

ENTROPY PRODUCTION MINIMIZATION IN A MULTICOMPONENT DIABATIC DISTILLATION COLUMN

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This work presents a procedure for direct numerical minimization of entropy production in a diabatic tray column with heat exchanged on the trays as control variables, as opposed to previously used procedures with temperature on the trays as control variables. The procedure, which had previously been demonstrated on a binary mixture, was in this work applied to a multicomponent mixture, with minor modifications. The procedure comprised the Complex optimization method and the Ishii-Otto method for solving the equations of a column model based on the iterative Newton-Raphson technique with partial linearization of the equations. The desired separation of the components was realized by the addition of a penalty function to the goal function, i.e. entropy production in the column. The required thermodynamic characteristics were calculated by the Soave equation of state. As an illustration, an industrial debutanizer with 5 components was used whose data, obtained by simulation, were compared with the optimization results of a diabatic column with the same desired separation and number of trays. After the diabatic column optimization procedure, the value of $91.91 \text{ JK}^{-1}\text{s}^{-1}$ was obtained as the best result for entropy production. According to the best solution, entropy production in the diabatic column was 23.2% lower than in the adiabatic column. The heat to be removed from the column increased by 24.7%, while the heat to be added to the column increased by 28.8%.

Key words: entropy production, multicomponent diabatic distillation, optimization

1. Introduction

Distillation is the most widely used separation method in chemical industry, based on the difference in volatility of the components that need to be separated. This process is characterised by low energy efficiency, i.e. high degradation of heat energy. In a standard distillation column heat is added in the reboiler at the highest temperature and removed from the condenser at the lowest temperature relative to the temperatures in the column. Low energy efficiency of classical distillation initiated research on different ways of enhancing it. One of the possibilities is the so-called *diabatic distillation*. In a diabatic column heat is added along the stripping section, i.e. on each stripping section tray, and removed along the rectifying section, i.e. on each rectifying section tray, if it is a tray column. As the temperature on the

stripping section trays is lower than the reboiler temperature (it decreases starting from the reboiler toward the feed tray), in a diabatic column heat is added at lower temperatures. Additionally, since the temperature on the rectifying section trays is higher than the condenser temperature (it increases starting from the condenser toward the feed tray), in a diabatic column heat is removed at a higher temperature than in classical (adiabatic) distillation. This results in decreased entropy production and exergy loss, *i.e.* in increased distillation efficiency in accordance with the Second Law of Thermodynamics. However, diabatic distillation requires a higher quantity of exchanged heat than classical distillation and a significantly more complex column design to enable the required heat exchange on all trays and adjustment to variable flows of the vapor and liquid phases on the trays. Namely, in classical distillation the vapor and liquid phase flows on the trays are approximately constant in both the rectifying and stripping sections (but with different values). In diabatic distillation, the vapor phase flow increases from the reboiler to the feed tray, and then decreases to the first tray, while the liquid phase flow increases from the first tray to the feed tray, and then decreases to the reboiler. With an increase in the number of trays and with heat exchanged on each tray, entropy production decreases; consequently, with an infinite number of trays a reversible column is obtained [1], which is of purely theoretical importance.

A more practical approach is entropy production minimization in a column with a finite number of trays, *i.e.* the determination of the optimal exchanged heat profile. Significant attention has been devoted to this problem and the following methods have been proposed: *Equipartition of entropy production* [2], *Equipartition of forces* (EoF) [3, 4], *Equal thermodynamic distance* (ETD) [5, 6], Equal quantities of heat removed from each rectifying section tray and equal quantities of heat added to each stripping section tray [7], Direct numerical optimization by different methods [8-10].

Theoretical methods have been presented only for binary columns and they represent an approximation of the optimal profile of the exchanged heat. The results of the application of these methods and their mutual comparison have been presented in several papers, among others in [8-12, 14-17, 19, 20]. Most direct numerical optimization procedures use tray temperatures as control variables and their application is also limited to binary columns. In paper [18] a new procedure of direct numerical optimization was developed, which uses the exchanged heat quantities as control variables. It was successfully applied to a frequently used example of distillation of a binary mixture of benzene and toluol. Out of five optimization methods used, the *Simplex* method proved to be the most advantageous. In this work, this procedure was applied to a multicomponent mixture using an industrial debutanizer, with minor modifications.

