Development and pilot-scale validation of a fuzzy-logic control system for optimization of methane production in fixed-bed reactors

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Elsevier Editorial System(tm) for Journal of Process Control

Manuscript Draft

Manuscript Number:

Title: Development and pilot-scale validation of a fuzzy-logic control system for optimization of methane production in fixed-bed reactors

Article Type: Research Paper

Keywords: Anaerobic digestion, bio-methane, fixed-bed reactor, fuzzy-logic control, optimization, winery wastewater

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Abstract: The objective of this study was to develop an advanced control system for optimizing the performance of fixed-bed anaerobic reactors. The controller aimed at maximizing the bio-methane production whilst controlling the volatile fatty acids content in the effluent. For this purpose, a fuzzy-logic controller was developed, tuned and validated in an anaerobic fixed-bed reactor at pilot scale (350 litres) treating raw winery wastewater. The results showed that the controller was able to adequately optimize the process performance, maximizing the methane production, with an average methane yield of about 0.29 LCH4 g-1 COD. On the other hand, the controller maintained the volatile fatty acids content in the effluent close to the established maximum limit (750 mg COD L-1). The outcomes of this study are expected to facilitate plant engineers to establish an optimal control strategy that enables an adequate process performance with the maximum bio-methane productivity.

Dear Editor,

Attached you will find the manuscript entitled "**Development and pilot-scale validation of a fuzzy-logic control system for optimization of methane production in fixed-bed reactors**" submitted for consideration as a research paper in Journal of Process Control. All the authors mutually agree for submitting this manuscript to Journal of Process Control. We confirm that it is the original work and that the work presented has not been submitted earlier to this Journal.

The main objective of this work was to develop an advanced control system for optimizing the performance of fixed-bed anaerobic reactors. The controller aimed at maximizing the bio-methane production whilst controlling the volatile fatty acids content in the effluent. To this aim, a fuzzy-logic controller was developed, tuned and validated in an anaerobic fixed-bed reactor at pilot scale treating raw winery wastewater. The outcomes of this study are expected to facilitate plant engineers to establish an optimal control strategy that enables an adequate process performance with the maximum bio-methane productivity.

The important findings that must be highlighted are:

- Simulation results show that the proposed controller is capable to achieve great process performances even when operating at high VFA concentrations.

- The controller was sufficient to capture the dynamics of the process around the corresponding set point.

- Pilot results showed the potential of this control approach to maintain the process working properly under similar conditions to the ones expected at full-scale plants.

Yours sincerely,

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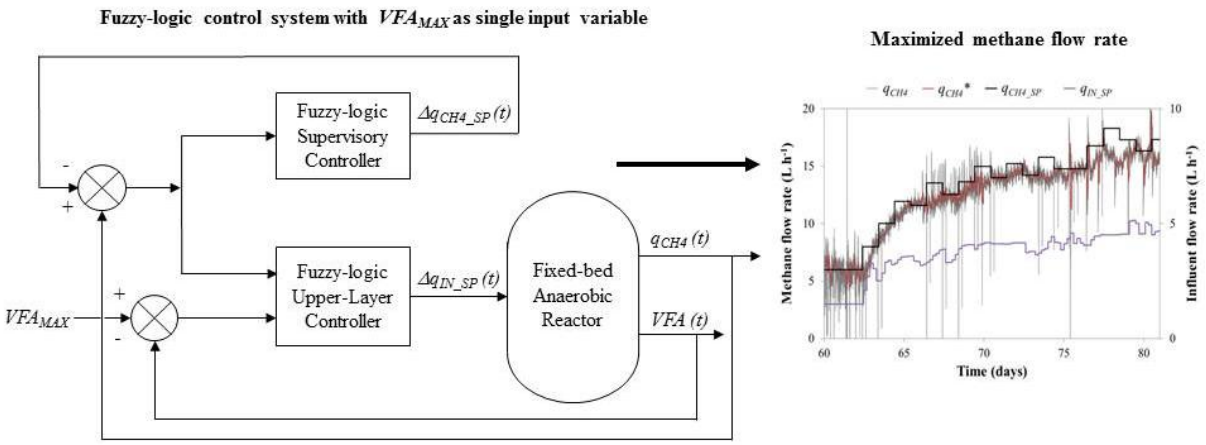
Graphical abstract



Highlights

- A fuzzy-logic control system for optimizing the methane production was proposed
- The controller was developed, tuned and validated at a 350 L pilot-scale system
- The controller aimed to maximize methane production whilst controlling VFA contents
- Methane yields up to 0.29 L CH_4 g^{-1} COD were achieved when running the controller

Development and pilot-scale validation of a fuzzy-logic control system for optimization of methane production in fixed-bed reactors

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Abstract

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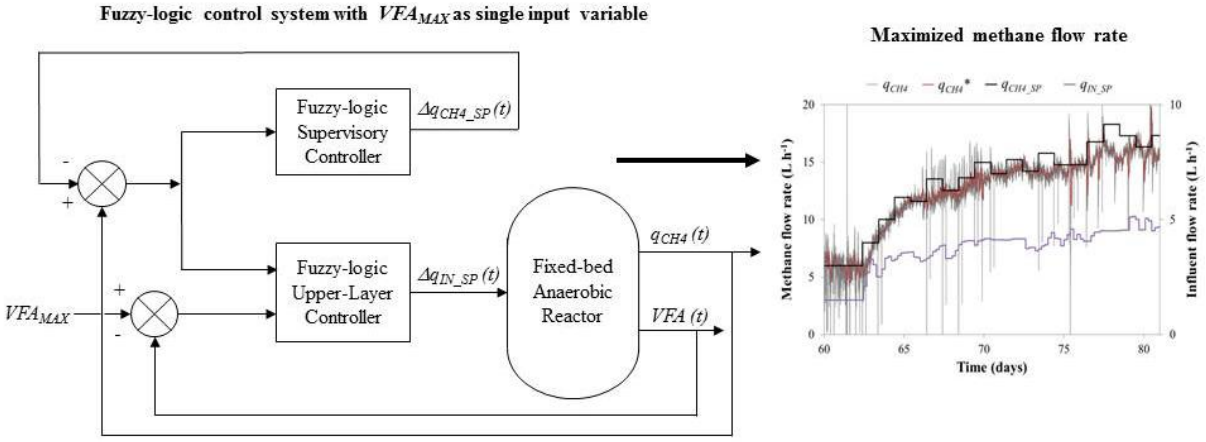

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1. Introduction

Nowadays, a major issue to overcome in order to achieve a global sustainable development is our dependency on fossil fuels for electricity production, which represents up to 80 % of the global energy consumption [1]. Therefore, one of the main challenges of this century is to develop new competitive sources of renewable energy, capable of replacing fossil fuels with a minimum impact on both environment and society [2]. In this context, alternative energy sources must be pursued [3]. Bio-methane production from anaerobic digestion (AD) of waste represents a promising option that can be considered as carbon neutral due to its net balance of greenhouse gases emissions.

Due to the high methane productivities that can be achieved by high-rate anaerobic reactors, a huge effort is currently being put on the study of systems such as up-flow anaerobic sludge blanket (UASB), expanded granular sludge blanket (EGSB), anaerobic membrane bioreactor (AnMBR) or fixed-bed bioreactor [4]. In these reactors, the biomass is self-immobilized, allowing uncoupling the hydraulic retention time (HRT) and the solid retention time (SRT).

However, the complexity and the diversity of the phenomena occurring in high-rate anaerobic reactors have delayed the understanding, and consequently the proper control, of this AD process. Due to the large number of factors that affect anaerobic processes, the selection of proper monitoring indicators and the development of advanced control systems

are crucial for a successful optimization of the process performance [5,6].

