Input/output transfer models of binary distillation columns derived from convection-diffusion partial differential equations

Stéphane Dudret, Fouad Ammouri and Pierre Rouchon

Abstract—Purity control in distillation columns often relies on linear models whose parameters (gains, delays, time constants) may be hard to fit satisfyingly. In this paper, we linearize a convection-diffusion model inspired from wave-models, in the case of vanishing diffusion. We obtain a linear transfer model which is explicitly parameterized with the usual column’s operating parameters. This model consists in gains and pure delays only. It is derived from an unusual asymptotic expansion (Laurent series with exponentially precise terms) versus the diffusion coefficient. We validate such linear transfer models in simulation and also on real plants data for cryogenic air separation units. Similar transfer models can be constructed for other cases of binary distillation.

I. INTRODUCTION

Distillation is amongst the most common processes in chemical industry. At high purity, the large marginal energy cost associated to a small decrease of the products’ impurity content motivates operating the distillation plant on the edge of the customer requirements. Efficient distillation plant control models are then necessary to optimize the process’ energy efficiency, in steady-state or during set-point changes. This is especially true regarding cryogenic air separation. First because the electricity costs are all the more prominent that the raw material, air, is free, second because of the huge amount of energy involved (cryogenic air separation is responsible of approximately 0.1% of the worldwide electricity consumption).

Classical stage-by-stage models are powerful tools to estimate the compositions dynamics inside distillation columns. Yet using them as straightforward control model requires substantial computational resources and may be problematic for real-time applications without use of specific techniques [5]. Thus many real-life distillation plants controllers limit to linear models, typically black-box transfer models limiting static gains, first-order filters and pure delays. Identifying such models turns out to be problematic. Invariant parameters models either limit to the vicinity of an operating point, or embody an average behavior which does not match the highly non-linear distillation dynamics. As for varying parameter models, they require time-consuming test campaigns which can disturb the production.

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We address here the development of new linear transfer model for the composition dynamics inside a packed cryogenic air separation column, allowing parameters scheduling depending on the operating point. This linear model has some relatedness with Marquardt’s wave-models [7] (see also [4], [6], [10]). Wave-models show that the compositions dynamics essentially results from traveling composition waves, triggered by reflux rate variations. They are yet not suited for high purity separation [1], where diffusive end-effects compete with convection. In [3] we proposed an enrichment of the wave-model for the high-purity context, in the form of a non-linear partial-differential equations (PDE) convection-diffusion model, with satisfying results compared to real industrial plant data. We briefly recall its structure.

Then we investigate a simplified linear version of this model in the case of vanishing diffusion. We show how the dynamics of the compositions at both ends of a packed column segment can be approximated with transfer function, in the form of an asymptotic expansion versus the diffusion coefficient. Due to the presence of exponentially precise terms, this asymptotic expansion is not trivial. It takes the form of a Laurent series multiplied by exponentially precise terms.

At the lowest order of expansion, our transfer model reduces to static gains and pure delays only. We express these gains and delays as explicit functions of the column’s internal flows and thermodynamic conditions at the operating point. This allows automated and confident parameters’ scheduling depending on the column’s operating point. We distinguish two cases, depending on the composition convection direction. Tables I and II provide the transfer matrix elements between the four inputs (flow-rates and compositions of the inflows) and the two outputs (compositions of the outflows) of the homogeneous packed segment depicted on Figure 1.

We then compare the developed linear transfer model to the non-linear convection-diffusion model on a toy-model column to illustrate how successive waves reproduce the composition dynamics. Then using real air separation plant data, we show that the simple transfer model can be satisfyingly tuned to reproduce the measured O2 composition in the lean liquid in response to multiple inputs variations.

II. NON-LINEAR DYNAMIC MODEL FOR BINARY DISTILLATION

Let us consider an homogeneous packed segment of an air separation column, such as depicted on Fig. 1 (left). As proposed in [2] and [3], the compositions dynamics along the
column s-axis can be modeled by the following convection-diffusion equation:

\[
(\sigma_L + \sigma_V k'(X)) \frac{\partial X}{\partial t} = \frac{\partial}{\partial s} \left( V k(X) - L X + \epsilon G(X) \frac{\partial X}{\partial s} \right),
\]

(1)
together with the static outputs:

\[
x(s, t) = X(s, t) - \epsilon \sigma_V \frac{G(X(s, t))}{\sigma_V L + \sigma_V V} \frac{\partial X}{\partial s}(s, t),
\]

(2)
\[
y(s, t) = k(X(s, t)) + \epsilon \sigma_V \frac{G(X(s, t))}{\sigma_V L + \sigma_V V} \frac{\partial X}{\partial s}(s, t),
\]

