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Surrogate Model-Based Optimization of Pressure-Swing Distillation Sequences with Variable Feed Composition

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ABSTRACT

Pressure-swing distillation (PSD) is a frequently applied method to separate pressure-sensitive azeotropic mixtures; however, its energy demand is very high. In continuous mode, PSD is performed in a system consisting of a high- and a low-pressure column. If the composition of the feed is between the azeotropic compositions at the two pressures, it can be introduced into any of the columns, leading to two possible column sequences. Depending on the feed composition, one of the sequences is optimal whether in terms of energy demand or total annual cost (TAC). In the present work, surrogate model-based optimization is applied to determine the optimal TAC value as a function of the feed composition between the azeotropic ones. As a first step, the column sequence with feeding into the high-pressure column is studied here. The mixture to be separated consists of water and ethylenediamine, which form a maximum-boiling azeotrope. The columns are modeled separately and a large number of simulations are performed in points generated by Latin hypercube sampling, then, algebraic surrogate models are fitted to the simulation results. The resulting surrogate optimization problem is solved for different feed composition values and the accuracy of the models is verified by rigorous simulations, as well. The energy cost and TAC has a maximum as a function of the composition. The error of the estimation of TAC remains under 6 % except at the lowest water concentration studied.

Keywords: Distillation, Machine Learning, Modelling and Simulations, Optimization, Surrogate Model

INTRODUCTION

For separating azeotropic mixtures, special distillation methods must be used, such as pressure-swing (PSD), extractive or heteroazeotropic distillation. The advantage of PSD is that it does not require the addition of a new component. However, it can only be applied if the azeotrope is pressure-sensitive, and its energy demand can also be high. The configuration of the system depends on not only the type of the homoazeotrope but also the feed composition (z). If z is between the azeotropic compositions at the pressures of the columns, the feed can be introduced into either the low- (LP-HP sequence) or the high-pressure column (HP-LP sequence). Depending on the value of z, one of the sequences will be optimal, whether with respect to energy demand or total annual cost (TAC).

Optimization of PSD systems is typically performed either by local methods (e.g. [1]) that might fail in finding the global optimum or by global, evolutionary algorithms (e.g. genetic algorithm, GA), which, however, require a large number of simulations. A potential approach to reduce the computational intensity is surrogate model-based optimization (SMBO), where the objective function is estimated with surrogate models fitted to the results of simulations. To the best of our knowledge, SMBO was only applied to optimize PSD systems by Bubel et al. [2] and Karaman et al. [3].

Bubel et al. [2] minimized the total energy demand of the separation of the maximum-boiling azeotropic mixture acetone-chloroform. They applied artificial neural networks as surrogate models, but did not consider the geometrical parameters (numbers of trays, feed locations) as optimization variables, only the operational ones. Two SMBO approaches were tested: fitting the models to individual columns or to the whole flow-sheets, with the first approach showing better results.

Karaman et al. [3] studied the separation of a maximum-boiling azeotropic mixture water-ethylenediamine by PSD, where z (35 mol% water) was between the azeotropes at 0.1 and 2.02 bar. The TAC of both sequences was minimized without and with heat integration. The LP-HP sequence was more favorable at this composition. The optimization was performed by two methods: a GA and SMBO. By SMBO, algebraic surrogate models were fitted to simulation results of the whole flow-sheet by the ALAMO software [4] and the resulting optimization problem was solved. Different decomposition techniques were tested with the models fitted (1) to elements of TAC (heat duty of LPC, column diameters), (2) to capital and energy costs or (3) to TAC itself. The best results were achieved with the highest level of decomposition. Although the TAC obtained by SMBO was lower than that of GA only once, the difference was always within 5 %.

