

# OPTIMIZATION OF CONTINUOUS DISTILLATION COLUMNS USING STOCHASTIC OPTIMIZATION APPROACHES

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The present work describes the use of two stochastic optimization formalisms, namely, genetic algorithms (GAs) and simultaneous perturbation stochastic approximation (SPSA), for the optimization of continuous distillation columns. Both the simple and azeotropic systems are considered in the analysis. In particular, for a specified degree of separation the problem of finding the optimal values of: (i) the number of stages, (ii) reflux ratio (entrainer quantity in the case of azeotropic distillation), (iii) feed location(s), have been addressed. The GA-based optimization has several attractive features such as: (i) convergence to the global rather than to a local minimum, (ii) the objective function need not satisfy smoothness, differentiability, and continuity criteria, (iii) robustness of the algorithm. The other optimization technique used in the study i.e., SPSA, is a rapid gradient-descent related method for multivariate optimization and is especially well-suited in situations where direct computation of the objective function gradient is not feasible, or the objective function measurements could be noisy. The feasibility of utilizing the GA and SPSA techniques has been demonstrated by considering the separation of three binary and two azeotropic systems of industrial relevance.

*Keywords:* distillation optimization; continuous distillation; azeotropic distillation; genetic algorithms; simultaneous perturbation stochastic approximation; Napthali-Sandholm model.

## INTRODUCTION

Continuous distillation is one of the most widely used separation techniques in the chemical process industry and exhaustive scientific and practical knowledge of this process has been gained over the last few decades. Being an energy intensive process, the optimum operation of continuous distillation columns assumes considerable economic importance. In a typical continuous distillation column operation (see Figure 1), the feed enters at any in-between stage and vapours are produced in the reboiler located at the bottom of the column. The vapours, while moving upwards inside the column, are progressively enriched in the more volatile component (MVC) and are condensed in the condenser. A portion of the condensate is then collected as the product while the rest is fed back to the column as a reflux; the reflux as it travels down the column gets enriched in the less volatile component (LVC).

Often, chemical mixtures comprise components forming azeotropes that are difficult to separate using simple distillation. In azeotropic distillation, a third component called the 'entrainer', is added to facilitate the separation of components to the desired purity. For instance, an azeotropic distillation utilizing benzene as the entrainer is widely used for separating an alcohol-water mixture. In this case, a ternary azeotrope is formed at the top of the column,

which upon condensation separates into the benzene-rich (organic) and water-rich (aqueous) phases. A typical schematic of the heterogeneous azeotropic distillation is depicted in Figure 2. The Figure shows that the principal stream ( $F$ ) is composed of the feed approaching the alcohol-water azeotrope and the other stream ( $L'_{N+1}$ ) comprises the entrainer-rich reflux. The ternary heterogeneous mixture that leaves from the top of the column, is fed to the decanter, where it separates into the organic and aqueous phases. While the entrainer-rich organic layer is returned to the column, the aqueous layer is fed to the stripper for the recovery of the dissolved entrainer and alcohol. The other distillation product i.e., anhydrous alcohol, is drawn out at the bottom of the column. Since the composition of the two phases in the decanter is different, in heterogeneous separation, the reflux ratio cannot be defined (as in homogenous distillation) as the rate ratio of the reflux stream ( $L'_{N+1}$ ) and the distillate ( $L''_{N+1}$ ). In heterogeneous azeotropic distillation, a make-up stream of the entrainer is needed to account for the losses caused by its presence in the product as well as vent streams; the make-up stream is taken along with the principal feed consisting of alcohol-water mixture (see Figure 2).

In a typical distillation column, the choice of the operating parameters include fixing of the feed location and reflux ratio (entrainer quantity for azeotropic

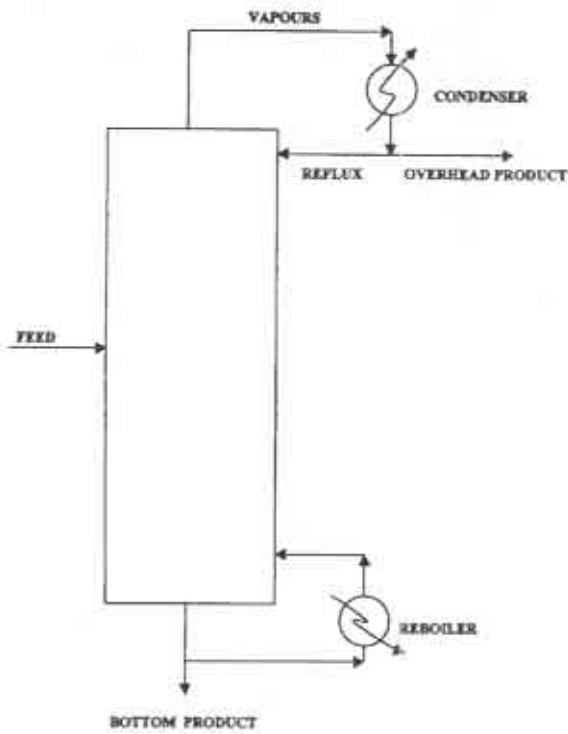


Figure 1. Schematic of continuous distillation column.

distillation), whereas the number of stages becomes the design variable. The total (minimum) reflux condition, gives the minimum (infinite) number of stages. Thus, it can be seen that all distillation columns operate between these extreme limits. In continuous distillation, the reboiler and condenser operations are energy intensive. While an increasing reflux ratio increases both the energy consumption and the tower diameter, it decreases the number of distillation stages. This eventually results in a trade-off between the energy and the fixed costs necessitating process optimization. The objective of such an optimization is the determination of the optimal operating and design parameters for achieving the desired degree of separation at the lowest total cost. The total cost of a distillation operation is made up of two major components. In the first component, the annual running cost of utilities i.e., the heating media to produce vapours, and the cooling media for vapour condensation, are accounted for. The second cost component refers to the annual fixed charges that take into account the interest and depreciation on the installation cost of the column, condenser, and reboiler; this component also includes maintenance of the installed equipment.

Several optimization studies that deal with various design aspects of continuous distillations are reported in the literature<sup>1-6</sup>. A comprehensive list of the pre-1989 references on the distillation column optimization can also be found in Edgar and Himmelblau<sup>7</sup>.

For optimizing the continuous distillation columns, the mixed integer non-linear programming (MINLP) seems to be an attractive approach<sup>4-5</sup>. A related optimization study<sup>3</sup> employing the outer loop method around the well-known Naphali-Sandholm procedure has also been performed. Both these techniques involve gradient computations at some stage in their optimization simulations. It is known

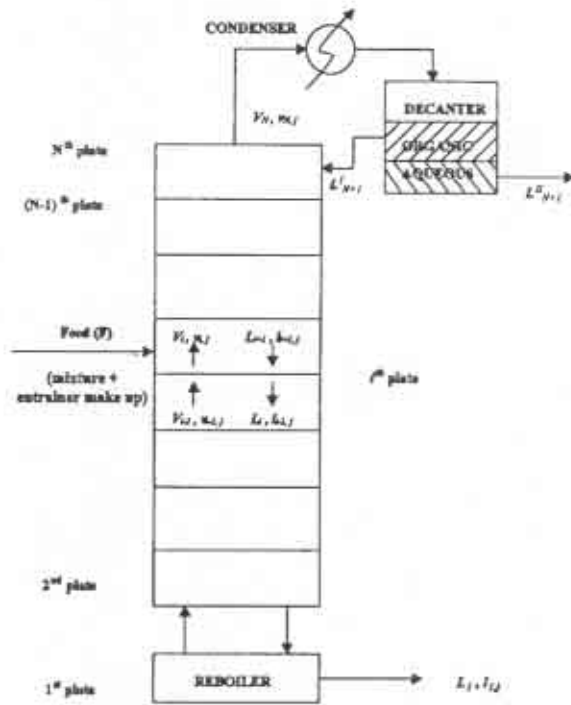


Figure 2. Schematic of azeotropic distillation column.

that the MINLP formalism can get entrapped into a locally optimum solution instead of the desired globally optimum one. Moreover, MINLP methods are complex, computationally intensive and many simplifications become necessary to make them affordable<sup>8</sup>.

