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Compartmental modeling of high purity air separation columns

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Abstract

High purity distillation columns are critical unit operations in cryogenic air separation plants. The development of nonlinear control technology is motivated by the need to frequently change production rates in response to time varying utility costs. Detailed column models based on stage-by-stage balance equations are too complex to be incorporated directly into optimization-based strategies such as nonlinear model predictive control. In this paper, we develop reduced order dynamic models for the upper column of a cryogenic air separation plant by applying time scale arguments to a detailed stage-by-stage model that includes mass and energy balances and accounts for non-ideal vapor–liquid equilibrium. The column is divided into compartments according to the locations of liquid distributors and feed and withdrawal streams. The differential equations describing each compartment are placed in singularly perturbed form through the application of a physically based coordinate transformation. Application of singular perturbation theory yields a differential–algebraic equation model with significantly fewer differential variables than the original stage-by-stage model. A rigorous column simulator constructed using Aspen Dynamics (Aspen Technology) is used to access the tradeoff between reduced order model complexity and accuracy as the number of compartments is varied.

Keywords: Air separation; Distillation column; Dynamic modeling; Nonlinear control

1. Introduction

Linear model predictive control (LMPC) is the most successful multivariable control technology in the chemical process industries (Qin & Badgwell, 1997). The success of LMPC is largely attributable to its optimization formulation that allows process constraints to be accommodated in a systematic fashion. The inherent assumption of linear process dynamics restricts the application of LMPC to relatively small operating regimes. Nonlinear control technology is often required for highly nonlinear processes that are subject to large and/or frequent changes in operating conditions. A prototypical example of this small but important class of nonlinear processes is continuous polymerization reactors used to produce multiple polymer grades (Debling et al., 1994; McAuley & MacGregor, 1992). While other controller design strategies such as feedback linearization (Henson & Seborg, 1997, chap. 4; Kravaris & Kantor, 1990) are available, nonlinear model predictive control (NMPC) appears to be the most promising nonlinear control technology for industrial applications (Allgower & Zheng, 2000).

NMPC is an extension of LMPC in which a nonlinear dynamic model is used to predict and optimize future process performance. NMPC controllers are commonly implemented using a simultaneous solution strategy in which the dynamic model equations are temporally discretized and posed as equality constraints in a nonlinear optimization problem (Henson, 1998; Meadows & Rawlings, 1997, chap. 5). Future input and state variables are treated as decision variables, thereby producing a potentially large nonlinear programming problem that must be solved repeatedly in real time. Computational complexity is inextricably linked to the nonlinear dynamic model used for controller design. Commercial NMPC technology (Qin & Badgwell, 1998) is effectively restricted to low-order nonlinear models such as those commonly developed for polymerization reactors.

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High purity distillation columns are an important class of nonlinear processes that are not directly amenable to the application of NMPC technology due to the high-order nature of stage-by-stage balance models (Luyben, 1973). Two implementation strategies have been pursued: (1) the development of sophisticated solution methods that allow direct incorporation of detailed column models and (2) the derivation of reduced order controller design models from stage-by-stage models to reduce the computational requirements. The first approach exploits the large number of algebraic variables typically present in differential–algebraic equation models of distillation columns (Leineweber, 1998). A significant fraction of NMPC computational overhead is attributable to calculation of the Jacobian and Hessian matrices. The NMPC optimization problem can be decomposed such that matrix elements associated with the algebraic variables can be excluded from these calculations. When combined with other computational enhancements including multiple shooting and reduced successive quadratic programming, the decomposition strategy allows the application of NMPC to stage-by-stage balance models of moderate complexity (Kronseder, von Stryk, Bulirsch, & Kroner, 2001; Nagy et al., 2000). However, the applicability of this approach is ultimately limited by the complexity of the column model.

The derivation of reduced order column models from more detailed stage-by-stage models has received considerable attention. While other methods are available, reduced order modeling techniques based on collocation, wave theory and compartmentalization are representative. Collocation methods involve the construction of a spatial mesh and the formulation of dynamic balance equations at the collocation points (Levien, Stewart, & Morari, 1985; Yang & Lee, 1997). A reduced order model is obtained by choosing the number of collocation points to be considerably less than the number of separation stages. Many studies are strictly focused on steady-state collocation models used for distillation column design (Seferlis & Grievink, 2001; Seferlis & Hrymak, 1994a, 1994b; Swartz & Stewart, 1986). As compared to the compartmentalization approach discussed below, collocation techniques suffer from a number of limitations including (Carta, Tola, Servida, & Morbidelli, 1995a, 1995b; Yang & Lee, 1997): (1) the collocation points may not correspond to actual separation stages; (2) state variables at other column locations must be determined by interpolation; (3) model accuracy is limited by the number of collocation points; (4) inaccurate predictions can be obtained for columns with steep profiles and/or discontinuities due to feed and withdrawal streams.

The wave modeling approach is based on the premise that the column profile can be described by a moving wave front with constant pattern (Kienle, 2000; Marquardt, 1986; Marquardt & Amrhein, 1994). Under simplifying assumptions including binary separation, ideal vapor–liquid equilibrium, constant equimolar overflow and constant molar holdups, a single differential equation for the wave position that allows the entire composition profile to be reconstructed can be derived. Unfortunately, the simplifying assumptions that underpin nonlinear wave theory are rarely satisfied in practice. For example, wave models are incapable of accounting for end effects and producing the self-sharpening behavior of non-ideal columns. As a result wave models have limited predictive capability.

Compartmental models are derived directly from stage-by-stage balance models by dividing the column into a small number of sections termed compartments (Benallou, Seborg, & Mellichamp, 1986; Horton, Bequette & Edgar, 1991; Levine & Rouchon, 1991). A dynamic model of each compartment is developed by combining stage-by-stage balances with overall balances over the entire compartment. If the number of stages in the compartment is sufficiently large and each stage has a comparable liquid holdup, then the overall dynamics of the compartment are much slower than the dynamics of any individual stage within the compartment. This time scale separation allows the compartment dynamics to be approximated with the differential equations for a representative stage whose holdup is equal to the total compartment holdup (Levine & Rouchon, 1991). The balance equations for other stages within the compartment are reduced to algebraic equations due to their relatively fast dynamics. Advantages of the compartmental modeling approach include: (1) perfect steady-state agreement with the associated stage-by-stage model is ensured; (2) a suitable tradeoff between model complexity and dynamic accuracy can be achieved by adjusting the number of compartments; (3) the triangular structure of the original stage-by-stage model is retained.

Compartmental models are comprised of relatively few differential equations representing the overall compartment dynamics and a potentially large number of algebraic equations derived from the dynamic stage balances. NMPC simultaneous solution methods require that the algebraic variables be included as decision variables and temporally discretized versions of the algebraic equations be posed as equality constraints. Consequently the computational advantage of reducing most of the differential equations to algebraic equations is not readily apparent. Recently, highly efficient NMPC solution methods for sparse differential–algebraic equation (DAE) models have been developed (Leineweber, 1998) and applied to distillation columns (Kronseder et al., 2001; Nagy et al., 2000). Although beyond the scope of this paper, we envision that compartmental models are ideally suited for these solution methods due to the large number of algebraic variables and the sparseness of the Jacobian matrix.

We are particularly interested in developing nonlinear modeling and control technology for cryogenic distillation columns in air separation plants. This research is motivated by the need to frequently adjust production rates in response to time varying electricity costs caused by deregulation of the electrical utility industry. Our previous work has focused on the derivation of reduced order nonlinear models using nonlinear wave theory (Zhu, Henson, & Megan, 2001) and compartmentalization (Khowinij, Henson, Belanger, & Megan, in press) as well as the development of a NMPC strategy based

on the wave model (Bian, Henson, Belanger, & Megan, 2005) for a nitrogen purification column. The purpose of this paper is to evaluate reduced order compartmental models for the upper column of a double column plant used to produce both nitrogen and oxygen products. A longer term goal is to utilize these compartmental models to design NMPC controllers through the application of customized solution methods that exploit the DAE model structure (Leineweber, 1998).

