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Stability and asymptotic observers of binary distillation processes described by nonlinear convection/diffusion models

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***Abstract*—Distillation column monitoring requires shortcut nonlinear dynamic models. On the basis of a classical wave-model and time-scale reduction techniques, we derive a one-dimensional partial differential equation describing the composition dynamics where convection and diffusion terms depend non-linearly on the internal compositions and the inputs. The Cauchy problem is well posed for any positive time and we prove that it admits, for any relevant constant inputs, a unique stationary solution. We exhibit a Lyapunov function to prove the local exponential stability around the stationary solution. For a boundary measure, we propose a family of asymptotic observers and prove their local exponential convergence. Numerical simulations indicate that these convergence properties seem to be more than local.**

I. INTRODUCTION

Distillation is one of the most commonly employed industrial separation processes. Distillation columns are known to exhibit highly nonlinear behaviors, all the more that they operate at high purity. To help stabilizing the products purity or managing transient operation, several approaches have been explored to develop nonlinear control models.

The broadly investigated stage-by-stage models suffer from their inherent complexity, resulting from the high number of involved elementary separation stages. Thus they hardly cope with real-time control and robust tuning requirements. Various reduced models can be obtained from the stage-by-stage approach, such as compartmental models ([1][2][3]) or collocation models, although not without decreasing their accuracy or range of validity, notably in dynamics.

A different approach was initiated in [4]. Distillation columns are envisaged as continuous beds along which molar fraction profiles move as propagating waves, depending on the liquid and gas flows. This wave model leads to a very concise formulation of the profiles. Yet the effects of the column's boundaries on the profiles' shape are not rendered with enough accuracy for high-purity separation purposes. More recent works on the wave-model use off-line estimated shape parameters thanks to steady-state plate-models [5] or Kalman filtering [6]. [4] is originally for binary mixture separation, but has been later extended to multi-component mixtures [7].

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In this paper, we investigate a non-linear dynamic model for binary mixture separation in a packed column. Similarly to [4], we use a continuous framework to establish local balance equations. Taking advantage of two different time scales in the system, we obtain a concise model: a partial differential equation (PDE) for an internal distributed variable (model (22)), and two output maps giving the molar fraction profiles ((19) and (20)). This model also accounts for the wave propagation and deformation concepts introduced within the wave-model approach. The associated Cauchy problem is shown to have a unique solution for all positive time: the solution behaves as regular molar fraction profiles and remains between 0 and 1. Then we investigate the stationary solutions, proving their existence, uniqueness, and strict monotonicity along the column s-axis. Next, we exhibit a strict Lyapunov function with exponential decay rate to prove the local exponential stability of the stationary solution in L^2 topology. Using the same Lyapunov function, we then propose a family of tunable non-linear state observers relying on the molar fraction measures on the boundary. Some simulation results illustrate the stationary solution stability and the observers performances, before we conclude on future works.

II. REDUCED BINARY DISTILLATION MODEL

We consider the separation of a binary mixture in a single packed distillation column, whose geometry and functioning are summarized on Fig. 1 (left). The column is fed at its bottom with gas molar flow V and heavy component molar fraction y_h. The gas rises through the packed section of height h, before it is totally liquefied in the top condenser. The obtained liquid is separated in an extracted product flow $L - V$, and a reflux flow L, which finally leaves the column at the bottom. Note that this geometry corresponds to a peculiarity of cryogenic air separation processes, where stripping and rectifying operations usually take place in two separated columns.

As the molar fractions always sum to 1, it is sufficient to compute the molar fraction of one of the chemical species to characterize the system. We focus arbitrarily on the heavy component, that is, the less volatile. We denote x (resp. y) its molar fraction in the liquid (resp. gas) phase.

A. Simplified model for phase-to-phase molar exchanges

The complex geometry of interlaced liquid and gas pathes is usually simplified as follows (see, e.g., [4]): we assume

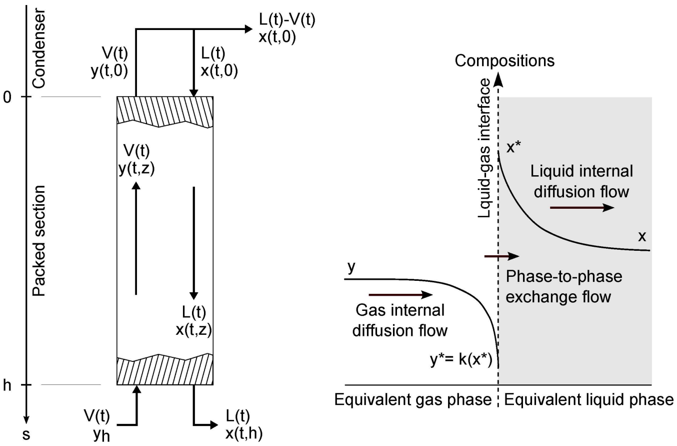

Fig. 1. *Left*: simplified scheme of a binary mixture separation column with liquid and gas flows, and heavy component molar fractions. *Right*: simplified model for the phase-to-phase exchanges in an infinitesimal column slice; inhomogeneous molar fractions in the radial dimension create internal diffusion flows coupled by the interface exchange flow.

that all the liquid (resp. gas) flows can be represented by a single equivalent liquid (resp. gas) flow. The equivalent liquid and gas phases are in contact through a single interface. In the following, subscript L (resp. V) stands for the equivalent liquid (resp. gas) phase.

Consider a slice of the distillation column of infinitesimal height such as illustrated on Fig. 1 (right). In the radial dimension, the molar fraction profiles result from the competition between vertical transport by liquid and gas flows, and radial exchanges between the liquid and gas phase. The latter predominate at the liquid-gas interface, where the two phases are permanently at the thermodynamical equilibrium. On the contrary, the farther from the interface, the less the thermodynamics couple the liquid and gas molar fractions. Consequently, far from the interface, vertical transport phenomenon predominates.

