Modelling and $L_1$ Adaptive Control of pH in Bioethanol Enzymatic Process

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Abstract—The enzymatic process is a key step in second generation bioethanol production. Pretreated biomass fibers are liquefied with the help of enzymes to facilitate fermentation. Enzymes are very sensitive to pH and temperature and the main control challenge in the nonlinear process is to ensure minimum deviations from the optimal pH level.

This article develops a mathematical model for the pH, which has not been reported earlier for this particular process. The new model embeds flow dynamics and pH calculations and serves both for simulation and control design. Two control strategies are then formulated for pH level regulation: one serves both for simulation and control design. Two control schemes are then designed, a PI controller and an $L_1$ adaptive output feedback controller. Model-based feed-forward terms are added to the controllers to enhance their performances. A new tuning method of the $L_1$ adaptive controller is also proposed. Further, a new performance function is formulated and tailored to this type of processes and is used to monitor the performances of the process in closed loop. The $L_1$ design is found to outperform the PI controller in all tests.

I. INTRODUCTION

Bioethanol is thought to become the primary renewable liquid fuel [1], [2] and extensive endeavors have been conducted to make the production process feasible on a large scale [3]. In order to reduce operating costs, bio-refineries are integrated with power plants following the IBUS principle [4], [5]. The conversion of lignocellulosic biomass to ethanol is performed only with steam and enzymes, therefore categorizing the technology as a green process.

In a second generation bioethanol production process, the enzymatic liquefaction step prepares the pretreated biomass for fermentation [4], [5]. In the pretreatment phase, the biomass is soaked with acetic acid and then pretreated with steam. Studies show that a steam/acetic acid combination improves the pretreatment process [6] and it will be considered that the stream of fibers is mildly acidic, thus lowering the pH level. Acetic acid is also produced in reduced quantities during the pretreatment stage.

The enzyme activity is influenced both by pH and temperature. Enzyme activity versus pH follows a certain bell-shaped curve [7]. Optimal pH activity differs for each enzyme type. For example, in the case of Accellerase TRIO, a pH level of 4.8 should be set 1. Another type of enzymes, i.e. Cellic CTec3 produced by Novozymes, requires a pH of 5.0 2. A small deviation from the pH optimal value, e.g. 0.2, can cause a significant drop in the process efficiency, e.g. 20%. Therefore, the main control challenge in such a process is to minimize deviations from the optimal pH level.

Controlling the pH has been the topic of many research activities and there are many generic solutions in the literature. A comprehensive review of the existing generic control strategies can be found in [8]. Control strategies vary from pure feed-forward control, where the inflow of hydroxide is manually adjusted by an operator, to more sophisticated schemes like adaptive fuzzy control [9], nonlinear adaptive control [10], [11] or model predictive control [12], [13]. The most common application of pH control is the neutralization process but, in an enzymatic liquefaction process, it is desired to keep the pH level somewhere in the range 4-6 depending on the enzyme type.

To our knowledge, pH modelling for an enzymatic liquefaction process has not been conducted earlier. Therefore, a process model is first formulated. Two control schemes are then designed, a PI controller and an $L_1$ adaptive output feedback controller. Classical PI control is widely spread in industry and is used as a reference for control performance. The $L_1$ adaptive controller represents a novelty in control theory and has never been applied to pH control of liquefaction processes. Major disturbances will be identified and model based feed-forward terms will be added to the controllers in order to improve their action. Also, a new tuning method of the $L_1$ controller is proposed as an enhancement of the method developed in [14]. Finally, the controllers are tested on large scale benchmark scenarios that cover both reference tracking and disturbance rejection cases. A performance function is formulated based on the enzymatic activity curve and the controllers are compared using the enzymatic efficiency associated with their action.

II. PROCESS DESCRIPTION

A generic enzymatic liquefaction process is illustrated in Figure 1.

The pretreated biomass was decomposed into fibers in the pretreatment stage and arrives in the liquefaction reactor having a high dry matter content TS typically between 25–30%. A high dry matter is required in order to make the technology cost-effective but it cannot increase indefinitely due to mixing technical problems that may appear [4], [15]. A sample of fibers is extracted automatically and
an NIR analysis is performed in order to determine its chemical composition, including the concentration of acetic acid denoted as \([\text{AcH}]_0\) or \(C_{A0}\), which is of interest in this case. The mass flow of fibers is also directly measured and denoted as \(F_{FF}\).