The remainder of the paper is organized as follows. In Section 2, our previous research on nitrogen purification columns is briefly reviewed to provide context for the present work. The development of an Aspen Dynamics simulator, a stage-by-stage balance model and reduced order compartmental models for the upper column is discussed in Section 3. Section 4 contains simulation results that demonstrate the relative performance of the various models. Finally, a summary and conclusions are presented in Section 5.

2. Previous work on nitrogen purification columns

Our previous research on reduced order modeling of cryogenic air separation plants focused on the nitrogen purification column shown in Fig. 1. The column has 42 equilibrium stages, a distributor located in the middle of the column to improve liquid flow characteristics, a sump located at the bottom of the column which holds a large liquid inventory and an integrated condenser/reboiler in which the partially flashed bottom stream is vaporized and the nitrogen vapor stream from the top of the column is condensed to produce the reflux and liquid nitrogen product streams (Zhu et al., 2001). The feed is introduced immediately above the liquid sump, and a portion of the overhead stream is withdrawn as the gaseous nitrogen product. A dynamic simulator of the column and the integrated condenser/reboiler (Bian et al., 2005) was developed using Aspen Dynamics (Aspen Technology) and used as a surrogate for the nitrogen plant in our simulation studies.

We have developed reduced order nonlinear models of the nitrogen purification column using nonlinear wave theory (Bian et al., 2005; Zhu et al., 2001). A single differential equation was used to track the position of the nitrogen composition wave front. When combined with a profile expression that approximates the wave pattern, the predicted wave position allowed the entire nitrogen composition profile to be constructed. Simulation tests for production rate changes showed that the nonlinear wave model captured the essential dynamic behavior of the Aspen model. However, the Aspen composition profiles exhibited self sharpening behavior due to nonlinear equilibrium relations as well as wave distortion near the ends of the column. The wave model produced large dynamic and steady-state prediction errors due to its inability to capture such non-ideal effects. We have shown that combined state and parameter estimation is a feasible approach to improve the predictive capability of the nonlinear wave model (Bian et al., 2005).

Recently we have developed reduced order compartmental models that provided a considerably more detailed description of the nitrogen purification column dynamics than was possible with the wave modeling approach (Khowinij et al., in press). The stage-by-stage model used for compartmental model derivation included ternary component balances and liquid hydraulic relations. The condenser, liquid distributor and feed stage were treated as single stage compartments due to their relatively large liquid holdups. Two reduced order models were formulated by dividing the column sections above and below the distributor into two compartments (five compartment model) or four compartments (seven compartment model). For example, the stage-by-stage model consisting of 130 differential equations was approximated by the seven compartment model with 19 differential equations and 111 algebraic equations. The two compartmental models were dynamically simulated using the DAE solver available in `MATLAB`. Each model produced close agreement with the stage-by-stage model and satisfactory predictions as compared to the considerably more detailed Aspen simulator. A typical simulation time for the seven compartment model was approximately 20% of that required for the stage-by-stage model.

In this paper, compartmental models for the upper column of the double column air separation plant shown in Fig. 1 are developed and evaluated. This work is an essential step towards our long-term goal of developing nonlinear modeling and control technology for double column and triple column air separation plants. With respect to our previous research on nitrogen purification columns, novel aspects of the present work include:

1. Construction of an Aspen simulator of the upper column that accounts for the multiple feed and withdrawal streams and liquid distributors located along the column.
2. Development of a more accurate stage-by-stage model for compartmental model derivation. Simplifying assumptions including constant equimolar overflow and constant relative volatility are relaxed by adding energy balances and non-ideal vapor–liquid equilibrium relations.
3. A more detailed investigation of different compartmentalization strategies on reduced order model complexity and computational efficiency.

3. Upper column modeling

A schematic representation of the upper column is shown in Fig. 1. The column has 59 stages including the reboiler and seven liquid distributors located throughout the column (stages 1, 10, 18, 26, 35, 43 and 51). The column receives two air feed streams (liquid air on stage 18 and turbine air on stage 35) following compression and expansion that achieves the cryogenic temperatures necessary for separation. The upper column is coupled to the lower column through the integrated condenser/reboiler, the reflux stream on stage 1 obtained from

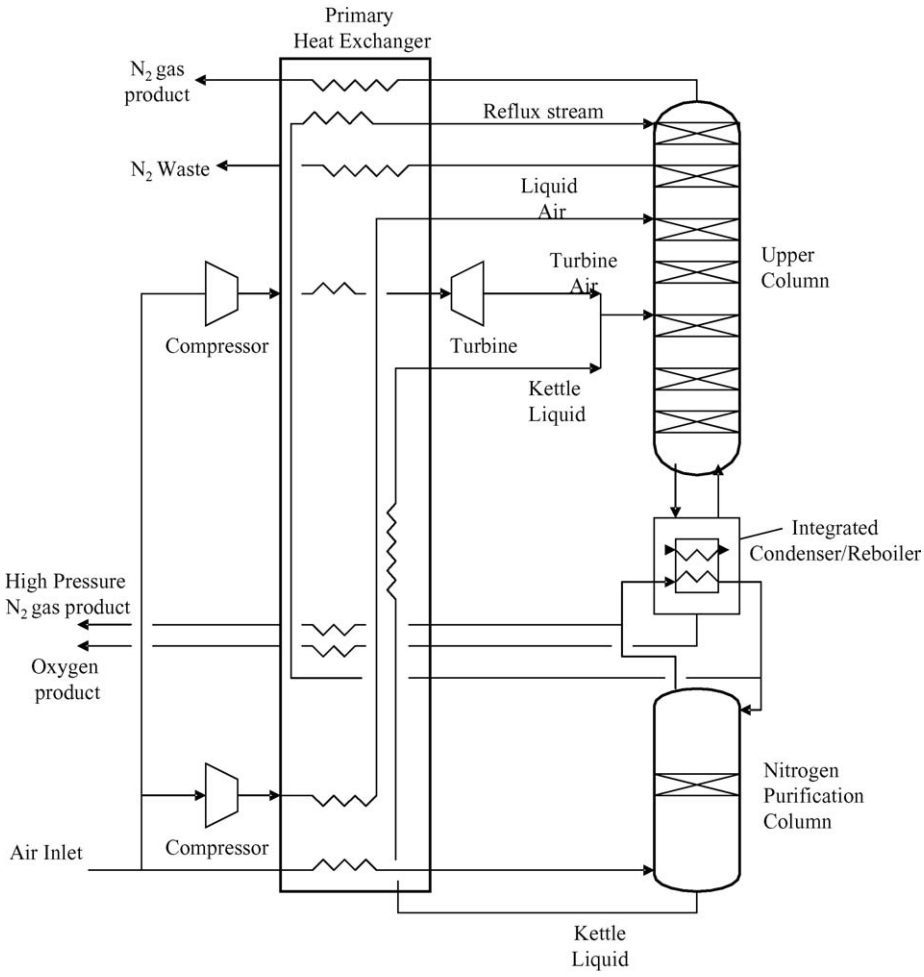

Fig. 1. Schematic diagram of a typical double column air separation plant.

the overhead of the lower column and kettle liquid feed obtained from the bottom of the lower column. The nitrogen product is withdrawn from stage 1 and a gaseous nitrogen waste stream is withdrawn from stage 10. Because the focus of this paper is upper column modeling, the lower column and the compression, expansion and heat exchange equipment are neglected. Therefore, the scope is limited to the upper column with reboiler, and the various feed and withdrawal streams are treated as independent inputs.

3.1. Aspen simulator

A detailed dynamic simulator of the upper column was developed using Aspen Dynamics (Aspen Technology). The Aspen column model `RadFrac` was used to solve the dynamic component balances and steady-state energy balances for each stage. Modeling of the lower column was avoided by treating the integrated condenser/reboiler as a simple reboiler with a constant condenser side temperature. The thermodynamic models used were NRTL for the liquid phase and Peng–Robinson for the vapor phase. Proprietary thermodynamic property data for the air components (nitrogen, oxygen and argon) were provided by Praxair. PID controllers were implemented for regulation of the reboiler level and the overhead pressure. Equipment specifications and the steady-state operating conditions listed in Table 1 were obtained from a typical Praxair double column air separation plant. The Aspen simulator was used to represent the upper column in our modeling studies. To evaluate the compartmental models over a reasonable range of operating conditions, two other steady states corresponding to liquid air feed flow rate changes of $\pm 25\%$ from the nominal value were also investigated. This feed stream was chosen for manipulation because it represents the major source of air to the column and is independent of the unmodeled lower column. A more realistic disturbance caused by a production rate change would require an integrated model of the lower and upper columns that is beyond the scope of this work.