This results in non-homogeneous profiles in the radial dimension, hence in radial diffusion flows. Orientating these flows as on Fig. 1 (right), we roughly write them $\frac{\lambda_L}{\epsilon}(x^*-x)$ in the liquid phase, $\frac{\lambda_V}{\epsilon}(y-y^*)$ in the gas phase, where $\lambda_{L,V} > 0$ are equivalent to diffusion coefficients, $0 < \epsilon \ll 1$ is a small scaling coefficient, and x^*, y^* are the molar fractions at the interface.

Assuming that the two components of the mixture have the same heat of vaporization, the coupling equation at the interface reads $\lambda_V(y^* - y) + \lambda_L(x^* - x) = 0$, while the thermodynamical equilibrium imposes the non-linear relation $y^* = k(x^*)$. One often, but non-necessarily, uses

$$k(x) = \frac{\alpha x}{1 + (\alpha - 1)x}\,, \tag{1}$$

where $0 < \alpha < 1$ stands for the relative volatility of the considered heavy component with respect to the second light one. For the moment, we do not impose any particular constraint on k.

Adding two last equations arising from molar balances in the liquid and in the gas phases, we obtain the system

$$\begin{cases} \sigma_L \partial_t x = -\partial_s(Lx) + \frac{\lambda_L}{\epsilon}(x^* - x), \\ \sigma_V \partial_t y = \partial_s(Vy) + \frac{\lambda_V}{\epsilon}(y^* - y), \\ 0 = \lambda_V(y^* - y) + \lambda_L(x^* - x), \quad y^* = k(x^*), \end{cases} \tag{2}$$

where L, V are the liquid and gas molar flows, respectively, and the $\sigma's > 0$ are the liquid and gas molar hold-ups, assumed to be constant. We use the notations $\partial_t := \frac{\partial}{\partial t}$, $\partial_s := \frac{\partial}{\partial s}$.

The boundary conditions of the model express on the heavy component molar flow rather than on molar fractions. At the bottom of the column, the inlet heavy component molar flow is directly $\Phi_{inlet} = V y_h$, where $y_h \in (0, 1)$ is the fixed inlet heavy component molar fraction. At the top of the column, neglecting the condenser's dynamics, the heavy component molar flow Φ_{out} exiting the packings separates in

- a heavy component product flow $\Phi_{prod} = \frac{V-L}{V}\Phi_{out}$,
- a heavy component recycled flow $\Phi_{in} = \frac{L}{V}\Phi_{out}$ which is re-injected in the packings.

B. Model reduction based on two time-scales

The system (2) is reduced for small yet non-zero values of ϵ. We remove here the stable and fast dynamics and provide an approximation of the slow dynamics including order one term versus the small positive parameter ϵ. Such model reduction is inspired of invariant manifold techniques applied to two-time scale systems and described in [8] for finite dimensional systems (see also [9]). The derivation of this order one slow system is only formal here and requires certainly additional mathematical justifications to cope with our infinite dimension framework.

The reduction is intended to preserve the slow balance equation on the total amount of heavy component

$$(\sigma_L + \sigma_V)\partial_t Z = -\partial_s(Lx) + \partial_s(Vy), \tag{3}$$

where

$$Z = \frac{\sigma_L x + \sigma_V y}{\sigma_L + \sigma_V}. \tag{4}$$

Thus, whatever the value of ϵ, that is, whatever the phase-to-phase exchanges, the reduced model of the 2-phases system remains conservative.

Let us define $X(s, t)$ such that

$$Z = \frac{\sigma_L X + \sigma_V k(X)}{\sigma_L + \sigma_V}, \tag{5}$$

and thus with the notation $k'(X) = \frac{dk(X)}{dX}$:

$$\partial_t Z = \frac{\sigma_L + \sigma_V k'(X)}{\sigma_L + \sigma_V}\partial_t X \tag{6}$$

Assuming that ϵ is small, let us consider the asymptotic development of the unknown molar fractions:

$$x = x_0(X) + \epsilon x_1(X) + \dots,\ y = y_0(X) + \epsilon y_1(X) + \dots,$$
$$x^* = x_0^*(X) + \epsilon x_1^*(X) + \dots,\ y^* = y_0^*(X) + \epsilon y_1^*(X) + \dots,$$

with the notations $x_i := \frac{\partial^i x}{\partial \epsilon^i}(\epsilon = 0)$, $y_i := \frac{\partial^i y}{\partial \epsilon^i}(\epsilon = 0)$, $x_i^* := \frac{\partial^i x^*}{\partial \epsilon^i}(\epsilon = 0)$, $y_i^* := \frac{\partial^i y^*}{\partial \epsilon^i}(\epsilon = 0)$, $\forall i \in \mathbb{N}$.

We assume in our derivations that neither V nor L depend on s. This assumption is reasonable for the gas, yet it certainly is a simplification of the liquid hydraulics in many separation columns.

Limiting the asymptotic development to the order 0 versus ϵ, the dynamical system (2) and the definition of Z give

$$x_0^* = x_0,\ y_0^* = y_0,\ y_0^* = k(x_0^*),\ Z = \frac{\sigma_L x_0 + \sigma_V y_0}{\sigma_L + \sigma_V}.$$

Thus at the order 0 versus ϵ we have

$$x_0 \equiv X, \quad y_0 \equiv k(X), \tag{7}$$

$$\partial_t X = \frac{-L + V k'(X)}{\sigma_L + \sigma_V k'(X)} \partial_s X. \tag{8}$$

Notice that the boundary conditions on the heavy component molar flows become

$$\begin{cases} \Phi_{inlet} = V y_0(t,h) = V y_h, \\ \Phi_{prod} = \frac{V-L}{V}(V y_0(t,0)) = (V-L) y_0(t,0), \\ \Phi_{in} = L y_0(t,0). \end{cases}$$

Since $\Phi_{in} = L x_0(t,0)$, we have $x_0(t,0) = y_0(t,0) = k(x(t,0))$. Thus $x(t,0) = 0$ or 1, which gives no valuable information on the small but non-zero value of the actual heavy component molar fraction in the product.