Biogas composition and production rate are the most commonly used variables acting as indicators of the process performance during AD. In addition, the methane yield (Y_{CH4}), which is usually defined as the amount of methane produced per unit of organic matter removed, is also used as an indirect parameter for evaluating the performance of anaerobic processes [7,8]. Nevertheless, these indicators can be insufficient to evaluate the overall process performance. This is because they usually indicate too late disturbances affecting the process, when there is no possible action to recover it immediately. To avoid this issue, the concentration of volatile fatty acids (VFA) has been proved to be an adequate state indicator for monitoring AD processes [9]. VFAs are main intermediate metabolites in AD and therefore, monitoring their concentration can be a useful tool for process diagnosis (*e.g.* to detect AD imbalances). Moreover, as this variable can be easily on-line monitored, for instance by means of titrimetric sensors, it gives a much faster and more reliable information than other common indicators applied for AD monitoring, such as pH, alkalinity, gas composition or gas production [10–14].

Many different alternatives, such as classical Proportional-Integral-Derivative (PID) control, fuzzy systems, neuron networks or model-based systems, have been applied for controlling AD process [15]. Amon these strategies, fuzzy-logic control has the main advantage of being applicable to control non-linear systems, such as AD. A fuzzy-logic controller [16] is able to optimize different types of processes under dynamic conditions by applying valuable expert knowledge [17–20]. Moreover, fuzzy-logic controllers do not require large amounts of data and/or rigorous mathematical models, thus allowing a much simpler calibration of the controller. In addition, these control systems allow the development

of multiple-input-multiple-output control schemes. Hence, it can be stated that fuzzy logic is a powerful tool for controlling anaerobic fixed-film reactors [21]. Therefore, fuzzy-logic control has been widely implemented in wastewater treatment over the last decades and has been successfully featured in several AD applications [22–26]. As listed in Jimenez et al. [15], different applications of fuzzy-logic control systems for AD control can be found in the literature. Taking some examples, Puñal et al [27] developed a PI-based fuzzy-logic controller which used the dilution rate as manipulated variable to control the concentration of VFAs in the effluent. In addition, Murnleitner et al. [28] applied fuzzy theory to avoid overloading of AD reactors. Recently, Robles et al. [29] demonstrated the suitability of fuzzy-logic systems for controlling the methane production in AD reactors using the methane flow rate and the VFA concentration as input variables. Nevertheless, only one study has been carried out so far for optimization of AD processes using fuzzy logic. Carlos-Hernandez et al. [30] proposed a fuzzy supervisory controller to optimize the AD performance by controlling alkali addition and the dilution rate. To the knowledge of the authors, no other study has been carried out to apply fuzzy-logic control systems for AD optimization.

Considering the aforementioned information, the main objective of this study was to develop an advanced control system for optimizing the methane production in fixed-bed anaerobic reactors. To this purpose, a fuzzy-logic system consisting of a supervisory controller to determine the set-point of methane flow rate and an upper-layer controller to define the inflow of substrate into the reactor was first developed by simulation and then validated in a 350 L pilot-scale fixed-bed anaerobic reactor treating industrial winery wastewater. The proposed controller aimed at maximizing bio-methane production whilst controlling the VFA concentration in the effluent. The main novelty of this study lies not only in developing a controller for optimizing the operation of fixed-bed anaerobic reactors, but

also in its validation under specific conditions that were similar to those found in full-scale plants.

2. Materials and methods

2.1. Pilot plant description and operation

Figure 1 shows the flow diagram and the instrumentation of the continuous fixed-bed anaerobic reactor used in this study. The plant had a total volume of 358 L. The support media (Cloisonyl: 180 m^2 m^{-3} specific surface) filled 34 L, leaving 324 L as effective volume. The anaerobic reactor was jacketed and connected to a water heating system for temperature control. Moreover, the plant was equipped with a pH control by feeding NaOH (30 %) to the system when necessary. The pH set-point was set at 7.2.

The plant was fed with industrial winery wastewater from local cellars located in the area of Narbonne, France. Table 1 shows the main average characteristics of the influent wastewater during the experimental period. The wastewater was stored in a feeding tank of 27 m^3 that was connected to a dilution system of 0.2 m^3. The main aim of this dilution system was to allow testing different organic loading rates (OLRs) in the plant. In the reactor, a portion of the mixed liquor was recycled from the bottom to the top for both improving the mixing conditions and favouring the stripping of the produced gases from the liquid phase. The influent wastewater was mixed with the recycled mixed liquor and then introduced at the top of the reactor. The recycling flow rate was controlled manually at approximately 550 L h^{-1}. The pilot plant was operated at a controlled temperature of 35 ºC.

144 *2.2. Pilot plant instrumentation, automation and control*

145

146 As shown in Figure 1, the plant was fully automated and instrumented. The on-line
147 equipment consisted of: one pH transmitter and one conductivity-temperature transmitter
148 located in the recycling pipe; one temperature transmitter in the anaerobic reactor; one gas
149 pressure transmitter in the head-space of the anaerobic reactor; two flow-rate transmitters (one
150 for the recycling pump and one for the feed pump); one gas flow-rate transmitter
151 (electromagnetic floater-based sensor) and one on-line CH_4/CO_2 sensor (Ultramat 22P
152 Siemens), both located in the biogas discharge pipeline; and one on-line titrimetric sensor
153 (Anaerobic Control Analyser AnaSense®, AppliTek S.L.) for the measurement of total VFA
154 and alkalinity in the reactor. On the other hand, a linear relationship (R^2 above 0.8) was
155 observed between the experimentally determined COD concentration in the effluent and the
156 VFA measurement from the on-line titrimetric sensor. Therefore, besides its experimental
157 determination, the COD concentration in the effluent was also predicted in real time from the
158 continuously on-line monitored VFA concentration.

159

160 The plant also included several lower-layer control loops, which consisted of classical
161 PIDs and on-off controllers, in order to control the influent flow rate, the temperature, and the
162 pH. The on-line sensors and the automatic equipment were connected to a network system
163 that included several transmitters, an input/output device, and a PC that was in charge of the
164 data acquisition and allowed performing multi-parameter control. The input/output device was
165 managed by a software developed at INRA-LBE. The main aim of this software was to carry
166 out data logging, advanced control action calculations and process supervision by using
167 Matlab® routines.

168

169 *2.3. Sampling and off-line measurements*

170

171 Besides the on-line process monitoring, samples from influent, effluent and biogas
172 streams were collected once per day. From both influent and effluent, the chemical oxygen
173 demand (COD) was determined twice/three times a week, whilst the composition of VFAs,
174 *i.e.* acetate (C2), propionate (C3), iso-butyrate and butyrate (iC4 and C4), and iso-valerate and
175 valerate (iC5 and C5), were analyzed once per day. Biogas composition (CH_4, CO_2, O_2, H_2S,
176 and N_2) was determined three times a week.

177

178 The COD was determined by the spectrophotometric micro-method (Tube Test MR,
179 AQUALYTIC®), according to Standard Methods [31]. The composition of VFAs was
180 determined by liquid chromatography (Perkin Elmer®, Clarus 580 Liquid Chromatograph).
181 0.5 mL of sample was introduced into a vial with the same amount of standard (1 g of ethyl-2-
182 butiric acid in 1 L of distilled water, acidified to 5 % (v/v) with H_3PO_4). Moreover, a control
183 solution containing the VFAs to be determined (1.078 g C2 L^{-1}; 1.022 g C3 L^{-1}; 1.068 g iC4
184 L^{-1}; 1.111 g C4 L^{-1}; 1.079 g iC5 L^{-1}; and 1.151 g C5 L^{-1}) was also analysed. The composition
185 of gas was measured using a gas chromatograph equipped with a thermic conductivity
186 detector (GC-TCD, Perkin Elmer®, Clarus 480 Gas Chromatograph). 0.2 mL of biogas were
187 collected by a gas-tight syringe and injected into the GC, which was maintained at
188 temperature of 65 ºC and pressure of 2.48 bars. The GC consisted of two columns: one
189 RtUBond (30m x 0.32mm x 10µm) allowing the separation of CO_2 and H_2S; and one Rt-
190 Molvieve 5A (30m x 0.32mm x 30µm) allowing the separation of the H_2, O_2, N_2 and CH_4.
191 The carrier gas was helium at a flow-rate of 4 mL min^{-1}.