3.2. Stage-by-stage balance model

A stage-by-stage balance model was derived to provide the basis for compartmental model development. Model derivation was based on the following simplifying assumptions:

Table 1
Nominal Operating Conditions of the Upper Column

Variable	Symbol (Units)	Value
N_2 product flow rate	P_{N_2} (kmol/h)	330.99
N_2 product impurity	$y_{O_2,P_{N_2}}$ (ppm)	1.15
O_2 product flow rate	P_{O_2} (kmol/h)	258.443
O_2 product impurity	$y_{O_2,P_{O_2}}$ (kmol/kmol)	0.9930
Average stage liquid holdup	$\bar{M}_L$ (kmol)	0.755
Average stage vapor holdup	$\bar{M}_V$ (kmol)	0.0970
Distributor liquid holdup	M_1 (kmol)	5.085
Distributor liquid holdup	M_{10} (kmol)	10.151
Distributor liquid holdup	M_{18} (kmol)	10.872
Distributor liquid holdup	M_{26} (kmol)	11.708
Distributor liquid holdup	M_{35} (kmol)	11.442
Distributor liquid holdup	M_{43} (kmol)	12.438
Distributor liquid holdup	M_{51} (kmol)	12.439
Reboiler liquid holdup	M_{59} (kmol)	43.560

(1) negligible vapor phase holdups; (2) ideal vapor phase behavior; (3) fast temperature dynamics; (4) linear pressure profiles across the column; (5) complete mixing of the vapor and liquid streams entering the feed stages; (6) constant reboiler medium temperature. The stage-by-stage model is comprised of dynamic component balances for oxygen and argon, dynamic overall mass balances, steady-state energy balances and an activity coefficient model to account for non-ideal liquid phase behavior. The model equations for the i-th separation stage are:

$$\begin{aligned}
M_i\frac{dx_{O_2,i}}{dt} &= L_{i-1}x_{O_2,i-1} + V_{i+1}y_{O_2,i+1} - L_i x_{O_2,i} - V_i y_{O_2,i} - x_{O_2,i}\frac{dM_i}{dt}\\
M_i\frac{dx_{Ar,i}}{dt} &= L_{i-1}x_{Ar,i-1} + V_{i+1}y_{Ar,i+1} - L_i x_{Ar,i} - V_i y_{Ar,i} - x_{Ar,i}\frac{dM_i}{dt}\\
\frac{dM_i}{dt} &= L_{i-1} + V_{i+1} - L_i - V_i\\
0 &= L_{i-1}h^L_{i-1} + V_{i+1}h^V_{i+1} - L_i h^L_i - V_i h^V_i - h^L_i\frac{dM_i}{dt}\\
1 &= y_{O_2} + y_{N_2} + y_{Ar}
\end{aligned} \tag{1}$$

where M is the liquid holdup, L and V the liquid and vapor molar flow rates, respectively, x and y component mole fractions in the liquid and vapor phases, respectively, and h^L and h^V are liquid and vapor phase enthalpies, respectively. Aspen physical property data were used to develop temperature dependent correlations for the pure component liquid and vapor enthalpies. Stream enthalpies were calculated from the pure component enthalpies using linear mixing rules. A linear relationship between liquid holdup and liquid molar flow rate was regressed from Aspen simulation data:

$$L_i = k_i M_i \tag{2}$$

where k is stage hydraulic coefficient.

The vapor–liquid equilibrium relation is written as:

$$\begin{aligned}
&y_{n,i} = \kappa_i\gamma_{n,i}K_{n,i}x_{n,i} + (1-\kappa_i)y_{n,i+1},\\
&n = N_2,\ O_2,\ Ar
\end{aligned} \tag{3}$$

where K is the ideal temperature dependent vapor–liquid equilibrium constant, γ the activity coefficient and κ is the Murphee stage efficiency. The ideal vapor–liquid equilibrium constant was calculated as:

$$K_{n,i} = \frac{P_n^{sat}(T_i)}{P_i} \tag{4}$$

where the Antoine equation was used to compute the pure component saturation pressures P_n^{sat}. A linear increase of the stage pressure from a constant overhead pressure (P_1) was assumed:

$$P_i = P_1 + (i-1)\Delta P \tag{5}$$

A constant individual stage pressure drop was correlated to the reboiler vapor rate (V_r) using Aspen simulation data:

$$\Delta P = \beta V_r^2 \tag{6}$$

where β is a constant. The activity coefficients were calculated using the Margules equation for multicomponent mixtures (Prausnitz, Lichtenthaler, & de Azevedo, 1986):

$$\begin{aligned}
\gamma_{N_2,i} &= \exp\left[\frac{A_{N_2,O_2}x^2_{O_2,i} + A_{N_2,Ar}x^2_{Ar,i} + (A_{N_2,O_2} + A_{N_2,Ar} - A_{O_2,Ar})x_{O_2,i}x_{Ar,i}}{RT_i}\right]\\
\gamma_{O_2,i} &= \exp\left[\frac{A_{N_2,O_2}x^2_{N_2,i} + A_{O_2,Ar}x^2_{Ar,i} + (A_{N_2,O_2} + A_{O_2,Ar} - A_{N_2,Ar})x_{N_2,i}x_{Ar,i}}{RT_i}\right]\\
\gamma_{Ar,i} &= \exp\left[\frac{A_{N_2,Ar}x^2_{N_2,i} + A_{O_2,Ar}x^2_{O_2,i} + (A_{N_2,Ar} + A_{O_2,Ar} - A_{N_2,O_2})x_{N_2,i}x_{O_2,i}}{RT_i}\right]
\end{aligned} \tag{7}$$

where T is the absolute temperature, R the ideal gas constant and the Margules constant, $A_{j,k}$, accounts for liquid phase interactions between components j and k. Proprietary values of the Margules and Antoine constants were provided by Praxair.

Steady-state mass and enthalpy balances were used to model mixing of the four feed streams shown in Fig. 2 with internal vapor and liquid streams. Each liquid distributor was modeled as a separation stage with negligible vapor holdup and a very small stage efficiency (5%). Modeling of the reboiler was simplified by assuming a constant condenser side temperature as in the Aspen simulator. A PI controller was designed to regulate the reboiler level by manipulation

of the oxygen product flow rate. The controller tuning parameters were identical to those used in the Aspen simulator: $k_c = -5000$ %level/(kmol/h) and $\tau_i = 2$ s. The complete stage-by-stage model consisted of 178 differential equations and 134 algebraic equations with the following unknowns: $x_{O_2,i}$, $x_{Ar,i}$, L_i, V_i and T_i. The MATLAB code `ode15s` was used to solve the DAE model.