In this work, our aim is to (1) improve the accuracy of surrogate models, thus, the performance of SMBO and (2) study the influence of z on the optimum of the HP-LP sequence, using the case study of Karaman et al. [3]. The first goal is achieved by fitting the models to the results of the single columns instead of the two-column system. Achieving the second goal requires repeated optimization at different feed compositions, which would be very time-consuming with conventional optimization methods. However, an advantage of SMBO is that z can be included as an input variable of the models. This enables quickly finding the optimum for any feed composition. By performing the optimization for the LP-HP in a future work, it will be possible to determine for both column sequences the range of z where the given sequence is optimal.

The novelty of our work consists of determining the optimal PSD system for the first time as a function of the feed composition by SMBO. Additionally, this is the first work that uses ALAMO to fit the models to be used in the optimization to the individual columns since Karaman et al. [3] fitted them to the two-column system. While Bubel et al. [2] did the model fitting to the individual columns, they only considered operational parameters as optimization variables.

PROCESS DESCRIPTION

Water(A) and ethylenediamine(B) form a maximum-boiling azeotrope below 4-4.5 bar. The azeotrope is pressure-sensitive, with its A content decreasing from 44.1 mol% at 0.10 bar to 25.6 % at 2.02 bar. The phase equilibrium calculations were performed by UNIQUAC [3].

The flow rate of the feed is F=100 kmol/h; its pressure and temperature are 2.02 bar and 46.85 °C, respectively. Its composition is assumed to be able to vary roughly between the two azeotropic compositions: $0.27 \leq z \leq 0.44$. Both components must be recovered with a purity of 99.5 %.

The feed is introduced into either the high- (HPC, 2.02 bar) or the low-pressure column (LPC, 0.101 bar), leading to the two column sequences HP-LP and LP-HP (Figure 1). Both columns have a pressure drop of 2.74 mbar/tray. (Note that Karaman et al. [3] used constant pressure drop values of 0.2407 and 0.0492 bar for HPC and LPC, respectively.) A is obtained as the distillate of HPC (D_{HP}). The bottom product's composition is close to the azeotropic one at the column pressure, and it is fed into LPC, where B is produced as distillate (D_{LP}). The bottom product is then led to HPC. The distillate flow rates can be calculated from the material balances written for the feed and the distillates as linear functions of z:

$$D_{HP} = F - D_{LP} \tag{1}$$

$$D_{LP} = F \frac{x_{DHP} - z}{x_{DHP} - x_{DLP}} \tag{2}$$

According to the purity specifications, the concentration A in D_{HP} and D_{LP} is x_{DHP}=0.995 and x_{DLP}=0.005, respectively.

In the present work, since only the HP-LP sequence is studied, the bottom product of LPC can be considered as a recycle stream ($F_{rec}=W_{LP}$).

CALCULATION METHOD

Simulation of the individual columns or the whole PSD process is performed with the professional flowsheet simulator CHEMCAD 7.

The total annual cost (TAC) is calculated from the total capital (TCC) and total energy cost (TEC) [1, 5]:

$$TAC = \frac{TCC}{PBP} + TEC \tag{3}$$

where PBP=3 y is the payback period. The capital cost includes the cost of the columns, condensers and reboilers. The energy cost is the cost of the low- and middle-pressure heating steam used to heat the reboiler of LPC and HPC, respectively. A detailed description of the cost calculation is given in Ferchichi et al. [5].

Apart from the numbers of theoretical trays (N_{HP}, N_{LP}), which are input parameters, the following results are needed for the calculation of TAC: the column diameters (Di_{HP}, Di_{LP}), the condenser (Q_{cHP}, Q_{cLP}) and reboiler heat duties (Q_{rHP}, Q_{rLP}) and temperature of the bottom products (T_{WHP} and T_{WLP}). (The temperatures of the distillates are also required to calculate the condenser areas, but they are constant due to the specified purities.) Therefore, surrogate models are fitted to the above results to calculate an estimation of TAC.

