For the biochemical process to be initiated, some initial concentrations of acidogenic and methanogenic bacteria, chemical oxygen demand and volatile fatty acids are needed [4].

In this context, the process should be monitored to prevent the inhibition of the methane production. In the batch mode this can be controlled by selecting the right amount of initial concentration of the substrate and biomass, otherwise may cause an extremely low methane production. It is evident the need of accurate models to predict the production of methane based on bioreactor initial concentrations. Many models have been proposed for anaerobic digestion (AD) processes, some of them for the process cognition and monitoring like the Anaerobic Digestion Model No.1 (ADM1) [5] and some for process control and monitoring as proposed by [3]. These two models are based on the mass balance, consumption and production yield of the biochemical reaction. With that, in one hand there is the ease of understand the process because of the parameter connections with physical and biological process, but on the other hand, depending on the available process measurements, these models may not be structurally identifiable [6]. A model with no structural identifiability permits infinite combination of parameters for the same process output, loosing its advantages of physical and biological parameter relations and process prediction properties, as presented by [7], [8].

In this article black-box model structures with the objective of process monitoring and prediction of methane are studied. The first model is the NAR (nonlinear autoregressive) model and the second is the NARMA (nonlinear autoregressive moving average) model. Black-box models do not have parameters with physical and biological meaning, but the structural identifiability is guaranteed as presented in [6], becoming an interesting class of models for process monitoring and prediction. Based on the aforementioned characteristics, the NAR and NARMA structures are used to predict methane production in a lab scale bioreactor process in batch mode.

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II. ANAEROBIC DIGESTIONS MODELS

In many cases, mathematical models are extremely important in order to understand, predict and control processes dynamics. It is quite common that simplified versions of processes are used to describe real one. These models may be classified in three different classes [6]: white-box, gray-box models and black-box models. The focus of this work is the black-box modeling, since our intention is based on the initial conditions concentrations, predict the process output without the need to investigate the process internal dynamics.

Many AD models are proposed in the literature [9]. To produce data set for our investigation, the model proposed by [3] is selected and adapted to a batch mode process (the choose of a bioreactor in batch mode will be explained in Section IV-C). In a batch mode, all the kinetics are based on the initial concentration of the bioreactor at time zero until the bioreactions end and the process stabilizes. To begin this section, the AD model proposed by [3] is presented and to conclude NAR and NARMA black-box models are presented in the following.

A. AD Macro Model

To study the anaerobic digestion process the nonlinear model proposed by [3] is used. This model is based on the mass balance modeling of the process and have been applied to many different AD processes. Campestrini et al. [10] reported the application of this model with success to the biogas production in batch bioreactors. The model is represented in a simplified version for a batch mode in (1).

$$
\begin{cases}
\dot{x}_1(t) = \nu_1(s_1(t))x_1(t) \\
\dot{x}_2(t) = \nu_2(s_2(t))x_2(t) \\
\dot{s}_1(t) = -k_1\nu_1(s_1(t))x_1(t) \\
\dot{s}_2(t) = -k_2\nu_1(s_1(t))x_1(t) - k_3\nu_2(s_2(t))x_2(t)
\end{cases}
\tag{1}
$$

where $x_1(t)$ and $x_2(t)$ are the concentration of acidogenic bacteria and concentration of methanogenic bacteria in mg/L, respectively; $s_1(t)$ and $s_2(t)$ represent the concentration of oxygen demand (COD) in mg/L and the concentration of volatile fatty acids (VFA) in $mmol/L$, respectively; k_1 ($mg\ COD/mg\ x_1$) and k_2 ($mmol\ VFA/mg\ x_1$) are the performance coefficients of COD degradation and production and k_3 (VFA $mmol/mg\ x_2$) is the yield coefficient of consumption of volatile fatty acids; ν_1 and ν_2 are specific microbial growth rates given by (2) and (3). Normally, (3) is expressed with a Haldane's law, but as presented by [10], when the substrate concentration is low and no inhibition VFA is observed, for model simplification reasons, is better than be used a Monod's law equation.

$$
\nu_1(s_1(t)) = \mu_{m1}\frac{s_1(t)}{K_{s1} + s_1(t)}
\tag{2}
$$

$$
\nu_2(s_2(t)) = \mu_{m2}\frac{s_2(t)}{K_{s2} + s_2(t)}
\tag{3}
$$

where K_{s1} (mg/L) and K_{s2} ($mmol/L$) are half saturation parameters associated with s_1 and s_2.