\(pH\) control is achieved by pumping a strong base i.e. sodium hydroxide, \(\text{NaOH}\), into the fiber fraction before entering the liquefaction tank. The inflow of base is measured as \(F_B\) and its concentration is 10 wt%.

Enzymes are pumped into the reactor proportional to cellulose quantity and the inflow of enzymes is measured as \(F_E\). A quantity of water \(F_W\) is also poured into the tank. The weight of the tank, the outflow and the \(pH\) value are directly measured as \(M_{FM}\), \(F_{FM}\), and \(pH\), respectively. Nominal values of the inflows, acetic acid concentration, tank load and total solids are presented in Table I. The data refer to a bioethanol plant that was designed to handle 1000 kg h\(^{-1}\) of fibers. The enzymatic liquefaction tank has a nominal load capacity of 5000 kg of fiber mash and the concentration of acetic acid varies around 5 g kg\(^{-1}\) depending on the pretreatment process parameters.

| \(F_{FF}\) | 1000 kg h\(^{-1}\) | \([\text{AcH}]_0\) | 5 g kg\(^{-1}\) |
| \(F_E\)  | 20 kg h\(^{-1}\)  | \(M_{FM}\)  | 5000 kg |
| \(F_W\)  | 80 kg h\(^{-1}\)  | TS           | 25 %    |

### Control Challenge

Several theoretical enzymatic activity bell-shaped curves are shown in Figure 2. The optimal \(pH\) level corresponds to an enzymatic activity of 100 %. In case of disturbances that occur in the pretreatment process, the acetic acid concentration in the inflow changes and affects the \(pH\) level. A small deviation from the optimal \(pH\) level can cause a significant drop in the enzymatic activity. This means that the quality of the outflow drops. An increase in enzyme quantity is necessary to meet the same quality constraints on the outflow but enzymes are very expensive. It is a lot cheaper to properly control the \(pH\) level with a base.

Another control challenge arises from the large scale nature of the process. The \(pH\) sensor is positioned on the outflow of the tank because it is easier to measure the \(pH\) level on a liquefied substance. The flow dynamics of the reactor are slow and a large quantity of fibers can be compromised due to a small deviation in the \(pH\) level. Therefore, a model and feed-forward terms for the controllers are necessary to obtain a high performance control strategy.

### IV. Process Model

The \(pH\) calculation is derived using the classical physico-chemical approach, which considers a set of weak acid/base equilibrium in liquid phase [16] and gas-liquid CO\(_2\) stripping process [17]. It is assumed that CO\(_2\) is not produced in the enzymatic liquefaction process but traces of bicarbonate can exist in the inflows due to upstream subprocesses or process water utilization. The process model captures flow dynamics and considers the mixture in chemical equilibrium at any given time.

#### A. \(pH\) Calculation

In total, the model has 6 weak acid/base equilibrium equations and CO\(_2\) stripping. It is also assumed that production and consumption of ions are negligible from enzymatic reactions.

Acetic acid is a weak acid and partially dissociates into Ac\(^-\) and H\(^+\) with an equilibrium constant \(K_A\) (1a). Sodium hydroxide is a strong base and fully dissociates into Na\(^+\) and OH\(^-\) (1b). Ac\(^-\) and Na\(^+\) combine to form the salt sodium acetate NaAc (1c). Water self-ionizes with an equilibrium constant \(K_W\) (1d). The liquid phase is not pure and is considered to contain CO\(_2\), which is the cause of a buffer formation that affects the \(pH\) level. Carbon dioxide forms carbonic acid H\(_2\)CO\(_3\) in water, which dissociates with an equilibrium constant \(K_{C2}\) into bicarbonate HCO\(_3^-\) and H\(^+\) as in (1e). The bicarbonate continues to decompose forming carbonate CO\(_3^{2-}\) and H\(^+\) with an equilibrium constant \(K_{C3}\) like in (1f).

\[
\text{AcH} \rightleftharpoons \text{Ac}^- + \text{H}^+ \quad (1a) \\
\text{NaOH} \rightarrow \text{Na}^+ + \text{OH}^- \quad (1b) \\
\text{Ac}^- + \text{Na}^+ \rightarrow \text{NaAc} \quad (1c)
\]
Three variables are defined:

\[ [A_T] = [\text{AcH}] + [\text{Ac}^-] \quad (2a) \]
\[ [B_T] = [\text{Na}^+] + [\text{NaOH}] \quad (2b) \]
\[ [C_T] = [\text{CO}_2] + [\text{HCO}_3^-] + [\text{CO}_3^{2-}] \quad (2c) \]