Contrary to our previous work [3], the two columns are modelled separately for the fitting of the surrogate models. An overview of the surrogate model system is

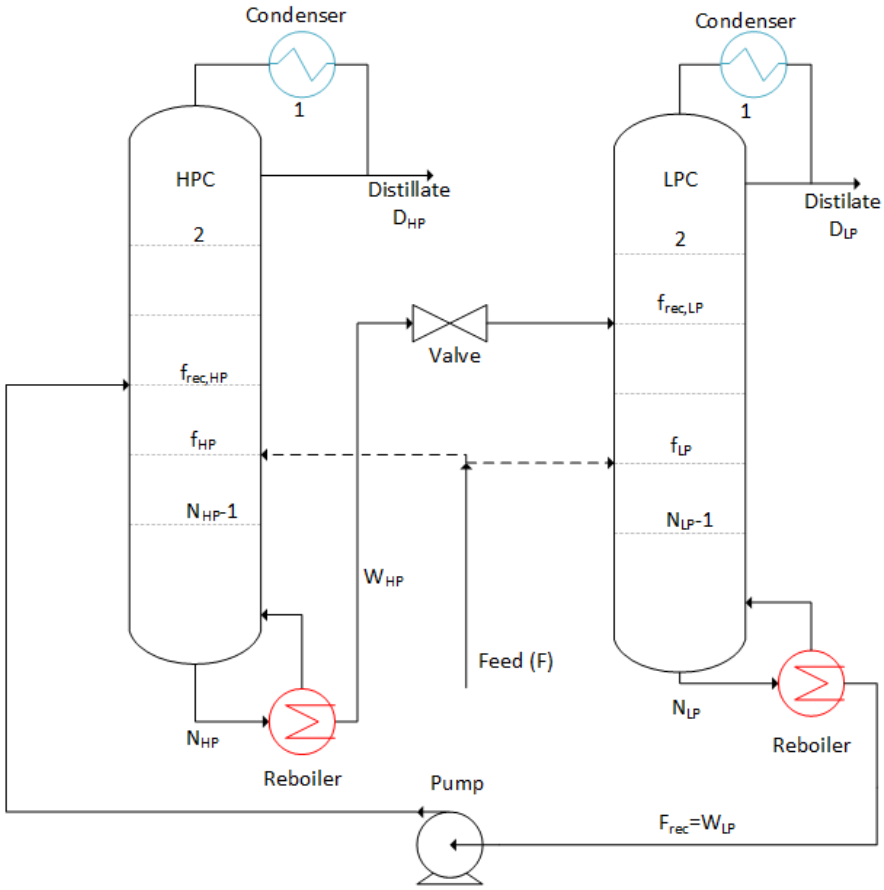

Figure 1: Common flow-sheet of the column sequences HP-LP and LP-HP. Depending on the sequence, the feed is either introduced into HPC or LPC (shown by dashed line)

shown in Figure 2. Symbols with * denote values calculated from surrogate models. The input variables for both columns are the number of trays, F_{rec}, the composition of the bottom product of LPC (x_{WLP}) and z. The flow rate (W_{HP}) and composition (x_{WHP}) of the feed of LPC are calculated by material balances from F_{rec}, x_{WLP} and D_{LP}. For both columns, 500-500 points are generated in the four-dimensional space of the input variables by using Latin hypercube sampling (LHS). The ranges of the input variables are given in Table 1. In each point, the column is simulated with the distillate composition and bottom product flow rate as column specifications. For the given input variables, the optimal feed locations are determined by minimizing the reboiler heat duty. Initial locations are set as $f_{rec}=0.1324 \cdot N_{HP}$, $f_{HP}=0.8971 \cdot N_{HP}$, and $f_{rec,LP}=0.2778 \cdot N_{LP}$ based on the optimal values of Karaman et al. [3]. The feed locations are then varied (in the case of HPC, one by one) until reaching the minimum of the heat duty. This procedure guarantees optimal feed locations while eliminating them as input variables, thereby reducing the dimensionality of the surrogate models.