In addition, the process output is represented by $q_M(t)$, which is the methane flow rate given by

$$
q_M = k_6\nu_2(s_2(t))x_2(t),
\tag{4}
$$

where k_6 ($mmol/g$) is the yield coefficient for the production of methane. To obtain the total amount of produced methane, the (4) is integrated from process beginning until process ends.

B. NAR and NARMA Models

The NAR (nonlinear autoregressive) and NARMA (nonlinear autoregressive moving average) models can be polynomial models that have been used in many different fields as in research on lithium-ion battery remaining useful life estimation [11], on retail sales forecasting [12], on wideband simulation of nonlinear power amplifiers [13], application to blood flow/pressure data [14] and so on. Polynomial models have the advantages of being simple in its structure, resulting in lower computation efforts for its simulation and parameters identification. A valuable aspect of these models is that the models have structural identifiability, which ensure a set of parameters for a determined and predict output dynamics.

The nonlinear autoregressive models are discrete in time and are used to predict the system output from their previous output values, as (5). Although the models are nonlinear, its parameter relations remain linear, in this manner it can be implemented the parameter identification using the Least Square (LS) method.

$$
y[k] = F^l[y[k-1], \ldots, y[k-k_y]]
\tag{5}
$$

where k is the discrete time, F is a polynomial function of $y[k]$ with degree of nonlinearity l, k_y is the maximum output delay.

NARMA models consist of a "NAR" part, the autoregressive nonlinear, and a "MA" part, the moving average. Similar to the NAR models, NARMA models also are discrete in time and use the previous output values to predict the current output. The difference between NAR and NARMA models is that the NARMA models consider its regressors the moving average factor to further approximate the system model, as can be seen in (6). Unlike the NAR models, NARMA models are characterized by pseudolineares parameters that prevent the direct application of the LS method, requiring the Extended Least Squares (ELS) method to be implemented.

$$
y[k] = F^l[y[k-1], \ldots, y[k-k_y], e[k-1], \ldots, e[k-k_e]]
\tag{6}
$$

where k_e is the maximum noise delay.

III. METHODS FOR SYSTEM IDENTIFICATION

System Identification (SI) is a procedure to explain the cause and effect between the input data and/or initial conditions and output data of a system through a mathematical model [6], [15], [16]. Firstly the parameters estimation is

performed, and subsequently, the model validation is evaluated with a cross-validation, wherein it is used different data sets to analyze the model behavior in different conditions.

The parameter estimation procedure can be executed with various different algorithms. Due to the simplicity of implementation, the LS [17] and ELS [6] methods for the NAR and NARMA models, respectively, these methods have been chosen in this study.

Is possible to apply the method of Least Square using the model:

$$M_{LS} : Y[k] = \Phi^T[k]\Theta \tag{7}$$

where Φ is the group of regressors output vector and θ is the group of parameter vector.

And the Extended Least Squares method M_{ELS} with the model:

$$M_{ELS} : Y[k] = \Phi^T[k]\eta + e[k] \tag{8}$$

where η is the group of parameter vector and e it is the noise.

The LS and ELS algorithm have the objective to minimize the squared difference between the actual ($y[k]$) and estimated ($\hat{y}[k]$) outputs. To evaluate this difference the relative error value (RE) is computed as follows:

$$RE = \frac{\|Y - \hat{Y}\|_2}{\|Y\|_2}.100 \tag{9}$$

The best model is the one that has the lowest RE possible, since it implies that the difference between the real output and the estimated output is also lower, as provided for under LS and ELS methods.

