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OPTIMAL DESIGN METHODOLOGY FOR AZEOTROPIC DISTILLATION COLUMNS

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Abstract
We develop a short and optimal design methodology for multicomponent mixtures, based on the concept of minimum difference in composition. The minimum reflux ratio is calculated through an analytical procedure, which allowing their use in multicomponent mixtures. The procedure also includes a simple criterion to optimize the structural design of the column, without increasing energy consumption. The method is tested with different azeotropic mixtures, and it is compared with the results of a multiobjective genetic algorithm with constraints. Results show that designs obtained with the new and stochastic methods are quite similar, but less time and computational resources are required.

Keywords: optimal design, azeotropic mixtures, short procedure

1. Introduction

Research in azeotropic distillation has increased in the last 20 years, particularly in the design area. The complicated nature of azeotropic mixtures makes the design procedure more complicated than that for ideal mixtures. An important step, previous to the design step, is the verification of the feasibility of the split, according to Stichlmair and Fair[1]. Once this is verified, then we can apply a design methodology. Several design methodologies were developed for this purpose, being the first the Boundary Value Method[2, 3], proposed by Doherty et al in 1985. The Boundary Value Method[2, 3], BVM, considers material balances in form of differential equations to calculate the number of plates, in each section of the column. The differential equations are solved from outside to inside the column, locating the feed stage where liquid compositions of both sections are equal. Nevertheless, in many cases the intersection of operation profiles is far away from feed composition, being the feed introduced in a stage where composition differs significantly. Moreover, the method uses residue curves to make calculations in total reflux; it is worth to mention that residue curves are operation lines at total reflux in packed columns, while in staged columns these lines are represented by distillation lines[1]. Also, the BVM assumes that at minimum reflux condition there is collinearity between the feed, the feed pinch point and the saddle pinch point. However, this collinearity is satisfied in ideal mixtures, while in azeotropic mixtures is just an approximation.

In 1990, Julka and Doherty[4] made the extension to multicomponent mixtures from the original BVM. The Zero-Volume Criterion method, ZVC, considers that minimum reflux ratio is obtained when there is collinearity between feed composition and the C-1 pinch points located in a hyperplane, where C is the number of components. However, this method works just in direct and indirect cuts, since pinch points that determine minimum reflux ratio must to be known a priori. Also, the idea of collinearity for minimum reflux ratio stands, in spite of being just an approximation in azeotropic mixtures.

The Minimum Angle Criterion, MAC, was presented in 1991 by Köhler et al[5]. The MAC considers that at minimum reflux ratio the angle between feed composition and a pinch point of each section of the column must to be a minimum. This criterion is empiric, equivalent to the collinearity of the BVM and ZVC, and it does not have a physical explanation for more than three components.

In 1994, Poellman et al[6] proposed the Eigenvalue Criterion, EC, which is an hybrid of the methods BVM and ZVC. In this method, the profiles are calculated just after the pinch point in the direction of the unstable eigenvector of the pinch point, using a tray to tray calculation. The minimum reflux ratio is the smallest value that allows the intersection of the profile of one section with a pinch point of the other section. However, the computational effort is increased when there is more than one unstable eigenvector in the pinch point, because a lot of profiles must be calculated.

In 1998, Bausa et al[7] presented the Rectification Bodies Method, RBM. A rectification body is a triangle delimited by all possible trajectories of pinch points, in stripping or rectification section, from products to feed. For each trajectory, a calculation of the generated entropy is performed, in order to verify that all of them are thermodynamically consistent. In the RBM, the minimum reflux ratio is found when rectification and stripping bodies touch each other in one point.

On the other hand, Liu et al[8] developed a method that considers that inside azeotropic regions there are compartments, which can be considered as ideal. A distillation region with one additional saddle node can be divided in compartments, which can be tried as ideal regions. So, in these compartments the Fenske-Underwood-Gilliland[9] method can be used, considering azeotropes as pseudo components. In their paper, they report that minimum reflux ratio and number of stages calculated with their method have deviations until 22% and 30%, respect to the values calculated in HYSIS.