2. Problem setting

The determination of the exchanged heat quantities on the trays (profiles) of a diabatic distillation column giving minimal entropy production in the column presents an optimization problem in the presence of limitations, where the goal function is entropy production, and the limitations are nonlinear equations of the column model and the desired separation, namely the purity of the products. Additional limitations are also possible, depending on the problem type. Solving the problem formulated in this way requires complex optimization methods, with a high number of iterations and unwarranted convergence. Namely, due to the emphasized nonlinearity of the column model equations, solving them is rather

complex, as testified by a number of specifically developed methods. The optimization problem can be set up differently, so that the quantities (temperatures and component separation) needed for the calculation of the goal function are obtained by solving a mathematical model of the column for an ongoing set of heats exchanged on the trays. The required separation may be provided by the addition of a penalty function to the goal function. This allows for the application of simpler optimization methods or some of the more common and tried methods for solving the model of a distillation column. In this work the latter approach was applied.

3. Model of a multicomponent separation column with ideal trays

A simple distillation column with trays intended for the separation of one feed stream into two products – the distillate and the bottom product – and equipped with a total condenser for the vapor flow from the column top and a reboiler for partial evaporation of the liquid flow from the column bottom, is shown in fig. 1. One part of the condensate is returned as reflux, while the other part is removed as the distillate. An analogous diabatic column with heat exchange (via the heat exchangers) on every tray is shown in fig. 2. This column does not use reflux from the condenser; instead, everything is removed as the distillate. A diabatic column with sequential heat exchangers is shown in fig. 3. This column uses one flow of the cooling fluid for the rectifying section, and one flow of the heating fluid for the stripping section.

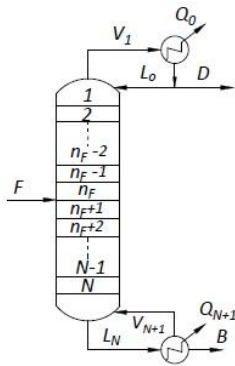


Figure 1. Simple column

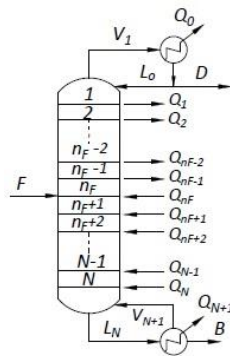


Figure 2. Diabatic column

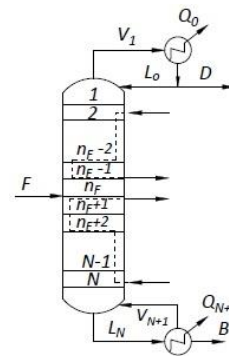


Figure 3. Diabatic column with sequential heat exchangers

In this work a general model was used of a complex column with ideal trays for multicomponent mixture separation based on the vapor-liquid equilibrium, *i.e.* on different volatilities, suitable for the calculation of both distillation and absorption columns. The tray of such a column is schematically presented in fig. 4. The model allows for the feed mixture to be added to each tray, the vapor and liquid side products to be removed, and heat to be exchanged using an appropriate heat exchanger. It is assumed that the tray is ideal, *i.e.* that the vapor and liquid phases are in equilibrium.

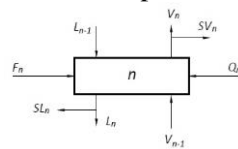


Figure 4. An equilibrium tray model

The mathematical model of a column with such trays is described by the standard MESH equation system: Total material balance equations:

$$\text{The condenser: } V_1 - L_0 - SL_0 - V_0 = 0$$

$$\text{The trays } F_n + L_{n-1} + V_{n+1} - L_n - SL_n - V_n - SV_n = 0 \quad n \in [1, N]$$

$$\text{The reboiler } L_N - L_{N+1} - V_{N+1} = 0 \quad (1)$$

where $SL_0 = D$ (distillate) and $L_{N+1} = B$ (bottom product).

