New Formulation of Optimization-Based Simulation of a Pressure Swing Adsorption Process: Hybrid Dynamic Optimization

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Introduction

A typical Skarstrom pressure swing adsorption (PSA) process consists of four basic operations, that is, pressurization, high-pressure adsorption, blow down and low-pressure purge. This sequence of operations is carried out in a series of fixed-beds in the same way but shifted in time. PSA processes, are, therefore, transient and cyclic in nature.

The PSA process models are described by sets of partial differential algebraic equations (PDAEs). They are mainly constituted by conservation equations and models for the equation of state, equilibrium and thermodynamic and transport properties.

The simulation of PSA processes consists in the determination of the cyclic steady-state (CSS). This is traditionally performed by means of successive substitution or Newton-type methods. These methods are well known in the literature and are extensively studied in order to improve their robustness and convergence rate. However, alternative and interesting approaches have recently emerged and are based on the use of optimization methods for the determination of CSS.[1–5] Their basic principle consists in the formulation of the simulation problem as a dynamic optimization problem where the performance index is defined as the sum of square differences between the state variables (gas-phase concentrations, solid-phase concentrations and temperature) obtained at the beginning and at the end of a cycle. The decision variables are the initial conditions (state variables at the beginning of a cycle), and the constraints are given by the process model, where the method of lines is used for spatial discretization and converts the PDAEs to differential-algebraic equations (DAEs). A gradient-based nonlinear programming (NLP) solver is then used to determine the initial state vector which minimizes the cyclic steady-state condition.

The main differences between the previous works on optimization-based simulation of PSA processes are the method of discretization of PDAEs (both single discretization and complete discretization), and the method used to compute the gradients for the NLP solver.

In Nilchan and Pantelides[1] and Ko et al.[3] the complete discretization approach, that is, space and time are discretized, is used and the resulting NLP problem is solved within the sensitivity-based code gPROMS. For simple models this approach is efficient, but for complicated models the complete discretization causes error accumulation and may lead to solver failure.

In Ko et al.[3,4] and Jiang et al.[2] the single discretization approach where only the space is discretized is used, and the method of lines used is the centered finite differences in Ko et al.,[3,4] whereas the finite volumes method is used in Jiang et al.[2] In the three works, the sensitivity method is used and the resulting optimization problem is solved within gPROMS. It is important to notice that the spatial discretization method involves quite a large number of decision variables and the use of sensitivity method for gradients computation results in a huge DAE system, and, consequently, in big computation times. On the other hand, since the number of constraints involved in an optimization-based determination of CSS is small, the adjoint system method becomes more interesting.

In Latifi et al.[5] the spatial discretization approach used is based on orthogonal collocation method, and the adjoint

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system method is used for gradients computation. They showed that the adjoint method has the fastest convergence rate compared to finite differences method and both numerical and analytical sensitivity methods. However, orthogonal collocation methods often introduce physically unrealistic oscillations near steep adsorption fronts, and lead to solutions with negative values for positive variables (e.g., mole fractions). The finite volumes method allows us to determine accurate solutions which in addition preserve the mass and energy balance in the spatial direction (conservative method).

This work focuses on the simulation of PSA processes, i.e., determination of the cyclic steady-state (CSS) using a new formulation, based on hybrid dynamic optimization approach, which relies on high-quality system models. The class of hybrid processes considered in this article refers to processes that require the use of different process models, each is valid in a given functioning time domain. Transitions between domains take place at different time instants (events). Like reverse flow reactors (Eingenberger and Nieken[6]), small-size wastewater treatment plants (Chachuat et al.[7]), emulsion polymerization processes (Salhi et al.[8]), etc., pressure swing adsorption (PSA) processes are typical examples of hybrid processes. In general, this class of hybrid processes is described by time-dependent, nonlinear dynamic models that exhibit model switching as a sequence of both time and state-dependent events.