In 2007, Gutiérrez-Antonio et al[10] presented a design method that overcomes the limitations of the previous ones[2-8]; it is based on distillation lines, and it uses algebraic material balances solved from the extremes to inside the column. The feed stage is located using as criterion the minimum composition difference between each stage in the column and the feed. However, calculation of minimum reflux ratio is restricted to ternary mixtures, since intersection of profiles must be verified. Also, in some cases large pinch zones are obtained, increasing the cost of the column without benefiting the separation.

In 2008, Lucia et al[11] use the shortest stripping line approach to present a method that minimize the energy requirements in distillation. The procedure is a global optimization algorithm that includes formulations as nonlinear and mixed integer nonlinear programming. A variety of examples are tested with this strategy, and results show that the minimum energy requirements are found for any number of components. In spite that this procedure is reliable and useful for multicomponent mixtures, deep knowledge in mathematical programming and considerable computational resources are required.

In general, all the procedures presented allow designing azeotropic columns. The BVM and its derivations[2-6] assume a collinearity criterion to calculate minimum reflux, in spite of being just an approximation in azeotropic mixtures. The method based on minimum composition difference[10] overcomes the previous limitations[2-9]; however, calculation of minimum reflux ratio is restricted to ternary mixtures, and in some cases designs with large pinch zones are obtained. Moreover, the optimization algorithm of Lucia et al[11] is reliable and useful for multicomponent mixtures, but considerable computer resources are required. It would be desirable to have a good methodology that allows having an optimal design with a short procedure.

In this work, we propose a short and optimal design methodology for azeotropic multicomponent mixtures, which considers material balances and the concept of minimum composition difference (based on our previous contribution[10]). The minimum reflux ratio is calculated through an analytical procedure, which allowing their use in multicomponent mixtures. The procedure includes a simple criterion to eliminate pinch zones in the design, without increasing energy consumption. The method is tested with different azeotropic mixtures, and their results are compared with those from a multiobjective genetic algorithm with constraints[12]. Results show that the new method generates similar designs to those from the optimization algorithm, but with less time and computational resources.

2. Design procedure

In the design of azeotropic columns, verification of the split feasibility is an important step previous to the design. A split is feasible if is located in the region delimited by distillation boundary, distillation line through feed composition, and material balances of direct and indirect separations. If the split is feasible, then we can apply a design procedure. The proposed methodology is based in distillation lines, and it uses algebraic material balances solved from outside to inside the column. Equations for rectifying (Eq. 1) and stripping (Eq. 2) sections (established on our previous contribution[10]) are:

$$y_{n+1,i} = \frac{r}{r+1} x_{n,i} + \frac{1}{r+1} z_{D,i} \tag{1}$$

$$x_{n+1,i} = \frac{s}{s+1} y_{n,i} + \frac{1}{s+1} z_{B,i} \tag{2}$$

$$\sum_{i=1}^{C} x_i = 1; \sum_{i=1}^{C} y_i = 1 \tag{3}$$

Where $x_{j,i}$ and $y_{j,i}$ are the liquid and vapor phase composition of component i in stage j, respectively; r and s are the reflux and reboil ratios, respectively; $z_{D,i}$ and $z_{B,i}$ are the compositions of component i in distillate and bottom products (liquid or vapor), respectively; and C is the number of components. The equation 3 represents the summation equations for liquid and vapor phases; thereby, equations 1 and 2 are applied to i=1 to C-1 components. Considering the thermal condition, q, of the feed stream, the reboil ratio can be calculated with Equation 4 (established on our previous contribution[10]):

$$s = (r+q)\left[\frac{z_{B,1} - z_{F,1}}{z_{F,1} - z_{D,1}}\right] + (q-1) \tag{4}$$

Moreover, feed, bottom and top products composition must to be collinear in order to satisfy the global material balance. In order to satisfy the global material balance for multicomponent mixtures, the bottom compositions for l components must be calculated with the next equation:

$$z_{B,l} = z_{F,l} + (z_{F,l} - z_{D,l})\left[\frac{s-q+1}{r+q}\right] \tag{5}$$

$$l = 2, \ldots, C-1 \tag{6}$$

Once the freedom degrees are fixed, we calculate the reboil ratio (Eq. 4), and the composition of l components (Eq. 5-6). Finally, we calculate rectifying and stripping profiles (Eq. 1-3) for a reflux given.