Material balance for the components:

$$V_1 y_{k,1} - (L_0 + SL_0) x_{k,0} - V_0 y_{k,0} = 0$$

$$F_n z_{k,n} + L_{n-1} x_{k,n-1} + V_{n+1} y_{k,n+1} - (L_n + SL_n) x_{k,n} - (V_n + SV_n) y_{k,n} \quad n \in [1, N]$$

$$L_N x_{k,N} - L_{N+1} x_{k,N+1} - V_{N+1} y_{k,N+1} = 0 \quad k \in [1, NK] \quad (2)$$

$$\text{Equation of the equilibrium: } y_{k,n} = K_{k,n} x_{k,n} \quad k \in [1, NK] \quad n \in [0, N+1] \quad (3)$$

Summation equations of molar fractions of the components:

$$\sum_{k=1}^{NK} x_{k,n} = 1 \quad \text{or} \quad \sum_{k=1}^{NK} y_{k,n} = 1 \quad n \in [0, N+1] \quad (4)$$

$$\text{Energy balance: } V_1 H_1^V + Q_0 - (L_0 + SL_0) H_0^L - V_0 H_0^V = 0$$

$$F_n H_n^F + L_{n-1} H_{n-1}^L + V_{n+1} H_{n+1}^V + Q_n - (L_n + SL_n) H_n^L - (V_n + SV_n) H_n^V = 0 \quad n \in [1, N]$$

$$L_N H_N^L + Q_{N+1} - L_{N+1} H_{N+1}^L - V_{N+1} H_{N+1}^V = 0 \quad (5)$$

The total produced entropy in the column is:

$$\frac{dS^{irr}}{dt} = L_{N+1} S_{N+1}^L + SL_0 S_0^L + V_0 S_0^V + \sum_{n=1}^N (SL_n S_n^L + SV_n S_n^V - F_n S_n^F) - \sum_{n=0}^{N+1} \frac{Q_n}{T_n} \quad (6)$$

The last term in Eq. (6) represents an approximation of entropy produced in the heat exchangers, *i.e.* the entropy change of the working fluids (heating and cooling). This change may be expressed in the following way [21, 22]:

$$\left(\frac{dS^{irr}}{dt} \right)_{\text{HEX}} = - \sum_{n=0}^{N+1} \frac{Q_n}{T_n^U} \quad (7)$$

For the determination of the optimal distribution of the exchangers' area, the authors [15] used the

$$\text{following expression: } \left(\frac{dS^{irr}}{dt} \right)_{\text{HEX}} = \sum_{n=0}^{N+1} \left(- \frac{Q_n}{T_n^U} + Q_n X_n \right) \quad (8)$$

$$\text{Where the mean driving force is modelled in the following way: } X_n = \frac{\delta}{\lambda_n T_n^2} \cdot \frac{Q_n}{A_n} \quad (9)$$

If the exchangers' area is calculated afterward based on the heat to be exchanged, the entropy produced in the exchangers is easier to express in the following manner:

$$\left(\frac{dS^{irr}}{dt}\right)_{\text{HEX}} = \sum_{n=0}^{N+1} U_n (S_{n,\text{out}}^{\text{U}} - S_{n,\text{in}}^{\text{U}}) \quad (10)$$

In the case of sequential heat exchangers, where only one flow of the cooling fluid is used for cooling and only one flow of the heating fluid for heating, Eq. (10) is simplified:

$$\left(\frac{dS^{irr}}{dt}\right)_{\text{HEX}} = U_{\text{cool}} (S_{\text{cool},\text{out}}^{\text{U}} - S_{\text{cool},\text{in}}^{\text{U}}) + U_{\text{heat}} (S_{\text{heat},\text{out}}^{\text{U}} - S_{\text{heat},\text{in}}^{\text{U}}) \quad (11)$$

The required data for the cooling and heating fluids may be calculated based on the heat to be exchanged, as follows:

$$U_{\text{cool}} = \frac{\sum_{n=0}^{N+1} Q_{n,\text{cool}}}{H_{\text{cool},\text{out}}^{\text{U}} - H_{\text{cool},\text{in}}^{\text{U}}} \quad (12), \quad U_{\text{heat}} = \frac{\sum_{n=0}^{N+1} Q_{n,\text{heat}}}{H_{\text{heat},\text{out}}^{\text{U}} - H_{\text{heat},\text{in}}^{\text{U}}} \quad (13)$$

The inlet and outlet temperatures of the working fluids may be estimated based on empirical experience of heat exchanger design and the required temperatures in the condenser and the last tray of the rectifying column section.