192

193 *2.4. Control system description*

Figure 2 shows a block diagram of the proposed fuzzy-logic controller for optimization of the performance of a fixed-bed anaerobic reactor. For that purpose the controller aimed at maximizing the bio-methane production whilst controlling the VFA content in the effluent. The proposed control structure consisted of: (i) an upper-layer controller that manipulated the influent liquid flow to maintain the methane gas flow rate close to a given set-point; and (ii) a supervisory controller that maximized the set-point of the methane flow rate to be controlled by the upper-layer controller.

The methane flow was calculated by means of the methane concentration in the gas phase and the measured biogas flow. The methane flow was corrected to account for the dependence of the biogas density on the volumetric flow. Thus, taking into account the on-line information from the biogas composition (% CH_4 and % CO_2) and the measured biogas flow ($G_{MEASURED}$), the methane flow (q_{CH4}) was calculated by Equation 1.

$$q_{CH\,4} = G_{CORRECTED} \cdot {\%CH_4}/{100} \qquad \text{(Eq. 1)}$$

where:

- $G_{CORRECTED} = G_{MEASURED} \cdot frho$ (Eq. 2)
- $$frho = \sqrt{\frac{rho_{AIR}}{\left(rho_{CH_4} \cdot \%CH_4 + rho_{CO_2} \cdot \%CO_2 + rho_{N_2} \cdot (100 - \%CH_4 - \%CO_2)\right)/100}} \qquad \text{(Eq. 3)}$$
- rho_{AIR}: volumetric weight of air (1.2930 kg m^{-3}),
- rho_{CH_4} : volumetric weight of CH_4 (0.7168 kg m^{-3}),
- rho_{CO_2} : volumetric weight of CO_2 (1.9768 kg m^{-3}),
- rho_{N_2} : volumetric weight of N_2 (1.2505 kg m^{-3}).

217

218 A 2h-moving average value for q_{CH_4} (q_{CH_4}*) was applied to the raw data to reduce the
219 noise from the measurements. Similarly, a 2h-moving average value (*VFA**) was also
220 considered for the effluent VFA concentration to take into account the sampling time of the
221 on-line titrimetric sensor. Both moving average values were also selected on the basis of AD
222 process dynamics through experimental observations. The control time of the upper-layer
223 controller was set to 5 h and the control time of the supervisory controller was set to 24 h. The
224 fuzzy-logic controller was defined following the Takagi-Sugeno structure.

225

226 *2.4.1. Upper-layer controller description*

227

228 The upper-layer controller determined the variation in the set-point of the influent flow
229 rate (Δq_{IN_SP}) to be applied to the corresponding PID controller on the basis of three inputs:
230 the error in the methane flow rate (eq_{CH4}; Equation 4), the variation in the error of the
231 methane flow rate (Δeq_{CH4}; Equation 5) and the difference between a maximum VFA
232 concentration (VFA_{MAX}) and the VFA content in the effluent (*dVFA*; Equation 6).

233

234 $$eq_{CH_4}(t) = q_{CH_4}(t) - q_{CH_4\_SP}(t) \quad \text{(Eq. 4)}$$

235 where:

236 - $eq_{CH_4}(t)$ *:* error in the methane flow rate at a given time *t*,

237 - $q_{CH_4}(t)$ *:* measured methane flow rate at a given time *t*,

238 - $q_{CH_4\_SP}(t)$: methane flow rate set-point at a given time *t*.

239

$$\Delta eq_{CH_4}(t) = \left|eq_{CH_4}(t)\right| - \delta \cdot \left|eq_{CH_4}(t-1)\right| \quad \text{(Eq. 5)}$$

where:

- $\Delta eq_{CH_4}(t)$: variation in the error of the methane flow rate at a given time *t*,
- $\left|eq_{CH_4}(t)\right|$: absolute value of the error in the methane flow rate at a given time *t*,
- δ: modifying algebraic factor (Equation 7),
- $\left|eq_{CH_4}(t-1)\right|$: absolute value of the error in the methane flow rate at the previous control action.

$$dVFA(t) = VFA_{MAX} - VFA(t) \quad \text{(Eq. 6)}$$

where:

- $dVFA(t)$: difference between VFA_{MAX} and the VFA content in the effluent at a given time *t*,
- $VFA(t)$: effluent VFA concentration at a given time *t*,
- VFA_{MAX}: maximum effluent VFA concentration.

Δeq_{CH_4} is negative or positive depending on whether $eq_{CH4}(t)$ tends to zero or not, respectively. Moreover, this equation features a modifying algebraic factor (δ) that is defined by Equation 7 to account for opposite signs between $\left|eq_{CH_4}(t)\right|$ and $\left|eq_{CH_4}(t-1)\right|$.

$$\delta = \frac{eq_{CH_4}(t) \cdot eq_{CH_4}(t-1)}{\left|eq_{CH_4}(t) \cdot eq_{CH_4}(t-1)\right|} \quad \text{(Eq. 7)}$$

For the fuzzification stage, three Gaussian membership functions, represented by Equation 8, were considered for eq_{CH_4} and Δeq_{CH_4}: Negative (*N*), Zero (*Z*) and Positive (*P*); and one Gaussian membership function was defined for *dVFA*: Zero (*Z*). As each Gaussian membership function is defined by two parameters (centre *c* and amplitude *a*), the control system had a total of 14 parameters as regards to the fuzzification stage. Concerning the defuzzification stage, four singleton membership functions were defined for Δq_{IN_SP}: High Negative (*HN*), Low Negative (*LN*), Low Positive (*LP*) and High Positive (*HP*). Therefore, the control system had a total of 4 parameters regarding the defuzzification stage.

$$\mu(p) = \exp\left(-\frac{(p-c)^2}{2\cdot\sigma^2}\right) \quad \text{(Eq. 8)}$$

where:

- $\mu(p)$: degree of membership of the input variable *p*,
- *p:* numerical value of the variable,
- *c:* centre of the Gaussian-type membership function,
- σ: amplitude of the Gaussian-type membership function.

Table 2 shows the resulting grade of membership to the different output linguistic labels that define the output fuzzy set. As this table shows, the effect of the input variable *dVFA* (represented by the third right-side term of rules #1, #2, #5a and #6b, *i.e.* 1 - μ $(dVFA)_Z$) on the output linguistic variable decreases as the effluent VFA concentration decreases (*i.e. if* μ $(dVFA)_Z = 0$ *then* 1 - μ $(dVFA)_Z = 1$). On the contrary, the effect of *dVFA* cancels the corresponding control action when the effluent VFA concentration is close to VFA_{MAX} (*i.e. if* μ $(dVFA)_Z) = 1$ *then* 1 - μ $(dVFA)_Z = 0$). Hence, the increase in the influent flow rate

controlled by the inference rules #1, #2, #5a and #6b is cancelled when the system is working at maximum VFA capacity.

The output linguistic variable (Δq_{IN_SP}) was obtained by applying Larsen's fuzzy inference method [32]. In the defuzzification stage, the Height Defuzzifier method was employed [33] to obtain a single output value from the output fuzzy set.