(3)
where \(L, V\) are the countercurrent liquid and gas molar flows, \(\sigma_L\) and \(\sigma_V\) the liquid and gas molar hold-ups, and \(t\) the time of the system. Considering air as a binary mixture \(\{N_2, O_2\}\), the actual liquid and gas \(O_2\) molar fractions \((x\) and \(y\), respectively) are obtained as static output from an internal dynamic variable \(X\), which can be seen as a lumped molar fraction \((X = \frac{\sigma_L x + \sigma_V v}{\sigma_L + \sigma_V})\). Application \(k(\cdot)\) corresponds to the thermodynamic equilibrium relation between the liquid and gas composition, and \(k'\) stands for its derivative. In the following, the non-linear model uses:

\[
k(X) = \frac{\alpha X}{1 + (\alpha - 1)X},
\]

(4)

with \(0 < \alpha < 1\) the relative volatility of \(O_2\) with respect to \(N_2\). The term \(G(X)\) actually depends on the internal flows:

\[
G(X) = \frac{k'(X)^2}{(\sigma_L + \sigma_V k'(X))^2} (\sigma_L V + \sigma_V L) ^ 2,
\]

(5)

where \(\lambda_L, \lambda_V\) are constant coefficients for the mass transfers in the liquid and gas phase, respectively. Coefficient \(\epsilon > 0\) tunes the diffusive effects and is related to the packing efficiency in the sense of [8]: the smaller \(\epsilon\), the higher the efficiency. For industrial-scale packed columns notably, one thus expects \(\epsilon\) to be vanishing. This motivates the expansions in \(\epsilon\) in the following sections.

III. LINEAR TANGENT MODEL

From the non-linear PDE model, we will derive a linear tangent model in the vicinity of an operating point. We will note with an overline the variables evaluated at the steady-state (for instance, \(\overline{X}\)). As illustrated on Fig. 1, the goal of this model is to connect the small variations of the packed segment outlet compositions \((\delta X_h, \delta X_0)\) to the small variations of the inlet compositions \((\delta x_0, \delta y_h)\) and of the outlet flows \((\delta L_{\text{in}}, \delta V_{\text{in}})\).

We simplify the PDE model by considering that \(k(\cdot)\) is linear in the vicinity of the steady-state profile. We take \(k(X) = \pi X\). This linearization is reasonable for short enough packed segments, along which the composition span is limited. We also replace the \(X'\)-- \(L'\)-- \(V'\)--dependent term \(G\) with a constant \(\overline{G}\), and consider that the hold-ups are constant. Neglecting the \(s\)-derivatives of \(L\) and \(V\), (1) rewrites:

\[
\frac{\partial X}{\partial t} = -v(s, \tau) \frac{\partial X}{\partial s} + \epsilon \overline{G} \frac{\partial^2 X}{\partial s^2},
\]

(6)

where

\[
v(s, \tau) = L(s, \tau) - \pi V(\tau) = L_0(\tau - \frac{\alpha}{\mu_L}) - \pi V(\tau),
\]

(7)
\[
\tau = \frac{t}{\sigma_L + \sigma_V} = \frac{t}{\sigma'},
\]

(8)
and \(\mu_L = V_L (\overline{\sigma_L} + \overline{\sigma_V})\) is the scaled liquid speed (we assume that the gas flow information propagates instantly along the column) and \(L_0\) is the liquid flow at the top of the segment. With \(x_0(t), y_h(t)\) the inlet compositions respectively at the top and the bottom of the packed segment, the boundary conditions for (6) read:

\[
\begin{align*}
L(0, \tau) x_0(\tau) &= (v(0, \tau) + \pi V(0, \tau)) x_0(\tau) - \epsilon \overline{G} \frac{\partial X}{\partial s}(0, \tau), \\
V(h, \tau) y_h(\tau) &= (v(h, \tau) - L(0, \tau)) x_h(\tau) - \epsilon \overline{G} \frac{\partial X}{\partial s}(h, \tau).
\end{align*}
\]

Switching to the Laplace domain, with \(p \sim d/dt\) the Laplace variable, the complete system rewrites, for small variations around the operating point:

\[
\begin{cases}
p \delta X(s, p) = -\overline{V} \frac{\partial X}{\partial X} (s, p) - \overline{X} \delta v(s, p) + \epsilon \overline{G} \frac{\partial^2 X}{\partial s^2}(s, p), \\
\overline{L} \delta x_0(p) = (\delta v_0(p) + \epsilon \overline{G} \delta v(p)) \overline{X}_0 + (\pi V + \overline{V}) \delta x_0(p) - \epsilon \overline{G} \delta X(s, 0), \\
-\overline{V} \delta y_h(p) = (\delta v_h(p) - \delta L(h, p)) \overline{X}_h + (\pi V + \overline{V}) \delta y_h(p) - \epsilon \overline{G} \frac{\partial X}{\partial X}(h, p),
\end{cases}
\]

(9)

where \(\overline{X}' = \partial s \overline{X}\) and

\[
\begin{align*}
\delta L(s, p) &= \delta L_0(p) e^{-\frac{\nu}{\nu'},} \\
\delta v(s, p) &= \delta L_0(p) e^{-\frac{\nu}{\nu'}} - \pi \delta v(p).
\end{align*}
\]

From now on, two cases must be distinguished, depending on the sign of the composition wave speed \(\nu = \overline{L} - \pi \overline{V}\).

