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Optimal control of a two stages process for ethanol production from lignocellulosic materials of sorghum

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Abstract. The production and purification of ethanol from sorghum lignocellulosic materials by a two stages process was modeled and optimized in the present work. The process involves a first stage for hydrolyzing sweet sorghum bagasse and simultaneously fermenting the generated sugars and a second stage for the separation of ethanol from the filtered culture. Kinetic and distillation equations were embedded into macroscopic balances in order to derive a mathematical model used to solve a two-stage optimal control problem. The aim was to maximize the productivity of the process by optimally managing the controlled flows and by optimally fixing the switching time between the fermentation phase and the separation phase.

Keywords: multi-stage optimal control; sorghum bagasse; simultaneous hydrolysis and fermentation; semi-continuous distillation.

1 Introduction

The determination of optimal control profiles for chemical processes described by differential and algebraic equations (DAEs) is a well know practice on chemical engineering. Given a system of state variables influenced by externally manipulatable control variables, the problem consists on finding the optimal control profiles minimizing/maximizing a stated objective. Sometimes the optimization of chemical processes would include a multi-stage optimal control problem that involve change of dynamics and control variables on several phases along the time. Many batch and semicontinuous process may include no smooth, switched optimal control problems and multi-stage problems. This kind of problems was mainly researched for aerospace applications but far less examples from chemical engineering can be found in the literature.

On the other hand, the oil price volatility and a long-term forecasted decline in worldwide petroleum reserves motivated the research on alternative energy sources which ideally should be renewable and sustainable. Bioethanol is regarded as a cred-

itable replacement-fuel because it has a positive impact on air quality and fulfils most criteria for an inexhaustible and renewable fuel [1]. Ethanol production from grains has been a very active research-field but the production of ethanol based on crop starches compete with food and fiber production. Therefore, research has shifted towards producing ethanol from lignocellulosic materials such as agricultural/forestry residues and from dedicated energy crops. Sweet sorghum is a variety in which the juice with high concentration of fermentable sugars (sucrose, glucose and fructose) can be extracted from the fresh stems [2]. A huge amount of lignocellulosic residue is produced as byproduct of sweet sorghum [3, 4] and this lignocellulosic residue, the so-called bagasse, has nonfood applications and is a very cheap carbohydrates source. Due to tight profits and high price volatility, a strong incentive for optimizing ethanol production exists. This work is concerned with the off-line optimization of a two stages process for producing ethanol from lignocellulosic materials of sorghum bagasse. The first stage involves the hydrolysis of sweet sorghum bagasse and the simultaneous fermentation of the generated sugars on a feed stirred tank reactor. The second one concerns to the separation of ethanol from the filtered culture by a semi-batch distillation column. The aim is to provide optimal open-loop trajectories for the solution flow with grinded bagasse to the reactor, the flow of filtered culture with ethanol toward the boiler of the distillation column, and the distillate flow. In addition, the switching time between the fermentation phase and the separation phase must be fixed. The optimization of the train involves not just the determination of optimal control trajectories but also the switching time between both phases because the final state values for the reaction stage determine the initial state values for the distillation stage. Consequently, it is not possible to independently optimize both units. I.e. optimal initial conditions for the distillation stage may be very different from end value determined by the independent optimization of the reaction phase. In addition, while the whole problem may be a fixed-time one, each stage is defined by a free time subproblem.

2 Modeling the simultaneous hydrolysis and fermentation process

Several ways to generate ethanol from lignocellulosic residues of sweet sorghum are possible. All are subject to active research but we restrict this study to the enzymatic hydrolysis of grinded bagasse and the simultaneous fermentation of sugars generated as products of the hydrolysis by *Saccharomyces cerevisiae*. Kinetic equations for the simultaneous saccharification and fermentation processes are given by:

$$r_M^h = -Y_{S/M}\gamma \frac{M}{M + k_1} M \quad (1)$$

$$r_S^h = \gamma \frac{M}{M + k_1} M \quad (2)$$

$$r_X = \mu_{\max} \frac{S}{k_S + S + \frac{M^2}{k_i}} \left(1 - \frac{P}{k_p} \right) X \quad (3)$$