Finally, the control action of the upper-layer controller was calculated as expressed by Equation 9.

$$q_{IN\_SP}(t) = q_{IN\_SP}(t-1) + \Delta q_{IN\_SP}(t) \quad \text{(Eq. 9)}$$

2.4.2. Supervisory controller description

The supervisory controller determined the variation in the set-point of the methane flow rate (Δq_{CH4_SP}) on the basis of two inputs: the error in the methane flow rate (Equation 4) and the accumulated error in the methane flow rate (Equation 10).

$$\Sigma eq_{CH_4}(t) = \Sigma eq_{CH_4}(t-1) + ST \cdot eq_{CH_4}(t) \quad \text{(Eq. 10)}$$

where:

- $\Sigma eq_{CH4}(t)$: accumulated error in the methane flow rate at a given time,
- $\Sigma eq_{CH4}(t-1)$: accumulated error in the methane flow rate at the previous sampling time (*ST*),

Regarding the fuzzification stage, three additional Gaussian membership functions were considered for Σeq_{CH_4} : Negative (*N*), Zero (*Z*) and Positive (*P*). Concerning the defuzzification stage, three singleton membership functions were defined for $\Delta q_{CH_4\_SP}$: Low Negative (*LN*), Low Positive (*LP*) and High Positive (*HP*). Thus, the supervisory controller added to the proposed fuzzy-logic controller a total of 6 and 3 parameters regarding fuzzification and defuzzification, respectively. Table 2 shows the resulting grade of membership to the different output linguistic labels that defined the output fuzzy set of the supervisory controller.

The output linguistic variable ($\Delta q_{CH_4\_SP}$) was determined following the method described in section 2.4.2. Finally, the control action of the supervisory controller was calculated as expressed by Equation 11.

$$q_{CH_4\_SP}(t) = q_{CH_4\_SP}(t-1) + \Delta q_{CH_4\_SP}(t) \quad \text{(Eq. 11)}$$

2.4.3. Simulation-based design and validation

The controller was firstly designed and tuned by simulation in Matlab® Simulink® using the Fuzzy Logic Toolbox™. To this aim, a simplified version of the model BNRM2 [34] was used. This model considers the main physicochemical and biological processes taking place during AD, including gas-liquid transfer (nitrogen, ammonia, oxygen, hydrogen, methane and carbon dioxide), a chemical model for pH calculation and biological steps such as acidogenesis, acetogenesis and acetoclastic and hydrogenotrophic methanogenesis. Therefore, this model allowed the simulation of the methane production rates and the concentrations of

VFAs in the effluent.

The control tuning was performed by a trial-error approach until obtaining an adequate response (*i.e.* a deviation of less than 5 % between the response and the set-point given by the supervisory controller).

3. Results and discussion

3.1. Simulation-based validation of the control system

Figure 3 shows the performance of the advanced controller obtained by simulation after control tuning. Figure 3a presents the evolution of the resulting methane flow rate and the corresponding set-point commanded by the supervisory controller, and the influent flow rate commanded by the upper-layer controller. Figure 3b shows the effluent VFA concentration and the VFA_{MAX} considered. VFA_{MAX} was set to 750 mg COD L^{-1} (value fixed from knowledge obtained from previous experiments). This maximum VFA concentration resulted in a minimum COD removal efficiency of 80%.

It must be mentioned that the value of VFA_{MAX} has to be carefully selected according to the control objectives (*i.e.* enhance AD performance and stability, minimize VFA contents in the effluent, meet COD discharge limits, achieve VFA requirements in downstream processes…) and process specificities. For instance, higher VFA_{MAX} values can be potentially applied without risk of reactor acidification if the controller performs in a high-alkalinity system. On the other hand, lower VFA_{MAX} values should be applied when the alkalinity of the system is low or when no pH control is possible, thus reducing the propensity of possible

acidification problems.

As Figure 3a shows, the controller was able to maintain the simulated methane flow at values close to the controlled set-point until reaching the constraint of the maximum VFA concentration (750 mg COD L^{-1}). This maximum VFA concentration was approached from days 5 to 6, thus the increase in the influent flow rate was almost null. Only when the VFA concentration was below its maximum threshold value it was possible to increase slightly the influent flow rate to compensate the negative error in the methane flow (see period from day 6 to end). As the differences between the measured and the desired values were getting smaller, also did the changes in q_{CH4_SP} and q_{IN_SP}. Within an infinite time and no external disturbances, the concentrations of VFA would eventually reach VFA_{MAX}, showing an optimal performance according to the desired VFA content in the effluent.

It is important to notice that during the first period of simulation, when the VFA concentrations were low, the methane production was higher than the one commanded by the supervisory controller (*e.g.* 2nd day). However, the supervisory controller did not increase more the set-point in order to avoid overloading the reactor.

3.2. Experimental validation of the control system

Figure 4 presents the evolution of the OLR and HRT throughout the experimental period. As it can be observed, the operational period is divided in 3 different sections: (I) reactor start-up; (II) transitory period including a pH-shock due to failure of the pH sensor; and (III) controlled process. As Figure 4 shows, the OLR was manually increased from 0 to 4 g COD L^{-1} d^{-1} from day 0 to around 40, whilst maintaining the HRT around 3 d. This progressive

increase in the OLR was carried out to minimize possible disturbances during the biofilm formation at the start-up process. During this period, the concentration of VFAs in the effluent was used as state indicator of the process performance. This allowed avoiding the inhibition of the newly-grown biomass due to overloading of the reactor. This is the reason for the decrease in the OLR from days 20 to 30. During this period, high VFA concentrations were observed in the effluent (up to 1500 mg COD L^{-1}) and, as the acetate inhibition coefficient of propionic-oxidizing bacteria is around 2500 mg COD L^{-1} (see, for instance, Siegert and Banks [35]), the OLR was reduced to avoid inhibition of these microorganisms. Around day 50 (period II in Figure 4), a significant increase of the pH in the reactor (up to around 9) occurred due to a failure in the lower-layer pH controller (data not shown). This resulted in a considerable decay of the anaerobic biomass. Therefore, the OLR and HRT were set to 1.4 g COD L^{-1} d^{-1} and 9 d, respectively, in order to recover the system to appropriate operating conditions. From day 61 on (period III in Figure 4), the advanced controller was turned on for optimizing the process performance. Figure 4 shows that the controller increased progressively the inflow to the reactor until reaching the maximum treatment capacity of the system, which was limited by VFA_{MAX} (set at 750 mg COD L^{-1}).

Figure 5 shows the evolution throughout the operational period of: the methane yield (Figure 5a); and the total COD removed in the system and the COD fraction removed for methane production (Figure 5b). As Figure 5 shows, no methane production was observed until day 20. This suggests that the removal of COD from days 10 to 20 was mainly related to the anabolism of the anaerobic biomass (*i.e.* initial growth, fixation, and acclimation of the biomass [7]) and to the production of the gas required for filling the headspace volume of the reactor and to achieve conditions of gas-liquid equilibrium within the system. Therefore, 20 days was identified in this study as the minimum time for obtaining a functional anaerobic

biomass consortium under conditions of equilibrium. The decrease in the COD removal observed from days 25 to 30 was related to the aforementioned accumulation of VFAs. After decreasing the OLR, the COD removal efficiency was restored. From day 30 to around 50, a quite stable COD removal efficiency (up to 85 %) was achieved. Concerning to the methane yields after day 20, this value increased greatly (reaching values up to 0.34 L_{CH4} per gram of COD removed) due to catabolism of methanogenic archaea. However, this value decreased from 0.32 to 0.10 L_{CH4} g^{-1} COD_{REM} from day 25 to day 30. According to Michaud et al. [7], this may have been caused by disturbances occurring during the initial contact of the microorganisms and the fixed support media. Therefore, even it after 20 days a functional anaerobic biomass existed, a minimum time of 35 days was needed to obtain a functional anaerobic biofilm. This value is in agreement with previous results reported in the literature (see, for instance, Michaud et al. [7]). Afterwards, the methane yield increased continuously throughout this operational period (except for period II), reaching again values up to 0.34 L_{CH4} g^{-1} COD_{REM}. This behaviour suggested the development and maturing of a stable biofilm.