The objectives of this article are: (1) to derive the necessary conditions of optimality for a hybrid system using the adjoint method, and to deduce the gradients required by the NLP solver; (2) to formulate the simulation of PSA processes as a hybrid dynamic optimization problem, where the model PDAEs are converted to DAEs using the finite volumes method, and (3) to present some new results obtained in the case of a nonisothermal PSA process.

PSA process model

The hybrid system considered here is a PSA process for solvent vapor recovery (SVR) with charcoal as adsorbent used for separation of a mixture of C_6H_6/N_2. The process used consists of four steps: pressurization, high-pressure adsorption, blow down and low-pressure purge. The process model is similar to those in Liu and Ritter[9] and Liu et al.[10] and is based on the following assumptions: a 1-D (one-dimensional) model is considered, the system is nonisothermal, the axial dispersion and pressure drop are negligible, the gas phase is ideal, the carrier gas is assumed to be inert, the mass-transfer rate is described by LDF (linear driving force) model.

The resulting model is constituted by the following equations:

Adsorbate mass balance in the fluid phase

$$\frac{\partial y}{\partial t}+u\frac{\partial y}{\partial z}+(1-y)\frac{RT}{P}\frac{1-\varepsilon}{\varepsilon}\rho_s\frac{\partial q}{\partial t}=0 \tag{1}$$

Overall mass balance

$$\frac{\partial u}{\partial z}+\frac{1}{P}\frac{\partial P}{\partial t}-\frac{1}{T}\frac{\partial T}{\partial t}-\frac{u}{T}\frac{\partial T}{\partial z}+\frac{1-\varepsilon}{\varepsilon}\frac{RT}{P}\rho_s\frac{\partial q}{\partial t}=0 \tag{2}$$

Table 1. Initial and Boundary Conditions for the Four Basic Steps

Step I (compression)	Step II (adsorption)
$P=f(t)$ $t=0,\ \forall z: y=y_{IV},\ q=q_{IV},\ T=T_{IV}$ $z=0,\ \forall t: y=y_{feed},\ T=T_{feed}$ $z=L,\ \forall t: u=0$	$P=P_H$ $t=0,\ \forall z: y=y_I,\ q=q_I,\ T=T_I$ $z=0,\ \forall t: y=y_{feed},\ T=T_{feed},\ u=u_H$
Step III (blowdown)	**Step IV (purge)**
$P=f(t)$ $t=0,\ \forall z: y=y_{II},\ q=q_{II},\ T=T_{II}$ $z=L,\ \forall t: \frac{\partial y}{\partial z}=0,\ T=T_{ads},\ u=0$	$P=P_L$ $t=0,\ \forall z: y=y_{III},\ q=q_{III},\ T=T_{III}$ $z=L,\ \forall t: y=y_{ads}\frac{P_L}{P_H},$ $T=T_{ads},\ u=u_L$

LDF model

$$\frac{\partial q}{\partial t}=k_a(q^*-q) \tag{3}$$

Energy balance

$$\left(\varepsilon\rho_g C_{pg}+(1-\varepsilon)\rho_s C_{ps}\right)\frac{\partial T}{\partial t}+\varepsilon\rho_g C_{pg}u\frac{\partial T}{\partial z}+(1-\varepsilon)\rho_s\Delta H\frac{\partial q}{\partial t}+\frac{2h}{r_b}(T-T_o)=0 \tag{4}$$

Equilibrium relationship for benzene

$$q*=\frac{q_s bPy}{1+bPy}$$

where

$$b=\frac{b_0}{R\sqrt{T}}\exp\left(-\frac{\Delta H}{RT}\right) \tag{5}$$

It is important to notice that Eqs. 1–5 are the same for the four basic steps. The associated initial and boundary conditions are however different and are given in Table 1.