The presence of other acids like lactic acid or succinic acid was also recorded but in negligible amounts and there might be other buffers in the stream. Therefore, the concentration of all unmodeled ions will be lumped into a single variable \([Z^-]\) and the charge balance is then formulated:

\[
[H^+] - [Z^-] + [Na^+] -
- [OH^-] - [Ac^-] - [HCO_3^-] - 2[CO_3^{2-}] = 0 \quad (3)
\]

Each term from the charge balance can be found as a function of variables \([A_T], [B_T], [C_T]\) or \([H^+]\) and equilibrium constants \(K_A, K_W, K_C, \text{and} K_{C_2}\). Therefore, \([H^+]\) can be determined by finding the real positive zero of (3). Afterwards, the pH level is computed using its definition:

\[
\text{pH} \overset{\text{def}}{=} -\log_{10}[H^+] \quad (4)
\]

There might be an offset between the estimated and measured pH level. The difference can be canceled by adjusting the concentration of \(Z^-\) i.e. \(C_{Z_0}\). An online estimation of \([Z^-]\) could be implemented in reality.

**B. State Space Model**

In this section, the flow dynamics and the pH calculation are embedded into a single model that has the structure from Figure 3. It is important to conduct all pH calculations only on the liquid part of the mixture. All notations are gathered in Table II with their measuring units.

\[
\begin{align*}
\text{F}_F & \quad C_{A_0} \quad F_E \quad C_{Z_0} \\
F_B & \quad C_{B_0} \\
F_W & \quad T_S \quad F_{FM}
\end{align*}
\]

**Fig. 3. Process flow scheme.**

Perfect and instantaneous mixing of the tank is assumed and this is represented as an Ideal Mixing block, which combines all inflows into a single input \(u_i\) without solids. The outflow without solids is denoted as \(u_o\). The inflow concentrations \(u_{a0}, u_{b0}\) and \(u_{c0}\) include a measuring unit transformation and are expressed in mol L\(^{-1}\).

The state variables of the dynamic model are defined as:

\[
x_z = [Z^-] \quad (5a)
\]

and \(x_m\), which is the mass of liquid inside the reactor.

Two more algebraic variables are also defined:

\[
x_h = [H^+] \quad (6a)
\]
\[
x_{CO_2} = [CO_2] \quad (6b)
\]

The system dynamics read as:

\[
\frac{dx_m}{dt} = u_i - u_o \quad (7a)
\]
\[
\frac{d(x_m x_z)}{dt} = u_i u_{a0} - u_o x_z \quad (7b)
\]
\[
\frac{d(x_m x_{a1})}{dt} = u_i u_{a0} - u_o x_{a1} \quad (7c)
\]
\[
\frac{d(x_m x_{b1})}{dt} = u_i u_{b0} - u_o x_{b1} \quad (7d)
\]
\[
\frac{d(x_m x_{c1})}{dt} = u_i u_{c0} - u_o x_{c1} + r_{ctr} x_m \quad (7e)
\]

where \(r_{ctr}\) is the CO\(_2\) stripping rate [17]:

\[
r_{ctr} = k_{ctr} \left( u^*_{co2} - x_{co2} \right) \quad (8)
\]

The dissolved concentration of CO\(_2\) is denoted as \(u^*_{co2}\) and is governed by the Henry law [17].