IV. IMPLEMENTATION AND RESULTS

In this section simulations are carried out to obtain the model regressors for NAR and NARMA. The parameters identification procedure is applied and the complete model (regressors + parameters) is validated. First, the results based on simulation are given and second, the results based on experimental data are presented and analyzed. In order to have a metric of comparison, a simulation using the anaerobic digestion macro model (1) is performed and the output data set is considered the control group (CG) data set. The parameter values for the CG data set are from [10] where $\mu_{m1} = 4.2912 \times 10^1$, $\mu_{m2} = 2.6493 \times 10^0$, $K_{s1} = 1.3065 \times 10^1$, $K_{s2} = 5.7127 \times 10^2$, $k_1 = 3.1204 \times 10^{-1}$, $k_2 = 6.2776 \times 10^{-2}$, $k_3 = 3.1473 \times 10^0$ and $k_6 = 2.7862 \times 10^2$ and the initial conditions of the CG are $x_1(0) = 0.2mg/L$, $x_2(0) = 0.8mg/L$, $s_1(0) = 74mg/L$ and $s_2(0) = 93mmol/L$.

Therefore, the investigation is performed in the following way. First the value of an IC, for example, $x_1(0)$, is changed while keeping the others ($x_2(0)$, $s_1(0)$, $s_2(0)$) fixed. The simulation is performed and the output data set is compared with the CG output data and the relative error (RE) is computed with (9). After, another IC is selected and changed, for instance x_2, keeping the others fixed and so on, until the tests of the four IC are executed.

Besides, in Section IV-C, to present the applicability of the black-box model structures for the prediction of the methane

production, a parameter identification and a model validation using experimental data is carried out. The experimental data is obtained from a laboratory scale plant where it is used two glass bottles with a capacity of one liter each. The biogas production is measured by an apparatus comprising a gas collector (U-shaped), an optical sensor, an expanded polystyrene bead, and an electronic circuit which records the stream and is able to calculate the volume of gas generated by bioreactor. To maintain a constant temperature (35°C, temperature suitable for the production of biogas), the experiment is conducted in a bacteriological incubator. Each of the $420mL$ bottles owned substrate (formed by sludge from the Wastewater Treatment Station of Ecological Cooperative of Vale do Caí - Ecocitrus, in Rio Grande do Sul, Brazil) and $180mL$ inoculum. The biogas measurements are taken four times a day (every 6 hours) for a total period of 37 days, resulting in 148 samples.

A. Selection of NAR and NARMA Regressors

It follows that the selection of the regressors for the NAR and NARMA model is important for the black-box models capture the main dynamics of the process. The NAR and NARMA models are selected from a test combining all possible regressors order of 3 to power 3. This combination results in 19 regressors for NAR model and 22 regressors for NARMA model. Furthermore, the parameter identification is performed for all combination of regressors. Those parameters that have contribution to model dynamics zero or values extremely close to zero are eliminated, which means that these regressors do not impact in the process output dynamic. With that the size of the regressors can be reduced. It is important to highlight that if a regressor is suppressed and the RE of the identification procedure increases, this regressor is central for a model and must be used even if the identified parameter is close to zero. The result of this exhaustive test is presented in (10) for NAR regressors and in (11) for NARMA regressors.

$$\Phi_{NAR}[k] = \begin{bmatrix} -(y[k-1]) \\ -(y[k-1]^2) \\ -(y[k-2]) \\ -(y[k-1]y[k-2]) \\ -(y[k-1]y[k-3]) \\ -(y[k-2]^2) \\ -(y[k-2]y[k-3]) \\ -(y[k-3]) \\ -(y[k-3]^2) \end{bmatrix} \tag{10}$$

$$\Phi_{NARMA}[k] = \begin{bmatrix} -(y[k-1]) \\ -(y[k-1]^2) \\ -(y[k-2]) \\ -(y[k-1]y[k-2]) \\ -(y[k-1]y[k-3]) \\ -(y[k-2]^2) \\ -(y[k-2]y[k-3]) \\ -(y[k-3]) \\ -(y[k-3]^2) \\ e[k-1] \\ e[k-2] \\ e[k-3] \end{bmatrix} \tag{11}$$