By using ALAMO, algebraic surrogate models are fitted to Q_{rHP}, Di_{HP}, Q_{cLP}, Q_{rLP} and Di_{LP} as functions of the four input variables by minimizing the Bayesian information criterion or, in the case of the mean square error. ALAMO is permitted to select, apart from a constant value, linear, exponential, logarithmic and polynomial functions of the variables, as well as different powers of the binary or tertiary products and ratios of the variables. Models of the diameters are forced to be integer or half-integer values in feet, as CHEMCAD calculates discrete values of diameters. Q_{cHP} is expressed as a linear function of Q_{rHP}. In order to be able to estimate the bottom product temperatures of the columns, it is also necessary to describe them by surrogate models. 400 boiling point values are generated by combining 20 pressure and 20 water concentration values. The pressure range is determined by calculating the minimum and maximum possible pressure drops from the range of N used. The concentration ranges used are $0.26 \leq x_{WHP} \leq 0.34$ and $0.40 \leq x_{WLP} \leq 0.44$. ALAMO is then used to fit models to the boiling points.

With the surrogate models, it is possible to calculate TAC*, an estimation of TAC, and minimize it for different values of z. The optimization problem can be written as:

$$\min_{N_{HP},N_{LP},F_{rec},x_{WLP}} TAC^*(N_{HP},N_{LP},F_{rec},x_{WLP},z) \qquad (4)$$