**TABLE II**

**EXPLANATION OF SYMBOLS THAT WERE USED IN THE PROCESS MODEL.**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>(u_i)</td>
<td>Liquid part of fibre fraction inflow</td>
<td>kg h(^{-1})</td>
</tr>
<tr>
<td>(u_o)</td>
<td>Liquid part of fibre mash outflow</td>
<td>kg h(^{-1})</td>
</tr>
<tr>
<td>(u_{a0})</td>
<td>Inflow concentration of acetic acid</td>
<td>mol L(^{-1})</td>
</tr>
<tr>
<td>(u_{b0})</td>
<td>Inflow concentration of sodium hydroxide</td>
<td>mol L(^{-1})</td>
</tr>
<tr>
<td>(u_{c0})</td>
<td>Inflow concentration of carbon dioxide</td>
<td>mol L(^{-1})</td>
</tr>
<tr>
<td>(x_m)</td>
<td>Mass of liquid fibre mash</td>
<td>kg</td>
</tr>
<tr>
<td>(x_{a1})</td>
<td>Total molar concentration of acid species</td>
<td>mol L(^{-1})</td>
</tr>
<tr>
<td>(x_{b1})</td>
<td>Total molar concentration of base species</td>
<td>mol L(^{-1})</td>
</tr>
<tr>
<td>(x_{c1})</td>
<td>Total molar concentration of carionic species</td>
<td>mol L(^{-1})</td>
</tr>
<tr>
<td>(x_h)</td>
<td>Molar concentration of unmodelled ions</td>
<td>mol L(^{-1})</td>
</tr>
<tr>
<td>(x_{co2})</td>
<td>Total molar concentration of CO(_2)</td>
<td>mol L(^{-1})</td>
</tr>
<tr>
<td>(u_{co2})</td>
<td>Molar concentration of CO(_2) in the atmosphere</td>
<td>mol L(^{-1})</td>
</tr>
<tr>
<td>(k_{ctr})</td>
<td>CO(_2) stripping process parameter</td>
<td>h(^{-1})</td>
</tr>
<tr>
<td>(K_W)</td>
<td>H(_2)O dissociation constant</td>
<td>-</td>
</tr>
<tr>
<td>(K_A)</td>
<td>AcH dissociation constant</td>
<td>-</td>
</tr>
<tr>
<td>(K_{C_1})</td>
<td>CO(_2) dissociation constant</td>
<td>-</td>
</tr>
<tr>
<td>(K_{C_2})</td>
<td>HCO(_3) dissociation constant</td>
<td>-</td>
</tr>
<tr>
<td>(\rho_{FM})</td>
<td>Fiber mash density</td>
<td>kg L(^{-1})</td>
</tr>
<tr>
<td>(M_A)</td>
<td>Molar mass of AcH</td>
<td>g mol(^{-1})</td>
</tr>
<tr>
<td>(M_B)</td>
<td>Molar mass of NaOH</td>
<td>g mol(^{-1})</td>
</tr>
<tr>
<td>(C_{A_0})</td>
<td>Inflow concentration of AcH</td>
<td>g kg(^{-1})</td>
</tr>
<tr>
<td>(C_{B_0})</td>
<td>Inflow concentration of NaOH</td>
<td>g kg(^{-1})</td>
</tr>
<tr>
<td>(C_{C_0})</td>
<td>Inflow concentration of CO(_2)</td>
<td>g kg(^{-1})</td>
</tr>
<tr>
<td>(C_{Z_0})</td>
<td>Inflow concentration of Z(^{-})</td>
<td>mol kg(^{-1})</td>
</tr>
<tr>
<td>(T_S)</td>
<td>Total solids in inflow stream</td>
<td>%</td>
</tr>
</tbody>
</table>

The charge balance can be rewritten as a polynomial:

\[ x_h^5 + p_1 x_h^4 + p_2 x_h^3 + p_3 x_h^2 + p_4 x_h + p_5 = 0 \quad (9) \]

where coefficients \(p_i\) are found by identification after expanding equation (3). The nonlinearity of the process arises from
the fact that coefficients \( p_i \) are functions of model states and change in time. \( x_h \) is then determined as the positive real zero of polynomial (9) and the output of the model is defined as:
\[
y_{pH} = -\log_{10} x_h
\]  

(10)

The numerical values of all model parameters are listed in Table III.

| \( K_W \) | \( 1 \times 10^{-14} \) | \( K_A \) | \( 1.7378 \times 10^{-5} \) | \( M_A \) | \( 60.052 \) | \( 21 \) \( g \) \( mol^{-1} \) | \( M_B \) | \( 39.997 \) | \( 15 \) \( g \) \( mol^{-1} \) | \( M_C \) | \( 44.01 \) | \( g \) \( mol^{-1} \) |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| \( K_{C1} \) | \( 4.3003 \times 10^{-7} \) | \( C_{A0} \) | \( 5 \) | \( g \) \( kg^{-1} \) |
| \( K_{C2} \) | \( 4.7995 \times 10^{-11} \) | \( C_{B0} \) | \( 100 \) | \( g \) \( kg^{-1} \) |
| \( u_{e0} \) | \( 1.71 \times 10^{-5} \) \( mol \) \( L^{-1} \) | \( C_{C0} \) | \( 7.1673 \times 10^{-4} \) | \( g \) \( mol^{-1} \) |
| \( \rho_{FM} \) | \( 1.05 \) \( kg \) \( L^{-1} \) | \( Z_{C} \) | \( 0.08 \) | \( mol \) \( L^{-1} \) |

**TABLE III**

**MODEL PARAMETERS.**

C. Titration Simulation

The model is tested by performing a classical titration simulation, i.e. feeding into the process an inflow of hydroxide resembling a stairway shape with a stair amplitude of 1 \( kg \) \( h^{-1} \) starting at time \( t = 20 \) h. The concentration of \( Z^- \) is disregarded. The results can be observed in Figure 4.