Spatial discretization

The method of lines methodology (Schiesser[11]) is used to convert the system of PDAEs to a system of differential algebraic equations (DAEs). In this work, the finite volumes method (Webley and He[12]) is used. It is particularly suitable for modeling hyperbolic conservation laws given its inherent conservative properties (Leonard[13]).

The discretized model of the PSA process under consideration may be written in the following hybrid form (Galàn et al.[14] and Ruban[15])

$$\dot{x}^{(k)}=f^{(k)}\left(x^{(k)},y^{(k)},p\right) \tag{6}$$

$$0=g^{(k)}\left(x^{(k)},y^{(k)},p\right) \tag{7}$$

The transitions conditions are

$$L_j^{(k)}=t-t_f^{(k)}=0 \tag{8}$$

The transitions functions associated with the transition conditions are

$$x^{(k+1)}\left(t_0^{(k+1)}\right) = x^{(k)}\left(t_f^{(k)}\right) (\text{i.e. } \Delta_j^{(k)} = 0)$$

for

$$k = 1, 2, 3, 4 \quad ; \quad j = k + 1. \tag{9}$$

The superscript k refers to a basic step of the Skarstrom PSA process considered.

A special case of the transition functions is given by the initial conditions for the compression step as

$$x^{(1)}(t_0^{(1)}) - x_0^{(1)} = 0 \tag{10}$$

where the differential state vector is given by

$$x^{(k),T} = \left(\bar{y}_1^{(k)}, \bar{y}_2^{(k)}, ..., \bar{y}_N^{(k)}, \bar{q}_1^{(k)}, \bar{q}_2^{(k)}, ..., \bar{q}_N^{(k)}, \bar{T}_1^{(k)}, \bar{T}_2^{(k)}, ..., \bar{T}_N^{(k)}\right) \tag{11}$$

and the algebraic state vector by

$$y^{(k),T} = \left(\bar{u}_1^{(k)}, \bar{u}_2^{(k)}, ..., \bar{u}_N^{(k)}\right) \tag{12}$$

The initial state is given by

$$x_0^{(1),T} = \left(\bar{y}_1^{(1),0}, \bar{y}_2^{(1),0}, ..., \bar{y}_N^{(1),0}, \bar{q}_1^{(1),0}, \bar{q}_2^{(1),0}, ..., \bar{q}_N^{(1),0}, \bar{T}_1^{(1),0}, \bar{T}_2^{(1),0}, ..., \bar{T}_N^{(1),0}\right) \tag{13}$$

On the other hand, from the basic steps durations, i.e., τ_{comp}, τ_{ads}, τ_{blow} and τ_{des}, the transitions times are given as

$$t_f^{(1)} = \tau_{comp},\ t_f^{(2)} = \tau_{comp} + \tau_{ads},\ t_f^{(3)} = \tau_{comp} + \tau_{ads} + \tau_{blow},\ t_f^{(4)} = \tau_{comp} + \tau_{ads} + \tau_{blow} + \tau_{des} \tag{14}$$

The last transition time is also the PSA cycle duration.

Optimization-based formulation of the simulation problem

The objective of the simulation problem is to determine the cyclic steady-sate, i.e., the state vector at the start of the cycle must be equal to the state vector at the end of the cycle. The classical formulation of this objective is expressed as

$$x^{(1)}\left(t_0^{(1)}\right) = x^{(4)}\left(t_f^{(4)}\right) \tag{15}$$

Different methods have been developed to solve Eq. 15, including fixed-point iteration approach, quasi-Newton and Newton methods.[1,17]

The optimization-based formulation developed in this work consists in treating the simulation problem as a single-dynamic optimization problem where the performance index is the CSS condition, the decision variables are the state variables at the start of the cycle, and the constraints are given by the hybrid model equations of the process with associated initial conditions, transition conditions and transition functions.

