A NEW APPROACH TO ENTROPY PRODUCTION MINIMIZATION IN DIABATIC DISTILLATION COLUMN WITH TRAYS

by

Momčilo Dj. SPASOJEVIĆ [a], ***Milovan R. JANKOVIĆ*** [b], ***and Damir D. DJAKOVIĆ*** [a*]

[a] Faculty of Technical Sciences, University of Novi Sad, Novi Sad, Serbia
[b] Faculty of Technology, University of Novi Sad, Novi Sad, Serbia

Original scientific paper
UDC: 66.045.1:66.021.3.048:519.876.5
DOI: 10.2298/TSCI1002317S

Previous approach to direct numerical minimization of entropy production in diabatic distillation column in order to determine heat quantity to be exchanged at trays was based on temperatures on trays as control variables and it was applied only to simple binary columns. Also, previously developed theoretical models for determining optimal exchanged heat profile were determined only at such columns and while they were approximated they produced worse results than numerical minimum of entropy production. In this paper, as control variables for minimization, exchanged heat on the trays is used. It enables application to complex multicomponent diabatic columns. Ishii-Otto global method, based on model linearization and iterative solution by Newton-Raphson technique, is applied for solving column mathematical model. Needed thermodynamical properties for ideal systems are calculated using Lewis-Randall ideal solution model, and for non-ideal slightly polar systems they are calculated using Soave equation of state. Five direct methods are used for numerical optimization. Applied approach is successfully demonstrated at frequently used example of distillation of benzene and toluol mixture by using for these purposes specially written program. Simplex method appeared to be the most convenient optimization method for the considered problem.

Key words: *entropy production, diabatic distillation, optimization*

Introduction

Distillation is still the most frequently used separation technique although it has small energy efficiency. During distillation, heat energy degrades and heat from higher temperature level shifts to a lower one while some fuel is spent in order to bring heat energy to a needed level. In the last 50 years, efforts have been made to increase distillation energy efficiency through heat integration of both other processes (exchangers network, coupled columns) and the distillation column by itself (heat pumps, heat integrated distillation column – HIDIC). In the last 35 years, a so called diabatic distillation has been investigated as one of possibilities. In traditional (adiabatic) distillation column with trays, heat is introduced in a reboiler, and removed in a con-

* Corresponding author, e-mail: djakovic@uns.ac.rs

denser while in a diabatic column, heat is exchanged at every tray (it is introduced in a stripping and removed in a rectifying section) by independent (intermediate heat exchanger for every tray) or sequential exchangers (with an exchanger for the stripping and rectifying sections) in order to minimize entropy production and exergy loss, respectively, which maximizes efficiency in accordance with the Second Law of Thermodynamics. Borderline case of diabatic distillation is reversible distillation (with a continual heat exchange) which is described, *e. g.* in [1-3]. Such column has infinite length (tray number) and is only of theoretical interest. More practical approach is addition of one or several exchangers in the column in order to decrease irreversibility. The effects and conditions of application of limited number of exchangers are investigated, among others, in [4-9], by the use of different methods. Minimization of entropy production in diabatic column with the exchangers at every tray has been more intensively investigated in the last 15 years and following methods are suggested:

– equipartition of entropy production [10],
– equipartition of forces (EoF) [5, 11],
– equal thermodynamic distance (ETD) [12, 13], and
– direct numerical optimization by different methods.

Equal distribution of driving forces is theoretically derived method which has arisen as the expansion of method of equal distribution of entropy production on irreversible thermodynamics basis. This method proposes that gaseous and liquid stream exchanging heat and mass on a column tray should work with uniform driving force through interfacial area. Equal thermodynamic distance method, also theoretically derived for binary distillation, orders that for every pair of consecutive trays there should be equal distance regarding Weinhold metrics [14] in the space of thermodynamic variables. The results of application of suggested methods and their mutual comparison are given in several papers, among others, in [15-25].

Previous investigations and application of suggested methods, both theoretical and numerical, have been limited to simple binary columns. Theoretical methods give only approximation of optimal exchanged heat profile, *i. e.*, they are more inferior than numerical optimization.

Problem statement

Distillation in simple binary columns represents only small share of distillation in total, so there is a need to develop a procedure for entropy production minimization, which should be applied at complex multicomponent diabatic distillation columns. Previous approach of minimization was based on temperatures as control variables. This approach can be applied to complex columns in two ways. The first way is that minimization of entropy production represents optimization in presence of limits given by equations of mathematical model of the column, except equations of energy balance from which needed quantities of exchanged heat are calculated. This problem is difficult to solve because of expressed non-linearity and narrow range of variables in which it is possible to calculate needed fluid characteristics. It also requires complex sophisticated optimization methods. In the second way, solving procedure of column model the equations, except energy balance equations, will be done in order to determine quantities which are needed for goal function calculation – entropy production. This will enable application of simpler optimization methods but it will also demand development of new algorithm for model equations solving.