In NAR case the model is reduced from the size of 19 to 9 regressors and for NARMA case from the size of 22 to 12 regressors. Note that the model (1) has 8 parameters and the models we have proposed 9 and 12, to NAR and NARMA, respectively. Although our models present more parameters, we ensure that all of these are identifiable, which is not necessarily true of the gray-box model proposed in the literature.

B. Simulation Results

The process dynamics presented in Figs. 1, 2 and 3 are simulation obtained from the AD macro model represented by (1). Figure 1 presents the acidogenic (x_1) and methanogenic (x_2) bacteria dynamics, Fig. 2 shows the methane production represented by (4) and in Fig. 3 the total amount of the methane production in one batch (i.e. the sum of the output concentration q_M from time zero until end time) is presented. For comparison purposes and to
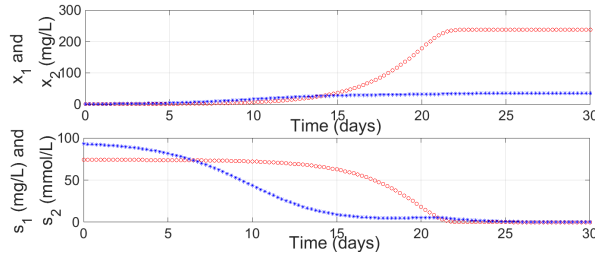

Fig. 1: Process dynamics simulation of acidogenic bacteria concentrations x_1 with circle markers and methanogenic x_2 in asterisk markers for model (1)
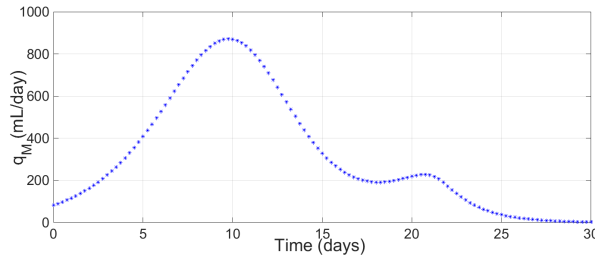

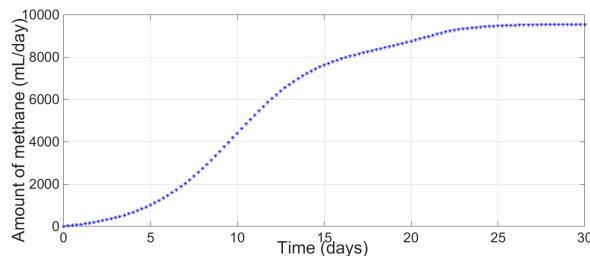Fig. 2: Process methane production q_M for model (1)

Fig. 3: Total amount of methane produced, simulation of model (1)

demonstrate the influence of the initial conditions into the process output, Fig. 4 presents the methane dynamic outputs when different initial conditions are applied to model (1)

where the square marker is the output to $x_1 = 0.0005 mg/L$, the cross marker is the output to $x_2 = 1.5mg/L$, the hexagon marker are the output to $s_1 = 210mg/L$, the x marker are the output to $s_2 = 190mmol/L$ and the circle marker is the output to CG. The parameter identification procedure has
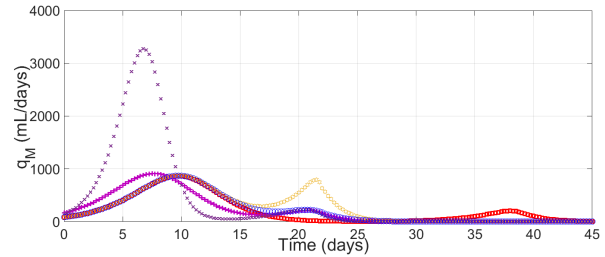

Fig. 4: System output dynamics for different IC and for the Control Group, the square marker to x_1, the cross marker to x_2, the hexagon marker to s_1, the x marker to s_2 and the circle marker to CG.

been carried out for the NAR and NARMA models with the regressors presented in (10) and (11), respectively. This identified parameters are presented in Table I.