It is noteworthy that the decision variables are the initial state vector. The time-independent vector of parameters, is, therefore, defined as

$$p = x^{(1)}\left(t_0^{(1)}\right) \tag{16}$$

The mathematical formulation of the optimization-based simulation is given by

$$\underset{p}{Min}\left\{J = \frac{1}{2}e^T e\right\}$$

where

$$e = x^{(1)}\left(t_0^{(1)}\right) - x^{(4)}\left(t_f^{(4)}\right) \tag{17}$$

subject to constraints, Eqs. 6–10.

Computational method

The computational method used consists in estimating the initial values of decision variables which are used in process model integration. The performance index and the gradients of both performance index and constraints with respect to decision variables are computed and provided to a gradient-based NLP solver, which in turn estimates a new vector of decision variables. The process is repeated until convergence where the optimal values of decision variables are obtained.

It is important to notice that in any gradient-based optimization solver, the convergence and its rate depend strongly on the accuracy of gradients computation. The computation of gradients by means of sensitivity method is best suited for optimization problems involving a large number of constraints and a small number of decision variables. The adjoint system method is, however, more efficient for problems with a large number of decision variables and few constraints.

In this work, finite differences and adjoint system methods are used and compared. They are based on the following general definition of the performance index

$$J = G\left[x\left(t_f\right), p\right] + \int_0^{t_f} F[x(t), p]dt \tag{18}$$

Finite differences method

The approximation of the gradient of the performance index J, with respect to a parameter p_i, by means of (centered) finite differences method consists in perturbing J, with a finite amount Δp_i of p_i as follows

$$\frac{\partial J}{\partial p_i} \approx \frac{J(p_i + \Delta p_i) - J(p_i - \Delta p_i)}{2\Delta p_i}$$

where typically

$$\frac{\Delta p_i}{p_i} = 1\% \tag{19}$$

Table 2. Physical Data used in the Model

Parameter	Value	Unit	Parameter	Value	Unit
r_b	0.0135	m	t_c	20	min
L	0.29	m	τ	8	min
ε	0.43	-	T_o	293	K
ρ_s	480	kg/m^3	ρ_g	1.308	kg/m^3
C_{ps}	1.05	$kJ/kg\ K$	C_{pg}	1.006	kJ/kgK
y_F	0.05	-	ΔH	-43.5	kJ/mol
V_F	0.0005	m^3 $(STP)/min$	k_a	0.086	s^{-1}
T_F	293	K	q_s	4.4	mol/kg
P_H	152	kPa	h	0.0314	$kJ/m^2s\ K$
P_L	7.6	kPa	b_o	3.88×10^{-8}	$m^3/mol.K^{0.5}$
γ	1.5	-			

Adjoint system method

The gradients of the performance index J, with respect to a parameter p_i, by means of adjoint system method, are derived based on the works of Bryson and Ho[18] and Ruban[15] and can be written as

$$\frac{\partial J}{\partial p_i} = \lambda^{(1)}\left(t_0^{(1)}\right)\frac{\partial x^{(1)}\left(t_0^{(1)}\right)}{\partial p_i} + \frac{\partial G}{\partial p_i} \tag{20}$$

The Hamiltonian function in each basic step is given by

$$H^{(k)} = F(x,p) + \lambda^{(k)^T} f^{(k)} \tag{21}$$

where the corresponding vector of adjoint variables is defined as

$$\dot{\lambda}^{(k)} = -\frac{\partial H^{(k)}}{\partial x^{(k)}} \tag{22}$$

with the terminal conditions

$$\lambda^{(4)}(t_f^{(4)}) = \left.\frac{\partial G}{\partial x}\right|_{t=t_f^{(4)}} = x^{(4)}(t_f^{(4)}) - p \tag{23}$$

k = 1, 2, 3, 4.

The computational algorithm for the adjoint system method is as follows:

(a) Estimation of initial values of decision variables p;

(b) integration of model Eqs. 6–10;

(c) backward integration of the system of adjoint Eqs. 22–23;

(d) computation of performance index (Eq. 18), and gradients (Eq. 20);

(e) estimation of new vector of decision variables p by NLP solver and repetition of the process from step (b) until the convergence criteria is satisfied.