In this paper, the third approach is applied to control variables for optimization. Instead of temperatures exchanged heat quantities at the trays are used as control variables. This choice enables application of simpler direct optimization methods, and for solving the whole column model, it is possible to apply some of numerous well known and checked methods. Column models, solution, and optimization approaches in previous use, and also in new suggested procedure are given.

A model of simple binary column with ideal trays

Simple adiabatic binary column with ideal trays and total condenser, shown in fig. 1, serves for binary mixture separation and includes feed mixture (F) and two products – distillate (D) and bottom products (B). The column is equipped with a condenser at the top (total) and a reboiler at the bottom. An analogous diabatic column with the exchanger at every tray is shown in fig. 2. Although a reflux into the column does not come back from the condenser at the top, it serves only for vapor condensation from the top into distillate.

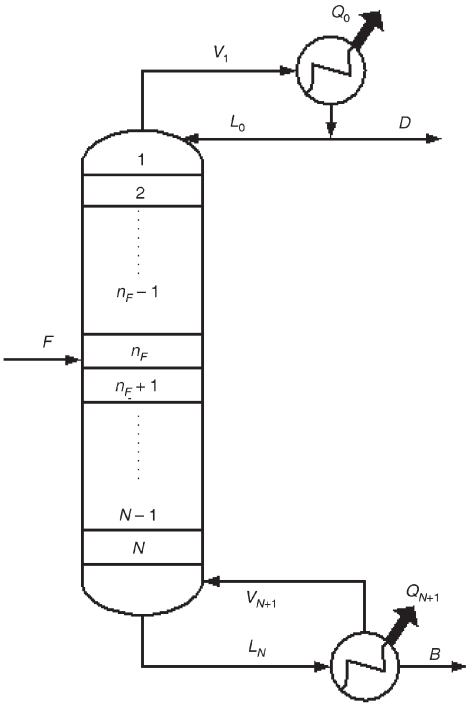

Figure 1. Simple adiabatic column with total condenser

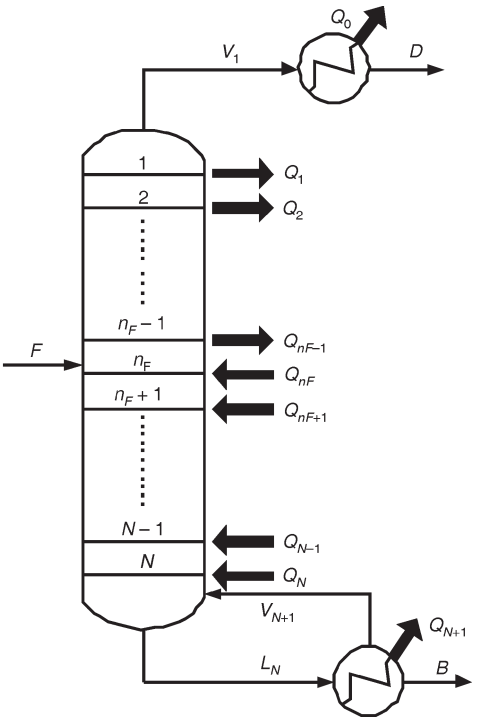

Figure 2. Simple diabatic column with total condenser

Diabatic column can be described by following equations of material and energy balances and using the equations of equilibrium and summation:

$$F = D + B \tag{1}$$

$$V_{n\ 1} \quad L_n \quad \begin{matrix} D & n & [0, n_F \quad 1] \\ B & n & [n_F, N] \end{matrix} \tag{2}$$

$$Fz_k \quad Dx_k^{\mathrm{D}} \quad Bx_k^{\mathrm{B}} \quad k \quad [1,2] \tag{3}$$

$$V_{n\ 1} y_{k\,n\ 1} \quad L_n x_{k\,n} \quad \begin{matrix} Dx_k^{\mathrm{D}} & n & [0, n_F \quad 1] \\ Bx_k^{\mathrm{B}} & n & [n_F, N] \end{matrix} \quad k \quad [1,\ 2] \tag{4}$$

$$Q_0 \quad D(H^{\mathrm{D}} \quad H_1^{\mathrm{V}}) \tag{5}$$

$$Q_n \quad V_n H_n^{\mathrm{V}} \quad L_n H_n^{\mathrm{L}} \quad V_{n\ 1} H_{n\ 1}^{\mathrm{V}} \quad L_{n\ 1} H_{n\ 1}^{\mathrm{L}} \quad \chi \quad n \quad [1,\ 2] \tag{6}$$

$$\chi \quad \begin{matrix} FH^{\mathrm{F}} & n \quad n_F \\ 0 & \text{otherwise} \end{matrix}$$

$$Q_{N\ 1} \quad V_{N\ 1} H_{N\ 1}^{\mathrm{V}} \quad BH^{\mathrm{B}} \quad L_N H_N^{\mathrm{L}} \tag{7}$$

$$y_{\mathrm{k\,n}} = K_{\mathrm{k\,n}} x_{\mathrm{k\,n}} \quad k \quad [1,\ 2] \quad n \quad [1,\ N+1] \tag{8}$$

$$\sum_{k\ 1}^{2} x_{\mathrm{k\,n}} \quad 1 \quad \sum_{k\ 1}^{2} y_{\mathrm{k\,n}} \quad 1 \quad n \quad [1,\ N \quad 1] \tag{9}$$