A. Case of a downwards composition wave \((\pi > 0)\)

Following the constants’ variation method, we make the following change of variables:

\[
\begin{align*}
\delta X(s, p) &= \lambda_1(s, p) e^{r_1(p)s} + \lambda_2(s, p) e^{r_2(p)s}, \\
\partial_s \delta X(s, p) &= \lambda_1(s, p) r_1(p) e^{r_1(p)s} + \lambda_2(s, p) r_2(p) e^{r_2(p)s},
\end{align*}
\]
where $r_1$, $r_2$ are the solutions of the characteristic equation

$$\varepsilon G r^2 - \varepsilon r - p = 0.$$ \hfill (10)

This yields:

$$\partial_s \lambda_1 e^{r_1 s} + \partial_s \lambda_2 e^{r_2 s} = 0,$$

$$\varepsilon G (\partial_s \lambda_1 r_1 e^{r_1 s} + \partial_s \lambda_2 r_2 e^{r_2 s}) = X' \delta v.

Let

$$A = \frac{X_h - X_0}{e^{\frac{\beta h}{\varepsilon G}} - 1}, \quad B = \frac{X_h - X_0}{e^{\frac{\beta h}{\varepsilon G}} - 1},$$

such that $X = Ae^b + B$. We have:

$$\partial_s \lambda_1 = \frac{A\pi \delta v(s, \sigma)}{e^G (r_1 - r_2)} e^{i(\frac{\pi}{\varepsilon G} - r_1)}$$

$$= \frac{A\pi \delta v(s, \sigma)}{e^G (r_1 - r_2)} \left( e^{i(\frac{\pi}{\varepsilon G} - r_1)} - e^{i(\frac{\pi}{\varepsilon G} - r_2)} \right),$$

$$= -\frac{A\pi \delta v(s, \sigma)}{e^G (r_1 - r_2)} e^{i(\frac{\pi}{\varepsilon G} - r_2)} s,$$

and

$$= -\frac{A\pi \delta v(s, \sigma)}{e^G (r_1 - r_2)} e^{i(\frac{\pi}{\varepsilon G} - r_2)} s,$$

thus:

$$\lambda_1(s, p) = C_1(p)$$

$$A\pi \left( e^{i(\frac{\pi}{\varepsilon G} - r_1)} - e^{i(\frac{\pi}{\varepsilon G} - r_2)} \right) \delta L_0 - \frac{A\pi \delta v(s, \sigma)}{e^G (r_1 - r_2)} e^{i(\frac{\pi}{\varepsilon G} - r_1)} \delta V,$$

$$\lambda_2(s, p) = C_2(p)$$

$$A\pi \left( e^{i(\frac{\pi}{\varepsilon G} - r_1)} - e^{i(\frac{\pi}{\varepsilon G} - r_2)} \right) \delta L_0 - \frac{A\pi \delta v(s, \sigma)}{e^G (r_1 - r_2)} e^{i(\frac{\pi}{\varepsilon G} - r_2)} \delta V,$$

where $C_1, C_2$ are integration constants to be determined with the boundary conditions of (9).

We will prove the following lemmas:

**Lemma 1:** $C_1(p) = C_1(p) e^{\frac{\beta h}{\varepsilon G}}$, where $C_1$ approximates as a Laurent series in $\varepsilon$ with exponential precision in $\frac{1}{\varepsilon}$, and $\delta X_h(p)$ approximates as a power series in $\varepsilon$, with exponential precision in $\frac{1}{\varepsilon}$.

**Lemma 2:** $C_2(p) = \delta X_0(p)$ with exponential precision in $\frac{1}{\varepsilon}$, and $\delta X_0(p)$ approximates as a power series in $\varepsilon$, with exponential precision in $\frac{1}{\varepsilon}$.