3.3. Compartmental models

The compartmentalization approach was used to derive reduced order dynamic models from the stage-by-stage model described above. To examine the effect of compartmentalization on reduced order model complexity and accuracy, the following three cases shown in Fig. 2 were investigated:

- 15 *Compartment model*: The reboiler (stage 59) and the seven distributors (stages 1, 10, 18, 26, 35, 43 and 51) were treated as separate compartments due to their large liquid holdups listed in Table 1. Equilibrium stages located between adjacent distributors were lumped together as multistage compartments.
- 9 *Compartment model*: Distributors located between feed and/or withdrawal points were lumped together with equi-

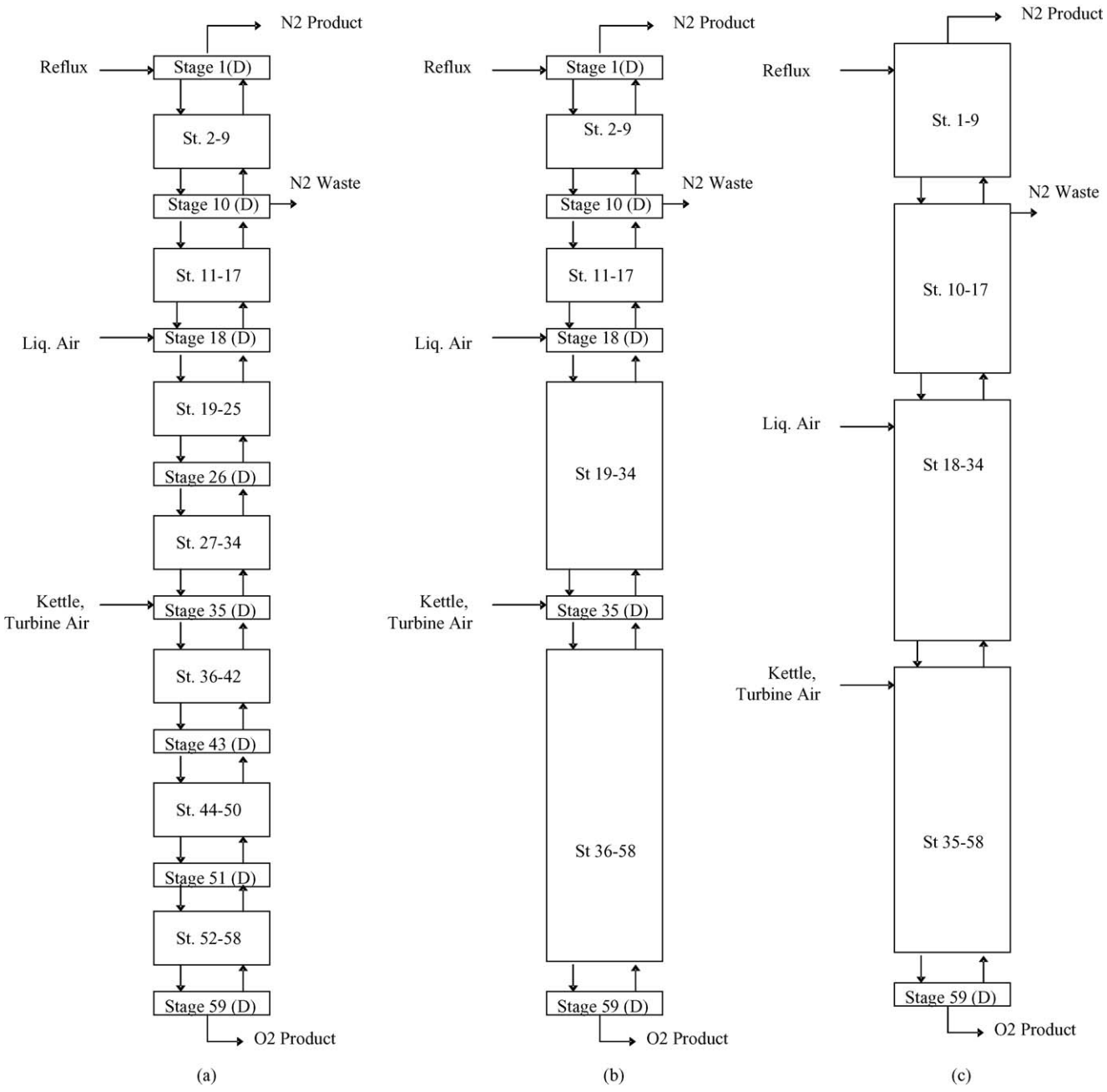

Fig. 2. Three alternative compartmentalization schemes: (a) 15 compartments; (b) 9 compartments; (c) 5 compartments.

librium stages such that only a single compartment was located between any two feed and/or product streams.

- 5 *Compartment model*: Each compartment contained a single feed and withdrawal stream located at the first stage of the compartment. The reboiler was treated as a single stage compartment due to its relatively large liquid holdup.

The compartmental models were derived by transforming the stage-by-stage model into singularly perturbed form and neglecting the fast dynamics (Levine & Rouchon, 1991). Single stage compartments cannot be simplified because the associated model equations do not exhibit a time scale separation. The model-order reduction procedure for multistage compartments can be summarized as follows: (1) replace the dynamic balances for a chosen equilibrium stage with dynamic balances derived for the entire compartment; (2) introduce a singular perturbation parameter that represents the ratio of the liquid holdup of an individual equilibrium stage to the liquid holdup of the entire compartment; (3) reduce the differential equations for the individual equilibrium stages to algebraic equations by setting the singular perturbation parameter to zero. As discussed earlier, we have applied this approach to a simpler nitrogen purification column model (Khowinij et al., in press) under the assumptions of equimolar overflow and ideal vapor–liquid equilibrium. Here we outline the compartmental modeling procedure for the upper column stage-by-stage model with steady-state energy balances and non-ideal vapor–liquid equilibrium relations. A detailed derivation is omitted for the sake of brevity.

Consider the dynamic balances for a multistage compartment comprised of m equilibrium stages. Overall component balances about the compartment yield:

$$\begin{aligned}
\frac{\mathrm{d}M_c x_{n,c}}{\mathrm{d}t} &= L_0 x_{n,0} + V_{m+1} y_{n,m+1} - L_m x_{n,m} - V_1 y_{n,1} \\
\frac{\mathrm{d}M_c}{\mathrm{d}t} &= L_0 + V_{m+1} - L_m - V_1 \\
h_c^{\mathrm{L}} \frac{\mathrm{d}M_c}{\mathrm{d}t} &= L_0 h_0^{\mathrm{L}} + V_{m+1} h_{m+1}^{\mathrm{V}} - L_m h_m^{\mathrm{L}} - V_1 h_1^{\mathrm{V}}
\end{aligned} \tag{8}$$

where the subscript n denotes oxygen or argon, and the subscripts 0 and $m+1$ are used to represent liquid properties from the stage immediately above the compartment and vapor properties from the stage immediately below the compartment, respectively. The total liquid holdup, holdup averaged compositions and holdup averaged liquid enthalpy of the compartment are defined as:

$$\begin{aligned}
M_c &= \sum_{i=1}^{m} M_i, \qquad x_{n,c} = \frac{\sum_{i=1}^{m} M_i x_{n,i}}{\mathrm{d}M_c}, \\
h_c^{\mathrm{L}} &= \frac{\sum_{i=1}^{m} M_i h_i^{\mathrm{L}}}{\mathrm{d}M_c}
\end{aligned} \tag{9}$$

A time scale separation is introduced by replacing the dynamic balances for the k-th stage in the compartment with the overall balances (8). Although previous studies have suggested that the location of this so-called "sensitive stage" does not have a significant effect on reduced order model accuracy (Khowinij et al., in press; Levine & Rouchon, 1991), we investigate this issue further in the next section. By defining $\frac{M_i}{M_c} = \alpha_i \epsilon$ where $\alpha_i \approx 1$ accounts for O(1) differences among individual stage holdups and $\epsilon \ll 1$, the stage-by-stage model equation can be placed in singularly perturbed form where the average compartment variables M_c and $x_{n,c}$ are the slow variables and the individual stage variables variables M_i and $x_{n,i}$ are the fast variables (Levine & Rouchon, 1991). The following reduced order model equations are obtained by setting $\epsilon = 0$ according to singular perturbation theory (Kokotovic, Khalil, & O'Reilly, 1999):