![Fig. 4. Titration simulation for verifying the implementation of the model.](image)

The acetic acid and the carbonic acid keep a low pH but after adding a substantial amount of base, approximately at time \( t = 500 \) h, the buffers are depleted and the pH increases to a value greater than 7, which is an expected result.

V. CONTROL DESIGN

The enzymatic liquefaction tank is assumed to have a mass controller, which can be easily constructed as a feed-forward strategy combined with feedback.

A bioethanol plant may switch between enzyme types but such an action does not happen often during production mode. Therefore, reference tracking is necessary but emphasis will be placed on disturbance rejection. There are many sources of disturbances that affect the pH level, e.g. pretreatment conditions, which influence the concentration of acetic acid, the presence of other weak acids and negative ions or the imperfect mixing effects, which might be significant since the tank has a large volume and a homogenous environment cannot be guaranteed.

A. Feed-forward Combined With PI Control

In this section, a classical PI controller combined with a nonlinear feed-forward signal is derived following a traditional design algorithm, i.e. linearization of the process model around the nominal operational point from Table I with \( pH = 5 \), and derivation of the controller using the Skogestad internal model control (SIMC) approach. The pH level is mainly disturbed by the initial concentration of AcH, which is measured through NIR analysis. A feed-forward term can be created using the nonlinear model to compensate for this type of disturbance. The block diagram of the closed loop system is shown in Figure 5.

![Fig. 5. Closed loop system with a feed-forward and a PI controller.](image)

The PI control law is defined as:
\[
u_{pI}(t) = K_P e(t) + K_I \int_0^t e(t) dt
\]  

(11)

where \( u_{pI}(t) \) is the feedback contribution to the flow of hydroxide, \( e(t) \) is the pH error signal defined as \( e(t) = r_{pH}(t) - y_{pH}(t) \), \( r_{pH}(t) \) is the pH reference signal, \( K_P \) is the proportional gain and \( K_I \) is the integral gain. Following SIMC rules [18], \( K_P \) and \( K_I \) were set to:
\[
K_P = 22.8 \) kg/h/pH unit \n\]  

(12a)

\[
K_I = 16.8 \) kg/h/pH unit \n\]  

(12b)

The feed-forward term is found from the charge balance (3) from where \( [Na^+] \) or \( u_{e0} \) is isolated:
\[
[Na^+] = -(H^+)[OH^-] + [Ac^-] + [HCO_3^-] + 2[CO_3^{2-}] + [Z^-]
\]  

(13)

The required concentration of \( [H^+] \) is found from the reference level:
\[
[H^+] = 10^{-r_{pH}}
\]  

(14)

and all the other concentrations are derived with the help of \( [H^+] \) and using the steady-state values for \( [C_T] \) and \( [A_T] \).
The contribution of the feed-forward term to the total inflow of hydroxide can then be computed. The total flow of base becomes:

$$F_{OH_i} = u_{FF} + u_{PI}$$  \hspace{1cm} (15)$$

where $$u_{FF}$$ is the feed-forward contribution and $$u_{PI}$$ is computed by the controller using feedback action.

B. $$L_1$$ Adaptive Control

The $$L_1$$ adaptive controller is a modified version of the model reference adaptive controller with a state predictor. In the MRAC architecture, the key to a high performance control strategy is the adaptation gain, which is subject to a compromise and there are no systematic ways of finding an optimal value. A high adaptation gain introduces high-frequency noise in the control channel and the stability margins are affected. In order to separate robustness from adaptation performance, a filter $$C(s)$$ is introduced in the control channel [19]. The analysis of the new controller i.e. the computation of the uniform bounds on outputs and control signals, is performed using the $$L_1$$ norm, hence the name of $$L_1$$ adaptive controller.

The process in open loop can be expressed as follows [19]:

$$y(s) = A(s)\{u(s) + d(s)\}$$  \hspace{1cm} (16)$$

where $$y(s)$$ is the pH level, $$A(s)$$ is an unknown transfer function, $$u(s)$$ is the system input and $$d(s)$$ lumps all the uncertainties and disturbances that affect $$A(s)$$. Transfer function $$A(s)$$ can be approximated as the linearized model around the nominal point from Table I with a base flow set such that $$pH = 5$$.