As mentioned before, during period II a system failure occurred due to a pH-shock. As Figure 5 illustrates, both COD removal for methane production and methane yield presented a sharp decrease. Nevertheless, when the control system was turned back on (period III), it was possible to quickly recover the system to the previous state, achieving values of methane yields and COD removals for methane production of around 0.34 L_{CH4} g^{-1} COD_{REM} and 85 %, respectively.

The fixed-bed anaerobic reactor achieved an efficient and stable performance when running the proposed advanced controller (period III). As Figure 5b shows, COD removal

efficiencies above 80 % were achieved during this period. In addition, a high stable methane yield of around 0.34 L_{CH4} g^{-1} COD_{REM} was reached (see Figure 5a). These results highlighted the suitable performance of the process under controlled conditions. Indeed, comparing the results from periods I and III, it can be stated that enhanced process performances were achieved in terms of COD removal, methane production and treatment capacity.

Figure 6 shows the performance of the advanced controller during the operational period III. As it can be observed, the supervisory controller increased continuously the set-point for the methane flow rate (see Figure 6a) until reaching the maximum effluent VFA concentration (see Figure 6b). Therefore, the upper-layer controller continuously increased the influent flow to reach the corresponding methane flow rate set-point. As a result, a maximum methane production of around 17 L h^{-1} was reached when operating with a VFA_{MAX} of 750 mg COD L^{-1}. A deviation of the methane flow rate from the established set-point lower than 10 % was achieved, whilst the methane yield was maintained around 0.35 L_{CH4} g^{-1} COD during the pseudo-stationary operational period (see days 65 to the end in Figure 5a). Throughout this period, a methane-rich biogas was also produced (with methane contents in the biogas around 85 ± 2 %).

As designed, the controller increased q_{CH4_SP} only if the concentration of VFA in the effluent was below VFA_{MAX}. The results from days 70 to 73 show that, even if eq_{CH4} was negative (*i.e.* q_{IN_SP} could be higher), the supervisory controller did not allow increasing the influent flow rate because the concentration of VFA was over VFA_{MAX}. The same occurred the days 77-78, verifying the correct performance of the controller.

As explained in section 3.1., without disturbances the process would reach eventually VFA_{MAX}, never overpassing it. However, this value was reached and overpassed in different occasions, suggesting that, as in any real process, disturbances affected the system. As the temperature and the pH were controlled and kept at barely constant values, the most likely sources of disturbances were the feed itself and the recirculation flow. Heterogeneity of the substrate may have caused small differences in the COD entering the reactor. In addition, as the substrate was kept into a feeding tank before entering the reactor, some extent of degradation had already occurred during the storage period, modifying the input concentration of VFA. Moreover, as the recirculation flow was manually controlled, there were sudden drops in the recycling flow rate due to partial clogging of the tubing. This caused significant variations of this parameter throughout the operational period (varying from 100 to 700 L h^{-1}). This may have affected the methane production, mainly by modifying the stripping rate of the produced gases from the liquid phase. Lower recycling flow rates might cause lower methane stripping rates from the liquid phase, leading to lower gaseous outflow rates of methane. However, the control action was able to compensate the disturbances in the methane production, achieving anyway the desired set-point. Therefore, the fuzzy-logic control action resulted in a suitable performance under disturbances which are likely to be similar to those expected in full-scale plants (*e.g.* variations in the recycling flow rate).

It can be concluded that, after a relatively simple calibration, the proposed fuzzy-logic controller was able to successfully optimize the process performance, maximizing the methane production and the VFA content in the effluent up to the chosen fixed values, whilst resulting in adequate COD removal efficiencies and methane yields. At this point, it is important to mention that, as the organic matter within the winery wastewater used as substrate is mainly composed of soluble COD, the AD kinetics were not limited by the

hydrolysis step. This allowed the application of a short period for the evaluation of the control strategy.

Finally, when considering the application of this fuzzy-logic control system at full-scale for control and optimization, different modifications might be considered to further improve the performance, such as optimization of the control dynamics for both controllers, fine adjustment of the knowledge-based fixed values (*i.e.*, VFA_{MAX}) and optimization of the tuning parameters (*i.e.* centre, amplitude and singleton values for the fuzzification and defuzzification stages), among others.

4. Conclusions

A fuzzy-logic based controller for optimizing the process performance of a 350 L fixed-bed anaerobic reactor treating winery wastewater was developed by simulation and validated under specific conditions that were similar to the ones expected at full-scale plants. The controller aimed at maximizing the methane productivity whilst controlling the VFA content in the effluent. By application of the fuzzy-logic control system, a deviation of the methane flow from the established set-point lower than 10 % was achieved. The methane yield resulted in values around 0.29 L_{CH4} g^{-1} COD, with COD removal efficiencies of up to 85 % obtained throughout the whole experimental period. On the other hand, the controller allowed an adequate control of the VFA content in the effluent, with values close to the established set-point (750 mg COD L^{-1}). Hence, the proposed fuzzy-logic controller was able to successfully control the system performance close to optimal conditions, maximizing the methane productivity and the VFA concentration, whilst resulting in adequate COD removal efficiencies and methane yields.

Acknowledgements

This research work has been supported by the Spanish Research Foundation (MICINN FPI grant BES-2009-023712), which is gratefully acknowledged.

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607

Figure and table captions

Figure 1. Flow diagram of the plant, including instrumentation. (Nomenclature: **FIT**: Flow-Indicator-Transmitter; **PIT**: Pressure-Indicator-Transmitter; **pH**: pH-Transmitter; **CT**: Conductivity-Transmitter; **T**: Temperature sensor; **PLC**: Programmable Logic Controller).

Figure 2. Flow diagram of the advanced fuzzy-logic controller

Figure 3. Simulation of the control system performance. Evolution of: **(a)** methane flow and influent flow; and **(b)** VFA content in the effluent

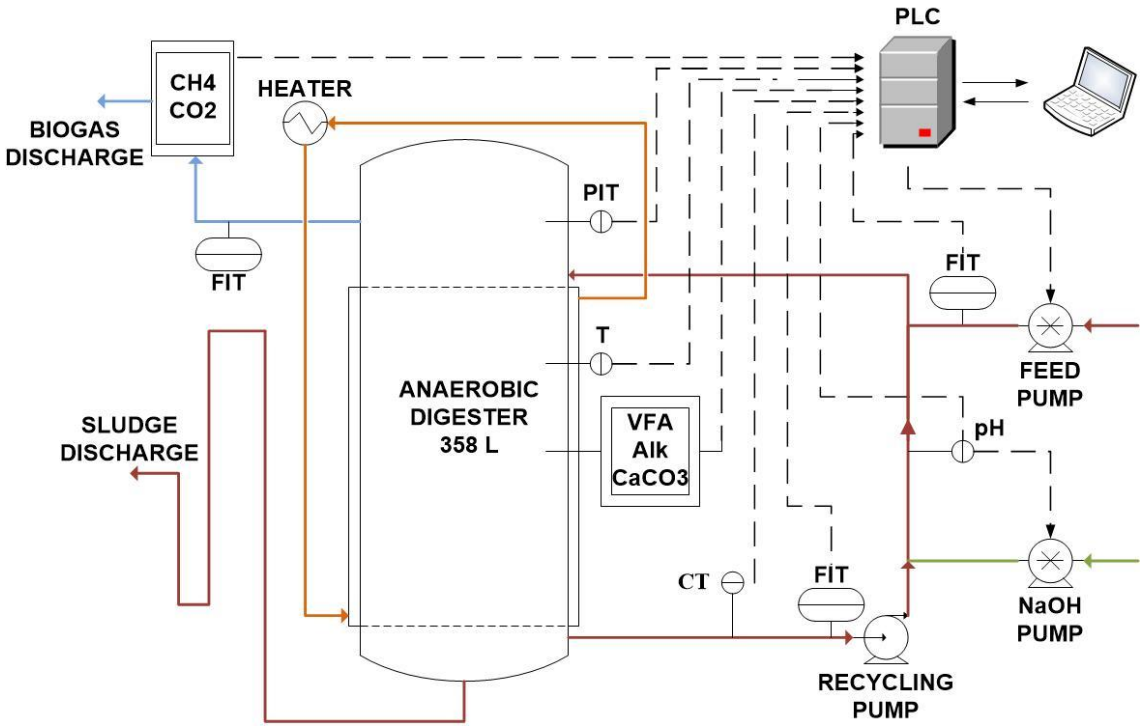
Figure 4. Evolution during the operational period of OLR and HRT. (I), (II) and (III) stand for the different sections of the operational period