$$r_P = a\mu_{\max} \frac{S}{k_S + S} X \quad (4)$$

$$r_S = -\frac{1}{Y} \mu_{\max} \frac{S}{k_S + S} X \quad (5)$$

In kinetic equations (1)-(5), M is a variable representing a fictitious concentration of hydrolyzable lignocellulosic material inside solid materials, S is the glucose concentration, X is the biomass concentration and P is the ethanol concentration; γ the maximum specific hydrolysis rate; k_i is the saturation constant; $Y_{S/M}$ is the observable yield of glucose on the lignocellulosic materials; μ_{\max} is the maximum specific biomass growth rate; k_s is the Monod constant on glucose; k_p and k_i are inhibition constants that takes into account the braking effect of ethanol and lignocellulosic solids on the biomass growth rate; a is the growth-associated Luedeking-Piret specific production rate and Y is a lumped yield of products (biomass and ethanol) on glucose. It is worth noting that, in addition to glucose, several other fermentable sugars are produced by the hydrolysis of the lignocellulosic material but glucose is the main product and is the limiting substrate in the biomass growth rate [5]. Rates r_M^h , r_S^h , r_X , r_P and r_S are respectively the depletion rate of hydrolysable materials; the glucose production rate; the biomass growth rate; the ethanol production rate and the glucose consumption rate. To model a reactor, kinetic rates linked by yield parameters must be embedded into macroscopic balances equations corresponding to the reactor type [6].

3 Modeling the separation process

Ethanol from aqueous solutions, like fermentation cultures, usually is separated by standard techniques like filtering and distillation. Although other options are feasible, filtering and distillation of cultures are usually employed to purify ethanol produced on fermentative process; so, we consider just these techniques. Models for a batch distillation depend mainly on the details level considered but there are currently quite standardized mathematical representations for this separation technique. We base our model on the one by [7] which considers the following assumptions: feeding an aqueous mixture at saturation temperature to the boiler; negligible vapor holdup in each tray and in the boiler; constant vapor flow and constant liquid holdup in trays and in the condenser; theoretical trays; constant operation pressure; adiabatic (energy balanc-

es neglected) column with n stages of equilibrium; and total condensation of the distillate. The mathematical model is given by eqs. (6) to (9):

$$\frac{dx_i}{dt} = \frac{L_f(x_{i-1} - x_i) + V_f(y_{i+1} - y_i)}{M_h} \quad i = 1, \dots, n \quad (6)$$

$$\frac{dB}{dt} = L_f - V_f \quad (7)$$

$$\frac{dx_0}{dt} = \frac{V_f}{M_{hc}}(y_1 - y_0) \quad (8)$$

$$\frac{dx_b}{dt} = \frac{L_f(x_n - x_{n+1}) - V_f(y_{n+1} - x_{n+1})}{B} \quad (9)$$

In eqs. (6)-(9), x_i represents the molar fraction of the volatile component in the liquid phase of tray i ; y_i is the molar fraction of the volatile component in the vapor phase of such a tray; L_f is the liquid flow on the column, V_f is the vapor flow; B is the molar quantity of aqueous solution in the boiler, M_h is the molar holdup in each tray and M_{hc} is the molar holdup in the condenser. Eq. (6) models the dynamics of the molar fraction of the volatile component on each tray. Subscript 0 indicates the condenser while $n + 1$ refers to the boiler. Eq. (7) computes the dynamics of the moles of filtered solution on the boiler. Eq. (8) computes the molar fraction of the volatile component, i.e. ethanol, on the condenser and eq. (9) does it for the boiler. Vapor-liquid relations are usually given by nonlinear algebraic equations like the Antoine equations but here a more complex, non-ideal vapor-liquid model is considered. It is expressed directly as a table as follows:

$$y_i = y(x_i) \quad (10)$$

4 Assembling models for describing and optimizing the train

The process involves a single reactor for simultaneously hydrolyzing solids and fermenting the produced sugars that feeds a distillation column as illustrated in Figure 2. Since a high concentration of solids may inhibit the biomass growth, its quantity in the reactor is managed by controlling the flow of the solution with solids in order to operate the reactor in fed-batch mode. Afterwards, the filtering of depleted solids and

biomass is performed to feed the boiler with the aqueous ethanol-solution. To model the train, kinetic equations rates linked by yield parameters must be embedded into macroscopic balances equations of the reactor [6] and, in the same way; the dynamics of the distillator must consider that the boiler is feed with the filtered solution. The problem involves two phases; (p_1) hydrolysis + fermentation and (p_2) distillation and three control variables: (u_1) the flow of the solution with hydrolysable solids toward the reactor; (u_2) the flow of the filtered culture toward the boiler of the distillation tower; and (u_3) the distillate flow.

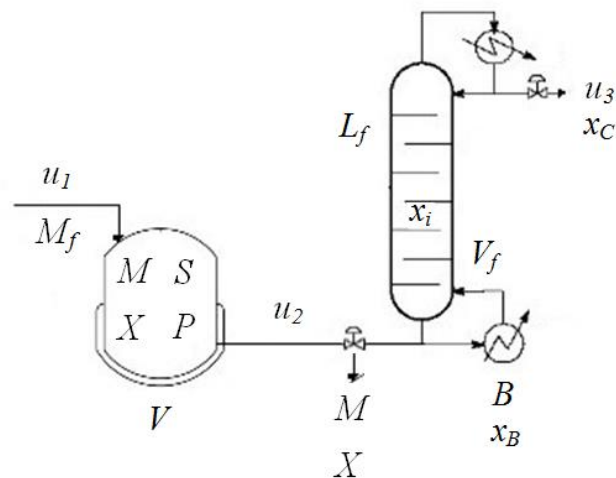


Fig. 1. Schematics of the train.

4.1. Dynamic equations

The dynamics of the whole train is defined by eqs. (11) to (20). Eqs. (11) to (14) respectively state the dynamics for the hydrolysable material, glucose, biomass and ethanol in the reactor. Eq. (15) gives the volumetric dynamics of the hydrolysis + fermentation reactor. Eq. (16) defines the dynamics of the molar fraction of ethanol in each tray. The dynamics for the quantity of solution in the boiler is given by eq. (17). The molar fractions of ethanol in the boiler and in the condenser are respectively defined by eqs. (18) and (19). Eq. (20) gives the dynamics of the distillate.

$$\frac{dM}{dt} = \left(r_M + u_1 \frac{M_f - M}{V} \right) p_1 \quad (11)$$

$$\frac{dS}{dt} = \left(r_S^h - \frac{1}{Y} r_X - u_1 \frac{S}{V} \right) p_1 \quad (12)$$

$$\frac{dX}{dt} = \left(r_X - u_1 \frac{X}{V} \right) p_1 \quad (13)$$

$$\frac{dP}{dt} = \left(r_P - u_1 \frac{X}{V} \right) p_1 \quad (14)$$

$$\frac{dV}{dt} = u_1 p_1 - u_2 p_2 \quad (15)$$

$$\frac{dx_i}{dt} = \left(\frac{L_f(x_{i-1} - x_i) + V_f(y_{i+1} - y_i)}{M_h} \right) p_2 \quad i = 1, \dots, n \quad (16)$$

$$\frac{dB}{dt} = (u'_2 + L_f - V_f) p_2 \quad (17)$$

$$\frac{dx_0}{dt} = \left(\frac{V}{M_{hc}} (y_1 - y_0) \right) p_2 \quad (18)$$

$$\frac{dx_B}{dt} = \left(\frac{u'_2(x_f - x_{n+1}) + L_f(x_n - x_{n+1}) - V_f(y_{n+1} - x_{n+1})}{B} \right) p_2 \quad (19)$$

$$\frac{dD}{dt} = u_3 p_2 \quad (20)$$

Binary indicators p_1 and p_2 take value 1 whenever their respective phases are active and 0 otherwise. The molar feeding flow u_2' is related to u_2 by algebraic equations to be next defined. Vapor-liquid data along the whole curve are computed by interpolation between the points provided by table expressed like the eq. (10).