An additional order in then required in the asymptotic development. To the order 1 versus ϵ, the system (2) rewrites

$$\sigma_L \partial_t X = -\partial_s (LX) + \lambda_L (x_1^* - x_1), \tag{9}$$

$$\sigma_V k'(X) \partial_t X = k'(X) \partial_s (VX) + \lambda_V (y_1^* - y_1), \tag{10}$$

$$0 = \lambda_L (x_1^* - x_1) + \lambda_V (y_1^* - y_1), \tag{11}$$

$$y_1^* = k'(X) x_1^*, \tag{12}$$

and the definition of Z gives

$$\sigma_L x_1 = -\sigma_V y_1. \tag{13}$$

Using (8),(9) and (10) rewrite

$$k'(X) \frac{\sigma_V L + \sigma_L V}{\sigma_L + \sigma_V k'(X)} \partial_s X = \lambda_L (x_1^* - x_1), \tag{14}$$

$$-k'(X) \frac{\sigma_V L + \sigma_L V}{\sigma_L + \sigma_V k'(X)} \partial_s X = \lambda_V (y_1^* - y_1). \tag{15}$$

According to (12) and (13), one has

$$\lambda_V (y_1^* - y_1) = -\lambda_L (x_1^* - x_1) = \lambda_V (k'(X) x_1^* + \frac{\sigma_L}{\sigma_V} x_1),$$

then multiplying (14) by $\frac{\lambda_V}{\lambda_L} k'(X)$ and subtracting (15) yields

$$x_1 = \frac{-\sigma_V \left(\frac{k'(X)^2}{\lambda_L} + \frac{k'(X)}{\lambda_V} \right)}{(\sigma_L + \sigma_V k'(X))^2} (\sigma_V L + \sigma_L V) \partial_s X, \tag{16}$$

$$y_1 = \frac{\sigma_L \left(\frac{k'(X)^2}{\lambda_L} + \frac{k'(X)}{\lambda_V} \right)}{(\sigma_L + \sigma_V k'(X))^2} (\sigma_V L + \sigma_L V) \partial_s X. \tag{17}$$

Let us define

$$G(X) = \frac{\frac{k'(X)^2}{\lambda_L} + \frac{k'(X)}{\lambda_V}}{\left(\sigma_L + \sigma_V k'(X)\right)^2} (\sigma_V L + \sigma_L V)^2.$$

Developing (3) up to order 1 versus ϵ yields

$$(\sigma_L + \sigma_V k'(X)) \partial_t X = \partial_s \left[V y_0 - L x_0 + \epsilon (V y_1 - L x_1) \right].$$

Injecting (7),(16) and (17), we obtain the following non-linear convection-diffusion partial-differential equation for X:

$$(\sigma_L + \sigma_V k'(X)) \partial_t X = \partial_s \left[-LX + V k(X) \right] + \epsilon \partial_s \left[G(X) \partial_s X \right]. \tag{18}$$

The profiles are available through output maps:

$$x(s,t) = x_0(X) + \epsilon x_1(X) = X - \tfrac{\epsilon \sigma_V G(X)}{\sigma_V L + \sigma_L V} \partial_s X, \tag{19}$$

$$y(s,t) = y_0(X) + \epsilon y_1(X) = k(X) + \tfrac{\epsilon \sigma_L G(X)}{\sigma_V L + \sigma_L V} \partial_s X. \tag{20}$$

At the top of the column, we now have

$$\Phi_{out} = V k(X(t,0)) + \epsilon G(X(t,0)) \partial_s X(t,0),\ \Phi_{in} = L X(t,0),$$

thus the top boundary condition leads to

$$V X(t,0) = V k(X(t,0)) + \epsilon G(X(t,0)) \partial_s X(t,0), \tag{21}$$

and the heavy component molar fraction in the product is

$$k(X(t,0)) + \epsilon \frac{G(X(t,0))}{V} \partial_s X(t,0).$$

At the bottom of the column, the boundary condition similarly rewrites $V y_h = V k(X(t,h)) + \epsilon G(X(t,h)) \partial_s X(t,h)$.

Notice that (large) radial diffusion flows in (2) *create* small nonlinear diffusion flows along the axis of the column in (18). Therein the column dynamics is distinguished from tubular chemical reactors, where the (large) axial diffusion term is on the contrary cut by perturbation reduction techniques as in [10]. The axial diffusion can be scaled with respect to the axial convection thanks to the diffusion scaling parameter ϵ. The axial diffusion is also directly impacted by the value of $(\sigma_V L + \sigma_L V)^2$, that is, by the operating set-point of the column. Roughly speaking, this would correspond to a set-point dependant wavefront shape in Marquardt's wave-model [4]. Boundary condition (21) would then describe the pinching profile due to the repelling effect of the condenser. In the wave-model, the wavefront speed is driven by the liquid and gas traffics; this is reflected here by the convection term of (18).

C. The nonlinear partial-differential model

Let us recall that

$$f(X) := \sigma_L + \sigma_V k'(X),$$

$$G(X) := \frac{\frac{k'(X)^2}{\lambda_L} + \frac{k'(X)}{\lambda_V}}{\left(\sigma_L + \sigma_V k'(X)\right)^2} (\sigma_V L + \sigma_L V)^2.$$

We consider from now on the following nonlinear partial-differential equation dynamic model for the binary mixture separation column:

$$\left\{ \begin{array}{l} f(X)\partial_t X = \partial_s\left[-LX + Vk(X) + \epsilon G(X)\partial_s X\right], \\ Vy_h = Vk\left(X_{(t,h)}\right) + \epsilon G(X_{(t,h)})\partial_s X_{(t,h)}, \\ VX_{(t,0)} = Vk\left(X_{(t,0)}\right) + \epsilon G(X_{(t,0)})\partial_s X_{(t,0)}, \end{array} \right. \quad (22)$$

for $t \in (0, +\infty)$, with the following hypotheses:

- $0 < y_h < 1$, $\epsilon > 0$, $h > 0$,
- $k : [0,1] \to [0,1]$ is a C^1, strictly increasing function on $[0,1]$ such that $k(0) = 0$, $0 < k'(0) < 1$, $k(1) = 1$, $1 < k'(1)$. By convention, the function k is extended to the whole line by the following definition

$$k(x) := \left\{ \begin{array}{l} k'(0)x \text{ if } x < 0, \\ 1 + k'(1)(x-1) \text{ if } x > 1. \end{array} \right. \quad (23)$$

- $L, V \in \mathbb{R}_+^*$ may be time varying, and satisfy

$$k'(0)V < L, \quad 0 < L < V, \quad y_h V < L. \quad (24)$$

III. Maximum principle for strong solutions

The existence and uniqueness of strong solutions to the Cauchy problem associated to (22) is a classical result (see [11]): if the initial condition is C^2 versus s, then for $t > 0$, the solution remains C^2 versus s. With the hypotheses of II-C, such solutions X remain inside $(0,1)$.