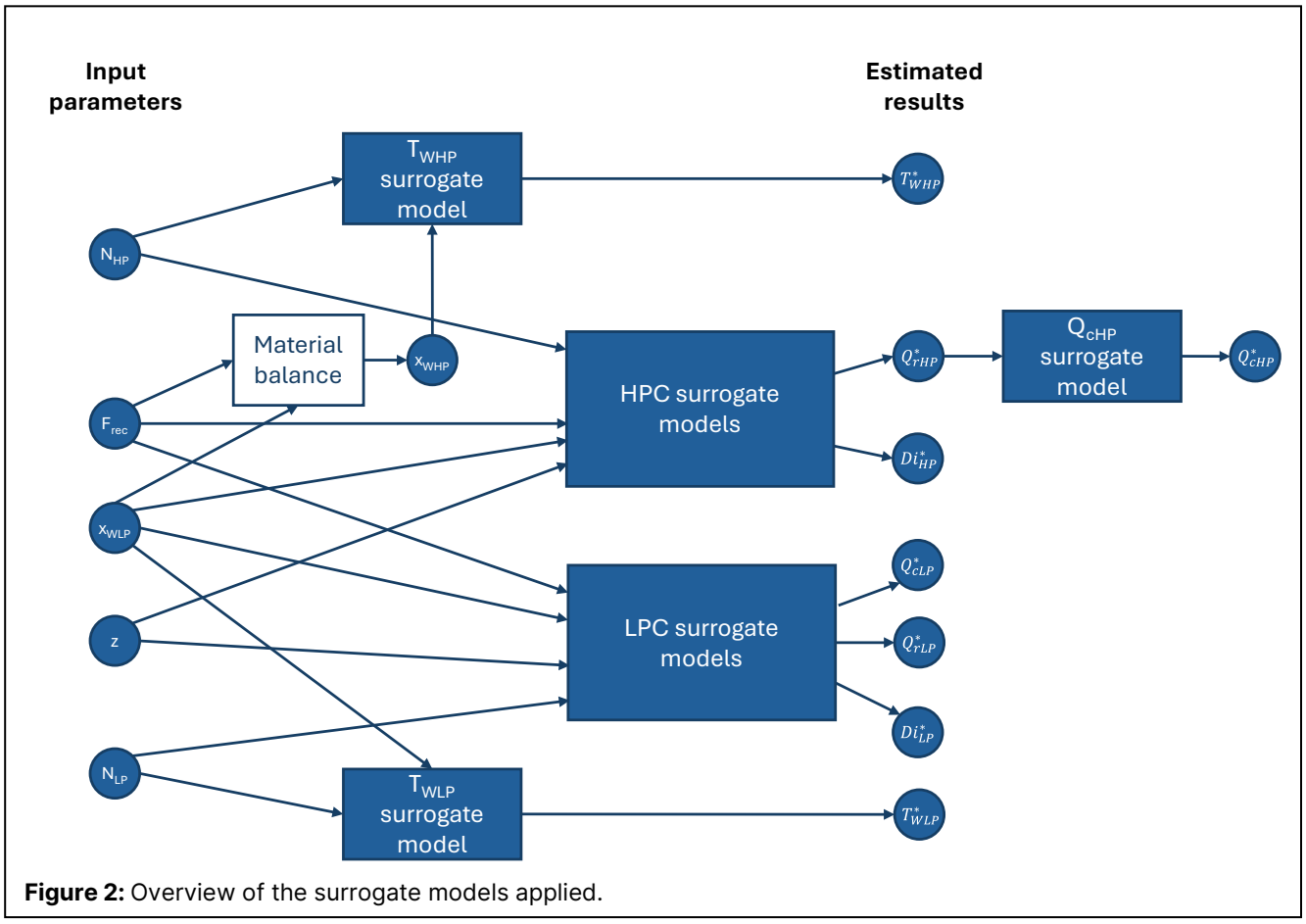

Figure 2: Overview of the surrogate models applied.

Table 1: Ranges of the input variables used for surrogate model fitting.

Variable	Minimum	Maximum
N_{HP}	50	100
N_{LP}	15	60
F_{rec}, kmol/h	80	220
x_{WLP}	0.42	0.44
z	0.27	0.43

The only constraints of the problem are the lower and upper bounds of the variables (Table 1) to avoid extrapolation with the surrogate models. The optimization problem is solved by sequential quadratic programming in Maple.

After solving the surrogate optimization problem, it is necessary to check the results by creating a three-dimensional plot of TAC* as the function of two variables. This can also help with the identification of local optima, but its most important aim is the following. It often occurs that the value of one or more variables is on the lower or upper bound. This might mean that the (constrained) optimum is truly on said bound, indicating that it is necessary to extend the range of the variable as the true optimum lies outside the current range. However, this can also be an artefact of the surrogate models, which can have a large error close to the bounds, where the concentration of simulation points is lower. If the direction of the errors improves the objective function, an erroneous optimum can be found. This can be prevented by the above graphical analysis and, if necessary, the application of a constraint to exclude the problematic region.

RESULTS

Karaman et al. [3] optimized the process at z=0.35 both by using a GA and SMBO (Table 2). The SMBO process was followed by determining the optimal feeding locations, as well. Note that the results are not completely comparable to the ones obtained here since they used constant pressure drop values.

In the present work, out of the 500 simulations, 408 converged in the case of HPC and 264 in the case of LPC. The size (number of elements), the adjusted coefficients of determination ($\bar{R}^2$) and root mean square error (RMSE) of the models are given in Table 3.

The linear model of Q_{cHP} and those of the bottom temperatures have a very good fit. The $\bar{R}^2$ values of the other heat duties are relatively high, as well. The least good fit occurs by the column diameters. However, for the majority of the data points, the error of the diameter is only one or two sizes.

Table 2: Optimization results of Karaman et al. [3] for z=0.35.

	GA	SMBO
N_{LP}	68	72
F_{rec}, kmol/h	161.1	184.8
N_{LP}	36	35
x_{WLP}	0.435	0.424
TCC, 10^6 $	1.901	1.992
TEC, 10^6 $/y	1.989	2.045
TAC, 10^6 $/y	2.622	2.709

Table 3: Ranges of the input variables used for surrogate model fitting.