It was demonstrated in work [23] that for a column (depropanizer) similar to the column investigated in this work, approximately equal values of entropy production are obtained, calculated using the entropy balance (6), exergy analysis, and far more complex and challenging non-equilibrium thermodynamics. In this work entropy production was used as the goal function and was calculated using eq. (6).

To solve the system of equations (1)-(5), which consists of $(2NK+3)(N+2)$ non-linear equations, it is necessary to assign the flows, the compositions and enthalpies of the feed streams, the flows of side products, the exchanged heat on the trays, and the pressure. The pressure on each tray may be calculated based on known or assumed pressure drops on the tray, in the condenser, and in the reboiler. If pressure drops are small compared with the pressure, it may be assumed that the pressure is constant in the whole column. The unknown variables to be determined are the flows and compositions of the vapor and liquid flows and temperatures on the trays (and in the condenser and reboiler). Their total number is also $(2NK+3)(N+2)$. Since the system of equations for the separation column's mathematical model is non-linear, it must be solved iteratively. There are a large number of methods and variations – general or specific to some of column classes such as absorber, reboiled absorber, rectification column and distillation column. In this paper, as well as in the previous one [18], the global *Ishii-Otto* method [24] was used in order to simulate all column classes, *i.e.* both adiabatic and analogous diabatic distillation columns. In the simulation of an adiabatic distillation column, flow V_0 (0 if the condenser is total) and the reflux rate are assigned. The values of flows L_0 and V_1 are thus fixed, and instead of them heat quantities to be exchanged in the condenser and the reboiler are determined as unknowns. According to the *Ishii-Otto* method, the condenser and the reboiler are seen as ideal trays. There is no reflux from the condenser in a diabatic column, so the calculation of the heat to be removed from the condenser, which is fixed by the composition of the vapor from the column top, may be done after solving the mathematical model of the column. Therefore, the calculation of a diabatic distillation column belongs to the class of reboiled

absorber calculations. With the reboiled absorber the vapor flow is assigned from the top of the column, and instead of it the heat to be added in the reboiler is calculated.

For the calculation of thermodynamic properties – vapor-liquid equilibrium constants, enthalpy and entropy – the SRK equation of state was used [25], and the characteristics of the components were taken from the literature [26].

The entropy of the real-fluid mixture was calculated as follows:

$$S = \sum_{k=1}^{NK} n_k \left[S_k^{*,0}(T^0, P^0) + \int_{T_0}^T \frac{C_{p,k}^*(T)}{T} dT - R \ln \frac{P}{P^0} \right] - R \sum_{k=1}^{NK} n_k \ln(n_k) + \Delta S \quad (14)$$

The ideal gas enthalpy at $T^0 = 298\text{K}$ and $P^0 = 101.325 \text{ kPa}$ was calculated by *Thinh's* method of group contributions [26]:

$$S_k^{*,0}(T^0, P^0) = \sum_{j=1}^{NG} \nu_j \Delta S_{s,j} \quad (15)$$

For the calculation of the heat capacity of an ideal gas at constant pressure, polynomial dependence on temperature was used with the coefficients taken from the literature [26]:

$$C_{p,k}^*(T) = a_k^* + b_k^* \cdot T + c_k^* \cdot T^2 + d_k^* \cdot T^3 \quad (16)$$

The departure function for entropy was calculated by the Soave equation of state [25], as follows:

$$\Delta S = \frac{1}{b} \cdot \frac{\partial a}{\partial T} \ln \left(1 + \frac{b \cdot P}{z_s \cdot R \cdot T} \right) + R \cdot \ln \left(z_s - \frac{b \cdot P}{z_s \cdot R \cdot T} \right) \quad (17)$$

The system of equations (1)-(5) was solved for different reflux values in order to determine the reflux required to achieve the desired separation of the components in an adiabatic column.

