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Fractal Analysis of pH Time series for Monitoring of an Anaerobic CSTR type Digester

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***Abstract*—Fluctuations of pH measurements from anaerobic digesters show an apparent random behavior. The monitoring of pH complex fluctuations can provide information about the operational conditions of anaerobic digesters. Then a suitable analysis can provide important insights on the dynamical behavior and operational stability of these processes. In this work, fractal scaling properties of pH signals from anaerobic digesters are explored by using fluctuation analysis. Results indicated that fractal dimension is directly related to key physical variables and can be used as indexes for the monitoring of anaerobic digesters.**

Keywords: Monitoring, Time series analysis, Wastewater treatment

I. INTRODUCTION

Biological wastewater treatments are used to decompose the organic compounds contained into domestic and industrial wastewaters in order to achieve the reduction of the pollutant concentration in the outlet stream below a specified value (Olson et al., 2005). Biological treatment can be classified in three basic categories: aerobic, anaerobic and anoxic. Particularly, anaerobic digestion (AD) refers to the natural biological degradation that happen when bacteria breaks down organic matter in the absence of oxygen through the cell metabolism. Therefore, additionally to the reduction of the pollutant concentrations, a high valued byproduct called biogas is produced which can be used as an energy source (Mata et al., 2000). Based on the operation mode, AD can be performed as batch or continuous process. In the former case, both substrate and the microbial consortium are added at once to a controlled and sealed environment where the biochemical reactions are allowed to run their course during a finite time. The anaerobic sequencing batch reactor (AnSBR) is the most commonly used batch AD bioreactor configuration. In regard to the continuous operation mode, organic matter and end products are continuously added and removed respectively, resulting in continuous pollutant degradation and biogas production. Examples of this operation mode of AD include continuous stirred-tank reactors (CSTR), fixed bed reactors (FBR), upflow anaerobic sludge blankets (UASB), expanded granular sludge beds reactors (EGSB) (Hwu et al., 1998; Agdag et al., 2005; Mendez-Acosta et al., 2011), among others. The continuous configuration is widely used in industrial wastewater treatment units. The selection and design of AD bioreactor systems depend on several factors such as organic loads, degradation kinetics, and environmental conditions. On the other hand, operating a wastewater treatment bioreactor is not a simple task since the quantity and composition of wastewater varies continuously. The microbial consortium also changes under the influence of internal and external factors, and is subject to complex and synergistic interactions between several functional groups of microorganisms. Moreover, the lack of on-line sensors that allow the real-time monitoring of the process state has been an obstacle to improve the understanding of the bioreactor performance. Therefore, nowadays one of the main objectives of the technical and scientific communities is related to the development of efficient devices that allow the real-time monitoring of key variables such as chemical oxygen demand (COD), volatile fatty acids (VFA) and biogas composition (Kleybocker et al., 2012).

Although considerable effort has been devoted to the development of sensors that allow the on-line monitoring of AD processes, nowadays its operation is usually restricted to the monitoring of secondary variables such as pressure, liquid and gas flow rates, temperature and pH. Several authors have considered that pH measurement can be used as an index of the AD performance given its direct relation with the microbial activity. In addition, equipment for pH measurements is inexpensive and easy to implement (Anderson and Yang, 1992). However, a drawback is that detailed information about efficiency and correct operation of the process can be hardly detected by using direct pH measurements (Kleybocker et al., 2012). Typically, on-line measurements exhibit complex fluctuations around time-varying trends, which are normally considered as uncorrelated noise. Consequently, such fluctuations are usually eliminated for the analysis of interest variables. However, the complex fluctuations can be an indicative of the complexity of physical phenomena involve in the AD process. Particularly, the pH signals taken from AD processes may reflect the complexity of the biological reactions as well as to provide relevant information of the digester performance (Olson et al., 2005).

Several studies have demonstrated that the fractal analysis of time series recovered from physical processes allow the identification of serial auto-correlations between fractal and physical parameters. In turn, an index of such auto-correlations can be used for characterization, evaluation or diagnosis tasks (Zunino et al., 2009). Recent studies have been shown that fractal analysis of pH fluctuations in bioreactors is a useful tool for the identification of correlations between fractal parameters and key physical parameters (i.e., Chemical Oxygen Demand (COD), Volatile

Fatty Acids (VFA) and biogas production.) in anaerobic digesters for the treatment of tequila vinasses (Mendez-Acosta et al., 2013). In this study, detrended fluctuation analysis (DFA) is proposed for exploring the presence of long-range correlations in pH time series obtained from an anaerobic CSTR type digester used in the tequila vinasses treatment. Our results indicate that the fractal analysis of pH time series from CSTR digester allows the identification of four characteristic regions, indicating that in pH signal can be identified four relevant physical phenomena. Moreover, by estimating the fractal dimension for different process stages, it is possible to identify direct correlations between the fractal parameters and three key variables in the AD process.