$$\begin{aligned}
0 &= L_0 x_{n,0} + V_2 y_{n,2} - L_1 x_{n,1} - V_1 y_{n,1} \\
0 &= L_0 + V_2 - L_1 - V_1 \\
0 &= L_0 h_0^{\mathrm{L}} + V_2 h_2^{\mathrm{V}} - L_1 h_1^{\mathrm{L}} - V_1 h_1^{\mathrm{V}} \\
&\vdots \\
0 &= L_{k-2} x_{n,k-2} + V_k y_{n,c} - L_{k-1} x_{n,k-1} - V_{k-1} y_{n,k-1} \\
0 &= L_{k-2} + V_k - L_{k-1} - V_{k-1} \\
0 &= L_{k-2} h_{k-2}^{\mathrm{L}} + V_k h_k^{\mathrm{V}} - L_{k-1} h_{k-1}^{\mathrm{L}} - V_{k-1} h_{k-1}^{\mathrm{V}} \\
M_c \frac{\mathrm{d}x_{n,c}}{\mathrm{d}t} &= L_0 x_{n,0} + V_{m+1} y_{n,m+1} - L_m x_{n,m} - V_1 y_{n,1} \\
&\quad - x_{n,c} \frac{\mathrm{d}M_c}{\mathrm{d}t} \\
\frac{\mathrm{d}M_c}{\mathrm{d}t} &= L_0 + V_{m+1} - L_m - V_1 \\
0 &= L_0 h_0^{\mathrm{L}} + V_{m+1} h_{m+1}^{\mathrm{V}} - L_m h_m^{\mathrm{L}} - V_1 h_1^{\mathrm{V}} - h_c^{\mathrm{L}} \frac{\mathrm{d}M_c}{\mathrm{d}t} \\
0 &= L_k x_{n,c} + V_{k+2} y_{n,k+2} - L_{k+1} x_{n,k+1} - V_{k+1} y_{n,k+1} \\
0 &= L_k + V_{k+2} - L_{k+1} - V_{k+1} \\
0 &= L_k h_c^{\mathrm{L}} + V_{k+2} h_{k+2}^{\mathrm{V}} - L_{k+1} h_{k+1}^{\mathrm{L}} - V_{k+1} h_{k+1}^{\mathrm{V}} \\
&\vdots \\
0 &= L_{m-1} x_{n,m-1} + V_{m+1} y_{n,m+1} - L_m x_{n,m} - V_m y_{n,m} \\
0 &= L_{m-1} + V_{m+1} - L_m - V_m \\
0 &= L_{m-1} h_{m-1}^{\mathrm{L}} + V_{m+1} h_{m+1}^{\mathrm{V}} - L_m h_m^{\mathrm{L}} - V_m h_m^{\mathrm{V}}
\end{aligned} \tag{10}$$

The 15 compartment model shown in Fig. 2a is completed by adding the balance equations and vapor–liquid equilibrium relations for the single stage compartments. An analogous procedure is used to derive the reduced order equations for the 9 compartment and 5 compartment models shown in Fig. 2b and c, respectively. The resulting reduced order model equations are not reported here. Table 2 shows that compartmentalization produces a large reduction in the number of differential variables as compared to the stage-by-stage model. The effect of model-order reduction on prediction accuracy and computational efficiency is explored in the next section.

Table 2
Complexity of the stage-by-stage and compartmental models

Model	Differential variables	Algebraic variables
Stage-by-stage	178	134
15 Compartment	46	266
9 Compartment	28	284
5 Compartment	16	296

4. Results and discussion

The first set of simulation results were generated by: (1) selecting the sensitive stage of each multistage compartment to be located in the center of the compartment and (2) adjusting stage efficiencies in the top section (stages 2–9) and the reboiler heat transfer coefficient of the stage-by-stage balance model such that the nitrogen and oxygen product purities of the Aspen model at the nominal steady state were matched. Fig. 3 provides a comparison of steady-state oxygen and argon composition profiles obtained from the Aspen simulator, the full-order stage-by-stage model (FOM) and reduced order models with 15, 9 and 5 compartments (Comp15, Comp9 and Comp5) at the nominal steady state. The composition profiles are plotted in physical coordinates and natural log transformed coordinates to facilitate comparison of the various models. The stage-by-stage model provides good agreement with the Aspen simulator over a very wide range of compositions. Although not illustrated here, the inclusion of energy balances and non-ideal vapor–liquid equilibrium relations in the stage-by-stage model was necessary to achieve satisfactory results. Sharp discontinuities occur at the stages where a feed stream is introduced, while relatively flat composition profiles are observed at the locations of the liquid distributors. As predicted by singular perturbation theory (Levine & Rouchon, 1991), each compartmental model provides perfect steady-state agreement with the stage-by-stage model. Any further improvements in the compartmental models would require the derivation of a more accurate stage-by-stage model.

The motivation for including more compartments in the reduced order model is to improve dynamic prediction accuracy. Fig. 4 shows the evolution of the nitrogen and oxygen product compositions for a 25% decrease in liquid air flow rate at $t = 1$ h. The stage-by-stage model yields a steady-state gain for the nitrogen product composition that is about 65% larger than the value produced by Aspen. By contrast, the steady-state gain for the oxygen product composition is predicted very accurately. Good dynamic agreement between the Aspen simulator and the stage-by-stage model responses is obtained for this step change. The reduced order models yield slower nitrogen composition responses than the stage-by-stage model, while very good agreement is obtained for the oxygen product purity except for the 5 compartment model.

The evolution of the oxygen and argon composition profiles for the −25% change in the liquid air flow rate is shown in Fig. 5. The profiles obtained from the Aspen simulator and the 15 compartment model are plotted at three time instants. While the compartmental model is able to track the two product compositions (see Fig. 4), the transient composition profiles are predicted less accurately. Large deviations between the Aspen and compartmental profiles are observed at the initial steady state ($t = 1$ h) and immediately af-

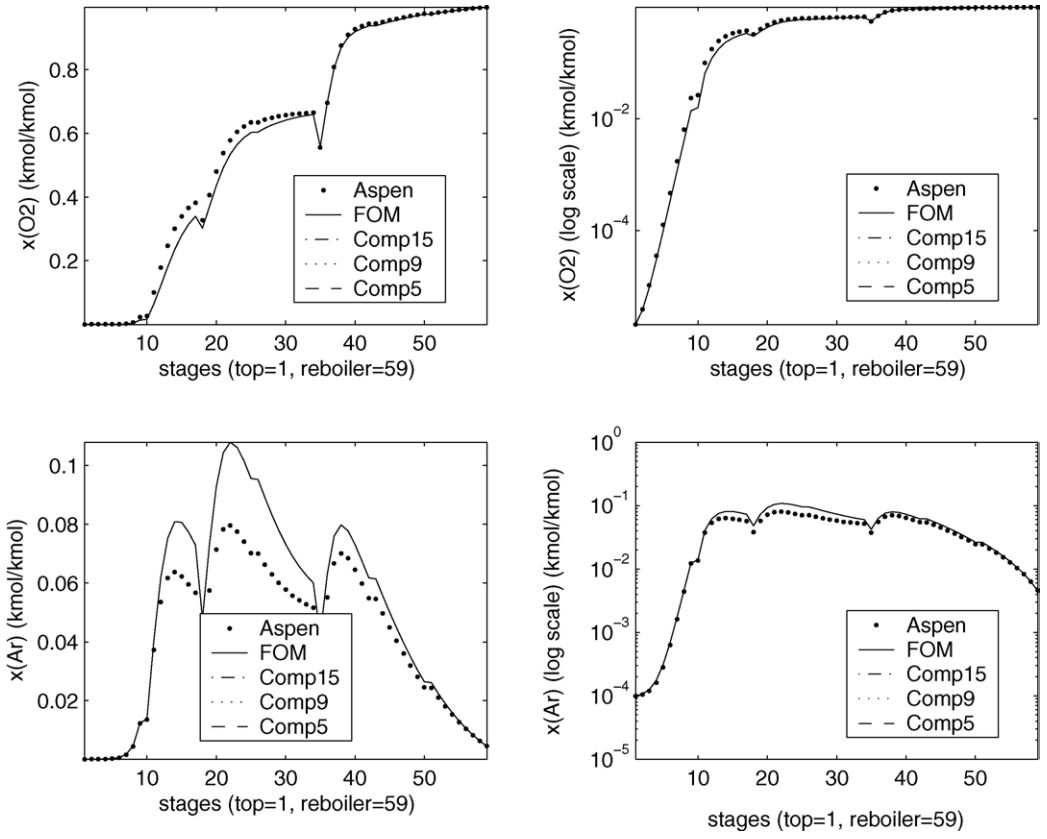

Fig. 3. Steady-state oxygen and argon composition profiles for the nominal operating point. The various models are denoted as: Aspen, Aspen simulator; FOM, full-order stage-by-stage model; Comp15, 15 compartment model; Comp9, 9 compartment model; Comp5, 5 compartment model.