Results

Data used

The time-varying pressure usually used is a quadratic function of time as suggested in Kapoor and Yang,[19] but for simplicity reasons and without loss of accuracy, the pressure profiles used here in the compression and blow down steps are considered to be linear and are given as

$$P(t) = P_H + (P_L - P_H)\left(-\frac{t}{\tau_{\text{comp}}} + 1\right)$$

and

$$P(t) = P_L + (P_H - P_L)\left(-\frac{t}{\tau_{\text{blow}}} + 1\right) \tag{24}$$

On the other hand, we assumed that the durations of adsorption and purge steps are equal ($\tau_{\text{ads}} = \tau_{\text{des}} = \tau$). In the same way, the durations of pressurization and blow down steps are the same and equal to one-fourth of the duration of adsorption and purge steps ($\tau_{\text{comp}} = \tau_{\text{blow}} = 0.25\tau$). The number of finite volume elements N is 40 and the rest of physical data are given in Table 2.

Results and Discussion

Simulations are carried out on a 3.40 GHz Intel Pentium 4 computer. The NLP solver used is NLPQL by Schittkowski[20] and DASSL code is used as the integrator (Brenan et al.[21]).

A fairly starting point of decision variables is necessary for convergence to the optimal values. The determination of initial values is not an easy task in optimization problems. However, for the PSA process considered, Liu and Ritter[9] carried out a study where the influence of the most influencing parameters is investigated and the initial values of decision variables are deduced from their results. With these initial values the SQP solver takes less than 10 iterations to converge to the optimal values.

Table 3 presents the optimal values of the performance index, i.e., the CSS condition (Eq. 17), and their corresponding CPU times for the two methods of gradients computation. It can be seen that adjoint system method has not only faster convergence rate, but it also leads to a more accurate value of the CSS condition. However, it is noteworthy that the implementation of the adjoint system method requires the forward integration of the process model first followed

Table 3. PSA optimization results

	Finite difference method	Adjoint system method
No. of iterations	30	6
CPU time (s)	1097	7
Performance index	2.08×10^{-3}	1.47×10^{-4}