This work is organized as follows. Section 2 the experimental setup is presented. Section 3 describes the computations involved in the DFA. Section 4 the main results are discussed in terms of the physics of the anaerobic digestion process. Finally, Section 5 presents some concluding remarks.

II. Materials and Methods

In this section, a brief introduction of the AD treatment of tequila vinasses and the description of CSTR bioreactors treating tequila vinasses are presented.

Anaerobic digestion and tequila vinasses treatment

Tequila liquor is produced from the distillation of fermented juice of an agave plant in four steps: cooking to hydrolyze inulin into fructose, milling to extract the sugars, fermentation with natural or commercial strains to convert the sugars into ethanol and organoleptic compounds, and, finally, distillation via a two-step process. Tequila industry produce high volumes of effluents with high pollutant loads called vinasses, which are discharged (untreated or partially treated) into natural receivers, thus causing severe environmental problems. Several technologies are applied for tequila vinasses treatment at full scale (Lopez et al., 2010). In particular, AD has shown its suitability for the treatment of tequila vinasses due to its high organic removal rates, low energy-input requirements, energy recovery from the production of methane, and low sludge production.

In AD processes organic contaminant compounds (substrates) are broken down into smaller molecules by chemicals and microorganisms, producing a mixture of methane and carbon dioxide, reducing the chemical oxygen demand of the influent (Mata et al., 2000). AD can be separated in four fundamental steps: hydrolysis, acidogenesis, acetogenesis, and methanogenesis. A variety of microorganisms coexist in an anaerobic bioreactor, and their appropriate interaction is necessary for the effective conversion of organic compounds to methane (McCarty, 1964). In order to provide high-rate oxidation of organic pollutants, microorganisms must be provided with an environment that allows them to thrive (Kim et al., 2006). Temperature, pH, dissolved oxygen and other factors affect the natural selection, survival and growth of microorganisms and their rate of biochemical oxidation. In AD, it is crucial to measure the pH throughout the entire process to favor the activity of methanogens. The microorganisms that are responsible for the creation of methane have an optimal pH between 6.5 to 8.

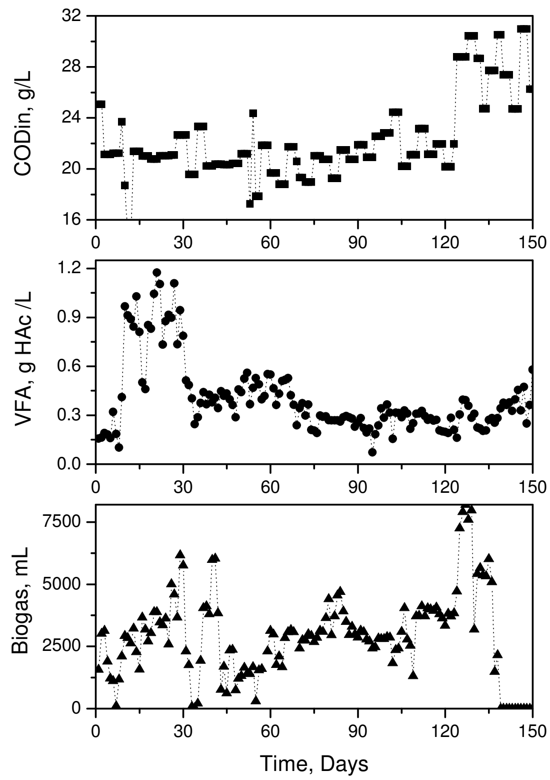

Figure 1. Off-line measurements collected from the anaerobic CSTR: a) COD inlet concentration, b) VFA concentration and (c) Biogas production.