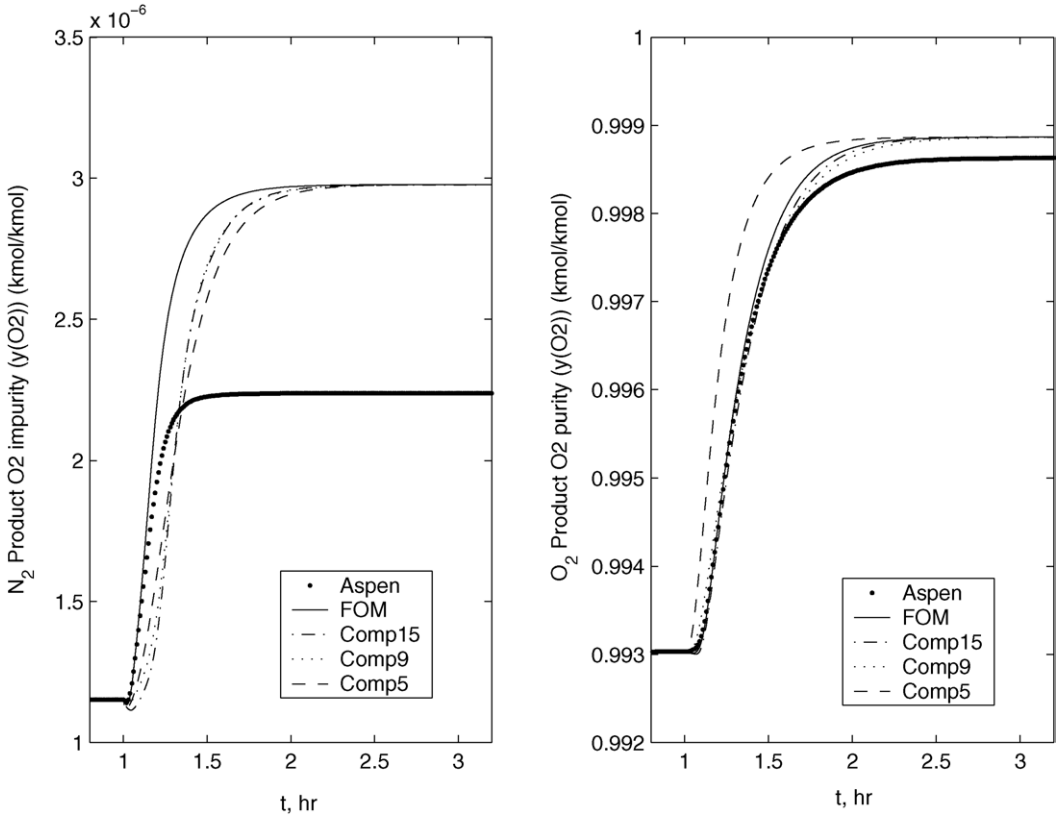

Fig. 4. Dynamic nitrogen and oxygen product composition responses for a −25% change in the liquid air feed flow rate at $t = 1$ h.

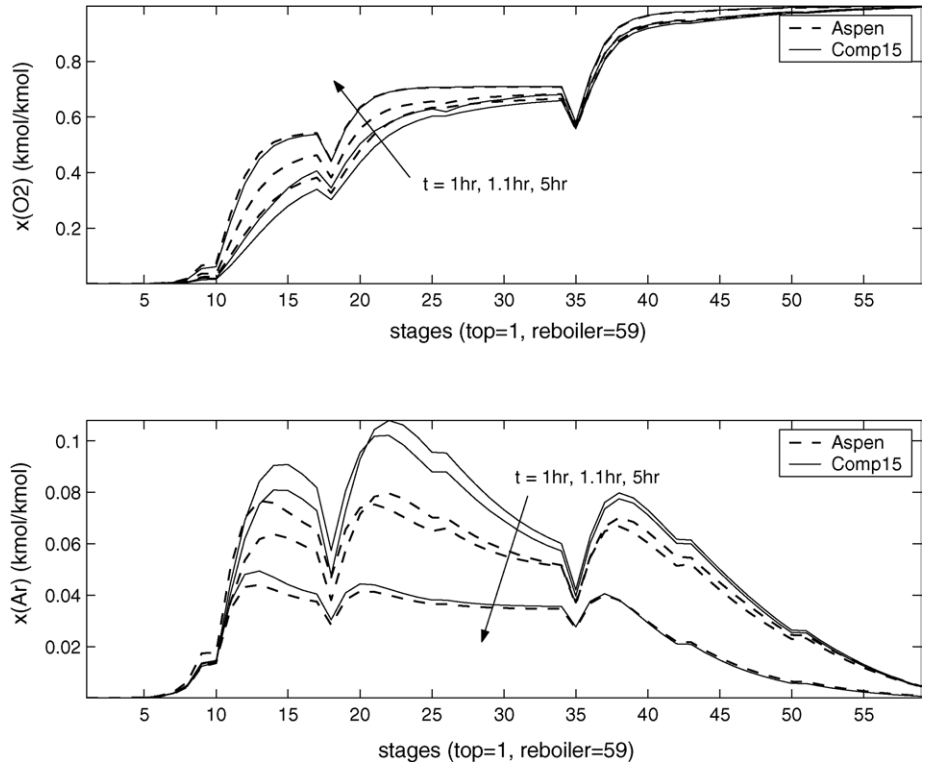

Fig. 5. Dynamic oxygen and argon composition profiles for a −25% change in the liquid air feed flow rate at $t = 1$ h.

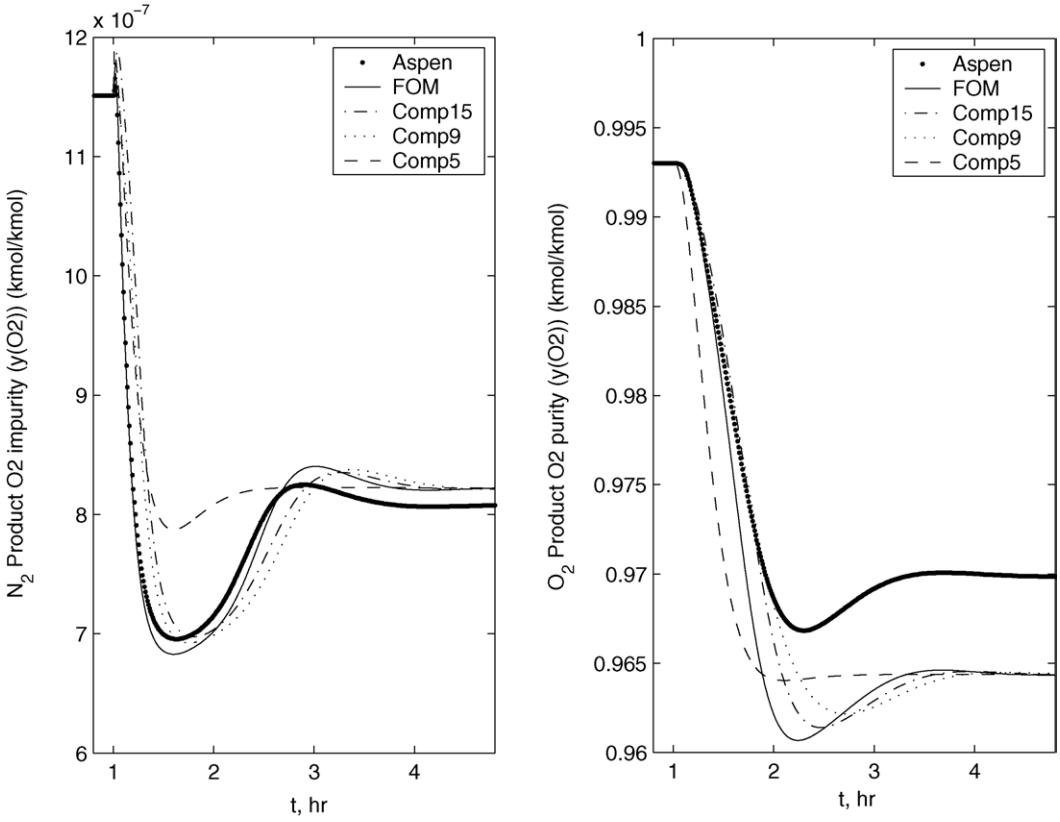

Fig. 6. Dynamic nitrogen and oxygen product composition responses for a +25% change in the liquid air feed flow rate at $t = 1$ h.

ter the step change ($t = 1.1$ h) in the upper portion of the column. By contrast the compartmental model yields very accurate predictions of the final steady-state composition profiles ($t = 5$ h).