The $$L_1$$ adaptive output feedback controller consists of an output predictor, an adaptation law and a control law [20], [19]. The structure of the closed loop system with this type of controller is presented in Figure 6.

The output predictor is built with the help of a first order model reference system:

$$\hat{y}(t) = -m\hat{y}(t) + m\{u(t) + \hat{\sigma}(t)\}$$  \hspace{1cm} (17)$$

where $$\hat{y}(t)$$ is the output estimation, $$1/m$$ is the desired time constant of the closed loop system, $$u(t)$$ is the inflow of hydroxide and $$\hat{\sigma}(t)$$ is the estimation of all uncertainties and unmodelled dynamics. The model reference system is also denoted as:

$$M(s) = \frac{m}{s + m}$$  \hspace{1cm} (18)$$

System (16) can be rewritten in terms of the model reference system [19]:

$$y(s) = M(s)\{u(s) + \sigma(s)\}$$  \hspace{1cm} (19)$$

where $$\sigma(s)$$ is identified as [19]:

$$\sigma(s) = \frac{(A(s) - M(s))u(s) + A(s)d(s)}{M(s)}$$  \hspace{1cm} (20)$$

The idea is to cancel all the uncertainties $$\sigma(s)$$ with the help of the control signal $$u(s)$$. Therefore, the control signal is defined as [19]:

$$u(s) = C(s)\{r(s) - \hat{\sigma}(s)\}$$  \hspace{1cm} (21)$$

where $$C(s)$$ is a first order filter in this application:

$$C(s) = \frac{c}{s + c}$$  \hspace{1cm} (22)$$

If Equation (21) is substituted in (19) then:

$$y(s) = M(s)C(s)r(s) + M(s)\{\sigma(s) - C(s)\hat{\sigma}(s)\}$$  \hspace{1cm} (23)$$

If $$\sigma(s)$$ is perfectly estimated then $$\hat{\sigma}(s) = \sigma(s)$$ and the disturbances will be rejected only in the bandwidth of $$C(s)$$.

The adaptive estimate $$\hat{\sigma}(t)$$ is updated using a projection algorithm that ensures boundedness of the estimate within a given ball [19]:

$$\hat{\sigma}(t) = \Gamma \text{Proj} (\hat{\sigma}(t) - \bar{y}(t))$$  \hspace{1cm} (24)$$

where $$\bar{y}(t) = \hat{y}(t) - \hat{y}(t)$$ is the estimation error of the output.

There are 3 parameters to set for the $$L_1$$ adaptive output feedback controller, i.e. the desired closed loop time constant $$1/m$$, the adaptation gain $$\Gamma$$ and the eigenfrequency of the control filter $$c$$.

The reference model and the control signal filter can be designed systematically [14]. Assuming perfect knowledge of disturbances, an ideal system $$y(s)$$ can be built and used for tuning [14]:

$$y(s) = H(s)C(s)r(s) + H(s)\{1 - C(s)\}d(s)$$  \hspace{1cm} (25)$$

where $$H(s)C(s)$$ is the transfer function from $$r(s)$$ to $$y(s)$$ and $$H(s)$$ is defined as:

$$H(s) = \frac{A(s)M(s)}{C(s)A(s) + \{1 - C(s)\}M(s)}$$  \hspace{1cm} (26)$$

Parameters $$m$$ and $$c$$ must be chosen such that $$H(s)$$ is stable and the following $$L_1$$ norm holds [19]:

$$\|G(s)\|_{L_1} < 1$$  \hspace{1cm} (27)$$
where \( G(s) = H(s)\{1 - C(s)\} \) is the transfer function from \( d(s) \) to \( y(s) \) and \( L \) is the Lipschitz constant required to guarantee BIBO stability (Lemma 4.1.1 in [19]).

By considering \( A(s) \) as the linearized system around the nominal point, a \((m, c)\) map can be built in order to facilitate the choice of these two parameters. The closed loop system has several poles, which can cause oscillations if they are complex. As a criterion for \((m, c)\) determination, the worst damping ratio \( \zeta \) should be higher than 0.7 in order to ensure acceptable oscillations. The damping ratio \( \zeta \) is plotted as a function of \((m, c)\) in the top plot of Figure 7.