Variable	Model size	$\bar{R}^2$	RMSE
Q_{cHP}	2	0.998	296 MJ/h
Q_{rHP}	37	0.962	1100 MJ/h
Di_{HP}	21	0.679	0.122 m
Q_{cLP}	24	0.909	980 MJ/h
Q_{rLP}	26	0.896	981 MJ/h
Di_{LP}	11	0.635	0.177 m
T_{WHP}	7	1.00	$2.93 \cdot 10^{-4}$ °C
T_{WLP}	13	1.00	$6.34 \cdot 10^{-5}$ °C

The optimization problem is solved for 9 different z values. Table 4 shows the optimal values of the optimization variables. N_{HP} and F_{rec} do not show a monotonous variation; their values are notably higher at z≥0.35. F_{rec} gradually decreases above this value, but both variables clearly show a decrease when approaching z=0.43. N_{LP} always equals 25 except for z=0.35. It must be noted that it was necessary to use a constraint of N_{LP}≥20 to avoid false optima at 15, the lower bound N_{LP}. x_{WLP} also reached its lower bound at z≤0.29 and z≥0.37. However, this seems not to be due to a particularly high error of TAC*, indicating that it would be necessary to decrease the lower bound of x_{WLP} when performing LHS. As shown in Figure 3 for z=0.35, it seems that two local optima exist, one at x_{WLP}=0.42 and another around x_{WLP}≈0.43. One of these is found at every z value.

Table 4: Optimal values of the optimization variables as a function of z, determined by SMBO.

z	N_{HP}	F_{rec}, kmol/h	N_{LP}	x_{WLP}
0.27	68	156.8	25	0.42
0.29	69	155.4	25	0.42
0.31	65	148.8	25	0.4296
0.33	65	152.8	25	0.4296
0.35	75	189.9	26	0.4295
0.37	75	187.4	25	0.42
0.39	76	179.9	25	0.42
0.41	71	169.4	25	0.42
0.43	67	150.1	25	0.42

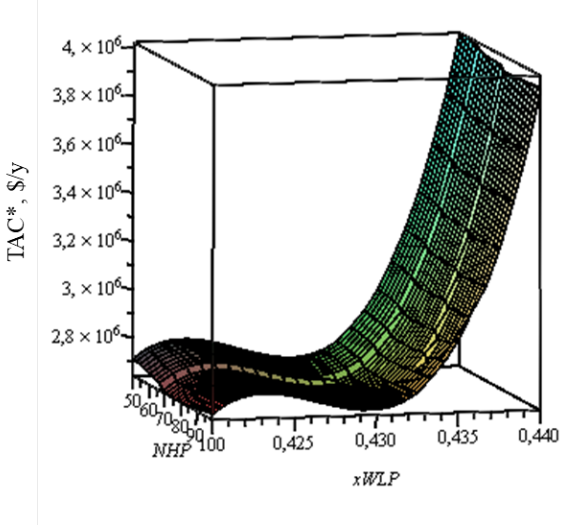

Figure 3. TAC* as a function of N_{HP} and x_{WLP} at z=0.35, F_{rec}=189.9 kmol/h and N_{LP}=26.

Figure 4 shows the costs calculated with the surrogate models in the optima given in Table 4 with dashed curves. At each z value, a simulation is also performed with the optimal values, and the costs are also calculated from the simulation results (shown with continuous curves). TCC* has a maximum as a function of z at z=0.35, but it hardly changes between 0.35 and 0.41. TEC* also has a maximum, but at a much higher value: z=0.41. Below z=0.33, a considerable increase can be observed on the increase of z. Due to the small changes in TCC*, the behavior of TAC* is similar to that of TEC* with a maximum at z=0.41.