Figure 5. Evolution during the operational period of: (a) methane yield; and (b) fraction of total COD removed and fraction of COD removed for methane production. (I), (II) and (III) stand for the different sections of the operational period

Figure 6. Control system performance. Evolution of: (a) methane flow and influent flow; and (b) VFA content in the effluent. SP stands for Set-points. The values marked with * represent the 2h-moving averages of the measured values (every 60 min)

Table 1. Average raw wastewater characteristics

Table 2. Advanced fuzzy-logic controller action: grade of membership to the output linguistic labels



627

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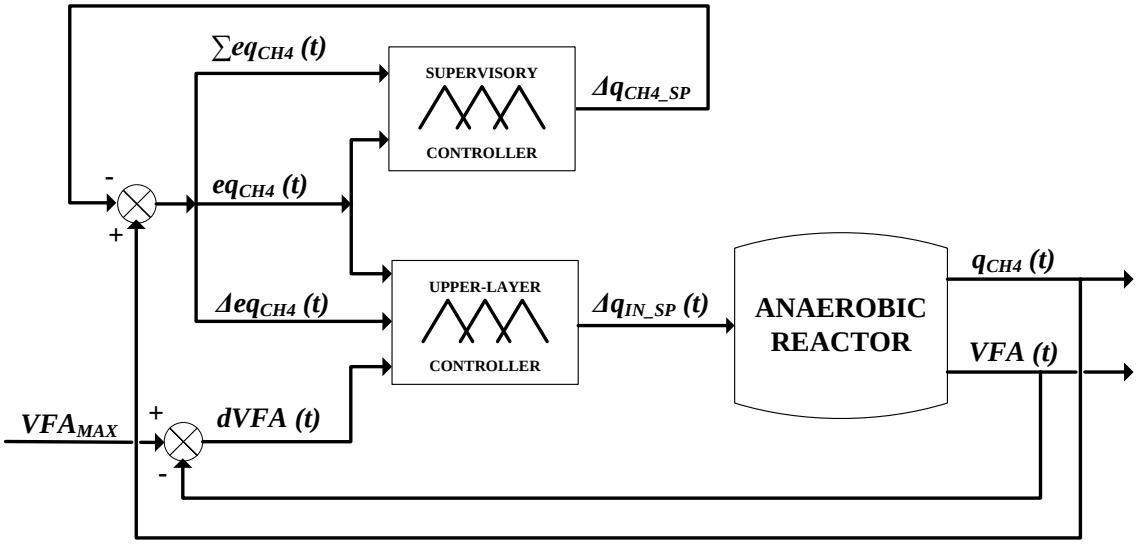

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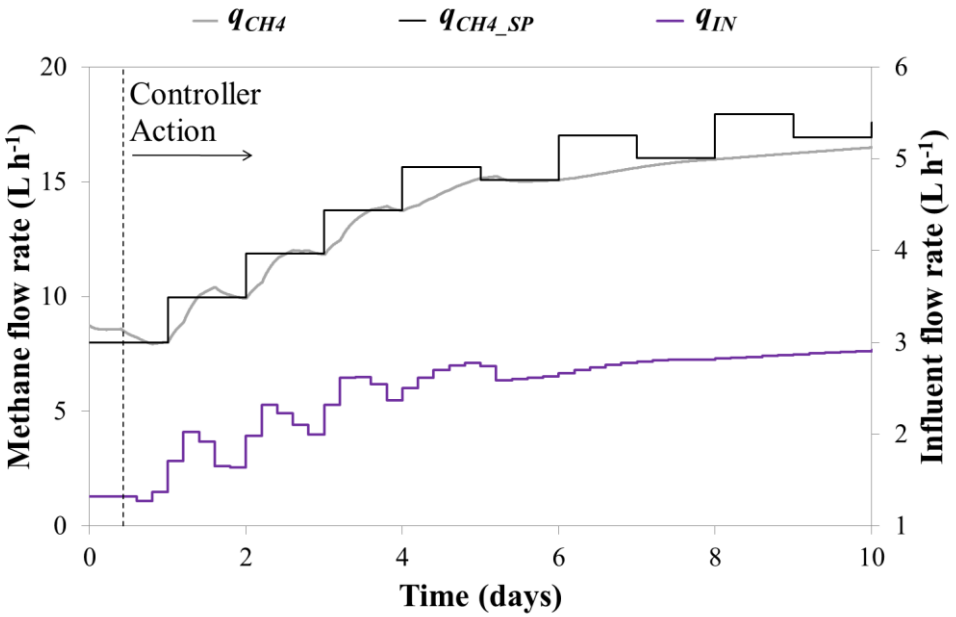
632 **Figure 2.** Flow diagram of the advanced fuzzy-logic controller



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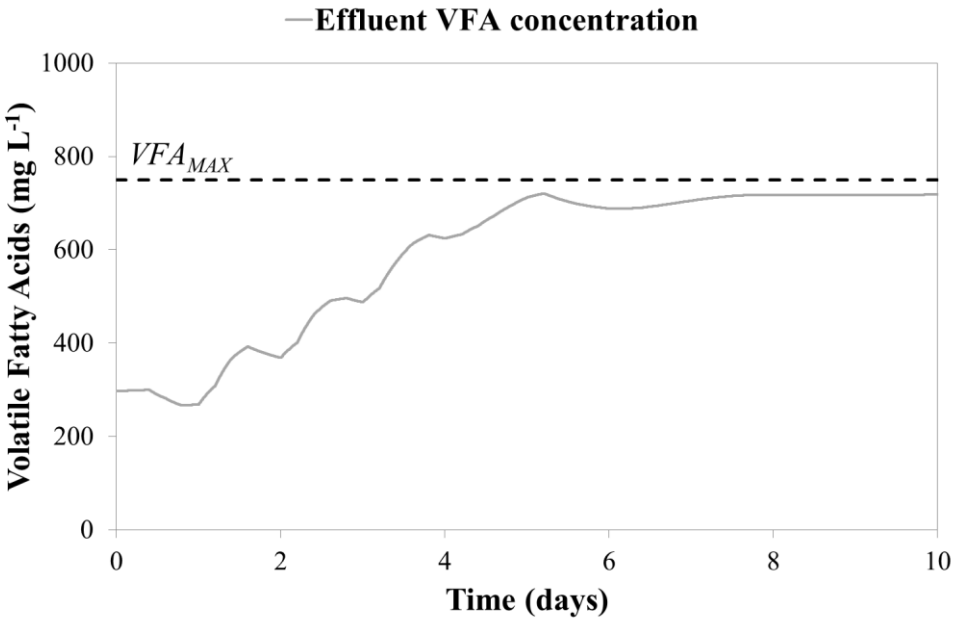
634 **(a)**