Anaerobic CSTR type digester

In Mendez-Acosta et al., (2010), a lab-scale CSTR-type digester for the treatment of Tequila vinasses was used. It was shown that 90-95% COD removal and the production of biogas rich in methane (>70%) can be achieved with this bioreactor configuration. The digester unit has an effective volume of 5 L which was made of polyvinyl chloride (PVC). The feeding was kept in a 6 L dilution tank, where known volumes of tap water and vinasses were mixed in different proportions allowing the manipulation of the influent COD concentration. The feeding was pumped from the bottom of the digester using a peristaltic pump. The vinasses pH in the dilution tank was regulated around 6.5-7.0 by adding a NaOH solution through an off-on control scheme. Fresh substrate is mixed with the recycled liquid just before entering to the reactor. The digester pH was regulated around 7.4 by adding a NaOH solution through an off-on control scheme. The digester temperature was regulated around 35°C by using an immersion circulator and water as heat transfer liquid which was conducted through the reactor jacket. A Thermo Scientific Orion 8256BN pH electrode was used to measure the digester pH whose values were recorded every second, time enough to detect small changes on this variable. Other on-line measured variables were temperature, pressure and the biogas flow rate. A National Instruments cRIO9004 device equipped with analogical and digital cards was used in the acquisition, treatment and storage of the data. The programming of this device was carried out by using the LabVIEW 8.2 software. Figure 1 shows dynamic profiles for three main bioreactor variables; namely, inlet COD (CODin), VFA, and biogas production, for an experimental run of 160 days. Notice that three phases can be identified: start-up, transition and stationary regulation.

III. Detrended fluctuation analysis

Commonly, signals from biological systems exhibit a regular long-term trend accompanied by high-frequency fluctuations. In general, it has been considered that such physiological noise is useless for understanding the physiological mechanisms underlying biological system. In recent years, several studies have shown the opposite by applying different methods for characterizing signal fluctuations. More specifically, it has been recognized that signal fluctuations are complex, even exhibiting fractal properties. Motivated by this, Peng et al. (1994) developed a method for characterizing the fractal scaling properties of DNA sequences, finding that the serial behavior of the sequence is affected by long-term correlations. The method, called as detrended fluctuation analysis (DFA), has been successfully applied for evaluating fractality and scaling characteristics of non-stationary sequences. DFA method can be described as follow. Consider a time series of length N given by $Y=\{y_1, y_2, \dots, y_N\}$, where $y_i = y(t_i)$ and $\Delta t = t_i - t_{i-1}$ is sampling period. Then:

- Divide the data series into D subseries Y_M of length M, where $M=sN$, and $s \in (0,1)$. s=1 indicate that the sample corresponds to the whole sequence Y. Compute the sequences of the partial summations,
$$x(i) = \sum_{k=1}^{i} y_k - \bar{y} \tag{1}$$
where $\bar{y}$ is the mean of each subseries Y_M calculated as
$$\bar{y} = \frac{1}{M}\sum_{k}^{M} y_k \tag{2}$$
- A polynomial function of degree m, denoted by $x_{s,m}(i)$, is used to approximated sequence in each segment $x(i)$. The interpolating curve $x_{s,m}(i)$ represents the local trend in each segment. The trend of the integrated sequence is removed by computing the fluctuation sequence as
$$z_{s,m}(i) = x(i) - x_{s,m}(i) \tag{3}$$
- The fluctuation function $F_m(s)$ is computed as the standard deviation value of the sequence $z_{s,m}(i)$ as
$$F_m(s) = \sqrt{z_{s,m}(i)^2/N} \tag{4}$$
The previous steps are repeated for a broad range of segment lengths s.

According to the guidelines by Peng et al. (1994), the following range of scales s_{min}=5 and s_{max}=N/4 should be selected. When the signal follows a scaling law, a power-law behavior $F_m(s) \approx s^{\alpha m}$ can be observed, where α_m is called the scaling exponent and is interpreted as self-affinity parameter representing the long-range power-law correlation properties of the signal.

The scaling exponent α_m is computed as the slope of the plot $\log(F_m(s))$ versus $\log(s)$. If the data is uncorrelated, the plot is roughly a straight line with slope α_m=0.5. If α_m >0.5, the time series is persistent which is characterized by long-run memory immerse in the data-series. Persistence implies that if the signal increases or decreases, it will continue increasing or decreasing in the future, respectively. Conversely, if α_m <0.5 the autocorrelations in the signal are antipersistent (i.e., an increment is very likely to be followed by a decrement). The value α_m=1.0 and α_m=1.5 correspond to the so-called pink or 1/f-noise and to the Brownian motion, respectively. The relationship between the fractal dimension D_f of the time series and the scaling exponent α_m can be expressed as $D_f = D_T - \alpha_m$, where D_T is the topological dimension (for time series D_T=2.0). So, by finding the scaling exponent, one can estimate the fractal dimension of the time series (Peng et al., 1994).