Entropy production in the condenser, column trays, and the reboiler is:

$$\frac{\mathrm{d}S^{\mathrm{irr}}}{\mathrm{d}t}_{0} \quad DS^{\mathrm{D}} \quad V_1 S_1^{\mathrm{V}} \quad \frac{Q_0}{T_0} \tag{10}$$

$$\frac{\mathrm{d}S^{\mathrm{irr}}}{\mathrm{d}t}_{n} \quad V_n S_n^{V} \quad L_n S_n^{\mathrm{L}} \quad V_{n\ 1} S_{n\ 1}^{\mathrm{V}} \quad L_{n\ 1} S_{n\ 1}^{\mathrm{L}} \quad \frac{Q_{\mathrm{n}}}{T_{\mathrm{n}}} \quad \kappa \quad n \quad [1, N] \tag{11}$$

$$\kappa \quad \begin{matrix} FS^{\mathrm{F}} & n \quad n_F \\ 0 & \text{otherwise} \end{matrix}$$

$$\frac{\mathrm{d}S^{\mathrm{irr}}}{\mathrm{d}t}_{N\ 1} \quad V_{N\ 1} S_{N\ 1}^{\mathrm{V}} \quad BS^{\mathrm{B}} \quad L_{\mathrm{N}} S_N^{\mathrm{L}} \quad \frac{Q_{N\ 1}}{T_{N\ 1}} \tag{12}$$

On the basis of expressions (10)-(12), total entropy production in the column is:

$$\frac{\mathrm{d}S^{\mathrm{irr}}}{\mathrm{d}t} \quad \sum_{n\ 0}^{n\ 1} \frac{\mathrm{d}S^{\mathrm{irr}}}{\mathrm{d}t}_{n} \quad DS^{\mathrm{D}} \quad BS^{\mathrm{B}} \quad FS^{\mathrm{F}} \quad \sum_{n\ 0}^{N\ 1} \frac{Q_n}{T_n} \tag{13}$$

If entropy production in the exchangers is taken into account, total entropy production will be [21]:

$$\frac{\mathrm{d}S^{\mathrm{irr}}}{\mathrm{d}t} \quad DS^{\mathrm{D}} \quad BS^{\mathrm{B}} \quad FS^{\mathrm{F}} \quad \sum_{n\ 0}^{N\ 1} \frac{Q_n}{T_n} \quad \sum_{n\ 0}^{n\ 1} Q_n X_n^{\mathrm{HX}} \tag{14}$$

Driving force for heat exchange in the exchangers, X_n^{HX}, can be approximated in the following way [21, 23, 24]:

$$X_n^{\mathrm{HX}} \quad \frac{\delta Q_n}{\lambda_n T_n^2 A_n} \tag{15}$$

Application of eqs. (14) and (15) is important when optimal distribution of exchanger area, A_n, is searched for the fixed total area of all exchangers.

In previous practice of entropy production in the column minimization, eq. (13) or eq. (14), respectively, in the case of independent exchangers at trays, temperatures from T_2 to T_{N} (T_0, T_1, and $T_{\mathrm{N+1}}$ are fixed by given composition of distillate and bottom products) are used as control variables. A solving procedure and determination of exchanged quantities of heat at trays is:

– molar fractions of heavier component are expressed from the equations of summation, eq. (9):

$$x_{2\,\mathrm{n}} = 1 - x_{1\,\mathrm{n}} \qquad y_{2\,\mathrm{n}} = 1 - y_{1\,\mathrm{n}}$$

and introduced into equilibrium, eq. (8):

$$\begin{array}{cc} y_{1n} & K_{1n}x_{1n} \\ 1 \quad y_{1n} & K_{2n}(1 \quad x_{1n}) \end{array} \tag{16}$$

Obtained system of two equations with two unknowns is linear if it is an ideal and constant vapor – liquid equilibrium, $K_{k\,n}$ is dependant only on temperature and pressure, respectively, and non-linear if the system is non-ideal and the constant is dependent both on liquid and vapor phases composition. The system is solved for all trays and in that way, vapor and liquid phase composition are determined.

– From total material balance equation, eq. (2), is expressed V_{n+1} and introduced in lighter component material balance equation, eq. (4), and then L_n, and V_{n+1} from the expression (2) are calculated for all trays.
– On the basis of known composition and temperatures vapor and liquid streams, enthalpies and then exchanged heat on the trays from energy balance equations, eqs. (5)-(7), and at the end, values of goal function, given by eqs. (13) or (14) are calculated.
– In accordance to the chosen optimization method, temperature profile on trays is changed and solving procedure is repeated until minimum goal function, given by eqs. (13) or (14) is established.