We have:

$$\delta X_0(p) = C_1(p) + C_2(p),$$ \hfill (11)

and

$$\delta X_h(p) = C_1(p) e^{r_1 h} + C_2(p) e^{r_2 h}$$

$$A\pi e^{\frac{\beta h}{\varepsilon G}} e^{i(\frac{\pi}{\varepsilon G} - r_1)} - e^{i(\frac{\pi}{\varepsilon G} - r_2)} \delta L_0$$

$$+ \frac{A\pi \delta v(s, \sigma)}{e^G (r_1 - r_2)} e^{i(\frac{\pi}{\varepsilon G} - r_2)} \delta V,$$

$$- \frac{A\pi \delta v(s, \sigma)}{e^G (r_1 - r_2)} e^{i(\frac{\pi}{\varepsilon G} - r_1)} \delta V.$$ \hfill (12)

Let us now have a look on the roots $r_1$, $r_2$. Given (10), the first one expands as a Laurent series in $\varepsilon$, while the second is a power series in $\varepsilon$:

$$r_1 = \frac{\pi}{\varepsilon G} + \frac{p}{\varepsilon G p^2} + O(\varepsilon^2),$$

$$r_2 = -\frac{p}{\varepsilon G p^2} + O(\varepsilon^2).$$

Thus in first approximation, all the right-hand terms in (12) but $C_2(p) e^{r_2 h}$ are of the size $e^{kh\varepsilon}$. Yet, since $X_h = Ae^b + B$ is finite whatever $\varepsilon$, we have necessarily $A = Ae^b$, where the constant $A$ admits a classical asymptotic development in $\varepsilon$. Thus, in (12), only $C_1(p) e^{r_1 h}$, which is a priori of size $e^{kh\varepsilon}$, could diverge when $\varepsilon \to 0$. This divergence would contradict the fact that $\delta X_h$ is finite whatever $\varepsilon$. Thus, necessarily:

$$C_1(p) = \tilde{C}_1(p) e^{\frac{\beta h}{\varepsilon G}}$$ \hfill (13)

to preserve the size of $\delta X_h$.

As an immediate consequence, $\delta X_0 = C_2$ with a residual error in $e^{kh\varepsilon}$. $C_2$ is straightforward from the boundary condition at $s = 0$ in (9):

$$\delta x_0 = (X_0 - \pi_0) \delta L_0 + C_2 (L_0 - \varepsilon G r_2) + O(e^{kh\varepsilon}).$$ \hfill (14)

Since $r_2$ writes as a power series in $\varepsilon$, we have proven Lemma 2.

Using (13) the symbolic equation (12) rewrites:

$$\delta x_h(p) = C_1(p) e^{r_1 h} + C_2(p) e^{r_2 h}$$

$$\frac{A\pi}{e^G (r_1 - r_2)} \left( e^{i(\frac{\pi}{\varepsilon G} - r_1)} - e^{i(\frac{\pi}{\varepsilon G} - r_2)} \right) \delta L_0$$

$$- \frac{A\pi \delta v(s, \sigma)}{e^G (r_1 - r_2)} e^{i(\frac{\pi}{\varepsilon G} - r_2)} \delta V.$$ \hfill (15)

Considering the exact expansions of the roots $r_1$ (Laurent series) and $r_2$ (power series) we see that:

$$\tilde{C}_1 = \frac{A}{e^G} \sum_{i=0}^{+\infty} \beta_i (e^G)^i + \frac{A}{e^G} \sum_{i=0}^{+\infty} \gamma_i (e^G)^i,$$

where the $\beta_i, \gamma_i$ are some constant or $p$-depending coefficients. Since $A$ admits an asymptotic expansion in $\varepsilon$, it comes that the Laurent series $\sum_{i=-1}^{+\infty} C_1 e^G$ is an approximation of $C_1$ with exponential precision in $\frac{1}{\varepsilon}$. Let us consider the first term $\tilde{C}_{1,-1}$. Considering only the terms in $\frac{1}{\varepsilon}$ in (15), we obtain:

$$\tilde{C}_{1,-1} e^{\frac{\beta h}{\varepsilon G}}$$

$$= \frac{A\pi}{e^G} \left( e^{i(\frac{\pi}{\varepsilon G} - r_1)} - e^{i(\frac{\pi}{\varepsilon G} - r_2)} \right) \delta L_0 - \frac{A\pi \delta v(s, \sigma)}{e^G (r_1 - r_2)} e^{i(\frac{\pi}{\varepsilon G} - r_2)} \delta V.$$ \hfill (16)
Thus (15) reads (exactly):
\[
\delta X_h(p) = \sum_{i=0}^{\infty} \tilde{C}_1,i(\epsilon G)^i + \tilde{A} G e^{-v \epsilon G} + \sum_{i=0}^{\infty} \gamma_i(\epsilon G)^i \right)
\]
\[\right) e^{(r_1 - \frac{v}{\epsilon G}) h} + \ldots \]
and given the expansions of \(r_1\) and \(r_2\), a power series in \(\epsilon\) can approximate \(\delta X_h(p)\) up to exponentially small residual terms in \(e^{-\frac{v}{\epsilon G}}\). We have proven Lemma 1.