For optimizing continuous azeotropic distillation, Prokatakis and Seider<sup>1</sup> used Powell's algorithm together with the material and energy balances, and equilibrium relations. The Powell's algorithm is a direct optimization methodology and simple to execute. The drawback of the direct methods, however, is that they are not as efficient and robust as the indirect methods, such as the gradient-based Newton's (quadratic) approximation techniques<sup>7</sup>. Kingsley and Lucia<sup>6</sup> studied the relationship between the two-phase and three-phase solutions to heterogeneous distillation simulation and optimization problems. Here, the problem necessitating a global optimization methodology was addressed using the tunnelling algorithm. This algorithm is again gradient-based and, therefore, involves derivative computations.

It is noticed, from the above discussion, that a majority of studies on continuous distillation optimization include applications of the deterministic optimization methods (MINLP, Powell's method, tunnelling method, etc.). These formalisms are mostly calculus-based involving direct computation of the gradient. The gradient-based techniques invariably require the objective function to be smooth, continuous and differentiable (well-defined slope values). When the objective function is multimodal, noisy, and fraught with discontinuities, simultaneous fulfillment of these criteria cannot be guaranteed, thus leading to suboptimal solutions. For instance, if the search space includes mixed integer (e.g., the number of distillation stages) and continuous (e.g., reflux ratio) variables, then the objective function could be non-monotonic and possess

multiple local minima. In recent years, the other class of optimization formalisms, known as 'stochastic (random) search algorithms' are becoming popular owing to their capability of performing multi-modal optimization<sup>9-11</sup>. The stochastic methods differ from their deterministic counterparts in that they involve a random component at some stage(s) during their implementation. It should, however, be noted that a randomized search does not necessarily indicate a direction-less search. The stochastic optimization algorithms although computationally intensive, are finding wide-spread acceptance in the chemical engineering applications owing to their affordability and the speed of the modern computing technology. In the present paper two stochastic search techniques, namely *genetic algorithms* (GAs) and *simultaneous perturbation stochastic approximation* (SPSA) have been successfully explored for the optimization of continuous distillation columns.

This paper is organized as follows. The following sections presents details of the optimization formulation corresponding to the continuous simple and continuous azeotropic distillations. This is followed by a review of the conceptual framework and the implementation procedures of the GA and SPSA methods. The final section discusses and compares the results of the optimization simulations conducted using the GA and SPSA strategies.

## PROBLEM FORMULATION

### Continuous Simple Distillation

The objective function used in this study is representative of the total annual cost ( $C_T$ ) that is made-up of two components, namely, the operating cost ( $C_1$ ), and the fixed cost ( $C_2$ ). While  $C_1$  accounts for the energy cost pertaining to the reflux ratio and reboiler duty, the cost component  $C_2$  accounts for the number of stages. The overall optimization objective is expressed as:

$$\text{Minimize } C_T(\mathbf{x}); \quad x_k^l \leq x_k \leq x_k^u \quad (1)$$

where  $C_T$  (\$) is a function of the  $K$ -dimensional decision variable vector,  $\mathbf{x} = [x_1, x_2, \dots, x_k, \dots, x_K]^T$  and  $x_k^l$  and  $x_k^u$  respectively, refer to the lower and upper bounds on  $x_k$ . The three decision variables ( $K = 3$ ) considered for optimization are: (i) the total number of stages ( $N$ ), which is a function of real-valued  $x_1$ , (ii) reflux ratio ( $x_2$ ), (iii) the feed location  $f_i$  (a function of  $x_3$  and  $x_1$ ). The evaluation procedure for the cost components  $C_1$  and  $C_2$  is discussed in Appendix I. The solution to the minimization problem defined in equation (1) should satisfy the following constraints:

- purity constraints:

$$\begin{aligned} x_d^{spec} - x_d^{simu} &\leq 0 \\ x_b^{spec} - x_b^{simu} &\leq 0 \end{aligned} \quad (2)$$

where  $x_d^{spec}$  and  $x_b^{spec}$  represent the desired top and bottom concentrations (mole %) of the MVC, and  $x_d^{simu}$  and  $x_b^{simu}$  refer to the optimized (simulated) values of the top and bottom concentrations of the MVC.

- equality constraints.

As defined by the material balance, equilibrium, summation of the mole fraction and heat balance (MESH) equations (see Appendix II).

In the present work, the matrix method developed by Naphthali and Sandholm<sup>12</sup> has been utilized for the simulation of simple multicomponent distillation to solve the steady-state MESH equations for each plate. The Naphthali and Sandholm (NS) method uses the Newton-Raphson technique to simultaneously solve all the variables in the MESH equations. For simulation purposes, the full NS matrix method for the continuous distillation simulation is combined with the UNIQUAC method for predicting the vapour-liquid equilibrium (VLE).

### Continuous Azeotropic Distillation

The optimization objective for the azeotropic distillation also involves minimization of the total cost,  $C_T$ , with the associated bounds on the three decision variables (see equation (1)). In the present case the decision variable,  $x_2$ , signifies the entrainer quantity. For a  $C$ -component azeotropic system, the optimized solution should satisfy the following purity constraints:

$$x_{ij}^{spec} - x_{ij}^{simu} \leq 0; \quad j = 1, 2, \dots, C \quad (3)$$

where  $x_{ij}^{spec}$  and  $x_{ij}^{simu}$  respectively refer to the desired and optimized bottom concentrations of the  $j$ th component of the azeotropic mixture.

By assuming constant molar overflow<sup>13</sup>, it is possible to simplify the equality constraints defined by the MESH equations. Under this assumption, the energy balance can be ignored and only the total stage mass balance and component balance equations, which are coupled with the equilibrium equations, need to be solved. The equality constraints in respect of the MES (mass balance, equilibrium and summation) equations are described in Appendix III.

In the case of heterogeneous azeotropic distillation, two phases are formed following the condensation of vapours on the top plate. Thus, additional computations taking into account the liquid-liquid equilibrium are necessary for determining the composition and the amount of reflux. For simulating an azeotropic distillation column, the simplified NS method described above which considers the material balance on each plate, is coupled with the phase separation calculations for the condenser; the prediction of the vapour-liquid and liquid-liquid equilibria has been made using the UNIQUAC method.

## STOCHASTIC OPTIMIZATION FORMALISMS

Solutions to the cost minimization problem defined in equation 1 were obtained using the GA and SPSA stochastic optimization formalisms. In the following section, the two optimization methodologies are described in detail along with their step-wise implementation procedures.

### GA-Based Function Optimization

The Genetic Algorithms<sup>14-15</sup> are based on the mechanisms of natural selection and genetics, which play a dominant role in the Darwinian evolution of biological organisms. The GAs are known to be efficient in searching noisy, discontinuous and non-convex solution spaces<sup>10</sup>, their characteristic features are:

- They are 'zero' order search techniques implying that they need only the scalar values of the objective function to be optimized.
- GAs perform a global search, and hence, they mostly converge to (or in the vicinity of) the global optimum of the objective function.
- The search procedure used by the GAs is stochastic and hence they can be utilized without invoking ad-hoc assumptions, such as smoothness, differentiability, and continuity, pertaining to the form of the objective function. Owing to this feature, GAs can be used to solve optimization problems that cannot be conveniently solved using the classical gradient-based algorithms, which require the objective function to simultaneously satisfy the above criteria.
- The GA procedure can be effectively parallelized, which helps in efficiently searching a large multi-dimensional solution space.

Owing to the above-stated attractive features, GAs have been utilized in diverse chemical engineering optimization applications<sup>16-23</sup>. In the earlier studies on the GA-based optimization of distillation processes, Fraga and Matias<sup>24</sup> optimized a reduced optimization problem in which a preselected sequence of distillation units and the associated heat exchanger network (HEN) was considered for the separation of ternary azeotropic mixtures. The objective function evaluation used by Fraga and Matias<sup>24</sup> is a design procedure in itself determining the unit design parameters (reflux rate and number of stages), and the HEN along with the full annualized operating cost. For this objective, they used a parallel version of the GA that was implemented using a distributed memory multicomputer in the form of a network of workstations. In the other study, Wang *et al.*<sup>8</sup> utilized an improved GA (IGA) for synthesizing an optimal distillation sequence and its HEN, with the objective of minimizing the total annual cost. The specific problems considered for validating the IGA methodology were separation of the five- and four-component (non-azeotropic) feed streams. As can be seen, the GA-based studies mentioned analyzed the HEN-augmented distillation columns. In the small and medium-scale industries, the distillation columns are often operated in a stand-alone mode i.e., without HEN-augmentation. This happens particularly when the hot and cold streams are not readily available. In the present paper, therefore, the basic continuous and azeotropic distillation optimization problem involving a single distillation column has been addressed.