Proposition 1: Let k, L, V be as in the previous section. Let $X_0 \in C^2([0,h], \mathbb{R})$ be such that $0 \le X_0(s) < 1, \forall s \in [0,h]$ and X be the solution of the system (18) associated to the initial data $X(0,s) = X_0(s), \quad s \in (0,h)$. Then, $\forall (t,s) \in [0,+\infty) \times [0,h], 0 \le X(t,s) < 1$.

Proof: The result is obtained by contradiction, applying the maximum principle to the function $Y_\lambda(t,s) := X(t,s)e^{-\lambda t}$, $\lambda > 0$, and limit arguments for $\lambda \to 0$. ∎

IV. Stationary solution

Consider $\mathbb{R} \ni \phi \mapsto \frac{LX_C(h)+C}{V} \in \mathbb{R}$ where X_C is the unique $C^1(\mathbb{R},\mathbb{R})$ solution of the Cauchy problem

$$-LX_C + Vk(X_C) + \epsilon G(X_C)\tfrac{dX_C}{ds} = C, \; X_C(0) = \tfrac{C}{V-L}.$$

Proposition 2: For every $\epsilon > 0$, with the hypotheses of II-C and L, V constant, there exists a unique stationary solution $\overline{X}_\epsilon \in C^\infty([0,h],\mathbb{R})$ to the system (22). Moreover, $[0,h] \ni s \mapsto \overline{X}_\epsilon(s)$ is strictly increasing and $0 < \overline{X}_\epsilon(0) < \overline{X}_\epsilon(h) < 1$.

Proof: Let us rewrite system (22) in steady-state:

$$\left\{ \begin{array}{l} -L\overline{X}_\epsilon + Vk(\overline{X}_\epsilon) + \epsilon G(\overline{X}_\epsilon)\frac{d\overline{X}_\epsilon}{ds} = C_\epsilon, \\ Vy_h = Vk\left(\overline{X}_\epsilon(h)\right) + \epsilon G(\overline{X}_\epsilon(h))\frac{d\overline{X}_\epsilon}{ds}(h), \\ V\overline{X}_\epsilon(0) = Vk\left(\overline{X}_\epsilon(0)\right) + \epsilon G(\overline{X}_\epsilon(0))\frac{d\overline{X}_\epsilon}{ds}(0). \end{array} \right. \quad (25)$$

We will prove that for every $\epsilon > 0$, there exists a unique $C_\epsilon \in \mathbb{R}$ such that the system (25) has a solution $\overline{X}_\epsilon \in C^\infty([0,h],\mathbb{R})$. Additionally, for every $\epsilon > 0$, we will have

- $0 < C_\epsilon < y_h(V-L)$,
- $0 < \overline{X}_\epsilon(s) < \overline{X}_\epsilon(h) = \frac{Vy_h - C_\epsilon}{L}, \forall s \in [0,h)$
- $\frac{d\overline{X}_\epsilon}{ds}(s) > 0, \forall s \in [0,h]$.

Notice also that if C_ϵ satisfies (25), then $\overline{X}_\epsilon(0) = \frac{C_\epsilon}{V-L}$ and $y_h = \frac{L\overline{X}_\epsilon(h)+C_\epsilon}{V}$.

First step: Let us define an auxiliary function ϕ. For every $C \in \mathbb{R}$, there exists a unique solution $X_C \in C^1(\mathbb{R},\mathbb{R})$ of the Cauchy problem

$$-LX_C + Vk(X_C) + \epsilon G(X_C)\frac{dX_C}{ds} = C, \; X_C(0) = \frac{C}{V-L}..$$

Indeed, the ordinary equation may be written $\frac{dX_C}{ds} = F(X_C) + \frac{C}{\epsilon G(X_C)}$, where $F(X) := \frac{LX - Vk(X)}{\epsilon G(X_C)}$. Thus the right hand side is a Lipschitz function of X_C with an affine growth when $|X_C| \to +\infty$. The function $\mathbb{R} \ni C \mapsto \phi(C) = \frac{LX_C(h)+C}{V} \in \mathbb{R}$ is C^1 and satisfies $\phi(0) = 0$ because $X_0 = 0$ (uniqueness in Cauchy-Lipschitz theorem).

Second step: Let us prove that ϕ is increasing. For every $C \in \mathbb{R}$, $\phi'(C) = \frac{LY_C(h)+1}{V}$ where $\frac{dY_C}{ds} = \alpha_C(s)Y_C + \frac{1}{\epsilon G(X_C)}$, $Y_C(0) = \frac{1}{V-L}$ and $\alpha_C := F'(X_C) - \frac{CG'(X_C)}{\epsilon G(X_C)^2}$. Thus $Y_C(h) = \left[\frac{1}{V-L} + \int_0^h \frac{e^{-\int_0^s \alpha_C(\theta)d\theta}}{\epsilon G(X_C)} ds\right] e^{\int_0^h \alpha_C(\theta)d\theta}$, $Y_C(h) > 0$ and then $\phi'(C) > 0$.