Choosing an expansion order for the roots \(r_1, r_2\), one can now identify, order by order, the terms of the series approximating \(C_1(p), C_2(p), \delta X_0(p)\) and \(\delta X_h(p)\), using (15), (14) and the boundary conditions in (9). Noting \(r_{1,i}\) (resp. \(r_{2,i}\)) the \(O(\epsilon^i), i \geq 0\) approximation of \(r_1\) (resp. \(r_2\)) we see in (15) that \(\pi - \epsilon G r_{1,i} + O(\epsilon) = 0\) and \(\epsilon G (r_{1,i} - r_{2,i}) + O(1) = 0\), whereas, in (14), \(C_2 \left( L_0 - \epsilon r_{2,i} \right) + O(1) = 0\). Consequently, whatever the chosen roots’ expansion order, one always will estimate \(\delta X_h(p)\) with one order lower precision that \(\delta X_0(p)\) (that is, in \(O(\epsilon^i)\) versus \(O(\epsilon^{i+1})\)).

Using the approximations \(r_{1,1}\) and \(r_{2,1}\), one obtains (detailed calculations are given in [3]):
\[
\delta X_0(p) = \frac{\delta x_0(p) - (X_0 - \pi_0) \delta L_0(p) L}{1 + \frac{\epsilon G}{L} p} + O(\epsilon^2),
\]
\[
\delta X_h(p) = \frac{\pi}{L} \delta y_h + \left( \bar{y}_h - \bar{X}_h \right) \frac{\delta V}{L}
\]
\[\left) + \frac{\pi}{L} \left( \delta x_0(p) - (X_0 - \pi_0) \frac{\delta L_0(p)}{L} \right) e^{-\frac{v}{\pi G}} \right)
\]
\[\left) - \bar{A} e^{-\frac{v}{\pi G}} \frac{\delta L_0(p)}{L} + \bar{\alpha} \bar{A} \frac{\delta V}{L} + O(\epsilon), \right)
\]

\[\delta X_h(p) = \frac{\delta y_h(p) + (\bar{y}_h - \bar{X}_h) \frac{\delta V(p)}{V}}{\bar{\alpha} \left( 1 - \frac{\epsilon G}{\pi V} \right)} + O(\epsilon^2),
\]
\[\delta X_0(p) = \frac{\pi}{L} \delta x_0(p) + \frac{\delta L_0(p) \left( \pi_0 - X_0 \right)}{\alpha V}
\]
\[\left) - \frac{\pi}{\alpha V} \left( \delta y_h(p) + \left( \bar{y}_h - \bar{X}_h \right) \frac{\delta V(p)}{V} \right) e^{-\frac{v}{\pi G}} \right)
\]
\[\left) + \bar{A} e^{-\frac{v}{\pi G}} \frac{\delta L_0(p)}{L} - \bar{A} \frac{\delta V(p)}{V} + O(\epsilon), \right)
\]

where
\[\bar{A} = \frac{X_0 - \bar{X}_h}{e^{-\frac{v}{\pi G}} - 1}.\]

C. Discussion on the approximated transfers

At the boundary where \(\bar{X}\) is the smallest (s = 0 in downward case, s = h in upwards case), the obtained transfer function is a simple mixing equation combined with a first-order filter. The time-constant (in the actual time-scale \(\pi G \left( \sigma_L + \sigma_V \right)\) or \(\pi G \left( \sigma_L + \sigma_V \right)\) depending on the sign of \(V\)) is determined by the local competition between the diffusive and convective effects. Large \(\pi G\) or \(L\) means that the entering composition information efficiently penetrates against the composition gradient. Conversely, large \(\epsilon G\) means that inner domain \(0; h\) feeds the boundary with the unaltered composition information it stores.

At the boundary where \(\bar{X}\) is the largest (s = h in downward case, s = 0 in upwards case), the transfer function is the sum of:

- an instant mixing equation (first line in (19) or (21)). The filtering effect can not be distinguished, since, as explained previously, this transfer function can not be approximated with the same precision in \(\epsilon\) than at the opposite boundary;
- a contribution of the composition variation \(\delta X\) at the opposite boundary (second line in (19) or (21), via the previously discussed mixing equation. Again, the filtering effect is lost. This contribution has traveled along the packed segment at the composition convection speed \(\pi G\) in the reduced timescale, that is, \(\pi G \left( \sigma_L + \sigma_V \right)\) in the natural timescale;
- contribution due to the variation of the liquid and gas flows, only taken into account when the new flows have established all along the segment (third line in (19) or (21)).

Note that, generally speaking, no composition information can travel against the composition convection wave in this model.