To understand the working principles of the GAs, consider a generic version of the optimization problem defined in equation (1):

$$\text{Minimize } f(x), x_k^l \leq x_k \leq x_k^u; \quad k = 1, 2, \dots, K \quad (4)$$

where  $x_k$  denotes the  $k$ th decision variable and  $f(x)$  represents the non-linear objective function. For obtaining a GA-based solution to the minimization problem, first a population of probable (candidate) solution vectors is randomly generated. The suitability of the candidate solution at fulfilling the optimization task is ascertained by evaluating its fitness value (score). It may be noted that the GAs essentially search for solutions possessing high fitness values. Upon evaluating the fitness scores of all candidate solutions in a population, three GA operators

namely, *selection*, *crossover* and *mutation* are used sequentially to produce a 'new generation' of probable solutions. The solutions represented by the new-generation chromosomes are usually better (in terms of their fitness values) as compared to those represented by the chromosomes in the current population. Subsequently, the new population is also subjected to the actions of the three GA operators and this procedure is iterated over successive populations till convergence is achieved. The essence of the GA-methodology can be stated as: the better solutions in the current population are selected for the reproduction and their offspring, generated via the crossover and mutation operations, replace the sub-optimal (i.e. with low fitness values) solutions. The solution population, owing to the repetitive actions of the three GA operators, improves itself from one generation to the next. The GA converges eventually and the best chromosome i.e., the one possessing the maximum fitness score, represents the solution to the optimization problem. Below, a detailed stepwise procedure for the GA implementation is described.

• *Step 1 (initialization)*: Randomly create a population of  $N_{pop}$  number of binary (base-2) strings where the length of each string equals  $l_{chr}$  elements (bits). Every string is divided into  $K$  segments where  $k$ th segment of length  $l_k$  refers to the base-2 representation of the  $k$ th decision variable,  $x_k$ ; the decimal equivalent of  $x_k$  can be evaluated using the corresponding lower and upper limits ( $x_k^l, x_k^u$ ) as given by:

$$x_k = x_k^l + \frac{(x_k^u - x_k^l) \times S_k}{2^{l_k} - 1}; \quad \sum_{k=1}^K l_k = l_{chr} \quad (5)$$

where  $S_k$  represents the base-10 equivalent value of the  $k$ th binary segment. Upon decoding all the  $N_{pop}$  strings in this manner, evaluate their fitness values,  $\xi_n (n = 1, 2, \dots, N_{pop})$ , using the pre-specified fitness function,  $\xi(x)$ . Depending whether the optimization task involves function maximization or minimization, the corresponding fitness function is defined by suitably transforming the objective function. More specifically, for the problems involving function maximization (minimization), the fitness value should increase (decrease) with the increasing value of the objective function. After evaluating the fitness scores of all the strings in the current population, rank the strings in the decreasing order of their fitness scores.

• *Step 2 (selection)*: From the current population, select  $N_{pop}$  number of strings possessing high i.e., above-average fitness values, for constructing the mating pool of parent strings. The parents are selected in a manner such that the strings with better fitness values (scores) get priority for inclusion in the pool. This pool may comprise multiple copies of the fitter chromosomes. Instead of the commonly employed noisy *Roulette-Wheel* (RW) method, it is advantageous to use a more stable method termed *stochastic remainder selection* (SRS), for choosing the parent chromosomes<sup>15</sup>. In the RW technique, the probability of selecting  $n$ th string,  $x_n$ , is directly proportional to its normalized fitness ( $Z_n$ ):

$$Z_n = \frac{\xi_n(x_n)}{\sum_{n=1}^{N_{pop}} \xi_n(x_n)} \quad (6)$$

whereas in the SRS scheme, a  $Z_n$ -dependent real-valued quantity ( $g_n$ ) is computed first:

$$g_n = Z_n \times N_{pop}; \quad 1 \leq n \leq N_{pop} \quad (7)$$

which is then separated into its integer ( $int_n$ ) and the corresponding remainder ( $rem_n$ ) parts, where  $int_n$  denotes the number of copies of the  $n$ th chromosome to be certainly included into the mating pool. The remainder part,  $rem_n$ , represents the probability of taking an additional copy of the  $n$ th chromosome into the mating pool. For instance, in the case of  $g_n = 2.6$  ( $int_n = 2$  and  $rem_n = 0.6$ ), the mating pool receives two copies of the  $n$ th chromosome with unit probability and an additional copy with probability equal to 0.6. This mating pool selection procedure is implemented with the successive chromosomes in the fitness hierarchy until a total of  $N_{pop}$  chromosomes are selected in the pool.

• **Step 3 (crossover):** Select a pair of parent chromosomes at random from the mating pool following which the decision to perform the crossover operation is made based upon the pre-specified value of the crossover probability ( $P_{cr}$ ); typically, a random number is generated between 0 and 1 and the crossover operation is performed on the chosen parent-pair if the number falls below  $P_{cr}$ . Note that the magnitude of  $P_{cr}$  chosen is usually high ( $P_{cr} \geq 0.9$ ). In the single point crossover, which is a widely employed crossover method, the chosen pair of parent strings is cut at the same randomly selected crossover point for generating two substrings per parent chromosome. The second (or the first) substrings of the respective chromosomes are then mutually exchanged and combined to form two offspring chromosomes. This crossover operation is repeated on the  $N_{pop}/2$  number of parent-pairs to obtain  $N_{pop}$  number of offspring chromosomes. The crossover operator in essence helps in improving the combinatorial diversity of the offspring population by utilizing the building blocks of the parent population<sup>10</sup>.

• **Step 4 (mutation):** Randomly mutate (flip) elements of the offspring strings, from zero to one and vice-versa, using  $P_{mut}$  as the probability of performing mutation. Unlike the crossover operation that exchanges the existing information (albeit partly) between the parent chromosomes, the mutation operation introduces characteristics hitherto absent in the offspring population. The mutation operation helps in conducting a local search around the point solutions represented by the unmutated offspring strings. A low value (0.01–0.05) of  $P_{mut}$  is desirable since it avoids creation of very different search sub-spaces, thereby preventing the GA search from becoming absolutely random. Since poor solutions need more corrections than their fitter counterparts, it is advantageous to use a mutation probability value,  $Q_{mut}$ , that is dependent on the chromosome's fitness score<sup>25</sup>.

$$Q_{mut} = (\xi_n(x_n^*) \times 1.01 - \xi_n(x_n)) \times P_{mut} \quad (8)$$

where  $\xi_n(x_n^*)$  refers to the fitness value of the fittest string,  $x_n^*$ .

The chromosome population resulting from the actions of the mutation operator represents a new generation. Thereafter, steps (2–4) are repeated for a sufficiently large number of generations ( $N_{gen}^{max}$ ) to achieve convergence. The criterion for the GA convergence could be that the fitness score of the best string in a population undergoes a very small, or no change, over a large number of successive generations. Upon convergence, the  $K$  number of binary

segments in the chromosome possessing highest fitness score, are decoded (see equation (5)) and the values of the decision variables obtained thereby represent the optimal solution searched by the GA.

### SPSA-Based Function Optimization

The SPSA<sup>9,26–28</sup> technique is based on a highly efficient 'simultaneous perturbation' scheme for approximating the objective function gradient. Here, the gradient approximation irrespective of the number of decision variables ( $K$ ) to be optimized, is performed using only two measurements of the objective function. This procedure is in contrast, for instance, to the standard two-sided finite-difference approximation needing  $2K$  objective function measurements for approximating the gradient. Although an effective stochastic optimization technique that has been successfully utilized in numerous optimization tasks<sup>9,29–30</sup>, the chemical engineering applications of the SPSA formalism are not as widespread as that of the GAs. Notwithstanding this fact, in a recent study<sup>31</sup>, the SPSA formalism has been successfully and advantageously utilized for optimizing an ANN-based process model to conduct the parameter and tolerance design concurrently. The primary virtues of the SPSA scheme are<sup>9</sup>:

- (i) ease of implementation;
- (ii) no need for measurements or direct calculation of the objective function gradient;
- (iii) robustness to noise in the objective function measurements;
- (iv) an empirical evidence of the ability to find the global minimum.