Third step: Let us prove that $\phi[y_h(V-L)] > y_h$ i.e. $\frac{LX_{C_*}(h)+C_*}{V} > y_h$ where $C_* := y_h(V-L)$. For this particular value of C_*, notice that $\frac{LX_{C_*}(h)+C_*}{V} > y_h \Leftrightarrow X_{C_*}(h) > y_h$. We have

$$\begin{array}{rl} \epsilon G(X_{C_*})\frac{dX_{C_*}}{ds} & = C_* + LX_{C_*} - Vk(X_{C_*}) \\ & = [y_h - k(y_h)]V > 0 \text{ at } s = 0 \end{array}$$

thus X_{C_*} is increasing in a neighborhood of 0^+. Let us assume the existence of $s^* \in [0,h]$ such that $\frac{dX_{C_*}}{ds}(s^*) = 0$. Then

$$[L - Vk'(X_{C_*})]\frac{dX_{C_*}}{ds} = \epsilon G'(X_{C_*})\left(\frac{dX_{C_*}}{ds}\right)^2 + \epsilon G(X_{C_*})\frac{d^2X_{C_*}}{ds^2}$$

with $\frac{dX_{C_*}}{ds}(s^*) = 0$. Thus (uniqueness in Cauchy-Lipschitz theorem) $\frac{dX_{C_*}}{ds} \equiv 0$, which contradicts the increasing behavior of X_{C_*} in a neighborhood of 0^+. Therefore, $\frac{dX_{C_*}}{ds}(s^*) > 0, \forall s \in [0,h]$ and $X_{C_*}(h) > X_{C_*}(0) = y_h$.

Fourth step: Let us prove the existence and uniqueness of $\overline{X}_\epsilon$. The function $\phi : [0, y_h(V-L)] \to \mathbb{R}$ is continuous, increasing and satisfies $\phi(0) = 0$ and $\phi[y_h(V-L)] > y_h$, thus (intermediate values theorem) there exists a unique $C_\epsilon \in \mathbb{R}$ such that $\phi(C_\epsilon) = y_h$. Moreover, $C_\epsilon \in (0, y_h(V-L))$. Then $\overline{X}_\epsilon := X_{C_\epsilon}$ gives the answer.

In a second part, let us prove that $\overline{X}_\epsilon$ is increasing on $[0,h]$. Let us assume the existence of $s^* \in [0,h]$ such that $\frac{d\overline{X}_\epsilon}{ds}(s^*) = 0$. Working as in the previous third step, one deduces that $\overline{X}_\epsilon$ is constant. But this is impossible because $\overline{X}_\epsilon(0) = \overline{X}_\epsilon(1) \Rightarrow C_\epsilon = y_h(V-L)$. We have proved $\frac{d\overline{X}_\epsilon}{ds}(s) \neq 0, \forall s \in [0,h]$. Necessarily, $\frac{d\overline{X}_\epsilon}{ds}(s) > 0, \forall s \in [0,h]$ and $\forall s \in (0,h)$,

$$0 < \frac{C_\epsilon}{V-L} = \overline{X}_\epsilon(0) < \overline{X}_\epsilon(s) < \overline{X}_\epsilon(h) = \frac{Vy_h - C_\epsilon}{L}$$

and consequently $0 < C_\epsilon < y_h(V-L) \quad \forall s \in (0,h)$. ∎

V. Local asymptotic stability

In this section we prove that the unique stationary solution $\overline{X}$ of (22) is locally exponentially stable by exhibiting a strict Lyapunov function for the tangent linearized system with at least exponential decay rate.

Let us define $X := \overline{X} + \delta x$. To the order 2 versus δx, (22) rewrites for $t \in (0, +\infty)$

$$\left\{ \begin{array}{l} f(\overline{X})\partial_t \delta x = \partial_s \left[\left(-L + g(\overline{X}) \right) \delta x + \epsilon G(\overline{X}) \partial_s \delta x \right], \\ g(\overline{X}(h))\delta x_{(t,h)} + \epsilon G(\overline{X}(h))\partial_s \delta x_{(t,h)} = 0, \\ V \delta x_{(t,0)} = g(\overline{X}(0))\delta x_{(t,0)} + \epsilon G(\overline{X}(0))\partial_s \delta x_{(t,0)}, \end{array} \right. \tag{26}$$

where $g(\overline{X}) := Vk'(\overline{X}) + \epsilon G'(\overline{X})\overline{X}'$, with the notations $\overline{X}' = \frac{d\overline{X}}{ds}$, $G' = \frac{dG}{dX}$. Introducing the new variable $\xi = \frac{\delta x}{\overline{X}'}$, (26) rewrites:

$$\left\{ \begin{array}{l} f(\overline{X})\overline{X}' \partial_t \xi = \partial_s \left[\epsilon G(\overline{X})\overline{X}' \partial_s \xi \right], \\ L\xi_{(t,h)} + \epsilon G(\overline{X}(h))\partial_s \xi_{(t,h)} = 0, \\ V\xi_{(t,0)} = L\xi_{(t,0)} + \epsilon G(\overline{X}(0))\partial_s \xi_{(t,0)}. \end{array} \right. \tag{27}$$

Proposition 3: the function

$$\mathcal{V}(\xi) := \int_0^h f(\overline{X}(s))\overline{X}'(s)\xi(s)^2 ds \tag{28}$$

is a strict Lyapunov function for (27): exists $\lambda_\epsilon > 0$ such that $d\mathcal{V}/dt \leq -\lambda_\epsilon \mathcal{V}$.

Proof: Let $f^* := \max_{s\in[0,h]} f(\overline{X}(s))\overline{X}'(s) > 0$, $\mu := \min_{s\in[0,h]} G(\overline{X}(s))\overline{X}'(s) > 0$. We have

$$\frac{1}{2}\frac{d\mathcal{V}}{dt} = \int_0^h \xi(t,s)\partial_s \left[\epsilon G(\overline{X})\overline{X}' \partial_s \xi(t,s) \right] ds.$$

Integrating by part yields

$$\frac{1}{2}\frac{d\mathcal{V}}{dt} = -L\overline{X}'(h)\xi(t,h)^2 - (V - L)\,\overline{X}'(0)\xi(t,0)^2 - \epsilon \int_0^h G(\overline{X})\overline{X}'(\partial_s \xi(t,s))^2 ds. \tag{29}$$

Thanks to the formula $\xi(t,s) = \xi(t,h) + \int_h^s \partial_s \xi(t,\theta)d\theta$, one finds a constant $P > 0$ such that

$$\int_0^h \xi(t,s)^2 ds \leq P \left(\xi(t,h)^2 + \int_0^h \left(\partial_s \xi(t,s)\right)^2 ds \right). \tag{30}$$

This inequality is proved by contradiction, similarly to the classical Poincaré inequality. Injecting (30) in (29) and taking $m := \min\left(L\overline{X}'(h)\,;\, \epsilon\mu\right) > 0$, one obtains

$$\frac{1}{2}\frac{d\mathcal{V}}{dt} \leq -m \left(\xi(t,h)^2 + \int_0^h \left(\partial_s \xi(t,s)\right)^2 ds \right) \leq -\frac{m}{Pf^*}\mathcal{V}.$$

■

VI. Asymptotic observers for time-varying profiles

The molar fraction is supposed to be measured at the top of the column. Let $y_{\mathrm{M}}(t) := k(X(t,0)) + \epsilon \frac{G(X(t,0))}{V}\partial_s X(t,0)$ be the measure. Let $L = \overline{L} + l(t)$, $V = \overline{V} + v(t)$, such that L and V still satisfy (24). Let $\overline{X}$ the stationary solution of (22) for $L = \overline{L}$, $V = \overline{V}$.