Concerning the study of the validity domain for NAR and NARMA models, the initial concentration of the acidogenic bacteria (x_1) is changed between 0.0005 and 2.2505, every $0.15mg/L$. This results in 16 different simulation for each different IC. It can be seen, in Figure 5, that the RE of process output remains practically constant, suggesting that the model is valid for all initial conditions tested. In addition, it is also clear that the lowest RE to the NAR model for IC x_1 are $1.9505mg/L$ with a RE of 0.28% and $0.1505mg/L$ with RE of 0.21% for NARMA model.
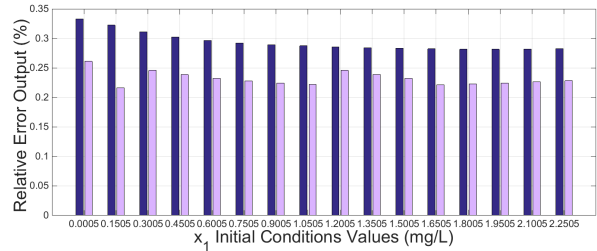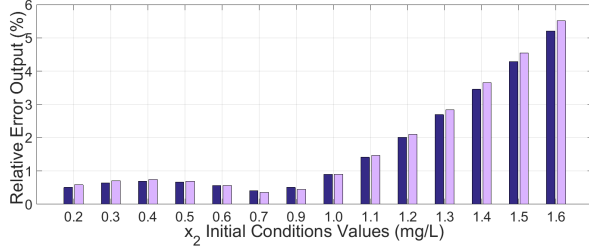

Fig. 5: Relative errors to each change of 0.15 mg/L for the initial condition x_1 with the NAR (dark bar) and NARMA (light bar) models
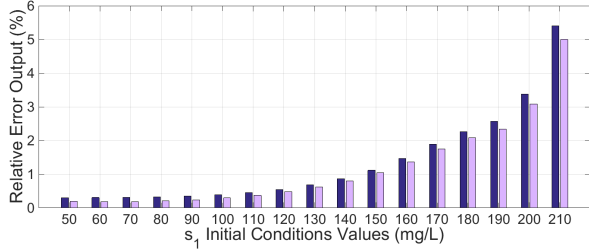
Similarly, the initial concentration of methanogenic bacteria (x_2) is changed between 0.2 and 1.5, every $0.1mg/L$. For each value a new simulation is proceeded and the RE of the output is computed. In Fig. 6 the smallest RE is found when $x_2(0) = 0.7mg/L$ for both models with 0.41% for NAR and 0.36% for NARMA. Note that after this value the RE increases fast until the RE equal to six where it is the maximum value admitted for this study to be in the validation domain. So, it can be seen that the model is valid, mainly between the range of values from 0.2 to $0.9mg/L$. Likewise, the initial conditions of chemical oxygen demand (s_1) is tested with values between 50 and 210 every $10mg/L$. Once more, simulations of each $s_1(0)$ have been carried out and its RE output between the simulation and CG are represented

TABLE I: Parameters values of the control group

Control Group	θ_1	θ_2	θ_3	θ_4	θ_5	θ_6	θ_7	θ_8	θ_9	θ_{10}	θ_{11}	θ_{12}
NAR	-2.4491	0.0175	1.8678	-0.0510	0.0156	0.0313	-0.0108	-0.4187	-0.0026	−	−	−
NARMA	-2.5288	0.0185	2.0219	-0.0536	0.0169	0.0323	-0.0113	-0.4942	-0.0027	0.8805	-0.2868	0.1466

Fig. 6: Relative errors to each change of 0.10 mg/L for the initial condition x_2 with the (dark bar) and NARMA (light bar) models

in Fig. 7. The RE presented shows that the error grows when larger $s_1(0)$ is presumed and the lowest RE corresponds to the value of $50mg/L$ with 0.30% for NAR model and the value of $60mg/L$ with for 0.18% NARMA model. Moreover, it is clear that the validity domain of the NAR models and NARMA for this IC will 50 to $90mg/L$ which are the values on which the smallest RE were obtained.