Tables I and II summarize the obtained transfers (in \(O(\epsilon)\) only), respectively for the downward and the upward convection case, between dimensionless inputs and outputs. At this order, the transfer functions consist in gains and pure delays only.
TABLE I
DIMENSIONLESS TRANSFER MATRIX (p LAPLACE VARIABLE) FOR A DOWNWARDS COMPOSITION CONVECTION SPEED \( \bar{v} = L - \bar{a} \bar{V} > 0 \).

<table>
<thead>
<tr>
<th>Input | Output</th>
<th>( \frac{\delta X_{h}(p)}{X_{h}} )</th>
<th>( \frac{\delta X_{0}(p)}{X_{0}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \frac{\delta L_{0}(p)}{L} )</td>
<td>( \frac{X_{0} - X_{0} e^{-p \bar{v}}} {X_{h}} )</td>
<td>( \frac{X_{0} - X_{0} e^{-p \bar{v}}} {X_{h}} )</td>
</tr>
<tr>
<td>( \frac{\delta V(p)}{V} )</td>
<td>( \frac{\hat{X}<em>{h}}{\hat{X}</em>{h}} - 1 )</td>
<td>( \frac{\hat{X}<em>{h}}{\hat{X}</em>{h}} - 1 )</td>
</tr>
<tr>
<td>( \frac{\delta x_{0}(p)}{X_{0}} )</td>
<td>( \frac{\hat{X}<em>{h} e^{-p \bar{v}}}{\hat{X}</em>{h}} )</td>
<td>( \frac{\hat{X}<em>{h} e^{-p \bar{v}}}{\hat{X}</em>{h}} )</td>
</tr>
<tr>
<td>( \frac{\delta y_{h}(p)}{Y_{h}} )</td>
<td>( \frac{\hat{X}<em>{h} e^{-p \bar{v}}}{\hat{X}</em>{h}} )</td>
<td>( \frac{\hat{X}<em>{h} e^{-p \bar{v}}}{\hat{X}</em>{h}} )</td>
</tr>
</tbody>
</table>

TABLE II
DIMENSIONLESS TRANSFER MATRIX (p LAPLACE VARIABLE) FOR AN UPWARDS COMPOSITION CONVECTION SPEED \( \bar{v} = L - \bar{a} \bar{V} < 0 \).

<table>
<thead>
<tr>
<th>Input | Output</th>
<th>( \frac{\delta X_{h}(p)}{X_{h}} )</th>
<th>( \frac{\delta X_{0}(p)}{X_{0}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \frac{\delta L_{0}(p)}{L} )</td>
<td>0</td>
<td>( \frac{X_{0} - X_{0} e^{-p \bar{v}}} {X_{h}} )</td>
</tr>
<tr>
<td>( \frac{\delta V(p)}{V} )</td>
<td>( \frac{\hat{X}<em>{h}}{\hat{X}</em>{h}} - 1 )</td>
<td>( \frac{\hat{X}<em>{h}}{\hat{X}</em>{h}} - 1 )</td>
</tr>
<tr>
<td>( \frac{\delta x_{0}(p)}{X_{0}} )</td>
<td>( \frac{\hat{X}<em>{h} e^{-p \bar{v}}}{\hat{X}</em>{h}} )</td>
<td>( \frac{\hat{X}<em>{h} e^{-p \bar{v}}}{\hat{X}</em>{h}} )</td>
</tr>
<tr>
<td>( \frac{\delta y_{h}(p)}{Y_{h}} )</td>
<td>( \frac{\hat{X}<em>{h} e^{-p \bar{v}}}{\hat{X}</em>{h}} )</td>
<td>( \frac{\hat{X}<em>{h} e^{-p \bar{v}}}{\hat{X}</em>{h}} )</td>
</tr>
</tbody>
</table>

IV. EXPERIMENTS
A. Comparison with the convection-diffusion model

We will compare the reduced transfer model to the non-linear PDE model (1) on the column configuration depicted on Fig. 2. Such configuration can be considered as a simplified representation of certain Air Separation Units (ASU) High-Pressure Column. The corresponding linear transfer model is obtained by connecting the downwards and upwards case transfer matrices, as shown on Fig. 3. Table III summarizes the chosen operating values. Note that for \( \hat{G}_{1}, \hat{G}_{2} \), we use the average value of the PDE model \( G(X) \) function over each segment. The liquid speed \( V_{L} = 0.004 \) m/s has been chosen deliberately small, to clearly separate the timescales.

The boundary condition at the top of segment 1 reads in a simplified manner \( \delta X_{1,0} = \alpha_{1} \delta X_{0,1} \), which reflects an instant, total liquefaction in the condenser. Note that one then has \( \delta X_{0,1} = \delta X_{0,1} = \alpha_{1} \delta L_{0,1} \), which resembles the phenomenon pointed out in [9]; in log-scale, the composition dynamics are almost independent from the operating point.

