Towards Computer-Aided Separation Synthesis

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DOI 10.1002/aic.10689
Published online November 30, 2005 in Wiley InterScience (www.interscience.wiley.com).

A novel computer-aided design methodology is introduced for synthesizing multi-component distillation networks. A single-objective unconstrained design problem formulation circumvents numerical challenges regarding lack of robustness and multiple local minima. A thermodynamically motivated temperature collocation approach drastically reduces the number of state variables occurring in rigorous column equilibrium and component balances. A novel chromosome encompassing all column configurations is proposed to solve the mixed-integer nonlinear programming (MINLP) using a stochastic genetic algorithm (GA) with minimum user input. The general formulation and robust performance of the computer-aided design approach naturally extends to multi-component mixtures. Applications include solutions to optimal sequencing problems for up to five species. The massive parallelism of the approach allows for the generation of complete solution maps classifying the design space into regions of optimality. The method also applies to optimal sequences of azeotropic columns. The temperature collocation approach constitutes a promising new design framework for synthesis problems previously not amenable to computer-aided design and analysis.© 2005 American Institute of Chemical Engineers AIChE J, 52: 1392–1409, 2006

Keywords: separation synthesis, azeotropic distillation, optimum column design

Introduction

Distillative separation processes constitute a significant portion of the total capital investment, energy requirement, and operating expenses of most chemical plants. Hence, the development of systematic approaches for synthesizing optimal separation sequences is of practical importance. The optimal separation synthesis problem can be described as follows: Given a feed mixture of known species, generate the best distillation train to achieve specified purity targets at minimum cost. Its optimal solutions encompass structural decisions like the configuration of columns, succession of intermediate and final products, as well as continuous parameters determining their operating conditions such as column reflux and reboil ratios.

Despite its significance, computer-aided synthesis of column sequences is still not a routine task.

reactive separation systems were studied by Pistikopoulos (Ismail et al., 1999, 2001). Shah and Kokossis (2002) developed mixed integer linear programs (MILP) for the synthesis of complex distillation systems. Biegler (e.g., Gopal and Biegler, 1999; Lang and Biegler, 2002) developed smoothing algorithms to eliminate the need for integer variables limiting column design, such as dry trays.

Most existing methods deploy simplified vapor-liquid equilibrium relations (that is, ideal or constant relative volatility) not applicable to many non-ideal and azotropic mixtures of industrial relevance. Separation network optimization with rigorous tray-by-tray models challenges numerical optimization codes, because the dissimilar mixture of structural decisions and continuous variables introduces discontinuous regions in the search space (Bauer and Stichlmair, 1995, 1996, 1998; Frey et al., 1997). Thus, derivative-based optimization algorithms may fail to locate attractive structural alternatives in separation synthesis. Moreover, the number of possible column sequences and state variables explodes with increasing species numbers. We propose a novel synthesis procedure featuring combinatorial exploration of all relevant structural alternatives with rigorous column profile computation to advance the capacity of computer-aided distillation design. Our novel problem transformation valid for sharp and non-sharp splits extends to general vapor-liquid equilibrium models for any number of species due to a massive problem size reduction. It will be demonstrated how to discover complete separation networks fully automatically with acceptable computational effort.

In this article we introduce a novel feasibility criterion for ascertaining the realizability of arbitrary column specifications based on a thermodynamically motivated transformation of the column tray equations into a dimensionless bubble point temperature space. The massive problem size reduction also replaces tray-by-tray vapor-liquid equilibrium and component balance constraints with a scalar metric, such that the resulting unconstrained optimization can be solved robustly by a problem-specific genetic algorithm. Further, we discuss industrially relevant applications including: (i) the optimal separation of mixtures of up to five species, (ii) the creation of complete solution maps for classes of separation problems with varying feed compositions, and (iii) the optimal design of azotropic distillation sequences. The article closes with a discussion of the results, followed by conclusions.