We consider the following 1-parameter family of observers:

$$\left\{ \begin{array}{l} f(\widehat{X})\partial_t \widehat{X} = \partial_s \left[-L\widehat{X} + Vk(\widehat{X}) + \epsilon G(\widehat{X})\partial_s \widehat{X} \right], \\ Vy_h = Vk\left(\widehat{X}_{(t,h)}\right) + \epsilon G(\widehat{X}_{(t,h)})\partial_s \widehat{X}_{(t,h)}, \\ \widehat{X}_{(t,0)} = (1-a)y_{\mathrm{M}}(t) \\ \qquad\qquad +a\left[k(\widehat{X}_{(t,0)}) + \frac{\epsilon G(\widehat{X}_{(t,0)})}{V}\partial_s \widehat{X}_{(t,0)} \right], \end{array} \right. \tag{31}$$

where $a \in \mathbb{R}$.

Let $X = \overline{X} + \delta x$, and $\widehat{X} = X + \widetilde{x}$. In the vicinity of $\overline{X}$, linearizing the PDE of the system (22) yields

$$f(\overline{X})\partial_t \delta x = \partial_s \left[\left(-\overline{L} + g(\overline{X}) \right) \delta x + \epsilon \overline{G} \partial_s \delta x \right] + \partial_s \left[-l(t)\overline{X} + \epsilon l(t)\overline{X}' \frac{\partial G}{\partial L}_{(\overline{X},\overline{L},\overline{V})} \right] + \partial_s \left[v(t)k'(\overline{X}) + \epsilon v(t)\overline{X}' \frac{\partial G}{\partial V}_{(\overline{X},\overline{L},\overline{V})} \right],$$

where $\overline{G} = G(\overline{X})$ with $L = \overline{L}$, $V = \overline{V}$.

Replacing δx by $\widetilde{x} + \delta x$ in this equation gives the linearized PDE of the observer (31), in the vicinity of $\overline{X}$ too. Thus the dynamics of the observation error $\widetilde{x}$ are given by

$$f(\overline{X})\partial_t \widetilde{x} = \partial_s \left[\left(-\overline{L} + g(\overline{X}) \right) \widetilde{x} + \epsilon \overline{G} \partial_s \widetilde{x} \right].$$

Since at $s = 0$

$$V\widehat{X} - Vy_M(t) = a\left(-Vy_{\mathrm{M}} + Vk(\widehat{X}) + \epsilon G(\widehat{X})\partial_s \widehat{X} \right) = aV(k(\widehat{X}) - k(X)) + a\epsilon\left(G(\widehat{X})\partial_s \widehat{X} - G(X)\partial_s X \right),$$

we obtain

$$\overline{V}\widetilde{x}_{(t,0)} = a\left(\overline{V}k'(\overline{X}(0))\widetilde{x}_{(t,0)} \right) + a\epsilon\left(\overline{G}(0)\partial_s \widetilde{x}_{(t,0)} + \frac{\partial G}{\partial X}_{(\overline{X},\overline{L},\overline{V})} \widetilde{x}_{(t,0)}\overline{X}'(0) \right).$$

Thus the boundary conditions for the observation error are

$$\begin{array}{l} \overline{V}\widetilde{x}_{(t,0)} = a\left(g(\overline{X}(0))\widetilde{x}_{(t,0)} + \epsilon \overline{G}(0)\partial_s \widetilde{x}_{(t,0)} \right), \\ g(\overline{X}(h))\widetilde{x}_{(t,h)} + \epsilon \overline{G}(h)\partial_s \widetilde{x}_{(t,h)} = 0. \end{array}$$

Replacing $\widetilde{x}$ by $\xi\overline{X}'$ yields

$$\begin{array}{l} f(\overline{X})\overline{X}'\partial_t \xi = \partial_s \left[\epsilon \overline{G}\overline{X}' \partial_s \xi \right], \\ \overline{L}\overline{X}'(h)\xi_{(t,h)} + \epsilon G(\overline{X}(h))\overline{X}'(h)\partial_s \xi_{(t,h)} = 0, \\ \overline{V}\overline{X}'(0)\xi_{(t,0)} = a\left(\overline{L}\overline{X}'(0)\xi_{(t,0)} + \epsilon \overline{G}(0)\overline{X}'(0)\partial_s \xi_{(t,0)} \right). \end{array}$$

Proposition 4: $\forall a \in [0, \overline{V}/\overline{L}]$, the function $\mathcal{V}$ defined in (28) is a strict Lyapunov function of the observation error,

Time (h)	0 to 1	1 to 5	5 to 9	9 to 11	11 to 13
L/V	0.61	0.5	0.61	0.64	0.61
$V(mol/s)$	70	70	70	70	70

TABLE I
REFLUX RATE STEPS FOR NUMERICAL SIMULATION IN VII-A

with exponential decay rate.