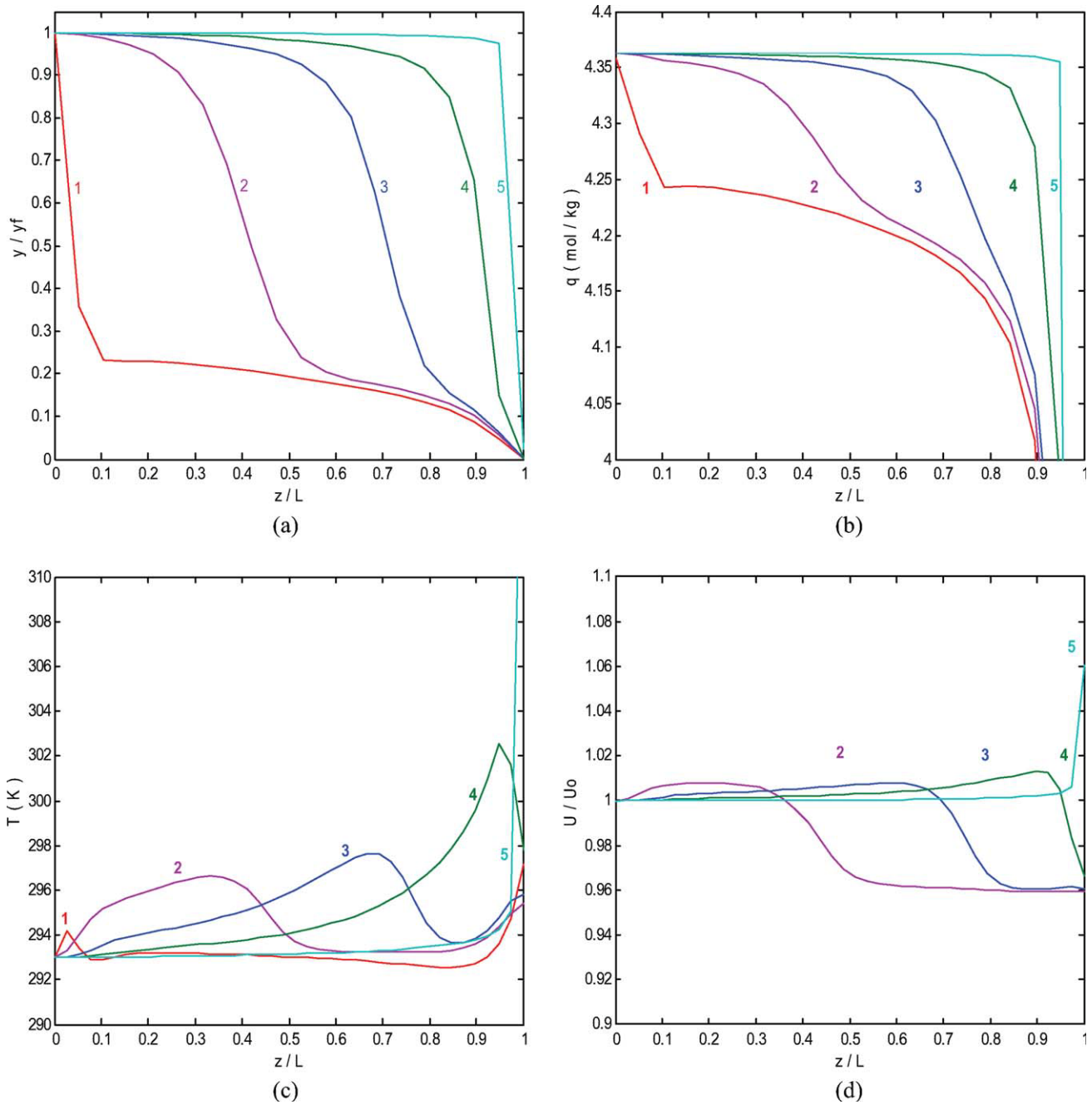

Figure 1. Fluid-phase concentration (a), solid-phase concentration (b), temperature (c), and velocity (d) profiles after adsorption step at CSS vs. dimensionless bed length. Curves 1, 2, 3, 4 and 5 correspond to profiles at τ_{ads} = 0, 2, 4, 6 and 8 min, respectively.

[Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

by the backward integration of the adjoint system where the state variables are needed. The latter are stored during the forward integration and recovered for the backward integration. For optimization problems where small to moderate number of variables are involved, the storage and recovery process is not expensive. This is the case for the PSA process considered here where only 40 finite volumes are used for spatial discretization, hence, leading to 120 decision variables. However, if we increase the number of finite volumes in order to improve the accuracy of optimal profiles, the resulting problem will involve a quite large number of decision variables. The storage and recovery process in this case is potentially very expensive and constitutes one of the limits of the adjoint system method. The remedy to overcome this limit would probably be the parallelization of the adjoint system integration as it was done for sensitivity calculation in Jiang et al.[22].

On the other hand, since the number of decision variables involved in the optimization problem considered here is not high the SQP solver used requires only few iterations.

However, when it is used for large-scale problems some difficulties may appear since the estimation of the Hessian matrix will become dense due to the BFGS update formula used by the solver. Reduced gradient methods are best suited for problems involving a significant number of decision variables and constraints (Murtagh and Saunders[23,24]).