Fig. 6 shows the dynamic responses of the nitrogen and oxygen products for a +25% change in the liquid air feed stream at $t = 1$ h. The stage-by-stage model generates very accurate predictions of the Aspen nitrogen product purity dynamics and steady-state gain. Although the stage-by-stage model produces a slightly larger steady-state gain than the Aspen simulator for the oxygen product composition, the undershooting dynamics caused by the reboiler level controller are faithfully captured. The 15 compartment model provides the best agreement with the stage-by-stage model for both product compositions. The 9 compartment model produces larger errors in the oxygen product purity dynamics, while the 5 compartment model exhibits large deviations for both compositions responses due to the lumping of equilibrium stages and liquid distributors into the same compartments. These results suggest that 15 compartments are needed to accurately track the product composition dynamics of the stage-by-stage model.

Oxygen and argon composition profile dynamics produced by the Aspen simulator and the 15 compartment model for the +25% change in liquid air flow rate are shown in Fig. 7. Despite generating satisfactory predictions of the product composition responses (see Fig. 6), the compartmental model exhibits large errors in the composition profile dynamics. Particularly large deviations are observed immediately after the step change ($t = 1.1$ h) and at the final steady state ($t = 5$ h). These results are partially attributable to the adjustment of the stage-by-stage model to reproduce the Aspen product compositions at the initial steady state. In effect, the stage-by-stage model has been tuned to predict the product compositions at the expense of the composition profiles.

The previous simulation results were generated by selecting the sensitive stage of each multistage compartment to be located in the center of the compartment. Fig. 8 shows the effect of the sensitive stage locations on product composition responses obtained with 15 compartments for a +25% change in the liquid air feed flow rate at $t = 1$ h. The most accurate predictions of Aspen composition dynamics are produced when the sensitive stages are located in the center of the compartments as in the previous simulation tests. However, very similar predictions are obtained when the sensitive stages are located in either the top or bottom of the compartments. The main advantages of center placement are less inverse response (see insets) and more accurate dynamic tracking of the oxygen product purity response. These results support the claim that the locations of the sensitive stages have little effect on reduced order model accuracy (Khowinij et al., in press; Levine & Rouchon, 1991).

The final set of simulation results were generated without adjusting the stage-by-stage model to reproduce the As-

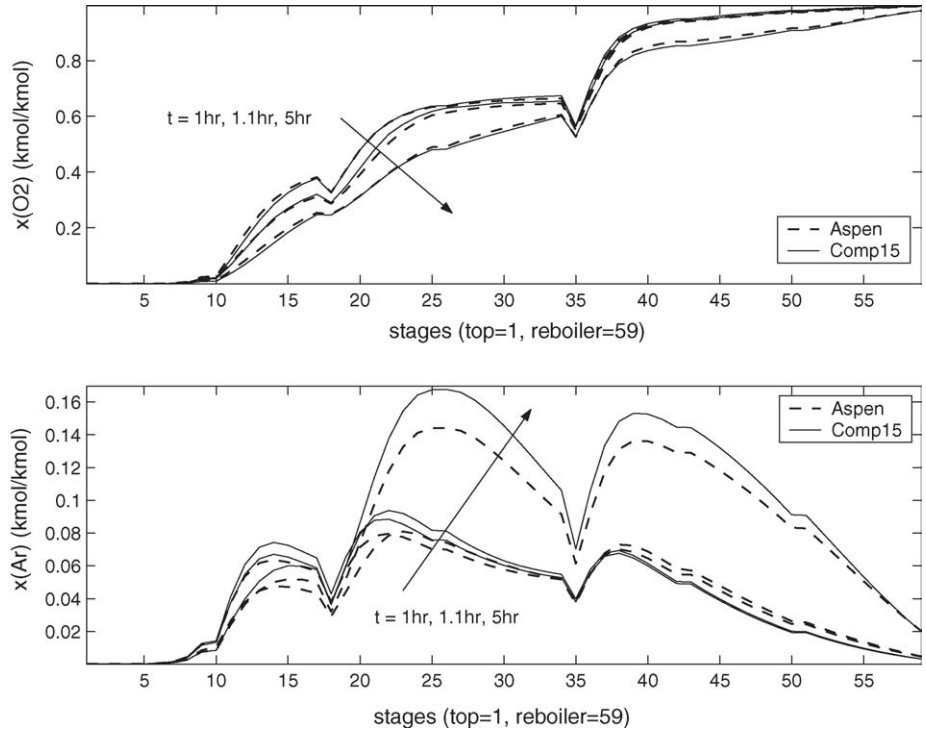

Fig. 7. Dynamic oxygen and argon composition profiles for a +25% change in the liquid air feed flow rate at $t = 1$ h.

pen product compositions at the initial steady state. Fig. 9 shows nitrogen and oxygen product composition responses for a +25% change in the liquid air feed flow rate at $t = 1$ h when the sensitive stage of each multistage compartment is located in the center of the compartment. As compared to the case where the stage-by-stage model was fine tuned to fit the initial Aspen product compositions (see Fig. 6), the stage-by-stage model produces a much smaller steady-state gain for

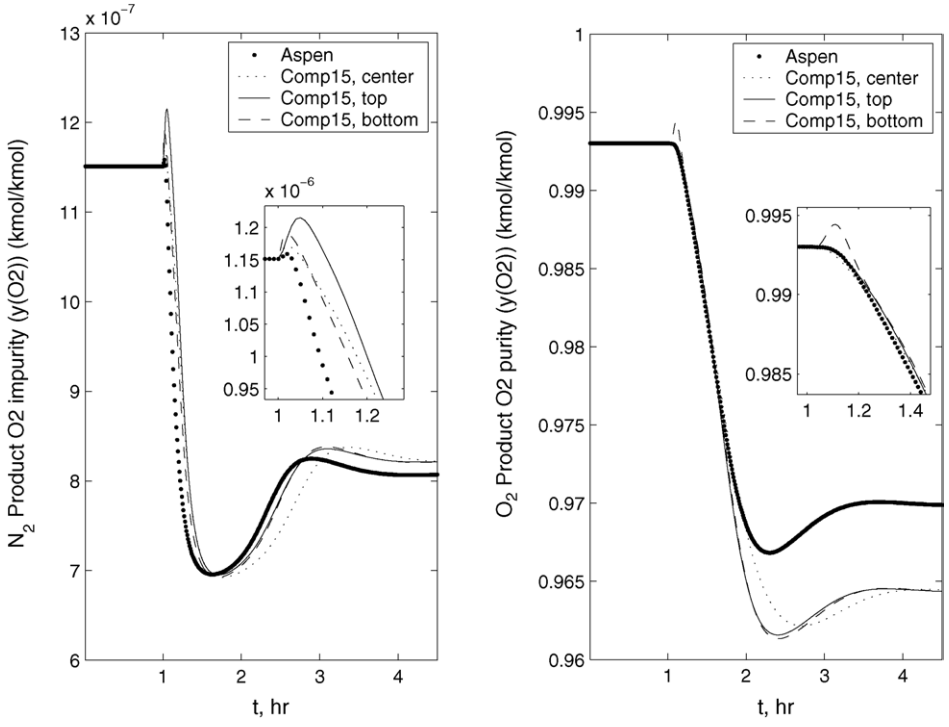

Fig. 8. The effect of sensitive stage locations on dynamic nitrogen and oxygen product composition responses of 15 compartment models for a +25% change in the liquid air feed flow rate at $t = 1$ h.