In order to accelerate the system, parameter \( m \) should be increased. To reduce oscillations in the system response, the bandwidth of filter \( C(s) \) needs to be enlarged such that a more responsive control action would be allowed. The pair \((0.6, 60)\) is chosen for this application, which corresponds to a \( \zeta > 0.9: \)

\[
m = 0.6 \quad c = 60 \tag{28}
\]

The model reference has a time constant that corresponds to 1.6 h. The Lipschitz constant, for the selected \( m \) and \( c \), is computed to be:

\[
L = 78.6851 \tag{29}
\]

which ensures a BIBO closed loop system according to Lemma 4.1.1 [19].

In theorem 4.1.1 from [19] it is shown that the tracking error between the real and the ideal system, which assumes perfect knowledge of the disturbance, is uniformly bounded with respect to a constant proportional to \( 1/\sqrt{\Gamma} \). Therefore, a high adaptation gain \( \Gamma \) is desired. At the same time, the stability and dynamics of \( \dot{\sigma} \) are dependent on \( \Gamma \). The transfer function from \( r(s) \) and \( d(s) \) to \( \dot{\sigma}(s) \) is [14]:

\[
\dot{\sigma} = F(s)[C(s)\{A(s) - M(s)\}r(s) + A(s)d(s)] \tag{30}
\]

where \( F(s) \) is identified as:

\[
F(s) = \frac{1}{\Gamma s + C(s)A(s) + \{1 - C(s)\}M(s)} \tag{31}
\]

\( \Gamma \) should be chosen such that \( F(s) \) is stable. To reduce the high frequency noise due to adaptation, \( \Gamma \) can be set such that the worst damping ratio of \( F(s) \), denoted as \( \zeta_\Gamma \), would be greater than 0.7. A plot of \( \zeta_\Gamma \) as a function of \( \Gamma \) is shown in the bottom plot of Figure 7. A value \( \Gamma = 1000 \) would be a good choice for the current application ensuring a damping ratio of \( \dot{\sigma} \) close to 1 and a relatively high adaptation gain.

Following the described procedure, the \( L_1 \) controller can be tuned in different pH operation points and parameters \( m, c \) and \( \Gamma \) could be adjusted in real time in order to maximize performances. Table IV contains the controller parameters in 3 different operating points.

### TABLE IV

<table>
<thead>
<tr>
<th>( pH )</th>
<th>( m )</th>
<th>( c )</th>
<th>( \Gamma )</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>1</td>
<td>50</td>
<td>800</td>
</tr>
<tr>
<td>5</td>
<td>0.6</td>
<td>60</td>
<td>1000</td>
</tr>
<tr>
<td>7</td>
<td>4</td>
<td>80</td>
<td>500</td>
</tr>
</tbody>
</table>

The feed-forward term developed in the classical control section is also added to the \( L_1 \) controller to test its efficiency.

### VI. BENCHMARK TESTS

Reference tracking is tested by performing a square wave of magnitude 2 as in Table V. The tests cover the entire pH interval \( 3 - 7 \), which includes most of the nonlinearity from the titration curve. Even though enzymes normally operate around \( pH = 5 \), it is of theoretical interest to test the controller for a wider range of operating points.

### TABLE V

REFERENCE TRACKING SQUARE WAVE SCENARIO.

<table>
<thead>
<tr>
<th>#</th>
<th>( r_{pH} )</th>
<th>Time interval</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>5</td>
<td>0 - 20</td>
</tr>
<tr>
<td>2.</td>
<td>( \uparrow 7 )</td>
<td>20 - 40</td>
</tr>
<tr>
<td>3.</td>
<td>( \downarrow 5 )</td>
<td>40 - 60</td>
</tr>
<tr>
<td>4.</td>
<td>( \downarrow 3 )</td>
<td>60 - 80</td>
</tr>
<tr>
<td>5.</td>
<td>( \uparrow 5 )</td>
<td>80 - 100</td>
</tr>
</tbody>
</table>

The disturbance rejection scenario is presented in Table VI and includes white noise perturbations with 0 mean and standard deviation \( \sigma \) on the feed concentrations of acetic acid, base and unknown buffers.

### TABLE VI

WHITE NOISE DISTURBANCE REJECTION SCENARIOS.