To illustrate the SPSA formalism, the authors reconsider the function minimization problem defined in equation (4). The implementation of the SPSA formalism is an iterative procedure that begins with a randomly initialized  $K$ -dimensional guess solution vector,  $\hat{x}$ ;  $\hat{x}_i^L \leq \hat{x}_i \leq \hat{x}_i^U$ . Unlike the GAs, the SPSA technique requires the objective function to be differentiable since it searches for the optimal solution vector ( $x^*$ ), such that the gradient of the objective function,  $g(x^*)$  attains a zero magnitude. That is:

$$g(x^*) = \partial f(x)/\partial x|_{x=x^*} = 0 \quad (9)$$

In each SPSA iteration,  $g(x)$  is approximated using the numerically efficient simultaneous perturbation technique mentioned earlier. The step-wise procedure for the SPSA implementation is presented below.

- **Step 1 (Parameter initialization):** Set the iteration index  $t$  to zero and initialize the non-negative SPSA parameters namely  $\alpha$ ,  $\phi$ ,  $\tau$ ,  $\kappa$  and  $\theta$ .
- **Step 2 (Gain-sequence computation):** Calculate the  $t$ -dependent values of the SPSA gain sequences ( $\theta$ , and  $\tau$ ) using:

$$\begin{aligned} \theta_t &= \theta/(\kappa + t + 1)^6 \\ \tau_t &= \tau/(t + 1)^9 \end{aligned} \quad (10)$$

- **Step 3 (Formation of simultaneous perturbation vector):** Construct a  $K$ -dimensional perturbation vector ( $\Delta$ ) using Bernoulli  $\pm 1$  distribution by taking 0.5 as the probability of occurrence of +1 or -1.
- **Step 4 (Evaluation of the objective function):** Obtain two

measurements  $f(\hat{x}_i^+)$  and  $f(\hat{x}_i^-)$  of the objective function where  $\hat{x}_i^+ = \hat{x}_i + \tau_i \Delta_i$  and  $\hat{x}_i^- = \hat{x}_i - \tau_i \Delta_i$ . It can be noticed in this step that all the elements of the current solution vector  $\hat{x}_i$  are perturbed simultaneously to obtain the respective perturbed vectors  $\hat{x}_i^+$  and  $\hat{x}_i^-$ .

• **Step 5 (Approximation of the gradient):** Generate the simultaneous perturbation approximation of the unknown gradient  $\hat{g}_i(\hat{x}_i)$  as given by:

$$\hat{g}_i(\hat{x}_i) = \left[ \frac{f(\hat{x}_i^+) - f(\hat{x}_i^-)}{2\tau_i} \right] \times [\Delta_{i1}^{-1}, \Delta_{i2}^{-1}, \dots, \Delta_{iK}^{-1}]^T \quad (11)$$

where  $\Delta_{ik}$  refers to the  $k$ th component (+1 or -1) of the perturbation vector,  $\Delta_i$ .

• **Step 6 (Update of  $\hat{x}_i$  vector):** Update the estimate of the decision variable vector  $\hat{x}_i$  to get its new value as given by:

$$\hat{x}_{i+1} = \hat{x}_i - \theta_i \hat{g}_i(\hat{x}_i) \quad (12)$$

• **Step 7 (Continuation or termination):** Increment the iteration counter  $t$  to  $t+1$  and return to step 2 unless convergence is attained. Any one of the following criteria may be used for determining the convergence: (i) successive iterations impart negligible or no corrections to the decision vector estimate,  $\hat{x}_{i+1}$ ; (ii) the algorithm has performed the prespecified maximum number ( $t_{SPSA}$ ) of iterations.

A judicious choice of the five SPSA-specific parameters, namely,  $\phi$ ,  $\alpha$ ,  $\tau$ ,  $\kappa$  and  $\theta$ , critically contributes to the success of the SPSA simulations. Although the detailed guidelines for selecting the five parameters can be found in Spall<sup>9</sup> they are summarized here for the sake of brevity:

- (i) The theoretically valid and practically effective values of  $\phi$  and  $\alpha$  are 0.602 and 0.101, respectively, although asymptotically optimal values of 1.0 and 0.1666 may also be used<sup>9,27</sup>.
- (ii) In a situation where perfect (i.e. noiseless) measurements of  $f(\hat{x})$  are possible (as in the present study), then  $\tau$  should be chosen as some small positive number.
- (iii) Choose  $K$  such that it is smaller (10% or less) than the maximum number of iterations allowed or expected.
- (iv) Select the  $\theta$  value such that  $\theta/(\kappa+1)^6$  times the magnitude of elements in  $\hat{g}_0(\hat{x}_0)$  is approximately equal to the smallest of the desired change magnitudes among the elements of the  $\hat{x}$  in the early iterations.

## RESULTS AND DISCUSSION

### Continuous Distillation Optimization

For evaluating the performance of the GA and SPSA techniques for optimizing the steady-state staged continuous distillation columns, the authors have considered the separation of three industrially important binary mixtures: (i) methanol-water; (ii) ethanol-water; (iii) water-acetic

acid. The distillation column is often operated with multiple feeds and therefore the methanol-water system with two feed streams has also been examined. The procedural details of the above-stated optimization case studies and the results obtained are described below.

Since the number of stages ( $N$ ) can assume only discrete values, the related decision variable ( $x_1$ ) is converted into its integer equivalent. Also, for the convenience of the procedure, the values of the feed location variable ( $x_3$ ) are normalized. The following expressions have been used for computing  $N$  and the discrete value ( $f_i$ ) of the feed location variable,  $x_3$ :

$$\begin{aligned} N &= \text{int}(x_1) \\ f_i &= \text{int}[1 + x_3(N - 2)]; \\ &\leq x_3 \leq 1; \quad (x_1 - 1) > f_i > 1 \end{aligned} \quad (13)$$

where  $\text{int}(\cdot)$  refers to the integer function. In equation (13), the reboiler and the condenser represent the first and the last distillation stages.

In the GA-based function minimization procedure, the fitness function value should scale inversely with the decreasing value of the objective function. The choice of an appropriate fitness function fulfilling this criterion is critical for obtaining good solutions. Accordingly, a number of fitness function forms were tested and the one given below, which in addition to the total cost also accounts for the purity constraints, yielded consistently good solutions;

$$\xi(x) = R_1 + R_2 \quad (14)$$

where:

$$R_1 = \left[ w_1 \times \left( 1 - \frac{1}{1 + \exp^{-C_T x_2}} \right) \right], \quad (15)$$

and:

$$R_2 = \left[ w_3 \times \left( \frac{1}{1 + \text{abs}(x_d^{\text{spec}} - x_d^{\text{simu}}) + \text{abs}(x_b^{\text{spec}} - x_b^{\text{simu}})} \right) \right] \quad (16)$$

In the above, the term defined by  $R_1$  takes into account the total distillation cost, while  $R_2$  accounts for the purity constraints;  $w_1$ ,  $w_2$  and,  $w_3$  are the weight factors used for: (i) avoiding the computational errors, such as, numerical overflow, division by zero, etc. during the evaluation of the objective function; (ii) expressing the relative importance of the cost components; (iii) making the fitness values more (or less) sensitive to the objective function values. In the GA procedure, a check for the constraint satisfaction is made before evaluating the fitness value of a candidate solution. If any of the equality or purity constraints are violated then the

Table 1. System parameters used for simulating simple continuous distillation simulation.

Binary system parameter	Methanol-water (single feed)	Methanol-water (two feeds)	Ethanol-water	Water-acetic acid
• Number of feeds	1	2	1	1
• Feed composition of MVC, mole %	50	60, 40	60	50
• Feed Flow rate, kmol hr <sup>-1</sup>	60	30, 30	60	100
• Top composition of MVC, $x_d^{\text{spec}}$ , mole %	99	99	95	90
• Bottom composition of MVC, $x_b^{\text{spec}}$ , mole %	5	5	1	1

Table 2. GA-based optimized solutions for continuous distillation\*.