According to Towler et al[13], the intersection of composition profiles anywhere in the composition space is a necessary and sufficient condition to establish the feasibility of the split. This necessary and sufficient condition is employed to calculate minimum reflux ratio. The calculation involves an iterative process where reflux ratio is varied in small increments (0.01 for instance). For each reflux the operation profiles are calculated and compared to find their first intersection; at this point, the reflux ratio is minimum. Due to the different configurations in the operation profiles, as consequence of the wide variety of azeotropic maps, the comparison of the operation profiles involves some considerations. It is clear that if we just compared all compositions of rectifying profile against those of the stripping profile, we would have an incorrect calculation; since, the compositions of the profiles are not continue, then in order to compare them we may have to establish a tolerance, but this tolerance just could quantify the distance between two compositions, but not guarantees the intersection of the profiles in all cases. So, in order to ensure the intersection of the operation profiles we construct lines for each two consecutive compositions of each profile, which are calculated with Eq. 1-6. Then, we verify the intersection of each line of the rectifying profile against each line of the stripping profile. The lines for rectifying (Eq. 7) and stripping (Eq. 8) sections are expressed as parametric equations:

$$\begin{gathered} R_{m \to m+1} = R_m + \alpha(R_{m+1} - R_m) \\ 0 \le \alpha \le 1 \end{gathered} \tag{7}$$

$$\begin{gathered} S_{k \to k+1} = S_k + \beta(S_{k+1} - S_k) \\ 0 \le \beta \le 1 \end{gathered} \tag{8}$$

$$\begin{gathered} R_m + \alpha(R_{m+1} - R_m) = S_k + \beta(S_{k+1} - S_k) \\ 0 \le \alpha, \beta \le 1 \end{gathered} \tag{9}$$

Where $R_{m \to m+1}$ is the line in the rectifying section that includes the composition points m and m+1, and $S_{k \to k+1}$ is the line in the stripping section that includes the composition points k and k+1. The parameters are introduced to consider that the intersection can take place in any part of the line, including the extremes (compositions). So, if there are two parameters α y β such that $R_m=S_k$, then there is intersection between the operation profiles. As result, we have a simple linear system with two unknown parameters and C equations, one for each component (Eq. 9).The procedure to calculate the minimum reflux ratio, Figure 1, can also be applied to verify the feasibility in multicomponent mixtures, since if there is no minimum reflux found in a very wide range of values, then the separation is not feasible. Once that the minimum reflux ratio is found, then the operating reflux ratio is fixed; for this, heuristic rules of Douglas[14] (1.1Rmin) or Doherty and Malone[15] (1.5Rmin) can be applied.