In the case of column with sequential heat exchangers, control variables are the flows and input temperatures of cold and hot fluids and exchanger surface distribution on trays. During optimization for every set of control variables, temperature profile which corresponds to a stationary state is searched [22], while part of previously described calculation for composition is used for flow and vapor and liquid streams enthalpies determination in the column. Different numerical methods are used for the optimization of diabatic column with independent and sequential heat exchangers:

– Powel method [17, 19, 20],
– optimization through small perturbations [18],
– Monte Carlo method [19, 21],
– simplex method [8, 22]

– least squares regression with Gauss-Newton procedure and one-dimensional search [23], and
– sequential quadratic programming using *fmincon* Matlab functions [21, 24, 25].

The use of temperatures on trays as control variables and described way of solving material and energy balances in order to determine optimal profile of exchanged heat restrict application to binary mixtures and simple columns only.

More logical choice is to take control variables to be the quantities of heat exchanged on trays, so for solving column model equation, some of numerous known procedures for complex multicomponent columns can be used. In that case, even choice of optimization method is not a critical one.

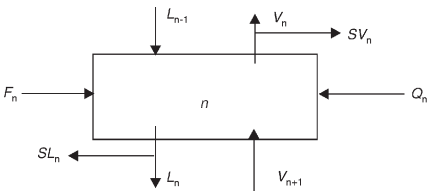

Figure 3. Equilibrium tray model

Complex multicomponent model with ideal trays

The tray of complex column for multicomponent mixture separation on the basis of vapor-liquid equilibrium is schematically shown in fig. 3. At every tray, feed mixture and removed vapor and liquid side products and exchanged heat can be introduced.

Mathematical column model with such trays is described by following equations:

– equation of total material balance

condenser $$V_1 - L_0 - SL_0 - V_0 = 0$$

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

reboiler $$L_N - L_{N+1} - V_{N+1} = 0 \tag{17}$$

where $$SL_0 = D \quad \text{and} \quad L_{N+1} = B$$

– component material balance

$$V_1 y_{k\,1} - (L_0 + SL_0)\, x_{k\,0} - V_0 y_{k\,0} = 0$$
$$F_n z_{k\,n} + L_{n-1} x_{k\,n-1} + V_{n+1} y_{k\,n+1} - (L_n + SL_n) x_{k\,n} - (V_n + SV_n) y_{k\,n} = 0 \quad n \in [1, N] \tag{18}$$
$$L_N x_{k\,N} - L_{N+1} x_{k\,N+1} - V_{N+1} y_{k\,N+1} = 0 \quad k \in [1, NK]$$

– energy balance

$$V_1 H_1^{\mathrm{V}} \quad Q_0 \quad (L_0 \quad SL_0) H_0^{\mathrm{L}} \quad V_0 H_0^{\mathrm{V}} \quad 0$$
$$F_n H_n^{\mathrm{F}} \quad L_{n\ 1} H_{n\ 1}^{\mathrm{L}} \quad V_{n\ 1} H_{n\ 1}^{\mathrm{V}} \quad Q_n \quad (L_n \quad SL_n) H_n^{\mathrm{L}} \quad (V_n \quad SV_n) H_n^{\mathrm{V}} \quad 0 \quad n \in [1, N] \tag{19}$$
$$L_N H_N^{\mathrm{L}} \quad Q_{N\ 1} \quad L_{n\ 1} H_{N\ 1}^{\mathrm{L}} \quad V_{N\ 1} H_{N\ 1}^{\mathrm{V}} \quad 0$$

– component molar fractions summation equation

$$\sum_{k\ 1}^{NK} x_{k\,n} \quad 1 \quad \text{or} \quad \sum_{k\ 1}^{NK} y_{k\,n} \quad 1 \quad n \in [0, N \quad 1] \tag{20}$$

– equilibrium equation

$$y_{k\,n} = K_{k\,n} x_{k\,n} \quad k \in [1, NK] \quad n \in [0, N+1] \tag{21}$$

Total produced entropy in column is:

$$\frac{\mathrm{d}S^{\mathrm{irr}}}{\mathrm{d}t} = L_{N+1} S^{\mathrm{L}}_{N+1} + SL_0 S^{\mathrm{L}}_0 + V_0 S^{\mathrm{V}}_0 + \sum_{n=1}^{N} (SL_n S^{\mathrm{L}}_n + SV_n S^{\mathrm{V}}_n + F_n S^{\mathrm{F}}_n) + \sum_{n=0}^{N+1} \frac{\dot{Q}_n}{T_n} \tag{22}$$

In eq. (22), a member for produced entropy in exchangers, as in eq. (14), can be added. For the system solution of $(2NK + 3)(N + 2)$ non-linear eqs. (17)-(21), the flows, compositions, feed streams enthalpies, exchanged heat on trays and the flows of all side products should be given.