System	$N$	$R$	$f_1$	$x_2^{top}$	$x_2^{bot}$	$C_T^*$ \$ year <sup>-1</sup>	$C_{PROD}^*$ \$ kg <sup>-1</sup>
(i) Methanol-water (single feed)	23	0.98	12	0.990	0.050	151117	0.021
(ii) Methanol-water (double feed)	25	0.93	9, 7	0.990	0.050	145906	0.020
(iii) Ethanol-water	47	2.73	33	0.948	0.009	137549	0.015
(iv) Water-acetic acid	54	2.53	17	0.900	0.010	576923	0.022

\* The desired purity values of the more volatile compound (MVC) in top ( $x_2^{top}$ ) and bottom ( $x_2^{bot}$ ) compositions were: (i) methanol-water system:  $x_2^{top} = 0.99$ ,  $x_2^{bot} = 0.05$ , (ii) ethanol-water system:  $x_2^{top} = 0.95$  and  $x_2^{bot} = 0.01$ , and (iii) water-acetic acid system:  $x_2^{top} = 0.90$ ,  $x_2^{bot} = 0.01$ .

<sup>†</sup>  $C_{PROD}$  is the cost per kg of distillate.

<sup>‡</sup> The weight factors ( $w_k$ ;  $k = 1, 2, 3$ ) used in the optimization simulations are: (i) 2.0, -0.0065, 1.0, (ii) 2.0, -0.0054, 1.0, (iii) 2.0, -0.0056, 1.0 and (iv) 2.0, -0.0065, 1.0.

corresponding candidate solution is penalized by assigning it a zero fitness score<sup>15</sup>. This is one way of ensuring that the constraint violating solution does not compete for a place in the mating pool. A more rigorous *penalty function* approach<sup>32</sup> could also be used as an alternative to the above-stated simple approach for the constraint handling.

Unlike the GAs that search solutions with high fitness scores, the SPSA formalism searches for a solution that minimizes the objective function directly. The following objective function form has been used in the SPSA-based optimization simulations:

$$f(x) = \frac{1}{1 + R_1 + R_2} \quad (17)$$

During the SPSA implementation, a check was made to ensure that the elements of the perturbed solution vectors ( $\hat{x}_1^+$  and  $\hat{x}_1^-$ ) satisfy the corresponding equality and purity constraints. In the event of constraint violation, the objective function (equation (17)) was penalized by equating it to its largest magnitude i.e., unity and simulations were restarted with a new guess solution vector. With these preliminaries, the results of the GA/SPSA-based optimization corresponding to the three case studies are described below.

The values of the GA-specific parameters used in the optimization simulations are:  $K = 3$ ;  $N_{pop} = 30$ ;  $P_{cr} = 0.95$ ;

$P_{mut} = 0.01$  and  $l_{chr} = 120$ . The values of  $K$ ,  $N_{pop}$  and  $l_{chr}$  are chosen such that: (i) the GA simulations do not take long CPU times to converge; (ii) the GA-searched solutions possess adequate precision. It may be noted that as the  $N_{pop}$  value (representing the size solution population) decreases, the CPU time required to manipulate the candidate solutions also goes down proportionately. On the other hand, it is essential that  $N_{pop}$  should be sufficiently large to explore the solution-space as widely as possible. The values of  $K = 3$  and  $l_{chr} = 120$  suggest that in a binary-coded solution string, each decision variable is represented with a 40 ( $= l_{chr}/K$ ) bit precision. This precision can be lowered (enhanced) by appropriately reducing (increasing) the  $l_{chr}/K$  ratio. The system specific parameter values corresponding to the three binary systems, are listed in Table 1. The GA-optimized values of the three decision variables namely: (i) number of stages ( $N$ ), (ii) reflux ratio,  $R(= x_2)$ , and feed location ( $f_1$ ) along with the respective purity values (for bottom and top composition) and the corresponding minimized value of the total cost, are listed in Table 2. For illustration purposes, the composition and temperature profiles as a function of the stage number, pertaining to the GA-based solution for the ethanol-water system, are shown in Figures 3 and 4.

The case studies described above were repeated using the SPSA as an optimization algorithm for which the following values of the SPSA-specific parameters were used:

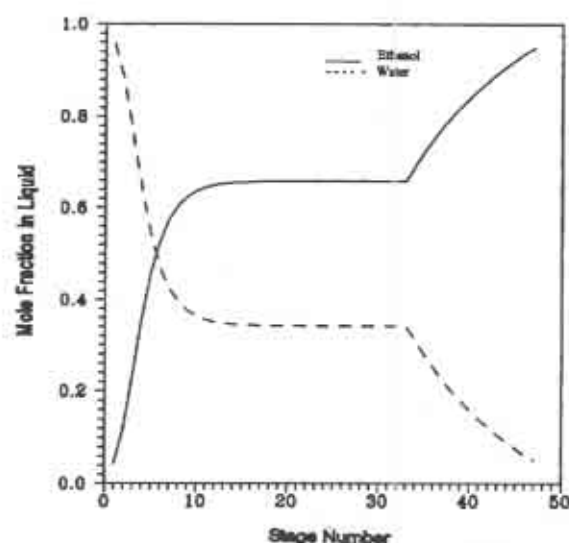


Figure 3. Composition profiles corresponding to the GA-based optimal solution for the ethanol-water system.

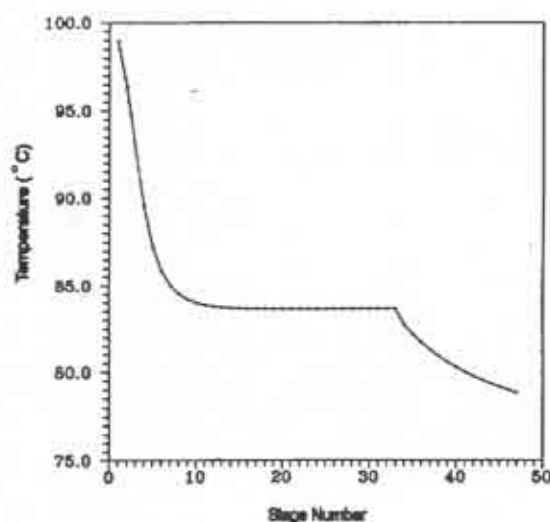


Figure 4. Temperature profile corresponding to the GA-based optimal solution for the ethanol-water system.

Table 3. SPSA-based optimized solutions for continuous distillation\*.

System	$N$	$R$	$f_i$	$x_D^{spec}$	$x_B^{spec}$	$C_T^{\dagger}$ \$ year <sup>-1</sup>	$C_{PROD}$ \$ kg <sup>-1</sup>
(i) Methanol-water (single feed)	29	0.98	13	0.990	0.0501	151240	0.021
(ii) Methanol-water (double feed)	25	0.91	8.5	0.990	0.0496	144413	0.020
(iii) Ethanol-water	43	2.74	34	0.948	0.0084	137899	0.015
(iv) Water-acetic acid	57	2.53	20	0.900	0.0101	578766	0.022

\* Same desired purity values as in the GA-based simulations were employed (see Table 2).

† The weight factors ( $w_k$ ;  $k = 1, 2, 3$ ) used are same as listed in Table 2.

$\alpha = 0.101$ ,  $\phi = 0.602$ ,  $\tau = 0.10$ ,  $\kappa = 250$  and  $\theta = 0.16$ ; the optimal solutions obtained are listed in Table 3. A comparison of the GA- and SPSA-based optimal solutions reveals that the total cost values minimized by both the approaches have nearly the same magnitudes (see Tables 2 and 3), although the optimized values of  $N$ ,  $R$  and  $f_i$  show minor differences. It is also observed that the optimal solutions very closely satisfy the purity constraints in respect of the top and bottom compositions of the MVC.