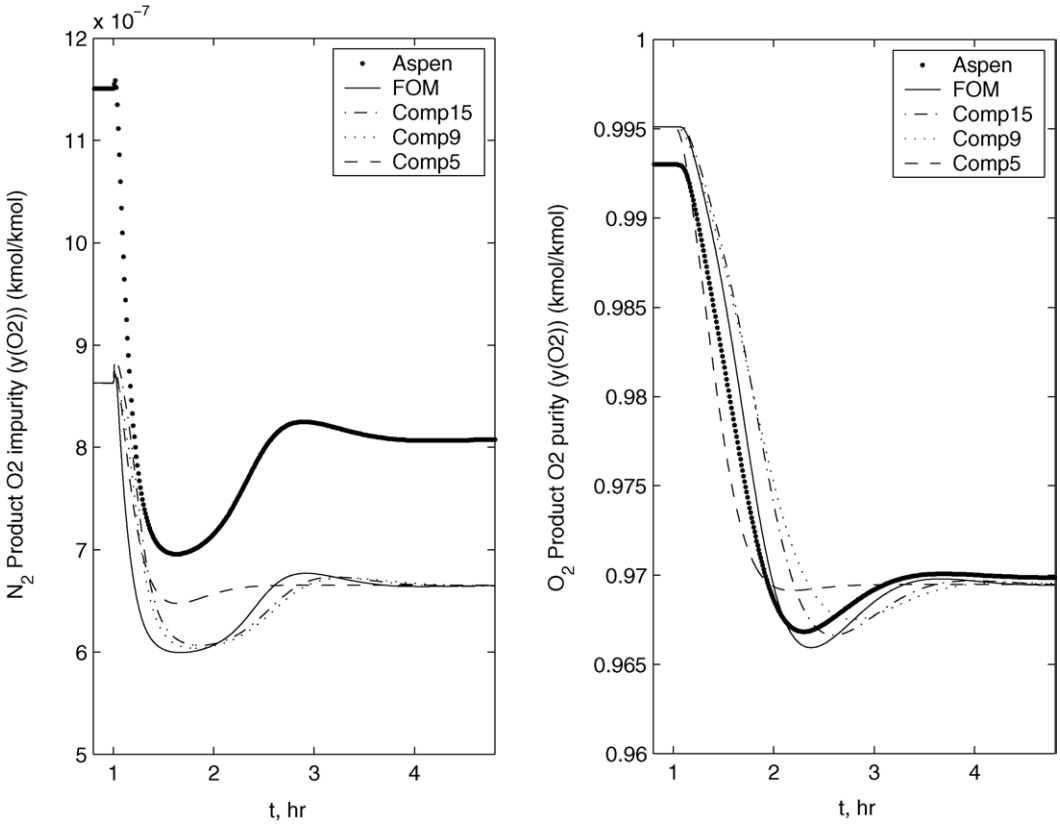

Fig. 9. Dynamic nitrogen and oxygen product composition responses for a +25% change in the liquid air feed flow rate at $t = 1$ h without matching the initial steady-state product compositions of the Aspen and full-order models.

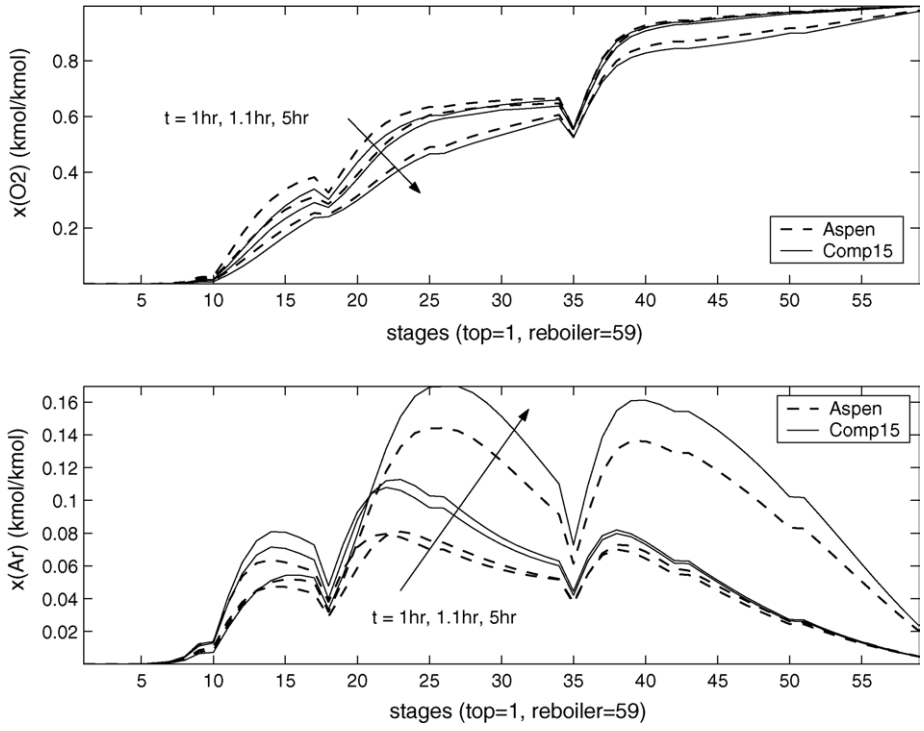

Fig. 10. Dynamic oxygen and argon composition profiles for a +25% change in the liquid air feed flow rate at $t = 1$ h without matching the initial steady-state product compositions of the Aspen and full-order models.

the nitrogen product purity but a more accurate steady-state gain for the oxygen product purity using the Aspen model as the basis for comparison. A comparison of the nitrogen and oxygen composition profiles in Figs. 7 (adjusted model) and 10 (unadjusted model) generated with 15 compartments shows that more accurate prediction of Aspen composition profiles is obtained with the unadjusted model. This result provides further support for the claim that the adjusted model has been tuned to reproduce Aspen product compositions at the expense of poor composition profile predictions.

As shown in Table 2, the compartmental modeling strategy allows a large reduction in the number of differential variables as compared to the stage-by-stage model. In our previous work on nitrogen purification columns, we observed that a seven compartment model produced a five-fold decrease in simulation time as compared to the stage-by-stage model from which it was derived (Khowinij et al., in press). For the upper column studied in this paper, we found that compartmental models produced roughly the same computation times as the stage-by-stage model regardless of the number of compartments used. We believe that these seemingly incongruous results are attributable to the more complex algebraic equations present in the upper column compartment models combined with the inefficiency of the MATLAB DAE solver. Although not investigated here, we conjecture that the computational efficiency of the compartmental models relative to the stage-by-stage model could be improved by using a more sophisticated DAE solver such as DASSL (Brenan, Campbell, & Petzold, 1989). However, our results suggest that the compartmental modeling approach can expected to yield significant reductions in open-loop simulation time when the stage-by-stage model contains relatively simple algebraic equations obtained under the assumptions of constant equimolar overflow and ideal vapor–liquid equilibrium. This conclusion would need to be verified with additional simulation studies for other types of distillation columns. As discussed earlier, we believe that the primary advantage of the compartmental modeling approach is that the resulting DAE models can be effectively incorporated into NMPC controllers by utilizing customized solution methods that exploit the sparse model structure (Leineweber, 1998).

5. Summary and conclusions

Reduced order compartmental models were derived from a detailed stage-by-stage balance model of a high purity distillation column used for cryogenic air separation. The stage-by-stage model with dynamic component balances, steady-state energy balances and non-ideal liquid phase relations was shown to provide good agreement with a rigorous Aspen simulator over a range of production rates. Reduced order models with different numbers of compartments were derived directly from the stage-by-stage model through the application of singular perturbation theory. The order reduction method produced compartmental models with relatively few differential variables but a large number of algebraic variables. The compartmental models were shown to yield good steady-state and dynamic agreement with the stage-by-stage model as long as a sufficiently large number of compartments were used. The compartmental models did not produce a significant decrease in open-loop simulation time despite the large reduction in the number of differential variables. We conjecture that improved compartmental model performance relative to the stage-by-stage model can be achieved with a more advanced differential–algebraic equation (DAE) solver than those currently available in Matlab. More importantly, the order reduction method produces sparse DAE models that can be exploited in simultaneous solution methods developed for nonlinear model predictive control. This direction is the focus of our future research.

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