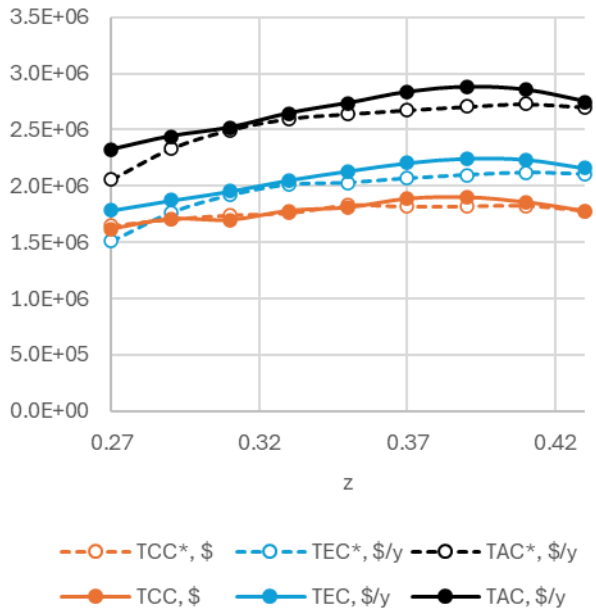

Figure 4. Estimated and rigorously calculated costs in the determined optima as functions of z.

The rigorously calculated TCC also shows a limited variation only, mostly explained by changes in Di_{HP}, and a maximum of z=0.39. Both TEC and TAC behave similarly to their estimated values, but their maxima are more pronounced and occur at a lower z value, also at z=0.39. The maximum in TEC, and thus in TAC, can be explained by the fact as z increases, more distillate has to be produced in HPC and its heat duty increases, while that of LPC decreases. It must be noted that these TAC values are probably not optimal, only approximations of the optima.

Comparing the estimated and rigorously calculated cost, it can be seen that the error of TCC is relatively low at all z values, with a maximum of 4 % at z=0.39. The error of TEC is low around z=0.32 and 0.43, and it is always below 7 % except at z=0.27 (15 %). The accuracy of TAC is similar to that of TEC. Its error is 11 % at z=0.27, but lower than 6 % otherwise. TEC and TCC are always underestimated by the surrogate models, but this is the case by TCC.

The values of N_{HP} and F_{rec} determined by Karaman et al. [3] by SMBO (Table 2) are close to the ones obtained here, while the values by GA are similar to those calculated for z ≤0.33. N_{LP} is considerably higher by both methods. The x_{WLP} value obtained here is between that of GA and SMBO. Karaman et al. [3] achieved a TAC lower by 1.0 % by SMBO and 4.2 % by GA. The error of TAC estimated by the surrogate models was 3.7 % compared to 3.5 % in this work.

CONCLUSIONS

The separation of maximum-boiling azeotropic mixture, water-ethylenediamine by pressure-swing distillation (PSD) was studied. The feed composition (z) was between the azeotropic compositions at the pressures of the columns. From the two possible column sequences, the HP-LP one is studied, where the feed is introduced into the high-pressure column (HPC). The total annual cost (TAC) of the PSD system was previously minimized by Karaman et al. [3] for a given feed composition (z=35 mol% water) using a genetic algorithm and surrogate-model based optimization (SMBO). By the latter method, surrogate models were fitted to a large number of simulation results of the two-column system.

In the present work, SMBO was applied by fitting the models to the results of the individual columns. Another novelty was that z was now included as an input variable of the models. The other input variables were the numbers of trays of the columns and the molar flow rate and water content of the bottom product of the recycle stream, that is, the bottom product of the low-pressure column. Using the surrogate models, it was possible to estimate TAC. The resulting surrogate optimization was solved for different values of z. The accuracy of the estimated TAC was verified by rigorous simulations, as well. The energy cost (TEC) and TAC had a maximum as a function of the composition. The capital cost was estimated with a maximal error of 4 %. The errors were higher for TEC and TAC. In the case of TAC, the error of the estimation of TAC remained under 6 % except at the lowest water concentration studied. The minimal TAC at z=35 % found in this work is only by 1 % higher than the SMBO result of Karaman et al. [3] and 4 % than the optimal value found by the genetic algorithm. In conclusion, the accuracy of the SMBO method is acceptable.

In the future, the SMBO of the LP-HP sequence will be performed, as well. This will make it possible to determine for both column sequences the range of z where the given sequence is optimal.

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