Proof: Integrating $\frac{d\mathcal{V}}{dt}$ by parts yields

$$\frac{1}{2}\frac{d\mathcal{V}}{dt} = -\overline{LX}'(h)\xi(t,h)^2 - \left(\frac{\overline{V}}{a} - \overline{L}\right)\overline{X}'(0)\xi(t,0)^2 - \epsilon \int_0^h G(\overline{X})\overline{X}'\left(\partial_s \xi(t,s)\right)^2 ds.$$

For any $a \in [0, \overline{V}/\overline{L}]$, $\left(\frac{\overline{V}}{a} - \overline{L}\right)\overline{X}'(0)\xi(t,0)^2 \geq 0$. Notice that this term is still well defined when $a = 0$, since $\frac{\xi(t,0)^2}{a} \to 0$ when $a \to 0$. Thus $\frac{1}{2}\frac{d\mathcal{V}}{dt} \leq -m\left(\xi(t,h)^2 + \int_0^h \left(\partial_s\xi(t,s)\right)^2\right)$, where $m := min\left(\overline{LX}'(h)\,;\,\epsilon\mu\right) > 0$. As in Proposition 3, one finds a constant $P > 0$ such that $\frac{1}{2}\frac{d\mathcal{V}}{dt} \leq -\frac{m}{Pf^*}\mathcal{V}$. ■

Consequently, $\forall a \in [0, \overline{V}/\overline{L}]$, the estimation given by observer (31) exponentially converges towards the actual time-varying profile X, provided that X is close enough to the stationary profile $\overline{X}$ associated to $(\overline{L}, \overline{V})$.

Tuning the parameter a allows making $\frac{d\mathcal{V}}{dt}$ more negative, thus fastening the convergence of the observer. It should also allow tuning the observer's robustness to measurement noise, by weighting the internal model effects in the top boundary condition of (31).

VII. TRANSIENT SIMULATIONS

In this section, we solve numerically the nonlinear model (22) and the nonlinear asymptotic observer (31), with the following parameters: $h = 8$ m, $\sigma_L = 2100$ mol.m^{-1}, $\sigma_V = 70$ mol.m^{-1}, $\lambda_L = 20$ mol.m^{-1}.s^{-1}, $\lambda_V = 10\lambda_L$, $\epsilon = 0.1428$, $y_h = 0.21$. We use the thermodynamical relation (1) with $\alpha = 0.42$. The partial differential equations are computed using an implicit finite difference method, with 1 s time-steps and 0.1 m space-steps. We have checked that smaller time and space steps do not significantly impact the numerical results.

A. Open-loop convergence of the nonlinear model

Fig. 2 illustrates the uniqueness and stability properties of the stationary solution. Reflux rate L/V undergoes several steps as summarized in Table I, with V kept constant to 70 mol/s. Simulation shows that the same L and V values lead to the same stationary solution, regardless of previous profile excursions. Incidentally, Fig. 2 also illustrates the column asymmetric dynamics, that is, transient behavior depending on whether the molar fraction increases or decreases over the flow change.

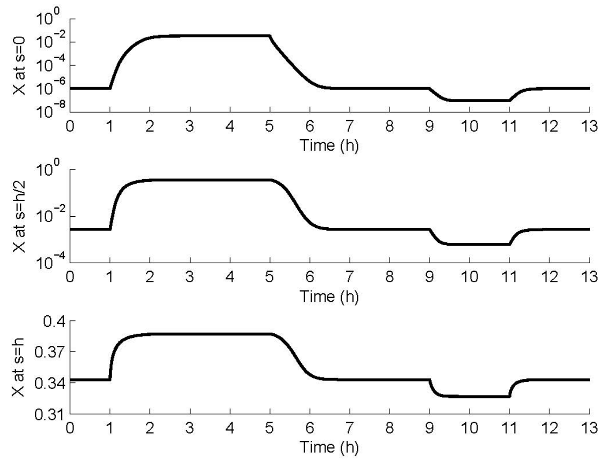

Fig. 2. Stability of the stationary solutions. *Top*: internal variable X at the top of the column (log scale). *Center*: internal variable X in the middle of the column (log scale). *Bottom*: internal variable X at the bottom of the column (linear scale). The column undergoes reflux step changes at $t = 1$ h, $t = 5$ h, $t = 9$ h and $t = 11$ h.

B. Observer convergence around a steady state

Fig. 3 illustrates the estimation of the stationary solution corresponding to $V = 70$ mol/s, $L/V = 0.61$ with various values of the tuning parameter a in nonlinear observer (31). The initial estimated solution is $\widehat{X}(0,s) = 2X(0,s)$. The bigger a, the less the top measurement is used, and the slower the convergence. Yet the estimation error vanishes for every $a \in [0, V/L]$.

The influence of a on the observation error is maximum at $s = 0$, yet rapidly decreases when s increases. Thus, when estimating molar fractions far from the top boundary, the influence of the top molar fraction estimation turns out to be negligible compared to the impact of measurement errors on L and V (illustrated on Fig. 3 (center and bottom) with 1% underestimation of L). Inversely, the value of a can be chosen freely to meet the top molar fraction estimation requirements (balancing the direct measurement and the estimated internal dynamics) without significantly impacting the rest of the estimated profile.

C. Observer convergence with a time-periodic L/V

We now consider the case of an unsteady liquid flow. L/V sinusoidally oscillates between 0.43 and 0.725 with period 1 hour and $V \equiv 70$ mol/s. The corresponding periodic regime of the top molar fraction is plotted on Fig. 4 (top) versus estimations obtained with various values of a in observer (31). The initial estimated solution is again $\widehat{X}(0,s) = 2X(0,s)$. Simulations show all the observations converge after some oscillations (yet high values of a lead to huge relative estimation error over the first cycles). Note that the variations of L cause the molar fractions to vary over a wide range (about four orders of magnitude at the top of the column) without compromising the convergence of the observers. This could indicate that the