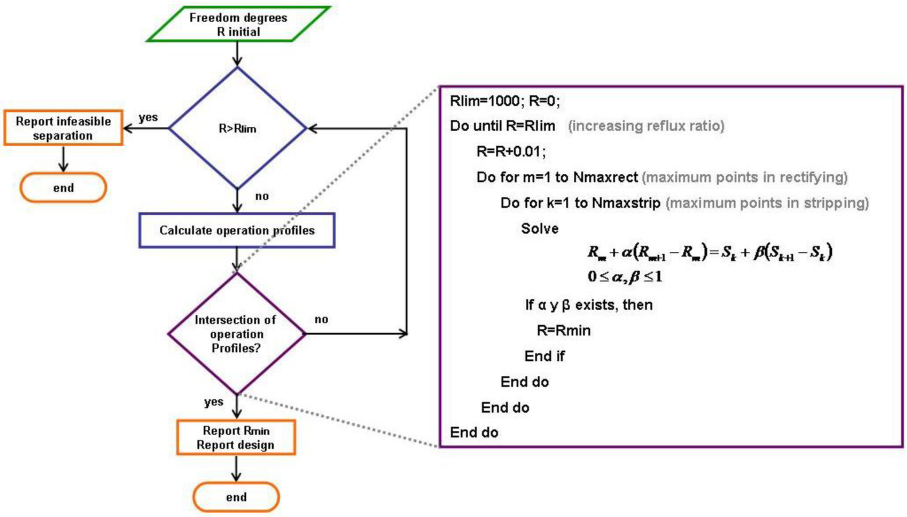

Figure 1. Algorithm and pseudo code to calculate the minimum reflux ratio

The complete design of the column involves the location of the feed stage. The feed stage is located where a minimum difference composition in rectifying and stripping sections is found. The two sections of the column are considered in the minimization procedure, so we can know the number of stages in each section and the location of the feed stage. The composition difference for each section of the column is calculated with (established on our previous contribution[10]):

$$d_R = \sqrt{\sum_{i=1}^{c}\left(z_{F,i} - x_{NR,i}\right)^2 + \sum_{i=1}^{c}\left(z_{F,i} - y_{NR,i}\right)^2} \tag{10}$$

$$d_S = \sqrt{\sum_{i=1}^{c}\left(z_{F,i} - x_{NS,i}\right)^2 + \sum_{i=1}^{c}\left(z_{F,i} - y_{NS,i}\right)^2} \tag{11}$$

Where d_R and d_S are the composition difference between the feed and some stage, *NR* or *NS,* in the rectifying or stripping sections. In this way, the feed is introduced with the minimal disturbance, making the design more efficient. Finally, once that the complete design is obtained, we can apply the criterion shown in Eq. 12 to the operation profiles, in order to eliminate pinch zones:

$$\left|\frac{d^2 x_i}{dn^2}\right| \le 1.0 * 10^{-4} \tag{12}$$

The differential equation (Eq. 12) is calculated as composition differences of the operation profiles. The optimal location of the feed stage along with the refinement of the final design allows having the better structure for the separation. Also, the algorithm for minimum reflux ratio allows identifying the minimum energy consumption, from which the column is designed. The most remarkable feature is that this new methodology just considers material balances and very simple equations.

3. Cases of study

The design method is applied to several azeotropic mixtures; due to spaces limitations the results of two of them are presented: acetone-isopropanol-water (M1), acetone-chloroform-benzene (M2). Feed compositions are: 0.63/0.07/0.30 (M1), 0.50/0.20/0.30 (M2); distillate compositions are: 0.93/0.02/0.05 (M1), 0.997/0.0015/0.0015 (M2); bottoms compositions are: 0.02/0.17/0.81 (M1), 0.30/0.28/0.42 (M2). Thermodynamic model employed for both mixtures is NRTL for liquid phase, while ideality is considered in vapor phase (according to results of a previous contribution[16]), and operation pressure is 14.7 psia. The feed stream is introduced as saturated liquid. On the other hand, these designs were optimized using a multiobjective genetic algorithm with constraints[12], coupled to Aspen Plus; so, all optimal designs are rigorous. As parameters of the evolutionary strategy, we selected 30 generations of 250 individuals each one. The resulting designs of this procedure were compared with those obtained by the short methodology.

4. Analysis of results

The proposed methodology and the evolutionary strategy were applied to the cases described in the previous section. From here, we called short design to that obtained from the proposed methodology, and evolutionary design to that obtained from the stochastic strategy. Also, it is worth of mention that the number of stages is counted from top to bottom, and since total condensers are considered, they are not counted as a stage. The mixture M1 has just one distillation boundary in the composition space, and the separation is performed in the left region, Figure 2. From Table 1 for mixture M1, we observe that the short design is quite similar to the evolutionary design, as in the structure of the column as in the reflux ratio value. In the short design, we calculate the minimum reflux ratio, and we used the heuristic of 1.1 times the minimal value to fix the operation reflux ratio. In the evolutionary design, there were no restrictions with respect to the value of the variables, or even heuristic rules are not considered. We also can observe that the purities are virtually reached in both designs; however, a little less energy is required in the short design.