For the determination of the optimal profile of the exchanged heat on the trays in a diabatic column, the total entropy produced in the column was used as the goal function, with the addition of a penalty function aimed at securing the desired separation of the components:

$$f = \frac{dS^{irr}}{dt} + PN_D (x_{TK,0} - x_{TK,0}^{det}) + PN_B (x_{LK,N+1} - x_{LK,N+1}^{det}) \quad (18)$$

$$\text{where } PN_D = \begin{cases} 10000, & \text{if } \mathbf{x}_{TK,0} - \mathbf{x}_{TK,0}^{det} > 0, \\ 0, & \text{if } \mathbf{x}_{TK,0} - \mathbf{x}_{TK,0}^{det} \leq 0 \end{cases} \quad PN_B = \begin{cases} 10000, & \text{if } \mathbf{x}_{LK,N+1} - \mathbf{x}_{LK,N+1}^{det} > 0, \\ 0, & \text{if } \mathbf{x}_{LK,N+1} - \mathbf{x}_{LK,N+1}^{det} \leq 0. \end{cases}$$

For the minimization of the goal function (18), the *Complex* method [27] was applied. This method falls into the category of direct methods, since it requires the knowledge of the function's value, but not the knowledge of partial derivatives' value. In this case, the latter value cannot be determined analytically, and numerical calculation would be very time-consuming. In the previous work [18], several methods were applied for the minimization of produced entropy in a binary diabatic tray column, of which the *Simplex* method showed the best results. Since a high number of local minimums with close values were observed, the *Complex* method was applied in this work, as it better covers the space of potential solutions by random choice of the points of the initial complex. This method was defined for optimization in the presence of explicit and implicit limitations in the form of inequalities on the variables' values, which are in this case heats exchanged on the trays. However, for the problem considered the limitations on these heats are not needed, so it is also possible to solve it by methods of unconditional optimization. In order to

observe the *Complex* method requirements, it is possible to artificially define lower and upper limitations, *i.e.* intervals of the values of heat exchanged on the trays (explicit limitations), so that they are never exceeded. Nevertheless, because of the initial points these intervals should be as realistic as possible, since the initial points are chosen by random numbers within the intervals. Unrealistic values of exchanged heat lead to divergence during the column operation simulation or require more iterations for the determination of the optimal profile of exchanged heat. The amounts of heat exchanged in the condenser and reboiler of a starting adiabatic column serve as orientation for the intervals' values. In a diabatic column most of the heat is exchanged on the trays at the column's ends and around the feed tray.

4. Calculation results and discussion

For the demonstration of direct optimization application for the determination of the optimal profile of the exchanged heat on the trays of a multicomponent diabatic column, an industrial debutanizer was used from a natural gas separation plant. There were 35 trays in the column, 19 of which were in the rectifying section, equipped with the condenser and reboiler. It operated at 690 kPa. A feed mixture with the following design composition (molar fractions): 0.0114 propane, 0.2912 isobutane, 0.313 n-butane, 0.1424 2-methylbutane, and 0.242 n-pentane was introduced at the temperature of 345 K. The feed mixture flow was 5540.3 kmol h⁻¹, and the reflux ratio 2.15. The required separation was defined through the molar fraction of 2-methylbutane in the distillate which must not be higher than 0.004, and the molar fraction of n-butane in the bottom product which must not be higher than 0.032. The distillate temperature was 327.7 K, and the bottom product temperature 372.2 K. For calculation purposes, it was assumed, using empirical correlations, that there were 23 theoretical trays in the column and that the 13th one was the feed tray. For the flow the value of 25 mol s⁻¹ was adopted. For the given conditions of composition, temperature and pressure, the vapor phase fraction in the feed mixture of 0.2676 was calculated using the *Soave equation of state*. Using the trial and error method it was found that the required separation with an adopted number of theoretical trays of the adiabatic column was achieved at the reflux ratio of 1.677, with the condenser and reboiler observed as ideal trays. The obtained distillate temperature (temperature inside the condenser) was 328.3 K and the bottom product temperature (temperature inside the reboiler) 374.1 K. The heat quantities exchanged in the condenser and the reboiler were 745802 and 637948 J s⁻¹, respectively, whereby the goal function value, *i.e.* entropy production was 119.68 JK⁻¹s⁻¹.