We focus on the \( \hat{G}_{2} \) composition dynamics at the junction between the two segments, that is, \( X_{n,1} \) on Fig. 3. The corresponding transfer is of the form:

\[
\delta X_{n,1}(p) = \left( K_{a} e^{-\theta_{1} p} + K_{b} e^{-\theta_{2} p} \right) \delta V^{n+1}(p) + \left( K_{c} e^{-\theta_{1} p} + K_{d} e^{-\theta_{2} p} \right) \delta V^{n+2}(p) + \left( K_{e} e^{-\theta_{1} p} + K_{f} e^{-\theta_{2} p} \right) \delta L^{n+1}(p) + \left( K_{g} e^{-\theta_{1} p} + K_{h} e^{-\theta_{2} p} \right) \delta L^{n+2}(p),
\]

with constant coefficients \( K_{a...j} \) and \( \theta_{1} = \frac{h_{1}}{\bar{v}_{1}}, \theta_{2} = \frac{h_{2}}{\bar{v}_{2}} \).

\( \Delta_{1} = \frac{h_{1}}{\bar{v}_{1}}, \Delta_{2} = \frac{h_{2}}{\bar{v}_{2}}, \Delta_{12} = \Delta_{1} + \Delta_{2} \). Figure 4 depicts the response of both the PDE and the transfer model to step changes of the top liquid draw \( L_{1}^{out} \). In agreement with the third line of (22), the composition’s response is caused by three successive waves. First, the change on \( L_{1}^{out} \) modifies the composition at the very top of the column, triggering a composition wave which reaches the intermediate liquid draw with delay \( \theta_{1} \approx 9.10^{-3} \) h (this negligible contribution is to be seen in the insert of Fig. 4). Second, a new reflux rate profile is established in the top segment after \( \Delta_{1} \approx 0.35 \) h. Third, after an additional delay \( \Delta_{2} \approx 0.35 \) h, a new reflux rate profile is established in the bottom segment, hence the last composition variation after delay \( \Delta_{12} \approx 0.7 \) h. Note that a change on the air injection \( V^{inj} \) would have triggered more waves. Waves are always in finite number since in the linear model no composition information travels against the composition wave, as mentioned earlier.

For the second and the third wave, the non-linear model is in advance with respect to the linear one, for compositions at a location \( \bar{s} \) are affected by reflux rate disturbances at \( \bar{s} < \bar{s} \) because of diffusive effects. Also, the numerical scheme used for the liquid propagation in the non-linear model is quite diffusive itself.

As seen on Fig. 4 (black line), the static gain of the transfer
model as is yields underestimated composition variations. We have considered in the linear model a linear application \( k() \) with constant first derivative \( \overline{\alpha} = 0.395 \). Yet the convection-diffusion model uses the definition (4), for which we obtain \( k'(X) = \overline{\alpha} = 0.4687 \) at the intermediate draw location, at the operating point. We then correct the linear model output with a multiplicative factor \( \overline{\alpha}/\alpha \), which yields the correct result (Fig. 4, grey line).

### B. Comparison against real plant data

We will now test the linear model against data acquired from the High Pressure (HP) column of an operational Air Separation Unit. The HP column consists in three packed segments, which can be represented by alternate upwards and downwards segments, as depicted on Fig. 5. The column is fed at its bottom with gaseous atmospheric air, and with a mixed liquid-gas air flow atop the bottom segment. Almost pure \( N_2 \) is drawn out at the top of the column.

We focus on the second liquid draw (lean liquid draw), where \( O_2 \) molar fraction is about 2%. These four flows are measured. Compositions in the flash drum are estimated using an isenthalpic flash model and upstream pressure and temperature measurements. \( O_2 \) composition is measured in the pure \( N_2 \) draw and the lean liquid draw only. Then, to dispose of the internal information required for the linear model tuning, we first fit a convection-diffusion model (1), (2), (3) to the column. The procedure is described in [3] yet essentially consists in fitting the diffusion parameter \( \epsilon \). As in the previous experiment, we obtain the linear model parameters as averaged values from the convection-diffusion model over each segment. Figure 6 shows the \( O_2 \) composition variations in the lean liquid, as estimated by the convection-diffusion model and the linear model, compared to the measured values, in response to the measured inlet and outlet flows variations and estimated inlet compositions variations. Input and output flows vary between +10 % and -15% of their initial value; inlet compositions vary between +0.5% and -1% of their initial value.

With fixed \( \epsilon \), the linear model reproduces the oscillations of the measured \( O_2 \) composition whose duration varies between 30 min and 2 h. Zoom on Fig. 7 shows in addition that shorter bumps are reproduced in an unfiltered yet synchronized manner.