4.2 Algebraic equations

Total sugars S_T extracted from lignocellulosic solids and the actual solids mass of can be estimated from the following expressions:

$$S_T \approx \frac{X}{Y_{X/S}} + \frac{P}{Y_{P/S}} + S \quad (21)$$

$$M_T = M_F - S_T \quad (22)$$

where M_F and M_T are respectively the total quantity of solids feed to the reactor and the actual quantity of solids in the reactor. Parameters $Y_{X/S}$ and $Y_{P/S}$ are the stoichiometric yields of biomass and ethanol on glucose or its isomers. The filtering factor c_f given by the volume of filtered solids-free solution per liter of non-filtered culture is computed by:

$$c_f = \frac{1000 - M_T - X}{1000} \quad (23)$$

This implies that the ethanol concentration on the solids-free solution is given by:

$$P_f = \frac{P}{c_f} \quad (24)$$

Since vapor-liquid equilibrium data are expressed in molar fractions, the following expressions must be computed to calculate the molar fraction of ethanol and the molar feeding flow to the distillation column:

$$x_f = \frac{\frac{P_f}{46}}{\frac{P_f}{46} + \frac{1000 - M_T - X - P_f}{18}} \quad (25)$$

$$u'_2 = 1e^{-3} \left(\frac{P_f}{46} + \frac{1000 - M_T - X - P_f}{18} \right) u_2 \quad (26)$$

It was assumed that sugars concentration in this flow is negligible because they have been almost exhausted in the fermentor.

4.3 Path, state control and end constraints

Optimal control of batch distillations usually involves the maximization of the quantity of obtained distillate (maximum distillate problem) subject to purity constraints on the distillate:

$$x_0^{\min}(t^{(2)}) \leq x_0 \leq 1 \quad (27)$$

In addition, a depletion constraint is imposed to the molar fraction on the boiler at the final distillation time:

$$x_{n+1}(t_f^{(2)}) \leq x_{n+1}^{\min} \quad (28)$$

Both constraints are applied just to the phase p_2 of the of the multi-stage optimal control problem. Also, limits to some states must be imposed to the achievable values for these variables. Here the following constraints must be considered:

$$0 \leq V \leq V^{\max} \quad (29)$$

$$0 \leq B \leq B^{\max} \quad (30)$$

Eq. (29) states that the culture volume is a nonnegative variable which must not exceed the reactor-vessel capacity while eq. (29) imposes the dome capacity as upper bound for the volume of filtered culture in the boiler. Flows to the fermentor, from the fermentor to the distillation column and the distillate flow are constrained as follows:

$$0 \leq u_1 \leq u_1^{\max} \quad (31)$$

$$0 \leq u_2 \leq u_2^{\max} \quad (32)$$

$$0 \leq u_3 \leq V_f - L_f \quad (33)$$

4.4 Objective function

When considering what it is desirable to optimize a train there are three obvious contenders; minimum duration to reach a stated target; maximum product content; and maximum productivity. We consider in this work a fixed time problem with the following objectives for each phase:

1. Phase 1: Maximum ethanol productivity in the reactor:

$$\text{Maximize } J^{(1)} = \frac{P(t_f^{(1)})V(t_f^{(1)})}{t_f^{(1)}} \quad (34)$$

2. Phase 2: Maximum distillate subject to purity constraint (27) and depletion constraint (28):

$$\text{Maximize } J^{(2)} = D(t_f^{(2)}) \quad (35)$$

So, while the whole problem is a fixed time one, each stage is defined by a free-time subproblem.