After multiple repetitions of the diabatic column optimization procedure, the value of 91.91 JK⁻¹s⁻¹ was obtained as the best result for entropy production. The profiles of the temperature, vapor and liquid flows, exchanged heat quantity and produced entropy for the adiabatic and diabatic columns are given in figures 5-9. These figures show the characteristic differences that are the same as in the case of binary mixture distillation, *i.e.* the temperature profile (curve) is steeper in diabatic distillation and the vapor and liquid flows change along the rectifying and stripping sections of the diabatic column, while they are approximately constant in the adiabatic column. Additionally, the produced entropy in the adiabatic column is high at the column's ends and low in the middle part, in comparison to the diabatic column. According to the best solution, entropy production in the diabatic column was 23.2% lower than in the adiabatic column. However, the heat to be removed from the column increased to 930108 J s⁻¹, which represents an increase of 24.7%, while the heat to be added to the column increased to 821944 J s⁻¹, which

represents an increase of 28.8%. When the values of several best solutions for the exchanged heat profiles in the diabatic column given in tab. 1 are compared, it can be seen that the required quantities of exchanged heat decrease with increased goal function. For example, for entropy production of $92.73 \text{ JK}^{-1}\text{s}^{-1}$, which represents a reduction of 22.5% compared to adiabatic distillation, the heat to be removed from the column is 870964 Js^{-1} , which represents an increase of 16.8%; the heat to be added is 762800 Js^{-1} , which represents an increase of 19.6%, compared to adiabatic distillation. Based on the aforementioned results, it may be concluded that a decrease in entropy production requires a disproportionate increase in the exchanged heat, *i.e.* that obtaining the cooling fluid at a higher (more usable) temperature requires supplying additional heat. This applies to the heat pump too, except that the heat pump, which increases the cooling fluid temperature from the condenser, is probably simpler than diabatic column design.

In paper [28] the addition of only one heat exchanger for the decrease in entropy production was considered, and it was concluded that an increase in the heat to be added to the column may be eliminated by adding several trays. The trends of the reduction of entropy production and the reduction of the exchanged heat with increased tray numbers can be observed based on the results in tab. 2, obtained when the number of trays in the diabatic column was increased by two and four, with the best three solutions given for each number of trays. It is noticeable that entropy reduction with the same number of trays requires an increase in exchanged heat. Based on the optimization results it may be concluded that the reduction of entropy production using a diabatic column requires an increase in operating and/or investment costs compared to an adiabatic column, which makes a self-standing diabatic column an inappropriate solution for efficiency increase in accordance with the Second Law of Thermodynamics. The heat integration of the rectifying and stripping sections (HIDiC) requires heat integration with other devices and processes or further diabatic column upgrade by the addition of new devices (compressors).

An increase in the total annual cost (TAC) was determined during partial or total column diabatization, with a fixed number of trays, in two of the three cases considered in paper [7]. A decrease in the TAC of the diabatic column compared with the adiabatic column was obtained for a system with high relative volatility of the key components. Besides entropy production, *i.e.* the number and size of additional heat exchangers, the TAC is affected by the choice of working fluids.

Based on entropy production on the feed tray of the diabatic column shown in fig. 9 and the molar fractions of the light key component in the bottom product given in tab. 2, which are better than required, it may be concluded that the feed tray in a diabatic column should not be the same as the one in an adiabatic column.

"Classical" numerical optimization methods – either direct ones, which only use the function goal values, or indirect ones, which also use the values of the second and/or first partial derivatives – are not efficient for the determination of the global optimum in the presence of a high number of local optimums. This is true of the applied direct *Complex* method of Box as well, despite it being the most suitable method because it starts from multiple points, *i.e.* sets of independent variables, obtained by random choice within the allowed area. In order to increase the probability of finding the global optimum, it would be necessary to apply methods that examine the area of possible solutions more systematically, *i.e.* meta-heuristic methods such as the method of simulated hardening (cooling) and its stochastic and deterministic modifications and genetic algorithms. The disadvantage of these methods is a large number of iterations

and dependence on the problem, since most of them encompass adjustable parameters for which the right choice of values is key to the efficiency of the method in a specific case. Examples of the application of these methods in distillation are recovery and minimization of the irreversibility index [29] and exergoeconomic optimization of distillation column sequencing [30]. The global optimum for the problem considered in this paper is not of practical interest because the exchanged heat disproportionately increases with the reduction in entropy production, which can be concluded by the analysis of the solutions, *i.e.* optimal values of local minima obtained.