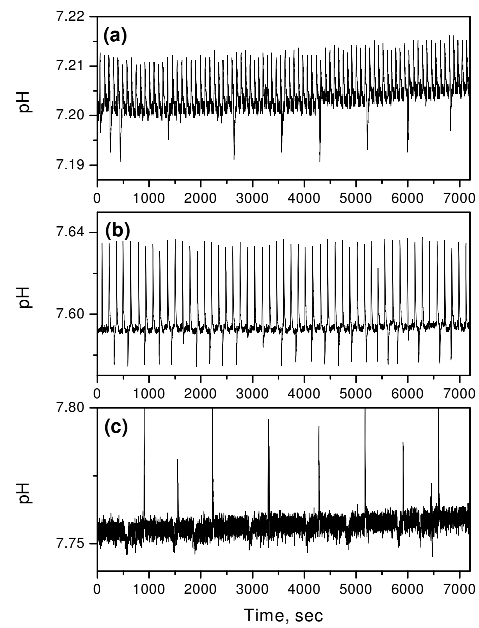

Figure 2. On-line pH time series recovered at different stages of AD process in the CSTR, a) 16 day, b) 54 day and c) 142 day.

IV. Results and discussion

The results obtained by the application of the fractal DFA on pH time series collected from the lab-scale wastewater treatment bioreactors used for the treatment of tequila vinasses are presented. The analyzed pH time series corresponds to two hours data sets taken at different stages of the bioreactor operation. Figure 2 shows three pH time series recovered in different stages of an experimental run, where it can be seen that the pH signals show complex fluctuations around their corresponding set-point values and several non-continuous perturbations along the time series. These fluctuations and discontinuities may be related to the regulation and correction of pH via the on-off controller, as well as the several reactions that are occurring simultaneously.

For our study, linear detrending (i.e., m=1) was used for estimating the scaling exponent. In this way, for simplicity in notation, the fluctuation function and the corresponding scaling exponent will be denoted by $F(s)$ and α_m, respectively. The fractal time series analysis was performed on pH time series data sets at the three different stages of the experimental run observed in Figure 1: start-up phase (days 30, 32, 34 and 36), transition phase (days 100, 102, 104 and 106) and stationary regulation phases (days 150, 152, 154 and 156). Figure 3 shows the fractal analysis results for the start-up phase, where it is observed that the fluctuation function $F(s)$ exhibits important variations and cannot be described by a uniform power law for all time scales. In fact, four different scaling regions can be clearly observed, at the scaling intervals of 4-17, 17-82, 82-510 and 510-2000 seconds, which is denoted as A, B, C and D, respectively. Each scaling region can be described with a different single scaling exponent value with its corresponding fractal dimension, denoted by $D_{f,A}$, $D_{f,B}$, $D_{f,C}$ and $D_{f,D}$, respectively.

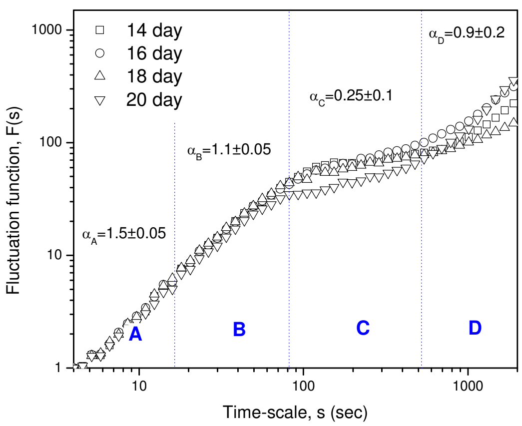

Figure 3. Fluctuation function as a function of the scale s for four pH time series. Notice that four scaling regions can be observed.

For transient and regulation phases, Figure 4 shows the fractal analysis, where it can be observed the same four scaling regions found in the start-up phase. This result indicates that the underlying physical phenomena in pH series cannot be described by a unique global fractal dimension, and that the degradation of tequila vinasses in anaerobic CSTR type digester is dominated by four principal phenomena. Fractal parameters for the three stages are summarized in Table 1.