On the long term a slowly time-varying bias appears (as for the convection-diffusion model). This can be corrected by slowly adapting the parameter \( \epsilon \), according to:

\[
\frac{d\epsilon}{dt} = \frac{1}{T_i} \frac{\dot{X}_{O_2}^{LL}(t) - \dot{X}_{O_2}^{LL}(t)}{\dot{X}_{O_2}^{LL}(t)},
\]

where \( \dot{X}_{O_2}^{LL}(t) \) is the lean liquid \( O_2 \) molar fraction estimated by the convection-diffusion model, \( \dot{X}_{O_2}^{LL}(t) \) is the measure, and \( T_i \) is about 1 h. This adaptive \( \epsilon \) can be used as a time-dependent parameter for the linear model, which yields the improved results of Fig. 8. As said in II, \( \epsilon \) is related to the packing efficiency, yet its dependence on the column’s set-point is unknown for the moment, hence the online estimation. Yet pre-estimations could be obtained by fitting the model off-line to reference steady-state simulations of the plant.

### V. CONCLUSION

From a non-linear convection-diffusion model for the dynamics of composition profiles in distillation columns, we derived a simplified, linear transfer functions model for binary separation. This model is obtained through an unusual asymptotic expansion versus the (small) diffusion term, involving Laurent series with exponentially precise terms.

The transfer model divides the distillation column in connected homogeneous sections; for each section, it connects the composition variations at the outlets with the variations of the composition and flow at the inlets. This model involves mixing effects, propagation of composition

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1One may also envisage to identify the gain coefficients of a parallel delay-lines formulation similar to (22) directly from on-site measurements; the delays would then be estimated from the column’s dimensions and the hydraulics at the investigated set-point.

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**TABLE III**

**OPERATING POINT PARAMETERS FOR THE 2-SEGMENTS COLUMN LINEAR TRANSFER MODEL**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>( h )</th>
<th>( \bar{\alpha} )</th>
<th>( G )</th>
<th>( \epsilon )</th>
</tr>
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<tbody>
<tr>
<td>Segment 1</td>
<td>5</td>
<td>0.395</td>
<td>250.16</td>
<td>0.2</td>
</tr>
<tr>
<td>Segment 2</td>
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<td>0.995</td>
<td>252.19</td>
<td>0.2</td>
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<td>Unit</td>
<td>( m )</td>
<td>( mol/m^3 )</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Parameter</td>
<td>( \dot{X}_0 )</td>
<td>( \dot{X}_h )</td>
<td>( \dot{x}_0 )</td>
<td>( \dot{g}_h )</td>
</tr>
<tr>
<td>Segment 1</td>
<td>( 1.64 \times 10^{-6} )</td>
<td>0.1355</td>
<td>0.0000</td>
<td>0.0035</td>
</tr>
<tr>
<td>Segment 2</td>
<td>0.0000</td>
<td>0.4022</td>
<td>0.0000</td>
<td>0.0021</td>
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<tr>
<td>Unit</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Parameter</td>
<td>( L )</td>
<td>( V )</td>
<td>( \dot{V} )</td>
<td>( \dot{\theta} )</td>
</tr>
<tr>
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<td>1786.10</td>
<td>857.33</td>
<td>5482.70</td>
</tr>
<tr>
<td>Segment 2</td>
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<td>1786.10</td>
<td>-26.79</td>
<td>5482.70</td>
</tr>
<tr>
<td>Unit</td>
<td>( mol/s )</td>
<td>( mol/s )</td>
<td>( mol/s )</td>
<td>( mol/m )</td>
</tr>
</tbody>
</table>

---

Fig. 4. **Top:** relative variations of the top liquid draw \( L_1^{out} \). **Bottom:** composition variations in the intermediate liquid draw, according to the convection-diffusion model (dashed), to the linear model (solid black), and to the linear model, with correction on the actual relative volatility (solid grey).
waves and progressive establishment of the internal reflux. At the chosen order of expansion, the transfers consist in static gains and pure delays only. The transfer model yet satisfyingly reproduces the dynamics of $N_2/O_2$ separation in an industrial air separation unit, compared to the convection-diffusion model or to plant measurements.

In addition, we show that the transfer model is easily fitted to the plant, since its parameters either are connected to design or operating data (such as inlet or outlet flows), or can be estimated using the convection-diffusion model. The latter can also be coupled to the transfer model to dynamically adapt the parameters which depend on the diffusion term, yielding improved plant-model agreement.

Further work should focus on turning the length of the sections into time-varying parameters, such that the inversion of the composition convection speed does not necessarily take place outside the physical segments. In addition, better handling of the impact of the internal flows on the diffusion terms should be considered. One should also address the direct identification of the column's transfer in the form of parallel delay-lines as outlined in Section IV.

REFERENCES