The number of the *Complex* method iterations ranged from about 500 to about 2000, depending on the initial points.

Table 1. Entropy generation and exchanged heat for several best solutions

Quantity	Adiabatic distillation	Diabatic distillation			
		1	2	3	4
Entropy production [$\text{JK}^{-1}\text{s}^{-1}$]	119.68	91.91	92.15	92.31	92.7
Removed heat [Js^{-1}]	745802	930108	925442	897666	870964
Added heat [Js^{-1}]	637948	821944	817279	789504	762800
Condenser [Js^{-1}]	745802	278917	278913	278914	278915
Reboiler [Js^{-1}]	637948	289932	295028	329543	306506
Molar fraction of 2-methylbutane in the distillate	0.003997	0.004001	0.003991	0.004000	0.004000
Molar fraction of n-butane in the bottom product	0.031988	0.031665	0.031665	0.031693	0.031658

Table 2. Entropy production and exchanged heat quantities for three best solutions with different tray numbers in the diabatic column

Quantity	Number of trays in the diabatic column								
	24			26			28		
Entropy production [$\text{JK}^{-1}\text{s}^{-1}$]	91.91	92.15	92.31	82.97	83.05	84.18	75.36	75.52	75.57
Removed heat [Js^{-1}]	930108	925442	897666	925125	891234	876330	865444	853055	834740
Added heat [Js^{-1}]	821944	817279	789504	816982	783093	768184	757324	744935	726622
Condenser [Js^{-1}]	278917	278913	278914	278903	278903	278907	278895	278892	278894
Reboiler [Js^{-1}]	289932	295028	329543	235695	270960	211737	216814	191590	223581
Molar fraction of 2-methylbutane in the distillate	0.00400	0.00399	0.00400	0.00400	0.00400	0.00400	0.00400	0.00399	0.00400
Molar fraction of n-butane in the bottom product	0.03167	0.03167	0.03169	0.03185	0.03188	0.03192	0.03200	0.03202	0.03200

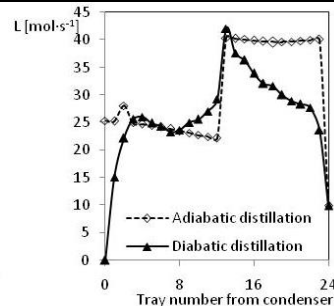
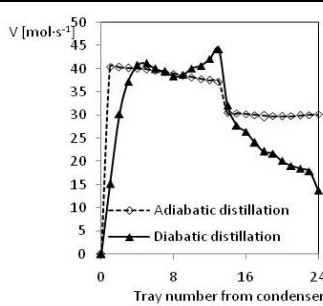
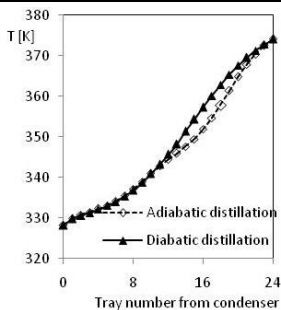


Figure 5. Temperature profile

Figure 6. Vapor flow profile Figure 7. Liquid flow profile

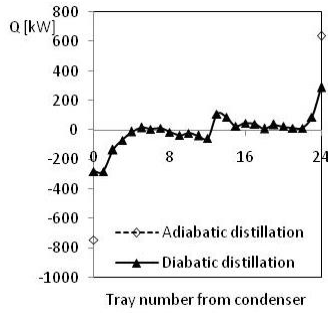


Figure 8. Exchanged heat profile

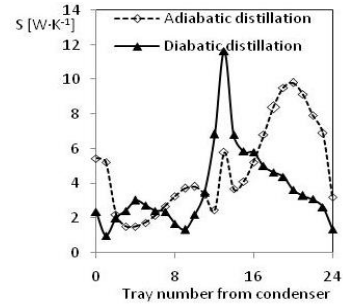


Figure 9. Produced entropy profile

5. Conclusion

The previously developed procedure for the determination of the optimal profile of exchanged heat by direct numerical minimization of entropy production in a diabatic tray column, where the exchanged heats are also independent variables, has been successfully applied to the distillation of a multicomponent mixture in a five-component industrial debutanizer.