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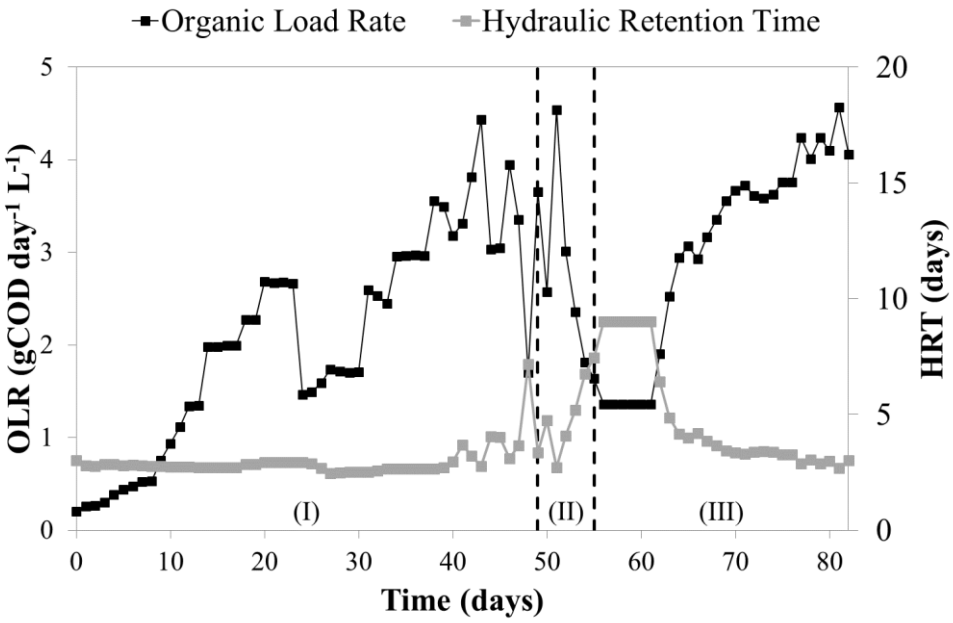
636 **(b)**

637 **Figure 3.** Simulation of the control system performance. Evolution of: (a) methane flow and

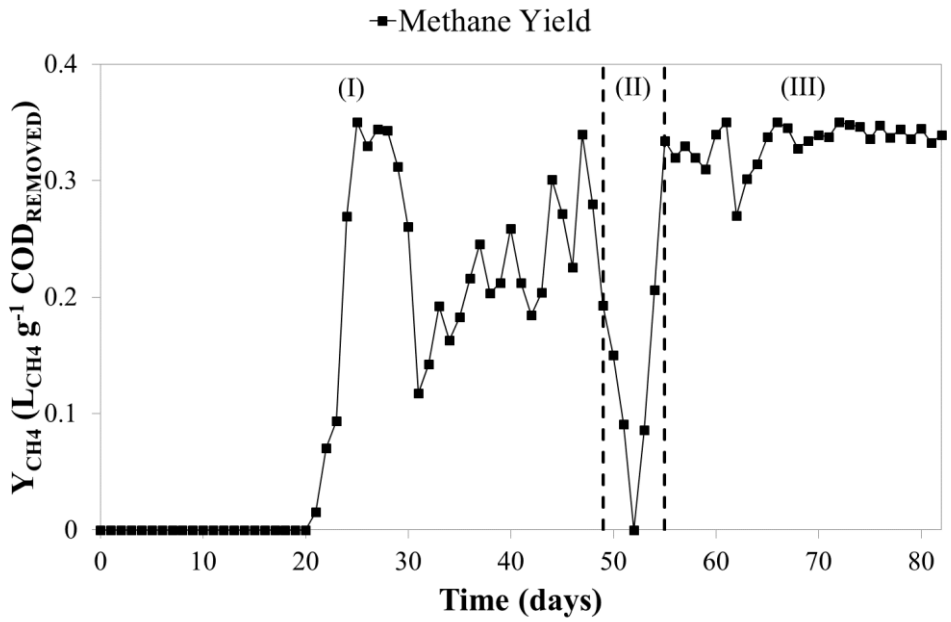
638 influent flow; and (b) VFA content in the effluent



639

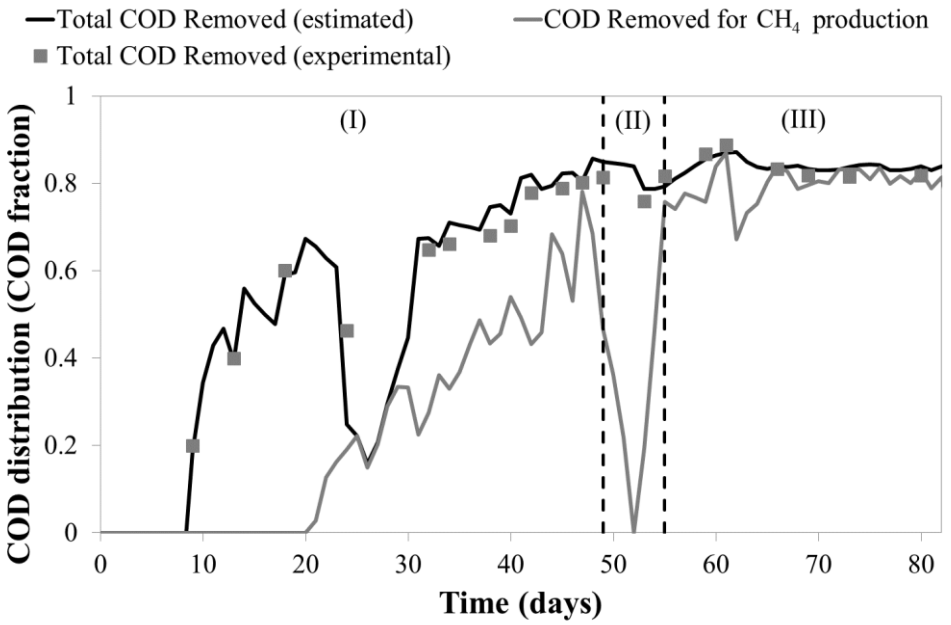
640 **Figure 4.** Evolution during the operational period of OLR and HRT. (I), (II) and (III) stand

641 for the different sections of the operational period



642

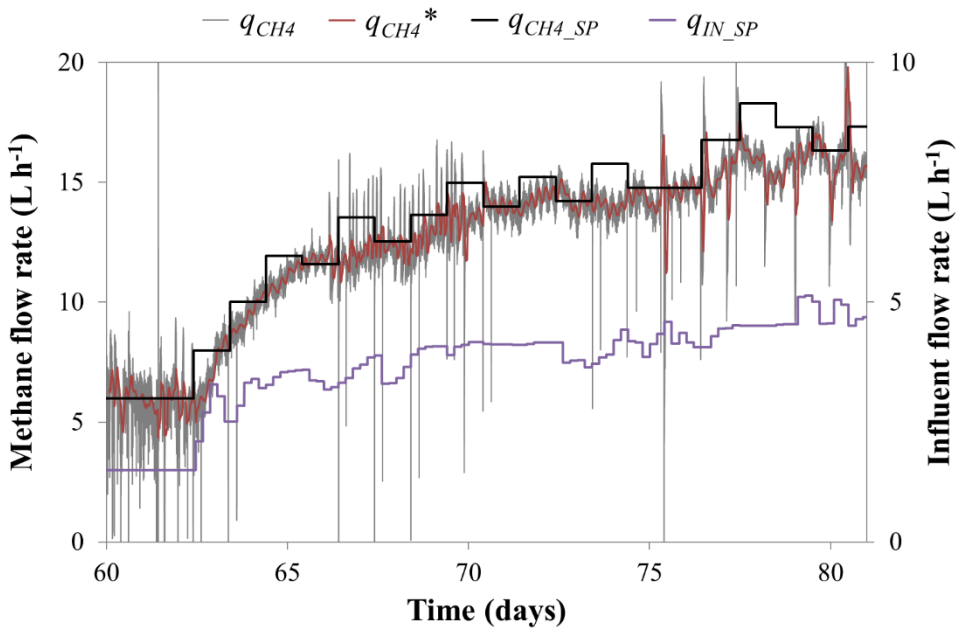
643 **(a)**



644

645 **(b)**

646 **Figure 5.** Evolution during the operational period of: (a) methane yield; and (b) fraction of
647 total COD removed and fraction of COD removed for methane production. (I), (II) and (III)
648 stand for the different sections of the operational period