It can be assumed for pressure that it is constant during the whole column or that for every tray it is calculated on the basis of known or assumed pressure drop. Number of variables to be determined, such as flows and compositions of vapor and liquid streams and temperature on trays (including condenser and reboiler), is also $(2NK + 3)(N + 2)$. There are four main classes of separation columns described by these equations, depending on specifications and these are: absorber (desorber – stripper), reboiled absorber, rectification column, and distillation column. The simplest case is absorber – column which does not have either condenser or reboiler, so the number of equations and independent variables to be determined is $N(2NK + 3)$.

For distillation column, flow V_0 (0 if the condenser is total) and reflux rate are given and that is why the values of flows L_0 and V_1 are fixed and instead of them, heat quantities to be exchanged in the condenser and reboiler as unknown are determined.

At reboiled absorber, which has no condenser, heat as an unknown quantity, which should be introduced in the reboiler, is determined instead of flow V_1, which is specified. Rectification column has condenser only and the quantity of heat, which should be removed in the condenser, is determined instead of V_0, which is specified (0 if condenser is a total one). At reboiled absorber and rectifying column, the number of independent variables and equations is $(2NK + 3)(N + 1)$. Some other specifications are possible (*e. g.*, some temperature, composition or some other property) but they are rarely used.

Because the system of separation column mathematical model equations is non-linear, it must be solved iteratively. There are a lot of methods and variations, general or specific for some of specified column classes.

At diabatic column, there is no reflux from the condenser, so the calculation of heat quantity which needs to be removed in the condenser is fixed by vapor composition from the top of the column. It is not connected with solving column mathematical model, *i. e.*, it is not determined simultaneously with other independent variables but after they have been determined.

Because of that, diabatic column calculation appertains to calculation class of reboiled absorber.

In order to simulate diabatic and analogous adiabatic column in the same method, one of general-global methods, is chosen [26]. Modifications and updates of this method allow simulation of coupled columns [27] and columns with non-ideal trays described by Murphy efficiency [28], and solution of hard high non-ideal separation problems [29, 30].

On the basis of this method, a program for produced entropy minimization given by the eq. (22) is made. The following direct methods for minimum searching are included in the program: Rosenbrock method of rotating directions [31], Hooke-Jeeves method [32], Powell method of conjugated directions [33], Simplex method [34], and Complex method [35].

Direct methods are used because they only require knowledge of function values and not the values of partial derivatives which, in this case, cannot be determined analytically and numerical calculation will be very time consuming. For thermodynamic properties calculation – vapor-liquid equilibrium constants, enthalpies, and entropies, the Soave-Redlich-Kwong (SRK) [36] is used for real systems, and for ideal systems, the procedures given in [17].

For binary column calculation, minor change to Ishii-Otto method is done because vapor temperature from the top and temperature from the first tray, respectively, is fixed by wanted distillate composition as dew point temperature. Instead of it, the unknown quantity of heat to be removed from the first tray is determined. According to this, for binary column control variables, optimization quantities of heat are Q_2, Q_3, ..., Q_N while for multicomponent column, they are Q_1, Q_2, ..., Q_N.

Results and discussion

For the new procedure of produced entropy minimization in diabatic distillation, the check is done on frequently used example of column for separation of benzene and toluol [5, 17, 18, 25]. Feed mixture with benzene molar fraction of 0.5 is introduced at the 12th tray with a flow of 1 mol/s at boiling temperature. Column is working at atmospheric pressure and has 24 trays, total condenser, and partial reboiler. Pressure drop at trays is neglected. Distillate flow is 0.5 mol/s and toluol molar fraction in distillate should not be higher than 0.05. The system is observed as the ideal one and for thermodynamic values calculation, the procedure described in [17] based on ideal solution model [37] is used, while data for components are taken from [38]. The most convenient optimization method for the observed problem has come out of the Simplex method with 370 iterations and 1090 calculations of function values following eq. (22). Minimum value of produced entropy in diabatic column is 1.056 J/sK while at adiabatic column, it is 2.998 J/sK which is a saving of 64.77%. Other results are shown graphically as the comparison of respected values of adiabatic and diabatic distillation. In fig. 4, where temperature profiles are shown, uniform temperature change at diabatic column trays can be seen, while the profile in adiabatic one is S-shaped, *i. e.*, the biggest temperature change is at trays at the end of the column.