The applied procedure enables unconditional optimization since the values (of temperature) needed for the calculation of the goal function, *i.e.* entropy production, are obtained by solving the mathematical model of the column, and the desired separation, defined by allowed molar fractions of the key components, is achieved by adding a penalty term to the goal function. Since the needed values are obtained by solving the column model, numerous methods can be applied. In this work, as well as in the work relating to the binary column, the *Ishii-Otto* method of partial linearization was used. Due to a high number of local minima, the *Complex* method was applied for optimization because it starts from multiple points obtained by random choice in the "allowed area", *i.e.* the area of exchanged heat quantities on trays of practical interest. To increase the probability of finding the global minimum, one of meta-heuristic methods would need to be applied. However, that would be of no practical interest since it was determined on the considered example of an industrial debutanizer that with the reduction in entropy production (for the best result – 23.2%) the exchanged heat in the diabatic column disproportionately increases (the heat to be removed from the column increased by 24.7%, while the heat to be added to the column increased by 28.8%) and is significantly higher than in the adiabatic column. Based on the results, it can be concluded that the regular diabatic column is not an energy efficient solution for irreversibility reduction in the case of the freestanding column. Heat integration with other processes (if they are available), or a higher degree of heat integration inside the column (HIDiC) is necessary.

6. Nomenclature

A – Area for heat exchange, [m ²]	PN – Penalty number, [–]
a – Parameter in the Soave equation of state [kPa·m ⁶ ·kmol ⁻²]	Q – Exchanged heat flow, [J·s ⁻¹]
B – Bottom product, [mol·s ⁻¹]	R – Universal gas constant, R = 8.3143 [J·mol ⁻¹ ·K ⁻¹]
b – Parameter in the Soave equation of state, [m ³ ·kmol ⁻¹]	S – Entropy, [J·mol ⁻¹ ·K ⁻¹]
C _p – Heat capacity at constant pressure, [J·mol ⁻¹ ·K ⁻¹]	ΔS – Residual entropy, [J·mol ⁻¹ ·K ⁻¹]
D – Distillate flow, [mol·s ⁻¹]	dS ^{irr} dt ⁻¹ – Total entropy production, [J·s ⁻¹ ·K ⁻¹]

F – Feed mixture flow, [mol·s ⁻¹]	SL – Liquid side stream flow, [mol·s ⁻¹]
f – Goal function, [J·s ⁻¹ ·K ⁻¹]	SV – Vapor side stream flow, [mol·s ⁻¹]
H – Enthalpy, [J·mol ⁻¹]	T – Temperature, [K]
K – Vapor-liquid equilibrium constant, [-]	U – Working fluid flow, [mol·s ⁻¹]
L – Liquid stream flow, [mol·s ⁻¹]	V – Vapor stream flow, [mol·s ⁻¹]
N – Total tray number, [-]	X – Driving force for heat transfer in an exchanger, [K ⁻¹]
NG – Total number of different kinds of functional groups in a molecule, [-]	x – Component molar fraction in liquid stream, [-]
NK – Total number of the components, [-]	y – Component molar fraction in vapor stream, [-]
n – Molar number of component, [-]	z – Component molar fraction in feed mixture, [-]
P – Pressure, [kPa]	zs – Compressibility factor, [-]

Greek letters	Subscripts
Δ_s – Total contribution for entropy, [J·mol ⁻¹ ·K ⁻¹]	B – Bottom product
δ – Film thickness, [m]	D – Distillate
λ – Heat conductivity, [W·m ⁻¹ ·K ⁻¹]	F – Feed mixture
ν – Number of functional groups of the same kind in a molecule, [-]	j – Functional group order number
Superscripts	k – Component order number
B – Bottom	LK – Light key component
D – Distillate	TK – Heavy key component
det – determined	n – Tray order number
F – Feed mixture	N – Last tray
L – Liquid	N+1 – Reboiler
U – Working fluid	S – Entropy
V – Vapor	0 – Condenser
0 – Standard state	1 – First tray

7. References

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