#### GA/SPSA-Based Optimization of an Industrial Distillation Column

The efficacy of the GA and SPSA techniques was also examined for the optimization of an existing six-stage continuous distillation column in a pharmaceutical industry. In the un-optimized column operation, a feed comprising 70 wt% methanol and 30 wt% water, used to be fed at the rate of 200 kg hr<sup>-1</sup> on the fourth stage from the bottom to produce the distillate containing 95 wt% methanol at the rate of 135 kg hr<sup>-1</sup>. The reflux ratio employed was 2.5:1. Being of lower purity (95% against the desired 98%), the top product methanol could not be recycled back to the process. A physical constraint existed that limited the total column height to a maximum of ten stages. This column was subjected to the GA- and SPSA-based optimization, wherein the number of stages were varied between 6 and

- feed location from the bottom ( $f_i$ ) = 3 (GA/SPSA).

It is seen from the above values that the solutions given by both the methodologies are the same except for the minor difference in the magnitude of the optimal reflux ratio. Next, the GA-based optimization solution ( $N = 9$ ,  $R = 1.82$ ,  $f_i = 3$ ) was physically implemented where the feed rate for the 70 wt% methanol was 200 kg hr<sup>-1</sup>. The column operated in this manner gave a distillate of the desired purity (98%) at the rate of 136 kg hr<sup>-1</sup>. It is noticed that the GA-based optimized solutions could be validated physically with excellent accuracy. The use of the optimized solution resulted in two major benefits to the industry: (i) since the methanol of the desired purity was obtained, it could be recycled to the process; (ii) the vapour load and the energy consumption were reduced by 20%.

#### Continuous Azeotropic Distillation Optimization

Two industrially important systems necessitating the azeotropic distillation are ethanol-water and ethylene diamine (EDA)-water. In both these systems, benzene is utilized as an entrainer. The column specifications used in the optimization simulations corresponding to the ethanol-water and EDA-water systems are given in Table 4. In these simulations, a fitness function similar to equation (14), but modified to account for the purity constraint with respect to the third component (benzene), has been utilized. In this

$$R_2 = \left[ w_3 \times \left( \frac{1}{1 + \text{abs}(x_{b1}^{spec} - x_{b1}^{simu}) + \text{abs}(x_{b2}^{spec} - x_{b2}^{simu}) + \text{abs}(x_{b3}^{spec} - x_{b3}^{simu})} \right) \right] \quad (18)$$

10; the desired purity values for the top and bottom products were 98 wt% and 10 wt% methanol, respectively. For these specifications, the optimal solutions given by the GA and SPSA are:

- number of stages ( $N$ ) = 9 (GA/SPSA);
- optimum reflux ratio ( $R$ ) = 1.82 (GA), 1.87 (SPSA);

function, while the term  $R_1$  remains unchanged, the term  $R_2$  is modified as:

where  $x_{bj}^{spec}$  and  $x_{bj}^{simu}$  ( $j = 1 - 3$ ) represent the desired and the simulated bottoms concentrations of the ternary system components. The GA-based solutions corresponding to the ethanol-water-benzene and the EDA-water-benzene systems are listed in Table 5. These simulations were

Table 4. Column specifications\* for continuous azeotropic distillation of EDA-water-benzene and ethanol-water-benzene systems.

Feed	Component flows, kmol hr <sup>-1</sup>			Component flows, kmol hr <sup>-1</sup>		
	Benzene	Ethanol	Water	Benzene	EDA	Water
• First feed, $F$	2	55.6	79.4	0.5	39.13	11.11
• Second feed at $N_n(L_{N+1})$	$x_2^{\dagger}$	0.5	0.5	$x_2^{\dagger}$	0	0

\* Column pressure: 1.1 atm.

† To be optimized.



Table 5. GA/SPSA-based optimized results for continuous azeotropic distillation\*.

System	$x_{D1}^{line}$	$x_{D2}^{line}$	$x_{D3}^{line}$	$N$	$L_{N+1}^i$ kmol hr <sup>-1</sup>	$f_1$	$C_T^*$ \$/year <sup>-1</sup>	$C_{PROD}^*$ \$/kg <sup>-1</sup>
• GA-based solution								
(i) Ethanol-water-benzene	0.9862	0.0003	0.0135	14	25.92	9	197510	0.073
(ii) EDA-water-benzene	0.9916	0.0018	0.0066	9	299.09	7	1782071	0.020
• SPSA-based solution								
(i) Ethanol-water-benzene	0.9853	0.0003	0.0144	14	25.97	7	196030	0.072
(ii) EDA-water-benzene	0.9836	0.0008	0.0156	9	299.47	6	1782145	0.020

\* The desired purity values of the bottom product for both the systems were:  $x_{D1}^{des} = 0.99$ ,  $x_{D2}^{des} = 0.005$ ,  $x_{D3}^{des} = 0.005$ .

†  $C_{PROD}^*$  refers to cost per kg of bottom product.

‡ The weight factors ( $w_k$ ;  $k = 1, 2, 3$ ) used in the optimization simulations are: 2.0, 1, 1.225 (ethanol-water-benzene) and 2.1, 1.0 and 1.0 (EDA-water-benzene).

performed using the same values of the GA-parameters as in the earlier case studies. The SPSA-based optimal solutions for the two azeotropic systems are also listed in Table 5, and they are in good agreement with those obtained using the GAs. The equilibrium composition and temperature profiles as a function of the stage number in respect of the GA-based optimal solution for the ethanol-water-benzene system are depicted in Figures 5 and 6. The trends of profiles shown in the figures are in good agreement with those obtained by Georgoulaki and Korchinsky<sup>33</sup>.

#### Issues Related to the GA/SPSA-Based Optimization

For non-linear objective functions, the decision surface may consist of multiple local minima of various shapes and sizes. In the case of problems involving function minimization, locating the deepest local or the global minimum assumes great importance. The stochastic nature of the GA and SPSA search methods assists in achieving this objective. Notwithstanding this observation, a special care was taken during the GA/SPSA implementations so that the search space was thoroughly explored. Specifically, for a fixed set of GA- and SPSA-specific parameter values, multiple optimization simulations were performed using a different random number sequence each time. Using different random number sequences changes the initial

candidate solution population in the GA-based optimization, and the initial guess solution vector in the SPSA-based optimization. In this way, a different search sub-space is explored each time, thereby, giving the optimization algorithm a fair chance of locating the deepest local or the global minimum on the objective function surface. Since multiple optimization simulations are necessary to obtain an overall optimal solution, it becomes necessary to examine the CPU time requirements of the GA/SPSA methodologies. The CPU times consumed by the two methods, while arriving at the optimal solutions (Tables 2, 3 and 5) are listed in Table 6. Both the optimization techniques required about ten to fifteen runs to arrive at the overall optimal solutions although the GA-based optimal solution in each run (in contrast to the SPSA-based solution) was nearly the same. It is seen from the CPU time values that the GA and SPSA methods are not computationally costly, even if multiple optimization runs need to be performed. Additionally it is noticed that, except in the case of EDA-water-benzene system, the SPSA methodology has consumed a significantly lower CPU time as compared to the GA method. It was also observed that the SPSA-based solutions are consistently more sensitive to the magnitudes of the SPSA-specific parameters. Thus, it can be inferred that the GA

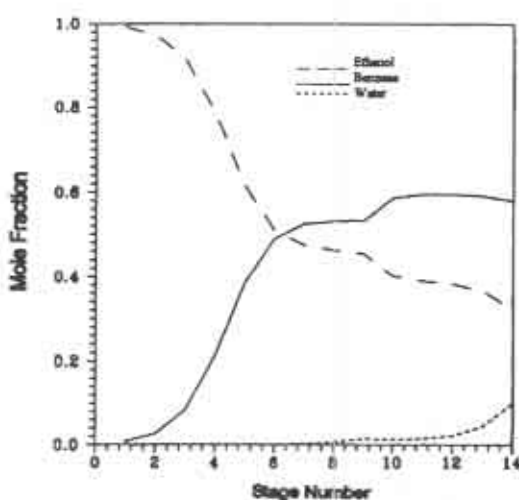


Figure 5. Composition profiles (GA-based) for ethanol-water-benzene system.