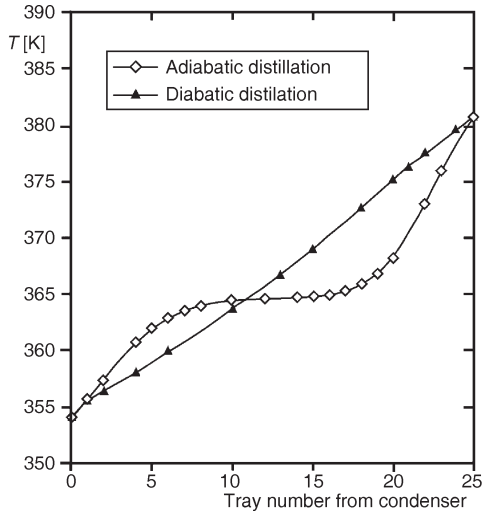

Figure 4. Temperature profile

In adiabatic column vapor and liquid phases, flows are almost constant in rectifying and stripping sections, respectively, while in diabatic column, these flows are the biggest at feed tray and they are uniformly decreased toward the end of the column as it can be seen in figs. 5 and 6. Figure 7 shows optimal profile of heat exchanged in diabatic column together with heat exchanged in condenser and reboiler of adiabatic column. Figure 8 gives produced entropy profiles calculated by eqs. (10)-(12) for both columns. Produced entropy through trays is more uniformly distributed at diabatic distillation.

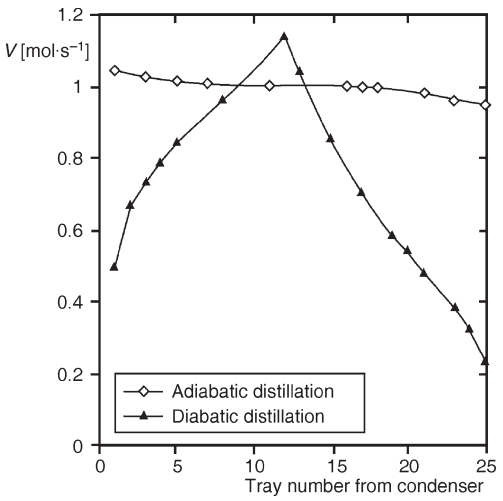

Figure 5. Vapor flow profile

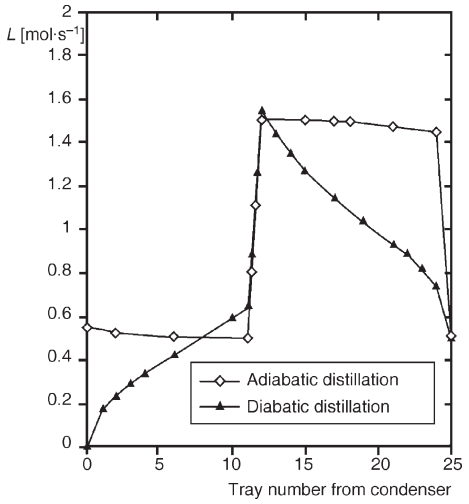

Figure 6. Liquid flow profile

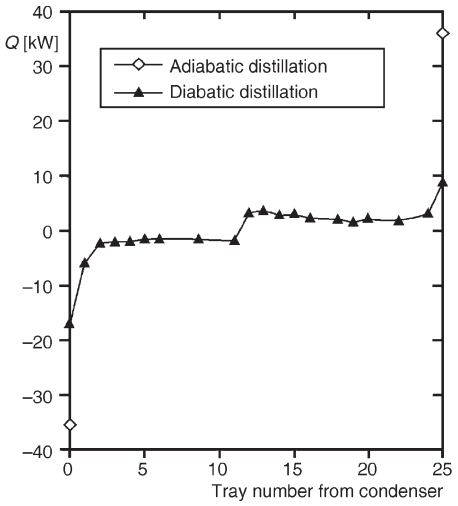

Figure 7. Heat profile

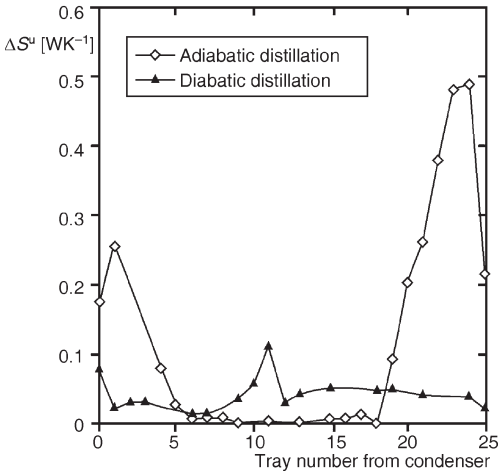

Figure 8. Total produced entropy profile

During minimization, previous notes are confirmed that goal function given by eq. (13) – for considered example eq. (22) is reduced to eq. (13) – has many local minimums with very close values. Since only direct optimization methods have been used, there is a need to repeat minimization procedure several times with different initial values of control variables in order to ensure that obtained solution is as close as possible to the global minimum.

Conclusions

The new procedure for direct numerical minimization of entropy production in diabatic column with independent heat exchangers at the trays is proposed. The backbone of this

procedure is the use of exchanged heat quantities as control variables for optimization. This approach enables the calculation of temperatures on the trays needed for calculation of goal function values – produced entropy to be done by solving mathematical model of column. For these purposes, global Ishii-Otto method based on total linearization of model equations, particularly Newton-Raphson technique, is used. For minimization, 5 well known direct optimization methods, *i. e.*, methods requiring only the knowledge of goal function values and not values of the derivatives, are used.