5 Multi-phase optimal control of the train

Due to the increasing complexity of engineering applications, over the past two decades the subject of optimal control has transitioned from theory to practical computing. Computational optimal control has become a science in and of itself, resulting in a variety of numerical methods and corresponding software implementations. GPOPS was developed in response to a demand from the research and academic community for a MATLAB software able to solve complex multi-stage optimal control problems. Its user-interface allows specifying multi-stage optimal control problems and we employed its 5.2 freeware version that implements the Radau pseudo-spectral collocation method [8]. To solve the problem, the following functions must be coded on GPOPS: (i) The cost functional here defined by eqs. (34) and (35). (ii) The right-hand side of the differential here given by eqs. (11) to (20), algebraic equalities (21) to (26), path constraint (27) and end-constraint (28). (iii) Boundary conditions and linkage constraints; i.e., time events and state values for connection between phases. (iv) Lower and upper limits on states, eqs. (29)-(30), and control variables, eqs. (31)-(33), at the beginning of each phase, during each phase and at the end of each phase $p = 1$ (fermentation), 2 (distillation). (v) Lower and upper limits on the total time-length of each phase and linkage constraints.

6 Solving the nominal problem

The nominal fixed final-time problem is defined by parameters summarized in Table 1 and solved in a 2.0 GHz 16 GB RAM PC. Kinetic and yield parameters for the hydrolytic production of glucose and isomers from Sorghum bagasse with FibreZyme G4 and for ethanol production with *Saccharomyces cerevisiae* were derived from experimental information obtained in Laboratorio de Fermentaciones (Facultad de Bioquímica Ciencias Biológicas; U.N.L). Non-ideal vapor-liquid equilibrium data

were gently provided by Dr. José Espinoza and coworkers from INGAR (U.T.N. – Conicet) in form of table (10).

The value $x_0^{min}(t^{(2)})$ was considered constant along the second phase. A higher purity value was not considered because of the energetic inefficiency inducted by high reflux ratios necessary to reach such values. Usually the obtained distillate is subject to a subsequent purification stage in another smaller distillation column. Note that a constant $x_0^{min}(t^{(2)})$ value reduces the path constraint (27) to a state constraint.

Table 1. Parameter values for the nominal solution.

Parameter type	Parameter	
Yields	Y_{SM} (g S/g M)	6.550
	Y (g X/g S)	1.126
Kinetics	γ (h^{-1})	0.039
	k_I (g M)	0.010
	μ_{Max} (h^{-1})	0.259
	k_S (g S)	10.00
	k_P (g P)	51.37
	k_I (g M)	5.000
	a (g P/ 10^6 cells X)	0.655
Distillation	V_f (kmol/h)	0.600
	M_h (kmol)	0.30
	M_{hc} (kmol)	0.90
	n	4
State bounds	V_{Max} (l)	1000
	B_{Max}	∞
	$x_0^{min}(t^{(2)})$	0.50
Control bounds	u_1^{Max} (l/h)	250
	u_2^{Max} (l/h)	100
	u_3^{Max} (kmol/h)	0.6
Initial conditions ($p = 1$)	M (g/l)	280
	S (g/l)	0.0
	X (g/l)	0.4
	P (g/l)	0.0
	I V (l)	500
Initial conditions ($p = 2$)	x_B	0.012
	x_i ($i = 1, \dots, 4$)	(equilib. w. boiler)
	x_C	0.807
	B (kmol)	1.00
	D (kmol)	0.00
End constraint	x_{n+1}^{min}	0.002

The output generated by GPOPS 5.2 states that the optimal quantity of distillate fulfilling purity constraints is $D(t_f^{(2)}) = 2.645$ kmol. The optimal switch time between

the production stage and the distillation stage is $t_f^{(1)} 63.601 h$. Optimal trajectories for control variables are depicted in Figs. 2. Evolution of states according the computed optimal switching time and control trajectories are depicted in Figures 3. The Fig 2.a. depicts the flow of solution with lignocellulosic material toward the reactor and the culture flow toward the boiler of the distillation column while Fig 2.b depicts the optimal distillate flow. Fig 3.a shows the evolution of states in the reactor and Fig. 3.b. depicts the evolution of the reactor volume according to the optimal flows presented in Figs. 2. Figs 3.c and 3.d depict respectively the evolution of the distillate and the quantity of filtered solution in the boiler. The evolution of molar fractions of ethanol in the boiler, in trays of the column and in the condenser is illustrated in Fig. 3.e and 3.f. Note that some numerical instability in the optimal distillate flow can be observed. This is a quite common phenomena observed in optimal control of batch distillations [7] that seems not to considerably distort the optimal evolution of the quantity of distillate.