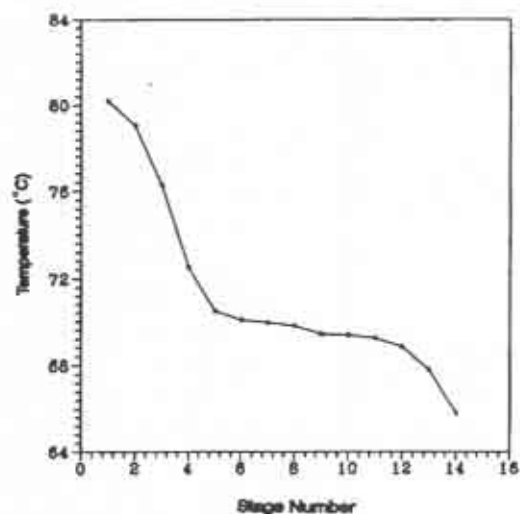


Figure 6. Temperature profile corresponding to the GA-based optimal solution for the ethanol-water-benzene system.

Table 6. CPU time consumed by GA/SPSA-based optimization simulations\*.

System	$N_{st}$	GA		SPSA	
		CPU time, sec	$t_{SPSA}$	CPU time, sec	
(i) Methanol-water (single feed)	5	34	150	22	
(ii) Methanol-water (double feed)	52	153	90	14	
(iii) Ethanol-water	100	293	166	43	
(iv) Water-acetic acid	101	747	30	32	
(v) EDA-water-benzene	5	25	64	88	
(vi) Ethanol-water-benzene	142	509	1	5	

\* Evaluated on 366MHz Pentium Celeron PC.

formalism is more robust compared to the SPSA, although the latter is numerically more efficient.

### CONCLUSION

To summarize, this paper presents two stochastic formalisms, namely GA and SPSA for the optimization of continuous simple and continuous azeotropic distillation columns. These optimization paradigms possess positive characteristics, such as: (i) only the objective function measurements and not the measurements (or direct calculation) of the objective function derivatives, are needed in their optimization procedures; (ii) simplicity of the algorithms; (iii) tolerance to noisy objective functions. A particularly significant advantage of the GA methodology is that unlike most commonly used gradient-descent optimization methods, it does not require the objective function to be smooth, differentiable and continuous, simultaneously. The efficacy of the GA/SPSA formalisms for the optimization objective involving minimization of the total distillation cost, has been demonstrated by considering three binary (non-azeotropic) and two tertiary (azeotropic) systems of industrial importance; the column parameters optimized are the number of stages, reflux ratio (entrainer quantity for azeotropic distillation) and the feed location. The optimization results obtained suggest that the GA and SPSA methodologies can be gainfully employed for optimizing the continuous distillation columns.

### APPENDIX I

#### Computation of Total Cost

The objective function ( $C_T$ ) for both, continuous simple and continuous azeotropic distillations represents the total annual cost (\$). This cost comprises two additive components:

$$\left\{ \begin{array}{l} \text{Total Cost} \\ C_T \end{array} \right\} = \left\{ \begin{array}{l} \text{Energy} \\ \text{Cost, } C_1 \end{array} \right\} + \left\{ \begin{array}{l} \text{depreciation + interest + maintenance} \\ \text{fixed} \\ \text{Cost, } C_2 \end{array} \right\} \quad (\text{A1})$$

where the energy cost,  $C_1$ , which is directly proportional to the heating cost is calculated according to:

$$C_1 = \frac{Q_r \times C_s \times N_D \times 24}{\lambda_{steam}} \quad (\text{A2})$$

where  $Q_r$  is the reboiler duty ( $\text{kcal hr}^{-1}$ ); ( $= 500 \text{ kcal hr}^{-1}$ ) is the latent heat of steam vaporization;  $C_s$  refers to the steam cost ( $= 0.0186 \text{ \$ kg}^{-1}$ ) and  $N_D$  denotes the number of yearly working days ( $= 330$ ).

The fixed cost,  $C_2$  ( $\text{\$ yr}^{-1}$ ), consists of packing ( $C_{pack}$ ) and column ( $C_{col}$ ) costs where  $C_{pack}$  is computed as:

$$C_{pack} = A_c \times N \times HETP \times C_{pack}^0 \quad (\text{A3})$$

Here,  $A_c$  representing the column area is calculated from the total vapour load on the basis of vapour velocity corresponding to the top temperature and capacity factor ( $C_f$ ) of the packing ( $C_f = 1.5$ );  $C_{pack}^0$  denotes the packing cost per unit volume ( $= 2325.58 \text{ \$ m}^{-3}$ ) and the HETP value for the packing is 0.6 m. The second cost component of  $C_2$  i.e.,  $C_{col}$ , is calculated on the basis of internals from the following correlation.

$$C_{col} = 3.14 \times 1.4 \times d_c \times N_{st} \times HETP \times W_s \times \rho_s \times C_{steel} \quad (\text{A4})$$

In this expression,  $d_c$ ,  $W_s$ ,  $\rho_s$  and  $C_{steel}$ , refer to the column diameter, column thickness ( $= 0.006 \text{ m}$ ), density ( $= 8000 \text{ kg m}^{-3}$ ) of the column material (steel) and its cost ( $\text{\$/kg}$ ). Assuming depreciation, interest, and maintenance costs of 18%, 15% and 2%, respectively, the total annual cost to be minimized is evaluated as:

$$C_T = C_1 + 0.35C_2 \quad (\text{A5})$$

In the operation of an azeotropic distillation column, a small quantity of entrainer is lost through the vent condenser and bottom product. Thus, the cost of entrainer loss approximately amounting to 3% of the total entrainer quantity, must be additionally considered while evaluating  $C_T$ :

$$C_T = C_1 + 0.35C_2 + C_3 \quad (\text{A6})$$

where,  $C_3$  ( $= 0.23 \text{ \$ kg}^{-1}$ ) refers to the cost of entrainer, i.e. benzene.

### APPENDIX II

#### Naphthali-Sandholm (NS) Model For Continuous Simple Distillation

The case of  $N$  stages separating  $C$  number of components is considered, where the first stage refers to the reboiler and the  $N$ th stage is the condenser. The NS model accounts for the material balance, equilibrium, summation and heat balances (MESH) on each stage, thus forming a set of  $N/(2C + 1)$  algebraic equations in as many unknowns. The assumptions for the model are: ideal vapour phase, constant column pressure and molar overflow, and ideal stages. The total liquid and vapour flows entering the  $i$ th ( $i = 1, 2, \dots, N$ ) stage are expressed as  $L_i$  and  $V_i$ , respectively, while the individual flow of the  $j$ th ( $j = 1, 2, \dots, C$ ) component is expressed as  $l_{ij}$  and  $v_{ij}$ , respectively. It is now possible to write the MESH equations as given below:

- component material balance:

$$M_{ij} = \left(1 + \frac{S_i}{V_i}\right) \times v_{ij} + \left(1 + \frac{S_i}{L_i}\right) \times l_{ij} - f_{ij} - l_{i+1,j} - v_{i-1,j};$$

$$i = 2, \dots, N - 1; \quad j = 1, 2, \dots, C \quad (\text{A7})$$

- material balance for the reboiler and condenser:

$$M_{1j} = \left(1 + \frac{S_1}{V_1}\right) \times v_{1j} + \left(1 + \frac{s_1}{L_1}\right) \times l_{1j} - f_{1j} - l_{2j} \quad (\text{A8})$$

$$M_{Nj} = \left(1 + \frac{S_N}{V_N}\right) \times v_{Nj} + \left(1 + \frac{s_N}{L_N}\right) \times l_{Nj} - f_{Nj} - v_{N-1,j} \quad (\text{A9})$$

where,  $M_{ij}$  represents the discrepancy function expressed in terms of moles  $\text{hr}^{-1}$ ;  $S_i$  and  $s_i$  are the vapour and liquid side-streams, and  $f_{ij}$  refers to the feed flow.

- equilibrium relationship:

$$Q_{ij} = \frac{\eta_i \times m_{ij} \times V_i \times l_{ij}}{L_i} + \frac{(1 + \eta_i) \times v_{i-1,j} \times V_i}{V_{i-1}} - V_{ij} \quad (\text{A10})$$

where  $Q_{ij}$  refers to the discrepancy function (moles  $\text{hr}^{-1}$ );  $\eta_i$  is the Murphree stage efficiency, and  $m_{ij}$  represents the equilibrium constant for the component  $j$  on  $i$ th stage. In equation (A10), the UNIQUAC method enters into calculations via the equilibrium constant,  $m_{ij}$ .