In the first step, the procedure is successfully applied to the example frequently used in literature in the column for separation of benzene and toluol mixture giving expected results similar to those in literature thus confirming the possibility of application to binary columns.

Nomenclature

A – heat transfer area, [m^2]
B – bottom products flow, [$mol \cdot s^{-1}$]
D – distillate flow, [$mol \cdot s^{-1}$]
F – feed mixture flow, [$mol \cdot s^{-1}$]
H – enthalpy, [$Jmol^{-1}$]
K – vapor-liquid equilibrium constant, [–]
k – component record number, [–]
L – liquid streams flow, [$mol \cdot s^{-1}$]
N – total trays number, [–]
NK – total component number, [–]
n – tray record number, [–]
n_F – feed tray record number, [–]
Q – flow of exchanged heat, [Js^{-1}]
S – entropy, [$Jmol^{-1}K^{-1}$]
dS^{irr}/dt – total entropy production, [$Js^{-1}K^{-1}$]
SL – liquid side stream flow, [$mol \cdot s^{-1}$]
SV – vapor side stream flow, [$mol \cdot s^{-1}$]
T – temperature, [K]
V – vapor streams flow, [$mol \cdot s^{-1}$]
X^{HX} – heat transfer driving force in heat exchanger, [K^{-1}]
x – component molar fraction in liquid stream, [–]
y – component molar fraction in vapor stream, [–]
z – component molar fraction in feed mixture, [–]

Greek letters

δ – film thickness, [m]
λ – heat conductivity, [$Wm^{-1}K^{-1}$]

Superscripts

B – bottom
D – distillate
F – feed mixture
L – liquid
V – vapor

Subscripts

k – component number
n – tray number
N – total tray number
0 – condenser
1 – first tray

References

[1] King, C. J., Separation Processes, McGraw-Hill, New York, USA, 1971
[2] Fonyo, Z., Thermodynamic Analysis of Rectification: 1. Reversible Model of Rectification, *International Chemical Engineering, 14* (1974), 1, pp. 18-27
[3] Kaiser, V., Gourlia, J. P., The Ideal Column Concept: Applying Exergy to Distillation, *Chemical Engineering, 92* (1985), 17, pp. 45-53
[4] Terranova, B. E., Westerberg, A. W., Temperature – Heat Diagrams for Complex Columns. 1. Intercooled/Interheated Distillation Columns, *Industrial & Engineering Chemistry Research, 28* (1989), 9, pp. 1374-1379
[5] Kjelstrup Ratkje, S., *et al.*, Analysis of Entropy Production Rates for Design of Distillation Columns, *Industrial & Engineering Chemistry Research, 34* (1995), 9, pp. 3001-3007