In order to compare the results obtained with those available in the literature, Figure 1 presents the same variables in the same format with the same system of axis as in Liu and Ritter.[9] It can be seen that they are in very good agreement with those obtained in Liu and Ritter[9] where the process model is discretized by finite differences method and solved by Newton-Raphson procedure. However, the computational time in Liu et al.[10] is huge compared to the adjoint system method developed here. Moreover, when the gradients are computed by means of sensitivity method and used either in the SQP solver or in a Newton-based approach, it was shown in Latifi et al.[5] that the adjoint system method is faster and more accurate.

It is worth noticing that the accuracy of the results obtained increases with the number of finite volumes used in the spatial discretization. However, the problem encountered here is that when the number of finite volumes is increased, the computational time becomes large and in most cases the solver fails to converge. Nevertheless, the results show that 40 finite volumes are quite enough to compute the same solution as in Liu and Ritter[9].

To overcome the problems induced by large optimization problems, an interesting approach was proposed in Ko et al.[3,4] where the initial spatial profiles of variables y, q and T are parameterized by means of functions of only four unknown parameters. The ranges of these parameters in the optimization are determined from the profiles of $y(t_f^{(4)})$, $q(t_f^{(4)})$ and $T(t_f^{(4)})$ resulting from successive substitution from the first cycle to CSS. The final optimization problem involves only 12 (4×3) parameters instead of 120 (40×3) parameters in the method developed in this work. The resulting number of decision variables is then quite reduced and the convergence of the optimization process is consequently improved. The optimal values of the 12 parameters are then determined from the optimization to the CSS conditions (Eq. 17). Although this approach is interesting, it may, however, not be as accurate as the approach developed in this work. This is due to the fact that in this work, the initial profiles can be specified freely at each node point and the CSS conditions are satisfied at each node point, whereas in Ko et al.[3,4] the shapes of the initial profiles are *a priori* captured using parameterization functions which may not be enough accurate to represent the freely computed profiles at CSS.

Conclusions

Optimization-based simulation of a typical Skarstrom PSA process is carried out by simultaneous treatment of simulation and optimization as a single problem. The optimization problem is formulated as a hybrid dynamic optimization where four modes (corresponding to the four steps of the PSA process) are involved. Each mode is characterized by a system of DAEs, transition conditions and transition functions. The necessary conditions of optimality (NCO) for a hybrid system are then derived using the adjoint system method. The resulting equations of NCO are used for computation of the gradients of the performance index and constraints with respect to decision variables. These gradients are then provided to the NLP solver (NLPQL (Schittkowski[20])) used for computation of optimal values of decision variables. The case study is a PSA process used for separation of a mixture of C_6H_6/N_2, and is described in detail in Liu and Ritter[9] and Liu et al.[10] The results obtained are in a good agreement with those reported in Liu and Ritter,[9] hence, showing that the optimization-based simulation developed is quite efficient. On the other hand the finite differences method is also used to compute the gradients and the results obtained show clearly that the adjoint system method leads to better values of the performance index and convergence rate. However, improvements are still to be brought to the method in order to increase the spatial accuracy (i.e., increase the number of finite volumes), and to correctly handle systems of high dimension.

Notation

b_o = isotherm parameter, $m^3/mol.K^{0.5}$
C_{pg} = gas-phase heat capacity, J/kgK
C_{ps} = adsorbent heat capacity, J/kg K
h = overall heat-transfer coefficient, J/m^2 s K
k_a = mass-transfer coefficient s^{-1}
L = bed length, m
P_H = high pressure, Pa
P_L = low pressure, Pa
q = amount adsorbed, mol/kg
q^* = equilibrium amount adsorbed, mol/kg
r_b = bed radius, m
T = temperature, K
t_c = total cycle time, s
T_F = feed temperature, K
T_o = ambient temperature, K
u = interstitial velocity, m/s
V_F = feed flow rate, m^3/s
y_F = feed mole fraction
τ = adsorption step time, s
ϵ = bed porosity
γ = purge to feed ratio
ρ_g = gas-phase density, kg/m^3
ΔH = heat of adsorption, J/mol
ρ_s = bed density, kg/m^3

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