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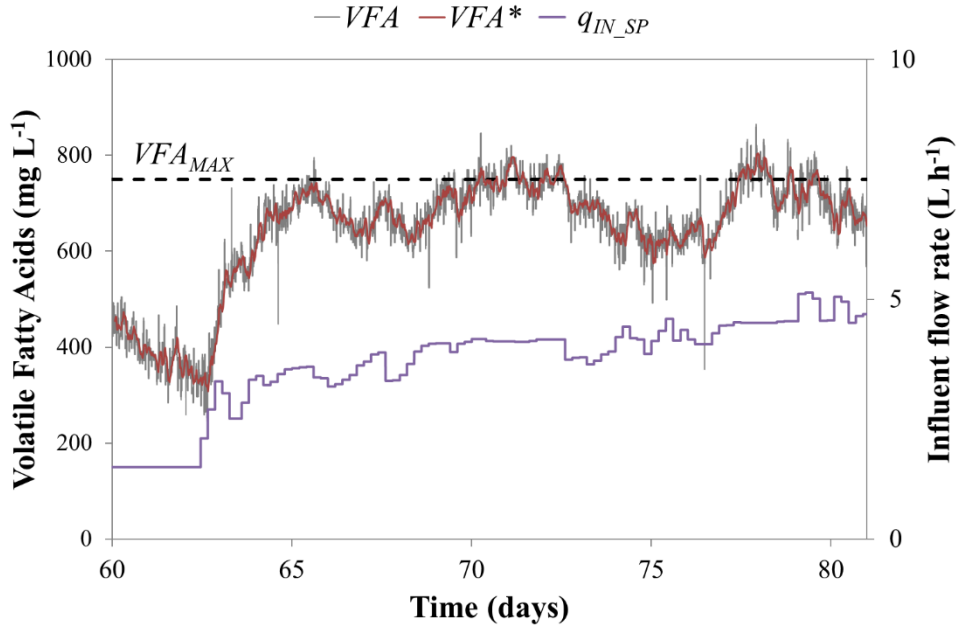
650 **(a)**



651

652 **(b)**

653 **Figure 6.** Control system performance. Evolution of: (a) methane flow and influent flow; and

654 (b) VFA content in the effluent. SP stands for Set-points. The values marked with * represent

655 the 2h-moving averages of the measured values (every 60 min)

656 **Table 1.** Average raw wastewater characteristics

Parameter	Unit	Mean ± SD
COD	g COD L^{-1}	21.6 ± 0.8
Acetate	g COD L^{-1}	3.7 ± 0.4
Propionate	g COD L^{-1}	4.6 ± 0.8
Butyrate	g COD L^{-1}	2.8 ± 0.3
Valerate	g COD L^{-1}	1.5 ± 0.7

657

658 **Table 2.** Advanced fuzzy-logic controller action: grade of membership to the output linguistic

659 labels

Inference Rule	**Grade of membership to the output linguistic variables**						
Supervisory controller							
A	$\mu(\Delta q_{CH4\_SP})_{HP}$	=	$\mu(eq_{CH4})_{Z}$	·	$\mu(\Sigma eq_{CH4})_{Z}$		
B	$\mu(\Delta q_{CH4\_SP})_{LN}$	=	$\mu(eq_{CH4})_{N}$	·	$\mu(\Sigma eq_{CH4})_{N}$		
C	$\mu(\Delta q_{CH4\_SP})_{LP}$	=	$\mu(eq_{CH4})_{P}$	·	$\mu(\Sigma eq_{CH4})_{P}$		
Upper-layer controller					$^{*}\ eq_{CH_4} < 0;\ ^{**}\ eq_{CH_4} > 0$		
1	$\mu(\Delta q_{IN})_{HP}$	=	$\mu(eq_{CH4})_{N}$	·	$\mu(\Delta eq_{CH4})_{Z}$	·	$(1 - \mu(dVFA)_{Z})$
2	$\mu(\Delta q_{IN})_{HP}$	=	$\mu(eq_{CH4})_{N}$	·	$\mu(\Delta eq_{CH4})_{P}$	·	$(1 - \mu(dVFA)_{Z})$
3	$\mu(\Delta q_{IN})_{HN}$	=	$\mu(eq_{CH4})_{P}$	·	$\mu(\Delta eq_{CH4})_{Z}$		
4	$\mu(\Delta q_{IN})_{HN}$	=	$\mu(eq_{CH4})_{P}$	·	$\mu(\Delta eq_{CH4})_{P}$		
5^{*}	$\mu(\Delta q_{IN})_{LP}$	=	$\mu(eq_{CH4})_{Z}$	·	$\mu(\Delta eq_{CH4})_{N}$	·	$(1 - \mu(dVFA)_{Z})$
5^{**}	$\mu(\Delta q_{IN})_{LN}$	=	$\mu(eq_{CH4})_{Z}$	·	$\mu(\Delta eq_{CH4})_{N}$		
6^{*}	$\mu(\Delta q_{IN})_{LP}$	=	$\mu(eq_{CH4})_{Z}$	·	$\mu(\Delta eq_{CH4})_{P}$	·	$(1 - \mu(dVFA)_{Z})$
6^{**}	$\mu(\Delta q_{IN})_{LN}$	=	$\mu(eq_{CH4})_{Z}$	·	$\mu(\Delta eq_{CH4})_{P}$		

660

List of Abbreviations

AD – Anaerobic Digestion
AnMBR – Anaerobic Membrane Bioreactor
CT – Conductivity-Transmitter
EGSB – Expanded Granular Sludge Blanket
FIT – Flow-Indicator-Transmitter
GC – Gas Chromatograph
HN – High Negative
HP – High Positive
HRT – Hydraulic Retention Time
LN – Low Negative
LP – Low Positive
N – Negative
OLR – Organic Loading Rate
P – Positive
PID – Proportional-Integral-Derivative
PIT – Pressure-Indicator-Transmitter
PLC – Programmable Logic Controller
SRT – Solid Retention Time
SR – Sampling Time
T – Temperature Sensor
UASB – Up-flow Anaerobic Sludge Blanket
VFA – Volatile Fatty Acid
Z – Zero

List of Symbols

q_{CH4} – Methane flow rate
$G_{CORRECTED}$ – Corrected biogas flow
$G_{MEASURED}$ – Measured biogas flow
frho – Volumetric correction factor
rho_{AIR} – Volumetric weight of air
rho_{CH4} – Volumetric weight of CH_4
rho_{CO2} – Volumetric weight of CO_2
rho_{N2} – Volumetric weight of N_2
eq_{CH4} (t) – Error in methane flow rate at a given time
q_{CH4} (t) – Methane flow rate at a given time
q_{CH4} * – 2-h moving average of **q_{CH4} (t)**
q_{CH4_SP} – Set-point of methane flow rate
Δeq_{CH4} (t) – Variation in the error of the methane flow rate at a given time
eq_{CH4} (t-1) – Error in methane flow rate at the previous control action
δ – Modifying algebraic factor
dVFA (t) – difference between VFA_{MAX} and the VFA content in the effluent at control time
VFA (t) – Effluent VFA concentration at a given time
VFA * – 2-h moving average of **VFA (t)**
VFA_{MAX} – Maximum effluent VFA concentration
Σeq_{CH4} (t-1) – Accumulated error in methane flow rate at the previous control action
Σeq_{CH4} – Accumulated error in methane flow rate at a given time
ST – Sampling time
p – Numerical value of a variable
c – Center of the Gaussian-type membership function
μ (p) – Degree of membership of the input variable p
σ – Amplitude of the Gaussian-type membership function
Δq_{CH4_SP} – Modification in methane flow rate set-point
q_{IN_SP} – Set-point of influent flow rate
Δq_{IN} – Modification of the influent flow rate
Y_{CH4} – Methane yield