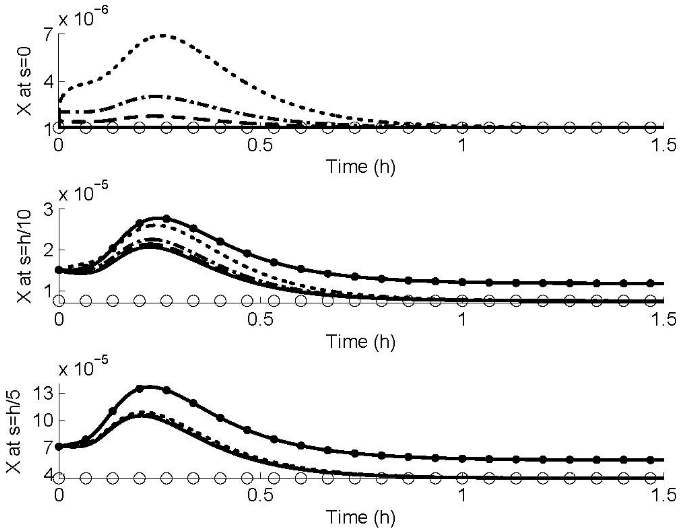

Fig. 3. Observation of a stationary solution. Several observers with various values of the tuning parameter (*solid line*: $a = 0$, *dashed*: $a = 0.5$, *dashdot*: $a = 1$, *dotted*: $a = V/L > 1$, *points*: $a = 0$ with 1% error on L) estimate the actual stationary solution (*circles*). *Top*: estimation at $s = 0$. *Center*: estimation at $s = h/10$. *Bottom*: estimation at $s = h/5$, the influence of a is almost negligible.

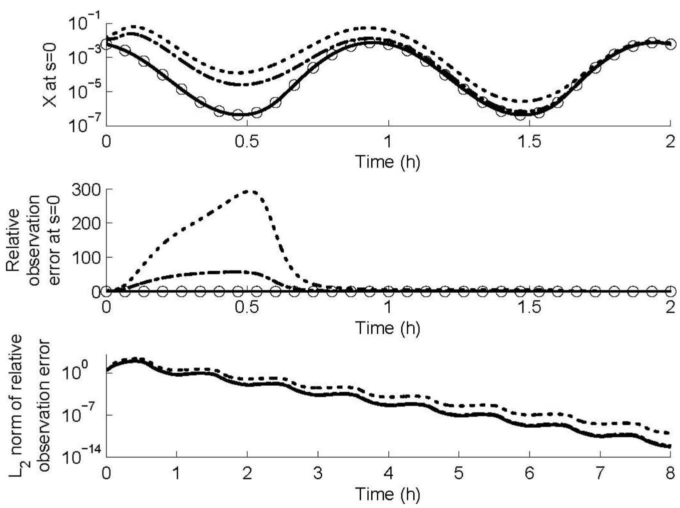

Fig. 4. Oscillating solution observation. The reflux rate L/V oscillates between 0.43 and 0.725 with period 1 h; the periodic regime (*circles*) is estimated with various values of the tuning parameter (*solid line*: $a = 0$, *dashdot*: $a = 1$, *dotted*: $a = V(t)/L(t)$). *Top*: estimation at $s = 0$ over the two first oscillations. *Center*: relative estimation error at $s = 0$ over the two first oscillations. *Bottom*: relative L_2-norm of the observation error, $\| \frac{\hat{X}-X}{X} \|_{L^2}$ over eight oscillations.

proposed family of observers has more global convergence properties than those we have proved in proposition 4.

VIII. Conclusions and Future Works

Inspired by the wave-model approach, we proposed a reduced non-linear partial differential equation dynamic model for binary distillation in a packed column. The reduction takes advantage of two separable time-scales in the system when an internal diffusion scaling coefficient is small enough. The resulting model accounts for non-linear wave propagation phenomenon, changing wave-front shape depending on internal flows, and repelling end-effects. Mathematical analysis of some of the model properties has been carried out, with only little assumptions on the thermodynamical equilibrium relation. The associated Cauchy problem admits a unique solution for all positive time. We proved the local exponential stability of the stationary solution *via* the use of a strict Lyapunov function. We proposed a tunable family of asymptotic observers to estimate time-varying profiles; using the same Lyapunov function, we guaranteed that the estimation error asymptotically vanishes provided the time-varying profile is close enough to a stationary profile. Through simulations, we showed that the proven local stability of the stationary solution and estimation error are in practice valid over wide molar fraction ranges. Future works should focus on determining whether or not these local properties can be globally extended.

References

[1] J. Lévine and P. Rouchon, "Quality control of binary distillation columns via nonlinear aggregated models," *Automatica*, vol. 27, no. 3, pp. 463–480, 1991.

[2] S. Khowinij, M. A. Henson, P. Belanger, and L. Megan, "Dynamic compartmental modeling of nitrogen purification columns," *Separation and Purification Technology*, vol. 46, no. 1-2, pp. 95–109, 2005.

[3] S. Bian, M. A. Henson, P. Belanger, and L. Megan, "Nonlinear state estimation and model predictive control of nitrogen purification columns," *Industrial and Engineering Chemistry Research*, vol. 44, no. 1, pp. 153–167, 2005.

[4] W. Marquardt, "Nonlinear model reduction for binary distillation," *Proceedings of the IFAC Control of Distillation Columns and Chemical Reactors*, pp. 123–128, 1986.

[5] G. . Zhu, M. A. Henson, and L. Megan, "Low-order dynamic modeling of cryogenic distillation columns based on nonlinear wave phenomenon," *Separation and Purification Technology*, vol. 24, no. 3, pp. 467–487, 2001.

[6] S. Bian, S. Khowinij, M. A. Henson, P. Belanger, and L. Megan, "Compartmental modeling of high purity air separation columns," *Computers and Chemical Engineering*, vol. 29, no. 10, pp. 2096–2109, 2005.

[7] A. Kienle, "Low-order dynamic models for ideal multicomponent distillation processes using nonlinear wave propagation theory," *Chemical Engineering Science*, vol. 55, no. 10, pp. 1817–1828, 2000.

[8] J. Carr, *Applications of Centre Manifold Theory*. Springer, 1981.

[9] P. Duchêne and P. Rouchon, "Kinetic scheme reduction via geometric singular perturbation techniques," *Chemical Engineering Science*, vol. 51, no. 20, pp. 4661–4672, 1996.

[10] D. Cohen and A. Poore, "Tubular chemical reactors: the "lumping approximation" and bifurcation of oscillatory states," *SIAM Journal on Applied Mathematics*, pp. 416–429, 1974.

[11] O. Ladyzhenskaia, V. Solonnikov, and N. Ural'ceva, *Linear and quasi-linear equations of parabolic type*, ser. Translations of Mathematical Monographs. American Mathematical Society, 1968, vol. 23.