- energy balance equation:

$$E_i = \left(1 + \frac{S_i}{V_i}\right) \times H_i + \left(1 + \frac{s_i}{L_i}\right) \times h_i - h_{ij} - H_{i-1} - h_{i+1}; \quad (\text{A11})$$

$$i = 2, \dots, N-1$$

- energy balance equations for the reboiler and condenser:

$$E_1 = \left(1 + \frac{S_1}{V_1}\right) \times H_1 + \left(1 + \frac{s_1}{L_1}\right) \times h_1 - h_{r1} - h_2 \quad (\text{A12})$$

$$E_N = \left(1 + \frac{S_N}{V_N}\right) \times H_N + \left(1 + \frac{s_N}{L_N}\right) \times h_N - h_{rN} - H_{N-1} \quad (\text{A13})$$

where  $E_i$  refers to the discrepancy function ( $\text{kcal hr}^{-1}$ ). In equation (A11), the enthalpy values,  $H_i$  and  $h_i$ , are for the vapour and liquid, respectively. Equations (A7–A13) were solved simultaneously using the Newton-Raphson method for obtaining the stage temperature and component liquid and vapour flow rates for which a linear pressure and temperature profile was assumed. The pressure on the  $i$ th stage ( $p_i$ ) is given by:

$$p_i = P_1 - i \times \Delta p \quad (\text{A14})$$

where,  $P_1$  is the bottom pressure and  $\Delta p$  refers to the pressure drop across the stage. The initial guess value for the temperature at each stage is given by:

$$T_i = T_1 + \frac{(i-1) \times (T_N - T_1)}{(N-1)} \quad (\text{A15})$$

where  $T_N$  and  $T_1$  are the temperatures of the condenser and reboiler, respectively, that assume values of the boiling points of the MVC and LVC.

### APPENDIX III

#### Simulation Model for Continuous Azeotropic Distillation

The behaviour of the azeotropic distillation was simulated using the simplified Naphthali-Sandholm model<sup>10</sup>, coupled with the phase separation calculations for the condenser. In the model, the independent variables are the temperature and component liquid flow at each stage. Under the assumptions stated in Appendix II, the NS model gets simplified and only the total stage mass balance and the component balances coupled with the equilibrium equations need to be solved. Here, the material balance on each stage for a  $C$ -component system is represented by a set of  $[C+1]$  algebraic equations (see Figure 2). For solving the case of phase splitting, the overall component flow ( $l_{ij}$ ), and the total flow,  $L_i$ , are used. The component balance equations applicable for the  $j$ th component on the  $i$ th stage are:

$$l_{i,j} + l_{i,j} \varphi_{i,j} - \varphi_{i-1,j} l_{i-1,j} - l_{i+1,j} - f_{i,j} = 0; \quad (\text{A16})$$

$$i = 2, \dots, N-1; \quad j = 1, 2, \dots, C$$

The material balance equations for the reboiler and  $N$ th stage are:

$$l_{1,j} + l_{1,j} \varphi_{1,j} - l_{2,j} - f_{1,j} = 0 \quad (\text{A17})$$

$$l_{N,j} + l_{N,j} \varphi_{N,j} - \varphi_{N-1,j} l_{N-1,j} - f_{N,j} = 0 \quad (\text{A18})$$

where,  $l_{ij}$  represents the moles of  $j$ th component in the liquid stream leaving the  $i$ th stage;  $f_{i,j}$  refers to the moles of  $j$ th component fed to the  $i$ th stage and  $\varphi_{i,j}$  is the stripping factor defined as:

$$\varphi_{i,j} = \frac{V_i \gamma_{i,j} P_{i,j}}{P_s L_i} \quad (\text{A19})$$

where  $L_i$  is the total liquid flow from the  $i$ th stage;  $V_i$  refers to the total vapour flow from the  $i$ th stage;  $\gamma_{i,j}$  describes the activity coefficient of the  $j$ th species in the  $i$ th stage;  $P_{i,j}$  represents the vapour pressure of  $j$ th pure component corresponding to the temperature of  $i$ th stage, and  $P_s$  denotes the system pressure.

- the total material balance on the  $i$ th stage is given as:

$$L_i - l_{i,1} - \dots - l_{i,C} = 0; \quad i = 1, 2, \dots, N; \quad (\text{A20})$$

$$j = 1, 2, \dots, C$$

Vapours leaving the  $N$ th stage usually form two immiscible phases after condensation.

Accordingly, the following equations apply for the condenser:

- component balance equations:

$$l'_{N+1,j} + l''_{N+1,j} = v_{N,j} \quad \text{for } j = 1 \text{ to } C \quad (\text{A21})$$

where  $l'_{N+1,j}$  represents the moles of  $j$ th component in the entrainer-rich phase which is refluxed to the column and  $l''_{N+1,j}$  represents the moles of  $j$ th component in the water-rich phase.

- phase equilibria equations for the decanter<sup>34</sup>:

$$\left( \frac{l_{N+1,i}^I}{\sum_{j=1}^C l_{N+1,j}^I} \right) \gamma_{N+1,j}^I = \left( \frac{l_{N+1,j}^{II}}{\sum_{j=1}^C l_{N+1,j}^{II}} \right) \gamma_{N+1,j}^{II} \quad (\text{A22})$$

where  $\gamma_{N+1,j}^I$  and  $\gamma_{N+1,j}^{II}$  represent the activity coefficients of the  $j$ th species in the decanter corresponding to the composition of phases I and II, respectively.

In the GA/SPSA optimization simulations, equations (A16–A20) were solved by the Gauss elimination method using a prespecified value for the reflux composition. The computed values of  $v_{N,j}$  were then used to solve equations (A21) and (A22) and to obtain the values of the reflux ( $l_{N+1,j}^I$ ). The calculations for the main column are then repeated till the assumed and the optimized reflux values match closely.

#### NOMENCLATURE

$C$	number of components
$C_T$	total annualized cost, \$
$C_1$	energy cost, \$
$C_2$	fixed cost, \$
$C_{PROD}$	product cost, \$ kg <sup>-1</sup>
$E$	discrepancy function for energy balance
$f_i$	integer-valued feed location
$F$	feed flow, kmol hr <sup>-1</sup>
$h$	enthalpy of liquid, kcal mol <sup>-1</sup>
$H$	enthalpy of vapour, kcal mol <sup>-1</sup>
$k$	number of decision variables
$l$	liquid flow of component, kmol hr <sup>-1</sup>
$L$	total liquid flow, kmol hr <sup>-1</sup>
$l_{chr}$	chromosome length
$m$	equilibrium constant
$M$	discrepancy function for component mass balance
$n$	string index
$N$	total number of stages including condenser and reboiler
$N_{gen}$	number of generations
$N_{pop}$	population size
$P_s$	system pressure, atm
$P_b$	bottom pressure, atm
$p$	vapour pressure, atm
$\Delta_p$	pressure drop in each plate
$P_c$	probability of crossover
$P_m$	probability of mutation
$Q$	discrepancy function for equilibrium relationship
$Q_{mut}$	variable probability of mutation
$Q_r$	reboiler duty, kcal hr <sup>-1</sup>
$R$	reflux ratio
$S$	vapour side-stream
$s$	liquid side-stream
$T_1$	reboiler temperature, °C
$T_N$	condenser temperature, °C
$V$	total vapour flow
$v$	vapour component flow
$x_j^{des}$	desired concentration of $j$ th distillate component
$x_j^{bot}$	desired concentration of $j$ th bottom component
$x_j^{opt}$	optimized concentration of $j$ th distillate component
$x_j^{bot}$	optimized concentration of $j$ th bottom component
$x$	decision variable vector
$x_1$	real-valued decision variable representing number of stages
$x_2$	reflux ratio; in the case of azeotropic distillation entrainer quantity
$x_3$	normalized feed location
$Z_n$	probability of selecting $n$ th string

#### Greek symbols

$\xi_n$	fitness value of $n$ th chromosome
$\gamma$	activity coefficient
$\beta$	decay rate

$\eta$	Murphree's stage efficiency
$\varphi$	stripping factor
$\lambda_{steam}$	latent heat of steam vaporization

#### Superscripts

$I$	organic liquid phase
$II$	aqueous liquid phase

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