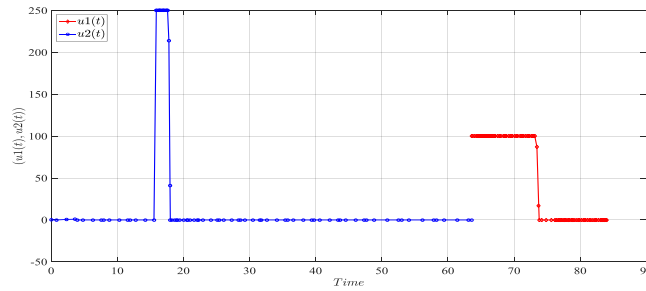


Fig. 2.a. Optimal trajectories for the flow u_1 of solution with lignocellulosic material toward the reactor and culture flow u_2 toward the boiler.

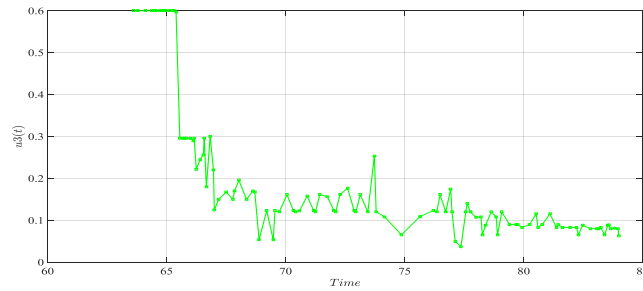


Fig. 2.b. Optimal distillate flow u_3 .

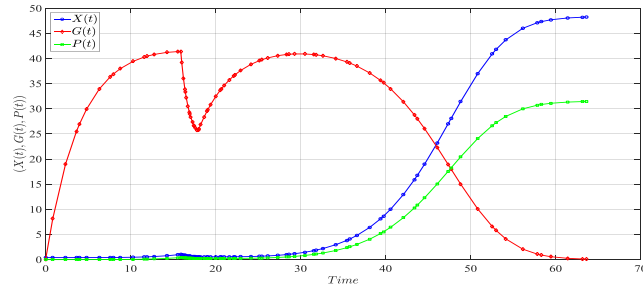


Fig. 3.a. Optimal evolution of glucose $G(t)$, biomass $X(t)$ and ethanol $P(t)$ concentrations in the reactor

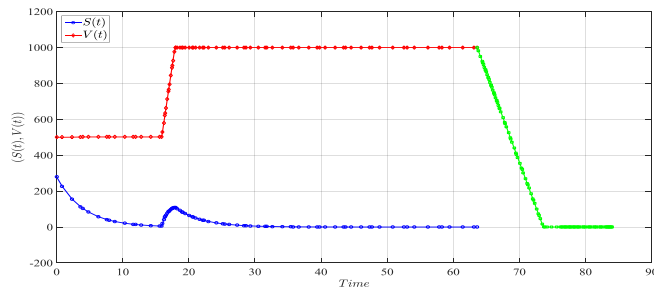


Fig. 3.b. Optimal evolution of variable $M(t)$ and dynamics of the culture volume in the reactor during *phase 1* and *phase 2*.