### Methodology

The optimal sequencing problem can be expressed as a constrained mathematical program (Eq. 1) in which \( d \) stands for the set of structural and parametric design variables and \( x \) are the state variables, such as the tray liquid and vapor concentrations. The equality constraints \( h(d,x) \) enforce tray equilibrium, component balances, and the column network connectivity; the inequalities \( g(d,x) \) represent the desired final product purity targets and operational limitations. For its numerical solution, this article proposes a novel computational approach combining a problem-specific genetic algorithm (GA) with a rigorous temperature collocation distillation design method. A high-level GA constructs different candidate separation configurations by varying the structural as well as key design variables (that is, different product sequences and operating conditions, like reflux ratio, bottoms, and distillate purities, respectively). The assessment of column realizability is delegated to a feasibility subroutine based on temperature collocation on finite elements (Zhang and Linninger, 2004). It ensures the satisfaction of rigorous tray equilibrium and component balances in a dimensionless bubble point space, thus eliminating the need for the equality constraints in Eq. 1. The resulting unconstrained problem is searched globally with a stochastic GA. The proposed problem formulation uses the smallest possible design variable set to completely represent the general sequencing problem.

\[
\begin{align*}
\min_{d,x} \quad & \text{Cost} \\
\text{s.t.} \quad & h(d,x) = 0 \quad \text{tray balance and connections between columns} \\
& g(d,x) \leq 0 \quad \text{product purities}
\end{align*}
\]

**Criteria for feasible distillation specifications**

The classical performance or rating problem predicts distillate and bottom compositions for a given feed and known column configuration. An inverse design problem, on the other hand, seeks optimal column dimensions and operating conditions for realizing the desired product purities. Solutions to the inverse problem are much harder to converge numerically and may only exist for certain specifications due to thermodynamic separation barriers. Rigorous proofs for the existence of feasible column specifications, in particular for complex column configurations, extractive, or reactive separations, are an open field of research (e.g., Barnicki and Siirola, 2004; Malone and Doherty, 2000; Siirola, 1996; Widago and Seider, 1996).

Recently, a unique distillation problem transformation offers a significant step towards solving inverse design problems rigorously (Zhang and Linninger, 2004). According to this novel temperature transformation approach, the realizability of a given separation target (such as three product purity specifications and the desired reflux) can be determined with the help of liquid tray composition profiles expressed in terms of equilibrium tray temperatures as independent variables, as given in Eq. 2 for the rectifying and Eq. 3 for the stripping section.

**Continuous rectifying profile equations:**

\[
\frac{\partial x_i^r}{\partial T} = - \left( x_i^r - \frac{r + 1}{r} y_i^r + \frac{1}{r} x_{D,i} \right) \\
\times \sum_{j=1}^{C} \left( \frac{\partial K_i}{\partial x_j} x_j^r \right) \\
+ \sum_{j=1}^{C} \left( x_j^r - \frac{r + 1}{r} y_j^r + \frac{1}{r} x_{D,j} \right) K_i
\]

**Continuous stripping profile equations:**

\[
\frac{\partial x_i^s}{\partial T} = - \left( \frac{s}{s + 1} y_i^s - x_i^s + \frac{1}{s + 1} x_{B,i} \right)
\]
The column profile Eqs. 2 and 3 describe the evolution of the liquid tray compositions, \( x_i^R \) and \( x_i^S \), as a function of the equilibrium bubble point temperature, \( T \). The stripping profile emerges from the bottom product composition; the rectifying profile starts from the distillate. In order to solve this boundary value problem with unknown intersection, the differential Eqs. 2 and 3 are discretized using orthogonal polynomial approximation. Multiple finite elements, each with a fixed number of collocation nodes, are implemented for high accuracy. The finite elements decompose the temperature range into segments; the nodes of the orthogonal collocation polynomials further divide each element according to a predetermined number of roots. This orthogonal collocation on finite elements (OCFE) method, also known as Spline Collocation (e.g., Villadsen and Michelsen, 1978), converts the system of Eqs. 2 and 3 into a set of nonlinear algebraic equations. These equations are enforced at discrete dimensionless bubble point temperatures \( \Theta_{\alpha,\beta} \) defined by Eq. 4:

\[
\Theta_{\alpha,\beta} = \frac{T - T_D}{T_B - T_D}
\]

The discretized profile equations are solved for the unknown compositions, \( x_i(\Theta_{\alpha,\beta}) \), according to Eqs. 5 and 6.