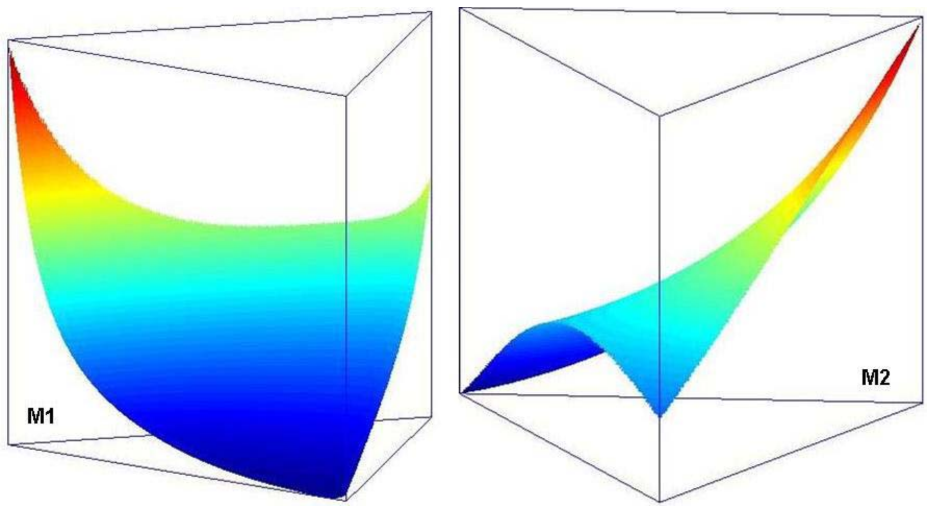

Figure 2. Boiling temperature surfaces for mixtures M1 and M2

Table 1. Resulting designs for both mixtures with the short and evolutionary strategies

	M1 acetone-isopropanol-water						M2 acetone-chloroform-benzene					
	R	N_F	N	X_D	X_B	Q, kW	R	N_F	N	X_D	X_B	Q, kW
Short design	0.96	18	13	0.92	0.02	534.8	2.75	49	46	0.997	0.29	404.3
Evolutionary design	0.99	17	11	0.93	0.02	541.6	2.78	49	46	0.997	0.30	407.5

In the second case, the mixture M2 has a boundary distillation with a pronounced curvature, as can be seen in Figure 2. From Table 1 for mixture M2, we observe that the short design is virtually the same that the evolutionary design, especially in the structure of the column, with a slightly difference in the reflux ratio value. In a similar way to the first case, in the short design we calculate the minimum reflux ratio, and we used the heuristic of 1.1 times the minimal value to fix the operation reflux ratio. In the evolutionary design, there were no restrictions with respect to the value of the variables, or even heuristic rules are not considered. We also can observe that the purities are virtually reached in both designs; however, a little less energy is required in the short design.

Then, it can be observed that the designs obtained with the short procedure are optimal, since they are quite similar to those obtained though a constrained mixed integer non linear and rigorous optimization strategy. This optimality is due to the location of the feed stage according to the concept of minimum difference in composition, minimizing at the same time the disturbance. Also, the simple criterion of the second derivate helps to eliminate the pinch zones that could appear. The most important aspect is that all these operations are made using just the operation profiles. Moreover, the algorithm to calculate the minimum reflux ratio, that uses operation profiles, can be used in multicomponent mixtures; additionally, it is useful to verify the feasibility in multicomponent mixtures, since if the intersection of profiles does not occur, then the separation is no feasible.

5. Conclusions

A short and optimal design procedure for azeotropic distillation columns has been presented. The procedure considers algebraic material balances, which are solved from outside to inside the column, satisfying the global material balance. Even when the intersection of composition profiles is verified in order to guarantees the feasibility, it is not used to locate the feed stage. The feed stage is located minimizing the composition difference between the stages of each section and the feed stream. In this way, the feed is better located, generating fewer disturbances, and making the design more efficient. Also, we propose an analytical procedure to calculate the reflux ratio in multicomponent mixtures that uses just the operation profiles; additionally, this algorithm could be useful to verify the feasibility in multicomponent mixtures, since if the intersection of profiles does not occur, then the separation is no feasible. Moreover, the procedure includes the simple criterion of the second derivate to eliminate pinch zones, in case that there exists. Finally, it is important to mention that the resulting designs from the short method are located in an optimal operation zone, since that they are so similar to the designs obtained with the evolutionary strategy.

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