[6] Agrawal, R., Fidkowski, Z.T., On the Use of Intermediate Reboilers in the Rectifying Section and Condensers in the Striping Section of Distillation Column, *Industrial & Engineering Chemistry Research, 35* (1996), 8, pp. 2801-2807
[7] Aguirre, P., *et al*., Optimal Thermodynamic Approximation to Reversible Distillation by Means of Interheaters and Intercoolers, *Industrial & Engineering Chemistry Research, 36* (1997), 11, pp. 4882-4893
[8] Andersen, T. R., *et al*., Energy Efficient Distillation by Optimal Distribution of Heating and Cooling Requirements, *Proceedings* (Ed. S. Pierucci), ESCAPE-10, Pisa, Italy, 2000, pp. 709-715
[9] de Koeijer, G. M., *et al.*, Positioning Heat Exchangers in Binary Tray Distillation Using Isoforce Operation, *Energy Conversion and Management, 43* (2002), 9-12, pp. 1571-1581
[10] Tondeur, D., Kvaalen, E., Equipartition of Entropy Production. An Optimality Criterion for Transfer and Separation Processes, *Industrial & Engineering Chemistry Research, 26* (1987), 1, pp. 50-56
[11] Sauar, E., Kjelstrup Ratkje, S., Lien, K. M., Equipartition of Forces: A New Principle for Process Design and Optimization, *Industrial & Engineering Chemistry Research, 35* (1996), 11, pp. 4147-4153
[12] Salamon, P., Nulton, J. D., The Geometry of Separation Processes: A Horse-Carrot Theorem for Steady Flow Systems, *Europhysics Letters, 42* (1998), 5, pp. 571-576
[13] Andersen, B., Salamon, P., Optimal Distillation Using Thermodynamic Geometry, in: Thermodynamics of Energy Conservation and Transport (Eds. A. DeVos, S. Sieniutycz), Springer Verlag, Berlin, 2000, pp. 319-331
[14] Weinhold, F., Metric Geometry of Equilibrium Thermodynamics, *The Journal of Chemical Physics, 63* (1975), 6, pp. 2479-2483
[15] Sauar, E., *et al*., Diabatic Column Optimization Compare to Isoforce Columns, *Energy Conversion and Management, 38* (1997), 15-17, pp. 1777-1783
[16] Andresen, B., Salamon, P., Thermodynamic Geometry Determines Optimal Temperature Profile in Distillation Column, in: Methods and Applications of Inversion, Springer Verlag, Berlin, 2000, pp. 15-30
[17] Schaller, M., *et al.*, Numerically Optimized Performance of Diabatic Distillation Columns, *Computers and Chemical Engineering, 25* (2001), 11-12, pp. 1537-1548
[18] Sauar, E., Siragusa, G., Andresen, B., Equal Thermodynamic Distance and Equipartition of Forces Principles Applied to Binary Distillation, *Journal of Physical Chemistry A, 105* (2001), 11, pp. 2312-2320
[19] de Koeijer G. M., *et al*., Comparison of Entropy Production Rate Minimization Methods for Binary Diabatic Distillation, *Industrial & Engineering Chemistry Research, 41* (2002), 23, pp. 5826-5834
[20] Schaller, M., *et al*., The Influence of Heat Transfer Irreversibilities on the Optimal Performance of Diabatic Distillation Columns, *Journal of Non-Equilibrium Thermodynamics, 27* (2002), 3, pp. 257-269
[21] de Koeijer, G., Røsjorde, A., Kjelstrup, S., Distribution of Heat Exchange in Optimum Diabatic Distillation Columns, *Energy, 29* (2004), 12-15, pp. 2425-2440
[22] Jimenez, E. S., *et al*., Optimization of a Diabatic Distillation Column with Sequential Heat Exchangers, *Industrial & Engineering Chemistry Research, 43* (2004), 23, pp. 7566-7571
[23] Røsjorde, A., Kjelstrup, S., The Second Law Optimal State of a Diabatic Binary Tray Distillation Column, *Chemical Engineering Science, 60* (2005), 5, pp. 1199-1210
[24] Johannessen, E., Røsjorde, A., Equipartition of Entropy Production as an Approximation to the State of Minimum Entropy Production in Diabatic Distillation, *Energy, 32* (2007), 4, pp. 467-473
[25] Shu, L., Chen, L., Sun, F., Performance Optimization of a Diabatic Distillation-Column by Allocating a Sequential Heat-Exchanger Inventory, *Applied Energy, 84* (2007), 9, pp. 893-903
[26] Ishii, Y., Otto, F. D., A General Algorithm for Multistage Multicomponent Separation Calculations, *Canadian Journal of Chemical Engineering, 51* (1973), 5, pp. 601-606
[27] Browne, D. W., Ishii, Y., Otto, F. D., Solving Multicolumn Equilibrium Stage Operations by Total Linearization, *Canadian Journal of Chemical Engineering, 55* (1977), 3, pp. 307-312
[28] Paunović, R., Janković, M., Škrbić, B., An Extended Ishii-Otto Algorithm for Multistage Multicomponent Separation Calculation with Stage Efficiencies Included, *Computers and Chemical Engineering, 8* (1984), 3-4, pp. 249-251
[29] Ishii, Y., Otto, F. D., An Efficient Simultaneous Correction Procedure for Multicomponent, Multistage Separation Calculations for Non-Ideal Systems, *Computers and Chemical Engineering, 25* (2001), 9-10, pp. 1285-1298
[30] Ishii, Y., Otto, F. D., A Method to Extend the Domain of Convergence for Difficult Multicomponent, Multistage Separation Problems, *Computers and Chemical Engineering, 27* (2003), 6, pp. 855-865

[31] Rosenbrock, H. H., An Automatic Method for Finding the Greatest or Least Value of a Function, *Computer Journal, 3* (1960), 3, pp. 175-184
[32] Hooke, R., Jeeves, T. A., Direct Search Solution of Numerical and Statistical Problems, *Journal of the Association for Computing Machines, 8* (1961), 2, pp. 212-221
[33] Powell, M. J. D., An Efficient Method for Finding the Minimum of a Function of Several Variables without Calculating Derivatives, *Computer Journal, 7* (1964), 2, pp. 155-162
[34] Nelder, J. A., Mead, R., A Simplex Method for Function Minimization, *Computer Journal, 8* (1965), 4, pp. 308-313
[35] Box, J. M., A New Method of Constrained Optimization and a Comparison with other Methods, *Computer Journal, 8* (1965), 1, pp. 42-52
[36] Soave, G., Equilibrium Constants from a Modified Redlich-Kwong Equation of State, *Chemical Engineering Science, 27* (1972), 6, pp. 1197-1203
[37] Lewis, G. N., Randall, M., Thermodynamics, McGraw-Hill, New York, USA, 1961
[38] Schaller, M., Numerically Optimized Diabatic Distillation Columns, Ph. D. thesis, Technische Universität Chemnitz, Chemnitz, Germany, 2007

Paper submitted: December 9, 2009
Paper revised: December 19, 2009
Paper accepted: January 12, 2010