For the start-up phase, scaling exponent values for Region A and B are 1.5±0.05 and 1.1±0.05 respectively, indicating strong long-range correlations for small time scales. For Region C, the scaling exponent value is 0.25±0.1, indicating anti-persistence behavior. Finally, the scaling exponent in Region D is 0.9±0.2, showing larger fluctuations than regions A, B and C, indicating a major complexity of physical phenomena in large time-scales. In general, fractal dimension values for Regions A exhibits constant fluctuations, from the start-up phase to the transition phase, increasing its maximum point in the regulation phase at 142 days. For Region C, fractal dimension values show a gradual decreasing, with moderate fluctuations, from the start-phase to the regulation phase. Although fractal dimension values exhibit considerable fluctuations in Region D, a clear trend can be observed. From the start-up phase to the transition phase, the fractal dimension increases abruptly from 1.0 to 1.8. From the transition phase to the regulation phase, the fractal dimension also presents a significant change from 1.8 to 1.4. Dynamical changes in fractal parameters displays similar trends than physical parameters of AD process showed in Figure 1.

Table 1. Fractal dimension values of the CSTR-type digester

Day	$D_{f,A}$	$D_{f,B}$	$D_{f,C}$	$D_{f,D}$
14	0.615	1.168	1.787	0.290
16	0.630	1.155	1.647	1.045
18	0.590	1.152	1.714	1.087
50	0.532	0.940	1.808	1.579
52	0.527	1.073	1.771	1.958
54	0.531	1.090	1.783	1.673
80	0.596	0.913	1.618	1.897
82	0.529	0.923	1.844	1.804
84	0.601	0.935	1.678	1.965
140	0.915	0.848	1.262	1.301
142	0.947	0.837	1.322	1.406
144	0.622	0.818	1.250	1.348

In particular, qualitative relations between $D_{f,A}$-COD inlet concentration, $D_{f,C}$-VFA concentration and $D_{f,D}$-biogas production are observed, , such that fractal parameters obtained with DFA of pH time series from CSTR type digester treating tequila vinasses can be used as qualitative monitoring indexes of important variables in the AD process.

V. CONCLUSIONS

This work proposed the application of fractal fluctuation analysis to pH time series from the anaerobic treatment of tequila vinasses in two continuous bioreactor configurations: up-flow fixed bed reactor and a CSTR-type digester. The fluctuation analysis allows the identification of four regions, suggesting four underlying phenomena in the continuous anaerobic digestion treatment of tequila vinasses. The dynamical changes in fractal parameters, scaling exponent values and fractal dimensions, obtained from the fluctuation analysis allow the qualitative description of the temporal evolution in three of key operation variables in AD processes: COD inlet concentration, total VFA concentration, and biogas production. The monitoring of these variables is of the great importance in the anaerobic digestion process. However, the on-line monitoring of these variables is expensive and requires considerable analysis time, such that our results provide a fast and reliable method to provide useful information in continuous AD processes.

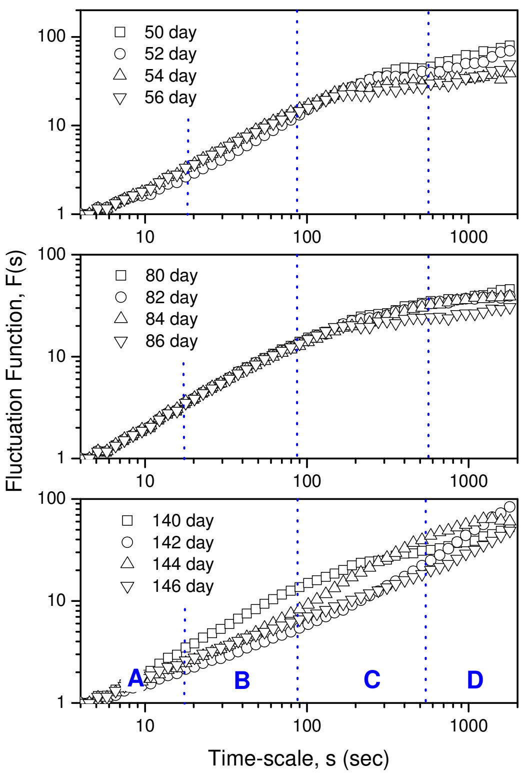

Figure 4. Fluctuation functions for a) 50-56 days, b) 80-86 days and c) 140-146 days. Notice that the slopes of each region exhibit considerable dynamical changes.

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