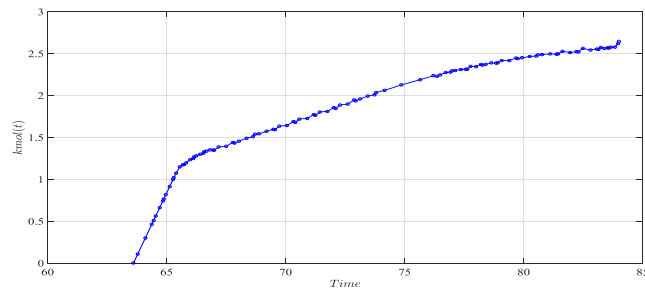


Fig. 3.c. Optimal evolution of the quantity of distillate $D(t)$.

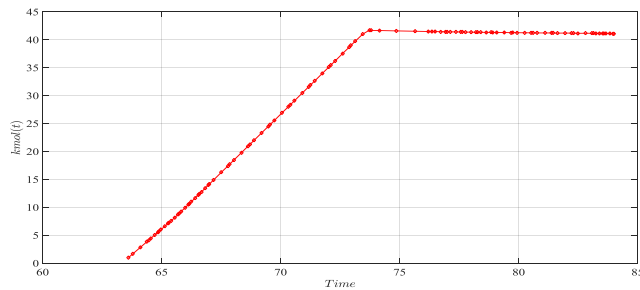


Fig. 3.d. Optimal evolution of the volume of filtered culture in the boiler.

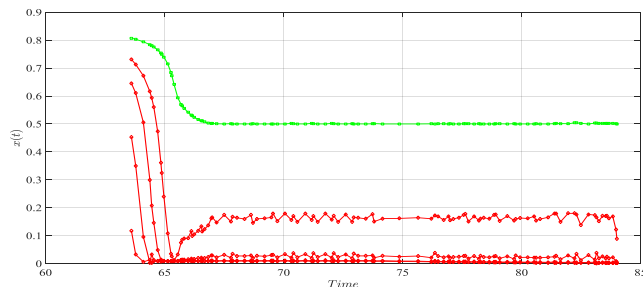


Fig. 3.e. Optimal evolution of molar fractions of ethanol in in trays and in the condenser.

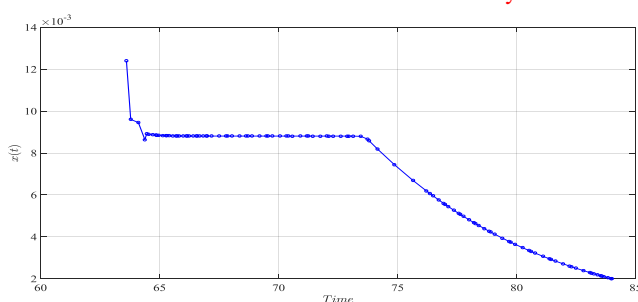


Fig. 3.f. Optimal evolution of molar fractions of ethanol in the boiler.

7 Conclusions

A two stages model for optimizing a train of a simultaneous hydrolysis and fermentation reactor and a distillation column has been developed in this work. The model involves experimental information for the hydrolytic production of glucose and isomers from Sorghum bagasse with FibreZyme G4 and the kinetics of ethanol production with *Saccharomyces cerevisiae* from the generated sugars; and bibliographic information for parameters of the distillation stage. Kinetic equations and distillation dynamics equations were introduced into macroscopic balances equations for modeling the train of both units. The assembled train involves three control variables: the flow of a solution with lignocellulosic solids toward the reactor, the flow of the filtered culture from the reactor toward the distillation column and the distilled flow out from such a column. These variables must be optimally profiled and time-scheduled. To reach that goal, the developed model, which involves $9 + n$ (number of trays) states, 3 control variables and 2 phases indicators, was solved by using GPOPS 5.2. As a consequence, the optimal flow profiles and the switch time between the fermentation and the separation stages were computed.

It is worth noting that multi-stage process involving a change of dynamics and change of control variables on several phases along the time are relatively common in (bio)chemical engineering but very few examples about the optimization of this kind of problems can be found (bio)chemical engineering literature. This work shows that

there are no big obstacles for using and optimizing more accurate multi-stage switching representations of these systems.

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