<table>
<thead>
<tr>
<th>#</th>
<th>Scenario</th>
<th>( \sigma )</th>
<th>Time interval</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>Acid disturbances</td>
<td>( 1 ) ( g ) ( kg^{-1} )</td>
<td>0 - 133</td>
</tr>
<tr>
<td>2.</td>
<td>Base disturbances</td>
<td>( 30 ) ( g ) ( kg^{-1} )</td>
<td>133 - 266</td>
</tr>
<tr>
<td>3.</td>
<td>Unknown buffers</td>
<td>( 0.02 ) ( mol ) ( L^{-1} )</td>
<td>266 - 400</td>
</tr>
</tbody>
</table>
Several theoretical bell shaped enzymatic activity curves were shown in Figure 2. The maximum efficiency of the enzymatic process is considered to be 1 and is reached when the pH level equals the optimal value. A small deviation from the optimal level can cause a significant drop in the enzymatic activity. The monitoring cost function is constructed by integrating the deviations from maximum enzymatic activity within a time window:

$$ J = \int_{t_0}^{t_1} \{1 - E(y_{pH})\} d\tau $$

(32)

where \( t_0 \) is the initial time, \( t_1 \) is the final time and \( E(y_{pH}) \) is the enzymatic activity associated with pH level \( y_{pH} \) and can be approximated with a Gaussian bell-shaped curve:

$$ E(y_{pH}) = \frac{1}{\sigma \sqrt{2\pi}} \cdot \exp \left(-0.5 \left( \frac{y_{pH} - \mu}{\sigma} \right)^2 \right) $$

(33)

where \( \sigma = 0.2 \) and \( \mu = 5 \) for this application. These values correspond to \( E_2 \) from Figure 2. The enzymatic activity is usually experimentally determined and can be represented as a table based map in reality.

Both control strategies are tested in the scenarios described above and the results are commented in the next section.

VII. RESULTS

The results for the reference tracking scenario can be viewed in Figures 8 and 10.

The classical control strategy has overshoots that increase as the system moves further from the design point. On the other hand, the \( \mathcal{L}_1 \) controller has a significant overshoot only in the neutral area of \( pH = 7 \). Otherwise, the \( \mathcal{L}_1 \) controller responds better than the PI controller following the model reference. The overshoots could be accommodated by a reference filter in both cases. The control signal is within acceptable limits.
The disturbance rejection scenario is shown in Figures 9 and 11 and the evaluation of the cost function is performed in Table VII. In the case of measured disturbances, i.e. acetic acid disturbances, the $L_1$ controller with feed-forward performs better than the PI controller leading to a full rejection of these disturbances. The nonlinear feed-forward term significantly helps both controllers.

In the second scenario, i.e. base disturbances, the $L_1$ controller has better results leading to smaller variations of the pH level, thus to a much lower $J$ value. The feed-forward term does not help in this case.

In the last scenario, i.e. unknown buffers, the $L_1$ controller outperforms the classical PI. Overall, regarding variations in the pH level, the $L_1$ controller has better performances.

<table>
<thead>
<tr>
<th>Scenario</th>
<th>PI</th>
<th>PI+FF</th>
<th>$L_1$+FF</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>62.87</td>
<td>18.67</td>
<td>6.93</td>
</tr>
<tr>
<td>2</td>
<td>188.19</td>
<td>187.16</td>
<td>24.42</td>
</tr>
<tr>
<td>3</td>
<td>237.39</td>
<td>184.95</td>
<td>21.44</td>
</tr>
</tbody>
</table>

VIII. CONCLUSIONS

In this article, for the first time to our knowledge, a pH model of the enzymatic liquefaction process has been developed. Reference tracking and disturbance rejection scenarios were formulated resembling the production mode of a large scale bioethanol plant and both measured and unmeasured disturbances were considered.

As classical control is widespread within industry, a PI controller with feed-forward action was first designed. A novel $L_1$ adaptive output feedback controller was then built and tuned in a systematic way in order to ensure high performance. The feed-forward term was also added to the $L_1$ controller. In the case of reference tracking, the PI controller performed well with small overshoots that increased as the process moved further from the design point. Re-tuning of the controller is necessary when switching to a different pH level in order to preserve performances. The $L_1$ controller proved to behave similarly regardless of the nominal pH level except in the neutral highly nonlinear area of $pH = 7$. However, the closed loop system remained stable.

In the case of measured disturbances, both control strategies were effective and it was shown that the feed-forward term considerably improves the system response. In the case of unmeasured disturbances, the $L_1$ adaptive controller had better performances.

A new cost function was derived from the enzymatic bell-shaped activity curve to assess closed loop performances. This cost function was further used to monitor the efficiency of the enzymatic process.

Finally, a tuning method was proposed for the $L_1$ controller that proved to be very effective for this application. This tuning procedure is an enhancement of the method presented in [14].

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REFERENCES