Fig. 7: Relative errors to each change of $10mg/L$ for the initial condition s_1 with the (dark bar) and NARMA (light bar) models

Finally, the validation domain study for the initial concentration of chemical oxygen demand (s_2) are proceeded. The investigation starts with concentration of 30 and goes until 190 $mmol/L$ with a step of 10 $mmol/L$. The profile of the RE is quite similar to the profile of s_1 as can be seen in Fig. 8. The smallest errors are 0.31% and 0.21% for the value of $90mg/L$ for both models. Low RE values for the IC of 30 to $70mmol/L$ suggest that this is valid domain of NAR and NARMA models.
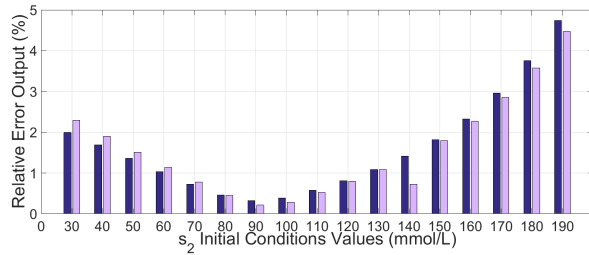
Fig. 8: Relative errors to each change (each $10mmol/L$) for the initial condition s_2 with the (dark bar) and NARMA (light bar) models

With the exception of x_1, as the value of the IC increase

TABLE II: Total amount of methane (in mL/day) for the best IC and CG for NAR, NARMA and first principles models

IC	Total amount of Methane Production		
	NAR	NARMA	First Principles Model (1)
$x_1(0)$	9.5400×10^3	9.5369×10^3	9.5509×10^3 and 9.5509×10^3
$x_2(0)$	1.0585×10^4	1.0586×10^4	9.5509×10^3 and 9.5509×10^3
$s_1(0)$	9.1136×10^3	9.2908×10^3	9.1235×10^3 and 9.3016×10^3
$s_2(0)$	9.2755×10^3	9.2743×10^3	9.2853×10^3 and 9.2853×10^3
CG	9.5407×10^3	9.5399×10^3	9.5509×10^3 and 9.5509×10^3

the RE also increases for x_2, s_1 and s_2. Another fact that stands out is that minors RE x_1, s_1 and s_2 to the NAR model are smaller than the RE found for the CG, the same goes for the IC s_1 for NARMA model also in relation to the CG.

From the evaluation of our results it is possible to note that the NAR and NARMA models are quite robust, mainly to the change in the IC x_1, as the RE for the variation of its initial conditions have a small effect in the process RE.

The Table II shows the simulation results for the total methane produced using NAR model (10), NARMA model (11) and the first principles model (1). To compute these values, the IC in which the minor RE were obtained are used. So, the IC to the NAR model are $x_1(0) = 1.9505mg/L$, $x_2(0) = 0.7mg/L$, $s_1(0) = 50mg/L$ and $s_2(0) = 90mmol/L$ and for the NARMA model are $x_1(0) = 0.1505mg/L$, $x_2(0) = 0.7mg/L$, $s_1(0) = 60mg/L$ e $s_2(0) = 90mmol/L$. For the macro model (1) the IC are the same as the NAR and the NARMA cases.