Rectifying profiles discretized with OCFE:

\[
A^R(\Theta_{\alpha,\beta}) x_i^R(\Theta_{\alpha,\beta}) - f^R(x_i, T_D, T_B, K, \Theta_{\alpha,\beta}) = 0
\]

Stripping profiles discretized with OCFE:

\[
A^S(\Theta_{\alpha,\beta}) x_i^S(\Theta_{\alpha,\beta}) - f^S(x_i, T_D, T_B, K, \Theta_{\alpha,\beta}) = 0
\]

\( \Theta_{\alpha,\beta} \) refers to the collocation node (i.e., the dimensionless bubble point temperature defined between the bubble point temperatures corresponding to the distillate and bottom compositions, respectively, \( T_D \) and \( T_B \)) in element \( \alpha \) at nodes \( \beta \). \( A^R \) and \( A^S \) are the gradient coefficient matrices of the orthogonal polynomials at collocation nodes, \( \Theta_{\alpha,\beta} \). \( f^R \) and \( f^S \) are the nonlinear right-hand sides of Eqs. 2 and 3 evaluated at nodes \( \Theta_{\alpha,\beta} \). \( x_i^R(\Theta_{\alpha,\beta}) \) and \( x_i^S(\Theta_{\alpha,\beta}) \) represent the unknown weights (i.e., composition of species \( i \)) at node, \( \Theta_{\alpha,\beta} \).

According to this temperature collocation approach, whose theoretical background is discussed in detail elsewhere (Zhang and Linninger, 2004), all liquid composition trajectories originate in bottom and top products and terminate in stationary pinch points, as illustrated in Figure 1. Feasible column profiles exist only within a temperature window delineated by the bubble points corresponding to product compositions and the stable nodes of the pinch equations. A design specification is termed feasible or realizable with simple distillation if stripping and rectifying profiles intersect. The bubble point distance is the Euclidean difference between a stripping and a rectifying composition profile for a specific bubble point temperature. The minimum bubble point distance (BPD) is a precise scalar quantity suitable for formulating a rigorous numerical feasibility criterion. Thus, column specifications leading to zero BPD are feasible; infeasible designs have non-zero BPD, as expressed in Eq. 7:

\[
\text{if } \min_{d,T} \text{BPD} = 0 \rightarrow d \text{ is feasible}
\]

In higher dimensions, exact intersection of trajectories is numerically more difficult to attain than in the two dimensional case, since the smallest error may cause the two profiles to miss each other. The use of the minimum BPD constitutes a numerically more robust intersection criterion than the exact equality of concentrations at the intersection. Moreover, small BPD values also indicate closeness to a feasible design, so that the BPD metric serves as a better guide in the search space than a Boolean hit-or-miss intersection criterion. Temperature collocation offers significant benefits over the regular tray-by-tray models: (i) its independent variable range is finite; (ii) the profile computations expressed in terms of equilibrium tray temperatures are not hampered by singular regions (i.e., saddle and pinch points) requiring infinite numbers of trays to overcome; and (iii) temperature collocation on finite elements drastically was shown to reduce the problem size by two orders of magnitude.

The efficiency of the novel thermodynamically motivated problem transformation has been demonstrated for a wide variety of sharp and sloppy separation problems (Zhang and Linninger, 2004). Apart from its robustness, its decisive advantage is its computational speed. Figure 2 documents a computational result for executing 10,000 design experiments for separating a non-ideal mixture with a high-boiling azeotrope. This computer simulation required only 1,272 sec on a Pentium II 450 Hz PC. The temperature collocation algorithm converges rapidly for both feasible and infeasible specs. For infeasible specifications, the length of the non-zero BPD can be