Analyzing the total amount of methane production obtained for each different IC for NAR and NARMA cases, Table II, the maximum difference is 10.95% to $x_2(0)$ and the smaller difference is 4.48% to $s_1(0)$ for the NAR case. For NARMA model the maximum difference is 10.95% to $x_2(0)$ and the smallest difference is 2.79% to $s_2(0)$. For the first principles model (1) with NAR IC, such differences are much smaller: with maximum of 4.47% and minimum of 2.78% for NARMA IC. Note that modifying the IC x_1 and x_2 the same methane production than CG is obtained.

C. Experimental Results

The 148 samples obtained from laboratory scale experiments mentioned above were divided into two groups: i) half of the data is used to perform the parameter identification; ii) other half for cross-validation of NAR (10) and NARMA (11) models.

In the same way as previous sections, the methods of LS and ELS for NAR and NARMA models are used for the parameter identification procedure with the experimental data. Note that the regressors are the same as in the early results. In Table III the estimated parameters are presented.

The RE of experimental data and NAR and NARMA the models are RE 16.82% and 24.18%, respectively. The Fig. 9 highlights the differences between the estimated output
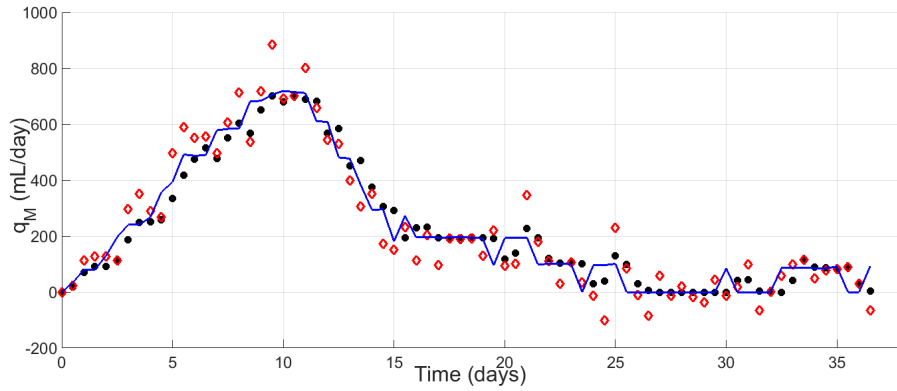
Fig. 9: Experimental methane concentration measurements (continuous line) and the cross-validation using NAR (dot marker) and NARMA (diamond marker) models.

TABLE III: Found parameters (θ) for NAR e NARMA models using experimental data.

$Parameters$	NAR	NARMA
θ_1	-0.5663	-0.6416
θ_2	0.0001	0.0001
θ_3	-0.5848	-1.2950
θ_4	-0.0036	-0.0026
θ_5	0.0013	0.0016
θ_6	0.0007	0.0003
θ_7	0.0028	0.0027
θ_8	0.1376	0.9437
θ_9	-0.0020	-0.0021
θ_{10}	-	-0.1329
θ_{11}	-	-0.8157
θ_{12}	-	0.4279

and real process measurement in the cross-validation process. Although RE is relatively high, it is important to highlight that experimental data is quite noisy due to the measurement method and the sensor accuracy, which may lead to major discrepancies between real and estimated values from the models. However, the model predictions for the dynamic process is extremely similar. Note that the parameters obtained for the real data have similarities between the NAR and NARMA models.

V. CONCLUSIONS

In this paper we have studied the development of black-box models for monitoring and prediction of methane production in anaerobic batch bioreactors. The validation of these models with a well established first principles model has been carried out, showing that the proposed model structures have a wide domain of validity. Experimental data from a lab scale methane production reactor have been used to test the models. Both models proposed provide appropriate performance in predicting the bioreactort's output, particularly taking into account the scarcity of data available for identification. Thus, the choice between using one or other model will depend on the focus of research. The NARMA model uses more parameters, but requires data with process information to work properly, since the NAR model, as is more restricted, do not need much. Thus, one should analyze the process studied to make the choice of model. Future research will focus on further experimental testing of the proposed approach.

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